NEUTRON SCATTERING IN THE 'NINETIES
FOREWORD

The International Atomic Energy Agency organized a series of symposia on neutron scattering during the period between 1960 and 1977 because of the importance of the technique in condensed matter research, and to promote the applications of this technique in various branches of applied science and technology. The study of condensed matter physics through neutron scattering techniques has now become a fully developed and mature scientific area and it is felt that much information in this field is more appropriately presented at topical conferences dealing with specific subjects.

However, the technological developments that have taken place since 1977 (the year of the last IAEA symposium on this subject) in new instrumentation and techniques, with emphasis on new types of sources, indicate that neutron scattering is poised for a new intense period of growth in terms of the use of the technique in basic research as well as its application to applied sciences and industry. The late 'seventies and early 'eighties saw intensive activity in a number of areas, in particular utilization of high flux reactors, perfection of techniques and instruments whose potential was recognized earlier, and recently the development of concepts and technology of spallation neutron sources. A number of pulsed neutron sources are already operating or will be operational by the next decade. Thus, the time was considered to be appropriate for a general meeting on the prospects of neutron scattering in the 'nineties.

This International Conference on Neutron Scattering in the 'Nineties, which took place in Jülich, was organized by the International Atomic Energy Agency in co-operation with the Jülich Nuclear Research Centre. These Proceedings emphasize the new developments in techniques, methodology, instrumentation and sources. In addition, the research utilization aspects are covered in a number of review papers which deal not only with condensed matter physics but also include biology and macromolecules, polymer studies, microemulsions and material science, all of which indicate considerable prospects for industrial applications.

The Agency acknowledges the substantial assistance of the Programme Committee and, in particular, Professors Springer and Stiller, its Chairman and Vice-Chairman respectively. The Agency also thanks the Jülich Nuclear Research Centre which was responsible for the local organization, and the Government of the Federal Republic of Germany, at whose invitation the conference was held in Jülich.
EDITORIAL NOTE

The papers and discussions have been edited by the editorial staff of the International Atomic Energy Agency to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants. In addition, the views are not necessarily those of the governments of the nominating Member States or of the nominating organizations.

Where papers have been incorporated into these Proceedings without resetting by the Agency, this has been done with the knowledge of the authors and their government authorities, and their cooperation is gratefully acknowledged. The Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard, in particular to achieve, wherever practicable, consistency of units and symbols and conformity to the standards recommended by competent international bodies.

The use in these Proceedings of particular designations of countries or territories does not imply any judgement by the publisher, the IAEA, as to the legal status of such countries or territories, of their authorities and institutions or of the delimitation of their boundaries.

The mention of specific companies or of their products or brand names does not imply any endorsement or recommendation on the part of the IAEA.

Authors are themselves responsible for obtaining the necessary permission to reproduce copyright material from other sources.
CONTENTS

INTRODUCTORY LECTURES (Session I)

Elementary excitations and phase transformations in solids
(IAEA-CN-46/36) ................................................................. 3
R.A. Cowley

New spallation neutron sources, their performance and applications
(IAEA-CN-46/12) ................................................................. 17
G.H. Lander, J.M. Carpenter

MAGNETIC DEVICES (Session II)

Novel beam bunching methods by perfect crystals and electromagnetic
means (IAEA-CN-46/40) ............................................................ 35
H. Rauch

Travelling wave and neutron magnetic resonance bunching
(IAEA-CN-46/69) ................................................................. 53
H. Rauch, J. Summhammer, H. Weinfurter

Pulsed reactor IBR-2 in the 'nineties (IAEA-CN-46/85) ...................... 63
V.D. Ananiev, Zh.A. Kozlov, V.I. Luschikov, Yu.M. Ostanevich,
E.P. Shabalin, I.M. Frank

MAGNETIC DEVICES AND ULTRA-COLD NEUTRONS (Session III)

Novel applications of polarized neutrons at pulsed sources
(IAEA-CN-46/11) ................................................................. 77
G. Badurek, W. Schmatz

Experience with spin analysis on a time-of-flight multidetector
scattering instrument (IAEA-CN-46/70) ........................................ 85
O. Schärpf

Production and use of ultra-cold neutrons (IAEA-CN-46/34) .......... 99
K.-A. Steinhauser

STEADY-STATE SOURCES (Sessions IV and V)

Magnetic scattering and polarized neutrons (IAEA-CN-46/57) .......... 109
G. Shirane, C.F. Majkrzak
A review of current and proposed reactor upgrades (IAEA-CN-46/28) ....... 119  
R.M. Moon  
Special neutron sources (IAEA-CN-46/16) ........................................ 135  
P. Ageron  
Very-high-flux research reactors based on particle fuels (IAEA-CN-46/38). 157  
J.R. Powell, H. Takahashi  
Upgrading of the Munich reactor with a compact core (IAEA-CN-46/53).... 165  
SINQ — A project for a continuous spallation neutron source  
(IAEA-CN-46/9) ....................................................................................... 171  
F. Atchison, W.E. Fischer, M. Pepin, Y. Takeda, Ch. Tschaaler  

INSTRUMENTS AND METHODS (Session VI)

Nuclear polarization and neutrons (IAEA-CN-46/81) ................................ 181  
H. Glättli  
Neutron scattering studies of nuclear orientation in the μK range  
(IAEA-CN-46/32) ....................................................................................... 185  
M. Steiner  
On-line evaluation of position-sensitive detector (PSD) diffraction data  
(IAEA-CN-46/64) ....................................................................................... 191  
R.F.D. Stansfield, G.J. McIntyre  
A device for combined neutron-photon processes in condensed matter  
(IAEA-CN-46/13) ....................................................................................... 199  
A. Stoeckli, A. Isacson, M. Koch, A. Furrer  
Interface investigation by slow neutron reflection (IAEA-CN-46/20) ....... 205  
B. Farnoux  
MIBEMOL: A six-chopper time-of-flight spectrometer installed on a  
neutron guide of the ORPHEE reactor (IAEA-CN-46/51) ........................ 211  
S. Hautecler, E. Legrand, L. Vansteelant, P.D'Hooghe, G. Rooms,  
A. Seeger, W. Schalt, G. Gobert  

PULSED SOURCES (Sessions VII and IX)

Instrumentation for pulsed sources (IAEA-CN-46/41) .............................. 219  
A.J. Leadbetter  
Instruments for high-flux sources with time structure of neutron flux  
(IAEA-CN-46/80) ....................................................................................... 247  
R. Scherm, B. Alefeld  
The spallation neutron source at the Rutherford Appleton Laboratory  
(IAEA-CN-46/14) ....................................................................................... 261  
D.A. Gray
The SNQ target and moderator system for an optimized neutron economy (IAEA-CN-46/79) ................................................................. 267  
G. Bauer, H. Conrad, D. Filges, H. Spitzer  
Resonance detection methods in spectrometers (IAEA-CN-46/26) ........... 279  
N. Watanabe  
Cold neutron scattering spectrometers at the spallation neutron facility, KENS (IAEA-CN-46/27) ............................................................. 285  
Y. Ishikawa, Y. Endoh, K. Inoue  
Time-of-flight spin echo (IAEA-CN-46/75) (Abstract only) .................... 291  
F. Mezei, D. Richter  
Real-time texture experiments (IAEA-CN-46/31) .................................. 293  
Dorte Juul Jensen

BIOLOGY AND MACROMOLECULES (Session VIII)  
Biomolecular applications of neutron spectroscopy: Current work and future developments (IAEA-CN-46/82) ................................. 303  
H.D. Middendorf  
Morphology and structure of biological systems (IAEA-CN-46/48) ........... 319  
H.B. Stuhrmann  
Neutron scattering from polymers in the 'nineties (IAEA-CN-46/42) ....... 335  
R.S. Stein  
Structure of micellar solutions and microemulsions (IAEA-CN-46/65)  
(Abbrevate only) ........................................................................... 349  
J.P. Cotton

POSTER PRESENTATIONS: PULSED AND STEADY-STATE SOURCES  
(Session X.a)  
Development of the spallation neutron source (SNS) at the Rutherford Appleton Laboratory (IAEA-CN-46/15P) ............................................. 353  
A. Carne  
The spallation neutron source SNQ (IAEA-CN-46/66P) ............................ 361  
B. Alefeld, G. Bauer, H. Halling, H. Lang, J. Schelten, G. Thamm,  
Z. Zettler  
Комплекс источников нейронов на основе протонных пучков Московской мезоной фабрики (IAEA-CN-46/52P) .......................................... 369  
Ю.М. Булкин, М.И. Грачев, Н.В. Колмычков, В.М. Лобашев,  
С.Ф. Сидоркин, Ю.Я. Стависский, В.В. Хмельщик, Н.А. Храстов  
(Assembly of neutron sources based on proton beams of the Moscow meson facility: Yu.M. Bulkin et al.)
Upgrading thermal neutron beams of light-water-moderated research reactors by reflector optimization (IAEA-CN-46/4P) ........................................... 377
E.J. Bleeker, U. Dahlborg, B. Nilsson, J. Skold, W.B. Yelon

A new medium-flux neutron source for pure and applied research
(IAEA-CN-46/62P) ................................................................................... 385
G. Dolling, R.F. Lidstone

The new neutron guide laboratory at the FRJ-2 (DIDO) reactor in Jülich
(IAEA-CN-46/54P) ................................................................................... 393
K. Werner, T. Springer, G. Duppich

POSTER PRESENTATIONS: INSTRUMENTS (PULSED SOURCES)
(Session X.b)

Resolution and intensity of a TOF-TOF spectrometer (IAEA-CN-46/72P). 401
R.E. Lechner

Bunching spectrometer (IAEA-CN-46/74P) (Abstract only) .................... 409
D. Richter, B. Alefeld

Inverted geometry multipurpose spectrometer for pulsed neutron sources
(IAEA-CN-46/46P) ................................................................................... 411

A spectrometer for diffuse scattering at a pulsed neutron source
(IAEA-CN-46/68P) ................................................................................... 413
J. Schelten, G. Bauer

Constant-q scan at a pulsed neutron source (IAEA-CN-46/67P) .......... 417
J. Schelten, B. Alefeld, A. Kollmar

Crystal and chopper spectrometers on pulsed neutron sources
(IAEA-CN-46/8P) ................................................................................... 421
C.J. Carlile, A.D. Taylor, W.G. Williams

Possibilities and restrictions of inelastic magnetic neutron scattering experiments beyond thermal energies (IAEA-CN-46/58P) ................... 435
M. Loewenhaupt

POSTER PRESENTATIONS: INSTRUMENTS (MISCELLANEOUS)
(Session X.c)

ΔT-window spectrometer (IAEA-CN-46/25P) ........................................... 443
B.A. Dasannacharya, P.S. Goyal, P.K. Iyengar, N.S. Satya Murthy,
J.N. Soni, C.L. Thaper

Experience of the Fourier time-of-flight (TOF) neutron techniques for high-resolution neutron diffractometry (IAEA-CN-46/35P) ........ 453
P. Hiismäki, V.A. Trunov, O. Anston, V.A. Kudryashev, H. Kukkonen,
H. Pöyry, A.F. Shechbetov, A. Tiitta, V.A. Ulyanov
A constant-energy crystal spectrometer with a one-dimensional multi-counter (IAEA-CN-46/44P) ................................................................. 461
R. Born, D. Hohlwein

Design of a neutron liquid diffractometer with a position-sensitive detector (IAEA-CN-46/2P) ................................................................. 469
R. G. Delaplane, U. Dahlborg, M. Sandström, K. Sköld

The use of the polychromatic method in elastic neutron diffraction (IAEA-CN-46/19P) ................................................................. 475
A. Delapalme, J. C. Marmeggi, M. Perrin

New velocity drive for backscattering spectrometers (IAEA-CN-46/7P) ........ 481

Area detector for the small-angle neutron scattering facility at Risø (IAEA-CN-46/6P) ................................................................. 489
J. K. Kjems, R. Bauer, B. Breiting, A. Thuesen

Small-angle neutron scattering facility at Risø (IAEA-CN-46/5P) .......... 495
J. K. Kjems, R. Bauer, P. Christensen, T. Freltoft, L. G. Jensen, J. Linderholm

Relaxation of aligned rod-like micelles (IAEA-CN-46/1P) ........ 501
L. Herbst, H. Hoffmann, J. Kalus, H. Thurn, K. Ibel

A pulsed neutron monochromatic beam at the ET-RR-1 reactor (IAEA-CN-46/33P) ................................................................. 507
M. Adib, A. Abdel-Kawy, Y. Eid, R. M. A. Maayouf

Multipurpose double-crystal monochromator for thermal neutrons (IAEA-CN-46/83P) ................................................................. 515
P. Mikula, J. Kulda, B. Chalupa, R. Michalec, M. Vrána, L. Sedláková

POSTER PRESENTATIONS: CRYSTALS AND TEXTURE (Session X.d)

Growth of large copper monochromator crystals (IAEA-CN-46/63P) .......... 527
J.-M. Welter, G. Jungerberg

Texture determination by neutron diffraction (IAEA-CN-46/60P) .......... 533
A. Oleś, J. Kulka, J. Szpunar, R. Wawszczak

Texture studies of cold rolled aluminium and copper sheets by neutron diffraction (IAEA-CN-46/47P) ................................................................. 539
M. M. Beg, N. M. Butt, Q. H. Khan, S. U. Cheema

NEUTRON DIFFRACTION, MATERIALS SCIENCE (Session XI)

Neutron diffraction studies of chemical structure and interaction (IAEA-CN-46/3) ................................................................. 551
M. S. Lehmann
Single-crystal diffractometry at the proposed pulsed neutron source SNQ
(IAEA-CN-46/71) (Abstract only) .......................................................... 565
W. Jauch, H. Dachs

High-resolution powder diffraction (IAEA-CN-46/30) ............................ 567
D.E. Cox

Neutrons for materials science (IAEA-CN-46/45) ................................. 575
C.G. Windsor, A.J. Allen, M.T. Hutchings, C.M. Sayers, R.N. Sinclair,
P. Schofield, C.J. Wright

CONCLUDING REMARKS ...................................................................... 595
M. Lambert, H. Stiller

Chairmen of Sessions and Secretariat of the Symposium ...................... 603
List of Participants .................................................................................. 605
Author Index .......................................................................................... 617
Transliteration Index .............................................................................. 619
Index of papers and posters by number .................................................. 621
INTRODUCTORY LECTURES

(Session I)
Chairman

M. BLUME
United States of America
Invited Paper

ELEMENTARY EXCITATIONS AND PHASE TRANSFORMATIONS IN SOLIDS

R. A. COWLEY
Department of Physics,
University of Edinburgh,
Edinburgh, Scotland,
United Kingdom

Abstract

ELEMENTARY EXCITATIONS AND PHASE TRANSFORMATIONS IN SOLIDS.

Neutron scattering is and will continue to be a uniquely powerful tool for the study of elementary excitations and phase transformations in solids. The paper examines a few recent experiments on molecular crystals, superionic materials, paramagnetic scattering and phase transitions to see what experimental features made these experiments possible, and hence to make suggestions about future needs. It is concluded that new instruments will extend the scope of neutron scattering studies to new excitations, that there is a need for higher resolution, particularly for phase transition studies, and that it will be important to use intensity information, discrimination against unwanted inelastic processes and polarization analysis to measure the excitations in new materials reliably.

1. INTRODUCTION

Neutron scattering is a uniquely powerful tool for the study of the elementary excitations and phase transitions of condensed matter. This is because thermal neutrons are the only probe for which the energy and wavevector are both well matched to the energies and wavevectors of the excitations which determine the thermal properties of condensed matter. It is this unique dual property of neutrons which has made them such a successful probe.

At this conference we are considering neutron scattering in the '90s, by when it will be over 40 years since the first thermal neutron scattering experiments to study condensed matter were performed at Oak Ridge. Are we certain that there will still be worthwhile experiments to perform in the '90s? Especially as by then there will be synchrotron sources providing a much brighter X-ray source than is conceivable for any neutron source, as well as the continuing progress with high resolution optical spectroscopy. Secondly if there are worthwhile experiments what type of equipment and facilities will
be needed? In this paper I shall examine these questions by looking back at several experiments on crystalline solids which have been performed over the past few years which have attracted my interest and which broadly illustrate, I believe, the types of experiments which we shall want to do in the '90s.

2. MEASUREMENT OF INTERATOMIC INTERACTIONS

The pioneering work of Brockhouse on the determination of phonon and magnon dispersion relations is now described in
elementary solid state physics textbooks and underpins a great deal of our detailed knowledge of interatomic forces and of exchange interactions. This work led to the development of the shell model and its various extensions to describe the forces in ionic materials and semiconductors, and provided essential support for the pseudo-potential theory of metals.

It is now possible to determine the phonon dispersion curves in more complicated crystals with up to about 10 atoms in the unit cell. A recent example is the determination [1] of the intermolecular modes of motion in \( C_2(CN)_4 \), shown in Fig. 1, which was chosen for study because it is one of the simplest materials in which there is a strong coupling between intermolecular and intramolecular modes. It was essential in this work to have available a fairly reliable theoretical model so that the normal modes could be identified by the intensities obtained with different scattering vectors, as well as a large single crystal and ample spectrometer time.

Currently the determination of phonon and magnon dispersion relations is unfashionable. We should not forget, however, that a detailed understanding of the phonon dispersion waves is essential before it is possible to understand the electrical and thermal conductivity and so measurements of the dispersion relations will provide basic information for any material which is likely to be of technological importance. Neutron scattering will remain the best way of determining the phonon dispersion relations, but the results may well be capable of interpretation only if intensity data is obtained for wave-vector transfers in different Brillouin zones, so that the normal mode eigenvectors can be identified with the help of a model or, with more difficulty, determined directly from the intensities.

3. NOVEL TYPES OF EXCITATIONS

In many materials the motions of the atoms are more complex than can be described by the theory of the small vibrations of regular crystals. One example is that of incommensurately modulated phases in which there is a pattern of modulation with wavevector \( \mathbf{q}_I \) which is incommensurate with the underlying crystal lattice. The energy of the crystal is then unchanged if the phase of the incommensurate modulation is altered. There are therefore modes [2] which describe changes in this phase, whose frequency is linearly proportional to the difference between their wavevector \( \mathbf{q} \) and \( \mathbf{q}_I \) namely \( \omega(\mathbf{q}) = C|\mathbf{q} - \mathbf{q}_I|\). These new acoustic-like modes are called phasons and, in a more general language, are the Goldstone modes resulting from the broken phase symmetry of the incommensurate phase.
Neutron scattering measurements of phasons, proved, however, to be difficult to obtain. There were two reasons for this: the difficulty in distinguishing between the phase modes and the normal acoustic modes, especially as they may interact with one another for $\mathbf{q} = \mathbf{q}_I$, and the fact that they are overdamped for small $|\mathbf{q} - \mathbf{q}_I|$ unless the temperature is very low [3]. In Fig. 2 are shown measurements [4] of the phason branch in ThBr$_4$. In this case it was known where in wavevector and energy to expect the phasons and the measurements were made possible partly because the acoustic modes had a very different slope from the phase mode, partly because the material has an incommensurate phase at very low temperatures so that the mode is not overdamped in the $\mathbf{q}$ region of the measurements, and finally because of the good wavevector and energy resolution of the spectrometer used.
Another type of novel excitation are the tunnelling modes which have now been observed [5] - Fig. 3, at low temperatures in the vibration spectra of, for example, CH$_3$ groups in CH$_3$NO$_2$. In this case the quantum mechanical tunnelling between different equivalent orientations of the CH$_3$ groups gives rise to a very small splitting of the lowest energy levels. The energy of this splitting has been observed directly in many materials by using
FIG. 4. A contour plot of the quasi-elastic scattering in CaF$_2$ at 1473K as compared with the theoretical model /6/.

the very high energy resolution of the backscattering spectrometers at the Institute Laue Langevin (ILL). These energies ~μeV are much smaller than can be observed with conventional spectrometers and the experiments would not have been possible without the development of these spectrometers.
Superionic materials are ones in which the ions become, at high temperatures, extremely mobile. In order to understand this behaviour it is necessary to identify the interstitials by which the ions can hop from one site to the next. These sites have been successfully identified in the fluorite class of superionic materials [6] by using neutron scattering techniques. The experiments consisted of measuring the intensity of the quasi-elastic scattering through a considerable volume of reciprocal space as illustrated in Fig. 4 for CaF$_2$, and then comparing this intensity with various different models for the interstitial sites. The good agreement between the experimental results and the calculations, Fig. 4, enabled the authors to
identify the interstitial sites and deformations around the Fe ions. In this experiment the diffuse elastic scattering was much less intense than the phonon scattering at these temperatures. Consequently it was necessary to use an inelastic spectrometer to eliminate the phonon scattering from the measurements of the quasi-elastic scattering. The requirement therefore is for a spectrometer which efficiently measures the quasi-elastic scattering over a large region of reciprocal space while discriminating against the inelastic scattering. It is likely that such measurements will play an essential role in the understanding of other partially ordered systems such as plastic crystals.
Neutrons are also ideal probes of the magnetic excitations although until recently most of the experiments had been performed using unpolarized neutrons because the loss of neutrons caused by the inefficient polarizers effectively prohibited the use of polarisation analysis for experiments on magnetic excitations except for a few favourable examples. Polarisation analysis does, however, give additional important information in enabling scattering from nuclear processes to be distinguished from magnetic processes, and in identifying the different contributions to the magnetic cross-section. The first application is particularly useful when the magnetic scattering is broad and difficult to distinguish from phonon scattering. In the ferromagnet Pd$_3$Fe unpolarized neutron scattering experiments suggested that near the [100] zone boundary the spin waves were broadened possibly because they interacted with the Stoner continuum of single particle states. The measurements were, however, tentative because of the presence of phonons in the same energy region. A polarized neutron scattering measurement [7] with an incident polarized beam, a horizontal magnet and an unpolarised analyser showed clearly, Fig. 5, that the magnetic scattering near the [100] zone boundary is broad in energy, which presumably results from the interaction with the Stoner modes.

More recent examples of the way in which polarization analysis enables the magnetic scattering to be obtained are the extensive studies [8,9] of paramagnetic scattering from metallic magnets. At the comparatively high temperatures involved it is essential to be able to separate the magnetic and nuclear cross-sections as illustrated by the measurements shown in Fig. 6. In this case of paramagnetic scattering a polarized incident beam and polarization analysis of the scattered beam were used, and the recent activity in the field results directly from the development of polarization analysis techniques at ILL and Brookhaven National Laboratory. In both of these measurements polarization analysis proved to be essential in enabling a broad distribution of magnetic cross-section to be separated from the phonon background.

A third example of the use of polarization analysis is in the study of soliton excitations in quasi-one-dimensional systems. In the case of CsNiF$_3$ [10], it has been difficult to distinguish between the two-magnon contribution and the soliton contribution to the scattering cross-section. Recent measurements using full polarization analysis have enabled the different magnetic cross-sections to be observed which have enabled the magnitude of the different components to be determined. These measurements required polarization analysis of the scattering occurring with a known well-defined energy and wave-vector.
Much of the recent progress in the understanding of phase transitions has been in understanding the fluctuations associated with continuous phase transitions. Neutron scattering is unique in that it enables the wavevector and frequency dependence of these fluctuations to be measured directly, and in no small part has the progress in the field been the result of neutron scattering experiments.

Most of the experiments on the fluctuations have been analysed by assuming the Ornstein-Zernike form for the scattering cross-section above the transition temperature, $T_c$, namely

$$I(Q) = \frac{A}{\kappa^2 + |Q - \vec{\kappa}|^2}$$

where $\vec{Q}$ is the wavevector describing the ordering and the inverse correlation length, $\kappa$, has a temperature dependence of the form

$$\kappa = \kappa_0\left(\frac{T - T_c}{T_c}\right)^\nu$$

where $\nu$ is the critical exponent. The susceptibility exponent, $\chi$, is also given by

$$\frac{A}{\kappa^2} = \chi_0\left(\frac{T - T_c}{T_c}\right)^{-\gamma}$$

Consequently neutron scattering measurements directly yield the amplitudes $\kappa_0$, $\chi_0$ and exponents $\nu$, $\gamma$ and $\beta$ for the order parameters.

In practice the situation is more complex than at first appears. The theory and exponents are only defined as $T \to T_c$, when $\kappa \to 0$ and the cross-section becomes very peaked in wavevector, while the resolution function is of fixed size. Consequently the most crucial data, with temperatures closest to $T_c$, are often very dependent upon the resolution corrections. It is clearly desirable to have better resolution. Indeed this is even more important as the Ornstein-Zernike form may not be the correct form of the cross-section as $T \to T_c$. This problem has only been studied in detail for the $d = 2$ Ising model for which the Ornstein-Zernike form is found to be a good approximation for $T > T_c$, but is very wrong for $T < T_c$ [11]. The analysis of experiments [12] on Rb$_2$CoF$_4$ showed that the Ornstein-Zernike form gave a reasonable description of the data, when convoluted with the resolution function, but that the
amplitudes obtained were very wrong. Analysis of the data for T < T_c, using a better form for the correlation function, gave a slightly better fit to the data, but more significantly provided satisfactory values for the amplitude ratios. In other systems it is not known whether the Ornstein-Zernike form is a satisfactory approximation especially for T < T_c, so that there is a real possibility that the conventional analysis of critical scattering data provides incorrect amplitude ratios. Clearly experiments are needed in which the resolution is sufficiently good that they can directly test whether the Ornstein-Zernike form is adequate.

There is a similar problem with the analysis of the 'central peak' at structural phase transitions. The analysis usually adopts a particular form of the cross-section and fits this to the experimental data. While this functional form is definitely reasonable, as is the Ornstein-Zernike form, there are also good reasons to suspect that it is a very oversimplified form and higher-resolution experiments are needed to test the accuracy of the assumed functional form. It is always more difficult to test the functional form than to fit a few parameters.

The need for higher resolution is well illustrated by experiments on the effect of applying a random field to an ordered magnetic system. Experiments are performed by applying a uniform field to a random antiferromagnet. When systems are cooled in a field then the systems no longer develop long-range order, Fig. 7, [13, 14], and the lineshape is of an unusual Lorentzian squared form. This result is currently in contradiction with the theory for d = 3 systems that predicts using equilibrium statistical mechanics, that the system should exhibit long range order. The behaviour at low temperatures is however dependent upon the past history of the sample below a well defined transition temperature, which depends on the strength of the applied field. The frequency or time dependence of the scattering in this low-temperature phase, and close to the phase transition is currently of interest in trying to reconcile the experimental results with theory as are higher resolution measurements to enable the experiments to be performed at smaller applied fields. The problem is an example of the type of physics which occurs in systems with competing interactions - namely lack of conventional long range order, metastability and long relaxation times. Another example of the contribution of neutron scattering to these systems is the study of the relaxation times in a Cu/Mn spin glass [15] Fig. 8 where the spin-echo technique has enabled the relaxation times to be studied for times intermediate between a microscopic time scale and macroscopic time scales, showing that the relaxation developed continuously over ten decades in time.
FIG. 7. The scattering from Fe$_x$Zn$_{1-x}$F$_2$ as a function of magnetic field at 10K. The increasing field destroys the long-range order and the solid lines are fits to a Lorentzian squared form /14/.

FIG. 8. The time-dependent spin correlation function for Cu$_5$Mn at various temperatures /15/.

5. SUMMARY AND CONCLUSIONS

In this paper I have illustrated the use of neutron scattering techniques to study the excitations and phase transitions in solids, by describing some recent experiments, which illustrate, I believe, the wide range of experiments which will
continue to be done with neutrons. It is difficult to see how any of the experiments can possibly be done in the '90s by using other techniques, such as synchrotron radiation or laser spectroscopy. We shall continue to need neutron scattering techniques.

Some of the experiments have proved to be possible because of the development of new instruments. This is particularly the case of the tunnelling experiments, Fig. 3, the spin echo experiments, Fig. 8, but also of the polarization analysis experiments, Figs 5 and 6. New instruments to enable neutron scattering measurements to be extended to new regions of frequency and wavevector will yield important results.

Many of the experiments described above need higher resolution in wavevector and frequency, especially those involving phasons, Fig. 2, and critical phenomena, Fig. 7. There will need to be an increasing effort to improve the resolution especially in wavevector during the '90s, if further progress is to be made in, say, phase transitions.

I suspect that during the '90s experiments which require a wider survey of reciprocal space or frequency will also play an important role such as those shown in Figs. 1, 4 and 6. These are likely to be particularly important for characterizing more complex materials, or for studying disordered or only partially ordered systems. In these cases, however, the experiments will be likely to need the full control and flexibility of the neutron scattering technique, to give useful results. It will be essential to use intensity information for equivalent points in reciprocal space to understand phonon dispersion curves. It was essential to be able to discriminate against inelastic phonon scattering in the experiments on both the superionic materials and paramagnetic materials.

I hope to see that neutron scattering will play an increasing role in understanding more about the electronic properties of materials with heavy fermions, with valence fluctuations, with superconducting properties and in understanding the properties of the electrons in actinides. All of these experiments will require measuring relatively weak and broad scattering under conditions in which it is possible to separate out clearly the electronic scattering, by polarisation analysis or other techniques. These experiments and those outlined above which require improved resolution will prove challenging to develop the necessary equipment and facilities for the '90s. This improvement in techniques is however necessary if neutron scattering is to be as useful in the study of excitations in solids during the '90s as it has been through the '60s, '70s and '80s.
REFERENCES

Invited Paper

NEW SPALLATION NEUTRON SOURCES, THEIR PERFORMANCE AND APPLICATIONS

G. H. LANDER, J. M. CARPENTER
Argonne National Laboratory,
Argonne, Illinois,
United States of America

Abstract

NEW SPALLATION NEUTRON SOURCES, THEIR PERFORMANCE AND APPLICATIONS.

Pulsed spallation sources now operating in the world are at the KEK Laboratory in Japan (the KENS source), at Los Alamos National Laboratory (WNR) and at Argonne National Laboratory (IPNS), both the latter being in the USA. The Intense Pulsed Neutron Source (IPNS) is currently the world's most intense source with a peak neutron flux of $4 \times 10^{14}$ n/cm$^2$/s at a repetition rate of 30 Hz, and globally producing $\sim 1.5 \times 10^{15}$ n/s. Present pulsed sources are still relatively weak compared to their potential. In 1985 the Rutherford Spallation Neutron Source will come on line, and eventually be about 30 times more intense than the present IPNS. Later, in 1986 the WNR/PSR option at Los Alamos will make that facility of comparable intensity, while a subcritical fission 'booster' at IPNS will keep IPNS competitive. These new sources will expand the applications of pulsed neutrons but are still based on accelerators built for other scientific purposes, usually nuclear or high-energy physics. Accelerator physicists are now designing machines expressly for spallation neutron research, and the proton currents attainable are in the milliamps range. (IPNS now runs at 0.5 GeV and 14 µA). Such design teams are at the KFA Laboratory Jülich, Argonne National Laboratory and KEK. Characteristics, particularly the different time structure of the pulses, of these new sources are discussed. Such machines will be expensive and require national, if not international, collaboration across a wide spectrum of scientific disciplines. The new opportunities for neutron research will, of course, be dramatic with these new sources.

1. Introduction

The first source using a proton accelerator and specifically equipped for neutron scattering from condensed matter systems was a prototype built at Argonne National Laboratory in 1974. Called ZING-P, it had an energy of 200 MeV and a time-averaged current of 0.1 µA. The ASPUN or SNQ projects now advanced by Argonne and KFA, Jülich, respectively, have energies of ~ 1600 MeV and projected currents of 4000 µA. These represent an increase in neutron intensity from the early ZING-P source at ANL by a factor of nearly $10^5$. Figure 1 shows a conceptual spallation source.
Fig. 1. Schematic of spallation source including linac and circular ring to produce short pulse proton bursts. Two targets are shown surrounded by scattering instruments.

Fig. 2. Plot of available proton intensity from spallation sources from 1974 to 2002.
TABLE 1. PROTON SPALLATION SOURCES

<table>
<thead>
<tr>
<th>Facility</th>
<th>Accelerator</th>
<th>Particle Energy MeV</th>
<th>Time-Average Current μA</th>
<th>Average Pulsing Frequency Hz</th>
<th>Source Pulse Width μs</th>
<th>Target Material</th>
<th>Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZING-P'</td>
<td>Synchrotron</td>
<td>500</td>
<td>3</td>
<td>30</td>
<td>0.1</td>
<td>$^{239}$U</td>
<td>Operated 1977-80</td>
</tr>
<tr>
<td>Argonne, USA</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WNR</td>
<td>Linac</td>
<td>800</td>
<td>3.5</td>
<td>120</td>
<td>4.0</td>
<td>W</td>
<td>Started 1977</td>
</tr>
<tr>
<td>Los Alamos, USA</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KENS-I</td>
<td>Synchrotron</td>
<td>500</td>
<td>2</td>
<td>15</td>
<td>0.07</td>
<td>W</td>
<td>Started 1980</td>
</tr>
<tr>
<td>KEK, Japan</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IPNS-I</td>
<td>Synchrotron</td>
<td>500</td>
<td>12</td>
<td>30</td>
<td>0.1</td>
<td>$^{239}$U</td>
<td>Started 1981</td>
</tr>
<tr>
<td>Argonne, USA</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SNS</td>
<td>Synchrotron</td>
<td>800</td>
<td>200</td>
<td>50</td>
<td>0.2</td>
<td>$^{239}$U</td>
<td>To start 1985</td>
</tr>
<tr>
<td>Rutherford, U.K.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KENS-I'</td>
<td>Synchrotron</td>
<td>500</td>
<td>10</td>
<td>15</td>
<td>0.5</td>
<td>$^{239}$U</td>
<td>To start 1985</td>
</tr>
<tr>
<td>KEK, Japan</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WNR-PSR</td>
<td>Linac +</td>
<td>800</td>
<td>100</td>
<td>12</td>
<td>0.27</td>
<td>$^{239}$U</td>
<td>To start 1986</td>
</tr>
<tr>
<td>Los Alamos, USA</td>
<td>Storage Ring</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SNQ</td>
<td>Linac and</td>
<td>1100</td>
<td>4000</td>
<td>100</td>
<td>$^{239}$U</td>
<td>Proposal Under Devel.</td>
<td></td>
</tr>
<tr>
<td>ASPUN</td>
<td>FFAG Synchrotron</td>
<td>1600</td>
<td>4000</td>
<td>60</td>
<td>0.4</td>
<td>Proposal Under Devel.</td>
<td></td>
</tr>
<tr>
<td>Argonne, USA</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

This table gives the specification of present (and proposed) proton spallation sources in the world. The first such proton source operated at Argonne in 1974-75 with 0.1 μA current and an energy of 200 MeV. We have not covered electron driven sources here, the largest of which HELIOS at Harwell, UK, also performs a good deal of condensed matter research.

We show in Figure 2 the progress of effective neutron flux from proton spallation sources (normalized to the Intense Pulsed Neutron Source, IPNS, at Argonne as of January 1985) over the period from ZING-P to the (hopefully) operating SNQ and ASPUN type sources, projected for the mid to late 1990's. Figure 2 shows a steady logarithmic increase of a factor of 10 about every 5 years. When one considers the almost nil progress in reactor fluxes over this same period, remembering that the world's highest flux reactor HFIR at Oak Ridge was commissioned in 1966, Figure 2 is an impressive testimony to the development of neutron fluxes from spallation sources. Details of proton spallation sources are given in Table I. To
obtain Figure 2 we have simply multiplied the energy and current of the proton sources. This gives one measure of usefulness, but is not by itself enough to assure a steady increase in neutron intensity for scattering experiments. Once the protons are produced they must be "converted" into neutrons; this is done through their interaction in targets of heavy nuclei (lead, tungsten, depleted or enriched uranium). The resulting neutron energy spectrum is far too energetic to be of use in neutron scattering experiments on condensed matter and moderators are required to slow down the neutrons.

2. Accelerators

To produce submicrosecond pulse proton beams the common method is with a synchrotron machine. However, the most advanced rapid cycling synchrotron designs presently completed have a goal of accelerating ~ $6 \times 10^{13}$ protons per pulse (ppp). IPNS runs at $3 \times 10^{12}$ ppp and the Rutherford at full power of 200 $\mu$A and 800 MeV will have $2.5 \times 10^{13}$ ppp. Scientists at KEK, Japan, have discussed a synchrotron (GEMINI) that could possibly deliver 500 $\mu$A at 800 MeV. This machine would have a space-charge limit of $7.2 \times 10^{13}$ protons and clearly be at the forefront of accelerator technology. To obtain higher currents one must either use linacs, in which case very large currents are possible, or another type of accelerator technology such as induction linacs or FFAGs. The most intense linac operating today is the LAMPF (Los Alamos Meson Proton Factory) which recently reached its design current of 1000 $\mu$A. The SNQ project is aiming for 4000 $\mu$A so is stretching the limits of linac technology. Such linacs have inevitably a relatively long pulse of protons. The macro-pulse length at LAMPF is now 750 $\mu$s, and would be 250 $\mu$s long at the SNQ facility. These long proton pulse lengths may be compressed in a storage ring that "compacts" the proton beam without accelerating it. At Los Alamos the proton storage ring, PSR, will accept one 750 $\mu$s macro-pulse containing $5 \times 10^{13}$ protons from LAMPF (frequency 120 Hz) and compress it into a pulse of 0.27 $\mu$s length. This will occur at a 12 Hz rate. A compressor ring, IKOR, has also been considered by the Jülich design team for an advanced stage of the SNQ project.

At Argonne, on the other hand, a different accelerator concept, the so-called Fixed-Field Alternating Gradient (FFAG) design has been introduced by R. L. Kustom and his collaborators[1]. In this design, which leads to the advanced machine called ASPUN, the proton orbits increase in radius as the energy increases. The magnetic field also increases with radius to provide increased bending strength. However, these fields are fixed in time, unlike a synchrotron. The DC
magnetic fields allow more efficient injection and capture of beam and more effective use of the rf acceleration system. The design goals for the current and beam power in the FFAG designs exceed the conventional rapid cycling synchrotrons by at least an order of magnitude. The FFAG synchrotron also allows the opportunity to internally stack beams, thus having an extracted beam repetition rate different from the injected beam repetition rate, leading to higher individual extracted pulse amplitudes. The prospective gain over linacs + compressor ring assemblies is so important that further work should continue on the FFAG.

3. Targets

Table I shows that the chosen target for medium intensity sources, as those operating in this decade, is depleted uranium. High-Z, high density targets are preferable because the neutron producing reactions are then most favorable, while the short range of the protons minimizes the size of the source. At moderate intensities the neutron intensity can be increased by using fissile material in a "booster" target. The gain factor is approximately $G \approx \frac{1}{(1-k)}$, where $k$ is the prompt-neutron multiplication factor. A booster with a gain factor of $x10$ was operated at the original Harwell Linac and is operating on the new accelerator, HELIOS for nuclear physics research. Plans are now fairly far advanced for a booster target for IPNS that will represent the first such target at a proton source when it is installed in 1986. The IPNS booster target should give a factor of ~3 in neutron beam intensity. Boosters may also be installed later at SNS and the WNR facility, although the power levels at both those facilities may be too high to allow correspondingly high gains. Because fission requires the dissipation of ~ 190 MeV per useful neutron produced, as opposed to ~ 50 MeV for the true spallation process, and boosters inescapably broaden the primary pulse and increase the number of delayed neutrons between pulses, their application is limited.

At higher power densities the heat deposition in stationary uranium is too great for any prospective cooling system. The SNQ project has devised a rotating water-cooled target. This target consists of 5916 cylindrical capsules of U (earlier versions specified W) each 2.4 cm diameter and 10 cm height, mounted on a wheel of 2.5 m diameter rotating at 0.5 Hz. The very considerable cooling required means that at these power densities the efficiency of neutron production decreases somewhat as compared to the more modest sources.
4. Moderators

Although accelerators are expensive and complex, and targets are technically challenging, the moderators and reflectors of pulsed sources might be said to represent the most arcane elements. Moderators could justifiably be called the "insertion devices" of pulsed neutron sources, for, similarly to the case of wigglers and undulators in synchrotron-light sources, they shift the radiation spectrum and tailor the time distribution differently, to suit different applications.

Figure 3 illustrates the general features of the shape of the moderated pulse. The intensity (normalized to 1.0 at the peak of the pulse) is plotted vs. energy and a length which represents the normalized time for each energy (neutron speed x time after the source pulse). For high energies, in
the "slowing-down" range, the normalized pulse shape is invariant with energy. For energies near kT, the exponential decay of thermalized neutrons produces a pronounced bulge, which remains roughly constant in absolute time, but becomes progressively narrower in normalized time, for energies below kT. We have two, orthogonally-placed handles on the time distribution through which we can optimize the response. Heterogeneous poisoning (subdividing the moderator with sheet(s) of absorber material) narrows the bulge by reducing the thermal neutron decay time. Cooling reduces kT, and moves the bulge to lower energy, thus extending the range of invariant pulse shapes to lower energy.

The time-averaged spectra for all moderators have two components: a thermal-neutron part dominant at low energies and a "slowing-down" (epithermal) part dominant at energies above about 5 kT. There are many tricks available to enhance moderator performance. Recently,[2] moderators with grooved surfaces have been developed, to provide higher fluxes, at some expense of pulse width. The choice of moderator is related to questions of heat transport and radiation damage; thus for example solid methane, wonderful from the point of view of providing intense, short pulses at low energy, is difficult to incorporate in high-intensity sources. Moderator design and optimization is a field which is still rapidly developing.

**Cold neutrons.** Here there is no difficulty for experiments such as small-angle scattering in which the time-averaged flux is the important factor. Using the equation,

\[ \lambda(\Delta) = 0.3955 \frac{t(\mu s)}{L(cm)} \]

and assuming L = 10 m, we find that a 5Å takes 12 ms to travel the distance. Thus if the timing uncertainty is ~ 100 μs this corresponds to only ~ 1% in Δλ/λ, as compared with the much greater Δλ/λ used at reactor SANS instruments. Source pulse widths do not play a central role in cold neutron research, so that the SNQ source, for example, has proposed using a cold D₂ moderator and a series of guide tubes for slow neutron experiments. Absorption of neutrons in D₂ is, of course, small, and losses occur due to the finite size of the moderator, absorption in structural material, and neutrons escaping through the beam holes, so that a large moderator is required. The difficulty with cold neutrons is that frame overlap, in which neutrons from pulse n + 1 catch up those from pulse n with the long flight paths necessary for good resolution, does not allow use of the full neutron bandwidth at repetition rates greater than ~ 40 Hz. To avoid these
limitations we have recently pointed out (see Fig. 1) that a pulsed source with two targets, one maximized for cold neutrons—say, a cold D\textsubscript{2} source and ambient D\textsubscript{2}O reflector, and operating at a repetition rate of 10 Hz, and the other using a hydrogenous moderator with a repetition rate of 40 Hz, would introduce a new flexibility into neutron research.

**Thermal and Epithermal Neutrons.** Moderators for these will certainly be hydrogenous, and it is here that the full technology of moderators, reflectors and poisons needs to be further explored. A general overview of these has been given recently\textsuperscript{[3]}.  

5. **Pulse Lengths**

Two general classes of accelerators and corresponding moderators are contemplated for pulsed sources. The "short-pulse" variety is based on accelerators which produce sub-microsecond pulses and incorporate rapidly-responding, small, dense, strongly-decoupled hydrogenous moderators which provide tailored, short $([\Delta t \sim (10-20\ \mu s/\text{Å}) \times \lambda(\text{Å})])$ pulses of moderated neutrons. The "quasi-steady" type is based on accelerators which produce pulses of duration of several hundred microseconds, and incorporate large, nondecoupled, D\textsubscript{2}O or hydrogenous moderators which provide highly-efficient moderation and long-time storage of thermal neutrons ($\sim 500\ \mu s$ independent of energy).

The moderators in quasi-steady sources are slower in response than those in short-pulse sources, roughly matching the source pulse length and capitalizing on the fact that these are more efficient in converting primary source neutrons to slow neutron beams, than rapidly-responding moderators. Time-average beam intensities of thermal neutrons per primary source neutron are thus greater in the case of the quasi-steady sources than in the short-pulse sources and their spectra are more nearly thermally equilibrated (appearing like reactor spectra) than those from short-pulse moderators. One may, of course, provide a slow moderator in a short-pulse source, with this same motive, for applications in which pulse length is not important and intensity is the governing consideration. The short-pulse sources provide much higher ratios of epithermal to thermal neutrons than the quasi-steady sources.

These are very significant differences in source pulse structure; either can be used highly effectively, but the mix of applications (which we can now only speculate about) will
certainly be different for the two. Very general arguments illustrating why short pulsed sources are desirable in spite of their typically lower efficiency (in the sense of converting fast to slow neutrons) have been presented some time ago by Michaudon\cite{4} and recently more precisely by Windsor\cite{5}.

In the quasi-steady sources, as in steady sources, monochromating devices such as crystals or choppers always define the incident-neutron energy. Although no direct use is made of the pulse, significant advantages accrue due to pulsing. In a chopper spectrometer, for example, (two choppers are needed in the quasi-steady source) the choppers can be phased to open so as to admit neutrons when the source flux is at its maximum, thus capitalizing on the duty cycle improvement in peak flux (but not capitalizing on the inverse square of the pulse length.) In crystal-monochromated beams, the counting can be arranged to take place while the source is "off" and the background is lower than average. In other applications, the fact that the source pulse is of considerable duration makes it possible to perform measurements at several distinct incident wavelengths all in the same set-up.

In the short-pulse source, at least one aspect of wavelength sorting is always done based on the definition of neutron flight times by the source pulse, whose length determines the resolution. Thus in this case there is a more-or-less strict requirement on having short pulses, stringently dependent upon the demands of the particular type of spectroscopy. The situation at higher neutron energies is easily appreciated. There, above say $E_0 = 300$ meV, conventional monochromating crystals lose efficiency for most purposes, guide tubes are ineffective, and chopper design is constrained; use of the already-prepared source pulse provides one step of monochromation which is 100% efficient. The short-pulse sources inherently provide more epithermal neutrons in relation to thermals, than the quasi-steady sources; this, combined with the progressive shortening of pulses with decreasing wavelength, makes the short-pulsed sources more effective in the higher-energy range than the quasi-steady ones.

6. Scientific Applications

We cannot hope to cover even a fraction of the scientific applications of pulsed sources in the short survey presented here; nevertheless it is worthwhile speculating on a few experimental methods that may well become routine and illustrating them with scientific examples.
High-Pressure Powder Diffraction. The special advantages of time-of-flight energy-dispersive powder diffraction are that all the information can be extracted through a single window in the pressure cell - thus allowing higher pressures to be reached.

High pressure neutron diffraction implies a small accessible sample volume because of the limitations of the materials used to enclose samples under pressure. The range of devices currently in use extends from hollow-cylinder, gas or liquid cells which enclose large volumes of several cm$^3$ at pressures up to about 10 kbar, to the recently popularized diamond anvil cells which achieve pressures in the Mbar range for micron-size samples. Each order of magnitude increase in pressure is accompanied by a quantum reduction in sample size. Neutron diffraction studies are currently flux-limited and can be done for 1 cm$^3$ samples in gas or liquid cells to 10 kbar or 0.1 cm$^3$ samples in supported piston-in-cylinder cells which can reach about 40 kbar. The next order of magnitude increase in neutron flux will allow the use of multiple anvil (cubic) presses which can achieve 100 kbar for effective sample volumes (accessible to neutrons) of about 0.01 cm$^3$. With still higher fluxes, opposed anvil designs similar to diamond anvil cells may extend this range to a few hundred kbar.

Synchrotron studies with diamond anvils can extend phase diagram studies up to the Mbar range. However, the absorption in the cell, preferred orientation with micron-size samples, and other systematic problems prevent accurate intensity measurements and the studies are confined to determination of diffraction line positions. Neutrons, with low absorption and a well-characterized spectral function, can determine these intensities with samples of the order of 10 mm$^3$ and so determine atomic positions. This, of course, is in addition of the usual advantage enjoyed by neutrons of being able to "see" light atoms in the presence of heavy ones. This advantage is of particular importance for geophysical materials.

Small-Q Magnetic Inelastic Scattering. A particularly challenging problem in neutron scattering from magnetic materials is to obtain data at small Q and relatively large energy transfers $\hbar \omega$. Pulsed sources are particularly good at probing the area of S(Q,E) space given by the coordinates 2 \textless Q \textless 5 Å$^{-1}$, 50 \textless E \textless 500 meV, because of the abundance of epithermal neutrons. Results from IPNS have already demonstrated[6,7] that this region of the (Q,E) diagram is rich in science. An area of potentially equal interest is that with 0.001 \textless Q \textless 1 Å$^{-1}$ and 5 \textless E \textless 20 meV. The low end of this Q and energy range can be reached with instruments at cold
Wide-Angle Detectors

Small-Angle (Position-Sensitive) Detector

Source (Moderator)

Chopper

7 m Sample

Spin Flipper

20 m Polarisng Filter

20 m Spin Analyzer

Fig. 4. Schematic of chopper spectrometer discussed in text. Incident flight path $L_0 = 20$ m, chopper to sample $L_2 = 1$ m, sample to detector $L_3 = 20$ m.

Sources (either at reactors or pulsed sources) with incoming or analyzed beams of energy $\sim 4$ meV (reflection from graphite) and usually have excellent energy resolution. However, for larger energy transfers in downscattering (i.e. the neutrons losing energy to the sample) we need the analog of a small-angle machine with inelastic scattering. Such a machine has been considered both by Crawford et al.\cite{10} and at the Shelter Island Workshop\cite{9} and is sketched briefly in Fig. 4. For example, with $L_1 = 20$ m, $L_2 = 1$ m, $L_3 = 20$ m, $E_1 = 1000$ meV, $\Delta E = 40$ meV, $Q = 0.5$ Å$^{-1}$, the energy resolution is $0.005 \frac{E_1}{E} = 5$ meV and the Q resolution is 10%. The scattering angle $\phi = 0.61^\circ$, and the beam is displaced $\sim 22$ cm. This design requires choppers rotating at 600 Hz (the present magnetic choppers bearing can achieve $\sim 1000$ Hz) and efficient ways to detect eV neutrons. Presumably scintillation detectors would be best.

What kind of scientific problems could one examine with such a capability? There are two areas of particular interest. The first is to examine the dynamic response function of the conduction electron states in metals, especially in intermediate valence and actinide systems. Such states are extended in real space so have a form factor that drops to zero by 1 Å$^{-1}$. The assumption is usually made that the response of the conduction electron states follows that of the f electrons in intermediate valence systems, but this is by no means obvious. A second type of scientific problem occurs in amorphous or glassy systems. These, of course, have no repeat distance in real space, nevertheless long-wavelength excitation modes (both atomic and magnetic in origin) exist at small wavevector. Such excitations occur at $Q \lesssim 1.5$ Å$^{-1}$, which is characteristicly the location of the first peak in the static structure factor, and have been predicted to extend in energy up to $\sim 50$ meV.
FIG. 5. The scattering function from $^3$He calculated using the chopper spectrometer in Fig. 4. Incident neutron energy is 250 meV with a resolution at a scattering angle of $\phi = 91.8^\circ$ of 1.5 meV. Note the change in slope that is a signature of the Fermi cut-off in the momentum distribution of $^3$He (see insert).

Also shown in Fig. 4 are polarization and analysis capabilities, that will often be essential in separating out the true magnetic contributions. The fact that we are discussing small-angle scattering with restricted beam geometry means that the white-beam polarizers presently developed, which rely on low-temperature nuclear polarization or differential absorption, can be used efficiently.

Momentum Distributions. At wave vectors sufficiently large for the impulse approximation to be valid, measurements of the scattering function $S(Q,E)$ give information about the momentum distribution of scattering nuclei, $n(p)$. In $^3$He at temperatures well below $T_F$ ($1.6^\circ$ K), $n(p)$ is expected to have a discontinuity at the Fermi surface, $p = p_F$, (see insert Fig. 5). This is reflected by kinks in $S(Q,E)$ at $E = \hbar Q^2/2M \pm Qp_F/M$ (see insert Fig. 5). Observation of these by neutron scattering would provide a direct observation of the Fermi surface in liquid $^3$He, analogous to measurements made in metals with electron Compton scattering.

The requirement of large $Q$ means that measurements of this kind are especially suited to pulsed-source instruments using epithermal neutron energies[10]. A preliminary measurement at IPNS[11] with $E_0 = 0.26$ eV yielded an approximate estimate of the width of $n(p)$, i.e. the mean kinetic energy per particle. However, the very high absorption cross section of $^3$He means that present pulsed sources cannot provide
sufficiently intense beams for high-resolution measurements to be carried out. To properly observe the kinks in $S(Q,E)$, we estimate that about five resolution elements will be needed on the relatively straight sections of the curve on either side of the kink. For $Q = 15 \text{ Å}^{-1}$, the curve in Fig. 5 implies that a resolution $\Delta E = 1.5 \text{ meV}$ will be required. This resolution must be accompanied by sufficient flux on sample to provide adequate statistics for a detailed shape analysis of $S(Q,E)$. An appropriate machine is the Ultra-High Resolution Chopper Spectrometer, Crawford et al. This instrument is designed for exactly the resolution required, $0.005 E_0$, only times have to be scaled up by 2 appropriate for an $E_0$ of 0.25 eV instead of 1.0 eV, and gives the following parameters:

\[
\begin{align*}
E_0 & = 0.25 \text{ eV} \\
\Delta E & = 1.25 \text{ meV} \\
\phi_s & = 1.1 \times 10^5 \text{ n cm}^{-2}\text{s}^{-1}
\end{align*}
\]

This flux is 20 times that available in the IPNS experiment, so that statistics should be 4-5 times better, certainly adequate for shape analysis.

In addition to studies of the effect of pressure, addition of $^4\text{He}$, etc. and other factors affecting the Fermi surface, we may anticipate the eventual capability to measure on polarized $^3\text{He}$. (This will require high magnetic fields and very low temperatures; however, measurements on solid $^3\text{He}$ at 0.5 mk and 0.1 T are already being carried out at IPNS.) In partially polarized $^3\text{He}$ one may expect two Fermi surfaces, for the majority and minority spins, both of which should show up in the n(p) measurement.

Neutron measurements at moderate to high Q have established the existence of the Bose condensate in $^4\text{He}$. The measurement of the effects of temperature, pressure, and addition of $^3\text{He}$ on the condensate fraction is currently an active area of investigation at present sources. Looking ahead to technological developments, we can anticipate n(p) measurements on spin-polarized hydrogen, another monatomic Bose system. In addition to measurements of broad features of n(p) such as the kinetic energy, it may be possible to ascertain the existence of a Bose condensate in spin-polarized hydrogen.

7. Conclusions

We have tried in this paper to review briefly four areas of pulsed-neutron science, accelerators that provide the
protons, targets into which they plunge, moderators which emit the neutrons for the scattering experiments, and certain aspects of pulse length. All four are areas requiring considerable research and it is safe to predict that ten years from now the ideas concerning all three will be radically different from those today. Moderators, in particular, are a rapidly developing field at pulsed sources.

We have ended this short paper with a discussion of three scientific areas that we believe will prosper at pulsed sources in the 1990s. This is, of course, by no means a complete list, but it is always important to realize that there is new science to be found and that neutron scattering in the 1990s will no doubt provide as many surprises and new results as it has over the last two decades.

Acknowledgments

We wish to thank the ANL staff for discussions on a number of points, in particular, R. K. Crawford, S. Ikeda, J. D. Jorgensen, R. L. Kustom, C. W. Potts, and D. L. Price.

REFERENCES

MAGNETIC DEVICES
(Session II)
Chairman

M. LAMBERT
France
Invited Paper

NOVEL BEAM BUNCHING METHODS BY PERFECT CRYSTALS AND ELECTROMAGNETIC MEANS

H. RAUCH
Atominstitut der Österreichischen Universitäten,
Vienna, Austria

Abstract

NOVEL BEAM BUNCHING METHODS BY PERFECT CRYSTALS AND ELECTROMAGNETIC MEANS.

The use of perfect crystals for installing new neutron small-angle scattering cameras provides advantages for measurements in the small \( \theta \)-range and for real-time experiments. A neutron resonator is proposed which is based on the combination of perfect crystal back-reflections in Zeman energy splitting. The neutron magnetic resonance system in combination with gated crystals can act as a pumping unit for neutrons and as a new pulse-shaping unit. It is shown how travelling magnetic waves can act as powerful neutron bunching units. The achievable velocity changes are around 5 m/s and, therefore, by a factor of 100 larger than in the case of neutron magnetic resonance systems. The advantage of expanding potentials for focusing neutrons from a source with a long pulse duration becomes obvious. Real gain factors higher than 10 are expected for properly designed systems.

1. INTRODUCTION

The Hamiltonian of the interaction of a neutron beam with a perfect crystal and with a magnetic field is known completely and, therefore, the wave function of the neutron beam behind such devices can be calculated. This enables the design of advanced beam tailoring systems.

The interaction with a perfect crystal is described by the dynamic diffraction theory, known for many decades in X-ray and electron diffraction /1-4/. One of its predictions is that there exists in Bragg diffraction a region of total reflection, not only in the forward but also in the Bragg direction \( \theta_B \). The width \( 2s \) depends on the coherent scattering length \( b_c \), the particle density \( N \), the geometrical structure factor \( F \), the wavelength of the neutrons \( \lambda \), and on the angle \( \chi \) between the reflecting plane and the surface of the crystal

\[
2s = \frac{b_c N |F| \lambda^2}{4\pi \sin 2\theta_B} \sqrt{\frac{\sin(\theta_B - \chi)}{\sin(\theta_B + \chi)}},
\]

This reflection width is for thermal neutrons around a few seconds of arc. It has been used for many years to achieve an extremely high-energy resolution by the invention of the backscattering
technique /5,6/, or an extremely high angular resolution by nondispersive double-crystal spectrometry /7-9/. Proper focusing effects and an increase of the reflection width can be achieved by using curved, vibrating, gradient or multi-stack systems /10-18/. Unfortunately, often the information about the wave function gets lost and a transition to the kinematic behaviour of mosaic crystal occurs.

Laue diffraction at monolithically designed multi-plate systems has been used to establish neutron interferometry /19,20/. This technique makes the phase difference of widely separated beams accessible as a new measuring quantity which permits a new kind of neutron physics experiment (e.g. /21,22/). Another interesting feature of dynamic diffraction is the appearance of a very narrow central peak in multi-plate Laue rocking curves /23,24/. Its width is given by the ratio of the lattice spacing d and the thickness D of the crystals (\(\Delta = 0.71 \frac{d}{D}\)) /25/), which is around a thousandth of a second of arc.

The interaction of neutrons with a magnetic field B is described by the magnetic dipole interaction of the magnetic moment \(\vec{\mu}\)

\[ H = -\vec{\mu} \cdot B \]  
which gives the basis of all magnetic neutron scattering phenomena, and for polarized neutron physics (e.g. /26,27/). The Larmor precession and the Zeeman energy splitting follow directly from this equation. The Larmor precession angle is used in neutron spin-echo spectroscopy as a measure for the neutron velocity which permits high-energy resolution with a less monochromatic and collimated beam /28,29/. There exists a coupling between the spin and the momentum space which becomes obvious by the static Zeeman splitting /30,31/ and by the neutron magnetic resonance energy shift /32/. These basic effects will be discussed in the following sections as additional components for advanced beam handling. In the case of static Zeeman energy splitting the total energy is conserved, but the kinetic energy changes owing to the force caused by the field gradient (\(F = \pm \mu \frac{\partial B}{\partial x}\)). However, in neutron magnetic resonance systems the total energy changes because of a time-dependent interaction, which causes an energy exchange between the neutron and the resonator of twice the Zeeman energy (\(\Delta E = \pm 2\mu B_0\) (see Fig. 1) /32/). For a 20 kG guide field \(\Delta E\) amounts to

\[ E = \pm 0.235 \text{ keV}, \]  
which is only a small fraction of the kinetic energy of thermal or cold neutrons, but nevertheless the ratio \(\Delta E/E\) is of the same order of magnitude as occurs for charged particles inside a single accelerator cavity - see Refs /33,34/. This has encouraged various proposals for electromagnetic beam tailoring systems /32,35-38/. These electromagnetic systems are in a certain sense alternative to mechanical systems like moving crystals /39,40/ or moving totally reflecting turbine plates /41/.

The reflection width of perfect silicon crystals for thermal neutrons and the Zeeman splitting of a field of about 10 kG are comparable and, therefore, various combinations of perfect crystal optics and magnetic energy transfer systems are also discussed in
FIG. 1. Proof of the energy exchange between the neutron and a resonance spin flip system /32/.
the following sections. It is shown how more neutrons of an available neutron flux can be compressed into a desired momentum, space or time interval.

2. PERFECT CRYSTAL SMALL-ANGLE CAMERAS

The measurement of the broadening of the very narrow rocking curve of perfect crystals in a non-dispersive arrangement can be used for small-angle scattering experiments in the \( Q \) range between \( 10^{-5} \) and \( 10^{-3} \) \( \text{Å}^{-1} \). The related resolution is much higher than for a standard neutron small-angle scattering facility where long beam paths and a fine collimation have to be used (e.g. /42/). The \( Q \)-resolution is strongly decoupled from the spectral and angular width of the beam, which gives a similar advance for momentum transfer measurements to that which exists for energy transfer measurements in spin-echo spectroscopy /28,29/.

The perfect crystal camera becomes superior to the standard small-angle scattering facilities for the investigation of objects larger than 710 Å and in certain cases even for objects larger than 960 Å /43,44/. This strongly favours the investigation of interparticle correlations which cause a strong \( \Omega \)-dependence of the scattering pattern at small \( \Omega \) values, but which are usually not observed in standard small-angle scattering experiments /42/. Prototype instruments exist at various places and they have shown their capacity for investigating large precipitates or voids, the domain structure and various lamella and membrane structures /45-51/. A sketch of an advanced set-up is shown in Fig. 2 /52/. The multiple spectrometer system provides a more effective use of the available neutron flux and permits a stereoscopic investigation of the sample. The rather short beam paths make the system also favourable for real-time experiments down to a time scale well below 1 ms. Channel-cut crystals should be used to reduce the tails of the rocking curves /53,54/ and Fankuchen cuts should be used to adapt high resolution to large samples, and less resolution to smaller samples because the beam cross-section varies conversely to the reflection width (Eq. (1)). These cameras measure in the same \( \Omega \) range as light scattering systems do, but the applicability of the first Born approximation, the sensitivity to magnetic systems, and the possibility of contrast variation make neutrons essential for investigations in this regime.

The perfect crystal camera is intrinsically a one-dimensional analysis system and, therefore, a deconvolution concerning the scattering in the vertical plane has to be performed as is routinely done in X-ray small-angle scattering /55/. The perfect crystal camera can easily be equipped with a polarization supplement by means of a multistage prismatically shaped magnetic field system /56,57/, separating the up and down polarization component sufficiently compared to the width of the diffraction pattern. To bridge the region between the perfect crystal and classical small-angle scattering regime the resolution of perfect crystal cameras has to be reduced. This can be achieved by using slightly mosaic crystals /46/ or, more effectively, by using curved, vibrating,
3. NEUTRON RESONATORS - GATED PERFECT CRYSTALS

Static macroscopic resonators for ultra-cold neutrons have been developed in the past on the basis of a Fabry-Perot interferometer known in light optics /60,61/. Here, I want to propose an active resonator based on magnetically gated perfect crystals placed in a backreflection position. The $\Delta k/k$ resolution for backreflection is determined by the crystal extinction length and it can be written as

$$\Delta k/k \sim 4 N_c b_c |F| d^2$$  \hspace{1cm} (3)

where $N_c$ is the number of unit cells per unit volume. The reflectivity curve can be well described by the Darwin formula (e.g. /2/) with a plateau of reflectivity 1 and a total width given by
Eq. (1). Owing to the Zeman splitting a magnetic field causes a shift of the \( k \) vector of the neutrons inside the field of

\[
\Delta k_m = \pm \mu B_m / H^2 k
\]

which corresponds to a relative shift of the crystal reflection curve to the neutron beam /62,31/. Therefore, neutrons can be caught between two perfect crystals if the magnetic field of the first crystal is changed properly. To get the system more effective a multi-plate system is proposed as shown in Fig. 3. The crystals are kept at different temperatures to shift the reflection curve of neighbouring crystals twice the Darwin width (Eq. (1)). The required temperature difference is calculated as

\[
\Delta T = \frac{1}{1 + \pi / 8N_b c |F| d_0^2} \cdot \frac{1}{\alpha}
\]

where \( \alpha \) is the coefficient of thermal expansion. By combination of these features a gated crystal system can be achieved as is shown in Fig. 3. The realization is facilitated by the fact that the reflection width of silicon crystal well matches with the required shifts induced by a temperature difference or by a magnetic field. For the (111) backreflection at silicon (\( \lambda = 6.275 \) Å) the required temperature difference between neighbouring plates is 16°C and the
FIG. 4. Resonator behaviour shown in a (x,t) diagram. The small dispersion of the beams is not visible.

magnetic field required to change from an open to a closed position is 12.6 kG. The shift occurs in both directions and therefore the beam remains almost unpolarized. The whole resonator should be packed into a totally reflecting guide tube to reduce the lateral losses. The losses during the crystal reflection owing to absorption, incoherent and inelastic scattering processes have been calculated on the basis of a removal cross-section of 0.4 b/63/ and the reflectivity formulae for absorbing crystals/2,3/ and they are quite small ($2.2 \times 10^{-5}$).

At pulsed systems the peak flux can be caught between the plates by a correct operation of the magnetic fields, and the neutrons can be released afterwards as is shown in Fig. 4 for a special situation. By means of correct time-dependent operation of the release magnets (R) very monochromatic pulses, time focusing effects, or a division of a long pulse into many short ones, can be achieved. An intense monochromatic and quasi-stationary beam can be achieved by putting an adjusted neutron magnetic resonance system (Section 4) between the resonator crystals. By means of this device neutrons are pumped from one crystal to the next and release the system with opposite polarization and with an energy corresponding to the ends of the reflection curves.

4. NEUTRON MAGNETIC RESONANCE BUNCHING

Inside a resonance flipper the Zeman energy can be exchanged between the neutron and the resonator system if the resonance conditions for spin inversion are fulfilled/64,31,32/. The kinetic energy changes at the entrance and at the exit of the guide field $B_0$ and the total (potential) energy changes inside the flipper coil (Fig. 1). It acts non-destructively on the wave function and therefore it represents a coherent optical component/65,32/. Owing to the smallness of the Zeman energy, a multi-stage system will
be needed; therefore, high flipping efficiencies and low depolarizations are required. To add the energy shift of the individual stages static flippers or a field inversion have to be applied between the stages. If the resonance frequency \( \nu_l = 2\mu B_l/h \) is applied, the flipping probability inside a constant guide field \( B_0 \) reads as:

\[
\eta = \sin^2 \frac{\mu B_l t}{2\pi}
\]

(6)

where \( B_l \) is the amplitude of the oscillating field and \( t \) is the time-of-flight through the coil, which depends on the velocity and the path angle \( \Phi \) through the coil \( t = l_0 \cos\Phi/v \). The acceptance for spin inversion and energy transfer can be enhanced if gradient fields are applied /68, 69, 37, 38/. This gives a flatter response curve which is of particular interest for ultra-cold neutron beam tailoring.

Neutron magnetic resonance devices can be applied to steady-state beams to pump neutrons into a certain energy interval or to polarize neutrons dynamically. The spatial dispersion of pulse beams can be used to influence neutrons of different velocity differently by a time-dependent operation of the neutron magnetic resonance system /32, 35, 36/. The pulsed and synchronized operation of flipper coils has been tested extensively in the past /70-74/. The action on the neutron beam of a correctly operating time-dependent neutron magnetic resonance system is shown schematically in Fig. 5a. Only small intervals \( \Delta v \) can be fed by this method; therefore, the conditions for bunching become very stringent if only the normal dispersion of the beam is used. To shift neutrons of a velocity around \( v_0 + n \Delta v_{HF} \) into an interval \( \Delta v \) around \( v_0 \) the dispersion has to be large enough to permit an individual energy change of the various beams. This determines the flight path \( L \) where the multi-stage system has to be placed behind the pulsed source providing a pulse of length \( \tau \):

\[
L > \frac{\nu_0}{\Delta v_{HF} - \Delta v} + \nu_0 (\Delta v_{HF} - \Delta v)
\]

(7)

where \( \nu_0 \) is the length of the individual neutron magnetic resonance units and \( \Delta v_{HF} \) its velocity change. This relation yields for the (111) Si backscattering interval (Eq. (3)), and for \( \nu = 10 \mu s, \nu = 5 \) cm and \( B_0 = 100 \) kG, a flight path \( D > 180 \) m. Although superconducting magnets can be used because the flipper coil can be switched on and off properly, the required parameters are difficult to achieve. The increase of the intensity of back-reflected neutrons is roughly \( 2^n \), where \( n \) is the number of stages.

The conditions for an effective use of such a device are strongly relaxed if the spatial separation is made by properly arranged backscattered crystals kept at different temperatures (see Section 3 and MUSIC spectrometer principle /75/).
FIG. 5. Chopping operation of a neutron magnetic resonance system for monochromatization of (a) a pulsed beam, and (b) in combination with correctly located backscattering crystals.
placed even 10 m behind the pulsed source if the distances $d$ of
the crystals fulfil the condition

$$d \geq \frac{\ell + \tau v_0 + (\Delta v + \Delta v_{HF}) D/v_0}{2}$$

(8)

Referring again to the (111)-Si backreflection case, the following
feasible parameters have to be fulfilled: $D = 10$ m, $B_0 = 20$ kG,
$\ell = 10$ cm, $\tau = 100$ $\mu$s, $d = 8.2$ cm, $\Delta T = 8^\circ$C and $\Delta B = 12.6$ kG at the
R crystals. Various other possibilities arise if a multiple L
crystal system and a proper energy transfer are also applied to the
re-reflected neutrons.

5. TRAVELLING WAVE FOCUSING

Following the techniques known in charged particle accelerator
physics it seems reasonable to consider forces travelling with the
particle beam. In the neutron case the electromagnetic force is
given as $F = \pm \mu_0 B/dz$ and its action can be increased considerably
compared to the static case (Fig. 1) if a proper magnetic potential
moves along the neutron path. Two typical cases of a fixed width
and an expanding oscillator potential are shown in Fig. 6. The
related Schrödinger equation for $x$- and $t$-dependent magnetic field
has to be solved but a lot can already be learned from the solution
of the classical equation of motion /38/. Within a fixed oscillator
potential which is moved with the velocity $v_0$ the acting force
on the neutron is $F = -2\mu_0 B_{max} x/w^2$. All neutrons within a certain
velocity interval fulfil an oscillatory movement with a circular
frequency

$$\omega = \sqrt{2\mu_0 B_{max}/mw^2} = \frac{v_{esc}}{w}$$

(9)

where $v_{esc}$ is the escape velocity for a neutron that is initially
at the centre $x = 0$. For a very narrow pulse ($\tau = 0$) where position
and the velocity are strictly correlated ($x = vt$) the phase of
the oscillatory movement is independent of $x$ and $v$, and all neutrons
reach a turning point at the same time and then all neutrons have
reached the velocity $v_0$ in the laboratory system. The focusable
velocity band is given as $2\Delta v = 2v_{esc}$, which corresponds to
6.8 m/s for a $B_{max} = 10$ kG field. This is by a factor $v/v_{esc} \sim 100$
larger than the energy shift of a neutron magnetic resonance system
(Section 4).

For practical purposes, pulse lengths $\tau \neq 0$ have to be
considered. When the oscillator potential is switched on at a
distance $D$ from the pulsed source, the length of the neutron pulse
is roughly given as $2\Delta x = \tau v_0 + \Delta v_B D/v_0$. Now, the phases of
the oscillatory motion are no longer independent of the velocity
and of the initial position. Nevertheless, pronounced focusing
effects can be achieved if the neutrons fulfil one quarter of a
full oscillation and the initial length of the neutron cloud is
smaller than the width of the potential ($\Delta x < w$). This determines
FIG. 6. Sketch of a travelling fixed width and an expanding magnetic potential.

the monochromatization time \( t_m (= \pi/2\omega) \), the length of the system \( (L = t_m v_o) \) and the velocity band \( 2\Delta v \) which can be monochromatized

\[
t_m = \frac{\Delta x}{v_{esc}} \left[ \frac{\pi}{2} - \arctan \sqrt{\frac{v_{esc}}{\Delta v} - 1} \right] \left[ 1 - \left( \frac{\Delta v}{v_{esc}} \right)^2 \right]^{-1/2}
\]

\[
2 \Delta v = 2v_{esc} \sqrt{1 - (\frac{\Delta x}{w})^2}
\]

The velocity distribution during and at the end of the monochromatization process is calculated from the formula

\[
\Delta v(t) = \sqrt{<v^2(t)>_{x,v} - <v(t)>^2_{x,v}}
\]

where \( v(t) \) corresponds to a classical oscillatory motion, the average \( < >_{x,v} \) has to be taken for all particles of the cloud.

Figure 7 shows typical results for an oscillator and a sinusoidal potential. The focusing effect and the gain factor are shown in Fig. 8 for two different pulse lengths of rectangular shape. At the focusing position the dimension of the neutron cloud reaches the dimension of the potential wall \( (2w) \) but the dispersion is less than it would be in vacuum. Instead of focusing in momentum space a similar system can be designed for focusing in space or time. Interesting acceleration or deceleration effects can be achieved if the neutron packet is initially located asymmetrically or on one side of the potential.

The basic limitations of the fixed width potential (Eq. (10)) do not exist for the expanding potential. In this case all particles
FIG. 7. Characteristic changes of the relative velocities of the neutrons along a travelling oscillator (above) and a sinusoidal (below) potential.
with $v \leq v_{\text{max}}$ can be kept within the potential if its expansion is properly chosen.

$$\frac{mv_{\text{max}}^2}{2} = 2\mu B_{\text{max}} \ln \left[ \frac{w(L)}{w(0)} \right]$$

(12)

Only the length of the system can become unreasonably long, which can be seen from the formula for the monochromatization time which follows from a series expansion

$$t_m^i = \frac{\Delta x}{v_{\text{esc}}} = \sum_{n=0}^{\infty} \left( \frac{\Delta v}{v_{\text{esc}}} \right)^{2n+1} \pi \frac{n}{j+\frac{1}{2}}$$

(13)

where $v_{\text{esc}}$ is defined by the initial width of the potential $w(0)$.

Figure 9 shows a comparison of the monochromatization times for a fixed width and an expanding potential for a very narrow pulse ($\tau \rightarrow 0$). The expanding potential is superior to the fixed width potential and a reasonable focusing effect can even be achieved for longer pulse widths as is shown in Fig. 8 for 100 $\mu$s pulse. These calculations have been done numerically, and more details will be given in a separate contribution to this conference /38/ and in a forthcoming paper /76/. There, some quantum mechanical considerations will also be discussed.
FIG. 9. Monochromatization times for a fixed width and an expanding potential for a narrow pulse (t - 0) as a function of the maximal focusing velocity \( t_{\text{esc}} = w/\nu_{\text{esc}} \).

The proposed layout of such an electromagnetic neutron bunching system is shown schematically in Fig. 10. It looks similar to an injector or extraction system of an accelerator and indeed much of the known accelerator technology can be adapted to the proposed neutron system /77,78/. For the neutron focusing system the required
rise times of the fields are considerably longer (\(\simeq\) \(\mu\)s) than they are for kicker magnets used at charged particle accelerators. Only the fields should be higher (> 10 kG). From the physical and technological aspect, no basic restrictions exist for the realization of such a beam tailoring system.

6. CONCLUSION

Up till now thermal neutrons are only produced by a statistical slowing down process inside the moderator of any source, and therefore they are widespread in real and momentum space. In future, there will be considerable interest in increasing the intensity of the experimental beams and in improving their definition concerning energy, divergence, polarization and phase. Therefore, advanced beam-handling devices will be required to shift neutrons into those intervals which are required by a certain experiment.

Perfect crystal devices act non-destructively on the wave function and can be used as phase-sensitive systems for new kinds of neutron physics experiments. Their high degree of perfection can also be used for an advanced beam collimation, which is essential for small-angle scattering, topography and interferometry. The comparability of the reflection width of perfect crystals and the Zeman energy shift of usual magnetic fields favours their combination as is discussed in Sections 3 and 4. This enables the realization of a neutron resonator and an advanced beam tailoring technique by a correct combination with a neutron magnetic resonance system. The most powerful tool seems to be the travelling wave bunching system proposed in Section 5. This method adapts experiences of charged particle accelerator beam handling devices to the neutron case. Instead of the electrical or magnetic field, the gradient of a magnetic field acts as the driving force. It should be again noted that the relative energy change per unit length is quite comparable for both kinds of radiation. Thus, the required equipment will be very expensive and, therefore, many guide tubes should be served by such a travelling wave unit. A high degree of flexibility should be kept to permit a beam modelling according to the requirements of the various experiments. Certainly this is facilitated by the fact that only electromagnetic forces are acting on the beam. In certain cases such devices can produce a continuous flux which is equivalent to the peak flux of a pulsed source. The phase space density is conserved for all proposed systems but the neutrons can be concentrated into those intervals which are used in a certain experiment. This increase is in many cases equivalent to an increase of the primary source strength, and therefore both possibilities should be used at advanced neutron scattering facilities in the 1990s.

ACKNOWLEDGEMENTS

Substantial support of J. Summhammer and L. Niel, as well as many fruitful discussions with G. Badurek, K. Hübner (CERN), D. Petrascheck (Linz) and H. Weinfurter, are gratefully acknowledged.
REFERENCES


ALVAREZ, L.W., BLOCH, F., Phys.Rev. 57 (1940) 111.


TRAVELLING WAVE AND NEUTRON MAGNETIC RESONANCE BUNCHING

H. RAUCH, J. SUMMHAMMER, H. WEINFURTER
Atominstitut der Österreichischen Universitäten,
Vienna, Austria

Abstract

TRAVELLING WAVE AND NEUTRON MAGNETIC RESONANCE BUNCHING.

A feasibility study has been performed to demonstrate the bunching and focusing properties of a travelling magnetic potential well on a pulsed neutron beam. Fixed width and widening potentials have been considered and it has been shown that both can be used for advanced and effective beam tailoring. A similar effect can be achieved by a multistage resonance flipper system acting on a beam in a pulsed and synchronous mode. The performance of the proposed devices is similar to beam handling components known at charged particle accelerators.

1. GRADIENT RESONANCE FLIPPER

Using the principle of magnetic resonance inversion developed by Rabi et al. [1], high frequency neutron spin flippers were developed [2], whose main advantage is to work within high magnetic fields, too. Later on interest was concentrated on another effect associated to the time-dependence of the magnetic field which causes an energy-shift in combination with a spin-flip [3,4]. After the experimental verification of this shift [5] suggestions were made to use this active component for different neutron optical devices [6,7].

The action of a space and time dependent magnetic field \( \mathbf{B}(x,t) \) on the neutron is described by the Pauli equation

\[
\left[ -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} - \mu \frac{\partial}{\partial t} \mathbf{B}(x,t) \right] \psi(x,t) = i\hbar \frac{\partial}{\partial t} \psi(x,t)
\]

where \( \mu \) is the magnetic moment of the neutron. \( \mathbf{B}(x,t) \) for a gradient HF-spin flipper can be written as

\[
\mathbf{B}(x,t) = \hat{x} B_1 \cos \omega t + \hat{y} B_1 \sin \omega t + \hat{z} B_0(x)
\]

The substitution \( \psi(x,t) = e^{-i(\omega t/2)\sigma_z} \phi(x,t) \) which corresponds to a transformation into the rotating frame, where the magnetic field depends on \( x \) only, gives

\[
\mathbf{B}_r(x) = \hat{x} B_1 + \hat{z} (B_0(x) + \frac{\mu \omega}{2\hbar})
\]
Separation of variables delivers

\[ \psi(x,t) = e^{-i(\frac{\omega}{2} \sigma_z + \frac{E}{\hbar})t} \chi(x) \]  

and

\[ [ -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} - \mu \sigma \cdot \vec{B}_r(x) ] \chi(x) = E \chi(x) \]  

Together with \( P := \chi^+ \sigma \chi \), Eqs. 3 and 4 describe the polarization and the final energy of the neutron beam as

\[ E_f = E + (P_2 - P_1) \frac{\hbar \omega}{2} \]  

which clearly shows the coupling of the energy shift to the change of polarization.

Since there is no time dependence in Eq. 4, one may conclude that there should exist a high-frequency analogue for each static neutron flipper and that the usual HF-flipper with homogeneous guide-field should not be the only way of getting time-dependent spin inversion. In principle, it is possible to get the typical shape of flip efficiency of the neutron resonators by other models too [8]. Proper resonance flippers using gradient fields were used for the spin inversion of a broad spectrum of UCN [9,10] and for the observation of the associated energy shift [11]. For these neutrons the condition

\[ \frac{1}{v} \left| \frac{dB_0}{dx} \right| \ll \gamma B_1^2 \]  

is fulfilled and, therefore, the beam behaves adiabatically [12]. For such fields \( (B_0(x) = B_0(0) + b_g x) \) there are still oscillations of the flip efficiency depending on the neutron wave length with the frequency

\[ \omega = \frac{\gamma m \hbar^2 B_1}{2h} \sqrt{C^2 + 1 + \frac{1}{C}} \arcsinh C \]  

In this equation \( C = (\hbar \sigma z)/(2B_1) \) denotes the relation between the amplitude of the HF-field \( (B_1) \) and the difference of the guide-field between the entrance and the center of the resonance coil of length \( \ell \) (Fig. 1).

Thus there are peaks of the flip efficiency close to 1 in the adiabatic as well as in the nonadiabatic range. Analyzing the numerical solution of Eq. (4) it was found that the positions of these peaks occur at
and that it is possible to design a HF-spin flipper with broad efficiency for thermal neutrons and with proper dimensions (Fig. 2). Such devices can be useful as optical active components because they can be combined to a multistage system acting on a broad neutron spectrum.
2. TRAVELING WAVE FOCUSING

The energy change of neutrons in the resonance system discussed before is caused by the time dependent interaction. Mechanical systems also exploiting this principle are the turbine for ultra cold neutrons [13] and Bragg reflection from moving crystals [14].

We now want to discuss a novel scheme, that uses time dependent magnetic fields and polarized neutrons. In comparison with resonance systems it has the advantage that no change of spin direction is needed and thus no depolarization occurs. The beam has to be pulsed to achieve the desired focusing effect. A simple representation of the scheme is given by the time-dependent potential barrier: polarized neutrons at first traveling in vacuum enter a region of space with a magnetic potential whose strength is purely time dependent. The magnetic field may be parallel or antiparallel to the neutron polarization. The neutrons have a total energy \( E = E_{\text{kin}} \) in vacuum. In the potential region their total energy is time dependent: \( E'(t) = E'_{\text{kin}} + \mu B(t) \). When they leave the potential region their energy has changed by an amount of \( \Delta E = \mu(B(t_2) - B(t_1)) \), with \( t_1 \), \( t_2 \) being the time of entrance and of exit from the potential region, respectively. This semiclassical consideration is reproduced fully by a recent quantum mechanical calculation for the special case of sinusoidal time dependence of the potential barrier [15]. Another aspect of the time dependent potential barrier, the interference phenomenon known as scalar Aharonov-Bohm effect, was also recently outlined [16].

But instead of having the neutrons pass successive regions of force, one can conceive of a region of force traveling with the neutrons. Let us consider a one-dimensional model where polychromatic polarized neutrons are emitted from a pulsed source located at the origin, with a pulse duration \( \tau = 0 \), causing the space time relation \( x = vt \). At the time \( t = t_s \) a traveling oscillator potential whose minimum position always coincides with the position of the neutron with velocity \( v_0 \), is switched on over the neutron cloud. The potential, in the rest frame of its minimum (now called potential frame), has the form:

\[
V(x') = \frac{\mu B_{\text{max}}}{x_b^2} x'^2
\]

(8)

\( B_{\text{max}} \) being the strength of the magnetic field at the position \( x_b \) (\( \mu B_{\text{max}} > 0 \) is assumed). At the switch-on time \( t_s \) a neutron with velocity \( v' \) and spatial coordinate \( x \) in the laboratory frame has velocity \( v'_A = v - v_0 \) and spatial coordinate \( x'_A = x - v_0 t_s \) in the potential frame. For times \( t > t_s \) the further movement of a neutron in the potential frame is an oscillation with frequency \( \omega \)

\[
x'(t) = x'_A \sqrt{1 + \frac{\omega^2}{\omega^2 t_s^2}} \sin(\omega t + \varepsilon)
\]

(9)
TRAVELLING MAGNETIC OSCILLATOR
POTENTIALS

TO EXPERIMENT

FIG. 3. A polarized neutron pulse travels for some time in a force field created by a magnetic oscillator potential. The original velocity interval is compressed to a narrow interval around $v_0$. A system of electromagnets can generate travelling waves of magnetic field strength to monochromate a steady stream of pulses.

$$v'(t) = \omega x'_A \sqrt{1 + 1/\omega^2 t^2} \cos(\omega t + \varepsilon)$$

where

$$\varepsilon = \arctg(\omega t_s) - \omega t_s \text{ and } \omega = \sqrt{\frac{2\mu B_{\text{max}}}{mx'_b^2}}$$

It is interesting now to see that the phase $\varepsilon$ does not depend on a particle's original velocity $v'_A$ or coordinate $x'_A$ at the switch-on time. Consequently, all particles simultaneously reach a turning point of their oscillation and thus $v' = 0$. If the traveling magnetic potential is switched off exactly at this moment, all neutrons of the pulse have velocity $v = v_0$ in the laboratory frame. A traveling oscillator potential applied to a neutron pulse thus acts as an active monochromator.

In practice, the applicability of this model is limited by the fact that the potential cannot become arbitrarily strong with increasing distance from the minimum. If only a field $B_{\text{max}}$ can be achieved, then the largest velocity interval that can be compressed is given by

$$\Delta v = 2 \sqrt{(2\mu B_{\text{max}}/m)[1 - (x'_w/x'_b)^2]}$$

where $x'_w/x'_b$ is the ratio of the width of the neutron cloud at $t = t_s$ to the distance of the two points of highest potential. For negligible $x'_w/x'_b$ and $B_{\text{max}} = 1$ Tesla one obtains $\Delta v = 6.8 \text{ m/s}$. Figure 3 shows a sketch of how such a system could be realized at a pulsed source having a high repetition rate.
FIG. 4. Comparison of the minimum monochromatization times for the monochromator with fixed potential width (M1) and with increasing potential width (M2) for a pulse of duration \( \tau = 0 \). At an energy ratio of maximum kinetic particle energy (in potential frame) to maximum potential of 0.8, M2 already needs only half the monochromatization time of M1. M1 cannot monochromate particle energies higher than the maximum potential.

The distance the potential must travel with the neutron cloud is given by \( L = v_0 t_m \), where \( t_m \) is the monochromatization time. For this model one obtains

\[
t_m = \frac{x_0}{2} \left\{ \pi - \arctg \left( \frac{\sqrt{2\mu B_{\text{max}}}}{m v_{\text{max}}^2} - 1 \right) / \sqrt{\frac{2\mu B_{\text{max}}}{m} - v_{\text{max}}^2} \right\}
\]

(12)

\( t_m \) becomes infinite for \( \frac{m v_{\text{max}}^2}{2\mu B_{\text{max}}} \) approaching 1 (see Fig. 4).

Shorter monochromatization times can be obtained with another scheme: a polarized neutron pulse is originally imbedded in a traveling oscillator potential in such a way that the extreme ends of the neutron cloud are situated at the points of the highest magnetic field, and thus at the points of the strongest force. In order to keep the slowest and the fastest neutrons from escaping the field region, the width of the potential increases with the dispersion of the neutron cloud. Let us follow the fastest particle again for a pulse duration \( \tau = 0 \). Then in the potential frame the force on the neutron is time dependent as \( x'_b \) is time dependent:

\[
F(t) = -\frac{2\mu B_{\text{max}}}{x'_b(t)}
\]

(13)
To decelerate the particle to \( v' = 0 \) the equation

\[
\frac{m v'^2}{2} \leq \frac{x_b^{\text{end}}}{x_b^0} = \int_{x_b^0}^{x_b^{\text{end}}} dx = 2 \mu B_{\text{max}} \max \ln \left( \frac{x_b^{\text{end}}}{x_b^0} \right)
\]

must be fulfilled. (\( 2x_b^0, 2x_b^{\text{end}} \) are the widths of the potential at the beginning and at the end of the monochromatization process.)

One can observe, that contrary to the former model, there is no limit to the maximum velocity that can be decelerated with a given achievable magnetic field \( B_{\text{max}} \).

Analytically this model does not lead to a simple expression for the particle motion. Here we will only give the monochromatization time \( t_{m2} \) [18]:

\[
t_{m2} = x_b^0 \sqrt{\frac{m}{4 \mu B_{\text{max}}}} \sum_{n=0}^{\infty} \left( \frac{m v'^2}{4 \mu B_{\text{max}}} \right)^{n+\frac{1}{2}} / \Pi (j+\frac{1}{2})
\]

A comparison of the monochromatization times of this and the former model is given in Fig. 4. For a given pulse and equal \( B_{\text{max}} \) the second model always has a shorter monochromatization time, meaning in practice shorter magnet systems.

Now the finite pulse duration time \( T \) has to be included. First, we will consider the monochromatization process in the model of a fixed width oscillator potential. Then the phase \( \varphi \) is no longer a constant for the whole ensemble. It becomes for a particle with \( x_A' \) and \( v_A' \):

\[
\varphi(x_A', v_A') = \arctg \left( \frac{\omega x_A' v_A'}{v_A'} \right) \quad \text{for} \quad v_A' > 0
\]

\[
= \pi + \arctg \left( \frac{\omega x_A' v_A'}{v_A'} \right) \quad \text{for} \quad v_A' < 0
\]

Thus, not all particles reach the velocity \( v' = 0 \) at the same time. What monochromaticity can still be obtained depends on the characteristics of the source in the velocity regime of interest. But when rewriting

\[
\omega x_A' v_A' = (2 \mu B_{\text{max}} / m)^{1/2} (x_A' / x_b^0 v_A')
\]

one notices that the possible values of \( \varphi \) can be confined to narrow intervals around \( \varphi = 0 \) and \( \varphi = \pi \), if \( x_A' / x_b^0 \) is small, that means that the neutron pulse must originally be fairly localized around the minimum of the potential. For a given spatial width of the pulse this means a low frequency \( \omega \) and thus a long monochromatization time.
As the then necessary magnet systems get linearly longer with the mean velocity $v_0$ of the neutrons, a realization for thermal neutrons seems feasible only in connection with backscattering instruments. Figure 5 shows a model calculation of the development with time of a pulse duration 20 μs and a rectangular velocity distribution in between $v_{\text{min}} = 637 \text{ m/s}$, $v_{\text{max}} = 643 \text{ m/s}$. With the parameters chosen the intensity increases by a factor 6.9 in a velocity interval of width 0.6 m/s, which is approximately the acceptance of backscattering instruments [17]. The length of the magnet system in this case has to be 15 m.

Using the monochromator of the traveling and widening potential, the final velocity distribution would hardly change, but the length of the magnet system could be cut to about half that length.

Finally, it should be noted that the above classical considerations can be shown to agree with quantum mechanical calculations, when the properties of the pulse as a whole are considered (Ehrenfest's theorem). Thus the procedure of first analyzing a classical
particle's behavior and then generalizing for the whole ensemble of a pulse is justified and will be described in a separate paper [18].

3. CONCLUSION

The calculation concerning the gradient resonance spin flipper has shown that the response function of such devices becomes remarkably broader than that of a standard HF-flipper. This enables the transfer of the resonance energy to a broader wave length band and to a more divergent beam which simplifies the use of neutron guides and the realization of multistage systems. A multistage system operating in a traveling wave mode can serve as an effective bunching device. There is no material needed in the beam and, therefore, this kind of monochromatization can be done while the neutrons are traveling along the guide tubes [19].

Even more effective systems can be realized by applying a traveling magnetic potential well with a fixed or a widening width. The calculations show that such a device provides an effective system for increasing the neutron density within certain velocity intervals and to reduce the spatial width of the beam. Various considerations show that a reasonable bunching and cooling effect can be achieved for feasible values of the pulse width, the length and the strength of the traveling magnetic potential well. This increases the capability of advanced neutron spectroscopy.

Useful discussions with G.Badurek and A.Zeilinger (Wien) and with K.Hübner (Geneve) are gratefully acknowledged. This work has been supported financially by Fonds zur Förderung der Wissenschaftlichen Forschung (project no. S42/02).

REFERENCES

PULSED REACTOR IBR-2 IN THE 'NINETIES

Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna

Presented by P. Pacher

Abstract

PULSED REACTOR IBR-2 IN THE 'NINETIES.

One may expect that in the 1990s a considerable part of neutron scattering experiments will be carried out on pulsed neutron sources. Pulsed reactors of the IBR-type are the founders of a family of pulsed sources of thermal neutrons. The first IBR-type reactor started operation in 1960 with a mean thermal power of 1 kW. In 1984 the IBR-2 reactor had a mean power of 2 MW, a pulsed power of 1,500 MW at 5 s⁻¹ repetition rate and a power pulse duration of 215 μs. The peak leakage current of thermal neutrons from the moderator surface reached 1 x 10¹⁶ n·cm⁻²·s⁻¹, which allows on the sample a mean flux of thermal neutrons up to 5 x 10⁷ n·cm⁻²·s⁻¹ monochromized by the time-of-flight method with an uncertainty of Δλ = 0.02 Å. The achieved parameters, being reasonably good for a wide range of experiments, are not to be considered as being a technical limit for this type of pulsed neutron source. The experience we already have allows us to expect development in the following directions: increasing of the reactor mean power, shortening of the pulse duration, installation of the liquid hydrogen moderator. The IBR-2 reactor is used in four main fields: physics of condensed matter, nuclear physics, fundamental and applied physics. The greatest activity is in condensed matter research. In addition to conventional structural and dynamic studies we intend to expand investigations to kinetics of transitions in the time range up to 10⁻⁴ s (time-resolved spectroscopy), properties of short-living non-equilibrium states, properties of matter under extreme conditions created in the pulsed mode (e.g., under the influence of superstrong magnetic fields). We expect that within the next decade, neutron methods will be increasingly introduced into different areas of research and technology.

Although the subject of the conference and our talk deals with the future developments in neutron scattering, it is worthwhile remembering that the future is based on the past. In this connection we should like to remind you that the first reactor that produced several power pulses per second and was used for neutron scattering experiments, was already constructed in Dubna at the Laboratory of Neutron Physics in 1960. Then this IBR reactor (Pulsed Fast Reactor) had an average thermal power of 1 kW and a pulsed power of 5 MW. Already the first experience indicated that the pulsed reactor is an efficient and convenient tool for investigations both in nuclear physics and in condensed matter research.

Since then, in our Laboratory a permanent activity has been in progress for the perfection of pulsed reactors. The idea of
Pulsed neutron sources soon became popular worldwide. At present about half the high flux neutron sources are the pulsed ones, and if we refer to new projects and sources under construction, this ratio will be about 2:1 for pulsed sources /1,2/.

The largest success in this field was the startup of the IBR-2 reactor /3,4/ (Fig. 1) which in 1984 achieved the following parameters /5/:

- Average thermal power: 2 MW
- Peak power: 1500 MW
- Power between the pulses: 100 kW
- Power pulse duration (FWHM): 215 \mu s
- Pulse repetition rate: 5 s\(^{-1}\)

The peak thermal neutron flux on the moderator surface is \(1 \times 10^{16} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}\) \((2)\)\(^{-1}\). The time structure of reactor power is shown in Fig. 2.

The visible surface of the moderator from the extracted beams is 400 - 800 cm\(^2\). The typical value of the time-average thermal neutron flux on the sample is about \(5 \times 10^7 \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}\) at a flight path of 15-20 m and at a monochromatizm of 0.05 Å. These parameters are advantageous for a wide range of research activities in condensed matter science, chemistry, biology and in other fields.
However, the reactor IBR-2 also has other advantages. In the economical sense it is not an expensive machine - this includes both the construction and operation expenses. In the event of 2500 hours of operation annually at 2 MW average power the nuclear refilling cycle is more than 10 years, the required electric power is about 0.5 MW.

The IBR-2 differs from other pulsed sources in the low repetition rate - 5 pulses/s. This allows one to use the time-of-flight method (TOF) even with very slow neutrons down to a velocity of 50 m/s. Finally, the spectrum of fast neutrons in the reactor core is up to 10 MeV, which makes the problem of shielding relatively simple and cheap.

The restricting factors on the IBR-2 are the relatively long power pulse duration and the limited life of the complicated mechanical equipment of the moving reflector (guaranteed lifetime is 10 000 h).

The parameters of the IBR-2 achieved in 1984 have not reached the limit and the next decade will be devoted to their improvement. The reactor operation experience indicates the possibility of a further increase of the average power level up to 3 - 4 MW, which means a proportional increase in the reactor flux. The further improvement is to reduce the power pulse duration. In the future we plan to use a new heterogeneous type moving reflector system /6/
FIG. 3. The core of the IBR-2 reactor with the planned heterogeneous moving reflectors.

(Fig. 3). This newly shaped reflector will have two forks rotating in opposite directions. Calculations and static experiments showed that this configuration will increase the rate of change of the reactivity significantly. The mechanical design is a rather hard job for the engineers. After solving this problem the power pulse duration is to be reduced to 120 µs. This will make the quality of the reactor much higher without any decrease of average power, or increase of pulse repetition rate. A more considerable decrease of pulse duration can be achieved by using an electron linear accelerator as an injector, i.e. in a booster mode of operation. The expected parameters in this mode are:

- power pulse duration: 10 µs
- average power: 0.5 MW
- repetition rate: 50 s⁻¹
- peak power: 1000 MW

The peak flux of epithermal neutrons (0.1 - 1 eV) is expected to be $1 \times 10^{17}$ n·s⁻¹·st⁻¹·eV⁻¹.

The third way of improving the quality of the IBR-2 reactor is connected with the construction of the liquid hydrogen moderator. The expected volume of hydrogen is about 1.5 - 2 litres and a heat emission due to radiational heating - about 200 W. This cold
The IRR-2 is mainly used in the following four fields: condensed matter physics, nuclear physics, fundamental physics and applied research. The main activity is in condensed matter physics; most of the beams are used for neutron scattering experiments. The main characteristics of some of the instruments are given in Table I. (The schemes for these instruments are shown in Figs 4-8). The subject of research covers a rather wide range of condensed matter problems. At present the traditional methods of studying the structure and dynamics are used for the investigation of more and more complicated systems. During recent years the following were investigated: The phase diagram of the super-ionic conductor CsHSO₄, the phase diagram of fast quenched nematic liquid crystals, the processes of hybridization of polyelectrolytes and biological membranes. Study of the dynamics was mainly concentrated on the inelastic incoherent neutron scattering from the hydrogen atoms in complicated molecular complexes (naphthalene + tetracyanobenzol, [Ni(NH₃)₆]⁺(NO₃)₂, dihalogenobiphenyl and others), in the hydrides of intermetallic complexes. Another incoherent process, the inelastic magnetic scattering in rare-earth intermetallics (TbNi₂, LaNi₂, HoNi₂, ErNi₂, LaCu₅, NdNi₅) was also successfully investigated at the IBR-2 reactor.

Having no time to go into the details of these investigations we should like only to note that the investigations in this field are characterized by more and more complicated structures and
<table>
<thead>
<tr>
<th>Spectrometer</th>
<th>Principal characteristics</th>
<th>Research programme</th>
</tr>
</thead>
<tbody>
<tr>
<td>Small-angle neutron scattering</td>
<td>$\phi \leq 3 \times 10^7 \text{ n cm}^{-2} \text{ s}^{-1}$</td>
<td>Low resolution structural investigation of physical, chemical and biological substances</td>
</tr>
<tr>
<td></td>
<td>$Q = 8 \times 10^{-3} - 2.0 \text{ Å}^{-1}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\Delta Q/Q \leq 0.1$</td>
<td></td>
</tr>
<tr>
<td>Four circle diffractometer</td>
<td>$\phi = 7.4 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$</td>
<td>Structural investigations of single crystals and polycrystals $d \leq 120 \text{ Å}$</td>
</tr>
<tr>
<td></td>
<td>$\lambda = 1.2 - 25 \text{ Å}$</td>
<td></td>
</tr>
<tr>
<td>Diffractometer for texture analysis</td>
<td>$\phi = 2 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$</td>
<td>Phase and texture analysis of low symmetry polycrystalline materials.</td>
</tr>
<tr>
<td></td>
<td>$0.5 - 4 \text{ Å}$</td>
<td></td>
</tr>
<tr>
<td>Polarization spectrometer</td>
<td>$\phi = 1.2 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$</td>
<td>Elastic and inelastic polarized neutron scattering. Structure and dynamics of magnetic materials.</td>
</tr>
<tr>
<td></td>
<td>$P \geq 94%$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\lambda = 0.7 - 7.5 \text{ Å}$</td>
<td></td>
</tr>
<tr>
<td>Inverted geometry spectrometer</td>
<td>$\phi = 1.3 \times 10^7 \text{ n cm}^{-2} \text{ s}^{-1}$</td>
<td>Incoherent inelastic neutron scattering. Dynamics of hydrogen. Investigation of crystal field splitting in rare-earth magnetics.</td>
</tr>
<tr>
<td></td>
<td>$\lambda = 0.4 - 4.5 \text{ Å}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\Delta \omega/\omega = 4 - 15%$</td>
<td></td>
</tr>
</tbody>
</table>
FIG. 5. Drawing of the polarization spectrometer. Curved neutron guides are the polarizer (1) and the analyser (2).

FIG. 6. Drawing of the single crystal diffractometer.

dynamical properties. The central problem is the relation between the dynamics and the structure. The manifestation of this relation in the case of simple systems is the phase transition. In the case of biological objects, where every macromolecule usually has its own function in a living cell, this problem acquires a new property and becomes a problem of accordance between the structure and the function of the macromolecule.
The pulsed reactor as a tool for neutron scattering experiments has some specific features which define the optimal areas of its use. It is possible to investigate simultaneously the structure and dynamics of a sample with the help of one instrument combining the time-of-flight diffraction and inverted geometry methods. This was done in the KDSOG-M spectrometer /7/, efficiently used in investigations of structures having the phase transitions of a bad reversibility, like CsHSO₄.

Another interesting feature of the pulsed reactor is the extremely high value of the peak flux on the sample. So, on the sample at a distance of 15 m from the moderator, the instantaneous thermal neutron flux reaches the value of $1.2 \times 10^9$ n·cm⁻²·s⁻¹ at an average flux of $3 \times 10^7$ n·cm⁻²·s⁻¹. This peculiarity allows one to investigate the kinetics of phase transitions, the nature of short-living non-equilibrium states, and the behaviour of the matter under extreme conditions available in a pulsed mode. For example, at present the construction of a magnetic device is being completed with the following parameters:

(a) pulse shape
- half period of a sinusoid
- pulse duration $0.7 - 1.5$ ms
- repetition rate $2 - 5$ s⁻¹
- amplitude of the magnetic field up to 500 kOe

(b) pulse shape rectangular
- pulse duration up to 400 µs
- leading edge 2 µs
- magnetic field up to 50 kOe
Of course we can apply not only magnetic pulsed fields. Much interest has been shown in the process of fast changes in structure, for example in muscles at an electro-stimulated muscle contraction, and in ferro-electrics in electric fields.

First we plan to deal with pure structural investigations only, but in the distant future they may possibly be extended to inelastic neutron scattering.

In the field of nuclear physics we want to emphasize the possibility of experiments with very small amounts of investigated materials, for example to use the targets from isotopes. Moreover, the high intensity of instantaneous neutron fluxes creates favourable conditions for the improvement of the effect-to-background ratio.

We have already noted that the low pulse repetition rate is the advantage of the pulsed source. Simple estimations show that at a flight path of 10 m and a 5 s⁻¹ pulse repetition rate, neutrons up to $\lambda \leq 80$ Å can be extracted without a frame overlap. Using a curved neutron guide the average flux around $\lambda = 30$ Å in the interval $\Delta \lambda = 0.1$ Å (which corresponds to the TOF resolution) is $6 \times 10^3$ n·cm⁻²·s⁻¹ even without a cold source. In this wavelength region a quite efficient quasi-elastic spectrometer can be built with a resolution of 1 μeV using the two-dimensional correlation technique /8,9/.

It is very difficult to make predictions for elementary particle physics, where new reactor experiments are rather rare. Nevertheless the IBR-2 reactor was successfully used in this field for searching for the hypothetical particle - axion /10/. The experiment made use of the periodic operation of the reactor to improve the signal-to-noise ratio, thus reducing the estimated upper limit for the cross-section of this particle by two orders of magnitude, and practically closing down the theoretical predictions.

Finally, the IBR-2 reactor is successfully used for applied research. Traditional fields, such as activation analysis and radiobiology /11/, have attained new possibilities as a result of
FIG. 9. Three-dimensional distribution function of orientations in granulite.

epithermal neutrons available with IBR-2. This allows one to extend the analysis to rare-earth materials, which need epithermal neutrons to become activated.

The high power doses in the extracted beams (up to 1000 rem/min) allows one to perform radiobiological investigations efficiently, for example to study the effects of pulsed irradiation on living organisms.

There exists increasing interest in texture studies, using the time-of-flight method on the pulsed reactor. In recent years it was shown /12/ that investigations of low symmetry materials with the time-of-flight method give a significant gain in time since all the reflections from polycrystalline materials are measured simultaneously. This allowed texture analysis to be introduced into geology.

To sum up the trends in applied research, one may say that in the future these investigations will show an upward tendency and will be used in different areas of science and technology. We have no doubt that the 'nineties will be characterized by further improvements of the existing pulsed sources, by the construction of
several new pulsed sources based on heavy current accelerators, and by the increasing spread of neutron methods in different research areas.

REFERENCES

/13/ BETEZL, M., et al., ibid, p. 753.
MAGNETIC DEVICES AND ULTRA-COLD NEUTRONS

(Session III)
Chairman

J. JANIK
Poland
NOVEL APPLICATIONS OF POLARIZED NEUTRONS AT PULSED SOURCES

G. BADUREK
Institut für Kernphysik,
Technische Universität Wien,
Vienna, Austria

W. SCHMATZ
Kernforschungszentrum Karlsruhe,
Institut für Nukleare Festkörperphysik,
Karlsruhe, Federal Republic of Germany

Abstract

NOVEL APPLICATIONS OF POLARIZED NEUTRONS AT PULSED SOURCES.
Two different kinds of beam deflecting systems are proposed that would shorten the length of the pulses emitted by a spallation neutron source without significant loss of neutron flux. One is based on magnetic prism refraction of long-wavelength polarized neutrons; the other exploits the possible modulation of the reflectivity of specially designed multi-layer neutron monochromators. Furthermore, a new polarized neutron inverted geometry spectrometer with electronically tunable magnetic resonator energy analysers is proposed.

1. INTRODUCTION

Neutron scattering experiments at pulsed neutron facilities require spectrometers which in general are different from those installed at continuous sources. The reason for this is primarily to make profitable use of the inherent time structure of the source, and consequently also of the very high available peak flux which exceeds the average flux in the thermal region of neutron energies by a factor of about 40.

A particular problem that arises in this context is the duration of the neutron bursts, which for the planned spallation source SNQ of the KFA Jülich will be approximately 300 µs in its initial LINAC version of the proton accelerator stage. This is too long, however, to achieve sufficient time-of-flight resolution without excessively long neutron flight paths. The pulse length has to be reduced therefore, either by means of mechanical choppers or by poisoning of the neutron moderator /1/. Since both methods lead to a decrease of neutron economy it is intended to shorten the primary pulses of the SNQ later on to about 120 µs by means of a special proton pulse compressor ring /2/.

In this paper we want to show an alternative approach for reducing the effective pulse length without necessarily losing much of the available neutron flux. The idea is to deflect half the
incident neutron packets rapidly from one direction into another so that at least two different experiments share the same pulsed beam. In the following section we will discuss two possibilities of realizing such a fast beam deflection system without mechanically movable components. Finally, in Section 3 the functional scheme of a new type of inverted geometry spectrometer with polarized neutrons is sketched, which allows optimal use to be made of the peak flux of a pulsed neutron source by proper variation in time of the scattered energy analyser windows.

2. PULSED BEAM DEFLECTION

Within a magnetic field \( \mathbf{B} \) the potential energy of neutrons is quantized into two Zeeman levels \( \pm \mu \mathbf{B} \) (\( \mu \)...magnetic moment of the neutron) according to the two possible spin orientations with respect to the field. The magnetic field therefore acts as a double refracting medium. If \( E_0 \gg |\mu| \mathbf{B} \) is the unperturbed kinetic energy of the neutrons outside the field region, the corresponding refractive indices for the two spin states are given as \( n_{\pm} = 1 \pm \mu B/2E_0 \). If a beam of polarized neutrons passes through a magnetic field region whose boundaries are mutually inclined to each other by an angle \( \phi \), it will be deflected upon polarization reversal by an angle \( \delta \),

\[
\delta = \frac{2|\mu| B}{E_0} \frac{\sin \phi}{\cos \phi + \cos 2\epsilon} \tag{1}
\]

In this equation \( \epsilon \) denotes the tilting angle of this field 'prism' with respect to symmetric neutron passage. Therefore, placing a fast spin-flipper in front of the deflection prism allows, in principle, a rapid change of the neutron beam propagation direction, as indicated in Fig. 1. Switching times of around a few microseconds are then readily achievable since the strong field of the magnet prism need not be commuted nor are mechanically movable components involved. But because of the smallness of the neutron magnetic moment (\( \mu = -6.03 \times 10^{-8} \) eV/T) it is by no means possible to achieve sufficiently large deflection angles with a single prism. However, if we assume about 10 prism stages each with an apex angle \( \phi = 120^\circ \), an asymmetry angle \( \epsilon = 25^\circ \), and a magnetic field \( B = 2T \), the angular deflection becomes as large as \( 1^\circ \) for a neutron wavelength of 10 Å (\( E_0 = 0.8 \) meV). So, at least for cold neutrons, it is absolutely feasible from a technical point of view to realize such a prism beam deflection system. Constructional details, as to whether the refracting fields should be produced by means of superconducting solenoids (\( B = 7T \)), or conventional ones that are driven by capacitor discharge power supplies (\( B = 25 \) T), naturally need much thorough theoretical and experimental examination. In particular, the latter system seems to be promising, since a small number of pulsed high field magnets would yield sufficiently large
deflection angles even for thermal neutrons. Further difficulties in methods, which have to be overcome, arise because of wavelength dispersion and beam depolarization effects.

The other, quite different method we propose for rapid beam deflection is sketched in Fig. 2. A thin-film multilayer consisting of alternating layers of two different materials is effectively a one-dimensional 'crystal', capable of Bragg-reflecting a mono-energetic neutron beam out of an initially poly-energetic spectrum /5/. The angular acceptance, and consequently also the reflected wavelength band of such a monochromator, can be strongly enhanced by sputtering several multilayers with different lattice spacings onto the same substrate /6/. It can also be used as a neutron polarizer if one of the bilayer materials is ferromagnetic, as for example in the case of Ge-Fe multilayer systems /7/. If we denote the scattering length density of the non-magnetic layer by $f_A = \rho_A b_A$, where $\rho$ is the particle density and $b$ the coherent nuclear scattering length, the corresponding scattering length density of the magnetic layer is given as $f_B = \rho_B(b_B \pm b_M)$. There, $b_M$ is the effective magnetic scattering length and the plus (or minus) sign depends on whether the neutron spin is parallel (or antiparallel) to the atomic spins which we assume to be aligned opposite to the direction of an applied magnetic field. It is important to note that the magnetic interaction of neutrons with atomic spins retains its full strength only if the spin magnetization $M$ and the neutron scattering vector $Q$ are oriented into orthogonal directions. It vanishes if both vectors have the same or mutually opposite direction, i.e. if $M \cdot Q = \pm 1$. According to the dynamical theory of diffraction, which takes into account extinction effects, for the integrated reflectivity of the first-order Bragg reflection of a multilayer, the relation

$$R \propto \left\{ \begin{array}{ll}
|\Delta f| \pm f_M \tanh \left[ \frac{2d^2N}{\pi} |\Delta f| \pm f_M \right] & \text{if } \hat{M} \cdot \hat{Q} = 0 \\
|\Delta f| \tanh \left[ \frac{2d^2N}{\pi} |\Delta f| \right] & \text{if } \hat{M} \cdot \hat{Q} = \pm 1
\end{array} \right. \tag{2}$$
The reflectivity of thin-film multilayers with equal nuclear scattering length density of the alternating magnetic and non-magnetic layers is zero if the layers are magnetized along the direction of the scattering vector $\mathbf{Q}$ (transmission case). A double-slit disk chopper in phase with the source pulses allows improvement of the signal-to-background performance of such a neutron beam deflection device (see text).

$$\Delta f = \rho_B a_B - \rho_B b_B$$

where $N$ is the total number of bilayers with lattice spacing $d$. One sees that for an application as a neutron polarizer the condition $\Delta f = f_M$ has to be fulfilled, since then the reflectivity is non-zero only for one spin state. On the other hand, for our purpose of beam deflection the nuclear scattering length densities of both layer materials have to be matched exactly so that $\Delta f = 0$. In this case the reflectivity can be suppressed completely by magnetizing the $B$ layers into the direction of the neutron momentum transfer vector as is indicated in Fig. 2. However, to achieve magnetic saturation a field of around $M_S/\mu_0$ ($M_S$ saturation magnetization, $\mu_0$ vacuum permeability) has to be applied in this case even for an ideally soft magnetic material, since the demagnetizing factor of thin layers is close to unity for a direction normal to their surface.

Since the multilayer removes only neutrons of a certain wavelength band from the incident beam, and also since its peak reflectivity is only around 90%, a disc chopper, which is synchronized with the source, has to be placed at some distance $L$ behind the multilayer beam deflector in order to achieve a reasonable 'on-off' intensity ratio. If the source pulse duration and period are denoted by $\Delta t_S$ and $T_S$, respectively, a single chopper slit of width $a$ yields a chopper opening time of just $\Delta t_S/2$ if the radius of the chopper disc is $r = aT_S/\pi \Delta t_S$ (SNQ: $\Delta t_S = 300 \mu s$, $T_S = 10 \text{ ms}$, $r = 30 \text{ cm}$ for $a = 3 \text{ cm}$). By means of a second slit located at a distance $D = L \tan \theta - 2a$ (if $D \ll r$) from the first one an appropriate time window is also set up for the deflected beam.
3. A NEW TYPE OF INVERTED GEOMETRY NEUTRON TOF SPECTROMETER

As was proposed first by Drabkin /9/ a zigzag-folded conductor, which produces a spatially alternating magnetic field oriented perpendicular to a constant field $B_0$, can be used as a wavelength selective neutron spin-flipper. On passage through such a field configuration each neutron creates, in its own frame of reference, a time-dependent high-frequency field with a frequency $\omega = 2\pi v/a$, where $v$ is the neutron velocity and $a$ the spatial period of the conducting foil oscillations. The spin-flip process takes place only for that neutron velocity

$$v_r = |\gamma|B_0 a/2\pi$$

(3)

which corresponds to a frequency equal to the Larmor frequency $\omega_0 = |\gamma|B_0$, with $\gamma = -1.83 \times 10^8$ rad/sT the neutron gyromagnetic ratio. Placing such a Drabkin resonator (in combination with a wavelength-independent spin-flipper) between two polarizing mirrors therefore leads to a neutron monochromator (or analyser) with a distinct transmission maximum at the velocity $v_r /10/$. 

![Diagram of the proposed neutron spectrometer](image_url)

**FIG. 3.** Sketch of the proposed neutron spectrometer. $S_1$, $S_2$, $S_3$ are supermirror polarizing guides, $R_1$, $R_2$ are a matched pair of Drabkin resonators. Their fields are perpendicular to the time-dependent field $B_0(t)$. The non-adiabatic flippers $F$ serve to achieve the correct orientation of the neutron polarization at the polarizers $S_2$ and $S_3$, as indicated by the arrows.
FIG. 4. Space/time diagram of the neutrons propagating from the source to the detector. By varying the analyser field $B_0$ inversely proportional to the time, all elastically scattered neutrons reach the detector irrespective of their initial velocity. The broken lines correspond to the case of an analyser with fixed scattered neutron energy.

To suppress the occurrence of unwanted side peaks in the transmitted spectrum, and to reduce the background caused by incomplete polarization, two Drabkin monochromators in series are optimal if their active lengths are in the ratio $L/L' = 1.56/1.1$. At least three polarizers, for example supermirrors /12/, are necessary for this purpose. The velocity or wavelength resolution of this double monochromator is $(\Delta v/v_r)^{1/2} = 0.68 a/l$.

We have proposed /13/ making use, at pulsed neutron sources, of the fact that such analyser systems can be tuned almost instantaneously to any desired neutron energy by rapid variation of $B_0$. In Fig. 3 an inverted geometry neutron time-of-flight spectrometer is sketched schematically, where in front of each detector a time-varying analyser is located. Each of these analysers consists of an appropriate pair of Drabkin resonators in combination with three polarizing supermirror neutron guides /14/. Two additional non-adiabatic wavelength-independent flippers provide for a proper orientation of the neutron polarization in front of each polarizer. For the sake of simplicity we assume for the moment that the analyser is placed immediately in front of the detector.
and that the time that neutrons need to pass through it is negligible compared with the time-of-flights $t_0$ and $t_1$ along the incident and scattered flight path, respectively. A neutron which arrives after a total time-of-flight $t = t_0 + t_1$ at the detector has undergone an energy transfer

$$\Delta E = \frac{m}{2} \frac{L_o^2}{(t - L/v_r)^2} - v_r^2$$  \hspace{1cm} (4)

upon scattering by the sample, where $m$ is the neutron mass and $v_r$ the velocity to which the analyser has been adjusted. From Eq. (3) it follows that for elastic scattering (i.e. $\Delta E = 0$) each neutron can reach the detector irrespective of its initial velocity if the field of the analyser varies inversely proportionally with time according to

$$B_o(t) = \frac{2\pi L}{|\gamma|a} \frac{1}{t} \text{ for } \frac{L}{v_{\text{max}}} \leq t \leq \frac{L}{v_{\text{min}}}$$  \hspace{1cm} (5)

as shown in Fig. 4 ($L = L_0 + L_1$). This field variation has to take place slowly enough, however, to consider $B_o$ as approximately constant during the neutron transit time $\Delta t = 1/a^2$ through an analyser with an active length $L_{\text{act}}$. This means that the condition

$$\Delta t \left| \frac{dB_o}{dt} \right| \ll B_o$$  \hspace{1cm} (6)

has to be fulfilled which leads to the condition $1/a^2 \ll t$. Without going into detail it has to be mentioned here that it is possible to reduce the active length of a Drabkin double
monochromator to $l_{act} = 1$ by means of a proper time shift between the two resonator fields, which in turn must increase with time according to the decrease of the incident neutron velocity. For elastic scattering the analyser could be placed anywhere between the sample and the detector if an appropriate scaling of $L$ and $t$ is taken into account in Eq. (5).

In the case of inelastic scattering a unique solution of the time-dependence of the resonator fields can be found only if the analyser is placed immediately behind the sample. Instead of Eq. (5) one then obtains

$$ B_0(t_o) = \frac{2\pi L_0}{|\gamma|a t_o} \left[ 1 - \frac{2\Delta E(t_o)}{m L_0} \right]^{1/2} \text{ for } \frac{L_0}{v_{max}} \leq t_o \leq \frac{L_0}{v_{min}} $$

Finally, in Fig. 5 reciprocal space scattering diagrams of possible constant energy-transfer scans are shown qualitatively for both elastic and inelastic processes.

REFERENCES

/13/ BADUREK, G., SCHMATZ, W., submitted to Z. Phys. B.
EXPERIENCE WITH SPIN ANALYSIS ON A TIME-OF-FLIGHT MULTIDETECTOR SCATTERING INSTRUMENT

O. SCHÄRPF
Institut Max von Laue - Paul Langevin, Grenoble

Abstract

EXPERIENCE WITH SPIN ANALYSIS ON A TIME-OF-FLIGHT MULTIDETECTOR SCATTERING INSTRUMENT.

By the simultaneous use of 28 supermirror spin analysers since January 1984 a variety of spin analysis experiments have been successfully carried out, such as scattering on liquid sodium to separate coherent and incoherent scattering, scattering by polymers in bulk and in solution, scattering by molecular crystals in the plastic phase, scattering by antiferromagnetic structures, spin glasses, Kondo alloys, paramagnets, garnets, semimagnetic semiconductors, with and without time-of-flight analysis. This paper gives a survey of special measurement methods and their evaluation with the simultaneous use of multidetector spin analysis and time-of-flight; for example, transmission calibration for different wavelengths, automatic adjustment of the analysers, multiple scattering corrections with and without flip of the spin, evaluation of paramagnetic scattering using multiple scattering vectors, and contour plots of magnetization density in para- or antiferromagnetic single crystals by rotation of the single crystals.

1. DESCRIPTION OF THE INSTRUMENT

The installation of 28 supermirror spin analysers with a cross-section of 50 x 100 mm² in front of 28 detectors was the essential change by which the conventional diffuse scattering machine D7 at the Institut Laue-Langevin (ILL) was modified into a powerful spin analysis instrument /1/. The possibilities of the mechanical chopper for time-of-flight and its electronics are simultaneously enlarged by the possible application of an electronic flipper chopper, which can be modulated with pseudo-statistical sequences or by other means, so that a fast chopper mode or even a Fourier chopper mode with maximum modulation frequency of 10⁵ Hz can be used, if one wants better resolution /2-5/. For this a small box with 16 detectors is foreseen with a thickness of 6 mm instead of 50 mm as in the normally used detectors. The smaller thickness enables better TOF resolution and a smaller width of 12 mm gives better Q resolution naturally with loss of count rate. If this is needed in a certain angular range the detector box can be fixed to the sides of each of the moveable detector tables. A doubly focusing pyrolytic graphite monochromator gives an increase of a factor of 5 of the intensity, which is very essential for polarization analysis, which suffers
always from too low an intensity. The monochromator is equipped with six step motors and six angular encoders, three for a reproducible wavelength change by rotation of the three monochromators in series and three for reproducible vertical focusing. One can select three different wavelengths: 3.1, 4.8 and 5.7 Å. For this purpose the whole machine, together with its outer shielding of 3.8 m diameter, can be moved on air cushions on a marble floor in the required direction.

2. ADJUSTMENT OF THE POLARIZERS

The essential feature discussed in this paper is polarization analysis with the possibility of time-of-flight analysis. By the simultaneous use of 28 supermirror spin analysers an immense gain in measuring time is accomplished. The use of polarizing crystals as analysers for polarization and energy would require 28 triple-axis instruments distributed throughout the whole angular range, an arrangement apparently impossible because of mechanical complications. In the case of the supermirror analysers, energy analysis can be done by TOF (time-of-flight) and it is only necessary to adjust the analysers for maximum transmission of the neutrons scattered by the sample. To do this each analyser has its own small back-and-forward driving synchronous motor, which can be controlled by a program that searches automatically for the direction of maximum count rate, moving the front end of the analyser around an axis at the detector end of the analyser, thus swivelling ±2°. As a sample for this objective one can use a quartz glass rod scattering very intensely without spin flip. Vanadium is less suitable because it scatters with and without spin flip. Without such an automatic adjustment the correct alignment of 28 analysers in a scattered beam would be unbearably tiresome.

3. FLIPPING RATIO: MEASUREMENT AND USE

The above-mentioned quartz glass scatterer is also well suited for measuring the resulting flipping ratio because it scatters very diffusely and only with non-spin flip. (Most suitable would be a pure isotope scatterer.) To measure the flipping ratio one uses a flipper between the polarizer and the quartz scatterer. This flipper consists of a Mezei coil with a horizontal field direction and a second coil wound directly on to this coil, but with vertical field direction. The latter serves for field correction purposes; it has to compensate inside the Mezei coil the vertical guide field, which is needed outside the flipper to avoid depolarization of the polarized beam /8,9/. If the neutron spins are first flipped and then scattered by the quartz, in an ideal case the analysers should transmit nothing. The really measured intensity behind the analysers consists then of the background, the neutrons transmitted by the polarizer or the analysers with the wrong spin direction, the neutrons which are not perfectly flipped, and the neutrons which are depolarized on the way. The measured flipping ratio \( R \), which is the
flipper-off/flipper-on counts ratio corrected for background, is
determined in the following way by the polarizer efficiency \( P_1 \),
the analyser efficiency \( P_2 \), the flipper efficiency \( f \) and the
depolarization \( \Pi \):

\[
R = \frac{(1+P_1P_2(1-N))}{(1+P_1P_2(1-N)(1-2f))}
\]

It should be as high as possible, normally between 20 and 50. It
is a measure of the imperfection of the whole set-up for
polarization and can be used to identify some faults.

The adjustment of the analysers can be done once and for all
at the beginning of a spin analysis period. The measurement of the
flipping ratio then shows faults in the analysers or the arrangement
of guide fields or flippers. If the flipping ratio is too poor (i.e.
less than 10) only for a certain analyser and there is no point on
the flight path of the neutrons where the guide field vanishes, the
respective analyser has to be repaired. It depends on the skill of
the experimenter to find and eliminate the faults. The measurement
of the flipping ratio has to be repeated at each experiment. It
changes with different guide field and flipper arrangement and is an
indicator of something depolarizing in the beam path - for example,
a zero-field region or a magnetic temperature switch in the cryostat
or a bad adjustment of the flipper. The required flipper current and
correction currents depend also on the fields surrounding the
flipper and must be adjusted each time something is changed, for
example from spin turn coils for paramagnetic scattering to simple
guide field coils for spin incoherent scattering or from an oven to
a cryostat.

The flipping ratio measurement is also essential for correcting
the data for finite flipping ratio. The inverse of \( R \) is a direct
measure of that part of neutrons with the wrong spin which is
transmitted by the analyser. This is seen immediately from its
definition:

\[
R = \frac{\text{intensity without flip}}{\text{intensity with flip}}
\]

For an unpolarized beam the incident intensities with and without
flip are equal and by the polarizer/analyser arrangement one
component is attenuated, in the ideal case to zero, the other
component is transmitted. \( R \) gives directly how many times the wrong
spin is more attenuated than the right spin. The inverse of \( R \) gives
the part of the wrong spin which is transmitted by the whole
assembly. This can be used for a simple correction. If one measures
the scattering of a sample with spin flip = \( I(\text{measured, sf}) \) and
without spin flip = \( I(\text{measured, nfl}) \) then the corrected value for
ideal flip ratio will be:

\[
\begin{align*}
I(\text{corrected, nsf}) &= I(\text{measured, nsf}) - (1/R) \cdot I(\text{measured, sf}) \\
I(\text{corrected, sf}) &= I(\text{measured, sf}) - (1/R) \cdot I(\text{measured, nsf})
\end{align*}
\]

This is an essential correction if the part without flip is high
and the flipping ratio is low for the scattered neutrons but high
for the neutrons in the direct beam. Its effect can be seen in
Fig. 1 for the case of a purely coherent scattering liquid Cs₂ showing I(sf) and I(nsf) as measured in Fig. 1(a) and corrected in Fig. 1(b).

4. VANADIUM CALIBRATION MEASUREMENT

A second necessary measurement is the vanadium calibration measurement. It is more complicated than the case without spin analysis, because it consists here of a measurement of scattering with and without spin flip. This has then to be corrected with background subtraction and with the flipping ratio measurement of the quartz. If the vanadium measurement is correct, the scattered neutrons with and without spin flip should show the flipping ratio 2. But even if one has only a 10% scatterer the flipping ratio
FIG. 2. Coherent scattering of (a) the pure solvent CS$_2$; (b) of the solvent +2% fully hydrogenated isotactic polystyrene upper curve corrected with corrected vanadium for absorption and multiple scattering, lower curve without this correction (0.8 scale); (c) Incoherent scattering of the 2% polystyrene, upper curve with corrected vanadium, lower curve without this correction; (d) Incoherent scattering of the solvent CS$_2$.

is never 2 without multiple scattering correction. It is usually 1.8 or less. This measure is a very good test for multiple scattering. This correction is normally done with a Monte Carlo program for neutron scattering with and without spin flip (see Section 6). I had to develop this since nothing corresponding to Blech and Averbach /10/, or Sears /11/, exists for polarized neutrons. As this correction must in any case be done one may take for the vanadium measurement larger rods to get more intensity and better statistics in a shorter time. Results of such calibrations show that the eight analysers first fabricated show nearly the same transmission. The boxes for the mirrors and the mirror adjustment were manufactured by a firm that worked well. The 16 analysers fabricated afterwards show transmissions that vary by a factor of 2 and I could not arrive at better results because of the poorer quality of the containers for the analysers. The flipping ratio shows similar variations. The effect of such a multiple scattering correction and absorption correction can be seen in Fig. 2, which shows the coherent and incoherent scattering of 2% fully hydrogenated isotatic polystyrene diluted in CS$_2$ as a function of the scattering vector, with and without this correction.

5. TRANSMISSION CALIBRATION FOR DIFFERENT WAVELENGTHS

For the time-of-flight measurements one needs the transmission for different wavelengths. As this will be different for each analyser, one needs 28 different transmission curves. The way to achieve this is as follows: the same scattering of a sample with wide enough quasi-elastic peaks is measured very carefully first without spin analysis, only with the polarizer installed, with
FIG. 3. Time-of-flight spectra of the coherent scattering (a) and the incoherent scattering (b) on liquid sodium drawn as curves over the $Q, \omega$ plane corresponding to the $Q, \omega$ values of the time-of-flight spectra of eight detectors at different scattering angles $2\theta$, $\lambda = 4.85 \, \AA$.

background subtraction and vanadium calibration but without multiple scattering and absorption correction. Then the same sample is measured in the same surroundings but with analysers installed. Under the assumption that scattering without spin flip together with scattering with spin flip should give the total scattering, after correction for background and finite flip ratio, the difference is due to the analyser transmission. This yields individual transmission curves for each analyser. A sample well suited for this purpose is liquid sodium. We also tried gaseous hydrogen of 18 atm pressure but the main intensity of this is too far away from the elastic peak in the TOF spectrum. For this procedure it would be desirable to have a simple reproducible method to remove the analysers out of the beam, something I am planning to carry out. At the moment this still has to be done manually. Here the question also arises as to what would be the highest energy that could be analysed. To date it is 10 meV, but for the next analysis period I shall shift this limit to more than 20 meV, already tested with one analyser. This option needs double the number of supermirrors, i.e. 3200
FIG. 4. The procedure described above for one angle of 62.4 degrees for a measurement with liquid sodium using a wavelength of 3.1 Å. The two measured TOF-spectra (a) without spin flip; (b) with spin-flip corrected for background and finite flip ratio; (c) is the TOF spectrum without analyser, and (d) is the TOF-spectra for coherent and (e) for incoherent scattering corrected for transmission of the analyser; (f) is the resulting transmission curve; the high values at the small wavelengths are due to frame overlap and belong to long wavelengths.

for all foreseen 32 detectors. I shall include it first for one bank with eight analysers to see whether it brings the expected gain.

Something else must be borne in mind. When measuring the coherent scattering of liquid sodium, shown in Fig. 3, it was difficult to understand why the peaks of the curves were not at the same energy and not at the elastic position. This is because the TOF measures always at a constant angle. In the case of a low incident energy of 4.85 Å wavelength these curves include very different Q values and therefore the peak of $S_{\text{coh}}(Q,\omega)$ is cut very obliquely (Figs 3 and 4).

6. CORRECTION FOR MULTIPLE SCATTERING

To get enough scattering one is inclined to use large samples with high scattering cross sections. It is then essential to be able to correct for multiple scattering and absorption. The Monte Carlo program for multiple scattering and absorption with and without spin flip can be used for elastic and quasi-elastic diffuse scattering. For liquid sodium for incoherent scattering this program uses a Lorentzian with Q-dependent width $DQ^2$ ($D =$ diffusion...
constant), and for the coherent scattering the Lovsey model of a monoatomic fluid /12/. For polymers in bulk a scattering law using a rotational EISF and a \( \Theta \)-independent Lorentzian in \( \omega \) with constant bulk temperature-dependent width /13/ is used. For elastic scattering the same program can be used - one need only to write for the number of wanted \( \omega \)-values the number 1.

For elastic scattering it is even easier to be independent of a model. \( S_{\text{coh}}(Q,\omega) \) and \( S_{\text{inc}}(Q,\omega) \) are introduced by a table of values at fixed \( Q \)-s and the program interpolates for the necessary \( Q \)-s at the fixed \( \omega \)-values. In elastic scattering one changes this table by trial beginning from the measured behaviour \( S_{\text{coh}}(0,0) \) so that the Monte Carlo program gives measured values again (self-consistently), and simultaneously the relation between coherent and incoherent scattering remains that measured for small \( Q \). With the same table the single scattering is then calculated without absorption \( J_{1\pm} \) with and without spin flip and a correction factor derived.

\[
J_{1\pm}^* = \frac{1}{(J_{1\pm} + J_{2\pm} + J_{3\pm} + J_{4\pm})}
\]

with \( J_{1\pm} \) = i-fold scattering intensity with or without flip. With this correction factor the measured curves are multiplied to obtain the corrected single scattering without absorption. Simultaneously the errors of the Monte Carlo averages are calculated by summing the squares of the differences from the averages. The program is a further development of the program of Johnson /14/ and runs in Fortran on the PDP 10 and in Basic on the PDP 11.

To show how essential such corrections are, I want to mention the following example. In measuring the temperature behaviour of the scattering by polymers in bulk one often observes a temperature-dependent decrease of intensity of scattering in small \( Q \) or even in transmission /15/. In a measurement of Gabrys et al. /13/ we also observed this, but using the Monte Carlo calculation including the rotational scattering law of Ref. /13/ I was able to correct for this. It results only from the high cross-section for inelastic multiple scattering.

7. PARAMAGNETIC AND ANTI FERROMAGNETIC SCATTERING

Many measurements were involved in order to separate paramagnetic and antiferromagnetic scattering from all other types of scattering, even in TOF. Here one essential trick to use in multidetector spin analysis is to measure with the incident spin direction in three orthogonal directions /7/. Then, according to

\[
\sigma_{sf}^{\text{Para}} \sim \frac{1}{2} (1 + \hat{\kappa} \cdot \hat{P})^2 \quad \text{and} \quad \sigma_{nsf}^{\text{Para}} \sim \frac{1}{2} (1 - (\hat{\kappa} \cdot \hat{P})^2)
\]
one obtains for the different cases:

\[
\begin{align*}
\sigma_{sf}^x &= \frac{2}{3} \sigma_{spin}^x + \frac{1}{2} (1+\cos^2\alpha) \sigma_{Para} \\
\sigma_{nsf}^x &= \frac{1}{3} \sigma_{spin}^x + \frac{1}{2} (1-\cos^2\alpha) \sigma_{Para} + \sigma_{isotop} + \sigma_{coherent} \\
\sigma_{sf}^y &= \frac{2}{3} \sigma_{spin}^y + \frac{1}{2} (1+\sin^2\alpha) \sigma_{Para} \\
\sigma_{nsf}^y &= \frac{1}{3} \sigma_{spin}^y + \frac{1}{2} (1-\sin^2\alpha) \sigma_{Para} + \sigma_{isotop} + \sigma_{coherent} \\
\sigma_{sf}^z &= \frac{2}{3} \sigma_{spin}^z + \frac{1}{2} \sigma_{Para} \\
\sigma_{nsf}^z &= \frac{1}{3} \sigma_{spin}^z + \frac{1}{2} \sigma_{Para} + \sigma_{isotop} + \sigma_{coherent}
\end{align*}
\]

with \(x,y,z\) = direction of the incident spin polarization, \(sf = \) spin flip, \(nsf = \) non-spin flip, \(\alpha = \) intensity, \(\alpha = \) angle between scattering vector \(\vec{k}\) and \(x\) direction, \(x\) and \(y\) in the scattering plane and \(z\) normal to the scattering plane, \(\vec{R} = \) unit vector in the direction of the scattering vector and \(\vec{P} = \) unit vector in the direction of the polarization. If one combines

\[\sigma_{sf}^x + \sigma_{sf}^y - 2\sigma_{sf}^z = \frac{1}{2} \sigma_{Para}\]

for spin-flip intensities and follows a similar procedure for the non-spin flip intensities one obtains directly the paramagnetic scattering alone or the antiferromagnetic scattering without even a special correction for the background. This is valid for each detector in the scattering plane, i.e. it is applicable for a multi-detector. Only if one has to correct in this case for finite flip ratio and the flip ratio is not the same for the three different directions, must the background also be measured. If the flip ratio is the same for the three different directions of spin one does not need the background and can correct with the same combination of \(x,y,z\) for the other spin state and the flip ratio, i.e.

\[\frac{1}{2} \sigma_{Para}^{corr} = (\sigma_{sf}^x + \sigma_{sf}^y - 2\sigma_{sf}^z) - \frac{1}{R} (\sigma_{nsf}^x + \sigma_{nsf}^y - 2\sigma_{nsf}^z)\]

and similarly for the other spin combination. The error in omitting this correction may be 10% if the flipping ratio is only 10.

We carried out such a measurement for the elastic scattering of a single crystal of a spin glass, where a superparamagnetic part of the magnetic scattering near the (000) direction can be seen and an antiferromagnetic part of the scattering in certain preferred directions. The single crystal was rotated about the (001) direction in steps of 5 degrees. We measured the scattering for each spin direction for 30 minutes. It was possible to observe the results even during the measurements by contour plots in the k-space. In the magnetic peaks found in this way we also looked with time-of-flight. This gave very narrow, practically only elastic peaks which could
FIG. 5. Magnetic scattering of the semi-magnetic semiconductor Cd$_{35}$Mn$_{65}$Te
(a) In the quasi-antiferromagnetic phase at 6K
(b) at 100K showing still structured magnetic scattering.
The points are the positions of the analysers.

not be resolved (we used the pseudo-statistical flipper chopper to get better resolution but then the background noise was too high and the background in this case cannot be measured separately). The elastic measurements correspond to the results of Ref. /16/. Measurements on a garnet powder for different temperatures showed that at T$_N$ the antiferromagnetic peaks disappear but the antiferromagnetic short-range order does not disappear till 200K. In this case the TOF spectra of the paramagnetic part were ~ 1.4 meV wide, corresponding to the T$_N$ of 10K. This width remains the same above and below T$_N$ as it is only a measure for the exchange coupling between the paramagnetic electron spins. In the case of solid oxygen, whose TOF spectrum we also tried to measure, the width was so wide that our present set-up was insufficient. For this new analysers with 2.1 Å cut off wavelength are required.

In the semi-magnetic semiconductor Cd$_{35}$Mn$_{65}$Te with the non-absorbing isotope of $^{114}$Cd we measured on a single crystal at 6K and at 100K. At 6K we saw clear antiferromagnetic peaks which at 100K were smeared out to an antiferromagnetic Debye-Scherrer ring that also indicated some beginning short-range order due to liquid-like next-neighbour interaction (see Fig. 5). Looking carefully at 6K it seems that this smeared-out ring was already there with the same size, indicating the same behaviour as with the garnets. Some samples such as CeMg and solid oxygen were also measured on the instrument D5 with polarization analysis. The measurements differ essentially. It seems that, owing to the large incident energy and the large analysed energy window on D5, the results include
FIG. 6. Separation of coherent and incoherent scattering of isotactic polymethyl methacrylate chains partially deuterated.

FIG. 7. Coherent scattering for different structures of polymethyl methacrylate.

Excitations which, in the case of the low incident energy of D7 of 3.48 meV or maximum 8.51 meV are not present. To see this clearer it must be possible to analyse also the energies on D5, maybe with TOF, which should be not so difficult.
8. SEPARATION OF COHERENT AND INCOHERENT SCATTERING IN POLYMERS

Here I only want to show some results which give an impression of what is possible in this field with polarization analysis. Without analysis the second coherent peak, shown in Fig. 6, disappears because the incoherent scattering behaves in a way that hides this peak. With polarization analysis this peak is well resolved and the behaviour of the incoherent scattering can be separately investigated without uncertain assumptions. Figure 7 shows the behaviour of the coherent scattering for different structures of polymethyl methacrylate. Even the fully hydrogenated sample clearly shows a coherent peak. The insert shows the monomer of the chain and how it is deuterated.

I would refer back also to Fig. 2, where the difference between the curves (a) and (b) gives practically the scattering of the holes in the solvent CS$_2$, filled by the fully hydrogenated and thus nearly not coherently scattering polystyrene.

The results of the measurements indicated here will be published more extensively elsewhere. They were carried out together with different groups - W. Gläser (liquid sodium), R. Gabryš (polymer in bulk), B. Barbara (CuMn spin glasses), M. Rawiso (polymer in solution), T. Brückel and W. Prandl (garnets), J. Schweizer and A. Plachty (solid oxygen), U. Steigenberger (semimagnetic semiconductors), J. Pierre and R.M. Galera (CeMg Kondo alloy). I publish here only my own contributions to the experiments, which I regard as interesting for all users and others interested in polarization analysis. Further developments in polarization analysis are expected from the use of efficient polarizers for short wavelengths, perhaps with the newly developed polarizing microguides /6, p.91/.

REFERENCES

/2/ SCHARPF, O., FUJARA, F., BADUREK, G., Test of a Mechanical Fourier Chopper at D7, Institut Laue-Langevin Rep. 84SC28T.
PRODUCTION AND USE OF ULTRA-COLD NEUTRONS

K.-A. STEINHAUSER
Fakultät für Physik,
Technische Universität München,
Federal Republic of Germany

Abstract

PRODUCTION AND USE OF ULTRA-COLD NEUTRONS.

Ultra-cold neutrons (UCNs), show a number of striking phenomena owing to their low energy (\( \sim 10^{-7} \) eV) and their long wavelength (\( \sim 1000 \) Å). They are strongly affected by gravity and magnetic fields and undergo total reflection from certain materials at any angle of incidence. As they are unable to penetrate material walls, UCNs cannot be extracted directly from a reactor source. Since their discovery in 1968 a number of different techniques of re-creating UCNs from originally faster neutrons have been developed: (a) down-scattering in a converter, (b) deceleration in the Earth's gravitational field, (c) spectral transformation to the UCN energy range by reflection from receding mirrors, (d) downscattering in liquid helium-4 at temperatures below 1K. As a result of their total reflection and sensitivity to magnetic fields, UCNs can be stored in material 'neutron bottles' or by suitable magnetic field configurations. This offers the possibility of investigating the properties of the neutron itself with very high accuracy. Such measurements include the search for the neutron electric dipole moment and the determination of the neutron beta decay lifetime. The unique properties of UCNs open up the possibility of new applications, such as the investigation of matter wave phenomena in synthetic systems of almost macroscopic dimensions or, as another example of neutron optics, the development of a neutron microscope. Recently, UCNs have been utilized as a tool for investigating condensed matter. The slow dynamics in macromolecules and biological substances can be measured, exploiting the high resolution of a gravity spectrometer.

INTRODUCTION

Neutrons moving at extremely low velocities (\( v \sim 5 \) m/s) are commonly known as "ultracold neutrons". Although they are, in principle, not different from faster neutrons, UCNs exhibit a number of interesting and unusual phenomena owing to their low energy (\( \sim 10^{-7} \) eV) and their long wavelength (\( \sim 1000 \) Å). Dealing with UCNs various interactions, which are usually negligible in thermal neutron research because of their weakness, have to be taken into account: (1) the earth's gravitational field (10^{-7}eV/m) causing strongly curved flight parabolas, (2) the interaction of the neutron's magnetic moment with a magnetic
field (6x10^-8 eV/Tesla), and (3) the interaction with the scattering potential. The latter gives rise to one of the most prominent features of UCNs - they are capable of being totally reflected from the surface of many substances, at any angle of incidence, even normal to the surface, if their kinetic energy is lower than the averaged Fermi scattering potential of the wall. For further information on the properties and peculiarities of UCNs see, e.g., Steyerl [1] and Golub and Pendlebury [2].

PRODUCTION OF ULTRACOLD NEUTRONS

One of the main problems of UCN physics is the difficulty of producing UCNs. Although they exist in the extreme low-energy fraction of the Maxwell-Boltzmann distribution, UCNs cannot be extracted directly from the moderator of a reactor as they are unable to penetrate material walls owing to total reflection or large absorption. They have to be re-created from originally faster neutrons. The initial experiments were performed at Garching by Steyerl [3] and at Dubna by Lushchikov et al. [4] in 1968. In the following years a number of different techniques of producing UCNs have been developed:

(a) Down-scattering in a small moderator - a converter - placed into a beam tube nose. In the same way as in a cold source, the UCN flux may be increased by cooling the converter.
(b) Deceleration by the action of gravity in a vertical or inclined neutron guide tube. Such a vertical guide tube has been used in the original facility at Garching [3] and is now being used at Leningrad [5], where it provides the strongest UCN beam at present available.
(c) Spectral transformation by the use of a mechanical "neutron-turbine". Applying Liouville's theorem to the deceleration of neutrons from velocity v1 to a velocity v2 by means of receding mirrors, we obtain an enhancement factor v1/v2, which may appear in several ways - in steady-state arrangements like the "Garching type turbine" [6], using totally reflecting mirrors, the beam cross-section is enlarged, whereas for a special geometry of moving Bragg reflecting crystals the short pulses, e.g. from a spallation neutron source, are stretched in time to produce a long intense UCN pulse [7].
(d) Down-scattering of 10 Å neutrons in, probably, the best converter, namely helium-4 at a temperature below 1K. In this "super-thermal source" invented by Golub [8] a high UCN density is accumulated in a neutron bottle filled with liquid helium. The maximum UCN density is limited by the storage time of the UCN bottle and can reach values much higher than in a normal converter with the same temperature.
As an example, the concept of the new "vertical source" at present in preparation at the Grenoble reactor may be described as follows: A vertical guide from the large D_2 cold source feeds a "total reflection neutron turbine" of the Garching type. In this device [6] the incident continuous beam of neutrons with an initial velocity of 50 m/s is transformed to an intense UCN beam of much wider cross-section by multiple reflections from a series of curved moving blades. Thus, the high intensity provided by a cold source of a high-flux reactor can be optimally used. We hope that this facility will supply a number of experiments with an extremely high-intensity beam of about 6000 n/cm^2/s.

USE OF ULTRACOLD NEUTRONS

It was first pointed out by Zel'dovich [9] in 1959 that UCNs can be trapped and stored in material cavities. The first systematic storage experiments were reported in 1971. But these experiments rendered storage times about ten times smaller than expected. This problem, which puzzled physicists for nearly ten years, has apparently been solved. It was convincingly demonstrated that the reduced storage times may be attributed to upscattering from surface contaminations [10,11]. Another way of confining neutrons arises from the fact that UCNs are strongly affected by magnetic fields. Hence they can be trapped in suitable magnetic field configurations.

This unique feature - the possibility to store UCNs for long periods in contrast to thermal neutrons, which have to be studied in flight - allows precise measurements of the properties of the neutron itself. A group at Bonn [12] has built a superconducting magnetic storage ring and has tested it at Grenoble. The losses in this torus are so small that an improved direct measurement of the neutron's lifetime for beta decay, which is at present known to an accuracy of about 1.5%, seems possible. Shapiro [13] was the first to suggest exploiting the confinement of UCNs in a material bottle for a sensitive search for the neutron's electric dipole moment. The existence of an electric dipole moment of any elementary particle would be of great interest as it would violate not only parity invariance but also time reversal symmetry. Two groups, one at Leningrad [14] and one at Grenoble [15], have constructed apparatus measuring the electric dipole moment in a magnetic resonance type experiment with bottled neutrons in an electric field. The best value for the electric dipole moment attained up to now is (-2 ± 1) x 10^{-25} e cm [14].

Another branch of UCN physics is UCN optics. Matter wave phenomena may be studied in macroscopic systems using UCNs. For this purpose a high-resolution gravity diffractometer has been
FIG. 1. Resonances observed in transmitted UCN intensity measured as a function of neutron energy. The two maxima correspond to a level splitting in a coupled-resonator potential created by the synthetic layer sequence shown in the inset diagram.

built at Garching [16]. This instrument makes use of the property of the gravitational potential to depend only on the vertical height. The height of fall between an entrance slit and the movable sample is a precise measure of the vertical component of neutron velocity. As no interference between horizontal and vertical motion occurs, a fairly wide horizontal velocity range can be exploited. Thus, in spite of the very high resolution of 2 neV the intensity is still appreciable. In a series of neutron optics experiments it was possible to demonstrate wave optical properties of particles in synthetic systems - mirror reflection, diffraction from a ruled grating, interference at a thin film, neutron resonances in stratified media, tunnelling splitting for two coupled resonators, and transmission bands in a system of many potential barriers. As an example to demonstrate a typical wave phenomenon, neutron resonances in a synthetic potential (quasibound states) are shown in Fig. 1 [17].

Another application of UCN optics is image formation. A neutron microscope should provide quite different information from that gained by light or electrons owing to the possibility of marking parts of, for example, biological samples by proton-deuteron exchange. For thermal or cold neutrons no satisfactory focusing systems are available. UCNs, however, owing to their unique property of being totally reflected, allow us to use special focusing mirrors. Thus, the development of efficient
systems for magnified image formation appears to be feasible. In contrast to light optics the strong curvature of UCN beams in the earth's gravitational field leads to chromatic aberrations. One way of compensating for these errors consists in "crossing" a concave mirror with a Fresnel-zone-plate, thus obtaining a "zone mirror", a focusing neutron-optical system similar to lenses or concave mirrors in light optics. Such a device has been successfully tested at the Grenoble reactor yielding sharp images of an object slit with a magnification of up to six [18]. In another set-up the chromatic aberrations of a parabolic mirror are compensated for by a second spherical mirror. This is possible if the neutrons are allowed to pass the maximum of their flight parabola between the two mirrors. A prototype of this "neutron microscope" has been installed at Grenoble, showing a sharp image of a slit with a magnification of 50. In a modified version of the two-mirror system, the chromatic aberrations are no longer corrected. But the overall chromatic errors are kept small by reducing the dimensions of the apparatus and using faster neutrons (~12 m/s), which are reflected by multi-layer mirrors.
Recently UCNs have been used as a tool for investigating matter. For this purpose the ultra-high resolution gravity spectrometer NESSIE (NEutronen Schwerkraft Spectrometer) has been developed and put into operation at Garching [19]. A scheme of this spectrometer is shown in Fig. 2. In this instrument a precise definition and analysis of the kinetic energy of the neutron is achieved by utilizing the reach of the flight parabolas of very slow neutrons (10 m/s) in the earth's gravitational field. Numerically, the reach dispersion has a value of 2 cm/neV. High-resolution spectroscopy most favourably uses UCNs because, at given resolutions in momentum and energy transfer, the intensity increases quadratically with wavelength. Additional intensity is gained by several focusing techniques like reach focusing (the maximum reach is stationary, thus allowing wide beam divergences) and application of elliptical mirrors in the analyser. By these means the world's best resolution of $(16.5 \pm 0.3)$ neV (FWHM) could be demonstrated. It is expected that an intensity gain by a factor $10^4$ can be achieved for this instrument when it is moved from the small FRM reactor at Garching to the new UCN source at Grenoble. The instrument can provide highest-resolution data on the slow dynamics in macromolecular systems. For a polymer in solution (polydimethylsiloxane in C₆D₆ at 70°C) a quasi-elastic line broadening of $(7.0 \pm 3.5)$ neV has been observed at a momentum transfer of 0.022 Å⁻¹. Furthermore, the instrument is suitable for investigating biological samples. Preliminary measurements on a model membrane show indications of quasi-elastic line broadening.

ACKNOWLEDGEMENTS

The author thanks Prof. A. Steyerl, who introduced him into the field of ultracold neutrons, and other colleagues for many stimulating discussions. The experiments of the Munich group gained from the help received from the institutes at Garching and Grenoble. Financial support was provided by the German Bundesministerium für Forschung und Technologie.

REFERENCES


STEADY-STATE SOURCES
(Sessions IV and V)
Chairmen

Y. ISHIKAWA
Japan
(Session IV)

J. BROWN
Institut Max von Laue - Paul Langevin
(Session V)
Invited Paper

MAGNETIC SCATTERING AND POLARIZED NEUTRONS

G. SHIRANE, C. F. MAJKRZAK
Brookhaven National Laboratory,
Upton, New York,
United States of America

Abstract

MAGNETIC SCATTERING AND POLARIZED NEUTRONS.

The last few years have witnessed considerable progress in the use of polarized neutron beam techniques for the study of condensed matter physics. Among the methods actively pursued at present are neutron spin echo, as pioneered by Mezel, for ultra-high-energy resolution and the energy-integrated magnetic scattering measurement technique developed by Brown and Ziebeck. We have concentrated on a medium-resolution range utilizing 5 to 100 meV polarized neutrons. This review deals with recent work at Brookhaven on the development of the triple-axis neutron scattering technique with polarization analysis. The stage has now been reached where quantitative characterization, in absolute units, can be done for a wide range of energy and momentum transfers in paramagnetic scattering. Some examples of recent inelastic measurements on 3d ferromagnets, as well as diffraction studies of multilayer thin film structures, are discussed.

I. INTRODUCTION

The polarized beam technique has been widely utilized for the study of spin density distributions in ferromagnets, following the pioneering work of Shull and Nathans[1] in the 1950's. This technique requires only a double axis configuration, without analyzer, and the magnetic scattering length $p$ is scaled to the nuclear length $b$ by the flipping ratio $R$:

$$ R = \frac{(b + p)^2}{(b - p)^2} $$

(1)

For this purpose the flipper $F$ (see Fig. 1) is placed between the monochromator $M$ and the sample $S$. Then in 1969, the classic paper by Moon, Riste and Koehler[2] appeared which advocated the use of polarization analysis after scattering. In particular, when a magnetic field $\mathbf{H}$ is applied parallel (HF) to the scattering vector $\mathbf{Q}$, then all magnetic scattering is spin-flip. In the configuration shown in Fig. 1, this scattering appears in the flipper ON channel.
FIG. 1. The schematic on the right represents a triple-axis neutron spectrometer where the monochromating crystal M and the analyser crystal A are also polarizers. \( a_0, a_1, a_2 \) and \( a_3 \) are collimations while S, F, and C denote sample, flipper and detector respectively. As shown, the sample is placed in a horizontal magnetic field (HF) directed along the scattering vector \( q \). The data on the left show magnon cross-sections for \(^{60}\text{Ni}\) obtained by the technique described in the text.

This flipper ON channel also contains the nuclear spin incoherent scattering (NSI) and all of the background. The straightforward way to eliminate these unwanted components and to obtain the magnetic scattering intensity \( I_M \) is to take the difference between the intensity \( I \) for HF and that for a vertical field (VF), both with flipper ON (see Table 1):

\[
I \left( HF - VF \right) = I_M / 2
\]  

(2)

This is the method used extensively by Ziebeck, Brown and their collaborators[4] in a series of pioneering studies of magnetic scattering from 3d metals and compounds. They used, intentionally, very broad energy resolution so that the analyzer essentially integrates over \( \omega \) giving \( S(Q) \) directly. The measured intensities were put on an absolute scale by comparison with powder intensities. Among the magnetic materials which have been studied are Fe, Ni, and MnSi.

Recent work[4,5,6] at Brookhaven has focused on the development of a polarized beam spectrometer with sufficient
TABLE I. SPIN-FLIP AND NON-SPIN-FLIP CROSS-SECTIONS$^a$

<table>
<thead>
<tr>
<th>Spin-Flip</th>
<th>Non-Spin-Flip</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\hat{H}</td>
<td>\hat{Q} (HF)$ $\sigma^{xx}<em>{\text{MAG}} + \sigma^{yy}</em>{\text{MAG}} + \frac{2}{3} \sigma^{\text{NSI}}$</td>
</tr>
<tr>
<td>$\hat{H} \perp \hat{Q} (VF)$ $\sigma^{xx}_{\text{MAG}} + \frac{2}{3} \sigma^{\text{NSI}}$</td>
<td>$\sigma^{zz}<em>{\text{MAG}} + \frac{1}{3} \sigma^{\text{NSI}} + \sigma^{\text{NC}} + \sigma^{z}</em>{\text{NCM}}$</td>
</tr>
</tbody>
</table>

$^a$ This table illustrates how various cross-sections $\sigma$ can be obtained by taking the difference of measured scattered intensities with an applied magnetic field parallel and perpendicular to $\hat{Q}$. $\sigma^{\text{NSI}}$ is the nuclear spin incoherent cross-section, $\sigma^{\text{NCM}}$ is a cross-term involving the product of nuclear coherent amplitudes and the $z$-component of magnetic amplitudes, and $\sigma^{\text{MAG}}$ is the magnetic scattering cross-section.

energy resolution to give direct information about $S(Q,\omega)$. So far we have centered our effort in the neutron energy range between 13 and 130 meV, with Heusler monochromator and analyzer. For future experiments at lower neutron energies, an extensive effort has been made to develop multilayer polarizers[5]. The use of a Drabkin[6] energy-dependent flipper in conjunction with polarizing supermirrors or multilayers makes it possible to decouple beam angular divergence from the energy resolution. Such a flipper has now been built and successfully tested at Brookhaven and is expected to be used routinely in the near future.

So far we have concerned ourselves with the use of polarized neutrons for studies of spin dependent cross sections. However, in 1972 Mezei[7] invented a novel application of a polarized beam, namely, neutron spin echo. This gives an extremely high energy resolution by measuring changes in the modulation of the neutron beam polarization caused by inelastic scattering. Thus polarized beams can be used to perform spin dependent measurements as well as to obtain ultrahigh resolution in non-magnetic inelastic scattering.

II. POLARIZATION ANALYSIS

Fig. 1 depicts our standard triple axis spectrometer converted into one utilizing polarized incident beams as well
FIG. 2. Evidence of antiferromagnetic spin waves (spin-flip scattering) in solid \( \text{O}_2 \) as obtained using a polarized-beam triple-axis spectrometer with polarization analysis. After Stephens et al. /8/.

as polarization analysis. This work was carried out as part of the USA/Japan collaboration on neutron scattering and a few key components, such as the Sm-Co permanent magnets for the analyzer, were supplied by the Japanese collaborators\(^1\). Large Heusler crystals, in the (111) transmission geometry, were kindly provided by the Institut Laue-Langevin\(^2\). Among the many technical problems to be solved is the production of polarized neutron beams with adequate intensity. In fact, this is the principal reason why the powerful technique of polarization analysis has not yet realized its full potential. For the energy range between 13 and 60 meV, we are now down about a factor of 20 in intensity using a fully polarized configuration as compared with our focused pyrolytic graphite triple axis instrument. This has allowed us to study several interesting magnetic systems.

Fig. 2 shows one of our earlier studies on spin waves in solid oxygen\([8]\). The spin-flip scattering is mainly of

---

\(^1\) Y. Ito and H. Yoshizawa of the Institute for Solid State Physics, University of Tokyo.

\(^2\) Courtesy of R. Pynn and A. Freund.
FIG. 3. Measurement of magnetic scattering in Fe using polarization analysis: (b) demonstrates the clear separation of magnetic (ON) and nuclear (OFF) cross-sections while (c) is a plot of I (HF-VF) for flipper ON (see Eq. 2). After Wickstad et al. /9/.
magnetic origin and characteristic of antiferromagnetic spin waves, in their powder averaged configuration. The non-spin-flip scattering is mainly nuclear consisting of Bragg peaks and phonons. As another example, consider the recent study of paramagnetic scattering in pure Fe. Fig. 3(b) demonstrates again the clear separation of magnetic (ON) and nuclear (OFF) cross sections. Fig. 3(c) shows the "difference" data given by Eq.(2), where a magnetic field of 20 Oe was applied parallel and perpendicular to the scattering vector to separate out the magnetic scattering, thereby eliminating the background subtraction problem. This type of measurement sheds light[9] on the puzzle of propagating spin waves above Tc.

Encouraged by these successes we have very recently investigated[10] the weak magnetic scattering from γFe at 1200K. The results shown in Fig. 4 confirm the ferromagnetic correlations reported in recent publications[11]. In addition, our study has provided direct information on the scattering function S(Q,ω) and the unusual Q-dependence of the diffusive line width. There are now a large number of interesting magnetic materials which appear within the technical capability of our spectrometer; these include heavy fermion super conductors, UPt₃ and UBe₁₃, and weak ferromagnets such as MnSi and Ni₃Al.
So far we have given examples of "bulk" magnetic scattering. There is the new and exciting area of study of layered magnets.

III. POLARIZED NEUTRON DIFFRACTION FROM THIN FILM MULTIBILAYERS

Considerable theoretical and experimental efforts to understand the electronic and magnetic structures of surfaces and interfaces have been undertaken in recent years\[12,13\]. Nevertheless, theories describing such fundamental physical phenomena as the critical behavior at the surface or interface of a ferromagnet [14-16] have remained largely untested although a number of measurements of the magnetization at an interface in thin film multibilayer structures by polarized neutron diffraction have been reviewed by Endoh[17].

Polarized neutron scattering is a powerful technique for studying such problems. As an example, we will discuss the investigation of an Fe-Ge thin film multibilayer structure[18]. The multilayers were prepared by radio frequency sputter deposition of alternating thin films of Fe and Ge onto a glass substrate. X-ray diffraction scans with the scattering vector \( \mathbf{Q} \) parallel and then perpendicular to the plane of the substrate reveal that the Ge films are amorphous while the Fe films are "textured", that is an Fe film is made up of microcrystallites each with a [110] direction normal to the plane of the substrate but with orthogonal directions in the plane randomly rotated about the normal [110] axis. The dimension of an Fe microcrystallite parallel to the plane of the substrate is of the order of 100Å. A rocking curve at a fixed scattering angle corresponding to the Fe(110) atomic plane spacing shows that the [110] directions normal to the plane of the substrate have an angular distribution with a full width at half maximum of several degrees. Thus, a diffraction scan as a function of \( |Q| \) and with the direction of \( \mathbf{Q} \) perpendicular to the substrate plane shows structure not only at low scattering angles where the principal diffraction peaks occur at values of \( Q = m \frac{2\pi}{D} \) where \( m \) is an integer and \( D \) is the thickness of a single Fe-Ge bilayer, but also at high angles with peaks at multiples of \( 2\pi/d \) where \( d \) is the Fe(110) plane spacing.

If a beam of polarized neutrons of intensity \( I_{0\pm} \) (where \( \pm \) denotes the spin eigenstate of the beam) is incident on a thin film multibilayer where one of the film materials is ferromagnetic with its magnetization aligned perpendicular
to \( \mathcal{Q} \), then two scattered intensities \( I_{S^+} \) and the corresponding "flipping ratio" \( R \equiv I_{S^+}/I_{S^-} \) can be measured. According to the kinematic theory of diffraction (where extinction is taken to be negligible), if \( \mathcal{Q} \) is perpendicular to the plane of the multilayer films (and fluctuations in bilayer thickness are neglected for now), then

\[
\frac{I_{S^\pm}}{I_0} = \left( \frac{4\pi}{\mathcal{Q}} \right)^2 \left| \sum_{j=1}^{N} \left( f_{Nj}^N \pm f_{Mj}^M \right) e^{iQZ_j} \right|^2 \cdot \left| \sin(M\mathcal{Q}D/2)/\sin(QD/2) \right|^2
\]

where \( f_{Nj}^N \) and \( f_{Mj}^M \) are the nuclear and magnetic scattering length densities, respectively, \( M \) is the number of bilayers and \( Z_j \) is the position of the \( j \)th atomic plane within a bilayer (assuming the bilayer is composed of \( N \) discrete, parallel atomic planes). By measuring the integrated intensities about each \( \mathcal{Q} = m \pi/D \) and/or the diffraction profile about \( \mathcal{Q} = 2\pi/d \) at high angles, it is possible to determine, in principle, the magnetization profile across the thickness of the bilayer. For large enough \( M \), however, extinction effects can become significant, particularly for the first order superlattice peak, and a dynamical theory of diffraction must be applied. For sufficiently small values of \( \mathcal{Q} \) where mirror reflection and refraction effects are important, the neutron is treated as a plane wave incident upon a layered but continuous medium.

Polarized neutron diffraction measurements were made on a number of Fe-Ge multilayer samples. In Fig. 5 the scattered intensity is plotted versus \( \mathcal{Q} \) for a multilayer consisting of 609 thin film bilayers of Fe and Ge. \( \mathcal{Q} \) is perpendicular to the film planes and \( D = 108\text{Å} \). Seven diffraction peaks with periodicity \( \mathcal{Q} = m \pi/D \) are shown beginning with \( m = 1 \). The "ON" ("OFF") data points correspond to incident neutrons in the "+" ("-" ) spin eigenstate. The fact that the flipping ratio \( R \) is not always \( >1 \) implies that the magnetization is not constant across the thickness of the bilayer. Both high- and low-angle polarized neutron diffraction data were fitted to various models for the structure factor. A consistent fit is obtained for a model where each bilayer is composed of three distinct regions; the first of pure amorphous Ge, the second of a random FeGe alloy with the same structure and orientation as the pure bcc Fe which constitutes the third region. An Fe moment in the pure Fe region has the full "bulk" moment while an Fe atom in the FeGe alloy region has a diminished moment depending upon the
FIG. 5. Polarized neutron diffraction scan of a Fe-Ge multi-bilayer with $Q$ perpendicular to the film planes and to the magnetization. Seven diffraction peaks with periodicity $Q = m \pi / D$ are shown beginning with $m = 1$ ($D = 108 \ \AA$). The "ON" ("OFF") data points correspond to incident neutrons in the "++" ("--") spin eigenstate. The fact that the flipping ratio $R$ as defined in the text is not always $> 1$ is indicative of the fact that the magnetization is not constant across the thickness of the bilayer. After Majkrzak et al. /18/.

concentration of Ge about the Fe atom's location in the layer. The Ge concentration is highest next to the pure Ge region and decreases to zero adjacent to the pure Fe region. It was therefore concluded that although a fraction of the Fe atoms have a moment whose magnitude is reduced from that in the bulk, this reduction is due primarily to the interdiffusion of Fe and Ge across the interface of the thin films which occurs during the deposition process. As shown in Ref. 18, the flipping ratios of some of the higher-order superlattice peaks can change by an order of magnitude or more for relatively small changes in the Fe moment in the interfacial region. Thus it is shown that polarized neutron diffraction can be an extremely sensitive technique for measuring such magnetization profiles.

Similar studies are now underway involving multibilayer structures consisting of ferromagnetic and superconducting thin films (e.g. Ni and V or Ni and Nb) of various
thicknesses. The idea is to investigate any interaction between the ferromagnetic and superconducting states which are in spatial proximity to one another.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the contributions of our fellow collaborators at Brookhaven, namely, J. D. Axe, P. Böni, L. Passell, O. Steinsvoll and J. Wicksted. Part of this research was supported by the U.S.-Japan Collaboration in Neutron Scattering. Research at Brookhaven was also supported by the Division of Materials Sciences, U.S. Department of Energy under contract No. DE-AC02-76CH00016.

REFERENCES

Invited Paper

A REVIEW OF CURRENT AND PROPOSED REACTOR UPGRADES

R. M. MOON
Solid State Division, Oak Ridge National Laboratory,
Oak Ridge, Tennessee,
United States of America

Abstract

A REVIEW OF CURRENT AND PROPOSED REACTOR UPGRADES.

In an effort to foresee the future health of neutron scattering, a survey of plans to upgrade reactors and associated experimental facilities was undertaken. The results indicate that we are now entering a period characterized by a substantial reinvestment in reactor sources and expansion in the number of neutron scattering instruments. For the group of institutions participating in this survey there will be a total investment in improved sources and experimental facilities of US $500 x 10^6 to $1000 x 10^6 over the next decade. This investment will result in a 30-40% increase in the total power of research reactors and an increase of 30-50% in the number of neutron scattering instruments. It is therefore reasonable to anticipate an approximate doubling in the number of reactor neutrons incident on samples in the mid-1990s compared to the present.

1. INTRODUCTION

Perhaps the best indication of the health and vitality of neutron scattering in the 1990s is the investment in new or improved sources and instrumentation which will be made in the remainder of the 1980s and early 1990s. Accordingly, a simple questionnaire aimed at discovering current plans for improvement was sent to prominent neutron scattering centers all over the world. Because my charge from the organizers of this conference dealt with reactor upgrades, these inquiries were restricted to reactor-based scattering centers. This paper summarizes the results of the survey.

My personal thanks go to those individuals and institutions who responded to the questionnaire. The response was very good; certainly the centers responding to my inquiry are responsible for more than 75% of the neutron-scattering publications. Therefore, although the results are not comprehensive, we should have no hesitation in drawing some general conclusions regarding the field as a whole. Details of the individual responses are given in the Appendix. Omission of an institution from the list in the Appendix means that either I failed to send a letter to that particular institution or that my correspondent failed to respond.
For this study a broad interpretation of "reactor upgrade" was adopted to include not only improvements in flux but also improvements in the number and quality of neutron scattering instruments and the addition of cold sources and guide halls.

2. GENERAL CONCLUSIONS

The responses gave information on the future plans for thirty-four reactors, most of which were constructed in the late 1950s and early 1960s. If neutron scattering at reactors is to be a healthy, growing field in the 1990s there should now be plans for major upgrades of a significant number of these reactors or for new research reactors. That this is indeed the case is demonstrated in Fig. 1 which shows the number of reactors in our sample which reached, or will reach, criticality in a given calendar year. The burst of activity from 1956 to 1968 is clearly evident, followed by a dry spell in the 1970s. The renewed activity in the 1980s and early 1990s is a greatly encouraging sign. In preparing Fig. 1 a reactor upgrade which would double the flux was counted as a new start or rebirth.

![NUMBER OF RESEARCH REACTORS ACHIEVING CRITICALITY](image)

**FIG. 1.** Number of new reactors per year from 1956 to 1996. An upgrade in which the reactor power is doubled is counted as a new start.
GROWTH OF NEUTRON SCATTERING AT RESEARCH REACTORS

FIG. 2. Growth in total investment, total reactor power, and number of scattering instruments for the institutions participating in the survey.

The questionnaire contained a question concerning funding in which the respondent was to indicate whether the funding was "assured", "probable", or "uncertain". The number of "probables" was so small that they were placed in the "assured" category in the final analysis. It should be recognized that the "uncertain" projects with regard to funding are also uncertain with regard to schedule.

The coming growth is indicated more quantitatively in Fig. 2. The cumulative investment in reactor sources and neutron scattering facilities is shown in Fig. 2a with mid-1984 as a starting point. Over the next decade, this investment will be between US $500 x 10^6 and 1000 x 10^6. A few respondents failed to indicate estimated costs. In these cases I added a reasonable, but unofficial, estimate of my own. The growth in the total power of this sampling of research reactors, shown in Fig. 2b, will be between 30% and 40%. Finally, the increase in the number of neutron scattering instruments, shown in Fig. 2c, will
be between 30% and 50%. Thus, considering the flux increase and the increase in the number of instruments, it is not unreasonable to conclude that there will be an approximate doubling in the number of neutrons incident on samples over the next decade at reactor sources. That is unquestionably a very healthy growth rate.

The importance of cold neutron beams is also evident in the survey results. Fifteen institutions are planning to install cold sources - a tribute to the value of existing cold sources at the ILL\(^1\) and other institutions.

The interest in improving reactor sources and associated neutron scattering facilities is world wide. Of the total investment now being contemplated (including my unofficial estimates for some projects), 41% originates in Asia, 34% in North and South America, and 25% in Europe.

My general conclusions can be stated quite simply. Reactor sources have proven their value for neutron scattering and other types of research. We are now entering a time period during which there will be a major, world-wide reinvestment in reactors and associated research facilities. With these new reactor facilities combined with the anticipated growth in pulsed sources, neutron scattering in the '90s will be a growing, exciting field of scientific research.

3. MAJOR UPGRADES AND NEW FACILITIES

3.1 Asia

The Bhabha Atomic Research Centre is completing a new reactor, DHRUVA, to be commissioned in 1984. This will be a 100 MW reactor with a peak thermal flux of \(1.8 \times 10^{14} \text{ n cm}^{-2}\text{.s}^{-1}\). A hot source (graphite at 1600 K) and a cold source (liquid methane at 110 K) will be installed. Neutron guides (10 cm x 2.5 cm) will transport cold neutrons to a guide hall. For thermal neutrons there will be four tangential beam tubes (10 cm diameter), one through tube and several radial beam tubes.

The National Atomic Energy Agency of Indonesia is building a new 30 MW multipurpose reactor in Serpong. This will be a pool type reactor using 20% enriched fuel with \(\text{H}_2\text{O}\) cooling and moderation and with a Be reflector. It will be used for materials testing, isotope production, neutron scattering, and training.

The Japan Atomic Energy Research Institute, Tokai, will completely rebuild the JRR-3 reactor. It will be operated at 20 MW using 20% enriched fuel and will have a peak thermal flux in the reflector of \(2.7 \times 10^{14} \text{ n cm}^{-2}\text{.s}^{-1}\). Six standard thermal

\(^1\) ILL = Institut Max von Laue — Paul Langevin.
beam tubes (6 cm x 11 cm) and one special radiography tube (3 cm x 3 cm) will deliver neutrons to the cylindrical reactor hall. Three cold guides (2 cm x 12 cm) from a liquid hydrogen source and two thermal guides (2 cm x 20 cm) will enter a spacious guide hall. A wide variety of modern instruments will be operated by both JAERI and university scientists.

3.2 North America

While we in North America have been regrettably slow in building a significant cold neutron capability, this weakness will be rectified if current plans are implemented. In a few years the NRU reactor at Chalk River will get a new vessel equipped with a large through tube. It is planned to install a liquid D$_2$ source at the center of this tube with new cold neutron spectrometers at either end. Brookhaven is planning to make better use of its cold source by constructing a guide hall into which three cold guides and two thermal guides will deliver neutrons. The National Bureau of Standards will install a large D$_2$O cold source and has plans for a guide hall with four cold guides to allow full utilization of this cold source. The proposed instruments for the BNL and NBS guide halls are given in the Appendix. The University of Missouri is planning to increase the power of its reactor from 10 to 21 MW, to add two new instruments, and to improve some existing instruments.

A long-range plan is being developed at Oak Ridge to construct a new reactor with flux of at least $5 \times 10^{15}$ n cm$^{-2}$ s$^{-1}$. Current thinking is that this reactor would have HFIR-type fuel with D$_2$O as coolant, moderator, and reflector. It would be fully equipped for neutron scattering research with a hot source, two cold sources, a large guide hall, and about 30 instruments. This would be a national, and perhaps international, user-oriented facility. The HFIR and the ORR (if still operating) would be shut down when this reactor becomes operational.

3.3 Europe

At the Institut Laue-Langevin the original cold source will be replaced with one of improved design to enhance the cold neutron flux in the existing guides. In addition, a second cold source and guide hall will be constructed, allowing an increased number of cold neutron instruments to be installed. Possibilities for increasing the reactor flux are being considered.

In the Federal Republic of Germany great improvements are now in progress with more in the planning stage. The BER II reactor in Berlin (West) will be reconstructed with a new compact core design, a new Be reflector, a high-pressure gaseous H$_2$ cold source, a guide hall, and a power increase from 5 to 10 MW.
These improvements will result in an order-of-magnitude increase in thermal flux and a greatly enhanced cold neutron capability. At Jülich an improved cold source and new guide hall featuring 58Ni guides will result in much improved cold neutron facilities. Among the instruments to be installed are a diffuse scattering spectrometer, a Bonse-Hart camera for very-small-angle scattering, a backscattering spectrometer, and two SANS units. A major reconstruction of the FRM reactor in Munich is being planned. Calculations based on a very compact core, high-density U3Si/Al dispersion fuel, H2O cooling, and a D2O reflector indicate a peak thermal flux in the reflector of 6-8 x 10^{14} n cm^{-2}.s^{-1} at a power of 20 MW. Such performance would make this reactor an extremely valuable research tool at a relatively modest cost.

The Central Research Institute for Physics in Budapest will reconstruct the WWR-SM reactor. The power will be increased from 5 to 15 MW, there will be a new core with a Be reflector, and a liquid H2 cold source will be installed. Three cold neutron guides and a variety of new instruments will be constructed.

The power of the SAPHIR reactor in Switzerland has been increased from 5 to 10 MW and the vertical divergence of the beam tubes has been doubled. When reconstruction of the instruments to take full advantage of the larger beams is completed, an order-of-magnitude increase in flux on the sample is anticipated.

At Harwell in the United Kingdom studies are in progress aimed at replacing PLUTO and DIDO with a single 50 MW reactor. This would be accomplished by a major upgrade of DIDO.

In the USSR the Kurchatov Institute of Atomic Energy will increase the power of the RR-8 reactor from 8 to 20 MW and new scattering instruments will be installed. The major omission in this survey is the new 100 MW reactor in Leningrad which should now be nearing completion. My unofficial information is that this reactor has H2O cooling, a D2O reflector, fuel geometry similar to that of HFIR, a cold source, a hot source, three through tubes, and six tangential tubes. Anticipated flux levels are 4 x 10^{15} n cm^{-2}.s^{-1} in the flux trap and 1.3 x 10^{15} n cm^{-2}.s^{-1} in the reflector. This reactor will be a major addition to the total world-wide capability for neutron scattering research.

ACKNOWLEDGEMENT

This research was sponsored by the Division of Materials Sciences, U. S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.
APPENDIX: RESPONSES TO REACTOR UPGRADE QUESTIONNAIRE

Country: Austria
Institution: Atominstutit der Österreichischen Universitäten
Reactor: TRIGA Mark-II, 250 kW, critical in 1962
Peak thermal flux: \(1 \times 10^{13} \text{ n.cm}^{-2}.\text{s}^{-1}\)
Present number of scattering instruments: 5
Upgrade plans:
  Reactor: no change
  Instruments: increase to 7
Cost: $60 000 (assured funding)²
It is planned to install a pulsed magnetic field synchronized with the reactor pulse for magnetic structure studies in very high fields, and to install a high resolution small-angle scattering facility based on channel cut perfect crystals to be used for real-time experiments. The completion date is in 1986.

Country: Belgium
Institution: S.C.K./C.E.N. MOL
Reactor: BR 2, 55 MW, critical in 1963
Peak thermal flux: \(5 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}\)
Present number of scattering instruments: 4
In collaboration with the Laboratorie Leon Brillouin a six-chopper time-of-flight spectrometer has been installed at the ORPHEE reactor.

Country: Canada
Institution: Chalk River Nuclear Laboratories
Reactor: NRU, 135 MW, critical in 1957
Peak thermal flux: \(3 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}\)
Present number of scattering instruments: 7
Upgrade plans:
  Reactor: new reactor vessel with cold source in large through tube
  Instruments: increase to 12
Cost: $4 000 000 for reactor upgrade (probable funding)
  $2 500 000 for instruments (uncertain funding)
When the NRU reactor is equipped with its third vessel, a large area (3.5 in. × 8.9 in.)³ through tube will be installed with a liquid D₂ source at its center. Three new spectrometers will be installed at either end of this tube. The earliest completion date is 1990. These plans are in addition to a continuing process of upgrading existing spectrometers.

---
² US dollars used throughout
³ 1 inch = 2.54 cm.
Country: Canada
Institution: McMaster University
Reactor: McMaster University Reactor, 2 MW, critical in 1959
Peak thermal flux: $3 \times 10^{13} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 3 with one more under construction
Upgrade plans: none

Country: China
Institution: Institute of Atomic Energy, Beijing
Reactor: HWRR, 15 MW, critical in 1958, upgraded in 1980
Peak thermal flux: $2.8 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 5
Upgrade plans:
- Reactor: increase power to 17.5 MW (1985)
- Instruments: install cold source and guide hall, increase instruments to 8 (1987)
Cost: unknown
The reactor power increase will be achieved by changing the fuel from $\text{UO}_2$ to metallic U. New instruments include a second powder diffractometer, a SANS unit, and either a polarized-beam SANS machine or a spin-echo spectrometer.

Country: Denmark
Institution: Risø National Laboratory
Reactor: DR-3, 10 MW, critical in 1960
Peak thermal flux: $1.5 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
- Reactor: increase power to 12 MW, flux-trap core, add hot source, reconstruct cold source
- Instruments: increase number to 7-8 including a higher resolution SANS facility
Cost: $1,000,000$ (funding probable)
The estimated completion date is 1988-89.

Country: France
Institution: Laboratoire Leon Brillouin
Reactor: ORPHEE, 14 MW, critical in 1980
Peak thermal flux: $2 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 15
For a description of this new reactor, see B. Farnoux and D. Cribier, Physica 120B (1983) 31. Additions and improvements to the present instruments will be accomplished gradually.

Country: France, Federal Republic of Germany, United Kingdom
Institution: Institut Laue-Langevin
Reactor: HFR, 57 MW, critical in 1971
Peak thermal flux: $1.2 \times 10^{15} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 33
Upgrade plans:
Reactor: some preliminary studies of possible flux increases are in progress.
Instruments: the original cold source will be improved (1985), a new cold source will be added (1986), and about eight additional instruments will be added (1986).
The redesign of the original cold source should produce an increase in cold neutron flux by a factor of about 1.8. New instruments on the second cold source include a triple-axis spectrometer, a high resolution spin-echo spectrometer, a SANS unit for biology and a low Q SANS unit.

Country: Federal Republic of Germany
Institution: Hahn-Heitner Institut, Berlin
Reactor: BER II, 5 MW, critical in 1973
Peak thermal flux: $2 \times 10^{13} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 5
Upgrade plans:
Reactor: increase power to 10 MW ($2 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$), add cold source and guide hall
Instruments: increase to about 11
Cost: $40,000,000 (funding assured)
This project involves redesign of the core, addition of a Be reflector, installation of a cold source, and construction of a guide hall. The project should be completed in 1987. See paper by A. Axman et al. in supplement to Atomkernenergie Kerntechnik 44 (1984) 356.

Country: Federal Republic of Germany
Institution: KFA Jülich
Reactor: FRJ-2, 23 MW, critical in 1962
Peak thermal flux: $2 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
Reactor: new cold source and guide hall
Instruments: increase to 15-18
Cost: $10,000,000 (assured funding)
A new liquid hydrogen cold source is to be installed which will feed three $^{58}\text{Ni}$ neutron guides. Eight to ten new instruments will be installed in a new guide hall. The project should be completed at the end of 1985. See paper in these proceedings by K. Werner et al.

Country: Federal Republic of Germany
Institution: Technische Universität München
Reactor: Forschungsreaktor München (FRM), 4 MW, critical in 1957
Peak thermal flux: $1.5 \times 10^{13} \text{ n.cm}^{-2}\text{s}^{-1}$  
Present number of scattering instruments: 7  
Upgrade plans:  
- Reactor: redesign core, increase power to 20 MW giving flux of about $7 \times 10^{14} \text{ n.cm}^{-2}\text{s}^{-1}$  
- Instruments: increase to about 15  
Cost: $\$70\,000\,000$ (funding is uncertain)  
See paper in these proceedings by K. Böning et al.  

Country: Hungary  
Institution: Central Research Institute for Physics, Budapest  
Reactor: WWR-SM, 4 MW, critical in 1959  
Peak thermal flux: $9 \times 10^{13} \text{ n.cm}^{-2}\text{s}^{-1}$  
Present number of scattering instruments: 5  
Upgrade plans:  
- Reactor: increase power to 15-20 MW giving flux of $2.5 \times 10^{14} \text{ n.cm}^{-2}\text{s}^{-1}$ (1988)  
- Instruments: a liquid H$_2$ cold source and five instruments will be installed  
Cost: $\$13\,000\,000$ (assured funding)  

Country: India  
Institution: Bhabha Atomic Research Centre  
Reactor: CIRUS, 40 MW, critical in 1960  
Peak thermal flux: $6 \times 10^{13} \text{ n.cm}^{-2}\text{s}^{-1}$  
Present number of scattering instruments: 8  
Upgrade plans:  
- Reactor: no change  
- Instruments: improve quality of existing instruments by updating data acquisition systems, adding polarization analysis capability to a diffractometer, and adding a position-sensitive detector to a powder unit.  

Country: India  
Institution: Bhabha Atomic Research Centre  
Reactor: DHRUVA, 100 MW, critical in 1984  
Peak thermal flux: $1.8 \times 10^{14} \text{ n.cm}^{-2}\text{s}^{-1}$  
Instruments: hot source, cold source, 9 instruments  
The following instruments will be installed at this new reactor: a medium resolution inelastic spectrometer, a polarization analysis spectrometer, a four-circle diffractometer, a Be filter spectrometer, a polarized neutron diffractometer, a high-resolution quasielastic spectrometer, a profile analysis diffractometer, a SANS unit, and an interferometer.  

---  

4 Böning, K., et al., IAEA-CN-46/53, these Proceedings.
Country: Indonesia
Institution: Research Centre for Nuclear Techniques
Reactor: TRIGA-Mark II, 1 MW, critical in 1964 (upgraded from 250 kW in 1971)
Peak thermal flux: $5 \times 10^{13}$ n.cm$^{-2}$s$^{-1}$
Present number of scattering instruments: 2
Upgrade plans:
  Reactor: no change
  Instruments: increase to 4
Cost: $50,000 (assured, but more is desired)
It is planned to construct a twin diffractometer (assured), a SANS unit, and a polarized beam diffractometer. The estimated completion date is 1985.

Country: Indonesia
Institution: National Atomic Energy Agency
Reactor: Multipurpose reactor, 30 MW, critical in 1987
Peak thermal flux: $2 \times 10^{14}$ n.cm$^{-2}$s$^{-1}$
Instruments: will probably have a cold source and several scattering instruments.

Country: Israel
Institution: Nuclear Research Centre-Negev
Reactor: IRR-2, 26 MW, critical in 1964
Peak thermal flux: $5 \times 10^{13}$ n.cm$^{-2}$s$^{-1}$
Present number of scattering instruments: 3
Upgrade plans:
  Reactor: no change
  Instruments: improve quality through use of multidetectors and curved monochromators

Country: Japan
Institution: Japan Atomic Energy Research Institute
Reactor: JRR-3, 10 MW, critical in 1962 (now shut down)
Upgrade plans:
  Reactor: complete redesign, 20 MW, $2.7 \times 10^{14}$ n.cm$^{-2}$s$^{-1}$
  Instruments: cold source, guide hall, about 17 instruments
Cost: $150,000.000 (approximate)
There will be seven beam tubes delivering neutrons to the reactor hall in addition to three cold guides and two thermal guides delivering neutrons to the guide hall. Some instruments will be constructed and operated by JAERI; others by universities.

Country: Japan
Institution: Japan Atomic Energy Research Institute
Reactor: JRR-2, 10 MW, critical in 1960
Peak thermal flux: $1 \times 10^{14}$ n.cm$^{-2}$s$^{-1}$
Present number of scattering instruments: 8
Upgrade plans: this reactor may be shut down when the rebuilt JRR-3 begins operations.
Country: Japan
Institution: Research Reactor Institute, Kyoto University
Reactor: KUR, 5 MW, critical in 1961
Peak thermal flux: $3 \times 10^{13} \text{ n.cm}^{-2}\cdot\text{s}^{-1}$
Present number of scattering instruments: 5
Upgrade plans:
  Reactor: repair and replacement of worn parts with no power increase, and addition of a cold source.
  Instruments: construction of a supermirror neutron guide
Cost: $1,500,000 (assured funding)
Completion near the end of 1985.

Country: The Netherlands
Institution: Interuniversitair Reactor Instituut, Delft
Reactor: Hoger Onderwijs Reactor (HOR), 2 MW, critical in 1963
Peak thermal flux: $2 \times 10^{13} \text{ n.cm}^{-2}\cdot\text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
  Reactor: no change in power, new liquid methane cold source
  Instruments: no change in number of instruments but various improvements are planned.
Cost: $250,000 (assured), $80,000 (probable), $40,000 (uncertain)
The instrumental improvements include adding a third axis to one of the depolarization instruments (1985) and increasing the detector area at one of the rotating crystal spectrometers (1986).

Country: The Netherlands
Institution: Joint Research Centre, Petten
Reactor: High Flux Materials Testing Reactor (HFR), 45 MW, critical in 1961
Peak thermal flux: $3 \times 10^{14} \text{ n.cm}^{-2}\cdot\text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
  Reactor: replacement of reactor vessel has been recently completed (October 1984)
  Instruments: no change in number, but enhanced flux in beam tubes
Cost: $5,000,000
The new reactor vessel has two new beam tubes and the potential for adding a cold source. After replacement of the primary heat exchangers in 1986-87, an increase of power to 60 MW would be possible.

Country: Norway
Institution: Institute for Energy Technology, Kjeller
Reactor: JEEP II, 2 MW, critical in 1966
Peak thermal flux: $3 \times 10^{13} \text{ n.cm}^{-2}\cdot\text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
   Reactor: operation at 3 MW during cold season is planned
   Instruments: no change

Country: Sweden
Institution: Institute of Reactor Physics, Royal Institute of Technology, Stockholm
Reactor: R2, 50 MW, critical in 1961
Peak thermal flux: $4 \times 10^{14}$ n.cm$^{-2}$.s$^{-1}$
Present number of scattering instruments: 5
Upgrade plans:
   Reactor: replacement of reactor vessel, redesign of reflector and beam tube geometry to optimize flux at beam tubes
   Instruments: increase number to 7
   Cost: US$3,000,000
It is expected that the sample flux will be 3 - 5 times greater. The work should be completed in 1985.

Country: Switzerland
Institution: Laboratory for Neutron Scattering, Würenlingen
Reactor: SAPHIR, 5 MW, critical in 1958
Peak thermal flux: $7 \times 10^{13}$ n.cm$^{-2}$.s$^{-1}$
Present number of scattering instruments: 4
Upgrade plans:
   Reactor: increase power to 10 MW
   Instruments: increase number to 5-6
   Cost: $2,000,000 (funding assured)
The work to increase the reactor power (new cooling system and improved control systems) was completed in March of 1984. New beam tubes have been installed with twice the vertical divergence of the old ones. The spectrometers are being rebuilt to take full advantage of the larger beams. This work will be completed in 1985.

Country: United Kingdom
Institution: Atomic Energy Research Establishment, Harwell
Reactor: DIDO, 25 MW, critical in 1956
Peak thermal flux: $1.8 \times 10^{14}$ n.cm$^{-2}$.s$^{-1}$
Present number of scattering instruments: 10
Upgrade plans:
   Reactor: increase power to 50 MW
   Instruments: plans not yet developed
Design studies for this upgrade are in progress. Schedule and cost are uncertain.

Country: United Kingdom
Institution: Atomic Energy Research Establishment, Harwell
Reactor: PLUTO, 25 MW, critical in 1957
Peak thermal flux: $2 \times 10^{14} \text{n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 4
Upgrade plans: none, this reactor may be shut down when DIDO is upgraded

Country: United States of America
Institution: Brookhaven National Laboratory
Reactor: High Flux Beam Reactor (HFBR), 60 MW, critical in 1965
Peak thermal flux: $1 \times 10^{15} \text{n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 11
Upgrade plans:
  - Reactor: no change
  - Instruments: it is planned to build a new guide hall and increase the number of instruments to 17
Cost: $29,000,000 (funding is uncertain)
New instruments proposed for the guide hall include a high-resolution powder diffractometer, a backscattering spectrometer, a spin-echo spectrometer, a low-energy time-of-flight spectrometer, a high-resolution SANS unit, and a single-crystal diffractometer. It is hoped to complete this work by 1989-90.

Country: United States of America
Institution: Massachusetts Institute of Technology
Reactor: MIT Research Reactor, 5 MW, critical in 1958
Peak thermal flux: $1 \times 10^{14} \text{n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 6
Upgrade plans:
  - Reactor: no change, reactor was upgraded in 1976
  - Instruments: no change

Country: United States of America
Institution: University of Missouri
Reactor: University of Missouri Research Reactor (MURR), 10 MW, critical in 1966
Peak thermal flux: $1.2 \times 10^{14} \text{n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 7
Upgrade plans:
  - Reactor: power increase to 21 MW
  - Instruments: increase to 9
Cost: $5,000,000 (funding uncertain)
The reactor upgrade involves redesign of the fuel and modification of the reflector. Instrument improvements include a new high-resolution powder diffractometer, a liquid and amorphous material diffractometer, a modification of the SANS unit to reach higher $Q$, and an interferometer modification to increase sensitivity. The estimated completion date is 1988.

Country: United States of America
Institution: National Bureau of Standards
Reactor: NBSR, 10 MW, critical in 1967
Peak thermal flux: $2 \times 10^{14} \text{ n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 9

Upgrade plans:
  Reactor: power increase to 20 MW (1984), cold source installation.
  Instruments: new guide hall and increase in number of instruments to 21-24
Cost: $26\,000\,000$ (funding uncertain)

A solid D$_2$O cold source will be installed during 1985-1987. The guide hall and new instruments should be completed by the end of 1988. Proposed instruments include a high resolution SANS unit, an extended range SANS unit, a low-energy time-of-flight spectrometer, a high-intensity time-of-flight spectrometer, a backscattering spectrometer, a cold neutron triple-axis spectrometer, a polarized-beam triple-axis spectrometer, and a neutron spin echo spectrometer.

Country: United States of America
Institution: Oak Ridge National Laboratory
Reactor: HFIR, 100 MW, critical in 1965
Peak thermal flux: $1.2 \times 10^{15} \text{ n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 9
Upgrade plans:
  Reactor: to be replaced by $5 \times 10^{15}$ reactor (1995).
  Instruments: hot source, cold source, guide hall, about 30 instruments.
Cost: $280\,000\,000$ (uncertain funding)

Country: United States of America
Institution: Oak Ridge National Laboratory
Reactor: ORR, 30 MW, critical in 1958
Peak thermal flux: $3 \times 10^{14} \text{ n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 4
Upgrade plans: none

Country: United States of America
Institution: University of Rhode Island
Reactor: RIAECR, 2 MW, critical in 1964
Peak thermal flux: $1.4 \times 10^{13} \text{ n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 3
Upgrade plans:
  Reactor: no change
  Instruments: a solid D$_2$O cold source to feed a SANS unit and an ultra-cold neutron facility is planned.
Cost: $150\,000$ (funding uncertain)

Country: USSR
Institution: Kurchatov Institute of Atomic Energy, Moscow
Reactor: RR-8, 8 MW, upgraded in 1981
Peak thermal flux: $2.5 \times 10^{14} \text{ n.cm}^{-2} \text{s}^{-1}$
Present number of scattering instruments: 4
Upgrade plans:
  Reactor: increase power to 20 MW
  Instruments: increase number to 6
Cost and schedule unknown

Country: Venezuela
Institution: Instituto Venezolano de Investigaciones Científicas (IVIC)
Reactor: RV-1, 3 MW, critical in 1960
Peak thermal flux: $3 \times 10^{13} \text{n.cm}^{-2}.\text{s}^{-1}$
Present number of scattering instruments: 2
Upgrade plans:
  Reactor: increase power to 5-10 MW
  Instruments: increase number to 3
  Cost: $18,000,000 (funding probable)
A new SANS unit is planned in addition to improvements on an existing triple-axis spectrometer. The work should be completed by 1990.
Invited Paper

SPECIAL NEUTRON SOURCES

P. AGERON
Institut Max von Laue - Paul Langevin,
Grenoble

Abstract

SPECIAL NEUTRON SOURCES.
In some specific energy ranges of interest the neutron intensities are so low
in conventional research reactors that they cannot be used for experiments which
would be possible only at these energies. Special devices have been developed in
order to enhance locally the intensity of such neutrons. They include, for hot
neutrons, moderators of limited sizes at high temperature (1500 to 2000K) and for
cold, very cold, and ultra-cold neutrons, moderators also of limited size at a low
temperature (around 20K) which increase the brightness of the source in the reactor
(flux per unit of solid angle), and neutron guides with total reflection, which
increase the solid angle of collection. Thus, combining the two, a large increase
of the total flux available to the user can be obtained. This paper summarizes some
attempts to optimize such moderators - size, shape, temperature, nature of
material - while taking into account the technical constraints (nuclear heating,
material strength, radiation damage, safety consideration, etc.). Despite this
optimization the net experimental results are far below the theoretical value which
can be expected, especially in the field of UCNs, which are very sensitive to tiny
defects. The fact that neutron lifetime in a material bottle has been increased
considerably allows one to expect similar improvements in the production and
guidance of UCNs.

1. INTRODUCTION

In a room-temperature moderator, after complete thermalization,
neutrons have an important intensity only in a narrow range of
energy: FWHM between 0.005 eV and 0.07 eV. Neutrons with an energy
between 0.1 and 0.5 eV (T \sim 1500 to 2000K) called 'hot neutrons',
or neutrons with energy lower than 0.005 eV to 0.001 or lower,
called 'cold neutrons' (T \sim 20K), are not special but very rare.
What are special are the devices enhancing the intensity of such
rare neutrons, which are moderators with a temperature higher than
room temperature for the hot neutrons and lower for the cold ones.
Owing to the finite size, the imperfection, and the absorption of
this moderator, the enhancement must be lower than the theoretical
limit (for instance \( \frac{T_{room}}{T_{cold}} \)).

Special mention must be made of the so-called ultra-cold
neutrons with energy lower than 1.8 \times 10^{-7} eV, velocity lower
than 6 m/s, wavelength higher than 670 Å, which have the unique
property of being contained by walls with a potential of
2 \times 10^{-7} eV (such as nickel or beryllium), elevation of 180 cm,
magnetic field gradient from 0 to 3 T. The lifetime in such a bottle
from 10-20 s in the early stages, has been improved by a glow-
discharge-cleaned surface, (∼100 s) by coating with a low absorber
Re at a low temperature (250 s), and by covering the walls with
a film of non-hydrogeneous oil which eliminates gaps (600 s) which
is close to the ultimate lifetime of neutrons by beta decay. Such
UCNs are unique in experiments which require a long observation
time (Electric Dipole Moment or neutron lifetime measurements).
Owing to their long wavelengths they are also used in neutron optics
experiments and, owing to their small energy, in high-energy
resolution inelastic scattering experiments. Between cold neutrons
(up to 15-20 Å) and UCN (λ > 600 Å) there exists a domain of
wavelengths called VCN, which is still unexplored although they
show promise for neutron optics, diffraction, surface studies etc.,
and as an intermediate source of UCN.

An effort has to be made for cold, very cold (VCN), and ultra-
cold neutrons (UCN), not only regarding the brightness of the source
(flux per unit of solid angle) but also regarding the efficiency of
the collection up to the point of use by improved neutron guides.

2. HOT NEUTRON SOURCES

Physical aspects

In a large D2O reflector at room temperature, surrounding
a fission (HFR, ORPHEE) or a spallation source (SIN, SNO), hot
neutrons (E > 0.2 eV) are still slowing down and have a spectrum
dφ/dE (E) = φepi/E where φepi is independent of energy. Figure 1
gives the measured spectrum in HFR at 25 cm from the core and an
estimate at a distance close to the core. Figure 2 shows also that
the ratio φth/φepi is about 7 near the core and increases
rapidly with distance while the thermal flux remains almost
constant /1/. This means that a moderator at high temperature which
enhanced the hot flux by a factor higher than 7 will give higher
fluxes of hot neutrons than their direct extraction, with the
additional advantage of a much lower parasitical flux of fast
neutrons, which is also illustrated in Fig. 2 and Ref. /1/.

This does not apply to a small moderator /2/ close to strongly
pulsed neutron sources where a smaller φth to φepi (between 1
to 5), depending on the size, the nature and the decoupling of the
moderator, and where a high peak to medium flux, higher than 100
with a pulse width of about 1 μs, makes them definitely superior
to steady-state hot moderators in TOF experiments.

In LIDO Harwell, Carter /3/ has investigated experimentally,
at a position where φth/φepi is about 25, different possible
hot moderators Be, BeO and graphite, with the same diameter of 15 cm,
but with varying thicknesses. The gain factors in hot neutrons of
Be and BeO were better by 1.3 than those with graphite at a
temperature between 800 and 1000°C. Beryllium with a melting point
of 1280°C is ruled out when higher temperatures are desired. BeO
has been used at DIDO Harwell /4/ but, owing to problems of
radiation damage, toxicity and poor thermal conductivity, graphite, with its well-known technology at high temperature, has been preferred for FR2 Karlsruhe, HFR Grenoble, and ORPHEE Saclay.

Abel /5/ has investigated the effect of size and temperature of a hot graphite block. The gain for a diameter of 11 cm to 20 cm is only 15%.

The effect of temperature between 1500 and 2000K was also investigated at SILOETTE Grenoble /6,7/. The spectrum measurement has shown that the gains for all hot neutron energy increases between 300 and 1500K, but only the gains for $E > 0.2$, 0.3 eV continue to increase between 1500 and 2000K. The choice of the
FIG. 2. Distance to the core versus (a) thermal, epithermal, fast fluxes; (b) Nuclear heating of H$_2$ by fast neutrons of D$_2$O by core and tank γ-rays at ILL.

Graphite temperature is thus determined by the desired hot neutron energy. At ORPHEE/8/ graphite temperature can be adjusted by varying the helium-nitrogen composition of the gas between the graphite and the container (between 900°C (pure helium) and 1650°C (pure nitrogen)). See Fig. 3.

Technical and safety aspects

Strong nuclear heating of the graphite is desirable, and thus a hot source can be placed at the maximum of thermal flux. At HFR it is about 0.55 W/g of which 0.5 is due to core gamma rays, 0.14 to capture gamma rays in the materials of the source and only 0.013 to fast neutrons (1.3 x 10$^{-14}$ $\phi_{\text{fast}}$). The insulation is made of graphite wool or multiple sheets of graphite or molybdenum. It can be made sufficient to ensure 1500K, even with poor nuclear
heating (0.1 W/g at FR2). More important is the thermal conductivity of the material to ensure a rather uniform temperature in the moderator and heat flux at its boundary heating (1.2 W/g at the inner side, 0.7 at the outer side at HFR).

A hot source causes two safety problems. (i) When hot, a large amount of heat is stored (50 MJ at HFR, 10 MJ at ORPHEE), and a sudden contact with water can cause a steam explosion. (ii) The large amount of void in the insulation, if suddenly flooded by D$_2$O, will cause a positive step of reactivity up to 0.5% $\Delta k/k$, which is not acceptable for the kinetics of HFR. To prevent this, the safety philosophy is to have a double container with an inert gas pressure in between, higher than the pressure of the surrounding water, and a still higher one around the graphite. These pressures are continuously monitored to raise the alarm if they rise or decrease.
The heat flux of about 10 W/cm² is evacuated from the inner to the outer container via a gas gap of about 1 mm thick; so the temperature of the inner wall is, under normal conditions, about 400-600°C; should a local failure of the insulation occur, the temperature rises above tolerable limits, and the thermocouples control this temperature. Nothing like that has occurred during the 13 years of operation of the HFR hot source, which can be considered both safe and reliable.

3. COLD NEUTRON SOURCES

Models for the energy transfer cross-section

The most widely used model for energy transfer is that of Young and Koppel /9,10/, who considered free molecules for their translational movements. For ortho- and para-hydrogen it agrees reasonably with the experimental cross-section value, and more sophisticated models /11/ do not result in further improvements.

For deuterium, taking into account the delayed diffusion and the intermolecular forces /12/, it gives a better agreement with the total experimental cross-section which gives better prediction for the production in the VCN and UCN ranges /13/.

Thermalization calculation

To calculate the space- and energy-dependent flux two main types of code can be used: (i) Monte Carlo codes (for example, TRIPOLI), which track individual neutrons from their source to regions or boundaries of interest, taking into account at each collision the differential cross-section for energy and direction change. They can easily use anisotropic cross-sections, work in three dimensions and be time-dependent. For reasonable computing times the 'stories' have to be short, i.e. the absorption and the leakage of the system should not be too low. They are therefore well suited to small hydrogenous cold sources near a spallation target, but much less to a deuterium source in a large D₂O reflector. (ii) Codes which solve, by iterations, the Boltzmann equation for several groups of energy (e.g. 15 groups from 0.1 to 200 meV) in one (e.g. THERMOS) or two dimensions (e.g. NET). The convergency of multigroup two-dimensional calculations is sometimes difficult. It may be useful, for rough parametric calculations, to represent \( \Phi(E,r) \) /14/.

\[
\Phi(E,r) = \Phi_1(r) S_1(E) + \Phi_2(r) S_2(E)
\]

where \( \Phi_2 \) is a cold flux, with an asymptotic spectrum \( S_2 \) in the cold moderator, and \( \Phi_1 \) a thermal flux with an asymptotic spectrum \( S_1 \) in the room-temperature moderator, the cross-section for transfer from one group to the other being determined by adjustments on a multigroup one-dimensional calculation.
For cold sources in a large $D_2O$ reflector, in order to limit the number of mesh points and to remain in two-dimensions, one restricts the volume of the moderator around the cold source to a cylinder in R,Z geometry, with the same Z axis for this volume for the cold source and for its extraction beam tube. Fictitious thermal neutron sources and albedo at the boundaries are then determined in order to obtain along the axis of the volume, without cold moderator or beam tube, the same thermal flux distribution as in the actual unperturbed reflector. In the case of the ILL reactor, Fig. 4 indicates the geometries for (a) the large $D_2O$...
source and its radial horizontal beam tube (the influence of the vertical service tube is neglected), and (b) for the small D\textsubscript{2} (or H\textsubscript{2}) source in a tangential horizontal beam tube (the transverse gradient is neglected).

In spite of these numerous approximations the overall transmitted cold flux spectrum, including losses in guide tubes, is in good agreement with the experimental one /15/ - see Fig. 5.

Hydrogen sources

Most of the cold sources at present in operation are discs of approximately 15 - 20 cm diameter and 5 - 7 cm thick inside radial or tangential beam tubes in D\textsubscript{2}O reflectors. Their measured gain factors as a function of the neutron wavelengths are defined as the ratio, at the beam tube exit, of the fluxes with or without hydrogen in the cell, except for the supercritical hydrogen source of DR3 Ris\o placed in a through-tube and compared to a H\textsubscript{2}O diffusor. The actual ortho/para compositions are not known. The calculated gains for pure para- and half-para/half-ortho hydrogen for 18 cm diameter and 5 cm thickness (with 20% void), frame the
different measured values, which are rather similar and not sensitive to differences in flux gradient and in material or void perturbations (Fig. 6).

Deuterium sources (Fig. 7)

The only flux measurements on deuterium cold sources are those made at SILLOETTE-Grenoble (cylinders of 18 and 38 cm diameter /16/) and at ILL-HFR (38 cm diameter sphere). The gain factor was defined by comparing the flux with $n_2O$ at the place of the source. Thus, for a beam tube (radial) in the direction of a flux gradient across the source, the gain factor depends also on this gradient. This explains, at least partly, the very different gains for similar source diameters, at HFR (70 for 10 Å) and at SILLOETTE (170) where the flux gradient was 2 to 2.5 higher. On the other hand, for a
tangential beam tube in the direction of which there is a negligible flux gradient across the source, the gain is more specific and depends only on the size of the source; 90 (at 10 Å) and 40 for diameters of 38 and 18 cm, respectively. These experimental gains are higher than the gains given by calculations in the SILOETTE geometry - 33 (18 cm), or by calculations reported in Ref. /17/ 36 (20 cm), 45 (30 cm) and 54 (40 cm).
Effect of a cavity in a large D₂ source

Experiments in SILUETTE with a tube (10 cm diameter), penetrating the centre of the large 38 cm cold D₂ source have shown an increase of the cold flux by a factor of up to 2.1 for a radial beam and 1.6 for a tangential one. A two-energy-group calculation on the HFR geometry (radial tube), for a 16 cm diameter cavity with different penetrations, shows that this increase is the maximum for a penetration equal to about 2/3 of the diameter of the source /18/.

An improved version of the first cold source in HFR, to be installed in 1985, has a cavity filled with D₂ gas, 20 cm high, 10 cm wide, and 25 cm depth of penetration in the 38 cm diameter D₂ sphere. It also includes a vertical guide inside the service tube for extraction of VCN and UCN. The expected increase of the cold flux as a function of the neutron wavelength has been calculated by TRIPOLI in the real geometry and by DOT in a simplified R,Z geometry (Fig. 3) and is given in Fig. 4B.

Comparison of D₂ with H₂ for a second cold source at HFR

For a limited diameter of 21 cm, an ortho/para H₂ source, with an optimum thickness of 5 cm, is compared to a D₂ source with an optimum thickness between 17 and 21 cm (for both of them a 20% vapour content is assumed). It appeared that the radial distribution of the brightness for cold neutrons at the boundary facing the neutron exit is rather uniform for H₂ but not for D₂. As it is mainly the central region of the source which illuminates the neutron guides with a 6 x 12 cm² section, one has to compare the gains at the centre and not the mean gain over the whole source. Calculated gains are similar for normal H₂ and D₂, whereas experimental results for similar dimensions indicate, at a long wavelength, a higher gain for D₂ (40) than for H₂ (20-30). A calculation with D₂ containing 10% H₂ indicates the possibility of further improvements.

Cold source associated with a pulsed source

For a steady-state (SIN) or modulated source (SNQ) with large pulses (5-10 ms), where the cold flux follows the flux in the moderator (at least, if not too far from the target), the previous optimization of a cold source is valid. For sources with very short proton pulses the cold moderator, with a thickness of 5 cm and a section of between 10 x 10 to 25 x 25 cm², which acts both as a moderator and a thermalizer, has to be placed very near the target. Qualitatively, a thicker moderator increases the total number of cold neutrons per pulse, but also increases their pulse width. As reported in Ref. /19/, for a given thickness of the moderator, the pulse width increases with the cold neutron wavelength and is about 100 μs at 5 A.

In sources with low nuclear heating, solid methane at 20K is considered the best cold moderator. When not possible, liquid
parahydrogen is a good substitute. Grooves in the cold moderator (1 cm thick, about 5 cm deep) in a pulsed source, are reported /20/ to enhance the cold flux by a factor of 2, whereas they seem to have no sensitive effect in a steady-state source.

Nuclear heating.

The nuclear heating of cold moderators is partly due to 'parasitic' radiations which do not contribute to the production of cold neutrons. For fission neutron sources they are: γ-rays from fission and fission products for a density \( S \) of sources of γ at \( r \), the nuclear heating at \( r_0 \) is

\[
W(r_0) = \int \int dE \int dr^3 1.6 \times 10^{-13} E \frac{\mu a(E)}{\rho} n(E) \frac{S}{4\pi (r_0-r)^2} \exp(-\sum \chi(E)t)
\]

where \( n(E) \) is the number of photons per disintegration in an interval \( dE \) around \( E \), \( \mu a/E \) is the coefficient of absorption in energy, and \( \chi \) the total cross-section in the different thicknesses \( t \) of materials between \( r \) and \( r_0 \). The γ nuclear heating is given for HFR as a function of the distance in Fig. 2.

Fast neutrons with a mean energy of 2 MeV (2.5 fission neutrons per fission):

\[
W(r_0) = \int \int dE 1.6 \times 10^{-13} \frac{\sum \chi(E)}{\rho} \frac{d\phi}{dE} (E, r_0)
\]

The fast neutron nuclear heating is given for HFR at a distance \( r \) from the core in Fig. 2. For hydrogen the fast neutron heating, in W/g, is roughly proportional to the fast flux (above 1 MeV) = 5 \times 10^{-13} \( \phi \); 3 times less for deuterium and 14 times less for heavy water.

For spallation neutron sources, if the target is \( ^{238}\text{U} \), about half the neutrons are fast fission neutrons and their nuclear heating can be treated as for pure fission sources. The remaining neutrons in \( ^{238}\text{U} \) and the total neutrons in the non-fissile target (lead, tungsten, etc.) are pure spallation neutrons with much higher energy than for fission neutrons (mean energy 25 MeV instead of 2 MeV); but their effect in nuclear heating, according to Reichardt /17/ is only higher by a factor of 2.5. Comparatively speaking, nuclear heating due to spallation γ-rays is negligible.

Calorimetric measurements made at SIN at 10 cm, in \( \text{D}_2\text{O} /21/ \), with a lead target receiving 600 MeV protons, give 3.6 W/g in \( \text{H}_2 \) and 1.9 in \( \text{D}_2 \) per mA of proton current, which produces a maximum thermal flux of 10\( 14 \cdot\text{cm}^{-2}\cdot\text{s}^{-1} \). A direct comparison, in view of the different geometry, is difficult but it seems that, chiefly for a cold source placed close to a target, the neutron heating per useful cold neutron is higher for spallation sources than for fission sources with an optimized position.
Nuclear heating produced by useful cold or thermal neutrons is thus proportional to the thermal flux. The main radiations produced by neutron capture are (i) prompt or delayed $\gamma$-rays - their nuclear heating depends on the nature and the quantity of structural materials. For optimized structures (1.5-2 mm of aluminium for the cryogenic cell, 5 mm for the zirconium vacuum vessel), the capture $\gamma$-ray heating is about $0.1 - 0.2 \times 10^{-15} \Phi_{th}$ W/g.

(ii) $\beta$-decay in the chain $^{27}\text{Al}(n,\gamma)\, ^{28}\text{Al},\, ^{28}\text{Al} \beta \rightarrow ^{28}\text{Si}$ - about $10^{-15} \Phi_{th}$ W/g, (iii) $\alpha$-decay in the chain

$$^{58}\text{Ni} (n\gamma)^{59}\text{Ni} \rightarrow ^{60}\text{Ni},\, \sigma_1 = 4.2 \text{ b},\, \alpha_1 = 79 \text{ b}$$

where $E = 4.75 \text{ MeV}$

$$^{56}\text{Fe} (n\alpha) \rightarrow \alpha_2 = 13 \text{ b}$$

after a time 

$$\ln \frac{\sigma_1 + \sigma_2 - \sigma_0}{\sigma_0} = \frac{3.5 \times 10^{12}}{\phi}$$

about 1 year at $\phi = 10^{15}$ W/g.

The nuclear heating due to (mean energy 4.75 MeV):

2.75 x $10^{-15} \phi$ for natural Ni;
0.27 x $10^{-15} \phi$ for stainless steel with 10% Ni.

Heat removal

The heat load generated in a cold source can be removed in three different ways: (i) By gaseous helium at low temperature, which cools the moderator cell through a heat exchanger (tubes at HFR-Brookhaven, or ribs at Reactor-Leningrad), directly welded or brazed to the cell; the moderator has to be at a higher temperature than the cell in order to transfer its heat to the cell. The heat needed to overcome the pressure drop in the forced circulation is simply supplied by the compressors working at room temperature in the helium circuit. This simple process has two drawbacks - it assumes that the thermal conductivity of the materials in the high flux is high and does not decrease during irradiation and it brings additional material, thus nuclear heating. (ii) By a forced circulation of the moderator itself in single-phase flow - supercritical hydrogen at a pressure of about 15 bars for a temperature of 30-50K. This implies (a) a circulator (or fan) for hydrogen at low temperature; (b) a thicker cell (6-8 mm Al) due to the 15 bar pressure instead of a thin (1.5-2 mm Al) for a pressure of 1.5 bar (cold) to 3-4 bar (hot), thus additional nuclear heating. (iii) By natural circulation of a boiling moderator. It is the simplest system if the service tubes can be vertical (one brings the liquid and the other takes out the gas in which case gravity rapidly separates out any droplets. Experience has shown that the heat removal possibilities of such thermosystems are enormous ($\approx 250$ kW), but that a small power ($\approx 300$ W) transferred
from the superheated outgoing gas to the incoming liquid may induce bubbles in it and cause a flow reversal. If this problem is solved by correct insulation of the return line, the free liquid level can always be adjusted to ensure a good filling of the useful cell volume.

More difficult is the natural convection in a partly horizontal tube where, with a two-phase flow in the return line, a possible overheating of the upper dome of the cell can occur. Such a system works well in existing sources (DIDO, EL3-Saclay, FRJ Jülich, etc.), but with a total power of 0.1-1 kW. The extrapolation to a power of 3 kW (2 in cell 1 in the liquid D$_2$) has necessitated a mock-up with liquid D$_2$ and with electrical heating on the wall, specially made with a low-thermal-conductivity stainless steel. The main conclusion of these experiments has been very encouraging /22/.

(1) The flow at nominal condition, although a strongly two-phase one (5/6 of the mass flow-rate was liquid in the return line) was stable without chugging or reversal.

(2) The total mass content of liquid in the cell (which is essential for neutronic performances), measured by γ-ray attenuation and a capacity probe, decreases at 0.8-0.85 at full power.

(3) The hot-spot temperature of the cell, when the cell becomes empty of liquid, owing to a total lack of refrigeration, reaches 150K only after 7 minutes, which allows sufficient time to stop the reactor or to restart the refrigerator.

It has to be pointed out that this satisfactory operation has been obtained with a mean heat density of 0.4 W/cm$^2$ in the liquid and a mean heat flux from the wall of 0.9 W/cm$^2$. Such a source placed in a 3-6 times higher flux, owing to a three times more powerful source, would cause a new problem, in particular if the critical heat flux for nucleate boiling (5 W/cm$^2$ for D$_2$) is overcome. A limit for natural connection operation could be reached.

From the safety aspects, 12 years of operation of the HFR cold source show satisfactory behaviour under a relatively low fast flux, but a high thermal flux (0.5%, Si build-up per year of operation with $\delta_{ph} = 10^{15}$). Higher, fast and thermal flux may lead to a shorter lifetime. The basic safety philosophy is that in the case of a failure in the cryogenic cell, the container vessel could withstand the following: The thermal shock and the overpressure due to the sudden flooding of the beam tube by the cold moderator; the detonation of a stoichiometric mixture at room temperature and pressure of D$_2$ (or H$_2$) with air; the sudden disappearance of the cryogenic fluid. This should not cause an excessively positive reactivity step (in the case of a reactor deuterium is in this respect better than hydrogen).

4. UCN AND VCN SOURCES

General remarks /23,24/

The fraction of the UCN flux to the total flux with a Maxwellian distribution at a temperature T,
$f = \frac{E_{\text{max}}}{2(\frac{kT}{h})^2}$

is very small at the moderator temperatures which can be achieved in a reactor: $2.5 \times 10^{-11}$ at $T = 300\text{K}$ and $2.5 \times 10^{-9}$ at $30\text{K}$ for neutron energies lower than $E = 1.8 \times 10^{-7} \text{eV}$ ($T = 2 \times 10^{-3}\text{K}$, $v = 6 \text{m/s}$); but the absolute UCN flux, for a total flux of $10^{14} - 10^{15} \text{cm}^{-2}\cdot\text{s}^{-1}$, is not negligible. One cannot calculate directly this flux by numerical integral or Monte Carlo codes. But as soon as the main part of the spectrum $d\phi(E)/dE$ is known, one can estimate the small energy $e$ current, $J(e)de$, emerging from the surface of the moderator and the equivalent neutrons density, $\rho(e)de$, by:

$$J(e)de = \frac{p(e)de}{4\pi t(e)}$$

where $p(e)de$ is the production rate $\text{cm}^{-3}\cdot\text{s}^{-1} = \frac{d\Sigma}{de}$

$$\rho(e)de = \frac{p(e)e}{v\Sigma t(e)}$$

$d\Sigma/dE$ is the differential down-scattering cross-section and $\Sigma_t(e)$ is the total cross-section including all causes which prevent a UCN born inside the moderator to reach the boundary - upscattering, capture, elastic scattering on inhomogeneities, etc., and neutron $\beta$-decay. The mean free path $\Sigma_t^{-1}$ can be considered as the useful thickness of the moderator (called 'the converter'), a few millimetres for UCN and $(v\Sigma_t)^{-1}$ as the UCN lifetime in the moderator.

If the incident neutrons have a Maxwellian distribution at the same temperature of the converter (thermal equilibrium), one can apply the principle of detailed balance, and $\rho(e)de$ becomes:

$$\rho(e)de = \frac{\phi \exp\left(-e/kT\right)}{v(kt)^2} \approx \frac{\Sigma_s}{\Sigma_t}$$

where the Boltzmann factor is close to 1, which is the lower tail of the Maxwellian distribution multiplied by the ratio of the upscattering to the total cross-section which is close to 1 for a room-temperature, low absorption material (water, polythene, etc.) but smaller than 1 for low temperature, very inhomogeneous solids (beryllium, graphite, etc.).

The total UCN density can be rewritten as

$$\rho \text{UCN} = \frac{\phi}{2\pi^2 \frac{v}{v_T}} x \frac{4}{3} \pi V_{\text{lim}}^3$$

The product of the phase space density by the volume in velocity space $(V_T = \sqrt{2kT/m})$. This expression still holds if UCNs are obtained by deceleration, assuming that the phase space density is the same at all energy much lower than $kT$ and is at best, unchanged during the deceleration process, if the faster neutrons come from a cold source.
Their phase space density is that of thermal neutrons at the position of the cold source multiplied by the actual gain factor \( g(E) \), at the considered energy, due to the cold source.

Direct extraction of UCN by a horizontal beam tube

The normal walls of beam tubes (around 5 mm aluminium with \( V_{\text{lim}} = 3.2 \text{ m/s} \)), by their nuclear potential and their absorption, constitute a barrier against UCN produced in the moderator, while they are transparent to the rest of the spectrum. This can be overcome by placing a thin converter made of material with an almost zero potential (ice, polythene, zirconium hydride) which restores the original Maxwellian spectrum. A further improvement is obtained if the converter is cooled down (between 70-20K) and more, in principle, if the incident neutrons are already cold ones.

Such UCN sources have been operated in different reactors, in the USSR, Canada, the United Kingdom. The UCNs are guided by electro-polished copper or stainless-steel pipes, with strongly curved sections in order to filter the UCN. The low efficiency of such devices (ratio of the measured current at the exit to the predicted one at the converter) seems to be due to the very large number of reflections, not completely specular, of UCN which are lost when backscattered to the source.

Extraction by vertical or inclined tubes

Deceleration by gravity from 10-15 m/s neutrons to 0-6 m/s UCN, even on modest elevation (6-10 m), is sufficient to permit materials for the converter (\( \text{D}_2\text{O} \) or beryllium) and membranes (Al, Zr) with \( V_{\text{lim}} \) between 3 and 7 m/s. Such a disposition has been used in particular at Garching and at Institut Max von Laue - Paul Langevin (ILL) with a room-temperature \( \text{H}_2\text{O} \) converter /25/, and at the Leningrad Nuclear Physics Institute where the first 3-cm-thick beryllium converter at 30K (the efficiency of which was very sensitive to the inhomogeneities) has been replaced by a 3-cm-thick liquid hydrogen in a zirconium cell, which also acts as a cold source /26/.

In this velocity range, the number of reflections in the exit tube is still high and its transmission low. This last one has been significantly improved (up to 30-50%) only for neutrons with higher velocities (50 to 400 m/s), which are decelerated to UCN velocities by Doppler shift machines, described below.

The total reflection UCN turbine (/27/ and Fig. 8)

Neutrons of 50 m/s at a mean angle of 0.1 rad with the direction of motion of the turbine, are reflected on the hemicylindrical blades (neutrons have a velocity of 25 m/s in the moving frame) which almost reverse their direction. They lose a further 25 m/s when leaving the reflector, and keep only their initial transverse velocities, between 0 and 10 m/s. The efficiency of this mechanical conversion is good (about 50%). The 3 cm width and the divergency, \( \pm 0.1 \) radian of incoming 50 m/s neutron beams, are
enlarged, respectively, to 20 cm and almost $2\pi$, which make possible the simultaneous use of 2-3 UCN beams. This turbine with a copper blade has been in operation at Garching since 1975. An improved version with nickel blades will be installed in 1985 at ILL on a vertical guide from the modified deuterium cold source. Taking into account predictable losses, the UCN current and density at the exit of the turbine will be $6000 \text{cm}^{-2}\text{s}^{-1}$ and $50 \text{cm}^{-3}$, respectively whereas direct extraction of UCN from the cold source through the service guide could yield figures 10 times lower.

Other turbines, with radial deceleration of the neutrons or with supermirror blades /28/ have been operated.

Doppler-shifted Bragg scattering on a pulse neutron source /29/.

A mica-crystal (d spacing $\sim 10 \text{Å}$) with its planes perpendicular to the incident 400 m/s neutrons, moving at an angle of about $30^\circ$ with the axis Z of the beam, at a velocity of 230 m/s, reflects the neutrons in the moving frame at a Bragg angle of about $60^\circ$. When leaving the crystal they are again decelerated to an almost zero velocity. The dimensions of the velocity volume reflected by the crystal are $\Delta V_x$ and $\Delta V_y$ (Fig. 9) comparable to those of the incident beam ($\pm 6 \text{m/s}$ for a nickel guide) but $\Delta V_Z = 1/2 V_{\text{incident}} \tan \beta n_x$ which requires a large mosaic, $n_x$, of several degrees in order to be comparable to $2 V_{\text{lim}} = 10 \text{m/s}$ acceptable by a UCN storage bottle, which is artificially obtained by Al wedges between the micas. In spite of the present efficiency of 10% at the reflection (reflectivity, no coincidence between reflected velocity volume and storable ones etc.) and of 15% at the collection to a bottle, the method is attractive.
FIG. 9. Principle of the Doppler-shifted Bragg conversion. The incident velocity $V_N$ becomes $V_{NR}$ in the moving frame, is reflected by the crystal and leaves the crystal with a velocity almost zero. The volume in the velocity space has a width of about $2V_{lim}$ but a height depending on the mosaic $n_x$ of the crystal.

because phase space density of UCN is related to the peak, not to the average flux of the pulsed incident beam, and the flow of UCN to the bottle is roughly continuous if the period of pulse repetition is about the same as the travel time of UCN through the production volume (around 10 ms). If larger, a phase chopper can help by opening the bottle entrance only during the time of the UCN pulse. The operation of such a device at ZING P1 Argonne gives information of its future possibilities.

Superfluid $^4$He UCN source /30/

Incident neutrons with energy $E_0$ about 1 meV are down-scattered at UCN energy $E$ by exciting a phonon in superfluid helium, with a cross-section according to Refs. /30,31/.

$$\sum (E_0 + E) = 2N \sigma S(k_0) \sqrt{E/E_0}$$

where $N$ is density of He atom $= 0.02 \times 10^{24} \text{ cm}^{-3}$
$\sigma$ is free He atom cross-section $= 1.1 \text{ b}$
$S$ is one phonon structure factor $= 0.1$
The production rate of UCN is then:

\[ p_{\text{UCN}} = \int_{0}^{E_{\text{lim}}} \sum(E_{0} \rightarrow E) \frac{d\phi}{dE_{0}} E_{0} E_{0} \, dE \]  

(2)

where \( \frac{d\phi}{dE_{0}} (E_{0}) = \frac{\phi_{\text{th}}}{\text{in reactor}} \frac{E_{0}}{(kT)^{2}} \times \frac{\Delta \Omega}{4\pi} \times g(E_{0}) \)

\( \frac{\Delta \Omega}{4\pi} \) = solid angle of collection \( \leq 7.5 \times 10^{-5} \) with a nickelled guide, 0.2 if helium placed inside the reactor.

\( g(E_{0}) = \text{gain due to a cold source (20-70)} \)

If the He is inside a UCN bottle, which is transparent to incident neutrons \( E_{0} \), the density of UCN can be built up to

\[ p_{\text{UCN}} = p_{\text{UCN}} \times \tau \]

where \( \tau \) is the lifetime of UCN in the bottle, including the beta decay of neutrons, the upscattering on \(^4\text{He}\) (negligible when its temperature is lower than 0.6K) the capture by \(^3\text{He}\) (negligible after a purification down to a \(^3\text{He}\) content lower than 10^{-11}), the loss reflection on the walls.

Such a source, proposed by Golub and Pendelbury /30/, has been operated at ILL /31,32/ - a polished stainless-steel tube (3 m long, 7 cm dia.) closed at one end by a fixed SS membrane and at the other end by a flap valve, is filled with purified \(^4\text{He}\) cooled down to 0.6K and acts as a guide for the incident neutrons. When the UCN equilibrium is reached the valve opens and the UCNs flow out through the cryogenic membranes to an outer detector or experiment. An overall lifetime of 60 s and a UCN density of 1.5 cm^{-3} have been measured. The losses at the detection owing to the membranes are important; therefore, the actual density in the helium is estimated to be 5-10 times higher; a 'cold exit' which avoids this membrane is under development. On the other hand, the lifetime has been improved up to 250 s (without helium) by coating the bottle with beryllium.

Table I is an attempt to compare the different kinds of existing UCN sources and to point out the causes of losses. A much clearer comparison will be available during 1985 when optimized UCN sources will be put in operation and, it is hoped, measured with more comparable methods - a turbine on the cold source and the improved helium source at ILL, the final version of the \( \text{H}_2 \) UCN source at Leningrad, and a Doppler-shifted UCN source at Los Alamos.

5. CONCLUSIONS

Hot graphite (1500-2000K) is at present the best source of steady-state beams of hot neutrons (0.1-0.5 eV), but will be definitely surpassed by pulsed neutron sources for TOF instruments.
<table>
<thead>
<tr>
<th></th>
<th>Thermal Gain</th>
<th>Theoretical</th>
<th>Measured</th>
<th>Loss factors</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>flux</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>cold source</td>
<td>(cm(^{-2})s(^{-1}))</td>
<td>(\rho) (cm(^{-3}))</td>
<td>(J) (cm(^{-2})s(^{-1}))</td>
</tr>
<tr>
<td>ILL inclined guide H(_2)O converter</td>
<td>(6 \times 10^{14}) x 1</td>
<td>30(^d)</td>
<td>3600(^e)</td>
<td>0.3(^a)</td>
</tr>
<tr>
<td>Leningrad vertical guide H(_2) cold source</td>
<td>(6 \times 10^{13}) x 25 (?)</td>
<td>75(^d)</td>
<td>9000(^f)</td>
<td>1200</td>
</tr>
<tr>
<td>Garching turbine</td>
<td>(1.6 \times 10^{13}) x 1</td>
<td>0.8(^d)</td>
<td>100(^f)</td>
<td>2</td>
</tr>
<tr>
<td>Argonne Doppler shifted on ZING P' Bragg conversion (peak)</td>
<td>(1.2 \times 10^{13}) x 20 (?)</td>
<td>12(^d)</td>
<td>1400(^f)</td>
<td>0.15(^b)</td>
</tr>
<tr>
<td>ILL superfluid helium (\tau = 60s\ \frac{\Delta \Omega}{4\pi} = 7.5 \times 10^{-5})</td>
<td>(2 \times 10^{14}) x 70</td>
<td>660(^e)</td>
<td>3300(^g)</td>
<td>1.5(^a)</td>
</tr>
</tbody>
</table>

- \(a\) Direct measurement
- \(b\) Deduced from current measurement
- \(c\) Guides non-adapted to the useful velocity
- \(d\) Calculated according to Eq. (1)
- \(e\) Calculated according to Eq. (2): \(\rho_{UCN} = \rho_{UCN} \times \tau\)
- \(f\) \(J = \rho \frac{\nu}{4}\)
- \(g\) Calculated according to Eq. (2): \(J(UCN) \times \rho_{UCN} \times \frac{Volume}{exit\ section}\)
For steady-state production of cold neutrons, deuterium sources in the 20-40 cm range are capable of a higher performance, especially at long wavelengths, than the well-established hydrogen sources, but involve more problems of geometry and location in a reactor. Nuclear heating and radiation damage in reactors, or in sources with higher mean powers, will cause severe technical problems; pulsed cold sources are advantageous for TOF experiments but to a lesser extent than for hot neutron sources (lower peak to mean ratio, longer pulse widths, about 100 µs).

For UCN and VCN sources, the very large margin between potential possibilities and present results offers great possibilities of improvements.

REFERENCES

/14/ AGERON, P., LIVOLANT, M., et al., Cryogenics 9 2 (1967) 42.
/16/ HARRIG, H.D., Etude de source de neutrons froids à hydrogène et deuterium liquides, Thèse à la Faculté de Grenoble (1967).

*Review papers on production and use of UCN*
Invited Paper

VERY-HIGH-FLUX RESEARCH REACTORS BASED ON PARTICLE FUELS

J. R. POWELL, H. TAKAHASHI
Department of Nuclear Energy,
Brookhaven National Laboratory,
Upton, New York,
United States of America

Presented by M. Blume

Abstract

VERY-HIGH-FLUX RESEARCH REACTORS BASED ON PARTICLE FUELS.

A new approach to high-flux research reactors is described, the VHFR (Very-High-Flux Reactor). The VHFR fuel region(s) are packed beds of HTGR-type fuel particles through which coolant (e.g., D_2O) flows directly. The small particle diameter (typically of the order of 500 microns) results in very large surface areas for heat transfer (~100 cm^2/cm^3 of bed), high power densities (~10 megawatts per liter), and minimal ΔT between fuel and coolant (~10K). VHFR designs are presented which achieve steady-state fluxes of ~2x10^{16} n/cm^2·s·1. Deuterium/beryllium combinations give the highest flux levels. Critical mass is low, ~2 kg ^235U for 20% enriched fuel. Refuelling can be carried out continuously on-line, or in a batch process with a short daily shutdown. Fission product inventory is very low, ~100 to 300 grams, depending on design.

We have carried out scoping studies of very-high-flux research reactors based on direct cooling of HTGR particle fuel. Thermal fluxes of 2 to 3x10^{16} n/cm^2·s can be generated in the central core of the reactor, with fluxes of ~10^{16} n/cm^2·s in the outer reflector.

The fuel particles are similar to the TRISO particles presently used in HTGR's. They consist of small kernels containing fissile UO_2, which are coated with several layers of pyrographite and silicon carbide. Particles can be fabricated to whatever diameter is desired. For VHFR's, diameters in the range of 400 to 500 microns appear optimum.

Unlike HTGR's, where the fuel particles are imbedded in large graphite blocks or balls, in the VHFR, the particles are held in packed beds and directly exposed to coolant. The large surface area for heat transfer (~100 cm^2/cm^3 of bed) allows high-power densities (e.g., 10 MW(th)/liter). By using efficient moderators such as Be, D_2O, or H_2O, critical volume is
FIG. 1. Annular fuel bed design for particle bed very-high-flux reactor.

FIG. 2. Thermal flux distributions for Be/D$_2$O VHFR as a function of $^{235}$U loading.
minimized. The combination of small size and high-power density results in very high flux levels.

Figure 1 illustrates one type of VHFR. The fuel particles are packed into an annular bed between two porous frits, with coolant flowing radially through the bed. In the design shown, the coolant is D_2O, Be metal is used for the inner moderator, and a combination of Be and D_2O for the outer moderator.

This design yields the highest flux levels of the various coolant/moderator combinations studied (D_2O and H_2O coolants/Be, H_2O, D_2O, and C moderators). Figure 2 shows thermal flux vs radial position with various 235U loadings, for a bed power density of 10 MW(th)/liter. Fluxes of 2 to 3x10^{16} n/cm^2/s are achieved. Other coolant/moderator combinations result in lower fluxes, typically by a factor of 2.

Figure 3 shows thermal flux distributions for the other VHFR coolant/moderators considered. As in Figure 2, the i.d. of the annular fuel bed is 40 cm, with a radial thickness of 5 cm. Core loading is constant at 5 kg 235U. The insert Table in Figure 3 shows k_{\infty} for the six cases examined. (For clarity, Cases 1 to 3 are vertically displaced from Cases 4 to 6.)

The choice of inner moderator strongly affects peak flux. Deuterium-oxide (Case 1) or carbon (Case 6) moderators yield much lower fluxes than beryllium (Cases 2, 4, and 5). The inner moderator need not be all Be, but can be a ring of Be next to the fuel bed, with a central D_2O channel. Comparing Cases 2 and 4, one sees that the difference in flux is relatively small. The combination of an H_2O ring and D_2O central channel also yields a high flux.

Neutronic analyses have been carried out with a 1-D, 15-group (9 thermal, 6 fast) ANISN model, using ENDFB-V cross sections. Based on previous analyses of gas-cooled particle bed reactors [1], the actual k_{eff} for an operating VHFR with 2-D end effects and poisoning (Xe, Sm, etc.) will be lower by ~0.20 than the 1-D value. This, together with fuel depletion effects, makes the critical mass for the VHFR in Figure 1 equal to ~1.5 kg 235U.

The VHFR can operate on 20% enriched fuel with only a small penalty in reactivity. Figure 2 compares k_{\infty} values for 20% enriched fuel (values in brackets) with those for fully enriched fuel at the same 235U loadings. The k_{\infty} values for partially enriched fuel are slightly lower; however, flux magnitudes and profiles are virtually identical with those for enriched fuel.

The ability to operate with partially enriched fuel is very important, as it greatly simplifies fuel safeguards requirements. Heat transfer performance is not affected, since the fuel particle kernels can easily accommodate the additional 238U.
FIG. 3. Thermal flux distribution for VHFRs with various coolants/moderators ($^{235}$U loading = 5 kg).
At a nominal value of 10 MW(th)/liter reactor power is 350 MW(th) for a 40-cm (i.d.) core with L/D=1. Reactivity burnup limits (~20% of initial $^{235}\text{U}$) are reached after one day of operation. The spent fuel particles can be hydraulically unloaded from the annular bed by a $\text{D}_2\text{O}$ side stream, and fresh fuel particles loaded. The process can be carried out daily on a batch basis during a short shutdown, or continuously during reactor operation.

Figure 4 illustrates one approach for hydraulically unloading and loading VHFR cores. Fuel transfer lines are symmetrically arranged azimuthally around the annular fuel bed. Inlet lines enter at the top and continue to the bottom of the bed, where they inject fresh fuel particles. Outlet lines collect spent fuel particles from the top of the bed. Typically, 6 inlet and 6 outlet transfer lines, each 1 cm in diameter, would be used. The inlet lines would be headered into a common inlet line, (not shown) above the core, as would the outlet lines.

Typically, particle volume flow rates for continuous fueling would be ~1 mm/s in the transfer lines. The reactor frits, solid moderators, end reflectors, and transfer lines would be a single integral unit that could be replaced through the top of
the D$_2$O coolant vessel when radiation damage limits were reached.

Transfer lines external to the core would be lined with a strong neutron absorber (e.g., gadolinium) to limit heat generation in the internal fuel. The inlet lines in the annular fuel bed would be porous, and the in fuel particles would be cooled by the existing radial coolant flow through the bed.

In contrast to HTGR particles, the fissile UO$_2$ in the kernels of the VHFR fuel particles must be diluted by some inert, low-cross-section material (e.g., C or Al$_2$O$_3$) to ensure that all particles effectively share the heat load. Typically, $^{235}$UO$_2$ concentration in the kernel will be only about 5 atom%, for kernels occupying 10% of the particle volume. With partially enriched fuel (e.g., 20% $^{235}$U), the corresponding total UO$_2$ concentration in the kernel would be higher, i.e., 25%. Because of the very small amount of $^{235}$U in the kernel, fractional volume burnup and gaseous fission product pressure are over two orders of magnitude smaller than in present HTGR particles. This, together with the low operating temperature (~100°C) and short-exposure time (~1 day), make the chances of fission product release from VHFR particles much smaller than from HTGR particles, which are already very low.

Other fuel bed geometries than that shown in Figure 1 can be devised. One attractive possibility is to split the annular fuel bed into two halves, axially separated by ~10 cm. Radially oriented beam tubes are then placed between the two halves of the core to increase the available experimental volume at high flux.

Another possibility is to form the reactor core by arranging a number of small diameter annular packed bed fuel elements (each about 10 cm o.d.) in a ring. The fuel elements can then be hydraulically unloaded and loaded on an individual basis.

Experiments [2] have demonstrated the long-term (2200 hours) compatibility of the particles with high-velocity water. Heat transfer experiments have demonstrated capability for 3 MW(th)/liter operation at coolant pressures of 1 atm. Higher pressures are expected to allow power densities well in excess of 10 MW(th)/liter. The flow geometry in Figure 1 allows operation up to ~5 MW(th)/liter at acceptable pressure drops (5 atm) for 5-cm-thick beds. Higher power densities can be achieved with various types of split-flow or folded-flow geometries. The cylindrical frits holding the packed particle bed would be made of wound zircaloy or Al wire, partially sintered to provide a very strong, low-pressure-drop structure.

At 5 MW(th)/liter, VHFR fluxes would be half those shown in Figures 2 and 3, and power levels would be reduced to 175 MW(th). This still allows very high flux performance, while it
reduces the power and fuel consumption rate and results in a lower, more conservative pressure drop and power density. More detailed study is needed to determine optimum VHFR parameters.

The VHFR has a number of attractive features important to safety.

1. Very low fission product inventory. The continuous throughput of fuel keeps fission product inventory down to the equivalent of a few hours of operation.

2. Fuel removal capability. In the event of a shutdown (planned or unplanned), the core fuel can be unloaded in a few seconds to a separately cooled holding vessel.

3. Excellent cooling capability. Upon shutdown, the VHFR core can be cooled by natural circulation if the coolant pumps fail to function and the fuel is not unloaded.

4. Good control. VHFR reactivity is very sensitive to neutron absorption in the moderators surrounding the annular fuel bed. Control rods (or drums) placed in these regions can provide both routine control and shutdown capability. Also, poison can be quickly injected into the coolant circuit for emergency shutdown.

In summary, the VHFR appears to be a very attractive approach for very-high-flux research reactors. More detailed design studies are needed to better define and optimize the concept including, for example, reduction in power and/or size.

REFERENCES


UPGRADING OF THE MUNICH REACTOR WITH A COMPACT CORE*

K. BÖNING, W. GLÄSER, J. MEIER, G. RAU, 
A. RÖHRMOSE, L. ZHANG**
Fakultät für Physik, Technische Universität München, 
Garching, Federal Republic of Germany

Abstract

UPGRADING OF THE MUNICH REACTOR WITH A COMPACT CORE.

An extremely small reactor core has been proposed for the project of substantial modernization of the FRM research reactor at Munich. According to the present status this 'compact core' will be a cylinder with a diameter of about 20 cm and 70 cm high. The new high-density $\text{U}_3\text{Si/AI}$ dispersion fuel of about 45% enrichment is contained in 20 concentric fuel plate rings. The compact core is surrounded by a large heavy-water tank which will incorporate the user installations (beam tubes and irradiation channels). However, the primary cooling circuit will contain light water which is not only more economic but also essential for the performance of the small core. An important optimization potential to decrease easily the power density peaks in the core is to reduce further the enrichment in those fuel plate rings where the neutron flux is particularly high. Two-dimensional neutron transport calculations show that such a core, containing about 7.5 kg $^{235}\text{U}$, should have an effective multiplication factor of about 1.22 and an unperturbed but realistic maximum thermal neutron flux in the heavy water tank of 7 to $8 \times 10^{14}$ cm$^{-2}$s$^{-1}$ at 20 MW reactor power.

CONCEPT

The FRM research reactor of the Technical University of Munich became critical in October 1957 as the first reactor in the Federal Republic of Germany. It is a conventional swimming pool reactor which is at present operated at a thermal power of 4 MW. It is used for beam tube and neutron irradiation applications.

To increase considerably both the flux and the volume where these thermal neutrons can be used, and also to achieve very pure thermal neutron spectra, we plan to modernize and upgrade the FRM substantially. Our project is to design an advanced, extremely small reactor core (giving rise to a high source density of fast neutrons per MW), to surround this core by a large volume of D$_2$O, the best available moderator (so that the thermalized neutrons have a...
particularly long lifetime), and to increase the reactor power up to about 20 MW. It is felt that this 'compact core' design will lead to a particularly high ratio of maximum thermal neutron flux $\phi_{\text{th}}^{\text{max}}$ (outside the core) to reactor power $P$, i.e. to a particularly high 'quality factor' $\phi_{\text{th}}^{\text{max}}/P$. It makes use of the new fuels with extremely high uranium densities developed within the international reduced enrichment programmes.

A preliminary, general design of the cylindrical compact core is shown in Fig. 1/1,2/. This somewhat earlier version, called 'compact core 2', has an active volume of $V = 15.9$ litres as defined by inner and outer radii of $R_1 = 3.47$ cm and $R_0 = 9.8$ cm, respectively, and $H = 60$ cm height. The 20 concentric fuel plate rings are fastened to six aluminium supports and are characterized by fuel and Al-cladding thicknesses of 0.51 and 0.38 mm, respectively, with 1.8 mm cooling channels in between. Assuming the fuel to consist of the new $U_3\text{Si}/\text{Al}$ dispersion fuel with a uranium density of 7.0 g/cm$^3$ and 45% enrichment, this core contains 6.99 kg $^{235}\text{U}$. At the power design value of $P = 20$ MW the average power density in the core, $P/V \approx 1.3$ MW/litre, is about that of the Grenoble (HFR-ILL) and Oak Ridge (HFIR) reactors. A more than 250 cm long, rigid zircaloy tube of about 10 cm inner radius and 10 mm thick surrounds the core and separates its $H_2O$ primary cooling circuit from the $D_2O$ moderator tank which has both diameter and height of 250 cm. The user installations (tangential beam tubes and
TABLE I. RESULTS OF THE TWO-DIMENSIONAL NEUTRONIC TRANSPORT CALCULATIONS WITH THE DOT 3.5 COMPUTER CODE
(For explanations see text).

<table>
<thead>
<tr>
<th></th>
<th>U235 mass (kg)</th>
<th>k$_{\text{eff}}$</th>
<th>$\phi_{\text{th}}^{\text{max}}$ (cm$^{-2}$s$^{-1}$)</th>
<th>maximum power density (MW/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) compact core 2</td>
<td>6.99</td>
<td>1.180</td>
<td>$7.5\times10^{14}$</td>
<td>3.74</td>
</tr>
<tr>
<td>b) &quot; &quot; with control rod</td>
<td>0.998</td>
<td>9.2$\times10^{14}$</td>
<td>4.54</td>
<td></td>
</tr>
<tr>
<td>c) compact core 3</td>
<td>8.15</td>
<td>1.249</td>
<td>$6.2\times10^{14}$</td>
<td>3.02</td>
</tr>
<tr>
<td>d) &quot; &quot; with control rod</td>
<td>1.095</td>
<td>7.4$\times10^{14}$</td>
<td>3.48</td>
<td></td>
</tr>
<tr>
<td>e) &quot; &quot; at operation temp.</td>
<td>1.235</td>
<td>6.3$\times10^{14}$</td>
<td>2.95</td>
<td></td>
</tr>
<tr>
<td>f) &quot; &quot; with enrichm.var.</td>
<td>7.48</td>
<td>1.219</td>
<td>$6.6\times10^{14}$</td>
<td>1.93</td>
</tr>
</tbody>
</table>

vertical irradiation channels) and the reactor shutdown rods will be incorporated in this D$_2$O tank, whereas the reactor control rod will be placed in the central H$_2$O channel of the core /1,2/.

CALCULATIONS

Two-dimensional neutron transport calculations have been performed using the DOT 3.5 computer code. The core and moderator/reflector configuration was assumed as described above, with the only modification that no installations were considered in the D$_2$O tank, because of which our results always represent unperturbed values.

Table I (a) shows the results for the effective multiplication factor k$_{\text{eff}}$, the maximum thermal neutron flux $\phi_{\text{th}}^{\text{max}}$ in the D$_2$O tank at P = 20 MW reactor power, and the corresponding maximum local power density (dP/dV)$_{\text{th}}^{\text{max}}$ of the cold, clean 'compact core 2'. Independent test calculations have been performed by the INTERATOM company using DOT 4.2 and Monte Carlo codes and their results are consistent with ours (after correction for different temperature assumptions) in the sense that k$_{\text{eff}}$ is up to 0.7% and $\phi_{\text{th}}^{\text{max}}$ is 3 or 8% smaller there /3/.

In another calculation we assumed a 3 mm thick AgInCd control rod to be located in the central H$_2$O channel of the compact core. Its outer radius was taken to be equal to the inner core radius R$_i$ and its height to be 6 cm greater on each side than the active core height H (in exactly the same way as the fuel plate rings of
the core were also always extended by 6 \, \text{cm} dummy Al-rings). The results are given in Table 1 (b). The corresponding change in reactivity, $\Delta \rho = 15.5\%$, should be large enough to control the reactor. The thermal neutron flux in the D2O tank is significantly larger now, since the central control rod 'pushes' the fission density rate more to the outer core regions.

Which unperturbed value of $k_{\text{eff}}$ will be required for an effective operation of the reactor under realistic conditions? A first burnup calculation for the compact core 2, which was performed by the INTERATOM company /3/, showed that the loss of reactivity, due to both fuel depletion and long-living fission products, is $\Delta \rho \approx 6.4\%$ assuming a reactor cycle length of 44 days as at the HFR Grenoble. Such a relatively low value of reactivity change was expected since it is a direct consequence of the large uranium mass in the small compact core. The saturation effect of xenon and samarium poisoning is $\Delta \rho = 5.4\%$ /3/. In making appropriate estimates for the other contributions to the reactivity requirement (e.g. $\Delta \rho = 3\%$) for the user installations in the D2O tank), we concluded that an unperturbed value of $k_{\text{eff}} = 1.22$ should be sufficient for effective operation of the reactor under realistic conditions /4/.

There are essentially two ways to increase the multiplication factor $k_{\text{eff}}$ of the compact core 2 up to the required value of about 1.22: to increase slightly the active volume and the average $\text{H}_2\text{O}/^{235}\text{U}$ ratio of the core. The main result is a slight relative increase of that number of neutrons which are moderated within the core volume (nevertheless the compact core remains highly undermoderated) /4/. So a modified design version, called 'compact core 3', has been established, which has a somewhat larger (gross) active volume of $V = 20.3$ litres as defined by inner and outer radii of $R_i = 3.14$ cm and $R_o = 10.1$ cm, respectively, and $H = 70$ cm height. The total mass of 8.15 kg $^{235}\text{U}$ is again contained in 20 concentric fuel plate rings which are fastened to 6 aluminium supports, and also the type ($\text{U}_3\text{Si}/\text{Al}$, etc.) and thickness (0.51 mm) of the fuel is as before. However, the Al-cladding thickness was reduced to 0.33 mm as suggested by the NUKEM company /5/ and, in particular, the $\text{H}_2\text{O}$ cooling channel thickness has been increased to 2.2 mm. At $P = 20$ MW the nominal average power density in the core is now $P/W = 1.0$ MW/litre.

The results of our DOT 3.5 calculations for the 'compact core 3' are shown in Table I (c). The multiplication factor $k_{\text{eff}}$ is now sufficiently large, but the maximum thermal neutron flux $\phi_{\text{th}}$ in the D2O tank at $P = 20$ MW reactor power is somewhat smaller than before because of the reduced fission density rate in the larger core volume /4/. A neutron history evaluation shows that, out of 100 fission neutrons, net numbers of 39 fast and 2 epithermal neutrons stream out of the core into the D2O tank of which, after thermalization, 16.5 diffuse back into the core. It is also very consistent with our discussion of the compact core behaviour that to replace all the $\text{H}_2\text{O}$ in the primary cooling circuit by $\text{D}_2\text{O}$ would dramatically reduce $k_{\text{eff}}$ to 1.054, since the stopping power of $\text{D}_2\text{O}$ is much smaller than that of $\text{H}_2\text{O}$.

The insertion of a central AgInCd control rod, defined in exactly the same way as above (Table I (b)), yields the results of
The corresponding decrease of reactivity, $\Delta \rho = 11.2\%$, seems not quite sufficient to control the reactor but the difference can presumably be easily compensated for by mixing some burnable poison (e.g. boron) with the $\text{H}_3\text{Si/Al}$ fuel.

In all calculations discussed so far the temperature was assumed to be $T = 293.6\text{K}$. Table I (e) shows the results obtained making reasonable assumptions for the operating temperatures (fuel $423.6\text{K}$, Al-cladding and $\text{H}_2\text{O} 335.6\text{K}$). The corresponding decrease in reactivity is $\Delta \rho = 0.9\%$.

A very important optimization potential of the compact core design is the possibility of varying the uranium enrichment (or density) from ring to ring, i.e. to smooth the power density distribution $\frac{dP}{dV}$ in the core by simply reducing the $^{239}\text{U}$ enrichment in those fuel plate rings where the neutron flux is particularly high. In Table I(f) the results of a calculation are presented in which the following enrichment profile was assumed for the 20 rings: 20, 33, 16 x 45, 33, and 20%. Obviously the value of $k_{\text{eff}}$ is smaller now because of the reduced mass of $^{239}\text{U}$, but still just about large enough, and the maximum thermal flux $\varphi_{\text{th}}$ is larger. But most important is the significant drop in the maximum local power density $(\frac{dP}{dV})_{\text{max}}$ from 3.02 MW/litre down to 1.93 MW/litre, which strongly relaxes the cooling requirements for the compact core.

In conclusion, if we realistically include the influence of both the central control rod and burnable poison or, alternatively and quite equivalently, that of burnup, we expect that a maximum unperturbed thermal neutron flux $\varphi_{\text{th}}$ can be achieved in the $\text{D}_2\text{O}$ tank at 20 MW reactor power. It seems very possible that this concept would also allow the compact core reactor to be operated at a power of somewhat more than 20 MW - with a proportional increase of $\varphi_{\text{th}}$.

Further design work is in progress.

REFERENCES


/5/ HROVAT, M., KOCH, K.H., NUKEM GmbH, private communication.
SINQ - A PROJECT FOR A CONTINUOUS SPALLATION NEUTRON SOURCE

F. ATCHISON, W. E. FISCHER, M. PEPIN, Y. TAKEDA,
Ch. TSCHALAER
SIN, Swiss Institute for Nuclear Research,
Villingen, Switzerland

Abstract

SINQ - A PROJECT FOR A CONTINUOUS SPALLATION NEUTRON SOURCE.

SINQ is a continuous spallation neutron source which is planned to be attached to the existing accelerator facility at SIN after its upgrading to higher currents. The overall project aims to provide the Swiss neutron user community with the facilities necessary to carry out scattering experiments in the wavelength range 1 to 10 Å; that is, the neutron source itself, experimental areas, sample conditioning apparatus, sample preparation areas and scattering instruments. Construction is planned to start in 1987, with the first neutrons expected in early 1990. Total costs (excluding spectrometers) are estimated to be 26.5 x 10^6 SFr. This report describes the general design concept and the expected performance of the neutron source.

1. INTRODUCTION

The Swiss Institute for Nuclear Research (SIN) operates a cyclotron system that produces a high-intensity proton beam of 592 MeV. The beam intensity at present available is about 150 μA, and mainly used for meson production at two special target stations. The secondary meson beams are used for nuclear and particle physics. This research forms the main activity for SIN. Some condensed matter physics is done at a muon beam.

After having passed through the meson target, about 75% of the power of the original primary proton beam is still left. This beam is at present stopped in a beam dump.

A new injector system is now being commissioned to allow routine operation of the SIN experimental programme with a proton current in excess of 1 mA. With such beam current the construction of a spallation neutron source with a performance comparable to that of a medium-flux reactor becomes feasible. Owing to the absence of any macro-pulse structure in the extracted proton beam this neutron source will be of continuous type.

The layout of the accelerator system and experimental facilities, following completion of the upgrade programme, and including SINQ, is shown in Fig. 1. The major components of SINQ are (i) an extension of the present proton channel, (ii) the neutron source itself, (iii) the building for the source and neutron beam-tube experimental area, (iv) the neutron guide hall, and (v) the scattering instruments.
FIG. 1. Layout of the accelerator system and the experimental facilities at SIN. The location of the neutron hall and the guide hall is shown.
2. THE GENERAL DESIGN CONCEPT FOR SINO

The source is to be mounted on an extension of the present proton channel beyond target 'E' in a new building on the east side of the experimental hall. This building is to contain the source itself and the experimental area for instruments requiring beam tubes. The non-interacting part of the proton beam will be collected beyond target 'E', deflected downwards to pass under the beam-tube area, hence to the centre of the source and finally pitched vertically upwards to the spallation target. This is illustrated in Fig. 2.

The proton energy at the spallation target will range from 560 to 590 MeV (depending on the thickness of target 'E') with an initial intensity of 1 mA.

A vertical section through the central region of the source is shown in Fig. 3. The target consists of a 'pot' of Pb/Bi eutectic mixture (molten) of inner diameter about 15 cm and 2 to 4 m high. Most of the beam power is deposited in the bottom 30 cm of the target. Natural convection is to be used to transport this heat to an exchanger mounted at the top of the target and hence away from the source /1/. The 'active' region of the target (the bottom 30 cm) is located at the centre of a tank of D₂O with a diameter and height of about 2 m. The D₂O thermalizes the fast neutrons resulting from high-energy proton interactions with the Pb/Bi in the target.
Radiation heating in the vicinity of the target by the neutron and gamma fields has been calorimetrically measured at the Thermal Neutron Facility at TRIUMF (Vancouver) for a similar geometry /2/.

Thermal neutrons are extracted from the $^{1}$H$_{2}$O with beam tubes; neutrons entering through the end window drift along the tube either to a monochromator or to the start of a neutron guide bundle. The wavelength range down a particular beam tube may be selected by choosing the temperature of the moderator at the beam-tube window. Particular emphasis is being placed on the provision of 'cold' neutrons at SINQ, allowing neutron scattering at wavelengths >3Å (a region not available at a domestic facility in Switzerland).
FIG. 4. Layout for the beam tubes and guides in the moderator tank. Version A: No irradiation station.

FIG. 5. Layout for the beam tubes and guides in the moderator tank. Version B: With irradiation station.
Selected beam tubes will view cold sources which consist of flasks of liquid hydrogen or deuterium at 20K. There is also interest in the provision of short wavelength neutrons, and a hot source (1000 to 2000K) is also allowed for in the planning.

The fast neutron production is also accompanied by high-energy particles (mainly neutrons). The D₂O tank is surrounded by a biological shield consisting of about 5 m of iron and concrete. The beam tubes and neutron guides pass through this shield to the outside of the source at about 6 m radius.

The annular region between 6 and 14 m from the centre of the source is to be used for mounting neutron scattering instruments that require beam tubes. Beam tubes with thermal, cold and (possibly) hot neutrons are planned. These beam tubes will, in general, require monochromators and auxiliary external shielding (both for high-energy background and thermal neutron dumps behind the experiments).

A neutron guide hall is to be built on the south side of the source/beam-tube building. A beam tube viewing a cold source will be terminated after about 1.5 m and the neutrons will then enter a guide-bundle; these will transport neutrons to scattering instruments in the guide-hall and up to about 50 m from the centre of the source.

Two layouts for the beam tubes in the moderator tank are shown in Figs 4 and 5. The second version reduces the number of beam-tube station is to provide a facility to irradiate material under various conditions with a hard neutron spectrum in order to simulate 'first wall conditions' in fusion reactors. Investigation of the performance of such a facility is in progress; the choice between the two versions will depend on these results.

3. EXPECTED PERFORMANCE

The design and performance of SINO are based on results from an experimental programme for the investigation of the neutronics. This programme has been realized by a Jülich-Karlsruhe-SIN collaboration on a secondary proton beam at SIN with a one-to-one mock-up model of the source /3/. One of the flux maps, relevant to the planned SIN source, is shown together with its comparison with a Monte-Carlo simulation in Fig. 6. By means of flux distributions such as these, the optimal position of the cold source as well as the position and size of the beam tubes may be determined.

The last experiment at the mock-up model examined several configurations of cold hydrogen sources and their spectra. For the following we assume an incident proton beam of 1.7 mA on to the spallation target (~1 mW of beam power). For a thermal beam tube with a tip surface of 175 cm² in the D₂O moderator and a collimator of 4 x 8 cm² along the last 4 m of the tube we obtain a 25 meV neutron flux at the monochromator of 1.3 to 1.7 x 10⁹ n/(cm²*sec*meV) value depends on the radial distance to the monochromator, which may be different for the various types of spectrometer.
The surface of the cold source has a flux of $1.2 \times 10^{14}$ n/(cm$^2$·s). Neutrons in the energy region 15 to 5 meV (2.3 to 4.0 Å) are better extracted through beam tubes. The corresponding fluxes profit from the gain factor of the cold source, which varies in this region from 1 to 5. Neutrons with a wavelength >4 Å will be extracted through guides. We plan to install guides with a nickel-58 surface; a flux gain with respect to guides with natural nickel can be expected.

REFERENCES


INSTRUMENTS AND METHODS

(Session VI)
Chairman

H. F. BERTAUD

France
NUCLEAR POLARIZATION AND NEUTRONS

H. GLÄTTLI
Service de Physique du Solide et de Résonance Magnétique,
CEN-Saclay,
Gif-sur-Yvette, France

Abstract

NUCLEAR POLARIZATION AND NEUTRONS.

Different possibilities for the use of polarized nuclei in thermal neutron scattering on condensed matter are reviewed. Highly polarized nuclei are the starting point for studying dipolar magnetic order. Systematic measurement of spin-dependent scattering lengths is possible on samples with polarized nuclei. Highly polarized hydrogen should help to unravel complicated structures in chemistry and biology. The use of polarized proton targets as an energy-independent neutron polarizer in the thermal and epithermal region should be considered afresh.

Scattering of neutrons with spin $\vec{S}$ on nuclei (spin $\vec{I}$) is mainly due to the strong interaction. It is spin dependent, and for thermal (s-wave) neutrons it can be described by two scattering lengths, $b^+$ and $b^-$, according to the two possible spin channels $J_\pm = I_\pm 1/2$. In most applications of neutron scattering, the nuclear spins are oriented at random and the spin dependence gives rise to incoherent scattering in much the same way as the isotopic incoherence, but this spin incoherence is modified as soon as some spin correlation occurs. This is in general due to nuclear polarization but it can also arise from spin symmetry requirements in a few molecules ($H_2$, $D_2$, $CH_4$) or molecular groups ($CH_3$) with widely spaced rotational states, i.e. where low moments of inertia are combined with low hindering potentials against molecular rotation. Two applications will be mentioned where the spin dependence manifests itself in a more distinctive way than just as incoherent scattering.

Possibly the most beautiful experiment, where the spin dependence has been put to its full use, was the observation of the long-range order of nuclear moments due to their dipolar interaction in a single crystal of LiH /1/. The advantage of studying nuclear dipolar order is obvious. The interaction leading to the magnetic order is the dipole-dipole interaction between the nuclear moments, situated at fixed and well-known positions. The Hamiltonian is a priori perfectly known. The price to pay for this clean model system, unequalled in electronic magnetism, may appear too high, and has in fact been found prohibitive for all but very few research groups studying magnetism.

Owing to the weakness of the nuclear magnetic moments, the transition temperatures $T_m$ are down in the nano-kelvin region. One way to overcome this difficulty in insulators is routinely applied in Saclay. Dynamic polarization of the nuclei, thermally decoupled from the lattice in high magnetic fields, can lead to effective
spin temperatures close to the mK and the subsequent demagnetization, generally but not necessarily performed in the rotating frame, brings it below $T_m$.

For Van Vleck ions, the enhanced (so-called pseudo-nuclear) moments can raise $T_m$ into the mK range. Pseudo-nuclear moments can be highly anisotropic, yielding new possibilities for nuclear ordering. Neutron diffraction studies have been reported on the ordered states of $\text{HoVO}_4/2$. For completeness it should be mentioned that nuclear magnetic order has also been reached in copper by means of a double stage of nuclear demagnetization /3/, but no neutron diffraction experiment has been reported so far.

The relevant parameter in neutron diffraction on polarized nuclei is the spin-dependent scattering length $b_N = (2/2l+1) (b^+-b^-)$. Up to quite recently $b_N$ was unknown for most nuclei. To measure $b_N$, a specific apparatus has been built in Saclay. It is based on the fact that the neutron spin precesses inside a sample around the so-called pseudo-magnetic field $H^*$ set up by the polarized nuclei /4,5/. The precession angle of the neutrons in a polarized beam can be measured as a function of the pseudo-magnetic field in a set-up analogous to the Ramsey molecular beam experiment /5,6/. Our results, combined with precise cross-section measurements, scattered data from different experiments like Bragg diffraction on polarized samples and diffuse scattering, allowed the determination of a large number of $b_N$'s. Today, $b_N$ is known with reasonable accuracy for $\sim 50$ nuclei. An estimate of its magnitude can be given for another $\sim 25$ nuclei. Recent compilations of neutron scattering lengths have been published by Koester and Rauch /7/, Mughabghab et al. /8/, Sears /9/ and ourselves /1,10/.

Besides being an important parameter in neutron scattering, $b_N$ is of course closely related to nuclear physics. The individual scattering lengths, $b^+$ and $b^-$, can in favourable cases be calculated from microscopic nuclear models /11/. They can also, in principle, be determined from resonance parameters or, at least, serve as a consistency check for the assignment of resonance spin values or for the existence of bound states /12/.

The ever-increasing complexity of structure research in chemistry and biology may use nuclear polarization as a particularly powerful tool. It has been proposed for a long time that the coherent scattering of hydrogen can be varied to a large extent by nuclear polarization. However, this method has not been applied, except for a demonstration exercise /13/, owing to the considerable experimental difficulty to obtain large hydrogen polarization. It should be worthwhile putting some effort into this method since it has two important advantages over the conventional deuteration. A much larger range of coherent scattering can be obtained in principle and this from a single sample without any chemical alteration /14/.

Finally, a last already old-time use of polarized nuclei should be mentioned here. Polarized hydrogen is an efficient broad-band neutron polarizer, down to very short wavelengths /15/. With the current polarized proton target technique, such an instrument should be considerably cheaper, smaller and easier to run than its ancestors in Dubna and Grenoble.
REFERENCES


/7/ KOESTER, L., RAUCH, H., Summary of Neutron Scattering Lengths, IAEA contract 2517/RR.


/14/ STUHRMANN, H.B., IAEA-CN-46/48, these Proceedings.

NEUTRON SCATTERING STUDIES OF NUCLEAR ORIENTATION IN THE \( \mu K \) RANGE

M. STEINER
Hahn-Meitner-Institut für Kernforschung,
Berlin (West)

Abstract

NEUTRON SCATTERING STUDIES OF NUCLEAR ORIENTATION IN THE \( \mu K \) RANGE.

In this paper the application of neutron diffraction to nuclear magnetism is discussed. Experiments have been performed with polarized and unpolarized neutrons on systems with hyperfine-induced, hyperfine-enhanced and magnetic-field-induced nuclear polarization. It is shown that these measurements will yield nuclear temperatures, nuclear spin lattice relaxation times and information about nuclear magnetic order. Nuclear \( T \) below 600 \( \mu K \) have been obtained. Experiments planned in the \( \mu K \) range are discussed.

In recent years significant progress has been achieved in the polarized neutron technique and in the performance of refrigerators reaching the \( \mu K \)-range. The use of the latest developments in both techniques and combining them, opens a new area of research for neutron scattering. The field of nuclear magnetism becomes accessible to neutron scattering by the use of the last, but commonly not used, part in the nuclear cross-section, namely \( b^+ - b^- \).

By reaching sufficiently large H/T values a static nuclear polarization \( \langle I_{\text{nuc}} \rangle \neq 0 \) can be produced, which can be detected through \( b^+ - b^- \). Some obvious applications of this new technique are the following:

(1) Having the \( \langle I_{\text{nuc}} \rangle \) variable for one constituent of the sample, the scattering length of this particular nucleus is variable and thus it is possible to label this nucleus and to obtain more information about structural details than without this marking possibility (applications in biology and metallurgy for example).

(2) One can determine \( b^+ - b^- \).

(3) Studies of nuclear magnetism, which up to now has been the domain of other techniques:
(a) Paramagnetic region

Measurement of nuclear polarization independent of external parameters without disturbing the system, which allows the nuclear temperature to be determined and can yield nuclear spin-lattice \( S-L \) relaxation times under specific conditions.

The direct measure of \( \langle I_{\text{nuc}} \rangle \) allows direct monitoring of the adiabaticity during demagnetization. This possibility makes studies possible of the onset of long-range nuclear order along the phase boundaries.
(b) Ordered region

The type of spontaneous nuclear order can be determined analogous to electronic magnetism, as well as phase diagrams.

Although the principle of studying nuclear polarization in static magnetic fields by means of neutron diffraction is of striking simplicity, many problems and difficulties exist in actually performing the experiment. To locate the main experimental difficulties and to find solutions to overcome them, and to check the different possibilities of the new technique, we have started a systematic study of the application of neutron scattering to nuclear magnetism at temperatures down to 600 μK. On the basis of our experiments different aspects and results of this technique are discussed. It is appropriate at the end of this introduction to mention the pioneering work by Abragam et al. /1/ who showed for the first time what neutrons can do in nuclear magnetism by obtaining the nuclear order in LiH and CaF₂. Their technique to obtain the low temperatures necessary, is a dynamic one based on the solid-state effect and therefore not generally applicable.

We have performed two essentially different types of experiments:

I. Study of hyperfine induced antiferromagnetic nuclear polarization, $I_{nuc}$, in CoF₂ and DyPO₄ using unpolarized neutrons.

II. Study of hyperfine enhanced nuclear polarization and adiabatic demagnetization in HoVO₄ using polarized neutrons.

In the type I experiments $I_{nuc}$ produces superlattice reflections as the electronic magnetic contribution does. However, if the electronic magnetic moment is parallel to the scattering vector, its contribution to the cross-section will vanish. Scattering from $I_{nuc}$ which does not have such selection rules will produce a Bragg-reflection with

$$ J \sim (b^+ - b^-)^2 I_{nuc}^2 $$

By scaling this intensity against the intensity of a reflection unaffected by nuclear polarization, $(b^+ - b^-)^2 I_{nuc}^2$ is obtained on an absolute scale. Thus, by knowing one of the above quantities the other one can be determined.

In CoF₂ $(b^+ - b^-)$ is known and thus, by measuring $J(001)$ $I_{nuc}^2$, the nuclear temperature can be obtained. Results of the short time experiments were published some time ago /2/ and therefore the results of a long time experiment is given here briefly. It takes approximately $10^3$ h to cool the nuclei to the temperature of the sample holder, which was kept at 9 mK all the time. The observation of the time evolution of beam heating effects demonstrated that the spin lattice relaxation time is around $10 - 40$ h at 12 mK. Apparently the cooling time of $10^3$ h is due to the famous 'thermal bottleneck' for the heat transfer crystal to sample-holder and the large nuclear specific heat to be removed.

In DyPO₄ the hyperfine field is so strong that for $T < 100$ mK $I_{nuc} \sim 1$, and thus the unknown $|b^+ - b^-|$ for Dy, can be
determined by measuring \( J(002) \). For a single crystal with natural isotope mixture \(|b^+ - b^-|_{\text{Dy}}\) was determined. Taking into account that only 45% of natural \(^{161}\text{Dy}, \; ^{163}\text{Dy}\) has \( I \neq 0 \) \((^{161}\text{Dy}, \; ^{163}\text{Dy})\) one obtains
\[ |b^+ - b^-|(^{161}\text{Dy}, \; ^{163}\text{Dy}) = 4.4 \times 10^{-12} \text{ cm} \]
a value which is very close to the exceptionally high value of
\((b^+ - b^-)\) for the proton.

It is clear that these conceptually very simple experiments
\( b^+ - b^- \) etc.

As a test system for the type II experiments we have chosen
\( \text{HoVO}_4 \) because there is detailed information about the nuclear magnetism /3/. Because \( I_{\text{nuc}} \) is achieved by the hyperfine enhanced external field, polarized neutrons have to be used and intensity changes are expected for several reflections due to \( I_{\text{nuc}} \). Two nuclear systems have to be taken into account: the \( ^{165}\text{Ho} \) and the
\( ^{51}\text{V} \)-system. Since all necessary parameters are known the \( H \) and \( T \)
dependence of \( J(200) \) can be calculated. Figure 1 shows results of
this calculation, demonstrating the importance of the contribution
of both nuclear systems to the change of the (200) intensity with \( T \)
and \( H \) - the characteristic of this system is the minimum in the \( J(T) \)
curves. This can be understood easily: \( I(\text{Ho}) \) dominates the high
\( T \) regime (because of the hyperfine enhancement), while \( I(\text{V}) \) becomes
important at low \( T \) only, and the minimum is due to the fact that the

**FIG. 1.** The \( T \)-dependence of the (200) Bragg intensity for \( \text{HoVO}_4 \) at different
applied fields.
sign of $b^+ - b^-$ for $^{165}$Ho and $^{51}$V is different. It is worth noticing that, because $(b^+ b^-)_V \gg (b^+ - b^-)_\text{Ho}$, even a very small polarization of V is sufficient to produce a significant change of $J(200)$.

In Figure 2 the time variation of $J(200)$ divided by $J(200)$ for $T \rightarrow \infty$ is shown for $H = 3T$. The expected minimum is clearly visible. From the final (200) intensity one finds $T_{\text{nu}} = 17 \text{ mK}$. The observed depth of the minimum is less than theoretically predicted, however. This is probably because the S-L relaxation time is different for the two magnetic systems. This is further substantiated by the cooling curve in 2T - there no minimum is observed. After 60 h cooling a slight heating of the lattice resulted in a slow reduction of of $J(200)$, indicating that the system is also in 2T on the low T side of the minimum.

It is thus possible to determine reliably the nuclear temperature by neutron diffraction, even in a rather complicated situation as in HoVO$_4$.

Having this possibility, the nuclear temperature can be followed during adiabatic demagnetization by just monitoring the respective Bragg intensities. We have performed such experiments, again on the HoVO$_4$ crystal. Figure 3 shows the low field part of such a demagnetization procedure from 3T. Just above 1 kG, $J(200)$ still corresponds to the same nuclear magnetization as in 3T ($T_{\text{nu}} = 17 \text{ mK}$). Thus, $T_{\text{nu}}$ at 1 kG = $17 \text{ mK} \times 1\text{ kG}/30\text{ kG} = 0.57 \text{ mK}$.

As can be seen from Figure 3 for $H < 1 \text{ kG}$, $J(200)$ raises with decreasing $H$, indicating the disappearance of the Ho polarization. This is interpreted as the onset of antiferromagnetic nuclear order as predicted by Bleaney /3/ and observed by neutron scattering by Suzuki et al. /4/. After demagnetization to 100 G, $J(200)$ decreased.
with time by a few per cent. This is clear evidence that the V system retains some polarization down to 100 G, thus being cooled to $T_{\text{nucl}} \approx 100 \mu K$.

During the course of those type II experiments, we had clear evidence of beam heating. Using different filters it became clear that it is the $\gamma$-contamination, which produces the beam heating with power of a few $n$-Watts.

The conclusion from the observed results is that neutron scattering is a powerful method for the study of nuclear magnetism, not only to determine spontaneous nuclear order (see /4/), but also, or even especially for, determining nuclear temperatures, studying nuclear spin-lattice relaxation times, determining the onset of non-adiabaticity etc. It is clear, however, that the combination of low temperatures and polarized neutrons is necessary to achieve all this.

The experiments described here paved the way for the most ambitious experiment imaginable in this field. Lounasmaa and his co-workers have studied the nuclear magnetism of Cu for the past 20 years, first in order to use Cu as a nuclear refrigerant and later for its own sake. Very recently they were able to reach temperatures of the Cu-nuclei as low as 25 nK, low enough to establish the spontaneous nuclear order of the Cu nuclei. This nuclear order is apparently antiferromagnetic /5/. To determine the unknown antiferromagnetic structure and the phase-diagram, cooperation was established between the neutron scattering groups at Risø, our Institute, and the low-temperature laboratory of the Technical University of Helsinki. On the basis of our results we believe that this neutron scattering experiment at 20 nK will be possible.
ACKNOWLEDGEMENTS

The contribution of our colleagues K. Kohlhoff, G. Rahn, U. Scheer and K. Siemensmeyer to this work is gratefully acknowledged. The fruitful co-operation with Dr. M. Kubota from the IFF of the KFA Jülich is also acknowledged. We are grateful for the help of Prof. Schärpf (ILL Grenoble), Dr. Weinfurter (Atominstitut Wien) and Dr. Wagner (PTB Braunschweig) during the installation of the polarized beam. The crystals used were grown by Dr. B. Wanklyn, Clarendon Laboratory, Oxford.

REFERENCES

ON-LINE EVALUATION OF POSITION-SENSITIVE DETECTOR (PSD) DIFFRACTION DATA

R. F. D. STANSFIELD, G. J. McINTYRE
Institut Max von Laue - Paul Langevin, Grenoble

Abstract

ON-LINE EVALUATION OF POSITION-SENSITIVE DETECTOR (PSD) DIFFRACTION DATA. The amount of raw data accumulated in a single-crystal diffraction experiment using a two-dimensional Position Sensitive Detector is usually so large that it is impracticable to store it. It is therefore necessary to reduce each local three-dimensional array of counts to a Bragg intensity, in a time not longer than the average time that one reflection is active. The statistically optimum procedure comprises an estimation of the background from a large number of counts, and an integration of peak intensity within a suitable three-dimensional envelope. A typical on-line method is described, using as an example the D19 diffractometer at the Institut Max von Laue - Paul Langevin (ILL) high-flux reactor. Current methods of PSD data reduction are reviewed. These fall into three groups according to the basis of the method used to find the integration envelope: (a) statistical criteria, (b) three-dimensional sigma(I)/I analysis, and (c) pre-calculation of the resolution function. On-line data reduction imposes special requirements on diagnostics to check the precision of the reduced data, especially at the start of an experiment, when any peculiarities must be identified and allowed for in the data-reduction procedure. The diagnostic possibilities resulting from the comparison of local with global characteristics of the background and the integration envelope are discussed.

1. INTRODUCTION

Two conditions that we wish a single-crystal neutron diffraction experiment to meet are a) efficient use of the neutron beam, and b) high accuracy of the estimated intensities of Bragg reflections. The first criterion is met by the experimental procedure, and the second by the data reduction procedure. An on-line method is one with concurrent procedures for collecting and reducing the data.

The complexity of data handling in a single-crystal diffraction experiment with a Position Sensitive Detector has been emphasized in a recent review by Convert and Forsyth [1a]. The instrument that we consider here consists of a crystal orienter and a two dimensional PSD, on a steady state neutron source. The orienter can have up to three degrees of freedom, such as the \( \omega \), \( \chi \) and \( \phi \) motions of a Eulerian cradle, and the
PSD can be positioned by rotating it about the sample position. Some of the methods that we describe are applicable to the analogous time-of-flight instrument on a pulsed source.

A typical experimental procedure has been described by Xuong et al. for an X-ray instrument with a PSD [2]. A three dimensional data array is built up from successive two dimensional frames of counts, by stepping the crystal about the scan axis. Compared with X-radiation, neutron beams have a much larger wavelength spread ($\Delta \lambda/\lambda$) and divergence. A peak centre can range over several detector elements on successive frames and a three dimensional search is required to locate it. A definition of the surface separating peak counts from background counts in three dimensions is also required for the best statistical estimation of the reflection intensity. The problem is exacerbated by the low signal to noise ratios obtained from neutron diffraction experiments with macromolecular (hydrogenous) samples.

The rate of accumulation of data is so large in such an experiment that it generally precludes storage of the raw data. A commonly adopted method of alleviating this problem is to pre-calculate the expected coordinates of all reflections. Then, either only small volumes of data are stored, or the raw data is stored temporarily and small volumes are extracted into computer memory. In both cases, the raw data must be reduced on-line and then discarded. The possibility to re-treat the raw data with the benefit of hindsight is lost. Therefore the data reduction procedure must include diagnostic checks to indicate when changes in the measurement or reduction procedures are necessary.

In this paper we outline the steps in a typical neutron experiment, using as an example the D19 diffractometer at the ILL high flux reactor. We describe our methods of data reduction and compare them with those of other workers. Finally we discuss how the available methods can satisfy the diagnostic requirements of on-line data reduction.

2. PSD DATA COLLECTION AND REDUCTION

The single-crystal diffractometer D19 is equipped with a relatively narrow, vertically curved multiwire proportional counter with 8192 cells [1b]. These are formed by 16 cathode wires which curve from top to bottom and 512 straight anode wires which cross the cathodes horizontally. The PSD surface subtends 64° vertically and 4° horizontally at 1.15 m from the sample position, which is also the centre of curvature. The PSD is mounted symmetrically about the horizontal plane and moves on a 20-arm. A Eulerian cradle is used to orient the sample. The instrument is controlled by a PDP 11/34 computer,
and a high speed link passes the raw data to a VAX 11/750 for data reduction.

From a crystal of decent diffracting power, the rate of data collection can be as high as one frame of 8192 integer counts per second, equal to 1 Mbyte per minute (with 2 bytes per integer). If the raw data were stored on magnetic tape, it would require one 2400 ft tape written at 1600 bpi\(^1\) every 40 minutes, which illustrates the need for on-line data reduction. The raw data from our experiments are not archived, but a 456 Mbyte disk does allow temporary storage.

The steps in a typical experiment are as follows.

2.1 Search for peaks

The first few reflections are found by performing scans at low angle, and allowing the data reduction program BRAGG VAX (described below) to find the coordinates of any peaks detected. A knowledge of the approximate crystal orientation can guide the choice of scans.

2.2 Index peaks and assign unit cell

In neutron experiments, the crystal unit cell is usually known in advance. A method for finding unknown cell dimensions from many observations with a PSD has been described by Wilkinson and Khamis, but is not in general use \([1c]\).

2.3 Determine the orientation matrix

The crystal orientation matrix, together with three translational offsets for the PSD and two angular offsets for the instrument angles, is determined by least-squares fit to the observed coordinates of reflections \([3]\). Note that scans for determining the orientation also yield valid intensity measurements of fully observed reflections, since no centring apertures are necessary with a two dimensional PSD.

2.4 Define a data collection strategy

A small number of crystal orientations defined by pairs of \(\chi\) and \(\phi\) are chosen to map volumes of reciprocal space within the angular limits of the instrument. At each \(\chi, \phi\) setting the PSD will be positioned a number of times depending on the chosen resolution limit, and for each of these positions, the crystal will be stepped about the vertical instrument axis, \(\omega\).

McIntyre has developed a program for fast prediction of reflection coordinates given \(\chi, \phi\), the PSD positions and an

\(^1\) bpi = bits/inch.
This program generates lists of scan commands optimised to receive groups of reflections and to skip empty regions of reciprocal space. For each reflection, the bounds of a three dimensional box are calculated in detector coordinates (cathode, anode) and ω coordinates. A look-up table is used to calculate the ω-extent as a function of χ, ψ (defined below).

2.5 Collect the data

The diffractometer control program reads the list of scan commands and positions the instrument angles, with a minimum of typically 15 steps in ω through each reflection [5]. The data are collected in normal-beam geometry, with angular coordinates χ across and ψ up the detector, and with the third dimension given by ω.

2.6 Treat the data

In an on-line experiment this step is performed concurrently with data collection. We have the choice of two data reduction procedures which differ in their algorithms for background calculation and single peak integration.

The first program, BRAGVAX, works sequentially on the complete frames of data from successive ω steps [1d]. The background is assumed to be those counts that obey Poisson’s statistics and a smooth ψ-dependent background curve is fitted to each frame. The program performs peak searches within the box areas predicted for reflections, and optionally over the rest of the frame. A contour is generated if enough neighbouring counts significantly above background are found. Summation of the background-corrected counts within the contour gives the contribution to the peak intensity. The program actually keeps the data for three frames in memory simultaneously so that a (quasi) three dimensional contouring can be effected by checking for significant neighbours on the preceding and succeeding frames. The contour line is not constrained to be any particular shape.

The second program, PEAKINT, was developed by Wilkinson and Khamis [1c]. It acquires and applies some knowledge of the resolution function during the data reduction, as opposed to the a priori approach of BRAGVAX. The box about the predicted peak position defines a set of counts which are extracted from temporary storage when a reflection has been completely scanned. Counts between boxes are not used at all. The extracted three dimensional data array is treated as a whole. For a strong peak, the shape and orientation of an ellipsoid is found from the moment-of-inertia tensor for equally weighted points contained within a suitable contour level (typically 5% of the peak height). By expanding and contracting the volume
of the ellipsoid while keeping the shape and orientation fixed, a "shape function" is derived from the intensity fraction contained in the ellipsoid as a function of volume. The limit to the peak extent is found by reference to the ellipsoidal surface for which \( \sigma(I)/I \) is a minimum. This is a generalisation of the method developed by Lehmann & Larsen for one dimensional profiles [6]. A library of strong peaks with their shape functions is kept and updated by the program. To increase the accuracy of weak peaks, their ellipsoid parameters and shape functions are deduced by interpolation amongst nearby strong peaks in the library. The statistically optimum volume in which peak-counts are summed is calculated as for strong peaks, and a correction factor to the intensity is deduced from the interpolated shape function. The treatment of a weak peak is purposely delayed in order to accumulate information in the library. The background for any peak is measured in a sufficiently distant ellipsoidal shell enclosing the peak, within the confines of the box.

3. COMPARISON OF METHODS

An advantage of BRAGGVAX is that only three frames of data are examined together, so that extension to a larger PSD is feasible. Because no assumptions are made beforehand, it successfully locates and integrates peaks without pre-calculation of their positions. A disadvantage is that such a method of contouring peaks has to be "tuned" to discriminate signal from noise, and therefore introduces bias in the estimation of weak peak intensities. The program PEAKINT, on the other hand, depends on a well-determined orientation matrix to extract boxes that fully enclose reflections. Analysis of recent measurements on deuterated lysozyme using both programs gave lower merging R-factors for PEAKINT reduced data than for BRAGGVAX reduced data [7].

Schoenborn [8], Roth & Lewit-Bentley [9] and Prince, Wlodawer & Santoro [10] have all described neutron PSD diffractometers for macromolecular studies which rely on the orientation matrix to predict the coordinates of reflections. Roth and co-workers are able to archive all their three dimensional data from low resolution experiments and perform their data reduction off-line. Schoenborn reduces his raw data by summing over the vertical detector coordinate \( \gamma \), to give a two dimensional data array in \( \phi, \omega \) which can be stored on magnetic tape and reduced off-line. In reducing the data from three to two dimensions he loses the advantages that a full three dimensional analysis would provide. Wlodawer uses a one dimensional detector with flat cone geometry so that the data array is also two dimensional, but in \( \gamma, \omega \). Film methods employed in neutron and X-ray diffraction provide two
dimensional data in $\gamma,\psi$, integrated over the crystal rotation angle.

Methods for the reduction of two dimensional neutron data have been described by Schoenborn [8], Spencer & Kossiakoff [11] and Sjölin & Wlodawer [12]. The only three dimensional procedures known to the authors in addition to those described in this paper, are those of Roth & Lewit-Bentley [9] and Wlodawer & Sjölin [13].

Both Schoenborn and Roth pre-calculate the shapes of the reflection envelopes taking into account the beam divergence, crystal mosaic, PSD resolution, monochromator spacing and wavelength band width.

Sjölin & Wlodawer find a two dimensional global background and employ a "dynamic mask" algorithm which is similar to the approach of BRAGG VAX. To avoid bias, they determine the integration envelope for weak reflections from a library of envelopes of strong reflections. The method of Spencer & Kossiakoff is essentially equivalent to that of PEAKINT but restricted to two dimensions. The major difference is that they employ a global background function.

Whatever the method used, its precision is improved if a global background is used. In an on-line application, this implies some temporary storage of the data, or at least of an array describing the global background. Of the methods described above, only PEAKINT performs a full three dimensional fit to the peak; all others work frame by frame to integrate the peak, once the background has been determined.

4. ON-LINE DIAGNOSTICS

On-line data reduction becomes advantageous when it provides diagnostic feedback on the performance of the data collection and of the data reduction [14]. Analysis of three dimensional data immediately indicates the discrepancies between expected and observed peak positions, and between predicted and observed peak extents. Because observed coordinates are found for every reflection measured, the orientation matrix can be continuously monitored. It can be updated if systematic discrepancies occur, using the coordinates of the most recently collected reflections, without necessarily losing time doing conventional centring.

Schoenborn and Sjölin & Wlodawer compare the local background of a reflection with the smoothed global background surface in order to signal anomalies in the data. These may be due, for example, to unexpected peaks that arise from movement or break-up of the crystal, or to parasitic scattering from the sample environment. Similarly, the observed shape parameters of strong reflections can be compared with those already in a library. Comparison of the reduced intensity with that from a
simple background-peak-background analysis of a one dimensional projection within the box, is a useful diagnostic that is included in BRAGG-VAX. Those methods that extract boxes should test the assumption that the peak fits completely within the box.

The extraction of three dimensional data into computer memory allows detailed analysis of the local background. The powder lines of cryogenic equipment, for example, may be removed by suitable treatment of the θ-projection of the background [15].

5. THE FUTURE

As detector technology improves, so the size and number of PSD's on one instrument will increase. More reflections will be observed simultaneously and the data reduction rate will need to be increased. With the computing power presently available the data reduction procedure may become the rate limiting step, if high accuracy and diagnostic feedback are to be maintained.

An investigation of multiprocessing has been initiated by S.Wilson at the ILL. The programming language OCCAM may be a suitable medium for constructing a system in which the software tasks required can be run in parallel, thus relieving the computational bottlenecks which can occur [16].

We believe that a combination of the various methods summarised in this paper will produce an efficient and accurate algorithm for on-line reduction of Bragg intensities from three dimensional data.

REFERENCES


A DEVICE FOR COMBINED NEUTRON-PHOTON PROCESSES IN CONDENSED MATTER

A. STOECKLI, A. ISACSON, M. KOCH, A. FURRER
Labor für Neutronenstreuung,
Eidgenössische Technische Hochschule Zürich,
Würenlingen, Switzerland

Abstract

A device has been developed for performing neutron scattering experiments with simultaneous irradiation by light at low temperatures. The light source is a halogen lamp which yields broad bands of wavelengths between 400 and 1000 nm by using appropriate filters. The light is guided by a series of lenses to the sample, which is mounted in a cooling system. A mechanical chopper may be inserted into the light beam in order to create light pulses with frequencies between 0.05 Hz and 5 Hz, and the neutron counts resulting from the dark and light experiments are separately stored. Our device has been used to study the light-induced dynamical behaviour of photosynthetic chlorophyll systems by neutron spectroscopy.

1. INTRODUCTION

When an optically active system is illuminated by light of a particular wavelength, it is electronically excited. After some time the excited system will return to its ground state. The corresponding relaxation mechanism can be either radiative, resulting in the emission of a photon (fluorescence), or radiationless, in which case a number of vibrational or phonon modes are created. Relaxation mechanisms generally take place through the simultaneous occurrence of both radiative and radiationless processes subject to energy conservation. Radiationless transitions increase the occupation number of the phonons and vibrons which can be directly measured by inelastic neutron scattering (INS). The INS technique appears to be the only experimental method which provides direct information on the radiationless transitions in terms of elementary quantum-mechanical processes, i.e. on the selection rules and lifetimes of the phonons and vibrons in the electronic-vibrational relaxation mechanisms.

The present paper describes how simultaneous photon absorption and neutron scattering can be experimentally realized at low temperatures. An instrumental device has been developed which is outlined in detail in section 2. An important aspect of the instrumental setup appears to be the possibility to create light pulses which is useful to unambiguously determine the amount of
heat dissipated into the system under light irradiation. In section 3 we present some preliminary results of INS experiments performed for a chlorophyll-protein system in D_2O solution. Further prospects of the method are summarized in section 4.

2. INSTRUMENTAL SETUP

Figure 1 shows a sketch of the instrumental setup. The light source is a 100 W halogen lamp whose efficiency is optimized by using a reflector. A parallel beam of 5 cm diameter is

created after transmission through a plane-convex lens. Various broad-band filters of variable band width have been used to yield wavelengths between 400 nm and 1000 nm. The light is guided to the sample through a lens system whose focal distance can be adjusted to the sample geometry. The sample is mounted in a closed-cycle He refrigerator. The light beam penetrates the sample chamber through a glass disk or lens inserted into the radiation shield of the cooling system. The maximum intensity at the sample position is 2 MLux.

In order to be able to unambiguously discriminate between heating effects and actual nonequilibrium phenomena in the INS experiments, a mechanical chopper has been inserted into the light beam. The light chopper is a rotating wheel with regularly arranged openings which periodically interrupt the light beam, thus creating light pulses whose frequencies can be varied between 0.05 Hz and 5 Hz. The neutron count-rates are separately stored for the dark and light experiments.

A crucial part of the instrumental setup is the sample cell which has to meet simultaneously three different experimental requirements, namely optimum scattering properties in the INS experiments, optimum efficiency concerning light irradiation and optimum cooling. Various types of sample cells realized at our laboratory have been described elsewhere \cite{1}.

3. EXPERIMENTAL RESULTS FOR A PHOTOSYNTHETIC CHLOROPHYLL SYSTEM

The function of photosynthetic systems is to absorb light energy and to convert it into chemical energy with optimum efficiency. The energy transfer mechanism occurs through several steps, e.g., energy transfer from the electronically excited antenna chlorophyll complexes to the reaction centre. Part of the excitation energy, however, may be lost because of the electronic-vibrational coupling in which case low energy phonons are created. This effect increases the phonon occupation number \( n(\omega) \) which can be directly measured by INS experiments through the incoherent cross section

\[
\sigma_{\text{inc}} \sim e^{-2W \frac{g(\omega)}{\omega}} <\frac{1}{2} + \frac{1}{2} + n(\omega)>.
\]  

We have performed INS experiments for various chlorophyll systems in solution. As an example we present the results obtained for a suspension of membranes of the photosynthetic bacterium Rhodopseudomonas viridis in \( D_2O \) buffer at a protein concentration of roughly 20 mg/ml \cite{2}. The effect of light irradiation (400 \( \leq \lambda \leq \) 800 nm) is shown in the normalized energy spectra of figure 2. Under light irradiation there is
a significant change of intensities; in particular, the intensity of the elastic line decreases, whereas the intensities in the inelastic part of the spectrum increase. It is evident from equation (1) that this can only be explained by increasing the phonon occupation number by $\Delta n(\omega)$. However, since a temperature gradient was set up under light irradiation ($T = 28$ K for the sample frame, $T = 55$ K for the centre of the sample), it is not clear whether $\Delta n(\omega)$ is due to a simple heating effect or to an effective increase of $n(\omega)$ through the creation of excited-state phonons.

Heating effects can be separated when the pulsed lightbeam technique described in section 2 is used. Typical results are shown in figure 3 which indicates that the major part of the observed intensity differences are due to heating effects.
FIG. 3. Intensity differences of the dark and light experiments versus inverse pulse frequency. The full and open circles refer to the inelastic and elastic parts of the observed spectra with energy windows of $\Delta E = 6.5 \pm 3.5$ meV and $\Delta E = 0 \pm 1$ meV, respectively.

However, small residual intensity differences remain between the light and dark experiments upon extrapolation of the pulse frequency to infinity. Obviously the anharmonic phonon coupling is rather strong, so that the system can almost be stabilized in thermal equilibrium.

4. CONCLUSIONS

An instrumental setup has been described which allows simultaneous photon absorption and neutron scattering experiments at low temperatures. In our case the light source is a halogen lamp, but it may sometimes be advantageous to use laser light which has an almost infinitely narrow bandwidth and generally more intensity, particularly when a pulsed laser is used. In addition laser light offers cryotechnical advantages, since the laser beam can penetrate the inside of a cryostat through a rather small hole in the radiation shield, so that low temperatures can more easily be stabilized.

When condensed matter is irradiated by light, there is a continuous production of "hot" phonons, so that the phonon system is no longer in thermal equilibrium. The actual non-equilibrium phenomena observed in the present experiments,
however, turned out to be rather small due to the short lifetimes of the phonons generated by light. More favourable conditions may exist in model systems such as ruby where lifetimes of the order of ns have been observed for the 3.6 meV phonons resulting from the nonradiative relaxation of the excited $^2E$ state (3). At present we are preparing such an experiment which has already been suggested twenty years ago (4).

Combined neutron-photon studies may also be used to enhance the energy transfer in the neutron scattering process into the eV-region by the photon energy (5). Thus the dynamic structure factors may be measured at large energies and modest wavevectors which is a domain that is usually difficult to access because of kinematic constraints.

Financial support by the Swiss National Science Foundation is gratefully acknowledged.

REFERENCES

INTERFACE INVESTIGATION BY SLOW NEUTRON REFLECTION

B. FARNOUX
Laboratoire Léon Brillouin*,
CEN-Saclay,
Gif-sur-Yvette, France

Abstract

INTERFACE INVESTIGATION BY SLOW NEUTRON REFLECTION.

Owing to their wave-like character, neutrons can be reflected or refracted at a boundary between two media. In many cases the index of refraction of the material is less than unity, so total reflection can occur when the incident beam is at an angle less than the critical glancing angle. The angular dependence of the reflectivity has been shown to be correlated to the variation of the index of refraction in a direction perpendicular to the surface. On the other hand, the depth of penetration in the medium is also angle-dependent. This provides a method of studying thin layers of material deposited on surface mirrors. This technique is now being developed and some preliminary results obtained by a time-of-flight (TOF) technique are presented.

The understanding of surface and interfacial phenomena is of considerable interest, both practical and theoretical. Many experimental techniques have been developed to study the near-surface of solids and liquids or layers of adsorbed molecules. In particular it has been shown /1-3/ that surface investigations can be performed by using a well-collimated monochromatic beam of X-rays, striking the surface at a glancing angle within the range for which total reflection occurs /4/. Recently it has been proposed /5,6/ to investigate the concentration profile near a surface by the reflection of a neutron beam.

This method has already been tested on known profiles /7-10/. The problem is now to propose experiments in which the profiles are unknown, and for which the use of neutron beams are essential. Such an experiment has actually been performed /11/, using the magnetic interaction of the neutron with electrons of a metallic sample in the superconducting state. Here we consider an interface in physical chemistry for which the interaction with the neutron is purely nuclear.

*Laboratoire commun CEA-CNRS.
For a non-magnetic material, and neglecting the absorption, the index of refraction \(n\), is given by the expression /12/:

\[
n - 1 = -\frac{1}{2} \frac{1}{\xi_n^2} K^2
\]

where \(K = 2\pi/\lambda\) is the wave number and \(\xi_n\) is the nuclear characteristic length of the medium, given by

\[
\xi_n = (4\pi Nb)^{-1/2}
\]

\(N\) is the number of scattering nuclei per unit volume and \(b\) the coherent scattering amplitude.

For slow neutrons the critical glancing angle for total reflection \(\theta_c\), expressed as \(\cos \theta_c = n\), is such that

\[
\theta_c = \frac{1}{K\xi_n}
\]

Values of \(\xi_n\) and \(\theta_c\) for common materials are given in Table I.

The interesting thing to note is that the reflected intensity does not fall to zero at the critical value \(\theta_c\). The angular dependence of the reflectivity \(R(\theta)\), expressed as the ratio of the reflected to incident intensity, is equal to the square of the reflection coefficient \(r\) of a semi-infinite slab of material /13/:

\[
r = \frac{\sin \theta - (n^2 - \cos^2 \theta)^{1/2}}{\sin \theta + (n^2 - \cos^2 \theta)^{1/2}}
\]
If the scattering amplitude density $N_b$ varies with the distance from the surface $z$, the characteristic length is no longer a constant and exhibits a spatial variation $\xi_n(z)$. Measurements of the reflectivity profile $R(\theta)$ are then quite sensitive to the surface inhomogeneities.

In most cases the reflectivity profile is measured by using a well-collimated monochromatic neutron beam, as a function of incident angle $\theta_i$. This technique is well suited for solid surfaces, but is difficult to use for liquid interfaces.

An alternative method is to measure the reflectivity profile at constant incident angle $\theta_i$ as a function of the neutron wavelength. In that case the neutron beam is reflected with an intensity which varies with the wavelength dependence of the index of refraction (1). The angle of incidence $\theta_i$ is chosen so that only long-wavelength neutrons are totally reflected. Using a wide incoming distribution of incident wavelengths, a single experiment at a fixed incident angle $\theta_i$ provides the complete reflectivity profile $R(\lambda)$.

This technique has been tested using a cold neutron beam of the ORPHEE reactor. The TOF measurement was performed with the small-angle spectrometer PAXY with the following set-up: The cold neutron beam provided by the neutron guide (characteristic wavelength 4 Å) is precisely collimated with 2 horizontal slits (0.4 x 5 mm$^2$) and falls on the horizontal sample with an incident angle of $\theta_i = 4.935 \times 10^{-3}$ rad ($\Delta \theta_i = 2.12 \times 10^{-4}$ rad).
Neutron pulses of 60 μs width are delivered by a small chopper with one slit of 1 x 10 mm², at a repetition rate of 35 pulse/s. The time range for data collection is 25 600 μs with a channel width of 100 μs. Direct and reflected beams are recorded by the time-of-flight technique with the XY multidetector (pixel size 0.5 x 0.5 cm²) set 5 metres away from the sample. Typical wavelength distributions are shown in Fig. 1.

The measured reflectivity profile for a liquid/air interface is plotted in Fig. 2. The data are collected in few hours.

Another good example of the possibilities of TOF technique is the determination of the magnetic depth profile of a magnetically active surface, recently performed by using the intense pulsed neutron source at Argonne /11/.

These examples show that the investigation of interface phenomena is feasible using cold neutron beams. This technique will provide new information on the density profile, especially by the use of the time-of-flight method. However, it will be developed more easily with the advent of the new pulsed neutrons sources.

REFERENCES

MI8EMOL: A SIX-CHOPPER TIME-OF-FLIGHT SPECTROMETER INSTALLED ON A NEUTRON GUIDE OF THE ORPHEE REACTOR

S. HAUTECLER, E. LEGRAND, L. VANSTEELANDT, P. D’HOOGHE, G. ROOMS*
CEN/SCK, Mol, Belgium
A. SEEGER, W. SCHALT
Zentralabor für Elektronik, Kernforschungsanlage Jülich GmbH, Jülich, Federal Republic of Germany
G. GOBERT
Institut Max von Laue - Paul Langevin, Grenoble

Abstract

MI8EMOL: A SIX-CHOPPER TIME-OF-FLIGHT SPECTROMETER INSTALLED ON A NEUTRON GUIDE OF THE ORPHEE REACTOR.

A variant of a four-chopper time-of-flight spectrometer obtained by replacing the first and the last unit by pairs of discs rotating in opposite directions has been built and operated. With respect to a similar four-rotor machine the resolution is increased by a factor of 2. Indeed, the elastic resolution of MI8EMOL is comparable with that of the IN5 spectrometer at the Institut Laue - Langevin, though smaller disc diameters and lower rotation speeds combined with a somewhat larger beam width are used. The computed resolution curve fully agrees with experimental results.

1. INTRODUCTION

A four-chopper time-of-flight spectrometer implanted on a neutron guide fed by a cold source is, within certain limitations, a very flexible instrument that is well adapted to the study of random motions (rotational re-orientations, translational diffusion) giving rise to quasi-elastic scattering. First of all, one is able to select the incident neutron energy $E_0$ according to the range of momentum transfers $Q$ one wants to cover. Then, the elastic resolution $\Delta E_0/E_0$ needed by the system under investigation can be achieved by choosing adequately the speed $\omega$ of the first, second and fourth choppers. Finally, the requirement of avoiding overlapping of the scattered neutron time distributions corresponding to successive bursts can be satisfied by reducing the speed of the third chopper. Thus, in such an instrument, represented by IN5 at the Institute Laue Langevin, the incident energy, the resolution and the repetition rate can be selected independently.

*Also IKW and U. I. Antwerp
FIG. 1. Schematic lay-out of the six-chopper time-of-flight spectrometer, MIBEMOL, at the ORPHEE reactor in Saclay. The energy of the neutrons incident on the sample is essentially determined by the phase angle between the two pairs of choppers 1 & 2 and 5 & 6. The resolution is dictated by the rotational speed chosen for these choppers. Higher-order contamination is eliminated by chopper 3 and the overall repetition rate of the neutron bursts is controlled by chopper 4, which may be allowed to rotate at a lower speed than the others.

TABLE I. SOME PARAMETERS OF THE SPECTROMETERS MIBEMOL AND IN5.

<table>
<thead>
<tr>
<th></th>
<th>MIBEMOL</th>
<th>IN5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chopper disc diameter (cm)</td>
<td>43.4</td>
<td>54</td>
</tr>
<tr>
<td>Radius at window centre (cm)</td>
<td>18</td>
<td>24</td>
</tr>
<tr>
<td>Maximum rotation speed (rev/min)</td>
<td>10 000</td>
<td>20 000</td>
</tr>
<tr>
<td>Beam width (cm)</td>
<td>2.5</td>
<td>2</td>
</tr>
</tbody>
</table>
FIG. 2. Calculated elastic resolution at various chopper speeds. Crosses indicate experimentally obtained values.

For any given incident energy the highest resolution of a four-rotor machine is limited by the maximum value \( \omega_{\text{max}} \) of the chopper speed. An easy way to gain a factor 2 in resolution consists in replacing the first and the last chopper by pairs of discs closely spaced but rotating in opposite directions. This arrangement was considered by Egelstaff /1/ already thirty years ago, whilst detailed characteristics were derived by Lowde /2/.

2. MIBEMOL

Figure 1 schematically shows the six-chopper time-of-flight spectrometer MIBEMOL which has been implanted at the end of the guide G6 of the ORPHEE reactor of the Laboratoire Léon Brillouin at the Centre d'Etudes Nucléaires de Saclay. Some of the instrument
parameters are given in Table I, together with the corresponding values for IN5. With respect to the latter instrument, the main differences lie in the utilization of somewhat smaller discs and lower maximum rotation speed. This choice allows the use of considerably simplified mechanics (designed at the ILL for D7) where a disc is directly fixed on the axis of a standard 3 pole-paired synchronous motor of the reluctance type. The ball bearings and the motor armature run in air, the disc itself is separated by a labyrinth seal and operates under vacuum. The chopper discs are made of aluminium, the annulus rotating in front of the guide being covered with neutron absorbing material made of a mixture of resin and gadolinium oxide. Two windows at 180° from each other are provided in this layer. The power units supplying the motors are static frequency converters in thyristor technology, controlled by a CAMAC instrumentation developed at KFA Jülich /3/. It has been checked that, in the long term, the speed and phase stabilities are better than $10^{-4}$ and 0.03°, respectively.

Figure 2 shows the computed elastic resolution $\Delta E_0/E_0$ as a function of the incident energy and for several chopper speeds. The interrupted line represents a typical resolution curve calculated for IN5. The three crosses give the full widths at half maximum of triangles fitted to the elastic component of vanadium runs; these experimental data fully agree with the calculated values at 6000 rev/min.

A full account of the characteristics and performance of MIREMOL will be given elsewhere /4/.

3. OUTLOOK

Regarding possible improvements one can mention the development of fibre-glass discs and of magnetic bearings of passive and active type. Also, high-speed synchronous motors with hysteresis armature in combination with new developed harmonic-free static frequency converters would in future allow the construction of electrically efficient and economic chopper systems.

ACKNOWLEDGEMENTS

The authors wish to thank Dr. M. Nève de Mévergnies, head of the Nuclear Metrology Department at the CEN/SCK, and Professors K. Michel from the U.I. Antwerpen and L. Van Gerven from the KU Leuven for their active support in this project, which was performed under the joint collaboration of the CEN/SCK Mol and the Belgian Interuniversity Institute IIKW.

Dr. L. Bevaart (IIKW) and Dr. R. Kahn (LLB) have taken part in the studies leading to the determination of the main parameters of the spectrometer. The programming performed by Dr. L. Bevaart (IIKW), Dr. R. Colson, Dr. W. Wegener (CEN/SCK), and Dr. G. Coddens (IIKW) is gratefully acknowledged.
REFERENCES


/4/ CEN/SCK BLG Report (to be published).
PULSED SOURCES
(Sessions VII and IX)
Chairmen

C. G. WINDSOR
United Kingdom
(Session VII)

J. KJEMS
Denmark
(Session IX)
Invited Paper

INSTRUMENTATION FOR PULSED SOURCES

A. J. LEADBETTER
Neutron Division, Rutherford Appleton Laboratory,
Chilton, Didcot, Oxon,
United Kingdom

Abstract

INSTRUMENTATION FOR PULSED SOURCES.

A review is given of most of the major types of neutron scattering experiments now being, or about to be, undertaken at pulsed sources. The various kinds of total scattering, polarized neutrons, inelastic and quasi-elastic scattering measurements are discussed and the methods are illustrated particularly using for example the instruments on the Spallation Neutron Source (SNS) at the Rutherford Appleton Laboratory. Typical data are shown to demonstrate current achievements using pulsed neutron sources and some of the first results obtained on the SNS are also shown.

1. INTRODUCTION

Pulsed neutron techniques are not only now well-established but are also still developing rapidly, although they are still relatively young compared with the use of continuous neutron sources. Windsor's book /1/ describes the principles, opportunities and problems of the use of such sources and very recently a number of excellent reviews have been published which include discussion of instrumentation for pulsed sources and its utilization /2-5/. This paper will not therefore include those aspects of the underlying principles of using pulsed neutron sources which have been adequately covered in these recent reviews. It will rather illustrate the types of measurement which can be made by reference to new results and novel developments at existing pulsed sources and to the new instruments being installed at the world's newest, and potentially most powerful spallation neutron source - the SNS - now nearing completion at the Rutherford Appleton Laboratory. This discussion should then also indicate the great potential for future scientific achievement as these sources and their instrumentation are developed.

Much discussion has taken place in recent years about the relative merits of pulsed and continuous neutron sources. This is not a simple problem and the difficulties are exacerbated by the fact that one is comparing on the one hand the performance of relatively new instrumentation on weak sources with correspondingly long extrapolations to fully developed instrumentation on powerful sources and, on the other hand, several decades of instrument development culminating in sophisticated instruments on existing
high-flux reactors and a short extrapolation to a more powerful source. However, enough results are now available in some areas, such as powder diffraction, for the performance of pulsed sources to speak for itself and this will be increasingly the case in other areas in the near future as techniques and source are further developed and especially as SNS operation approaches full power. Also the complementarity of the two types of sources should be emphasized with reactors generally better below some cross-over energy above which pulsed sources become increasingly better. This is a useful generalization, although it should certainly be used with caution, but for SNS at full power the cross-over energy is estimated to be less than 10 meV with an effective performance about 3 orders of magnitude better than a high flux reactor at 1 eV ($\sim 0.3 \AA$).

The principal pulsed sources either now operational or under construction are given in Table I which also contains a brief indication of the major types of instrument at each source. This list is not exhaustive because of lack of space and the fact that the instruments are often modified and new ones are always under development. It does, however, give an adequate guide for present purposes to the range of instruments currently available.

2. NEUTRON SPECTRA

Moderators for pulsed sources have to be designed to slow down the neutrons to produce the maximum flux at the required energy while maintaining an adequately sharp neutron pulse. With the short source pulses of the facilities listed in Table I the neutron pulse widths are determined by the moderator assembly. The moderator material should be of dense hydrogenous composition and water, $l$-CH$_2$, $\ell$-CH, s-CH$_4$ and polythene at various temperatures have all been used.

At the SNS four moderators are installed in wing geometry: two ambient temperature Gd-poisoned water moderators above the target, and a $l$-CH$_4$ at $\sim 95K$ plus a H$_2$ moderator at 25K below the target. These have been designed to give a variety of spectral characteristics optimized for the different types of instrument.

The spectral distributions of neutrons were measured during the first full SNS operation run in December 1984 and are shown in Fig. 1. These spectra are typical of such moderator assemblies on any of the pulsed sources listed in Table II. The rich epithermal region of the spectrum together with the high flux of neutrons out to 10Å produced by the H$_2$ moderator should be particularly noted. Comparison of observed and predicted values for spectral shape and absolute flux presented in Fig. 1 and in Table II show excellent agreement between the two.

The pulse shapes are complicated functions of time but may usefully be characterized by the half-width and by the long time exponential decay. The predicted half-widths for each of the moderators is given in Fig. 2 and preliminary analysis of the first results shows the observations to be consistent with these
TABLE I. PULSED SOURCES AND THEIR INSTRUMENTS

KENS-1' at KEK, Japan (1985)
Proton current: 10 μA at 500 MeV into \(^{238}\text{U}\) Target
Proton Pulse width 0.5 μS, frequency 15 Hz
Instruments: 3 powder diffraction, 1 small-angle,
2 polarized neutron, 1 single-crystal diffraction,
1 phonon spectrometer, 1 quasi-elastic crystal analyser,
1 electronvolt spectrometer, 1 crystal analyser inelastic.

IPNS-1 at Argonne, USA (operational)
Proton current: 12 μA at 500 MeV into \(^{238}\text{U}\) Target
Proton Pulse width 0.1 μS, frequency 30 Hz
Instruments: 2 powder diffraction, 1 single-crystal,
2 chopper, 1 small-angle, 1 crystal analyser inelastic,
1 electronvolt spectrometer, 1 polarized neutron.

WNR-PSR at Los Alamos, USA (1986)
Proton current: 100 μA at 800 MeV into \(^{238}\text{U}\) Target
Proton Pulse width 0.3 μS, frequency 24 Hz
Instruments: 2 powder diffraction, 1 single-crystal,
1 filter difference inelastic, 1 phonon spectrometer,
1 small angle, 1 electronvolt.

SNS at Rutherford Appleton Laboratory, UK (1985)
Proton current: 200 μA at 800 MeV into \(^{238}\text{U}\) Target
Proton Pulse width 0.4 μS, frequency 50 Hz
Instruments: 2 powder diffraction, 1 small-angle,
1 chopper, 1 crystal analyser inelastic,
1 quasi-elastic backscattering analyser,
1 single-crystal diffraction,
1 electronvolt spectrometer,
1 polarized neutron.

HELIOS at AERE, Harwell, UK (operational)
Electron linac of 45 kw
Electron Pulse width 2.0 μS, frequency 150 Hz
Instruments: 2 powder diffraction, 1 small-angle,
1 chopper, 1 phonon spectrometer, 1 electronvolt,
1 active sample diffractometer.

predictions. The exponential decay constants have also been obtained from the data and are also compared with predicted values in Table II: again the agreement is excellent.

Final points to note are that the power pulse at zero time is very sharp with a decay constant of about 30 μs and the background of time incoherent neutrons is extremely low, showing the shielding and collimator designs to be highly efficacious.
3. DETECTORS

The vast majority of neutron scattering measurements have hitherto used gas detectors with RF$_3$ and now mostly He as the active component. Position sensitive detectors have also been increasingly used and their status has recently been extensively reviewed /7/. A new development has been the production at SNS of Li-loaded glass scintillator detectors. The absorption of a neutron releases energetic charged particles which produce ion pairs along their track and the resulting electrons excite the active component of the glass Ce$_3^+$, resulting in a flash of light which is detected by a photomultiplier. The advantages of these detection systems are a) their efficiency, especially for higher energy neutrons, b) their short dead times - making possible higher count rates, c) the varied detector shapes possible, and d) their cost, which is about half of that for an equivalent area of gas detectors. The main disadvantage in their development has been their $\gamma$-sensitivity but this has now been reduced to a level such that they may be used with advantage at
TABLE II. SUMMARY OF SNS MODERATOR PERFORMANCE

<table>
<thead>
<tr>
<th>Moderator</th>
<th>Ambient water (1.5 cm poison depth)</th>
<th>Liquid methane</th>
<th>Liquid hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermalization ratio</td>
<td>2.1 ± 0.2 (2.4)</td>
<td>1.8 ± 0.2 (1.8)</td>
<td>2 ± 0.5 (2.6)</td>
</tr>
<tr>
<td>Effective moderator</td>
<td>35 ± 1 (34)</td>
<td>13 ± 2 (10)</td>
<td>2.6 ± 1 (2.8)</td>
</tr>
<tr>
<td>temperature (meV)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Slowing-down exponent</td>
<td>0.09 ± 0.02 (0.1)</td>
<td>0.08 ± 0.02 (0.1)</td>
<td>-</td>
</tr>
<tr>
<td>Transition between</td>
<td>150 ± 20 (80 ± 10)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maxwellian and</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>epithermal (meV)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Long-time exponential</td>
<td>22 ± 5 (20)</td>
<td>31 ± 2 (32)</td>
<td>25λ ± 2λ (25λ)</td>
</tr>
<tr>
<td>decay constant of the</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>moderator pulse (μs)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^{a}\)figures in parentheses are predicted values from Ref. /6/.

least for pulsed sources where the γ-flash occurs at very short times.

Two examples of these detectors and their use will be given: the first is a simple new design of monitor counter invented by P. Davidson, the efficiency of which may be easily varied over a very wide range and the second an annular multidetector with fibre-optic coding designed for use on the high-resolution powder diffractometer (HRPD) at SNS.

The design of the new monitor counters is based on a mesh of glass scintillator cemented to glass fibres as shown in Fig. 3. These are cheap and easy to construct and their detection efficiency is given by

\[ n = \left[ 1 - \exp \left( -2.375 \frac{x}{\sqrt{E}} \right) \right] K \]

where \(x\) is given in cm, \(E\) in eV, and \(K\) the ratio of scintillator to total area. For example 0.5 mm cubes at 5 mm separation gives \(K = 10^{-2}\). The γ-sensitivity of these detectors to \(^{60}\)Co (∼ MeV)
The principle of the HRPD detector system has already been described [7] and the area detector configuration is shown in Fig. 4. Although such detectors indeed have relatively high γ-sensitivity the γ-backgrounds in pulsed spallation sources should be much more favourable, the main source on HRPD being n-γ conversions in the sample. The results from the first SNS neutron run appear to confirm this. Another possible problem with these detectors was thought to be 'cross-talk' between spatial channels but this has now been shown to be at a level of less than 1%. The long-term stability has also been shown to be good - the variation in count rate over four weeks on a fixed source was ± 0.5%. The detection efficiency should be ~70% for 1Å neutron and the first results are consistent with this.

Actual diffraction results obtained with this detector system are shown in Section 4 and although more calibration remains to be done the excellent performance is already clear and it is now evident that these scintillator detection systems are going to be extremely useful for pulsed source measurements in the future.
small pieces of scintillator glass cemented to 25 micron glass supporting fibres, e.g., 0.5 mm pieces at 5 mm pitch gives a detection efficiency of ~ $5 \times 10^{-4}$ at 1 Å

FIG. 3. General arrangement of the scintillator monitor counters on SNS,

4. TOTAL SCATTERING INSTRUMENTS

With all of these instruments a diffraction pattern is obtained by simple time-of-flight measurement in each detector element with no determination of energy changes on scattering. Such total scattering experiments give, in the static approximation, information about the instantaneous structures of materials. Three broadly different types of instrument will be considered.
4.1. Diffractometers for studies of crystalline powders, amorphous materials, liquids and diffuse scattering

This heading covers a very wide range of scientific research but the basic principles of the diffractometers are essentially the same, although the details will depend on the major applications envisaged. The major variables to be considered are the intensity, the resolution and the detector configuration. High resolution can be chosen at the expense of intensity (and vice versa) although the loss of intensity may be ameliorated by the use of guide tubes. The detector configuration is chosen to maximize count rate consistent with consideration of resolution, background and cost but also, as appropriate, to give azimuthal and hence, for example, textural information and to obtain diffraction patterns at low angles with high neutron energies so as to minimize inelasticity effects.

4.1.1. Medium-resolution diffractometers

Such instruments exist on all pulsed sources, where for this purpose medium resolution is defined as $0.003 < Q/Q_0 < 0.01$, and this resolution is obtained over a wide $Q$ range from $< 1\text{Å}^{-1}$ to $> 30\text{Å}^{-1}$. In order to provide this resolution the sample is typically ~10 m from the source with a sample detector distance of
FIG. 5. The sample and detector configuration on the LAD instrument on SNS which is typical of a medium-resolution powder diffractometer. The sample is at 10 m from the $^1$CH$_4$ moderator.

about 1 m and a variety of scattering angles. A typical configuration is illustrated in Fig. 5 by the liquids and amorphous materials diffractometer (LAD) on the SNS /8/. Note the presence here of relatively low-angle detectors which are important in studies of non-crystalline materials.

Instruments of this type have already provided a wealth of new data on both crystalline and non-crystalline samples. A recent example is given in Fig. 6 which shows results obtained using the 0.4% resolution general purpose powder diffractometer at Argonne on Cr$_2$(MoO$_4$)$_3$ /9/.

Profile refinement of the results enabled 114 structural parameters to be determined which is at least comparable to the best which has been achieved on reactor instruments.

For non-crystalline materials and especially for liquids containing light atoms it is now clear /10,11/ that in order to minimize inelasticity effects measurements should be made at relatively low angles - for example, less than 20° for light elements - and using appropriately high-energy neutrons so as to
FIG. 6. Time-of-flight powder diffraction data on $\text{Cr}_2(\text{MoO}_4)_3$ [9]. Profile refinement fitting, differences between calculated and experimental data and the location of the Bragg peaks are shown.

give the required $\theta$ range. This was beautifully demonstrated recently [12] by comparison of reactor and pulse source ifffraction data on liquid $\text{H}_2\text{O}$ showing the virtual removal of inelastic effects with the liquids diffractometer at the WNR pulsed source. A preliminary design for an instrument at SNS called SANDALS [13] envisages a $\theta$ range of $0.05 \, \text{Å}^{-1}$ to $50 \, \text{Å}^{-1}$ using rings of 5 mm thick glass scintillator detectors to cover azimuthally the angular range $0.5$ to $50^\circ$ with resolution $\varphi/\theta$ varying from $24\%$ at $0.5^\circ$ through $3\%$ at $5^\circ$ to $0.6\%$ at $50^\circ$. This kind of instrument, which is unique to pulsed sources with their rich epitheral spectrum, will make a great impact on structural studies on non-crystalline materials in the future.

4.1.2. High-resolution powder diffractometers

High resolution is obtained by the use of long flight paths and high scattering angles. The highest resolution instrument so far constructed is the HRPD at SNS which is illustrated in Fig. 7. The diffractometer is situated at the end of a 96 m curved guide which has a radius of 18 km, the transmission of which is illustrate in Fig. 8, which shows how effective such a guide is in eliminating high-energy neutrons. Choppers are used to remove pulses or restrict the wavelength transmission because of the frame overlap encountered with such long flight paths.

The best attainable resolution of $0.4\%$ is obtained using a sample of radius $\geq 15$ mm at 2 m from the detector. The first results
FIG. 7. The high-resolution powder diffractometer (HRPD) on SNS. The sample is shown at the highest resolution position 2 m from the detector and 96 m from the CH₄ moderator.

FIG. 8. The transmission of the 96 m guide of HRPD showing the efficient removal of high-energy neutrons.

obtained with this instrument are shown in Figs 9 and 10. The former shows a combined diffraction pattern in one ring of one octant of the detector (cf. Fig. 4) from a single crystal of pyrolytic graphite at 2 m and a sample of NiO at 1 m from the detector. The latter shows an enlarged plot of the 111 and 111 peaks in NiO obtained from a sum over five detector segments. The full beam size is 2.5 x 8 cm², the NiO sample was 1.5 x 8 cm² and the large graphite crystal was illuminated by the beam passing by, and through, the NiO sample. These plots illustrate the high resolution
and high signal-to-background obtained with this instrument. It is also worth emphasizing that the run was made with a very low proton intensity so that the data shown would have been obtained in less than 1 s of running time at full SNS power and there would also be the full 8 detector octants each with 20 segments instead of the single segment shown for the powder data of Fig. 9 and the five segments of Fig. 10.

4.1.3. High-intensity powder and diffuse-scattering instrument

The high brightness of the neutron pulses may also be used to optimize count rate for moderate resolution. This is useful for kinetic powder diffraction measurements and studies of diffuse scattering of both structural and magnetic origin in a wide variety of crystals. A preliminary design study for such an instrument on SNS has given very promising results /14/ by reducing the moderator-sample distance to 7 m, and using a sample detector distance of 1 m with an azimuthal array of counters and a sample size of up to 1.5 cm horizontal by 3 cm vertical, an extremely high count rate instrument may be achieved as shown in Table III where the predicted performance of the diffractometer is compared with other existing

---

**FIG. 9.** The diffraction patterns of a pyrolytic graphite (PG) single crystal at the 2 m sample-detector position and a NiO powder sample at the 1 m position measured simultaneously on one of the 20 detector rings of a single detector octant on HRPD at SNS.
FIG. 10. A larger-scale plot of the 111 and 11T reflection of NiO from Fig. 9. The two NiO peaks arise from a small magnetic lattice distortion from cubic symmetry. The relative peak splitting is $1.5 \times 10^{-3}$ and the resolution in this experiment is $\sim 1.3 \times 10^{-3}$.

and planned instruments in terms of a quality factor defined as incident flux on sample ($\phi$) times sample volume ($v$) and detector solid angle ($\Omega$).

It is clear that such an instrument would enormously broaden the range of diffuse scattering and kinetic studies possible with neutrons - for example it would be possible to obtain a useful powder diffraction pattern in one pulse (20 ms) of SNS operation.

4.2. Single-crystal diffractometers

The method involved is time-of-flight Laue diffraction and the idea is very simple: A two-dimensional position-sensitive detector with time-of-flight analysis is used to measure a large volume of reciprocal space simultaneously /15/. The method is not simple in practice since it requires a detector of high enough spatial resolution and uniformity of response, a pulse structure sharp enough to give adequate temporal resolution and considerable capability in computer hardware and software. Nevertheless, already the method has been developed to the state where accurate crystal structure determinations have been achieved /16/ and detailed
TABLE III. COMPARISON OF POWDER DIFFRACTOMETERS

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Resolution $\Delta Q/Q(%)$</th>
<th>Quality Factor $\phi V\Omega$</th>
</tr>
</thead>
<tbody>
<tr>
<td>GPPD (Argonne)</td>
<td>0.3</td>
<td>$4 \times 10^4$</td>
</tr>
<tr>
<td>DIB (ILL)</td>
<td>0.6</td>
<td>$2 \times 10^5$</td>
</tr>
<tr>
<td>D20 (ILL)</td>
<td>0.3</td>
<td>$1 \times 10^7$</td>
</tr>
<tr>
<td>D7 (ILL)</td>
<td>3</td>
<td>$5 \times 10^6$</td>
</tr>
<tr>
<td>HRPD (SNS)</td>
<td>0.04</td>
<td>$1 \times 10^5$</td>
</tr>
<tr>
<td>High-intensity Diffractometer (SNS)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\begin{cases} (7^\circ - 25^\circ) \ (30^\circ - 60^\circ) \ (85^\circ - 95^\circ) \ (138^\circ - 150^\circ) \end{cases}$</td>
<td>$\begin{cases} 3-10 \ 1-2 \ 0.75 \ 0.6 \end{cases}$</td>
<td>$\begin{cases} 1 \times 10^8 \ 4 \times 10^7 \ 1 \times 10^7 \ 2 \times 10^8 \end{cases}$</td>
</tr>
</tbody>
</table>

calculations /17/ suggest that potentially the use of high-intensity pulsed sources like SNS should be considerably more powerful than reactors for single-crystal studies.

4.3. Small-angle (low-q) scattering

Small-angle neutron scattering ($10^{-3} < q/Å^{-1} < 1Å^{-1}$) has been one of the major growth areas of neutron scattering applications in the last decades, led by the ILL Grenoble. In all cases 2-d position sensitive detectors have been used and the great advantage of pulsed sources is again the wide $\Omega$ range covered by a single setting of the detector as a result of time-of-flight.
analysis, while the high-flux reactor instruments have a somewhat higher intrinsic count rate than the low Q SNS instrument in the narrow $0$ range obtained by a given incident wavelength band ($\Delta \lambda / \lambda \sim 10\%$) and detector setting. Two pulsed source instruments have now been used to produce high-quality scientific results: the KENS low Q instrument (see Y. Ishikawa et al.)\(^1\) situated at the end of a curved guide about 19 m from a 20K cold source and that at Argonne /3/ which is not on a guide but directly views on 80K moderator at a distance of 8 m.

The low-Q diffractometer being constructed at SNS is shown in Fig. 11 /18/. Here the sample views the $H_2$ moderator at a distance of 16 m but is removed from direct line-of-sight by means of a super mirror Soller bender which bends neutrons of $\lambda > 1$ Å through $\sim 25$ m rad so removing the fast neutrons emerging from the primary collimator. Choppers will be used to avoid frame overlap or to restrict the wavelength band of neutrons incident on the sample. The detector will be a fibre optic coded glass scintillator in five parts: an inner dartboard with annuli between 5 and 38 cm radius divided into $20^\circ$ segments, and four rings all moving on rails in an evacuated tank of 20 m length. The $0$ range available will be $0.002$ to $2$ Å$^{-1}$. No results are yet available with the SNS instrument but extensive measurements have been made with the low-Q diffractometers at Argonne and especially at KENS. An example which illustrates the considerable advantages of the wide Q-range directly available with pulsed source is given in Fig. 12, which shows the Ostwald ripening process in the crystallization of $TiO_2$ gel as a function of heat treatment obtained with the Argonne spectrometer /19/. Extrapolation of the performance of these instruments to

\(^1\) ISHIKAWA, Y., et al., IAEA-CN-46/27, these Proceedings.
the more powerful SNS indicates that this will be much better than any reactor source when measurements over a wide Q-range (like that of Fig. 12) are required as in many materials science and biological applications. Other advantages are easier separation of elastic and inelastic scattering and the study of time-dependent phenomena.

5. POLARIZED NEUTRONS

The major problem in the development of polarized neutron spectrometers for pulsed sources is the requirement for broad-band neutron polarization. Three approaches are being used but none has yet reached the stage where polarized neutron scattering experiments are being routinely performed. The TOP instrument at KENS /20/ uses a soller super-mirror system but this is limited to wavelengths longer than ~3Å and the flux is very low. Nevertheless interesting depolarization measurements have been made. The second method involves the use of dynamically polarized proton filters and two methods have been used for polarizing the protons. The Polarized
Epithermal Neutron (PEN) instrument at KENS uses an ethylene glycol + Cr filter /21/ and has produced neutron polarizations of better than 80% for E < 2 eV. Improvements are planned for removing $^3$He, used in cooling the filter, from the beam and so improving the transmission.

A novel spin refrigerator method has recently been suggested /22/ which involves the use of a spinning single crystal containing ytterbium ions whose electron spins strongly couple with the surrounding protons. Promising initial results have been obtained.

At the SNS a polarized neutron instrument (POLARIS) /23/ is being developed which uses the third method: resonance absorption polarizing filters containing $^{149}$Sm. The method is based on the preferential absorption of one spin state when neutrons pass through the filter containing polarized nuclei. The polarization thus requires the use of very low temperatures and the best results so far have been obtained using a SmCo$_5$/tin matrix permanent magnet at 12 mK /24/. These are shown in Fig. 13, which demonstrates the great promise of the method, especially, as the metallurgical problems required to be overcome to improve the performance of the filter have now been identified. The instrument in which the filter is to be used for further development of the technique and for the first applications is shown in Fig. 14.

6. INELASTIC SCATTERING INSTRUMENTS

The instruments existing or being developed on pulsed sources may cover much the same range of $\omega$ and $Q$ as for reactor instruments.
but the pulsed epithermal rich spectrum leads to important differences. For example, for cold neutron applications such as high-resolution quasi-elastic scattering the intrinsic count rates for narrow ranges of energy transfer are lower than for reactors but advantages may accrue when wider energy windows are required. Pulsed sources also have great advantages for high energy transfer applications such as magnetic excitations in solids, momentum distributions for solids and quantum liquids, vibrational excitations of light atoms in solids and excitations in liquids, glasses and amorphous solids. Examples will be given of different kinds of instruments for these studies. The question of the study of single-crystal excitations at medium or low-energy transfer using pulsed sources has received much attention but this is an area still requiring much development work before it can be determined to what extent pulsed sources can be competitive with reactors. This area was thoroughly reviewed in Ref. /3/ and no such spectrometer is yet under construction at SNS so for reasons of space no further discussion will be given here.

6.1. Direct geometry chopper and crystal spectrometers

These instruments are very similar in principle to the equivalent reactor instruments and give a very versatile coverage of \( \theta \) and \( \omega \). In fact no crystal monochromator instruments currently exist at the sources listed in Table I, but chopper instruments exist at Harwell and at Argonne, the latter especially having produced a considerable amount of excellent scientific data.
illustrated by recently published $S(Q,\omega)$ data on water /25/. Analysis of the performance of the two types of instrument show clearly that choppers are superior at higher energies /26/ (greater than about 30-100 meV depending on the detailed designs) where the crystal reflectivities become small, while at lower energies the crystal monochromator can provide much better intensities. It would clearly be useful to combine these two approaches if it can be done without too much complication. A preliminary design for such a hybrid instrument on SNS has been carried out /26/ and is illustrated in Fig. 15. It involves a vertical movement of sample area and counter bank by about 30 cm between crystal and chopper options.

For energy transfers to beyond 500 meV a spectrometer (HET) with a 1 $\mu$s burst-time, 600 Hz chopper is being installed at SNS and is illustrated in Fig. 16. It is characterized not only by high-energy transfers but high resolution ($\Delta E_0/E_0 = 0.7\%$) and a concentration on low scattering angles so as to keep $Q$ as low as possible for magnetic excitations and reduction of multiphonon scattering and recoil broadening. The $\omega$-$Q$ ranges obtained with various incident energies are shown in Fig. 17.

6.2. Inverted geometry inelastic spectrometers

Current versions of this type of spectrometer use either filter difference or crystal analyser methods. In both cases a very wide range of energy transfers are available with high count rates and a fixed final energy of about 3 to 5 meV but little or no control over momentum transfer. The fact that the high $\omega$ is necessarily associated with high $Q$ may restrict the applications, which are generally in the field of vibrational spectroscopy. At Los Alamos a Be/Be $Q$ filter difference spectrometer, which has an intrinsic
FIG. 16. The high-energy transfer spectrometer HET which views an ambient moderator on SNS.

FIG. 17. The range of energy-momentum space covered by the HET spectrometer for various detector angles and incident energies.
resolution of around 1 meV, has been operated very successfully /27/ while at KENS a crystal analyser spectrometer using a time-focused pyrolytic graphite analyser has been operated extremely successfully /28/. A similar type of spectrometer is being installed at SNS and its design is shown in Fig. 18. This is situated at 12 m from an ambient moderator using Be filters to clean up the background and remove higher orders and has an energy resolution of 2-5%. Typical data obtained with this type of crystal analyser instrument are given in Fig. 19 which shows scattering from TiH₂ measured on the CAT instrument at KENS /29/.

6.3. Inverted geometry quasi-elastic instruments

These instruments are required to have high-energy resolution, certainly < 100 μeV, preferably for a Q value of up to at least 2 Å⁻¹ for the study of quasi-elastic and very-low-energy transfer
FIG. 19. Data on TiH₂ showing five orders of the hydrogen stretching vibration measured on the CAT spectrometer at KENS [29].

FIG. 20. The high-resolution inelastic scattering spectrometer IRIS situated at the 25K H₂ moderator on SNS.

(e.g. tunnelling levels) inelastic scattering. They need to be situated on a cold source and only two such instruments currently exist. The first is the energy-focused crystal analyser LAM at KENS, which has an energy resolution of 0.1 meV and an elastic Q range of
FIG. 21. The measured resolution function for the Be/Be window detection system on IRIS.

TABLE IV. PARAMETERS OF THE IRIS DETECTION SYSTEMS

<table>
<thead>
<tr>
<th></th>
<th>Be/Be</th>
<th>Graphite</th>
<th>Silicon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Final energy/meV</td>
<td>5.22</td>
<td>1.82</td>
<td>2.07</td>
</tr>
<tr>
<td>Energy window/meV</td>
<td>6.4</td>
<td>1.2</td>
<td>0.09</td>
</tr>
<tr>
<td>$\Delta Q/\AA^{-1}$</td>
<td>0.9-3.4</td>
<td>0.3-1.8</td>
<td>0.1-2.0</td>
</tr>
<tr>
<td>Energy resolution/µeV</td>
<td>50</td>
<td>12</td>
<td>1</td>
</tr>
</tbody>
</table>

0.5 to 2.8 $\AA^{-1}$, described by Ishikawa (IAEA-CN-46/27, these Proceedings). The second is the IRIS spectrometer at SNS. Three alternative detector options are envisaged as shown in Fig. 20. The first of these constructed by the Bhabha Atomic Research Centre, Trombay, has already been installed. This utilizes a beryllium window analyser consisting of a cooled (80K) Be filter, the transmitted neutrons from which are reflected in near backscattering geometry into an annular counter by polycrystalline Be at a different temperature. When the reflector is at 300K an instrumental resolution of $\sim$50 µeV is obtained and the first measurement made with the detector in the direct beam is shown in Fig. 21. This confirms the expected resolution but also shows the presence in these first measurements of a significant inelastic background. The
The arrangement of the two detection systems being installed on the electronvolt spectroscopy beamline of the SNS.

Other two detection systems utilize backscattering reflection from silicon or graphite crystals to give the maximum possible energy resolution. The expected range of performance from the three detector options is given in Table IV which shows that IRIS will be an extremely powerful and versatile spectrometer.

7. ELECTRONVOLT SPECTROSCOPY

The relatively high fluxes of epithermal neutrons available from pulsed sources makes it possible for the first time to contemplate neutron scattering measurements with energy transfers in the region of 1 eV and higher. Two methods are under development, both using resonant interaction of neutrons to define an incident or scattered energy and the resonance widths make possible intrinsic resolutions of < 0.1 eV. A discussion of the principles and a list of possible resonances is given in Ref. /3/ and see also Refs /30,31/. The first method is an inverse-geometry method which detects the \( \gamma \)-rays emitted on neutron capture from a foil placed after the sample. This method is being used at Argonne and at Harwell. The major difficulty lies in reaching small \( Q \) values which demands low scattering angles (< 5°) where there is considerable difficulty in separating signal from background.

The second method may be used in either direct or inverse geometry; it is a difference method in which data are collected with a resonantly absorbing filter both in and out of the beam. The difference neutron time-of-flight spectra then give either the response of the sample to the resonance energy incident neutrons or, in the inverse geometry case, identify those neutrons scattered to the resonance energy. The inverse geometry is generally the better one for analysing energies up to \( \sim 1 \) eV and this method is being pursued at Los Alamos and at SNS. The filtered beam technique is
FIG.23. Inelastic scattering from ZrH$_2$ using filter difference spectrometers at
the WNR source. The Pu-240 resonance was used as an energy analyser at
(a) 20°, (b) 15°, (c) 10°. The curves are calculated from a simple harmonic
oscillator model. (d) was obtained using Sm-149 at 5° with elastic
scattering suppressed.
technically simpler than the resonance detector but its main
disadvantage is clearly that it is a difference method, although
data quality can in fact be improved by using additional filters to
remove resonance energy neutrons from the incident beam and
selectively attenuating neutrons at energies $E > E_R$ in the
scattered beam which are elastically scattered by the sample.
Because of the uncertainties about which is the better technique, at
least in the short to medium term, both resonance detector and
filter methods will be pursued at SNS as a joint project with
Harwell. The proposed arrangement of spectrometers is shown in
Fig. 22. Test measurements of the SNS prototype filter spectrometer
have in fact been made on WNR beam lines at Los Alamos and some
results on $\text{ZrH}_2$ are illustrated in Fig. 23 /32/.

8. CONCLUSIONS

With the beginning of operation of the SNS and the upgrades in
progress on the KENS, Argonne and Los Alamos sources the use of
pulsed neutron sources will certainly mature rapidly in the next
decade. The results obtained so far on relatively weak sources are
already impressive but we are only at the beginning. It can be
confidently forecast from extrapolation of what has been done
already that higher intensities and new instruments will make a
major scientific impact, but more exciting is the prospect, surely
to be fulfilled, of new developments so far unforeseen.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the considerable help received
from my colleagues at SNS in the preparation of this paper.

REFERENCES

/1/ WINDSOR, C.G., Pulsed Neutron Scattering, Taylor and Francis
    (London) 1981.
/4/ WINDSOR, C.G., in Neutron Scattering, (SKOLD, K., PRICE, D.L.,
    Eds) in series, Methods of Experimental Physics, New York
/6/ TAYLOR, A.D., Rutherford Appleton Laboratory, Rep. RAL-84-120
/7/ CONVERT, P., FORSYTH, J.B., Eds, Position Sensitive Detection
/16/ Not used.
Invited Paper

INSTRUMENTS FOR HIGH-FLUX SOURCES WITH TIME STRUCTURE OF NEUTRON FLUX

R. SCHERM
Physikalisch-Technische Bundesanstalt, Braunschweig, Federal Republic of Germany

B. ALEFELD
Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Jülich, Federal Republic of Germany

Abstract

INSTRUMENTS FOR HIGH-FLUX SOURCES WITH TIME STRUCTURE OF NEUTRON FLUX.

Most of the paper is based on the results of two workshops which were held to discuss how a proper instrumentation of the planned spallation source SNQ can make the best use of the enhanced neutron flux during the pulse. First the flux properties of the SNQ are described and an "effective" peak flux $<\phi>$ is introduced. Then a short comparison of steady state and pulsed methods and the definition of a gain factor is given. According to their operational modes the neutron scattering instruments were arranged into three classes and gain factors compared to a steady-state source with the same time-averaged flux are estimated.

1. INTRODUCTION

The concept of the spallation neutron source SNQ, which is planned in the Federal Republic of Germany, differs from that of existing spallation sources in the United States of America and Japan as well as from the SNS source in the United Kingdom, which will go into operation soon. These sources are especially designed for neutron scattering with hot and epithermal neutrons on the expanse of thermal and subthermal neutrons. It is impossible to build a source with optimum parameters for all applications and techniques of neutron scattering. For example, very short pulses for hot and epithermal neutron scattering can best be produced with a proton pulse length of the order of microseconds and with small poisoned moderators without reflectors. On the other hand, to obtain a high thermal and subthermal flux, unpoisoned moderators with reflectors should be installed. The SNQ is designed for thermal and cold neutron scattering. While the time-averaged flux is already comparable to that of a high flux reactor, its time structure allows a considerable improvement of nearly all conventional neutron scattering techniques. Many of the advantages of pulsed neutron sources have been realized long ago and an extensive treatment of the whole subject is given in the book, Pulsed Neutron Scattering.
by Windsor /1/. The prospect in having a new dimension of time-dependent thermal neutron flux was a great challenge to create new ideas to exploit the costly neutrons of this source more efficiently than is possible on a continuous source. Here we briefly describe the flux properties of the SNQ, followed by a comparison between DC and time-of-flight methods, and then a coarse gain factor is defined for instruments operated on a pulsed source over those on a steady-state source. Instruments at the SNQ will be divided in three subgroups: DC, time-of-flight and multiplexing instruments. Tentatively a gain factor is assigned to each instrument. Special techniques, which may become attractive with a pulsed source, are discussed.

2. THE NEUTRON FLUX AT THE SNQ

The most important property of the neutron flux at the SNQ is its pulse structure /2/. Two different and well-separated moderators are to be installed - a thermal H2O moderator and a cold N2 source. The shape of the neutron pulse is determined by the rectangular proton pulse of length 250 μs and the decay time τd of the neutron field in the moderator.

The resulting pulse shape is shown in the upper part of Fig. 1 for both the thermal and cold moderator. Table I shows the main flux parameters.

As a future option it is planned to shorten the proton pulses in a compressor ring to 200 ns. This will improve Φ by a factor of two in the thermal region. Much larger gains are expected for hot and epithermal neutrons. While the average flux of $1.2 \times 10^{15}$ is equivalent to that on existing high flux reactors, the flux during the pulse is enhanced up to a factor 35. If it were only the average flux Φ which counts, regardless of the pulse structure, this source would just be an expensive way of copying a reactor. The justification of this high effort is identical with properly utilizing the pulse structure. Ideally we want to invent instruments whose detectors permanently count relevant neutrons which emerge from the peak flux during the pulse. This would correspond to a steady-state source of $4 \times 10^{15}$ n·cm⁻²·s⁻¹. The pulse shapes

Table I. FLUX PROPERTIES OF THE SNQ (Φ in n·cm⁻²·s⁻¹).

<table>
<thead>
<tr>
<th>Decay time</th>
<th>Thermal moderator</th>
<th>Cold moderator</th>
</tr>
</thead>
<tbody>
<tr>
<td>FWHM</td>
<td>120 μs</td>
<td>350 μs</td>
</tr>
<tr>
<td>Time averaged flux</td>
<td>$1.2 \times 10^{15}$</td>
<td>$0.8 \times 10^{15}$</td>
</tr>
<tr>
<td>Peak flux</td>
<td>$42 \times 10^{15}$</td>
<td>$16 \times 10^{15}$</td>
</tr>
<tr>
<td>$\dot{\Phi}/\Phi$</td>
<td>35</td>
<td>16</td>
</tr>
</tbody>
</table>
3. TIME-OF-FLIGHT VERSUS STEADY STATE

Crystal diffraction and time-of-flight analysis are the conventional methods of defining $k_0$ and $k_1$ and hence the energy and momentum transfer of the scattered neutrons. Both methods have their merits and drawbacks. Here we take as an example powder diffraction, where one has to measure many Debye Scherrer lines, i.e. one scans the diffracted intensity $I(0)$ as a function of momentum transfer. In a steady-state experiment the wavelength is fixed through a crystal monochromator and the intensity, scattered in various angles $2\theta$ is registered. In a pulsed diffraction
experiment a short white pulse with a repetition rate \( v \) is produced either by a chopper or in the moderator. After some distance \( L \), different wavelengths arrive consecutively at the sample. Neutrons scattered into a fixed angle are now registered as a function of time \( t = \lambda \cdot L \cdot m / h \) which now serves to identify the wavelength \( \lambda \) and hence \( Q \). Both methods are illustrated in Fig. 2 in a \( \lambda - t \) diagram as well as in the neutron distance-time diagram.

The steady-state instrument selects a wavelength band \( \delta \lambda \) permanently, while at the pulsed instrument the wavelength at the sample varies with time. If both experiments are properly designed they are equivalent, which can be shown as follows. In the steady-state experiment the intensity is proportional to the resolution

\[
J \propto \frac{\delta \lambda}{\lambda} = \frac{\delta V}{V}
\]

In the time-of-flight method the same resolution is obtained by

\[
\frac{\delta V}{V} \propto \frac{t}{T} = \frac{t}{L} \cdot v
\]

where \( t \) is the flight time from the chopper to the detector. The available wavelength band between two pulses is given by
\[ \frac{\Delta \lambda}{\lambda} = T \frac{v}{L} \]

where \( T \) is the time between two pulses.

The intensity within one resolution element is reduced by the factor \( T/T \) but the number of available resolution elements is given by \( T/t \). Cutting down the intensity within one resolution element by chopping the neutron beam is exactly compensated for by using more resolution elements consecutively. In many practical cases only a limited part of the \( \Delta \lambda \) band contains interesting or "publishable results", a fact which we respect by defining a time utilization factor

\[ \text{TUF} = \frac{\Delta t_{PR}}{T} \]

\( \Delta t_{PR} \) is the time between two pulses where publishable results (in Phys. Rev.) are obtained. We now define a coarse gain factor \( G \) for an instrument operated at a pulsed source compared to an instrument at a steady-state source with the same time-averaged flux. The useful intensity on the sample at a DC-instrument is proportional to

\[ J_{DC} \sim \phi \cdot \delta \lambda \cdot \delta \Omega \cdot \text{TUF} \]

The same quantity for an instrument on a pulsed source using the pulse width \( \tau \) is

\[ J_p \sim \langle \phi \rangle \tau \cdot \delta \lambda \cdot \delta \Omega \cdot \Delta t_{PR}/T. \]

Requiring identical resolution \( \delta \lambda \) for both cases we obtain for \( G \)

\[ G = \frac{J_p}{J_{DC}} \sim \frac{\langle \phi \rangle \tau \cdot \Delta t_{PR}}{\phi \cdot T} \]

Here we have assumed that a DC instrument can be operated very economically with TUF = 1 and we have set \( \delta \Omega_{DC} = \delta \Omega_p \). Often the latter assumption is not quite justified. The moderator size of 10 x 10 cm², as well as the substantial 6 m shield of the target block at the SNO, reduce the brightness \( \delta F \cdot \delta \Omega_p \) compared to the reactor. A crystal instrument can further increase the intensity if a curved monochromator focuses a large beam on to a small sample. Intensity losses due to crystal reflectivity or chopper transmission are also neglected. Nevertheless, we believe that the above defined gain factor is a simple but reasonable measure for understanding the basic virtue of different types of instrument.

As a matter of fact the gain factor can never be higher than the ratio between the effective peak flux and the time-averaged flux.

4. INSTRUMENTATION OF THE SNQ

To initiate the discussion regarding how correct instrumentation of the SNQ can make use of the enhanced flux during
the pulse, two workshops were held in 1984 in Bad Kissingen and in Maria Laach /3/. Some of the ideas discussed there are also presented at this conference. The following account is mainly based on the results of these workshops. An evaluation of SNQ instruments is simplified by grouping them into classes according to their operational mode:

- DC instruments
- time-of-flight instruments
- multiplexing instruments

4.1 DC instruments

The most important representatives of this class of instruments are the triple-axis spectrometer and the two-axis crystal diffractometer. They simply use the time-averaged flux $\phi$. Because of their well-established merits for specifically investigating a "point" of $S(Q,\omega)$ (e.g., special phonons, super-reflex near-phase transitions), we expect them to survive even at a pulsed source. With the gain factor $G = 1$, SNO is just equal to a high flux reactor. But even here the pulse structure offers certain advantages. Gating the detector suppresses the background, i.e., detection of the valuable neutrons is performed between two proton pulses, where the fast background of the source is virtually zero. Certainly the background of the source can be reduced by a coarse monochromatization of the beam with phased choppers within the target shielding. Also, part of the sample background can be removed by time-of-flight separation. Different order reflections from the monochromator and analyser can be distinguished without using filters and allows $\lambda/2$ or $\lambda/3$ to be used deliberately for the measurement.

4.2 Time-of-flight methods (TOF)

The SNO is a dedicated neutron source for time-of-flight methods with the inherent restriction that the pulse width $\tau$ and the repetition rate $\nu$ cannot be matched to all different types of instrument simultaneously. This class of instruments profits fully from the effective peak flux $\langle \phi \rangle$ , which leads to the gain factor $G = \frac{\tau}{\nu \nu_{ideal}}$.

$$G = \frac{\tau}{\nu \nu_{ideal}}$$

($\nu_{ideal} \geq 100$)

The many different versions of TOF instruments can be grouped into two main methods, the direct and the inverse methods. The spectrometers of the first use a short monochromatic pulse which hits the sample, and the energy of the scattered intensity is analysed by TOF. On the other hand, in the inverted geometry a short white pulse is produced which falls on to the sample after a distance $L$, where neutrons with different velocities are separated by their arrival time. After scattering, a fixed energy is filtered out from the spectrum by Bragg reflection.
4.2.1. Direct geometry

As a prototype instrument we discuss the IN 5, a double chopper instrument /4/. It is one of the most efficient instruments at the Institut Max von Laue - Paul Langevin (ILL). The schematic operation of the instrument is shown in the time-distance diagram of Fig. 3 for a typical neutron energy of 2 meV and a resolution of 100 meV. If the flight path from the sample to the detector is 4 m, a pulse length at the sample of 80 μs is needed. This pulse is produced by the chopper CH2 which is placed 14 m from the moderator. Another chopper CH1 at a distance of 8 m from the moderator acts partly as tailcutter and a monochromator.

It can easily be seen from Fig. 3 that the effective pulse length $\tau$ at the moderator is 290 μs. From Fig. 2 an effective peak flux $<\phi>/<\delta>$ of 17 is obtained. The gain factor

$$G = \frac{<\phi> \tau \nu}{\phi \nu_{\text{ideal}}}$$

is 17 for $\nu_{\text{ideal}} = 100$ Hz and 8.5 for $\nu_{\text{ideal}} = 200$ Hz.

It should be mentioned that for this type of instrument a strong reduction of the moderator pulse length (retaining the same integrated flux) would not much improve the performance.

4.2.2. Inverse TOF

Inverse TOF instruments have proved their potentialities on continuous sources /5/, and especially on the existing pulsed
This method is schematically illustrated in Fig. 4. The velocity resolution at the sample is given by $\frac{\delta v}{v} = \frac{v \cdot \tau}{L}$ where $\tau$ is the flight path from the moderator to the sample. The velocity band between two pulses is $\Delta v/v = T \cdot v/L$. Hence the number of resolution elements within the available velocity band is given by $T/\tau$. This number amounts to 38 or 25 if the pulse length at half maximum at the thermal or cold moderator, respectively, is used. In many cases the scan range over 38 and/or 25 resolution elements is already too large, so the most efficient way to operate an inverted TOF instrument is to use the whole pulse length of the source. However, for high resolution instruments there are practical difficulties. For example, an inverted TOF instrument with the resolution of the backscattering instrument of $\delta v/v = 10^{-4}$ needs a flight path of 2.2 km ($\tau = 350 \mu s$ and $v = 630 \text{ m/s}$). This length is at least by a factor of 10 far from any practical considerations. If many more resolution elements than 38 or 25 can be used with a large time utilization factor, the pulse length should be reduced, together with a proportionate reduction of the flight path. This is certainly recommended for powder diffraction. A practical difficulty should be mentioned at this point. A chopper at some distance from a pulsed source acts as a monochromator. To obtain the full wavelength...
FIG. 5. The modified backscattering instrument MUSICAL. At a pulsed source the velocity drive with one monochromator is replaced by a multi-single-crystal monochromator (insert) in which consists of thin silicon slices with slightly different lattice spacings and a distance $l$ between the crystals.

band between two pulses at the detector, the distance between the chopper and the moderator may not exceed a maximum value, which in special cases may be as small as 2 m. Apart from the fact that chopper positions must be foreseen at a very early stage of design, considerable heating and safety problems may arise.

4.3 Multiplexing instruments

Conventional DC methods like small-angle scattering, spin-echo spectroscopy and backscattering can be operated on a pulsed source in a multiplexing mode of operation by using a wider wavelength band than in the DC mode without the resolution deteriorating, since the different wavelength can be separated by time-of-flight. The gain factor for these types of instrument depends on the desired resolution. For low-resolution small-angle scattering, as well as for quasi-elastic spin-echo spectroscopy, a primary velocity resolution of 10% is adequate. At a distance of 15 m from the moderator one obtains a wavelength band of 40% for a velocity of 600 m/s. This means that a gain of about 4 can be expected. The gain rise is proportional to the improvement of the desired wavelength resolution, reaching asymptotically $\frac{\phi}{\phi}$. A special kind of multiplexing was proposed for the backscattering
FIG. 6. Energy-time diagram of the MUSICAL instrument. The different energies can be separated by time-of-flight.

instrument /3,9/. This spectrometer, also called "MUSICAL", is sketched in Fig. 5. Apart from the sophisticated monochromator the instrument is very similar to IN 10. On IN 10 the monochromator is one silicon crystal, mounted on a velocity drive. At the MUSICAL spectrometer the monochromator consists of several silicon crystals with the same orientation but with lattice spacings which differ by one Darwin width, i.e. $2 \times 10^{-5}$ from each other. The crystals are mounted at a certain distance between each other. Therefore, differently coloured neutrons arrive at different times leading to a simple relation between the arrival time and the energy transfer as is shown in Fig. 6. The gain factor $G$ of MUSICAL is proportional to the number of crystals of the monochromator. The optimum number of crystals is given by $n_{opt} = T/2 \tau = 13$ (for a pulse length of 350 $\mu$s). Finally, the gain with 13 crystals is given by $G = \text{TUF} \cdot <\delta>/\delta$. For most types of experiment TUF can be set equal to one, so finally $G$ is obtained at about 15.

5. SPECIAL METHODS

Recently a number of futuristic experimental techniques on a pulsed source were discussed. Probably the most fascinating technique is the dynamic or time-dependent energy analysis of the scattered neutrons, since this method may raise the gain factor of the scattered neutrons considerably in many experimental situations. Three quite different methods have been proposed up to now. The Drabkin device is proposed by Badurek and Schmatz for dynamical energy analysis of polarized neutrons /3/. The second example is the diffuse scattering instrument proposed by Schelten and Bauer /3/. The third example is a vibrating crystal on an
FIG. 7. The Doppler Instrument BALDI /12/. A white neutron beam passes the graphite crystal and hits the Doppler crystal which is mounted on a wheel with two arms. The wheel rotates with a frequency of 50 Hz and is synchronized to the source. The back-reflected neutrons are deflected by the graphite crystal to the sample. The rest of the spectrometer is conventional.

inverted TOF instrument proposed by Schelten and Alefeld /3/. Our feeling is that this field of experimental innovations for neutron scattering on a pulsed source is far from exhausted. Schelten and Alefeld have proposed a phase space transformation with a moving mosaic crystal in order to increase the divergency of a monochromatic neutron beam /3/. Finally, we would like to describe briefly a method to increase the intensity of a pulsed thermal neutron beam. If the moderator temperature is optimized for each velocity, then the intensity within the velocity interval $\Delta v$ around $v$ is proportional to $v^{-3}$. If neutrons are first cooled to a low velocity $v_1$ in a cold moderator and then shifted by a Doppler moving crystal to the velocity $v_2$, a theoretical gain of $(v_2/v_1)^3$ is expected. For example, if neutrons are accelerated from 800 m/s to 1800 m/s the intensity is raised by a factor of /11/. Because it is impossible to build an optimum moderator for each velocity the gain is probably higher on a real neutron source. If there exists only a cold moderator with $v_T = 800$ m/s ($T = 40$K) and a thermal moderator with $v_T = 2400$ m/s ($T = 350$K), a theoretical gain factor of 25 can be expected. Taking into account
Table II. GAIN FACTORS OF DIFFERENT TYPES OF INSTRUMENT /3/.

<table>
<thead>
<tr>
<th>Mode of operation</th>
<th>Type of instrument</th>
<th>Gain</th>
</tr>
</thead>
<tbody>
<tr>
<td>DC</td>
<td>Triple-axis spectrometer</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Two-axis spectrometer</td>
<td>1</td>
</tr>
<tr>
<td>TOF</td>
<td>Powder diffraction</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>Single-crystal diffraction</td>
<td>3 - 35</td>
</tr>
<tr>
<td></td>
<td>Direct TOF, thermal</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>$v_{\text{ideal}} = 250$ Hz</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Direct TOF, cold,</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>$v_{\text{ideal}} = 100$ Hz</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Inverted TOF, MAX, thermal</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Inverted TOF, MAX, cold</td>
<td>12 - 15</td>
</tr>
<tr>
<td>Multiplexing</td>
<td>Small-angle scattering</td>
<td>2 - 20</td>
</tr>
<tr>
<td></td>
<td>Diffuse scattering</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Spin echo</td>
<td>3 - 6</td>
</tr>
<tr>
<td></td>
<td>MUSICAL</td>
<td>15</td>
</tr>
</tbody>
</table>

reflectivity losses of the Doppler crystal of 50%, and the fact that the peak flux at the cold moderator is lower by about a factor of 2 than at the thermal moderator, there should remain a gain factor of about six. This method has not been used on a steady-state source up to now; however, it may become very attractive on a pulsed source. A sketch of a TOF instrument with a Doppler wheel is shown in Fig. 7.

6. CONCLUSION

We did not intend to discuss in detail all possible instruments on a pulsed source. In particular, those instruments which have shown their great potential on existing pulsed sources, such as the MAX spectrometer at KENS, or several instruments for elastic and inelastic scattering at Argonne and Los Alamos, have not been dealt with. The importance of very high quality neutron guide tubes for the SNQ should at least be mentioned. We also believe that a method by Hossfeld /10/ - the combination of pseudo-random choppers operating asynchronously to the moderator pulse - could help very much to reduce or even supress the influence of the large nuisance background of the classical method.

Table II gives a list of possible instruments for the SNQ, and the gain factor assigned to each of them. Our personal feeling is that a pulsed source with the SNQ parameters is comparable to a steady-state reactor with $10^{16}$ n·cm$^{-2}$·s$^{-1}$. This statement is equivalent to an evaluation of pulsed sources by Maier-Leibnitz /11/ thirteen years ago.
ACKNOWLEDGEMENT

This paper is based on the work of many people and the discussions with them. We are grateful to all of them, in particular to those who contributed to the SNO workshops, which are the basis of this report.

REFERENCES

/5/ STEICHELE, E., ARNOLD, P., Phys. Lett. 44A (1973) 165-
/8/ Los Alamos Rep. LALP 83-17, IS 1-73-83.
THE SPALLOATION NEUTRON SOURCE AT THE RUTHERFORD APPLETON LABORATORY

D. A. GRAY
Rutherford Appleton Laboratory,
Chilton, Didcot, Oxon,
United Kingdom

Abstract

THE SPALLATION NEUTRON SOURCE AT THE RUTHERFORD APPLETON LABORATORY.

The paper reports progress and plans for commissioning the Spallation Neutron Source, based on an 800 MeV 200 μA mean current synchrotron with a depleted uranium target, giving 0.4 μs proton pulses at 50 Hz. Commissioning of the synchrotron has, in a very short time, given close to 10% of the design current per pulse and this has been extracted successfully from the synchrotron. It is planned to produce the first neutrons during 1984 and to start a sustained run for neutron scattering research in about April 1985.

1. INTRODUCTION

The Spallation Neutron Source (SNS), which is the world's most powerful accelerator-based pulsed neutron source under construction, was approved in 1977, is planned to produce its first neutrons during 1984, to be in regular operation during 1985 and can confidently be expected to be a major contributor to neutron scattering research in the 1990's.

2. DESCRIPTION OF THE SNS

Some characteristics of the SNS are given in Table I. A more detailed description is given in (1) with updating in (2) and (3). Fig 1 shows the layout of the SNS. Intense 0.4 μs pulses of protons are accelerated to 800 MeV in the synchrotron of the SNS and are then guided through the extracted proton beam channel to the target station. In the target, made of plates of depleted uranium clad with zircaloy, 25 to 30 neutrons with MeV energies are produced. Above the target there are 2 ambient temperature water moderators and below there are 2 moderators, one of methane at 95°K and one of supercritical hydrogen at 25°K. The fluxes and the pulse lengths expected from the moderators are given in (4). The target/moderator assembly is surrounded by a reflector of beryllium rods cooled by D₂O. The whole assembly is supported at the centre of a stainless steel vessel some 3m in diameter filled with helium.
Table I: CHARACTERISTICS OF THE SNS

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Final proton energy</td>
<td>800 MeV</td>
</tr>
<tr>
<td>Final proton current</td>
<td>200 μA mean</td>
</tr>
<tr>
<td>Pulse repetition rate</td>
<td>50 Hz</td>
</tr>
<tr>
<td>Number of protons per pulse</td>
<td>$2.5 \times 10^{13}$</td>
</tr>
<tr>
<td>Proton pulse structure</td>
<td>2 x 100 ns pulses centres separated by 300 ns</td>
</tr>
<tr>
<td>Target heat deposition</td>
<td>225 kW</td>
</tr>
<tr>
<td>Number of fast neutrons</td>
<td>$3 \times 10^{16}$ per second</td>
</tr>
<tr>
<td>Number of neutron holes</td>
<td>18</td>
</tr>
<tr>
<td>Injector</td>
<td></td>
</tr>
<tr>
<td>Pulse length</td>
<td>400 μs</td>
</tr>
<tr>
<td>Pulse current</td>
<td>20 mA</td>
</tr>
<tr>
<td>Synchrotron</td>
<td></td>
</tr>
<tr>
<td>Number of magnet superperiods</td>
<td>10</td>
</tr>
<tr>
<td>Mean radius</td>
<td>26m</td>
</tr>
<tr>
<td>Bending radius of dipole magnets</td>
<td>7m</td>
</tr>
<tr>
<td>Magnet power supply</td>
<td>Biassed 50 Hz sinusoidal</td>
</tr>
<tr>
<td>Vacuum chamber</td>
<td>Ceramic in magnets, stainless steel elsewhere</td>
</tr>
<tr>
<td>Injection scheme</td>
<td>H– stripped to protons in the synchrotron with orbit control using 4 dipoles pulsed during injection</td>
</tr>
<tr>
<td>No. of RF accelerating stations</td>
<td>6</td>
</tr>
<tr>
<td>Frequency of RF</td>
<td>1.3 - 3.1 MHz</td>
</tr>
<tr>
<td>Extraction</td>
<td>Vertical with 3 fast kicker magnets of rise time 200 ns giving 14 mr deflection</td>
</tr>
<tr>
<td>Extracted proton beam line</td>
<td>150m approx</td>
</tr>
<tr>
<td>Distance from extraction point to neutron station</td>
<td></td>
</tr>
<tr>
<td>Distance from intermediate transmission target to neutron target</td>
<td>20m</td>
</tr>
<tr>
<td>Target station</td>
<td></td>
</tr>
<tr>
<td>General thickness of shielding</td>
<td>4.3m (thicker in forward direction)</td>
</tr>
<tr>
<td>Outer radius of main shield</td>
<td>5.9m</td>
</tr>
</tbody>
</table>
Surrounding the vessel is the biological shield through which there are 18 neutron beam holes. Each neutron hole can be shut off by a 2m thick steel shutter supported within the shield. Collimation of the neutron line can be tailored within the shutters and through the remainder of the shield by hand-stacking in 'insert boxes' set into the permanent shield.

A portfolio of instruments has been designed, of which some 6 partial but engineered instruments and 3 development instruments are expected to be available at the beginning of regular operation.

A large amount of equipment existed from previous accelerators in the UK (Nimrod and Nina). The facility has been built in existing buildings with some relatively small modifications. It is estimated that a facility which would cost about £100M on a 'green field' site has been built for approximately £50M. The mean power in the 800 MeV proton beam is designed to be 160 kW. Control of beam loss in the synchrotron has therefore been of primary concern in the design. It is estimated that no more than 1% of the 800 MeV beam can be lost if induced activities are to be kept to a level such that conventional maintenance techniques can be used. A system for collecting 50% of the injected beam, likely to be lost between 70 and 90 MeV, has been installed.
There is also a large amount of diagnostic equipment including provisions for measurement of beam position vertically and horizontally, beam profile in the vertical and horizontal planes, bunch shape and betatron frequencies.

3. COMMISSIONING HIGHLIGHTS

Excellent progress has been made with comparatively little operating time. A timetable of the most significant achievements is as follows:

Jan 84 - 1 μs long 70 MeV beam pulses injected into the main ring (with dc excitation of the magnets) and several hundred turns circulated at the first attempt.

Mar 84 - 0.8 μs, 70 MeV beam pulses successfully circulated with ac + dc excitation of the main ring magnets.

Apr 84 - 10 μs beam pulses successfully accelerated at the first attempt to 140 MeV using 2 rf cavities. With longer injected beam pulses the maximum accelerated beam intensity was $2.8 \times 10^{12}$ protons per pulse (ppp) or about 10% of full intensity per pulse.

Jun 84 - $1.2 \times 10^{12}$ ppp accelerated, at the first attempt using 4 rf cavities, to an energy of 550 MeV.

Sep 84 - $1.5 \times 10^{12}$ ppp successfully extracted into a graphite beam dump, again at the first attempt.

A major problem before the extraction system was commissioned in September was that it was not possible to have a beam dump so that all accelerated beam was lost at unknown points in the magnet ring. This meant that beam operating time had to be severely limited to avoid activating the accelerator and delaying installation work on the remainder of the facility. The pulse repetition frequency was also restricted to 1 Hz or less for the same reason.

Approximately $10^{13}$ ppp have been successfully injected into the SNS as a 4.5 mA, 400 μs long beam (Fig 2). This should be enough to produce an accelerated beam intensity of at least $5 \times 10^{12}$ ppp or 20% of the SNS full intensity. The maximum intensity accelerated to 550 MeV to date is about $2 \times 10^{12}$ ppp, the beam being limited by instabilities. The extracted beam pulses at 550 MeV are shown in Fig 3.
FIG. 2. 70 MeV protons building up during the 400 µs injection period at the end of which there are $10^{13}$ protons circulating.

FIG. 3. A composite showing the two 550 MeV 100 ns proton pulses in the synchrotron and their detection in the extracted proton channel after firing of the extraction kicker magnets.
4. PLANNED PROGRAMME OF COMMISSIONING

It is expected that by the time of the Conference the SNS will have produced its first neutrons. This will require the completion of the extracted proton beam line. It is planned to have the depleted uranium target filled with static D\textsubscript{2}O, the 2 ambient moderators, the 2 cryogenic moderators with circulating cryogenic fluids, and the reflector all available in the target station with the biological shield completed to the stage where about 5 x 10\textsuperscript{11} protons per pulse at 1 Hz can be put on to the target for a limited period.

It is planned to make neutron measurements in 6 neutron beam lines, in most cases using the installed neutron spectrometers. Neutron flux, spectra and time structure will be measured for the 4 moderators.

In the period following this commissioning run, the target station will be completed and engineering runs carried out on accelerator equipment to bring the repetition rate towards 50 Hz.

It is planned to start a long scheduled run for the neutron scattering programme in April 1985 with an aim of having at least 10% of the final performance which will make the SNS the most powerful operational accelerator-based pulsed neutron source.

Acknowledgement

This paper reports on the work of about 300 individuals. As Project Leader I take the opportunity to record my appreciation of all the efforts put in to achieve success for the SNS.

REFERENCES

THE SNQ TARGET AND MODERATOR SYSTEM FOR AN OPTIMIZED NEUTRON ECONOMY

G. BAUER*, H. CONRAD**, D. FILGES***, H. SPITZER**

* Projekt Spallations-Neutronenquelle
** Institut für Festkörperforschung
*** Institut für Reaktorentwicklung
Kernforschungsanlage Jülich GmbH, Jülich, Federal Republic of Germany

Abstract

THE SNQ TARGET AND MODERATOR SYSTEM FOR AN OPTIMIZED NEUTRON ECONOMY.

By taking advantage of the particular features of the spallation process for generating neutrons, and viewing the neutron source and the scientific instruments as a system to be optimized, it became possible to work out a design which, starting from only one third of the number of neutrons produced per second in the Grenoble high-flux reactor, will supply useful scientific information at a rate at least ten times higher. This is achieved by careful choice of materials and geometry in the target-moderator-reflector region, adopting a philosophy that provides a good time structure of the neutron flux at a high time average level and by a careful balance between the needs of different types of experiments. The most prominent design features of the source are: a slab-type target shape; a grooved moderator of high slowing-down density and with a fast reflector; no absorbing material close to the moderators (apart from the target itself); a pulse duration of the proton beam matched to the natural decay time of the neutron field in the moderators; a high degree of flexibility in arranging instruments around the source and matching the beam characteristics to individual needs.

1. Neutron flux and source characteristics

In the modern high flux research reactor at the Institute Laue Langevin (ILL) in Grenoble, about $4 \times 10^{18}$ fast neutrons per second are produced by nuclear fission with 57 MW of heat generated at the same time. Obviously, progress by orders of magnitude in terms of neutron generation is not to be expected.

In the SNQ concept [1], the number of neutrons produced per second is even lower: $1.4 \times 10^{18}$ per second. The fact that a comparable time-average thermal flux can be obtained in the moderator is due to the special design options one has with a spallation neutron source. Another option that can be exploited in the spallation neutron source is the time structure of the neutron flux. Detailed studies [2] have shown that, as a rule of thumb, the effective gain in neutrons carrying useful information at the detector of an experiment is between $1/3$ and $1/2$ of the peak-to-average flux ratio at the moderator. The design
goals for the target-moderator-reflector-beam tube arrangement of a spallation neutron source are therefore threefold:

1. With a given proton beam power, to produce as many neutrons as possible in as small a volume as possible.
2. To design for efficient moderation and leakage of moderated neutrons into the beam ports.
3. To provide enough flexibility to optimize the arrangement and cross section of the beams to accommodate the largest possible number of instruments with a minimum of mutual interference.

2. The SNQ arrangement of beam tubes

The biological shield of the SNQ target will have 12 penetrations leading into the target hall and 2 penetrations, each equipped with 6 neutron guides, leading into an adjacent guide hall. While multiple use of guide tubes is standard, special provisions are made for flexibility in using the thermal beam ports. As shown in Fig. 1 the 12 penetrations leading into the target hall are each equipped with a 2 m diameter rotating shutter in such a way that installation of beam-shaping devices is possible from the top of the shielding when the shutter is closed. These devices range from collimators which can provide beams from very small to very large outer cross-section and divergence to beam splitters which allow the extraction of more than one beam with horizontal or vertical separation from any beam port.

To retain maximum flexibility in beam shaping to meet everyone's needs, provisions have been made for the installation of choppers, tail cutters, filters and other beam-shaping devices inside the biological shield.

3. The thermal neutron source

3.1 The moderator

The moderator designed for the thermal neutron source is located below the target. It is of H2O because this has the highest slowing-down density for fast neutrons and is easily pumped and handled for heat removal.

The moderator (see Fig. 2) has a basal plane of 25 x 16 cm² with the long side extending parallel to the direction of the proton beam. It consists of a central part, 5 cm wide and completely filled with water and 6 cm wide fins, 1 cm thick and separated by 1 cm extending to both sides of the moderator in the directions of the beam ports. This has the following advantages:
FIG. 1. Schematic vertical (top) and horizontal (bottom) section through the SNQ target block. (a) Rotating slab target; (b) bundle of six neutron guides; (c) wells for chopper installations; (d) access port for shutter inserts; (e) disc-shutter with exchangeable inserts; (f) H$_2$O-moderator for thermal neutrons; (g) D$_2$-source for cold neutrons; (h) reflector for cold neutron source (graphite); (i) recess with flexible arrangement of shielding material. In the horizontal section only four of the 12 disc-shutters are shown.
3.2 The reflector

Lead was chosen as a reflector material for the thermal neutron source, because it has good scattering properties and does not moderate neutrons. Relative to a Be-reflector lead gives a marginal advantage in terms of neutron intensity (see Fig. 3) but results in a cleaner pulse shape as can be seen from Fig. 4. The reflector is designed to surround the moderator as closely as possible and to leave open only a section necessary for the beam tubes to view the grooved surface of the moderator. Technically, the water flowing through the moderator is also used to cool the reflector.
FIG. 3. Experimental data obtained with a graphite analyser crystal at 90° scattering angle for the integrated and peak intensity of the Bragg reflections for various target-moderator-reflector combinations. The term 'flat' refers to a moderator of optimized thickness with no grooves on its surface. Grooved moderator as in Fig. 2.

3.3 The source characteristics

The SNQ thermal neutron source is characterized by a decay constant of its neutron field of about 100 - 120 μs. For a proton pulse of 250 μs duration this leads to a shape of the thermal neutron pulse as shown in Fig. 5. Without a pulse compressor for the protons, the width of the neutron pulse is about 250 μs in the whole spectral range, leading to a peak-to-average flux ratio of 35 in the thermal regime and approaching 40 as the pulse shape becomes more rectangular for the epithermal energies. Since the lifetime of neutrons in a given
FIG. 4. Bragg reflections from a graphite analyser crystal at 90° scattering angle shown in semilogarithmic plot for a reflector of Pb and Be around a polyethylene moderator (roughly equivalent to H₂O). The poor separation of the peaks is due to the short flight path of only 5.53 m used in the experiment.

energy interval decreases rapidly with increasing neutron energy in the epithermal regime due to the slowing-down process, much higher peak-to-average flux ratios can be obtained there by use of a proton pulse compressor as planned as an optional supplement to the SNQ-facility.

It should be noted, however, that the question whether a uranium target can be used in this case needs further investigation.

The shape of the neutron spectrum in the thermal regime can be represented reasonably well by a Maxwellian distribution for the SNQ H₂O moderator, as shown in Fig. 6, where measured data are compared to a Maxwellian distribution convoluted with the time behaviour of the source pulse. The effective moderator temperature deduced from this fit is Tₘ = 316 K.

Unlike in pulsed neutron sources [3], no decoupling or poisoning is provided for in the concept of DIANE, as the SNQ neutron source is called. Such a decoupling and poisoning would at best make any sense in conjunction with a pulse compressor and, even then, it would affect the time-average neu-
FIG. 5. Time-dependency and relative intensities expected for the neutron pulses of the room-temperature H$_2$O moderator and the cold D$_2$O source with graphite reflector. The peak-to-average flux ratio is 35 and 20, respectively. The time average cold flux is 66% of the time average thermal flux.

electron flux very seriously, as can be seen from Fig. 7. This is an obvious disadvantage if many of the instruments rely on a high time-average flux as is at present the case.

4. The Cold Neutron Source

4.1 The moderator and reflector system

Investigations to optimize the SNQ cold neutron source are still going on. The present concept is to have a cylindrical source with 30 cm diameter of liquid D$_2$ with an H$_2$O-premoderator facing the target and embedded in a graphite reflector. The design has been proposed on the basis of recent experimental investigations. It replaces the older one, which had a D$_2$O tank as reflector, because of the superior time characteristics that can be expected and which was found to be of great interest to the users even in the regime of cold neutrons [2].
4.2 The source characteristics

The expected time constant of this source is 350 µs and its average flux is about 66% of that of the thermal neutron source. The peak-to-average flux ratio will be about 20 (cf. Fig. 5). The effective moderator temperature has not yet been determined for the D₂ source with graphite reflector. Therefore, no spectral distribution is given here.

5. The Target

The target material will be a high uranium content alloy because it was found that depleted uranium yields about two times higher neutron intensity than lead (or tungsten), as can be seen from Fig. 3. The target has slab geometry with the large area placed horizontally. This has the following advantages:

(1) Neutrons produced in the relatively small reaction zone where the beam hits the slab are scattered from the adjacent material and will leave the target predominantly in the directions where the moderators are placed. The enhancement factor was found to be about 1.4, as can be seen from Fig. 8. The minimum width of the slab necessary for this effect is 30 cm and the optimum target thickness is 8 - 10 cm.
By the same token the material at both sides of the reaction zone provides an efficient shielding against fast neutrons which might otherwise enter the beam tube. In an arrangement where the beam tubes look at a slab moderator and see the target behind it, the fast neutron background at the exit of the beam tube is 3 orders of magnitude higher, as shown in Fig. 9. This compares to a potential gain of a factor of 2 in thermal neutron leakage for this geometry.

By forming the slab as a disc of large diameter (2.5 m in the SNQ-case) and rotating it around its axis perpendicular to the proton beam, it becomes possible to cope with the enormous heat density in the reaction zone. Calculations have shown [4] that in a target of depleted uranium the power dissipation at SNQ beam parameters is 10 MW time average with a local peak density up to 15 kW/cm³, running as high as 600 kW/cm³ during the 250 µs pulse. This high power density can only be managed in a moving target which is heated during one pulse only and then cooled during a full period of revolution (2 s in the SNQ design). Although not directly related to the optimization of the neutron economy, this is an important factor because it allows operation of the source during a reasonable period of time. The expected lifetime of the SNQ-target exceeds 12 000 hours at full beam. Another advantage of the target rotation is that those delayed neutron precursors which decay more than 10 ms...
FIG. 8. Optimization of dimension of slab target. (a) Thermal neutron leakage from a moderator placed above the target as a function of the lateral width of the target with the target thickness as a parameter. (b) Plot of values from (a) for a slab width of 300 mm as a function of target thickness (curve 1). Curve (2) gives the relative background of fast neutrons for the same targets (not to scale with thermal neutrons.).

FIG. 9. High-energy neutron flux at the end of a 6 m beam tube for (1) the SNO target-moderator-beam tube arrangement; (2) a geometry where the beam tube views a moderator in front of the target, yielding a gain in thermal neutron flux of a factor of two.
after the pulse are not close to the moderators when emitting their delayed neutrons. In addition to the overall dilution of delayed neutron precursors by a factor 200 (circumference of the rotating target to width of the primary reaction zone) this helps to keep the delayed neutron background between pulses negligibly low.

The primary reaction zone in the target is determined by the effective width of the proton beam (4 cm) and the depth, at which most of the neutrons have been generated. As can be seen from Fig. 10, this is about 25 cm for a uranium target with the necessary dilution for cladding and cooling. Obviously the size of the moderator (see above) is well matched to the active source area at the target surface.

6. The System

The SNO thermal and cold neutron sources have been designed and optimized under continuous interaction with the developing ideas about instrument design. These instruments, the design philosophy of which is described in a different paper [5], together with the neutron source, form the system to be optimized for economic deployment of useful information on the properties of matter. The fact that, relative to the best existing reactor, only one third of the number of neutrons are originally produced, but are expected to yield more than an order of magnitude more useful information, shows that, so far, some success is to be seen in this effort.
ACKNOWLEDGEMENT

The work reported here is the result of an ongoing collective effort of a large number of people working together to achieve one common goal. This includes staff from inside the KFA Jülich as well as from other institutions. Discussions with many people from research centres and universities have been extremely helpful in promoting this work. The experiments were performed at the proton accelerators at CERN (Geneva, Switzerland), SIN (Villigen, Switzerland) and LNS (Saclay, France). The authors are particularly grateful to the management and technical staff of these laboratories for their active support.

REFERENCES


LANDER, G.H., CARPENTER, J.M., IAEA-CN-46/12, these Proceedings.


RESONANCE DETECTION METHODS IN SPECTROMETERS

N. WATANABE
National Laboratory for High Energy Physics,
Oho-machi, Thukuba-gun, Ibaraki, Japan

Abstract

RESONANCE DETECTION METHODS IN SPECTROMETERS.

Resonance detection methods in combination with pulsed spallation neutron sources make electronvolt neutron spectroscopy possible, which can enlarge the experimentally accessible range of energy and momentum transfer by a factor greater than 10, compared with conventional techniques using a reactor. This paper describes these methods, the present status of instrument development and actual performance, with some measured results. Present performance is already good for measurements of the momentum distribution of atoms where large momentum and energy transfer are important, but not sufficient for experiments in which high-energy transfer must be measured at low momentum transfer, as in the case of high-energy magnetic excitations, mainly because of poor signal-to-background ratio or sensitivity. Significant improvements are necessary, and such efforts are in progress at various laboratories.

1. INTRODUCTION

A resonance instrument is defined as a spectrometer which utilizes sharp nuclear resonances to define the neutron energy before or after scattering. Figure 1 shows the basic principle of such spectrometers. When a foil filter including resonance nuclei is placed in the incident flight path (position (A)), resonance interactions remove neutrons at the resonance energy $E_r$ from the incident beam and the scattered neutron energy $E_f$ is determined by time-of-flight. The difference between spectra measured with and without the filter gives the scattered neutron spectrum for fixed initial energy. We call this instrument a Resonance Filtered Beam Spectrometer (RFBS)/1,2/. But, if the foil is placed in the scattered beam (position (B), an inverted geometry of the RFBS is realized and this is called a Resonance Filtered Detector Spectrometer /1-4/. In the Resonance Detector Spectrometer (RDS) /5-10/, neutrons scattered to $E_r$ are detected through prompt capture gamma rays (or other secondaries) produced by resonance interactions in the foil placed at position (c), and the incident neutron energy distribution is determined by time-of-flight.

There are merits and demerits for each of these methods /11,12/. The RDS can yield better statistics than the other two, which is the major advantage of this instrument because the desired spectrum is obtained directly on the RDS, while it is obtained from a difference spectra on the other two. By contrast, the RFBS and the RFDS are
very simple to build and are easily adaptable to a multi-counter layout, while the RDS requires a more sophisticated detector system and shielding effective for both neutrons and gammas /10/. Figure 2 shows an example of an RDS with heavy shielding /10/. Polarization of resonance foils is possible in all three cases /13,14/.
TABLE I. SOME IMPORTANT CHARACTERISTICS OF TYPICAL RESONANCE FOILS /15/

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Resonance energy $E_r$(eV)</th>
<th>Intrinsic peak cross-section $a_0$(b)</th>
<th>Peak cross-section Doppler broadened width at 300K at low temp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{149}$Sm</td>
<td>0.872</td>
<td>57.9</td>
<td>21 170</td>
</tr>
<tr>
<td>$^{165}$Ho</td>
<td>3.92</td>
<td>85.0</td>
<td>9 510</td>
</tr>
<tr>
<td>$^{181}$Ta</td>
<td>4.28</td>
<td>57.0</td>
<td>23 800</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>6.67</td>
<td>27.5</td>
<td>21 790</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>1.056</td>
<td>33</td>
<td>172 000</td>
</tr>
</tbody>
</table>

* See Ref. /1/

2. ENERGY RESOLUTION

The resonance foil determines the major contribution to energy resolution in resonance instruments. Some important characteristics of typical resonance foils are summarized in Table I. The best resolution ever achieved is 53.5 meV by $^{238}$U at low temperature /15/. Resolution better than the intrinsic resonance width can never be achieved by cooling, because Doppler broadening exists even at zero temperature. Some improvements are possible by taking difference spectra using two foils of different thickness or at different temperatures /15,16/. The expected resolution by this method is about 37 meV /15/. Much improved resolution may be realized by a polarized RDS in combination with the basic idea of spin precession energy analysis /17/.

Resolution broadening caused by the finite extent of the source, sample and detector was studied /18/, and a time-focusing method was proposed /18/ and confirmed experimentally /19/.

3. DETECTOR SYSTEM AND SIGNAL-TO-BACKGROUND

In the RFBS and the RFDS, one can utilize the usual neutron sensitive detectors. In the RDS, two detectors are most commonly used for $\gamma$-ray detection: a bismuth germanate crystal (BGO;= Bi$_4$Ge$_3$O$_{12}$), and a high purity Ge detector (HPGe) /10,12,20/. The former can detect high-energy gammas with high efficiency and the latter can select gammas at a specific energy.

Typical performance of the RDS using a half-inch$^1$ thick BGO detector with a $^{181}$Ta foil in front of the BGO was such that the detection efficiency was about 0.5 counts per capture event, with a

$^1$ 1 in. = 2.54 cm
threshold equivalent to 2 MeV, and signal-to-background ratio (S/N) (which strongly depends on sample, scattering angle and shielding) of about 2.5 for graphite and 12 for a bismuth sample at a scattering angle of 90° /9,10/.

A HPGe detector has good energy resolution and hence can selectively detect strong low-energy characteristic X-rays produced by internal conversion in the neutron capture process /20/. A 5-mm-thick HPGe detector was compared with a 5-cm-thick BGO detector by measuring 90° scattering from an aluminium sample /20/. The results indicate that the HPGe detector gives a better S/N by a factor of about two over the BGO detector.

To improve S/N in the RDS with a BGO detector at low scattering angles (1.5-3°), a ring detector system, which consists of eight 1-in.-diameter BGO crystals has been developed and tested /21/. For a thick polyethylene sample, an S/N of about 2.3, obtained by simple lower-level discrimination, has been improved to about 15 by taking more than two coincidences, sacrificing counting efficiency by a factor of 15-20. This may correspond to a sensitivity of about 0.1 b/sr if one can assume that inelastic peaks larger than S/N = 0.2 are measurable. Direct activation of the BGO detector by fast neutrons may occur, and therefore tests of a massive chopper which can suppress the fast neutron burst are encouraged to reduce the background level further /22/.

To improve the statistical problem in the RFDS, use of fixed energy selective filters was proposed and tested /4/.

4. EXPERIMENTS AT LARGE Q

Neutron scattering at very large momentum transfer (say \( Q < 100 \, \text{Å}^{-1} \)) is essential to determine the momentum distribution of the scattering particle. Several experiments have been successfully completed /1,23-26/. One of the most exciting examples is the measurement of the Bose-condensate fraction \( \alpha_0 \) in the superfluid \(^4\text{He}\). As an example, Fig. 3 illustrates a very high Q neutron scattering \((Q \sim 150 \, \text{Å}^{-1})\) from liquid \(^4\text{He}\) performed with an RDS /26/. The 6.67 eV resonance in \(^{238}\text{U}\) was used for defining the scattered neutron energy at a scattering angle of 167°. This corresponds to a recoil energy of about 11 eV. In the spectrum for He-II, we can recognize a distinct condensate peak at the central part of a broad spectrum corresponding to the momentum distribution of the normal component in He-II.

5. EXPERIMENT AT LARGE \( \mathcal{K}_\omega \) WITH SMALL Q

To measure high energy excitations, low Q experiments (say \( Q < 5 \, \text{Å}^{-1} \)) become essential, especially in magnetic systems. The kinematic constraint requires high-energy incident neutrons and small scattering angles \( Q \). For example, \( E_i \sim 10 \, \text{eV} \) with \( Q \sim 3^\circ \) is an indispensable setting for measuring the scattering at \( \mathcal{K}_\omega \sim 0.5 \, \text{eV} \) with \( Q \sim 4 \, \text{Å}^{-1} \). Such measurements are very difficult with chopper spectrometers, and hence resonance instruments may become
important. The present performance of these instruments, however, is not sufficient to perform such experiments, mainly because of poor signal-to-background ratio or sensitivity. A test experiment measuring the local vibrational modes in ZrH2 with low Q was performed using an RDS with a 241Pu foil /3/. The lowest Q values with which observation of excitations at 140, 280 and 420 meV was possible were about 4, 7 and 9 Å⁻¹ with scattering angles of 10°, 15°, and 20°, respectively. A measurement of high-energy magnetic excitation at about 650 meV in HoF₂ (spin-orbit excitation) was also tried, but a null result was obtained /3/. Current sensitivity of this technique is estimated to be about 1.5 b·sr⁻¹, while the cross-section for the excitation cited above is a few hundred mb·sr⁻¹. As mentioned already, the RDS with coincidence gave better sensitivity, but is not sufficient to detect such scattering. Further improvement is desirable.

REFERENCES

284

/10/ WATANABE, N., Resonance detector instruments, Proc. 1984, HEECM (see Ref. /2/).
/14/ WILLIAMS, W.G., Proc. 1984 HEECM Workshop (see Ref. /2/).
/20/ JOHNSON, R.G., Experimental tests of BGO and HPGe detectors for resonance detector systems, Poster presented at 1984 HEECM Workshop (see Ref. /2/).
/21/ CRAWFORD, K., et al., private communication.
/22/ CARPENTER, J.M., private communication.
COLD NEUTRON SCATTERING SPECTROMETERS AT THE SPALLATION NEUTRON FACILITY, KENS

Y. ISHIKAWA, Y. ENDOH
Physics Department, Tohoku University, Sendai
K. INOUE
Department of Nuclear Engineering, Hokkaido University, Sapporo, Japan

Abstract

COLD NEUTRON SCATTERING SPECTROMETERS AT THE SPALLATION NEUTRON FACILITY, KENS.

The paper discusses the advantages of the pulsed cold neutron sources based on the experimental results obtained with the cold neutron spectrometers SAN, TOP, LAM-40 and LAM-80 installed at the spallation neutron facility, KENS. One advantage of using pulsed cold neutrons is their potential for simultaneously measuring a wide range of momentum and energy transfers (SANS, LAM), which is shown to be quite important for the studies of non-equilibrium phenomena. Another advantage is the possibility of obtaining new information by employing white cold neutrons. The automatic separation of inelastic components from small-angle scattering (SAN) and the detection of magnetic states by simple transmission measurements (TOP) are examples of the possibility. These results indicate that the pulsed spallation neutron source has also good potentially useful for cold neutron scattering.

The KENS is the world's first spallation neutron facility. It has been operative since 1980 and is a unique facility where pulsed cold neutrons have been extensively utilized /1/. It has long been believed that the pulsed spallation neutron sources have fewer advantages for cold neutron scattering because of elongation of the pulsed width in the process of moderation /2/. Therefore, in most pulsed neutron facilities the main research has been confined to the problems of high energy and momentum transfers. Since, however, the demand for cold neutron scattering is quite strong in many fields of science, such a trend for the spallation neutron source has been considered a seriously weak point of this source. We have installed five spectrometers, SAN, TOP, LAM-40, LAM-80 and HRP on our cold neutron source and the results with these spectrometers have proved the good potential of the pulsed spallation neutron source, even in the field of cold neutron scattering. This paper makes a critical assessment of its potential.

One of the best examples of the potential is provided by SAN, a small-angle scattering spectrometer, installed at an exit of an 11 m cold neutron guide /3/. By employing white cold neutrons with
FIG. 1. (a) Temporal variation of scattering intensity during the process of spinodal decomposition of Al-6.8 at% Zn at \( T_a = 25^\circ C \). (b) Dynamic scaling of \( S(Q,t) \).

wavelength \( \lambda \), ranging between 3 and 5 Å, a two-dimensional PSD as well as several normal \(^3\)He counters around the sample, the spectrometer makes it possible to measure simultaneously the moment transfers \( Q \) between \( 4 \text{ Å}^{-1} \) and \( 3 \times 10^{-3} \text{ Å}^{-1} \). Such a wide dynamic range of \( Q \) measurements was found to be quite important for pursuing non-equilibrium phenomena such as spinodal decomposition. For example, recent studies on the spinodal decomposition process in the FeCr and Al-Zn alloys with SAN /4/ have revealed the very early stages of decomposition process which has been predicted by non-linear theory /5/, but has not been observed experimentally because of spreading of the scattering over a wide \( Q \) space. One example is shown in Fig. 1(a) where the temporal evolution of the scattering...
function $S(0,t)$ of Al--6.8% Zn, in the process of decomposition at 25°C, is displayed. The sample was annealed in-situ for a required time $t$, followed by quenching to 77°K for neutron scattering measurement. In Fig. 1(b) is plotted the normalized scattering function defined by $F(O/O_1(t)) = 0_1(t)^3 \cdot S(A,t)$ against $O/O_1$ with $O_1$ the first moment of the scattering function. The result suggests that there are two distinctly different stages in the decomposition process. In the later stage ($t > 7$ min), the scaling law holds and all $F(O/O_1)$ fall on a single curve. This has been predicted theoretically/6/ and has also been supported experimentally for a number of alloys/7/. The scattering function at high $O$ side obeys the $O^{-4}$ law (Porod law) as is indicated in a log-log plot in the inset on the figure. This is the stage where the precipitates already have a definite interface. We have, however, found that there is an earlier stage ($t < 7$ min) where the scaling law does not hold and $S(0,t)$ exhibits the $O^{-2}$ dependence at high $O$ side as can be seen in the figure. Such an early stage has also been observed in the Fe-Cr alloys/4/ and is found to be a general feature of the spinodal decomposition process. We should note that this early stage can be detected only with a machine with a wide dynamic range.

The SAN has another unique characteristic in that the small-angle scattering from the inhomogeneity can be separated from inelastic scattering as spin-wave scattering only by making a careful analysis of the $\lambda$ dependence of the scattering function; in case of elastic scattering, the scattering function does not show any $\lambda$ dependence. On the other hand, if the inelasticity is involved in the scattering, the $\lambda$ dependence appears and we can distinguish both types of scattering from each other. This technique was used to separate the spin wave contribution from the small-angle scattering in typical Invar alloy such as Fe$_8$Pt, Fe$_6$Ni$_{35}$ and Fe$_8$B$_1$, and the $O$ dependence of the real small-angle scattering could be deduced.

The TOP is the cold polarized neutron spectrometer which polarizes white cold neutrons with $\lambda$ ranging between 3 and 10 Å by means of total reflection from the curved Soller mirrors made of Fe-Co magnetic film evaporated on oriented polypropylene sheets (OPP)/8/. A simple application of the spectrometer is the transmission measurements with similar Soller mirror-type polarization analysers, which have been found to give new information on the magnetic states in the random system. Figure 2 displays an example of the results/9/. The Fe$_{0.7}$Al$_{0.3}$ sample used in the experiment is a re-entrant spin glass; the ferromagnetic magnetization in a weak field starts to decrease below a temperature $T_g$, disappearing at the lowest temperature as shown in Fig. 2(a). The $\lambda$ dependent depolarization measured in the different magnetic states (A-J) is plotted in Fig. 2(b) and (c), which exhibits quite different features depending on the magnetic states. In both paramagnetic (A) and re-entrant spin glass (J) states, no depolarization occurs over the whole wavelength, but in approaching $T_C(R)$ or $T_g(I)$, a linearly $\lambda$ dependent depolarization was observed. The most interesting result is an oscillatory depolarization observed in the ferromagnetic state.
FIG. 2. (a) Magnetization of Fe$_{0.7}$Al$_{0.3}$ measured in a weak field of 100 Oe, and (b,c) $\lambda$-dependent depolarization of each state.
(N-O), suggesting that the state is not the simple ferromagnetic one. This is also a new technique realized by employing white cold neutrons.

Both LAM-40 and LAM-80 are the crystal analyser spectrometers with similar large analyser mirrors /10/. The spectrometers are designed specially for the studies of the quasi-elastic scattering from hydrogen containing materials as polymers. They also proved that the pulsed cold neutron source is quite suitable for this kind of spectrometer; the resolution of the spectrometer can be modified in three orders of magnitude simply by changing spectrometer parameters. The incident neutron energy resolution $\Delta E_1$ can be controlled by changing the flight path $L_1$ between the sample and source, while final energy resolution $\Delta E_2$ can be improved by making the Bragg reflection angle $\Theta_R$ larger; $\Delta E_2/E_2 = \cot \Theta_R \Delta \Theta$.

By selecting the parameters ($L_1$ and $\Theta_R$) as ($6 m, 40^\circ$) and ($30 m, 80^\circ$) respectively, the LAM-40 and LAM-80 attain the energy resolution of about 100 $\mu$eV and 10 $\mu$eV, respectively. The LAM-80 has a special characteristic where the energy transfers can be measured simultaneously up to 1 meV by keeping the same energy resolution, providing another example of the wide dynamic range of the measurements with the KENS cold neutron source. It is impossible to have such a wide energy transfer range with a conventional spectrometer employing monochromatized incident neutrons.

A brief description of a few examples of the measurements with four different kinds of spectrometers has shown that the pulsed cold neutron source has good potential for competing with the cold neutron source in the steady reactor because of enlargement of the dynamic range of the measurements as well as new information obtained by simultaneous measurements of $\lambda$ dependent scattering. The economical advantage that the cold neutron can be obtained with a small refrigerator should also be mentioned. The KENS-I cold neutron source is cooled with a refrigerator of 40 W (PGH 105) and our recent mock-up experiment suggests that the solid methane moderator can be cooled with the same refrigerator even for the KENS-I' where the neutron intensity is increased one order of magnitude compared with the KENS-I.

REFERENCES

TIME-OF-FLIGHT SPIN ECHO*

F. MEZEI
Hahn-Meitner-Institut,
Berlin (West)

D. RICHTER
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

TIME-OF-FLIGHT SPIN ECHO.

The basic set-up for a spin-echo spectrometer at a pulsed source is outlined. Significant intensity gains compared to a steady-state source are possible in using wide neutron wavelength bands. At 50 Hz repetition rate and $\lambda = 5 \, \text{Å}$ the gain factor amounts to an order of magnitude while at the same time the Q resolution is about 10 times better.

REAL-TIME TEXTURE EXPERIMENTS

Dorte JUUL JENSEN
Riso National Laboratory,
Roskilde, Denmark

Abstract

A technique for fast in-situ texture determination by neutron diffraction is described. The basic instrument is variable wavelength neutron spectrometer modified to incorporate a linear position-sensitive detector and a fully automatic Eulergoniometer. At present a complete textural analysis with an accuracy of 2-3% determined by counting statistics, requires from 15 to 30 minutes measuring time depending on the sample material. By on-line recording of the change in texture, which commonly accompanies such processes as deformation, recrystallization and grain growth in crystalline materials, information about the process and its kinetics is obtained. This is illustrated by in-situ kinetic investigations of recrystallization and grain growth in various polycrystalline metals; the potential of the technique for problems related to metallurgical practice is demonstrated by an approach of texture tailoring in commercially pure aluminium.

1. INTRODUCTION

The great majority of technologically applied materials have a polycrystalline structure. In general the distribution function of the crystal orientations, or the texture of the material, is not random but has certain preferred orientations with respect to a set of macroscopic sample axes. The properties of the material, and therefore also the possibilities for applications, are strongly dependent on the texture; a precise knowledge and control of the texture development is therefore important from an industrial viewpoint. Texture measurements can, however, also be used as a probe to study fundamental metallurgical processes. As most metallurgical processes, like for example plastic deformation, recrystallization and grain growth, are orientation dependent, the texture is commonly changed during these processes. By following the change in texture, kinetic information about the crystallographic processes that take place in the bulk material is obtained. This type of information is not directly accessible through other measuring techniques. Using optical or electron microscopy the structure can be determined in great detail at any stage during the process; however, these techniques sample only a relatively thin layer and the processes occurring here are in general not the same as the bulk processes. Other techniques rely on changes in physical properties, e.g. a change in stored energy hardness or in electrical resistivity, but these methods are of a more indirect type than texture measurements, and the results cannot always be directly related to the micromechanisms taking place in the material.
The texture of polycrystalline materials is usually determined by pole figure measurements, i.e., diffraction measurements where the intensity of the diffracted signal from a particular set of lattice planes is recorded. Standard techniques, mostly based on X-ray diffraction /3/, are, however, too time-consuming to be applied for on-line kinetic studies of fast processes. Only recently has work been done to modify existing techniques for more rapid data acquisition. In 1976 Gerward et al. /4/ showed that the changes in texture during tensile deformation of an aluminum wire could be followed in-situ using energy-dispersive X-ray diffraction. The potential of neutron diffraction as a tool for investigating recrystallization kinetics of individual texture components was demonstrated by Hansen et al. in 1981 /5/. One of the latest developments in the field is the use of multidetectors and Juul Jensen and Kjems /6/ have shown that the data collection rate can be greatly enhanced by the use of a linear position-sensitive detector. This describes a further improved experimental set-up for rapid and automatic determination of several different (hkl) pole figures. The potential of the instrument is demonstrated via kinetic investigations of recrystallization and grain growth in various polycrystalline metals and via an attempt to produce specific recrystallization textures in commercially pure aluminum.

2. EXPERIMENTAL TECHNIQUE

The texture of a material is characterized by the three-dimensional orientation distribution function (ODF) \( f(g) \) defined as

\[
\frac{dv(g)}{v} = f(g)dg
\]

where \( dv(g) \) is the volume of all crystallites having orientations in the range \( g - g + dg \), and \( v \) is the total volume of the sample. In practice the ODF is not measured directly but calculated from several (hkl) pole figures from different (hkl) reflections, each of which represents an integral of the ODF in the plane perpendicular to the scattering vector. (A review of the experimental techniques and the methods of analysis is given in /7/).

2.1. The spectrometer

The basic unit for the fast neutron texture measurements is the multipurpose spectrometer TAS3, placed at the Risø DR3 reactor. The spectrometer can easily be changed from a standard triple axis spectrometer to a double axis especially equipped with a linear position-sensitive detector, PSD, and an Euler goniometer. The set-up is sketched in Fig. 1. A given wavelength is selected by a curved pyrolytic graphite crystal monochromator. The sample is mounted in the Euler goniometer, where it can be rotated about three axes under
computer control. And the detector is placed to record the intensity simultaneously along \( \sim 50^\circ \) of a Debye-Scherrer ring. The same diffraction condition is fulfilled for the entire detector only if the scattering angle \( 2\theta_s \), between the incident and the diffracted beam, is equal to \( 90^\circ \) where the Debye-Scherrer cone turns into a plane perpendicular to the incident beam. The continuous spectrum from a thermal reactor, however, allows one to choose a wavelength such that \( 2\theta_s = 90^\circ \) for most low-index reflections from common metals and alloys.

The detector is a \(^3\)He detector operated in the proportional regime. It is a 0.5 m long, 5x10\(^{-2}\) m diameter stainless steel tube with a 3x10\(^{-4}\) m diameter quartz wire coated with colloidal graphite as the resistive anode (2.5 k\(\Omega\)/cm) \( /8/ \). The position resolution of the detector system is about 1 cm (full-width half-maximum), and it can handle counting rates up to \( \sim 20,000 \) counts/s without appreciable dead time (for higher counting rates, dead time corrections are handled by a multichannel analyser system MCA). For the present use, the data recorded in the MCA are summed to give 18 "reduced channels" representing equivalent segments along the detector. This means that the detection system can be regarded as 18 independent detectors placed along the Debye-Scherrer ring. (For further experimental detail, see \( /6/ \)).

The complete spectrometer, temperature and processing of the signals from the PSD are controlled by a PDP-11/23 computer. A CANON computer with a colour screen and an 8-colour jet-ink plotter is directly coupled to the PDP 11 providing an on-line graphical presentation of the measured data.

2.2. Experimental procedure

With one setting of the sample the PSD records the diffracted intensities along \( \sim 50^\circ \) of a Debye-Scherrer ring, corresponding to \( \sim 34^\circ \) of a small circle in a (hkl) pole figure. A complete pole figure is obtained by rotating the sample discontinuously in the Euler goniometer, with the sample fixed while counting. Typically,
180 settings of the sample corresponding to 180x18 measuring points
are required for a complete pole figure measurement. However, owing
to symmetry this number can be reduced. For example, for rolled
cubic materials, which are of current interest in metallurgical
studies, only a quarter pole figure is needed. In this case 61
settings of the sample are used, and typical measuring times (with
an accuracy of 2-3% as determined by counting statistics) range
from 7 to 15 minutes depending on the scattering cross-section of
the sample material. This can be compared with standard X-ray
techniques where a measuring time of about 2 hours is required for
similar measurements.

On applying the calculation method introduced by Bunge /9/
at least two pole figures from different reflections are needed
to determine the ODF (assuming cubic crystallite and orthorhombic
sample symmetry). A typical time resolution is therefore about 20
minutes. Thus, it is possible to make reasonably accurate ODF-based
kinetic investigations of processes, which involve a texture
transformation over at least a couple of hours.

The kinetics of very fast processes, where this time resolution
is insufficient, can be followed by means of partial pole figures
that focus on selected texture components. Here the sample
orientation is changed only between a few preselected positions.
In the limiting case the sample is kept fixed during the
measurements, but as the PSD covers 34° of a small circle in the
pole figure plane, it is possible to position the sample so that
the detector records the change in intensity of one or even a few
texture components simultaneously, and thus a time resolution of
the order of seconds is obtained.

3. APPLICATIONS

3.1 Recrystallization

The energy stored in a plastically deformed material may be
reduced by annealing. During recrystallization almost dislocation-
free nuclei develop and grow.

In general one or both these basic processes are orientation
dependent, resulting in a change in texture from the deformed to
the recrystallized state. An example of this is given in Fig. 2.
Here the ODF for commerically pure aluminium heavily deformed by
rolling is shown (Fig. 2a), and the ODF for the same material after
annealing at 240°C for 15 hours is shown in Fig. 2b /10/. The ODFs
are represented in a standard form, where the orientations of the
crystallites are described by the Euler angles $\phi_1, \phi_2$ and
$f(g)(1)$ is plotted by equidensity lines in sections of the Euler
space with $\phi_2$ = constant /7/. A significant change of a rather
complex nature is observed between the two states and a full ODF
investigation is needed to determinate the overall kinetics. From
ODF data measured during annealing, the volume fraction of the main
texture components is calculated and plotted vs. time (Fig. 3). From
such results precise kinetic information is obtained, which can be
FIG. 2. ODFs for aluminium of commercial purity.
(a) Cold-rolled to 95% reduction in thickness.
(b) Cold-rolled to 95% reduction and annealed at 240°C for 15 hours.
(hkl)<uvw>, (hkl) are assigned to the normal direction of the sheet plane
and <uvw> to the rolling direction.

FIG. 3. The development of specific texture components during isothermal annealing
at 240°C of aluminium of commercial purity, cold-rolled to 95% reduction.
used to determine important overall characteristics such as, for example, the volume fraction of recrystallized material as a function of time and thereby also the time for 50% recrystallization. Combined with microscopical observations the kinetic texture results give an improved understanding of both the nucleation and growth processes taking place /11/. Furthermore, the results can be used to evaluate kinetic models. As an illustration of this (see Fig. 3) the measured data are fitted to an Avrami equation /12/, which has been commonly used to describe recrystallization kinetics:

\[ X = 1 - \exp(-A \cdot t^\beta) \]  

where \( X \) is the recrystallized volume fraction, \( t \) is the time and \( A, \beta \) are constants characteristic of the process.

It is seen that the equation gives a reasonable fit to the measured points and thereby an accurate determination of the fitted parameters \( A, \beta \). For the aluminium data, shown in Fig. 3, \( \beta \)-values below 2 are found. According to the Avrami interpretation this corresponds to growth of recrystallized grains in one or two dimensions (needle- or plate-shaped grains) /12/. However, the recrystallized grains are, via microscopical investigations, found to be equiaxed, meaning that a simple growth model, as assumed in the Avrami interpretation, fails to describe the present recrystallization process. This result is in agreement with earlier observations of the recrystallization of pure copper based on measurements of single-texture components /5/.

### 3.2. Grain growth

Annealing of a recrystallized specimen at elevated temperatures can cause grain growth, whereby the stored energy in the material is further lowered owing to reduction of the total grain boundary area /13/.

The texture development during grain growth in cold drawn and recrystallized pure copper rods has been followed by the neutron diffraction technique in parallel with the microscopic investigations of the evolution of the grain-size distribution upon annealing /14/. It was found that grains having the (111) normal direction parallel to the fibre axis grow at the expense of grains with the (100) direction along the fibre axis, and that the growth process was of the anomalous type /13/. Furthermore, an increase in annealing temperature leads to a higher \((111)/(100)\) ratio (see Fig. 4). The development of these two main texture components has been followed via the fast kinetic measurements to determine the growth/decrease rate of the components for various annealing temperatures.

### 3.3. Other applications

The macroscopic anisotropy and thereby the properties of the material depend on the texture. By controlled "texture tailoring", materials with a certain application profile might be produced.
FIG. 4. The volume ratio of grains having the (111) and (100) direction parallel to the fibre axis after grain growth in pure copper rods at various temperatures. (O and □ refer to two determination methods).

This requires knowledge of the texture changes during the possible fabrication processes for a variety of materials. For cold-rolled aluminium of commercial purity the effect of different thermal treatments has been studied, using the neutron diffraction technique /15/. On-line texture measurements, together with a fast response furnace system, make it possible to adjust parameters such as heating rate, time and temperature according to the actual texture development and thereby control the direction of the development.

4. CONCLUSIONS

The use of a standard neutron spectrometer modified to incorporate an Euler goniometer and a linear position-sensitive detector enables rapid determination of the texture of bulk specimens. With the present set-up a complete texture analysis, based on an ODF calculated from two pole figures, requires a measuring time of about 20 minutes. An on-line investigation of processes, which involve a texture transformation over at least a couple of hours, is therefore possible. The method is non-destructive and it samples the crystallographic changes within a bulk material. This type of in-situ information is not directly accessible from other standard measuring techniques, and combined with, for example, microscopical investigations of the change in microstructure fast in-situ texture measurements can serve as an important tool for a further understanding of basic physical metallurgical processes.
REFERENCES


/12/ AVRAMI, M., J. Chem. Phys. 8 (1940) 212.


/15/ JUUL JENSEN, D., HANSEN, N., "Texture and microstructural changes following thermal treatments" (7th Int. Conf. Montréal, 1985) (to be published).
BIOLOGY AND MACROMOLECULES

(Session VIII)
Chairmen

B. FENDER
Institut Max von Laue - Paul Langevin

K. NIEHAUS
Federal Republic of Germany
Invited Paper

BIOMOLECULAR APPLICATIONS OF NEUTRON SPECTROSCOPY: CURRENT WORK AND FUTURE DEVELOPMENTS

H. D. MIDDENDORF*
Department of Biophysics,
King's College, London,
United Kingdom

Abstract

BIOMOLECULAR APPLICATIONS OF NEUTRON SPECTROSCOPY: CURRENT WORK AND FUTURE DEVELOPMENTS.

Since around 1980, as the result mainly of instrumental advances, biophysical and biochemical applications of quasi-elastic and inelastic neutron scattering have been pursued more actively. Following a brief outline of biomolecular dynamics from the perspective of a neutron spectroscopist, this paper focuses on two areas where progress is being made: dynamics of hydration processes, and intramolecular dynamics of proteins. The spatio-temporal information obtained makes neutron scattering a valuable experimental complement to numerical simulation studies, and the prospects for developing biomolecular applications of neutron spectroscopy are very good.

1. INTRODUCTION

A great variety of physical techniques are at present being used in studies of the structure and function of living matter at the molecular level. Most of these techniques were devised originally by physicists; at some stage in their development they have passed into the hands of the biophysicist, biochemist or molecular biologist, and subsequently they have often undergone considerable adaptation and refinement before becoming part of the impressive array of tools with which it is possible nowadays to probe complex macromolecular systems.

Neutron scattering is a relative newcomer among biophysical techniques. Despite an auspicious start in 1966 with crystallographic work on vitamin B\textsubscript{12} at Harwell by Moore, Willis, and Hodgkin /1/ and inelastic scattering experiments on polypeptides in the USA by Boutin, Whittemore, Gupta, and Trevino /2/, it was only during the 1970s, after the establishment of research centres with high-flux reactors at Brookhaven and Grenoble, that bio-neutron studies began to make an impact /3/. These were almost exclusively

*Present address: Clarendon Laboratory, Oxford University,
Parks Road, Oxford OX1 3PU, United Kingdom
devoted to structural work, i.e. the analysis of small-angle interference patterns produced by coherently scattered neutrons. From a purely biophysical or biochemical point of view, all such applications of neutron scattering represent straightforward extensions of X-ray diffraction, the principal new features being the emphasis on light elements and the scattering contrast between hydrogen and deuterium. From a neutron physicists's perspective, however, diffraction studies exploit only part of the information content of the scattered radiation field. Any energy changes due to coherent or incoherent scattering from thermal motions, or changes in the spin states of scattered neutrons, are not analysed in conventional diffraction work. If one considers that experiments of this kind are quite expensive, whatever the particular set-up, it seems worthwhile even in the case of complex biomolecular samples to go beyond diffraction and explore the full potential of neutron scattering techniques.

2. NEUTRONS AS PROBES OF BIOMOLECULAR DYNAMICS

What dynamic features will cold or thermal neutrons be able to 'see' in biomolecules, and on what functional aspects can neutron spectroscopy provide information not accessible by other techniques?

To refer to 'biomolecules' or 'biopolymers' as a single class of organic substances is, first of all, a gross oversimplification, even if we are a little more precise and distinguish between proteins, nucleic acids, and lipid assemblies. The biomolecules that interest us here, disregarding the last category, are all linear polymers twisted or folded into a well-defined three-dimensional shape, with a characteristic sequence of side groups (amino acid side chains, nucleotide bases) that determines much of their biological function. The two prototypical forms are globular and fibrous: the former are all proteins, whereas the latter may be proteins (collagen, intermediate filaments, muscle components) or nucleic acids (DNA fibres).

The main types of motion observable by quasi-elastic and inelastic neutron scattering may be classified as follows.¹

(a) Optical and acoustical phonons. Co-operative vibrational excitations that propagate according to a dispersion relation \( \omega(q) \) require regular, spatially extended structures. The low-frequency branches (\( \hbar \omega < 100 \text{ cm}^{-1} \)) of the phonon dispersion diagram have been mapped out for a number of molecular crystals of biophysical interest, e.g. for single crystals of deuterated amino acids and of hydrogenous DNA-type pyrimidines /4/. Because of

¹Numerical values for the energy transfer \( \hbar \omega \) will be quoted in units of cm\(^{-1} \), defined by 1 cm\(^{-1} \) = 124 \( \mu \text{eV} \) (\( \hbar = \text{Planck's constant}/2\pi \)).
solvent damping, defects and other intrinsic irregularities, it seems difficult to observe phonons in more 'natural' biomolecular samples. Low-frequency acoustic phonons have, however, been implicated in several important energy transduction and control processes in proteins, DNA and muscle /5/. Non-linear mechanisms for energy transfer over relatively long distances by means of mobile, soliton-like regions of conformational changes have in particular received a good deal of attention. In α-helices, such excitations depend on the coupling between amide-I vibrations and longitudinal acoustic modes, with energies (from analytical models and numerical simulations) in the 5 to 50 cm⁻¹ region.

(b) Fluctuations and Breathing Modes. Many biomolecules, especially proteins, are only marginally stable and possess a broad spectrum of 'soft modes' (KT ≥ ℏω ≥ 1 cm⁻¹) with nuclear amplitudes that can be much larger than those associated with the more familiar high-frequency vibrations. These low-frequency fluctuations are functionally significant since they reflect the structural heterogeneity which enables biological macromolecules to perform highly specific recognition, interaction and transduction processes. Stochastic fluctuations of the secondary and tertiary structure of proteins were first postulated in connection with isotope exchange studies /6/ to explain the accessibility of interior groups ('breathing modes'). Neutron diffraction studies /7/ have provided structural details of local fluctuations resulting in the cooperative breaking of several hydrogen bonds ('regional melting'). Theoretical approaches to describing stochastic fluctuations of this kind include computer simulations of the molecular dynamics of structural elements and entire molecules /8/, and (for long wavelengths) calculations based on continuum models /9/ (density fluctuations in visco-elastic media). The difference between these fluctuations and those represented by a 'sea' of short-lived acoustical phonons is only conceptual, and the frequency range is the same.

(c) Side Group Motions. The sequence of side groups (amino acid side chains, nucleotide bases) along the main chain(s) of a biomolecule are to a lesser or greater degree involved in the dynamics of the structure as a whole. At one extreme, side chains at the surface of proteins tend to be weakly coupled to the backbone and their dynamics is largely determined by solvent interactions. The other extreme is that side groups are buried in the interior where they are more of an integral part of the structure and therefore contribute appreciably to its overall dynamical properties. Torsional modes and hindered rotations are prevalent in side groups, with frequencies in the 150-400 cm⁻¹ region (methyl groups) down to very low values for groups inside hydrophobic domains. In the interior and in clefts, the steric hindrances may be such that side groups can only perform rapid jumps
between a few relatively well-defined potential minima (as is the case, for example, for 180° flips of aromatic rings). At the surface or in an environment with considerable local flexibility, on the other hand, their mobility may assume a diffusive character. Apart from amino acid side chains, other covalently attached groups with distinct functional properties (prosthetic groups, cofactors) will often play an important role in the dynamics of surface and near-surface interactions.

(d) Inter-Domain and Inter-Submit Motions /10/. The domain structure of many globular proteins, and also the quaternary structure of larger proteins, gives rise to low-frequency motions of massive parts of the molecule relative to each other. Nominal frequencies derived on the assumption of harmonic vibrations are of the order of 10 cm⁻¹ or less; these however will be highly damped in solution and the domains or sub-units are more likely to perform restricted Brownian motions around their equilibrium positions. The active sites of enzymes are usually located in clefts which separate two domains, and the inter-domain motions of interest here are bending modes around the 'hinges' given by the polypeptide chain segments connecting the two domains. Inter-domain motions of this kind are sometimes also called 'breathing modes'.

(e) Solvent Interactions /11/. In aqueous solutions, depending on the scale length considered, there are two levels of dynamic interactions: Over scale lengths in the 1-10 Å region, the translational and rotational degrees of freedom of water molecules in a hydration shell are reduced by interactions with the macromolecular surface and its various features (ionic groups, polar and nonpolar regions). Over scale lengths of the order of 100 Å, the integrated effects of these short-range interactions determine the translational and rotational Brownian motion of the macromolecule as a whole, and thus its properties as observed by macroscopic techniques (diffusion coefficients and viscosity).

While it is useful to classify the atomic and molecular motions in and around biomolecules according to frequency range and structural features involved, there is evidently considerable overlap between the five categories discussed here and to some extent these reflect different experimental and theoretical approaches to the subject. One may discern, broadly, three main lines of thought: phenomenological descriptions developed along with particular experimental methods, theoretical concepts adapted from solid-state physics, and interpretations based on numerical simulations of the molecular dynamics /12/.

The last of these approaches provides the most powerful methods and is of major interest in the present context. Ab initio simulations start from the crystallographically known co-ordinates of all atoms in a structure, and generate their trajectories over a time interval of the order of 100 ps by solving the Newtonian
equations of motion in steps of 0.002 - 0.005 ps for an empirical potential energy function. This function represents a superposition of terms describing bond stretching and bending, torsion, electrostatic and van der Waals interactions, hydrogen bonding and polarization effects. For small proteins with molecular weights \( MW < 10^4 \), or segments of \( \alpha \)-helices and DNA, it is possible already to account for all of these interactions; for larger systems it is necessary to introduce constraints which limit the results to lower frequencies or spatially to particular regions of interest. The inclusion of a hydration shell also requires simplifications. Although full numerical simulations have only been performed for a few proteins so far, it is certain that this method will be applied more extensively during the next ten years, and that simulation results will become available for most biophysically interesting proteins with \( MW \leq 50,000 \). Once a good set of trajectories is known for a given system, it is relatively straightforward to calculate the time-correlation functions and dynamic structure factors needed for a detailed interpretation of inelastic neutron scattering data.

3. TWO MAJOR PROBLEM AREAS

Following this brief outline of biomolecular dynamics and some of the challenging problems arising here, I would like to focus on two areas where experimental progress is being made /13/.

3.1. Biomolecular hydration

A comprehensive description of the spatiotemporal properties and the functional role of water associated with biomolecules and in living cells is an important but exceedingly difficult task. Since Henderson's classic discussion /14/ of the "biological significance of the properties of matter", more than half a century of research on biological water has given us some understanding of its nature, but this work has also spawned a number of controversial ideas and a vast amount of data from disparate experimental techniques that are not easily integrated into a coherent picture. A good appreciation of the extraordinary range of problems encountered here, and of the experimental and theoretical approaches used in current work, may be gained from the proceedings of a recent conference /15/.

Since Whittemore obtained the first hydration difference spectrum of a polyopeptide in 1968 /16/, inelastic neutron scattering techniques have been used in a number of studies attempting to characterize the dynamics of hydration phenomena in systems ranging from biochemically pure preparations to live cells. By measuring \( S(Q,\omega) \) over a region of momentum and energy transfers matched to the microscopic distance and time-scales of interest, and by exploiting
the wide range of H/D contrast that can be realized in hydration experiments involving biosynthetically deuterated macromolecules, such studies can provide much new information, which should eventually lead to a better understanding of the nature and function of biological water.

An attractive feature of neutron spectroscopy as applied to hydration problems is that it is feasible to examine both the diffusive domain (\( \nu \omega \lesssim 1 \, \text{cm}^{-1} \)) and the transition region up to several 100 \( \text{cm}^{-1} \) where the scattering from more localized, discrete modes begins to dominate. Neutron spectra in the latter region, between a few \( \text{cm}^{-1} \) and \( \approx 800 \, \text{cm}^{-1} \), enable hydrogen-weighted spectral density functions to be determined which are closely related to thermodynamic properties. A successful separation of such functions, by H/D contrast measurements, into three components (bulk biopolymer, interfacial region, bulk water) could provide a much-needed basis for the quantitative description of surface-associated water and the coupling of intramolecular fluctuations to an aqueous environment.

The experiments that have been performed to date fall into two categories:

(a) Sorption studies where \( 0 < h < 1 \) (\( h = g \text{ water}/g \text{ dry mass} \)), on samples that are being hydrated in steps from the dry or nearly dry state up to levels of water content necessary for functional interactions. By analysing sets of contrast-dependent difference spectra taken along the sorption isotherm, it is possible to examine the structural and dynamical changes accompanying the gradual activation of the various degrees of freedom of a biomolecule and its interaction with increasing amounts of closely associated water /17/.

(b) Studies of the hydration dynamics in more or less concentrated systems for which \( h \gtrsim 1 \), such as solutions of purified proteins or nucleid acids, protein crystals, and whole cells or tissues. Here all biomolecular constituents are fully hydrated, and attention focuses on how the properties of the water of hydration are modified relative to the bulk water component.

It must be stressed that these two kinds of experiment probe different aspects of the molecular dynamics of hydration processes: the changes occurring at or near the macromolecular surface as the result of vapour pressure changes cannot be expected to relate in any simple manner to the behaviour of the interfacial region under fully solvated conditions. For live cells and organisms, \( h \) is usually larger than 1 and may reach values of 10 in plants. The full transition from (a) to (b), i.e. the progressive activation of a biological system by hydration from \( h \approx 0 \) to \( h > 1 \), is a very common and important process (plant growth from seeds) the molecular details of which are largely unknown. A few microorganisms tolerate
virtually complete dehydration in a reversible way; this unique phenomenon has been exploited recently in a quasi-elastic scattering study /18/ of the water dynamics in cysts of the brine shrimp Artemia for 0.1<h<1.2.

Apart from an early study at Harwell on the mobility of water in a solution of oriented tobacco mosaic virus particles /19/, all inelastic scattering experiments under (b) have been on whole cells. The questions of greatest interest here relate to the existence and properties of an 'ice-like' component in the water of hydration /20/, and to hypotheses about long-range correlations (of the order of 100 Â) in intracellular water /21/. There is still considerable disagreement among biophysicists and physiologists about the spatiotemporal properties of intra-cellular water, and the interpretation of NMR experiments in this area remains controversial /22/.

In experiments on hydrogenous samples, it is mainly the incoherent scattering due to a protonated component undergoing large-amplitude displacements (i.e. H$_2$O) that is observed against a 'background' of relatively immobile protons (i.e. those belonging to the macromolecular constituents, including strongly associated water protons). The spectra are often interpreted on the basis of a composite scattering law given by

\[ fS^b_{\text{inc}}(0,\omega) + (1-f)S^m_{\text{inc}}(0,\omega) \]

where f is the fraction of water molecules that appear to be 'bound' with respect to the time scale resolved, and (1-f) is the 'mobile' fraction. The motions of the former are approximated by a Debye-Waller factor, whereas the latter, i.e. $S^m_{\text{inc}}(0,\omega)$, is expressed as the convolution of a translational model with one or more terms of the expansion of a rotational scattering law. Results obtained for Artemia /18/, for example, show very nicely how the motional freedom of much of the intracellular water is reduced by association with macromolecular constituents. Comparison with parallel NMR and dielectric relaxation measurements leads to the conclusion that this is not due to obstructions, compartments, or exchange with minor phases, but is a consequence of microscopic interactions over distances between 1 and 10 Â.

Fully complementary H/D contrast variation experiments can only be done with samples that are also available in biosynthetically deuterated form. These are of particular value in sorption experiments. High-resolution quasi-elastic and inelastic neutron spectra have been reported /23/ for partially and fully in-vivo deuterated samples of the phycobiliprotein C-phycocyanin isolated from blue-green algae. C-phycocyanin is a multimeric chromoprotein located between the thylakoid membranes, and its function is that of a light-harvesting protein involved in photosystem II. The cross-section ratio between sorbed H$_2$O
molecules and the bulk of a protein is particularly favourable for covalently deuterated samples, and changes in the vibrational properties become measurable already at hydration levels of about 2% (g water/g protein), i.e. during the first hydration stage when interactions with ionizable side chains predominate. Because of the large number of modes in a heterogeneous macromolecular system, the absence of selection rules in neutron scattering, and various broadening effects, such spectra always consist of a continuous intensity distribution carrying numerous more or less differentiated features. The complex pattern of intensity changes seen here reflects both the net increase and the repartition of the various hydration-activated degrees of freedom of the protein water system. For any fixed covalent deuteration level of the protein, by increasing the D2O fraction at constant total water content, one may 'fade out' the proton signal from the water of hydration and those labile hydrogen positions that exchange on the time-scale of the experiment. This reduces the incoherent scattering throughout the Q-range observed, but it also shifts the emphasis in the wet-minus-dry difference spectra from single-particle motions to two-particle correlations, and gives characteristic Q-dependences which carry information on the collective aspects of the water dynamics and its coupling to the protein matrix. At higher hydration levels, equivalent to monolayer coverage and beyond, the fraction of bulk-like water molecules increases and we expect the vibrational coupling to the protein to become 'softer' overall; this is borne out by the difference spectra which show a number of distinct intensity changes and peak shifts.

3.2. Intramolecular dynamics

The study of hydration and exchange processes in biological macromolecules is of course closely related to, and often inseparable from, studies of their intramolecular dynamics. The majority of the latter have focused on proteins because protein molecules, as a class, are distinguished by a diversity of structural and functional properties not found in other biomolecules. This diversity is reflected in a very wide range of dynamical phenomena which extend spectrally over more than 15 decades, from 10^15 Hz down to processes with frequencies below 1 Hz /12/.

The characteristic levels of structural organization found in a biomolecule are to some extent reflected in the frequency spectrum it exhibits. The skeletal modes of vibration associated with the primary and secondary structure of polypeptides (proteins) and polynucleotides (DNA, RNA) are generally accessible to modern time-of-flight spectrometers and the lower frequencies (\(\hbar \omega < kT \approx 200 \text{ cm}^{-1}\)) are of greatest interest here. The modes associated with the tertiary and quaternary structure of proteins, i.e. the way in which structural elements and building blocks are assembled into larger entities capable of performing complex
biochemical reactions and control processes, fall typically in the 1 to 50 cm\(^{-1}\) region. Whereas spectral changes observed at high frequencies are, as a rule, indirectly due to functionally interesting dynamical processes, future work will be concerned much more with detecting as directly as is possible the co-operative low-frequency motions in biomolecular systems. Experimental information is lacking here, and neutron spectroscopy is uniquely capable of bridging the ns to ps region by overlapping with NMR on the low-frequency side and with optical techniques at higher frequencies.

At very low frequencies, corresponding to energy transfers between 10\(^{-5}\) and 10\(^{-2}\) cm\(^{-1}\) (neV to µeV), intramolecular fluctuations naturally involve massive parts of a biomolecule. In the sorption studies discussed above, it is possible to \(\text{D}_2\text{O-}
\) hydrate a hydrogenous multimeric protein and observe (on a backscattering instrument) small \(\text{D}\)-dependent line broadenings due predominantly to incoherent scattering. Using a spin-echo spectrometer it is also possible to measure the slope of the intermediate scattering function at low \(Q\) as \(t \to 0\) for the complementary situation, i.e. for \(\text{H}_2\text{O-}
\) hydration of a covalently deuterated protein /23/. Here the motions of the water protons and of H-exchanged groups at or near the surface are fast enough to contribute only to a 'depolarized' background signal. Both types of experiment are being performed on C-phycocyanin /23/, and the results represent the first hard data relating to the concept of 'hydration-controlled fluctuations' /24/, i.e. restricted Brownian motions of the sub-units relative to each other. Similar solution scattering experiments using the spin-echo spectrometer IN 11 at Grenoble have been carried out on haemoglobin and n bromo grass mosaic virus. A recent, more detailed IN 11 study of the flexibility of immunoglobulin /25/ is of considerable interest in the context of current ideas on antigen-antibody dynamics /26/.

At higher frequencies, between about 1 and 1000 cm\(^{-1}\), intramolecular fluctuations in globular proteins involve primarily elements of the secondary and tertiary structure. Their dynamical properties have been widely discussed in connection with simulation studies and specific processes such as enzymatic catalysis and oxygen transport. Two kinds of experimental technique have been brought to bear on these problems: 'Selective' methods probing the motions of certain natural or artificially introduced atoms or groups (e.g. resonance Raman scattering or Mössbauer spectroscopy), and techniques making use of interactions that are less specific and therefore give averages of one sort or another. Of the latter, crystallographic temperature factor data and neutron spectra contain information averaged in complementary ways: for a few proteins, the mean-square displacements \(<x^2>_i\) derived from X-ray diffraction are individually known for each heavy atom \((i)\) but the \(\omega\)-dependence is lumped inside Debye-Waller integrals; incoherent neutron scattering data, on the other hand, provide spectral resolution but the differential intensities are spatially averaged over all hydrogens in the structure.
Although the specific cross-section of a hydrogenous, D$_2$O-exchanged protein is an order of magnitude larger than that of heavy water, much of the inelastic scattering from a protein solution will be due to the perdeuterated buffer even when the solution is very concentrated. At concentrations in the 1 to 10% range, time-of-flight spectra with exceptionally good counting statistics are required to resolve intramolecular modes of proteins and to detect spectral changes resulting from ligand binding, sub-unit interaction, or complex formation. Considerable impetus for solution scattering experiments has come from the time-focusing spectrometer IN6 at Grenoble which combines moderately good resolution with relatively high flux. In current solution scattering work using IN6 and the multi-chopper spectrometer IN5, time-of-flight data are being collected for four systems: hexokinase with and without glucose, lysozyme with and without N-acetylglucosamine (GlcNAc) or its trimer, bovine pancreatic trypsin inhibitor (BPTI), and lactate dehydrogenase (LDH) with and without its co-enzyme. These experiments address a number of important questions on the dynamics of proteins in relation to numerical simulation studies and to molecular interpretations of their thermodynamic properties. Two closely related but in practice somewhat distinct aims are being pursued here:

(a) Interpretation of the data obtained in terms of frequency distributions that can be compared with the results of ab initio molecular dynamics simulations in the range 0.05 to 100 ps (corresponding to GGO to 0.3 cm$^{-1}$). This requires realistic simulations (including solvent effects) and is at present feasible only for relatively small and crystallographically well-characterized proteins.

(b) Interpretation of difference spectra from two well-defined states of a system, obtained under conditions similar to those used in microcalorimetric studies. The primary aim is to analyse certain integral properties of these difference spectra and to relate overall changes to the thermodynamic quantities characterizing the two states.

The first goal in enzyme experiments must be to resolve spectrally and quantify the well-known 'stiffening' of an enzyme upon ligation, and to look for evidence in support of inter-domain motions such as the much-discussed hinge-bending mode in the 1 to 30 cm$^{-1}$ region. Initial results obtained for lysozyme (Lz) at rather high concentrations (85 or 200 mg/ml) suggest that enhanced co-operative fluctuations of the tertiary structure of Lz relative to Lz(GlcNAc)$_n$ occur at low Q, where the dynamics is sampled over distances between 10 to 15 Å; to substantiate this observation it will be essential to extend the data to lower concentrations. Similar results have recently been obtained for LDH. As regards (a), extensive simulations of the molecular dynamics exist for BPTI,
and this is obviously a good candidate for first comparisons. Spherically average single-phonon spectra of D$_2$O-exchanged BPTI have been calculated on the basis of a normal-mode analysis /27/, and the results are being used to examine the relationship between hydrogen-weighted frequency distributions derived from IN6 spectra, the single-phonon scattering, and further calculations aimed at determining two-phonon corrections. For the small, compact protein investigated, this work is expected to shed light on the key question of how closely the hydrogen-amplitude weighted frequency distribution approximates the 'true' frequency distribution in different spectral regions.

4. FUTURE DEVELOPMENTS

Although the potential of neutron scattering techniques for the study of biomolecular structure and dynamics is undoubtedly very great, their exploitation has for a long time been hampered by low fluxes and the small number worldwide of high-performance instruments. For diffraction work, these limitations have to some extent been overcome or have at least become more tolerable during the past five to seven years. Structural studies using neutrons have already reached a certain degree of maturity, and neutron diffraction is clearly developing into an important tool of structural molecular biology. The same cannot yet be said for biophysical and biochemical applications of neutron spectroscopy: here the flux and access requirements are larger by one to two orders of magnitude, and this has meant that most dynamical studies are still at an exploratory stage.

There are signs however that this first phase of work is coming to an end, and that we are entering a period of growth during which it will become possible to study a number of selected systems in much greater detail and in parallel with analytical model calculations or numerical simulations of the scattering laws. There are two reasons for looking forward with optimism to the 1990s:

(1) The first has to do with the tremendous increase generally in experimental and theoretical investigations of the molecular dynamics of proteins, nucleic acids, and membranes. Not surprising, against this background, is the interest shown by biophysicists and biochemists in a 'new' technique capable of providing spatiotemporal information in the crucial ns and ps region over distances between 1 and 100 Å. Although the sample sizes and run times required are often a disappointment to prospective collaborators, it is gratifying to note that the new results obtained have already attracted a good deal of attention. One consequence of this has been that in recent years the 'neutrocratic' hurdles for work of this kind have not been as high as they used to be, and that it is possible nowadays to obtain instrument time for experiments that were considered far too adventurous during the 1970s.
(2) The $Q,\omega$-regions covered by neutron spectrometers have expanded substantially in recent years and continue to expand. Considered in isolation from other factors, the $Q,\omega$-coverage and the resolution of spectrometers in current use must be regarded as excellent for developing biomolecular applications of neutron spectroscopy. Flux and access are the overriding factors limiting progress here, and we can expect that these constraints will be relaxed gradually in the near future with the commissioning of high-intensity spallation sources and the development of fast spectrometers duplicating or complementing those existing at steady-state reactors.

Perhaps the most striking fact to emerge from a survey /13/ of the small body of published work and of reports on work in progress is that inelastic scattering experiments have already been performed on an extremely wide range of samples, from small proteins in solution to membrane stacks, oriented DNA and collagen fibres, photosensitive preparations and whole cells. This must be surprising because the structural and dynamical complexity of biological systems is such that the domain of essential sample parameters (concentration, pH, H/D contrast, temperature, etc.) is very large, and also because biomolecular spectroscopy typically calls for difference experiments: a well-defined perturbation is applied to a big and complicated system, and the resulting difference signal can be quite small. While a good idea of what can be done with neutrons has been gained in these experiments, it is clear that much of the future work in this field will be concerned with substantiating the results already obtained and with extending them to determine the effects of changes in temperature and other important sample parameters that have not been explored at all. In parallel with the experimental work, an essential part of future research will be to develop in greater detail the theoretical interpretation of inelastic scattering from heterogeneous samples of complex structure. It is more difficult, beyond this predictable phase of work, to forecast intrinsically new applications of neutron spectroscopy in molecular biology, or even to single out particular experiments that have not already been proposed or attempted. Possible applications of anomalous or magnetic scattering, isotope effects, and ultra-cold neutrons spring to mind here, but it is difficult to imagine at present how these could be exploited except in very unusual circumstances. It seems certain, however, that the more widespread use of pulsed neutron sources from around 1986 onwards will stimulate new experimental approaches to probing $S(0,\omega)$ by intense polychromatic pulses with repetition rates that are comparable with the characteristic relaxation times of several processes of biophysical and biochemical interest.

Judging from the considerations sketched under (1) and (2) above, we can expect biomolecular applications of inelastic scattering to expand considerably during the next ten years. More
specifically, we may look forward to new results in some or all of the following problem areas:

(a) Determination of space-time correlation functions for phonon-like and stochastic intramolecular fluctuations in the 1 to 1000 cm\(^{-1}\) range, together with efforts to identify and separate contributions due to side-chain motions.

(b) Studies of solvent-dependent changes in the quasi-elastic and inelastic scattering from biomolecules over a wide range of H/D contrast, with the aim of developing the quantitative description of solvent-solute interactions at the molecular level: dynamics of diffusive processes, repartition of the motional degrees of freedom at higher frequencies.

(c) Measurements of changes in the low-frequency spectra of proteins (mainly 0.1 to 200 cm\(^{-1}\)) as the result of specific functional interactions involving their tertiary and quaternary structure, with a view to relating these 'soft mode' difference spectra to thermodynamic and kinetic data.

(d) Quasi-elastic studies of the mobility of one component in a heterogeneous aggregate of biomolecules relative to another, in cases where sufficient H/D contrast can be achieved either by selective in-vivo deuteration or by reconstitution of hydrogenous and deuterated components.

(e) 'Stroboscopic' probing of certain time-dependent processes in systems which can be excited periodically with \(\Delta t = 10^{-2}\) to \(10^{-4}\) s, by analysing spectral changes as a function of phase shifts between incident neutron pulses and externally applied sample stimuli.

The progress that can be made in each of these areas will depend on three factors: A reasonable share in the concerted use of high-performance instruments covering the \(Q,\omega\)-ranges required, substantial efforts to advance the interpretation of inelastic scattering from biomolecules by model calculations and simulation studies, and increased collaboration with biochemists specializing in the preparation of selectively or fully deuterated samples. Because of a somewhat fragmented, instrument-oriented approach to allocating beam time, the first of these factors will be of overriding importance for the future.

In 1975, during the first Brookhaven Symposium on bio-neutron work /3/, Egelstaff's discussion of the possible biological applications of inelastic scattering led to the conclusion that "the next stage is up to the biologists". Now that the results of about a dozen initial studies have been published and several more ambitious projects are well under way, the conclusion must be that it is up to
the neutron scattering community to recognize fully the scientific potential of inelastic scattering for the molecular dynamics of proteins, nucleic acids, and composite systems, and to enable a wider group of molecular biologists interested in time-dependent processes to take advantage of the many opportunities for innovative work existing in this field. It will be essential to sustain a rate of development commensurate with the general growth in the use of advanced spectroscopic techniques in biomolecular research, especially at a time when an integrated picture of a number of important processes based on thermodynamic, kinetic, and molecular dynamics data is beginning to emerge.

ACKNOWLEDGEMENT
This work is being supported by the UK Science and Engineering Research Council.

REFERENCES

/15/ Biophysics of Water (FRANKS, F., MATHIAS, S.F., Eds), Chichester, Wiley (1982). 
Invited Paper

MORPHOLOGY AND STRUCTURE OF BIOLOGICAL SYSTEMS

H. B. STUHRMANN
Institute of Physical Chemistry,
University of Mainz,
Federal Republic of Germany

Abstract

MORPHOLOGY AND STRUCTURE OF BIOLOGICAL SYSTEMS.

About half the atoms of living cells are hydrogens and nearly all biological applications of neutron scattering rely on the well-known difference in the scattering length of the proton and deuteron. Contrast variation in $H_2O/D_2O$ mixtures is often used to differentiate between various moieties of a macromolecule. In lipoproteins, for instance, a mixture of 40% $D_2O$ in $H_2O$ matches the scattering density of the protein moiety, rendering visible to neutron scattering the lipid structure only. This technique has become particularly important in investigating the interaction of proteins with nucleic acids in viruses, chromatin and ribosomes. Another wide class of applications is the molecular structure and function of membrane proteins. Specific labelling by isotopic substitution is a still more sensitive technique. Its full power can only be exploited by polarization variation of proton spins using intense polarized neutron beams as they are now provided by reflection from magnetized supermirrors.

INTRODUCTION

Of the many techniques that have been used to study biological structures, the use of thermal neutrons has been one of the more recently applied. Within the last 15 years since 1969, neutron beams have been shown to provide useful information about the properties of subcellular structures. The emphasis is mainly on small-angle scattering experiments. More than 80% of the biological projects submitted to the Institute Max von Laue - Paul Langevin at Grenoble fall into the category of contrast variation. The majority of small-angle scattering experiments has been carried out at the ILL instrument D11. It is useful to remember the basic concept of the instrument (Fig.1).

Neutrons released from the fuel element are thermalized in heavy water. They are further cooled down by liquid deuterium and reflected into the small-angle camera by a 28 m long bent guide tube (horizontal width 3 cm, radius 2700 m) at grazing incidence which eliminates $\gamma$-radiation and fast neutrons. The resulting wavelength distribution is centred at 6 Å wavelengths.
FIG. 1. Small-angle camera D11 at the high-flux reactor of the Institut Max von Laue–Paul Langevin in Grenoble.

approximately. The spectral distribution is further narrowed to 9% full half width by a helical slot selector which can be tuned to any wavelength between 4 and 25 Å just by changing its rotatory speed. The beam is collimated in a path the length of which can be changed from 2 to 40 m by introducing of straight neutron guide sections. The typical sample diameter is 20 mm. The detector is a boron trifluoride containing multielectrode chamber with gas amplification. Its sensitive area consists of 64x64 channels, the cross-section of one being 1 cm². The direct beam is caught by a rectangular beam stop (3x5cm) in front of the area detector.

Small-angle scattering is mainly elastic scattering. It is therefore characterized by the change of the flight direction of the incident neutron only. The momentum transfer \( Q \) is simply related to the wavelength \( \lambda \) and the scattering angle \( 2\theta \) by

\[
Q = \frac{4\pi}{\lambda} \sin \theta
\]

Time-of-flight (TOF) analysis allows the identification of neutrons with different wavelengths. As TOF analysis can be done at any small angle \( 2\theta \) small, low-Q scattering is performed in a more efficient way at a pulsed neutron source. A broader interval of the wavelength spectrum can be used and its monochromatization can be handled in a very flexible way. The gain of low-Q scattering at the planned spallation neutron source SNQ, with respect to the instrument D11 operated at a source with equal mean flux, depends on the distance of the detector from the source and the frequency at which thermal or cold neutrons are emitted (Fig. 2).
If a wavelength resolution better than 9% is needed, then TOF analysis of pulsed neutrons meets this requirement easily. If samples are very small, then a good angular resolution can be obtained at a relatively short distance between the sample and detector. The reduction of the flight path would allow the use of a larger portion of the spectrum [1].

**BASIC FEATURES OF LOW-Q SCATTERING**

The central objective of scattering studies on biological macromolecules is to determine molecular conformations and arrangements, particularly those which may contribute to specific functions. Usually this must be done from a very limited amount of scattering data and so independent methods of analysis are valuable. The principles of neutron scattering are very similar to those of X-rays, but some important differences exist.

If the scattering specimen is a single macromolecule with no regular separations between neighbouring structures, then the scattering is continuous. The amplitude of the scattered radiation is de-
terminated by the scattering density distribution $\varrho(\mathbf{r})$ of the macromolecule according to the Fourier relationship

$$F(\mathbf{s}) = \int \varrho(\mathbf{r}) e^{-2\pi i \mathbf{s} \cdot \mathbf{r}} d^3r$$  \hspace{1cm} (2)$$

The Fourier inverse of this expression is

$$\varrho(\mathbf{r}) = \int F(\mathbf{s}) e^{2\pi i \mathbf{s} \cdot \mathbf{r}} d^3s$$  \hspace{1cm} (3)$$

where $\mathbf{s} = \mathbf{Q}/2\pi$. It gives a three-dimensional coherent scattering density distribution from the values $A(\mathbf{s})$, which must be determined from the scattering experiment. The problem of phase determination results from only being able to measure the magnitude of $A(\mathbf{s})$, since the measured intensity $S(Q)$ is proportional to $|F(Q)|^2$.

Random orientation of the macromolecule results in averaging the scattering intensity $S(Q)$. This process is more easily understood by developing $\varrho(\mathbf{r})$ as series of spherical harmonics

$$\varrho(\mathbf{r}) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} \varrho_{\ell m}(r) Y_{\ell m}(\omega)$$  \hspace{1cm} (4)$$

The scattering function of a dilute solution of macromolecules then is:

$$I(Q) = 4\pi^2 \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} |F_{\ell m}(Q)|^2$$  \hspace{1cm} (5)$$

where $F_{\ell m}(Q) = (2/\pi)^{3/2} i^l \int \varrho_{\ell m}(r) J_l(Qr) r^2 dr$  \hspace{1cm} (6)$$

The striking feature of Eq. (5) is that the scattering $|F_{\ell m}(Q)|^2$ of each multipole $\varrho_{\ell m}(r) Y_{\ell m}(\omega)$ adds independently to the total scattering $I(Q)$. Independent rotation of

$$\varrho_{\ell m}(\mathbf{r}) = \sum_{m=-\ell}^{\ell} \varrho_{\ell m}(r) Y_{\ell m}(\omega)$$  \hspace{1cm} (7)$$

does not affect the scattering function, although it does change the structure (2). Recently a new algorithm was discovered by Svergun, Feigin and Schedrin (3) which allows the separation of the scattering functions of $\varrho_{\ell m}(\mathbf{r})$. The same authors have used this method for the structure determination of T phages which have cylindrical symmetry ($m = 0$).

In writing the above relationships, the structure was expressed as the density of coherent scattering lengths. For X-rays, the scattering centres are the electrons of the atoms. Every electron has the same X-ray coherent scattering length ($= 0.28 \times 10^{-12}\text{cm}$ for $\theta = 0$). Consequently, $\varrho(\mathbf{r})$ is just the familiar electron density of the structure. For neutrons, the interaction with atoms is essentially described by the scattering amplitude operator $A(4)$:

$$A = b + 2 A \mathbf{\mathbf{M}} \cdot \mathbf{s} + 2 B \mathbf{\mathbf{I}} \cdot \mathbf{s}$$  \hspace{1cm} (8)$$
The first and the third term take into account the interaction of the neutron with the atomic nucleus, \( b \) is the isotropic nuclear scattering length. The third term describes the interaction of the neutron with spin \( s \) with nuclei having the spin \( I \).

The second term corresponds to dipolar interaction between the magnetic moment of atomic electrons. \( M_\perp \) is the component of the magnetization perpendicular to the vector momentum transfer \( \mathbf{Q} \). The magnetic scattering length of an atom is the product of the number of its Bohr magnetons multiplied with \( 0.27 \times 10^{-12} \) cm.

\[ b \text{ and } B \text{ can be expressed by } b \text{ and } b^* \text{. These are the scattering \text{ lengths which correspond to coupling of } I \text{ and } s \text{ to } I + 1/2 \text{ and } I - 1/2 :} \]

\[ b = \frac{(I + 1) b^{(4)} + I b^{(-)}}{2 I + 1} \quad B = \frac{b^{(4)} - b^{(-)}}{2 I + 1} \quad (9) \]

Example: an assembly of isotopes of the same kind with nuclear spin \( I \) of diamagnetic atoms gives coherent forward scattering

\[ I_{coh} = |F|^2 = b^2 + b^* b + 2 b B \mathbf{I} \cdot \mathbf{P} \cdot \mathbf{n} \quad (10) \]

and incoherent scattering

\[ I_{inc} = B^2 I(I+1) - B^* \mathbf{P} \cdot \mathbf{P} - b^* B \mathbf{I} \cdot \mathbf{P} \cdot \mathbf{n} \quad (11) \]

where \( \mathbf{P} \) and \( \mathbf{n} \) are unit vectors describing the direction of polarisation of the target and the incident neutrons, respectively. In the case of hydrogen (protons) with \( I = 1/2 \) we have

\[ b^{(4)} = + 1.085 \times 10^{-12} \text{ cm} \quad b^{(-)} = -4.74 \times 10^{-12} \text{ cm} \quad (12) \]

and therefore

\[ b = -0.375 \times 10^{-12} \text{ cm} \quad B = 2.92 \times 10^{-12} \text{ cm} \quad (13) \]

There is hardly any regularity of the nuclear scattering amplitudes as there is with electrons. This is because neutron scattering is the result of nuclear forces and is dominated by a process known as resonance scattering. Some nuclei (e.g. \(^{149}\text{Sm}, \quad ^{133}\text{Cd}, \quad ^{157}\text{Gd}) exhibit a strong wavelength dependence (dispersion) at thermal neutron energies. As is shown in Fig.3 there is a change of the magnitude and phase of the scattering length \( b \) near the resonance energy which is taken into account by adding the dispersion of the real \( b' \) and the imaginary \( b'' \) to the constant \( b_0 \).

\[ b(\lambda) = b_0 + b'(\lambda) + b''(\lambda) \quad (14) \]

\( b' \) is an order of magnitude larger than \( b_0 \), and it assumes both negative and positive values. A consequence of this is that the scattering length of nuclei may be negative even at energies far away
from the resonance energy, e.g. $b'(\lambda)$ of hydrogen (Eq.12). The dispersion of resonant (or anomalous) X-ray scattering, especially at many $L_3$-absorption edges, compares to that of neutron scattering by resonant nuclei (Fig. 3). The dispersion of the imaginary part $b''$ is related to the cross-section of total absorption $6(\lambda)$ by the optical theorem:

$$6'(\lambda) = 2\lambda b''(\lambda) \quad (15)$$

As the scattering lengths vary strongly among different nuclei, isotopic substitution provides a direct means of isomorphous changes of the scattering amplitudes. The large difference between the coherent scattering lengths of the proton and the deuteron allow a wide variation of scattering length at the site of a bound hydrogen atom:

$$b = (-0.374 + 1.04 X) 10^{-12} \text{ cm} \quad (16)$$

where $X$ is the occupancy by $^2H (=D)$. It is excelled by the spin dependence of the scattering length of the proton:

$$b = (-0.374 \pm 1.444 P) 10^{-12} \text{ cm} \quad (17)$$

where $P$ is the degree of polarization of the proton spins. $P$ is positive if the neutron spin is parallel to the proton spin and it is negative in the case of antiparallel spin orientation. Polarization changes the scattering length of the proton three times more efficiently than isotopic substitution.
CONTRAST VARIATION

The easiest way of isomorphous replacement in a biological system is to diffuse D\textsubscript{2}O into its aqueous phase. The effect on the neutron scattering will be tremendous. It is particularly informative in the case of those macromolecules which consist of two different components, like nucleic acids and proteins in viruses (5), chromatin (6) and ribosomes (7). But also other systems like carbohydrate-protein macromolecules (8), and lipoproteins (9) were rewarding subjects of contrast variation in H\textsubscript{2}O/D\textsubscript{2}O mixtures.

In dilute solutions the neutron scattering intensity originates from the difference between the scattering density distribution $\rho(\vec{r})$ of the dissolved particles and the density of the solvent.

$$\rho(\vec{r}) = \rho_{\text{solute}} - \rho_{\text{solvent}} \quad (18)$$

The solvent density has been assumed to appear uniform at low Q. A similar equation holds for the mean values

$$\bar{\rho} = \bar{\rho}_{\text{solute}} - \bar{\rho}_{\text{solvent}} = \text{CONTRAST} \quad (19)$$

At vanishing contrast, $\rho_c(\vec{r})$ describes the deviations of the density distribution of the dissolved particle $\rho(\vec{r})$ from its mean value. Consequently, there is no coherent scattering of the particle at zero angle, and only relatively weak scattering is encountered at wider angles. It is due to the internal structure, e.g. the mutual arrangement of lipids and proteins in lipoprotein.

On decreasing the scattering density of the solvent, the excess scattering density of the particle with respect to the solvent increases everywhere inside the particle boundaries. The added contrast is taken into account by

$$\rho(\vec{r}) = \rho_s(\vec{r}) + \bar{\rho} \rho_c(\vec{r}) \quad (20)$$

where $\rho_c(\vec{r}) = 1$ everywhere inside the particle volume and it is zero elsewhere. It is the product $\bar{\rho} \rho_c(\vec{r})$ which describes the uniform increase of the excess scattering density $\rho(\vec{r})$. The idea of contrast variation was first developed in crystallographic studies on haemoglobin by Bragg and Perutz (10).

With neutron scattering in H\textsubscript{2}O/D\textsubscript{2}O mixtures, $\rho_c(\vec{r})$ and $\rho_s(\vec{r})$ need a slightly different interpretation. About one quarter of the hydrogen atoms in proteins may dissociate and are therefore readily exchanged by deuterium. The variation of the contrast therefore is somewhat smaller than the change in the density of the H\textsubscript{2}O/D\textsubscript{2}O mixture. This is taken into account by $\rho_c(\vec{r}) < 1$. In fatty acid domains no hydrogen exchange will occur ($\rho_c(\vec{r}) = 1$) whereas polar regions are described by $\rho_c(\vec{r}) \sim 0.7$. $\rho_s(\vec{r})$ however, represents the internal structure with the hydrogen atoms partly exchanged by deuterium according to the H\textsubscript{2}O/D\textsubscript{2}O ratio of the solvent at buoyancy.
Contrast variation by polarization of the proton spins of a biomolecule embedded in \( D_2O \) does not suffer from H/D exchange in the above sense, i.e. \( \rho_c(\vec{r}) \) is strictly unity. As the polarization of proton spins is catalyzed by paramagnetic centres, inhomogeneities in \( \rho_c(\vec{r}) \) may occur at lower degree of polarization (11).

With this in mind we can start to analyze neutron scattering. As the intensity is the square of the amplitude, its contrast dependence always assumes the following form (12):

\[
I(Q) = I_s(Q) + \rho I_{cs}(Q) + \bar{\rho}^2 I_c(Q) \tag{21}
\]

One of the basic scattering functions, \( I_s(Q) \), has already been discussed, as it is the scattering function of \( \rho_s(\vec{r}) \). \( I_c(Q) \) is the scattering function of the volume excluded to the solvent and \( I_{cs}(Q) \) is a cross term originating from the convolution of the macromolecular shape \( \rho_s(\vec{r}) \) with the internal structure \( \rho_s(\vec{r}) \). The measurement of neutron scattering in at least three different \( H_2O/D_2O \) mixtures provides the three basic scattering functions.

It is interesting to note that polarization variation allows the direct measurement of each of the basic scattering functions. Flipping the spin of a polarized neutron beam from parallel to antiparallel orientation to the proton spins of the target changes the sign of the cross term. The difference between these two types of measurement immediately yields twice the value of \( I_{cs}(Q) \). Any variation of the sum of both scattering intensity measurements is due to \( I_c(Q) \) only.

The analysis of \( I_c(Q) \) is most promising. The known family of possible structures belonging to a one-particle scattering curve can be greatly reduced, if the assumption of a shape model can be made. Using a multipole expansion and restriction to spherical harmonics \( Y_{l,m} \) with \( l = 0,1,2 \) and 3, a rather detailed model has been found in the case of the large ribosomal subunit of E.coli ribosomes (13). It agrees remarkably well with the model from observations in an electron microscope (Fig.4).

In a first approximation the analysis of the internal structure relies on the evaluation of the quadratic terms of the power series of the basic scattering functions. It defines the contrast dependence of the radius of gyration \( R \) of the dissolved particle.

\[
R^2 = R_e^2 + \alpha / \bar{\rho}^2 - \beta / \bar{\rho}^2 \tag{22}
\]

where

\[
\alpha = \int \rho_s(\vec{r}) \bar{r}^2 d^3r / V
\]

\[
\beta = \int \int \rho_s(\vec{r}) \rho_s(\vec{r}') \bar{r} \bar{r}' d^3r d^3r' / V
\]

and

\[
V = \int \rho_s(\vec{r}) d^3r
\]
FIG. 4. Shape of the large sub-unit of \textit{E. coli} ribosomes.

FIG. 5. Dependence of the square of the radius of gyration on the square of the reciprocal of the contrast $\tilde{\beta}$. The results from the large ribosomal sub-unit and of two of its derivatives are shown. They differ in the degree of deuteration of the protein moiety. In one derivative half of the protons of the ribosomal proteins are exchanged, in the other nearly all of them have been replaced by deuterium, the rRNA remaining native /15/.

This relation is widely used (7,8,9,15). $\alpha$ and $\beta$ reflect the lowest resolution terms of the monopole and dipole of $q_s(\tilde{\beta})$, respectively. With chromatin and water soluble proteins $\alpha$ is positive, whereas molecules with a high density core, like viruses, ribosomes etc., exhibit a negative $\alpha$ (Fig.5). $\beta$ is always positive.
This is a requirement of modern biological structure research which can best be demonstrated in the case of _E. coli_ ribosomes. To clarify some terms and the reasons for our engagement in this area, some of the properties of ribosomes are mentioned.

Each living cell contains ribosomes. This cell organelle translates the genetic information, which is coded as a sequence of four different nucleotides in nucleic acids (DNA or mRNA) into the sequence of twenty different amino acids of proteins. Each triplet of nucleotides (codon) corresponds to either an amino acid or to a processing signal for the ribosome. Most of the investigations on protein biosynthesis are performed on the ribosome of _E. coli_ bacteria. This ribosome, with a relative molecular mass of 2.6 million, can be separated into two unequal subunits. The small subunit consists of one ribosomal RNA and 21 different proteins (S1-S21), the large subunit of two (one short and a very long) rRNA molecules and 32 different proteins (L1-L34). The 55 components of the ribosome can be separated and reconstituted again to fully active ribosomes (14).

It is an accepted idea of structure research that specific staining of the sample helps to understand the pattern it gives. For structural studies on dissolved ribosomes the easiest way to identify the spatial distribution of rRNA and proteins is to diffuse heavy water D\(_2\)O into its aqueous phase. The contrast dependence shown in Fig. 5 clearly shows that the rRNA is closer to the centre of the particle than the ribosomal proteins.

This is a very rough description of the ribosomal subunit. More detailed information about the structure can be obtained from specific labelling of parts of the macromolecule. Ribosomal proteins lend themselves easily to specific staining. This is the basis for two different ways to elucidate the architecture of the ribosome.

**Label Triangulation.**

This method is has proved to be very suitable for the determination of distances between pairs of ribosomal proteins. From a large number of protein-protein distances the relative co-ordinates of the ribosomal proteins can be determined by triangulation. Following an idea of Kratky and Worthmann (17), Hoppe (18) and Engelman and Moore (19) independently proposed to measure a pair distance interference term, which is obtained as the difference of two scattering experiments using the following four equimolar solutions of the complex (Fig. 6).

The scattering \(I(1)+I(2)\), diminished by \(I(3)+I(4)\), yields the pair distance interference term arising from the vectors connecting volume elements in the one protein with the other exclusively. Following the reasoning of Hoppe (18) we verify this within the frame of the multipole expansion of \(I(Q)\) in Eq. 5. Let \(u(r)\) and \(v(r)\) be
the structures of two different ribosomal proteins in the ribosomal subunit \( w(r) \). The corresponding amplitudes are \( U(Q), V(Q) \) and \( W(Q) \). According to Eq.5 each multipole component may be treated separately. We therefore identify \( U(Q), V(Q) \) and \( W(Q) \) with an arbitrary multipole \( F_{\alpha \nu \mu}(Q) \) of the amplitude. Omitting \( Q \) and characterizing the degree of staining of the two proteins by \( a \) and \( b \) we obtain:

\[
I = \left[ I(1) + I(2) \right] - \left[ I(3) + I(4) \right]
\]

\[
= \left( aU + aV + W \right)^2 + \left( bU + bV + W \right)^2 - \left( aU + bV + W \right)^2 - \left( bU + aV + W \right)^2
\]

\[
= 2 \left( a - b \right)^2 UV
\]

At first approximation, this term can be interpreted as the distance \( d \) between the centres of mass between the two ribosomal proteins.

\[
I(Q) \sim \frac{\sin dQ}{dQ}
\]

On this basis the co-ordinates of nearly all proteins of the small subunit of \( E. \text{coli} \) ribosomes have been determined (20). Further analysis of Eq. (23) also allows the determination of possible asymmetries of individual protein shapes and their mutual orientation. A more direct approach to shape determination of ribosomal proteins relies on contrast variation.

Label contrast variation

This is the generalized form of contrast variation. The scattering of one or two proteins is measured as \( I_c(Q) \) in Eq. 21. Variation of the scattering length of the label yields the other two basic scattering functions, \( I_c(Q) \) and \( I_b(Q) \). It is the contrast of the labelled protein which has to be changed. As the boundaries
of the label do not coincide with those of the whole particle, measure­ments in various H₂O/D₂O mixtures would not be the right way to identify the scattering of the label. One way to perform contrast variation correctly would involve the synthesis of various ribo­osomes that differ in the contrast of their labelled proteins. This would be a prohibitively difficult task in most cases.

Nierhaus (21) therefore proposed to eliminate two basic scattering functions, $I_C(Q)$ and $I_S(Q)$. For this purpose any kind of long­range spatial fluctuation of scattering density - those of the labelled proteins excepted - had to be avoided (Fig. 8).
The scattering density of both rRNA and the ribosomal proteins was matched to that of D$_2$O. The labelled protein was added during the recombination of the appropriately deuterated components, rRNA and proteins, during recombination. In the same way the unlabelled large ribosomal subunit was prepared. The difference between the scattering of the labelled particles and the reference sample is considered as the scattering of the protein (Fig 8 and 9a). The validity of the assumptions concerning the invisibility of the reference particle could be shown by numerous neutron small-angle scattering experiments within the accuracy of the data shown in Fig. 8.

A complete study of contrast variation can best be achieved by polarization variation of the target. For this purpose the native
ribosomal protein is preferably (not necessarily) embedded in a fully deuterated matrix (Fig. 9). It happens that the scattering density of the fully deuterated rRNA nearly equals that of the deuterated proteins, i.e. this ribosomal subunit can be rendered 'invisible' to neutrons by using a solvent of slightly higher density than that of D$_2$O (e.g. D$_2$O : deuterated glycerol 3:1).

The contrast of the labelled protein is directly proportional to the polarization of its proton spins. Polarization variation unambiguously discriminates the scattering of the label $I_C(Q)$ in Eq. 20 against the other two basic functions. And it does it by extending the range of contrast considerably beyond the possibilities of isotopic substitution (Fig. 9). As is shown in Fig. 9a, the scattering from a labelled ribosomal protein in situ amounts to about 3% of the total scattering (from D$_2$O and background). Polarization variation increases this term to about 8% if $I_C(Q)$ is involved only, or even 30% if the model of the 'invisible' ribosome can be maintained. The precision of the scattering of the labelled protein may increase by one order of magnitude with proton spin polarization variation, not to mention the unambiguity of the results which is perhaps the significant advantage of polarization variation. Alternatively, one would need one to two orders of magnitude more beam time to obtain the same result with isotopic substitution.

The cross term is most easily measured, not only because it can be obtained directly as the "flipping ratio", but also because it exhibits relatively large scattering cross-sections (Fig. 9c). The contrast dependence of the apparent radius of gyration is mainly due to this term and it is a direct measure of the distance of the labelled protein from the centre of mass of the large ribosomal subunit. Adding the small ribosomal subunit will lead to a substantial shift of the centre of mass. Repeating the measurement in the new system would also fix the polar height of the labelled protein.

CONCLUSIONS

Neutron small-angle scattering will develop to a more powerful technique of macromolecular structure research, once the spin dependence of the coherent scattering length of protons can be used routinely. Intense polarized neutron beams and a high degree of polarization of the target are needed. Very efficient broad-band spin selectors (magnetic super mirrors) meet the requirements of small-angle scattering (22) perfectly. The dynamic polarization of proton spins in biological samples has hardly been started. The development of this technique would open new ways of molecular structure research in biology and materials sciences.
ACKNOWLEDGEMENT

The support of this work by the Bundesminister für Forschung und Technologie is gratefully acknowledged.

REFERENCES

Invited Paper

NEUTRON SCATTERING FROM POLYMERS IN THE 'NINETIES

R. S. STEIN
Polymer Research Institute,
University of Massachusetts,
Amherst, Massachusetts,
United States of America

Abstract

NEUTRON SCATTERING FROM POLYMERS IN THE 'NINETIES.

The technique of small-angle neutron scattering (SANS) has led to many advances in the understanding of the structure of polymer molecules, both in solution and in the solid state. Notable contributions are (a) characterization of single molecule parameters in dilute and concentrated solution and in the solid state, (b) the characterization of molecular interaction parameters in solutions under the above conditions, (c) the study of critical phenomena in phase separation, (d) the study of the chain morphology on aggregated systems such as networks and crystalline polymers, and (e) the characterization of deformation and orientation processes in solids on a molecular level. Future advances are expected to ensue from progress in instruments leading to higher flux, longer wavelength, and pulsed sources. These lead to possibilities for studies of molecular dynamics in concentrated systems. The SANS technique is expected to reach a new level of sophistication involving studies on polymeric systems with selective isotopic labelling.

INTRODUCTION

The SANS technique offers some special advantages to the polymer scientist. In addition to providing a source of radiation with wavelength considerably shorter than light but somewhat longer than x-rays, a great gain arises because of the very different scattering lengths for the different kinds of radiation. X-rays essentially ignore hydrogen atoms, a very important component of most polymeric structures, whereas hydrogen has a large scattering length for neutrons. In this respect, the techniques are complementary for structure determination.

Those acquainted with the light scattering technique know that an important experimental problem is to obtain systems that are optically clean. The scattering by a particle varies with the sixth power of its radius so that a few large particles may completely dominate the scattering. For dilute solutions it is necessary to carefully filter or centrifuge to remove dust particles. This difficulty has precluded extensive application of the technique to concentrated solutions or bulk systems. They are too difficult to clean. While the theory exists to interpret light scattering from concentrated solutions 1, little application has been made for this reason. While it is possible to use light scattering (or x-ray scattering 2) to study amorphous polymer blends, not much has been done.
Contrast for light scattering depends upon refractive index difference, while that for x-ray scattering depends upon electron density difference. These differences between polymers and their contaminants are not great. However if a polymer is extensively labelled with a highly scattering species such as deuterium, its contrast with respect to other components (other polymer or solvent) is usually greater than that of the other components with respect to the contaminant. Thus, removal of the contaminant is not necessary. Studies may then be conducted on concentrated systems or blends without employing the difficult cleaning process. Of course, problems result if the contaminants have high contrast (if they were deuterated or were voids).

An outstanding advantage of SANS is the possibility for isotopic labelling. X-ray scatterers have used this in the past through the introduction of heavy atoms into structures. A problem is that such atoms often perturb the system greatly, so that the labelled structure may substantially differ from the unlabelled. The replacement of hydrogen (H) in polymer molecules or structures with deuterium (D) changes scattering greatly without causing significant structural perturbation. This effectively makes possible the task of "coloring a molecule red" which was assigned to the author by his mentor, the late Professor Arthur V. Tobolsky, for his doctoral dissertation at Princeton University. The challenge was to be able to see an individual molecule in a condensed phase so as to be able to witness its changes during phenomena such as phase changes and deformation. Many less effective means for "coloring" molecules have been developed, but with SANS we have the best way to date to "color them red" with deuterium. This has led to much exciting work on concentrated solutions, blends, networks and gels, crystal topology, and orientation.

REVIEW OF PAST ACCOMPLISHMENTS

Many significant results have arisen from the application of the SANS technique to polymers during the past 10 years. Much of this has been reviewed so details will not be given here. With fear of omission, some notable achievements will be mentioned:

1. The Characterization of the Molecular Weight, Radius of Gyration, and Conformation of Single Molecules in Dilute and Concentrated Solution

Dilute solution molecular weight measurements are redundant with and agree with light scattering results, although the greater contrast possible with deuterated polymer (or solvent) makes possible the extension to lower molecular weights without employing excessive cleaning techniques. Because of the long wavelength of light, radius of gyration measurements have as the lower limit a few hundred angstroms. With extraordinary efforts, measurements have been extended to 50 Å. The lower wavelength of neutrons makes possible the extension of measurements down to a few angstroms.

Intermolecular interference prevents the use of light scattering for measurement of radius of gyration in concentrated systems. This may be done, however, with SANS through isotopic labelling of a portion of the polymer. While it was originally thought necessary that the labelled component be dilute, it has more recently been demonstrated that this is not necessary. Working with a high concentration of label has the obvious advantage of increasing contrast and decreasing data acquisition time.

Very limited information about internal conformation of polymer molecules may be obtained by light scattering. Because of the long wavelength, measurements are restricted to the low q region \( q = \frac{4 \pi}{\lambda} \sin (\theta/2) \). SANS allows extension to the intermediate and high q region where information about more local structure of the polymer chain is obtained. This permits Kratky plots of \( I(q)q^2 \) vs. q which should level off to a constant value at high q for a gaussian coil. Experiments show deviations from this at high q.
arising from non-gaussian separation of scattering elements on chains at small
distances. The theory for this effect has been approached by Yoon and Flory
using the rotational isomer approximation. They predict difference in
this region dependent upon chain microstructure, as occurs for example between
syndiotactic and isotactic poly(methyl methacrylate) as demonstrated by Kirste,
et al. 8.

An important contribution deals with the elucidation of the microstructure
of block copolymers using contrast matching. The theory for this was originally
worked out by Benoit 9 for light scattering, where mixed solvents were used
to match refractive indices of the two blocks. The problem was, however, that
by varying the solvent composition, its interaction with the polymer changed,
resulting in conformational perturbation. The approach with SANS proved much
better in that one of the blocks could be deuterated and contrast matching
could then be accomplished using mixtures of H and D containing species of the
same solvent 10. This results in negligible perturbation of
conformation.

2. The Extension of the Above Studies to Bulk Polymers and Polymer Blends

The measurement of molecular dimensions in bulk polymers has proved
evasive. Attempts by x-ray scattering on halogen end labelled molecules were
unsuccessful because of aggregation of the halogenated ends. Use of solutions
of D containing polymers in their H containing counterparts was outstandingly
successful, especially with the extension to higher concentrations of the D
containing component 11. This settled an important controversy in
polymer science and verified Flory’s contention that bulk polymers are theta
solvents for themselves in which molecules have their unperturbed conformations
12.

A more demanding test is to see whether local chain conformation in the
bulk corresponds to the unperturbed value. This proves to be the case. The
method has been applied to amorphous polymer blends where the solvent is
polymeric. Kirste 13 has demonstrated the application of the Zimm plot
14, developed for light scattering, to the determination of the
molecular weight, radius of gyration and second virial coefficient (to be
discussed below) to dilute polymer blends. Hadziioannou and Stein 15
have extended the measurements to concentrated blends and have also shown
16 how the Zimm plot technique may be extended to this regime.

3. The Determination of Interaction Parameters Between Components of Amorphous
Blends

Flory and Huggins 12 showed how the interaction parameter, $\chi$, may
be obtained from the second virial coefficient derived from osmotic pressure or
light scattering experiments. As previously mentioned, Kirste 13
demonstrated the extension of the technique to dilute solutions of polymer
blends, and Hadziioannou and Stein 15 applied a variation to
concentrated blends. In this case, the second virial coefficient is not
applicable, and the method gives $\chi$ directly. The method involves
extrapolation of scattering data to zero q. The results prove to be a special
case of a more general formulation by deGennes 17. In this way values
of $\chi$ as a function of temperature and concentration have been determined. A
variation of the procedure suggested by Koningsveld has been applied where
is a function of concentration 18.

4. The Study of Critical Fluctuations for Solutions

It is known that for low molecular weight systems, concentration
fluctuations of increasing size occur as a critical temperature is approached.
The theory for this has been developed by Ornstein and Zernike 19 who
show how one may obtain a correlation length from the q dependence of scattered intensity. Such techniques have been applied to polymer solutions, both in low molecular weights and polymeric solvents. Using their approach, or a modification due to deGennes, one may extrapolate to the temperature at which the correlation length becomes infinite, which may be identified with the spinodal.

This has been done, for example, for blends of deuterated polystyrene with poly(vinyl methyl ether) which exhibit lower critical solution temperatures. From the dependence of correlation lengths and intensities on degree of superheating, critical exponents may be determined which agree well with theoretical predictions. The advantage of using SANS for these studies arises not only from the greater contrast possible because of labelling but also as a result of the smaller correlation length that may be observed as compared with light scattering.

5. The Study of Early Stages of Phase Separation for Polymer Mixtures

The advent of phase separation for polymer mixtures is conventionally detected by observing light scattering cloud points, where the intensity of scattering appreciably increases when the temperature is shifted from a value in the miscible region to one in the immiscible region. The measurement of "neutron scattering cloud points" has recently been demonstrated. Values for cloud points for spinodal separation agreed well with those obtained by light scattering.

The kinetics of phase separation have been studied by observing the evolution of the q dependence of scattering with time. Initial stages have been shown to be spinodal and obey, when plotted in terms of reduced variables, the same kinetics as seen for low molecular weight systems. At later times, phase growth by Ostwald ripening and hydrodynamic processes is seen.

6. The Study of Screening Lengths and Distribution of Crosslinking Points in Network Polymers

The characterization of network heterogeneity through observation of the enhancement of light scattering upon swelling a network polymer was suggested by Stein and demonstrated by F. Bueche. More recently, its application to the characterization of rubber networks crosslinked by different procedures was described by Stein and Soni. They showed that swollen networks scattered more than uncrosslinked polymer solutions of the same concentration, and that the excess scattering characterized the network heterogeneity. These workers suggested that the same experiment could be done using SANS, thereby extending measurements to higher q values and observing heterogeneities at a smaller distance scale. Contrast is enhanced through swelling with a deuterated solvent. Thus, it is possible to bridge the gap in q between light and neutron scattering and observe heterogeneities ranging from micron to angstrom size scales.

Preliminary measurements have indicated structure at a level smaller than that of the mesh size of the network. Data has been interpreted in terms of the Debye-Bueche theory for scattering from inhomogeneous systems. Correlation functions are obtained by Fourier transformation and lead to correlation lengths in the 10-20 Å range. This has been interpreted in terms of a "correlation hole" associated with partial exclusion of solvent in regions close to the crosslink. Further experiments are necessary to confirm this assignment. Larger correlation distances are found to be related to the spacings between these correlation holes and may prove to be a means of measuring distances between "spatial neighbors" in a network (as contrasted with "topological neighbors" as studied using labelled chains). Data may be interpreted using the concept of screening lengths in networks as proposed by deGennes.
7. The Study of Chain Orientation and Relaxation of Polymer Molecules in Stretched Amorphous Polymers

The anisotropic SANS arising from stretched rubbers has been studied by Han, et al \(^{32}\) in an effort to test rubber elasticity theory. The purpose was to decide between two different theories of deformation of rubber networks, the affine deformation of crosslinks model and the phantom network model. The results were somewhat ambiguous, partly because of the limited precision of the measurements, but to a great extent because of the ill-defined character of the network (as discussed below).

Experiments were also carried out to test the affine assumption utilized in theories of swelling of rubbers such as that by Flory and Rehner \(^{12,33}\). Preliminary measurements \(^{31}\) indicate that the increase in the radius of gyration of network chains is less than the cube root of the increase in volume upon swelling as predicted by the affine theory. This result is still not understood. Either the experiments or their interpretation is in error or swelling theory must be modified.

Studies have also been carried out to observe the anisotropy of radius of gyration occurring upon deforming linear (uncrosslinked) polymers. Samples of high molecular weight (500,000) atactic polystyrene were oriented by capillary extrusion using the technique of Porter, et al \(^{34}\). The extrudate was rapidly quenched to below its glass transition temperature \((T_g)\) so as to freeze in its orientation. Guinier plots were then made for \(I(q)\) in planes parallel to and perpendicular to the extrusion direction so as to determine the values of \(R_g\) in these two planes \(^{35}\). The data were consistent with the model of affine transformation of chain ends up to the highest draw ratio measured \((10X)\). However, for low molecular weight \((50,000)\) samples, the change in \(R_g\) was less than predicted by the affine model. If the high molecular weight sample was heated to a temperature above its \(T_g\) for a short time and then quenched again, its \(R_g\) was found to decrease toward its undeformed value.

Such studies can be carried out in real time (without quenching) provided sufficient neutron flux is available. By measuring the change with time of the entire \(I(q)\) vs. \(q\) curve, it is possible to follow the evolution of molecular conformation accompanying relaxation \(^{36,37}\). It provides an excellent way to test theories of chain relaxation in the solid state such as that of "reptation" \(^{38-39}\). Preliminary attempts have been made to measure SANS under dynamic (oscillatory strain) conditions \(^{40}\), and future studies of these phenomena using this approach appear promising.

8. The Characterization of Chain Topology of Isolated Polymer Single Crystals and Solid Semi-Crystalline Polymers

Chain topology in crystalline polymers has been controversial \(^{41}\). Early concepts of polymer crystals envisioned the "fringed micelle" model in which chains passed through polymer crystals into the amorphous phase and then back into another crystal. A negligible fraction of these amorphous chains was believed to return to the same crystal. Then, based upon consideration of conservation of density at the phase boundary, Flory proposed that a reasonable fraction (of the order of a half) returned to the same crystal \(^{42}\).

With the discovery of polymer single crystals grown from dilute solution \(^{43-45}\), the concept of chain topology was revised. Because of these crystals being thin (about 100 Å) as compared with the molecular chain length; and because of their regular shape and growth surfaces that appeared to be of crystallographic origin, it was proposed that the emerging chains regularly re-entered the crystal in positions adjacent to where they emerged. It was postulated that this morphology persisted in the solid state, and that the lamellae observed by electron microscopy and small angle x-ray scattering
(SAXS) could be identified with these single crystals. The extreme view was that the majority of chains emerging from such lamellae returned through adjacent re-entry and that only occasional chains (tie chains) interconnected crystals. This concept led to a very different picture of the amorphous phase than that of a two-phase model in which the morphology of amorphous regions of crystalline polymers was considered to be identical with that of completely amorphous polymers.

In an attempt to decide among these concepts, techniques for establishing chain topology in semicrystalline polymers have been employed. One of these pioneered by S. Krimm involves studying infrared spectra of mixed crystals of H and D containing polyethylene. SANS studies on such systems have been extensively employed, although their interpretation has been controversial. It is generally agreed that the value of $R_s$ for molecules in solid state crystalline polymers is close to that in the amorphous melt, although there is a difference in the case of solution grown single crystals. Fischer has referred to this as the "solidification model" in which the large-scale dimensions of the chain are preserved during crystallization as a consequence of kinetic considerations.

Crystallization involves rearrangement of smaller sections of chains, best examined by SANS in the intermediate and larger $q$ regions. Such studies and their comparison with models generated by Monte-Carlo simulations form the basis of studies by Yoon and Flory and Gutman, et. al. The conclusions differ because of differences in the models associated with the density gradient at the crystal-amorphous interface. Experiments also offer difficulties because of problems associated with segregation of the D containing polyethylene during crystallization which occurs because of the appreciable difference in melting point from the H containing polyethylene. This problem is less serious for other polymers such as isotactic polystyrene and polypropylene.

In any case, it appears that the truth lies somewhere between the two extremes of random re-entry and adjacent re-entry, probably being closer to the former for bulk crystallized polymers and to the latter for polymers crystallized from dilute solution. The topology undoubtedly depends upon crystallization conditions, with more randomness associated with more rapid crystallization. Efforts have been made to account for this in terms of kinetic concepts. The ideal is to be able to classify the amorphous phase into chain types such as chain folds of varying length and adjacency, tie chains and chain ends. This has not yet been achieved, but there is hope that combination of SANS with other techniques (NMR, IR and raman spectroscopy) may lead to a solution.


As discussed above, the SANS from semi-crystalline polymers arises from chains, portions of which lie in both crystalline and amorphous regions. Thus, changes in SANS occurring on stretching such systems must be explained in terms of the changes in orientation and morphology of both of these regions. Preliminary studies of changes in SANS on drawing crystalline polymers in the crystalline state indicate that the change in $R_s$ is less than affine. There is no reason to expect that it should be, considering the two phase contributions to the process. The interpretation of the SANS results requires model building in which changes in the two regions are separately considered. Two cases are apparent:

(a) At small elongations, the crystalline and amorphous regions preserve their integrity, so that changes may be interpreted in terms of separate processes occurring in the crystalline and amorphous phases. It is evident that these cannot be uniquely characterized in terms of data.
obtained from SANS alone. Thus it is desirable to introduce information relating to crystalline orientation obtained by wide angle x-ray diffraction (WAXD) and lamellar orientation obtained from SAXS. From this information, it is possible to calculate the crystalline contribution to SANS.

The amorphous contribution depends upon the nature and connectivity of the amorphous chain segments with the crystals (as discussed above), and in the manner in which the amorphous layers between crystals change their orientation and size. Preliminary attempts to model this process have been made by Petraccone, et. al. It is useful to include information obtainable by birefringence and infrared dichroism to characterize changes in amorphous orientation and conformation.

(b) At higher elongations, where there is an interchange of chains between crystalline and amorphous regions, is more difficult to treat. It is known that at higher elongations, WAXD and SAXS indicate changes in size and shape of crystals. In this regime, microscopy and small angle light scattering (SALS) show a transformation from spherulitic to fibrillar morphology. It is not clear whether this process occurs by melting and recrystallization or by plastic deformation of crystals without melting. It is quite evident that model building to describe these processes will require parameters obtained through a variety of other experiments.

10. The Study of the Morphology of Block Copolymers in Solution and in the Solid State

It has been indicated above that SANS may be used to characterize the chain structure of isolated block copolymer molecules. In concentrated solutions or the bulk such systems may phase segregate, leading to ordered structures which may be idealized as having spherical, cylindrical, or lamellar morphology, dependent upon the molecular weight of the blocks. The chain conformation within the domains is determined by the size and shape of the domains, the necessity of a given block to primarily reside within one of the domains, chain connectivity between blocks, and maintenance of constant density. As with crystalline polymers, chain conformation in concentrated solution or the bulk has been studied by SANS measurements where one of the blocks is partly substituted with D. Considerations analogous to those for crystalline polymers apply to deformation of phase segregated block copolymers. Some such measurements have been reported.

11. Studies of the Dynamics of Single-Chain Molecules in Solution

For scattering from moving molecules, there may be loss or gain of momentum of the neutron with a consequent wavelength shift from that of the incident neutron beam. This observation, as with quasi-elastic (QE) light scattering provides information about molecular dynamics. Analysis of the momentum distribution of scattered neutrons with appropriate precision is difficult, but may be accomplished using spin-echo techniques, as reported, for example, by Higgins. The analysis of the time structure of scattered neutrons using pulse sources appears promising.

While QE-SANS is experimentally difficult, there are certain advantages as compared with QELS that justify its application. The larger q which is achievable makes possible the study of the dynamics of smaller regions of the molecule. Selective isotope labelling of portions of molecules makes possible the selective study of certain dynamic modes of motion.

12. Use of SANS to follow Chemical Reactions involving Polymers

It is usually easier and cheaper to employ conventional techniques to follow the course of chemical reactions. However there have been cases where SANS has been very useful:
(a) *Studies of Trans-Esterification of Polyesters*: There is indication that blends of poly(ethylene terephthalate) (PET) and poly(butylene terephthalate) are miscible in the melt. However, within a few minutes they tend to trans-esterify, leading to a copolymer. A similar problem is observed when one attempts to measure single molecule properties of PET in the melt by mixing the H-containing and D-containing species. Trans-esterification leads to the formation of a copolymer containing a mixture of H and D. Ultimately, all the molecules become identical and contrast is lost. A study of the consequent decrease in intensity and its q dependence with time serves to characterize the course of the reaction.

(b) *Studies of the Formation of an Epoxy Resin*: Composites, which are of rapidly growing commercial importance, contain a filler (glass fibers, kevlar, etc.) in a polymeric matrix, which is often an epoxy resin. Since the resulting polymer is crosslinked and intractible, parts are usually fabricated by polymerizing the monomer in the form of the finished product. The polymerization reaction is complex and often involves phase separation. Recent experiments by Wu have involved carrying out such reactions using D labelled monomers in the SANS apparatus to follow the course of the reaction.

13. *Studies of the Self-Diffusion Coefficient of Polymer Melts*

The measurement of the diffusion coefficient of a liquid into another liquid is often accomplished by establishing a sharp interface between the two liquids and following the concentration gradient across the interface as a function of time. This may be done, for example, by observing the refractive index gradient. For measurement of the self-diffusion coefficient of a polymer melt, there are two problems: (a) Diffusion coefficients for polymeric melts are low so long times are required to establish a macroscopic concentration gradient, (b) both polymers are identical so techniques like refractive index will not work for observing the concentration gradient. Here SANS comes to the rescue! A sandwich of alternate thin layers of H-containing and D-containing species of the polymer is made. If the layers are appreciably thicker than the wavelength of the neutrons in a SANS experiment, scattering arises only from density fluctuations within the layers and these are small. When diffusion occurs, there is a mixing of components so scattering increases due to the onset of concentration fluctuations. The intensity can be appreciable because of the contrast provided by the D. For thin layers, distances are small so that times for diffusion are relatively short. Such measurements have been made for polyethylene by Stamm at Julich and Graessley at Exxon.

**LIMITATIONS OF EXISTING TECHNIQUES FOR POLYMER STUDIES**

Limitations of current techniques are primarily related to problems of (a) low flux, (b) inadequate resolution, (c) detection problems and (d) sample preparation. As compared with standards of light and x-ray scattering, fluxes for SANS are dreadfully low. The advent of the laser led to a flux increase of from $10^3$ to $10^6$ over conventional light sources with an accompanying improvement in collimation, monochromaticity and coherence, leading to the feasibility of new classes of experiments. Corresponding gains are achieved by employing synchrotrons for SAXS. With SANS, the situation is similar to that of the pre-laser days for light scattering. Long times are required for data acquisition, even with relatively poor collimation and monochromaticity.

Prospects for increasing fluxes with steady state reactors lead to estimates of a 50-100X change. The situation may be brighter for pulsed sources and a few orders of magnitude gain may be possible. This not only leads to decrease in data gathering time for conventional experiments, but also leads to the possibility of achieving better resolution through better collimation.
Also, time resolved experiments in which changes of systems are observed in real time become feasible.

As mentioned previously, it is possible to close the gap in values of $q$ attained in light scattering and SANS experiments at present. However, it remains desirable to extend the $q$ range for SANS to lower values overlapping the light scattering region so as to make use of the isotope substitution techniques in this region. For example, values of radii of gyration of molecules soon exceed 1000Å on deformation, so that indirect means must be used to follow molecular extension processes.

With conventional equipment the minimum $q$ achieved is related to the path length in the apparatus and the linear resolution of the detector. It is not feasible to construct apparatus appreciably bigger than those presently available. There appears to be a possibility for solid state detectors with greater linear resolution. Alternatively, one may turn to different apparatus design, such as the Bonse-Hart type. This requires point-by-point counting, but times for this will be reduced with higher fluxes.

I believe the greatest limitation for SANS experimentalists is the securing of suitable samples. To take proper advantage of the power of SANS and the isotope substitution technique, one should have samples which are selectively labelled in designated places. The investment for SANS experiments is great enough that the samples employed should be well characterized with respect to molecular weight, tacticity, etc.

The synthesis of such samples for SANS usually involves routine techniques of organic chemistry. They can be tedious and time consuming, since the preparation of a suitable polymer often first involves the synthesis of a suitably labelled monomer starting from simpler compounds containing the label. The problem is to a large extent an organizational one of how to bring the skills of a qualified organic chemist to bear on this need of the SANS practitioner.

In "crystal gazing" concerning the future of SANS studies of polymers, I shall assume that solutions will be found to the above problems, and will now explore what additional things might be done.

**FORECASTS FOR THE 90's**

The following are expected to be some of the more significant developments in SANS during the next fifteen years:

1. **New and upgraded SANS facilities** will come on line in the U.S., Japan and Europe. Polymer scientists will be more prone to use the technique with the increasing availability of beam time at convenient locations. The research style of going to central facilities will be more accepted as polymer scientists also go to centers to use synchrotrons, super-electron microscopes, surface analysis equipment, etc. In addition to higher flux steady state sources and more general use of cold neutrons, pulse sources will become more prevalent and will be used by those who have need for exploiting the time structure of the beam or those working under difficult experimental conditions (like high pressure) where it will be easier to scan $q$ by exploring variation of intensity with wavelength (using time-of-flight techniques) than with angle. Solid state detectors will be developed leading to higher linear resolution and permitting better $q$ resolution and/or more compact equipment.

2. **Equipment will become more "user friendly".** Changes in sample-to-detector distance and changes in beam guide configuration will be more easily accomplished. Apparatus will be interfaced with faster and larger computers permitting more sophisticated real-time data analysis and comparison.
between experimental results and theoretical models. Real-time optimization of theoretical parameters through difference minimization techniques will become possible, much as is now done in structure refinement in x-ray crystallography. Absolute calibration techniques will be improved and equipment techniques will become available, so that parallel measurements on the same sample by SANS, SAXS, and SALS will become readily possible. This will permit studies over a wide range of \( q \) using different contrast weighting factors.

(3) Increasing cooperation will ensue between SANS practitioners and synthetic chemists so as to generate polymers which are selectively deuterated at particular locations. These will permit more sophisticated interpretations of SANS results, so that detailed conformational data for many polymeric systems will become available. With increasing information about conformational properties of single molecules and with increasing information about interaction between molecules, modelling of aggregated systems will become better.

(4) Higher fluxes will make possible the study of relaxation processes in real-time of isolated molecules in solution, polymer melts, and crystalline and liquid crystalline systems. Faster data acquisition and computing will facilitate the compilation of \( I(q) \) vs. \( q \) data and direct comparison with dynamic theories. Measurements will be carried out under oscillatory strain, acquiring data (a) using pulsed neutron sources where samples are oscillated at a frequency equal to the pulse rate and where the phase angle between the pulse and the strain is varied, or (b) using steady state sources where a Fourier analysis of scattered intensity with respect to strain is carried out in a manner similar to what is now used for dynamic WAXD and dynamic SAXS. In addition to mechanical strain, the effect of other varying fields will be explored. For example, the effect of oscillating electrical fields on the response of a piezo-electric or polar liquid crystalline polymer will be of interest.

(5) Higher fluxes will make feasible the study of phase changes in real time for processes such as polymer crystallization or phase separation by amorphous blends, much as is now being carried out by WAXD with or without the use of synchrotron or by light scattering.

(6) The types of studies now being carried out to determine chain topology of semi-crystalline polymers will be extended to include the study of liquid crystalline and other partly ordered systems. Polymers will be synthesized possessing mesogens and flexible spacers in which one or the other will be labelled. The nature of crystal defects will be explored where potential defect sites are labelled. For example, studies will be carried out on linear low-density polyethylene where the side groups are labelled so that their inclusion in the crystal and location in the morphology might be explored. The orientation and relaxation of such defect-containing systems will be of special interest.

(7) Studies will be conducted on high modulus polymers to establish the degree of chain extension. A polymer chain is extremely strong along the chain axis (like diamond) but weak in a perpendicular direction as a consequence of secondary forces binding it to other chains. Unoriented polymers are relatively weak because of the contribution of the low strength perpendicular direction. As a polymer is stretched, the chains become oriented parallel to the stretching direction and the strength in this direction increases (at the expense of a decrease in strength in the direction perpendicular to stretching). If one could produce fibers within which the chains approached almost complete extension, they would be extremely strong in the fiber direction, approaching the intrinsic strength of the single molecule. In practice, values of about 1/3 of this have been achieved.
Satisfactory methods for measuring degree of chain extension are not available. SANS on partly labelled material is a possibility. The value of $R_b$ parallel to the fiber axis will exceed the instrumental resolving power in most cases. However, the perpendicular $R_b$ will decrease with extension and approach values characteristic of the perpendicular dimension of single chains (a few angstroms). These are measurable (at high $q$). It is suggested that this application of SANS will be of major importance in characterizing these ultra-strong polymeric materials.

(8) With increased flux and greater capability for time resolved measurements, it seems likely that additional applications for following reaction mechanisms using labelled reactants will emerge. A typical example will be to follow the polymerization of segmented polyurethanes and/or nylon block copolymer which are gaining increasing importance in "RIM" polymerization.

(9) It seems likely that increased use of low molecular weight labelled probes added to polymeric systems will be made, analogous to the previously discussed use of swelling by a deuterated solvent to study network homogeneity. Examples would be their use for studies of microporosity, absorption and diffusion of low molecular weight liquids in polymers, swelling of amorphous regions in crystalline polymers, and swelling of polymer in the presence of filler. A far-out example might be the study of "free-volume" in glassy polymer by introducing a highly scattering small labelled probe under pressure (such as O$_2$ or NO$_2$ gas).

(10) If the SANS technique becomes sufficiently commonplace, it could be used to monitor experiments much as FTIR and NMR are used today. One might visualize a SANS detector on a gel permeation chromatograph (GPC), or a DSC built into a SANS cell so that scattering changes can be used to identify calorimetric changes. Specially prepared samples are sufficiently precious that the same samples might be examined by a number of techniques. For example, if oriented model networks with D-labelling are prepared, these might be examined by FTIR (comparing bands associated with H and D) and deuterium NMR as well as SANS.

(11) With the development of more intense and pulsed sources, there will undoubtedly be a proliferation of dynamic SANS experiments. I am least familiar with this area, so speculations will be withheld.

CONCLUSIONS

"Crystal ball gazing" is often dangerous. Techniques and problems not conceived today will most certainly evolve in the next decade. However it is evident from the above that many challenging problems exist which utilize current ideas and technologies likely to develop. Thus, we may rest assured that SANS will grow in importance and will become a major tool for polymer scientists in the foreseeable future.

REFERENCES

29. R. S. Stein and V. Soni, to be submitted for publication.
37. A. Hill, R. Lo, R. S. Porter and R. S. Stein, to be submitted for publication.
43. A. Keller, Phil. Mag., 2 , 1171 (1957).
46. L. Mandelkern, Ref.41, p.310.
47. S. Krimm and T. C. Cheam, Ref.41, p.244.
49. M. Stamm, E. W. Fischer and M. Dettenmaier, Ref.41, p.263.
52. C. M. Guttman, J. D. Hoffman and E. A. DiMarzio, Ref.41, p. 297.
54. J. Klein and R. C. Ball, Ref.41, p.198.


64. W. Graessley (Exxon), private communication, 1984.


Invited Paper

STRUCTURE OF MICELLAR SOLUTIONS AND MICROEMULSIONS*

J. P. COTTON
**Laboratoire Léon Brillouin, CEN-Saclay,
Gif/Yvette, France

Abstract

STRUCTURE OF MICELLAR SOLUTIONS AND MICROEMULSIONS.

When amphiphilic molecules (surfactant) are dispersed in water (or in water and oil), they form interfaces which separate aqueous from paraffinic (or oil) media. These interfaces form a large variety of colloidal structures depending on composition and temperature. Once the phase diagram obtained by thermodynamic methods, the problem lies in the determination of the structure and the interactions (organization) of these colloids. This is in principle a classical problem of radiation scattering but actually hard to solve because colloids are heterogeneous materials and their interaction is quite important. The difficulty lies in the separation of intra and intercolloid contributions to the intensity. (Moreover, both parts depend on the same parameters.) Small-angle neutron scattering with its large possibility of isotropic labelling is very useful in this field. A review of the applicable methods is given in connection with examples from the physics of micellar solutions and microemulsion (as a special case, the micellar inversion is discussed). A new method is also proposed.

*The full text of this paper was not available in time for inclusion in these Proceedings.

**Laboratoire mixte CEA - CNRS
POSTER PRESENTATIONS
PULSED AND STEADY-STATE SOURCES
(Session X.a)
Chairmen

T. SPRINGER
W. SCHMATZ

Federal Republic of Germany
DEVELOPMENT OF THE SPALLATION NEUTRON SOURCE (SNS) AT THE RUTHERFORD APPLETON LABORATORY

A. CARNE
Rutherford Appleton Laboratory, Chilton,
Didcot, Oxon,
United Kingdom

Abstract

DEVELOPMENT OF THE SPALLATION NEUTRON SOURCE (SNS) AT THE RUTHERFORD APPLETON LABORATORY.

When operating at full design specification the SNS will be the highest intensity pulsed neutron source in the world and will remain so well into the 1990s. The SNS is capable of development in many areas which will keep it in the forefront, not only as a pulsed neutron source but as a source of secondary particles of many types. Such developments are described, including accelerator, target and secondary beams.

1. INTRODUCTION

The Spallation Neutron Source (SNS) at the Rutherford Appleton Laboratory is scheduled to become operational at low intensity in April 1985. As this Conference has already heard (1,2), during 1984 a number of successful commissioning tests on the accelerator were performed, culminating in a short run in December 1984 when the whole facility was operated for the first time to produce neutrons for experimental purposes. In this run a low intensity 550 MeV proton beam was used on the full target assembly containing a depleted uranium target, a beryllium and heavy water reflector, 2 ambient water moderators and 2 cryogenic (CH₄ and H₂) moderators.

From the nominal 10% intensity at 550 MeV in early 1985, the SNS will be worked up to the full design specification during 1986 with 180-200 μA of protons at 800 MeV interacting with the depleted uranium 238 target to produce 6.5 x 10¹⁴ fast neutrons per pulse at the repetition rate of 50 Hz. The SNS will become the most powerful pulsed neutron source in the world and will remain so into the 1990's. Even when compared with the most intense continuous source, viz the ILL reactor at Grenoble, the SNS as presently conceived will be markedly superior in intrinsic brightness in the epithermal energy region, comparable at thermal energies and inferior only in the cold neutron energy region.
The SNS has potential for development in many areas which will keep it in the forefront well into the 1990's: not only as a pulsed neutron source but as a source of secondary particles of many types, some (eg muons) to complement it in the field of condensed matter science, others (eg neutrinos) for the study of fundamental physics problems. Such developments are described below and their advantages given. Developments of the SNS in its prime function as a pulsed neutron source are considered first.

2. DEVELOPMENT OF THE SNS

a) Accelerator

Clearly, increasing the proton beam current from the 50 Hz, 800 MeV synchrotron would increase the neutron source intensity. As a worthwhile goal consider an increase in intensity of a factor 3. The present design intensity of 2.3-2.5x10^{13} protons per pulse at 50 Hz (corresponding to 180-200 μA) is limited by transverse incoherent space charge effects, by emittance growth during acceleration and by acceptance limitations on extraction. The space charge limit can be reduced at injection by increasing the injection energy according as \( \beta^{2} \gamma^{3} \) where \( \beta, \gamma \) are the usual relativistic terms. Thus if this were the only limitation, an increase of the injection energy from the present 70 MeV to about 185 MeV is required. However during acceleration under heavy space charge the emittance decrease does not vary with momentum to the expected \( \frac{1}{4} \) on the present machine but is reduced by some factor. The limited evidence available elsewhere suggests an emittance 'blow-up' factor of 2 for the SNS, indicating a \( \varepsilon / \beta \gamma \) variation. Also the aperture available at extraction has already been determined for the initial parameters of the SNS and this will provide a further restriction as the emittance blow-up increases with current. Taking these 2 features into account, together with the reducing momentum range with injection energy, suggests the intensity can be increased according as \( \beta^{2} \gamma^{3} / \sqrt{\beta \gamma} \), ie \( \beta^{1.5} \gamma^{2.5} \). Hence to achieve a factor 3 increase in intensity requires an injection energy of approximately 230 MeV but perhaps as high as 300 MeV.

New equipment for a new injector at 230 MeV would include an induction linear accelerator at 600 μA or an RF linac. An induction linac for protons at 50 Hz has not been built but can offer a number of advantages. It can produce the required 7.5x10^{13} protons in a single bunch with the required emittances for the SNS. By modifying the synchrotron RF to harmonic number 1 (\( h = 1 \)), single shot,
loss-free injection is possible, giving a bunch length of 420 ns, reducing to 300 ns at 800 MeV. It would also be necessary to modify the injection system. The alternative RF linac would require modifications to the synchrotron magnet power supply and magnet coils, change of synchrotron RF to \( h = 1 \), and a new injection bump magnet and foil system. Though better known than the induction linac, it is less attractive because of its associated beam loss on injection. A third alternative, an FFAG synchrotron is also being considered. Such injection systems are complex and necessarily expensive, with rough indications of price being in the region £50–£60 million.

b) Target

The present SNS target consists of 34 kg of 0.2% depleted uranium, arranged in a stack of 23 parallel disks of varying thickness. Each disk is clad in zircaloy-2 and mounted in a stainless steel picture frame. Detailed descriptions of the target and its performance together with the moderators and reflector have been given (3,4). The intensity of the SNS can be enhanced by provision of a multiplying 'booster' target, containing fissile material (5). Such a target would be optimised for the cold neutron regime, to make it complementary to the best reactors, but concomitant increases in the higher energy regions would be an added bonus.

A collaborative project was set up in 1983 between RAL and Kernforschungsanlage, KFA, Jülich, to study multiplying sub-critical targets. The collaboration includes contributors from Birmingham University and Ispra. The project is considering topics such as scientific case, high power booster concepts, neutronic performance and moderator fluxes, technical problems (fuel and coolant), criticality and safety. The scope for such targets is two-fold:

i) To enrich the present SNS parallel plate target, with closely coupled moderators within a reflector assembly. This would serve the existing experimental facilities.

ii) An entirely new target station with a target consisting of a core (perhaps with fissile material) within a blanket of fissile material, surrounded by a fast reflector, with the moderators located at optimal positions within the assembly.

The design of the extracted proton beam and the layout of the SNS on the RAL site are such that both options are possible.
The beam could be split between the existing target station and the new one to allow optimisation of performance: the resultant reduced repetition rate for each target aids the time-of-flight technique (avoidance of frame overlap) and lowers the mean power ratings. The apportioning of beam is a major factor in the overall scheme.

Using the HETC/MORSE and DOT code packages studies have been made of uniformly enriching the existing SNS target, using a simplified benchmark model. Fig 1 summarises the dependence on percentage enrichment of various parameters, e.g. fission source, total source, net leakage, total power.
generation and ratio of total power to net leakage. Comparison is made always to the original depleted case. A 65% enrichment ($k_{\text{eff}} = 0.86$) produces an increase of net leakage (decoupler to reflector) of 4.6 for an increase of total power of 7.8, i.e., the ratio of power to net leakage is almost double that of the depleted case. A more detailed study of modifying the practical SNS target with 65% enrichment has been made. Moreover in the present SNS target assembly the neutron flux from the downstream 25K hydrogen moderator could be increased by enriching the back end of the target only. The features of such a system are still under study.

A booster target consisting of a core, with or without fissile material, surrounded by a fissile blanket ($^{238}\text{U}O_2 + ^{239}\text{Pu}O_2$), with a nickel reflector is also being studied. ANSN and DOT code calculations have been performed on a model with simple spherically symmetric geometry with a 20% volume occupation by sodium coolant. The calculations indicate that a core surrounded by a blanket all uniformly enriched is superior to a system with depleted core, producing a more compact assembly with a corresponding increase in neutron surface brightness. There is however little advantage in enhancing further the enrichment of the core. The alternative $^{235}\text{U}O_2$ has also been included in the calculations but the results show there is a substantial critical mass advantage of $^{239}\text{Pu}$ over $^{235}\text{U}$ in any compact system. To produce the same $k_{\text{eff}}$ at the same enrichment requires an all-uranium system of radius about 1.5 times the plutonium enriched assembly.

The goal of the order of 10x improvement is aimed at in these booster studies. Detailed work including engineering design and criticality/safety studies will determine what gains will actually be achievable. However developments in this area are likely to be considerably more cost-effective than in the accelerator.

3. THE SNS AS A SOURCE OF SECONDARY PARTICLES: THE $\mu$SR FACILITY

Installation of an intermediate transmission target in the extracted proton beam upstream of the main SNS target allows the provision of secondary beams, in particular to develop a world class $\mu$SR facility. The pulsed nature of the SNS synchrotron affords an enormous advantage both as a neutron source and as a muon source (compared with the high intensity continuous current meson factories at LAMPF, SIN and TRIUMF). In the case of $\mu$SR studies with positive $\mu$-muons produced from macroscopically DC sources, the useful $\mu^+$
stopping rates that can be tolerated are limited by the long muon lifetime in the sample \((\tau = 2.2 \mu s)\). However a pulsed muon source can utilise all of the incoming muons by implanting them in a sample in short bursts (10-100 ns) which are separated in time long compared with the muon lifetime (on the SNS this time is 20 ms). Increases of more than a factor 100 in the useful \(\mu^+\) stopping rate in samples are possible in the SNS pulsed muon source. Most experiments will benefit simply from these increased stopping rates, but the use of pulsed muons offers further advantages together with the possibility of developing new \(\mu\)SR techniques:

a) A very clean \(\mu\)SR time spectrum, which will allow \(\mu\)SR measurements up to at least 10 radioactive half-lives. This will facilitate development of techniques for the measurement of slow relaxation functions particularly the long time behaviour of the longitudinal or zero field relaxation functions, for diffusion and spin relaxation studies.

b) Possibility of pulsed stimulation of the sample. The low duty cycle of the SNS allows the efficient use of pulsed experimental conditions, eg magnetic fields, RF fields etc, and has applications in studies of hyperfine fields in magnetism and free radical chemistry.

c) Time domain spectroscopy. By varying the time of the pulsed stimulus to the sample relative to the muon burst, the response function can be explored. Such a technique extends the \(\mu\)SR timescale and will be used in the study of magnetic relaxation functions - especially in spin glass systems - using pulsed magnetic fields.

Funds have been provided by the EEC, Germany\(^1\) and France, whilst the UK has provided beam line components, sufficient to build the surface muon beam shown in Fig 2. As can be seen in the figure the source (a 20 mm thick graphite target) is located 20 m upstream of the main SNS target. This target will interact with some 5% of the incoming proton beam to produce high intensity, fully polarised surface muons at 28 MeV/c for \(\mu\)SR science. At full SNS intensity, the 28 MeV/c surface \(\mu\) intensity is \(1.6 \times 10^6 \mu^+/s\). Positron contamination of the muon beam will be effectively eliminated by the use of a cross-field electrostatic separator shown in the figure. By increasing the field in this separator the polarisation of the muon can be

\(^1\) Federal Republic of Germany.
rotated from 100% longitudinal to 100% transverse. The time structure of the beam arriving at the experiment position will be largely determined by the time structure of the protons at the intermediate target, i.e., double 100 ns pulses separated by 230 ns repeated at 50 Hz. An early development of the beamline will include a fast kicker magnet which will produce separation of the 100 ns pulses to separate experimental areas and, by shaping the kicker waveform, reducing their time widths to give improved time resolution (6). The beamline can also be used as a cloud muon channel with unpolarised muons $\mu^+$ of momentum up to 70 MeV/c; it can also be used as a higher momentum channel with pulsed pions $\pi^-$ of momentum up to 200 MeV/c.

Beyond this muon facility a second beamline could be built from the intermediate target on the opposite side of the extracted proton beam. There are two possibilities, both of which are being studied at RAL:

i) To build a superconducting decay channel for $\mu^+$ of momentum in the range 80-150 MeV/c and polarisation about 80% (this beamline would be similar to that at SIN).
ii) To build a high intensity $\pi^-$ beam line for cancer therapy studies, similar to the SIN Piotron line.

It is particularly appropriate that the $\mu$SR facility be built and developed at the SNS. The additional advantage of having a source dedicated to $\mu$SR in a laboratory oriented towards the study of a wide range of problems in Physics, Chemistry, Biology and Materials Science, is of great importance. Since $\mu$SR ideally complements the neutron scattering technique it is fortunate that the world's most powerful pulsed neutron and pulsed muon sources can be located side by side.

Acknowledgement

A number of colleagues have contributed towards the work on development of the SNS. It is a pleasure to acknowledge their efforts, particularly Grahame Rees, Tim Broome, Gordon Eaton, David Picton and Derek Beynon of Birmingham University and Winfreid Scherer of KFA.

REFERENCES

(1) D A GRAY, "The Spallation Neutron Source at the Rutherford Appleton Laboratory". This Conference IAEA-CN-46/014.
(2) A J LEADBETTER, "Instrumentation for Pulsed Sources". This Conference IAEA-CN-46/041.
THE SPALLATION NEUTRON SOURCE SNQ


*KFA Institut für Festkörperforschung
**KFA-SNQ
***KFA-Zentrallabor für Elektronik
++KFA Anlagenplanung

Kernforschungsanlage Jülich GmbH, Jülich, Federal Republic of Germany

Abstract

THE SPALLATION NEUTRON SOURCE SNQ.

The spallation neutron source, planned in the Federal Republic of Germany, is conceived primarily for experiments with thermal and subthermal neutrons. It is to be optimal not only for time-of-flight spectroscopy but also for "multiplexing" measurements of steady state character. For this reason the source is pulsed with rather large pulse lengths. The spallation will be induced by protons accelerated to 1100 MeV with a time average current of 5 mA, in pulses of 100 Hz repetition rate and 250 μs duration. For this acceleration a linac is foreseen with two H⁺ sources and two radiofrequency quadrupoles in parallel, the two beams merging in a funneling section for injection into an Alvarez and from there into a single cell structure. The spallation material will be first W and later 238U, mounted on a wheel which rotates through the proton beam. Neutron beams will be extracted through 10 twin beamholes and 12 neutron guides from two moderators, a thermal and a cold one. Further facilities will be provided for nuclear and particle physics and for irradiation purposes.

The planned spallation neutron source in the Federal Republic of Germany, SNQ (Spallations-Neutronen-Quelle), is designed primarily for experiments with beams of thermal and subthermal neutrons. The design goal is to surpass existing high flux reactors, not only for epithermal but also with regard to thermal and to cold neutron fluxes. This will be achieved with a time-average thermal flux

\[ \bar{\phi}_{th} = 1.2 \times 10^{14} \text{ n cm}^{-2} \text{s}^{-1}, \]

equal to the one of the Institut Laue Langevin (ILL) reactor in Grenoble, and a peak flux

\[ \phi_{th} = 4.5 \times 10^{16} \text{ n cm}^{-2} \text{s}^{-1}, \]
in pulses of 100 Hz repetition rate and 260 μs duration. For cold neutrons the fluxes will be

\[ \phi_{c} = 8 \times 10^{14} \text{ n cm}^{-2} \text{s}^{-1}, \]
Table I. COMPARISON OF $\Phi_{th}$ VALUES

<table>
<thead>
<tr>
<th>Neutron Source</th>
<th>HFR (ILL)</th>
<th>SNS (RAL)</th>
<th>SNQ(KFA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time average thermal flux $\Phi$ ($10^{13}$ cm$^{-2}$s$^{-1}$)</td>
<td>100</td>
<td>0.7</td>
<td>120</td>
</tr>
<tr>
<td>Peak thermal flux $\Phi$ ($10^{13}$ cm$^{-2}$s$^{-1}$)</td>
<td>100</td>
<td>450</td>
<td>4 500$^a$</td>
</tr>
<tr>
<td>Pulse repetition rate $\nu$ (s$^{-1}$)</td>
<td>---</td>
<td>50</td>
<td>100</td>
</tr>
<tr>
<td>Effective peak thermal flux $\phi_v$ ($10^{17}$ cm$^{-2}$s$^{-2}$)</td>
<td>1$-2^b$</td>
<td>2.2</td>
<td>45$^a$</td>
</tr>
</tbody>
</table>

$^a$ Not including a pulse compressor
$^b$ Assuming a chopper frequency of 100 — 200 s$^{-1}$

again somewhat comparable to the ILL, and

$$\Phi_c = 2.2 \times 10^{16} \text{ n cm}^{-2} \text{ s}^{-1},$$

with 390 $\mu$s pulse duration. These values were determined in mock-up experiments at the SIN and at the SATURNE proton accelerators. In Table I the values for $\Phi_{th}$ are compared to those of the Grenoble reactor and to those expected for the spallation source, SNS, under construction at the Rutherford Appleton Laboratory in the United Kingdom. The time structure of the SNQ - 100 Hz repetition rate and 260 or 390 $\mu$s pulse duration, respectively - was chosen as an optimal compromise between different demands.

It has been shown by intensive elaborations$^1$ that with the above values, not only time-of-flight spectroscopy but nearly all kinds of neutron scattering measurements gain considerably in intensity compared to the ILL reactor. A summary of these elaborations is presented in Table II. It shows that, for most types of measurement, the gain is larger than a factor of 10. For the non-time-of-flight types of experiment, this is achieved mostly by redesigning conventional instruments in such a way that they profit from the source time structure by accepting a band of incident wavelengths much broader than the final resolution. For the instruments gaining little (small-angle scattering with poor resolution, spin-echo spectrometer) or nothing (triple axis spectrometer), there are yet considerable advantages, for instance reduction of background.

Table II. SUMMARY ON AN INTENSITY COMPARISON FOR INSTRUMENTS AT SNO AND HFR.

The gain G refers to SNO, Linac and U target but no compressor.

<table>
<thead>
<tr>
<th>Type of instrument</th>
<th>Gain</th>
</tr>
</thead>
<tbody>
<tr>
<td>TOF powder diffractometer (200 m) res. $2 \times 10^{-14}$</td>
<td>35</td>
</tr>
<tr>
<td>TOF single-crystal diffractometer $\text{LiSO}_4$ cell = 6 Å $\text{protein cell} = 50$ Å</td>
<td>10-3</td>
</tr>
<tr>
<td>Small-angle diffraction $\text{protein cell} = 300$ Å</td>
<td>3</td>
</tr>
<tr>
<td>Small-angle scattering $\text{beam } \phi 10$ mm res: 10% $\text{10 m } 1%$ $\text{1 mm } 10%$</td>
<td>2</td>
</tr>
<tr>
<td>Diffuse scattering</td>
<td>7</td>
</tr>
<tr>
<td>TAS triple axis spectrometer</td>
<td>1</td>
</tr>
<tr>
<td>TOF inelastic thermal neutrons $v_{\text{exp}} = 250$ Hz $\text{cold neutrons} v_{\text{exp}} = 100$ Hz</td>
<td>14</td>
</tr>
<tr>
<td>MAX inverted TOF with multi-arm analyser</td>
<td>17</td>
</tr>
<tr>
<td>IRIS$^a$ - inverted TOF with backscattering analyser $\text{res. 10-50 } \mu$eV</td>
<td>12-15</td>
</tr>
<tr>
<td>MUSIC multi-crystal backscattering $\text{res. 0.1-1 } \mu$eV</td>
<td>12</td>
</tr>
<tr>
<td>Spin echo $\text{res. 20 }\text{neV}$</td>
<td>3-6</td>
</tr>
</tbody>
</table>

$^a$ designed (for higher resolution) and under construction at SNS.

separation of different orders of reflections, enhancement of the
dynamic time scale, etc.

These gains for scattering experiments with thermal and subthermal neutrons are achieved by a high time-average power of the primary proton beam and by keeping the neutron moderators unpoisoned and coupled to the source, the spallation target, as well as to reflectors. This has to be paid for by the pulses being too long for some of the measurements. Those measurements then require pulse shaping, sometimes by choppers close to the moderators. The provision of short primary pulses, of around 200 ns, is left open as an option.

The spallation process is induced by protons, accelerated in a linac of the following characteristics:
The most prominent feature by which this accelerator must surpass other machines is its high current in pulses of high repetition rate. Yet, the design shown in Fig. 1 is based on known and tested components only. Two multi-cusp \( H^+ \) sources with an extraction voltage of 50 keV, operating in parallel, inject into two 100 MHz radiofrequency quadrupoles for acceleration to 2 MeV and simultaneous focusing and bunching of the beams. In a funnelling section, the two 100 mA beams then are merged together as in a zipper so that the resulting one 200 mA beam has a 200 MHz microstructure. It is injected into a more or less standard 200 MHz Alvarez accelerator for acceleration to 100 MeV. Acceleration from 100 to 1100 MeV will be achieved through 640 identical cavities, with the phases of the accelerating voltage controlled electro­nically. This single cell structure was chosen for the high-energy part because it offers the following advantages:

- There is no energy flow along the accelerating structure. This is important in view of the fact that the energy extracted from the cavities by the beam is so high that it has to be supplied from the generators during the pulse, which is difficult with energy flow along the structure.

- There are a large number of rf amplifiers, but each of comparatively low power. If one or a few of them fail, the phase in the following cavities can be readjusted with practically no beam interruption. This guarantees a high availability of the machine.

- The end energy can be varied and, within certain limits, balanced with the current. This is of threefold interest:
(i) Variable energy is important for some nuclear physics experiments.

(ii) Bunch manipulations, desirable for meeting the special requirements of some experiments, are feasible in various ways by using some of the accelerator length as drift space. In this way, bunches of very narrow energy spread or particularly high intensity can be produced for special purposes.

(iii) During the commissioning period, and perhaps even later on, it will be necessary to study carefully all sources of beam losses to keep the activation of the facility as low as possible. This is considerably facilitated with the possibility of varying the energy at any point of the accelerator. Similar considerations hold for the optimization of operating parameters with respect to source exploitation and cost.

The energy variation is possible as a long-term option as well as from pulse to pulse or even between the main pulse and a satellite provided for deflection to the nuclear physics facilities. Multiple-energy operation will, therefore, not impede the neutron production.

The main feature of the spallation target, shown in Fig. 2, is its design for removing the heat produced and for keeping the operating temperature low. For this purpose, the target material is contained in 5916 cylindrical capsules, mounted on a wheel of 2.5 m diameter which rotates horizontally through the proton beam with a frequency of 0.5 Hz so that two consecutive proton pulses hit neighbouring surface elements without overlap. The material is cooled by water entering and leaving the interior of the wheel through its axis. The neutrons, produced in the target elements, will stream upwards and downwards into moderators for slowing down to thermal and subthermal energies - a light water moderator for thermal and a cold moderator (liquid deuterium) for subthermal energies. Figure 3 shows the shape of the neutron pulses leaving the moderators. For neutron scattering experiments, 10 beam holes with 20 neutron beams and 12 neutron guides will be pointing on to these moderators in a way very similar to arrangements around the cores of research reactors. Two further beam holes are foreseen for fundamental studies on the neutron and its interactions and, eventually, for nuclear physics. The target block will also contain some facilities for irradiating samples, for instance for isotope production. Figure 4 shows the area for neutron scattering experiments. Space will be provided for about 40 or more instruments on two floors in the hall around the target shielding and in an adjacent hall to which subthermal and cold neutrons are transported through 12 guide tubes of different curvatures.

The aforementioned option for shorter pulses can be realized by inserting a proton compressor ring between the linac and the spallation target and by introducing another moderator, decoupled from the source and the reflector and with neutron absorbing material inside. For the compressor, an isochronous ring has been conceived, able to compress the linac pulse of 250 μs duration into a pulse of 700 ns or into three pulses of 200 ns, separated from each other by intervals of 10 μs. Such short pulses will be required
FIG. 2. The SNO spallation target. The target material (lead, tungsten or depleted uranium) is mounted on a wheel of 2.5 m diameter rotating through the proton beam with a frequency of 0.5 Hz. The target elements are cooled by water.

FIG. 3. Pulse form d(t) from the thermal H₂O moderator and the cold D₂ moderator in C reflector. Flux enhancement \( \delta_{\text{th}}/\delta_{\text{th}} \) as averaged over a pulse length t cut out from the moderator pulse.
for work with epithermal neutrons as well as for some nuclear physics and neutrino experiments. Provisions will be made for further targets to be installed at a later stage: a pion production target and a thin isotope-recoil target in the mainproton beam and a further spallation target for isotope production directly connected to a mass separator in a small part of the beam branched off from the main beam. Moreover, another small part of the proton beam will be branched off for nuclear structure research with energies between 350 and 1100 MeV, part of which can also be injected into a cooler synchrotron at present being designed for use with an already existing isochronous cyclotron. As to neutrino physics, a special target is studied, dedicated to the production of "beam dump" neutrinos.
ASSEMBLY OF NEUTRON SOURCES BASED ON PROTON BEAMS OF THE MOSCOW MESON FACILITY.

The authors describe an assembly of neutron sources for physical research based on proton beams from the Moscow meson facility. The expected neutron intensity at the $4\pi$ solid angle in the regime of the resonance selector is $\sim 3 \times 10^{16}$ n/s (25 ns, 400 Hz), while the peak density of the thermal neutron flux at the eliminating surface of the moderator is $\sim 5 \times 10^{15}$ n-cm$^{-2}$-s$^{-1}$ and the pulse duration $\sim 50$ $\mu$s.

Комплекс источников нейтронов на основе протонных пучков Московской мезонной фабрики.

Дается описание комплекса источников нейтронов для физических исследований на основе протонных пучков Московской мезонной фабрики. Ожидаемая интенсивность нейтронов в телесном угле $4\pi$ в режиме резонансного селектора $\sim 3 \times 10^{16}$ нейтр./с (25 ns, 400 Гц), пиковая плотность потока тепловых нейтронов на светящейся поверхности замедлителя $\sim 5 \times 10^{15}$ нейтр./см$^2$ при длительности импульса $\sim 50$ мкс.

Сооружаемая в ИЯИ АН СССР, в Троицком научном центре, мезонная фабрика — сильноточный ускоритель протонов и ионов $Н^+$ на энергию 0,6 ГэВ с током 0,5-1,0 мА (рис. 1). Интенсивные пучки протонов, в том числе поляризованных, пучки $π^-$, $μ$-мезонов и нейтрино, получаемые при взаимодействии протонов с веществом мезонных мишеней и ловушек протонных пучков, позволяют разывать широкий круг исследований — от фундаментальных проблем строения материи [1] до прикладного применения, как, например, изучение радиационного поведения материалов для атомной энергетики и установок термоядерного синтеза [2].

Протоны средних энергий позволяют получать и интенсивные потоки нейтронов. Действительно, при взаимодействии протона с энергией 0,6 ГэВ с веществом пруткеной мишени из тяжелых ядер (вольфрам, свинец) испускается 10-12 нейтронов, из них более 90% — испарительные, со средней энергией 2-3 МэВ. В случае мишени из урана-238 число вторичных нейтронов — 25, примерно половина — нейтроны деления. При доступных ныне интенсивностях протонных пучков от сильноточных ускорителей протонов — ме-
зонных фабрик, средние интенсивности нейтронов пока еще ниже, чем достигнутые в совре-ременных исследовательских реакторах, однако возможность изменения в широких пределах временной структуры протонного пучка и, соответственно, временной структуры потока нейтронов открывает уникальные экспериментальные возможности [3].

Формирование коротких импульсов протонов средних энергий опирается на использование перезарядной инъекции ионов H⁺ в накопители. Перезарядная инъекция, предложенная в свое время Альварецом [4] и осуществленная впервые Г.И. Будкером с сотрудниками [5], открывает пути накопления в кольцевых магнитных системах больших циркулирующих токов протонов. Быстрый вывод протонов из накопителя Московской мезонной фабрики (рис.1) на внешнюю мишень позволит генерировать импульсы нейтронов длительностью 5-200 нс со средней интенсивностью нейтронов в телесном угле $4\pi \sim 6 \cdot 10^{16}$ нейтр./с.

На основе сгруппированного пучка протонов могут быть получены импульсные потоки медленных нейтронов для исследований по физике конденсированных сред (изучение упругого и неупругого рассеяния холодных и тепловых нейтронов в конденсированных средах), уникальные по интенсивности импульсные потоки резонансных нейтронов. Спектрометры резонансных нейтронов на основе импульсного источника Московской мезонной фабрики позволяют осуществить широкий круг исследований парциальных процессов взаимодействия нейтронов с ядрами в изолированных резонансах и в области перекрывающихся уровней.
Рис. 2. Комплекс источников нейтронов: 1 — протонные пучки; 2 — квазистационарный источник тепловых и холодных нейтронов; 3 — импульсный источник нейтронов.
Импульсный характер потока первичных нейтронов при малом выходе нейтронов между импульсами (в случае урани-238 этот выход — 1%, свинца — 0,1%, а для мишени среднего веса, например из молибдена < 10^{-5} % [3]) позволит, применяя временную селекцию событий, получить в хороших замедлителях (D₂O, Be, C) квазистационарные потоки тепловых и холодных нейтронов с малым вкладом натепловых и быстрых ("временная тепловая колонна"). Это открывает возможность постановки таких экспериментов, как прямое изучение рассеяния нейтрона на нейтроне, поиск нейтрон—антинейтронных осцилляций [7]..

Комплекс источников нейтронов создается на основе двух пучков протонов (рис. 2), которые могут быть использованы одновременно. Первый пучок — пучок протонов из ускорителя со средним током до 1 мА, имеющий временную структуру в виде последовательности макроимпульсов длительностью ~ 100 мкс, следующих с частотой повторения 100 Гц. Этот пучок попадает в нейтронную мишень квазистационарного источника тепловых и холодных нейтронов (рис. 2, 3, 4). Второй пучок, сгруппированный в накопителе с перезарядной инжекцией и однооборотным выводом [3, 6] в виде последовательности импульсов длительностью 5-200 нс (100-400 Гц) при среднем токе до 500 мкА, попадает в нейтронную мишень импульсного источника нейтронов (рис. 2, 3, 5).

В импульсном источнике нейтронов наряду с импульсами протонов, сгруппированными в накопителе, может использоваться микроструктура протонного пучка, формиру-
Рис. 4. Квазистационарный источник тепловых и холодных нейтронов: 1 — нейтронная мишень; 2 — тяжеловодный замедлитель; 3 — жидкодеутерийный замедлитель; 4 — канал п—п-эксперимента.

ющаяся в линейном ускорителе. Импульсы микроструктуры имеют длительность 0,2 нс и следуют с частотой 200 МГц в пределах импульса макроструктуры. Для времени—пролетных исследований они могут быть использованы после селекции, при частоте повторения до 200 кГц, что, естественно, приводит к соответствующей потере интенсивности.

Оба источника располагаются в цилиндрических боксах радиационной защиты из стали и бетона (рис.3) и “просматриваются” системой каналов, снабженных шиберами гильотинного типа.
Квазистационарный источник тепловых и холодных нейтронов (рис. 4) включает водоохлаждаемую нейтронную мишень в виде плотной упаковки стержней из естественного урана в алюминиевых оболочках, тяжеловодный замедлитель в алюминиевом баке, жидкодейтериевый замедлитель. Жидкодейтериевый замедлитель "светит" в широкопертурный канал эксперимента по обнаружению осцилляций нейтрон—антинейтрон, каналы для исследований с холодными и ультрахолодными нейтронами. Сквозной тангенциальный канал позволит проводить исследования в интенсивных полях тепловых нейтронов в тяжеловодном замедлителе.
При токе протонов \( \sim 0,5 \text{ мА} \) расчетное значение плотности потока тепловых нейтронов на дне экспериментального канала \( \sim 3 \cdot 10^{13} \text{ нейтр./}(с \cdot \text{см}^2) \), плотность потока холодных нейтронов на светящей поверхности холодного замедлителя \( \sim 10^{13} \text{ нейтр./}(с \cdot \text{см}^2) \) при эффективной температуре \( \sim 30 \text{ К} \).

При этом полное тепловыделение в нейтронной мишени \( \sim 1 \text{ МВт} \), в тяжелой воде \( \sim 40 \text{ кВт} \), в холодном замедлителе \( \sim 0,5 \text{ кВт} \).

Тепло, выделяемое в мишени, отводится водой с входной температурой \( \sim 30^\circ \text{С} \), передающей тепло воде второго контура.

Тепло, выделившееся в тяжелой воде, отводится в дополнительном теплообменнике. Жидкодейтериевый замедлитель охлаждается за счет конденсации паров дейтерия в теплообменнике, обдуваемом гелием с входной температурой 15 К от холодильной установки, работающей в рефрижераторном режиме.

Импульсный источник нейтронов [8] включает мишень — сборку из урановых (вольфрамовых) стержней в алюминиевых оболочках, охлаждаемую водой, и систему замедлителей (рис.5). Легководные замедлители примыкают к верхней и нижней поверхностям мишени. Тонкий (~2 см) верхний замедлитель "светит" в каналы и нейтроноводы резонансных и быстрых нейтронов с пролетными базами до 500 м, выходящие из зала экспериментального комплекса (рис.1). Нейтроноводы снабжены промежуточными павильонами. Нижний замедлитель, толщиной около 6 см, предполагается использовать в экспериментах с тепловыми и холодными нейтронами. Соответствующие каналы выходят в экспериментальный зал.

Предусмотрена возможность использования холодного жидкокварцодородного замедлителя, легководных замедлителей, отравленных кадмием, отражателей замедлителей. Мишень и система замедлителей импульсного источника нейтронов располагаются в вакуумном баке в полости защиты (рис.3).

Средняя интенсивность нейтронов, испускаемых в телесном угле 4\( \pi \) в режиме резонансного селектора (25 нс, 400 Гц) \( \sim 3 \cdot 10^{16} \text{ нейтр.}/с \) при длительности импульса \( \sim 50 \text{ мкс} \). Использование отражателя из железа позволит получить пиковые потоки плотностью до \( \sim 5 \cdot 10^{15} \text{ нейтр.}/(с \cdot \text{см}^2) \) на светящей поверхности \( \sim 100 \text{ см}^2 \).

Средняя интенсивность быстрых нейтронов из мишени (испарительных и каскадных) в режиме быстрого селектора при длительности импульсов \( \sim 0,2 \text{ нс} \) и частоте повторения 200 кГц \( \sim 3 \cdot 10^{13} \text{ нейтр.}/с \) в телесном угле 4\( \pi \).

**Литература**


UPGRADING THERMAL NEUTRON BEAMS OF LIGHT-WATER-MODERATED RESEARCH REACTORS BY REFLECTOR OPTIMIZATION

E. J. BLEEKER
Netherlands Energy Research Foundation ECN, Petten, Netherlands

U. DAHLBORG
Institute for Reactor Physics, Royal Institute of Technology, Stockholm, Sweden

B. NILSSON, J. SKOLD
Studsvik Energiteknik AB, Studsvik, Sweden

W. B. YELON
University of Missouri Research Reactor, Columbia, Missouri, United States of America

Abstract

UPGRADING THERMAL NEUTRON BEAMS OF LIGHT-WATER-MODERATED RESEARCH REACTORS BY REFLECTOR OPTIMIZATION.

A series of reflector and beam tube configurations has been studied at the R2-0 research reactor at Studsvik with respect to the relation between the flux density in the reflector and the neutron current in the beam tube and to the ratio of thermal to fast neutrons. H2O, D2O and Be were chosen as reflector materials with due attention to the actual and possible future configuration of HFR, Petten and R-2, Studsvik, where reactor vessel replacement has just been completed or is in progress. The experiments were designed to reveal the dependence of the flux density on the depth and width of the D2O reflector, the amount of D2O between the core and the beam tube, and a Be layer inside or outside the core-box. Flux density levels in the reactor core, in the H2O and D2O reflector, and in the beam tube have been monitored by activation of Co wires and stacks of different metal foils and gold foils.

1. INTRODUCTION

Dedicated reactors for neutron beam research use D2O reflectors and tangential beam tubes, but an entire generation of multipurpose reactors are equipped with Be, H2O or other reflectors and radial beam tubes. Since a number of these reactors are in the process of, or approaching, upgrade (e.g. HFR-Petten, R2-Studsvik, MURR-Missouri) it is appropriate to examine possible modifications to beam tube geometry and reflector configuration in order to achieve better performance.
for neutron beam research (especially neutron scattering) while maintaining the radial geometry. In addition, computer codes for calculating reactor performance have been improved in recent years and these codes may now be capable of describing beam tube performance as well as general reactor conditions. It is thus useful to provide data in order to critically test these codes.

We have performed a series of measurements to obtain such data, in which neutron fluxes and spectra and beam tube currents are measured for a variety of reflector materials and for a number of beam tube configurations.

2. EXPERIMENTAL

The experiments were performed at the R2-0 reactor at Studsvik, an MTR type reactor which is uncontained and convection cooled, allowing easy access of variable reflector and beam tube configurations. The mock-up arrangement is shown in Fig. 1. The overall dimensions of the box, entirely made of

---

1 Materials Testing Reactor.
aluminium, are 50 x 60 x 60 cm and it is divided into eight compartments. The compartments can be individually filled with He gas, H$_2$O or D$_2$O. Thus, by filling and emptying compartments, it is possible to simulate different beam tube and moderator configurations. As it is often advantageous in neutron experiments to have high and narrow beams, the cross-sectional area of compartments 1 through 5 was 8 x 34 cm$^2$.

Four different irradiation facilities were available, two wet wells and two dry wells. One of the dry wells is positioned at the end of a "beam tube" at a distance of 190 cm from the front wall of the box. Activities obtained in this well measure neutron current while the other three irradiation facilities measure neutron fluxes. In order to ensure that compartment walls are not seen from the dry well D, a collimator of polyethylene and cadmium was placed in the "beam tube" (Fig. 1). Different reflectors could be inserted between the mock-up volume and the reactor core. The three combinations which are relevant for past and future situations at the reactors in Petten and Studsvik were used in these investigation, namely 5 cm Be + 4 cm Al, 4 cm Al + 5 cm Be and 4 cm Al.

The nominal reactor power during the irradiations was 20 kW. For normalization purposes, six Co-wires were inserted into the reactor core for each experiment in positions indicated in Fig. 1. The average thermal flux as determined from the activity of the six Co-wires was used for relative normalization of the measurements.

3. RESULTS

The flux densities were determined by foil activation at various locations inside the reflector tank and the beam tube for 26 different reflector configurations. In order to reach criticality for each configuration, six different cores were needed. In total about 380 foils have been activated and measured by the method of gamma counting and, in some cases, by beta-gamma coincidence counting. Measurements of flux within the reflector volume (wet wells 1 and 3) are referred to as "unperturbed", $\Phi$, while fluxes and currents within the beam tube are denoted by $\Phi$.

In addition to the s$\bar{b}acial$ flux distribution, we also determined the neutron spectral distribution in the thermal ($\Phi$), intermediate ($\Theta$) and fast ($\Phi_f$) regions (where $\Theta$ is flux per unit lethargy). As flux density detectors Sc-45, Fe-58, Au-197, U-235 and Ni-58 were chosen. The first four measure the thermal/intermediate fluxes whereas the fast flux is derived from the activation of Ni-58.

---

3.1. Unperturbed fluxes

For measurements inside the reflector volume, Au foils 5 cm apart, and foil stacks, 10 cm apart, were inserted in wet wells 1 or 3 on flexible Al strips (bayonets) and activated for 2-3 hours. Uncertainties in \( \Phi \) range from 7\% to 16\%. Because of the lower intermediate flux, uncertainties in \( \Theta \) are between 40\% and 250\% in \( \mathrm{D}_2\mathrm{O} \) and between 11\% and 800\% in \( \mathrm{H}_2\mathrm{O} \). Three basic configurations were considered, reflector 1 or 2 (Be-Al or Al-Be) with \( \mathrm{D}_2\mathrm{O} \) in the tank, reflector 1 or 2 with \( \mathrm{H}_2\mathrm{O} \) in the tank or reflector 3 (Al) with \( \mathrm{D}_2\mathrm{O} \) in the tank. The filling of compartments 1-4 and 7 were also studied, but they had only small effect on \( \Phi \) and will not be discussed here. In Fig. 2 the results of the thermal flux densities for the Be-Al-\( \mathrm{D}_2\mathrm{O} \), Al-\( \mathrm{D}_2\mathrm{O} \), Be-Al-\( \mathrm{H}_2\mathrm{O} \) and Al-Be-\( \mathrm{H}_2\mathrm{O} \) cases are compared. In \( \mathrm{H}_2\mathrm{O} \), \( \Phi \) drops by one order of magnitude in the first 15 cm, in \( \mathrm{D}_2\mathrm{O} \) it is reduced by 30\% only. The Al-\( \mathrm{D}_2\mathrm{O} \) configuration seems to be favored over the Be-Al-\( \mathrm{D}_2\mathrm{O} \) case by about 60\%. Only a small part of this gain can be explained by the fact that the tank is 6 cm nearer to the reactor core. In Fig. 3 the three components of the neutron spectrum are presented for the Al-\( \mathrm{D}_2\mathrm{O} \) and the Be-Al-\( \mathrm{H}_2\mathrm{O} \) cases. It can be seen that, within the limits of accuracy, the distribution of \( \Phi \) and \( \Theta \) are similar for the two cases, although the \( \mathrm{D}_2\mathrm{O} \) case is favored by a slightly higher \( \Phi \)/\( \Theta \) ratio. On the other hand, the fast flux density is relatively higher in \( \mathrm{H}_2\mathrm{O} \).

3.2 Beam Tube Fluxes (Dry Well B)

The dry well in the reentrant part of the beam tube, enables us to investigate the effect of a central void in the reflector. Activations were undertaken in well B for three representative configurations. A foil of Ag-109 was added to the spectrum set of foils in these cases. The values of \( \Phi \) are plotted at the appropriate position in Fig. 2. It appears that the thermal flux in the void surrounded by \( \mathrm{D}_2\mathrm{O} \) follows the unperturbed flux at the same depth in the reflector, whereas for a void in \( \mathrm{H}_2\mathrm{O} \), \( \Phi \) is...
FIG. 3. Unperturbed flux densities in the reflector tank.

FIG. 4. Current densities in the beam tube; well D.

nearly a factor 5 higher than inside the reflector. Also, the thermal flux density for D₂O is higher by a factor of 10 compared to Be-H₂O although at the front of the tank the ratio \( \Phi(D₂O)/\Phi(Be-H₂O) \) is less than 2. In fact, the ratio \( \Phi/\Phi_p \) with \( \Phi \) taken at the depth where the beam tube begins, and \( \Phi_p \) at well B, is 1.6 for D₂O and 17 for Be-H₂O.

3.3 Beam Tube Currents (Well D)

The measurements in well D are the most essential part of the experiments, since they give the thermal neutron current density in the beam tube. Only gold foils were activated in this position. Some of the foils were covered on both
sides by 0.5 mm of cadmium to distinguish between thermal and resonance activation. The current density for the Al-D$_2$O case of Fig. 4 shows shallow flux peaking at a D$_2$O thickness of $\sim$ 15 cm. The slopes of $\phi$ and $\theta$ turn out to be quite comparable to that in the reflector (Fig. 3). In Fig. 5, two other cases are compared with that of Fig. 4: in the first case, beryllium has been added to the heavy water reflector (the Al-Be-D$_2$O case) and in the second case, compartments 2, 3 and 4 are successively filled with H$_2$O (with D$_2$O in the preceding compartments) in order to investigate the effects of a hydrogenous medium in a heavy-water environment; i.e. to simulate a H$_2$ cold source. As measurements with Cd were not done for these configurations, only total saturation activities are compared in Fig. 5. As can be seen, what applies inside the reflector also applies at the end of the beam tube: the Al-D$_2$O values are higher than the Al-Be-D$_2$O values. Furthermore, the loss in current density by the introduction of 5 cm of water in a region with a weak flux gradient does not seem to be prohibitive. Comparison of the D$_2$O and the Be-H$_2$O cases reveals that the current density at the end of the beam tube with D$_2$O is 6 times higher than with Be-H$_2$O. This surprisingly large gain factor leaves much for speculation, but is consistent with the gain of 10 at the nose of the beam tube.
4. DISCUSSION

Although the results reported here represent only a fraction of the data collected, important trends are already apparent. As expected, the radial decrease of unperturbed flux is much slower in D2O than in H2O. Consequently, the nose of a beam tube may be positioned further from the reactor core in D2O, in a region of more favorable thermal to fast flux ratio. The radial dependence of the perturbed beam tube flux in D2O is only slightly different from the unperturbed reflector flux, which is due to the large neutron mean free path in D2O.

In comparison, the beam tube flux $\Phi$ is strongly depressed compared to the unperturbed flux in the Be-H2O case. This is thought to arise from two effects: first, the void (beam tube) strongly perturbs the reflector, and the power is depressed in the fuel adjacent to the void. Unfortunately, the core fluxes in positions E1 and F1 were not measured. However, the Co wire activations in positions D1, G1 and H1 support this interpretation. Secondly, the H2O reflects few neutrons back towards the core from more than a few cm away, and consequently the beam tube cannot gain from neutrons emerging along the core away from E1 and F1, while with D2O, neutrons can be reflected into the beam tube from a large part of the core surface.

In comparison to Be-H2O or Be-D2O, the D2O reflector gives a significant gain in $\Phi$. This is due to the smaller cross-section of D2O compared to Be, which allows more neutrons to leak from the core, and the greater neutron mean free path, which allows neutrons which have diffused out to return towards the core. These effects are well known and largely account for the difference of the MURR (Be-C) and Orphée (D2O) reactors, which, apart from reflector configuration, have similar geometries. It should not be forgotten, however, that greater leakage implies lower reactivity and some loss in fuel cycle can be expected from the replacement of Be by D2O. Even more complex is the analysis of flux tilting within the reactor core as a result of a mixed reflector, e.g. Be on 3 sides and D2O on one side. This must be accounted for in any detailed design.

The gain of 6 in the beam tube flux is unexpected and it would be of great interest to see if calculations can reproduce these results. If representative, the gains associated with D2O may be larger than estimated from the unperturbed fluxes. This result emphasizes the need for new, sophisticated reactor codes. Such calculations are presently underway at Argonne.
A NEW MEDIUM-FLUX NEUTRON SOURCE FOR PURE AND APPLIED RESEARCH

G. DOLLING
Atomic Energy of Canada Research Company,
Chalk River Nuclear Laboratories,
Chalk River, Ontario

R. F. LIDSTONE
Whiteshell Nuclear Research Establishment,
Pinawa, Manitoba, Canada

Abstract
A NEW MEDIUM-FLUX NEUTRON SOURCE FOR PURE AND APPLIED RESEARCH.

A new multipurpose reactor concept is described that is intended for primary utilization as a medium-flux neutron source, or for a combination of materials testing, radiisotope production and beam-tube applications. The MAPLE (Multipurpose Applied Physics Lattice Experimental) concept combines light- and heavy-water-moderated lattices within a heavy-water-filled calandria. Thermal neutron fluxes in the range of 1 to $3 \times 10^{18} \text{n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ are produced by aluminium-clad, low-enrichment uranium silicide aluminium dispersion fuel elements. Heat removal of 10 to 25 MW is provided by light water flowing upwards from a lower plenum through flow channels in the core and an open chimney to plate-type heat exchangers. Two fuelling schemes are considered, compact-core loading for primary use as a neutron source, and distributed loading for general applications including fuel and materials testing. The relatively modest cost of the MAPLE reactor would make possible a much wider exploitation of the unique properties of thermal neutrons in materials research worldwide, exploitation that has hitherto been severely restricted by the great expense and limited availability of adequate neutron sources. As examples of neutron applications in engineering, descriptions are given of two examples of internal strain measurements, in (i) a curved portion of steam generator tubing, and (ii) an accidentally over-rolled section of Zircaloy pressure tube of the type used in CANDU power reactors.

1. INTRODUCTION

Neutrons from existing research reactors around the world have been used in many areas of materials research, non-destructive testing, isotope production, etc. Even though rather low flux sources ($< 10^{16} \text{n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$) such as the SLOWPOKE reactor [1] can be successfully employed in certain fields, most applications to date have been explored with sources in the $10^{17}$ to $10^{18} \text{n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ range, and these would benefit from even higher fluxes by way of shorter test times, higher throughput or better resolution. A general review of neutrons in engineering and technology has been given recently by Walker [2] and neutron radiography in particular is discussed by Barton [3]. The many uses of neutrons
FIG. 1. MAPLE reactor concept.
in medicine, for example for the production of radioisotopes for tracers, of \(^{60}\)Co \(\gamma\)-ray sources, boron capture therapy, and so on, have been reviewed by Fowler [4]. In addition, there is a large body of fundamental research in physics, chemistry and biology made possible by the advent of medium and high flux neutron sources [5]. Nevertheless, it is clear that exploitation of the uniquely valuable properties of the neutron is still at an early stage, severely restricted by the low flux of many present-day sources and the limited accessibility of the extremely few and extremely expensive high flux-sources. The relatively modest cost of the MAPLE reactor, described in section 2, should enable it to fill a much-needed role in materials research world-wide by making adequate neutron fluxes more affordable and hence more widely available.

In section 3, as examples of neutron applications in engineering, we describe how neutron diffraction provides a unique and non-destructive method for measuring internal strains in two manufactured components: a curved portion of steam generator tubing [6] and an accidentally over-rolled section of Zircaloy pressure tube [7] of a type used in CANDU power reactors.

2. THE MAPLE REACTOR CONCEPT

The MAPLE reactor concept, the horizontal layout of which is illustrated schematically in Fig. 1, comprises an open-tank reactor assembly within a light-water pool. A heavy-water-filled calandria vessel (1.80 m diameter by 0.80 m high) surrounds a central hexagonal grid that accommodates 29 fuel assemblies, reflector modules, or irradiation rigs with a lattice spacing of 83.5 mm. Fourteen fuel channels in the calandria surround the central lattice (167 mm spacing). The basic fuel unit is an aluminum-clad rod of uranium-silicide-aluminum-dispersion fuel meat, 600 mm long, containing 60 to 86 g of 20% enriched uranium. Standard assemblies for the central core contain 36 rods; 18-rod assemblies are used in the outer core and control sites in the inner core. Reactivity control absorbers operate in the water annulus outside the fuel of the control site.

Heat removal of 10 to 25 MW is provided by light water pumped to a lower plenum, upward through the core to the open chimney, and then through plate-type heat exchangers back to the lower plenum. About 10% of the primary coolant from the heat exchangers is delayed and then returned to the pool bottom to create a downward flow in the chimney that suppresses upward migration of activation products, e.g. \(^{15}\)N.

The MAPLE reactor is intended for primary use as a neutron source, or for multipurpose utilization for materials testing, radioisotope production, and beam-tube applications. Compact core loading of 12 to 14 fuel assemblies may be employed to maximize thermal neutral flux levels in the reflector for a given core power output. Larger core loadings of about 19 fuelled
TABLE I. UNPERTURBED THERMAL NEUTRON FLUX IN THE REFLECTOR AT 10 MW TOTAL CORE POWER OUTPUT

<table>
<thead>
<tr>
<th>Distance from Grenoble (mm)</th>
<th>HFR</th>
<th>ORPHEE</th>
<th>HFBR</th>
<th>JRR-3M</th>
<th>MAPLE (14 fuel sites)</th>
<th>MAPLE (28 fuel sites)</th>
<th>MAPLE (19 fuel sites)</th>
</tr>
</thead>
<tbody>
<tr>
<td>150</td>
<td>1.4</td>
<td>2.1</td>
<td></td>
<td>2.5</td>
<td>1.8</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>2.0</td>
<td>2.7</td>
<td>0.9</td>
<td>2.5</td>
<td>1.8</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>250</td>
<td>2.4</td>
<td>2.5</td>
<td>1.5</td>
<td>1.2</td>
<td>2.3</td>
<td>1.7</td>
<td>1.0</td>
</tr>
<tr>
<td>300</td>
<td>2.6</td>
<td>2.4</td>
<td>1.8</td>
<td>1.4</td>
<td>2.1</td>
<td>1.6</td>
<td>0.8</td>
</tr>
<tr>
<td>350</td>
<td>2.4</td>
<td>2.1</td>
<td>1.7</td>
<td>1.3</td>
<td>1.8</td>
<td>1.4</td>
<td>0.7</td>
</tr>
<tr>
<td>400</td>
<td>2.1</td>
<td>1.8</td>
<td>1.4</td>
<td>1.2</td>
<td>1.5</td>
<td>1.2</td>
<td>0.6</td>
</tr>
<tr>
<td>450</td>
<td>1.8</td>
<td>1.5</td>
<td>1.2</td>
<td>1.1</td>
<td>1.2</td>
<td>1.0</td>
<td>0.5</td>
</tr>
<tr>
<td>500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* cited in Reference [10].

sites facilitate a combination of isotope production and beam-tube applications. Of course, the core may also be fully loaded for high-power materials-testing applications.

The anticipated performance of MAPLE as a neutron source is compared in Table I to that of several existing heavy-water reflected designs for two compact core loadings and a fully loaded inner core. A compact loading of 14 fuel sites provides a flux quality comparable to ORPHEE [8] or HFR-Grenoble; a 19-site fuel loading permits fluxes per unit power equivalent to HFBR; full core loadings produce similar performance to the planned modification of JRR-3 [9].

The MAPLE neutronics calculations were performed with the multigroup three-dimensional diffusion code 3DDT [11], using the multigroup transport code, WIMS [12] to generate five-group microscopic cross-section sets for the various reactor regions. The WIMS library used 69 energy-group microscopic cross-sections based on ENDF/B-IV data.

3. SOME APPLICATIONS OF THERMAL NEUTRONS

3.1 Internal strains in steam generator tubes

The steam generator tubes in CANDU nuclear generating stations are made of a special alloy, Incoloy-800, formed into hairpins. Residual strains in a given tube after it has been bent to its design shape are undesirable, because of the increased possibility of stress-corrosion cracking when a concentration of salts in areas of localized dryout coincides with high tensile stress.

The spacings of the \{111\} and \{200\} planes in the longitudinal direction of the tube were measured as a function of position around the circumference. The "relaxed" value of plane spacing was measured on a piece of straight tubing. The strains derived from \{220\}, \{200\} and \{111\} planes in the radial and hoop directions were also measured as a function of circumferential
The longitudinal (or axial) lattice strain (Fig. 2) shows a maximum just above the neutral plane of the bent tube and a minimum just below it. The maximum longitudinal strain derived from \{200\} planes is almost twice that derived from \{111\} planes. The magnitude of the strains within the wall came as a surprise, since conventional X-ray methods [1] had indicated lower strains. The fact that the strain in the longitudinal direction could be different in differently aligned grains of a cubic material was also a surprise. Model calculations for a thin walled tube with elastic/plastic bending [14], followed by elastic unloading, confirm the shape of the residual strain curve. They suggest, moreover, that the anisotropy of Young's modulus in Incoloy accounts for the difference between the \{111\} and \{200\} strains. The Incoloy tubing is textured such that there are 5 times more \{111\} planes oriented along the tube axis than \{002\} planes. The \{111\} behaviour, a balanced bending moment, is more representative of the tube. However, the texture is not so strong that we can determine the complete strain tensor. The variation of lattice strain in both the hoop and radial directions around the circumference is the reverse of the longitudinal strain and about 25% of its value.

### 3.2 Lattice strains in over-rolled pressure tubes

In a CANDU reactor, the seal between the zirconium alloy pressure tube and its stainless steel end-fitting is a rolled
joint, made by inserting a rolling tool and expanding the tube into the end-fitting. The mouth of the fitting was slightly tapered for easy location and insertion of the tube in the fitting. In many cases the rolling tool was inserted too far, so that the tube was expanded into the taper, giving rise to a large circumferential residual tensile stress [15]. Under operating conditions, hydrogen diffused to the regions of high tensile stress. Platelets of zirconium hydride were formed perpendicular to the residual tension, and cracking eventually occurred. We have measured the lattice strain in the overrolled region with the neutron diffraction technique.

Figure 3 shows measurements of lattice strain in the hoop direction in a complete over-rolled tube as a function of distance from a point in the tube within the end fitting (0 mm) through the point of maximum insertion of the rolling tool (17.5 mm) to a region characteristic of the production tube. The lattice strain is determined with respect to the interplanar spacing at the free "unstrained" end and corresponds to the incremental elastic strain induced by rolling. Well within the end-fitting, where the tool presses the tube against the fitting the lattice is compressed, as one would expect; however, in the unsupported region there is a sharp increase in the (0002) interplanar spacing corresponding to the hoop direction. This tensile strain extends beyond the insertion point of the tool. A coupon was subsequently cut from the complete over-rolled tube for X-ray line broadening measurements and Fig. 3 also shows the lattice strain in the hoop direction of this coupon. Although the macroscopic hoop stresses no longer exist, as much as 50% of the original lattice strain is still present in the coupon. We believe that this phenomenon arises from grain-to-grain interactions. One conventional method of measuring residual stress involves observing the change in strain gauge readings when such a coupon is cut from a tube, and assuming that no stresses remain in the coupon. This approach would clearly fail to reveal the effects we have observed with neutron diffraction. Also shown in Fig. 3 is the lattice strain profile for a complete tube that was given a stress-relief heat treatment after the rolled joint was made.
REFERENCES


THE NEW NEUTRON GUIDE LABORATORY AT THE FRJ-2 (DIDO) REACTOR IN JÜLICH

K. WERNER, T. SPRINGER, J. DUPPICH
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

THE NEW NEUTRON GUIDE LABORATORY AT THE FRJ-2 (DIDO) REACTOR IN JÜLICH.

A hydrogen cold source has been working at the FRJ-2 (DIDO) reactor in Jülich since 1969 in connection with a neutron guide laboratory. In view of the anticipated needs of cold neutron facilities during the next 10 to 15 years, both the cold source and the guide laboratory are being replaced by a new installation. The following improvements are foreseen: (i) Neutron guides which are better illuminated, i.e., which obtain a beam whose divergency is close to the critical angle; (ii) $^{58}$Ni is used instead of natural nickel as coating of the guides; and (iii) a great number of new and more efficient instruments is foreseen, in particular in the field of small-angle scattering and high-resolution spectroscopy. The laboratory, the guides and the new source are nearly completed. The neutrons should be available during 1985. At present, seven new instruments are under construction.

1. INTRODUCTION

The FRJ-2 (DIDO) at the KFA Jülich is a heavy water reactor with a power of 23 MW and a peak thermal flux of $2 \times 10^{14} \text{s}^{-1} \cdot \text{cm}^{-2}$. In 1969 the reactor was equipped with a hydrogen source. The cold neutrons were conducted by a neutron guide to the reactor wall. At this point the guide was split into two separate parts, leading the neutrons to four instruments in a small external laboratory. A time-of-flight machine was connected with the beam inside the reactor hall /1/. The instruments have been intensively used for many research subjects, in particular for small-angle scattering in polymer and metal physics research, for disorder scattering, and for high resolution spectroscopy, mainly on molecular crystals (for a few examples see Refs /2-4/). Also, in the future, cold neutrons will play an important role in many research fields, in particular where high resolution in momentum and in energy transfer is desired. Therefore, in view of the anticipated needs in these fields, in 1982 it was decided to rebuild the whole laboratory and its instrumentation. Also, the cold source had to be replaced. In the meantime the project is reaching its final stage. The neutron guides and the instruments are under construction and are expected to be operating in 1985. The new laboratory should provide approximately 10 beam positions - one or two of them to serve for the developments and tests for the Jülich spallation source, SNO. In the following, a brief report on the project is given.
2. THE IN-PILE PART OF THE SYSTEM

The new cold source is similar to the earlier installation as described in /1/. Liquid hydrogen circulates in a closed loop inside the beam tube, which is condensed and cooled by helium gas from a refrigerator. The diameter of the moderator vessel is 15 cm. Its thickness has been optimized by the Monte-Carlo calculations /5/, using the Young-Koppel hydrogen gas cross-sections /6/. In addition, measurements were carried out at the SIN spallation source facility on moderators with different geometries. These studies yielded a value for the optimum thickness of 4 cm. For this value the gain of the source is fairly insensitive to the parahydrogen concentration in the range between 25 and 90%. (The thickness of the vessel which has been used so far was 6 cm).

The old beam tube, as well as the new one, contains a bismuth filter for the absorption of fast neutrons and gamma rays. It is a 48 cm long single crystal with (111) parallel to the beam axis. The bismuth is cooled by helium taken from the main source circuit. The filter is necessary and has been maintained because it allows the installation of straight guides. An essential design requirement of the beam tubes was to minimize the distance L between source and guide entrance which had to be 195 cm. As seen from the guide entrance, the range of angles which covers the full surface of the source is given by

\[ \gamma = \frac{(D-d)}{2L} \]

where D and d are the source and guide widths (in the horizontal or vertical direction), respectively. Full use of the critical angle of the guide \( \gamma_c(\lambda) \) is made if

\[ \gamma \geq \gamma_c(\lambda) = 2.04 \times 10^{-3} \times \lambda(\text{A}) \]

for a \(^{58}\text{Ni}\) surface.

The entrance of the in-pile section of the neutron guide has a cross-section of 10 x 10 cm\(^2\). It immediately bifurcates into two separate guides with dimensions of 10 x 7 cm\(^2\) and 10 x 3 cm\(^2\), respectively.

3. THE EXTERNAL NEUTRON GUIDES

The rectangular guides are constructed from ground and polished boron glass plates (1.5 cm thick) which are glued together. The inner surface is covered with a \(\sim 1000\) Å layer of \(^{58}\text{Ni}\), produced by vacuum evaporation. Measurements of the critical angle and of the reflectivity were carried out at the ILL reactor\(^1\) (to be published). The whole lay-out of the guide system is presented in Fig. 1.

---

\(^1\) ILL = Institut Max von Laue – Paul Langevin, Grenoble.
FIG. 1. Schematic sketch of the layout of the neutron guide system and the instrumentation. Starting from the cold source (CS) and passing the Bi-single crystal filter (Bi) the neutrons reach the guide entrance (A), (dimensions 10 x 10 cm²). A guide bifurcation splits the beam in two parts: NLI (7 x 10 cm²) and NLII (3 x 10 cm²). After the main shutter (S), NLII splits horizontally in the guides NLII A and NLII R (B) (3 x 4.5 cm² each). NLII and NLII A are straight, NLII R is bent with a radius R = 490 m. Before the reactor wall, the neutron guides cross-section is shown by (C). At about 25 m there are three independent guides (D). At 30 m a bender is built in guide NLII (E), which divides the beam: cross-sections are two of 2 x 4.5 cm² and one of 5 x 10 cm². At the end of the system, there are four separated guides (F).

Instruments guide I: TOF (time-of-flight instrument in the reactor hall), DNS (instrument for diffuse scattering) and BSI (backscattering machine I); Guide I A: Beta-NMR (in-beam magnetic resonance experiment); Guide I B: TAS (triple axis spectrometer) and SNO (test facilities for the spallation neutron source); Guide II A: DC (double crystal small-angle instrument), BS2 (backscattering machine 2) and SA 1 (small-angle scattering experiment 1); Guide II B: SA 2 (small-angle scattering experiment 2).
The part of the neutron guides inside the reactor hall is mounted in a vacuum casing, whereas the other part of the guides is directly evacuated. Two guides are straight, which provide a range of wavelengths between 2 and 10 Å. One of the guides is curved (not polygonal), with a radius of curvature of 490 m, which corresponds to a wavelength cut-off of 5 Å. A "bender" is introduced into one of the straight guides in order to create an additional end position. This bender has a length of 3.5 m and the radius of curvature is 57 m. It consists of nine channels each 0.17 cm wide, separated by glass walls of 0.05 cm thickness. The resulting deflection angle at the end of the bender is about 3.5° and the wavelength cut-off is at 3.8 Å. Some of the instruments will be equipped with neutron guide funnels with a parabolic profile, in order to increase the intensity at the sample.

4. INSTRUMENTS

The first generation of instruments under construction is nearly finished. Two of the end positions will be occupied by small-angle scattering instruments. For these instruments the neutrons are monochromatized by helical slit selectors with wavelengths between 3 and 10 Å and a resolution of \( \Delta k/k = 0.2 \). A slit system with variable collimation is foreseen with the object of adapting the incident and the outgoing divergencies. The total length (collimator section plus sample-detector distance) is 40 m for each of these instruments. A magnetized S-shaped multimirror can be introduced for work with polarized neutrons. A two-dimensional scintillation detector (Anger camera) is under construction.

One of the end positions is used for an in-beam NMR experiment where nuclei are excited by the capture of polarized neutrons. The fourth end position is foreseen for SNO test and prototype facilities. A neutron irradiation facility is also foreseen.

For the time being six instruments will be situated on side positions of the guides, namely

- a time-of-flight instrument with a focusing multicrystal; this instrument is inside the main reactor hall;
- an instrument to study diffuse scattering on disordered systems with a modest Q-resolution but high sensitivity and low background. The neutrons are detected and analysed by time-of-flight, or by a multicrystal analyser which consists of 32 focusing graphite crystals with counters;
- a backscattering spectrometer with 0.2 μeV resolution. A hydraulic Doppler drive allows preselected velocity scans to be carried out (see paper IAEA-CN-46/7P);
- a triple-axis spectrometer with a double monochromator as a test facility for the SNO spallation source;

\[ \text{2 Schätzler, R., et al., IAEA-CN-46/7P, these Proceedings.} \]
- a double crystal diffractometer. Two parallel ideal silicon single crystals serve as a collimator for scattering at very small angles. This instrument has a $Q$-resolution of $0.5 \times 10^{-5} \text{Å}^{-1}$.
- a second backscattering spectrometer without Doppler drive. It allows scans at zero energy transfer with an energy window of about 0.2 µeV ("fixed window spectrometer").

Part of these instruments and part of the instrumental time will be available for research groups from universities. It is our hope that the new guide laboratory will strengthen the high resolution work with cold neutrons. Furthermore, it will help to bridge the gap until a new neutron source becomes available.

REFERENCES
/1/ SCHMATZ, W., STILLER, H.H., Kerntechnik 13 (1971) 525.
    (b) PRESS, W., KOLLMAR, A., Solid State Comm. 17 (1975) 405.
/3/ SCHMATZ, W., "Treatise on Materials Science and Technology",
/4/ SCHMATZ, W., SPRINGER, T., SCHELTEN, J., IRBEL, K., J. Appl.
POSTER PRESENTATIONS
INSTRUMENTS (PULSED SOURCES)
(Session X.b)
Chairmen

T. SPRINGER
W. SCHMATZ

Federal Republic of Germany
RESOLUTION AND INTENSITY OF A TOF-TOF SPECTROMETER

R. E. LECHNER
Hahn-Meitner-Institut,
Bereich Kernchemie und Reaktor,
Berlin (West)

Abstract

RESOLUTION AND INTENSITY OF A TOF-TOF SPECTROMETER.

Intensity and resolution of multi-disc chopper spectrometers and aspects of their optimization are discussed. Theoretical results are compared to measurements and the two existing instruments (IN5 and MIBEMOL) are compared with each other.

Time-of-flight (TOF) spectrometers for inelastic neutron scattering using TOF techniques for the monochromatization of the incident neutron beam, as well as for the energy analysis of scattered neutrons have been applied for more than 25 years in the study of condensed matter. A particularly useful technique, which is known for its flexibility regarding the variation of the experimental resolution, employs a series of two or more disk choppers for creating the incident monochromatic neutron pulse /1/. It has been used with great success since 1973 in the IN5 multichopper spectrometer at the Institut Max von Laue - Paul Langvin (ILL) in Grenoble /2/. Another, rather similar instrument of this type was recently commissioned at the Orphée reactor in Saclay (MIBEMOL) /3/. In this paper we discuss intensity and energy resolution of such spectrometers as well as aspects of their optimization. For this purpose it will be sufficient to consider two choppers only, because the use of further choppers is not relevant in the present context, except for the definition of the TOF period. Minor effects on the intensity due to neutron flux attenuation along the flight path, and the influence of neutron guide divergence or sample geometry on the resolution, will not be considered here. Approximate formulae where such effects have
been neglected were derived elsewhere /4/. The number of neutrons incident on the sample is given by

\[ I \left[ \text{cm}^{-2} \text{s}^{-1} \right] = (1.7055/\lambda_0^4 \left[ \text{Å}^4 \right]) (\tau_1 \left[ \mu\text{s} \right] \tau_2 \left[ \mu\text{s} \right]) \]

\[ L_{12} \left[ \text{m} \right] L_{SD} \left[ \text{m} \right] (d\phi/dE_0 \left[ \text{cm}^{-2} \text{s}^{-1} \mu\text{eV}^{-1} \right]) \]

(1)

where \( d\phi/dE_0 \) is the average neutron flux per unit of energy incident on chopper 1 during its burst time \( \tau_1 \); \( \tau_2 = \) burst time of chopper 2 (FWHM); \( L_{12} = \) distance chopper 1 - chopper 2; \( L_{SD} = \) distance sample - detector; \( \lambda_0 \) is the incident neutron wavelength. According to this, for a given value of \( \lambda_0 \), the experimental intensity is governed by the factor

\[ F = (\tau_1 \tau_2) / (L_{12} L_{SD}) \]

(2)

The FWHM of the energy resolution function at the detector can be approximately expressed as

\[ \Delta(\hbar\omega) \left[ \mu\text{eV} \right] = 647.2 \left( A^2 + B^2 + C^2 \right)^{1/2} / (L_{12} L_{SD} \lambda^3) \]

(3)

with

\[ A = 252.77 \delta L \lambda L_{12} \]

\[ B = \tau_1 (L_{2S} + L_{SD} \lambda^3/\lambda_0^3) \]

(4)

\[ C = \tau_2 (L_{12} + L_{2S} + L_{SD} \lambda^3/\lambda_0^3) \]

Here \( \lambda \) is the scattered neutron wavelength, \( \delta L \) is the uncertainty in the length of the neutron flight path (finite sample and detector size), \( L_{2S} = \) distance chopper 2 - sample. The quantities \( L_{12}, L_{2S}, L_{SD}, \delta L \) must be given in \( \text{m} \), \( \lambda_0 \) and \( \lambda \) in \( \text{Å} \), \( \tau_1 \) and \( \tau_2 \) in \( \text{µs} \). It can be shown that the energy resolution width \( \Delta(\hbar\omega) \) is
Table I. Elastic energy resolution $\Delta(h\nu)$ [meV] for various ($\tau_1$, $\tau_2$) combinations, keeping constant the spectrometer distances and the intensity factor $F$.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Optimum values</th>
<th>Actual values</th>
<th>Optimum values interchanged</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>($\tau_1$ = 78.54 $\mu$s) ($\tau_2$ = 35.9 $\mu$s)</td>
<td>($\tau_1$ = 53.1 $\mu$s) ($\tau_2$ = 53.1 $\mu$s)</td>
<td>($\tau_1$ = 35.9 $\mu$s) ($\tau_2$ = 78.54 $\mu$s)</td>
</tr>
<tr>
<td>INS $\lambda_0 = 6.1\AA$</td>
<td>70.6 meV 17.3 meV</td>
<td>80.2 meV 19.3 meV</td>
<td>108.2 meV 25.4 meV</td>
</tr>
<tr>
<td>MIREMOL $\lambda_0 = 6.1\AA$</td>
<td>($\tau_1$ =110.8 $\mu$s) ($\tau_2$ = 39.68 $\mu$s)</td>
<td>($\tau_1$ = 66.3 $\mu$s) ($\tau_2$ = 66.3 $\mu$s)</td>
<td>($\tau_1$ = 39.68 $\mu$s) ($\tau_2$ =110.8 $\mu$s)</td>
</tr>
<tr>
<td></td>
<td>75.0 meV 18.6 meV</td>
<td>90.8 meV 21.8 meV</td>
<td>140.8 meV 32.8 meV</td>
</tr>
</tbody>
</table>

minimized for a given value of the intensity factor $F$ under the condition that $B = C$, i.e.

$$\frac{\tau_1}{\tau_2} = \frac{(L_{12} + L_{2S} + L_{SD} \lambda^3/\lambda^3)}{(L_{2S} + L_{SD} \lambda^3/\lambda^3)}$$

This equation shows that the conventional spectrometer design with $\tau_1 = \tau_2$ is generally not completely optimized, although the deviation from the optimum resolution width in existing instruments (see Table I) is only around 10 to 20%.
FIG. 1. Effect of the ($\Delta$($\hbar$ω))-dependent energy resolution $\Delta(\hbar\omega)$ (full curve) on pairs of tunnelling peaks of the substances Mn acetate, Pb acetate, CH$_4$-II and MDBP. It causes a difference in the measured peak widths for the same absolute values of energy gain and loss; measured elastic peak widths are close to the theoretical value; (experiments performed on IN5 with $\lambda_0 = 10 \ \text{Å}/5, 6/7$).

Table II. Parameter values of the spectrometers IN5 and MIREMOL

<table>
<thead>
<tr>
<th>Parameter</th>
<th>IN5</th>
<th>MIBEMOL</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L_{12}$</td>
<td>6.25</td>
<td>8.96</td>
</tr>
<tr>
<td>$L_{2S}$</td>
<td>1.286</td>
<td>1.42</td>
</tr>
<tr>
<td>$L_{SD}$</td>
<td>3.977</td>
<td>3.58</td>
</tr>
<tr>
<td>$T_1$</td>
<td>53.1</td>
<td>66.3</td>
</tr>
<tr>
<td>$T_2$</td>
<td>53.1</td>
<td>66.3</td>
</tr>
</tbody>
</table>
Let us now compare resolution calculations based on Eqs. (3) and (4) to experimental results. Within the approximations used good agreement is expected for cases where geometrical effects are small, i.e. for low scattering angles and for slab-shaped samples in reflection geometry, if the sample plane roughly bisects the scattering angle. Unfortunately, except for elastic scattering, no sufficiently precise scattering data from $\delta$-function-like cross-sections were available to us. An indirect comparison is, however,
possible using intrinsically broadened tunnelling peaks. Figure 1 suggests that the measured widths of such peaks for four different samples /5,6/ reflect the almost linear variation with $h_\omega$ of the resolution width $\Delta(h_\omega)$ (full curve) in the considered energy range. The measured elastic peak widths, which exhibit no natural line broadening, are rather close to the theoretical curve (within 2 to 5%).

Finally we would like to use formulae (1) to (5) in order to compare the two existing multi-disk-chopper spectrometers in their standard modes of operation: IN5 at 15000 rpm and MIREMOL (with pairs of counter-rotating choppers /3/) at 10000 rpm. The relevant parameter values are given in Table II; for $\delta l$ we have assumed a value of 0.02 m. We obtain $F(IN5)/F(MIREMOL) = 0.828$ for the ratio of intensity factors. The energy resolutions of the two spectrometers as a function of $\lambda_0$ and $\lambda$ are compared in Figs. 2(a) and 2(b) with the aid of quasi-three-dimensional plots. It is seen that the qualitative behaviour of the resolution functions is very similar. Quantitative differences of up to $\sim 20\%$ are observed, the resolution of IN5 being slightly better for $\lambda < \lambda_0$, whereas that of MIREMOL is somewhat better for $\lambda >> \lambda_0$.

ACKNOWLEDGEMENTS

Thanks are due to Professor S. Clough, Dr. A.H. Horsewill and Dr. M. Prager for making available their line-width data.

REFERENCES

/2/ DOUCHIN, F., LECHNER, R.E., SCHERM, R., to be published; (see also: Neutron Research Facilities at the ILL High Flux Reactor (Dec. 1983) 75-77.


BUNCHING SPECTROMETER*

D. RICHTER, B. ALEFELD
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

BUNCHING SPECTROMETER.

A time-of-flight focusing spectrometer is proposed which achieves time focusing through a monochromator moving at constant acceleration. Its performance is compared with that of conventional TOF machines on the low resolution side, and with TOF backscattering spectrometers for high resolution. In the resolution range of $10^{-2}$ a bunching spectrometer gains an order of magnitude in intensity compared to a double-chopper spectrometer. For $10^{-3}$ resolution it is comparable with graphite backscattering machines.

INVERTED GEOMETRY MULTIPURPOSE SPECTROMETER FOR PULSED NEUTRON SOURCES


*Institute of Nuclear Physics, Kraków, Poland
**Joint Institute for Nuclear Research, Dubna

Abstract

INVERTED GEOMETRY MULTIPURPOSE SPECTROMETER FOR PULSED NEUTRON SOURCES.

The spectrometer is designed to study inelastic (INS) and quasi-elastic (QNS) neutron scattering with simultaneous phase control of a sample by neutron powder diffraction (NPD). The INS spectra can be analysed in a wide range of energy transfer (0 - 2500 meV) with different resolutions given by a composite Be filter and single crystal analysers. The QNS can be studied in the range of momentum transfer 0.1 - 3.0 Å⁻¹ with resolutions varying from 600 peV down to 20 peV.

Analysis of the neutron energy by a time-of-flight method allows a simultaneous investigation of the dynamics and structure of condensed matter /1/. Such types of investigation are particularly important for the substances exhibiting polymorphism, especially when the phase transitions are accompanied by the simultaneous changes of the structure, lattice dynamics and stochastic motions of the molecules or molecular groups.

The scheme of the spectrometer is shown in Fig. 1. It consists of four modules which enable a simultaneous measurement of inelastic (INS) as well as quasi-elastic (QNS) neutron scattering and also neutron diffraction (NID) on the polycrystalline samples. Module (11) contains three sections: two of them are for high-resolution QNS studies (ΔE ≈ 20 µeV) at the small or large scattering angles, and the third for diffraction. Modules (12) and (14) are identical. Each consists of eight sections shifted by 10°. They are assigned for the INS and QNS (ΔE ≈ 20 - 600 µeV) measurements. Module (13) contains four sections: two of them for diffraction measurements, the other two for the INS studies at the high scattering angles (170° and 175° as in the figure) or small angles (5° and 10°) when module (13) is exchanged with module (11) by rotating around the spectrometer axis. Operating modes and physical parameters of the spectrometer at the IBR-2 pulsed reactor and the spallation neutron source (SNS) have been discussed.
Simple versions of this type of spectrometer /2,3/ have been recently constructed at the IRR-30 and IBR-2 pulsed reactors of JINR at Dubna. First experiments performed with the use of these spectrometers show the usefulness of simultaneous measurements of INS, ONS and NO spectra for studies of different solid phases of complex molecular crystals /2,4/, ferroelectric and superionic materials /5/ and metal hydrides /6/.

REFERENCES

A SPECTROMETER FOR DIFFUSE SCATTERING AT A PULSED NEUTRON SOURCE

J. SCHELTEN, G. BAUER
Projekt Spallations-Neutronenquelle (SNQ),
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

A SPECTROMETER FOR DIFFUSE SCATTERING AT A PULSED NEUTRON SOURCE.
To measure elastic-diffuse defect scattering of subthermal neutrons from a pulsed neutron source at the SNQ, a new spectrometer is proposed. Its essential feature is a highly symmetric mechanical velocity selector with concentric cylinders of bars and gaps rotating around the sample. The velocity selector is transparent for incoming and outgoing neutrons of a given wavelength within a relative wavelength band of ±10%. Thus, only elastically scattered neutrons can reach the detector. Such a spectrometer at a pulsed neutron source has an intensity gain of 10 to 20 with respect to a conventional spectrometer for diffuse scattering at a continuous neutron source if both sources have identical average fluxes.

1. INTRODUCTION

The elastic diffuse-scattering of subthermal neutrons contains valuable information about the defect structure of a crystal. From the measured scattering intensity as a function of the scattering vector \( \mathbf{Q} \) one is able to determine

(i) The small defect concentration in the range of 100 ppm;
(ii) The defect site, i.e. whether the defect is a substitutional or interstitial one, whether it is at an octaeder or tetraeder site, etc.;
(iii) The distortion field caused by the defect;
(iv) The effective interaction potentials between the atoms of the host lattice by fitting a theoretical model to the experimental data.

The optimum parameters for a spectrometer for diffuse-elastic scattering are:

(i) The spectrometer should be placed at the end of a long neutron guide and should utilize 4-8 \( \AA \) neutrons.
(ii) It may use a relatively broad wavelength band of \( \Delta\lambda/\lambda \sim 10\% \).
(iii) A poor energy and momentum resolution are sufficient.
(iv) A crude separation of the weak elastic defect scattering from the much stronger thermal diffuse scattering of the host lattice is mandatory.
In conventional spectrometers for diffuse scattering at continuous neutron sources the separation is accomplished in a time-of-flight method, where only about 5% of the primary neutron intensity can be utilized. In conventional spectrometers a 2 cm wide neutron beam illuminates the sample. The scattered intensity is recorded by a series of 2 cm dia. counters at a distance of 80 cm from the sample. A chopper in front of the sample produces ≈ 100 μs neutron pulses with a duty cycle of ≈ 5%. The energy resolution is around 1 meV.

2. DIFFUSE SPECTROMETER AT A PULSED SOURCE

At the end of a 20 m long neutron guide a burst of neutrons with a wavelength band of Δλ/λ = 10% from a pulsed neutron source has already a pulse width of 2 and 4 ms for 4 and 8 Å neutrons, respectively. Thus, the pulse width becomes comparable with the pulse distance of 10 ms for the SNO. Therefore, a separation between elastic and inelastic scattering cannot be achieved by time-of-flight without introducing a chopper which is at the expense of scattering intensity.

The idea of the proposed spectrometer for diffuse scattering is based on a mechanical velocity selector which simultaneously monochromatizes the neutron beam and analyses the scattered beam, i.e. only neutrons of wavelength λ preselected by the rotation per minute and within the wavelength band Δλ/λ arrive at the sample and only elastically scattered neutrons can pass the mechanical velocity selector in order to reach the detector.

The new mechanical velocity selector has a vertical axis of rotation. It consists of six concentric cylinders which rotate around the sample. Each cylinder is equipped with gaps and bars of equal widths. The rotational phase angles of the cylinders are zero or π, i.e. bars and gaps oppose bars and gaps or gaps and bars. Owing to this highly symmetric arrangement a neutron beam of a particular wavelength, which passes all concentric cylinders from outside, can also pass them from inside. A possible arrangement of bars and gaps on six cylinders is shown in Fig. 1. In the figure legend further details are presented.

One easily verifies that the velocity selector is transparent for neutrons with wavelengths λ, 3λ, 6λ, etc. Thus, a suppression of the contaminants 3λ and 6λ is needed, which can easily be done either by an additional conventional mechanical velocity selector or by time-of-flight.

The proposed spectrometer for diffuse scattering with the velocity selector for incoming and outgoing neutrons is shown in Fig. 2. The spectrometer still has the parameters of optimum performance as mentioned above. It can be placed at the end of a 20 m long neutron guide, it uses neutrons of 4 to 8 Å wavelength with a relatively broad wavelength band of Δλ/λ = 10%, and performs a crude-energy resolution in the 0.5 meV range.
FIG. 1. Mechanical velocity selector with six concentric cylinders consisting of bars and gaps of equal width. Radii of the largest and smallest cylinders are 80 cm and 30 cm. Rotational phase angles of cylinders 1 to 4 and 6 are zero and the phase angle of cylinder 5 is $\pi$. The transmission is 50%. For a wavelength resolution of 12.5% the specifications are: the largest bar/gap width $d_6 = 1$ cm, the smallest $d_1 = 0.38$ cm. The rotation frequencies are 50.9 Hz and 25.5 Hz for 4 Å and 8 Å neutrons, respectively. The technical performance of such a velocity selector does not cause serious problems.

FIG. 2. Layout of the proposed spectrometer for diffuse scattering. N - neutron guide; S - sample; V - velocity selector with concentric cylinders; D - neutron counters; sample to detector distance ~80 cm.
3. DETECTOR SIGNAL AND INTENSITY GAIN

The time-dependent detector signal (time proportional wavelength $\lambda$) is given by (1)

$$I(\lambda) = g(\lambda) \cdot \int S(\lambda,\lambda') g(\lambda') \, d\lambda'$$

(1)

where $g(\lambda)$ is the triangular-shaped characteristic of the velocity selector for incoming and outgoing neutrons which are scattered according to $S(\lambda,\lambda')$. This intensity distribution may be recorded with 40 time-channels of $\approx 250 \mu s$ width over a range of a few half widths.

For purely elastic scattering the detector signal is

$$I_{el}(\lambda) = g^2(\lambda)$$

(2)

which can be assumed to be known. Its half width is $\sqrt{3}/\sqrt{5}$ times the half width of $g(\lambda)$. If both elastic and inelastic contributions with small energy transfers are present the purely elastic contribution can be obtained by fitting $I_{el}(\lambda)$ to the measured intensity distribution $I(\lambda)$ of (2). Such a peak profile analysis is helpful in separating the purely elastic defect scattering from multi-phonon scattering of the host lattice which can lead to unresolved scattering contributions with small energy changes.

The intensity gain of the proposed spectrometer with respect to the sketched conventional spectrometer at a continuous source is large since no neutrons are wasted by chopping the beam. Its ultimate gain factor of 10 to 20 depends on the transmission factor $T$ of the new velocity selector since the scattered intensity is proportional $T^2$. Designs with $T > 50\%$ are possible.
CONSTANT-q SCAN AT A PULSED NEUTRON SOURCE

J. SCHELTON, B. ALEFELD, A. KOLLMAR
Projekt Spallations-Neutronenquelle (SNQ),
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

By modifying a triple axis spectrometer with a vibrating collimator and analyser crystal a constant-q scan can be performed during each neutron pulse from a pulsed neutron source. Formulas for the amplitudes of the angular vibrations at a frequency equal to the repetition rate are derived and their values are discussed. Typically such a time-of-flight constant-q scan covers an energy transfer range of 10 half-widths of a measured phonon peak and leads to an intensity gain of up to 10.

1. INTRODUCTION

In classical neutron spectroscopy scattered intensity is deliberately measured with a triple-axis spectrometer step by step at particular points in the four-dimensional \( q_x \omega \) space. Frequently, constant-q or constant \( \omega \) scans are pursued through phonon dispersion planes. These traditional methods can be performed at a pulsed neutron source for each neutron burst with a modified triple-axis spectrometer.

This paper describes the modifications of the spectrometer, demonstrates that a constant-q scan can be pursued, and estimates the intensity gain of the spectrometer at pulsed sources.

2. MODIFIED TRIPLE-AXIS SPECTROMETER

To perform a scan for each neutron burst the spectrometer parameters have to be changed periodically with the repetition rate \( \omega \) of the pulses. For this purpose the analyser crystal is mounted on a torsion pendulum with a resonance frequency close to the repetition rate which vibrates with frequencies \( \omega \). This angular motion of the analyser crystal changes sinusoidally the Bragg angle \( \theta \) and thus the final neutron energy \( E_f \) about an average value. Simultaneously, the initial neutron energy \( E_i \) decreases continuously with time for each pulse. Amplitude and phase of the vibration can be adjusted such that - to first order - scans can be made for which the direction of the phonon vector remains constant.
FIG. 1. Layout of the modified triple-axis spectrometer at a pulsed neutron source.

$N$ - neutron guide; $V$ - mechanical velocity selector; $S$ - sample; $A$ - analyser crystal; $D$ - detector; $\varphi$ - scattering angle; $\theta$ - Bragg angle; $\Delta \varphi, \Delta \theta$ - vibration amplitudes; $k_i, k_f$ - neutron wave numbers before and after the scattering process.

Additional adjustable scan parameters are needed to keep the direction and modulus of the phonon vector constant. Such parameters are obtained from another vibrational mode. After the sample the collimator is also mounted on a torsion pendulum with resonance frequency close to $\omega$. This angular motion changes sinusoidally the scattering angle $\varphi$. Amplitude and phase of both vibrations are adjusted with respect to the neutron burst in order that - at first order - the scan becomes for each neutron pulse a constant-$q$ scan about a deliberately chosen $\omega_0, q_0$ value. The layout of the modified triple-axis spectrometers for constant-$q$ scans is shown in Fig. 1.

Neutrons of 2 to 4 Å wavelength from a 25 to 50 m long neutron guide are used. For a pulsed neutron source with SNO values ($\omega/2\pi = 100$ Hz, $\Delta t = 250$ μs) the wave-number resolution is due to the large flight time $t_i$ in the range of 1%. The mechanical velocity selector prevents neutrons with much shorter and longer wavelengths from earlier and later pulses arriving at the spectrometer at the same time. The time dependence of the primary wave-number is $k_i + \Delta k_i(t)$ and at first order determined by

$$\Delta k_i(t) = -\frac{k_i}{t_i} t \quad (1)$$

The angular motion of the collimator, which tunes the scattering angle, and of the analyser crystals which in addition change the Bragg angle, are described by

$$\varphi = \varphi_0 + \Delta \varphi_0 \sin \omega t \quad (2)$$

$$\theta = \theta_0 + \Delta \theta_0 \sin \omega t + \Delta \theta_0 \sin \omega t \quad (3)$$
The values of $k_1$, $\varphi$, and $\theta$ at $t = 0$ are set in such a way that, at a value $\omega_q$ close to or on a phonon dispersion curve, the scattered intensity is measured.

So far the actual parameter changes have been described. From the triangle $P_1P_2P_3$ in Fig. 2 one derives how the parameters should change in order to pursue a constant-$q$ scan. The conditions are

$$\Delta \varphi = - \frac{\Delta k_1}{k_f} \sin \varphi_0$$  \hspace{1cm} (4)

$$\Delta k_f = + \Delta k_1 \cos \varphi_0$$  \hspace{1cm} (5)

With the definitions (1) to (3) and the Bragg equation in its differential form

$$\Delta k_f = - k_f \cot \theta \left[ \Delta \theta_0 \sin \omega t - \Delta \varphi_0 \sin \omega t \right]$$  \hspace{1cm} (6)

inserted in (4) and (5), the amplitudes of the angular vibrations are determined with

$$\Delta \varphi_0 = \Delta \varphi_{\text{max}} \times \sin \varphi_0$$  \hspace{1cm} (7)

$$\Delta \theta_0 = \Delta \varphi_{\text{max}} \times \left[ \frac{\tan \delta_0}{\tan \varphi_0} + 1 \right]$$  \hspace{1cm} (8)

$$\Delta \varphi_{\text{max}} = \frac{1}{2\pi} \frac{T}{T_1} \frac{k_1}{k_f} \quad \text{and} \quad T = 2\pi/\omega$$  \hspace{1cm} (9)
For typical values $T = 10$ ms, $4 \, \text{Å}$ neutron, 25 m distance and $k_f = k_i/2$, $\Delta \varphi_{\text{max}}$ is $7^\circ$.

The condition of a constant-$q$ scan can only be approximately fulfilled during the time $T/2$ of a half wave. Furthermore, during this time there is a mismatch since the angular motion is sinusoidal; however, it should be linear. Therefore, the useful scan time is limited to the time interval $[-T/8, T/8]$ since, within this interval, changes of the direction and modulus are sufficiently small in almost all practical cases.

A specialized case is a backscattering instrument which is modified by a sinusoidally moving analyser crystal with a matching of this Doppler motion to the time dependence of the wave number of the incoming neutrons.

3. INTENSITY GAIN

Within the useful time interval of $\pm T/8$ during a scan one spans the range $\Delta h\omega$ of energy transfer which may be related to the primary energy resolution $\delta E$. The ratio of both is

$$\frac{\Delta h\omega}{\delta E} \approx \frac{T}{4\Delta t} \left[ 1 - \frac{k_f}{k_i} \cos \varphi_0 \right]$$

For the SNQ with $T = 10$ ms and $\Delta t = 250$ μs the ratio is around ten. Thus, a phonon peak is scanned over about 10 half-widths. If this relatively broad scan of a phonon peak is really needed, for instance for a decent background subtraction and for a careful line profile analysis, the intensity gain of the modified triple axis spectrometer at a pulsed source with respect to a conventional instrument at a continuous source is just this factor of 10.

4. TORSION PENDULUM

The following data demonstrate that the vibrations of the collimator and crystal are not difficult to realize. A 1-m-long steel post of 1 cm radius carrying the collimator represented by the moment of inertia of 10 kg $\times$ (10 cm)$^2$ has a torsion resonance frequency of 100 Hz. The sheer angle is $\gamma = 0.01 \Delta \varphi_0$. The stored energy is $U = 20 \times \Delta \varphi_0^2$ kW·s.

With a quality factor of $Q = 300$ for this mechanical resonator the dissipated energy is $P = 20 \times (\Delta \varphi_0)^2$ kW. For $\Delta \varphi_0 = 20^\circ$ the shearing of $\gamma = 3 \times 10^{-3}$ is still in the elastic region and the energy dissipation is $P = 2$ kW.
CRYSTAL AND CHOPPER SPECTROMETERS ON PULSED NEUTRON SOURCES

C. J. CARLILE, A. D. TAYLOR, W. G. WILLIAMS
Neutron Division, Rutherford Appleton Laboratory,
Chilton, Didcot,
United Kingdom

Abstract

This paper discusses the choice of a monochromating device for the incident beam of a direct geometry inelastic scattering spectrometer on a pulsed source such as the United Kingdom Spallation Neutron Source (SNS). The resolutions, dynamic ranges and fluxes achievable with crystals and phased choppers are calculated for the incident energy range $20 < E_i$ (meV) $< 500$. The practicalities of realizing a spectrometer to provide 1% energy transfer resolutions over a large $(Q,\epsilon)$ range are also considered. It is shown that the chopper spectrometer is the better choice for the upper part of this incident energy range, but that the crystal method, particularly in a double monochromator arrangement, may offer advantages at lower energies. It is felt that the most versatile spectrometer is a hybrid one, and preliminary recommendations are made on a suitable spectrometer design for the SNS.

1. INTRODUCTION

Direct geometry time-of-flight inelastic neutron scattering spectrometers have long been used on steady state sources to measure the dynamical structure factor and frequency distribution of isotropic systems. The scientific interest [1] in such spectrometers comes from fields as diverse as excitations in solids, liquids, amorphous and disordered systems, magnetic scattering and molecular spectroscopy. In this paper we discuss the characteristics of the equivalent pulsed source spectrometer and show that an advanced spallation source such as SNS [2] may be used to improve the resolution and expand the kinematic range available. Such a spectrometer must cover a wide and variable region of $Q$-$\epsilon$ space. This may be achieved by a versatile monochromating device capable of providing a variety of incident energies at an appropriate high resolution, say 1%, and a wide but closely spaced set of scattering angles. The latter is provided straightforwardly by a large array of time-sorted detectors. The choice of the former, covering 20 meV to 500 meV, is the subject of this paper.
2. RESOLUTION

We shall restrict our discussion to two monochromating devices, namely Bragg scattering from a single crystal or velocity selection by a mechanical chopper phased to the pulsed source. These determine the incident energy, with some associated uncertainty, and time-of-flight to the detector, with an associated uncertainty, is used to determine the scattered neutron energy. A description of the important parameters which determine the resolution is aided by reference to the distance-time diagram shown in Figure 1. The moderator, chopper, scattering sample and detector are denoted by M, CH, S and D respectively. The crystal acts as a wavelength selector with resolution

\[ R' = \frac{\Delta \lambda}{\lambda} = \cot \theta \Delta \theta = \frac{\Delta t'}{t} \]  

independent of its location in the primary beam. \( E_1 \) and \( E_2 \) represent the incident and final neutron energy and the energy transfer, \( \epsilon \), is given by

\[ \epsilon = E_1 - E_2 \]  

Primes are used to denote time widths (FWHM) which are related to velocity uncertainties and double primes indicate pure timing errors. The chopper open time, \( \Delta t'_{CH} \), is a combination of an intrinsic burst time defined by the slit package and a sweep time.
across the moderator face. Focussing conditions [3], which may be used to reduce the sweep contributions and are only important at high energies, will not be discussed here.

For both spectrometers the measurable parameter is the time-of-flight to the detector, \( t_D \). An effective energy transfer is obtained by assuming a well-defined incident energy thus allowing the final flight time, and hence the final energy, to be determined. In order to determine the error in \( \varepsilon \) we associate the timing error at the detector \( \Delta t_D \) with an energy uncertainty in \( E_2 \) (and hence \( \varepsilon \)):

\[
\Delta \varepsilon = \frac{\partial \varepsilon}{\partial t} \Delta t = 2 \frac{E_2}{t_2} \Delta t_D
\]

For the crystal spectrometer a purely dispersive time width \( \Delta t_S' \) at the sample due to the resolution of the monochromator is propagated, after inelastic scattering, to an observed width at the detector

\[
\Delta t_D' = \Delta t_S' \left[ 1 + \frac{L_2 E_1}{L_1 E_2} \right]^{3/2}
\]

Interpreting this width as an uncertainty in \( \varepsilon \), from equation (3):

\[
\Delta \varepsilon' = \frac{\Delta t_D'}{E_1} \left[ 1 + \frac{L_1}{L_2} \left( 1 - \frac{\varepsilon}{E_1} \right) \right]^{3/2}
\]

where \( R' = \Delta t_1'/t_1 \) is the resolution of the monochromator. Non-dispersive time uncertainties, such as the moderator pulse width, contribute a term, from equation (3):

\[
\Delta \varepsilon'' = \frac{\Delta t_M}{t_2} = \frac{\delta_m}{L_2} \text{ and } \delta_m \text{ is (for the SNS) the 28 mm uncertainty associated with the moderator in the slowing down part of the spectrum. The overall resolution of the crystal spectrometer is then the sum in quadrature of equations (5) and (6):
}

\[
\frac{\Delta \varepsilon}{E_1} = \left[ \left\{ 2R' \left[ 1 + \frac{L_1}{L_2} \left( 1 - \frac{\varepsilon}{E_1} \right) \right]^{3/2} \right\}^2 + \left\{ 2R'' \left[ 1 - \frac{\varepsilon}{E_1} \right] \right\}^2 \right]^{1/2}
\]
A similar expression may be derived for the case of a chopper spectrometer. The contribution to the velocity selection from the chopper open time $\Delta t_{CH}$ gives, in an analogous way to the crystal:

$$\frac{\Delta \varepsilon'}{E_1} = 2R_1' \left[ 1 + \frac{d_1 + d_3}{d_2} \left( 1 - \frac{\varepsilon}{E_1} \right)^{3/2} \right]$$  \hspace{1cm} (8)

with $R_1' = \Delta t_{CH}/t_{CH}$. The chopper however also has an additional dispersive component due to the moderator pulse width:

$$\frac{\Delta \varepsilon'}{E_1} = 2R_2' \left[ 1 + \frac{d_3}{d_2} \left( 1 - \frac{\varepsilon}{E_1} \right)^{3/2} \right]$$  \hspace{1cm} (9)

with $R_2' = \Delta t_{M}/t_{CH} = \delta_m/d_1$

The overall resolution of a chopper device is then the sum in quadrature of the two dispersive terms:

$$\frac{\Delta \varepsilon}{E_1} = \left[ \left( 2R_1' \left[ 1 + \frac{d_1 + d_3}{d_2} \left( 1 - \frac{\varepsilon}{E_1} \right)^{3/2} \right] \right) + \left( 2R_2' \left[ 1 + \frac{d_3}{d_2} \left( 1 - \frac{\varepsilon}{E_1} \right)^{3/2} \right] \right) \right]^{1/2}$$  \hspace{1cm} (10)

Since both $R_1'$ and $R_2'$ are inversely proportional to $d_1$, the chopper spectrometer requires large values of the primary flight path for high resolution. In the crystal spectrometer $R'$ and $R''$ do not depend on $L_1$, and in fact a short $L_1$ is preferable. The functional dependence of the dimensionless resolution $\Delta \varepsilon/E_1$ on the fractional energy transfer $\varepsilon/E_1$ is clearly different in the two cases so that it is not immediately obvious at what value of $\varepsilon/E_1$ we should compare performances. As an example, we calculate the resolution of a chopper spectrometer with $d_1 = 9m$, $d_3 = 1m$ and $d_2 = 4m$ and with $\Delta t_m$ matched to $\Delta t_{CH}$ and compare this to a $L_1 = 7m$, $L_2 = 4m$ crystal spectrometer with $R'$ chosen to match the chopper's resolution at $\varepsilon = 0$. These calculations are illustrated in Figure 2. From this figure we note that the overall behaviour of the two functions is sufficiently similar that we may choose to match at other values of $\varepsilon/E_1$ without introducing a significant bias. In the $\varepsilon = 0$ limit, the resolution of both spectrometers is the convolution of $R'$ or $R''$ terms, with the $R'$ terms amplified by geometric factors. The $\varepsilon = E_1$ limit differs in the two cases. For the chopper it is a convolution of two $R'$ terms, but in the crystal case it simply contains one $R'$ term, that due to the resolution of the monochromator itself. Figure 2 also illustrates the fractional energy transfer required to provide a given fractional energy resolution $\Delta \varepsilon/\varepsilon$. The area of $Q-\varepsilon$ space accessible at a given
FIG. 2. The resolution in energy transfer versus energy transfer in reduced units for a crystal and a chopper monochromator matched at $\varepsilon = 0$. Fractional energy transfer resolutions are also indicated.

FIG. 3. The variation in fractional energy transfer resolution over $Q-\varepsilon$ space.

Resolution is significantly less than that given by the kinematic condition:

$$\frac{h^2 Q^2}{2m} = E_1 + E_2 - 2\sqrt{E_1 E_2} \cos \phi$$  \hspace{1cm} (11)

where $\phi$ is the scattering angle.
Figure 3 shows contours of $Q-\xi$ space accessible for a set of resolutions in the case of a direct geometry instrument with infinitely variable $E_1$. A reasonable 'rule of thumb' which balances $Q-\xi$ range and resolution is the condition

$$\frac{1}{3} \leq \frac{\xi}{E_1} \leq \frac{2}{3}$$

(12)

We have not considered the scattering vector resolution $\Delta Q$ in any detail in this work, however the main contributions come again from $\Delta t_D$, (see equation (3)) together with a term due the uncertainty in the scattering angle $\phi$. For the crystal spectrometer and isotropic beam divergences we find:

$$\Delta Q = \frac{2mE_2}{h^2Q}\left\{\frac{2E_2}{mL_2}\left[E_1^{\frac{1}{2}}\cos\phi - E_2^{\frac{1}{2}}\right]^2 \Delta t_D^2 + E_1\sin^2\phi \Delta \phi^2\right\}^{\frac{1}{2}}$$

(13)

with $d_2$ replacing $L_2$ for the chopper spectrometer. In general a more detailed expression which resolves the parallel and perpendicular components of $Q$ will be necessary.

3. INTENSITIES

For comparison purposes we consider the crystal and chopper spectrometers to have identical secondary flight paths of $4m$, and to view the 95K SNS liquid methane moderator. The comparison therefore entails calculating the intensities at the scattering sample positions for the same energy transfer resolution which we set at 1% for $\epsilon = E_1$ i.e. $\Delta E_1/E_1 = 0.01$. The intensity at the sample is given by:

$$I(E_1) = \frac{6 \times 10^{12}}{L^2} \frac{\Delta E_1}{E_1} T(E_1)$$

(14)

where $L \equiv L_1 \equiv d_1 + d_3$ and $T(E_1)$ is the transmission efficiency of the device.

a) Crystal Spectrometer

We begin with the premise that the instrument has a fixed take-off angle (20), this being largely dictated by the difficulty in rotating a large heavily-shielded detector. At fixed resolution this immediately determines the acceptable beam divergence at the monochromator since $(\Delta E_1/E_1) = 2\cot\theta_0 \Delta \theta$. This relation imposes appreciable intensity penalties at high incident energies where both $\theta$ and $\Delta \theta$ are small, and this is the principal reason why
The crystal spectrometer intensities were calculated using:

\[ I(E_1) = \frac{6 \times 10^{12}}{L_1} \frac{2R(\theta) \cot \theta T'}{L_1} \]  

(15)

where \( R(\theta) \) is the integrated crystal reflectivity as calculated using the transmission geometry formalism of Popovici and Gelberg [4] which includes extinction, and \( T' = 0.8 \) is a factor to account for real crystal performances compared with calculated ones; this latter factor has been justified experimentally [5] for copper crystals. Intensities were calculated for a room temperature Cu
monochromator with $L_1 = 10 \text{m}$, which provides full illumination and 1% incident energy resolution when $\theta = 90^\circ$. These intensities are shown in Figure 4A, though it must be emphasised that this performance cannot be achieved continuously over the whole range, as the calculation is based on optimised crystal thicknesses and mosaics at selected energies. These fluxes can be readily enhanced by i) a factor $\times 2$ by using reflection rather than transmission geometry and ii) a further factor $\times 2.5$ by using a number of monochromators in a 'vertical' focussing array [6]; the estimated intensities are then given by Figure 4B. Pyrolytic graphite (PG) is a more efficient monochromator than Cu in the thermal and cold neutron region and we give estimates of the intensities for a single PG monochromator in Figure 4C using the method of Riste and Ottes [7]. Finally we have considered the implications of using a double PG crystal array in a vertical focussing geometry and show the calculated fluxes in Figure 4D. We believe that these fluxes should be relatively easy to achieve in practice, and this arrangement provides the facility of allowing the incident energy to be changed continuously. (See Section 4).

b) Chopper Spectrometer

It is first necessary to determine the instrument geometry i.e. the source-chopper distance $d_1$ that gives, for matched moderator and chopper open times (or $R_1' = R_2'$), a resolution $2 R_\text{CH}$ of 1%. A solution of equations:

$$R_\text{CH} = \left[ (R_1')^2 + (R_2')^2 \right]^{1/2} = 0.005 \quad (16)$$

and

$$R_2' = \frac{5 \text{m}}{d_1} = \frac{1}{36d_1} \quad (17)$$

determines the geometry $d_1 = 7.86 \text{m}$, $d_3 = 1 \text{m}$, $d_2 = 4 \text{m}$. The intensity on the sample, equation (14), is in this case:

$$I_\text{CH}(E_1) = \frac{6 \times 10^{12}}{(d_1 + d_3)^2} \frac{2R_\text{CH}(E_1)}{T(E_1)} \quad (18)$$

$$= 5.4 \times 10^4 \frac{T(E_1)}{\text{n/cm}^2/\text{s}} \quad (19)$$

where $T(E_1)$ is the chopper transmission. For a Fermi chopper-slit package this depends on several factors which include geometrical attenuation, collimation, shadowing and imperfections in manufacture. These have been discussed in [8] and estimated to be:
where $E_i$ is expressed in eV.

The resultant intensities calculated at the sample position over the slowing down region are shown in Figure 4E. It should be noted that this continuous behaviour is again not achievable in practice for the chopper spectrometer, since only a finite number of slit packages each optimised to a given $E_i$ can be made.

c) Discussion

The general conclusion from Figure 4 is that the chopper spectrometer provides greater intensities towards large neutron energies, but that crystal monochromators can perform better towards lower energies. The cross-over energies are approximately 100 meV for a vertically focused Cu crystal array and 40 meV for a double PG monochromator system. It should be noted that these curves are only appropriate for the fixed incident energy resolution $\Delta E_i/E_i = 0.01$, and that the cross-over shifts towards lower $E_i$ values as the resolution is improved.

4. PRACTICALITIES

a) Number of Incident Neutron Energies

In a direct geometry spectrometer with a large angular array detector the main limitation to the $(Q,\epsilon)$ range which can be accessed is determined by the number of incident energy ($E_i$) settings available. In the case of the chopper spectrometer this is only a relatively minor restriction, since a set of rotors can, in principle, be phased with the source to provide a continuously variable $E_i$. For a fixed take-off angle crystal spectrometer a given combination of crystals or crystal planes can only provide a finite number of $E_i$ values.

Equation (12) provides a first indication of the number of $E_i$ values required to give reasonable resolution and $(Q,\epsilon)$ space coverage. If the maximum incident energy needed is $E_M$, then this condition can be met by producing other monochromatic beams with energies $E_M/2$, $E_M/4$, $E_M/8$, etc. This criterion can be satisfied by both types of spectrometer, and provides broad $(Q,\epsilon)$ range coverage with fractional energy transfer resolutions better than approximately 5% (see Figure 2).

The current trends in the scientific applications of these spectrometers are now directed towards the measurement of absolute cross-sections and line shapes, and in order to minimise corrections to measured data it is widely accepted that

$$T(E_i) = 0.72 - 0.6E_i$$  \hspace{1cm} (20)
TABLE I. \((\varepsilon/E_1)\) REGIONS ACCESSIBLE WITHOUT ORDER CONTAMINATION IN A CRYSTAL SPECTROMETER WHERE \((L_1/L_2) = 2.5\)

<table>
<thead>
<tr>
<th>Reflection Order (n)</th>
<th>ELASTIC SCATTERING FROM ((n - 1))th ORDER</th>
<th>UP-SCATTERING FROM ((n - 1))th ORDER</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\lambda_2/\lambda_1)</td>
<td>4.5  2.75  2.17  1.88  1.70</td>
<td>2.5  1.25  0.83</td>
</tr>
<tr>
<td>(\varepsilon/E_1)</td>
<td>0.95  0.87  0.79  0.72  0.65</td>
<td>0.84  0.36 ---</td>
</tr>
</tbody>
</table>

resolutions ~ 1% are now needed even for broad scattering distributions. Figures 2 and 3 show that this can only be achieved at the expense of \((Q,\varepsilon)\) range, and also that fractional energy losses \(\varepsilon/E_1 \sim 0.90\) are necessary. One consequence of this is that it requires the incident energies \(E_1\) to be continuously changeable in steps ~ 10%; this is in principle achievable up to incident energies \(E_1 \sim 500\) meV only in the case of the chopper spectrometer. Continuous changes in \(E_1\) are also possible with a double monochromator system, and we have shown that they give high intensities at low energies.

b) Higher Order Reflections

A further practical issue for the crystal spectrometer is that the monochromator produces, in addition to a beam of wavelength \(\lambda_1\), order contamination at \(\lambda/2, \lambda/3\) ... etc. This has the possible advantage of simultaneously producing several utilisable incident energies, but has the disadvantage that inelastic spectra from the different orders may overlap. When using \(n\)th order reflection (with an incident wavelength \(\lambda_1\) and a scattered wavelength \(\lambda_2\)) and restricting the discussion to downscattering only (low sample temperature) then the condition for no contamination from elastic scattering from the \((n - 1)\)th reflection is

\[
\frac{\lambda_2}{\lambda_1} \leq \frac{(L_1/L_2) + n}{(n + 1)}
\]  

(21)
and this restricts the observable $\epsilon/E_1$ range to:

$$\frac{\epsilon}{E_1} < 1 - \left( \frac{n - 1}{(L_1/L_2) + n} \right)^2$$

(22)

Table I gives these ranges for $L_1/L_2 = 2.5$, showing that even at 5th order there is no significant order overlap in the useful $(Q,\epsilon)$ range defined by equation (12). We emphasise however that much of this $\epsilon/E_1$ range is only accessed at relatively modest $\Delta \epsilon/\epsilon$ resolutions, up to 5%, and this resolution worsens with decreasing $\epsilon/E_1$ range i.e. with the order of the reflection.

If up-scattering from the $(n-1)$ order is also allowed (finite sample temperatures) then this overlap problem becomes more severe. No contamination occurs for:

$$\frac{\lambda_2}{\lambda_1} < \frac{L_1/L_2}{n - 1}$$

(23)

and

$$\frac{\epsilon}{E_1} < 1 - \left( \frac{n - 1}{L_1/L_2} \right)^2$$

(24)

Table I also gives calculated values of these quantities for $L_1/L_2 = 2.5$. We conclude that, in this more general scattering case, order contamination is problematic for reflection orders greater than 2, even when low resolution large $(Q,\epsilon)$ range experiments are being attempted. High resolution (1%) experiments using second order monochromated neutrons will not in general be possible in this $L_1/L_2$ geometry.

To summarise, we consider on balance that higher order reflections from a crystal monochromator are undesirable, particularly since this simple treatment has neglected the possibility of down-scattering from all orders higher than $n$. Higher order reflections are unlikely in general to produce good quality data, and may cause unwanted backgrounds.

c) Instrumental Factors

In deciding the best choice of monochromator for a direct geometry instrument it is also necessary to consider several more practical factors which also influence its performance. Table II summarises the advantages and disadvantages of the two spectrometers. Space does not permit these points to be discussed in detail, however references are made to some of the existing literature on this subject.
<table>
<thead>
<tr>
<th>Instrument</th>
<th>Advantages</th>
<th>Disadvantages</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. Crystal Spectrometer</td>
<td>1. Sample out of main beam gives lower backgrounds.</td>
<td>1. Restricted number of incident energies $E_i$ in a fixed geometry spectrometer.</td>
</tr>
<tr>
<td></td>
<td>2. The spectrometer can be designed to have no high speed moving parts.</td>
<td>2. Simultaneous reflections reduce the effective monochromator reflectivities [5].</td>
</tr>
<tr>
<td></td>
<td>3. There is scope for improving intensities by a) Using focusing techniques [6] and b) cooling the crystals [9].</td>
<td>3. Higher order reflections are always present and may give problems.</td>
</tr>
<tr>
<td></td>
<td>4. Crystal monochromators require little maintenance.</td>
<td>4. The use of crystals in incident beams is relatively new to pulsed sources. [10]</td>
</tr>
<tr>
<td>B. Chopper Spectrometer</td>
<td>1. The incident energy $E_i$ is in principle continuously variable, though with some penalty in transmission.</td>
<td>1. The sample is in the direct beam; background suppression requires using additional choppers [12].</td>
</tr>
<tr>
<td></td>
<td>2. This is a well-established technique on reactor and pulsed sources. [11]</td>
<td>2. Mechanically and electronically complex since the chopper needs to be phased precisely with the pulsed source which is not always absolutely periodic. [11]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3. Rotors often require a considerable amount of maintenance.</td>
</tr>
</tbody>
</table>
5. CONCLUSION

We are led to the conclusion that two different monochromating devices are required ideally to provide incident energies in the range 20 meV to 500 meV. A vertically focussed, pyrolytic graphite double monochromator is the obvious choice at low energies and a phased mechanical chopper is favoured at high energies. The secondary spectrometer is however a large and very expensive array of detectors covering the maximum solid angle and only one can be built on SNS. We therefore propose that such a secondary spectrometer is shared between the two monochromating devices on the one beam hole. This scheme, which involves periodic raising and lowering of the secondary chamber by inbuilt jacks, is illustrated in Figure 5.

REFERENCES


POSSIBILITIES AND RESTRICTIONS OF INELASTIC MAGNETIC NEUTRON SCATTERING EXPERIMENTS BEYOND THERMAL ENERGIES

M. LOEWENHAUPT
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

Hot neutron sources, especially spallation neutron sources, offer high-energy neutrons (eV) which can be used to do neutron scattering far beyond thermal energies. In this paper, possible and some already performed, experiments in this 'high-energy region' are discussed. The topic is restricted to magnetic excitations of systems with 4f and 5f magnetic moments and the merits and the limits of the methods are shown.

The properties of thermal neutrons (energy 26 meV, wavelength 1.8 Å) are ideally suited for studying the crystallographic and magnetic structure and the elementary excitations (phonons, magnons) of condensed matter. Cold neutron sources and spin-echo techniques, on the one hand, have extended the neutron scattering experiments into the low-energy, high-resolution (µeV, neV) region, while, on the other hand, hot neutron sources, especially spallation neutron sources, offer high-energy neutrons (eV) which can be used to do neutron scattering far beyond thermal energies. This paper deals briefly with possible experiments in this "high-energy region" but is restricted to magnetic excitations of systems with 4f and 5f magnetic moments. Because of the finite spatial extent of the wave functions of the "magnetic electrons" the envelope of all magnetic scattering intensity falls off with increasing Q, such as

\[ f^2(0) = e^{-aQ^2} \]

with \( a \) typically between 0.04 and 0.08 (Fig. 1). Thus, the main magnetic scattering intensity is confined to

\[ 0 < 4 \text{Å}^{-1} \].

If large-energy transfers are desired, the kinematics of the scattering process require small scattering angles and rather large incident neutron energies of several times the desired energy transfer (see Fig. 2 and the tabulated data in Fig. 1). These diagrams reveal two rather unfavourable aspects:

(a) While for lower incoming energies (\( E_0 < 100 \text{ meV} \)) the energy transfer in a magnetic scattering experiment can be nearly as large as \( E_0 \), this is not the case for higher \( E_0 \). Maximum attainable energy transfers (at \( \Theta = 0^\circ \)) are only 33% (\( E_0 = 1 \text{ eV} \)), 11% (\( E_0 = 10 \text{ eV} \)) and 3.6% (\( E_0 = 100 \text{ eV} \)) of...
FIG. 1. Typical Q-dependence of the square of the magnetic form factor for 3d, 4f and 5f moments (and hence of the magnetic scattering intensity). The table below gives the scattering angles $\theta$ for different sets of $E_o$ and $\hbar \omega$ at momentum transfers of 2, 4 and 6 Å$^{-1}$, respectively.

<table>
<thead>
<tr>
<th>$E$ (eV)</th>
<th>$\hbar \omega$ (eV)</th>
<th>$\theta$ (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.10</td>
<td>0.03</td>
<td>14.9</td>
</tr>
<tr>
<td>1</td>
<td>0.15</td>
<td>2.8</td>
</tr>
<tr>
<td>10</td>
<td>0.5</td>
<td>0.8</td>
</tr>
<tr>
<td>100</td>
<td>1</td>
<td>0.4</td>
</tr>
<tr>
<td>100</td>
<td>2</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.4</td>
</tr>
</tbody>
</table>

the incident energies (taking, e.g., Q = 4 Å$^{-1}$ as desirable momentum transfer). The necessary incoming energies are proportional to the square of the maximum energy transfer divided by the desired momentum transfer.

(b) The scattering angles become drastically smaller for increasing energy transfers; the step from a few degrees ($\hbar \omega = 0.5$ eV) to only a few tenths of a degree ($\hbar \omega = 2$ eV) will of course have severe consequences for the design of any such instrument.
To illustrate the rapidly increasing difficulty in performing high-quality high $\tilde{H}_\omega$ measurements at low $Q$ we give a general expression for the counting rate in the detector

$$\tilde{C} \cdot \tilde{S} \cdot f_0 \cdot d \Omega_0 \cdot dE_0 \left[ \frac{N}{F_0} S(Q, \omega) \right] \cdot d \Omega_1 \cdot dE_1 \cdot f_1$$

where the different factors characterize:

(a) The source where $\tilde{S}$ is the flux per solid angle and energy interval; in the slowing down region this is proportional to $1/E$. 

FIG. 2. Diagrams of the accessible $Q$- and $\omega$-regions for different incoming neutron energies $E_0$ in the low $Q$ range of interest for magnetic neutron scattering. The slope of the $\theta = 0^\circ$ line at $(0,0)$ is given by $\omega/\Omega \approx 2k_o^{-1}$. It determines the necessary incoming energy $E_{min} \sim (\tilde{H}_\omega_{ex}/Q_{ex})^{-1}$ if one wants to measure an excitation with energy $\tilde{H}_\omega_{ex}$ at momentum transfer $Q_{ex}^{-1}$. 

(b) The incoming and outgoing collimation which is chosen to be about equal and is mainly given by the perpendicular components of the momenta; these in turn determine the Q-resolution (because of the small scattering angles we have to assume a point geometry instead of the usually used slit geometry where the collimation perpendicular to the scattering plane may be more relaxed than in the plane).

\[ d\Omega_0 = d\Omega_1 = \left( \frac{k_j}{k} \right)^2 \left( \frac{d\Omega}{k} \right)^2 \]  \hspace{1cm} (2)

(c) The incident and final energy resolution \( dE_0 \) and \( dE_1 \) which again should be equal and of the order of \( dE \approx 0.1 \text{ eV} \) or better (this requires \( dE/E < 10^{-3} \) for \( E_0 = 100 \text{ eV} \)...).

(d) Attenuation of the beam before and after the scattering; the factor \( f_0 \) may not depend on the neutron energy; \( f_1 \) contains among others the counter efficiency \( \epsilon \).

(e) The scattering law \( S(Q,\omega) \) and the number \( N \) of scatterers in the beam; for the case of high energies we have \( k_1 \approx k_0 \) and the expression in the brackets independent of the neutron energy.

Counting all energy or momentum dependent factors together we obtain (with \( E_0 = E_1 = E \sim k^2 \) and \( E \sim (\omega/Q)^2 \) as mentioned before).

\[ \dot{\zeta} \sim (E^{-3} \text{ or } k^{-6}) \text{ or } (\frac{Q}{\omega})^6 \text{ or } \epsilon \text{ or } (dE)^2 \text{ or } (d\Omega)^4 \]  \hspace{1cm} (3)

This strong dependence of the counting rate on the neutron energy means that any significant increase in energy transfer at low \( Q \), or decrease of \( Q \) at high \( \omega \) will immediately create a severe intensity problem, unless relaxed \( Q \) and \( \omega \) resolutions may be tolerated.

From the experience with experiments on existing spallation sources I estimate an upper limit of 0.5 to 1 eV of energy transfers at momentum transfers around 2 to 5 Å\(^{-1}\) as achievable in inelastic neutron scattering using advanced spallation sources.

In the following I discuss very briefly some possible and some already performed experiments which will take advantage of this increase of the energy window to higher energies by more than one order of magnitude as compared to conventional neutron scattering. Figure 3 shows schematically the excitations of 4f and 5f magnetic moments in solids up to about 1 eV. Typical crystal field splittings and magnon dispersions of stable 4f systems usually fall in the conventional energy range of thermal and cold neutron scattering. There is, however, an increasing number of 4f compounds with larger splittings which require higher incident neutron energies. The first results were obtained from experiments on the chopper instruments LRCS and HRCS at the Intense Pulsed Neutron Source, Argonne, USA. They include the determination of the crystal field splitting in CeB\(_6\) /1/, PrO\(_2\) /2/, PrBaO\(_3\) and UO\(_2\) /3/ and of the spin-orbit splitting in PrAl\(_2\) /4/. In the intermetallic 5f compounds USn\(_3\) and UA1\(_2\) the broad quasi-elastic line (\( \Gamma/2 = 4 \text{ meV} \) and 35 meV for USn\(_3\) and UA1\(_2\), respectively) could be followed up to energy.
The regions of interest in 'high-energy spectroscopy with neutrons' are indicated by question marks and discussed in the text.

transfers of 10 times the line width /4/. The data obtained so far extend the observation of magnetic scattering to energy transfers of up to 260 meV. Future work in this field will, it is hoped, give answers to the question marks in Fig. 3 and help us to understand the magnetic behaviour of condensed matter containing stable and unstable 4f and 5f magnetic atoms.

REFERENCES

POSTER PRESENTATIONS

INSTRUMENTS (MISCELLANEOUS)

(Session X.c)
Chairmen

T. SPRINGER
W. SCHMATZ

Federal Republic of Germany
Abstract

\( \Delta T \)-WINDOW SPECTROMETER.

The paper describes a spectrometer based on a window filter, the neutron energy window being produced by a pair of beryllium blocks maintained at different temperatures, thus obtaining a window width variable between 15 and 40 \( \mu \)eV at about 5.2 meV. The spectrometer covers a wave-vector transfer from 0.8 to 3.0 \( \AA^{-1} \) and gives an energy transfer coverage of -1.5 to +3 meV. Natural width of Bragg cut-off for Be is first measured for different temperatures. Feasibility of producing a window is then demonstrated. Bragg steps in transmitted neutrons from Be are also measured for (1\( \bar{0} \)T1) and (0002) reflection and the possibility of producing additional windows is mentioned. Considerations for optimizing intensity consistent with resolution are discussed and the relevant features of the spectrometer to be installed at SNS at Rutherford Appleton Laboratory, United Kingdom, are described. Expected intensity is calculated.

1. INTRODUCTION

With the growth of neutron scattering and a need for superior resolution it was realised \( /1/ \) that it is possible to narrow down the width of the usual window of a window filter \( /2/ \) by two orders of magnitude by using two beryllium blocks at different temperatures as a filter/-reflector pair, and a spectrometer based on this was proposed \( /3/ \). In this paper the details of such a spectrometer designed and built at BARC, Trombay, for installation at the spallation neutron source (SNS) at the Rutherford Appleton Laboratory, United Kingdom, are described. First, the principle of the \( \Delta T \)-window spectrometer is described (Section 2) and then a set of measurements performed to ascertain some of the design parameters required for the spectrometer (Section 3) is given. The factors involved in the optimization of the spectrometer design with respect to resolution and intensity are discussed in Section 4. The shape of the resolution function and the intensity anticipated in a typical elastic scattering experiment is evaluated in Section 5. Finally, a summary is presented.

2. PRINCIPLE

Figure 1 is a schematic diagram of a \( \Delta T \)-window analyser. It consists of two blocks of beryllium, Be(I) and Be(II), kept at
temperatures $T_1$ and $T_2$, and a suitable annular detector. Neutrons scattered from the sample are first transmitted by a 15 cm long Be(I) which produces a sharp-stepped transmitted beam, allowing neutrons of energy $E < E_1$ to pass through. Out of these, neutrons with energies $E_1 < E < E_{II}$, are reflected in a nearly backward direction and detected in the detector, thus allowing a narrow energy window of width $\Delta = E_1 - E_{II}$ ($E_{II}$ actually corresponds to a reflection at an angle slightly less than $\pi$). By varying $T_2$ from 300 to 500K it is possible to obtain a window varying in width from 15 to 40 $\mu$eV with average energy, $E$, of the window changing from 5229.5 to 5217.5 $\mu$eV.

It is also possible to obtain additional windows with a reduced length of Be(I). Beryllium has three closely spaced Bragg reflections with indices (1010), (0002) and (1011), with spacings corresponding to Bragg cut-off energies 5235, 6399 and 6854 $\mu$eV. If one uses about 5 cm long Be(I) (instead of 15 cm), more than 95% of neutrons below 5235 $\mu$eV pass through, but 30% of neutrons between 5235 and 6854 $\mu$eV are also transmitted. Above 6854 $\mu$eV the
transmission is still very small. Be(II) placed in this beam will reflect neutrons back on to the annular detector. Depending on the angular coverage of the detector, three different windows will be produced near the three Bragg cut-offs.

3. OBSERVATIONS

A single crystal spectrometer with variable high incident collimation and a Ge(220) analyser was used to measure (1010) cut-off for 5, 10 and 15 cm long pieces of Be placed in the incident reactor beam, and also for blocks of Be with three different metallurgical treatments, at 300K and 116K with an energy resolution of 5 μeV /4/. A typical observation is shown in Fig. 2. The width of the step was found to be nearly the same in all the experiments mentioned above, giving an average step width around 15 μeV which is much larger than the resolution. The shift is consistent with the reported thermal expansion of Be. The figure also shows that it is possible to build a window detector with resolution down to about 15 μeV.

To further ensure the feasibility of the method the single crystal spectrometer was modified (see Fig. 3). A shielded annular detector was placed in the beam reflected by the crystal, germanium being now replaced by Cu(111). The detector receives neutrons back-scattered from a second Be bloc, thus creating the required window.
FIG. 3. Energy analysis of the ΔT-window, using the spectrometer shown, at 300 and 515K.

FIG. 4. Bragg edges for (10\bar{1}0), (0002) and (10\bar{1}1).
The window was then measured by making a $\theta$--$2\theta$ scan with Be(II) and annular detector moving as a composite unit through $2\theta$. The estimated resolution of the incident spectrometer was about 25 $\mu$eV. The measured window is shown in Fig. 3, clearly demonstrating the $\Delta T$-window and also showing that the intensity nearly doubles by increasing the temperature from 300 to 515K. The width of the peak shown here is determined mainly by the incident resolution and geometry of the spectrometer and not by the width of the window. The shift is consistent with the geometrical and resolution requirements.

Measurements were also made for the steps of (10\overline{1}0), (00\overline{0}2) and (101\overline{1}) reflections with a somewhat poorer incident collimation (Fig. 4). The energy difference between the steps being 1164 and 455 $\mu$eV, respectively, it should be possible to utilize them for quasi-elastic scattering measurements if the quasi-elastic width is not too large (say less than 150 to 200 $\mu$eV (FWHM).

The measurements of this section establish the feasibility of the $\Delta T$-window analyser.

4. DESIGN OPTIMIZATION

The spectrometer is designed for installation at the end of a guide tube facing a liquid hydrogen source at SNS, which gives 50 pulses per second. Incident pulse duration, $\Delta t(E_0)$, and the neutron intensity, $I(E_0)$ for neutron energy $E_0 = 5229$ $\mu$eV are 98 $\mu$s and $1.84 \times 10^9$ n-$\mu$eV$^{-1}$s$^{-1}$ per sterad, respectively /5/. Neutrons from the moderator (100 mm x 100 mm) will be guided on to the sample (42 mm x 42 mm) at a distance of 31m (= L1) giving an incident resolution of 31 $\mu$eV. The neutrons scattered from the sample will then be analysed by the $\Delta T$-window analyser.

The analyser is designed /6/ to obtain the maximum possible intensity consistent with the incident resolution for a situation when Be(I) is at 100K and Be(II) is between 300 and 500K. The minimum thickness required for Be(I) is 15 cm. The 40 mm deep BF$_3$ detector contributes 13 $\mu$eV to resolution and gives 59% efficiency. The backscattered reflectivity from Be(II) is given by

$$R = R_1 + R_3 = \left[ \frac{\mu_s}{2\mu} + \left( \frac{\mu_s}{2\mu} \right)^3 \right] (1-e^{-\mu D_2})$$

where $D_2$ is the thickness and $\mu_s$ and $\mu$ are macroscopic cross-sections for coherent elastic scattering and for total attenuation. It is the sum of reflectivities for once ($R_1$) and thrice ($R_3$) scattered neutrons. The chosen thickness of 50 mm gives reflectivity varying from 30 to 17.5% as temperature is varied from 300 to 500K. Beyond this thickness the rate of increase of reflectivity tapers off and hence it is not economical to choose a larger thickness.

It is useful to define a figure of merit, $F$, for the analyser (see Fig. 1):

$$F = (1/4 \pi) \left( \pi r^2/\xi^2 \right) (1-P).$$
FIG. 5. Figure of merit for ΔΤ-window analyser.

TABLE I. PARAMETERS OF THE ΔΤ-WINDOW SPECTROMETER
(see Fig. 1)

<table>
<thead>
<tr>
<th>Beryllium (I): r = 25 mm; D₁ = 150 mm; T₁ = 100K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transmission at T₁: Tᵣ = 0.92 for 5219 &lt; E &lt; 5236 μeV</td>
</tr>
<tr>
<td>0.15 for 5236 &lt; E &lt; 6390 μeV</td>
</tr>
<tr>
<td>0.001 for E &gt; 6390 μeV</td>
</tr>
<tr>
<td>Beryllium (II): R = 80 mm (arm I), 100 mm (arm II); D₂ = 50 mm</td>
</tr>
<tr>
<td>T₂ variable; Reflectivity of Be(II) = 0.30 (at 300K) &amp; 0.175 (at 500K)</td>
</tr>
<tr>
<td>Detector: D₃ = 40 mm; r₁ = 35 mm; r₂ = 100 mm; n = 0.59 at 5229 μeV</td>
</tr>
<tr>
<td>Distances: l = 490 to 690 mm; L = 350 to 700 mm</td>
</tr>
<tr>
<td>E = 5229.5 μeV at T₂ = 300K &amp; 5217.5 μeV at T₂ = 500K</td>
</tr>
<tr>
<td>Energy range: -1450 to +3000 μeV</td>
</tr>
<tr>
<td>Wave-vector range: 0.8 to 3.0 Å⁻¹; (Δθ/θ) = 7.5% to 1%</td>
</tr>
</tbody>
</table>
$P = \left[ \frac{r_1}{\ell \tan (\pi - \theta)} \right]^2$ gives the fraction of nearly backscattered neutrons not received by the detector because of its central hole. Given the geometrical constraints, one finds $\ell = 630 \ \text{mm}$; for this condition the figure of merit is given by Fig. 5. Based on this, $R$ is chosen to be $80 \ \text{mm}$, which leads to $L = 693 \ \text{mm}$ and $r_2 = 100 \ \text{mm}$. Table I gives the physical design parameters, the $\theta$ and $E$ ranges, and the $\theta$ resolution.

To keep the figure of merit the same as a function of temperature of Be(II), $L$ has been made continuously variable between 350 and 700 mm. Provision has also been made to vary $\ell$ between 490 and 690 mm; this allows an improvement in the figure of merit by as much as 65% without any change in resolution.

A photograph of the spectrometer with its shields removed is shown in Fig. 6. It has been provided with two arms, one covering an angle $+30$ to $+150$ and the other $-30$ to $-90$.

5. INTENSITY AND RESOLUTION

The intensity observed in a given time channel is given by a lengthy expression involving five variables. This expression has been evaluated assuming an instantaneous incident pulse and
The paper reports the first measurements of the natural width of the Bragg cut-off of Re. It demonstrates the feasibility of the
A ∆T-window spectrometer should be able to give a resolution of 40 µeV at 5230 µeV and allow a momentum transfer range up to 3.0 Å⁻¹ with an energy range of about 4500 µeV. If it is possible to use the additional windows as described in the text it would be possible to increase the ∆T-range as well as the throughput at some cost to the energy range. Further, if one is interested in an even more limited energy range for quasi-elastic scattering experiments, it is possible to increase the resolution substantially by using a matching double crystal monochromator on the incident side, because then the time-of-flight contributions to the analyser resolution from Be(II) and detector are not present. The calculated intensities suggest the viability of the spectrometer for use at the SNS.

REFERENCES

EXPERIENCE OF THE FOURIER TIME-OF-FLIGHT (TOF) NEUTRON TECHNIQUES FOR HIGH-RESOLUTION NEUTRON DIFRACTOMETRY

Technical Research Centre of Finland, Reactor Laboratory, Espoo, Finland

Abstract

EXPERIENCE OF THE FOURIER TIME-OF-FLIGHT (TOF) NEUTRON TECHNIQUES FOR HIGH-RESOLUTION NEUTRON DIFRACTOMETRY.

Fourier TOF techniques for neutron powder diffraction is being developed as a joint Finnish-USSR effort referred to as the Mini-SFINKS project. Presented are the first test results as obtained with the Mini-SFINKS TOF diffractometer put together mainly from the Astacus diffractometer components transported from Otaniemi to Gatchina. Experience gained so far suggests that substantial improvement in neutron economy can be obtained with rather straightforward modifications.

INTRODUCTION

An extensive programme for providing the new high-flux reactor PIK, scheduled to start its operation in 1987, with research equipment is in progress at the Leningrad Nuclear Physics Institute. As a part of this programme a joint Finnish-USSR effort, the SFINKS-project, aiming at constructing a time-of-flight powder diffractometer with polarization analysis, has been started. To obtain practical experience about Fourier TOF diffractometry, the Astacus device [1] was transported and re-installed at a 19 m long curved neutron guide at the 16 MW VVR-M reactor in Gatchina near Leningrad. This paper reports the experience gained so far.

The principles and the accomplishment of the Fourier synthetization of the TOF spectra were described earlier [1 – 4]. The present hardware is based on a parallel binary correlator capable of processing high counting rates.

* Leningrad Nuclear Physics Institute, Leningrad District, USSR
A profile-refinement program based on the estimation procedures for correlated data described by Tili [5] is available for conventional low-pass-filter synthetized data and can be readily exploited for refinements. A computer program for the estimation scheme for a band-pass filter synthetized data [6] has not yet been completed.

GEOMETRICAL CONSIDERATIONS

In high-resolution powder TOF diffraction time focusing is a key feature. If the orientation angles of the disk-shaped chopper, sample and detector are properly chosen, focusing conditions can be fulfilled with high precision in the range of scattering angles $90^0 < 2\theta < 180^0$, provided that neutrons can fly freely from one component to the next. In practice the desired shape of the sample is a cylinder of a few millimetres in diameter, when requirements due to high pressure, for example, are considered. The neutrons are best conducted to a narrow sample with a neutron guide between the chopper and the sample. In this case, however, the focusing possibility of the chopper is lost and some further constraints have to be accepted in order to obtain the desired compromise between resolution and intensity. The situation can be studied with the aid of Fig. 1, representing the focal loci of detectors at two different sample-to-detector distances while the chopper-to-sample distance is kept constant at 6.445 m. For different directions of incident neutrons the whole picture must be rotated around the sample axes. Obviously at $2\theta = 180^0$ only second-order effects on the total time-of-flight are generated, owing to finite incident collimation. Once the desired resolution is fixed, then, for the incident collimation as provided by the neutron guide, the detector can be extended from $2\theta = 180^0$ down to some lower limit. For shortest wavelengths the largest contribution to the relative error of the lattice spacings, $\Delta d/d$, comes from the time window of the chopper. Therefore, a very large detector can be used without further deteriorating the resolution. For longest wavelengths, on the other hand, the time window contribution to $\Delta d/d$ is marginal, and the geometrical contribution becomes the dominant one. Resolution can be improved, if desired, either by reducing the size of the detector from the smaller scattering angle side,
or by introducing extra collimation in the incident beam. One should recognize, perhaps, that, in ordinary powder diffraction work, there are fewer peaks at long wavelengths and thus less need for high resolution. An extra collimator in the incident beam with slight off-adjustment might also give an option for filtering out the shorter wavelengths, which could significantly improve the signal-to-noise ratio in a Fourier TOF measurement of a restricted range of wavelengths.

EXPERIMENTAL

The time-of-flight patterns are recorded in 2048 channels with the channel width adjusted to 2 μs. An external time delay can be chosen in multiples of the channel width in order to shift the
FIG. 2. Profile refinement of $\alpha$-$\text{Al}_2\text{O}_3$: The measured pattern (top) and the error pattern (bottom). Data were collected in 64 hours.

Table I. Comparison of refined parameters.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Lattice parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$a$ [Å]</td>
<td>5.12968 (6)</td>
<td>5.13448 (5)</td>
<td>5.1340 (1)</td>
</tr>
<tr>
<td>$\alpha$ [°]</td>
<td>55.288 (1)</td>
<td>55.270 (1)</td>
<td>55.270 (1)</td>
</tr>
<tr>
<td><strong>Atomic positions</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$u$ (Al)</td>
<td>0.3523 (2)</td>
<td>0.3522 (1)</td>
<td>0.35222 (5)</td>
</tr>
<tr>
<td>$u$ (O)</td>
<td>0.5562 (2)</td>
<td>0.5562 (2)</td>
<td>0.55642 (5)</td>
</tr>
<tr>
<td><strong>Isotropic temp. parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$B$ (Al)</td>
<td>$\sim$ 0</td>
<td>0.19 (5)</td>
<td>Anisotropic parameters</td>
</tr>
<tr>
<td>$B$ (O)</td>
<td>$\sim$ 0</td>
<td>0.34 (3)</td>
<td></td>
</tr>
</tbody>
</table>
available 4096 µs time-of-flight interval to cover the desired part of the diffraction pattern. The sample can be positioned at a goniometer table at 6.445 m distance from the Fourier chopper. The chopper itself is provided with a mechanism for adjusting the angle of the chopper plane relative to the incident beam and is located at the exit of a 19 m long curved 10 mm x 70 mm neutron guide.

A 2 mm thick circular Li-glass scintillator, 190 mm in diameter, was used as the detector at a distance of 3.017 m from the sample at scattering angle 120°.

Tailoring and measuring the incident spectrum are among the most important tasks in TOF diffractometry. At present the need for improvements are seen in this very area. In the present configuration neutrons emerging from the exit of a curved neutron guide through the chopper fly freely to the sample. Owing to the wavelength-dependent angular divergence and the curvature of the guide, the spectrum of the incident neutrons at the sample varies from point to point. It is therefore essential that when measuring the incident spectrum an even sampling both over the sample cross section and the guide exit cross section is assured. In fact, the incident spectrum used in the refinement of α-Al₂O₃ data was summed up from 112 individual measurements. The sample was dried in hot atmosphere for removing adsorbed water and filled a container 50 x 50 x 10 mm³. The international standard reference sample of cylindrical shape was not yet available but will be measured later.

RESULTS

Figure 2 shows the measured pattern (top) and the error pattern (bottom) of α-Al₂O₃, as measured in 64 hours with the Minisfinks facility. The refined parameters are given and compared with published data in Table I. The Chi² value of the refinement was 4.0.

The exact lattice constant is not of particular interest here, since no calibration of the instrument had been made earlier and no special measures were taken to control the temperature of the sample or the equipment. The rhombohedral angle α is fully
determined from the mutual positions of the peaks and does not involve peak intensities. Thus, it should come out rather accurately. The deviation from published values may indicate differences in sample preparation or in the temperature and emphasizes the need for using a standard reference sample. The relative atomic positions depend also on the intensities of the peaks and are in good agreement with published values. Temperature parameters are most difficult to estimate. In the present refinement no values significantly different from zero could be obtained. Solving of this problem would benefit from careful absorption corrections omitted so far, from better statistics and from extending the range of lattice spacings to shorter spacings.

CONCLUSIONS AND FUTURE PLANS

Up till now the efforts in the implementation of the Fourier TOF techniques for neutron powder diffraction have been focused establishing an operational system at the Gatchina reactor mainly from the existing components. A major new development was to build the control and data system using SM-4 minicomputer. The experience so far has been most promising and has demonstrated both the high resolution and the large range of lattice spacings. However, from the neutron economy point of view, the present configuration is far from optimum. The first straightforward improvement is to increase the solid angle of the detector. This can be achieved by adapting a larger detector and installing it closer to sample at higher scattering angle. Calculations show that by a 400 x 400 mm² detector a 30-fold increase in the counting rates can be achieved. Secondly, it is advisable to install a straight neutron guide between the chopper and the sample for conducting the neutrons to a smaller sample. Depending on the wavelength this would increase the neutron economy by a factor of 3 to 6. The overall effect would be an advantage of factor 100 - 200 over the present counting rates. Owing to the guide the focusing possibility of the chopper is lost but, as the new detector is designed to be installed at a higher scattering angle, the expected change in the resolution is not significant. If very small Δd/d is needed, a proper Soller collimator may easily be installed at the end of the straight neutron guide.
REFERENCES

A CONSTANT-ENERGY CRYSTAL SPECTROMETER WITH A ONE-DIMENSIONAL MULTICOUNTER*

R. BORN, D. HOHLWEIN
Institut für Kristallographie,
Universität Tübingen,
Tübingen,
Federal Republic of Germany

Abstract

A CONSTANT-ENERGY CRYSTAL SPECTROMETER WITH A ONE-DIMENSIONAL MULTICOUNTER.

A preliminary version of a novel crystal spectrometer has been set up at the BER II reactor in Berlin (West). The instrument allows the registration of scattered neutrons with a chosen energy transfer in a large and continuous range of scattering angles simultaneously. This is achieved by flat analyser crystals which reflect the neutrons out of the horizontal plane into a one-dimensional position-sensitive detector. Resolution measurements are compared with calculations. There is the possibility of "gradient" focusing as for conventional triple axis machines. The first measurements of acoustical phonons are analysed. All possible scattering events in the horizontal plane can be recorded systematically for a given energy transfer. With the flat-cone technique upper layers in reciprocal space can also be analysed in the same way, giving a complete set of inelastic data.

1. INTRODUCTION

Crystal spectrometers in combination with a position-sensitive linear detector have already been described with flat analyser crystals /1/ (MARX-spectrometer) and with curved ones /2/. In both cases the reflected neutrons stay in the horizontal plane. Therefore, only a small angular range of scattered neutrons can be detected simultaneously.

The constant-energy multicounter spectrometer proposed some years ago /3/ is able to analyse the scattered neutrons coming from the sample for a large and continuous range of scattering angles simultaneously. This is achieved by a reflection out of the horizontal plane at the analyser crystals. We report on a preliminary set-up of such an instrument, the resolution and the measurements of acoustical phonons, followed by a conclusion.

* Work in collaboration with the Hahn-Meitner Institute, Berlin (West)
A preliminary version of the novel spectrometer has been put into operation by modifying the flat-cone diffractometer /4/ which is installed at the BER II reactor in Berlin (West).

The beam coming from the reactor is reflected at a graphite monochromator. The neutrons scattered by the sample are reflected out of the horizontal plane by the analyser crystals (Fig. 1). At the moment one flat composite analyser crystal (graphite) is used which is 75 mm high and 200 mm long covering 8 degrees of scattering angles. Ten such crystals would cover 80°, which is the range of the curved linear multidetector. The detector has 400 detection elements (steps of 0.2°) and is commercially available.

The distance between analyser crystals and multidetector is small. Therefore, only a small vertical translation mechanism for the multidetector (15 cm) had to be installed. The detector height is 10 cm, allowing the detection of a relatively large interval in energies (about 7 meV) without lifting the counter.

The geometry is shown schematically in Fig. 1. The azimuthal scattering angle $\phi_A$ changes slightly with $\phi$. With the polar angle $\mu$ between the horizontal plane and the scattered beam /3/ one gets

\[
\tan \phi_A = \tan \phi / \cos 2\theta_A \tag{1}
\]
\[
\tan \mu = \sin 2\theta_A / (\tan^2 \phi + \cos^2 2\theta_A)^{1/2} \tag{2}
\]
\[
\Delta k/k \approx 1 - (\sin^2 \theta_A \cos \phi + \cos^2 \theta_A / \cos \phi) \tag{3}
\]

**FIG. 1.** Spectrometer configuration in the horizontal and vertical plane. (C = collimator, M = monochromator, S = sample, A = analyser, D = multidetector).
For our arrangement \((2 \Theta_A = 42^\circ, \phi = 4^\circ)\) the azimuthal angle \(\Phi_{\text{Det}}\) is expanded by 0.08\(^\circ\) relatively to \(\phi\), and \(\Delta k/k\) is 0.19\%. Therefore, we have practically a constant energy-transfer for the range of 8\(^\circ\) covered by the analyser. In reciprocal space this corresponds to a circular scan concentric with the Ewald circle.

3. RESOLUTION

To calculate the resolution function we followed the procedure of Cooper and Nathans /5/ given for a conventional triple-axis instrument. They introduced a transmission function \(P(\Delta k_i, \Delta k_f, \gamma_1, \gamma_2, \delta_1, \delta_2)\), defining the probability that a neutron with deviations in lengths of wave vectors and directions relative to a scattering process, given by \(k_i\) and \(k_f\), will be detected. \(\gamma_1, \gamma_2\) are horizontal divergences and \(\delta_1, \delta_2\) vertical ones for incoming and scattered neutrons. Neglecting the vertical divergences for the moment we have only to change the transmission function for the analyser in their formula (6). In our case we have

\[
P(\Delta k_i, \gamma_2) = P_A \exp\left(-\frac{1}{2} \frac{\Delta k_f/k_f \tan \Theta_A}{\eta_A} \right)^2
\]

with the mosaic angle \(\eta_A\) and the Bragg angle \(\Theta_A\) of the analyser crystal. This function now is independent of \(\gamma_2\) because of Eq. (3). The consequence of this difference can easily be incorporated in their formulas for the resolution ellipsoids (the term \(a_6 = 0\)). The vertical divergence \(\delta_2\), which is important for the out-of plane reflection at the analyser, is taken into account by an effective enlargement of the mosaic angle \(\eta_A\):

\[
\eta'_A = (\eta_A^2 + \delta_2^2)^{1/2}
\]
FIG. 3. Transverse and longitudinal phonon 'ellipses' at a deviation of $\Delta \theta = 9^\circ$ from the Bragg reflection. $E_I$ is the incident, $E_F$ the analysed energy.

FIG. 4. Measured and calculated phonons without energy analysis for the silicon (111)-reflection, $\Delta \theta = 9^\circ$. 

REL INTENSITY

SCATTERING ANGLE $2\theta_s$ [DEGREE]
FIG. 5. Measured phonons for the silicon (111)-reflection with energy transfer $E_F = 4.6$ meV. The solid line shows a calculated longitudinal phonon with sound velocity of 9.14 km/s.

Resolution measurements have been done with a perfect silicon crystal using the (111)-reflection. This reflection appears at $2\theta_S = 44.6^\circ$ with $\lambda = 2.4$ Å.

Experimental data together with calculation as outlined above are shown in Fig. 2. There is a good agreement for collimations $\alpha_0 = 0.3^\circ$, $\alpha_1 = 0.6^\circ$, $\alpha_2 = \alpha_3 = 0.5^\circ$ and mosaic angles $\eta_M = 0.4^\circ$, $\eta_A = 0.8^\circ$. As with a conventional triple axis instrument there is a large anisotropy in the $(0,\omega)$-plane allowing the focusing of a phonon branch.

4. MEASUREMENTS OF ACOUSTICAL PHONONS

Working on a small reactor only relatively strong inelastic scattering events can be measured like acoustical phonons near the zone centre. This has been done at a perfect silicon crystal with a volume of about $4 \text{ cm}^3$.

At the wavelength of 2.4 Å the scattering surface for acoustical phonons in silicon is elliptical - Fig. 3. At the outer 'ellipse' all phonon vectors end with transverse polarization, which diffract simultaneously for a lattice point not far from the Bragg position. The projection of such an ellipse can be seen on a multicounter if no energy analysis is done. From the intensity distribution one can determine in favourable cases the sound velocities as has been demonstrated with a photographic detector /6/. More information can be obtained with an energy analysis. With our instrument (constant energy transfer) a circular cut is made through the scattering surfaces and only well-defined phonons can diffract simultaneously - Fig. 3.
An example for the measurement of acoustical phonons without analyser crystals is shown in Fig. 4. The excited phonons belong to the (111) lattice point which is turned away from the Bragg position by 9° (energy gain). A program has been written to calculate the intensity distribution. There is already a relatively good agreement with the measurements - Fig. 4.

The intensity distribution for an energy transfer of 4.6 meV is shown in Fig. 5. Now the number of the measured phonons is reduced. The outer peaks belong to transverse phonons; near the centre is a peak of a longitudinal phonon identified by a calculation (sound velocity 9.14 km/s).

5. CONCLUSIONS

Because of the small energy variation with scattering angle (Eq. (3)) all scattering events in the horizontal plane can be measured simultaneously for a given energy transfer ($2\Theta_A$) and a given sample orientation. By turning the sample crystal around a vertical axis all possible scattering processes in the horizontal plane can be recorded systematically for a given energy transfer. For example, in this way the elastic diffuse scattering of disordered crystals can be measured.

Varying the take-off-angle of the analyser crystals ($2\Theta_A$) - or the incident wavelength - all scattering events in the horizontal plane can be recorded.

In the horizontal plane we will have in general one reciprocal plane called 0. layer. Upper layers can be analysed in the same way at the flat-cone diffractometer. Then the detector moves out of the horizontal plane together with the analyser crystals.

The resolution of the instrument is similar to a conventional 3-axis instrument allowing gradient focusing of phonon branches. Measurements will not only be done in the main-symmetry directions. Therefore, all data should be included in model calculations.

At a pulsed source the same technique can be applied. For fixed analyser angles and a monochromatic beam a range of energy transfers can be measured by time-of-flight.

This investigation has been supported by the Bundesministerium für Forschung und Technologie (projekt number FKZ 03-B13A13-8).

REFERENCES

156.
DESIGN OF A NEUTRON LIQUID DIFFRACTOMETER WITH A POSITION-SENSITIVE DETECTOR

R. G. DELAPLANE
The Studsvik Science Research Laboratory, Nyköping

U. DAHLBORG
Department of Reactor Physics, The Royal Institute of Technology, Stockholm

M. SANDSTRÖM
Department of Inorganic Chemistry, The Royal Institute of Technology, Stockholm

K. SKÖLD
The Studsvik Science Research Laboratory, Nyköping, Sweden

Abstract

DESIGN OF A NEUTRON LIQUID DIFFRACTOMETER WITH A POSITION-SENSITIVE DETECTOR.

The design of a neutron diffractometer for structural Investigations of liquids and amorphous materials to be Installed at the R2 reactor at Studsvik, Sweden is described. The Berl1ner-MURR detector system is utilized which features a stationary array of 16 linear position-sensitive He counters which simultaneously collect diffracted neutron intensities for scattering angles from 2 to 140°. The sample position is variable and can be displaced up to 50 cm upstream in the primary beam from the centre of the detector circle for experiments which require high resolution at small scattering angles. The predicted instrumental resolution for typical operating conditions is discussed.

1. INTRODUCTION

Structural studies of liquids and amorphous substances using diffraction techniques require high statistical precision of the measured diffracted intensities, while resolution requirements are less demanding than in the case of powder diffractometry. These considerations make the use of a linear position-sensitive-detector (PSP) system particularly attractive as intensities diffracted from a large range of scattering angles can be measured simultaneously. Efficient collection rates are essential for the precise determination of partial structure factors in polyatomic systems where isotopic substitution techniques are used in which limited quantities of sample are available. Furthermore, simultaneous data
collection covering a wide scattering angle is necessary for experiments with samples under extreme conditions of temperature and pressure and for investigations of kinetic phenomena such as chemical reactions and phase transformations.

2. INSTRUMENT DESCRIPTION

The instrument is shielded from background radiation by a water-filled steel tank with an inner radius of 110 cm as shown in Fig. 1. A perspective side view in Fig. 2 gives a more detailed description. The monochromatic beam of neutrons is first defined by a Soller collimator to limit the horizontal divergence and is diffracted by the sample mounted on a movable table. A portion of the scattered neutrons passes through an oscillating collimator and is collected by an array of PSD counters. After processing, the diffraction pattern is displayed as a histogram of diffracted intensity as a function of the scattering angle 2θ.

The Berliner-MURR detector system with associated electronics is an amplification of the basic design used in a neutron powder diffractometer constructed by the Research Reactor Facility at the University of Missouri at Columbia, Missouri, USA /1/. The detector system comprises 16 Reuter-Stokes $^{3}$He counters mounted in four interlocking banks in order to simultaneously subtend scattering angles ranging from 2° to 140°. Each counter consists of a cylindrical stainless steel tube, 2.54 cm in diameter with an active length of about 61 cm. The counter gas is a mixture of 8 atm of $^{3}$He, and 4 atm of argon with 5% CO$_2$ to give a calculated average efficiency of 65% for $\lambda = 1.0$ Å. A thin nickel-chrome wire with
a resistance of 3200 Ω serves as the central anode. The position of each detected neutron along the counter length is determined by the charge division method /2/. The resulting analog-digital converter (ADC) output is processed via an interface with a PDP 11/24 computer system to give a histogram of the number of detected neutrons as a function of scattering angle. Corrections are applied for variations in detection efficiency, resolution and subtended solid angle as a function of the position along the axis for each detector.

An oscillating radial collimator consisting of radially spaced steel blades coated with gadolinium is mounted between the sample and the detectors to minimize background radiation, especially that due to Bragg scattering by vacuum windows on furnaces, cooling devices or other auxiliary equipment. An oscillatory motion of 5° about the sample axis prevents shadowing effects on the detector. The sample holder which can be rotated continuously about the ω axis is mounted on a movable table which can be displaced up to 50 cm upstream in the primary beam from the normal centred position. This permits the sample-detector distance to be varied from 100 to 150 cm at 2θ = 0° in order to increase the resolution at lower scattering angles.

3. DIFFRACTOMETER RESOLUTION

As summarized by Caglioti et al. /3/, for a two-axis powder diffractometer the resolution of a diffraction peak at a given value
FIG. 3. Calculated instrumental resolution as a function of $2\theta$. Horizontal beam collimation preceding the monochromator is $0.25^\circ$; collimation between monochromator and sample, $0.60^\circ$. Mosaic distribution of the monochromator crystal is $0.4^\circ$; take-off angle $2\theta_m$ is $90^\circ$. Dashed lines exclude effects of vertical divergence. Upper curves are for a sample width and height of 10 and 30 mm, respectively. The lower curve for a sample width of 5 mm. The solid curves denoted by integers give the total resolution for each detector vertically displaced by $n \times 2.54$ cm from the horizontal plane defined by a vector coincident with the primary beam and another perpendicular to the sample axis.

FIG. 4. Calculated resolution function expressed as $Q(\AA^{-1})$ for $\lambda = 1.0 \AA$. Effects of vertical divergence are not included. Experimental conditions are those given in Fig. 3. Lines 1a, 1b, 2a, 2b are for sample widths of 10 and 5 mm, respectively. Solid lines 1a and 2a are for a normal centred sample position; dashed lines 1b and 2b refer to a sample position displaced 50 cm upstream in the primary neutron beam.
of $\theta$ is approximately Gaussian and may be expressed as
$$A^2 = U \tan \theta + V \tan \theta + W$$
where $A$ is the instrumental full width at half maximum (FWHM) in degrees and $U, V, W$ are constants for a specific experimental arrangement, i.e. beam collimation, mosaic distribution and scattering angle ($\theta_m$) of the monochromator crystal. The contribution to the angular resolution due to the detectors at an average radius of 100 cm is about $\Delta \theta = 0.2^\circ$. In addition, instrumental broadening due to the curvature of the Laue diffraction cones becomes important at low angles. These effects depend on the vertical dimensions of both the sample and the detector which become critical for detectors displaced out of the horizontal scattering plane.

The diffraction peak profile as observed by each detector has been calculated as a function of the scattering angle $\theta$ for a variety of experimental arrangements. The total instrumental resolution is derived by convoluting the peak profiles with the above expression $A$. The resolution function was calculated for a model consisting of a horizontal array of five curved detectors each subtending an angle of 140° about a radius of 100 cm (Fig. 3). The poor resolution near $\theta = 0^\circ$ due to vertical divergence can be improved by masks of cadmium strips to decrease the effective vertical dimensions of the detectors in this region. The resolution function expressed in terms of $Q (\AA^{-1})$ for $\lambda = 1.0 \AA$ in Fig. 4 shows the improved resolution at low $\theta$ gained by moving the sample position 50 cm upstream in the primary neutron beam. For a PSD system the horizontal beam collimation between the sample and detector is determined by the effective widths of both. These resolution curves illustrate the advantages of using small samples if the diffracted intensity is sufficient for good counting statistics.

Future plans include the utilization of a monochromator crystal which can be vertically focused. Expected flux at the sample position is estimated to be $10^9 \text{n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ at $\lambda = 1.0 \AA$.

REFERENCES

Abstract

THE USE OF THE POLYCHROMATIC METHOD IN ELASTIC NEUTRON DIFFRACTION.

Generally, the polychromatic method used to test the quality of crystals (twins, polycrystals etc.) or to orientate single crystals. It is also possible to use it in a much more quantitative way - each time an appearance of the commensurate and/or incommensurate long-range order occurs, the polychromatic method seems to be the most appropriate means of disclosing it. This quality arises from the gain in the exposure time and the simplicity of the method. The panoramic view of the symmetry and of its evolution with temperature and pressure makes this method valuable for studying displacive phase transitions. The fundamental reflections are easily indexed directly, but the incommensurable reflections can only be indexed by the use of some tricks (rotating crystal in marked incident spectrum, gnomonic projection). By using computers it is easy to process measurements with the incident neutron beam deviated from the principal mirrors of the crystal sample. In this case, the amount of information is multiplied by a factor 2 owing to this unsymmetrical Laue pattern. Theoretically it is possible to estimate the structure factors for the unknown spots by comparison with those of the fundamental reflections, but the overlapping of the spots presents a serious limitation for an extensive measurement of intensities. The displacive phase transition of a-U at low temperature, and the results on single crystal growth of oxygen are examined. Preliminary attempts to extend the detection with television camera tubes are presented.

1. INTRODUCTION

Since the first experiments carried out by Wollan, Shull and Marney in Laue Photography of Neutron Diffraction /1/, the development of this technique has awaited an improvement of the detectors such as a scintillator-film system /2/ or an XY electronic detector /3/.

In 1979 Marmeggi showed the capabilities of the white incident beam for studying the set of propagation vectors of a modulated structure /4/. Here we first report some advantages of the polychromatic method used in the study of displacive phase transitions and single crystal growth; second, we explore the possibilities of adding a television camera tube for a rapid examination of the crystal orientation.
2. THE POLYCHROMATIC METHOD (P.M.)

The characteristics of the polychromatic method are a polychromatic incident beam \( n \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \cdot \AA^{-1} \) (see Fig. 1a), a fixed position of the sample \( S \) and a multidetector (photographic or electronic system—Fig. 1b). The elastic neutron diffraction obeys the Bragg law \( (2d \sin \theta = \lambda) \), with the interplanar spacing \( \lambda \), and the results are mainly the measure of the Bragg angle \( \theta \) and the recognition of the plane of the incident and diffracted beam. Another important result is the disclosure of the zone \( \{u,v,w\} \) for some \( (hkl) \) spots, leading to the expression:

\[ hu + kv + tw = 0 \]

which can be an important relation in identifying the incommensurable \( (hkl) \) reciprocal knots.

Two possibilities arise from the Laue pattern:

1. All the unknown spots come from the commensurate structure of which we know the crystallographic parameter. In this case, it can be easily shown that the coordinates of the gnomonic projection of the Laue pattern are linearly related to the ratio of crystallographic parameters and Miller indices. For example, for an incident beam parallel to the \( b \) axis of an orthorhombic structure \( (a,b,c) \), the \( (X,Y) \) co-ordinates of the gnomonic projection corresponding to the \( (hkl) \) planes are given by the expressions

\[ X = \left( \frac{h}{k} \right) a^* b^* \]

and

\[ Y = \left( \frac{l}{k} \right) c^* b^* \]

where \( a^*, b^*, c^* \) are reciprocal lattice parameters.

In the case of commensurate structure, one deduces without difficulty the values of \( k \) which permit one to obtain the set of the smallest \( hkl \) integers; this achieves the indexation (standard deviation on \( h,k \) and \( l \) is typically \( \sigma = 0.003 \)).

2. Some unknown spots \( (q_1, q_2, q_3) \) arise from an incommensurate new structure. The problem is then to know the wavelength \( \lambda \) responsible for such spots, because we do not know the \( a^*, b^*, c^* \) parameters of the new unit cell corresponding to this new long-range order.
One way to disclose the wavelength corresponding to such a spot is to follow its intensity when the crystal is rotating. If the incident spectrum is marked (by the lack of one known wavelength \( \lambda_0 \) due to a monochromatization of the incident spectrum (Fig. 1a), the intensity of the unknown spot must disappear for the setting corresponding to \( \lambda_0 \). On the other hand, the use of the gnomonic projection leads also to a disclosure of the zone of the unknown satellites, owing to the characteristic where the planes \( (q_a, x, y, z) \) of the zone are represented by aligned points on the gnomic projection. We then obtained the zone equation for these satellites \( uq_x + vq_y + wq_z = 0 \) and, for known values of \( uvw \), we get an important relationship between \( q_x \), \( q_y \) and \( q_z \) of the unknown spot.

3. EXAMPLE OF THE \( \alpha \)-U PHASE TRANSITION /5/

The \( \alpha \)-U presents an orthorhombic structure at \( T < 880K \) (Cmcm) /6/. At low temperature this crystal showed a second-order phase transition at 43K, but apparently the structure stayed with the same long-range order except for small changes in \( a, b \) and \( c \) parameters and little evolution of the \( y \) parameter of the unique atom position \( (A; 0, y, 1/4) \). Figure 2 shows the Laue pattern obtained at 70K and 6.5K for incoming neutrons parallel to \( (\bar{a}) \) axis.
Because in the P.M. we impose no constraints on the possible diffracted rays, one easily observes new spots which reveal a new long-range order below 43K. Using the gnomonic projection, it has been easy to indicate the different new satellites by points on Fig. 3 on the gnomonic projection of co-ordinates \((a*/b*\) and \(c*/b*)\).

On examining satellite Nos 7, 9, 13, 15 and 17, we find a zone with the axis \([011]\); it results in a relation \(q_xu + q_yv + q_zw = 0\) which in our case becomes simplified into \(0 + q_y + q_z = 0\) or \(|q_y| = |q_z|\) (we write the wave vector of the transition as \(\vec{q} = \pm q_x \hat{a}^* \pm q_y \hat{b}^* \pm q_z \hat{c}^*\)). In attempting an indexation with \(\lambda = 1.25\ \text{Å}\) (the most probable wavelength in the spectrum) we find that satellite 16 has \(\vec{q}\) components \((1.46, 0.78, 1.12)\), which are easily interpreted as particular values of the general form \((q_x; 1-q_y; 1+q_z)\); it is straightforward to deduce \(q_y = q_z = 0.18\) and \(q_x = 0.5\). With a twice exposed Laue photograph, at two different temperatures, one could find an evolution of the satellites with physical parameters (temperature, pressure) /5/ and conclude that \(q(6.5K) = \pm 0.5\hat{a}^* \pm 0.176\hat{b}^* \pm 0.181\hat{c}^*\) and \(q(36K) = \pm 0.5\hat{a}^* \pm 0.138\hat{b}^* \pm 0.207\hat{c}^*\) with a standard deviation of \(\sigma = 0.003\).
4. THE GROWTH OF OXYGEN SINGLE CRYSTAL

During the past few months, we were interested in growing a single crystal of oxygen to carry out neutron diffuse scattering experiments in $\alpha$, $\beta$ and $\gamma$ phases. A system was built up with an oxygen generator and a Dioplex cryogenerator, and Laue photographs were obtained for different programs of the temperature evolution. We could define precisely the temperature range of the $\gamma$-oxygen single-crystal stability and the process to obtain true crystallite powder by crossing the $\beta$-$\gamma$ transition temperature ($T_c = 44K$; $\Delta v/v = 15\%$).

5. FIRST RESULTS WITH TELEVISION CAMERA TUBE

An important goal of the P.M. could be to obtain a rapid examination of the crystal orientation. The average exposure time for Laue photography is between 3 min for normal reflections to 2h for small satellites. Even for a 3 min exposure time, the total duration of the photographic process (exposure and development) is about 20 min; it is too much to easily orientate single crystals. By adding an electronic camera we hope to get a Laue pattern between 15s and 3 min.

Two characteristics are important: (a) Owing to the low luminous flux given by the convector

\[
\left(^{1}n + ^{6}Li \rightarrow ^{4}He + ^{3}H + 4.8\ MeV\ ZnS \rightarrow 10^4\ photons\ of\ \lambda \sim 4400\ \AA;\right)

\text{few } 10^{-12}\ W\cdot\text{cm}^{-2},
\]  

the detector must be very sensitive and with a very low background level. (b) The low diffracted neutron flux needs the use of tubes which can integrate the signal in a few minutes. The "Secondary electron conduction tubes" (used in astronomical photometry) have a target with a very high resistivity that permits a long integration time (up to 30 min). Images, with 3 min exposure time, have been easily obtained on a television camera and we hope to decrease this duration with a refrigerated light intensifier. Single-crystal orientation could be carried out in few minutes by this technique.

6. CONCLUSION

The main features of the polychromatic method is the fixed position of the sample (no adjustment, no scanning) throughout the experimental time; it results in the signal being permanent throughout the experiment, producing a considerable increase in efficiency. Secondly, there are no constraints on the sample owing to the method leading to good detection of the real diffracted beam; one can say it is one of the best methods to place the diffracted spots and to appreciate the symmetry elements parallel to the incident neutron beam and their evolutions with physical parameters.
REFERENCES

/1/ WOLLAN, E.O., SHULL, C.G., MARNEY, M.C., Phys. Rev. 73 (1948) 527L.
NEW VELOCITY DRIVE FOR BACKSCATTERING SPECTROMETERS


*Institut für Festkörperforschung
**Technische Dienste, Mechanische Werkstätten
Kernforschungsanlage Jülich GmbH, Jülich, Federal Republic of Germany

Abstract

NEW VELOCITY DRIVE FOR BACKSCATTERING SPECTROMETERS.

A new concept is presented for the Doppler drive of a high resolution neutron backscattering spectrometer with a large flexibility concerning its velocity profiles. It is based on a high-precision hydraulic servo-cylinder system and - when equipped with Si-111 single crystals - allows energy transfers of up to 20 MeV.

1. FUNCTION OF THE DOPPLER DRIVE IN A NEUTRON BACKSCATTERING SPECTROMETER

The concept and the prototype of the neutron backscattering spectrometer has been developed at the Munich Research Reactor and is described in Ref. [1]. Shortly afterwards real backscattering spectrometers went into operation at Jülich's DIDO reactor [2] and at Grenoble's HFR reactor. Both spectrometers are installed at cold-neutron guides and, together with an about ten years younger thermal neutron backscattering spectrometer at Grenoble's HFR reactor, are the only backscattering spectrometers in the world up to now.

A backscattering spectrometer is a neutron crystal spectrometer with an exceptionally good energy resolution. This resolution is achieved by working with the largest possible Bragg angles at both the monochromator and analyser crystals (backscattering).

Figure 1 explains the mode of operation. Neutrons are produced in a research reactor; by means of a cold source (liquid H$_2$/D$_2$) [7] they are moderated to such energies that the maximum of their Maxwellian velocity distribution corresponds to wavelengths between 4 and 6 Å. In neutron guide tubes ((1) in Fig. 1) the neutrons are extracted and fly to the different instruments. They hit the monochromator (2) of the backscattering spectrometer which consists of Si-111 single crystals and reflects the neutrons with wavelengths of around $\lambda = 6.27083$ Å. The energy resolution is given by

$$\frac{\Delta E}{E} = 2 \cot \Theta \Theta + 2 \frac{\Delta \tau}{\tau}$$

where $\Theta$ is the Bragg angle, $\Theta \Theta$ contains the angular divergence of the neutron beam (and the mosaic spread of the crystal which,
However, vanishes for ideal Si crystals), $\tau = 2 \pi / d$, $d$ is the lattice plane spacing, and the term $\Delta \tau$ is a measure of the lack of sharpness of the lattice plane spacing and of the extinction of the crystal. Thus, a resolution of 0.3 keV FWHM can be achieved. The exact wavelength of the reflected neutrons depends on the instantaneous velocity of the monochromator crystal: the Doppler drive creates an energy spectrum in the same manner as a Mössbauer drive, and the energy spectrum spans $\pm 10 \mu$eV for mechanical drives. All reflected neutrons are directed on to the sample by the graphite deflecting crystal (3). In the sample (4) the neutrons are scattered and hit one of the spherical segments (5) plated with Si-111 analyser crystals. These neutrons which, in the scattering process by neutron energy loss or gain, have regained the wavelength 6.27083 Å, make a 180° reflection, fly through the sample a second time and reach their detector (6) where they are counted. According to their time of arrival, which corresponds to a certain start time at the monochromator, and thus to a certain Doppler shift of their incident energy, these neutron counts are stored in certain energy channels of the resulting neutron scattering spectrum.

In the two existing cold neutron (high resolution) backscattering spectrometers the Doppler drives consist of stripped-down petrol motors, whose crankshafts are rotated by means of
electromotors. The analyser crystals mounted on the pistons thus move back and forth with nearly sinusoidal velocity-time characteristics as shown in Fig. 2a. Each velocity corresponds to a certain energy shift of the backscattered neutrons; thus, a certain incident energy spectrum is created, which in the case of a Si-111 monochromator is distributed around \( E_0 = 2.0805 \text{ meV} \) with a time structure according to the floppier drive frequency. The disadvantages of such a crank drive are: (i) the velocity distribution is not uniform - high positive and negative Doppler velocities (and thus neutron energies) occur much more frequently in the spectrum than low ones (see Fig. 2a); (ii) the velocity-time characteristics are fixed for a mechanical drive and cannot be changed. Possible alternative drive systems are pneumatic, electric and hydraulic Doppler drives; in a prestudy /3/ the hydraulic system gave by far the best performance.

2. THE HYDRAULIC DOPPLER DRIVE

Our hydraulic Doppler drive consists of three main components: a servo-cylinder system, a compressor system (both from REXROTH GmbH, Fed. Rep. Germany), and an electronic control system. Its incorporation into the backscattering spectrometer is shown in Fig. 1.
2.1. The servo-cylinder system

The piston (dia. 32 mm) and the piston rod (dia. 28 mm) of the servo-cylinder (type ST) move on hydrostatic tapered bearings. This fractionally optimized system guarantees both a high position and phase accuracy. The maximum stroke of the cylinder is 200 mm. On top of it a 4-way servo-valve (type 4 WS 2 EE 10) is mounted. This valve is electrically driven (with a hydraulic amplifier) and allows a maximum velocity of 3 m/s for the piston of the servo-cylinder, corresponding to a neutron energy modulation of ± 20 µeV. In the event of future experimental requirements the valve can easily be replaced by a larger one (servo-cylinder and compressor system are laid out for a maximum velocity of 4.5 m/s). The servo-cylinder is equipped with an ultrasonic displacement transducer (accuracy ± 0.1 mm) which produces the feedback signal for the electronics regulating the control piston of the servo-valve (see Section 2.3). On the other hand, the servo-cylinder is equipped with an inductive velocity transducer; according to its signal in the dedicated MicroVAX II computer system of the backscattering spectrometer, the arriving neutron counts are set for certain velocity (energy) channels. An important requirement on the drive system was that no high-frequency vibrations be superimposed on the movement of the monochromator (even at the turning point) because a high-frequency modulation of the nominal Doppler velocity creates a corresponding modulation of the neutron energies and thus spoils the high resolution of the spectrometer. The absence of these high-frequency vibrations was verified in our prestudy /3/ by (i) simulation of the whole system on an analogous computer of the Institut für Hydraulik of the RWTH Aachen, and (ii) by test measurements on similar servo-cylinders.

The servo-cylinder is mounted on a cast-iron support together with two round guide bars on which the monochromator holder moves back and forth - see Fig. 1. The movement is absolutely free from play owing to special ball race bearings, also visible on the figure. The monochromator holder, which is driven by the piston of the servo-cylinder, consists of a honeycomb-like aluminium plate with a high bending rigidity and low neutron attenuation; the construction of the whole support is complex because of the requirement that our Doppler drive be transparent to all neutrons in the neutron guide with wavelengths other than our nominal wavelength of 6.27 Å - the backscattering spectrometer is not situated at the end position of the neutron guide and all the neutron spectrometers downstream must also be supplied with neutrons.

2.2. The compressor system

The main parts of the compressor system are a high-pressure circulation pump (feed performance 48 L/min; resulting oil pressure, 140 bar), a hydrostorage container with a gas pre-tension of 100 bar, which attenuates the pressure pulsation, an oil-cleaning and oil-temperature regulating system with the necessary pumps and heat exchangers, and an oil storage container with a volume of 250 L.
2.3. The electronic control system

The velocity of the Doppler drive is not regulated by velocity regulation, but by means of a position regulation of the servo-cylinder piston, i.e., the central cylinder of the servo-valve is coupled to the ultrasonic position transducer via a PID position controller. The rated position values at 1024 time increments within one half period of the piston motion are controlled by a microprocessor. This is periodically repeated during the 6 to 24h duration of a run for recording a neutron spectrum. According to the experimental requirements the microprocessor is programmed at the beginning of each run by means of the instrument computer which, for example, for a desired maximum energy transfer, calculates the rated positions at the 1024 time increments and automatically takes care of boundary conditions (maximum stroke, maximum velocity, etc.); if experimental requirements cannot be realized it creates error messages. By this type of task division between microprocessor and instrument computer, the computer is relieved from routine work.

3. PHYSICAL ASPECTS

The main application fields of a high-resolution backscattering spectrometer are slow diffusion (quasi-elastic neutron scattering - an example is shown in Fig. 3) and inelastic neutron scattering at very low energy transfers (e.g. rotational tunnelling - Fig. 4). In both fields the hydraulic Doppler drive has considerable advantages compared to its mechanical predecessor. This is illustrated in Fig. 2. In the velocity profile of a crank drive (Fig. 2a) high positive and negative velocities occur much more frequently than low ones, i.e. when a neutron scattering spectrum is thus recorded, most of the measuring time is spent at the upper and lower borders of the spectrum, whereas the information of interest is either in the centre of the spectrum (quasi-elastic neutron scattering) or at certain energy transfers (tunnelling peaks). With a hydraulic Doppler drive, however, the experimenter can largely spend his measuring time at the energy transfers he desires; thus, the counting statistics in the interesting parts of the spectra can be considerably increased. Standard operation modes of the new Doppler drive will be triangular velocity-time characteristics (every velocity occurs equally frequently (Fig. 2b) and scanning trapezoid velocity-time characteristics (Fig. 2c), which allows scanning mainly at selected regions of the spectrum, e.g. at Doppler velocities corresponding to \(-E_2 < \hbar \omega < -E_1\) and \(E_1 < \hbar \omega < E_2\), which would allow, for example, certain tunnelling peaks to be investigated in detail.

We are convinced that our Doppler drive concept will find many applications in future neutron scattering spectrometers, particularly in instruments designed for pulsed neutron sources where the main aim is to make somehow use of the high peak flux of the neutron pulses. One example is the bunching spectrometer /6/.
FIG. 3. Hydrogen diffusion in the hydrogen storage compound $\text{Ti}_{0.8}\text{Zr}_0.2\text{CrMnH}_2$ as observed by high-resolution quasi-elastic neutron scattering /4/.

FIG. 4. Rotational tunnelling of methyl groups in Lutidine as observed by high-resolution inelastic neutron scattering /5/.
REFERENCES

/2/ ALEFELD, B., Kerntechnik 14 (1972) 15.
/6/ RICHTER, D., ALEFELD, B., IAEA-CN-46/74P, these Proceedings.
Area Detector for the Small-Angle Neutron Scattering Facility at Risø

J. K. Kjems, R. Bauer*, B. Breiting, A. Thuesen
Risø National Laboratory, Roskilde, Denmark

Abstract

The 3He multiwire area-sensitive neutron detector system for the Risø small-angle neutron scattering facility is described. The active detector volume is 40 cm x 40 cm x 4.5 cm with a gas filling of 0.5 atm 3He and 2 atm argon with 5% CO2 giving 75% detection efficiency at 8 Å neutron wavelength. The position of a neutron event is determined via the rise-time difference for pulses arriving at either side of three multiwire planes. The anode plane is used for one co-ordinate and the two cathode planes, coupled in parallel, are used for the other. The overall position resolution is 9 mm in both co-ordinates and the maximum reliable count rate is 10 kHz.

1. Introduction

Area-sensitive detector systems are indispensable for small-angle neutron scattering (SANS) experiments in order to obtain sufficient intensity and reproducibility without loss of resolution. Several systems are available, including a few commercial systems and, for an overview of this topic, the reader is referred to the recent proceedings of an ILL* conference /1/. A 3He multiwire system, inspired by the Oak Ridge design /2/, was chosen for the Risø-SANS facility, partly owing to previous experience with linear position sensitive 3He detectors and partly because it appeared to be the least complicated for an initial attempt in this field. The size of the active area, 40 cm x 40 cm, was determined conservatively to avoid problems with the curvature in a single entrance-window design. It also appeared to match the dimensions of the SANS spectrometer which only allows up to 4 m distance between the sample and the detector /3/. The detector system has been proved to achieve all the design goals and it has now been operating satisfactorily since autumn 1982 without any interruptions.

* Present affiliation: Royal Veterinary and Agricultural University, Copenhagen, Denmark

1 ILL = Institut Max von Laue – Paul Langevin, Grenoble

489
2. MECHANICAL DESIGN

An "exploded" view of the detector is shown in Fig. 1. It has a circular outer shape, 720 mm diameter, with a stainless-steel backplate on which the three 40 cm x 40 cm fibreglass planes are mounted with a separation of 10 mm. The detector lid is made of aluminium with an entrance-window thickness of 8 mm over the active area. It is sealed with a Vithon O-ring. The overall detector thickness is 160 mm and the active section is 45 mm thick. There is a total of eight high-voltage feed-throughs and two holes for gas inlet and circulation. The preamplifiers are also mounted on the back of the detector together with the gas purification system. The detector is supported by the beam-stop unit, which seals against the spectrometer vacuum. The whole unit rests on air-cushions and can be moved manually to different scattering angles. The sample/detector distance is changed by removing segments of the vacuum vessel. The detector shielding is made from boronated polyester and it encloses the whole unit.

FIG. 1. Exploded view of the Risø SANS area detector with the support and the beam stop arrangement.
3. ELECTRICAL DESIGN

3.1. Anode and cathode planes

The detector works in the proportional mode and the neutrons are detected by the reaction

\[ ^1\text{H} + \frac{3}{2}\text{He} \rightarrow \frac{3}{2}\text{T} + \frac{1}{2}\text{p} + 764 \text{ keV}. \]

The central anode plane is made with 68 25-\(\mu\)m-diameter stainless steel wires (the two outermost have diameters of 60 \(\mu\)m and 100 \(\mu\)m) which are interconnected by standard 200 \(\Omega\) (± 1\%) resistors. The wire spacing is 6 mm. The two cathode planes are similar except that all the wires have a diameter of 60 \(\mu\)m. The anode operating voltage is 3.6 kV and the cathode is kept at a neutral potential of 1.7 kV, i.e. the electric field pattern is nearly the same as it would be without the cathodes mounted. This choice optimizes the electron drift towards the anode and it also minimized the high-voltage induced noise. The detector gas is 0.5 atm ^3He and 2 atm argon with 5% \(\text{CO}_2\), which gives 75% efficiency at a neutron wavelength of 8 Å.

At the usual working conditions the gas amplification is approximately 500 and the neutron pulse height distribution as shown in Fig. 2 has a width of 14% FWHM. To maintain this resolution it is necessary to circulate the detector gas through a small alumina filter mounted on the back of the detector. The circulation is driven by convection caused by a small heating coil.
3.2. Delay line electronics

The positioning of the neutron events is based on the delay line principle. The total resistance in each plane is approximately 13 kΩ giving an overall time constant of 2.4 μs. Each plane is connected to two charge-sensitive preamplifiers and the impact position is determined from the difference in rise-time for pulses arriving at these. The anode plane is used for one co-ordinate and the induced signals in the two cathode planes, coupled in parallel after the preamplifiers, are used for the other co-ordinate. The rise-time is determined by the zero-crossings of the doubly differentiated signals after the main amplifiers and a delay is used to ensure that the sequence is always the same. The difference in zero-crossing times is converted to a pulse-height in coincidence with a discriminator pulse selecting the neutron events from the sum of all the preamplifier signals. The pulses with amplitudes proportional to the impact co-ordinates are converted to addresses in a 64 x 64 array in a dual parameter multi-channel analyser system. The maximum reliable processing rate for neutron events is 10 kHz. The data can be displayed on-line either as contours or in isometric projection on a cathode screen, or they can be transferred to the SANS PDP-11 computer and displayed on a colour monitor.

Fig. 3. Relation between impact position and final channel assignment for the Risø SANS area detector system. The non-linearity is corrected for by using fits to a fifth-order polynomial (solid line). The inserts show the response to a pinhole beam giving the spatial resolution.
4. PERFORMANCE

The detector is calibrated by a measurement in a diffuse flux of neutrons with a perforated cadmium mask in front of the entrance window. The holes in the mask are 1 cm in diameter and are 2.4 cm apart. This test shows that there is no correlation in the determination of the two co-ordinates of a neutron impact. It also shows that the response is not completely linear owing to the increase in the grid capacitance near the edges. This effect is corrected for using a fifth-order polynomial which is determined from fit to the mask test data. Examples of such fits are shown in Fig. 3, which also shows the response to a pin-hole beam from which the spatial resolution, \( \sim 9 \text{ mm (FWHM)} \), is determined. The detection efficiency is uniform (± 5%) except very near the edges and the small local variations are normally corrected for using the spectra from incoherent scatterers such as water, vanadium or perspex. At shorter wavelengths (\( \lambda < 5 \text{ Å} \)) the scattering in the entrance window may cause additional background seen as a broad peak under the narrow pin-hole peak. The \( \gamma \)-sensitivity is very low with a background of less than 0.1 count/s total counts under normal operating conditions. The overall stability of the detector system is adequate. Changes in the efficiency have been observed over periods of a month whereas the calibration of positional response remains unaltered over periods of six months.

5. PLANS FOR IMPROVEMENTS

The detector performance can be improved on several points. The interwire resistance can easily be tuned to improve the linearity in the positional response and the planes may be provided with guard electrodes that would eliminate the small background (< 0.02 counts/s) arising from to high-voltage leakage currents. Tests are also being made to use circular rather than square wire grids, which would increase the sensitive area from 1600 to 2800 cm\(^2\) without changing the outer dimensions of the detector.

ACKNOWLEDGEMENTS

The design of the electronics for the detector systems was carried out in collaboration with P. Christensen, and T. Freltoft has assisted with the computer software. The design and construction of the Risø SANS facility have been supported by grants from the Swedish and the Danish Natural Science Research Foundations.

REFERENCES

/3/ KJEMS, J.K., et al., IAEA-CN-46/P5, these Proceedings.
SMALL-ANGLE NEUTRON SCATTERING FACILITY AT RISØ

J. K. KJEMS, R. BAUER*, P. CHRISTENSEN, T. FRELTOFT, L. G. JENSEN, J. LINDERHOLM
Riso National Laboratory, Roskilde, Denmark

Abstract

SMALL-ANGLE NEUTRON SCATTERING FACILITY AT RISØ.

The new six metre small-angle neutron scattering facility at Riso National Laboratory is described. It is situated at a cold source beam outside the OR-3 reactor confinement at the end of a 20 m neutron guide. A variable tilt, mechanical velocity selector provides the monochromatization with $\Delta\lambda/\lambda$ in the range of 0.05 to 0.20. As a novel feature the sample is placed in high vacuum, $<10^{-6}$ mbar, without windows between the entrance slit and the 40 cm x 40 cm area detector. The sample can be rotated and translated in the vacuum which allows the use of cryostats, cryomagnets and furnaces without the introduction of extra windows. The instrument uses neutron wavelengths ranging from 4 to 20 $\AA$ with momentum transfers from 0.2 to 0.003 $\AA^{-1}$.

1. INTRODUCTION

The Risø SANS facility has been established with support from the Danish and the Swedish Science Research Councils. It came into operation in 1982 and it is used for structural studies in solid-state physics, chemistry, metallurgy and molecular biology.

The design has been inspired by the Laue-Langevin Institute (ILL) D17 instrument /1/ and by the Oak Ridge small-angle scattering facility /2/. The choice of dimensions was more or less dictated by the available space in the neutron house outside the DR3 reactor where the instrument is placed. The mechanical parts were all designed and constructed at Risø, including the area detector /3/, the dimensions of which, 40 cm x 40 cm, represent a manageable conservative first choice for the initial attempt in this field.

The first real experiments were carried out in autumn 1982 and so far the instrument has matched all the design goals.

2. TECHNICAL DESCRIPTION

The layout of the instrument is shown in Fig. 1. The main components are (1) the mechanical velocity selector, (2) the high vacuum collimator tubes, (3) sample chamber, (4) flight path and (5) the neutron area-sensitive detector.

* Present affiliation: Royal Veterinary and Agricultural University, Copenhagen, Denmark
2.1. Mechanical velocity selector

The neutron velocity selector consists of a rotating drum (max. 5000 rev/min) with tilted slits for the passage of neutrons. The spectral properties after passage through the monochromator are characterized by $\lambda = 7920 \times \theta / \nu$ (Å) and $\Delta \lambda / \lambda = 316 \theta / (\nu \lambda)$, where $\theta$ is the tilt angle in degrees and $\nu$ is the rotation of the drum in rev/min. For the peak of the cold neutron source, $\lambda = 3.3$ Å and $\nu = 5000$ rev/min we have $\Delta \lambda / \lambda = 0.19$ and an intensity of about $10^6$ n·s⁻¹·cm⁻² at the sample for circular apertures of 16 and 8 mm diameter.

2.2. Vacuum system

The high-vacuum system begins at the exit of the velocity selector, leaving 50 mm space for a beam monitor and exchangeable boron plast slits, typically 12 mm to 16 mm in diameter. The evacuated volume extends all the way to the area detector and is furnished with arrays of boron plast lined cadmium apertures that eliminate any stray scattering of neutrons. The distance from the entrance slit to the sample is 2.25 m and the sample-to-detector distance can be chosen to be either 3 or 4 m. The cylindrical sample chamber is placed on top of the pumping system, which consists of a rotary fore-pump, 38 m³/h, and a turbo-molecular pump separated
by a molecular sieve filter which removes traces of pumping oil. The high vacuum is better than 10^{-6} mbar and allows direct mounting of cryostats, furnaces and cryomagnets in the sample chamber - Fig. 2. A special sample changer with Peltier cell cooling/heating has been designed primarily for biological samples in standard quartz cuvettes. An xy-translation stage and a rotation stage allow for precise positioning and orientation of the samples in vacuum. Other auxiliary equipment includes permanent magnet sample holders, precision two-stage temperature controls and a special apparatus for stretching and quenching polymer samples. The flight path to the detector is surrounded by a conical, shielded tube which can be set at angles up to 20° relative to the direct beam. The beam stop is placed in front of the detector, suspended in thin steel wires and it is manoeuvrable in vacuum under remote control.

2.3. Area detector

The 40 cm x 40 cm area-sensitive neutron detector functions as a proportional gas counter /3/. The detector gas consists of 2 atm Ar containing 5% CO₂ and 0.5 atm $^3$He. The latter gas is used to produce ionized hydrogen and tritium from the reaction $n + ^3$He → p + $^3$H + 764 keV. The electrons from the ionization
tracks in the gas drift towards the anode and are multiplied near the 64-wire anode grid at +3.6 kV. The resulting currents are also picked up by the two parallel coupled cathode grids each with 64 wires held at a potential of about +1700 V. Both anode and cathode grids are used to determine the position of the above-mentioned neutron reaction impact via the difference in arrival times for the current pulse at the ends of the grids. The cathode wires are chosen to have a total delay of 2.4 µs (T = RC). The anode grid has small 200 Ω resistors interconnecting the wires and giving a similar delay. By measuring the arrival times at each end of the grids the neutron signal can be converted to a (x,y) position by standard electronic circuitry. The positional resolution is ~ 9 mm full width at half maximum.

2.4. Electronics and computer

A two-parameter Canberra 80 64 x 64 store the x and y co-ordinates corresponding to the neutron events in the detector. The two-dimensional spectra can be displayed either as a contour plot or as a three-dimensional overview. By means of a PDP 11-23 computer on line with the dual parameter system, it is possible to convert the spectra into (kx,ky) space co-ordinates and thereafter to analyse the data in various ways. The computer is also linked to a CAMAC system which, among other features, controls the (x,y) translation of the sample holder, the velocity of the velocity-selector drum and the temperature of the sample. In addition, the computer is equipped with a 512 x 512 pixel colour graphic screen and a sixteen-colour plotter for graphical data analysis.

The data acquisition is performed by the multi-channel analyser independent of the computer, and hence the user may carry out preliminary data analysis and storage during normal counting operations. The standard programs feature the usual Guinier and Porod plots as well as the corrections for background and detector sensitivity that are customary for such facilities. At any stage of the analysis the data may be transferred to floppy discs and/or the central computer system for further processing. A separate, identical PDP-11 working station is available in the close vicinity of the spectrometer for this purpose.

2.5. Neutron flux

The neutron flux at the exit of the guide at which the SANS instrument is situated has been measured in collaboration with R.E. Lechner /4/. The spectrum is shown in Fig. 3 as the total differential flux integrated over the angular divergence. This divergence is determined by the total reflection angles in the Ni-coated guide and it depends linearly on the neutron wavelength. To derive the flux at the sample position in the SANS spectrometer this divergence and the apertures have to be taken into account. A typical value with a 12 mm diameter entrance slit is $10^6$ n.cm$^{-2}$.s$^{-1}$ at 3 Å falling approximately exponentially to $10^4$ n.cm$^{-2}$.s$^{-1}$ at 18 Å.
3. USE OF THE FACILITY

So far the Risø-SANS facility has been used for studies in biology, polymer and colloid chemistry and physics, metallurgy and solid state physics (magnetism) with approximately equal weight in these areas. It is managed by the solid state physics section of the Physics Department and the use is supervised by a committee with representatives from the Danish and Swedish Natural Science Research Councils, Risø National Laboratory and the users.

Outside users are welcome on a collaborative basis.

REFERENCES

/4/ LECHNER, R.E., private communication.
RELAXATION OF ALIGNED ROD-LIKE MICELLES

L. HERBST, H. HOFFMANN, J. KALUS, H. THURN
Physikalisches Institut und Institut für Physikalische Chemie
der Universität Bayreuth,
Bayreuth,
Federal Republic of Germany

K. IBEL
Institut Max von Laue - Paul Langevin,
Grenoble

Abstract

RELAXATION OF ALIGNED ROD-LIKE MICELLES.

Rod-like micelles of Cetylpyridiniumsalicylate (CPS) in D₂O were aligned in
a couette flow experiment with a shear gradient of Π = 37 s⁻¹. The small-angle
neutron scattering (SANS) intensity showed a marked anisotropy, reflecting the
alignment of the micelles. The relaxation of the aligned micelles was studied after
the sudden decrease of Π to zero in time intervals of multiples of 200 ms. For each
time an effective rotational diffusion coefficient D can be extracted. D shows a
tendency to decrease with time.

1. INTRODUCTION

Systems of rod-like particles have been studied for a long
time. Usually this is done by rheological, optical, electro-optical
methods or combinations of these methods. With SANS we have the new
possibility of seeing the distribution function f of the orientation
of the rods as well as the time-dependence of f in great detail.
It can be assumed that a study of f can be of importance with
respect to the peculiar behaviour (CPS is visco-elastic and shows a
time-dependent viscosity) of these micellar solutions.

2. THE SAMPLE AND THE SHEAR APPARATUS

In a solution of 10 mM Cetylpyridiniumsalicylate in D₂O with
10 mM NaCl rod-like micelles, with a diameter of 42 Å and a length
of ~500 Å, exist. The micelles were aligned in a shear apparatus
and studied in a SANS experiment at the instrument D11 from ILL
(Institut Max von Laue - Paul Langevin) /1/.

The principal features of the scattering apparatus are shown in
Fig. 1. Figure 2 shows the apparatus in more detail. Between the two
fixed windows, 1 and 2, rotates a disk, 3. The windows, which are
glued to the parts 6 and 7, and the disk, are made of quartz glass
and are each 0.1 cm thick. The distance between windows and disk is
0.1 cm. The disk is connected to an axis, 4, which is driven by a NC
motor not shown in Fig. 2. Parts 6 and 7 are made of stainless steel. Teflon is the material used for the bearings, 5, and the seals are O-rings of rubber. Copper tubes, soldered to the part, 6, can be connected to a cryostat, allowing the temperature of the apparatus to be changed. The shear rate $\Gamma$ can be varied between 300 s$^{-1}$ and zero. The shear rate, $\Gamma$, is not constant over the window. For our purpose the deviation from the mean value $\Gamma_0$ is not severe. Generally, the diameter of the neutron beam is 0.1 cm; this has to be compared with the distance of 6.5 between the centre of the axes and the window. Therefore $\Gamma = \Gamma_0(1 \pm 0.08)$. The temperature was fixed at 25°C for our experiment.
FIG. 3. SANS curve of equal intensities for time $t = 225$ ms after the sudden change to zero of the shear gradient $\Gamma$. The intensities are 500, 1000, 1500, 2000 and 3000, respectively. The disturbance in the middle of the figures is due to the beam stop.

FIG. 4. As Fig. 3, but $t = 425$ ms.
3. RESULTS OF THE MEASUREMENTS

The experiment was done as follows: for 15 s the shear rate $\Gamma$ was fixed at 37 s$^{-1}$. At time $t = 0$ we reduced $\Gamma$ within a time of less than 0.1 s to zero and observed, in intervals of multiples of 200 ms, the changing anisotropic scattering pattern. The scattering curves, shown in Figs 3, 4 and 5, are highly anisotropic, indicating a substantial alignment of the rods. The scattering pattern is related to the distribution function $f$ of the rod axis. In fact, for $t < 0$ we prepared a non-equilibrium state of $f$, whereas for $t > 0$ we observed the time relaxation of $f$. Figure 5 shows the scattering after a relatively long time, by when anisotropy has nearly disappeared. Figure 4 shows an intermediate state.

4. ANALYSIS OF THE EXPERIMENTAL RESULTS

Currently we are analysing the time evolution of the scattered intensity by solving the Langevin equation /2/

$$\frac{\partial f}{\partial t} = D \left\{ \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left( \frac{\partial f \cdot \sin \theta}{\partial \theta} \right) + \frac{1}{\sin^2 \theta} \frac{\partial^2 f}{\partial \phi^2} \right\}$$

$$- \Gamma \left\{ \frac{\partial (f \cdot \omega(\theta) \cdot \sin \theta)}{\sin \theta \cdot \partial \theta} + \frac{\partial (f \cdot \omega(\phi))}{\sin \theta \cdot \partial \phi} \right\}$$

(1)
FIG. 6. The coordinate system for calculating the distribution function $f$.

for a rotational diffusion of particles. The relevant angles of the rod and the direction of the velocity vectors are shown in Fig. 6. Here, $f$ is the distribution function of the rod axis, $D$ the rotational diffusion constant, and $\Gamma$ the velocity gradient.

$$\omega(\theta) = -\sin^2 \cdot \sin \theta$$

$$\omega(f) = \frac{1}{4} \sin \theta \cdot \sin 2\theta$$

(2)

The state for $t < 0$ is characterized by $\partial f / \partial t = 0$, for $t > 0$ by $\Gamma = 0$. As mentioned above, the alignment of the rods is rather high. A calculated orientation distribution function,
approximately valid for our experimental situation at $t = 0$, is shown in Fig. 7. The results of our model calculations are shown in Figs 3 to 5 as smooth solid lines. At the moment it seems that the rotational diffusion constant $D \approx 0.5 \, s^{-1}$ depends on the actual distribution function, probably reflecting the influence of the interaction between the charged micelles. Such an interaction can be taken into account in the first order by the introduction of an effective $D$ in Eq. (1). Eventually the length of the rods depends on the actual shape of the distribution function. Up to now we have not been able to disentangle these two possible effects by using only the SANS measurements.

ACKNOWLEDGEMENTS

The authors would like to thank the Bundesministerium für Forschung und Technologie for financial support.

REFERENCES

A PULSED NEUTRON MONOCHROMATIC BEAM AT THE ET-RR-1 REACTOR*

M. ADIB, A. ABDEL-KAWY, Y. EID, R. M. A. MAAYOUF
Reactor and Neutron Physics Department,
Nuclear Research Centre,
Atomic Energy Authority,
Cairo, Egypt

Abstract

A PULSED NEUTRON MONOCHROMATIC BEAM AT THE ET-RR-1 REACTOR

A pulsed neutron monochromatic beam, at the ET-RR-1 reactor, is produced by two 32 cm diameter rotors suspended in magnetic fields, whose centres are 126 cm apart rotating at speeds up to 16 000 rev/min. Each of the rotors has two slots, which are of constant cross-section in area: 7 x 10 mm², and are curved so that they have a maximum transmission for neutrons whose speed is 8.2 times that of the rotor tip. The jitter of the phase between the rotors at different rotation rates is found not to exceed ±1 µs. It has been found that both the observed time distribution and the TOF distribution of the neutrons at different rotation rates are in good agreement with the calculated ones. The observed intensity of the monochromatic neutrons of wavelength 2.74 ± 0.09 Å, obtained by the rotors rotating at a speed of 10 500 rev/min with 864 ± 1 µs difference in phase between them, is 66.8 n/s. This value is found to be less than the predicted one by a factor of 5.5.

INTRODUCTION

Egelstaff et al. /1/ obtained almost neutron mono-kinetic pulses by using two curved-slot rotors placed at a suitable distance from each other, rotating at the same speed. By adjusting the difference in phase between the two rotors to correspond to the time-of-flight (TOF) between them, of neutrons with the desired velocity, it is possible to select these neutrons, and to reject all others except for those with velocities in a narrow range close to the one selected.

Several authors /2,3/ have shown that, for a required wavelength resolution and rotor radius, the obtained neutron intensity is higher at high angular velocity w. However, the strength function of the rotor material, together with the dynamic forces, encounter serious limitations on the maximum angular velocity which can be obtained. To reduce the enumerated difficulties Egelstaff /1/ and Krebs /2/ reported a phased rotor

* Work Supported by the Division of Technical Assistance, IAEA, Vienna
system with a wire suspension. However, as reported by Mook et al. /4/, such high-speed rotating equipment is expensive to construct and quite often needs a fair amount of maintenance. Kalebin /5/ designed a phased rotor system using a magnetic suspension instead of the wire one.

The present work deals with measurements of the main parameters of the pulsed neutron monochromatic beam, produced by the two-rotor-system suspended magnetic fields. This system was received through IAEA technical assistance and recently installed in front of one of the ET-RR-1 reactor's horizontal channels.

EXPERIMENTAL FACILITY

The facility consists of two similar rotor systems, each of which is mounted on its mobile platform. The general view of the rotor system is shown in Fig. 1. The rotor is 32 cm in diameter, is made from nickel alloy, and has two slots, which are of constant cross-sectional area (7 x 10 mm²) and a curvature radius of 65.65 cm. The distance between the centres of the rotors is 126 cm, while the analysing flight path is 224 cm. A helium-3 neutron detector battery is mounted at the end of the flight path. A 1024 multi-channel time analyser, with channel width varying from 0.25 to 64 μs, is used during the measurements. The double-rotor system is described in detail in Ref. /6/.

FIG. 1. Schematic diagram of one of the rotors.
Channels

Time μs

10 -2 -1 0 1 2

Counts x 10^-4

- - 7300 rev/min
• 9655 rev/min

FIG. 2. Jitter phase between the rotors.

PULSED NEUTRON PARAMETERS

Jitter phase between the rotors is considered one of the most important parameters of the system. As reported by Adib et al. /6/, the jitter of the phase between the two rotors was found not to exceed 1.5 μs and had a double-peak behaviour around the zero phase. This was because of the damping unit used. A new damping unit has been designed specially to decrease such fluctuations. The jitter of the phase between the two rotors at different rotation rates is presented in Fig. 2, where it can be seen that the double peak has disappeared with the improvement of the jitter phase. The new value does not exceed ±1.0 μs.
The neutron monochromatic beam, typically observed at a rotation rate of 10 500 rev/min is presented in Fig. 3 for different values of the phase between the two rotors. The TOF is measured from the centre of the first rotor to the detector where the flight path is 350 cm. The maximum transmission for neutrons of wavelength 2.74 Å is found to correspond to 864 µs difference in phase between the rotors.

Figure 4 shows the time distribution of neutrons in the pulse at the detector position, along with the calculated one for maximum transmission /3/. One can see that both distributions are in good agreement. This means that the standard deviation of the pulses at a sample position 40 cm from the centre of the second rotor will be 11 µs.

The neutron TOF distribution in pulse obtained for maximum transmission at rotation rates 10 500 and 11 650 are presented in
Fig. 5. The solid lines are the calculated distributions following Royston /3/. One can observe that the experimental distributions are in good agreement with the calculated ones. From Fig. 5 one can also obtain the wavelength resolution $\Delta \lambda / \lambda$ as 3.2% for neutrons with $\lambda = 2.74 \, \text{Å}$. The observed intensity of the monochromatic neutrons of wavelengths $2.74 \pm 0.09$ and $2.47 \pm 0.08 \, \text{Å}$ are found to be 66.8 and 92.2 n/s, respectively, while the calculated intensity, taking into account the attenuation in air and the detector efficiency, are 370 and 507 n/s, respectively.

At the present stage, and in order to increase the intensity of the neutron monochromatic beam, we started to realign the whole system using a specially designed optical system, along with neutron guide collimators coated with $^{58}$Ni and located between the rotors. It is expected that this will increase the neutron intensity by one order.
FIG. 5. The neutron TOF distribution in the pulse.
ACKNOWLEDGEMENT

The authors would like to express their gratitude to the Laboratory of Dr. Kalebin for manufacturing the double-rotor system.

REFERENCES

MULTIPURPOSE DOUBLE-CRYSTAL MONOCHROMATOR FOR THERMAL NEUTRONS

P. MIKULA, J. KULDA, B. CHALUPA, R. MICHALEC*, M. VRANA*, L. SEDLÁKOVÁ*
Nuclear Physics Institute, Řež, Czechoslovakia

Abstract

MULTIPURPOSE DOUBLE-CRYSTAL MONOCHROMATOR FOR THERMAL NEUTRONS.

A new type of a highly efficient double-crystal monochromator (DCM) has been proposed and experimentally tested for thermal neutrons based on two elastically bent silicon single crystals, both set in a strongly asymmetric and parallel (1,-1) setting. It has been found that the reflectivity properties of this DCM may be even compared with the best single-crystal mosaic monochromators. Such a simple arrangement ensures easy control of the effective mosaic aspect and very low background at the place of the sample. Relatively high angular resolution of the (1,-1) double-crystal (DC) setting enables the DCM to be used as a medium-resolution instrument for the study of small-angle neutron scattering. Further, the use of simultaneous diffraction, e.g. on two sets of planes, enables two diffractometers to be installed on one incident polychromatic beam.

1. INTRODUCTION

Despite considerable progress in the production of good quality mosaic monochromators in recent years, much effort is still being devoted to this problem. Considerable attention is chiefly being paid to achieving a routine control of the growth of a mosaic crystal with "ideal" distribution and dimensions of mosaic subgrains from materials with promising neutron-optical properties /1/.

One of the best materials for producing mosaic monochromators has proved to be pyrolitic graphite /2/, which, owing to its large spacing and usually broad mosaic spread, is mainly used for monochromatization with a neutron wavelength above 0.2 nm. At present at the Institut Max von Laue - Paul Langevin (ILL), Grenoble, a programme concerning the production of beryllium monochromators is being conducted and a fair chance exists that the growth of high quality Be crystals may become a routine matter in the near future /3/. Then a correct mosaic distribution could be established by means of plastic deformation.

Alternatively, it is possible to control easily the resolution and reflecting power by the elastic deformation of perfect crystals through bending, applying a thermal gradient etc. For ordinary
reflection and transmission geometries, the use of elastically deformed crystals as monochromators has been restricted because of a relatively small effective "mosaic spread" $\delta \Theta$, being of the order of $10^{-3}$ rad. By applying a greater deformation gradient on the crystal one can break it, or the enlargement of $\delta \Theta$ may be accompanied by a decrease of the peak reflectivity $r$. In our case this problem was solved by using a fully asymmetrical geometry permitting $\delta \Theta \sim 10^{-2}$ rad /4/ to be reached. The associated effect of the reflected beam widening on the first crystal is compensated for by the second crystal in the $(1,-1)$ setting in opposite geometry.

2. PARALLEL $(1,-1)$ SETTING OF TWO BENT Si CRYSTALS

Let us suppose the experimental arrangement as schematically displayed in Fig. 1, and that incident neutrons enter the first crystal through its end-face and pass through it along its longest edge. The total change in Bragg angle for the incident beam on its path through the first crystal $(\delta \Theta)_I$, which is considered as an effective mosaic spread, is given by relation /4/

$$ (\delta \Theta)_I = L/R, $$

(1)

![Fig. 1. Schematic arrangement of the proposed monochromator set-up.](image-url)
FIG. 2. The optimum length and radius of curvature and the corresponding integrated reflectivity as functions of the wavelength for 111, 220 and 400 reflections of a Si crystal with fixed "effective mosaicity", $\delta \phi$.

FIG. 3. The efficiency $\rho / \delta \phi$ (full lines) and the peak reflectivity $r$ (dashed line) as functions of the radius of curvature $R$. Curves 1, 2, 3, 4 correspond to different lengths of the crystals equal to 5, 10, 15 and 20 cm, respectively.
where $L$ is the length of the crystal bar and $R$ the radius of curvature. Further, let us set $(\delta_0 \theta)_I = \delta \theta$. The neutrons reflected on the first crystal may be successively reflected on the second crystal, when both crystals are in the precise parallel position and are bent in the same sense with the same radii, $R_I = R_{II}$. On the basis of the lamellar model of diffraction by an elastically deformed crystal, integrated reflectivity $\rho^0$ of this DC system may be written in the form /4/

$$\rho^0 = (\delta_0 \theta \theta R) \exp(r(R) \pi^2 A(\mu, L)), \quad (2)$$

where $r(R)$ is the peak reflectivity /5/ and $A(\mu, L)$ the attenuation factor with the linear attenuation coefficient $\mu$. In this case $A(\mu, L) = \exp(-\mu, L)$ and $r(R) = 1 - \exp(-\nu R)$, where $\nu = F^2_R \lambda^2 / (V_c \sin 2\theta)$, $F_R$ and $V_c$ being the structure factor and the volume of the unit cell, respectively. The calculation of an extremum of $\rho^0$ for fixed $\delta \theta$ and $\lambda$ yields $L_{opt}$ in the form

$$L_{opt} = (\delta_0 \theta / 0) \log \left[1 + 20 / (\mu \delta \theta)\right], \quad (3)$$

$R_{opt}$ is given by $R_{opt} = L_{opt} / \delta \theta$ from Eq. (1).

Figure 2 displays the dependences of $L_{opt}$, $R_{opt}$ and $\rho^0(L_{opt}, R_{opt})$ as functions of $\lambda$ for 111, 220 and 400 reflections of an Si crystal with fixed $\delta \theta$.

Besides using all the optimum parameters for $\delta \theta$ and $\lambda$ fixed in advance, another approach, consisting in an arbitrary choice of the bending radius $R$ for fixed $L$ and $\lambda$, which yields the possibility of an easy control of $\delta \theta$ and $\rho^0$, may be employed. In this case it is convenient to treat the ratio $\rho^0 / \delta \theta$ (the efficiency of DCM) as a function of $R$ for several values of $L$. Corresponding dependences for 111 reflection are demonstrated in Fig. 3.

3. EXPERIMENTAL

The test experiments were performed at the 5 MW WWR-S reactor by means of a triple-axis spectrometer TKSN-400 /6/ with a Zn mosaic monochromator with a mosaicity of about 17' on the first axis. The Si crystals having the same dimensions 200 x 30 x 5 mm$^3$ (length x height x thickness) were placed on the second and third axes. The Si plates were cut from a dislocation-free perfect crystal ingot grown in the 111 direction. In our experiments we used 111 and 400 diffracting planes, making an angle of about 18.6° and 36.3° with the largest surface which, in the case of fully asymmetric geometry, correspond to $\lambda = 0.2$ nm and 0.157 nm, respectively. The beam path between the Si crystals was $L = 50$ cm.

Figure 4(a) and 4(b) display the experimental results of the efficiency $\rho^0 / \delta \theta$ versus the angular departure $\Delta \theta$ of the second crystal from parallel setting for 111 reflection, $R = 78$ m and 400 reflection, $R = 36$ m, respectively. An agreement between the experiment and the theory may be considered according to the ratio $(\rho^0 / \delta \theta)_{exp} / (\rho^0 / \delta \theta)_{th}$ at 0.94 and 0.78. In both cases this ratio
FIG. 4. The efficiency $\rho_0 / \rho_0$ of the DC monochromator versus the rocking angle $\Delta \phi$ of the second crystal in the vicinity of the precise parallel setting (curve 2). Curve 1: maximum theoretical efficiency.

FIG. 5. Double-crystal rocking curves for different curvature radii. Curves 1, 2 and 3 correspond to the radii equal to 78, 36 and 20 m, respectively.
FIG. 6. The dependence of the FWHM of the double-crystal rocking curve versus the reciprocal value of the bending radius.

FIG. 7. An example of the rocking curves measured with (o) and without (•) a SANS sample (2 mm Teflon foil).
FIG. 8. Time-of-flight distribution of double-reflected neutron beam for an angle deviation from the parallel setting $\Delta \theta = 20''$, $R_i = 150$ m and the vibration amplitude of the second crystal $u_{||} = 20$ $\mu$m. The channel width is 4 $\mu$s.

was found to be constant within 5% for different radii employed, up to $R = 20$ m. Figure 3 reveals that, for sufficiently large $R$, the efficiency $\rho(\theta)$ on an absolute scale is mainly influenced by the attenuation $A(\rho, L)$. Further, it is clear from Fig. 2 that $L = 20$ cm of the crystal plates available is much longer than the optimum length $L_{opt}$ for $\theta_0 = 8.73 \times 10^{-3}$ rad fixed in advance. The rather low value of the efficiency corresponding to 400 reflection originates from the horizontal divergence of the incident beam $\Delta \omega$. It reveals that neutrons reflected in any point of the first crystal are, on the second crystal, spread over an area $\Delta y = 2\Delta \omega / \cos 2 \theta_0$ along the axis perpendicular to the largest surface. In the case of 400 reflection $\Delta y = 7.4$ mm considerably exceeds the thickness of the second crystal. Hence, the thickness of the second crystal should be about the value $\Delta y$ greater than the one of the first crystal.

The FWHM of the DC rocking curve is determined by convolution of the intrinsic diffraction patterns of both crystals with approximately rectangular forms of widths $(\delta_{\theta_0})_I$ and $(\delta_{\theta_0})_I$ being of the order of $5 \times 10^{-4}$ rad. The symbols $(\delta_{\theta_0})_I$ and $(\delta_{\theta_0})_I$ mean the total changes in Bragg angle experienced by the reflected and incident beams on their paths through the first and second crystals, respectively.

Figure 5 displays the DC rocking curves measured on the 111 planes, with the neutron wavelength $\lambda = 0.2$ nm for different bending radii.
Figure 6 displays the dependence of FWHM of the DC rocking curves as a function of the reciprocal value of the bending radius. A very important property characteristic of any DCM is a very low background-to-peak ratio. In our case at the 4 x FWHM angular distance from the peak of the rocking curve, a background-to-peak ratio of $3 \times 10^{-5}$ was observed.

The relatively small value of FWHM, low peak-to-background ratio, and the high flux of monochromatic neutrons make it possible to use this DCM as a medium-resolution DC instrument for studying small-angle neutron scattering (SANS).

Figure 7 demonstrates an example of the SANS experiment performed with Teflon foil. Owing to the large dimensions of the beam between the crystals, DCM might be especially suitable for the SANS study of a strongly absorbing sample which could be in the form of a thin sheet of a large square. In such a case a relatively large volume of the sample might be studied without a significant loss of the neutron flux due to the absorption.

By exciting both bars in flexural mode vibrations with a suitable phase shift between the crystals, one can obtain a pulsed output monochromatic beam, as was demonstrated in Ref. /7/ for symmetric Bragg geometry.

In our experimental geometry we succeeded in obtaining a pulsed output beam by exciting only the second crystal in a co-operative action with the first bent one to avoid parasitic pulses in the TOF spectrum, a Cd slit was placed between the crystals, screening the first half of the second crystal.

Other experimental results including those on the focusing may be found in Ref. /8/.

4. CONCLUSION

The maximum usable crystal thickness, and hence the width of the output beam, are determined by a limit value of the bending radius. Consequently, a use of such a simple arrangement of DCM seems suitable for experiments with small samples and for instruments on neutron guide tubes, where the width of the incident polychromatic beam and its horizontal divergence are relatively small.

The use of simultaneous diffraction, e.g. on two sets of planes, enables two diffractometers to be installed on one incident polychromatic beam.

REFERENCES

/6/ PETRZILKA, V. et al., Jad. Energ. 11 (1972) 367.
POSTER PRESENTATIONS
CRYSTALS AND TEXTURE

(Session X.d)
Chairmen

T. SPRINGER
W. SCHMATZ

Federal Republic of Germany
GROWTH OF LARGE COPPER MONOCHROMATOR CRYSTALS

J.-M. WELTER, G. JUNGERBERG
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH, Jülich,
Federal Republic of Germany

Abstract

GROWTH OF LARGE COPPER MONOCHROMATOR CRYSTALS.

To fabricate copper monochromators for neutrons, large single crystals with
the following specifications were grown - diameter: 6.5 cm; length: up to 20 cm;
orientation: (110); orientation and mosaic spreads: less than 2'. The starting
material was high-purity electrolytic copper (99.999%) which was vacuum remelted to
make the blanks. Single crystals were obtained by directional solidification of a
seeded melt contained in a high-purity graphite crucible (Bridgman technique with
stationary crucible and moving furnace). The structural quality of the crystals was
controlled by diffractometry using 468 keV γ-rays emitted by an 192Ir source.

1. INTRODUCTION

Our aim is to describe the production line of copper crystals which was started three years ago at the request of the Institut Laue - Langevin for the fabrication of neutron monochromators. The specifications of the crystals dimensions and structural quality were fixed by the technique used during this fabrication [1]. Crystal blocks with the dimensions 8 x 4 x 4 cm³ were plastically deformed by controlled hot-pressing. To produce an anisotropic mosaic distribution the long edge has a (110) orientation. A homogeneous mosaic spread is only obtained if the starting crystal block is nearly perfect, i.e. if it is free of small-angle grain boundaries. Therefore, the requirements for the crystals were:

- diameter: 6.5 cm
- length: up to 20 cm
- orientation: (110)
- orientation constancy: ± 1'
- mosaic spread: < 2'

Owing to existing equipment and infrastructure we succeeded after a relatively short period of trials in growing such crystals on a routine basis using a Bridgman technique. At present the yield is better than 66%. The principal failure mode is the development of an excessive mosaic spread in the final solidifying part of the crystal.
Although the growth process appears to be the most important step of the production, two other aspects must be pointed out. First, careful selection of the raw copper material and preparation of the blanks is crucial. As the crystals weigh 6 to 7 kg, cheap copper brands must be used. On the other hand, high-structural quality is only obtained at a very low impurity level. To find the optimal material a copper market survey was made together with the Institut für Metallkunde and Metallphysik of the RWTH Aachen [2,3]. Secondly, good in-house control possibilities of the structural perfection is essential. Our γ-ray diffractometer was an invaluable tool which allowed for immediate feedback between the structural quality of the crystal and the growth conditions. An apparatus was even built to grow small (seed) crystals in-beam on the γ-ray diffractometer and to check whether this technique should also be used in the case of the large crystals [4,5].

2. BLANK PREPARATION

Impurities have a detrimental influence on the structural quality. Precipitates in copper (usually oxides and sulphides) may nucleate dislocations and subgrain boundaries. Inhomogeneously distributed metallic impurities may lead to chemico-elastic strains. Such a segregation can result from constitutional supercooling which occurs at excessive impurity levels [6]. Thus, high-purity, low-cost copper is needed. Fig. 1 shows the result of our market survey [3]. The quality index is the residual resistivity ratio RRR measured on directionally solidified rods (ф 1.2 cm). Selected high-purity cathodes produced on an industrial scale are available at a low price (field I-C) and may even reach the quality of the best laboratory-made copper specimens (field L). So-called high-purity vacuum-remelted bars (field I-B) can be disregarded.

Selected copper cathodes leading to RRR values between 3000 and 5000 were used to prepare the blanks by melting. Spark-source mass-spectrometry analysis detected the following impurities (in at. ppm):

<table>
<thead>
<tr>
<th></th>
<th>N</th>
<th>O</th>
<th>Na</th>
<th>Cr</th>
<th>Fe</th>
<th>Ag</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1.3</td>
<td>4.5</td>
<td>1.7</td>
<td>1.6</td>
<td>0.9</td>
<td>0.6</td>
</tr>
</tbody>
</table>

The combination of high-purity graphite crucibles, medium-frequency induction heating which stirs the melt well and a residual atmosphere below 1 mPa, led to a thorough degasing of the material.
3. CRYSTAL GROWTH

The crystals were grown by directional solidification of the seeded melt in graphite crucibles. As for the growth of nearly perfect large crystals, the advantages of soft crucibles made from graphite powder have not been demonstrated unambiguously [7], we preferred to use hard crucibles in view of the ease of fabrication and handling.

The (110) oriented seed crystals had a diameter of 0.6 cm and a length of 6 cm. At the beginning perfect crystals grown by the Czochralski technique were used [8]. As it turned out that the seeds were badly damaged by the melting blank, the neck region was optimized and seeds with a lower quality could be used. They were either grown in the in-beam γ-ray diffractometer apparatus or machined out of a large crystal by spark erosion.

The crucible was placed on a water-cooled support and the pressure of the ambient atmosphere was lowered below 10 mPa. After melting the blank, the furnace was moved upwards at a speed between 1 and 2 cm/h. A feature of the furnace was its very high thermal capacity. Therefore, the total duration of the heating, growth and cooling periods was between 60 and 72 h. To minimize the influence of building vibrations, the furnace was mounted in a heavy 2.65 × 1.60 × 1.35 m$^3$ frame made from section steel.

FIG. 1. Price versus residual resistivity ratio \( R (300\text{K})/R (4.2\text{K}) \) for various copper brands. L: laboratory scale; I: industrial scale; C: cathodes; B: bars.
FIG. 2. Map of the FWHM of γ-ray rocking curves measured at different locations of a copper crystal. The (200) scattering vector is perpendicular to the crystal axis.

FIG. 3. Map of the FWHM of γ-ray rocking curves of a copper crystal. a: location of the analysed volumes; b: (220) scattering vector parallel to the crystal axis; c: (200) scattering vector parallel to the crystal axis. The deviation of the bars in b and c from the dashed line shows the local misorientation of the lattice planes.
4. STRUCTURAL PERFECTION

The tilts of the lattice planes in the crystal, which are due to dislocations and subgrain boundaries, were investigated by γ-ray diffraction [9]. The γ-ray source was a neutron activated iridium foil. The beam had a cross-section of $1 \times 0.02 \text{ cm}^2$ and a minimal divergence of $12^\circ$. The 468 keV line ($\lambda = 2.64 \text{ pm}$) of $^{192}\text{Ir}$ was used. The absorption length of 1.3 cm allows to analyze thick copper crystals with this radiation. Furthermore, the large (average) extinction length of 90 μm leads to minimal extinction effects. Consequently, the full width at half maximum (FWHM) of the rocking curves can be used as a quality index for the mean mosaic spread.

Fig. 2 shows the perfection map of a crystal which illustrates two aspects mentioned previously. First, the seed crystal and the re-solidified part have a very poor structural quality. Indeed, the rocking curve of the crystal part below the neck reveals various pronounced subgrains. But only one grain passed the neck constriction and led to a nearly perfect cone region. Secondly, the perfection deteriorated somewhat again in the upper part of the crystal.

In Fig. 3 control measurements made at the Institut Laue-Langevin on one of our nearly perfect crystals are shown.

A question which was discussed was the possibility of growing crystals under γ-ray diffraction control. A pilot unit demonstrated the feasibility of this concept [4,5] - when more than one subgrain passed the neck constriction the growing crystal could be remelted and a new trial was started. Nevertheless, we did not pursue this concept for the growth of the large crystals, as the quality deterioration usually occurred at the end of the growth process. Therefore, the time saving would not compensate the efforts associated with the modifications of the equipment.

5. CONCLUSION

The reasons why large, nearly perfect copper crystals could be grown, are:

- optimal neck design;
- high-purity starting material;
- smooth temperature profile in the furnace;
- low vibration level.

The first measure ensured a good selection process and the other ones minimized the strains in the growing crystals.
ACKNOWLEDGEMENTS

This work could only be performed with the help of many specialists. The authors want to thank Dr. H. Beske, Mr. F.J. Bremer, Mrs. R. Fischer, Dr. A. Freund, Mrs. L. Gain, Dr. D. Lenz, Mr. Ch. Mambor, Dr. W. Uelhoff, Prof. H. Wenzl for discussions and assistance.

REFERENCES

TEXTURE DETERMINATION BY NEUTRON DIFFRACTION

A. OLEŚ, J. KULKA, J. SZPUNAR, R. WAWSZCZAK
Institute of Physics and Nuclear Techniques,
Academy of Mining and Metallurgy,
Kraków, Poland

Abstract

TEXTURE DETERMINATION BY NEUTRON DIFFRACTION.

Since 1968 the neutron diffraction method has been used in our Institute to determine the crystallographic texture. The main advantages of this method are low absorption of neutrons and large geometrical cross-section of neutron flux. The original texture goniometer and the electronic equipment for the texture experiment have been constructed. The whole equipment gives the maximum measurement flexibility and ensures comfortable operating in long-lasting experiments. The neutron diffraction method for texture determination was used to investigate polycrystalline materials as follows: the texture of industrial and mineral graphites, Goss texture of Fe-Si transformer sheets; textures of Fe-Ni meteorites; correlation between the texture and fatigue cracking; texture of reactor fuels, wires and rods. The correlation between texture and physical properties was investigated.

INTRODUCTION

The first neutron diffraction experiments with texture were carried out in 1953 /1/ and were soon followed by others /2-7/.

Since 1968 we have been using the neutron diffraction method to determine the crystallographic texture. The grain-orientation distribution (texture) provides important information about the polycrystalline materials. The main advantages of the neutron diffraction method as compared with the X-ray diffraction are low absorption of neutrons in most industrial materials, and large geometrical cross-section of neutron flux. These traits make it possible to perform texture studies in coarse-grained materials. Neutron diffraction provides information on the average texture. Quite often the material investigated is not homogenous. In such cases it is important to determine the average texture, which influences the anisotropy of the physical properties.

THE DESCRIPTION OF THE METHOD

A beam of neutrons with a fixed wavelength is scattered on the sample. The position of the detector is stable and corresponds with a proper Bragg angle for the chosen plane (hkI). The sample is mounted on a goniometer in the centre of the table of the spectrometer. It is rotated in such a way that each plane in the sample can be set to reflect the neutrons into the detector. The pole figure can be determined. The intensity of neutrons is proportional to the density of poles on the pole figure.
An important variant of the neutron diffraction method of texture determination is based on the time-of-flight idea. This method is especially useful for pulsed neutron sources.

The main aim of the diffraction experiments is to determine the crystalline orientation distribution function (ODF). This function can be calculated on the basis of pole figures. In most cases three pole figures have to be measured, e.g. type of (110), (200), and (211). The calculations of the ODF were described by Bunge /8,9/. Many details on texture determination are given by Szpunar /10/.  

TEXTURE GONIOMETER AND ELECTRONIC EQUIPMENT  

The mechanical construction of the goniometer fulfils the requirement for the sample rotation. There are two axes (see Fig. 1). The inner one performs the β rotation and its resolution is 10', the outer one performs the α rotation with resolution 1'. Both axes are driven by stepping motors. The sample holder is fixed in the centre of the β worm. Thanks to the female screw in this worm the sample holder can be lifted up and down to set the sample precisely in the centre of the goniometer. The α worm-wheel rolls on three small ball-bearings installed in an outer frame. To remove the backlash in the work worm and the worm-wheel, the work worm has one end fixed in an eccentric ball-bearing holder. Both motors have elastic connections with the worms.

The electronic equipment (Fig. 2) used in the texture experiment was constructed as a single crate system, following the CAMAC standards. The analog part contains high voltage power supplies for two detectors (one working as a monitor of the neutron flux) and threshold amplifiers forming pulses of both channels.

FIG. 1. (a) The sample rotation in the goniometer; (b) The α and β angles in stereographic projection.
FIG. 2. The conceptual scheme of the electronic system.

FIG. 3. Industrial graphite texture: (a) A pole figure; (b) An anisotropy of electrical conductivity /11/. 
Those pulses are counted and operated in the digital part. The programmed crate controller based on the microprocessor Intel 8080 defines the sequence of the experiment, controls the work of the digital blocks and organizes the data registration by a tape puncher or a miniature printer. The operator chooses limits for the motion of the sample, the density of the measurement points, duration time (measured by intensity of the beam or by the real time) and writes to the memory the actual axes positions after switch-on. The processor, the counters, the stepping motor drivers and the line printer interface have been manufactured especially for this experimental set-up. On the processor front panel the status of the address, data buses and some bits of the CAMAC control bus are displayed. One can write and read the memory using the hexadecimal keyboard. The dual six-decade scaler displays its contents, and the switches on the front panel make it possible to have some changes in the running measurement to stop, start, erase data, set the range of monitor counts, etc.

Two stepping motor axis positioners for two axes ensure the motion with proper velocity, according to the information written in the processor program. This block contains and displays the actual position of the axis. It is possible to control all its functions manually.

The whole equipment gives the maximum flexibility of measurements, comfortable operating in the long-lasting experiments and little expense for the input-output devices.

**STUDIES OF DIFFERENT MATERIALS**

Neutron diffraction is very useful when studying the structure and texture of graphites. The structure of industrial graphite is quite often not homogenous and not all parts of a specimen are well graphitized. The main component constitutes graphitized particles of cokes which stick together with the help of adhesive substances. All
materials are cut through by pores and inclusions. The grain size of the industrial graphite investigated was about 0.3 to 3 mm. Porosity was very high. Pores occupied about 30% of the volume of the specimen. Neutron diffraction is strongly recommended for the determination of graphite texture. The pole figure of industrial graphite and the related anisotropy of electrical conductivity are shown in Fig. 3.

The pole figure given in Fig. 4 contains information about the grain orientation of the Fe-Si sheet. It shows a texture of the coarse-grained material of Goss type \( \{110\} <001> \). This type of sheet texture is interesting in the production of transformer cores. We have investigated the texture at different stages of the transformer sheet production. The induction saturation strongly depends upon Goss texture perfection.

The texture of wire has an axial symmetry. Metals like copper have a preferred orientation of \(<111>\) direction along the wire. Many other metals in the form of wire and rods have been tested.

The crystallographic texture of iron-nickel meteorite has been determined by neutron diffraction /12/. The Fe-\( \gamma \) single crystal, composed from numerous lamellas separated by Fe-\( \alpha \) lamellas, has been found (Fig. 5). This single crystal has very low mosaic. Evidently, the collision with the Earth and the Fe-\( \alpha \) phase growth were not followed by destruction of the single crystal structure. The Fe-\( \alpha \) crystals, correlated spatially with a Fe-\( \gamma \) single crystal, exhibit granulation and dispersion. A polycrystalline material with statistical grains orientation has not been observed. These data relate the meteorite history.

The correlation between texture and fatigue cracking was studied in hexagonal metals /13/. The experiment confirms the given theoretical model. The neutron diffraction texture determination was applied for the prediction of the anomalous fracture directions in metals.

There are only few examples. Further texture studies for different polycrystalline materials are being continued.
REFERENCES

TEXTURE STUDIES OF COLD ROLLED ALUMINIUM AND COPPER SHEETS BY NEUTRON DIFFRACTION

M. M. BEG, N. M. BUTT, Q. H. KHAN, S. U. CHEEMA
Pakistan Institute of Nuclear Science and Technology, Rawalpindi, Pakistan

Abstract

TEXTURE STUDIES OF COLD-ROLLED ALUMINIUM AND COPPER SHEETS BY NEUTRON DIFFRACTION.

A systematic study of the texture of copper and aluminium cold-rolled sheets has been made by using the triple axis neutron spectrometer. Aluminium samples were made from pure aluminium ingots. Aluminium samples with 92% reduction in thickness showed (200) [200] texture. Cold-rolled sheets of copper, obtained both from heat-treated commercial ingots and from single crystal samples, were also studied. Sheets rolled from a single crystal follow the texture of the original single crystal. The results showed that the sheet texture depends on the history of the material. The measurements were made using neutrons of wavelength 1.175 Å obtained from a medium-flux swimming-pool reactor of 5 MW.

1. INTRODUCTION

During the industrial production of materials, various physical treatments such as pressing, rolling and drawing are commonly performed. In such processes, the polycrystalline distribution of the materials is grossly affected and preferred orientations of the small crystallites take place. Diffraction measurements on such samples do not give the same intensity for all positions of the sample in the beam; rather specific textures occur in such a sample /1-5/.

Texture is determined by diffraction techniques by setting the spectrometer for a Bragg reflection and rotating the sample in the beam to find if any special position of the sample gives a higher reflected intensity. Both the X-ray and the neutron diffraction methods are used for this purpose. In X-ray measurements the results are affected by the surface texture more than the interior texture, which may not be exactly similar. The neutron scattering is a bulk technique and a far greater volume of material is exposed /6-8/ to the beam by this method (about 1 cm thick). The surface effects are less significant in such measurements.

When rolling the sheets of the face centred cubic metals, (011) [211] texture is developed /7,9/ which means that the rolling axis is towards the [211] direction and the (110) plane is parallel to the surface of the sheet. Generally, a combination of more than one texture is observed. Two good review works – Szupunar /8/ and Richards /2/ – can be consulted for further details and more references. Some detailed theoretical works that predict the texture in metals are given in references /10-12/.
The presence of texture in the materials changes their mechanical properties such as tensile strength, Young's modulus and elastic anisotropy. Therefore, it is important to know the texture of the material when the above properties are important.

2. EXPERIMENTAL MEASUREMENTS

For texture measurements special goniometers are required. These help to rotate the sample in all possible directions.

A manually operated goniometer was made which enabled the sample to be rotated about a horizontal axis through an angle of 270°. The angle on this scale could be easily read to an accuracy of 1/4°. The sample could be rotated about the vertical axis over a range of 360°. This simple goniometer was good enough for texture measurements in steps of 2° x 2°. To mount the sample in the goniometer the rolling direction was marked on the face of the sample which was mounted so that the rolling direction was vertical.

FIG. 1. Schematic diagram of texture studies by neutron diffraction.
The neutron diffraction measurements were made with the triple axis spectrometer TKSN-400 installed at the 5 MW swimming pool research reactor. The available thermal neutron flux at the sample, using a graphite monochromator, was about $10^5$ n.cm$^{-2}$.sec$^{-1}$. Measurements of neutron intensities were made corresponding to reflections (111), (200) and (220). For each reflection the intensities were recorded, corresponding to 5° steps of angles $\alpha$ and $\phi$. $\alpha$ is the angle about the vertical axis and $\phi$ the rotation angle about the horizontal axis. The procedure of the measurement was that $\phi$ was fixed and $\alpha$ was moved in steps of 5°, and at each step neutron intensities were recorded. For the next series of intensity recording $\phi$ was moved to the next step and a series of measurements were made in that position by changing $\alpha$ in steps of 5°. $\phi$ was thus moved in steps of 5° to cover the complete quadrant. The starting position of the angle $\alpha$ was such that the plate sample bisected the angle $180-2\theta$ where $2\theta$ is the Bragg angle (transmission mode). The experimental arrangement is shown in Fig. 1.

3. TEXTURE MEASUREMENTS IN ALUMINIUM

The texture in aluminium wires has been measured by Schlafer and Bunge /10/, and later on cold-rolled aluminium sheets by Eder and Klemencic /5/ by X-ray and neutron diffraction. Eder and Klemencic prepared their samples from 99.9% pure aluminium slabs reduced in 20 passes to a thickness of 2 mm, which corresponds to a reduction of 90%. Ten circular discs of 35 mm each were stacked together so that the rolling direction was parallel throughout.
Thick samples were used to obtain good intensity. Eder and Klemencic mainly obtained the (124) [211] texture in their sheets. Generally for f.c.c. metals (110) [112] texture is expected.

Aluminium sample I

In the present experiment two independent samples were used for the studies. The first sample was made from a 5 cm thick (99.99%) purity aluminium bar, which was cold-rolled by several passes to 92% reduction to a final thickness of 2 mm. From this sheet three pieces measuring 2 cm x 2 cm were cut. One of the samples was annealed at 200°C for 3 hours to remove any strains other than the rolled texture. The third piece was annealed at 400°C for two hours. The ingot was neither annealed nor mechanically treated before rolling.

Figure 2 shows that, for the (200) reflection, the concentration of intensities is around angular positions α = 90° and φ = 0° or 90°. From these results it is found that the crystal planes (200) are aligned along the surface of the sample and the direction [100] is along the rolling direction. To reconfirm the (200) cubic orientation of the sheets all the six face planes of the cube were checked. Further, the (111) reflection shown in Fig. 3 also confirms the above results. This (200) [002] texture was observed in all the three samples cut from the same sheet referred to in the preceding paragraph. The above texture is also confirmed by the (111) pole figure shown in Fig. 3.
Aluminium sample II

The experiment was repeated on material from another ingot. The ingot was annealed at 400°C before rolling. The material (99.99% purity) was reduced by 92% in thickness by cold rolling. The piece was rolled by giving 2.5 - 3% deformation per pass. The initial thickness of the material was 25.3 mm, which was reduced to 2 ± 0.05 mm. Three rectangular pieces, cut from the same strip, were joined together to increase the scattering intensity. The sample was mounted with the rolling direction vertical. (200), (220) and (111) polar diagrams shown in Figs 4, 5 and 6 were measured. The results observed from this sample were different compared with the first sample. The observed texture was a super-position of (110) [112] and a weaker (111) [211] texture. The results on sample II were in agreement with previous works /1,2/.

To investigate the reason for the observed differences in the two aluminium samples it was decided to study another f.c.c metal like copper with two samples, one a single crystal and another a polycrystalline sample. The former sample had an initial texture while the latter ingot had no preferred orientation.

4. TEXTURE OF COPPER

Sample I

To determine the effect of the initial orientation on the final texture, a piece cut from a single crystal of copper was cold-rolled. The thickness was reduced by about 90%. (200), (220) and (111) poles of original single crystal, and the rolled sheets from it, were measured. The two sets of data essentially resembled each other, with a little systematic angular slip in the planes.

The experiment showed that the sample essentially retained the initial texture, even after 90% reduction by rolling.

Sample II

The second copper sample was prepared by alternate annealing and cold working /13/ a 15.5 mm cube piece cut from a commercial polycrystalline copper ingot. The sample was annealed at 450°C for a few hours. Then the first step was to compress the billet alternately 10 to 20% in each of the three orthogonal directions and then to anneal it. The second step was to compress the sample in only two directions. Finally, the sample was worked only in one direction and then annealed. This process reduced the grain size of the sample by about half. The copper piece was then cold-rolled to a strip of 2 mm thickness corresponding to a reduction in thickness by 87%. Figure 7 shows the (200) pole figure of the sample. The sample was then further reduced in thickness to 1.0 mm to give a final reduction of 94% - Figs 8, 9 and 10 give the (200), (220) and (111)
FIG. 4. (200) pole figure of cold-rolled aluminium sheet (sample II).

FIG. 5. (220) pole figure of cold-rolled aluminium sheet (sample II).
FIG. 6. (111) pole figure of cold-rolled aluminium sheet (sample II).

FIG. 7. (200) pole figure of cold-rolled copper sheets (sample II, 87% reduction).
FIG. 8. (200) pole figure of cold-rolled copper sheet (sample II, 94% reduction).

FIG. 9. (220) pole figure of cold-rolled copper sheet (sample II, 94% reduction).
FIG. 10. (111) pole figure of cold-rolled copper sheet (sample II, 94% reduction).

pole figures of sample II. The comparison on intensities /1/ give the rolling texture as a superposition of the (110) [112] and (111) [211] textures. The 94% reduction sample gave sharper poles.

5. DISCUSSION AND CONCLUSION

Generally in f.c.c. metals (110) [112] texture superimposed on some secondary textures is observed. The texture of the first aluminium sample was found to be (200) [002], which is completely different from the predicted texture. Earlier, Eder and Klemencic /5/ observed (124) [211] texture for aluminium which is somewhat different from the standard texture. (200) [002] texture was not observed earlier in aluminium. However, in copper (200) [002] has been observed in a sample by Richards /2/. He measured textures of seven samples of copper with different original textures and grain sizes. All the samples changed to standard f.c.c. rolling textures except the one with an original (200) [002] recrystallization texture. Now, similar behaviour has been observed in aluminium. The work also showed that in the case of samples with large grain sizes and with strong original orientations, conventional rolling textures are not achieved.
ACKNOWLEDGEMENTS

We wish to acknowledge the Metals Advisory Service, Lahore, for rolling some of the samples. Technical support of the Neutron Diffraction Group staff is also acknowledged. This work was done under IAEA contract RR/1142 awarded to H.M. Butt.

REFERENCES

NEUTRON DIFFRACTION, MATERIALS SCIENCE

(Session XI)
Chairman

G. H. LANDER
United States of America
Invited Paper

NEUTRON DIFFRACTION STUDIES OF CHEMICAL STRUCTURE AND INTERACTION

M. S. LEHMANN
Institut Max von Laue - Paul Langevin,
Grenoble

Abstract

NEUTRON DIFFRACTION STUDIES OF CHEMICAL STRUCTURE AND INTERACTION.

The analysis of chemical structure, using neutron crystallography, is reviewed. First the various aspects of neutron diffraction are discussed and special advantages are outlined. Then various themes within the study of three-dimensional crystalline material are treated. The most traditional of these, the location of light atoms in structures, is shown to remain important, and likewise high-precision work still gives much new information. Within the last decade powder analysis has become a tool in chemical analysis. This is partly due to developments in monochromator technology, computer applications and large area detectors. For very similar reasons studies of materials in real time and under extreme conditions are becoming frequent. We therefore observe both a continuing use of traditional neutron crystallographic techniques, and a growth of new methods and areas which depend on instrumental developments. With the advent of new sources and larger detector systems further advances can thus be expected in the application of neutrons in structural chemistry.

INTRODUCTION

Crystallographic techniques probably offer the most direct way of investigating chemical structure. If suitable crystalline material is available, both molecular geometry and intermolecular arrangements can nowadays be obtained easily. The main drawback is that diffraction methods only give the time-averaged chemical structure for one or more equilibrium states, and do normally not supply any direct information on chemical reactions and pathways. Nevertheless, a large number of structural studies using diffraction methods form an important part of the framework upon which our knowledge of chemical structure and reactivity is built.

Following a long period of development of methods for solution of crystal structures and structure determination of a series of key organic, inorganic and biological molecules, the interests and efforts in crystallography have now also turned to the study of the behaviour of chemical structures under various conditions. Many parameters are available, ranging from "physical" variables such as temperature and pressure to "chemical" variables such as crystal growth conditions and chemical substitution. A similar development has been seen in neutron diffraction, being a part of
crystallography. In the early days an important effort was put into getting the structural information that was not easily obtained from X-ray analysis, i.e. the location of light atoms next to very heavy atoms. As part of this task was done, more emphasis was laid on the variation of environment conditions. It would be fair to say, though, that even in the early days of neutron scattering both furnaces and cryostats were often used, and that the precise location of light atoms still remains an important activity. Indeed, in very many cases the two intermix.

In this paper we shall try to outline these developments. They are of course related to the nature of the interactions between the neutron and matter, so in order to understand in which way the analysis of chemical structure can take advantage of neutron diffraction, we shall first briefly repeat and discuss the nature of these interactions. As other radiations - notably X-rays - are used for the analysis of chemical structure, and as the X-ray tube is easily available, a list implicitly becomes a comparison between a measurement done with neutrons and with X-rays. Keeping this in mind we can summarize:

The neutron scattering length is not a simple, monotonic function of the atomic number. This favours the observation of light atoms and other selected groups of atoms.

The scattering is a nuclear interaction, and the atomic form factor is a constant, the scattering length. This simplifies considerably the structure factor equation and its interpretation.

The spectral distribution of the beam can be tailored to the need of a given kind of experiment.

The neutron beam can be made homogeneous over a very large area. Samples up to several square centimeters large can be used.

The absorption of neutrons in most materials is low. This simplifies the construction of environment control units.

The neutrons can have magnetic interactions with matter. This is a large domain of research, but will only be discussed briefly here.

Altogether there is a whole series of points, where the employment of neutron beams in chemical structure analysis are favourable. In comparing with X-rays one might even conclude that these last are only better when a study of the detailed electron distribution in materials is needed, and that indeed the dominant use of X-rays is mainly due to their easy accessibility.

A series of books and reviews have been written on the application of neutrons in chemistry /1/, and the reader is referred to these for very extensive bibliographies.

In the following, the analysis of chemical structure has been divided into a number of themes, centred on various aspects of chemical interactions. These themes tend to follow the points outlined above for the various aspects of neutron interactions, but it seems that this parallelism is less pronounced for the themes of more recent occurrence such as powder studies. There are thus indications that we are still in the process of learning how to use neutrons in chemical research in an optimal manner.
In this review we do not discuss the dynamic aspects of chemistry that might be observed using inelastic neutron scattering, and likewise we do not discuss studies of surface interactions, although again these can tell us much about chemical structure and interactions. We shall therefore mainly review the chemistry that can be inferred from the study of structures and habits of three-dimensional crystalline material.

THE LOCATION OF LIGHT ATOMS (HYDROGEN) IN A MOLECULE FROM SINGLE CRYSTAL DATA

Although an ultimate goal in chemistry is to describe the molecular interactions using the complete electronic distributions, much can be learned from a rough knowledge of the atomic locations. The distance between two carbon atoms, for example, will instantly tell us whether the bond between the carbon atoms is single, double or triple, or even something in between, and the many structural studies available have both confirmed and extended our ideas on chemical bonding. The most labile atom is hydrogen, and this has been one of the most difficult to deal with. When hydrogen is bound to a valence-electron rich, and therefore easily polarizable, atom such as nitrogen, oxygen or fluorine, local chemical environment changes can easily introduce a bond-breaking followed by proton transfer. In general terms we call this proton acidity, and this phenomenon is intimately linked with aqueous solution chemistry. It is therefore not only because hydrogen is easier to observe with neutrons that many studies of hydrogen positions have been done, but the information sought is much needed.

In most of the cases where the hydrogen atom is labile, it is bound to several atoms in what we term hydrogen bonding, and we can learn much about the pathway and the mechanisms by determination of the precise interatomic distances. Many studies of this kind have been done /2,3/ and they can be generalized in relationships between the various distances as given in Fig. 1. Here, the length relationship in bonds of the type \( \text{N-H...O} \) is given. The scatter of points around an expected smooth curve comes partly from experimental error and partly from other molecular effects. These can be separated out, and via a study of the interatomic forces one can arrive at empirical potential functions /4/. Employing a reaction path picture one can likewise compare observed and calculated curves /5/. This work is still underway, and as theory develops we can expect more experimental data to be required.

If we now look at the hydrogen bonding to metal atoms the picture is much more complex. Figure 2 shows the metal-hydrogen bonding in \( \text{FeH}_2\text{Mg}^4\text{Br}_3.5\text{Cl}_{0.5}(\text{C}_4\text{H}_8\text{O})_8 \) where the shaded atoms are the metal-bound hydrogens /6/.

Up to now around fifty studies have been done /7/ and some trends emerge. This applies both to the metal-hydrogen bond lengths as a function of the hydrogen site in the cluster as well as the cluster size relationship to the number of hydrogen atoms. This
FIG. 1. Relationship between bond length N-H and H...O in N-H...O hydrogen bonds. Only N-H involving N being sp hybridized is given.

again has implications on the behaviour of metals upon hydrogen absorption. Altogether, these systems are very complex, and even though general principles are being laid down, there is a need for further studies. In many cases an additional problem is the small crystal sizes available, which indicate that higher flux and larger detector systems are needed.
Similar requirements for high flux and large detectors are found for the analysis of macromolecules at atomic resolution. Several reasons for these studies can be identified. Most macromolecular chemistry takes place in aqueous solutions, and water is essential for the correct confirmation of the macromolecule. A few studies are now available /8/, but we are far from any general picture. As mentioned above, many hydrogen atoms are labile, and their exchange rates give information about the flexibility of the macromolecule /9/. Neutron diffraction can be used for these studies owing to the large difference in scattering length between hydrogen and deuterium. If we leave a macromolecule in D_2O the amount of deuterium found at a given site is related to the exchange ability for this atom. Deuterium, which gives a higher signal than hydrogen, can also be used to identify better the individual hydrogen atoms which are of crucial importance, for example for the enzyme mechanism /10/. An example is given in Fig. 3 for the enzyme lysozyme /11/. The scattering density in the plane of the carboxyl group of residue 35, glutamic acid, is shown. The presence of a hydrogen (deuterium) 35DE2 at the carboxyl group is in agreement with the proposed enzyme mechanism. Deuteration can be carried further, for example in studies of small-molecule interaction with a macromolecule /12/. If the small molecule is deuterated then its signal would be considerably higher than in the similar X-ray case.

Finally, one should note that neutrons are equally well used to distinguish neighbouring atoms in the periodic system, where X-rays might fail, and this kind of study occurs regularly.
The most serious limitation of neutron diffraction studies is often the low neutron flux leading to a weak signal. Even then one observes in very many cases that the quality of the results is better for neutrons than for X-rays. The reason for this is favourable experimental conditions. The cross-section of the homogeneous beam can be made very large, ensuring that for all orientations the crystal is exposed to the same flux. This is often not true in the X-ray case. For crystal monochromatized X-rays the full width at half height of the homogeneous beam in one direction is only ≈ 0.8 mm /13/. Moreover, the spectral shape of the neutron beam can be tailored to needs, and this is essential when integrating a Bragg reflection. In this process we have to decide the range in the scanning angle over which to integrate, and we have to fix the background. If the beam contains long-tail components (Lorentzian-type spectrum) as in the X-ray case, it is difficult to obtain an objective result. For neutrons one normally has Gaussian functions and even a triangular function can be obtained.

Other effects, such as absorption, are in a majority of the cases handled very easily, where this is not so for heavy atoms and X-rays, and problems occurring with secondary extinction and thermal diffuse scattering are much the same in the two cases. When we finally arrive at the stage of interpretation, again neutron data are simpler to deal with. The structure factor equation as normally used is given by

$$F(h) = \sum f_j \exp(2\pi i h \cdot r_j) T_j$$

where the sum is over atoms at positions $r_j$, and $T_j$ is the Debye-Waller factor, which describes the time-averaged distribution of atom $j$ around its equilibrium position. If the form factor $f_j$ is a function of $h$, as in the case of X-ray data, the $f_j$ and $T_j$ will correlate, and interpretation of $T_j$ will depend on the assumptions made for $f_j$. For neutron data $f_j$ is constant, and our interpretation of the scattering in terms of mean atom positions and atomic distributions is unbiased.

The Debye-Waller factor is normally assumed to describe a harmonic motion, but this would clearly not be sufficient in most cases. An example from KH$_2$PO$_4$ studied 2K above the phase-transition (Fig. 4) shows the disordered proton distribution to be as that obtained from a Fourier density map /14/. In this case a more advanced description is needed, and this can be obtained by adding terms to the structure factor description /15/. This then allows one to get an idea of the various vibrational and librational modes in the crystal, without resorting to spectroscopic studies, and to get a more precise estimate of the atomic equilibrium position. Much work has been devoted to this kind of analysis /16/, giving information ranging from the behaviour of the Debye-Waller factor in metals and simple inorganics as a function of temperature, to the determination of torsional frequencies for methyl groups in complex organic compounds.
FIG. 4. Section through the neutron scattering density along the O...H...O bond in KH$_2$PO$_4$. Only the H distribution is given.

FIG. 5. Electron deformation density section for H$_2$O$_2$ (X-N map). The plane contains two oxygen and one hydrogen atom. Hydrogen bonds are indicated with dotted lines.

The interatomic distances of high precision obtained from neutron diffraction studies at very low temperatures have recently been used to compare with molecular structures as obtained from advanced quantum chemical ab-initio calculations /17/. Reasonable agreement can be obtained and one can certainly expect further studies of this kind.

The combined used of high precision positions and thermal motion is common when the charge density of a molecule is studied from X-ray measurements. For the reasons given above neither the co-ordinates nor the thermal motion are reliably determined from X-rays, and neutron results are therefore used for the theoretical
calculations of electron densities. An example of such a calculation is given for hydrogen peroxide /18/ in Fig. 5. This shows the difference between the observed electron density and the density if the atoms had spherical electron density distribution. The lack of deformation charge accumulation in the O-O bond can be explained by evoking the Pauli exclusion principle /18/. When two electron-rich atoms bind, the charge rearrangement moves electrons away from the overlap region. Although in this case the neutron data mainly serve as a reference, the fact of having an independent precise measure of atomic configuration does not only help in the electron density analysis but also advances the general understanding and improvement of the diffraction data.

HIGH-RESOLUTION POWDER ANALYSIS

For many compounds single crystals are not available. The reason for this is most often that large crystals cannot be grown, but it also happens that crystals break when going through phase transitions. In this case the only access to a structure analysis is via a measurement on powders. This has undoubtedly biased our view of structural chemistry, as some classes of compounds have not had the attention they deserve. Over the last decade this has changed owing mainly to the development of new analysis techniques using high-resolution powder diffraction data.

Similar to the case of high precision the progress is based on several aspects of neutron technology. As it is impossible, except for extremely simple compounds, to separate completely the individual powder lines, it is necessary to unravel the spectrum by some profile fit technique that allows for extensive overlap of Bragg reflections. This requires a simple and well-defined spectral shape, and this can easily be produced with neutron beams. Adding to this the low absorption coefficient for most materials, environment control units are relatively easily constructed. With a refinement method /19/ and some convincing examples of application /20/ the method of neutron profile refinement developed quickly, and attained widespread use.

The powder techniques are being used in all areas of chemical crystallography, where the number of parameters in the structural problem is small. There is therefore no reason to enumerate the various subjects, except for a general comment. The neutron powder refinement techniques are used by a much wider group of scientists than are normally doing neutron scattering, in a manner similar to the way spectroscopic methods and routine X-ray structure solutions are employed for chemical analysis. The subjects therefore reflect at any time both the issues of interest for the chemical community as well as the industrial technical problems, that are tractable by structural analysis.

At present the number of variable parameters in an analysis is one to two hundred. New instruments are being built that double the resolution and, in favourable cases, this should lead to the solution of problems containing 500 parameters. This of course
requires the observation of an approximately similar number of peaks, which are not individual Bragg reflections. With the advent of the new sources, where short wavelength neutrons are available, such spectra can be recorded if the crystals are of sufficient quality to give a diffraction pattern at high momentum transfers.

In most cases powder neutron work is not aimed at crystal structure solutions, but rather at observations of changes when chemical or physical parameters are varied. The profile refinement techniques can, however, be used to extract intensities /21/, and then standard crystallographic structure solution techniques can be employed. Some attempts have already been made, and based on results of these and model calculations /22/, it has been estimated that structures with up to twenty independent atoms in a centrosymmetric space-group can be resolved. A limiting factor, apart from instrumental resolution, is undoubtedly the quality and size of the crystallites involved. If single crystals cannot be made large enough to carry out an X-ray analysis, they are necessarily in the micron size. Particle broadening effects can then become important. Even then there will still be many structures that can be handled with these methods.

LOW-RESOLUTION POWDER DIFFRACTION AND SOME REAL CHEMISTRY IN REAL TIME

Structure changes are mainly studied as a function of temperature and pressure. Other variables are just as interesting but often more difficult to handle. Changes of structure in time, i.e. chemical reactions, can be studied if the time constants are sufficiently large. Measurements can then be done using multidetectors, and at present several minutes are required for each recording. Often the work is done on powders. In this way a range of Bragg reflections can be monitored simultaneously. In most cases no full structure analysis is done at each point of the reaction, although this might be possible.

To get a maximum of flux the requirements on wavelength and angle resolution are normally reduced, and the reaction is then followed from a selected number of reflections. The first model system to investigate was the layered dichalcogenide $2H$-$TaS_2$ and the reaction products obtained from electrochemical reduction in $K_2SO_4/D_2O$ solutions /23/. In this case the intercalation of $K^+$ and $D_2O$ was followed from variation of d-spacings and intensities of the layer-reflections of the $2H$-$TaS_2$ with time. Later, similar kinds of studies were done on the deuteropyridine intercalation in the same compounds. The time scale for these experiments is some hours, so with the new generation of large-area detector powder diffractometers, reactions taking less than an hour to completion are certainly feasible.

Growth of crystals or change of crystal modification can equally well be followed in real time. In this case no direct structure information is obtained, and the main interest is to monitor the amount of the various components in the system. This
is done by following the intensity of powder lines belonging to the different components. Examples of this is the hydrothermal growth of iron oxides /24/ and the hydration of compounds /24/. An example is given in Fig. 6 /26/. The powder spectrum as a function of time is given for the reaction CaSO\(_4\cdot\frac{1}{2}\)D\(_2\)O + D\(_2\)O = CaSO\(_4\cdot2\)D\(_2\)O (gypsum). The reaction takes place over a period of about one hour.

Other experiments are of a similar nature, using different variables. If a very complicated set of phase transitions occur as a function of, for example, temperature, then similar techniques can be used to survey the range of possible structures. Low-resolution neutron powder studies have likewise been used to study the chemisorption on surfaces, and an interesting example of a surface chemical reaction is the study of the electro-chemical passivation of Ni powder /27/. By following the (111) and (200) reflection of Ni as the potential of a pressed Ni powder in a 0.05 M Na\(_2\)SO\(_4\) + 0.5 M K\(_2\)SO\(_4\) solution was varied, it was shown that about 2% of the Ni atoms were reversibly oxidized to Ni\(_2\)O\(_3\).

These types of experiment will probably not become as common as the powder refinement analysis or the location of light atoms, where neutrons have obtained a status as a routine analysis. On the other hand, because of large beams and high transmission factors, there will be a continuous demand for neutrons to do this kind of experiment, especially as larger multi-detector units become available.

**SPIN-DENSITY**

Information about unpaired electrons can be obtained from the study of magnetic materials, using polarized neutron beams. Much work has been done, but the number of analyses of more direct "chemical" interest is still limited. These studies have been aimed
at studying the distribution of unpaired 2p electrons as found in N-O and N-N radicals /28/, and the spin population and covalency in 3d transition metal compounds /29/.

The structure factors for the magnetic components are obtained from the ratio between the intensity of the scattered neutrons using a beam with neutron spin up and with spin down. Eventually they are dependent on the value and quality of the nuclear structure factor. Moreover, as the measurement times are rather long, full three-dimensional data sets are normally not collected. The modelling of the unpaired electron distribution is therefore sometimes done via a least-squares fit using a multipole expansion of the density. An example is given in Fig. 7 for the di(2,2,6,6-tetramethyl-4-piperidinyl-1-oxyl)-suberate /30/. This is the unpaired spin density projected on to a plane orthogonal to the plane of the molecule, showing clearly the distribution of the 2p(non-bonding) orbital. In this case there is an even distribution of unpaired electrons on two atoms of the N-O radical.

As the magnetic scattering techniques do probe specific electrons, which can be of chemical interest, these methods are potentially very useful, and we can expect more work of this kind as the measurement method improves, and as neutron fluxes increase.

CONCLUDING REMARKS

From the present review several trends appear.

There is obviously still a need for the location of hydrogen atoms, but the tendency is to study very large systems such as metal-coordination complexes and biological macromolecules. There
is likewise a similar strong interest in precision studies, both as a support for the analysis of electron density distribution, and as a means to learn more about interatomic potential function from the distribution of the nuclei.

Analysis of simpler systems that can be managed by powder analysis, is of steadily growing importance, and also measurements of structural changes in real-time studies would attract much interest.

A common factor for all these themes is that they have developed because of improvements in measurement and computer technology. For the very large structures only multidetectors can give an acceptable data collection rate, and for the powder work improvements in monochromator technology has been of much help. In a similar manner real-time studies would not be feasible if multidetectors were not available.

Altogether, we can therefore conclude that much of the advance is coming from increases in effective count-rate, and that these have inspired new ways of using neutrons. Consequently, with further development of both sources and equipment we can expect new fields of research to occur, although we cannot of course predict where and when.

REFERENCES


/5/ BORGI, H.R., Angew. Chem. 87 (1975) 461.


/29/ FIGGIS, B., ibid, p. 17.
Invited Paper

SINGLE-CRYSTAL DIFFRACTOMETRY AT THE PROPOSED PULSED NEUTRON SOURCE SNQ*

W. JAUCH, H. DACHS
Hahn-Meitner-Institut für Kernforschung, Berlin (West)

Abstract

SINGLE-CRYSTAL DIFFRACTOMETRY AT THE PROPOSED PULSED NEUTRON SOURCE SNQ.

The performance of single crystal time-of-flight diffraction at the proposed pulsed neutron source SNQ (time-averaged flux $\bar{\phi} = 1.2 \times 10^{15}$ n/s$^{-1}$cm$^{-2}$) is compared with the conventional monochromatic technique at a steady-state reactor of equal thermal flux. The comparison is based on the total counting time necessary to obtain a sufficiently accurate data set. Applications in different fields are treated: (i) Structure analysis of biological macromolecules at very high resolution; (ii) high-precision studies of positional parameters for small unit cells; (iii) polarized neutron diffraction to determine precise magnetization densities.

* This paper was based on a more detailed presentation, the text of which is to be found in Neutron Scattering Instrumentation for SNQ, Proc. Workshop, Maria Laach, 3–5 Sep. 1984, Rep. Jül-1954, KFA-Jülich (Oct. 1984).
HIGH-RESOLUTION POWDER DIFFRACTION

D. E. COX
Physics Department, Brookhaven National Laboratory,
Upton, New York,
United States of America

Abstract

HIGH-RESOLUTION POWDER DIFFRACTION.

In the past decade there has been a dramatic increase in the application of neutron powder diffraction data to structural analysis, particularly in many areas of solid-state chemistry and materials research. This increase has been accompanied by the growth of a large user community, and can be attributed mainly to the introduction of the Rietveld technique of refinement, which has proved to be a very powerful structural tool. An important factor in this progress has been the availability of high-resolution neutron data, and a brief description is given of the capabilities of present-day instruments and also the two next-generation instruments nearing completion of the Rutherford Appleton Laboratory and the Institute Max von Laue - Paul Langevin (ILL). Some representative examples of structural investigations in the area of materials research are listed, with specific reference to the rhombohedral phase of Na-A zeolite, which represents a difficult problem to solve with present-day techniques. Emphasis is placed on the importance of a proper description of the peak shapes which accounts for sample effects such as particle size and strain broadening in a physically plausible way rather than by empirical parametrization. Finally, some likely future developments are summarized.

INTRODUCTION

In the decade which has elapsed since Rietveld introduced his method for profile refinement of neutron powder data [1] there has been a dramatic increase in the application of powder diffraction techniques to structural analysis. In a 1977 review article, Cheetham and Taylor [2] listed almost 200 examples of structure refinements of this type (now generally known as "Rietveld refinement" to avoid confusion with traditional methods of individual line profile analysis), and by now the number must have grown to well over 1000. More recently, encouraging progress has been reported in the refinement of x-ray powder data which can be expected to further accelerate this growth.

Not only has the Rietveld technique become established as a powerful structural tool in its own right, but also its widespread application to neutron data has created a very large and
diverse user community, many of whom are concerned with the characterization of materials with useful properties which are often related to their polycrystalline nature such as catalysts. This community is very well aware of the unique role of neutrons in structural investigations and may be expected to play an active part in the development of new facilities in the next decade. One of the principal requirements is likely to be improved resolution, since it is this which ultimately sets a limit to the complexity of a structure which can be handled.

In recent years, the demand for time on high resolution instruments has far outstripped supply, and this situation appears likely to continue, particularly since the user community shows no sign of reaching a saturation level yet. Increasing demand may also be anticipated for instruments optimized for high intensity rather than high resolution for time-resolved studies of chemical, ceramic, and metallurgical processes. This aspect is also an important one, but beyond the scope of the present paper.

INSTRUMENTATION

The major application of high resolution powder diffraction has been to structural studies of inorganic compounds, and this has greatly influenced the design features for optimum performance. For constant wavelength (CW) instruments operating in the step-scan mode, these have been discussed in detail by Hewat [3], the essential point being that the minimum in the angular (2θ) resolution should be at about 90°, where the density of peaks is highest. This is achieved by the use of a monochromator set to diffract at a Bragg angle θM of about 60°. High resolution is obtained with appropriate combinations of high-efficiency Soller slits, and intensity is optimized with the use of wide vertical apertures, a large, vertically focussing monochromator with appropriate mosaic width, and as many counters as practical mounted on the scattering arm. The resolution Δd/d of such an instrument as a function of Q is fairly well matched to the density of peaks in the pattern and has a minimum around Q = 4πsinθM/λ, i.e. quite close to Qmax, which is about 8Å⁻¹ for a typical wavelength of 1.5Å. This minimum value (Δd/d)min therefore represents a reasonable index of performance for structure refinement. In the case of D1A at the Institute Laue Langevin (ILL), (Δd/d)min is about 2x10⁻⁴ [4], a figure matched by the recently commissioned HRPD at Lucas Heights.[5]

An alternative to a multicounter detector operating in the step-scanning mode is a linear position sensitive detector, and
two fairly high resolution instruments of this type have recently been described, one at KFA, Jülich using a \(^{6}\)Li scintillation detector [6] and the other at the University of Missouri, Columbia using a \(^{3}\)He gas-filled detector [7]. The design features for optimum performance are very similar to those described above except that the resolution after the sample is governed by the size of the sample, the spatial resolution of the detector and the distance between them instead of the divergence of a Soller collimator. In both cases, a \((\Delta d/d)_{\text{min}}\) of about \(5 \times 10^{-3}\) is achieved.

In the past few years the Rietveld technique has also been very successfully applied to high resolution time-of-flight (TOF) data obtained at pulsed neutron sources, and the corresponding design features have been described in detail by Jorgensen and Rotella [8]. In this case the important considerations are the time distribution of the neutron pulse, the length of the flight path and the sample and detector geometry, both time and geometrical contributions to \(\Delta d/d\) being essentially independent of wavelength. Optimization for high resolution is relatively straightforward, involving a suitable combination of short pulse width, long flight path and a high scattering angle, \(\Delta d/d\) for the two high angle detector banks on the instruments SEPD and GPPD at IPNS, Argonne, being about \(3.5 \times 10^{-3}\) and \(2 \times 10^{-3}\) respectively.

Within the next year, the commissioning of two next-generation high resolution instruments will undoubtedly have a major impact on neutron powder structural analysis. One of these is a TOF instrument being installed at SNS at the Rutherford Appleton Laboratory [9], and the other is a CW instrument (D2B) at the reactor face at ILL [10]. Both of these have been designed to give optimum performance with \(\Delta d/d\), or \((\Delta d/d)_{\text{min}}\), equal to about \(5 \times 10^{-4}\), a factor of at least four better than present day instruments. In addition, upgrading of existing diffractometers and planning and construction of new ones is underway at a number of other neutron centers.

**STRUCTURAL STUDIES**

It is impossible to make any comprehensive summary of the enormous range of structural problems tackled by Rietveld analysis. Instead a few recent examples are given in Table 1 of systems which are of interest from a materials viewpoint, where the inter-relation between structure and physical and chemical properties is the primary motivation. Generally, not only are single crystals of these materials unavailable, but also the
Some recent examples of Rietveld refinement in materials research

<table>
<thead>
<tr>
<th>Application</th>
<th>Compound</th>
<th>Space Group</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Superconductivity</td>
<td>YbMo$_6$S$_8$</td>
<td>R$_3$</td>
<td>[11]</td>
</tr>
<tr>
<td>Fast ion conductivity</td>
<td>Na$_3$Sc$_2$(PO$_4$)$_3$</td>
<td>Cc(293K)</td>
<td>[12]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>R$_3$c(&gt;317K)</td>
<td></td>
</tr>
<tr>
<td>Magnetism</td>
<td>Nd$<em>2$Fe$</em>{14}$B</td>
<td>P$_4$2$_1$mm</td>
<td>[13]</td>
</tr>
<tr>
<td>Hydrogen Storage</td>
<td>γ-FeTiD</td>
<td>P2/m</td>
<td>[14]</td>
</tr>
<tr>
<td>Catalysis</td>
<td>Na-A Zeolite</td>
<td>Fm3c</td>
<td>[16,17]</td>
</tr>
<tr>
<td></td>
<td>(NaAlSiO$_4$)</td>
<td>R$_3$c?(&lt;335K)</td>
<td></td>
</tr>
</tbody>
</table>

Properties of interest may be critically dependent on the polycrystalline nature of the material, as example in zeolites, which are an important class of industrial catalysts. The Rietveld technique has proved to be particularly valuable in studies of this type.

The structure of Na-A zeolite provides an interesting example of the application of the Rietveld technique. X-ray single crystal studies showed this to be cubic, space group Fm3c, with the Si and Al ions completely ordered.[18] Following this, three independent neutron studies were described in close succession. The first reported a slight rhombohedral distortion at 290K [15], the second a cubic structure in agreement with the x-ray results [16], and the third a rhombohedral-to-cubic transition at 335K [17]. This transition was interpreted in terms of partial ordering of one set of Na ions, with a consequent lowering of the symmetry to rhombohedral, space group R$_3$c, involving a large increase in the number of structural parameters. A model of this type, but with cubic constraints on the parameters, gave a satisfactory profile fit, as can be seen by inspection of the original data in Figs. 7 and 8 of reference [17]. It seems likely that the Si:Al ratio and the residual H$_2$O content play an important role in determining the structure, but the small distortion, the degree of peak overlap and the number of variable parameters in the rhombohedral phase make this a difficult problem to solve at present day levels of resolution.
The importance of modelling peak shapes properly has been emphasized by many authors, and the current thrust towards higher resolution has focussed more attention on how these shapes are modified by sample effects, such as particle size and strain broadening, and instrumental effects, such as vertical divergence. For example, application of the Scherrer formula in the form $\Delta d/d = d/L$ shows that a mean particle size, $L$, of 0.1 $\mu$m gives a $\Delta d/d$ of around $10^{-3}$, which should be readily visible on a high resolution instrument. If the line profile shape is assumed to be Lorentzian, as often appears to be the case, the instrumental peak shape is appreciably modified. Although this is a familiar problem in the refinement of x-ray powder data [19], which has significantly hindered the application of the Rietveld technique, only recently have systematic attempts been made to allow for such effects in refinement of neutron data. [20-25] For CW instruments, where the instrumental peak shape closely approximates a Gaussian function, there is a lot to be said for the use of the Voigt function (a numerical convolution of Lorentzian and Gaussian functions) or an approximation to this, the "pseudo-Voigt" function, which is a simple linear combination of the two having the form [26]

$$I(\Delta 2\theta) \propto \eta L(\Delta 2\theta, \Gamma) + (1-\eta)G(\Delta 2\theta, \Gamma)$$

$\Delta 2\theta$ and $\Gamma$ are the displacement from the peak and its FWHM respectively, and $\eta$ is a "mixing" parameter. However, although $\eta$ is a convenient parameter to incorporate in the Rietveld code, it has little physical significance per se, and no simple dependence on $2\theta$ over an extended range. Instead of some empirical parametric form for $\eta$, it is clearly desirable to incorporate sample effects in more explicit form through the convolution of Gaussian and Lorentzian components of the type

$$\Gamma_G = (U \tan^2 \theta + V \tan \theta + W)^{1/2}$$

$$\Gamma_L = X \tan \theta + Y/\cos \theta$$

where $X$ and $Y$ are strain and particle size parameters respectively. This approach has yielded very satisfactory preliminary results, and appears to be equally valid for both conventional and synchrotron x-ray data [27].

FUTURE DEVELOPMENTS

There seems little doubt that the next decade will witness a steady growth in the application of the Rietveld technique to
structural analysis from neutron powder data. Together with this will be a general trend towards the optimization of present instruments through improved neutron optics, monochromators, and more efficient collimators and detectors. In particular, the new generation of high resolution instruments will have a very significant impact, not only by greatly extending the ability to refine complicated structures, containing perhaps 200-300 positional parameters, but also by opening up the possibility of solving completely unknown structures by direct methods, hitherto a single crystal technique.

More emphasis is likely to be placed on structural changes as a function of some external variable such as temperature, pressure, magnetic field or chemical environment. Increased rates of data collection will permit in situ studies of such changes in favorable cases. High resolution data are also likely to find increasing application to other types of investigation, such as indexing of unknown phases, line broadening, phase transitions, and stress and texture measurements.

Considerable effort will need to be devoted to improvements in software, including a more sophisticated approach to the peak shape problem with respect to instrumental aberrations and sample effects, possibly anisotropic in nature. More reliable criteria for assessing the accuracy of the results and what constitutes a significant improvement will be required. Extension of the Rietveld technique in a systematic way to more generalized models of structural and thermal disorder, and incorporation of distance and angle constraints is yet another area for development.

Last but not least, the development of high resolution x-ray powder techniques ($\Delta d/d \leq 10^{-4}$) at synchrotron facilities[28] is sure to stimulate considerable activity, which in some cases will be a challenge but in general more likely to be complimentary to neutron techniques.

This work is supported by the Division of Materials Sciences, U. S. Department of Energy, under contract DE-AC02-76CH00016.

REFERENCES

Invited Paper

NEUTRONS FOR MATERIALS SCIENCE

C. G. WINDSOR, A. J. ALLEN, M. T. HUTCHINGS,
C. M. SAYERS, R. N. SINCLAIR, P. SCHOFIELD, C. J. WRIGHT
Materials Physics and Metallurgy Division,
Atomic Energy Research Establishment,
Harwell, Didcot, Oxon,
United Kingdom

Abstract

NEUTRONS FOR MATERIALS SCIENCE.

The discussion will be limited to applied materials research performed on a customer/contractor basis. The information obtained using neutrons must therefore compete both scientifically and financially with information obtained using other techniques, particular electron microscopy, X-ray, NMR, infra-red and Raman spectroscopy. It will be argued that the unique nature of the information gained from neutrons often outweighs the undoubted difficulties of access to neutron beams. Small-angle scattering has emerged as the neutron technique of widest application in applied materials research. The penetration of neutron beams through containment vessels, as well as through the sample, allows the measurement of in situ time-dependent experiments within a furnace, cryostat, pressure vessel or chemical reactor vessel. Examples will be given of small-angle scattering projects from the nuclear metallurgy, coal, oil, cement, detergent and plastics industries. High-resolution powder diffraction is another technique with wide applications. Structural studies are possible on increasingly complex phases. The structure and volume fraction of minority phase can be measured at levels appreciably below that possible by X-ray diffraction. A rapidly growing field at present is the measurement of internal strains through the small shifts in lattice spacing. Neutron diffraction is unique in being able to measure the full strain tensor from a specified volume within a bulk specimen. Inelastic scattering measurements exploit the unique property of neutrons to measure the orientations of vibrating molecules. Examples will be chosen from the field of catalysis where inelastic spectroscopy has revealed the nature of the bonding of hydrocarbon molecules.

1. THE SCOPE OF THIS ARTICLE

Materials science covers a wide field from aerosols to zeolites. This article will be limited to applied materials science in the sense that someone has commissioned the research and will be willing to pay the market price for it. This "contract research" has long been a speciality of the Harwell laboratory in England. The first steps to enter this field using neutron scattering were taken in the late sixties by Dr Peter Egelstaff. One of his maxims was "never do anything with neutrons that can be done in any other way!". He saw
that the unique properties of a neutron beam could allow a flourishing applied neutron programme even within this maxim. In June 1970 Dr Egelstaff had a historic meeting with heads of industrial research departments. They agreed in predicting a growth in applied neutron beam expenditure which would match the academic programme in the UK at that time (£400K in 1970 prices). They saw neutrons as following in the long line of physical techniques, the electron microscope, X-rays, nuclear magnetic resonance (NMR) and Raman infra-red spectroscopy, which were discovered and first used by physicists in the universities. These techniques spread first to scientists of other disciplines, and then to almost every important laboratory to become the universal tools of materials science they are today. Although this prediction failed to take into account the vast increase in spending on new neutron sources and instrumentation since then, a number of laboratories, uncluding Harwell in the UK, are finding an increasing number of industries which are seeing the need to fund neutron beam research as part of their R & D programmes. In this paper, we indicate some of the areas and some of the problems which are covered.

2. THE USE OF NEUTRON BEAMS BY INDUSTRY

The uses of neutron beams in materials science are complementary to the very techniques we mentioned in the last paragraph; the electron microscope, X-rays, NMR and infra-red and Raman spectroscopy. These techniques are commonplace in every industrial laboratory. This multiplicity may be expensive and inefficient — those instruments cost much the same as neutron spectrometers and they probably stand idle for two thirds of the day — something abhorrent to the neutron beam scientist. However it does mean that if an industrialist wants to measure a precipitate diameter in an alloy he can realistically know the answer in a day or two. Consider the procedure if he requires to do a neutron beam experiment. First he must search for possible centres where he might do his research. The list is quite long on a world scale but very short on a national scale. Next he must contact someone at the reactor and ask for some beam time. He will receive a spectrum of replies. At one extreme the neutrons are free, the schedule is completely flexible, but some one person controls it and his chances of getting the time depend on the interest of this person in his problem. At the other extreme is Harwell. A paying customer has priority over any other use of the neutron beam. Schedules will be changed for an urgent experiment.

Having obtained an allocation of beam time, the investigator has to perform the experiment. Back home in his laboratory he will probably have to hand technicians who will take his sample and run it for him. In most neutron centres he will have to do this himself. Some technicians may be to hand to help if there
is some fault condition but generally "helping users" is not their job! Harwell scientists and technical support staff have long been trained to put the users first! They may be contracted to perform, analyse and write up any experiment as required.

3. UNIQUE CHARACTERISTICS OF NEUTRON BEAMS FOR MATERIALS SCIENCE

3.1 Penetration

Let us return to our list of the rival techniques to neutron scattering mentioned in both previous paragraphs. Electron microscopy, X-rays, NMR, infra-red and Raman spectroscopy all have the disadvantage for many materials studies, certainly for metals studies, of being surface sensitive. Neutrons are unique in observing bulk average properties on the scale of centimetres. This means that interior properties of components can be studied without careful cutting procedures.

The neutron low absorption makes sample environment control a much easier and broader field than is possible with competing materials research techniques. "In situ" experiments within furnaces, cryostats, pressure cells and chemical reaction vessels are all routine in the neutron field.

3.2 Isotopic substitution

The rival techniques using electrons or X-rays are sensitive to the electrons in the sample and not to the nuclear distribution. In fact the vast majority of materials problems are insensitive to its nuclear isotopes. This makes neutron scattering from isotopic samples a uniquely useful technique for identifying effects from a particular nucleus. In particular hydrogen with its strong scattering power not shared by deuterium can be picked out selectively in radiography and in vibrational spectroscopy. In small angle scattering the ability to vary the contrast of a defect, by changing the isotopes in the defect or its surrounding matrix, gives unique information. An important example is the extensive use of H₂O/D₂O mixtures to identify macromolecules in aqueous media.

3.3 The applicability of the first Born approximation

What more esoteric reason could you give to an industrialist for using neutrons! What it means is that neutron cross-sections can be routinely measured on an absolute scale and calculated absolutely by simple single scattering theory [1]. In contrast, electron diffraction intensities are difficult to calculate without extensive computer codes. Infra-red and Raman vibrational mode intensities depend on electric dipole and polarizability transition probabilities. Both are much more difficult to evaluate than the simple vibrational amplitude transition probability of neutron scattering.
3.4 Neutrons the multi-technique

Neutron beams combine features from many techniques. Defect detection from electron microscopy, diffraction from X-rays, diffusion from NMR and vibrational spectroscopy from Infra-red and Raman methods. Often this feature is irrelevant - you only book time on one instrument! However the simultaneous use of several neutron techniques is possible and could be uniquely useful during for example in-situ heat treatments on a precious sample. Multi-purpose instruments are particularly suited to a pulsed neutron beam. Figure 1 shows an update of the "metallurgical facility" proposed for the Harwell linac in 1978. A similar multi-facility would be possible on a reactor guide tube.

4. SMALL ANGLE NEUTRON SCATTERING (SANS)

In the last decade SANS has emerged as the neutron beam technique of widest application to materials science - certainly it is the one most easy to sell. SANS measures the scattering from inhomogeneities in the material. They scatter strongly at low angles so that run times are measured in minutes or hours rather than the hours or days typical of most neutron experiments. Its problem is that a user cannot deduce how many types of defect he is looking at simultaneously. For this reason SANS needs to be supplemented by other techniques, particularly analytic electron microscopy which can usually give the principal defect in a given size range.
4.1 Copper clustering in steels: the electrical industry

SANS becomes particularly useful for defects too small to be resolved by conventional microscopy. The problem of copper clustering arises from the parts per million of copper arising in steels from the copper wires in scrap cars sometimes added during the steel making process. In the absence of radiation this copper remains stable at the operating temperatures of a reactor pressure vessel. However radiation has the effect of "shaking down" alloys to their most stable thermodynamic state. In this case it induces aggregation of copper atoms to form very small 20 \AA diameter copper rich clusters which can cause embrittlement of the steel. Figure 2 shows the amount of clustering as measured by SANS as a function of copper concentration by Jones and Buswell [2]. The effect is clear, as is its solution which is simply to specify a sufficiently low copper content of the steel.

4.2 Porosity: the coal industry

The microporosity of coals is of great importance to the industry particularly for direct conversion to gas processes. It is one of the few quantitative measures of coal quality. The standard technique for measuring coal porosity and surface area is by nitrogen adsorption. However this requires the coal to be degassed by evacuation before examination and is insensitive to closed pores. X-ray small angle scattering suffers from the fact that the contrast from pores (air or water filled) is relatively small.
FIG. 3. The size distribution of pores in coals [3].

compared with that from several mineral inclusions. The neutron contrast between coals and pores is large and comparable for either air or water filled pores. The contrast from most minerals in coals is small. Figure 3 is from the work of the Tricker et al [3] showing the volume weighted mesopore size distribution for several coals. The anthracite coals have a low angle scattering and a broad pore size distribution extending from 100 Å to 300 Å diameter. They have a high carbon content or "rank". Lower rank coals such as subbituminous and lignite have pores of around 60 Å diameter. General agreement between the nitrogen adsorption method and SANS was obtained providing a firm foundation for using SANS to probe porosity when conventional methods cannot be used.

4.3 Porosity: the oil industry

The porosity of oil-bearing rocks is of importance in estimating the migration of hydrocarbons and aqueous liquids under capillary and hydrostatic pressure. The standard techniques of nitrogen adsorption and mercury intrusion give variable results for the smaller pores with diameters below 50 Å or so. Transmission electron microscopy is a vital tool for measuring pore
shapes but is not a straightforward method for pore size distributions. In practice neutron and X-ray small angle scattering provide complementary inputs to the measurement of pore size distributions. For neutrons the oil to rock contrast is close to that of void to rock. For X-rays the oil to rock contrast is low but that to void to rock is large. The two techniques together therefore give information on the three component system of oil and void pores in rock. Figure 4 shows measurements taken on the Missouri Research Reactor by Hall, Mildner and Borst [4] on the cumulative pore size distribution from a horizontal cut of East Kentucky oil shale. Neutrons can in these shales observe the anisotropy from the flattening of the pores under natural compression.

4.4 Porosity: the cement industry

Snails' shells and ordinary Portland cement (OPC) are both silicate networks. Why are the shells so much stronger. The answer lies in their defect structure. Birchall, Howard and Kendall [5] added polymer gel to the cement. This produces macro-defect-free (MDF) cements which can take a strain of $4 \times 10^{-3}$ giving properties more like a plastic. The porosity of both OPC and MDF cements can be measured by neutron scattering without the drying pretreatment necessary in most other porosity measurements. Figure 5 shows the quite different pore size distributions measured for the two types [6].
4.5 Surfactant structure: the detergent industry

Detergents are precisely structured surfactants. Their job is to take globules of grease and disperse them in stable droplets surrounded by an ordered layer of the surfactant. In this field $H_2/D_2O$ contrast can be of enormous importance. Dispersed in $H_2O$ the SANS from a selection of products looks very similar. Dispersed in $D_2O$ the structures reveal their distinctive shapes [7].

5. NEUTRON DIFFRACTION

A diffractometer would certainly appear in any list of neutron instruments for applied materials research. As this section shows, one diffractometer can perform a number of roles provided its resolution is high. Essentially different information is provided by diffraction peak positions, intensities, line widths and shifts over the sample.

5.1 Structure determination: the chemical industry

X-ray diffraction is the standard tool of crystallography. Neutron powder diffraction takes over in the many cases where single crystals cannot be made, and in cases where hydrogen atom positions need to be known. The technique of powder profile refinement is now well developed for both reactor and pulsed
FIG. 6. The cage structure of sodium zeolite A and its diffraction pattern. It undergoes distortion when the cations are replaced by lithium [8].

neutron sources. Figure 6 shows such a "routine" structural problem in zeolite chemistry solved using both X-ray and neutron refinements at the Argonne IPNS source [8].

5.2 Micro-crystallinity of catalysts: the gas industry

Diffraction line positions and intensities give structural information. Line widths give information on the size, strain
and defect nature of the crystallites responsible for the diffraction. In particular, the widths of lines in $Q$ can have the contributions:

i) Crystallite mean size ($L$)  $\Delta Q = \frac{2\pi}{L}$

ii) Strain ($S$)  $\Delta Q = QS$

iii) Paracrystallinity ($\sigma$)  $\Delta Q = Q^2\sigma$

Thus a plot of linewidth $\Delta Q$ against $Q$ reveals an intercept giving the crystallite size and a flat, linear or quadratic variation with $Q$. Pulsed neutron powder diffractometers are particularly valuable for this work since they give good resolution over a very wide range of $Q$. Figure 7 shows measurements on co-precipitated nickel/alumina catalysts made on the Back Scattering Spectrometer (BSS) on the Harwell linac [9]. The linear plots reveal a high degree of paracrystalline disorder. Further measurements revealed the changes in disorder throughout the reduction of the catalyst.

5.3 Diffuse scattering in a ceramic: the aerospace industry

A perfectly ordered crystalline material gives only Bragg scattering together with a little temperature dependent thermal
FIG. 8. Two samples of zinc sulphide prepared by different methods. The upper sample gives diffuse scattering between the Bragg peaks.

FIG. 9. Experimental and calculated line intensities for diffraction peaks observed in stainless steel welds containing 16% ferrite (high).

diffuse scattering. Any disorder gives diffuse scattering between Bragg peaks. The magnitude of diffuse scattering may therefore be used as a test of the perfection of crystalline materials. Figure 8 shows two samples of zinc sulphide used for infra-red windows showing very similar properties in many respects but
differing greatly in diffuse scattering. These were measured on the Harwell linac [10]. In this case very few quality control techniques showed differences between specimens.

5.4 Minority phases in welds: the nuclear industry

Neutron diffraction can be a quantitative measure of minority phases at less than 1% volume fraction. Electron diffraction readily detects such phases but quantitative analysis is hard. X-ray diffraction suffers from an intrinsic background from Compton scattering and fluorescence limiting detection sensitivity. Figure 9 shows studies of stainless steel welds [11] showing both austenite (γ) and ferrite (α) peaks as welded and after a simulated in-service life of 1000 hours at 625°C. The deleterious sigma phase growth is clearly seen. Its growth has been followed as a function of time "in situ" in a furnace.

5.5 Texture: the steel industry

A polycrystalline sample that has been rolled, forged or drawn in general develops a tendency for its crystallites to be orientated along some defined direction with respect to the sample axes. This texture can be desirable as in the manufacture of transformer steels where the preferred orientation approaches that of a deformed single crystal. More often it is undesirable particularly in the prevention of cracking in components formed by hot pressing. X-ray diffraction remains the standard technique but neutron diffraction has the vital advantage of performing the measurement over a cc or so of material. The sample generally contains sufficient grains and the pole figure to be obtained is a true statistical average. With X-rays especially with large grained samples, the scattering volume may cover only a few grains, or even only one grain. The statistical fluctuations due to grains are large and the final pole figure is irregular and noisy. Figure 10 shows a 110 pole figure measured by Szpuner and Ojanen [12] on the FIRII reactor Otaniemi on Fe3%Si steel for the electrical industry.

5.6 Internal stress: the welding industry

The importance of residual stress in components such as welds is hard to over-emphasise. After welding the stresses will typically be a large fraction of the yield stress. Most design codes force the user to assume internal stresses equal to the yield stress thus leading to overspecification of the weld thickness. This in turn leads to reduced heat transfer and an aggravation of the problem. In this important field the information possible with neutrons is unique. Given a piece of unstrained material they can give the full stress tensor nondestructively at a point within a bulk sample. Even with no unstressed sample the principal unstressed directions can
FIG. 10. The measured torque curve for rolled and annealed Fe3%Si alloy compared to calculation (points) from measured pole figures (right hand side) [12].

FIG. 11. The variation of residual stress with position through a double-V weld sample as shown inset. The shaded area represents the results of strain gauge measurements by Leggatt.
always be obtained. Figure 11 shows the stresses measured in a typical weld provided by the Welding Institute, Abington, UK compared with the results from conventional destructive techniques.

6. QUASI-ELASTIC AND VIBRATIONAL SCATTERING

Moving nuclei give rise to Doppler shifts in neutron energy giving rise to inelastic scattering. Perhaps because of their generally longer counting times inelastic instruments have not yet had the impact on applied materials science of diffractometers. Diffusional motion gives rise to a broad inelastic peak centred about the elastic energy. Sometimes this quasi-elastic scattering can be recognised by the fact that, like diffuse scattering, it occurs between Bragg peaks. It can then be measured by a simple diffractometer as in the first example below. More usually it requires an inelastic spectrometer to separate the quasi-elastic from the elastic scattering.

6.1 Structural disorder: reactor safety

An understanding of the properties of UO₂ at temperatures right up to its melting point is vital to the safety case of nuclear reactors in possible fault conditions. In fact UO₂ has quite anomalous thermodynamic properties in the temperature range from 2000 to 3000°K. Neutron quasi-elastic measurements made in a high temperature furnace up to 2930°K show the anomalies to be caused by oxygen disorder. Figure 12 shows the temperature dependence of the quasi-elastic scattering [13]. Its Q dependence identifies the form of the oxygen defect motions.
6.2 Diffusion in aqueous solutions: the food industry

The rate of diffusion of water molecules is sensitive to only a few percent molar concentrations of many solvents. We believe the earliest experiment performed for repayment by industry was of this form. It was a study of the motions of water molecules in dilute solutions of tertiary butyl alcohol - a hydrophobic molecule. Figure 13 shows the quasi-elastic width as a function of Q value by Franks et al [14]. A 10% molar solution slows the rate of diffusion by a factor of two. This work suggested that the t-Bu-OH molecules may affect the neighbouring water molecules by a co-operative mechanism.

6.3 Diffusion of ethylene on a zeolite catalyst: the chemical industry

The nature of the rotational jump diffusion and translational diffusion of an ethylene molecule adsorbed on a catalyst can be tracked down by quasi-elastic neutron scattering. The high contrast of hydrogen and the open nature of the zeolite cages structure make it relatively easy to pick out the scattering from the ethylene. Figure 14 shows the quasi-elastic
FIG. 14. Quasi-elastic scattering from ethylene on a zeolite catalyst. The dashed line denotes the rotational scattering involving 180° jumps.

scattering measured at medium (0.1 meV) resolution. The broad rotational diffusion is well separated from the translational diffusion which can be measured at higher resolution [15], [16].

7. CONCLUSIONS

The growth in industrial use of neutron beams suggested 15 years ago has occurred, if not at the predicted rate. The applications are now demonstrated, and the task is to train industrial scientists and their colleagues at nuclear centres to use the new tools available to them.

ACKNOWLEDGEMENT

We are grateful to those customers who have allowed us to quote their results.

REFERENCES

CONCLUDING REMARKS
CONCLUDING REMARKS

M. LAMBERT
France

H. STILLER
Federal Republic of Germany

This conference has been devoted to experimental techniques in neutron scattering. Nevertheless, seven papers on scientific topics were also included because one naively feels that effort on advanced instrumentation does not make much sense if one does not know what it is good for. We wish to say that the seven scientific reviews and prognoses certainly ought to convince everyone that neutrons are and will remain the most suitable tools for studies on a very wide range of many-particle problems, with atoms and molecules and unpaired electrons as particles. The problems range from applied materials research as illustrated from so many new aspects by Windsor, (IAEA-CN-46/45), to phenomena of a more and more complex character, such as the phasons in incommensurate systems, tunnelling modes, fluctuations at phase transitions with still some unexpected features, as we have heard, to the structure and morphology as well as the molecular kinetics in enormously complex systems, systems of polymers and of biological macromolecules: for instance of polymer blends, mixtures of synthetic and natural rubbers, cellular organels, viruses, micellar solutions and micro-emulsions and many others. For studies on such complex systems, the idea presented by Stuhrmann (IAEA-CN-46/48) - to replace isotope substitution by a variation of the coherent proton scattering length via polarization of the neutrons and the nuclei in the sample - seemed to us especially interesting and promising. Great effort will doubtless be spent on developing this technique.

We have seen how, on the one hand, at present one approaches increasingly complex phenomena in comparatively simple systems, and, on the other hand, comparatively simple phenomena in increasingly complex systems. Soon will arise the urgent need to understand complex phenomena in complex systems. Thus, we now know that future studies will require enhanced intensity, improved resolution and the most sophisticated instrumental techniques.

For the experimentalists that most of us are, it is most comforting to know this, even though we all are aware that in real life things do not develop in this naive logical way.
One usually does not know at first what it is one wishes to know. Rather the tools are developed independently of their precise purpose. In practice, science has often been advanced primarily by methodical progress preceding the definition of problems. This holds, we think, for theoretical methods as it does for experiments.

Reports have been presented about existing neutron sources, sources being upgraded, novel sources in statu nascendi, and about novel sources being conceived, steady-state and pulsed sources; furthermore, about entirely novel instruments, adapted to the respective sources, as well as about new or improved components for such novel instruments and also for existing ones. The general impression is certainly that a very large amount of development is in progress.

As to the sources, a steady-state reactor is, of course, something straightforward. If the beam ports are well designed everything is up to the experimentalists. If anything is not optimal, they can only blame themselves. The situation is very different with pulsed sources. The pulse repetition rate, for instance, is a source parameter, and different people may want quite different values for it. Messrs Leadbetter (IAEA-CN-46/41) and Alefeld (IAEA-CN-46/80) both illustrated this with examples. For instance, a repetition rate of, say, 50 Hz may be too low for someone who wishes to measure quasi-elastic scattering only or inelastic scattering in a very limited energy range. At a reactor he would perhaps choose 250 Hz. However, the same 50 Hz may be too high, for instance, for a small-angle scatterer, because of frame overlap. At a reactor everyone can choose his own chopper frequency. At a pulsed source a compromise is required, such that everyone gains as much as possible and nobody loses too much.

At most pulsed sources, the pulse duration is also a source characteristic. Again, different experimentalists may want different values. It is only at first sight that one wishes the source pulses in every case to be as short as possible. For measurements which use the time structure of the source not for resolution but for other purposes, quite often longer pulses are better. Alefeld (IAEA-CN-46/80) has shown this for a multiplexing backscattering spectrometer and for small-angle scattering. Long source pulses (around 100 µs) provide a large time-average flux and have the advantage, of course, that everyone can choose his own pulse length for his beam provided that one can place choppers close enough to the moderators.
We have learned that, for all these reasons, it does not make sense to compare two sources by just one number, by saying source A is 50% better than source B, as desirable as such a statement would be when one faces committees and applies for money. It cannot be made. Even a comparison of fluxes as a function of energy is inadequate. The quantity that currently at least needs to be considered as a function of energy is

\[
\frac{\phi_A \cdot T_A}{\phi_B \cdot T_B}
\]

where the first factor is the ratio of fluxes, best taken at sample positions of course, and the \( T \) are what may be called time utilization factors, that is the fraction of the total time actually available for collecting interesting and relevant data. For a classical time-of-flight measurement, for instance, this would be for a comparison of a pulsed source (PS) with a steady-state source (SS)

\[
\frac{\phi_{PS} \cdot v_{PS}}{\phi_{SS} \cdot v_{ideal}}
\]

if \( v_{ideal} \) is the repetition rate you would choose at a steady-state source with a chopper. If, for instance, \( v_{PS} = 50 \text{ Hz} \), while at a steady-state source we would take \( v_{ideal} = 250 \text{ Hz} \), then this total ratio is 1/5, so that the flux ratio must be larger than 5 if this measurement is to be better than at a reactor. But of course this quantity is different for different experiments. We shall return to this subsequently.

The optimization of instruments is just as important as the optimization of sources. The expense for new sources would be partly wasted if the instruments were not best adapted to them.

General improvements of scattering instruments, independent of the sources, naturally primarily concern instrumental components. We were most impressed by the devices for handling neutron spins: various new polarizers, the Drabkin wiggler as a monochromator or analyser, in particular if it is used together with inverted time-of-flight such that, by decreasing the perpendicular field linearly with
time, one can accept a very wide frequency band without worsening resolution. At least intellectually, we found most fascinating the devices proposed by Rauch and Summhammer (IAEA-CN-46/69) for focusing and bunching neutron pulses with the help of either a series of magnetic force fields or one force field travelling with the neutron. We cannot judge how easily this method can be accomplished in practice; it certainly would be of great importance for improving resolution without loss of intensity.

Then there were the different librating, rotating, swinging or otherwise dancing instrumental parts of which the Jülich people are so particularly fond; for instance, the concept for constant Q scans by inverted time-of-flight or the diffuse scattering with six concentric cylinders rotating around the sample to eliminate inelastic scattering. I shall not examine the many ideas here because of our limited time. Last but not least, the great progress achieved with detectors, in particular area detectors, must be mentioned.

With respect to the congruence of instruments and sources we think that the most essential perception presented at this conference is the realization that the time structure of the source can be exploited not only with time-of-flight measurements of the kind you do at a reactor by chopping the steady-state beam but also by multiplexing. One performs a steady-state measurement, as at a reactor, and uses the time structure of the source to do a number of measurements simultaneously. The gain, in comparison to a steady-state source of equal time-averaged flux, is then the number of such simultaneous measurements one can perform in the intervals between pulses. For such measurements one needs a large time-averaged flux and hence a long pulse. The pulse length does not determine the resolution. As examples, small-angle scattering, diffuse scattering, backscattering and the spin-echo instrument have been discussed.

CONCLUSIONS

Higher neutron fluxes are needed for

- more extensive use of polarization (not only for magnetic studies but also, for instance, to separate coherent and incoherent scattering)
- higher resolution
- studies with more and larger ranges of parameters
- smaller and/or more dilute samples
- the study of events of smaller probability (small cross-sections, difference measurements).

In general: more complex phenomena in more complex systems, i.e. quantitative as well as qualitative progress is necessary.

Moon (IAEA-CN-46/28) estimates that in the 1990s the number of neutrons hitting samples per unit time will perhaps be doubled. Actually, that is not a tremendous enhancement. However, not only more neutrons are desirable as such, but more effective sources are needed to make possible studies on questions which have arisen from the answers found with the sources and the instruments at present available.

As of today, we have in Europe two advanced steady-state sources, the high-flux reactor at the ILL in Grenoble and the ORPHEE reactor in Saclay. This is quantitatively not enough. The demand for beam time at the ILL is more than double what is available, even under normal reactor conditions. Many proposals are rejected, not always because they are below a certain standard of quality but, one must admit, almost arbitrarily. The arbitrariness is unavoidable. And the demand is still increasing.

A third advanced neutron source, the spallation source SNS in the United Kingdom is about to go into operation. The construction of a fourth advanced source, a further spallation source, the SNQ, is now proposed in the Federal Republic of Germany. One may ask if this is not too much? From the point of view of condensed matter research, including biology, the answer must be no! As to the quantity of problems awaiting investigation, we have learned here, from the many different fields of research, that this statement is obviously true. As to progress in the quality of sources, we already mentioned at the beginning that to assess this requires detailed discussions and calculations. Such discussions and calculations have been carried out in and between some of the workshops which took place in 1984 and have been the central point of many discussions at this conference, as well as last October at the Shelter Island conference in the United States of America. An important elaboration of this kind is given in the paper by Jauch and Dachs (IAEA-CN-46/71).

Figure 1 shows the peak and the time-averaged fluxes expected for the SNS and the SNQ. One sees how well the SNS and the SNQ will complement each other as long as the SNQ is not equipped with a pulse compressor, and this will
probably be the case for quite a while. The SNQ surpasses the SNS in peak flux for neutron energies below 200 meV. For higher energies, 200 meV and above, the SNS becomes superior. And this is very consistent with the pulse length requirements at the experiments. At high energies one needs short pulses.
For the SNQ, the figure also indicates what the effective fluxes - the peak fluxes times the time utilization factors mentioned above - will be for various types of measurement. The indicated areas show the ranges of effective fluxes one may obtain, depending on the special experimental requirements. The best effective flux one can obtain is, of course, the nominal peak flux of the source, as obtained for instance for a classical time-of-flight measurement, if the source repetition rate happens to be the optimal frequency also for one's measurement. That is, the time utilization factor is 1. If it is not so, then the effective flux is lower. The figure reflects the analyses and numerical work done last year.

We conclude from all this that new sources, combined with new instruments, will lead to true and substantial progress in the physics and chemistry of condensed matter, in materials science and, in particular, in molecular biology. We should not hesitate to state this in all our countries.
CHAIRMEN OF SESSIONS

Session I
M. BLUME
United States of America

Session II
M. LAMBERT
France

Session III
J. JANIK
Poland

Session IV
Y. ISHIKAWA
Japan

Session V
J. BROWN
Institut Max von Laue — Paul Langevin

Session VI
H.F. BERTAUD
France

Session VII
C. WINDSOR
United Kingdom

Session VIII
B. FENDER
Institut Max von Laue — Paul Langevin

K. NIEHAUS
Federal Republic of Germany

Session IX
J. KJEMS
Denmark

Session X
T. SPRINGER
Federal Republic of Germany

W. SCHMATZ
Federal Republic of Germany

Session XI
G.H. LANDER
United States of America

SECRETARIAT

Scientific Secretaries:
M. LEISER
M. MEHTA
Division of Research and Laboratories, IAEA

Administrative Secretary:
Gertrude SEILER
Division of External Relations, IAEA

Editors:
Monica KRIPPNER
R. PENISTON-BIRD
Division of Publications, IAEA

603
LIST OF PARTICIPANTS

Adib, M.
Reactor and Neutron Physics Department,
Nuclear Research Centre, Atomic Energy Authority,
Cairo, Egypt

Alefeld, B.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Allain, Y.
Laboratoire Léon Brillouin,
CEA, Centre d’études nucléaires (CEN) de Saclay,
P-91191 Gif-sur-Yvette Cedex, France

Andreeff, A.
Technische Universität Dresden,
Mommsenstrasse 13,
DDR-8027 Dresden, German Democratic Republic

Arnold, H.
Institut für Kristallographie,
Technische Hochschule Aachen,
Templergraben 55,
D-5100 Aachen, Federal Republic of Germany

Badurek, G.
Atom Institut der Oesterreichischen Universitäten,
Schüttelstrasse 115, A-1020 Vienna, Austria

Bell, H.G.
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Bergsma, J.
Netherlands Energy Research Foundation (ECN),
P.O. Box 1, NL-1755 ZG Petten, Netherlands

Bertaut, E.F.
Laboratoire de cristallographie,
CEA, Centre d’études nucléaires (CEN) de Grenoble,
166 X, F-38042 Grenoble, France

Blume, M.
Brookhaven National Laboratory,
Upton, NY 11973, United States of America

Böning, K.
Fakultät für Physik,
Technische Universität München,
D-8046 Garching, Federal Republic of Germany

Born, R.
Institut für Kristallographie,
Universität Tübingen,
Charlottenstrasse 33,
D-7400 Tübingen, Federal Republic of Germany

Bowman, C.
Los Alamos National Laboratory,
Los Alamos, NM 87545, United States of America

Buchenau, U.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

605
LIST OF PARTICIPANTS

Bührer, R.
Labor für Neutronenstreuung,
Eidgenössische Technische Hochschule Zürich,
c/o Eidgenössisches Institut für Reaktorforschung,
CH-5303 Würenlingen, Switzerland

Burke, R.
Westinghouse Hanford Co.,
P.O. Box 1970,
Richland, WA 99352, United States of America

Burkel, E.
Sektion Physik, Universität München,
Geschwister Scholl Platz 1,
D-8000 Munich 22, Federal Republic of Germany

Butt, N.M.
Pakistan Institute of Nuclear Science and Technology (PINSTECH),
Nilore, Rawalpindi, Pakistan

Carlile, C.J.
Neutron Division, Rutherford Appleton Laboratory,
Chilton, Didcot, Oxon OX11 OQX, United Kingdom

Carne, A.
Rutherford Appleton Laboratory,
Chilton, Didcot, Oxon OX11 OQX, United Kingdom

Chalupa, B.
Nuclear Physics Institute,
250 68 Rež, Czechoslovakia

Cloth, P.
Institut für Reaktorentwicklung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Cotton, J.-P.
Laboratoire Léon Brillouin, CEA, CEN de Saclay,
F-91191 Gif-sur-Yvette Cedex, France

Coulter, C.L.
Biotechnology Resources Program,
Division of Research Resources,
National Institute of Health,
Building 31, Room 5B 41, 9000 Rockville Pike,
Bethesda, MD 20205, United States of America

Cowley, R.A.
Department of Physics, Edinburgh University,
Mayfield Road, Edinburgh EH9 2LX, Scotland,
United Kingdom

Cox, D.E.
Physics Department, Brookhaven National Laboratory,
Building 510 B,
Upton, NY 11973, United States of America

Dachs, H.
Hahn-Meitner-Institut für Kernforschung Berlin GmbH,
Glienicker Strasse 100, D-1000 Berlin 39

Dahlbör, U.
Department of Reactor Physics,
Royal Institute of Technology,
S-100 44 Stockholm, Sweden

Dasannacharya, B.A.
Nuclear Physics Division,
Bhabha Atomic Research Centre,
Trombay, Bombay 400 085, India

Davidovic, M.
Boris Kidrić Institute of Nuclear Sciences,
Vinča, P.O. Box 522, Belgrade, Yugoslavia
Delapalme, A. Laboratoire Léon Brillouin, CEA, CEN de Saclay, P-91191 Gif-sur-Yvette Cedex, France

Delaplane, R. Studsvik Research Laboratory, S-611 82 Nyköping, Sweden

Deraman, Mohamad Bin Nuclear Energy Unit, Prime Minister Department, Komplek Puspati, Bandar, Selangor, Malaysia

Dolling, G. Atomic Energy of Canada Ltd, Chalk River Nuclear Laboratories, Chalk River, Ontario K0J 1J0, Canada

Driesen, G.M. Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Eckold, G. Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Faraqo, B. (see also under Farnoux, B.) Institut Max von Laue – Paul Langevin, 156X, P-38042 Grenoble, France

Farnoux, B. Laboratoire Léon Brillouin, CEA, CEN de Saclay, P-91191 Gif-sur-Yvette Cedex, France

Felcher, G.P. Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439, United States of America

Filges, D. Institut für Reaktorentwicklung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Fischer, W.E. Swiss Institute for Nuclear Research, CH-5234 Villigen, Switzerland

Frick, B. Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Fuess, H. Institut für Kristallographie und Mineralogie, Universität Frankfurt, Senckenberanlage 30, D-6000 Frankfurt/Main 1, Federal Republic of Germany

Furrer, A. Labor für Neutronenstreuung, Eidgenössische Technische Hochschule Zürich, c/o Eidgenössisches Institut für Reaktorforschung, CH-5303 Würenlingen, Switzerland

Geick, R. Physikalisches Institut, Universität Würzburg, Röntgenring 8, D-8700 Würzburg, Federal Republic of Germany
LIST OF PARTICIPANTS

Glättli, H.
Service de physique du solide et de résonance magnétique,
CEA, CEN de Saclay,
P-91191 Gif-sur-Yvette Cedex, France

Grapengeter, H.H.
Institut für Physikalische Chemie,
Universität Hamburg,
Laufgraben 24,
D-2000 Hamburg 13, Federal Republic of Germany

Gray, D.A.
Rutherford Appleton Laboratory,
Chilton, Didcot, Oxon OX11 OQX, United Kingdom

Grimm, H.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Grondey, S.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Gudowski, W.
Institute of Reactor Physics,
Royal Institute of Technology,
Drottning Kristinas Väg 47,
S-114 28 Stockholm, Sweden

Haensel, R.
Institut für Experimentalphysik, Universität Kiel,
Olshausenstrasse 40-60,
D-2300 Kiel 1, Federal Republic of Germany

Häfele, W.
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Hahn, T.
Rheinisch-Westfälische Technische Hochschule (RWTH),
Templergraben 59,
D-5100 Aachen, Federal Republic of Germany

Haji-Saeid, S.M.
Atomic Energy Organization of Iran,
P.O. Box 12-1198, Teheran, Iran

Hauck, J.
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Hautecler, S.
Centre d'étude de l'énergie nucléaire (CEN/SCK),
Boeretang 200, B-2400 Mol, Belgium

Hecker, R.
Institut für Reaktorentwicklung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Heiroth, M.K.
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany
LIST OF PARTICIPANTS

Hempelmann, R. Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Hiismäki, P. Reactor Laboratory, Technical Research Centre of Finland, Otakaari 3 A, SF-02150 Espoo, Finland

Hildebrand, E. Institut für Neurobiologie, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany


Hohlwein, D. Institut für Kristallographie, Universität Tübingen, Charlottenstrasse 33, D-7400 Tübingen, Federal Republic of Germany

Inoue, H. National Institute of Agrobiological Resources, Tsukuba Science City, Yatabe, Ibaraki-ken 305, Japan

Ishikawa, Y. Physics Department, Tohoku University, Aramaki Sendai 980, Japan

Iyengar, P.K. Bhabha Atomic Research Centre, Trombay, Bombay 400 085, India

Janik, J. Institute of Nuclear Physics, PL-30-059 Cracow, Poland

Jansen, E. Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Jauch, W. Hahn-Meitner-Institut für Kernforschung Berlin GmbH, Glienicker Strasse 100, D-1000 Berlin 39

Jung, W. Institut für Physikalische Chemie, Rheinisch-Westfälische Technische Hochschule (RWTH), Templergraben 59, D-5100 Aachen, Federal Republic of Germany

Juul Jensen, D. Risø National Laboratory, Postbox 49, DK-4000 Roskilde, Denmark

Kalus, J. Physikalisches Institut und Institut für physikalische Chemie, Universität Bayreuth, Postfach 3008, D-8580 Bayreuth, Federal Republic of Germany

Kalvius, G.M. Department für Physik des, Technische Universität München, D-8046 Garching, Federal Republic of Germany
<table>
<thead>
<tr>
<th>Name</th>
<th>Institution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kammermeier, H.</td>
<td>Institut für anorganische Chemie, Universität München, Meiserstrasse 1, D-8000 Munich 2, Federal Republic of Germany</td>
</tr>
<tr>
<td>Kampmann, R.</td>
<td>GKSS-Forschungszentrum GmbH, Max-Planck-Strasse 1, D-2054 Geesthacht, Federal Republic of Germany</td>
</tr>
<tr>
<td>Kjems, J.K.</td>
<td>Risø National Laboratory, Postbox 49, DK-4000 Roskilde, Denmark</td>
</tr>
<tr>
<td>Lambert, M.</td>
<td>Laboratoire Léon Brillouin, CEA, CEN de Saclay, F-91191 Gif-sur-Yvette Cedex, France</td>
</tr>
<tr>
<td>Lander, G.H.</td>
<td>Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439, United States of America</td>
</tr>
<tr>
<td>Larsson, K.E.</td>
<td>Institute of Reactor Physics, Royal Institute of Technology, Drottning Kristinas Väg 47, S-114 28 Stockholm, Sweden</td>
</tr>
<tr>
<td>Leadbetter, A.J.</td>
<td>Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, United Kingdom</td>
</tr>
<tr>
<td>Lechner, R.E.</td>
<td>Hahn-Weitner-Institut für Kernforschung Berlin GmbH, Glienicker Strasse 100, D-1000 Berlin 39</td>
</tr>
<tr>
<td>Leciejewicz, J.</td>
<td>Institute of Nuclear Chemistry and Technology, Dorodna 16, PL-03-195 Warsaw, Poland</td>
</tr>
<tr>
<td>Leqrand, E.</td>
<td>CEN/SCK, Boeretand 200, B-2400 Mol, Belgium</td>
</tr>
<tr>
<td>Loewenhaupt, M.</td>
<td>Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany</td>
</tr>
<tr>
<td>Marsongkohadi</td>
<td>Research Centre for Nuclear Techniques, Jalan Tamansari 71, Bandung, Indonesia</td>
</tr>
<tr>
<td>Mezei, F.</td>
<td>Hahn-Weitner-Institut für Kernforschung Berlin GmbH, Postfach 39 01 28, D-1000 Berlin 39</td>
</tr>
<tr>
<td>Middendorf, H.D.</td>
<td>Department of Biophysics, King's College, London WC2B 5RL, United Kingdom</td>
</tr>
<tr>
<td>Mikula, P.</td>
<td>Nuclear Physics Institute, 250 68 Ústí, Czechoslovakia</td>
</tr>
<tr>
<td>Möller, H. B.</td>
<td>Risø National Laboratory, Postbox 49, DK-4000 Roskilde, Denmark</td>
</tr>
<tr>
<td>Monkenbusch, M.</td>
<td>Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany</td>
</tr>
</tbody>
</table>
LIST OF PARTICIPANTS

Montoya Zavaleta, M. Universidad Nacional de Ingeniería, Facultad Ciencias, Apt. 1301, Lima, Peru

Moon, R.M. Solid State Division, Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, TN 37830, United States of America

Müller, K.D. Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Neef, R.D. Institut für Reaktorentwicklung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Neumann, J.-M. Département de biologie, CEA, CEN de Saclay, F-91191 Gif-sur-Yvette Cedex, France

Nève de Mévergnies, M. CEN/SCK, Boeretang 200, B-2400 Mol, Belgium

Nicklow, R.M. Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, TN 37830, United States of America

Niel, L. Atom Institut der Oesterreichischen Universitäten, Schüttelstrasse 115, A-1020 Vienna, Austria

Nierhaus, K. Max-Planck-Institut für Molekulare Genetik, Ihnestrasse 63-73, D-1000 Berlin 33

Oleś, A. Institute of Physics and Nuclear Techniques, University of Mining and Metallurgy, Ul. Mickiewicza 30, PL-30-059 Cracow, Poland

Olovsson, I. Institute of Chemistry, University of Uppsala, Box 531, S-751 21 Uppsala, Sweden

Otnes, K. Institute for Energy Technology, P.O. Box 40, N-2007 Kjeller, Norway

Pépy, G. Laboratoire Léon Brillouin, CEA, CEN de Saclay, F-91191 Gif-sur-Yvette Cedex, France

Pfeiffer, F.W. Aussenstelle D, Technische Hochschule Darmstadt, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Prager, M. Institut für Festkörperforschung, Kernforschungsanlage Jülich GmbH, Postfach 1913, D-5170 Jülich, Federal Republic of Germany

Priesmeyer, H.G. Institut für Kernphysik, Universität Kiel, c/o GKSS-Forschungszentrum Geesthacht GmbH, Max-Planck-Strasse 1, D-2054 Geesthacht, Federal Republic of Germany
LIST OF PARTICIPANTS

Rauch, H. 
Atominstitut der Oesterreichischen Universitäten,  
Schüttestrasse 115, A-1020 Vienna, Austria

Richter, D. 
Institut für Festkörperforschung,  
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany

Rittinghaus, K.F. 
Zentraallabor für Elektronik,  
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany

Robinson, A.L. 
American Association for the  
Advancement of Science,  
1515 Massachusetts Avenue, NW,  
Washington, DC 20005, United States of America

Robl, C.R. 
Institut für Anorganische Chemie,  
Universität München, Meiserstrasse 1,  
D-8000 Munich 2, Federal Republic of Germany

Rosta, L. 
Central Research Institute for Physics,  
P.O. Box 49, H-1525 Budapest, Hungary

Roult, G. 
Laboratoire de diffraction neutronique,  
CEA, CNRS de Grenoble,  
85X, F-38041 Grenoble Cedex, France

Sadikov, I. 
Kurchatov Atomic Energy Institute,  
Kurchatov Place, Moscow,  
Union of Soviet Socialist Republics

Sangarivavanich, A. 
Office of Atomic Energy for Peace,  
Vibhavadi Rangsit Road,  
Bangkhen, Bangkok 10900, Thailand

Schaal, H. 
Institut für Reaktorentwicklung,  
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany

Schätzler, R. 
Institut für Festkörperforschung,  
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany

Schelten, J. 
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany

Scherm, R. 
Physikalisch-Technische Bundesanstalt,  
Postfach 3345,  
D-3300 Braunschweig, Federal Republic of Germany

Schilling, W. 
Institut für Festkörperforschung,  
Kernforschungsanlage Jülich GmbH,  
Postfach 1913,  
D-5170 Jülich, Federal Republic of Germany
LIST OF PARTICIPANTS

Schmatz, W.
Institut für Nukleare Festkörperphysik,
Kernforschungszentrum Karlsruhe GmbH,
Postfach 3640,
D-7500 Karlsruhe, Federal Republic of Germany

Schofield, P.
Atomic Energy Research Establishment,
Harwell, Didcot, Oxon OX11 ORA, United Kingdom

Schwahn, D.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Schweizer, J.
Laboratoire de diffraction neutronique,
CEA, CEN de Grenoble,
85X, F-38041 Grenoble Cedex, France

Seeqer, A.
Central Laboratory for Electronics,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Seeliger, D.
Technische Universität Dresden,
Mommsenstrasse 13,
DDR-8027 Dresden, German Democratic Republic

Shirane, G.
Physics Department, Brookhaven National Laboratory,
Building 510 B,
Upton, NY 11973, United States of America

Smith, H.J.
Atomic Energy of Canada Ltd,
Whiteshell Nuclear Research Establishment,
Pinawa, Manitoba ROE 1L0, Canada

Somenkov, V.
Kurchatov Atomic Energy Institute,
Kurchatov Place, Moscow,
Union of Soviet Socialist Republics

Sosnowska, I.
Institute of Experimental Physics,
Warsaw University,
Hoza 69, PL-00-681 Warsaw, Poland

Springer, T.
Institut für Festkörperforschung,
Kernforschungsanlage Jülich GmbH,
Postfach 1913,
D-5170 Jülich, Federal Republic of Germany

Stein, R.S.
Polymer Research Institute,
University of Massachusetts,
701 Graduate Research Center,
Amherst, MA 01003, United States of America

Steiner, M.
Hahn-Meitner-Institut für
Kernforschung Berlin GmbH,
Glienicker Strasse 100, D-1000 Berlin 39

Steinhauser, K.-A.
Fakultät für Physik,
Technische Universität München,
James-Franck-Strasse,
D-8046 Garching, Federal Republic of Germany
LIST OF PARTICIPANTS

Stiller, H.  Projekt Spallationsneutronenquelle,  
            Kernforschungsanlage Jülich GmbH,  
            Postfach 1913,  
            D-5170 Jülich, Federal Republic of Germany

Stockmeyer, R.  Institut für Festkörperforschung,  
            Kernforschungsanlage Jülich GmbH,  
            Postfach 1913,  
            D-5170 Jülich, Federal Republic of Germany

Stuhrmann, H.B.  Deutsches Elektronen-Synchrotron (DESY),  
            D-2000 Hamburg 2, Federal Republic of Germany

Suck, J.-B.  Institut für Nukleare Festkörperphysik,  
            Kernforschungszentrum Karlsruhe GmbH,  
            Postfach 3640,  
            D-7500 Karlsruhe, Federal Republic of Germany

Sumhammer, J.  Atom-Institut der Österreichischen Universitäten,  
            Schütztenstrasse 115, A-1020 Vienna, Austria

Taylor, A.D.  Neutron Division, Rutherford Appleton Laboratory,  
            Chilton, Didcot, Oxon OX11 OQX, United Kingdom

Tellgren, R.  Institute of Chemistry, University of Uppsala,  
            Box 531, S-751 21 Uppsala, Sweden

Tourand, G.J.  Laboratoire Léon Brillouin, CEA, CEN de Saclay,  
            F-91191 Gif-sur-Yvette Cedex, France

Truong, Quang-Vinh  Hahn-Meitner-Institut für  
            Kernforschung Berlin GmbH,  
            Glienickerstrasse 100, D-1000 Berlin 39

Uhlhorn, C.D.  Bundesministerium für Forschung und Technologie,  
            Postfach 20 07 06,  
            D-5300 Bonn 2, Federal Republic of Germany

Van Loef, J.J.  Interuniversitair Reactor Instituut (IRI),  
            Mekelweg 15, NL-2629 JB Delft, Netherlands

Vorderwisch, P.  Hahn-Meitner-Institut für  
            Kernforschung Berlin GmbH,  
            Glienickerstrasse 100, D-1000 Berlin 39

Vratislav, S.  Technical University of Prague,  
            Nuclear Faculty,  
            Prague, Czechoslovakia

Watanabe, N.  National Laboratory for High Energy Physics,  
            Oho-machi, Tsukuba-gun, Ibaraki-ken 305, Japan

Wegener, W.  CEN/SCK,  
            Boeretang 200, B-2400 Mol, Belgium

Weinfurter, H.  Atom-Institut der Österreichischen Universitäten,  
            Schütztenstrasse 115, A-1020 Vienna, Austria

Welter, J.-M.  Institut für Festkörperforschung,  
            Kernforschungsanlage Jülich GmbH,  
            Postfach 1913,  
            D-5170 Jülich, Federal Republic of Germany
LIST OF PARTICIPANTS

Werner, K.  
Institut für Festkörperforschung, 
Kernforschungsanlage Jülich GmbH, 
Postfach 1913, 
D-5170 Jülich, Federal Republic of Germany

Wilkinson, K.  
Solid State Division, 
Oak Ridge National Laboratory, 
PO Box X, 
Oak Ridge, TN 37831, United States of America

Williams, W.G.  
Neutron Division, 
Rutherford Appleton Laboratory, 
Chilton, Didcot, Oxon OX11 OQX, United Kingdom

Windsor, C.G.  
Materials Physics and Metallurgy Division, 
Atomic Energy Research Establishment, 
Harwell, Didcot, Oxon OX11 ORA, United Kingdom

Wolf, L.  
Institut für Reaktorenentwicklung, 
Kernforschungsanlage Jülich GmbH, 
Postfach 1913, 
D-5170 Jülich, Federal Republic of Germany

Woods, R.  
Los Alamos National Laboratory, 
P.O. Box 1663, MS H805, 
Los Alamos, NM 87544, United States of America

Yang, Zhen  
Institute of Atomic Energy, 
P.O. Box 275(30), Beijing, China

Zabel, H.  
Department of Physics, University of Illinois, 
1110 W. Green, 
Urbana, IL 61801, United States of America

Zhou, Huiming  
Institute of Atomic Energy, 
P.O. Box 275(30), Beijing, China

Zinn, W.  
Institut für Festkörperforschung, 
Kernforschungsanlage Jülich GmbH, 
Postfach 1913, 
D-5170 Jülich, Federal Republic of Germany

ORGANIZATIONS

EURATOM

Kley, W.  
Joint Research Centre, 
I-21020 Ispra (Varese), Italy

INSTITUT Max von LAUE - PAUL LANGEVIN (ILL)

Ageron, P.  
Avenue des Martyrs, 156X, 
P-38042 Grenoble Cedex, France

Brown, J.  
Avenue des Martyrs, 156X, 
P-38042 Grenoble Cedex, France

Faraqo, B.  
Avenue des Martyrs, 156X, 
P-38042 Grenoble Cedex, France 
(see also under Hungary)
<table>
<thead>
<tr>
<th>Name</th>
<th>Address</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fender, B.</td>
<td>Avenue des Martyrs, 156X, F-38042 Grenoble Cedex, France</td>
</tr>
<tr>
<td>Lehmann, M.S.</td>
<td>Avenue des Martyrs, 156X, F-38042 Grenoble Cedex, France</td>
</tr>
<tr>
<td>Michaudon, A.</td>
<td>Avenue des Martyrs, 156X, F-38042 Grenoble Cedex, France</td>
</tr>
<tr>
<td>Schärpf, O.</td>
<td>Avenue des Martyrs, 156X, F-38042 Grenoble Cedex, France</td>
</tr>
<tr>
<td>Stansfield, R.F.D.</td>
<td>Avenue des Martyrs, 156X, F-38042 Grenoble Cedex, France</td>
</tr>
<tr>
<td>Michalec, R.</td>
<td>Head Post Office, P.O. Box 79, 101000 Moscow, Union of Soviet Socialist</td>
</tr>
<tr>
<td>Natkaniec, I.</td>
<td>Laboratory of Neutron Physics, Head Post Office, P.O. Box 79, 101000</td>
</tr>
<tr>
<td></td>
<td>Moscow, Union of Soviet Socialist Republics</td>
</tr>
<tr>
<td>Pacher, P.</td>
<td>Head Post Office, P.O. Box 79, 101000 Moscow, Union of Soviet Socialist</td>
</tr>
<tr>
<td></td>
<td>Republics</td>
</tr>
</tbody>
</table>
AUTHOR INDEX

Abdel-Kawy, A.: 507
Adib, M.: 507
Ageron, P.: 135
Alefeld, B.: 247, 361, 409, 417
Allen, A.J.: 575
Ananiev, V.D.: 63
Anston, O.: 453
Atchison, F.: 171
Badurek, B.: 77
Bauer, G.: 267, 361, 413
Bauer, R.: 489, 495
Beg, M.M.: 539
Bleeker, E.J.: 377
Bönig, K.: 165
Born, R.: 461
Breiting, B.: 489
Bulkin, Yu.M.: 369
Butt, N.M.: 539
Carlile, C.J.: 421
Carne, A.: 353
Carpenter, J.M.: 17
Chalupa, B.: 515
Chazipetros, J.: 481
Cheema, S.U.: 539
Christensen, P.: 495
Conrad, H.: 267
Cotton, J.P.: 349
Cowley, R.A.: 3
Cox, D.E.: 567
Dachs, H.: 565
Dahlborg, U.: 377, 469
Dasannacharya, B.A.: 443
Delapalme, A.: 475
Delaplane, R.G.: 469
D’Hooghe, P.: 211
Dolling, G.: 385
Duppich, J.: 393
Eid, Y.: 507
Endoh, Y.: 285
Farnoux, B.: 205
Filges, D.: 267
Fischer, W.E.: 171
Frank, I.M.: 63
Freltoft, T.: 63, 495
Furrer, A.: 199
Gläser, W.: 165
Glättli, H.: 181
Gobert, G.: 211
Goyal, P.S.: 443
Grachev, M.I.: 369
Gray, D.A.: 261
Halling, H.: 361
Hauetecler, S.: 211
Hempelmann, R.: 481
Herbst, L.: 501
Hiismäki, P.: 453
Hoffmann, H.: 501
Hohlwein, D.: 461
Hutchings, M.T.: 575
Ibel, K.: 501
Inoue, K.: 285
Isacson, A.: 199
Ishikawa, Y.: 285
Iyengar, P.K.: 443
Janik, J.A.: 411
Jauch, W.: 565
Jensen, L.G.: 495
Jungerberg, G.: 527
Juul Jensen, Dorte: 293
Kalus, J.: 501
Khan, Q.H.: 539
Khmel'shchikov, V.V.: 369
Khryastov, N.A.: 369
Kirsch, O.: 481
Kjems, J.K.: 489, 495
Koch, M.: 199
Kollmar, A.: 417
Kolmychkov, N.V.: 369
Kozlov, Zh.A.: 63
Kudryashev, V.A.: 453
Kukkonen, H.: 453
Kulda, J.: 515
Kulka, J.: 533
Lander, G.H.: 17
Lang, H.: 361
Lambert, M.: 595
Leadbetter, A.J.: 219
Lechner, R.E.: 401
Legrand, E.: 211
Lehmann, M.S.: 551
Lidstone, R.F.: 385
Linderholm, J.: 495
Lobashev, V.M.: 369
Leowenhaupt, M.: 435
Luschikov, V.I.: 63
Maayouf, R.M.A.: 507
Majkrzak, C.F.: 109
Marmeggi, J.C.: 475
Mayer, J.: 411
Majkrzak, C.F.: 109
McIntyre, J.J.: 191
Meier, J.: 165
Mezei, F.: 291
Michalec, R.: 515
Middendorf, H.D.: 303
Mikula, P.: 515
Moon, R.M.: 135
Natkaniec, J.: 411
Nilsson, B.: 377
Noack, W.: 481
Olés, A.: 533
Ostanevich, Yu.M.: 63
Pawelczyk, J.: 411
Pepin, M.: 171
Perrin, M.: 475
Powell, J.R.: 157
Pöyry H.: 453
Rau, G.: 165
Rauch, H.: 35, 53
Richter, D.: 291, 409, 481
Röhrmoser, A.: 165
Roome, G.: 211
Sandström, M.: 469
Satya Murthy, N.S.: 443
Sayers, C.M.: 575
Schärpf, O.: 85
Schalt, W.: 211
Schätzler, R.: 481
Schelten, J.: 361, 413, 417
Scherm, R.: 247
Schmatz, W.: 77
Schofield, P.: 575
Sciesinski, J.: 411
Sedláková, L.: 515
Seeger, A.: 211
Shabalina, E.P.: 63
Shchebetov, A.F.: 453
Shirane, G.: 109
Sidorkin, S.F.: 369
Sinclair, R.N.: 575
Skold, J.: 377
Sköld, K.: 469
Son, J.N.: 443
Spitzer, H.: 267
Springer, T.: 393
Stansfield, R.P.D.: 191
Stavisskij, Yu.Ya.: 369
Stein, R.S.: 335
Steiner, M.: 185
Steinhauser, K.-A.: 99
Stiller, H.: 595
Stoeckli, A.: 199
Stuhrmann, H.B.: 303
Szmhammer, J.: 53
Szpunar, J.: 533
Takahashi, H.: 157
Takeda, Y.: 171
Taylor, A.D.: 421
Thamm, G.: 361
Thaper, C.L.: 443
Thuesen, A.: 489
Thurn, H.: 501
Tiitta, A.: 453
Trunov, V.A.: 453
Tschaaler, Ch.: 171
Ulyanov, V.A.: 453
Urban, S.: 411
Vansteelandt, L.: 211
Vrána, M.: 515
Watanabe, N.: 279
Wawszczak, R.: 533
Weinfurter, H.: 53
Welter, J.-M.: 527
Werner, K.: 393
Williams, W.G.: 421
Windsor, C.G.: 575
Wright, C.J.: 575
Wyrwich, H.: 481
Yelon, W.B.: 377
Zettler, Z.: 361
Zhang, L.: 167
TRANSLITERATION INDEX

Булкин, Ю. М.  Bulkin, Yu.M.
Грачев, М. И.  Grachev, M.I.
Колмычков, Н. В.  Kolmychkov, N.V.
Лобашев, В. М.  Lobashev, V.M.
Сидоркин, С. Ф.  Sidorkin, S.F.
Хмельщиков, В. В.  Khmel'shchikov, V.V.
Хрястов, Н. А.  Khryastov, N.A.
INDEX OF PAPERS AND POSTERS BY NUMBER

(The letter P indicates a poster presentation)

<table>
<thead>
<tr>
<th>IAEA-CN-46/-</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1P</td>
<td>501</td>
</tr>
<tr>
<td>2P</td>
<td>469</td>
</tr>
<tr>
<td>3</td>
<td>551</td>
</tr>
<tr>
<td>4P</td>
<td>377</td>
</tr>
<tr>
<td>5P</td>
<td>495</td>
</tr>
<tr>
<td>6P</td>
<td>489</td>
</tr>
<tr>
<td>7P</td>
<td>481</td>
</tr>
<tr>
<td>8P</td>
<td>421</td>
</tr>
<tr>
<td>9</td>
<td>171</td>
</tr>
<tr>
<td>11</td>
<td>77</td>
</tr>
<tr>
<td>12</td>
<td>17</td>
</tr>
<tr>
<td>13</td>
<td>199</td>
</tr>
<tr>
<td>14</td>
<td>261</td>
</tr>
<tr>
<td>15P</td>
<td>353</td>
</tr>
<tr>
<td>16</td>
<td>135</td>
</tr>
<tr>
<td>19P</td>
<td>475</td>
</tr>
<tr>
<td>20</td>
<td>205</td>
</tr>
<tr>
<td>25P</td>
<td>443</td>
</tr>
<tr>
<td>26</td>
<td>279</td>
</tr>
<tr>
<td>27</td>
<td>285</td>
</tr>
<tr>
<td>28</td>
<td>119</td>
</tr>
<tr>
<td>30</td>
<td>567</td>
</tr>
<tr>
<td>31</td>
<td>293</td>
</tr>
<tr>
<td>32</td>
<td>185</td>
</tr>
<tr>
<td>33P</td>
<td>507</td>
</tr>
<tr>
<td>34</td>
<td>99</td>
</tr>
<tr>
<td>35P</td>
<td>453</td>
</tr>
<tr>
<td>36</td>
<td>3</td>
</tr>
<tr>
<td>38</td>
<td>157</td>
</tr>
<tr>
<td>40</td>
<td>35</td>
</tr>
<tr>
<td>41</td>
<td>219</td>
</tr>
<tr>
<td>42</td>
<td>335</td>
</tr>
<tr>
<td>44P</td>
<td>461</td>
</tr>
<tr>
<td>45</td>
<td>575</td>
</tr>
<tr>
<td>46P</td>
<td>411</td>
</tr>
<tr>
<td>47P</td>
<td>539</td>
</tr>
<tr>
<td>48</td>
<td>319</td>
</tr>
<tr>
<td>51</td>
<td>211</td>
</tr>
<tr>
<td>52P</td>
<td>369</td>
</tr>
<tr>
<td>53</td>
<td>165</td>
</tr>
<tr>
<td>54P</td>
<td>393</td>
</tr>
<tr>
<td>57</td>
<td>109</td>
</tr>
<tr>
<td>58P</td>
<td>435</td>
</tr>
<tr>
<td>60P</td>
<td>533</td>
</tr>
<tr>
<td>62P</td>
<td>385</td>
</tr>
<tr>
<td>63P</td>
<td>527</td>
</tr>
<tr>
<td>64</td>
<td>191</td>
</tr>
<tr>
<td>65</td>
<td>349</td>
</tr>
<tr>
<td>66P</td>
<td>361</td>
</tr>
<tr>
<td>67P</td>
<td>417</td>
</tr>
<tr>
<td>68P</td>
<td>413</td>
</tr>
<tr>
<td>69</td>
<td>53</td>
</tr>
<tr>
<td>70</td>
<td>85</td>
</tr>
<tr>
<td>71</td>
<td>565</td>
</tr>
<tr>
<td>72P</td>
<td>401</td>
</tr>
<tr>
<td>74P</td>
<td>409</td>
</tr>
<tr>
<td>75</td>
<td>291</td>
</tr>
<tr>
<td>79</td>
<td>267</td>
</tr>
<tr>
<td>80</td>
<td>247</td>
</tr>
<tr>
<td>81</td>
<td>181</td>
</tr>
<tr>
<td>82</td>
<td>303</td>
</tr>
<tr>
<td>83P</td>
<td>515</td>
</tr>
<tr>
<td>85</td>
<td>63</td>
</tr>
</tbody>
</table>

621
HOW TO ORDER IAEA PUBLICATIONS

An exclusive sales agent for IAEA publications, to whom all orders and inquiries should be addressed, has been appointed in the following country:

UNITED STATES OF AMERICA UNIPUB, P.O. Box 433, Murray Hill Station, New York, NY 10157

In the following countries IAEA publications may be purchased from the sales agents or booksellers listed or through your major local booksellers. Payment can be made in local currency or with UNESCO coupons.

ARGENTINA Comisión Nacional de Energía Atómica, Avenida del Libertador 8250, RA-1429 Buenos Aires
AUSTRALIA Hunter Publications, 58 A Gipps Street, Collingwood, Victoria 3066
BELGIUM Service Courrier UNESCO, 202, Avenue du Roi, B-1060 Brussels
CHILE Comisión Chilena de Energía Nuclear, Venta de Publicaciones Amunategui 95, Casilla 188-D, Santiago
CZECHOSLOVAKIA S.N.T.L., Mikulandska 4, CS-116 86 Praha 1
FRANCE Office International de Documentation et Librairie, 48, rue Gay-Lussac, F-75240 Paris Cedex 05
HUNGARY Kultura, Hungarian Foreign Trading Company P.O. Box 149, H-1389 Budapest 62
INDIA Oxford Book and Stationery Co., 17, Park Street, Calcutta-700 016
ISRAEL Heiliger and Co., Ltd., Scientific and Medical Books, 3, Nathan Strauss Street, Jerusalem 94227
ITALY Librería Scientifica, Dott. Lucio de Biasio "aeiou", Via Meravigli 16, I-20123 Milan
JAPAN Maruzen Company, Ltd., P.O. Box 5050, 100-31 Tokyo International
NETHERLANDS Martinus Nijhoff B.V., Booksellers, Lange Voorhout 9-11, P.O. Box 269, NL-2501 The Hague
PAKISTAN Mirza Book Agency, 65, Shahrah Quaid-e-Azam, P.O. Box 729, Lahore 3
POLAND Ars Polona-Ruch, Centrala Handlu Zagranicznego, Krakowskie Przedmiescie 7, PL-00-068 Warsaw
ROMANIA liexim, P.O. Box 136-137, Bucharest
SOUTH AFRICA Van Schaik Bookstore (Pty) Ltd., P.O. Box 724, Pretoria 0001
SPAIN Díaz de Santos, Lagasca 95, E-28006 Madrid
U.S.S.R. Mezhunarodnaya Kniga, Smolenskaya-Sennaya 32-34, Moscow G-200
YUGOSLAVIA Jugoslovenska Knjiga, Terazije 27, P.O. Box 36, YU-11001 Belgrade

Orders from countries where sales agents have not yet been appointed and requests for information should be addressed directly to:

Division of Publications
International Atomic Energy Agency
Wagramerstrasse 5, P.O. Box 100, A-1400 Vienna, Austria