USE OF COLD NEUTRONS FOR CONDENSED MATTER RESEARCH AT THE NEUTRON GUIDE LABORATORY ELLA IN JÜLICH

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ABSTRACT

Cold neutrons produced in the FRJ-2 DIDO reactore are guided into the external hall ELLA. It hosts 10 instruments that are fed by three major neutron guides. Cold neutrons allow for diffraction and small angle scattering experiments resolving mesoscopic structures (1 to 100 nm). Contrast variation by isotopic substitution in chemically identical species yields informations uniquely accessible by neutrons. Inelastic scattering of cold neutrons allows to investigate slow molecular motions because the low neutron velocity results in large relative velocity changes even at small energy transfers. The SANS machines and the HADAS reflectometer serve as structure probes and the backscattering BSS1 and spin-echo spectrometers NSE as main dynamics probes. Besides this the diffuse scattering instrument DNS and the lattice parameter determination instrument LAP deal mainly with crystals and their defects. Finally the beta-NMR and the EKN position allow for methods other than scattering employing nuclear reactions for solid state physics, chemistry and biology/medicine.

1. Introduction

For the research with cold neutrons in Jülich an external neutron guide laboratory ELLA that hosts a number of instruments (see Fig. 1) is attached to the confinement building of the reactor. Thermal neutrons from the D_2O moderator near the FRJ-2 DIDO reactor core diffuse into the liquid H_2 volume ($\approx$ 0.7l) of the cold source and by multiple scattering are cooled to the H_2 temperature. This shifts the Maxwellian spectrum of the neutrons from 2300 m/s to 600 m/s most probable velocity. These neutrons pass a cooled Bi single crystal filter of 50 cm thickness which prevents the core γ- and fast neutron radiation from entering the

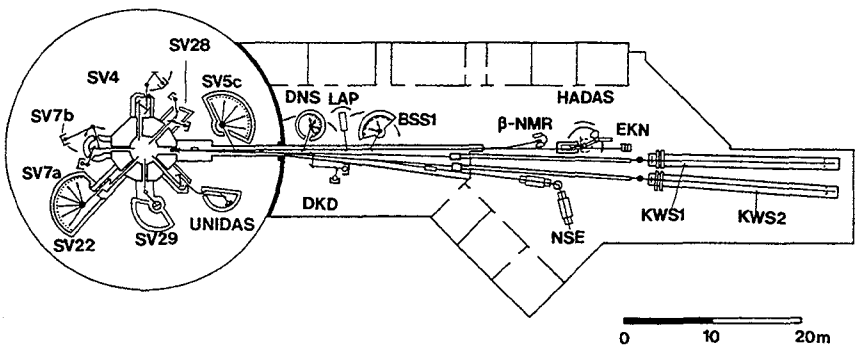

Figure 1: Layout of the DIDO reactor and ELLA with instrument positions.

neutron guide system. Thereby a rather clean beam of "cold" neutrons is provided and fed to the instruments in the guide hall ELLA by a manifold of ^{58}Ni coated neutron guides. The total flux in the guides at their entrance into the ELLA is $4 \cdots 5 \times 10^8 \mathrm{cm}^{-2}\mathrm{s}^{-1}$, after monochromatization and collimation $10^4 \cdots 10^7 \mathrm{neutrons/cm}^2\mathrm{s}$ –depending on the instrument– are left at the sample positions. By the cooling gain factors compared to a thermal beam of more than 20 are achieved for neutrons with velocities below about 500 m/s. These neutrons are especially suited to investigate structures and motions of mesoscopic size, e.g. of macromolecules, molecular aggregates, nano particles and early stages of precipitations and phase separations. The notions "soft matter" and "complex fluids" cover many of the thus treated research topics. Cold neutrons have wavelengths λ from about $5 \cdots 15$Å at the same time –in contrast to X-ray photons– their energy corresponds to low lying vibrational or relaxational molecular excitations. This coincidence enables the simultaneous investigation of structure and dynamics (motions that change/modulate the structure) by neutrons. The other unique advantage of neutrons is the possibility to create contrast and visibility by selective isotopic substitution –especially H/D replacement in soft matter samples. Further due to the cm penetration depth of neutrons and the availability of highly transparent window materials that may have thicknesses of several cm equilibrium samples may be studied under high pressures and/or high and low temperatures. However the associated low interaction cross sections in combination with the limited available neutron fluxes poses several restrictions on the instrument design and use. The art of building neutron scattering instruments consists in guiding as many neutrons from the source via the sample scattering process to the detector. To a large extend this is effected by selecting a resolution as low as compatible with the typical structures in the scattering intensity, since starting from a Maxwellian neutron gas any narrowing of either the directions (divergence) or the used velocity band (wavelength) reduces the available flux of neutrons. Typically a large detecting area (many counting tubes, area detector) is essential for a reasonable data collection rate. Another consequence of the finite luminance of the source emitting non-directed radiation is that the number of neutrons that hit the sample and therefore might contribute to the signal scales with the sample area, i.e. the beam diameter. Since usually all other dimensions scale with the sample size this is the reason for the large size of the typical neutron instruments. It simply results from a compromise of feasibility and cost of enlarging with intensity.

2. Diffraction Instrumentation

The notion diffraction is used for experiments that measure the angular distribution of the scattering without analysing the spectrum of the scattered beam. The information gained corresponds to a "shap-shot" picture of the micro(meso)scopic structure. Whereas diffractometers that aim at the atomistic length scale in liquids and anorganic crystals require a wavelength shorter than typical atom-atom distances (a few Å) are located at "thermal" and "hot" beams, the diffraction instruments in the ELLA guide hall allow for the investigation of large scale structures. Especially the two small angle neutron scattering (SANS) instruments **KWS1** and **KWS2** cover the range from $10 \cdots 1000$Å. This is achieved by the combined effect of the use of long wavelength neutrons $\lambda = 6 \cdots 16$Å and of small scattering angles. The latter measure –together with the sample size argument given in the introduction– yields a very large overall length of about 40m, consisting of 20m collimation length and 20m sample-detector distance. The actual distances may be shortend by (automatic) insertion of neutron guides into the collimation track and moving of the detector to a closer position inside the evacuated flight tube of 1.5m diameter. The detectors have a sensitive area of $60 \times 60\mathrm{cm}^2$ with a spatial resolution of $0.5 \cdots 0.8$cm. One of the largely identical SANS machines is equipped with a FZJ developped ^{6}Li detector the other has a ^{3}He gas counter of the Geesthacht type. The incoming neutrons are filtered by a mechanical velocity selector with a FWHM for $\Delta\lambda/\lambda$ of 10% or 20%. The broad velocity band ensures a high neutron flux at the sample, the resulting broadening of

the resolution is acceptable for most of the investigated problems. Whereas the SANS technique has a wide spectrum of application from the shape distribution of proteins over precipitations in metals, magnetic flux lines in superconductors to particle size distribution in technical powders like soot or concrete, the current mainstream application in Jülich are soft matter problems, all of which rely heavily on the (H/D) contrast variation and matching techniques. The topics extend from the configuration determination of polymer chains in the melt over to the investigation of demixing phase transitions of polymer and block-copolymer melts under variation of thermodynamical parameters as temperature and pressure. Polymer aggregation phenomena in solution (see Fig. 2) as well as structures and phases of microemulsion as well as the microscopic chain deformations due to strain of crosslinked rubber networks are other fields of research. As a supplement to the conventional SANS instruments a so called double crystal spectrometer **DKD** is operated, the spatial resolution of which extends into the range of light microscopy. Such a resolution requires the detection of extremely small scattering angles (a few μradian) which is realized by subsequent reflection of the neutron beam by perfect silicon crystals.

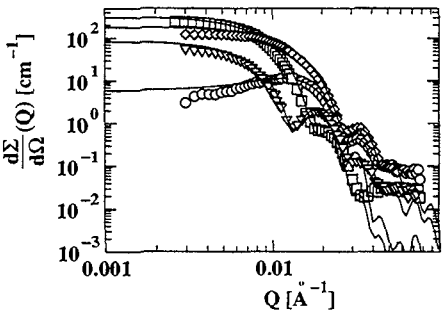

Figure 2: SANS from spherical blockcopolymeraggregates in three different contrasts. Lines correspond to a model fit.

A specialized diffractometer with some degree of spectral analysis is the diffuse neutron "spectrometer" **DNS**, the setup of which (see Fig. 3) resembles a neutron time of flight spectrometer. It utilizes neutrons which are reflected out of one of the guides by a graphite crystal monochromator ($\lambda = 3 \cdots 5$Å). Before hitting the sample the thus prepared monochromatic beam is periodically interrupted by a chopper consisting of a rotating neutron absorbing disc with a transmitting window at its periphery. The resulting neutron bursts are scattered by the sample and the scattered radiation is detected by 56 ^{3}He tubes arranged in a circle around the sample. Currently an option for polarization analysis is installed.

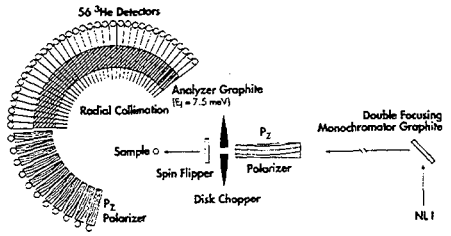

Figure 3: DNS layout

The primary purpose of this instrument is the measurement of diffuse scattering resulting from defects in crystals. A perfect periodic lattice yields scattering intensity only under the very restrictive Bragg condition, both the scattering angle and the crystal orientation must have special values. However if the crystal contains defects –either compositional and/or lattice distortions by interstitials– a low intensity scattering intensity contribution occurs virtually at any angular setting however with a typical smooth intensity distribution. The dependence of this so called diffuse scattering on the angles of scattering and orientation contains the desired information on the types of defects –and after model calculations– on the interaction potentials. The advantage of neutrons for these investigations is the ability to discriminate elastic scattering from inelastic thermal diffuse scattering. The latter is due to transient distortion of the lattice by thermal fluctuation, i.e. phonons. This may bury (e.g. in the X-ray scattering situation) the searched for defect signals if they are not discriminated by their inelasticity. In addition the DNS instrument may be used as a low resolution time-of-fligth spectrometer. The ploarization option will allow for the separation of incoherent and coherent and magnetic scattering contribution in the measurement of structure factors. An important application of this features is the investigation of the spatial correlations in amorphous polymers.

A former triple axis spectrometer **HADAS** has meanwhile be converted into a reflectometer. By using thin slits the monocromatic beam from a graphite crystal monochromator is collimated such that specular reflection from the sample surface at low incident angles (a few degrees). The specular intensity is sensitive to the (scattering length) density profile near ($\approx$ 1000Å) the surface or an buried interface. Polymer surfaces which exhibit scattering length density

variation due to enrichment/depletion of a H/D-labelled component near the surface as well as magnetic layers are investigated. The latter will benefit from a planned installation of neutron polarization analysis.
The high resolution lattice parameter determination setup **LAP** is a dedicated instrument to measure the lattice parameter of typical semiconductor materials –especially GaAs– with an accuracy of 10^{-6} with respect to a reference crystal. The method employs the Doppler shift of the neutron wavelength of Bragg reflected neutrons from a moving perfect crystal at a scattering angle of 180^0. Effects of defects, growing conditions, stoechiometry etc. on the lattice parameters are studied.

3. Spectrometers for high resolution inelastic scattering

Excitations like lattice vibrations, magnons or electronic crystal field transitions correspond to frequencies in the THz domain corresponding to energies in the meV range. Since thermal neutrons have energies in the 10^1meV region the above mentioned inelastic processes lead to a considerable, easily detectable change. However for low lying excitation like tunneling transitions or slow relaxative motions in the samples this change amounts only to energies in the μeV range. Use of incident neutrons with less energy, i.e. "cold" neutrons ($\approx 10^0$ meV), helps to increase the relative effect of these excitations on the neutron velocity. But additional specialized techniques have to be used to achieve the required resolution. The backscattering (π-) spectrometer **BSS1** utilizes the Bragg reflection from perfect silicon crystals at a scattering angle near 180^0 (=backscattering) where the reflected wavelength depends only to second order on the direction therby preserving a narrow wavelength band even for a divergent beam. Preparation of the incoming and the scattered radiation is performed by the same type of $\approx 180^0$ Bragg reflection, scanning of the energy transfer is performed by moving the monochromator crystal utilizing the Doppler shift of the neutron velocity (see Fig. 4).

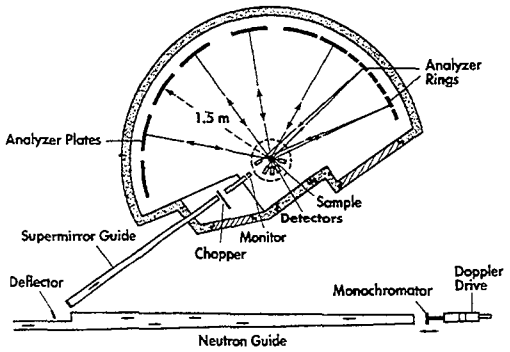

Figure 4: BSS1 layout

Due to the extremely narrow band of velocities selected by the monochromator from the continuous spectrum only a tiny fraction ($\approx 10^{-3}$) of the neutrons reach the sample. Therefor it is necessary to compensate for the loss due to the severe spectral filtering by collecting neutrons from very large solid angles onto a small amount (12) of detectors by a focussing arrangement of the analyzer crystals on 1m sized shperical reflectors that image the sample on one of the counting tubes located close to the sample. A coarse chopper interrupts the primary beam such that neutrons, that made the way from the sample to the analyzer mirrors and then back to the detectors close to the sample, may be discriminated from those, that went directly from the sample to the closeby counting tubes, by their time of flight. Typical experiments include the observation of tunneling spectra –mostly associated with the rotation of CH_3-groups– at low temperatures (< 15K) and their gradual transition from quantum mechanical tunneling to "classical" diffusive reorientation with increasing temperature. Also slow diffusive or relaxative motions –especially of protons with their high incoherent scattering cross section– are observed in a vast variety of different samples. The problems range from hydrogen diffusion in metals, ion motion in materials for electrochemical fuel cells to relaxative motions due to the glassy structure of polymers. The neutron spin-echo spectrometer **NSE** is complementary to the π-spectrometer. It covers roughly the same frequency range, however yielding data in the time domain ($\approx 0.04 \cdots 30$ns) rather than in the frequency space.

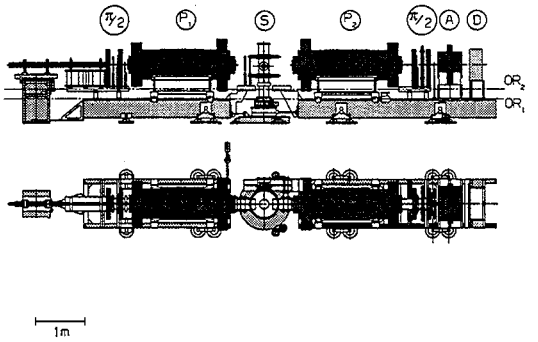

Figure 5: Mechanical layout fo the NSE spectrometer.Most conspicious the two symmetric main precession coils with compensating loops at both ends.

The method to keep reasonable intensity even in a limited solid angle corresponding to small angle scattering and with the detectability of velocity changes $< 10^{-4}$ is done by a tricky manipulation of the neutron spins which are treated as a kind of individual "stop watch" attached to each neutrons. The initially longitudinal polarized beam is extracted from the guide feeding KWS2 by a magnetic FeGe multilayer. Precession in magnetic fields effects the rotation of the "stop watch pointers". Since close to the sample (S) a magnetic element, the π-flipper effectively reverses the "stop watch pointer" (i.e. precession) angle, the passage through a precession track (P_2) exactly symmetric to P_1 before the sample leads to a resulting zero net angle at the end of the track (at the $\pi/2$-flipper). The beam has regained its full polarization, irrespective of the individual starting velocities of the neutrons ! This effect is called spin-echo. Any velocity change at the sample leads to polarization loss in this echo and therefore contains the information on the scattering spectrum. The decoupling of individual starting velocity and velocity change effect allows for the use of a broad (10% $\cdots$ 20% FWHM) incoming wavelength band which yields an intensity advantage of at least 1000 compared to direct filtering. By the use of a large 30cm^2 supermirror analyzer in combination with a matching ^{3}He area detector another data collection rate gain is achieved. For the soft matter and complex fluid research the NSE uniquely opens the field of dynamics to the small angle scattering regime. The investigation of polymer chain dynamics (see Fig. 6), fluctuation in microemulsion and aggregates is largely within the range of NSE and due to its relaxative nature benefits from the fourier transform property. Relaxation data are more readily interpretable in the time domain.

4. Nuclear solid state and chemical research

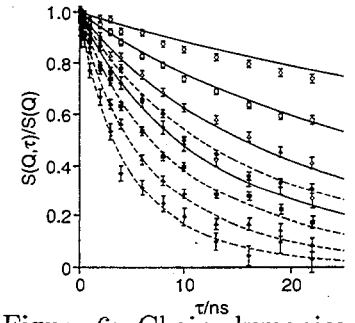

Figure 6: Chain dymanics of a 2.5% polymer solution.

The β-**NMR** spectrometer utilizes the asymmetry of the direction of β-radiation from spin polarized short lived nuclei (e.g. ^{8}Li($T_{1/2} = 0.8$s), ^{12}B($T_{1/2} = 20$ms), ^{20}F($T_{1/2} = 11$s), ^{110}Ag ($T_{1/2}$ =24s), ^{116}In($T_{1/2} = 14$s)). These nuclei are created in the polarized state by capture of a polarized cold neutron by the stable precursor isotope. The sample is located in a homogeneous magnetic field, by the temperature and field dependence of the decay of the β-radiation assymmetry the spin relaxation times of the probe nuclei are investigated. This contributes to the investigations on atomic displacements and diffusion, defect kinetics, spin diffusion, spin-lattice relaxation, phase transitions and interactions with the electrons in the sample. The position **EKN** is a multipurpose position at the end of one of the guides with a flux of 2×10^8cm^2/s over 10×4.8cm^2. Currently the main use is chemical analysis of trace elements by the prompt γ-radiation accompanying virtually all neutron captures.