INTERESTING RESULTS FROM NEUTRON SCATTERING

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Neutron scattering was first recognized as a powerful tool for condensed-matter physics studies in the late 1940,s. One of its first big triumphs was the direct confirmation of antiferromagnetic structures and another, the location of hydrogen atoms in crystals. Such measurements were done with low-flux neutron sources and tended to skim off the cream while leaving many interesting problems untouched. The higher-flux facilities which were rapidly being developed addressed themselves to increasingly difficult new challenges leaving behind a host of interesting unsolved problems which were very suitable for a student-oriented low-flux facility.

In this talk, which makes no claim to being comprehensive, I want to show you a few examples of diverse results which have appealed to me for a variety of reasons. Some of the results appealed to me because I was involved in their acquisition, others because I needed them before I could proceed with my own research and still others because they represent pretty examples of interesting phenomena. Some of these results are old, others are quite recent.

ONE OF THE FASCINATING TOPICS STUDIED EXTENSIVELY BY NEUTRON SCATTERING OVER THE YEARS HAS BEEN THE LATTICE DYNAMICS OF METALS AND THE ROLE PLAYED BY THE CONDUCTION ELECTRONS. UNTIL THE NEUTRON SCATTERING EXPERIMENTS WERE DONE IN THE LATE 1950'S IT WAS SERIOUSLY DEBATED WHETHER OR NOT WELL-DEFINED NORMAL MODES OF VIBRATION EXISTED IN METALS. MOST OF THE EARLY PIONEERING WORK IN THIS FIELD WAS DONE BY BROCKHOUSE. HE NOT ONLY INVENTED THE POWERFUL AND NOW WORLD-WIDE-USED CONSTANT-MOMENTUM-TRANSFER TECHNIQUE DURING THE COURSE OF THESE STUDIES, BUT HE ALSO SHOWED THAT THE INTERATOMIC FORCE SYSTEMS IN LEAD, FOR EXAMPLE, WAS UNEXPECTEDLY LONG RANGED. THE FIRST SLIDE SHOWS THE DISPERSION CURVES FOR LEAD. WE NOW HAVE A MUCH BETTER UNDERSTANDING OF THE PHYSICS BEHIND THESE LONG RANGE FORCES ALTHOUGH THE DETAILED CALCULATIONS STILL STIR SPIRITED DISCUSSION AT INTERNATIONAL MEETINGS. I WOULD LIKE TO SHOW YOU A SIMPLE SLIDE OF SOME OLD MEASUREMENTS WE DID AT CHALK RIVER WHICH ILLUSTRATES THE WEALTH OF DETAIL CONTAINED IN SOME OF THESE MEASUREMENTS. THESE RESULTS ARE FOR THREE DIFFERENT METALS ALL OF WHICH HAVE THE SAME STRUCTURE, SODIUM, NIOBIUM AND MOLYBDENUM - THEY ARE BODY-CENTRED CUBIC WITH ONE ATOM PER PRIMITIVE UNIT CELL. NATURE DOES NOT

SLIDE 1

SLIDE 2

PROVIDE US WITH ANY SIMPLER CRYSTALS. THIS SLIDE SHOWS THE FREQUENCY VS WAVELENGTH RELATIONSHIP FOR LONGITUDINAL WAVES PROPAGATING ALONG THE CUBE-EDGE DIRECTION. THE ABSCISSA IS ACTUALLY THE LATTICE SPACING DIVIDED BY THE WAVELENGTH.

THE CURVES START OUT FROM THE ORIGIN AS STRAIGHT LINES, A REFLECTION OF THE FACT THAT AT LONG WAVELENGTHS THE VELOCITY OF THE WAVES, WHICH ARE JUST SOUND WAVES, IS INDEPENDENT OF FREQUENCY. WHEN THE WAVELENGTH IS COMPARABLE WITH THE INTERATOMIC SPACING HOWEVER, THE SHAPES OF THE CURVES CHANGE AND THIS IS WHERE THE INTERESTING PHYSICS LIES. THE CURVE FOR SODIUM IS VERY NEARLY A SIMPLE SINE CURVE AND IS REMINISCENT OF THE CALCULATED CURVES FOR THE SIMPLE SYSTEMS TREATED IN ELEMENTARY TEXT BOOKS ON SOLID-STATE PHYSICS. THE CURVES FOR NIOBIUM AND MOLYBDENUM HAVE COMPLICATED SHAPES. NIOBIUM AND MOLYBDENUM ARE BOTH TRANSITION METALS, THEY HAVE SIMILAR MASSES, SIMILAR LATTICE SPACINGS, AND ARE NEIGHBORS IN THE PERIODIC TABLE. THE PRINCIPAL DIFFERENCE BETWEEN THEM IS THAT MOLYBDENUM HAS AN EXTRA ELECTRON AND THUS ITS FIRMI LEVEL IS DIFFERENT. IT DOES NOT TAKE MUCH IMAGINATION TO APPRECIATE THAT THE PRESENCE OF THIS EXTRA ELECTRON DRASTICALLY ALTERS THE WAY IN WHICH THE ATOMS VIBRATE.

THEORETICIANS ARE STILL DEBATING HOW THESE RESULTS ARE RELATED TO THE VERY DIFFERENT SUPERCONDUCTING PROPERTIES OF THESE TWO METALS AND THE PROS AND CONS OF THE VARIOUS THEORETICAL MODELS PROPOSED TO EXPLAIN THE LATTICE DYNAMICS OF THESE METALS. TWO RELATED INTERESTING DEVELOPMENTS HAVE COME ABOUT MORE RECENTLY. AT THE HFBR IN BROOKHAVEN, HIGH-RESOLUTION EXPERIMENTS WERE DONE TO SHOW THAT THE PHONONS (THE NAME GIVEN TO QUANTA OF LATTICE

SLIDE 3

VIBRATIONAL ENERGY) IN NIOBIUM CHANGE BOTH THEIR ENERGIES AND WIDTHS WHEN THE METAL BECOMES SUPERCONDUCTING. PROFESSOR BROCKHOUSE AND I HAD LOOKED FOR A SIMILAR EFFECT IN LEAD IN 1961 BUT COULD DETECT NOTHING SIGNIFICANT. JUST LAST YEAR A GROUP WORKING AT THE HFBR REACTOR AT OAK RIDGE REPORTED A VERY STRIKING EFFECT ON THE DISPERSION CURVE

SLIDE 4

FOR BODY-CENTRED CUBIC ZIRCONIUM WHICH MAY BE RELATED TO THE TRANSITION TO THE HEXAGONAL CLOSE-PACKED STRUCTURE WHICH OCCURS AT LOWER TEMPERATURES.

ONE OF THE MOST FASCINATING AREAS FOR NEUTRON SCATTERING HAS BEEN IN THE DETERMINATION OF MAGNETIC

SLIDE 5

STRUCTURES AND THE EXCITATIONS IN THESE STRUCTURES. THE NEXT SLIDE SHOWS A RESULT NOT AMENABLE TO ANY OTHER TECHNIQUE. IT IS A VERY OLD DIFFRACTION PATTERN TAKEN AT

TWO DIFFERENT TEMPERATURES ON THE NRX REACTOR AT CHALK RIVER ON A POWDER SAMPLE OF UO_2. AT THE LOWER TEMPERATURE THERE ARE EXTRA PEAKS WHICH CONFIRM THE ANTIFERROMAGNETIC NATURE OF UO_2. MATERIALS SUCH AS RARE EARTHS OFTEN HAVE VERY MUCH MORE COMPLICATED MAGNETIC STRUCTURES.

SLIDE 6 IN A SERIES OF FASCINATING EXPERIMENTS IN THE EARLY 1960'S THE OAK RIDGE GROUP DISCOVERED THE SPIRAL AND CONE STRUCTURES CHARACTERISTIC OF SOME OF THESE ELEMENTS. THE EXCITATIONS IN THESE STRUCTURES WERE STUDIED AT OAK RIDGE, CHALK RIVER

SLIDE 7 AND RISØ. THE NEXT SLIDE SHOWS SOME RESULTS OBTAINED AT CHALK RIVER FOR ERBIUM, ONE OF THE MORE DIFFICULT EXPERIMENTS WE HAD TACKELED UP TO THAT TIME MAINLY BECAUSE

SLIDE 8 OF ERBIUM'S VERY HIGH NEUTRON ABSORPTION CROSS SECTION. THE NEXT SLIDE SHOWS SOME RESULTS FROM RISØ ILLUSTRATING HOW THE CURVE FOR THE C-DIRECTION IN TERBIUM CHANGES WITH TEMPERATURE. THESE ARE BIG CHANGES AND HAVE BEEN ANALYZED TO TELL US A LOT ABOUT THE PHYSICS OF THE MAGNETISM IN RARE-EARTH SYSTEMS - THESE RESULTS WERE ALL OBTAINED ON REACTORS WITH NEUTRON FLUXES EXCEEDING 10^{14} NEUTRONS $.CM^{-2}$ S^{-1}; IT IS UNLIKELY THAT THE TRIUMF SOURCE COULD MAKE A SIGNIFICANT CONTRIBUTION. ON THE OTHER HAND, IF A LARGE SINGLE CRYSTAL OF THE APPROPRIATE ISOTOPE OF ERBIUM, FOR EXAMPLE, SUCH AS THE ONE USED AT OAK RIDGE, COULD BE OBTAINED, SOME STRUCTURE WORK WOULD NO DOUBT BE POSSIBLE

AND THIS COULD FORM A USEFUL BASIS FOR FURTHER WORK AT ANOTHER LABORATORY. IN MY OPINION THERE IS STILL PLENTY OF SCOPE FOR INGENUITY AT LOW-FLUX INSTALLATIONS. WE ONLY HAVE TO LOOK AT SOME OF THE FASCINATING WORK DONE AT KJELLER, IN NORWAY, BY RISTE AND HIS GROUP TO GET AN IDEA OF THE POSSIBILITIES.

AN EXAMPLE OF AN INTERESTING EXPERIMENT DONE IN RECENT YEARS AT CHALK RIVER BY POWELL AND MARTEL IS THE STUDY OF STRUCTURAL CHANGES IN 1-METHLYTHYMINE, ONE OF THE BASES OF DNA, AS A RESULT OF IRRADIATION BY ULTRA-
SLIDE 9 VIOLET LIGHT. YOU CAN SEE FROM THE CURVES THAT THE EFFECT OF THE IRRADIATION IS TO SHIFT THE POSITIONS OF SOME PEAKS, MAKE OTHERS DISAPPEAR, AND GENERALLY TO CHANGE THE WHOLE DIFFRACTION PATTERN.

SOME OTHER DNA BASES SHOWED NO CHANGE WITH IRRADIATION INDICATING THAT LITTLE, IF ANY, DAMAGE OCCURRED. THESE RESULTS ARE INTERESTING AND HOPEFULLY WILL INPUT INTO OUR EVENTUAL BASIC UNDERSTANDING OF RADIATION DAMAGE IN DNA.

ANOTHER INTERESTING SYSTEM BEING STUDIED AT CHALK RIVER, AND IN OTHER LABORATORIES AROUND THE WORLD, IS THAT OF PLASTIC CRYSTALS. PLASTIC CRYSTALS ARE ONES WHICH CONSIST OF COMPLICATED MOLECULES SITTING ON WELL-DEFINED

SITES, JUST AS IN AN ORDINARY CRYSTAL, BUT THE MOLECULES ARE FREE, OR NEARLY FREE, TO ROTATE; THUS THE POSITIONS OF THE INDIVIDUAL ATOMS WITHIN THE MOLECULES ARE NOT WELL-DEFINED AND, HENCE, THE CORRESPONDING DIFFRACTION PATTERN SHOULD BE MORE LIKE THAT FROM A LIQUID, I.E. BROAD PEAKS INSTEAD OF THE SHARP PEAKS CHARACTERISTIC OF A SOLID. SOME

SLIDE OF THESE PLASTIC CRYSTALS, CBr_4 FOR EXAMPLE, HAVE DIFFRACTION PATTERNS WHICH ARE A MIXTURE OF THE TWO - SHARP PEAKS AND BROAD PEAKS AS SHOWN ON THE SLIDE. VARLEY SEARS OF CHALK RIVER DISCUSSED THESE RESULTS THIS MORNING AND I ONLY MENTION THEM TO EMPHASIZE THAT THERE IS STILL A GREAT DEAL OF INTERESTING STRUCTURAL WORK MUCH OF WHICH CAN BE DONE WITHOUT HAVING VERY HIGH NEUTRON FLUXES AVAILABLE.

ANOTHER VERY ACTIVE FIELD WHICH HAS PRODUCED FASCINATING RESULTS IS THAT OF STRUCTURAL PHASE CHANGES IN FERROELECTRIC-TYPE MATERIALS. IN MANY CASES CERTAIN MODES OF VIBRATION DECREASE IN FREQUENCY AS THE TEMPERATURE IS LOWERED TOWARDS THE TRANSITION POINT AND THE MODE BECOMES UNSTABLE, THUS TRIGGERING THE PHASE TRANSITION. THE FIRST EXPERIMENT DEMONSTRATING SUCH AN EFFECT WAS DONE BY COWLEY AT CHALK RIVER AND SINCE THEN THERE HAS BEEN A LOT OF ACTIVITY IN THIS FIELD, PARTICULARLY AT CHALK RIVER AND BROOKHAVEN.

FINALLY, I WANT TO TALK A LITTLE ABOUT MY FAVORITE NEUTRON-SCATTERING SPECIMEN, LIQUID HELIUM. IT WAS IN 1957

THAT PALEVSKY FROM BROOKHAVEN, WORKING IN STOCKHOLM WITH LARSSON AND HIS GROUP, PUBLISHED THE RESULTS OF THEIR NEUTRON-SCATTERING EXPERIMENTS GIVING THE FIRST DIRECT EVIDENCE OF THE EXISTENCE OF WELL-DEFINED EXCITATIONS IN SUPERFLUID LIQUID HELIUM-4. PROBABLY NO OTHER SINGLE ELEMENT HAS BEEN SO EXTENSIVELY STUDIED BY NEUTRON SCATTERING SINCE THAT TIME. WE HAVE DONE A LOT OF WORK AT CHALK RIVER AND MAJOR CONTRIBUTIONS HAVE BEEN MADE IN SWEDEN, AT LOS ALAMOS, BROOKHAVEN, OAK RIDGE, ARGONNE, INSTITUT LAUE-LANGEVIN AND DUBNA. IT IS THE EXCITATIONS NEAR THE ROTON MINIMUM, WHICH ARE RESPONSIBLE FOR MUCH OF THE THERMODYNAMIC AND HYDRODYNAMIC BEHAVIOUR OF SUPERFLUID HELIUM-4, THAT HAVE BEEN STUDIED MOST THOROUGHLY. THE VARIOUS MEASUREMENTS OF THE ROTON ENERGY CARRIED OUT IN THE LATE 1950'S AT STOCKHOLM, LOS ALAMOS AND CHALK RIVER WERE REALLY VERY PRECISE BUT THE NEED FOR EVEN MORE PRECISE MEASUREMENTS BECAME VERY APPARENT WHEN THE BEAUTIFUL LIGHT-SCATTERING MEASUREMENTS OF GREYTAK AND HIS CO-WORKERS AT MIT INDICATED THAT TWO ROTONS COULD BE BOUND AND THAT THE WEAK LINK IN DETERMINING THE BINDING ENERGY WAS THE NEUTRON RESULT WE HAD BEEN SO PROUD OF.

IN A COLLABORATIVE EXPERIMENT BETWEEN CHALK RIVER AND THE INSTITUT LAUE-LANGEVIN WE CARRIED OUT WHAT I BELIEVE

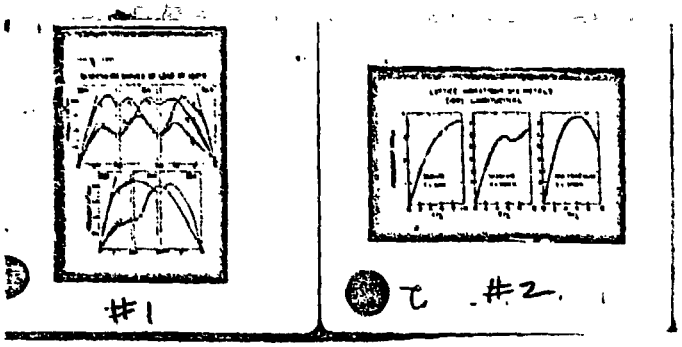
IS THE MOST PRECISE INELASTIC NEUTRON-SCATTERING MEASURE-
SLIDE MENT DONE USING CONVENTIONAL TECHNIQUES. THE NEXT SLIDE SHOWS THE MEASUREMENTS NEAR THE ROTON-MINIMUM SECTION OF THE DISPERSION CURVE. THE DETAILS ARE NOT IMPORTANT BUT THE RESULT IS A VERY GOOD EXAMPLE OF INTERNATIONAL CO-OPERATION IN THIS FIELD AND THE USE OF UNIQUE FACILITIES IN OTHER LABORATORIES. IN ORDER TO MAKE THE BEST USE OF SUCH FACILITIES IT IS ESSENTIAL TO HAVE AN ACCESSIBLE FACILITY CLOSE TO HOME WHERE SUFFICIENT FAMILIARITY WITH THE NECESSARY TECHNIQUES CAN BE OBTAINED.

I AM CONVINCED THAT THE TRIUMF FACILITY CAN MAKE AT LEAST TWO VERY USEFUL CONTRIBUTIONS TO THE NEUTRON-SCATTERING FIELD:

1. PROVIDE THE TRAINING AND EXPERTISE NECESSARY TO MAKE EFFICIENT USE OF HIGHER-FLUX INSTALLATIONS.

2. WITH THE APPLICATION OF A LITTLE INGENUITY, DO SOME COMPLETE EXPERIMENTS FOR WHICH NO APOLOGIES HAVE TO BE MADE.

I WISH TONY ARROTT AND HIS COLLEAGUES GREAT SUCCESS WITH THIS VENTURE AND I FEEL CERTAIN THAT IT WILL BE ONE TO BE PROUD OF.



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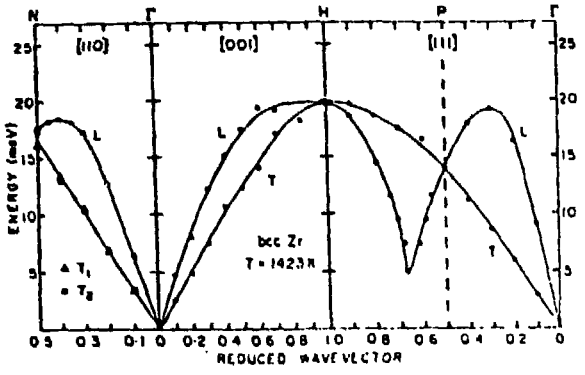

FIG. 1. Dispersion curves along the [100], [110], and [111] symmetry direction of bcc Zr at 1423 K. The frequencies obtained for the two crystals examined in these experiments (see text) were found to agree to within experimental errors. The solid lines were drawn as a guide to the eye.

#4

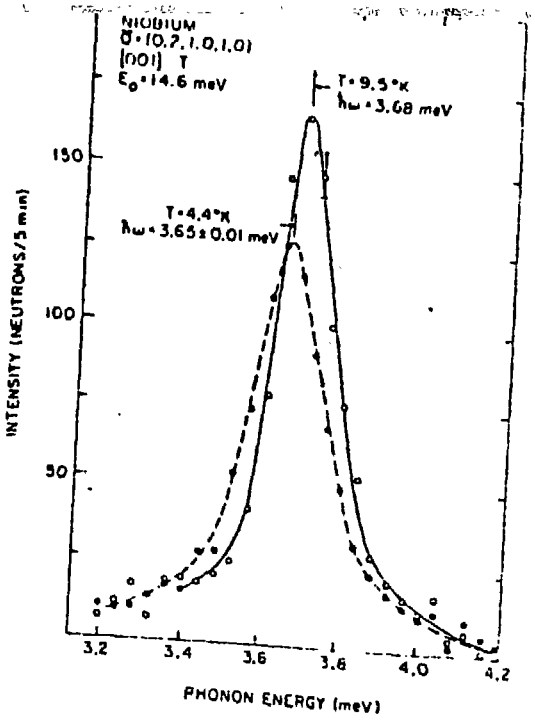

FIG. 11. Scattered-neutron spectra of the [00ζ]T, $\zeta = 0.2$ phonon for $T > T_c$ (solid line) and $T < T_c$ (dashed line) showing decrease in energy with decrease in temperature.

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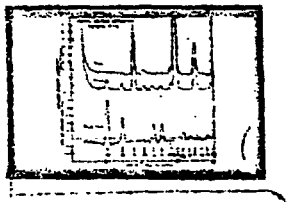

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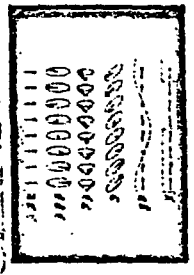

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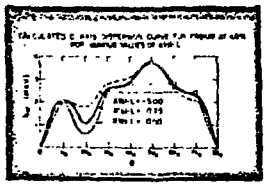

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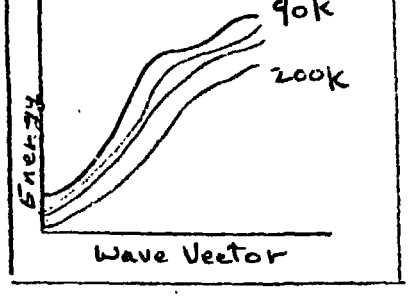

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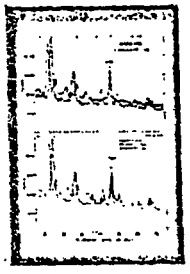

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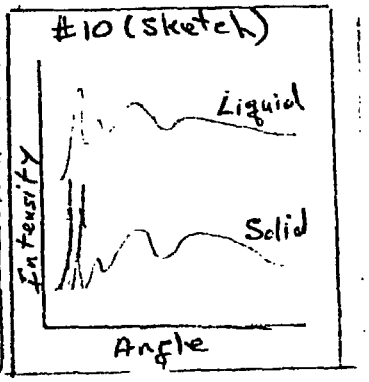

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