The First Decade of Neutron Scattering at Chalk River: 1949-59

by Thomas M. Holden

The completion of the National Research Experimental Reactor, NRX, at Chalk River in 1947 set the stage for nuclear research in Canada. NRX provided space for in-reactor irradiations as well as external beams of neutrons and γ-rays for experiments outside the biological shielding. The earliest neutron beam experiments were aimed at understanding not only the structure and dynamics of solids liquids and gases but also nuclear structure and fission processes. According to G.C. Hanna in [1], "W.B. Lewis (then director of Chalk River)…left the scientific direction of pure research to the scientists". It appears that the staff scientists were given the initiative to carry out the research that seemed most likely to them to lead to advances in science and technology.

According to A.D.B. (Dave) Woods in a private communication, "There is no doubt in my mind that Don Hurst (then director of the Reactor Research and Development Division) was the inspiration behind the neutron scattering program at Chalk River". In the late 1940s Hurst worked with Norman Z. Alcock and John A. Spiers on neutron scattering from gases and on nuclear physics problems. To augment the program, Hurst hired Bertram N. Brockhouse in 1950 and Dave G. Henshaw in 1951. The goal of measuring the detailed vibrational spectrum of solids by inelastic neutron scattering was formulated in 1950 in study group meetings in Hurst's home between himself, Brockhouse, G. H. (Trudi) Goldschmidt and Noel K. Pope.

The first monochromatic neutron beams were extracted from NRX by diffraction from natural crystals such as NaCl and shortly afterwards a second axis was added to create a diffractometer [2]. The beam had a cross section of 2.5 × 1.25 cm^2 to maximize the intensity and the distance from the monochromator to the sample was 183cm to allow low diffraction angles. The monochromator was surrounded by massive shielding which was required for adequate reduction of unwanted background intensity. With care, the lower and upper limits on neutron energy were 1 meV and 50 eV!

Summary

Contributions from Chalk River Nuclear Laboratories in the first decade, 1949-59, to neutron scattering research in Canada are described.

The first experiment on the scattering of neutrons [3] described diffractometer measurements of pressurized O_2 and CO_2 with 70 meV (1.06 Å) neutrons selected by a NaCl monochromator from the NRX reactor spectrum. The experiment was designed to elucidate the atomic structure of the molecules and the measurements were compared with a calculation based on the scattering of the neutron by systems of two and three point entities assuming no inelastic scattering. The calculation for O_2 matched the variation of the scattered intensity with angle reasonably well, but lay above the experiment for CO_2 at low momentum transfers. It was surmised that, since the CO_2 gas was quite close to the liquid phase, some condensate in the sample chamber might have changed the scattering pattern. The experiments were clearly right at the limit of what could be done at that time and great care had to be taken to avoid systematic errors.

The second experiment reported [4] was on the scattering lengths of the deuteron where the interest was on the impact on the theory of nuclear forces rather than atomic physics. 1.063 Å neutrons from the NRX reactor were selected by reflection from the (100) planes of NaCl. The scattering of slow neutrons by the deuteron of nuclear spin 1 is defined by the two possible spins of the compound nucleus, 3/2 or 1/2. The form of the scattering is a quadratic function of the ratio $\frac{a_{1/2}}{a_{3/2}}$, which had two roots 3.2 ± 0.3 or 0.12 ± 0.04 where $a_{1/2}$ and $a_{3/2}$ are the relevant scattering lengths. As noted in their paper, the remaining ambiguity could only be resolved by experiments with polarized neutrons. The presently accepted values of the scattering lengths [5] of the deuteron are $a_{3/2}$ = 9.53 ± 0.03 fm and $a_{1/2} = 0.975 \pm 0.06$ fm and their ratio $\frac{a_{1/2}}{a_{3/2}}$ is 0.102 ± 0.007. This modern result is within the careful error estimates made by Hurst and Alcock in 1951 for the second of their quoted values. The experiment is a textbook case of considering the sources of systematic and random errors and assigning uncertainties to the measured quantities and carrying these through to the final ratios.

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The first experiment with an impact on solid state physics emerged from measurements of the resonant scattering of slow neutrons by Cd. The one-to-one correspondence between the intensity of neutrons transmitted by Cd near 350 meV and the neutron wavelength led to its use as an analyzer to test the average wavelength of neutrons scattered by Pb, Al and C. The significance of the experiment for the possibility of a measurement of inelastic scattering was recognized by Brockhouse in his Nobel address [6]. The intensity of neutrons scattered by Pb, Al or C was measured for each of ten thicknesses of Cd covering an assembly of six symmetrically arranged BF_3 counters. The actual transmission through Cd showed deviations from the transmission calculated on the basis of elastic scattering alone that correspond to a net increase in neutron energy upon scattering. The deviations increase in the order Pb, Al, C corresponding to larger energy transfers to the neutron as the mass of the scattering nucleus decreases. An Einstein model of the vibrational energy levels of a monatomic solid using reported Einstein temperatures of 65.5, 300.4 and 1740K (5.65, 25.9 and 150.0 meV) for Pb, Al and C, respectively, gave a satisfactory account of the net change in wavelength on scattering from these materials. Significantly, it suggested that the possibility of measuring energy transfers directly was close to being feasible.

The first neutron measurements from Chalk River on the structure of liquid He^4 were reported by Henshaw and Hurst in 1953 [7]. The intensities, corrected for background and the change of effective volume with scattering angle, were accurate to ±7% while double scattering and the effect of resolution were noted to be smaller than the statistical error. The main finding was the position of the first peak in S(Q) at 2.15 ± 0.0.11 $Å^{-1}$ corresponding to a peak in the radial distribution function of atoms, g(r), at 3.6 Å. Further measurements [8] of the intensities normalized by the differential scattering cross section of a free He^4 atom indicated that the coherent scattering, which reveals the liquid structure, was restricted to the peak around 2.057 $Å^{-1}$ and that at greater wave vectors the scattering was essentially free atom scattering. The principal result of the temperature variation of the coherent scattering was that there was no change in the structure of He^4 at the λ-point, where He^4 turns into a superfluid. This was not unexpected since the λ-transition was thought to be essentially a Bose-Einstein condensation in momentum space which would result in a change in the velocity distribution of the atoms but no change in the spatial distribution.

By 1955, large Al single crystals grown by Henshaw had replaced natural single crystals as monochromators providing major improvements in monochromatic beam intensities. The first reports of inelastic neutron scattering using the "crude triple-axis crystal spectrometer", shown in Fig. 1 [6], came in 1955 with the publication of "The scattering of neutrons by phonons in an Al single crystal" [9] by Brockhouse and Alec T.Stewart, which was the culmination of the study group discussions held four years previously. Bragg scattering is far stronger than phonon scattering, but when the single crystal sample is angled to avoid Bragg scattering, the phonons may be observed. When the conditions of wave vector conservation and energy conservation are met simultaneously, a peak may be seen in the scattered intensity plotted as a function of the energy transfer to the crystal. For a monatomic lattice the frequencies are expected to separate into three branches, corresponding in the low frequency limit, to the familiar longitudinal and two transverse sound waves.

In the experiment, neutrons of constant wavelength 1.148 Å (62.2 meV) from a crystal spectrometer fell on an Al sample crystal with [01 $\bar{1}$] axis vertical. Measurements were made with

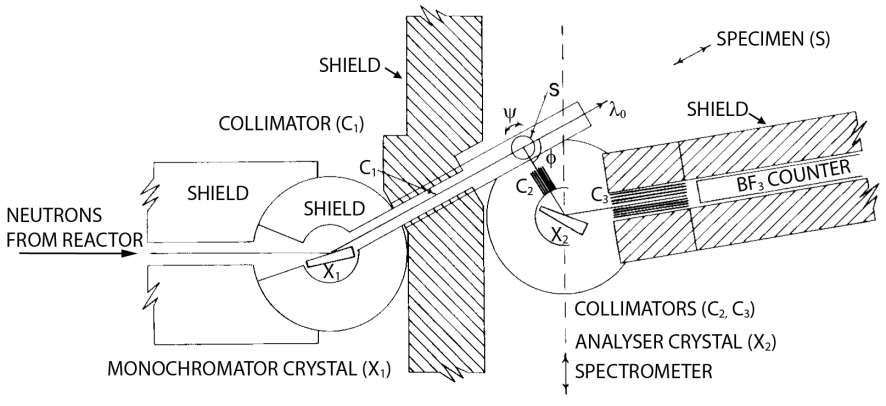

Fig. 1 The first crude version of the triple-axis spectrometer. Monoenergetic neutrons are selected by the large single crystal monochromator (X_1) and impinge on the specimen (S) which is located on a table whose orientation (ψ) with respect to the incident beam can be selected. This table can be moved along the incident beam as desired. The analyzing spectrometer, which employs crystal X_2, is a diffractometer of especially large aperture which can be translated as a unit. The angle, Φ, through which the examined neutrons are scattered, is determined by triangulation. Thick shielding, shown hatched, between X_1 and S, blocks unwanted background neutrons and γ-rays. Thick shielding also surrounds the BF_3 counter. Collimators C_1, C_2 and C_3 determine the precise direction of the neutrons.

the sample crystal angle rotated by increments, δ, from the angle for Bragg scattering for the (333) reflection**.** The energy distribution was measured with a second crystal spectrometer and revealed neutron peaks in both neutron energy loss and gain.

Four points on the TA branch were established and one on the LA branch shown in Fig. 2. The position of the TA phonons matched a sinusoidal curve with an initial linear slope corresponding to the transverse velocity of sound in Al. The authors noted that similar inelastic neutron scattering experiments were being carried out by B. Jacrot in Paris and the group of R.S. Carter, D.J. Hughes, H. Palevsky and R.L. Zimmerman at Brookhaven in the United States.

A more complete series of experiments on the lattice vibrations in Al was reported in the epochal paper [10] in *Reviews of Modern Physics* in 1958. Enormous progress had been made in the intervening four years in obtaining the phonon dispersion relations in three high symmetry directions and checking that the observed results were in agreement with the cross section and selection rules for one-phonon and two-phonon processes. Two different crystals were examined both by crystal and time-of-flight spectrometry. The various processes that give accidental sharp peaks that masquerade as excitations and are usually referred to as "spurions" were identified. Finally, the results were discussed in terms of the existing theories of lattice dynamics and compared with the results of diffuse x-ray experiments. That is, all possible checks were made to establish that the inelastic scattering observed was due to lattice vibrations.

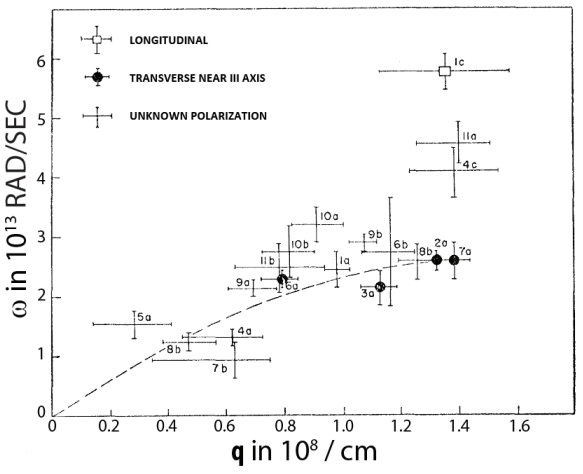

Fig. 2 The relation between the phonon frequency, ω (10^{13} radians/sec) and the phonon wavevector, q (10^{8} cm^{-1}), near the [111] direction in aluminium (Al). Positively identified transverse (TA) and longitudinal (LA) modes are indicated by solid and open circles. Phonons of unknown polarization are indicated by dots. The dashed curve is a sine curve whose initial slope corresponds to a velocity of sound of 3080 m/s for Al.

The first measurements of the dispersion relation, $\hbar\omega(q)$, for magnetic excitations in an ordered magnet Fe_3O_4, were reported [11] in 1957. To put the paper in context, it was generally thought that the excitations out of the fully ordered magnetic state were wave-like by analogy with the vibrations of the crystal lattice. However, the only manifestations of the excitations were found from thermodynamic average measurements like magnetization and low temperature specific heat. The sample was a large natural single crystal with about 10% of the Fe sites replaced by vacancies, and impurities such as Ti, Mn, Al, and Si. There are six interpenetrating magnetic sub lattices in Fe_3O_4, and so there is one "acoustic" mode which goes to zero energy at $\mathbf{q} = 0$ and five "optic" branches. In spite of the large coherent phonon scattering and large incoherent scattering from the impurities, the spin-wave dispersion relation was established over an energy range of 16 meV and wave vectors up to 0.04 $Å^{-1}$. Over this small range, it was not possible to decide on the basis of the initial experiments whether the dispersion relation was linear or quadratic. However, a straight line through the data did not correspond to the velocity of sound in magnetite. For a quadratic dispersion relation, J_{AB}, the exchange parameter between the A and B sites was 2.0 meV and with this value, the calculated Curie temperature on a molecular field model was 1050K compared with the actual value of 850K. On this physical basis, it was concluded that a quadratic dispersion relation was more reasonable. In a paper written shortly afterwards, Brockhouse showed that the intensity ratio of the spin wave scattering with and without a magnetic field was 1.42 ± 0.05 compared with the theoretical value of 1.5 thus confirming the magnetic character of the excitations.

Measurements of the energy-momentum relation in liquid He^4 were reported [12] in 1958 by time-of-flight with a rotating crystal spectrometer and the Chalk River filter chopper spectrometer. The results agreed with previous neutron time-of-flight measurements and followed a form consistent with a linear phonon relation at small wave vectors and a roton minimum around 0.7 meV. These experiments marked the beginning of a series of experiments on the inelastic scattering from He^4 over a period of 50 years by Henshaw, Woods, Eric C. Svensson and collaborators.

1957 was an important year for neutron research in Canada since the National Research Universal, NRU, reactor started in November with a power of 200MW and thermal neutron flux of 3×10^{14} neutrons cm^{-2} sec^{-1}, about a factor of 10 higher than NRX. A newly designed triple-axis spectrometer [13] was rapidly deployed. In 1958 A.D.B. Woods was hired by Brockhouse who was embarking on studies of phonons in semiconductors, metals and alkali halides. At this point, measurements of dispersion relations in high symmetry directions had to be made by an iterative process since there was no guarantee that measurements with a fixed incident energy, E, and varying E′ to measure the scattered peak would yield a phonon wave vector on a symmetry direction. Woods, in a private communication, described the discovery of the constant-**Q** method in this way:

> *I remember the Monday morning that Bert came in and announced his idea of the constant-**Q** method of observing phonons. A few weeks earlier R.G. Stedman from Sweden had arrived at Chalk River to work for a year in P.A. Egelstaff's United Kingdom Atomic Energy group on scattering from neutron moderators. Dr. Stedman had explained to us attempts made in Sweden to observe phonons in NaCl on the initial steep branch of the dispersion relation by moving at constant energy transfer across the curve. Bert brilliantly clued into this and realized that if you could control the angle of scattering and the sample crystal orientation along with the energy transfer you could do a scan without changing the momentum transfer [hence constant-**Q**].*

This immediately changed the speed and accuracy of measurements of inelastic scattering in crystals and led to an enormous outpouring of papers on phonons in metals and semiconductors, Kohn anomalies, magnetic excitations, paramagnetic scattering, liquids, crystal fields and an unprecedented understanding of the physics of materials. Brockhouse was awarded the Nobel Prize in Physics in 1994 for his formidable achievements in solid state physics, for the development of the triple axis crystal spectrometer and for the constant-**Q** technique. The measurements made in the first decade at Chalk River led the world in neutron scattering and were remarkable, as in later work, for their ingenuity and rigorous attention to sources of experimental error.

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