

Today's increasing effort to understand condensed matter using neutrons was largely defined by the pioneering work of C.G. Shull and B.N. Brockhouse, the winners of the 1994 Nobel Prize in Physics, that is now part of every elementary textbook on the subject.

# Probing Matter with Neutrons

## Where Atoms Are

The idea that magnetism is derived from arrangements of unpaired electrons in a solid goes back to the 1920s, but Cliff Shull, together with the late Ernie Wollan, were the first to demonstrate that the exact arrangement of these microscopic "magnets" located on atomic sites could be determined by neutron scattering. The first antiferromagnetic structure examined was that of MnO (see Fig. 1) reported in 1949. The theory for the interaction of the neutron and the magnetic moments derived from the unpaired electrons in solids had been worked out by Halpern and Johnson in 1939, but it was by no means clear how to perform these experiments, or how to separate the weak magnetic interaction from the stronger interaction that the neutron has with the nuclei. The experiments verified directly the ideas of Néel (Nobel Prize, 1970) that underpin our understanding of magnetic arrangements at the microscopic level. Forty-five years, later neutron scattering is still the technique of choice for determining magnetic arrangements.

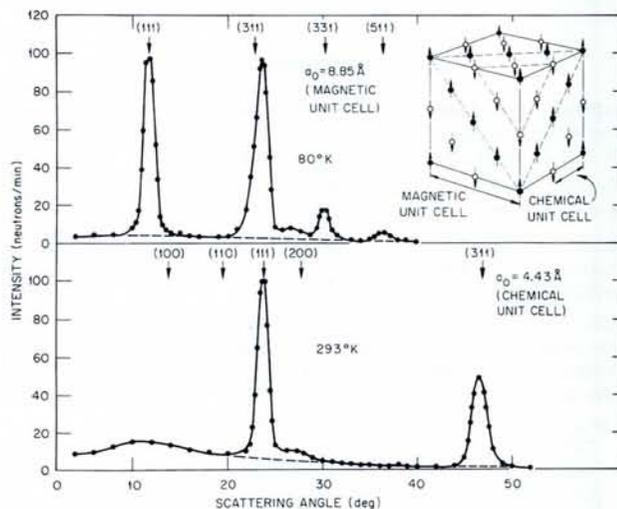
These experiments were performed at the graphite reactor at Oak Ridge National Laboratory. Simultaneously with their research on magnetism, Cliff Shull and his associates performed experiments to locate the hydrogen atoms in a series of simple crystal structures. The most common method for determining crystal structure is that of X-ray scattering (for which von Laue and the Braggs, father and son, became Nobel laureates in 1915). But X-rays are scattered by the charge (*i.e.*, the number of electrons), and for hydrogen there is only a single electron so that the contribution to the scattering of X-rays from hydrogen is very small and essentially negligible. In contrast, it was already known, from among other things the experiments of Fermi (Nobel Prize, 1938), that the interaction between a thermal neutron and a proton is relatively large. This led to the first determination of the position of hydrogen atoms in a solid by Shull and his associates at Oak Ridge in the late-1940s.

Shull left Oak Ridge in 1955 and went to MIT. He continued to perform beautiful neutron experiments at Oak Ridge, Brookhaven National Laboratory and MIT. He further developed the idea of the magnetic interaction between the neutron and the unpaired electrons in solids, and this led to the use of polarized neutrons to determine the precise distribution of unpaired electrons around the atoms [see *EN 22* (1991) 87]. Later his interests turned to the dynamical diffraction of neutrons and the building of

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*Fig. 1 — Neutron diffraction patterns from polycrystalline MnO at temperatures above and below the Néel temperature of 122 K. Neutron diffraction was pioneered by Clifford Shull, photographed below with E.O. Wollan (on the left) in the late-1940s behind the first two-axis diffractometer built specifically for neutron diffraction work which was installed at the Oak Ridge National Laboratory (ORNL) in the US.*



*C.G. Shull became Professor Emeritus of Physics at the Massachusetts Institute of Technology upon his retirement in 1985, having been appointed Full Professor in 1955. He was born in Pittsburg, PA, in 1915 and studied there, graduating in physics from the Carnegie Institute of Technology (now Carnegie Mellon University) in 1937 after starting out in aeronautical engineering. He received his Ph.D. from New York University in 1941 and worked as a research physicist at the Texas Co. (now Texaco) in Beacon, NY until 1946 and at the ORNL until he moved to MIT in 1955. He was awarded the American Physical Society's Buckley Prize in 1965, and was elected to the American Academy of Arts and Sciences in 1956 and to the National Academy of Sciences in 1975.*



neutron interferometers that are able to measure the important interactions of matter with neutron standing waves. Many of these experiments have implications in particle physics, as does his first experiment to determine the electric dipole moment of the neutron.

## How Atoms Move

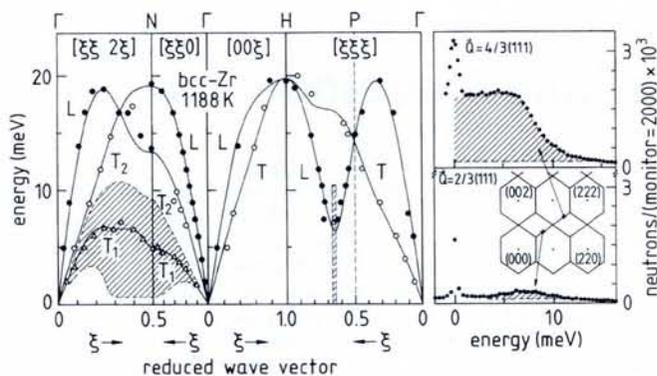
While Cliff Shull was developing the neutron scattering technique to determine where atoms and unpaired electrons "are", Bert Brockhouse at the Chalk River National Laboratory in Canada started to ask the question of how atoms "move". The theories of the dynamical motion of atoms in solids had been developed by Born, von Karmen, and others, and ultrasonic measurements were able to provide data on the long-wave-

length phonons in solids that determine elastic properties. However, no technique was available to determine the relationship between the shorter wavelength motions of atoms (phonons) and their energy. It is these quantized atomic motions that arise from the forces between atoms in solids, and are responsible for materials undergoing phase transformations and, finally, melting.

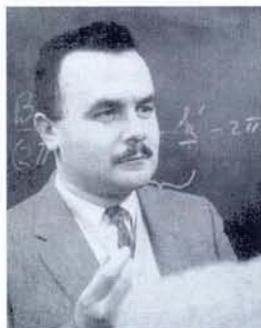
The key to Brockhouse's success was to recognize that not only do the neutrons have a wavelength comparable to the spacing between atoms in a solid (a fact, of course, used by Shull in his experiments on the static properties of solids) but also that their energy is comparable to that of the elementary excitations in solids, *e.g.*, phonons. This led to the invention of the triple-axis spectrometer in neutron spectroscopy.

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Fig. 2 — Phonon dispersion curves of Zr in its high-temperature bcc phase. The unusual feature is that some phonons have low energy and extremely short lifetimes. This is indicative of displacive transformations of bcc-Zr to close-packed structures. [From W. Petry, *Phase Trans.* **31** (1991) 119]. Brockhouse in his pioneering work showed that phonons in different Brillouin zones coincide, but this rule is clearly broken in a strongly anharmonic, vibrating crystal such as bcc-Zr.



B.N. Brockhouse (photographed) was appointed Professor of Physics in 1962 at the University of Toronto, where he remained until his retirement in 1984. Born in 1918 in Alberta, Canada, he



moved to Vancouver at an early age and worked as a laboratory assistant and self-employed radio repairman, both in Vancouver and Chicago, after graduating from high school. Following war service, he studied mathematics and physics at the University of British Columbia before moving to the University of Toronto where he obtained his Ph.D. in 1950 for work on ferromagnetic materials. He then worked for 12 years at the Chalk River National Laboratories of the Atomic Energy of Canada, Ltd., becoming Head of the newly formed Neutron Physics Branch in 1960. Member of The Royal Society of London and of Canada, he has been awarded the American Physical Society's Buckley Prize, the Institute of Physics and Physical Society's (UK) Duddell Medal and Prize, and the Canadian Association of Physicists' Gold Medal.

The first report, together with A.T. Stewart, of phonon dispersion curves was on aluminium and was published in 1955. A series of pioneering experiments performed in the late-1950s at Chalk River illustrated the power of the triple-axis instrument and the unique information that could be obtained on the dynamical properties of solids with neutron spectroscopy. An example of a more recent investigation of the phonon curves in Zr, with its incipient phonon anomaly owing to displacive phase transformations, is shown in Fig. 2. The data illustrate the advance the field has made because the dispersions are difficult to measure given that the body-centred cubic (bcc) Zr single crystal only exists at high temperatures.

The equivalent propagation of the excitations of the magnetic ground state are called spin waves or magnons, and these again were first reported by Chalk River. Brockhouse and his collaborators went on to examine a variety of solids, but also extended their work to liquids. In the early 1950s the late L. Van Hove formulated the idea of the correlation function to describe the average configuration of atoms in a liquid and how it would change with time. Neu-

trons were able to probe directly this correlation function, and Brockhouse and his team reported the first results on lead and light ( $H_2O$ ) and heavy ( $D_2O$ ) water.

### A Lasting Impact

By a delightful coincidence the news of the Prize came at the moment 400 from among the world's approximately 3000 neutron scatters were gathered for the triennial conference on the subject in Sendai, Japan. Many of us, of course, asked why the award was so late. Indeed, it is no secret that the Laureates had been nominated before, and they have both received many other awards. The Nobel Committee would no doubt answer that one needs not only

pioneering work, but also an indication of its long-term impact.

Neutron scattering is no longer performed in a dusty setting next to a nuclear reactor being used for testing materials needed in the nuclear power business. There is considerable activity in modern settings (see insert) which is a direct consequence of the unique role that neutrons play in our efforts to understand condensed matter, a role defined to a large extent by the Laureates.

In an age when relevance is the touchstone of success, it was fitting that the opening plenary talk at the recent Sendai conference was given by Sunil Sinha (Exxon, USA) on "Neutrons in Industry". Examples in his talk were drawn from the petroleum, chemical, polymer, and electronic industries. Later there was a special session on the determination of residual stress in mechanical components. Tom Holden (Chalk River, Canada) explained that the stress-corrosion cracking of steam turbine tubes is a 3000 M\$US problem in the US, and that neutrons are making a contribution towards at least ameliorating it. Neutrons have played an important role in polymer science (the difference between the scattering from H and D, as demonstrated by Shull and his associates, being the key).

These studies are not only aimed at developing new polymers, but they have also been used to demonstrate the validity of many of the theories of de Gennes (Nobel Prize, 1991). As our world becomes more complex, the clarity of understanding brought by neutron experiments stands out as more and more significant — no doubt a point recognized by the Nobel Committee in awarding this year's Prize.

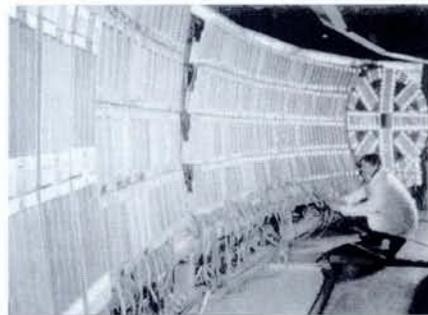
In this age of "selling", when there is often little difference between a scientist and the proverbial used-car salesman, the personalities of this year's Laureates merit attention. Both go out of their way to avoid publicity: they enjoy the company of friends, but not vast impersonal conferences. They led by example and their scientific and personal lives are perhaps best characterized by the simple word "honesty", making the award all the more satisfying.

### ACKNOWLEDGEMENT

The photographs of Figs. 1 and 2 and the diffraction pattern of Fig. 1 are taken from *Physica B* **136 & 137** (North-Holland; 1986) comprising the *Proceedings of the International Conference on Neutron Scattering* (Santa Fe, NM; 1985), and includes appreciations of the 1994 Nobel Laureates, and from the *Proceedings of the Conference on Frontiers of Neutron Scattering* (Cambridge, MA; 1985), which honoured C.G. Shull on the occasion of his 70th birthday.

### The Modern Era

A large neutron detector operated in conjunction with the High Flux Reactor at the Institut Laue-Langevin (ILL), Grenoble, the world's premier facility for neutron scattering. The ILL is operated by France, Germany and the UK, with other European countries as partners. Commissioned in the early 1970s, the reactor is ready to restart operation after a major refurbishment [EN 25 (1994) 155]. With at least 2000 scientific visits by external users each year, and some 40 working instruments, this is a far cry from the early days of neutron scattering.



Plans for a new reactor at Oak Ridge are awaiting approval from the US Congress (the cost is projected as over 2000 M\$US). At the UK's Rutherford Appleton Laboratory, an accelerator-based spallation source (ISIS) for neutron scattering now has approaching 1000 scientific visits a year. It builds on an alternative technology to produce neutrons that goes back to Cockcroft and Lawrence, and early machines at Argonne (US), Tohoku (Japan), and Harwell (UK). The next project on the drawing board for Europe is the European Spallation Source, a machine some 30 times more powerful than ISIS [EN 25 (1994) 37]. Japan also has plans for a major new neutron source.