

EARLY DEVELOPMENT OF NEUTRON SCATTERING

Nobel Lecture, December 8, 1994

by

CLIFFORD G. SHULL.

Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

Neutrons were discovered by Chadwick in 1932 when he observed a penetrating form of radiation emanating from beryllium metal when activated by alpha-particles from a radium source. Further study showed this to be neutral particle radiation which could be degraded in kinetic energy to thermal energy upon successive inelastic scattering by light atoms in a medium. With this Fermi thermalization, it was realized that thermal neutrons, because of the wave-particle duality principle, should exhibit a wave character with DeBroglie wavelength comparable to the atom spacing in solids. Thus diffraction effects should be expected in the scattering of neutrons by crystals, just as with x-radiation, and early experiments in 1936 showed in a crude way that this was true.

At this time, x-ray diffraction had been developed to become an important tool in establishing the structure and atomic interactions in materials and it seemed unlikely that neutron diffraction would develop as a useful tool because of very low source intensity. This limitation changed dramatically in the early war years of 1939 - 1943 with the discovery of nuclear fission by Hahn and Meitner and the subsequent demonstration by Fermi of a self-sustaining, neutron chain-reacting assembly. Following this Fermi demonstration, immediate design and construction of a full scale nuclear reactor, or "pile" as it was called then, was effected at Oak Ridge, Tennessee.

This reactor, then called the Clinton Pile, was meant to produce the first measurable quantities of plutonium and to serve as a pilot plant for the much larger production reactors which were being designed for construction in the state of Washington. The Clinton Pile was a graphite moderated, air-cooled assembly which operated at a power level of about 3.5 megawatts, thereby producing a slow neutron flux density of about 10^{12} neutrons/cm² sec. During this same period, another assembly was designed and constructed at Chicago with the feature of heavy water moderation, the CP-3 assembly. I show in Fig. (1) the start-up dates of these early and post-war reactors. Both the Clinton and the CP-3 pile assemblies operated through the war years producing man-made elements and isotopes and to a limited extent their neutron radiation was used to obtain some critically needed cross section data.

Fig. (2) shows an early photograph of the loading face of the Clinton Pile

with operators performing some function on a movable platform in front of the assembly. The pile is cubic in form with edge length about 7 m and consists of a nested collection of graphite "stringers", rectangular blocks of square cross sections about 10 cm on a side and length 150 cm. Pure uranium metal serves as the fissionable fuel for the assembly and is positioned in a lattice-array of channels in the graphite extending from the loading face to the back, discharge face. The uranium metal is in the form of cylindrical rods, about 2 cm in diameter and 10 cm in length, and each is carefully sealed in an aluminum can.

Start-up Dates of Early "Piles"

1942 (<i>Dec.</i>)	Fermi Demonstration (<i>Chicago</i>)
1943 (<i>Nov.</i>)	Clinton Pile, graphite, (3 MW)
1944	Argonne CP-3, heavy water
1947	NRX – Chalk River
1948	BEPO – Harwell
1950	LITR – Oak Ridge (enriched U)
	BGRR – Brookhaven

Fig. 1: Dates of Operation of Early Nuclear Reactors.

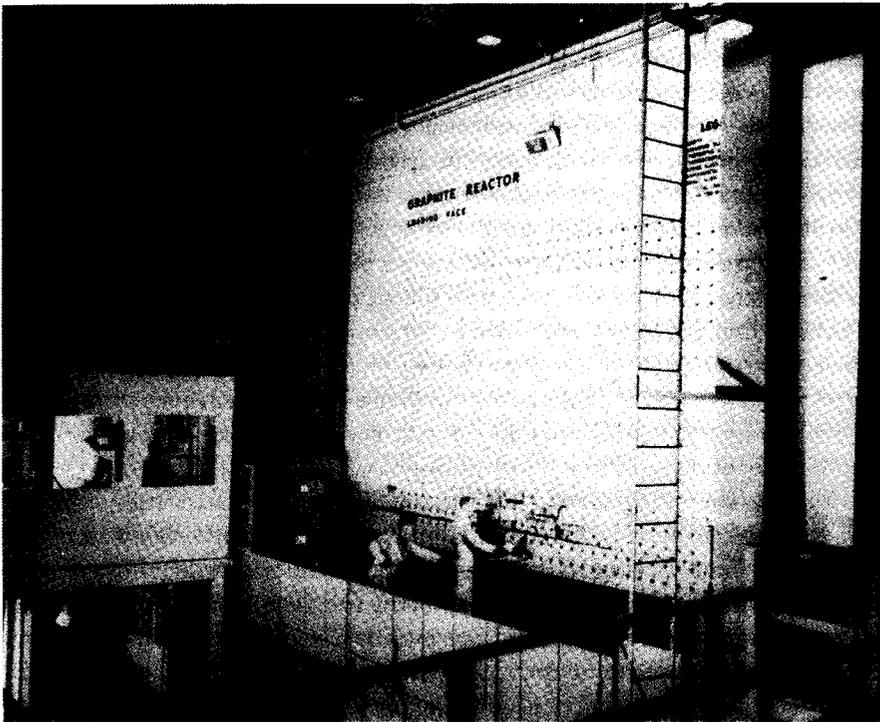


Fig. 2: Photograph of the Loading Face of the Clinton Pile.

Cooling air is drawn through the channels to the back face by large suction fans and control of the nuclear reaction level is effected by the positioning of various neutron-absorbing control rods within the assembly. Surrounding the cubic graphite pile on all sides is a radiation protection shield of concrete of thickness about 1.5 m. With foresight, the designers had arranged some channels through the concrete shield for access to the active volume of the assembly.

During the war period, a single-axis spectrometer had been used at one of these channels to supply monoenergetic neutrons for transmission cross section measurements. At the end of the war Ernest Wollan decided to set up a two-axis spectrometer for full diffraction pattern study with the first axis providing a monochromatic beam, hopefully of enough intensity to permit second axis diffraction patterns to be studied. Wollan had arranged for delivery to Oak Ridge of a base spectrometer that he had used in his earlier thesis work on x-ray gas scattering under Arthur Compton at Chicago. This spec-

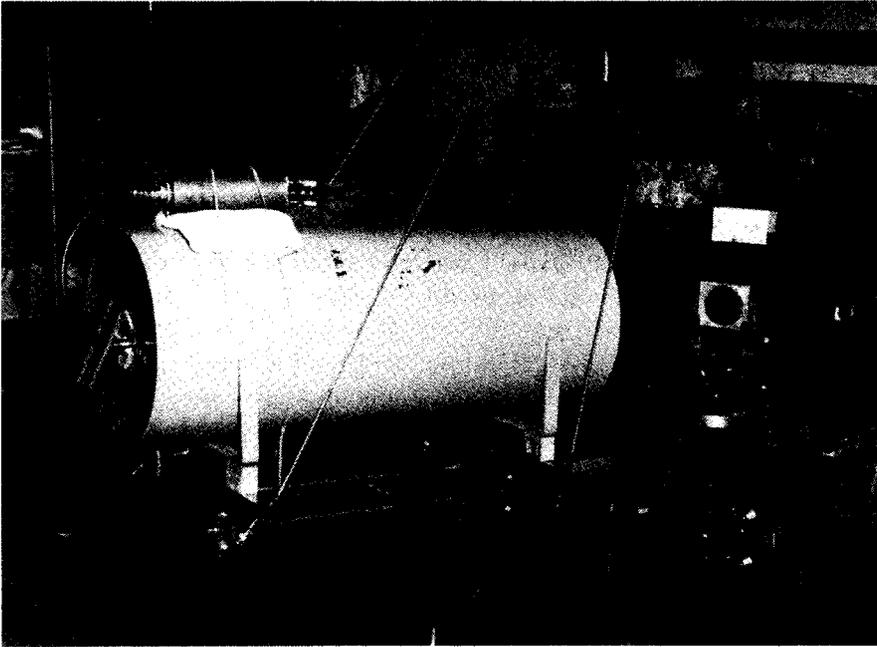


Fig. 3: Photographs of the First Double Axis Neutron Spectrometer Used to Take Powder- Diffraction Patterns at the Clinton Pile.

trometer, with coaxial control of specimen and detector positions, had to be modified to support the larger loading of a neutron detector with its necessary shielding, and this was done with support cables running to a swivel-bearing in the ceiling directly above the spectrometer axis. Fig. (3) is a photograph of this assembly taken during the first year of use but after some improvements in the shielding around the detector and the monochromating crystal had been made.

Operation of the spectrometer was entirely by hand and it was a time-consuming chore for Wollan and his early colleague, R. B. Sawyer, to collect what were the first neutron diffraction patterns of polycrystalline NaCl and light and heavy water in the early months of 1946. I was shown these patterns on a visit that I made to Oak Ridge in the spring of that year prior to my relocating there and joining Wollan in further work in June 1946. I show in Fig. (4) a portion of the NaCl pattern and, although the background level is high, it is seen that the diffraction peaks can be measured and interpreted in a quantitative way. This was important because Wollan had hoped by using powder-diffraction patterns to avoid the uncertainties in single-crystal intensities caused by extinction effects. The light and heavy water patterns were taken to see if there was any suggestion of hydrogen (deuterium) atom contribution to the diffuse, liquid patterns as taken with neutrons. Some differences in the coherent features of the two patterns were to be seen but no attempt to analyze this was made.

During the early period of study with this spectrometer the emphasis was centered on understanding the intensity that was being measured in the diffraction pattern, both within the coherent diffraction peaks and in the diffuse scattering. At that time, the only pertinent, quantitative information that was available were values of the total scattering and absorption cross sections as measured with thermal neutrons for many elements. Although these quantities might be expected to establish the total intensity in all of the diffraction pattern, the existence of isotopes for some elements, the possible presence of different spin state scattering amplitudes for nuclei with spin, and the presumed thermal disorder or excitation scattering were all factors that had to be considered case by case. Progress in isolating and defining these various factors came very slowly because of the choresome mode of data collection on the instrument, which was entirely by hand operation.

Towards the end of the first year, we were joined by a young technician, Milton Marney, who assisted in the tedious data collection and by two volunteer physicist-scientists from the Oak Ridge Training School, William Davidson and George Morton. The Training School was organized in late 1946 and was meant to bring a blue-ribbon group of established scientists from academia, industry and government to Oak Ridge for an extended period so as to attend lectures, collaborate with research groups and generally to learn the new technology and science that had been developed during the war period. Davidson and Morton took interest in our neutron scattering work and helped with the data collection: however, Morton, who was skilled in instrumentation, soon tired of this and decided to design an automatic control system that would permit step-scanning with intensity measurement and its recording. This took the form of motor drives, cam wheels and microswitches, and most importantly a print-out recorder of the neutron counter measurement. These changes took several months to effect and, once operational, it permitted continuous, uninterrupted data collection and this represented a very important step in the progress of the program.

Along with these improvements in the instrumentation, regular changes were being made in the shielding around the monochromating crystal and the movable detector on the outer spectrometer. Initially, the background intensity in the diffraction patterns was comparable to the peak intensity, as seen in Fig. (4) and about as much time was spent in measuring background as in studying the pattern itself. Regular background intensity measurements through insertion of a solenoid-activated cadmium filter into the incident beam were a part of the initial automatic program system.

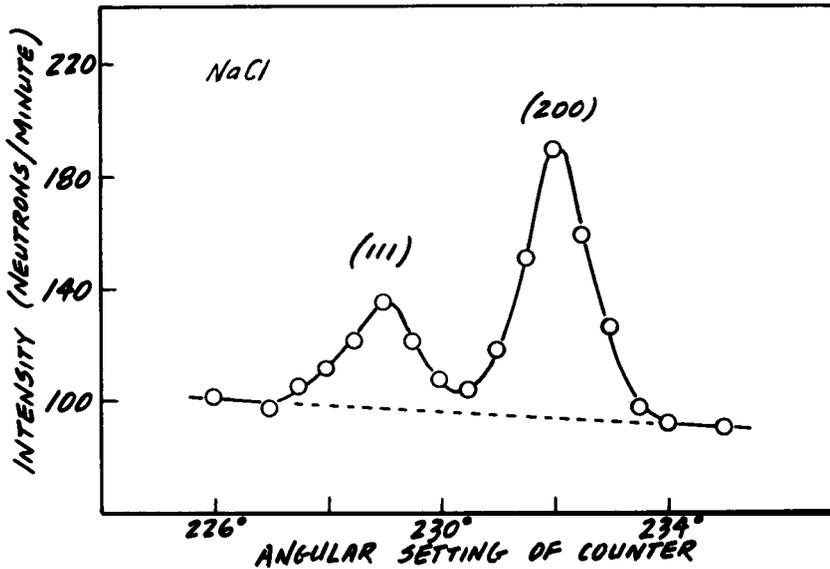


Fig. 4: Portion of the First Powder Diffraction of NaCl taken at the Clinton Pile. I

With this new system of data accumulation, we returned to our initial program of classifying various elements according to their coherent scattering amplitudes and of exploring various novel cases where neutron diffraction might supply useful information beyond that obtained with x-ray scattering. In the special case of a monoisotopic element with zero nuclear spin (nearly satisfied in oxygen and carbon), the coherent scattering amplitude, which defines the coherent peak intensity in the diffraction pattern, can be obtained from the total scattering cross section. This can then be used as an internal standard in quantifying the diffractive properties of other elements when combined with the standard element. This program of classification went on for several years continuing into the period when separated isotopes became available.

Rather early in the program, a puzzling feature of the intensity distribution was noted, namely that there was larger than expected diffuse scattering in the pattern. Careful intensity measurements on an absolute scale showed the diffuse intensity to be larger than that caused by thermal diffuse scattering or isotopic and nuclear spin incoherence. This was eventually identified

as resulting from multiple scattering within the scattering samples, a novel effect characteristic of neutron scattering and never encountered in equivalent x-ray scattering technology,

With these improvements in instrumentation and a better understanding of the components that were being seen in the patterns, attention was then turned to the important crystallographic case of hydrogen-containing crystals. It was known at the time that the coherent scattering amplitude of hydrogen for neutrons must be small since that quantity could be derived from measurements of total scattering of neutrons by ortho- and parahydrogen. However the precision of those measurements did not specify either its finiteness or its algebraic sign. There was also the additional suspicion that hydrogen atoms might display anomalously large thermal motion in crystals and that this would wipe out coherent scattering. This all became settled when powder patterns of NaH and NaD were obtained, with parts being shown in Fig. (5). Analysis of the intensities in these full patterns yielded the coherent scattering amplitudes for hydrogen and deuterium (they are of opposite sign) and showed the hydrogen centers to possess rather normal

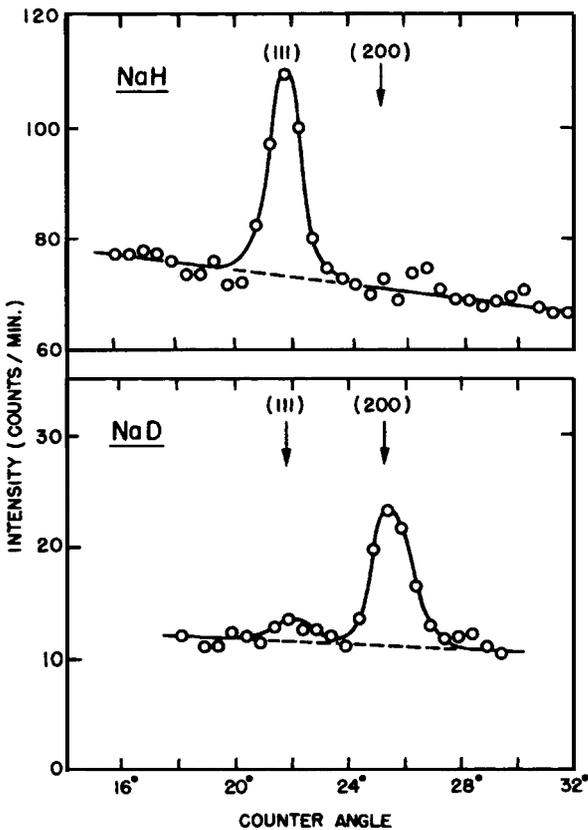


Fig. 5: Patterns for NaH and NaD showing that hydrogen contributes normally to the pattern.

thermal oscillation. The large diffuse scattering seen for NaH arises from anomalously large nuclear spin incoherent scattering from hydrogen centers.

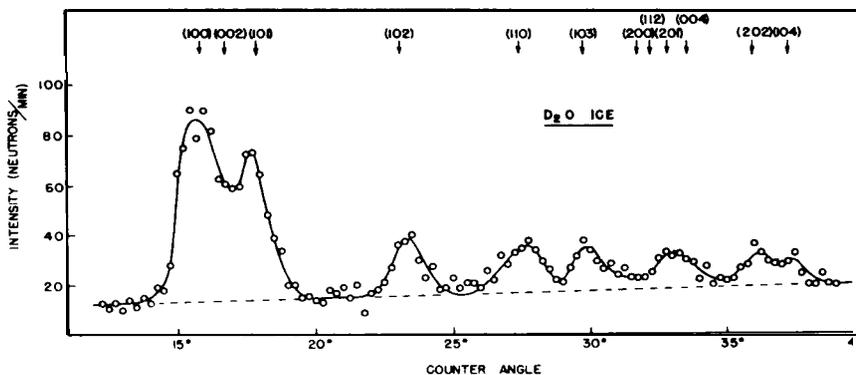


Fig. 6: Pattern Taken for a Sample of Powdered D₂O Ice.

Another interesting hydrogen structure that attracted our attention was that of common ice. It was known from x-ray diffraction studies that the basic structure was formed by interlocking oxygen atom tetrahedra but the bonding location of the hydrogen atoms was unknown and several models for this had been proposed. Fig. (6) shows a powder pattern that was taken with a deuterated sample of ice and the analysis of it shows clearly the validity of the

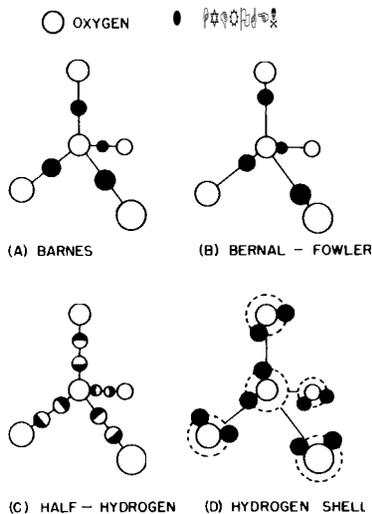


Fig. 7: Suggested Models for the Hydrogen Atom Distribution in Ice.

“half-hydrogen or Pauling” model of the structure as illustrated in Fig. (7) Preparing a powder sample of D₂O ice presented new problems and a new specimen cryostat (with dry ice-acetone coolant) was built which some peo-

ple called an "ice-ball machine". D_2O vapor was frozen on the inside surface of a cylindrical specimen tube, followed by reaming and light packing of successive layers without contamination from normal atmospheric moisture. Other hydrogen structures were studied in the next two years, mostly transition metal and heavy atom hydrides including the interesting, extreme case of uranium hydride.

In mid-1949 a second spectrometer was installed at a beam port adjoining the early one. This spectrometer, again a two-axis unit, was really the first prototype of present-day neutron spectrometers, having components that were built for specific neutron use rather than being improvised from x-ray scattering units. An elegant cylindrical shield (A) surrounded the monochromating crystal and this was supported on a large bearing which permitted easy change of beam exiting angle or beam wavelength. The specimen spectrometer (B) was supported from the monochromator shield and was now sturdy enough to support the counter arm without the use of support cables that characterized the earlier unit.

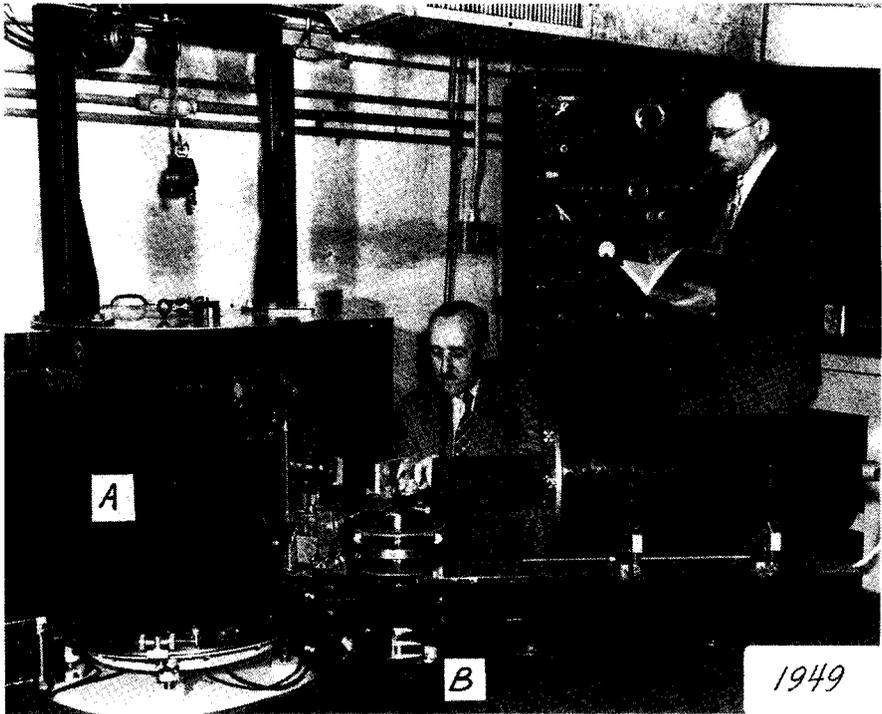


Fig. 8: Shows this spectrometer with Ernest Wollan (kneeling) and the author in an early photograph.

Along with the use of these spectrometers in which neutron-sensitive counting tubes were used to detect diffraction intensity effects, we were interested in exploring photographic detection as used effectively in x-ray technology. It was realized that photographic film would be very insensitive to neutron radiation and that a converter screen placed next to the emulsion would be required. After some tests, it was found that an indium sheet would serve this

purpose: indium has sizable absorption cross sections for thermal neutrons and emits beta-particles in its decay which will produce photographic action. This screen-film detector was used to record the first neutron Laue pattern, taken for a NaCl crystal with an exposure time of about 10 hours as shown in Fig. (9). A direct, full spectrum beam was taken from the reactor through a long shielding collimator and this passed on to the crystal with the film positioned about 10 cm behind the crystal. Among the features to be seen in this first pattern is the apparent double structure of the Laue spots. This puzzled us at first, until later investigation showed it to be an artifact caused by twinning in our first selected crystal. One can notice also in the background, a series of narrow band images running from top to bottom. This results from the presence of Scotch Tape ribbon holding adjacent strips of indium sheet together. So not only is this the first neutron Laue pattern but, as well, it is the first neutron radiograph!



Fig. 9: Neutron Laue Pattern Taken with NaCl Crystal

My abbreviated description of the developments that occurred during the first few years of neutron scattering investigation at Oak Ridge has failed to mention that many other colleagues and collaborators beyond those specifi-

cally mentioned were involved in the effort. My heartfelt appreciation is due them as well as to other members of the Physics Division and the Oak Ridge laboratory administration for unfailing support. But mostly I appreciate the association, collaboration and close friendship, extending for nearly a decade, that I enjoyed with Ernest Wollan, who first guided me to the wonders of neutron scattering research. I regret that he did not live long enough to share in the honors that have come to me.