NEUTRON SPECTROSCOPY: HOW IT ALL BEGAN

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Abstract

Abstract: The early history of neutron spectroscopy is briefly presented.

1932-1945

Most of James Chadwick's scientific contemporaries saw his discovery of the neutron in 1932 as the end of a long and exceedingly difficult search for the identity of the basic building blocks of the atomic nucleus. But to Enrico Fermi and his collaborators at the University of Rome - newly involved in a study of artificial radioactivity - it was both an end and a beginning: an end in that the elusive neutral particle had at last been found and a beginning in that the neutron appeared to offer expanded possibilities for the production of radioactive nuclei. Before the year was out the University of Rome Group was deeply immersed in a systematic study of neutron-induced radioactivity.

Some two years later, in 1934, while attempting - unsuccessfully - to reproduce an earlier-reported neutron activation investigation, the Rome Group found, to their considerable astonishment, that the level of induced activity seemed to depend on where in their laboratory the measurement was made. Searching for the origin of this curious effect, they began interposing various materials into the space between the target and the Ra-Be source that provided the neutrons. One material they tried, paraffin wax, produced a dramatic increase in the level of activation. Fermi, a theorist who had only recently turned his hand to experimental nuclear physics, was initially puzzled by this unexpected result. But it led him to what he afterwards referred to as "the most important discovery I ever made". After pondering the matter he came to the conclusion that the Ra-Be neutrons were losing energy by colliding with protons in the paraffin and that this "moderating effect" was what had increased the capture probability.

The following year, at Columbia University, Dunning, Pegram, Fink, Mitchell and Segre\(^{(1)}\), using the device shown in Figure 1, demonstrated that paraffin wax was indeed an effective neutron moderator, exactly as Fermi had surmised. As the forerunner of a long line of spectrometers operating on the time-of-flight principle, the device they built to put Fermi’s idea to the test is worth looking at in detail. Basically, it consisted of a pair of identical aluminum disks mounted 54 cm apart on a common shaft, each with alternating cadmium-covered and open sectors. As the speed of rotation was increased, the downstream disk absorbed neutrons with progressively greater velocities. Dunning et al found that the greatest drop in intensity occurred at rotation speeds of 2900 to 3000 rpm from which they inferred that the most probable velocities in the moderated part of the incident neutron spectrum were in the range of 2250 to 2700 m/sec.
The discovery that neutrons could be slowed down to energies in the thermal range had profound ramifications. One was that it inspired Leo Szilard to consider the possibility of a self-sustaining, neutron-producing chain reaction. Another was that it led to speculation that thermal neutrons might some day be useful spectroscopically since they were known to be not only charge-neutral but also to have energies and de Broglie wavelengths comparable to the collective excitation energies and interatomic spacings in condensed matter. Lacking at the time, of course, were sources that could provide thermal beams of adequate intensity. Nonetheless, soon after Fermi’s epic discovery attempts were made to establish that neutrons did, indeed, have a spectroscopic potential, remote though it then appeared to be.

In 1936 Mitchell and Powers (2), following up on the earlier neutron studies at Columbia, built the device shown in Figure 2 to explore the possibility that thermal neutrons, like x-rays, could be diffracted by ordered crystalline materials. To compensate for the extremely low thermal beam intensities produced by their paraffin-moderated Ra-Be source, Mitchell and Powers mounted an array of MgO single crystals around a central, cylindrical shield designed to screen the detector from direct, line-of-sight source neutrons. When the crystals were oriented to Bragg reflect neutrons with wavelengths in the neighborhood of 1.6 Å (2475 m/sec) - which had earlier been observed to be the most probable wavelength in the thermal part of the spectrum - count rates were found to be six to eight times higher. It thus became evident that coherent reflection could, at least in principle, be employed both as a monochromating method and for atomic-scale structural determinations.
Fig. 2 Schematic of the apparatus built by D.P. Mitchell and P.N. Powers in 1936 for their investigation of neutron scattering in ferromagnets. By this time investigations of the hyperfine splitting of spectral lines had established that neutrons (like protons and electrons) had an intrinsic spin and (like their charged counterparts) must therefore possess a magnetic moment. In 1939 Louis Alvarez and Felix Bloch - utilizing the more intense accelerator-produced neutron beams then available - were able to demonstrate that neutron scattering in ferromagnets was a spin-dependent process and that partially polarized beams could be produced by simply passing them through blocks of magnetized iron. Once a way had been found to polarize neutrons, it was then possible to employ the magnetic resonance methods pioneered by Isidore Rabi and his collaborators at Columbia University to measure the magnetic moment of the neutron. The spectrometer built by Alvarez and Bloch (3) in 1940 to make the original measurement is shown in Figure 3. It was the first of many neutron instruments to utilize polarized neutrons and it was also the first in which resonance techniques were employed.

While these initial evaluations of the properties and possible uses of thermal neutrons were being made in the United States, Fermi and his collaborators were pressing ahead with their exploration of neutron-induced radioactivity. Progressing slowly through the periodic table, they ultimately began to concentrate their attention on neutron capture in uranium, the heaviest known element, in the belief it would lead them to even heavier elements (transuranics) not present in nature. They were soon joined in the search for transuranics by Irene Curie and her collaborators in Paris and by Otto Hahn, Lise Meitner and Fritz Strassmann in Berlin. Initially, the efforts of all three groups were fruitless: no evidence of radioactive atoms with the expected chemical properties could be found. Finally, in 1939, Hahn and Strassmann made an exhaustive chemical analysis of the products of thermal neutron capture by uranium and established that intermediate-mass radioactive nuclei, not transuranics, were being produced. They were thus led to the startling conclusion that neutron capture in uranium led to a totally new process, neutron-induced-fission. Soon thereafter it was determined that both neutrons and intermediate-mass nuclei were produced in the fission process. Hahn and Strassmann’s astonishing discovery had thus moved Szilard’s idea of a self-sustaining neutron-generating chain reaction an important step closer to reality.
VII. 3. Neutron magnetic moment

The earliest resonance measurement of the neutron magnetic moment was that of Alvarez and Bloch (ALV 40). A schematic diagram of their apparatus is shown in Fig. VII. 3. Their neutrons were produced by deuteron bombardment of Be; they were polarized and analysed by passing them through magnetized pieces of Swedish iron about 4 cm thick. With these pieces of iron the neutron transmission increased about 6 per cent. on application of the magnetic field. A Rabi type single-oscillating field was used to induce the transitions. A typical neutron resonance curve is shown in Fig. VII. 3. The magnetic field was calibrated in terms of the magnetic field and frequency inside a cyclotron tuned to accelerate protons. The result of this experiment was \( \mu_n = 1.93 \pm 0.02 \) nuclear magnetons.

L.W. Alvarez and F. Bloch,
Phys. Rev. 57, 111 (1940)

Fig. 3 Schematic of the apparatus built by Alvarez and Bloch in measurement of the magnetic moment of the neutron.

These crucial experiments were carried out in a political atmosphere that ultimately became so hostile that it forced many of Europe’s most prominent scientists, among them Fermi and Szilard, to relocate in the United States. By September of 1939, when World War II engulfed Europe, the European emigre scientists and their American counterparts had moved the United States to the forefront in nuclear research. At some time between September, 1939 and December, 1941 (when the United States entered the War), Szilard, who had settled in New York City, succeeded in his quest to persuade an initially skeptical Fermi (then a member of the Columbia University faculty) that a neutron-generating chain reaction based on the fission process was a practical possibility.

Late in 1942, a group under Fermi’s direction completed construction of what would come to be known as the CP-1 reactor and demonstrated the first self-sustaining fission chain reaction... In the excitement of the moment it is doubtful that anyone present thought to consider that the fission process - apart from its potential to produce weapons of unparalleled destructive power - could also produce thermal neutron beams of unprecedented intensity. Be that as it may, once the
practicality of generating and controlling fission chain reactions had been demonstrated, the nuclear weapons development program escalated rapidly. Reactor construction, as part of this program, was given the highest possible priority. A little more than two years later reactors capable of producing beams of thermal neutrons were in operation at both the Argonne and Clinton Laboratories. Not surprisingly, the needs of the weapons program pre-empted the possible use of reactors as neutron sources for basic science investigations.

1945-1960

It was only after World War II came to an end in August 1945 that reactor-produced thermal neutron beams finally became available to the scientific community. Once given the opportunity, the select few at the Argonne and Clinton Laboratories who had access to these beams immediately set to work to determine what role (if any) neutrons might be able to play in exploring the atomic-scale properties of crystalline solids and other forms of condensed matter. Characteristically, Fermi led the way in this new endeavor.

From the beginning it was recognized that improvements in reactor design as well as a realistic evaluation of possible spectroscopic applications required, first and foremost, a more complete understanding of the neutron scattering and absorptive properties of nuclei. Thus in the immediate post-war years the primary focus was on two kinds of instruments: those intended to measure scattering and absorption cross sections and those designed to determine - for spectroscopic purposes - the signs and magnitudes of scattering amplitudes.

As would be expected, the first reactor-based neutron instruments employed the same operating principles as their earlier, pre-war counterparts. They were of two basic types, those utilizing diffraction from single crystals and those employing time-of-flight to define neutron energies. In the “single crystal” category were instruments developed in early 1946 by Lyle Borst and his coworkers (6) at the Clinton Laboratory (now known as the Oak Ridge National Laboratory) and by Walter Zinn (7) - probably in collaboration with Fermi - at the Argonne Laboratory. A schematic of Zinn's spectrometer is shown in Figure 4. Consisting of a pair of cadmium-slits to define the incident polychromatic beam, a monochromating (calcite) single crystal and a shielded, BF3 neutron detector, it would be classified today as a “single-axis” spectrometer. Energy scanning required a combined (1:2) rotation of the crystal and the arm supporting the detector. Cross sections were determined by measuring the energy dependence of transmission through a sample of the material of interest placed in the monochromatic beam between the crystal and the detector.
First experimental arrangement.

W.H. Zinn
Phys. Rev. 71, 752 (1947)

Fig. 4 Schematic of Zinn’s 1947 single-axis spectrometer.

During the war years the time-of-flight approach had been extensively used for accelerator-based neutron cross section measurements. Aware of this, Fermi and his collaborators \(^6\) decided it would be useful to adapt these well-established time-of-flight techniques to reactor-based cross section studies. Figure 5 shows the original “Fermi chopper” built at the Argonne Laboratory reactor and called (somewhat misleadingly) by its creators “a velocity selector”. Small by current standards, it had a rotor only two inches in diameter and was driven (at speeds as high as 15000 rpm) by a belt attached to the motor of a shop grinder. A stack of aluminum plates separated by cadmium foil spacers in the center of the rotor introduced the requisite pulsed structure into the transmitted polychromatic beam. Neutron energies were determined by measuring their time-of-flight from the rotor to a bank of four shielded BF\(_3\) detectors 1.46 meters away. The corresponding time dependence of the transmission through a sample placed between the rotor and the bank of detectors determined the energy dependence of the cross section.

Fig. 5 Schematic of the rotor of the original built in 1947.
Fermi and his coworkers were also the inventors of the polycrystalline neutron filter. The original test of the concept was made at the Argonne reactor early in 1946 using a 23 cm long block of crystalline graphite. Subsequently the group demonstrated that blocks of polycrystalline beryllium and beryllium oxide were also excellent filter materials.

From the viewpoint of condensed matter studies, there was at least as much need to measure scattering amplitudes as cross sections. This led to the next step forward in instrument development. Both the Argonne and Oak Ridge Groups were aware that only in the case of mono-isotopic (or nearly so), spin-zero nuclei, such as helium, carbon, oxygen, etc., was it possible to derive scattering amplitudes directly from the measured scattering cross sections. Most scattering amplitudes, it was recognized, would have to be determined either from studies of diffraction from two-element crystalline materials or from measurements of critical angles for total reflection. The construction of what we would now call two-axis diffractometers was thus undertaken as one part of this two-pronged attack on the scattering amplitude problem.

Fermi's Group at the Argonne reactor built the first two-axis instrument. Adapted from Zinn's original single-axis spectrometer, it had a CaF$_2$ monochromating crystal on the first axis and, on the second axis, a goniometer designed to support and align a single crystal sample. An arm carrying a shielded detector rotated around the second axis. Today we would describe the upgraded Zinn spectrometer as a single crystal neutron diffractometer. With it Fermi and Leona Marshall measured the intensities of low-index Bragg reflections from a number of two-element single crystals. By selecting crystals of known structure containing one element whose scattering amplitude had already been determined, they were able to derive from the observed diffracted intensities both the relative sign and magnitude of the unknown scattering amplitude.

Ernest Wollan and Clifford Shull chose to attack the scattering amplitude problem in a slightly different way. The two-axis instrument shown in Figure 6 was built for them at Oak Ridge not long after the Argonne diffractometer became operational. Of the same basic design as the Argonne instrument, the Oak Ridge diffractometer had one important advantage: it was motor-driven and had an automated data collection system and thus could operate unattended in a continuous-scanning mode. Where the Wollan and Shull approach differed from Fermi and Marshall's was that they chose to work with powder samples. Even though the diffracted intensities were lower, using powders eliminated the need to make extinction corrections to the diffracted intensities, a source of significant systematic error in the single crystal measurements. With today's perspective, we see the Wollan-Shull two-axis instrument as the first in a long line of automated neutron powder diffractometers.
Diffraction measurements determined only the relative signs of the scattering amplitudes. Casting about for a way to put the determinations of sign on an absolute basis, the Argonne Group turned to reflection techniques: the existence (absence) of finite-angle total reflection being identified, with, respectively, positive (negative) scattering amplitudes. Reflectometry also had another attractive feature: in cases where finite-angle total reflection was observed, the scattering amplitude could be directly derived from the measured critical angle. The first neutron reflection measurements were made by Fermi and Marshall \(^{(8)}\) using mirrors of selected metals mounted on the sample axis of the Argonne diffractometer. It is interesting to note that these measurements and others that followed were made solely to determine the signs and (in some cases) the magnitudes of scattering amplitudes. Almost three decades would pass before neutron reflection would be reintroduced as a microscopic-scale probe of surfaces and interfaces.

Another major step forward in neutron spectroscopy, although it wasn't apparent at the time, was P. R. O'Connor and G. T. Seaborg's \(^{(10)}\) 1948 investigation of the neutron-producing spallation reaction. More than two decades would elapse before spallation would be seen as a practical alternative to fission as a means of generating copious neutron beams for research.

Although the beams produced by the first reactors were considerably more intense than those provided by either Ra-Be sources or, for that matter, the accelerators of the day, they were still only marginally adequate for diffraction measurements. Aware that neutron diffraction was unlikely to be competitive for the kinds of structural studies that traditionally fell within the domain of x-ray crystallography, the early neutron spectroscopists chose, instead, to take advantage of the magnetic moment of the neutron and concentrate on an area that was not then accessible with x-rays and about which little was known; i.e. magnetism and magnetically ordered structures. It was, in fact, an early attempt to explore magnetic ordering in iron with neutrons that led Donald Hughes and his collaborators \(^{(11)}\) to the realization that small angle neutron scattering was a potentially valuable source of information concerning structures of intermediate scale (in this case magnetic domains). Figure 7 shows the facility they built in 1949 for small-angle studies at the Argonne reactor. Utilizing a polychromatic beam defined by cadmium slits, their instrument can be seen today as the first step towards the creation of the small-angle-neutron-scattering spectrometers that are used today to explore the microscopic-scale configurations of polymers and biological systems.
Equipment for study of small-angle scattering; the neutron intensity as a function of $\theta$ is measured by moving the counter and slit in the plane of the figure (horizontal plane).


Fig. 7 Schematic of the Hughes, Burgy, Heller and Wallace for small angle investigations.

As a unique source of information relating to magnetic structures and magnetic ordering transitions, neutron diffraction investigations soon began to attract the attention of the scientific community. This rapid rise of interest in magnetic systems was the inspiration for the next step forward in instrumentation: a polarization-analysis capability, it was recognized, would increase the sensitivity and selectivity of magnetic measurements. Thus a search began for better ways to polarize neutrons.

Up to the end of 1949, neutron instruments and instrument components were basically copies of their x-ray and accelerator physics counterparts. But in attacking the polarization problem, a purely neutron approach emerged. Both the Argonne and Oak Ridge groups decided to try exploiting a concept that derived from Otto Halpern and M.H. Johnson's theoretical analysis of neutron interactions with magnetic atoms; namely, that the nuclear and magnetic parts of the scattering were coherent and would therefore interfere. Looking through tables of the commonly available ferromagnetic materials, Morton Hamermesh at Argonne found one, cobalt, in which the magnetic scattering amplitude was larger than the nuclear amplitude. He suggested that one of the two spin states of the neutron would have a positive scattering amplitude and therefore be totally reflected at finite angles, while the other, having a negative scattering amplitude, would only totally reflect at zero angle. Capitalizing on Argonne's earlier experience with mirrors, Hughes and Merle Burgy constructed a pair of identical cobalt mirrors bonded to copper substrates, one to serve as polarizer and the other as polarization analyzer. Although they were not able to align the cobalt magnetic moments completely with the available field, Hughes and Burgy nevertheless found they could produce substantially better broad-band polarizations by reflection than could be achieved by the earlier method of transmitting the beams through magnetized iron. Theirs was the first demonstration of neutron polarization by reflection.

Characteristically, the Oak Ridge Group chose to apply the interference concept somewhat differently. Based on their familiarity with diffraction techniques, Shull, Wollan and Walter Koehler made a search for ferro or ferri-magnetic single crystals with low-index Bragg reflections in which the nuclear and magnetic parts of the scattering were so closely matched that only one of the two spin states of the neutron would scatter coherently. Using transmission through magnetically saturated iron to analyze the polarization of the Bragg-reflected beam, they were able to show that the monochromatic beam produced by the (220) Bragg reflection from a magnetically saturated single crystal of magnetite (a ferrimagnet) was essentially 100 percent
Schematic diagram of the apparatus. The angle of scattering $\phi$ can be changed by moving the specimen and the spectrometer as indicated.


Fig. 8 Schematic of Brockhouse's original triple-axis spectrometer. This striking result inspired the subsequent building of a number of crystal diffractometers with a polarization analysis capability.

Another instrument introduced during these early years was the helical-groove, slow-neutron velocity selector. By replacing the slotted-disk rotor of Dunning et al. with a rotor containing spiral grooves, J.G. Dash and H.S. Sommers, Jr. were able to produce continuous, monochromatic, slow neutron beams by mechanical means alone. Although the helical-groove velocity selector subsequently fell into disfavor because its resolution and spectral range were limited, it is now coming back into its own both as a higher-order filter and as a monochromator for special applications (such as small angle scattering) where lower energy neutrons are preferred and high intensity is more important to the over-all performance of the instrument than energy resolution.

Because Argonne and Oak Ridge were the only places where facilities for neutron spectroscopy were available, almost all neutron instrument development work up to this point was concentrated at these two laboratories. But within a few years of the end of World War II new laboratories for nuclear and condensed matter research were under construction, not only in the United States but in Canada, western Europe and the Soviet Union, and as fast as these new institutions were completed they began to build their own reactors and organize neutron scattering programs. With facilities for neutron research becoming more generally accessible, the focus of instrument development then began to shift from Argonne and Oak Ridge to the newer institutions.

In the early post-war years, reactor-produced beam intensities were barely sufficient for elastic studies and inelastic scattering investigations weren't given any serious consideration. But by the early 1950's a second generation of reactors producing more intense beams had come online and Bertram N. Brockhouse decided that the time had come to try adding a third axis (for an energy analyzing crystal) to the two-axis spectrometer. The first triple-axis spectrometer, which he built and installed at the NRX reactor at Canada's Chalk River Laboratory (16), is shown in Figure 8. With it Brockhouse made the first direct observations of propagating and diffusive modes in condensed matter systems launching thereby the new field of inelastic neutron spectroscopy.
It is interesting to note that in cases where instruments based on either crystal diffraction or time-of-flight could serve experimental needs equally well, both types tended to be developed at about the same time and were, for the most part, equally convenient and effective. Thus while plans to build a triple-axis spectrometer were moving forward at Chalk River, a variety of time-of-flight and combined crystal diffraction and time-of-flight approaches to inelastic spectroscopy were also under active consideration at other laboratories. At Harwell, for example, Peter Egelstaff was thinking in terms of building a phased, two-rotor system to produce bursts of monochromatic neutrons \(^{(17)}\). But the technical problems of operating such systems and handling the large amounts of data they produced were not then easily resolved, and it would be well into the next decade before instruments based on time-of-flight became available for inelastic scattering investigations.

With increasing emphasis on inelastic neutron spectroscopy, a demand arose for spectrally tailored beams, particularly beams of sub-thermal energy. Interestingly, attempts to down-shift the spectrum of Ra-Be neutrons with cryogenically cooling hydrogenous moderators had actually been made in pre-war days, but the results had been inconclusive \(^{(18)}\). But by the 1950’s numerous advances had been made in neutron technology and Egelstaff and his collaborators at Harwell decided to have another look at the problem. Coming ultimately to the conclusion that liquid hydrogen and deuterium offered the best prospects for success, they designed and constructed a liquid hydrogen moderator that was installed in Harwell’s BEPPO reactor \(^{(19)}\). It performed so well that another was soon made for their DIDO reactor. Building on this early experience, liquid hydrogen (deuterium) moderators were subsequently installed in a number of other research reactors.

Retrospective

Looking back, 1960 appears as a watershed year for neutron spectroscopy. At that point, most of the basic instrumental concepts were in-place and neutron-based experiments were beginning to have a considerable impact on condensed matter physics. But it was a field still plagued by many problems. Certainly the major one was that it was very much source-constrained: beam intensities were marginal even at the best reactors and collecting statistically reliable data was a time-consuming and not always successful process. And because the vacuum tube electronics of the day were prone to drift and malfunction, even in the rare instances when sufficient beam time was available, reproducing data tended to be difficult, particularly when small cross section processes were the major focus of the investigation. Beyond that, the performance of the instruments themselves left much to be desired. Those employing crystal diffraction for monochromation were handicapped by the inadequate reflectivity of the available crystals; those based on time-of-flight had problems relating to support bearings and speed and phase control systems and were, in addition, adversely affected by the limitations of their vacuum-tube-based data collection and data analysis systems.

In the decades that followed, high flux research reactors would be designed and built, stable transistorized electronics and digital computers would become commonly available, neutron guides would make their appearance, much better monochromating and polarizing crystals would be found and the development of magnetic suspension bearings and computerized instrument control, data collection and data analysis systems would bring time-of-flight-based spectrometers to full parity with their crystal-based counterparts. But even with these many technological advances, it is still easy to discern in today’s state-of-the-art instruments the pervasive influence of instruments built by the first neutron spectroscopists long ago.

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