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EARLY DEVELOPMENTS IN POLARIZED NEUTRON TECHNOLOGY

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Abstract.- Early developments in the production and use of polarized beams of slow neutrons were coupled closely with improvements in the understanding of the magnetic scattering process and with the establishment of magnetic structures by neutron scattering. These aspects will be summarized in the present review.

1. Introduction and Early Theoretical Developments

In the years immediately following Chadwick's discovery of the neutron, there was intense interest in studying the interactions of this new form of radiation with nuclei and in establishing its characteristic physical properties such as its mass, spin, and magnetic moment. Theoretical arguments were available at the time suggesting that the spin was probably 1/2 and that a magnetic moment existed which should be near that of the proton but of opposite sign relative to the spin direction. But of course this needed confirmation and this encouraged Felix Bloch to investigate [1] the interaction between the supposed magnetic neutrons and magnetic atoms in matter as a means of assessing the neutron properties. Bloch showed that this interaction would result in a scattering cross section comparable to that of other nuclear interaction cross sections and, in prophetic terms, he proposed that studies of neutron transmission through ferromagnetic iron would yield information on the neutron moment, the preparation of polarized neutrons, and on details of the atomic magnetization. All of these were to follow in due course and my present task is to outline the early developments in the magnetic scattering of neutrons that were to lead to a polarized neutron technology -- the theme of our Conference.

The seminal contribution of Bloch was to lead very shortly to a detailed quantum-mechanical treatment of magnetic scattering by Schwinger [2] with resultant basic cross section expressions somewhat different than had been developed by Bloch. This difference arose in the directional dependence of the cross section and it was found [3] to originate from different assumptions about the intrinsic magnetic structure of the neutron and whether it is to be described as a point-like dipole or as a microscopic amperian current -- a question that was not to be settled for another dozen years. Schwinger also formulated expressions for different experiments that might become tractable, recognizing again the polarization-producing features of these.

The Schwinger analysis, along with the Bloch treatment, had concentrated on the effects that might arise from the coherence between magnetic and nuclear scattering in the specific case of iron atoms in the ferromagnetic metal. It remained for Halpern and Johnson [4] to suggest that scattering by other magnetic ions in a paramagnetic state might be more favorable in terms of experimental measurement and...
moreover the theoretical interpretation of the results would offer less complication. At this time the interpretation of the magnetic state of uncoupled paramagnetic ions was much better understood than was the case for the magnetic metals. These early published notes were followed by three comprehensive theoretical papers by Halpern and Johnson [5], Halpern and Holstein [6], and Halpern, Hamermesh and Johnson [7] which set the stage for all of the later experimental work. In these remarkable papers, the authors again address the question of neutron scattering by individual magnetic atoms and as well, the consequences of coherence that would arise in the ferromagnetic and partially coupled paramagnetic states. For the first time, the specific effects arising from the crystal structure of the magnetic medium were analyzed and a comprehensive treatment was given of the polarization, and depolarization, action that would be expected in transmission experiments of neutrons through ferromagnetic media. These authors subscribed to the Schwinger point-of-view concerning the basic form of the magnetic interaction but nevertheless believed that the question should be left to observation.

2. Establishment of Magnetic Scattering and the Sign and Magnitude of the Magnetic Moment.

Experiment groups at various locations, most notably at Cornell, Columbia and Copenhagen, were not long in responding to the challenges set forth by the first Bloch paper and the next two years saw a flurry of short papers dealing with the transmission of neutrons through magnetized iron. These are discussed in two summary papers by Frisch, von Halban, and Koch [8] and by Powers [9] both appearing in 1938. It must be recognized that the only neutron source available at the time was the Ra-Be source surrounded by a hydrogen-containing paraffin howitzer for thermal moderation and low intensity was the rule-of-the-game. Some control on the average speed of the neutrons was available from the temperature of the howitzer and detection was accomplished in rather crude BF3-gas absorbing chambers. The experiments concentrated on the passage of thermal neutrons through magnetized and unmagnetized iron and, considered collectively, they demonstrated that

(i) there was a single transmission effect in which the transmitted intensity increased upon magnetization of the iron, upon increasing the thickness of the iron, and upon reducing the average neutron speed,

(ii) double transmission effects, with successive transmission through two iron plates magnetized parallel or anti-parallel, were to be found which suggested that nonadiabatic transitions of the neutron spin state could occur between the plates, and

(iii) that controlled neutron spin precession between the plates with auxiliary fields could be used to obtain the sign and magnetic moment of the neutron.

From these significant observations, it was concluded that the sign of the neutron magnetic moment was negative (moment vector opposite to spin vector) and that the moment value was about 2 nuclear Bohr magnetons (within 50%) in rough agreement with theoretical expectation. Furthermore, the experiments showed conclusively that there was magnetic scattering of neutrons as originally suggested by Bloch with consequent neutron polarization in neutron beams transmitted through magnetized ferromagnetic media and that the sense of polarization could be modified by auxiliary magnetic fields.

An important extension of the technique was introduced in 1940 by Alvarez and Bloch [10] through the use of resonance modification of the neutron polarization between two magnetized iron plates (polarizer and analyzer). Using cyclotron produced neutrons of much higher intensity than that from Ra-Be sources and calibrating the Larmor precession field in terms of the proton cyclotron field, they obtained the first accurate value for the neutron moment to be 1.93 (± 0.02) nuclear Bohr magnetons. Further progress in defining this important quantity was limited until
after the war years when Arnold and Roberts [11] reported the first precision value using essentially the same techniques as had been developed by Alvarez and Bloch. By this time, 1947, neutron beams of much higher intensity from a nuclear reactor (or pile as they were known then) were available and they were able to obtain the moment value of 1.9103 (± 0.0012) nuclear Bohr magnetons. This value was important in that it was the first to show clearly the non-additivity of the proton and neutron moments in forming the deuteron moment as had been suggested by Rarita and Schwinger [12]. Further improvement in this value was obtained in the contemporary experiment of Bloch, Nicodemus, and Staub [13] who reported the value 1.91307 (± 0.0006) along with additional evidence that the sign was negative.

3. Polarization of Neutrons Passing Through Magnetized Iron

Following the initial demonstration experiments showing magnetic scattering of neutrons, the primary interest lay in the quantitative establishment of the moment which resulted in the remarkable precision as chronicled in the above section. During the same period however, attention was also given to measurement of the cross section for magnetic scattering as is evidenced in the single transmission measurement and to the neutron polarization that would be produced by such means. Although the earliest experiments on this with Ra-Be neutron sources had shown some of the features predicted by the early theory, it remained for later experimentation with much higher intensity from cyclotron and reactor sources to obtain quantitative comparison. Thus in 1943, Bloch, Hamermesh, and Staub [14] showed the predicted dependence of the effect upon the square of the iron plate thickness and demonstrated the need for obtaining high magnetic saturation of the transmitting iron plate. However their value for the polarization cross section as interpreted from the single transmission effect (fractional change of intensity upon magnetization) turned out to be about twice as large as would be calculated using the coherent nuclear scattering amplitude and best form factor distribution for iron as known at the time. This study was extended in later papers by Fryer [15] using bands of different mean neutron energy from a pulsed cyclotron and by Bloch, Condit and Staub [16] with more highly saturated iron samples. There still remained the disquieting disagreement with the theory and this was emphasized even more in a study of Hughes, Wallace and Boltzman [17] who attacked the problem with neutrons from a nuclear reactor. Using both thermal spectrum neutrons and monoenergetic neutrons with a velocity selector, they found experiment results to be about three times larger than expected.

In considering possible reasons for this discrepancy, some attention was paid at the time to a revision of the neutron spin upward to a value 3/2 in spite of strong, independent evidence favoring 1/2. This was discarded however when new calculations by Steinberger and Wick [18] appeared in 1949. These authors recalculated the magnetic form factor for iron allowing for exchange effects and, with an improved value for the coherent nuclear scattering cross section for iron, established agreement with the Hughes values. Further experiments by Fleeman, Nicodemus, and Staub [19] resolved some of the smaller differences that had occurred in the various experiments. It was of course recognized in these experiments on the single transmission effect that the neutrons were partially polarized in the transmitted beam. In the most favorable case, this was expected to be about 60% and, in order to study this in most direct fashion, Burgy et al [20] resurrected the double transmission technique that had been used in the earliest experiments [8,9]. In this arrangement a neutron beam is passed successively through two iron plates with the first serving as the polarizer and the second as the analyzer. New complications are introduced in this technique such as depolarization within the plates, adiabatic changes of polarization between the plates, beam hardening, and grain size effects. These were studied by Burgy and colleagues and, although quantitative results on the degree of polarization were uncertain, the experiments gave general support to the expected behaviour.
4. Polarization of Neutrons by Reflection from Magnetized Mirrors

In their pioneering theoretical paper [7], Halpern, Hamermesh, and Johnson had pointed out the double-valued index of refraction that neutrons would experience in a ferromagnetic medium. This led Halpern [21], and independently Achieser and Pomeranchuck [22], to propose that there would be two critical angles (depending upon the neutron spin state) for total reflection from a magnetized iron surface and that this could be used to prepare a completely polarized neutron beam. This suggestion was given significant extension by Hamermesh [23] who pointed out that complete polarization would also be expected in the reflected beam from a magnetized cobalt surface with lesser demands on the collimation of the incident beam and its wavelength purity. In this material the scattering amplitudes are such that the two indices are on opposite sides of unity and this implies that only neutrons of one spin state will suffer external reflection.

These very interesting suggestions were shortly studied by Hughes and Burgy [24] using BeO filtered and well-collimated neutrons (λ about 4.5Å) falling upon magnetized mirror surfaces of iron and cobalt. Figure 1 reproduces their reflectivity data as a function of mirror angle for the case of iron showing the presence of two critical angles corresponding to the reflection of the two neutron spin states. The presence of two critical angles is of fundamental importance since it serves to distinguish between the original Bloch and Schwinger formulations of magnetic scattering theory as mentioned earlier. The curve on the figure labelled Bloch represents the single index of refraction effect that would be expected for the conditions of the experiment in a dipole representation of the neutron magnetic structure. On the other hand for a Dirac amperian current representation as introduced by Schwinger, one expects the double index curve agreeing nicely with the experiment points as shown. Further theoretical discussion on this topic was provided by Ekstein [25]. This fundamental conclusion was arrived at simultaneously by Shull, Wollan, and Strauser [26] in a different type of magnetic scattering experiment that will be discussed in the next section. They studied the pure magnetic scattering (uncontaminated by nuclear scattering) in a Bragg reflection from ferromagnetic Fe3O4 as a function of magnetization direction relative to the scattering vector with results shown in figure 2. In this figure, the two curves labelled sin²α and cos²α correspond, respectively, to the Dirac and Bloch representation with the data offering full support to the former model.

![Fig. 1: Reflectivity of iron mirror surface as a function of glancing angle (Hughes and Burgy [24]).](image1)

![Fig. 2: Variation of magnetic scattering intensity with angle between the magnetization and scattering vectors (Shull, Wollan, and Strauser [26]).](image2)
Hughes and Burgy also studied surface reflection from a thin layer of cobalt deposited on a copper base where complete polarization was to be expected. This was done in a double mirror reflection arrangement with the second, downstream mirror serving as polarization analyzer and with both mirrors being magnetized in the plane of the layer and parallel. Upon inserting an unmagnetized sheet of iron between the mirrors, thereby depolarizing the beam from the first mirror, the intensity ratio (polarized to unpolarized, now referred to as the shim ratio) was found to correspond to 100% polarization in the beam from the first mirror.

This demonstration of complete polarization in a mirror reflected neutron beam was important in showing that neutron beams of high intensity with full polarization could be prepared for further use. Exploitation of this technique has occurred in many experimental studies since that time and of course technological improvements have been introduced, most notably by Mezei [27] with "super-mirror" fabrication. It still remains as a favored technique with competition only from the Bragg reflection method that will be discussed in the following section.

5. *Magnetic Scattering Developments and Polarization by Bragg Diffraction*

At the same time as the demonstration of complete polarization by ferromagnetic mirror reflection, other magnetic diffraction developments had occurred leading to Bragg diffraction as a means of obtaining fully polarized neutron beams. The polarization aspects of this are so intricately linked to advancements in the magnetic scattering and diffraction of neutrons that the author wishes to include some discussion of this topic in the present outline. In doing so it is hoped that the reader will bear indulgence with the author for the rather personal vein in which it is presented.

Neutron diffraction as a complementary tool to X-ray and electron diffraction can be said to have originated in 1946 with studies by Fermi at Chicago on single crystal Bragg reflections and by Wollan and Shull at Oak Ridge on polycrystalline sample diffraction. The earliest studies concentrated on classifying the coherent nuclear scattering from various elements and isotopes and on exploring some of the areas where novel crystallographic information might become available. Included in these was the possibility that the paramagnetic scattering predicted by Halpern and Johnson [4] could be measured since this would serve as a new test of magnetic scattering theory and as well provide information on the magnetic form factor. A very early pattern of MnO taken at Oak Ridge had exhibited a larger-than-normal diffuse scattering and we were suspicious that this might be a representation of paramagnetic scattering. This was studied in greater depth in late 1948 and early 1949 and, after correction for multiple scattering, nuclear incoherent scattering and temperature diffuse scattering, the residual diffuse scattering expressed in cross section units

![Radial distribution function](image1)

![Magnetic diffuse scattering](image2)

**Fig. 3:** Magnetic diffuse scattering by Mn**++ **ions in various compounds.

**Fig. 4:** Radial distribution function for 3d electrons in Mn**++ **ions.
of figure 3 was found. The peaking in this curve for MnO was rather unusual for an atomic form factor and other Mn\textsuperscript{++}-ion containing compounds were studied which showed more normal form factor dependence upon scattering angle. The form factor suggested by these curves was then Fourier transformed to get the radial distribution function of the 3d magnetic electrons in Mn\textsuperscript{++} as shown in figure 4. This was given comparison with a theoretical calculation by Dancoff using a self-consistent field analysis with exchange effects included.

During this period the author had been in communication with Louis Maxwell and Samuel Smart at the Naval Ordinance Laboratory near Washington and they pointed out Prof. L. Néel's work on magnetic materials [28]. This suggested to the author that the short range order character of the MnO diffuse scattering at room temperature should show further development at low temperature and possibly to long range order below the antiferromagnetic ordering temperature (now Néel Temperature) of 122 K. This belief was strengthened by an early observation of extra, unidentified diffraction lines in the neutron patterns for α-Fe\textsubscript{2}O\textsubscript{3} at room temperature. Néel had considered this material to be antiferromagnetic below 680°C and it was possible that the extra structure in the pattern at room temperature was to be associated with this long range magnetic order. Encouraged by Smart to test this, both low and high temperature patterns were obtained for MnO and α-Fe\textsubscript{2}O\textsubscript{3} respectively which confirmed the initial suspicion. These patterns are shown in figures 5 and 6 as reported by Shull and Smart [29] and by Shull, Strauser, and Wollan [30].

![Fig. 5: Development of coherent antiferromagnetic scattering in MnO at low temperature.](image1)

![Fig. 6: Collapse of coherent antiferromagnetic scattering in α-Fe\textsubscript{2}O\textsubscript{3} at high temperature.](image2)

With this confirmation of both incoherent paramagnetic and coherent antiferromagnetic scattering, the Oak Ridge group studied the magnetic structures of other materials, most notably that of ferromagnetic magnetite, Fe\textsubscript{3}O\textsubscript{4}, containing both Fe\textsuperscript{++} and Fe\textsuperscript{+++} ions distributed among the tetrahedral and octahedral sites. Néel had suggested a model of this magnetic structure which he called ferrimagnetic with unbalanced antiferromagnetic coupling of tetrahedral and octahedral Fe ions as shown in figure 7. The neutron diffraction pattern shown in figure 8 gave full confirmation of this postulated magnetic structure as reported by Shull, Wollan, and Koehler [31]. Two very interesting features of this pattern were to be noticed in the (111) and (220) reflections. The former reflection was analyzed to be almost completely magnetic in origin with minimal contribution from nuclear scattering and since the material is ferromagnetic with ion orientation depending on the magnetization direction, it could be used in a sensitive test distinguishing the Bloch and Schwinger formulations of the fundamental magnetic scattering theory. This has been discussed in section 4. On the other hand, the (220) reflection was found to consist of closely similar
amplitude contributions, $0.95 \times 10^{-12}$ cm from nuclear scattering and $0.97 \times 10^{-12}$ cm from magnetic scattering. This suggested to the author that this should serve as a fully effective polarizing reflection because the cross section for scattering would be drastically different for the two spin states of an unpolarized beam.

This anticipated neutron polarization was sought as soon as a single crystal of Fe$_3$O$_4$ became available. The crystal was held in a magnetizing field and the degree of polarization was assessed by passing the (220) reflected beam through a block of iron held in a separate and parallel magnetic field. Measurement of the single transmission effect in the iron block along with a similar monitoring measurement with an unpolarized beam from a copper crystal yielded a polarization value of $100(\pm 5)\%$. With the same arrangement, a similar measurement was performed on the polarization produced in a (110) reflection beam from an Fe crystal and this yielded 41% which is rather lower than the calculated value of 60% because of extinction in the iron crystal. These measurements were reported by Shull [32].

The Bragg reflection method of preparing a polarized neutron beam has been widely used since then in many dedicated spectrometer assemblies. The merit of this method arises from having both polarization and monochromatization available in a single diffraction process. Such monochromatization is invariably required in magnetic structure analysis. Of course other Bragg reflections from other crystals can be expected to offer high polarization when the scattering amplitudes are suitably distributed and mention can be made to two cases which have found frequent use. These are the ferromagnetic metal crystal Co-8% Fe in (200) reflection and the Heusler alloy Cu$_2$MnAl in (111) reflection both offering higher intensity than does Fe$_3$O$_4$. The latter has been developed by Delapalme and Schweizer [33].

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6. References

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