LA NOUVELLE VAGUE IN POLARIZED NEUTRON SCATTERING

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Polarized neutron research, like many other subjects in neutron scattering developed in the footsteps of Cliff Shull. The classical polarized neutron technique he pioneered was generalized around 1970 to vectorial beam polarizations and this opened up the way to a "nouvelle vague" of neutron scattering experiments. In this paper I will first reexamine the old controversy on the question whether the nature of the neutron magnetic moment is that of a microscopic dipole or of an Amperian current loop. The problem is not only of historical interest, but also of relevance to modern applications. This will be followed by a review of the fundamentals on spin coherence effects in neutron beams and scattering, which are the basis of vectorial beam polarization work. As an example of practical importance, paramagnetic scattering will be discussed. The paper concludes with some examples of applications of the vector polarization techniques, such as study of ferromagnetic domains by neutron beam depolarization and Neutron Spin Echo high resolution inelastic spectroscopy. The sample results presented demonstrate the new opportunities this novel approach opened up in neutron scattering research.

1. Introduction

There are few areas in neutron scattering research in which Cliff Shull was not associated with the first, pioneering steps. Polarized neutrons are no exception. Cliff is one of the authors of a decisive experiment [1] which provided the experimental proof that the neutron spin has to be considered as a microscopic Amperian current loop and can not be modelled by a microscopic dipole, i.e. positive and negative magnetic poles separated by an infinitesimal distance. An other, also decisive experimental proof of the same rather surprising fact was published by Hughes and Burgy [2] in the same volume of Phys. Rev., 16 pages apart from that of Shull, Wollan and Strauser [1]. Actually, the two papers appeared in inverse order of submission, and their quasi-simultaneity does not diminish the merit of either of the two, in particular because they were of very different types. In the same year of 1951 Cliff published another seminal paper on polarized neutron diffraction from ferromagnetic crystals (actually ferrite and iron) [3] which became the starting point of a major chapter of neutron scattering research, the study of magnetic structures with the tremendously enhanced sensitivity polarized neutrons provide compared to unpolarized beam diffraction work.

In this kind of polarized neutron work, as introduced by Cliff, the neutron beam polarization is always parallel to the magnetic field, which has to be present all along the neutron trajectory in order to maintain the polarization. Thus the polarization is defined as a scalar quantity \( P = p_\uparrow - p_\downarrow \) where \( p_\uparrow \) and \( p_\downarrow \) stand for the occupation probabilities of the "up" and "down" spin states. This approach seemed to be quite satisfactory in view of the beautiful results it produced in magnetic crystallography, and also because it appeared to be quite natural on the basis of the happy emphasis quantum mechanics textbooks put on the very much quantic nature of 1/2 spins, i.e. that only the two states "up" and "down" appeared to be of physical relevance. It took as much as 18 years before the next fundamental step in polarized neutron techniques had been mastered by the extension of the experimental capabilities to the production and analysis of vectorial (3 dimensional, 3D) spin polarizations, which essentially means polarization vectors \( P \) not parallel to the magnetic field. The original scalar "up-down" polarization thus corresponds to the component of the vector polarization parallel to the magnetic field direction, which we define as the z-axis, i.e. \( P_z = p_\uparrow - p_\downarrow \).
This generalization of the concept of beam polarization has been independently introduced by several, actually out-of-the-main-stream research groups \([4,5,6,7]\), using quite different experimental techniques. It was probably fortunate that these groups were not aware of each other's work, thus a number of very different lines of thought were followed and applications in very different directions were pursued. Special credit has to be given to the group of G.M. Drabkin in Leningrad who were doing three-dimensional polarization analysis as early as 1969, the year in which the beautiful paper by Moon, Riste and Koehler was published on conventional “up-down” polarization analysis \([8]\). A detailed account of the system developed in Leningrad was only published several years later \([4]\).

The generalized, vectorial polarized neutron work is an area in neutron scattering whose debut did not benefit of Cliff's interest. This just might be a sign for those of us who spent more than a decade with it that our field is eventually not that fascinating after all. Nevertheless, it did produce some fundamentally rather new concepts and quite some science via its applications such as Neutron Spin Echo (NSE). The phrase “nouvelle vague” in the title was meant to refer to developments which started with and followed up the advent of vectorial polarization analysis techniques. As we will see below, it is the interference between the “up” and “down” spin states which leads to 3D spin polarizations. Thus we will fundamentally be concerned with the coherence between these two states. Coherence can even be maintained through scattering processes which allow us to shape neutron spin wave function “by hand”, as it was shown to be an aspect of the action of neutron polarizers \([9]\). Paradoxically, the full quantum-mechanical treatment of neutron spin behaviour leading to the vector polarization picture as opposed to the “popular” quantum image of “up-down” states leads to results in full agreement with predictions of a classical mechanical description \([10]\). Without any doubt, innovative thinking in this area has crucially benefited of this unexpectedly recovered liberty of reflecting in simplest classical terms. Landau was said to consider a classical mechanical theory that worked as the ultimate pleasure of a theoretician. It is a very appreciable thing for experimentalists, too! One fundamental goal in our quest for understanding nature, in my opinion, is to find out which is the simplest theory, based on the simplest principles that can explain a given observation. It goes without saying that all more complex theories built on deeper levels of our knowledge are without substance for the same observation.

This evolution of the basic approach to polarized neutrons was quite naturally also accompanied by a “new wave” of technical developments and “down to earth” applications. Neutron polarizer and analyzer systems we have today are much more efficient than those of 10 years ago. This is giving more and more importance to a second kind of applications of polarized neutrons, viz. to use polarization as a beam modulation technique for the purpose of shaping beams and performing energy analysis. NSE spectroscopy proved to be the first spectacular success of this kind of applications, but the idea of polarized neutron beam modulation goes back to at least 10 years earlier, viz. to Drabkin’s proposal of a magnetic neutron monochromator \([11]\). Another example of this kind is represented by the various schemes proposed and tested in spin flip chopping in time-of-flight spectroscopy \([12]\).

In what follows, as a homage to Cliff’s widely admired taste for the clearest possible understanding of fundamental questions and concepts, I will mostly concentrate on a description of what one might wish to include about our new understanding of spin polarization phenomena in a college textbook, some of it correcting errors figuring in some existing textbooks. A few recent experimental results presented at the end will illustrate the utility of all that footwork. This paper is partly based on lectures I held during the last semester at Technical University Berlin, and it also reflects what I have learned from the questions by the audience. Much of the contents of this paper has not been published before.

Before turning to the subject matter I would like to stress that my above account on the debut of vectorial polarization analysis only refers to a “nouvelle vague” of experiments. Theoreticians, once again, were way ahead of us. The amazing
paper by Halpern and Johnson of 1939 on paramagnetic scattering of polarized neutrons [13] presents the main result in general vectorial form. It took exactly 40 years to prove it experimentally in its generality, as I will describe below. The complete vectorial polarization theory of neutron scattering from samples of any thinkable complexity was also provided by our theoretician colleagues as early as 1962 [14]. These results are of such complexity and contain so many sample parameters that investigation of very special, simplified combinations could or can experimentally be envisaged only. I will elaborate a bit on this aspect of generalized polarization analysis. Let me just stress here that we know that the theory is exact, but we might never find a sample well enough characterized for a meaningful illustration of all the subtleties predicted.

2. The nature of neutron magnetic moment and the Hamiltonian

As mentioned above, this question was one of those Cliff was involved with at the beginnings of polarized neutron research. The reason I come back to it is twofold. Partly, because the old controversy that lasted from 1936 to the experimental settlement in 1951 has mostly been forgotten, together with the real relevance of the question. Partly, because the new experimental methods of our interest involve coils in which neutrons travers regions with non-zero current density, and it is not a trivial question to decide the form of the Hamiltonian to be used in calculating forces and energy changes in such a situation. In fact, the old results contain the answer, but in no obvious way.

Let us first state the problem by considering the force acting on a magnetic moment in a magnetic field, as predicted by the two models of classical microscopic dipole or a classical microscopic Amperian current loop, as illustrated in fig. 1. We will consider the forces given by ordinary electrostatics and magnetostatics in the limit of making the size of the object going to zero with the magnetic moment \( \mathbf{\mu} = ed \) and \( \mathbf{\mu} = I f/c \) in the two cases, respectively, where \( f \) is the surface element vector of the current loop) kept constant. Surpris-

![Fig. 1. Magnetic forces acting on a dipole and on a current loop.](image)

ingly, the two results are different:

\[
F_d = F^d + F^w = (\mathbf{\mu} \cdot \text{grad}) \mathbf{H} = \text{grad}(\mathbf{\mu} \cdot \mathbf{H}) - \mathbf{\mu} \times \text{curl} \mathbf{H},
\]

\[
F_c = \oint \mathbf{F} = \frac{1}{c} \oint \mathbf{f} \times \mathbf{H} = \frac{1}{c} \int \text{grad}(\mathbf{H} \cdot \mathbf{f}) - \frac{1}{c} \int \text{div} \mathbf{H} \, df = \text{grad}(\mathbf{\mu} \cdot \mathbf{H}).
\]

The difference between the force acting on the dipole \( F_d \) and the one acting on the current loop \( F_c \) is nevertheless only non-zero, if the current density at the position of the magnetic moment is non-zero. Thus the well-known equivalence of these two models in electromagnetism of continuous media is fully compatible with this difference, since materials do not penetrate each other. We can also observe that eq. (2) suggests that the energy of the magnetic moment in a field has the simple form \( -\mathbf{\mu} \cdot \mathbf{H} \), while the situation is more complex for the dipole model. (Knowing nature's penchant for simplicity and aesthetics, we could just discard the dipole model at this point. In fact this would not be all that arbitrary; it just turns out to be logically difficult to accommodate in a microscopic theory both dipoles and currents as sources of fields.)

If we consider a magnetic moment inside a medium, we have to think of \( \mathbf{H} \) as the microscopic field. Since we attribute this internal field to the Amperian microcurrents of the electrons, the averaged of the internal microfields on a macroscopic scale gives what is customarily called the magnetic induction of the medium \( \mathbf{B}_m \). (On the microscopic level it does not make sense to distinguish between \( \mathbf{B} \) and \( \mathbf{H} \), and except otherwise stated I will write \( \mathbf{H} \). In some textbooks \( \mathbf{B} \) is used in this context, i.e. as the real physical field in vacuum.) On the other hand, if we would postulate that magnetism in
media is due to microscopic dipoles, then the internal microscopic field would average to what is usually referred to as the magnetic intensity in a medium, \( H_m = B_m - 4\pi M \), with \( M \) being the macroscopic magnetic moment density. Thus there will be two simple limiting cases considering the average force exerted by a magnetic medium on our particle:

a) If both the particle moment and the magnetism of the media is described by Amperian currents:

\[
F_\text{a} = \text{grad}(\mu \cdot B_m); \tag{3}
\]

b) If both of them are described by dipoles:

\[
F_\text{b} = \text{grad}(\mu \cdot H_m). \tag{4}
\]

I have listed these two cases not only because they are the ones which obviously correspond to simple Hamiltonians, but also because the actual debate on the nature of the neutron magnetic moment in the 1930's and 40's ultimately boiled down to the choice between these two Hamiltonians \[15\].

Sure enough, the original controversy never appeared as such a simple matter at that time and there was quite some confusion. In his, with Cliff's expression, "prophetic" paper Bloch \[16\] introduced in 1936 what we today call magnetic neutron scattering, but unfortunately he used the dipole model. Schwinger reexamined the problem using a formal quantum mechanical treatment and arrived at a different expression for the cross section \[17\]. Incorrectly, he believed that this was due to the use of quantum mechanics from the outset in contrast to the classical character of some of Bloch's arguments. In his answer \[18\] Bloch has correctly demonstrated that the difference has nothing to do with the use of quantum mechanics (which would violate the fundamental principle of correspondence), but with the dipole vs. current loop assumption. He has shown that if we assume kind of a Lorentz hole in the electron density around the neutron, the result will depend on the shape of this exclusion hole. This actually demonstrates the old difficulty in magnetostatics with the description of interpenetrating magnetizations, viz. what Bloch was trying to avoid was a not curl-free magnetic field, in contrast to our eq. (1). In a little known paper from 1938 \[19\] Migdal claimed to have demonstrated that the model character of the neutron moment is irrelevant, which would restore the equivalence of the dipole and Amperian current pictures in magnetostatics. In view of the simple example of eqs. (1) and (2) this is of course incorrect, and on closer examination one finds that he used the Amperian model from the outset. Finally Ekstein \[15\] summarized the formal problem as a choice between the two types of Hamiltonians in eqs. (3) and (4), but he did not offer any interpretation for the two assumptions, and incorrectly he maintained the distinction between \( H \) and \( B \) even on the microscopic level. What this hid was a fact that nobody realized at that time, namely that the dipole vs. current loop character of the microscopic sources of magnetism of the medium itself is as much part of the problem as the nature of the neutron moment.

The simplest experiment distinguishing between models (3) and (4) is the total reflection of neutrons on a ferromagnetic mirror magnetized parallel to its surface. As well known, in this field configuration \( H_m \) inside the magnet is the same as the external field, and therefore the neutron will experience no magnetic force on crossing the surface. On the other hand, \( B_m \) jumps at the surface by \( 4\pi M \) leading to an attractive effect on the moment "up" state (moment parallel to \( B_m \)) and a repulsive effect on the "down" state. This latter situation was the one observed by Hughes and Burgy \[2\], confirming that both the neutron magnetic moment and the microscopic nature of magnetism corresponds to the Amperian model.

Beyond the purely intellectual curiosity, a clear understanding of the problem of magnetic forces acting on neutron beams also became an important technical issue in high resolution NSE spectroscopy \[7\]. The resolution of the next instrument to be completed by 1988 at ILL will attain a fraction of a neV. In this method the neutron energy change in the sample scattering process is determined by directly comparing the incoming and outgoing velocities for each neutron individually. This is achieved by making the neutron spins perform large numbers of Larmor precessions, in the opposite sense before and after the scattering, as illustrated in fig. 2. The Larmor precessions are initiated by turning the neutron spins perpendicular-
lar to the magnetic field direction with the help of the first $\pi/2$ flipper coil. The $\pi$ spin flipper next to the sample inverts the apparent sense of the precessions and the second $\pi/2$ coil together with the polarization analyser serves for the observation of the precessing polarization of the scattered beam. The problem with the extreme high resolution now planned is that the flipper coils used to initiate and handle the precessions produce about $10^{-6}$ fields, i.e. Zeeman energies of the same magnitude as the energy resolution of the spectrometer. Thus it became inevitable to analyse the influence of the flipper coils on the velocity of the neutrons. This will be part of the next chapter.

We will conclude this chapter by examining the physical origin of the Hamiltonian $-\mu \cdot H$. It is commonly believed that it corresponds to the mixed term in the energy of the total magnetic field $H + H_n$, where $H_n$ is the magnetic field of the neutron:

$$E_{\text{field}} = \frac{1}{4\pi} \int H \cdot H_n \, dV = \frac{1}{4\pi} \int H_n \cdot \text{curl} \, A \, dV$$
$$= \frac{1}{4\pi} \int A \cdot \text{curl} \, H_n \, dV = \frac{1}{c} \int A \cdot j_n \, dV$$
$$= \int A \cdot \text{curl} \, M_n \, dV = \int M_n \cdot \text{curl} \, A \, dV$$
$$= \mu \cdot H,$$

where $A$ is the vector potential for $H$, and $j_n = c \text{curl} \, M_n$ the current density for $H_n$. This argument gives the correct result in electrostatics, but as we see, in magnetism it gives the negative of the correct answer. (This wrong sign has been overlooked in some otherwise valuable textbooks.) The explanation of this apparent discrepancy is that if magnetic moments are moving with respect to each other there is a mutual induction effect which tends to change the currents of which the fields originate. It can be shown [20] that the electric energy required in order to keep these currents constant (e.g. provided by the power supply if the field is produced by a coil) is exactly equal to the change of $-\mu \cdot H$ for both the neutron and the source of the external field $H$. Thus the total energy balance is indeed:

$$\Delta E_{\text{field}} + \Delta E_{\text{neutron}} + \Delta E_{\text{induction}} = \Delta (-\mu \cdot H).$$

It might appear somewhat surprising that the apparently local interaction term $-\mu \cdot H$ in reality implies the action by distant objects, which might just stress the fundamental nonlocality of quantum mechanics.

3. Vectorial beam polarization

In order to be able to describe not just “up” and “down” but general spin states we only have to consider the general form of a spin-1/2 wave function, which is a coherent superposition of the two basic states:

$$|\psi\rangle = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$$

It is well known, that with this wave function the expectation values of the Pauli (spin component) operators form a unit vector $S = (S_x, S_y, S_z)$, which can be considered as the corresponding classical spin direction vector and can be characterized by the two polar angles $(\theta, \phi)$ [10]. The relation between these angles and the coefficients are:

$$\alpha = \cos \frac{\theta}{2}, \quad \beta = e^{i\phi} \sin \frac{\theta}{2}.$$

Furthermore, it can be shown by rigorous quantum mechanical analysis [10] that for a neutron beam in a magnetic field in which the neutron
velocity changes produced by the magnetic forces due to the inhomogeneities of the field [cf. eq. (2)] are in any direction negligible compared with the beam velocity spread, the "classical spin vector" \( S \) evolves according to the classical equation of motion

\[
\frac{dS}{dt} = \gamma_1 [S \times H]. \quad \gamma_1 = 2.916 \text{ kHz/Oe.} \tag{10}
\]

In other words, in the absence of appreciable Stern–Gerlach beam splitting effects, the neutron spin motion reduces to classical Larmor precessions around the instantaneous magnetic field direction at the position of the pointlike neutron. Thus, incidentally, for spin-1/2 particles the real quantum behaviour coincides with the classical one, in contrast to the partial picture of "up" and "down" quantum states. Obviously, if we deal with beams composed of neutrons with different spin vectors \( S \), the observable average expectation value of the spin components will define the beam polarization as \( P = \langle S \rangle \).

All of the spin flip (or turn) devices developed by now [4,5,6,7] contain sheets of current carrying material across the neutron beam, i.e. the neutrons traverse a non curl-free field region. Thus in view of eqs. (1) and (2), we really need to know that it is actually the Amperian model of eq. (2) which applies. Being the special case of interest for NSE spectroscopy we will examine the action of the flat coil flipper device introduced in ref. [7], and shown in fig. 3. Since in eq. (2) the force is derived from a bona fide potential energy \(-\mu \cdot H\), we can apply the method of wavefunction matching at finite potential steps. For this purpose we can consider the current sheet forming the side of the coil as infinitely thin. Assume that the neutron arrives with \( S \) parallel to the external field \( H \), i.e. \( |\chi\rangle = |\uparrow\rangle \). On the internal side of the coil the same \( S \) vector will be represented by the state

\[
|\chi\rangle' = \cos \frac{\theta}{2} |\uparrow\rangle' + \sin \frac{\theta}{2} |\downarrow\rangle'. \tag{11}
\]

where "up" and "down" refers to the direction of the resulting field \( H' \) inside the coil (the angle \( \phi \) is measured with respect to the coil plane). These two representations of the spin state at the current sheet will only be identical for all times, if the total energy of each stationary eigenstate involved is equal, i.e.

\[
\frac{\hbar^2 k^2}{2m} - \mu H = \frac{(h k' - \mu H')^2}{2m} + \mu H'. \tag{12}
\]

where \( k, k' \) and \( k' \) are the neutron momenta associated with the \( |\uparrow\rangle \) and \( |\downarrow\rangle \) spin states. Thus inside the coil where the spin flipping Larmor precession takes place (cf. fig. 3), we have to do with two different neutron wave numbers. It can be shown that the Larmor precession, which is fundamentally an interference effect between the "up" and "down" spin states (cf. eq. (8) and e.g. \( S = \alpha \beta + \beta \alpha \)), is due to the wave beating between these slightly different wave vectors [21]. The same analysis shows that on leaving the coil after e.g. 180° precession, with an \( S \) vector perpendicular to the external field \( H \), as illustrated in the figure, the wave function will again be the superposition of two, different wavenumber components with the same total energy \( E \).

\[
E = \frac{(h k)}{2m} - \mu H = \frac{(h k')}{2m} + \mu H. \tag{12}
\]

Thus our conclusion is that the flipper action conserves the neutron energy and does not interfere with the determination of the inelasticity of the scattering process.

It is worth stressing that the Larmor precessions as interference effects reproduce all fundamental features of any other beam interference phenom-
enon. They can be produced by introducing a beating between two different wave numbers, as we have seen, or by switching on a homogeneous magnetic field, in which case the energies of the "up" and "down" spin components are split by the amount of $2\mu H$. Thus the precessions occur as beating between two slightly different frequencies in time. This is essentially what happens in nuclear magnetic resonance [10]. Fig. 4 illustrates Larmor precessions observed as function of the precession field value (actually in a spin echo configuration). Exactly the same signal can be obtained as function of distance (flightpath) in a constant field. The decay of the precessing polarization on both sides corresponds to a dephasing of the precessions due to the neutron velocity spread in the beam, if we consider the beam as an ensemble of pointlike classical particles. If we consider the neutron beam as an ensemble of plane waves with different momenta, the dephasing is due to the fact that the same magnetic field will introduce different beating distances for different initial neutron momenta. Finally, if we consider the beam as consisting of identical wave packets, the same observation will be interpreted as an image of the wave packet, showing its minimal dimension in terms of wavelength units. The observation of Larmor precessions does not allow us to distinguish between these various assumptions on the particle states in the beam. This is a common fundamental feature of all neutron beam interference experiments performed by now.

4. Coherence and vector polarization analysis

In conventional "up-down" polarization analysis we can describe the neutron spin change in the scattering process by 4 scattering cross sections [8] which give the transition probabilities from an initial fundamental spin state into a final one: $t_{\uparrow\uparrow}$, $t_{\uparrow\downarrow}$, $t_{\downarrow\uparrow}$, and $t_{\downarrow\downarrow}$. (These cross sections will in general depend on the orientation of the magnetic field determining the "up-down" directions at the sample with respect to the neutron momentum transfer vector and the sample orientation.) If, for example, the scattered beam polarization would happen to be not parallel to $H$, this would imply that there is a coherence between the scattered "up" and "down" wave components, cf. eqs. (8) and (9). In order to observe the vectorial character of the scattered beam polarization we need to make sure that it will not loose its precessing (perpendicular to $H$) component by dephasing due to inhomogeneities of $H$ across the beam cross section or to the neutron velocity differences in the beam. No such precautions are required if we are only interested in the "up-down" $P_z$ component, and this is the reason why there are only very few examples of vectorial polarization analysis, other than 3-dimensional depolarization studies and NSE. Fundamentally, vector polarization analysis requires that the sample be surrounded by a well defined region of very small and homogeneous field. All the "new wave" techniques mentioned in the introduction can then be applied for the analysis, essentially by turning the various components of the spin direction into that of $H$ for the purpose of determining them one by one using a usual, $P_z$-sensitive analyser. This possibility was first demonstrated by Alperin in 1972 [22] by the observation of a scattered beam polarization perpendicular to the incoming beam polarization in a particular Bragg reflection on a non-centrosymmetrical crystal.

The very general principles of quantum mechanics provide a simple framework for the description of this kind of through-the-scattering coherence. Considering the scattering on an object in a well defined quantum mechanical state, the incoming and outgoing spin states (actually for each given neutron momentum and energy transfer) will be related by a linear operator, the so-called the $\hat{S}$-matrix:

$$|x\rangle = \hat{S} |\alpha\rangle = \begin{pmatrix} a_{\uparrow\uparrow} & a_{\uparrow\downarrow} \\ a_{\downarrow\uparrow} & a_{\downarrow\downarrow} \end{pmatrix} |\alpha\rangle.$$  (13)
Obviously, the above mentioned 4 transition probabilities \( t_{11}, t_{12}, \ldots \) correspond to \( |a_1^2 \rangle, |a_1^3 \rangle, \ldots \). The new aspect in 3D polarization analysis is the determination of cross products of these complex matrix elements. A simple, experimentally verified example is the spin turning effect produced by polarizing neutron mirrors [9,23]. In this particular case the nondiagonal elements of the \( \hat{S} \)-matrix were found to be negligible. Thus the reflected beam spin wave function is given as

\[ |\chi'\rangle = a_{\uparrow}, \alpha |\uparrow \rangle + a_{\downarrow}, \beta |\downarrow \rangle. \]

The effect of the polarizer is that it changes the ratio of the amplitudes of the "up" and "down" components in the coherent superposition, i.e. in view of eq. (9), the angle \( \theta \) between the spin vector and the field \( H \). This angle can be experimentally determined by taking the ratio of the \( x \) and \( z \) spin components at \( \phi = 0 \). Theoretically, in view of eqs. (8) and (9),

\[ \frac{\theta'}{2} = \frac{|a_{\downarrow}, \beta|}{|a_{\uparrow}, \alpha|} = \frac{1}{\sqrt{R}} \frac{|\beta|}{|\alpha|} = \frac{1}{\sqrt{R}} \frac{\tan \theta}{2}. \]  

(14)

where \( R \), the flipping ratio of the polarizer denotes \( |a_{\downarrow}^2|/|a_{\uparrow}^2| \). The results shown in fig. 5 were found to be in good agreement with eq. (14), which implies nearly perfect phase coherence between the two matrix elements in question.

The evaluation of the most general consequences of the existence of the \( \hat{S} \)-matrix is straightforward but tedious, let alone the experimental verification of other than special cases. For completeness I just give the results, which are well known from nuclear theory [24]. In order to simplify matter we make use of the fact that any \( 2 \times 2 \) complex matrix can be represented as

\[ \hat{S} = h\hat{I} + a \cdot \hat{a}. \]  

(15)

where \( \hat{I} \) is the unit matrix and \( h \) a complex number, and \( \hat{a} \) stands for the three Pauli matrices with \( a \) being a complex vector. In these terms one gets:

\[ \langle \chi'|\chi\rangle = h^*b + a^* \cdot a + S^*(h^*a + a^*h) \]

\[ + iS^*(a^* \times a) \]  

(16)

and

\[ S'<\chi'|\chi'> = h^*a + a^*b - i(a^* \times a) + Sh^*b \]

\[ + ib^*(a \times S) + i(S \times a^*)b + (a^* \cdot S)a \]

\[ + a^*(S \cdot a) - S(a^* \cdot a). \]  

(17)

The fact that we have obtained these well-known relations without assuming anything specific about the sample is an important generalization. It just stresses that no matter what kind of complexities a sample might display, from the point of view of what the neutron spin sees it will be fully characterized by a complex scalar and a complex vector, \( b \) and \( a \) (which of course depend on the neutron momentum and energy transfer \( q \) and \( \omega \)). An interesting feature is that if the incoming beam polarization is unity, i.e. it corresponds to a single spin vector \( S \), the scattered beam polarization will also be unity, viz. \( P' = S' \).

This latter appears to contradict experimental observations such as that scattered beam polarization in the case of nuclear spin incoherent scattering is \( P' = -P/3 \). We have to remember, however, that eqs. (16) and (17) only apply for a sample in a well defined initial quantum state. At finite temperatures this can not in principle be fulfilled. The effect of finite temperature is that the sample state changes with time during the experiment, so there is an averaging over inhomogeneities in time taking place. Most samples are
also inhomogeneous in their volume. Furthermore, if the scattering cross section has a finer structure in \( q \) and \( \omega \) than the instrumental resolution, there will be an averaging over these variables, too. What is called “incoherent” scattering is an example for the latter. With a \( q \) resolution corresponding to the inverse of the sample size we would only observed coherent scattering reflecting in all detail the scattering length of each individual nucleus at each atomic site. With the practically available \( q \) resolutions all we see is a \( q \) average of this coherent scattering (fortunately) and this average is called “incoherent” scattering. Thus for most samples we have to perform all this averaging over the space-time inhomogeneities, and eventually also take into account the final resolution. This is actually the much harder task, because in general it is not possible to introduce an average \( \hat{S} \)-matrix, since averaging first of all destroys the coherence the \( \hat{S} \)-matrix is meant to express. We can only maintain the \( \hat{S} \)-matrix description for scattering effects due to features constant over the time of data collection period in spatially homogeneous samples. Since the lifetime of vibration phenomena is always rather short, only the structural features of solid samples fall into this category. In particular eqs. (16) and (17) apply to Bragg scattering on monodomain single crystals, also allowing for nuclear spin order as part of the global order structure.

Averaging over the space-time inhomogeneities of the sample fundamentally boils down to evaluating average values of various products of matrix elements of the \( \hat{S} \)-matrix. This is rather simple matter for the self products of the type \( |a_j^2| \), where phase relations (coherence) play no role. Quite obviously, the averaged absolute squares of the 4 \( \hat{S} \)-matrix elements will now correspond to the 4 scattering probabilities in classical “up-down” polarization analysis [8]. The cross products, on the other hand, are related to phase correlations between the various matrix elements, and can lead to a tremendous variety of behaviors, even if the average absolute square of each element is fixed by an “up-down” type study. For example by changing the magnetizing field applied on polarizing mirrors various degrees of coherence could be observed between different matrix elements [23]. Ultimately the full polarization behavior will be determined by the 16 independent averages \( a_{ij}^*a_{kl} \). (Note the cross products of elements in inverted order are the complex conjugates of each other, but since both the real and imaginary parts of the averages are of interest such a pair counts as two independent averages.) On the other hand, experimentally we can also determine 16 independent parameters. Namely, due to the two degrees of freedom of the spin-1/2 Hilbert space, measured quantities must linearly depend on the beam polarization \( P \) (see e.g. ref. 10). Thus the crosssection and the scattered beam polarization \( P' \) most generally can be given as

\[
\langle x' | x' \rangle = A + B \cdot P, \quad P'\langle x' | x' \rangle = C + TP.
\]

Where \( A \) is a scalar, \( B \) and \( C \) are vectors and \( T \) is a tensor, all of them real, of course. It remains to be shown that the system of 16 linear equations formed by the relations between the averages \( a_{ij}a_{kl} \) \((i, j, k, l = \uparrow \) or \( \downarrow \)) and the components \( B, C \) and \( T \) is solvable. This is conveniently achieved by indentifying the 16 relevant averages in terms of parameters \( b \) and \( a \) introduced in eq. (15), namely

\[
\begin{align*}
\hat{b}^*b, & \quad a^*_aa_a, \quad \text{Re}(a^*_aa_b), \quad \text{Im}(a^*_aa_b), \quad \text{Re}(b^*a_a), \quad \text{Im}(b^*a_a)
\end{align*}
\]

\((\alpha = x, y, z \) and \( \beta \neq \beta \)). It can simply be verified comparing eqs. (16), (17) and (18), that the 4 averages \( \hat{b}^*b \) and \( a^*_aa_a \) \((\alpha = x, y, z \) are uniquely determined by \( A \) and the diagonal elements of \( T \), and the remaining 12 equations break down in 6 simply solvable pairs, e.g.

\[
B_\alpha = 2 \text{Re}(b^*a_a) + 2 \text{Im}(a^*_aa_a),
\]

\[
C_\alpha = 2 \text{Re}(b^*a_a) - 2 \text{Im}(a^*_aa_a),
\]

where \( \alpha, \beta, \gamma \) form a cyclical permutation of \( x, y, z \).

With the usual terminology, these averages are correlation functions, and they are functions of \( q \) and \( \omega \). Thus, as a consequence of the spin degrees of freedom, any kind of behaviour in a sample can be described by not more than 16 independent correlation functions in the \((q, \omega) \) space. It has to be stressed that this number 16 is determined by the neutron spin value of 1/2 (2-dimensional spin variable space) and fully independent of the number of spin degrees of freedom and complexity of the sample.
In sum, if a sample can be described by a single $\hat{S}$-matrix in the spin variable, the polarization behaviour of the sample scattering at given $q$ and $\omega$ can be characterized by maximum 7 independent parameters (fixing the phase of one matrix element arbitrarily). These 7 parameters, i.e. the $\hat{S}$-matrix can be fully determined by vectorial polarization analysis (which can provide up to 16 experimental parameters) but not with the 4 parameters provided by conventional “up-down” polarization analysis. If, on the other hand, we have to average over the space-time inhomogeneities of the sample, there can be up to 16 independent correlation functions characterizing the degree of incoherence introduced by these inhomogeneities, which are also uniquely determined by the equal number of measurable quantities vectorial polarization analysis experiments can provide.

To conclude this chapter, we will examine the very instructive case of paramagnetic scattering. Halpern and Johnson’s original expression is a vectorial relation [13]:

$$P' = -q (P \cdot q) / q^2.$$  \hfill (19)

The fact that the scattered beam polarization vector $P'$ can be nonparallel to the incoming one $P$ implies in view of eqs. (8) and (9) that there is a coherence between the “up-up” non-spin-flip and “up-down” spin flip scattering amplitudes. This is quite amazing since the paramagnetic state is defined by the lack of any correlations between the various spin components. Closer analysis, which can be most simply based on eq. (17), reveals [25] that this correlation is simply due to the fact that the neutron spin only couples to the component of the sample magnetization perpendicular to the momentum transfer vector $q$. This dependence on the direction of $q$ is the only correlation the neutron will experience between the fully uncorrelated paramagnetic spins. The same analysis also shows that eq. (19) is more generally valid not only for paramagnets, but for any macroscopically isotropic system, such as multidomain or polycrystalline antiferromagnets.

For 40 years relation (19) has only been verified in terms of the “up-down” picture, i.e. taking only the component of $P'$ parallel to $P$ (which latter one will be taken of unit absolute value, somewhat simplifying the experimental situation):

$$P' = P' \cdot P = - (P \cdot q)^2 / q^2 = - \cos^2 (P, q).$$  \hfill (20)

This equation itself has a very interesting feature. The right hand side is the square of the direction cosine of the momentum transfer vector $q$ with respect to $P$. If we determine $P'$ for three mutually perpendicular but otherwise arbitrary directions of $P$, say $X$, $Y$ and $Z$, the sum of the three $P'$ values will be minus unity:

$$P'(X) + P'(Y) + P'(Z) = - \cos^2 (X, q) - \cos^2 (Y, q) - \cos^2 (Z, q) - 1.$$

In contrast, for nuclear scattering effects with $P' = P$ the same sum is 3. (Nuclear spin incoherent scattering, however, also gives a sum of $-1$ in view of the relation $P' = -P/3$. This is no accident.) This allows to identify the paramagnetic scattering cross section even if the direction of $q$ is not uniquely determined (e.g. broad inelasticity or use of large detector areas). This property was first put to application by the Leningrad group [26]. If we know more about the direction of $q$, the scheme can be simplified and made more efficient. For example if $q$ lies in the $X$, $Y$ (say horizontal) plane, $P'(Z)$ will be zero, i.e. with $P$ vertical one can observe the contribution of the background nuclear scattering alone, and subtract it from the sum $P'(X) + P'(Y)$ in order to obtain the full paramagnatic contribution, separated from the nuclear background including nuclear spin incoherent scattering this time [25]. The situation is even simpler if $q$ is fixed in a direction, say parallel to $X$, in which case $P'(Y)$ is also zero.

Performing a sequence of conventional “up-down” polarization analysis measurements with different incoming polarization directions $P$, as just described, is an important generalization of the original scheme of ref. 8. In practice it implies that each time a guide field of 10-100 Oe and parallel to $P$ is applied to the sample, and therefore the whole scheme can only work if this field has no effect on the state of the sample, e.g. as in high temperature paramagnets. It can be shown that such a sequence of measurements with three independent $P$ directions can provide the following 10 out of 16 independent correlation functions
described above: $b^*a_{b}, a^*a_{a}$, $\text{Re}(b^*a_{a})$, $\text{Im}(a^*a_{a})$. If we know for sure that the sample can be described by a single $S$-matrix, these 10 products allow in general the determination of the $S$-matrix, i.e. $b$ (arbitrarily taken as real), and the six parameters of the complex vector $a$. However, one could not decide if the system corresponds to a single $S$-matrix or to an average. (E.g. $\text{Re}(b^*a_{a}) = 0$ and $\text{Im}(a^*a_{a}) = 0$ can as well mean that $a_{a}$'s are all purely imaginary and $b$ real in a single $S$-matrix, as well as that they are incoherent.) In contrast, the general vectorial polarization scheme allows such distinctions to be made, of course. Namely, single $S$-matrix samples are characterized by equalities of the type

$$ (b^*b)(a^*a_{a}) = \text{Re}(b^*a_{a})^2 + \text{Im}(b^*a_{a})^2. $$

The vector character of eq. (19), as opposed to the scalar relation eq. (20), has only been experimentally tested up to now in the so called Paramagnetic Neutron Spin Echo (PNSE) method [25]. In this method the spin flipper coil next to the sample is removed (cf. fig. 2) and thus nuclear scattering effects will not give an echo signal. However, paramagnetic scattering will do so just because of the polarization change given by eq. (19). With the $q$ vector lying in the Larmor precession plane, i.e. perpendicular to the small magnetic field applied to the sample, neutrons arriving to the sample after having precessed over an angle $\phi$ (which can be of many thousands of radians), will assume after the scattering a polarization $P' = \cos \phi$ in the direction of $q$. (We measure $\phi$ with respect to the direction of $q$, for simplicity.) After the scattering they will precess by another angle $\phi$, and thus the final polarization component in the direction corresponding to $q$ will be given as

$$ P_{\text{NSE}} = \langle \cos^2 \phi \rangle = \frac{1}{2}. \tag{21} $$

Here the average is taken over the barely monochromatic beam, in which $\phi$ varies by many times $2\pi$. This result means in practice, that with a fully polarized incoming beam, the polarization of the PNSE signal is $1/2$, if there are no dephasing effects due to the inelasticity of the scattering (which would make before and after scattering precession angles different). This has been found experimentally. Note, that if we would only know of eq. (20) and assume that $P'$ is parallel to $P$, we would still have a PNSE signal but with a polarization amplitude of $\langle \cos \phi \cdot \cos 2\phi \rangle = 1/4$. The observation of the $1/2$ amplitude of the PNSE signal in 1979 [27] was the first confirmation of the vectorial implications of eq. (19) dating from 1939.

5. Applications and examples

Vectorial polarization analysis has been demonstrated in neutron diffraction [20], but it has not been systematically applied yet. This is mainly due to the delicate magnetic environment it requires around the sample region. A dedicated instrument using sophisticated magnetic shielding techniques is being tested at the ILL by Francis Tasset and collaborators and will be operational in the near future [28].

On the other hand, 3D polarization analysis has been quite widely used in studying the depolarization of a neutron beam transmitted through a ferromagnetic sample. This provides information on the sample domain structure, and the observation of a $3 \times 3$ depolarization matrix describing the vectorial relation between the incoming and outgoing polarizations [29] provides much more information than the 3 parameters observable in the conventional $P||P'$ approach. An interesting example showing the rearrangement of domains in time after a field reversal is given in fig. 6 [30].

NSE experiments represent the bulk of vectorial polarization work performed by now. In this method, the Larmor precessions are primarily used for beam modulation in order to perform high resolution analysis of the scattering inelasticity. However, as we have seen above, in the PNSE variant this is combined with an unambiguous identification of the magnetic scattering effects, too. The figures below show some recent results which highlight the particularly useful features of this method. All these experiments have been performed using the IN11 machine at ILL [31], schematically illustrated in fig. 2. Fig. 7 illustrates the determination of the apparent diffusion constant of pig Immunoglobulin-G molecules in D$_2$O solution [32]. The measured linewidths are the ultimate limits of what can be resolved by neutrons today. The straight line represents a diffu-
molecule on the length scale corresponding to the $q$ range of the NSF experiment. The $q$ and $\omega$ range shown in the figure is at present only accessible to NSF measurements.

The NSF method is particularly well adapted for the study of inelastic lineshapes. This is primarily due to the Fourier transformation inherently performed by the Larmor precessions, which reduces the instrumental resolution corrections to a simple division as opposed to the usual deconvolution. Furthermore, in the two examples shown in figs. 8 and 9 the instrumental resolution was so much better than the effects investigated (respectively 0.05 $\mu$eV and 0.1 $\mu$eV) that such corrections were negligible anyway. In fig. 8 the directly measured $S(q, t)$ lineshape is shown in EuO powder sample at the ferromagnetic Curie point $T_c = 69.3$ K [33]. The best agreement was obtained by the assumption of a simple exponential decay $\exp(-t')$, which corresponds to a Lorentzian line in the energy $\omega$ domain (continuous line, $t' = 0.79$ $\mu$eV). The dashed line illustrates the best possible fit assuming the lineshape proposed by Wegner and Hubbard on the basis of mode-coupling theory [34], and it is clear that this lineshape is at variance with the experiment. Note that the data were obtained at a small $q$ value where the theory was expected to apply. This is the first experiment

![Fig. 6](image1)

**Fig. 6.** Various elements of the transmitted beam depolarization matrix $D$ as a function of time, following a fast reversal of the magnetizing field applied on a Ni sample [30].

![Fig. 7](image2)

**Fig. 7.** Relaxation rate as a function of wave number in a D$_2$O solution of pig Immunoglobin-G, as observed in a NSF experiment [32]. The straight line corresponds to a diffusion constant $D = 2.74 \times 10^{-7}$ cm$^2$ s$^{-1}$.

![Fig. 8](image3)

**Fig. 8.** The $S(q, t)$ spectrum of spin fluctuations in EuO at the ferromagnetic Curie point [33]. The continuous line and the dashed line correspond to best fits to Lorentzian and Hubbard–Wegner lineshapes, respectively.

![Fig. 9](image4)
The inset shows the $S(q, \omega)$ spectrum corresponding to the curve fitted to the $T_c - 3$ K data using magnon theory with dipolar interaction and including a central diffusive component (dotted line) [35].

in which it was technically possible to distinguish between these two lineshapes at a constant $q$. Fig. 9. shows the temperature dependence of the $S(q, t)$ scattering spectra in another ferromagnet, Fe just below $T_c = 1040$ K [35]. Since the sample is fully depolarizing the beam, a new twist of NSE had to be used, the Intensity Modulation NSE, which is technically related to the Neutron Spectral Modulation (NSM) method, which also makes use of Larmor precession modulation [36]. The insert gives the $T_c - 3$ K results transformed into the energy domain. The spectrum reveals no third peak between the two magnon lines, which should correspond to the longitudinal spin fluctuations. Closer analysis shows (using a rough estimate of the intensity of the longitudinal contribution) that the width of this quasielastic line is rather broad (see dotted line in the inset) and this is why the expected central peak is absent and, of course, consistently escaped detection over the years. This spectrum shows the lowest energy magnon peaks ever observed.

The last example is of a quite different application of NSE spectroscopy, which is rather more complicated to explain. It concerns the measurement of the linewidth of phonons with a resolution of about 0.1% of the phonon energy. The principle of the method has been described in detail elsewhere [37,25], and I only include it because it is bound to become more important in the future. The latest results on the linewidth of acoustic phonons in superfluid $^4$He at various temperatures and momenta are shown in fig. 10. [38]. The open symbols are NSE results, the others high resolution triple axis data. The phonon energies in this $q$ range vary between 640 and 890 $\mu$eV. The NSE results at $q = 0.4$ and 0.5 Å$^{-1}$ set an upper limit for the simple 3 phonon decay rate, which happens to be somewhat lower than preliminary theoretical estimates [39]. This decay process becomes kinematically forbidden for higher $q$ values, and the drop of the linewidth at the lowest to temperatures beyond $q = 0.5$ Å$^{-1}$ might be a hint to this effect. The temperature dependence of the linewidth is due to 4 phonon processes. NSE is the only currently available method for the determination of the lifetime of elementary excitation with this kind of resolution.

6. Conclusion

Those of us who were involved in polarized neutron scattering work undoubtedly experienced the special fun it offers. Probably the most amazing about it are the huge effects one can provoke by manipulating the only handle we can grasp at on the neutron, i.e. its spin. The manipulations only involve changing of some currents energizing
some simple coils, most importantly, without any physical movement of spectrometer parts. Cliff played a decisive role in the debut of polarized neutron work and he might well be the first one who realized the great potentials polarized neutrons provide to precision experiments by allowing measurements made without any changes in spectrometer geometry. The generalization of the classical “up-down” polarization methods he pioneered to vectorial beam polarizations opened up a variety of opportunities in pursuing both types of goals Cliff set the example of, and for which he has educated the taste of all of us in the neutron field. One of them is the constant search for more precise and more powerful tools for what is the everyday bread of neutron science: the study of condensed matter. The other of them is the demonstration and verification of the fundamental principles of microphysics with the best achievable precision, which also makes a body of beautiful and fascinating experimental material available for making the teaching of physics richer, deeper and more fun. As all of us in neutron scattering, I am most grateful to Cliff for the inspiration his example means to us.

References