Neutrons and Magnetism

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1. Introduction

Neutrons, particularly neutron scattering techniques, are fundamentally important for the study of magnetic structures and dynamics. Indeed, one of the definitions for an ordered magnetic structure is that neutron diffraction experiments must reveal magnetic Bragg peaks, and neutron scattering is currently the only technique capable of making a complete study of magnetic excitations. Conversely, the need to understand the physics of magnetism is probably the greatest driving force behind the increasing demand for neutron sources, with developments in diverse fields spanning spintronics to superconductivity and quantum critical points. Only the simplest magnetic problems can be solved without some information from neutrons.

The success of neutron techniques is due to the strength of the interaction between the magnetic moment of the neutron, given by its spin angular momentum, with the magnetic induction of a sample. This interaction is similar in magnitude to that between the neutron and a nucleus, thus the magnetic signal can often be clearly distinguished from nuclear structure and dynamics.

The two-hour lecture will cover the basics of neutron scattering, starting with the wave equation and developing to the first Born Approximation. A discussion, with examples, of the application of neutron scattering to problems of magnetic structure (associated with elastic scattering) and dynamics (inelastic scattering) will follow, with emphasis on the extra information that can be determined with polarized neutrons. A brief introduction to the rapidly expanding field of experiments with neutron dynamical scattering will be made. Finally, mention will be made of spherical neutron polarimetry, which enables the well known ‘phase problem’, common to all scattering techniques, to be overcome.

2. The Born Approximation

The theory behind all the techniques that use neutrons to probe magnetic properties starts with solving the wave equation for the neutron/sample ensemble:

$$\left[-\left(\hbar^2/2M\right)\nabla^2 + \hat{V}(\mathbf{r})\right]\psi = E\psi \ . \qquad (1)$$

For scattering experiments, the probability of scattering, or *cross-section*, can be derived by solving the wave equation and finding the expectation values for E.

The interaction between the neutron and the sample is generally weak, so equation (1) is often solved by first-order perturbation theory. If the neutron is assumed to be a plane wave, the probability of it scattering in to a certain solid angle with a certain energy change, $\mathrm{d}^2\sigma/(\mathrm{d}\Omega\cdot\mathrm{d}E)$, comes from Fermi’s Golden Rule:

$$\begin{aligned}\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\cdot\mathrm{d}E} &= \frac{k'}{k}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \sum_{\zeta,s} p_\zeta p_s \sum_{\zeta',s'} \left|\left\langle \boldsymbol{k}',s',\zeta' \middle| \hat{V}(\mathbf{r}) \middle| \boldsymbol{k},s,\zeta \right\rangle\right|^2 \delta\left(\hbar\omega + E_\zeta - E_{\zeta'}\right) \\ &= \frac{k'}{k}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \sum_{\zeta,s} p_\zeta p_s \sum_{\zeta',s'} \left|\left\langle s',\zeta' \middle| \int \hat{V}(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \middle| s,\zeta \right\rangle\right|^2 \delta\left(\hbar\omega + E_\zeta - E_{\zeta'}\right)\end{aligned} \ . \qquad (2)$$

The expression $\mathrm{d}^2\sigma/(\mathrm{d}\Omega\cdot\mathrm{d}E)$ is known as the *partial cross-section* for scattering. Eq. (2) shows that the cross-section depends upon the Fourier transform of the scattering potential over all positions in the sample, thus a Fourier analysis of an experiment can reveal all the structural and dynamic properties determined by $\hat{V}(\mathbf{r})$.

The two assumptions necessary for the derivation of eq. (2) make up the first Born Approximation, and the majority of all neutron scattering experiments are interpreted from theory developed from this equation.

2.1 Magnetic scattering

The potential energy operator, $\hat{V}(\mathbf{r})$, sometimes known as the *pseudo-potential*, describes the interaction of the neutron with the sample. It can be broken up in to two parts:

$$\hat{V}(\mathbf{r}) = V_n(\mathbf{r}) + V_m(\mathbf{r}) \ . \tag{3}$$

The second term in eq. (3), $V_m(\mathbf{r})$, describes the interaction of the neutron with the magnetic induction, $\mathbf{B}(\mathbf{r})$, and may be written:

$$V_m(\mathbf{r}) = -\gamma\mu_N\hat{\boldsymbol{\sigma}}\cdot\mathbf{B}(\mathbf{r}) \ . \tag{4}$$

where $\hat{\boldsymbol{\sigma}}$ is the Pauli operator defining the neutron spin. The magnetic induction incorporates all the magnetic and electromagnetic properties in and around the sample. Magnetism due to unpaired electrons, domain walls, induction, external fields, and local circuits are all measurable with neutron scattering, although very different instruments are often needed to probe different length and energy scales.

The interaction of the neutron with the nucleus, $V_n(\mathbf{r})$, may be written:

$$V_n(\mathbf{r}) = \frac{2\pi\hbar^2}{m_n}\left(b + B\hat{\mathbf{I}}\cdot\hat{\boldsymbol{\sigma}}\right)\delta(\mathbf{r}) \ , \tag{5}$$

where b is defined as the nuclear scattering length, $B\hat{\mathbf{I}}$ is the nuclear spin. While rarely investigated, it is possible to use neutrons to measure magnetic properties and correlations between nuclei, which are modulated by the nuclear spin.

The cross-section is proportional to components of the magnetization in the sample. Following from the laws of electrodynamics, and in the limit that $\hat{V}(\mathbf{r}) = V_m(\mathbf{r})$, eq. (2) may be written:

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\cdot\mathrm{d}E} = \frac{k'}{k}(\gamma r_0)^2\sum_{\zeta,s}p_\zeta p_s\sum_{\zeta',s'}\left|\langle s',\zeta'|\hat{\boldsymbol{\sigma}}\cdot\mathbf{M}_\perp(\mathbf{Q})|s,\zeta\rangle\right|^2\delta(\hbar\omega + E_\zeta - E_{\zeta'}) \ , \tag{6}$$

where $\mathbf{M}_\perp(\mathbf{Q})$ is the Fourier transform of the components of the magnetization at positions $\mathbf{r}$ in the sample that are perpendicular to the momentum transfer of the neutron beam, $\mathbf{Q}$.

The spatial distribution of the magnetization must be accounted for in the Fourier-transformed $\mathbf{M}_\perp(\mathbf{Q})$. Accounting for the spatial distribution gives rise to a modulation of the intensity of the magnetic scattering as a function of $\mathbf{Q}$, known as the *magnetic form factor*. No form factor is observed for nuclear scattering – the nucleus is assumed to be point-like, as shown by the delta function in eq. (5).

Most neutron scattering experiments are carried out with a random orientation of the neutron spin relative to the sample and the instrument. Further information can be obtained if the neutron beam is *polarized*, that is, the incident neutron spins all have the same direction. The cross-section then becomes a function of the change in the neutron direction, energy and moment direction. Polarized neutron experiments can be used for the separation of magnetic and nuclear scattering, for the unambiguous determination of complex magnetic structures, and for the measurement of the direction of the magnetization inside a sample. Most measurements with polarized neutrons constrain the initial and final neutron spin directions to be co-linear, either parallel (+) or antiparallel (–) to a given direction. In this case the pseudopotential can be split in to four possibilities depending on the neutron spin orientation before and after scattering, and may be written:

$$\begin{aligned} U^{ss'} &= \langle s' | \hat{V} | s \rangle, \\ U^{\pm\pm} &= b \pm BI_z \mp \gamma r_0 M_{\perp z}, \\ U^{\pm\mp} &= -\gamma r_0 \left(M_{\perp x} \pm iM_{\perp y} \right) + B\left(I_x \pm iI_y \right) \end{aligned} \tag{7}$$

The subscripts x, y, and z are here defined such that z is the direction of the initial neutron polarization. The polarization dependent cross-sections are then derived from eqs. (2) and (7) and are a function of the polarization orientation relative to $\mathbf{Q}$.

2.2 Elastic scattering

Neutron elastic scattering, or diffraction, is used to determine magnetic structures, both local and long-ranged. In the elastic limit, eq. (2) may be written:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \propto \int \left| \left\langle \hat{V} \right\rangle \right|^2 e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot \mathrm{d}\mathbf{r} + \left(\left| \left\langle \hat{V}^2 \right\rangle \right| - \left| \left\langle \hat{V} \right\rangle \right|^2 \right) \int z(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot \mathrm{d}\mathbf{r} \ . \tag{8}$$

where the brackets show that all the relevant averages have been made.

The first term in eq (8) is due to the mean structure of the sample and gives rise to Bragg scattering. Complicated magnetic structures will have magnetic Bragg peaks that can be indexed and analysed using crystallographic techniques. Proof of the existence of antiferromagnetism, double- and triple-Q structures, incommensurate and helical magnetic structures and the measurement of magnetic transition temperatures are feasible with elastic neutron scattering.

2.3 Polarized elastic scattering

While more complicated and lengthy in execution, polarized neutron diffraction is often essential. Separation of magnetic from nuclear scattering is useful for the measurement of ferromagnets and single-Q structures. Polarized neutron measurements of Bragg peaks determine magnetic form factors, being the Fourier transform of the unpaired electron density in a unit cell. Sometimes measurements with unpolarized neutrons will not be able to distinguish between models for magnetic structures. Moment directional components can be measured with polarized neutrons, which can determine the best magnetic model. A very important application of polarized neutron diffraction is for the measurement of short-range order.

The second term in eq. (8) is depends on the Fourier transform of a short-range order correlation function, $z(\mathbf{r})$. Knowing the behaviour of magnetic short-range order is vital for the understanding of amorphous magnetism, spin-glasses, and magnetic frustration. Scattering from short-range order will be broad and diffuse, between the Bragg peaks, and a mixture of magnetic and nuclear scattering, often in roughly equal magnitudes. It is necessary to use neutron polarization analysis to separate the magnetic from the nuclear scattering.

2.4 Inelastic scattering

Neutron scattering is currently the only technique capable of measuring magnetic dynamics of all frequencies and momenta in a sample. If the sample has only one type of magnetic atom, eq. (6) can be rewritten:

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\cdot\mathrm{d}E} = \frac{k'}{k} N f^2(Q)(\gamma r_0 \mu_\perp)^2 S(\mathbf{Q},\omega), \tag{9}$$

where $\mu_\perp$ is the component, perpendicular to **Q**, of the moment per atom, $f(Q)$ is the magnetic form factor, and $S(\mathbf{Q},\omega)$ is the Fourier transform of the magnetic positions and vibrations in the sample and is known as the *scattering function*. Measurement of $S(\mathbf{Q},\omega)$ covers the physics of the magnetic properties of the sample, whether it be due to magnons (quantized, dispersive excitations), crystal fields (quantized, non-dispersive), cluster dynamics or amorphous structures (frequently overdamped, non-quantized), impurities or paramagnetism (quasielastic). Neutron inelastic scattering is also crucial for the study of magnetic phase transitions, with measurements of critical phenomena requiring either the measurement of soft modes, or with integration over all inelastic scattering.

2.5 Polarized inelastic scattering

While magnetic elastic scattering, and in particular Bragg diffraction, is often well-localised and relatively easy to separate without the need for polarized neutrons, the scattering function, $S(\mathbf{Q},\omega)$, generally represents surfaces or diffuse features in the $(\mathbf{Q},\omega)$ landscape. Magnetic excitations often mix and interact with nuclear excitations, giving rise to macroscopic features like magnetostriction, superconductivity, and the INVAR effect. Measurements with polarized neutrons are frequently used to separate magnetic from nuclear scattering.

3. Dynamical scattering

The first Born approximation will fail if the interaction between the neutron and the sample cannot be described by first-order perturbation theory, or if the neutron cannot be described by a plane wave function. Both cases hold when scattering from a surface with very small scattering angles, where the phenomena of refraction and total reflection may be observed. Neutrons will still interact with the magnetic properties of the sample, however the interaction now becomes dependent on the wavelength and the energy of the neutron. A more advanced theory, known as a *dynamical* theory, is now needed to describe the observed scattering.

Dynamical scattering for neutrons has, to date, been limited to elastic scattering. It assumes that the incident wave function is still well described by plane waves, and uses Fresnel theory to calculate the changes in the wave functions at interfaces. Fresnel theory requires an expression for the refractive index of a material, n, which, for neutrons with wavelength λ, is given by the equation:

$$n = 1 - \frac{N\lambda^2}{2\pi}\langle \hat{V} \rangle - i\frac{N\lambda}{4\pi}\sigma_a(\lambda). \tag{10}$$

The absorption cross-section, $\sigma_a(\lambda)$, is normally very small and is often neglected.

3.1 Reflectivity

If the angle that the incident beam subtends to the mean surface is equal to the angle of the scattered beam, the ratio of the scattered to the incident intensity is called the *reflectivity*. The reflectivity can be modelled by matching boundary conditions for refracted and reflected waves at interfaces, and the measurement gives the mean scattering length density, $N\langle \hat{V} \rangle$, as a function of depth beneath the mean surface. Neutron reflectivity is popular in the field of magnetic thin films and multilayers, particularly when combined with polarization. It has been used to measure the size and distribution of magnetization as a function of depth as well as more exotic phenomena such as magnetic proximity effects, percolating spin-glasses, 'dead' magnetic layers, and vortices in superconductors.

3.2 Off-specular scattering

Measurement of the off-specular scattering, where the incident angle differs to the scattered angle, can give information on in-plane magnetic correlations from planar samples. Studies of magnetic roughness at interfaces, domain structures, and correlations between elements on patterned samples are all possible. The demand for these measurements is rapidly increasing as they are of great interest to physicists and materials scientists alike, being greatly applicable in the fields of nanomagnetism and information storage. Analysis of off-specular scattering is tremendously difficult, however, particularly when combined with neutron polarization analysis. The theory to describe these types of measurements is still in development.

4. Solving the Phase Problem

Most neutron scattering experiments measure the neutron intensity. As such, they measure a probability amplitude, while the probability amplitude *and* phase are necessary for an unambiguous solution to the sample wave function. Conventional scattering experiments must therefore rely on fitting a model to the data. It is possible, however, to measure the phase information using polarized neutrons. The neutron magnetic moment may rotate on interaction with the sample. Eq. (7) gives the projection of this rotation on to one axis, however the final spin orientation may be anywhere in 4π steradians. If the neutron polarization is measured, rather than the neutron intensity, a unique solution for a magnetic structure may be found. The theory for this is well developed within the Born approximation and experimental methods exist for carrying out these measurements on complex antiferromagnets.

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