INTERACTION OF THERMAL NEUTRONS WITH MATTER

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Introduction

Thermal neutrons are a powerful tool for investigating a variety of properties of condensed matter. Their usefulness arises from the lack of charge on the neutron, the value of its mass, and the fact that it has a magnetic moment. Neutrons at room temperature have a wavelength of about 2 Å, which is of the order of interatomic distances in solids and liquids, and an energy of about 25 meV, which is of the same order as the energy of many excitations in condensed matter. Thus the scattering of thermal neutrons yields information on the positions and motions of the particles in the scattering system.

Neutrons interact with matter in two ways. The first is via the nuclear force between the neutrons and the nuclei in the scattering system. The second is via the interaction between the magnetic moment of the neutron and the magnetic field of unpaired electrons in the atom. Thus the nuclear interaction is always present. The magnetic interaction occurs additionally when the scattering atoms are magnetic, and provides information on the magnetic properties – both static and dynamic – of the scattering system.

Nuclear scattering

We consider first scattering by a single nucleus fixed at the origin. The wavelength of thermal neutrons is very much larger than the range of nuclear forces. The scattering is therefore spherically symmetric, and the wavefunction for the incident and scattered waves may be written in the form

$$\psi(\vec{r}) = e^{ikz} - \frac{b}{r} e^{ikr}. \tag{1}$$

The quantity b is known as the <u>scattering length</u> and represents the strength of the scattering from the particular nucleus.

We may distinguish two types of nuclei. In the first type the scattering length is complex and varies rapidly with the energy of the neutron. The scattering is associated with the formation of a compound nucleus with energy close to an excited state. Examples of such nuclei are ^{113}Cd and ^{157}Gd. The imaginary part of the scattering length corresponds to absorption, and such nuclei are highly absorbing.

In the second type of nuclei, which comprises the great majority, the compound

nucleus is not formed near an excited state. The scattering length is almost entirely real and is independent of neutron energy. We shall confine the subsequent discussion to such nuclei.

The value of the scattering length depends on the nucleus (i.e. its Z, N values) and also on the spin value of the nucleus-neutron system. If the nucleus has spin $I(\neq 0)$ it can form two states with the nucleus with spins $I \pm \frac{1}{2}$. Each state has its own scattering length denoted by b^+ and b^-. For a nucleus with $I = 0$ there is only one scattering length. As we do not have a proper theory of nuclear *forces* for calculating the scattering lengths, we have to treat them as parameters to be determined experimentally. Several methods are available, e.g. the gravity refractometer, giving values accurate to about 1 in 10^3. Unlike their counterparts in X-ray scattering which vary smoothly with the atomic number Z, neutron scattering lengths vary erratically from one nucleus to its neighbour. A selection of values is given in Table 1.

Table 1   Values of scattering length b (in units of 10^{-15} m)

Nuclide:	^{1}H		^{2}H		^{23}Na		^{59}Co	
T:	1	0	3/2	1/2	2	1	4	3
b:	10.8	−47.5	9.5	1.0	6.3	−0.9	−2.8	9.9

T is the spin of the nucleus-neutron system.

If we calculate $\psi(\vec{r})$ using the Born approximation, we obtain the result

$$\psi(\vec{r}) = e^{ikz} + \frac{f(\theta)}{r} e^{ikr},$$ (2)

where

$$f(\theta) = -\frac{m}{2\pi\hbar^2} \int V(\vec{r}) e^{i\vec{q}\cdot\vec{r}} d^3r .$$ (3)

$V(\vec{r})$ is the potential of the nucleus-neutron force, and m is the mass of the neutron. This result is consistent with (1) provided

$$V(\vec{r}) = \frac{2\pi\hbar^2}{m} b \, \delta(\vec{r}) .$$ (4)

Only this δ-function form of $V(\vec{r})$, known as the Fermi pseudopotential, gives the required result of isotropic scattering in the Born approximation.

For a general system of particles the scattering is obtained by adding the waves scattered by each nucleus with due regard to phase. If the nuclei were fixed in position the cross-section would be

$$\frac{d\sigma}{d\Omega} = \left| \sum_j b_j \exp(i\vec{q}\cdot\vec{R}_j) \right|^2 = \sum_{jj'} b_j b_{j'} \exp\left\{ i\vec{q}\cdot(\vec{R}_j - \vec{R}_{j'}) \right\},$$ (5)

where b_j is the scattering length, and $\vec{R}_j$ the position of the j th nucleus. The scattering vector $\vec{q}$ is defined by $\vec{q} = \vec{k}_o - \vec{k}$, where $\vec{k}_o$ and $\vec{k}$ are the wavevectors of the

incident and scattered neutrons. For this scattering system, only elastic scatter-
ing can occur, i.e. $k_o = k$.

In practice the nuclei are not fixed in position. In liquids they move around,
and in solids they oscillate about their equilibrium positions. Inelastic scatter-
ing is therefore possible. We put $\hbar\omega = E_o - E$, where $E_o (= \hbar^2 k_o^2/2m)$ and $E(= \hbar^2 k^2/2m)$
are the initial and final energies of the neutron. The cross-section has the form

$$\frac{d^2\sigma}{d\Omega\,dE'} = \frac{k}{k_o}\frac{1}{2\pi\hbar}\sum_{jj'} b_j b_{j'} \int_{-\infty}^{\infty} dt\, \exp(-i\omega t)$$

$$\times \left\langle \exp\left\{-i\vec{q}\cdot\vec{R}_{j'}(0)\right\} \exp\left\{i\vec{q}\cdot\vec{R}_j(t)\right\}\right\rangle. \tag{6}$$

In this expression $\vec{R}_j(t)$ is the Heisenberg operator for the position of the j th
nucleus, i.e.

$$\vec{R}_j(t) = \exp(iHt/\hbar)\,\vec{R}_j\,\exp(-iHt/\hbar), \tag{7}$$

where H is the Hamiltonian of the scattering system. The symbol < > denotes the
thermal average of the operator enclosed, at the temperature of the scattering system.
Classically the quantity $\vec{R}_j(t)$ may be regarded as the position of the j th nucleus
at time t.

Coherent and incoherent scattering

Consider a scattering system consisting of a single element where the scattering
length b varies from one nucleus to another owing to nuclear spin or the presence of
isotopes or both. We rewrite (6) as

$$\frac{d^2\sigma}{d\Omega\,dE'} = \sum_{jj'} b_j b_{j'} \left\langle j, j'\right\rangle, \tag{8}$$

where <j,j'> stands for the entire j,j' term on the right-hand side of (6) apart
from the two scattering lengths. We assume there is no correlation between the
value of the scattering length and nuclear site, and average (8) over all distribu-
tions of the scattering lengths among the sites. This gives

$$\frac{d^2\sigma}{d\Omega\,dE'} = (\bar{b})^2 \sum_{\substack{jj' \\ j \neq j'}} \left\langle j, j'\right\rangle + \overline{b^2} \sum_j \left\langle j, j\right\rangle$$

$$= (\bar{b})^2 \sum_{jj'} \left\langle j, j'\right\rangle + \left\{\overline{b^2} - (\bar{b})^2\right\} \sum_j \left\langle j, j\right\rangle. \tag{9}$$

The first term in this expression is the <u>coherent</u> cross-section, and the second is
the <u>incoherent</u> cross-section. Coherent scattering depends on the relative positions
of all the nuclei and therefore contains interference terms. It corresponds physic-
ally to the scattering that would occur from a hypothetical system containing the
same atomic positions and motions, but with all the scattering lengths equal to the

mean of the actual scattering lengths. Incoherent scattering does not show inter-
ference and is the scattering due to the random variation of the scattering lengths
about their mean value. If all the scattering lengths are equal, then $\overline{b^2} = (\bar{b})^2$,
and the incoherent scattering is zero.

For a single isotope with spin I, the scattering lengths b^+ and b^- occur with
relative weights $I + 1$ and I. For several isotopes, each scattering length is fur-
ther weighted by the relative abundance of the isotope. These factors determine
the quantities $(\bar{b})^2$ and $\overline{b^2} - (\bar{b})^2$. Numerical values are usually quoted in terms
of

$$\sigma_{coh} = 4\pi \, (\bar{b})^2 \qquad \text{and} \qquad \sigma_{inc} = 4\pi \left\{ \overline{b^2} - (\bar{b})^2 \right\} . \tag{10}$$

Some representative values are given in Table 2. The extension of the theory to
systems containing more than one element is readily made.

Table 2   Values of σ_{coh} and σ_{inc} (in units of 10^{-28} m^2)

Element or nuclide:	^{1}H	^{2}H	C	O	Mg	Al	V	Fe	Cu	Zn
σ_{coh}:	1.8	5.6	5.6	4.2	3.6	1.5	0.02	11.5	7.5	4.1
σ_{inc}:	80.2	2.0	0.0	0.0	0.1	0.0	5.0	0.4	0.5	0.1

Correlation functions

From (6) and (9) the coherent cross-section may be written as

$$\frac{d^2\sigma}{d\Omega \, dE'} = \frac{\sigma_{coh}}{4\pi} \frac{k}{k_o} N \, S(\vec{q}, \omega), \tag{11}$$

where

$$S(\vec{q}, \omega) = \frac{1}{2\pi \hbar} \int G(\vec{r}, t) \exp\left\{ i(\vec{q} \cdot \vec{r} - \omega t) \right\} d^3r \, dt , \tag{12}$$

$$G(\vec{r}, t) = \frac{1}{(2\pi)^3 N} \int \sum_{jj'} \left\langle \exp\left\{-i\vec{q} \cdot \vec{R}_{j'}(0)\right\} \exp\left\{ i\vec{q} \cdot \vec{R}_j(t) \right\} \right\rangle \exp(-i\vec{q} \cdot \vec{r}) d^3q . \tag{13}$$

N is the number of particles in the scattering system. Thus the cross-section is
proportional to the scattering function $S(\vec{q}, \omega)$, which is the Fourier transform in
space and time of the time-dependent pair-correlation function $G(\vec{r}, t)$. The quan-
tity $G(\vec{r}, t) \, d^3r$ may be interpreted classically as the probability that, given a
particle at the origin at time $t = 0$, any particle (including the origin particle)
is in the element of volume d^3r at position $\vec{r}$ at time t.

The relations (11) to (13) are an extension of the result for Fraunhofer
diffraction of light that the amplitude of the scattered wave is the Fourier trans-

form in space of the density function of the scattering system. In the case of light (and X-rays) the radiation travels so fast that the particles in the scattering system do not have time to move in the interval between the incident radiation reaching neighbouring atoms in the system. The radiation 'sees' the particles frozen in some configuration, and the scattering depends only on the value of $G(\vec{r},t)$ at $t = 0$. Thermal neutrons, however, have velocities of the order of 10^3 m s^{-1}. The time for the neutron to travel between particles separated by distances of about 10^{-10}m is $\sim 10^{-13}$ s. This is of the order of the period of oscillation for atoms in solids and liquids. Thermal neutron scattering therefore gives information on $G(\vec{r},t)$ at general values of t, which is one of the reasons neutrons are so useful for investigating the properties of condensed matter.

Magnetic scattering

We confine the discussion to the scattering that arises from the interaction between the magnetic moment of the neutron and the magnetic field due to the spins of the electrons, i.e. we ignore orbital effects. For a single electron at the origin the field $\vec{B}$ at position $\vec{R}$ is

$$\vec{B} = \operatorname{curl} \vec{A} , \qquad \vec{A} = \frac{\mu_0}{4\pi} \frac{\vec{\mu}_e \times \vec{R}}{R^3} , \qquad \vec{\mu}_e = -2\mu_B \vec{s} , \qquad (14)$$

where $\vec{\mu}_e$ is the magnetic moment of the electron, $\vec{s}$ its spin angular momentum in units of $\hbar$, and μ_B is the Bohr magneton.

The magnetic moment of the neutron may be expressed as

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma} , \qquad (15)$$

where μ_N is the nuclear magneton, $\vec{\sigma}$ the Pauli spin operator of the neutron, and $\gamma = 1.91$. The magnetic potential is

$$V_{mag} = -\vec{\mu}_n \cdot \vec{B} . \qquad (16)$$

Inserting this in the Born approximation expression (3) we obtain for the magnetic counterpart of the scattering length

$$b_{mag} = -\gamma r_e \vec{\sigma} \cdot \vec{Q}_\perp , \qquad (17)$$

where

$$\vec{Q}_\perp = \hat{q} \times (\vec{s} \times \hat{q}) , \qquad (18)$$

and $r_e = \mu_0 e^2/4\pi m_e$ is the classical radius of the electron. $\hat{q}$ is a unit vector in the direction of $\vec{q}$.

It can be seen that $\vec{Q}_\perp$ is the vector projection of $\vec{s}$ on to the plane perpendicular to $\vec{q}$. Thus b_{mag} is zero when $\vec{q} \parallel \vec{s}$. This result may be seen on physical grounds. When $\vec{q} \parallel \vec{s}$, the phase difference between the waves scattered by two atoms in a plane perpendicular to $\vec{s}$ is zero (Fig. 1). The contribution to the integral

$\int \vec{\mu}_n . \vec{B} \exp(i\vec{q}.\vec{r})d^3r$ over a plane perpendicular to $\vec{s}$ is therefore proportional to $\int \vec{B}$ dS, where dS is an element of area in the plane, and the integral is taken over the whole plane. Now the contribution to the integral from the component of B perpendicular to the plane is zero, because div $\vec{B} = 0$, and the contribution from the component of $\vec{B}$ in the plane is likewise zero, from the symmetry of the field due to a dipole.
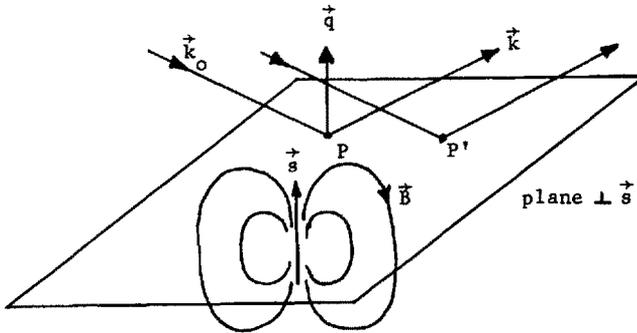

Fig. 1   Diagram for the case $\vec{q} \parallel \vec{s}$. The phase difference between the waves scattered at P and P' is zero.

It can be seen that the expression for b_{mag} is more complicated than its nuclear counterpart. The nuclear force is short-range and central. Thus the nuclear scattering length, which is proportional to the Fourier transform of the potential, is a constant, independent of $\vec{q}$. The magnetic force is long-range and non-central. Thus the magnetic scattering length depends on $\vec{q}$, and has the geometrical dependence given in (17) and (18).

For a single atom with L-S coupling the spins of the electrons combine to form a resultant spin angular momentum $\vec{S}$ for the atom, and b_{mag} becomes

$$ b_{mag} = -\gamma r_e \vec{\sigma} . \left\{ \hat{q} \times (\vec{S} \times \hat{q}) \right\} F(\vec{q}) . \tag{19} $$

The magnetic form factor $F(\vec{q})$, which arises from the phase difference between the waves scattered by the different electrons in the atom, is the Fourier transform of $s(\vec{r})$, the normalised spin density of the unpaired electrons, i.e.

$$F(\vec{q}) = \int s(\vec{r}) \exp(i\vec{q}.\vec{r}) \, d^3r. \tag{20}$$

For a scattering system of many atoms we add the amplitudes of the scattered waves from each atom just as in the nuclear case. Coherent scattering occurs in systems with magnetic order, e.g. ferromagnets and antiferromagnets, and incoherent scattering occurs in paramagnets.

Elastic and inelastic scattering

For crystalline systems the scattering may be divided into elastic and inelastic processes. Coherent elastic scattering, both nuclear and magnetic, is the exact analogue of Bragg scattering of X-rays. For nuclear scattering and magnetic scattering from ferromagnets, the Bragg peaks occur at the same reciprocal lattice points as for X-rays. But for antiferromagnets and helimagnets the peaks are, in general, at other points in reciprocal space.

Inelastic scattering involves exchange of energy between the neutron and excitations such as phonons and magnons in the scattering system. All the scattering apart from coherent elastic scattering is sometimes referred to as thermal diffuse scattering, as it does not give peaks at points in reciprocal space. The term derives from X-ray scattering where it is used to denote the inelastic (phonon) scattering. This type of scattering produces a negligible fractional change in the energy of the X-ray quantum (in sharp contrast to the neutron case), but a significant change in the momentum, and thus gives rise to diffuse spots and streaks on X-ray photographs.

Applications

Coherent elastic scattering of thermal neutrons gives information on the location of atoms (neutrons are particularly useful for locating hydrogen) on the degree of structural order (e.g. in binary alloys), and on magnetic structures. Inelastic scattering gives information on phonon and magnon frequencies and lifetimes, time-dependent pair-correlation functions, and spin-correlation functions. Considerable work has been done in a number of fields, such as liquids, polymers, liquid crystals, molecular crystals, diffusion in solids, critical phenomena, and so on.

Additional information may be obtained by polarising the neutrons and analysing their spin states after scattering. Interference between nuclear and magnetic scattering provides a method for polarising neutrons and is also used to enhance the accuracy of some measurements, for example the determination of spin densities.

R E F E R E N C E S

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