Neutrons for magnetism

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Neutrons

• Neutron discovered by James Chadwick in 1932
• Used in magnetic neutron diffraction in 1949 (Clifford Shull)
• Neutron mass close to proton mass
• Neutron decays in ~15 minutes
• Spin \( \frac{1}{2} \)
• Magnetic moment \( 1.9 \mu_N \approx 0.001 \mu_B \)

Neutrons

• Neutron has no electrical charge
• Interaction is with atomic nuclei (strong force, short range) and unpaired electrons (EM)
• Neutron-matter interaction is weak: perturbation theory adequate (very roughly, probability of interaction in a solid \(-10^3\), mean free path \(-1\) cm)
• Neutron interaction with nuclei and electrons similar in magnitude
• Scattering cross section with proton is huge: 82 barns. For deuterons, it is an order of magnitude smaller, \([1 \text{ barn} = 10^{-28} \text{m}^2]\)

Neutrons

• Energy given by \( E = \frac{h^2 k^2}{2m} \)
• Energies are often given in meV = \(10^{-3} \text{ eV}\)
• Wavelength \( \lambda \) given in Angstroms
• Useful results: \( \frac{h^2}{2m} = 2.08 \text{ meV A}^{-2} \)
  \[ \lambda = \left( \frac{h^2}{2mE} \right)^{1/2} = \frac{9.04}{\sqrt{E}} \]
  \(1 \text{ meV} \approx 0.24 \text{ THz} \approx 8.07 \text{ cm}^{-1} \approx 11.61 \text{ K}^{-1}\)

Neutrons

• Measure distribution of neutrons scattered from sample
• Interaction potential determines properties measured
• Scattering must be coherent for correlations to be measured
• Scattering of neutrons is very weak and so can use the Born approximation
• Means scattering depends on the Fourier transform of the interaction potential, and system responds linearly

Moderator

\[
n(v) \propto v^3 \exp \left( -\frac{1}{2} \frac{m_n v^2}{k_B T} \right) \quad v_{\text{max}} = \sqrt{\frac{3k_B T}{m_n}}
\]
Neutrons

- The Born approximation assumes coherence and superposition
- Detected amplitude = $\phi_1 + \phi_2 + \cdots$
- Detected intensity = $|\phi_1 + \phi_2 + \cdots|^2$
  
  $$= |\phi_1|^2 + |\phi_2|^2 + \phi_1^*\phi_2 + \phi_2^*\phi_1 + \cdots$$

  depends on relative positions of atoms 1 and 2

- Measurement non-destructive (though they might activate the sample!)
- Bulk, not surface probe
- Samples have to be big (~1 mm$^3$ for diffraction experiments, ~1 cm$^3$ for inelastic measurements)
- Technique expensive – need a nuclear reactor with holes in it (!) or a spallation source. These are not cheap. Fortunately, Europe has a number of excellent sources of neutrons.

Kinematics

- Conservation of energy
  $$\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$
- Conservation of momentum
  $$Q = k_i - k_f$$
- Scattering event characterized by $(Q, \omega)$
- (i) Elastic scattering
  $$\hbar\omega = 0$$
- (ii) Inelastic scattering
  $$\hbar\omega \neq 0$$
- Both processes occur in every experiment
- Can set $Q$ and $\omega$ independently in the experiment

Elastic neutron scattering

- Conservation of energy
  $$\hbar\omega = E_i - E_f = 0$$
- $Q = k_i - k_f$

Elastic neutron scattering

- Conservation of energy
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Nuclear scattering

- Scalar potential, short-range: \( V_N = \frac{2\pi \hbar^2}{m_n} b_0(r) \)
- Scattering length \( b \sim 10^{-14} \text{ m} \)
- Scattering function
  \[ S(Q) = | \sum_j b_j \exp(i Q \cdot r_j) |^2 \]
  - Rigid crystal
  \[ S(Q) = N \left( \frac{2\pi)^3}{V_0} \sum_G |F(G)|^2 \delta(Q - G) \]
  - \( N = \) number of unit cells
  - \( V_0 = \) volume of unit cell

Coherent and incoherent

- \( b \) varies with isotope and spin orientation
- separate \( b \) into an average value \( \bar{b} \) and
- coherent scattering results from \( b \) and gives rise to diffraction peaks
- incoherent scattering results from \( \bar{b} \) and gives rise to an incoherent background
  \[ S_{\text{inc}}(Q) = \sum_j (\sigma_{\text{inc}})_j \]

Magnetic neutron diffraction

- Magnetic interaction potential energy
  \[ V_M = -\mu_n \cdot B(r) \]
- depends on spin and orbital currents
- depends on direction of neutron spin
- vector, not scalar, interaction
- anisotropic scattering
- derives from electronic states and so the magnetic form factor is important

Magnetic neutron diffraction

- Magnetic interaction potential in Q-space
  \[ V_M(Q) = -\mu_n \cdot B(Q) \]
- Maxwell: \( \nabla \cdot B = 0 \implies i Q \cdot B(Q) = 0 \)
- \( B(Q) \) is therefore perpendicular to \( Q \)
- Neutrons scatter from \( m_l \), the component of the magnetic moment perpendicular to \( Q \)
Magnetic neutron diffraction

Scattering cross section

Given the symbol \( \sigma \)

- Neutrons scattered into various final energies and also various bits of space (solid angle), so deal with the double differential cross section:

\[
\frac{d^2\sigma}{dE_f d\Omega}
\]

- This sums up to the total cross section

\[
\sigma = \int \int dE_f d\Omega \frac{d^2\sigma}{dE_f d\Omega}
\]

Example

\( \text{para-nitro} \)-phenyl

nitronyl nitroxide

An organic ferromagnet with \( T_c = 0.67 \) K

Example

Crystal structure strongly affects magnetic coupling.

Only the \( \beta \) -phase of \( p \)-NPNN is ferromagnetic.

One can use the polarized neutron diffraction maps to understand the important overlaps between regions of positive and negative spin density.

Polarized neutron diffraction

The scattered intensity \( I(k) \) for scattering vector \( k \) is given by

\[
I(k) = |F_0(k)|^2 + P \cdot |F_0(k)|^2 F_0(k) + F_0(k) F_0(k) + |F_0(k)|^2
\]

where \( F_0(k) = \sum \Delta_{\alpha}(h,k) \cdot \psi_i \) is the nuclear structure factor, \( F_0(k) \) is the projection of the magnetic structure factor \( F_0(k) = \sum M_j e^{ik \cdot r_j} \) onto the plane perpendicular to \( k \), \( P \) is the polarization of the beam, \( \psi_i \) is a Debye-Waller factor for the \( i \) th atom in the unit cell at position \( r_i \) with scattering length \( b_i \).

Measure \( I(k) \) for neutron polarization parallel or antiparallel with the magnetic field, and the sign of the interference term can be varied, allowing the magnetic structure factor to be deduced.

Example

spin density of \( p \)-NPNN

Zheludev et al
JMMM 135 147 (1994)
Inelastic neutron scattering

Scattering cross section

\[ \frac{d^2 \sigma}{dE_{\text{f}} d\Omega} = \frac{\text{Number scattered } \pi^{-1}}{I_0 d\Omega dE_f} \]

- Numerator contains (i) \( dE_f \) (ii) \( d\Omega \) (iii) speed of scattered neutrons \( v_f = \frac{\hbar k_f}{m} \) (iv) density of incident neutrons \( \rho_i \) (v) transition probability in which the sample changes its momentum by \( Q \) and its energy by \( \omega \), i.e. \( S(Q, \omega) \)
- Denominator contains (i) \( I_0 = \rho_i v_i = \rho_i \hbar k_i / m \) and (ii) \( dE_f \) (iii) \( d\Omega \)

\[ \Rightarrow \frac{d^2 \sigma}{dE_{\text{f}} d\Omega} = \frac{k_f}{k_i} S(Q, \omega) \]

Detailed balance

\[ S(Q, -\omega) = \exp\left(-\frac{\hbar \omega}{k_B T}\right) S(Q, \omega) \]

Calculation of \( S(Q, \omega) \)

\[ S(Q, \omega) = \sum_i P_i \sum_j \left|M_{ij}\right|^2 \delta(\hbar \omega - E_j + E_i) \]

- Can look at local magnetic excitations, e.g. crystal field level spectroscopy

Calculation of \( S(Q, \omega) \)

\[ S(Q, \omega) = \frac{1}{\pi} \chi''(Q, \omega) \]

\[ M_\alpha(Q, \omega) = \chi_{\alpha\beta}(Q, \omega) H_\beta(Q, \omega) \]

\[ \chi(Q, \omega) = \chi'(Q, \omega) + i\chi''(Q, \omega) \]
Partially filled 3d shell gives rise to a magnetic moment.

\[ \text{Cu}^{II} = 3d^9 \]

\[ \text{KCuF}_3 \]

\[ \text{La}_2\text{CuO}_4 \]

\[ \text{CoNb}_2\text{O}_6 \]

\[ \text{CoNb}_2\text{O}_6 \]
Magnetic moment $\mu$ in a field $B$

$$E = -\mu \cdot B$$

$$\mu = \gamma L$$

Spin precession classical

$$\frac{d\mu}{dt} = \gamma \mu \times B$$

$$\dot{\mu}_x = \gamma B\mu_y$$

$$\dot{\mu}_y = -\gamma B\mu_x$$

$$\dot{\mu}_z = 0,$$

$$\mu_x(t) = |\mu| \sin \theta \cos (\gamma B t)$$

$$\mu_y(t) = |\mu| \sin \theta \sin (\gamma B t)$$

$$\mu_z(t) = |\mu| \cos \theta.$$

Spin precession quantum

$$\hat{H} = g\mu_B B \cdot \hat{S} = g\mu_B B \hat{S}_z.$$

$$[\hat{S}_x, \hat{S}_y] = i\hat{S}_z,$$ or equivalently: $$[\hat{S} \cdot X, \hat{S}] = i\hat{S} \times X.$$

$$\frac{d}{dt} \langle \hat{S} \rangle = \frac{1}{i\hbar} \langle \hat{S}, \hat{H} \rangle = -\frac{g\mu_B}{\hbar} \langle \hat{S} \rangle \times B.$$

$$\gamma = -\frac{g\mu_B}{\hbar} = -\frac{e}{2m_e}.$$
Polarized neutron reflection

\[ V_{\alpha} = \frac{\hbar^2}{2\pi m} p_{\alpha} B_{\alpha} - \mu_{\alpha} B_{\alpha} \]


\[ M = D^{-1}(q_\perp) \prod_{j=1}^{N-1} \left[ D(q_j) D(q_j', q_j, q_j') \right] D(q_N) \]

where

\[ D(q_j) = \begin{bmatrix} D(q_j') & 0 \\ 0 & D(q_j) \end{bmatrix} \]

is the 4x4 generalization of the transmission matrices,

\[ D(q_j', q_j, q_j') = \begin{bmatrix} D(q_j') & 0 \\ 0 & D(q_j', q_j') \end{bmatrix} \]

is the 4x4 generalization of the propagation matrices, and

\[ R(\theta_{j+1}) = \begin{bmatrix} \cos(\theta_{j+1}/2) & \sin(\theta_{j+1}/2) \\ -\sin(\theta_{j+1}/2) & \cos(\theta_{j+1}/2) \end{bmatrix} \]

are the matrices for rotating the quantization axis by \( \theta_{j+1} \) at the \( j+1 \) interface.