Magnetic Neutron Scattering

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- Magnetism and Neutron Scattering – A Killer Application
- Magnetism in Solids
- Bottom lines on magnetic neutron scattering
- Examples
C. G. Shull et al, 1951  

Magnetic Structure of MnO

Various diagrams illustrating magnetic structures and measurements at different temperatures.

- **Paramagnet** $T > T_C$
- **Ferromagnet** $T < T_C$
- **Antiferromagnet** $T < T_N$
Magnetic Neutron Scattering directly probes the electrons in solids

**Killer Application:** Most powerful probe of magnetism in solids!
**Magnetism = Net Angular Momentum**

**TABLE 1 Nuclear Magnetic Resonance Data**

For every element the most abundant magnetic isotope is shown. After Varian Associates NMR Table, 4th ed., 1964.

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</table>

- **d-electrons: 10 levels to fill**
- **4f: 14 levels**
- **5f**
$\text{eg orbitals}$

$\text{t}_{2g}$ orbitals
$e_g$ orbitals

$3d^5 : Mn^{2+}$

$t_{2g}$ orbitals
$e_g$ orbitals

$3z^2 - r^2$

$x^2 - y^2$

$t_{2g}$ orbitals

$zx$

$yz$

$xy$

$3d^9 : Cu^{2+}$

- Red arrows for electron occupation
- Blue arrows for electron occupation
Superexchange Interactions in Magnetic Insulators

\[ H = J \sum_{i,j} S_i \cdot S_j \]

RKKY exchange in Itinerant Magnets (eg. Rare Earth Metals)
$T = 0.9 \, T_C$

$T = T_C$

$\vec{k} = (0, 0, \frac{1}{2})$

$T = 1.1 \, T_C$

$\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$
Magnetic Neutron Scattering

Neutrons carry no charge; carry $s=1/2$ magnetic moment

Only couple to electrons in solids via magnetic interactions

$$\mu_n = -\gamma \mu_N \sigma \quad \gamma = 1.913 \quad \text{nuclear magneton} = e \frac{\hbar}{2m_n} \quad \text{Pauli spin operator}$$

How do we understand what occurs when a beam of mono-energetic neutrons falls incident on a magnetic material?
Calculate a “cross section”:

What fraction of the neutrons scatter off the sample with a particular:

a) Change in momentum: \( \kappa = k - k' \)

b) Change in energy: \( \hbar \omega = \hbar^2 k^2 / 2m - \hbar^2 k'^2 / 2m \)

- Fermi’s Golden Rule
- 1\textsuperscript{st} Order Perturbation Theory

\[
\frac{d^2 \sigma}{d\Omega \ dE} : k, \sigma, \lambda \rightarrow k', \sigma', \lambda'
\]

\[= \frac{k'}{k} \left( \frac{m}{2\pi \hbar^2} \right)^2 |< k' \sigma' \lambda' | V_M | k \sigma \lambda >|^2 \delta (E_\lambda - E_\lambda' + \hbar \omega)\]

- kinematic
- interaction matrix element
- energy conservation
Understanding this means understanding:

$V_M$: The potential between the neutron and all the unpaired electrons in the material

$V_M = -\mu_n B$

Magnetic Field from spin ½ of Electron: $B_S$

Magnetic Field from Orbital Motion of Electrons: $B_L$
The evaluation of \( | <k' \sigma' \lambda' | V_M | k \sigma \lambda> |^2 \) is somewhat complicated, and I will simply jump to the result:

\[
d^2\sigma/d\Omega \ dE' = (\gamma r_0)^2 \ k'/k \ \Sigma_{\alpha \beta} \ (\delta_{\alpha \beta} - \kappa_\alpha \kappa_\beta) \\
\times \ \Sigma \Sigma_{\text{All magnetic atoms at } d \text{ and } d'} \ F_{d'}^*(\kappa)F_d(\kappa) \\
\times \ \Sigma_{\lambda \lambda'} \ p_\lambda < \lambda | \exp(-i\kappa R_{d'})S_{d'}^\alpha | \lambda' >= < \lambda' | \exp(i\kappa R_d)S_d^\beta | \lambda > \\
\times \ \delta (E_\lambda - E_{\lambda'} + \hbar \omega)
\]

With \( \kappa = k - k' \)

This expression can be useful in itself, and explicitly shows the salient features of magnetic neutron scattering.
We often use the properties of \( \delta (E_\lambda - E_\lambda' + \hbar \omega) \) to obtain \( d^2\sigma/d\Omega \ dE' \) in terms of *spin correlation functions*:

\[
d^2\sigma/d\Omega \ dE' = \frac{(\gamma r_0)^2}{(2\pi\hbar)} \frac{k'}{k} N \{1/2 \ g \ F_d(\kappa)\}^2 \\
\times \sum_{\alpha \beta} (\delta_{\alpha \beta} - \kappa_\alpha \kappa_\beta) \sum_l \exp(i\kappa \cdot l) \\
\times \int <\exp(-i\kappa \cdot u_0))\exp(i\kappa \cdot u_1(t))> \\
\times <S_0^\alpha(0) \ S_1^\beta(t)> \ \exp(-i\omega \ t) \ dt
\]

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**Dynamic Spin Pair Correlation Function**

**Fourier transform:** \( S(\kappa, \omega) \)
Comparable in strength to nuclear scattering

\( \{1/2 \, g \, F(\kappa)\}^2 \) : goes like the magnetic form factor squared

\[ \sum_{\alpha \beta} (\delta_{\alpha \beta} - \kappa_\alpha \kappa_\beta) \] : sensitive only to those components of spin \( \perp \kappa \)

Dipole selection rules, goes like:

\[ < \lambda' | S_\beta^d | \lambda > \]

where \( S_\beta^\beta = S^x, S^y \) (\( S^+, S^- \)) or \( S^z \)

**Diffraction type experiments:**

Add up spin correlations with phase set by \( \kappa = k - k' \)

\[ \Sigma_l \, \exp(i\kappa \cdot l) \, <S_0^\alpha(0) \, S_l^\beta(t)> \quad \text{with } t=0 \]
Magnetic form factor, \( F(\kappa) \), is the Fourier transform of the spatial distribution of magnetic electrons –

\( \text{usually falls off monotonically with } \kappa \text{ as } \pi / (1 \text{ A}) \sim 3 \text{ A}^{-1} \)
Three types of scattering experiments are typically performed:

- Elastic scattering
- Energy-integrated scattering
- Inelastic scattering

**Elastic Scattering**

\[ \hbar \omega = (\hbar k)^2/2m - (\hbar k')^2/2m = 0 \]

measures time-independent magnetic structure

\[
\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left\{ \frac{1}{2} g F(\kappa) \right\}^2 \exp(-2W) \\
\times \sum_{\alpha \beta} (\delta_{\alpha \beta} - \kappa_\alpha \kappa_\beta) \Sigma_1 \exp(i\kappa \cdot l) \langle S_0^\alpha \rangle \langle S_1^\beta \rangle
\]

- \( S \perp \kappa \) only
- Add up spins with \( \exp(i\kappa \cdot l) \) phase factor
**URu$_2$Si$_2$**

\[ \mathbf{k} = 0,0,1 \]
\[ a^* = b^* = 0: \]

everything within a basal plane \((a-b)\) adds up in phase

\[ c^* = 1: \]

\(2\pi\) phase shift from top to bottom of unit cell

\(\pi\) phase shift from corners to body-centre –good …..

but \(\mu \parallel \mathbf{k}\) kills off intensity!

Try \(\mathbf{k} = 1,0,0:\)

\(\mu \perp \mathbf{k}\) good!
\( \text{Mn}^{2+} \text{ as an example: } \frac{1}{2} \text{ filled 3d shell } S = \frac{5}{2} \)

\( (2S+1) = 6 \text{ states: } |S(S+1), m_z > \)

\( m_z = +\frac{5}{2} \hbar, +\frac{3}{2} \hbar, +\frac{1}{2} \hbar, -\frac{1}{2} \hbar, -\frac{3}{2} \hbar, -\frac{5}{2} \hbar \)

\[ \text{H}=0; \text{ 6 degenerate states} \]

\[ \text{H} \neq 0; \text{ 6 non-degenerate states} \]

\[ < \frac{3}{2} | S^- | \frac{5}{2} > \neq 0 \rightarrow \text{ inelastic scattering} \]
Magnetic sites are coupled by exchange interactions:

\[ H = J \sum_{i,j} S_i \cdot S_j \]

Spin Wave Eigenstate:

“Defect” is distributed over all possible sites
Inelastic Magnetic Scattering: $|k| \neq |k^0|$

Study magnetic excitations (e.g., spin waves)

Dynamic magnetic moments on time scale $10^{-9}$ to $10^{-12}$ sec

$S(k, \omega) = n(\omega) \chi''(k, \omega)$

Bose (temperature) factor  Imaginary part of the dynamic susceptibility

Figure 9 A spin wave on a line of spins. (a) The spins viewed in perspective. (b) Spins viewed from above, showing one wavelength. The wave is drawn through the ends of the spin vectors.
Sum Rules:

One can understand very general features of the magnetic neutron Scattering experiment on the basis of “sum rules”.

1. $\chi_{DC} = \int (\chi''(\kappa=0, \omega)/\omega) \, d\omega$ ;

   where $\chi_{DC}$ is the $\chi$ measured with a SQUID

2. $\int d\omega \int_{BZ} d\kappa \, S(\kappa, \omega) = S(S+1)$
Symmetry broken

\( T = 0.9 \, T_C \)

\( \xi \sim \text{very large} \)

Origin of universality

\( T = T_C \)

\( T = 1.1 \, T_C \)
\[ M(Q) \]

- Bragg scattering gives square of order parameter; symmetry breaking
- Diffuse scattering gives fluctuations in the order parameter

\[ T < T_C \]

Intensity

\[ M(Q)^2 \]

\[ Q = 2\pi/d \]

\[ \chi(Q_{\text{ord}}) \]

\[ \kappa \]

Diagram showing Bragg and diffuse scattering peaks with temperature below \( T_C \).
CsCoBr₃

Bragg scattering

\[ Q = (2/3, 2/3, 1) \]

\[ I = M^2 = M_0^2 (1 - T/T_C)^{2\beta} \]

Energy-integrated critical scattering

\[
\frac{d\sigma(\vec{Q})}{d\Omega} = \frac{\chi(\vec{Q}_{ord})}{1 + \frac{q_a^2 + q_b^2}{\kappa_{ab}^2} + \frac{q_c^2}{\kappa_c^2}},
\]
Geometrical Frustration:

The cubic pyrochlore structure;
A network of corner-sharing tetrahedra

Low temperature powder neutron diffraction from \( \text{Tb}_2\text{Ti}_2\text{O}_7 \)
A $^{3+}$ site within a distorted cube of 8 $O^{2-}$ ions – unique direction pointing into or out of tetrahedra.

$\text{Tb}^{3+} : S=3, L=3, J=6$

$(2J+1) = 13$ states split by the crystalline electric field.
Inelastic neutron scattering on polycrystalline Tb$_2$Ti$_2$O$_7$

( $\Delta$: Ho$_2$Ti$_2$O$_7$ $\sim$ 240 K ; Dy$_2$Ti$_2$O$_7$ $\sim$ 380 K)
Time-of-flight neutron scattering from DCS on Tb$_2$Ti$_2$O$_7$
One Transition in Zero Field

Five Transitions in Non-Zero Field
Conclusions:

- Neutrons probe magnetism on length scales from 1 – 100 Å, and on time scales from $10^{-9}$ to $10^{-12}$ seconds.

- Magnetic neutron scattering goes like the form factor squared (small $\kappa$), follows dipole selection rules $< \lambda | S^{+,-,z} | \lambda >$, and is sensitive only to components of moments perpendicular to $\kappa$.

- Neutron scattering is the most powerful probe of magnetism in materials; magnetism is a killer application of neutron scattering (1 of 3).
Magnetic Structures can be complicated

Incommensurate structures in rare earth metals

Multiple-\(\vec{k}\) structures in high-symmetry antiferromagnets