Magnetic neutron diffraction

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Magnetic moment-Rare earths

- Progressive filling of 4f levels
  - Strong Hund’s rules
  - Strong spin-orbit interaction
  - Weak CEF

- Unpaired electrons
  - Total angular momentum
    \[ J = L + 2S \]

\[ \mu = g_J \mu_B J \approx g_J J \frac{e\hbar}{2m_e} \]
Transition metals

- **Progressive filling of 3d levels**
  - Strong Hund’s rules interactions
  - Strong CEF
  - Weak spin-orbit interaction
- **Unpaired electrons**
  - Spin moment
  - Orbital moment (quenched)

\[
\mu = g \mu_B S \approx 2S \frac{e \hbar}{2m_e}
\]
Magnetic structures

- Exchange coupling between moments leads to ordering
  - Direct exchange
  - Superexchange (insulators)
  - RKKY (metals)
  - Dipolar
- Magnetic anisotropy leads to moment direction
- Magnetic structures defined by
  - Propagation vector(s)
  - Moment size
  - Moment direction(s)

Elastic scattering - Bragg’s Law

\[ 2ds\sin\theta = n\lambda \]
1-D cartoons

- **nuclear structure**
  atoms separated by lattice spacing \( a \)

- **ferromagnet**
  collinear moments; commensurate

- **simple ferrimagnet**

- **simple antiferromagnet**

- **antiferromagnet with larger unit cell**

- **non-collinear antiferromagnet**

- **incommensurate antiferromagnet**
Neutron magnetism

- Spin-1/2 particle
- Magnetic moment

\[ \mu_n = -\gamma \mu_N = -1.913 \frac{e\hbar}{2m_p} \]

\[ \frac{\mu_n}{\mu_e} \approx \frac{m_e}{m_p} = 1/2000 \]
Dipole interaction

Interaction between neutron and electron

\[ U = -\mu_n \cdot B = \frac{\mu_0 \gamma e^2}{4\pi m_e} \sigma \cdot B = \gamma r_0 \sigma \cdot B \]

\[ U^{uv} = \langle u| b - pS_\perp \cdot \sigma |v \rangle \]

\[ p = \gamma r_0 g S f(Q) \quad S_\perp = \hat{S} - (\hat{S} \cdot \hat{Q})\hat{S} \]

Only moment projection perp. to \( \mathbf{Q} \) will scatter neutrons

\[ U^{++} = b - pS_{\perp z} \]
\[ U^{--} = b + pS_{\perp z} \]
\[ U^{-+} = -p(S_{\perp x} + iS_{\perp y}) \]
\[ U^{+-} = -p(S_{\perp x} - iS_{\perp y}) \]
Magnetic cross-section

\[(\gamma r_0)^2 = 291 \text{ millibarns/steradian}\]

\[b^2(\text{Fe}) = 895 \text{ mb/Sr}\]
Magnetic form factor

\( f(\mathbf{Q}) \): Fourier transform of the atomic magnetization density
Magnetic structure factor is actually a vector quantity, but for collinear structure, can be simplified

\[ F_M(\tau) = \sum_d \frac{1}{2} g_d \langle S_d \rangle \sigma_d F_d(Q) \exp(-W_d) \exp(i \tau \cdot d) \]

Scattering differential cross-section for \emph{unpolarized} beam

\[ \frac{d\sigma}{d\Omega} = N r_0^2 (1 - \hat{\tau}_z^2) \left| F_M(\tau) \right|^2 \]

More generally

\[ \frac{d\sigma}{d\Omega} = N r_0^2 \sum_{\tau} \delta(Q - \tau) \left| \hat{Q} \times \{M(\tau) \times \hat{Q}\} \right|^2 \]
1-D Cartoons

**Configuration in Real Space**

- $a$

**Diffraction Pattern in Reciprocal Space**

- $a^*$

- **nuclear Bragg peaks**

- **magnetic intensities on top of nuclear Bragg peaks**

- **half-indexed magnetic and integer-indexed nuclear and magnetic Bragg peaks**

- **half-indexed magnetic Bragg peaks**

- **quarter-indexed magnetic Bragg peaks**

- **quarter-indexed magnetic Bragg peaks**

- **magnetic satellites**

- **nuclear intensities only**

- **nuclear and magnetic intensities**

- **magnetic intensities only**
Determine magnetic structure

- **Prescription**
  - Measure the magnetic propagation vector(s)
  - Magnetic space group
    - Limits the possible structures
    - You need to know the crystal structure
  - Determine moment direction(s) (refinement)

- **Potential problems**
  - Magnetic domains
  - Crystallographic twinning
  - Multiple wavevectors/multi-q structures
In 1949, Clifford Shull observed additional magnetic reflections in MnO which led to the confirmation of antiferromagnetism.

Shull and J. S. Smart, Phys Rev 76, 1256 (1949).

**Table II.** Comparison between observed MnO antiferromagnetic intensities and those calculated for various models of magnetic orientation with respect to crystallographic axes.

<table>
<thead>
<tr>
<th>Calculated for various oriented models</th>
<th>(a)</th>
<th>(b)</th>
<th>(c)</th>
<th>Observed (neutrons/min)</th>
</tr>
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<tbody>
<tr>
<td>(111)</td>
<td>1038</td>
<td>0</td>
<td>1560</td>
<td>1072</td>
</tr>
<tr>
<td>(311)</td>
<td>460</td>
<td>675</td>
<td>...</td>
<td>308</td>
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<tr>
<td>(331)</td>
<td>129</td>
<td>109</td>
<td>...</td>
<td>132</td>
</tr>
<tr>
<td>(511)</td>
<td>54</td>
<td>24</td>
<td>...</td>
<td>70</td>
</tr>
<tr>
<td>(333)</td>
<td>54</td>
<td>24</td>
<td>...</td>
<td>70</td>
</tr>
</tbody>
</table>
Cone structure of Er

- incommensurate
- Alternating cone structure
- Spin slips from magnetoelastic effect


FIG. 6. Diffraction pattern from the $q=(5/21)c^*$ phase at 0 T and 10 K along the [001] direction. The insert shows the first eight layers of the basal-plane spin-slip model for this structure.
Cone structure of Er
Neutron polarization analysis

- Why use polarization?
  - Separate magnetic/nuclear scatt. (q=0 structures)
  - Refine structure determination (e.g. canting)
  - Separate coherent/incoherent (diffuse scattering, mag. densities)

\[
\begin{align*}
U^{++} &= b - p S_{\perp z} \\
U^{--} &= b + p S_{\perp z} \\
U^{+-} &= -p (S_{\perp x} + iS_{\perp y}) \\
U^{-+} &= -p (S_{\perp x} - iS_{\perp y})
\end{align*}
\]
Instrumentation

Monochromator

Crystal magnetization

\[ U^{++} = b - pS_{\perp z} \]

\[ U^{--} = b + pS_{\perp z} \approx 0 \]

Cu$_2$MnAl (111) (Heusler)

Spin flippers
Spin-flip vs. Non-spin-flip

Useful modes

- \( P \parallel Q \) (in-plane polarization): All magnetic scattering is SF
- \( P \perp Q \) (vertical polarization): magnetic scattering can be SF & NSF
Polarized experiments

Separation of magnetic/nuclear Paramagnetic scattering

Polarization @ pulsed source

Heusler mono won’t work for wide angle scattering

$^3$He polarizers
Further references

- **Magnetic neutron scattering**

- **Structural refinements**
  - GSAS http://www.ncnr.nist.gov/xtal/software/gsas.html
  - FullProf http://www.ill.eu/sites/fullprof/

- **Magnetic space groups**
  - Izyumov, Ozerov, “Neutron diffraction of magnetic materials”
  - Sarah program (representational analysis)