Magnetic neutron diffraction

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September 19, 2018
Magnetic moment-Rare earths

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

- Unpaired electrons
  - Total angular momentum
    \[ J = L + S \]
  - Total moment
    \[ \mu = L + 2S \]
    \[ \mu = g_J \mu_B J = g_J \beta \frac{eH}{2m_e} \]
Magnetic moment - 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)

\[ \mu = g \mu_B S = 2S \frac{eh}{2m_e} \]

<table>
<thead>
<tr>
<th>Transition Metal</th>
<th>J x S</th>
<th>( \frac{2}{2\sqrt{(S+1)}} )</th>
<th>( \frac{2}{gJ} )</th>
<th>Magnetic Moment (( \mu_B ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti³⁺</td>
<td>3d x 1</td>
<td>1/2</td>
<td>1.73</td>
<td>1.0 ( \mu_B )</td>
</tr>
<tr>
<td>V³⁺</td>
<td>3d x 2</td>
<td>1</td>
<td>2.83</td>
<td>2.0 ( \mu_B )</td>
</tr>
<tr>
<td>Cr³⁺</td>
<td>3d x 3</td>
<td>3/2</td>
<td>3.67</td>
<td>3.0 ( \mu_B )</td>
</tr>
<tr>
<td>Cr²⁺ Mn³⁺</td>
<td>3d x 4</td>
<td>2</td>
<td>4.00</td>
<td>4.0 ( \mu_B )</td>
</tr>
<tr>
<td>Mn²⁺ Fe³⁺</td>
<td>3d x 5</td>
<td>5/2</td>
<td>5.92</td>
<td>5.0 ( \mu_B )</td>
</tr>
<tr>
<td>Fe³⁺ Co³⁺</td>
<td>3d x 6</td>
<td>2</td>
<td>4.00</td>
<td>4.0 ( \mu_B )</td>
</tr>
<tr>
<td>Co²⁺</td>
<td>3d x 7</td>
<td>3/2</td>
<td>3.67</td>
<td>3.0 ( \mu_B )</td>
</tr>
<tr>
<td>Ni²⁺</td>
<td>3d x 8</td>
<td>2</td>
<td>2.80</td>
<td>2.0 ( \mu_B )</td>
</tr>
<tr>
<td>Cu²⁺</td>
<td>3d x 9</td>
<td>1</td>
<td>1.70</td>
<td>1.0 ( \mu_B )</td>
</tr>
</tbody>
</table>
Itinerant magnetism

Weak or variable moment size

- Stoner ferromagnetism (Fe, Ni)

- Fermi surface nesting driven antiferromagnetism (Cr)
Magnetic structures

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

Elastic scattering - Bragg’s Law

\[ 2d \sin \theta = n \lambda \]
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**
1-D Cartoons

Configuration in Real Space

Diffraction Pattern in Reciprocal Space

- 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
Typical 3D magnetic structures
Neutron magnetism

• Spin-1/2 particle
• Magnetic moment

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

\[ \mu_n/\mu_e \approx m_e/m_p \approx 1/2000 \]

Neutron magnetic moment is very small compared to an electron
Dipole interaction

Interaction between neutron and electron

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

\[ U^{\alpha \beta} = \langle \alpha | b - pS_\perp \cdot \sigma | \beta \rangle \]

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

strength \quad moment direction

Neutron spin state processes

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

\[ \left( \frac{1}{2} \gamma r_0 \right)^2 = 73 \text{ millibarns/steradian} \cdot \mu_B^2 \]

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

\( f(\mathbf{Q}) \): Fourier transform of the *atomic* magnetization density

Magnetic neutron scattering signal falls off with \( Q \). Similar to x-ray form factor.

Note that nuclear neutron scattering does NOT fall off with \( Q \).
Magnetic structure factor can be simplified for a *collinear* structure,

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

Scattering differential cross-section for *unpolarized* beam

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

More generally

\[ \frac{d\sigma}{d\Omega} = N(\gamma r_0)^2 \sum_{\tau} \delta(Q - \tau)|\hat{Q} \times \{M(\tau) \times \hat{Q}\}|^2 \]
**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 propagation vectors/multi-q structures
Confirmation of AF structure

In 1949, Clifford Shull observed additional magnetic reflections in MnO using neutron diffraction, which led to the confirmation of antiferromagnetism.

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

<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>
</thead>
<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>
</tr>
<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></td>
<td></td>
<td></td>
<td></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

CAM
Slip cone
Cone

Temperature

MAGNETIC FIELD (T)

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Ames Laboratory

IOWA STATE UNIVERSITY
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Why use neutron polarization?

- Separate magnetic/nuclear scattering (q=0 structures)
- Refine structure determination (eg. canting)
- Separate coherent/incoherent (diffuse scattering, mag. densities)

Non-spin flip scattering

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

Spin flip scattering

\[ U^{+-} = -p(S_{\perp x} + iS_{\perp y}) \]
\[ U^{-+} = -p(S_{\perp x} - iS_{\perp y}) \]

What happens when the neutron polarization is parallel to Q?
Instrumentation

Polarizing monochromator

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

Cu$_2$MnAl (111) (Heusler)

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

• **Useful modes**
  - $\mathbf{P} \parallel \mathbf{Q}$ (in-plane polarization): All magnetic scattering is SF
  - $\mathbf{P} \perp \mathbf{Q}$ (vertical polarization): magnetic scattering can be SF & NSF
Polarized experiments

\[ \text{Fe}_2\text{O}_3 \]

\[ \text{NSF} \]

\[ \text{SF} \]

P || Q: separation of magnetic/nuclear paramagnetic scattering

Polarization @ pulsed source

Neutron absorption by $^3$He is polarization dependent

$^3$He polarizers

Heusler mono won’t work for wide angle scattering

Need to keep $^3$He spins aligned in a gas. Optical pumping + magnetic field.
Further references

• Magnetic neutron scattering

• Structural refinements
  – FullProf: http://www.ill.eu/sites/fullprof/

• Magnetic space groups
  – Izyumov, Ozerov, “Neutron diffraction of magnetic materials”
  – Sarah program (representational analysis):
    http://fermat.chem.ucl.ac.uk/spaces/willsgroup/software/