Neutron scattering for magnetism

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The neutron as a probe of condensed matter
Neutron-matter interaction processes
Diffraction by a crystal: nuclear and magnetic structures
Inelastic neutron scattering: magnetic excitations
Use of Polarized neutrons
Techniques for studying magnetic nano-objects
Complementary muon spectroscopy technique
Conclusion
The neutron as a probe of condensed matter
The neutron as a probe of condensed matter

Subatomic particle discovered in 1932 by Chadwik
First neutron scattering experiment in 1946 by Shull

**Properties:**

\[ m_N = 1.675 \times 10^{-27} \text{ kg}, \ s = 1/2, \ \tau = 888 \text{ s} \]

- Neutron: particle/plane wave with \( E = \frac{\hbar^2 k^2}{2m_N} \) and \( \lambda = \frac{2\pi}{k} \)

- Wavelength of the order of few Å (thermal neutrons) \( \approx \) interatomic distances
  Interference \( \rightarrow \) diffraction condition
The neutron as a probe of condensed matter

Subatomic particle discovered in 1932 by Chadwick
First neutron scattering experiment in 1946 by Shull

**Properties:**

\( m_N = 1.675 \times 10^{-27} \text{ kg}, \, s = 1/2, \, \tau = 888 \text{ s} \)

- Neutron: particle/plane wave with \( E = \frac{\hbar^2 k^2}{2m_N} \) and \( \lambda = \frac{2\pi}{k} \)

- Wavelength of the order of few Å \( \approx \) interatomic distances \( \rightarrow \) diffraction condition

- Energies of thermal neutrons \( \approx 25 \text{ meV} \)
  \( \approx \) energy of excitations in condensed matter

- Neutral: probe volume, nuclear interaction with nuclei
The neutron as a probe of condensed matter

**Properties:**

- carries a spin $\frac{1}{2}$: sensitive to the magnetism of unpaired electrons (spin and orbit)
  - Probe magnetic structures and dynamics
  - Possibility to polarize the neutron beam

\[ \vec{\mu}_n = -\gamma \mu_N \vec{\sigma} \]

- Better than X rays for light or neighbor elements or isotopes (ex. H, D): complementary
- Neutron needs big samples!
The neutron as a probe of condensed matter

≠ TYPES OF NEUTRON SOURCES FOR RESEARCH:

• **Neutron reactor** (continuous flux)
  ex. Institut Laue Langevin in Grenoble

• **Spallation sources** (neutron pulses)
  ex. ISIS UK or ESS future European spallation source (Lund)

• **Compact source** projects (neutron pulses)

Images ILL and ESS websites
The neutron as a probe of condensed matter

≠ TYPES OF NEUTRON SOURCES FOR RESEARCH:

\[ \lambda = 5 \, \text{Å} \]
The neutron as a probe of condensed matter

**VARIOUS ENVIRONMENTS:**

- **Temperature:** 30 mK-2000 K
- **High magnetic steady fields** up to 26 T  
  Pulsed fields up to 40 T
- **Pressure** (gas, Paris-Edinburgh, clamp cells)  
  up to 100 kbar
- **Electric field**
- **CRYOPAD** zero field chamber  
  for polarization analysis

![Cryopad](image)

D23@ILL  
15 T magnet
The neutron as a probe of condensed matter

**Use of neutron scattering for magnetic studies:**

- Most powerful tool to determine complex magnetic structures (non-collinear, spirals, sine waves modulated, incommensurate, skyrmion lattice)
- Complex phase diagrams (T, P, H, E) under extreme conditions
- Magnetic excitations and hybrid excitations
- In situ, in operando measurements
- Magnetic domains probe
- Short-range magnetism (ex. spin liquid/glass/ice)
- Magnetic nano-structures/mesoscopic magnetism
- Chirality determination
- Materials with hydrogen

*Example:* orthorhombic $R$MnO$_3$

Goto et al. PRL (2005)
Neutron-matter interaction processes
**SCATTERING PROCESS: INTERFERENCE PHENOMENA**

Born approximation

\[ \vec{Q} = \vec{k}_i - \vec{k}_f \]

Momentum transfer = scattering vector

Source \( \rightarrow \) sample \( \rightarrow \) detector \( \rightarrow \) plane waves

Detector \( d\Omega \)
SCATTERING PROCESS: INTERFERENCE PHENOMENA

\[ \vec{Q} = \vec{k}_i - \vec{k}_f \]

Scattering vector

Energy transfer:

\[ \hbar \omega = E_i - E_f = \frac{\hbar^2}{2m} (k_i^2 - k_f^2) \]

Elastic scattering: \( \hbar \omega = 0 \)

\[ E_i = E_f \quad |k_i| = |k_f| \quad Q = 2 \sin \theta / \lambda \]
**Neutron-matter interaction processes**

**Scattering process: interference phenomena**

\[ \vec{Q} = \vec{k}_i - \vec{k}_f \]

**Scattering vector**

**Energy transfer:**

\[ \hbar \omega = E_i - E_f = \frac{\hbar^2}{2m} (k_i^2 - k_f^2) \]

Inelastic scattering: \( \hbar \omega \neq 0 \)

\[ E_i < E_f \quad |k_i| < |k_f| \]
Scattering process: interference phenomena

\[ \mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f \]

Scattering vector

Energy transfer:

\[ \hbar \omega = E_i - E_f = \frac{\hbar^2}{2m} (k_i^2 - k_f^2) \]

Inelastic scattering: \( \hbar \omega \neq 0 \)

\[ E_i > E_f \quad |k_i| > |k_f| \]
**Scattering process: interference phenomena**

The cross-sections (in barns $10^{-24}$ cm$^2$) are quantities measured during a scattering experiment:

- **Total cross-section** $\sigma$: number of neutrons scattered per second / flux of incident neutrons

- **Differential cross section** $\frac{d\sigma}{d\Omega}$: per solid angle element

- **Partial differential cross section** $\frac{d^2\sigma}{d\Omega dE}$: per energy element
Fermi’s Golden Rule

Partial differential cross section

\[
\frac{d^2 \sigma}{d\Omega dE} = \frac{k_f}{k_i} \left( \frac{m_N}{2\pi \hbar^2} \right)^2 \sum_{\lambda, \sigma_i} \sum_{\lambda', \sigma_f} p_{\lambda, \sigma_i} p_{\lambda', \sigma_f} |\langle k_f \sigma_f \lambda_f | V | k_i \sigma_i \lambda_i \rangle|^2 \delta(\hbar \omega + E - E')
\]

Energy conservation

Initial and final wave vector and spin state of the neutrons

Initial and final state of the sample

Interaction potential = Sum of nuclear and magnetic scattering
Neutron-matter interaction processes

Interaction potential $= \text{Sum of nuclear and magnetic scattering}$

Nuclear interaction potential
- very short range
- isotropic

$$V(\vec{r}) = \left(\frac{2\pi\hbar^2}{m_N}\right) \sum_i b_i \delta(\vec{r} - \vec{R}_i)$$

Magnetic interaction potential
- Longer range (e$^-$ cloud)
- Anisotropic

Dipolar interaction of the neutron magnetic moments $\mu_n$ with magnetic field from unpaired e$^-$

$$V(\vec{r}) = -\mu_n \cdot \vec{B}(\vec{r})$$

$b$ Scattering length
depends on isotope and nuclear spin

$$\vec{B}(\vec{r}) = \frac{\mu_0}{4\pi} \sum_i \text{rot} \left( \frac{\vec{\mu}_{ei} \times (\vec{r} - \vec{R}_i)}{|\vec{r} - \vec{R}_i|^3} \right) - \frac{2\mu_B}{\hbar} \vec{p}_i \times \frac{(\vec{r} - \vec{R}_i)}{|\vec{r} - \vec{R}_i|^3}$$

Spin contribution  Orbital contribution
Neutron-matter interaction processes

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \left( \frac{m_N}{2\pi\hbar^2} \right)^2 \sum_{\lambda,\sigma_i} \sum_{\lambda',\sigma_f} p\chi p\sigma_i \left| \langle k_f \sigma_f \lambda_f | V | k_i \sigma_i \lambda_i \rangle \right|^2 \delta (\hbar \omega + E - E')
\]

Some algebra (hyp. no spin polarization)

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \sum_{jj'} \int_{-\infty}^{+\infty} \langle A_j^*(0) A_{j'}(t) e^{-i\vec{Q}\cdot\vec{R}_j(0)} e^{i\vec{Q}\cdot\vec{R}_j(t)} \rangle e^{-i\omega t} dt
\]

with \( A_j(t) \) the scattering amplitude

Scattering experiment
\[\rightarrow\] FT of interaction potential
Neutron-matter interaction processes

\[
\frac{d^2 \sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{1}{2\pi \hbar} \sum_{jj'} \int_{-\infty}^{+\infty} \langle A^*_j, (0) A_j(t) e^{-i \mathbf{Q} \cdot \mathbf{R}_j, (0)} e^{i \mathbf{Q} \cdot \mathbf{R}_j(t)} \rangle e^{-i \omega t} dt
\]

\[
b_j = \sum_{jj'} \langle A^*_j, (0) A_j(t) e^{-i \mathbf{Q} \cdot \mathbf{R}_j, (0)} e^{i \mathbf{Q} \cdot \mathbf{R}_j(t)} \rangle e^{-i \omega t} dt
\]

\[
p = 0.2696 \times 10^{-12} \text{ cm}
\]

Magnetic form factor of the free ion

X-rays

neutrons

\[
pf_j(Q) \mathbf{M}_{j\perp}(\mathbf{Q}, t)
\]
Neutron-matter interaction processes

\[
\frac{d^2 \sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{1}{2\pi \hbar} \sum_{jj'} \int_{-\infty}^{+\infty} \langle A^*,(0)A_j(t)e^{-i\vec{Q}\vec{R}_j(0)}e^{i\vec{Q}\vec{R}_j(t)} \rangle e^{-i\omega t} dt
\]

Projection of the magnetic moment \( \perp \vec{Q} \)
Neutron-matter interaction processes

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \sum_{jj'} \int_{-\infty}^{+\infty} \langle A^*_{jj'}(0) A_{jj'}(t) e^{-i\vec{Q}\cdot\vec{R}_{jj'}(0)} e^{i\vec{Q}\cdot\vec{R}_{jj'}(t)} \rangle e^{-i\omega t} dt
\]

\[
b_j \quad p f_j(Q) \vec{M}_{jj} (\vec{Q}, t)
\]

= Double FT in space and time of the pair correlation function of the nuclear density magnetic density \( \perp \vec{Q} \)
Separation elastic/inelastic:

Keeps only the time-independent terms in the cross-section and integrate over energy

→ elastic scattering (resulting from static order)
Diffraction by a crystal: nuclear and magnetic structures
Diffraction by a crystal: nuclear and magnetic structures

**Nuclear Diffraction**

Crystal = lattice + basis

Crystal lattice: \( \vec{R}_n = u_n \vec{a} + v_n \vec{b} + w_n \vec{c} \)

Reciprocal lattice: \( \vec{H} = h\vec{a}^* + k\vec{b}^* + l\vec{c}^* \)

\[
\frac{d\sigma}{d\Omega} = \sum_{j,j'} <b_j b_{j'}, e^{-i\vec{Q}(\vec{R}_{j'} - \vec{R}_j)}> 
\]

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} |F_N(\vec{Q})|^2 \delta(\vec{Q} - \vec{H}) 
\]

Coherent elastic scattering from crystal

→ Bragg peaks at nodes of reciprocal lattice

**Diffraction Condition (lattice)**
**Nuclear diffraction**

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} |F_N(\vec{Q})|^2 \delta(\vec{Q} - \vec{H})
\]

For a non Bravais lattice:

\[\nu\] atoms/unit cell

\[
\vec{R}_{n\nu} = \vec{R}_n + \vec{r}_\nu \quad \text{with}
\]

\[
\vec{r}_\nu = x_\nu \vec{a} + y_\nu \vec{b} + z_\nu \vec{c}
\]

\[
F_N(\vec{Q}) = \sum_\nu b_\nu e^{i\vec{Q} \cdot \vec{r}_\nu}
\]

Information on atomic arrangement inside unit cell
Magnetic diffraction

Magnetic ordering may not have the same periodicity as the nuclear one → propagation vector $\vec{\tau}$ → periodicity and propagation direction

Moment distribution is a periodic function of space → can be Fourier expanded:

$$\vec{\mu}_{n,\nu} = \sum_{\vec{\tau}} \vec{m}_{\nu,\vec{\tau}} e^{-i\vec{\tau}.\vec{R}_n}$$

Magnetic moment of atom $\nu$ in $n^{th}$ unit cell

Fourier component associated to $\vec{\tau}$

Example: For a unique propagation vector $\vec{\tau} = (1/2, 0, 0) = a^*/2$

$$\vec{\mu}_{n,\nu} = \vec{m}_{\nu} e^{-i2\pi n/2}$$

Staggered magnetic moments = doubling of the nuclear cell
Diffraction by a crystal: nuclear and magnetic structures

**Magnetic Diffraction**

Magnetic ordering may not have the same periodicity as nuclear one → **propagation vector** \( \vec{\tau} \) → periodicity and propagation direction

Moment distribution is a periodic function of space → can be Fourier expanded:

\[
\vec{\mu}_{n,\nu} = \sum_{\vec{\tau}} \vec{m}_{\nu,\vec{\tau}} e^{-i\vec{\tau} \cdot \vec{R}_n}
\]

Magnetic moment of atom \( \nu \) in \( n^{th} \) unit cell

Fourier component associated to \( \vec{\tau} \)

Magnetic periodicity = \( x \) times nuclear periodicity → \( \vec{\tau} = (1/x, 0, 0) \)
MAGNETIC DIFFRACTION

Magnetic ordering may not have same periodicity as nuclear one
→ propagation vector $\vec{\tau}$ → periodicity and propagation direction

For a non-Bravais lattice

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_M \perp (\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})$$

→ Bragg peaks at satellites positions $\vec{Q} = \vec{H} \pm \vec{\tau}$
**Magnetic Diffraction**

Magnetic ordering may not have the same periodicity as the nuclear one

→ **propagation vector** \( \vec{\tau} \) → periodicity and propagation direction

For a non-Bravais lattice

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_M \perp (\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})
\]

**Magnetic structure factor: information on magnetic arrangement in unit cell**

\[
\vec{F}_M (\vec{Q} = \vec{H} + \vec{\tau}) = \rho \sum f_\nu (\vec{Q}) \vec{m}_\nu,\vec{\tau} e^{i\vec{Q}.\vec{r}_\nu}
\]

Fourier component
Diffraction by a crystal: nuclear and magnetic structures

**Magnetic diffraction**

Magnetic ordering may not have same periodicity as nuclear one

\[ \rightarrow \text{propagation vector } \vec{\tau} \rightarrow \text{periodicity and propagation direction} \]

For a non-Bravais lattice

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_M\perp(\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})
\]

- What is the magnetic structure described by a zero propagation vector?
- What is the propagation vector describing a type A antiferromagnet?
- What is the propagation vector associated to a magnetic helix of periodicity 8a?
**Magnetic Diffraction: Classification of the Magnetic Structures**

If $\vec{\tau} = 0$, magnetic/nuclear structures same periodicity  
$\rightarrow$ Bragg peaks at reciprocal lattice nodes $\vec{Q} = \vec{H}$

- Bravais lattice $\vec{\tau}=0 \Rightarrow$ ferromagnetic structure

![Diagram showing direct and reciprocal space with magnetic and nuclear structures](image-url)
**Magnetic Diffraction: Classification of the Magnetic Structures**

If $\vec{\tau} = 0$, magnetic/nuclear structures same periodicity
→ Bragg peaks at reciprocal lattice nodes $\vec{Q} = \vec{H}$

• Non Bravais lattice $\vec{\tau} = 0 \Rightarrow$ ferromagnetic or antiferromagnetic structure

Intensities of magnetic peaks
→ Arrangement of moments in cell

Direct space

Reciprocal space

Magnetic • Nuclear
Magnetic diffraction: classification of the magnetic structures

If \( \vec{r} \neq 0 \), magnetic satellites at \( \vec{Q} = \vec{H} \pm \vec{r} \)

Ex. \( \vec{r} = \vec{H}/2 = (1/2,0,0) \), collinear antiferromagnetic structure
Magnetic Diffraction: Classification of the Magnetic Structures

If $\vec{\tau} \neq 0$ and $\vec{\tau} \neq \vec{H}/2$, magnetic satellites at $\vec{Q} = \vec{H} \pm \vec{\tau}$

Sine wave amplitude modulated and spiral structures $\vec{\tau} = (\tau, 0, 0)$

\[
\mu_{n\nu} = \mu_{\nu} \hat{u} \cos(\vec{\tau} \cdot \vec{R}_n + \Phi_{\nu})
\]

\[
\mu_{n\nu} = \mu_{1\nu} \hat{u} \cos(\vec{\tau} \cdot \vec{R}_n + \Phi_{\nu}) + \mu_{2\nu} \hat{v} \sin(\vec{\tau} \cdot \vec{R}_n + \Phi_{\nu})
\]
Magnetic diffraction: classification of the magnetic structures

If \( \vec{\tau} \neq 0 \) and \( \vec{\tau} \neq \vec{H}/2 \), magnetic satellites at \( \vec{Q} = \vec{H} \pm \vec{\tau} \)

Sine wave amplitude modulated and spiral structures \( \vec{\tau} = (\tau, 0, 0) \)

Rational/irrational \( \vec{\tau} = \text{commensurate/incommensurate} \) magnetic structure
Diffraction by a crystal: nuclear and magnetic structures

**Magnetic Diffraction: Classification of the Magnetic Structures**

Multi-\(\vec{\tau}\) magnetic structure

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_{M\perp}(\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})
\]

Ex. canted structure with \(\vec{\tau} = 0\) and \(\vec{\tau} = \vec{H}/2 = (1/2,0,0)\)

![Diagram of Direct space and Reciprocal space with magnetic and nuclear structures]
Diffraction by a crystal: nuclear and magnetic structures

Magnetic diffraction: classification of the magnetic structures

Complex magnetic structures:
Sine wave amplitude modulated spiral (helix, cycloid), canted structures → due to frustration, competition of interactions, Dzyaloshinskii-Moryia/anisotropic interaction…

Ex. in rare earth metals
Magnetic diffraction: classification of the magnetic structures

Sine wave amplitude modulated spiral (helix, cycloid), canted structures

Ex. in multiferroics

\[ \text{TbMnO}_3 \]

Kenzelmann et al., PRL 2007

28 K < T < 41 K: incommensurate sine wave modulated \( \rightarrow \) paraelectric

T < 28 K: commensurate spiral (cycloid) \( \rightarrow \) ferroelectric
**Magnetic Diffraction: Techniques**

**Powder Diffraction**
- Bragg’s law: \( Q = \frac{2 \sin \theta}{\lambda} \)
- Example: Fixed \( \lambda \) and varying \( \theta \) (or multidetector)

\[ I(|\vec{Q}|) \]

**Single-Crystal Diffraction**
- Complex structures, magnetic domains, bulky environments
- Bring a reciprocal node in coincidence with \( \vec{Q} = \vec{k}_i - \vec{k}_f \)
- Then measure the integrated intensity (rocking curve)

\[ I(\vec{Q}) \]

- 4-circles mode
- Powder diffractometer
- Lifting arm
**Magnetic Diffraction: Solving a Magnetic Structure**

**Finding the propagation vector** $\vec{\tau}$ (periodicity of magnetic structure):
powder diffraction $\rightarrow$ difference between measurements below and above $T_c$.
Indexing magnetic Bragg reflections with $\vec{Q} = \vec{H} \pm \vec{\tau}$

**Refining magnetic Bragg peaks intensities** (powder and single-crystal)
and domain populations (single-crystal)
$\rightarrow$ moment amplitudes and magnetic arrangement of atoms in the cell
(use scaling factor from nuclear structure refinement) with programs like Fullprof

https://www.ill.eu/sites/fullprof/

**Help from group theory and representation analysis**
Use of rotation/inversion symmetries to infer possible magnetic arrangements compatible
with the symmetry group that leaves the propagation vector invariant $\rightarrow$ constrains the refinement
MAGNETIC DIFFRACTION: EXAMPLES

Original powder diffraction experiment in MnO from Shull et al. Phys. Rev. (1951)

\[ T_N = 116 \text{ K} \]
**Diffraction by a crystal: nuclear and magnetic structures**

**MAGNETIC DIFFRACTION: EXAMPLES**

Original powder diffraction experiment in MnO from Shull *et al.* *Phys. Rev.* (1951)

$T_N = 116$ K

Propagation vector

$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$

$T < T_N$

$80$ K

$T > T_N$

$293$ K

Mn atoms in MnO

---

Chemical unit cell

Magnetic unit cell

---

Neutron counts

Scattering angle

ESM 2019, Brno
Magnetic diffraction: Examples

Original powder diffraction experiment in MnO from Shull et al. *Phys. Rev.* (1951)

- Confirmation of antiferromagnetism
  Predicted by *Louis Néel* in 1936
Diffraction by a crystal: nuclear and magnetic structures

**Magnetic diffraction: Examples**

\( \text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14} \quad \text{Marty et al., PRL 2008} \)

![Diagram of a triangular lattice of Fe\(^{3+}\) triangles, S=5/2](image)

粉末衍射

磁性转变在 \( T_N = 28 \text{ K} \)

粉末衍射

\( 2 \theta \) (°)

\( \text{Neutron counts (a. u.)} \)

\( \rightarrow \) 传播矢量 \( \vec{\tau} = (0, 0, 1/7) \)
**Magnetic Diffraction: Examples**

**Ba$_3$NbFe$_3$Si$_2$O$_{14}$**  *Marty et al., PRL 2008*

- **Refinement of integrated neutron intensities**

![Graph showing measured vs. calculated intensity](image)

- Single-crystal diffraction
**Magnetic Diffraction: Examples**

**Ba$_3$NbFe$_3$Si$_2$O$_{14}$  Marty et al., PRL 2008**

- Triangles of magnetic moments in (a, b) plane
- Magnetic helices propagating along $c$ with period $\approx 7c$
Diffraction by a ill-ordered magnetic systems

atomic states

Magnetic states

crystallized solid

Magnetic order

liquid

Spin liquid

gas

paramagnet
Diffraction by a ill-ordered magnetic systems

Ex.: Spin liquid = no order/strong fluctuations despite presence of spin pair correlations

Diffuse neutron scattering map of spin ice

*Fennell et al., Science 2009*

\[ S(Q) \]

- Paramagnetic scattering
- Bragg peaks
- Correlated diffuse scattering

\[ Q \]

Powder diffraction

Single-crystal diffraction
Inelastic neutron scattering: magnetic excitations
Inelastic neutron scattering: nuclear and magnetic excitations

Collective excitations

- Magnons
- Spin relaxation (spin glass, spin ice etc.)

Critical fluctuations

Crystal field levels

Itinerant magnetism

Inter-multiplets splittings

- Adapted from T. Perring, lecture at the Oxford Neutron School

Elastic peak

\[ S(Q, \omega) \]

\[ \hbar \omega \text{ (meV)} \]

\[ t \text{ (sec.)} \]

0.1 \[ 10^{-11} \]

1 \[ 10^{-12} \]

10 \[ 10^{-13} \]

100 \[ 10^{-14} \]

1000 \[ 10^{-15} \]
Inelastic neutron scattering: magnetic excitations

https://europeanspallationsource.se/science-using-neutrons
Inelastic neutron scattering: magnetic excitations

**Inelastic scattering: magnetic**

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \sum_{jj'} \int_{-\infty}^{+\infty} \langle A_j^*(0) A_j(t) e^{-i\vec{Q}\vec{R}_j'(0)} e^{i\vec{Q}\vec{R}_j(t)} \rangle e^{-i\omega t} dt
\]

with \( A_j(t) = pf_j(Q) \vec{M}_j \perp (\vec{Q}, t) \)

\[ \text{again some algebra} \]

\[
\frac{d\sigma^2}{d\Omega dE} = (\gamma r_0^2) \frac{k_f}{k_i} f^2(\vec{Q}) \sum_{\alpha, \beta} \left[ \delta_{\alpha, \beta} - \frac{Q\alpha Q\beta}{Q^2} \right] S^{\alpha, \beta}(\vec{Q}, \omega)
\]
\[ \frac{d\sigma^2}{d\Omega dE} = (\gamma r_0^2) \frac{k_f}{k_i} f^2(Q) \sum_{\alpha, \beta} \left[ \delta_{\alpha, \beta} - \frac{Q_\alpha Q_\beta}{Q^2} \right] S^{\alpha, \beta}(\vec{Q}, \omega) \]

- **Magnetic form factor (squared)**
- **Polarization factor**

**Scattering function:** spin-spin correlation function \( \langle \vec{S}_j^\alpha(0) \vec{S}_j^\beta(t) \rangle \)
related to the dynamical susceptibility via the fluctuation-dissipation theorem

\[ S^{\alpha, \beta}(\vec{Q}, \omega) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \chi^{''\alpha, \beta}(\vec{Q}, \omega) \]
Inelastic neutron scattering: magnetic excitations

**Inelastic scattering: spin waves**

Quantum description: spin wave mode = quasi-particle called magnon

Creation/annihilation processes in cross-section

\[
\frac{d^2\sigma}{d\Omega dE} = (\gamma r_0)^2 \frac{k_f}{k_i} \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{q}} f(Q)^2 |F(\vec{Q})|^2 < n_\pm > \delta(\omega \mp \omega_{\vec{q}}) \left\{ \begin{array}{l} \delta(\vec{Q} - \vec{H} - \vec{q}) \\ \delta(\vec{Q} - \vec{H} + \vec{q}) \end{array} \right. 
\]
**Inelastic neutron scattering: magnetic excitations**

**Inelastic scattering: spin waves**

*Spin waves* (magnons): elementary excitations of magnetic compounds = transverse oscillations in relative orientation of the spins

Characterized by wave vector $\vec{q}$, a frequency $\omega$

Only certain spin components involved

*Ferromagnetic* $J > 0$

\[ \mathcal{H} = - \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j \]

*Antiferromagnetic* $J < 0$
**Inelastic scattering: magnetic excitations**

**Inelastic scattering: spin waves**

Spin waves (magnons): elementary excitations of magnetic compounds= transverse oscillations in relative orientation of the spins

Characterized by wave vector $\vec{q}$, a frequency $\omega$

Only certain spin components involved

**Dispersion relation** $\omega(\vec{q})$

Crystal with $p$ atoms/unit cell:

$p$ branches

\[ E(q) = 4JS(1 - \cos(qa)) \]

Ferromagnetic $J > 0$

\[ E(q) = -4JS|\sin(qa)| \]

Antiferromagnetic $J < 0$
Inelastic neutron scattering: magnetic excitations

**INELASTIC SCATTERING: TECHNIQUES**

**Instrument time-of-flight**
- Neutron pulses (spallation/chopped): time and position on multidetector give final $E$ and $\vec{Q}$
- Powder and single-crystal: access to wide region of reciprocal space

**Instrument triple-axis**
- Position at $\vec{Q}$ point and energy analyzer: single-crystal, bulky sample environment, polarized neutrons

\[
Q = k_i - k_f
\]
\[
E = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)
\]
**Inelastic neutron scattering: magnetic excitations**

**INELASTIC SCATTERING: NUCLEAR VERSUS MAGNETIC**

**Nuclear excitations (phonons)**

- Form factor $\propto Q^2$
- Intensity max for $\vec{Q} \parallel \vec{e}$ and zero for $\vec{Q} \perp \vec{e}$
- with $\vec{e}$ the polarization of the mode

**Magnetic excitations (spin waves)**

- Form factor $\propto Q$
- Intensity maximum for $\vec{M} \perp \vec{Q}$

**Purposes of inelastic scattering experiments:**

**Nuclear:** Information on elastic constants, sound velocity, structural instabilities…

**Magnetic:** Information on magnetic interactions and microscopic mechanisms yielding the magnetic properties…

**In multiferroics:** Spin-lattice coupling, hybrid modes ex. electromagnons
Spin waves in MnO

$J_1 = 0.77 \pm 0.02, \ J_2 = 0.89 \pm 0.02 \text{ meV, } + \text{ (anisotropies, exchange striction…)}$

Bonfante et al. Solid State Com. 1972
Kohgi et al. Solid State Com. 1972
Inelastic neutron scattering: magnetic excitations

Inelastic scattering: examples

Spin waves in $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$ single crystal

Loire et al. PRL 2011
Chaix et al. PRB 2016

Analysis of spin waves dispersion using Holstein-Primakov formalism in linear approximation
Magnetic structure and Hamiltonian are inputs of existing programs (SpinWave, SpinW)

http://www-llb.cea.fr/logicielsllb/SpinWave/SW.html
https://www.psi.ch/de/spinw
Inelastic neutron scattering: magnetic excitations

**INELASTIC SCATTERING: EXAMPLES**

**Spin waves in Ba$_3$NbFe$_3$Si$_2$O$_{14}$ single crystal**

**Experiment**

**Calculation**

- Determination of the Hamiltonian
- Interpretation of multiferroic properties

\[ H = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{ij} D_{ij} \mathbf{S}_i \times \mathbf{S}_j + \sum_{i,\alpha} K_{\alpha} (\hat{n}_{\alpha} \cdot \mathbf{S}_i)^2 \]
Inelastic neutron scattering: magnetic excitations

**Inelastic scattering: examples other than spin waves**

**Localized excitations**

Transition between energy levels:

*Discrete non dispersive signal*

Example crystal field excitations in rare-earth ions

Ho\(^{3+}\) in Ho\(_2\)Ir\(_2\)O\(_7\), Lefrançois et al. Nat. Com. 2017
**Inelastic neutron scattering: magnetic excitations**

**INELASTIC SCATTERING: EXAMPLES OTHER THAN SPIN WAVES**

**Localized excitations**

Transition between energy levels:
**Discrete non dispersive signal**
Example crystal field excitations in rare-earth ions

**Quantum excitations**

Spinons (≈ domains walls)
→ Freely propagate
→ Ungapped **continuum**

**Dispersion relation**

KCuF₃: 1D antiferromagnets with spin S=1/2, *Nagler et al.* *PRB* 1991

Ho³⁺ in Ho₂Ir₂O₇, *Lefrançois et al.* *Nat. Com.* 2017

\[
(0, 0, l)
\]
Use of Polarized neutrons
Use of Polarized neutrons

Cross section depends on the spin state of the neutron. Polarized neutron experiment uses this spin state and its change upon scattering process to obtain additional information.

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \left( \frac{m_N}{2\pi\hbar^2} \right)^2 \sum_{\lambda,\sigma_i} \sum_{\lambda',\sigma_f} \rho_{\lambda\sigma_i} p_{\lambda\sigma_i} \left| \langle k_f \sigma_f \lambda_f | V | k_i \sigma_i \lambda_i \rangle \right|^2 \delta(\hbar\omega + E - E')
\]

Different techniques using polarized neutrons depending on the way initial \( P_i \) and final \( P_f \) polarizations are analyzed:
- Half polarized experiments (either \( P_i \) or \( P_f \))
Use of Polarized neutrons

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Different techniques using polarized neutrons depending on the way initial $P_i$ and final $P_f$ polarizations are analyzed:
- Half polarized experiments (either $P_i$ or $P_f$)
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Different techniques using polarized neutrons depending on the way initial $P_i$ and final $P_f$ polarizations are analyzed:
- Half polarized experiments (either $P_i$ or $P_f$)
- Longitudinal polarization analysis
- Spherical polarization analysis
→ Used in diffraction and inelastic scattering
Use of Polarized neutrons

- Amplifies magnetic signal
- Measurement of magnetic form factor
- Atomic site susceptibility tensor
- Magnetization density map

Spin density maps in URu$_2$Si$_2$

*Ressouche et al PRL 2012*
Use of Polarized neutrons

- Amplifies magnetic signal
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- Magnetization density map

**Unique magnetic chirality**
in \( \text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14} \) *Marty et al. PRL 2008*

- Separation magnetic/nuclear
- Access to spin components \( M_x, M_z \)
- Access to magnetic/nuclear chirality

Spin density maps in \( \text{URu}_2\text{Si}_2 \)
*Ressouche et al PRL 2012*
Use of Polarized neutrons

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Unique magnetic chirality in \( \text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14} \) [Marty et al. PRL 2008]

- Separation magnetic/nuclear
- Access to spin components \( M_y, M_z \)
- Access to magnetic/nuclear chirality

- Refine complex magnetic structure
- Probe magnetic domains

Spin density maps in \( \text{URu}_2\text{Si}_2 \) [Ressouche et al PRL 2012]

Antiferromagnetic domains in \( \text{MnPS}_3 \)

Magnetic moments reversed [Ressouche et al PRB 2010]
Techniques for studying magnetic nano-objects
Techniques for studying magnetic nano-objects

SMALL ANGLE SCATTERING AND REFLECTOMETRY

Techniques to probe various kinds of nanostructures
Use of polarized neutrons
Reflectometry, SANS,
combination of both (GISANS)

SANS: small q = large objects

Mühlbauer et al. Rev. Mod. Phys. 2019
Techniques for studying magnetic nano-objects

**Small Angle Scattering and Reflectometry**

Applications: long wavelength spin textures, vectorial magnetization profile of ordered or diluted magnetic nanoparticles/nanowires/domain walls and of magnetic multilayers down to the monolayer (depth and lateral structure) in absolute values.
Techniques for studying magnetic nano-objects

**Small Angle Scattering and Reflectometry**

Applications: long wavelength spin textures, vectorial magnetization profile of ordered or diluted magnetic nanoparticles/nanowires/domain walls and of magnetic multilayers down to the monolayer (depth and lateral structure) in absolute values.

Example SANS in MnSi: ordered lattice of skyrmions

Length scales

- 0.3 nm
- 2 nm
- 300 nm
- 100 µm

Techniques

- Off-specular reflectivity
- Grazing Incidence Ultra Small Angle Scattering
- Spin-Echo Grazing Incidence
- Grazing Incidence SANS
- Reflectivity
- Diffraction

*Mühlbauer et al. Science 2009*
Complementary muon spectroscopy technique
Muons are light elementary particles produced by decay of pions. Muons have a spin $\frac{1}{2}$, and remain implanted in matter until their decay = local probe

Muon decay: anisotropic emission of the positron recorded by forward and backward detectors, correlated to muon spin direction.
Muon Spin Spectroscopy (μSR=muon spin resonance/rotation/relaxation)

Internal fields → Larmor precession of the muon spin: oscillations on top of asymmetric decay

Number of positrons collected in the two detectors vs time

Asymmetry of the muon decay

Larmor frequency vs T ∝ internal fields

Use of μSR:
Detection of small static/dynamic internals fields (ordered moments or disordered systems) with high sensitivity ≈ 0.01 μB → Phase diagrams
**Neutron scattering** = best method to determine the magnetic arrangement in bulk matter, especially for complex orders. Also unique tool to measure the magnetic excitations especially at low energies. Drawbacks: needs of big samples → This can be improved with novel sources. Formalism well established.

Internal fields in matter can be measured with alternative highly sensitive techniques such as NMR, Mössbauer, **muon spectroscopy**.

**X-ray scattering** complementary tool. Magnetic scattering rather weak effect (5 orders of magnitude smaller than non-magnetic scattering) compensated by very high brilliance of synchrotron sources and use of resonant techniques (chemically selective) → small samples can be used. Huge progress in RIXS techniques. However still unable to reach low energies accessible by neutron scattering.
Further reading

• Material borrowed from presentations of B. Grenier, L. Chaix, N. Qureshi, E. Ressouche, thanks to them!


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