Neutron scattering
a probe for multiferroics and magnetoelectrics

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Outline of the lecture

The neutron as a probe of condensed matter
  Properties
  Sources
  Environments
  Scattering processes

Diffraction by a crystal
  Nuclear, magnetic structures
  Instruments type
  Examples

Inelastic scattering
  Phonons, spin waves, localized excitations
  Instruments type
  Examples

Polarized neutrons
  Longitudinal, spherical polarization analysis
  Examples
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The neutron as a probe of condensed matter

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

Properties:

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 \( \approx k_B T \approx 25 \text{ meV} \)
  of the order of the excitations in condensed matter

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

Properties:

- carries spin $\frac{1}{2}$: sensitive to spins of unpaired electrons
  - probe magnetic structure and dynamics
  - possibility to polarize neutron beam

- Better than XR for Light or neighbor elements
- Neutron needs big samples

Scattering length

$X$ rays

$\propto Z$

$\text{neutrons}$

ESMF2010, L’Aquila
The neutron as a probe of condensed matter

Two types of neutron sources for research:

- Neutron reactor (ex. ILL Grenoble 57 MW),
- Spallation sources (ex. ISIS UK 1.5 MW)

FISSION

\[ ^{235}\text{U} \]

SPALLATION

\[ \text{U, W, Hg} \]
The neutron as a probe of condensed matter

Various environments:

- Temperature: 15 mK-700 K
- High magnetic fields up to 15 T (D23, ILL)
  Pulsed fields up to 30 T (IN22, ILL)
- Pressure (gas, Paris-Edinburgh, clamp cells)
- Electric field
- CRYOPAD zero field chamber for polarization analysis

CRYOPAD

D23@ILL
15 T magnet
The neutron as a probe of multiferroics and magnetoelctrics

- Complex magnetic structures: spirals, sine waves modulated, incommensurate
- Link with atomic positions
- Complex phase diagrams (T, P, H, E)
- Hybrid excitations: electromagnons
- Domains manipulation under E/H fields
- Chirality determination and manipulation

Example: orthorhombic RMnO$_3$

Goto et al. PRL (2005)
Counts neutrons scattered by sample in solid angle \( d\Omega \) for energy transfer:

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

Cross section in barns \((10^{-24} \text{ cm}^2)\) after integration on energy and solid angle.

Scattering vector:

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

**Scattering processes**

- **Inelastic scattering**
  \( E_{f2} > E_i \) \( |k_{f2}| > |k_i| \)

- **Elastic scattering**
  \( E_f = E_i \) \( |k_f| = |k_i| \) \( Q = 2\sin\theta/\lambda \)

- **Inelastic scattering**
  \( E_{f1} < E_i \) \( |k_{f1}| < |k_i| \)

**Sample**

**Detector**

**Incident neutrons** \( E, k_i \)
Scattering processes

Fermi’s Golden rule

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

Interaction potential \( V \)

Energy conservation

Sum of nuclear and magnetic scattering
Scattering processes

Interaction potential
- very short range
- isotropic

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

b Scattering length depends on isotope and nuclear spin

Interaction potential
- Longer range (e\textsuperscript{-} cloud)
- Anisotropic

dipolar interaction:
neutron magnetic moments with magnetic field from unpaired e\textsuperscript{-}

\[ V(\vec{r}) = -\vec{\mu}_N \cdot \vec{B}(\vec{r}) \]

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

Spin contribution Orbital contribution
Scattering processes

with $A_j(t)$ the scattering amplitude

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

Projection of the Magnetic moment $\perp$ to $\vec{Q}$

maximum intensity for $\vec{M}_\perp(\vec{Q})$

neutron

nucleus $j$

isotropic

neutron

electron $i$
Scattering processes

with $A_j(t)$ the scattering amplitude

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

Scattering experiment $\rightarrow$ FT of interaction potential

$p = 0.2696 \times 10^{-12}$ cm

magnetic form factor of the free ion
Scattering processes

with $A_j(t)$ the scattering amplitude

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

= Double FT in space and time of the correlation function of the nuclear and magnetic density in $(\vec{r}, t)$ and $(\vec{0}, 0)$
Separation elastic/inelastic:
Keeps only the time-independent (static) terms in the cross-section and integrate over energy ➔ elastic scattering

\[
\frac{d\sigma}{d\Omega} = \sum_{j,j'} < A^*_j A_{j'}, e^{-i \bar{Q}(\vec{R}_{j'}, - \vec{R}_j)} >
\]
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**Diffraction from a crystal**

**Nuclear diffraction**

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**

\( \Rightarrow \) **Bragg peaks at nodes of reciprocal 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 Bravais lattice: 1 atom/unit cell

\[ F_N(\vec{Q}) = b \]

Non Bravais lattice: \( \nu \) atoms/unit cell

\[ \vec{R}_{n\nu} = \vec{R}_n + \vec{r}_\nu \text{ with} \]
\[ \vec{r}_\nu = x_\nu \vec{a} + y_\nu \vec{b} + z_\nu \vec{c} \]

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

Information on atomic arrangement inside unit cell
Magnetic diffraction

magnetic ordering may not have same periodicity as nuclear one

→ propagation vector $\vec{\tau}$ → periodicity and propagation direction
Diffraction from a crystal

Magnetic diffraction

magnetic ordering may not have same periodicity as nuclear one

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

for a non-Bravais lattice

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

diffraction condition

\( \rightarrow \) Bragg peaks at satellites positions \( \vec{Q} = \vec{H} \pm \vec{\tau} \)
Diffraction from a crystal

**Magnetic diffraction**

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

for a non-Bravais lattice

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

**magnetic structure factor**: info. on magnetic arrangement in unit cell

$$\vec{F}_M(\vec{Q} = \vec{H} + \vec{\tau}) = p \sum f_\nu(\vec{Q}) \vec{m}_\nu,\vec{\tau} e^{i\vec{Q} \cdot \vec{r}_\nu}$$

$p = 0.2696 \times 10^{-12}$ cm

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

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

Fourier component associated to $\vec{\tau}$
Magnetic diffraction: classification of magnetic structures

If $\mathbf{\tau} = 0$, magnetic/nuclear structures same periodicity

$\Rightarrow$ Bragg peaks at reciprocal lattice nodes

• Bravais lattice $\mathbf{\tau} = 0 \Rightarrow$ ferromagnetic structure
**Magnetic diffraction: classification of magnetic structures**

If $\vec{r} = 0$, magnetic/nuclear structures same periodicity

$\Rightarrow$ Bragg peaks at reciprocal lattice nodes

• **Non Bravais lattice** $\vec{r} = 0 \Rightarrow$ ferromagnetic structure or not

Arrangement of moments in cell $\Leftrightarrow$ intensities of magnetic peaks
Diffraction from a crystal

**Magnetic diffraction: classification of magnetic structures**

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

Ex. $\vec{\tau} = \vec{H}/2$, antiferromagnetic structure

- **Rational $\vec{\tau}$**, commensurate magnetic structure
- **Irrational $\vec{\tau}$**, incommensurate magnetic structure
Diffraction from a crystal

Magnetic diffraction: classification of magnetic structures

\[ \vec{\tau} \neq 0 \text{ and } \vec{\tau} \neq \vec{H}/2 \]

Sine wave modulated and spiral structures

\[ \vec{\mu}_{n\nu} = \mu_{1\nu} \hat{u} \cos(\vec{\tau} \cdot \vec{R}_n + \Phi_{\nu}) + \mu_{2\nu} \hat{u} \sin(\vec{\tau} \cdot \vec{R}_n + \Phi_{\nu}) \]
Diffraction from a crystal

Magnetic diffraction: classification of magnetic structures

\( \vec{r} \neq 0 \) and \( \vec{r} \neq \vec{H} / 2 \)

Sine wave modulated and spiral structures

Example TbMnO\(_3\)  

\( 28 \text{ K} < T < 41 \text{ K} : \) incommensurate sine wave modulated paraelectric

\( T < 28 \text{ K} : \) commensurate spiral (cycloid) ferroelectric

Kenzelmann et al., PRL 2007
Diffraction from a crystal

Magnetic diffraction: classification of magnetic structures

Single-\(\vec{\tau}\) and multi-\(\vec{\tau}\) magnetic structures:

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

ex canted arrangement
Diffraction from a crystal

Solving a magnetic structure

Find the propagation vector (periodicity of magnetic structure): 
powder diffraction $\Rightarrow$ difference between measurements $< \text{and} > T_c$. 
Indexing magnetic Bragg reflections with $\vec{Q} = \vec{H} \pm \vec{\tau}$

Refining Bragg peaks intensities $\Rightarrow$ moment values and 
Magnetic arrangement of atoms in the cell 
(using scaling factor from nuclear structure refinement) 
with programs like Fullprof, CCSL, MXD...

Help with group theory and representation analysis 
Use of rotation/inversion symmetries to infer possible magnetic 
arrangements compatible with the symmetry group that leaves 
$\vec{\tau}$ invariant (pgm like basireps...) (cf. L. Chapon’s lecture)
Diffraction from a crystal

Techniques: powder diffractometers

Bragg’s law \[ Q = \frac{2 \sin \theta}{\lambda} \]

Fixed \( \theta \), varying \( \lambda \):
- time-of-flight diffractometer (spallation source)

Fixed \( \lambda \), varying \( \theta \) (or multidetector):
- 2-axes diffractometer (neutron reactor)
Diffraction from a crystal

Techniques: single-crystal diffractometers

Bring a reciprocal lattice node in coincidence with $\vec{Q} = \vec{k}_i - \vec{k}_f$ then measure its integrated intensity (rocking curve)

4-circles mode

lifting arm (bulky environments)
Diffraction from a crystal

Techniques: difference powder/single-crystal diffraction

All reciprocal space accessible in same acquisition
\[ I(|\vec{Q}|) \]

Best to find propagation vector

Sometimes not enough information to solve structure
(Several Bragg peaks at same \(|\vec{Q}|\) positions)

Each reciprocal space point accessible by set up positioning
\[ I(\vec{Q}) \]

Usually better signal/noise

More information to solve complex magnetic structures

Sometimes problem with magnetic domains
Diffraction from a crystal

Purpose: solve nuclear and magnetic structures, distortion under external parameters, variation of unit cell...

Example: YMnO$_3$
Influence of AF transition on ferroelectric order

Powder diffraction:
Nuclear & magnetic structures ➔ Mn-O distance variation

Lee et al., PRB 2005
Diffraction from a crystal

Example: YMnO$_3$

$\Rightarrow$ Mn-O distance variation

Coupling spin lattice at $T_N$

Lee et al., PRB 2005
Diffraction from a crystal

Example: $\text{Ba}_3\text{NbFe}_2\text{Si}_2\text{O}_{14}$

$\rightarrow$ powder and single-crystal diffraction

Complex magnetic structure

Powder diffraction

Propagation vector $\vec{\tau} = (0, 0, \approx 1/7)$

triangular lattice of $\text{Fe}^{3+}$ triangles, $S=5/2$
Diffraction from a crystal

Example: $\text{Ba}_3\text{NbFe}_2\text{Si}_2\text{O}_{14}$

Single-crystal diffraction

$\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$, magnetic structure

D15 (ILL), $\lambda=1.174\text{Å}$, $T=10\text{ K}$

120° moments on triangles in (a, b) plane

Helices propagating along c

*Marty et al., PRL 2008*
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Inelastic scattering from a crystal

**Reminder on phonons**

Displacement from equilibrium harmonic forces $\vec{R}(t) = \vec{R}_0 + \vec{u}(t)$

Normal modes (stationary waves)
Characterized by wave vector $\vec{q}$
Frequency $\omega$
Polarization $\vec{e}$
- $\vec{e} \parallel \vec{q} \rightarrow$ longitudinal
- $\vec{e} \perp \vec{q} \rightarrow$ transverse

Frequency related to $\vec{q}$ by dispersion relation $\omega(\vec{q})$
Inelastic scattering from a crystal

Reminder on phonons

Displacement from equilibrium
harmonic forces \( \vec{R}(t) = \vec{R}_0 + \vec{u}(t) \)

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Frequency related to \( \vec{q} \) by dispersion relation \( \omega (\vec{q}) \)
Reminder on phonons

Quantum description:

modes = quasi-particles called phonon

Creation/annihilation processes in cross-section

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i} \frac{(2\pi)^3}{V} \sum \sum \frac{|F(Q)|^2}{\omega_q} \langle n_\pm \rangle \delta(\omega + \omega_q) \left\{ \delta(Q - H - q) \right\}
\]

dynamical nuclear structure factor
Inelastic scattering from a crystal

Reminder on spin waves

Spin waves as elementary excitations of magnetic compounds = transverse oscillations in relative orientation of the spins

Characterized by wave vector $\vec{q}$
Frequency $\omega$
Certain spin components involved
Frequency related to $\vec{q}$ by dispersion relation $\omega(\vec{q})$

Spin waves in ferromagnet

$1$ atom/unit cell

$\hbar \omega = 4JS(1 - \cos(\frac{q}{a})\frac{\pi}{a})$
Inelastic scattering from a crystal

Reminder on spin waves

1 atom/unit cell

Crystal with p atoms/unit cell:
p branches

Spin waves in antiferromagnet

\[ \hbar \omega = -4JS|\sin(qa)| \]

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

Creation/annihilation processes in cross-section

\[ \frac{d^2\sigma}{d\Omega dE} = (\gamma r_0)^2 \frac{k_f}{k_i} \frac{(2\pi)^3}{V} \sum_{\vec{q}} \sum_{\vec{Q}} f(Q)^2 |F(\vec{Q})|^2 < n_\pm > \delta(\omega + \omega_{\vec{q}}) \delta(\vec{Q} - \vec{H} - \vec{q}) \]

dynamical magnetic structure factor
Inelastic scattering from a crystal

**Nuclear excitations**

- Form factor: $\propto Q^2$
- Intensity max for $\vec{Q} // \vec{e}$ and zero for $\vec{Q} \perp \vec{e}$

**Magnetic excitations**

- Form factor: $\downarrow$ with $Q$
- Intensity maximum for $\vec{M} \perp \vec{Q}$

**Localized excitations**

- Transition between energy levels: non dispersive signal
- Example crystal field excitations in rare-earth ions
Inelastic scattering from a crystal

**Instruments: triple-axis**

Measure scattered neutrons as a function of $\vec{Q} = \vec{k}_i - \vec{k}_f$

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

Detector

Analyser

Sample

Monochromator

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Inelastic scattering from a crystal

**Instruments: time-of-flight**

Neutron pulses chopped from a monochromatic beam
Arrival time and position on multi detector
give final energy and $Q$

$$\frac{\hbar^2 Q^2}{2m_n} = \hbar \omega + 2E_i - 2E_i \sqrt{1 + \frac{\hbar \omega}{E_i}} \cos(2\theta)$$
Inelastic scattering from a crystal

**Instruments:** Time-of-Flight versus Triple-Axis

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<th>Powder (sometimes single-crystal)</th>
<th>Single-crystal (sometimes powder)</th>
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<td>t-scans at fixed $2\theta \Rightarrow I(</td>
<td>\mathbf{Q}</td>
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<td>Multidetector (all reciprocal space accessible in one acquisition)</td>
<td>Scattering plane accessible only</td>
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<td>Resolution $\frac{\Delta E}{E} \approx 1%$</td>
<td>Resolution $\frac{\Delta E}{E} \approx 5%$</td>
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<td>Used for localized excitations</td>
<td>Use to study collective excitations</td>
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<td>Often not enough to analyze collective excitations</td>
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Inelastic scattering from a crystal

**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 order...

**In multiferroics:**
Spin-lattice coupling, hybrid modes ex. electromagnons
Inelastic scattering from a crystal

Example: hexagonal RMnO$_3$

When spin waves helps solving magnetic structures

Different possible Mn $120^\circ$ magnetic orders: impossible to distinguish with unpolarized neutrons diffraction

![Diagram of hexagonal RMnO$_3$]

$R$ crystal field levels transitions

depends on the two $J_z$ exchange interactions: Given by spin waves

Fabrèges et al., PRL 2009
Inelastic scattering from a crystal

Example: YMnO$_3$
Spin and lattice excitations

Petit et al., PRL 2008

Oxygens
Mn

Phonon mode polarized along the ferroelectric c axis

Example: YMnO$_3$
Spin and lattice excitations

Petit et al., PRL 2008

Oxygens
Mn

Spin wave dispersion

(q 0 12)

(q 0 6)
Inelastic scattering from a crystal

Example: YMnO$_3$  

Petit et al., PRL 2008

Opening of a gap in phonon mode below $T_N$ locked on the spin wave mode

Resonant interaction between both modes = Strong spin-lattice coupling
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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.

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

Polarization \( P = \text{two spin state populations' difference} \)
Use of polarized neutrons

\[
\frac{d^2\sigma}{d\Omega \, dE_f} = \langle N_Q \cdot N_Q^\dagger \rangle_\omega + \langle M_{\perp Q} \cdot M_{\perp Q}^\dagger \rangle_\omega \\
= iP_0 \cdot \langle M_{\perp Q} \wedge M_{\perp Q}^\dagger \rangle_\omega \\
+ P_0 \cdot \left[ \langle N_Q \cdot M_{\perp Q} \rangle_\omega + \langle M_{\perp Q}^\dagger \cdot N_Q \rangle_\omega \right]
\]

Blume-Maleyev equations

Unpolarized neutrons

\[
P_f \frac{d^2\sigma}{d\Omega \, dE_f} = P_0 \cdot \langle N_Q \cdot N_Q^\dagger \rangle_\omega - P_0 \cdot \langle M_{\perp Q}^\dagger \cdot M_{\perp Q} \rangle_\omega \\
+ \langle N_Q^\dagger \cdot M_{\perp Q} \rangle_\omega + \langle M_{\perp Q}^\dagger \cdot N_Q \rangle_\omega \\
+ i\langle M_{\perp Q} \wedge M_{\perp Q}^\dagger \rangle_\omega \\
+ \langle M_{\perp Q} \cdot (P_0 \cdot M_{\perp Q}) \rangle_\omega + \langle (P_0 \cdot M_{\perp Q})^\dagger \cdot M_{\perp Q} \rangle_\omega \\
+ iP_0 \wedge \left[ \langle M_{\perp Q}^\dagger \cdot N_Q \rangle_\omega - \langle N_Q^\dagger \cdot M_{\perp Q} \rangle_\omega \right]
\]

Information about correlations between nuclear-magnetic terms, between different spin components: ex. chiral term
**Use of polarized neutrons**

**Principle of an experiment**

- Select spin state and direction of incident neutron
- Analyze polarization of scattered beam in same direction: longitudinal polarization analysis
- Analyze polarization of scattered beam in any direction: spherical polarization analysis
Use of polarized neutrons

Longitudinal polarization analysis

- Separation magnetic/nuclear
- Access to spin components $M_x$, $M_y$, $M_z$
- Separation coherent/incoherent
- Access to chirality and nuclear/magnetic interference terms
Use of polarized neutrons

Spherical polarization analysis

CRYOPAD
Zero magnetic field chamber

Polarization matrix $P$

Final polarization

$\begin{pmatrix}
P_{XX} & P_{XY} & P_{XZ} \\
P_{YX} & P_{YY} & P_{YZ} \\
P_{ZX} & P_{ZY} & P_{ZZ}
\end{pmatrix}$

→ refine complex magnetic structure
→ Access to non-diagonal terms of the polarization matrix
→ probe domains
Use of polarized neutrons

Example: YMnO$_3$

Longitudinal polarization analysis

\[ \vec{Q} \parallel \vec{P} \parallel \vec{X} \]

Spin flip: magnetism \( T < T_N \)

Non Spin flip: nuclear \( T > T_N \)

Hybrid Goldstone mode of multiferroic phase

“spin waves dresses with atomic fluctuations”

Pailhès et al., PRB 2009
Chirality and ferroelectricity

Chirality: sense of spin precession in spiral or triangular arrangements

\[ \propto \vec{S}_i \times \vec{S}_j \]

Magnetic spiral order \( \Rightarrow \) ferroelectricity with

\[ \vec{P} \propto \vec{r}_{ij} \times (\vec{S}_i \times \vec{S}_j) \]

Katsura et al. PRL (2005)
Mostovoy PRL (2006)
Sergienko et al. PRB (2006)
Use of polarized neutrons

Chirality and ferroelectricity

Example: $\text{Ba}_3\text{NbFe}_2\text{Si}_2\text{O}_{14}$

Static Chirality

2 chiralities associated to helices and triangular arrangements

$\Rightarrow$ 4 possible chiralities $(+1,+1), (-1,-1), (-1,+1), (+1,-1)$

Unpolarized neutron single-crystal diffraction $\Rightarrow$

2 possible magnetic structures $(+1,-1)$ and $(-1,+1)$
Use of polarized neutrons

Chirality and ferroelectricity

Example: $\text{Ba}_3\text{NbFe}_2\text{Si}_2\text{O}_{14}$

Static Chirality

Spherical polarization analysis with CRYOPAD

$\Rightarrow$ A single possibility is selected (+1, -1)

Marty et al. PRL (2008)

Link with electrical polarization?
Use of polarized neutrons

Chirality and ferroelectricity

Example: $\text{Ba}_3\text{NbFe}_2\text{Si}_2\text{O}_{14}$

Dynamical Chirality

Chiral scattering

Totally chiral spin wave branch measured by longitudinal polarization analysis

Loire et al. submitted
Use of polarized neutrons

Chirality and ferroelectricity

\[ \exists P_E \text{ at } T_N \Rightarrow \text{proportional to the chirality domain unbalance} \]

**TbMnO\textsubscript{3}: electric control of spin helicity in a magnetic ferroelectric**

(polarized neutrons)

*Yamasaki et al. PRL (2007)*

**RbFe(MoO\textsubscript{4})\textsubscript{2}**

*Kenzelmann et al. PRL (2007)*

Triangular magnetic structure ⇒ 2 domains of triangular chiralities
Use of polarized neutrons

Domains in multiferroics/magnetoelectrics

Ni$_3$V$_2$O$_8$: coupled magnetic and ferroelectric domains
(longitudinal polarization analysis)

Cabrera, PRL 2009

YMn$_2$O$_5$: Electric field switching of antiferromagnetic domains
(spherical polarization analysis)

Radaelli, PRL 2008
Use of polarized neutrons

Domains in multiferroics/magnetoelectrics

Example: MnPS$_3$

T$_N$=78 K

Magnetic group 2'/m
Collinear antiferromagnet with $\vec{\tau}$=0
⇒ allows ferrotoroidicity
⇒ allows linear ME effect

$$F(\vec{E}, \vec{H}) = -\alpha_{ij} E_i H_j$$

with non diagonal tensor in the (a, b, c*) frame

$$\alpha = \begin{pmatrix} 0 & \alpha_{12} & 0 \\ \alpha_{21} & 0 & \alpha_{23} \\ 0 & \alpha_{32} & 0 \end{pmatrix}$$

cf. antiferromagnetic domains handling in linear magnetoelectric Cr$_2$O$_3$

Brown JPCM 1998

ESMF2010, L’Aquila
Use of polarized neutrons

Domains in multiferroics/magnetoelectrics

Example: MnPS$_3$

Antiferromagnetic/ferrorotoroidic domains manipulated in crossed E and H fields when cooled through $T_N$

Spherical polarization analysis

Ressouche et al. PRB (2010)
What I did not speak about:

- Debye-Waller factor omitted in the scattering expressions
- Incoherent versus coherent
- Diffuse scattering (short-range order)
- Reflectometry and small angle scattering: heterostructures
- Flipping ratio technique: magnetization density map

... 

Also possible to study films

Neutron diffraction study of hexagonal manganite YMnO$_3$, HoMnO$_3$, and ErMnO$_3$ epitaxial films

Gélard et al APL (2008)

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Thanks to B. Grenier and E. Ressouche (CEA-Grenoble, France)
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