

What do we see with neutrons in magnetism ?

L.C. Chapon

Institut Laue-Langevin, Grenoble, France

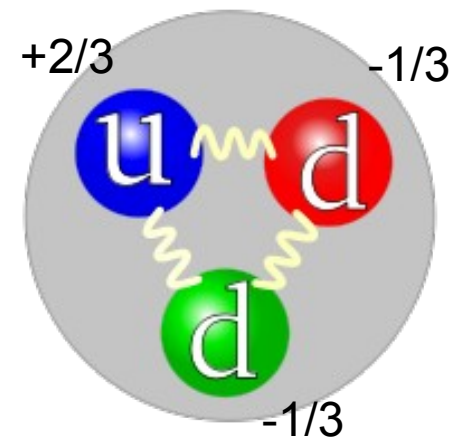
The neutron

- Non-charged particle $m_N = 1.67 \cdot 10^{-27} \text{ kg}$
 - Total angular momentum (« nuclear spin ») **$I=1/2$**
 - The magnetic moment is extremely small compared to the electron.
- > The interaction potential is small, Born approximation is valid

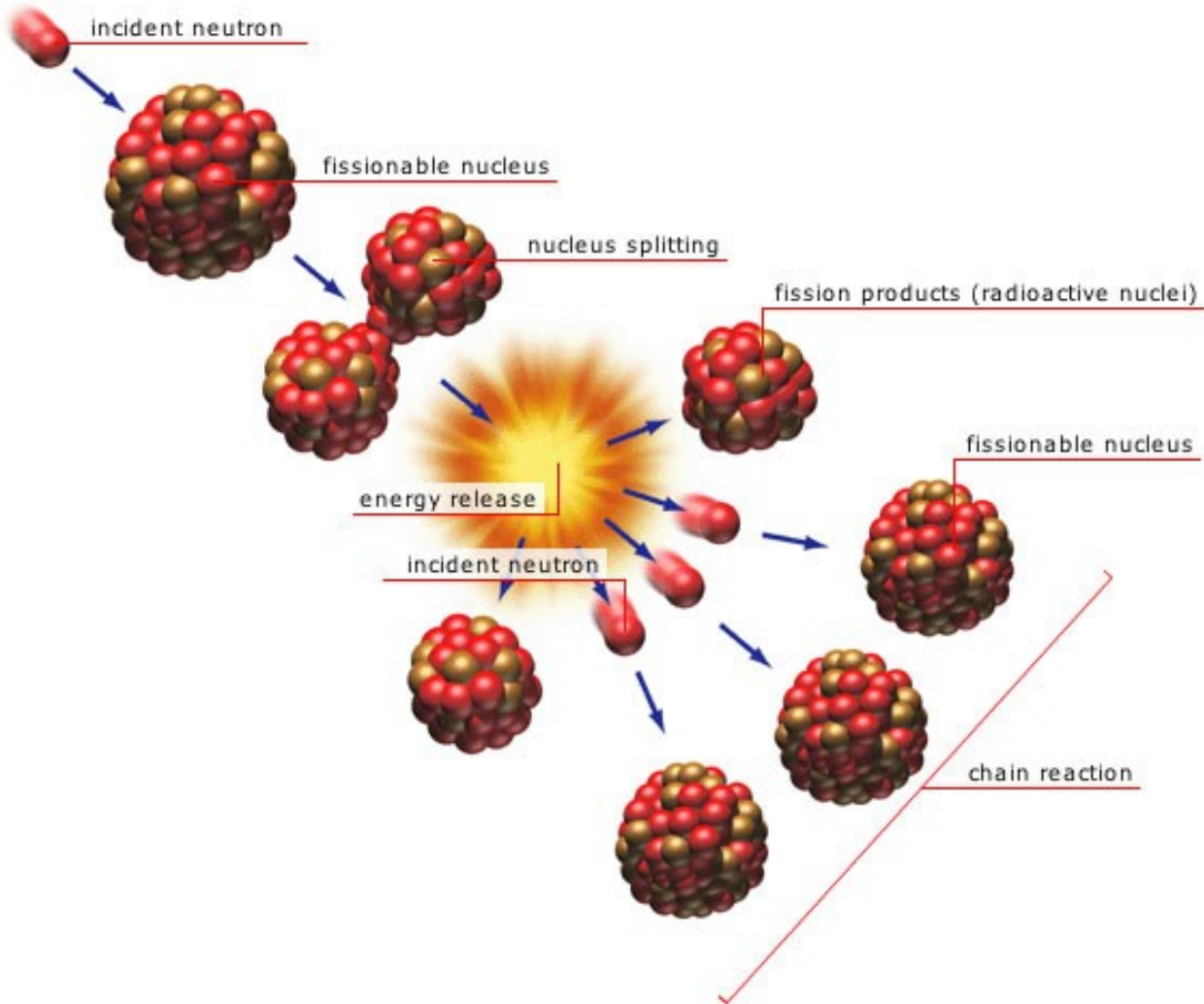
$$\mu = \gamma \mu_N \sigma \text{ with } \gamma = -1.913$$

$$\mu_N = \frac{e\hbar}{2m_p} = 5.05 \cdot 10^{-27} \text{ J.T}^{-1}$$

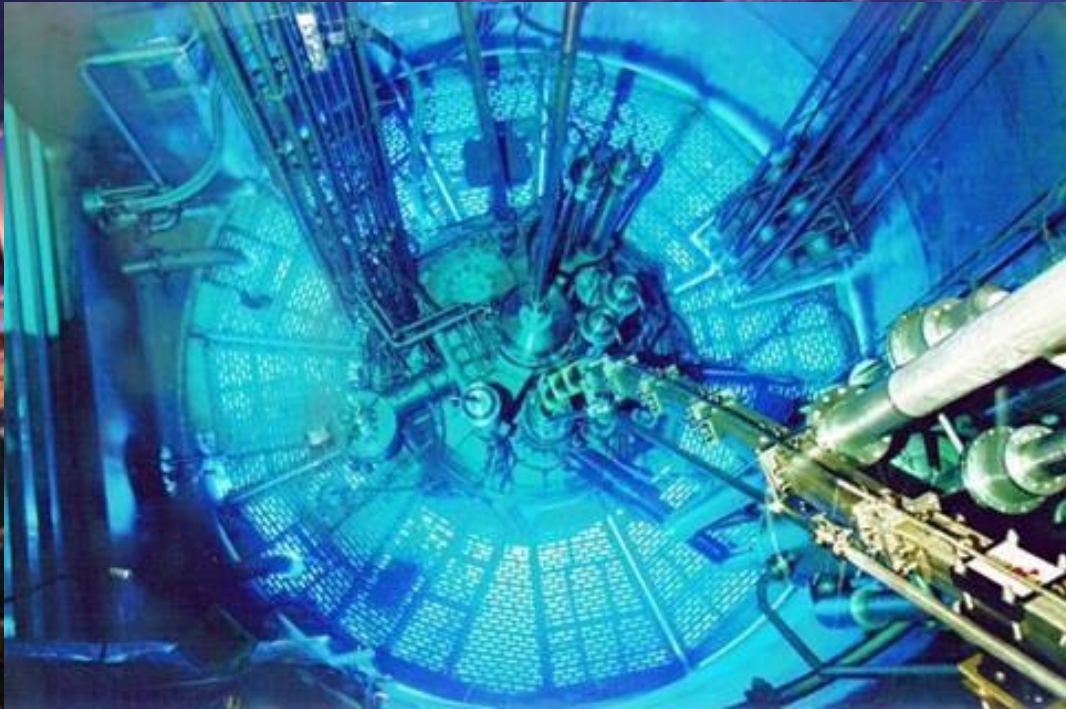
$$\mu_B = \frac{e\hbar}{2m_e} = 9.28 \cdot 10^{-24} \text{ J.T}^{-1}$$



Production : Nuclear fission



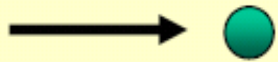
ILL, Grenoble, 58MW



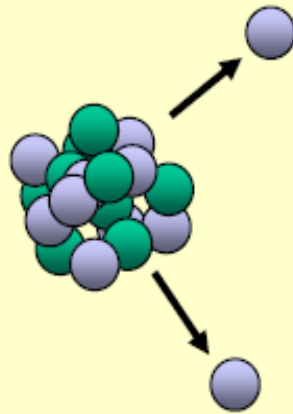
58MW 'Swimming pool' reactor
20K liquid D₂ moderator
2000K graphite moderator
1.5 x 10¹⁵ n/s/cm² – the most powerful
neutron source in the world

Production : spallation

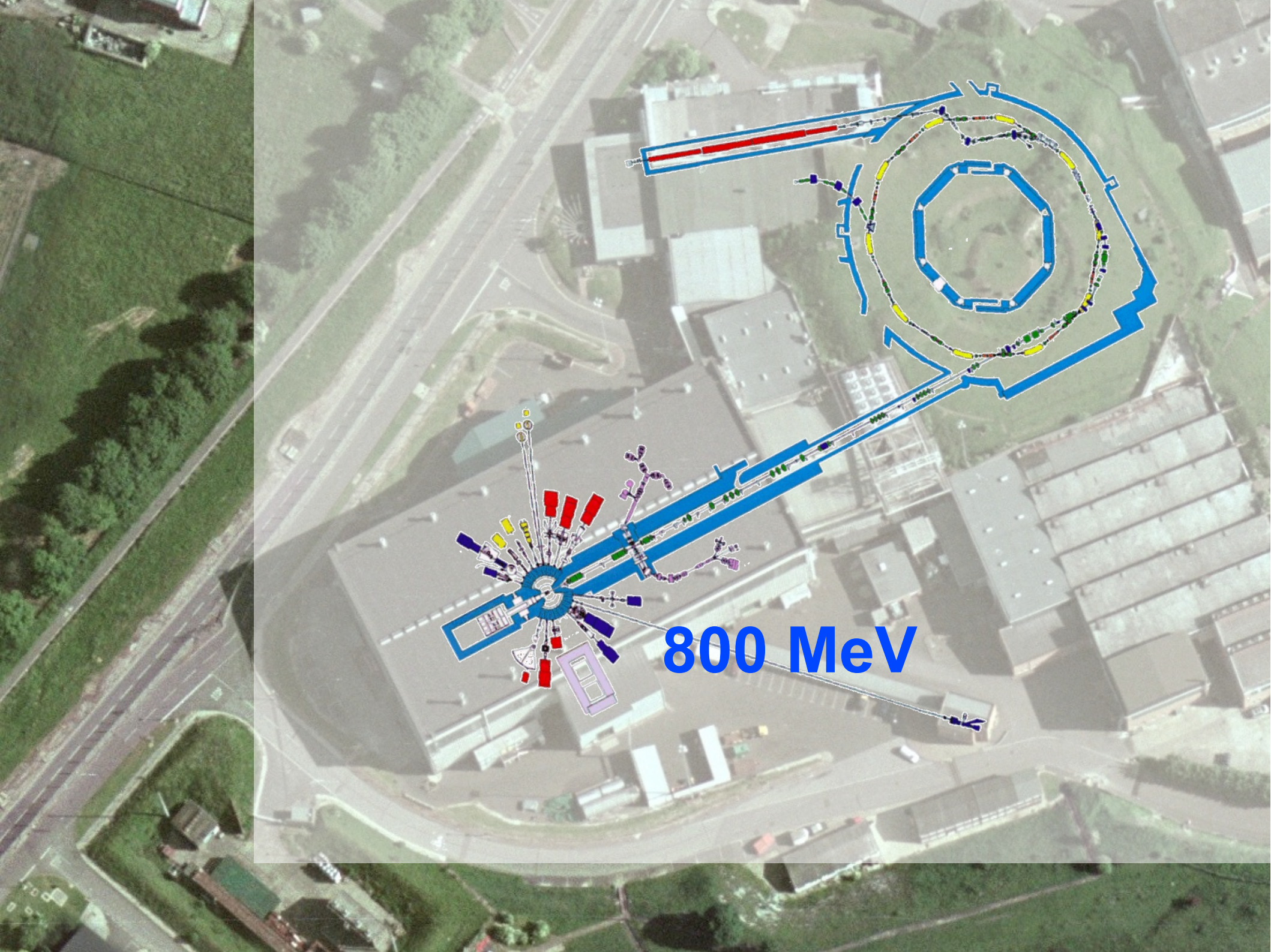
Fast proton



Heavy nucleus (Ta, U, Hg)

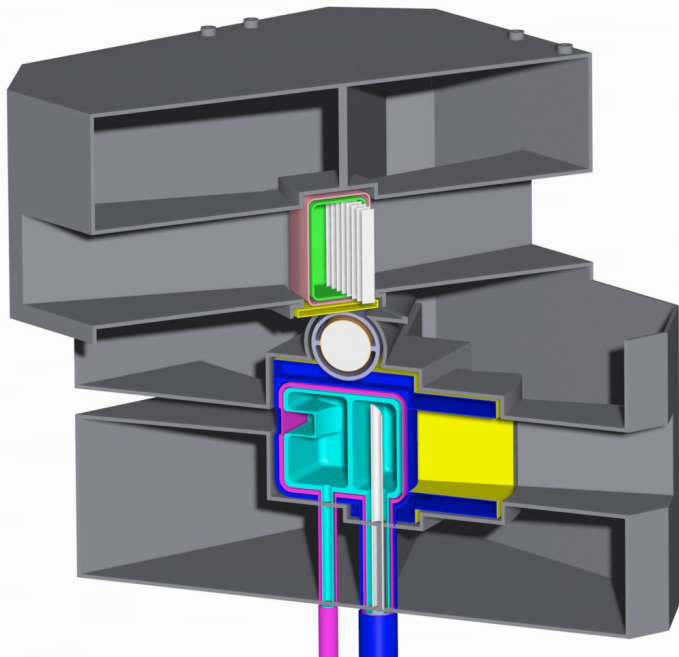
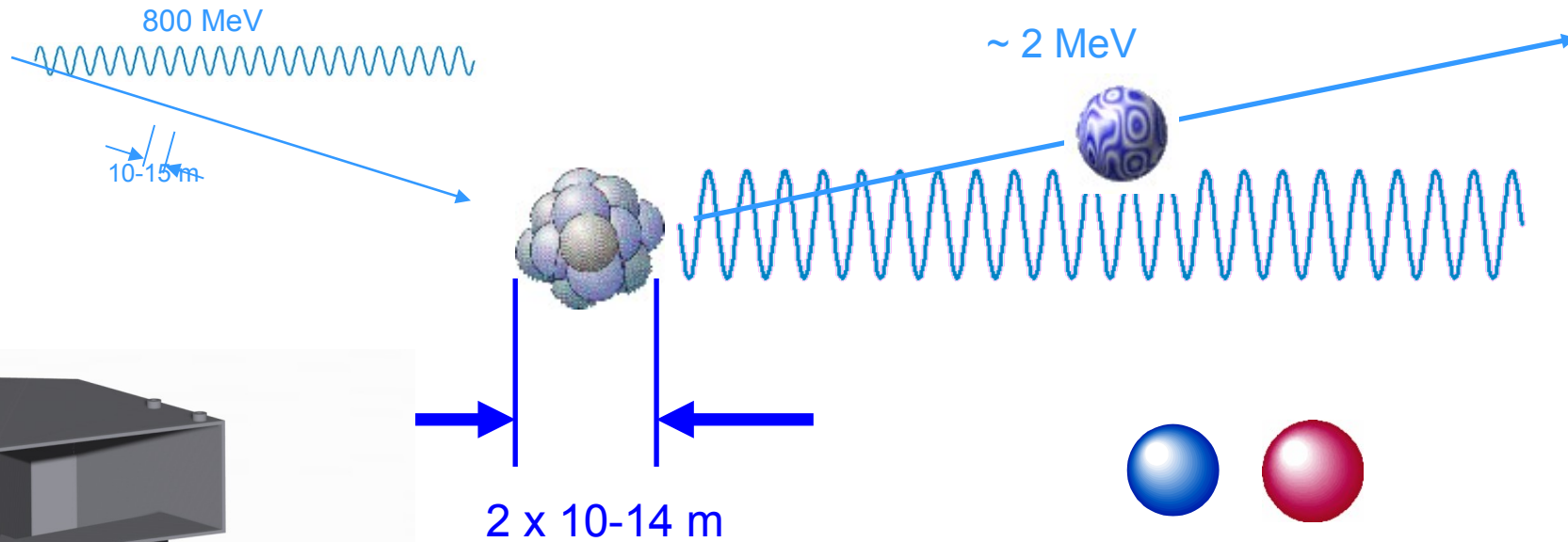


Fast Neutrons are slowed by collisions in a moderator (CH_4 , H_2O , D_2O)



800 MeV

Production : spallation




Neutrons slowed down by « moderators » :

- H_2
- CH_4
- H_2O

What do we see with neutrons in magnetism ?


The Nobel Prize in Physics 1994



Neutrons behave as particles and as waves

The Royal Swedish Academy of Sciences has awarded the 1994 Nobel Prize in Physics for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter.


Clifford G. Shull, MIT, Cambridge, Massachusetts, USA, winner one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.



S Shull made use of elastic scattering i.e. of neutrons which change direction without losing energy when they collide with atoms.


Because of the wave nature of neutrons, a diffraction pattern can be recorded which indicates where in the sample the atoms are situated. Even the placing of light elements such as hydrogen in metals, hydrides, or hydrogens, carbon and oxygen in organic substances can be determined.

The pattern also shows how atomic dipoles are oriented in magnetic materials, since neutrons are affected by magnetic forces. Shull also made use of this phenomenon in his neutron diffraction techniques.




Neutrons see more than X-rays

X-rays are scattered by electrons, neutrons by nuclei. This means that neutrons can see through light elements like hydrogen which are invisible to X-rays.




Neutrons reveal inner stresses

It has been predicted in an experiment that about 10% of the atoms in a crystal are displaced from their regular positions. Neutrons can see these displacements and measure the stresses.



Neutrons show what atoms remember

Of their order positions when they were randomly in solution. This is a very important property of some materials, especially polymers, which can be studied by neutron scattering.

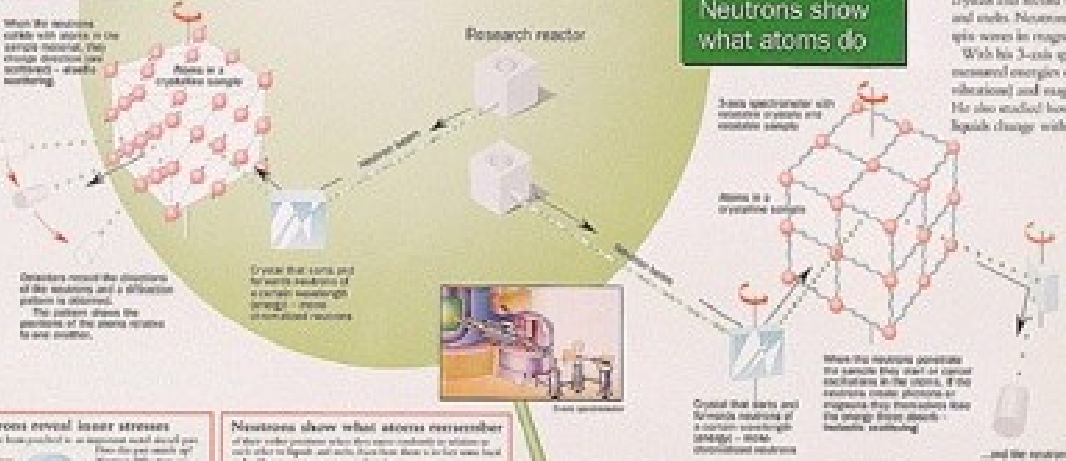


Neutrons reveal structure and dynamics

Neutrons show where atoms are

Neutrons bounce against atomic nuclei. They also react to the magnetism of the atoms.

Neutrons show what atoms do



When the neutrons collide with atoms in the sample material, they change direction and scatter - elastic scattering.


Crystal that acts as a diffraction grating.

Crystal that acts and diffracts neutrons of a certain wavelength (energy) - Bragg's diffraction condition.

When the neutrons penetrate the sample they start to interact with the atoms. If the neutrons excite phonons or magnons they transfer some of the energy they carry - inelastic scattering.

...and the neutrons that scattered in a detector.

Bertalan P. Brockhouse, McMaster University, Hamilton, Ontario, Canada, winner one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.



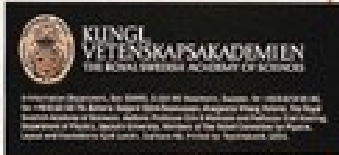
B Brockhouse made use of inelastic scattering i.e. of neutrons, which change both direction and energy when they collide with atoms. They then start to reveal atomic oscillations in crystals and record movements in liquids and solids. Neutrons can also interact with spin waves in magnets.

With his 3-axis spectrometer Brockhouse measured energies of phonons (atomic vibrations) and magnons (magnetic waves). He also studied how atomic structures in liquids change with time.

When it started Brockhouse and Shull made their pioneering contributions to the first nuclear reactors in the USA and Canada back in the 1940s and 1950s. It was then that the technique of the neutron became available for scientific research.

... from its applications.

Thousands of new experiments and new work in the many neutron research centres throughout the world. New and very advanced neutron scattering installations have been built and more are planned in Europe, the USA and Asia. In these super-collaborative installations we combine the strengths of new neutron superconductors, molecular magnets, complex surfaces of metals for analysis, neutron scattering, spin neutrons and the combination between the neutron and the whole progress of polymers.

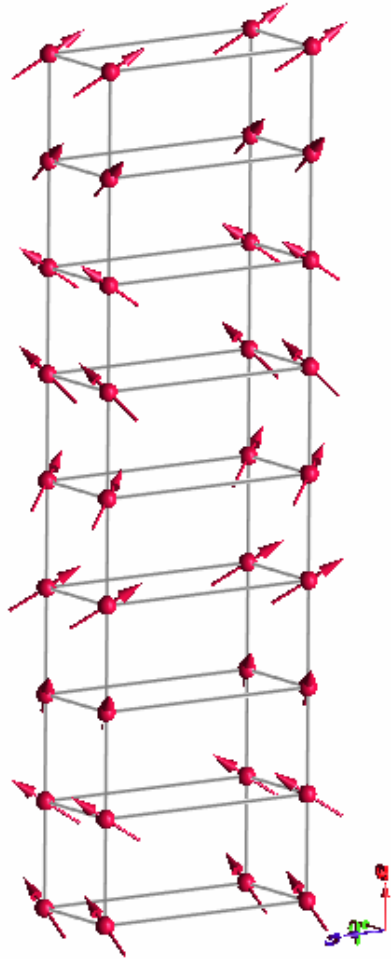


Further reading:

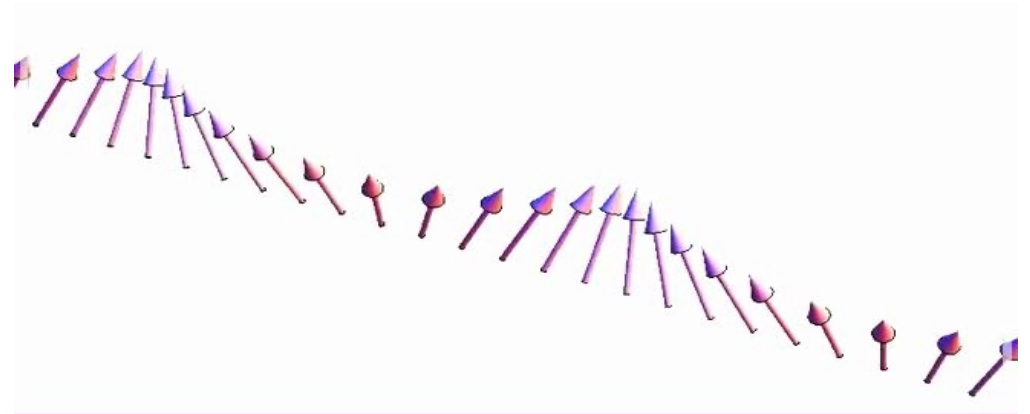
- 1. The Nobel Prize in Physics 1994. Stockholm: The Nobel Foundation, 1994. 100 pp. ISBN 91-7319-111-1.
- 2. Shull, C.G. and Brockhouse, B.P. (eds) Neutron Scattering: Techniques and Applications. New York: Academic Press, 1978. 400 pp. ISBN 0-12-080000-0.
- 3. Brockhouse, B.P. and Shull, C.G. (eds) Neutron Scattering: Techniques and Applications. New York: Academic Press, 1978. 400 pp. ISBN 0-12-080000-0.

What do we see with neutrons in magnetism ?

Static spin arrangements



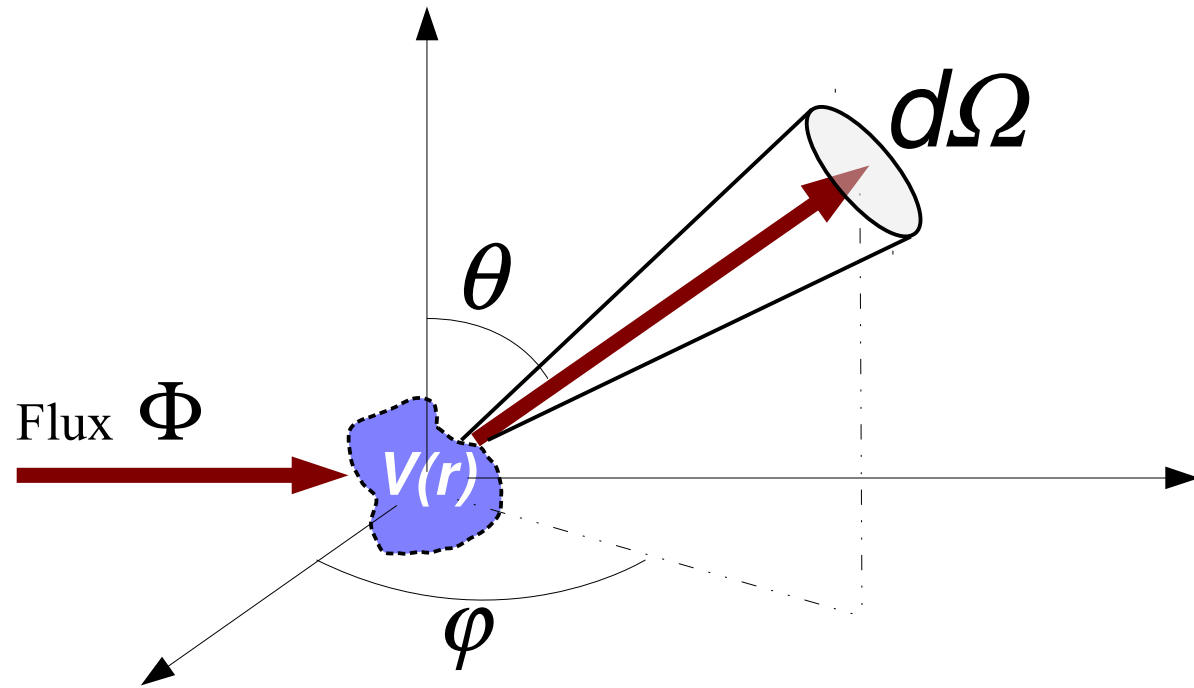
Local or collective excitations



Outline

- Reminder of scattering theory, neutron nuclear scattering
- Magnetic scattering theory:
 - Spin contribution
 - Orbital contribution
 - Density matrix formalism, non-polarized and polarized cases
 - Magnetic form factors
- Probing different magnetic states:
 - Magnetic Bragg scattering: Long-range ordered structures
 - Diffuse scattering, short-range correlations
 - Small angle scattering, skyrmions
 - Inelastic scattering, crystal field excitations, magnons

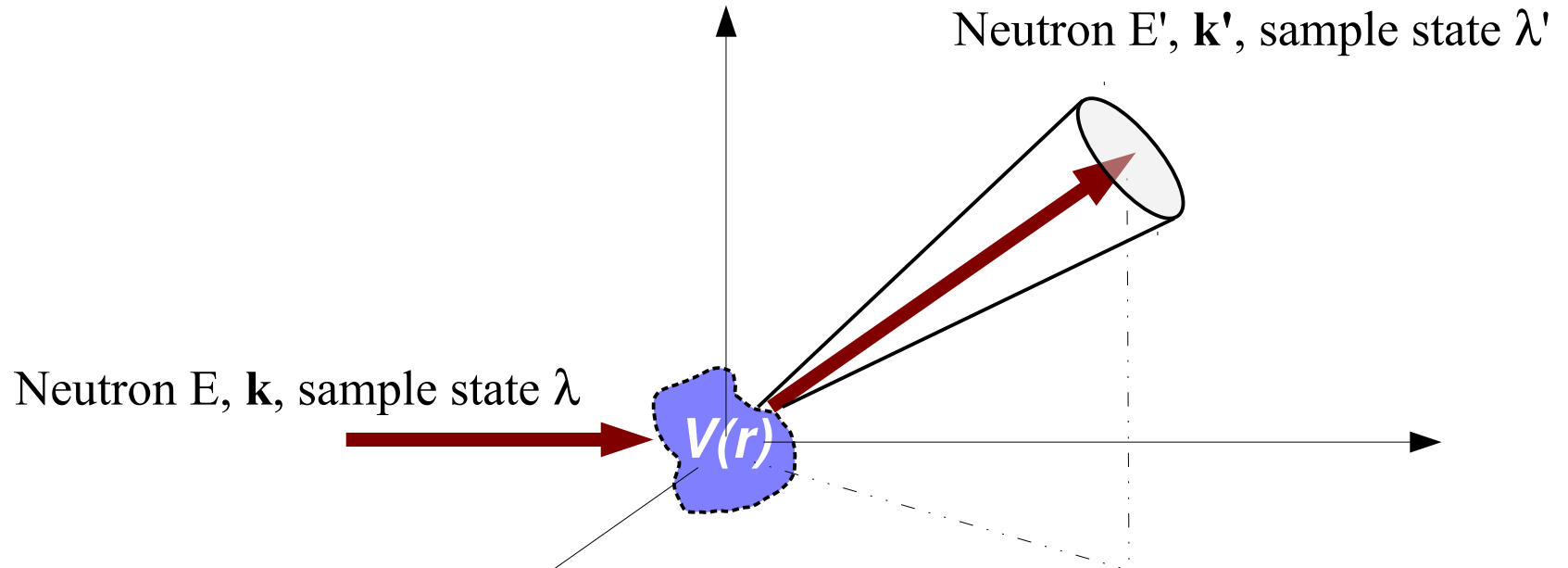
Scattering by a potential $V(r)$



$$\overbrace{dn}^{n \cdot s^{-1}} = \overbrace{\Phi}^{n \cdot cm^{-2} \cdot s^{-1}} \overbrace{d\Omega \sigma(\theta, \phi)}^{n \cdot u}$$

- σ has the dimension of a surface
- Usually in barns= 10^{-24} cm^2

Differential cross section, Fermi's Golden rule



$$\frac{d^2 \sigma}{d\Omega dE'} = \sum_{\lambda} p_{\lambda} \sum_{\lambda'} \left(\frac{d^2 \sigma}{d\Omega dE'} \right)_{\lambda \rightarrow \lambda'}$$

$$= \frac{k'}{k} \left(\frac{m}{2\pi\hbar^2} \right)^2 \sum_{\lambda} p_{\lambda} \sum_{\lambda'} |\langle k' \lambda' | V | k \lambda \rangle|^2 \delta(E_{\lambda} - E_{\lambda'} + E - E')$$

Born approximation

- In the quantum mechanical treatment of scattering by a central potential, the stationary states $\varphi(\mathbf{r})$ verify:

$$[\Delta + k^2]\varphi(\mathbf{r}) = \frac{2\mu}{\hbar^2} V(\mathbf{r})\varphi(\mathbf{r})$$

- In the integral equation of scattering, the stationary wave-function is written :

$$v_k^{scat}(\mathbf{r}) = e^{i\mathbf{k}_i \cdot \mathbf{r}} + \frac{2\mu}{\hbar^2} \int G_+(\mathbf{r} - \mathbf{r}') V(\mathbf{r}') v_k^{scat}(\mathbf{r}') d^3 r'$$

- One can expand iteratively this expression (Born expansion).

If the potential is weak, one can limit the expansion to the first term, this is the first Born approximation. In this case the scattering cross section (amplitude) is related to the Fourier transform of the potential function.

$$\sigma_k(\theta, \phi) = \frac{m^2}{4\pi^2 \hbar^2} \left| \int V(\mathbf{r}) e^{i\mathbf{q} \cdot \mathbf{r}} d^3 r \right|^2$$

Quantum Mechanics,
Claude Cohen-Tannoudji et al., Vol 2, Chapt 8

The phase problem

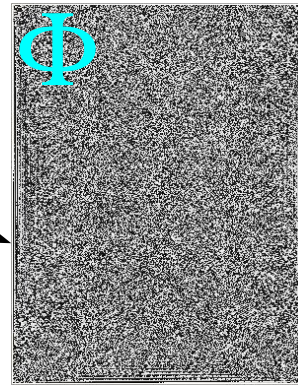
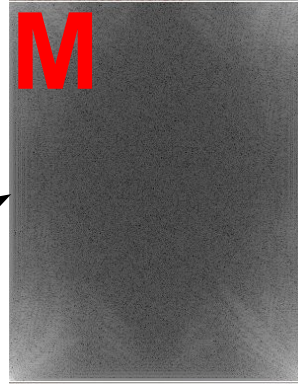
Loss of information in a physical measurement

$$F(u, v) = M(u, v) e^{i\Phi}$$

$$F(u, v) = \frac{1}{N_x N_y} \sum_x \sum_y f(x, y) e^{(-2\pi i(\frac{xu}{N_x} + \frac{yv}{N_y}))}$$



James Chadwick

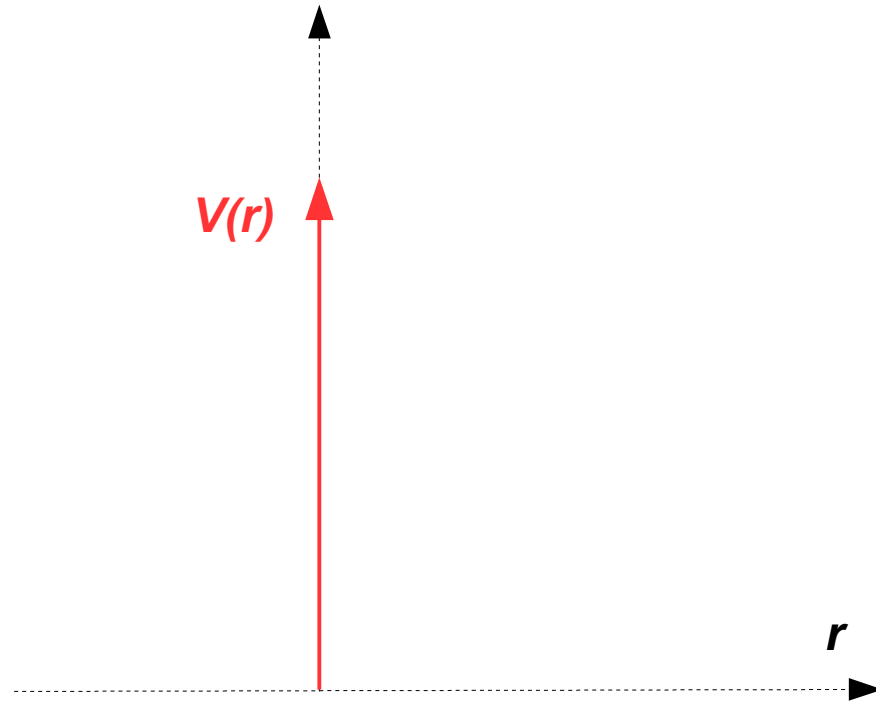


$$f'(x, y) = \frac{1}{N_u N_v} \sum_u \sum_v M(u, v) e^{(2\pi i(\frac{xu}{N} + \frac{yv}{N}))}$$



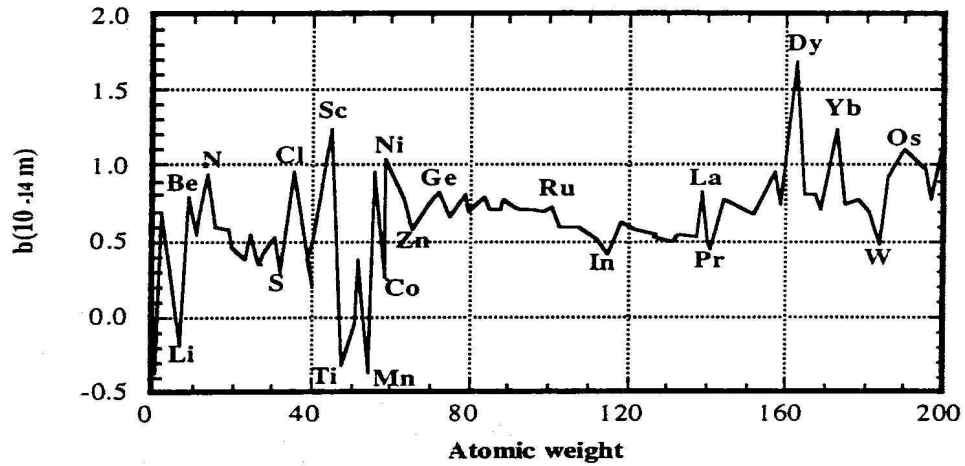
Nuclear scattering

- Nuclear scattering mediated by the strong force, extremely short range ($\text{fm}=1.10^{-15} \text{ m}$).
- Neutron wavelength much larger ($\text{\AA}=1.10^{-10}\text{m}$), can not probe internal nuclear structure: scattering is isotropic.
- The interaction between the neutron and the atomic nucleus is represented by the Fermi pseudo-potential, a **scalar field** that is zero except very close to the nucleus (**δ function**).

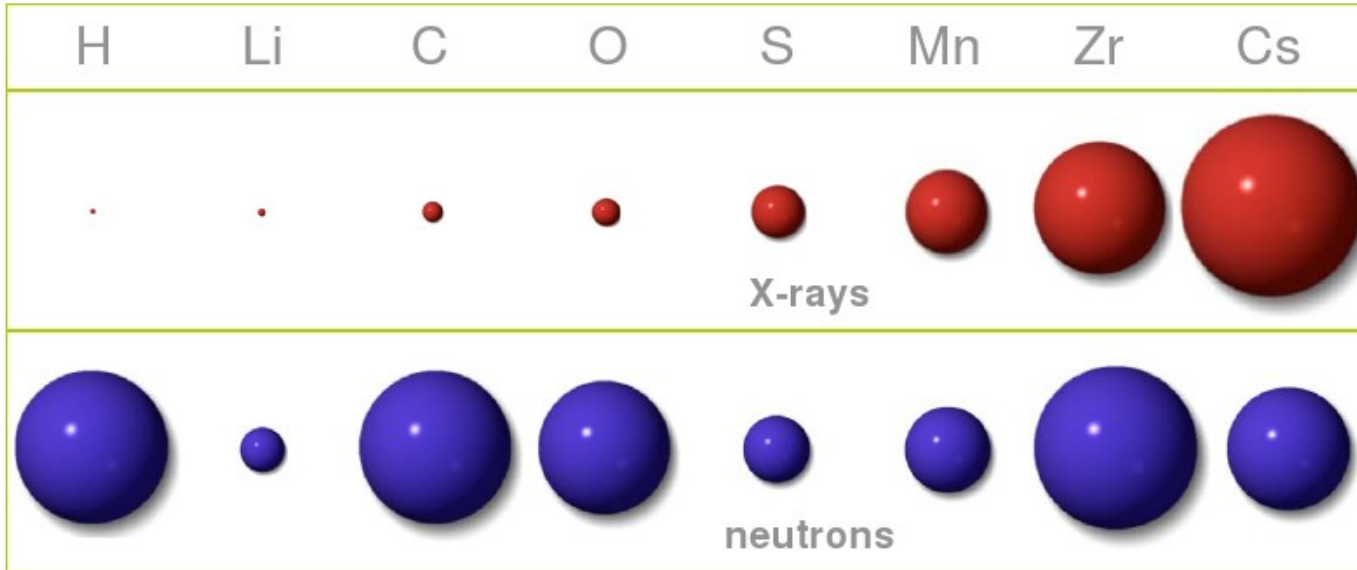


$$V(\vec{r}) = \frac{2\pi\hbar^2}{m_r} a \delta^3(\vec{r})$$

Scattering lengths



Typically a few fm



X-ray vs Neutron

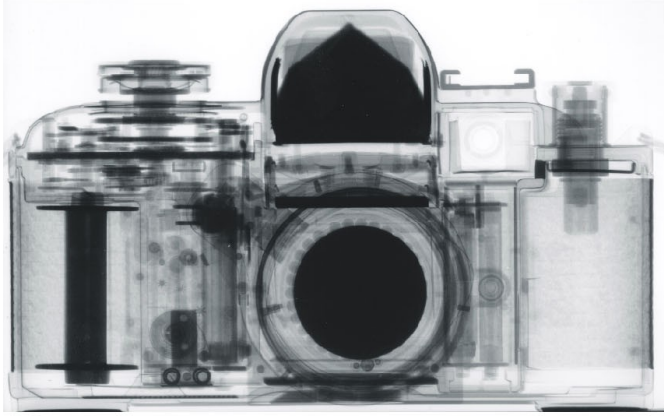


Fig. a: Neutron radiography of a camera

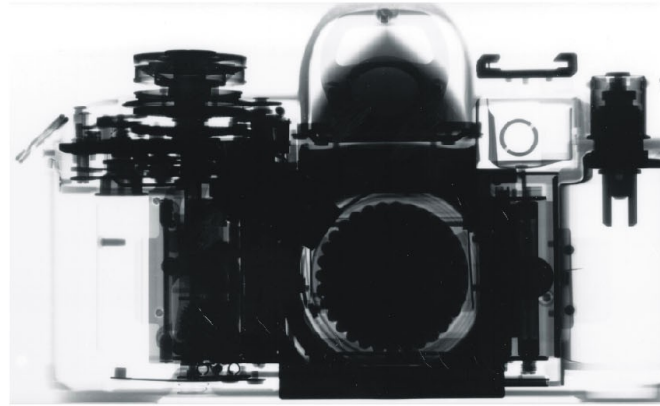
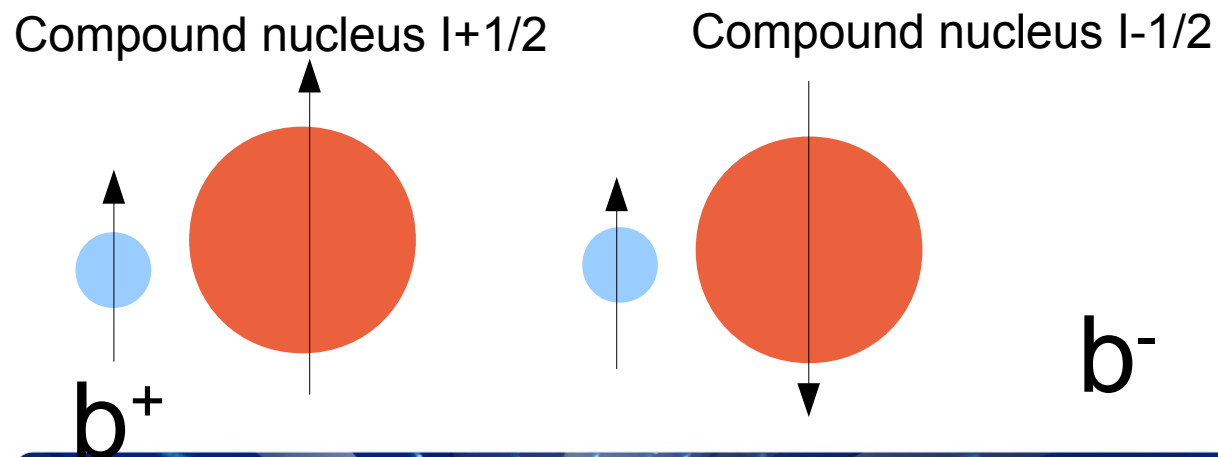


Fig. b: Radiographic image of a camera made X-rays

Also: - b depends on the isotope
 - b depends on the spin states of the neutron and nucleus



$$b_{coh} = \bar{b}$$

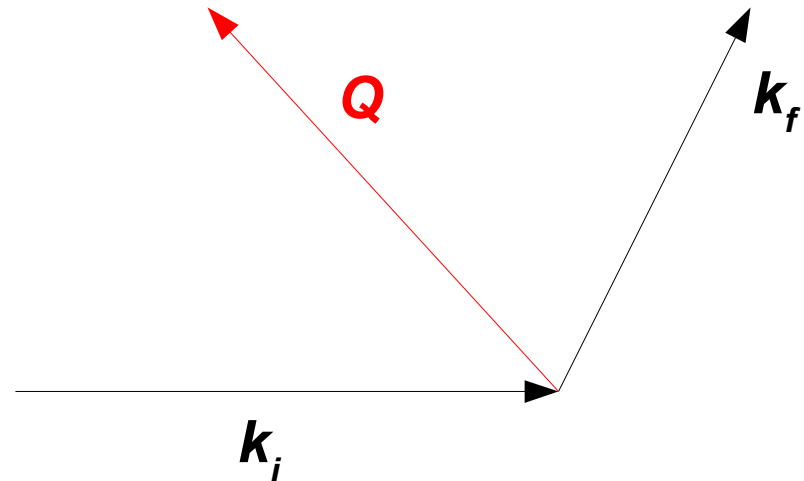
$$b_{inc} = \sqrt{b^2 - \bar{b}^2}$$

Magnetic cross section

For elastic neutron magnetic scattering, one needs to evaluate (in the Born approximation), the cross section:

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\sigma,\lambda\rightarrow\sigma',\lambda'} = \frac{k'}{k} \left(\frac{m}{2\pi\hbar}\right)^2 |\langle \mathbf{k}', \sigma' | V_m | \mathbf{k}, \sigma \rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

- σ initial spin-state of the neutron
- σ' final spin-state of the neutron
- \mathbf{k} incident wave-vector
- \mathbf{k}' scattered wave-vector
- \mathbf{Q} momentum transfer
- V_m magnetic interaction potential



Magnetic interaction (spin)

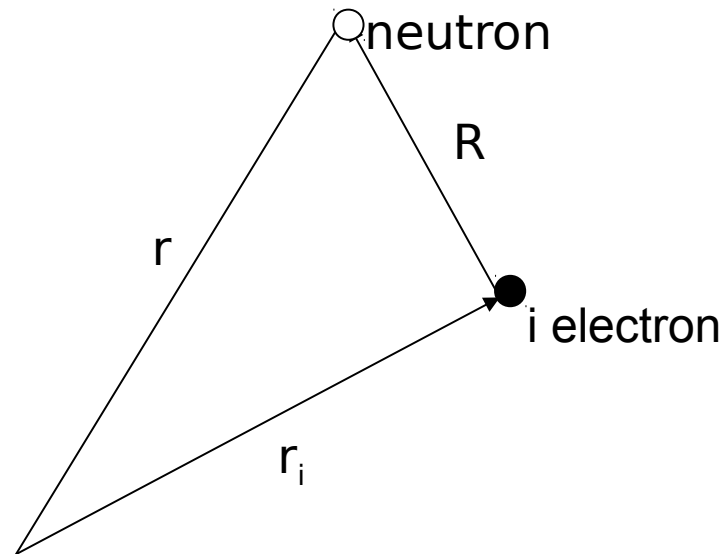
Considering a single unpaired electron:

$$\mathbf{A}(\mathbf{R}) = \frac{\mu_0}{4\pi} \frac{\boldsymbol{\mu}_e \times \hat{\mathbf{R}}}{R^2}$$

$$\mathbf{B} = \nabla \times \mathbf{A}$$

$$V = -\boldsymbol{\mu} \cdot \mathbf{B}$$

$$V = -\gamma \mu_N 2 \mu_B \frac{\mu_0}{4\pi} \boldsymbol{\sigma} \cdot \nabla \times \frac{\mathbf{s} \times \hat{\mathbf{R}}}{R^2}$$



$$V_S = \gamma \mu_N 2 \mu_B \frac{\mu_0}{4\pi} \boldsymbol{\sigma} \cdot \nabla \times \mathbf{s} \times \nabla \frac{1}{|R|} = cste. \boldsymbol{\sigma} \cdot \mathbf{W}_S$$

Introduction to the Theory of Thermal Neutron Scattering
G. L. Squires, Dover Publications

Magnetic interaction (spin)

$$\nabla \times \mathbf{s} \times \nabla \frac{1}{|R|} = \frac{1}{2\pi^2} \int \frac{1}{\mathbf{x}^2} \nabla \times \mathbf{s} \times \nabla e^{i\mathbf{x}R} d\mathbf{x} = \frac{1}{2\pi^2} \int (\hat{\mathbf{x}} \times \mathbf{s} \times \hat{\mathbf{x}}) e^{i\mathbf{x}R} d\mathbf{x}$$

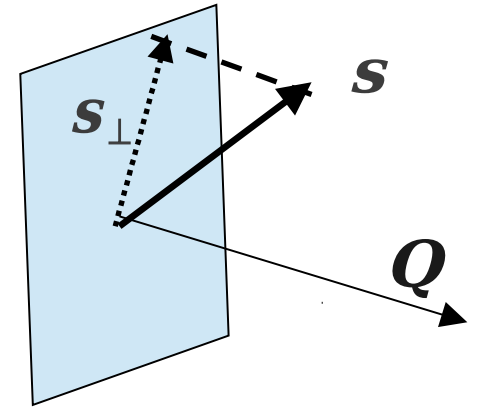
$$\left\langle \mathbf{k}' \left| \nabla \times \mathbf{s} \times \nabla \frac{1}{|R|} \right| \mathbf{k} \right\rangle = \frac{1}{2\pi^2} \int e^{i\mathbf{Q}r} \int (\hat{\mathbf{x}} \times \mathbf{s} \times \hat{\mathbf{x}}) e^{i\mathbf{x}R} d\mathbf{x} d\mathbf{r} = 4\pi \hat{\mathbf{Q}} \times \mathbf{s} \times \hat{\mathbf{Q}} \cdot e^{i\mathbf{Q}r_i}$$

This quantity is the **projection of \mathbf{s} perpendicular to \mathbf{Q}** :

$$\mathbf{s}_{\perp}(\mathbf{Q}) = \hat{\mathbf{Q}} \times \mathbf{s} \times \hat{\mathbf{Q}}$$

$$\left\langle \mathbf{k}' \left| W_s \right| \mathbf{k} \right\rangle = 4\pi \hat{\mathbf{Q}} \times \mathbf{s} \times \hat{\mathbf{Q}} \cdot e^{i\mathbf{Q} \cdot \mathbf{r}_i}$$

Introduction to the Theory of Thermal Neutron Scattering
G. L. Squires, Dover Publications



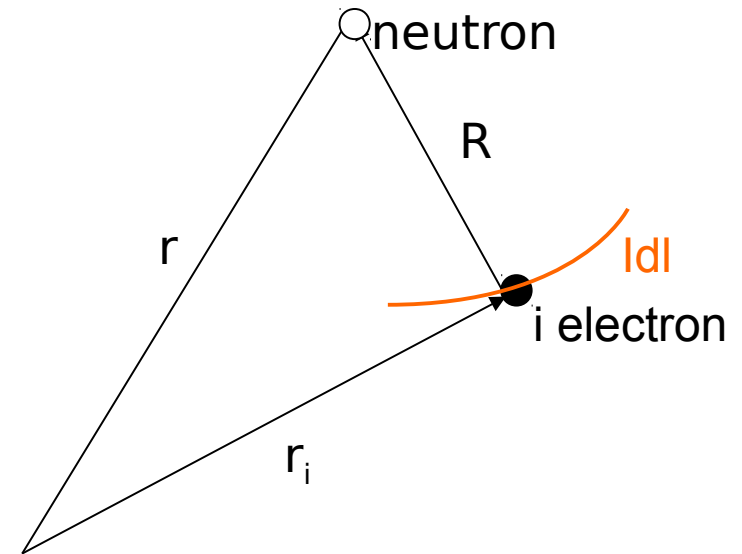
Magnetic interaction (orbital)

$$\mathbf{B}(\mathbf{R}) = \frac{\mu_0}{4\pi} \frac{I d\mathbf{l} \times \hat{\mathbf{R}}}{R^2} = \frac{\mu_0 e}{4\pi m_N} \frac{\mathbf{p}_i \times \hat{\mathbf{R}}}{R^2} = \frac{2\mu_0 \mu_B}{4\pi \hbar} \frac{\mathbf{p}_i \times \hat{\mathbf{R}}}{R^2}$$

$$V_L = \gamma \mu_N 2 \mu_B \frac{\mu_0}{4\pi \hbar} \boldsymbol{\sigma} \cdot \mathbf{p}_i \times \nabla \frac{1}{|\mathbf{R}|} = \text{cste} \boldsymbol{\sigma} \cdot \mathbf{W}_L$$

$$\mathbf{W}_L = \frac{1}{\hbar} \frac{\mathbf{p}_i \times \hat{\mathbf{R}}}{R^2}$$

$$\langle \mathbf{k}' | W_L | \mathbf{k} \rangle = \frac{4\pi i}{\hbar Q} \mathbf{p}_i \times \hat{\mathbf{Q}} e^{i\mathbf{Q} \cdot \mathbf{r}_i}$$



Use the Fourier transform:

$$\int \frac{\hat{\mathbf{R}}}{R^2} e^{i\mathbf{K} \cdot \mathbf{R}} = 4\pi i \frac{\mathbf{K}}{K}$$

Magnetic interaction strength

Collecting the **pre-factors**

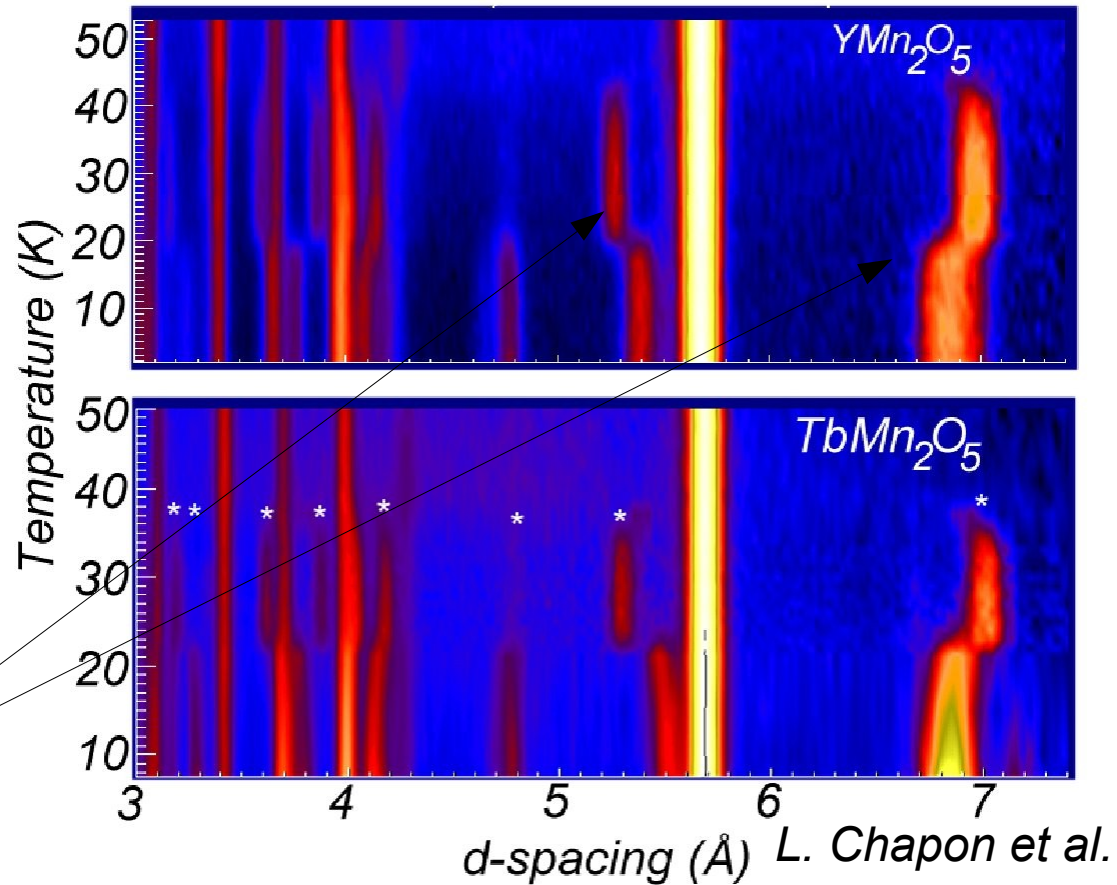
$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{k,\sigma \rightarrow k',\sigma'} = \frac{k'}{k} \left(\frac{m}{2\pi\hbar}\right)^2 |\langle \mathbf{k}', \sigma' | V_S + V_L | \mathbf{k}, \sigma \rangle|^2 \delta \dots = \left(\frac{m}{2\pi\hbar} 2\gamma\mu_N\mu_B\mu_0\right)^2 \frac{k'}{k} \dots$$

$$\left(\frac{m}{2\pi\hbar} 2\gamma\mu_N\mu_B\mu_0\right)^2 = (\gamma r_0)^2$$

r_0 : free electron radius = $2.8 \cdot 10^{-15}$ m

Magnetic scattering length **is comparable in magnitude to nuclear scattering !**

Magnetic scattering



(Spin + orbital) magnetic scattering

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\sigma,\lambda\rightarrow\sigma',\lambda'} = \frac{k'}{k} \left(\frac{m}{2\pi\hbar}\right)^2 |\langle \mathbf{k}', \sigma', \lambda' | V_m | \mathbf{k}, \sigma, \lambda \rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

Defining:

$$\mathbf{M}_\perp = \sum_i e^{i\mathbf{Q}\cdot\mathbf{r}_i} (\hat{\mathbf{Q}} \times \mathbf{s}_i \times \hat{\mathbf{Q}} + \frac{i}{\hbar Q} \mathbf{p}_i \times \hat{\mathbf{Q}})$$

The spin and orbital part of \mathbf{M}_\perp are the transverse components of the Fourier transform of the spin and orbital magnetization density

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\sigma,\lambda\rightarrow\sigma',\lambda'} = \frac{k'}{k} (\gamma r_0)^2 |\langle \lambda', \sigma' | \sigma \cdot \mathbf{M}_\perp | \lambda, \sigma \rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

Density matrix formalism

Calculations are enormously simplified by using the **density-matrix** formalism to describe mixed-states, incomplete polarization of beam, analysis....

Suppose a quantum system in a mixed state, i.e. probability p_1 to be in state 1, probability p_i to be in state i etc....

One defines a density operator $\hat{\rho} = \sum_i p_i |\psi_i\rangle \langle \psi_i|$

Chosen an orthonormal basis, $|u_n\rangle$, one can define a density matrix whose elements are

$$\rho_{mn} = \langle u_m | \hat{\rho} | u_n \rangle$$

The expectation value of an operator A is simply:

$$\langle A \rangle = \text{tr}(\rho A)$$

Fano, U.

Description of States in Quantum Mechanics by Density Matrix and Operator Techniques Rev. Mod. Phys., American Physical Society, 1957, 29, 74-93

Density matrix formalism - Neutron spin states

Neutron, a spin $\frac{1}{2}$ particle

$$|\uparrow\rangle = |1/2, 1/2\rangle$$

$$|\downarrow\rangle = |1/2, -1/2\rangle$$

Spin operators:

$$\hat{S}_+|\uparrow\rangle = 0 \quad \hat{S}_+|\downarrow\rangle = |\uparrow\rangle \quad \hat{S}_z|\uparrow\rangle = 1/2|\uparrow\rangle \quad \hat{S}_+ = \hat{S}_x + i\hat{S}_y$$

$$\hat{S}_-|\uparrow\rangle = |\downarrow\rangle \quad \hat{S}_-|\downarrow\rangle = 0 \quad \hat{S}_z|\downarrow\rangle = -1/2|\downarrow\rangle \quad \hat{S}_- = \hat{S}_x - i\hat{S}_y$$

Pauli spin operators and matrices:

$$\hat{\sigma}_x = 2\hat{S}_x, \hat{\sigma}_y = 2\hat{S}_y, \hat{\sigma}_z = 2\hat{S}_z$$

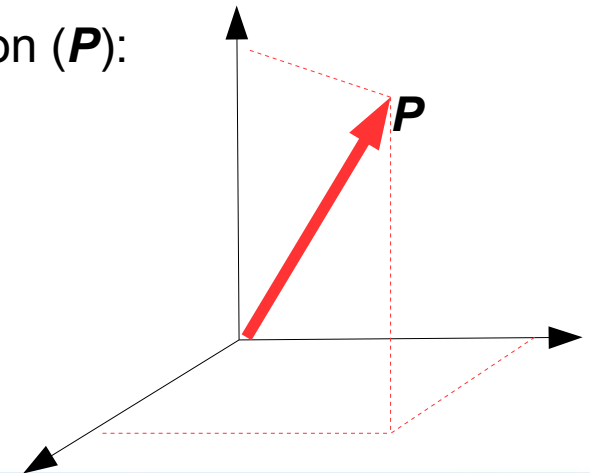
$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$$

$$\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$$

$$\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

Density matrix representing the incident neutron beam polarization (\mathbf{P}):

$$\hat{\rho} = \frac{1}{2} (I + \mathbf{P} \cdot \hat{\boldsymbol{\sigma}}) = \frac{1}{2} (I + P_x \cdot \hat{\sigma}_x + P_y \cdot \hat{\sigma}_y + P_z \cdot \hat{\sigma}_z)$$



Magnetic interaction(elastic case)

A very easy way to averaging over all spin states:

Scattered intensity:
$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \text{tr} [(\mathbf{M}_\perp \cdot \boldsymbol{\sigma}) \rho (\mathbf{M}_\perp \cdot \boldsymbol{\sigma})^+]$$

Final polarization:
$$\mathbf{P}_f \cdot \frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \text{tr} [\boldsymbol{\sigma}_f (\mathbf{M}_\perp \cdot \boldsymbol{\sigma}) \rho (\mathbf{M}_\perp \cdot \boldsymbol{\sigma})^+]$$

Blume, M.

Polarization Effects in the Magnetic Elastic Scattering of Slow Neutrons Phys. Rev., American Physical Society, 1963, 130, 1670-1676

Magnetic interaction (unpolarized)

In the case of unpolarized neutron beam (and no analysis), there are no interference terms between nuclear and magnetic scattering. The scattered intensity is simply the square of the amplitude derived before.

For example if magnetic structure with $k=0$,

$$\frac{d\sigma}{d\Omega} = F_N(\mathbf{Q})^2 + |\mathbf{M}_\perp(\mathbf{Q})|^2$$

Magnetic interaction (polarized)

$$I = \frac{1}{2} \text{tr} [(N\mathbb{1} + \mathbf{M}_\perp \cdot \boldsymbol{\sigma}) (\mathbb{1} + \mathbf{P}_i \cdot \boldsymbol{\sigma}) (N^*\mathbb{1} + \mathbf{M}_\perp^* \cdot \boldsymbol{\sigma})]$$

$$I = NN^* + \mathbf{M}_\perp \cdot \mathbf{M}_\perp^* + \mathbf{P}_i \cdot (N\mathbf{M}_\perp^* + N^*\mathbf{M}_\perp) + i\mathbf{P}_i (\mathbf{M}_\perp^* \times \mathbf{M}_\perp)$$

$$P_f \cdot I = \frac{1}{2} \text{tr} [\boldsymbol{\sigma} (N\mathbb{1} + \mathbf{M}_\perp \cdot \boldsymbol{\sigma}) (\mathbb{1} + \mathbf{P}_i \cdot \boldsymbol{\sigma}) (N^*\mathbb{1} + \mathbf{M}_\perp^* \cdot \boldsymbol{\sigma})] \quad (15)$$

$$P_f I = \mathbf{P}_i (NN^* - \mathbf{M}_\perp \cdot \mathbf{M}_\perp^*) + (\mathbf{P}_i \cdot \mathbf{M}_\perp) \mathbf{M}_\perp^* + (\mathbf{P}_i \cdot \mathbf{M}_\perp^*) \mathbf{M}_\perp - i(N\mathbf{M}_\perp^* - N^*\mathbf{M}_\perp) \times \mathbf{P}_i$$

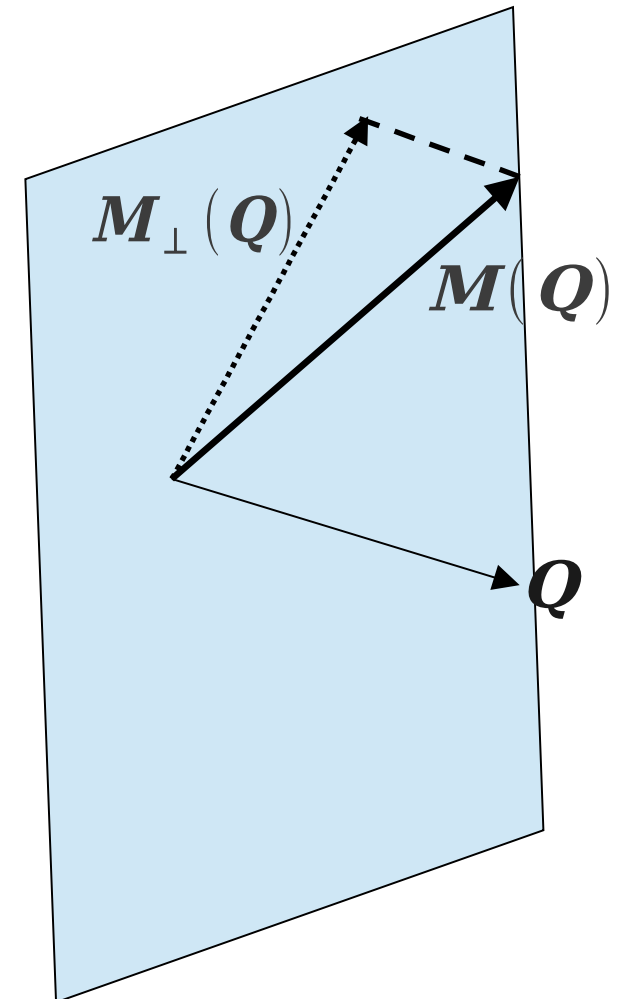
$$+ (N\mathbf{M}_\perp^* + N^*\mathbf{M}_\perp) - i(\mathbf{M}_\perp^* \times \mathbf{M}_\perp)$$

Blume, M.

Polarization Effects in the Magnetic Elastic Scattering of Slow Neutrons Phys. Rev., American Physical Society, 1963, 130, 1670-1676

Magnetic “extinction”

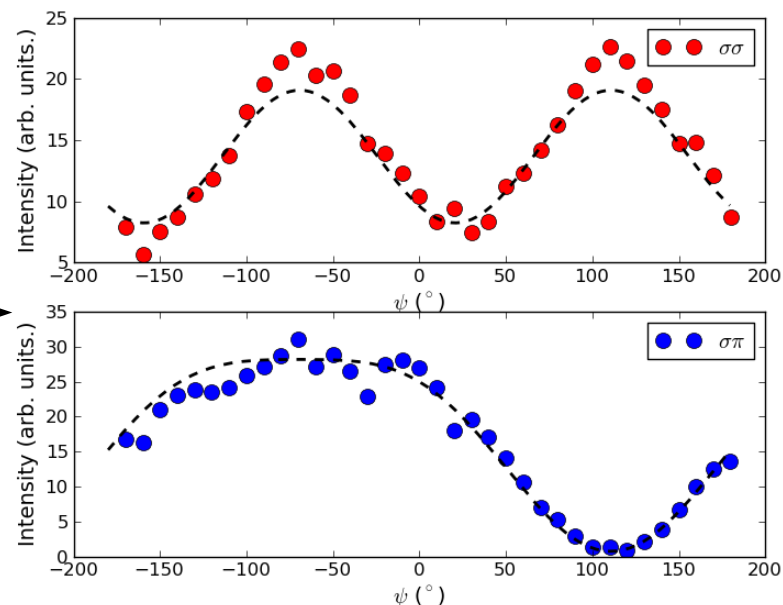
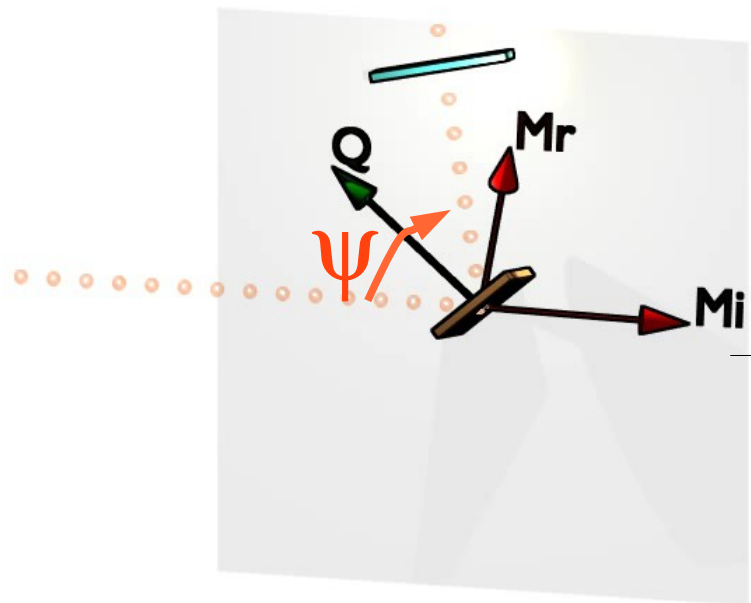
- From the projection operation emerges a very important extinction condition (if M parallel to Q , scattering is null)
- However, the only directional information about $\mathbf{M}(\mathbf{Q})$ comes from the projection operation, so great loss of information from this projection. We are **only** sensitive to the norm of the interaction vector.
- We will see that using polarized neutrons (3D polarimetry) allows to access directly the direction information (and phase).



Magnetic “extinction” (comparison with X-ray)

In non-resonant X-ray magnetic scattering, the cross-section depends upon projections on \mathbf{k}_i , \mathbf{k}_f

Consequence: signal depends on the azimuthal angle

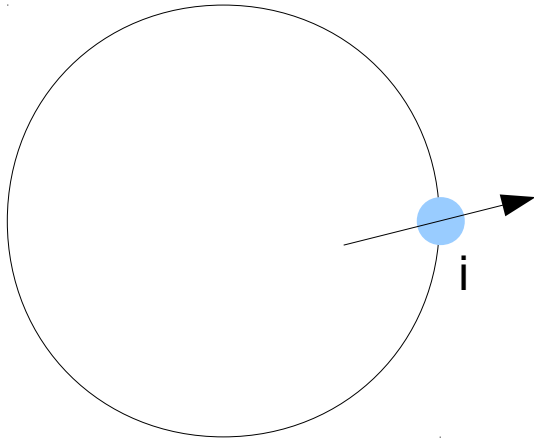


Scattering amplitude ($\sigma\sigma$): $-i. \mathbf{M}(\mathbf{Q}). \hat{\mathbf{k}}_i \times \hat{\mathbf{k}}_f$

Scattering amplitude ($\pi\sigma$): $-2.i.\sin^2(\theta). \mathbf{M}(\mathbf{Q}). \hat{\mathbf{k}}_i$

Unit-cell magnetic structure factor

Need to take into account the spatial distribution of the electron-spins and sum over all magnetic sites in the unit-cell:



$$M(\mathbf{Q}) = \sum_j f_j(\mathbf{Q}) \cdot \mathbf{m}_j \cdot T_j(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{r}_j}$$

Magnetic structure factor (complex vector)

- Magnetic form factor: $f_j(\mathbf{Q}) = \frac{1}{|m_j|} \int m_j(\mathbf{R}) e^{i\mathbf{Q} \cdot \mathbf{R}} d\mathbf{R}$

- Thermal parameter (Debye-Waller factor): $T_j(\mathbf{Q})$

$$M_{\perp}(\mathbf{Q}) = \hat{\mathbf{Q}} \times M(\mathbf{Q}) \times \hat{\mathbf{Q}} = M(\mathbf{Q}) - (M(\mathbf{Q}) \cdot \hat{\mathbf{Q}}) \cdot \hat{\mathbf{Q}}$$

Magnetic interaction vector (complex vector)

Magnetic form factor

$$f_j(\mathbf{Q}) = \frac{1}{|m_j|} \int m_j(\mathbf{R}) e^{i\mathbf{Q}\cdot\mathbf{R}} d\mathbf{R}$$

In the most general case, the magnetization distribution is non-spherical:

$$\mathbf{m}(\mathbf{R}) = \sum_l R_l^2(R) \sum_{m,p} \beta_l^{m,p} \cdot y_l^{m,p}(\hat{\mathbf{R}})$$

Using the addition theorem:

$$f(\mathbf{Q}) = 4\pi \sum_l i^L \langle j_L(\mathbf{Q}\cdot\mathbf{R}) \rangle \sum_{m,p} \beta_l^{m,p} \cdot y_l^{m,p}(\hat{\mathbf{Q}})$$

with:

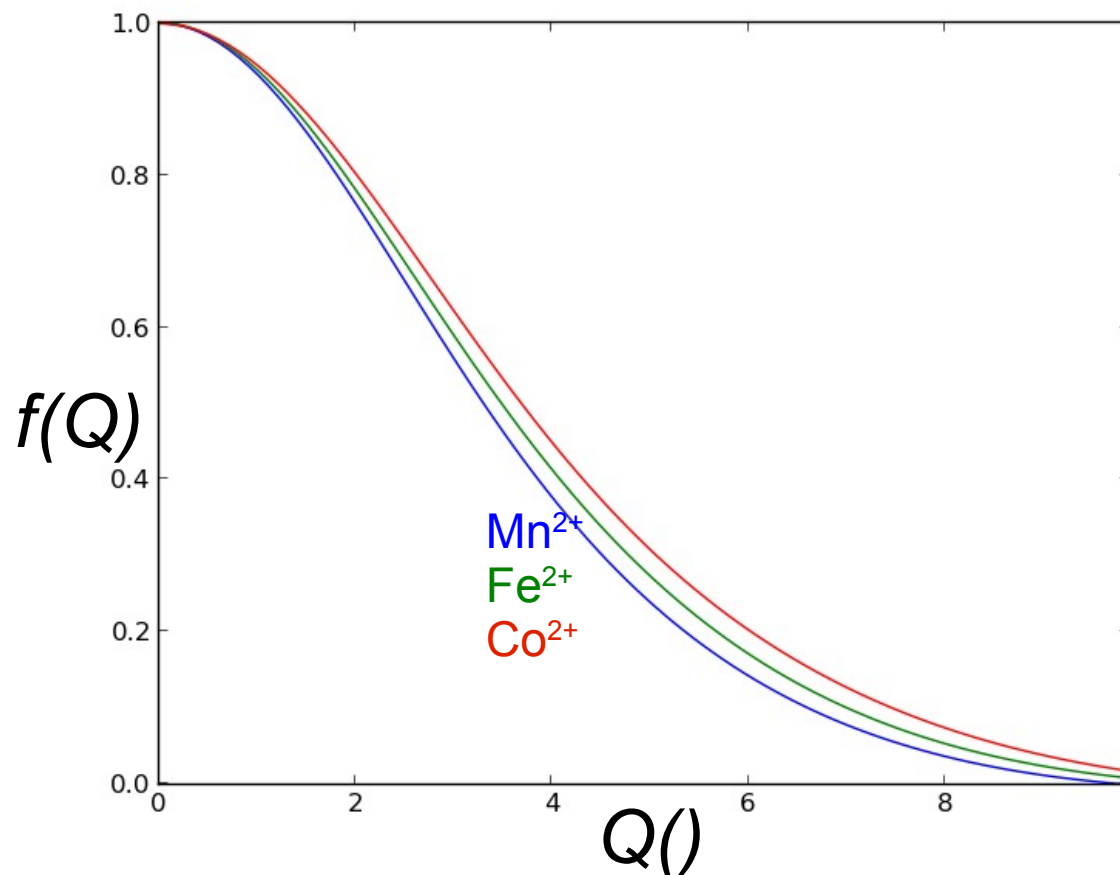
$$\langle j_L(\mathbf{Q}\cdot\mathbf{R}) \rangle = \int R_L^2(R) \cdot j_L(\mathbf{Q}\cdot\mathbf{R}) R^2 dR$$

Very often used in the dipolar approximation when modeling magnetic structures.

Magnetic form factor (Dipolar limit, j_0 term)

*Magnetic form factor tabulated
by Brown:
International Tables for
Crystallography, Volume C,
sect. 4.4.5)*

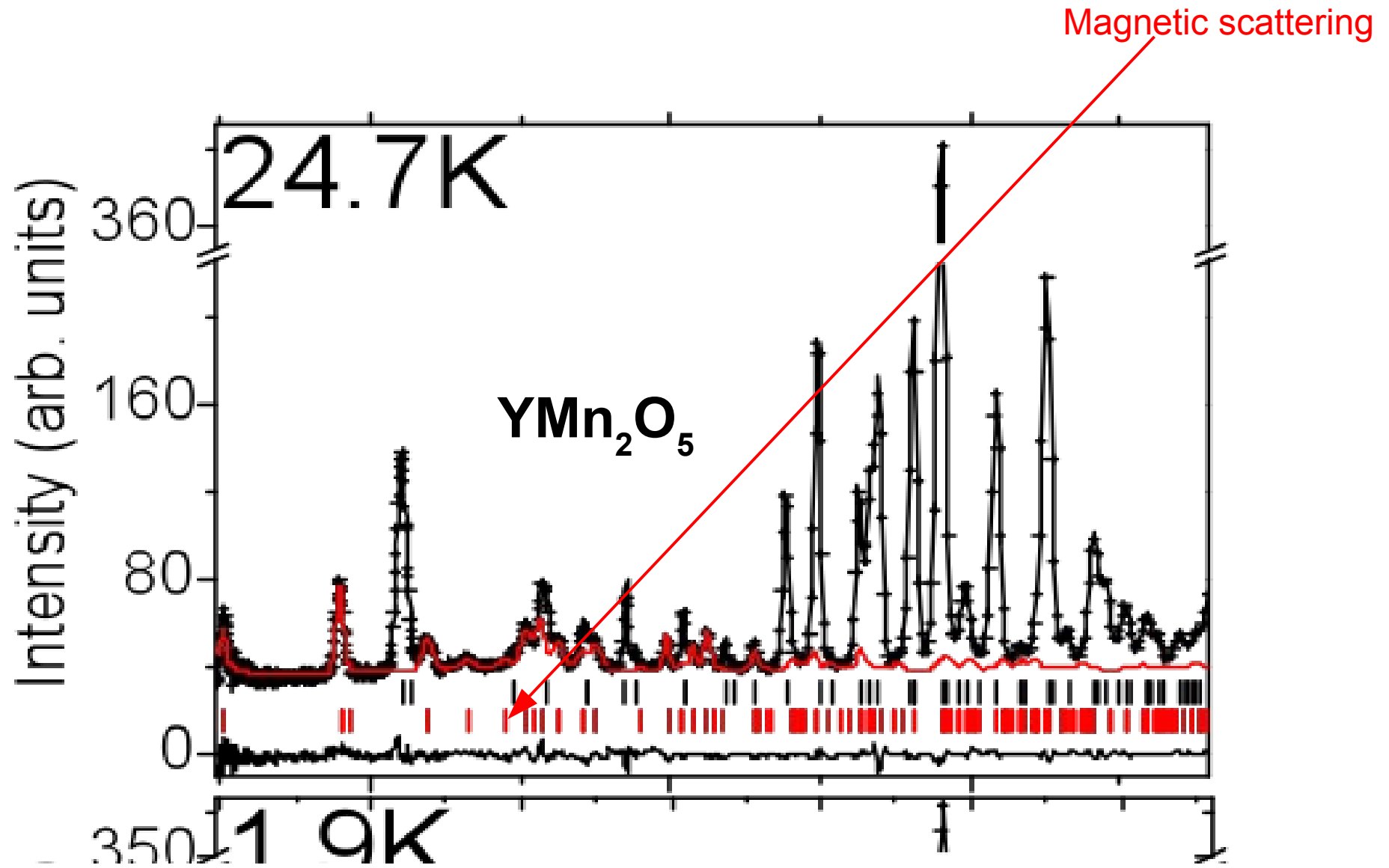
*Calculated by Hartree-Fock method
using Slater type orbitals
then fitted using
analytic approximations (expansion
in exponentials)*



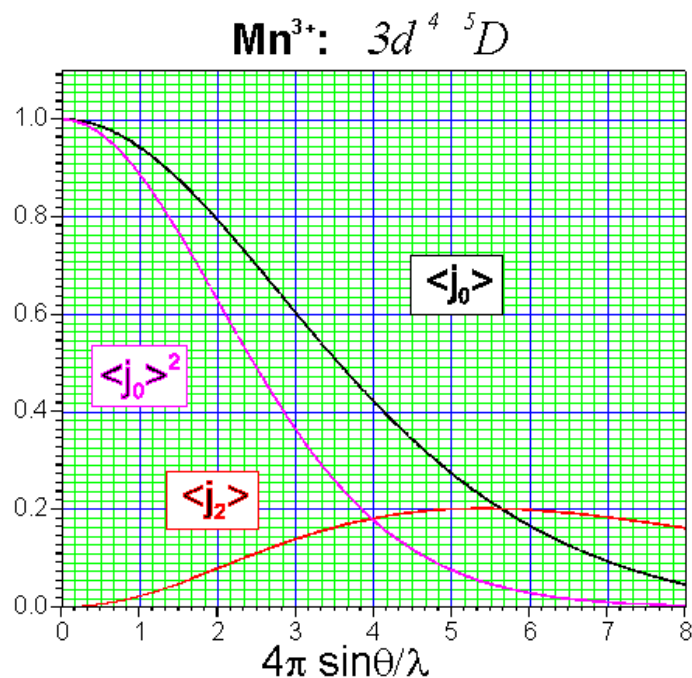
$$\langle j_l(s) \rangle = s^2 \left(A_l \exp\{-a_l s^2\} + B_l \exp\{-b_l s^2\} + C_l \exp\{-c_l s^2\} + D_l \right)$$

$$\langle j_0(s) \rangle = A_0 \exp\{-a_0 s^2\} + B_0 \exp\{-b_0 s^2\} + C_0 \exp\{-c_0 s^2\} + D_0$$

Magnetic form factor (example)



Magnetic form factor (dipolar limit, j₀,j₂)

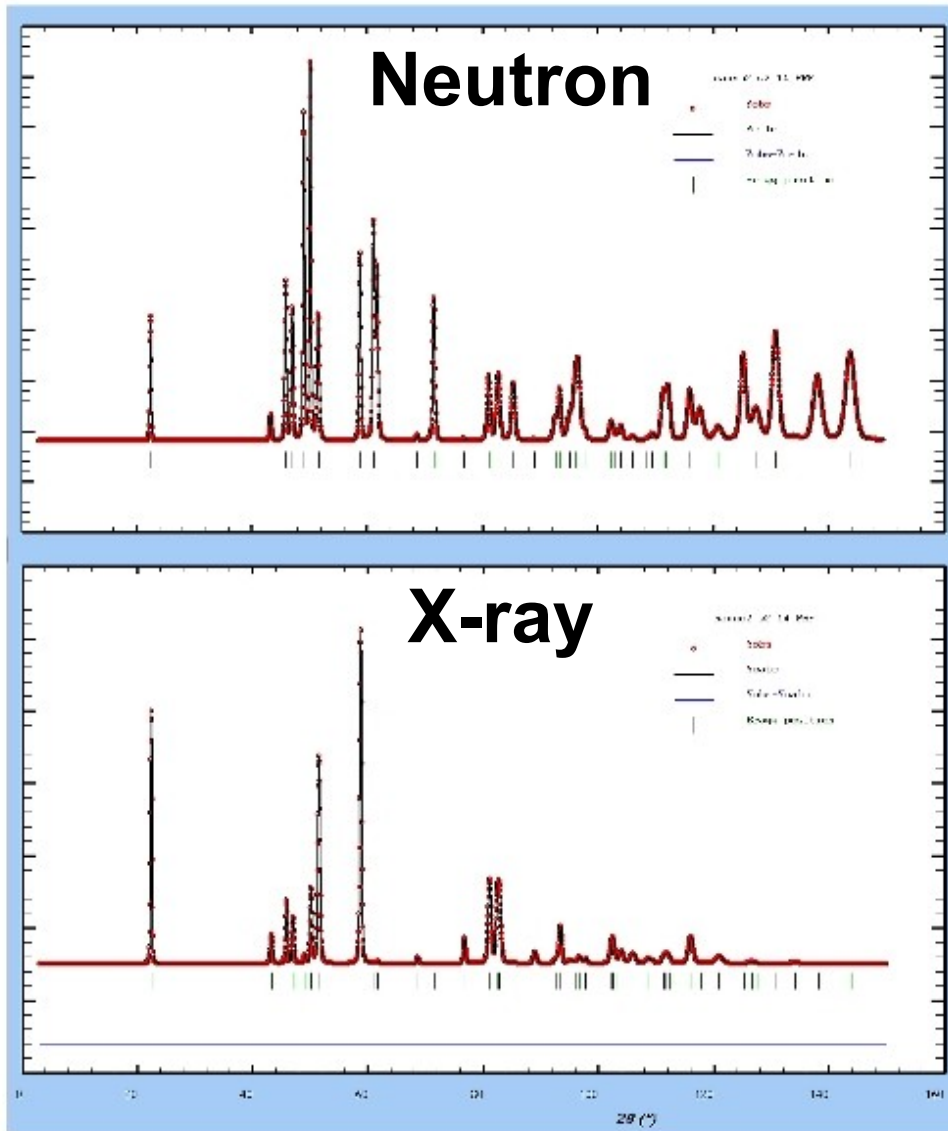


In the dipole approximation:

$$f(Q) = \langle j_0(Q) \rangle + \left(1 - \frac{2}{g}\right) \langle j_2(Q) \rangle$$

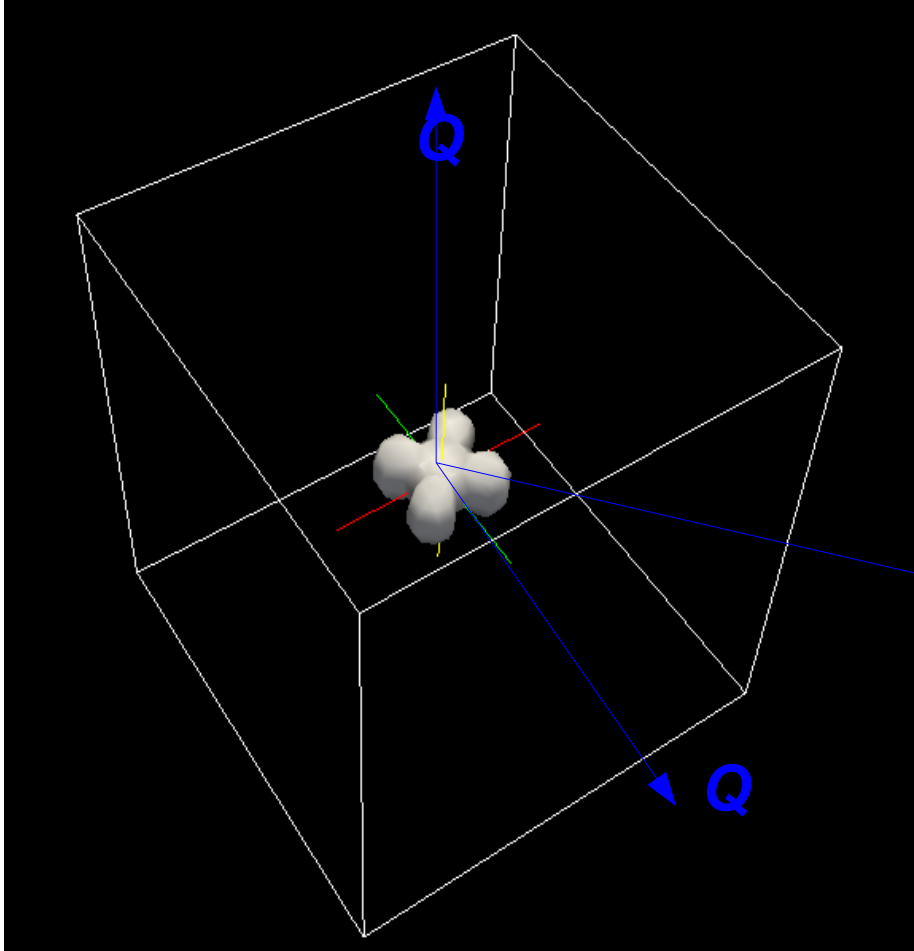
*International Tables of Crystallography, Volume C,
ed. by AJC Wilson, Kluwer Ac. Pub., 1998, p. 513*

Magnetic form factor (example)



- Form factor is similar to the X-ray form factor except that in the case of X-ray, the scattering arise from all electrons and not simply the unpaired electrons.

Magnetic form factor



Form factor depends not only on the modulus of Q but also the direction.

$$m(\mathbf{R}) = \sum_l R_l^2(R) \sum_{m,p} \beta_l^{m,p} \cdot y_l^{m,p}(\hat{\mathbf{R}})$$

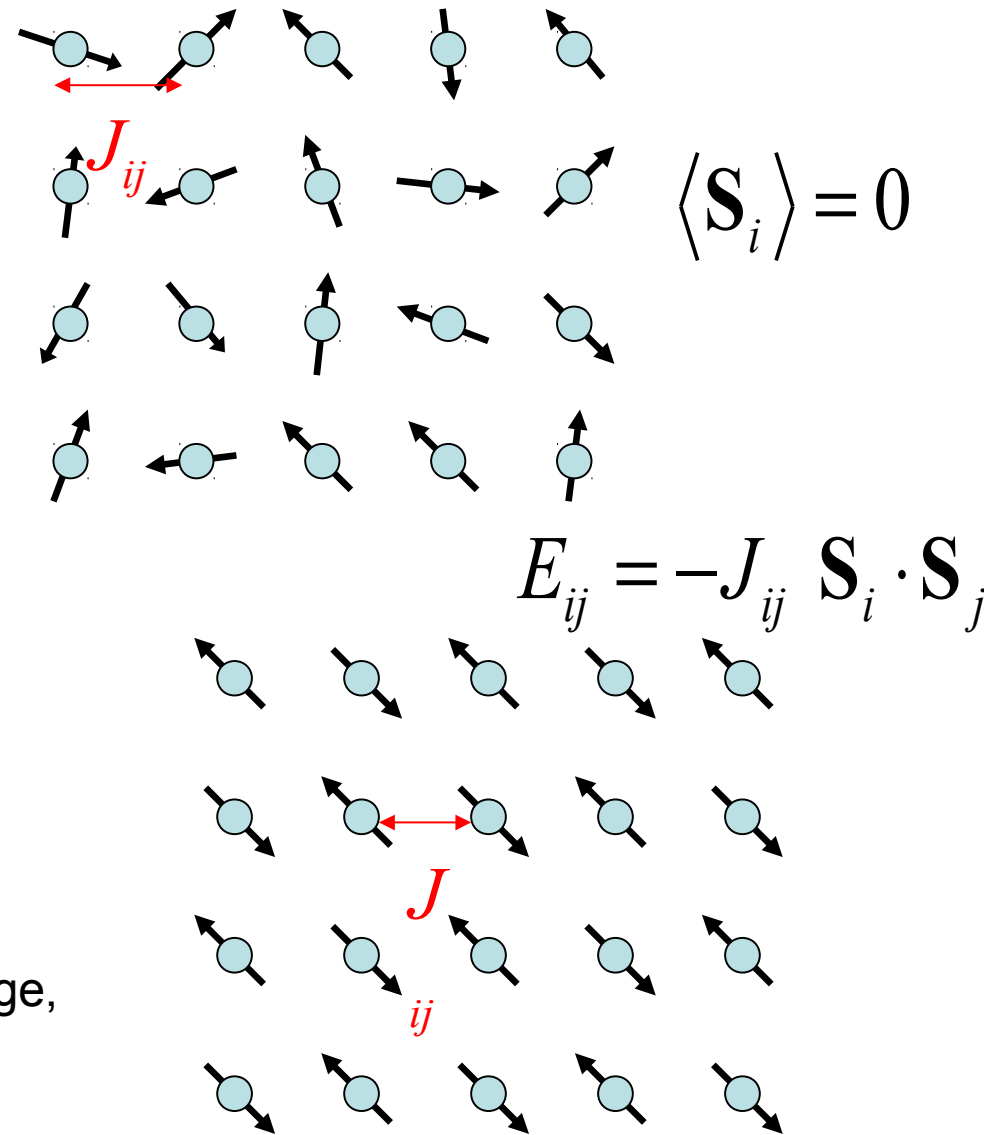
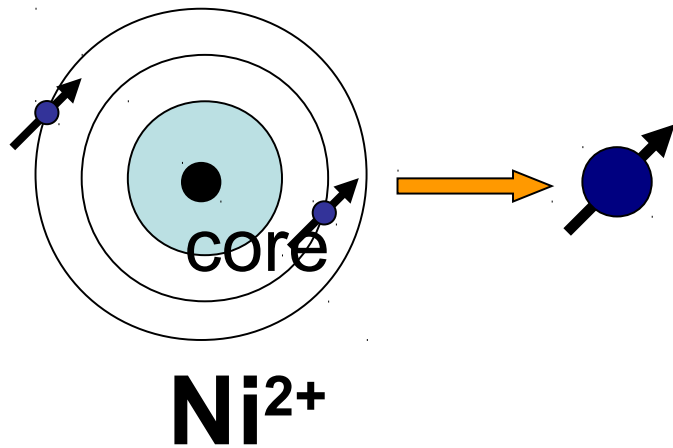
Q

Q

Examples of magnetic scattering experiments

Different magnetic state(s)

- In some crystals, some of the atoms/ions have unpaired electrons (transition metals, rare-earths).
- The intra-atomic electron correlation, Hund's rule, favors a state with maximum S/J, the ions possess a localized magnetic moment



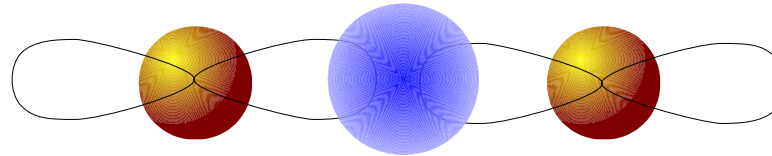
- When exchange interactions (direct, superexchange, double exchange, RKKY, dipolar) stabilizes a long range magnetic order.

Different magnetic state(s)

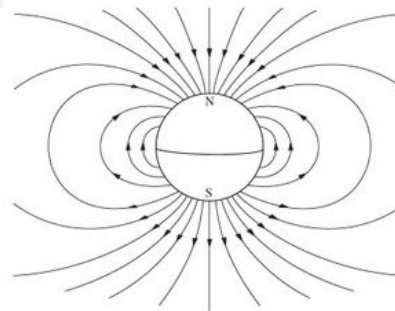
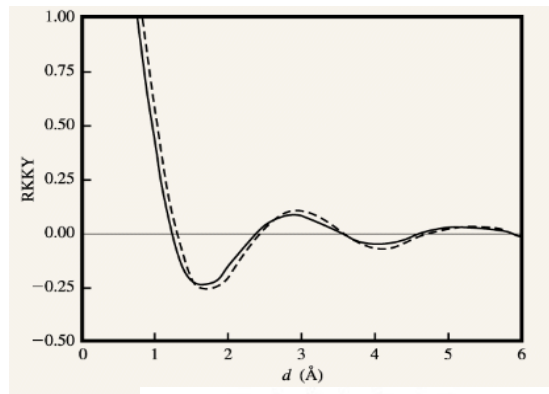
- Direct exchange interaction
(direct overlap of orbital wave-functions)
AFM for short-distance



- Indirect exchange interaction
- Super-exchange (M-O-M)
- Super super-exchange (M-O-O-M)
coupling through a diamagnetic anion or
more complex exchange paths



- RKKY interactions
(coupling of localized moments
through conduction electrons)



- Dipole-dipole interaction.
Decrease rapidly with distance.
Usually relevant for large moments at
low T

First study of antiferromagnetism with neutrons

1945



Oak Ridge Nat. Lab.

1949

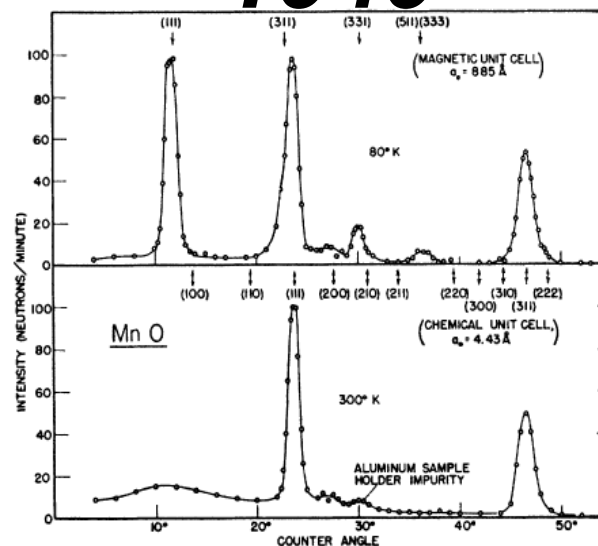


FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.

Detection of Antiferromagnetism by Neutron Diffraction*

C. G. SHULL

Oak Ridge National Laboratory, Oak Ridge, Tennessee

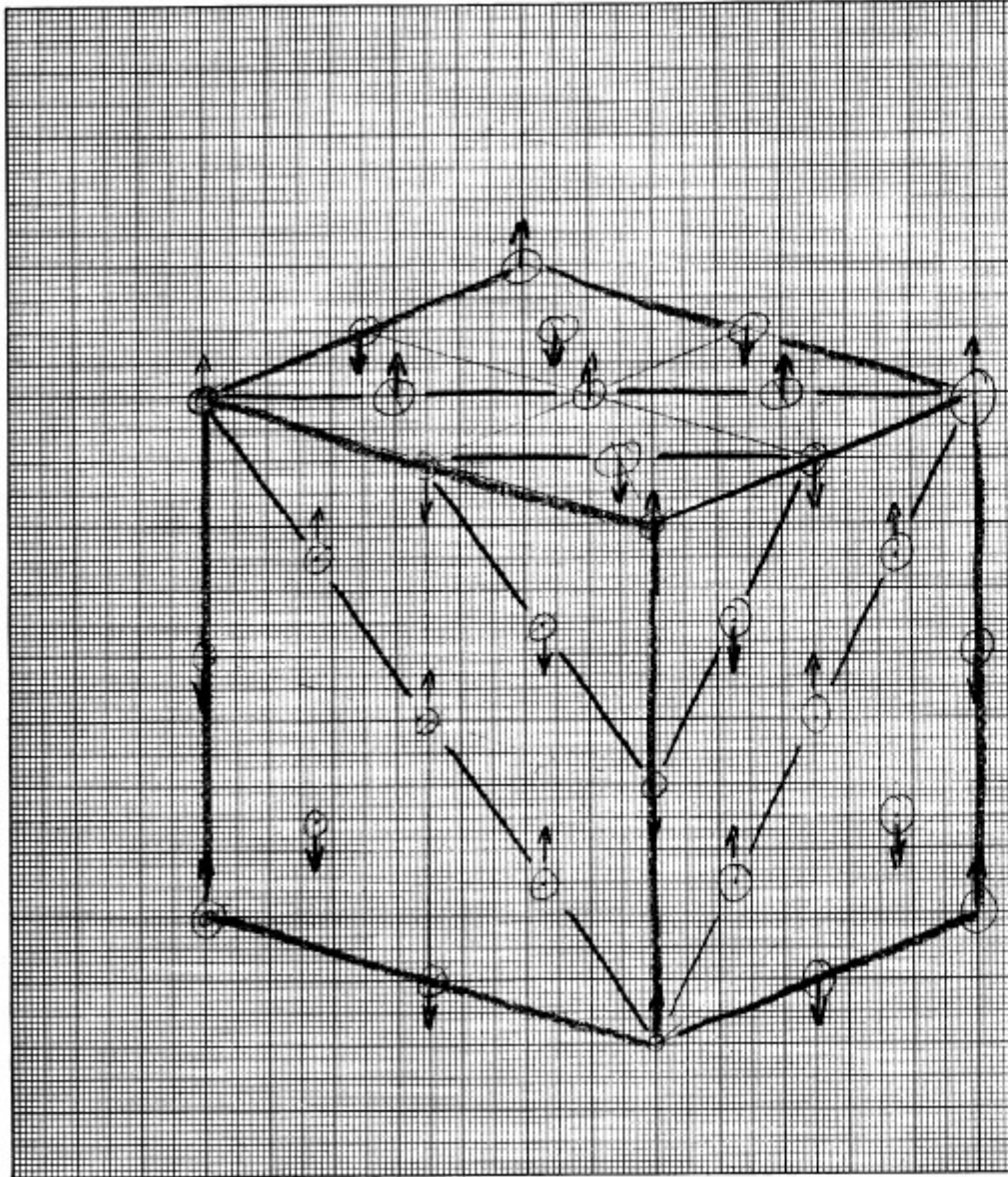
AND

J. SAMUEL SMART

Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland

August 29, 1949

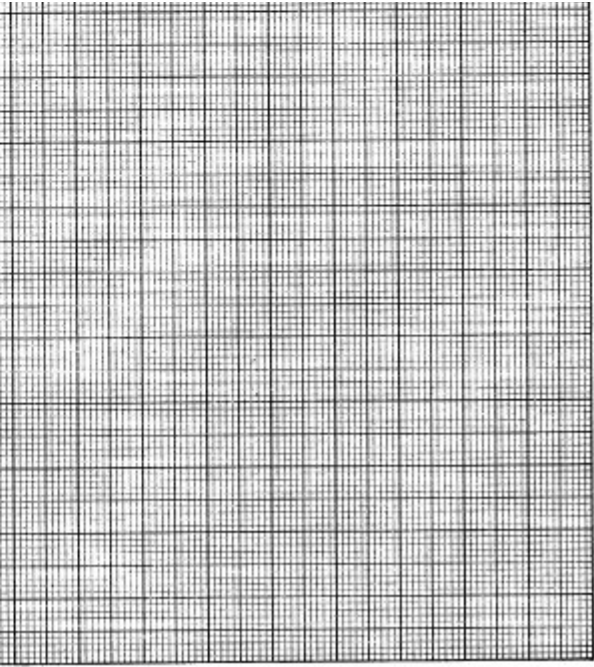
MnO



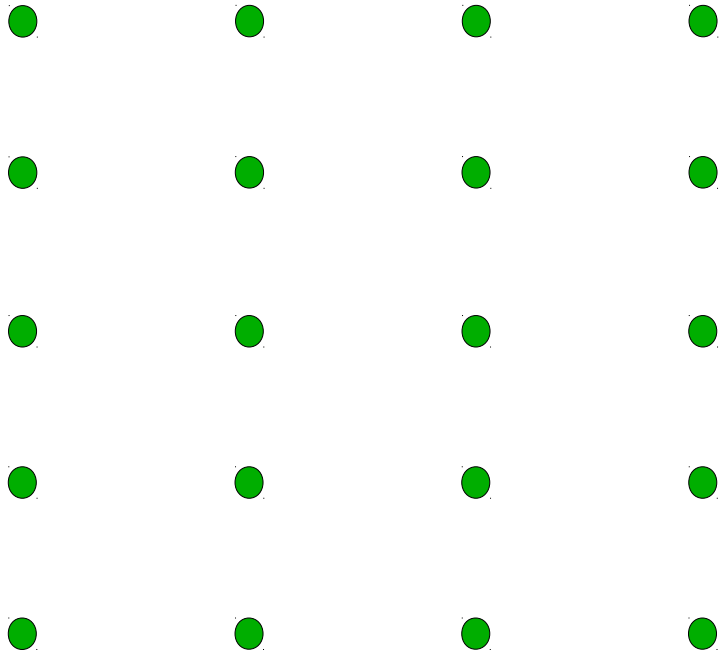
L. Neel

Ann. de Physique, 3 p.137 (1948)
 C.R. Acad Sci Paris 228 64-6 (1949)

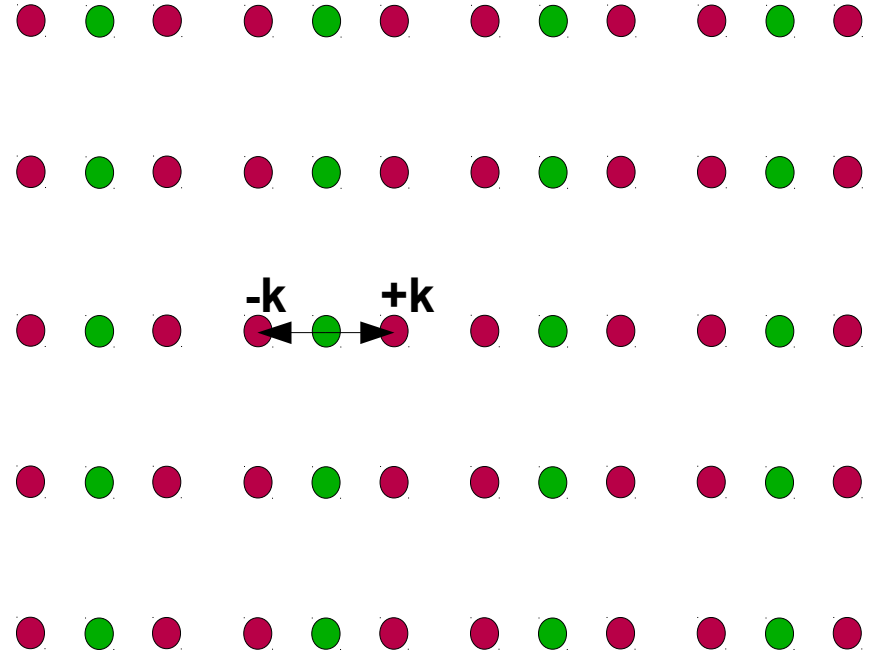
	MnO	MnS	FeO	FeF ₂	MnF ₂	α Fe ₂ O ₃
d (M-M)	3.12	3.68	3.03	3.36	3.30	2.91 ^(2.88) _(2.94)
d' (M-O)	2.21	2.61	2.14	2.12	2.11	2.03
θ_N °K	-610	-528	-570	-117	-113	-2000
θ_P °K	122	165	198	79	72	950
C	4.4	4.3	6.24	3.88	4.08	4.4



More complex example(s)



Reciprocal lattice



Reciprocal lattice
(incommensurate magnetic peaks)

Formalism of propagation vector(s)

For simplicity, in particular for wave-vector inside the **BZ**, one usually describe magnetic structures with Fourier components:

$$\mathbf{m}_{ij}(\mathbf{R}_L) = \sum_k \mathbf{S}_{kj} \cdot e^{-2\pi i \mathbf{k} \cdot \mathbf{R}_L}$$

which for a single propagation vector :

$$\mathbf{m}_{ij}(\mathbf{R}_L) = \mathbf{S}_{kj} \cdot e^{-2\pi i \mathbf{k} \cdot \mathbf{R}_L} + \mathbf{S}_{-kj} \cdot e^{2\pi i \mathbf{k} \cdot \mathbf{R}_L}$$

Since \mathbf{m}_{ij} is a real vector,

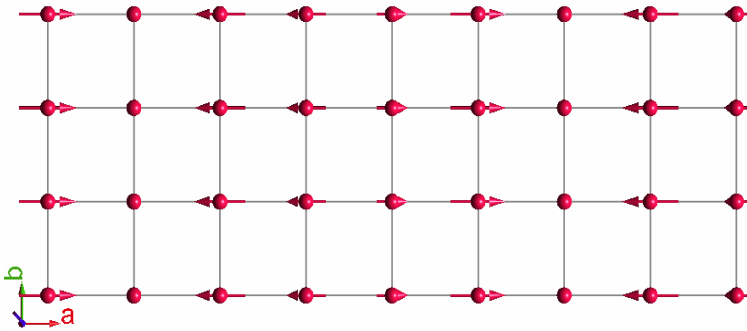
one must imposes the condition $\mathbf{S}_{-kj}^* = \mathbf{S}_{kj}$

Here \mathbf{S}_{kj} is a complex vector made of linear combinations of basis vectors that, in the most general case, do not span necessary the same irreducible representations.

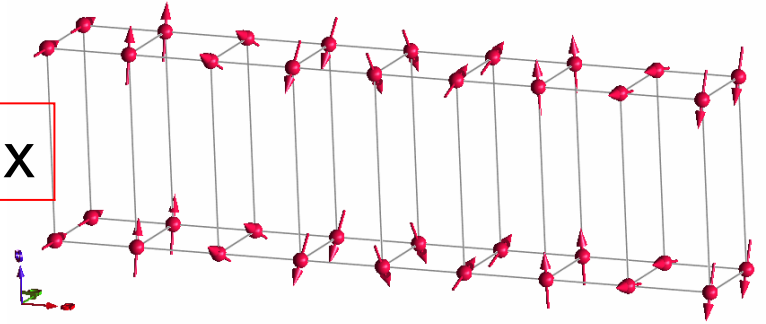
k inside BZ

- k interior of the Brillouin zone (pair k, -k)

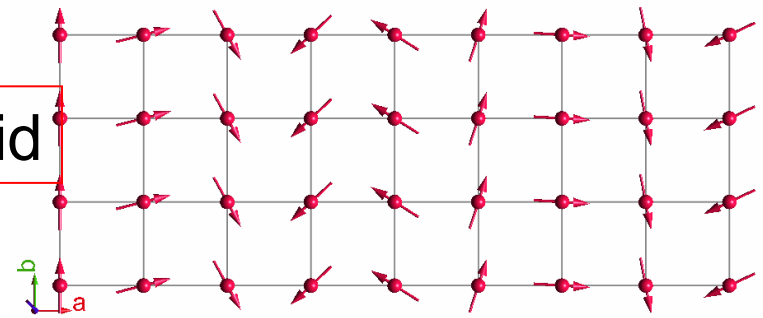
Amplitude modulation



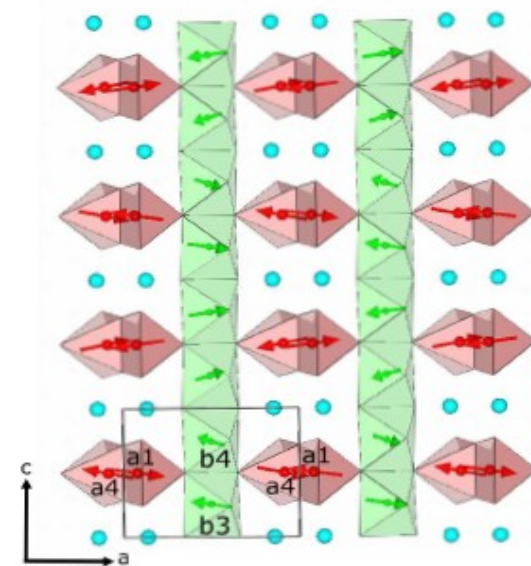
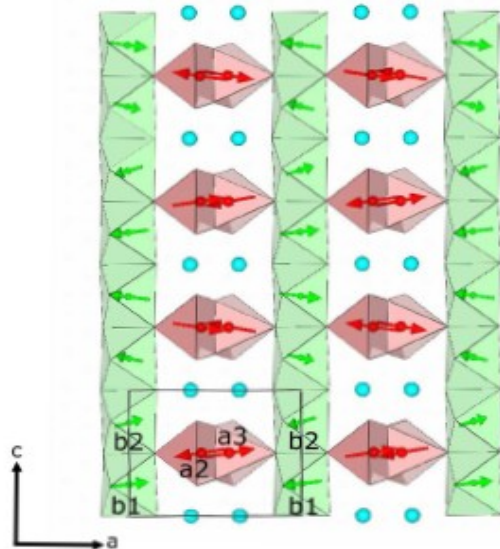
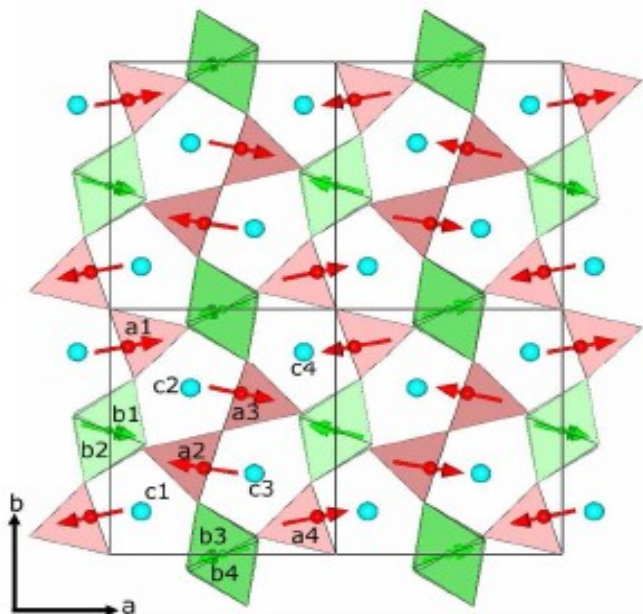
Helix



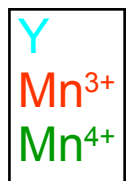
Cycloid



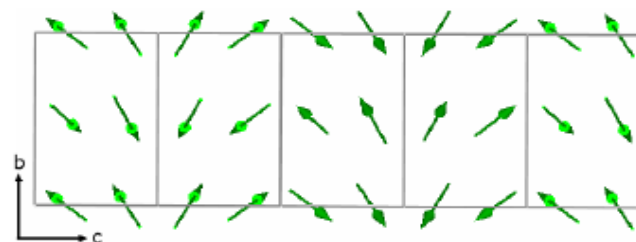
Quite complex ordered states (RMn_2O_5)



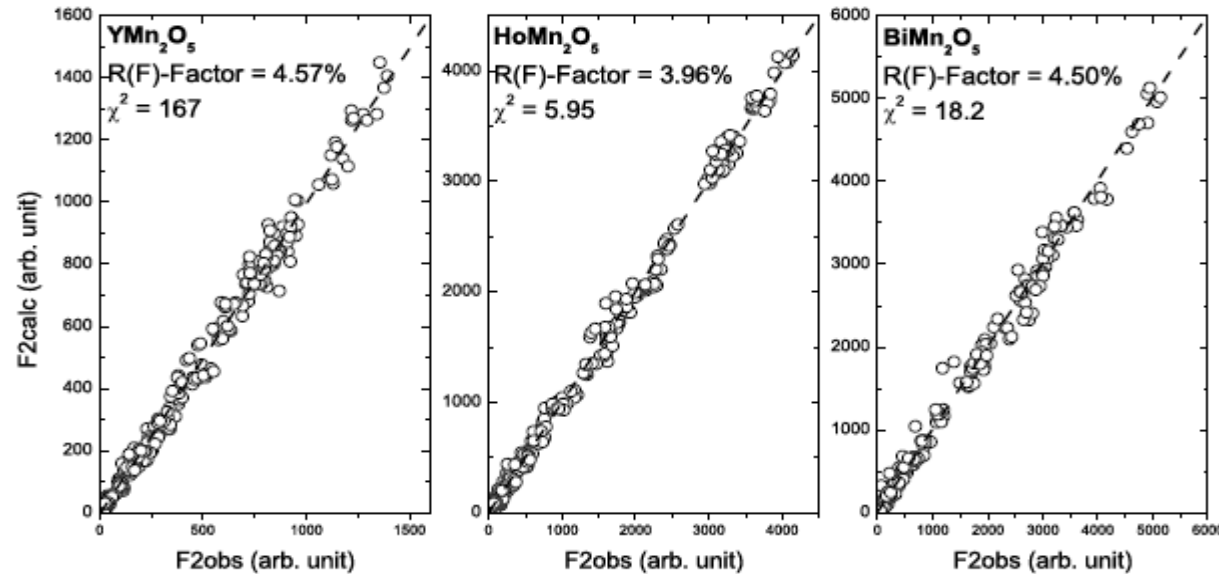
$$k=(1/2, 0, 1/4)$$



AFM magnetic chains (ab-plane)
Cycloidal component (c-direction)

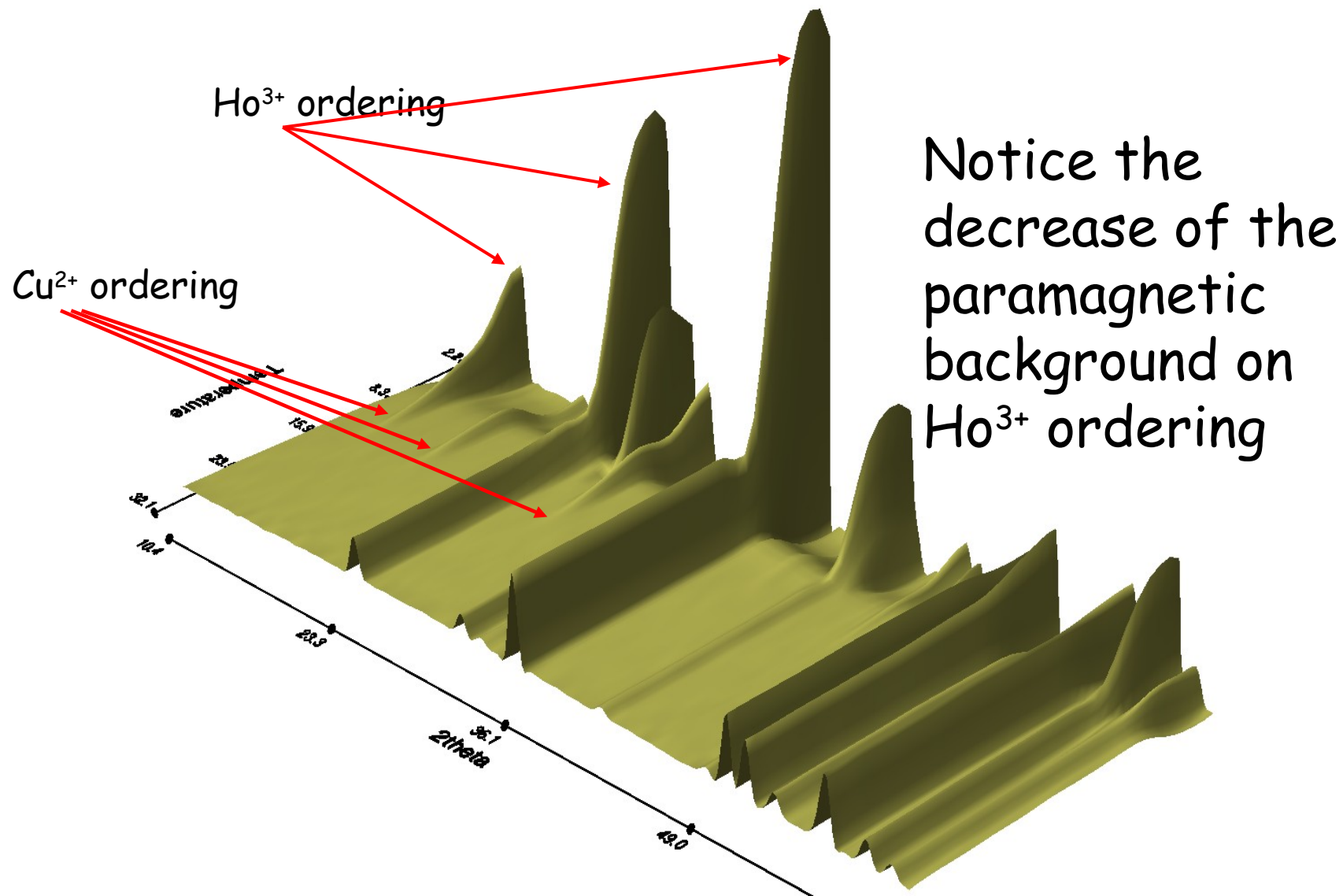


Quite complex ordered states (RMn_2O_5)



YMn_2O_5 355 independent reflections 37 refined parameters: {Rx-Ry-lz-MagPh(Mn ³⁺ fixed)-ext4} $m(\text{Mn}^{4+})=2.4 \mu_{\text{B}}$ $m(\text{Mn}^{3+})=3.1 \mu_{\text{B}}$	HoMn_2O_5 381 independent reflections 53 refined parameters: {Rx-Ry-lz-MagPh(Mn ³⁺ fixed)-ext4} $m(\text{Mn}^{4+})=2 \mu_{\text{B}}$ $m(\text{Mn}^{3+})=2.5 \mu_{\text{B}}$ $m(\text{Ho})=0/1 \mu_{\text{B}}$	BiMn_2O_5 204 independent reflections 24 refined parameters: {Rx-Ry-Rz-ext4} $m(\text{Mn}^{4+})=2.1 \mu_{\text{B}}$ $m(\text{Mn}^{3+})=2.8 \mu_{\text{B}}$
--	--	---

Magnetic ordering of Ho and Cu ions in $\text{Ho}_2\text{BaCuO}_5$ (D1B)



Competing multi-q magnetic structures in HoGe₃ (I & II)

P Schöbinger-Papamantellos, J Rodríguez-Carvajal, LD Tung, C Ritter and KHJ Buschow
 J. Physics: Condensed Matter **20** (2008) 195201 (12pp)
 195202(13pp)

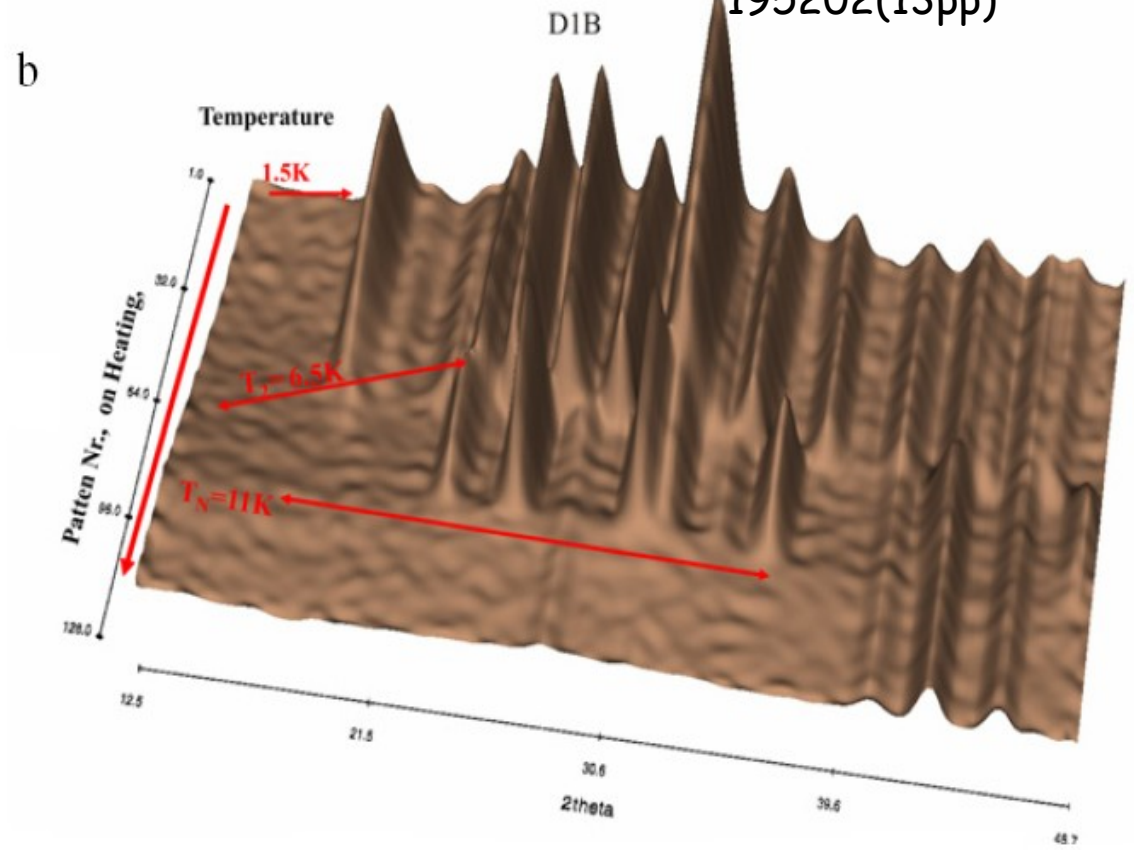
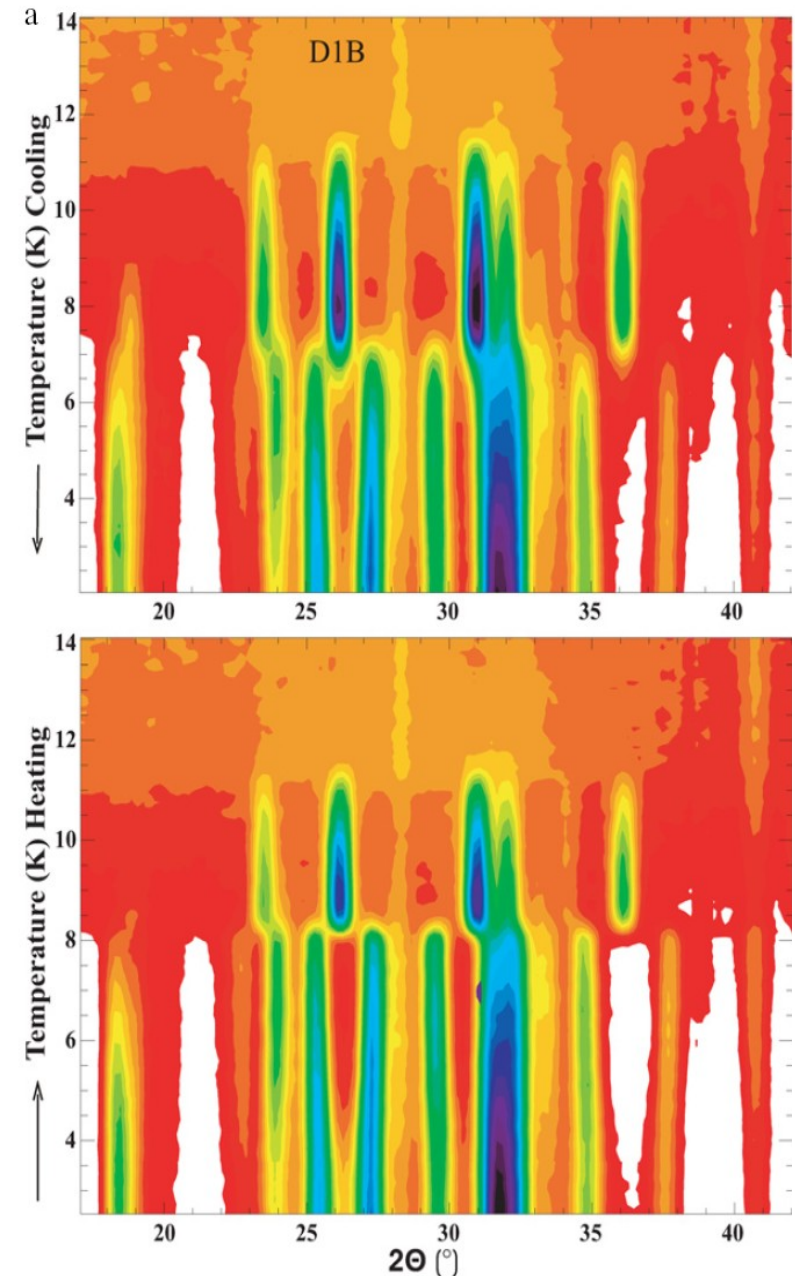
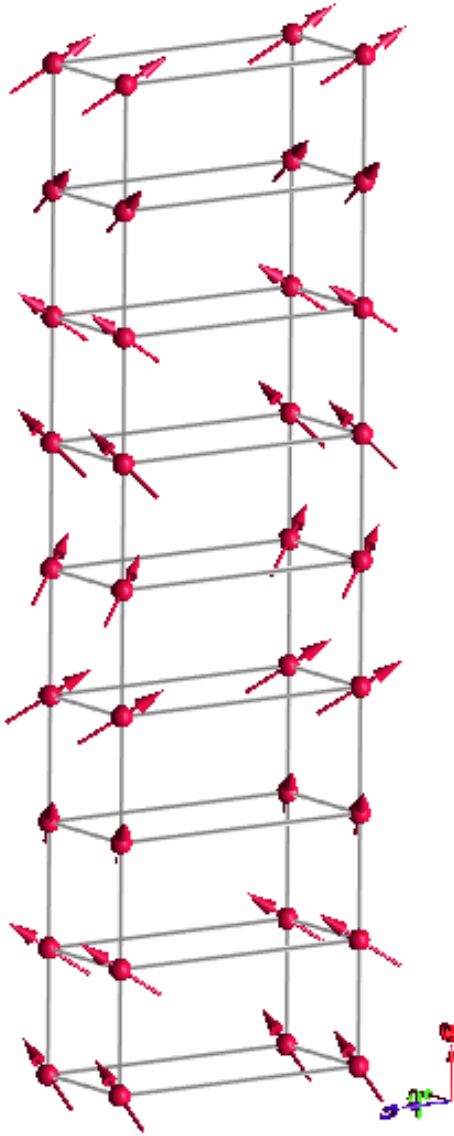


Figure 6. Thermodiffractogram of HoGe₃: (a) in a 2D projection on heating and cooling showing the succession of magnetic phase transitions below $T_N = 11$ K at $T_2^H = 8.1$ K and $T_3^H = 4.8$ K (temperatures given on heating) and (b) in a 3D view on cooling.

Multi-k structure: conical example

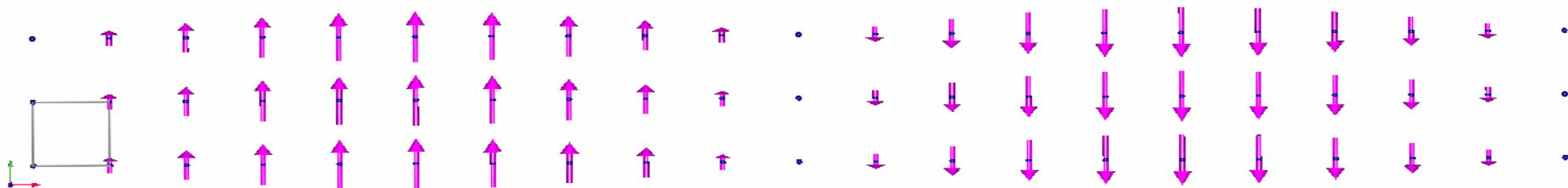
Conical



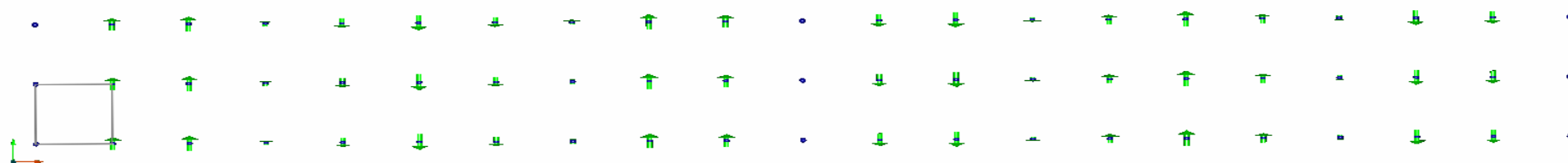
Multi-k structure with:

- **Helical modulation**
- **Ferromagnetic component**

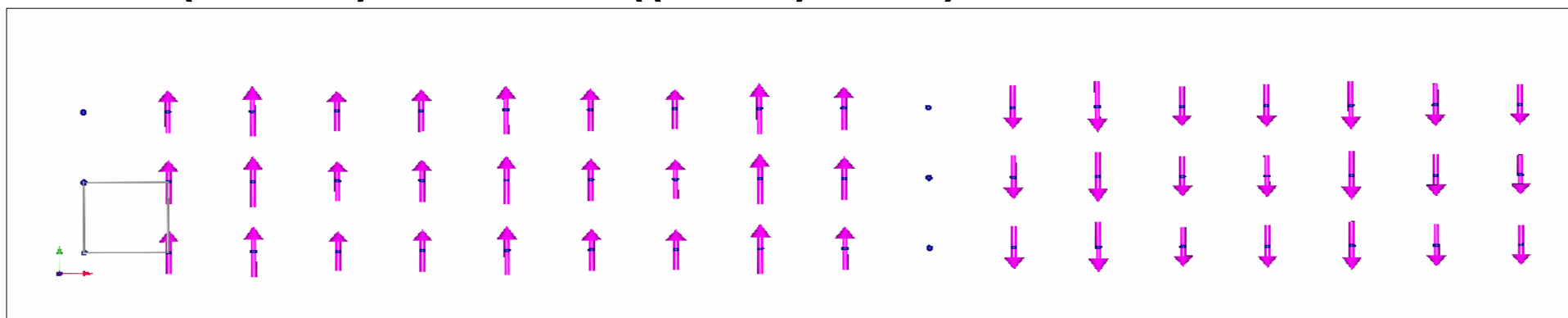
Multi-k structures : Bunched modulations



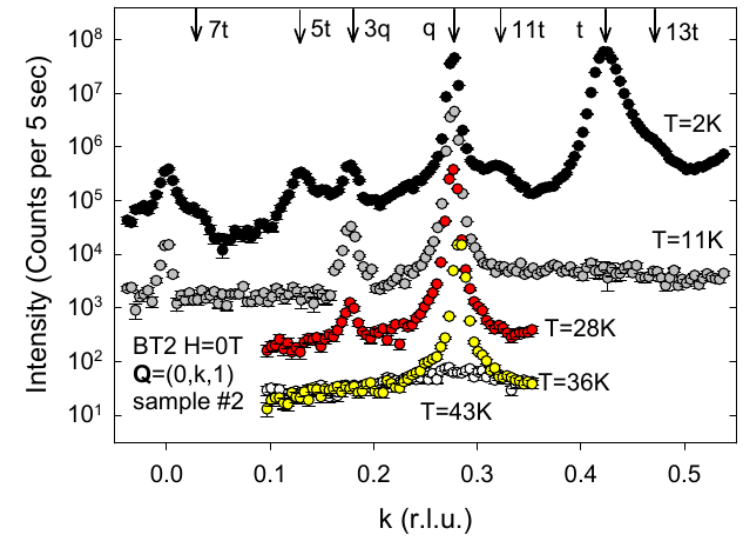
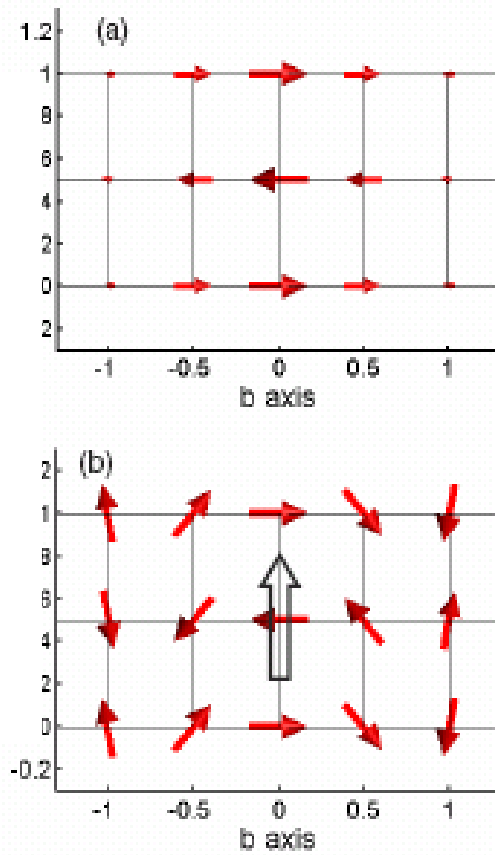
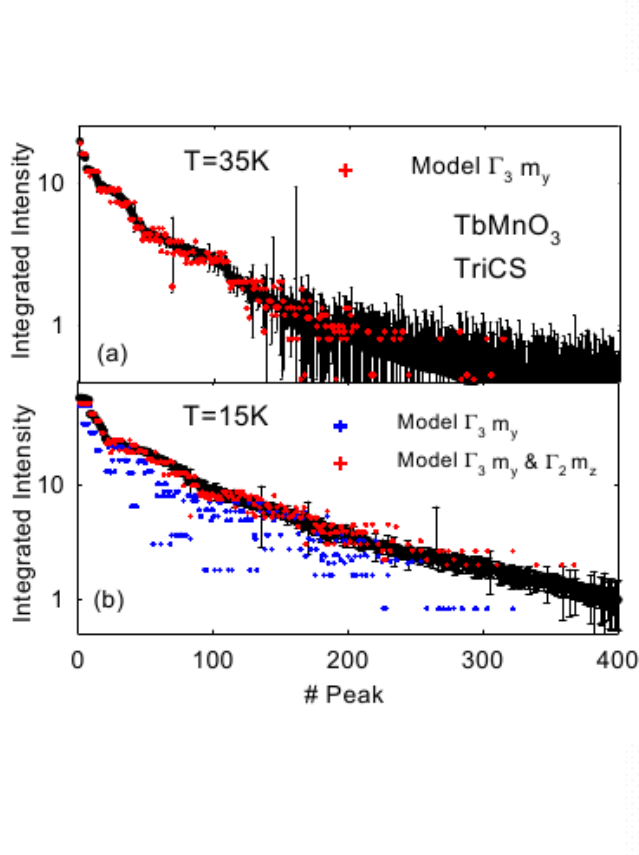
$$\mathbf{k}=(d,0,0)$$



$$+ \mathbf{k}=(3d,0,0) + \dots + \mathbf{k}=(2n+1)d,0,0)$$



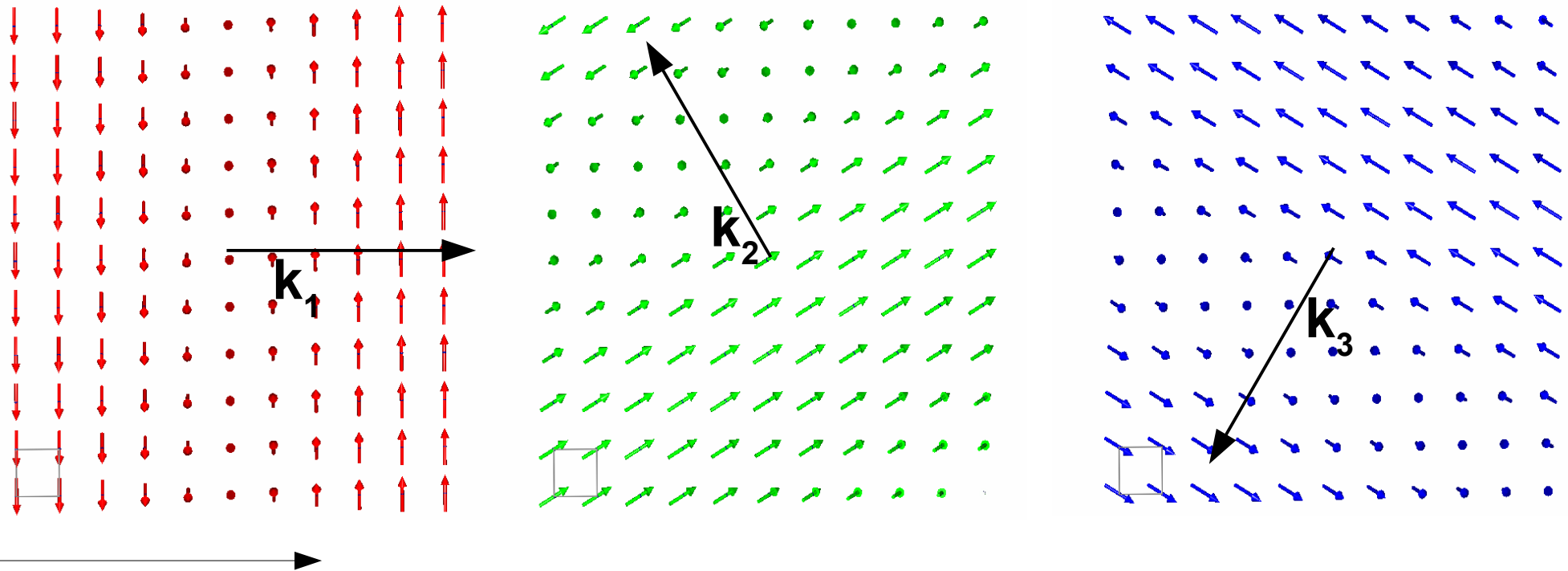
TbMnO₃



Kenzelmann, PRL 95, 087206 (2005)

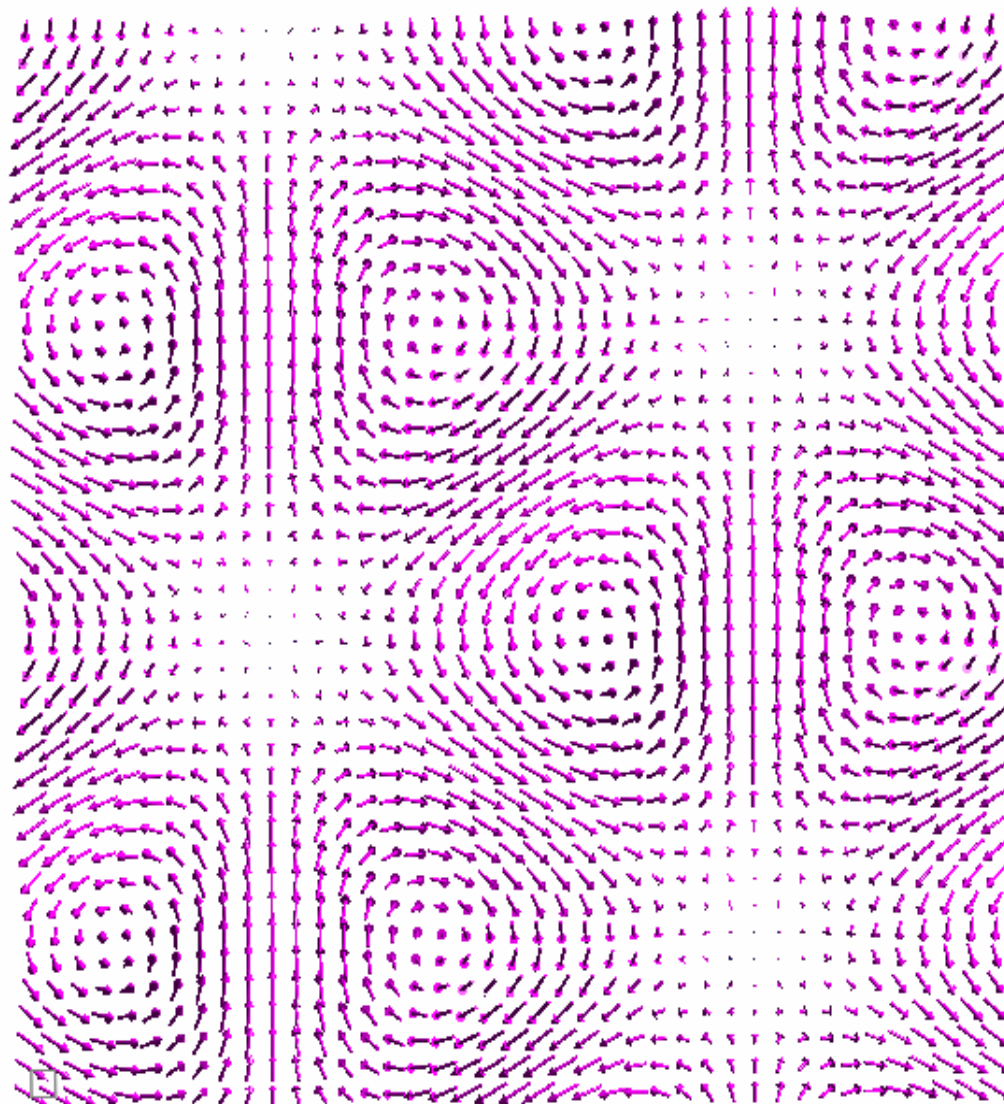
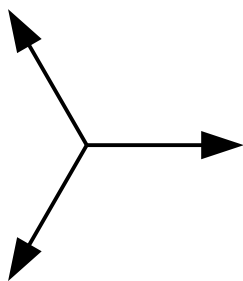
Multi-k structures

Example of a 4-k structure: the skyrmion lattice



- $k_1 + k_2 + k_3 = 0$, same chirality for k_1, k_2, k_3
- Ferromagnetic component

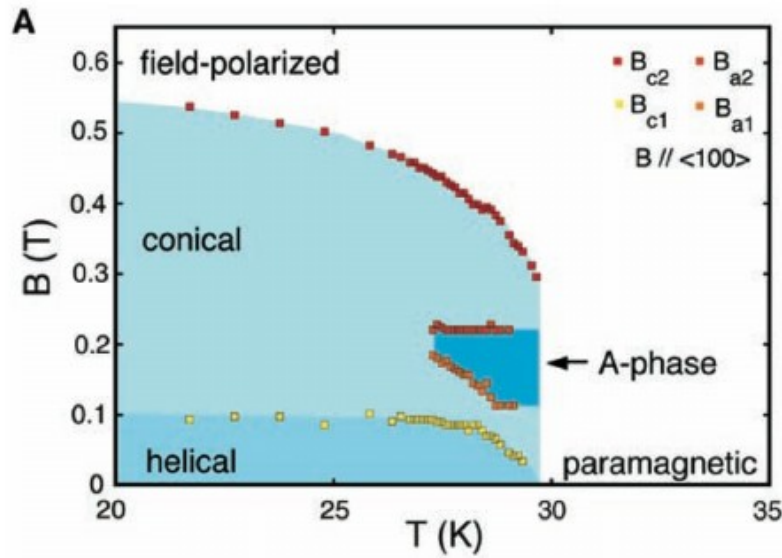
Multi-k structures



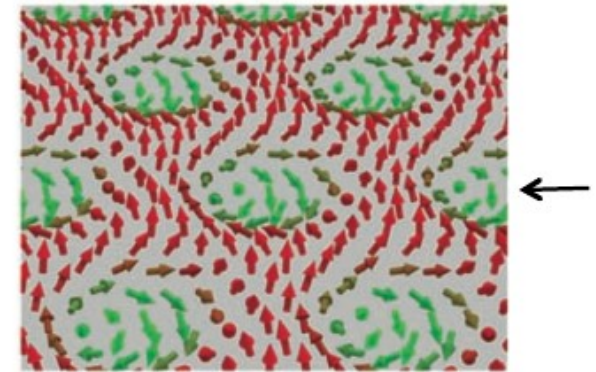
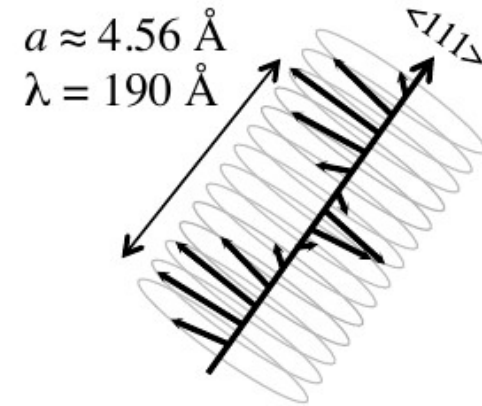
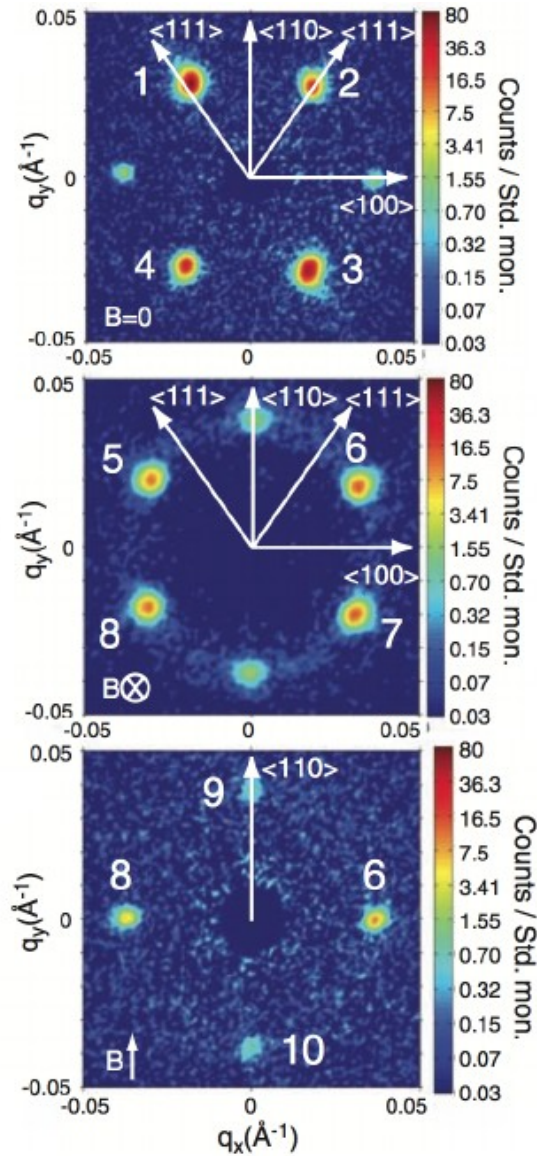
“Skyrmion”-type lattice stabilized by energy terms of the type:

$$F = \dots + S_1 e^{ik_1 + \varphi_1} \cdot S_2 e^{ik_2 + \varphi_2} \cdot S_3 e^{ik_3 + \varphi_3} \cdot M$$

Skymion in MnSi



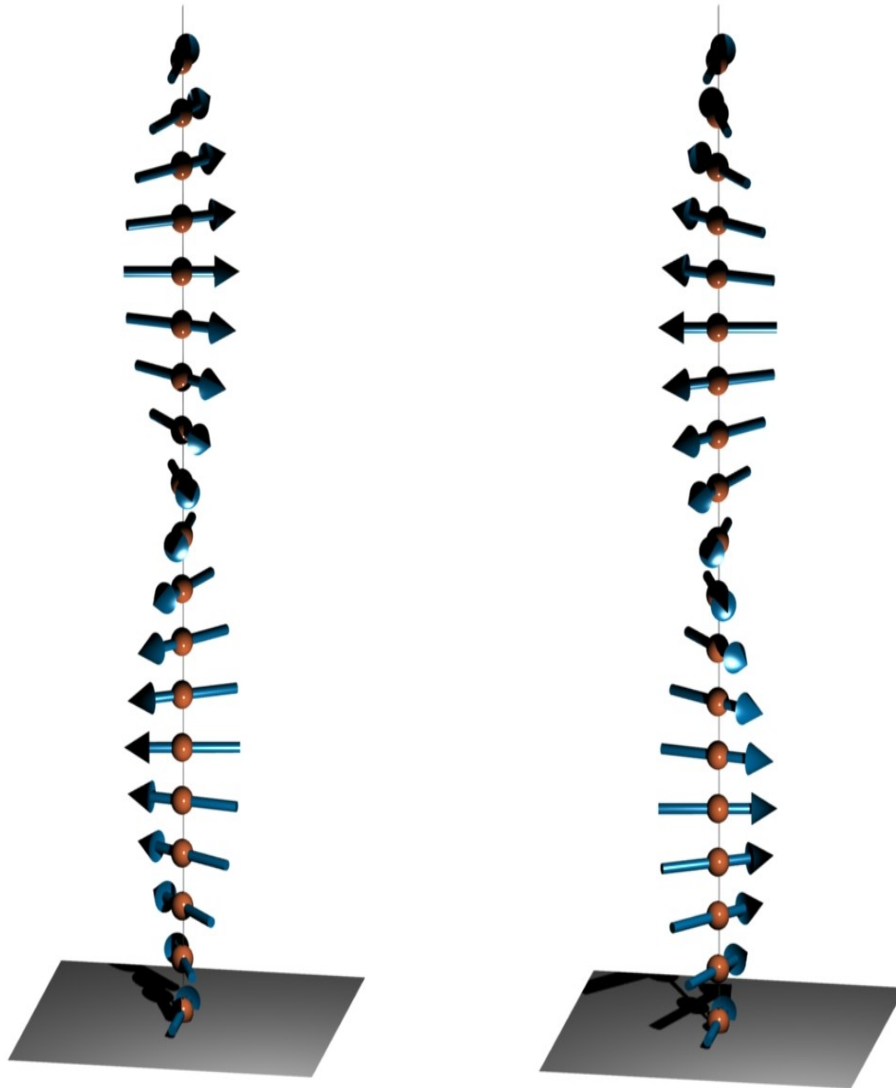
S. Mühlbauer et. al. Science **323** 915 (2009)



Domains

- Because the **symmetry** of the ordered magnetic state is **lower** than that of the paramagnetic state (loss of certain symmetry elements)
- If the order of the paramagnetic group \mathbf{G}_0 is \mathbf{g} and the order of the ordered group \mathbf{G}_1 is \mathbf{h} , there will be $\mathbf{g/h}$ domains.
- The different types of domains:
 - configuration domains (k-domains) : loss of translational symmetry
 - orientation domains (S-domains): loss of rotational symmetry
 - 180 degrees domains (time-reversed domains): loss of time-reversal symmetry
 - chiral domains: loss of inversion symmetry

Chiral-domains

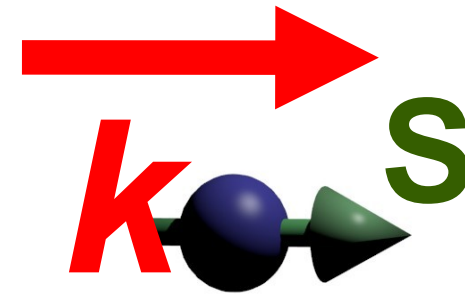
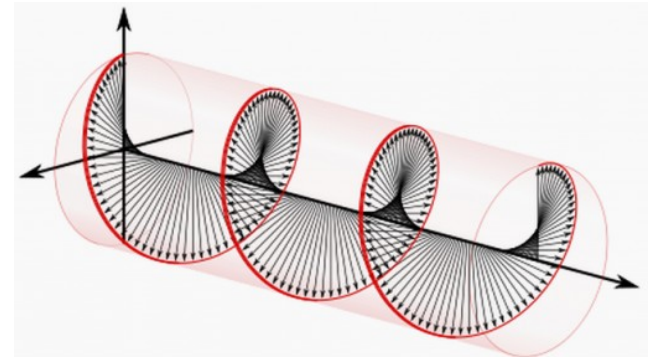


- Loss of inversion symmetry generates two domains of opposite handedness

- Note however that this is not the case if the paramagnetic group is a chiral group, in which case a single handedness is stabilized (no energy degeneracy)

Inversion domains (“chiral” scattering)

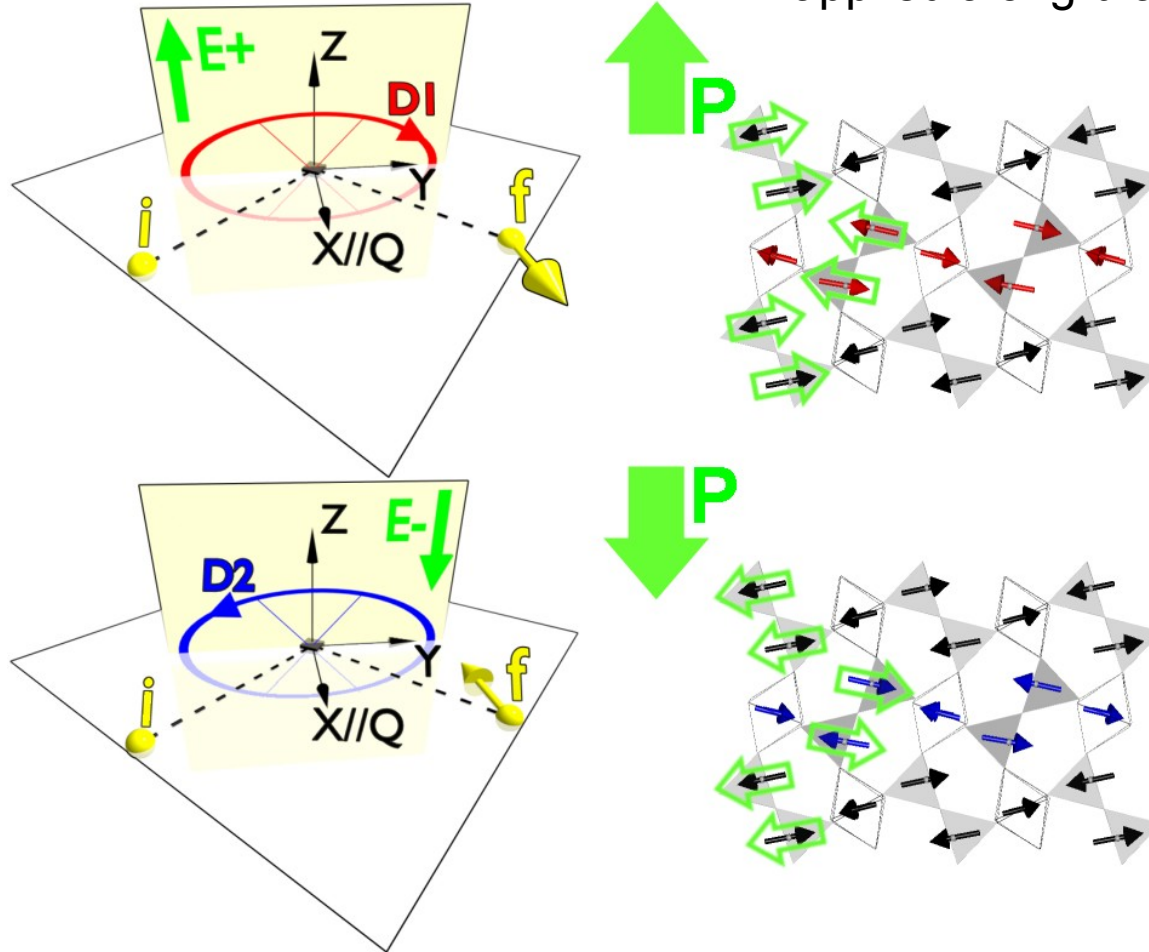
Spherical Polarimetry, ILL CRYOPAD



$$\Im(M_{\perp} \times M_{\perp}^*)$$

Inversion domains

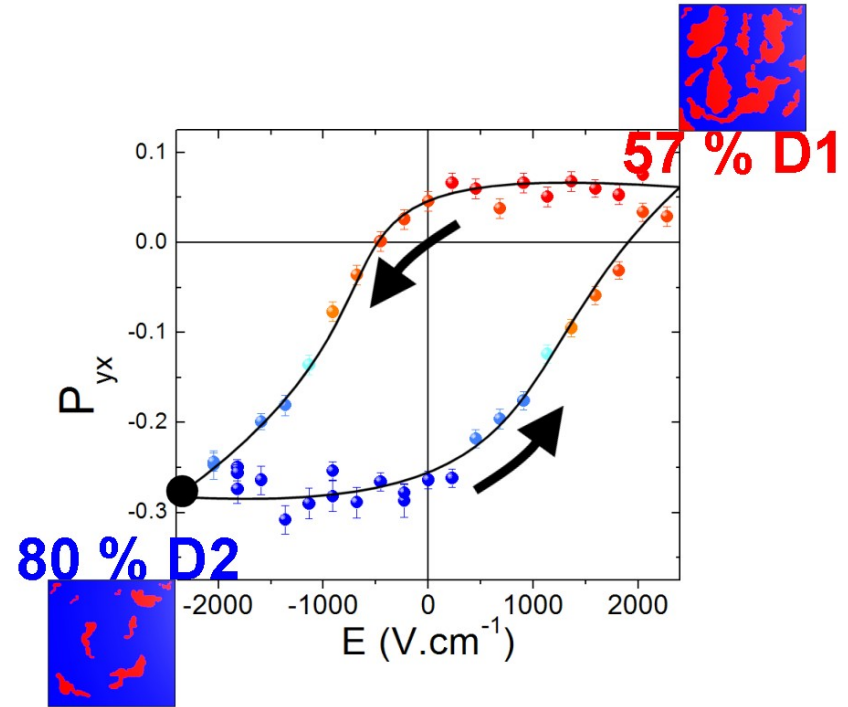
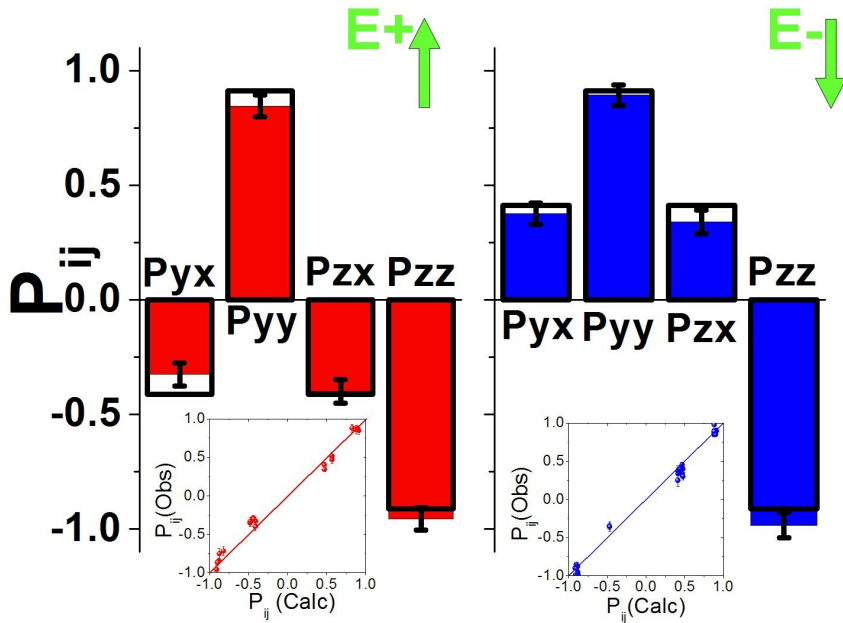
Crystal: b-axis along z-direction
Electric field up to 2.2 kV/cm
applied along b-axis



Inversion domains

$$P_{yx} = P_{zx} \text{ created polarization}$$

For the same hkl reflection, the sign of the created polarization is opposite for inversion domains

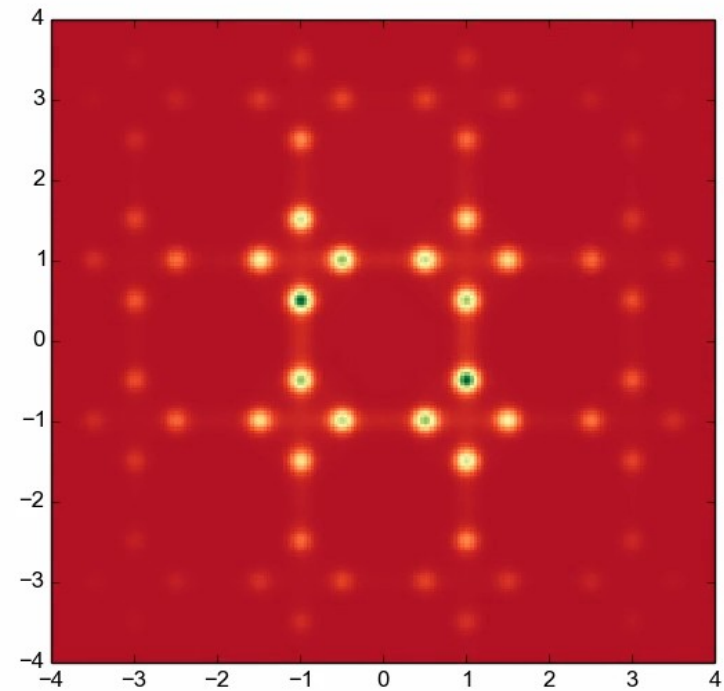
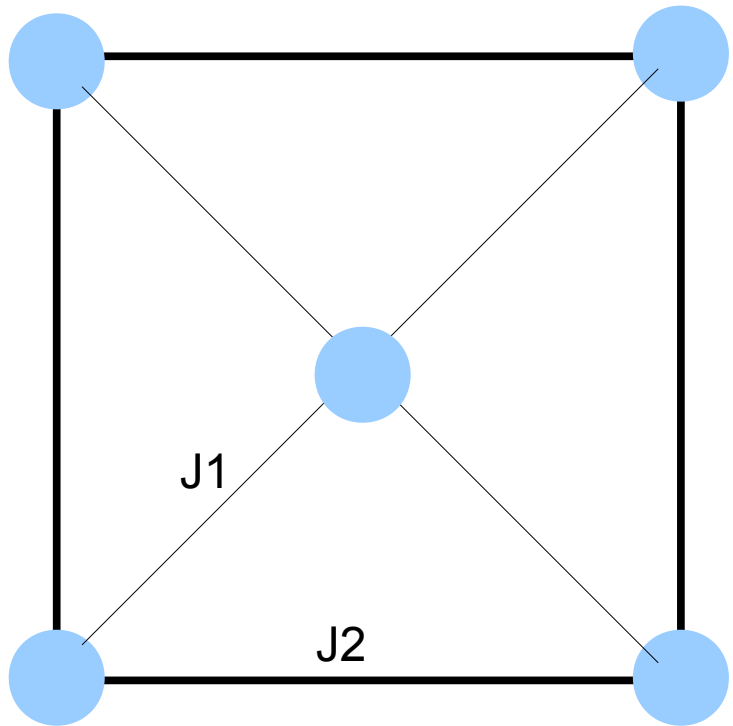


Radaelli et al., PRL 2008

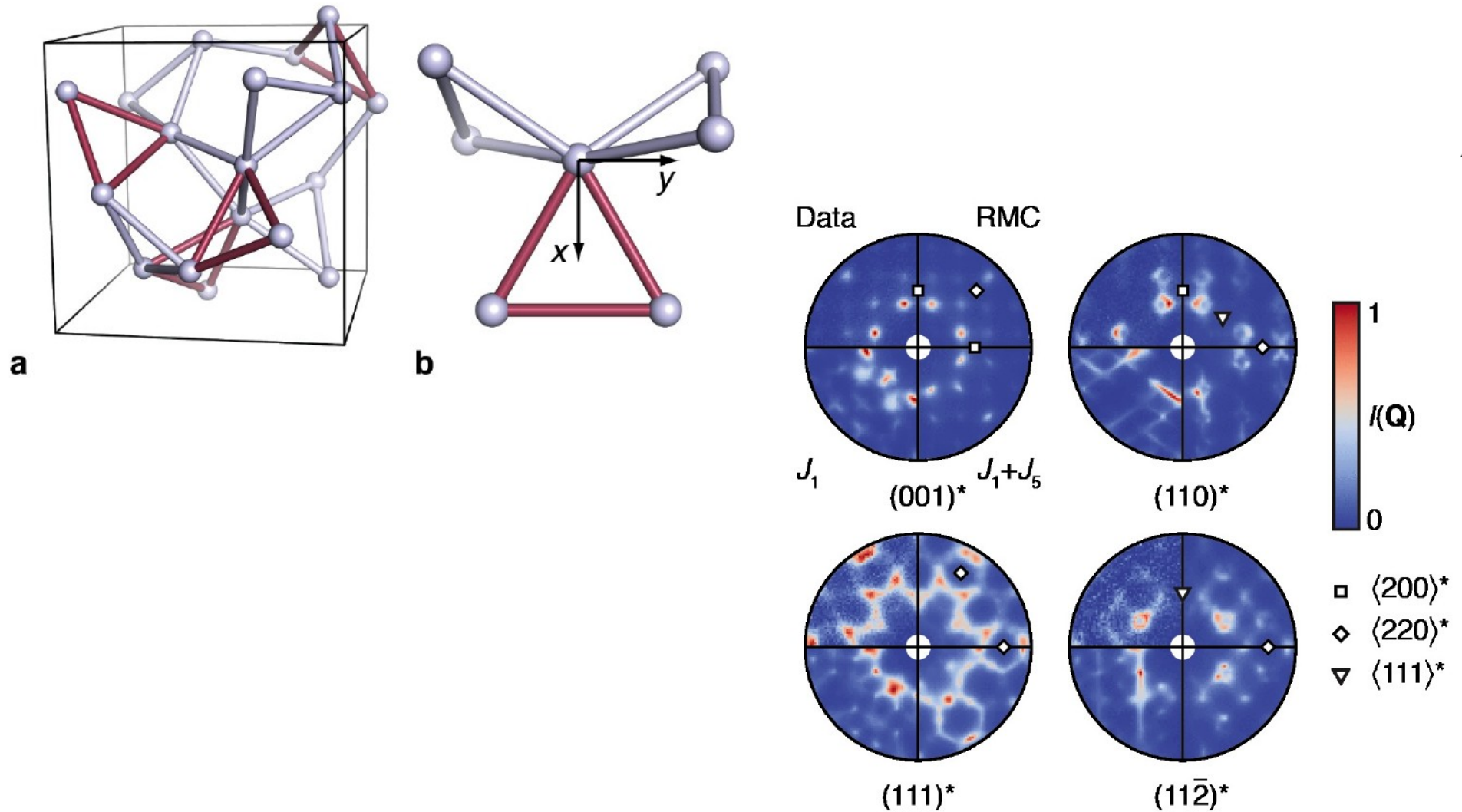
Short-range correlations

Probing short-range correlations
Via diffuse magnetic scattering

Simple J_1 - J_2 cubic fcc magnet

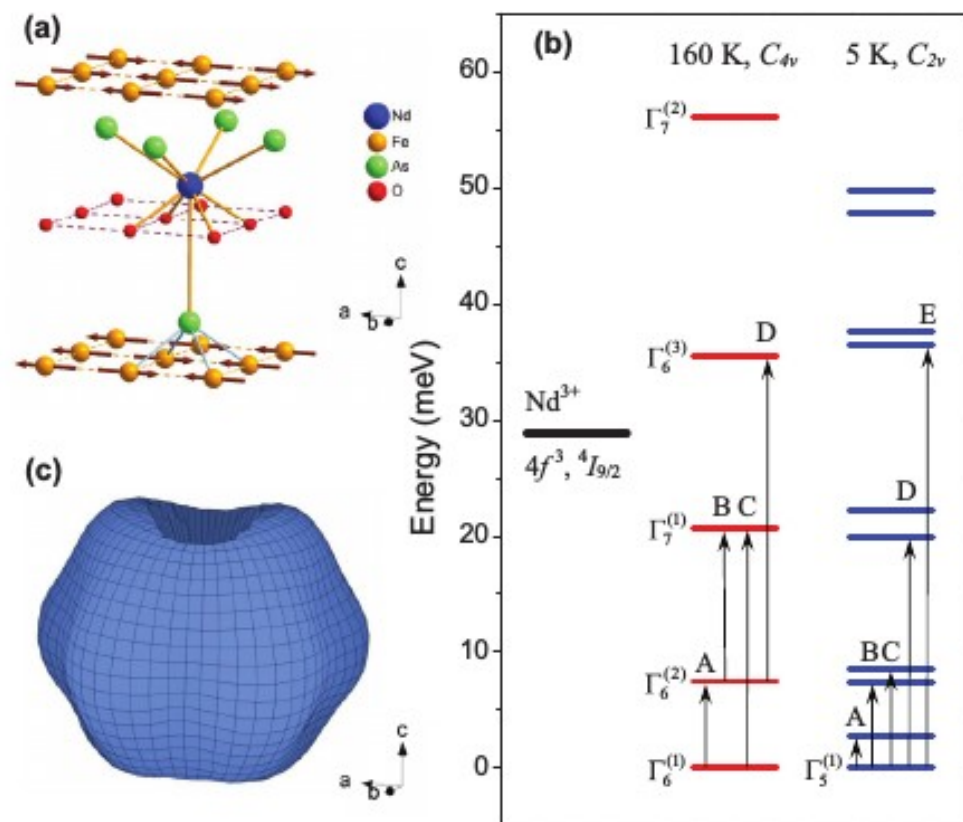
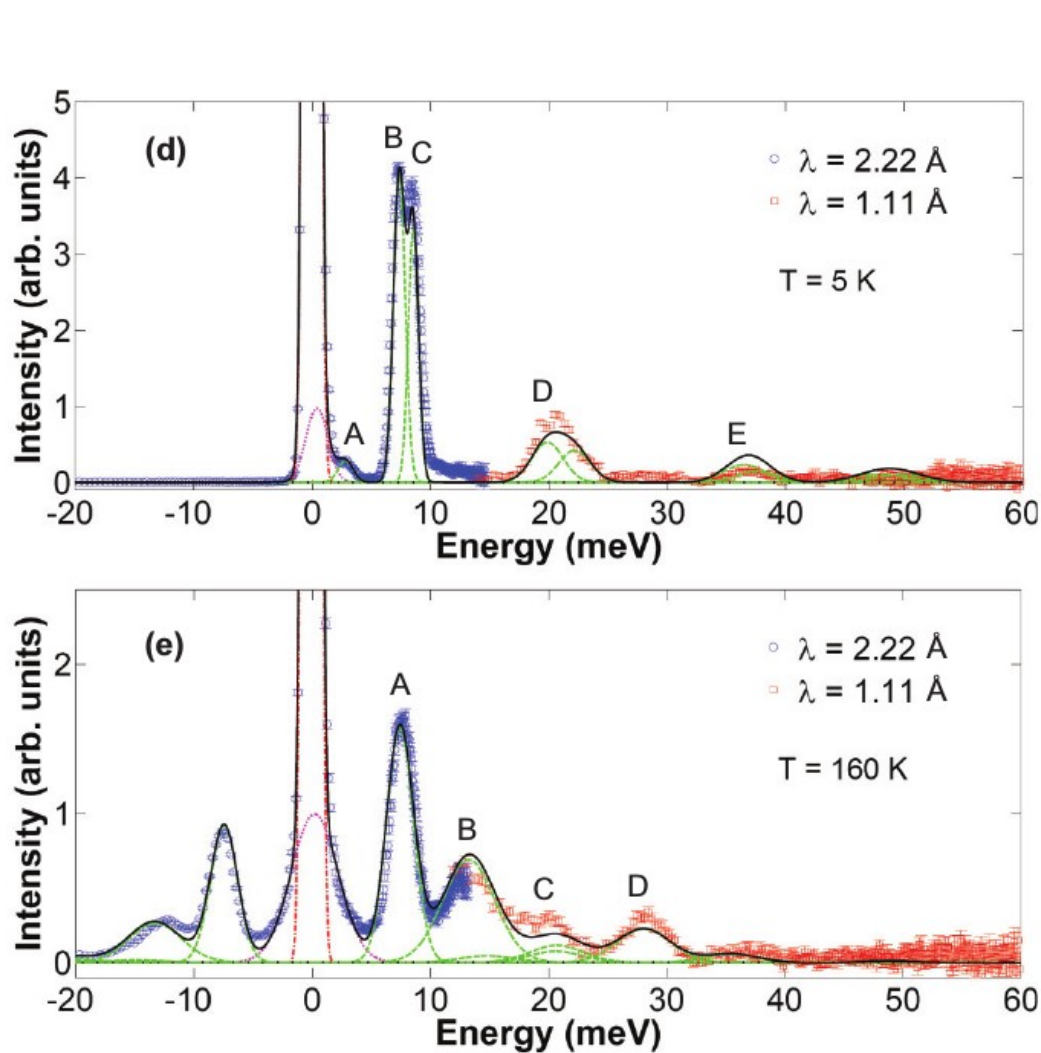


Short range correlations in frustrated beta-Mn



JAM. Paddison et al. PRL (2013)

Crystal field excitation(s)



$$\frac{d^2\sigma(i \rightarrow j)}{d\Omega dE'} = N \frac{k_f}{k_i} \left(\frac{\hbar\gamma e^2}{mc^2} \right)^2 e^{-2W} \left| \frac{1}{2} g_J F(\mathbf{Q}) \right|^2 \times \sum_{i,j} n_i |\langle j | J_{\perp} | i \rangle|^2 \delta(E_i - E_j + \hbar\omega),$$

Y. Xiao et al., PRB 88, 214419 (2013)

Spin excitations

Martin Mourigal et al., Nature Physics, 9, 435 (2013)

