What do we see with neutrons in magnetism ?

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NEUTRONS FOR SCIENCE®

The neutron

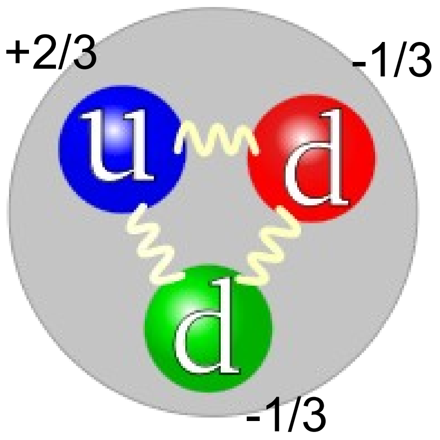
- Non-charged particle $m_N = 1.67.10^{-27}\, kg$
- Total angular momemtum (« nuclear spin ») **I=1/2**
- The <u>magnetic moment is extremely small</u> compared to the electron.

-> The interaction potential is small, Born approximation is valid

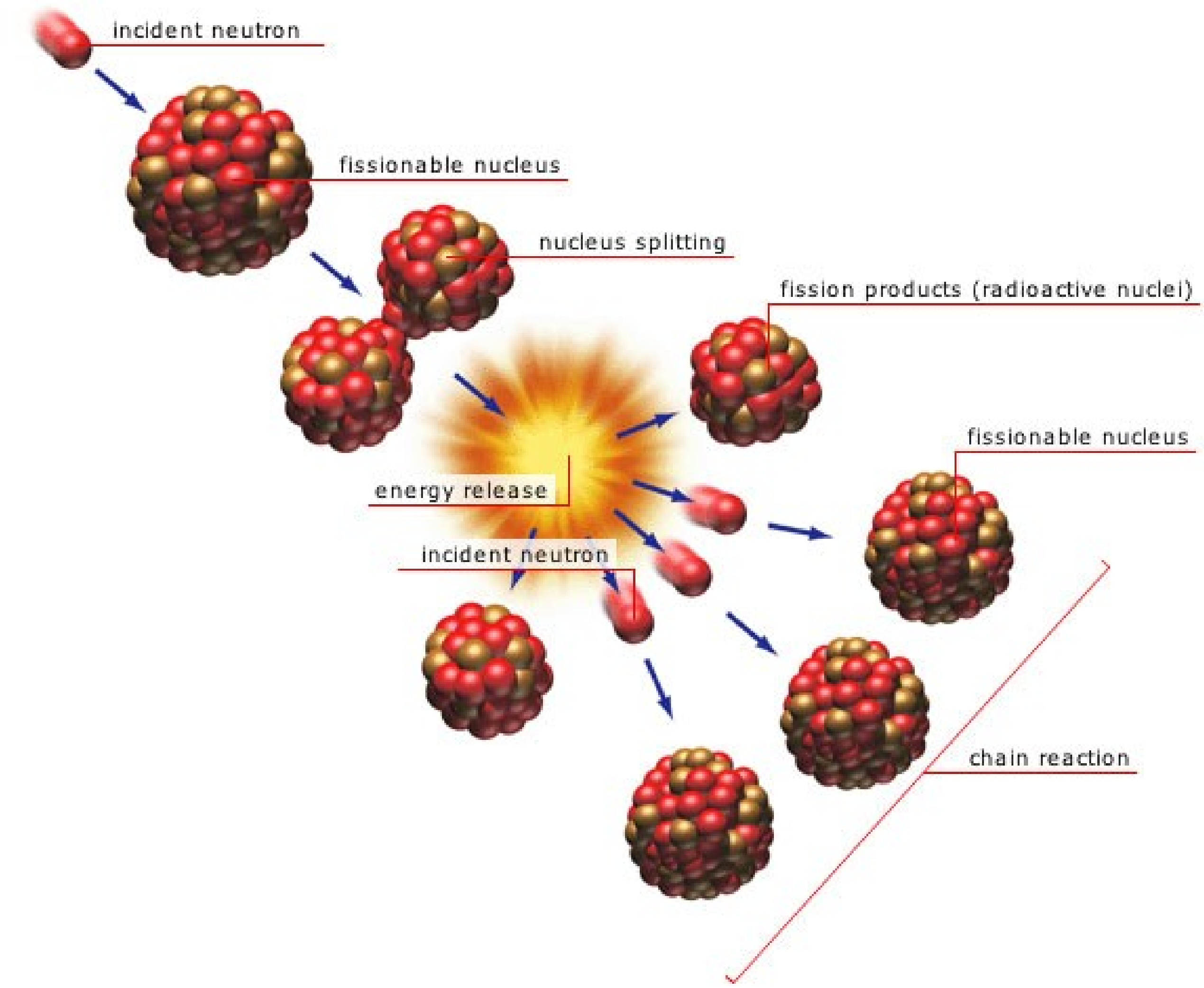
$$\boldsymbol{\mu} = \gamma \mu_N \boldsymbol{\sigma} \; with \gamma = -1.913$$

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

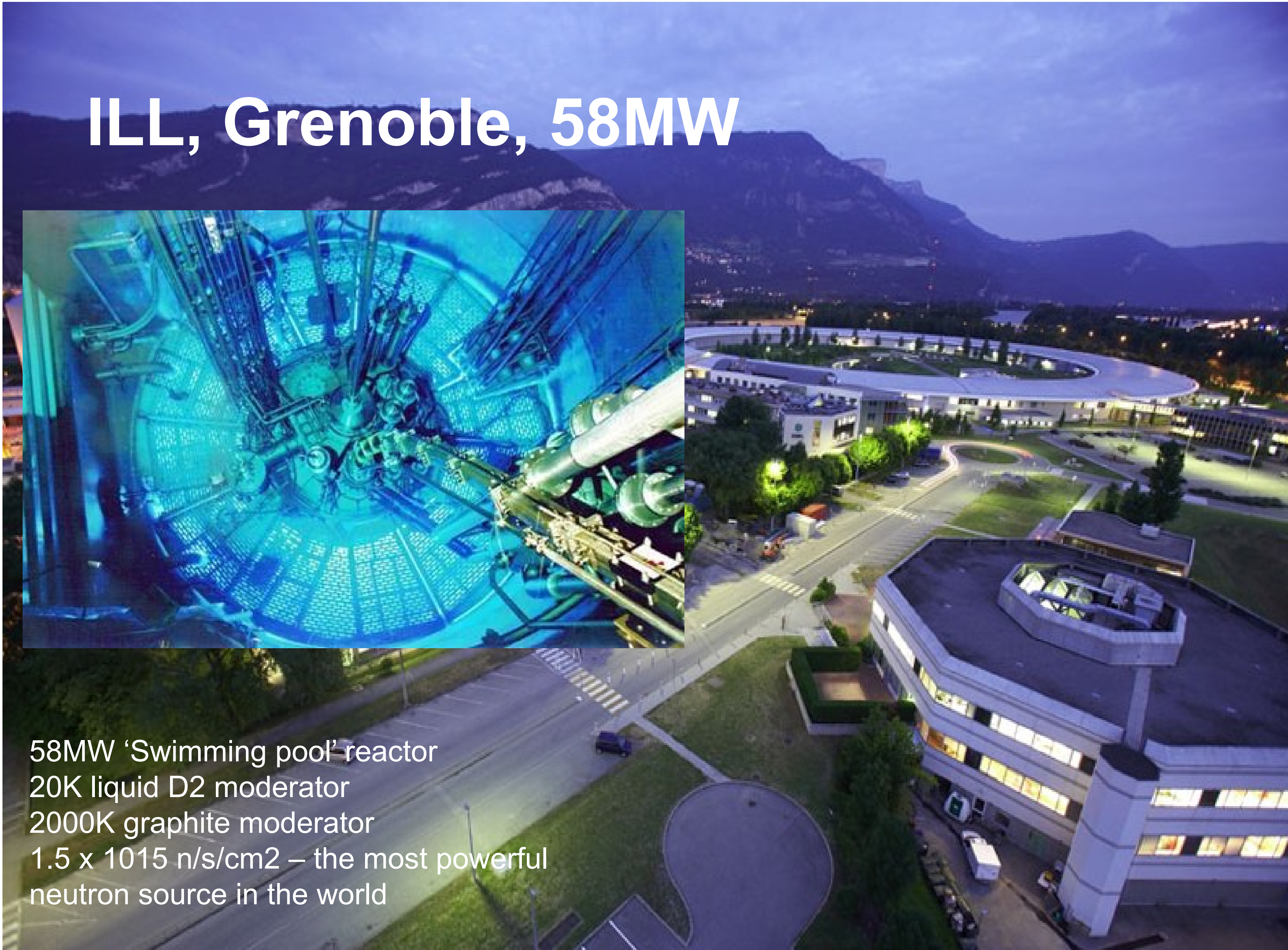
$$\mu_B = \frac{e\hbar}{2m_e} = 9.28\,10^{-24}\, J.T^{-1}$$



Production : Nuclear fission



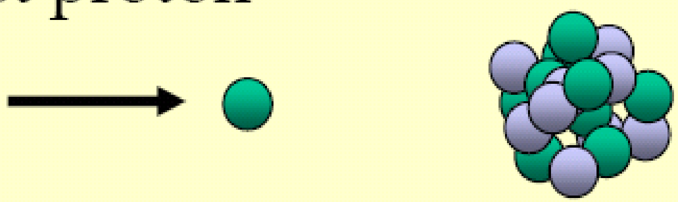
ILL, Grenoble, 58MW



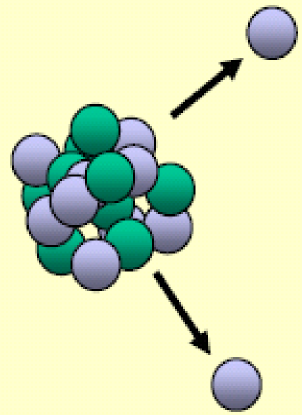
58MW ‘Swimming pool’ reactor
20K liquid D2 moderator
2000K graphite moderator
1.5 x 1015 n/s/cm2 – the most powerful
neutron source in the world

Production : spallation

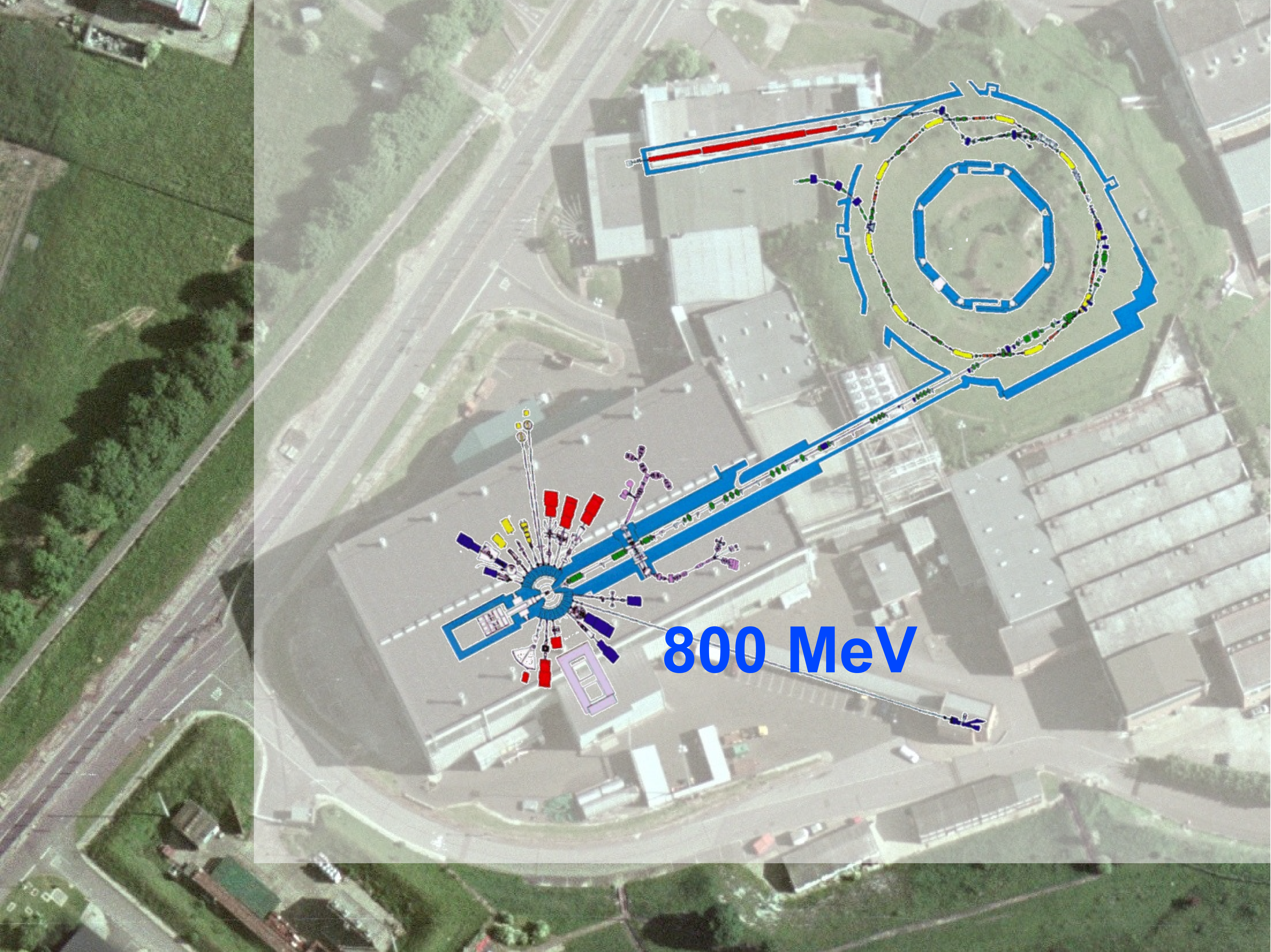
Fast proton



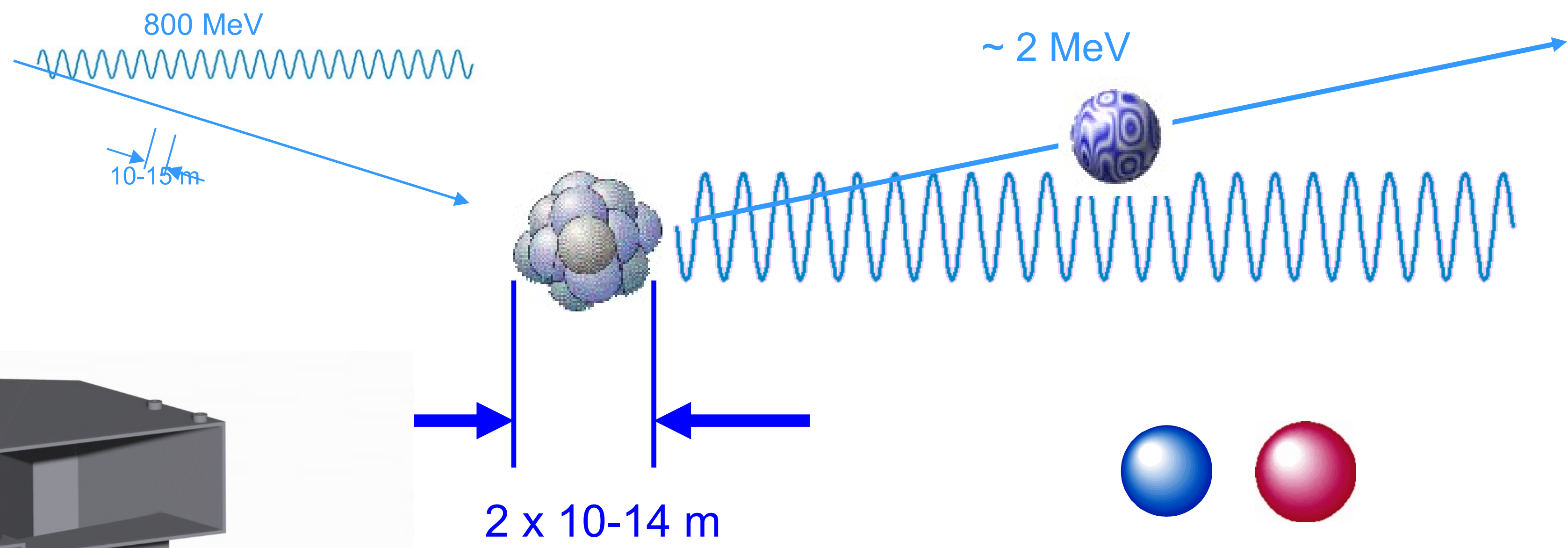
Heavy nucleus (Ta, U, Hg)

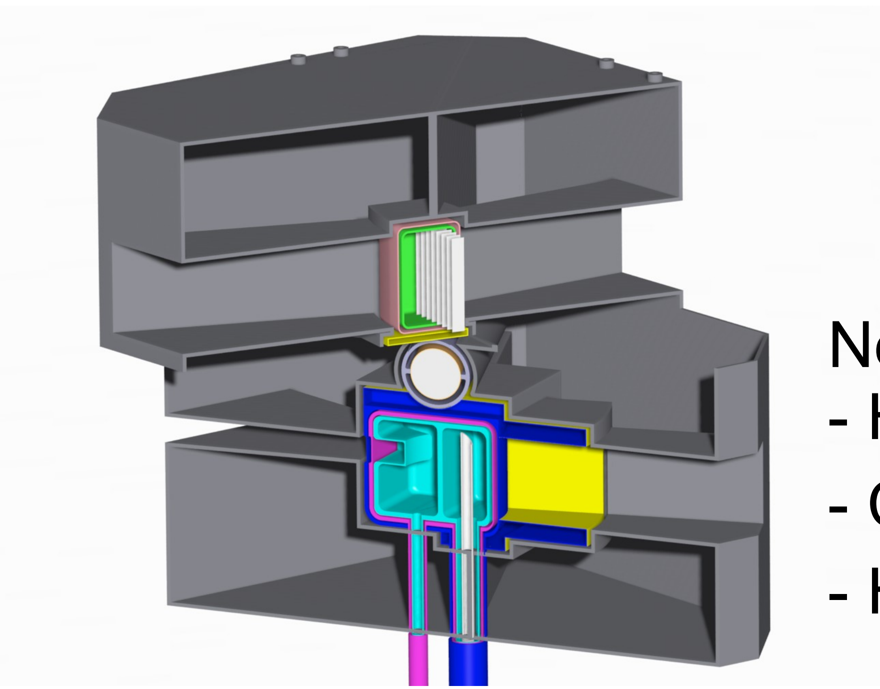


Fast Neutrons are slowed by collisions in a moderator (CH_4, H_2O, D_2O)


800 MeV

Production : spallation





Neutrons slowed down by « moderators » :

- H_2
- CH_4
- H_2O

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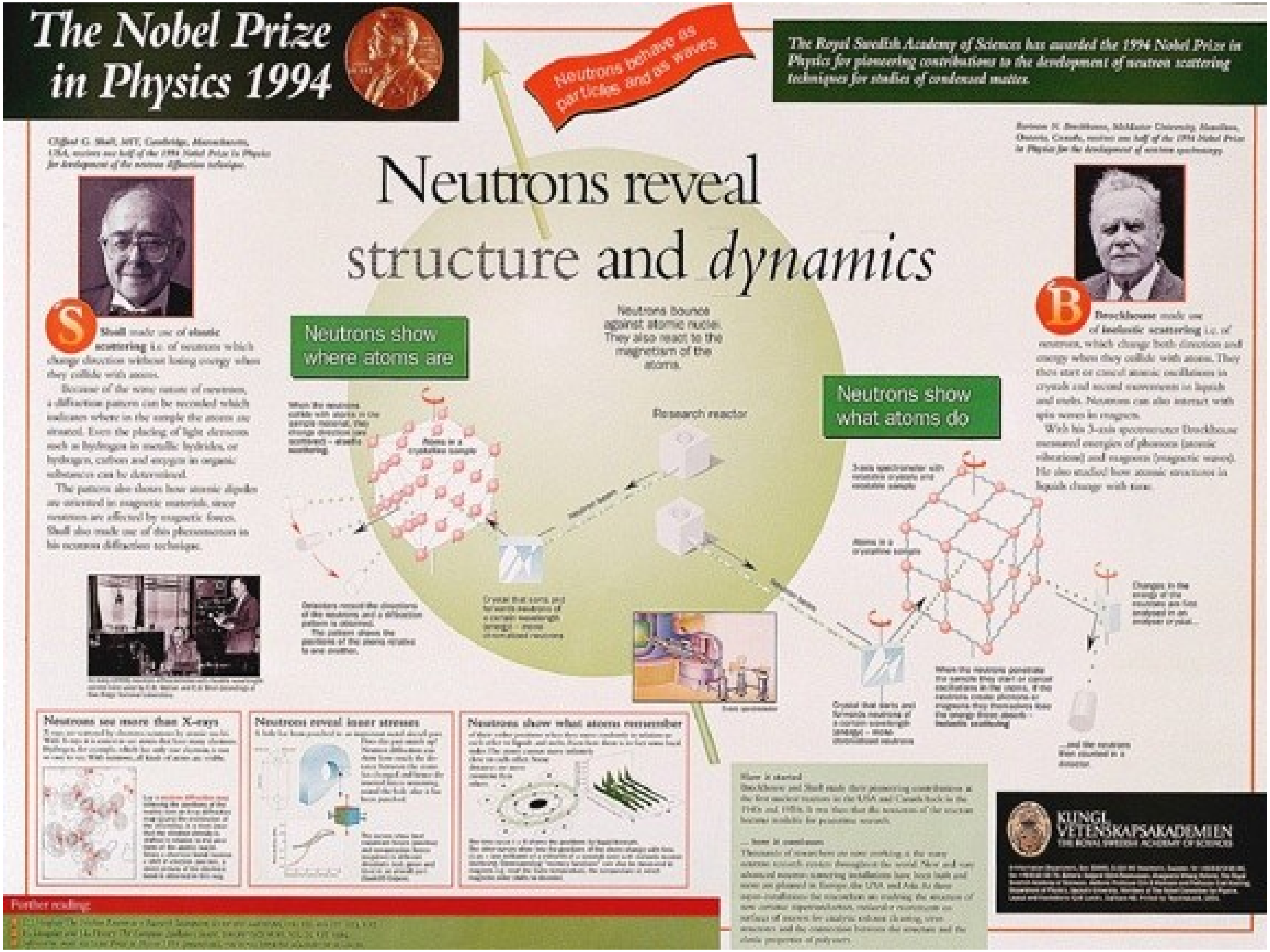
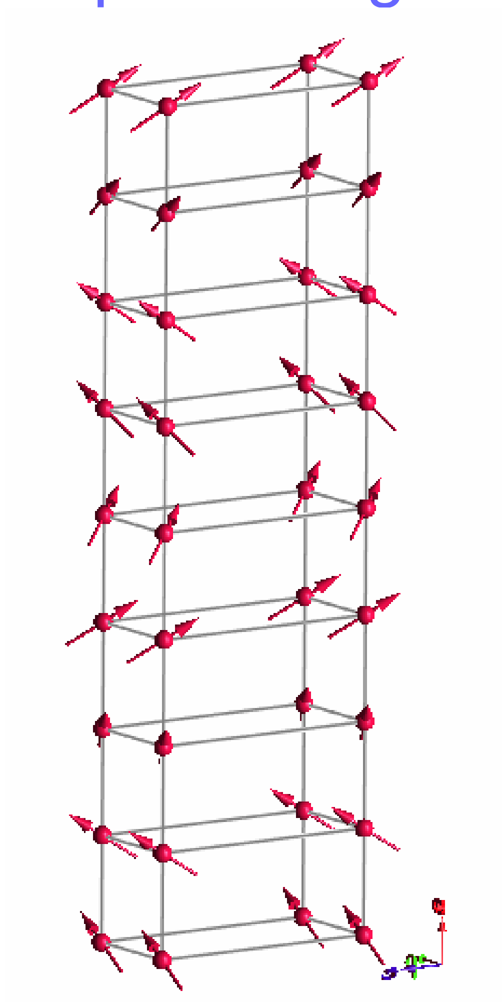
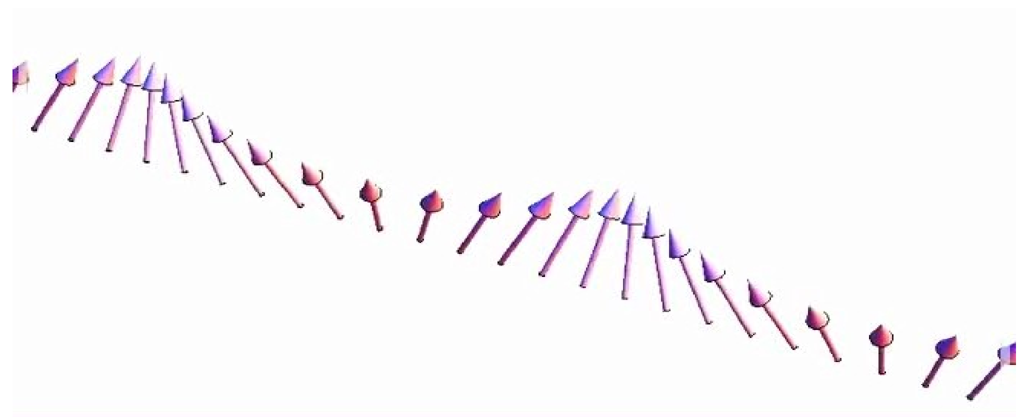


What do we see with neutrons in magnetism ?

Static spin arrangements



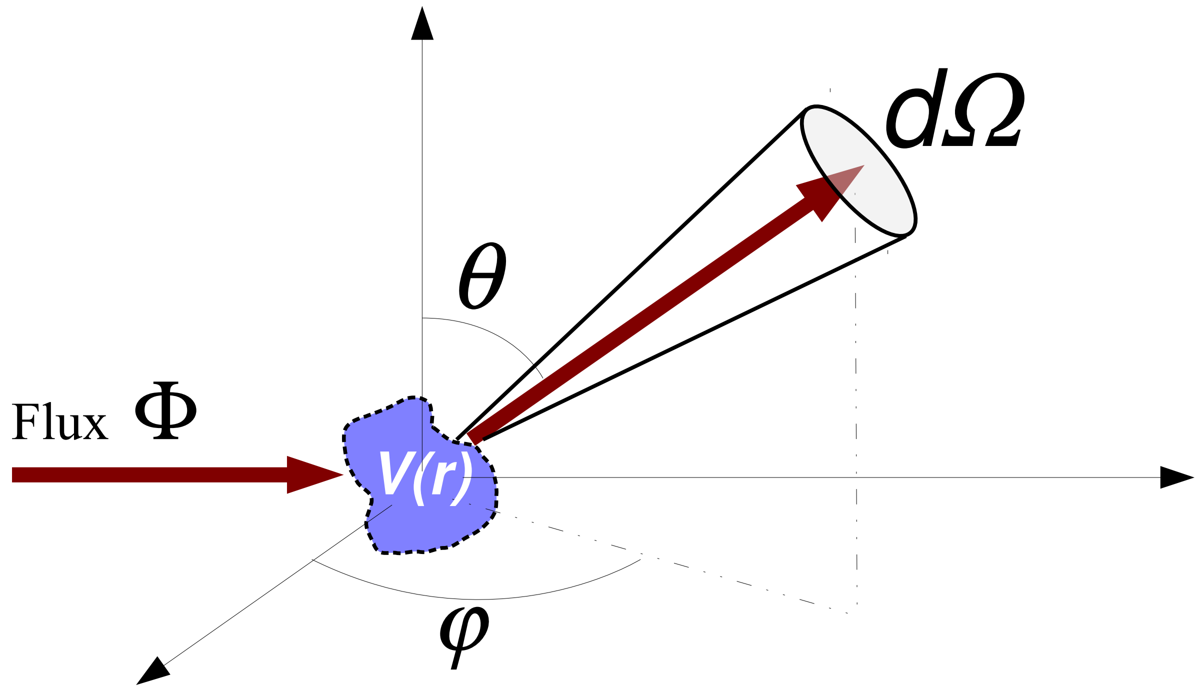
Local or collective excitations



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

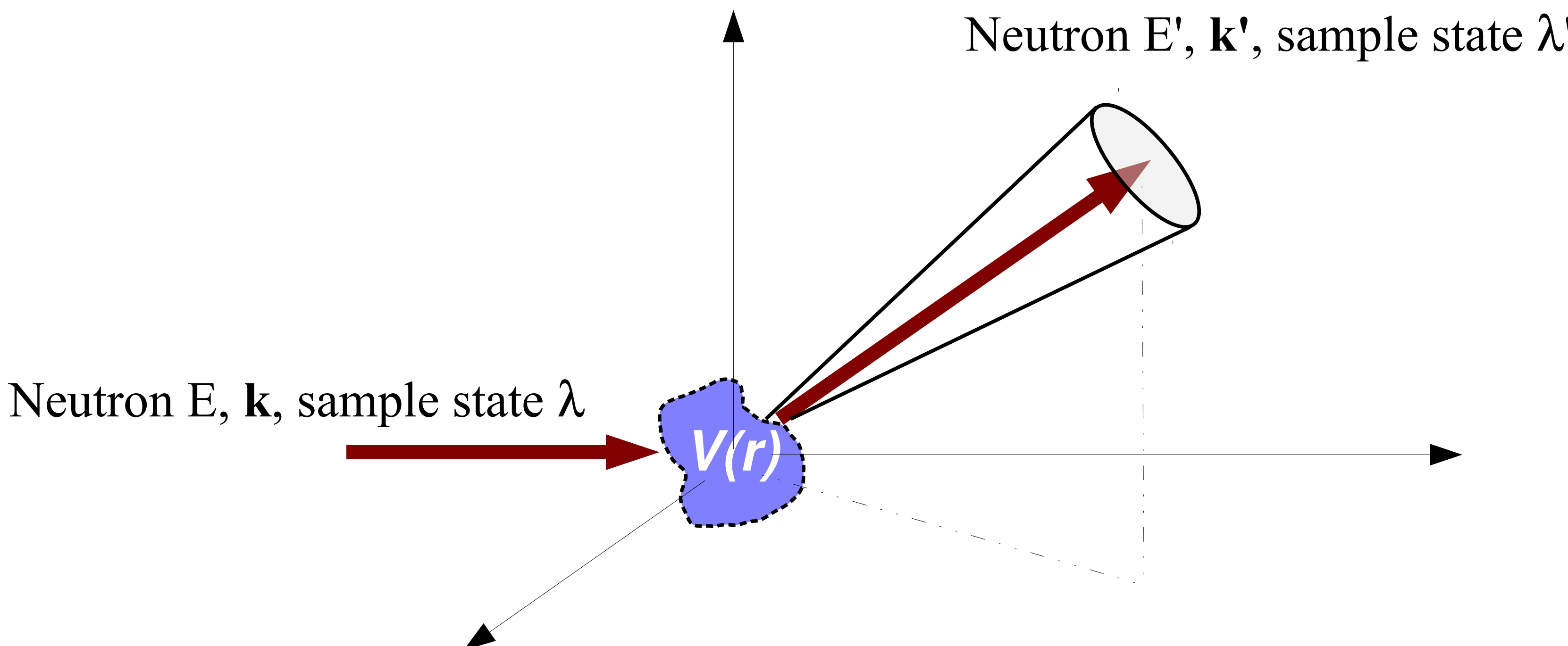
Scattering by a potential *V(r)*



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

- 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 φ(r) verify:

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

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

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

•One can expend 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.

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

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

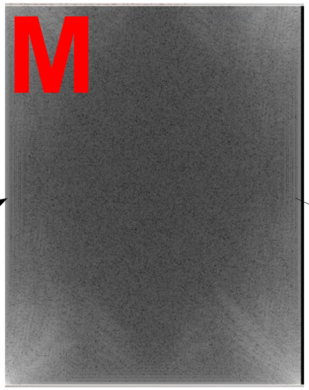
The phase problem

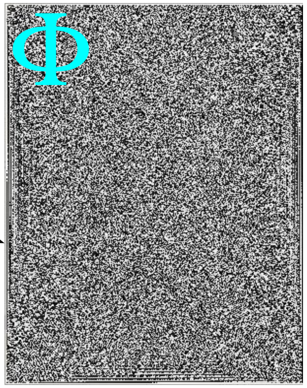
Loss of information in a physical measurement

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

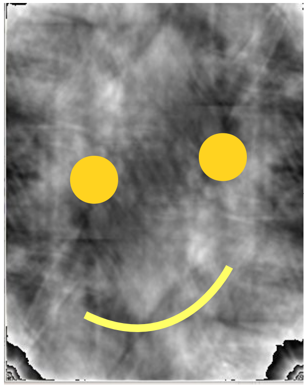
$$F(u,v)=\frac{1}{N_x N_y}\sum_x\sum_y f(x,y)\,e^{\left(-2\pi i\left(\frac{xu}{N_x}+\frac{yv}{N_y}\right)\right)}$$

James Chadwick



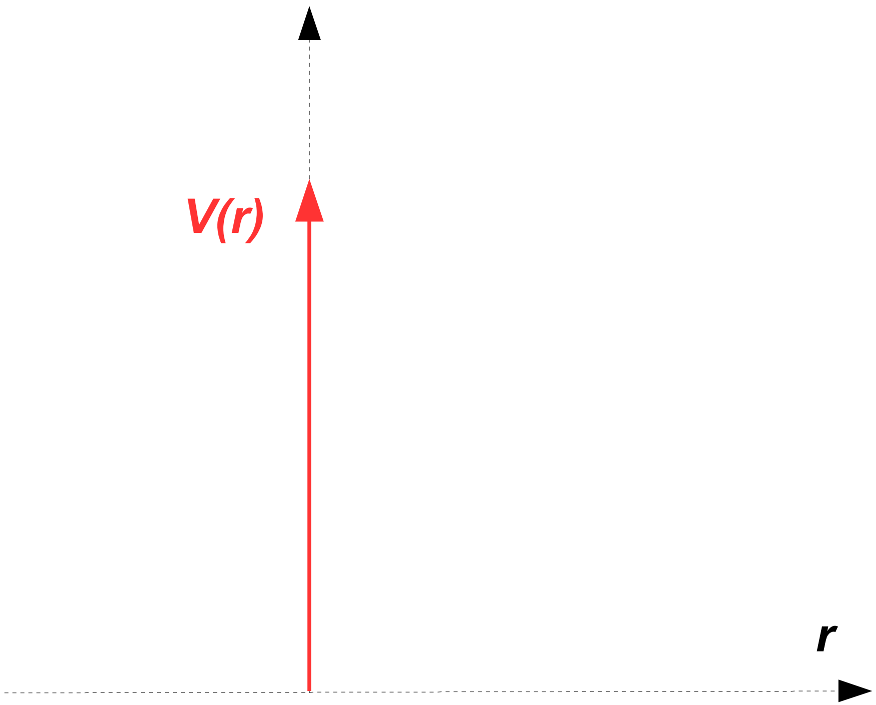


$$f'(x,y)=\frac{1}{N_u N_v}\sum_u\sum_v M(u,v)\,e^{\left(2\pi i\left(\frac{xu}{N}+\frac{yv}{N}\right)\right)}$$



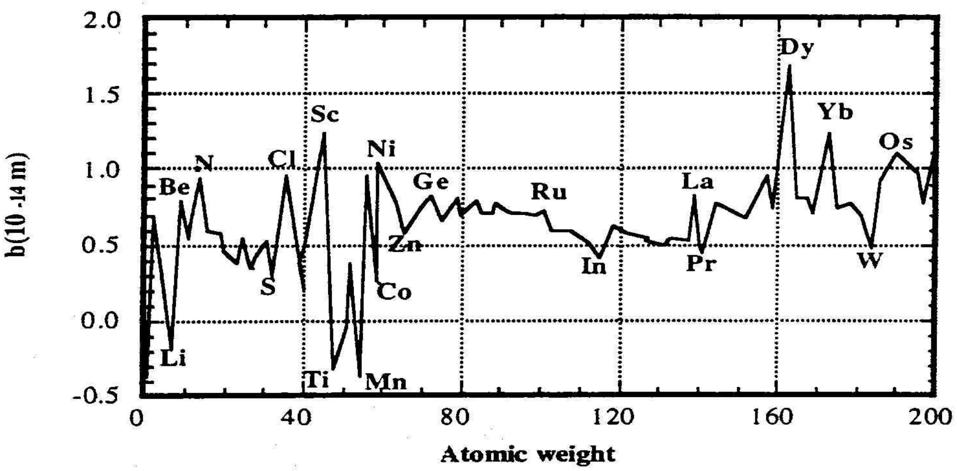
Nuclear scattering

- Nuclear scattering mediated by the strong force, extremely short range (fm=1.10^{-15} m).
- Neutron wavelength much larger (Å=1.10^{-10}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).

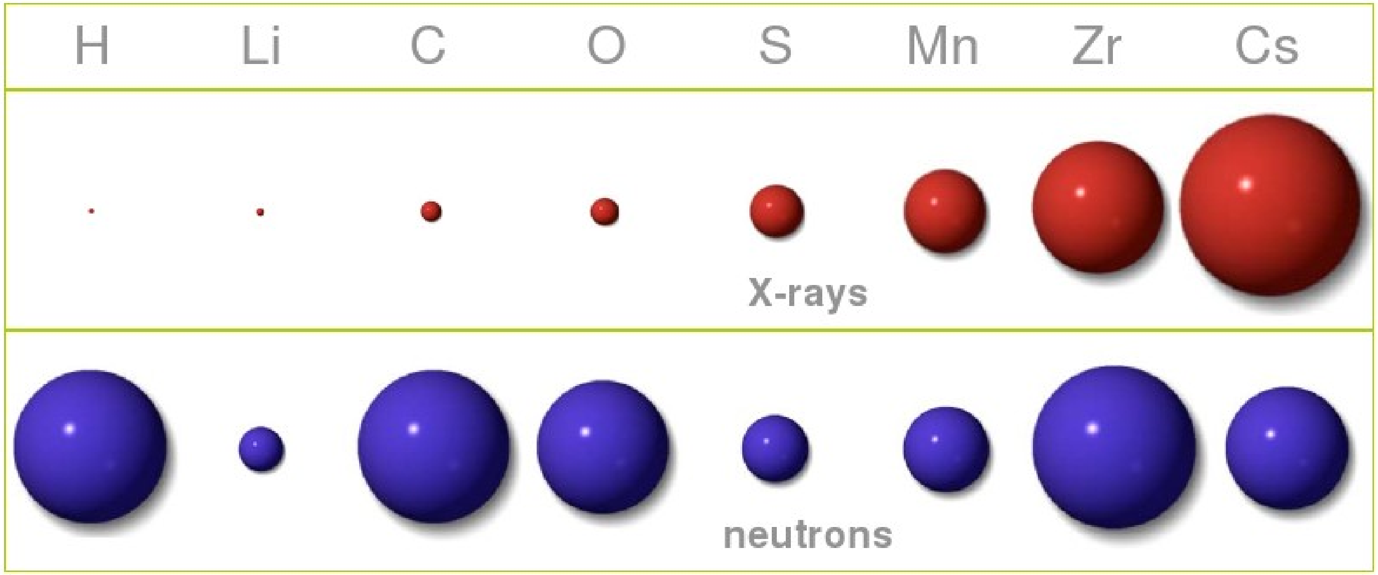


$$V(\vec{r}) = \frac{2\pi\hbar^2}{m_r} a\,\delta^3(\vec{r})$$

Scattering lengths



Typically a few fm



X-ray vs Neutron

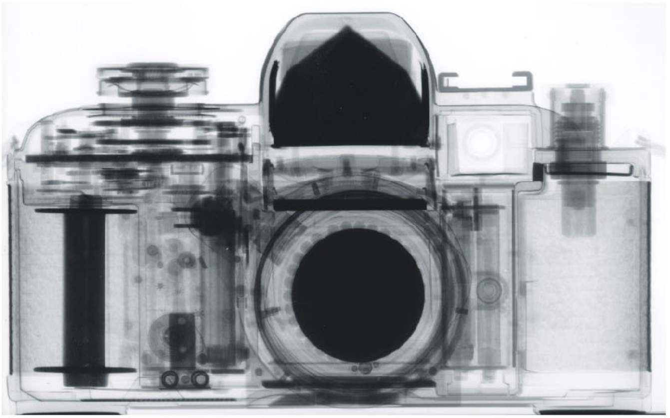


Fig. a: Neutron radiography of a camera

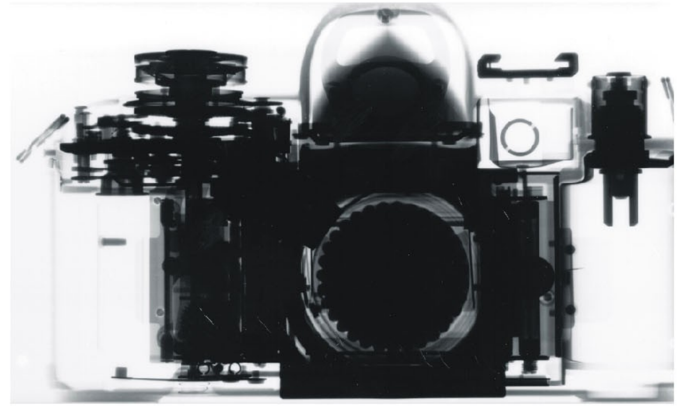


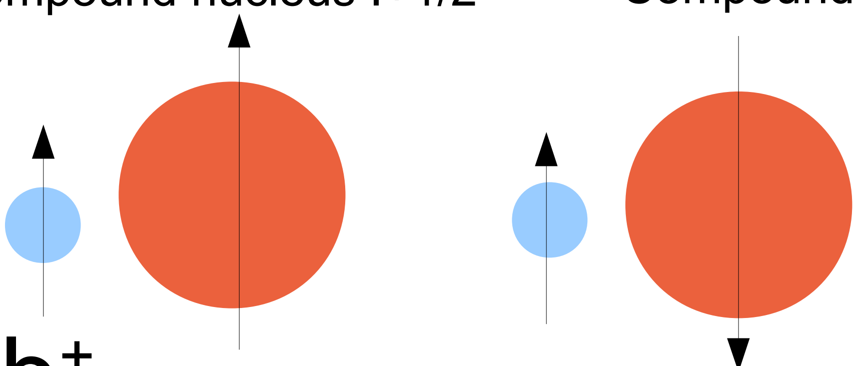
Fig. b: Radiographic image of a camera made X-rays

Also: - *b* depends on the isotope

- *b* depends on the spin states of the neutron and nucleus

Compound nucleus I+1/2

Compound nucleus I-1/2



b^+

b^-

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

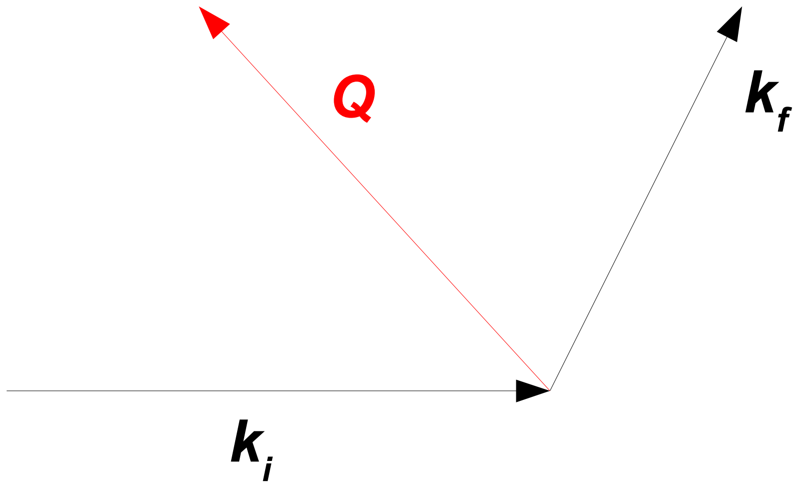
$$b_{inc} = \sqrt{\overline{b^2} - \bar{b}^2}$$

Magnetic cross section

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

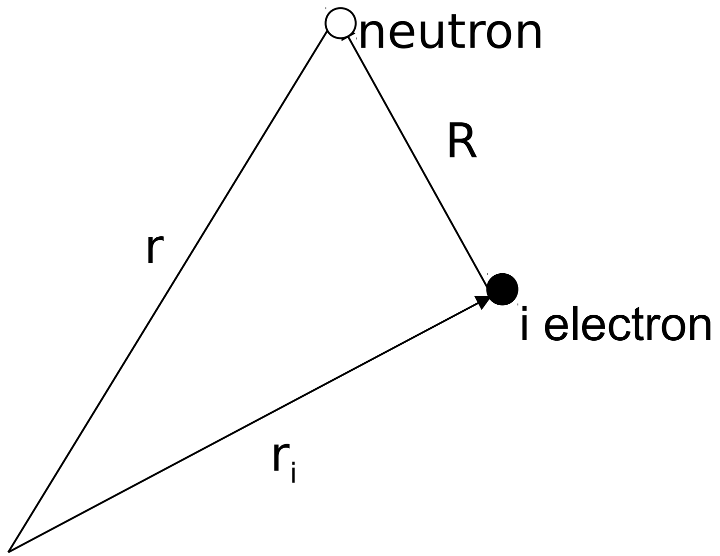
$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\sigma,\lambda\rightarrow\sigma',\lambda'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar}\right)^2 |\langle \mathbf{k}',\boldsymbol{\sigma}'|V_m|\mathbf{k},\boldsymbol{\sigma}\rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

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



Magnetic interaction (spin)

Considering a single unpaired electron:



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

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

$$V = -\boldsymbol{\mu} . \boldsymbol{B}$$

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

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

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

Magnetic interaction (spin)

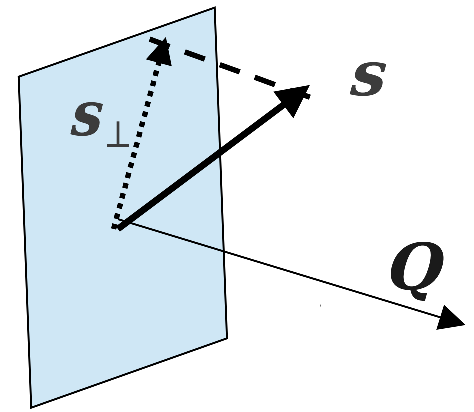
$$\nabla\times\boldsymbol{s}\times\nabla\frac{1}{|R|}=\frac{1}{2\pi^2}\int\frac{1}{\boldsymbol{x}^2}\nabla\times\boldsymbol{s}\times\nabla\, e^{i\boldsymbol{x}\boldsymbol{R}}d\boldsymbol{x}=\frac{1}{2\pi^2}\int\left(\hat{x}\times\boldsymbol{s}\times\hat{x}\right)e^{i\boldsymbol{x}\boldsymbol{R}}d\boldsymbol{x}$$

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

This quantity is the **projection of s perpendicular to Q:**

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

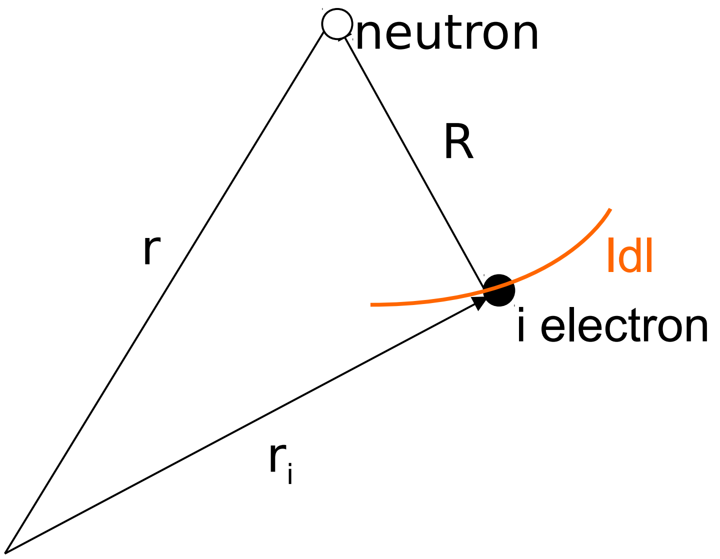
$$\left\langle\boldsymbol{k}'\middle|W_s\middle|\boldsymbol{k}\right\rangle=4\pi\hat{\boldsymbol{Q}}\times\boldsymbol{s}\times\hat{\boldsymbol{Q}}.e^{i.\boldsymbol{Q}.\boldsymbol{r}_i}$$



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

Magnetic interaction (orbital)

$$\boldsymbol{B}(\boldsymbol{R}) = \frac{\mu_0}{4\pi}\frac{I\boldsymbol{dl}\times\hat{\boldsymbol{R}}}{R^2} = \frac{\mu_0 e}{4\pi m_N}\frac{\boldsymbol{p_i}\times\hat{\boldsymbol{R}}}{R^2} = \frac{2\mu_0\mu_B}{4\pi\hbar}\frac{\boldsymbol{p_i}\times\hat{\boldsymbol{R}}}{R^2}$$



$$V_L = \gamma\mu_N 2\mu_B\frac{\mu_0}{4\pi\hbar}\boldsymbol{\sigma}.\boldsymbol{p_i}\times\nabla\frac{1}{|\boldsymbol{R}|} = cste\,\boldsymbol{\sigma}.\boldsymbol{W_L}$$

$$\boldsymbol{W_L} = \frac{1}{\hbar}\frac{\boldsymbol{p_i}\times\hat{\boldsymbol{R}}}{R^2}$$

Use the Fourier transform:

$$\int\frac{\hat{R}}{R^2}e^{i\kappa R} = 4\pi i\frac{\boldsymbol{\kappa}}{\kappa}$$

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

Magnetic interaction strength

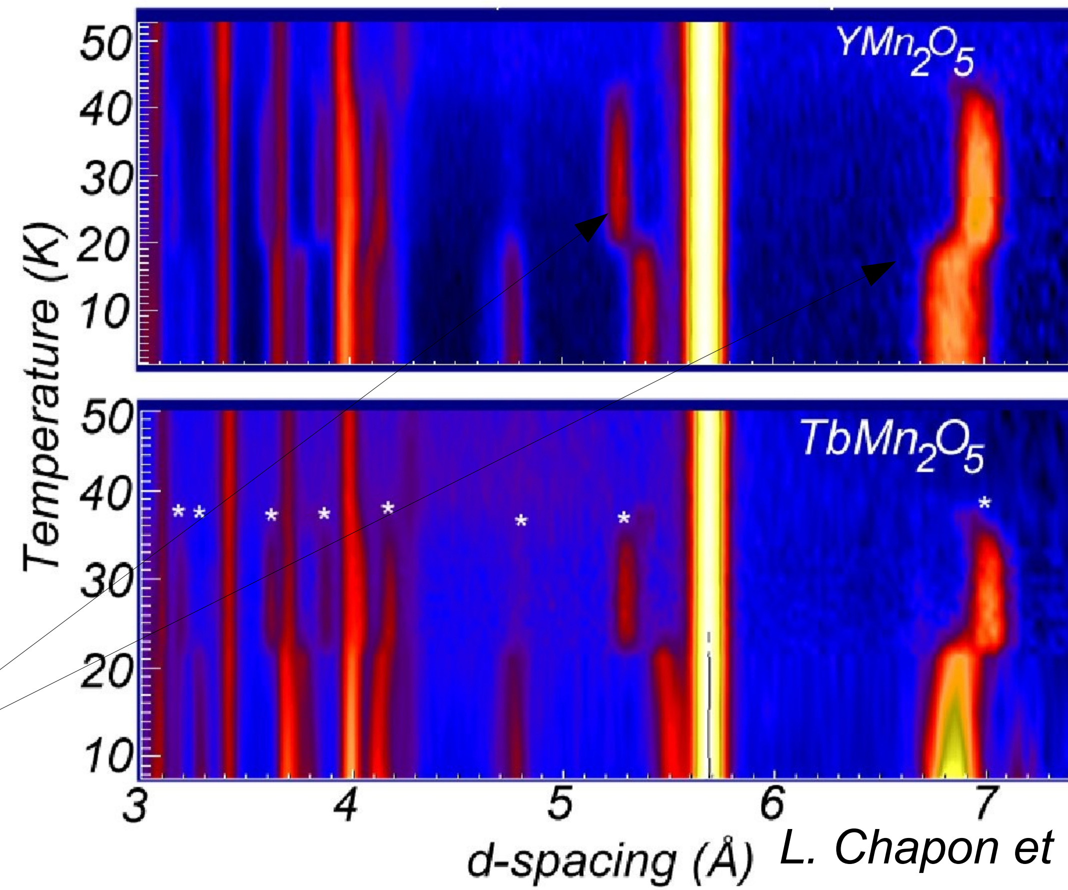
Collecting the **pre-factors** .....

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

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

r_0: free electron radius =2.8.10^{-15} m

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



Magnetic scattering

L. Chapon et al.

(Spin + orbital) magnetic scattering

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

Defining:

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

The spin and orbital part of $M_\perp$ are the transverse components of the Fourier transform of the spin and orbital magnetization density

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

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:

$$\langle A \rangle = tr(\rho A)$$

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 ½ particle

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

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

Spin operators:

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

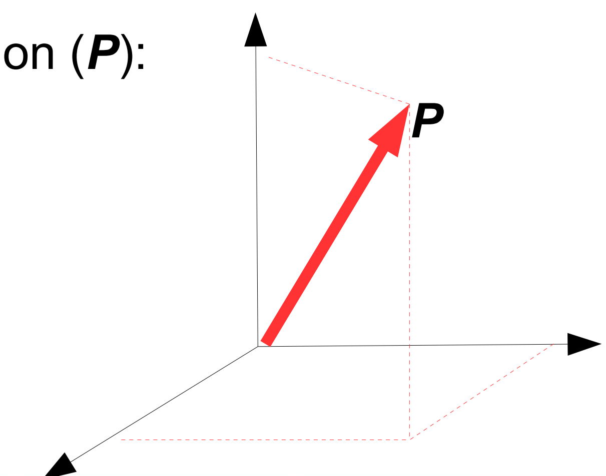
$\hat{S}_-|\uparrow\rangle = |\downarrow\rangle \quad \hat{S}_-|\downarrow\rangle = 0 \quad \hat{S}_z|\downarrow\rangle = -1/2|\downarrow\rangle \quad \hat{S}_- = \hat{S}_x - i\hat{S}_y$

Pauli spin operators and matrices:

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

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

Density matrix representing the incident neutron beam polarization (***P***):



$$\hat{\rho} = \frac{1}{2}\left(I + \boldsymbol{P}.\hat{\boldsymbol{\sigma}}\right) = \frac{1}{2}\left(I + Px.\hat{\sigma}_x + Py.\hat{\sigma}_y + Pz.\hat{\sigma}_z\right)$$

Magnetic interaction(elastic case)

A very easy way to averaging over all spin states:

Scattered intensity:

$$\frac{d\sigma}{d\Omega}=(\gamma r_0)^2 tr\left[(\boldsymbol{M}_\perp.\boldsymbol{\sigma})\rho(\boldsymbol{M}_\perp.\boldsymbol{\sigma})^+\right]$$

Final polarization:

$$\boldsymbol{P}_f.\frac{d\sigma}{d\Omega}=(\gamma r_0)^2 tr\left[\boldsymbol{\sigma}_f(\boldsymbol{M}_\perp.\boldsymbol{\sigma})\rho(\boldsymbol{M}_\perp.\boldsymbol{\sigma})^+\right]$$

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,

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

Magnetic interaction (polarized)

$$I = \frac{1}{2} tr \left[\left(N\mathbb{1} + \boldsymbol{M}_{\perp} . \boldsymbol{\sigma} \right) \left(\mathbb{1} + \boldsymbol{P_i} . \boldsymbol{\sigma} \right) \left(N^{*}\mathbb{1} + \boldsymbol{M}_{\perp}^{*} . \boldsymbol{\sigma} \right) \right]$$

$$I = NN^{*} + \boldsymbol{M}_{\perp} . \boldsymbol{M}_{\perp}^{*} + \boldsymbol{P_i} . (N\boldsymbol{M}_{\perp}^{*} + N^{*}\boldsymbol{M}_{\perp}) + i\boldsymbol{P_i} \left(\boldsymbol{M}_{\perp}^{*} \times \boldsymbol{M}_{\perp} \right)$$

$$\boldsymbol{P_f} . I = \frac{1}{2} tr \left[\boldsymbol{\sigma} \left(N\mathbb{1} + \boldsymbol{M}_{\perp} . \boldsymbol{\sigma} \right) \left(\mathbb{1} + \boldsymbol{P_i} . \boldsymbol{\sigma} \right) \left(N^{*}\mathbb{1} + \boldsymbol{M}_{\perp}^{*} . \boldsymbol{\sigma} \right) \right] \qquad (15)$$

$$\boldsymbol{P_f} I = \boldsymbol{P_i} \left(NN^{*} - \boldsymbol{M}_{\perp} \boldsymbol{M}_{\perp}^{*} \right) + \left(\boldsymbol{P_i} . \boldsymbol{M}_{\perp} \right) \boldsymbol{M}_{\perp}^{*} + \left(\boldsymbol{P_i} . \boldsymbol{M}_{\perp}^{*} \right) \boldsymbol{M}_{\perp} - i \left(N\boldsymbol{M}_{\perp}^{*} - N^{*}\boldsymbol{M}_{\perp} \right) \times \boldsymbol{P_i}$$

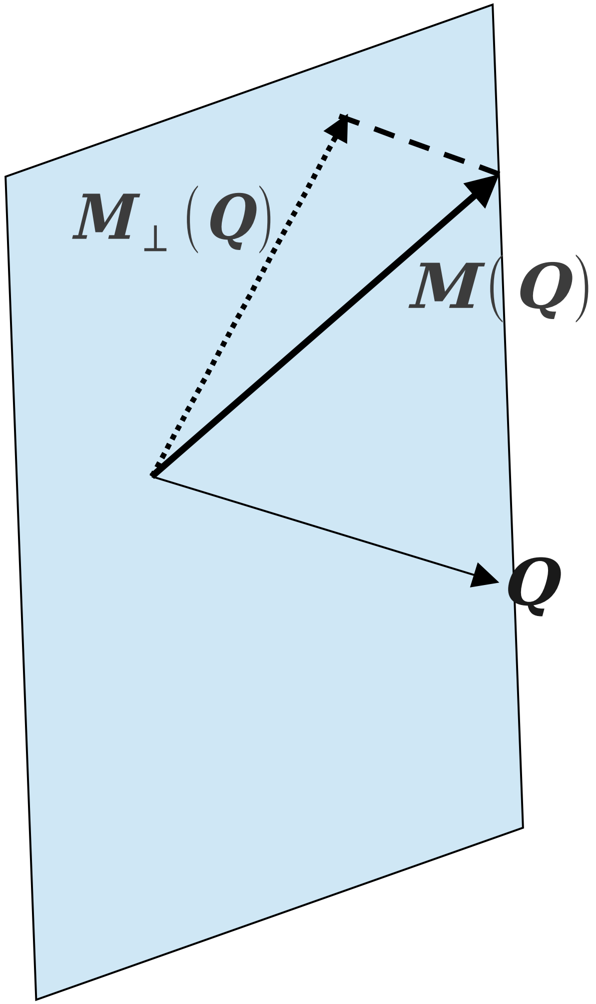
$$+ \left(N\boldsymbol{M}_{\perp}^{*} + N^{*}\boldsymbol{M}_{\perp} \right) - i \left(\boldsymbol{M}_{\perp}^{*} \times \boldsymbol{M}_{\perp} \right)$$

Blume, M.

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

Magnetic "extinction"

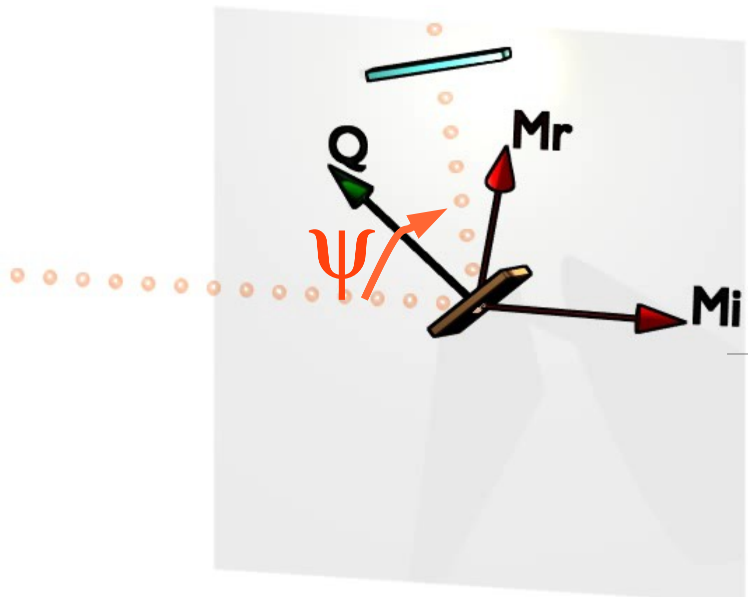
- From the projection operation emerges a very important extinction condition (if M parallel to Q, scattering is null)
- However, the only directional information about **M**(**Q**) comes from the projection operation, so great loss of information from this projection. We are **only** sensitive to the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

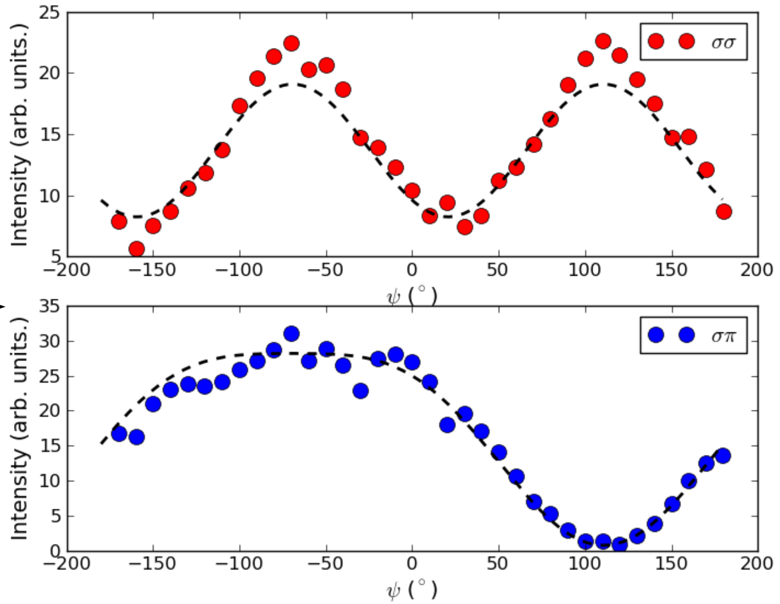


Magnetic "extinction" (comparison with X-ray)

In non-resonant X-ray magnetic scattering, the cross-section depends upon projections on $\mathbf{k_i}$, $\mathbf{k_f}$

<u>Consequence: signal depends on the azimuthal angle</u>



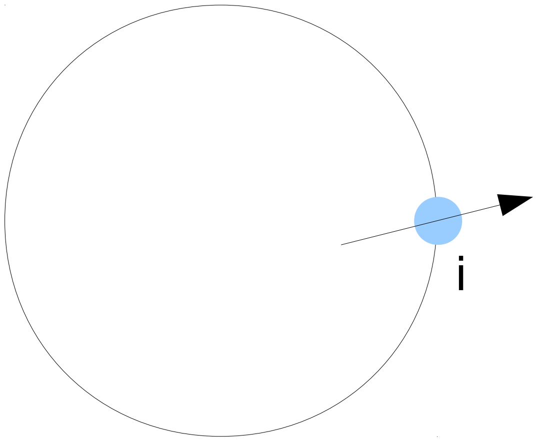


Scattering amplitude (σσ): $-i.\,\boldsymbol{M}(\boldsymbol{Q}).\,\widehat{\boldsymbol{k}}_i \times \widehat{\boldsymbol{k}}_f$

Scattering amplitude (πσ): $-2.\mathrm{i}.\sin^2(\theta).\,\boldsymbol{M}(\boldsymbol{Q}).\,\widehat{\boldsymbol{k}}_i$

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:



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

Magnetic structure factor (complex vector)

- Magnetic form factor: $f_j(\boldsymbol{Q}) = \frac{1}{|\boldsymbol{m}_j|} \int \boldsymbol{m}_j(\boldsymbol{R}) e^{i\boldsymbol{Q}\boldsymbol{R}} d\boldsymbol{R}$
- Thermal parameter (Debye-Waller factor): $T_j(\boldsymbol{Q})$

$$\boldsymbol{M}_\perp(\boldsymbol{Q}) = \hat{\boldsymbol{Q}} \times \boldsymbol{M}(\boldsymbol{Q}) \times \hat{\boldsymbol{Q}} = \boldsymbol{M}(\boldsymbol{Q}) - (\boldsymbol{M}(\boldsymbol{Q}).\hat{\boldsymbol{Q}}).\hat{\boldsymbol{Q}}$$

Magnetic interaction vector (complex vector)

Magnetic form factor

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

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

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

Using the addition theorem:

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

with:

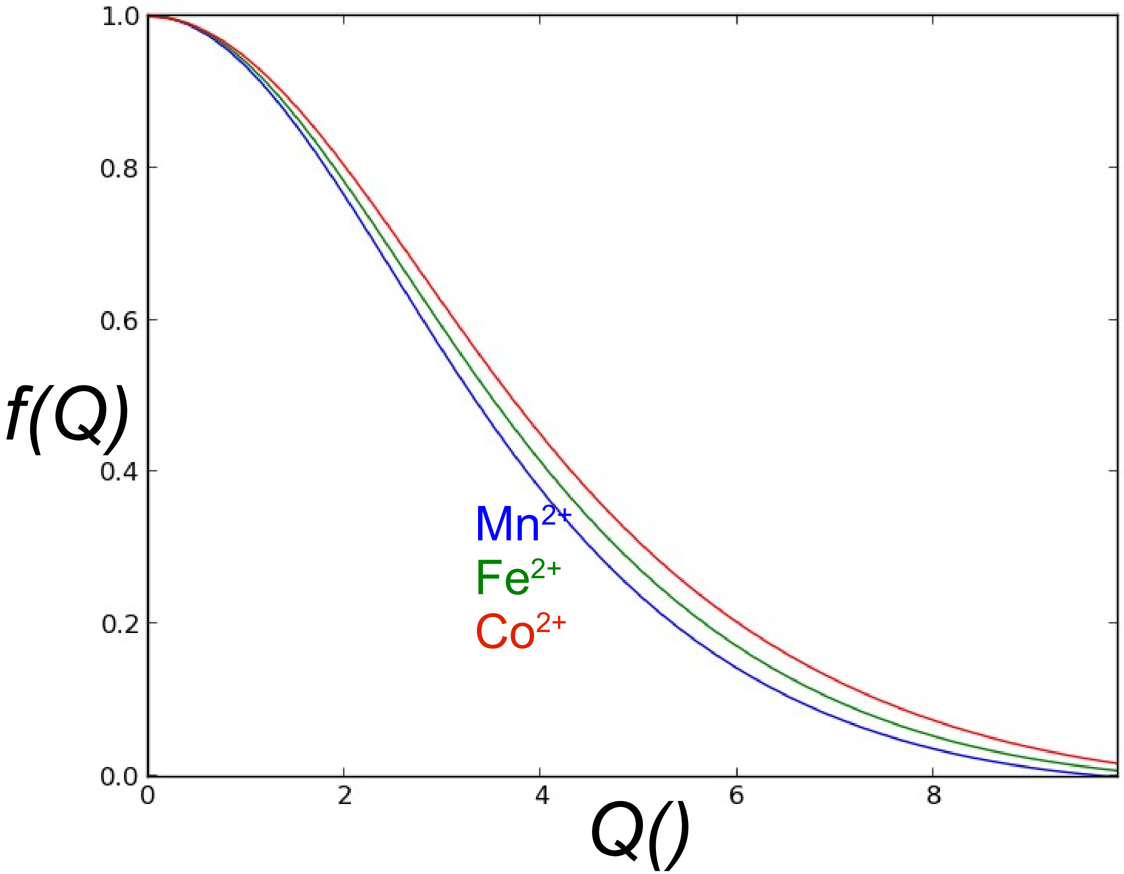
$$\langle j_L(Q.R) \rangle = \int R_L^2(R) . j_L(Q.R) R^2 dR$$

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

Magnetic form factor (Dipolar limit, j0 term)

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

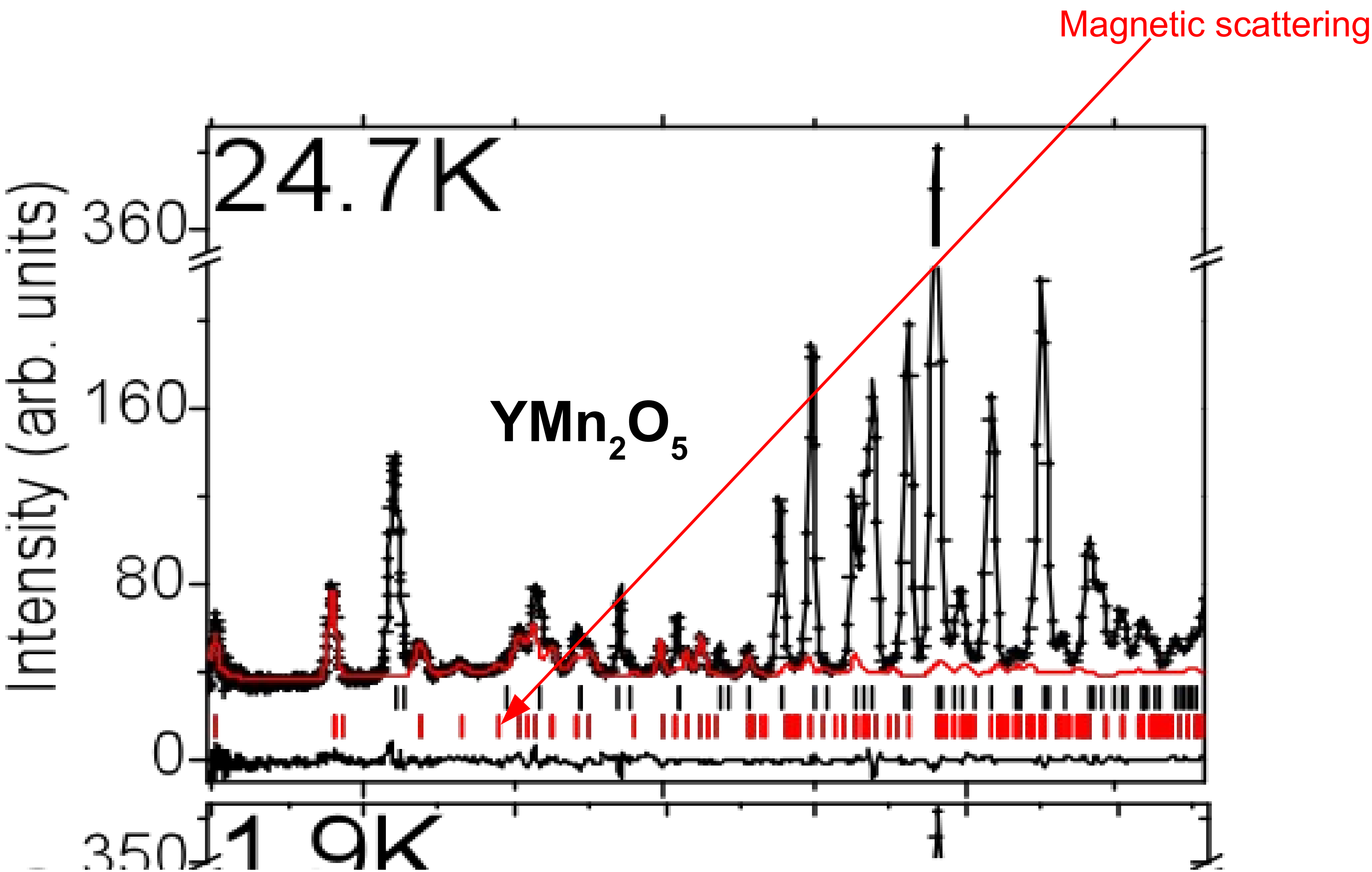
Calculated by Hartree-Fock method using Slater type orbitals then fitted using analytic approximations (expansion in exponentials)



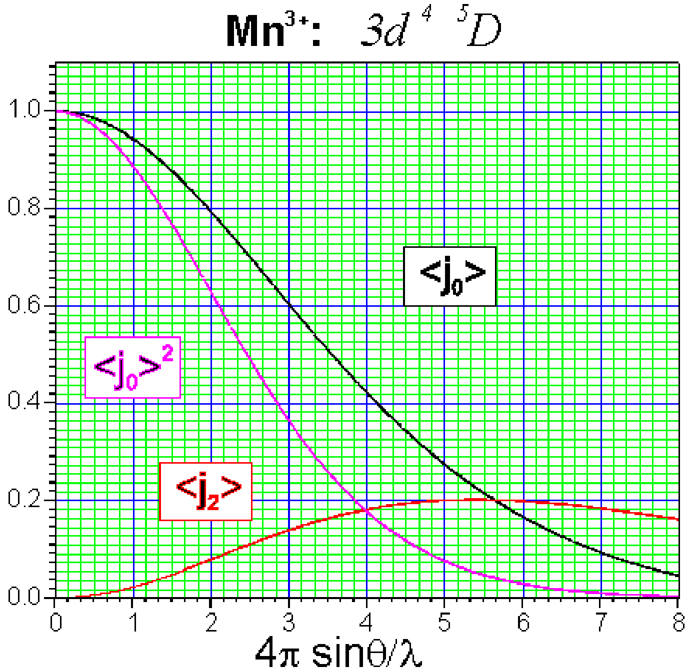
$$\langle j_l(s)\rangle = s^2\left(A_l \exp\{-a_l s^2\} + B_l \exp\{-b_l s^2\} + C_l \exp\{-c_l s^2\} + D_l\right)$$

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

Magnetic form factor (example)



Magnetic form factor (dipolar limit, j0,j2)

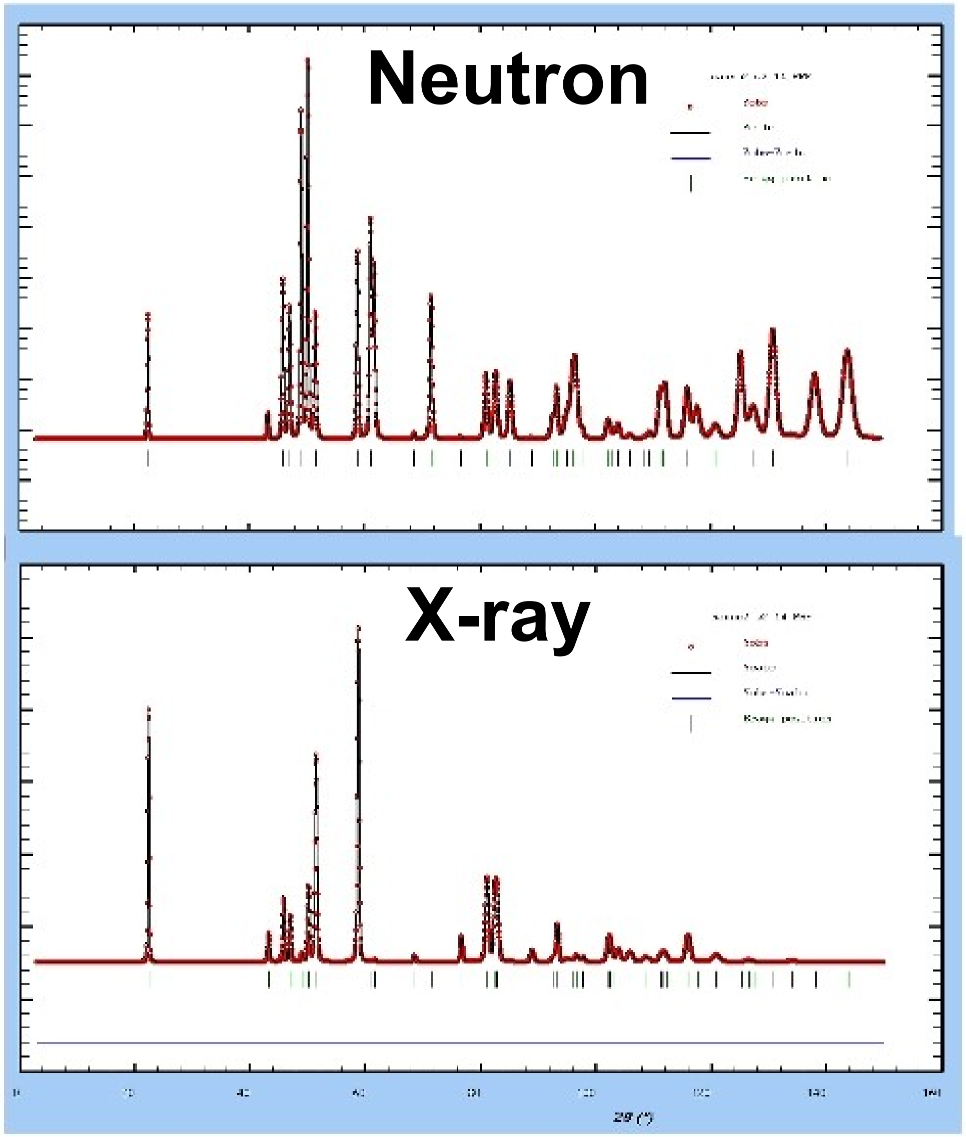


In the dipole approximation:

$$f(Q) = < j_0(Q) > + (1 - \frac{2}{g}) < j_2(Q) >$$

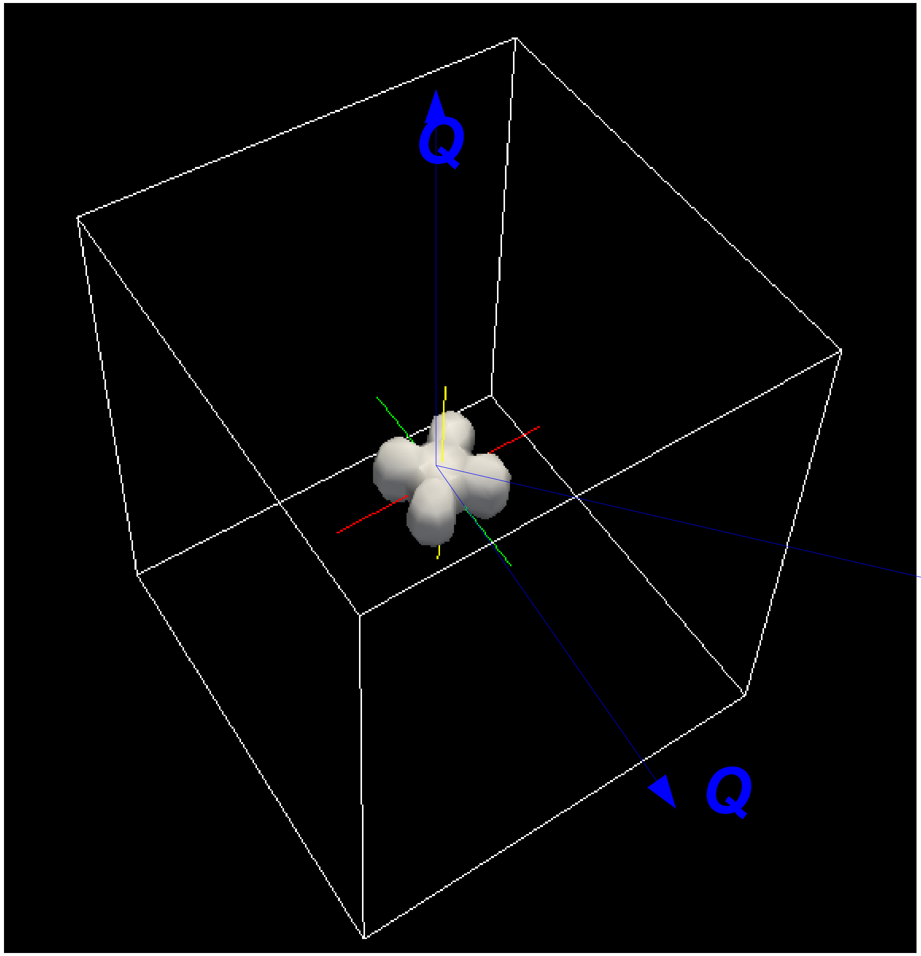
International Tables of Crystallography, Volume C, ed. by AJC Wilson, Kluwer Ac. Pub., 1998, p. 513

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



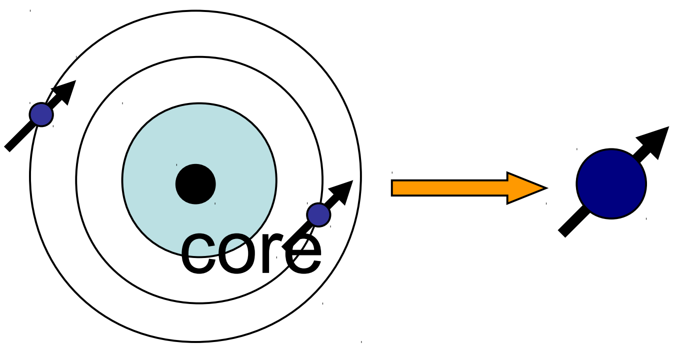
Form factor depends not only on the modulus of Q but also the direction.

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

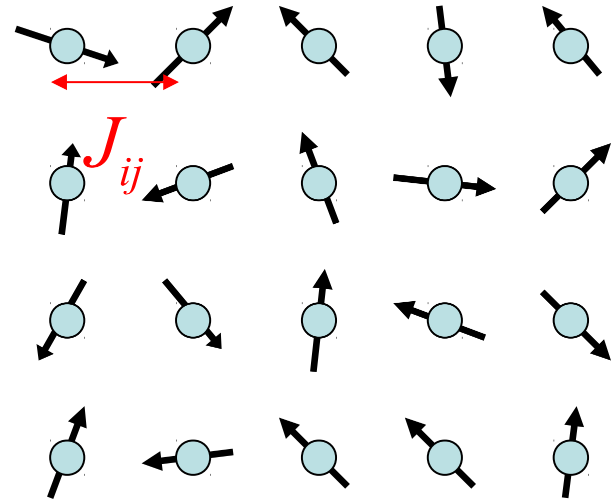
Examples of magnetic scattering experiments

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 posses a localized magnetic moment

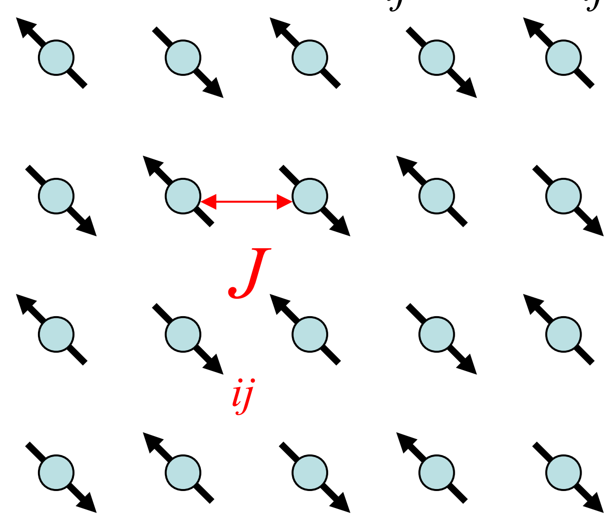


Ni^{2+}



$$\langle \mathbf{S}_i \rangle = 0$$

$$E_{ij} = -J_{ij}\, \mathbf{S}_i \cdot \mathbf{S}_j$$

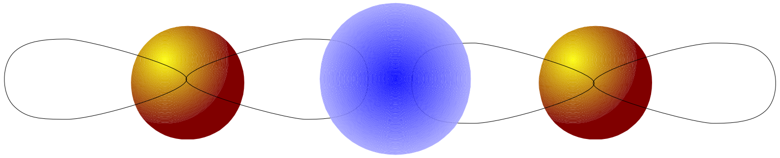


- When exchange interactions (direct, superexchange, double exchange, RKKY,dipolar ….) stabilizes a long range magnetic order.

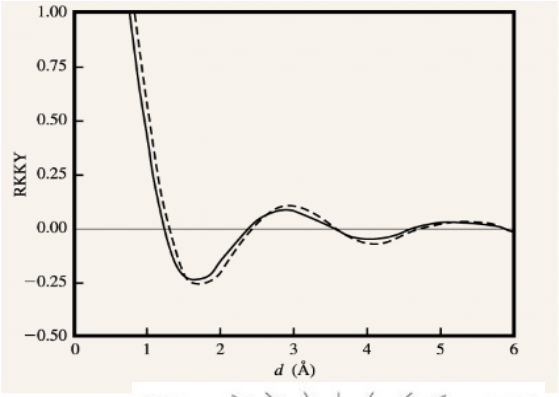
Different magnetic state(s)

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

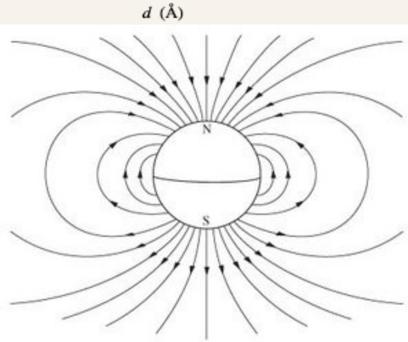
• Indirect exchange interaction
• Super-exchange (M-O-M)
• Super super-exchange (M-O-O-M)
coupling through a diamagnetic anion or more complex exchange paths



•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

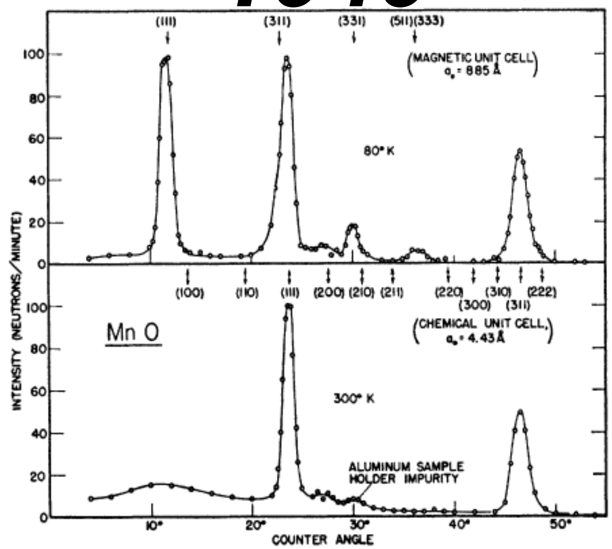


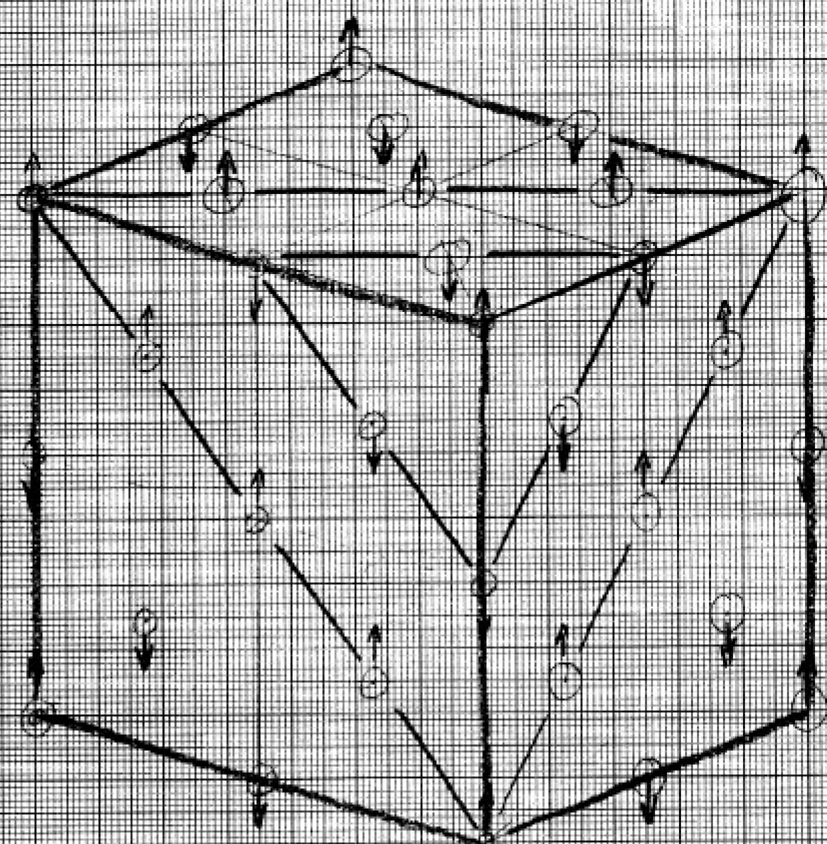
FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.

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

NEUTRONS FOR SCIENCE ®

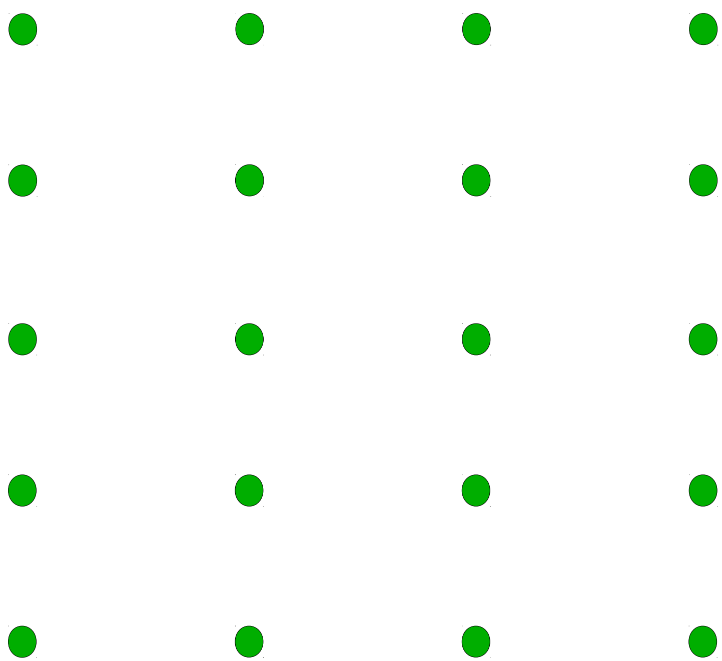
MnO



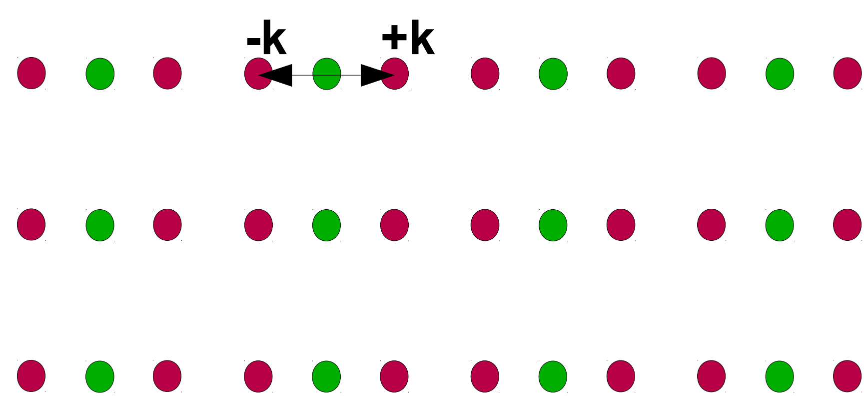
L. Neel — Ann. de Physique, 3 p.137 (1948)
C.R. Acad Sci Paris 228 64-6 (1949)

	MnO	MnS	FeO	FeF_2	MnF_2	αFe_2O_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_a °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:

$$\mathbf{m}_{lj}(\mathbf{R}_L) = \sum_{\mathbf{k}} \mathbf{S}_{\mathbf{k}j} . e^{-2\pi i \mathbf{k}.\mathbf{R}_L}$$

which for a single propagation vector :

$$\mathbf{m}_{lj}(\mathbf{R}_L) = \mathbf{S}_{\mathbf{k}j} . e^{-2\pi i \mathbf{k}.\mathbf{R}_L} + \mathbf{S}_{-\mathbf{k}j} . e^{2\pi i \mathbf{k}.\mathbf{R}_L}$$

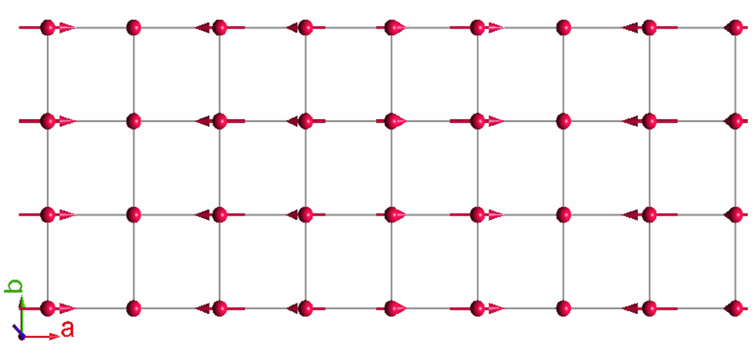
Since $\mathbf{m}_{lj}$ is a real vector,
one must imposes the condition $\mathbf{S}_{-kj}^{*} = \mathbf{S}_{kj}$

Here 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.

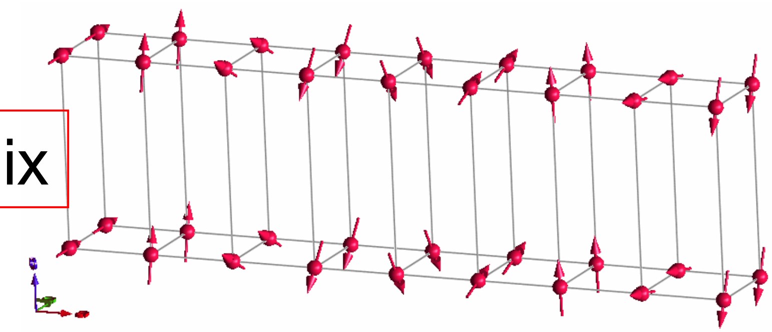
k inside BZ

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

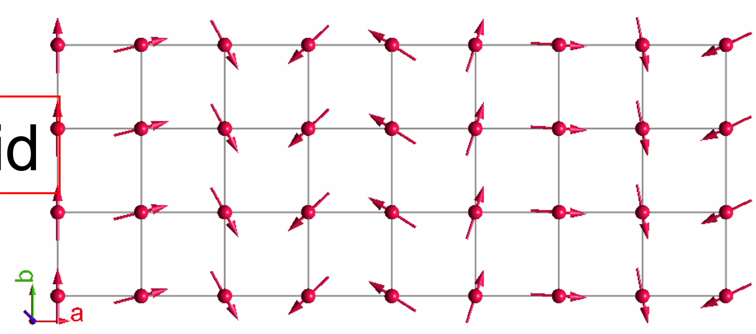
Amplitude modulation



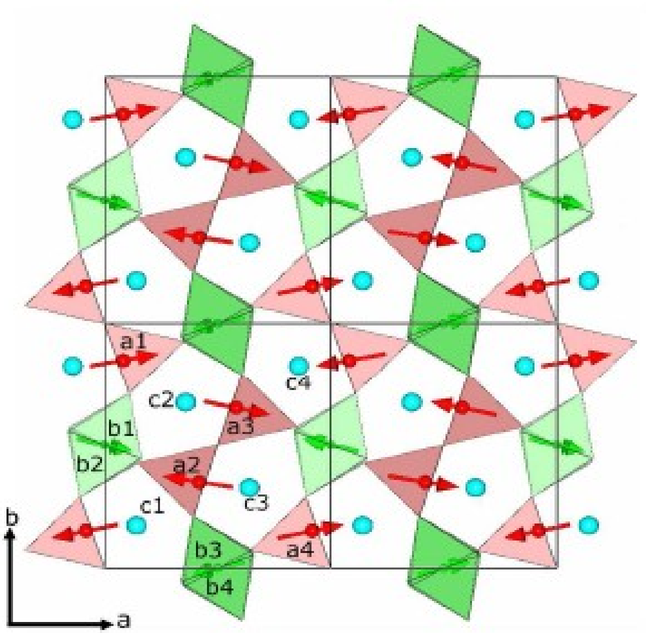
Helix

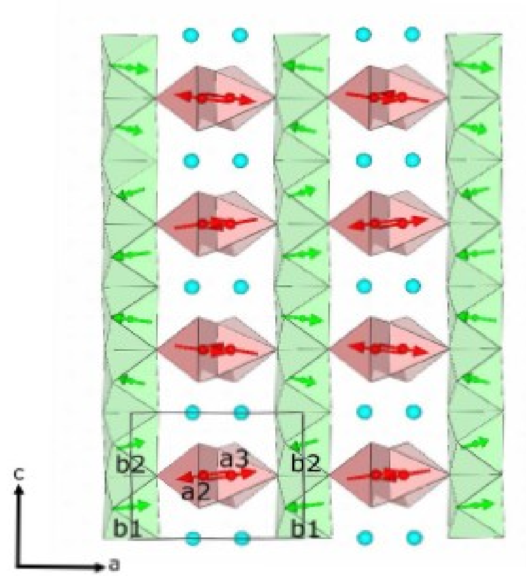


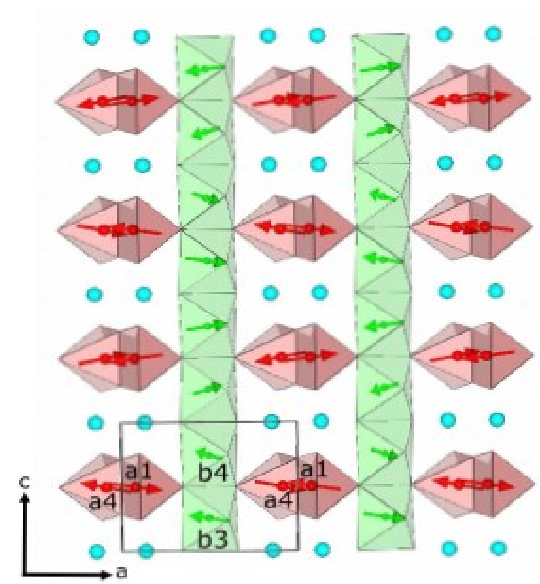
Cycloid



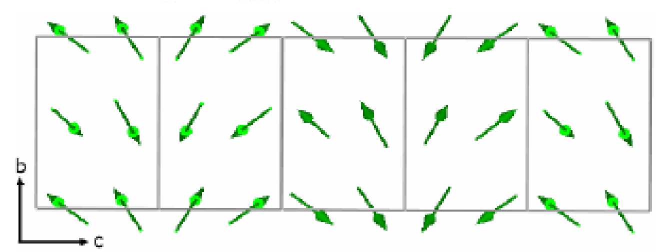
Quite complex ordered states (RMn_2O_5)







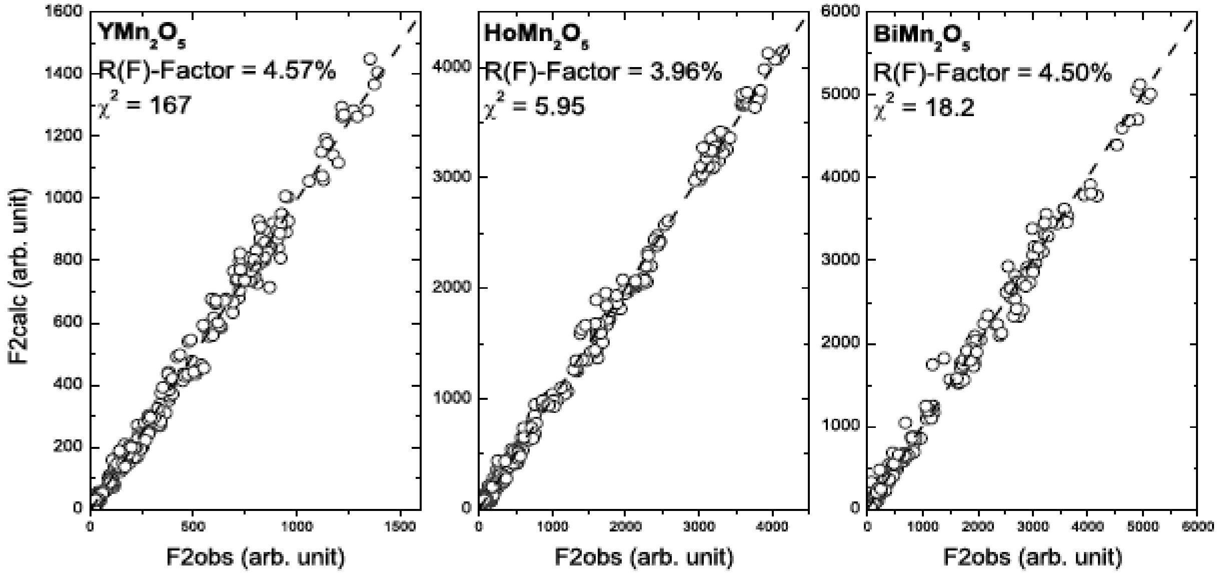
k=(1/2, 0, 1/4)



Y
Mn^{3+}
Mn^{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-Iz-MagPh(Mn^{3+} fixed)-ext4}

m(Mn^{4+})=2.4μ_B
m(Mn^{3+})=3.1 μ_B

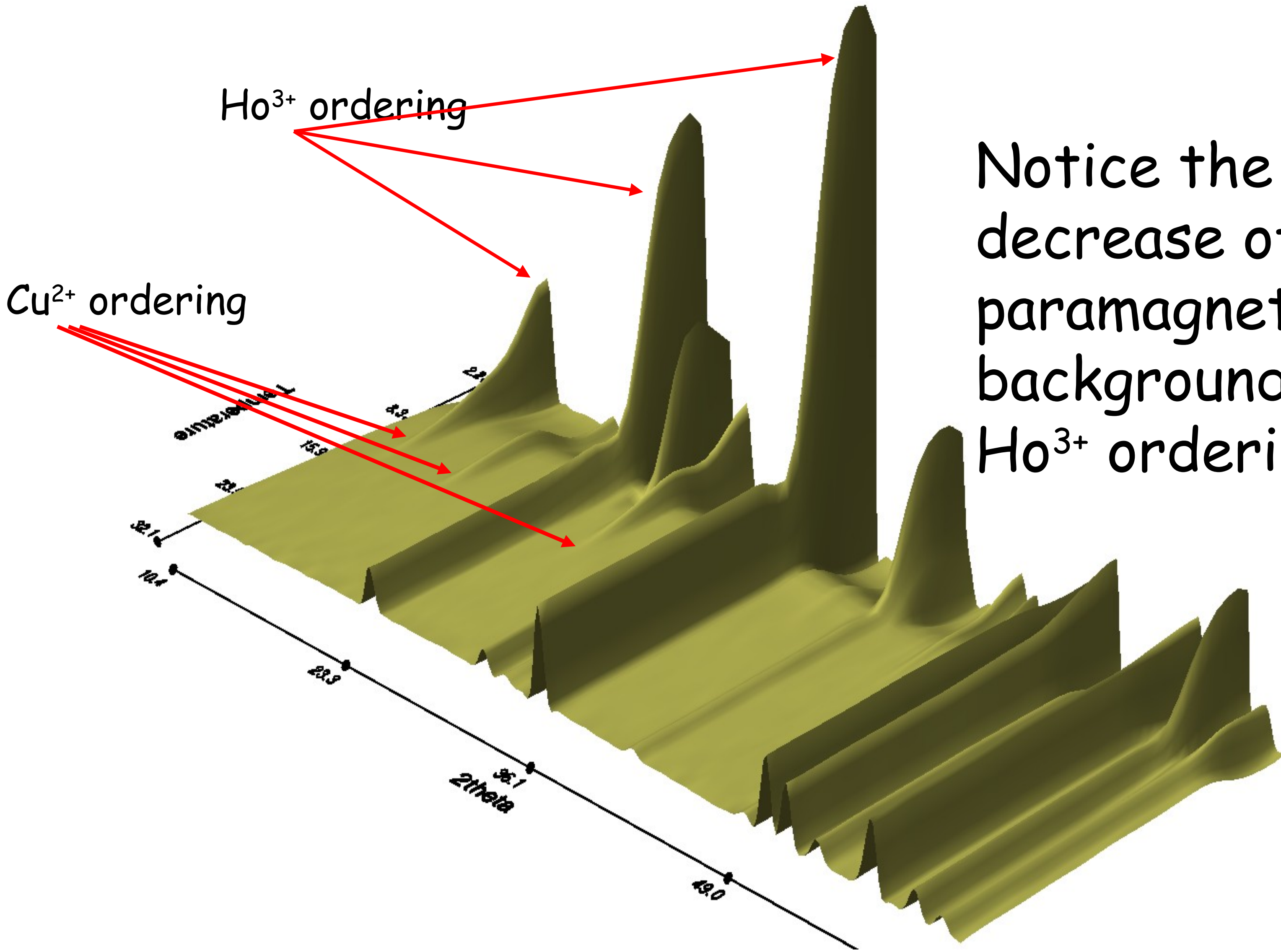
$HoMn_2O_5$
381 independent reflections
53 refined parameters:
{Rx-Ry-Iz-MagPh(Mn^{3+} fixed)-ext4}

m(Mn^{4+})=2 μ_B
m(Mn^{3+})=2.5 μ_B
m(Ho)=0/1 μ_B

$BiMn_2O_5$
204 independent reflections
24 refined parameters:
{Rx-Ry-Rz-ext4}

m(Mn^{4+})=2.1 μ_B
m(Mn^{3+})=2.8 μ_B

Magnetic ordering of Ho and Cu ions in Ho_2BaCuO_5 (D1B)



Notice the decrease of the paramagnetic background on Ho^{3+} ordering

Competing multi-q magnetic structures in $HoGe_3$ (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)

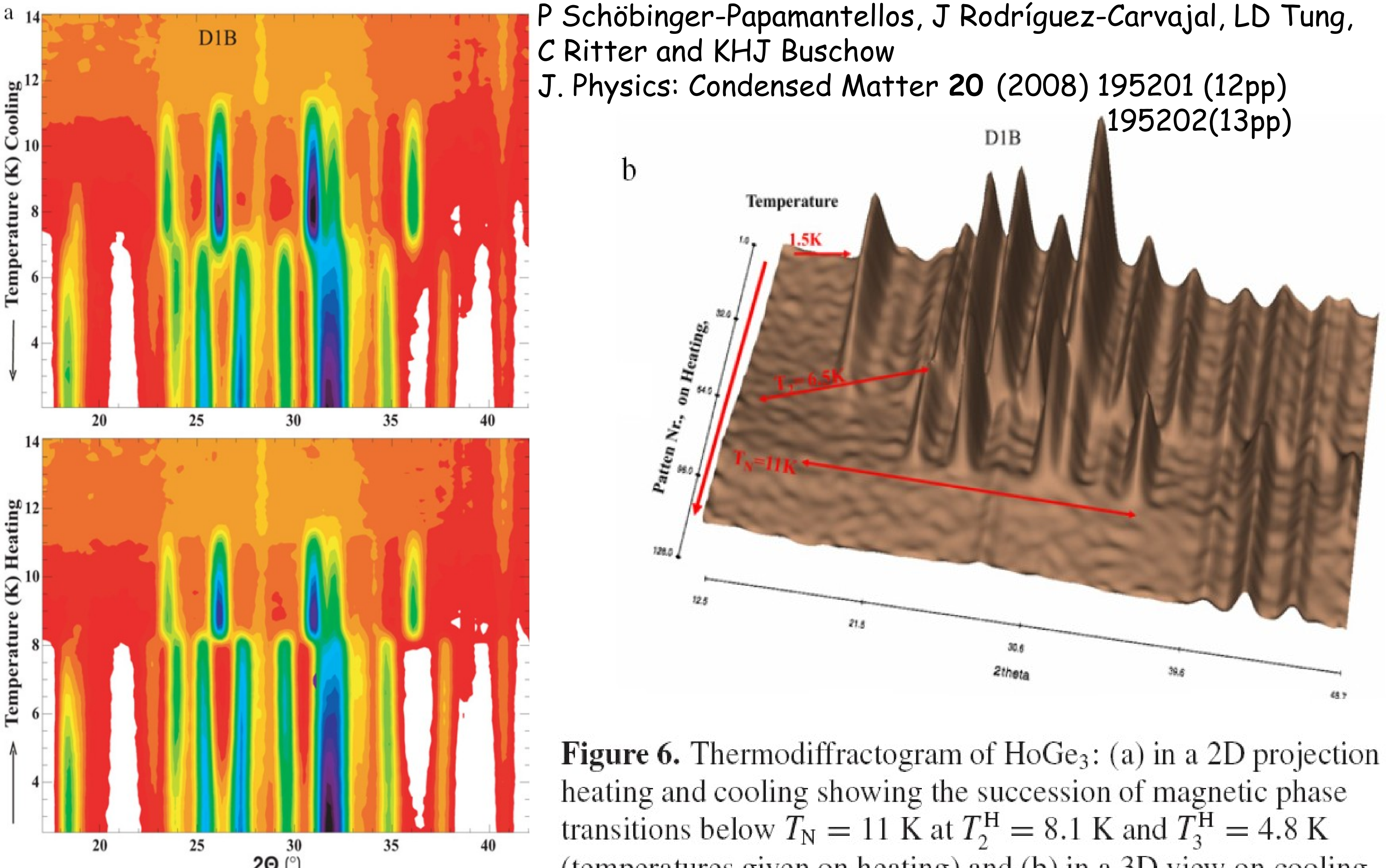
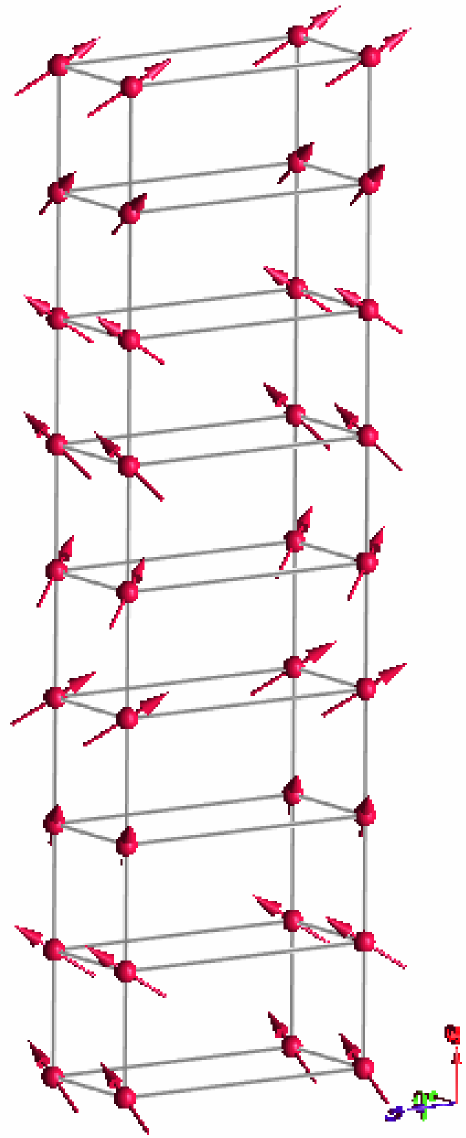


Figure 6. Thermodiffractogram of $HoGe_3$: (a) in a 2D projection on heating and cooling showing the succession of magnetic phase transitions below $T_N = 11$ K at $T_2^H = 8.1$ K and $T_3^H = 4.8$ K (temperatures given on heating) and (b) in a 3D view on cooling.

Multi-k structure: conical example

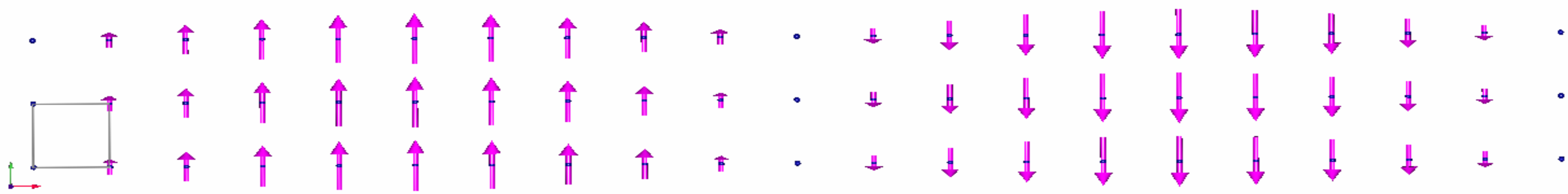
Conical



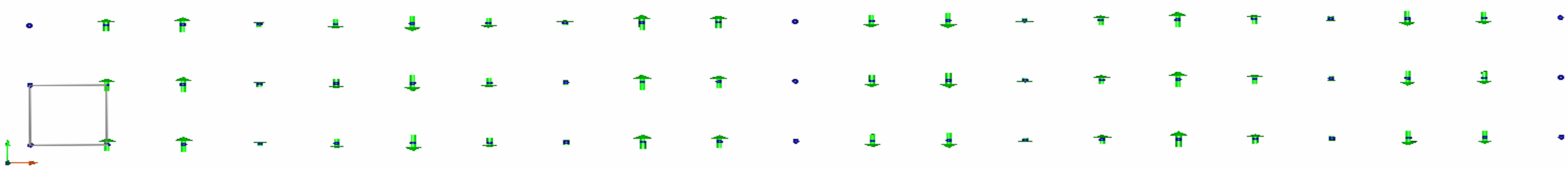
Multi-k structure with:

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

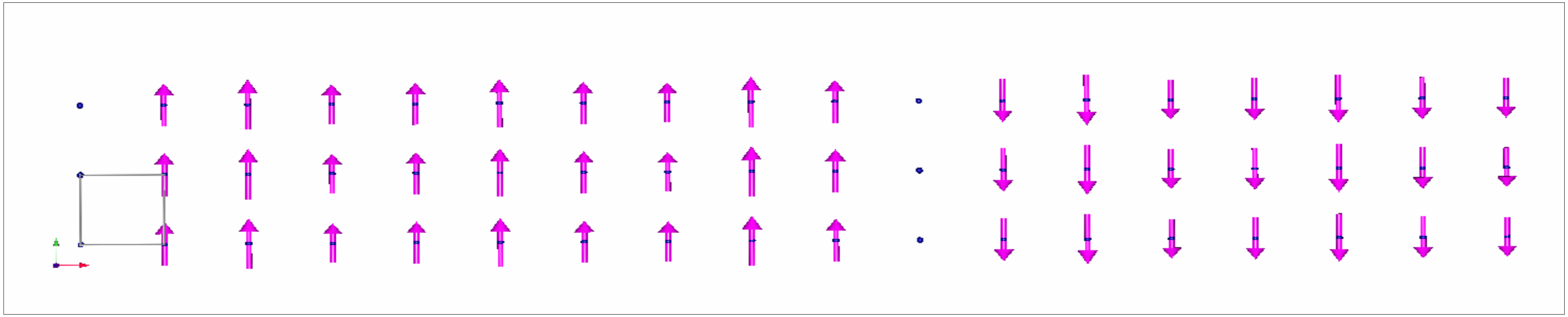
Multi-k structures : Bunched modulations



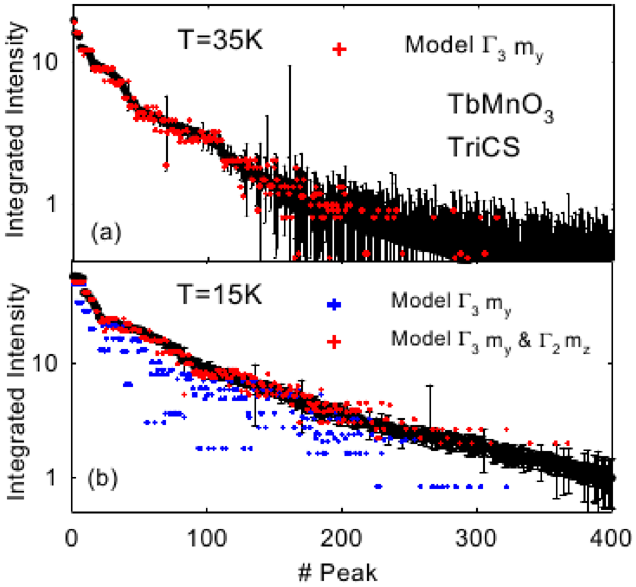
k=(d,0,0)

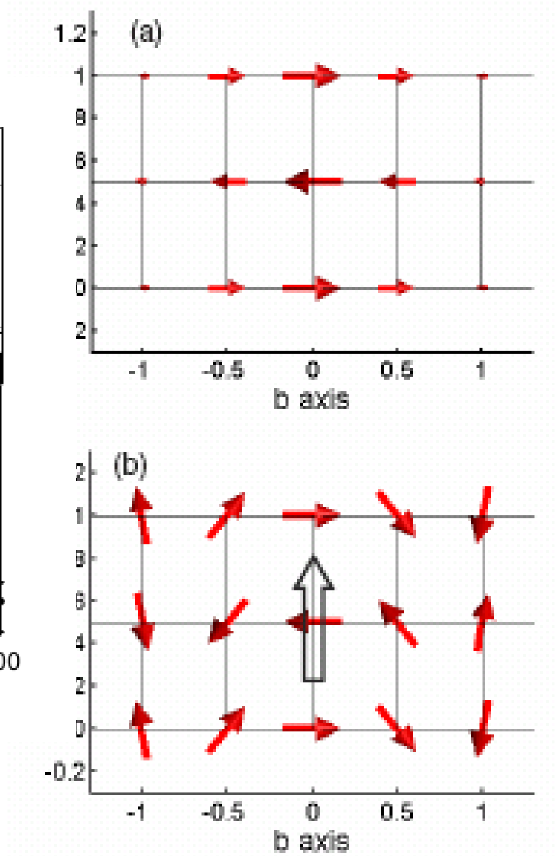


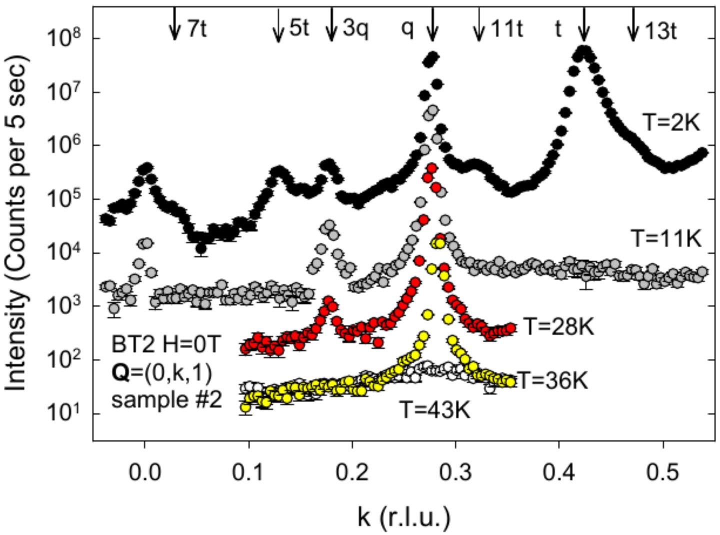
+ k=(3d,0,0) + … + k=((2n+1)d,0,0)



$TbMnO_3$



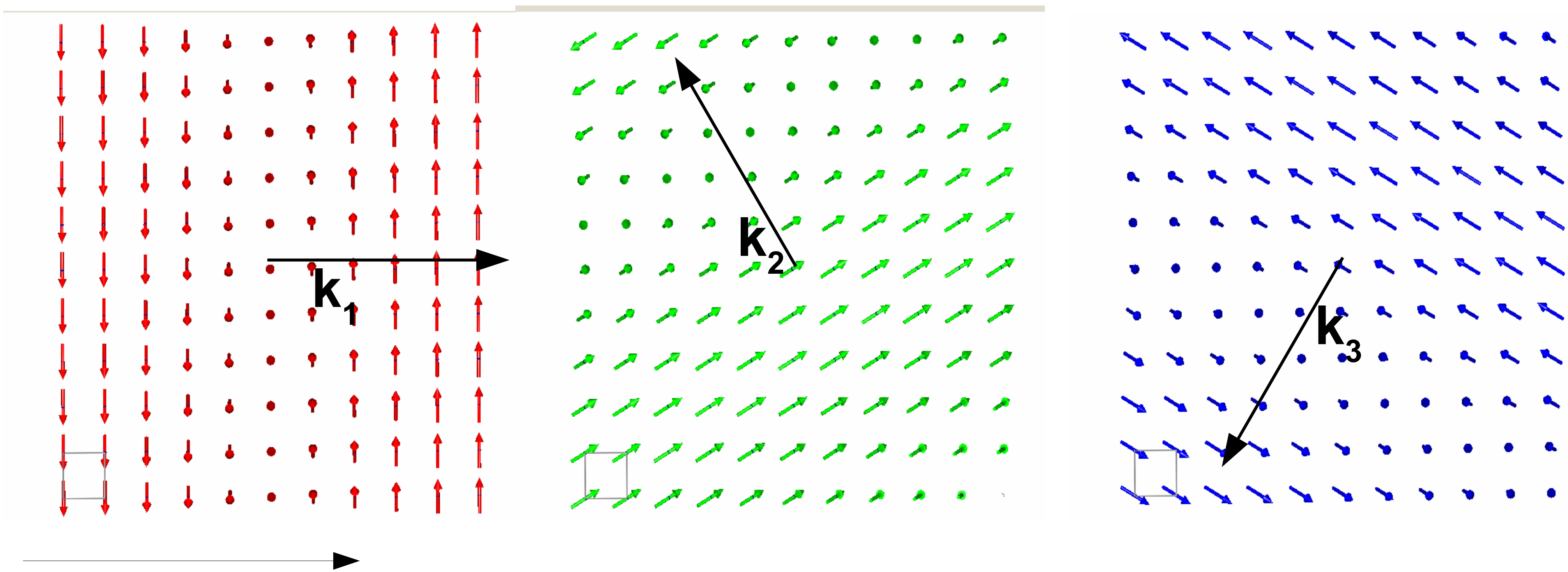




Kenzelmann, PRL 95, 087206 (2005)

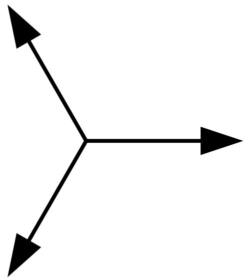
Multi-k structures

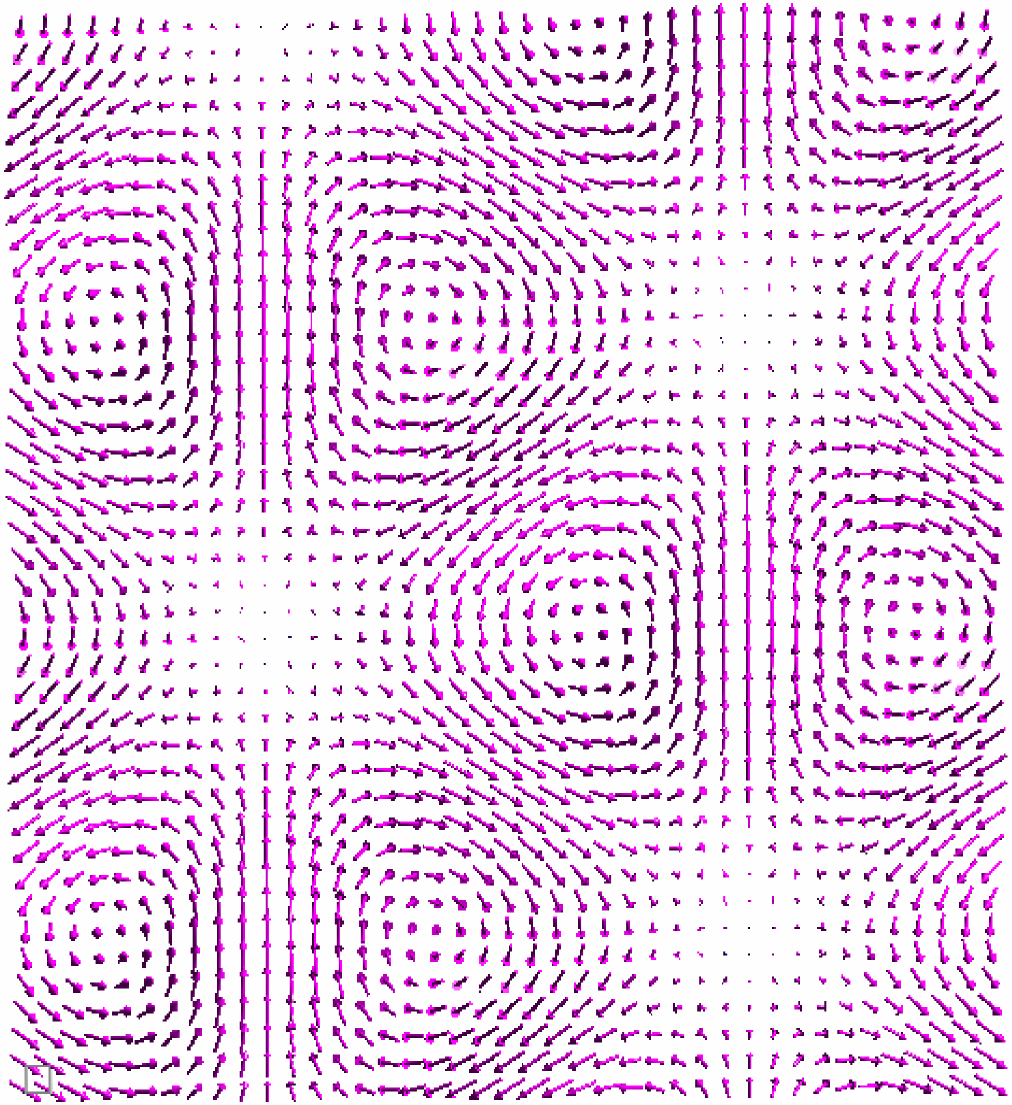
Example of a 4-k structure: the skyrmion lattice



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

Multi-k structures

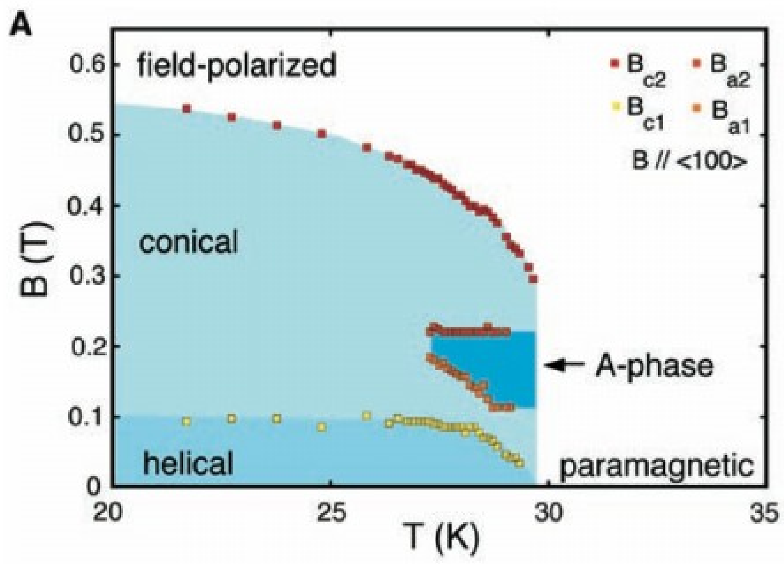




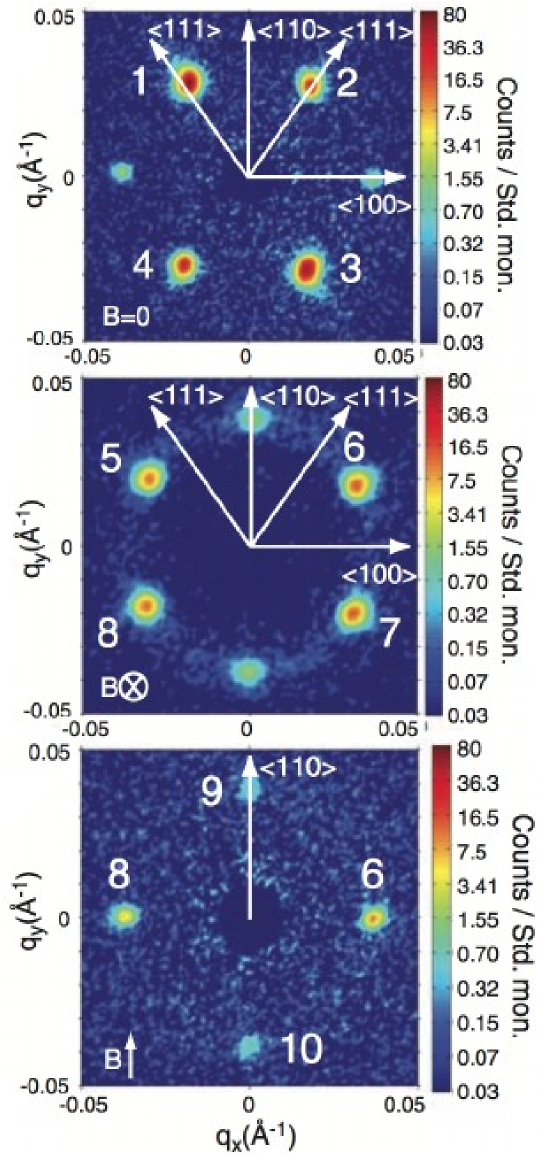
“Skyrmion”-type lattice stabilized by energy terms of the type:

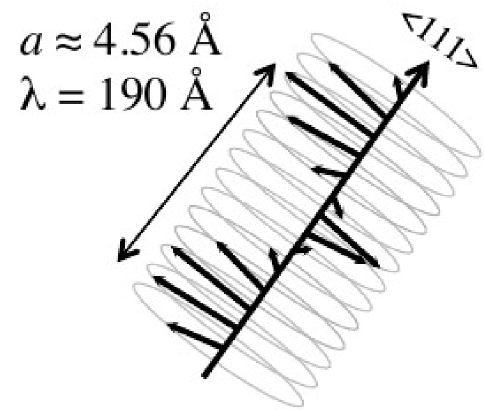
$$F = \ldots + S_1 e^{ik_1 + \varphi_1} . S_2 e^{ik_2 + \varphi_2} . S_3 e^{ik_3 + \varphi_3} . M$$

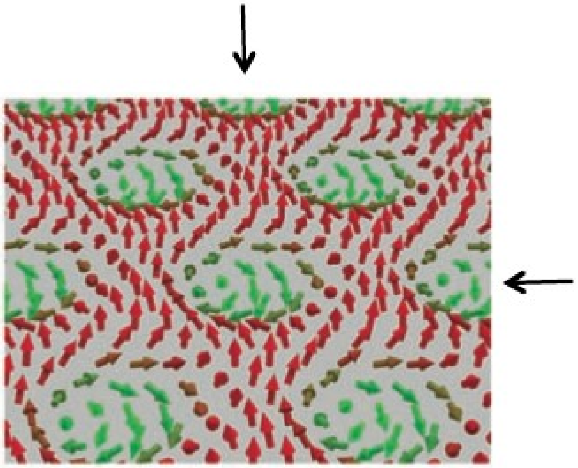
Skyrmion in MnSi



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







Domains

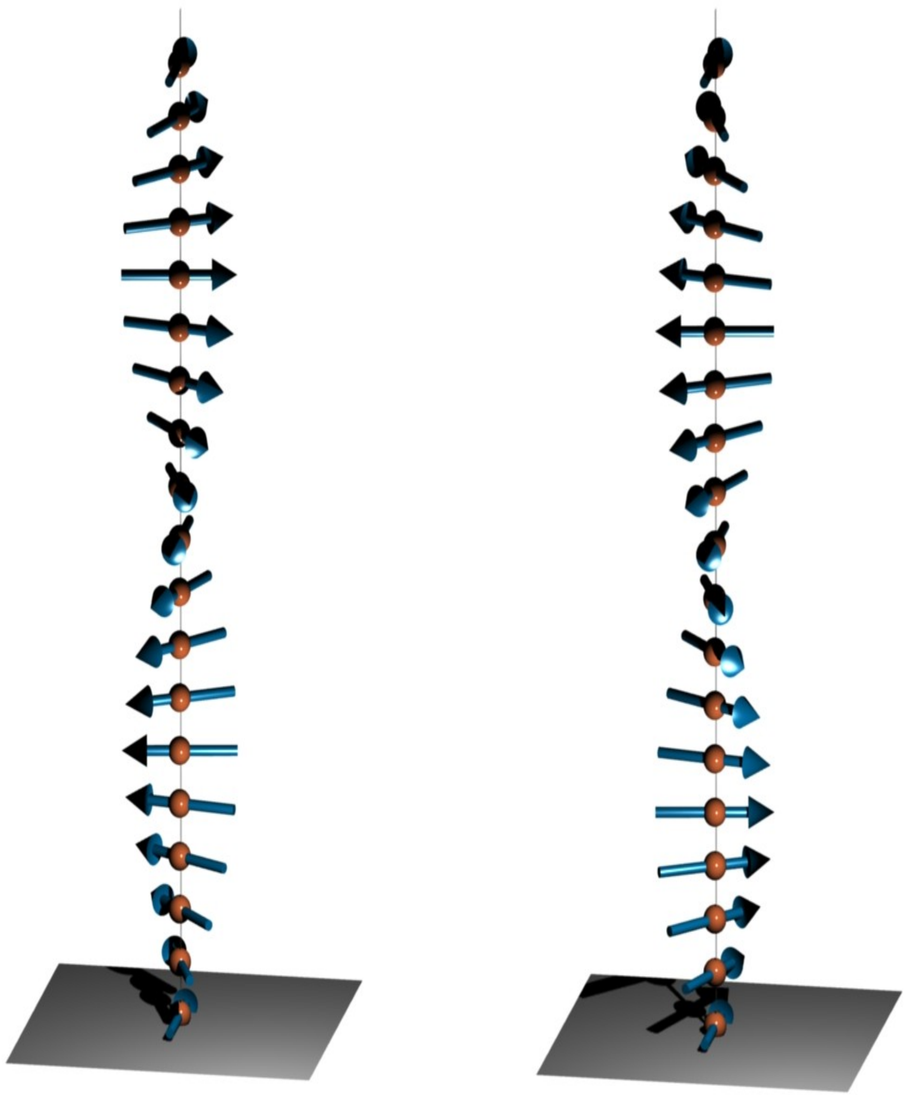
- Because the **symmetry** of the ordered magnetic state **is lower** than that of the paramagnetic state (loss of certain symmetry elements)

- If the order of the paramagnetic group $\mathbf{G_0}$ **is g** and the order of the ordered group $\mathbf{G_1}$ **is h**, there will be **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

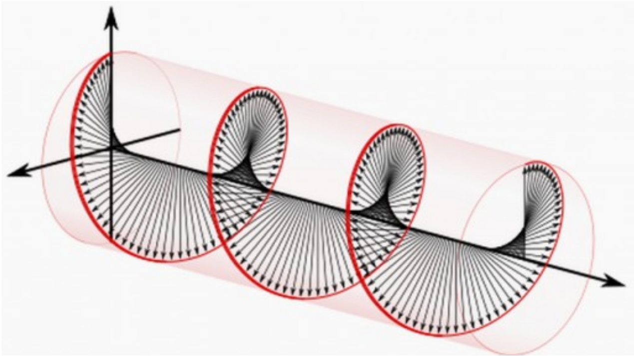
Chiral-domains

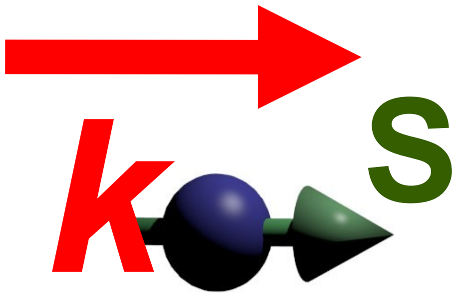


- Loss of inversion symmetry generates two domains of opposite handedness
- Note however that this is not the case if the paramagnetic group is a chiral group, in which case a single handedness is stabilized (no energy degeneracy)

Inversion domains ("chiral" scattering)

Spherical Polarimetry, ILL CRYOPAD

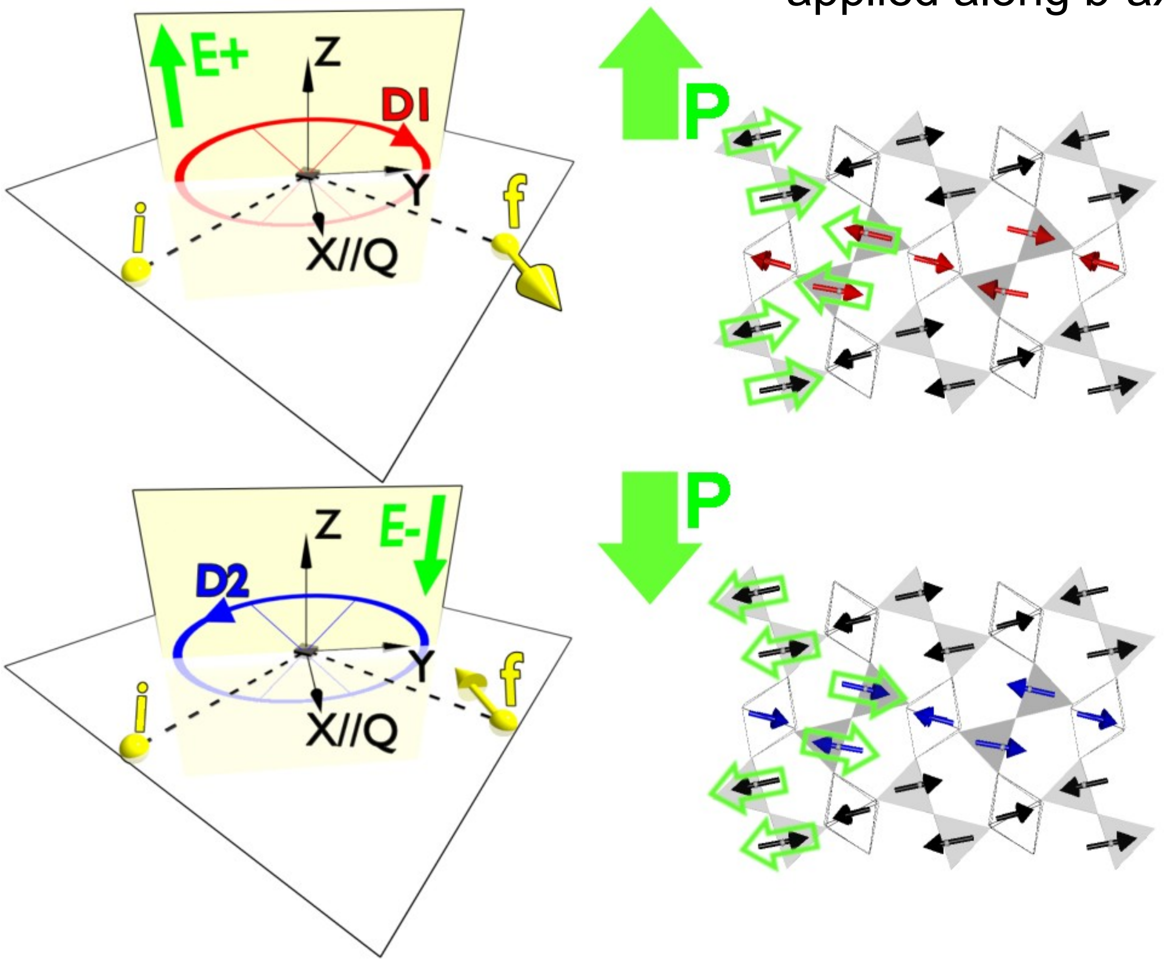




$$\Im\left(M_{\perp} \times M^{*}_{\perp}\right)$$

Inversion domains

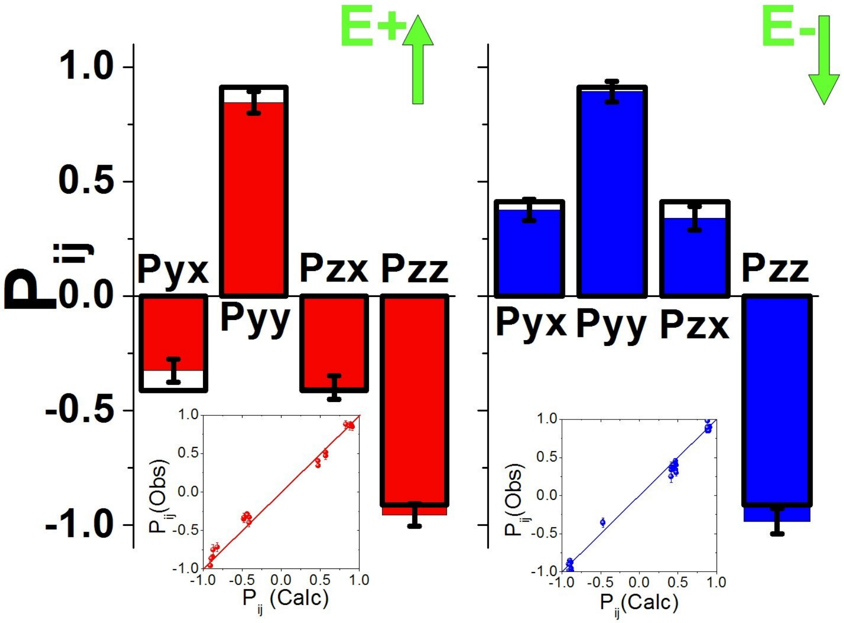
Crystal: b-axis along z-direction
Electric field up to 2.2 kV/cm applied along b-axis

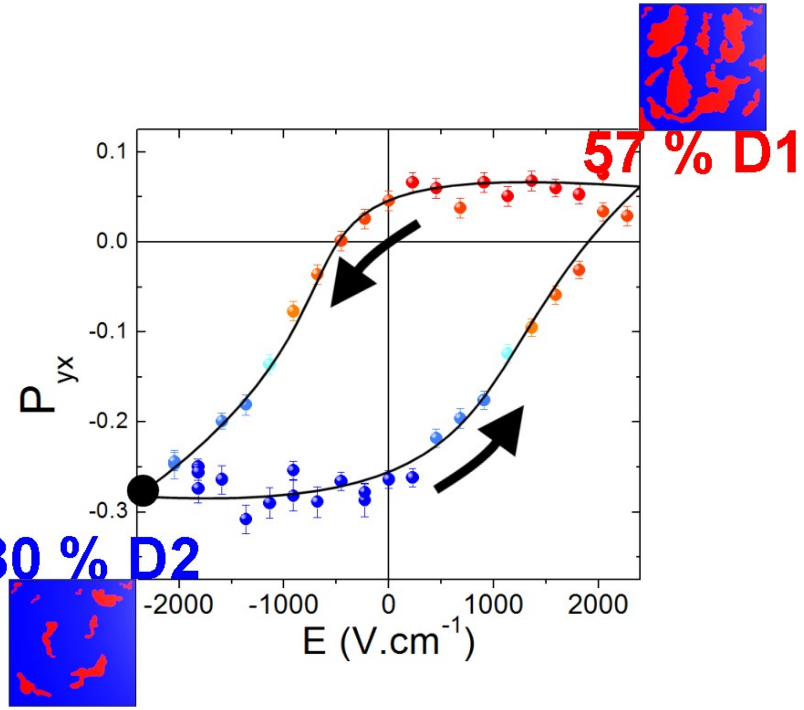


Inversion domains

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

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



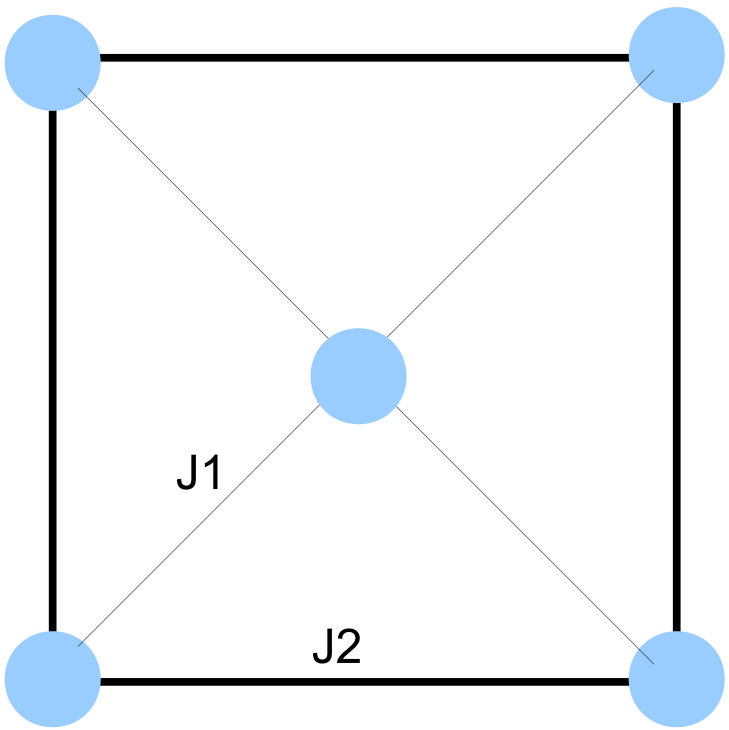


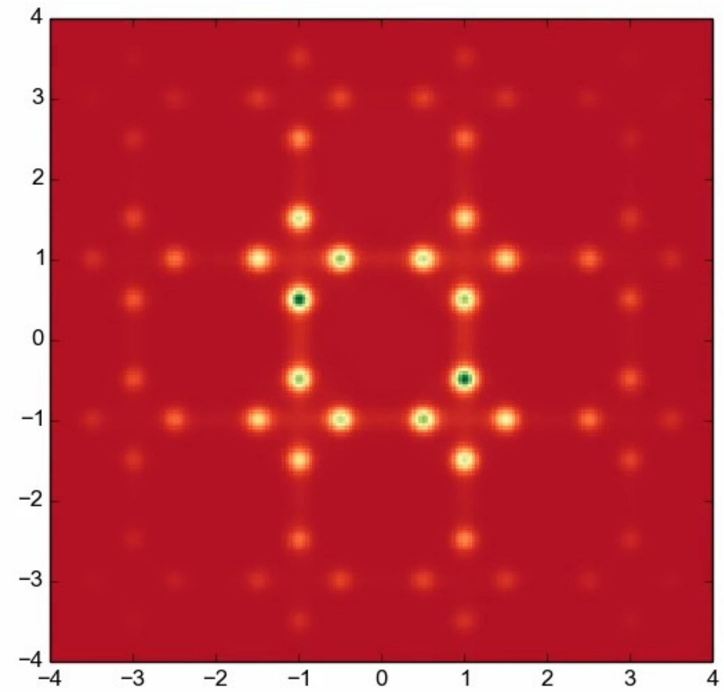
Radaelli et al., PRL 2008

Short-range correlations

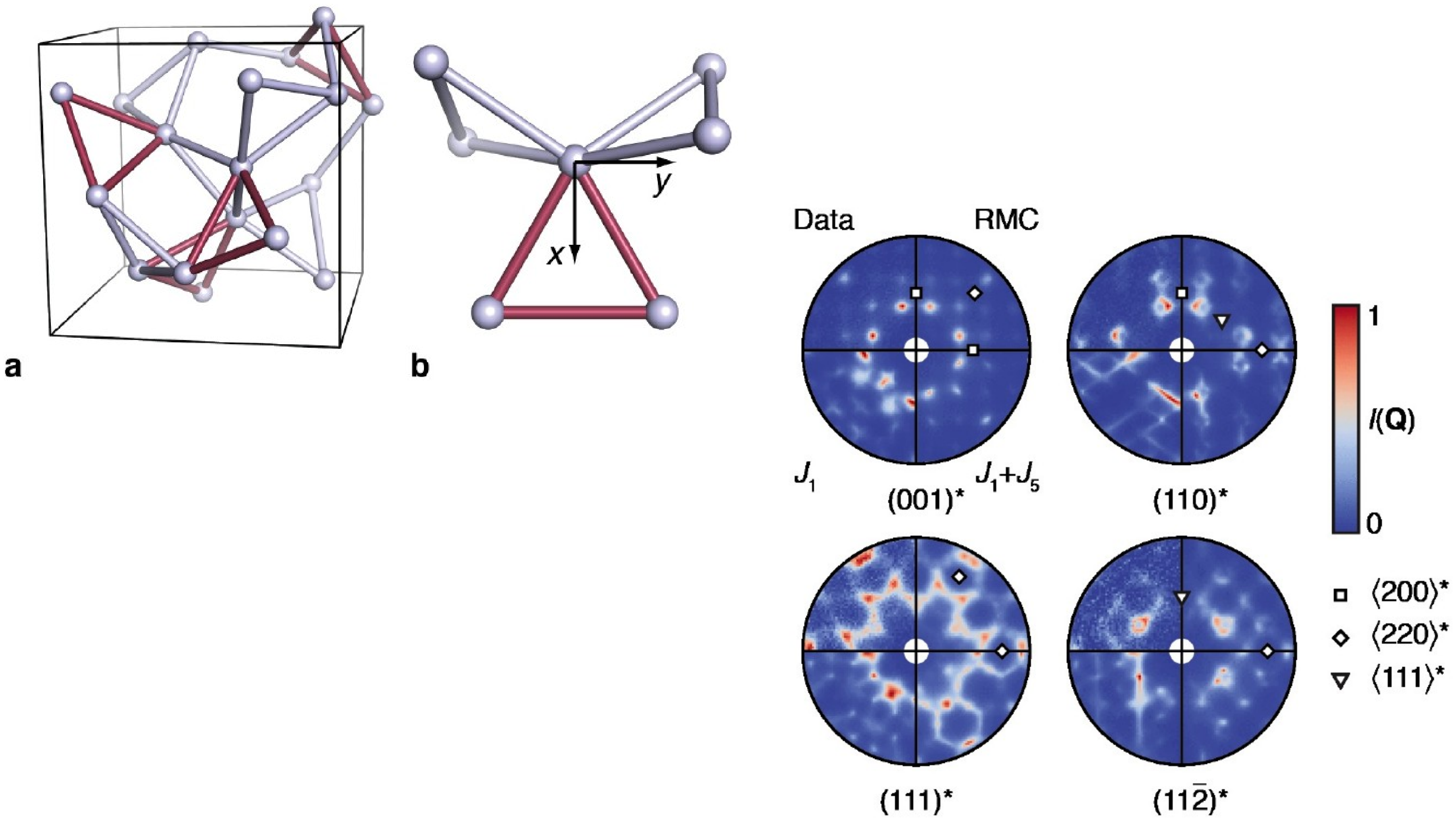
Probing short-range correlations
Via diffuse magnetic scattering

Simple J1-J2 cubic fcc magnet



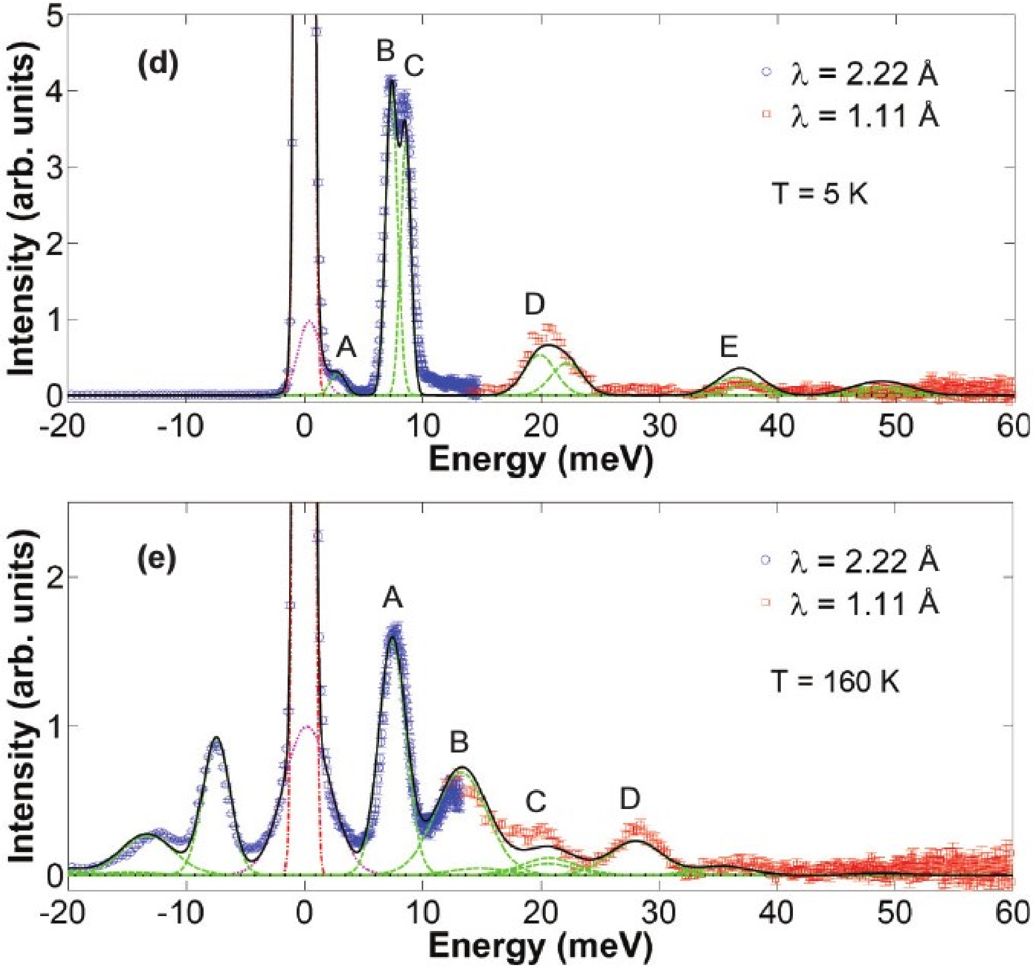


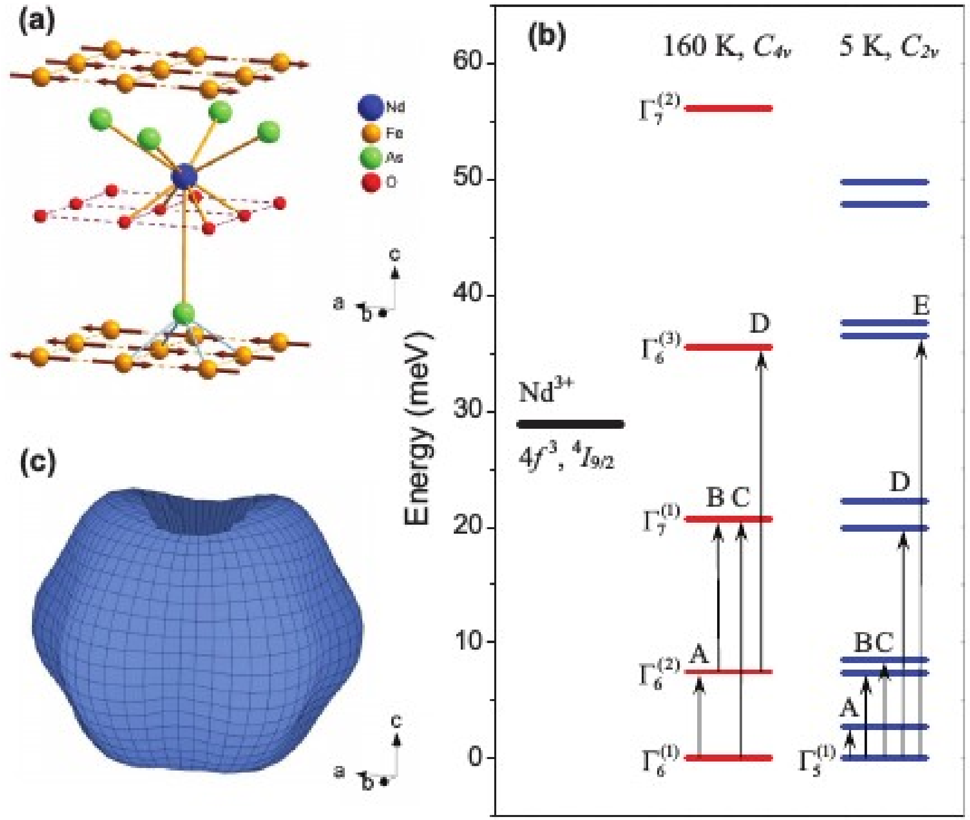
Short range correlations in frustrated beta-Mn



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)