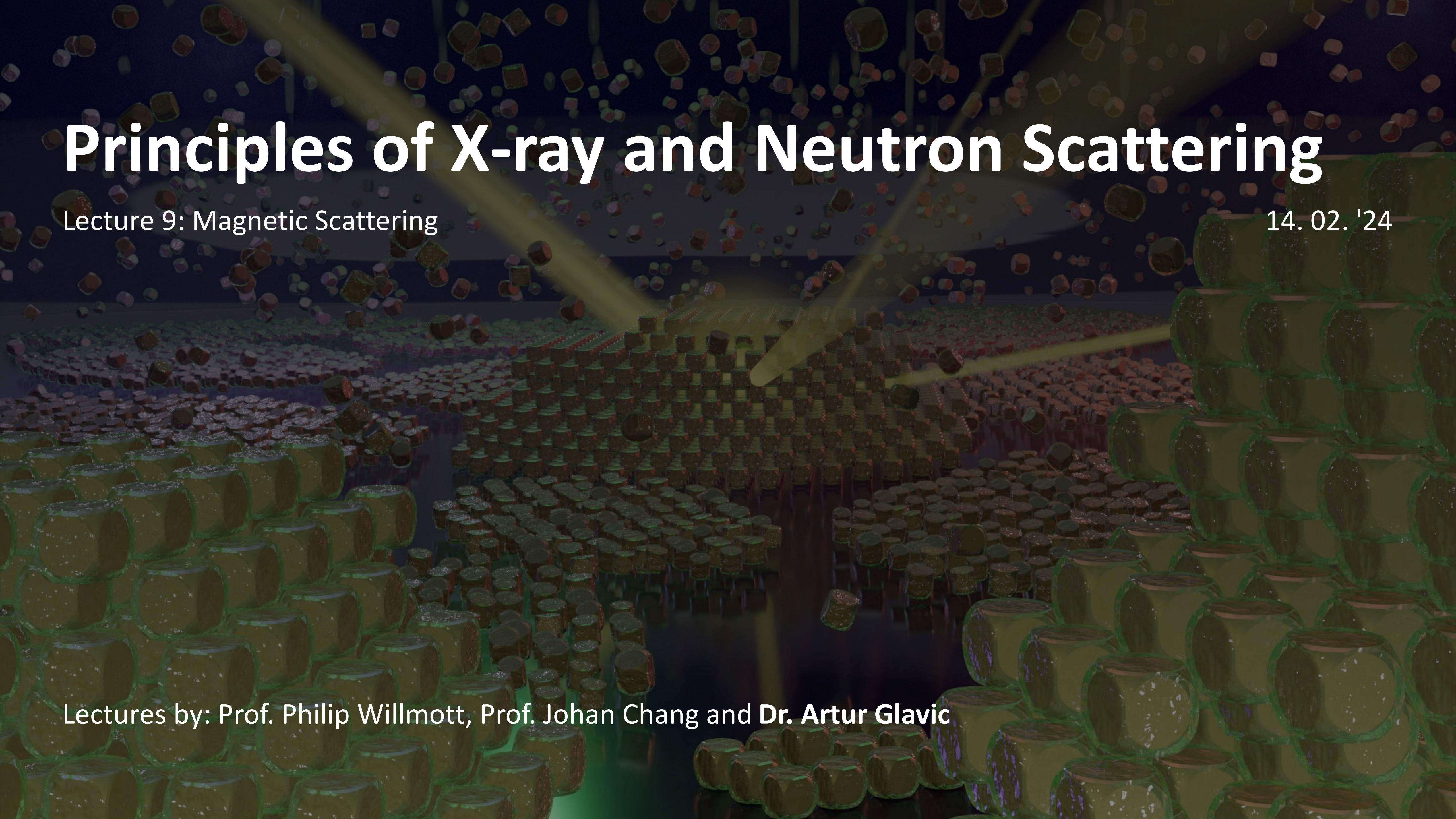


Principles of X-ray and Neutron Scattering

Lecture 9: Magnetic Scattering

14. 02. '24

Lectures by: Prof. Philip Willmott, Prof. Johan Chang and **Dr. Artur Glavic**

Course Outline

Monday	Tuesday	Wednesday	Thursday	Friday
Lecture 1 10-10h45 Philip	**Lecture 4** 10-10h45 Philip	**Lecture 7** 10-10h45 Artur	**Lecture 10** 10-10h45 Artur	**Lecture 13** 10-10h45 Johan
Lecture 2 11-11h45 Philip	**Lecture 5** 11-11h45 Philip	**Lecture 8** 11-11h45 Artur	**Lecture 11** 11-11h45 Artur	**Lecture 14** 11-11h45 Johan
Lunch - Mensa	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**
Lecture 3 13h00-13h45 Philip	**Lecture 6** 13h00-13h45 Philip	**Lecture 9** 13h00-13h45 Artur	**Lecture 12** 13h00-13h45 Artur	**Lecture 15** 13h00-13h45 Johan
		Exercise Class 14h30-16		Exercise Class 14h30-16

X-ray scattering

Neutron Scattering

Resonant x-ray scattering

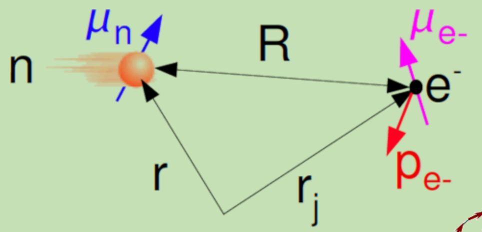
Neutron Lectures:

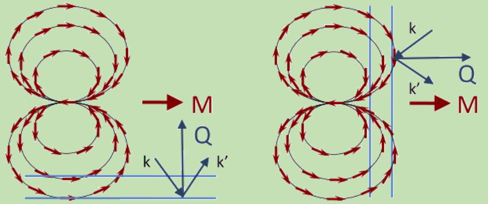
7: Neutrons & Scattering to Determine Structure
8: Inelastic Neutron Scattering to Investigate Dynamics
9: Magnetic Scattering
10: Neutron Polarization Analysis
11: Studying quantum matter for nanoscale applications
12: Neutron Instrument Development

Lecture 9: Magnetic Scattering

Theoretical Background

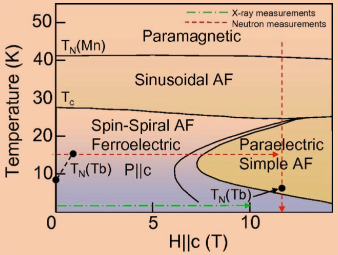
- Neutron magnetic interaction
- Magnetic scattering selection rules
- Magnetic form factor

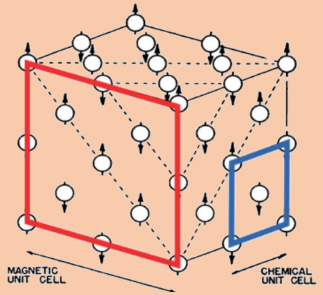


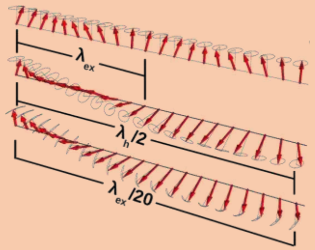


Example Application

- Experimental form factor
- Anti-ferromagnetic order
- Inelastic scattering from (heli-)magnons







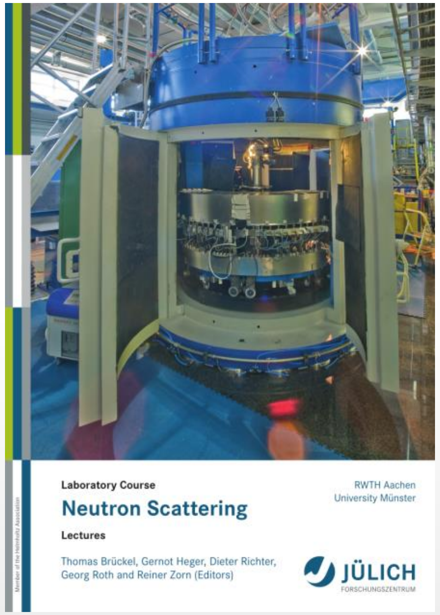
Further Reading

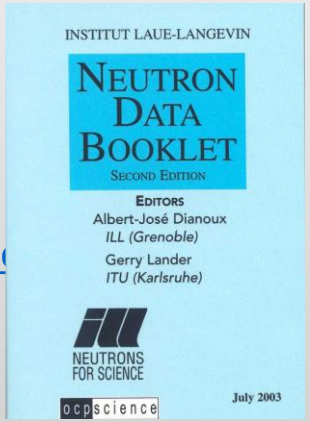
- **“Neutron Diffraction of Magnetic Materials”**
  Y. A. Izyumov, V. E. Naish, and R. P. Ozerov.
  Plenum Publishing Corporation, New York (1991)

- “Introduction to the Theory of Thermal Neutron Scattering”
  G. L. Squires
  Dover Publication (1978)
- “Theory of Neutron Scattering from Condensed Matter” Vol.I/II.
  S. W. Lovesey
  Oxford Science Publications (1984).

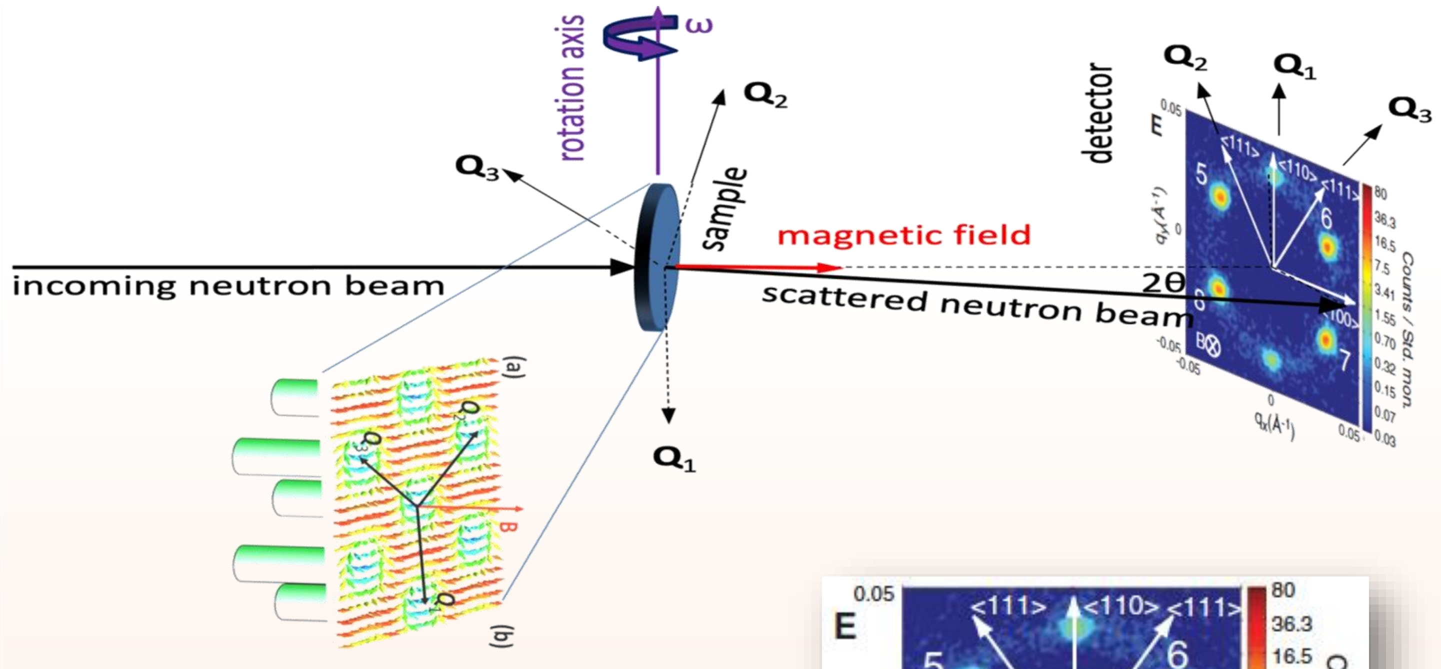
- “Neutron Scattering”
  T. Brückel, et al. (2012) / Available Open Access:
  https://juser.fz-juelich.de/record/136390/files/Schluesseltech_39.pdf

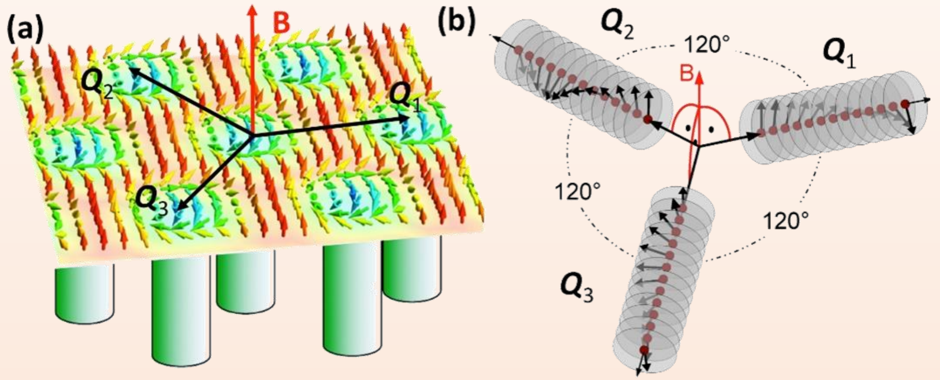
- “Neutron Data Book”
  Albert-José Dianoux and Gerry Lander
  https://www.ill.eu/fileadmin/user_upload/ILL/1_About_ILL/Documentation/NeutronDataBooklet.p

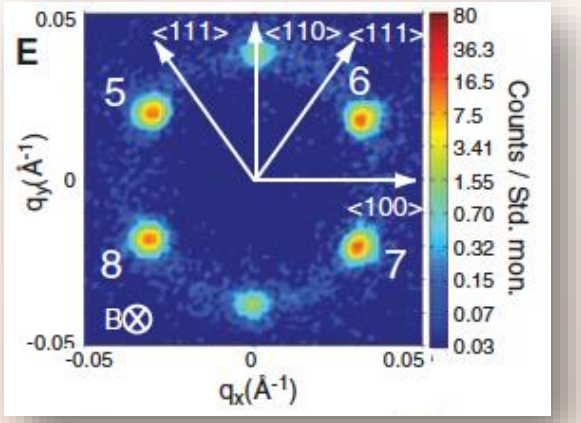




Example for Magnetic Scattering



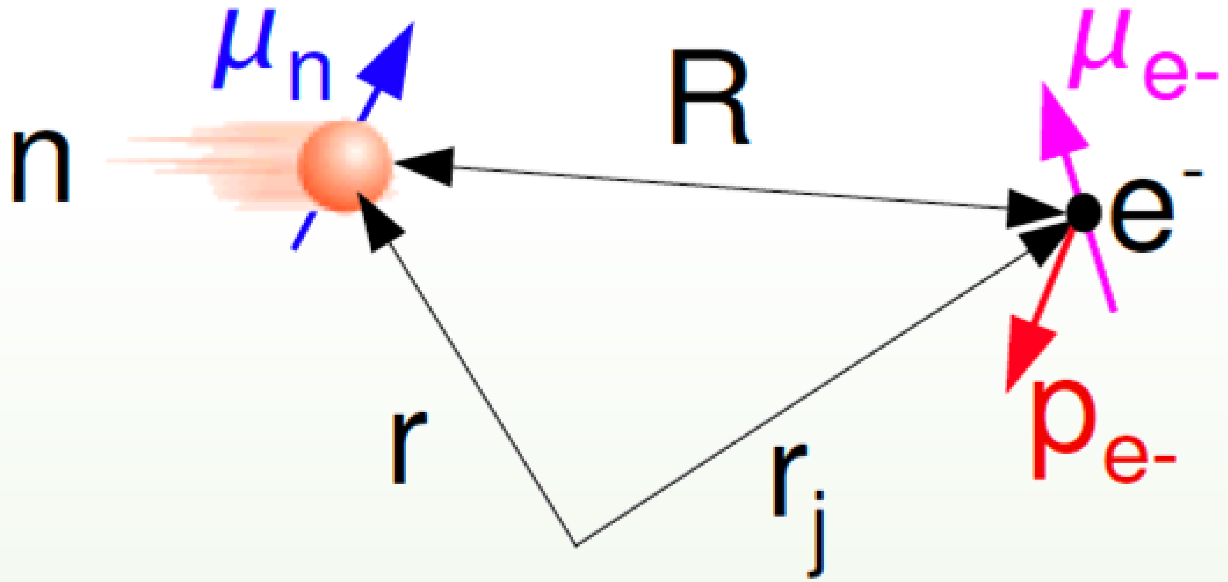




S. Mühlbauer et al. Science (2009).

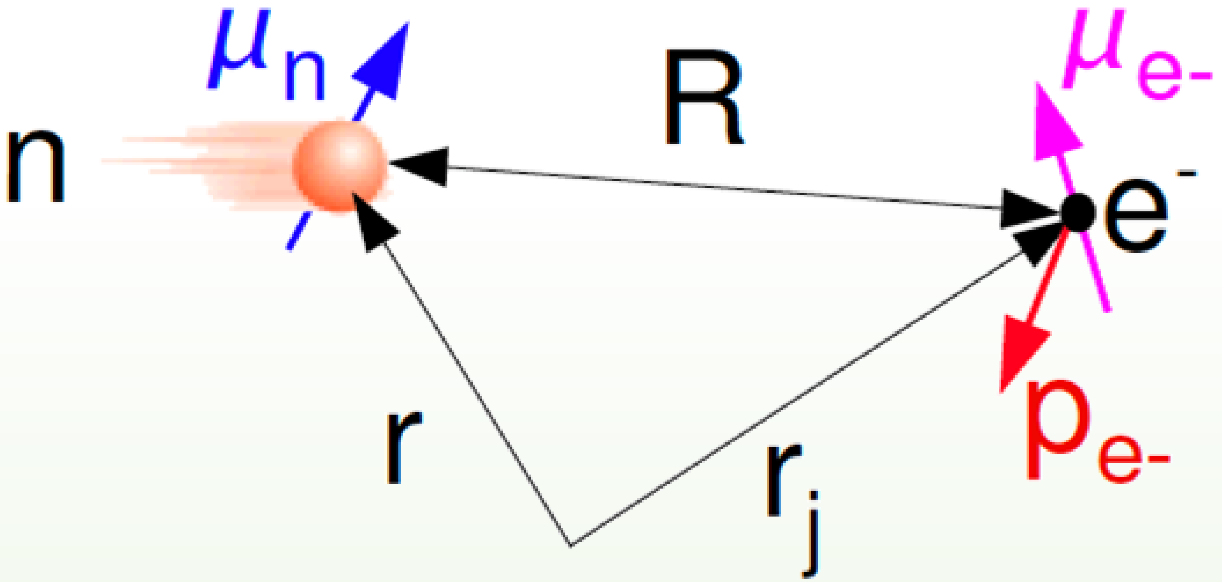
➠ **SANS discovered the Skyrmion lattice in MnSi for the first time!**

Magnetic Interaction



- Neutrons carry a spin ½ that interacts with any magnetic field.
- In solid states magnetic fields are generated by the electrons that also carry spin ½.
- Electrons additionally carry an orbital momentum L also creating a magnetic field.

Magnetic Interaction



$$V_m = -\vec{\mu}_n \vec{B} \text{ with } \vec{\mu}_n = -\gamma \mu_N \vec{\sigma}$$

$\gamma = 1.913$ is the neutrons gyromagnetic ratio

$\mu_N = \frac{e\hbar}{2\, m_p}$ is the nuclear magneton (e is the elementary charge and m_p is its mass)

$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ are the Pauli spin matrix operators.

Magnetic Field Due To Electron

- Magnetic dipole moment of the electron $\mu_e = -2\mu_B \vec{s}$ produces a magnetic field at a distance $\vec{R}$

$$B_s = \vec{\nabla} \times \vec{A}, \text{ with } A = \frac{\mu_0}{4\pi} \frac{(\vec{\mu}_e \times \vec{R})}{R^3},$$

- Because the electron represents a moving charge e^- it additionally generates the field

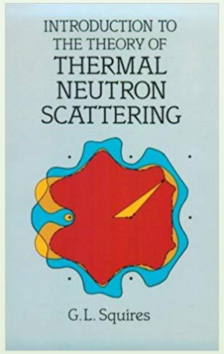
$$B_L = \frac{\mu_0}{4\pi} \frac{(2\mu_B)}{\hbar} \frac{(\vec{p} \times \vec{R})}{R^3},$$

at the point $\vec{R}$. Here $\vec{L} = \vec{R} \times \vec{p}$ is the angular momentum of the electron.

- In total we obtain the magnetic interaction potential between neutron and electrons (see Squires)

$$V_\mathrm{m} = -\frac{\mu_0}{4\pi} \gamma \mu_N 2\mu_B \vec{\sigma} (\vec{W_s} + \vec{W_L})$$

where $\vec{W_s} = \vec{\nabla} \times (\frac{\vec{s} \times \vec{R}}{R^3})$ and $\vec{W_L} = \frac{1}{\hbar} \left(\frac{\vec{p} \times \vec{R}}{R^3} \right)$.



Reminder: Cross-Section via Fermi's Golden Rule

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_I P(I) \sum_F |\langle k', F|V(x)|k, I\rangle|^2 \delta(E_I - E_F + \hbar\omega)$$

➔ Because V_m explicitly contains the neutron spin operator $\vec{\sigma}$ we have to introduce the spin state σ when evaluating the double-differential cross-section.

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{I,\sigma} P(I)P(\sigma) \sum_{F,\sigma'} |\langle k', F, \sigma'|V(x)|k, I, \sigma\rangle|^2 \delta(E_I - E_F + \hbar\omega)$$

➔ Here it is possible to separate the computation of the transition matrix into two parts:

$$\langle k', F, \sigma'|V_m|k, I, \sigma\rangle = \left\langle F, \sigma' \middle| \underbrace{\left\langle k' \middle| (\vec{W}_s + \vec{W}_L) \middle| k \right\rangle}_{\text{Does not depend on neutron spin!}} \middle| I, \sigma \right\rangle$$

The Magnetic Interaction Vector

➔ Evaluating the neutron spin-independent part for electrons i with position $\vec{r}_i$, spin s_i, and momentum $\vec{p}_i$, we obtain:

$$\left\langle k' \right| (\vec{W}_{si} + \vec{W}_{Li}) \left| k \right\rangle = 4\pi \vec{M}_{\perp\vec{Q}},$$

$\vec{M}_{\perp\vec{Q}} = \sum_i e\text{^}(i\vec{Q}\vec{r}_i)\left\{\hat{\vec{Q}} \times \left(\vec{s}_i \times \hat{\vec{Q}}\right) + \frac{i}{\hbar Q}(\vec{p}_i \times \hat{\vec{Q}})\right\}$ is called the ***magnetic interaction vector*** that only contains the position dependent part of V_m. $\hat{\vec{Q}}$ is a unit vector in the direction of $\vec{Q}$.

➔ It can be shown (involving a lengthy calculation) that $M_{\perp\vec{Q}}$ can be expressed as a function of the ***magnetization density*** $\vec{M}(\vec{r})$ ***of the scattering system***:

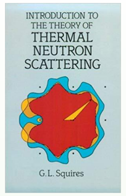
$$\vec{M}_{\perp\vec{Q}} = \hat{\vec{Q}} \times \left(\vec{M}_{\vec{Q}} \times \hat{\vec{Q}}\right) \qquad \vec{M}_{\vec{Q}} = -\frac{1}{2\mu_B}\int d^3r \vec{M}(\vec{r}) e^{i\vec{Q}\vec{r}}$$

➔ $\vec{M}_{\vec{Q}}$ is called the ***magnetic structure factor*** and is the Fourier transform of $\vec{M}(\vec{r})$.

The Magnetic Cross-Section

➔ Now we can evaluate the entire interaction matrix:

$$\sum_{\sigma,\sigma'} P(\sigma)\ |\langle k'\lambda'\sigma'|V_m|k\lambda\sigma\rangle|^2 = \sum_{\sigma,\sigma'} P(\sigma) \left|\left\langle \lambda'\sigma' \middle| \vec{\sigma}\cdot\vec{M}_{\perp\vec{Q}} \middle| \lambda\sigma \right\rangle\right|^2$$



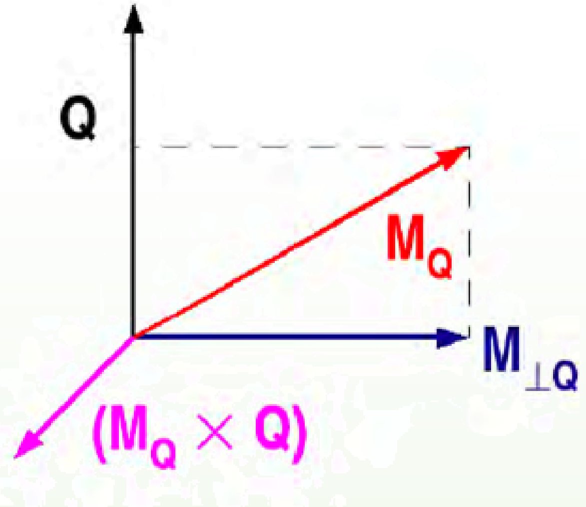
$$= \sum_\alpha \left\langle \lambda \middle| \vec{M}^{\alpha\dagger}_{\perp\vec{Q}} \middle| \lambda' \right\rangle \left\langle \lambda' \middle| \vec{M}^{\alpha}_{\perp\vec{Q}} \middle| \lambda \right\rangle$$

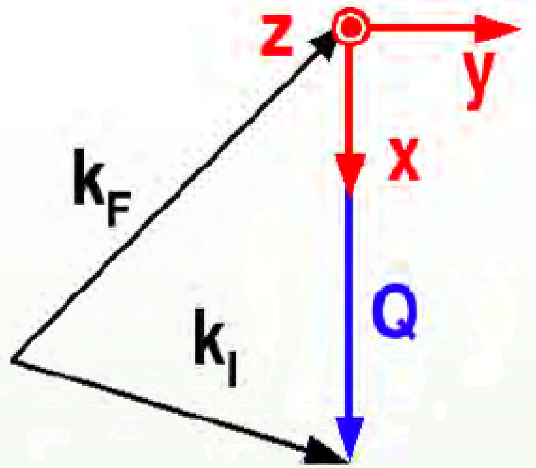
➔ With this we can write down the full magnetic cross-section:

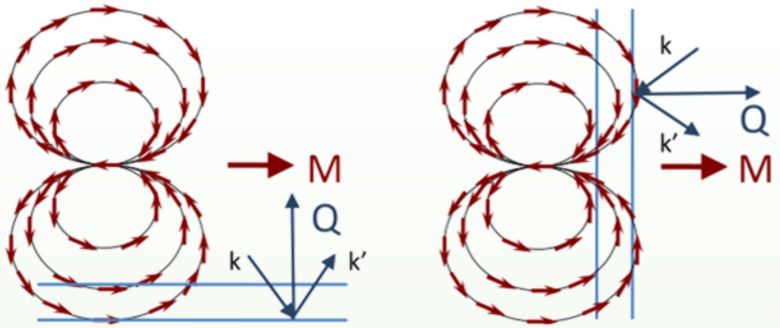
$$\frac{d^2\sigma}{d\Omega dE'} = (\gamma r_0)^2 \frac{k'}{k} \sum_{I,F} P(I) \sum_\alpha \left\langle I \middle| \vec{M}^{\alpha\dagger}_{\perp\vec{Q}} \middle| F \right\rangle \left\langle F \middle| \vec{M}^{\alpha}_{\perp\vec{Q}} \middle| I \right\rangle \delta(E_I - E_F + \hbar\omega)$$

Here $r_0 = 2.82 \cdot 10^{-15}$ m is a collection of prefactors and corresponds to the classical electron radius.

The Magnetic Selection Rule







➔ Note that due to the double cross-product in the magnetic interaction vector only magnetic moments in the sample that are perpendicular to the momentum transfer $\vec{Q}$ will contribute to scattering.

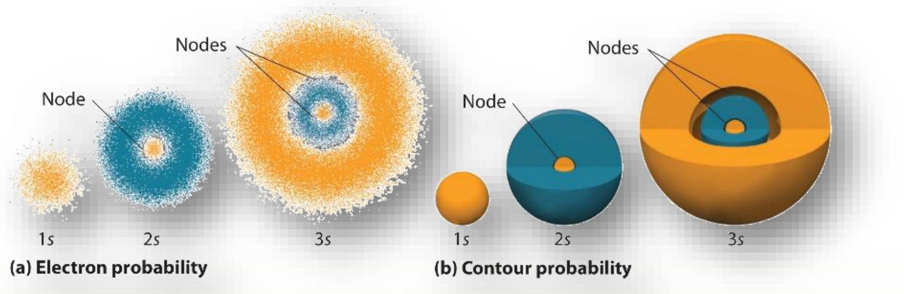
$$\vec{M}_{\perp\vec{Q}} = \hat{\vec{Q}} \times \left(\vec{M}_{\vec{Q}} \times \hat{\vec{Q}}\right)$$

➔ Note that this is essentially a consequence of magnetic scattering arising from a ***dipole-dipole interaction.***

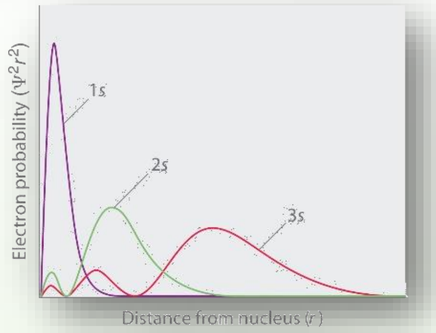
➔ This can be used to differentiate the direction in which the moments point.

Magnetic Form Factor

What is the form factor for a magnetic atom?



➔ Besides the structure factor in Born-approximation we make two more assumptions:
   - The Heitler-London model is valid, thus unpaired electrons are near to equilibrium positions of the magnetic ions.
   - The total angular momentum L and the total spin are good quantum numbers and therefore LS coupling is assumed.



➔ For L = 0, we get (see for example Lovesey): $\mathrm{F_d}\left(\vec{\mathrm{Q}}\right) = \int d^3r \, e^{i\vec{Q}\vec{r}} s_d(\vec{r})$.

Here $s_d(\vec{r})$ is the density of unpaired electrons around ion *d* normalized to their number.

$\mathrm{F_d}\left(\vec{\mathrm{Q}}\right)$ is the ***magnetic form factor*** that allows to consider all electrons of one magnetic atom together and regard $\vec{S}_{ld}$ as the total spin of that atom.

➔ Because the electrons form a cloud around the magnetic ion, and are not centered at the position of the nucleus the form factor falls off as function of $\vec{Q}$.

Magnetic Form Factor

➔ For L ≠ 0, the magnetic form factor gets replaced by:

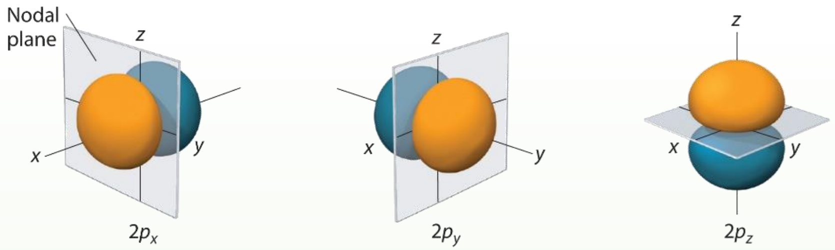
$$\frac{1}{2} g F_d(\vec{Q}) = \frac{1}{2} g_S \mathfrak{I}_0 + \frac{1}{2} g_L \left(\mathfrak{I}_0 + \mathfrak{I}_2\right)$$

where $g = g_S + g_L$,

$$g_S = 1 + \frac{S(S+1) - L(L+1)}{J(J+1)},$$

$$g_L = 1 + \frac{L(L+1) - S(S+1)}{2J(J+1)},$$

$$\mathfrak{I}_n = 4\pi \int_0^\infty j_n\left(Qr\right) r^2 dr$$



Here g is the Landé splitting factor and $j_n(Qr)$ is the nth order spherical Bessel function.

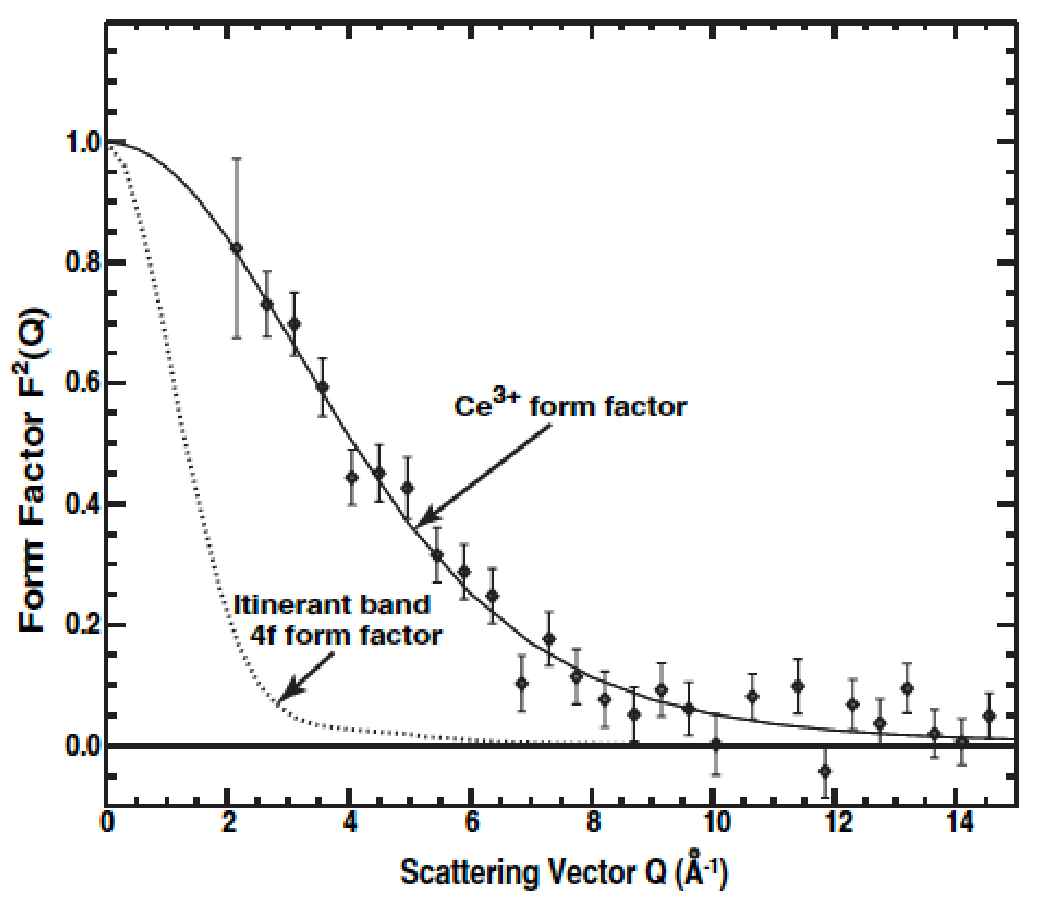
➔ In this case $\vec{S}_{ld}$ needs to be considered as the total angular momentum operator

➔ The magnetic form factor can be approximated by analytical functions of the form:

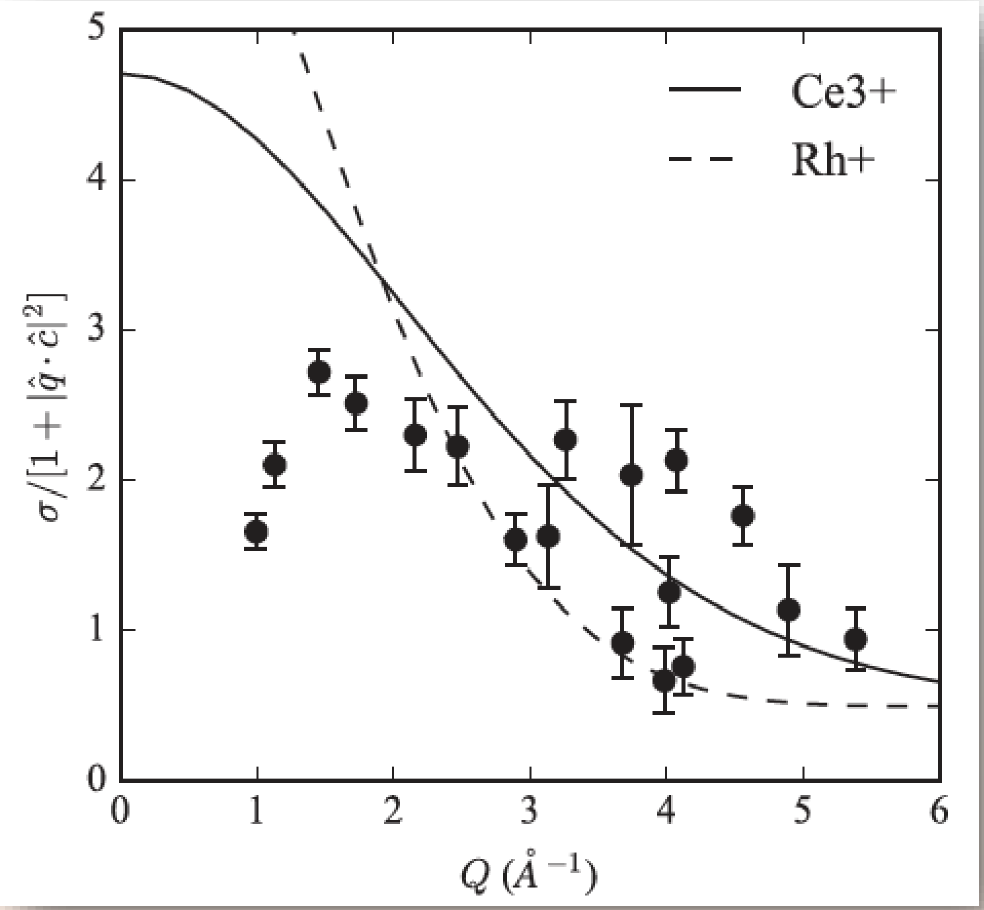
$$\begin{aligned} \langle j_0(s) \rangle &= A\exp(-as^2) + B\exp(-bs^2) + C\exp(-cs^2) + D \quad \text{for} \quad l = 0 \\ \langle j_l(s) \rangle &= As^2\exp(-as^2) + Bs^2\exp(-bs^2) + Cs^2\exp(-cs^2) + Ds^2 \quad \text{for} \quad l \neq 0. \end{aligned}$$

➔ All the parameters are tabulated (for example Neutron Scattering Handbook)

Magnetic Form Factor

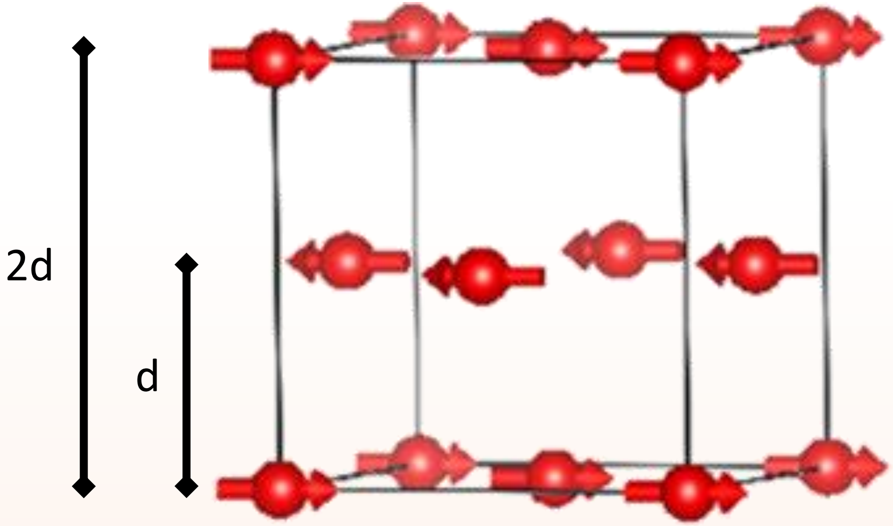


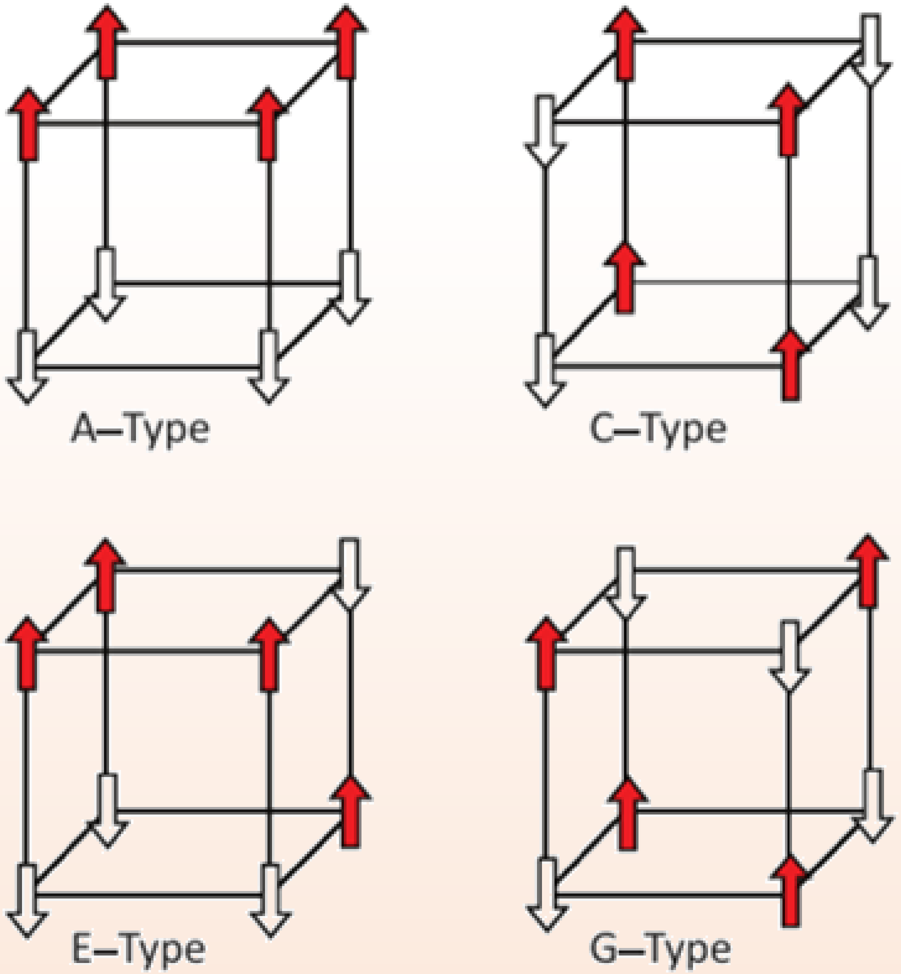
A. P. Murani, *et al.*, PRL **95,** 256403 (2005)



D. M. Fobes *et al.*, JPCM 29 17LT01 (2017)

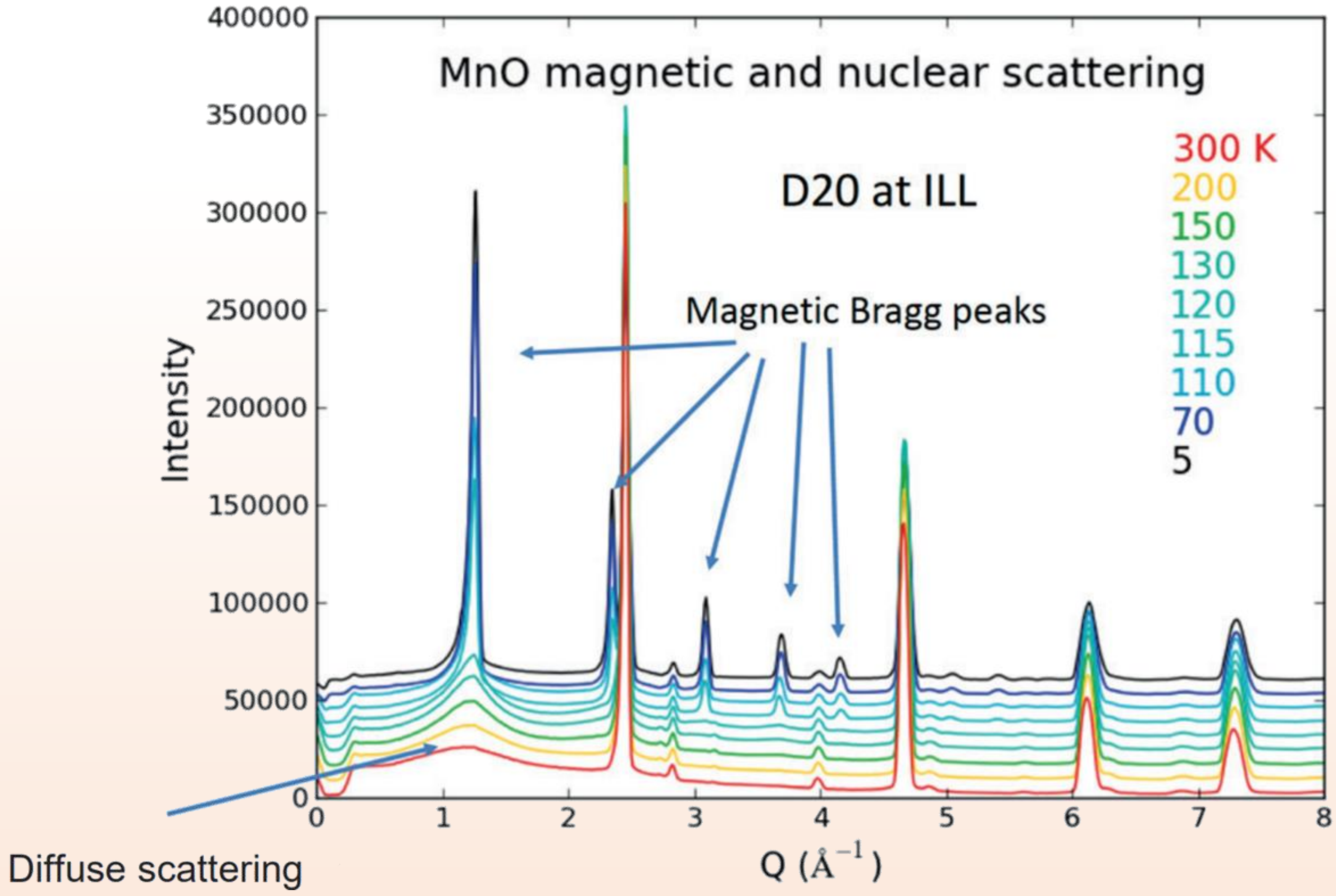
Diffraction from Anti-Ferromagnet

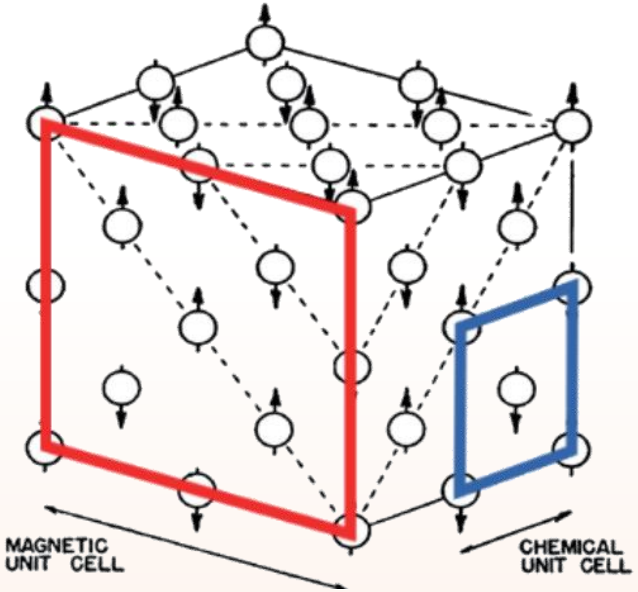




- Periodicity of anti-ferromagnets is larger then the underlying atomic structure
- In the simplest case there is a doubling of the unit cell in one direction
- This leads to additional Bragg-peaks at forbidden positions (1/2 order Bragg-peaks)

Diffraction from Anti-Ferromagnet





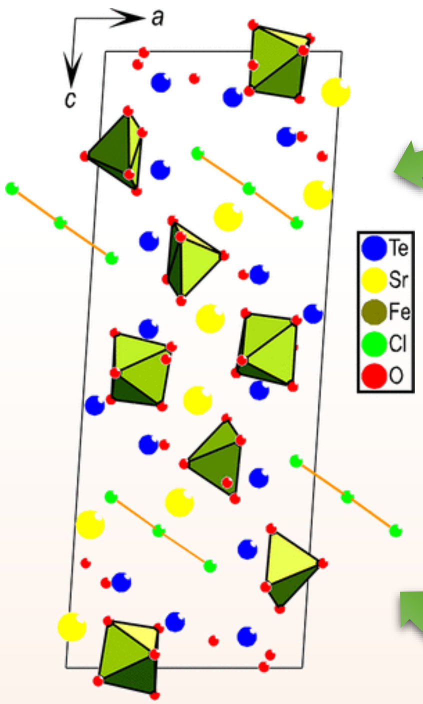
B. C. Chakoumakos and J. B. Parise,
https://www.elementsmagazine.org/probing-phase-transitions-and-magnetism-in-minerals-with-neutrons/

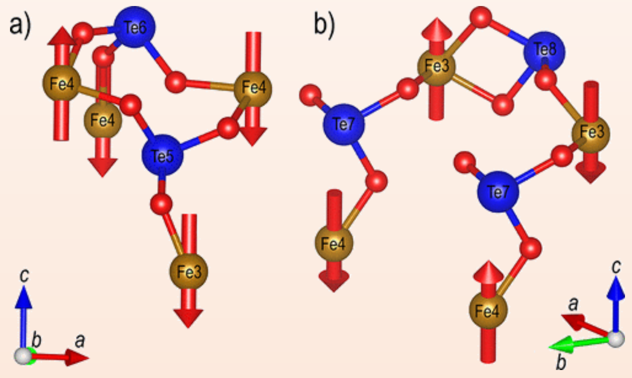
Diffraction from Anti-Ferromagnet

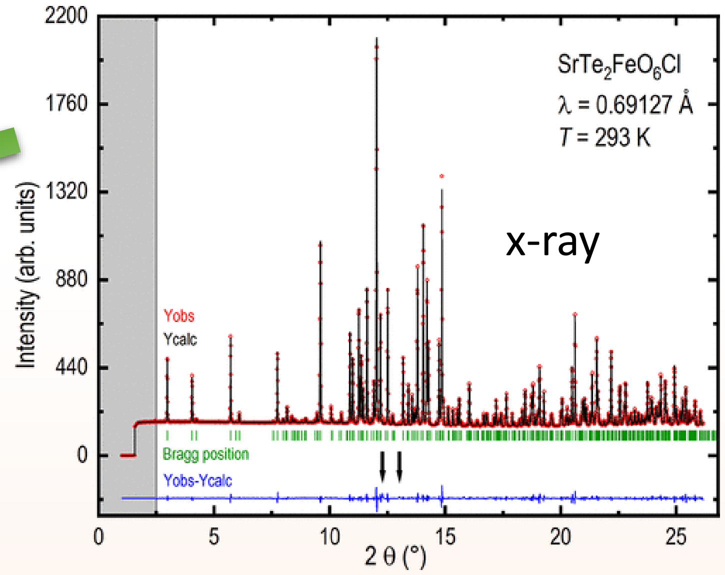
Recent results from HRPT+DMC at SINQ:

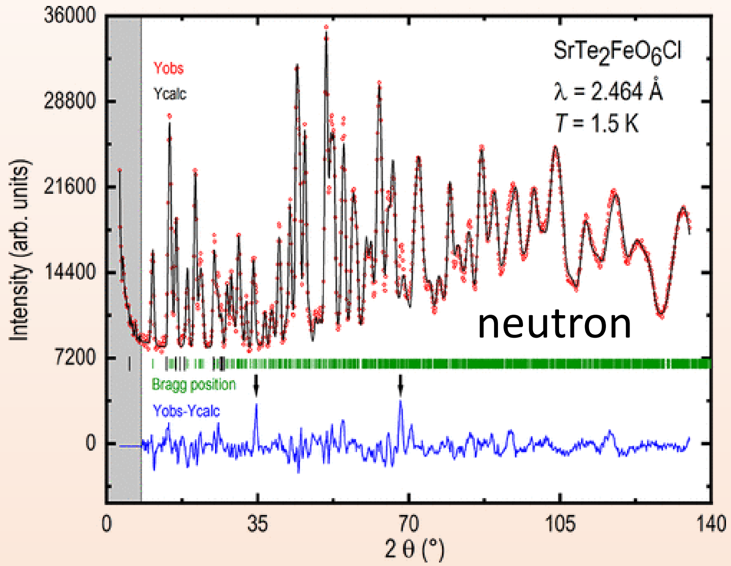
- Complex system $SrTe_2FeO_6Cl$
  - monoclinic with 88 atoms per UC
  - heavy and light elements
  - low temperature magnetic structure
- Combined use of x-ray and neutron diffraction (at two temperatures) to solve nuclear and magnetic structure
- Refinement yields very precise lattice parameter + atomic positions and thermal motion parameters

Results XRD+ND refinement:	
chemical formula	$SrTe_2FeO_6Cl$
crystal system	monoclinic
space group	$P12_1/n1$ (no. 14)
a (Å)	10.2604(1)
b (Å)	5.34556(5)
c (Å)	26.6851(3)
β (°)	93.6853(4)
R_P (%)	1.32

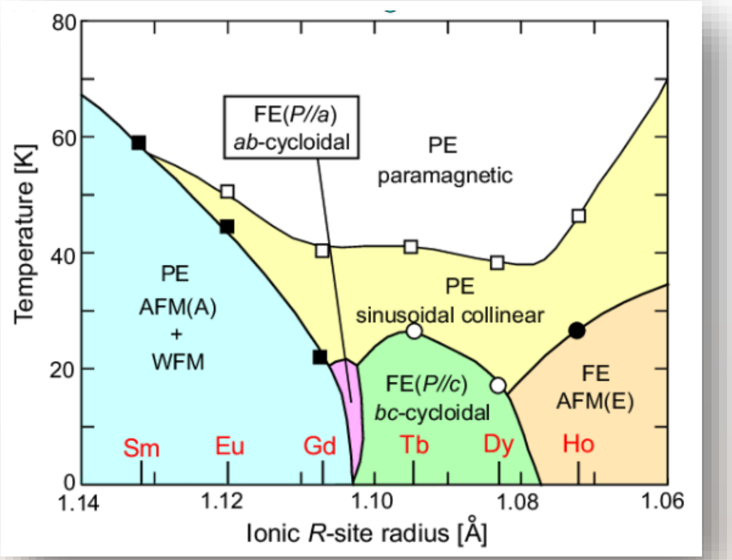






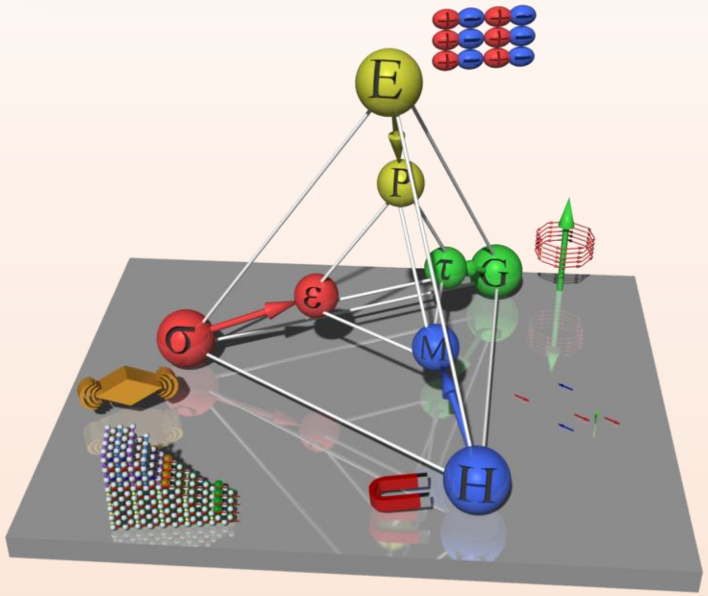


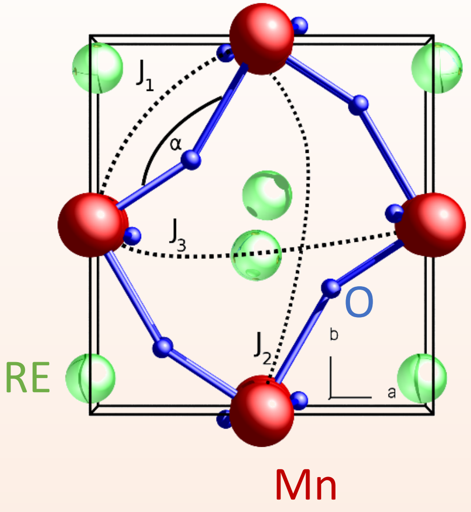
Incommensurate Magnetic Structures in $TbMnO_3$

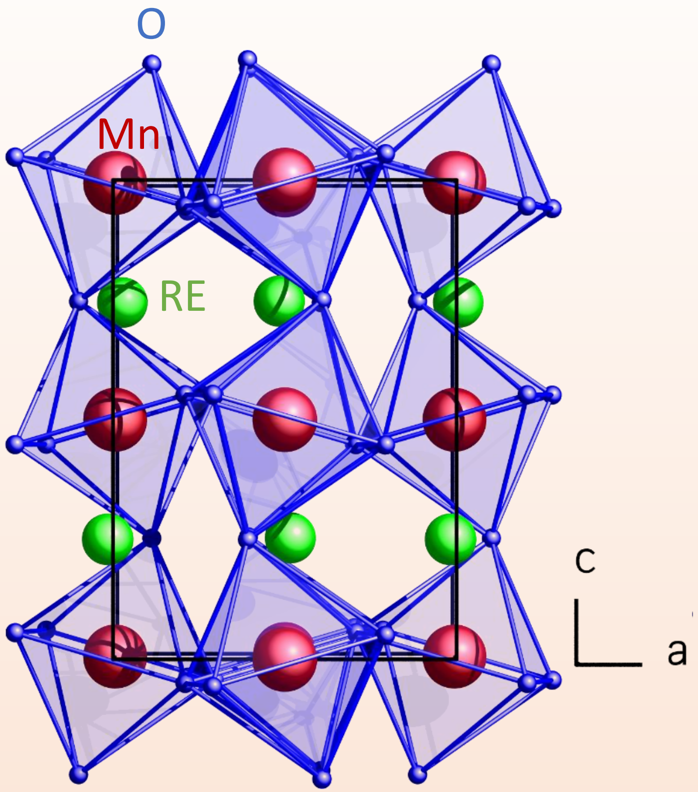


$TbMnO_3$ is a multiferroic material:

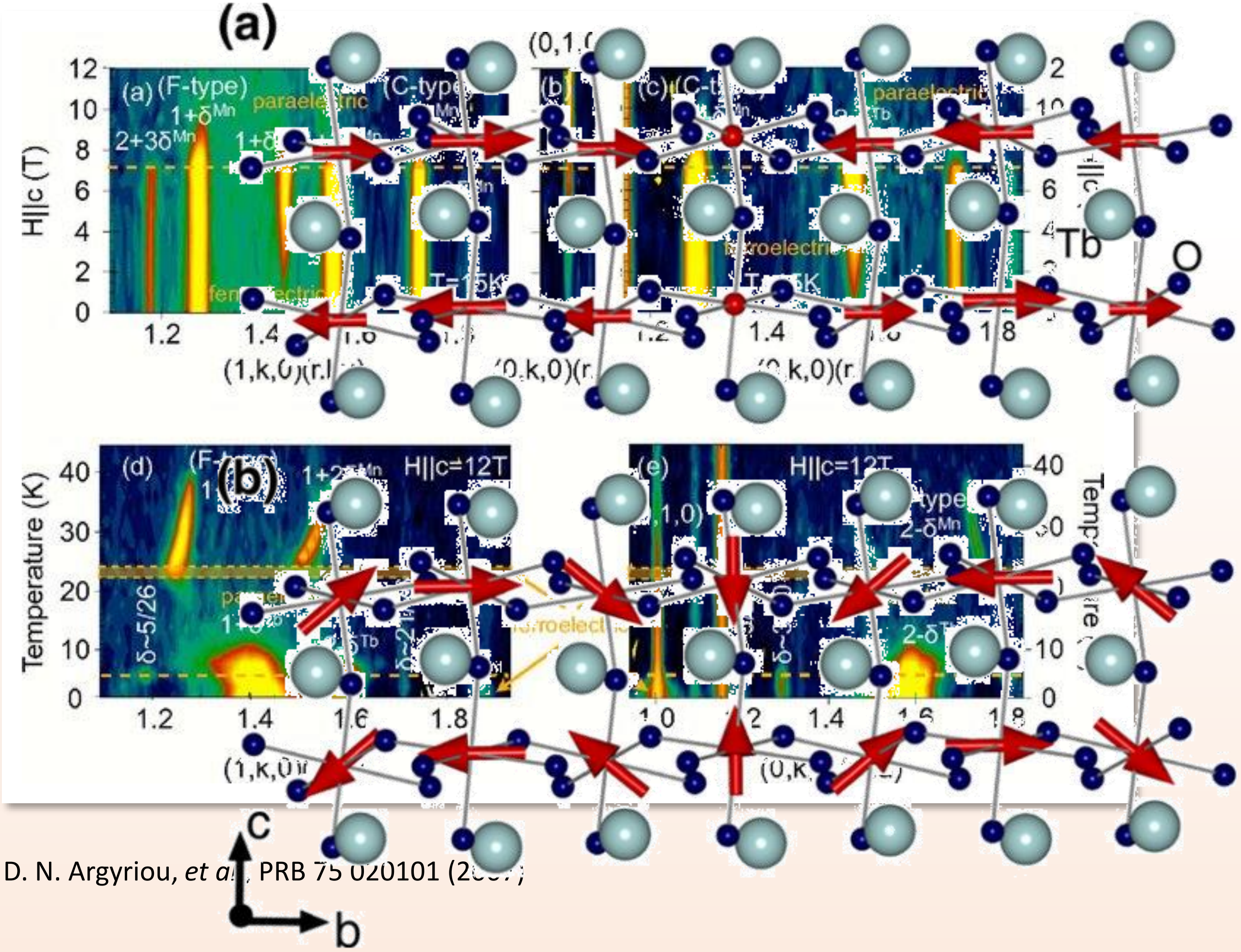
simultaneous magnetic and ferro-electric order



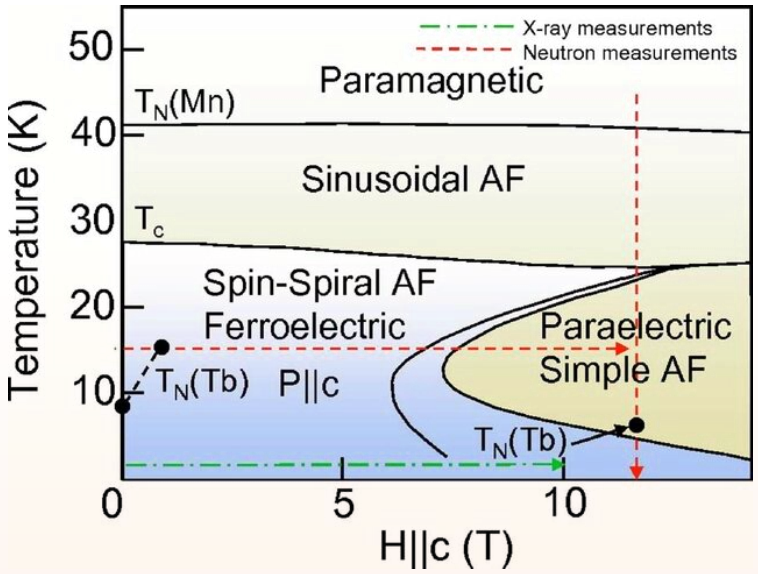


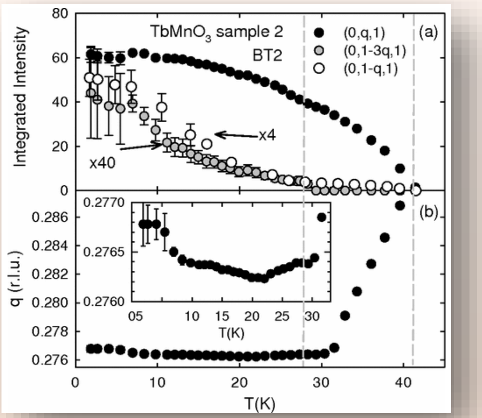


Incommensurate Magnetic Structures in $TbMnO_3$



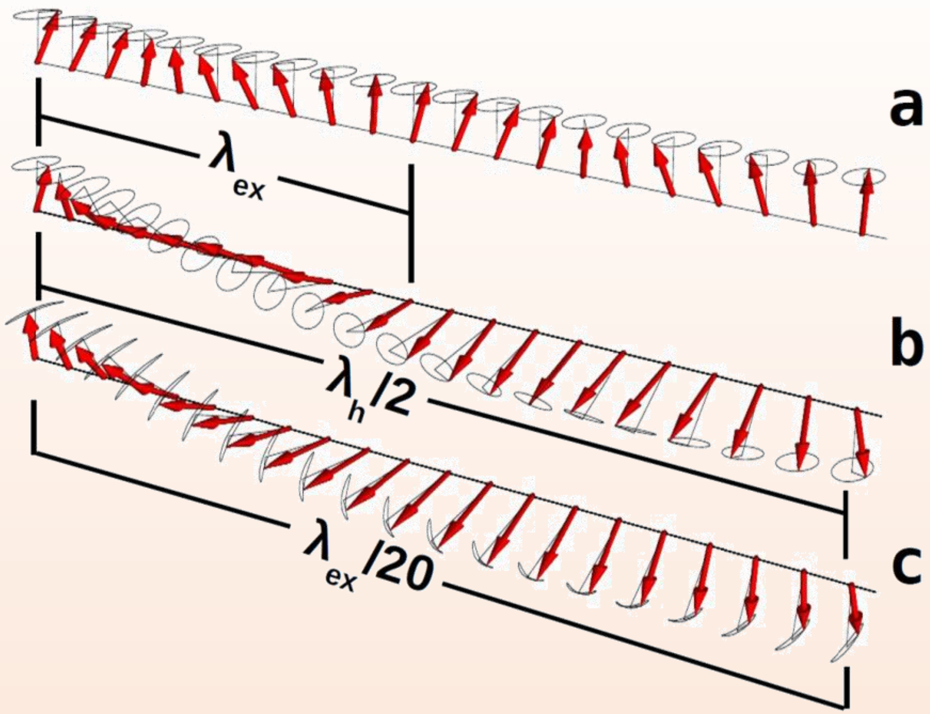
D. N. Argyriou, *et al.*, PRB 75 020101 (2007)





M. Kenzelmann, *et al.*, Phys. Rev. Lett. 95, 087206 (2005)

Helimagnon



M. Kugler, *et al.*, Phys. Rev. Lett. 115, 097203 (2015)