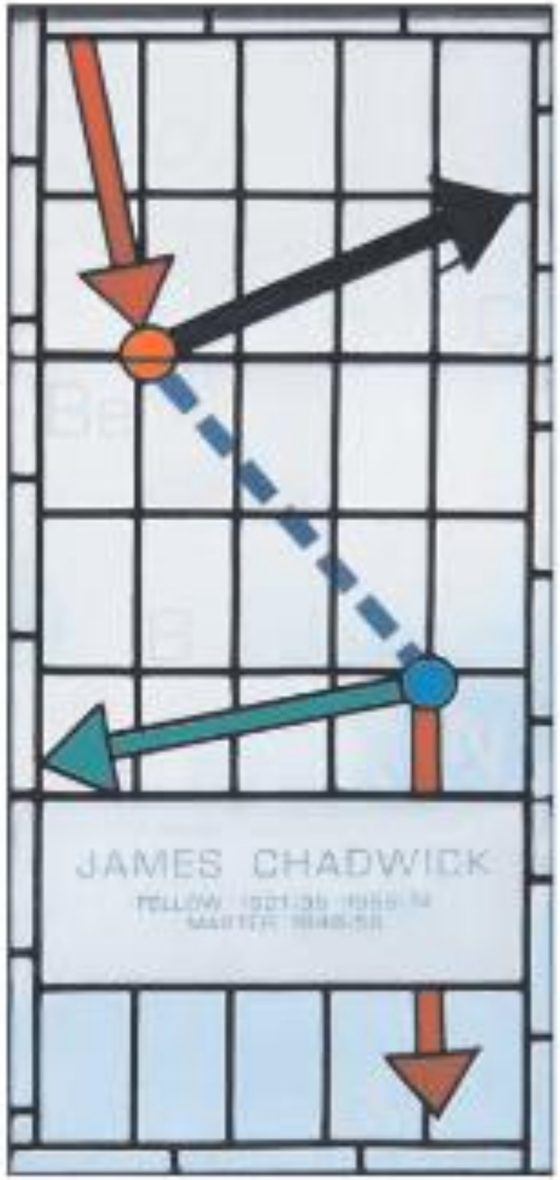
Magnetic SANS

ELIZABETH BLACKBURN, 2021-06-01

Outline

- The difference between nuclear and magnetic scattering of neutrons
- What does that mean for our experiments?
- How can we optimize our experiments to take advantage of this?
- What do we need to account for when measuring magnetic materials?

$${}^{4}_{2}\alpha + {}^{9}_{4}\mathrm{Be} \rightarrow {}^{12}_{6}\mathrm{C} + {}^{1}_{0}\mathrm{n}$$



What do neutrons interact with?

Basic properties of the neutron	
mass	1.675×10^{-27} kg
charge	0
spin	½
magnetic dipole moment	-1.913 nuclear magnetons (= 0.001 μ_B)

Density of what? What do neutrons couple to?

- Nuclei, via the strong force.
  - Short-range interaction (especially compared to the neutron wavelength λ).
  - So, we approximate the scattering potential of a single nucleus at position $\boldsymbol{r}$ as:

$$V(\boldsymbol{r}) = \frac{2\pi\hbar^2}{m} b\, \delta(\boldsymbol{r})$$

  - b is the scattering length of the nucleus.

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The **Bohr magneton**

$$\mu_B = \frac{e\hbar}{2m_{\mathrm{electron}}} = 9.274 \times 10^{-24}\ [\mathrm{Am}^2]$$

- Magnetic fields
  - In a solid, these are generated by magnetic nuclei and electrons.
  - Because of the enormous difference in strength between the nuclear magneton and the (electron) Bohr magneton, we will ignore scattering from magnetic nuclei here (although it can be studied).
  - We mostly assume that we can model the magnetic fields by placing magnetic moments on atoms/ions.

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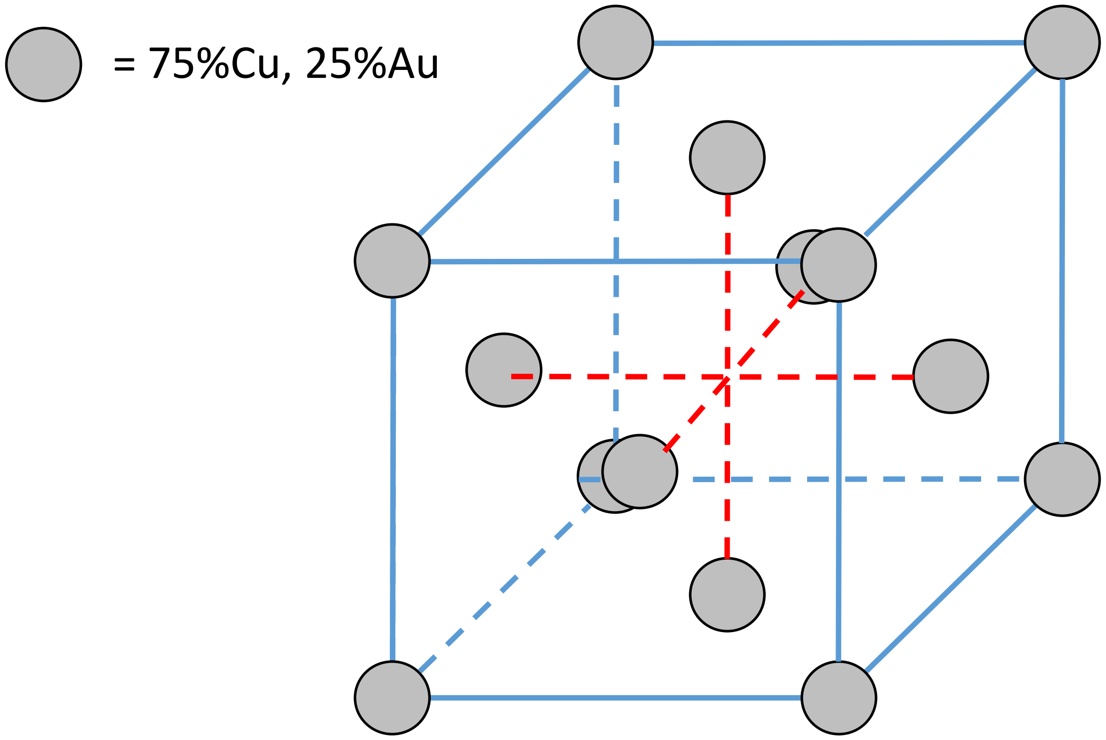
How does this help us with looking at magnetic structures?

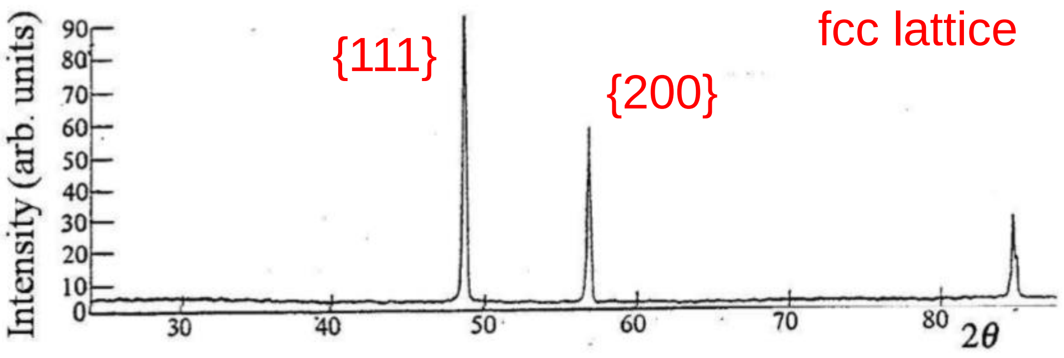
The variations in magnetic field brought about by the electronic magnetic moments are not uniform in space, and so provide spatial variations that can be scattered off.

To start with, we can consider it as providing different labels for scattering sites.

Cu_3Au at high temperatures



X-ray powder diffraction



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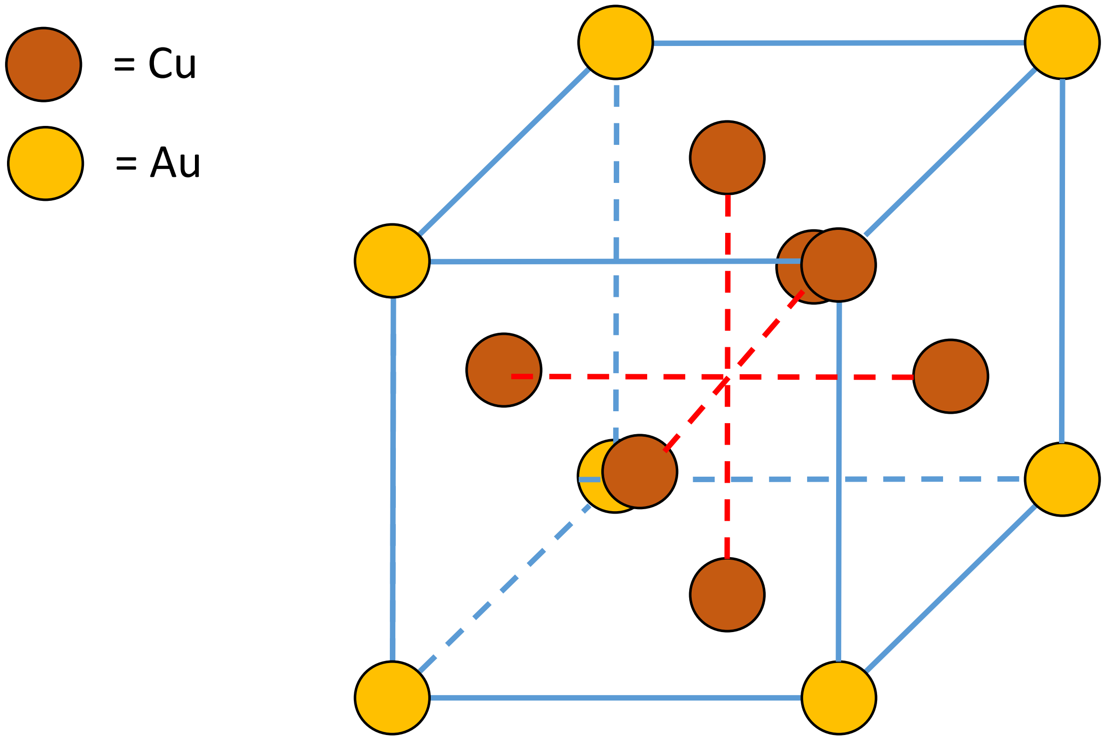
Cu_3Au cooled down



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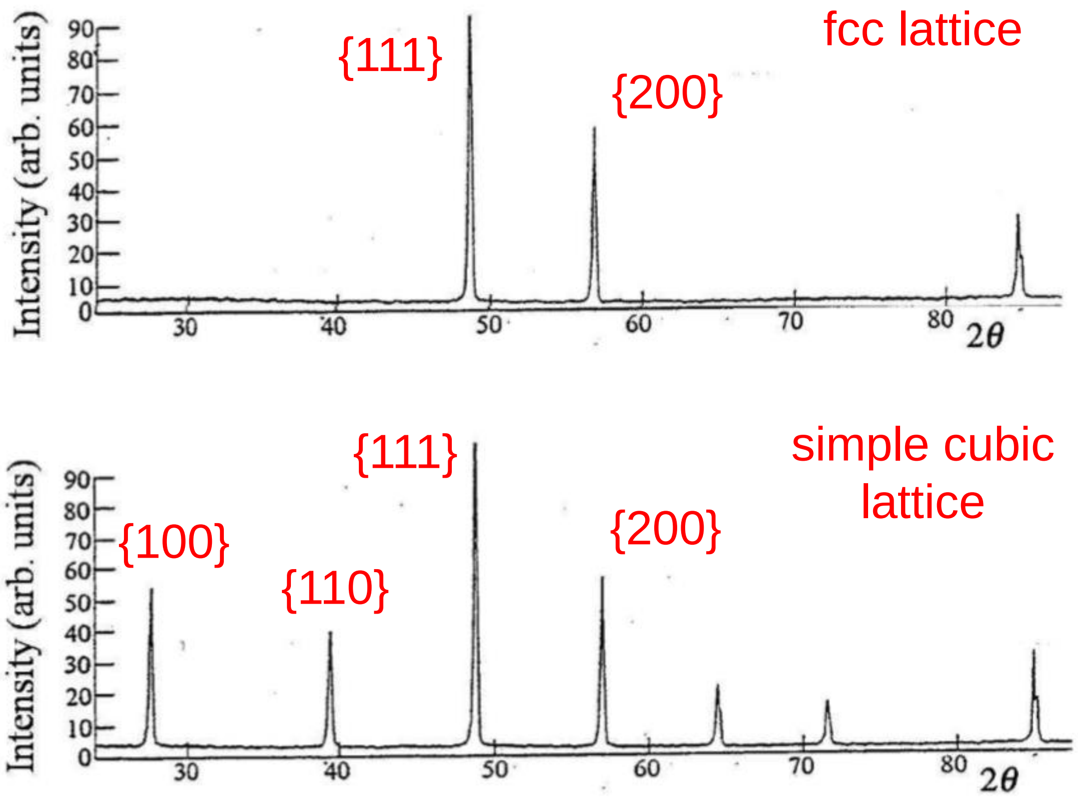

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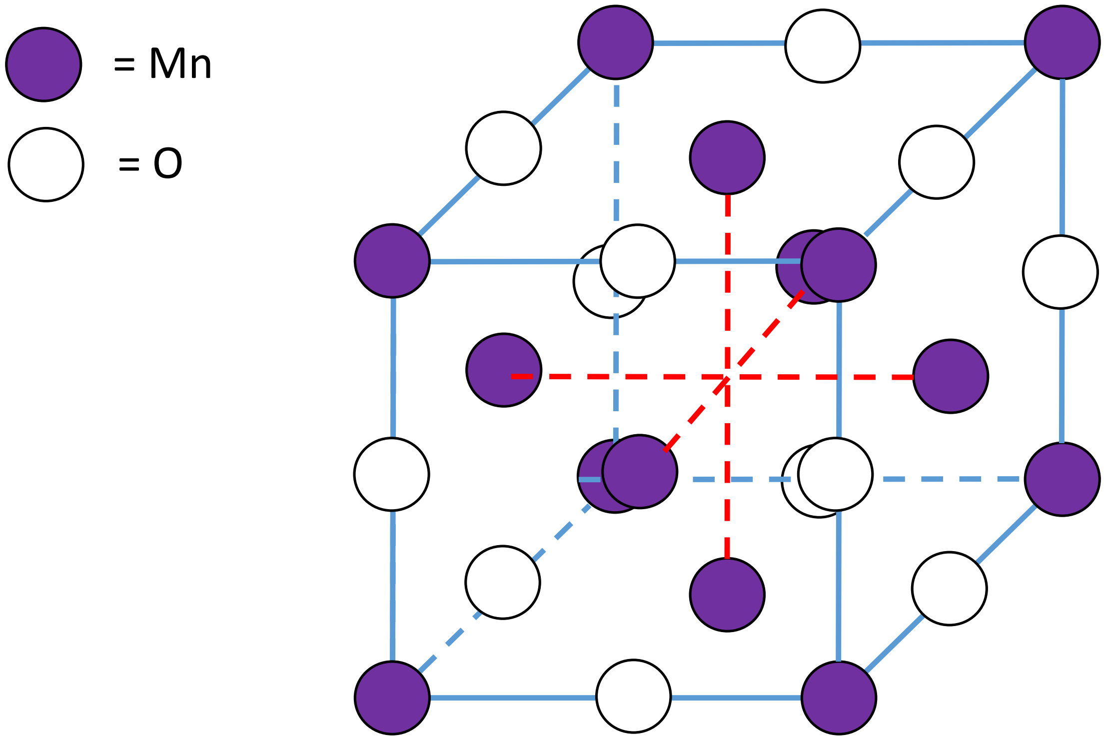
MnO – now a magnetic example



Neutron powder diffraction

300 K – sample is paramagnetic; fcc lattice

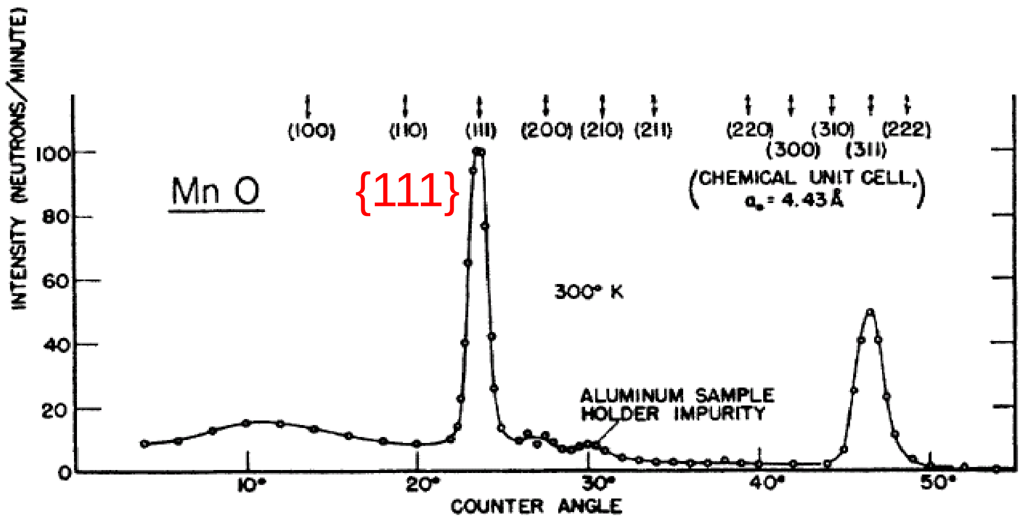


FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.

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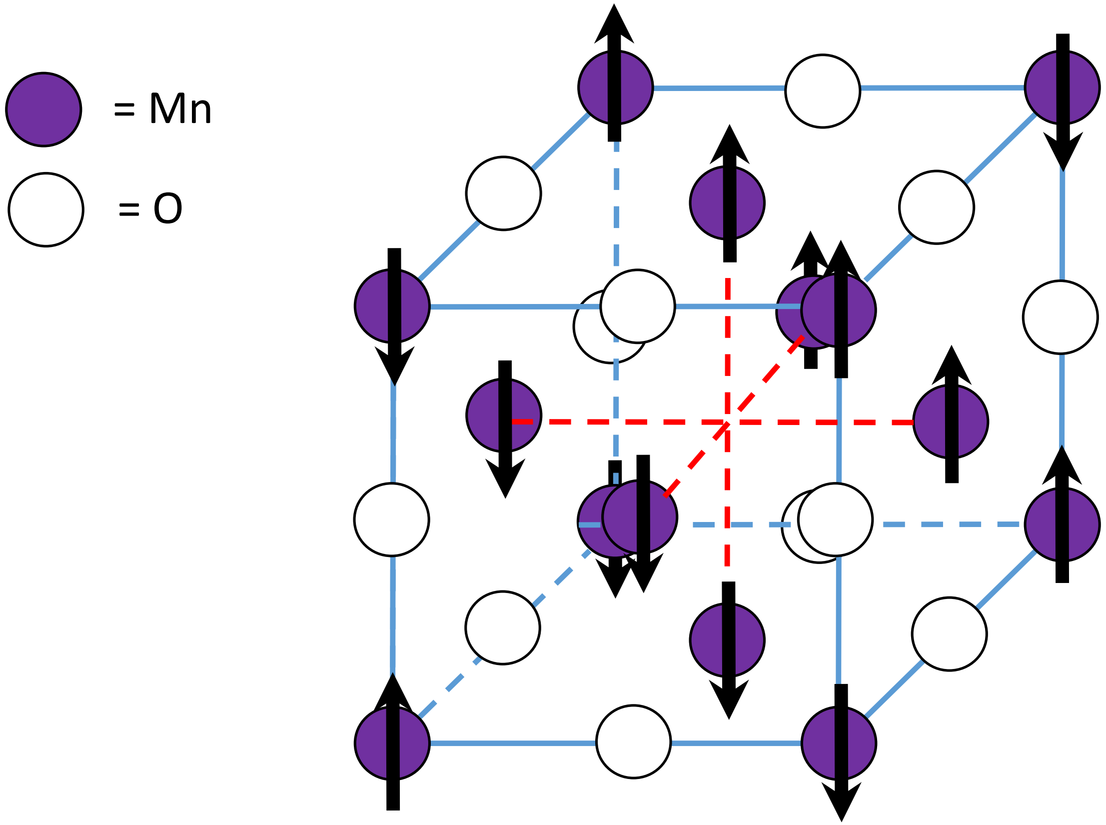
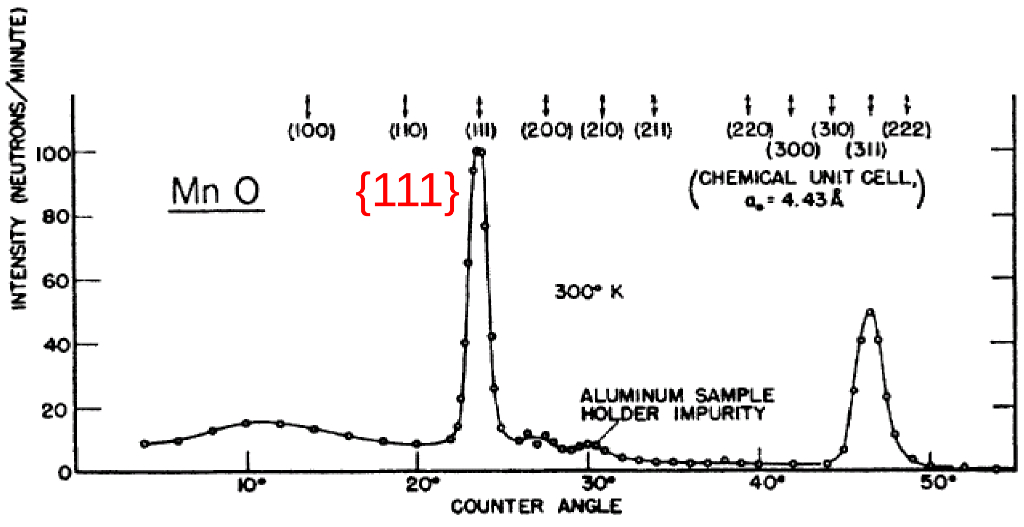


Neutron powder diffraction



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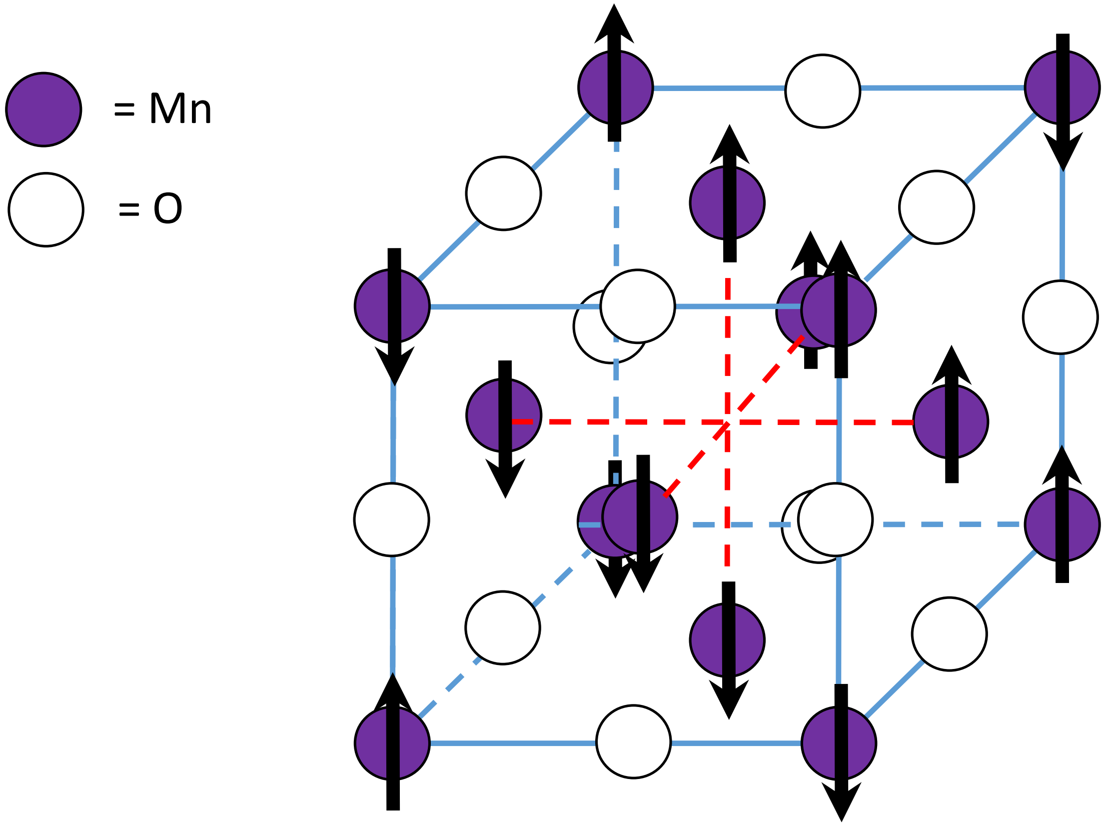

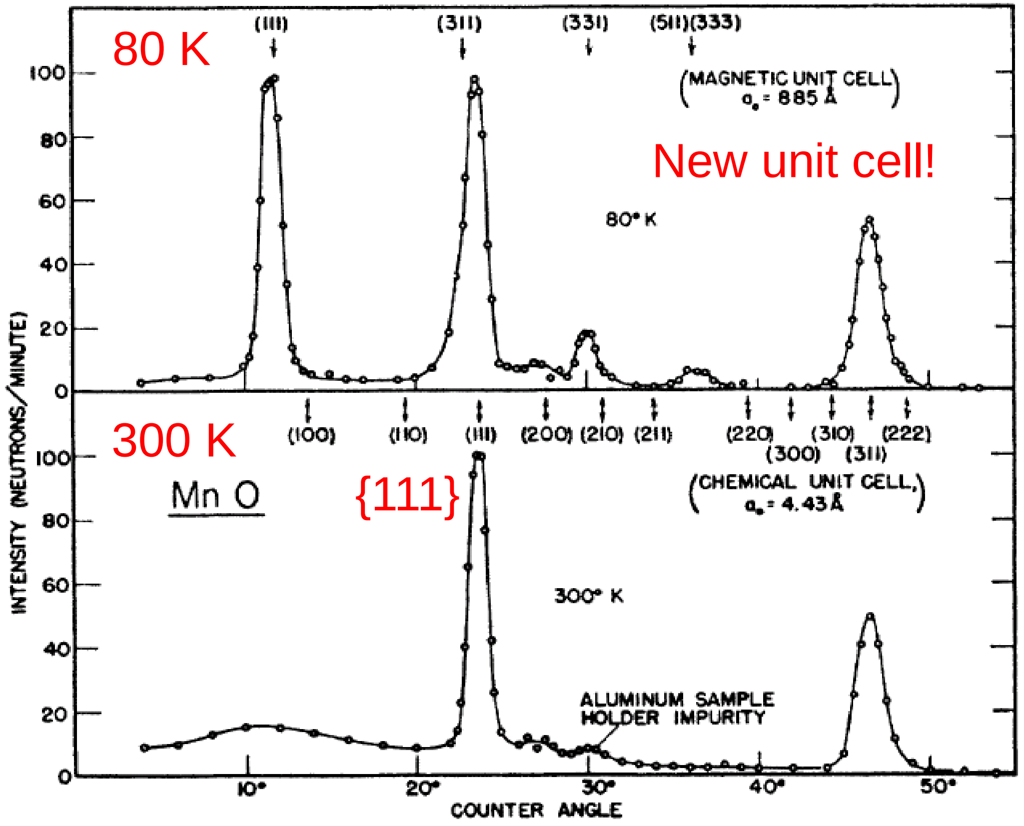

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Magnetic moments are vectors

This perhaps obvious statement has a number of important consequences.

- The magnetic moment is not just a label; the moment direction matters.
  - Possible directions may be restricted by the nature of the atom, or the orientation of the electron orbitals in the crystal lattice.

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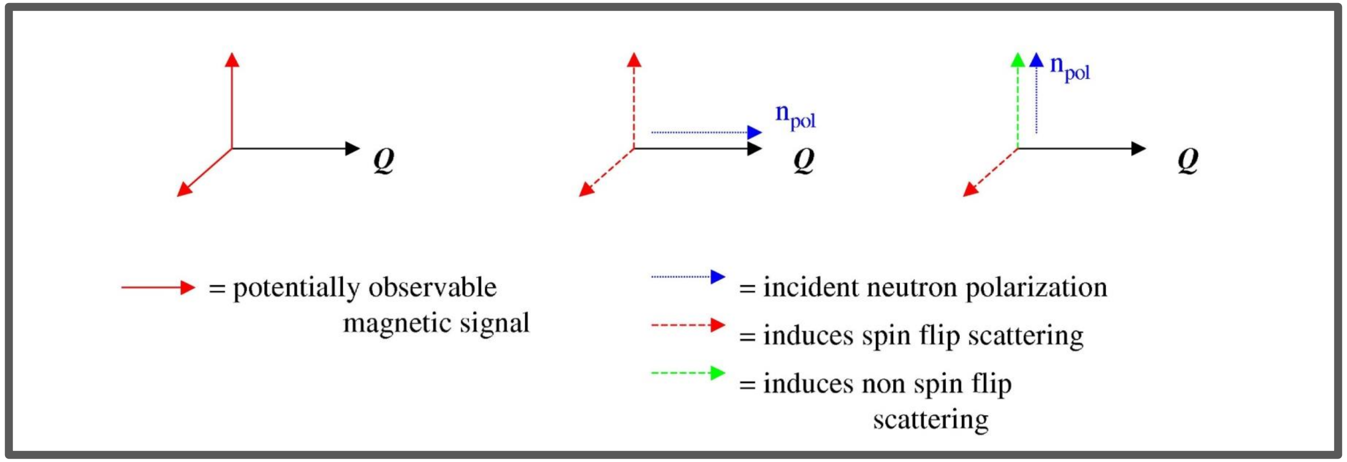
- We can detect the magnetic moment direction because the scattering interaction is, at root, one between two dipoles – the atomic magnetic moment and the neutron magnetic dipole.
  - Only magnetization (magnetic moments) that is perpendicular to ***Q***, the scattering vector, generates a signal.
  - Extra info if the neutron beam is polarized.



An interlude on neutron scattering theory

In a neutron scattering experiment, the measured quantity is the number of neutrons into a detector.

From this, we can get the *differential cross-section*.

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{\textit{number of neutrons scattered per second into } \mathrm{d}\Omega \textit{ in the direction } \theta, \phi}{\textit{neutron flux } (\Phi) \,.\, \mathrm{d}\Omega}$$

If we have the experimental capability, we can also measure the *partial differential cross-section,* which adds energy discrimination.

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'} = \frac{\textit{as above, with final energy between } E' \textit{and } E' + \mathrm{d}E'}{\textit{neutron flux } (\Phi) \,.\, \mathrm{d}\Omega . \mathrm{d}E'}$$

The full and complete derivation can be found in the books in the bibliography; one uses Fermi's golden rule to get:

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'}\right)_{\sigma\lambda \to \sigma'\lambda'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar 2}\right)^2 |\langle \boldsymbol{k}'\sigma'\lambda'|V|\boldsymbol{k}\sigma\lambda\rangle|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

where V is the interaction potential, $\boldsymbol{k}$ refers to the neutron wavevector, σ refers to the neutron spin state, and λ refers to the state of the system being scattered off. $\hbar\omega$ is the energy change of the neutron.

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An interlude on neutron scattering theory

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

To proceed further, we need to know about the interaction potential. For nuclear scattering, as already stated, where the strong force is involved, we use an approximation

$$V(\boldsymbol{r}) = \frac{2\pi\hbar^2}{m} b\,\delta(\boldsymbol{r})$$

we treat the nucleus as a point scatterer

where b is the scattering length for a given nucleus.

For magnetic scattering, the interaction potential is more complicated, as the forces are not central and exist over longer ranges, and we have to consider the vectorial nature of the magnetic moments.

The magnetic equivalent for scattering from one unpaired electron is

the electron is distributed over a larger spatial area

$$V = -\boldsymbol{\mu}_n \cdot \boldsymbol{B} = -\frac{\mu_0}{4\pi}\gamma\mu_N 2\mu_B \boldsymbol{\sigma} \cdot \left(\mathrm{curl}\left(\frac{\boldsymbol{s}\times\widehat{\boldsymbol{R}}}{R^2}\right) + \frac{1}{\hbar}\frac{\boldsymbol{p}\times\widehat{\boldsymbol{R}}}{R^2}\right)$$

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Magnetic interaction between a neutron and an electron

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

We can insert our potential for the neutron in the magnetic field of an electron, eventually adding in multiple electrons or moments at periodic sites (labelled as i). The spin and orbital components cannot be separated as they manifest as one magnetic field. We can then write the cross-section in terms of the Fourier transform of the magnetization.

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

where

$$\boldsymbol{M}_\perp(\boldsymbol{Q}) = \widehat{\boldsymbol{Q}} \times \left(\boldsymbol{M}(\boldsymbol{Q}) \times \widehat{\boldsymbol{Q}}\right)$$

and

$$\boldsymbol{M}(Q) = \sum_i f_i(\boldsymbol{Q})\boldsymbol{m}_i^{\boldsymbol{k}} \exp(i\boldsymbol{Q}.\boldsymbol{r}_i)\exp(-Wi)$$

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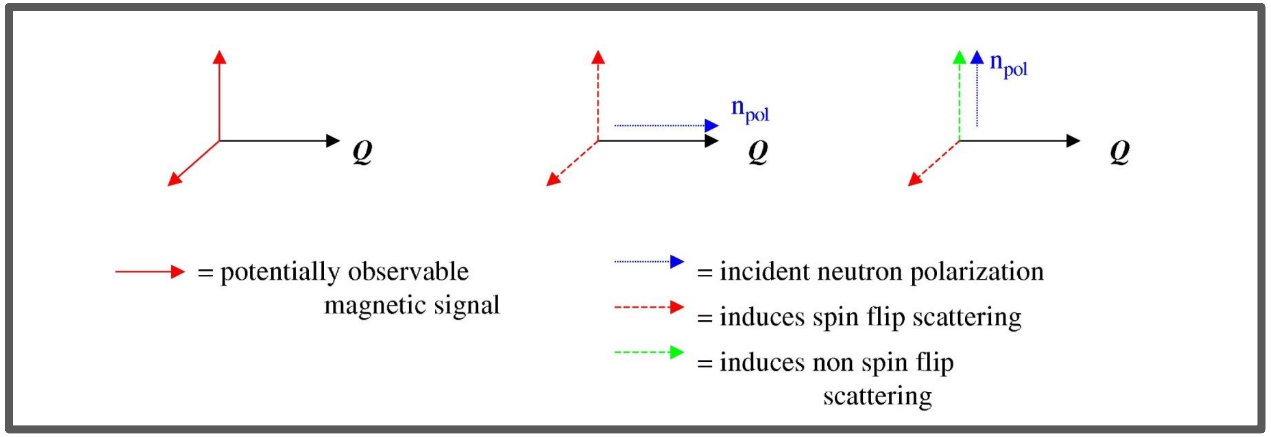
Magnetic interaction between a neutron and an electron

$$\boldsymbol{M}_{\perp}(\boldsymbol{Q}) = \widehat{\boldsymbol{Q}} \times \left(\boldsymbol{M}(\boldsymbol{Q}) \times \widehat{\boldsymbol{Q}}\right)$$



individual moments

Debye-Waller factor

$$\boldsymbol{M}(Q) = \sum_i f_i(\boldsymbol{Q})\boldsymbol{m}_i^{\boldsymbol{k}} \exp(i\boldsymbol{Q}.\boldsymbol{r}_i) \exp(-Wi)$$

lattice information

form factor

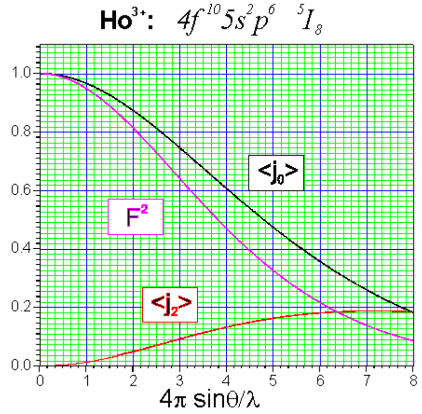


Image: ETHZ

Form factor: the Fourier transform of the magnetization distribution of a single atom/ion. Tabulated as a series of Bessel functions for all ions in the Neutron Data Booklet.

The magnetization distribution is long-range in real space and so this envelope function kills off magnetic signal at large $\boldsymbol{Q}$.

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Magnetic moments are vectors

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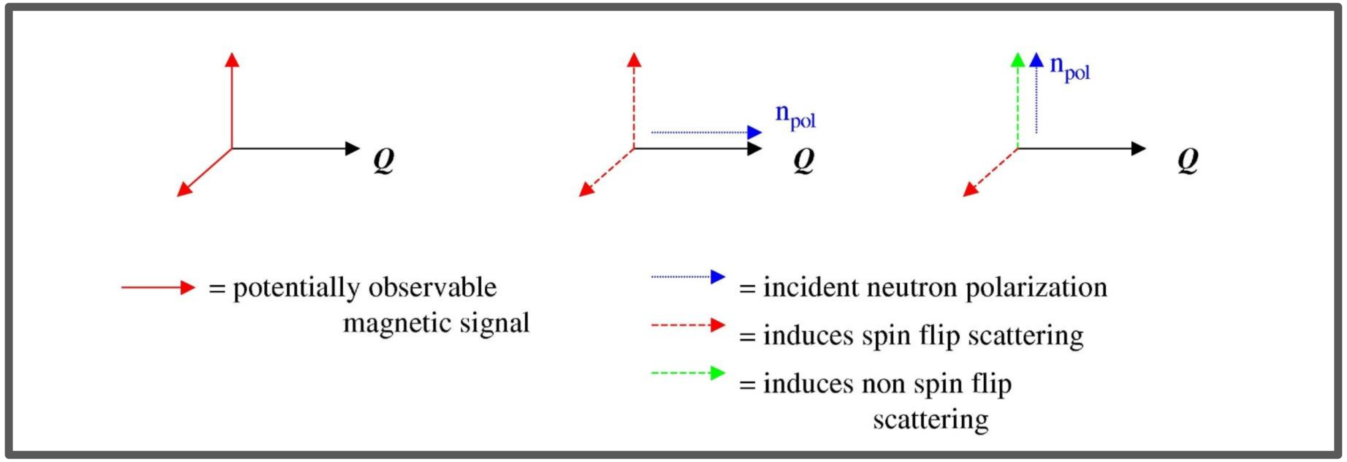

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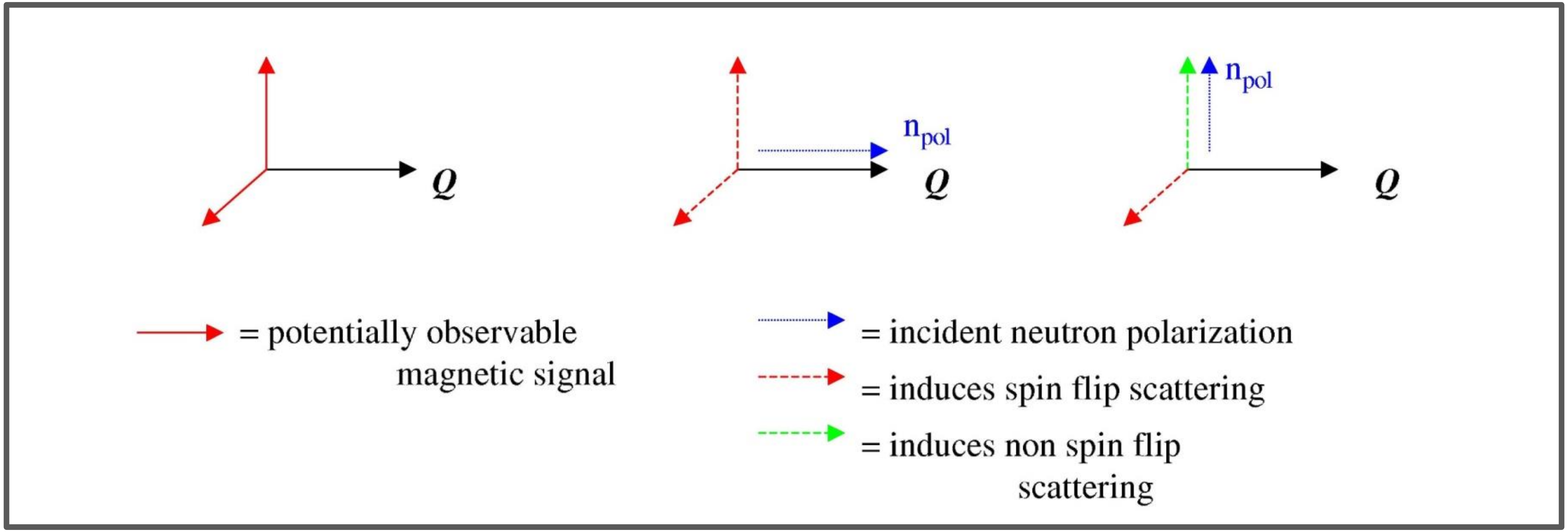
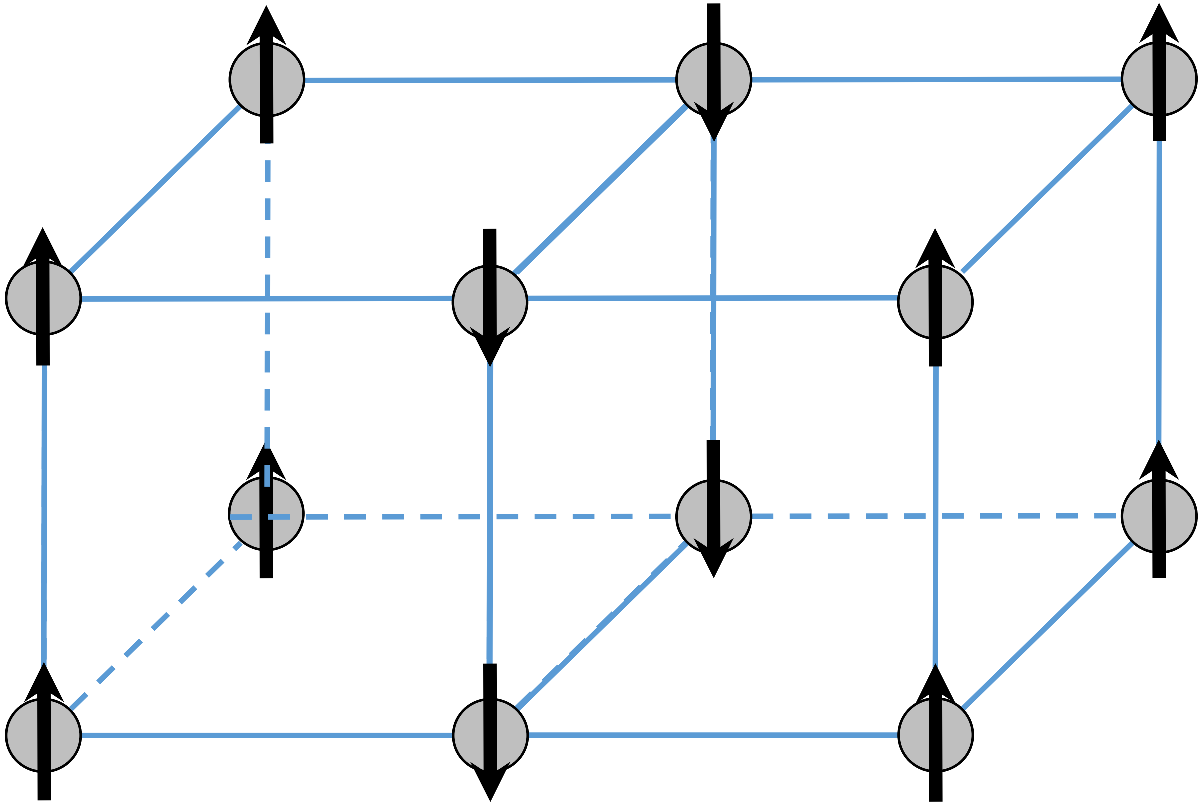
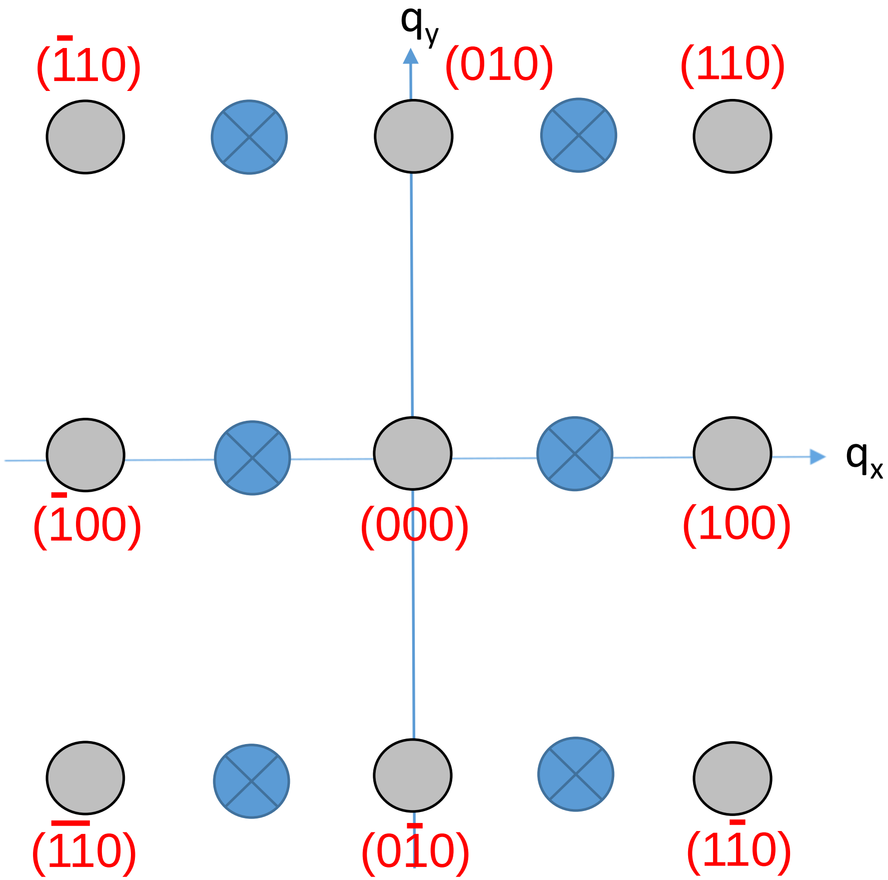


What does that mean for our experiments?

Real space



Reciprocal space



$$\boldsymbol{q}_{\mathrm{magnetic}} = (½\ 0\ 0)$$

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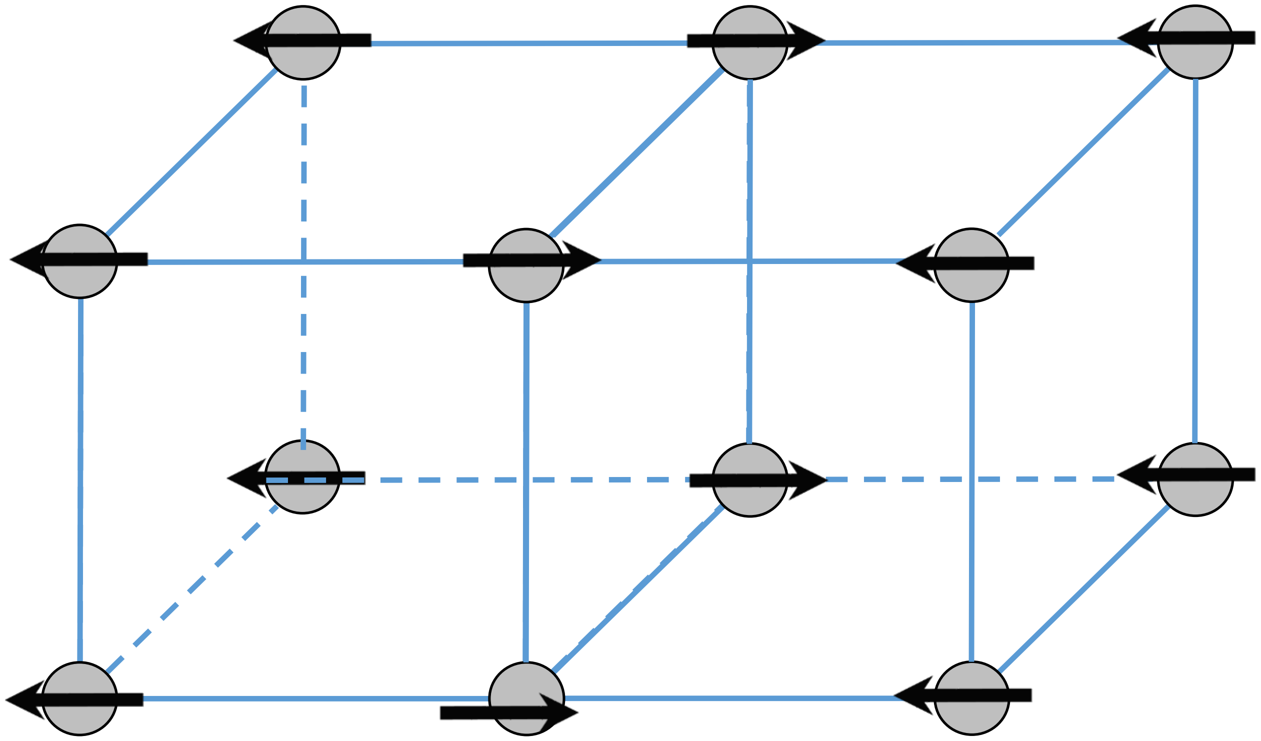
What does that mean for our experiments?

Real space



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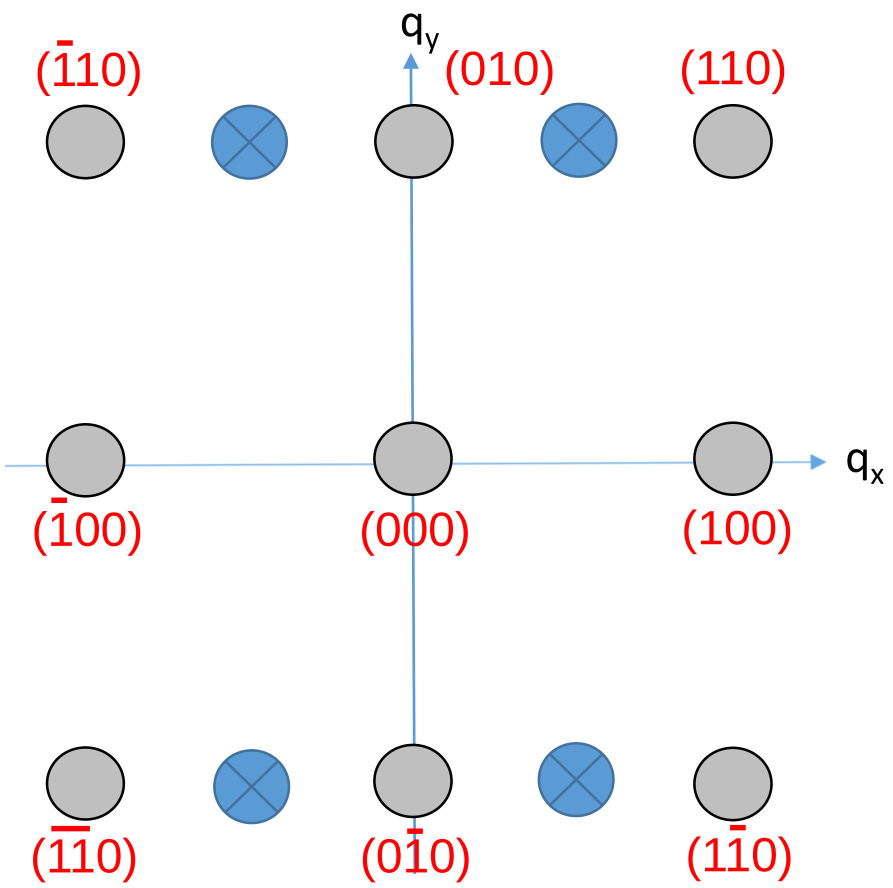
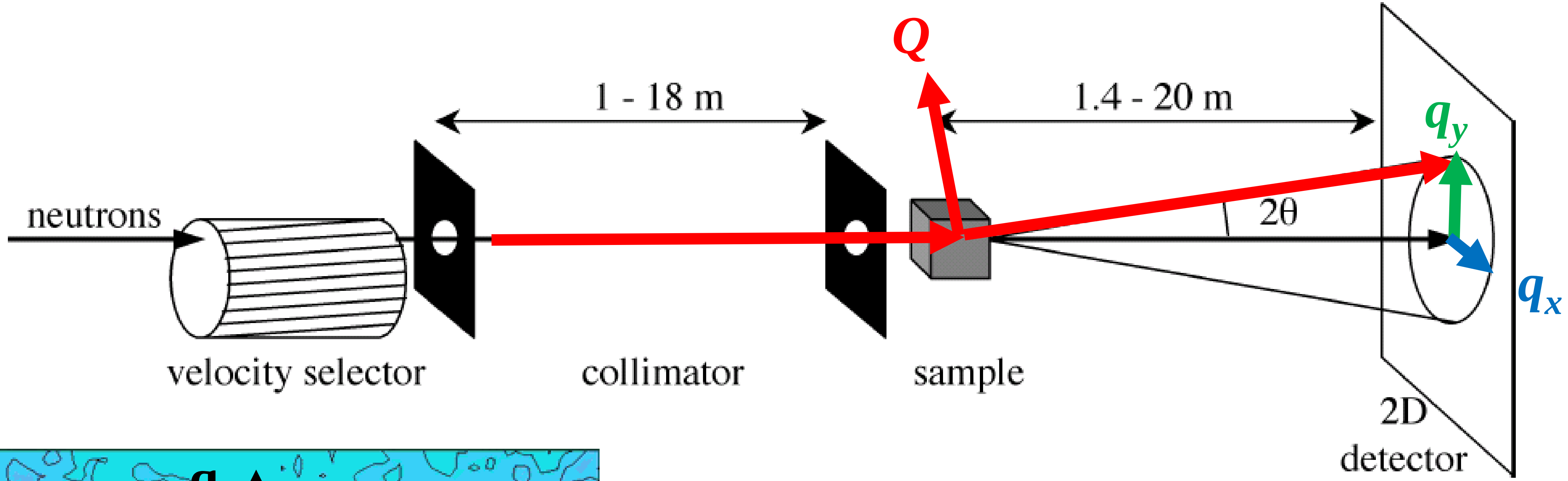


$$\boldsymbol{q}_{\text{magnetic}} = (\tfrac{1}{2}\ 0\ 0)$$

What does that mean for our experiments?



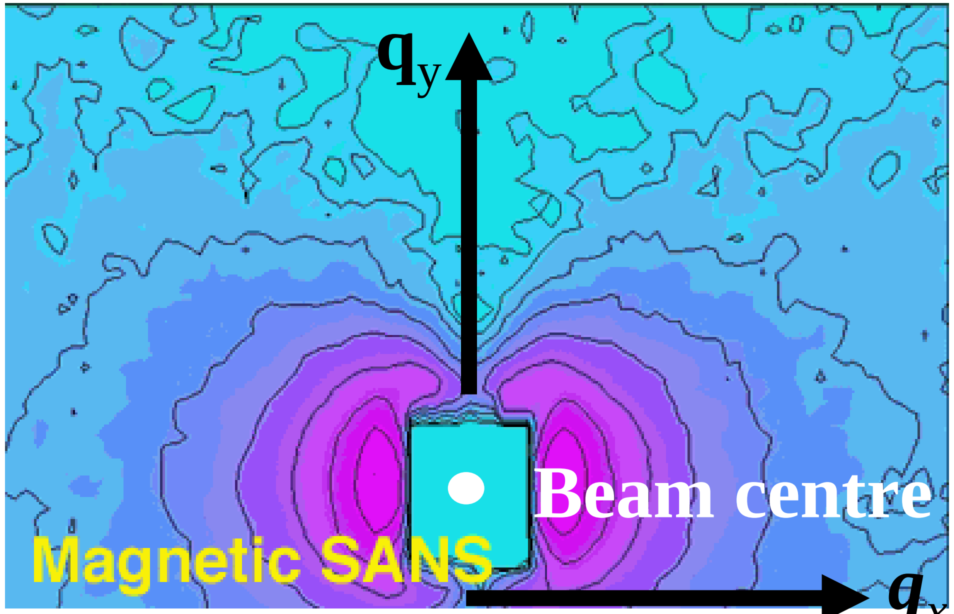
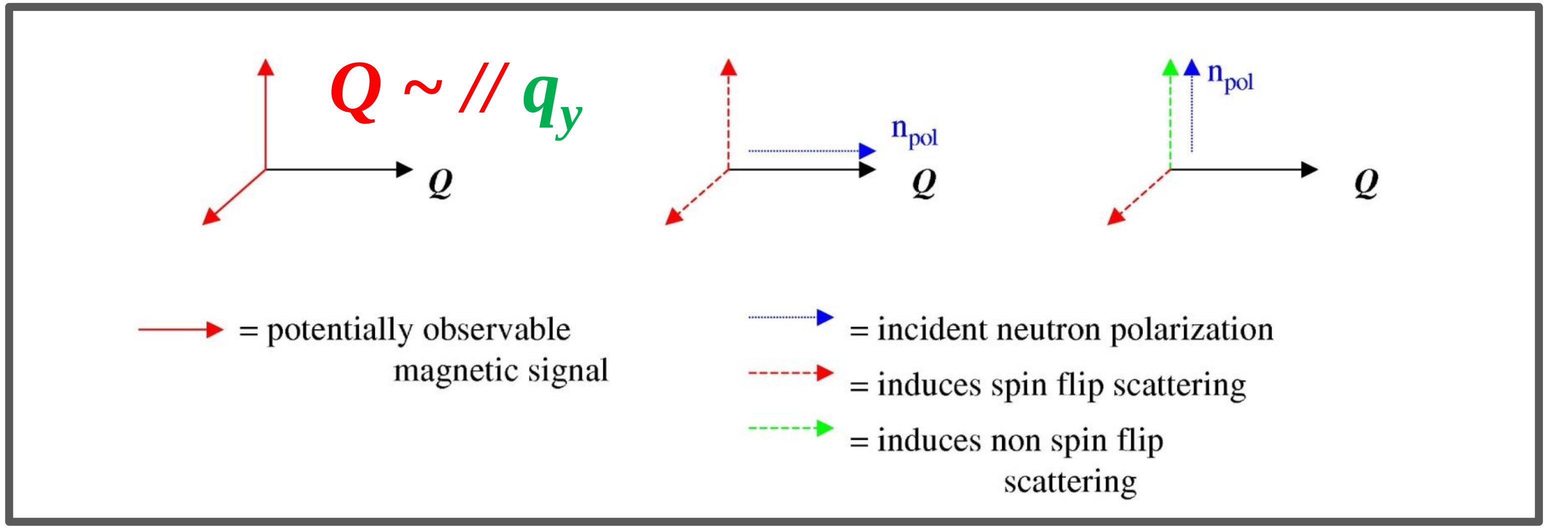


Image credit: ILL



Changes in the magnetization direction

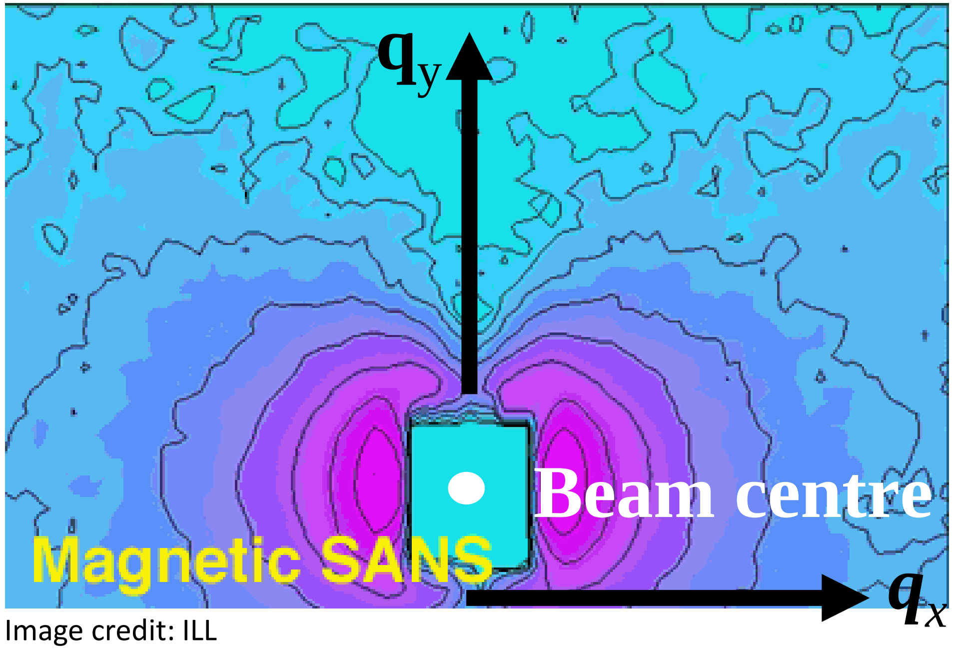
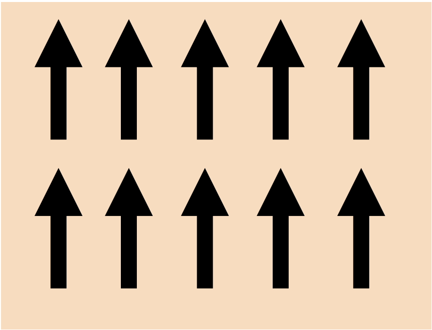


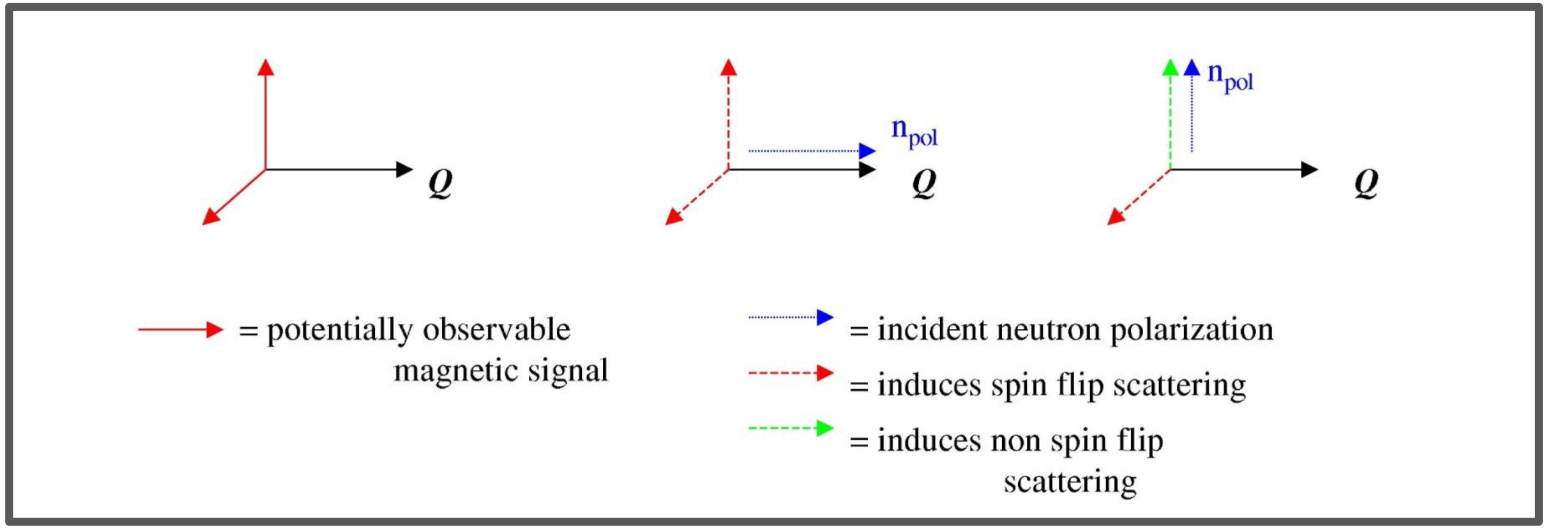
Image credit: ILL

Q // q_y **no additional magnetic signal seen**

Q // q_x **additional magnetic signal seen**

Scattering system has magnetization parallel to q_y





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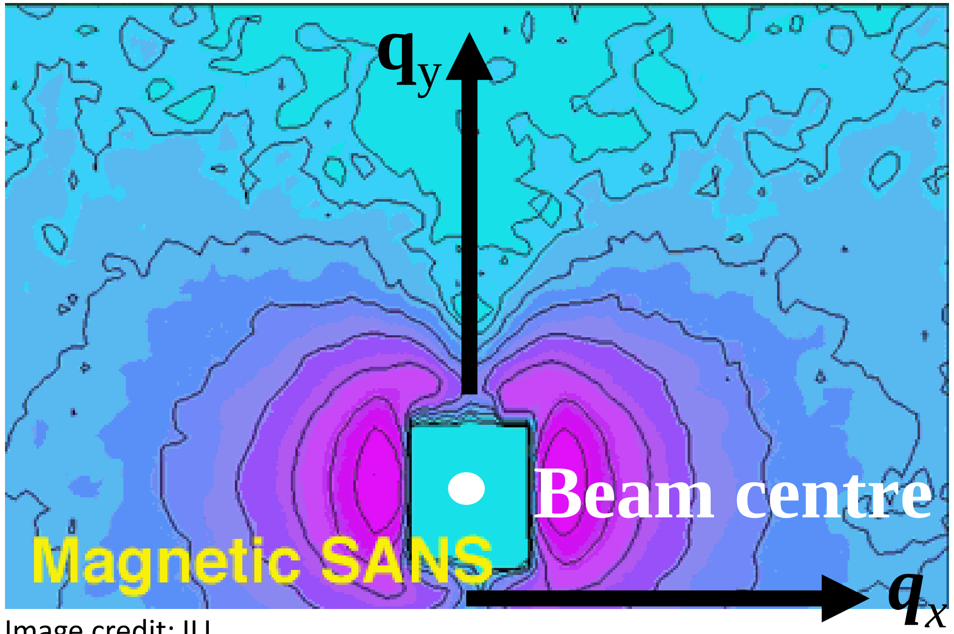

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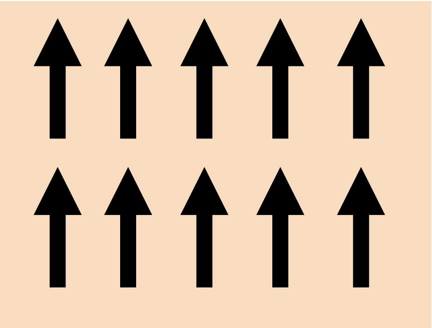

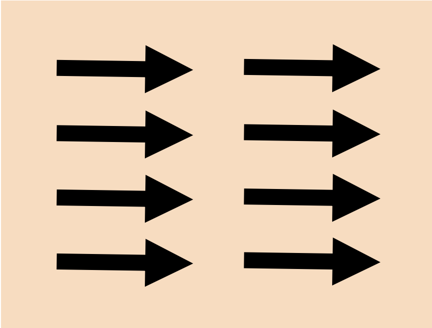

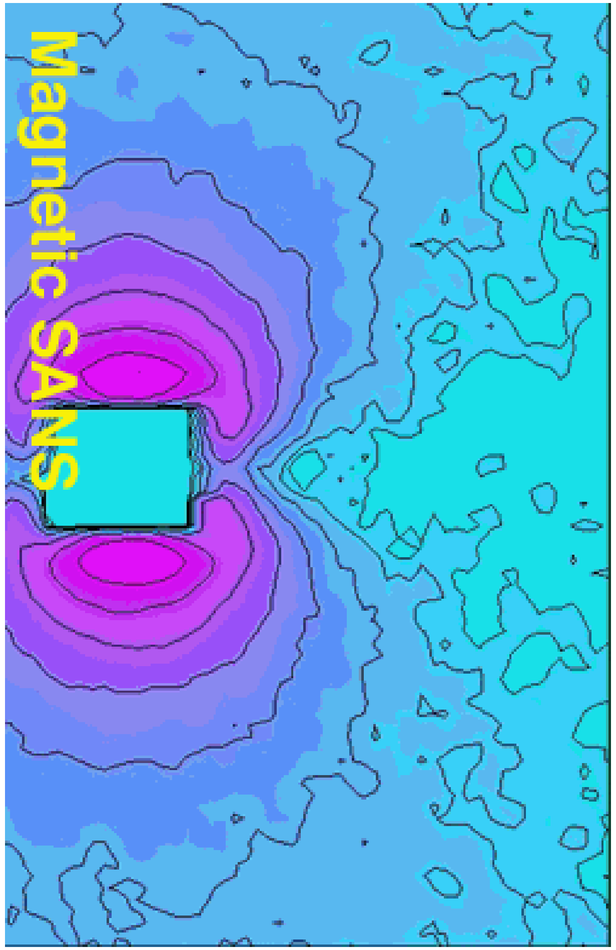

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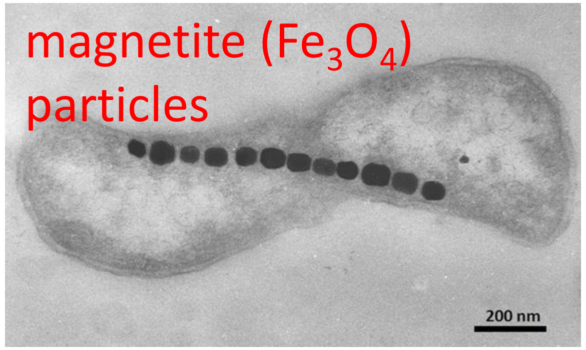
We can use an external field to control the magnetization

Magnetotactic bacteria



Nature Education (2010)

Image: Heraeus

Scattering from a close packed array of magnetite particles

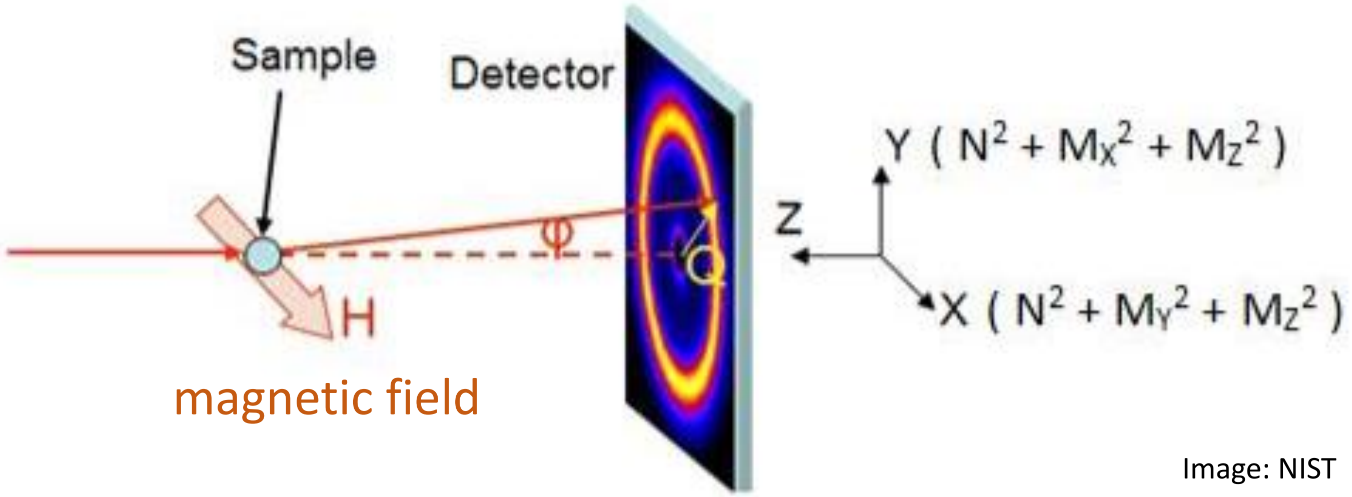


Image: NIST

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

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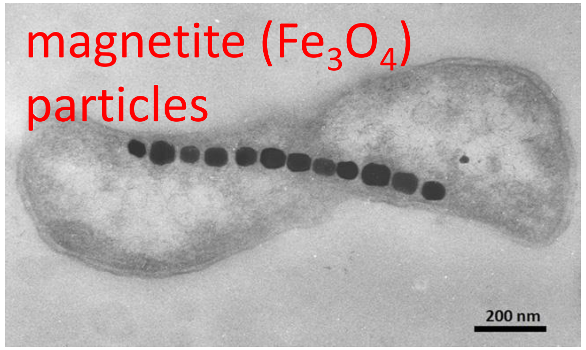

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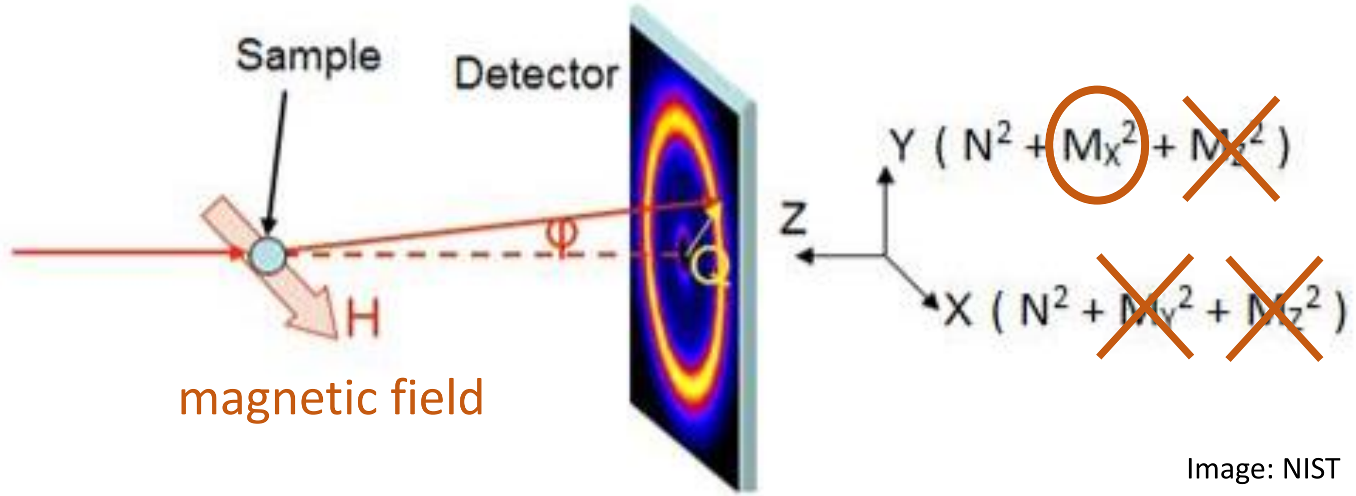

Image: NIST

Image: Heraeus

If the magnetic field applied is large enough, the magnetization of the particles will all point in the same direction, parallel to the field.

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

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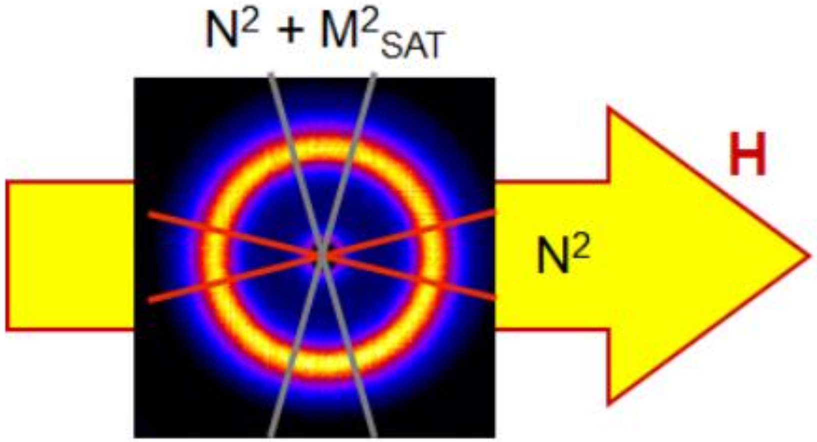
We can use an external field to control the magnetization

Scattering from a close packed array of magnetite particles

NB: magnetic SLD same order of magnitude as nuclear SLD



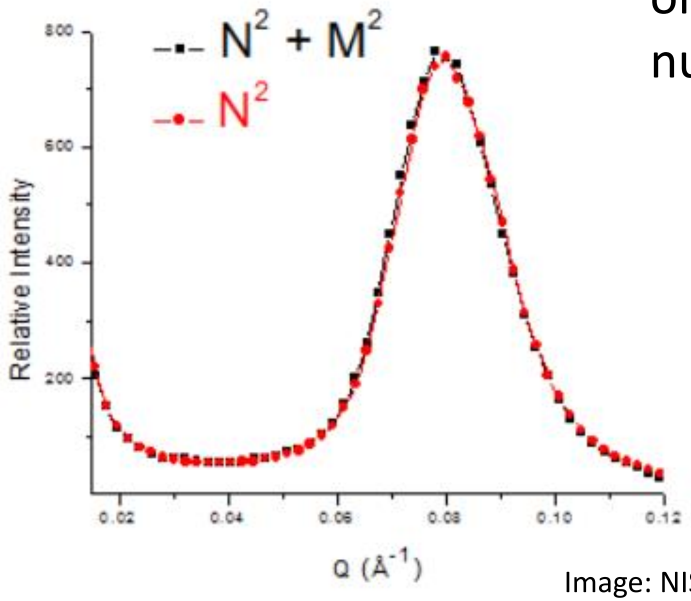


Image: NIST

Material (bulk)	Chemical Formula	SLD_nuclear (Å^{-2})	SLD_magnetic (Å^{-2})
Magnetite	Fe_3O_4	6.97×10^{-6}	1.46×10^{-6}

Nuclear scattering excess = 45.58
Magnetic scattering excess = 2.13

→ Magnetic scattering fraction = 2.13/(45.58+2.13) = 4.5%

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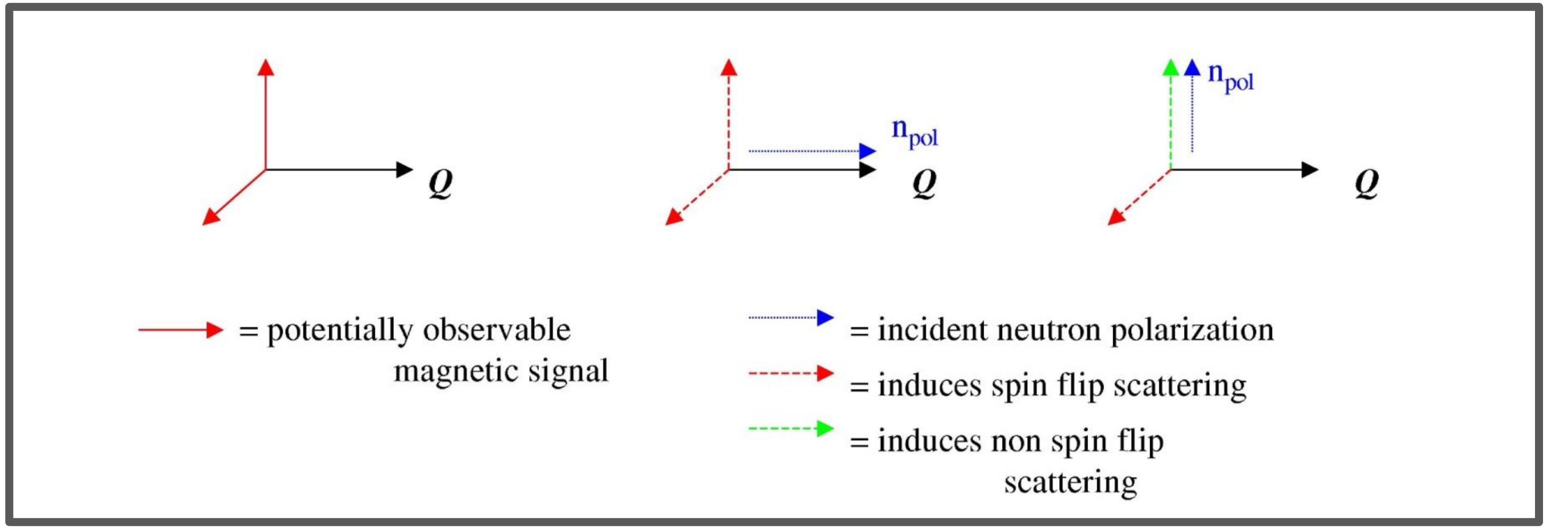
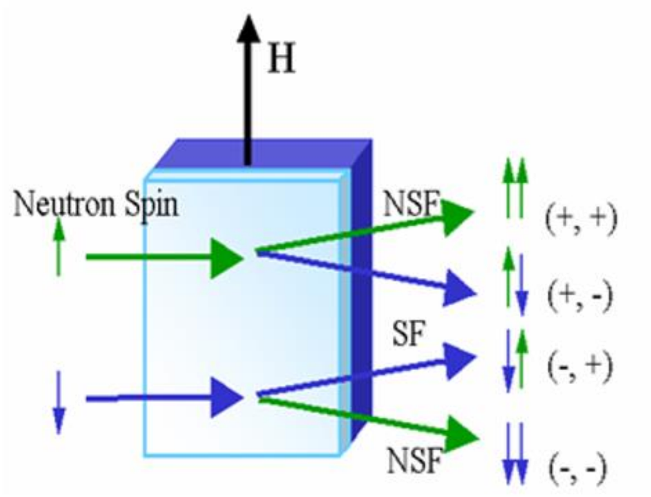
Directing the polarization of the neutrons





Polarizing before the sample:

- We measure with the spins polarised along a given axis, $I+$
- And then measure with the spins polarised in the opposite direction, $I-$.
- We can therefore measure the Flipping Ratio for a given axis, $I+ / I-$

Polarizing before and after the sample:

- We can measure the specific cross-sections for spin-flips or non-spin-flips with the neutrons polarized in a particular direction.
- We can separate out magnetic (spin-flip) from non-magnetic scattering in certain cases.

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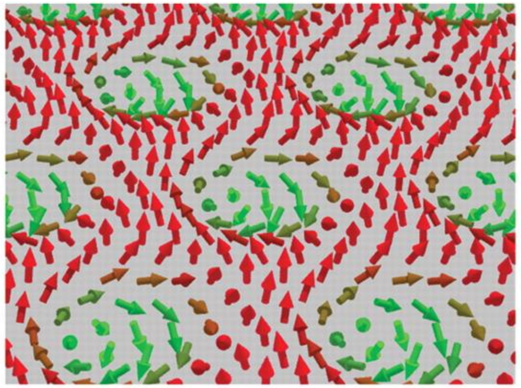
What can we actually study with SANS?

Diffraction at small angles

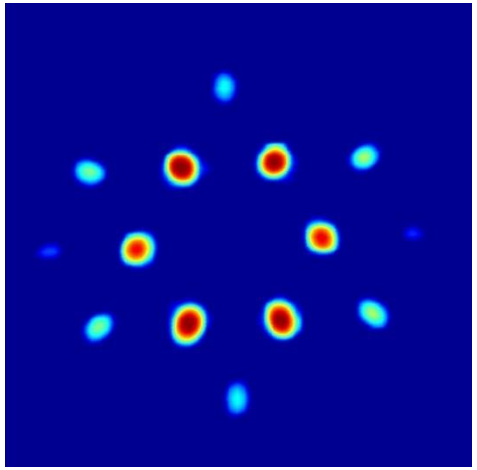
Long lengthscale periodic structures

Skyrmions

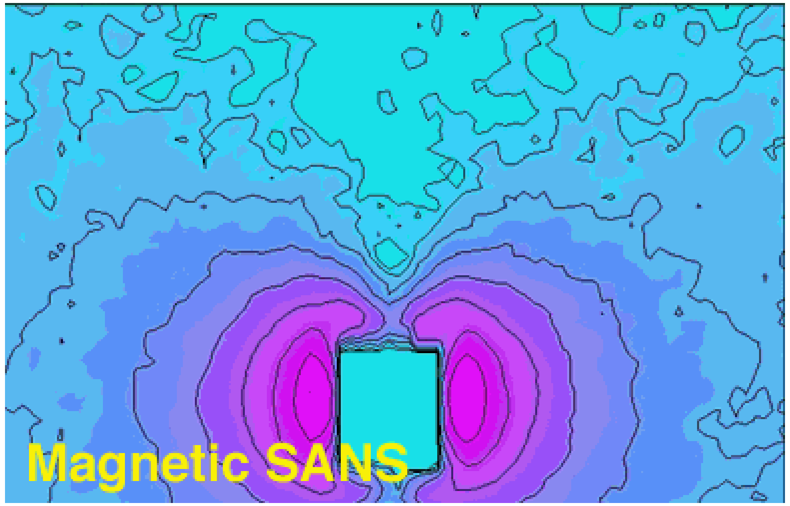


S. Muehlbauer *et al.*, Science **323**, 915 (2009).

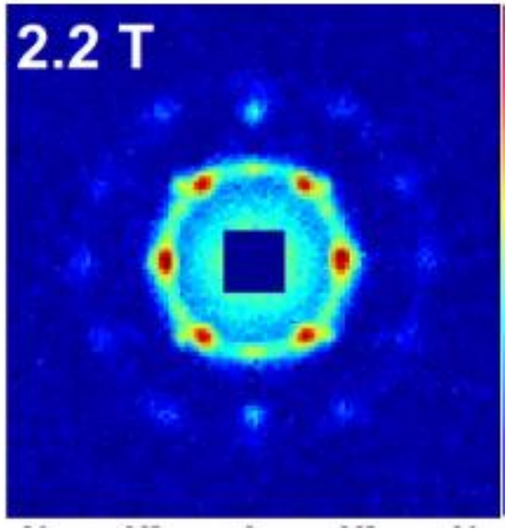
Vortex lattices in superconductors



Diffuse SANS



Fe-O NPs – self assembly



Z. Fu *et al.*, Nanoscale **8**, 18541 (2016)

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What influences the observed magnetic small angle scattering?

Everything you have already heard about in Lectures 2 and 3

- Particle shape (including size)
- Packing of particles
- Polydispersity (level of variations in shape and size)
- Interparticle interactions

And in addition we have to consider:

- Magnetic anisotropy
- Domain walls
- Demagnetization factors
- Magnetic dead layers
- Magnetic interparticle interactions

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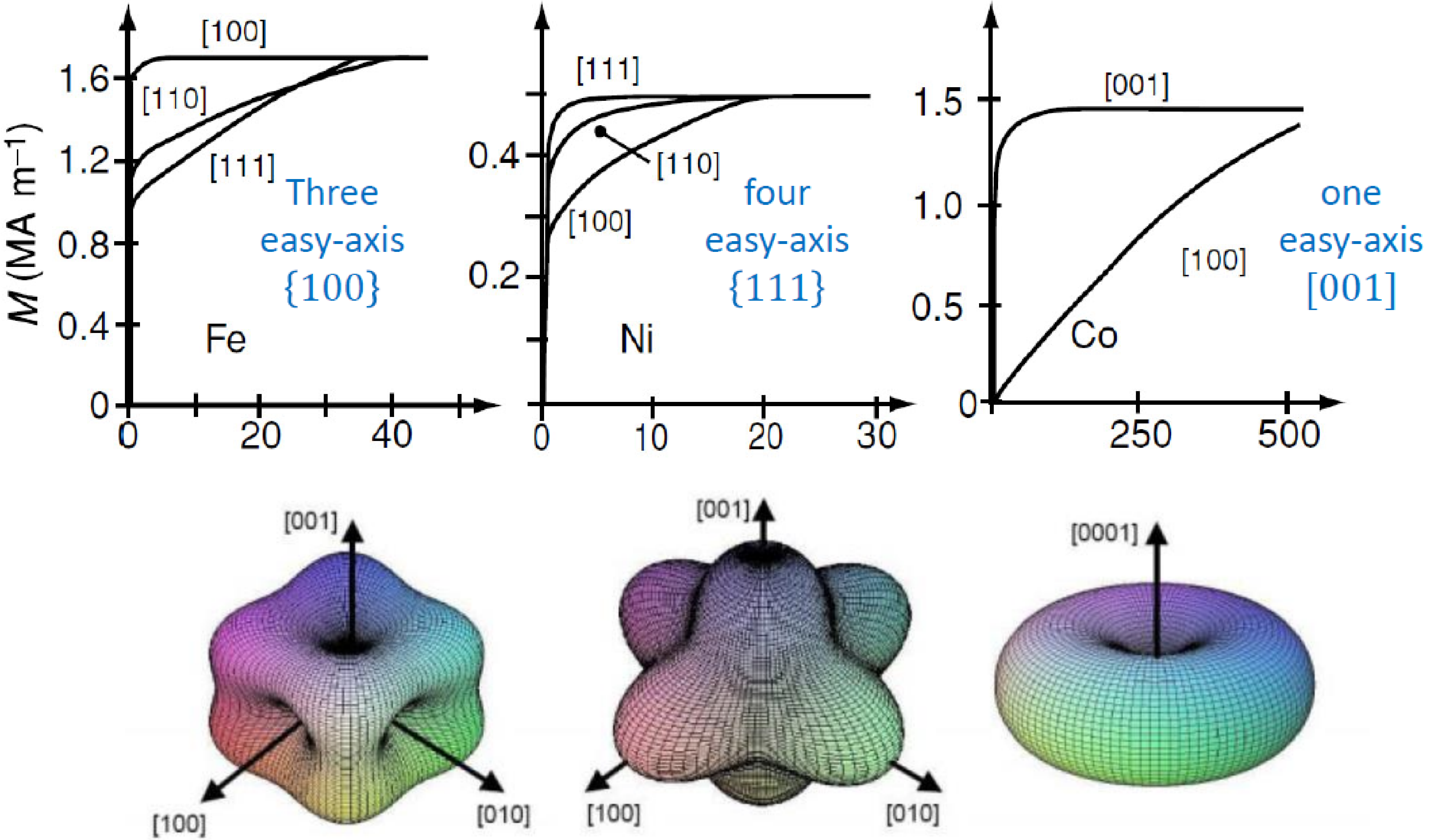
Magnetic anisotropy



http://magnetism.eu/esm/2011/slides/bayreuther-slides.pdf

Domain walls

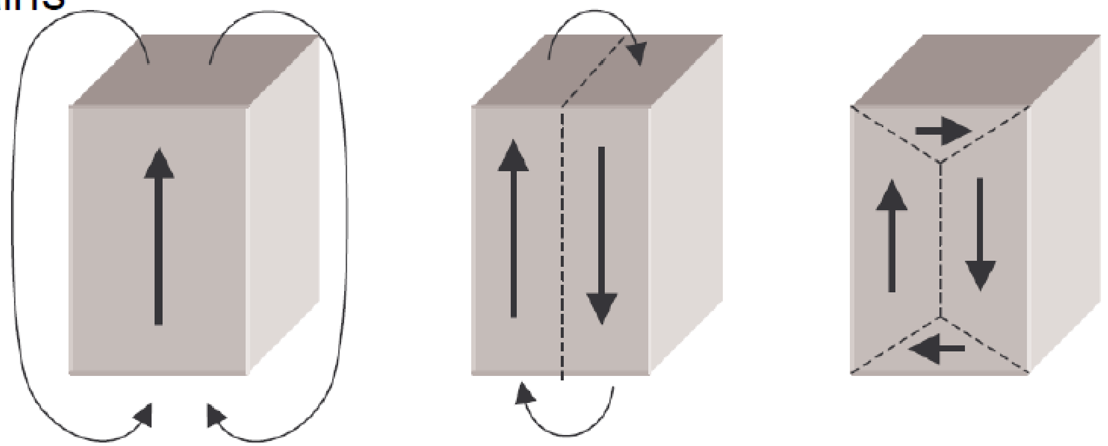
The magnet can minimize its energy by forming domains

Two competing effects

1. Domains minimizes the magnetostatic energy

2. The formation of domain walls costs energy

The domain wall energy $\gamma = 4\sqrt{AK}$ $[\mathrm{energy} \cdot \mathrm{m}^{-2}]$



A = exchange energy
K = anisotropy energy

Image credit: Rasmus Westerström

Domain walls

Hard magnets: Large coercivity B_0 → large resistance towards demagnetization.
Difficult to move domain walls

Soft magnets: Small coercivity B_0 → low resistance towards demagnetization.
Easy to move domain walls

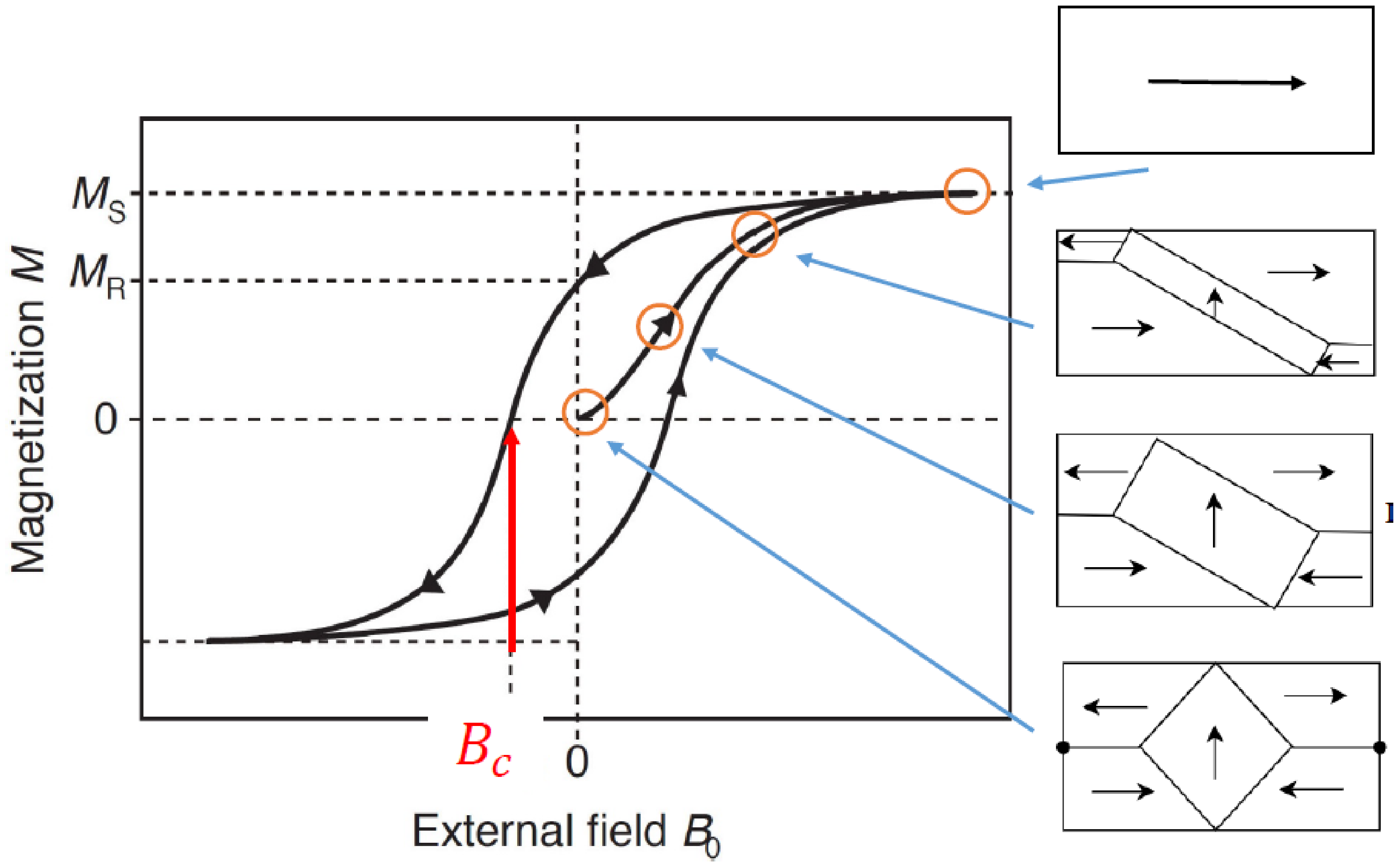
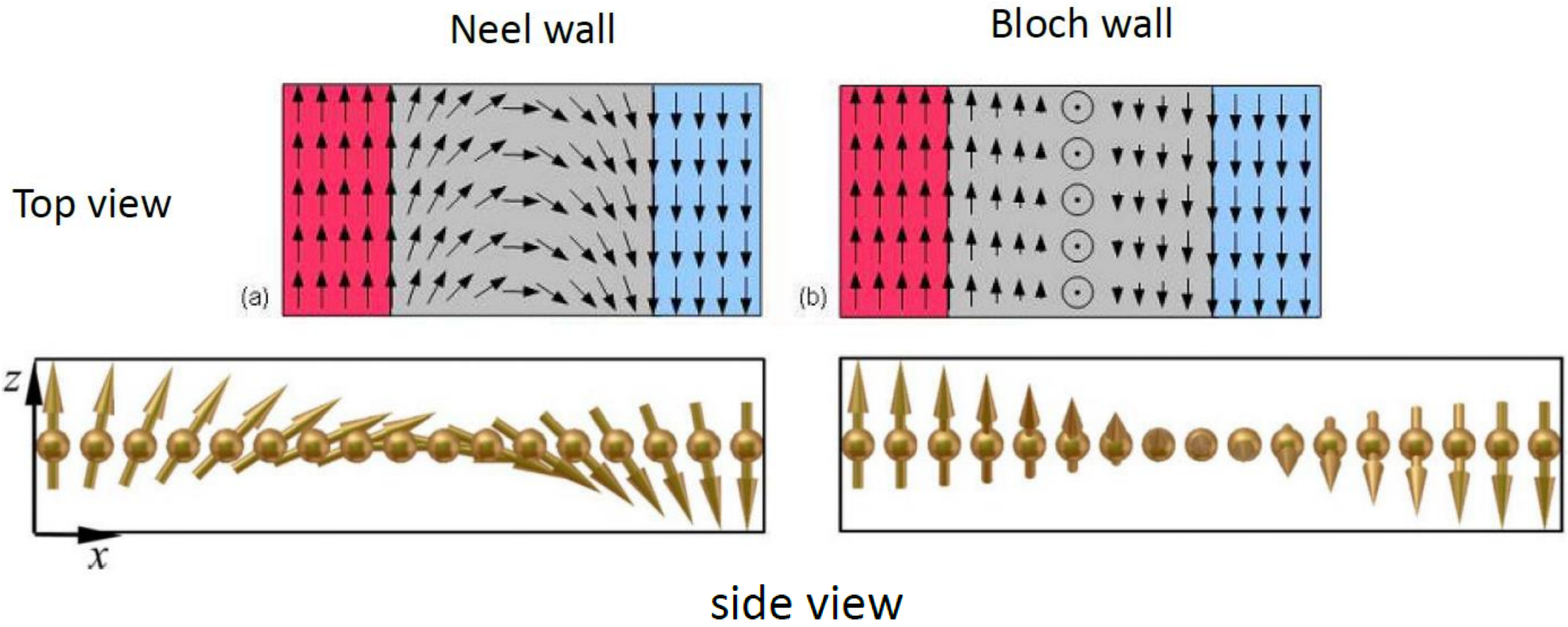


Image credit: Rasmus Westerström

Domain walls



Domain walls are not sharp interfaces – the magnetisation changes over a relatively long distance. This means that the assumptions built into, e.g. Porod's law, regarding sharp interfaces are not always appropriate.

Image credit: Rasmus Westerström

Demagnetization factor

Any uniformly magnetized sample having the form of an ellipsoid also has an uniform demagnetizing field $\boldsymbol{H}_d$ with the relation

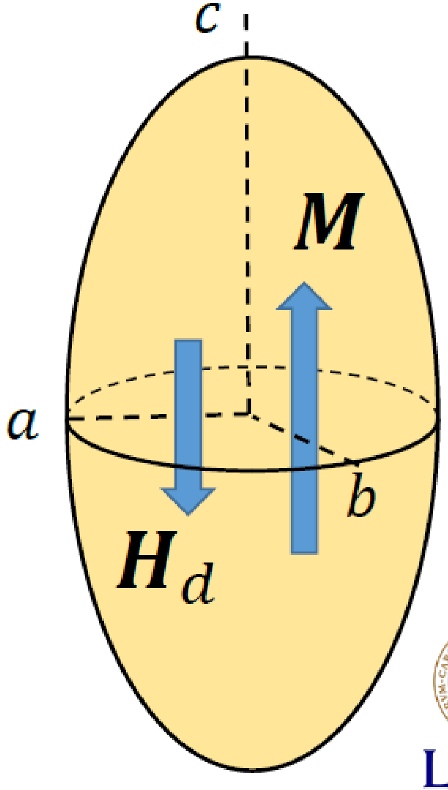
$$\boldsymbol{H}_d = -N_d \boldsymbol{M}$$

Where N_d is the demagnetization tensor. Along the principle axis (a, b, c) of the ellipsoid, $\boldsymbol{H}_d$ and $\boldsymbol{M}$ are collinear and the corresponding principle components (N_a, N_b, N_c) of N_d are called the demagnetization factors with the relationship

$$N_a + N_b + N_c = 1$$

This also approximately true for shapes that are limiting cases of ellipsoid, such as thin films or a wire

Shape	**Magnetization direction**	N_d
Sphere	Any direction	$1/3$
Thin film	In-plane	0
	Normal to the plane	1
General ellipsoid of revolution $a = b \neq c$		$N_c = (1 - 2N_a)$



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What influences the observed magnetic small angle scattering?

Everything you have already heard about in Lectures 2 and 3

- Particle shape (including size)
- Packing of particles
- Polydispersity (level of variations in shape and size)
- Interparticle interactions

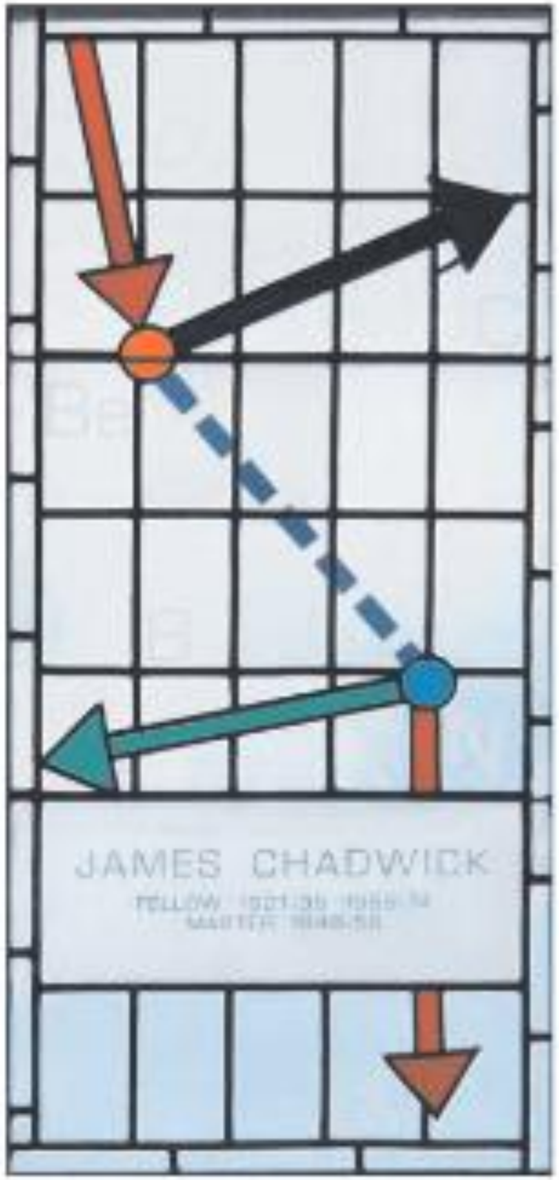
And in addition we have to consider:

- Magnetic anisotropy
- Domain walls
- Demagnetization factors
- Magnetic dead layers
- Magnetic interparticle interactions

Summary

- The difference between nuclear and magnetic scattering of neutrons
- What does that mean for our experiments?
- How can we optimize our experiments to take advantage of this?
- What do we need to account for when measuring magnetic materials?

$${}^{4}_{2}\alpha + {}^{9}_{4}\mathrm{Be} \rightarrow {}^{12}_{6}\mathrm{C} + {}^{1}_{0}\mathrm{n}$$



Useful references

Introduction to the Theory of Thermal Neutron Scattering, G. L. Squires, Dover (1996).

Theory of Neutron Scattering from Condensed Matter (2 volumes), S. W. Lovesey, Clarendon Press (1984).

Neutron Data Booklet: https://www.ill.eu/fileadmin/user_upload/ILL/1_About_ILL/Documentation/NeutronDataBooklet.pdf

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Magnetic small-angle neutron scattering

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