Elastic polarized neutron scattering: theory and practice

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Aims

1. Review some basic concepts in polarised neutron scattering
2. Give an overview of polarised neutron instrumentation.
3. Discuss some applications of polarised neutron diffraction, particularly for magnetic systems (the dark side of PA)

Outline

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1. Introduction and motivation
   - Pauli matrices, polarization and cross sections
   - The scattering potential and the rules of polarised neutron scattering
   - Example: paramagnetic scattering
2. Components of a polarized neutron instruments
   - Polarizers and analysers
   - Guide fields and flippers
3. Polarized elastic scattering
   - The Blume-Maleev equations
   - Longitudinal polarisation analysis (polarimetry if time allows)

Neutron spin

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The neutron is an S = 1/2 particle, like the electron. Its spin degree of freedom is represented by the spin angular momentum operator:

$$\boxed{\mathbf{S} = \frac{\hbar}{2}\sigma}$$

where σ is a vector of the three **Pauli matrices**:

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

These have eigenvalues of ±1, meaning the spin can take on a value of ±1/2h. The **expectation value** $\langle\sigma\rangle$ for a given spin wave-function is called the **spin direction**, and may be considered the direction in which the spin points.

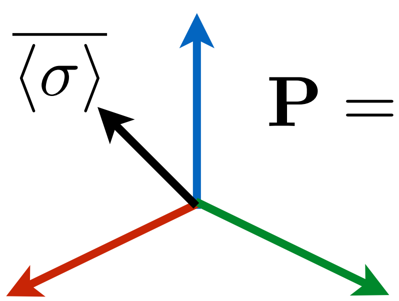
We will label the corresponding eigenstates $|+_\alpha\rangle$ and $|-_\alpha\rangle$, where α represents the spin direction.

Polarized neutron beams

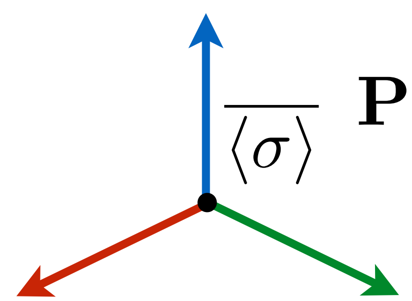
For an ensemble (beam) of neutrons, the **polarization** is defined as:

$$\boxed{\mathbf{P} = (2/\hbar)\overline{\langle \mathbf{S} \rangle} = \overline{\langle \sigma \rangle}}$$

where the right hand side is the mean expectation value of the spin direction

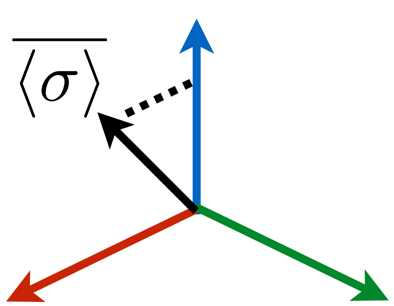


$$\mathbf{P} = \left(\frac{1}{2}, 0, \frac{\sqrt{3}}{2} \right)$$

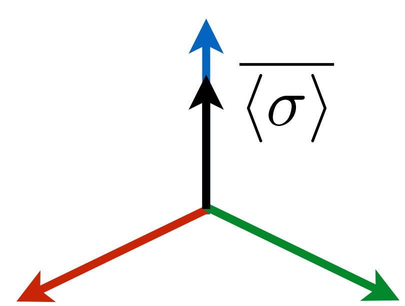


$$\mathbf{P} = (0, 0, 0)$$

In the presence of a magnetic field, the polarization is defined as the projection of **P** onto the field axis (the components perpendicular to the field precess)



$\xrightarrow{\mathbf{H} \parallel z}$



$$\boxed{P = \frac{N_+ - N_-}{N_+ + N_-}}$$

Basic results from unpolarised scattering

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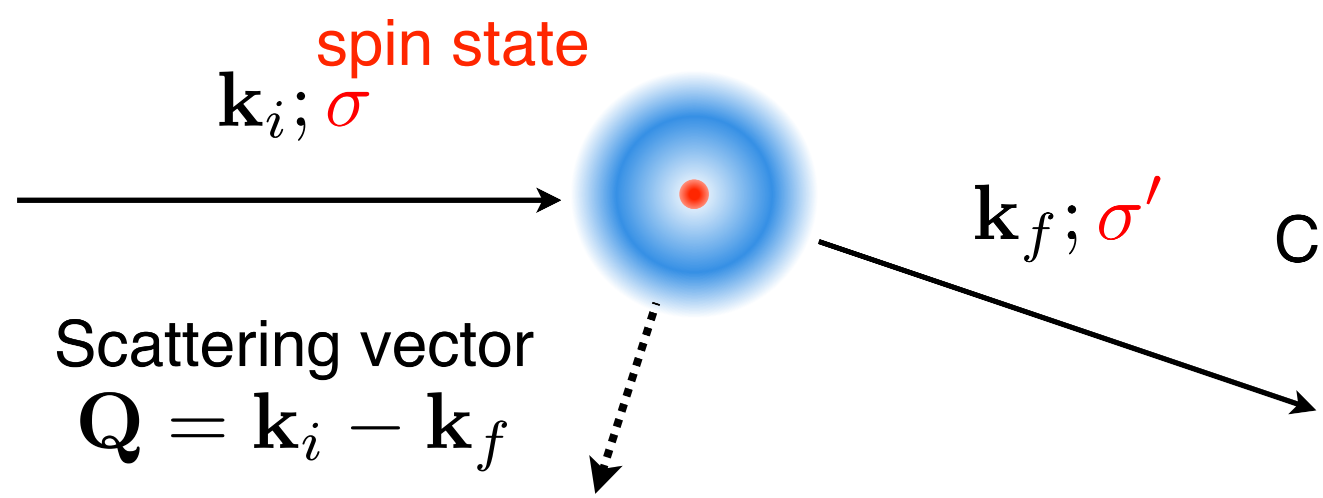


Cross section

$$\frac{d\sigma}{d\Omega}$$

Magnetic

Magnetic scattering length

$$\propto \hat{\mathbf{Q}} \times \mathbf{M}(\mathbf{Q}) \times \hat{\mathbf{Q}}$$

(components of **M** $\perp$ **Q**)

Magnetic form factor

$$\rho_s(\mathbf{r}) \xrightarrow{\mathrm{FT}} f(\mathbf{Q})$$

Nuclear

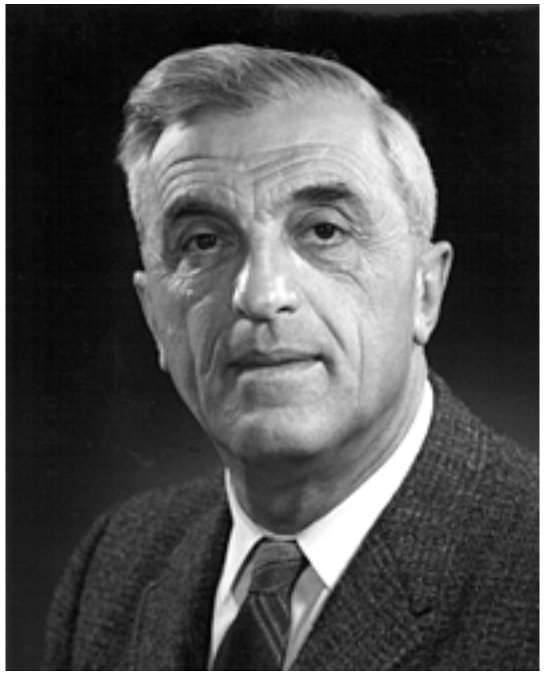
Scattering length depends on nuclear spin and isotope

Nuclear coherent cross section
- mean scattering length $(\bar{b})^2$

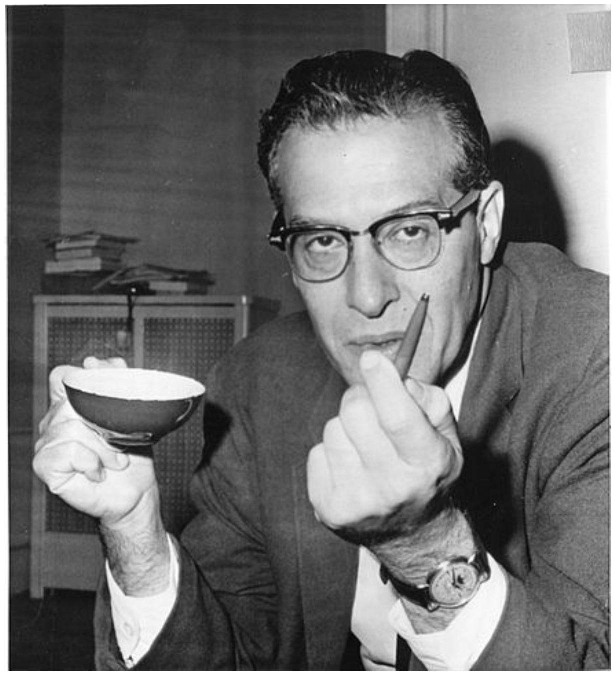
Incoherent cross section
- variance in scatt. length $\overline{b^2} - (\bar{b})^2$

What effect do these scattering processes have on the polarization and vice versa?

Historical digression



Felix Bloch



Julian Schwinger

Otto Halpern

All the ingredients for polarised neutrons were present 7 years after the discovery of the neutron, and 5 years before the first neutron scattering experiments

MAY 15, 1939 PHYSICAL REVIEW VOLUME 55

On the Magnetic Scattering of Neutrons

O. HALPERN AND M. H. JOHNSON
New York University, University Heights, New York, New York

(Received December 3, 1938)

The scattering potential

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Reminder: the scattering cross section is given by the First Born Approximation/ Fermi's golden rule as the square of the matrix element:

$$\left(\frac{d\sigma}{d\Omega}\right) \propto \left|\langle \sigma' \lambda' \mathbf{k}_f | V(\mathbf{r}) | \sigma \lambda \mathbf{k}_i \rangle\right|^2 \longrightarrow \left(\frac{d\sigma}{d\Omega}\right) \propto \left|\langle \sigma' | V(\mathbf{Q}) | \sigma \rangle\right|^2$$

neutron spin state

scattering system states

Fourier transform potential
Elastic scattering only

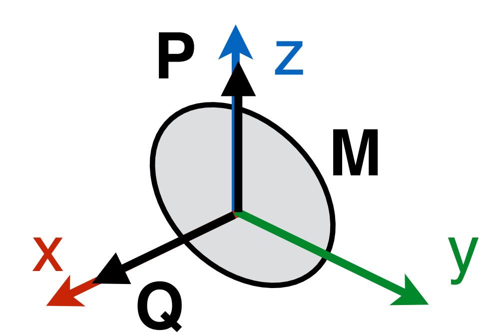
How does the interaction with the neutron spin state enter the potential?

$$V_m(\mathbf{Q}) = \boxed{\sigma \cdot \hat{\mathbf{Q}} \times \mathbf{M}(\mathbf{Q}) \times \hat{\mathbf{Q}}} \qquad V_n(\mathbf{Q}) = A(\mathbf{Q}) + \boxed{B(\mathbf{Q})\mathbf{I} \cdot \sigma}$$

e.g. **M(Q)** ⊥ **Q** || **x** (*i.e.* in the yz plane), **S** || **z**

$\sigma_z M_{\perp z} |+_z\rangle = M_{\perp z} |+_z\rangle$ **S unchanged**

$\sigma_y M_{\perp y} |+_z\rangle = i M_{\perp y} |-_z\rangle$ **S flipped**



Rules of polarized neutron scattering

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For the moment, only consider processes like these, that either leave the neutron spin (beam polarization) unchanged, or flip it by π, *i.e.*:

Non spin-flip $|\sigma\rangle \rightarrow |\sigma\rangle$ $\left\{ \begin{array}{l} |+\rangle \rightarrow |+\rangle \\ |-\rangle \rightarrow |-\rangle \end{array} \right.$

$\left. \begin{array}{l} |+\rangle \rightarrow |-\rangle \\ |-\rangle \rightarrow |+\rangle \end{array} \right\}$ **Spin-flip** $|\sigma\rangle \rightarrow |-\sigma\rangle$

Magnetic

Components of **M** perpendicular to the polarization:

$$|\sigma\rangle \rightarrow |-\sigma\rangle \quad \textbf{SF}$$

Components parallel:

$$|\sigma\rangle \rightarrow |\sigma\rangle \quad \textbf{NSF}$$

Nuclear

Nuclear coherent scattering and isotope incoherent scattering:

$$|\sigma\rangle \rightarrow |\sigma\rangle \quad \textbf{NSF}$$

Spin-incoherent scattering:

$$\frac{1}{3}(|\sigma\rangle \rightarrow |\sigma\rangle), \quad \frac{2}{3}(|\sigma\rangle \rightarrow |-\sigma\rangle)$$

The scattered polarization is different! We will look at these in more detail later.

Example: paramagnet

Generally, magnetic scattering shows a complex dependence on the angles between **M**, **P**, and **Q**. In a paramagnet, the situation is much simpler.

The cross sections corresponding to **NSF** and **SF** scattering are:

$$\left(\frac{d\sigma}{d\Omega}\right)^{\alpha}_{\mathrm{NSF}} = C[1-(\hat{\mathbf{P}}\cdot\hat{\mathbf{Q}})^2] \qquad \left(\frac{d\sigma}{d\Omega}\right)^{\alpha}_{\mathrm{SF}} = C[1+(\hat{\mathbf{P}}\cdot\hat{\mathbf{Q}})^2]$$

Using the definition of scalar polarization $P = (N_+ - N_-)/(N_+ + N_-)$

Halpern-Johnson equation

$$\mathbf{P}' = \hat{\mathbf{Q}}[\mathbf{P}\cdot\hat{\mathbf{Q}}]$$

This means that if **P** ll **Q**, all the magnetic **NSF** scattering vanishes, and the magnetic scattering should appear in the **SF** channel only.

What do we need to measure this?

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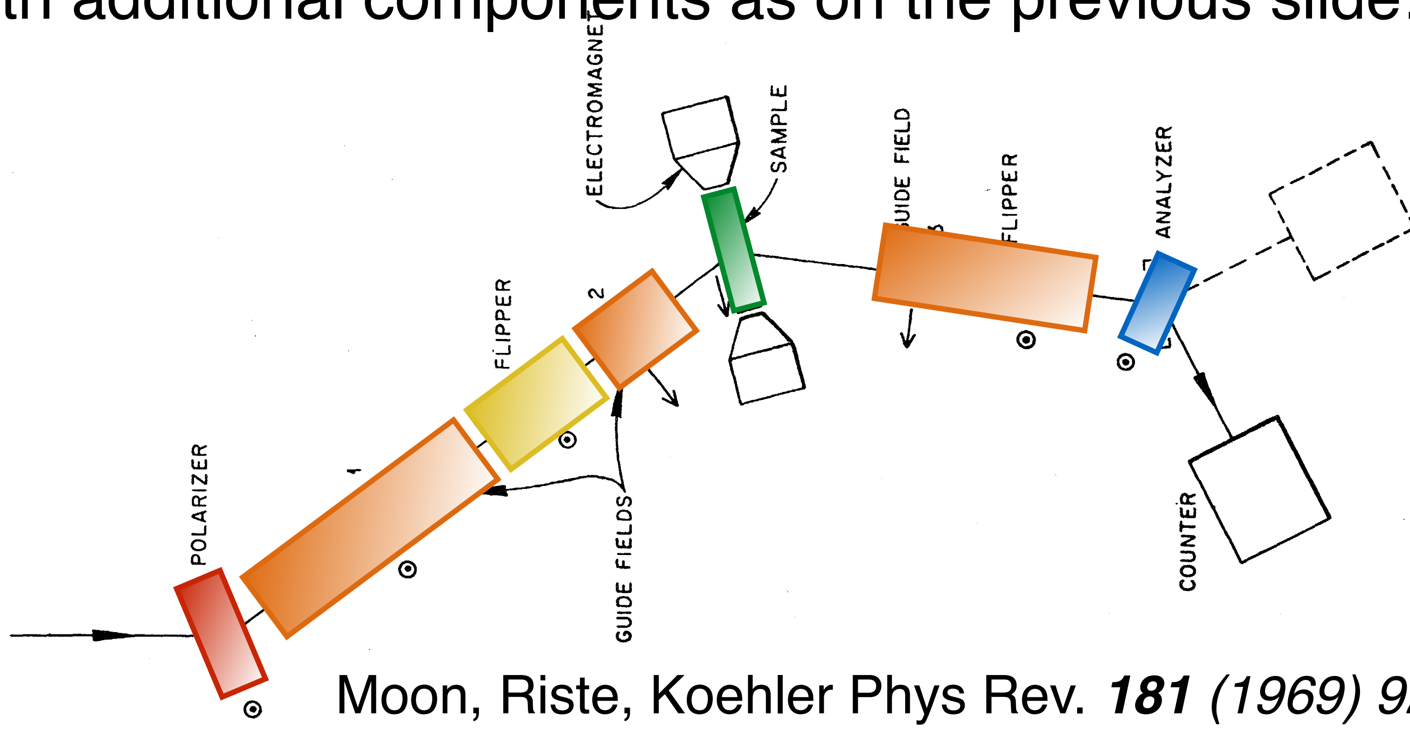
All we need to do to verify this is to measure the **SF** cross section with **P** ll **Q**. How do we achieve this? In practise, need 5 different components along the instrument:

Component	Beam	Box
1. **Polarizer**; select single polarization from unpolarised incident beam $\sqrt{2}^{-1}(\vert+\rangle + \vert-\rangle)$	$\vert+\rangle$	P
2. **Guide fields** to both maintain **P** and rotate it ll **Q**	$\vert+_{\vert\vert\mathbf{Q}}\rangle$	G
3. A **flipper**, to change the orientation	$\vert-_{\vert\vert\mathbf{Q}}\rangle$	F
4. A paramagnetic sample (magnetic scatt. **SF**)	$\vert+_{\vert\vert\mathbf{Q}}\rangle$	S
2. **Guide fields** to rotate **P** back to original direction	$\vert+\rangle$	G
5. **Analyzer**; selects a single polarization from scattered beam	$\vert+\rangle$	A

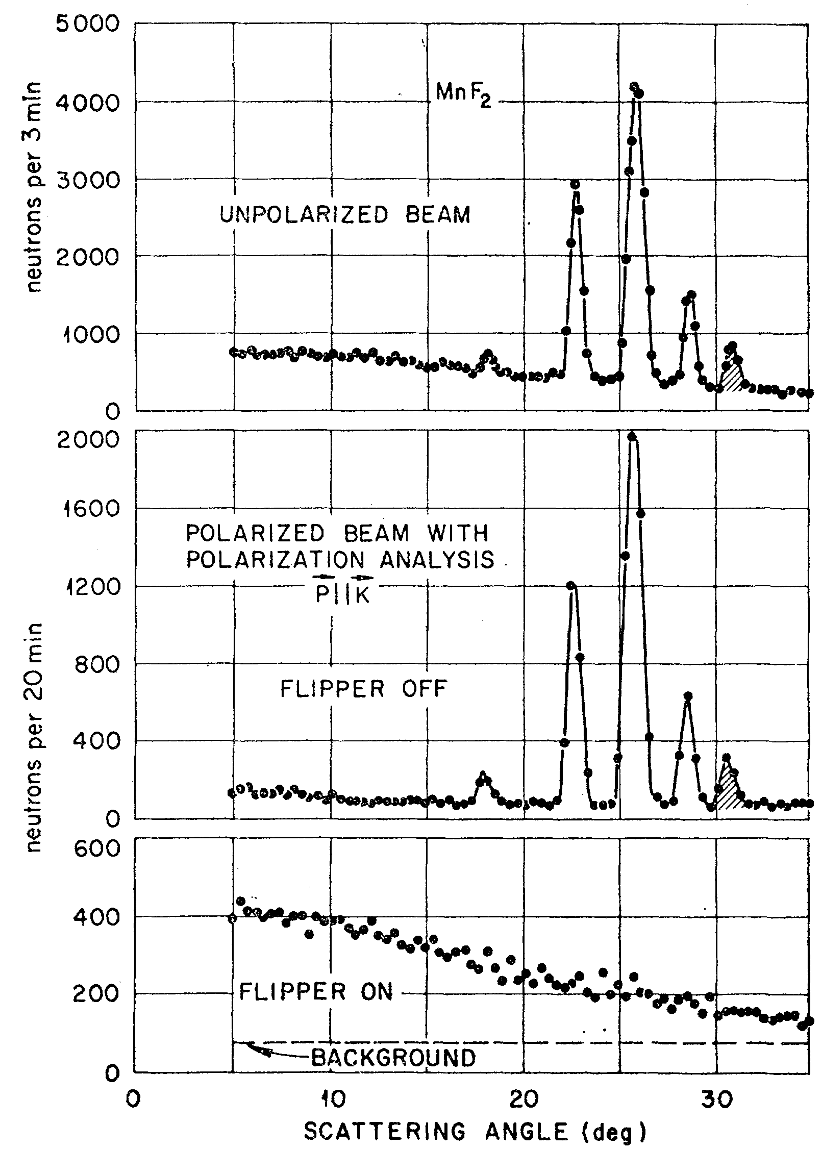
Paramagnetic scattering: experiment

This experiment was first performed by Moon, Riste, and Koehler in 1968

The instrument used was a triple-axis spectrometer with additional components as on the previous slide:



Moon, Riste, Koehler Phys Rev. ***181*** *(1969) 920*



Flipper off

$$|+\rangle \rightarrow |+\rangle$$

Flipper on

$$|-\rangle \rightarrow |+\rangle$$

Before into more detail on the full separation of cross section components, let's have a look at the various polarised elements

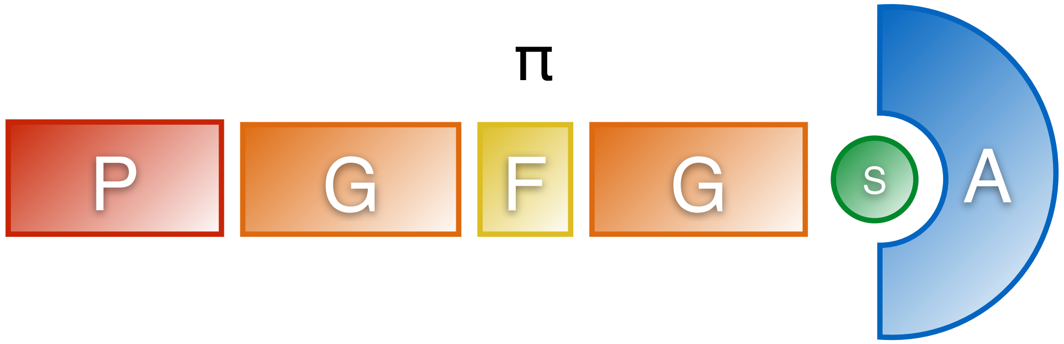
The elements of a polarized beamline

Layout of a polarized instrument

As shown before, polarised instruments consist of some combination of:

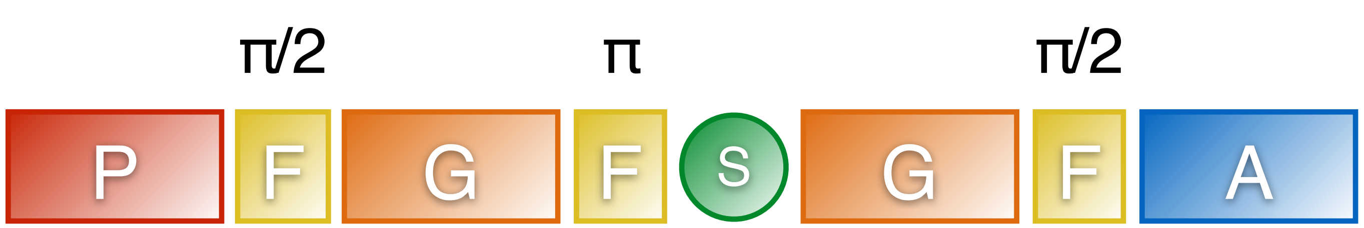
Polarizer (Guide) Field Flipper Analyzer

e.g. wide-angle diffractometer



e.g. reflectometer

e.g. spin-echo



precession

Polarizing/analyzing the beam

Historically, the first means of polarising/analyzing a neutron beam was by Bragg reflection from a ferromagnetic crystal. The coherent elastic cross section is:

$$\left(\frac{d\sigma}{d\Omega}\right) \propto \left| F_N - \langle \psi' | \sigma \cdot \hat{\mathbf{Q}} \times \mathbf{M}(\mathbf{Q}) \times \hat{\mathbf{Q}} | \psi \rangle \right|^2$$

If **M** ⊥ **Q**, **M** || **B** || **z**, and for a single magnetic domain:

$$\left(\frac{d\sigma}{d\Omega}\right)_{++} \propto |F_N - \langle +_z | \sigma_z M_z(\mathbf{Q}) | +_z \rangle|^2 = |F_N - F_M|^2$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{--} \propto |F_N - \langle -_z | \sigma_z M_z(\mathbf{Q}) | -_z \rangle|^2 = |F_N + F_M|^2$$

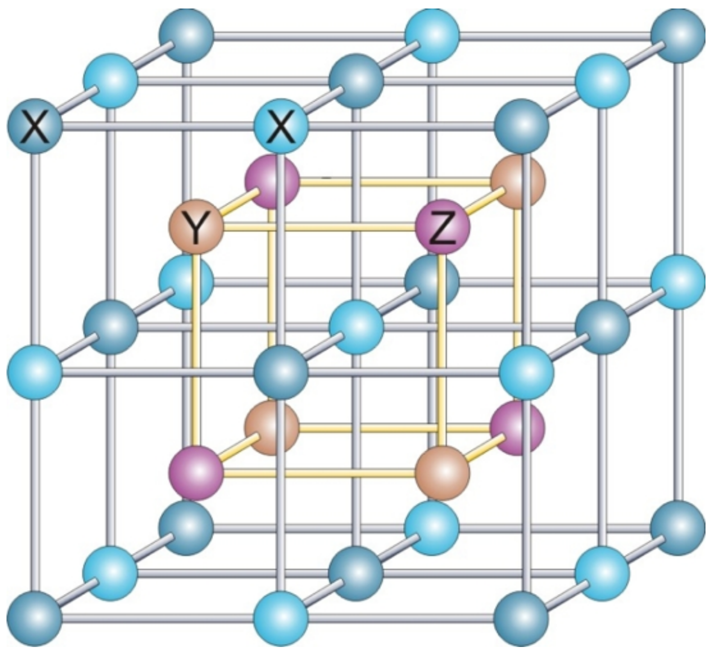
All the **SF** cross sections are zero. We can quickly see that a polarised beam will result if $F_N = F_M$. The polarising/analyzing efficiency is:

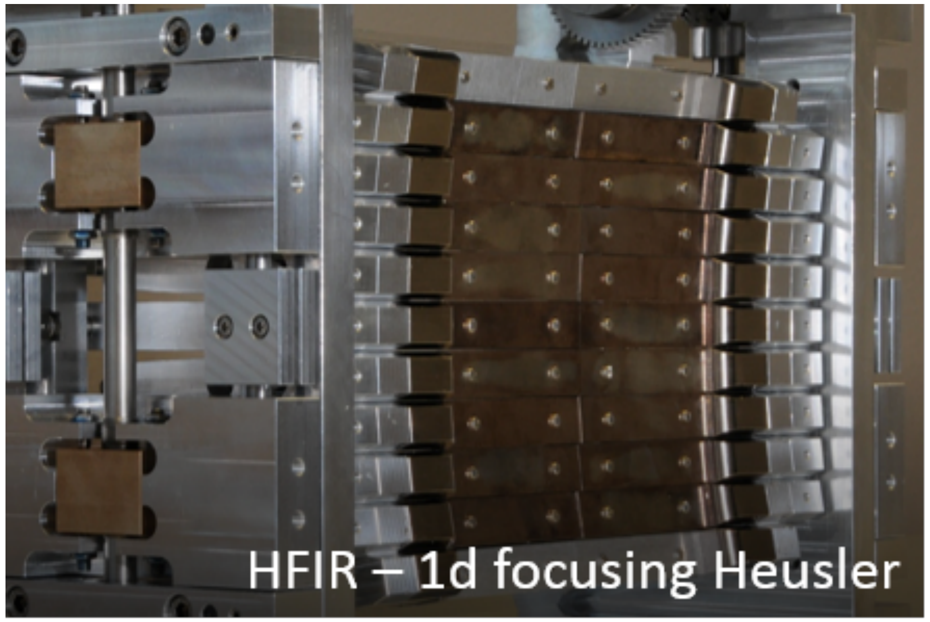
$$P = \frac{(d\sigma/d\Omega)_{--} - (d\sigma/d\Omega)_{++}}{(d\sigma/d\Omega)_{--} + (d\sigma/d\Omega)_{++}} = \frac{|F_N + F_M|^2 - |F_N - F_M|^2}{|F_N + F_M|^2 + |F_N - F_M|^2}$$

Example: Heusler crystal

e.g. Cu_2MnAl Heusler alloy

Al @ (0,0,0), Mn @ (1/2,1/2,1/2) — (111) reflection structure factors:





$$F_N^{2n+1} = b_{\mathrm{Al}} - b_{\mathrm{Mn}} = 7.179 \text{ fm} \qquad F_M^{2n+1} = \frac{1}{2}\gamma_n r_0 g f(Q)\langle M_z \rangle = 6.740 \text{ fm}$$

$$P = \frac{(d\sigma/d\Omega)_{--} - (d\sigma/d\Omega)_{++}}{(d\sigma/d\Omega)_{--} + (d\sigma/d\Omega)_{++}} = \frac{|F_N + F_M|^2 - |F_N - F_M|^2}{|F_N + F_M|^2 + |F_N - F_M|^2} \sim 95\%$$

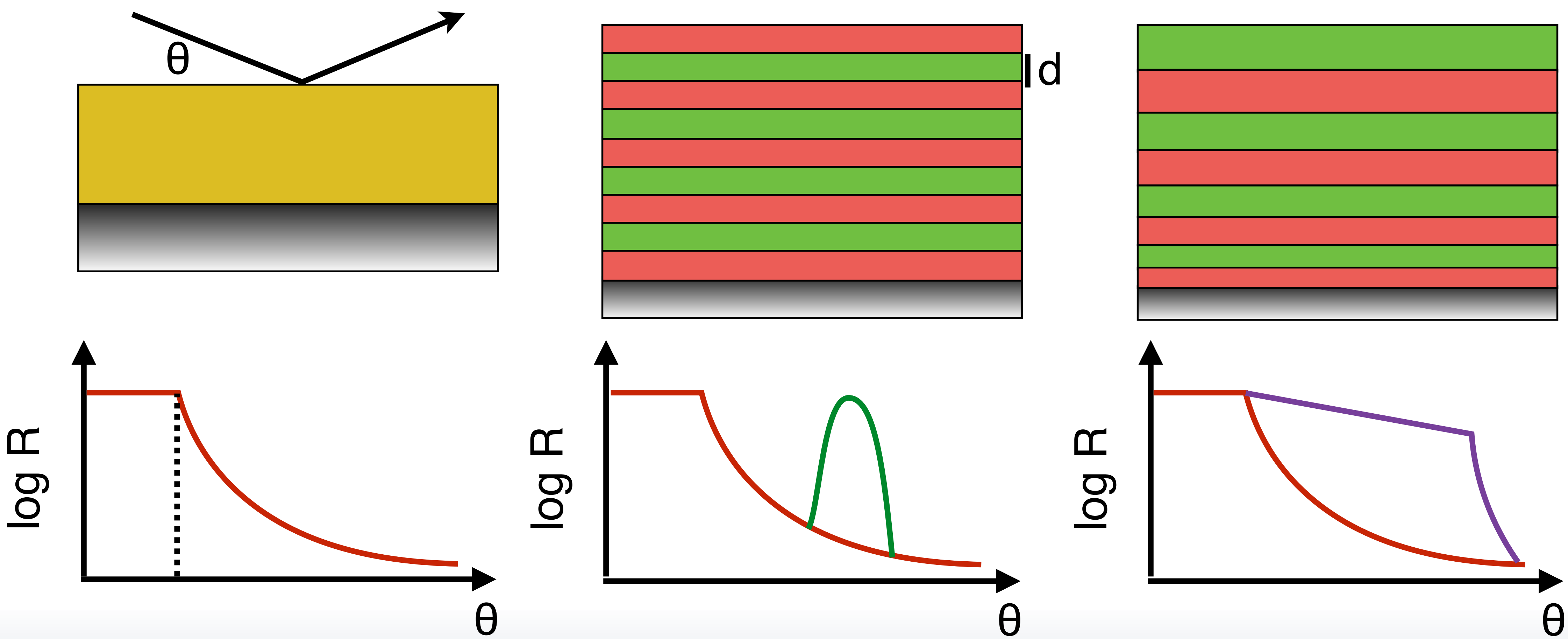
+ stable, monochromates beam, high P **—** monochromatic, range ~0.8Å — 6.5 Å

Supermirrors I

A major development in neutron polarization was the discovery of supermirrors in the mid 1970's. Mezei, Commun. Phys. **1** (1976) 81

$$n_{\pm} \simeq 1 - \frac{\langle V_N \rangle \mp \langle V_M \rangle}{2E_k} = 1 - \left(\frac{N\lambda^2}{2\pi} \right) (\bar{b} \mp \bar{p}) \longrightarrow \theta_{c\pm} = \lambda \left[\frac{N}{\pi} (\bar{b} \pm \bar{p}) \right]$$

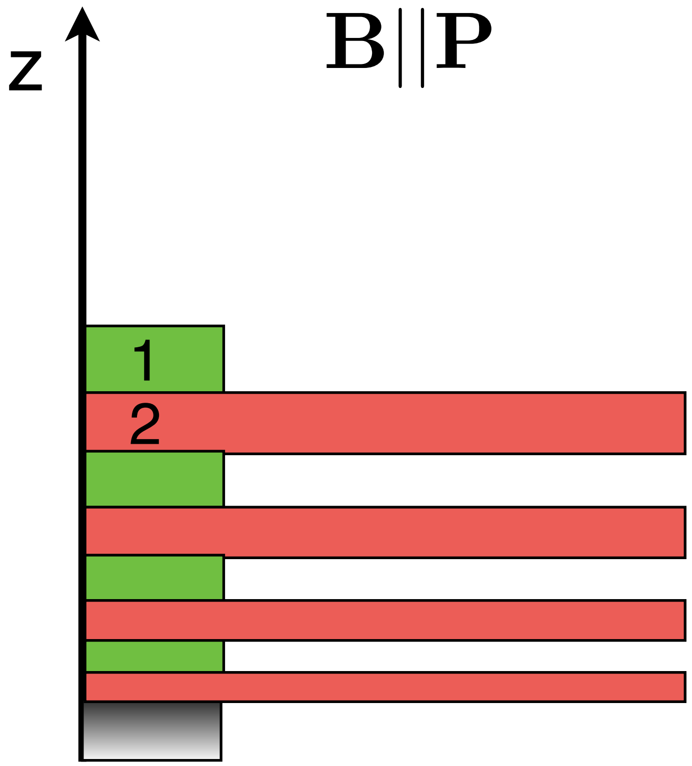
For a single state or unpolarised neutrons:



Supermirrors II

As for the single crystal ferromagnet, the supermirrors may be made polarising if the nuclear and magnetic scattering lengths are matched:

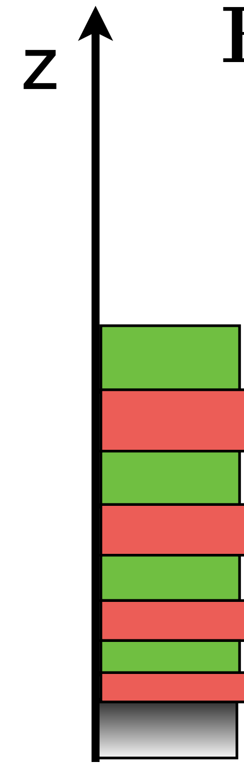
$$\mathbf{B}||\mathbf{P}$$



$$(\bar{b}+\bar{p})$$

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2}\right)^2$$

$$\mathbf{B}|| - \mathbf{P}$$



$$(\bar{b}-\bar{p})$$

For the polarization direction on the right, the interfaces are nearly invisible.

+ stable, broad-band, high P, large divergences **—** cutoff below ~2Å

Examples: supermirror devices

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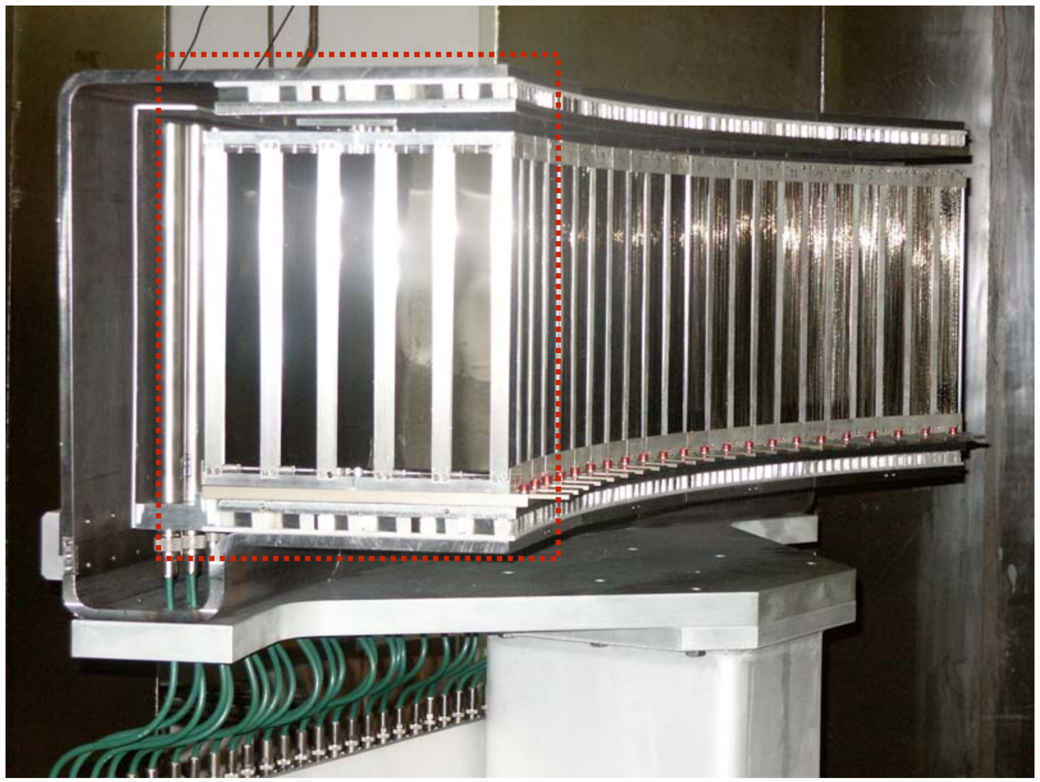
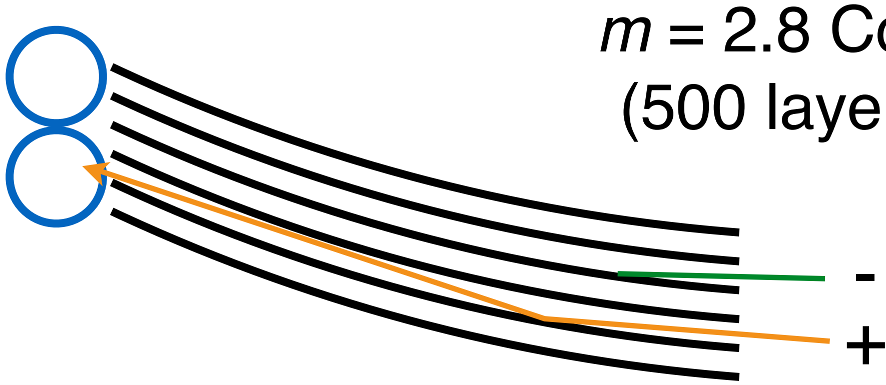
Bender (D7, ILL)

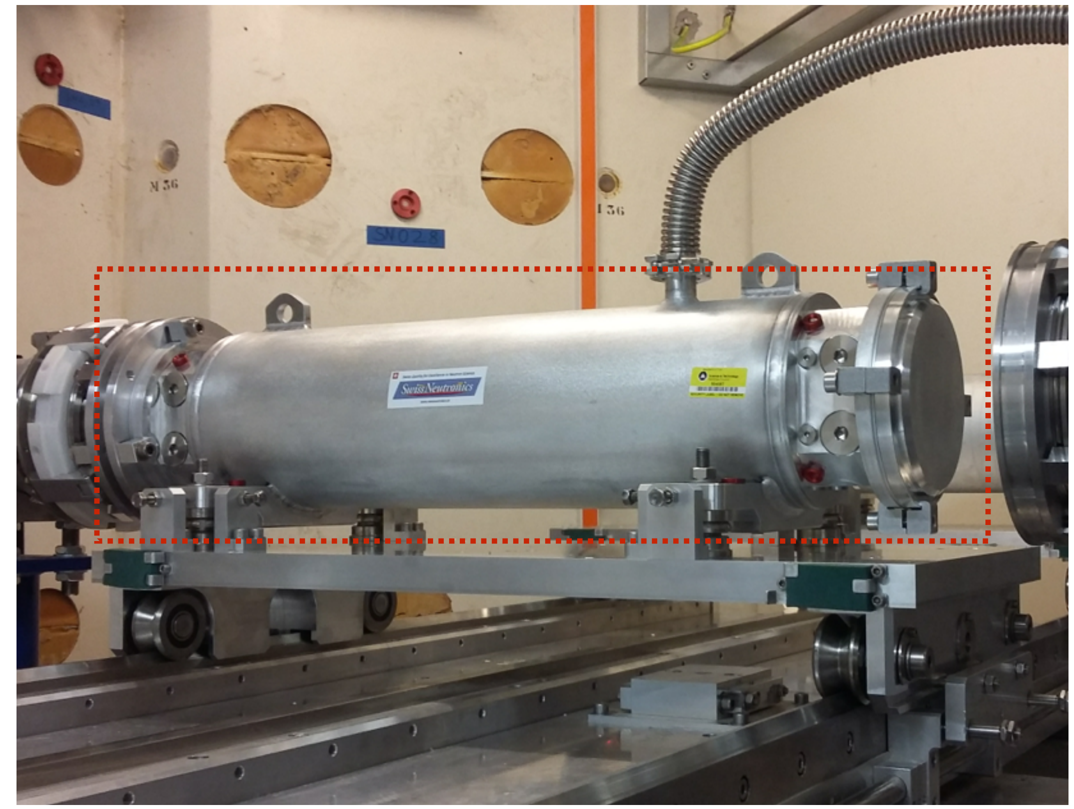


reflects desired spin state

m = 2.8 Co/Ti
(500 layers)

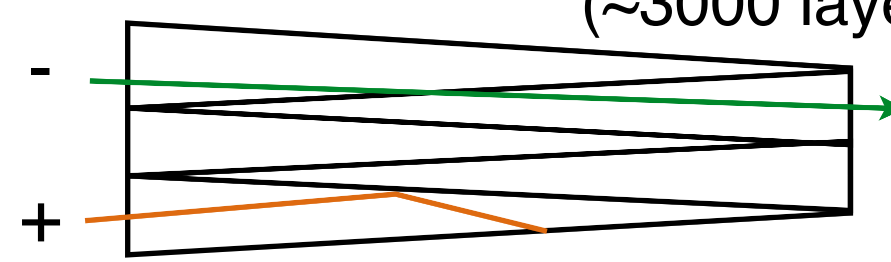


V-cavity (LET, ISIS)



transmits desired spin state

m = 5 Fe/Si
(~3000 layers)



^{3}He

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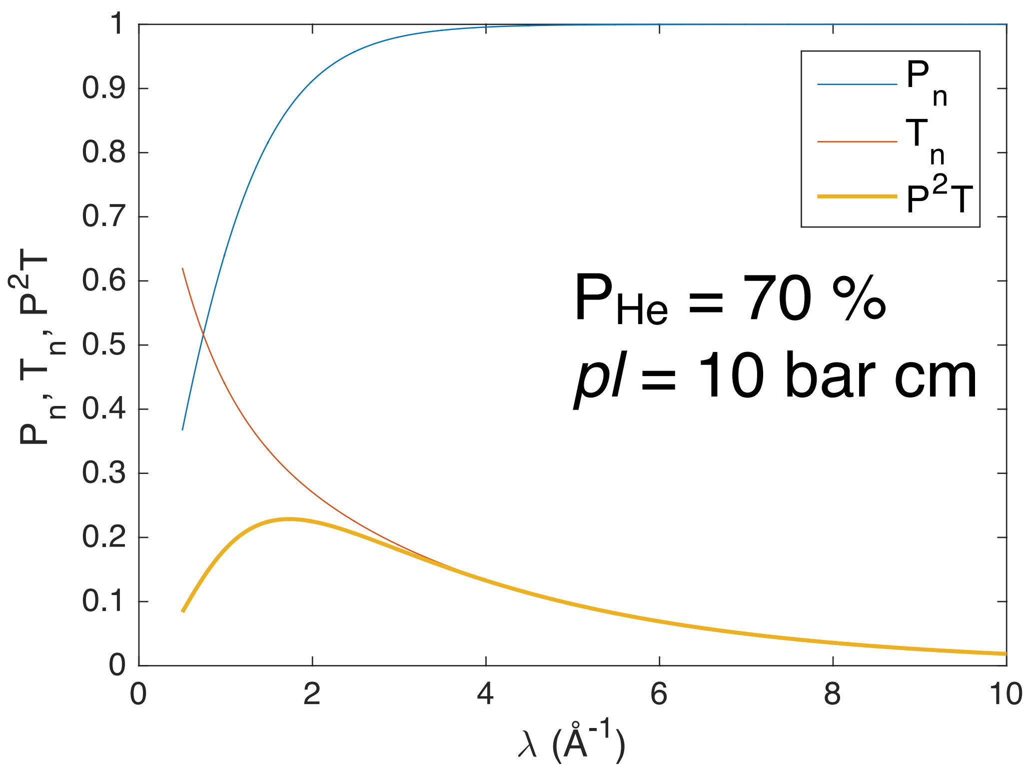
In the 80's and 90's, ^{3}He was developed as a polarizer/analyzer. The absorption of ^{3}He depends on the relative orientation of its nuclear spin to the neutron spin:



total absorption

spin-dependent

$$\sigma_\pm = \sigma_0 \pm \sigma_p$$

spin-independent

$$T = \exp(-O)\cosh(OP_{He})$$

$$P_n = \tanh(0.0733 pl\lambda P_{He}) = \tanh(OP_{He})$$

^{3}He can be polarised by either optically pumping metastable ^{3}He atoms directly (**MEOP**), or by pumping an alkali metal vapour, and transferring the polarisation via spin exchange collisions (**SEOP**)

\+ broad-band (<1 Å), flip in-situ — sensitive to magnetic field, time-dependent

Manipulating the neutron spin: guide fields and flippers

Adiabatic versus non-adiabatic transitions

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As might be expected, the polarization of the beam can be rotated and manipulated using spatially varying magnetic fields. But what type of field profile leads to a flip, and what profile causes a field rotation?

Consider a neutron moving through a magnetic field changing at a constant angular rate $d\theta_B/dt$. The equation of motion is:

$$\boxed{\frac{d\langle\sigma\rangle}{dt} = \gamma\langle\sigma\rangle \times \mathbf{B}}$$

The solution in a homogenous field is of course Larmor precession of $\langle\sigma\rangle$ about the field direction at frequency ω_L. In the changing field, we may identify two limits:

Adiabatic

$$\boxed{\omega_L \gg d\theta_B/dt}$$

Non-adiabatic

$$\boxed{\omega_L \ll d\theta_B/dt}$$

Preserving polarization vs. flipping it

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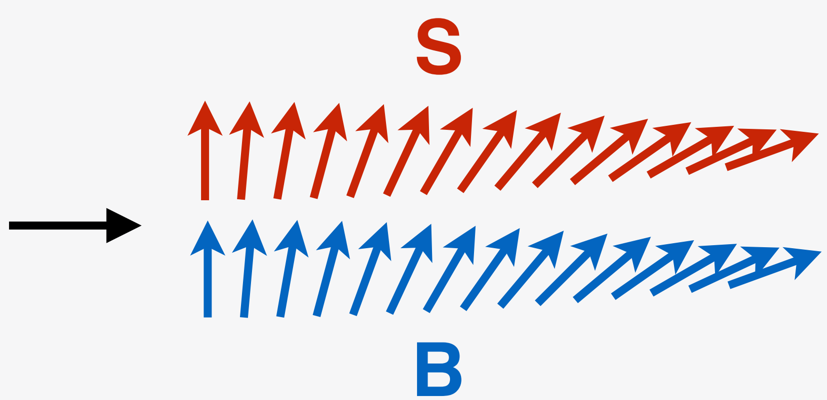
Adiabatic

$$\omega_L \gg d\theta_B/dt$$

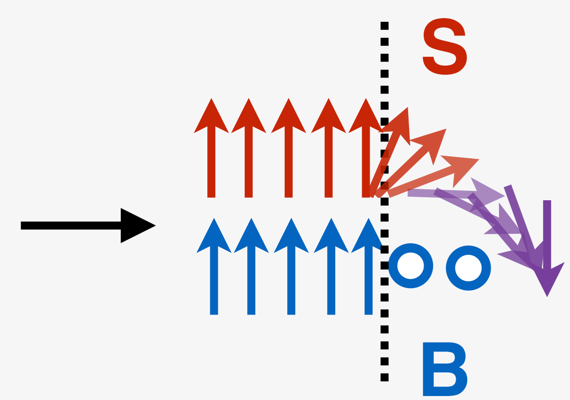
The spin follows the rotating field direction



Non-adiabatic

$$\omega_L \ll d\theta_B/dt$$

The spin immediately begins precessing about the new direction



The degree of adiabaticity may be quantified by the adiabaticity parameter:

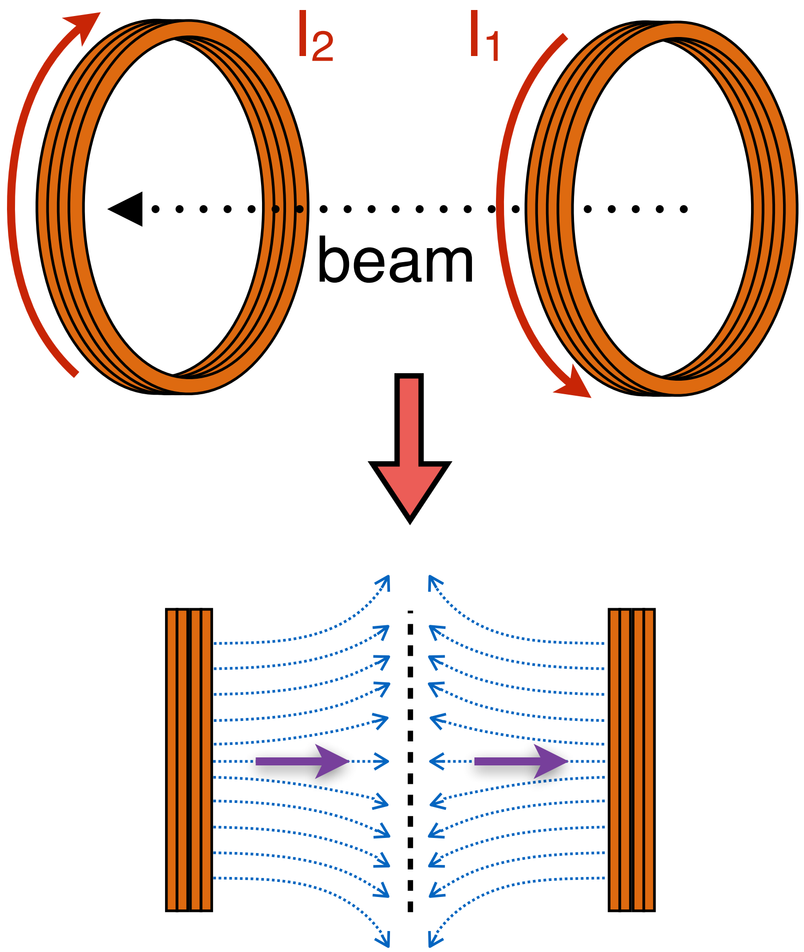
$$\boxed{A = \frac{\omega_L}{\omega_B} = \frac{|\gamma| B}{v_n (d\theta_B/dx)}}$$

For good transport of **P** along the guide field, **A** should be in excess of 10

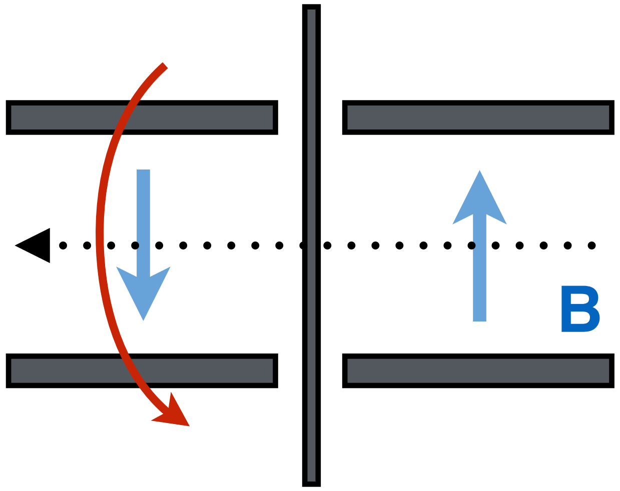
Flippers I

On the contrary, flippers require small values of A << 1

Drabkin



Cryoflipper



Meissner screen (Nb or YBCO)

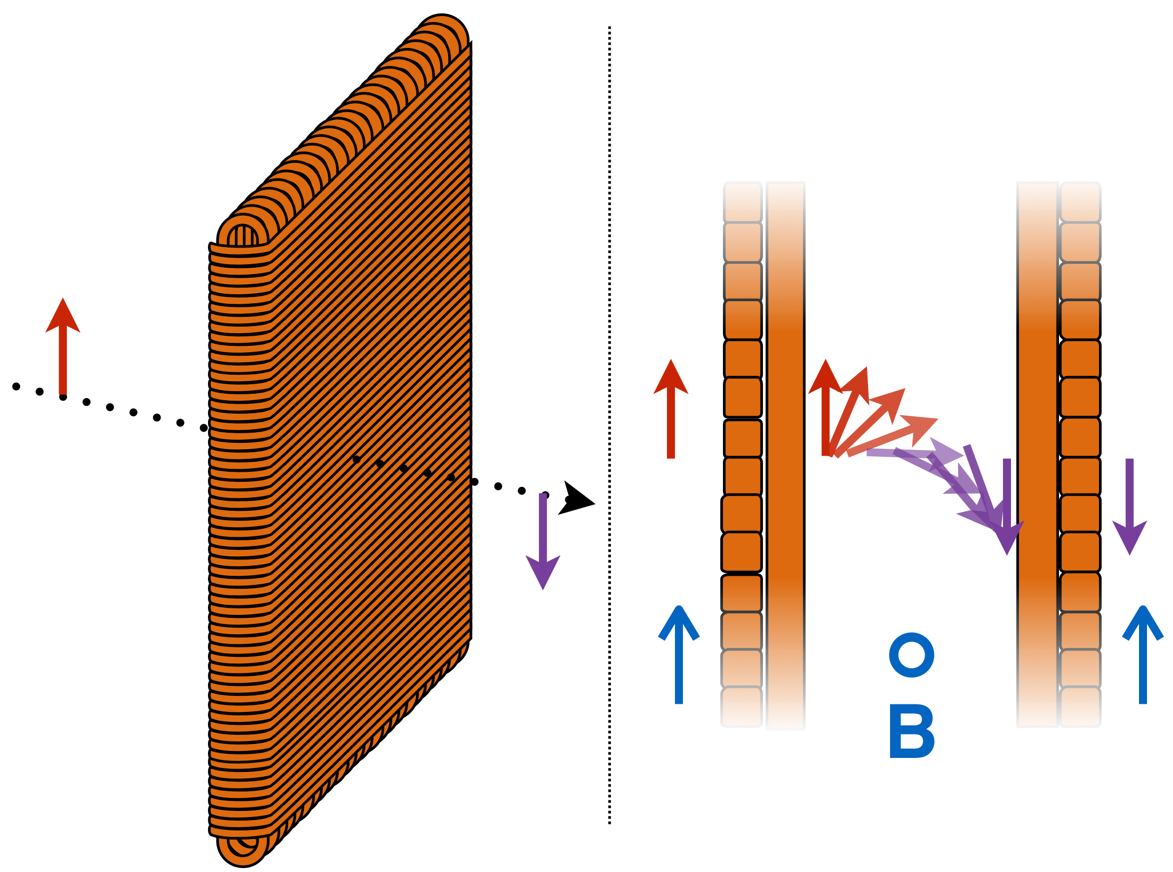
+ Broad band, no mat. in beam, simple
— Low efficiency, poor for large beams

+ Broad band, 100% efficient
— Material in beam, trapped flux

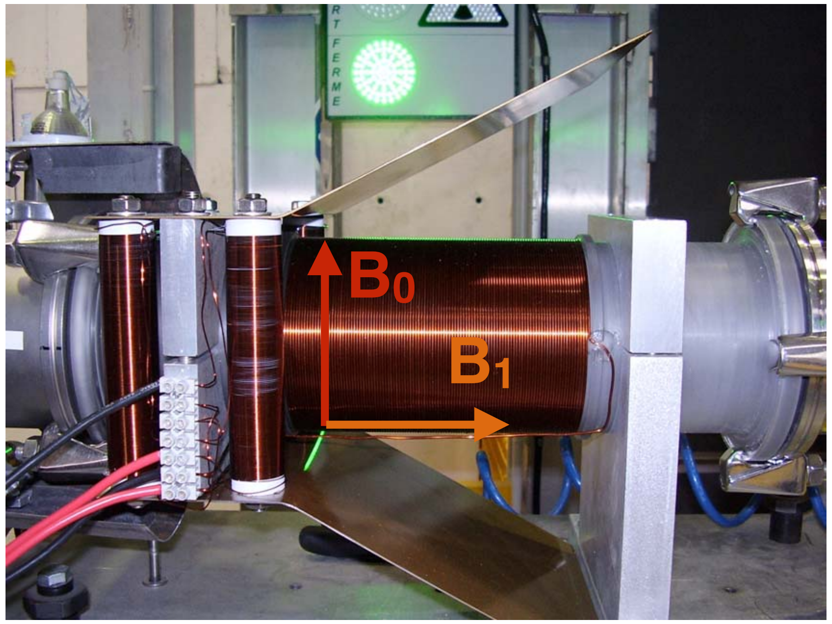
Flippers II

Larmor precession can also be used as a tool to flip the neutron

Mezei flipper



RF AFP



$$\mathbf{B}_{tot} = \left(B_0 + \frac{\omega}{\gamma}\right)\hat{z} + B_1\hat{x}$$

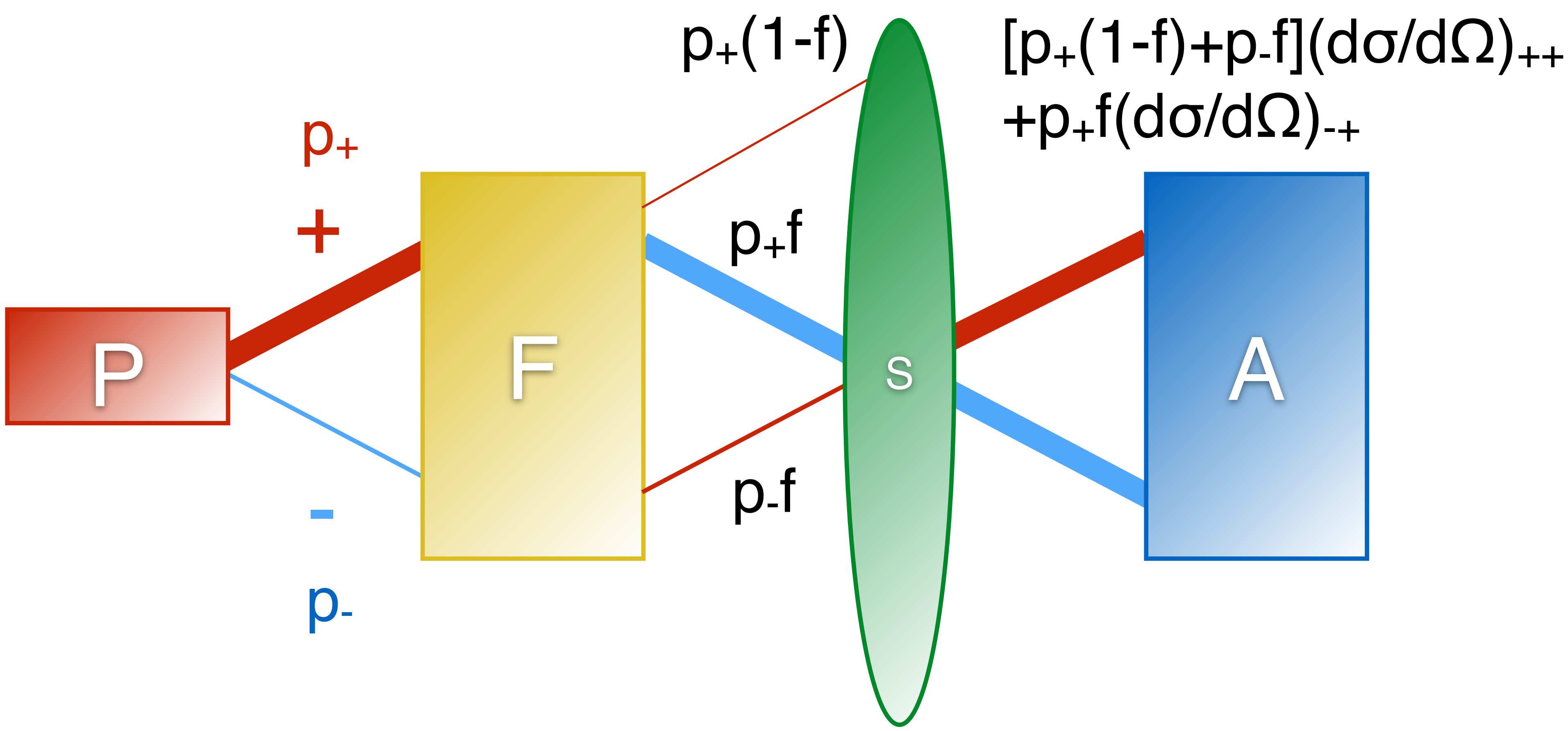
+ Any angle, high efficiency, simple
— Mat. in beam, monochromatic*

+ Broadband, high efficiency
—

A brief note on efficiency

Inevitably, the components on a neutron instrument have a finite efficiency, which has to be corrected for.

e.g. diffractometer with a single flipper, measuring -+



It is essential to characterise the instrument components!

Elastic polarized scattering

Blume-Maleev equations I

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Although it is instructive to consider the matrix elements for specific processes, it would be useful to generalise the dependence of the cross section and scattered polarization on the incident polarization. This can be achieved using the **density matrix**:

$$\langle A \rangle = \mathrm{Tr}\{\rho \mathbf{A}\} \qquad \rho = \sum_{\nu} p_\nu |\psi_\nu\rangle\langle\psi_\nu| \qquad \rho = \frac{1}{2}(\mathbf{I}_2 + \mathbf{P}\cdot\sigma)$$

density matrix

over spin states in the beam

ρ related to polarization

Let's now expand out the matrix element in the expression for the cross section shown previously. In terms of the density matrix:

$$\left(\frac{d\sigma}{d\Omega}\right) \propto |\langle\psi'|V(\mathbf{Q})|\psi\rangle|^2 = \mathrm{Tr}\{\rho V(\mathbf{Q})^\dagger V(\mathbf{Q})\}$$

Similarly, the scattered polarization is:

$$\mathbf{P}'\left(\frac{d\sigma}{d\Omega}\right) \propto \mathrm{Tr}\{\rho V(\mathbf{Q})^\dagger \sigma V(\mathbf{Q})\}$$

Blume (Phys Rev **130**, 1670, 1963, Physica B **267-268**, 211, 1999); Maleev JETP **14** (1962) 1168

Blume-Maleev equations II

Using the properties of the Pauli matrices (and their products) and the identity:

$$(\mathbf{a}\cdot\sigma)(\mathbf{b}\cdot\sigma) = \mathbf{a}\cdot\mathbf{b}\mathbf{I} + i(\mathbf{a}\times\mathbf{b})\cdot\sigma$$

… we arrive at the **Blume-Maleev equations** (ignoring incoherent, dropping **Q**):

chiral

$$\left(\frac{d\sigma}{d\Omega}\right) \propto NN^* + \underbrace{\mathbf{M}_\perp\cdot\mathbf{M}_\perp^* - i\mathbf{P}\cdot(\mathbf{M}_\perp\times\mathbf{M}_\perp^*)}_{\text{Magnetic}} + \underbrace{\mathbf{P}\cdot(\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N)}_{\text{Nuclear-magnetic interference}}$$

Not observed in unpolarized experiment

$$\boxed{\begin{aligned}\mathbf{P}'\left(\frac{d\sigma}{d\Omega}\right) \propto{}& \mathbf{P}NN^* - \mathbf{P}(\mathbf{M}_\perp\cdot\mathbf{M}_\perp^*) + \mathbf{M}_\perp(\mathbf{P}\cdot\mathbf{M}_\perp^*) + \mathbf{M}_\perp^*(\mathbf{P}\cdot\mathbf{M}_\perp) + \ldots \\ & \underbrace{i(\mathbf{M}_\perp\times\mathbf{M}_\perp^*) + \mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N}_{\text{create polarization}} + i(\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N)\times\mathbf{P}\end{aligned}}$$

Half-polarised instrument

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Let us first look at a simple application of this; a half polarised diffractometer

$$\left(\frac{d\sigma}{d\Omega}\right) \propto NN^* + \mathbf{M}_\perp \cdot \mathbf{M}_\perp^* - i\mathbf{P} \cdot (\mathbf{M}_\perp \times \mathbf{M}_\perp^*) + \mathbf{P} \cdot (\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N)$$

If we assume (as in the polarizer example), $\mathbf{M} \parallel |\mathbf{P}| \perp \mathbf{Q}$, and $\mathbf{M}_\perp$ and N are real, the ratio of flipper **on** to **off** intensities is

$$R = \frac{F_N^2 + F_M^2 + 2PF_NF_M}{F_N^2 + F_M^2 - 2PF_NF_M}$$

If the nuclear structure factors are also known (they can be found from an unpolarised experiment), the magnetic structure factor F_M can be extracted for any number of Bragg peaks.

Spin density measurement

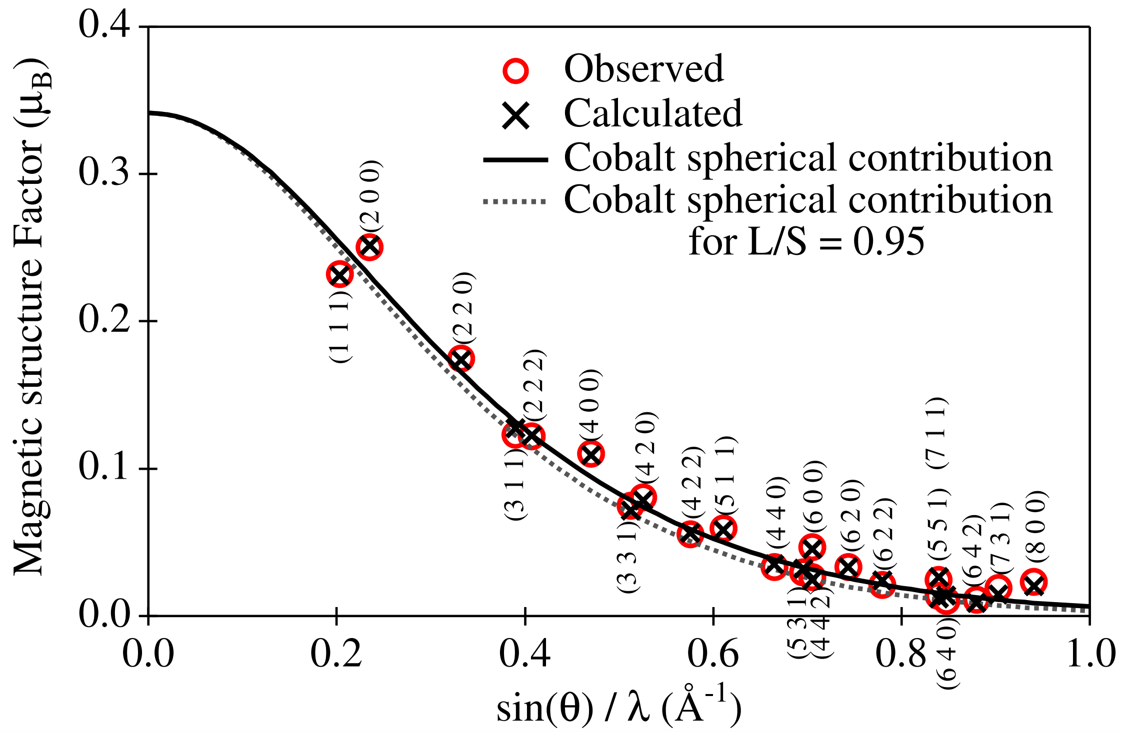
In a ferromagnet, the magnetic scattering amplitude depends only on the form factor (the spatial Fourier transform of the spin density)

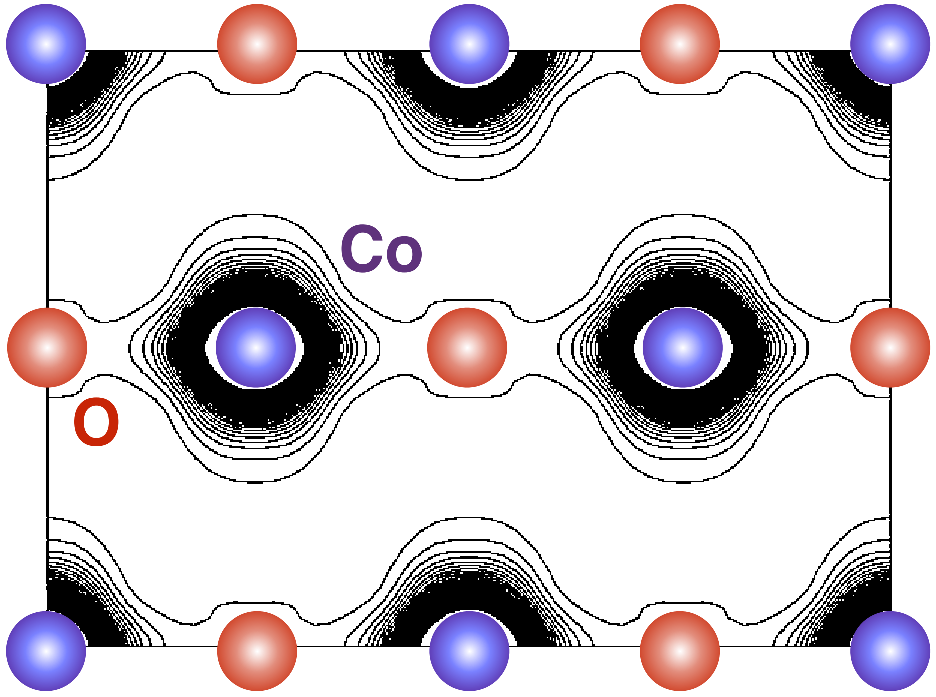
$$\rho_s(\mathbf{r}) \xrightarrow{\mathrm{FT}} f(\mathbf{Q})$$

Thus, flipping ratio measurements can be used to extract real space spin density maps for both ferromagnets and materials which be polarised in a magnetic field

e.g. CoO

projection along [0$\bar{1}$1]





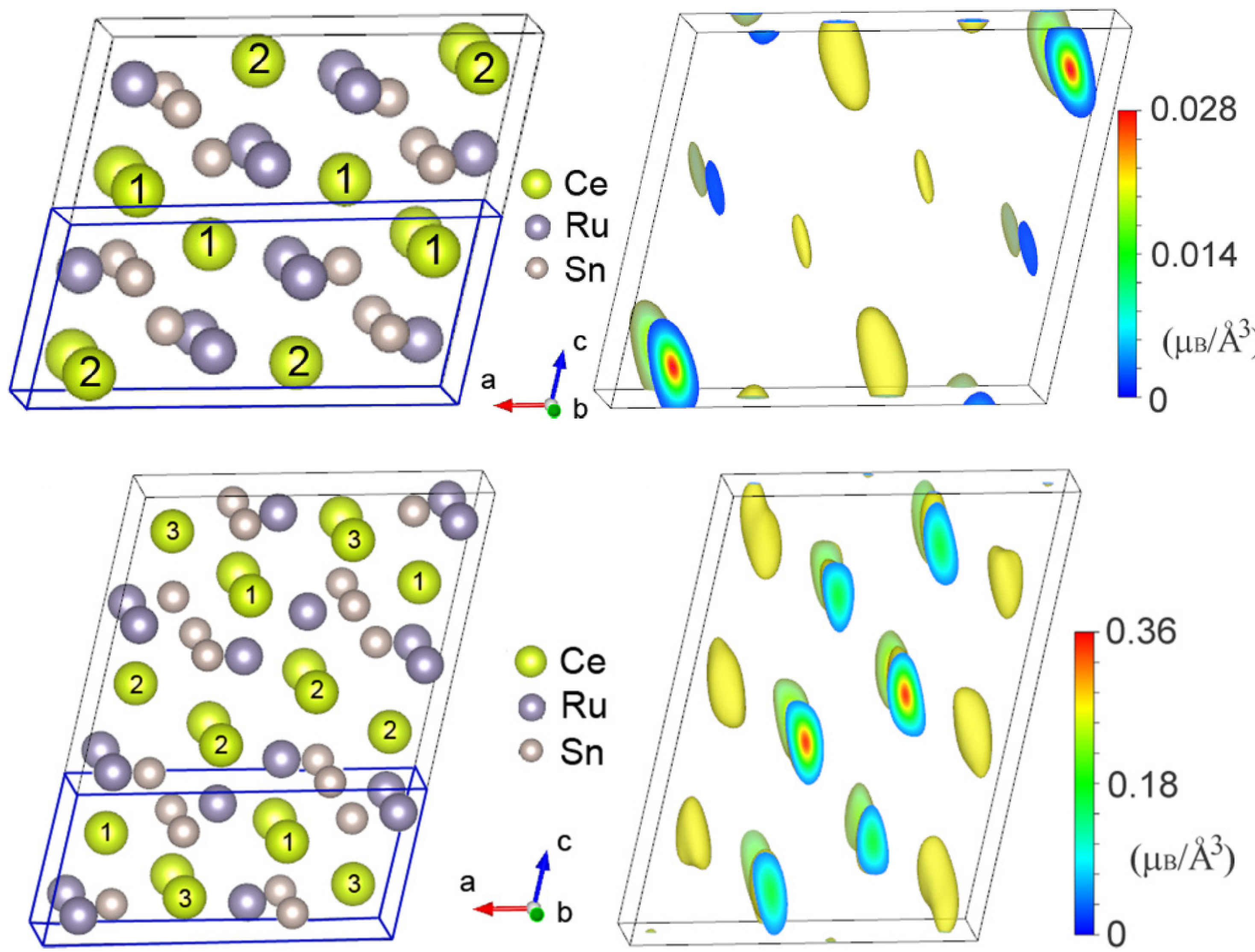
Kernavanois JPCM **15** (2003) 3433

Example: CeRuSn

System related to both famous Ce-based Kondo materials and elemental Ce.
Two Ce sites at RT, unusual structural phase transition at ~250 K

275 K
$\mu_{Ce1} = 0.003\mu_B$
$\mu_{Ce2} = 0.030\mu_B$

1.8 K
$\mu_{Ce1} = 0.60\mu_B$
$\mu_{Ce2} = 0.72\mu_B$
$\mu_{Ce3} = 0.43\mu_B$



Prokes et. al. Phys. Rev. B **91** 014424

Example: Cr spin wheel

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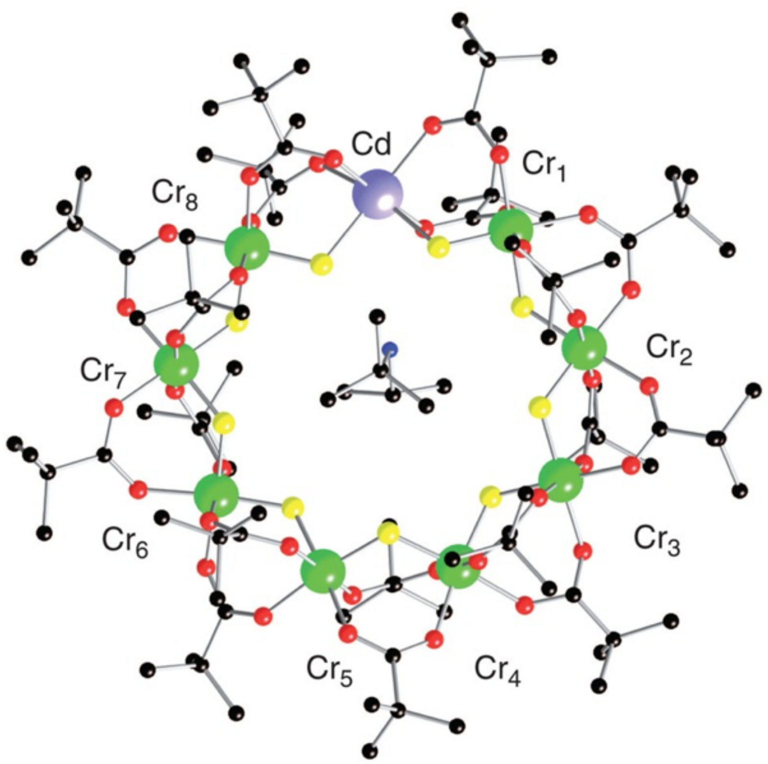
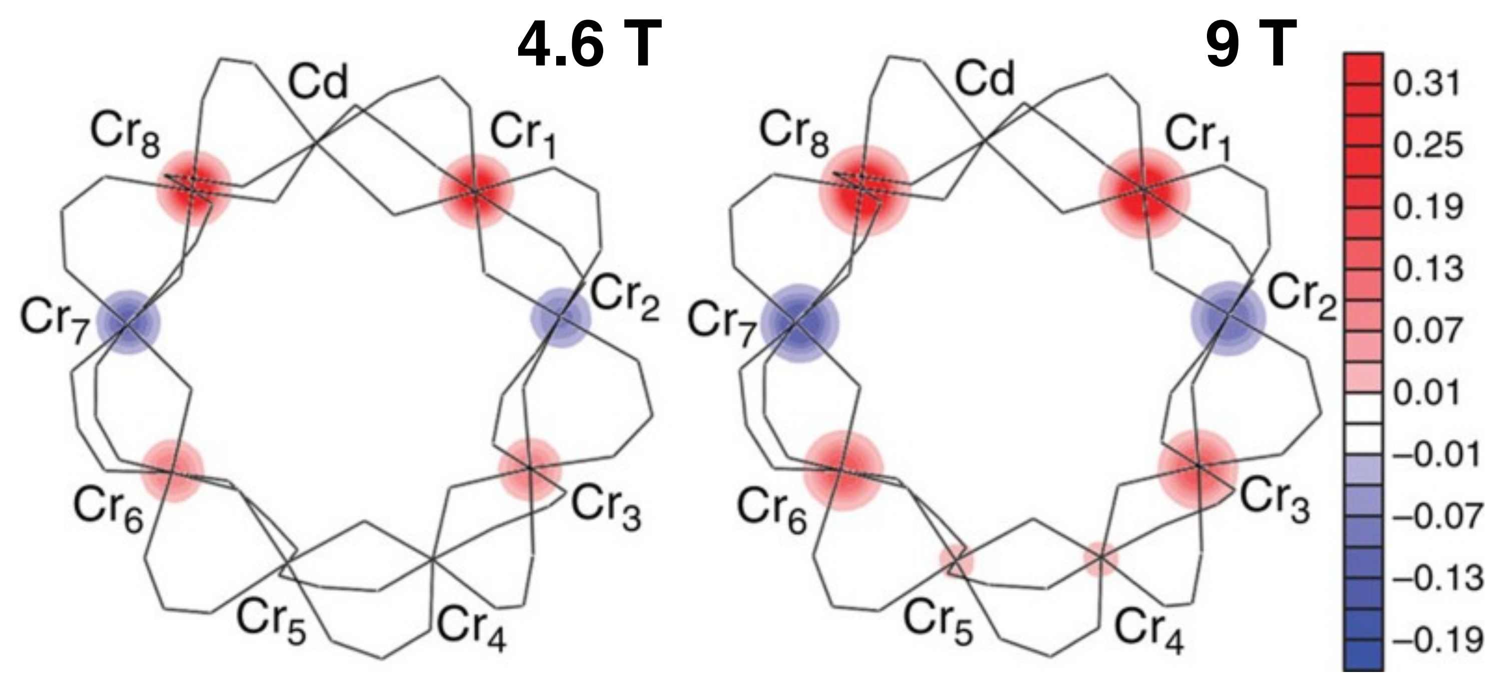
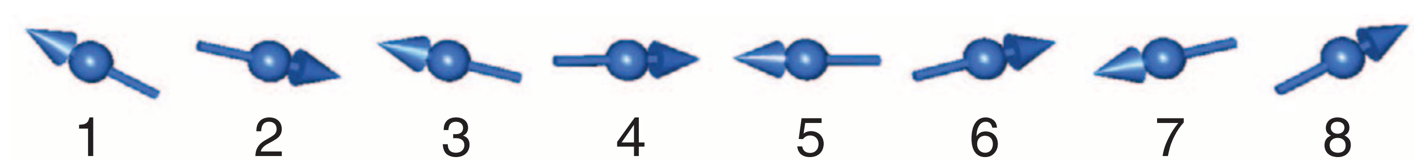
Ring of Cr^{3+} ions joined by organic linkers. One site in the ring is substituted with Cd^{3+}. Theory predicts a decrease in moment moving away from the Cd site.





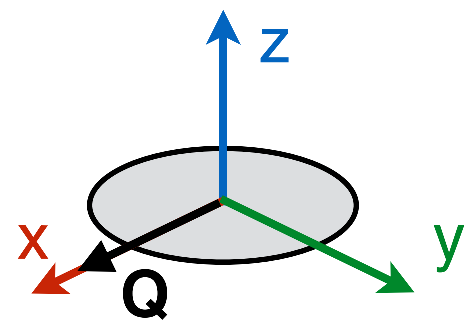
Spin structure:



Guidi et. al. Nature Comms. **6** 7061

Longitudinal polarization analysis

Longitudinal polarization analysis corresponds to the cases we looked at previously - where **P**′=±**P**. For a single crystal, define the coordinate system:



From the Blume-Maleev equations:

$$\left(\frac{d\sigma}{d\Omega}\right)^{x}_{\pm\pm} = NN^{*} \qquad \left(\frac{d\sigma}{d\Omega}\right)^{x}_{\pm\mp} = |M_{\perp}^{\perp x}|^2 \mp iP(M_{\perp}^{z*}M_{\perp}^{y} - M_{\perp}^{x}M_{\perp}^{z*})$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{y}_{\pm\pm} = |N|^2 + |M_{\perp}^{||y}|^2 \pm P(NM_{\perp}^{y*} + N^{*}M_{\perp}^{y}) \qquad \left(\frac{d\sigma}{d\Omega}\right)^{y}_{\pm\mp} = |M_{\perp}^{\perp y}|^2$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{z}_{\pm\pm} = |N|^2 + |M_{\perp}^{||z}|^2 \pm P(NM_{\perp}^{z*} + N^{*}M_{\perp}^{z}) \qquad \left(\frac{d\sigma}{d\Omega}\right)^{z}_{\pm\mp} = |M_{\perp}^{\perp z}|^2$$

These are equivalent to the Moon, Riste, and Koehler equations for P_x II **Q**

LPA: Single detector

If we only have a single detector (like the example discussed previously), we can ensure that **x** || **Q** for any **Q** using adiabatic rotations of the polarization.

Measuring the 12 cross sections on the previous slide allows us to perform a separation of the components (again, ignoring incoherent scattering):

$$|N|^2 = \left(\frac{d\sigma}{d\Omega}\right)^x_{\pm\pm} \quad |M_\perp^{||y}|^2 = \frac{1}{2}\left[\left(\frac{d\sigma}{d\Omega}\right)^y_{++} + \left(\frac{d\sigma}{d\Omega}\right)^y_{--} - \left(\frac{d\sigma}{d\Omega}\right)^x_{++} - \left(\frac{d\sigma}{d\Omega}\right)^x_{--}\right]$$

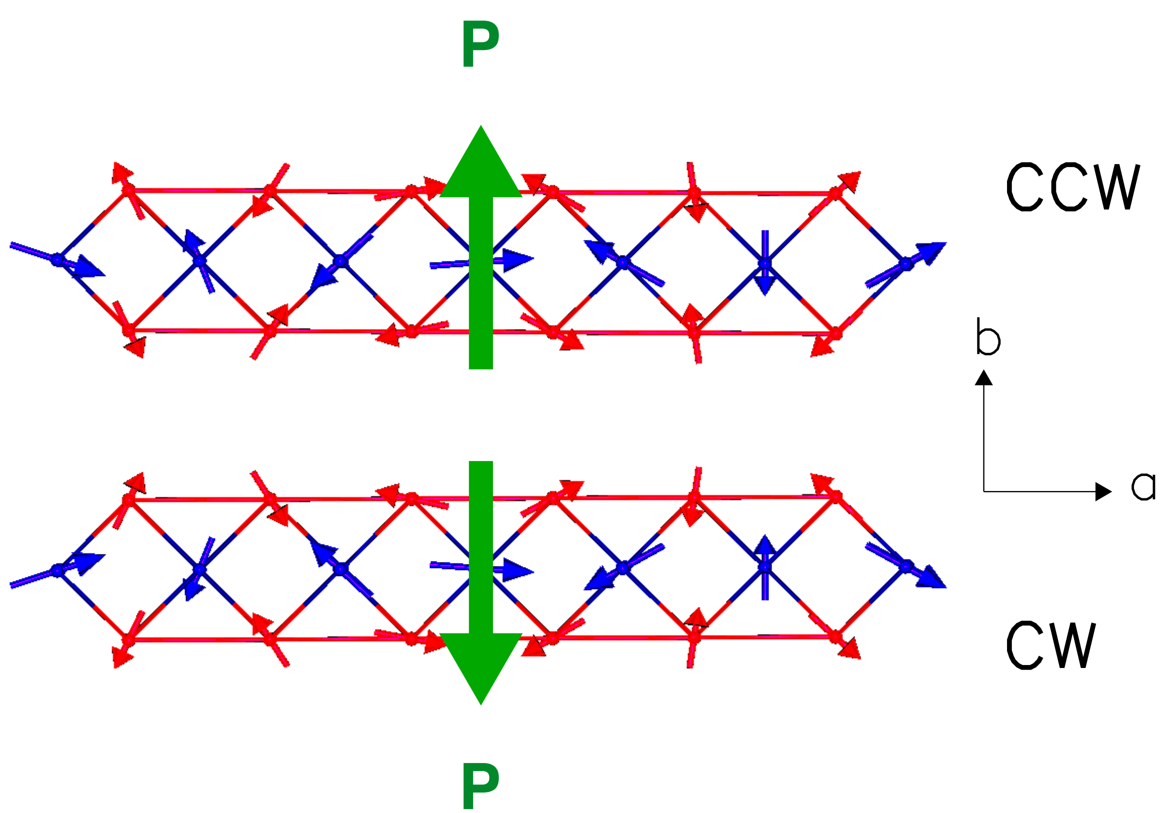
$$|M_\perp^{||y}|^2 = \frac{1}{2}\left[\left(\frac{d\sigma}{d\Omega}\right)^z_{++} + \left(\frac{d\sigma}{d\Omega}\right)^z_{--} - \left(\frac{d\sigma}{d\Omega}\right)^x_{++} - \left(\frac{d\sigma}{d\Omega}\right)^x_{--}\right]$$

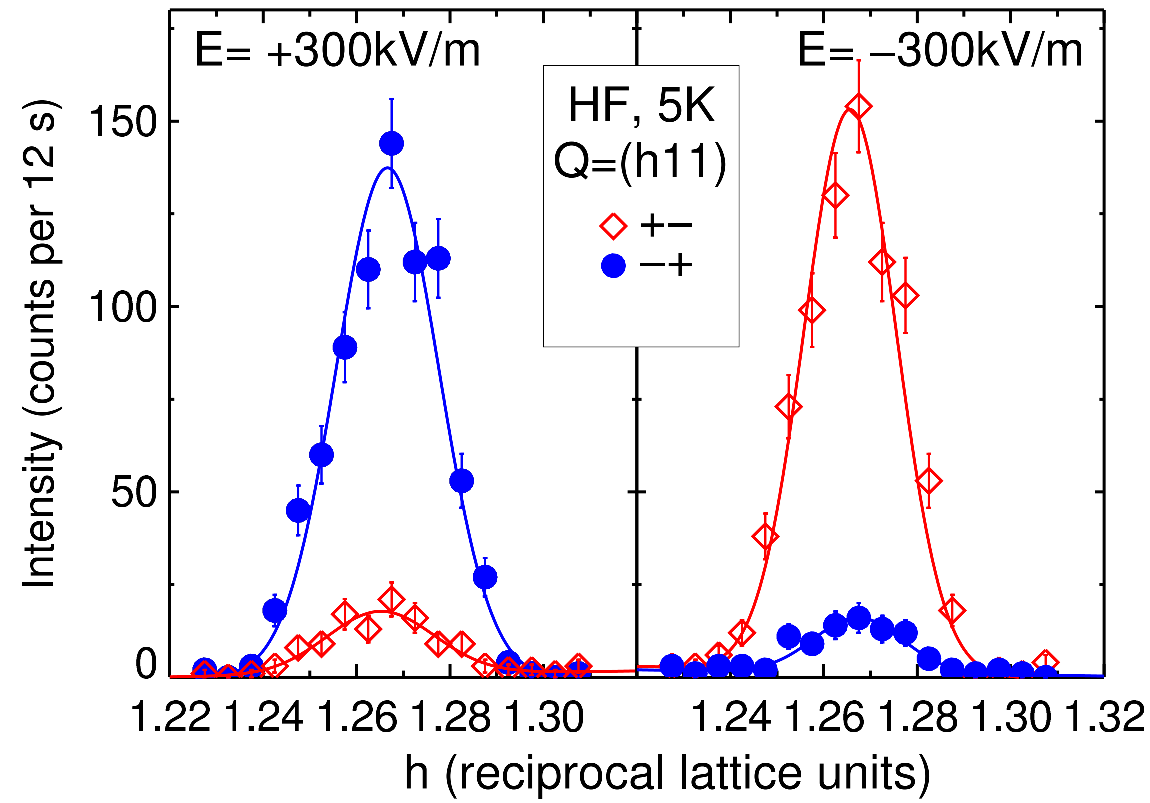
$$i(M_\perp^{z*}M_\perp^y - M_\perp^x M_\perp^{z*}) = \frac{1}{2P}\left[\left(\frac{d\sigma}{d\Omega}\right)^x_{+-} - \left(\frac{d\sigma}{d\Omega}\right)^x_{-+}\right]$$

… and so on …

Example: $Ni_3V_2O_8$

$Ni_3V_2O_8$ comprises Ni^{2+} spins on a complex, buckled kagome lattice. The spin order is helicoidal, which induces a ferroelectric polarization below ~6.5 K. The direction is linked with the chirality of the helix. Can this be observed?



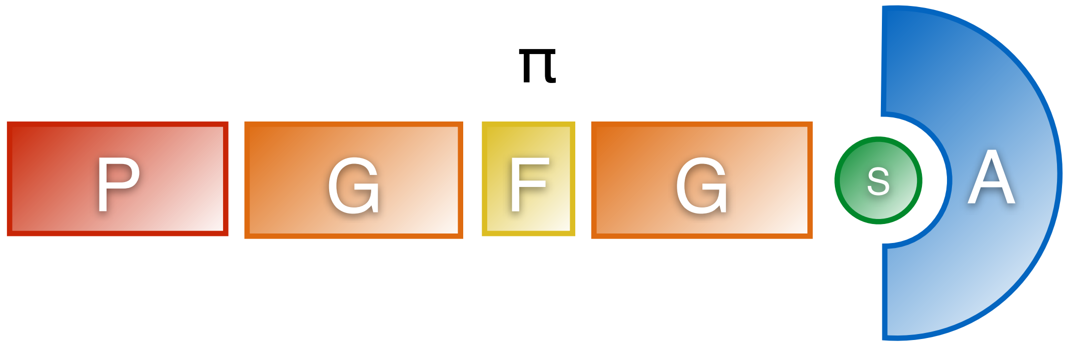


$$\left(\frac{d\sigma}{d\Omega}\right)^{x}_{\pm\mp} = |M_\perp^{\perp x}|^2 \mp iP(M_\perp^{z*} M_\perp^{y} - M_\perp^{x} M_\perp^{z*})$$

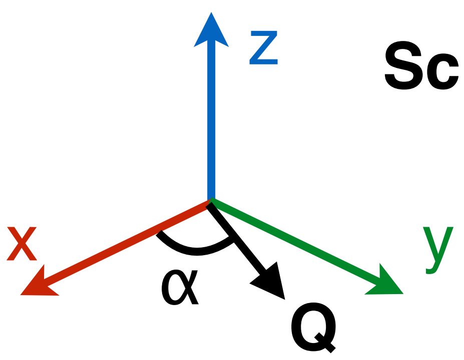
Cabrera et. al. Phys. Rev. Lett. **103** (2009) 087201

LPA: 2D detector

If there is more than one detector, it is no longer possible to simultaneously align the polarization with **Q**. It is also generally difficult to flip over a wide angle. These two limitations mean some assumptions have to be made:



1. The detector is 2D and lies in the plane containing **x**, **y**, and **Q** *i.e.*



Schärpf angle α

Otto Schärpf

D7

LPA: 2D detector

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2. There sample does not have a net magnetic moment, nor are there any no chiral or nuclear-magnetic interference contributions:

$$\left(\frac{d\sigma}{d\Omega}\right)^{\alpha}_{-+} = \left(\frac{d\sigma}{d\Omega}\right)^{\alpha}_{+-}$$

3. The magnetic component is isotropically distributed in space, i.e. the scattering is paramagnetic-like and the Halpern-Johnson equation applies:

$$\boxed{\mathbf{P}' = \hat{\mathbf{Q}}[\mathbf{P} \cdot \hat{\mathbf{Q}}]}$$

All the direction-dependent components replaced by averages.

Including the incoherent cross sections, which have been neglected so far, the result is the **Schärpf equations…**

Schärpf and Capellmann phys. stat. sol. (a) **135**, (1993), 359
Stewart J. Appl. Cryst **42** (2009) 69

Schärpf equations

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$$\left(\frac{d\sigma}{d\Omega}\right)^{z}_{++} = \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc}$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{z}_{-+} = \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si}$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{x}_{++} = \frac{1}{2}\cos^2\alpha\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc}$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{x}_{-+} = \frac{1}{2}(\sin^2\alpha + 1)\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si}$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{y}_{++} = \frac{1}{2}\sin^2\alpha\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc}$$

$$\left(\frac{d\sigma}{d\Omega}\right)^{y}_{-+} = \frac{1}{2}(\cos^2\alpha + 1)\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{si}$$

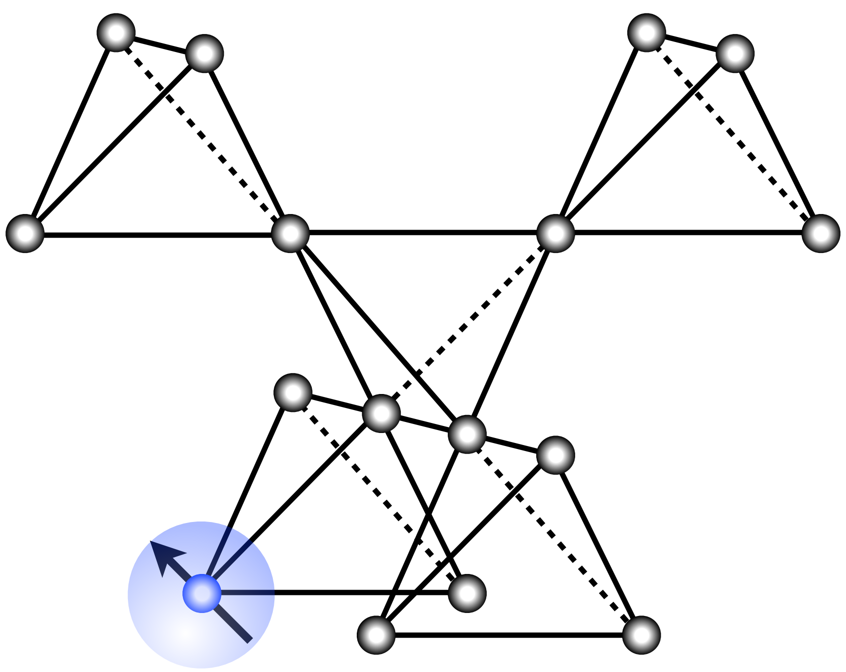
Schärpf and Capellmann phys. stat. sol. (a) **135**, (1993), 359
Stewart J. Appl. Cryst **42** (2009) 69

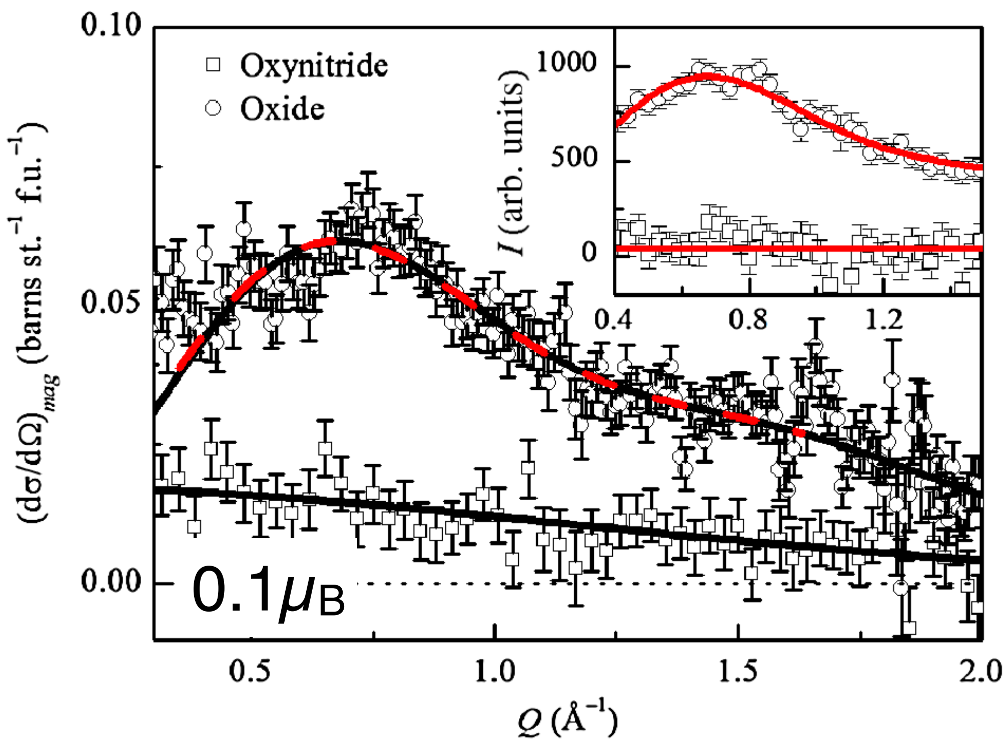
Example: diffuse magnetic scattering

These conditions are generally met for diffuse scattering from powder samples:

$$\left(\frac{d\sigma}{d\Omega}\right)_{mag} = 2\left[\left(\frac{d\sigma}{d\Omega}\right)^{x}_{-+} + \left(\frac{d\sigma}{d\Omega}\right)^{y}_{-+} - 2\left(\frac{d\sigma}{d\Omega}\right)^{z}_{-+}\right]$$

e.g. frustrated pyrochlore antiferromagnetic $Lu_2Mo_2O_5N_2$ and $Lu_2Mo_2O_7$…





L. Clark, GJN, et. al. PRL **113** (2014) 117201

Example: Spin ices

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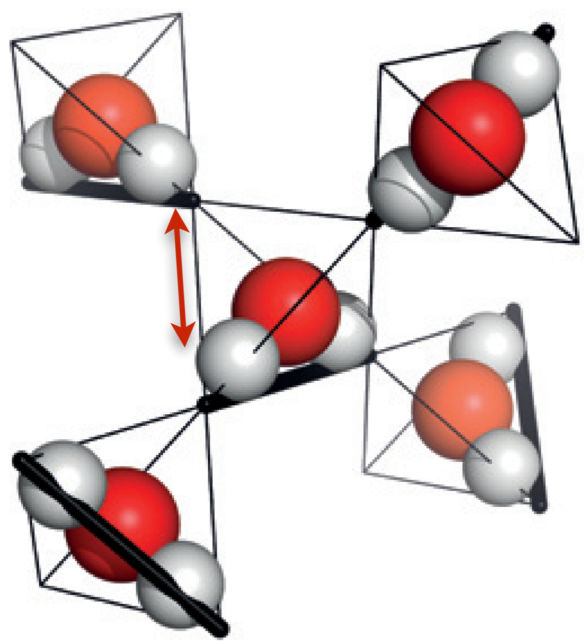
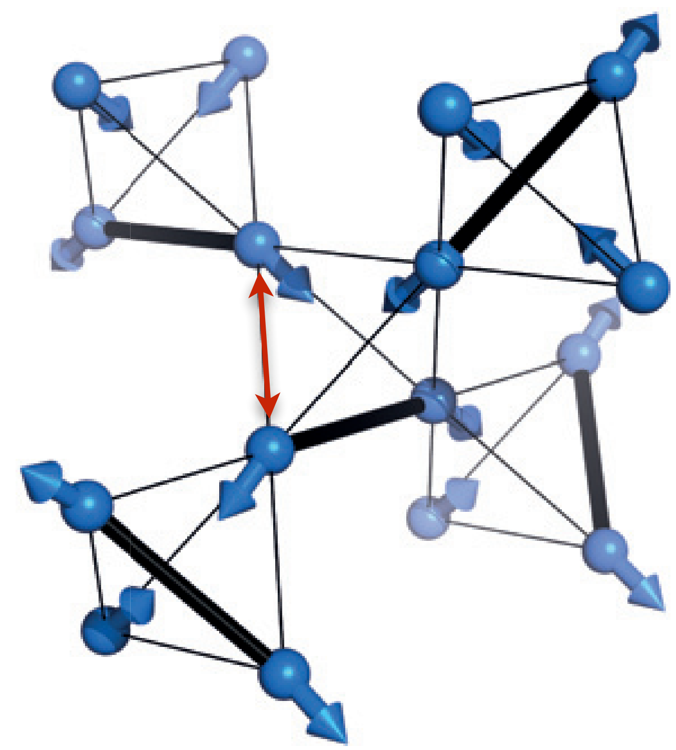
3D water ice



Coulomb repulsion
2-near 2-far

3D spin ice



Dipolar coupling & anisotropy
2-in 2-out

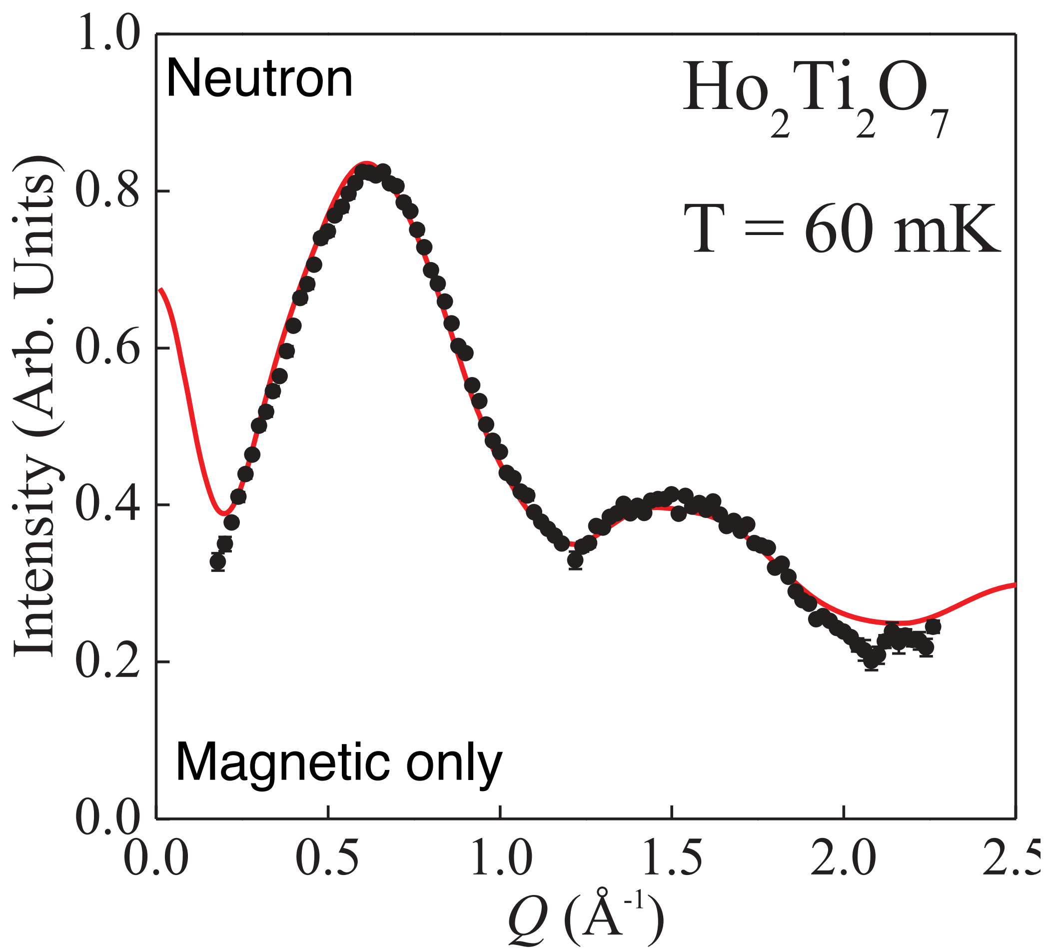
Both are under-constrained - infinite number of ways to satisfy interactions

Keen Nature **521** 303; Bernal, Fowler J. Chem. Phys **1** 515
Ramirez, Nature **399** 333; Bramwell PRL **87** 047205

Example: spin ices

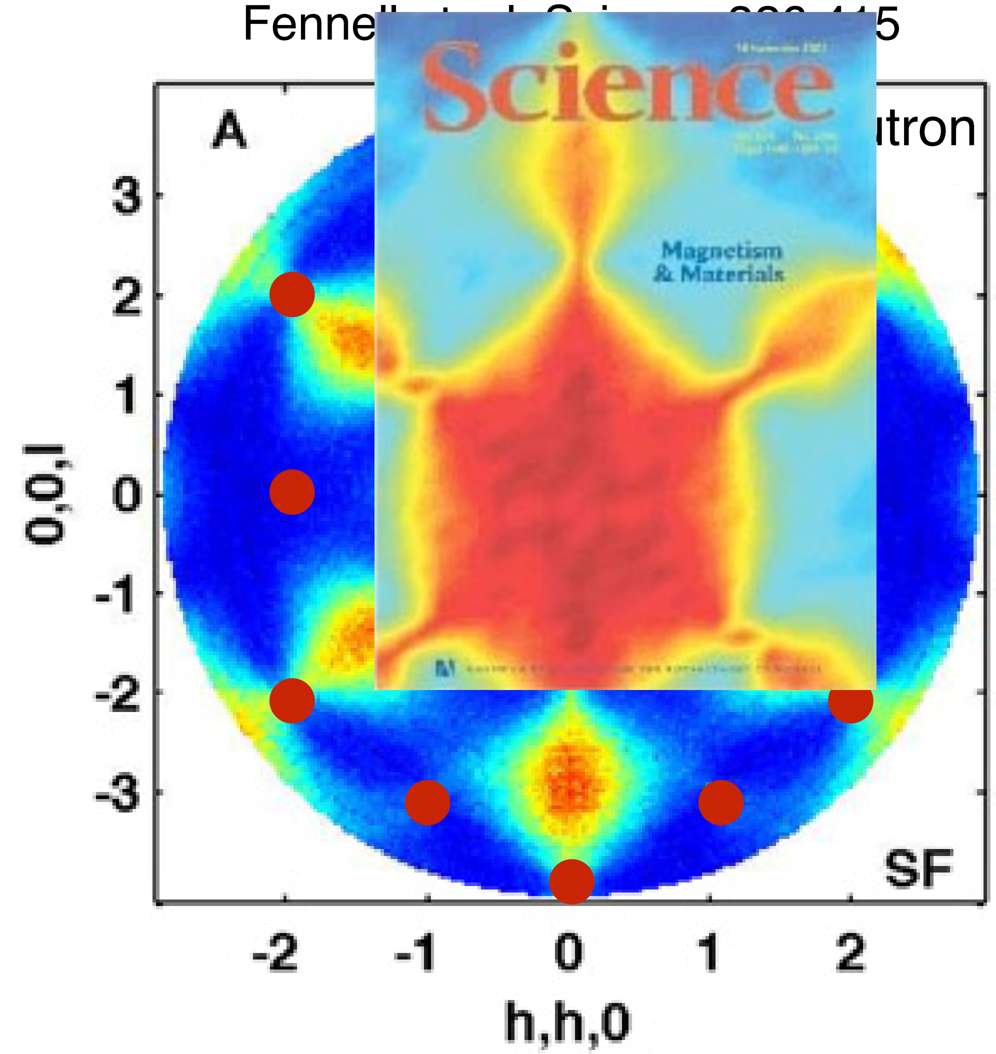
Powder

Hallas et. al. PRB 86 134431



Single crystal

Fennell et. al. Science 326 415



Spherical neutron polarimetry

Slides: R. Stewart

Neutron Polarimetry

To this point we have only been concerned with applying uniaxial polarization analysis - i.e. with measuring the scattered intensity associated with a scalar change of polarization along a particular axis.

This is "uniaxial (longitudinal) polarization analysis"

For a full description of the scattering processes we must perform "polarization analysis" in its true sense, i.e. we must measure all components of the polarization vector

This is "neutron polarimetry"

$$\sigma = \begin{cases} NN^* & \text{Nuclear} \\ \mathbf{M}_\perp \cdot \mathbf{M}_\perp^* + i\mathbf{P}_i \cdot \left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right) & \text{Magnetic} \\ \mathbf{P}_i \cdot \left(\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N\right) & \text{NM Interference} \end{cases}$$

and

$$\mathbf{P}_f \sigma = \begin{cases} \mathbf{P}_i NN^* & \text{Nuclear} \\ -\mathbf{P}_i\left(\mathbf{M}_\perp \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp\left(\mathbf{P}_i \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp^*\left(\mathbf{P}_i \cdot \mathbf{M}_\perp\right) - i\left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right) & \text{Magnetic} \\ \mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N - i\left(N\mathbf{M}_\perp^* - N^*\mathbf{M}_\perp\right) \times \mathbf{P}_i & \text{NM Interference} \end{cases}$$

Blume (*Phys Rev* *130*, *1670*, *1963*, *Physica B* *267-268*, *211*, *1999*)

Neutron Polarimetry

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$$\sigma = \begin{cases} NN^* & \text{Nuclear} \\ \mathbf{M}_\perp \cdot \mathbf{M}_\perp^* + i\mathbf{P}_i \cdot \left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right) & \text{Magnetic} \\ \mathbf{P}_i \cdot \left(\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N\right) & \text{NM Interference} \end{cases}$$

$$\mathbf{P}_f\sigma = \begin{cases} \mathbf{P}_i NN^* & \text{Nuclear} \\ -\mathbf{P}_i\left(\mathbf{M}_\perp \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp\left(\mathbf{P}_i \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp^*\left(\mathbf{P}_i \cdot \mathbf{M}_\perp\right) - i\left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right) & \text{Magnetic} \\ \mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N - i\left(N\mathbf{M}_\perp^* - N^*\mathbf{M}_\perp\right) \times \mathbf{P}_i & \text{NM Interference} \end{cases}$$

Points to note are:

1) Pure nuclear scattering does not effect the neutron polarization
2) The cross-terms are non zero only for non-collinear (e.g. spiral) structures where $M_\perp^*$ and $M_\perp$ are not parallel.
3) Scattering by NM interference will only occur when the nuclear and magnetic contributions occur with the same wavevector.

Where there is no NM interference and no chiral terms (which is generally true for paramagnets and glassy systems) the above equations reduce to the uniaxial equations.

Flipping-ratios revisited

$$\sigma = \begin{cases} NN^* & \text{Nuclear} \\ \mathbf{M}_\perp \cdot \mathbf{M}_\perp^* + i\mathbf{P}_i \cdot \cancel{\left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right)} & \text{Magnetic} \\ \mathbf{P}_i \cdot \left(\mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N\right) & \text{NM Interference} \end{cases}$$

$$\mathbf{P}_f \sigma = \begin{cases} \mathbf{P}_i NN^* & \text{Nuclear} \\ -\mathbf{P}_i\left(\mathbf{M}_\perp \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp\left(\mathbf{P}_i \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp^*\left(\mathbf{P}_i \cdot \mathbf{M}_\perp\right) - \cancel{i\left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right)} & \text{Magnetic} \\ \mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N - i\cancel{\left(N\mathbf{M}_\perp^* - N^*\mathbf{M}_\perp\right)} \times \mathbf{P}_i & \text{NM Interference} \end{cases}$$

The imaginary part of $M_\perp$ contains the phase information on the magnetic order. For non-chiral structures,

$$\mathbf{M}_\perp = \mathbf{M}_\perp^*$$

$$\Rightarrow \mathbf{M}_\perp^* \times \mathbf{M}_\perp = 0$$

Interaction vector

$$\left.\begin{array}{c} \mathbf{M}_\perp \propto \mathbf{Q} \times \mathbf{M} \times \mathbf{Q} \\ \mathbf{M} \perp \mathbf{Q} \end{array}\right\} \Rightarrow \mathbf{M}_\perp \| \mathbf{M} \Rightarrow \mathbf{M}_\perp \| \mathbf{P}_i$$

Sample is magnetised

$$\Rightarrow \mathbf{P}_i = \mathbf{P}_f$$

So we recover the form of the cross-section for magnetic diffraction - and the observation that there is no spin-flip scattering

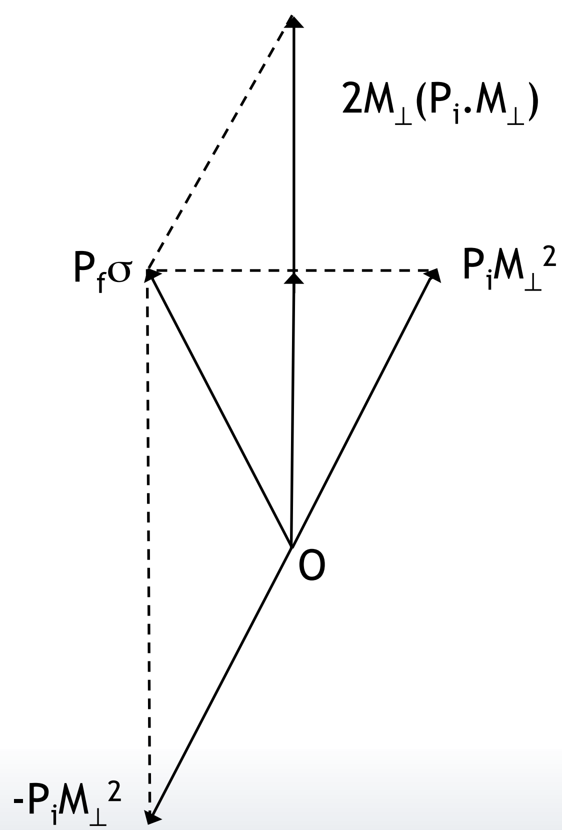
Polarimetry examples

Science & Technology Facilities Council

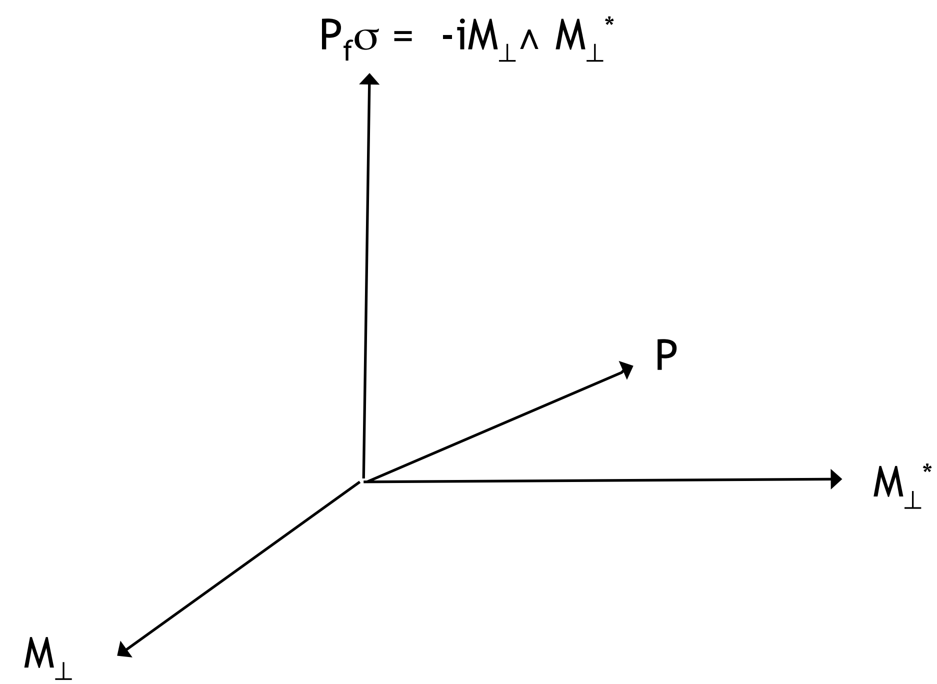
$$\mathbf{P}_f\sigma = \begin{cases} \mathbf{P}_i N N^* \\ -\mathbf{P}_i\left(\mathbf{M}_\perp \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp\left(\mathbf{P}_i \cdot \mathbf{M}_\perp^*\right) + \mathbf{M}_\perp^*\left(\mathbf{P}_i \cdot \mathbf{M}_\perp\right) - i\left(\mathbf{M}_\perp^* \times \mathbf{M}_\perp\right) \\ \mathbf{M}_\perp N^* + \mathbf{M}_\perp^* N - i\left(N\mathbf{M}_\perp^* - N^*\mathbf{M}_\perp\right) \times \mathbf{P}_i \end{cases}$$

- e.g. $M_\perp$ and $M_\perp^*$ are parallel -
- no nuclear scattering

$$\mathbf{P}_f\sigma = -\mathbf{P}_i\mathbf{M}_\perp^2 + 2\mathbf{M}_\perp\left(\mathbf{P}_i \cdot \mathbf{M}_\perp\right)$$



- e.g. $M_\perp$ and $M_\perp^*$ are perpendicular -
- no nuclear scattering - *chiral systems*



Creation of polarization - independent of incident $\mathbf{P}_i$

Polarimetry - M real

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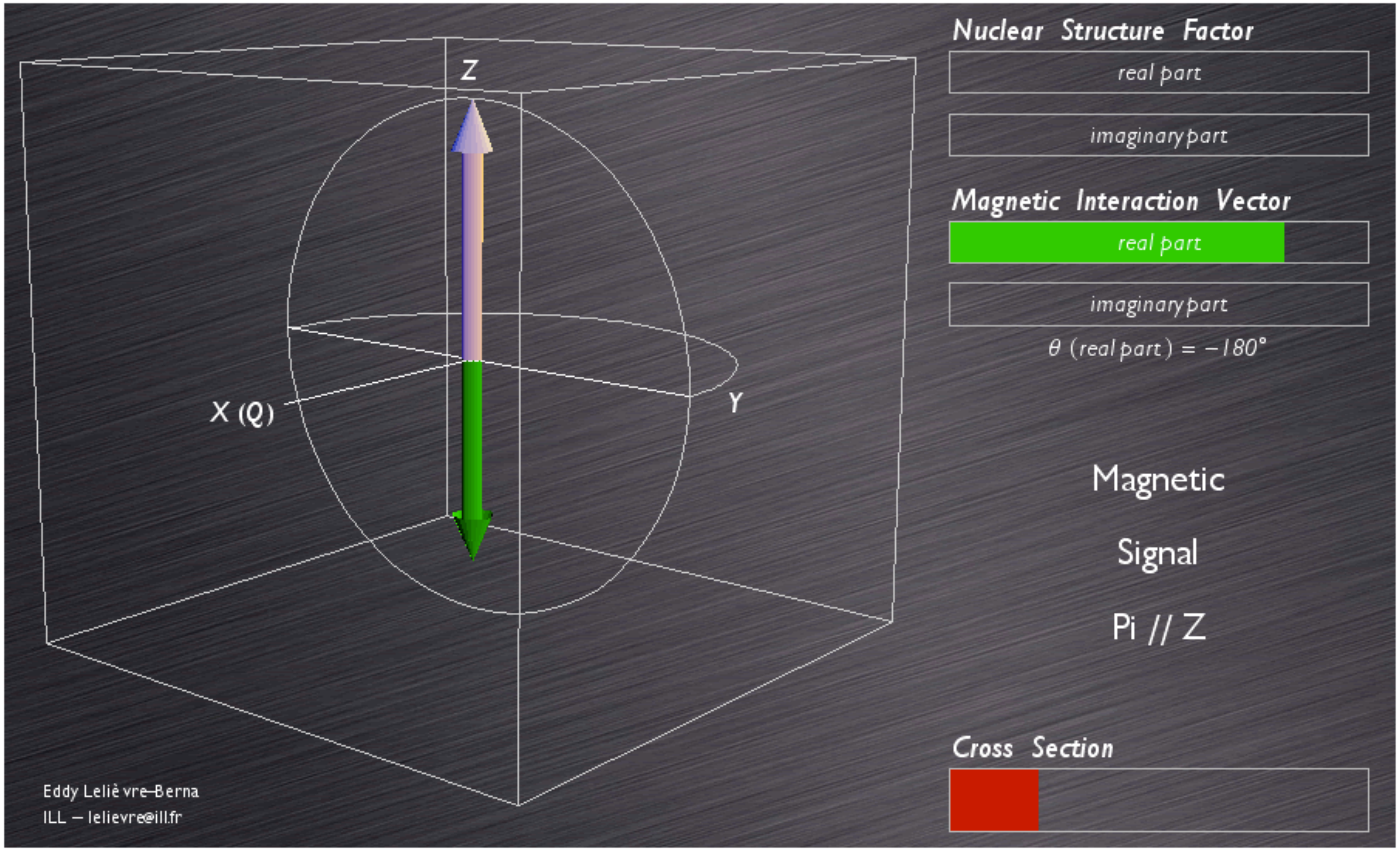
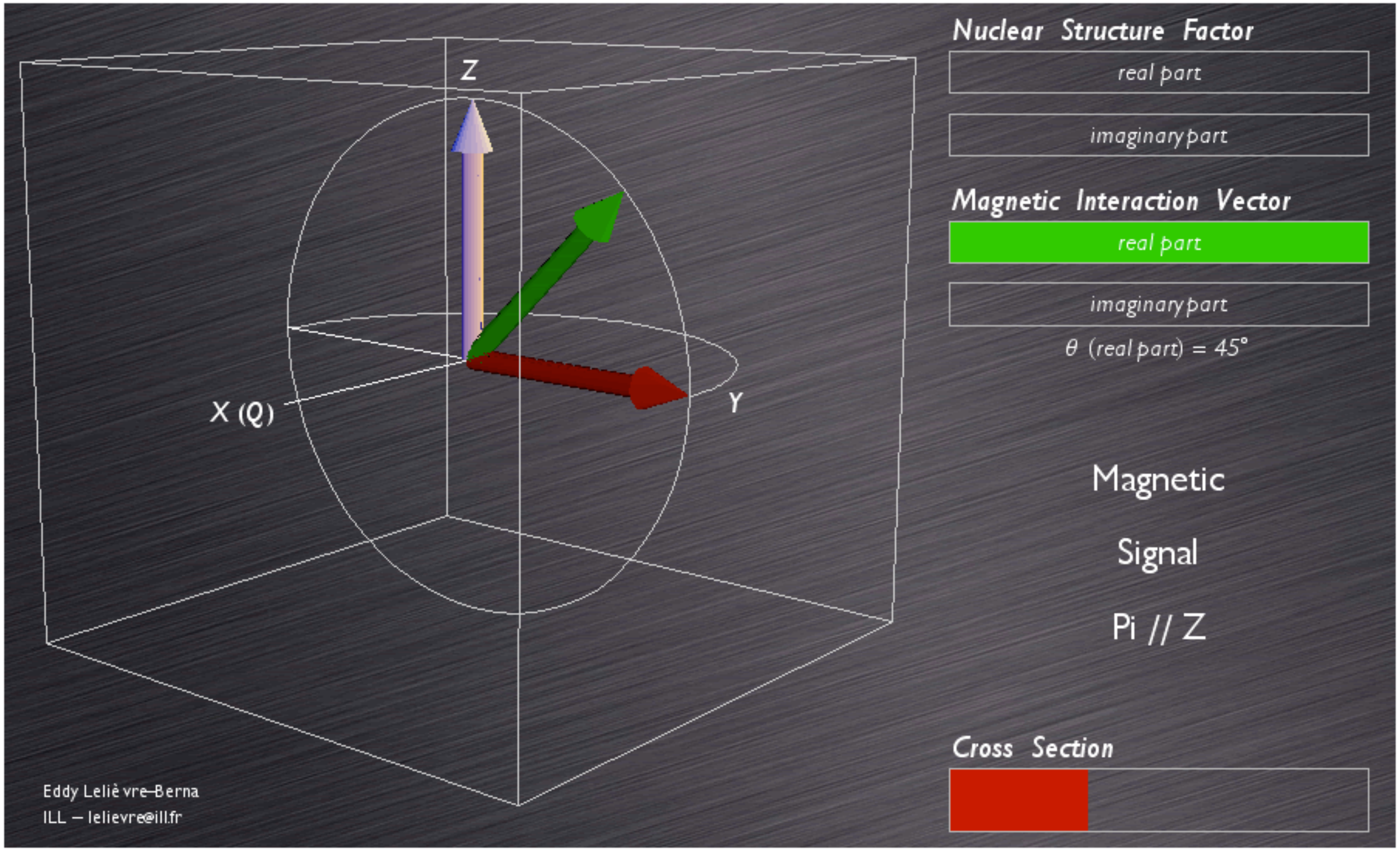
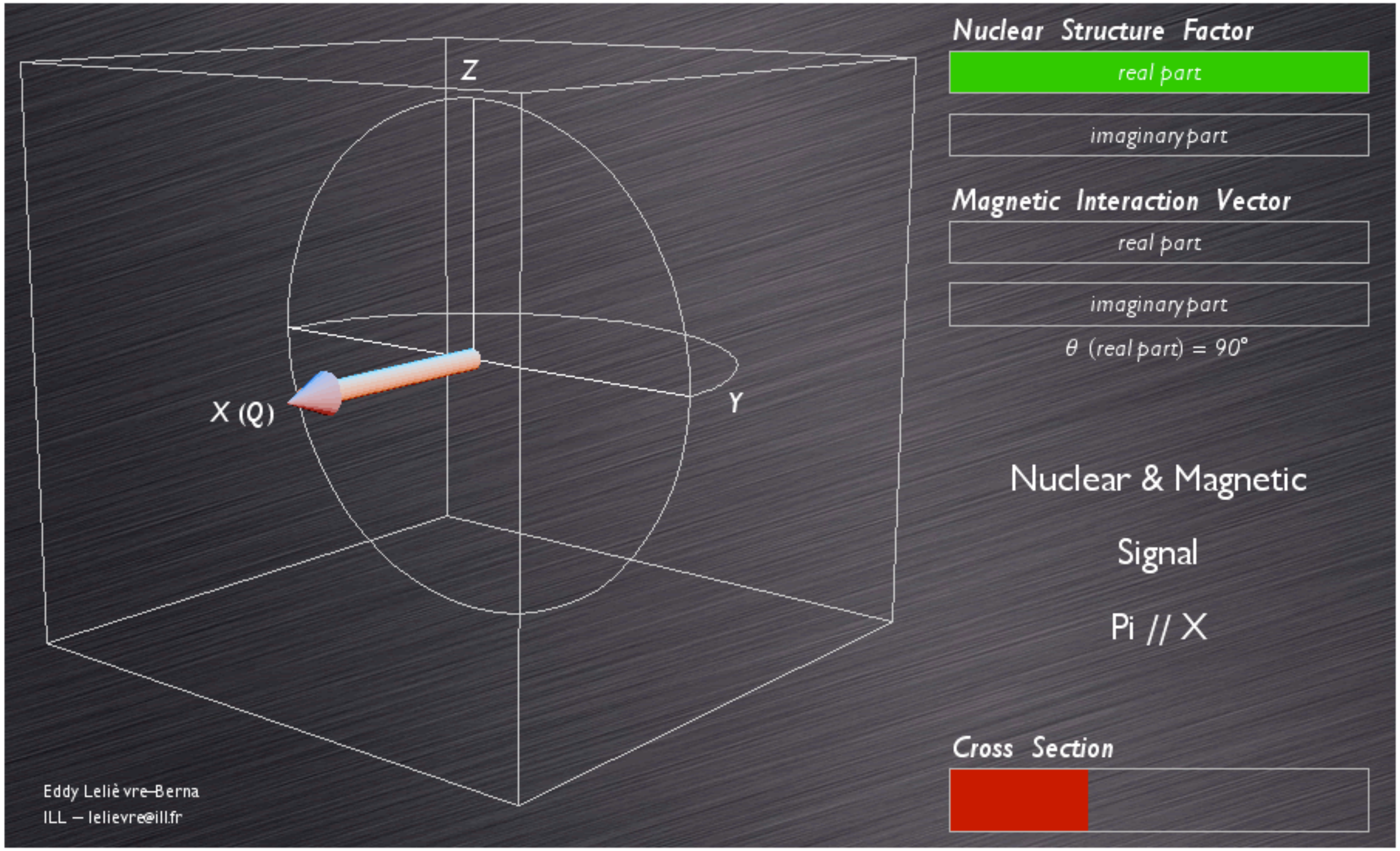


Polarimetry - M complex



Polarimetry - NM (real/real)



The Polarization Tensor

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The goal is to determine complex magnetic structures. In practice this is done by measuring the polarization tensor **P** which is unambiguously defines all the terms in the Blume equations

$$\mathbf{P}_f = \boldsymbol{P}\mathbf{P}_i \qquad \text{where} \qquad \boldsymbol{P} = \begin{bmatrix} P_{xx} & P_{xy} & P_{xz} \\ P_{yx} & P_{yy} & P_{yz} \\ P_{zx} & P_{zy} & P_{zz} \end{bmatrix}$$

In practice, this is what is measured in a neutron polarimetry measurement

As an illustrative example, in the case of collinear antiferromagnet - aligned in the z-direction, the polarization tensor would be

$$\boldsymbol{P} = \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

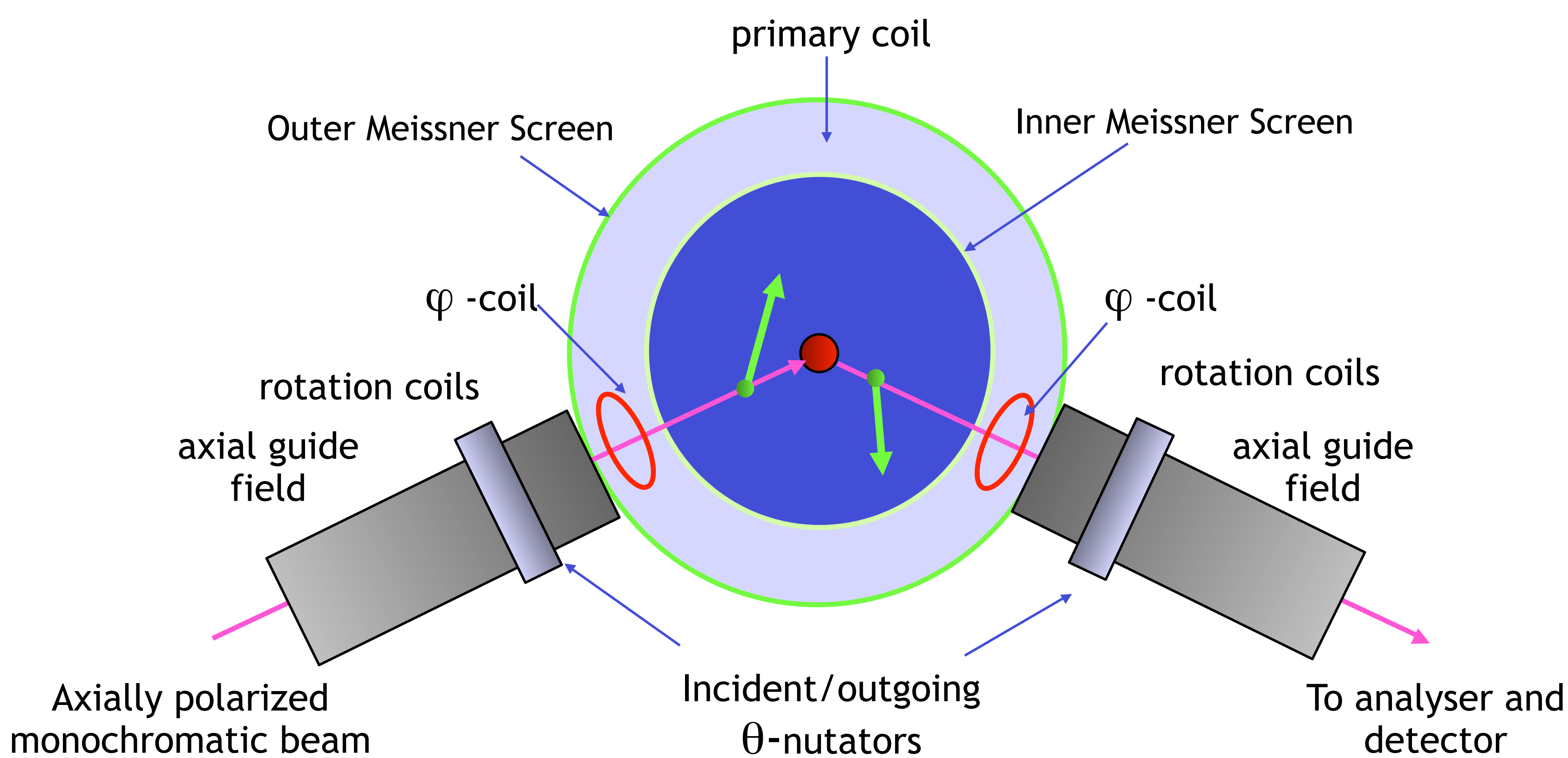
The appearance of any non-collinear magnetism would feed through into the off-diagonal components

NB The 3-directional PA method (D7) only measures the diagonal components of this matrix and would therefore miss this information

CRYOPAD

CRYOPAD has been developed by Tasset and co-workers at ILL in order to determine the vector polarization of the scattered beam for any predetermined direction of the vector polarization of the incident beam (i.e. measurement of the polarization tensor)

The field along the whole of the neutron beam is perfectly defined with the help of spin nutators, precession coils, Meissner screens, in order to align and analyse the polarization in any direction in space.



Tasset et al, Physica B 267-8, 69, (1999)

Examples from CRYOPAD

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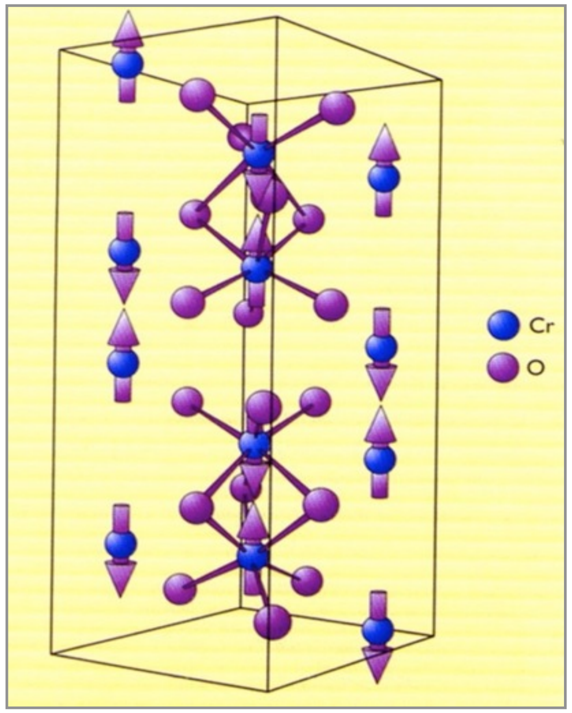
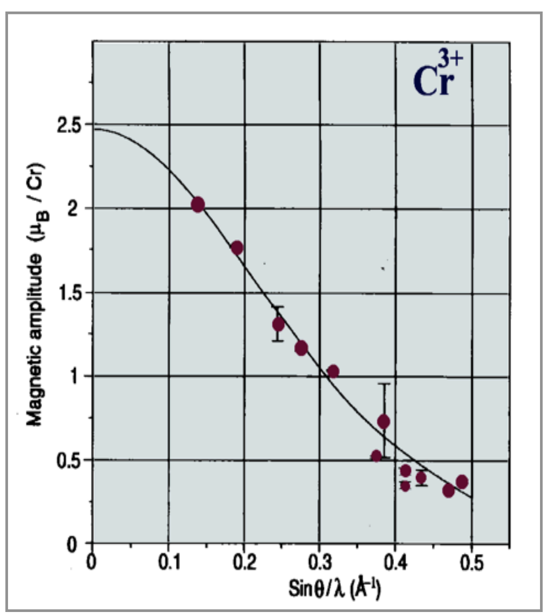


Cr_2O_3 *Brown et al, Physica B 267-268, 215, 1999)*

Cr_2O_3 is a collinear antiferromagnet with zero propagation vector for which the magnetic and nuclear scattering are phase shifted by ˚90

It is anti-centrosymmetric and therefore information about 180^o antiferromagnetic domains cannot be obtained by measuring just the cross section or with uniaxial polarized neutron measurements

By cooling under various conditions of electric and magnetic fields an imbalance in domain populations is achieved - the crystal is then measured in zero field



Not only are the magnetic structures for the cooling conditions obtained - but for the first time the zero field magnetic form factor of an antiferromagnet is determined

Other studies include inelastic scattering measurements of, for example, $CuGeO_3$ *(Regnault et al, Physica B 267-268, 227, 1999)* and structural studies of complex magnetic phases e.g. Nd

Summary

Important things to remember

- The neutron cross section and the scattered polarization of the beam depends on the incident polarization
  - For magnetic scattering, components parallel to the polarization leave the spin unflipped while components perpendicular flip the spin
  - Chiral scattering and nuclear-magnetic interference may be observed
- Polarized neutron scattering can thus separate
  - Components of the cross section
  - Components of the magnetisation
- All of this can be achieved using just a few polarised neutron components
  - Polarizer/analyser, flipper, guide fields