Polarized neutron scattering

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ISIS Facility

Oxford School on Neutron Scattering 2019

Take-home message

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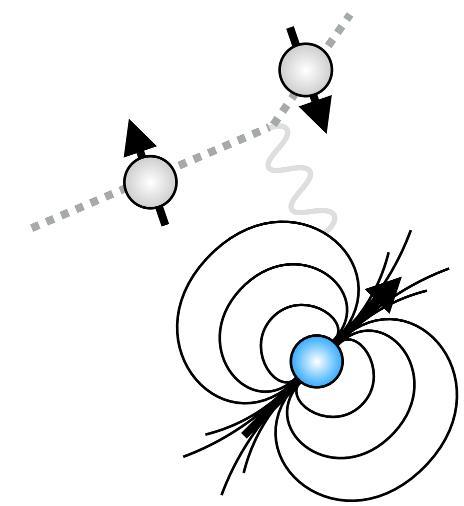
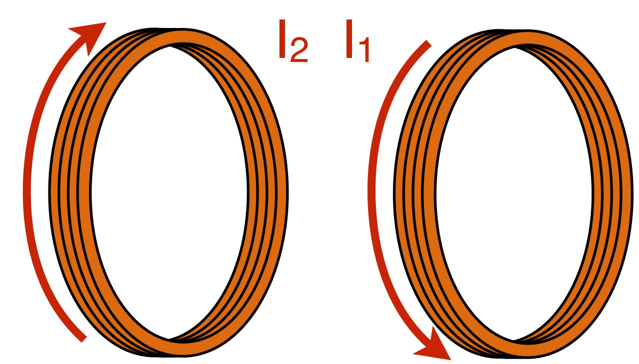
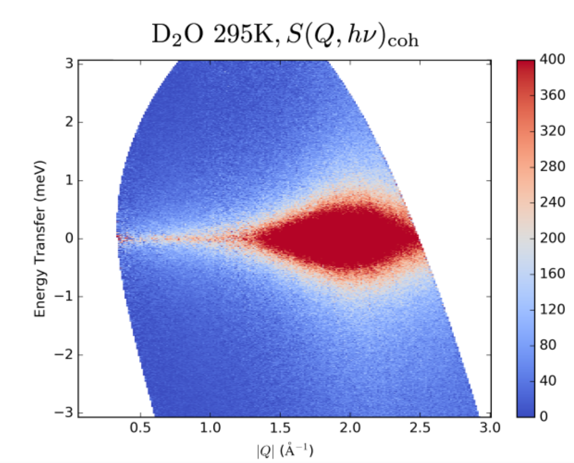
Polarized neutrons can be used to enhance (nearly) any neutron scattering experiment, either by providing additional information (this lecture), or improving the resolution or range using Larmor precession (A. Faraone)

Overview

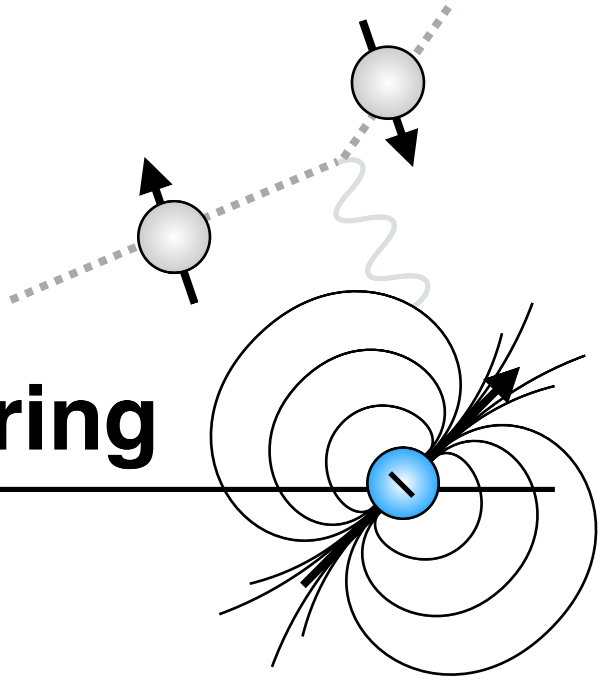
- **Principles of polarized neutron scattering**
  - What is a polarized neutron beam?
  - How do polarized neutrons interact with matter?
  - What extra information can be gained by using polarized neutrons?
- **Practical polarized neutron scattering**
  - Polarized devices: polarizers/analyzers, flippers, and guide field
- **Advanced applications of polarization analysis**
  - Magnetic diffraction and diffuse scattering
  - SANS, reflectometry spectroscopy





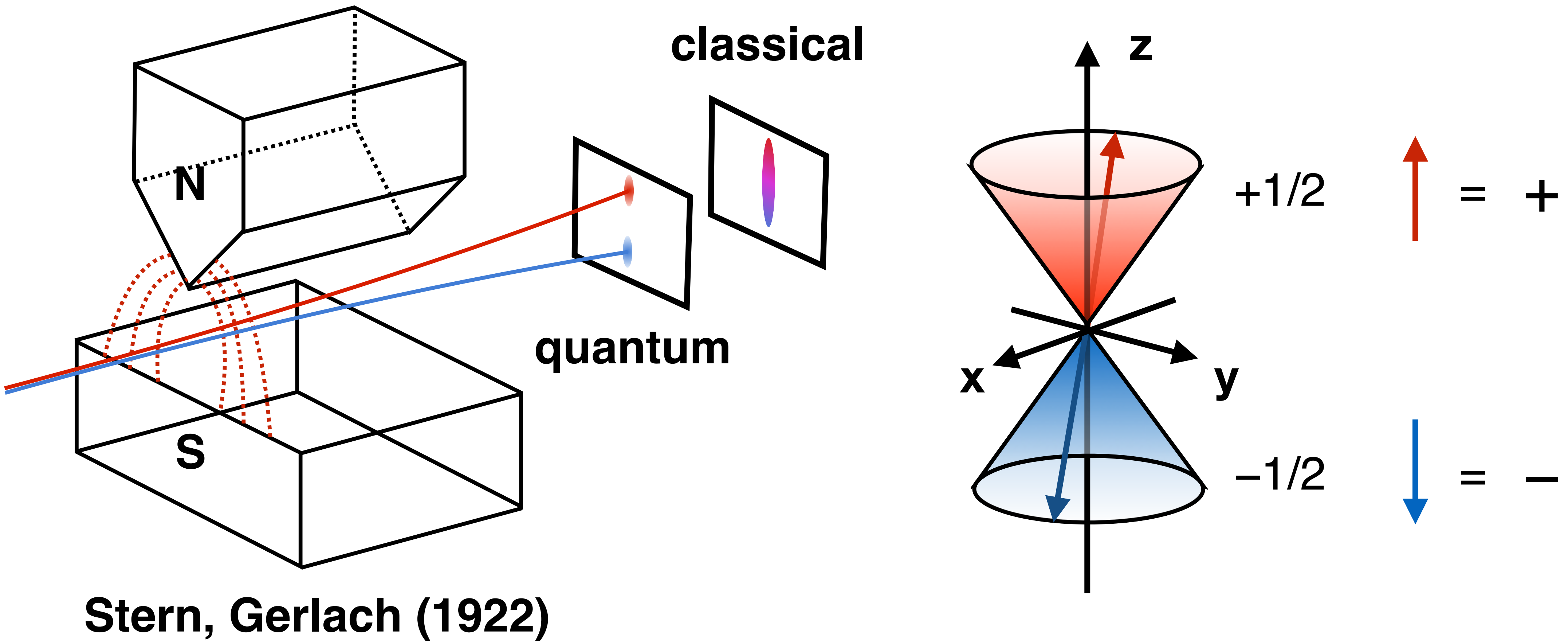


Principles of polarised neutron scattering



Spin angular momentum

Neutrons possess an inherent **magnetic moment** related to their **spin-angular momentum** S = 1/2



The **spin** has three components — *x*, *y*, and *z*. In a magnetic field, only the component along the field, conventionally *z*, is well defined.

Vector and Scalar Polarization

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In a magnetic field, the polarization of a beam is a vector pointing in the direction of the field, with the length of the vector defined as the **scalar polarization**:

$$\boxed{P = \frac{N_+ - N_-}{N_+ + N_-}} \quad \text{or} \quad \boxed{P = \frac{F-1}{F+1}; \quad F = \frac{N_+}{N_-}}$$

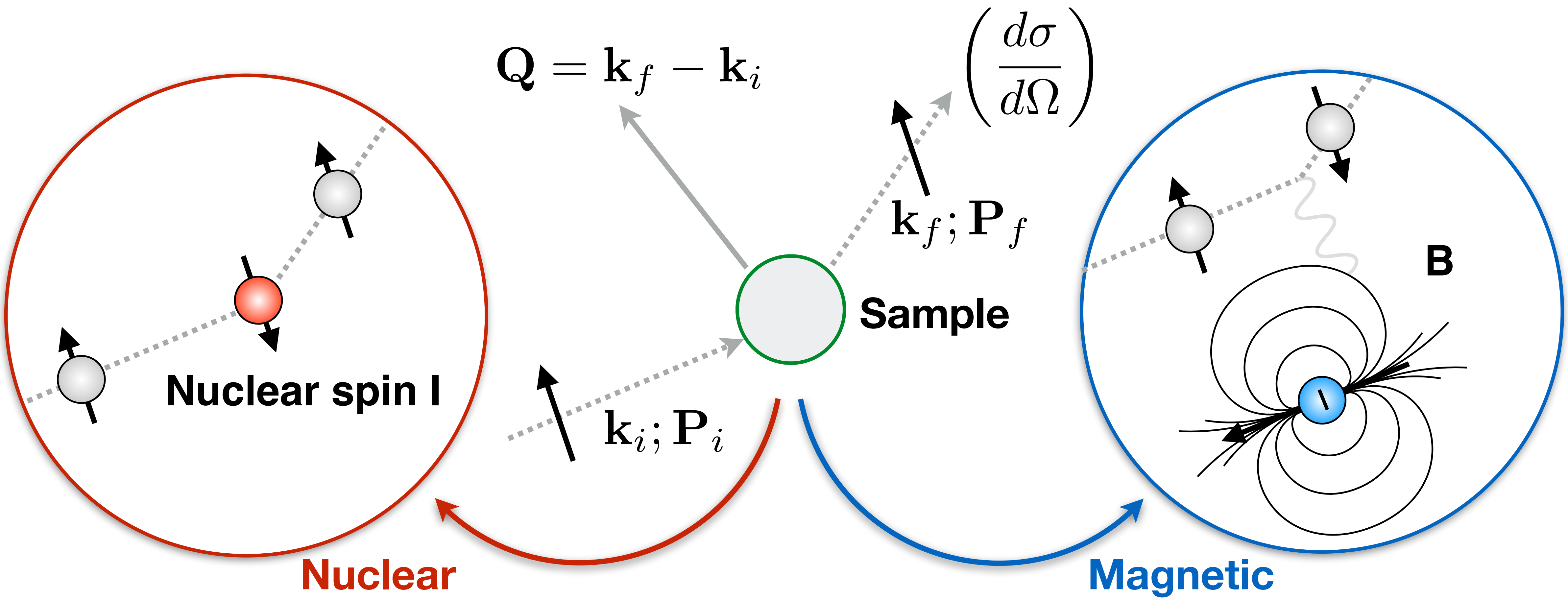
Where F is the **flipping ratio**, a frequently measured experimental quantity.

To determine the polarisation of a beam, we insert a device that selects either ↑ or ↓ from the beam (*e.g.* another SG apparatus). This is called **polarization analysis**.

$$P = \frac{1200}{3000} = 40\%; \quad F = \frac{7}{3}$$

Polarized neutron scattering

Most samples also contain magnetic moments, originating either from nuclei or the electrons — **magnetism**.



The **scattered polarization** and **cross section** (intensity) depends on the relative orientation of the beam polarization and the magnetic moments in the sample.

➔ Analyzing the scattered beam can provide us with this information!

Spin-flip and non-spin-flip scattering

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In most cases, it is sufficient to analyse the scattered polarization along the same direction as the incident. This is called **longitudinal polarization analysis**.

We then only need to consider two types of process:

Non-spin-flip (NSF)

$\downarrow \rightarrow \downarrow$ "$--$"

$\uparrow \rightarrow \uparrow$ "$++$"

Cross sections

$$\left(\frac{d\sigma}{d\Omega}\right)_{++} \qquad \left(\frac{d\sigma}{d\Omega}\right)_{--}$$

If equal: $\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{NSF}}$

Spin-flip (SF)

$\uparrow \rightarrow \downarrow$ "$+-$"

$\downarrow \rightarrow \uparrow$ "$-+$"

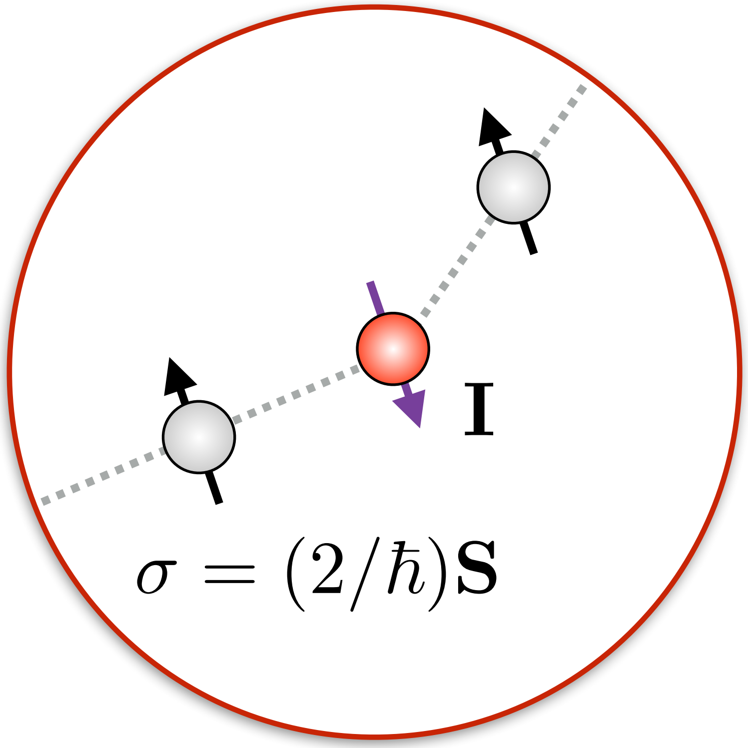
Cross sections

$$\left(\frac{d\sigma}{d\Omega}\right)_{+-} \qquad \left(\frac{d\sigma}{d\Omega}\right)_{-+}$$

If equal: $\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{SF}}$

Nuclear scattering

The neutron interacts with the nucleus via the **strong nuclear force** (Squires Ch. 9 and A. Boothroyd):



$$\mathbf{b} = \boxed{A} + \boxed{B\sigma \cdot \mathbf{I}}$$

$$b_{coh} = \overline{\mathbf{b}}; \quad b_{inc} = \sqrt{\overline{\mathbf{b}^2} - \overline{\mathbf{b}}^2}$$

Nuclear coh.
Isotope inc.

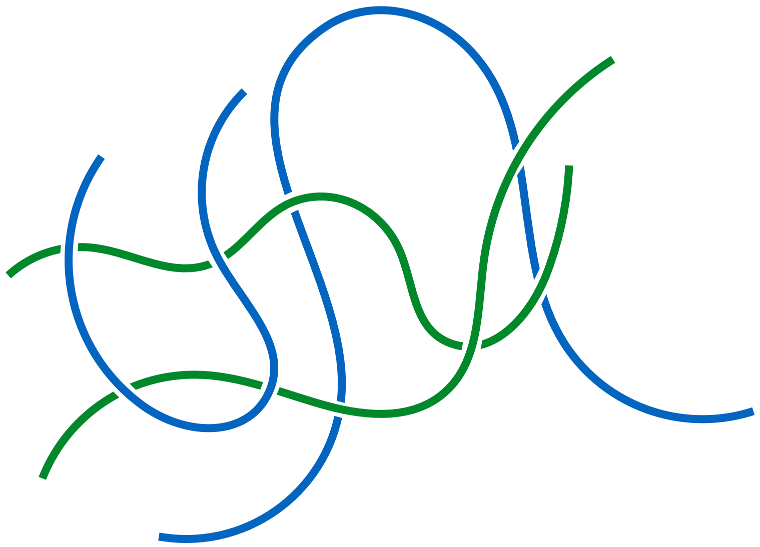
Spin inc.
$\sqrt{B^2 I(I+1)}$

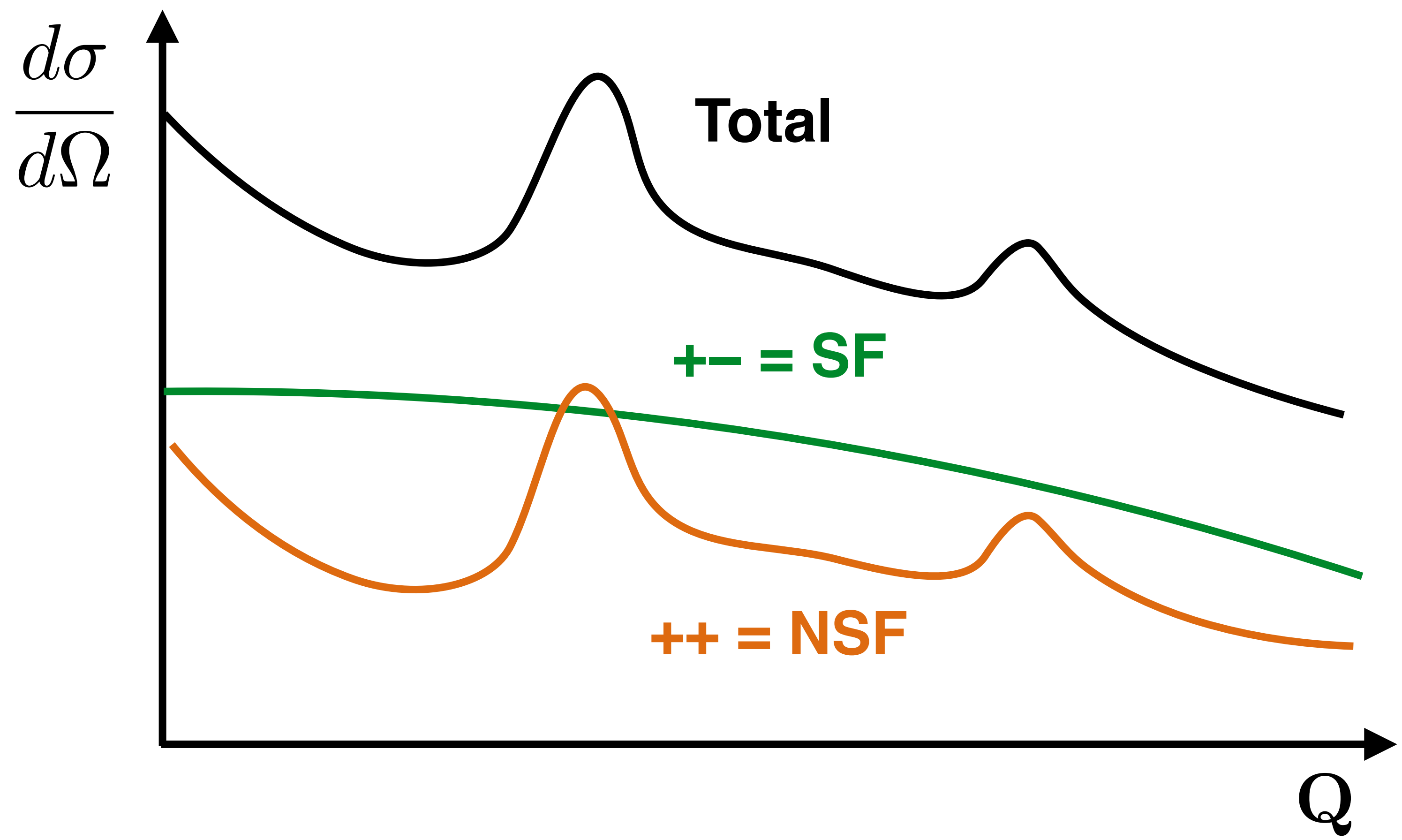
NSF: $\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{++} = \left(\frac{d\sigma}{d\Omega}\right)_{--}} = \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh+II}} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}$

SF: $\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{+-} = \left(\frac{d\sigma}{d\Omega}\right)_{-+}} = \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}$

Example 1: Polymer

Consider a hydrocarbon polymer:





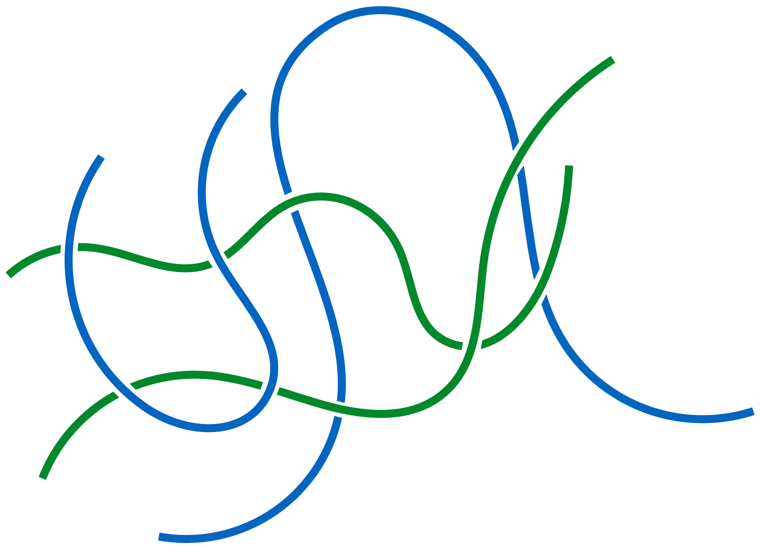
	σ_{coh}	σ_{SI}
C	**5.551**	0.001
H	**1.757**	**80.26**

If we perform longitudinal polarization analysis, we can separate the contributions:

$$\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{++}} = \boxed{\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh+II}}} + \boxed{\frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}} \qquad \boxed{\left(\frac{d\sigma}{d\Omega}\right)_{+-}} = \boxed{\frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}}$$

Example 1: Polymer

Consider a hydrocarbon polymer:



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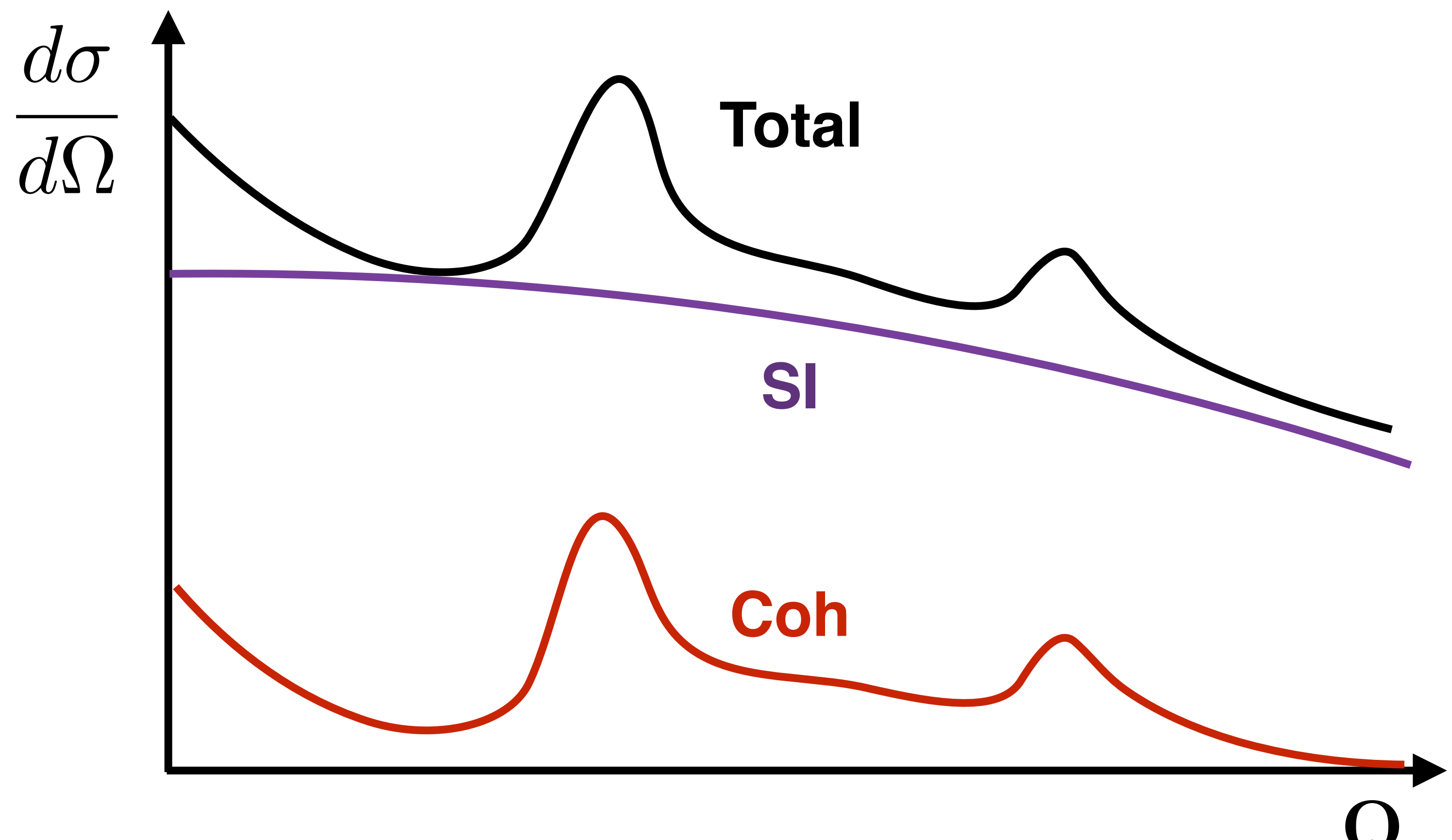

If we perform longitudinal polarization analysis, we can separate the contributions:

$$\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{coh}}} = \boxed{\left(\frac{d\sigma}{d\Omega}\right)_{++}} - \boxed{\frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{+-}} \qquad \boxed{\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}} = \boxed{\frac{3}{2}\left(\frac{d\sigma}{d\Omega}\right)_{+-}}$$

Magnetic scattering

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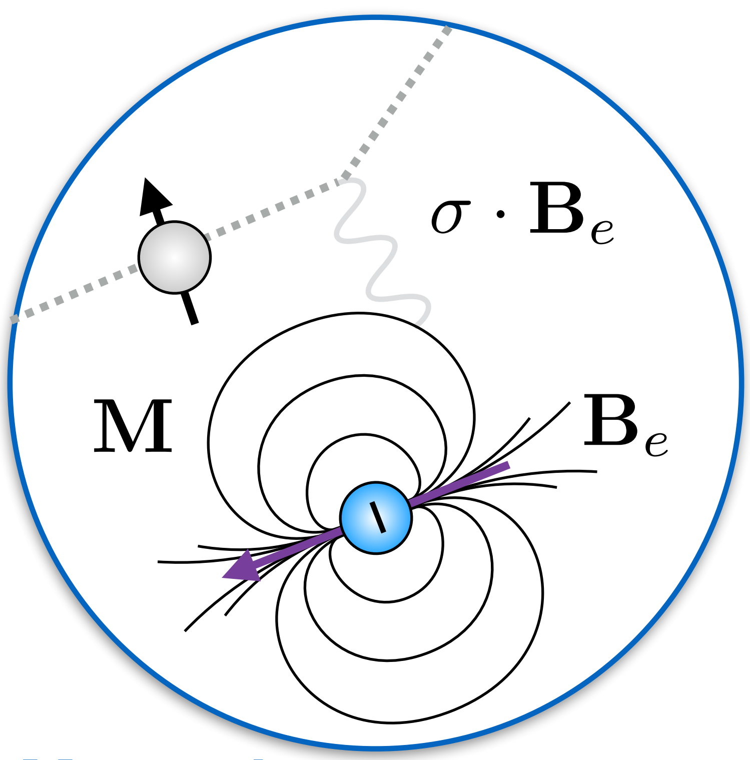
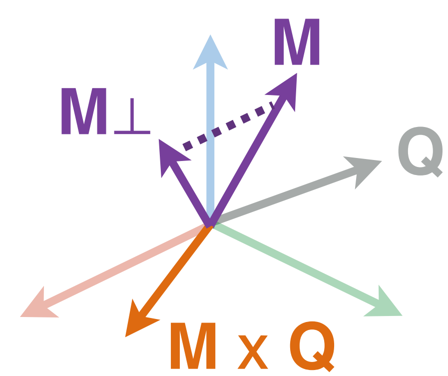
Magnetic scattering dominated by the **neutron-dipole** interaction (see N. Qureshi)



Magnetic

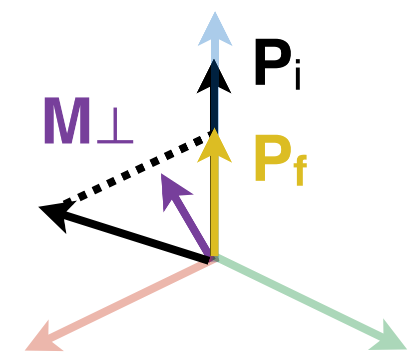
A Only measure components $\mathbf{M}\perp\mathbf{Q}$



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

Squires Ch. 7

B $\mathbf{M}_\perp \parallel \mathbf{P}_i$ - NSF
$\mathbf{M}_\perp \perp \mathbf{P}_i$ - SF



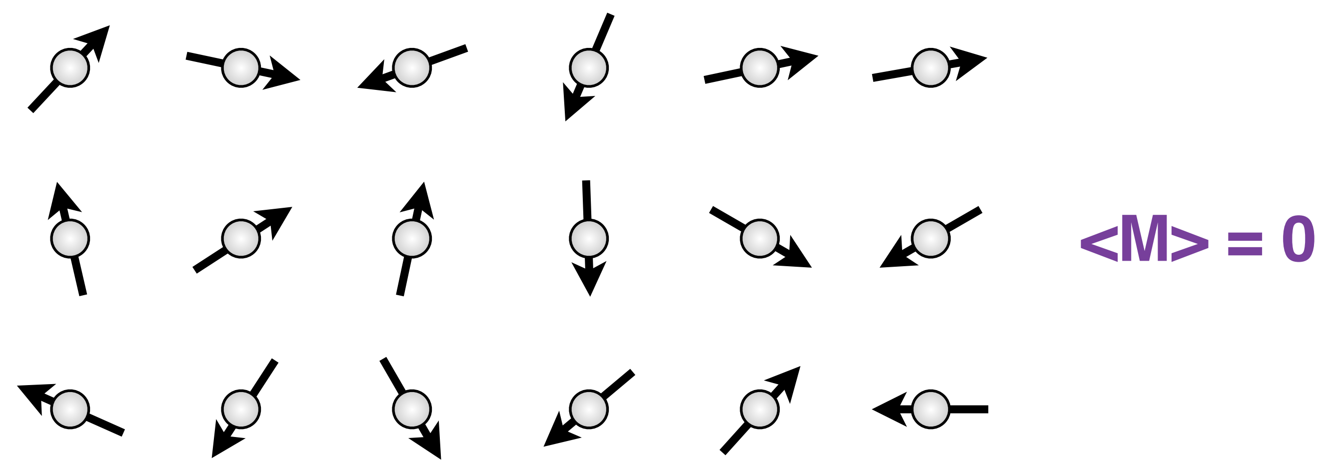
1. Rot. $\mathbf{P}_i$ 180° about **M**
2. Project onto $\mathbf{P}_i$
3. Find ratio **NSF**:**SF**

Brown, Forsyth, Tasset

This means we now have to worry about the relative directions of the sample moment (magnetisation) **M** (often ordered), **Q**, and $\mathbf{P}_i$. Complicated in general!

Example 2: Paramagnetic scattering

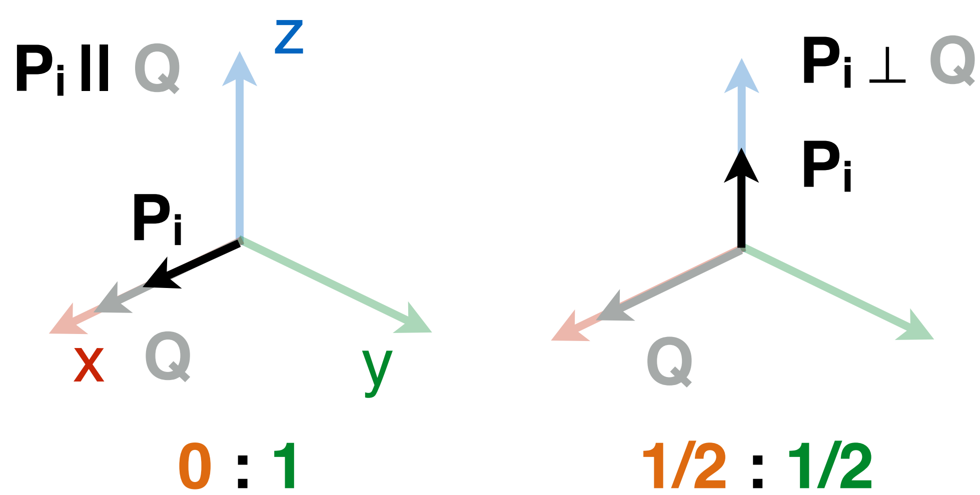
Let us consider the case where the electronic moments are disordered.



After averaging over the random direction of **M**, the magnetic elastic scattering cross section only depends on angle between the incident polarization $\mathbf{P_i}$ and **Q**:

$$\left(\frac{d\sigma}{d\Omega}\right)_{++} = \left(\frac{d\sigma}{d\Omega}\right)_{--} \propto 1 - (\hat{\mathbf{Q}} \cdot \hat{\mathbf{P}}_i)^2$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{+-} = \left(\frac{d\sigma}{d\Omega}\right)_{-+} \propto 1 + (\hat{\mathbf{Q}} \cdot \hat{\mathbf{P}}_i)^2$$

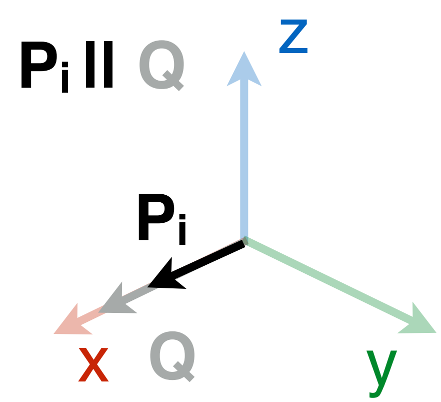


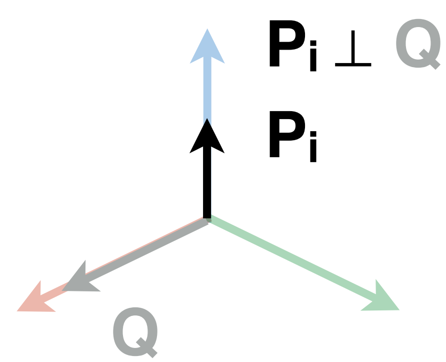
Squires Ch. 9, p. 179

Example 2: the || - ⊥ method

Combining this with example 1, what if all three types of scattering are present?

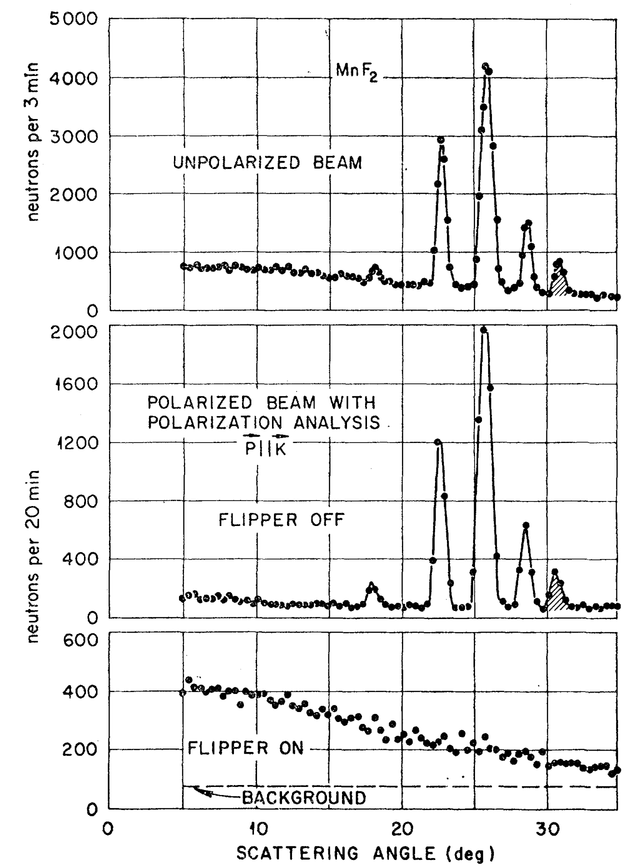
$(d\sigma/d\Omega)_{NSF}$: $(d\sigma/d\Omega)_{SF}$





	$P_i \parallel Q$	$P_i \perp Q$
Magnetic	0 : 1	1/2 : 1/2
Spin incoherent	1/3 : 2/3	1/3 : 2/3
Nuclear	1 : 0	1 : 0

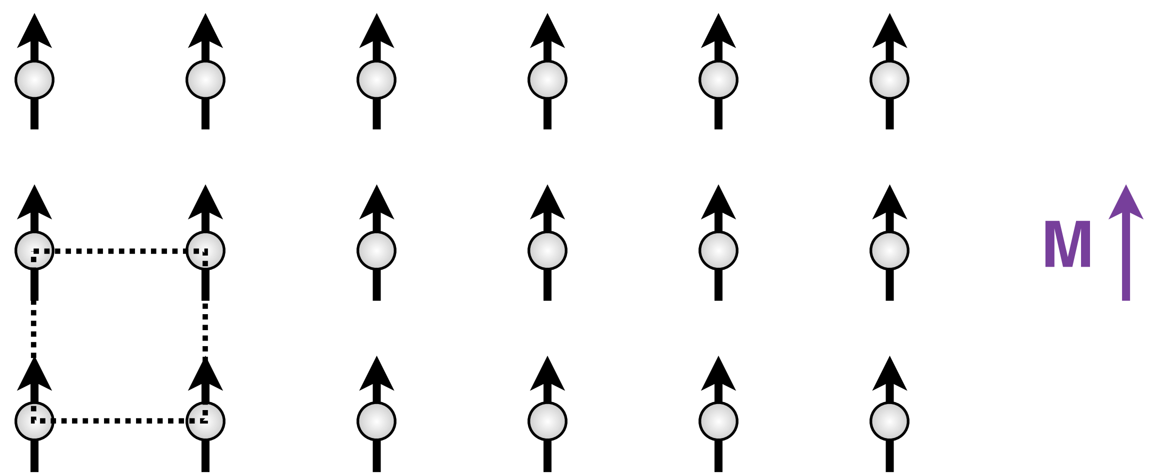
MnF2
Moon, Riste, Koehler



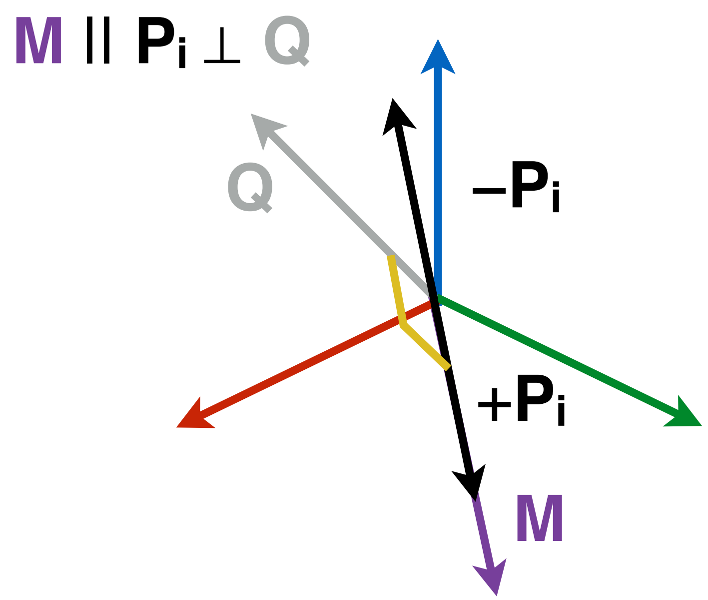
e.g.
$$\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{mag}} = 2\left[\left(\frac{d\sigma}{d\Omega}\right)^{\mathbf{P}||\mathbf{Q}}_{\mathrm{SF}} - \left(\frac{d\sigma}{d\Omega}\right)^{\mathbf{P}\perp\mathbf{Q}}_{\mathrm{SF}}\right]$$

Example 3: collinear ferromagnet

Another case involves the electronic moments in the sample all being aligned



Bragg peak cross section now depends on the orientations of the magnetisation **M**, $\mathbf{P}_\mathbf{i}$, and **Q**. It also includes **both** nuclear and magnetic contributions. For **M** II $\mathbf{P}_\mathbf{i}$ ⊥ **Q**:



1. **M** ⊥ **Q** : measure all of **M**
2. $\mathbf{P}_\mathbf{i}$ II **M**⊥ : all scattering **NSF**

+$\mathbf{P}_\mathbf{i}$ II **M** : $\left(\frac{d\sigma}{d\Omega}\right)_{++} \propto |F_N - F_M|^2$

–$\mathbf{P}_\mathbf{i}$ II **M** : $\left(\frac{d\sigma}{d\Omega}\right)_{--} \propto |F_N + F_M|^2$

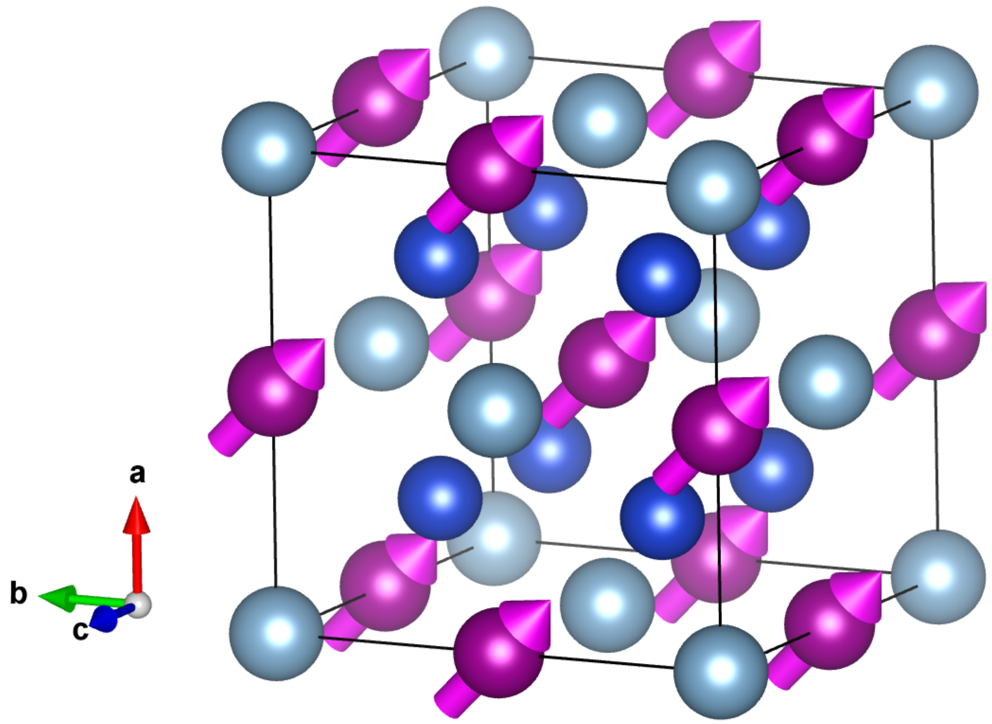
NM interference

Squires Ch. 9, p. 181

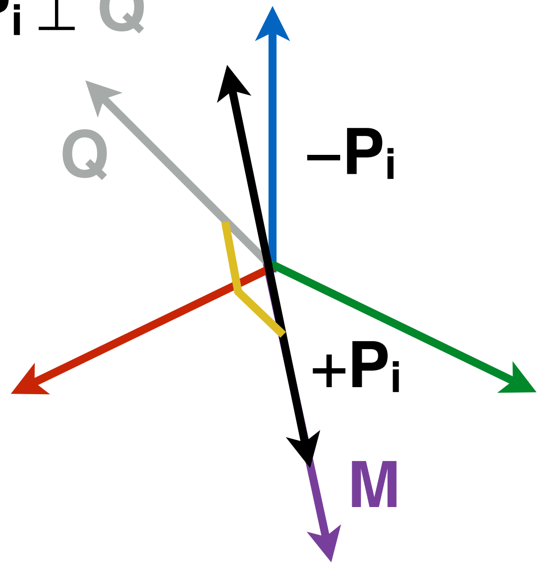
Example 3: magnetic crystal polarizer

e.g. Cu_2MnAl

M || (110)
Q = (1-11)
F_N = 7.2 fm
F_M = 6.8 fm



M || **P$_i$** ⊥ **Q**



1. **M** ⊥ **Q** : measure all of **M**
2. **P$_i$** || **M**$_\perp$: all scattering **NSF**

+**P$_i$** || **M** : $\left(\frac{d\sigma}{d\Omega}\right)_{++} \propto |F_N - F_M|^2 \sim 0.16 \text{ barns}$

–**P$_i$** || **M** : $\left(\frac{d\sigma}{d\Omega}\right)_{--} \propto |F_N + F_M|^2 \sim 200 \text{ barns}$

Summary

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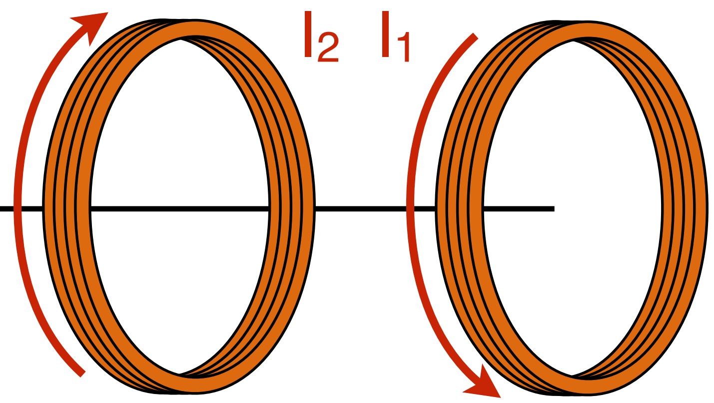
Rules

1. The **nuclear** coherent and isotope incoherent scattering is entirely NSF
2. The **spin incoherent** scattering is 1/3 **NSF** and 2/3 **SF**
3. The components of the sample **magnetisation** perpendicular to **Q** and…
    - … parallel to $\mathbf{P}_i$: **NSF**
    - … perpendicular to $\mathbf{P}_i$: **SF**

Consequences

1. We can separate the components of the cross section (Examples 1,2)
2. We are also sensitive to the direction of magnetic moments

Practical polarised neuton scattering



What do we need?

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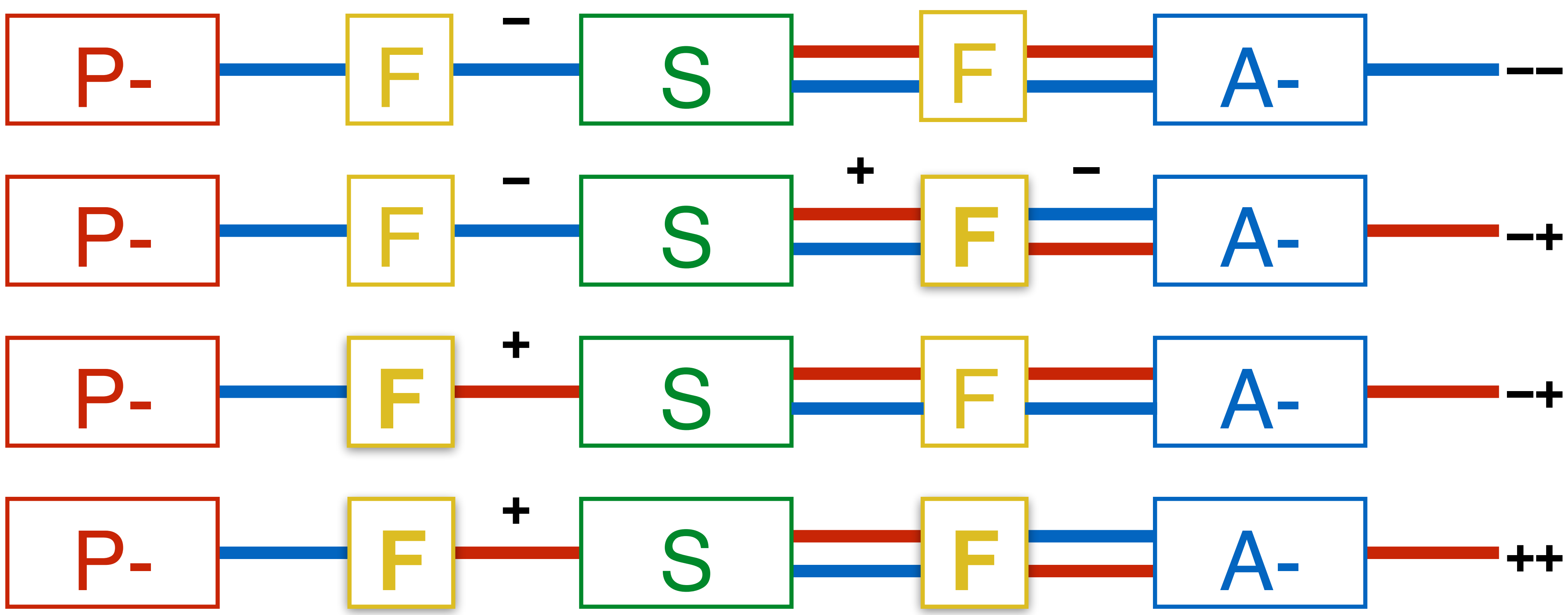
We've seen that we can polarise and analyse a beam with crystals like Cu_2MnAl. However, these are normally fixed to accept only one state — need **flippers**



We have also seen that it can be useful to rotate the polarisation versus Q and M — **guide field**. The guide field also preserves the polarisation between the elements.

Polarized neutrons in practice

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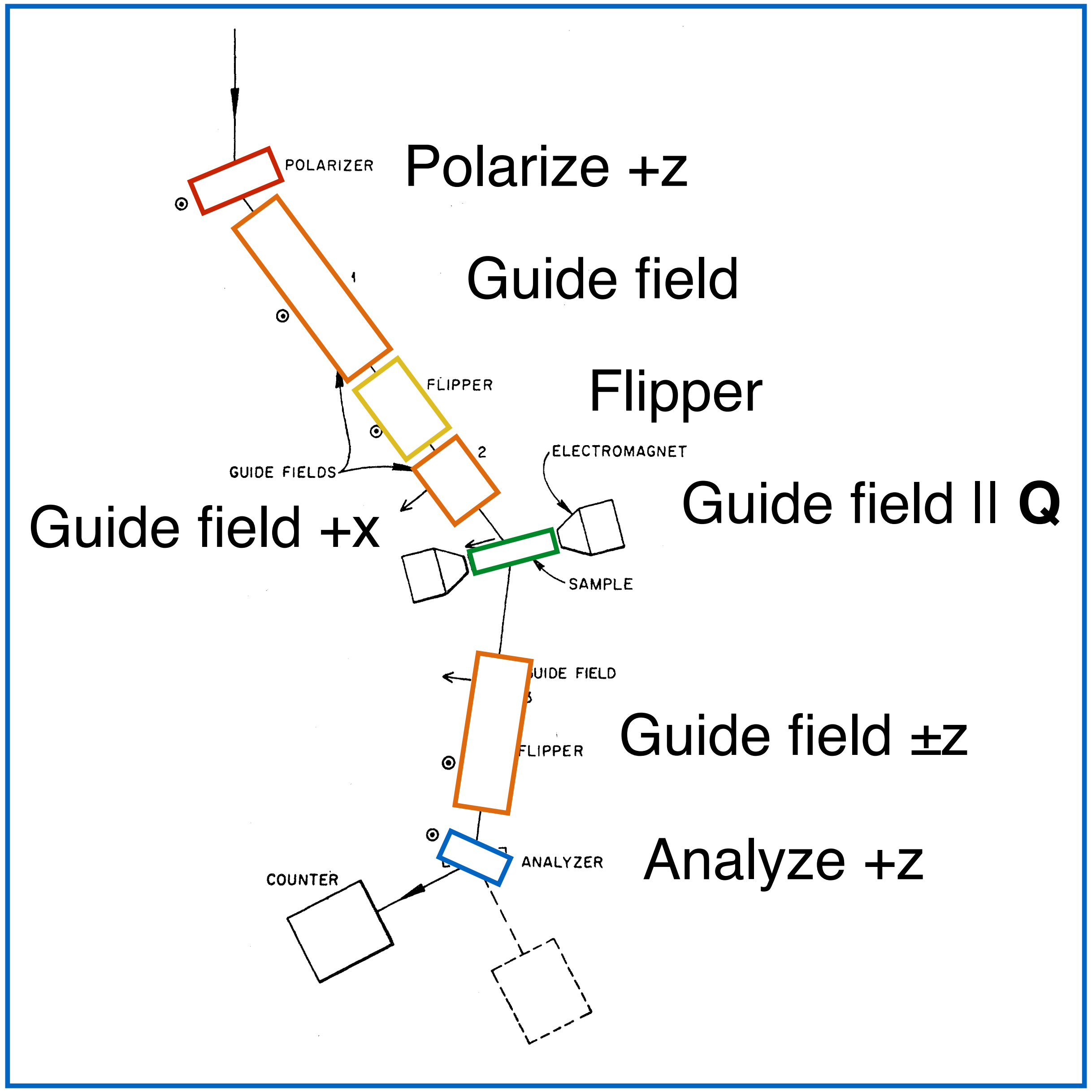
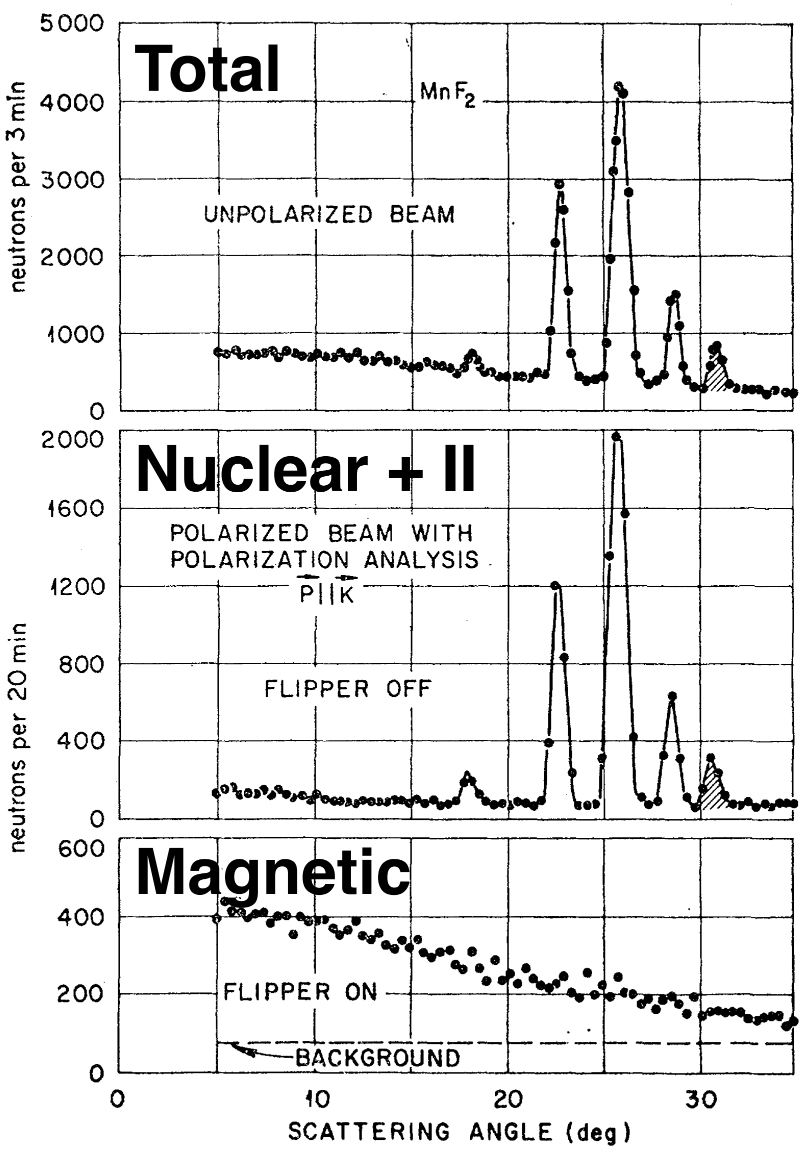
The first instrument of this kind was built by Moon, Riste, and Koehler in 1968

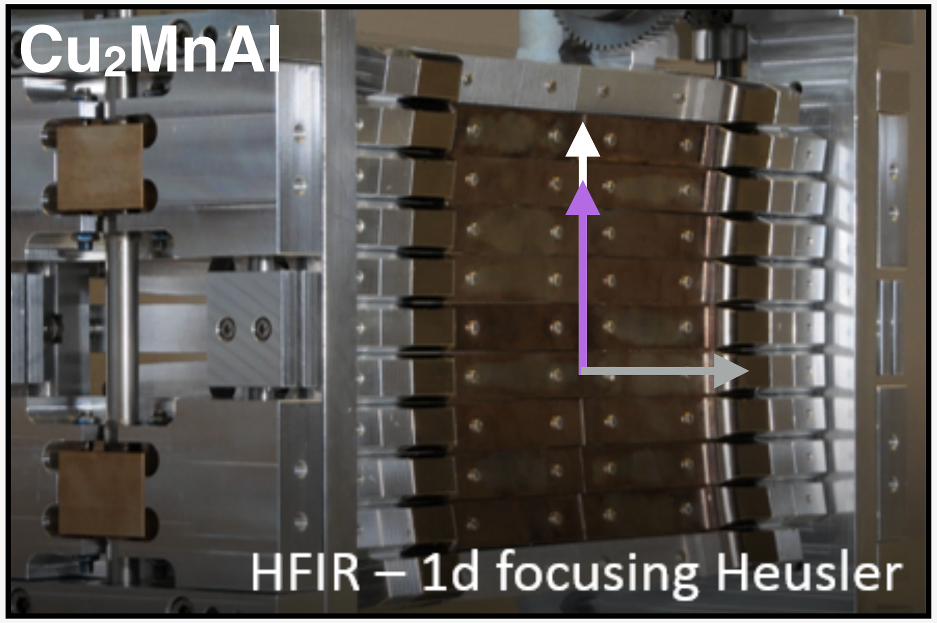




Moon, Riste, Koehler

Neutron polarizers and analyzers

1. Magnetic crystal



$$\left(\frac{d\sigma}{d\Omega}\right)_{\pm\pm} \propto |F_N \mp F_M|^2$$

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

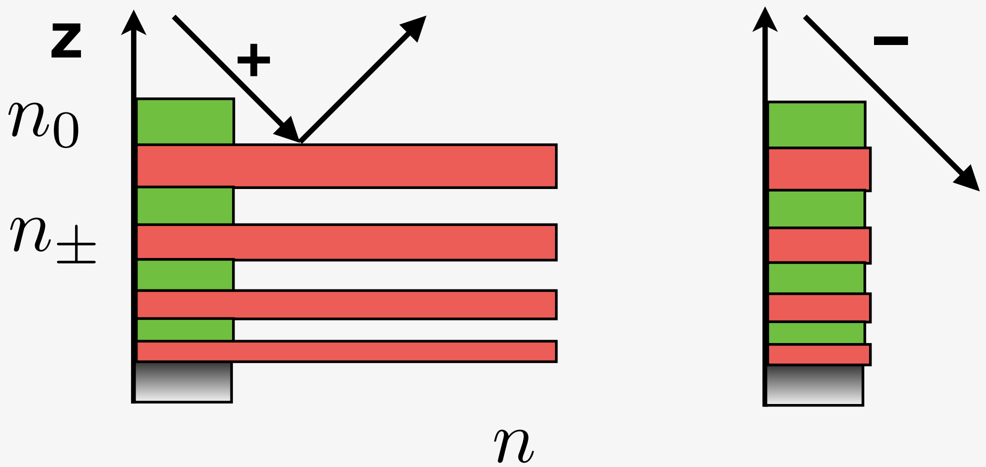
If $F_N = F_M$, polarized beam!

(see Example 3)

2. Polarizing mirrors

Alternating nonmagnetic and magnetic layers

$$n_\pm \propto \sqrt{\rho_{\mathrm{coh}} \mp \rho_{\mathrm{mag}}}$$



Reflectivity at the interface:

$$R = \left(\frac{n_0 - n_\pm}{n_0 + n_\pm}\right)^2$$

If $n_0 = |n_\pm|$, polarized beam!

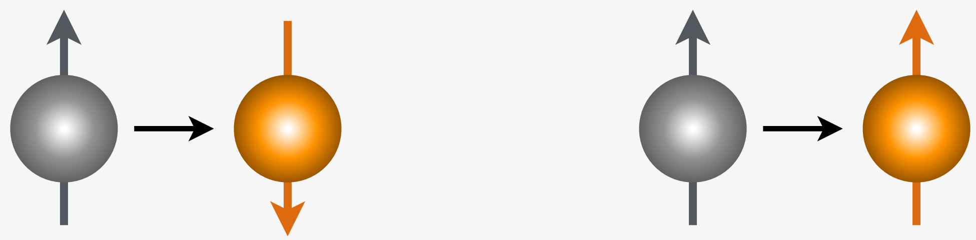
(see S. Langridge lecture)

Neutron polarizers and analyzers

3. ^{3}He spin filter

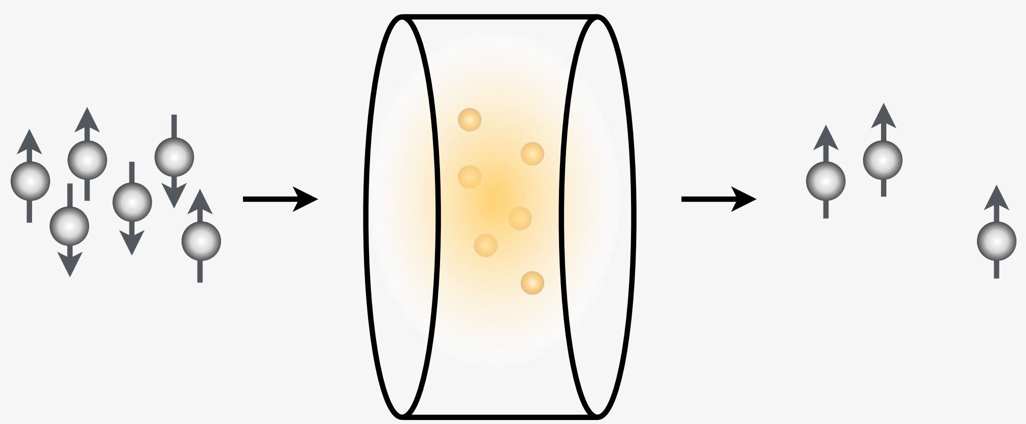
^{3}He (nuclear spin I = 1/2) has a spin-dependent absorption cross section:

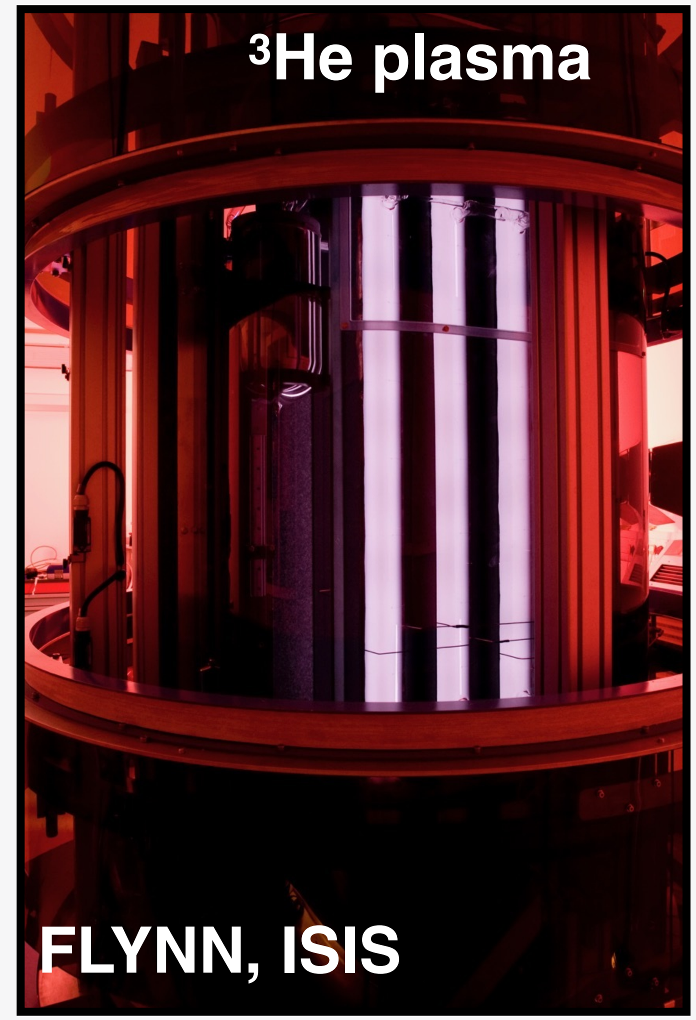
neutron ^{3}He



$\sigma_{\mathrm{abs}} \sim 6000$ barns $\qquad \sigma_{\mathrm{abs}} \sim 0$ barns

unpolarized beam | spin filter ~1 bar ^{3}He | polarized beam



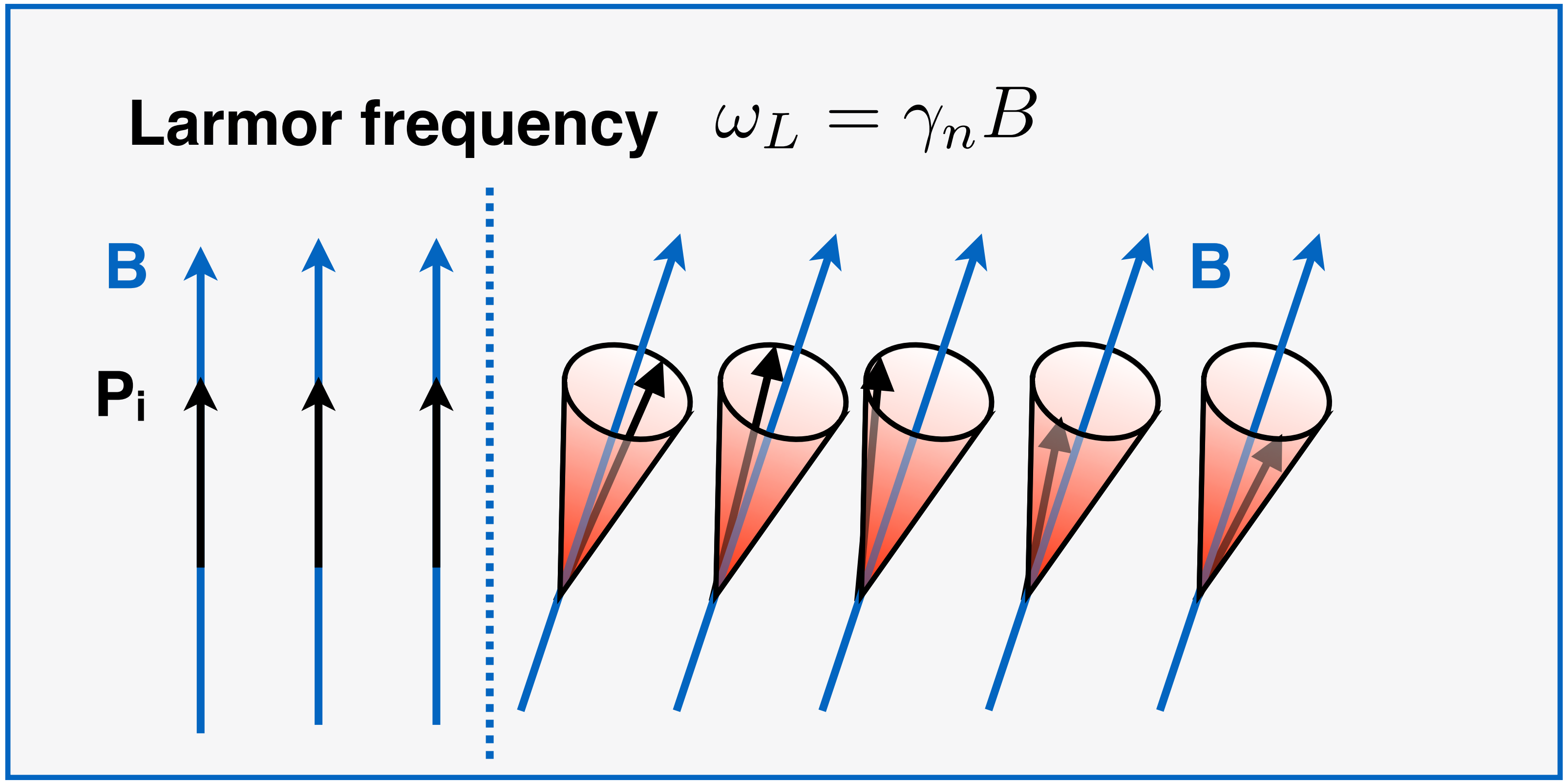


Require high ^{3}He polarization for good neutron polarization → lasers!

Manipulating the polarization

After creating polarised beam, need to **guide/rotate** it and **flip** its direction. This is done using magnetic fields.

If the direction of the magnetic field changes, the polarization **Larmor precesses** around the new field direction.



The angle of the cone depends on the angle between the original field direction and the new field direction.

Manipulating the polarization

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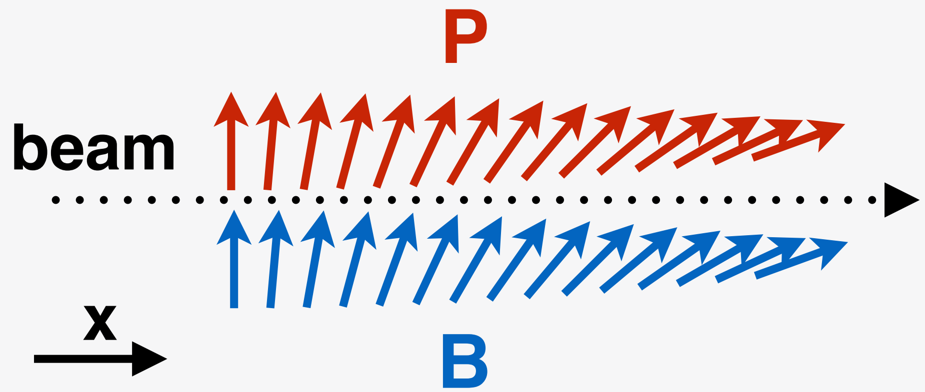
Let us imagine we have a field changing at a rate $\omega_B = d\theta_B/dt$. We may then identify two cases by comparing this rate with the Larmor frequency and neutron velocity:

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

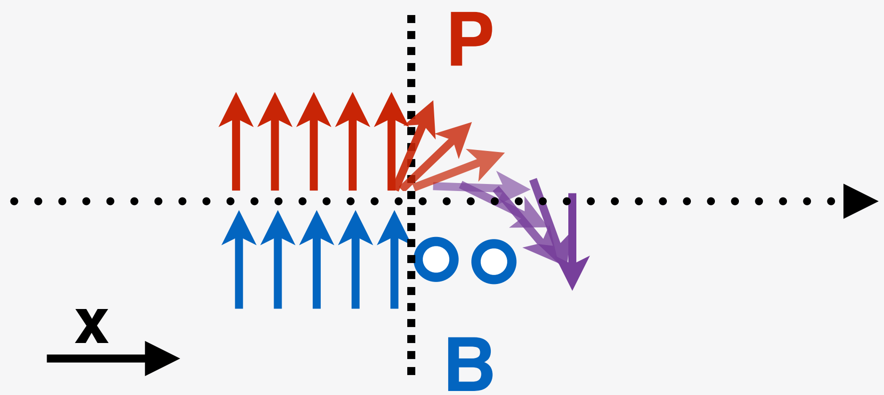
Adiabatic (A > 10)

The spin follows the rotating field direction



Non-adiabatic (A < 0.1)

The spin immediately begins precessing about the new direction



Slow changes → field rotation. Fast changes → precession/flipping

Guide fields/field rotators

Guide/rotating field is typically constructed using either permanent magnets or electromagnets:

XYZ field rotator

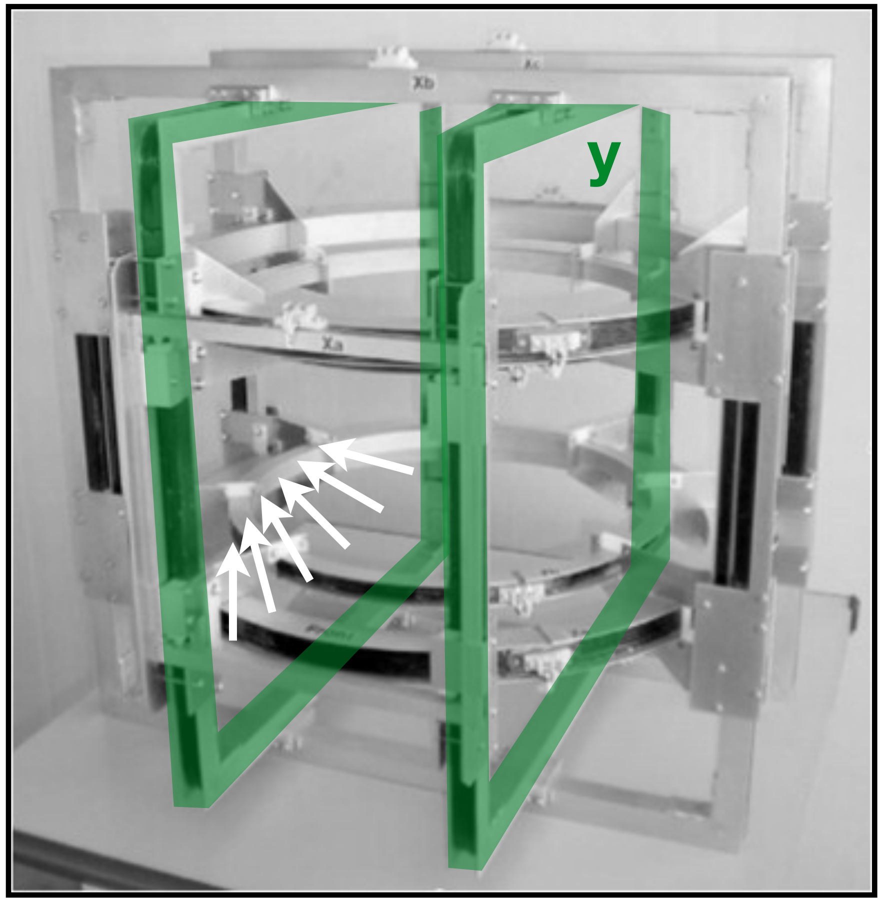


Photo: R. Stewart

Guide field

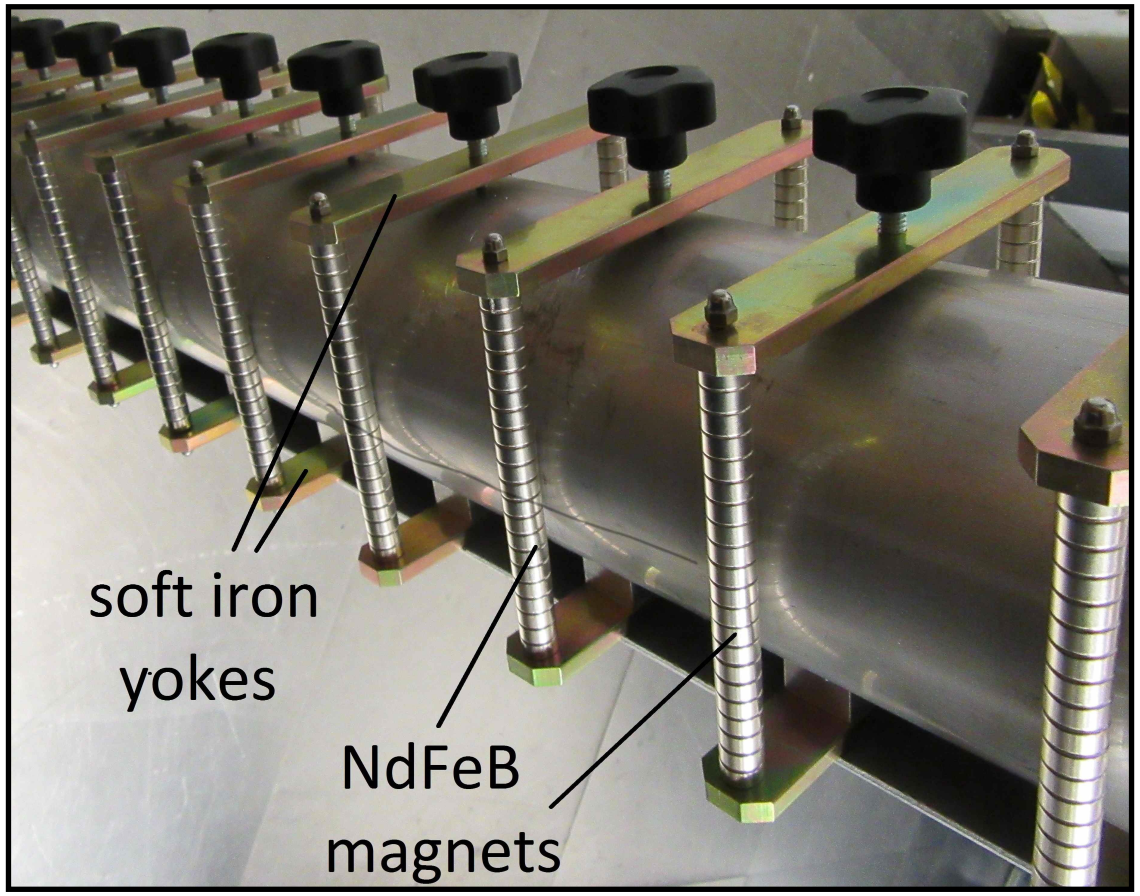
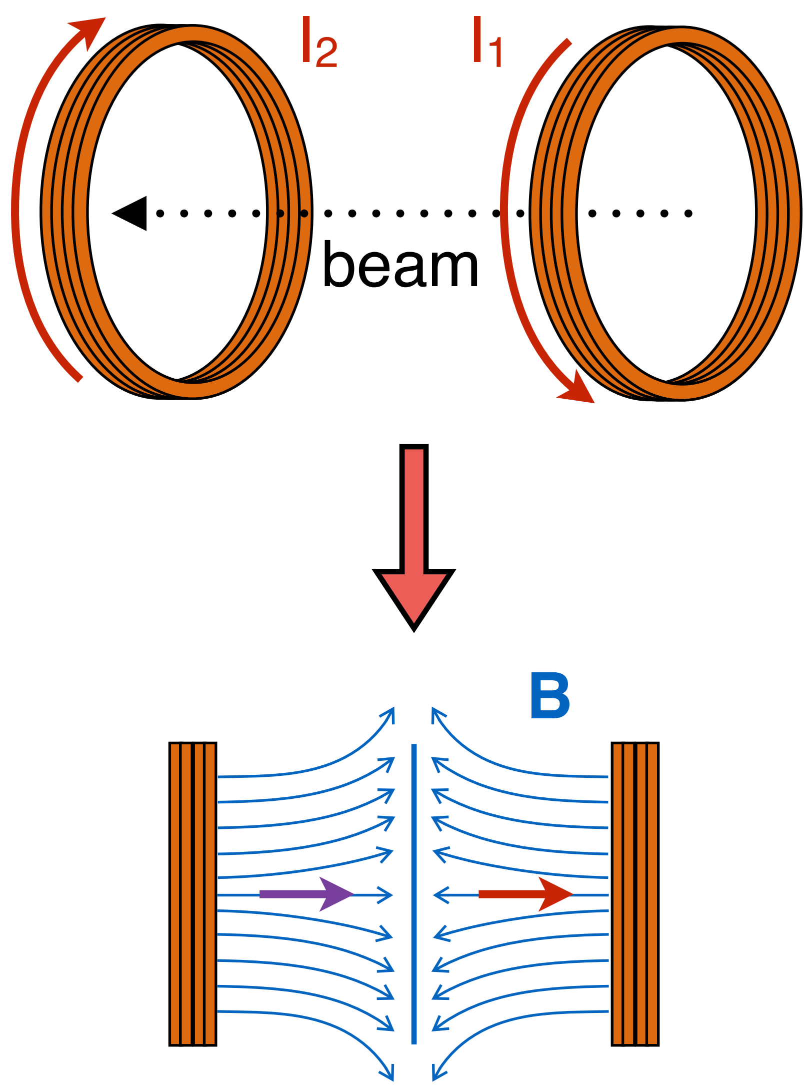


Photo: J. Kosata

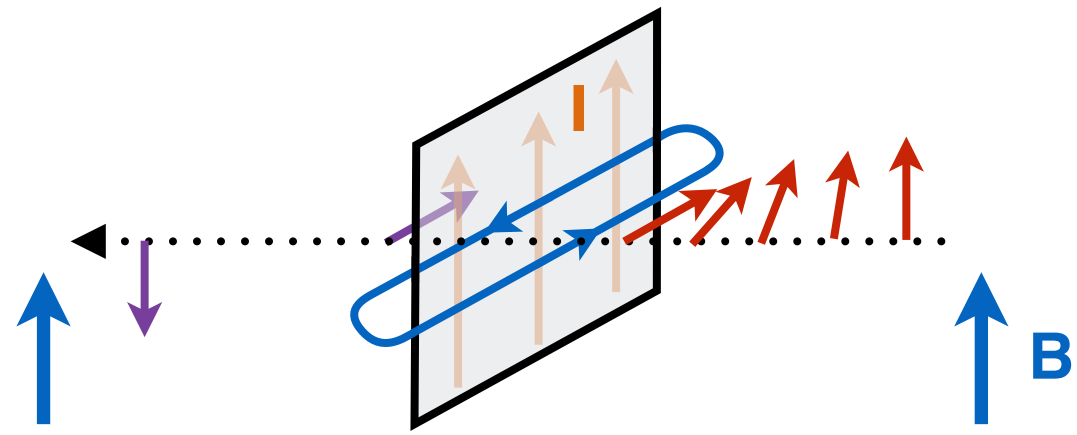
Non-adiabatic spin flippers

Drabkin

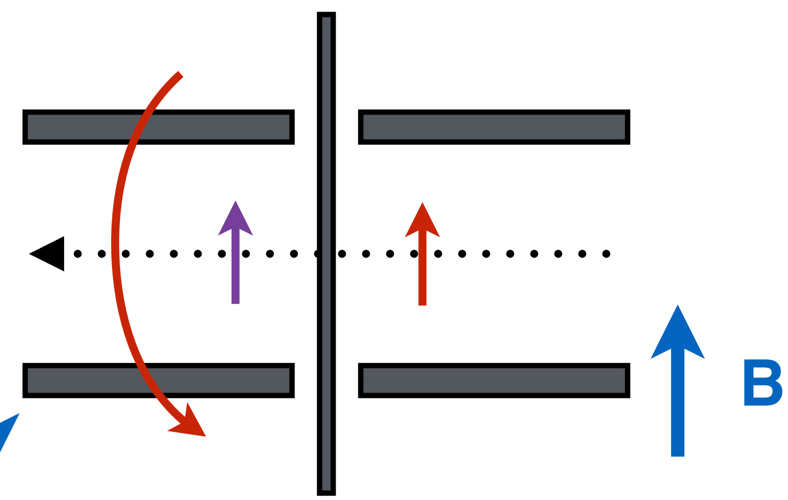


Field changes direction in the middle.

Dabbs foil



Cryoflipper

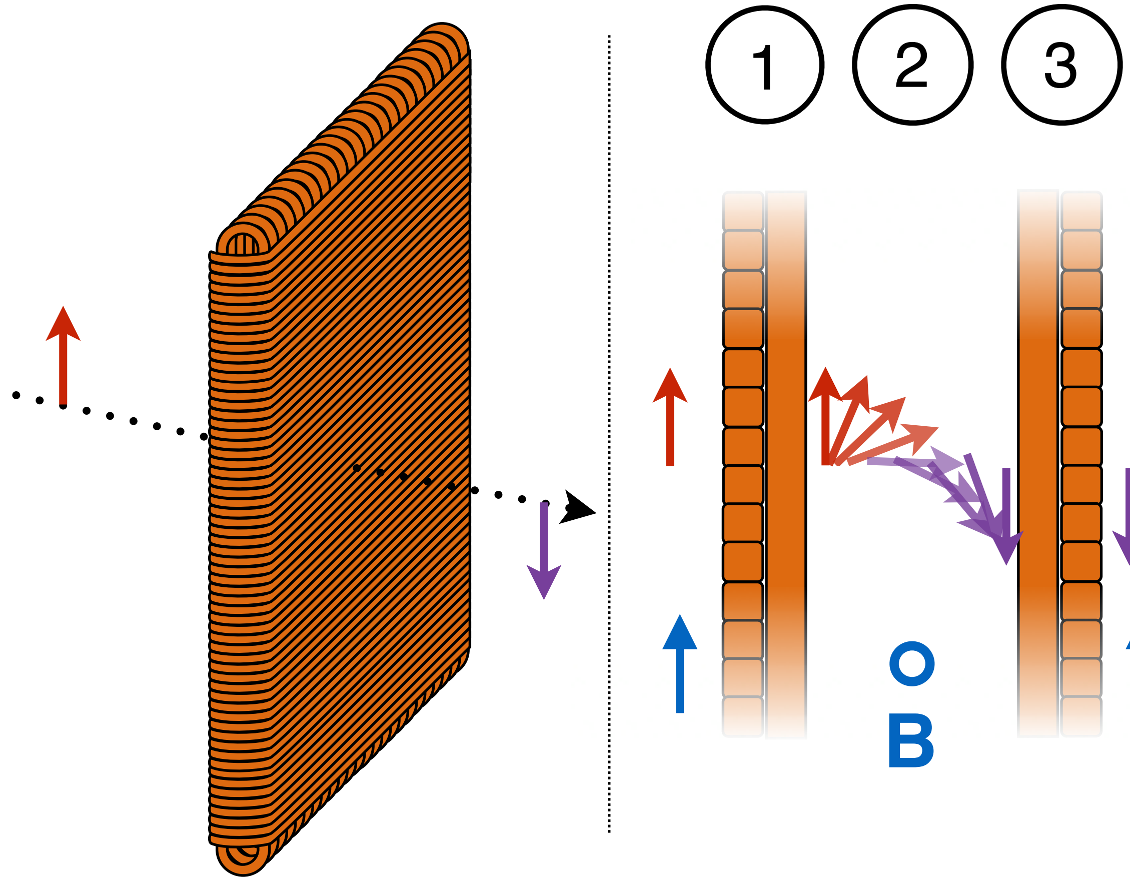


Meissner screen (Nb or YBCO)

Other types of spin flipper

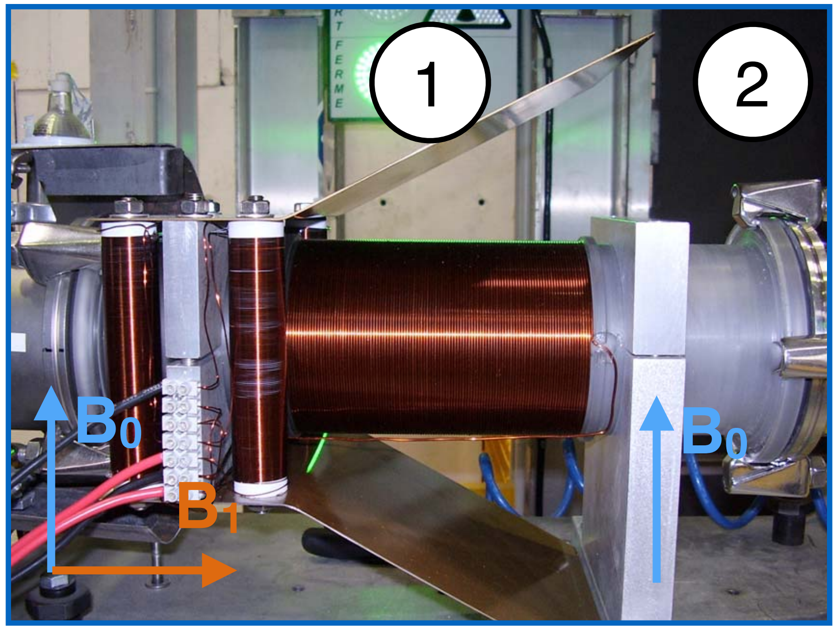
Alternatively, we can use Larmor precession combined with non-adiabatic trans.

Mezei



1. Non-adiabatic transition
2. Half a precession (π)
3. Non-adiabatic transition

Adiabatic Fast Passage

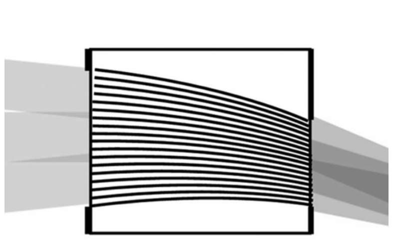


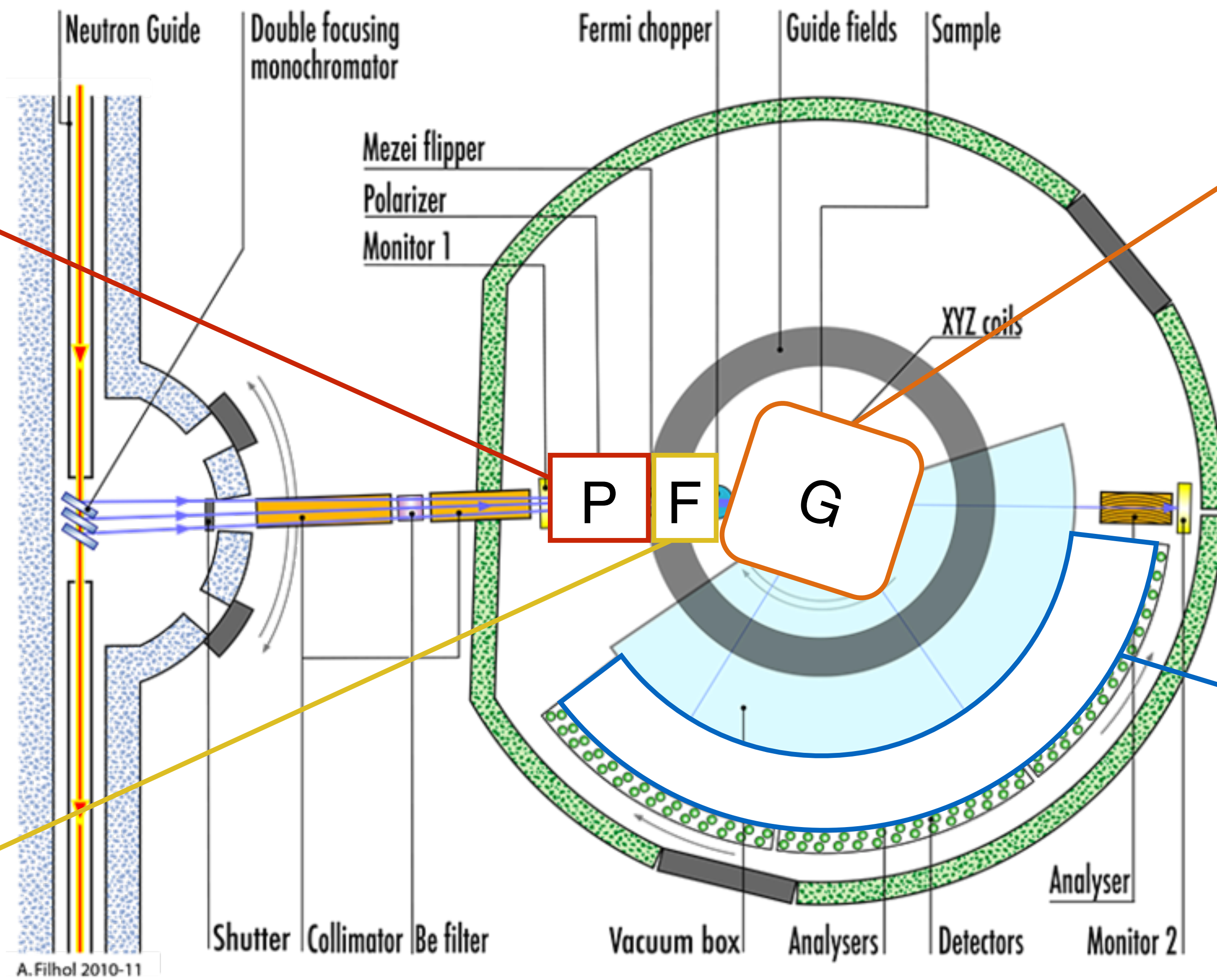
$$\mathbf{B}_{tot} = \left(B_0 + \frac{\omega}{\gamma}\right)\hat{z} + B_1\hat{x}$$

1. Reversal of B_{tot} with RF field
2. Non-adiabatic transition

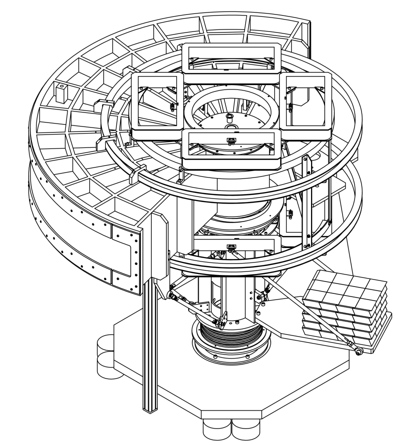
Example instrument: D7, ILL

Polarizer
Supermirror bender





Guide field
XYZ



Flipper
Mezei

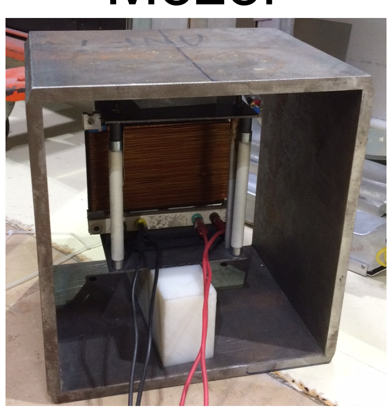
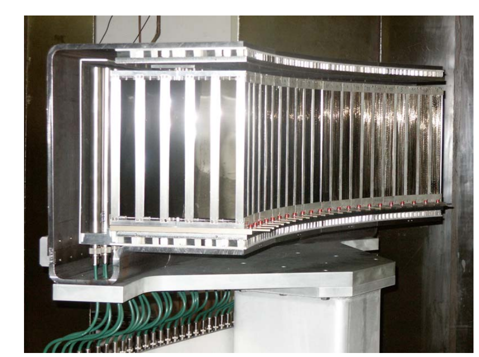


photo: A. Wildes

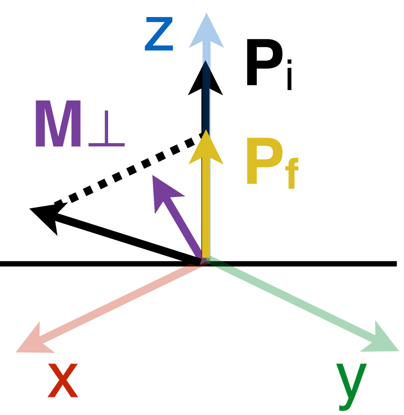
Analyzer
Supermirror



(wide angle rare)

Stewart et. al.; Schärpf

Advanced polarised neutron scattering: Generalising polarisation analysis



Reminder: rules of polarised neutron scattering

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Nuclear

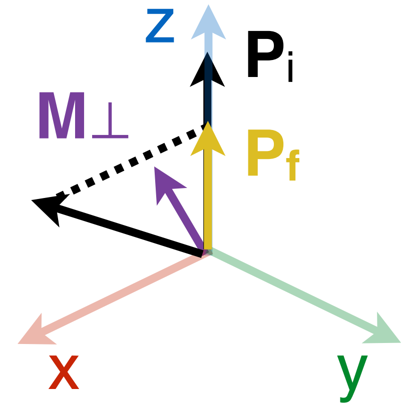
1. The nuclear coherent and isotope incoherent scattering is entirely NSF

2. The spin incoherent scattering is 1/3 **NSF** and 2/3 **SF**

Magnetic

3. The components of the sample magnetisation perpendicular to **Q** and…

- … parallel to $\mathbf{P}_i$: **NSF**
- … perpendicular to $\mathbf{P}_i$: **SF**



Reminder: The || - ⊥ method

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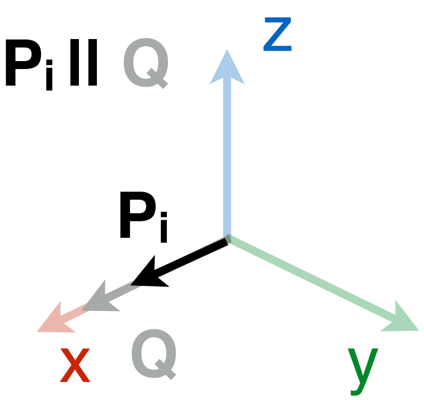
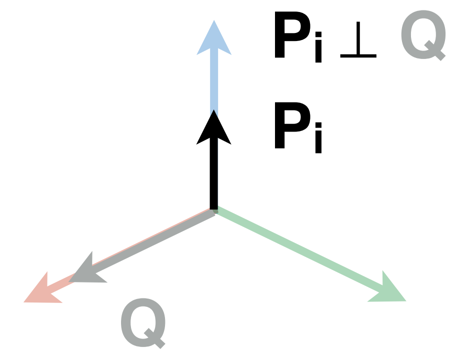
$(d\sigma/d\Omega)_{NSF}$: $(d\sigma/d\Omega)_{SF}$





	$P_i \parallel Q$	$P_i \perp Q$
Magnetic	0 : 1	1/2 : 1/2
Spin incoherent	1/3 : 2/3	1/3 : 2/3
Nuclear	1 : 0	1 : 0

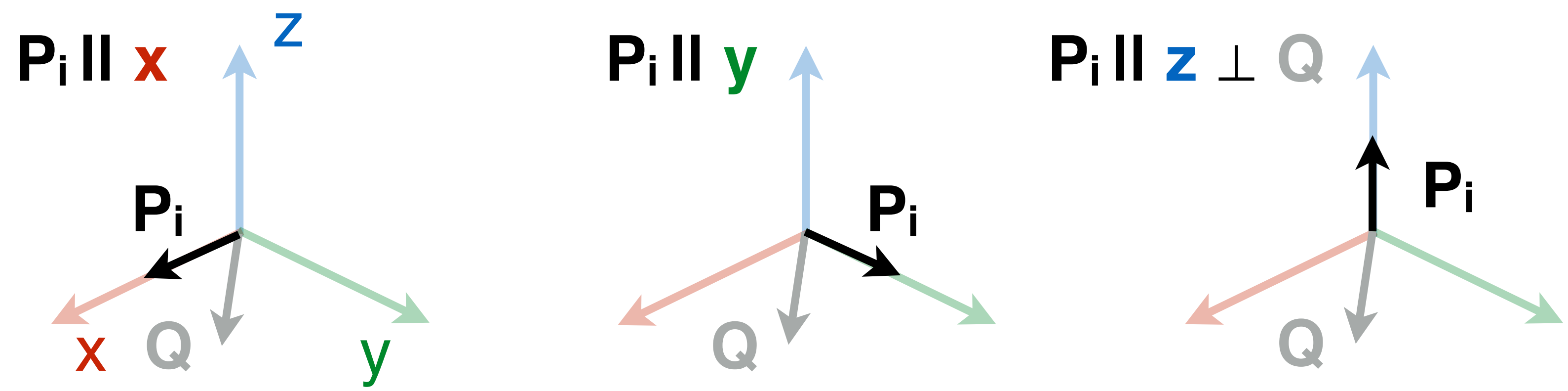
e.g.

$$\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{mag}} = 2\left[\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{SF}}^{\mathbf{P}||\mathbf{Q}} - \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{SF}}^{\mathbf{P}\perp\mathbf{Q}}\right]$$

2D XYZ polarization analysis

In the case where we have a 2D detector, like in a powder diffractometer (*e.g.* D7), it is no longer possible to align **Q** and $\mathbf{P}_i$ for every detector. However:

$(d\sigma/d\Omega)_{NSF} : (d\sigma/d\Omega)_{SF}$



	$\mathbf{P}_i \parallel x$	$\mathbf{P}_i \parallel y$	$\mathbf{P}_i \parallel z \perp Q$
Magnetic	$1/2(\cos^2\alpha) : 1/2(\cos^2\alpha+1)$	$: 1/2(\sin^2\alpha+1)$	$1/2 : 1/2$
Spin incoherent	$1/3 : 2/3$	$1/3 : 2/3$	$1/3 : 2/3$
Nuclear	$1 : 0$	$1 : 0$	$1 : 0$

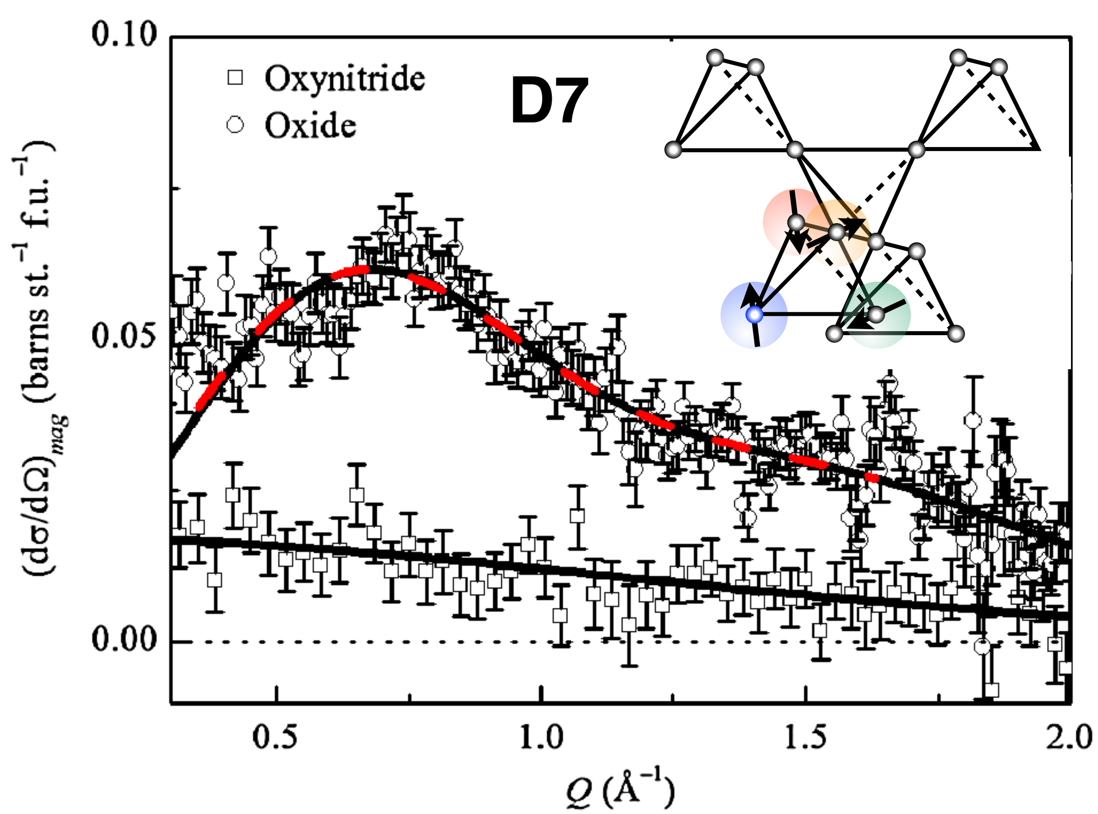
e.g.

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

Examples: 2D XYZ PA

This technique can be used to separate very small signals in magnetically disordered powders (scatter like paramagnets):

Frustrated magnets

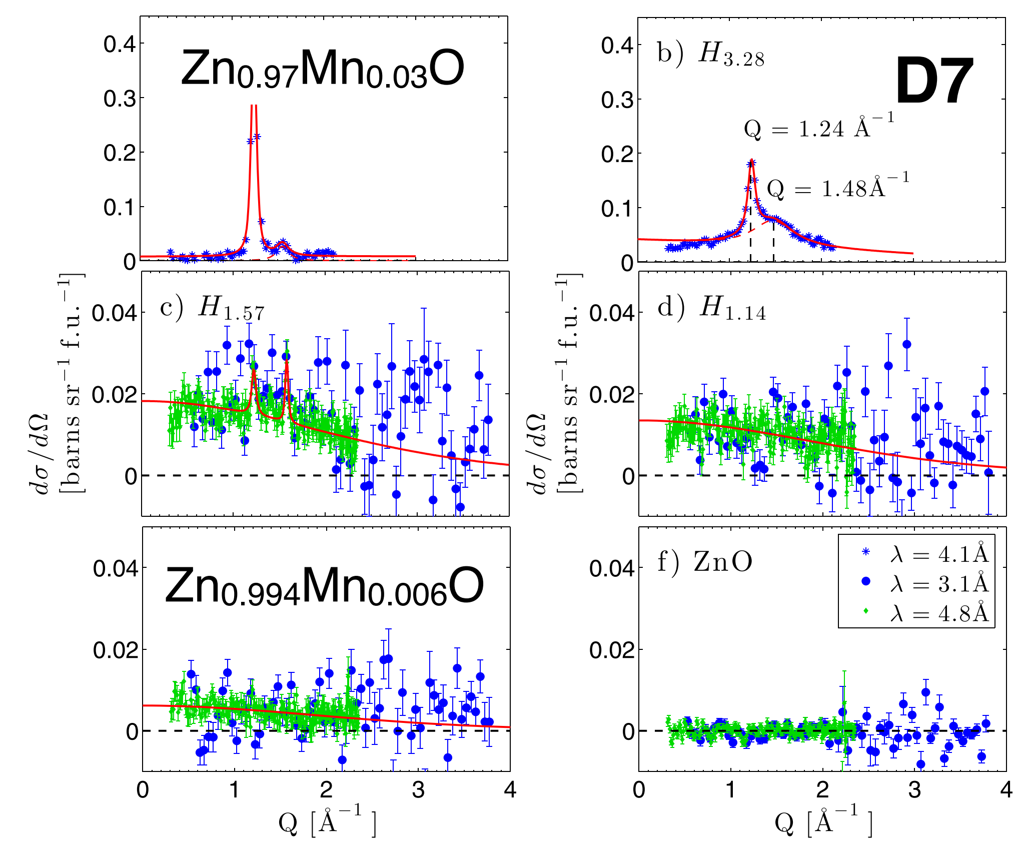


Magnetic frustration destroys magnetic order, only short range order

$Lu_2Mo_2O_5N_2$: **6% of S = 1/2!**

Clark et. al.

Magnetic semiconductors



Is the RT ferromagnetism intrinsic?

No! Clusters at high concentrations.

Lancon et. al.

XYZ LPA: Magnetic single crystals

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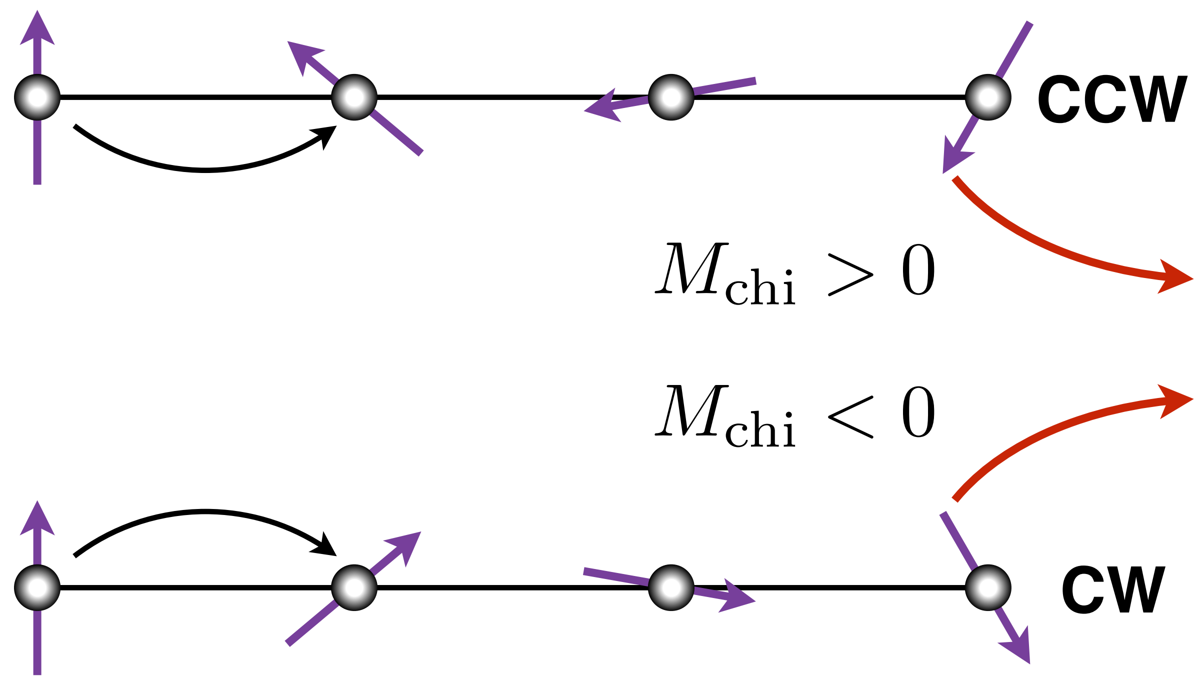
If the scattering is not paramagnetic-like, we're back to having to consider the directions of **Q**, **M**, and $\mathbf{P}_i$. This is usually the case for single crystals.

Other complications we may encounter are the presence of **nuclear-magnetic interference** (Example 3), and **chiral scattering** for non-collinear structures:



$M_{\mathrm{chi}} > 0$

$M_{\mathrm{chi}} < 0$

SF cross section || **Q** contains handedness

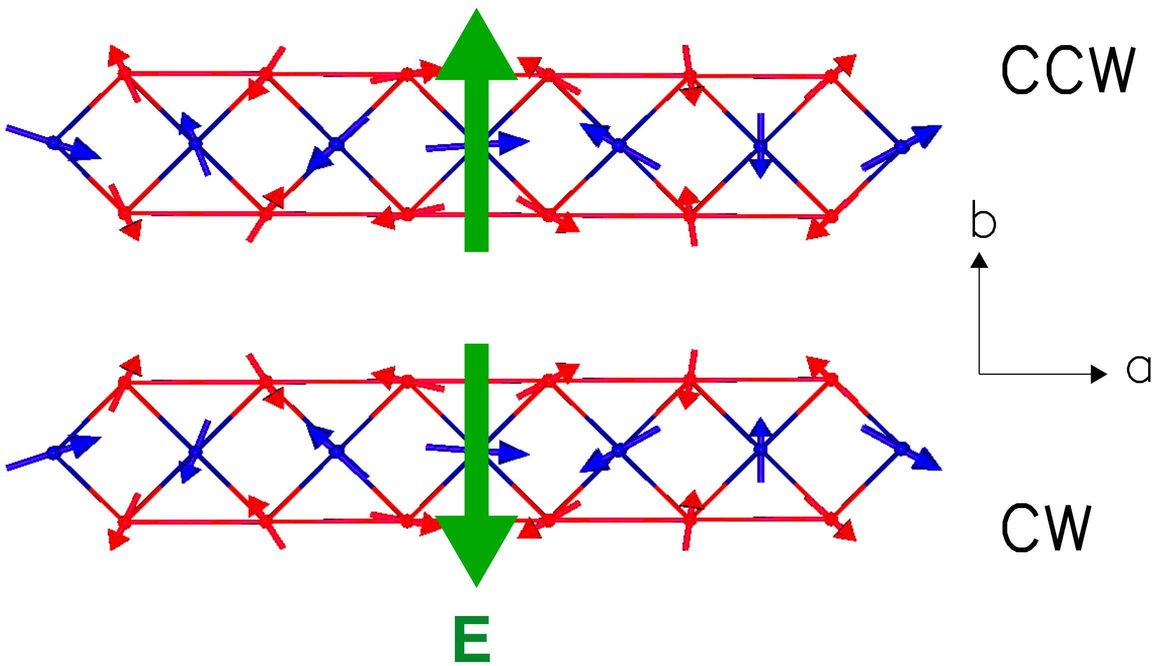
$$\left(\frac{d\sigma}{d\Omega}\right)^{\mathbf{P}_i||\mathbf{Q}}_{+-} \propto |M_{\perp}^{\perp\mathbf{P}_i}|^2 - PM_{chi}$$

Not visible in unpolarized!

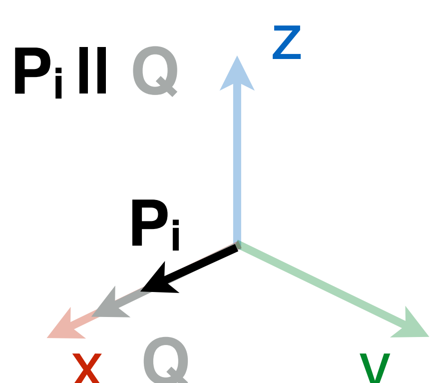
If we can set **x** || **Q**, and if we use two flippers, it is still possible (in most cases) to separate all of the components (see Blume, Ressouche for the maths).

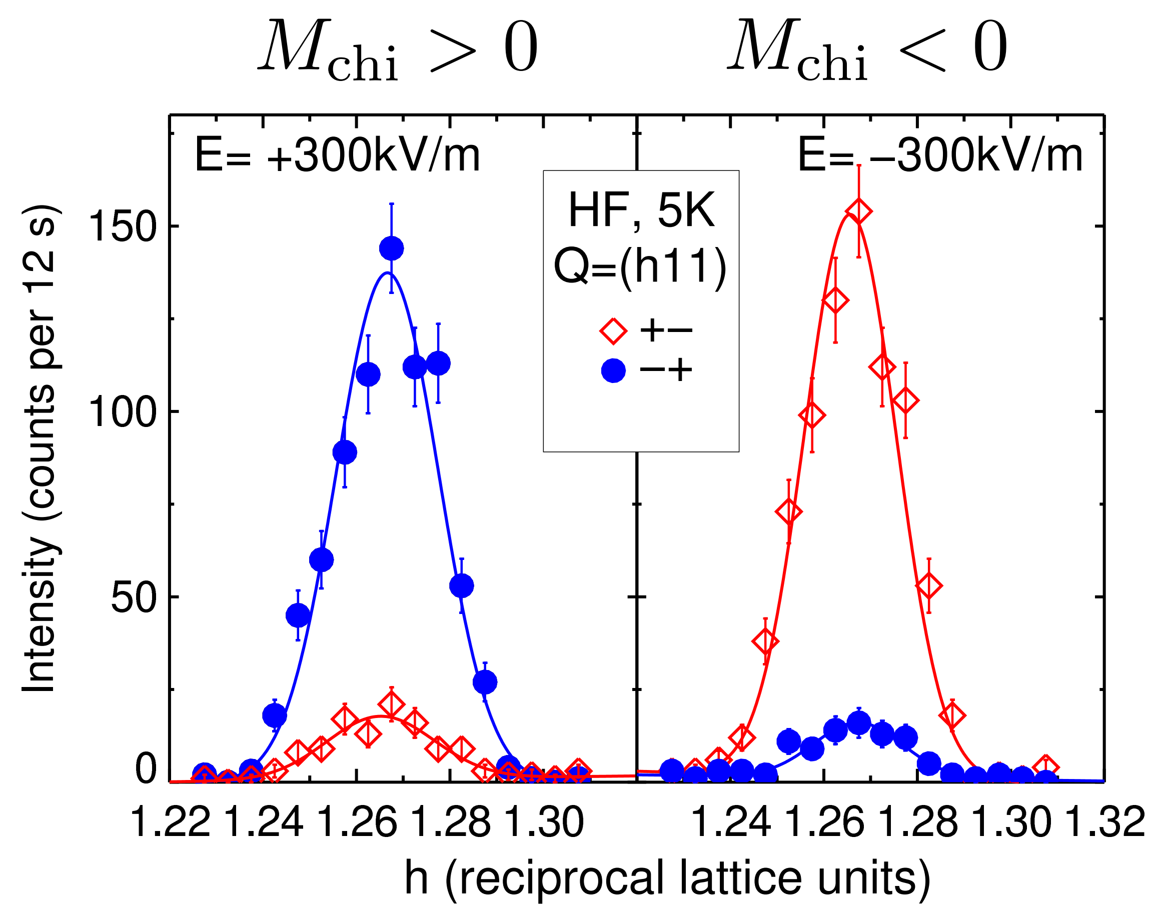
Example: non-collinear structure

In the multiferroic $Ni_3V_2O_8$, we can select handedness by applying an electric field:



$$\left(\frac{d\sigma}{d\Omega}\right)_{+-}^{\mathbf{P}_i||\mathbf{Q}} \propto |M_\perp^{\perp \mathbf{P}_i}|^2 - PM_{chi}$$





Cabrera et. al.

Spherical polarimetry

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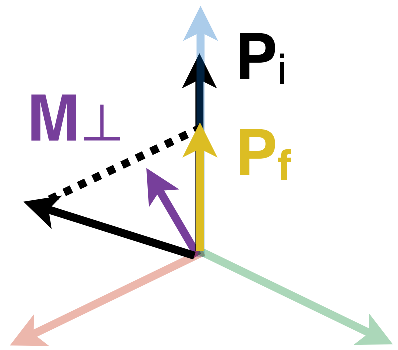
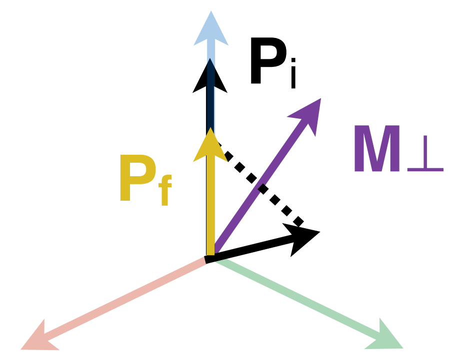
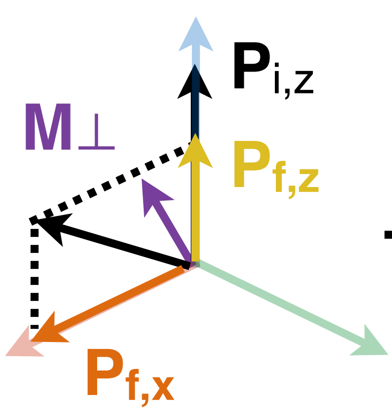
In some cases, crystal symmetry means that different magnetic structures look identical in LPA. This is a result of the projection onto the **P**$_i$ (field) direction:





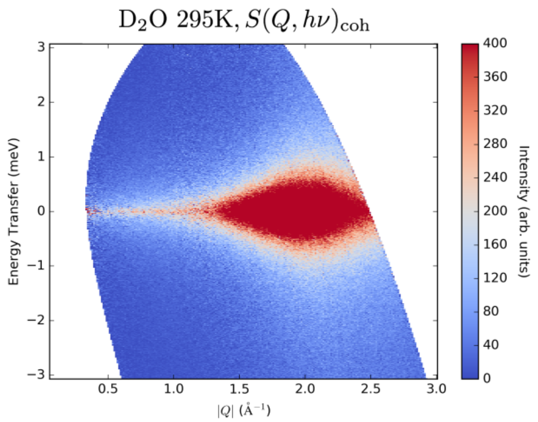
In this case, LPA is insufficient, and we need to measure all components of the scattered polarization. This is achieved by performing **spherical polarimetry**



$$\longrightarrow \begin{pmatrix} P_{f,x} \\ P_{f,y} \\ P_{f,z} \end{pmatrix} = \begin{pmatrix} P_{xx} & P_{xy} & \boxed{P_{xz}} \\ P_{yx} & P_{yy} & P_{yz} \\ P_{zx} & P_{zy} & \boxed{P_{zz}} \end{pmatrix} \begin{pmatrix} P_{i,x} \\ P_{i,y} \\ P_{i,z} \end{pmatrix}$$

In spherical polarimetry, projection avoided by placing sample in zero field, and carefully controlling **P**$_i$ and **P**$_f$ with fields and flippers (see Brown, Forsyth, Tasset).

Advanced polarized neutron scattering: Beyond (magnetic) diffraction



PA beyond magnetic diffraction

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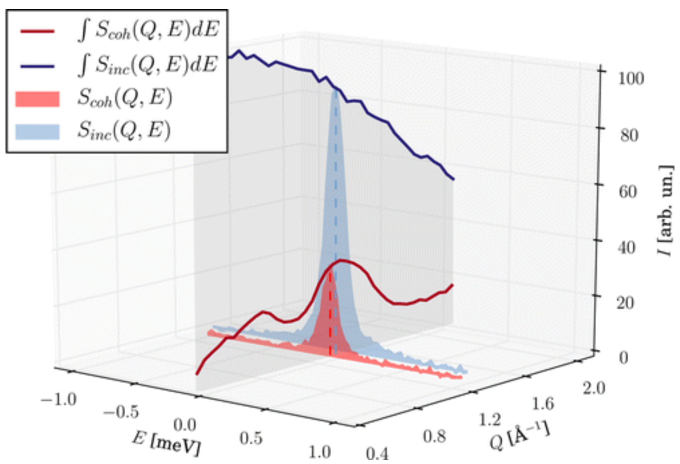
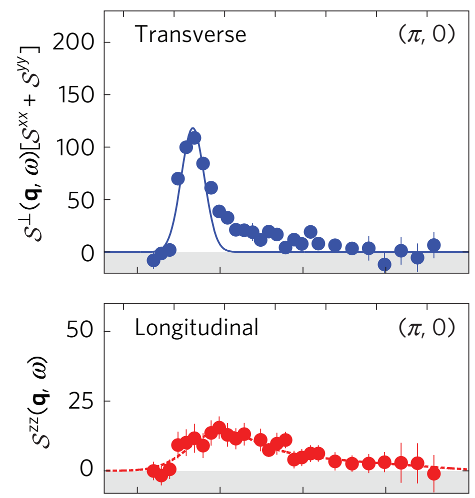
Inelastic scattering

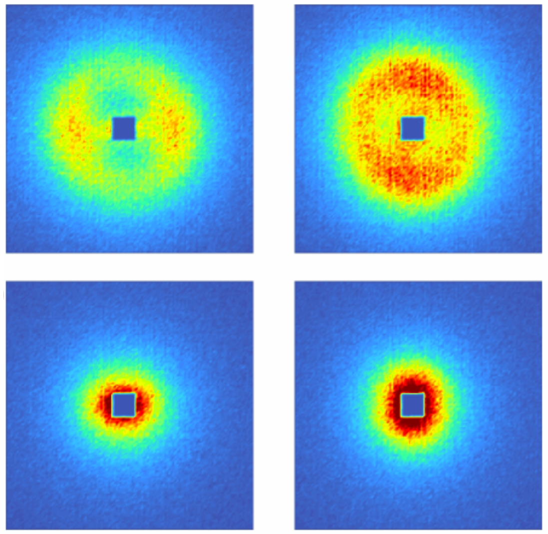


Burankova *J. Phys Chem. B*



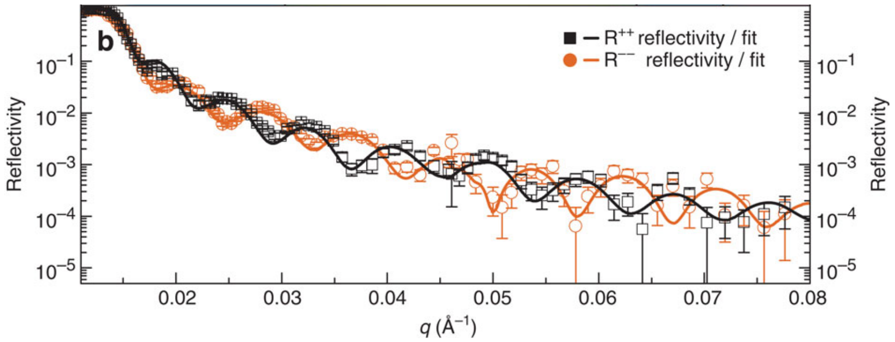
Dalla Piazza *Nat. Phys.*

Polarized SANS

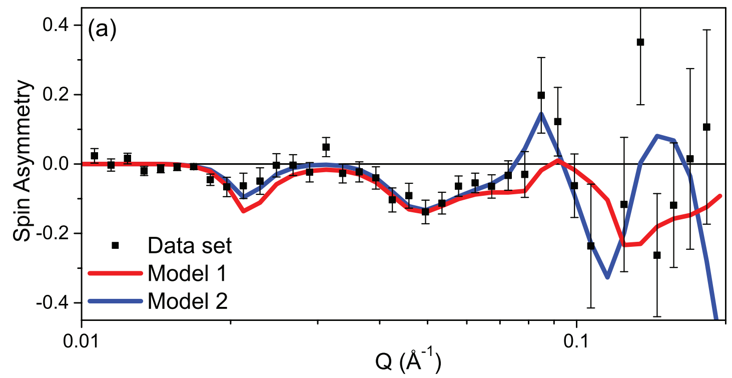


Klepp, *Materials*

Polarized Reflectometry (S. Langridge)

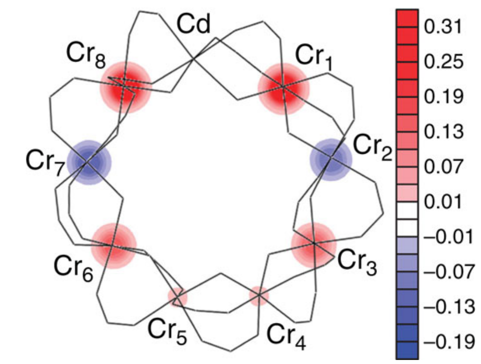


Spurgeon *Nature Comms.*



Satchell *J. Phys.: Condens. Matter*

Spin Density

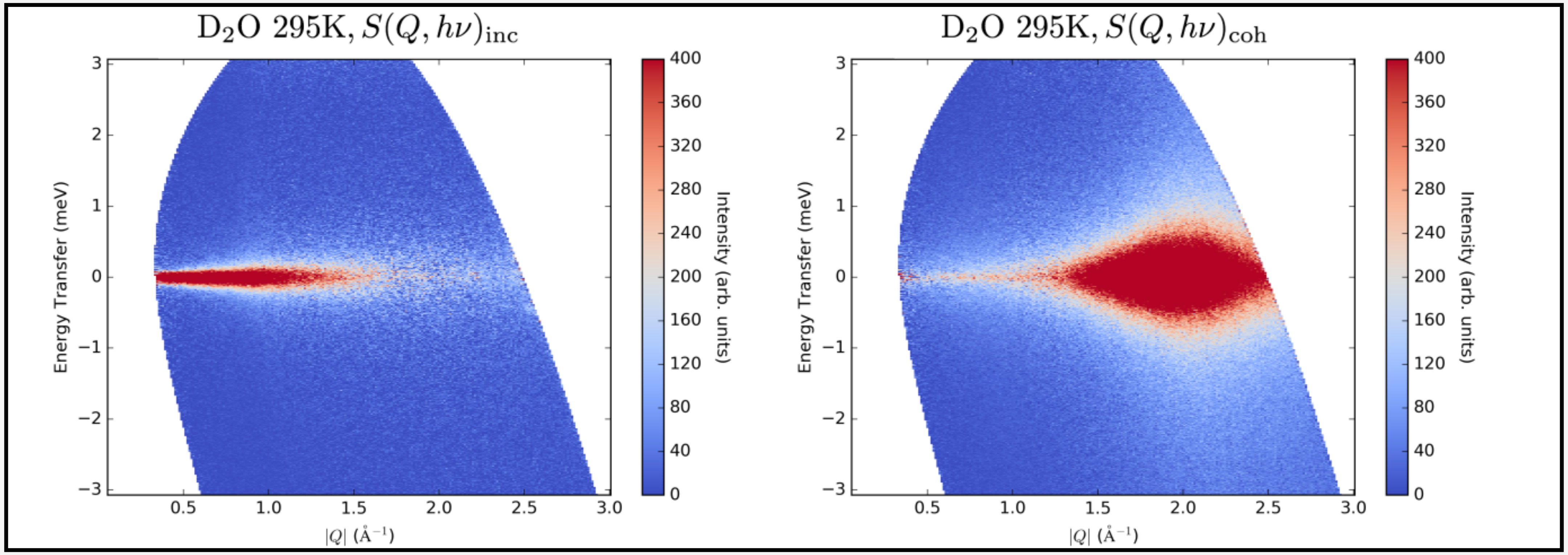


Guidi *Nature Comms.*

Example: Inelastic scattering

One of the most promising future applications is inelastic polarised neutron scattering on wide-angle inelastic spectrometers.

D_2O (LET, ISIS)



Polarized neutrons allow for the separation of collective $\boldsymbol{S_{coh}(Q,E)}$ and single-particle dynamics $\boldsymbol{S_{inc}(Q,E)}$. This has resulted in a revision of the model for the dynamics in water.

Arbe et. al.

Conclusion

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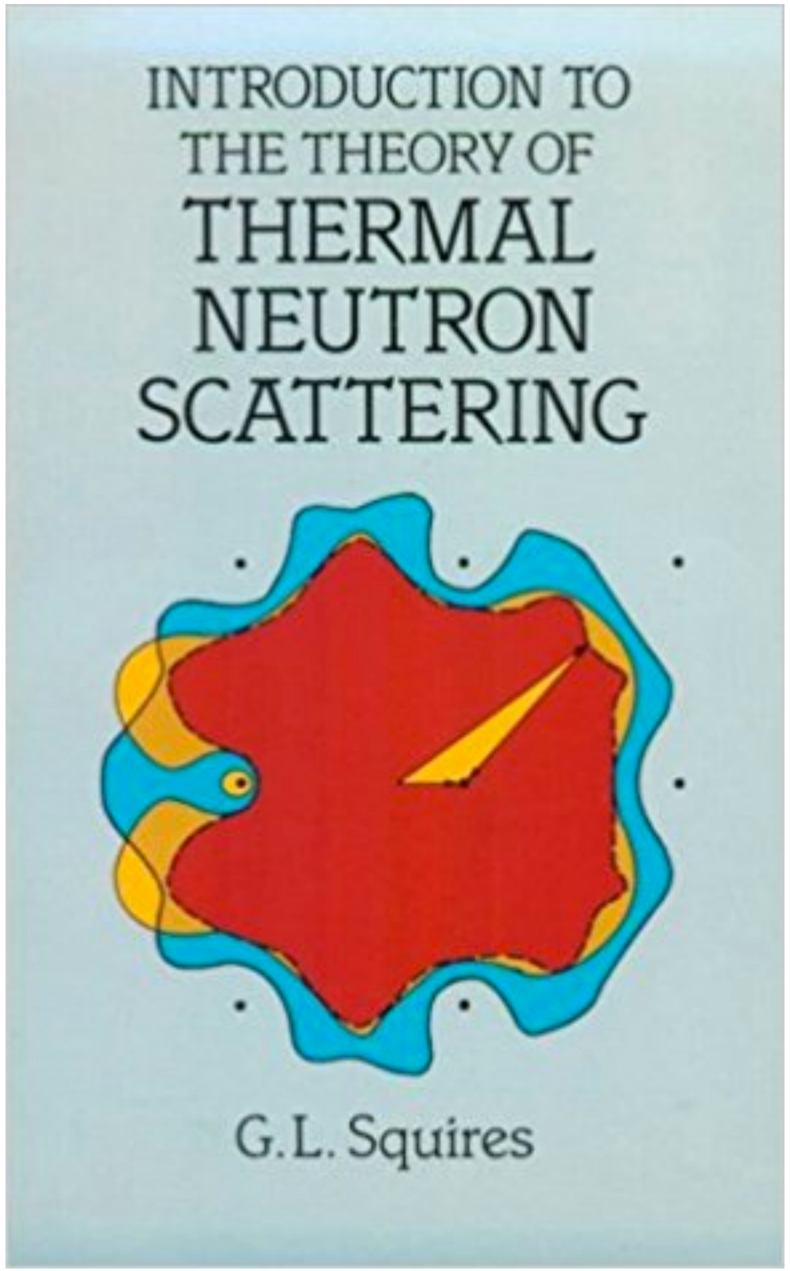
- Polarized neutron beams interact with magnetic moments (both nuclear and electronic) in samples. **The scattered polarization and cross section depends on the type of scattering process** (nuclear coherent, spin incoherent, or magnetic).

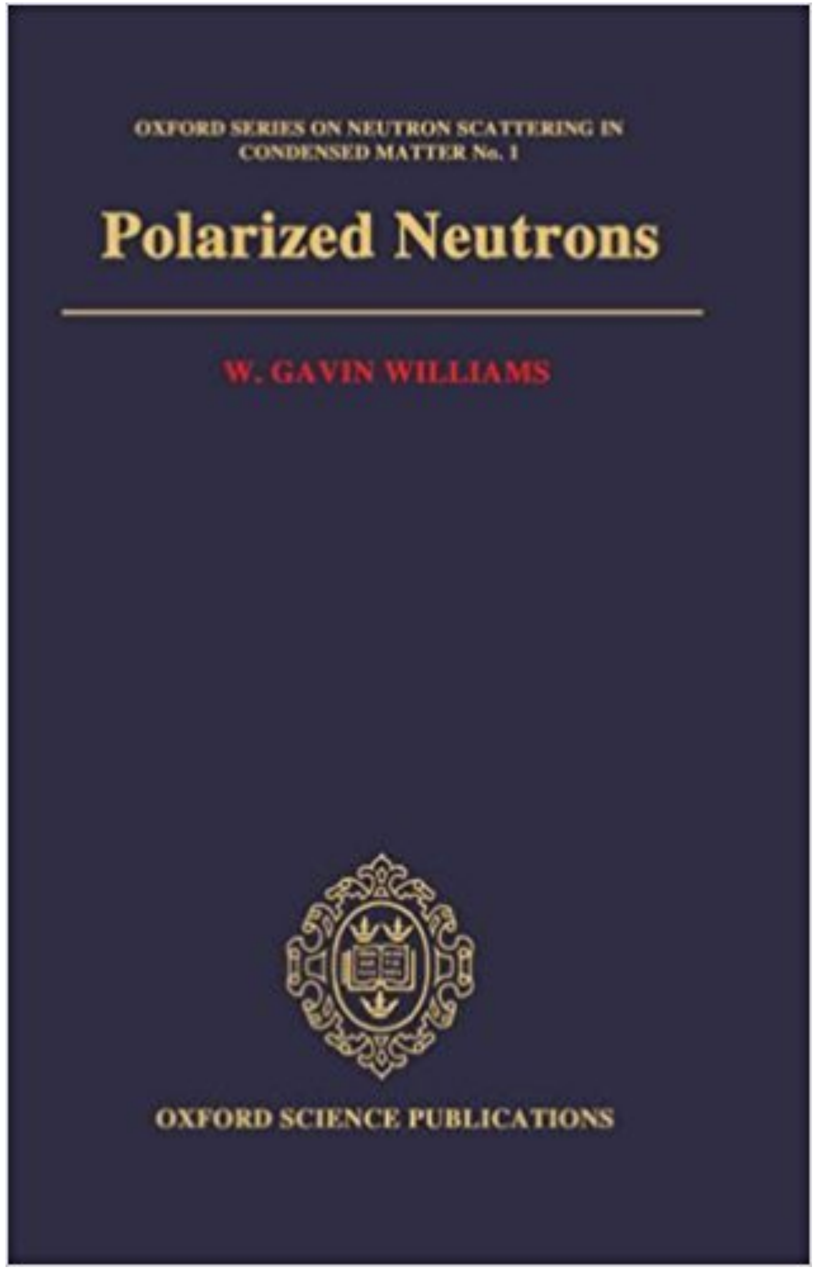
- Polarized neutron beams can therefore be used to:
    - **Separate cross section components**
    - **Determine magnetic moment orientations**
    - **Access parts of the cross section inaccessible to unpolarised neutrons**

- Polarized neutron beams can also be used to improve the resolution of neutron scattering (A. Faraone)

Books



Basic theory



Devices

Further reading

Theory

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

LPA: Blume, Phys. Rev. **130** (1963) 1670

Polarimetry: Brown, Forsyth, Tasset, Proc. Roy. Soc **442** (1969) 147

2D XYZ: Schärpf and Capellmann, phys. stat. sol. a **135** (1993) 359

LPA+Polarimetry: Ressouche Collection SFN **13** (2014) 02002

Examples

Multiferroic $Ni_3V_2O_8$: Cabrera et. al. Phys. Rev. Lett. **103** (2009) 087201

Ionic liquids: Burankova J. Phys. Chem. B **118** (2014) 14452

Frustrated magnet $Lu_2Mo_2O_5N_2$: Clark et. al. Phys. Rev. Lett. **113** (2014) 117201

Magnetic semiconductor Mn:ZnO: Lancon et. al. Appl. Phys. Lett. **109** (2016) 252405

Instrumentation

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

D7 and 2D XYZ: Stewart et. al. J. Appl. Cryst. **42** (2009) 69

Polarimetry: Tasset, Physica B **267** (1999) 69

Take-home message

Polarized neutrons can be used to enhance (nearly) any neutron scattering experiment, either by providing additional information (this lecture), or improving the resolution or range using Larmor precession (A. Faraone)