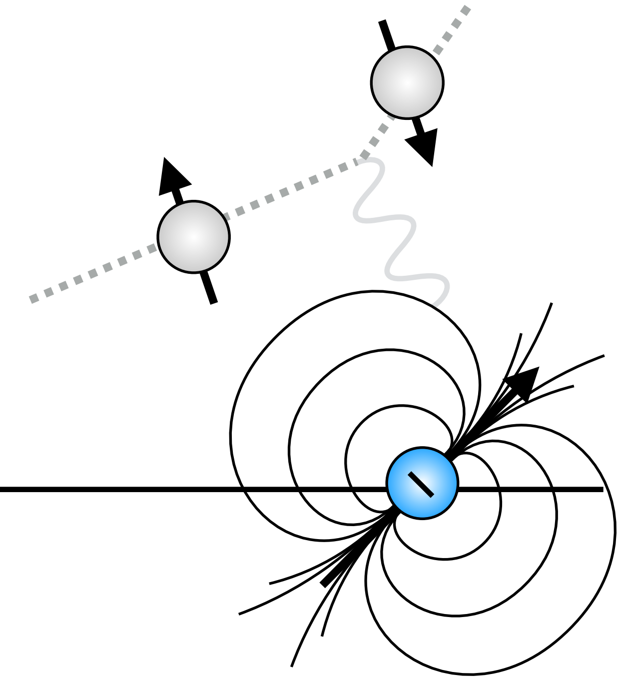
Polarized neutron scattering



Gøran Nilsen

https://creazilla.com/nodes/1597-oxford-skyline-silhouette

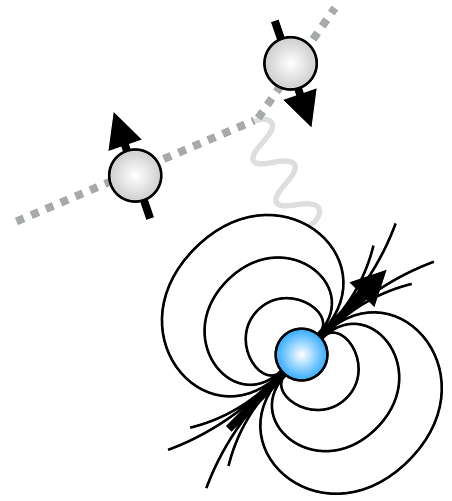
Take-home message

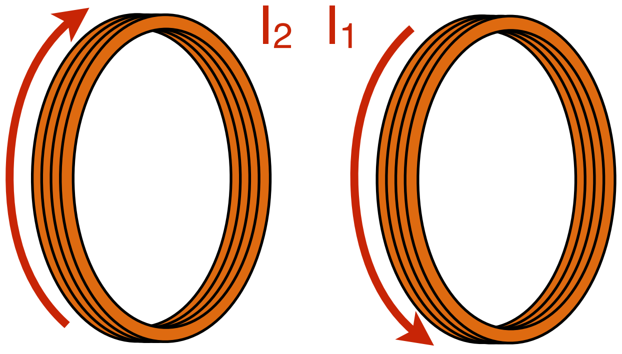
Polarized neutrons can be used to enhance (nearly) any neutron scattering experiment, either by:

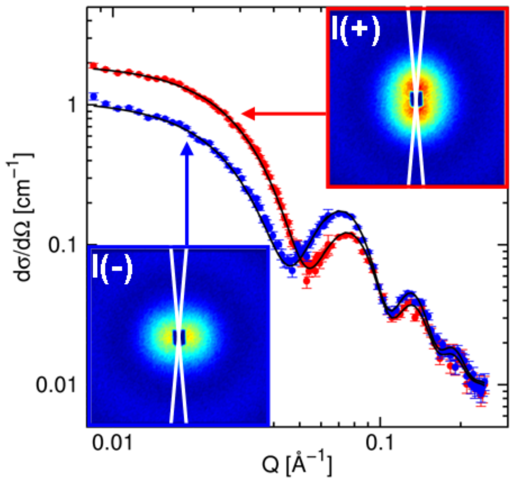
1. **Providing additional information on the scattering components (coherent, incoherent, magnetic)**
2. Improving the resolution or range

Overview

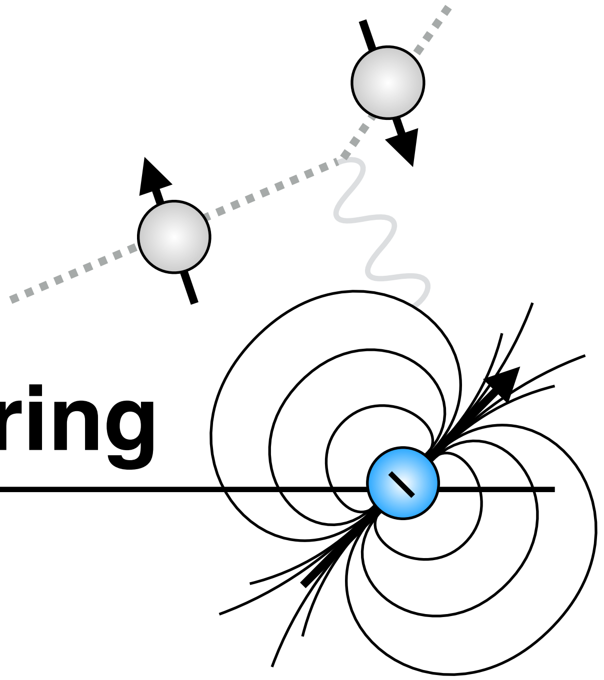
- **Principles of polarized neutron scattering**
  - What is a polarized neutron beam?
  - How do polarized neutrons interact with matter?
  - What extra information can be gained by using polarized neutrons?
- **Practical polarized neutron scattering**
  - Devices: polarizers/analyzers, flippers, and guide field
- **Techniques and applications of polarized neutrons**
  - Half-polarized
  - Longitudinal polarization analysis
  - Spherical polarimetry
- **Spin echo**





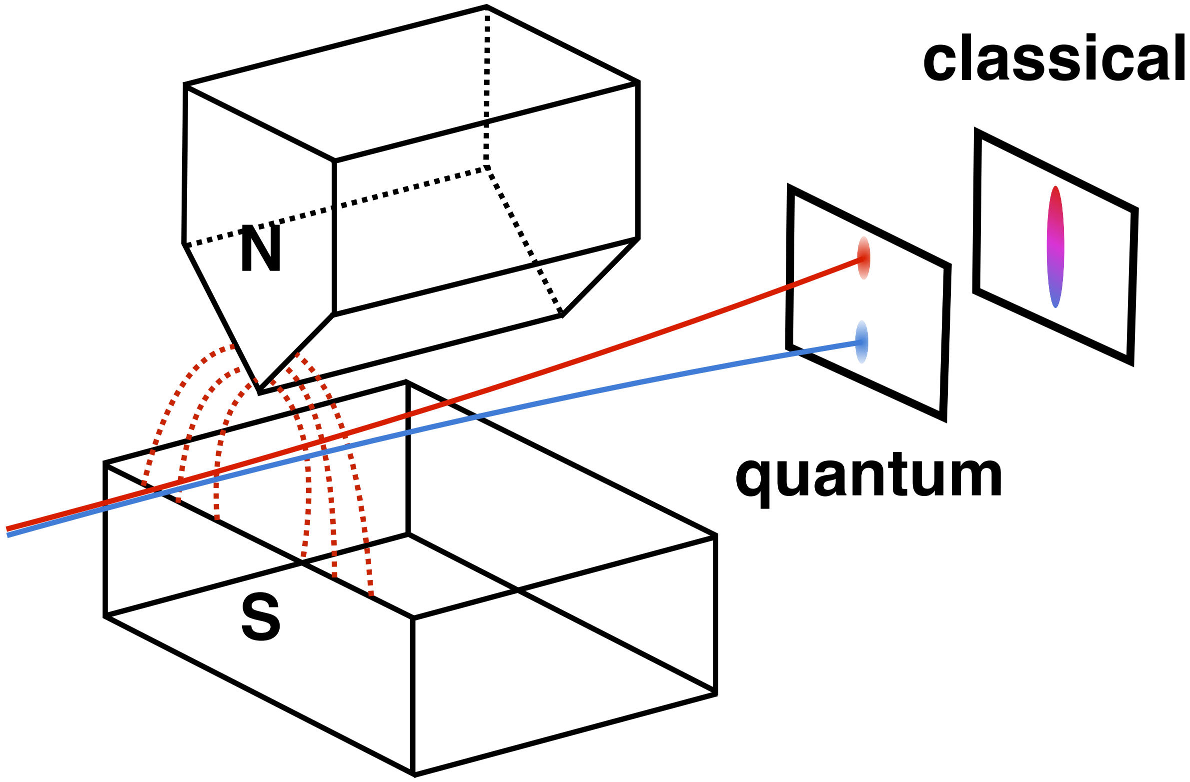


Principles of polarised neutron scattering

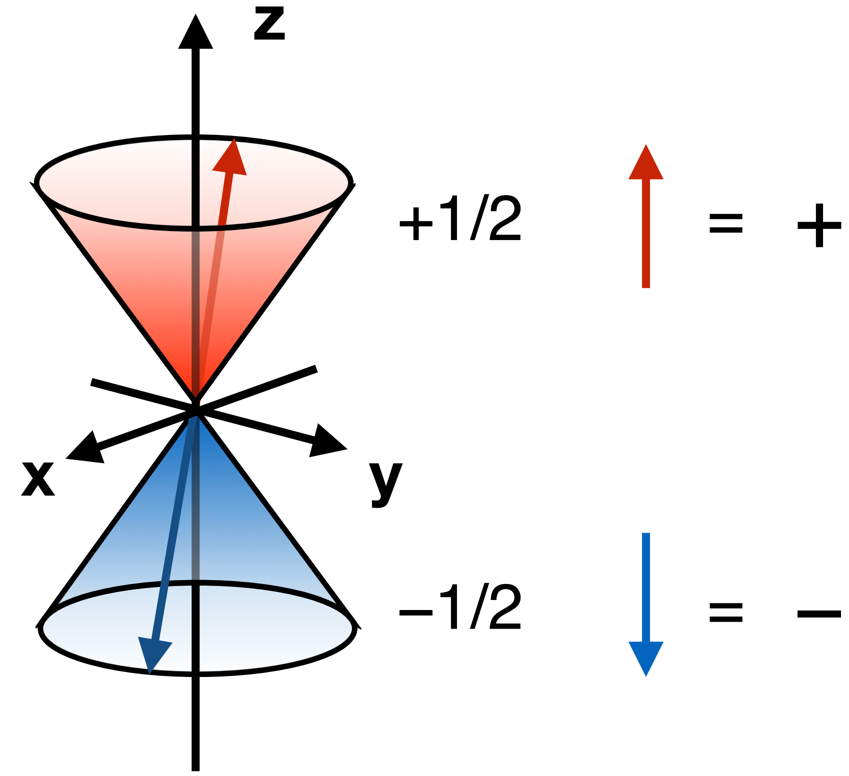


Spin angular momentum

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



Stern, Gerlach (1922)



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

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**:

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

or

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

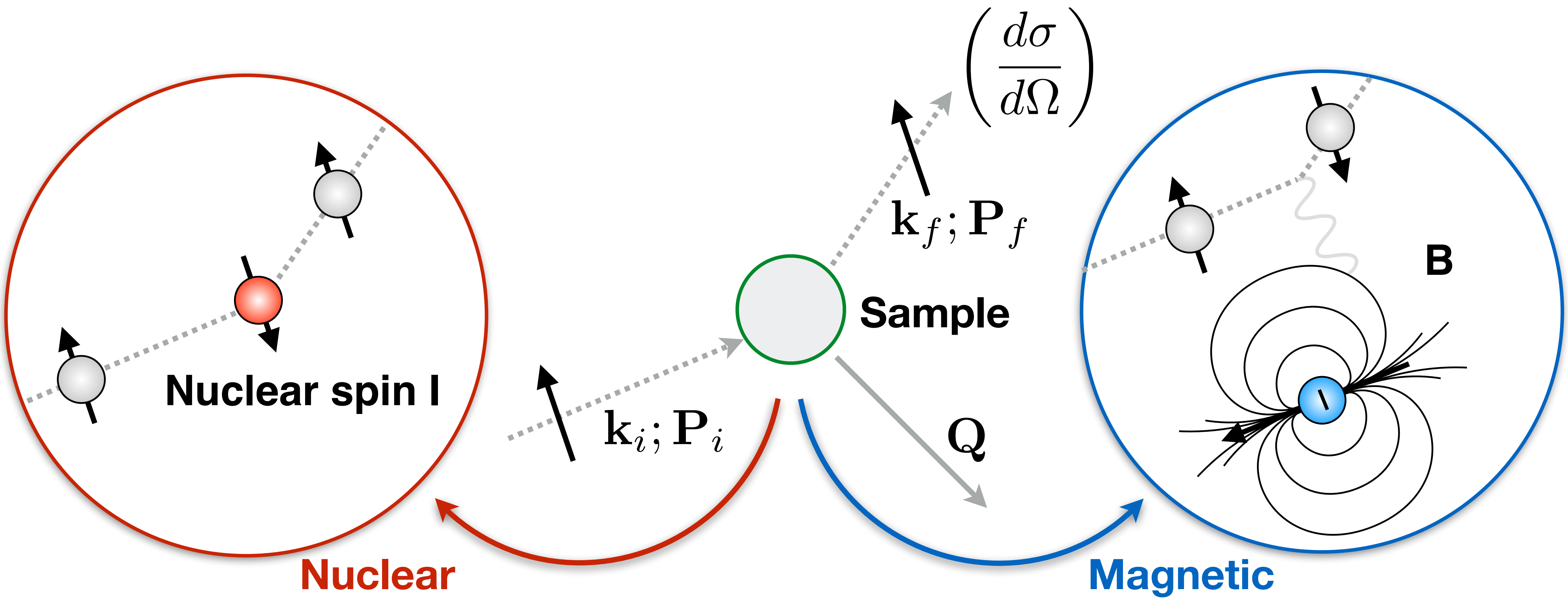
Where F is the **flipping ratio**, a frequently measured experimental quantity.

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 elastic scattering

In most experiments, 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)

$\uparrow \rightarrow \downarrow$ $\downarrow \rightarrow \uparrow$

“+−” **“−+”**

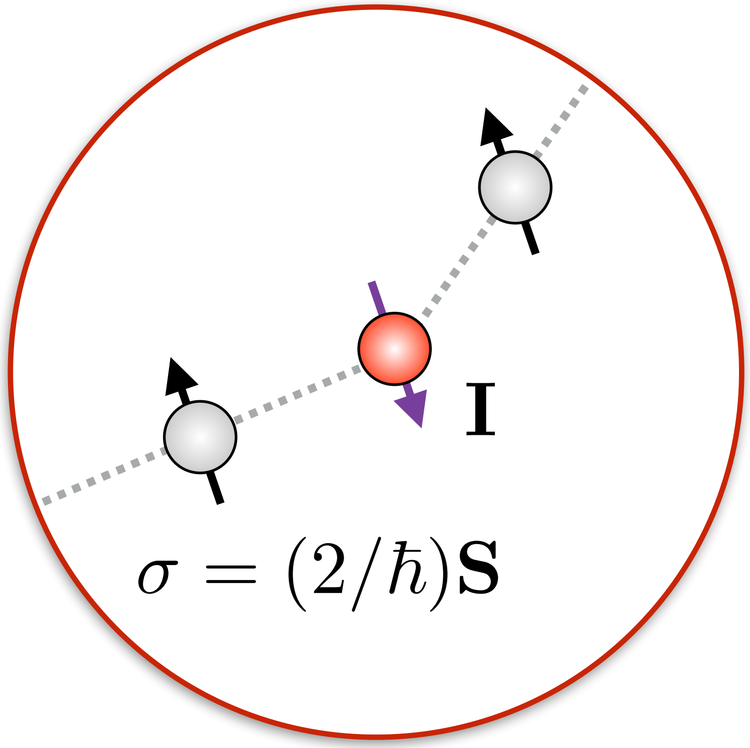
Cross sections

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

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 Boothroyd Ch. 4):



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

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

Nuclear coh.
Isotope inc.

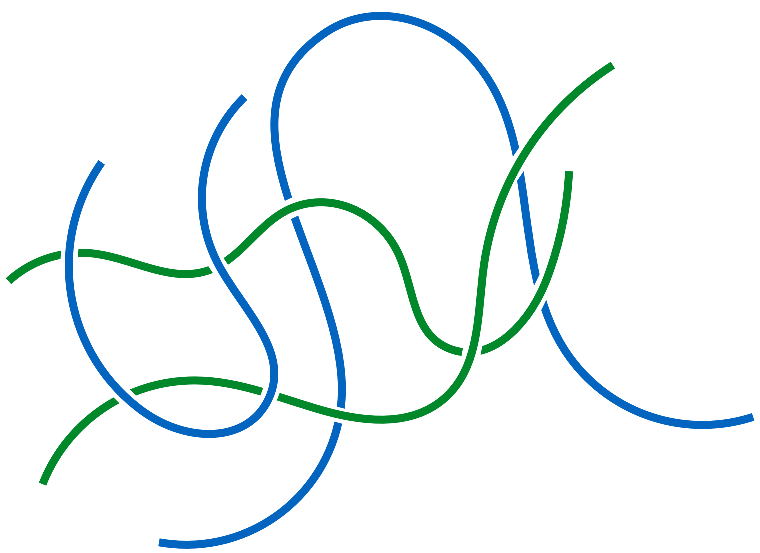
Spin inc.
$\sqrt{B^2 I(I+1)}$

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

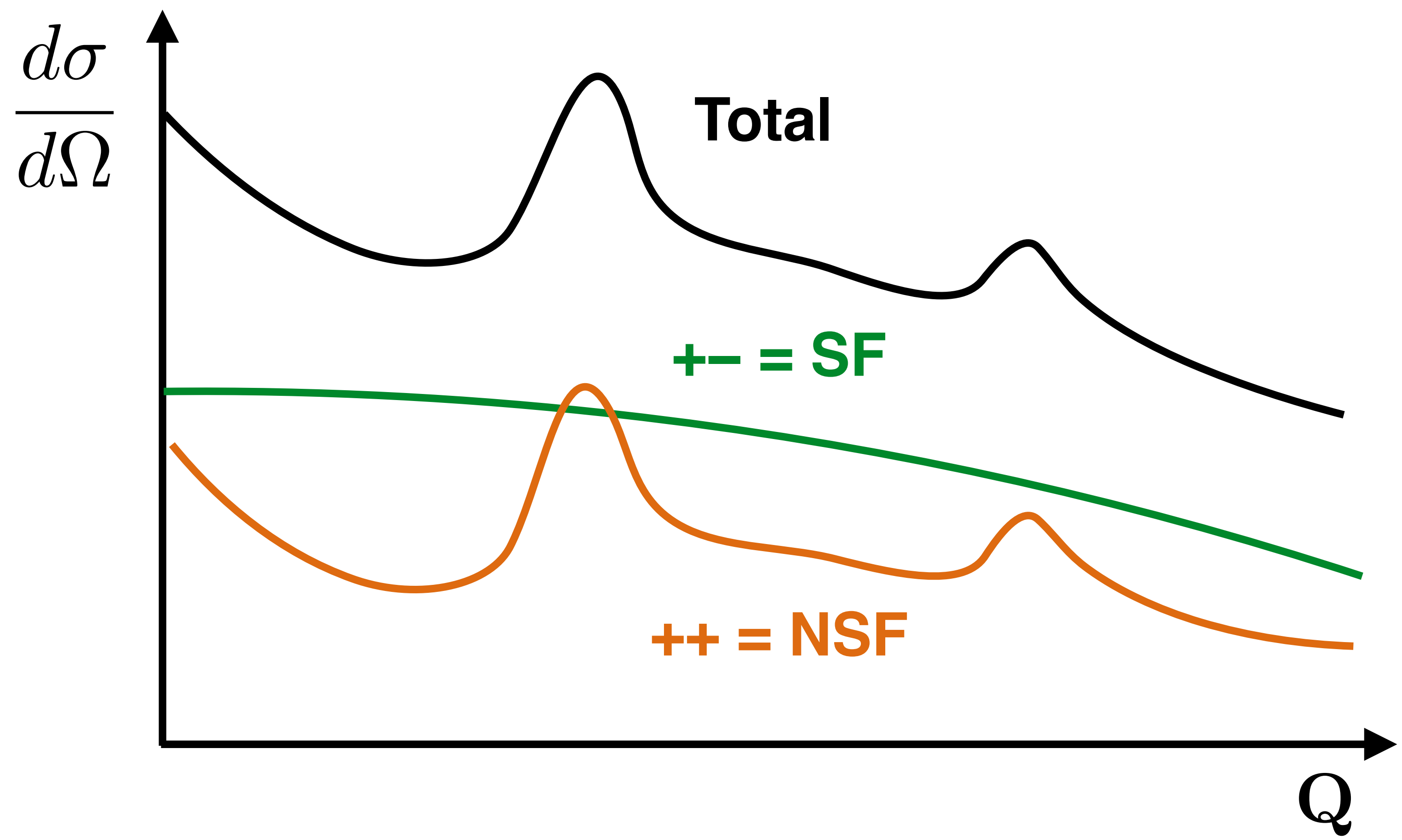
SF: $\boxed{\left(\frac{d\sigma}{d\Omega}\right)_{+-} = \left(\frac{d\sigma}{d\Omega}\right)_{-+}} = \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{inc}}$

Example 1: Polymer

Consider a hydrocarbon polymer:



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

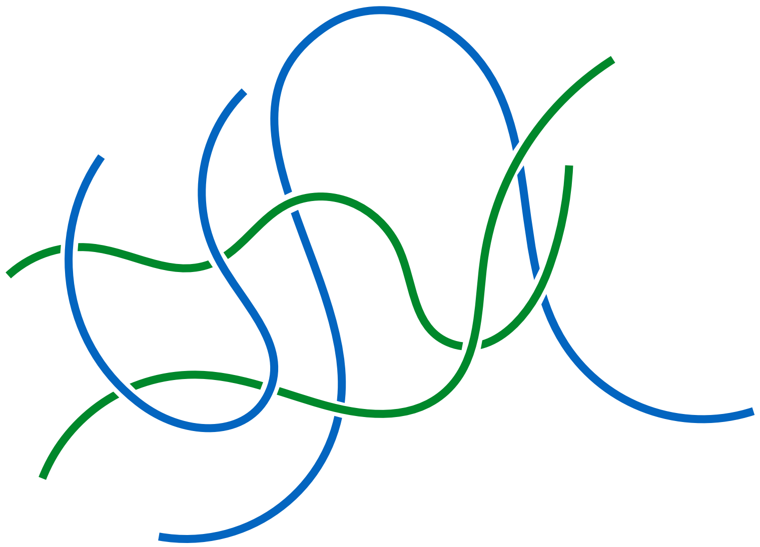


If we perform longitudinal polarization analysis, we can separate the contributions:

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

Example 1: Polymer

Consider a hydrocarbon polymer:



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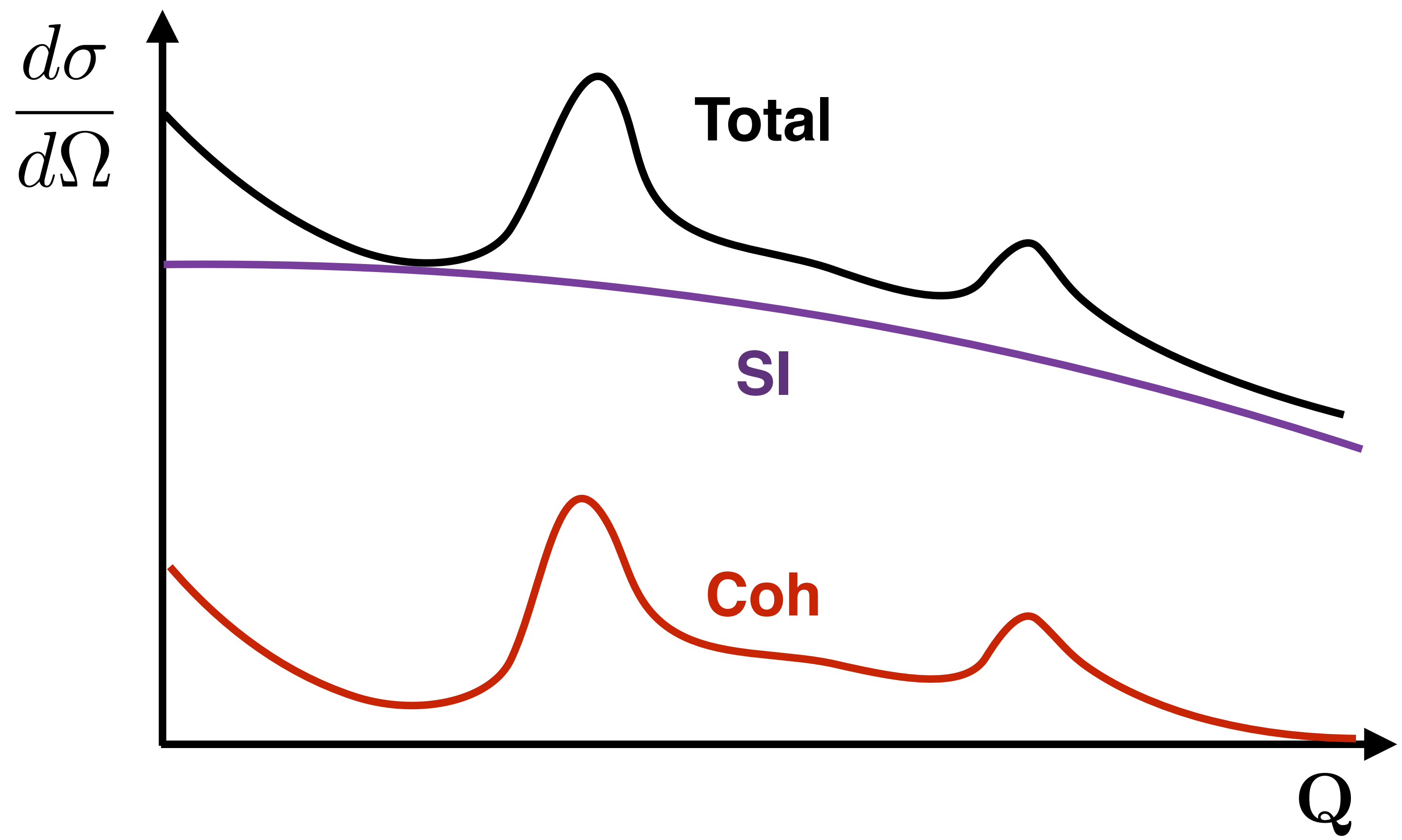
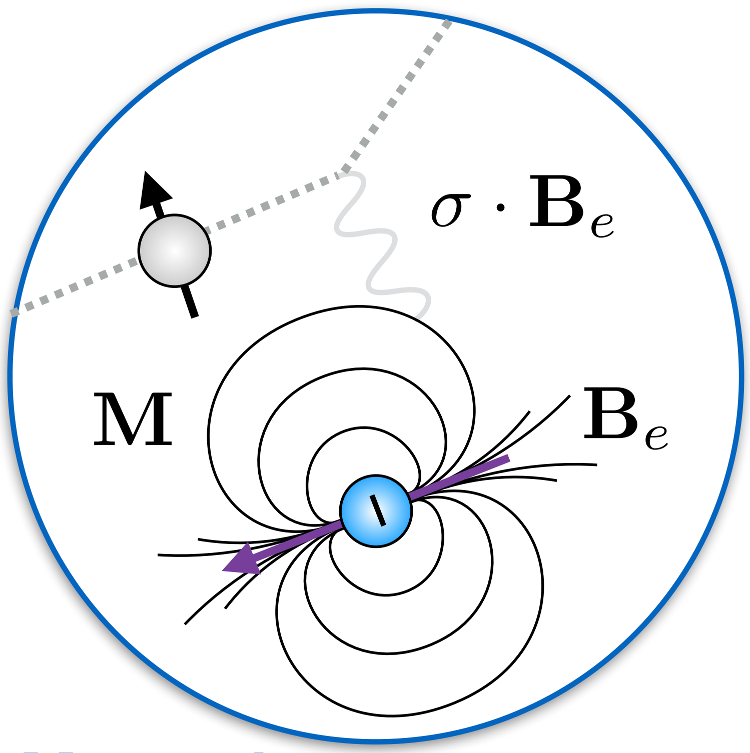


If we perform longitudinal polarization analysis, we can separate the contributions:

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

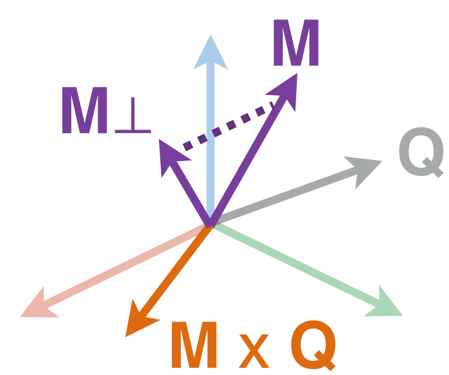
Magnetic scattering

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



Magnetic

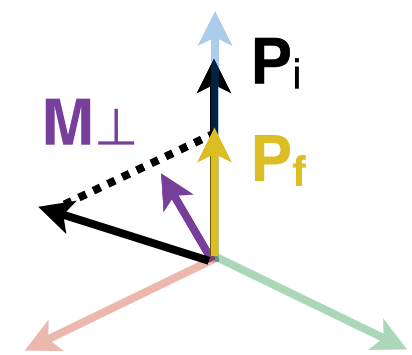
A Only measure components $\mathbf{M}\perp\mathbf{Q}$



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

Boothroyd Ch. 4
Squires Ch. 7

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



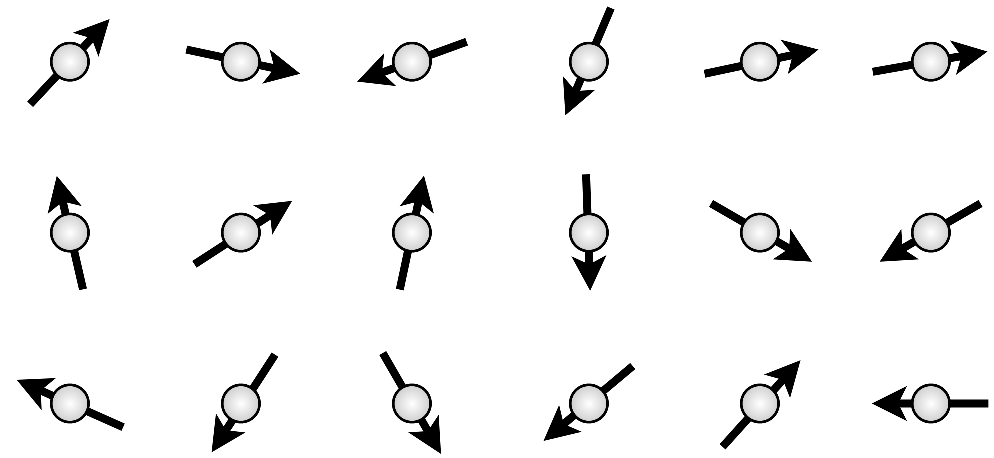
1. Rot. $\mathbf{P}_i$ 180° about **M**
2. Project onto $\mathbf{P}_i$

Boothroyd Ch. 4
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.

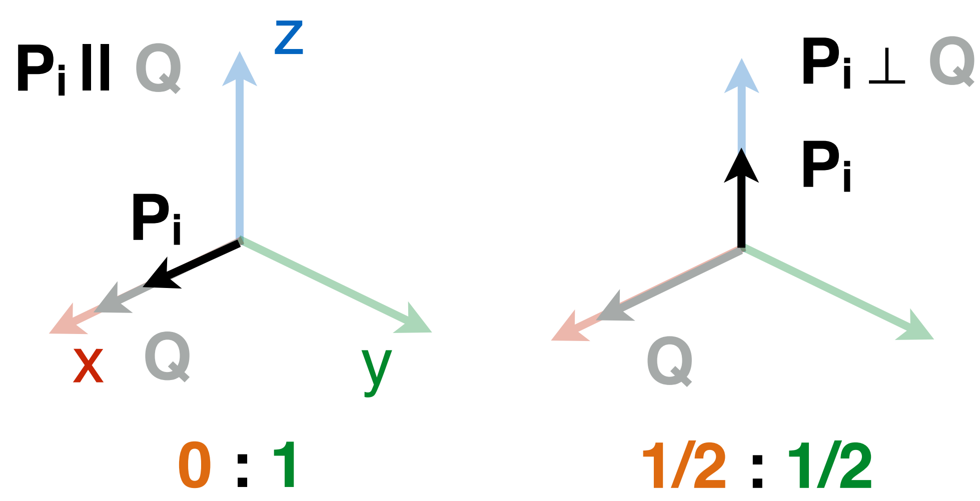


$\langle M \rangle = 0$

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:

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

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

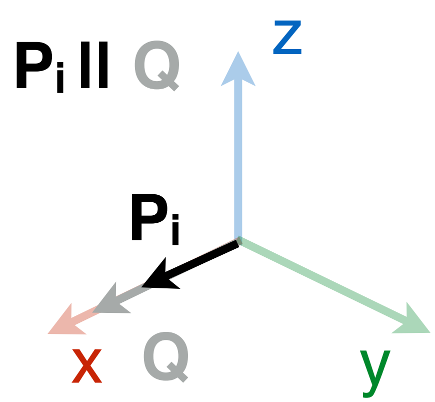


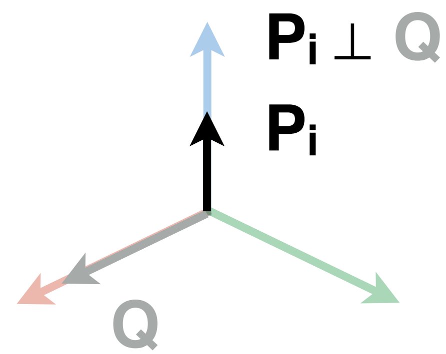
Squires Ch. 9, p. 179

Example 2: the || - ⊥ method

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

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





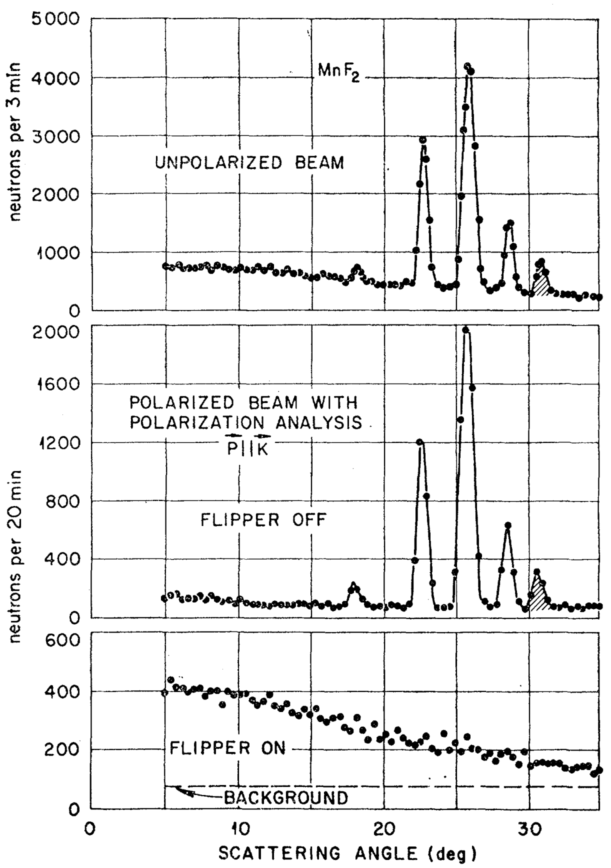
	$P_i \parallel Q$	$P_i \perp Q$
Magnetic	0 : 1	1/2 : 1/2
Spin incoherent	1/3 : 2/3	1/3 : 2/3
Nuclear	1 : 0	1 : 0

e.g.

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

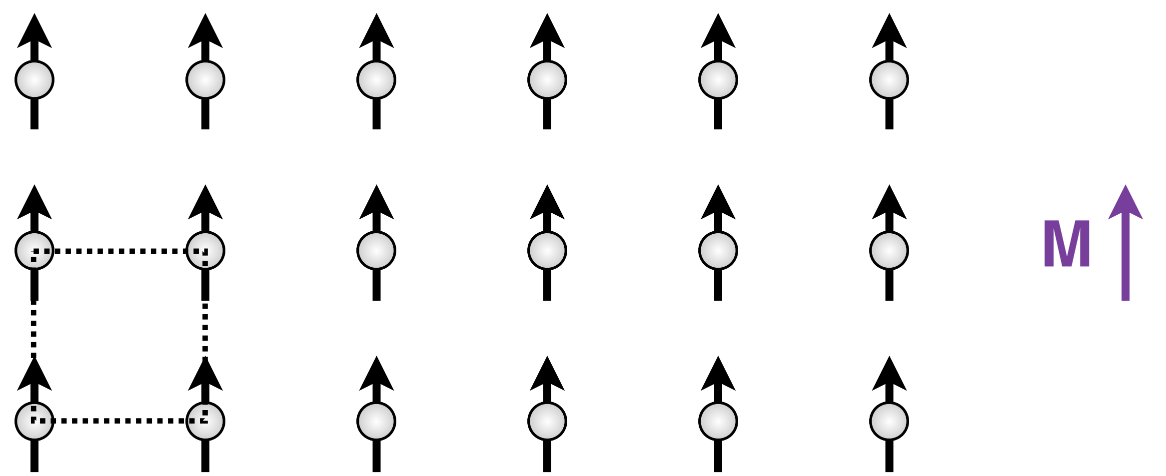
MnF_2

Moon, Riste, Koehler



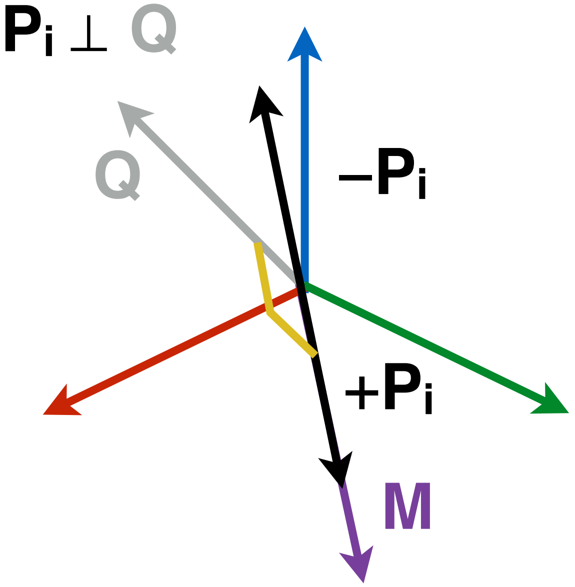
Example 3: collinear ferromagnet

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**, **P**$_\mathbf{i}$, and **Q**. It also includes **both** nuclear and magnetic contributions. For **M** || **P**$_\mathbf{i}$ ⊥ **Q**:

M || **P**$_\mathbf{i}$ ⊥ **Q**



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

$$\left.\begin{array}{ll} +\mathbf{P}_\mathbf{i} \parallel \mathbf{M} : \left(\dfrac{d\sigma}{d\Omega}\right)_+ & \propto |F_N + F_M|^2 \\ -\mathbf{P}_\mathbf{i} \parallel \mathbf{M} : \left(\dfrac{d\sigma}{d\Omega}\right)_- & \propto |F_N - F_M|^2 \end{array}\right\} \text{NM interference}$$

Example 3: magnetic crystal polarizer

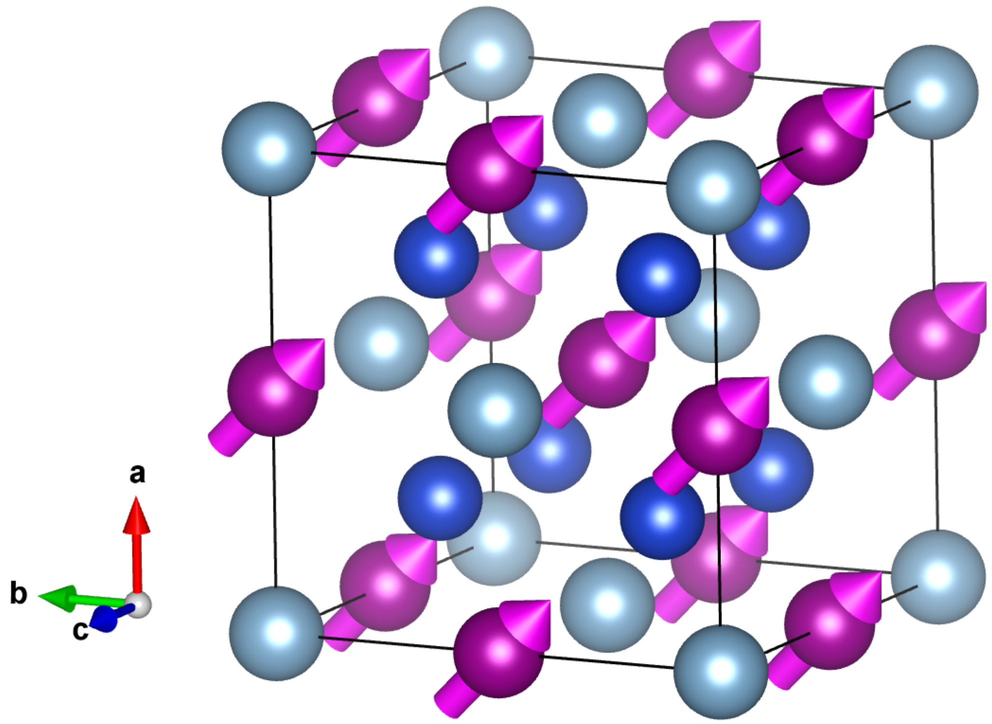
e.g. Cu_2MnAl

M || (110)

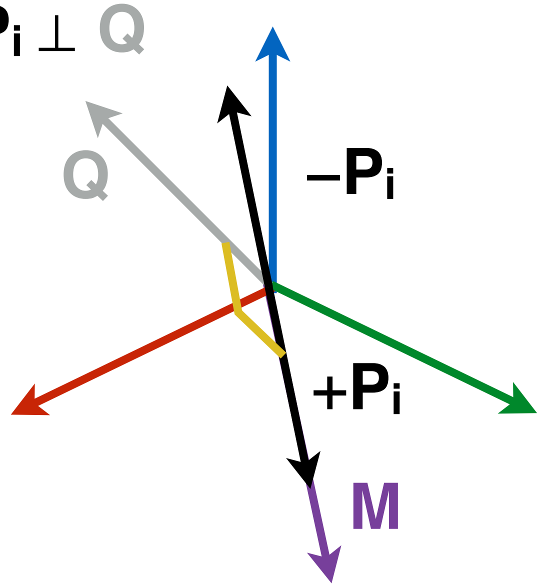
Q = (1-11)

$F_N = 7.2$ fm

$F_M = 6.8$ fm



M || $\mathbf{P_i} \perp$ **Q**



1. **M** $\perp$ **Q** : measure all of **M**
2. $\mathbf{P_i}$ || $\mathbf{M}_\perp$: all scattering **NSF**

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

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

Summary

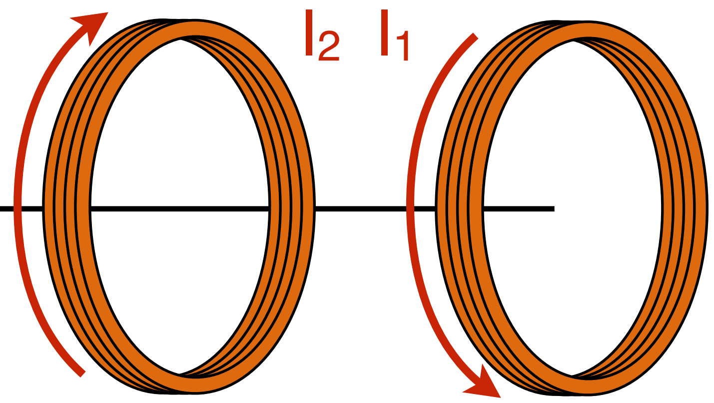
Rules

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

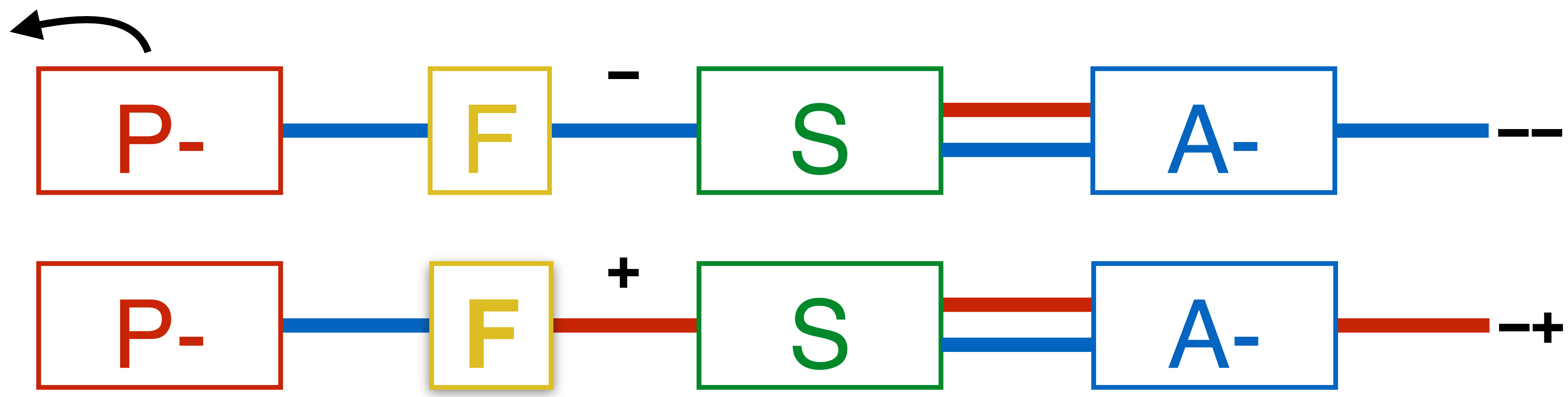
1. We can separate the components of the cross section (Examples 1,2)
2. We are also sensitive to the direction of magnetic moments through either the cross section (ferromagnets) or the polarization

Practical polarised neuton scattering



What do we need?

Returning to examples 1 and 2: how do we measure the SF and NSF scattering? We've seen that we can polarise and analyse a beam with crystals like Cu_2MnAl:

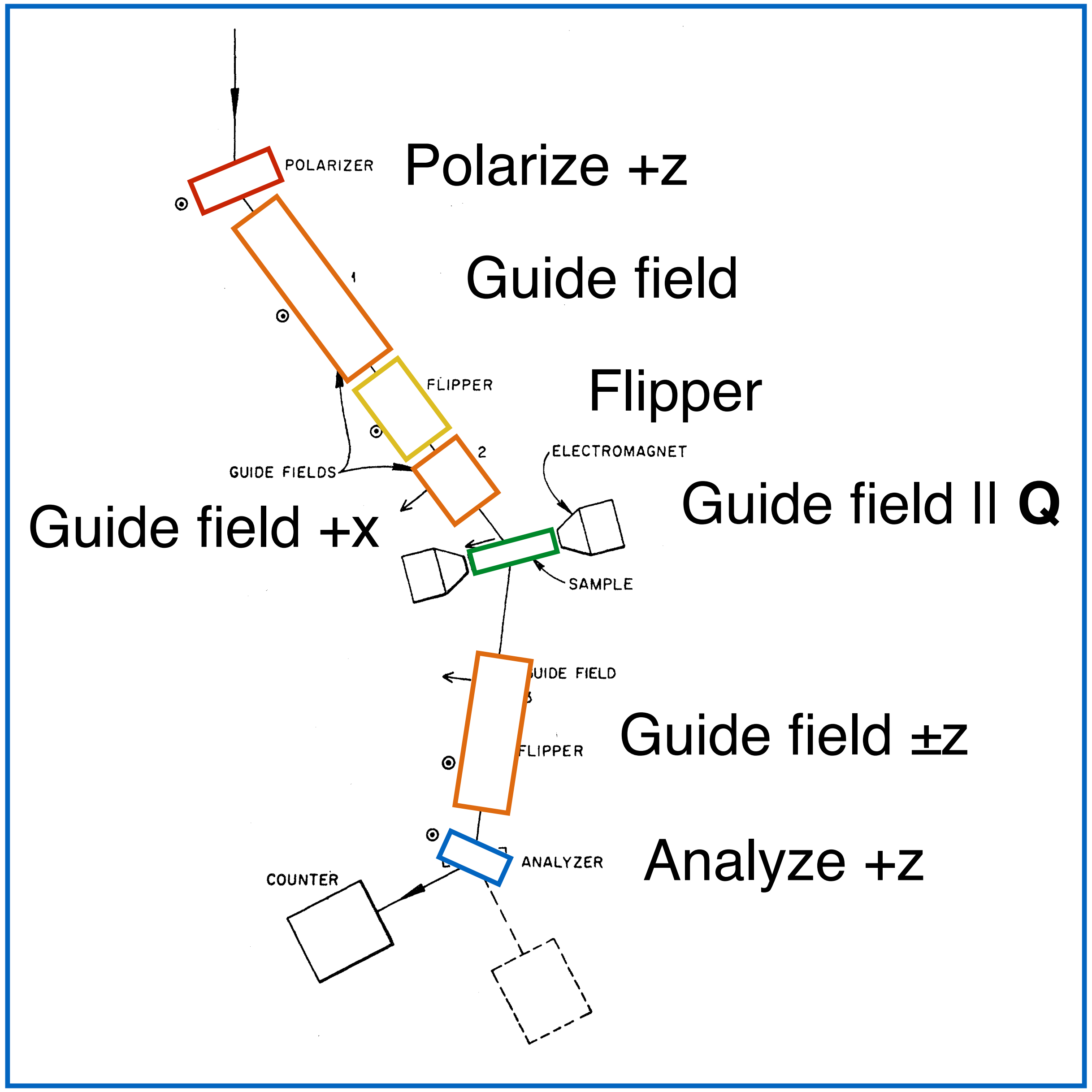


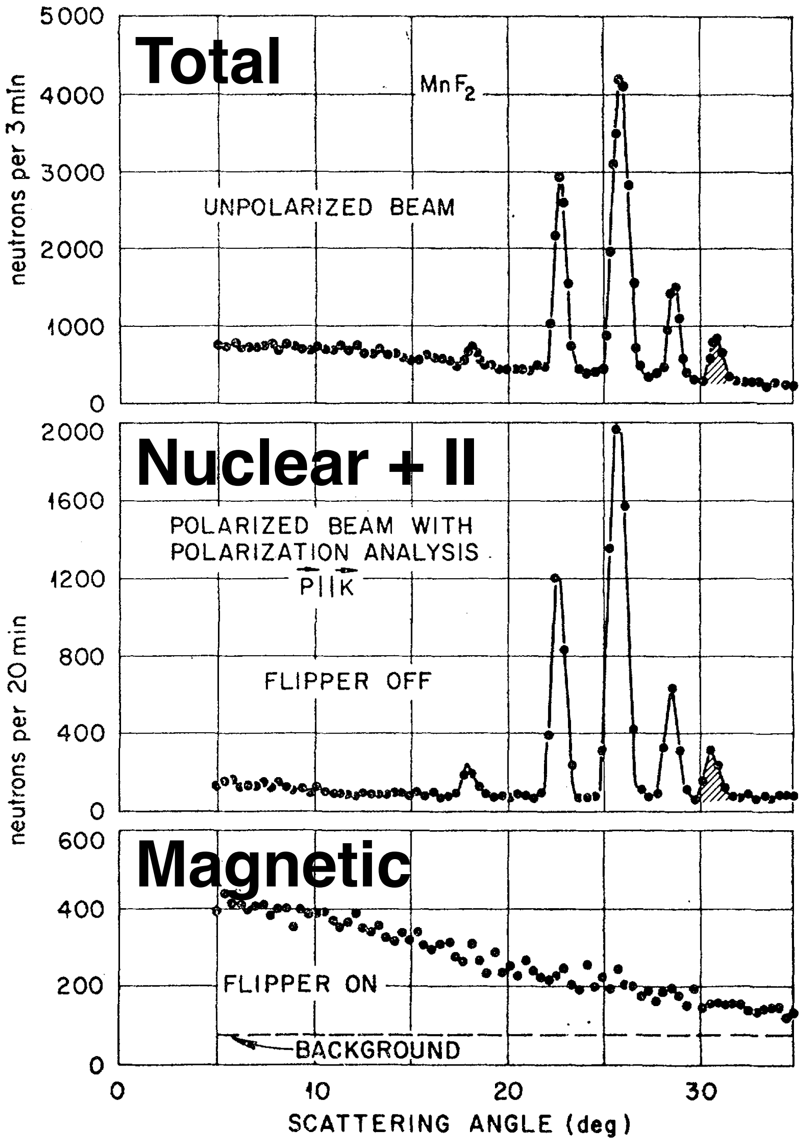
However, these are normally fixed to accept only one state — need **flippers**

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

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

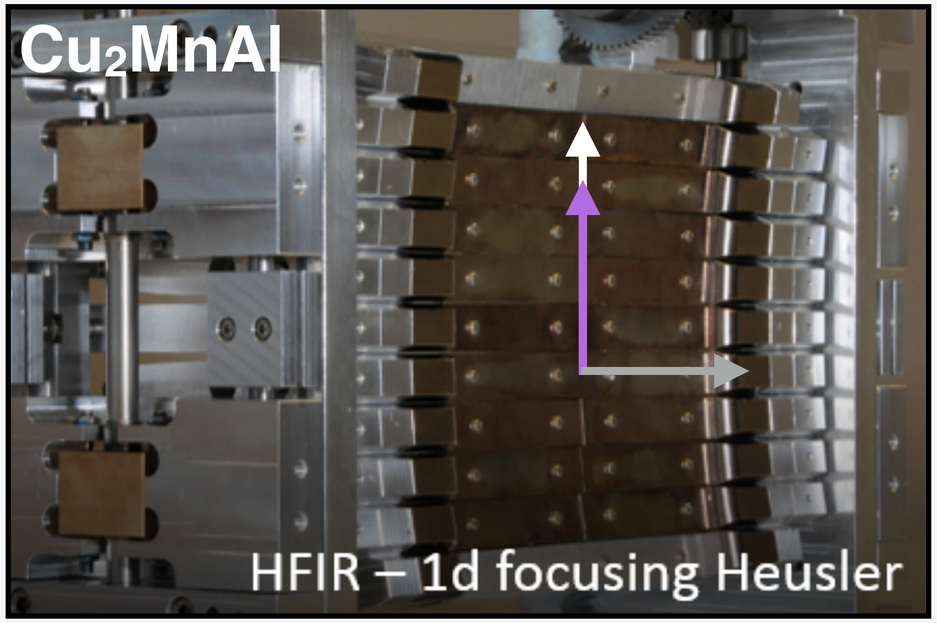




Moon, Riste, Koehler

Neutron polarizers and analyzers

1. Magnetic crystal



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

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

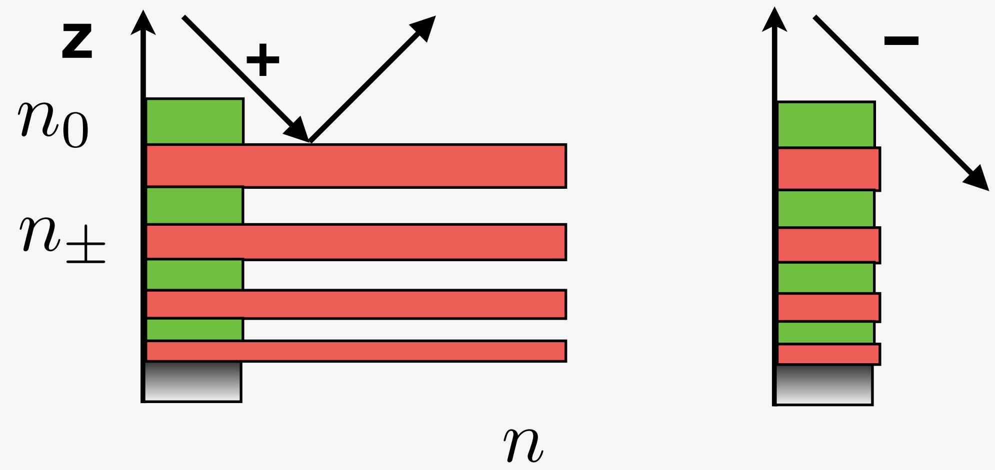
If $F_N = F_M$, polarized beam!

(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:

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

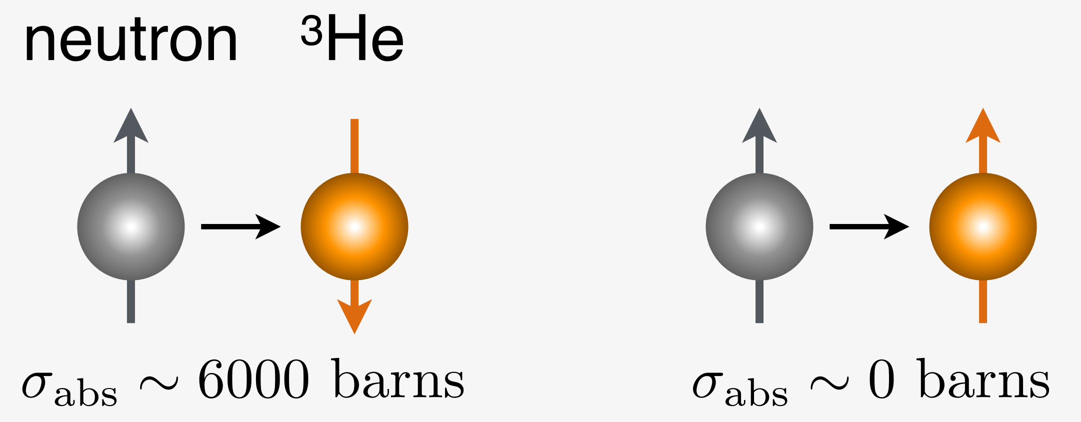
If $n_0 = |n_{\pm}|$, polarized beam!

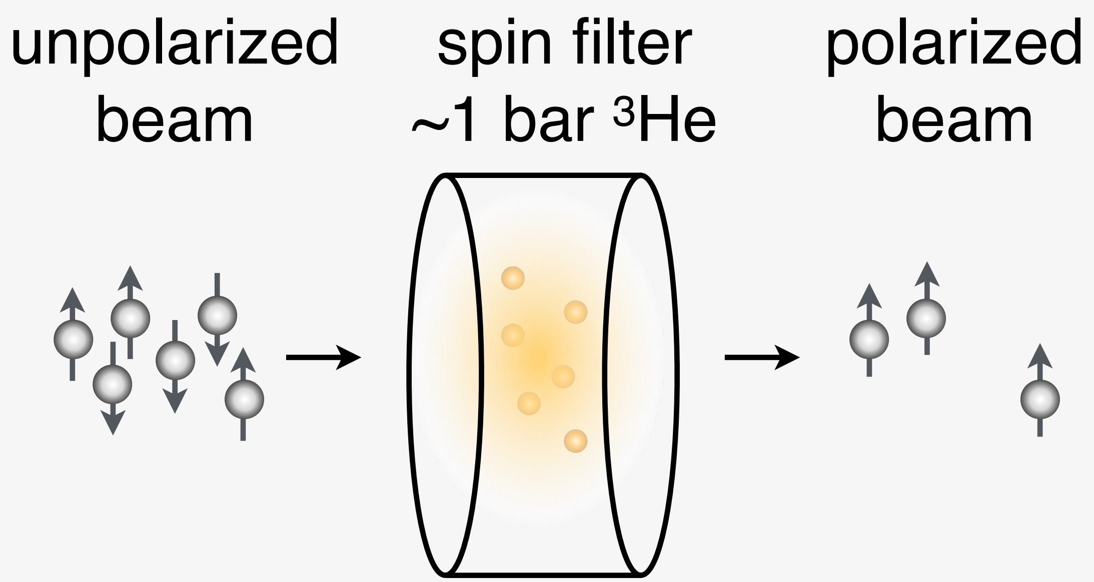
(see S. Langridge lecture)

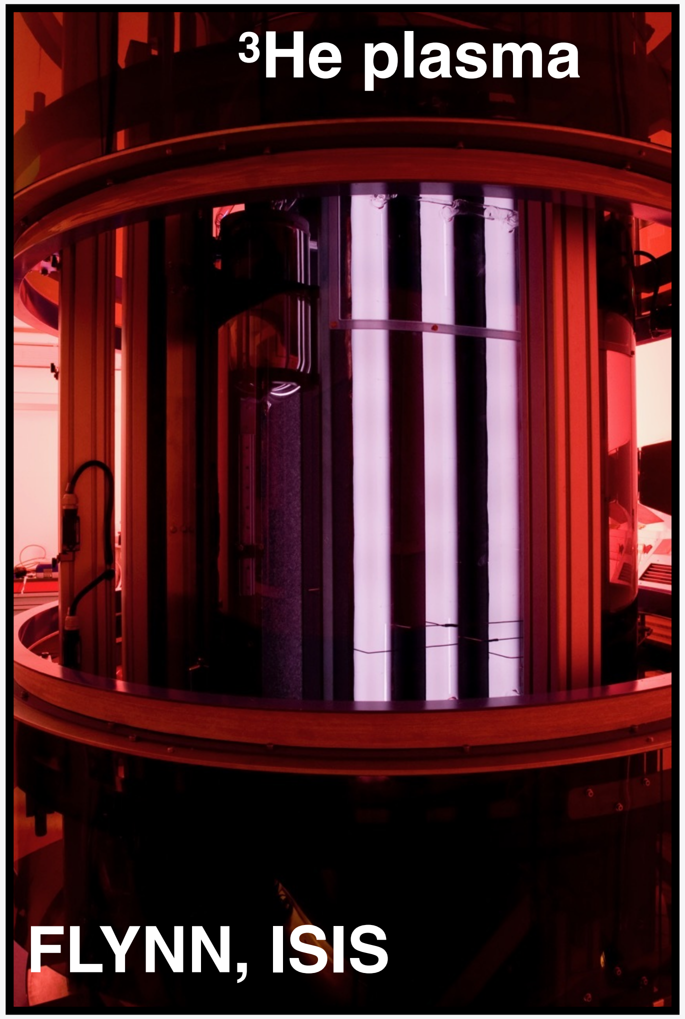
Neutron polarizers and analyzers

3. ^{3}He spin filter

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





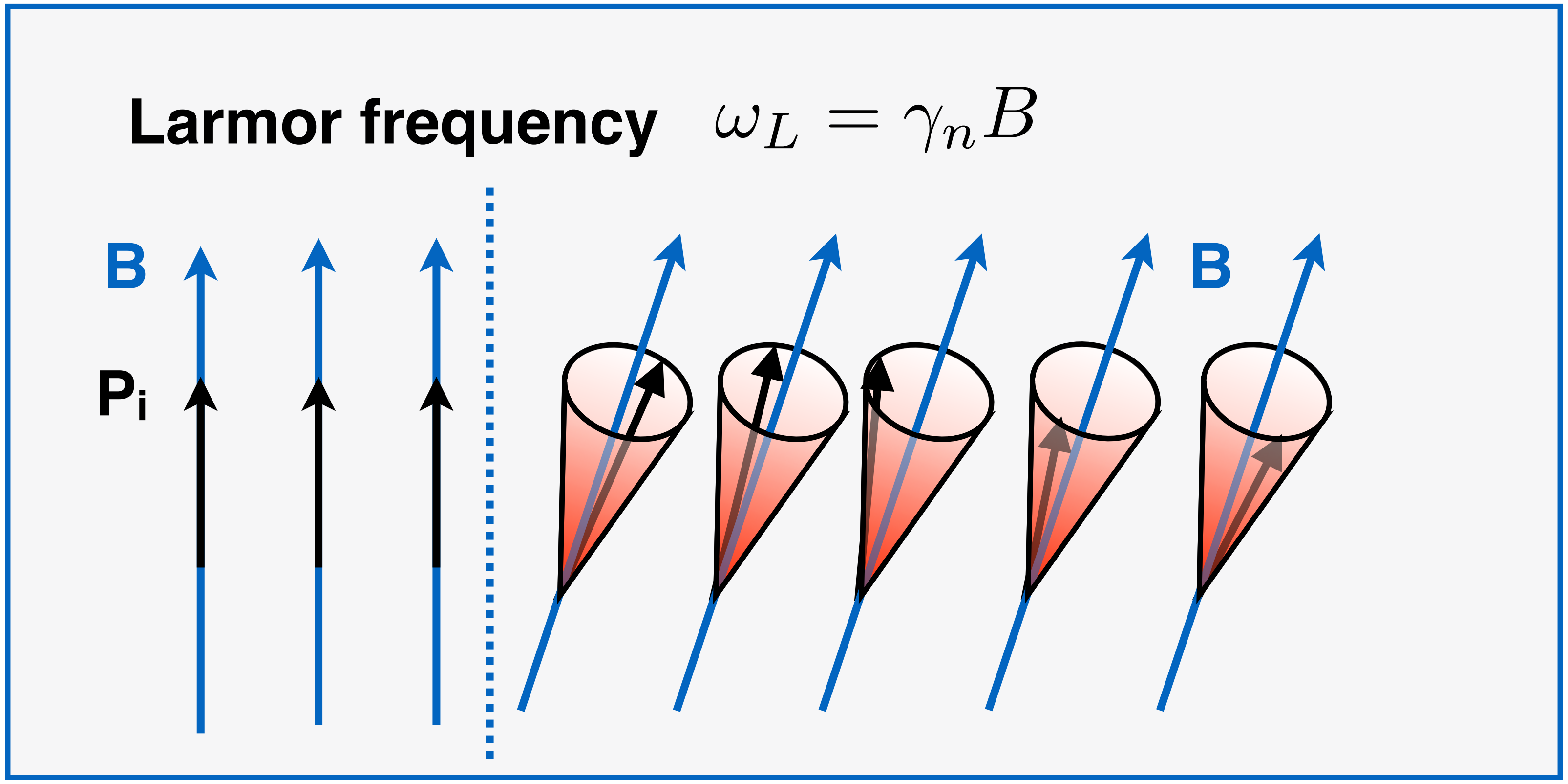


Require high ^{3}He polarization for good neutron polarization → lasers!

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.

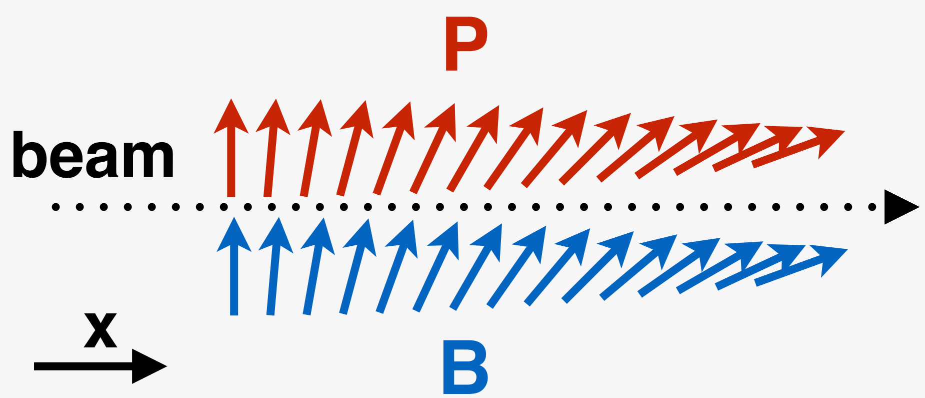
Manipulating the polarization

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:

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

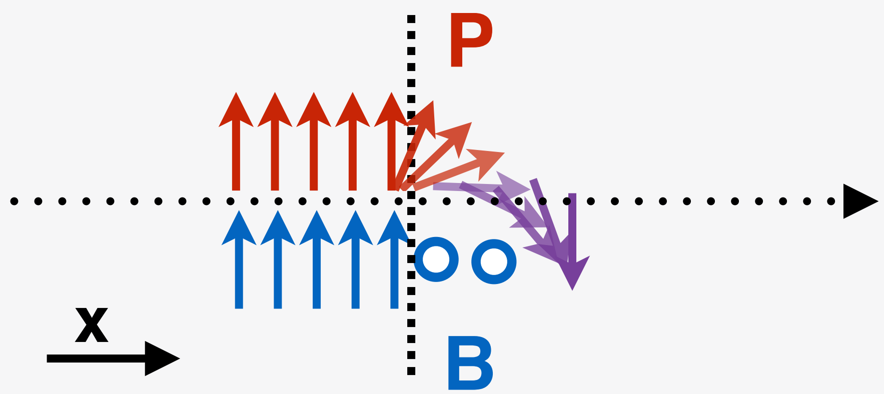
Adiabatic (A > 10)

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

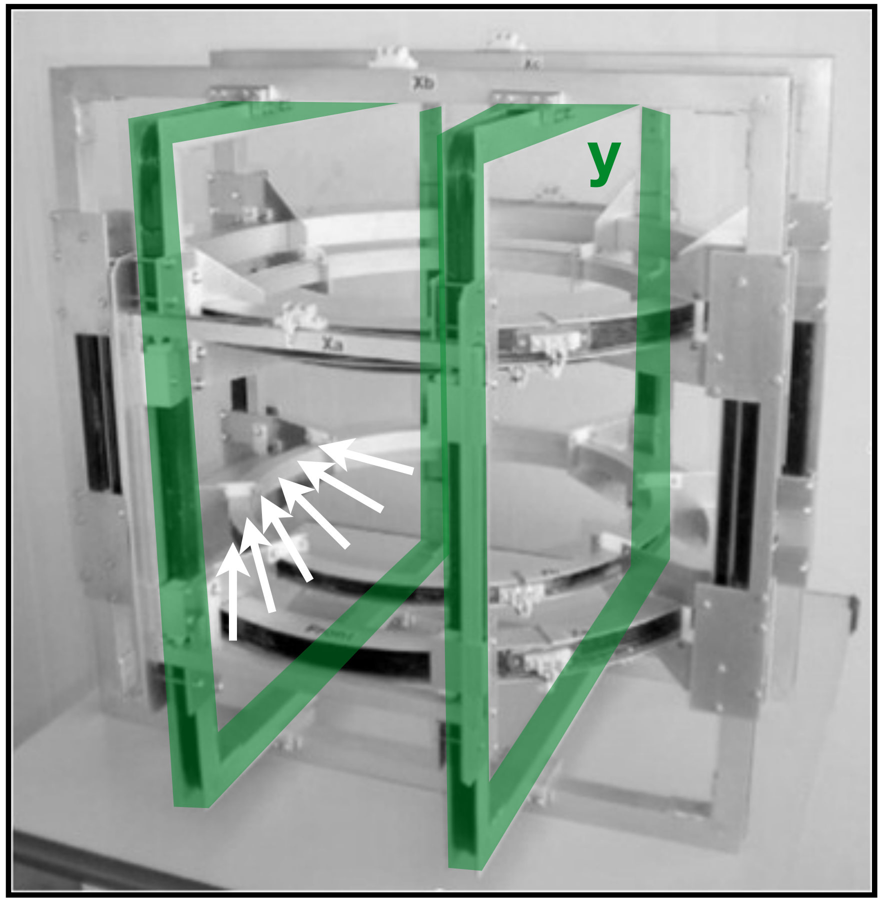


Photo: R. Stewart

Guide field

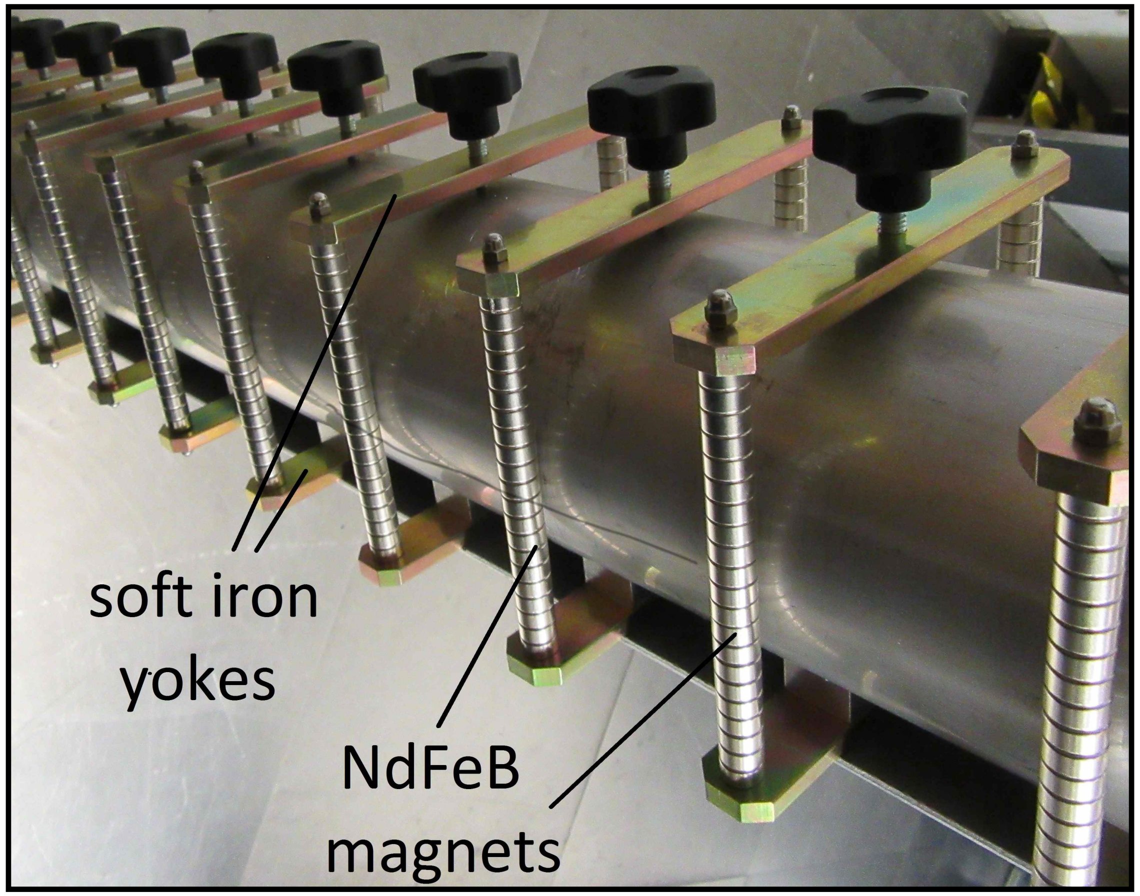
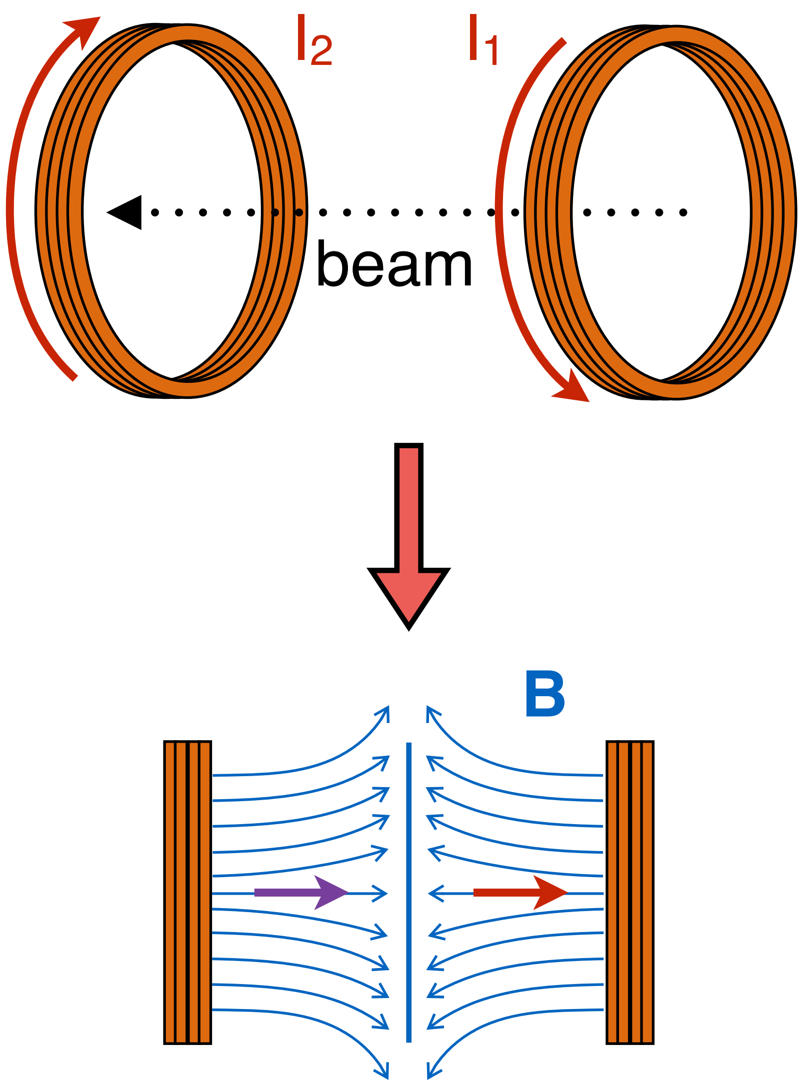


Photo: J. Kosata

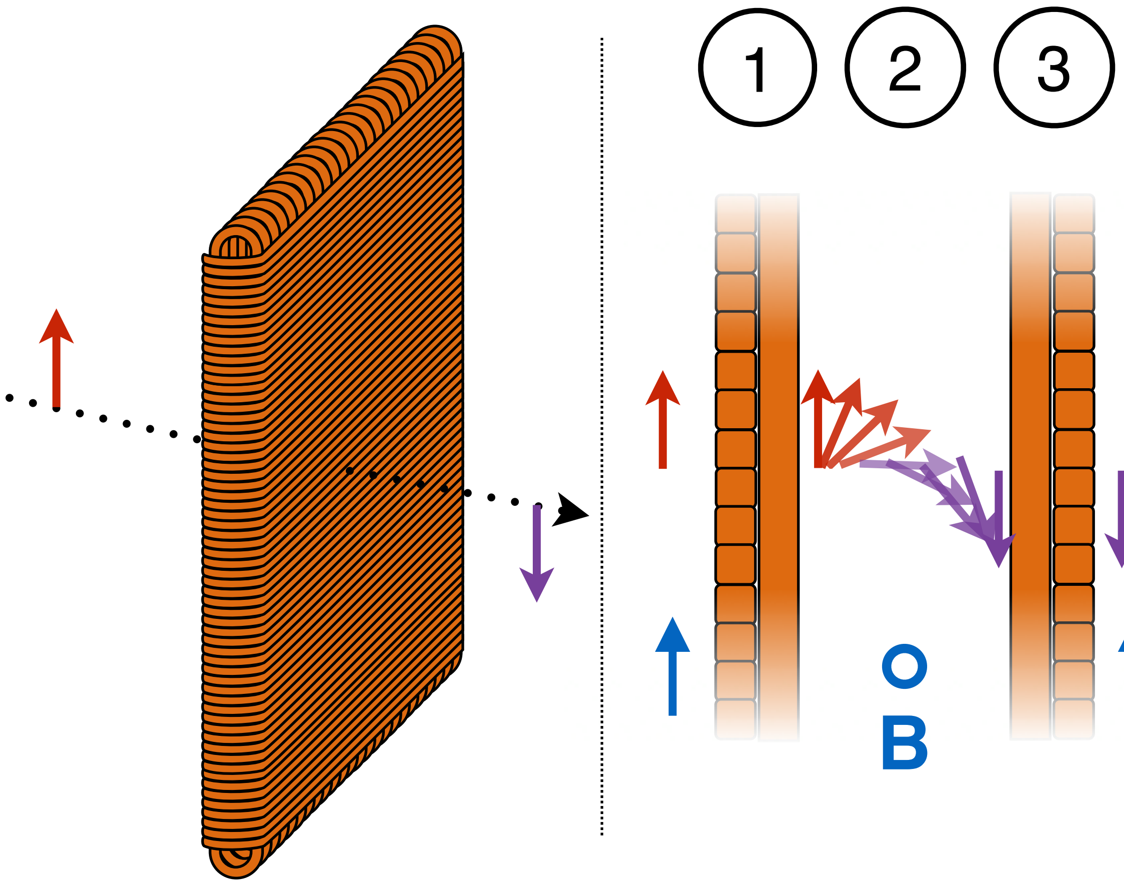
Spin flippers: a few examples

Drabkin



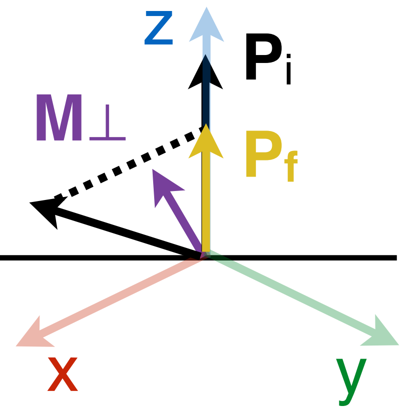
Field changes direction in the middle.

Mezei



1. Non-adiabatic transition
2. Precession (π)
3. Non-adiabatic transition

Techniques and applications



Reminder: rules of polarised neutron scattering

Nuclear

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

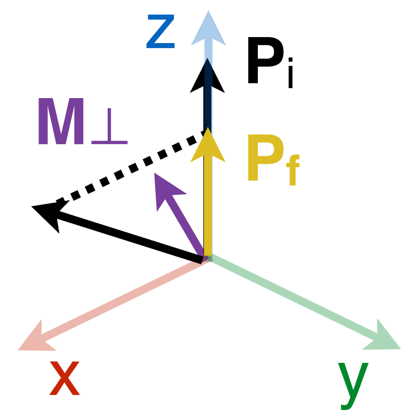
2. The spin incoherent scattering is 1/3 **NSF** and 2/3 **SF**

Magnetic

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

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

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

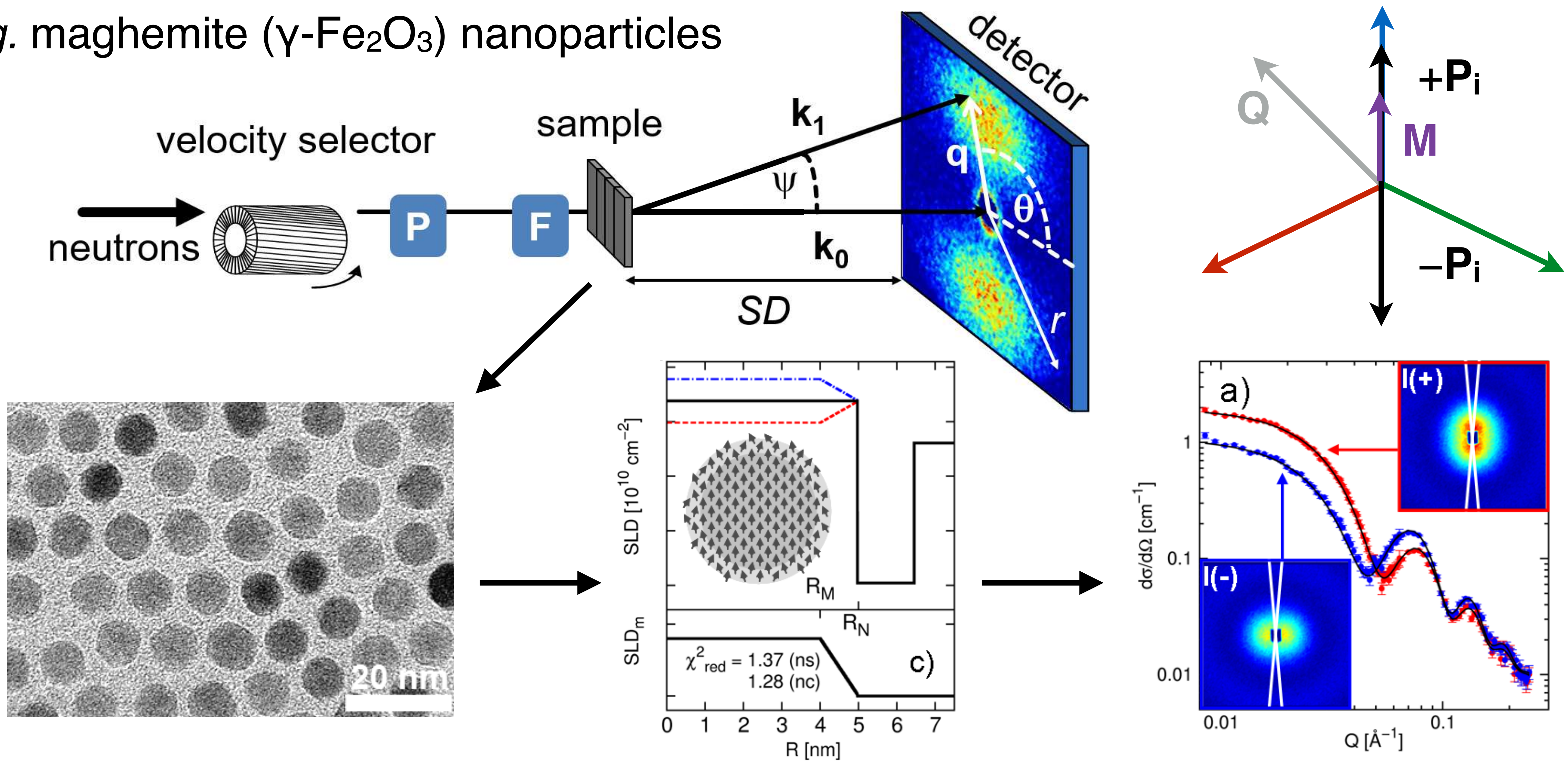


“Half”-polarized techniques

The simplest implementation involves just a polarizer and flipper. These techniques typically rely on nuclear-magnetic interference (Example 3):

POLSANS

e.g. maghemite (γ-Fe_2O_3) nanoparticles



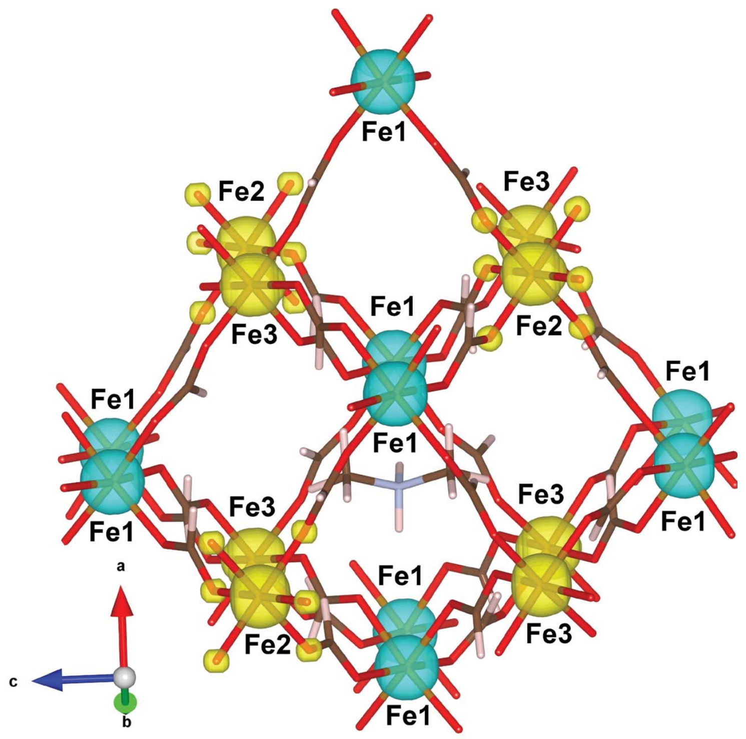
Muhlbauer et al.; Disch et al.

"Half"-polarized techniques

The simplest implementation involves just a polarizer and flipper. These techniques typically rely on nuclear-magnetic interference (Example 3):

Spin density

e.g. $[NH_2(CH_3)_2][Fe^{3+}Fe^{2+}(HCOO)_6]$

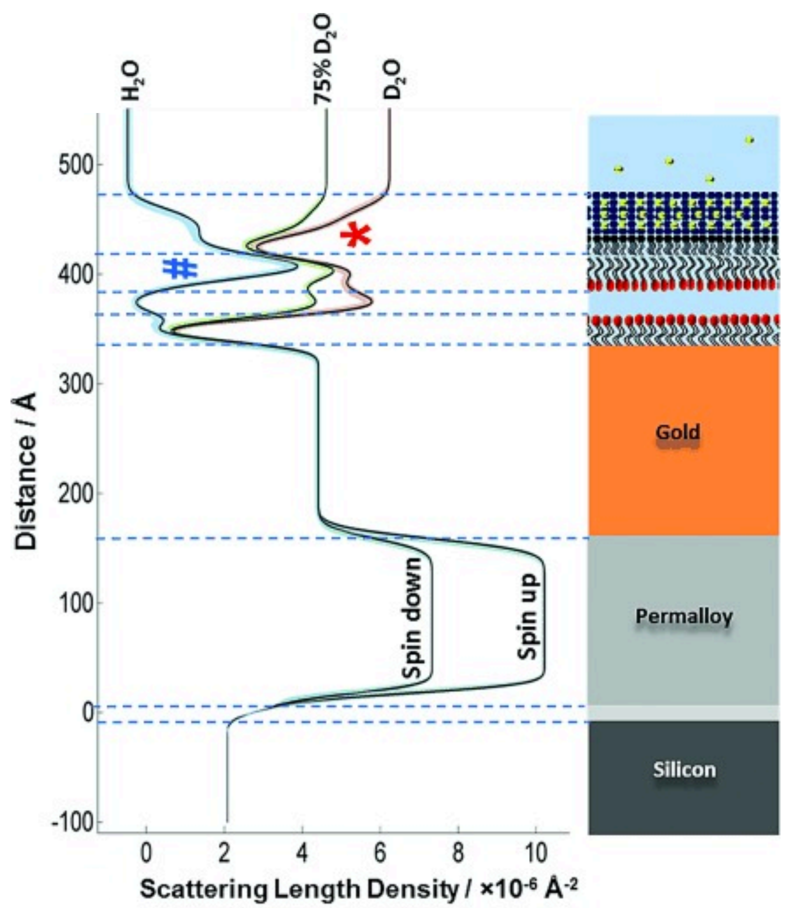


$$\left(\frac{d\sigma}{d\Omega}\right)_{\pm\pm} \propto |F_N \pm F_M|^2$$

Canadillas-Delgado et al.

Polarized neutron reflectometry

e.g. model E-coli lipid membrane



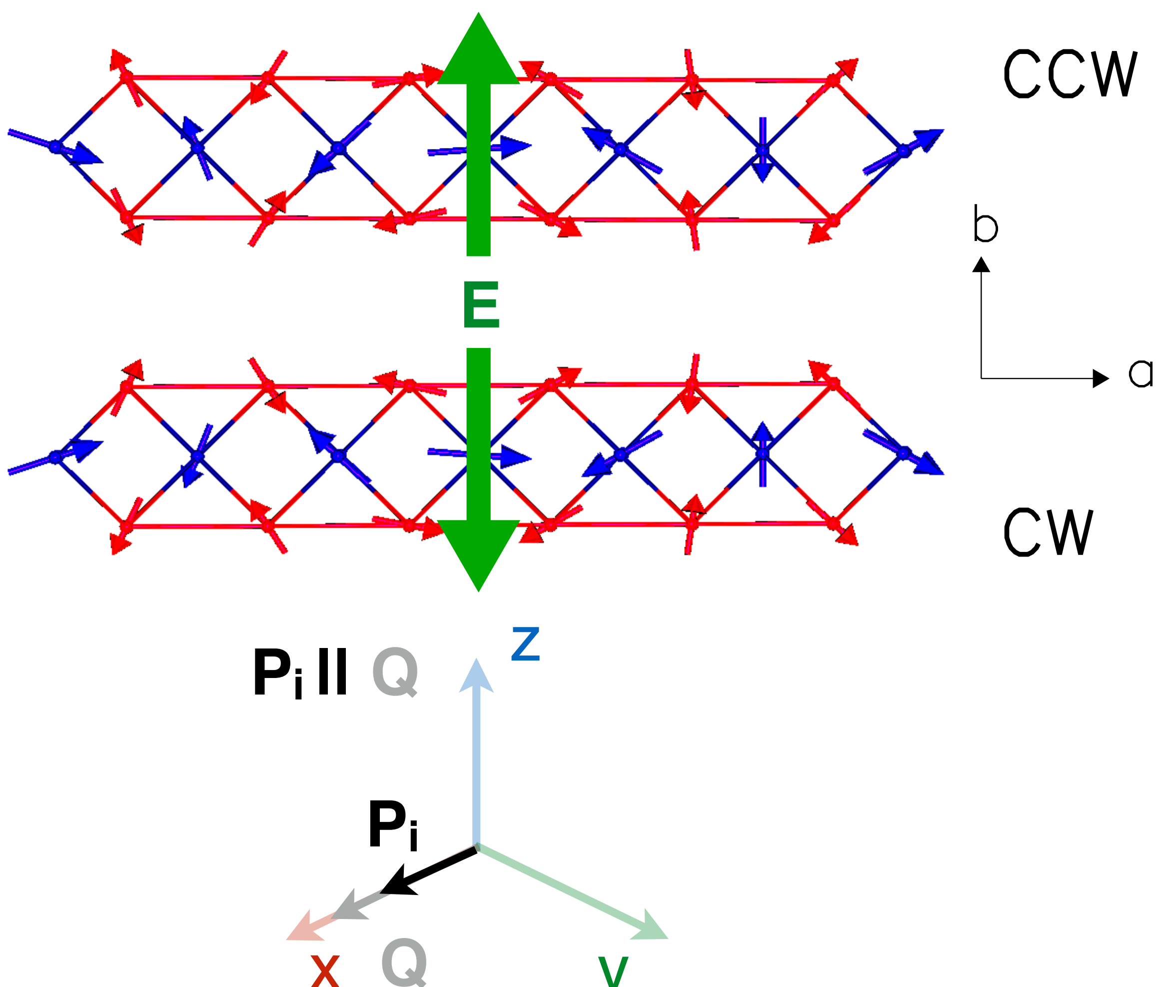
Clifton et al.; see S. Langridge for other examples

Longitudinal polarization analysis: chiral scattering

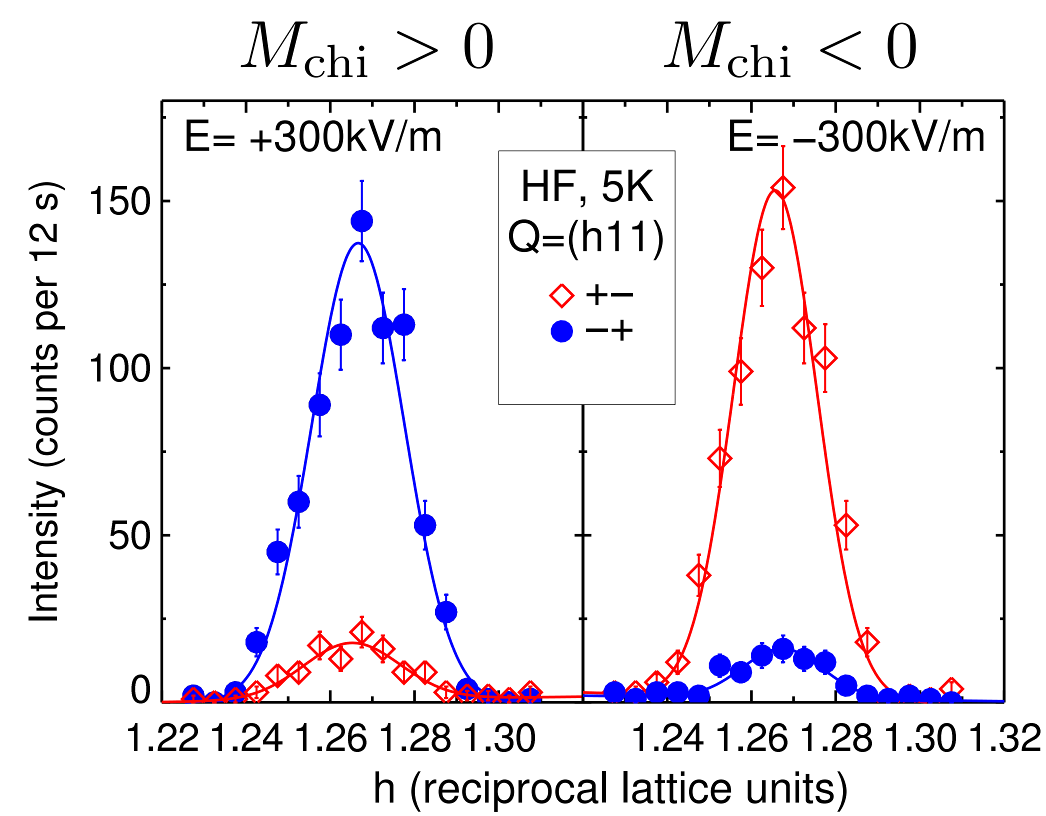
Beyond component separation, longitudinal polarization analysis is also able to observe cross section components that are invisible to unpolarized neutrons:

Chiral scattering

e.g. multiferroic $Ni_3V_2O_8$



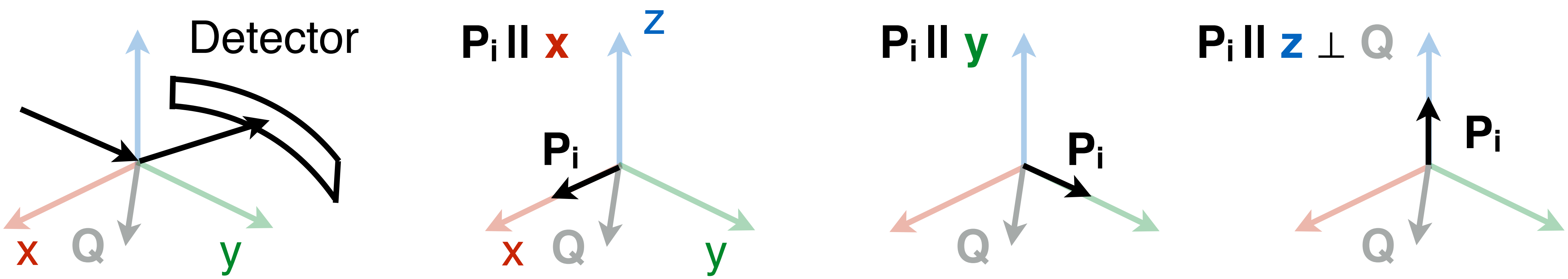
$$\left(\frac{d\sigma}{d\Omega}\right)_{+-}^{\mathbf{P}_i||\mathbf{Q}} \propto |M_\perp^{\perp\mathbf{P}_i}|^2 - PM_{chi}$$



Cabrera et. al.; see R. Johnson lecture

2D XYZ polarization analysis

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



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

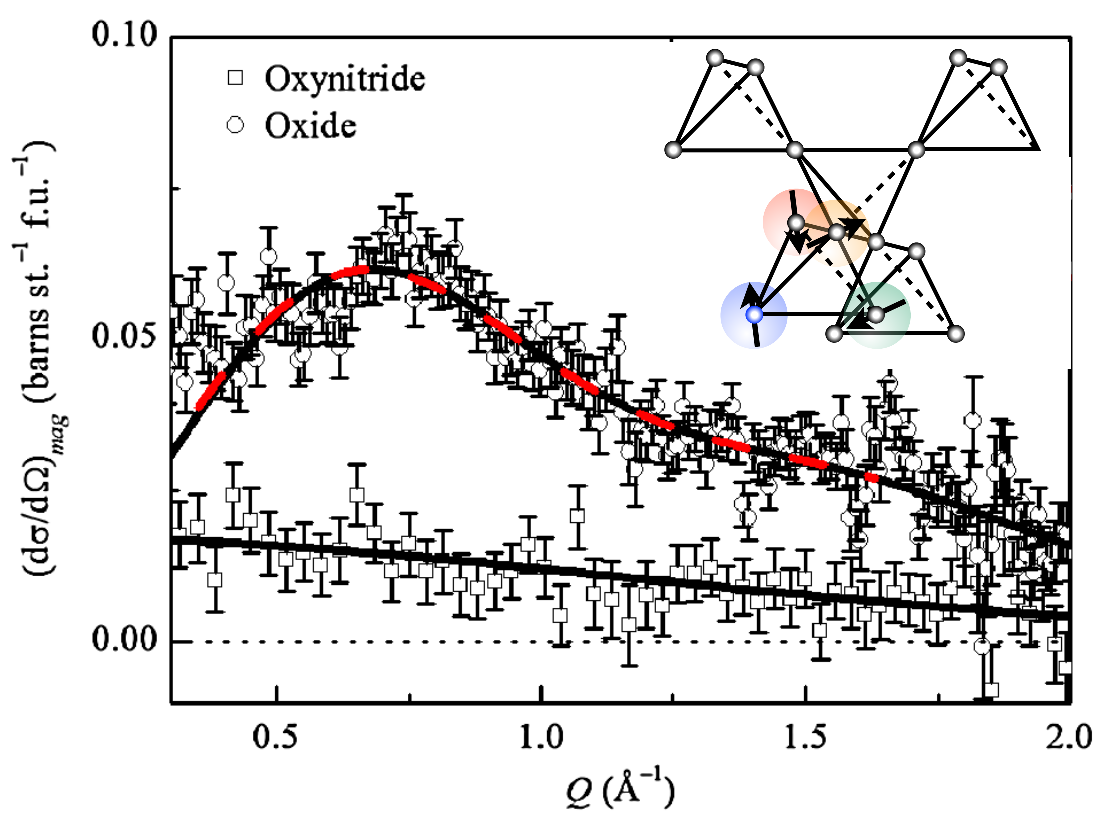
	$\mathbf{P}_i \parallel x$	$\mathbf{P}_i \parallel y$	$\mathbf{P}_i \parallel z \perp Q$
Magnetic	$1/2(\cos^2\alpha)$: $1/2(\cos^2\alpha+1)$	: $1/2(\sin^2\alpha+1)$	1/2 : 1/2
Spin incoherent	1/3 : 2/3	1/3 : 2/3	1/3 : 2/3
Nuclear	1 : 0	1 : 0	1 : 0

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 or distinguish magnetization components in magnetically disordered systems:

Frustrated magnets

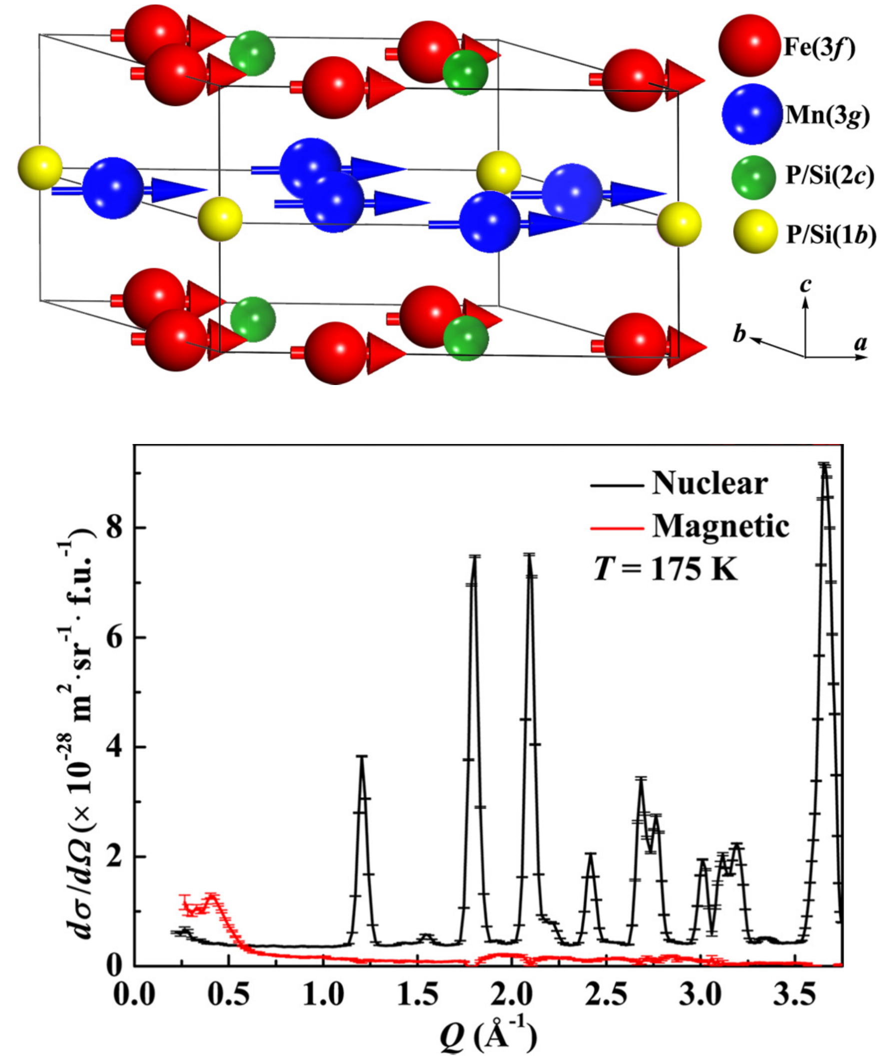


Magnetic frustration destroys magnetic order, only short range order

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

Clark et. al.

Magnetocaloric materials



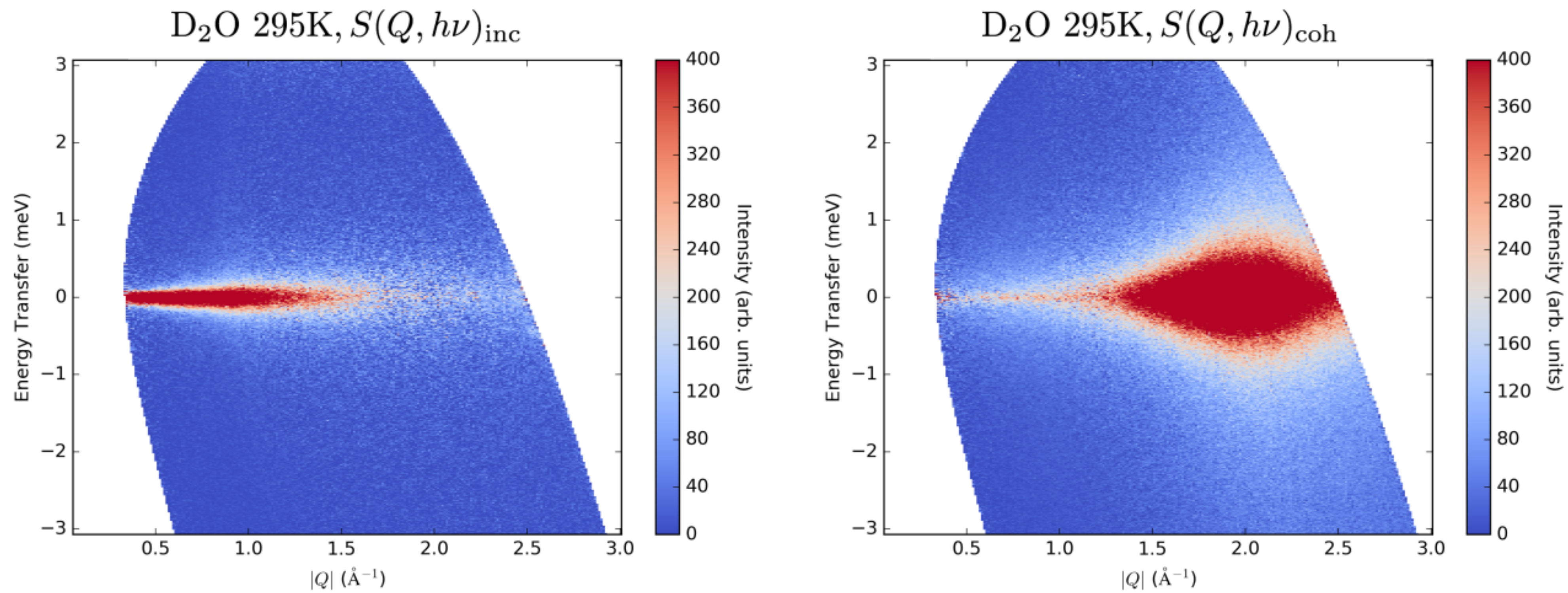
Miao et. al.

Longitudinal polarization analysis

We have already looked at a few examples of longitudinal PA. Wide-angle LPA has recently come into more widespread use for inelastic scattering:

Polarized spectroscopy

e.g. Coherent and incoherent dynamics in D_2O

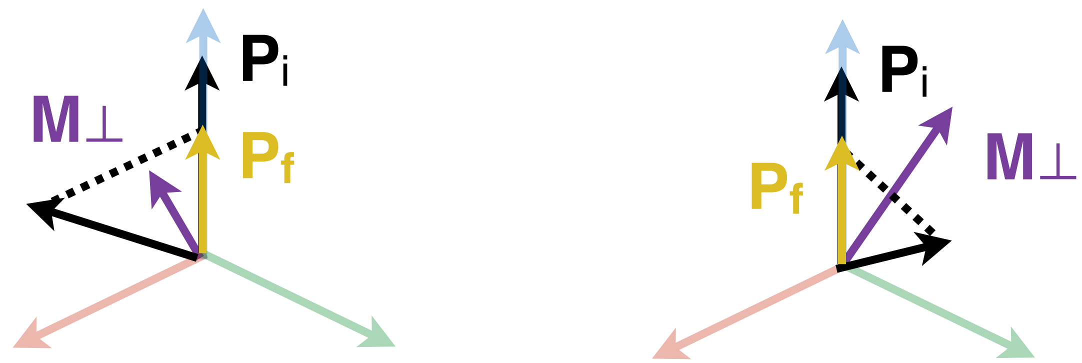


$S_{inc}(Q,E)$ contains the collective (and single-molecule) dynamics while ***$S_{inc}(Q,E)$*** contains only the single-molecule motions. This has resulted in a revision of the model for the dynamics in water.

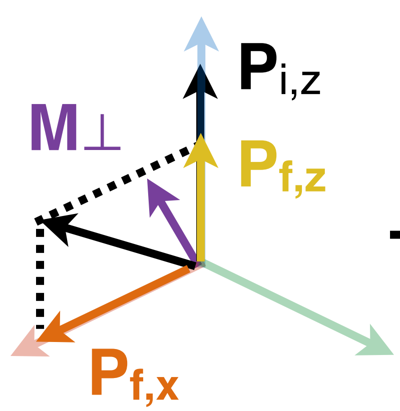
Arbe et. al.

Spherical polarimetry

In some cases, the 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 doing **spherical polarimetry**

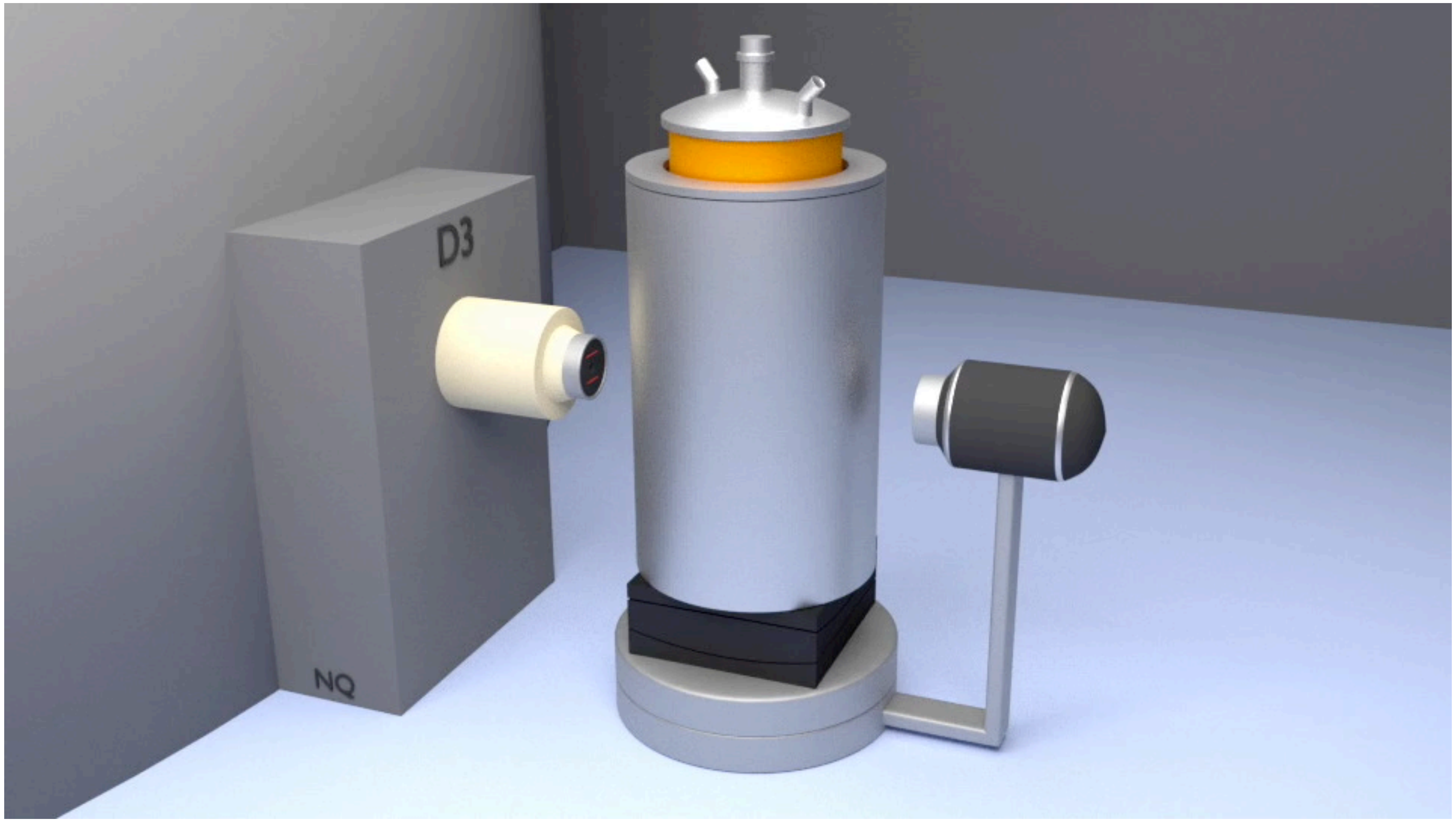


$$\longrightarrow \begin{pmatrix} P_{f,x} \\ P_{f,y} \\ P_{f,z} \end{pmatrix} = \begin{pmatrix} P_{xx} & P_{xy} & \boxed{P_{xz}} \\ P_{yx} & P_{yy} & P_{yz} \\ P_{zx} & P_{zy} & \boxed{P_{zz}} \end{pmatrix} \begin{pmatrix} P_{i,x} \\ P_{i,y} \\ P_{i,z} \end{pmatrix}$$

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).

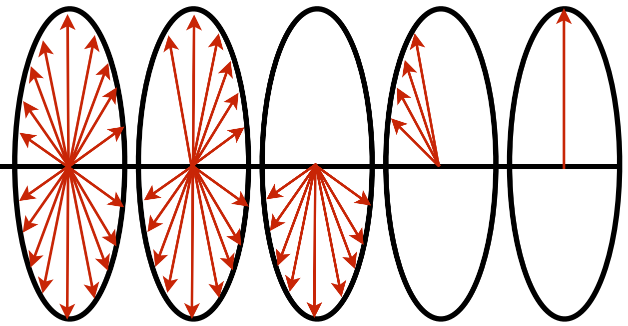
Spherical polarimetry

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

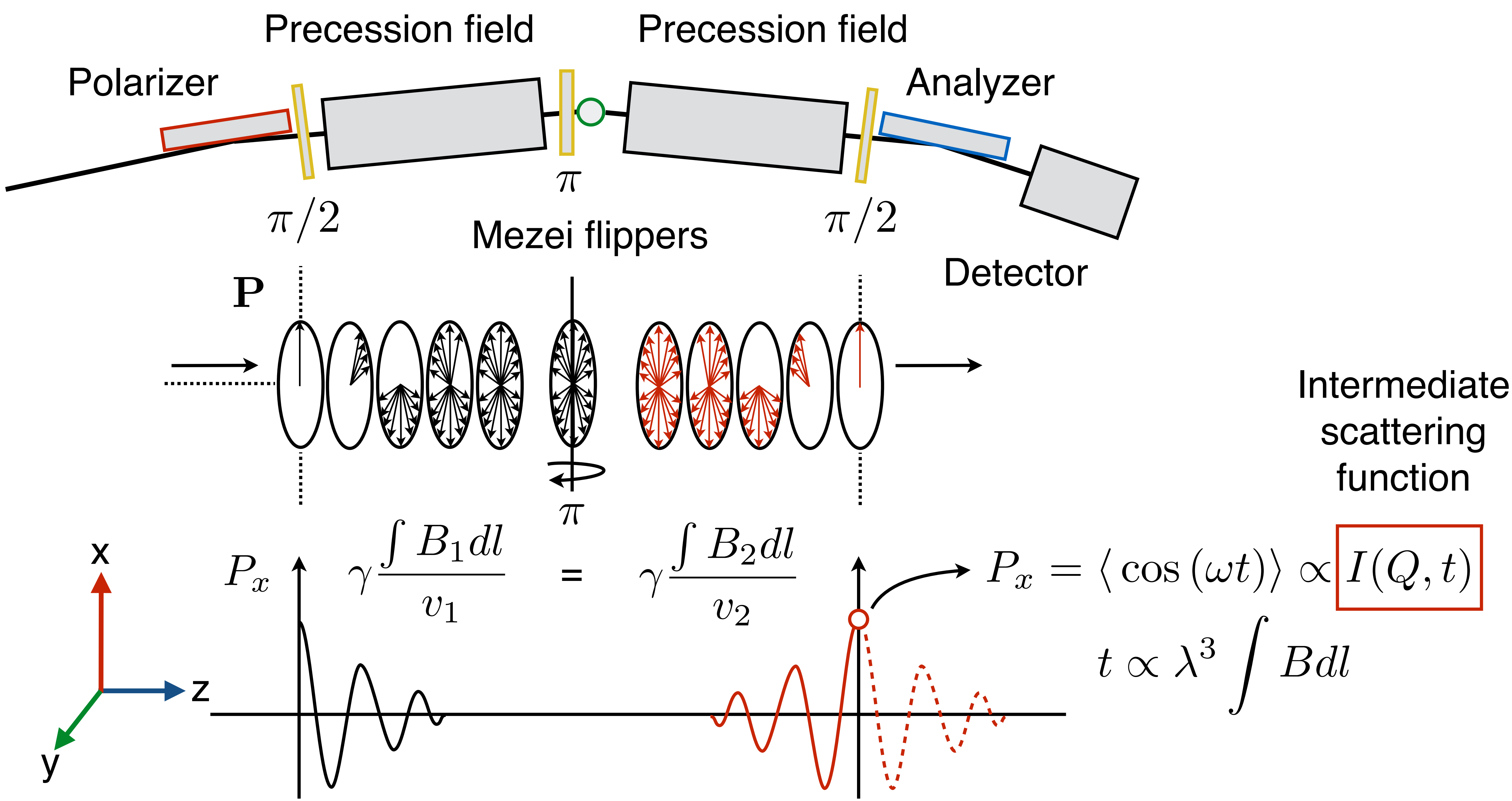


N. Qureshi; see R. Johnson lecture for examples

Neutron spin echo

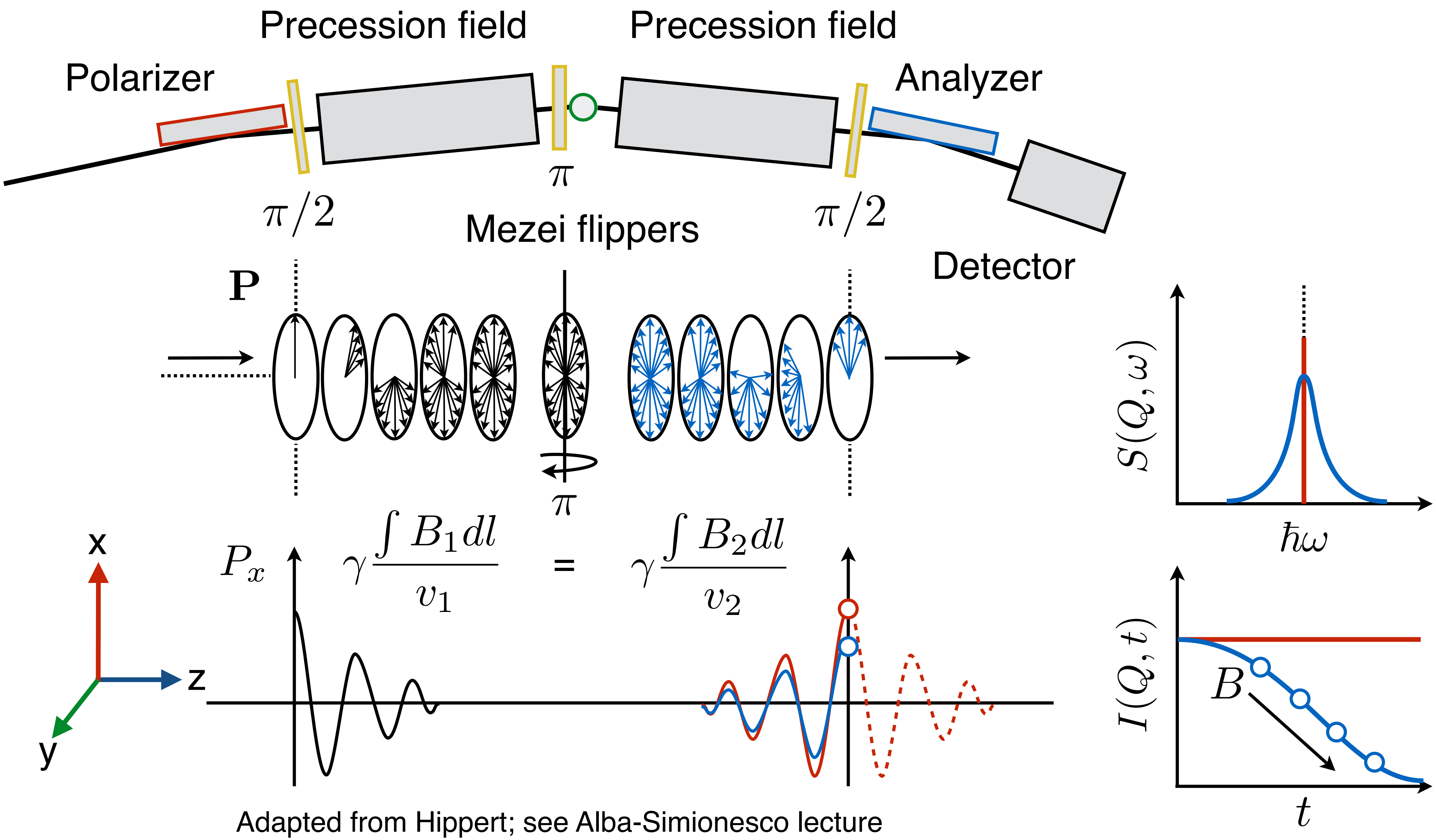


Principle: “classical” spin echo



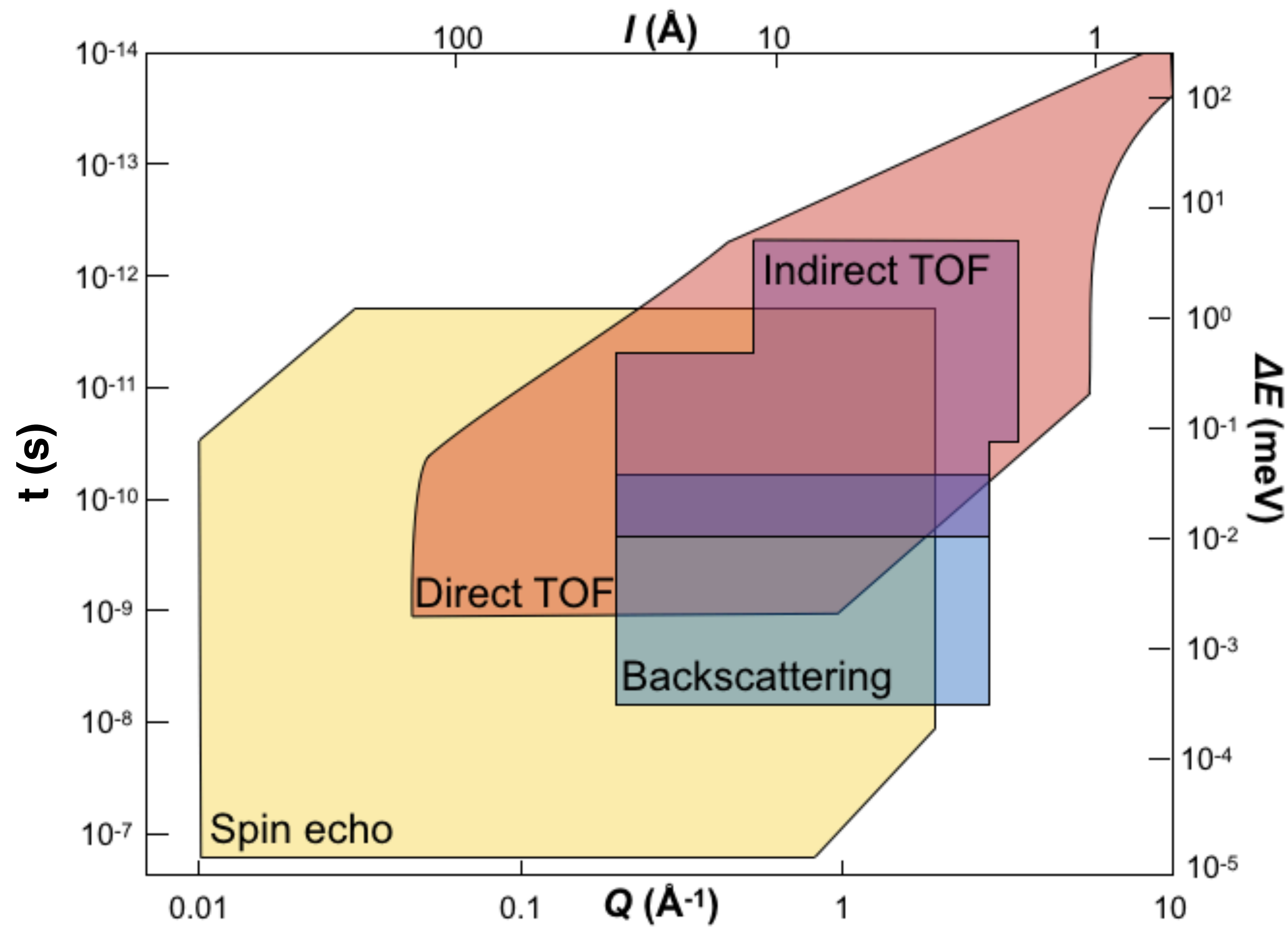
Adapted from Hippert; see Alba-Simionesco lecture

Principle: “classical” spin echo



Adapted from Hippert; see Alba-Simionesco lecture

Resolution

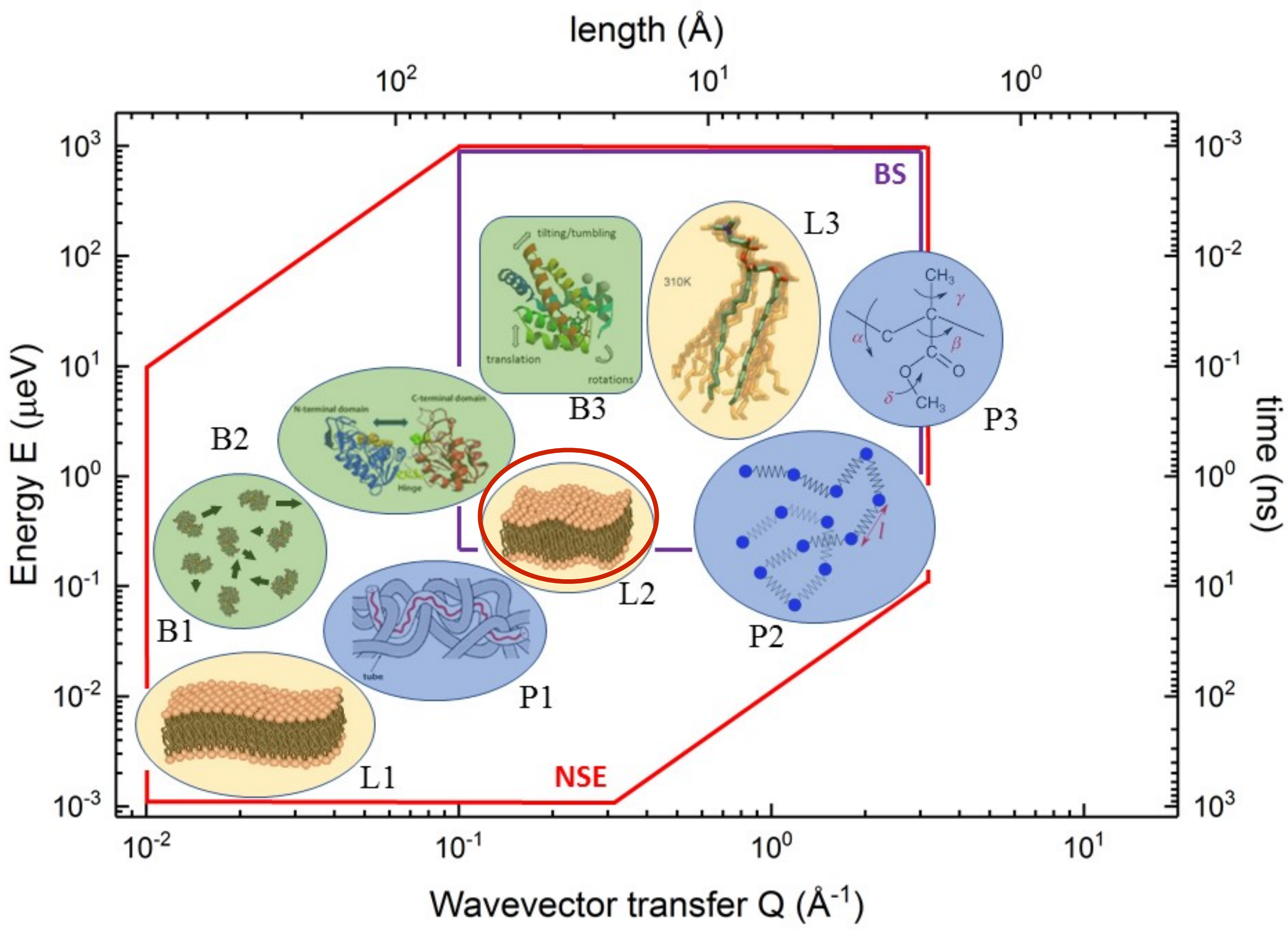


l (Å)
100
10
1
10^{-14}
10^{-13}
10^{-12}
10^{-11}
10^{-10}
10^{-9}
10^{-8}
10^{-7}
t (s)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
ΔE (meV)
Indirect TOF
Direct TOF
Backscattering
Spin echo
0.01
0.1
Q (Å⁻¹)
1
10

Example

Spin echo is frequently used to observe “slow” (~ns-μs) dynamics in polymers and biological systems, as well as magnetic systems (with some modifications…)

Dynamics in soft matter and biological systems



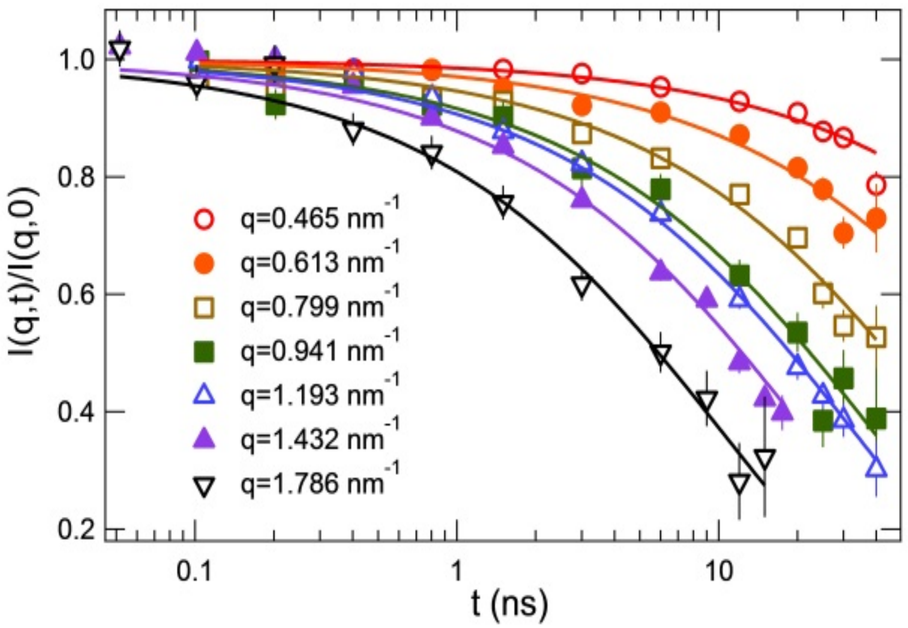
Gardner et. al.

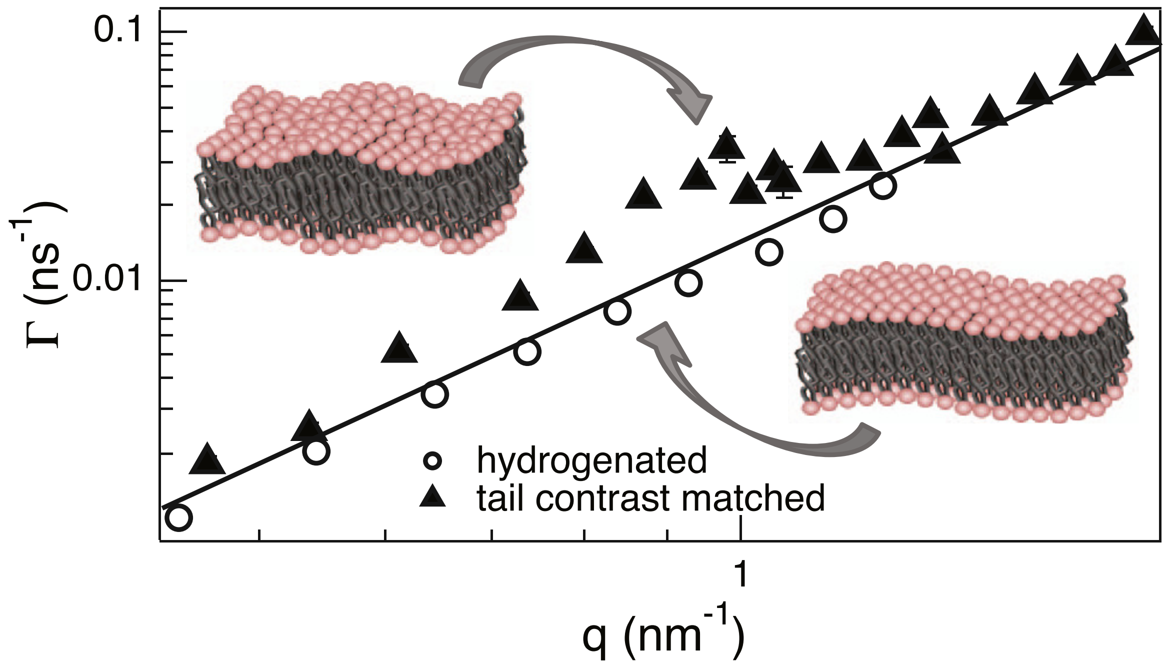
Example

Spin echo is frequently used to observe "slow" (~ns-μs) dynamics in polymers and biological systems, as well as magnetic materials (with some modifications)

Dynamics in soft matter and biological systems

e.g. Thickness fluctuations in model lipid bilayer systems





Lipid bilayers make up the cell membranes of many cells. Their thickness fluctuations determine many aspects of cell function. The ns dynamics in a deuterated sample reveal these fluctuations.

Ashkar et. al.

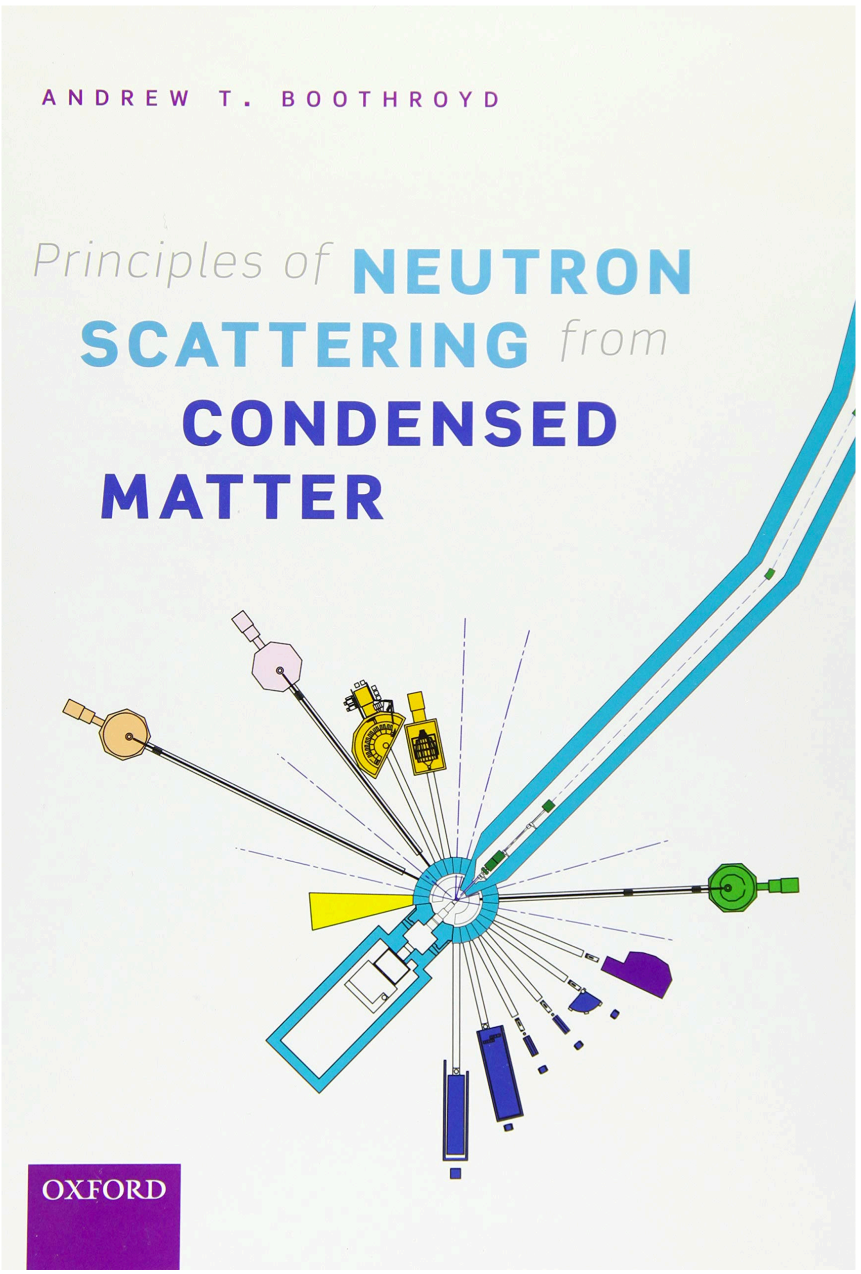
Conclusion

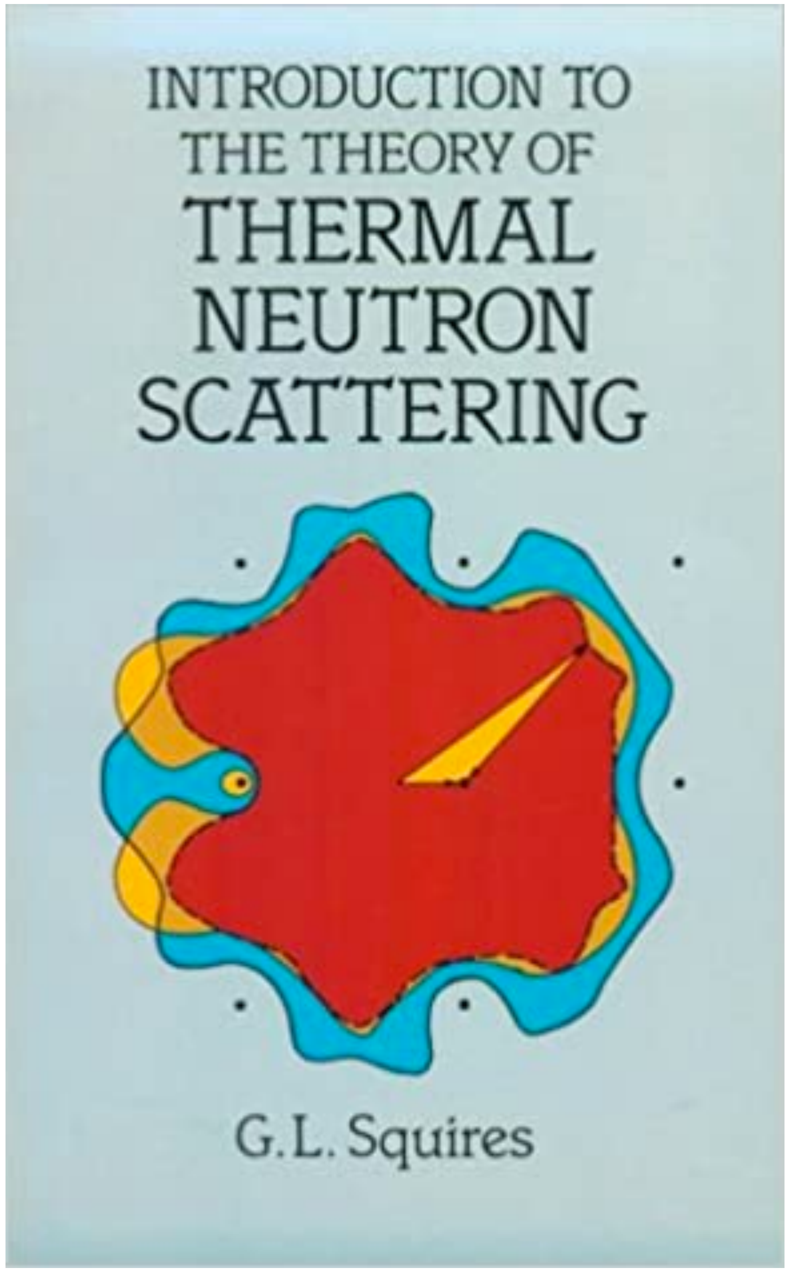
- Polarized neutron beams interact with magnetic moments (both nuclear and electronic) in samples. **The scattered polarization and cross section depends on the type of scattering process** (nuclear coherent, spin incoherent, or magnetic).

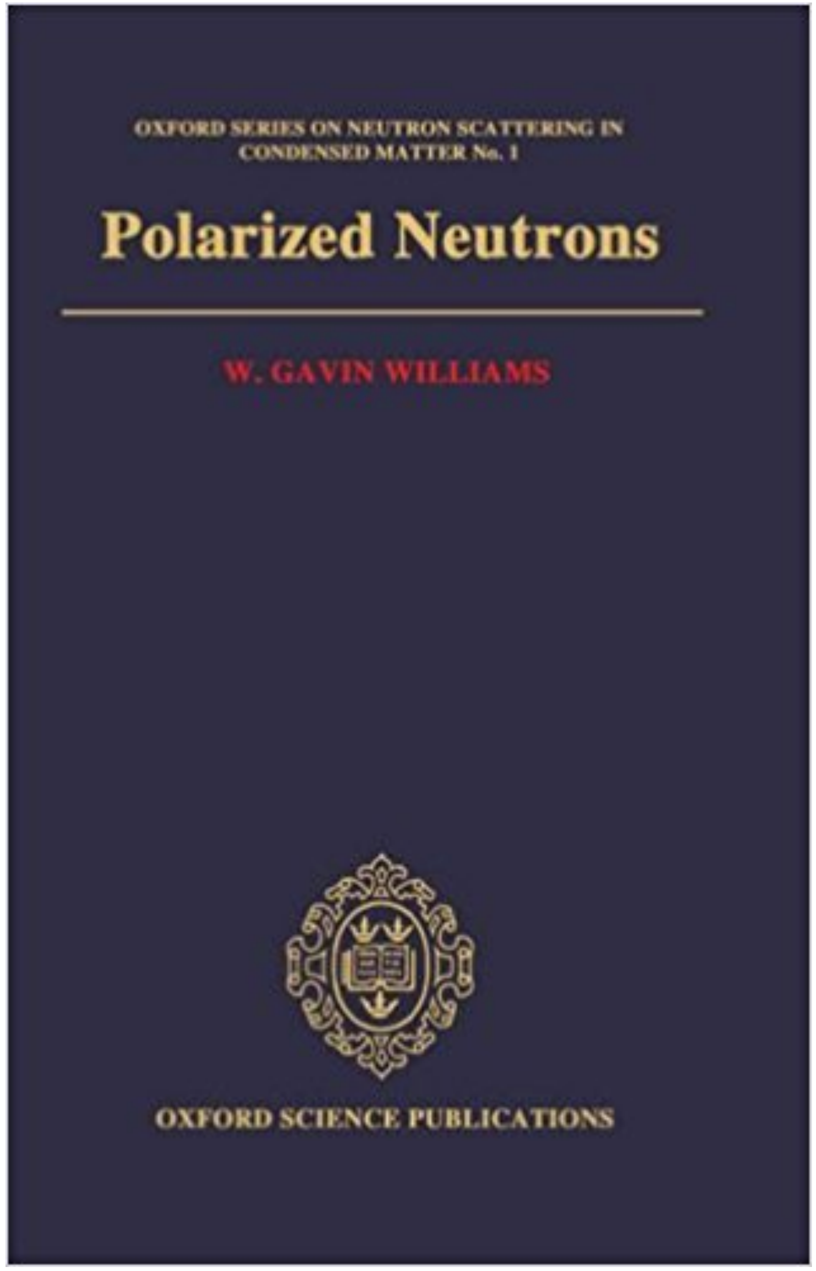
- Polarized neutron beams can therefore be used to:
  - **Separate cross section components**
  - **Determine magnetic moment orientations**
  - **Access parts of the cross section inaccessible to unpolarised neutrons**

- Polarized neutron beams can also be used to improve the resolution of neutron scattering by exploiting Larmor precession

Books







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

Polarized SANS: Mühlbauer Rev. Mod. Phys **13** (2014) 02002

Instrumentation

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

XYZ: Stewart et. al. J. Appl. Cryst. **42** (2009) 69

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

Spin echo: Gardner et al. Nature Reviews Phys. **2** (2020) 103

Further reading

Examples

D_2O dynamics: Arbe et al. Phys. Rev. Research **2** (2020) 012015

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

Magnetic contrast for lipid reflectometry: Clifton et al. Angew. Chem. **54** (2015) 11952

Magnetic nanoparticles: Disch et al. New J. Phys. **14** (2012) 013025

Magnetocaloric $(Mn,Fe)_2(P,Si)$: Miao et al. Phys. Rev. B 94 (2016) 014426

Membrane dynamics: Ashkar et al. Biophys. J. **109** (2015) 106

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

Multiferroic spin density: Canadillas-Delgado et al. IUCrJ **7** (2020) 803