Polarized Neutrons
Intro and Techniques
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Polarized neutron beams

Each individual neutron has spin $s = \frac{1}{2}$ and an angular momentum of $\pm \frac{1}{2}\hbar$

Each neutron has a spin vector $\vec{s}_n$ and we define the polarization of a neutron beam as the ensemble average over all the neutron spin vectors, normalised to their modulus

$$\vec{P} = \frac{\langle \vec{s}_n \rangle}{\frac{1}{2}} = 2\langle \vec{s}_n \rangle$$

If we apply an external field (quantisation axis) then there are only two possible orientations of the neutrons: parallel and anti-parallel to the field. The polarization can then be expressed as a scalar:

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

where there are $N_+$ neutrons with spin-up and $N_-$ neutrons with spin-down
**History of Polarized Neutrons**

1932  Discovery of the neutron  
Chadwick (*Proc Roy Soc* A136 692)

1937  Theory of neutron polarization by a ferromagnet  
Schwinger (*Phys Rev*, 51, 544)

1938  Partial polarization of a neutron beam by passage through iron  

1940  Magnetic moment of the neutron determined by polarization analysis  
Alvarez and Bloch (*Phys Rev* 57, 111)

- 1941  **Theory of magnetic neutron scattering**  
Halpern and Johnson (*Phys Rev* 51, 992; 52, 52; 55, 898)

1951  Polarizing mirrors, proof of the neutron’s μ·B interaction  
Hughes and Burgy (*Phys Rev*, 81, 498)
History of Polarized Neutrons

1951 Polarizing crystals (magnetite Fe$_3$O$_4$, Co$_{92}$Fe$_8$)

1959 First polarized beam measurements (of magnetic form factors of Ni and Fe)

1963 General theory of neutron polarization analysis
Blume (*Phys Rev* **130**, 1670)

1969 First implementation of neutron polarization analysis, Oak Ridge, USA
History of Polarized Neutrons

1972  Invention of neutron spin echo (IN11, ILL)
      Mezei (Z Phys Rev 255, 146)

1982  XYZ polarization analysis on a multidetector spectrometer (D7, ILL)
      Schärpf (AIP Conf. Proc. 89, 175)

1987  Invention of neutron resonance spin-echo (leading to SESANS, MIEZE, ...)
      Golub and Gähler (Phys. Lett. A 123, 43)

1988  Development of neutron polarimetry measurements with CRYOPAD
      Tasset et. al. (J. Appl. Phys. 63, 3606)

2000  Routine use of $^3$He neutron spin-filters for polarizing neutrons
Polarized neutrons today

- Single crystal diffraction
- Diffuse scattering
- Inelastic scattering (3-axis and TOF)
- Reflectometry (on and off-specular)
- SANS - magnetic and non-magnetic
- Neutron Spin-Echo
- Neutron Resonance Spin-Echo
- SESANS
- Larmor Diffraction
- Neutron Depolarization
- Polarized Neutron Tomography
- ....
Polarized neutron beams

What we often would like to do in polarized neutron experiments is measure the scalar polarization of the beam.

\[ P = \frac{N_+ - N_-}{N_+ - N_-} \]

\[ = \frac{(N_+/N_-) - 1}{(N_+/N_-) + 1} \]

\[ = \frac{F - 1}{F + 1} \]

Where \( F = \frac{N_+}{N_-} \) is called the Flipping Ratio and is a measurable quantity in a scattering experiment.

This description of a polarized beam is OK for experiments in which a single quantisation axis is defined: **Longitudinal Polarization Analysis**

The technique of 3-dimensional neutron polarimetry, however is termed: **Vector (or Spherical) Polarization Analysis**
A Uniaxial PA experiment

First attempted by Moon, Riste and Koehler (Oak Ridge 1969)

Phys Rev. 181 (1969) 920
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Polarizers

Stern-Gerlach experiment (1922)

Cu$_2$MnAl (Heusler) crystal grown at ILL

Supermirror systems (eg Co/Ti, Fe/Si etc)

3He spin-filter
**Flippers**

Drabkin flipper: useful for white beams of limited size

Dabbs Foil: “current sheet”

Mezei Flipper: “current sheet”

AFP Flipper: “adiabatic fast passage”
Uniaxial Polarization Analysis
Neutron polarization and scattering

We start with the (elastic - $|k_i| = |k_f|$) scattering cross-section

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 |\langle k'S'|V|kS\rangle|^2$$

Where the spin-state of the neutron $S$ is either spin-up $|\uparrow\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ or spin down $|\downarrow\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$

For nuclear scattering (no spin) $V$ is the Fermi pseudopotential, and the matrix element is

$$\langle S'|b|S\rangle = b\langle S'|S\rangle = \begin{cases} b \begin{pmatrix} \uparrow \rightarrow \uparrow \\ \downarrow \rightarrow \downarrow \end{pmatrix} & \text{Non-spin-flip} \\ 0 \begin{pmatrix} \uparrow \rightarrow \downarrow \\ \downarrow \rightarrow \uparrow \end{pmatrix} & \text{Spin-flip} \end{cases}$$

where we have used the fact that the spin states are orthogonal and normalised

$$\langle \uparrow|\downarrow\rangle = \langle \downarrow|\uparrow\rangle = 0, \quad \langle \uparrow|\uparrow\rangle = \langle \downarrow|\downarrow\rangle = 1$$
Neutron polarization and magnetic scattering

V is the magnetic scattering potential given by

\[ V_m(Q) = -\frac{\gamma_{n}r_{0}}{2\mu_{B}} \sigma \cdot M_{\perp}(Q) = -\frac{\gamma_{n}r_{0}}{2\mu_{B}} \sum_{\zeta} \sigma_{\zeta} \cdot M_{\perp\zeta}(Q) \]  (see e.g. Squires)

where \( \zeta = x, y, z \). Here \( M_{\perp}(Q) \) represents the component of the Fourier transform of the magnetisation of the sample, which is perpendicular to the scattering vector \( Q \) - i.e. the neutron sensitive part. \( \sigma_{\zeta} \) are the Pauli spin matrices

\[
\begin{align*}
\sigma_x &= \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}
\end{align*}
\]

Substitution of these into the magnetic potential gives us the matrix elements

\[
\langle S'|V_m(Q)|S \rangle = -\frac{\gamma_{n}r_{0}}{2\mu_{B}} \begin{cases} 
M_{\perp z}(Q) & \uparrow \rightarrow \uparrow \\
-M_{\perp z}(Q) & \downarrow \rightarrow \downarrow \\
M_{\perp x}(Q) - iM_{\perp y}(Q) & \uparrow \rightarrow \downarrow \\
M_{\perp x}(Q) + iM_{\perp y}(Q) & \downarrow \rightarrow \uparrow 
\end{cases}
\]

Non-spin-flip  Spin-flip
Magnetic scattering rule

The non-spin-flip scattering is sensitive only to those components of the magnetisation parallel to the neutron spin

The spin-flip scattering is sensitive only to those components of the magnetisation perpendicular to the neutron spin

NB This is one of those points that you should take away with you. It is the basis of all magnetic polarization analysis techniques.
Neutron polarization and nuclear scattering

In general a bound state is formed between the nucleus and the neutron during scattering with either spins antiparallel (spin-singlet) or spins parallel (spin-triplet). The scattering lengths for these situations are different and are termed \( b_- \) and \( b_+ \).

The scattering length operator is

\[
\hat{b} = A + B \sigma \cdot I
\]

\[
A = \frac{(I+1)b_+ + Ib_-}{2I+1}, \quad B = \frac{b_+ - b_-}{2I+1}
\]

(see e.g. Squires, p173)

The calculation of the matrix elements now proceeds analogously to the case of magnetic scattering

\[
\langle S' | \hat{b} | S \rangle = \left\{ \begin{array}{c}
A + BI_z \quad \uparrow \rightarrow \uparrow \\
A - BI_z \quad \downarrow \rightarrow \downarrow \\
B(I_x - iI_y) \quad \uparrow \rightarrow \downarrow \\
B(I_x + iI_y) \quad \downarrow \rightarrow \uparrow 
\end{array} \right\}
\]

Non-spin-flip

Spin-flip

Since the nuclear spins are (normally) random \( \langle I_x \rangle = \langle I_y \rangle = \langle I_z \rangle = 0 \)

Therefore with the coherent scattering amplitude proportional to \( \overline{b} \), we can write

\[
\overline{b} = A \quad \text{i.e. the coherent scattering is entirely non-spin-flip}
\]
Moon-Riste-Koehler Equations

Bringing all this together, we get

\[ |\uparrow\rangle \rightarrow |\uparrow\rangle = \bar{b} - \frac{\gamma n r_0}{2 \mu_B} M_{\perp z} + BI_z \]

\[ |\downarrow\rangle \rightarrow |\downarrow\rangle = \bar{b} + \frac{\gamma n r_0}{2 \mu_B} M_{\perp z} - BI_z \]

\[ |\uparrow\rangle \rightarrow |\downarrow\rangle = -\frac{\gamma n r_0}{2 \mu_B} (M_{\perp x} - iM_{\perp y}) + B(I_x - iI_y) \]

\[ |\downarrow\rangle \rightarrow |\uparrow\rangle = -\frac{\gamma n r_0}{2 \mu_B} (M_{\perp x} + iM_{\perp y}) + B(I_x + iI_y) \]

Remember that: \[ \vec{M}_{\perp} (\vec{Q}) = \hat{Q} \times (\vec{M} (\vec{Q}) \times \hat{Q}) = \vec{M} (\vec{Q}) - (\vec{M} (\vec{Q}) \cdot \hat{Q}) \hat{Q} \]

If the polarization is parallel to the scattering vector, then the magnetisation in the direction of the polarization will not be observed since the magnetic interaction vector is zero. i.e. all magnetic scattering will be spin-flip.
Spin-incoherent scattering

Now, let’s take another look at the nuclear incoherent scattering. We know that this is given by \( \bar{b}^2 - \langle \bar{b} \rangle^2 \)

Applying this to the \( |\uparrow\rangle \rightarrow |\uparrow\rangle \) transition, and neglecting magnetic scattering, we get

\[
\bar{b}^2 = \langle (\bar{b} + B\hat{I}_z)^2 \rangle = \langle \bar{b} \rangle^2 + \langle B^2\hat{I}_z^2 \rangle + 2\langle \bar{b}B\hat{I}_z \rangle
\]

Now, for a randomly oriented distribution of nuclei of spin \( I \), we have

\[
\langle \hat{I} \rangle = \sqrt{I(I+1)} = \sqrt{I_x^2 + I_y^2 + I_z^2}
\]

\[
\Rightarrow I_x^2 = I_y^2 = I_z^2 = \frac{1}{3}I(I+1) \quad \text{since the distribution is isotropic}
\]

Therefore we can write

\[
\bar{b}^2 - \langle \bar{b} \rangle^2 = \langle \bar{b} \rangle^2 - \langle \bar{b} \rangle^2 + \frac{1}{3}B^2I(I+1)
\]

Isotope incoherent scattering

spin incoherent scattering

The other transitions are dealt with in a similar way
Moon-Riste-Koehler II

Finally, we get

\[
\begin{align*}
|\uparrow\rangle \rightarrow |\uparrow\rangle &= \bar{b} - \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} + b_{II} + \frac{1}{3} b_{SI} \\
|\downarrow\rangle \rightarrow |\downarrow\rangle &= \bar{b} + \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} + b_{II} + \frac{1}{3} b_{SI} \\
|\uparrow\rangle \rightarrow |\downarrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} \left( M_{\perp x} - i M_{\perp y} \right) + \frac{2}{3} b_{SI} \\
|\downarrow\rangle \rightarrow |\uparrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} \left( M_{\perp x} + i M_{\perp y} \right) + \frac{2}{3} b_{SI}
\end{align*}
\]

The details of the magnetic scattering will in general depend on the direction of the neutron polarization with respect to the scattering vector, and also on the nature of the orientation of the magnetic moments.
Scientific Examples
Polarized magnetic diffraction

For a ferromagnetic sample aligned in a field perpendicular to the scattering vector we have

\[ \vec{M}_\perp(\vec{Q}) = \vec{M}(\vec{Q}) - \left( \vec{M}(\vec{Q}) \cdot \hat{Q} \right) \hat{Q} = \vec{M}(\vec{Q}) \]

and \( M_\perp \) has no component in the xy-plane, so that the spin-flip scattering is zero. This implies that we don’t need to analyse the neutron spin, it will always end up in the same direction it started in. Therefore

\[
d\sigma/d\Omega = \left[ F_N(\vec{Q}) - F_M(\vec{Q}) \right]^2 \quad \text{for neutrons polarized parallel to the field}
\]

\[
d\sigma/d\Omega = \left[ F_N(\vec{Q}) + F_M(\vec{Q}) \right]^2 \quad \text{for neutrons polarized antiparallel to the field}
\]

where

\[
F_N(\vec{Q}) = \sum_i b_i \exp(i\vec{Q} \cdot \vec{r}_i)
\]

\[
F_M(\vec{Q}) = \gamma_n r_0 \sum_i g_{ij} J_i f_i(\vec{Q}) \exp(i\vec{Q} \cdot \vec{r}_i)
\]

Notice that to simulate an unpolarized measurement, we simply average the two polarized cross sections

\[
\frac{d\sigma}{d\Omega} = \frac{1}{2} \left[ (F_N(\vec{Q}) - F_M(\vec{Q}))^2 + (F_N(\vec{Q}) + F_M(\vec{Q}))^2 \right]
\]

\[
= F_N^2(\vec{Q}) + F_M^2(\vec{Q})
\]

NB we have neglected incoherent scattering here.
Polarized magnetic diffraction

Using a spin flipper to access these two polarized cross sections we can determine the “flipping ratio”, $R$, of a particular Bragg reflection:

\[
R = \frac{d\sigma/d\Omega}{d\sigma/d\Omega} = \frac{[F_N(Q) + F_M(Q)]^2}{[F_N(Q) - F_M(Q)]^2} = \left(\frac{1 + \gamma}{1 - \gamma}\right)^2
\]

with \( \gamma = \frac{F_M(Q)}{F_N(Q)} \)

So, for example, in the case of Ni we measure a flipping ratio of 1.7 at the (111) reflection and 1.1 at the (400) reflection.

After all the reflections have been measured, $F_M(Q)$ can be deduced (assuming careful measurements of $F_N(Q)$ have been taken - at low fields/high temps). Then $F_M(Q)$ can be inverse Fourier transformed to get the real-space magnetisation density.
D3, ILL

Hot neutron 2-axis diffractometer at the ILL

Crystal mounted on low-temperature goniometer to access reflections out of the equatorial plane

Other PND instruments at LLB, Oak Ridge, SINQ, FRM-II...
Form factor measurements

Crystal structure
$z = c/2$

Magnetisation density

Derived from inverse Fourier transform of $F_m(Q)$

Spin density measurements

SrFe$_2$As$_2$

![Graph showing spin density measurements](image)

**Theory with**
- Experiment $z_{As}$
- Optimized $z_{As}$

**Normalized Factor**
- 1.676 $\mu_B$
- 0.879 $\mu_B$

**Form Factor vs $|K|$ (Å$^{-1}$)**

**Normalized Factor**
- 1.040 $\mu_B$
- 1.676 $\mu_B$
- 2.220 $\mu_B$

**Legend**
- Experiment
- Theory
- BCC Fe

*Y Lee, et. al., Phys Rev B 81, 060406R (2010)*
Polarization analysis - paramagnets

It can be shown (see Squires p 179) that in the case of a fully disordered paramagnet these expressions reduce to

\[
\frac{d\sigma}{d\Omega}_{NSF}^\zeta = \frac{1}{3} \left( \frac{\gamma r_0}{2} \right)^2 g^2 f^2 (Q) J(J+1) \left[ 1 - (\hat{P} \cdot \hat{Q})^2 \right]
\]

\[
\frac{d\sigma}{d\Omega}_{SF}^\zeta = \frac{1}{3} \left( \frac{\gamma r_0}{2} \right)^2 g^2 f^2 (Q) J(J+1) \left[ 1 + (\hat{P} \cdot \hat{Q})^2 \right]
\]

where we have replaced the z-direction with the general direction \( \zeta = x, y, \) or \( z \)

Therefore, the scalar polarization becomes is given by

\[
P = \frac{\left[ 1 - (\hat{P} \cdot \hat{Q})^2 \right] - \left[ 1 + (\hat{P} \cdot \hat{Q})^2 \right]}{\left[ 1 - (\hat{P} \cdot \hat{Q})^2 \right] + \left[ 1 + (\hat{P} \cdot \hat{Q})^2 \right]}
\]

This is easily simplified to give the Halpern-Johnson Equation

\[
P' = -\hat{Q} \cdot (\hat{P} \cdot \hat{Q})
\]

first derived in 1939, and valid for all paramagnetic and disordered magnets
Halpern-Johnson Equation

\[ P' = -\hat{Q} \cdot (P \cdot \hat{Q}) \]

where \( P' \) is the scattered polarization direction and \( P \) is the incident polarization direction.

We can immediately see that setting the polarization direction along the scattering vector has the desired effect of rendering all the magnetic scattering in the spin-flip cross-section.

Now we suppose that we have a multi-detector in the x-y plane. In this case the unit scattering vector is

\[ \hat{Q} = \begin{pmatrix} \cos \alpha \\ \sin \alpha \\ 0 \end{pmatrix} \]

where \( \alpha \) is the angle between \( Q \) and an arbitrary x-axis - the “Schärpf angle”.

\[ P' = P \cdot \hat{Q} \cdot (P \cdot \hat{Q}) \]

where \( P' \) is the scattered polarization direction and \( P \) is the incident polarization direction.
The Schärpf Equations

Substituting this unit scattering vector into the Halpern-Johnson Equation, and directing \( \mathbf{P} \) in three orthogonal directions, \( x, y \) and \( z \), leads to six cross sections (3 non-spin flip and 3 spin-flip).

Including the nuclear coherent, isotope incoherent and spin-incoherent terms we have

\[
\begin{align*}
\left( \frac{d\sigma}{d\Omega} \right)_{\text{NSF}}^X &= \frac{1}{2} \sin^2 \alpha \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{1}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}} + \left( \frac{d\sigma}{d\Omega} \right)_{\text{nuc+II}} \quad \text{x-direction} \\
\left( \frac{d\sigma}{d\Omega} \right)_{\text{SF}}^X &= \frac{1}{2} \cos^2 \alpha + 1 \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{2}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}} \\
\left( \frac{d\sigma}{d\Omega} \right)_{\text{NSF}}^Y &= \frac{1}{2} \cos^2 \alpha \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{1}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}} + \left( \frac{d\sigma}{d\Omega} \right)_{\text{nuc+II}} \quad \text{y-direction} \\
\left( \frac{d\sigma}{d\Omega} \right)_{\text{SF}}^Y &= \frac{1}{2} \sin^2 \alpha + 1 \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{2}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}} \\
\left( \frac{d\sigma}{d\Omega} \right)_{\text{NSF}}^Z &= \frac{1}{2} \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{1}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}} + \left( \frac{d\sigma}{d\Omega} \right)_{\text{nuc+II}} \quad \text{z-direction} \\
\left( \frac{d\sigma}{d\Omega} \right)_{\text{SF}}^Z &= \frac{1}{2} \left( \frac{d\sigma}{d\Omega} \right)_{\text{mag}} + \frac{2}{3} \left( \frac{d\sigma}{d\Omega} \right)_{\text{SI}}
\end{align*}
\]

D7, ILL

Cold neutrons (to avoid too much Bragg scattering)

Can be used as a diffuse scattering diffractometer or a cold time-of-flight spectrometer


Other wide angle polarized instruments at FRM-II (DNS) NIST (Macs) - others coming soon
Supermirrors

Supermirror “bender” analyser array on D7, ILL. There is over 250 m² of supermirror in the full analyser array. (c.f. doubles tennis court is 260 m²)
Polarized magnetic diffraction - powders

A difference map between parallel and antiparallel cross-sections leaves the nuclear-magnetic interference term

$$\Delta \frac{d\sigma}{d\Omega} = \frac{d\sigma}{d\Omega \uparrow} - \frac{d\sigma}{d\Omega \downarrow} = 4F_N(Q)F_M(Q)$$

In the case of ferrimagnets, where some Bragg reflections are due to entirely one sublattice, this can lead to positive and negative peaks in the difference pattern.

Fe$_3$S$_4$ - Greigite
(NB can’t warm above T$_c$)

Chang, et. al., J Geophysical Res. 114 B07101 (2009)
Diffuse scattering examples
Spin-Ice

Frustrated ferromagnetic system with strong CF anisotropy

Polymer diffraction

Polyisoprene: \((CH_2CH = C(CH_3)CH_2)_n\)
Alvarez, et al.

- Complete separation of SI scattering
- Internal normalisation (inc. D-W factor)
- Careful analysis of multiple scattering
- Close comparison with MD simulations

Inelastic magnetic scattering

Pyrochlore - Tb$_2$Sn$_2$O$_7$

Science with Polarized Neutrons

Magnetic slow-relaxation in “spin-ice”


Glass transition in polymer-glass, polybutadiene