#### Polarized Neutrons Intro and Techniques Ross Stewart



#### Polarized neutron beams

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

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

 $\vec{P} = \langle \vec{s}_n \rangle / \frac{1}{2} = 2 \langle \vec{s}_n \rangle$ 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 Frisch et al (Phys Rev 53, 719), Powers (Phys Rev 54, 827)

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 <u>μ.B</u> interaction Hughes and Burgy (*Phys Rev*, **81**, 498)



### History of Polarized Neutrons

1951 Polarizing crystals (magnetite Fe<sub>3</sub>O<sub>4</sub>, Co<sub>92</sub>Fe<sub>8</sub>)

Shull et al (Phys Rev 83, 333; 84, 912)





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

Nathans et al (Phys Rev Lett, 2, 254)

1963 General theory of neutron polarization analysis Blume (Phys Rev 130, 1670) Maleyev (Sov. Phys.: Solid State 4, 2533)



1969 First implementation of neutron polarization analysis, Oak Ridge, USA Moon, Riste and Koehler (*Phys Rev* 181, 920)









### History of Polarized Neutrons

1972 Invention of neutron spin echo (INII, 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 <sup>3</sup>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.

$$P = \frac{N_{+} - N_{-}}{N_{+} - N_{-}}$$
$$= \frac{\left(N_{+} / N_{-}\right) - 1}{\left(N_{+} / N_{-}\right) + 1}$$
$$= \frac{F - 1}{F + 1}$$

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











 First attempted by Moon, Riste and Koehler (Oak Ridge 1969) *Phys Rev. 181 (1969) 920*





 First attempted by Moon, Riste and Koehler (Oak Ridge 1969) *Phys Rev. 181 (1969) 920*





#### Polarizers

Stern-Gerlach experiment (1922)



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



#### Cu<sub>2</sub>MnAI (Heusler) crystal grown at ILL



3He spin-filter





#### Uniaxial Polarization Analysis



# Neutron polarization and scattering

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

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

Where the spin-state of the neutron **S** is either spin-up

$$\begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \text{or spin down} \quad \left| \downarrow \right\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

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

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

 $|\uparrow\rangle =$ 

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

$$\langle \uparrow | \downarrow \rangle = \langle \downarrow | \uparrow \rangle = 0, \ \langle \uparrow | \uparrow \rangle = \langle \downarrow | \downarrow \rangle = 1$$



# Neutron polarization and magnetic scattering

V is the magnetic scattering potential given by

$$V_m(\mathbf{Q}) = -\frac{\gamma_n r_0}{2\mu_B} \boldsymbol{\sigma} \cdot \mathbf{M}_{\perp}(\mathbf{Q}) = -\frac{\gamma_n r_0}{2\mu_B} \sum_{\zeta} \boldsymbol{\sigma}_{\zeta} \cdot \boldsymbol{M}_{\perp\zeta}(\mathbf{Q}) \qquad \text{(see e.g. Squires)}$$

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

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

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

$$\left\langle \mathbf{S}' | V_m(\mathbf{Q}) | \mathbf{S} \right\rangle = -\frac{\gamma_n r_0}{2\mu_B} \begin{cases} M_{\perp z}(\mathbf{Q}) & |\uparrow\rangle \rightarrow |\uparrow\rangle \\ -M_{\perp z}(\mathbf{Q}) & |\downarrow\rangle \rightarrow |\downarrow\rangle \\ M_{\perp x}(\mathbf{Q}) - iM_{\perp y}(\mathbf{Q}) & |\uparrow\rangle \rightarrow |\downarrow\rangle \\ M_{\perp x}(\mathbf{Q}) + iM_{\perp y}(\mathbf{Q}) & |\downarrow\rangle \rightarrow |\downarrow\rangle \end{cases}$$
 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  $b_1$  and  $b_+$ .

The scattering length operator is

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

$$A = \frac{(I+1)b_{+} + Ib_{-}}{2I+1}, \quad B = \frac{b_{+} - b_{-}}{2I+1} \quad \text{(see e.g. Squires, p173)}$$

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

$$\langle \mathbf{S}' | \hat{\mathbf{b}} | \mathbf{S} \rangle = \begin{cases} A + BI_z & |\uparrow\rangle \rightarrow |\uparrow\rangle \\ A - BI_z & |\downarrow\rangle \rightarrow |\downarrow\rangle \\ B(I_x - iI_y) & |\uparrow\rangle \rightarrow |\downarrow\rangle \\ B(I_x + iI_y) & |\downarrow\rangle \rightarrow |\uparrow\rangle \end{cases}$$
 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  $\overline{b} = A$  i.e. the coherent scattering is entirely non-spin-flip



#### Moon-Riste-Koehler Equations

Bringing all this together, we get

$$\begin{split} |\uparrow\rangle \rightarrow |\uparrow\rangle &= \overline{b} - \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} + BI_z \\ |\downarrow\rangle \rightarrow |\downarrow\rangle &= \overline{b} + \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} - BI_z \\ & \text{Moon, Riste and Koehler (Phys Rev 181 (1969) 920} \\ |\uparrow\rangle \rightarrow |\downarrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} (M_{\perp x} - iM_{\perp y}) + B(I_x - iI_y) \\ |\downarrow\rangle \rightarrow |\uparrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} (M_{\perp x} + iM_{\perp y}) + B(I_x + iI_y) \end{split}$$

Remember that:  $\vec{M}_{\perp}(\vec{Q}) = \hat{Q} \times \left(\vec{M}(\vec{Q}) \times \hat{Q}\right) = \vec{M}(\vec{Q}) - \left(\vec{M}(\vec{Q}) \cdot \hat{Q}\right) \hat{Q}$ 

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  $\overline{b^2} - (\overline{b})^2$ 

Applying this to the  $|\uparrow\rangle \rightarrow |\uparrow\rangle$  transition, and neglecting magnetic scattering, we get  $\overline{b^2} = \left\langle \left(\overline{b} + BI_z\right)^2 \right\rangle$  $= \left\langle \left(\overline{b}\right)^2 \right\rangle + \left\langle B^2 I_z^2 \right\rangle + 2 \left\langle \overline{b} BI_z \right\rangle$ 

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

$$\langle \mathbf{I} \rangle = \sqrt{I(I+1)} = \sqrt{I_x^2 + I_y^2 + I_z^2}$$
  

$$\Rightarrow I_x^2 = I_y^2 = I_z^2 = \frac{1}{3}I(I+1) \text{ since the distribution is isotropic}$$
  
Therefore we can write  

$$\overline{b^2} - \left(\overline{b}\right)^2 = \left\langle \left(\overline{b}\right)^2 \right\rangle - \left(\overline{b}\right)^2 + \frac{1}{3}B^2I(I+1)$$
  
Isotope incoherent scattering spin incoherent scattering

The other transitions are dealt with in a similar way



#### Moon-Riste-Koehler II

Finally, we get

$$\begin{split} |\uparrow\rangle \rightarrow |\uparrow\rangle &= \overline{b} - \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} + b_{II} + \frac{1}{3} b_{SI} \\ |\downarrow\rangle \rightarrow |\downarrow\rangle &= \overline{b} + \frac{\gamma_n r_0}{2\mu_B} M_{\perp z} + b_{II} + \frac{1}{3} b_{SI} \\ |\uparrow\rangle \rightarrow |\downarrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} (M_{\perp x} - iM_{\perp y}) + \frac{2}{3} b_{SI} \\ |\downarrow\rangle \rightarrow |\uparrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} (M_{\perp x} + iM_{\perp y}) + \frac{2}{3} b_{SI} \end{split}$$
 where  $b_{SI} = \sqrt{B^2 I(I+1)} \\ |\downarrow\rangle \rightarrow |\uparrow\rangle &= -\frac{\gamma_n r_0}{2\mu_B} (M_{\perp x} + iM_{\perp y}) + \frac{2}{3} b_{SI} \end{split}$ 

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

 $\frac{d\sigma}{d\Omega} = \left[F_N(\mathbf{Q}) - F_M(\mathbf{Q})\right]^2 \text{ for neutrons polarized parallel to the field}$  $\frac{d\sigma}{d\Omega} = \left[F_N(\mathbf{Q}) + F_M(\mathbf{Q})\right]^2 \text{ for neutrons polarized antiparallel to the field}$ 

where

$$F_N(\mathbf{Q}) = \sum_i b_i \exp(i\mathbf{Q} \cdot \mathbf{r}_i)$$

$$F_M(\mathbf{Q}) = \gamma_n r_0 \sum_{i} g_{J_i} J_i f_i(\mathbf{Q}) \exp(i\mathbf{Q} \cdot \mathbf{r}_i)$$

Notice that to simulate an unpolarized measurement, we simply average the two polarized cross sections  $\frac{1}{2}$ 

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

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:

$$R = \frac{\left(\frac{d\sigma}{d\Omega}\right)_{\downarrow}}{\left(\frac{d\sigma}{d\Omega}\right)_{\uparrow}} = \frac{\left[F_{N}(\mathbf{Q}) + F_{M}(\mathbf{Q})\right]^{2}}{\left[F_{N}(\mathbf{Q}) - F_{M}(\mathbf{Q})\right]^{2}} = \left(\frac{1+\gamma}{1-\gamma}\right)^{2}$$
  
with  $\gamma = \frac{F_{M}(\mathbf{Q})}{F_{N}(\mathbf{Q})}$ 

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,  $F_M(\mathbf{Q})$  can be deduced (assuming careful measurements of  $F_N(\mathbf{Q})$ have been taken - at low fields/high temps). Then  $F_M(\mathbf{Q})$ 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 lowtemperature goniometer to access reflections out of the equatorial plane

updated AF 2011

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



#### Form factor measurements



P Javorsky, et. al., Phys. Rev. B 67 224429 (2003)



Derived from inverse Fourier transform of  $F_m(\mathbf{Q})$ 





#### Polarization analysis - paramagnets

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

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

where we have replaced the z-direction with the general direction  $\zeta = x$ , y, or z Therefore, the scalar polarization becomes is given by

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

This is easily simplified to give the Halpern-Johnson Equation

$$\mathbf{P}' = -\hat{\mathbf{Q}} \cdot (\mathbf{P} \cdot \hat{\mathbf{Q}})$$

first derived in 1939, and valid for all paramagnetic and disordered magnets



# Halpern-Johnson Equation

 $\mathbf{P}' = -\hat{\mathbf{Q}} \cdot (\mathbf{P} \cdot \hat{\mathbf{Q}})$ 

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



### The Schärpf Equations

Substituting this unit scattering vector into the Halpern-Johnson Equation, and directing 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

$$\left(\frac{d\sigma}{d\Omega}\right)_{X}^{NSF} = \frac{1}{2}\sin^{2}\alpha\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc+II} \qquad \text{x-direction}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{X}^{SF} = \frac{1}{2}\left(\cos^{2}\alpha + 1\right)\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc+II} \qquad \text{y-direction}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{Y}^{NSF} = \frac{1}{2}\cos^{2}\alpha\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc+II} \qquad \text{y-direction}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{Y}^{SF} = \frac{1}{2}\left(\sin^{2}\alpha + 1\right)\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \left(\frac{d\sigma}{d\Omega}\right)_{SI} + \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{1}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{nuc+II} \qquad \text{z-direction}$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{Z}^{SF} = \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{mag} + \frac{2}{3}\left(\frac{d\sigma}{d\Omega}\right)_{SI} + \frac{1}{2}\left(\frac{d\sigma}{d\Omega}\right)_{nuc+II} \qquad \text{z-direction}$$

Schärpf and Capellmann Phys Stat Sol A135 (1993) 359



# 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

JRS, J. Appl. Cryst 42 (2009) 69

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 m<sup>2</sup> of supermirror in the full analyser array. (c.f. doubles tennis court is 260 m<sup>2</sup>)





Science & Technology Facilities Council

JRS, J. Appl. Cryst 42 (2009) 69

#### Polarized magnetic diffraction - powders



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

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

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.

 $Fe_3S_4$  - Greigite (NB can't warm above  $T_c$ )



Science & Technology Facilities Council

Chang, et. al., J Geophysical Res. 114 B07101 (2009)



T. Fennell, et al. Science 326, 415 (2009)

#### 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

Alvarez, et.al., Macromolecules 36 (2003) 238



#### Inelastic magnetic scattering



#### Pyrochlore - Tb<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>

Rule, et. al., Phys. Rev. B 76 212405 (2007)



#### Science with Polarized Neutrons



Ehlers et al, J Phys: Condens Matter 18, R231 (2006)

