

# Polarised Neutron Scattering

Summer School on Neutron Scattering

Chalk River, June 3<sup>rd</sup> – 7<sup>th</sup> 2013



D.H. Ryan

Centre for the Physics of Materials and Physics Department  
McGill University, Montreal, Canada



# Outline

- What is polarised neutron scattering?
- Why use polarised neutron scattering?
- How do you do polarised neutron scattering?
- What can you learn from polarised neutron scattering?

# What is polarised neutron scattering?

It is neutron scattering using a beam of *polarised* neutrons.

Neutrons have a spin.

The interaction between this spin and the atomic moments in a sample makes neutrons the unquestioned first choice in studying magnetic ordering.

We can represent the spin of the neutron by a vector  $\mathbf{s}$ , but we can only ever measure one component of this vector along some arbitrary axis at any instant.

If we pick our axis parallel to  $\mathbf{s}$  then the projection is 1 and the neutron is fully polarised with a polarisation vector  $\mathbf{P}$  and magnitude  $|\mathbf{P}|=1$ .

Unfortunately, the processes that are used both to produce neutrons (typically fission or spallation) and to moderate them, neither create, nor preserve a specific polarisation, so that in a beam of neutrons, the average of  $\mathbf{P}$  over  $N$  neutrons:

$$\mathbf{P} = (\sum_i \mathbf{P}_i) / N$$

is an average over random, unit-length vectors, and is therefore zero.

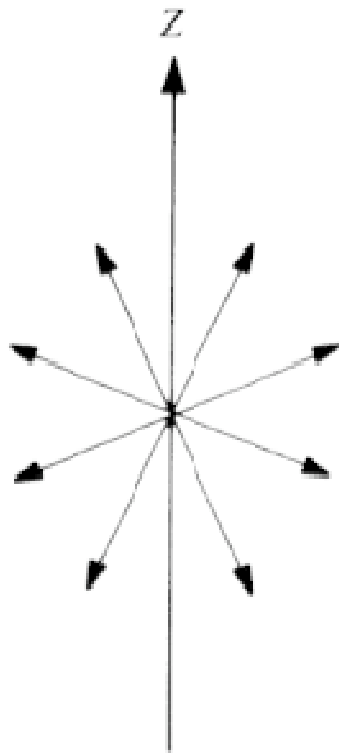
Since the neutron is a spin- $1/2$  particle, it can only have two spin angular momentum values ( $\pm 1/2 h$ ), so once we define our axis (*e.g.* with an applied magnetic field), we can divide our neutrons into two sets:

$N_+$  and  $N_-$  or 'up' and 'down'.

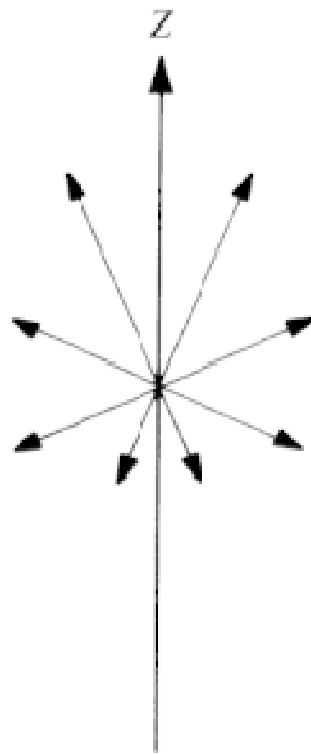
Our polarisation then becomes:

$$\mathbf{P} = [(N_+ - N_-)/(N_+ + N_-)] \mathbf{h}$$

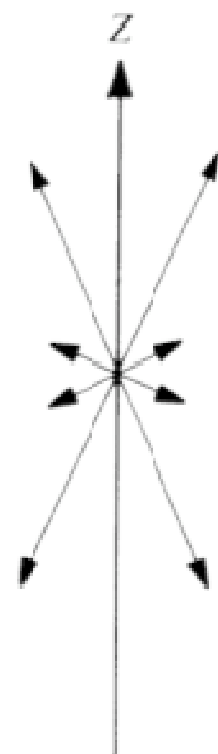
with  $\mathbf{h}$  being a unit vector parallel to the magnetic field.



Random



Polarised



Oriented

# Why use polarised neutron scattering?

The simplest view of neutron scattering has the neutron and nucleus interacting via the strong force: (effectively) point particle on point particle.

However, if there is a moment on the atom involved in the neutron scattering event, then an electromagnetic interaction becomes possible. Now we are dealing with *vector on vector* scattering and the relative orientations of the atomic moment and the scattering vector affect the scattering probability, while the orientation of the neutron and target moments affects the nature of the scattering.

→ *more* information is available.

For example, if the target moment is perpendicular to the neutron polarisation, then the scattering leads to a reversal of the polarisation.

In a conventional neutron scattering experiment, you measure the number of neutrons scattered into some solid angle  $d\Omega$  with energies  $E'$  to  $E' + dE'$ , normalised to the incident flux. This is denoted:

$$\frac{d^2\sigma}{d\Omega dE'}$$

but with a polarised beam we also care about the *orientation* of the neutron's polarisation vector.

Using a polarised neutron beam, we can resolve this scattering into two distinct components depending on whether the incident neutron moment is 'up' (+) or 'down' (-):

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_+ \quad \left(\frac{d^2\sigma}{d\Omega dE'}\right)_-$$

Furthermore, with polarisation analysis on the *scattered* beam, we can resolve these scattering cross-sections into two further groups depending on whether the neutron polarisation is changed in the scattering event:

Non-spin-flip (**NSF**):

$$\left( \frac{d^2\sigma}{d\Omega dE'} \right)_{++} \quad \left( \frac{d^2\sigma}{d\Omega dE'} \right)_{--}$$

and spin-flip (**SF**):

$$\left( \frac{d^2\sigma}{d\Omega dE'} \right)_{+-} \quad \left( \frac{d^2\sigma}{d\Omega dE'} \right)_{-+}$$

With:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{2} [()_{++} + ()_{--} + ()_{+-} + ()_{-+}]$$

Coherent nuclear scattering arises from strong force interactions and is therefore *always* NSF. Depending on the scattering geometry, nuclear and atomic spin scattering can be SF, NSF or a mixture of both.



The nuclear (strong force) interaction is isotropic (no scattering vector dependence) and very short ranged (point on point).

Including the interaction between the neutron spin and that of the scattering nucleus leads to a scattering length (no form factor):

$$b = A + B(\hat{s} \cdot \mathbf{I})$$

where  $s$  is the neutron spin and  $\mathbf{I}$  is the nuclear spin.

Since the neutron is a spin- $1/2$  particle, only two possibilities exist:  $b^+$  and  $b^-$  for parallel and antiparallel configurations respectively.

The first term ( $A$ ) does not depend on either the nuclear or the neutron moments and so gives rise only to NSF scattering.

The term proportional to  $B$  causes NSF scattering through the  $I_z$  term in the dot product, and SF scattering through the  $I_x$  and  $I_y$  terms.  
(Neutron spin 'up' =  $+z$ )

By contrast, the EM interaction between the electron and neutron spins is longer-ranged and involves an extended object (the electron cloud) so the scattering length has a form factor that falls off quite rapidly with  $|k|$ . Furthermore, the interaction is *anisotropic*.

Once we allow for an interaction between the neutron spin and the spins in the sample, we have *three* vectors to deal with:

1. The neutron polarisation  $\mathbf{P}$
2. The scattering vector  $k$
3. The atomic moment  $S$

and our magnetic form factor becomes:

$$b_m = (\text{constants})(\hat{s} \cdot \vec{S}_p)$$

where  $s$  is the neutron spin (parallel to  $\mathbf{P}$ ) and  $S_p$  is the component of  $S$  that is perpendicular to the scattering vector  $k$ .

We can get NSF scattering through the  $s_z$  term of the dot product and SF scattering through the  $s_x$  and  $s_y$  terms. Placing  $S \parallel k$  switches the magnetic scattering 'off' as  $S_p$  is then zero.

Assuming that the neutron polarisation vector  $\mathbf{P}$  is directed along the 'z'-axis, then the balance between SF and NSF scattering for all possible processes is:

Type of scattering	$\mathbf{P} \parallel \vec{k}$		$\mathbf{P} \perp \vec{k}$	
	SF	NSF	SF	NSF
Nuclear coherent	0	1	0	1
Nuclear isotope incoherent	0	1	0	1
Nuclear spin incoherent	2/3	1/3	2/3	1/3
Magnetic	$S_x + S_y$	0	$S_x$	$S_z$

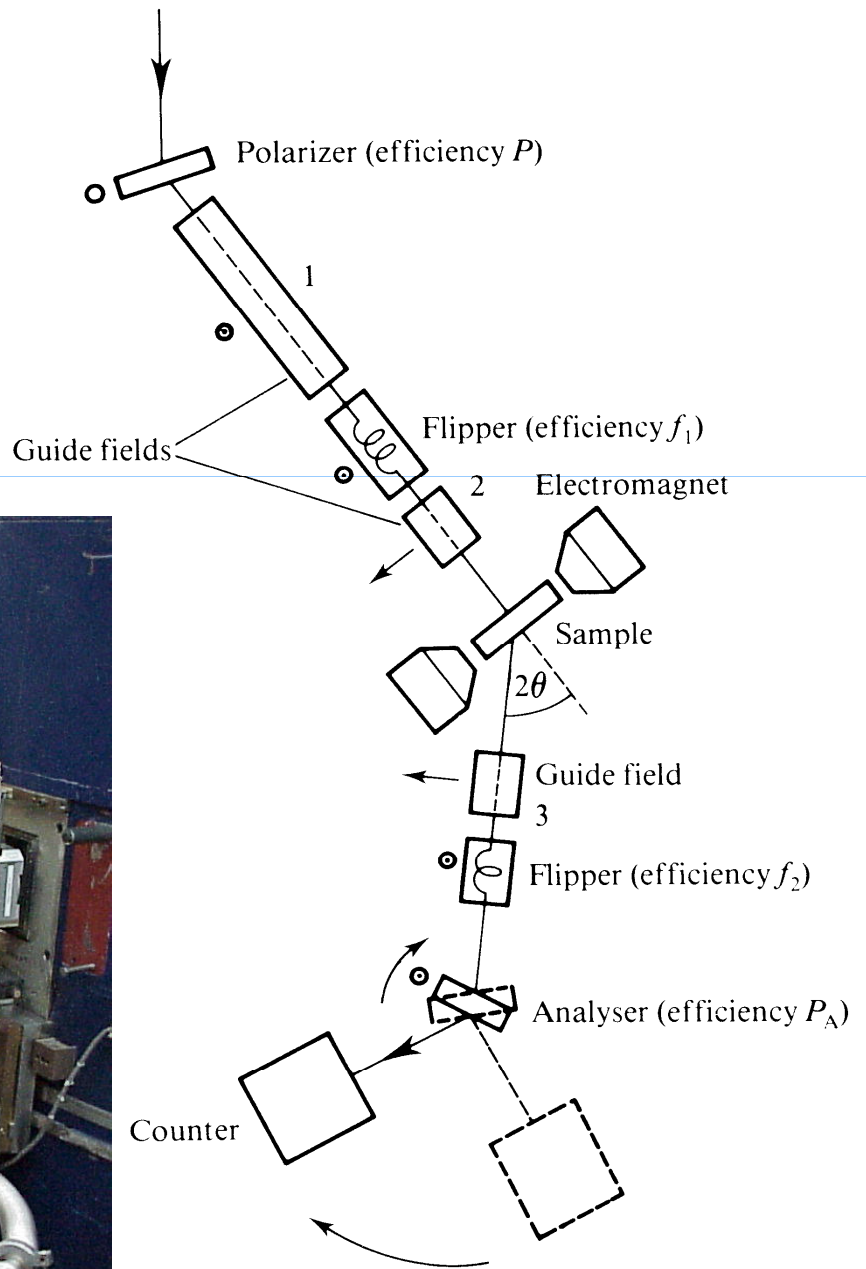
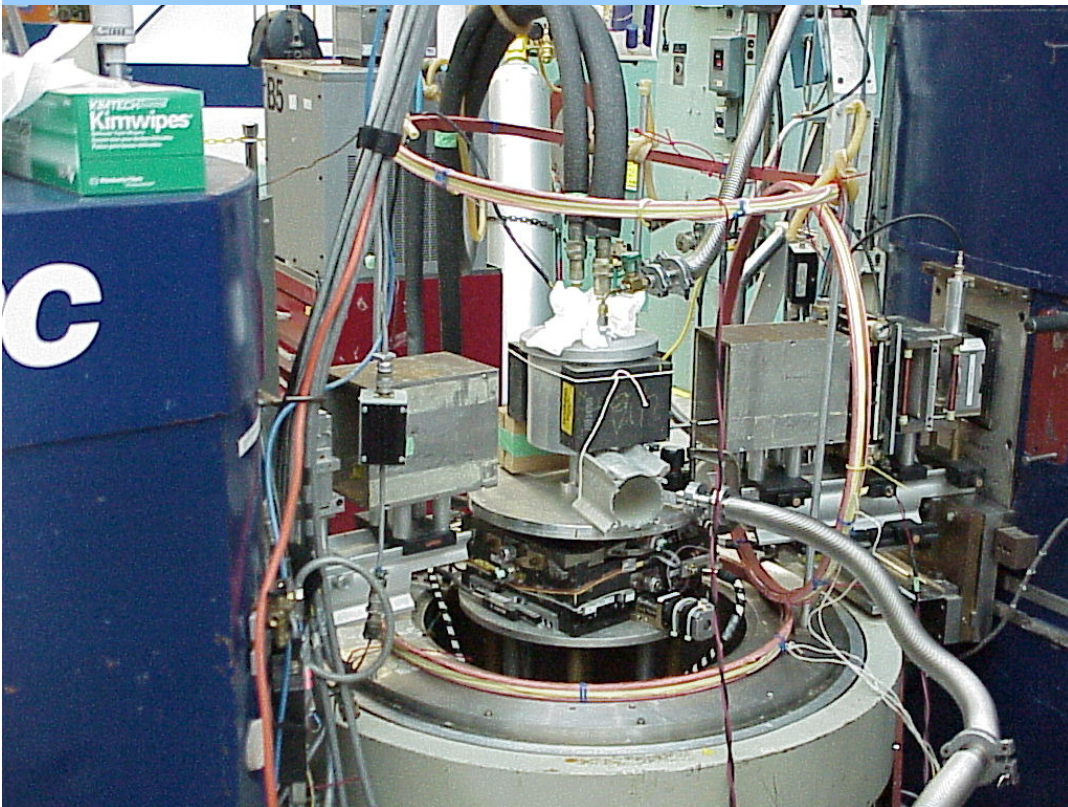
Only the atomic spin components perpendicular to  $k$  contribute to the magnetic scattering, and only spin components perpendicular to  $\mathbf{P}$  can cause a spin flip.

# How do you do polarised neutron scattering?

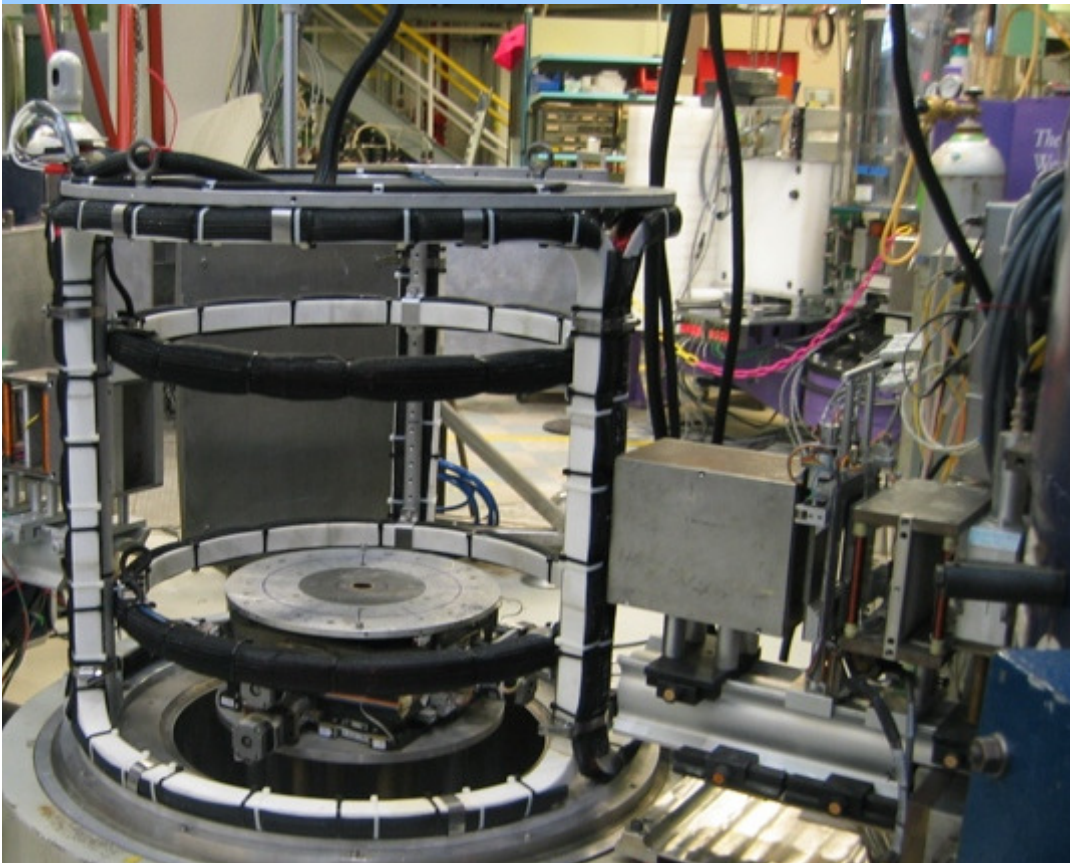
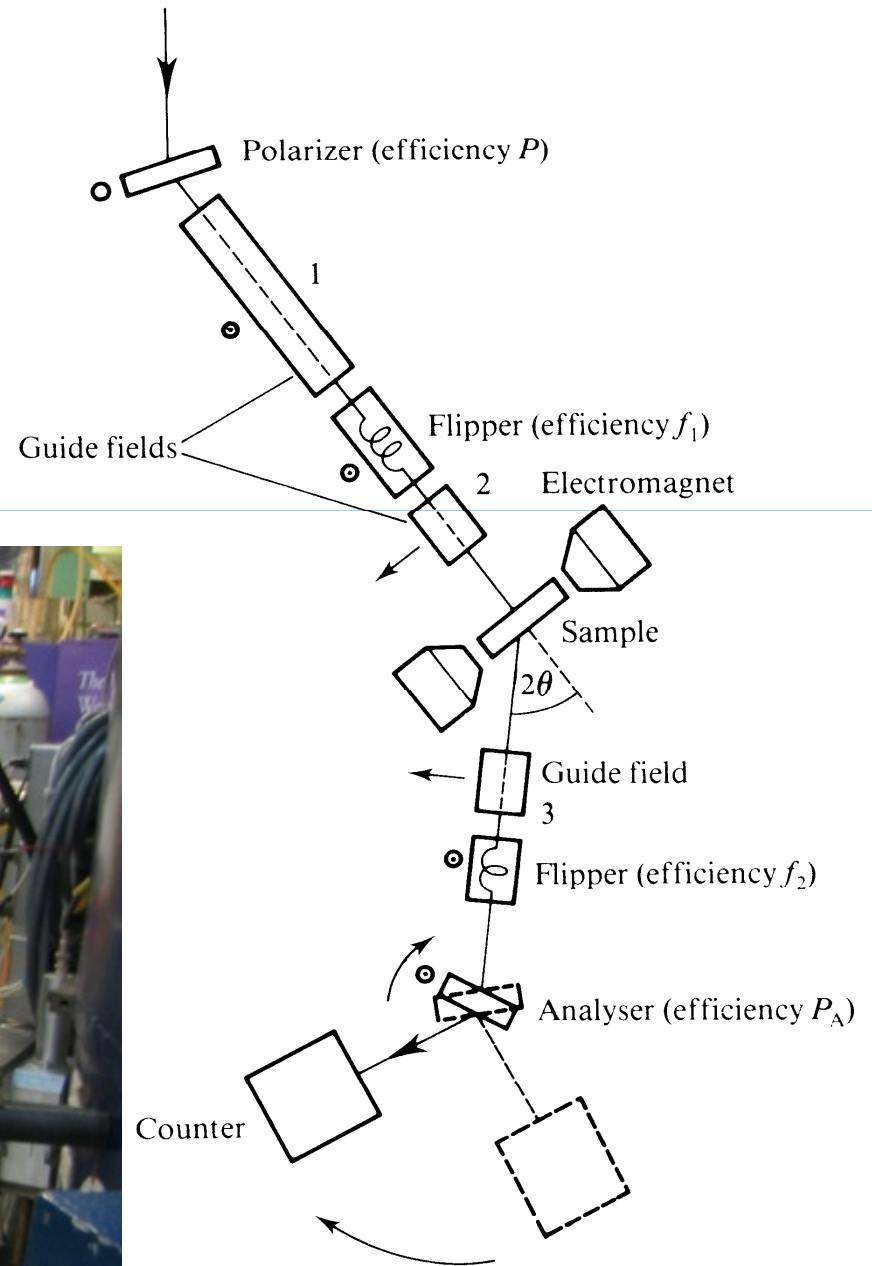
This breaks down into several stages, with an obvious symmetry :

- Produce a polarised neutron beam
- Preserve the polarisation of the beam
- Manipulate the polarisation of the beam
- Deliver it to the sample
- Manipulate the polarisation of the scattered beam
- Preserve the polarisation of the scattered beam
- Measure the final polarisation of the scattered beam

# Spectrometer components



# Spectrometer components (C5)



# Produce a polarised neutron beam

The reactor is a white, unpolarised source, and we will typically want a monochromatic, fully polarised beam. We will be using the processes that make polarised neutron scattering interesting to obtain a polarised beam.

As there is no (practical) way to change the polarisation vector of each neutron as it arrives, we will have to settle for dumping all of the 'wrong' ones.

Several basic methods exist:

- Scattering filters
- Absorption filters
- Bragg reflection
- 'Optical' reflection

# Polarising Filters

Filters are (ideally) broad-band devices with no energy or wavelength selectivity. A separate monochromator must therefore be used. This can be an advantage as a high-quality monochromator crystal can be used for tight energy resolution or looser resolution can be obtained with a velocity chopper.

Scattering filters rely on selectively scattering one polarisation state out of the beam and allowing the other state through.

The earliest polarised neutron beams were produced by transmission through a magnetised polycrystalline iron sheet. Unmagnetised polycrystalline iron sheets make excellent beam depolarisers (but for very different reasons) and are frequently used to assess the effectiveness of other parts of the polarised neutron instrument.



Two parameters determine the efficiency of a polarising filter:

1. Polarising efficiency *i.e.* the polarisation of the transmitted beam, given by:

$$P = (T_+ - T_-) / (T_+ + T_-)$$

2. Total transmittance

$$T = (T_+ + T_-) / 2$$

Using a thicker sheet will increase the polarisation by improving the rejection of the unwanted neutrons, but it also leads to more scattering of the desired neutrons, reducing the overall transmission.

Aim to maximise  $P\sqrt{T}$ .

The scattering cross-sections for the (+) and (-) spin states are given by:

$$\sigma_{\pm} = \sigma_0 \pm \sigma_p$$

where  $\sigma_0$  is the spin-independent cross-section and  $\sigma_p = 1/2(\sigma_+ - \sigma_-)$  is the polarisation cross-section.

The transmitted intensity of each spin state is just:

$$I_{\pm} = I_0 \exp(-Nt \sigma_{\pm})$$

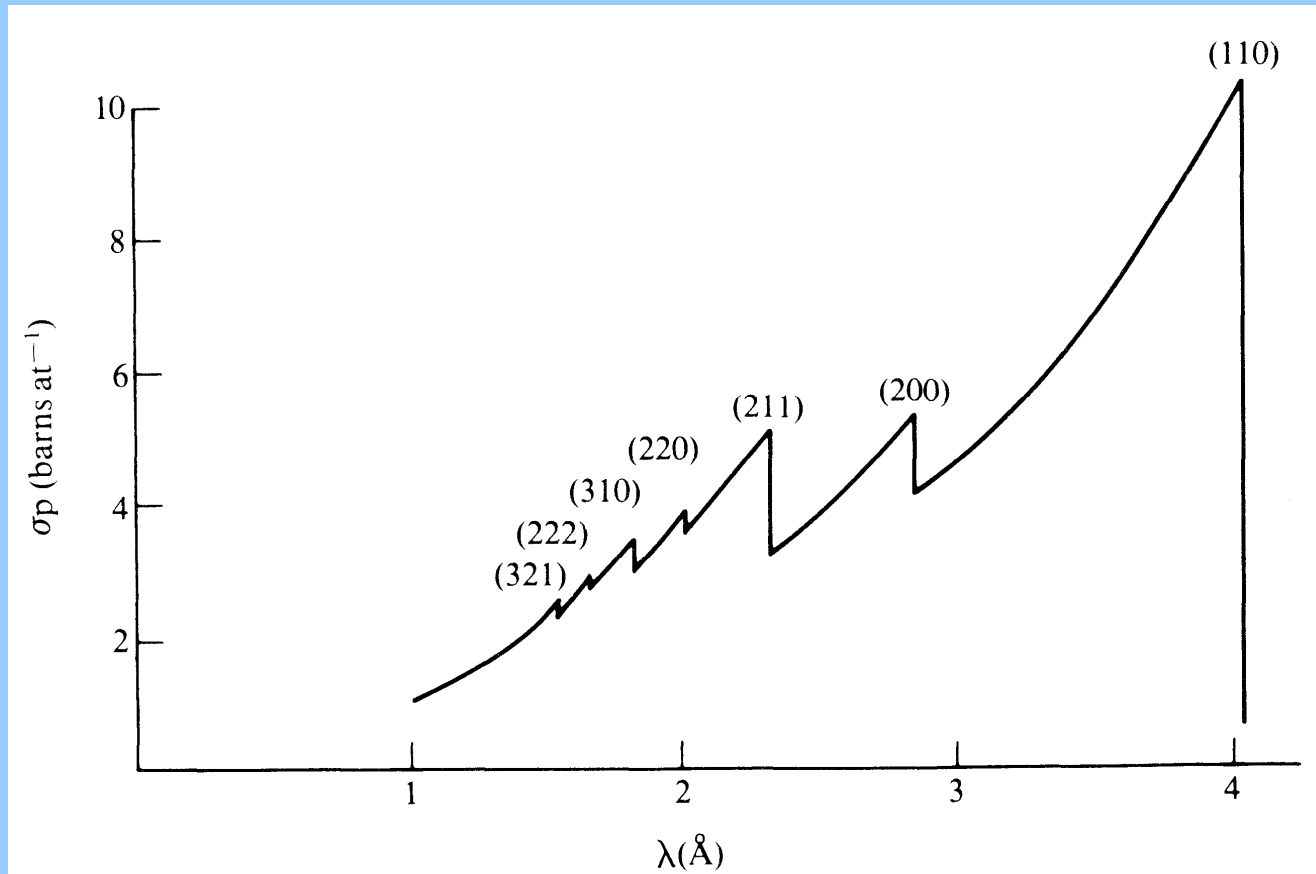
so that

$$P = -\tanh(\sigma_p Nt)$$

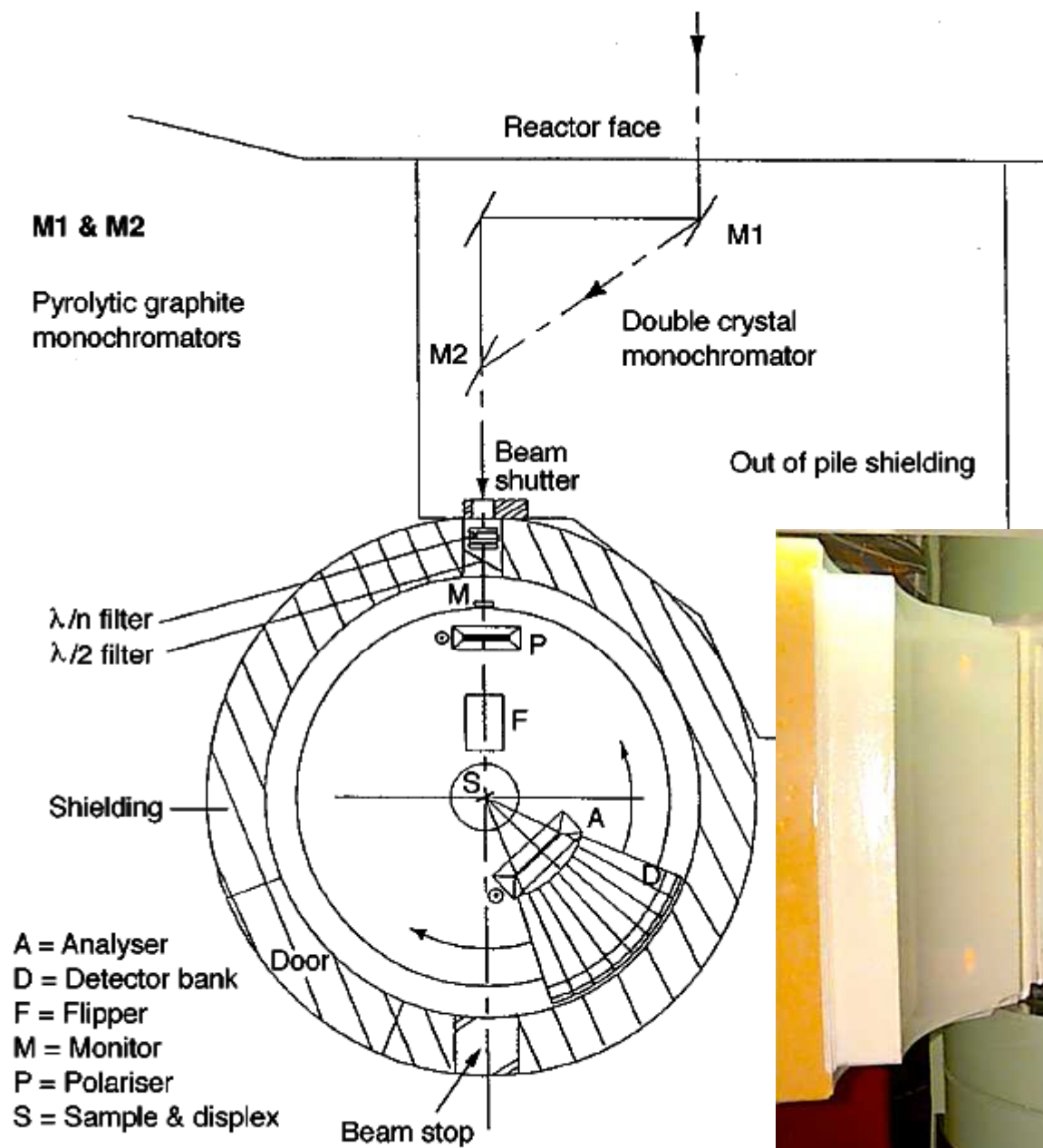
and

$$T = \exp(-\sigma_0 Nt) \cosh(\sigma_p Nt)$$

For iron,  $\sigma_p$  is about 10b near  $\lambda = 4 \text{ \AA}$ , with  $\sigma_0$  about 16.8b so it should be possible to achieve  $P \sim 0.8$  with  $T \sim 0.3$ , however it is difficult to fully magnetise a sheet of iron and in practice this performance is not achieved.



LONGPOL, where this technique was used to great effect, operated with  $P \sim 0.50$  and  $T \sim 0.25$  at  $\lambda = 3.6 \text{ \AA}$ .



Schematic diagram of the LONGPOL diffractometer-spectrometer.

*T. J. Hicks*

Department of Physics, Monash University

*Aust. J. Phys.*, 1997, 50, 1127-39

# Absorption filters

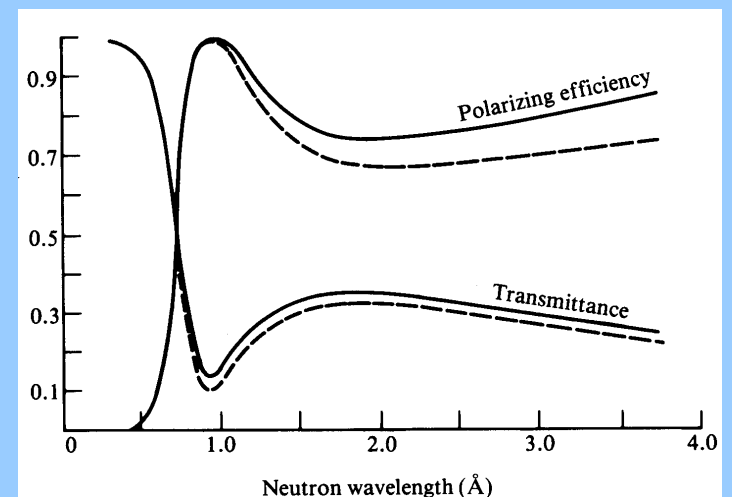
These use polarised nuclei to selectively absorb one neutron polarisation and transmit the other. The two absorption cross-sections can be written in the same form as we used for the scattering filter:

$$\sigma_{A\pm} = \sigma_A(1 \pm \rho P_N)$$

where  $\rho$  depends on the nuclear absorption resonance being used, and  $P_N$  is the *nuclear* polarisation.

Achieving a high degree of nuclear polarisation is the limiting factor in these filters. Traditionally, absorption filter development has employed a “brute force and ignorance” approach:

Cool to a few mK and apply a *big* magnetic field.



$^{149}\text{SmCo}_5$  at 15 mK

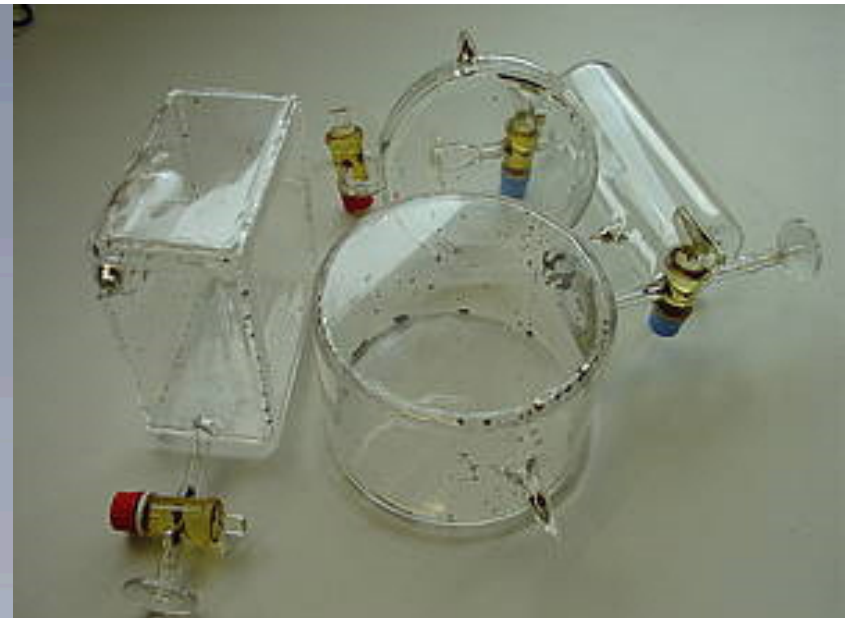
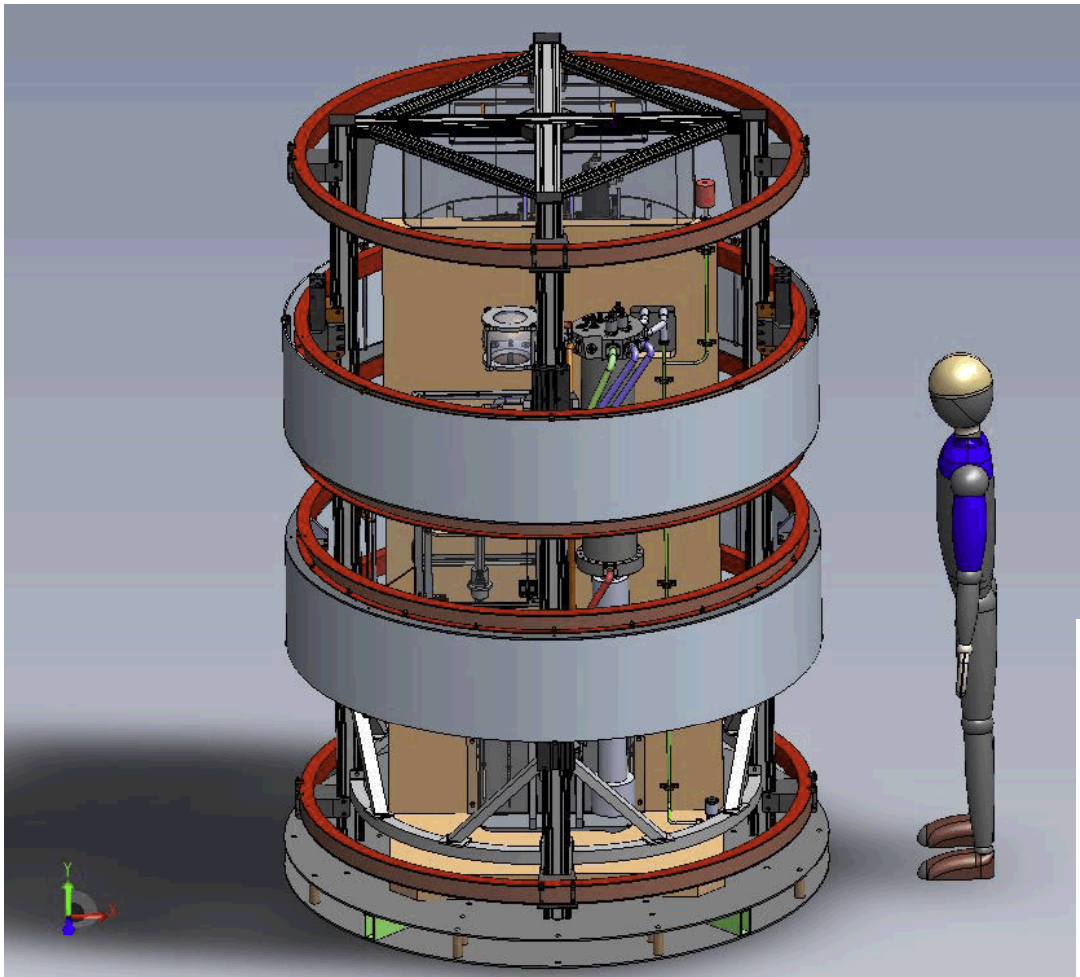
# Optically pumped $^3\text{He}$ filters

These are rapidly growing in popularity as the technology improves. Broad band, largely wavelength independent, they do not cause any beam deviation and they are insensitive to beam divergence.

$^3\text{He}$  has a large (5000 b) capture cross-section for thermal neutrons that involves the creation of a spin-0  $^4\text{He}$  state from a pair of spin- $1/2$  particles ( $^3\text{He}$  and the neutron). The reaction only occurs if the two spins are anti-parallel.

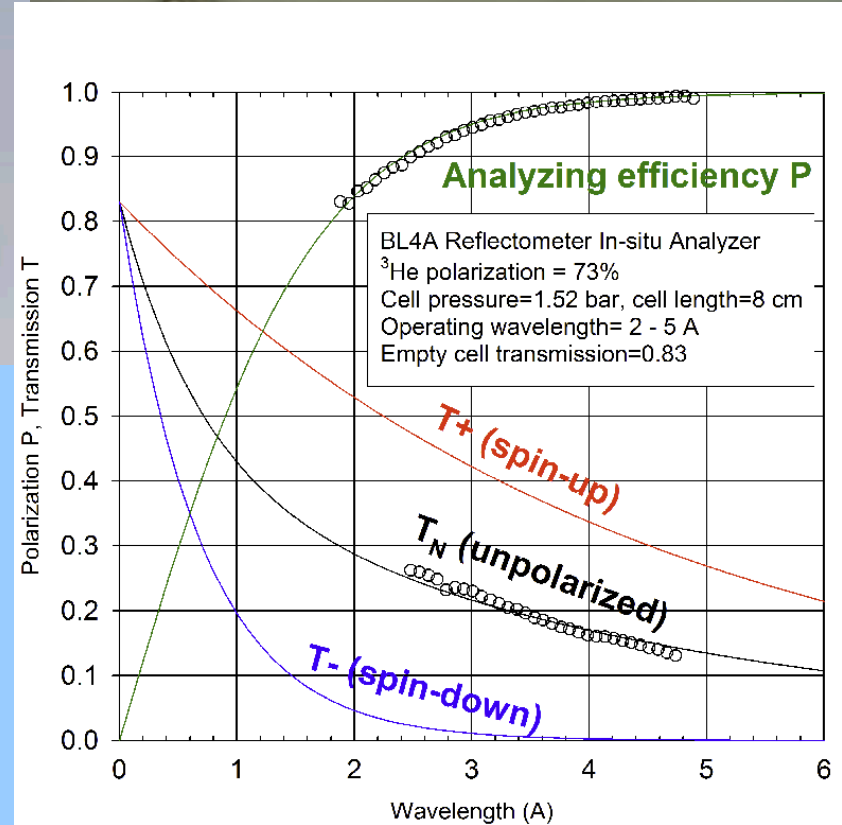
It is possible to polarise  $^3\text{He}$  gas indirectly by spin exchange with optically pumped  $^{85}\text{Rb}$  vapour. Circularly polarised laser light is used to pump the  $^{85}\text{Rb}$  atoms into a specific hyperfine state. These polarised atoms then collide with the  $^3\text{He}$  gas atoms, transferring their polarisation.

The effort goes into preserving the  $^3\text{He}$  polarisation as any scattering involving a magnetic moment can flip the  $^3\text{He}$  spin. Hold-times of 100 hours are now commercially available.



The  $^3\text{He}$  is polarised in a central station, then it is loaded into special cells and moved to the instrument.

As with polarised neutrons, the polarised  $^3\text{He}$  must be kept in an field to preserve the polarisation.



# Bragg reflection

Polarising elements that rely on Bragg diffraction also act as monochromators. This can restrict the range of wavelengths that can be accessed and usually enforces a relatively narrow band-pass at any given wavelength.

Bragg polarisers rely on two key aspects of magnetic neutron scattering:

1. Nuclear and magnetic scattering lengths are of comparable magnitude
2. The magnetic scattering length depends on the relative orientation of the atomic moments and the neutron spin.

If we take a very simple geometry:

- Ferromagnet saturated along 'z'
- Scattering in 'xy'-plane

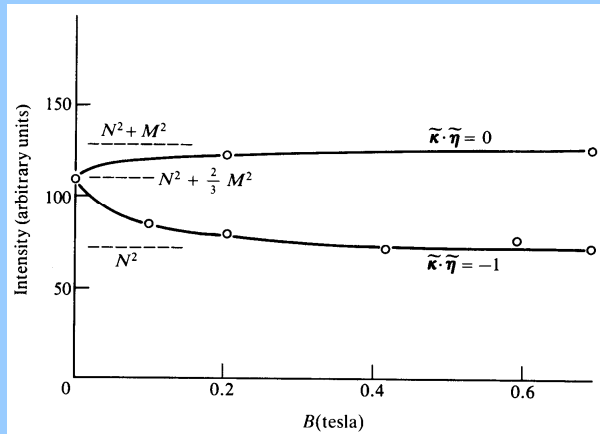
then the scattering cross-section takes a very simple form:

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

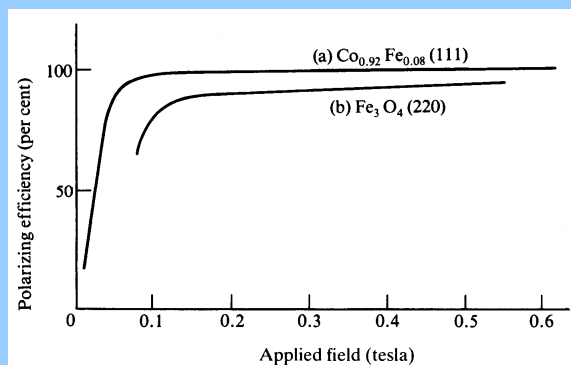
where  $F_N(\kappa)$  and  $F_M(\kappa)$  are the nuclear and magnetic structure factors associated with the specific Bragg reflection (we include all of the form factors etc. here).



If we can locate a reflection for which  $|F_N(\kappa)| = |F_M(\kappa)|$  then the magnetic and nuclear scattering will cancel for one polarisation state, and add for the other. An unpolarised beam undergoing Bragg diffraction from those planes will emerge fully polarised.

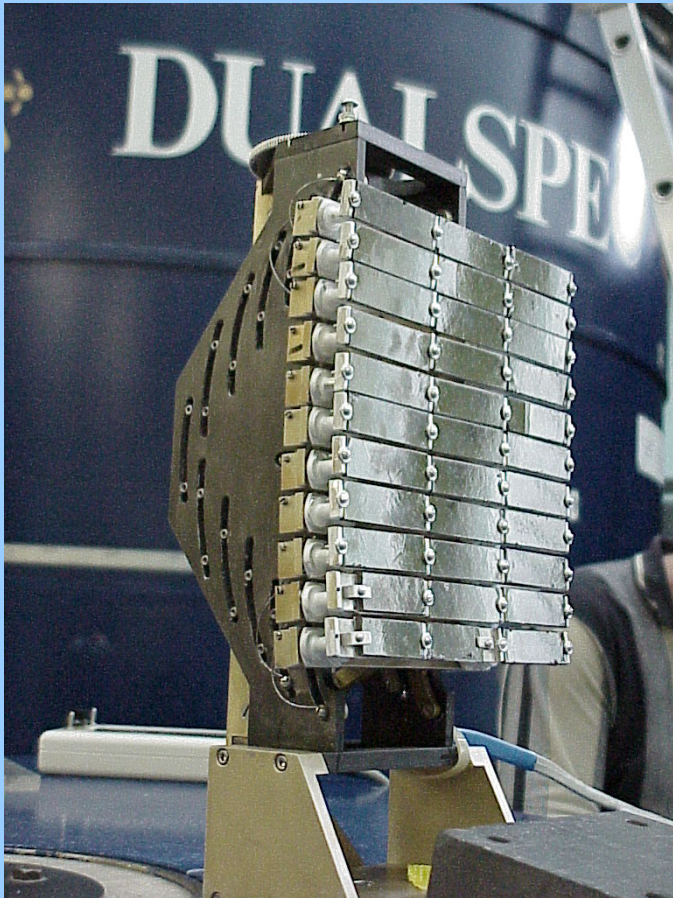


The first such polariser was the (220) reflection from magnetite ( $\text{Fe}_3\text{O}_4$ ), but  $\text{Co}_{0.92}\text{Fe}_{0.08}$  was soon found to yield better polarisation from two reflections: (111) (~98%) and (200) (~100%).



The high absorption cross-section of Co means that you lose about a factor of 4 in intensity compared with an unpolarised monochromator, and when used in a triple-axis system this translates into an overall loss factor of ~15.

The current favourite is the (111) reflection from the Heusler alloy  $\text{Cu}_2\text{MnAl}$ , which gives much better transmission and reflectivity. Unfortunately the (222) structure factor is quite large, leading to some  $\lambda/2$  contamination that must be eliminated using a PG filter. With  $\text{Cu}_2\text{MnAl}$  at both the polariser and analyser positions on C5, the observed polarisation is typically 96% at 2.37 Å.



# 'Optical' polarisers

These are broad-band non-monochromating polarising elements that rely on a very different view of neutron scattering.

If we consider the passage of a neutron through some medium then we are only concerned with forward scattering (*i.e.*  $\kappa = 0$ ) which does not depend on either the structure or the dynamics of the material. In this situation, we can define a refractive index,  $n$ , for the medium.

The refractive index for the two polarisation states is:

$$n_{\pm} = 1 - ( \lambda^2 N / (2\pi) ) ( b \pm (B/B_s)p )$$

where  $N$  is the number density of scattering nuclei,  $b$  is the average nuclear coherent scattering length,  $B$  and  $B_s$  are the average and saturation magnetic polarisations of the material and  $p$  ( $= 2\pi m\mu_n B_s / h^2 N$ ) is the magnetic coherent scattering length at  $\kappa = 0$ .

$n$  is always close to 1, with  $|n - 1| \sim 10^{-6}$

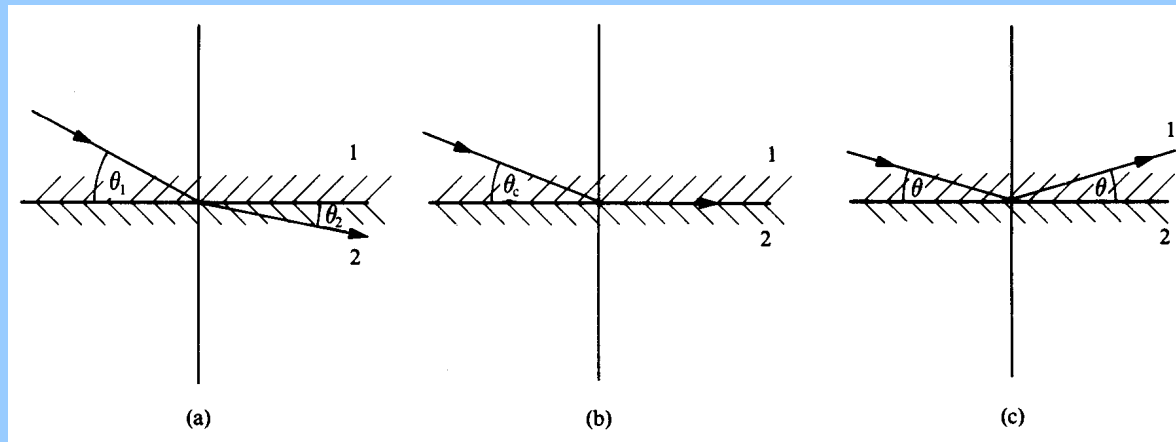
For most materials,  $b > 0$  and  $n < 1$  so a neutron entering the material from vacuum is passing into a lower 'density' region and can undergo total external reflection at sufficiently small incident angles.

Snell's law gives the critical angle for total external reflection as:

$$n = \cos \theta_c \sim 1 - (\theta_c^2 / 2)$$

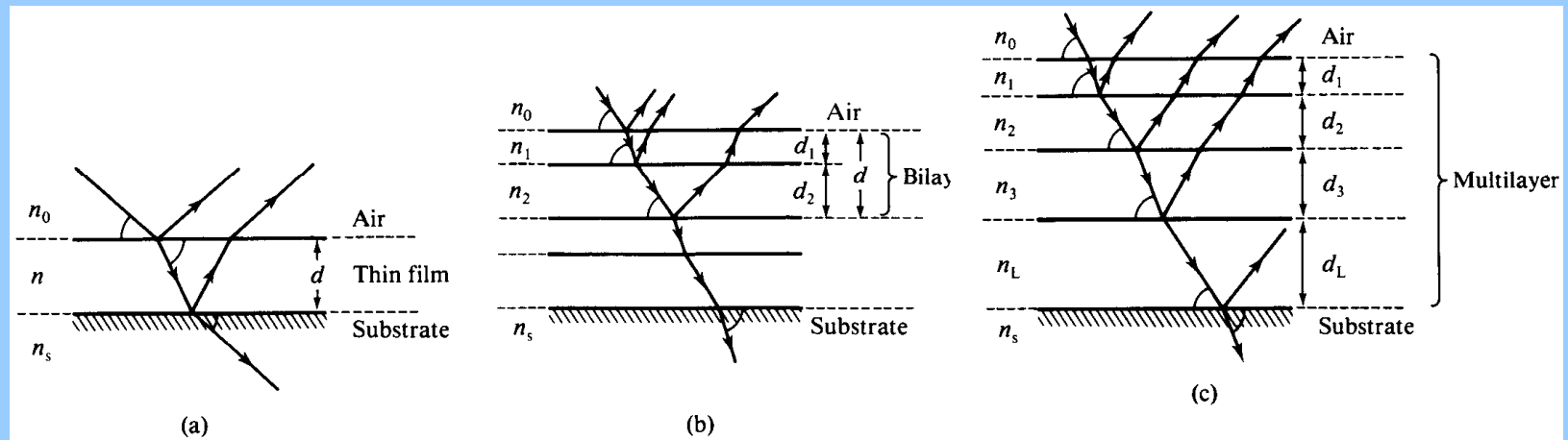
for small angles. Thus:

$$\theta_c = \lambda(N/\pi)^{1/2} [(b \pm (B/B_s)p)]^{1/2}$$



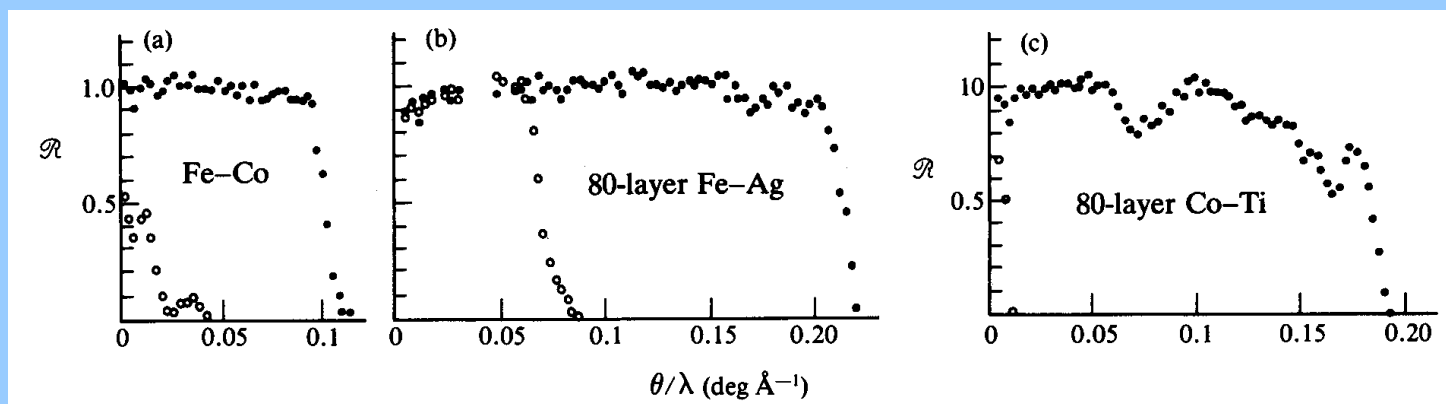
One immediate problem is that  $n$  is very close to 1 and so  $\theta_c$  tends to be of order  $1^\circ$ , and getting a reasonably wide beam demands a very long mirror. A second problem involves 'disposing' of the unwanted polarisation state.

If a thin-film geometry is used, then the unwanted neutrons can be either absorbed in an index-matched underlayer (e.g. a-Gd<sub>15</sub>Ti<sub>85</sub> under a Co<sub>60</sub>Fe<sub>40</sub> polarising mirror) or simply scattered incoherently by a hydrogen-containing polymer substrate.

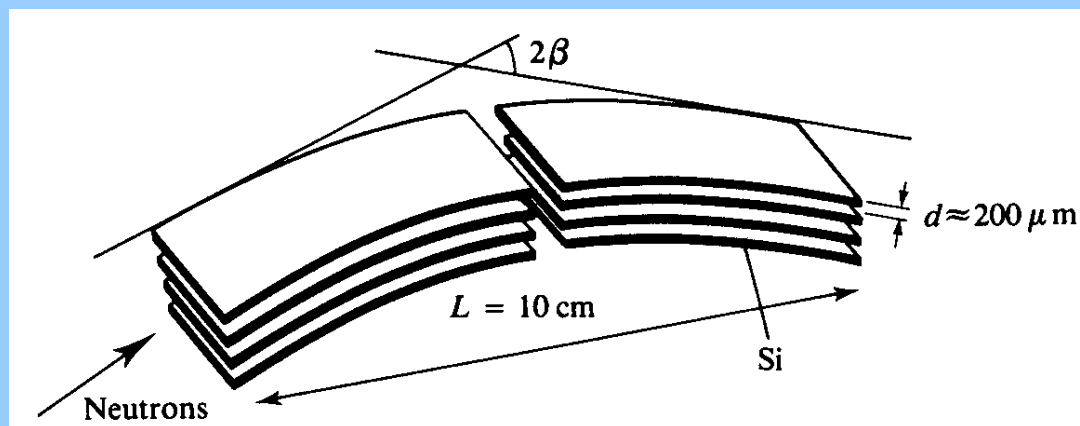


Once you start with thin-films the possibilities for tailoring the reflectivity are greatly increased. A stack of Fe/Ge bilayers yields a monochromatic polarised beam as 'ups' experience a square-wave modulated index and so 'Bragg scatter' off the periodic structure, while the 'downs' see no contrast and simply pass through.

The most extreme form of this arrangement is the 'supermirror' which uses alternating Co and Ti layers but with a gradient in the layer thicknesses so as to extend the critical angle for total reflection for one polarisation state.



Often an array of supermirror sheets will be stacked to form a polarising Soller collimator with a slight curvature used both to eliminate straight-through beam contamination such as  $\gamma$ 's, and to force every neutron to be reflected at least once.



# Preserve the polarisation of the beam

This might seem obvious, but once the beam is polarised, you need it to *stay* polarised.

The problem is that you have created a beam of neutrons with all of the spins in a specific state ('up') with respect to some reference, usually an external magnetic field. They *need* this field to be present in order to know which way is up. Guide fields of about 1 mT are therefore provided all of the way along the neutron flight path from polariser to analyser.

The polarisation can be rotated intentionally by allowing the neutrons to pass through a region in which the field magnitude is constant, but the direction changes 'slowly'. The scale is set by the Larmor frequency of the neutron

$$f_L \sim 29 \text{ MHz/T} \quad \omega_L \sim 183 \times 10^6 \text{ rad/s/T}$$

and the time-of-flight through the region. The precession angle in a field  $B_0$  is given by:

$$\varphi_L = \gamma l B_0 / v$$

where  $l$  is the distance and  $v$  ( $\sim 2000$  m/s) is the neutron velocity. For  $B_0 = 1$  mT, the neutron completes a full rotation in about 7 cm.

If the field direction changes slowly in the neutron's frame of reference, then the neutron follows the change adiabatically. If the change is fast, then the neutron moment remains spatially fixed in what is known as 'non-adiabatic fast passage'.

Both types of field changes are used in polarised neutron instruments to manipulate the polarisation of the beam.



# Manipulate the polarisation of the beam

We can now generate a beam of 'up' neutrons and deliver it to the sample. The other half of the instrument collects the scattered beam, preserves its polarisation and delivers it to an analyser and detector that count the number of 'up' neutrons that arrive.

This gets us  $(d\sigma/d\Omega)_{++}$ , but to get at the other three cross-sections we need a way to turn 'up' neutrons into 'down' neutrons.

This is the job of *flippers*.

Many designs exist, but I will discuss two:

- RF spin flippers
- Precession (Mezei) flippers

# Radiofrequency (RF) spin flippers

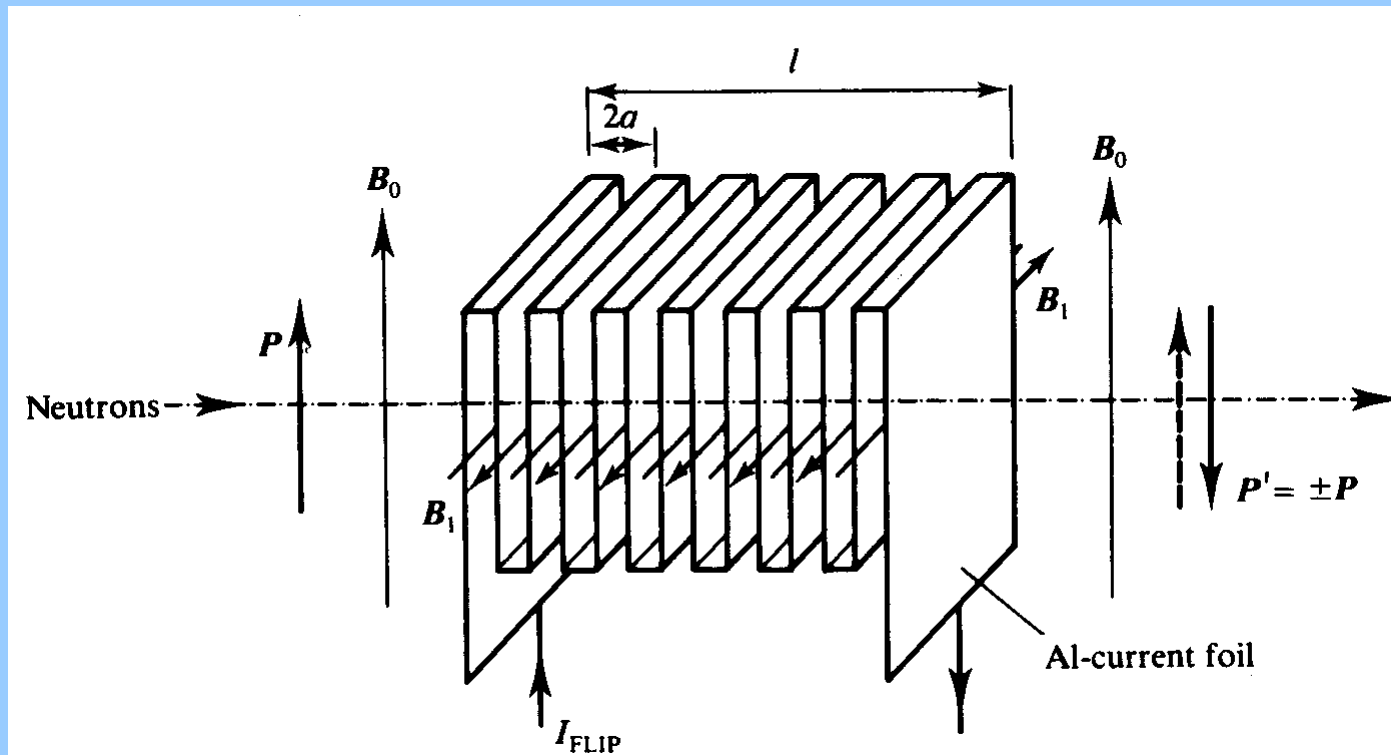
These drive the spin reversal using a brief  $180^\circ$  or  $\pi$  RF field pulse applied perpendicular to a dc guide field.

The duration of the 'pulse' is set by the flight of the neutron through the device, and the frequency is tuned to the Larmor frequency of the neutron in the guide field.

At 10 mT,  $f_L=300$  kHz.

These devices require a homogeneous dc field (better than 0.5%) and have a strong wavelength dependence to their efficiency. The neutron energy is not *strictly* conserved.

An interesting alternative design uses a field that oscillates in *space* rather than time. The neutrons pass through a sheet bent into the form of a sinusoid. A current in the sheet generates a field  $B_1$  oriented perpendicular to the guide field. As with the RF flipper, the dc guide field  $B_0$  sets  $\omega_L$  and the  $B_1$  field appears in the neutron's rest frame with a frequency  $\omega = \pi v/a$ .

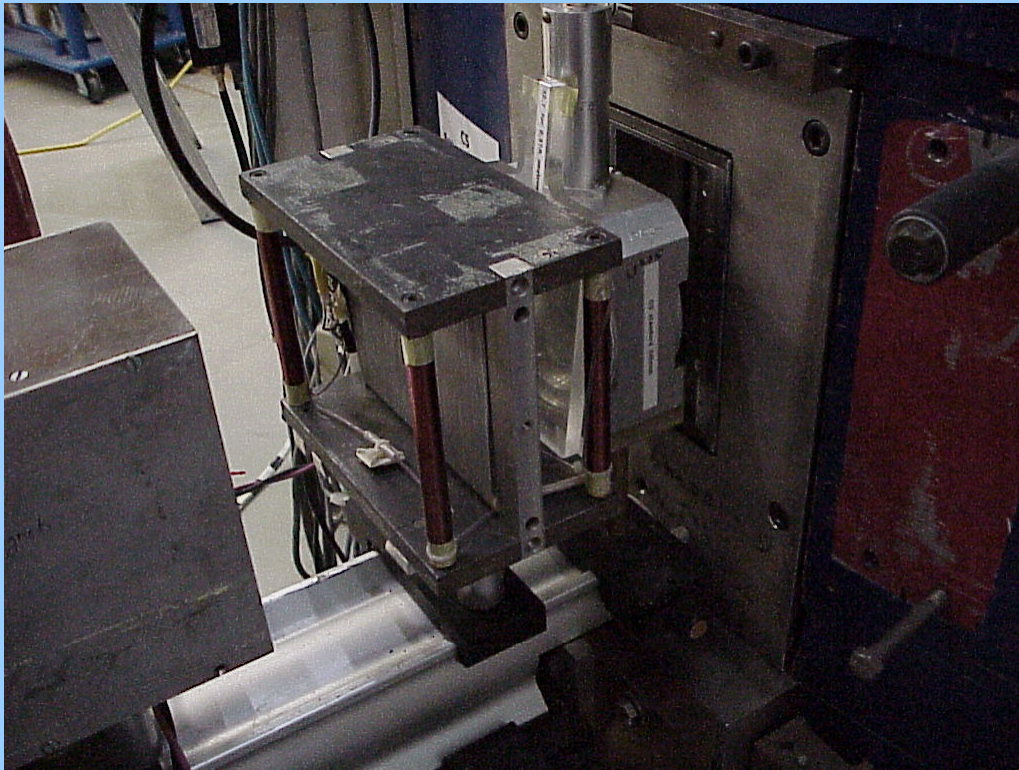


# Precession (Mezei) flippers

A rectangular coil is used to generate an abrupt change in field direction that does not affect the guide field outside the flipper coil. The neutron enters and exits the coil non-adiabatically *i.e.* its polarisation direction does not follow the new field axis.

The neutron spin precesses around the field inside the coil, then it exits with a modified direction. Various combinations of coils, currents and orientation can be used to cause neutron spin rotations of any angle with high efficiency.

The configuration on C5 involves a pair of coils, one inside the other, with the outer one generating a vertical field and the inner one being horizontal and perpendicular to the neutron's flight path. The vertical field is tuned to exactly cancel the guide field at the flipper location, while the horizontal field is set so that the neutron rotates by exactly  $180^\circ$  as it passes through the coil.



The introduction of flippers leads to a characterisation of the beam polarisation in terms of the 'flipping ratio' ( $\mathcal{R}$ ) the ratio of counts observed with the flipper off to that with the flipper on.

$$\mathcal{R} = \frac{f_{off}}{f_{on}}$$

For a perfectly polarised beam this ratio is infinite, on C5 we get about 22.  $\mathcal{R}$  is related to the polarisation by:

$$P = \frac{(N_+ - N_-)}{(N_+ + N_-)} = \frac{\mathcal{R} - 1}{\mathcal{R} + 1}$$

# What can you learn from polarised neutron scattering?

## Some examples

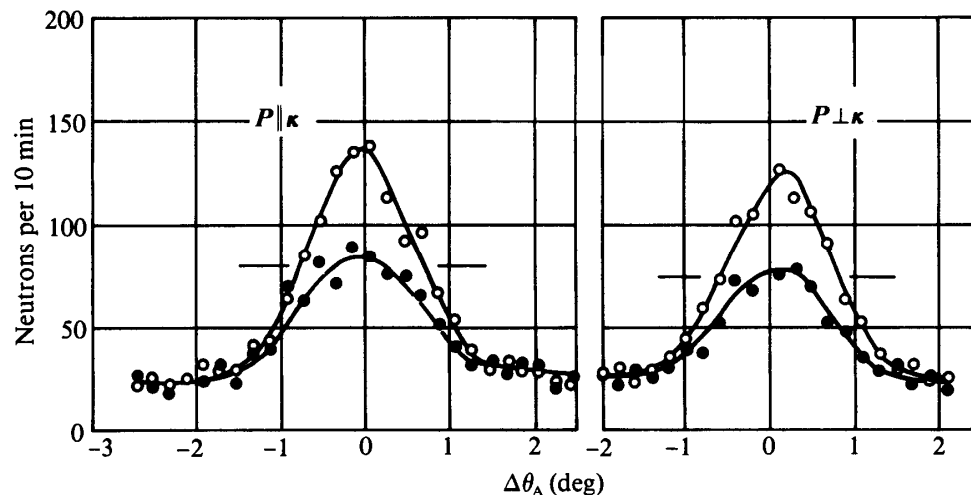
- Distinguish nuclear isotopic incoherent scattering from nuclear spin incoherent scattering.
- Isolate atomic paramagnetic scattering and so measure form factors
- Separate nuclear and magnetic structure scattering
- Study domain formation and magnetisation
- Measure polarisation of non-magnetic material due to contact with a magnetic layer

# Nuclear spin incoherent scattering

At any reasonable temperature and field, nuclear moments are oriented randomly at each site in a material. The interaction of the neutrons with this array of random vectors leads to incoherent scattering in much the same way as random chemical substitution in an alloy leads to incoherent x-ray scattering.

If the nuclear moment is perpendicular to the neutron polarisation vector then the neutron moment is flipped by the scattering event, whereas if nuclear moment is parallel to the neutron polarisation vector, the scattering involves no flip.

Since there are two ways for a moment to be perpendicular to  $\mathbf{P}$  and only one way to be parallel (in 3 dimensions), the spin-flip scattering has twice the intensity of the non-spin-flip scattering.

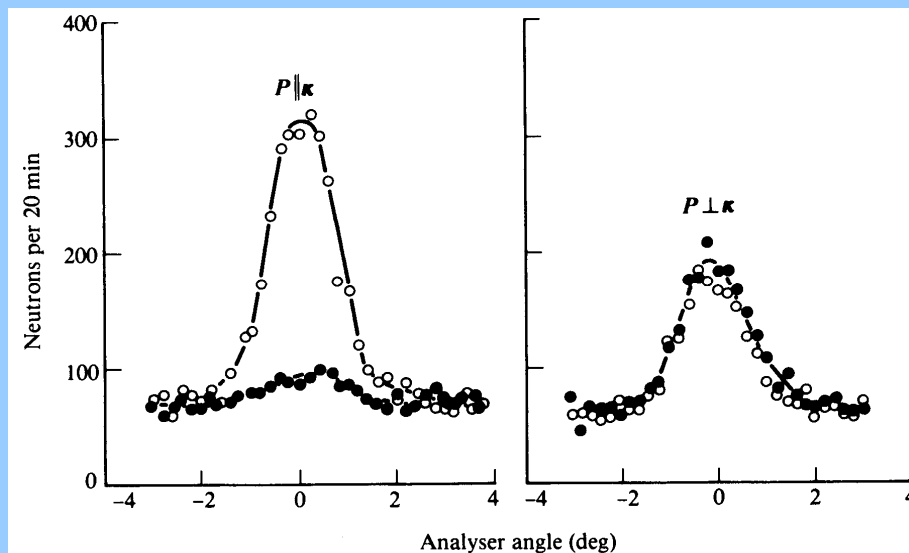


Nuclear spin incoherent scattering from  $^{51}\text{V}$ :  
○ SF, ● NSF.

# Paramagnetic scattering

A paramagnet is an array of randomly oriented atomic moments whose directions also change constantly in time. Scattering from this disorder leads to an elastic incoherent signal, much as nuclear spin disorder does. However scattering from atomic moments involves the EM interaction, not the strong force, and the vector nature of both moments matters. Only the atomic moment components perpendicular to the scattering vector  $\kappa$  can scatter the neutrons, and only those components perpendicular to the polarisation vector  $\mathbf{P}$  cause spin-flip scattering.

Thus, if  $\kappa \parallel \mathbf{P}$  all of the magnetic scattering is SF (it is not possible to be simultaneously parallel to the polarisation and perpendicular to the scattering vector), while if  $\kappa \perp \mathbf{P}$ , then the SF and NSF scattering signals are equal.

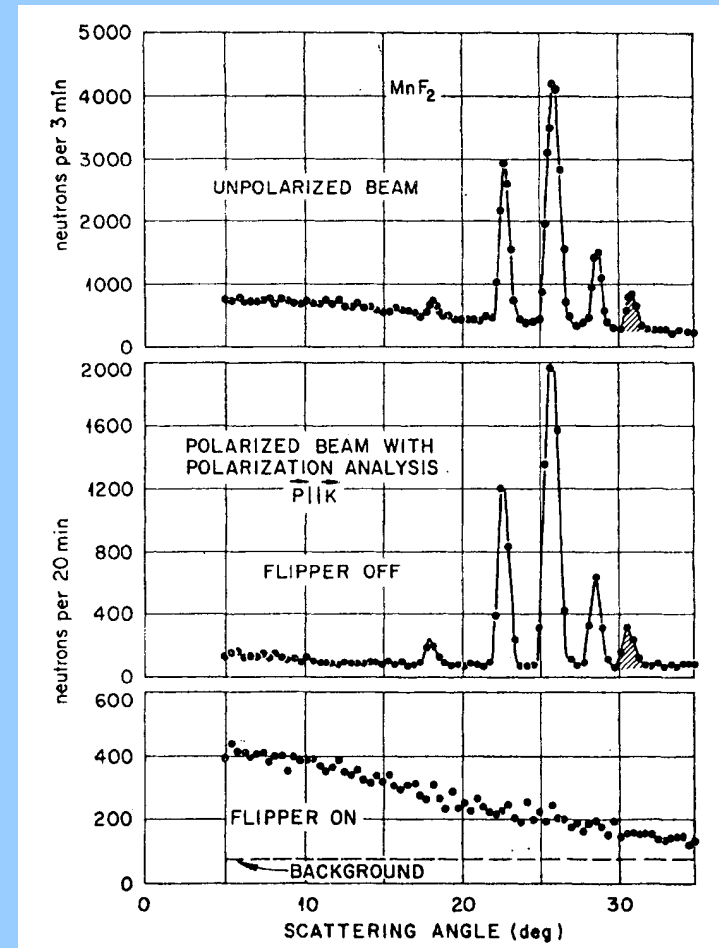


Paramagnetic scattering from  
 $\text{MnF}_2$ :  
○ SF, ● NSF.



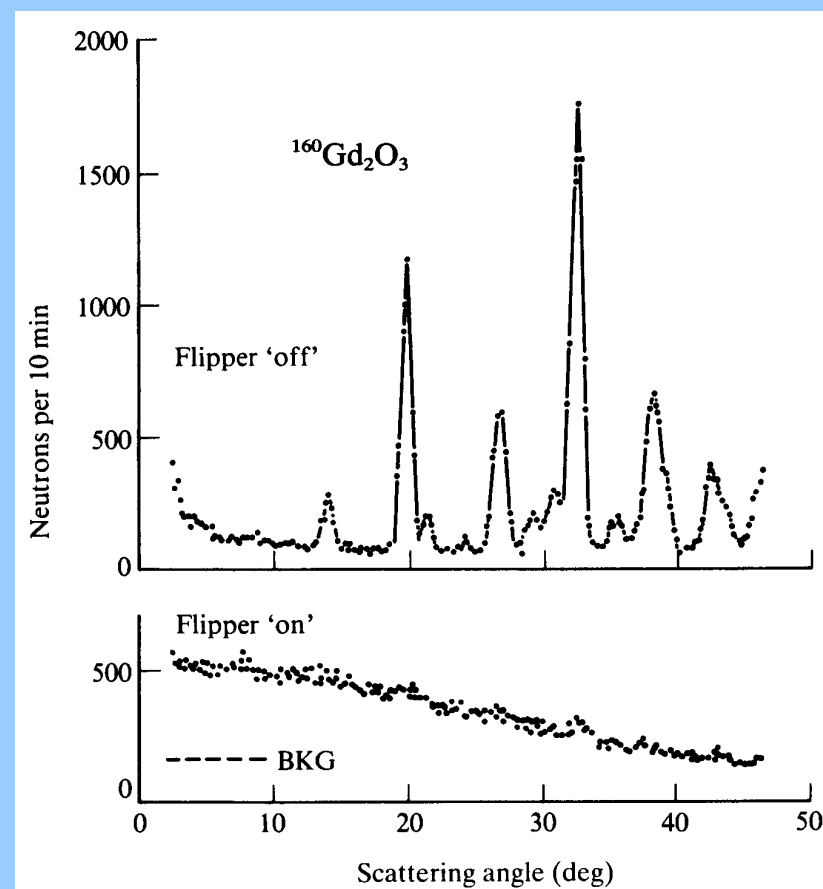
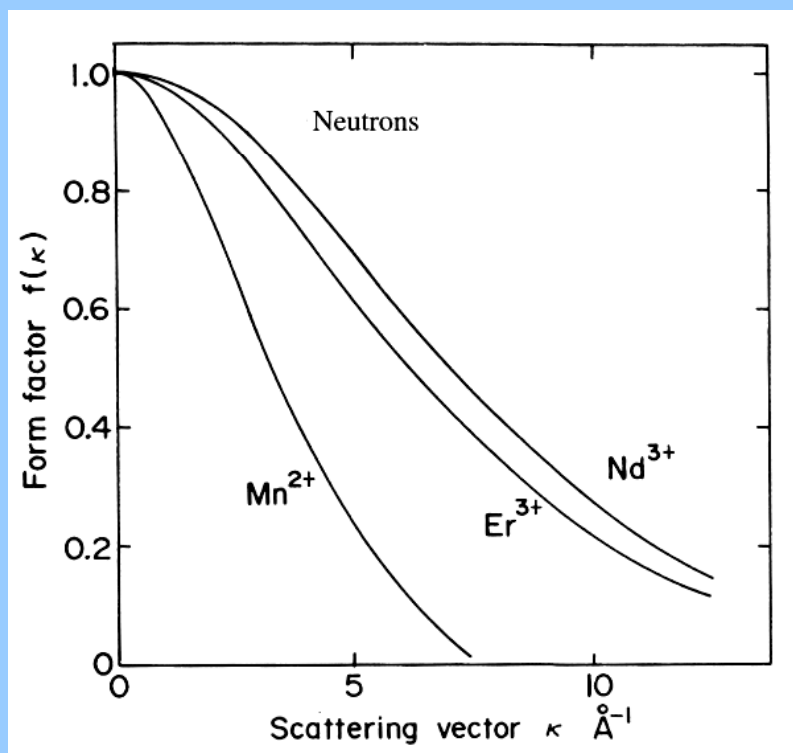
# Magnetic form factors

Unlike nuclear spin scattering, atomic magnetic scattering involves an interaction with an extended object – the atomic electron cloud – and so the associated form factor is more restricted in extent. This form factor can be measured directly using polarisation analysis to separate magnetic from nuclear scattering. If we work with  $\kappa \parallel \mathbf{P}$  then the NSF scattering is entirely nuclear in origin, while the SF scattering comes only from the atomic moments

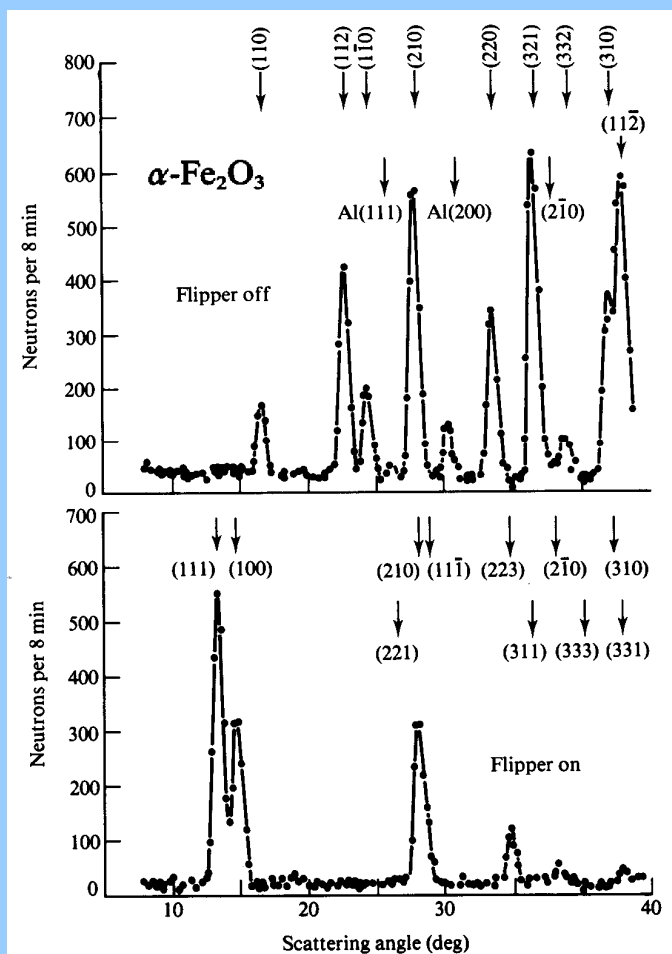


The unpolarised beam shows both the paramagnetic and nuclear scattering signals. Switching to a polarised beam leads to an immediate loss of intensity (check the vertical scales) but permits a complete separation of the two scattering signals. The paramagnetic signal is clear, even under the Bragg peaks.

The moments of the rare earths (*e.g.* Gd) are associated with the 4*f* electrons which are more compactly distributed than the 3*d* electrons that carry the moments of the transition metals (*e.g.* Mn). The form factor of Gd falls off more slowly than that of Mn.



# Separation of nuclear and magnetic scattering



Again, if we work with  $\kappa \parallel \mathbf{P}$  it is possible to distinguish nuclear Bragg from magnetic Bragg. This configuration is not useful for ferromagnets as the guide field will force the moments to be parallel to  $\kappa$  and so no magnetic scattering will be observed.

However, it can be used to look for non collinear order, and it is especially powerful for antiferromagnets where magnetic and nuclear scattering can be separated even when they overlap.

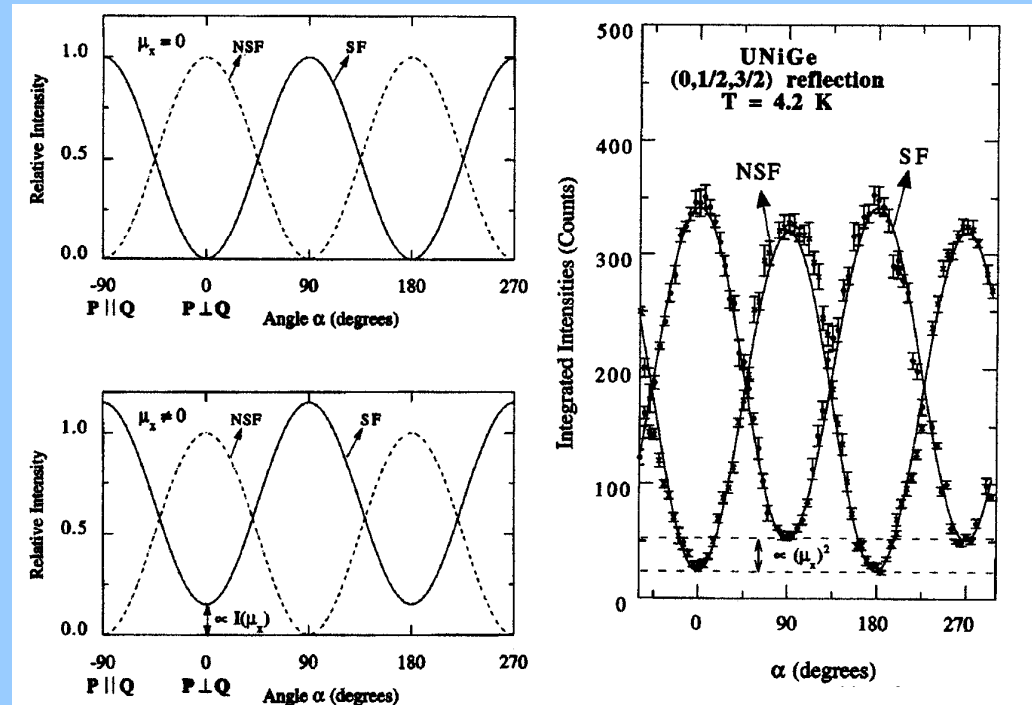
# Search for weak non-collinearity

Analysis of unpolarised neutron scattering data on UNiGe admitted two possible models:

1. all moments in the  $bc$ -plane
2. some canting out of the  $bc$ -plane

To distinguish these two cases, a measurement was made at fixed scattering vector  $\kappa$  but with the neutron polarisation held in the horizontal plane by a rotatable magnet so that  $\mathbf{P}$  could be swept around in the  $bc$ -plane.

The SF scattering does not go to zero when  $\mathbf{P} \perp \kappa$  (at 0 and 180 degrees). The departure from zero implies a canting angle of  $17 \pm 4$  degrees out of the  $bc$ -plane.



# Neutron Depolarisation

Polarisation analysis of scattering from ferromagnets is of very limited use. If we saturate the FM using the guide field then the moments are always parallel to  $\mathbf{P}$  and no SF scattering occurs in any geometry. If we fail to saturate the sample, then random precessions within the ferromagnetic domains will depolarise the beam.

At first sight this might appear to be a problem, however we can turn the situation to our advantage and use the depolarisation signal to *measure* the domains present in our sample.

As the neutrons pass through an array of domains in a ferromagnet they will undergo a series of random precessions. The domain walls are far too sharp for the neutron to be able to follow the changes in magnetisation direction adiabatically so the moment orientation remains fixed in space during the passage. Within each domain, the neutron moment will then precess about the local field before passing on to the next domain.

For a single, uniformly magnetised region of thickness  $d$ , the final polarisation,  $P$ , is given by:

$$P = P_o \left( \frac{B_{\parallel}^2}{B^2} + \frac{B_{\perp}^2}{B^2} \cos( c B d \lambda ) \right)$$

where:

$P_o$  is the initial polarisation

$$c = 4.633 \times 10^{14} \text{ m}^{-2} \text{ T}^{-1}$$

$B$  is the domain magnetisation.

Note: only  $B_{\perp}$  causes precession.

For many domains of average thickness  $\delta$ :

$$P = P_o \exp(-\alpha \lambda^2)$$

With

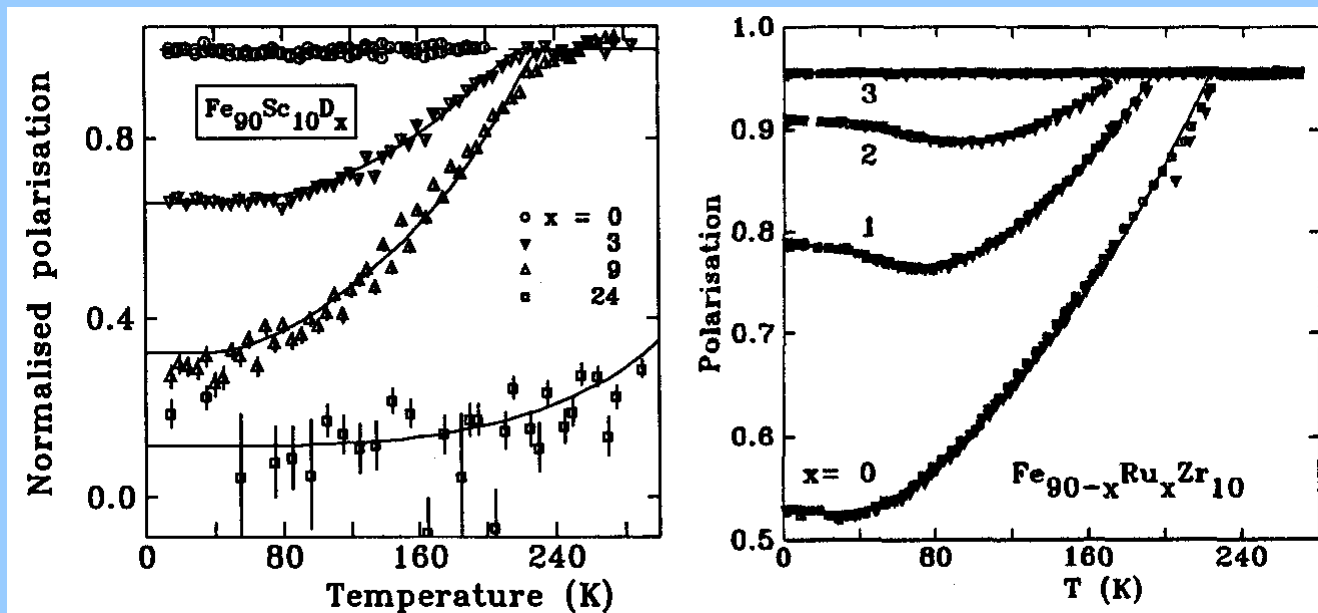
$$\alpha = \frac{1}{2} c^2 \langle B_{\perp}^2 \rangle d \delta$$

where:

$d$  is the sample thickness

$\langle B_{\perp}^2 \rangle$  is the mean square domain magnetisation perpendicular to the beam polarisation

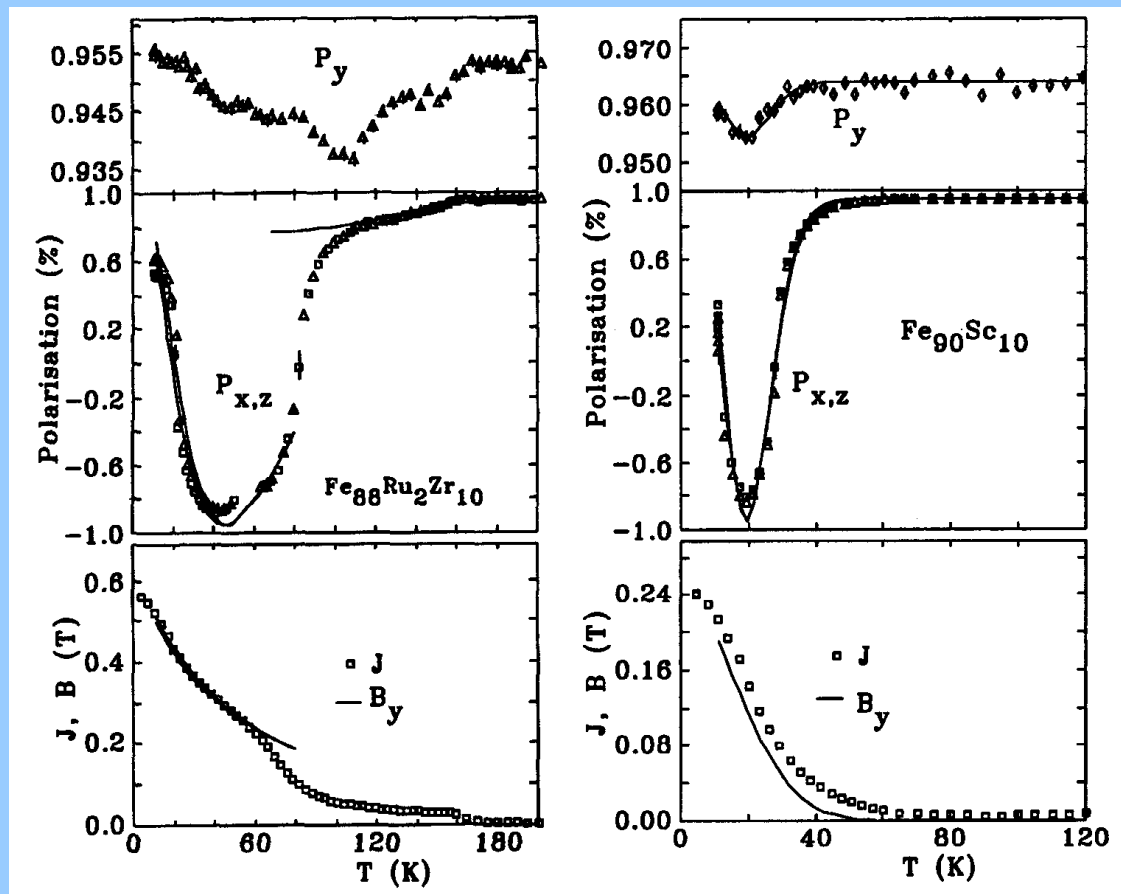
If we know the sample thickness and the bulk magnetisation then we can use the depolarisation signal to demonstrate that domains are actually present and to measure their average size through the thickness of the sample.



One weakness in the analysis is the assumption that the domain distribution is isotropic. Demagnetising fields and magnetostrictive stresses can greatly modify the domain texture. Furthermore, we observe a clear tendency for some samples to develop a field-cooled magnetisation in the guide fields used to control the neutron polarisation.

We added a set of Helmholtz coils around the sample so that we could adiabatically rotate the neutron polarisation parallel to  $x$ ,  $y$  and  $z$  and so probe all three domain texture axes.

We used the system to study the decay of field-cooled magnetisation in partially and fully frustrated magnets.





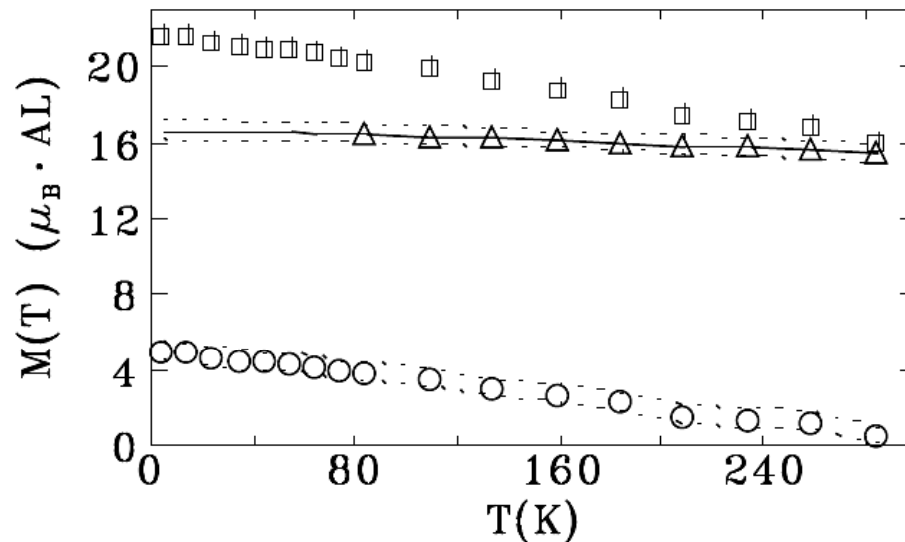
# Polarised Neutron Reflectometry (PNR)

Palladium is a metal that is *almost* ferromagnetic, and when placed in contact with a magnetic material it will develop a small moment. Even as little as 0.1% of Fe doped into bulk Pd will lead to long ranged order. An isolated Fe atom in Pd metal will induce a 'giant' moment of about  $10\mu_B$  within a cloud about 10 Å in radius.

These estimates came from measurements on doped bulk samples and were complicated by disorder effects which convolute the moment size with its extent. In order to obtain better estimates of both the size of the moment induced on the Pd and also how far the polarisation extends into the Pd metal, we reduced the problem to an ordered 2-d system and turned to PNR.

The first step was to optimise the Pd/Fe multilayer sample.

# Setting a scale



□ total magnetisation,  $\triangle$  Fe layer magnetisation deduced from LT-CEMS,  $\circ$  difference attributed to the Pd layers.

A combination of bulk magnetisation and low-temperature conversion electron Mössbauer spectroscopy (LT-CEMS) was used to establish the magnitude of the Pd polarisation and its temperature dependence.

# Defining the problems

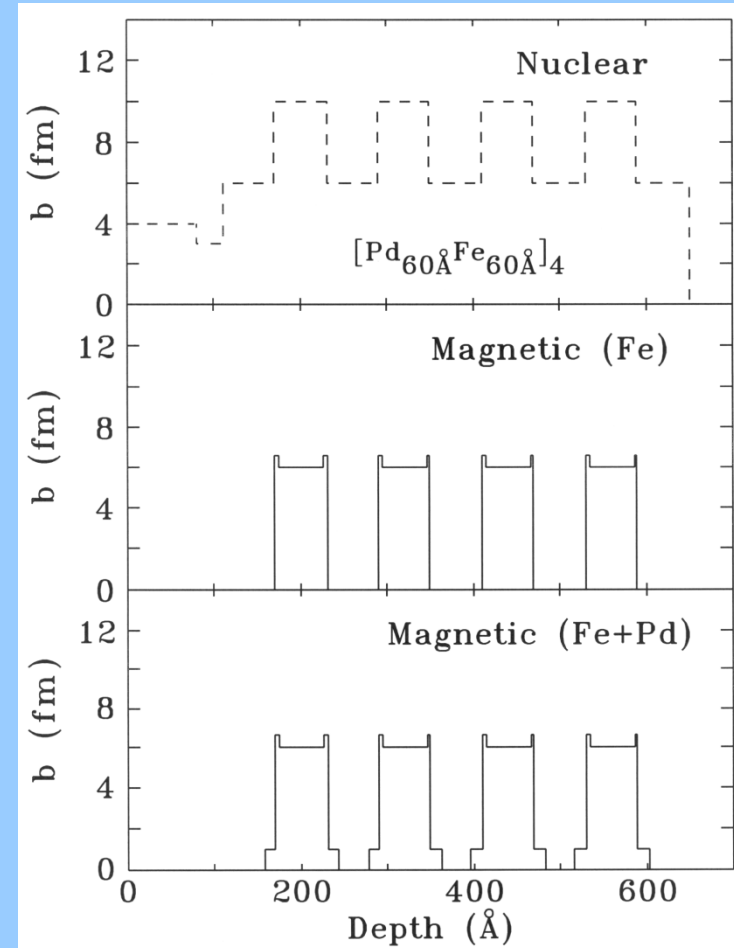
- Previous attempts have failed to find a signal
- Scattering due to small Pd moment will be about 1 *fm*
- Nuclear scattering is much larger (  $b_{\text{Fe}} \sim 10 \text{ fm}$ ,  $b_{\text{Pd}} \sim 6 \text{ fm}$  )
- Magnetic scattering from Fe is about 6 *fm*
- Pd moment is strongly temperature dependent so we need to work at 5K. Even then, the signal will be weak.

# Optimisations

- Match Pd and Fe layer thicknesses
- Tune layer thickness to expected polarisation depth
- Work with polarised neutrons so that we can compare 'up—up' and 'down—down' scattering
- Exploit strong temperature dependence of Pd moment: scattering changes between RT and 5K must be due primarily to Pd

# Why match the layer thicknesses?

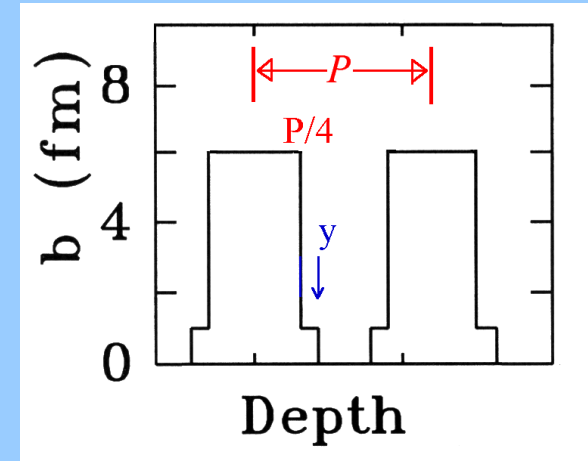
- Low-angle scattering gives Fourier transform of layer structure
- Accurately matched layer thicknesses means that the *chemical* scattering length profile is square-wave modulated
- Only *odd* harmonics present in transform
- Magnetic scattering from Fe layers is fully correlated with chemical scattering, so it does not affect the square-wave modulation
- Magnetic scattering from polarised Pd breaks symmetry and leads to *even* harmonics



# Tune layer thickness

$$\begin{aligned} a_1 &= \int_0^{P/4} 1 \cdot \cos(\omega x) dx + \int_{P/4}^{P/4+y} b \cdot \cos(\omega x) dx \\ &= \frac{P}{2\pi} \left[ 1 + b \left( \cos\left(\frac{2\pi}{P}y\right) - 1 \right) \right] \end{aligned}$$

$$\begin{aligned} a_2 &= \int_0^{P/4} 1 \cdot \cos(2\omega x) dx + \int_{P/4}^{P/4+y} b \cdot \cos(2\omega x) dx \\ &= (-1) \frac{P}{4\pi} \frac{b}{2} \sin\left(\frac{4\pi}{P}y\right) \end{aligned}$$



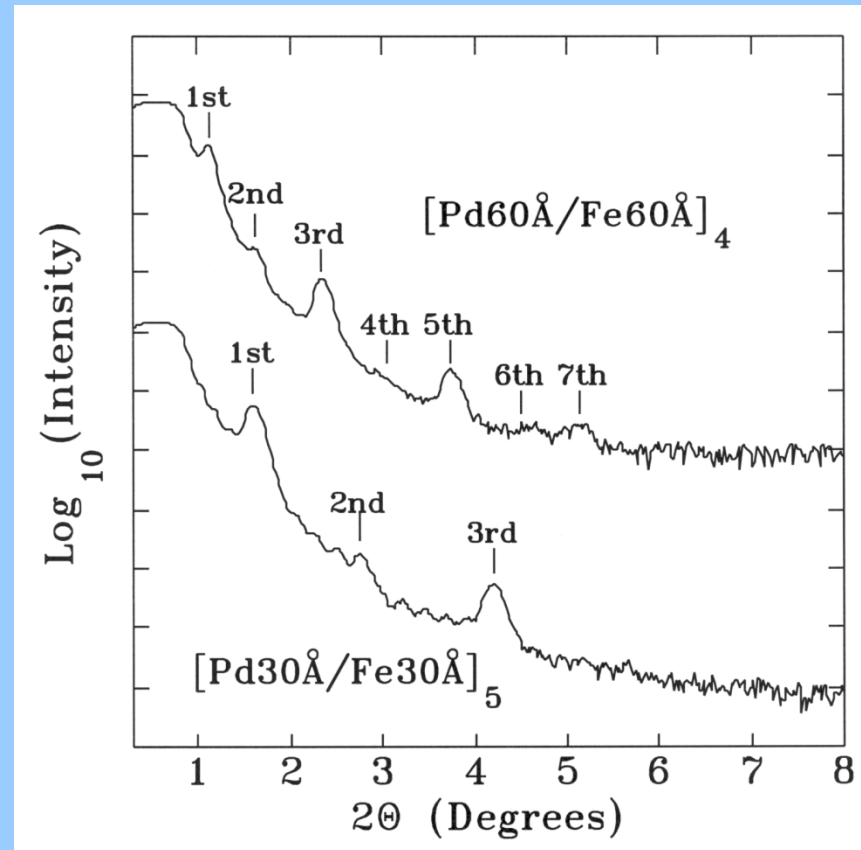
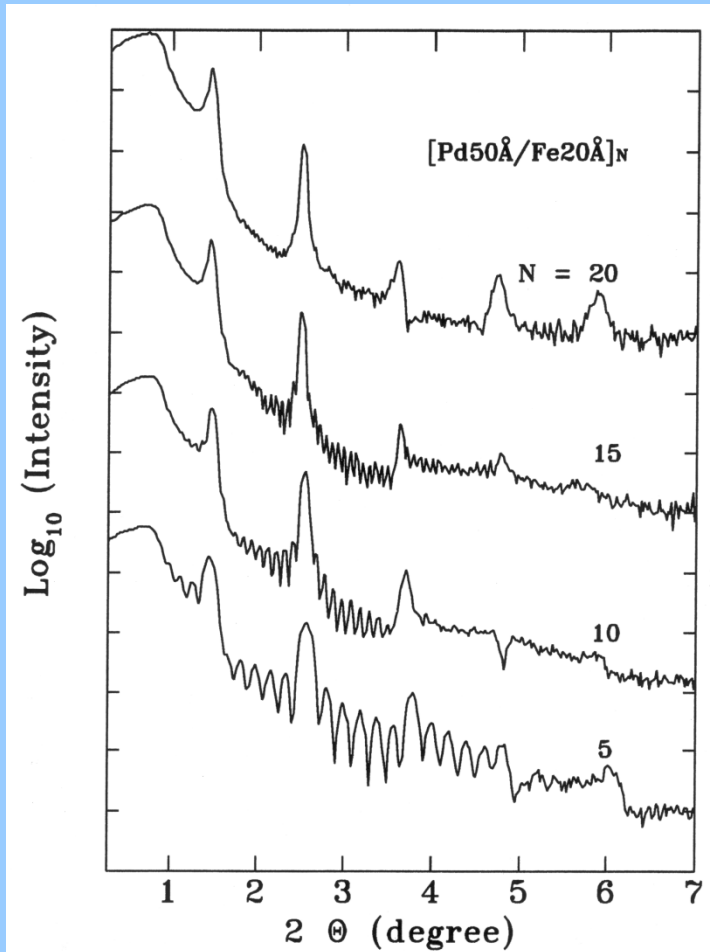
Second order term is maximised for  $y = P/8$

*i.e.* we want half of palladium layer to be polarised

CEMS estimate of  $t_{pol} \Rightarrow 2y = 25 \pm 3 \text{ \AA}$

$\rightarrow$  Optimum period is  $\sim 100 \pm 12 \text{ \AA}$

# Effects of layer matching

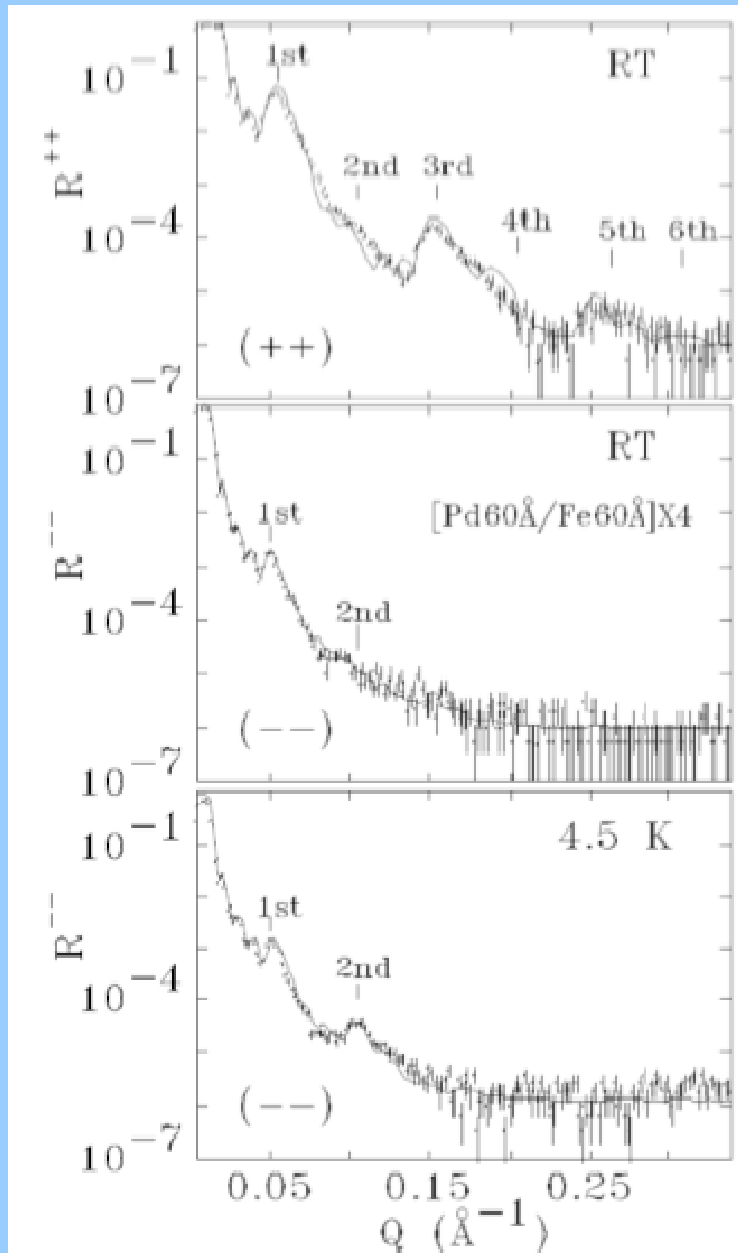


At room temperature the estimated Pd polarisation is essentially zero, so the magnetic and nuclear scattering densities simply add in the  $R^{++}$  channel to yield a scattering pattern very similar to that seen by x-ray reflectometry.

However, in the  $R^{--}$  channel the magnetic and nuclear scattering from the Fe layers almost cancel, and the net scattering density of the Fe almost matches that of the Pd layers. The PNR signal is quite weak.

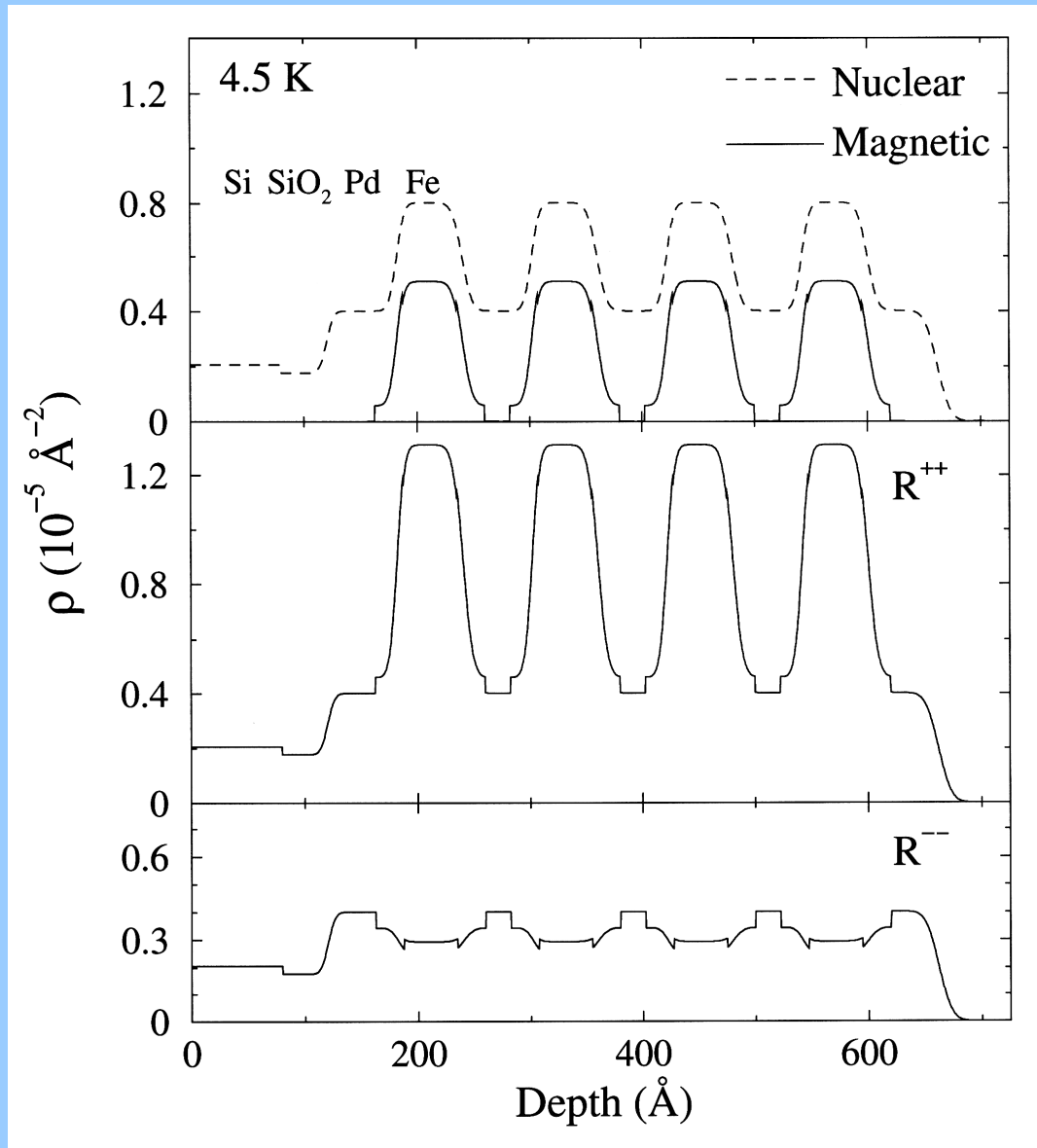
At 4.5 K the polarisation that develops in the Pd layer greatly changes the net scattering density profile and a strong signal appears in the second-order position.

This can only arise from a breaking of the simple square-wave symmetry of the scattering density and so must come from new magnetic scattering in the Pd layers.





The rest is an exercise in computer fitting and model building.



•  $\mu_{\text{Pd}} = 0.32 \pm 0.02 \mu_{\text{B}}$

•  $t_{\text{pol}} = 20 \pm 4 \text{ \AA} = 8.9 \pm 1.8 \text{ ML}$

# Some useful references

## **General**

- “*Polarised Neutrons*”, W.G. Williams, Oxford Science Pubs. (1988)

## **Magnetic Scattering**

- R.M. Moon *et al.*, Phys. Rev. 181, (1969) 920.
- C.G. Schull *et al.*, Phys. Rev. 83, (1951) 333.
- C.G. Schull *et al.*, Phys. Rev. 84, (1951) 912.
- R.M. Moon *et al.*, Phys. Rev. B5, (1972) 997.

## **Instrumentation**

- R. Pynn, Rev. Sci. Instrum. 55, (1984) 837

## **LONGPOL**

- S.J. Campbell *et al.*, J.Phys.E 7 (1974) 195.
- N. Ahmed *et al.*, J.Phys.E 7 (1974) 199.

## **Polarisers**

- A. Delapalme *et al.*, NIM 95 (1971) 589.
- J.W. Lynn *et al.*, J. Appl. Cryst. 9 (1976) 454.
- F. Tasset, Physica B 213 & 214 (1995) 935.

## **Depolarisation**

- O. Halpern and T. Holstein, Phys. Rev. 59 (1941) 960.
- M.T. Rekveldt, Physica B 267–8 (1999) 60.

# *Many* thanks to...

- **Zin Tun and Zahra Yamani**, (CNBC) who have tried so hard to teach me about neutron scattering.
- **Ron Donabarger, Chad Boyer and Raymond Sammon** (CNBC) for making things work every time I visit.
- **Larry McEwan** (CNBC) for making things, or making them fit, at the last moment.
- **Mel Potter** (CNBC) for creating a unique operating mode on C5 just for me
- **Sean Cadogan** (U.Manitoba) for doing all of the neutron fitting.

and the *many* others that have made it such a pleasure to visit the CNBC for so many years.