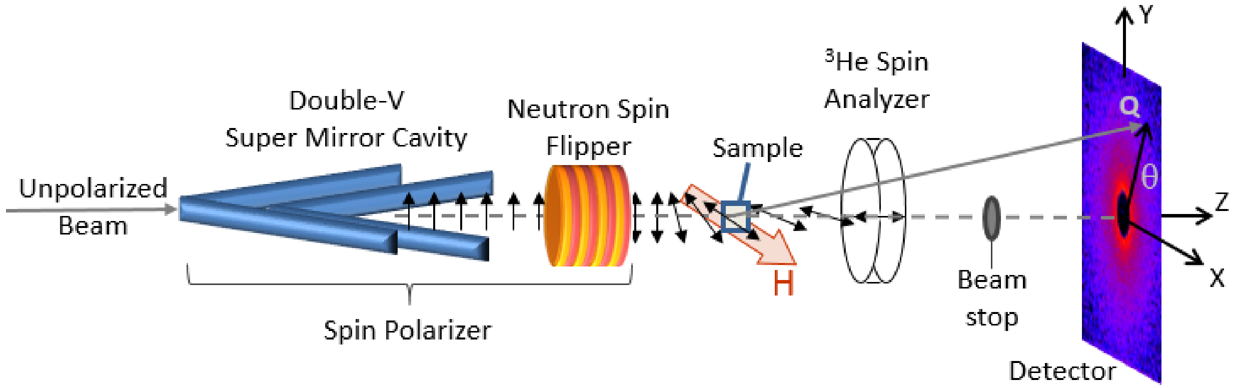
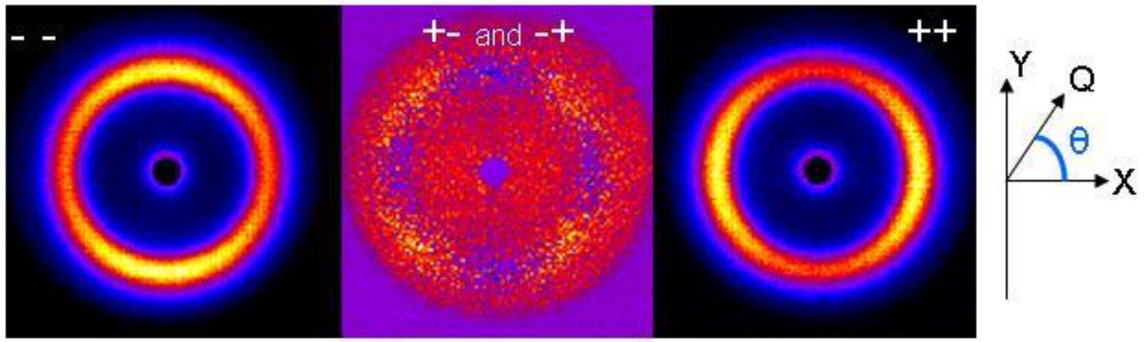
Overview of Polarization Analysis for Neutron Scattering

(Originally given June 17, 2020 at the National School on Neutron and X-Ray Scattering)

Kathryn Krycka

NIST Center for Neutron Research, Gaithersburg, MD





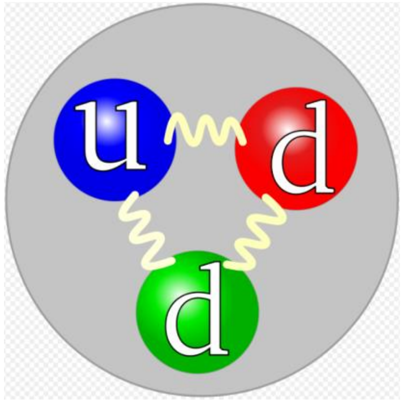
Support has been provided by the Center for High Resolution Neutron Scattering, a partnership between the National Institute of Standards and Technology and the National Science Foundation under Agreement No. DMR-1508249.

Outline

- How to prepare and characterize a polarized neutron beam
- What magnetic information polarization analysis can provide, motivated by a nanoparticle sample
- Beam and sample depolarization considerations
- A more detailed look at the cross-terms and chiral structures
- Discussion of alternative geometries

Neutron Properties

- Hadron comprised of three quarks; no net charge (insensitive to electric fields)



https://en.wikipedia.org/wiki/Neutron

- Free lifetime of 881.5±1.5 seconds (15 minutes)
- Spin ½ (fermion) that gives rise to a magnetic moment
- $\mu_n = -1.913\ \mu_N$ (nuclear magneton) $= -9.662 \times 10^{-27}$ J/T (or $\approx \mu B / 1000$)
- Spin and magnetic moment are oppositely oriented, complicating what “up” and “down” mean with respect to an applied field.

Neutron's Response to a Static Magnetic Field*

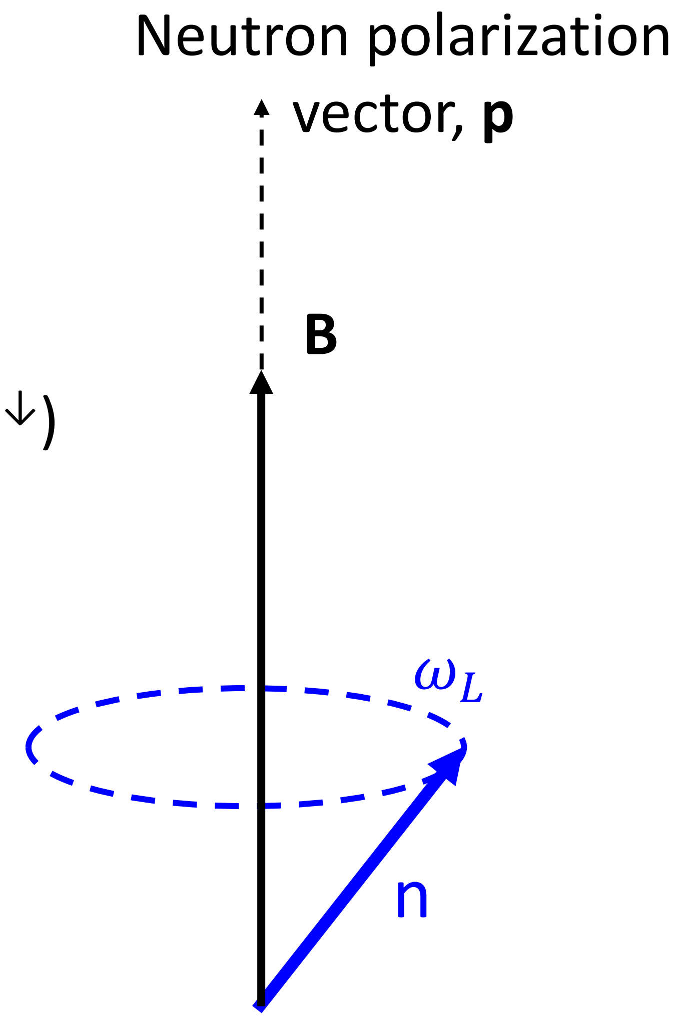
- A neutron can be represented by a spinor wave function with spin eigenstates "up" and "down" (+,- or ↑,↓):

- Polarization of a single neutron is the expectation value of the appropriate Pauli matrix

- Neutron beam polarization (many neutrons), P ≡ (n↑- n↓)/(n↑+ n↓)

- The time dependence a two-state quantum system can be represented by a classical vector, $$\frac{d\vec{P}}{dt} = -\gamma_L \vec{P} \wedge \vec{B}$$

- Gyromagnetic ratio $\gamma_L = -1.833E4$ rad/Gauss−sec and Larmor frequency $\omega_L = -\gamma|B|$



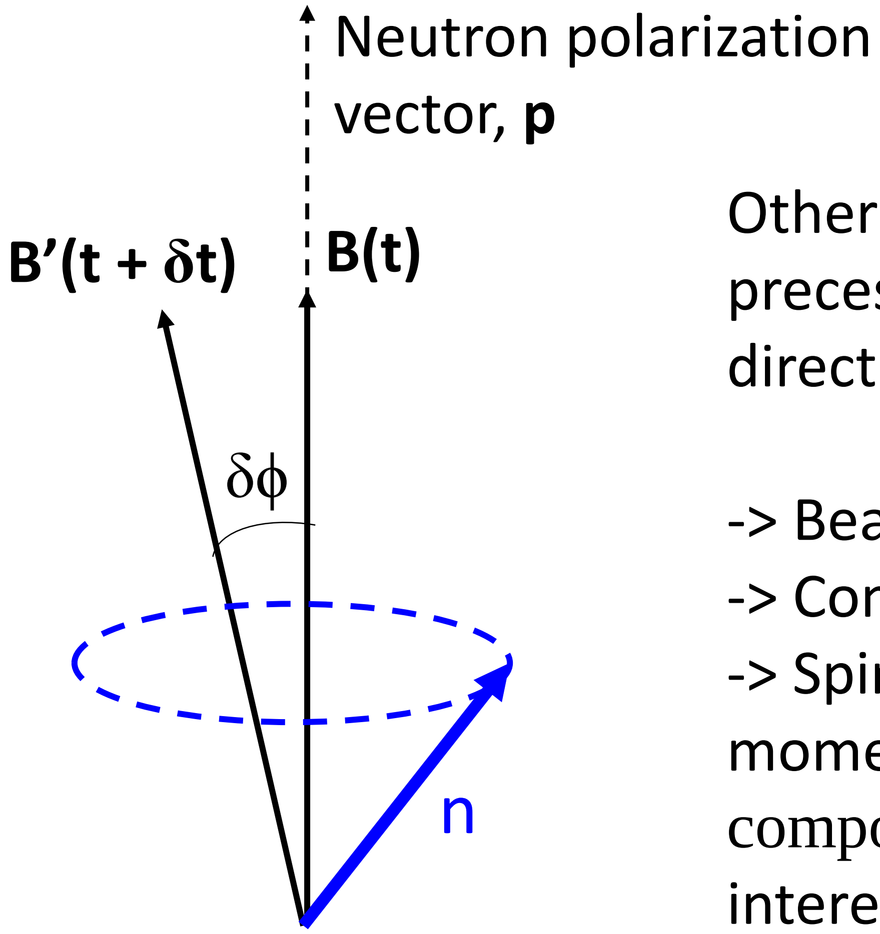
*Reproduced largely from Roger Pynn's Neutron Scattering Lectures

Neutron's Response to Varying Magnetic Field

Neutron adiabatically follows field (retains polarization) if

$$\frac{d\phi}{dt} << |\varpi_L|$$

$\omega_L = -\gamma|B| = -1.833E4$ rad/Gauss−sec



Otherwise, the neutron will precess about a new field direction

-> Beam depolarization
-> Controlled flipping devices
-> Spin flip from magnetic moments ($\perp$Q and with component $\perp$ p) in sample of interest

Neutron's Response to Material

- Neutrons are sensitive to changes in (structural) scattering length density, and this is independent of the neutron's spin direction and doesn't alter the resulting spin direction

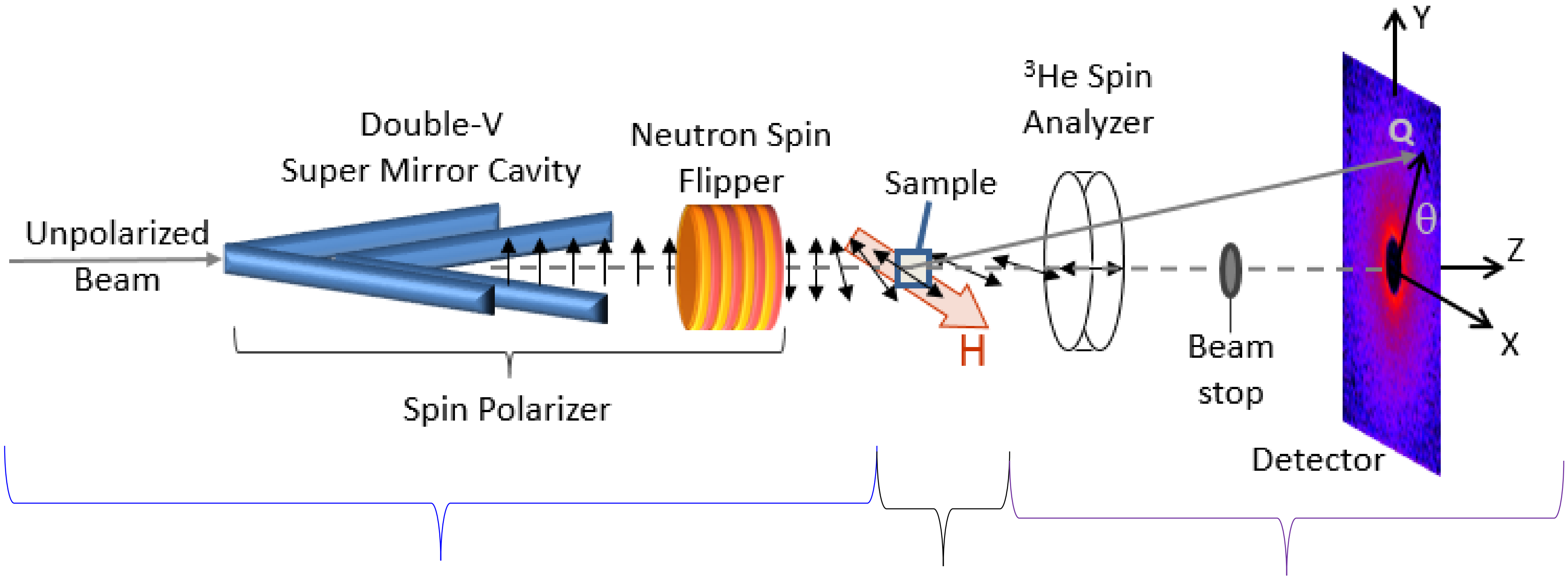
- Only the component of the magnetic moment (or magnetic form factor), M, that is $\perp$ Q may participate in neutron scattering. This is embodied in the Halpern-Johnson vector (Phys. Rev. 55, 898 (1939)) as:

$$\Upsilon(\hat{\mathbf{Q}}) = \mathbf{M} - (\hat{\mathbf{Q}} \cdot \mathbf{M})\,\hat{\mathbf{Q}} = |\mathbf{M}|\,[\hat{\mathbf{M}} - (\hat{\mathbf{Q}} \cdot \hat{\mathbf{M}})\,\hat{\mathbf{Q}}]$$

- Of M $\perp$ Q (defined by Υ), the portion || **p** contributes to non-spin flip (NSF), while the portion perp **p** contributes to spin-flip (SF) scattering -- Moon, Riste, Koehler (Phys. Rev. 181, 920-931 (1969)) where A || **p** and B x C = A:

$$\sigma^{\downarrow\downarrow}_{\uparrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}\left|N \pm \Upsilon_A\right|^2, \quad \sigma^{\uparrow\downarrow}_{\downarrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}\left|(-\Upsilon_B \mp i\Upsilon_C)\right|^2$$

General Experimental Set-Up (SANS example shown)



Front-End:
Typically polarize , maintain polarization, and modify direction of neutrons for a non-divergent beam

Sample Scattering:
Often 1D analysis w/ B-field, but 3D analysis w/o field possible (spherical polarimetry)

Back-End:
Maintain polarization, analyze neutron spin direction for non-divergent (reflectivity, diffraction) or divergent (SANS, off-specular) beam

Beam Polarization

- In the presence of an applied field (B), half the neutrons precess about direction || B, and half precess anti-|| to B

- P ≡ (n↑- n↓)/(n↑+ n↓), so the goal is to absorb or reflect away the undesired spin state

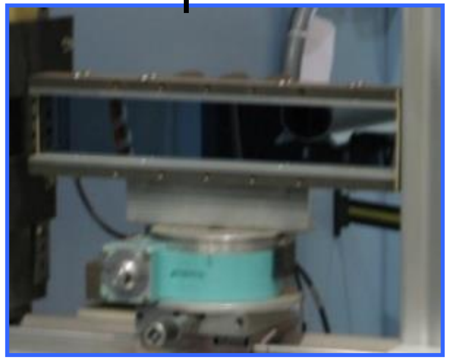
- Polarizing Monochromator (Heusler, Cu_2MnAl)
  -Good when monochromatic, non-divergent beam is required

- Supermirror (FeSi multilayers)
  -Pros: Polarizes a range of wavelengths; high efficiency
  -Con: Beam must be non-divergent (or benders required)

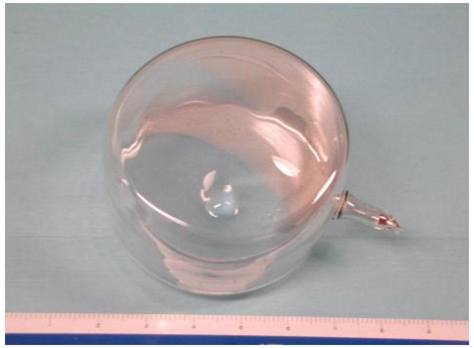
- Spin Filter (^3He)
  - Pro: can analyze wide range of angles, ability to flip neutrons
  -Cons: Needs to be repolarized over time; λ-dependent

Heusler

Fe|Si



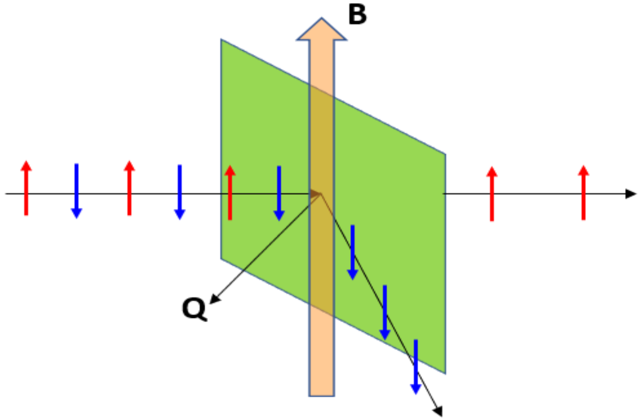
^3He

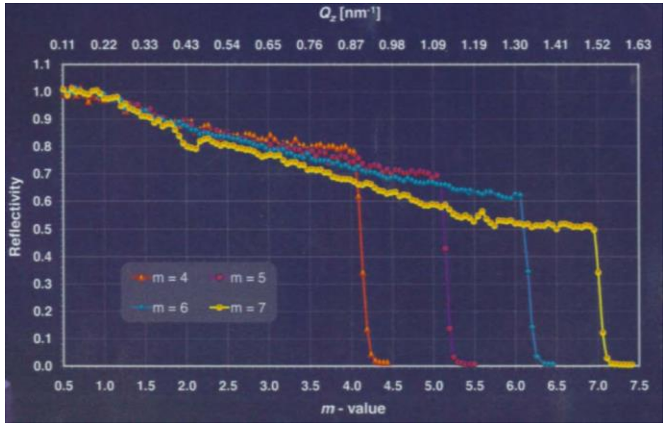


Polarizing Crystal, Supermirror

- Neutrons only scatter from moments ⊥ Q. Neutron with spins || B experience a decrease in potential, while those anti-|| to B experience an increase in potential.

SLD for n↑, n↓ = $b_{nuclear}$ -,+ $b_{magnetic}$

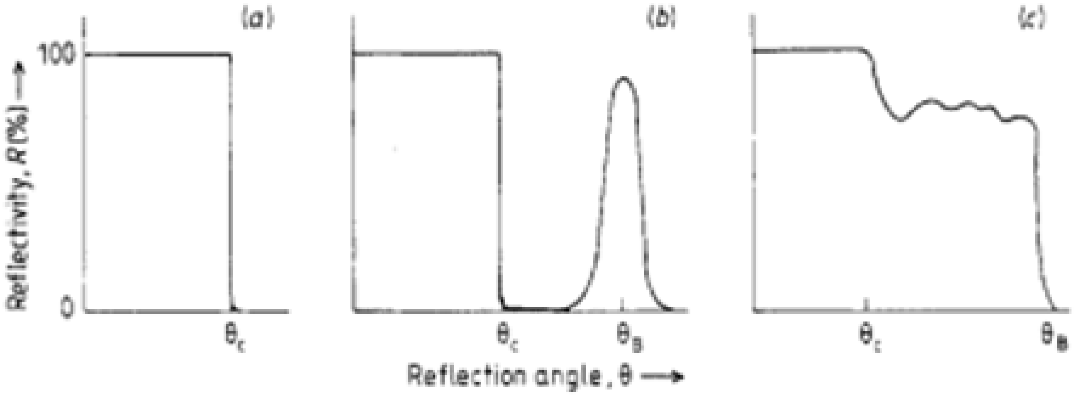




Courtesy of Swiss Neutronics

- By choosing materials with same nuclear and magnetic SLDs, obtain reflection for the n↓ state and transmission for the n↑ state.

- The same idea applies the *average* nuclear and magnetic SLDs of layered materials. By varying the distance between layers, multiple critical angles may be achieved, extending the angular acceptance for reflection.

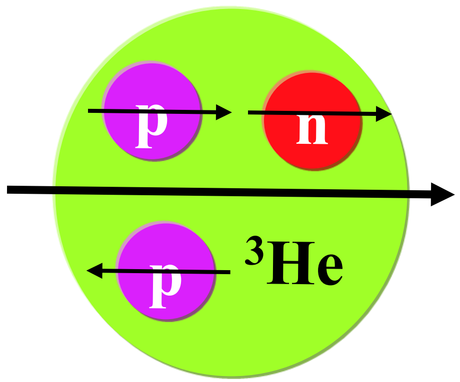


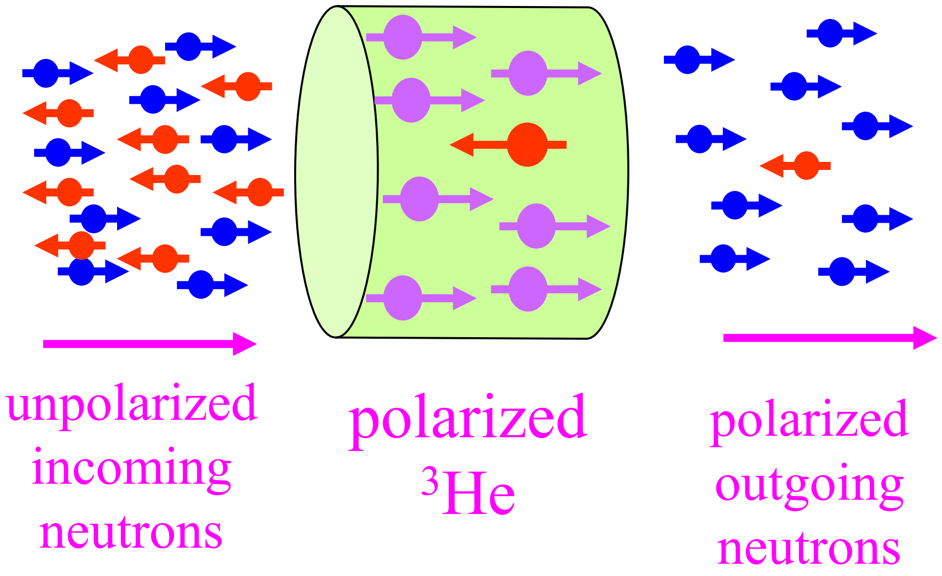
(a) Reflecting mirror, (b) multilayer, (c) supermirror
J W White and C G Windsor, Rep. Prog. Phys. V 47, 707-765, 1984

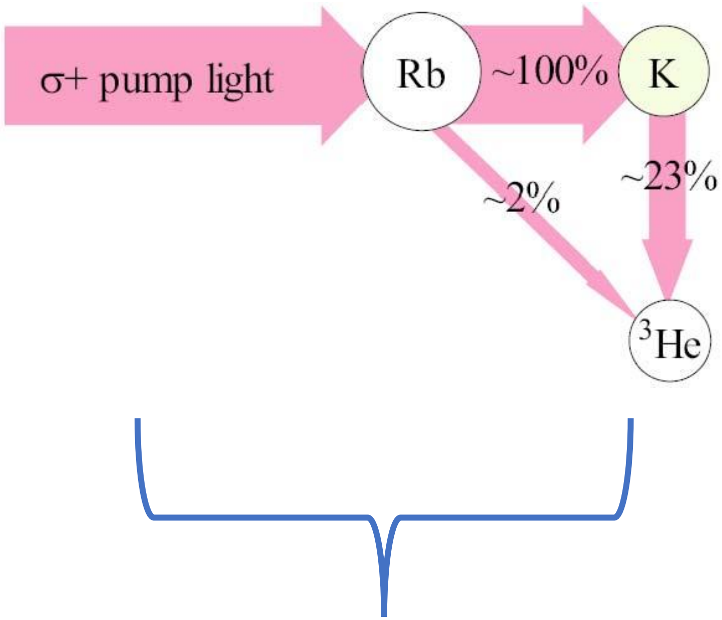
^{3}He Spin Filters

^{3}He nuclear spin carried mainly by the neutron

K.P. Coulter et al, NIM A 288, 463 (1990)







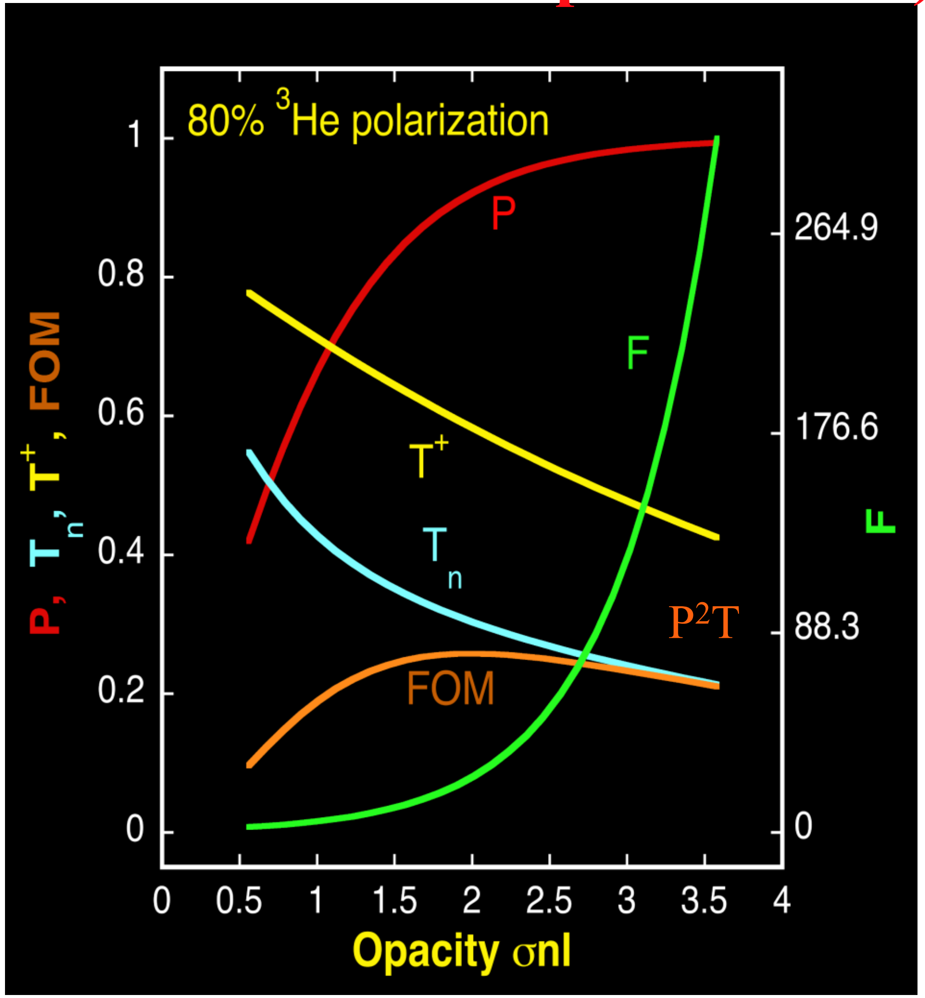
^{3}He optically pumped to desired spin state

- strongly spin-dependent neutron absorption cross section.
- anti-aligned neutrons see a thick absorption target, aligned neutrons see a thin target.

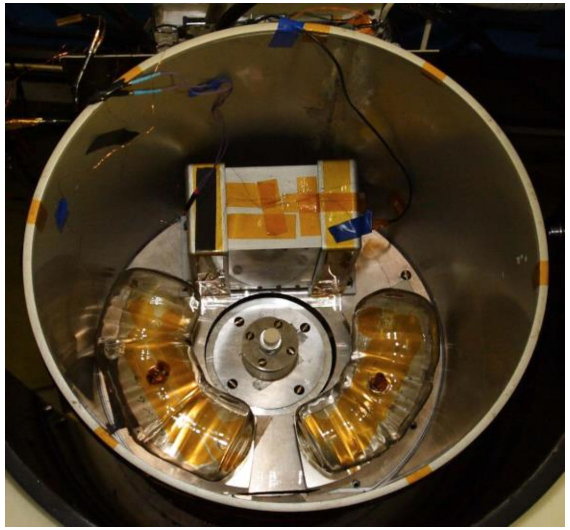
Courtesy of Wangchun Chen. https://www.nist.gov/ncnr/spin-filters/spin-filter-info/seop-method/optical-pumping

Polarized ^{3}He Neutronic Performance

80% ^{3}He polarization (higher effective neutron polarization)



Courtesy of Wangchun Chen



Spin Filters can be made in a wide variety of shapes to accommodate many forms of wide angle diffraction

Neutron Spin Flipping

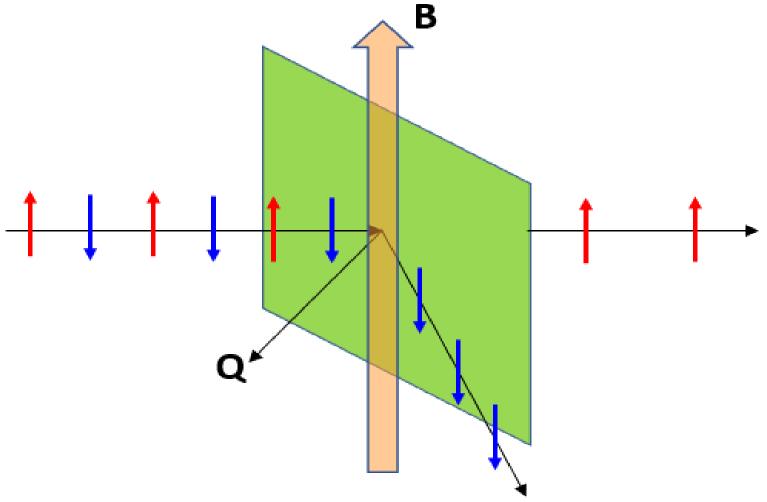
- Spin reversal must occur with respect to the polarization axis (a simple change in the polarization axis direction does not work)

- For ^3He, spin reversal is built in by reversing ^3He spins via RF pulse

- If can rotate your supermirror angle, may be able to vary between spin states (transmission vs. reflection)

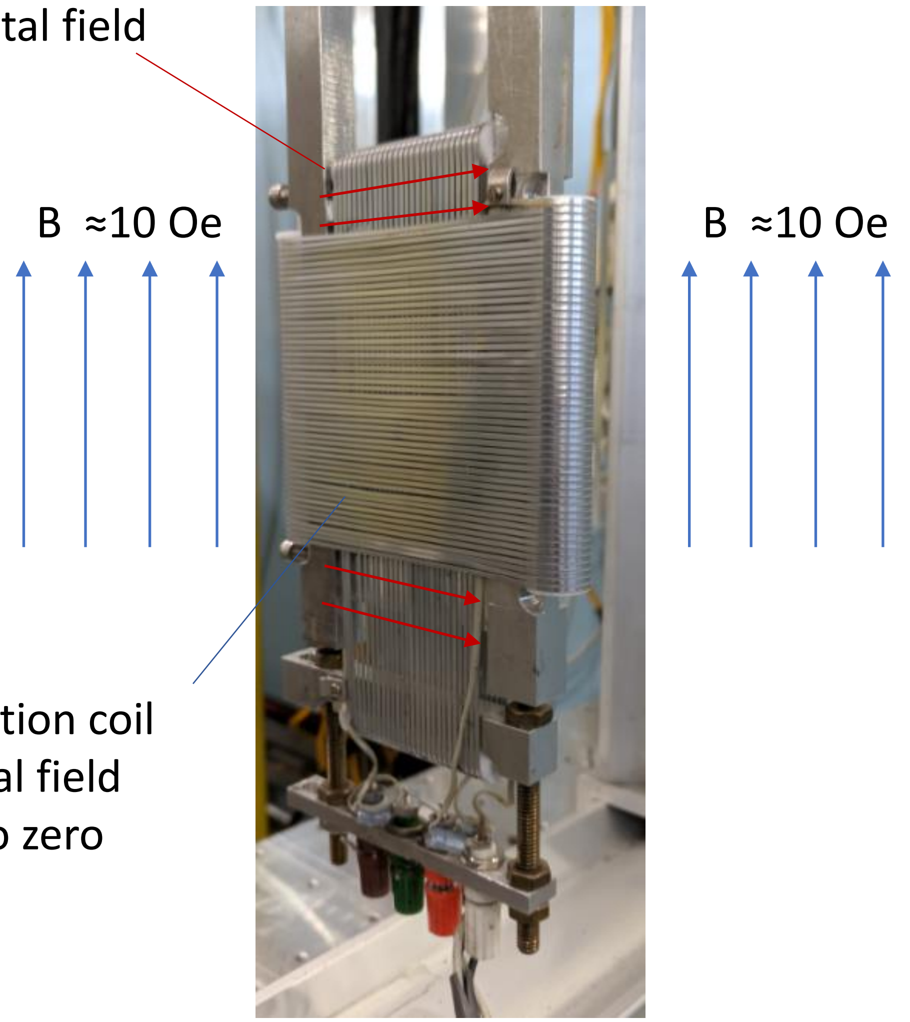
- Mezei or coil flipper (tuned for specific neutron wavelength, material in beam)

- White-beam, gradient field spin-flipper (appropriate for multiple wavelengths, no material in beam)



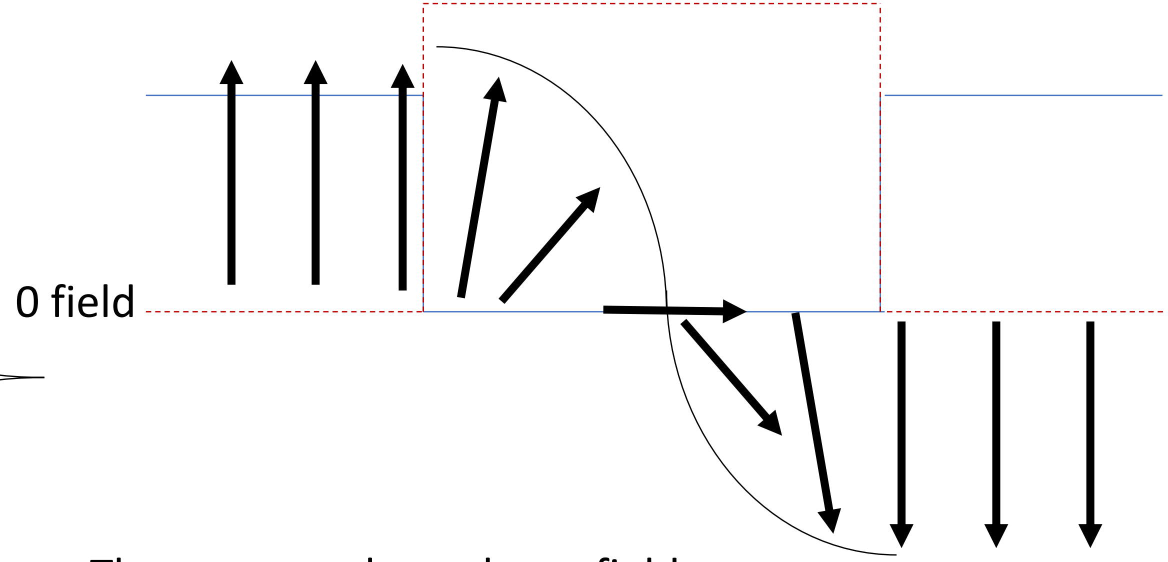
Mezei or Coil Flipper

Flipper current sets up a horizontal field (≈ 15 Oe)

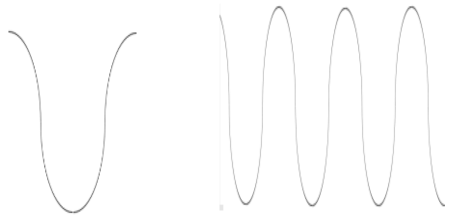


Compensation coil sets vertical field abruptly to zero

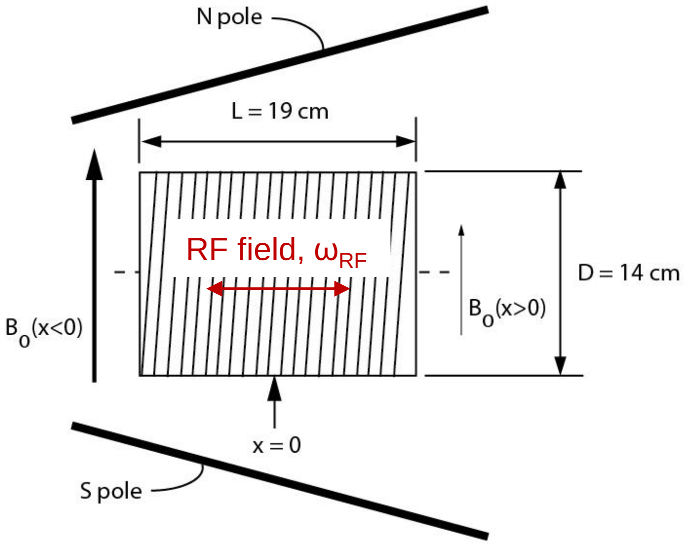
Blue is vertical field
Red is horizontal field
Black neutron spins



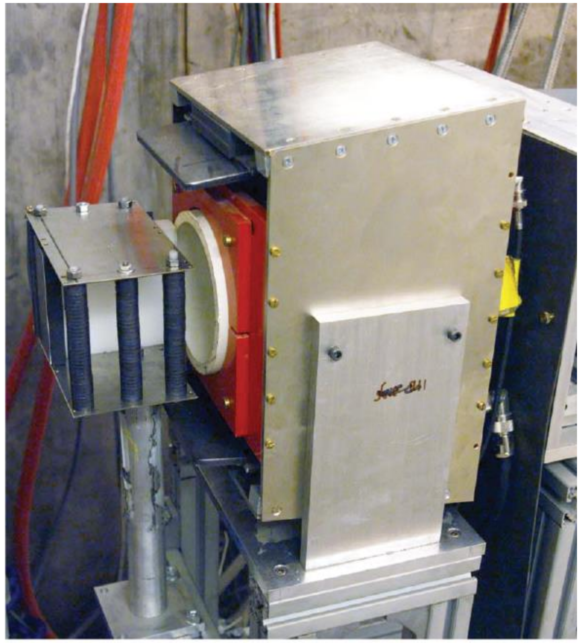
- The neutron always has a field to follow (never loses polarization axis)
- But the spin is reversed with respect to the applied field upon completion
- Other rotations possible:



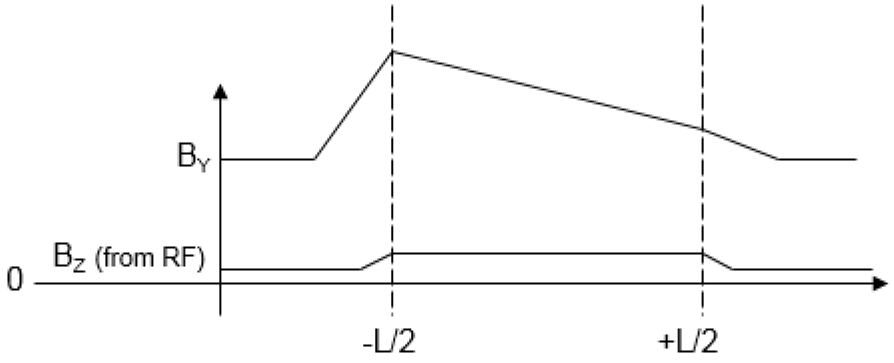
Spin Gradient, RF-Flipper



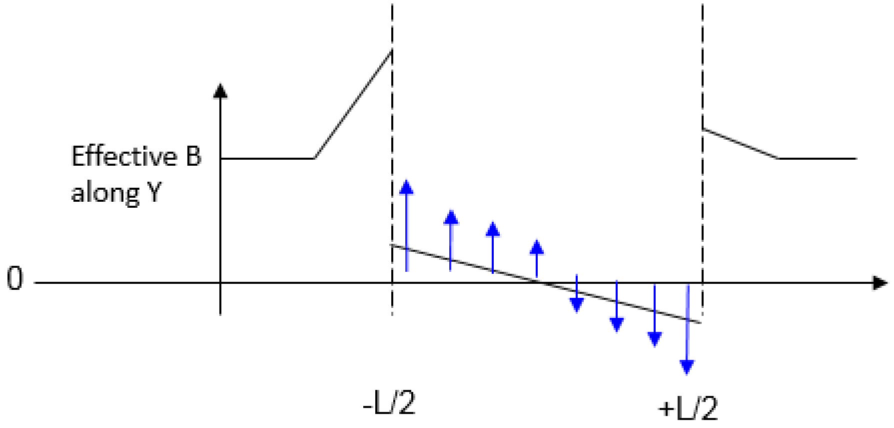
300 kHz ~ 100 G



Asterix Flipper, M. Fitzsimmons



$$B_{Y,\ effective} = B_Y - \omega_{RF} / \Upsilon$$



- If satisfy adiabatic condition along Z for fastest neutron, then neutrons of higher λ's will also flip
- Accommodates wide range of wavelengths
- Very high efficiencies
- Nothing in the neutron beam

C.P. Slicther, Principles of Magnetic Resonance, (Springer Verlag, Berlin 1980).

Polarized Beam Characterization

- Flipping ratio (F.R.) = n↑ / n↓, measures on transmitted beam (assume sample scattering is negligible if sample present)
  Flipping ratios > 30 decent; can be much higher

- Polarization, P ≡ (n↑- n↓)/(n↑+ n↓) = (F.R. – 1)/(F.R. + 1) and polarization efficiency, ε, ≡ (n↑)/(n↑+ n↓) = (1 + P)/2

- ^{3}He atomic polarization ($\wp_{^3\text{He}}$)can be determined from unpolarized transmissions where T_E = glass transmission and μ = opacity of cell (dependent on neutron wavelength, gas pressure, and cell length – typically determined in advance from transmission measurements of unpolarized ^{3}He cell)

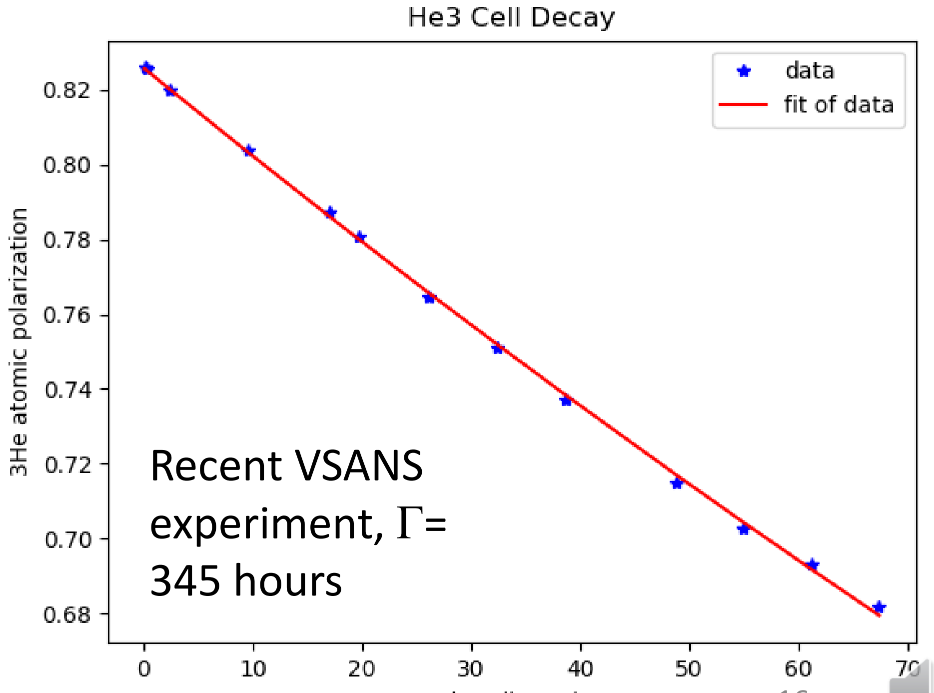
$$T^{\text{majority, minority}}_{^3\text{He}} = T_{\text{E}} \exp\left[-\mu(1 \mp \wp_{^3\text{He}})\right]$$

$$\wp_{^3\text{He}} = a\cosh\left[\frac{T^{\text{unpol beam}}_{\text{(polarized) }^3\text{He cell}} - T_{\text{background noise}}}{T^{\text{unpol beam}}_{^3\text{He cell OUT}} - T_{\text{background noise}}}\,\frac{1}{T_{\text{E}}\exp(-\mu)}\right]/\mu.$$

$$P_{\text{cell}} = \tanh(\mu\wp_{^3\text{He}})$$

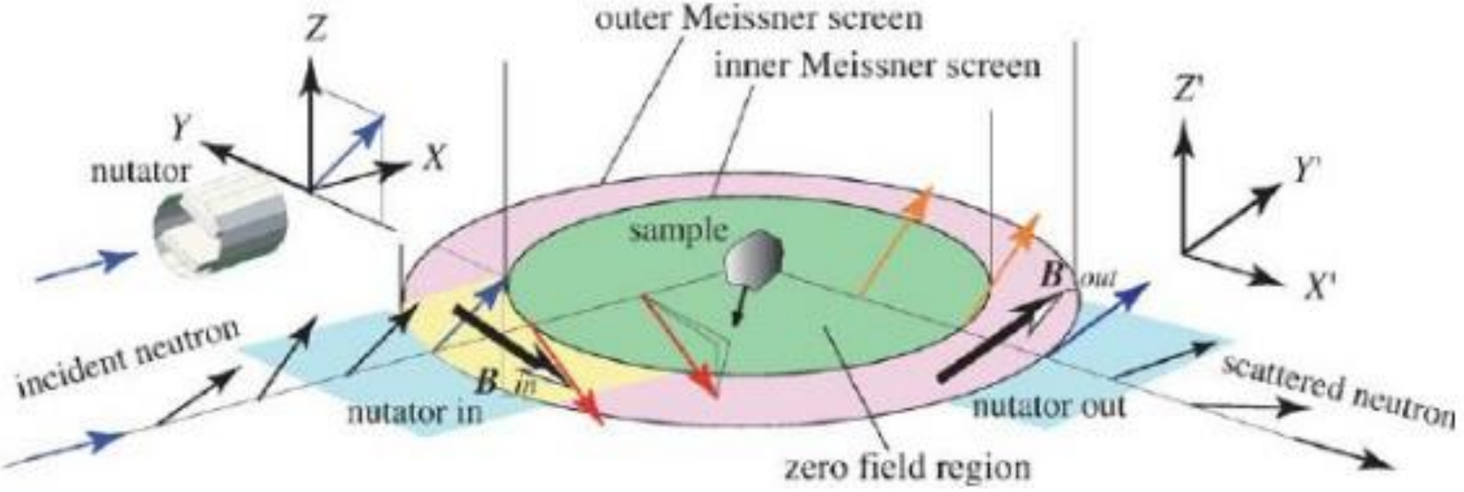
- Time dependence of ^{3}He polarization decay should be accounted for:

$$\mu\wp_{^3\text{He}}(t_n) = \mu\wp_{^3\text{He}}(t_0)\exp\left[(t_0 - t_n)/\Gamma\right]$$



Spherical Neutron Polarimetry (in comparison to 1D polarization analysis)

- Zero-applied magnetic field at sample
- Neutron free to rotate and is not constrained projections along +/- B
- Outside of sample region B-fields again define neutron polarization axes
- Up to 9 (or 18) measurement combinations allow detailed measurements of helical and chiral spin structures



CRYOPAD on the triple-axis spectrometer TAS-1 at JAERI

Masayasu Takeda[a,*], Mitsutaka Nakamura[a], Kazuhisa Kakurai[a], Eddy Lelièvre-berna[b], Francis Tasset[b], Louis-Pierre Regnault[c]

Physica B 356 (2005) 136–140

[1] M. Blume, Phys. Rev. 130, 1670 (1963)

[2] S.V. Maleyev et al., Soviet Phys. Solid State 4, 2533 (1963)

[3] F. Tasset, Physica B 157, 627 (1989)

[4] P.J. Brown, PhysicaB 297, 198 (2001)

Neutron Interactions with Sample Revisited

Strong interaction:

$$V(\vec{\mathbf{x}}) = \frac{2\pi\hbar^2}{m_\mathrm{n}} b\, \delta(\vec{\mathbf{x}} - \vec{\mathbf{R}})$$

Atomic position

Scattering length

Dirac delta function

$$\delta(x) = \begin{cases} \infty, & x = 0 \\ 0, & x \neq 0 \end{cases}$$

Strong scattering is sensitive to the position of atomic nuclei i.e. structure

Electromagnetic interaction:

$$V(\mathbf{x}) = -\vec{\mu}_\mathrm{n} \cdot \vec{\mathbf{B}}(\mathbf{x})$$

Magnetic field

Local magnetization

$$\vec{\mathbf{B}}(\mathbf{x}) = \nabla \times \left[\frac{\mu_0}{4\pi} \int \frac{\vec{\mathbf{M}}(\mathbf{x}') \times (\vec{\mathbf{x}} - \vec{\mathbf{x}}')}{|\vec{\mathbf{x}} - \vec{\mathbf{x}}'|^2} d^3x' \right]$$

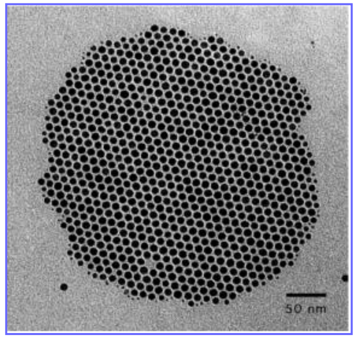
Magnetic scattering is sensitive to local magnetic fields i.e. magnetization

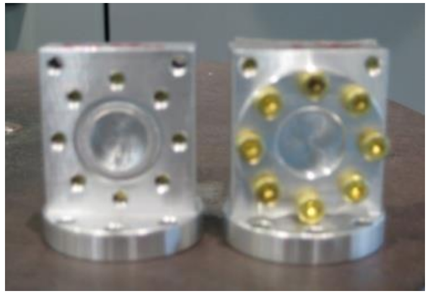
- magnetic SLD, ρ_m = M (in A/m) x 2.853 x 10^{-6} m/(A Å^2) where 1000 A/m = emu/cc
- Electric dipole moment of neutron negligible
- Magnetic moment of interacting nuclei are usually unpolarized
- This can lead to incoherent background scattering (example hydrogen) – 2/3 in spin-flip and 1/3 in non spin-flip channels

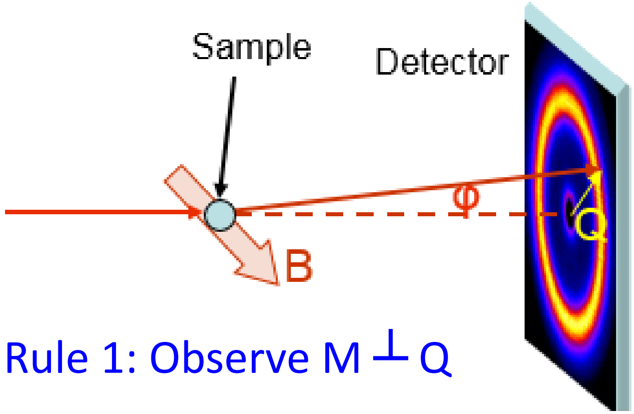
Courtesy of Chris Bertrand, NCNR Summer School 2014

Example to Motivate Polarization Analysis

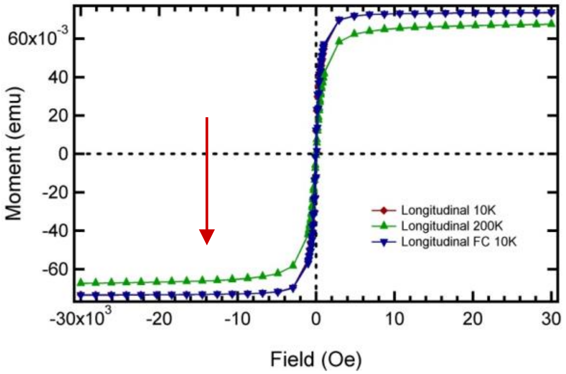
Monodisperse, 9 nm, ferrimagnetic magnetite (Fe_3O_4) particles crystallize into a face-centered cubic crystallites ≈ μm. These crystallites are randomly oriented and form a powder. Magnetite is commonly used due to bio-compatibility stability, and a moment comparable to Ni.



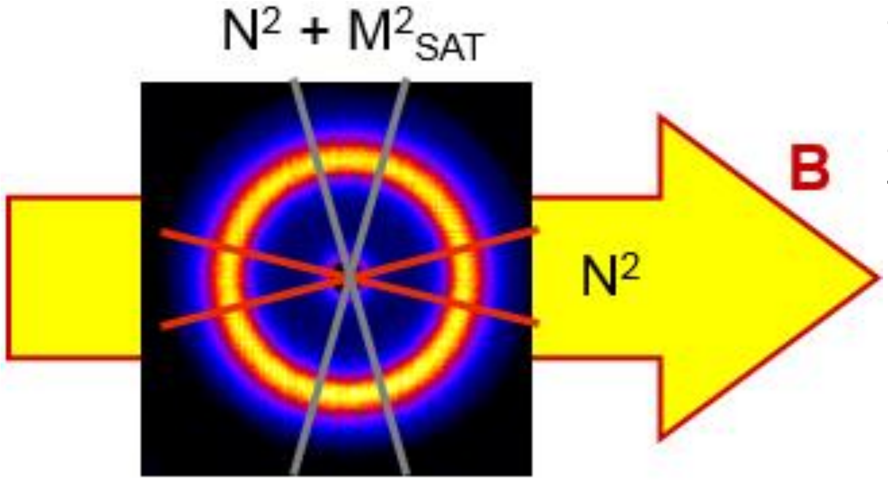




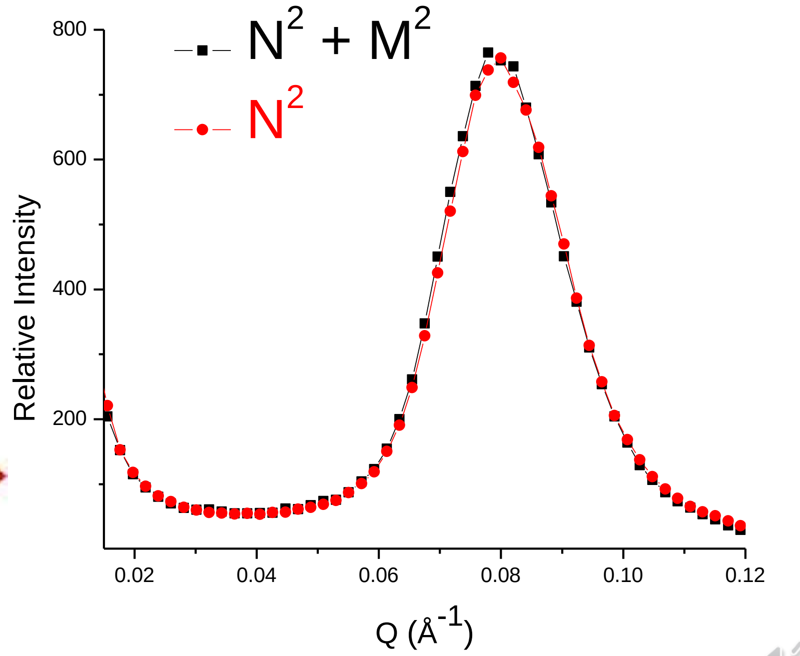
Rule 1: Observe M ⊥ Q



$$N, M_J(Q) = \sum_K \rho_{N,M_J}(K) e^{i\vec{Q}\cdot\vec{R}_K}$$

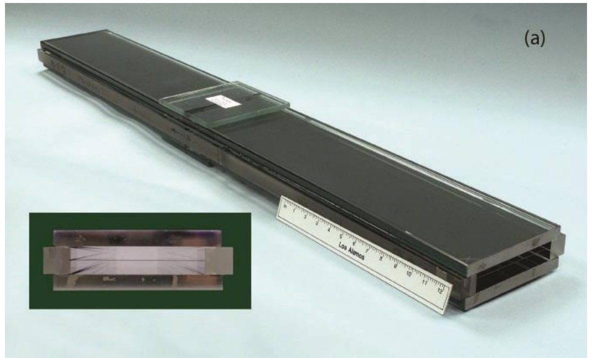


$(\rho_M / \rho_N)^2 = 4\%$



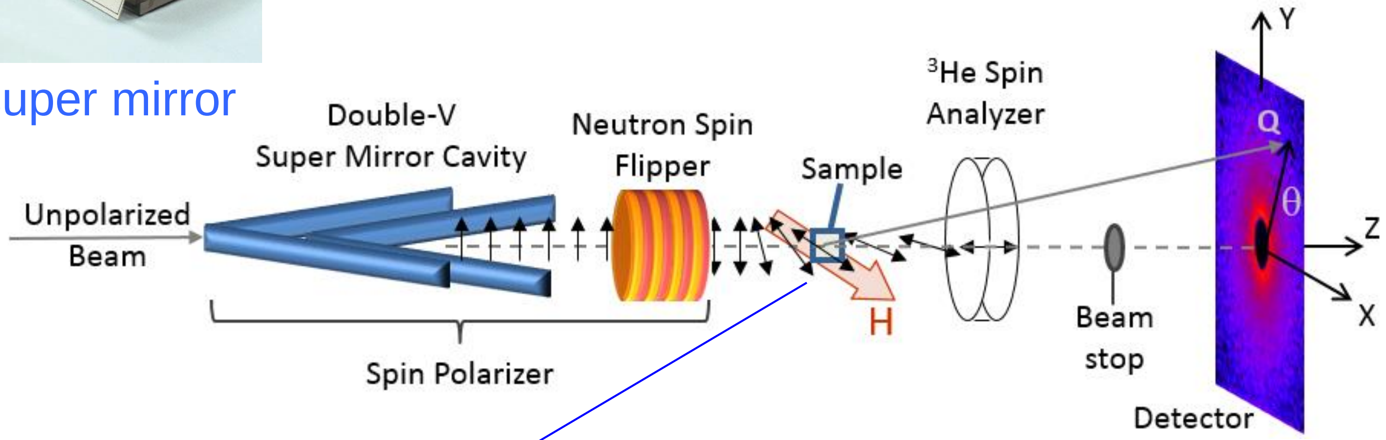
0.08 $Å^{-1}$ peak (111) reflection in 13.6 nm FCC lattice

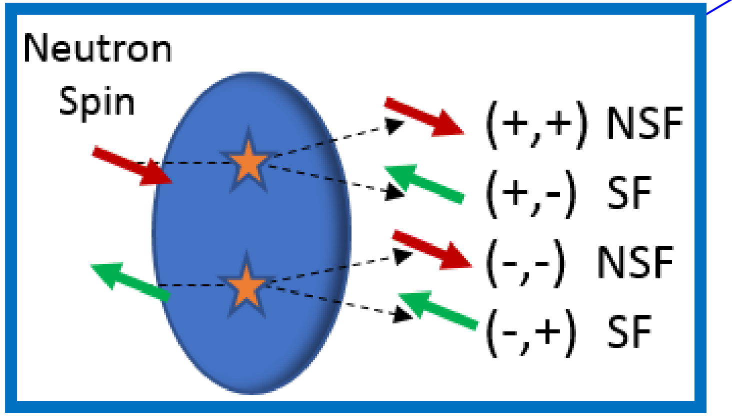
Polarization Analyzed SANS (SANSPOL, POLARIS, PASANS)



FeSi super mirror

^{3}He neutron spin analyzer [W.C. Chen et al., Physica B, 404, 2663 (2009)]





Non spin-flip (NSF) vs. Spin-flip (SF) scattering

NSF → all structural scattering (N)
→ projection of (M ⊥ Q) that is || H

SF → the projection of (M ⊥ Q) that is ⊥ H

Thus, spin-flip is entirely magnetic!

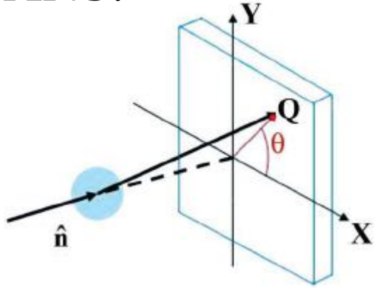
Rules of 1D Polarization (polarization axis, **p**, defined by B)

- Rule 1: Only the component of the magnetic moment (or magnetic form factor), M, that is ⊥ Q may participate in neutron scattering. This is embodied in the Halpern-Johnson vector (Phys. Rev. 55, 898 (1939)) as:

$$\Upsilon(\hat{\mathbf{Q}}) = \mathbf{M} - (\hat{\mathbf{Q}} \cdot \mathbf{M})\,\hat{\mathbf{Q}} = |\mathbf{M}|\,[\hat{\mathbf{M}} - (\hat{\mathbf{Q}} \cdot \hat{\mathbf{M}})\,\hat{\mathbf{Q}}]$$

- Often it is conceptually simpler to define M in terms of three orthogonal components labeled A, B, and C, where A || **p** and B x C = A. ω is the angle between axes, which can be recast in terms of θ for SANS:

$$\Upsilon_{J=A,B,C}(\hat{\mathbf{Q}}) = \sum_{L=A,B,C} M_L[\cos(\omega_{L,J}) - \cos(\omega_{\mathbf{Q},J})\cos(\omega_{\mathbf{Q},L})]$$

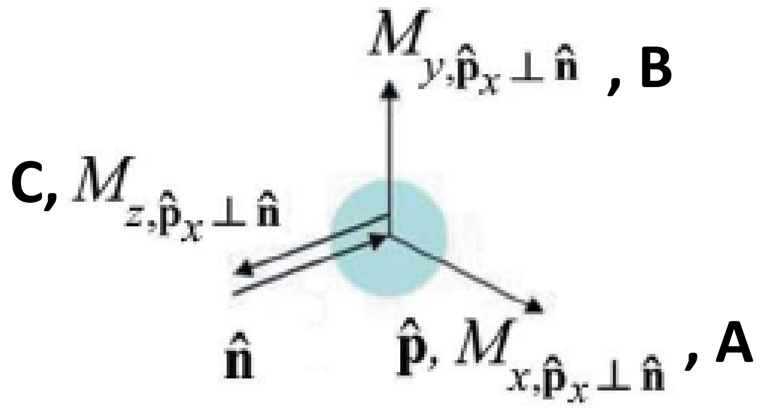


- Rule 2: Of M ⊥ Q (defined by ϒ), the portion || **p** contributes to non-spin flip, while the portion ⊥ **p** contributes to spin-flip (Moon, Riste, Koehler, Phys. Rev. 181, 920 (1969)). Note we are here neglecting any nuclear magnetic scattering, which is often unpolarized and negligible. A common exception is incoherent H-scattering, which shows up as a flat background with 2/3 of the scattering in the spin-flip channel and 1/3 in the non-spin-flip channel.

$$\sigma^{\downarrow\downarrow}_{\uparrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}\left|N \pm \Upsilon_A\right|^2, \quad \sigma^{\uparrow\downarrow}_{\downarrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}|(-\Upsilon_B \mp i\Upsilon_C)|^2$$

Specifics for **p** ⊥ n-beam

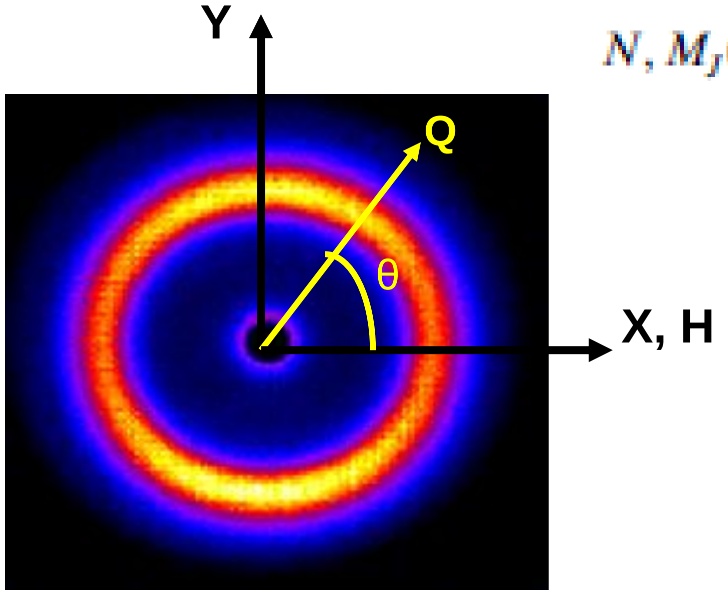
$$\Upsilon_A(\mathbf{Q}) = M_A \sin^2(\theta) - M_B \sin(\theta)\cos(\theta)$$
$$\Upsilon_B(\mathbf{Q}) = M_B \cos^2(\theta) - M_A \sin(\theta)\cos(\theta)$$
$$\Upsilon_C(\mathbf{Q}) = M_C$$

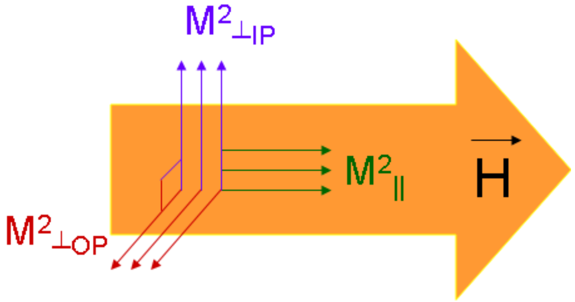


$$\begin{aligned}
\sigma^{\overset{\downarrow\downarrow}{\uparrow\uparrow}}_{\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) &= N(\mathbf{Q})N^*(\mathbf{Q}) + M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\sin^4(\theta) \\
&+ M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\cos^2(\theta)\sin^2(\theta) \\
&- [M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
&+ M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin^3(\theta)\cos(\theta) \\
&\pm [N(\mathbf{Q})M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) + N^*(\mathbf{Q})M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin^2(\theta) \\
&\mp [N(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) + N^*(\mathbf{Q})M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin(\theta)\cos(\theta)
\end{aligned}$$

$$\begin{aligned}
\sigma^{\overset{\uparrow\downarrow}{\downarrow\uparrow}}_{\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) &= M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
&+ M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\cos^4(\theta) \\
&+ M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\sin^2(\theta)\cos^2(\theta) \\
&- [M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
&+ M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin(\theta)\cos^3(\theta) \\
&\pm i[M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
&- M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin(\theta)\cos(\theta) \\
&\mp i[M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) - M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\cos^2(\theta)
\end{aligned}$$

Specifics for p ⊥ n-beam





$$N, M_J(\mathbf{Q}) = |N, M_J| \exp(i\varphi_{N,M_J}) \;\Longrightarrow\; \alpha\alpha^* = |\alpha|^2,$$

$$\alpha\beta^* + \alpha^*\beta = 2|\alpha|\,|\beta|\,\overline{\cos}(\varphi_\alpha - \varphi_\beta),$$

$$i(\alpha\beta^* - \alpha^*\beta) = -2|\alpha|\,|\beta|\,\overline{\sin}(\varphi_\alpha - \varphi_\beta)$$

$$I^{--,++} = |N|^2 + \sin^2(\theta)\cos^2(\theta)|M_{\perp ip}|^2 + \sin^4(\theta)|M_{||}|^2$$

$$-2\cos(\theta)\sin^3(\theta)|M_{||}||M_{\perp ip}|\cos(\varphi_{||}-\varphi_{\perp ip})$$

$$\pm 2\sin(\theta)\cos(\theta)|N||M_{\perp ip}|\cos(\varphi_N-\varphi_{\perp ip})$$

$$\mp 2\sin^2(\theta)|N||M_{||}|\cos(\varphi_N-\varphi_{||})$$

$$I^{+-,-+} = |M_{\perp op}|^2 + \cos^4(\theta)|M_{\perp ip}|^2 + \sin^2(\theta)\cos^2(\theta)|M_{||}|^2$$

$$-2\sin(\theta)\cos^3(\theta)|M_{||}||M_{\perp ip}|\cos(\varphi_{||}-\varphi_{\perp ip})$$

$$\pm 2\sin(\theta)\cos(\theta)|M_{||}||M_{\perp op}|\sin(\varphi_{||}-\varphi_{\perp op})$$

$$\mp 2\cos^2(\theta)|M_{\perp ip}||M_{\perp op}|\sin(\varphi_{\perp op}-\varphi_{\perp ip})$$

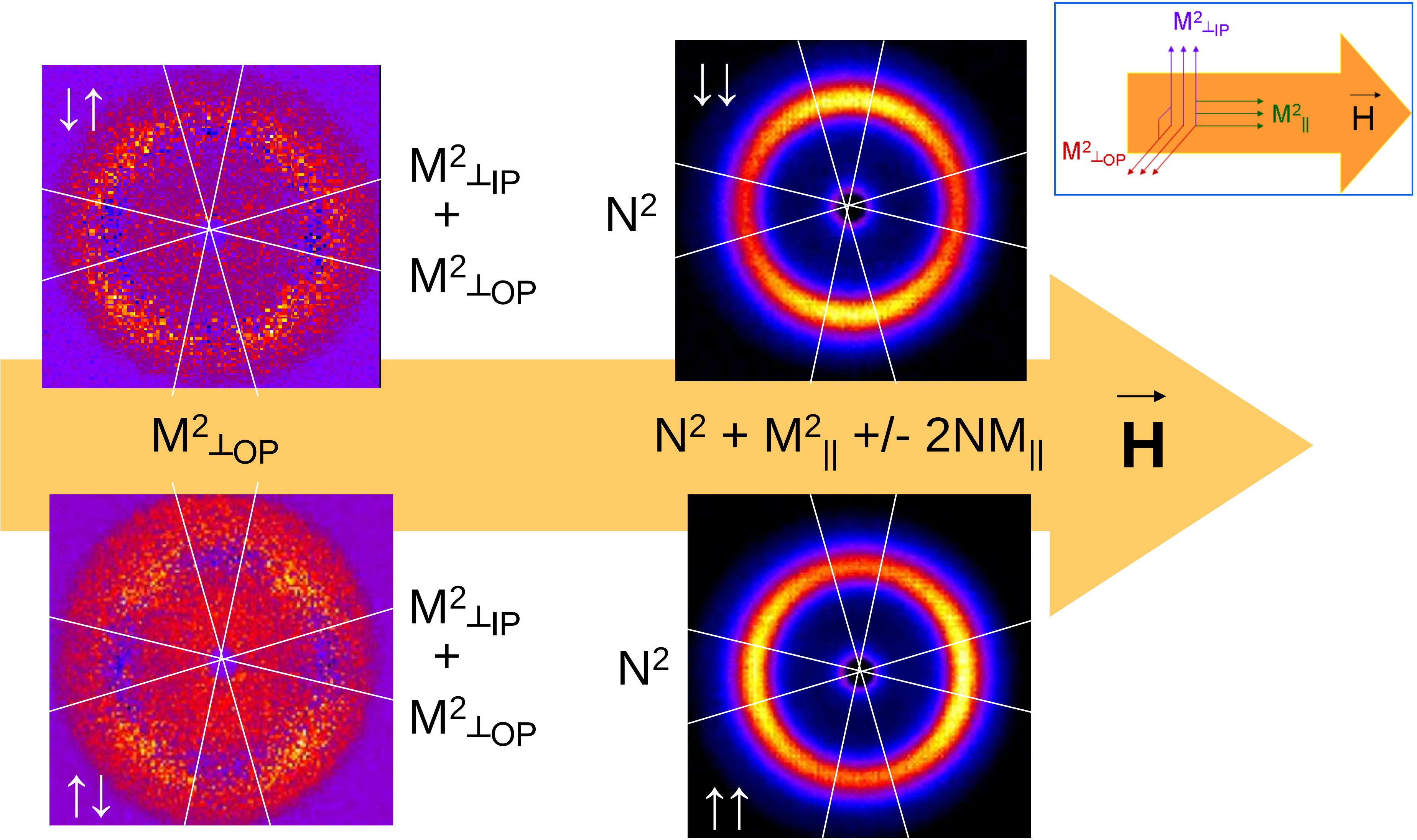
R. M. Moon, T. Riste, and W. C. Koehler, Physical. Review 181, 920 (1969)

A. Wiedenmann *et al*., Physica B 356, 246 (2005)

A. Michels and J. Weissmüller, Rep. Prog. Phys. 71, 066501 (2008)

K. Krycka *et al*., J. Appl. Cryst. 45, 554 (2012)

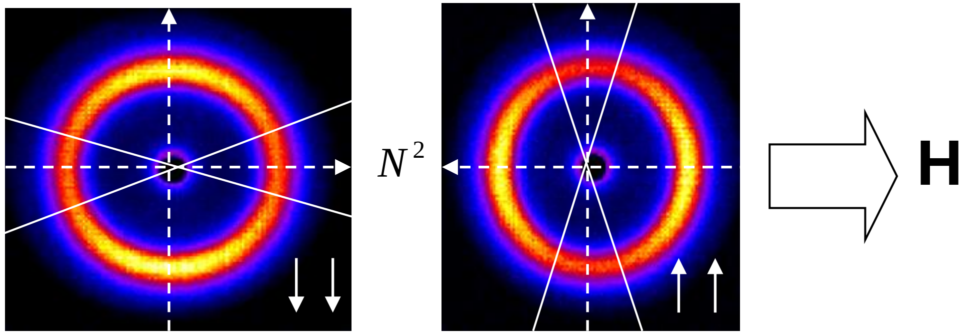
Coordinate Axes Simplification (p ⊥ n)

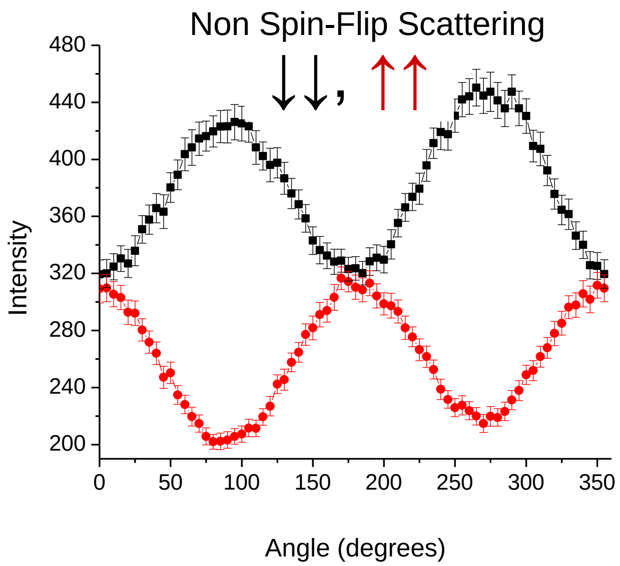


If sample is structurally isotropic, we can determine $M^2_{\parallel}$

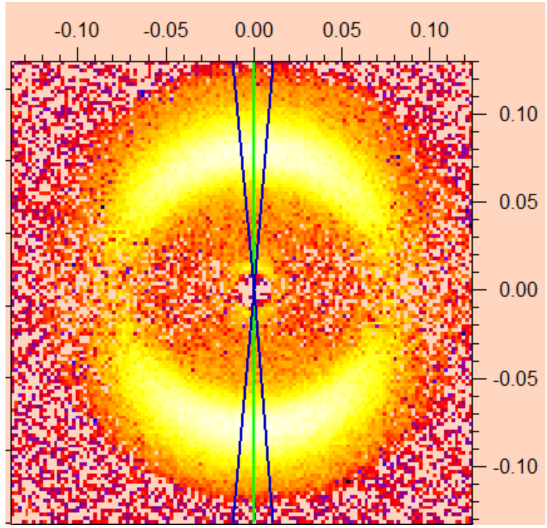
Non Spin-Flip Scattering at 1.2 Tesla, 200 K

$N^2 + M_{\parallel}^2 + 2NM_{\parallel}$ $\qquad$ $N^2 + M_{\parallel}^2 - 2NM_{\parallel}$





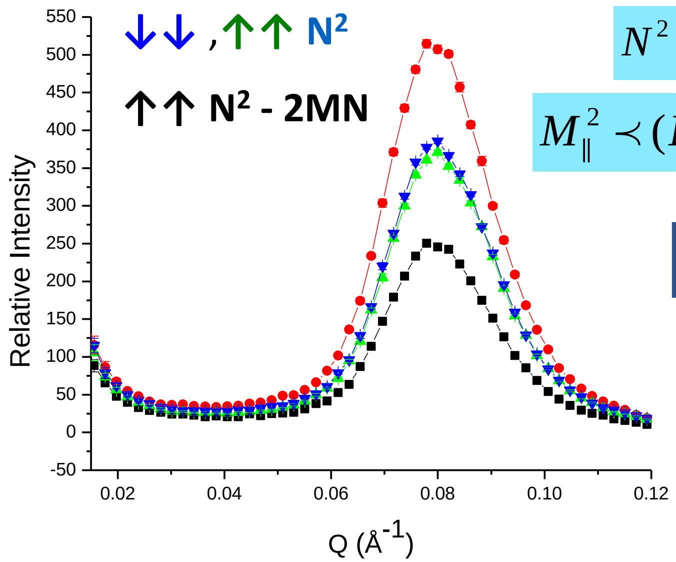
↓↓ - ↑↑ Scattering



↓↓ N^2 + 2MN

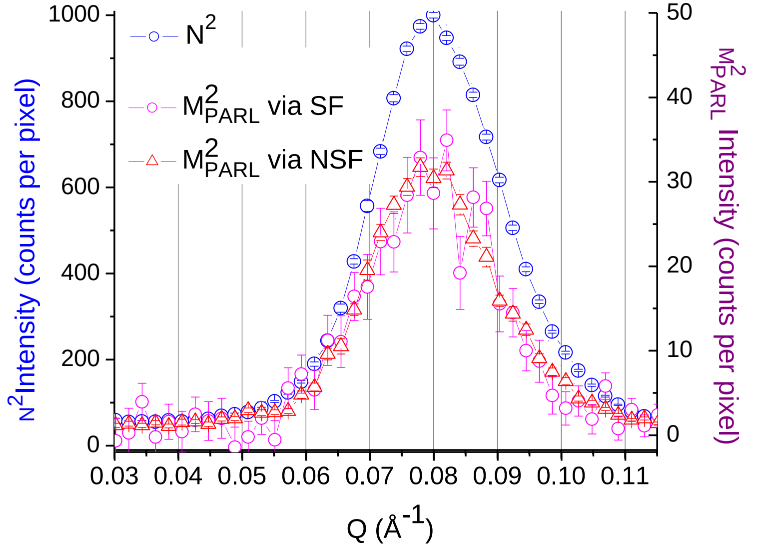
↓↓ ,↑↑ N^2

↑↑ N^2 - 2MN



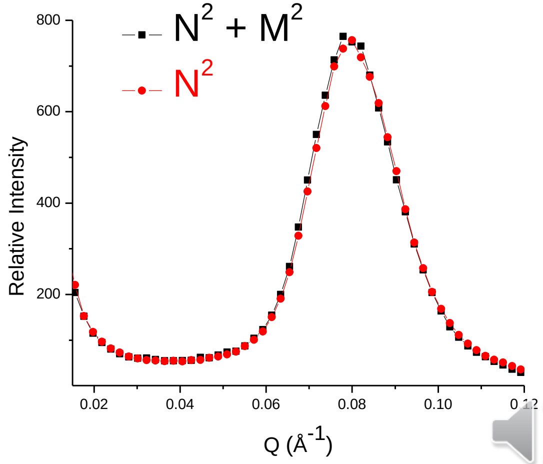
$$N^2 \prec (I_X^{\uparrow\uparrow} + I_X^{\downarrow\downarrow})$$

$$M_{\parallel}^2 \prec (I_Y^{\downarrow\downarrow} - I_Y^{\downarrow\downarrow})^2 / 8N^2$$



vs.

Unpolarized



M || B from Spin-Flip Scattering at 1.2 Tesla, 200 K

$$I^{+-,-+} = |M_{\perp op}|^2 + cos^4(\theta)|M_{\perp ip}|^2 + sin^2(\theta)cos^2(\theta)|M_{||}|^2$$

$$-2sin(\theta)cos^3(\theta)|M_{||}||M_{\perp ip}|cos(\varphi_{||}-\varphi_{\perp ip})$$

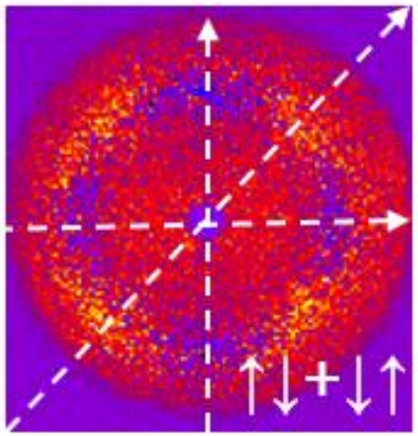
$$\pm 2sin(\theta)cos(\theta)|M_{||}||M_{\perp op}|sin(\varphi_{||}-\varphi_{\perp op})$$

$$\mp 2cos^2(\theta)|M_{\perp ip}||M_{\perp op}|sin(\varphi_{\perp op}-\varphi_{\perp ip})$$

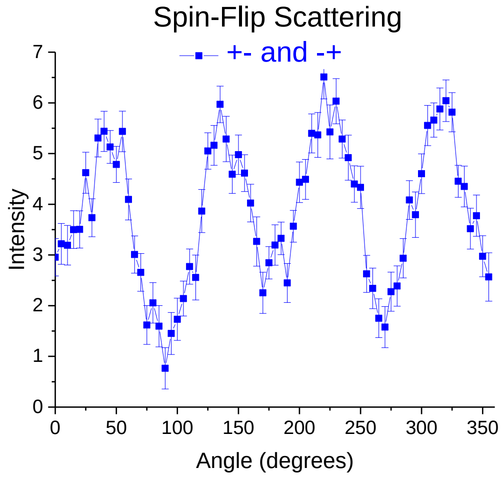
$$M^2_{PARL} = I^{\uparrow\downarrow,\downarrow\uparrow}_{45^o} - 1.25M^2_{PERP}$$

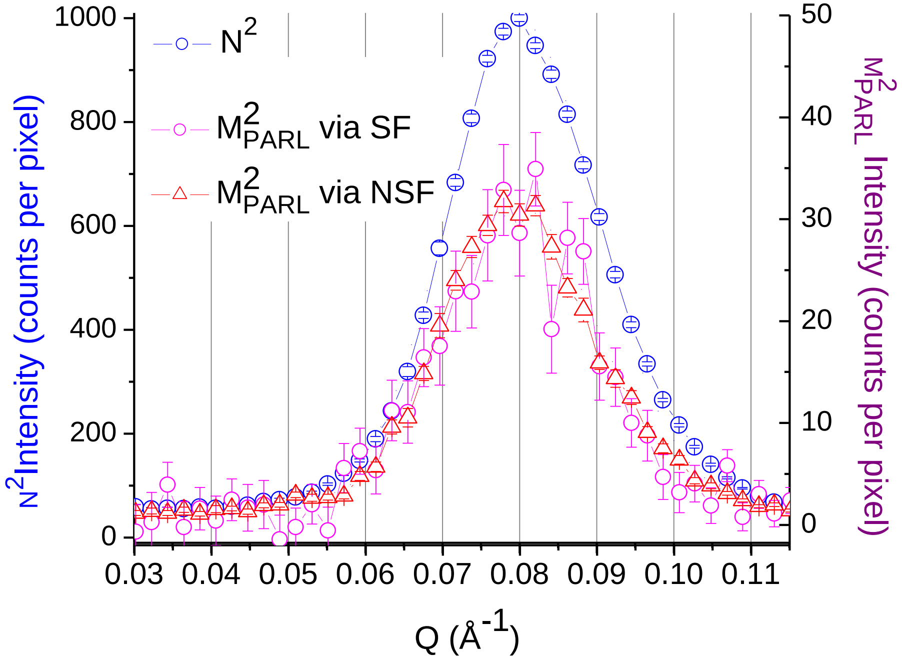
$$M^2_{PERP} = (I^{\uparrow\downarrow,\downarrow\uparrow}_X + I^{\uparrow\downarrow,\downarrow\uparrow}_Y)/3$$

$M^2_{\perp OP}$ $M^2_{||} + 1.25(M^2_{\perp IP} + M^2_{\perp OP})$



$M^2_{\perp IP} + M^2_{\perp OP}$



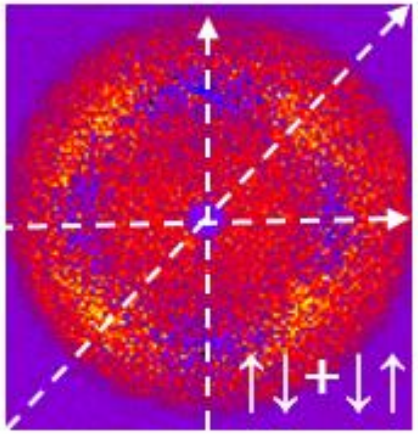


Note Magnetic / Nuclear ~ 0.03

M ⊥ B from Spin-Flip Scattering at 1.2 Tesla, 200 K

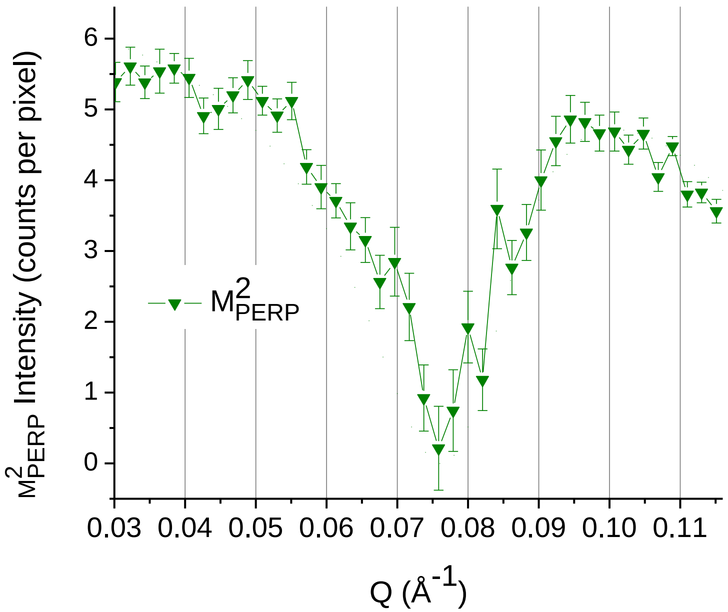
$$M^2_{PERP} = (I_X^{\uparrow\downarrow,\downarrow\uparrow} + I_Y^{\uparrow\downarrow,\downarrow\uparrow})/3$$

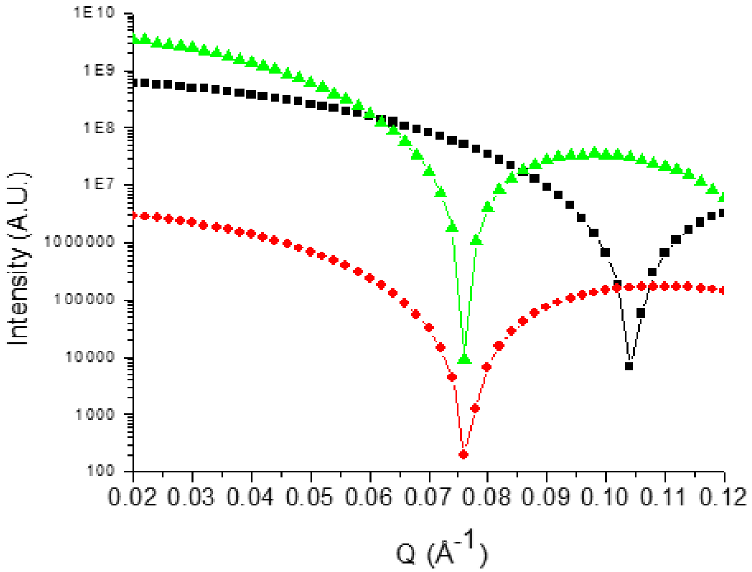
$M^2_{\perp OP}$ $\quad$ $M^2_{\parallel} + 1.25(M^2_{\perp IP} + M^2_{\perp OP})$

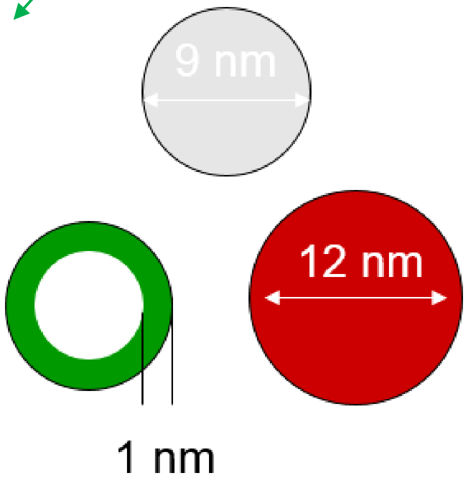


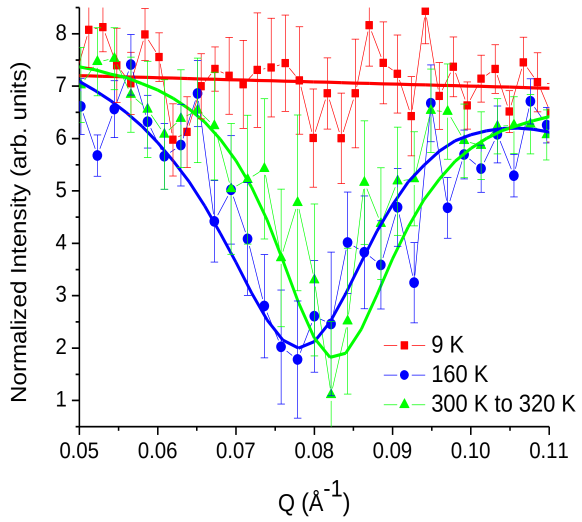
$M^2_{\perp IP} + M^2_{\perp OP}$

Dip at 0.075 Å^{-1} is reminiscent of spherical scattering



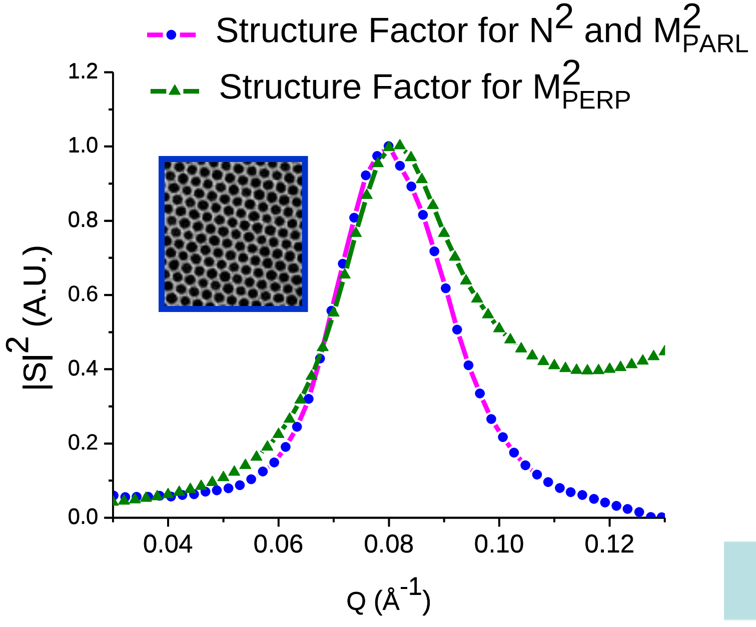






Canted magnetic shell changes with temperature

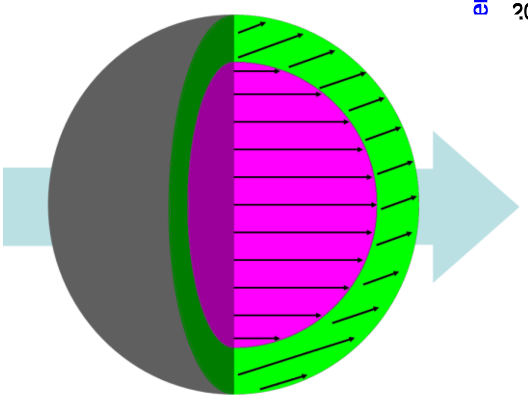
Putting It Together

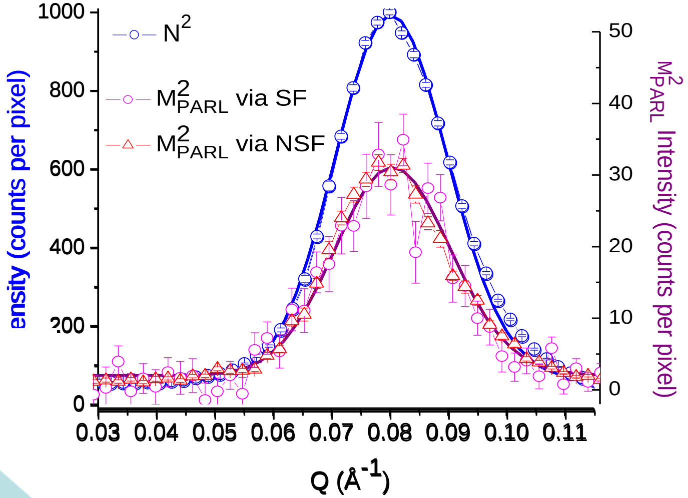


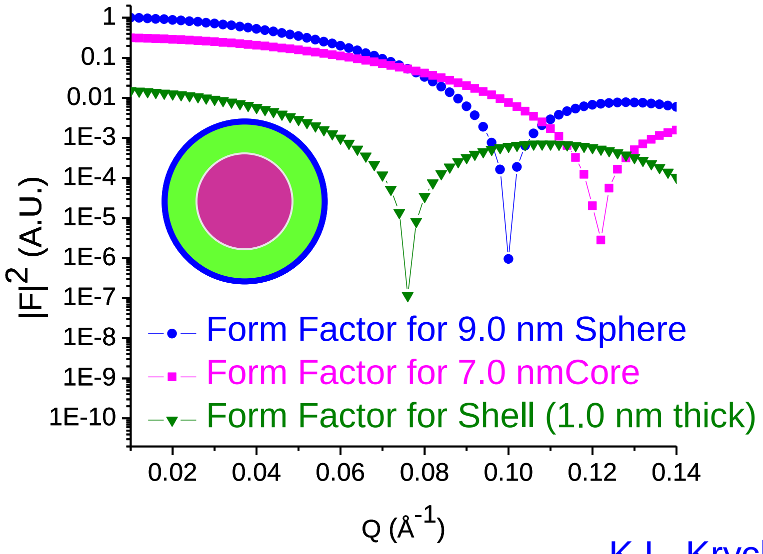
ρ_N **= 6.97E-6 Å^{-2}**

ρ_M **= 1.46E-6 Å^{-2}**

(513 emu / cc)





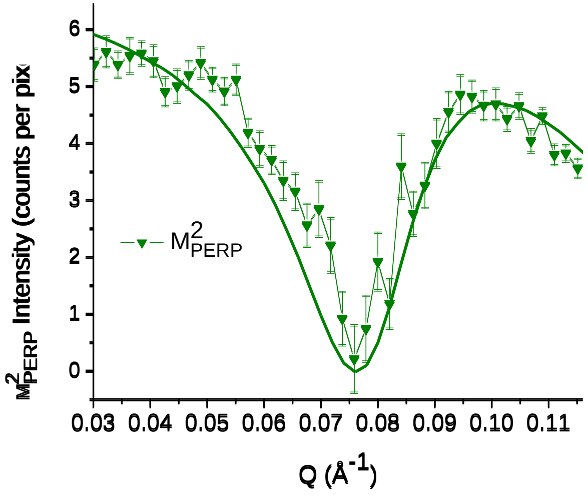


Modeled Diameters:

Sphere 9.0 nm

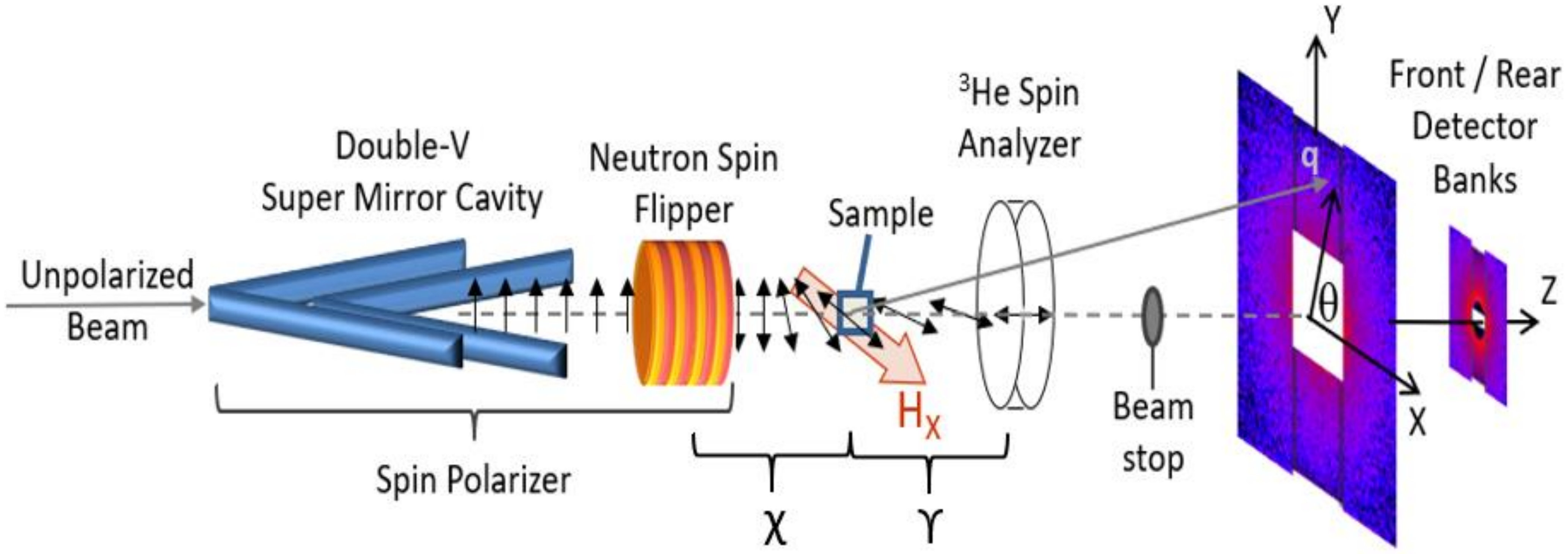
Ferrimagnetic core 7 nm

Canted shell 7 to 9 nm (±0.2 nm)



K.L. Krycka et al., *Phys. Rev. Lett.* 104 207203 (2010)

Neutron Beam and Sample Depolarization

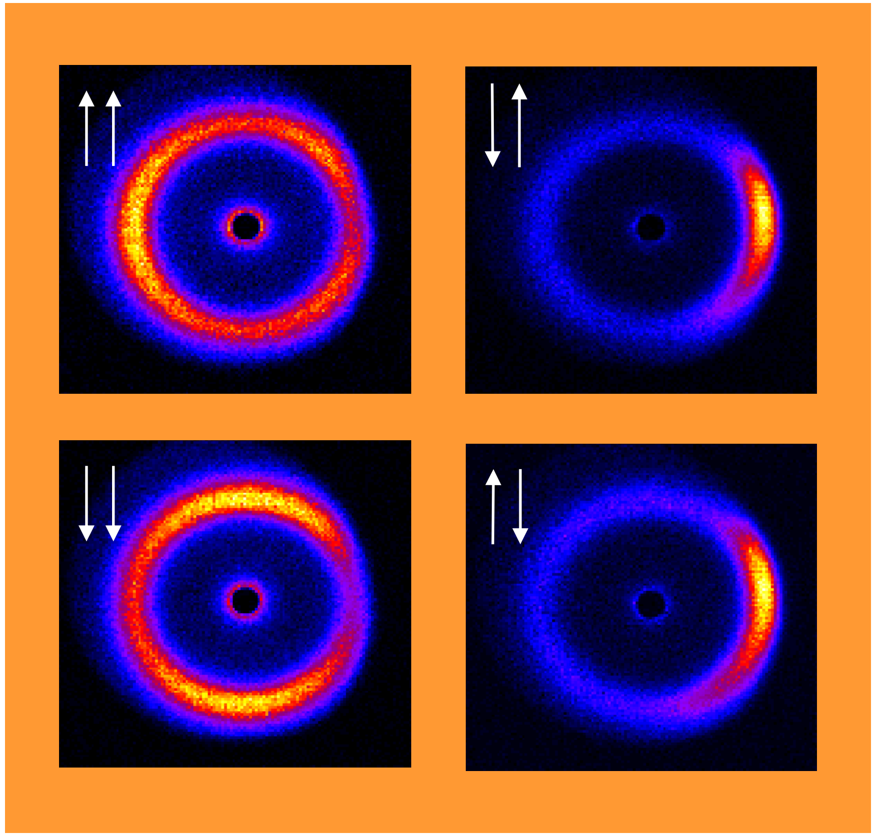


1. Make sure off-axis (beam-path) depolarization is minimized
2. Measure and correct for depolarization contributions using transmission measurements

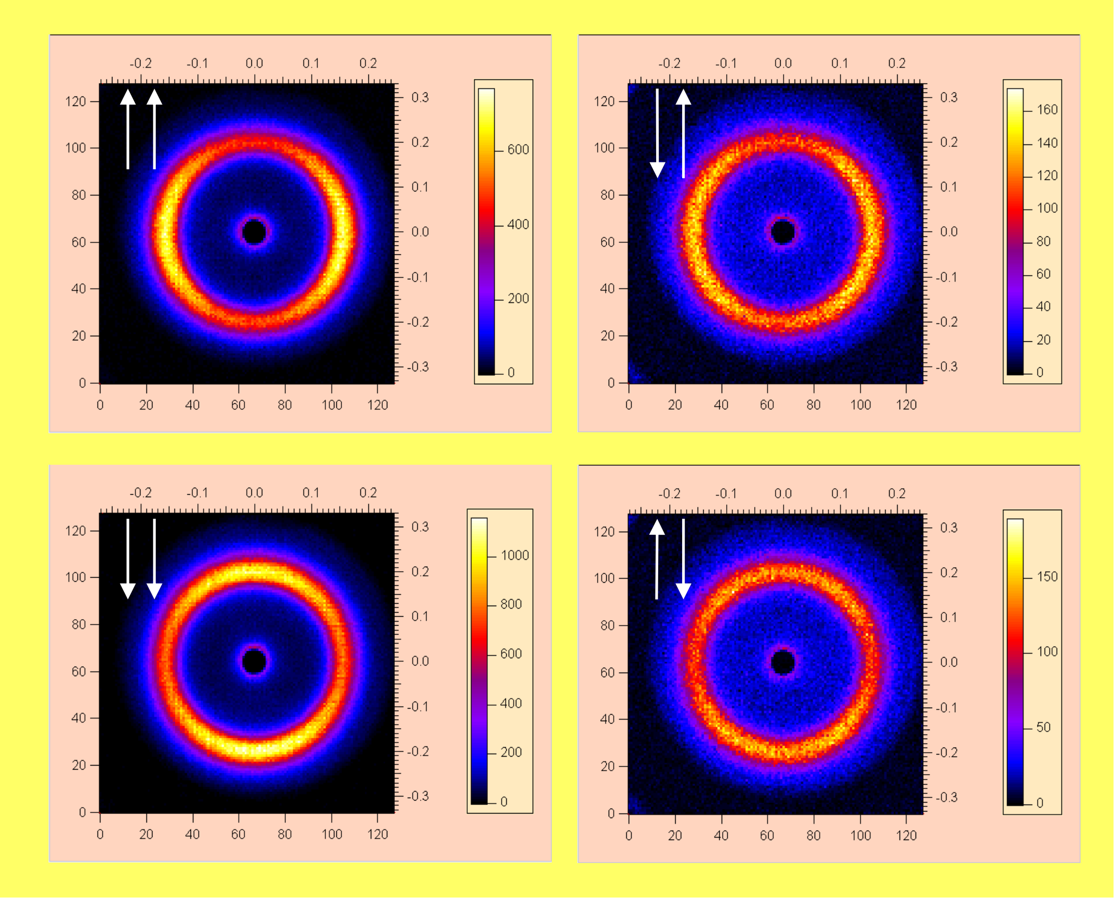
Note that Χ and ϒ encompass both beam-path and sample depolarization

Off-Axis Beam De-Polarization

- Although transmission captures depolarization up to and through the sample, depolarization of a *divergent beam* requires a visual inspection:

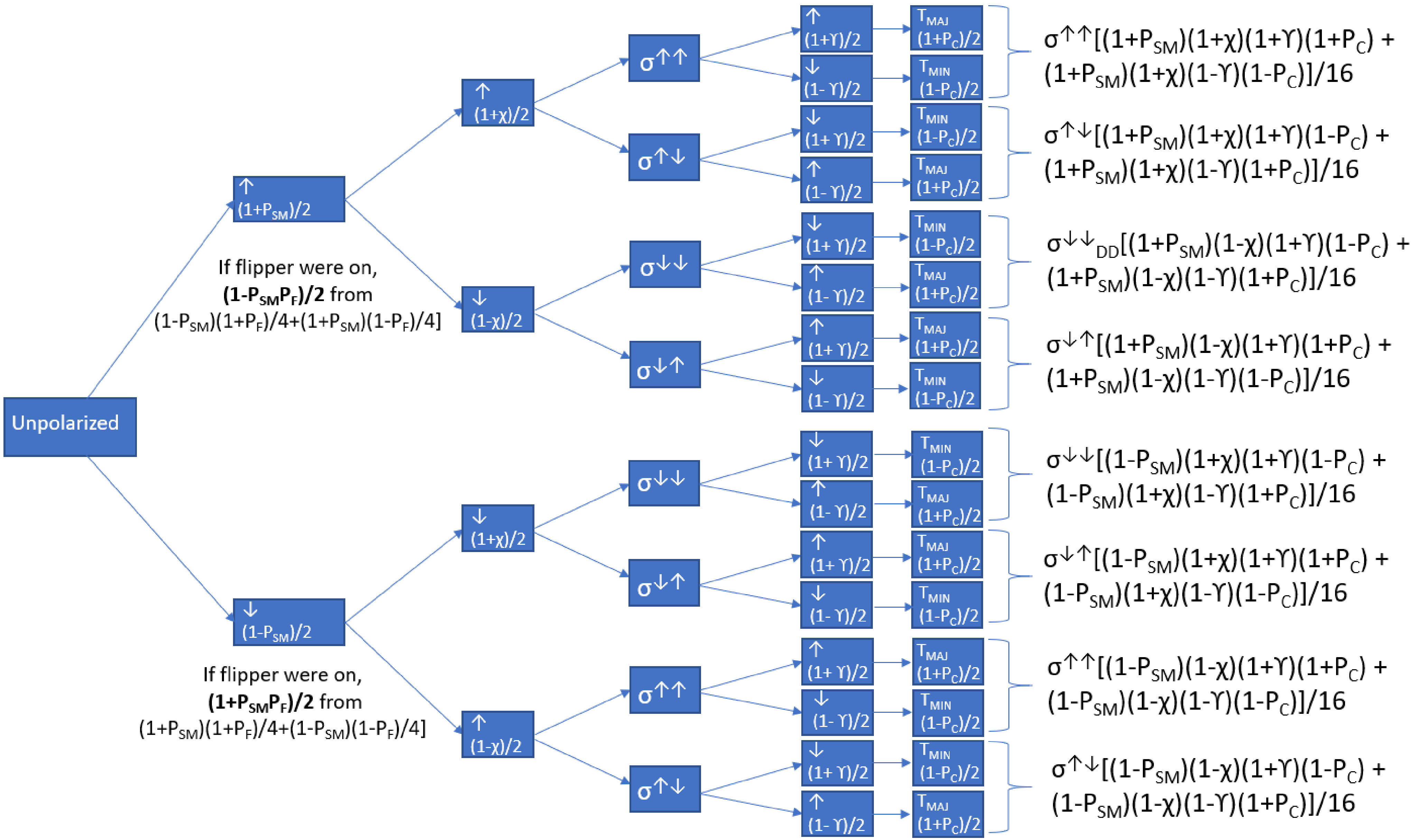


versus



Diffraction ring example shown where leakage of NSF into SF channel is common sign of depolarization

Spin Cascade (Shown for Majority ↑↑)



And there are 3 other variations to consider…

More Compact Form

$$\frac{1}{4}\underbrace{\begin{bmatrix} (+a+b+c+1) & (-a-b+c+1) & (+a-b-c+1) & (-a+b-c+1) \\ (-a-b+c+1) & (+a+b+c+1) & (-a+b-c+1) & (+a-b-c+1) \\ (+a-b-c+1) & (-a+b-c+1) & (+a+b+c+1) & (-a-b+c+1) \\ (-a+b-c+1) & (+a-b-c+1) & (-a-b+c+1) & (+a+b+c+1) \end{bmatrix}}_{Measured\ Polarization\ Efficiencies} \underbrace{\begin{bmatrix} \sigma^{\uparrow\uparrow} \\ \sigma^{\downarrow\uparrow} \\ \sigma^{\downarrow\downarrow} \\ \sigma^{\uparrow\downarrow} \end{bmatrix}} = \underbrace{\begin{bmatrix} S_{\uparrow\uparrow} \\ S_{\downarrow\uparrow} \\ S_{\downarrow\downarrow} \\ S_{\uparrow\downarrow} \end{bmatrix}}$$

Cross-sections — Measured scattering

$$a \equiv P_{SM} P_F P_\chi P_\Upsilon P_C; \quad b \equiv P_{SM} P_F P_\chi; \quad c \equiv P_\Upsilon P_C$$

$$\text{In transmission, } \sigma^{\uparrow\uparrow} = \sigma^{\downarrow\downarrow} = 1 \text{ and } \sigma^{\downarrow\uparrow} = \sigma^{\uparrow\downarrow} = 0.$$

The goal is to determine a, b, and c from transmissions, then multiply both sides by the inverse of the Measured Polarization Efficiencies matrix. That allows the true cross-sections (σ) to be determined from the four measured scattering patterns (S).

Solving Matrix Polarization Coefficients

In transmission, $\sigma^{\uparrow\uparrow} = \sigma^{\downarrow\downarrow} = 1$ and $\sigma^{\downarrow\uparrow} = \sigma^{\uparrow\downarrow} = 0$.

$$P_\chi P_\Upsilon P_{SM} = (2T_{\uparrow\uparrow}/T_{Unpol} - 1)/P_C$$

$$P_\chi P_\Upsilon P_{SM} = (1 - 2T_{\uparrow\downarrow)}/T_{Unpol})/P_C$$

$$P_\chi P_\Upsilon P_{SM} P_F = (2T_{\downarrow\downarrow}/T_{Unpol} - 1)/P_C$$

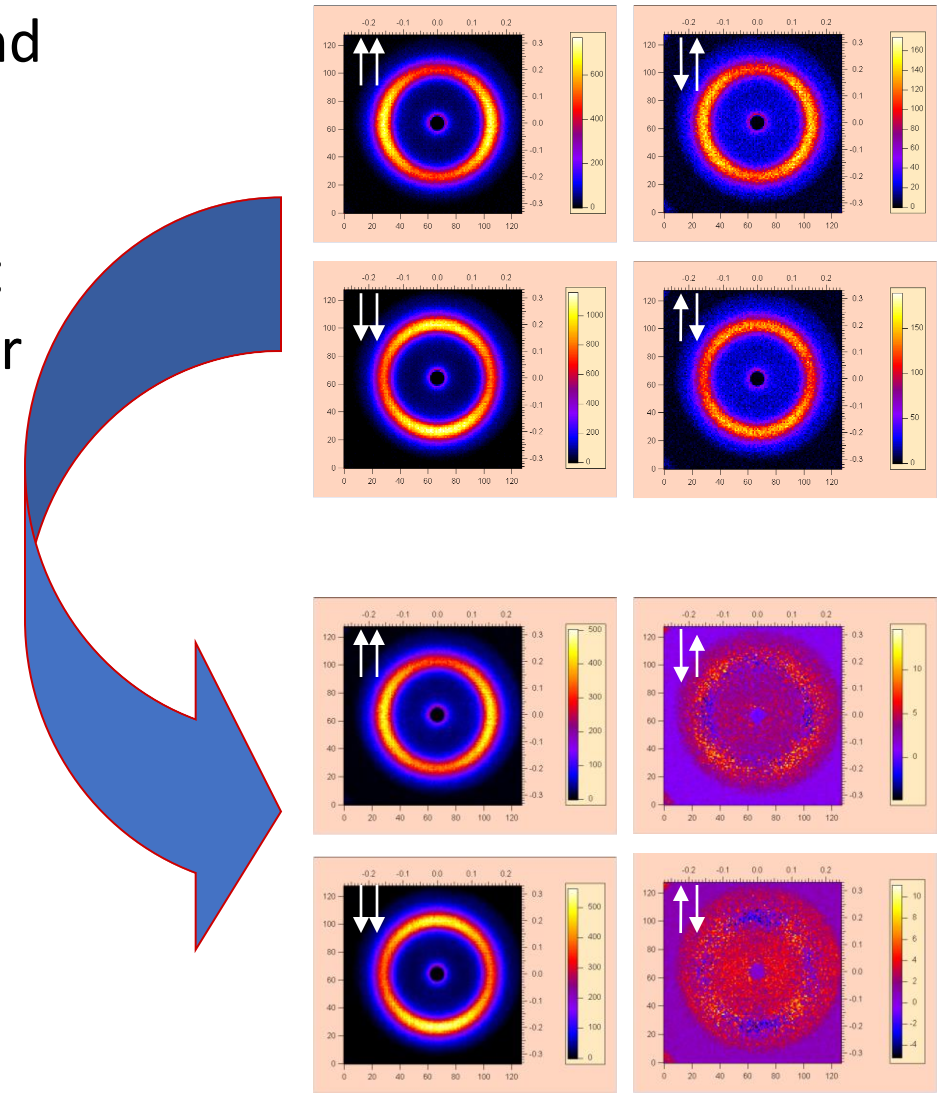
$$P_\chi P_\Upsilon P_{SM} P_F = (1 - 2T_{\downarrow\uparrow}/T_{Unpol})/P_C$$

The polarizations of the super mirror, χ, and ϒ are difficult to separate. Often P_{SM} can be measured on an empty sample holder, and we set χ = ϒ.

$$P_F = [(2T_{\downarrow\downarrow}/T_{Unpol} - 1)/P_C]/[(2T_{\uparrow\uparrow}/T_{Unpol} - 1)/P_C]$$

Polarization Efficiency Correction Effect

- Spin leakage from supermirror, polarizer, and ^{3}He analyzer (time-dependent) are all important
- Sample itself can be depolarizing, and must be measured and corrected for if looking for small signals
- Typically corrections are most relevant for spin-flip scattering
- Multiple scattering (around a Bragg peak) can be difficult to properly polarization correct
- Too much wavelength spread (say > 15%) can also cause issues around sharp scattering features



Magnetic Correlations and Sample Depolarization

Directional depolarization

Magnetic coherence length (which may be < domain size)

$$D_{ii} = e^{-(\gamma^2/v^2)L\{\xi - \alpha_{ii}\}} \qquad i = x, y, z$$

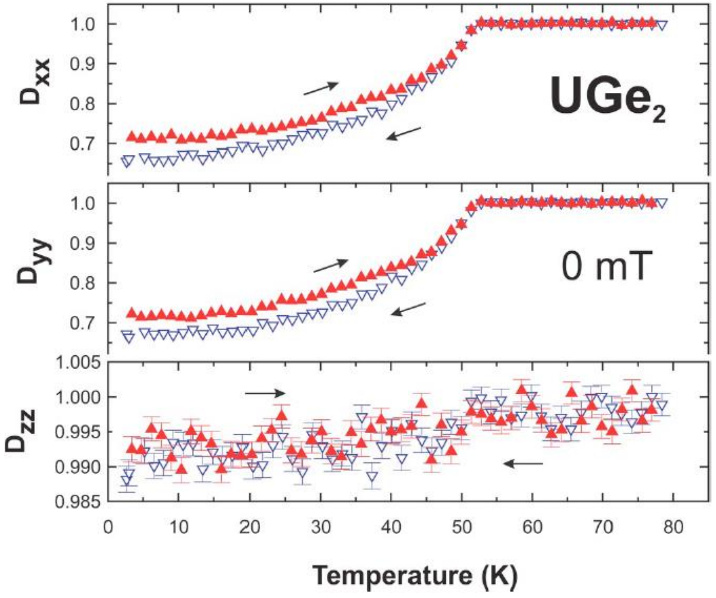
Assume → 0

Sample length

Gyromagnetic ratio of neutron / velocity

S. Sakarya, N. H. van Dijk, E. Brück

PHYSICAL REVIEW B 71, 174417 (2005)

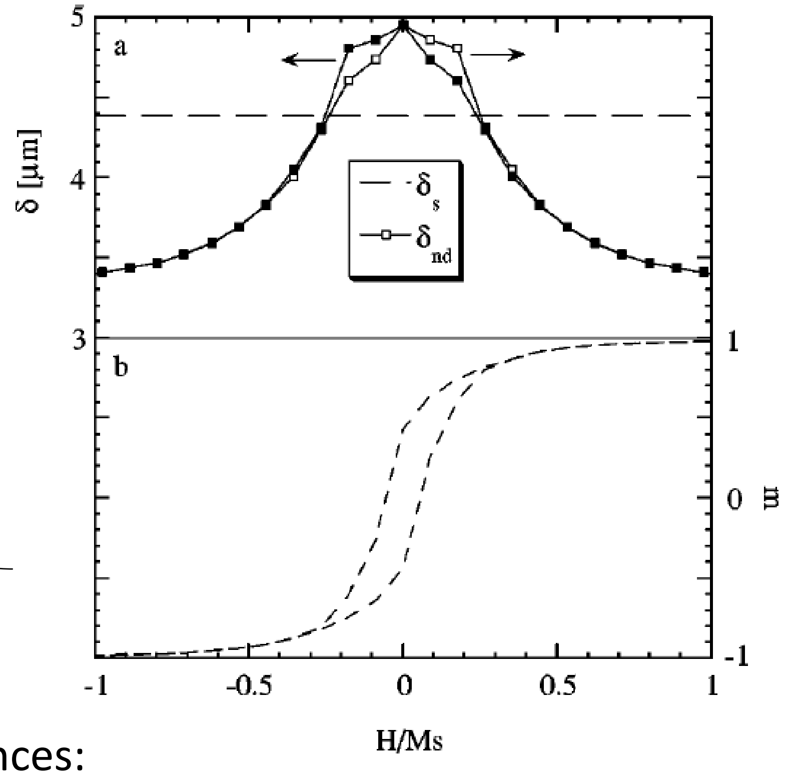


$\mu_0 H$ (mT)	Temp. incr./decr.	$d_\uparrow$ (μm) $T \approx T_C$	$d_\downarrow$ (μm) $T \approx T_C$
0	ZFC, incr.	4.4(1)	4.4(1)
0	FC, decr.	5.1(2)	5.1(2)
4	ZFC, incr.	150(20)	25(5)
4	FC, decr.	60(10)	13(2)
8	ZFC, incr.	85(20)	10(2)
8	FC, decr.	85(20)	10(5)

S. G. E. te Velthuis, et al.

J. Appl. Phys., Vol. 89, No. 2, 15 January 2001

Simulations based on ferromagnetic assembly of spherical domains

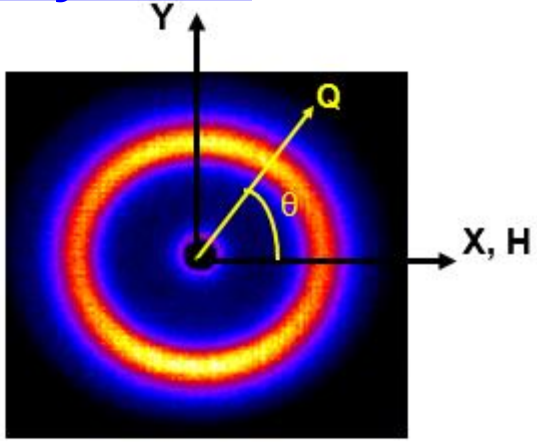


Additional references:

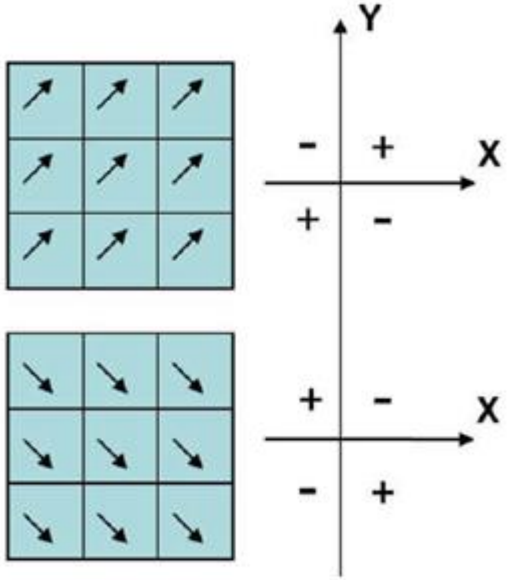
M. Th. Rekveldt, Z. Physics, 259, 391-410 (1973)

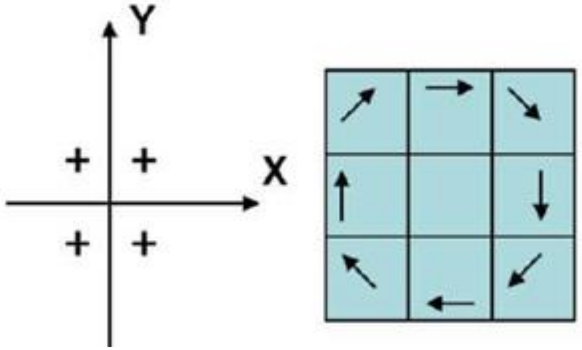
M. Th. Rekveldt, Texures and Microstructures, 11, 127-142 (1989)

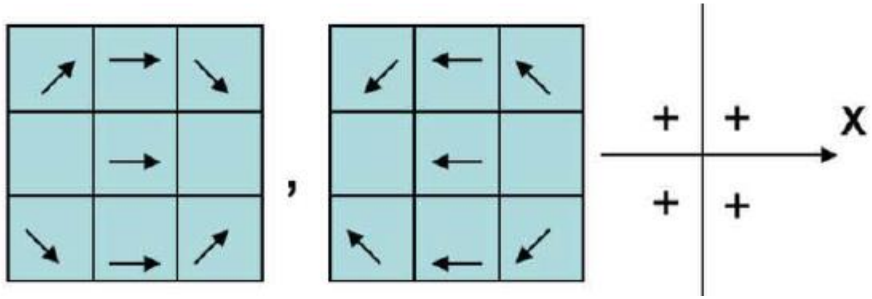
Impact of Cross-Term Depends on Symmetry of System



$$-2\sin(\theta)\cos^3(\theta)|M_{||}||M_{\perp ip}|\cos(\varphi_{||}-\varphi_{\perp ip})$$







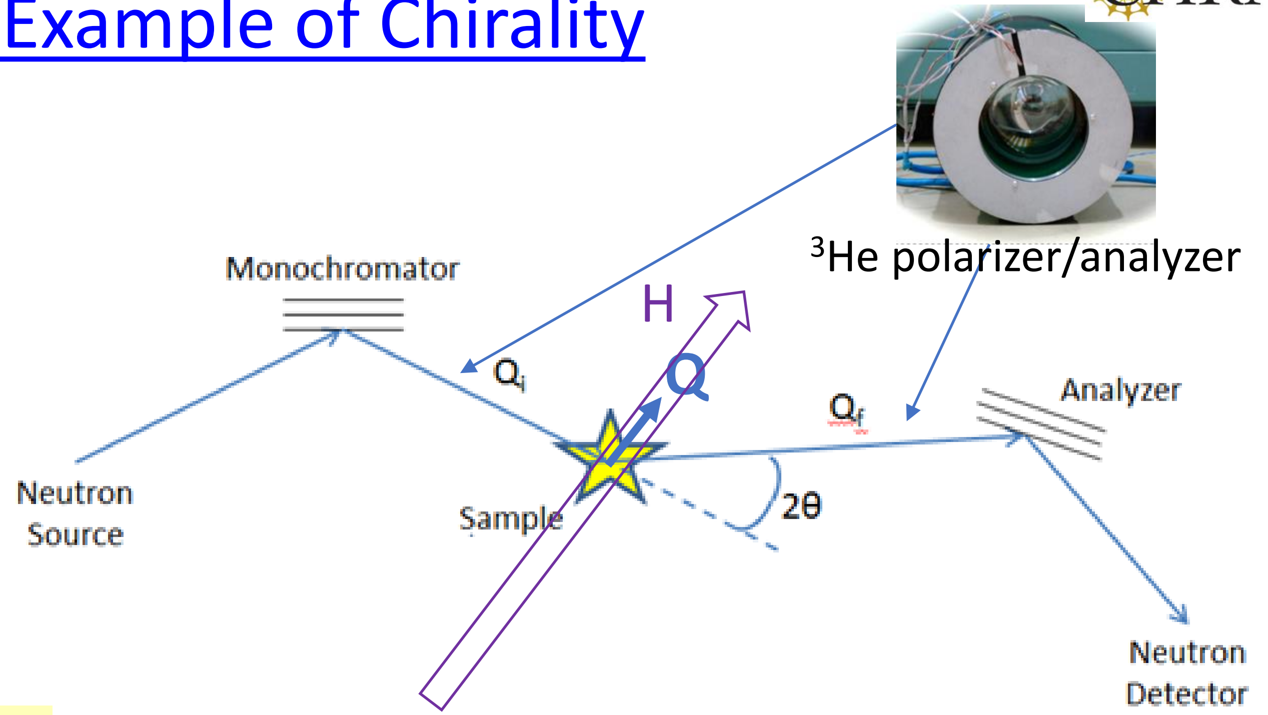
Random canting about field direction shouldn't result in 2:1 spin-Flip deviation

Chiral Systems often results in noticeable cross-terms (and also ↑↓-↓↑differences, too)

Other structures may have non-zero contribution

Diffraction Example of Chirality

$$
\begin{aligned}
\sigma^{\uparrow\downarrow}_{\downarrow\uparrow\,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) = {} & M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
& + M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\cos^4(\theta) \\
& + M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})\sin^2(\theta)\cos^2(\theta) \\
& - [M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
& + M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin(\theta)\cos^3(\theta) \\
& \pm i[M_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) \\
& - M^*_{x,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\sin(\theta)\cos(\theta) \\
& \mp i[M_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M^*_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q}) - M^*_{y,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})M_{z,\hat{\mathbf{p}}_x\perp\hat{\mathbf{n}}}(\mathbf{Q})]\cos^2(\theta).
\end{aligned}
$$

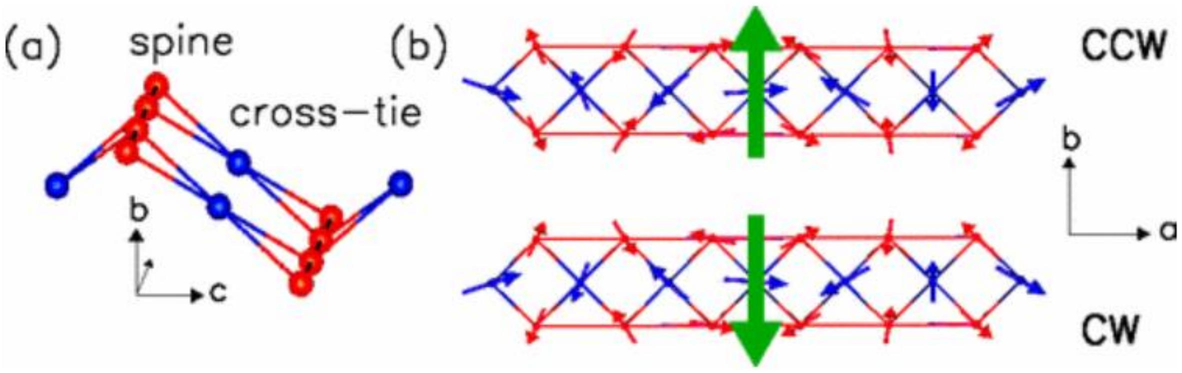


PRL 103, 087201 (2009) PHYSICAL REVIEW LETTERS week ending 21 AUGUST 2009

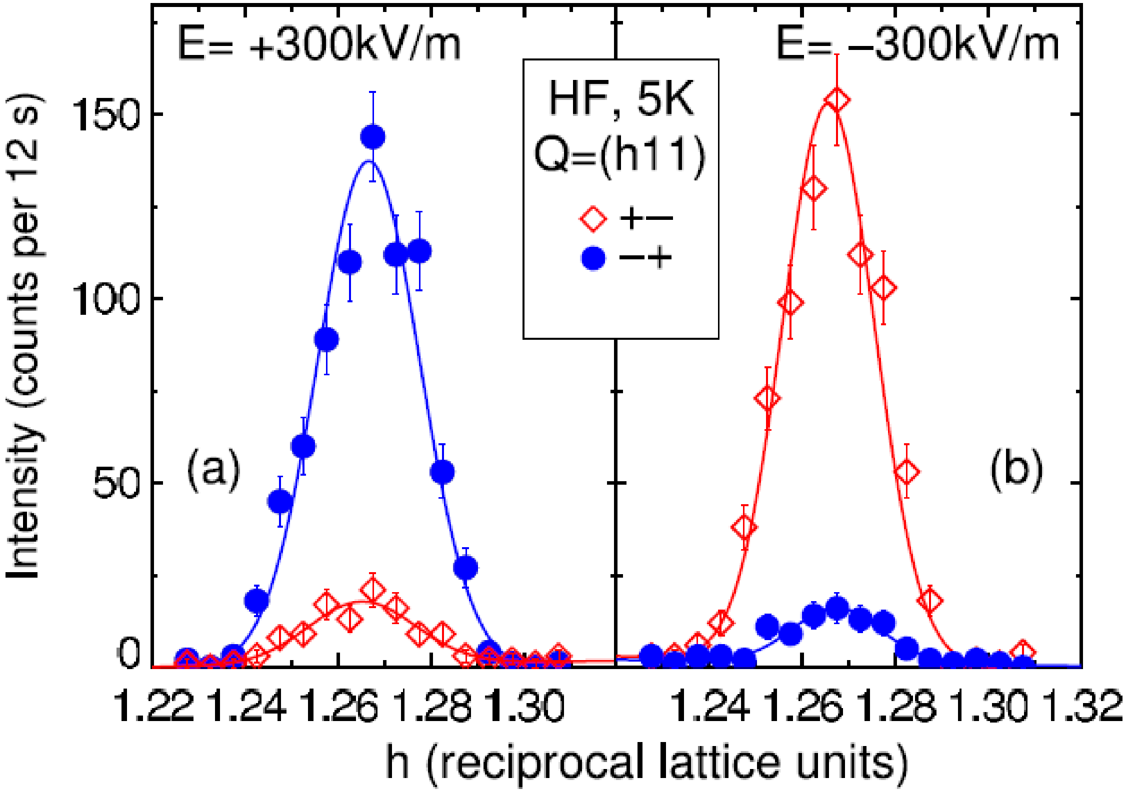
Coupled Magnetic and Ferroelectric Domains in Multiferroic $Ni_3V_2O_8$

I. Cabrera,[1,2] M. Kenzelmann,[3] G. Lawes,[4] Y. Chen,[2] W. C. Chen,[2] R. Erwin,[2] T. R. Gentile,[2] J. B. Leão,[2] J. W. Lynn,[2] N. Rogado,[5] R. J. Cava,[6] and C. Broholm[1,2]

Diffraction Example of Chirality Continued



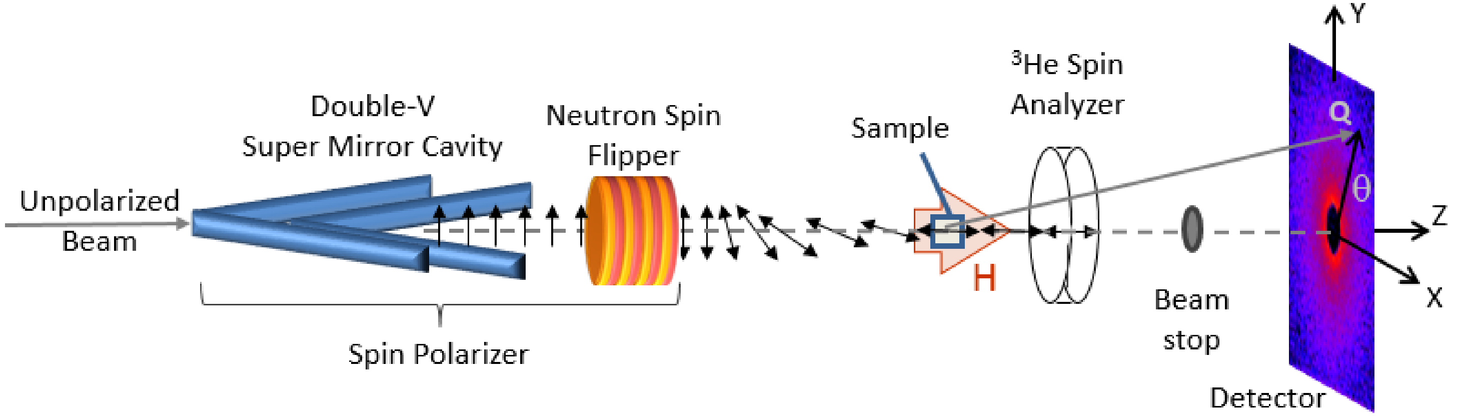
(a) NVO crystal sublattice showing Ni^{2+} spine (red) and cross-tie (blue) sites. (b) Counterclockwise (top) and clockwise (bottom) spin cycloids propagating along the $\mathbf{a}$ axis. The (green) vertical arrow indicates the direction of electric polarization.



DOI: https://doi.org/10.1103/PhysRevLett.103.087201

Consider **p ||** n-beam

$$\Upsilon_A(Q) = M_A$$
$$\Upsilon_B(Q) = M_B \cos^2(\theta) - M_C \sin(\theta)\cos(\theta)$$
$$\Upsilon_C(Q) = M_C \sin^2(\theta) - M_B \sin(\theta)\cos(\theta)$$



$$\sigma^{\downarrow\downarrow}_{\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}{}^{\uparrow\uparrow}(\mathbf{Q}) = N(\mathbf{Q})\,N^*(\mathbf{Q}) + M_{z,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,M^*_{z,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q}) \pm [M_{z,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,(\theta)\,N^*(\mathbf{Q}) + M^*_{z,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,N(\mathbf{Q})]$$

Nuclear and M || B are entirely non spin-flip

$$\sigma^{\uparrow\downarrow}_{\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}{}^{\downarrow\uparrow}(\mathbf{Q}) = M_{x,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,M^*_{x,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\sin^2(\theta) + M_{y,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,M^*_{y,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\cos^2(\theta) - \sin(\theta)\cos(\theta)[M_{x,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,M^*_{y,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q}) + M^*_{x,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})\,M_{y,\hat{\mathbf{p}}_z\|\hat{\mathbf{n}}}(\mathbf{Q})]$$

M ⊥ B is entirely spin-flip

Pro: Cleaner separation of M || B and M ⊥ B ; Con: can't point large of a field at ^{3}He analyzer

Comparison of **p** ⊥ n-beam and **p** || n-beam

Terms for $\hat{\mathbf{p}} \perp \hat{\mathbf{n}}$.

	$\sigma^{\downarrow\downarrow}+\sigma^{\uparrow\uparrow}$	$\sigma^{\downarrow\downarrow}-\sigma^{\uparrow\uparrow}$	$\sigma^{\uparrow\downarrow}+\sigma^{\downarrow\uparrow}$	Unpol
$\lvert N\rvert^2$	1	0	0	1
$\lvert M_x\rvert^2$	$\sin^4(\theta)$	0	$\sin^2(\theta)\cos^2(\theta)$	$\sin^2(\theta)$
$\lvert M_y\rvert^2$	$\sin^2(\theta)\cos^2(\theta)$	0	$\cos^4(\theta)$	$\cos^2(\theta)$
$\lvert M_z\rvert^2$	0	0	1	1
$2\lvert N\rvert\,\lvert M_x\rvert\,\overline{\cos}(\varphi_N-\varphi_{M_x})$	0	1	0	0
$-2\lvert N\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_N-\varphi_{M_y})$	0	1	0	0
$-2\lvert M_x\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_{M_x}-\varphi_{M_y})$	$\sin^3(\theta)\cos(\theta)$	0	$\sin(\theta)\cos^3(\theta)$	$\sin(\theta)\cos(\theta)$

Can get M || B from Y-cut and X-cut subtraction or N-M cross-term; also seen in spin-flip channel with 4-fold symmetry.

Spin-flip has all three magnetic components

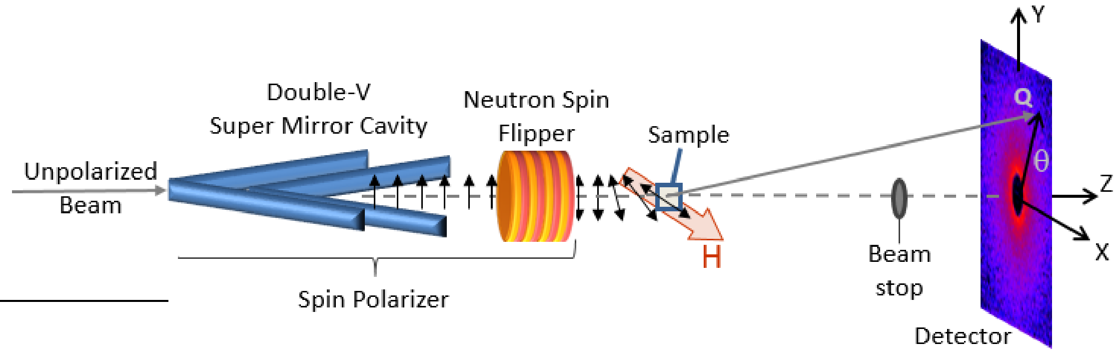
Terms for $\hat{\mathbf{p}} \parallel \hat{\mathbf{n}}$. The choice of X and Y axes within the plane $\perp\hat{\mathbf{n}}$ is arbitrary.

	$\sigma^{\downarrow\downarrow}+\sigma^{\uparrow\uparrow}$	$\sigma^{\downarrow\downarrow}-\sigma^{\uparrow\uparrow}$	$\sigma^{\uparrow\downarrow}+\sigma^{\downarrow\uparrow}$	Unpol
$\lvert N\rvert^2$	1	0	0	1
$\lvert M_z\rvert^2$	1	0	0	1
$\lvert M_x\rvert^2$	0	0	$\sin^2(\theta)$	$\sin^2(\theta)$
$\lvert M_y\rvert^2$	0	0	$\cos^2(\theta)$	$\cos^2(\theta)$
$2\lvert N\rvert\,\lvert M_z\rvert\,\overline{\cos}(\varphi_N-\varphi_{M_z})$	0	1	0	0
$-2\lvert M_x\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_{M_x}-\varphi_{M_y})$	0	0	$\sin(\theta)\cos(\theta)$	$\sin(\theta)\cos(\theta)$

Always have nuclear + M || B in non-spin-flip, so separation of the two may be tricky (unless can turn off magnetism in the sample)

Spin-flip is cleanly M ⊥ B

Consider Half-Polarization



Terms for $\hat{\mathbf{p}} \perp \hat{\mathbf{n}}$.

	$\sigma^{\downarrow\downarrow} + \sigma^{\uparrow\uparrow}$	$\sigma^{\downarrow\downarrow} - \sigma^{\uparrow\uparrow}$	$\sigma^{\uparrow\downarrow} + \sigma^{\downarrow\uparrow}$	Unpol
$\lvert N\rvert^2$	1	0	0	1
$\lvert M_x\rvert^2$	$\sin^4(\theta)$	0	$\sin^2(\theta)\cos^2(\theta)$	$\sin^2(\theta)$
$\lvert M_y\rvert^2$	$\sin^2(\theta)\cos^2(\theta)$	0	$\cos^4(\theta)$	$\cos^2(\theta)$
$\lvert M_z\rvert^2$	0	0	1	1
$2\lvert N\rvert\,\lvert M_x\rvert\,\overline{\cos}(\varphi_N - \varphi_{M_x})$	0	1	0	0
$-2\lvert N\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_N - \varphi_{M_y})$	0	1	0	0
$-2\lvert M_x\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_{M_x} - \varphi_{M_y})$	$\sin^3(\theta)\cos(\theta)$	0	$\sin(\theta)\cos^3(\theta)$	$\sin(\theta)\cos(\theta)$

↓-↑ would be same as ↓ ↓ - ↑ ↑ except for an extra My-Mz (chiral) cross-term

Terms for $\hat{\mathbf{p}} \parallel \hat{\mathbf{n}}$. The choice of X and Y axes within the plane $\perp \hat{\mathbf{n}}$ is arbitrary.

	$\sigma^{\downarrow\downarrow} + \sigma^{\uparrow\uparrow}$	$\sigma^{\downarrow\downarrow} - \sigma^{\uparrow\uparrow}$	$\sigma^{\uparrow\downarrow} + \sigma^{\downarrow\uparrow}$	Unpol
$\lvert N\rvert^2$	1	0	0	1
$\lvert M_z\rvert^2$	1	0	0	1
$\lvert M_x\rvert^2$	0	0	$\sin^2(\theta)$	$\sin^2(\theta)$
$\lvert M_y\rvert^2$	0	0	$\cos^2(\theta)$	$\cos^2(\theta)$
$2\lvert N\rvert\,\lvert M_z\rvert\,\overline{\cos}(\varphi_N - \varphi_{M_z})$	0	1	0	0
$-2\lvert M_x\rvert\,\lvert M_y\rvert\,\overline{\cos}(\varphi_{M_x} - \varphi_{M_y})$	0	0	$\sin(\theta)\cos(\theta)$	$\sin(\theta)\cos(\theta)$

↓-↑ would be same as ↓ ↓ - ↑ ↑

If want M || B, then half-pol may be the best way to go.

Rough Guidelines For (Magnetic) Polarized Scattering

- Often start with unpolarized scattering to get a feel of overall scattering intensity
- If sample magnetically saturates, will check (a) Intensity ⊥ B vs. Intensity || B or (b) high-field minus low field , or (c) changes in magnetism above and below blocking to get a feel for magnetic signal strength. The caveat is that moments ⊥ B may interfere with simple subtraction methods.
- Half-polarization can be exceptionally helpful in boosting the M || B signal in the form of ↓ minus ↑ nuclear-magnetic cross-term
- If interested in in moments ⊥ B, can attempt low-field minus high field (especially along direction || B), but if signal is supposed to be small relative to structural scattering, full-polarization may be the only way to extract it
- Cost of polarization analysis: loose half the intensity with polarizer, and double the counting channels if take ↑ and ↓ cross-sections (factor of 2 to 4)
- If add ^{3}He analyzer, for example, the starting transmission of the desired spin state is about 50% (higher if using simple supermirror). Will also need to take 4 cross-sections. Yet, if spin-flip is the goal, won't have to count as long to overcome (typical) dominant structural scatting.
- Often bias the spin-flip to non spin-flip in a ratio of 2 or 3: 1. Typically don't take scans longer than an hour each in order to do a good time corrections for the ^{3}He spin filter, if used.

Summary

- Polarization analysis of the neutron spin is ideal for separating (1) nuclear from magnetic scattering and (2) magnetism || and ⊥ to **p**
- Magnetic scattering <1% that of the structural scattering can be measured with careful correction of any polarization leakage
- Polarization analysis can be used to effectively measure phase – unambiguously solve structures (magnetic reference layers) and measure changes in energy (NSE)
- Polarization analysis can be used to separate (and/or remove) incoherent background scattering
- Consider whether full polarization, half-polarization, and/or unpolarized scattering best suit the your needs for a specific sample