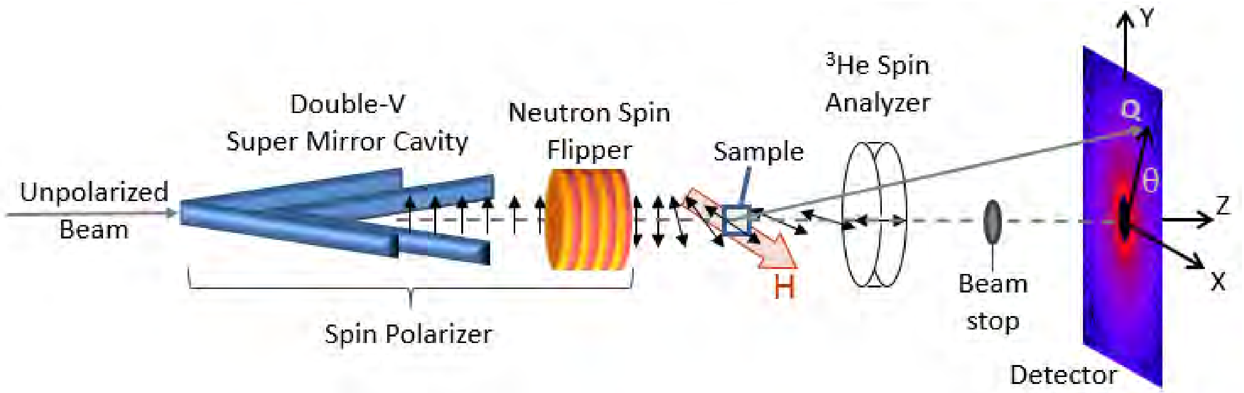
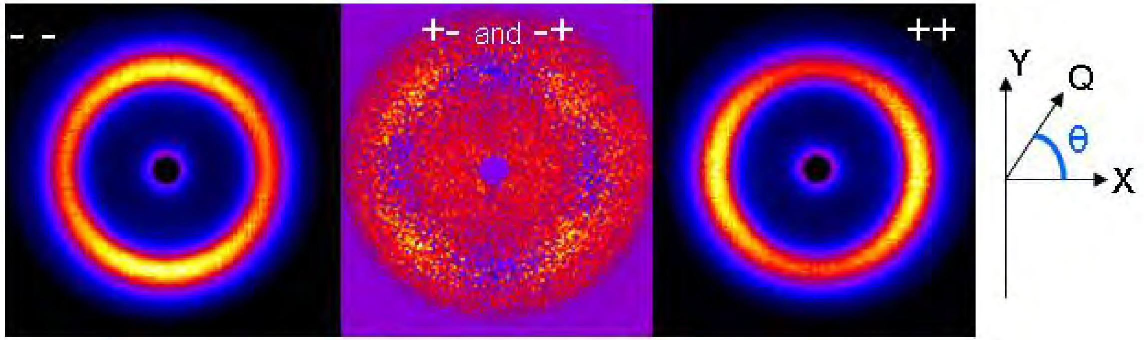
National School on Neutron and X-Ray Scattering
Argonne and Oak Ridge National Laboratories

Polarized Neutron Scattering

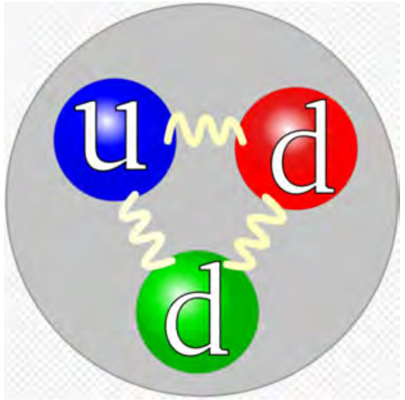
June 19, 2019





Kathryn Krycka
NIST Center for Neutron Research, Gaithersburg, MD

Neutron Properties

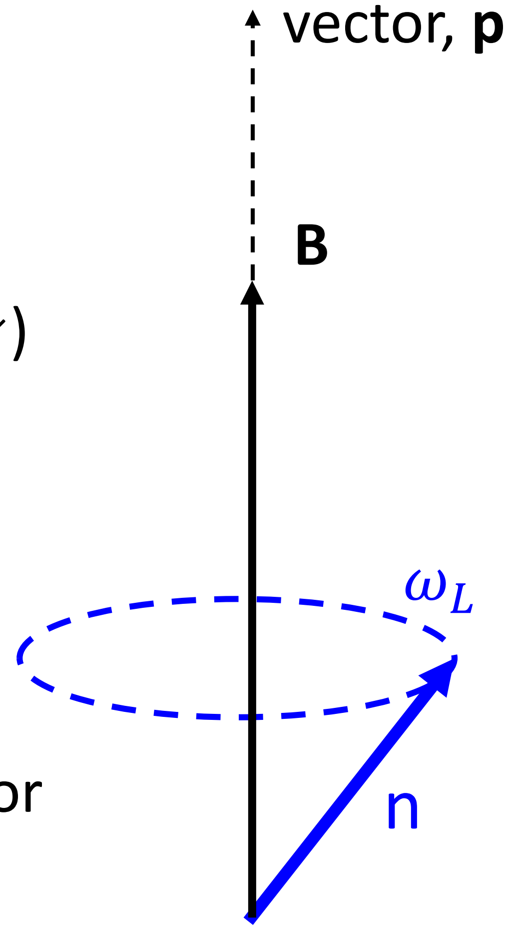


https://en.wikipedia.org/wiki/Neutron

- Hadron comprised of three quarks; no net charge (insensitive to electric fields)
- Free lifetime of 881.5±1.5 seconds (15 minutes)
- Spin ½ (fermion) that gives rise to a magnetic moment
- μ_n = - 1.913 μ_N (nuclear magneton) = -9.662 x 10^{-27} J/T (or ≈ µB / 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 neurons), 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 $\gamma_L =$ −1.833 x 104 rad/Gauss−sec and Larmor frequency $\omega_L = -\gamma|B|$



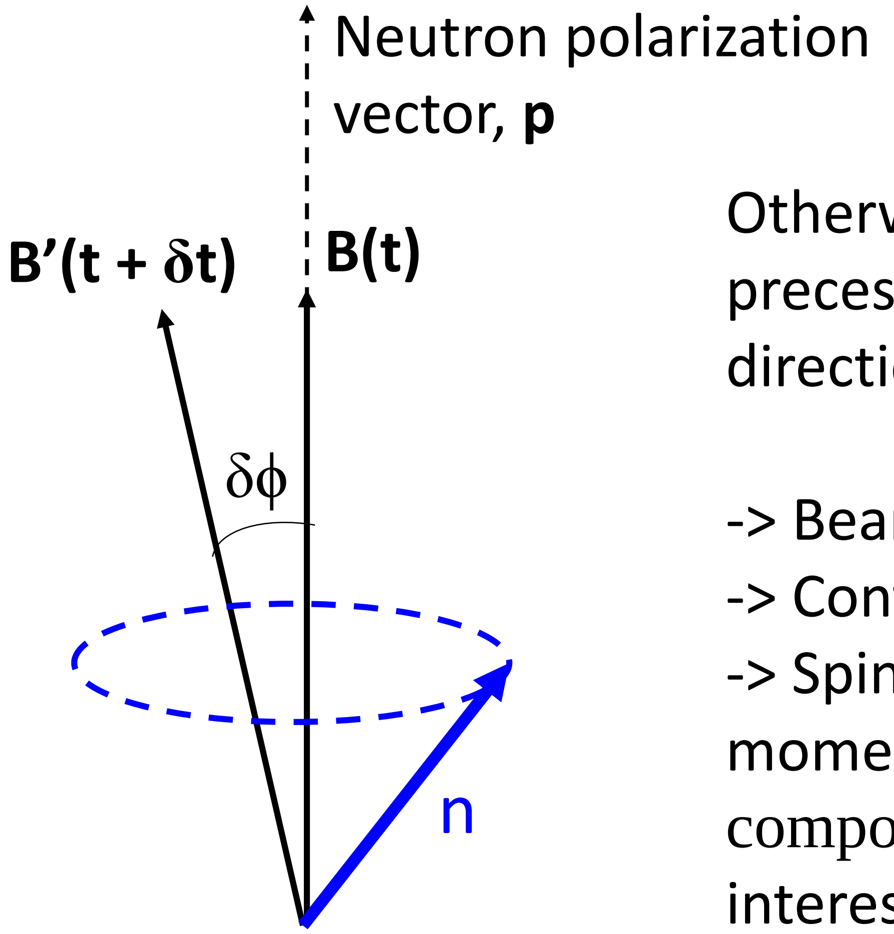
Reproduced largely from Roger Pynn's Neutron Scattering Lecture 10

Neutron's Response to Varying Magnetic Field

Neutron adiabatically follows field (retains polarization) if

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

$\omega_L = -\gamma|B| = -1.833 \times 104$ rad/Gauss−sec



Otherwise, the neutron will precess about a new field direction

-> Beam depolarization
-> Controlled flipping devices
-> Spin flip from magnetic moments (⊥Q and with component ⊥ 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

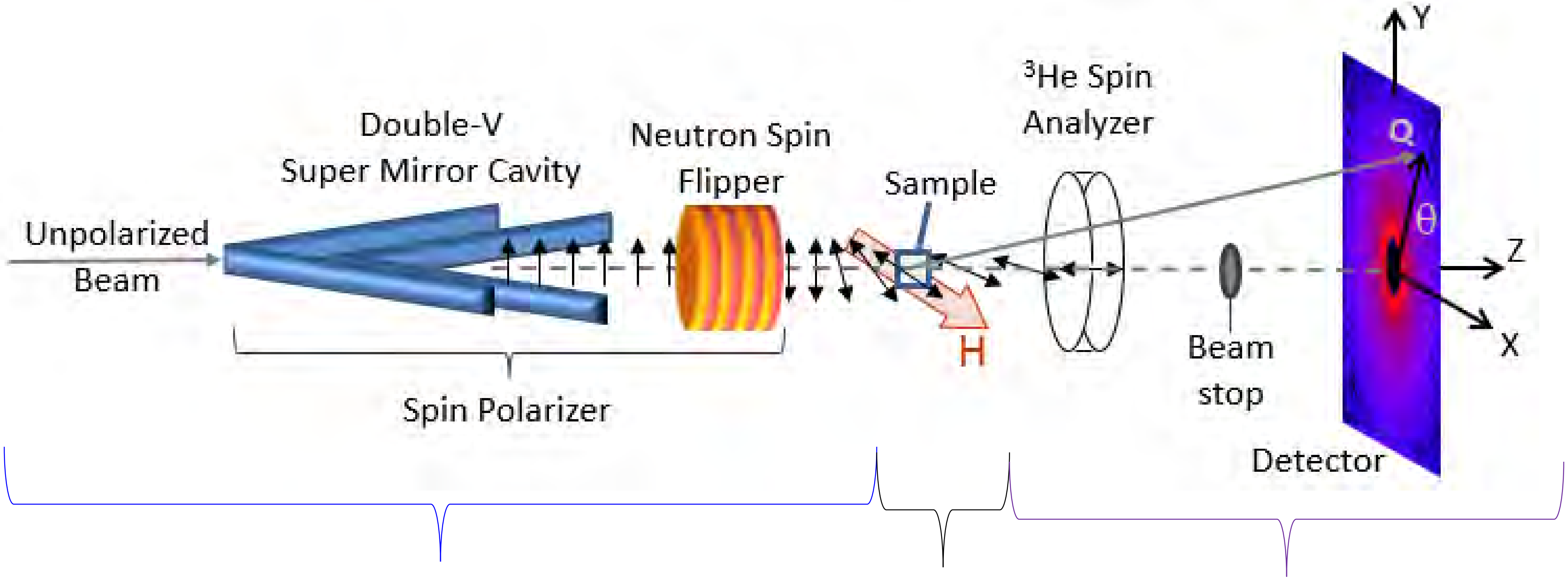
- Only the component of the magnetic moment (or magnetic form factor), M, that is $\perp$ Q may participate in neutron scattering. This is embodied in the Halpern-Johnson vector (Phys. Rev. 55, 898 (1939)) as:

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

- 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)):

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

General Experimental Set-Up (SANS example shown)



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

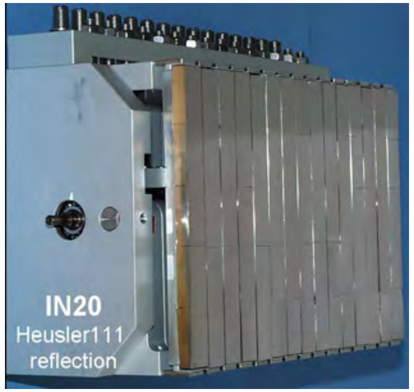
Sample Scattering:
Often 1D analysis w/ B-field, but 3D analysis w/o field possible (spherical polarimetry)

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

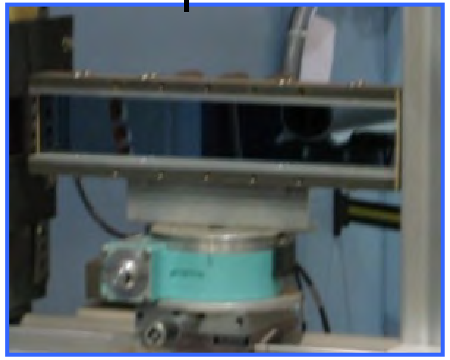
Beam Polarization

- 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

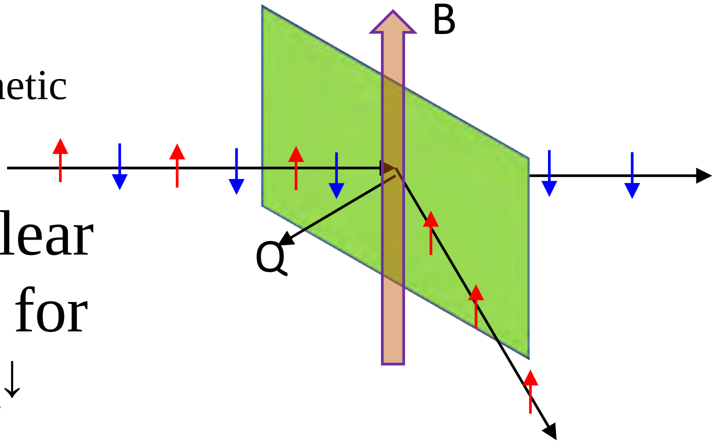


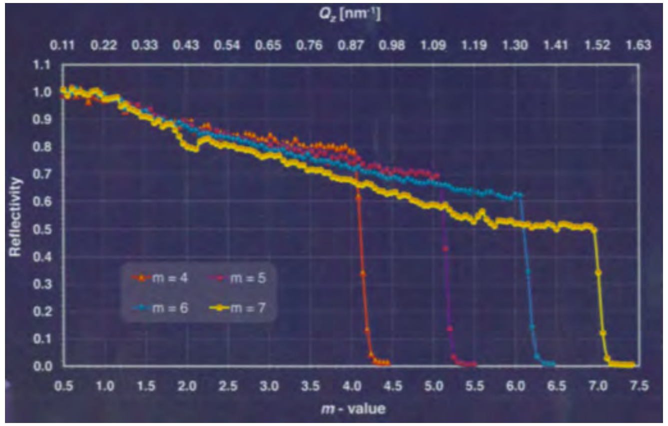
^3He

Polarizing Crystal, Supermirror

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

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

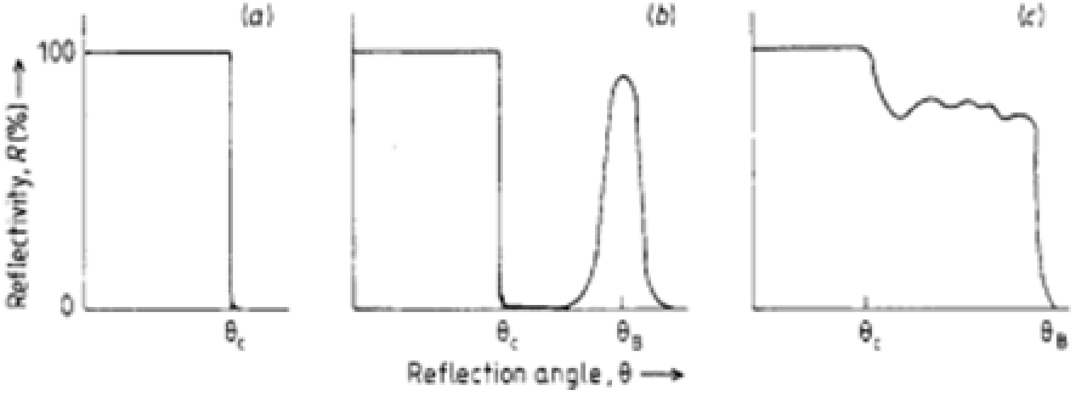




Courtesy of Swiss Neutronics

- 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.

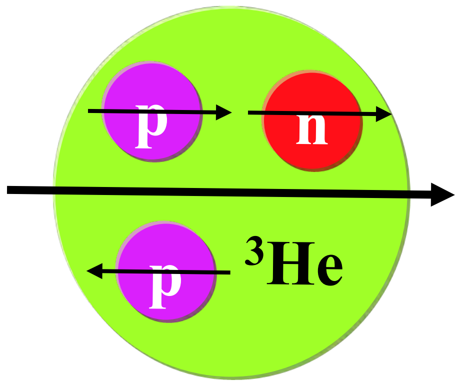


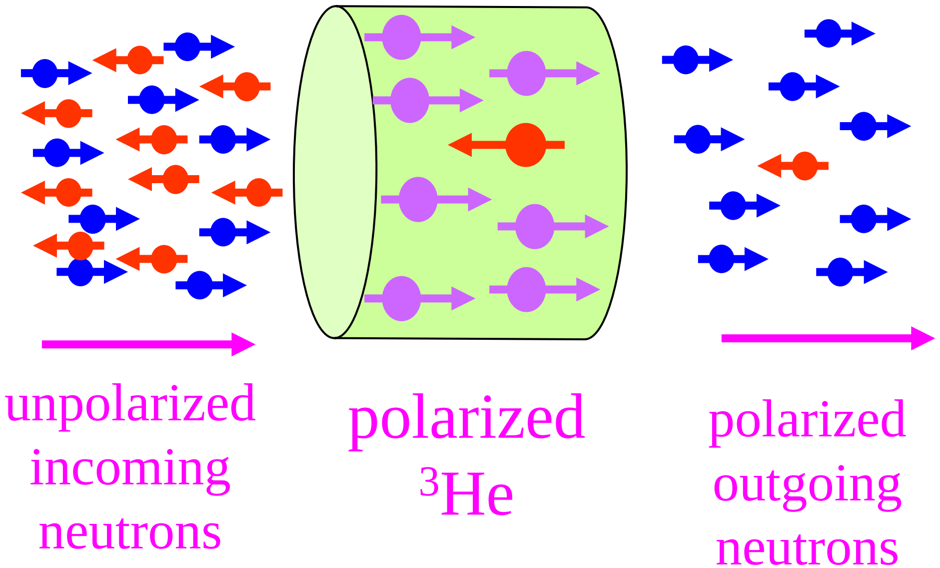
(a) Reflecting mirror, (b) multilayer, (c) supermirror
J W White and C G Windsor, Rep. Prog. Phys. V 47, 707-765, 1984

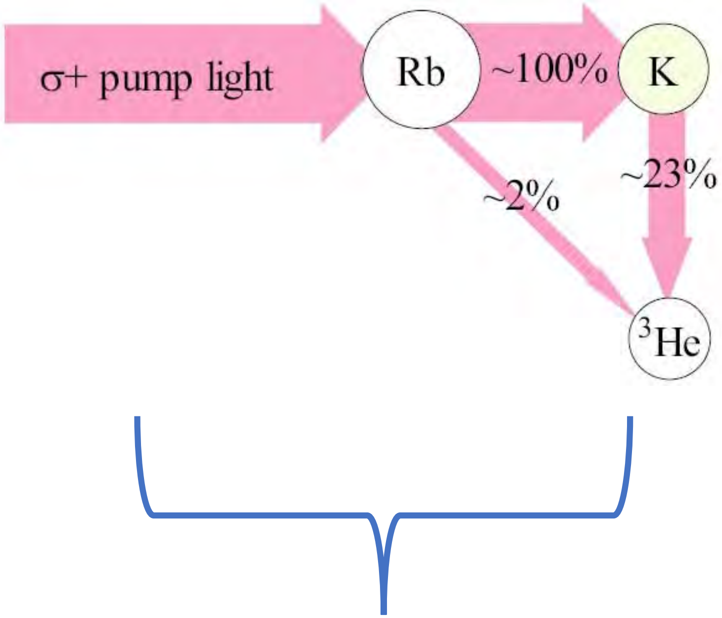
^{3}He Spin Filters

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

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







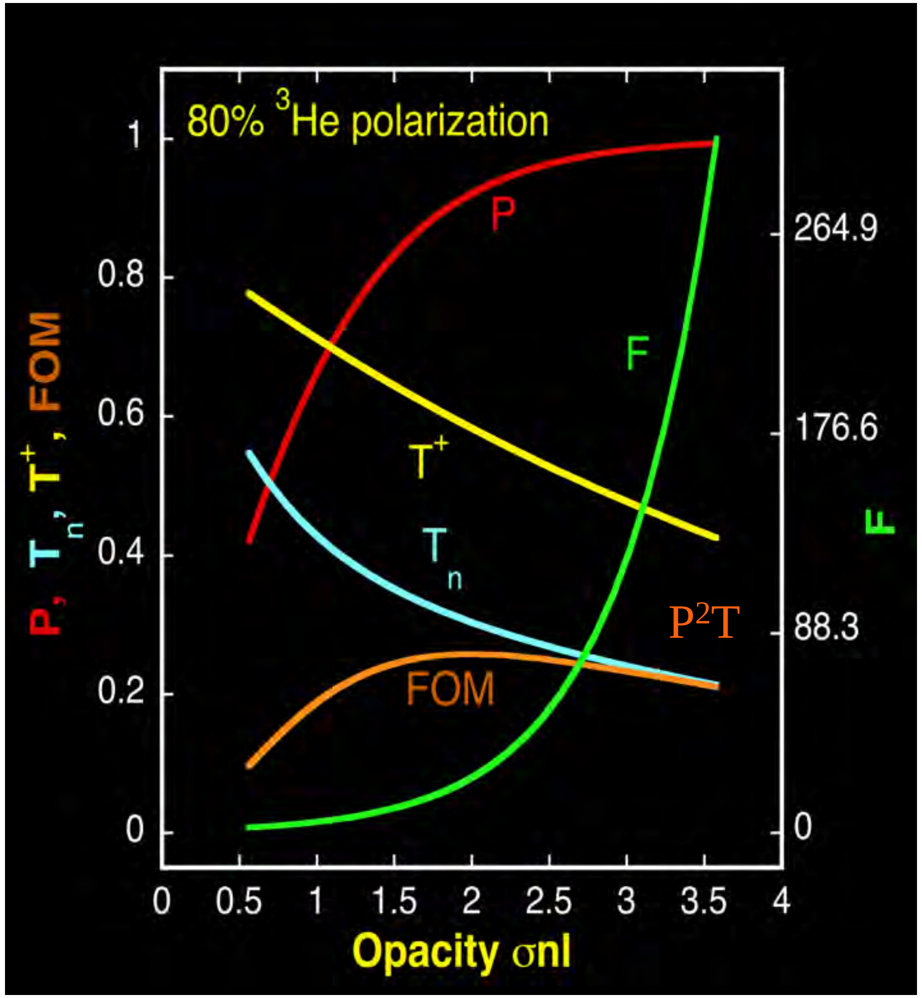
^{3}He optically pumped to desired spin state

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

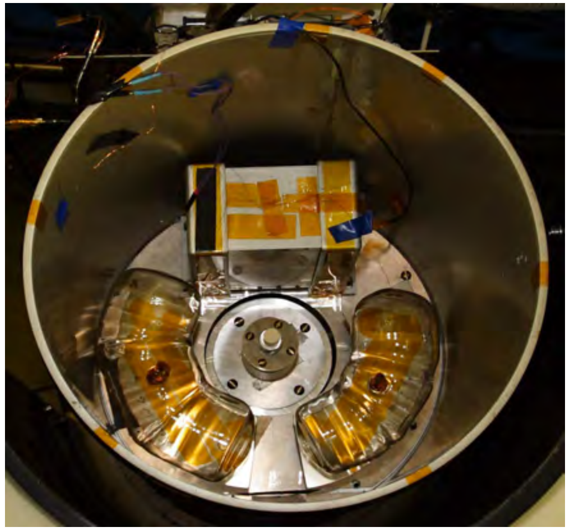
Courtesy of Wangchun Chen

Polarized ^{3}He Neutronic Performance

80% ^{3}He Pol.



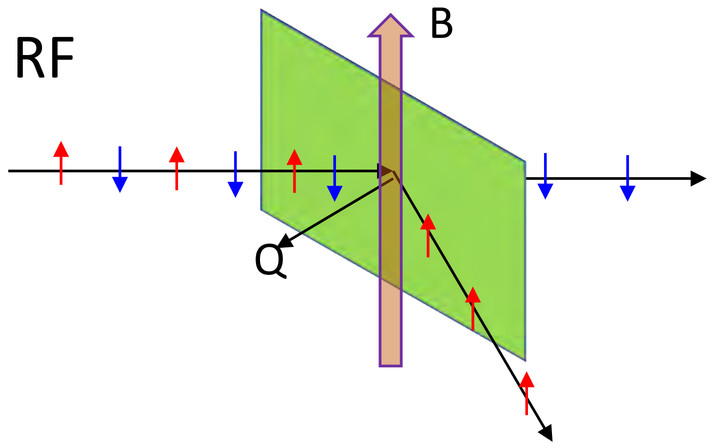
Courtesy of Wangchun Chen



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

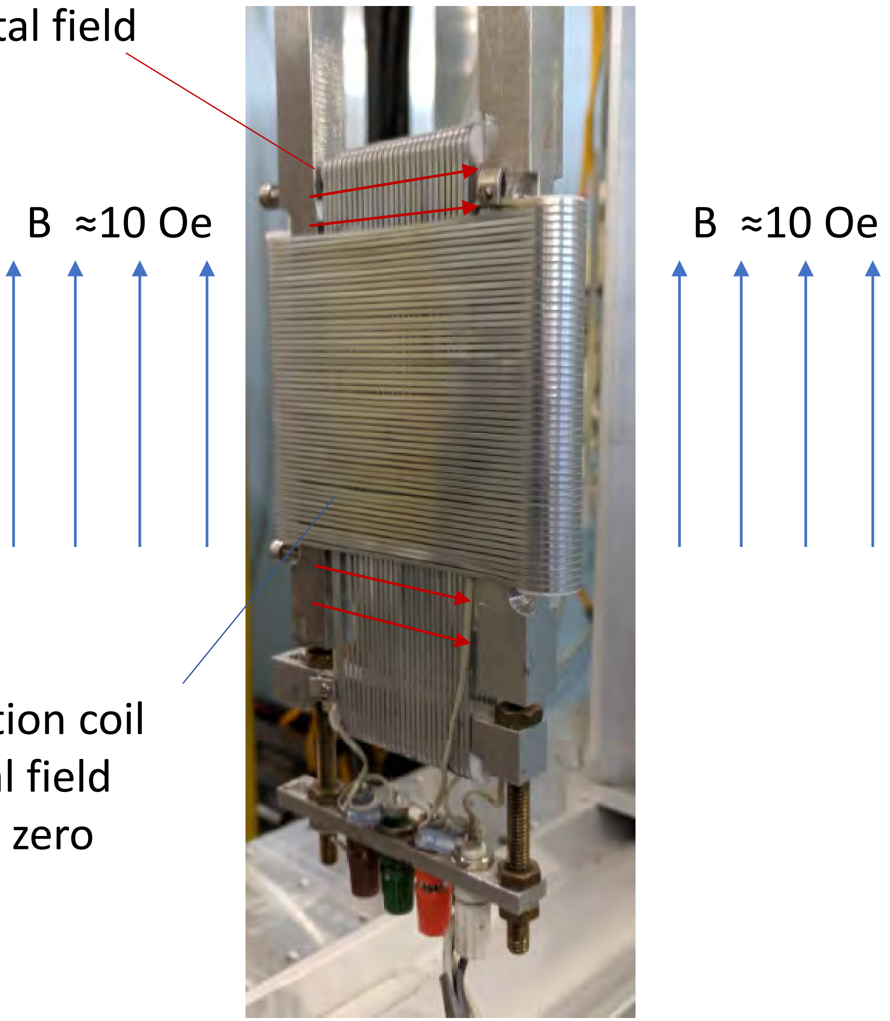
Neutron Spin Flipping

- Spin reversal must be with respect to B-field (not a simple adiabatic transition)
- For ^{3}He, spin reversal is built in by reversing ^{3}He spins via RF pulse
- If can rotate your supermirror angle, may be able to vary between spin states
- Current sheet (tuned for specific neutron wavelength, material in beam)
- White-beam, gradient field spin-flipper (appropriate for multiple wavelengths, no material in beam)



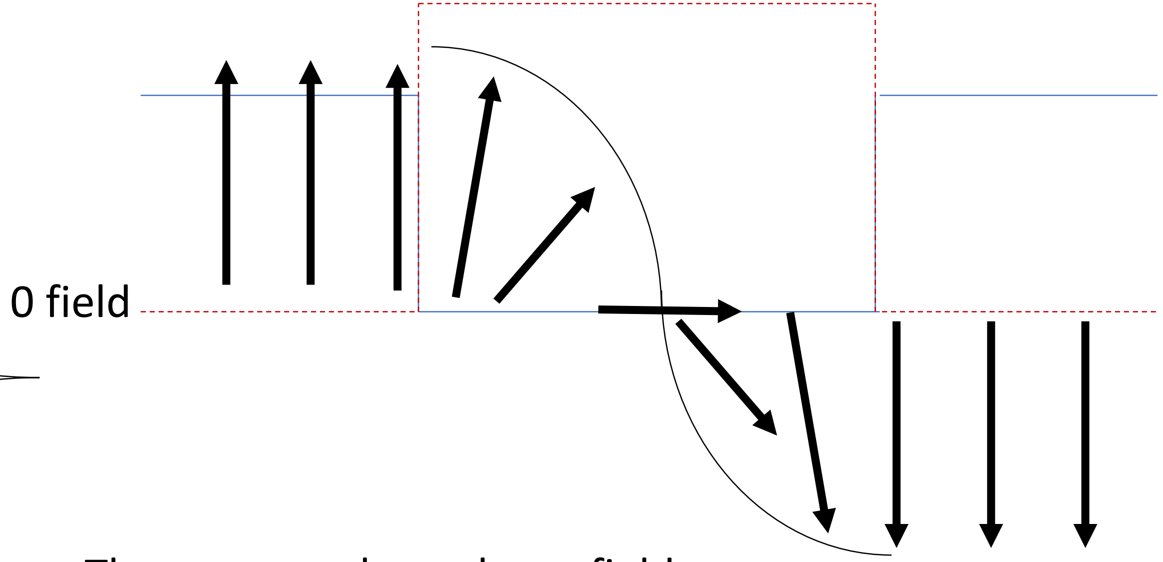
Current Sheet

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

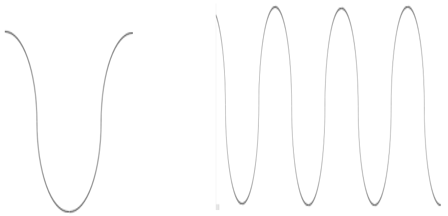


Compensation coil sets vertical field abruptly to zero

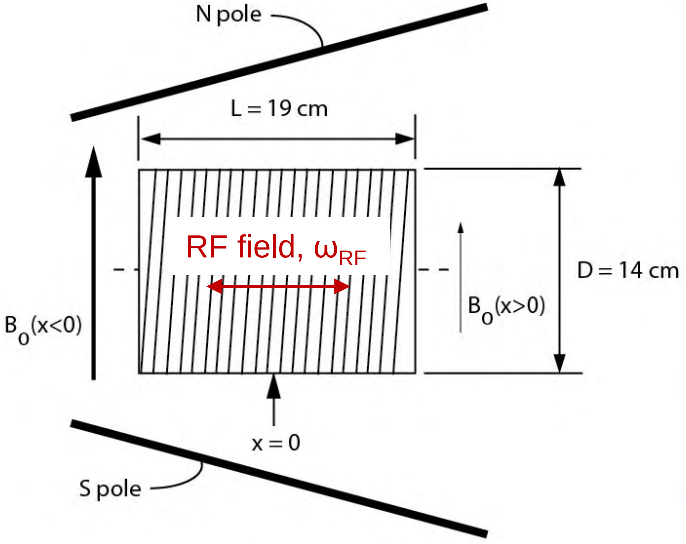
Blue is vertical field
Red is horizontal field
Black neutron spins



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

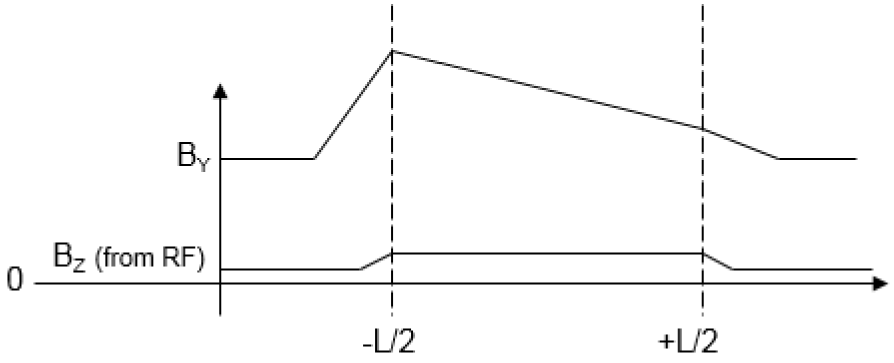


Spin Gradient, RF-Flipper

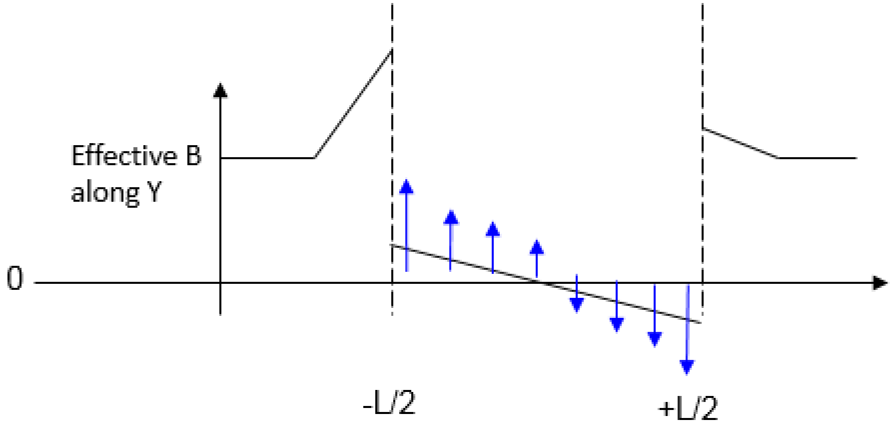


300 kHz ~ 100 G

Asterix Flipper, M. Fitzsimmons



$$B_{Y,\ effective} = B_Y - \omega_L / \Upsilon_L$$



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

- 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

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 ($\wp_{^3\mathrm{He}}$)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)

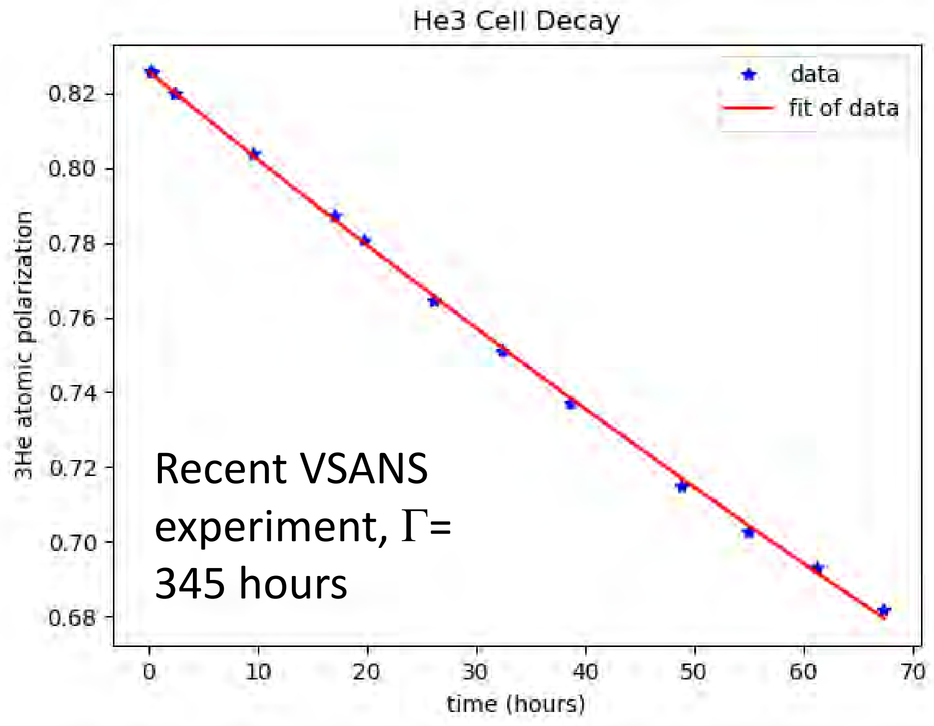
$$T^{\text{majority, minority}}_{^3\mathrm{He}} = T_{\mathrm{E}} \exp\left[-\mu(1 \mp \wp_{^3\mathrm{He}})\right]$$

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

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

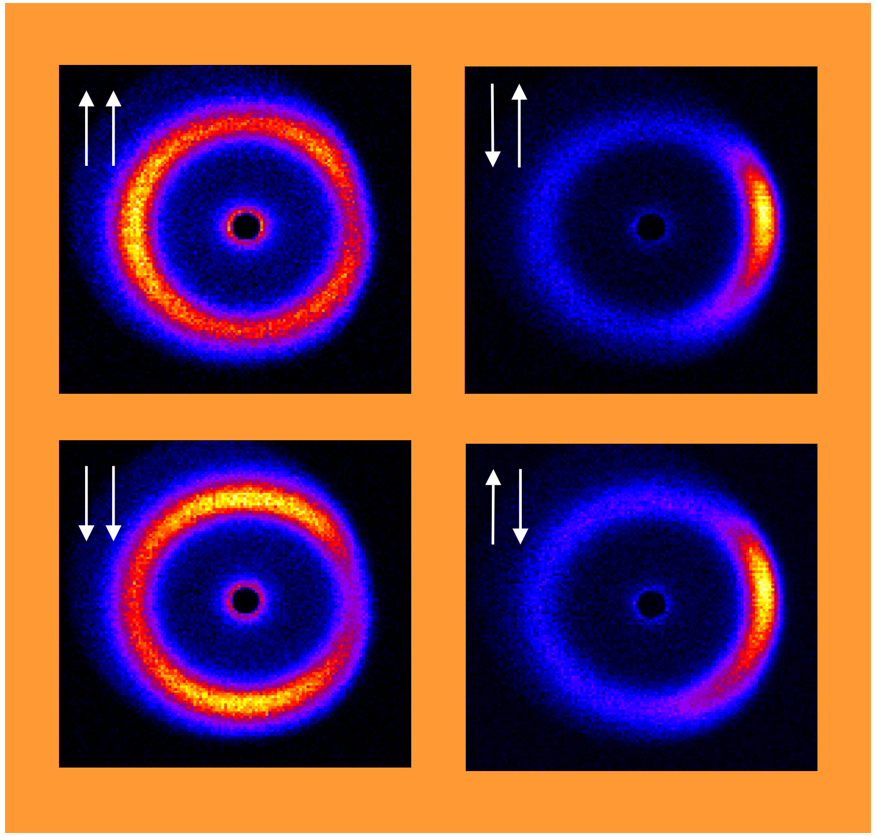
- Time dependence of ^{3}He polarization decay should be accounted for:

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

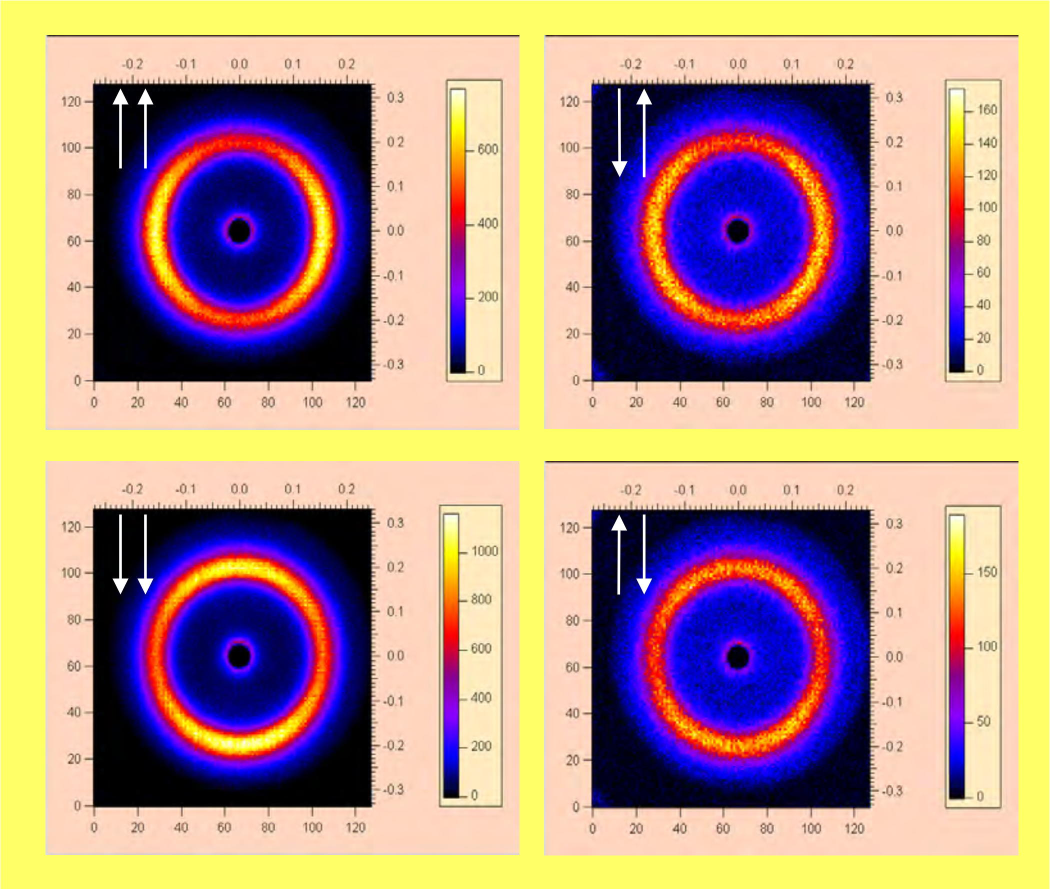


Polarized Beam Characterization II.

- 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

Neutron Interactions with Sample

Strong interaction:

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

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:

$$V(\mathbf{x}) = -\vec{\mu}_n \cdot \vec{\mathbf{B}}(\mathbf{x})$$

Magnetic field

Local magnetization

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

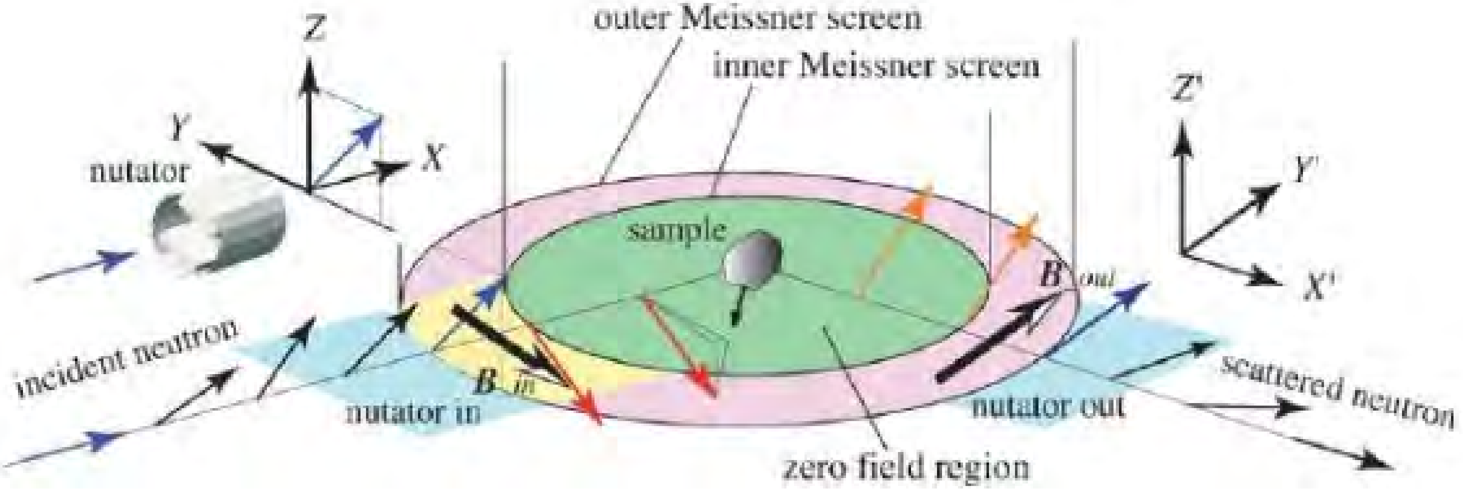
Magnetic scattering is sensitive to local magnetic fields i.e. magnetization

- 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

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)

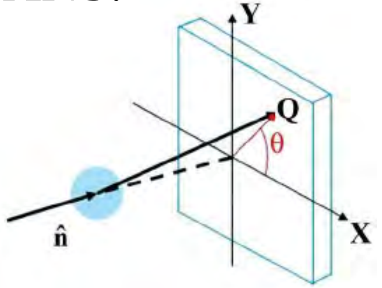
Focus on 1D Polarization (polarization axis, **p**, defined by B at sample)

- 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:

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

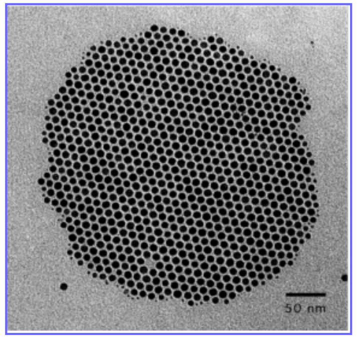


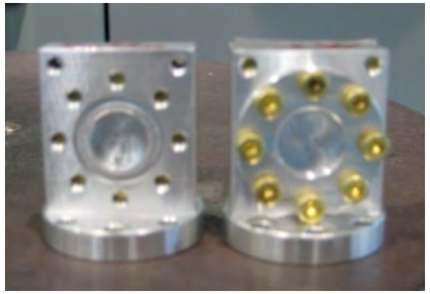
- Rule 2: Of M ⊥ Q (defined by ϒ), the portion || **p** contributes to non-spin flip, while the portion ⊥ **p** contributes to spin-flip (Moon, Riste, Koehler, Phys. Rev. 181, 920 (1969)). Note we are here neglecting any nuclear magnetic scattering, which is often unpolarized and negligible. A common 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.

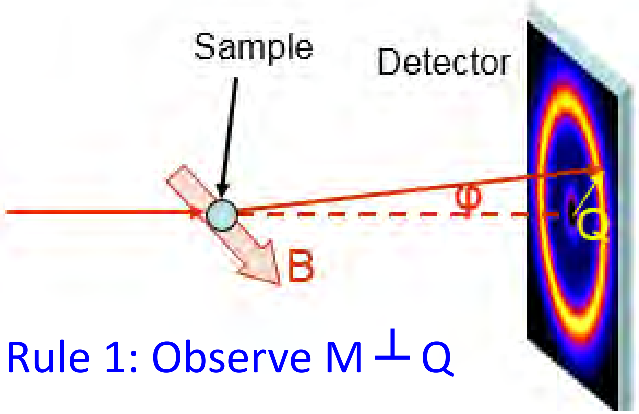
$$\sigma^{\downarrow\downarrow}_{\uparrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}\left|N \pm \Upsilon_A\right|^2, \quad \sigma^{\uparrow\downarrow}_{\downarrow\uparrow}(\mathbf{Q}) = \tfrac{1}{2}\left|(-\Upsilon_B \mp i\Upsilon_C)\right|^2$$

Example to Motivate Polarization Analysis

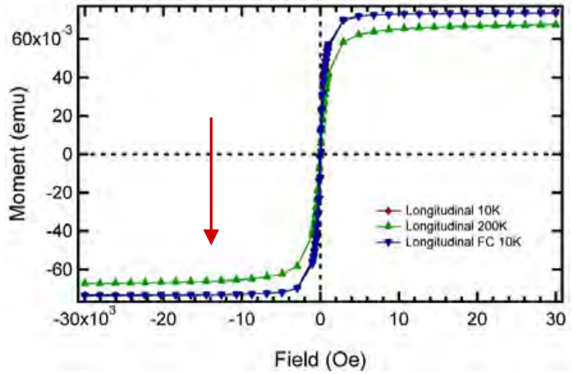
Monodisperse, 9 nm, ferrimagnetic magnetite (Fe_3O_4) particles crystallize into a face-centered cubic crystallites ≈ μm. These crystallites are randomly oriented and form a powder. Magnetite is commonly used due to bio-compatibility stability, and a moment comparable to Ni.



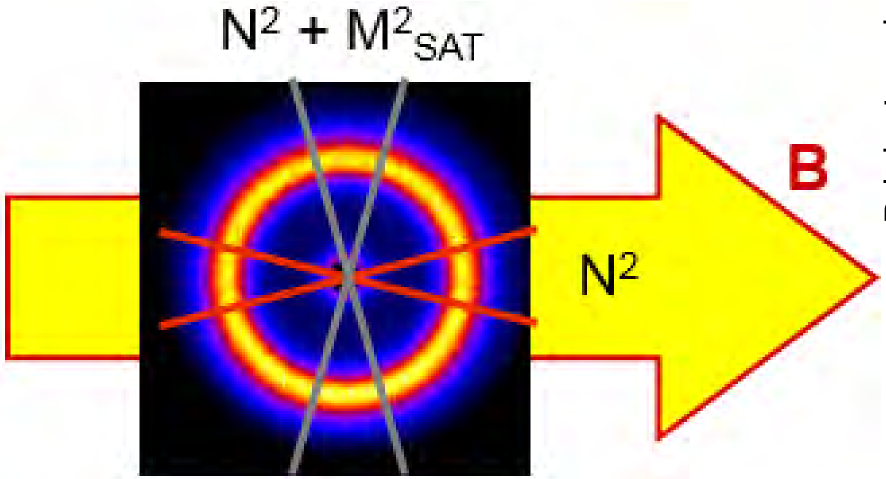




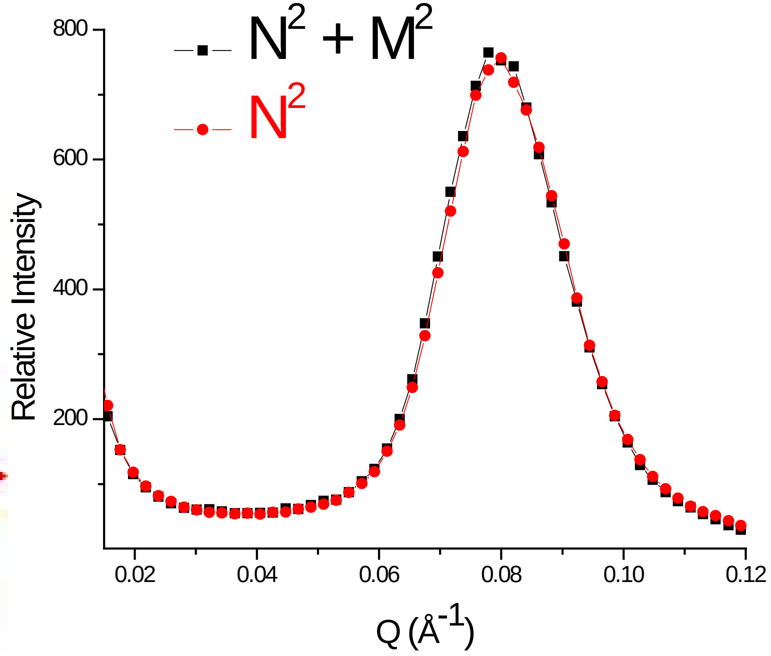
Rule 1: Observe M ⊥ Q



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

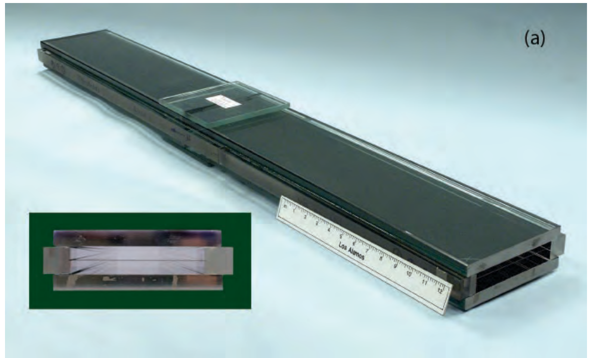


$(\rho_M / \rho_N)^2 = 4\%$



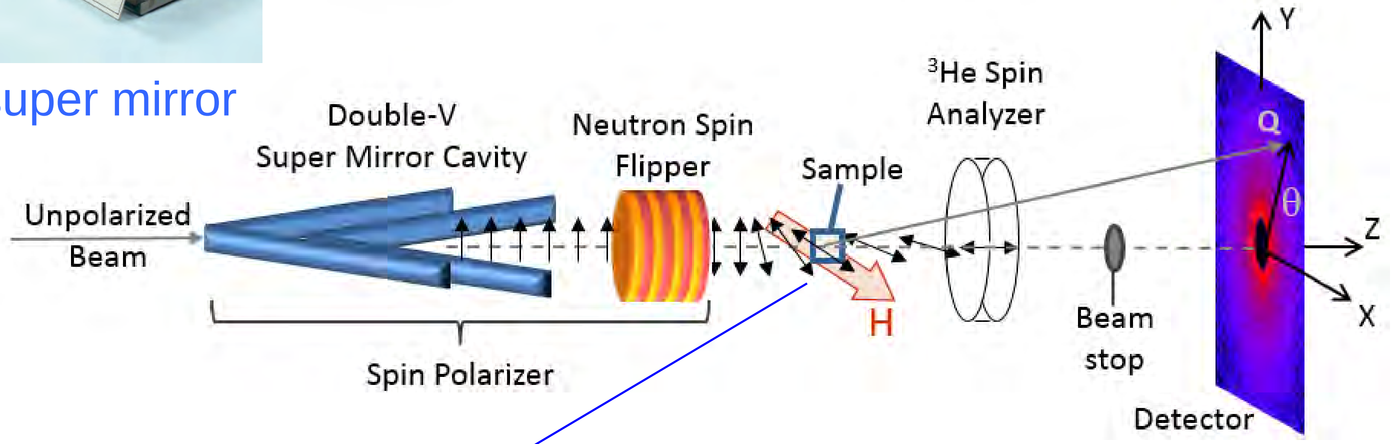
0.08 $Å^{-1}$ peak (111) reflection in 13.6 nm FCC lattice

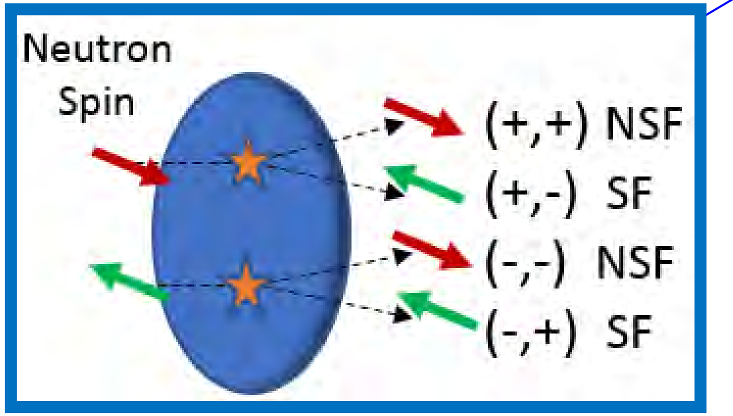
Polarization Analyzed SANS (SANSPOL, POLARIS, PASANS)



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 ($M^{\perp}$ Q) that is || H

SF → the projection of ($M^{\perp}$ Q) that is ⊥ H

Thus, spin-flip is entirely magnetic!

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

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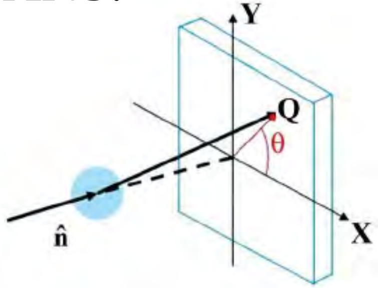
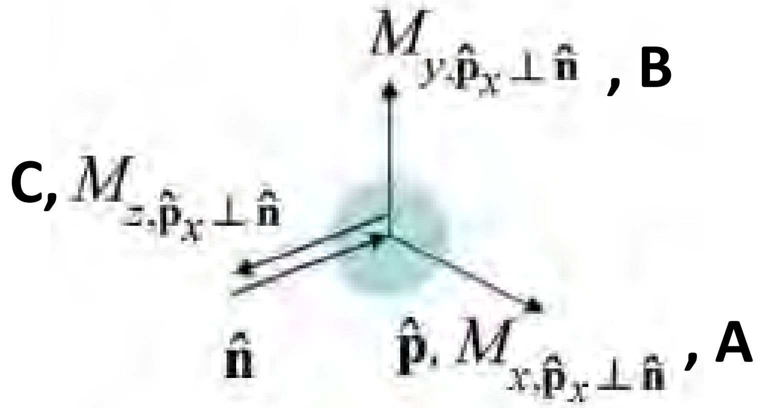
$$\Upsilon_{J=A,B,C}(\hat{\mathbf{Q}}) = \sum_{L=A,B,C} M_L[\cos(\omega_{L,J}) - \cos(\omega_{\mathbf{Q},J})\cos(\omega_{\mathbf{Q},L})]$$



- 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.

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

Specifics for p ⊥ n-beam ($N, M_J(\mathbf{Q}) = |N, M_J| \exp(i\varphi_{N,M_J})$)



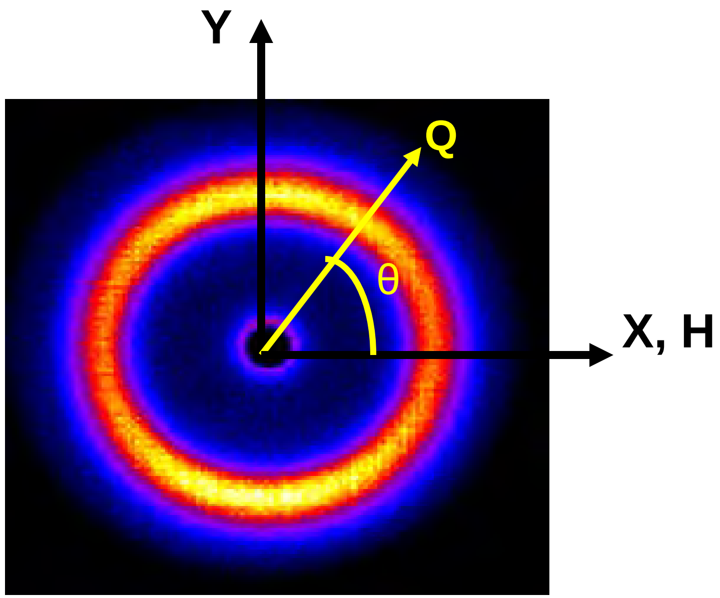
$$\Upsilon_{J=A,B,C}(\hat{\mathbf{Q}}) = \sum_{L=A,B,C} M_L[\cos(\omega_{L,J}) - \cos(\omega_{\mathbf{Q},J})\cos(\omega_{\mathbf{Q},L})]$$

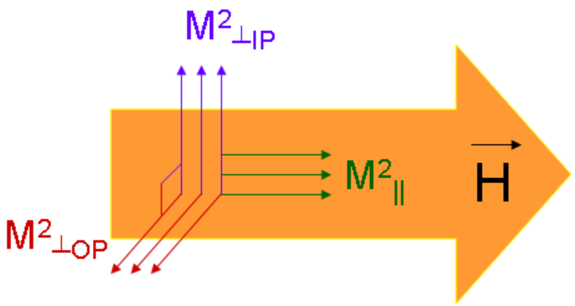
$$\Upsilon_A(\mathbf{Q}) = M_A \sin^2(\theta) - M_B \sin(\theta)\cos(\theta)$$
$$\Upsilon_B(\mathbf{Q}) = M_B \cos^2(\theta) - M_A \sin(\theta)\cos(\theta)$$
$$\Upsilon_C(\mathbf{Q}) = M_C$$

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

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

Specifics for p ⊥ n-beam





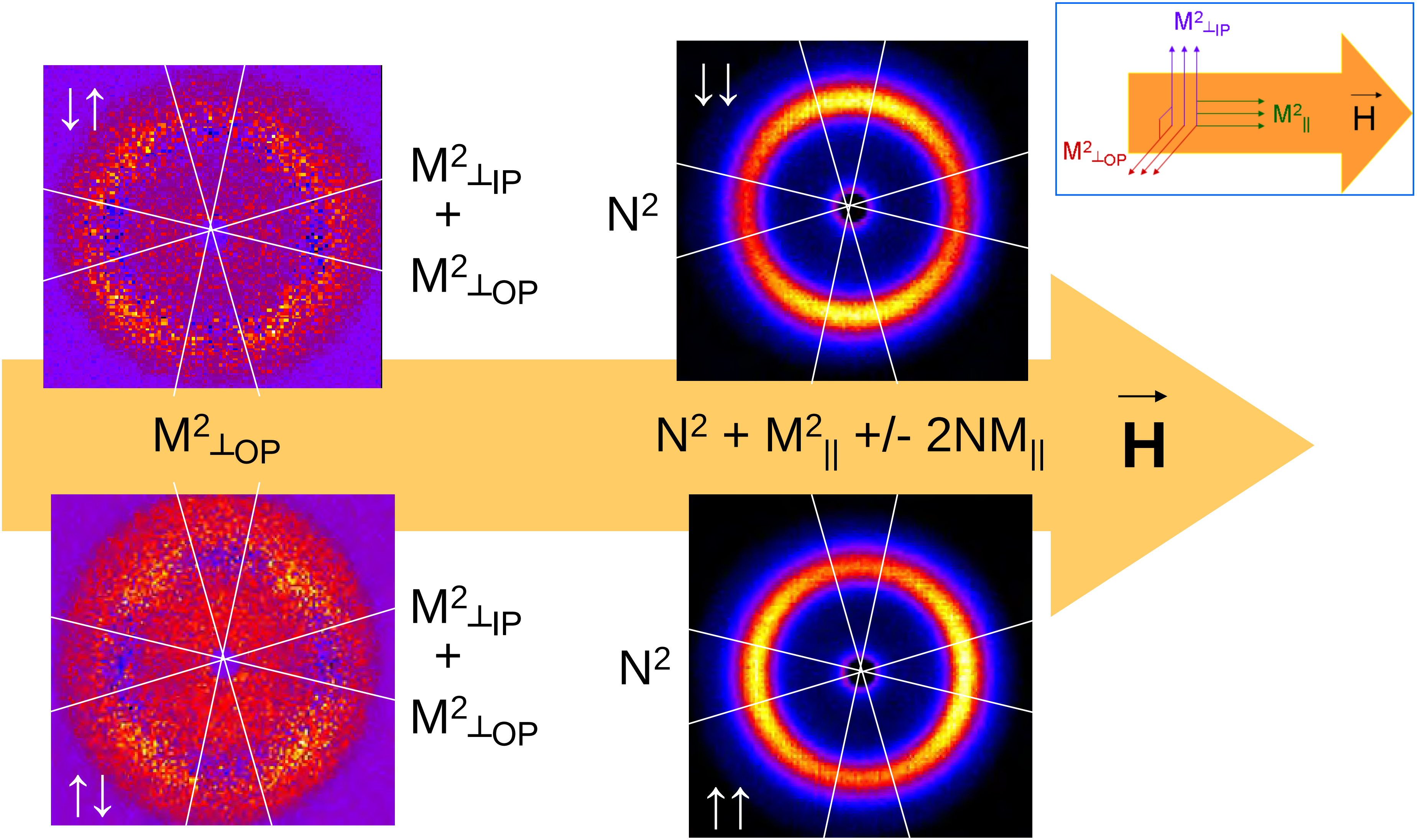
$$\alpha\alpha^* = |\alpha|^2,$$
$$\alpha\beta^* + \alpha^*\beta = 2|\alpha|\,|\beta|\,\overline{\cos}(\varphi_\alpha - \varphi_\beta),$$
$$i(\alpha\beta^* - \alpha^*\beta) = -2|\alpha|\,|\beta|\,\overline{\sin}(\varphi_\alpha - \varphi_\beta)$$

$$I^{--,++} = |N|^2 + \sin^2(\theta)\cos^2(\theta)|M_{\perp ip}|^2 + \sin^4(\theta)|M_{||}|^2$$
$$-2\cos(\theta)\sin^3(\theta)|M_{||}||M_{\perp ip}|\cos(\varphi_{||}-\varphi_{\perp ip})$$
$$\pm 2\sin(\theta)\cos(\theta)|N||M_{\perp ip}|\cos(\varphi_N-\varphi_{\perp ip})$$
$$\mp 2\sin^2(\theta)|N||M_{||}|\cos(\varphi_N-\varphi_{||})$$

$$I^{+-,-+} = |M_{\perp op}|^2 + \cos^4(\theta)|M_{\perp ip}|^2 + \sin^2(\theta)\cos^2(\theta)|M_{||}|^2$$
$$-2\sin(\theta)\cos^3(\theta)|M_{||}||M_{\perp ip}|\cos(\varphi_{||}-\varphi_{\perp ip})$$
$$\pm 2\sin(\theta)\cos(\theta)|M_{||}||M_{\perp op}|\sin(\varphi_{||}-\varphi_{\perp op})$$
$$\mp 2\cos^2(\theta)|M_{\perp ip}||M_{\perp op}|\sin(\varphi_{\perp op}-\varphi_{\perp ip})$$

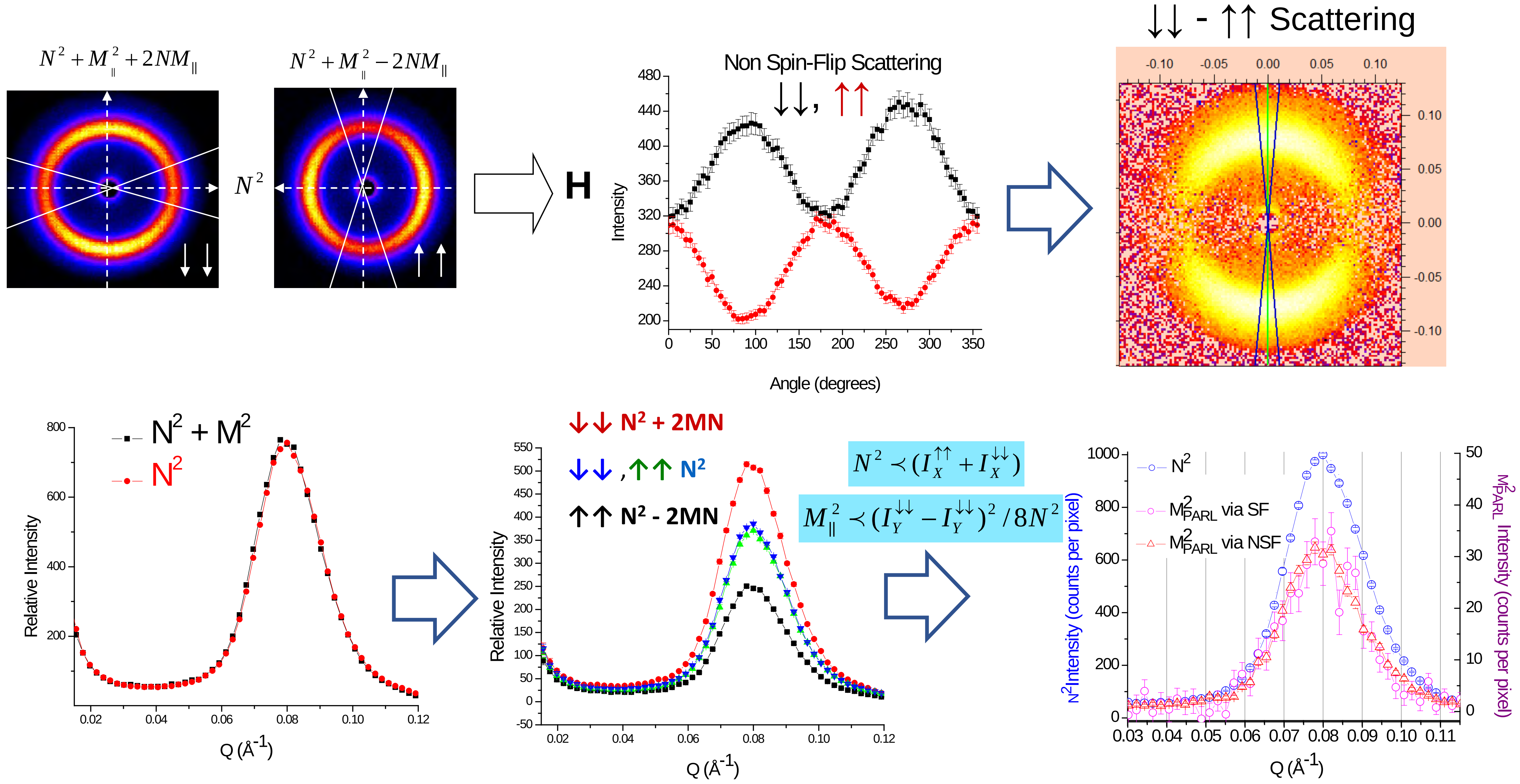
R. M. Moon, T. Riste, and W. C. Koehler, Physical. Review 181, 920 (1969)
A. Wiedenmann *et al*., Physica B 356, 246 (2005)
A. Michels and J. Weissmüller, Rep. Prog. Phys. 71, 066501 (2008)
K. Krycka *et al*., J. Appl. Cryst. 45, 554 (2012)

Coordinate Axes Simplification ($p \perp n$)



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

Non Spin-Flip Scattering at 1.2 Tesla, 200 K



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

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

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

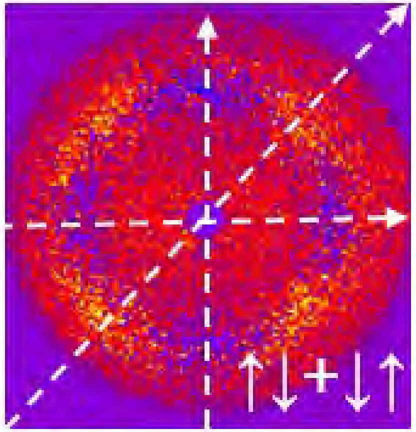
$$\pm 2sin(\theta)\cos(\theta)|M_{||}||M_{\perp op}|\sin(\varphi_{||}-\varphi_{\perp op})$$

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

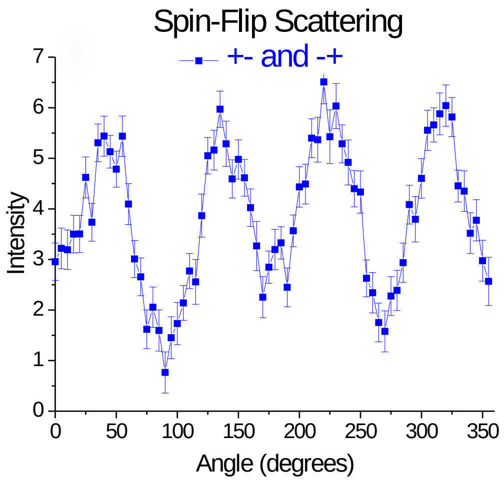
$$M^2_{PARL} = I^{\uparrow\downarrow,\downarrow\uparrow}_{45^o} - 1.25 M^2_{PERP}$$

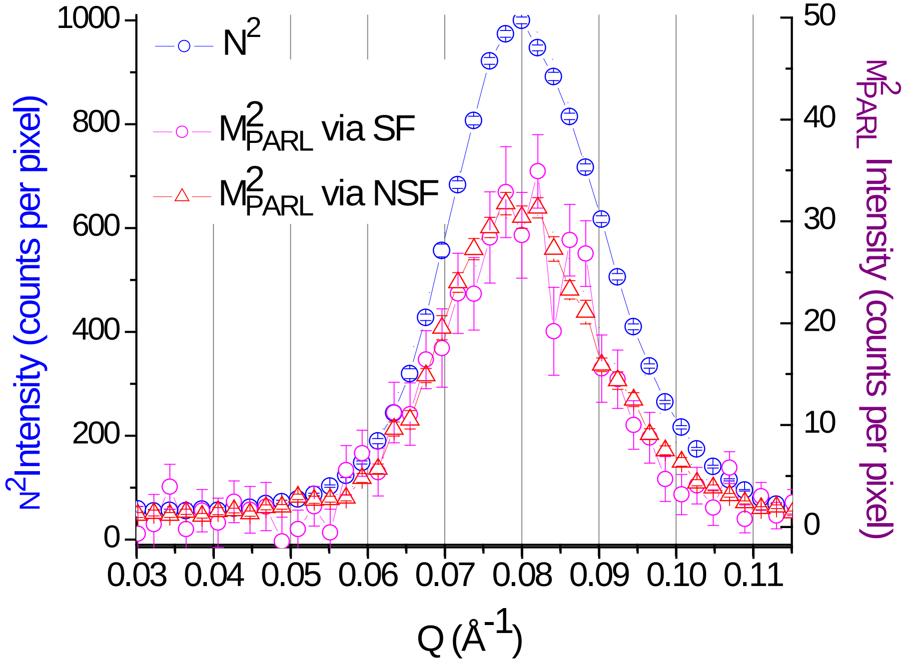
$$M^2_{PERP} = (I^{\uparrow\downarrow,\downarrow\uparrow}_X + I^{\uparrow\downarrow,\downarrow\uparrow}_Y)/3$$

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



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

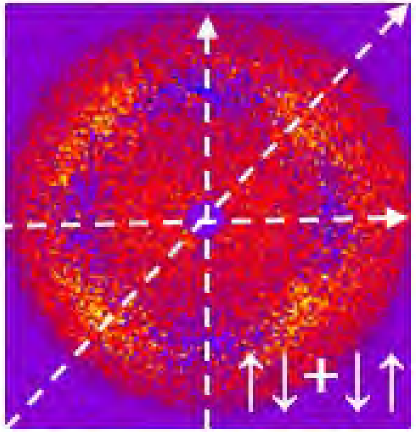




Note Magnetic / Nuclear ~ 0.03

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

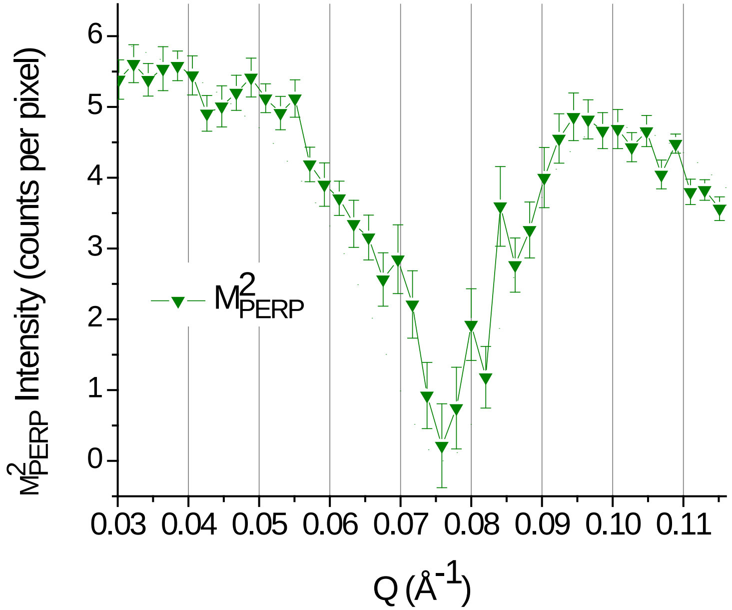
$$M^2_{\perp OP} \qquad M^2_{\parallel} + 1.25(M^2_{\perp IP} + M^2_{\perp OP})$$

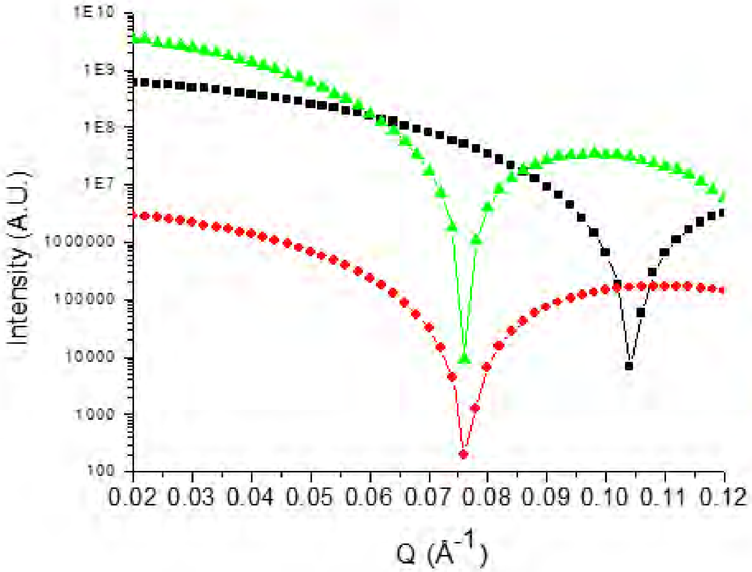


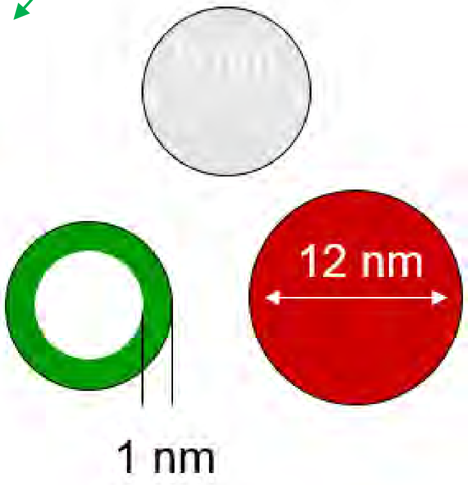
$$M^2_{\perp IP} + M^2_{\perp OP}$$

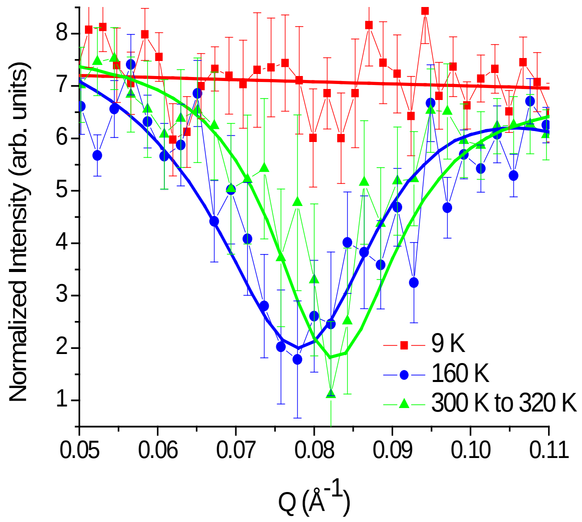
Dip at 0.075 Å^{-1} is reminiscent of spherical scattering

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



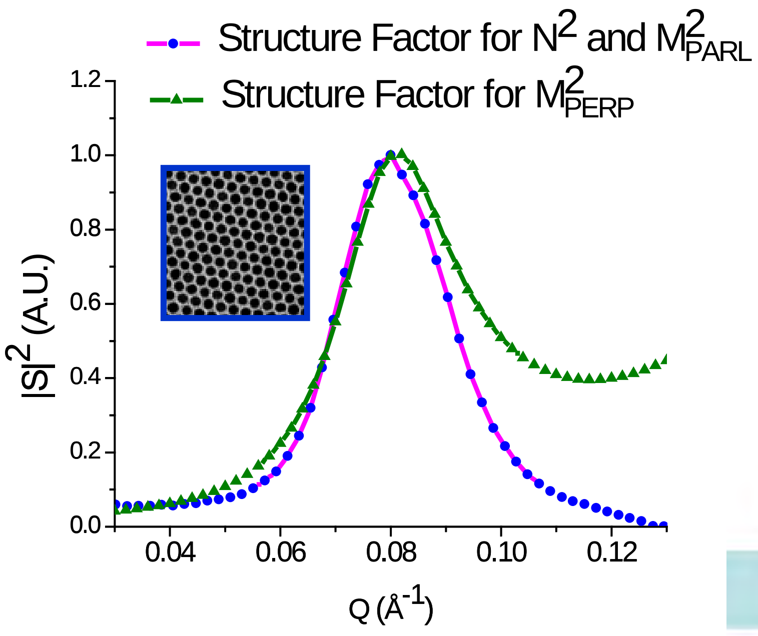






Canted magnetic shell changes with temperature

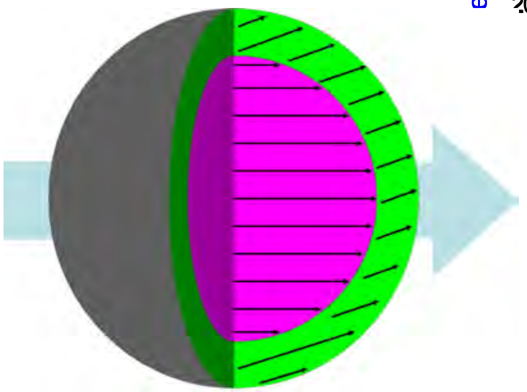
Putting It Together

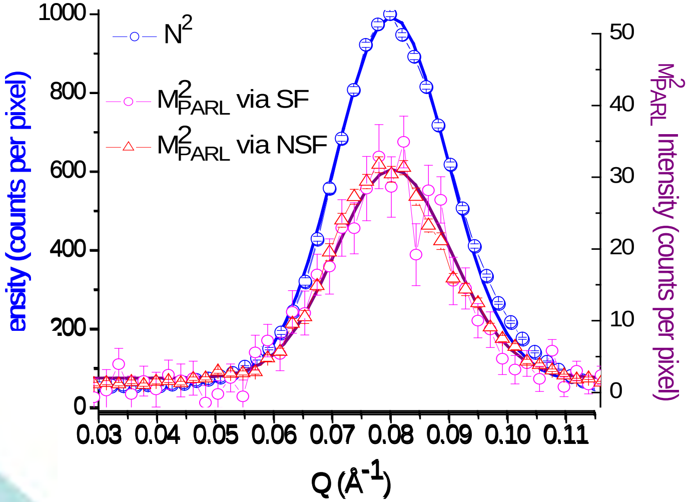


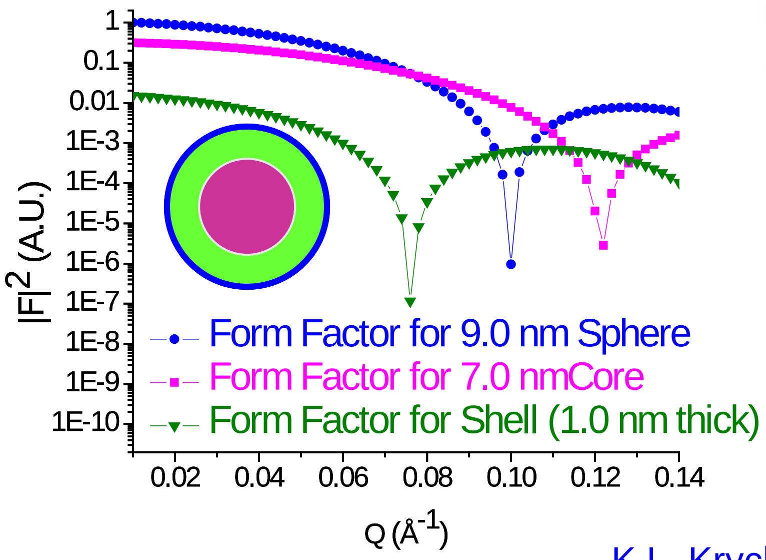
ρ_N = 6.97E-6 Å^{-2}

ρ_M = 1.46E-6 Å^{-2}

(513 emu / cc)





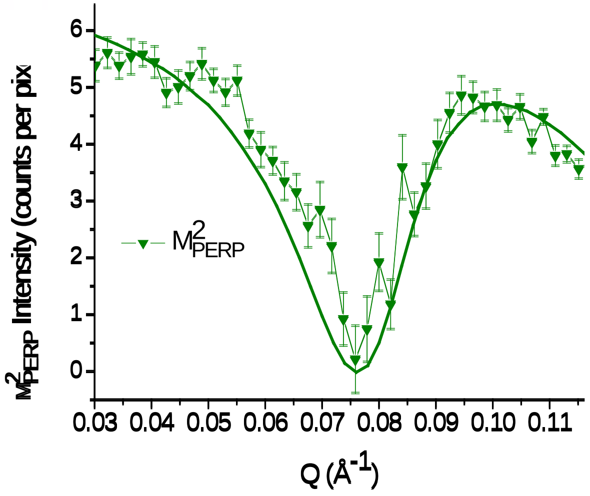


Modeled Diameters:

Sphere 9.0 nm

Ferrimagnetic core 7 nm

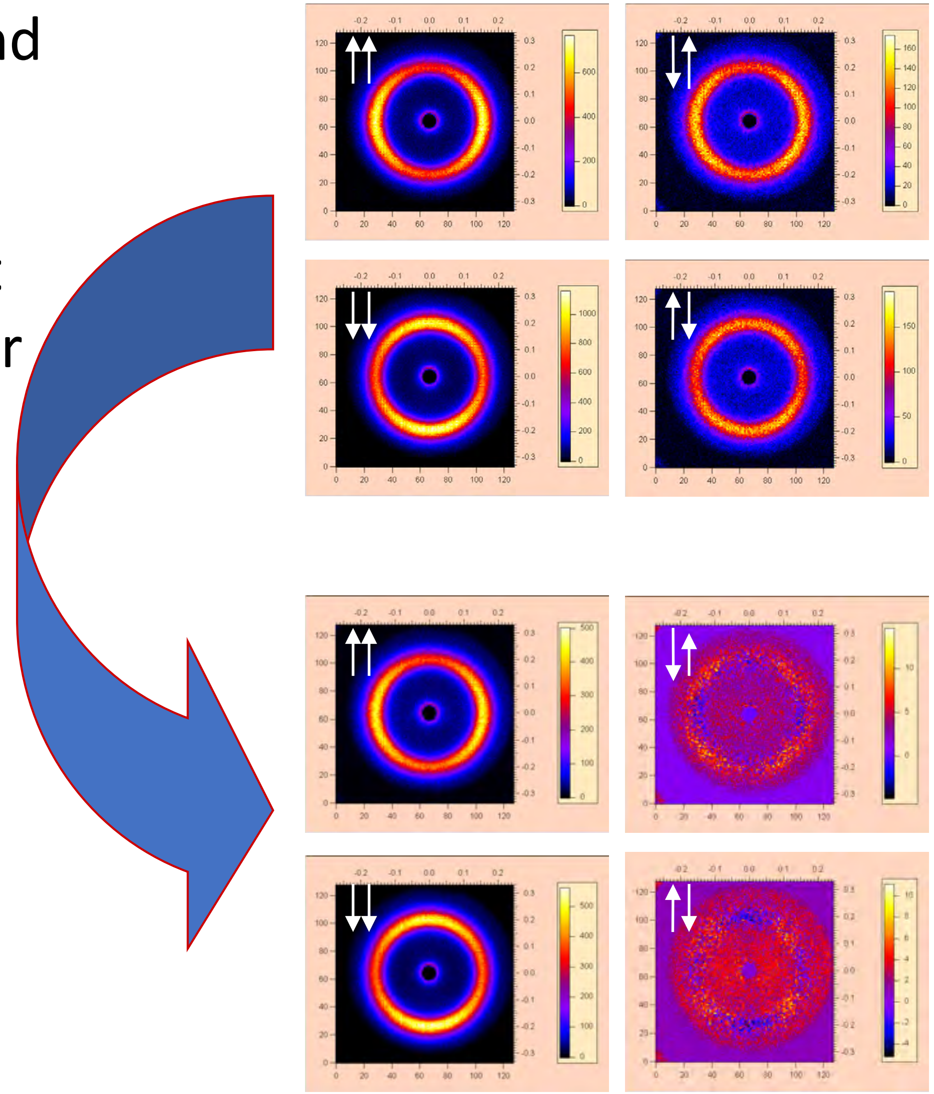
Canted shell 7 to 9 nm (±0.2 nm)



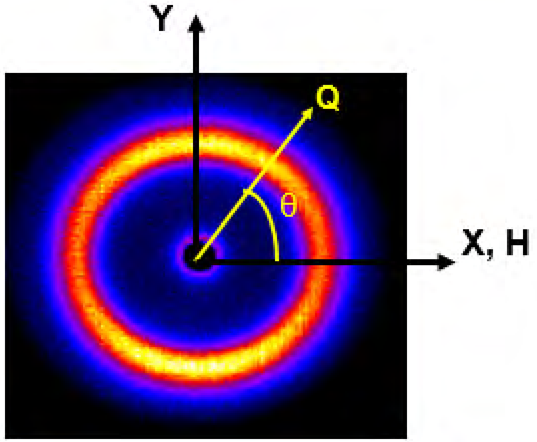
K.L. Krycka et al., *Phys. Rev. Lett.* 104 207203 (2010)

Polarization Efficiency Corrections Required

- Spin leakage from supermirror, polarizer, and ^3He analyzer (time-dependent) are all important
- Sample itself can be depolarizing, and must be measured and corrected for if looking for small signals
- Typically most important for small, 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



Revisiting the Cross-Terms (p ⊥ n-beam)



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

$$-2cos(\theta)sin^3(\theta)|M_{||}||M_{\perp ip}|cos(\varphi_{||}-\varphi_{\perp ip})$$
$$\pm 2\,sin(\theta)cos(\theta)|N||M_{\perp ip}|cos(\varphi_N-\varphi_{\perp ip})$$
$$\mp 2sin^2(\theta)|N||M_{||}|cos(\varphi_N-\varphi_{||})$$

Used this previously to get $M_{||B}$

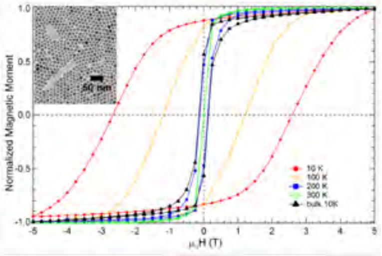
$$I^{+-,-+} = |M_{\perp op}|^2 + cos^4(\theta)|M_{\perp ip}|^2 + sin^2(\theta)cos^2(\theta)|M_{||}|^2$$

Expect 2:1 ratio along X and Y directions in spin-flip

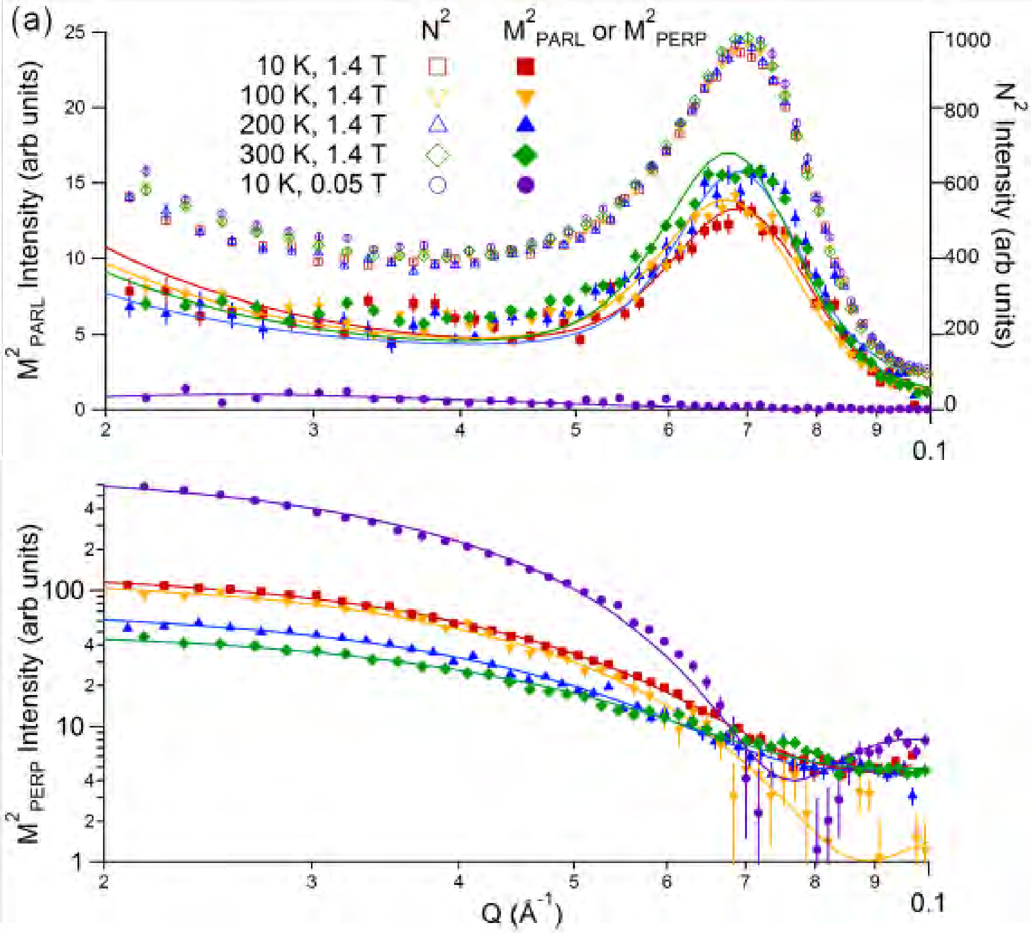
$$-2sin(\theta)cos^3(\theta)|M_{||}||M_{\perp ip}|cos(\varphi_{||}-\varphi_{\perp ip})$$
$$\pm 2sin\,(\theta)cos\,(\theta)|M_{||}||M_{\perp op}|sin(\varphi_{||}-\varphi_{\perp op})$$
$$\mp 2cos^2(\theta)|M_{\perp ip}||M_{\perp op}|sin(\varphi_{\perp op}-\varphi_{\perp ip})$$

Does not disappear win the sum of ↑↓ +↓↑

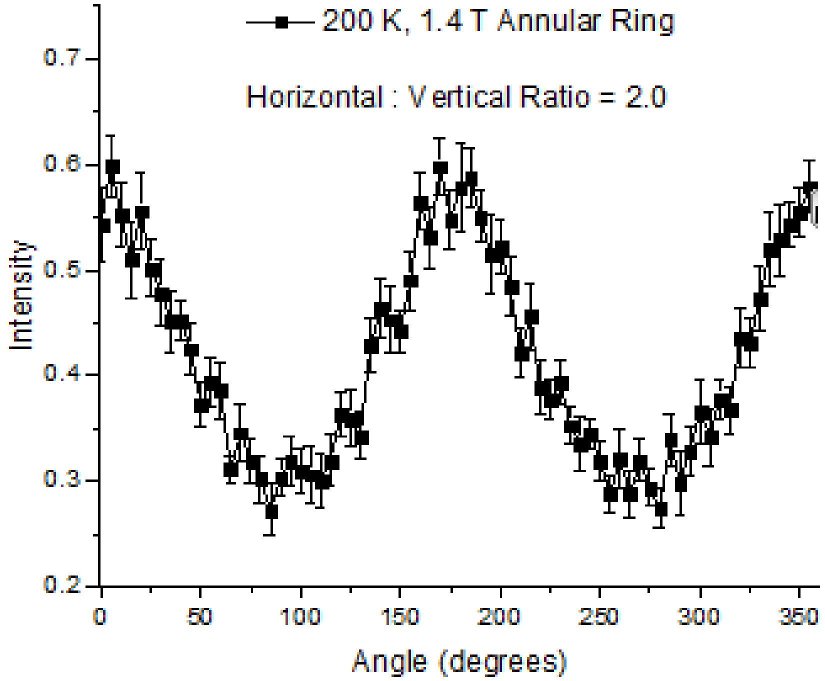
Canting Alone Doesn't Cause 2:1, X:Y Spin-Flip Deviation



10 nm $CoFe_2O_4$ Nanoparticles



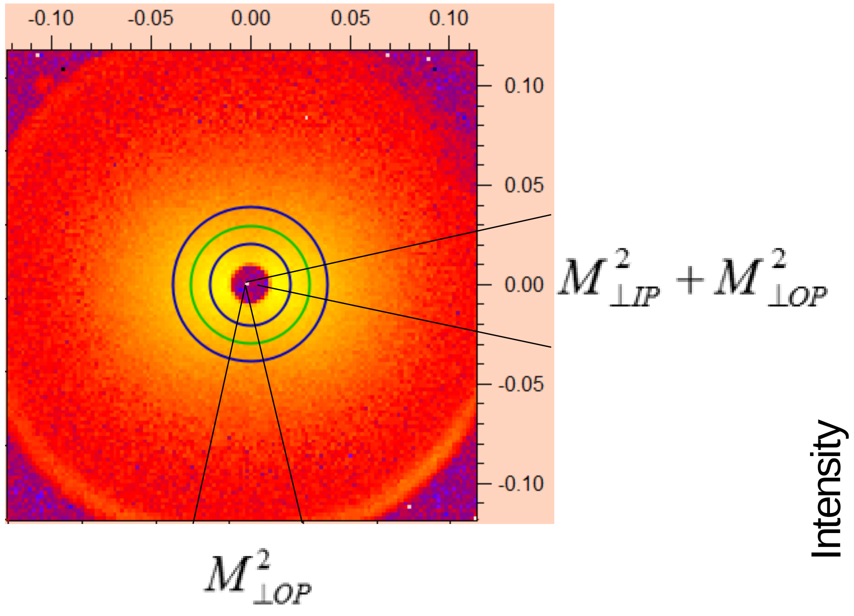
Condition	M^2_{PERP}	M^2_{PARL}	Measured θ	Modeled θ
10 K, 1.4 T	2.9± 0.2	13.5±0.5	33±2°	33±4°
100 K, 1.4 T	2.8±0.2	14.2±0.5	32±2°	32±4°
200 K, 1.4 T	1.5± 0.2	15.8±0.5	24±2°	26±4°
300 K, 1.4 T	0.86± 0.07	17.1±0.5	17±2°	17±4°

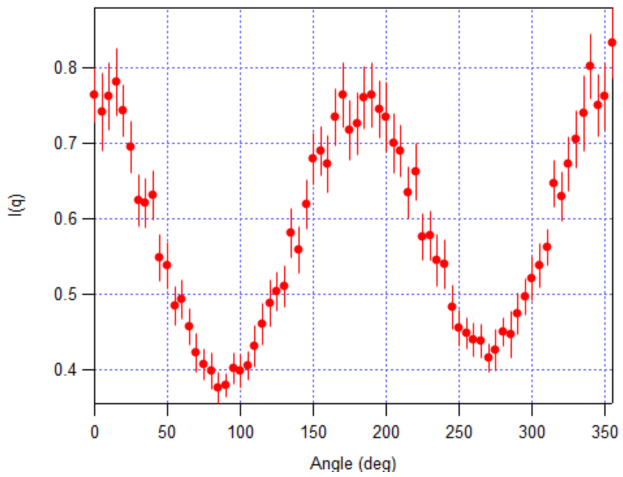


K. Hasz et al., Phys. Rev. B 90, 180405(R) (2014)

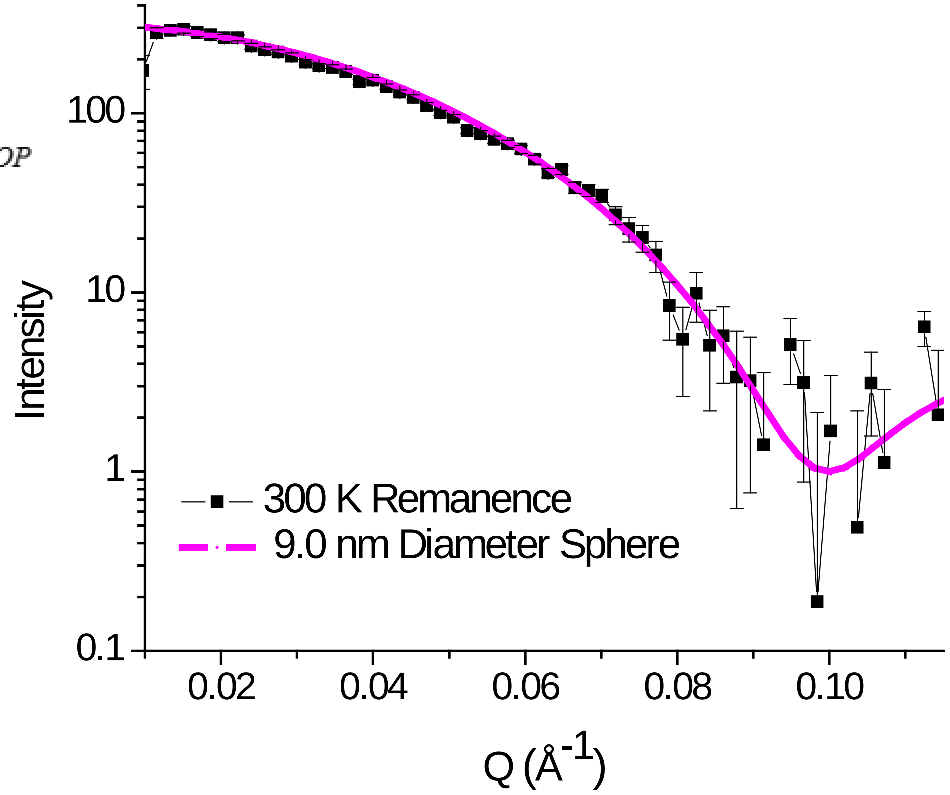
Revisiting the Cross-Terms: Near Zero Field

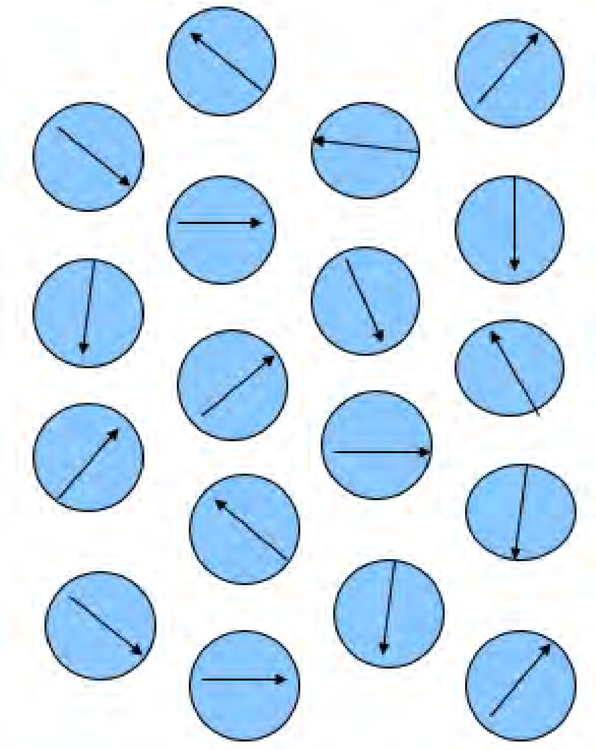
Expected 2:1 ratio





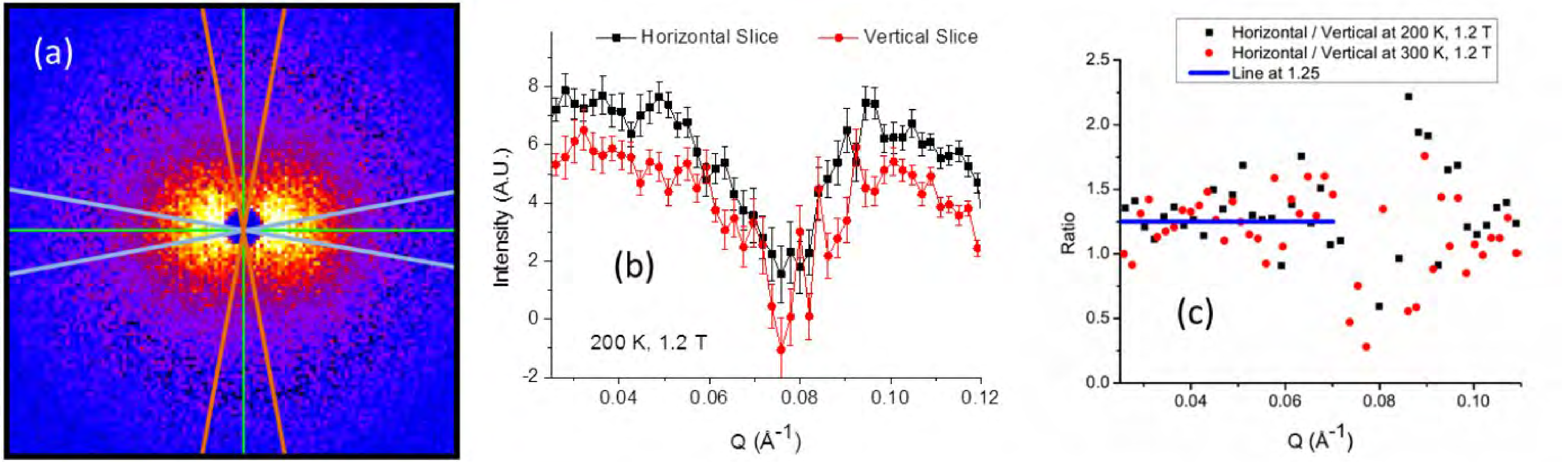
- Field and magnetic correlations eliminated at 300 K, 0.005 T





- Nanoparticles show **no shell features** and behave as uniform, ferrimagnetic 9 nm spheres randomly oriented in space. **Thus, shell is magnetic in origin.**

Revisiting the Cross-Terms: 1.2 Tesla Field



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

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

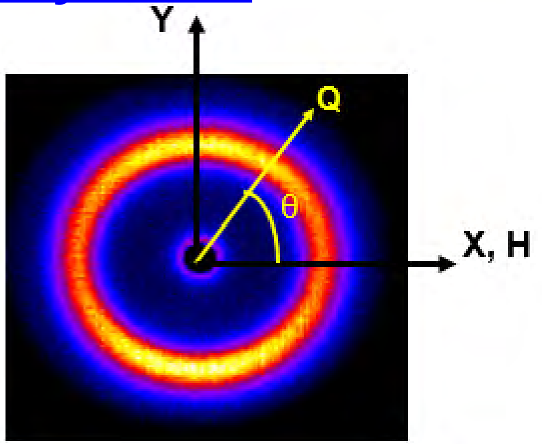
$$\pm 2\sin(\theta)\cos(\theta)|M_{||}||M_{\perp op}|\sin(\varphi_{||}-\varphi_{\perp op})$$

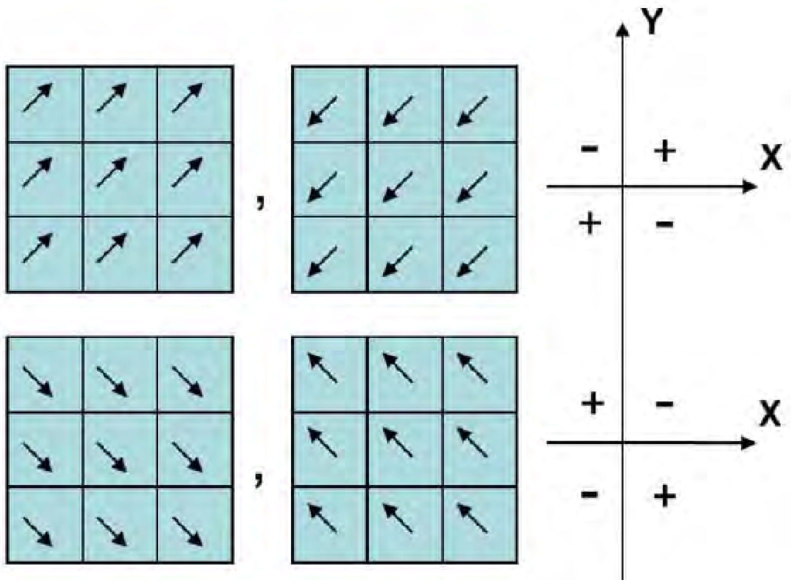
$$\mp 2\cos^2(\theta)|M_{\perp ip}||M_{\perp op}|\sin(\varphi_{\perp op}-\varphi_{\perp ip})$$

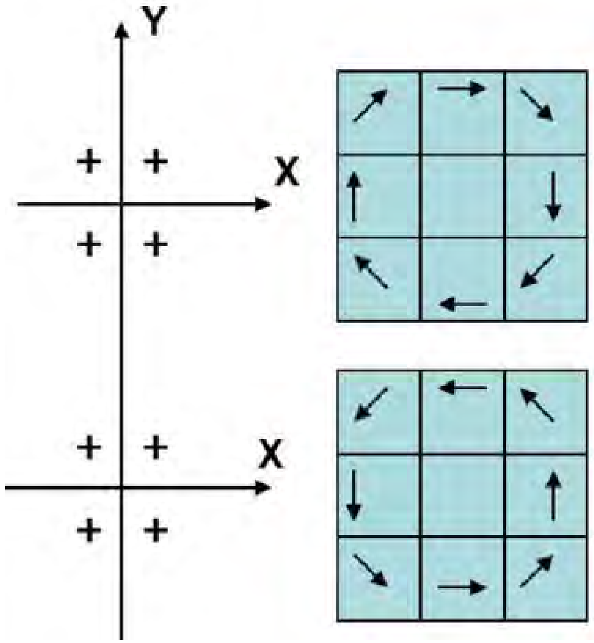
One explanation of the spin-flip deviation could be shell canting with average angles of 20 to 40 degrees (Phys. Rev. Lett. 113, 147203 (2014)).

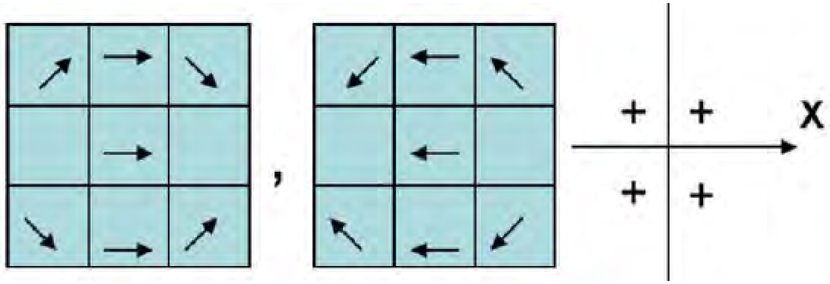
Impact of Cross-Term Depends on Symmetry of System

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







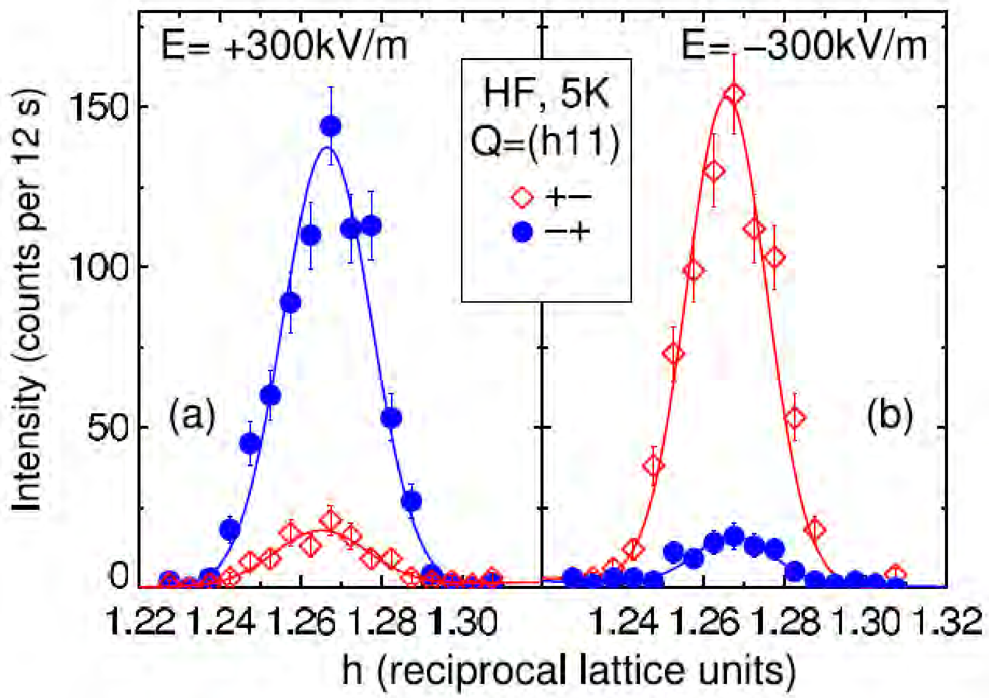


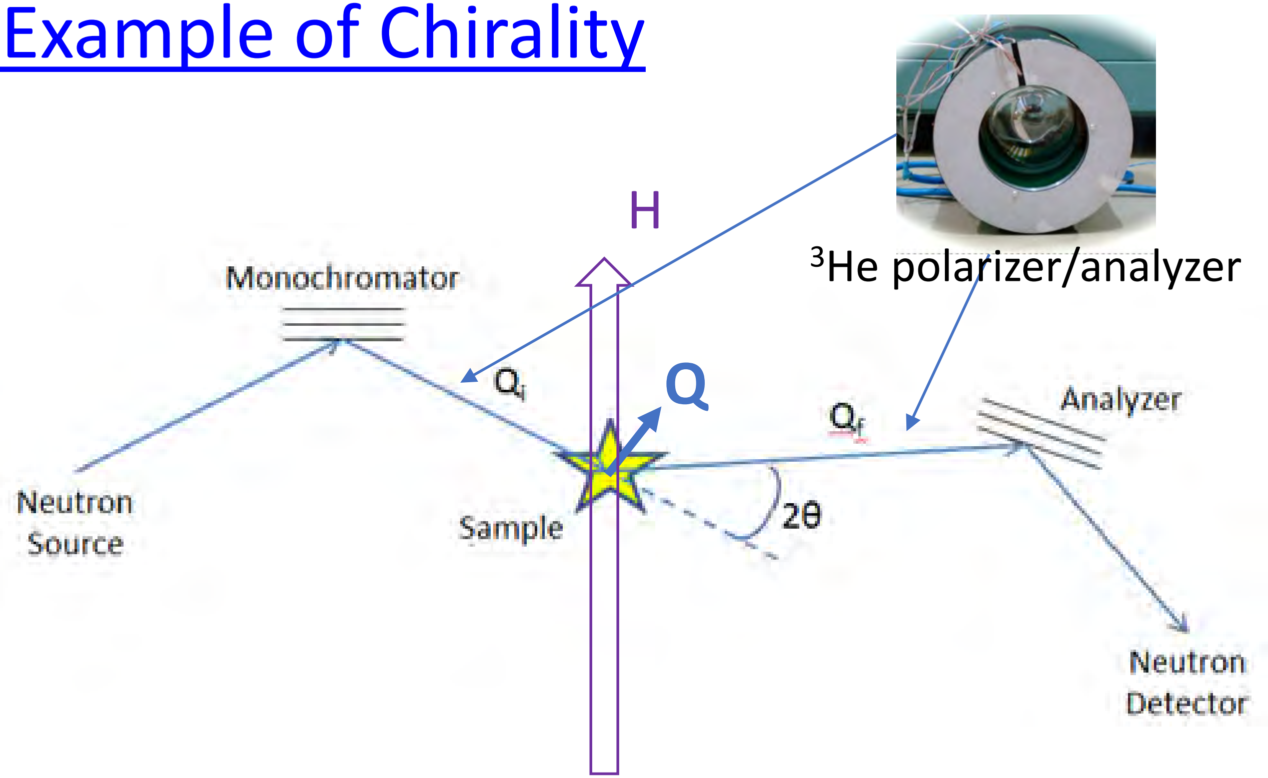
Random canting about field direction shouldn't result in 2:1 spin-Flip deviation

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

Other structures may have non-zero contribution

Diffraction Example of Chirality





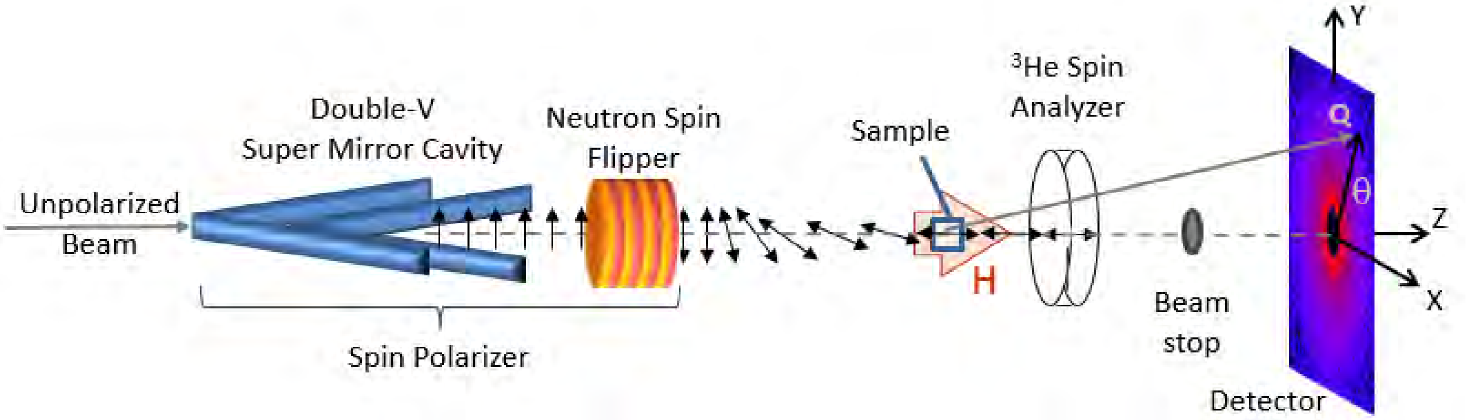
PRL **103,** 087201 (2009) PHYSICAL REVIEW LETTERS week ending 21 AUGUST 2009

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]

Consider **p ||** n-beam

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



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

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

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

M ⊥ B is entirely spin-flip

Increased statistics, but can't point too large of a field at ³He 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
$\|N\|^2$	1	0	0	1
$\|M_x\|^2$	$\sin^4(\theta)$	0	$\sin^2(\theta)\cos^2(\theta)$	$\sin^2(\theta)$
$\|M_y\|^2$	$\sin^2(\theta)\cos^2(\theta)$	0	$\cos^4(\theta)$	$\cos^2(\theta)$
$\|M_z\|^2$	0	0	1	1
$2\|N\|\,\|M_x\|\,\overline{\cos}(\varphi_N-\varphi_{M_x})$	0	1	0	0
$-2\|N\|\,\|M_y\|\,\overline{\cos}(\varphi_N-\varphi_{M_y})$	0	1	0	0
$-2\|M_x\|\,\|M_y\|\,\overline{\cos}(\varphi_{M_x}-\varphi_{M_y})$	$\sin^3(\theta)\cos(\theta)$	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

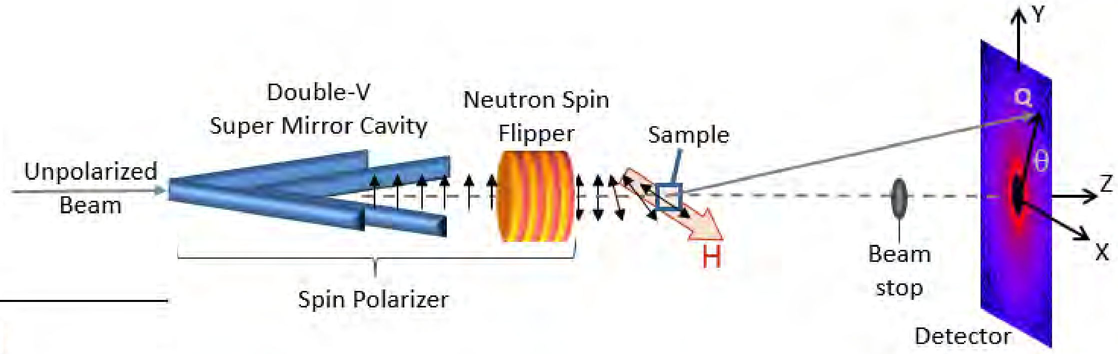
Terms for $\hat{\mathbf{p}} \parallel \hat{\mathbf{n}}$. The choice of X and Y axes within the plane $\perp \hat{\mathbf{n}}$ is arbitrary.

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

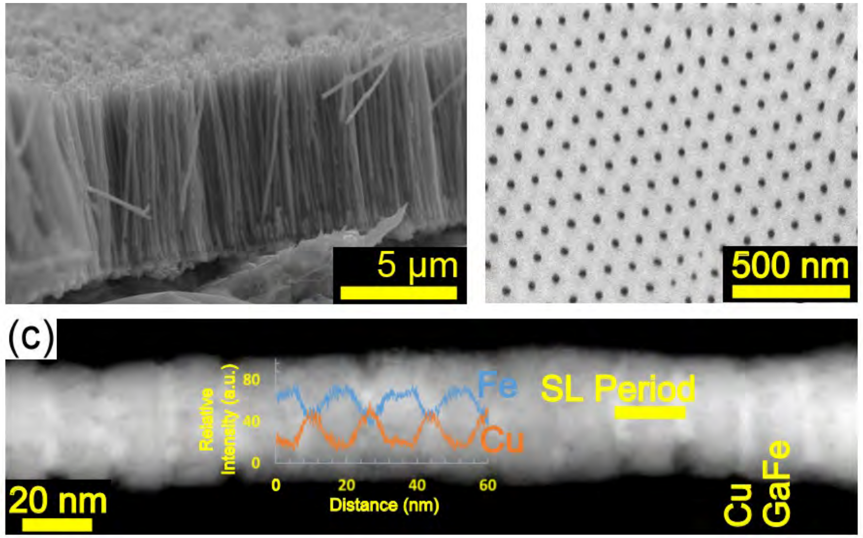
↓-↑ would be same as ↓ ↓ - ↑ ↑

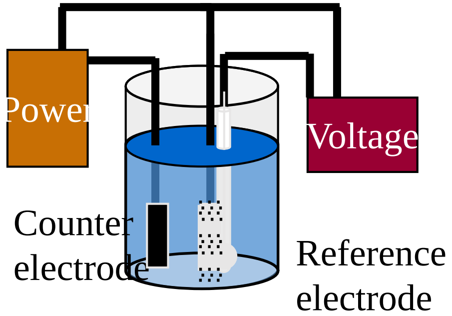
If want M | | B, then half-pol may be the best way to go.

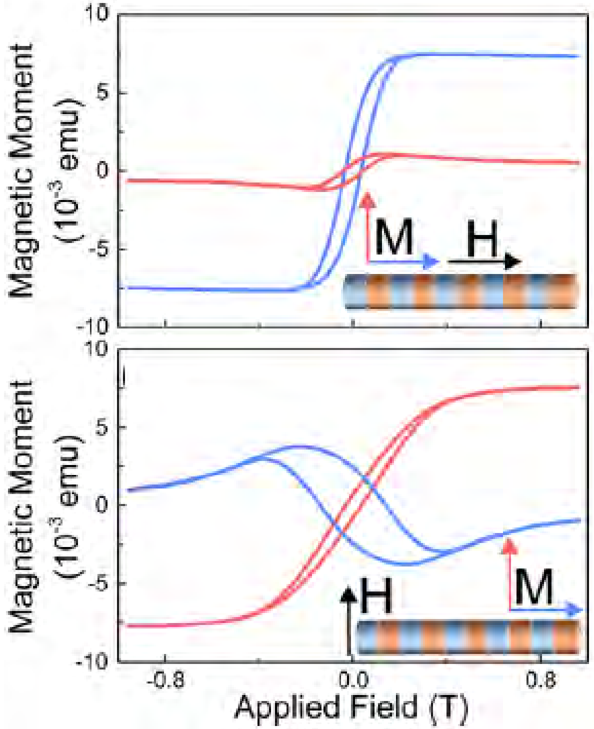
Rough Guidelines To Approaching Polarized Scattering

- Often start with unpolarized scattering to get a feel of overall scattering intensity
- If sample magnetically saturates, will check either (a) Intensity ⊥ B and Intensity || B to look for obvious magnetic scattering with or (b) high-field minus low field and/or changes in magnetism above and below blocking temperatures for either B ⊥ n-beam or B || beam configurations. The caveat is that moments ⊥ B may interfere with simple subtraction methods
- If still having trouble extracting an expected magnetic signal || B, then half-polarization can be helpful in boosting the M || B signal in the form of ↓ minus ↑ nuclear-magnetic cross-term
- If interested in in moments ⊥ B, can try 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 (a total factor of 4)
- If add 3He 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 => factor of 16. yet, if spin-flip is desired, won't have to count as long to overcome (typical) dominant structural scatting.
- Often bias the non spin-flip to 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.

Nanowire Example: $Ga_{20}Fe_{80}$ (11.6 nm) / Cu (5.0 nm) × 350

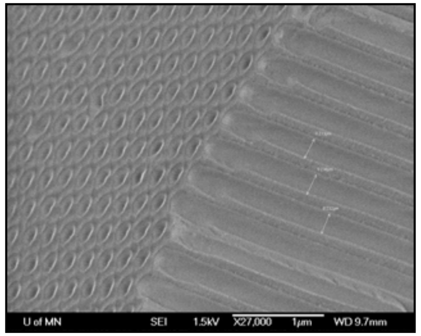


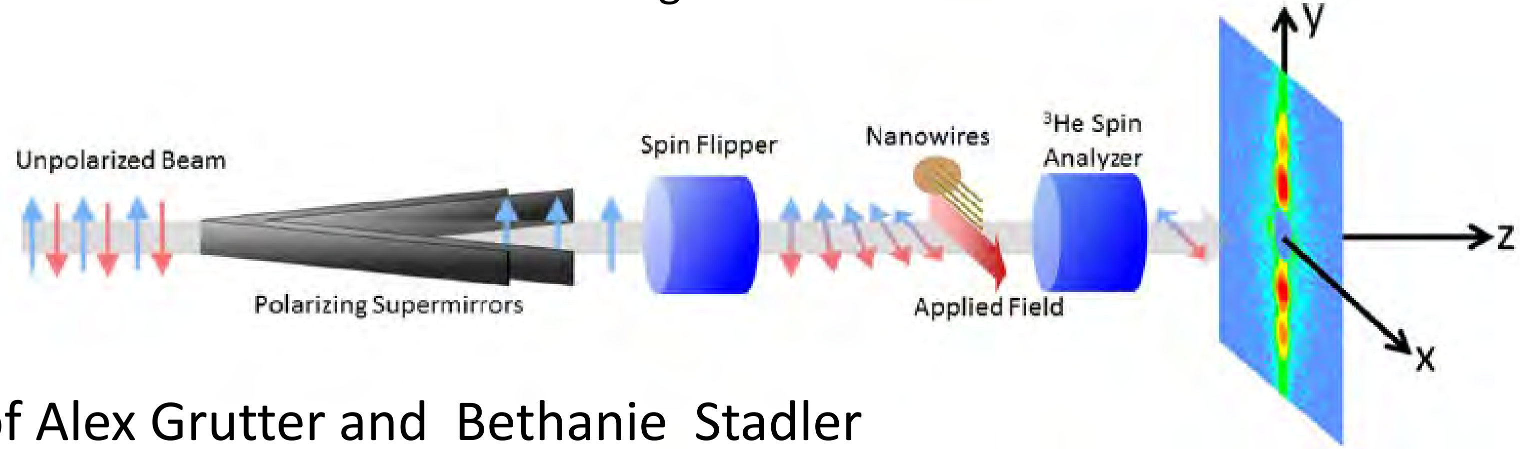




Electroplate Cu and GaFe segments Into the pores

- Al_2O_3 nanopores impose uniform diameter and interwire spacing
- Achieve uniform thickness over large areas

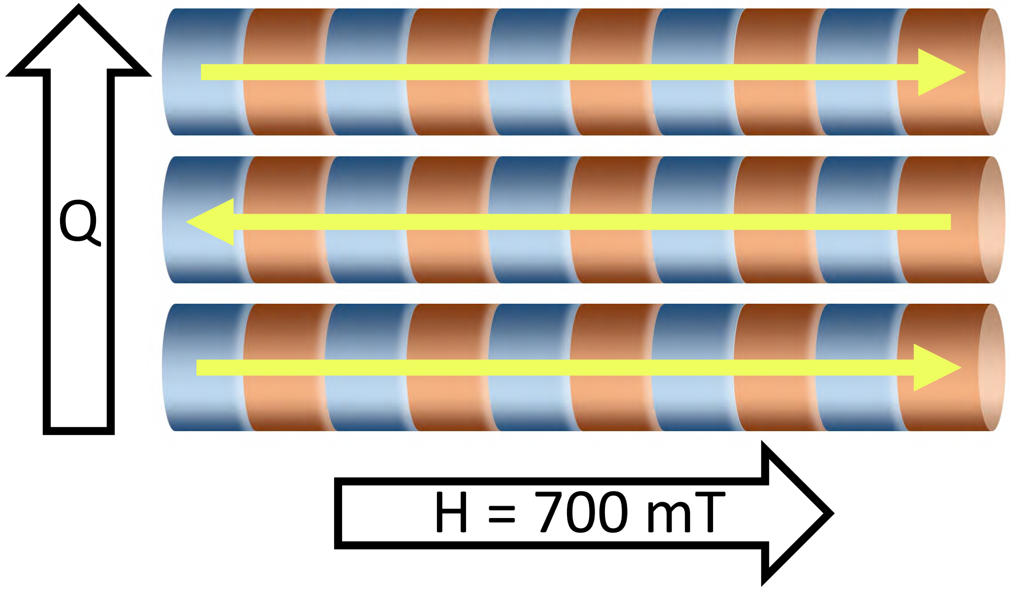


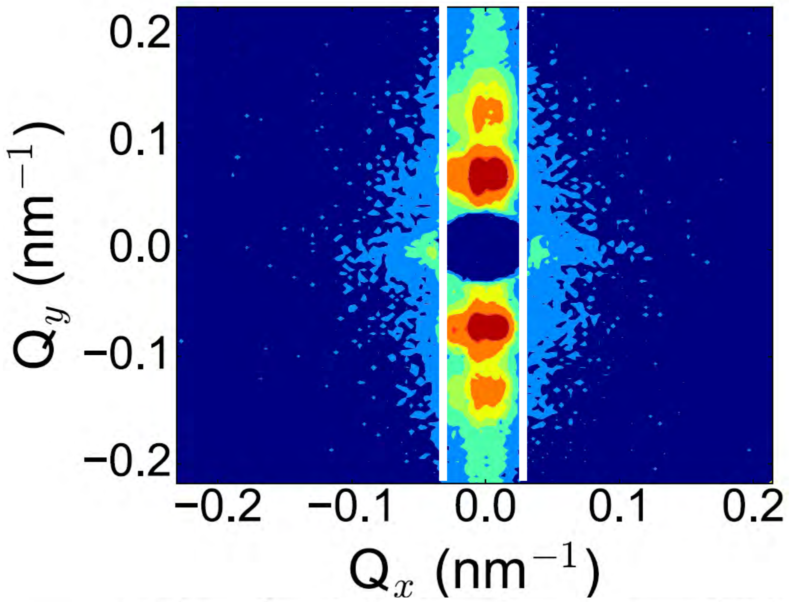


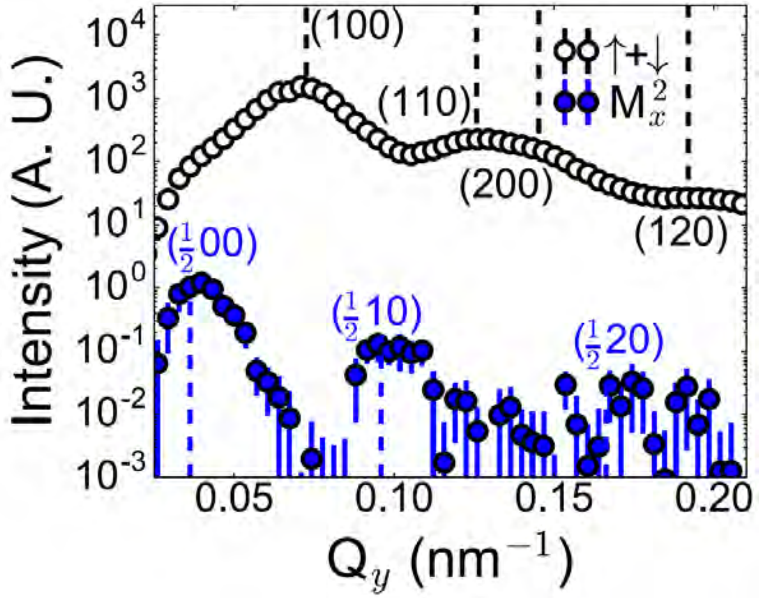
Hybrid SANS/Re-flectivity Example

Many images courtesy of Alex Grutter and Bethanie Stadler

High Field Interwire Scattering





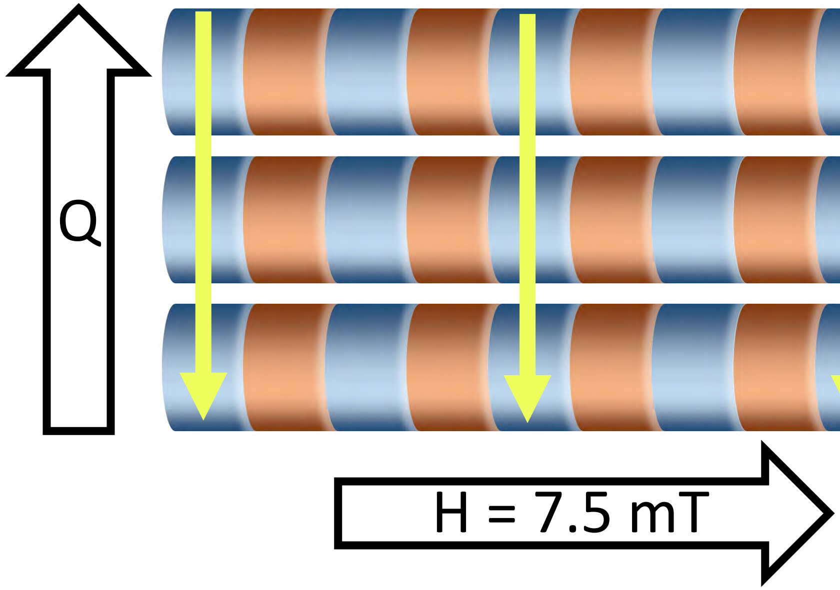


Non Spin-flip Scattering

↑↑ + ↓↓ = Structural Peaks
↓↓ -↑↑ (or ↓-↑)= Magnetic Peaks

AFM ordering of magnetization along the field between wires

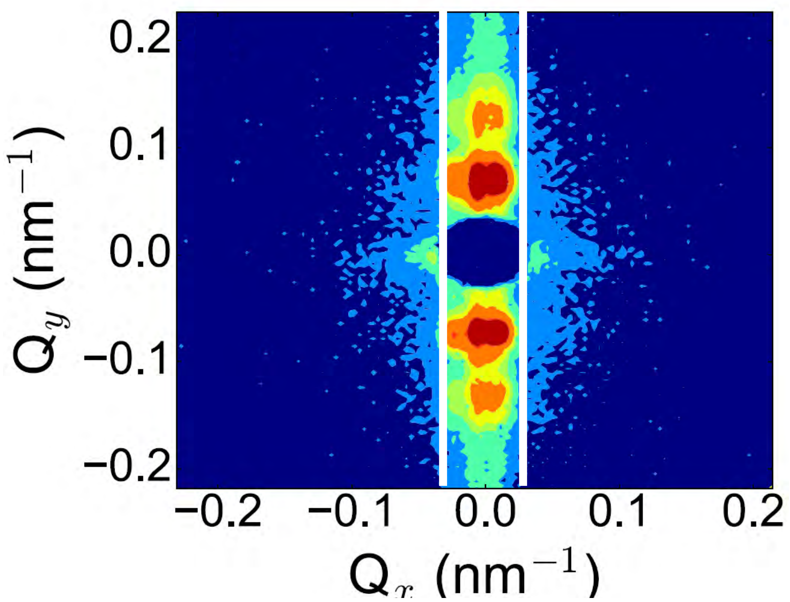
Low-Field Interwire Scattering

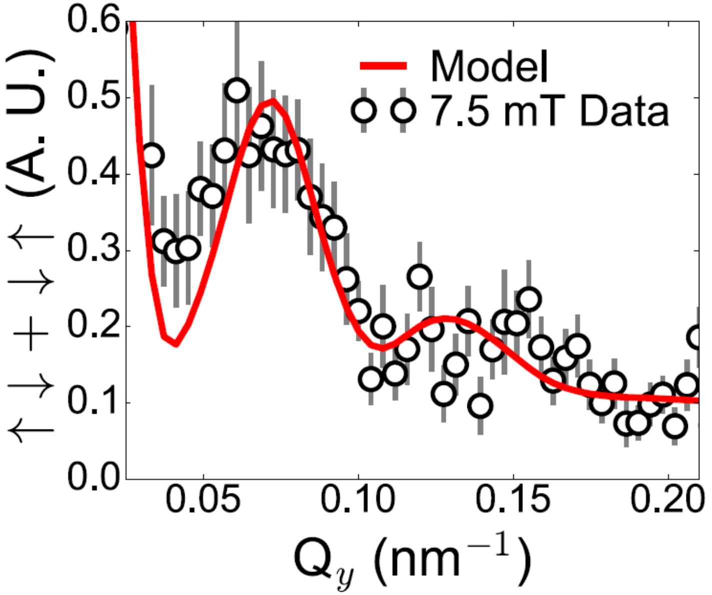


Spin-flip Scattering

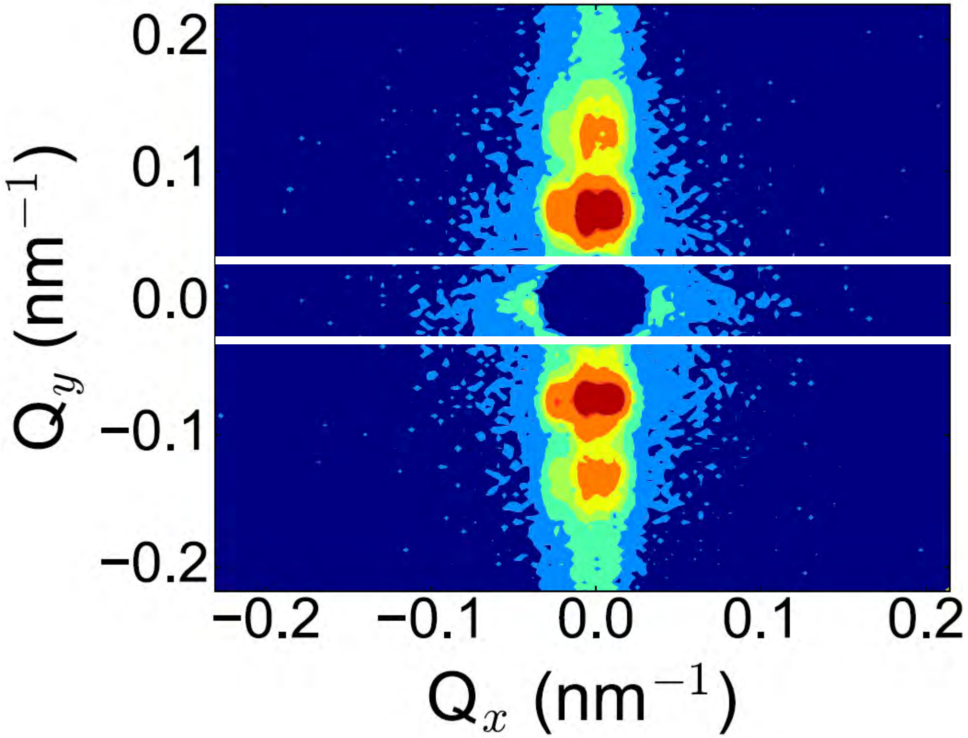
↑↓ + ↑↓ = $M_\perp$ peaks

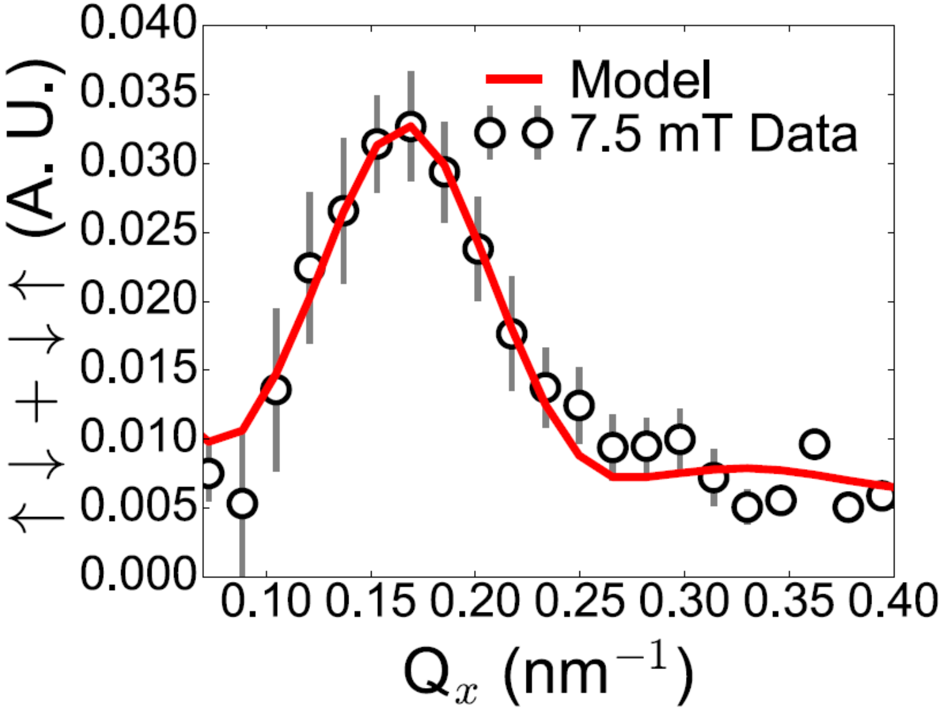
In-plane magnetization aligns ferromagnetically between wires

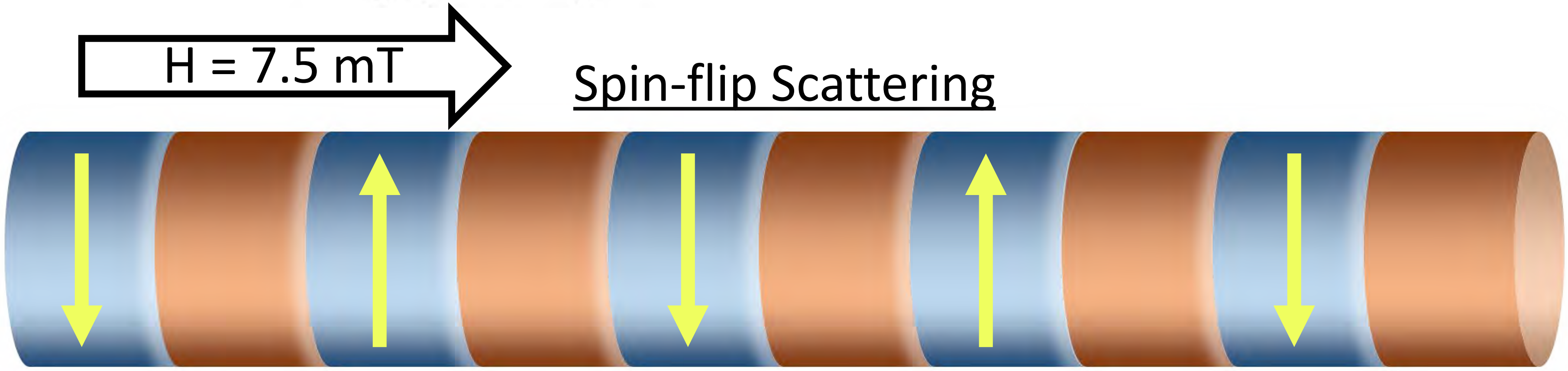




Low Field Intersegment Scattering





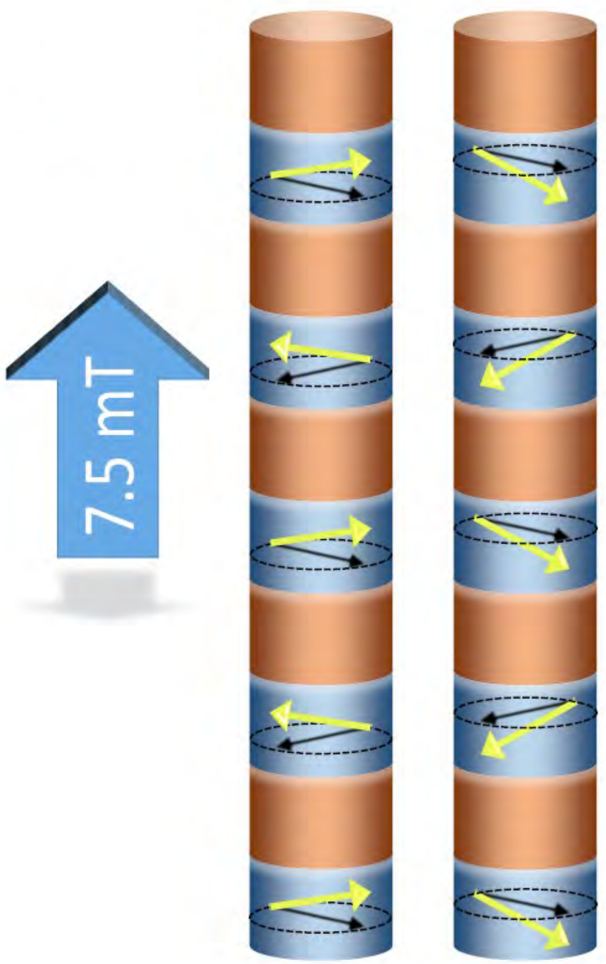


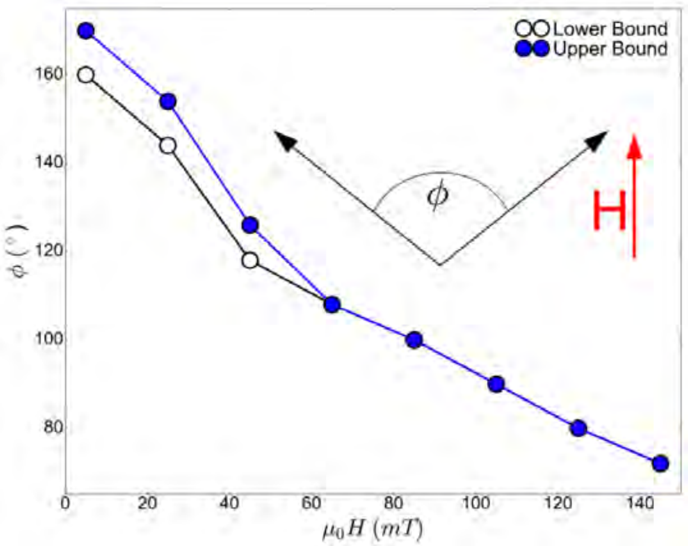
True AFM or Fan Structure?

But wait! There are ↑↓ peaks along:
Q_X = 0
AND
Q_Y = 0

- AFM order of $M_{in\text{-}plane}$ means there NO net moment in-plane
- Therefore NO spin-flip scattering along Q_X = 0
- Must be ***some*** canting away from pure AFM

Fan Structure!



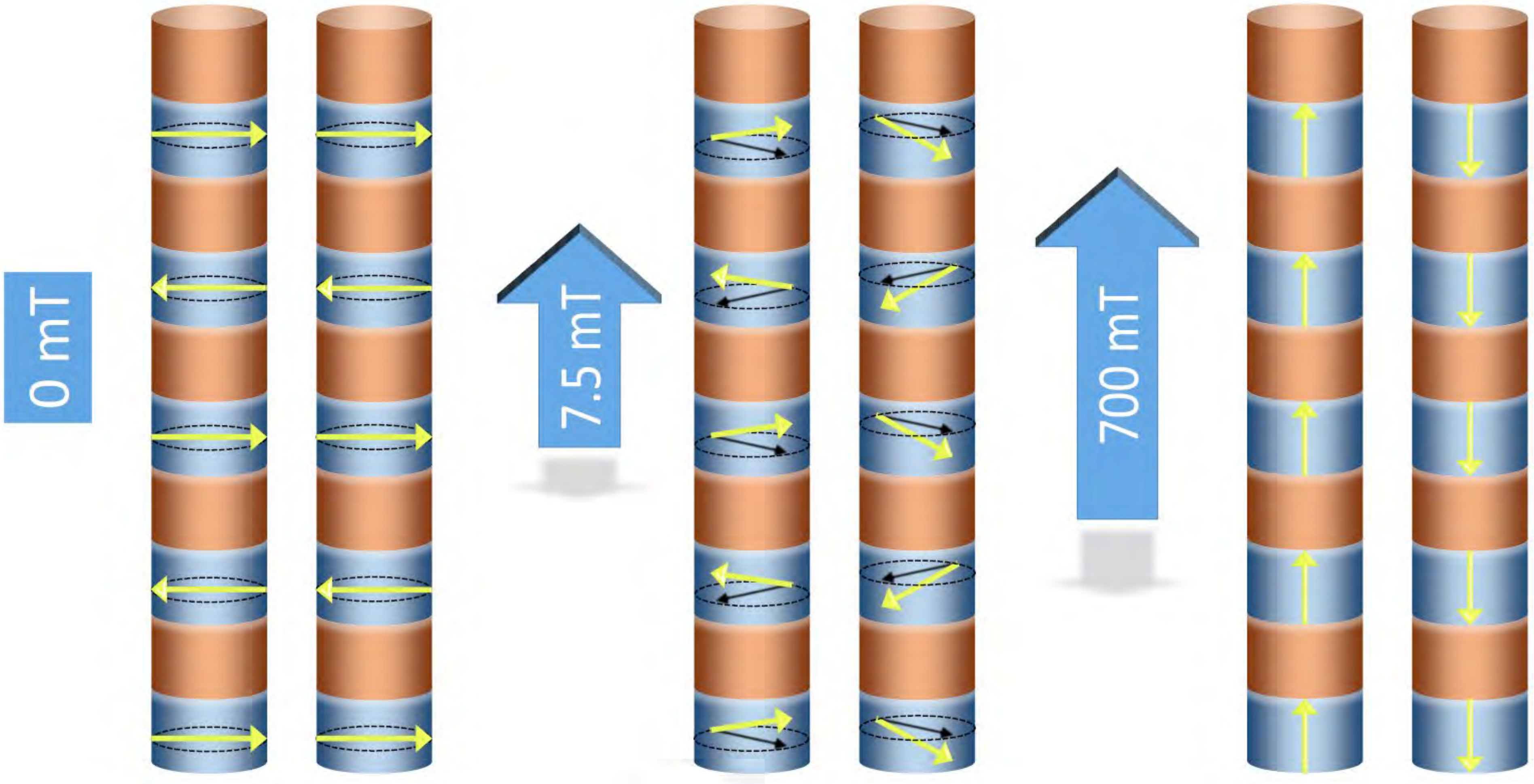


Even small in-plane field projection yields significant canting

1.3° canting in this case
≈ 0.15 mT in-plane field

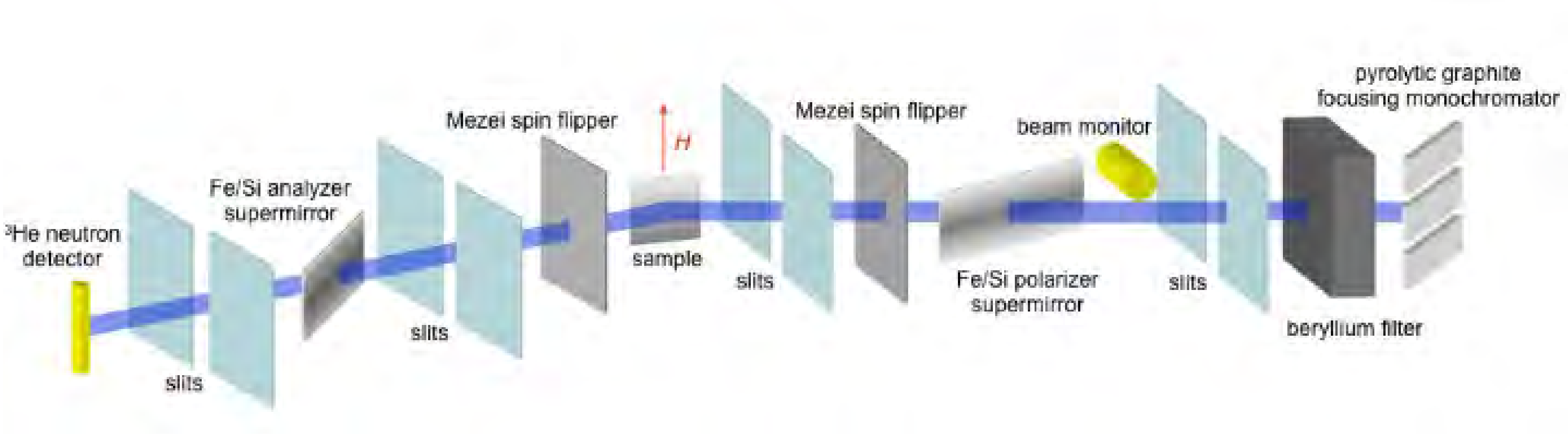
Courtesy of Alex Grutter

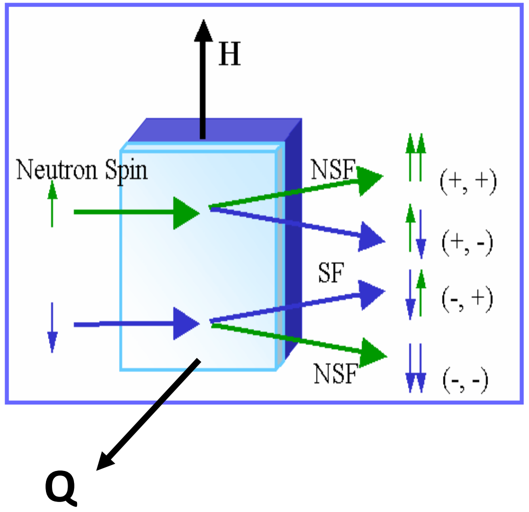
Field Dependent Structures



A. J. Grutter *et al.,* ACS Nano 11, 8311-8319 (2017)

Polarized Reflectometry





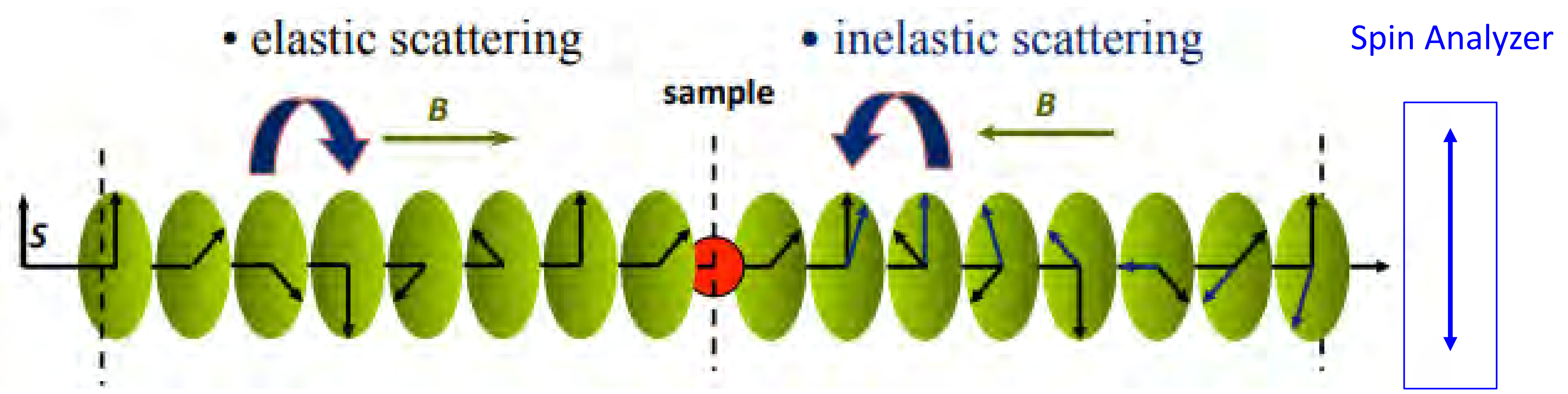
- NSF measures nuclear and M || H scattering, SF measures M $\perp$ H scattering averaged within the sample plane

- Additionally, magnetic reference layers with polarized beam serve as a means to obtain phase information and extract non-ambiguous scattering density profiles. Similar to hydrogen-deuterium substitution or changing substrate material, but these measurements are performed on the *same* sample.

Majkrzak C F and Berk N F 1995 *Phys. Rev.* B **52** 827–30
de Hann V O and van Well A A 1995 *Phys. Rev.* B **52** 831–3
Majkrzak C, Berk N, Krueger S, Dura J, Tarek M, Tobias D, Silin V, Meuse C, Woodward J and Plant A 2000 *Biophys. J.* 79 3330–40
Majkrzak C F, Berk N F and Perez-Salas U F 2003 *Langmuir* 19 7796–810

Neutron Spin Echo

Image from Antonio Farone's 2012 NCNR Summer School Talk

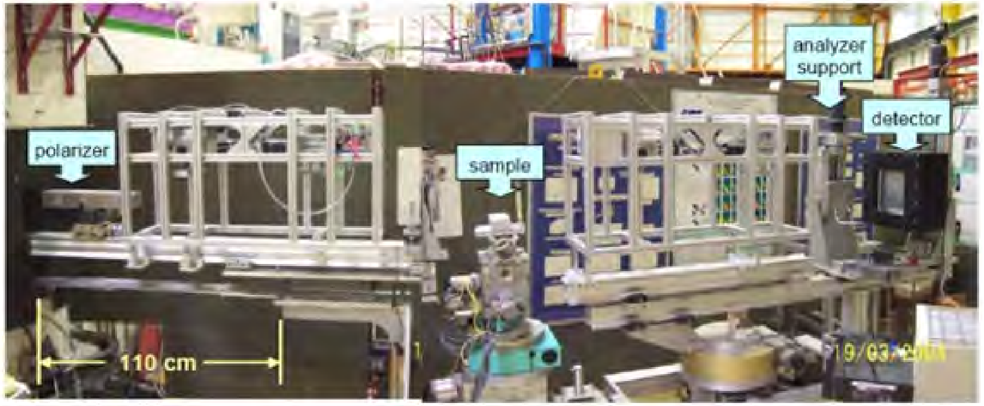


- Start with polarized neutrons; apply a B-field in the perpendicular direction
- Neutrons precess with Larmor frequency based on applied field strength (wavelength independent, although the time spent in the field changes slightly)
- Set-up to "wind" and "unwind" with the same number of rotations for elastic scattering (echo)
- Inelastic scattering causes changes in velocity, which in turn causes the neutrons to partially lose their final polarization state

Neutron Spin Echo

Begin with F. Mezei, Z. Physik 255, 146 (1972)

Spin Tagging Demonstration[1]
on EVA at the ILL, France



[1]J. Major, H. Dosch, G.P. Felcher, *et al.*, Physica B (2003).
Also, M. Th. Rekveldt, *et al.*, Rev. Sci. Instr. (2005).
And R. Pynn, M.R. Fitzsimmons, *et al.*, Rev. Sci. Instr. (2005).

(Courtesy of Chuck Majkrzak)

- Changing applied field allow for changes in scattering as a function of time (based on neutron velocity) to be determined
- Sensitive to neutron velocity changes on the order of 10^{-4}, even for distribution of wavelengths
- Sensitive to small energy changes on the neV level
- Incoherent scattering, as usual, places 1/3 onto non spin-flip and 2/3 into spin-flip channels