Principles of X-ray and Neutron Scattering

Lecture 10: Neutron Polarization Analysis 15. 02. '24

Lectures by: Prof. Philip Willmott, Prof. Johan Chang and **Dr. Artur Glavic**

Course Outline

Monday	Tuesday	Wednesday	Thursday	Friday
Lecture 1 10-10h45 Philip	**Lecture 4** 10-10h45 Philip	**Lecture 7** 10-10h45 Artur	**Lecture 10** 10-10h45 Artur	**Lecture 13** 10-10h45 Johan
Lecture 2 11-11h45 Philip	**Lecture 5** 11-11h45 Philip	**Lecture 8** 11-11h45 Artur	**Lecture 11** 11-11h45 Artur	**Lecture 14** 11-11h45 Johan
Lunch - Mensa	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**
Lecture 3 13h00-13h45 Philip	**Lecture 6** 13h00-13h45 Philip	**Lecture 9** 13h00-13h45 Artur	**Lecture 12** 13h00-13h45 Artur	**Lecture 15** 13h00-13h45 Johan
		Exercise Class 14h30-16		Exercise Class 14h30-16

- X-ray scattering
- Neutron Scattering
- Resonant x-ray scattering

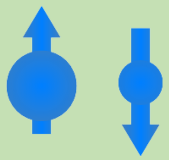
Neutron Lectures:

7: Neutrons & Scattering to Determine Structure
8: Inelastic Neutron Scattering to Investigate Dynamics
9: Magnetic Scattering
10: Neutron Polarization Analysis
11: Studying quantum matter for nanoscale applications
12: Neutron Instrument Development

Lecture 10: Magnetic Scattering and Neutron Polarization Analysis

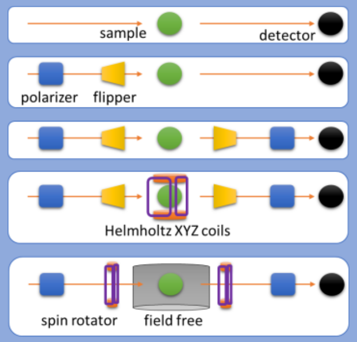
Theoretical Background

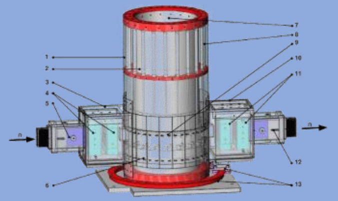
- Polarization considerations in scattering



Practical Implementation

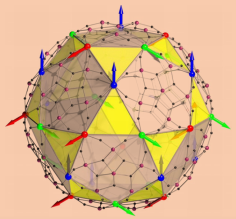
- Instruments with polarization (analysis)
- Polarization control devices
- Full polarimetry





Example Application

- Separating nuclear and magnetic inelastic scattering
- Molecular magnets



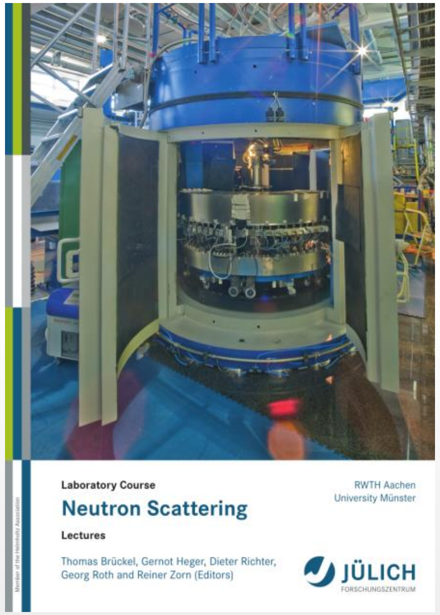
Further Reading

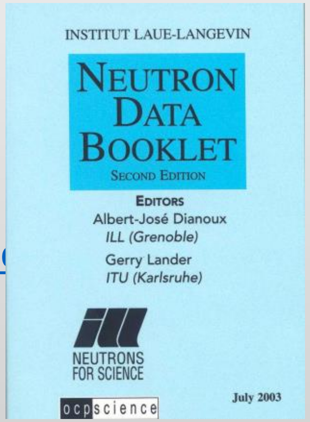
- **“Neutron Diffraction of Magnetic Materials”**
  Y. A. Izyumov, V. E. Naish, and R. P. Ozerov.
  Plenum Publishing Corporation, New York (1991)

- “Introduction to the Theory of Thermal Neutron Scattering”
  G. L. Squires
  Dover Publication (1978)
- “Theory of Neutron Scattering from Condensed Matter” Vol.I/II.
  S. W. Lovesey
  Oxford Science Publications (1984).

- “Neutron Scattering”
  T. Brückel, et al. (2012) / Available Open Access:
  https://juser.fz-juelich.de/record/136390/files/Schluesseltech_39.pdf

- “Neutron Data Book”
  Albert-José Dianoux and Gerry Lander
  https://www.ill.eu/fileadmin/user_upload/ILL/1_About_ILL/Documentation/NeutronDataBooklet.p





Polarization of a Neutron Beam

➔ A neutron is a spin-1/2 particle and its spin can be expressed as

$$\chi = a\chi_\uparrow + b\chi_\downarrow = a\begin{pmatrix}1\\0\end{pmatrix} + b\begin{pmatrix}0\\1\end{pmatrix}$$

$|a|^2$ and $|b|^2$ are the probabilities to be in the up or down state.

For normalization:

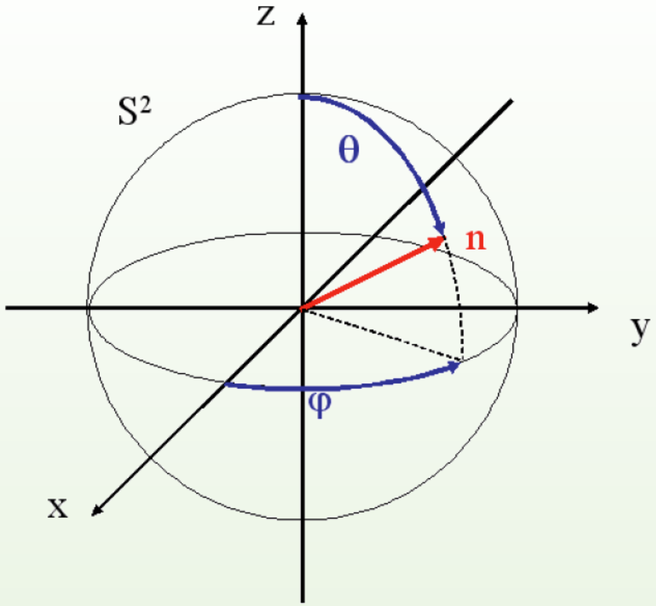
$$\chi^\dagger\chi = |a|^2 + |b|^2 = 1$$

➔ We can define the polarization of a single neutron as the unit vector pointing in the direction of the spin (see for example: Lovesey, Pauli matrices $\hat{\sigma}$ as before):

$$\boldsymbol{P} \equiv <\hat{\boldsymbol{\sigma}}>= \chi^\dagger\hat{\boldsymbol{\sigma}}\chi = Tr(\hat{\varrho}\hat{\boldsymbol{\sigma}}),$$

$$\text{where} \quad \hat{\varrho} = \chi\chi^\dagger = \begin{pmatrix}|a|^2 & ab^* \\ ba^* & |b|^2\end{pmatrix}$$

is a ***density matrix operator*** that gives the probability of a certain spin state.



➔ The following choice for *a* and *b* is a valid normalization:

$$a = \cos\frac{\theta}{2}e^{i\frac{\phi}{2}} \qquad b = \sin\frac{\theta}{2}e^{-i\frac{\phi}{2}}$$

➔ And we find a more physical representation (Bloch sphere):

$$\boldsymbol{P} = \begin{pmatrix}2\Re(a^*b)\\2\Im(a^*b)\\|a|^2-|b|^2\end{pmatrix} = \begin{pmatrix}\sin\theta\cos\phi\\\sin\theta\sin\phi\\\cos\theta\end{pmatrix} = \hat{\boldsymbol{n}}$$

Polarization of a Neutron Beam

➔ The polarization of a beam is then: $\boldsymbol{P} = \frac{1}{N}\sum_i \boldsymbol{P}_i = << \hat{\boldsymbol{\sigma}} >>_{beam}$,

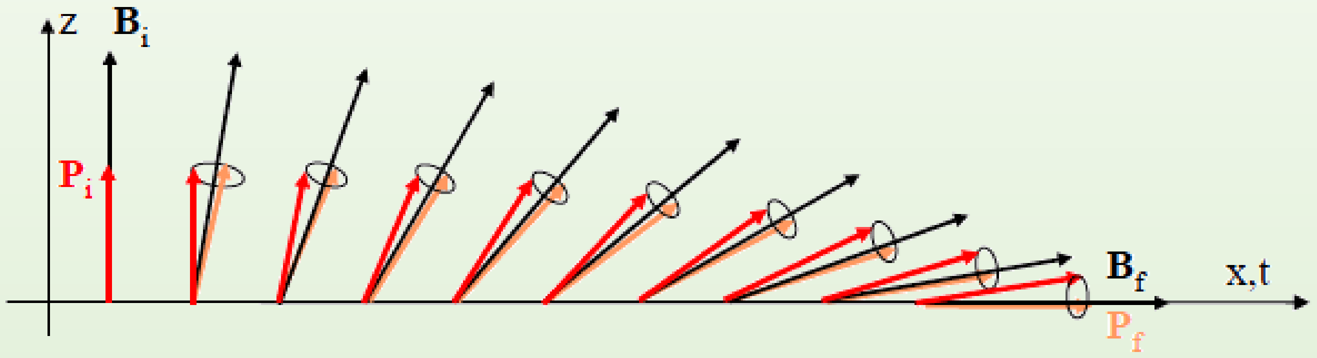
➔ A polarized beam can be manipulated by magnetic fields. This is known as ***Larmor precession:***

$$\frac{d\boldsymbol{P}(t)}{dt} = -\gamma_L(\boldsymbol{P}(t) \times \boldsymbol{B}).$$

➔ The precession frequency is $\omega_L = \frac{\gamma e}{m_p} B = \gamma_L B$ where $\gamma_L = 2\pi \cdot 2913 \frac{rad}{sG}$

➔ the direction parallel to the magnetic field is conserved. This implies that, if the magnetic field direction is changed with a frequency slow compared to the Larmor frequency, the polarization vector will follow.

adiabatic
field
change



The Blume-Maleyev Equations (from Lovesey)

➔ The most general expression for the cross-section, taking into account the neutron spin is:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\int dt \sum_{\sigma_i,\sigma_f} p_\sigma \langle\sigma_i|V^\dagger_{\boldsymbol{Q}}(0)|\sigma_f\rangle\langle\sigma_f,|V_{\boldsymbol{Q}}(t)|\sigma_i\rangle \exp(-i\omega t),$$

where $V_{\boldsymbol{Q}}(t) = \langle \boldsymbol{k}_f|V(t)|\boldsymbol{k}_i\rangle = N_{\boldsymbol{Q}}(t) + (\gamma r_0)\hat{\boldsymbol{\sigma}}\cdot \boldsymbol{M}_{\perp\boldsymbol{Q}}(t).$

Nuclear Structure Factor   Magnetic Interaction Vector

➔ The probability function p_σ can be expressed using the density matrix operator:

$$\hat{\varrho} = \sum_{\sigma_i,\sigma_f} p_\sigma|\sigma\rangle\langle\sigma|,$$

➔ With this, the sum and average can be expressed such as that

$$\sum_{\sigma_i,\sigma_f} p_\sigma\langle\sigma_i,|V^\dagger_{\boldsymbol{Q}}(0)|\sigma_f\rangle\langle\sigma_f,|V_{\boldsymbol{Q}}(t)|\sigma_i\rangle = \sum_{\sigma_i}\langle\sigma_i|V^\dagger_{\boldsymbol{Q}}(0)V_{\boldsymbol{Q}}(t)\hat{\varrho}|\sigma_i\rangle = Tr(\hat{\varrho}V^\dagger_{\boldsymbol{Q}}(0)V_{\boldsymbol{Q}}(t))$$

➔ And the corresponding cross-section is

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\int dt Tr(\hat{\varrho}V^\dagger_{\boldsymbol{Q}}(0)V_{\boldsymbol{Q}}(t))\exp(-i\omega t).$$

The Blume-Maleyev Equations (from Lovesey)

$$
\begin{aligned}
\frac{d^2\sigma}{d\Omega dE'} \quad = \quad & \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\int dt\Big\{ \\
& \langle N_{\boldsymbol{Q}} N_{\boldsymbol{Q}}^{\dagger}\rangle + && \text{pure nuclear contribution} \\
& +(\gamma r_0)^2 \langle \boldsymbol{M}_{\perp\boldsymbol{Q}} \boldsymbol{M}_{\perp\boldsymbol{Q}}^{\dagger}\rangle + && \text{pure magnetic contribution} \\
& +(\gamma r_0)\boldsymbol{P_0}\left[\langle N_{\boldsymbol{Q}}^{\dagger}\boldsymbol{M}_{\perp\boldsymbol{Q}}\rangle + \langle \boldsymbol{M}_{\perp\boldsymbol{Q}}^{\dagger} N_{\boldsymbol{Q}}\rangle\right] - && \text{nuclear-magnetic interference} \\
& -i(\gamma r_0)\boldsymbol{P_0}\langle \boldsymbol{M}_{\perp\boldsymbol{Q}} \times \boldsymbol{M}_{\perp\boldsymbol{Q}}^{\dagger}\rangle && \text{chiral magnetic contribution} \\
& \Big\}\exp(-i\omega t),
\end{aligned}
$$

➔ Here $\boldsymbol{P}_0$ describes the polarization of the incident beam.

➔ This equation describes a half-polarized experiment (incident beam polarized but no polarization analysis).

➔ Can this already be useful?

The Blume-Maleyev Equations

➔ Change of polarization during scattering process is given by

$$\boldsymbol{P'} = \frac{Tr(\hat{\varrho} V^{\dagger}_{\boldsymbol{Q}} \hat{\boldsymbol{\sigma}} V_{\boldsymbol{Q}})}{Tr(\hat{\varrho} V^{\dagger}_{\boldsymbol{Q}} V_{\boldsymbol{Q}})}, \quad \boldsymbol{P'} \frac{d^2\sigma}{d\Omega dE'} = \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \int dt Tr(\hat{\varrho} V^{\dagger}_{\boldsymbol{Q}}(0) \hat{\boldsymbol{\sigma}} V_{\boldsymbol{Q}}(t)) \exp(-i\omega t)$$

$$\begin{aligned} \boldsymbol{P'} \frac{d^2\sigma}{d\Omega dE'} &= \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \int dt \Big\{ \boldsymbol{P}_0 \langle N_{\boldsymbol{Q}} N^{\dagger}_{\boldsymbol{Q}}(t) \rangle - (\gamma r_0)^2 \boldsymbol{P}_0 \langle \boldsymbol{M}_{\perp \boldsymbol{Q}} \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}}(t) \rangle + \\ &+ (\gamma r_0)^2 \langle (\boldsymbol{P}_0 \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}}(t)) \boldsymbol{M}_{\perp \boldsymbol{Q}} \rangle + (\gamma r_0)^2 \langle \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}}(t) (\boldsymbol{P}_0 \boldsymbol{M}_{\perp \boldsymbol{Q}}) \rangle + \\ &+ (\gamma r_0) \Big(\langle N^{\dagger}_{\boldsymbol{Q}} \boldsymbol{M}_{\perp \boldsymbol{Q}}(t) \rangle + \langle \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}} N_{\boldsymbol{Q}}(t) \rangle \Big) + \\ &+ i(\gamma r_0) \boldsymbol{P}_0 \times \Big(\langle \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}} N_{\boldsymbol{Q}}(t) \rangle - \langle N^{\dagger}_{\boldsymbol{Q}} \boldsymbol{M}_{\perp \boldsymbol{Q}}(t) \rangle \Big) + \\ &+ i(\gamma r_0)^2 \langle \boldsymbol{M}_{\perp \boldsymbol{Q}} \times \boldsymbol{M}^{\dagger}_{\perp \boldsymbol{Q}}(t) \rangle \Big\} \exp(-i\omega t). \end{aligned}$$

M. Blume. Phys. Rev., 133 (5A) (1964) Yu. A. Izyumov and S.V. Maleyev. Sov. Phys. JETP, 14, 1668 (1962)

The Blume-Maleyev Equations

$$\boldsymbol{P}' = \tilde{\boldsymbol{P}} \boldsymbol{P}_0 + \boldsymbol{P}''$$

$$\sigma \tilde{\boldsymbol{P}} = \begin{pmatrix} (N - M^y - M^z) & iI^z & -iI^y \\ -iI^z & (N + M^y - M^z) & M_{mix} \\ iI^y & M_{mix} & (N - M^y + M^z) \end{pmatrix}, \qquad \sigma \boldsymbol{P}'' = \begin{pmatrix} C \\ R^y \\ R^z \end{pmatrix}$$

$$\sigma \equiv \frac{d^2\sigma}{d\Omega dE'} = \underbrace{N + M^y + M^z}_{\text{independent of } \boldsymbol{P}_0} - \underbrace{P_0^x C + P_0^y R^y + P_0^z R^z}_{\text{dependent on } \boldsymbol{P}_0}.$$

➔ The actually measured quantity is the polarization tensor

$$\mathrm{P}_{ij} = (P_{i0} \tilde{P}_{ji} + P_j'')/|\boldsymbol{P}_0|,$$

with *i* and *j* (*i, j = x, y, z*) being the components of the incident and final polarization vectors.

P. J. Brown, Physica B, 297, 198 (2001) M. Janoschek Physica B, 397, 125 (2007).

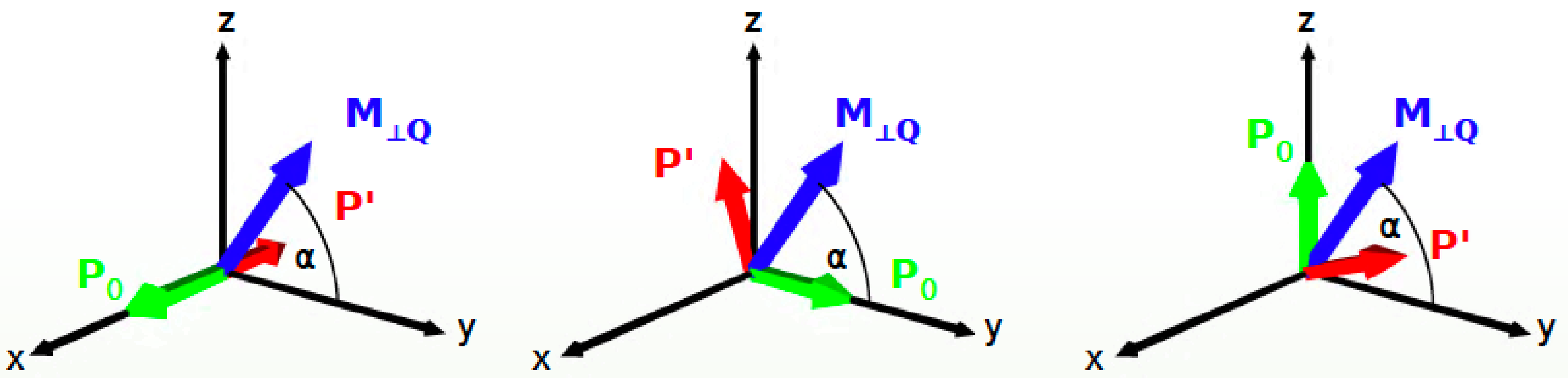
Relevant Terms in Polarization Tensor

$$\sigma\tilde{\boldsymbol{P}} = \begin{pmatrix} (N-M^y-M^z) & iI^z & -iI^y \\ -iI^z & (N+M^y-M^z) & M_{mix} \\ iI^y & M_{mix} & (N-M^y+M^z) \end{pmatrix}, \qquad \sigma\boldsymbol{P}'' = \begin{pmatrix} C \\ R^y \\ R^z \end{pmatrix}$$

Item	correlation functions	description
N	$\frac{k_f}{k_i}\langle N_{\boldsymbol{Q}} N^{\dagger}_{\boldsymbol{Q}}\rangle_\omega$	nuclear contribution
$M^{y/z}$	$(\gamma r_0)^2\frac{k_f}{k_i}\langle M^{y/z}_{\perp\boldsymbol{Q}} M^{\dagger y/z}_{\perp\boldsymbol{Q}}\rangle_\omega$	y- and z-components of the magnetic contribution.
$R^{y/z}$	$(\gamma r_0)\frac{k_f}{k_i}\langle N^{\dagger}_{\boldsymbol{Q}} M^{y/z}_{\perp\boldsymbol{Q}}\rangle_\omega + \langle M^{\dagger y/z}_{\perp\boldsymbol{Q}} N_{\boldsymbol{Q}}\rangle_\omega$	real parts of the nuclear-magnetic interference term.
$I^{y/z}$	$(\gamma r_0)\frac{k_f}{k_i}\langle N^{\dagger}_{\boldsymbol{Q}} M^{y/z}_{\perp\boldsymbol{Q}}\rangle_\omega - \langle M^{\dagger y/z}_{\perp\boldsymbol{Q}} N_{\boldsymbol{Q}}\rangle_\omega$	imaginary parts of the nuclear-magnetic interference term.
C	$i(\gamma r_0)^2\frac{k_f}{k_i}(\langle M^{y}_{\perp\boldsymbol{Q}} M^{\dagger z}_{\perp\boldsymbol{Q}}\rangle_\omega - \langle M^{z}_{\perp\boldsymbol{Q}} M^{\dagger y}_{\perp\boldsymbol{Q}}\rangle_\omega)$	chiral contribution
M_{mix}	$(\gamma r_0)^2\frac{k_f}{k_i}(\langle M^{y}_{\perp\boldsymbol{Q}} M^{\dagger z}_{\perp\boldsymbol{Q}}\rangle_\omega + \langle M^{z}_{\perp\boldsymbol{Q}} M^{\dagger y}_{\perp\boldsymbol{Q}}\rangle_\omega)$	mixed magnetic contribution or magnetic-magnetic interference term

P. J. Brown, Physica B, 297, 198 (2001) M. Janoschek Physica B, 397, 125 (2007).

Meaning of Terms



➔ Consider: $M_{\perp Q} = m\,[\hat{e}_y \cos(\alpha) + \hat{e}_z \sin(\alpha)]$

➔ $M^y + M^z = m^2 \cos^2(\alpha) + m^2 \sin^2(\alpha) = m^2.$

$-M^y + M^z = -(M^y - M^z) = -m^2 \cos^2(\alpha) + m^2 \sin^2(\alpha) = -m^2 \cos(2\alpha).$

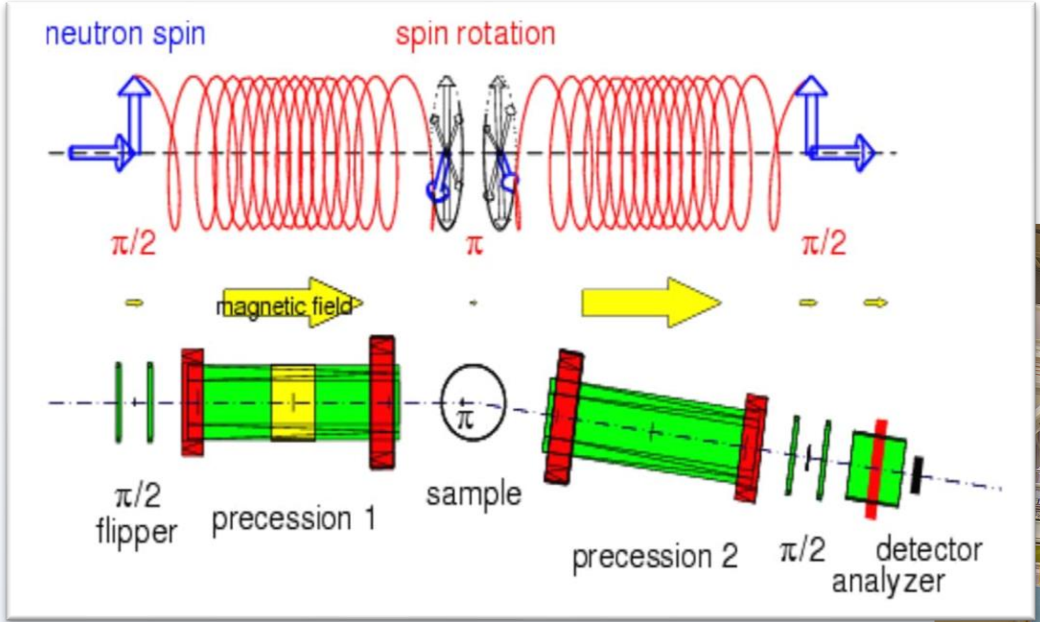
$M_{mix} = m^2 \cos(\alpha) \sin(\alpha) + m^2 \sin(\alpha) \cos(\alpha) = m^2 \sin(2\alpha).$

$C = 0$ as $M_{\perp Q}$ is real and therefore $M_{\perp Q} \parallel M^{\dagger}_{\perp Q}$.

➔ $\sigma = m^2$ and $\mathrm{P} = \begin{pmatrix} -1 & 0 & 0 \\ 0 & \cos(2\alpha) & \sin(2\alpha) \\ 0 & \sin(2\alpha) & -\cos(2\alpha) \end{pmatrix}$

Take home message:
- Polarization component parallel to $M_{\perp Q}$ stay
- Polarization component perpendicular to $M_{\perp Q}$ flip

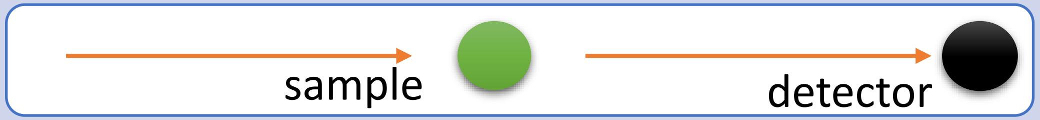
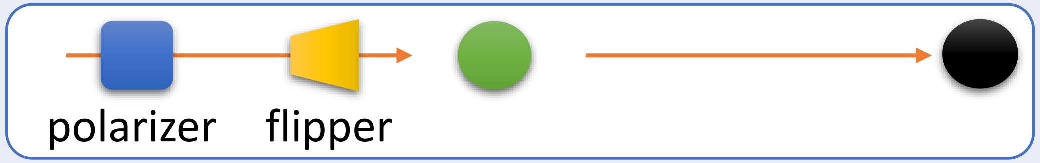
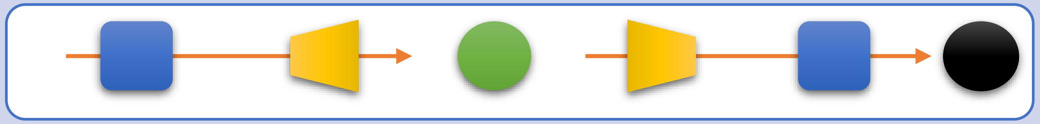
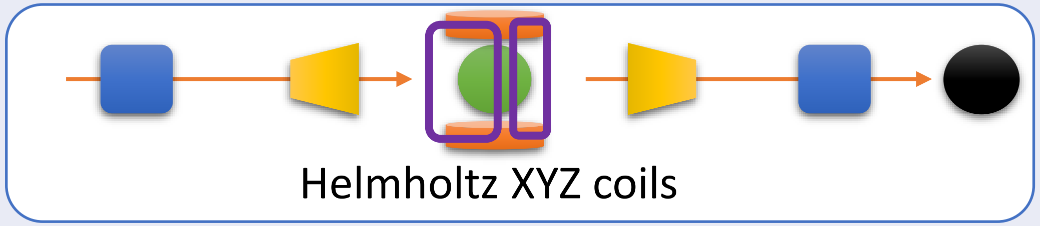
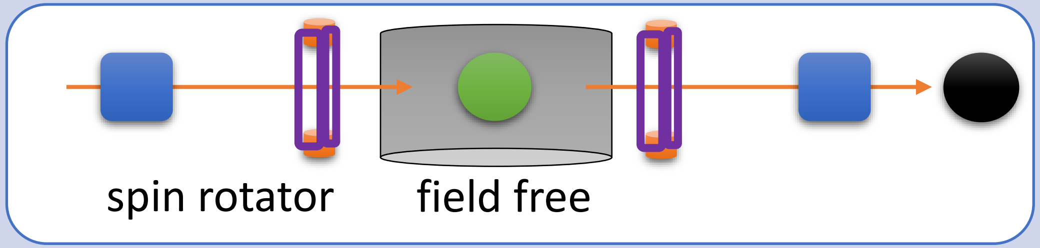
Neutron Spin Echo – Polarization as Clock



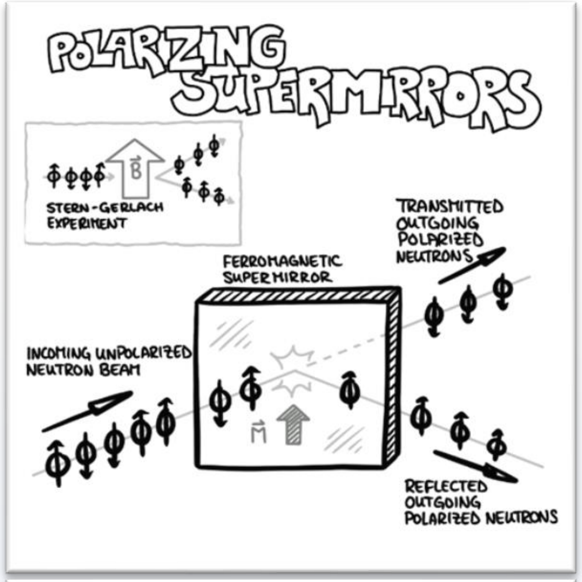
Classes of polarized neutron experiments

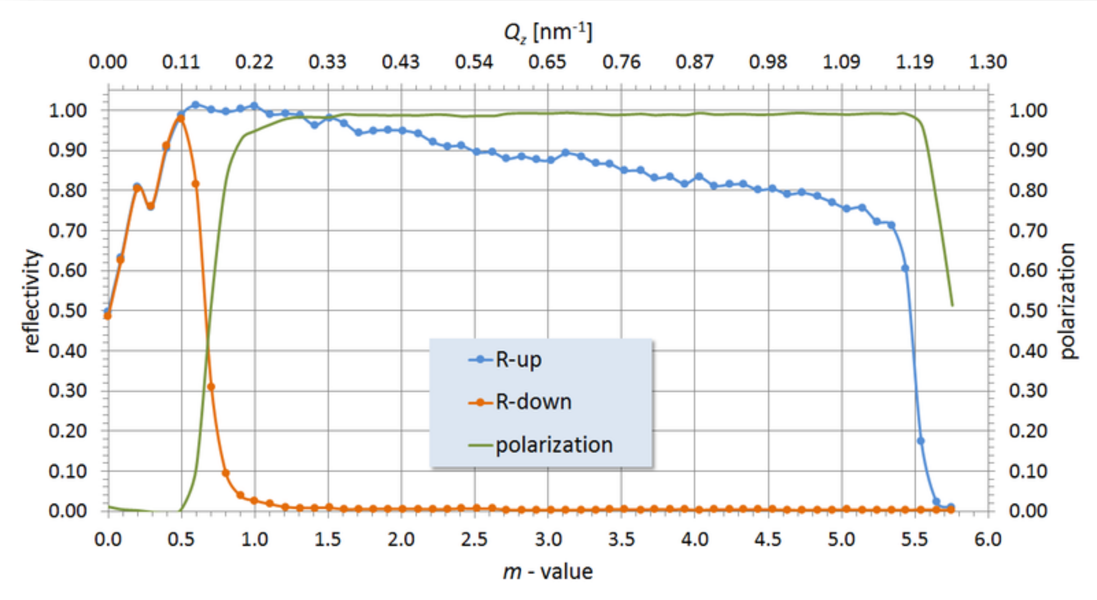
Experiment	P. Incoming	P. Outgoing	Information
Unpolarized	X	X	Propagation vector of magnetic order, μ AF-ordered (few exceptions)
(“Half”) polarized	$P_{\pm Z}$	X	+ μ FM order along B || P_Z
Polarization analysis	$P_{\pm Z}$	$P_{\pm Z}$	+ Separate $M_{\|\|Z}$ and $M_{\perp Z}$ components + Separate N from $M_{\perp Z}$
XYZ polarization analysis	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	+ Separate spin-incoherent scattering + Separate N from $M_{\|\|Z}$ - No fixed direction magnetic field at sample
Polarimetry (Spherical pol. analysis)	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$ $P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$ $P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	$P_{\pm X}$ / $P_{\pm X}$ / $P_{\pm X}$ $P_{\pm Y}$ / $P_{\pm Y}$ / $P_{\pm Y}$ $P_{\pm Z}$ / $P_{\pm Z}$ / $P_{\pm Z}$	+ Magnetic interaction terms (complex spin structures) - No magnetic field around sample

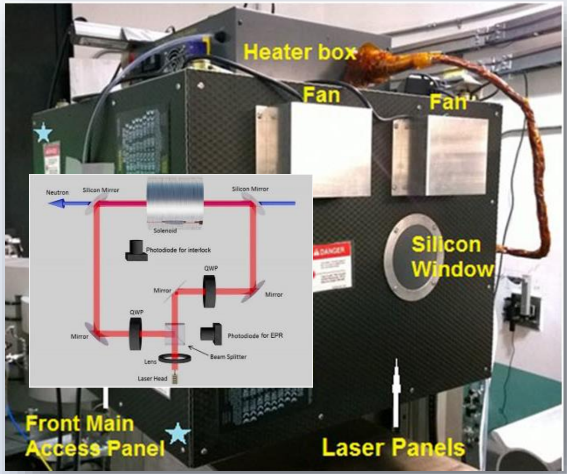
Classes of polarized neutron experiments

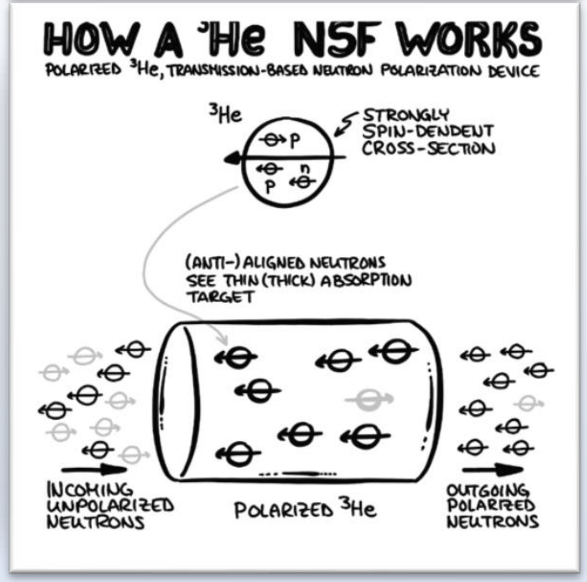
Experiment	P. Incoming	P. Outgoing	Implementation
Unpolarized	X	X	
("Half") polarized	$P_{\pm Z}$	X	
Polarization analysis	$P_{\pm Z}$	$P_{\pm Z}$	
XYZ polarization analysis	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	
Polarimetry (Spherical pol. analysis)	$P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$ $P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$ $P_{\pm X}$ / $P_{\pm Y}$ / $P_{\pm Z}$	$P_{\pm X}$ / $P_{\pm X}$ / $P_{\pm X}$ $P_{\pm Y}$ / $P_{\pm Y}$ / $P_{\pm Y}$ $P_{\pm Z}$ / $P_{\pm Z}$ / $P_{\pm Z}$	

Neutron polarizers





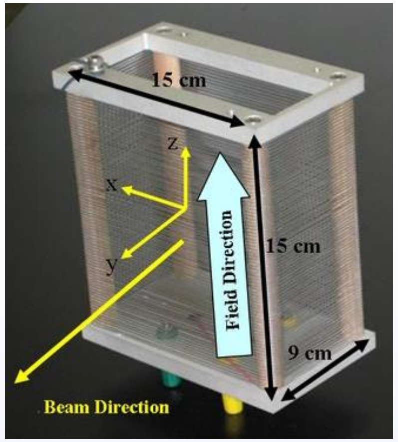




@Rob_Dimeo

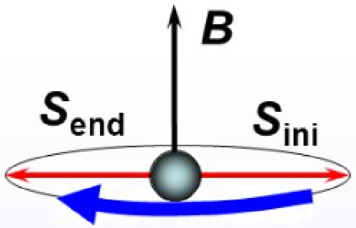
Neutron scattering lengths and cross sections							
Isotope	conc	Coh b	Inc b	Coh xs	Inc xs	Scatt xs	Abs xs
He	---	3.26(3)	---	1.34	0	1.34	0.00747
3He	0.00014	5.74-1.483*i*	-2.5+2.568*i*	4.42	1.6	6	5333.(7.)
4He	99.99986	3.26	0	1.34	0	1.34	0

Neutron spin-flippers



Mezei flipper

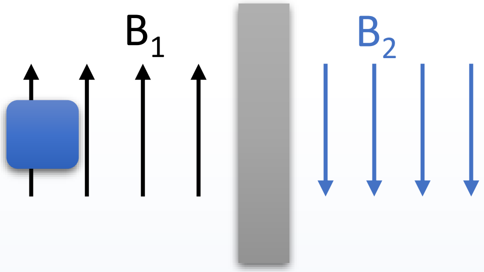
Lamor-precession



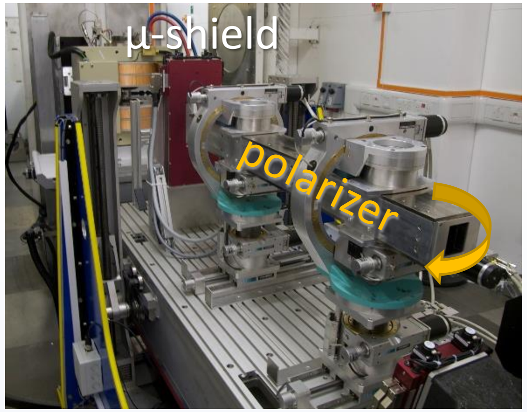
+ simple and cheap

- only single wavelength

non-adiabatic



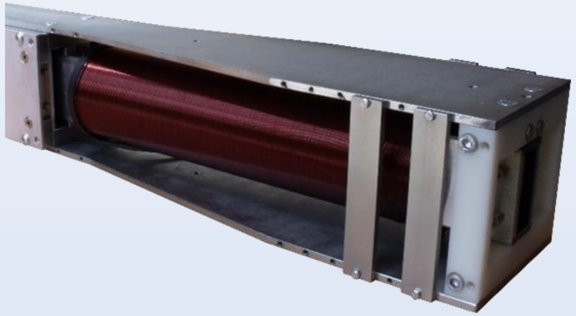
μ-metal or superconductor

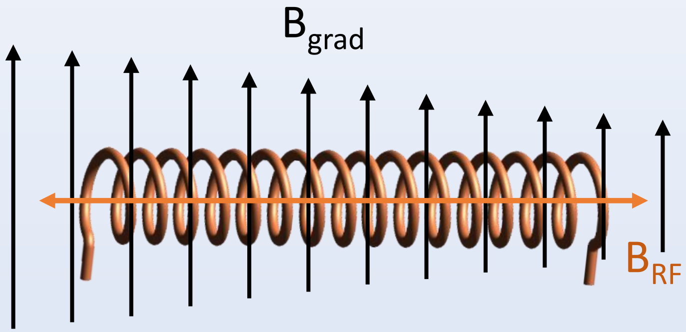


+ broad wavelength band (ToF)

- losses in intensity
- can lead to partial depolarization

Resonant / radio frequency



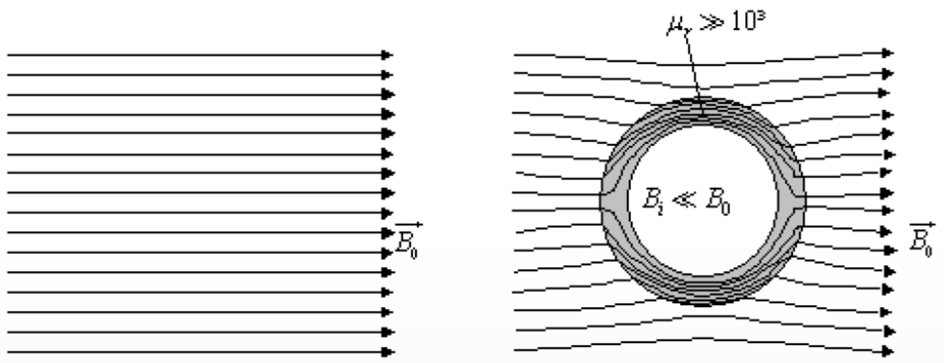


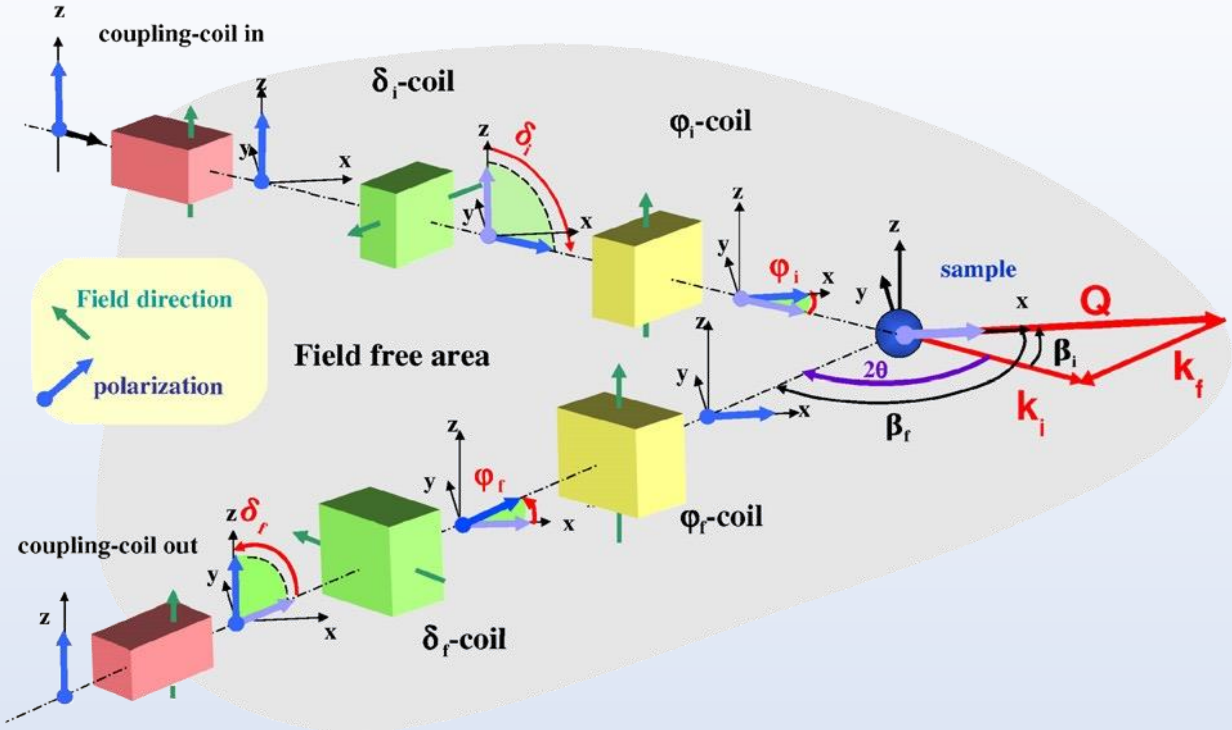
+ broad wavelength band

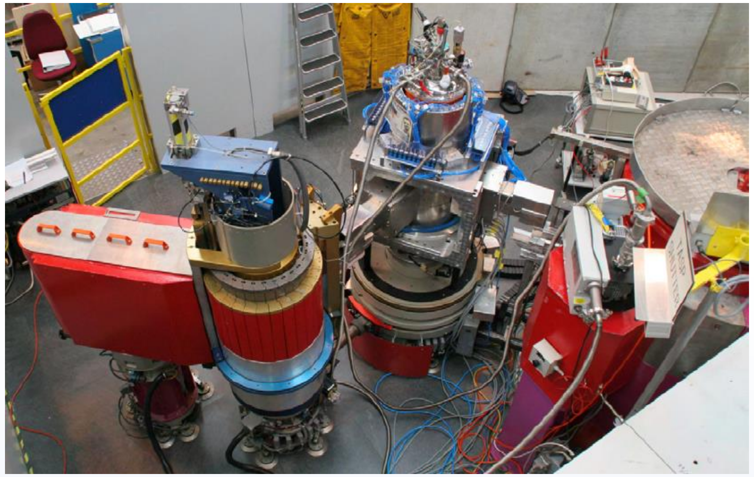
+ very high efficiency (>99%)

- more costly
- requires more space

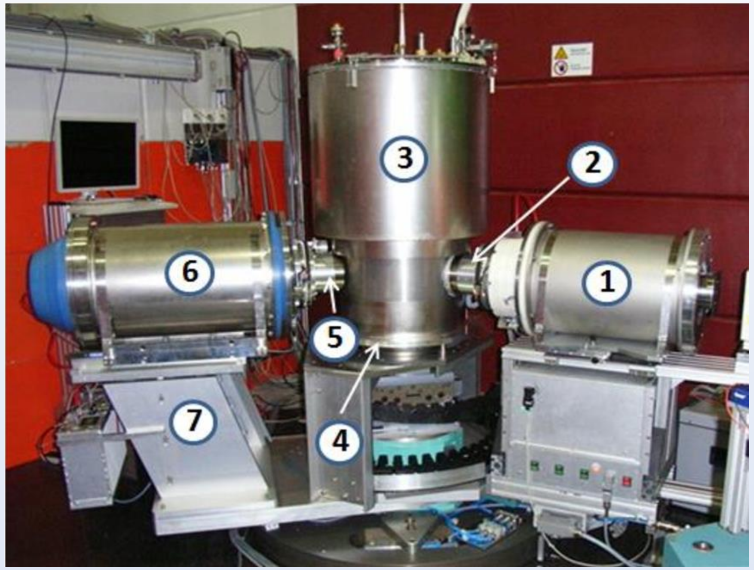
Neutron polarimetry devices







MuPAD



CryoPAD

Discerning Magnetic from Nuclear Inelastic Scattering

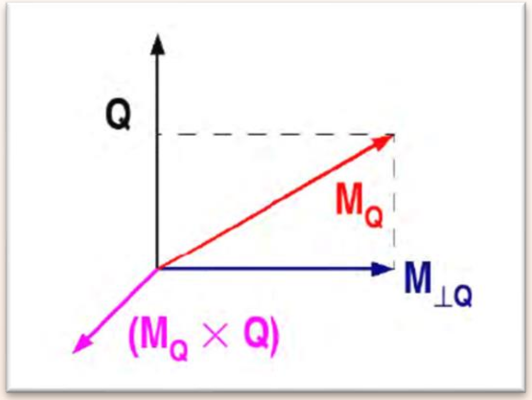
1. The best way is to try to find Brillouin zones that only have nuclear or magnetic scattering (nuclear vs. magnetic structure factor).

TABLE I. Nuclear (F_N^2) and magnetic (F_M^2) structure factors of MnSi at 0 K. F_M^2 includes the magnetic form factor. $a=4.558$ Å, $u_{Mn}=0.138$, $u_{Si}=0.845$, $M=0.4\mu_B$.

h	k	l	F_N^2	F_M^2
1	0	0	0	0
0	1	1	2.362	0.0269
1	1	1	2.624	0.0230
2	0	0	0.143	0.0015
2	0	1	3.479	0.0225
0	2	2	0.036	0.0000
3	0	0	0	0
1	2	2	0.308	0.0005
2	2	2	8.016	0.0122

2. In general measure phonons at higher and magnons at lower *Q* to take advantage of Q^2 *vs* $f(Q)$.

3. Magnetic scattering can sometimes be excluded via geometric selection rule.

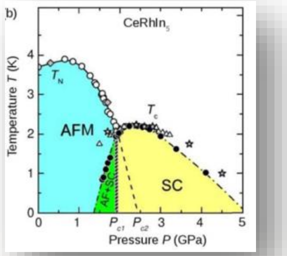


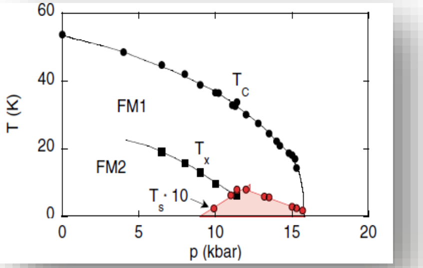
4. Subtract intensities that are unwanted by using independent measurements (careful).
5. **Polarization analysis** (very powerful, unfortunately decreases intensity by roughly factor 2-3 and you have to measure 4-18 different combinations to get information)

Subtracting Unwanted Background/Phonons

$CeRhIn_5$

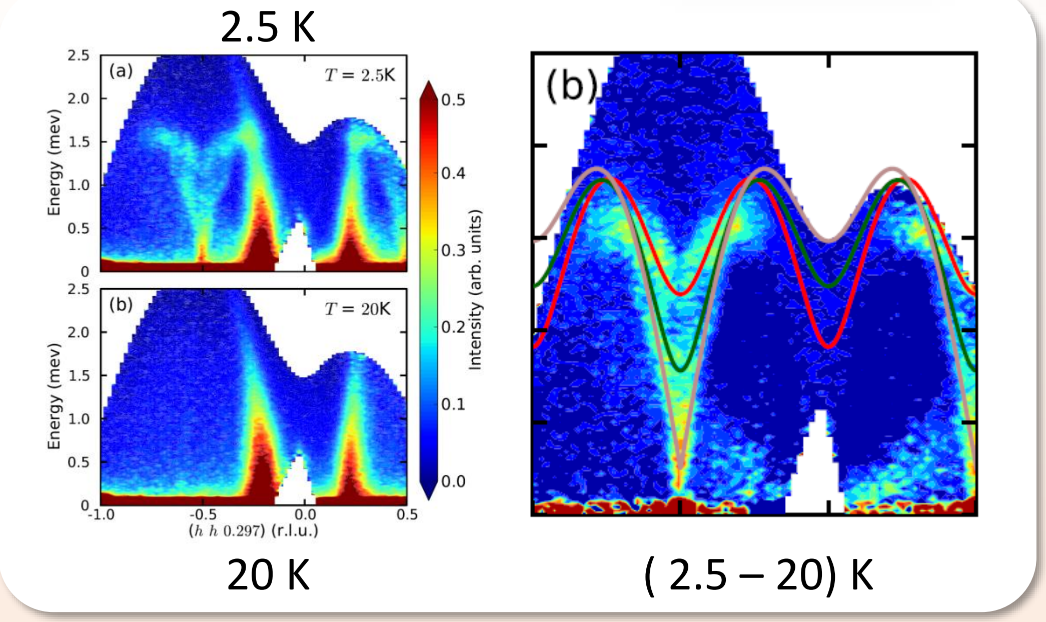
$T_N = 3.8$ K



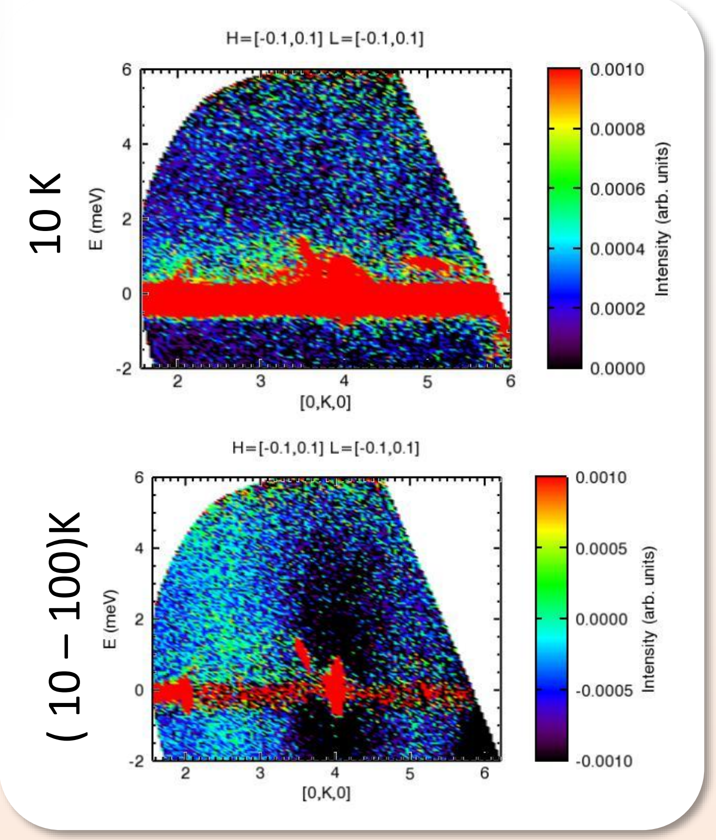


UGe_2

$T_C = 53$ K



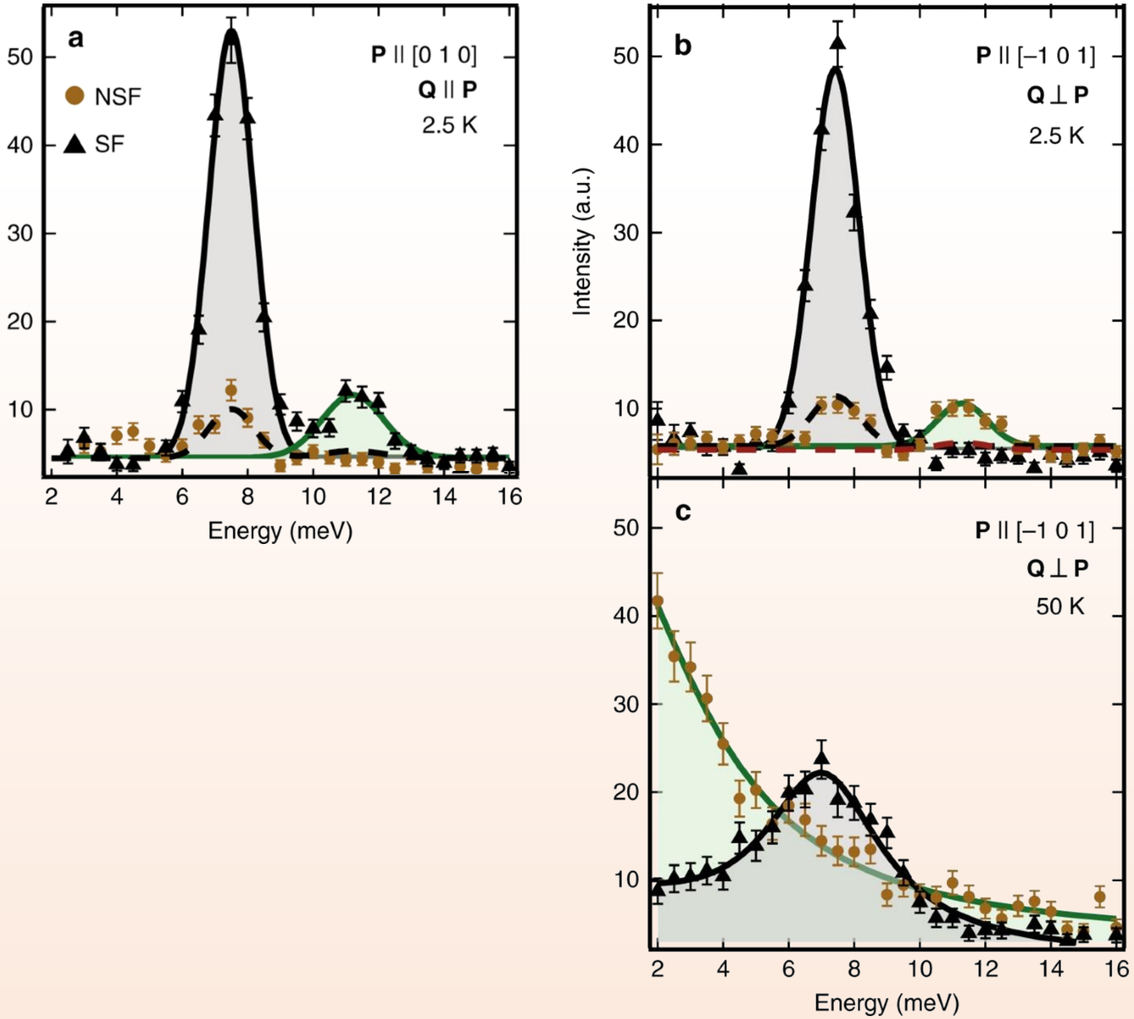
WORKS NICELY

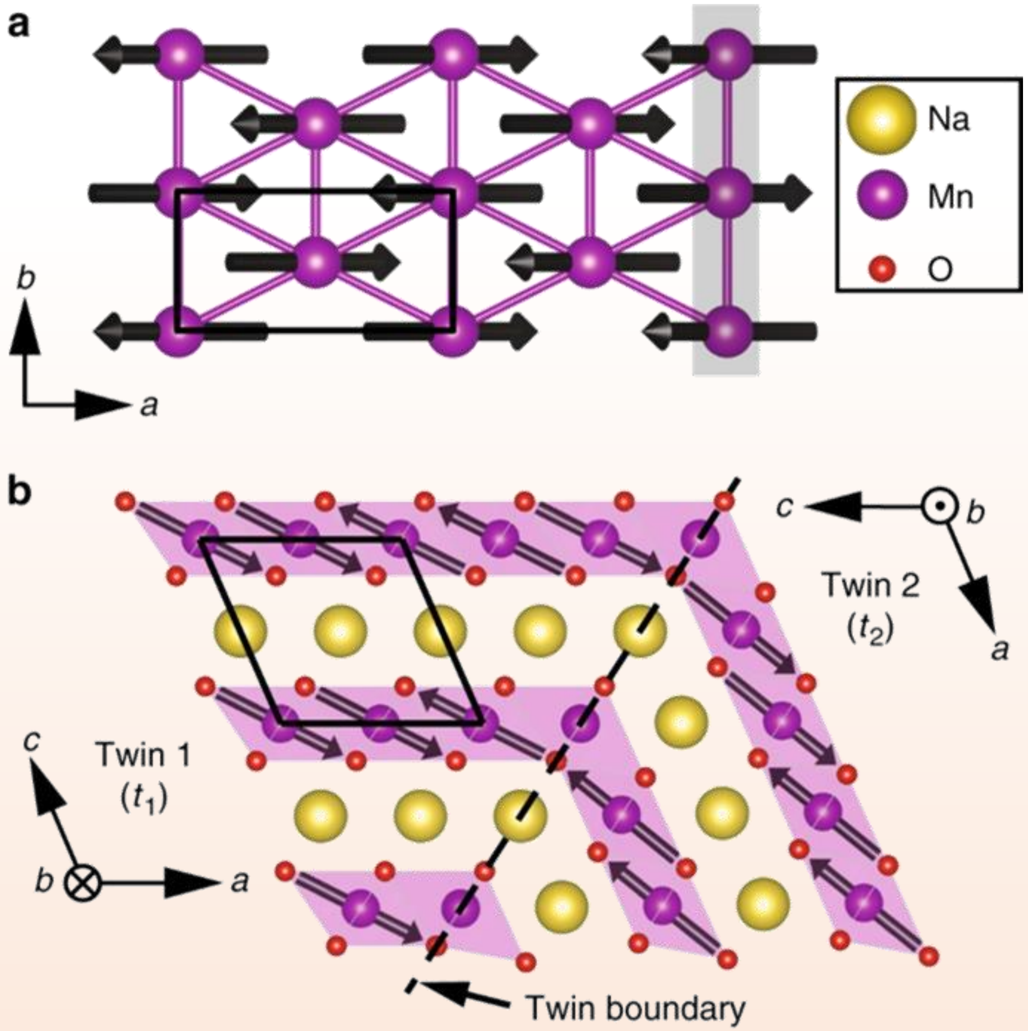


DOES *NOT* WORK

What is the issue with the second example?

Separating magnetic and nuclear scattering (inelastic)

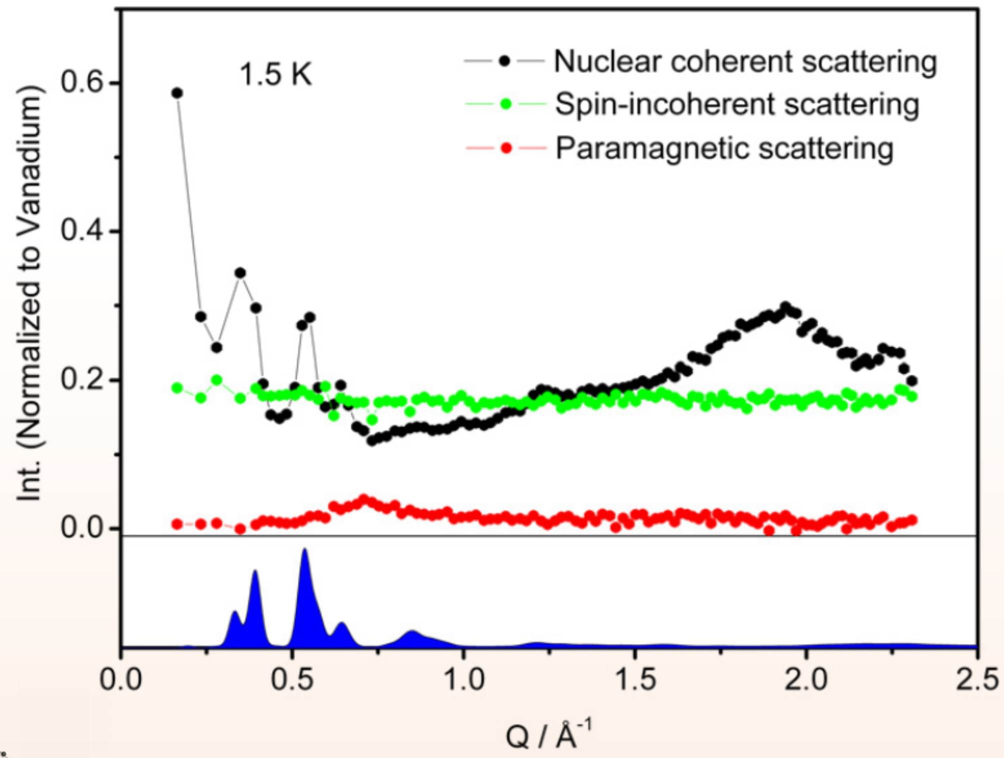


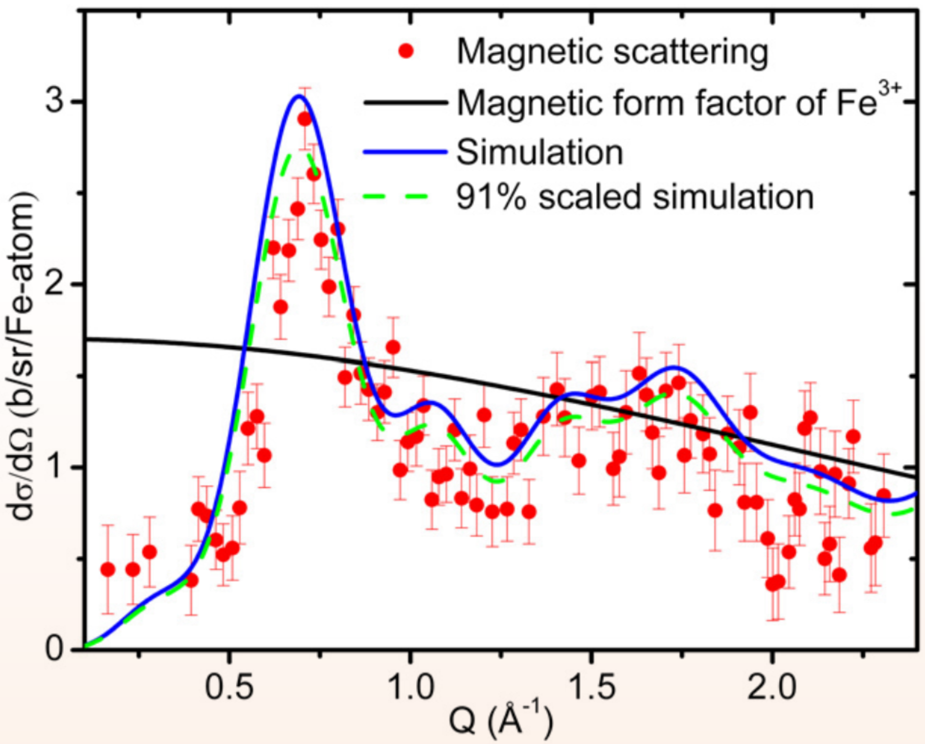


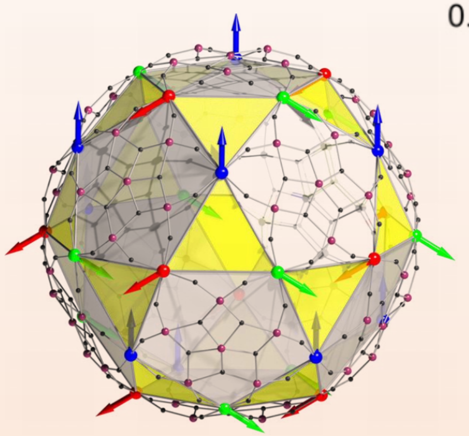
$Na_{0.9}MnO_2$

R. L. Dally, *et al.*, Nat. Comm. **9**, 2188 (2018)

Separating magnetic and nuclear scattering (elastic)







Molecular magnets are tiny crystals with a well defined atomic structure and some magnetic atoms that have a magnetic order.

The small magnetic signal can be extracted from the nuclear dominated scattering by the use of XYZ-polarization analysis measurements.

Z. Fu, *et al.*, New Journal of Physics **12**, 083044 (2010)