Polarized Neutron Scattering

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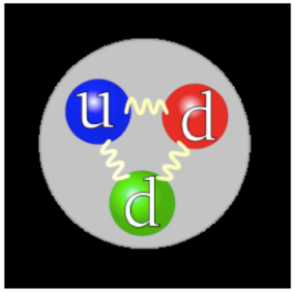
SwedNess/NNSP, Tartu Estonia, September 19th 2017

The neutron



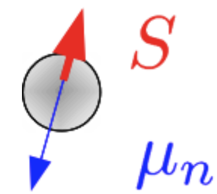
Source: Wikipedia

Charge 0
Spin 1/2

Quarks

	Charge	Spin
u	2/3	1/2
d	-1/3	1/2

Neutron is a **spin ½ particle**, the spin is tied to a **magnetic moment**.



$$\mu_n = \gamma_n \mu_N = g_n S \mu_N$$

$$\gamma_n = -1.913 \qquad \mu_N \equiv \frac{e\hbar}{2m_p} \qquad \text{compare} \quad \mu_B \equiv \frac{e\hbar}{2m_e}$$

neutron interacts with nuclei

Its **spin** interacts with spin of nuclei

Its **magnetic moment** interacts with magnetic moments of unpaired electrons

=> Structure and dynamics of atoms and magnetic moments

Why polarized neutron scattering?

to see more and to separate scattering terms

Do it with polarized neutron scattering?

PHYSICAL REVIEW VOLUME 83, NUMBER 2 JULY 15, 1951

Neutron Diffraction by Paramagnetic and Antiferromagnetic Substances

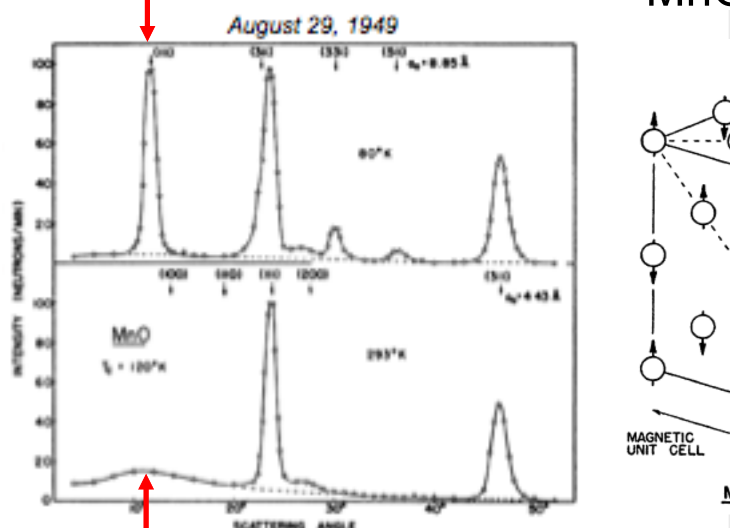
C. G. Shull, W. A. Strauser, and E. O. Wollan
Oak Ridge National Laboratory, Oak Ridge, Tennessee

(Received March 2, 1951)

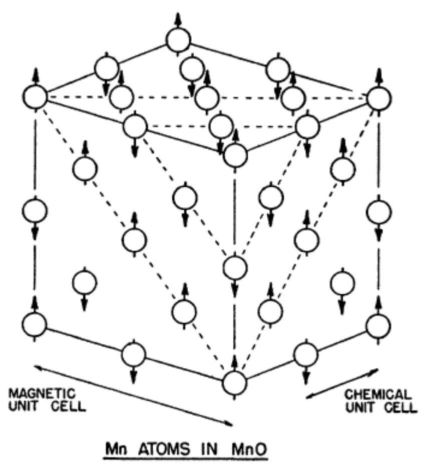
Detection of Antiferromagnetism by Neutron Diffraction 1949 *Shull et al.* *Nobel prize 1994*

"for the development of neutron diffraction technique"

(111) alternating layers



MnO



paramagnetic
spin fluctuations

spin structure from intensities

spins

|| to [100] *Shull et al. PR 1951*

in (111) planes *Shaked et al. PRB 1988*

|| to $[11\bar{2}]$ *Goodwin et al. PRL 2006*

polarized neutron scattering provides more information

intensities and polarization

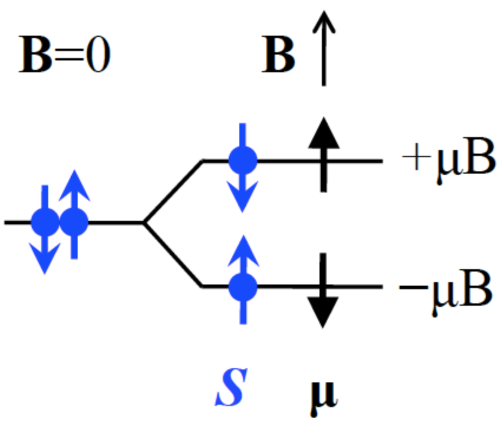
Outline

- **Neutron spins in magnetic fields**
  - experimental devices => instruments
- **Scattering and Polarization** *Moon, Riste, Koehler*
  - spin-dependent nuclear interaction
  - magnetic interaction
- **Blume-Maleyev Equations**
  - examples
- **outlook for ESS**

$$\gamma = 2\gamma_{\mathrm{n}}\mu_{\mathrm{N}}/\hbar = -1.83 \cdot 10^8\,\mathrm{s}^{-1}\mathrm{T}^{-1}$$
$$\gamma/2\pi = -2916\,\mathrm{Hz/Oe.}$$

Neutron spins in magnetic fields

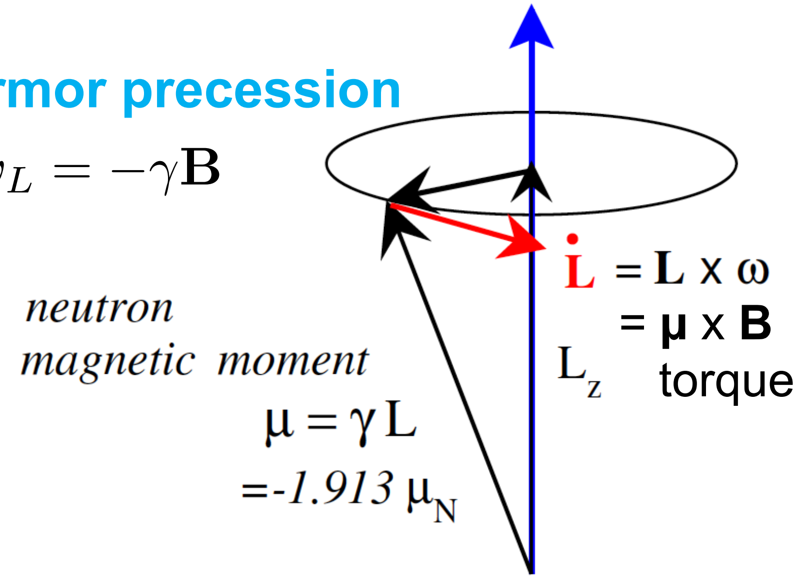
Zeeman splitting



$\hbar\omega_L$

Larmor precession

$\omega_L = -\gamma \mathbf{B}$



Bloch equation of motion

$$\dot{\boldsymbol{\mu}} = \gamma\, \boldsymbol{\mu} \times \mathrm{B}$$

QM: no nutation

Neutron beam polarization

with respect to magnetic field

average of spins: $\mathbf{P} = 2\langle \mathbf{S} \rangle$ $-1 < \mathbf{P} < 1$

normalized difference of intensities neutron spin up $n_\uparrow$ and down $n_\downarrow$

$$\mathrm{P} = \frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow}$$ $0 < \mathrm{P} < 1$

Absorption and transmission

Polarized He-3 filter

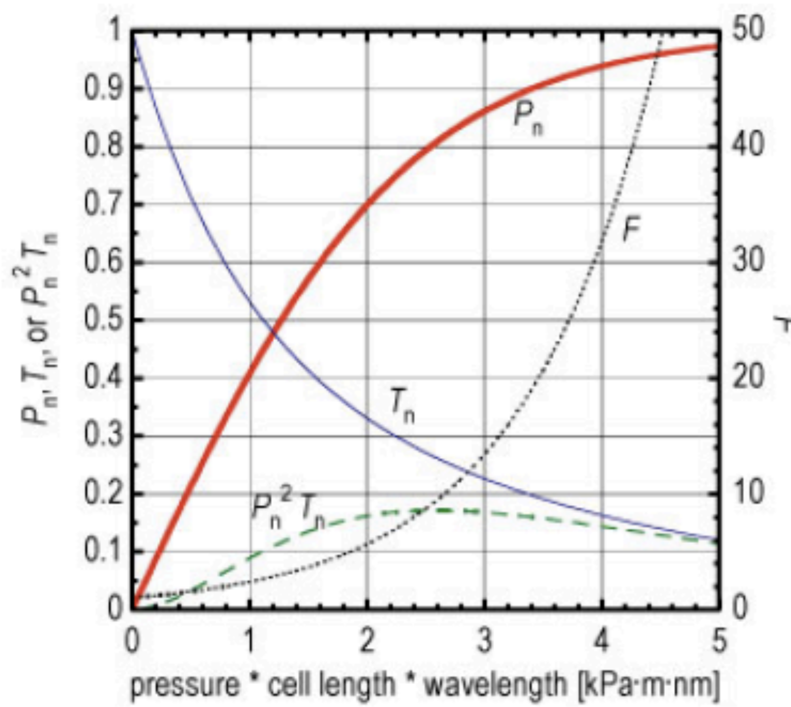
$$\sigma_{abs}(n_{\uparrow}, {}^{3}He_{\uparrow}) = 0$$

$$\sigma_{abs}(n_{\uparrow}, {}^{3}He_{\downarrow}) = 5333b$$

- SEOP Spin-exchange-optical pumping
- Laser polarizes Rb
- exchange with K then ^{3}He-spin
- very homogeneous field

^{3}He cells used at JCNS

Tunable efficiency
by pressure and volume
typically *good for thermal neutrons*



Scattering
constructive interference of nuclear b and magnetic scattering amplitudes p

$$\sigma_\pm \propto (b \pm p)^2$$ *Lecture 7*

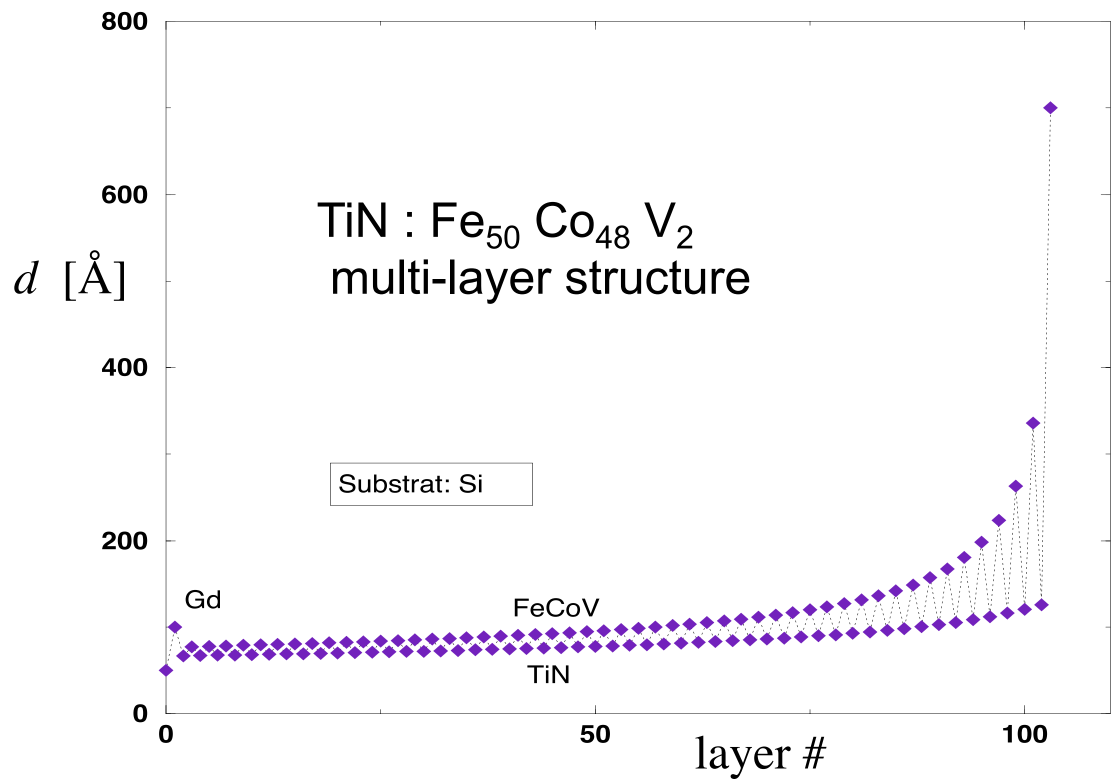
- **Magnetic Bragg scattering**
  e.g. Heusler crystals, Cu_2MnAl (111), P= 0.95
  single ferro domain needed, low reflectivity

  see in following: Moon, Riste, Koehler

Scattering
constructive interference of nuclear and magnetic scattering

- Total reflection by of magnetic “super-mirrors” (Mezei, Schärpf)

Bragg-diffraction from an
multi-layer structure
of varying layer thickness d

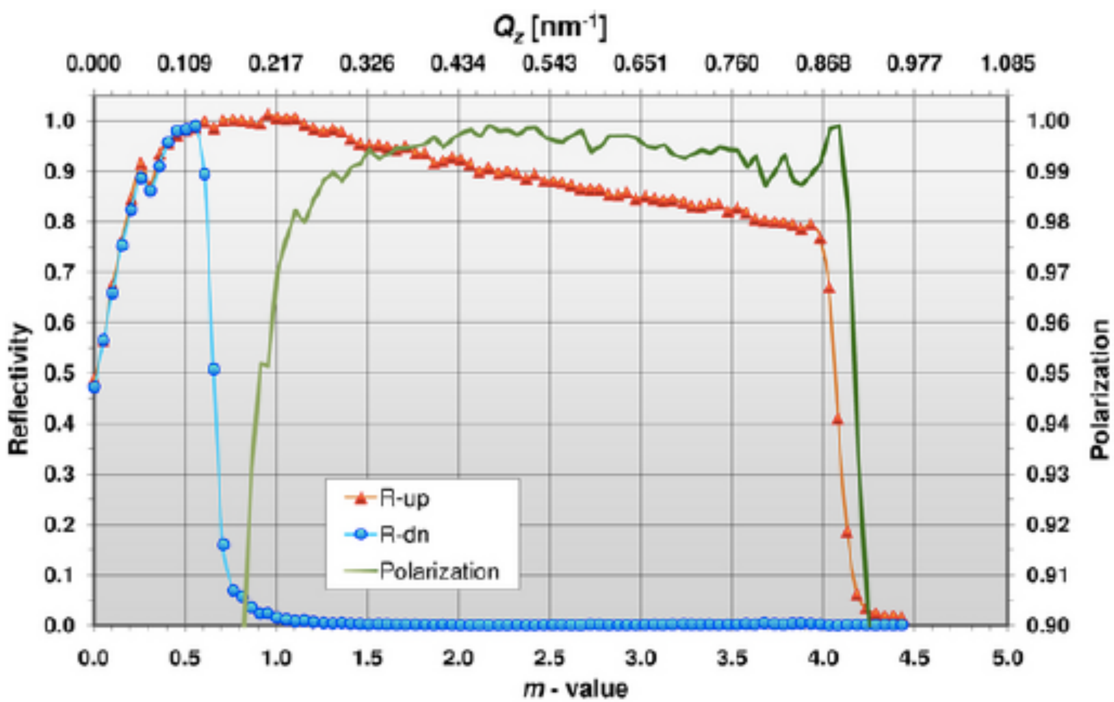


Scattering
constructive interference of nuclear and magnetic scattering

- Total reflection by of magnetic "super-mirrors" (Mezei, Schärpf)

$$\Theta_c^{\pm} = \lambda\sqrt{n(b \pm p)/\pi}$$

Surface of
FeSi multilayers

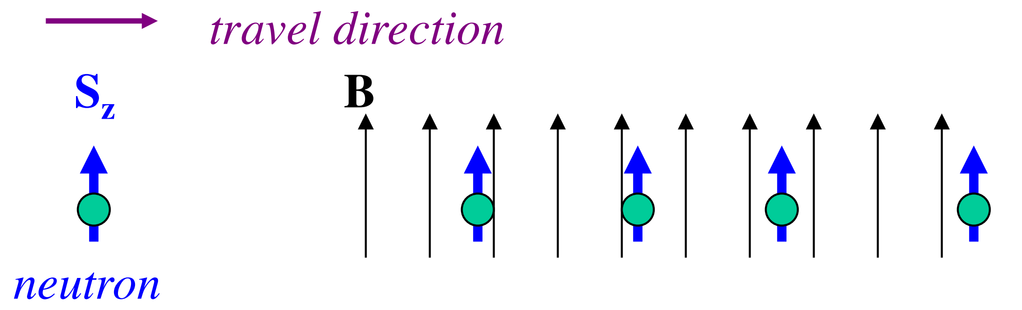


much better polarization
at the interface of
Si : FeSi multilayers

Source: Swiss Neutronics

Guide fields

constant B

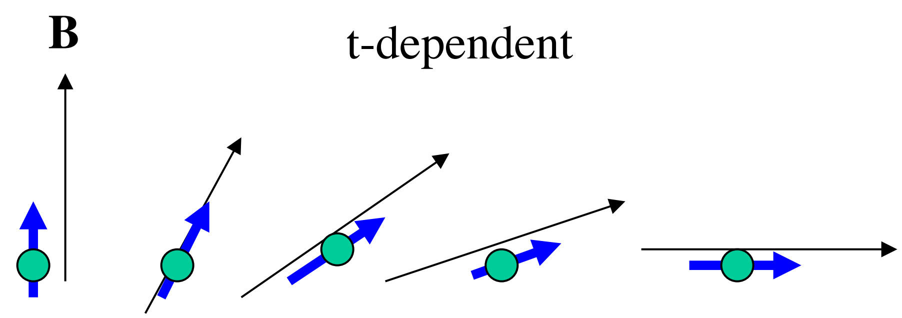


Asymptotic behaviour

adiabatic field change

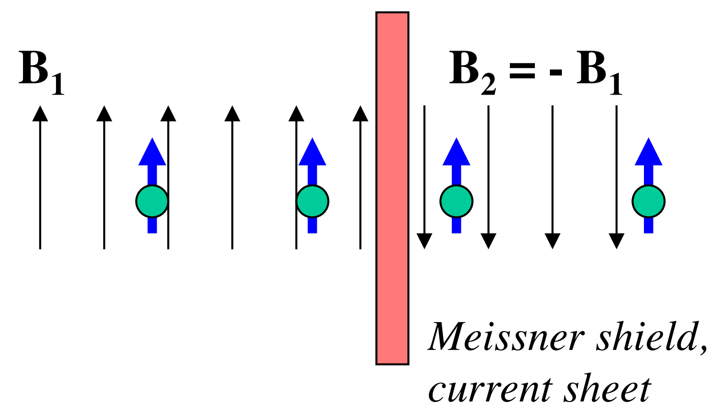
slowly varying "strong" B(t)

fast precession around B



sudden change

no change of S_z but change wrt B

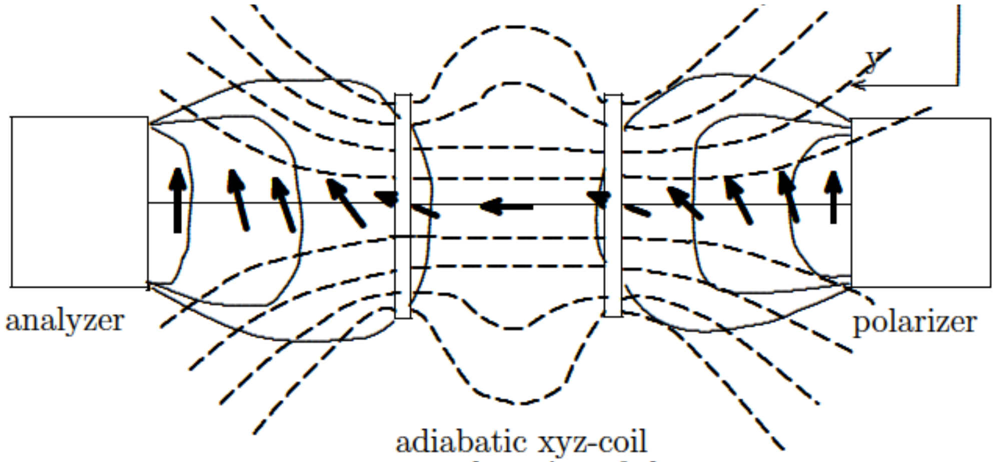


General behaviour

Solve Bloch equation of motion $\dot{\boldsymbol{\mu}} = \gamma\, \boldsymbol{\mu} \times \mathrm{B}$

Guide fields

nutators, xyz-coils



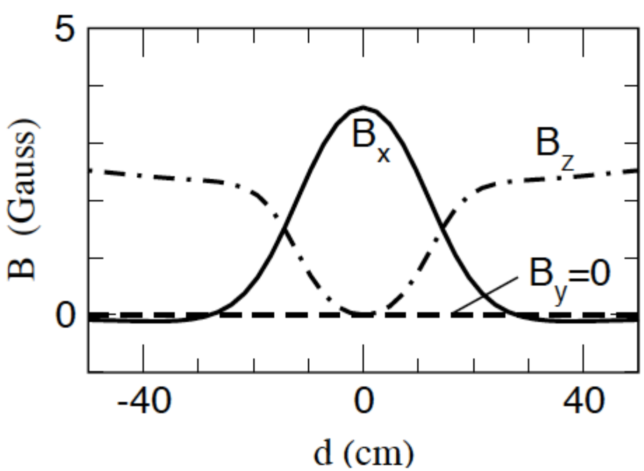


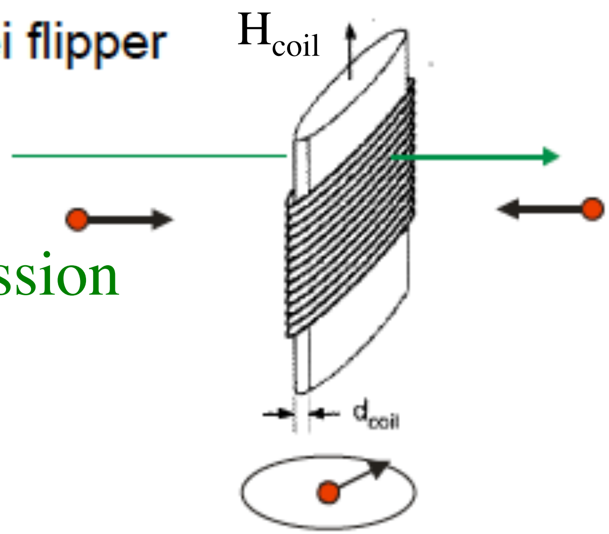
Fig. 6: *(left) Magnetic field setting in a xyz-coil system for an adiabatic nutation of the polarization of cold neutrons in horizontal x-direction at the sample turning to a vertical (guide) field B_z at further distance from the sample. (right) A photo of the xyz-coil system in the DNS instrument at the FRM-2.*

Flipper

Objective: change neutron polarization with respect to the applied field

Adiabatic and non-adiabatic changes of P ll B

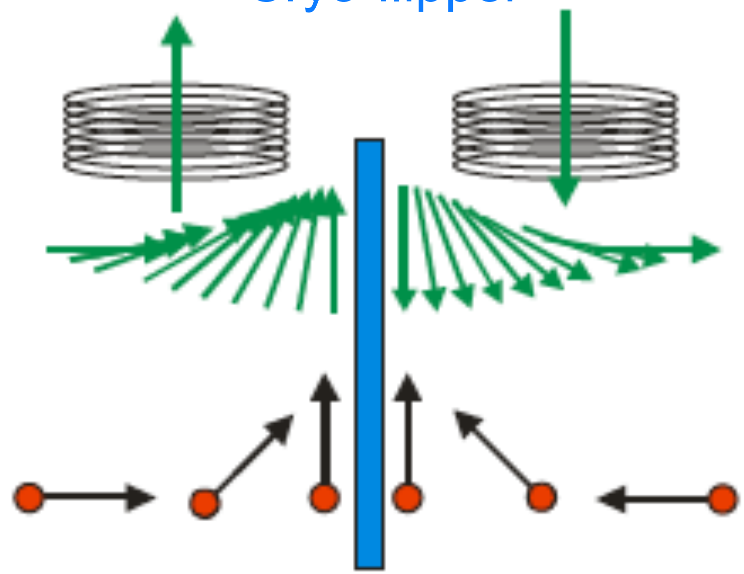
- Mezei flipper



precession

precession coil

Cryo-flipper



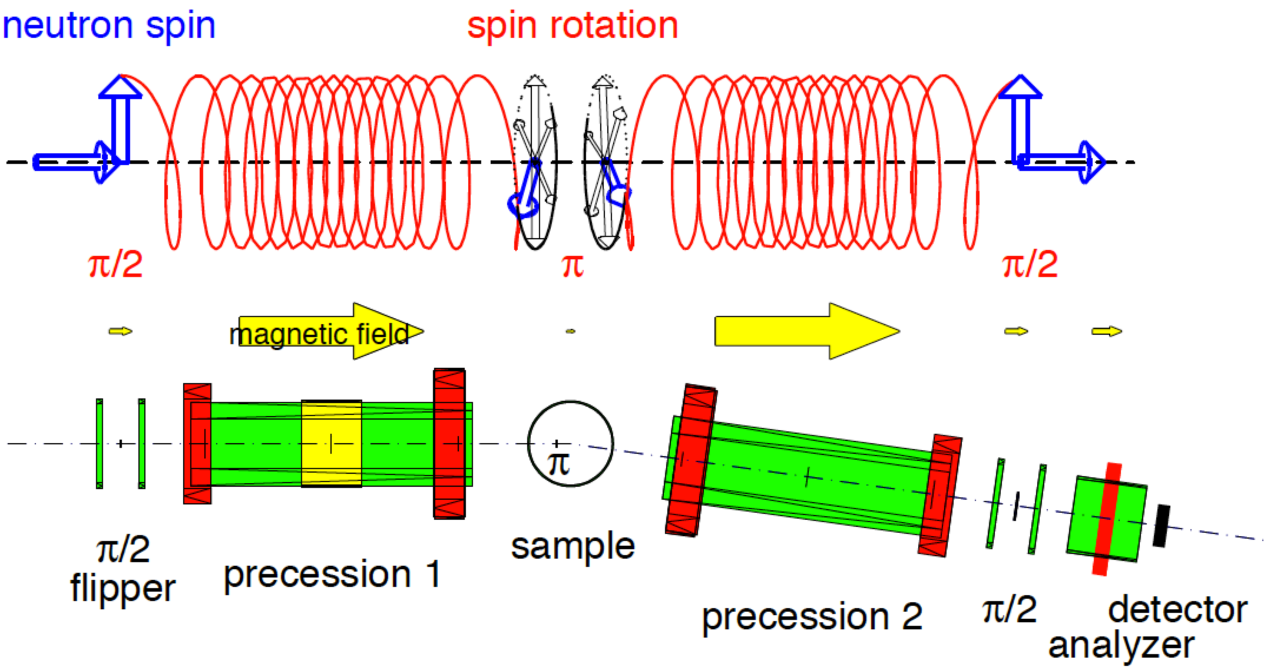
Meissner shield *superconducter*

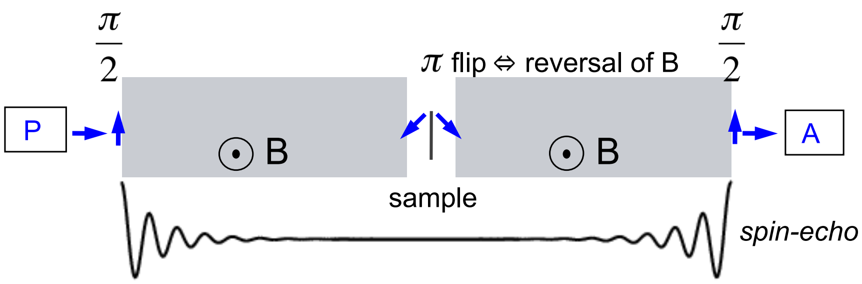
π- flipper

$$\omega \cdot t = -\gamma B \cdot d/v = \pi$$

$$B = \frac{\pi}{d}(\mathrm{m/s} \cdot \mathrm{Å}/\lambda)/(2916 \cdot 2\pi\,\mathrm{Hz/Oe}) = \frac{67.83}{d\lambda}\,\mathrm{cm\,Å\,Oe}$$

Spin-echo technique



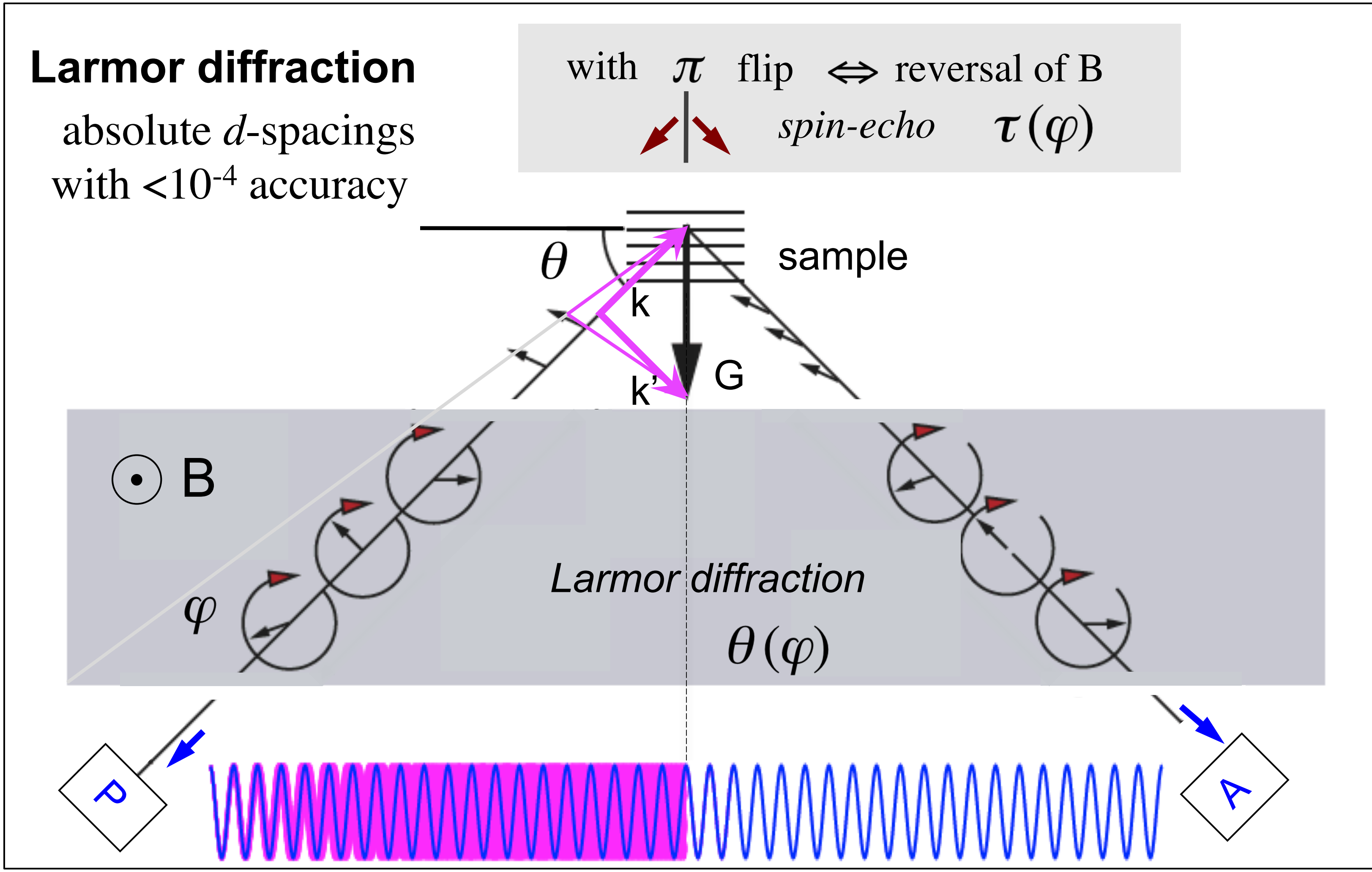


$$I \propto \int d\omega\, P_{(z)} S(\mathbf{Q},\omega) = \frac{1}{2}\left(S(Q) + \int d\omega\, \cos(\phi) S(\mathbf{Q},\omega)\right),$$

$$\phi = \omega t = \text{const} \cdot \omega \int ds\, B$$

$$I \propto \frac{1}{2}(S(Q) + S(Q,t))$$

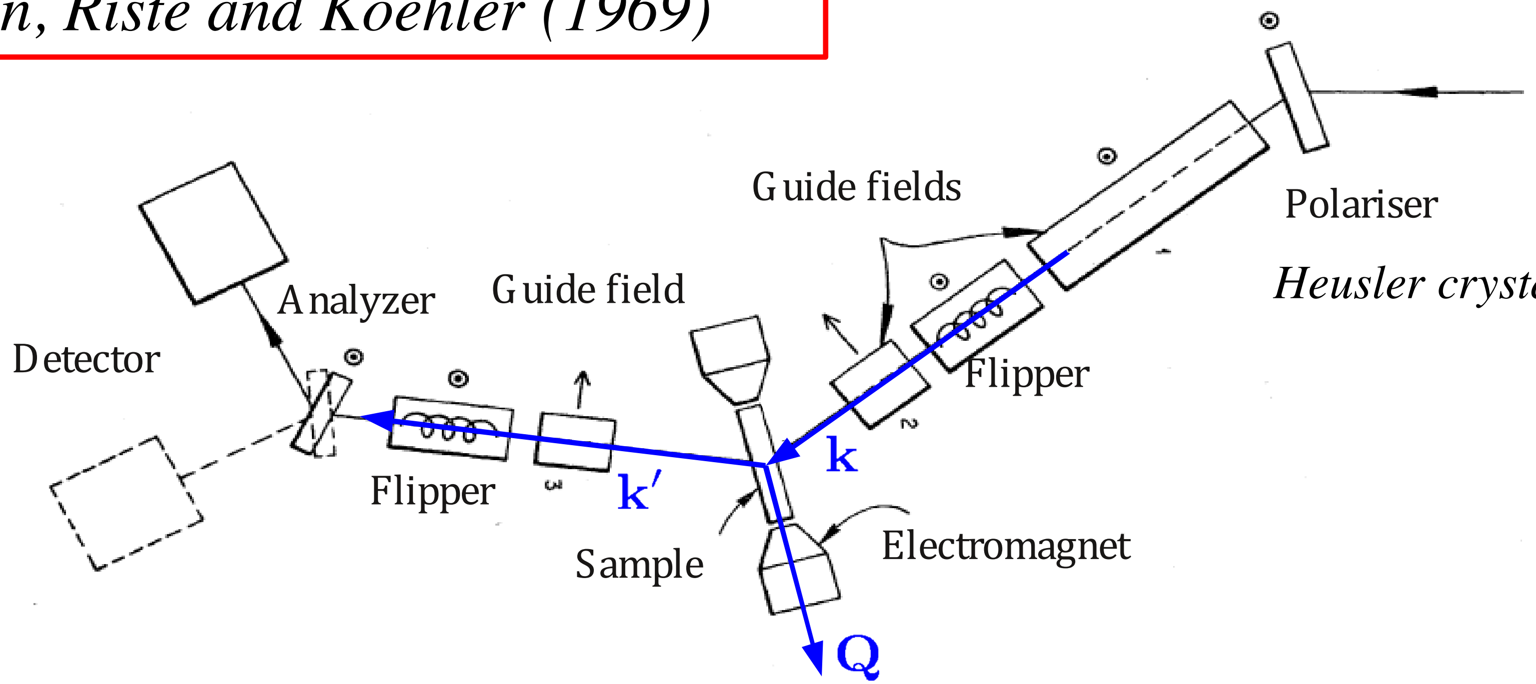
=> Relaxation times



Larmor diffraction
absolute *d*-spacings
with $<10^{-4}$ accuracy
with π flip $\Leftrightarrow$ reversal of B
spin-echo $\tau(\varphi)$
θ
sample
k
k'
G
B
φ
Larmor diffraction
$\theta(\varphi)$
P
A

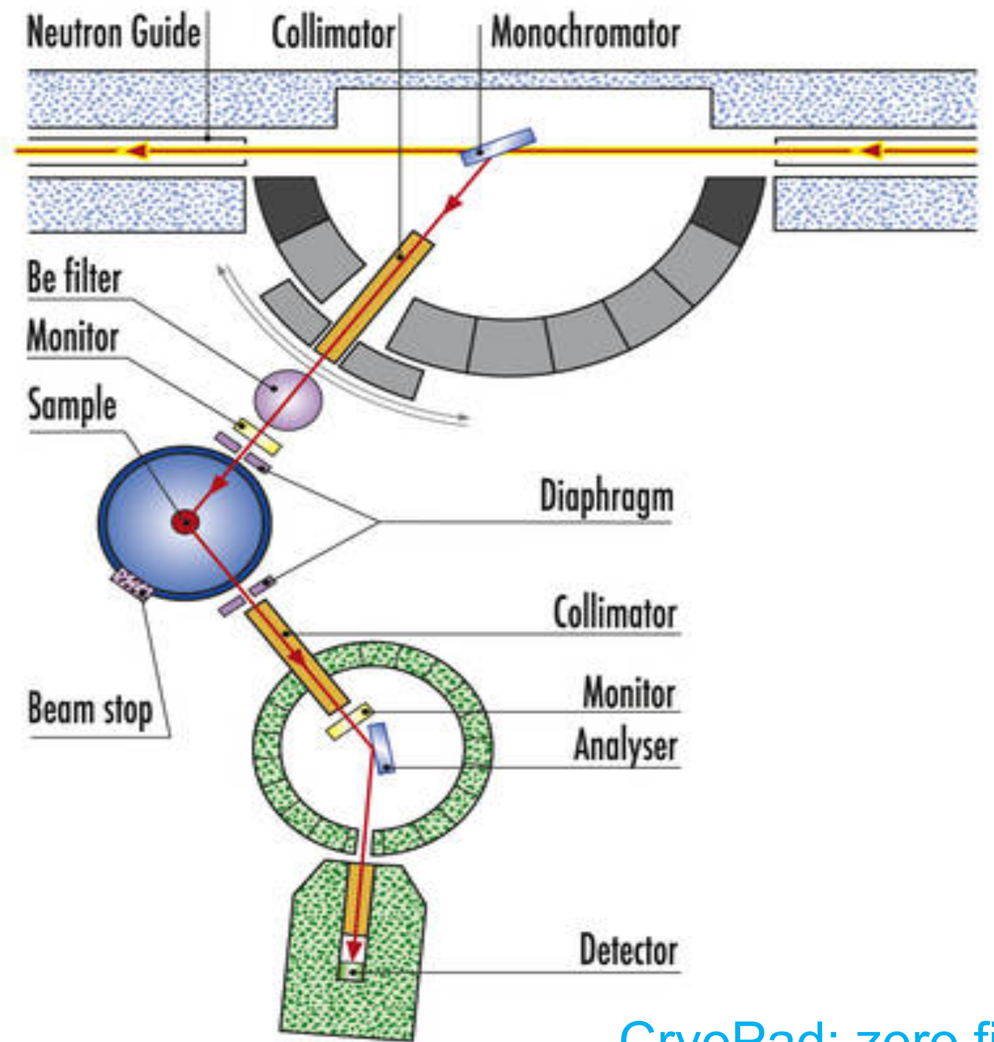
Triple axis instrument
with polarization analysis

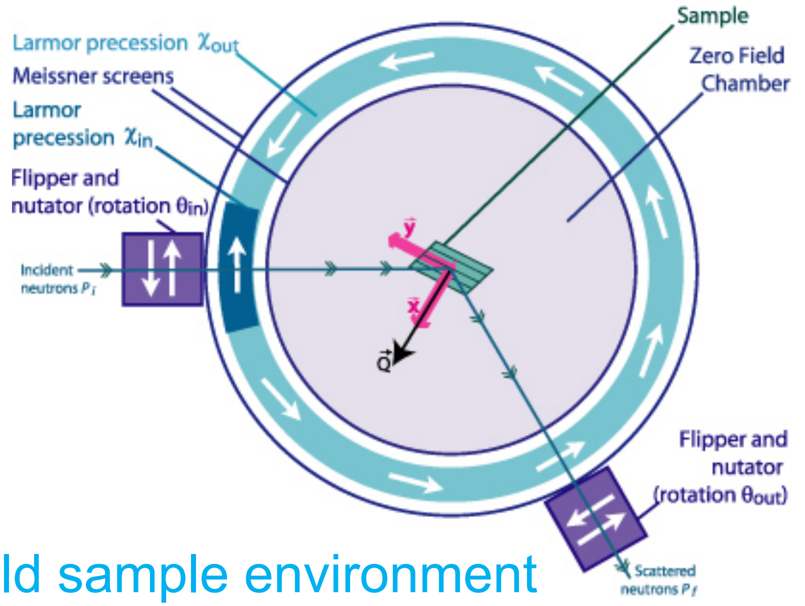
Moon, Riste and Koehler (1969)



Triple axis instrument with polarization analysis

IN12 @ ILL





CryoPad: zero field sample environment

Outline

- **Neutron spins in magnetic fields**
  - experimental devices => instruments
- **Scattering and Polarization**
  - spin-dependent nuclear interaction
  - magnetic interaction
- **Blume-Maleyev Equations**
  - examples
- **outlook for ESS**

Coherent nuclear scattering

Differential scattering cross section

now including initial and final spin states

$$\boxed{\frac{d\sigma}{d\Omega} = (\frac{m_n}{2\pi\hbar})^2 |< \mathbf{k}'\mathbf{S}'|V|\mathbf{k}\mathbf{S} > |^2}$$

1st Born approximation

$$V(\mathbf{r}) = \frac{2\pi\hbar^2}{m} b\delta(\mathbf{r} - \mathbf{R})$$

Point like nucleus

$$\langle \mathbf{k}'|V|\mathbf{k}\rangle = \frac{2\pi\hbar^2}{m} \underbrace{\sum_l b_l e^{i\mathbf{Q}\mathbf{R}_l}}_{= b(\mathbf{Q})}$$

Conservation of momentum and plane wave scattering

Scattering amplitude – transition matrix element

$$A(\mathbf{Q}) = < S_Z'|b(\mathbf{Q})|S_Z >= b(\mathbf{Q}) < S_Z'|S_Z >$$

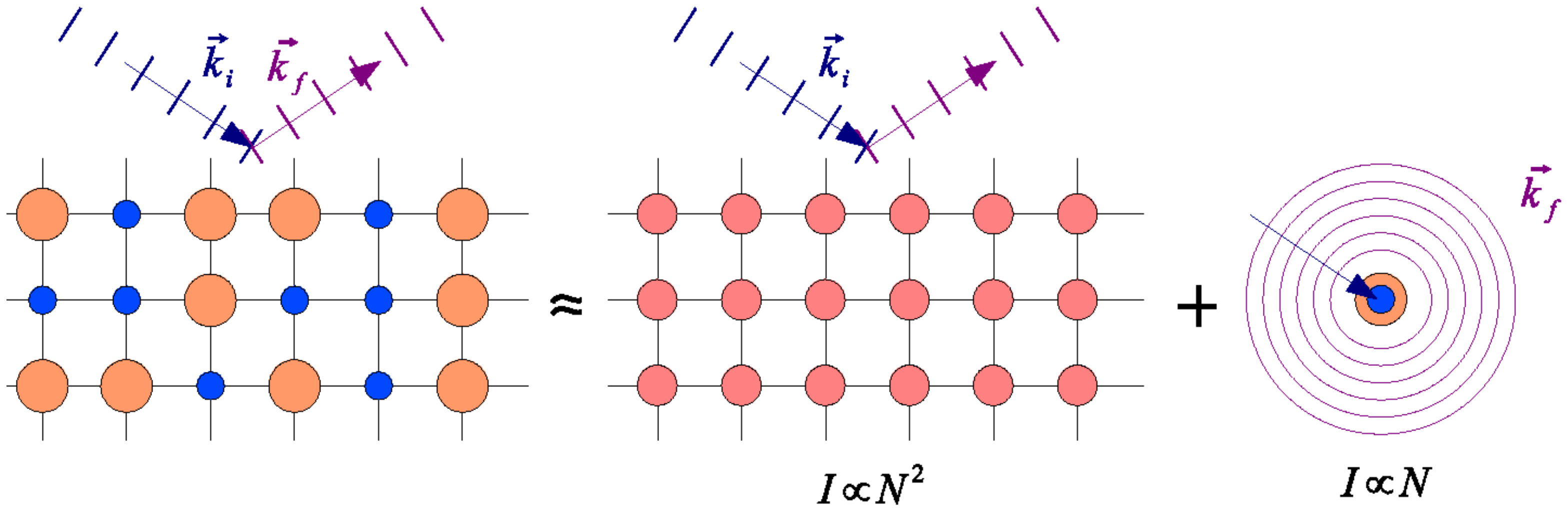
$$= b(\mathbf{Q}) \quad \text{no spin-flip}$$

$$= 0 \quad \text{spin-flip}$$

$$\frac{d\sigma}{d\Omega} = \overline{b}^2 \sum_{ll'} e^{i\mathbf{Q}(\mathbf{R}_l - \mathbf{R}_l')}$$

Coherent & incoherent scattering

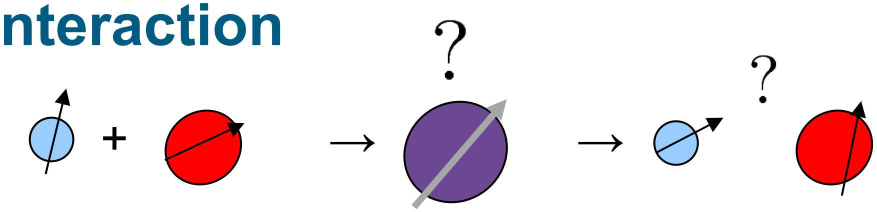
$$\frac{d\sigma}{d\Omega} = \bar{b}^2 \sum_{ll'} e^{\mathrm{i}\mathbf{Q}(\mathbf{R}_l - \mathbf{R}_l')} + N\left(\overline{b^2} - \bar{b}^2\right)$$



coherent

incoherent
spin and isotope

Spin dependent interaction



Spin $\quad 1/2 \quad I \quad\quad J = I \pm 1/2$

Two possibilities	Multiplicities $2J+1$	Probabilities	Scattering length
$J = J_+ = I + 1/2$ Triplet	$2I+2$	$p_+ = \dfrac{I+1}{2I+1}$	b_+
$J = J_- = I - 1/2$ Singlet	$2I$	$p_- = \dfrac{I}{2I+1}$	b_-
	$4I+2$		

$$\bar{b} = p_+ b_+ + p_- b_- = \frac{(I+1)b_+ + I b_-}{2I+1} \equiv A \qquad \frac{b_+ - b_-}{2I+1} \equiv B$$

$$\overline{b^2} = \textstyle\sum_i p_i b_i^2 = p_+ b_+^2 + p_- b_-^2$$

$$b^2_{\substack{spin\\inc}} \equiv \bar{b^2} - \bar{b}^2 = p_+ p_- (b_+ - b_-)^2$$

$$\frac{d\sigma}{d\Omega} = \bar{b}^2 \underbrace{\sum_{ll'} e^{\mathrm{i}\mathbf{Q}(\mathbf{R}_l - \mathbf{R}'_l)}}_{\text{coherent}} + \underbrace{N\left(\overline{b^2} - \bar{b}^2\right)}_{\text{incoherent}}$$

$$b^2_{\substack{isotope\\inc}} \equiv \bar{b^2} - \bar{b}^2 = c_A c_B (b_A - b_B)^2$$

How about spin states after scattering?

Spin dependent nuclear scattering amplitude

$$A(\mathbf{Q}) = \langle \mathbf{k}'\mathbf{S}' | A + B\hat{\sigma} \cdot \hat{\mathbf{I}} | \mathbf{k}\mathbf{S} \rangle$$

Spin operator $\hat{\boldsymbol{\sigma}} = \{\underline{\hat{\boldsymbol{\sigma}}}_x, \underline{\hat{\boldsymbol{\sigma}}}_y, \underline{\hat{\boldsymbol{\sigma}}}_z\}$

Pauli Matrices $\underline{\hat{\boldsymbol{\sigma}}}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ $\underline{\hat{\boldsymbol{\sigma}}}_y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}$ $\underline{\hat{\boldsymbol{\sigma}}}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

spin states, quantization axis z $|+\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ $|-\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$

$$\underline{\hat{\boldsymbol{\sigma}}}_x |+\rangle = |-\rangle \qquad \underline{\hat{\boldsymbol{\sigma}}}_x |-\rangle = |+\rangle$$

2/3 spinflip

$$\underline{\hat{\boldsymbol{\sigma}}}_y |+\rangle = \mathrm{i}|-\rangle \qquad \underline{\hat{\boldsymbol{\sigma}}}_y |-\rangle = -\mathrm{i}|+\rangle$$

$$\underline{\hat{\boldsymbol{\sigma}}}_z |+\rangle = |+\rangle \qquad \underline{\hat{\boldsymbol{\sigma}}}_z |-\rangle = -|-\rangle$$

1/3 non-spinflip

Spin dependent nuclear scattering amplitude

$$A(\mathbf{Q}) = \langle \mathbf{k}'\mathbf{S}' | A + B\hat{\sigma} \cdot \hat{\mathbf{I}} | \mathbf{k}\mathbf{S} \rangle$$

$I = 0$ $\quad$ $$A(\mathbf{Q}) = \langle S_z' | \bar{b} | S_z \rangle = \bar{b} \langle S_z' | S_z \rangle$$

$$\langle + | + \rangle = \langle - | - \rangle = 1$$

$$\langle + | - \rangle = \langle - | + \rangle = 0$$

No spin flip in absence of a nuclear spin

$I \neq 0$

$$A(\mathbf{Q})^{\mathrm{NSF}} = A + BI_z \qquad \text{for the } ++ \text{ and } -- \text{ case}$$

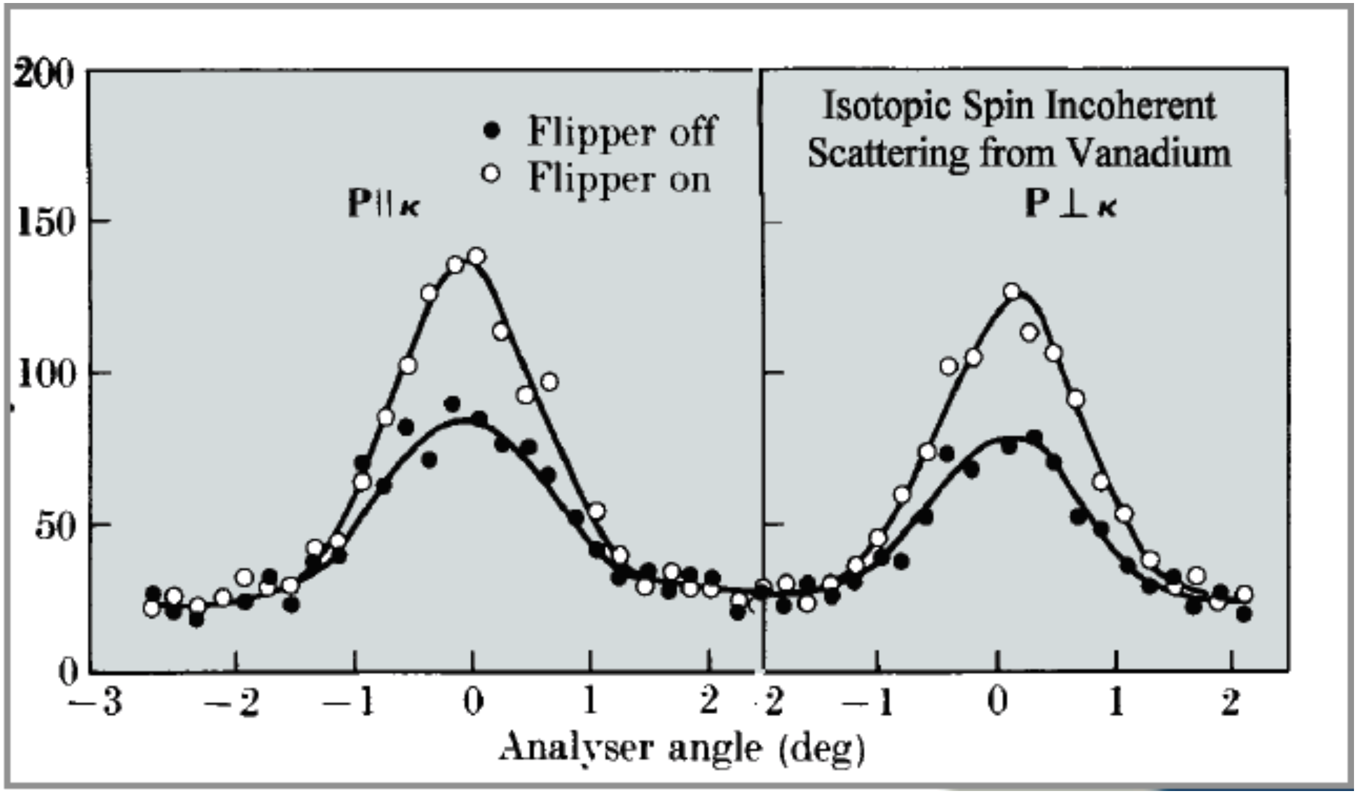
$$A(\mathbf{Q})^{\mathrm{SF}} = B(I_x + \mathrm{i}I_y) \qquad \text{for the } +- \text{ and } -+ \text{ case}$$

A perpendicular nuclear spin flips the neutron spin!

A parallel nuclear spins flip does not

2/3 of spin-incoherent scattering is spin-flip for disordered nuclear spins, independent of the direction of P

Moon, Riste and Koehler (1969)

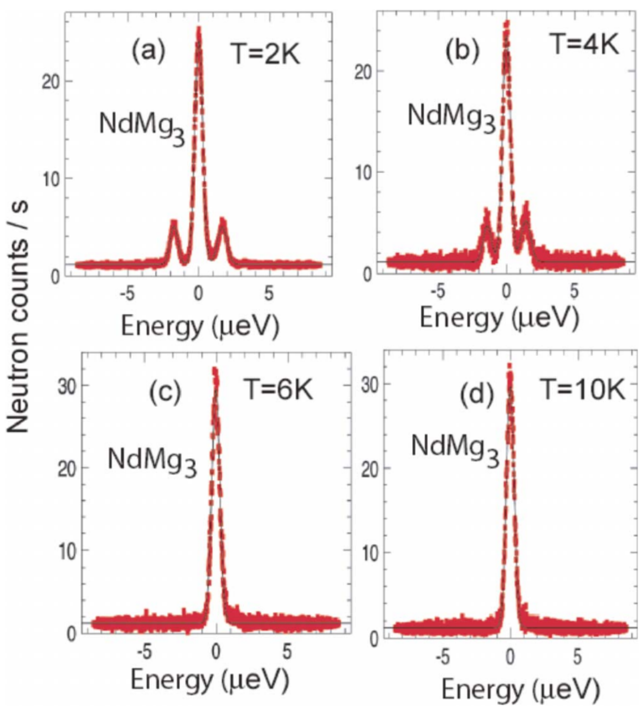


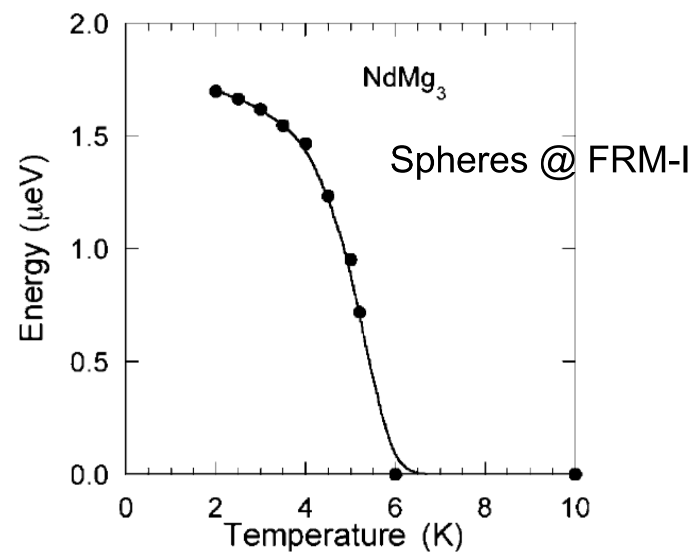
2/3 of spin-incoherent scattering is spin-flip independent of the direction of P

Spin dependent nuclear scattering

non spin-flip scattering is elastic
spin-flip scattering maybe inelastic

^{143}Nd $I = 7/2$ H=0, disorder — hyperfine splitting — magnetic order





Phys. Rev. B 78, 012411 (2008)

Quiz: why are only two side peaks visible at low T?

Separation of spin incoherent scattering

In the absence of nuclear polarization and magnetic scattering

$$\langle I_x \rangle = \langle I_y \rangle = \langle I_z \rangle = 0$$

$$\langle I_x^2 \rangle = \langle I_y^2 \rangle = \langle I_z^2 \rangle = \frac{1}{3}\langle I(I+1) \rangle$$

$$\frac{d\sigma}{d\Omega}^{\mathrm{NSF}} = \tfrac{1}{3} N B^2 \langle I(I+1) \rangle + \frac{d\sigma}{d\Omega}_{\mathrm{coh}} + \frac{d\sigma}{d\Omega}_{isotope-inc}$$

$$\frac{d\sigma}{d\Omega}^{SF} = \tfrac{2}{3} N B^2 \langle I(I+1) \rangle$$

only spin-incoherent

$$\frac{d\sigma}{d\Omega}_{\mathrm{spin\ inco}} = \frac{3}{2}\frac{d\sigma}{d\Omega}^{\mathrm{SF}}$$

$$\frac{d\sigma}{d\Omega}_{coh} + \frac{d\sigma}{d\Omega}_{isotope-inc} = \frac{d\sigma}{d\Omega}_{NSF} - \frac{d\sigma}{d\Omega}_{SF}$$

Polarization analysis: Spin-flip and non-spin-flip scattering

Separation of spin-incoherent and coherent nuclear scattering

Applications to hydrogeneous materials, soft matter, etc.

$$\frac{d\sigma}{d\Omega}^{N}_{Q,coh} + \underbrace{\frac{d\sigma}{d\Omega}^{N}_{isotop-inc}}_{small} = \frac{d\sigma}{d\Omega}^{NSF} - \frac{1}{2}\frac{d\sigma}{d\Omega}^{SF}$$

$$\frac{d\sigma}{d\Omega}_{spin-inc} = \frac{3}{2}\frac{d\sigma}{d\Omega}^{SF}$$

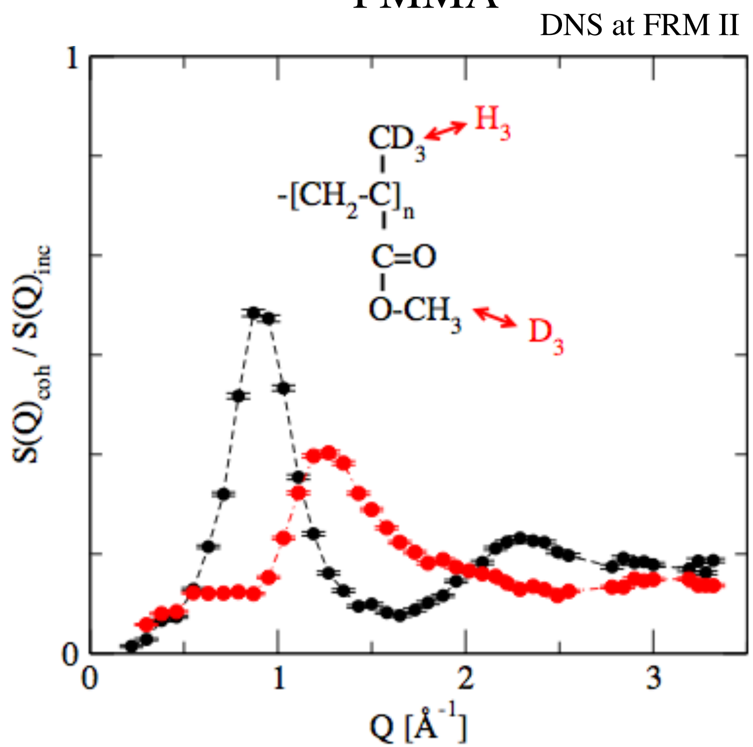
$$\sigma^{H}_{coh} = 1.75b \qquad \sigma^{H}_{inc} = 80.26b$$

$$b^{H}_{coh} = -3.74\,fm \qquad b^{D}_{coh} = +6.67\,fm$$

* Separating huge spin-incoherent background of H

* Intrinsic calibration

PMMA



from intensities to partial pair-correlation functions to compare with MD and MC simulations

A.C. Genix et al
Macromolecules 39, 3947 (2006)

About spin incoherent scattering:

(Spin) incoherent scattering does not contain phase information between distinct particles

→ single particle behavior is accessible
self correlation function, Chap. 11.2)

phase information on the identical particle: $\exp(iQ(R(t)-R_0(t_0))+i\omega(t-t_0))$

(Spin) incoherent scattering is isotropic *if integrated in energy*

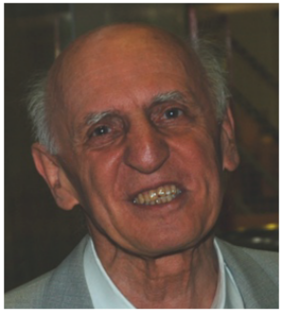
→ calibration of multi detector instruments
internal standard for absolute intensity measurements

Conservation of angular momentum

→ Spin incoherent scattering has an effect on the neutron spin

while isotope incoherent scattering does not

Liquid sodium at 840 K
(homepage Otto Schärpf)



Neutron scattering lengths and cross sections							
Isotope	**conc**	**Coh b**	**Inc b**	**Coh xs**	**Inc xs**	**Scatt xs**	**Abs xs**
Na	100	3.63	3.59	1.66	1.62	3.28	0.53

www.ncnr.nist.gov

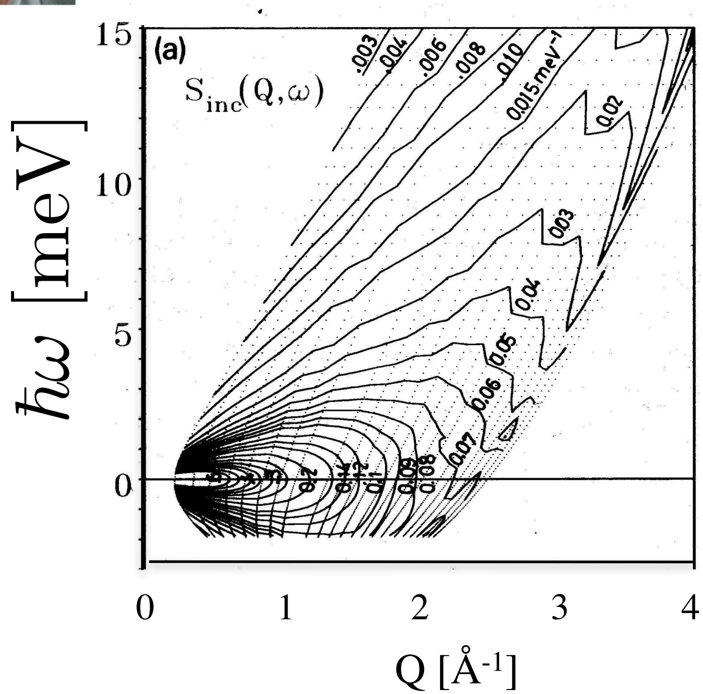
a good motivation to think about how to separate scattering

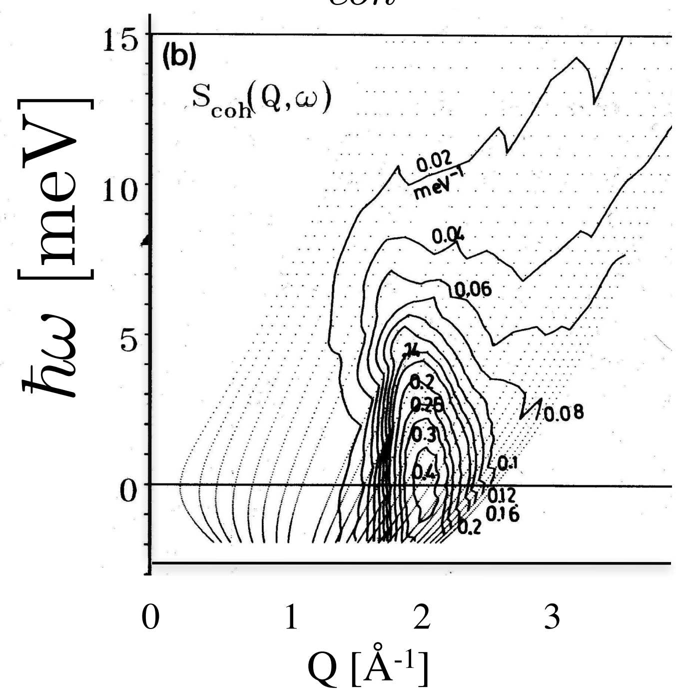
spin-incoherent

$$\sigma_{inco} = 1.62\text{b}$$

coherent

$$\sigma_{coh} = 1.66\text{b}$$





FT (self correlation)

single particle diffusion

FT (pair correlation)

collective behavior

precursors of Bragg scattering

Outline

- Introduction
- Neutron spins in magnetic fields
- Scattering and Polarization

Spin dependent nuclear scattering

magnetic scattering

Reminder

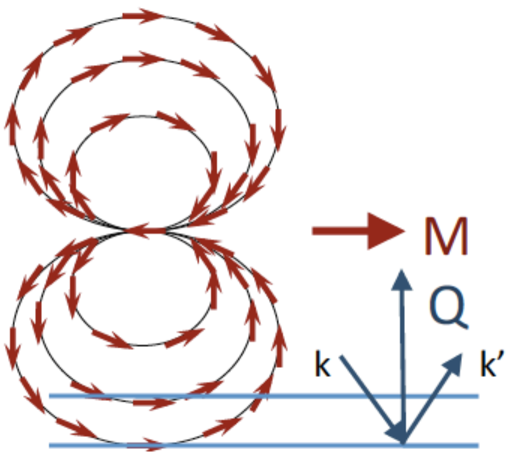
Neutron spins

dipole-dipole interaction with magnetic fields of unpaired electrons

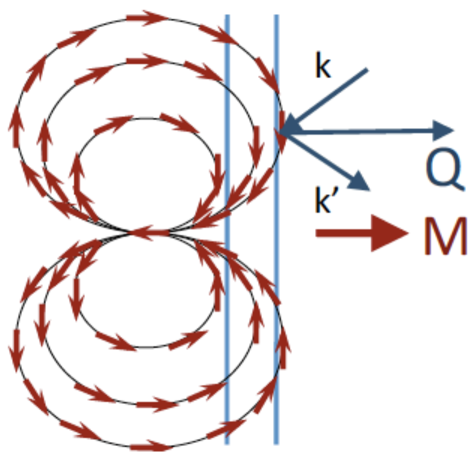
$$V_m = -\mu_{(n)} \cdot (\mathbf{B}_S + \mathbf{B}_L)$$

$$V_m = -(\gamma_n r_0/2)\hat{\boldsymbol{\sigma}} \cdot \hat{\mathbf{M}}^{\perp}_{\mathbf{Q}}$$

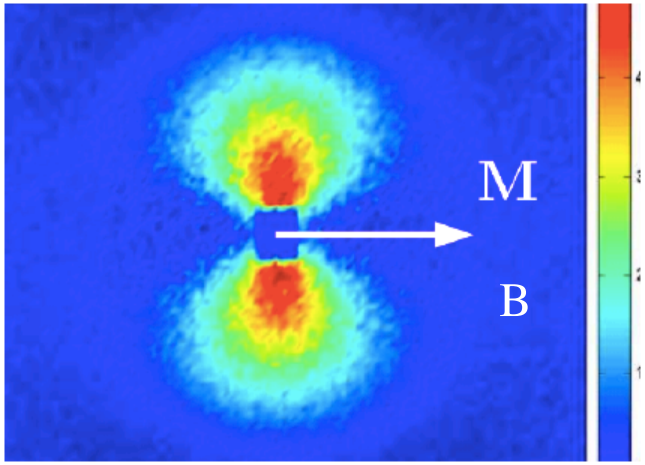
$$\mathbf{M}^{\perp}_{\mathbf{Q}} = \mathbf{e_Q} \times \mathbf{M_Q} \times \mathbf{e_Q}$$



Constructive interference



Destructive interference



initial and final spin states

$$
\begin{aligned}
A(\mathbf{Q}) &= \langle S_z'| - \frac{\gamma_{\mathrm{n}} r_0}{2\mu_{\mathrm{B}}} \hat{\boldsymbol{\sigma}} \cdot \mathbf{M}_{\perp}(\mathbf{Q}) | S_z \rangle \\
&= -\frac{\gamma_{\mathrm{n}} r_0}{2\mu_{\mathrm{B}}} \sum_{\alpha} \langle S_z' | \underline{\hat{\boldsymbol{\sigma}}}_{\alpha} | S_z \rangle \mathbf{M}_{\perp\alpha}(\mathbf{Q}) \qquad \alpha = x, y, \text{ or } z
\end{aligned}
$$

$$
\underline{\hat{\boldsymbol{\sigma}}}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \underline{\hat{\boldsymbol{\sigma}}}_y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix} \quad \underline{\hat{\boldsymbol{\sigma}}}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}
$$

Choosing *z* as quantization axis

$$
A(\mathbf{Q}) = -\frac{\gamma_{\mathrm{n}} r_0}{2\mu_{\mathrm{B}}} \cdot \begin{cases} \mathbf{M}_{\perp\mathbf{Q},z} & \text{for the } ++ \text{ NSF case} \\ -\mathbf{M}_{\perp\mathbf{Q},z} & \text{for the } -- \text{ NSF case} \\ \cancel{\mathbf{M}_{\perp\mathbf{Q},x}} - \mathrm{i}\mathbf{M}_{\perp\mathbf{Q},y} & \text{for the } +- \text{ SF case} \\ \cancel{\mathbf{M}_{\perp\mathbf{Q},x}} + \mathrm{i}\mathbf{M}_{\perp\mathbf{Q},y} & \text{for the } -+ \text{ SF case} \end{cases}
$$

coordinate system say x ll Q

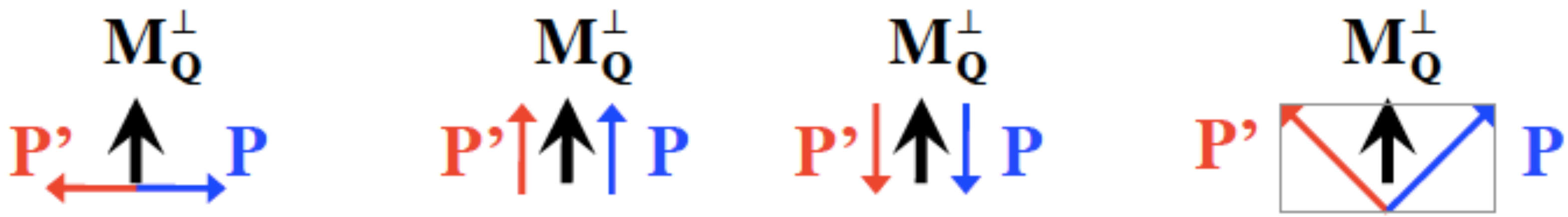
we have seen this before:

A perpendicular component flips the neutron spin!
A parallel component does not

direction of **P, M, Q** matters!

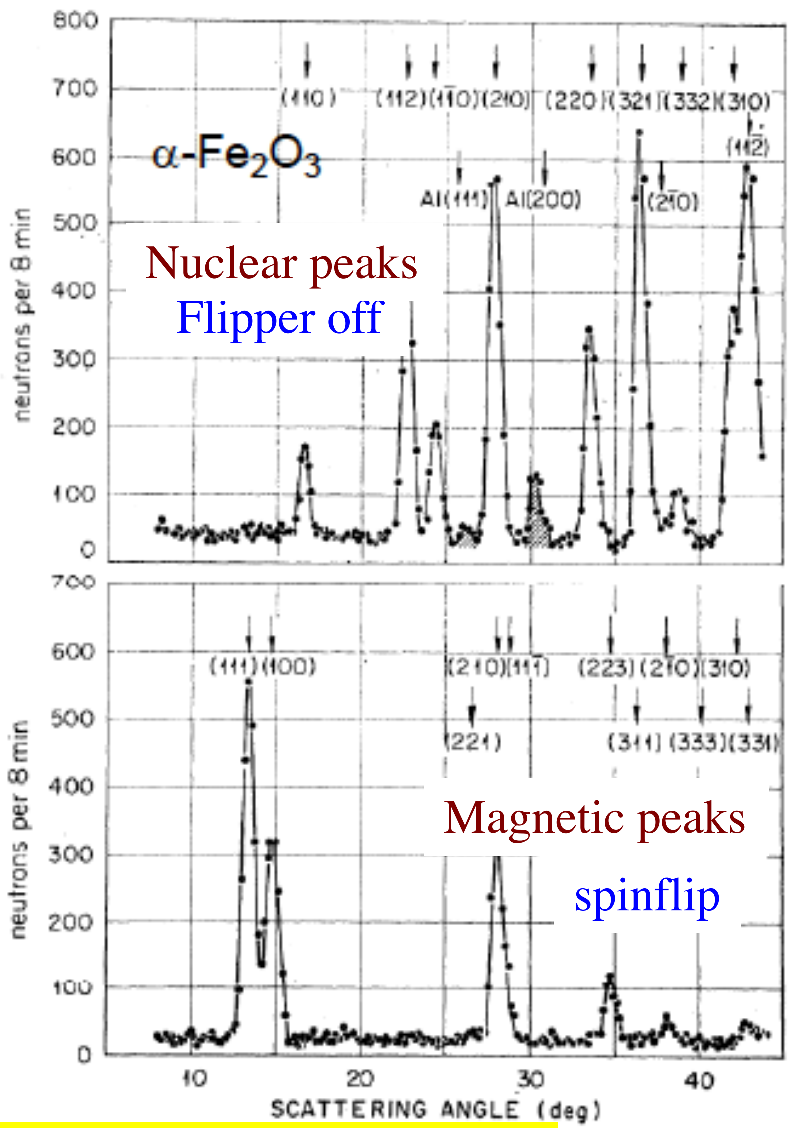
$$\langle +|\,\hat{\boldsymbol{\sigma}}\cdot\hat{\mathrm{M}}^{\perp}_{\mathbf{Q}}|+\rangle \;=\; \mathrm{M}^{\perp}_{z,\mathbf{Q}}$$

$$\langle -|\,\hat{\boldsymbol{\sigma}}\cdot\hat{\mathrm{M}}^{\perp}_{\mathbf{Q}}|+\rangle \;=\; \mathrm{i}\mathrm{M}^{\perp}_{y,\mathbf{Q}}$$



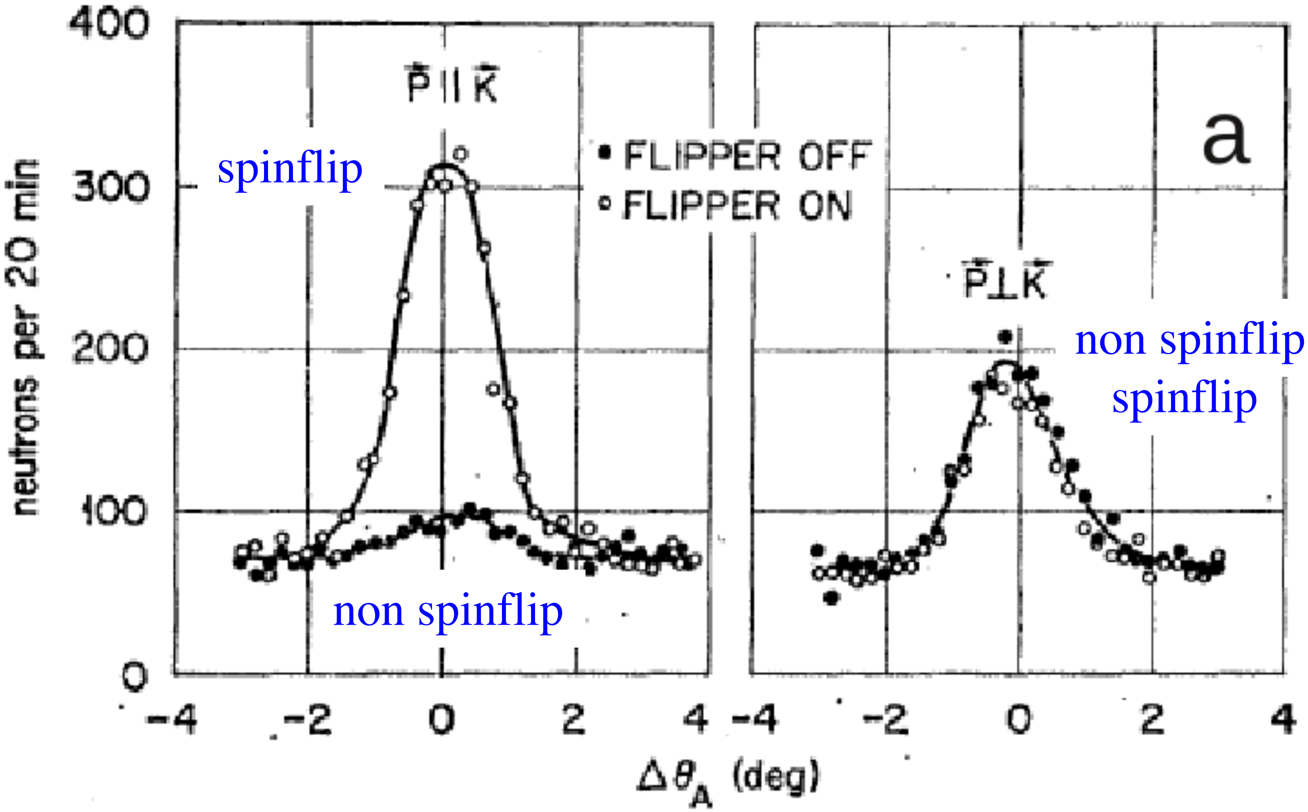
Separating nuclear and magnetic scattering

- Antiferromagnet: Bragg scattering



Moon, Riste and Koehler (1969)

Example: MnF_2 paramagnet



$$\frac{d\sigma}{d\Omega}_{\mathrm{mag}}^{\mathbf{M}_z^{\perp}} = \frac{d\sigma}{d\Omega}_{\perp}^{\mathrm{NSF}} - \frac{d\sigma}{d\Omega}_{\parallel}^{\mathrm{NSF}} = \frac{d\sigma}{d\Omega}_{\parallel}^{\mathrm{SF}} - \frac{d\sigma}{d\Omega}_{\perp}^{\mathrm{SF}}$$

Separation of magnetic scattering

by differential methods

XZ Difference: nuclear coherent and (spin) incoherent terms and background vanish

for paramagnets, antiferromagnets and powders, weak fields and isotropic M

$$\boxed{|\mathbf{M}_x| = |\mathbf{M}_y| = |\mathbf{M}_z|}$$

Polarization/Field	Spin-flip — If there is no chirality …
$\mathbf{P} \parallel x \parallel Q$	$\frac{2}{3}\frac{d\sigma}{d\Omega}_{\mathrm{inc}} + bg + \frac{d\sigma}{d\Omega}^{\mathbf{M}_y^{\perp}}_{\mathrm{mag}} + \frac{d\sigma}{d\Omega}^{\mathbf{M}_z^{\perp}}_{\mathrm{mag}}$
$\mathbf{P} \parallel z \perp Q$	$\frac{2}{3}\frac{d\sigma}{d\Omega}_{\mathrm{inc}} + bg + \frac{d\sigma}{d\Omega}^{\mathbf{M}_y^{\perp}}_{\mathrm{mag}}$

Separation of magnetic scattering

by differential methods

XZ Difference: nuclear coherent and (spin) incoherent terms and background vanish

Polarization/Field	Non spin-flip
$\mathbf{P} \parallel x \parallel Q$	$\frac{d\sigma}{d\Omega}_{\rm coh} + \frac{1}{3}\frac{d\sigma}{d\Omega}_{\rm inc} + bg$
$\mathbf{P} \parallel z \perp Q$	$\frac{d\sigma}{d\Omega}_{\rm coh} + \frac{1}{3}\frac{d\sigma}{d\Omega}_{\rm inc} + bg + \frac{d\sigma}{d\Omega}^{\mathbf{M}_z^\perp}_{\rm mag}$

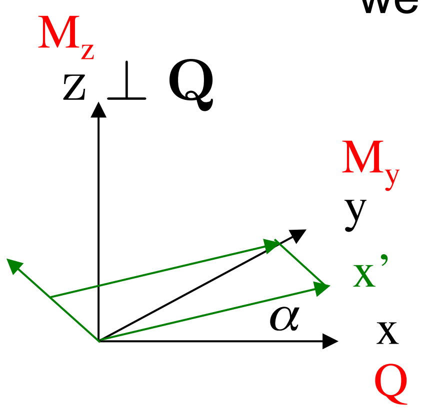
If there is no N-M interference …

XYZ-method for multi-detector instruments

Schärpf

If we cannot set **P** || to all **Q** simultaneously

we can still determine magnetic scattering by 3 measurements



$\cos^2\alpha + \sin^2\alpha = 1$

valid for all scattering angles

for paramagnets, powders, isotropy

$$|\mathbf{M}_x|^2 = |\mathbf{M}_y|^2 = |\mathbf{M}_z|^2$$

P||x P||y P||z

$$\frac{d\sigma}{d\Omega}_{\text{mag}} = 2\left(\underbrace{\frac{d\sigma}{d\Omega}^{SF}_{\mathrm{X\bar{X}}} + \frac{d\sigma}{d\Omega}^{SF}_{\mathrm{Y\bar{Y}}}}_{I_{P\parallel Q} + I_{P\perp Q}} - 2\,\frac{d\sigma}{d\Omega}^{SF}_{\mathrm{Z\bar{Z}}}\right) = -2\left(\frac{d\sigma}{d\Omega}^{NSF}_{\mathrm{XX}} + \frac{d\sigma}{d\Omega}^{NSF}_{\mathrm{YY}} - 2\,\frac{d\sigma}{d\Omega}^{NSF}_{\mathrm{ZZ}}\right) = 2|\mathbf{M}_i|^2$$

spin-flip *non-spin-flip*

$I_{P\parallel Q} + I_{P\perp Q} - 2\, I_{P\perp Q}$

$$|\mathbf{M}_y|^2 + 2|\mathbf{M}_z|^2 - 2|\mathbf{M}_y|^2 \qquad 0 + |\mathbf{M}_y|^2 - 2|\mathbf{M}_z|^2$$

Outline

- **Neutron spins in magnetic fields**
  - experimental devices => instruments
- **Scattering and Polarization**
  - spin-dependent nuclear interaction
  - magnetic interaction
- **Blume-Maleyev Equations**
  - examples
- **outlook for ESS**

Blume – Maleyev (1963)
general theory for polarized neutron scattering

… yields two expressions

for **scattering intensity**

$$\sigma_{\mathbf{Q}} = \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotope\text{-}inc}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

$$\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} = |N_{\mathbf{Q}}|^2$$

$$+ \underbrace{|\mathbf{M}^{\perp}_{\mathbf{Q}}|^2}_{\textit{magnetic}} + \underbrace{\mathbf{P}(N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}})}_{\textit{magnetic-nuclear interference}} + \underbrace{i\mathbf{P}(\mathbf{M}^{\perp}_{-\mathbf{Q}} \times \mathbf{M}^{\perp}_{\mathbf{Q}})}_{\textit{chirality}}$$

and **final polarized intensity**

$$\mathbf{P}'\sigma_{\mathbf{Q}} = \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotop\text{-}inc}} - \tfrac{1}{3}\mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$
$$+\mathbf{M}^{\perp}_{\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{-\mathbf{Q}}) + \mathbf{M}^{\perp}_{-\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}) - \mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}$$
$$+\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}} + i(\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} - \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}}) \times \mathbf{P} + i\mathbf{M}^{\perp}_{\mathbf{Q}} \times \mathbf{M}^{\perp}_{-\mathbf{Q}}$$

$$\mathbf{P}' = \sigma_{\mathbf{Q}}/\mathbf{P}'\sigma_{\mathbf{Q}}$$

Blume – Maleyev (1963)
general theory for polarized neutron scattering

… yields two expressions

$\mathbf{P} = 0$

for **scattering intensity**

$$\sigma_{\mathbf{Q}} = \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotope\text{-}inc}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

$$\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} = | N_{\mathbf{Q}} |^2$$

$$+ | \mathbf{M}^{\perp}_{\mathbf{Q}} |^2 + \mathbf{P}(N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}} N_{\mathbf{Q}}) + i\mathbf{P}(\mathbf{M}^{\perp}_{-\mathbf{Q}} \times \mathbf{M}^{\perp}_{\mathbf{Q}})$$

magnetic *magnetic-nuclear interference* *chirality*

and **final polarized intensity**

$$\mathbf{P}'\sigma_{\mathbf{Q}} = \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotop\text{-}inc}} - \tfrac{1}{3}\mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

$$+\mathbf{M}^{\perp}_{\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{-\mathbf{Q}}) + \mathbf{M}^{\perp}_{-\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}) - \mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}$$

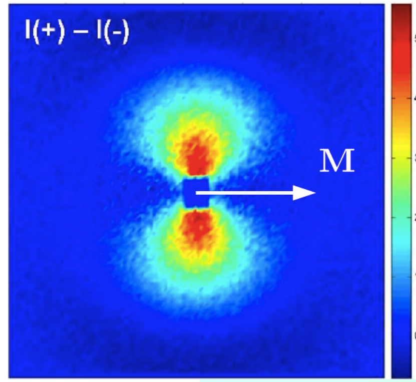
$$+\mathbf{M}^{\perp}_{\mathbf{Q}} N_{-\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}} N_{\mathbf{Q}} + i(\mathbf{M}^{\perp}_{\mathbf{Q}} N_{-\mathbf{Q}} - \mathbf{M}^{\perp}_{-\mathbf{Q}} N_{\mathbf{Q}}) \times \mathbf{P} + i\mathbf{M}^{\perp}_{\mathbf{Q}} \times \mathbf{M}^{\perp}_{-\mathbf{Q}}$$

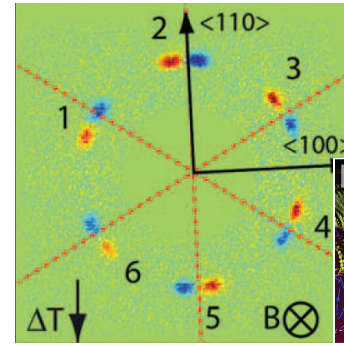
$\mathbf{P}' = \ ?$

$\mathbf{P}' = \sigma_{\mathbf{Q}} / \mathbf{P}'\sigma_{\mathbf{Q}}$

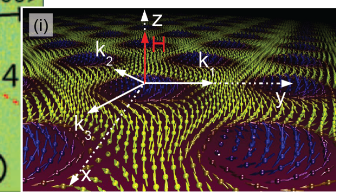
Blume – Maleyev general theory for polarized neutron scattering

Ref.[5, 6]





F. Jonietz et al. Science 2010



Skyrmions

… yields two expressions

for **scattering intensity**

Polarization reversal

$$\sigma_{\mathbf{Q}} = \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotope\text{-}inc}} + \sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

$$+ |\mathbf{M}^{\perp}_{\mathbf{Q}}|^2 + \mathbf{P}(N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}}) + i\mathbf{P}(\mathbf{M}^{\perp}_{-\mathbf{Q}} \times \mathbf{M}^{\perp}_{\mathbf{Q}})$$

magnetic *magnetic-nuclear interference* *chirality*

and **final polarized intensity**

$$\mathbf{P}'\sigma_{\mathbf{Q}} = \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotop\text{-}inc}} - \tfrac{1}{3}\mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

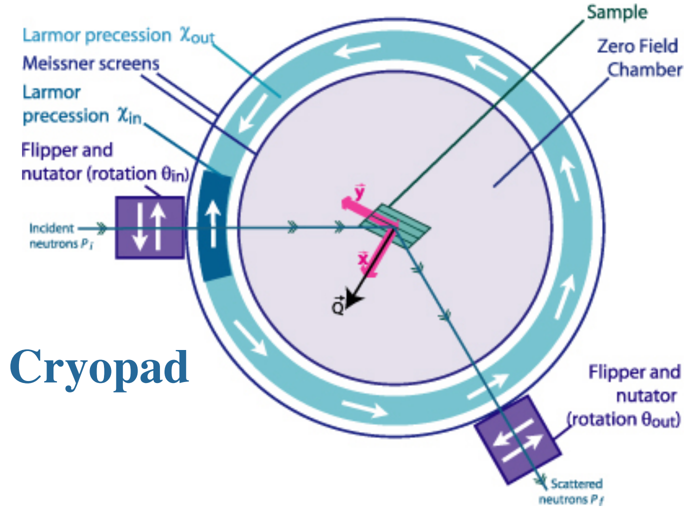
$$+\mathbf{M}^{\perp}_{\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{-\mathbf{Q}}) + \mathbf{M}^{\perp}_{-\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}) - \mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}$$

$$+\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}} + i(\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} - \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}}) \times \mathbf{P} + i\mathbf{M}^{\perp}_{\mathbf{Q}} \times \mathbf{M}^{\perp}_{-\mathbf{Q}}$$

creates **P’**

Spherical neutron polarimetry

Jane Brown



Cryopad

$$\mathbf{P}'\sigma = (|N|^2 + \boldsymbol{\mathcal{R}})\mathbf{P} + \mathbf{P}''$$

P” created polarization

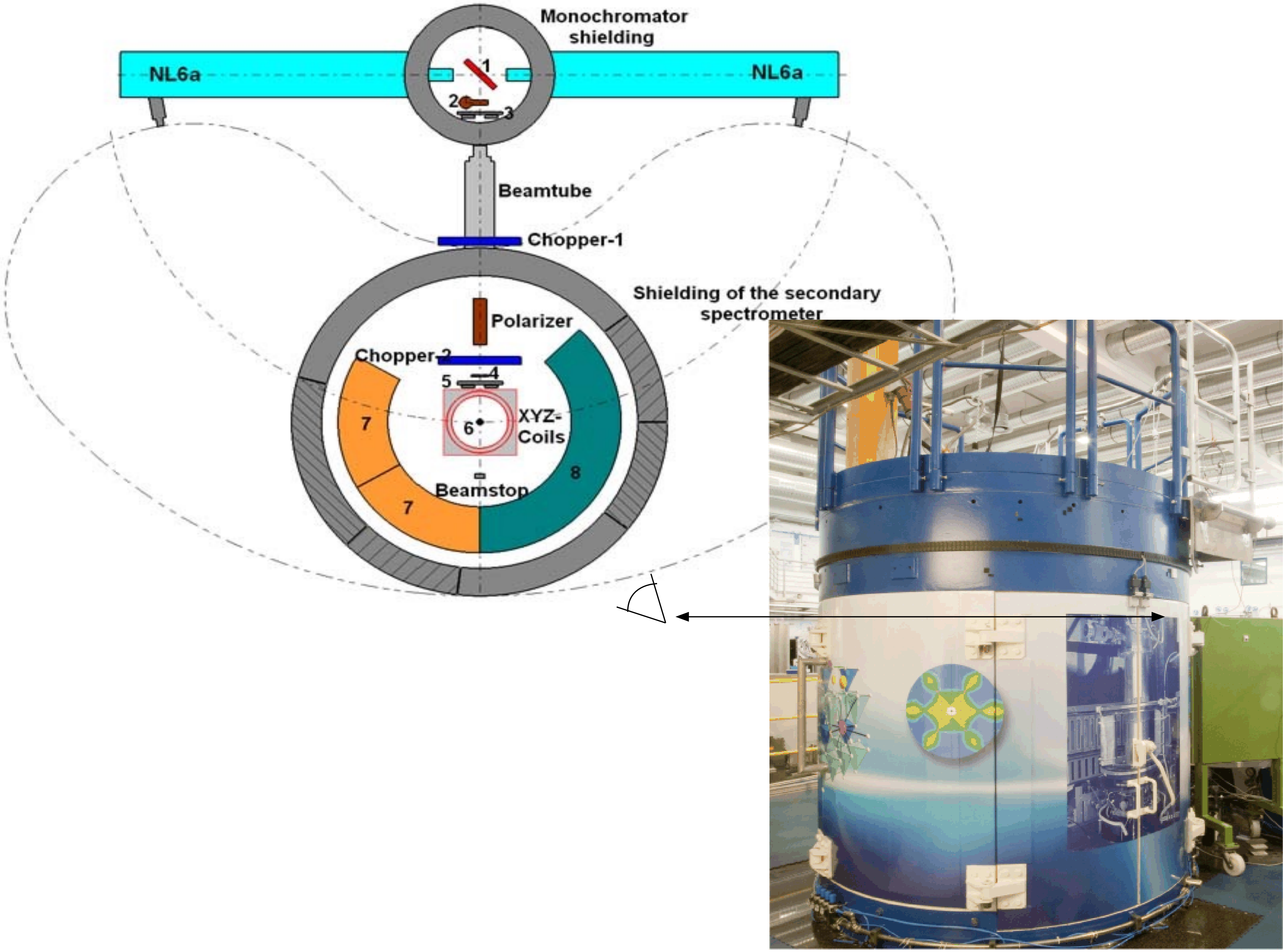
$$\boldsymbol{\mathcal{R}} = \begin{pmatrix} -|M_y|^2 - |M_z|^2 & 2\,Im\,[NM_z] & 2\,Im\,[NM_y] \\ -2\,Im\,[NM_z] & +|M_y|^2 - |M_z|^2 & 2\,Re\,[M_yM_z] \\ -2\,Im\,[NM_y] & 2\,Re\,[M_zM_y] & -|M_y|^2 + |M_z|^2 \end{pmatrix}$$

$$\mathbf{P}'' = (-2\,Im\,[M_yM_z]\,,\,2Re[NM_y]\,,\,2Re\,[NM_z])$$

Half-polarized experiments – polarization reversal

$$\begin{aligned} \sigma_{\mathbf{Q}}(\mathbf{P}) - \sigma_{\mathbf{Q}}(-\mathbf{P}) &= 2\mathbf{P}(N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}}) + 2\mathrm{i}\mathbf{P}(\mathbf{M}^{\perp}_{-\mathbf{Q}} \times \mathbf{M}^{\perp}_{\mathbf{Q}}) \\ &= -2\,Im\,[M_yM_z]_x\,,\,2Re[NM_y]\,,\,2Re\,[NM_z] \text{ for } \mathbf{P} = P_x,\ P_y, \text{and } P_z \end{aligned}$$

Multi detector instruments: DNS @ FRM-II



“XYZ- separation”

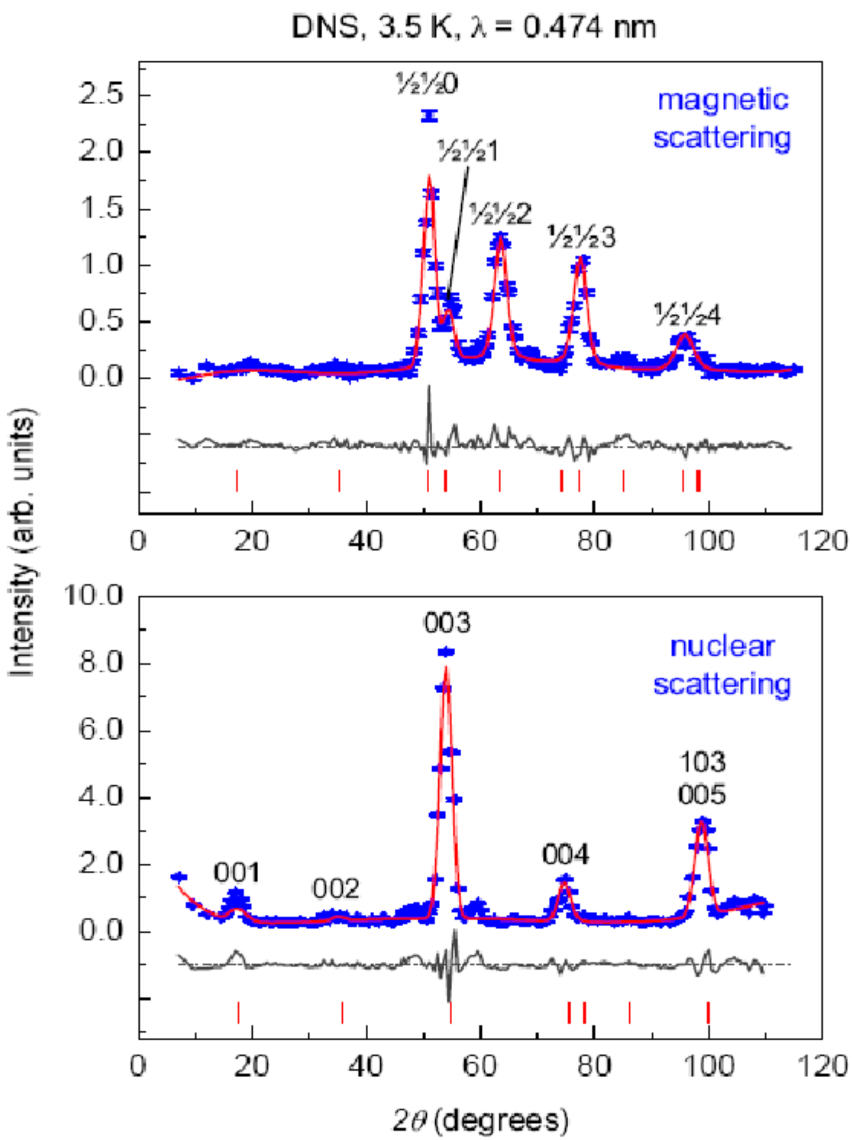
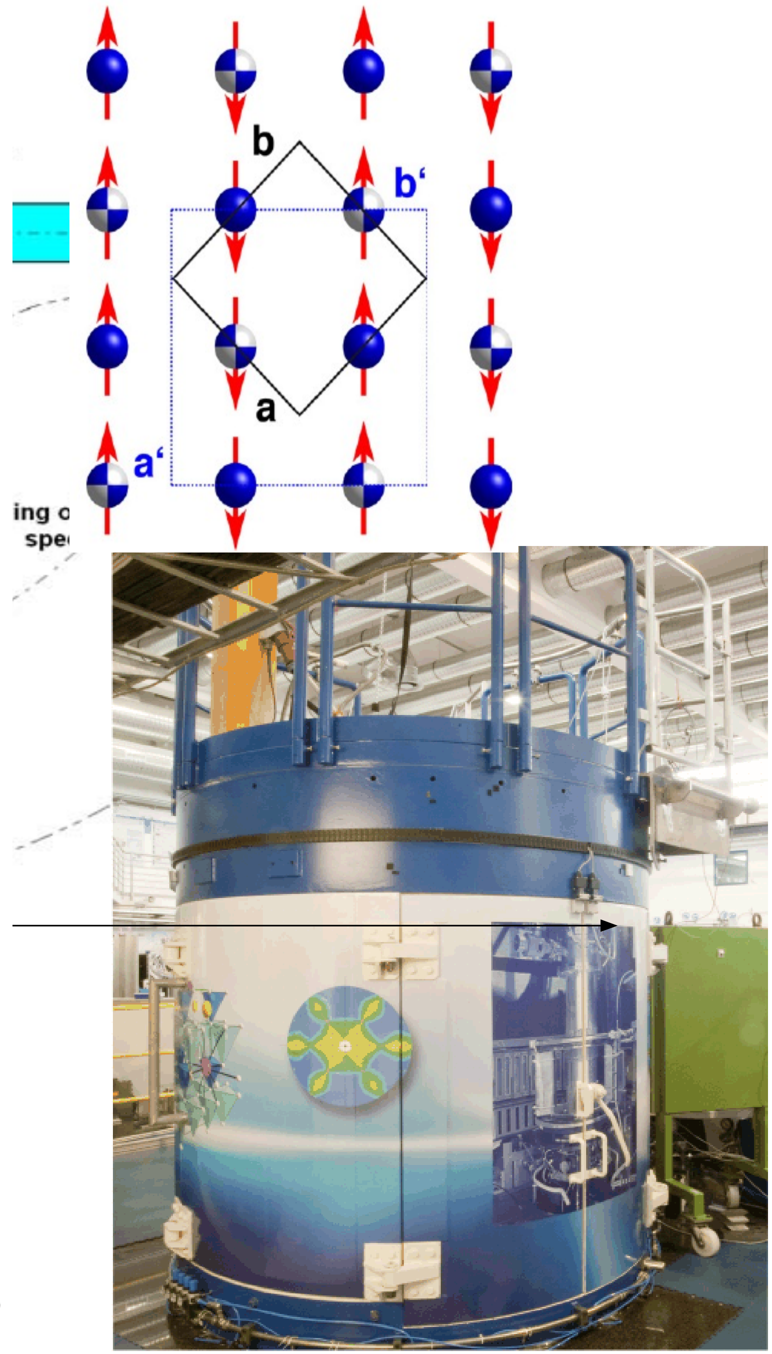
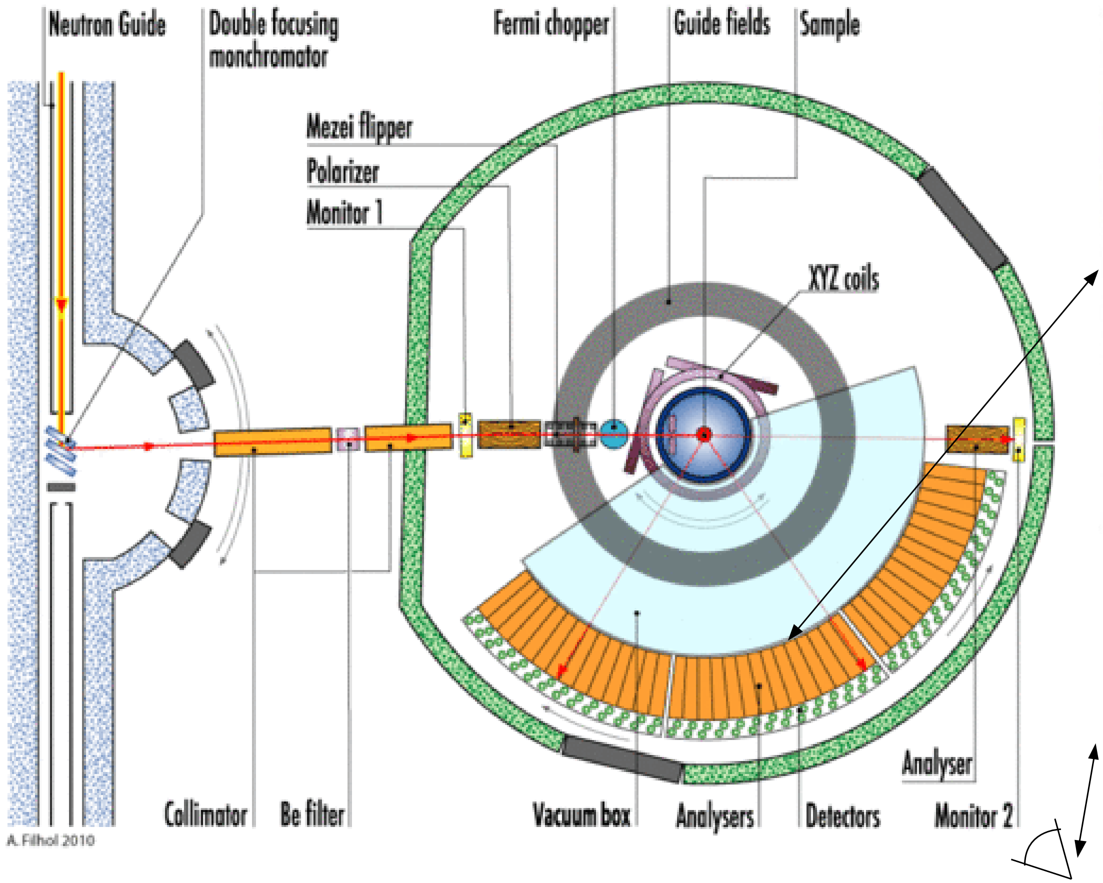
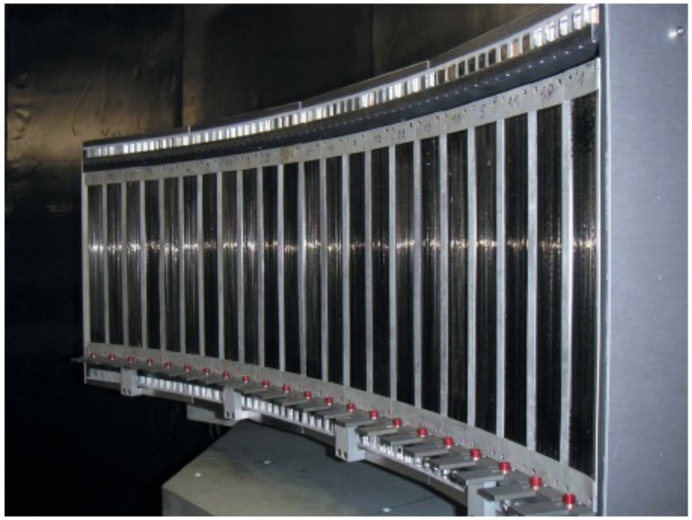


FIG. 3: Magnetic and nuclear reflections of Sr_2CrO_3FeAs (blue) and Rietveld fit (red) at 3.5 K measured at DNS [3].



Multi detector instruments: D7 @ ILL





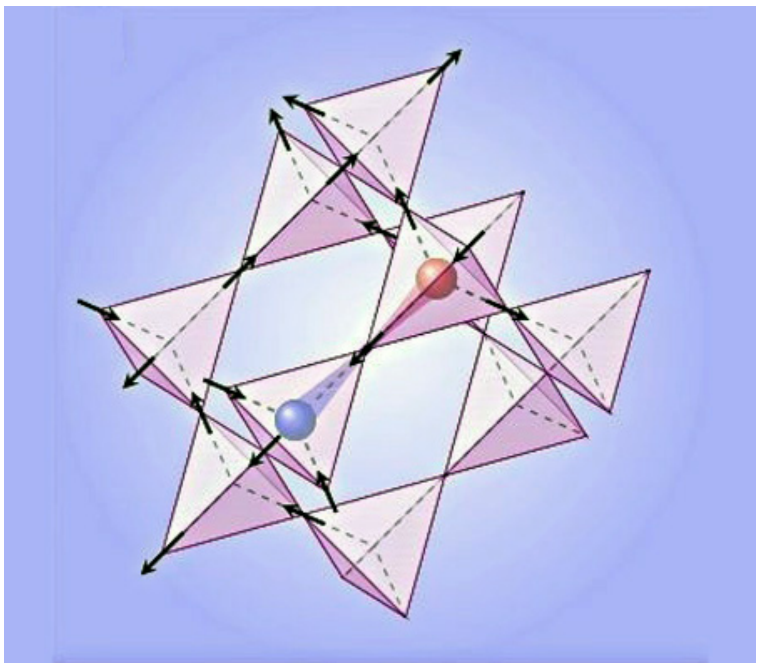
Both DNS and D7 have a time of flight option: inelastic scattering with polarization analysis

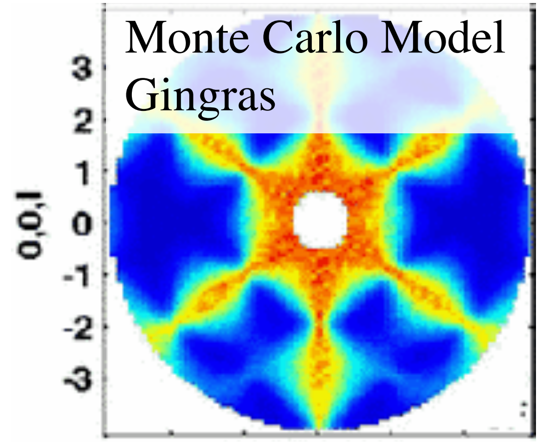
Single-crystal experiments

Magnetic monopoles proposed by Paul Dirac 1931
“topological monopoles in “spin-ice”

Magnetic Coulomb Phase in the Spin Ice $Ho_2Ti_2O_7$

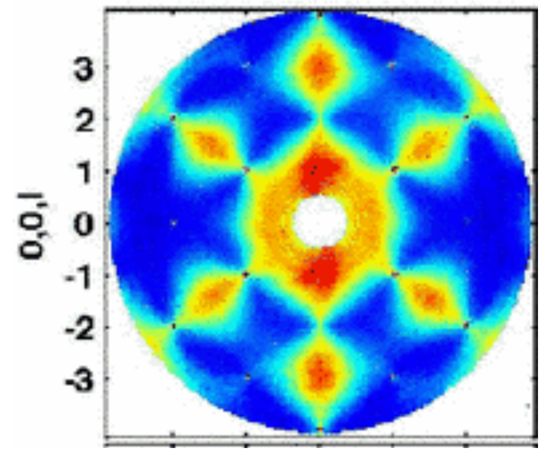
T. Fennell et al. Science 2009



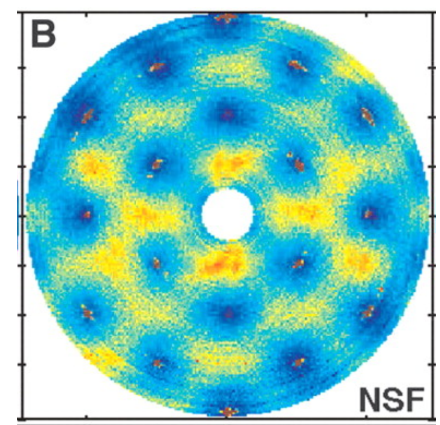


P || z

Spin-flip
Scattering
shows
pinch-points



NSF



h,h,0

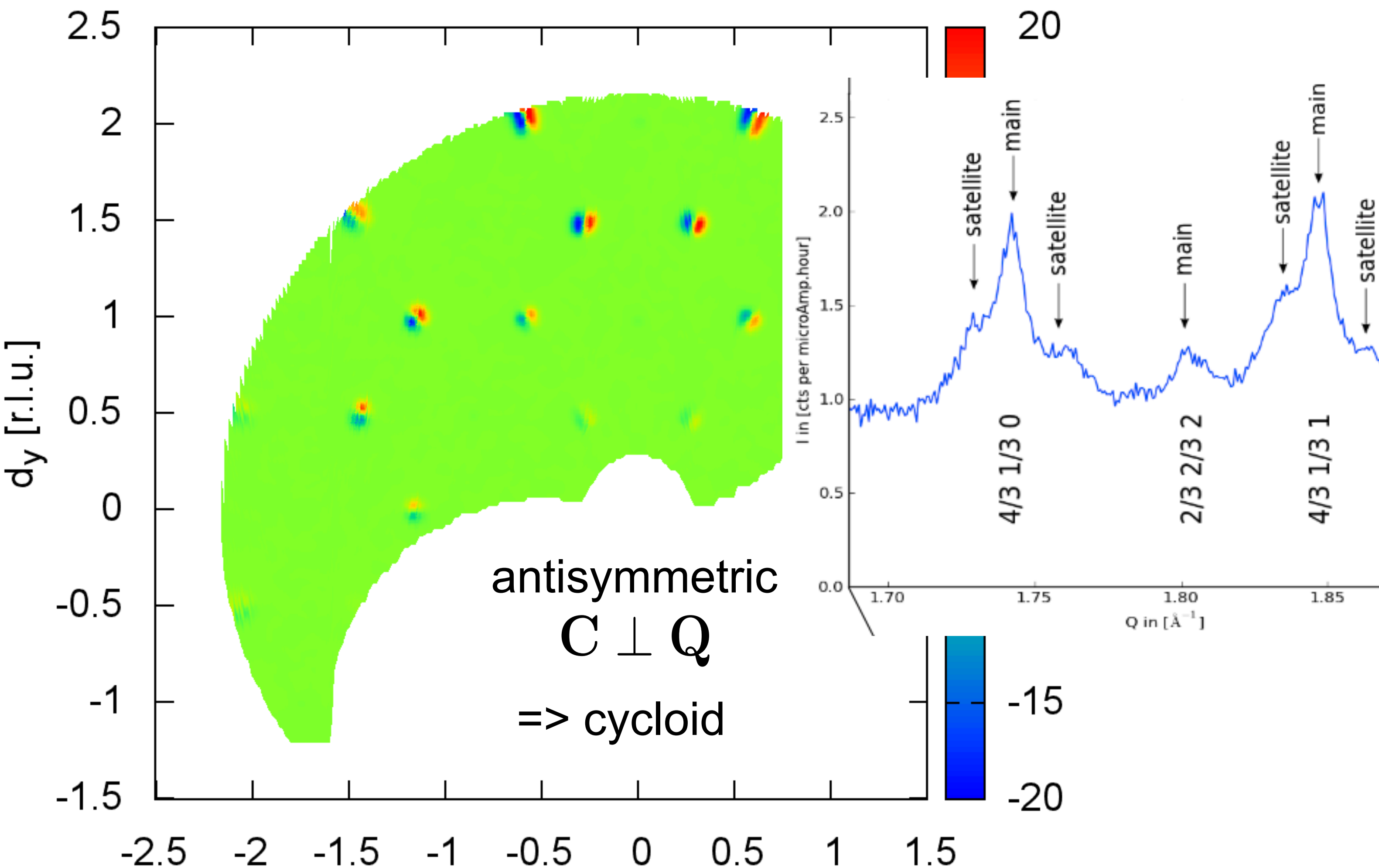
Poles apart. A pyramid with three ions pointing in (blue) acts as a north monopole; one with one ion pointing in (red) acts as a south monopole. By flipping other spins, the monopoles can be moved apart.

DNS
FRM II

XYZ for single crystals Journal of Physics: Conference Series **211** (2010) 012026

chiral scattering

T = 4 K



$$I_{chiral,x'} = 2\mathrm{i}(\mathbf{M}_\perp^\dagger \times \mathbf{M}_\perp)_{x'} = \Delta^{sf}_{x'} = \cos\alpha\,\Delta^{sf}_{x}$$

Depolarisation of the neutron spins are observed …

Higgs transition from a magnetic Coulomb liquid to a ferromagnet in $Yb_2Ti_2O_7$

depolarization of the neutron spins are observed with thermal hysteresis, indicating a first-order ferromagnetic transition. Our results are explained on the basis of a quantum spin-ice model, whose high-temperature phase is effectively described as a magnetic Coulomb liquid, whereas the ground state shows a nearly collinear ferromagnetism with gapped spin excitations.

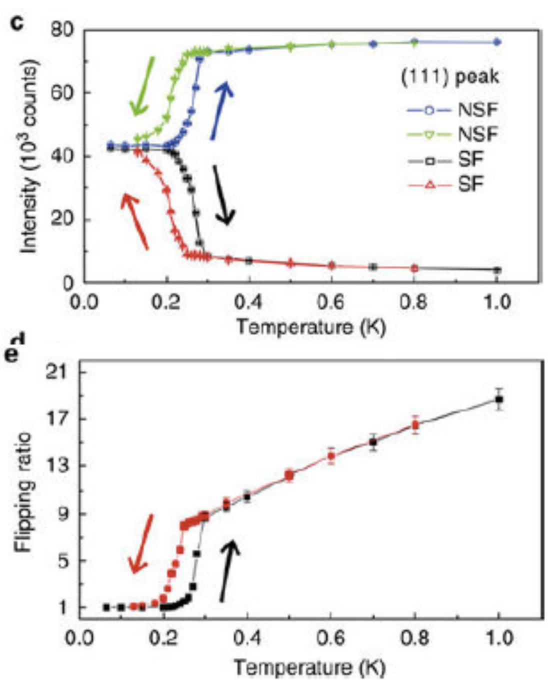
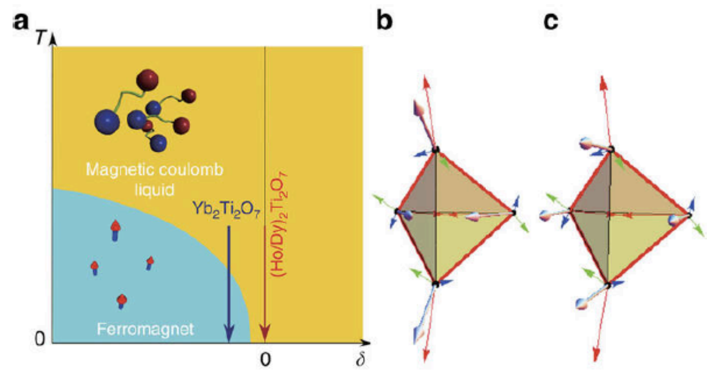


Figure 1: Schematic phase diagram and hypothetical ordered structures for $Yb_2Ti_2O_7$.



(**a**) Schematic phase diagram as a function of temperature T and the relative strength δ of the U(1)-symmetric planar exchange interaction and the Ising exchange. A first-order Higgs transition appears between a Coulomb liquid phase and a Higgs phase of magnetic monopoles. The other two model parameters q and K in equation (2) are assumed to be negligibly small for dipolar spin ice $(Dy/Ho)_2Ti_2O_7$, whereas they are finite for $Yb_2Ti_2O_7$ as obtained in the present work. Monopoles (blue balls) and antimonopoles (red balls) are illustrated for both the phases. In the magnetic Coulomb liquid phase (yellow), magnetic monopoles are carried by pseudospin-1/2 fractionalized gapped spinon excitations out of quasi-degenerate spin-ice manifold, obeying a Coulombic law. In the Higgs phase (cyan), monopolar spinons are condensed to form local magnetic dipole moments (arrows) showing ferromagnetic long-range order. (**b**,**c**) Hypothetical ferromagnetically ordered structures of the pseudospins (**b**) and the magnetic moments (**c**) in the low-temperature Higgs phase. The finite planar components of the pseudospins are ascribed to a condensation of monopolar spinons in the U(1) gauge theory[2,28].

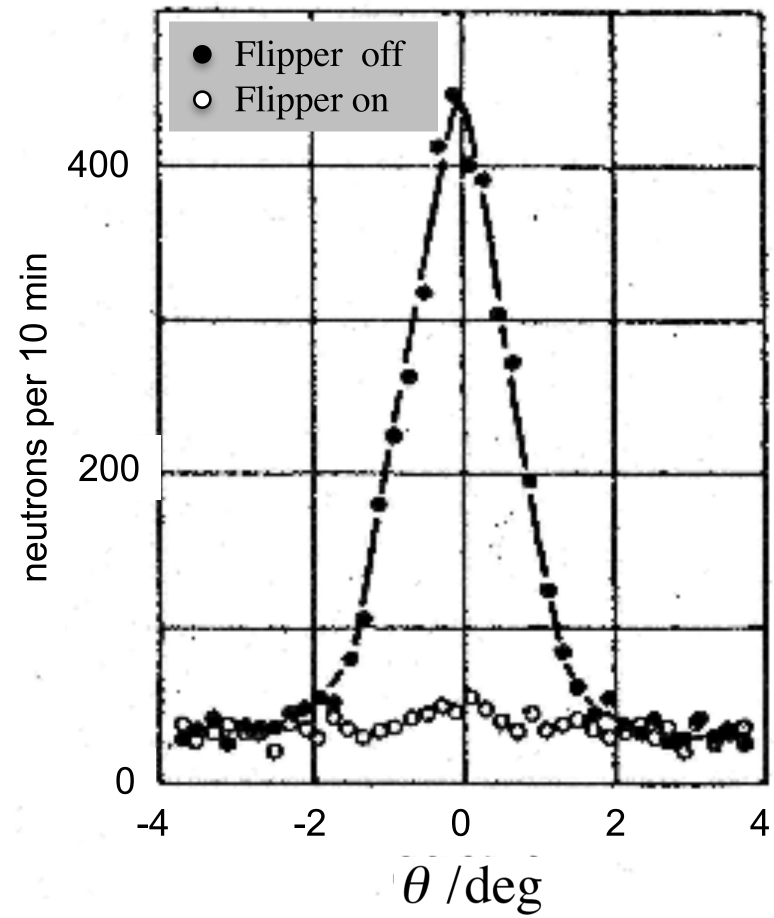
Moon, Riste & Koehler, Phys. Rev. 1969

Ni nuclear scattering

different isotopes - different b
isotopic incoherent scattering
no nuclear spins involved
=> no spin-flip scattering

$$\frac{d\sigma}{d\Omega}^{N}_{\mathbf{Q}} = \frac{d\sigma}{d\Omega}^{N}_{\mathbf{Q},coh} + \frac{d\sigma}{d\Omega}^{N}_{isotop-inc}$$

$$\mathbf{P}' = \mathbf{P}$$



Q: Ni is a ferromagnet, how can this picture be true?

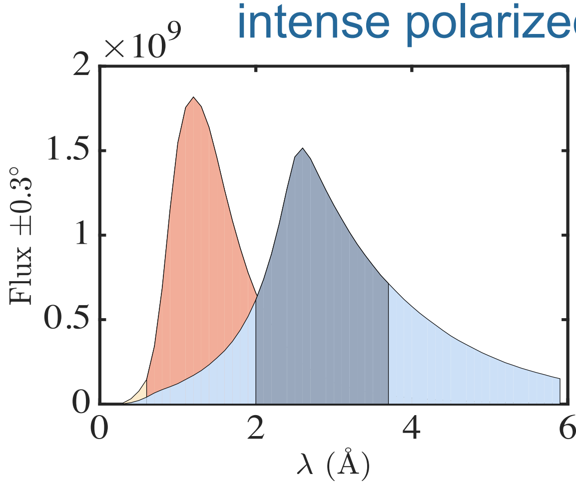
Outline

- **Neutron spins in magnetic fields**
  - experimental devices => instruments
- **Scattering and Polarization** *Moon, Riste, Koehler*
  - spin-dependent nuclear interaction
  - magnetic interaction
- **Blume-Maleyev Equations**
  - examples
- **outlook for ESS**

ESS polarised single crystal instrument MAGiC

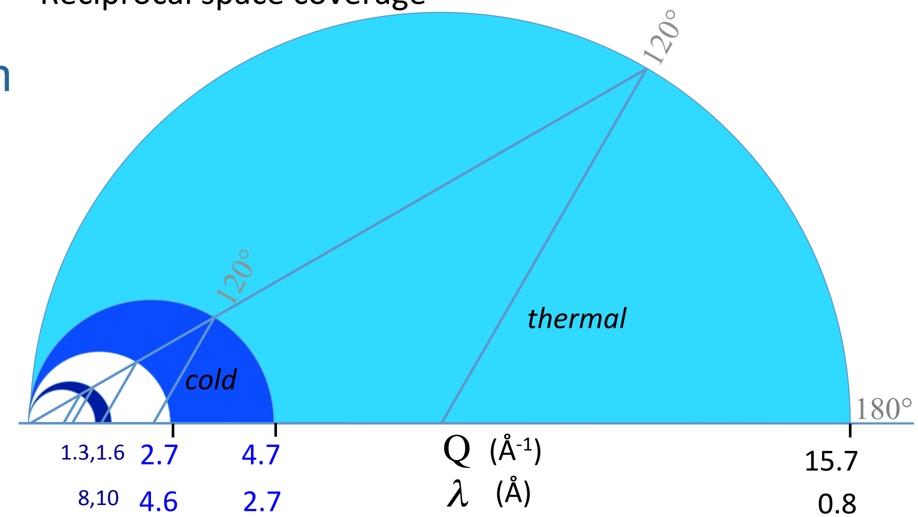
in construction LLB, JCNS, PSI
User operation 2023

intense polarized white beam

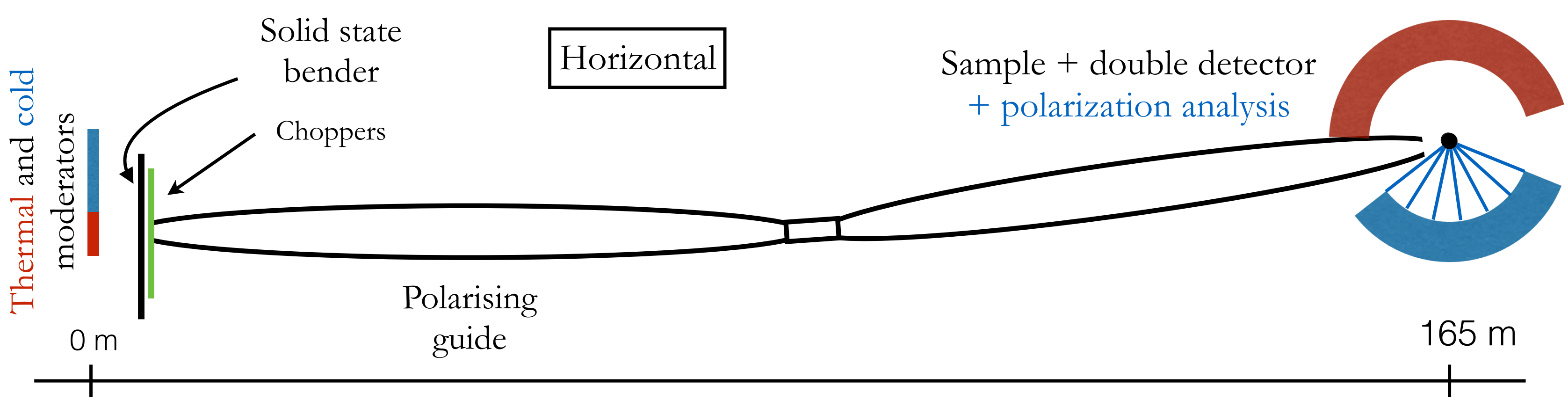


Neutron time-of-flight Laue instrument

Reciprocal space coverage



large position sensitive detector



Virtual experiments using MAGiC

Cases

HoMnO$_3$

BiFeO$_3$

Spin ice

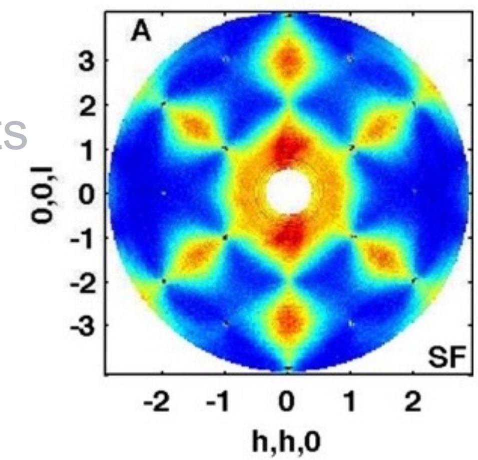
Bucky ball

Molecular magnets

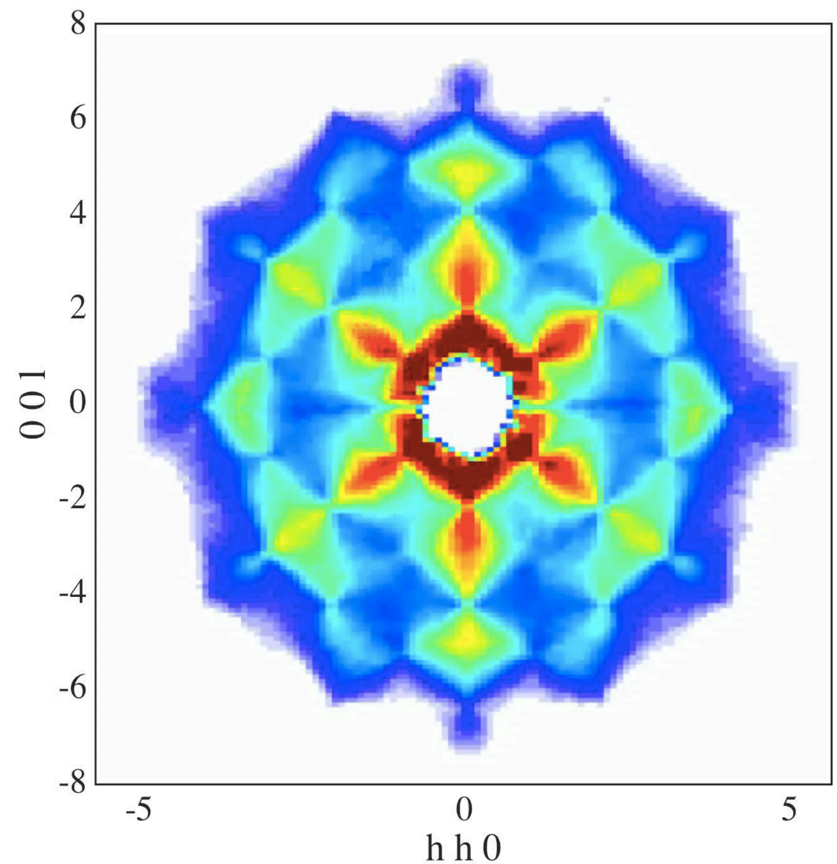
Ho$_2$Ti$_2$O$_7$

T. Fennell *et* al.
Science 2009

D7: 2×10^6 n/s/cm^2



MAGiC: $2 \times 10^{\mathbf{9}}$ n/s/cm^2



10 min & 10 mm^3

Virtual experiments using MAGiC

Cases

HoMnO$_3$

BiFeO$_3$

Spin ice

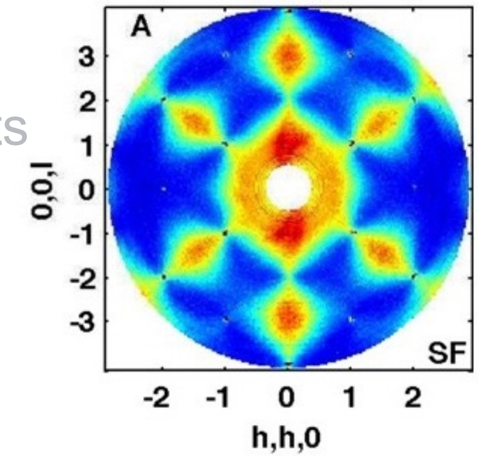
Bucky ball

Molecular magnets

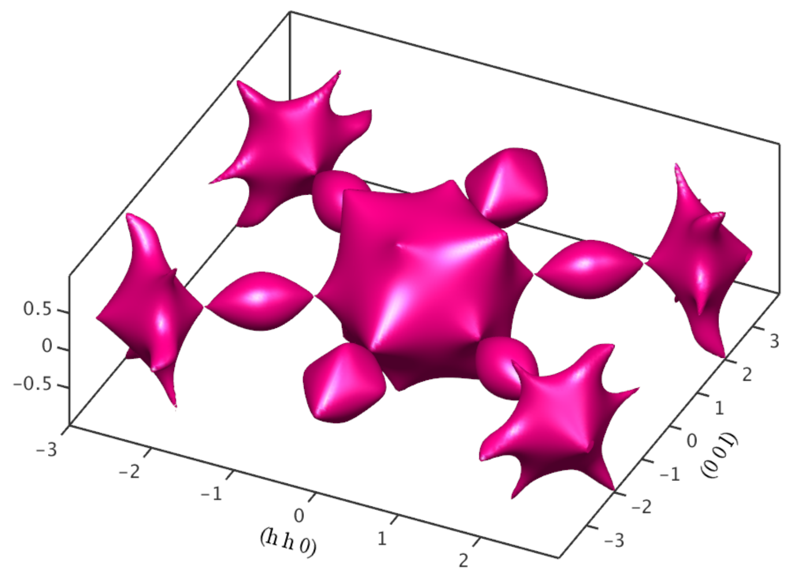
$Ho_2Ti_2O_7$

T. Fennell *et* al.
Science 2009

D7: 2×10^6 n/s/cm^2



MAGiC: 2×10^{9} n/s/cm^2

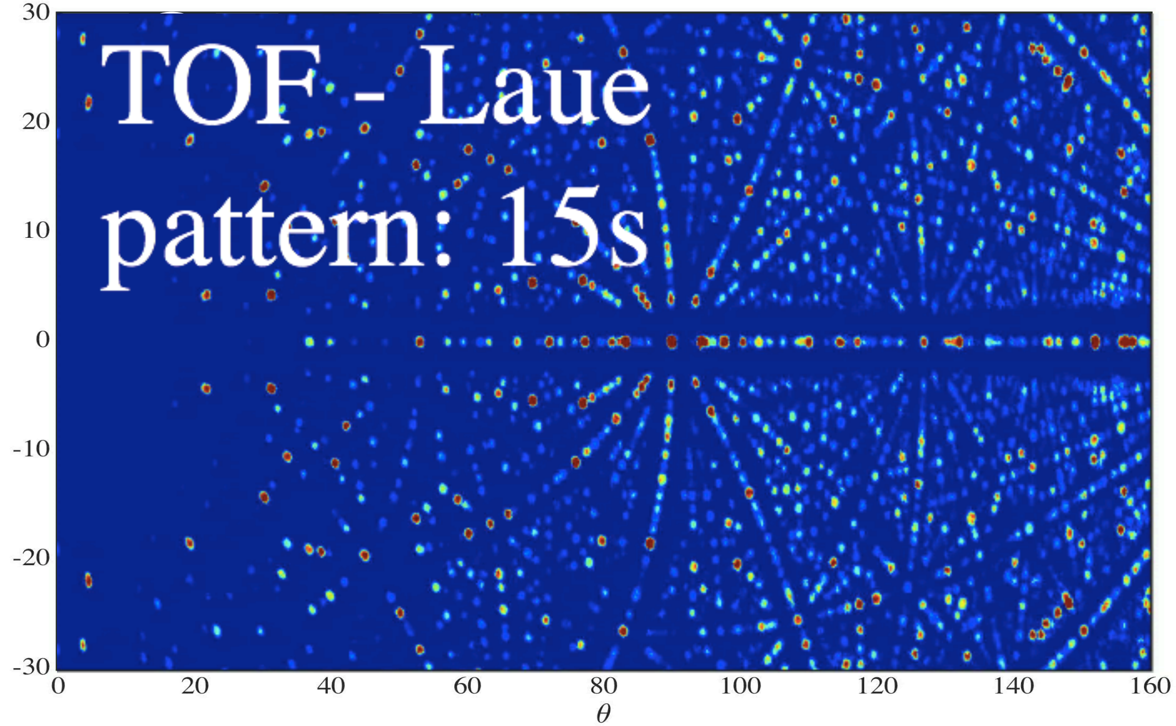


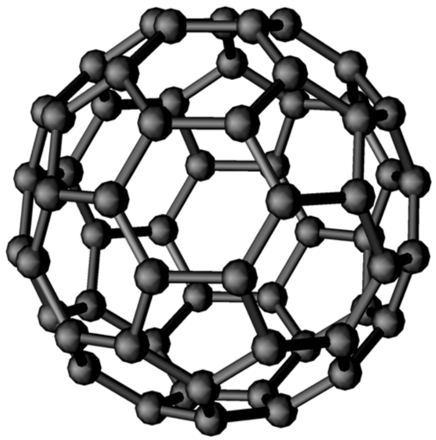
10 min & 10 mm^3

virtual MAGIC experiments

Cases

HoMnO$_3$
BiFeO$_3$
Spin ice
Bucky ball
Molecular magnets





C_{60}: a=14 Å, 1 mm^3 sample
Thermal spectrum @ full pulse length

Full data collection: 1mm^3 ~ minute

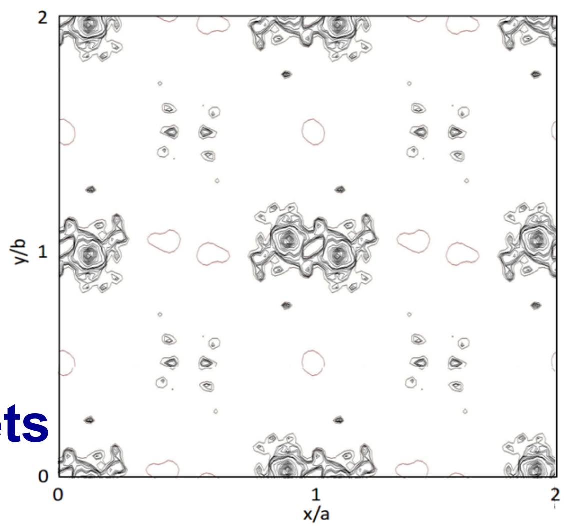
virtual MAGIC experiments

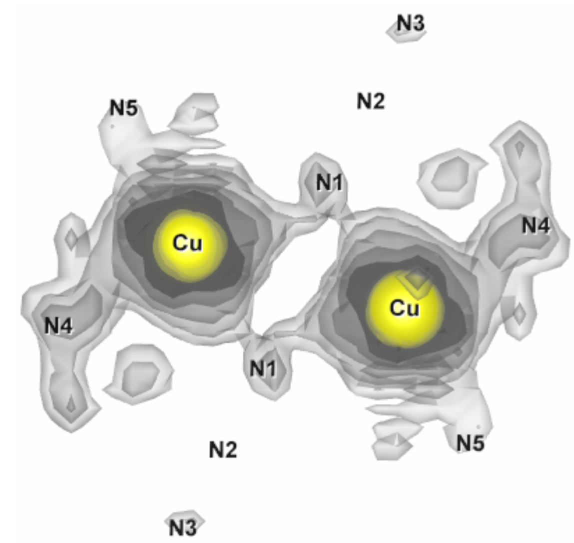
2 weeks D3   will be a few minutes on MAGIC

spin density map

Cases

HoMnO$_3$
BiFeO$_3$
Spin ice
Bucky ball
Molecular magnets





	DFT calculations (μ_B/ion)	MAGiC refinement (1σ)	Refinement Ref.[62]
Reflections used	NA	600	549
Cu	0.774	0.75 (1)	0.87(2)
N_1	0.069	0.08(1)	0.06(2)
N_2	-0.015	-0.014(10)	-0.04(2)
N_3	0.054	0.05 (2)	0.08(2)
N_4	0.067	0.07(1)	0.04(1)
N_5	0.048	0.06(2)	0.06(2)

ESS polarised single crystal instrument MAGiC

in construction LLB, JCNS, PSI
User operation 2023