Polarized Neutron Scattering

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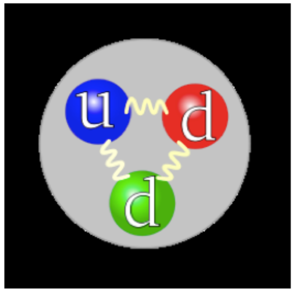
SwedNess/NNSP, Tartu Estonia, September 18th 2019

Repetition / Basics of

neutron properties

magnetic neutron scattering

The neutron



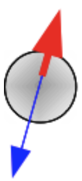
Source: Wikipedia

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**.



S

$$\mu_n = g_{\mathrm{n}} \cdot S \cdot \mu_{\mathrm{N}} \simeq \mp 1.913 \mu_{\mathrm{N}} = \pm \gamma_{\mathrm{n}} \mu_{\mathrm{N}}.$$

$$\mu_N \equiv \frac{e\hbar}{2m_p}$$

$$\frac{\mu_N}{\mu_B} = \frac{m_e}{m_n} \approx \frac{1}{1836}$$

Note the gyromagnetic factor $\gamma_n = -1.913$ for the neutron is negative, *i.e.* spin and magnetic moment are antiparallel

neutron interacts with nuclei

Its **spin** interacts with spin of nuclei

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

magnetic scattering

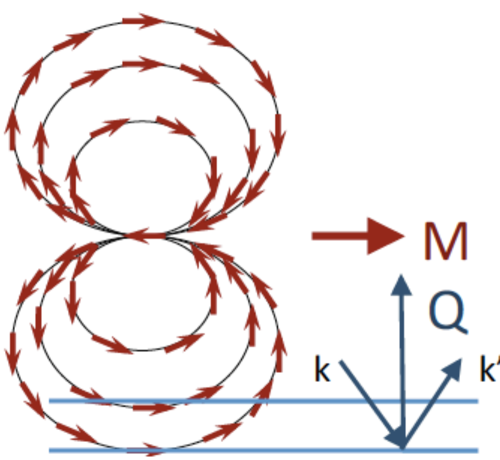
Neutron spins

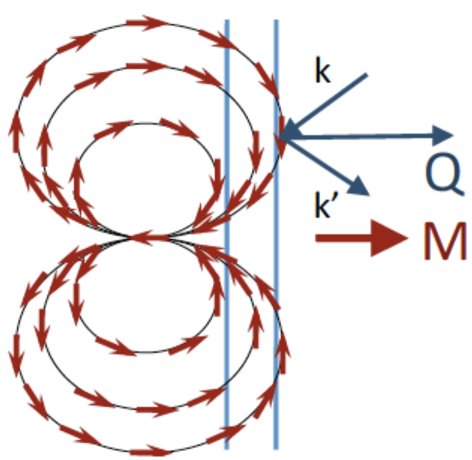
dipole-dipole interaction with magnetic fields of unpaired electrons

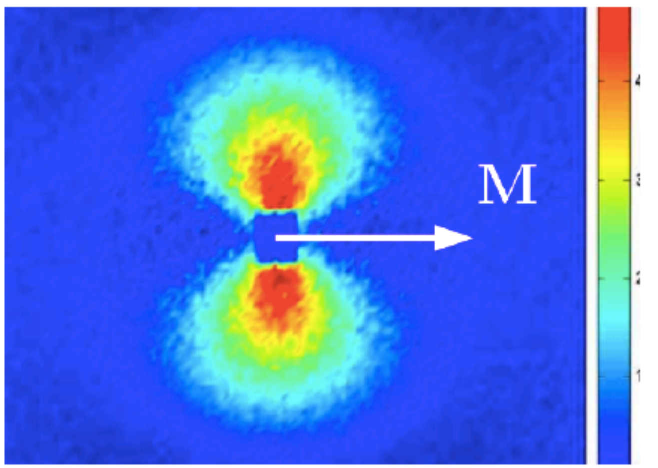
$$V_m = -\mu_{(n)} \cdot (\mathbf{B}_S + \mathbf{B}_L)$$

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

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







we can only see the moments perpendicular to Q !

magnetic scattering

Neutron spins

dipole-dipole interaction with magnetic fields of unpaired electrons

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

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

Magnetic scattering amplitudes (Bacon 1975)

$$p = -0.27 \times 10^{-12}\text{cm} \; \langle\mu\rangle f(Q)$$

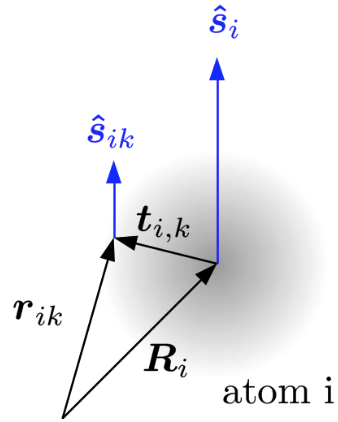
neutron electron coupling constant

magnetic moment in units μ_B

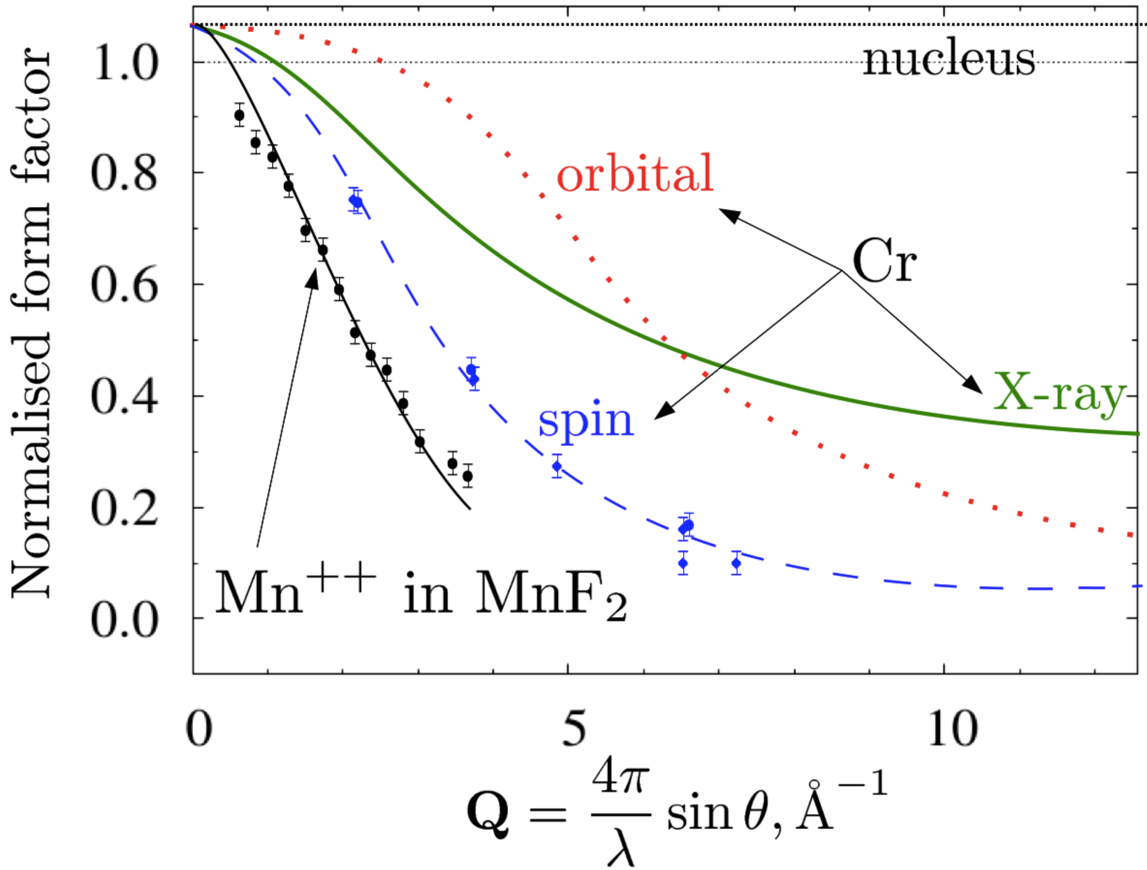
magnetic formfactor – Fourier transform of the spin density

(analog to x-ray form factor related to electron density)

Example: Fe, low T, saturated
$\mu = 2.2$
$f(Q)=0.59$ @ ~3.1Å^{-1}
$p = 0.35 \times 10^{-12}$cm



Spin density form-factors of Cr and Mn



Neutron properties are suitable to study structure and dynamics of atoms and magnetic moments

Why polarized neutron scattering?

PHYSICAL REVIEW VOLUME 83, NUMBER 2 JULY 15, 1951

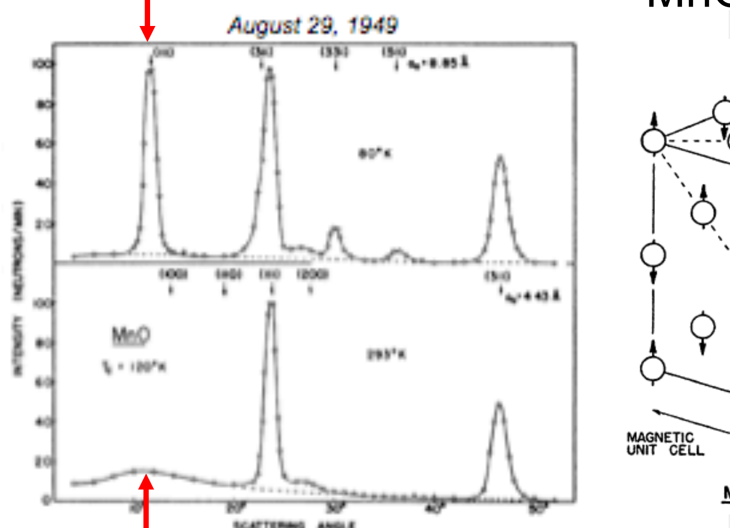
Neutron Diffraction by Paramagnetic and Antiferromagnetic Substances

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*

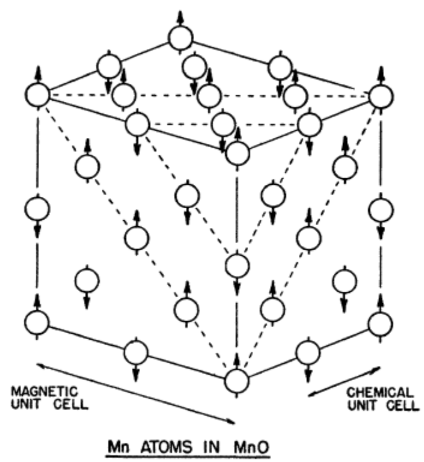
"for the development of neutron diffraction technique"

(111) alternating layers



paramagnetic
spin fluctuations

MnO



spins

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

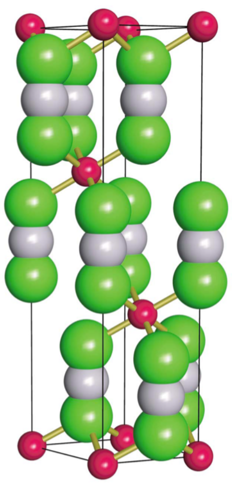
in (111) planes *Shaked et al. PRB 1988*

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

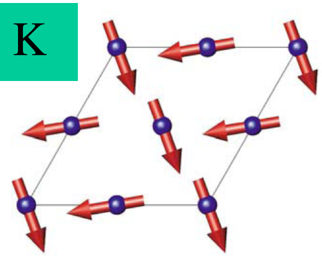
MnO => MnNCN
Replacing O^{-2} by NCN^{-2}

M. KROTT *et al* PHYSICAL REVIEW B **80, 024117 2009**

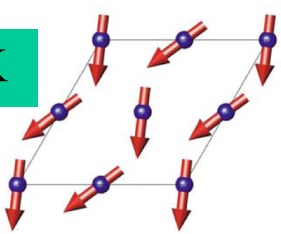
MnNCN



T = 4.5 K

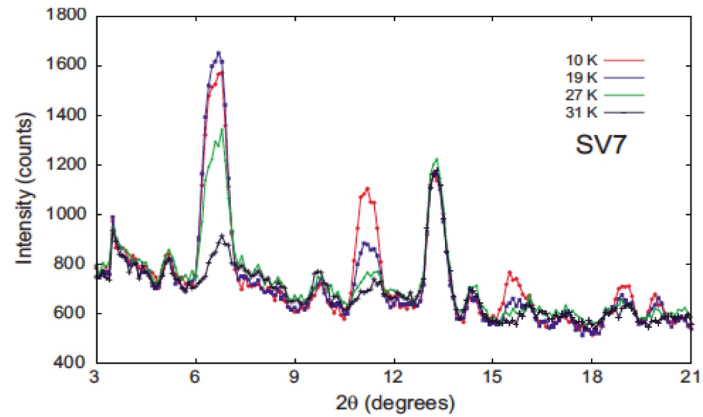


T = 20 K

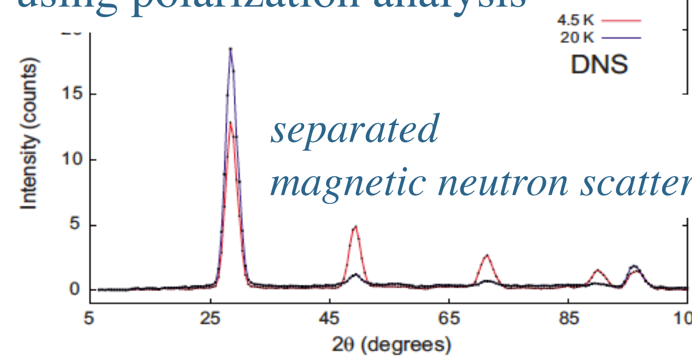


very clean results

conventional neutron diffraction



… using polarization analysis



separated
magnetic neutron scattering

Outline

- **Neutron spins in magnetic fields**
  - toolbox of experimental devices => instruments

- **Scattering and Polarization**
  - spin-dependent nuclear interaction
  - magnetic interaction

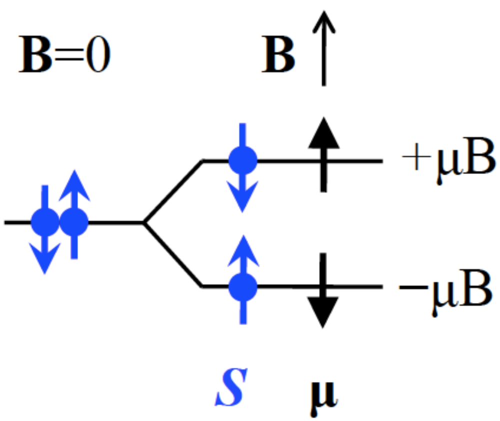
- **Blume-Maleyev Equations**
  - examples

- **outlook for ESS**

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

Neutron spins in magnetic fields

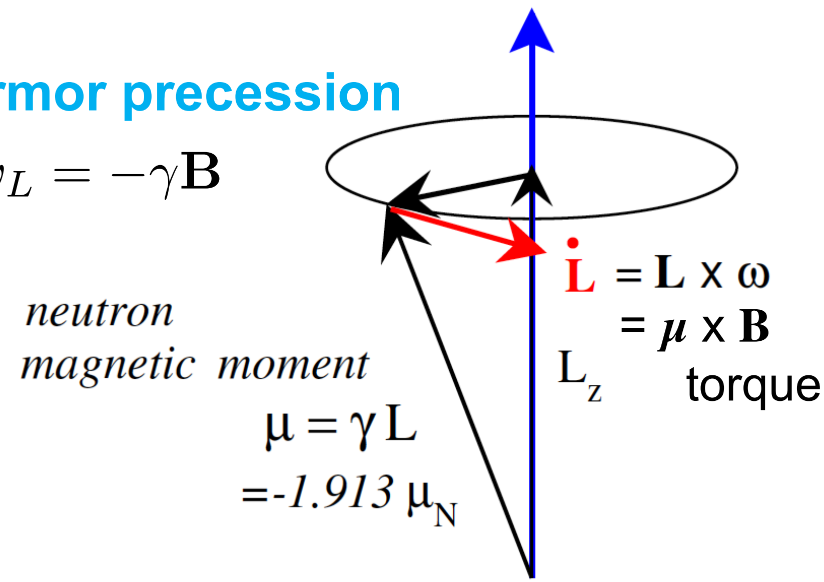
Zeeman splitting



$\hbar\omega_L$

Larmor precession

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



Bloch equation of motion

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

Neutron beam polarization

expectation value
average of spins:

$$\mathbf{P} = 2\langle \mathbf{S} \rangle$$

$-1 < |\mathbf{P}| < 1$

for a specific quantization axis

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

$0 < \mathrm{P} < 1$

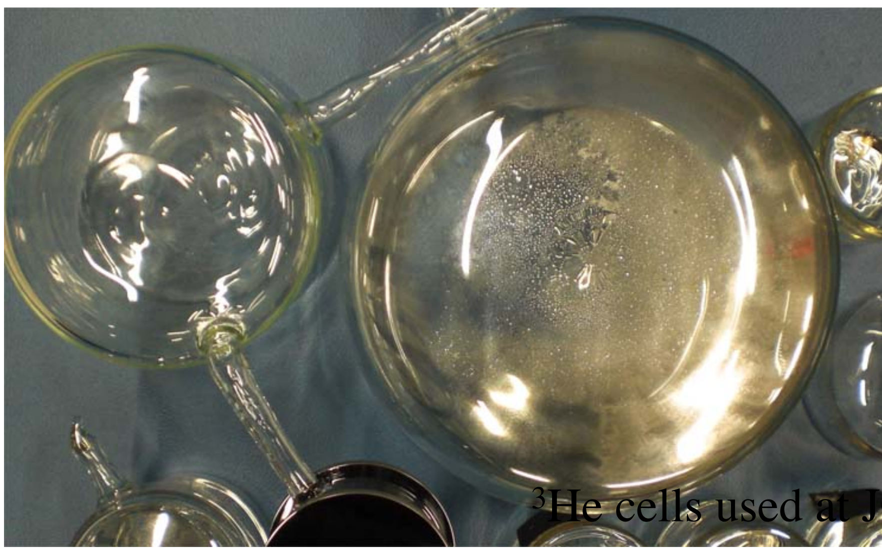
Neutron beam polarization

Absorption and transmission

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

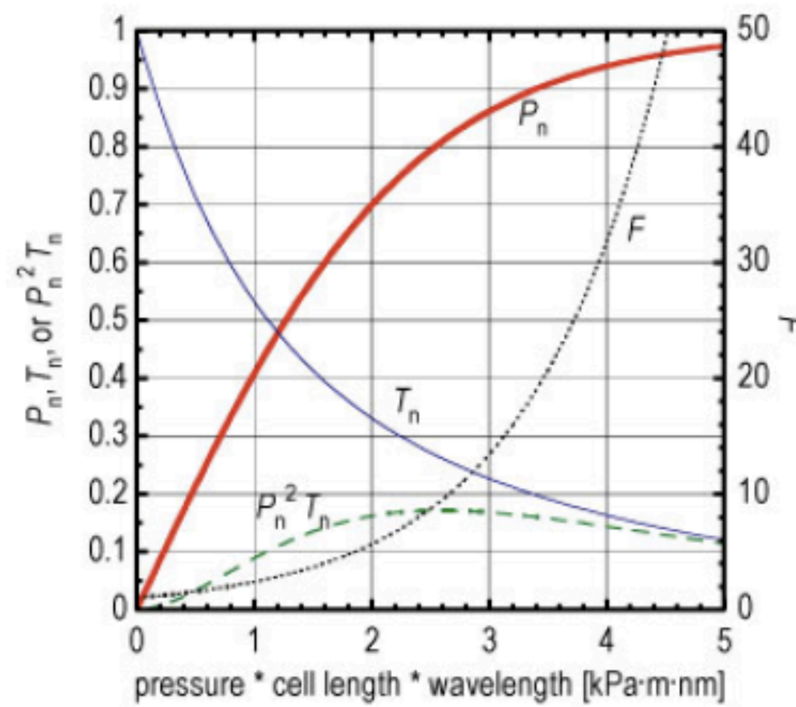
$$\sigma_{abs}(n_\uparrow, {}^3He_\downarrow) = 5333b$$

- 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



Neutron beam polarization

Scattering
constructive interference of nuclear and magnetic scattering

$$\sigma_{\pm} \propto (b \pm p)^2$$

-

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

Neutron beam polarization

Scattering

constructive interference of nuclear and magnetic scattering

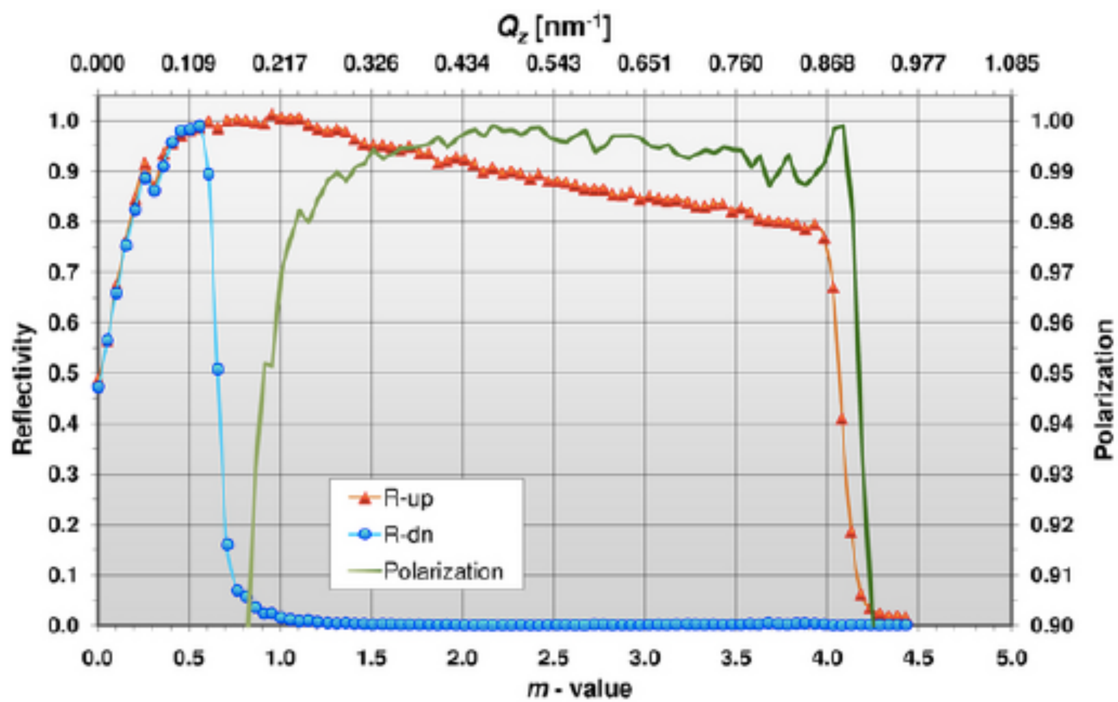
$$\sigma_{\pm} \propto (b \pm p)^2$$

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

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

Surface of
FeSi multilayers

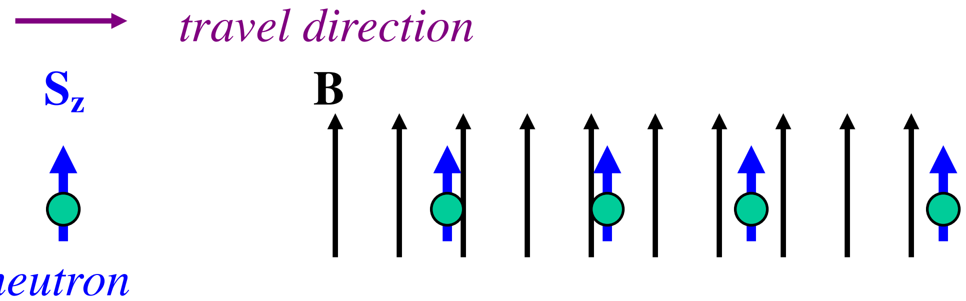
much better polarization at the interface of Si : FeSi multilayers



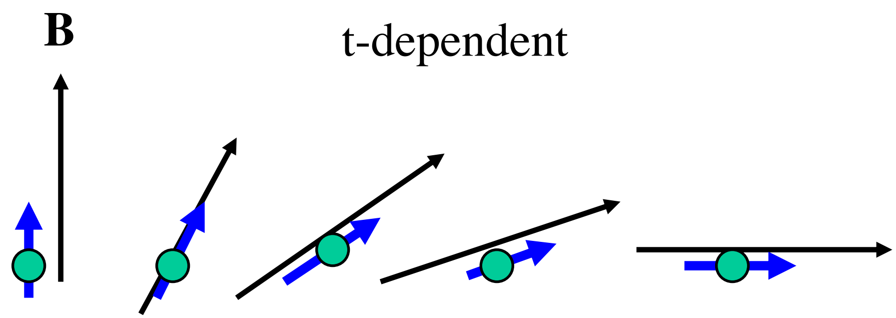
Source: Swiss Neutronics

Guide fields

constant B



Asymptotic behaviour

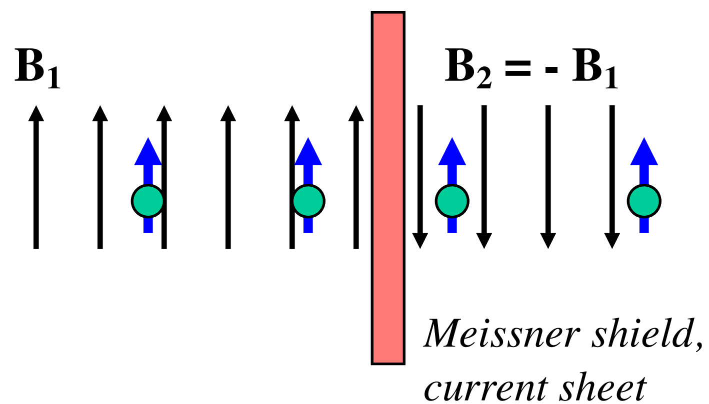


adiabatic field change

slowly varying “strong” B(t)
fast precession around B

sudden change

no change of S_z but change wrt B

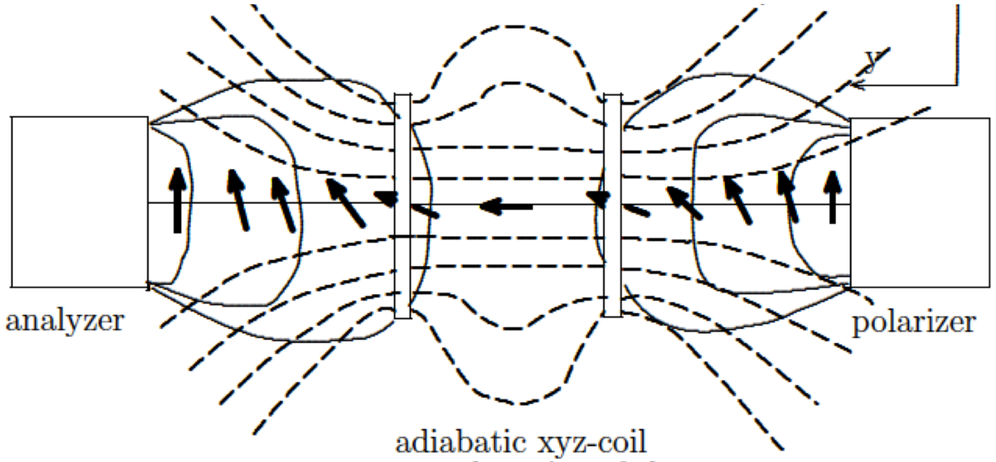


General behaviour

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

Guide fields - adiabatic field change

nutators, xyz-coils



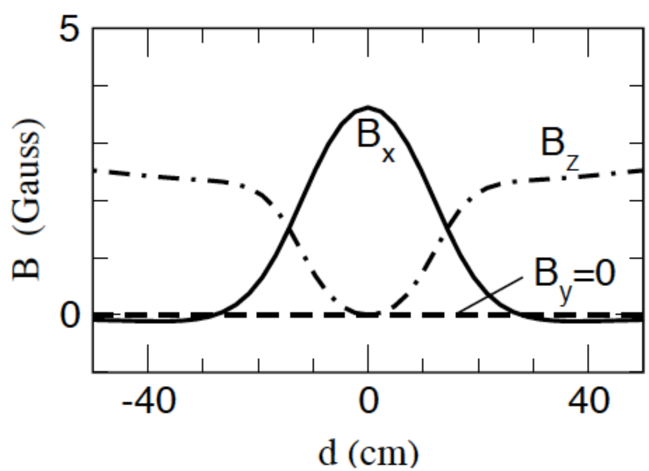


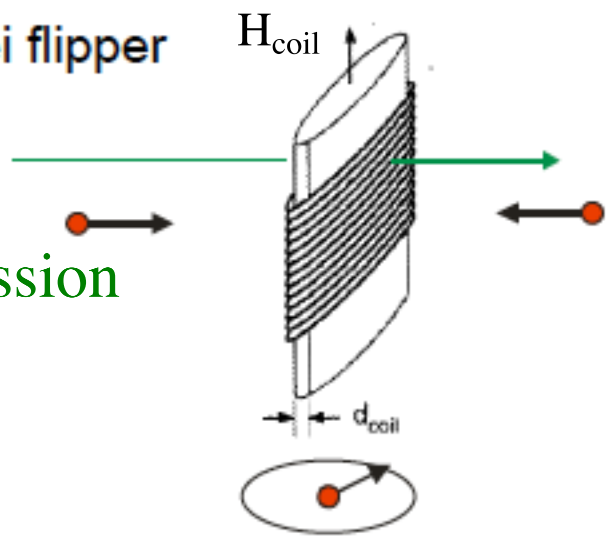
Fig. 6: *(left) Magnetic field setting in a xyz-coil system for an adiabatic nutation of the polarization of cold neutrons in horizontal x-direction at the sample turning to a vertical (guide) field B_z at further distance from the sample. (right) A photo of the xyz-coil system in the DNS instrument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

Flipper

Objective: change neutron polarization with respect to the applied field

Adiabatic and non-adiabatic changes of P || B

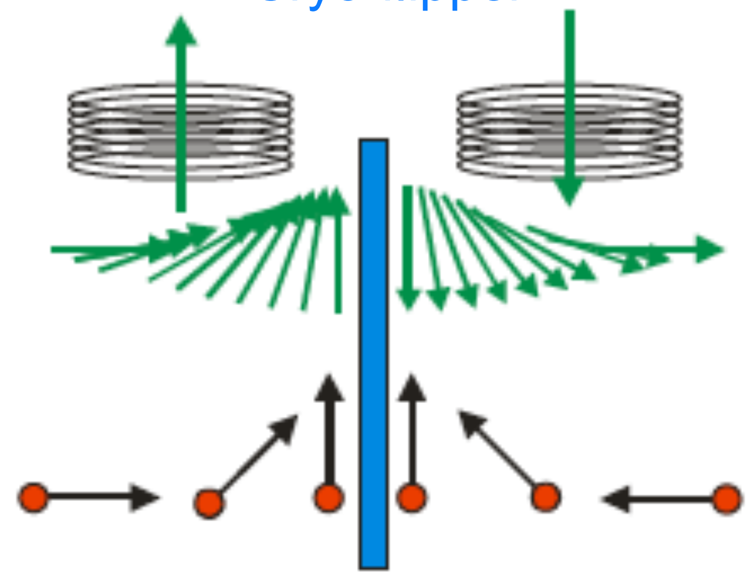
- Mezei flipper



precession

precession coil

Cryo-flipper



Meissner shield *superconducter*

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

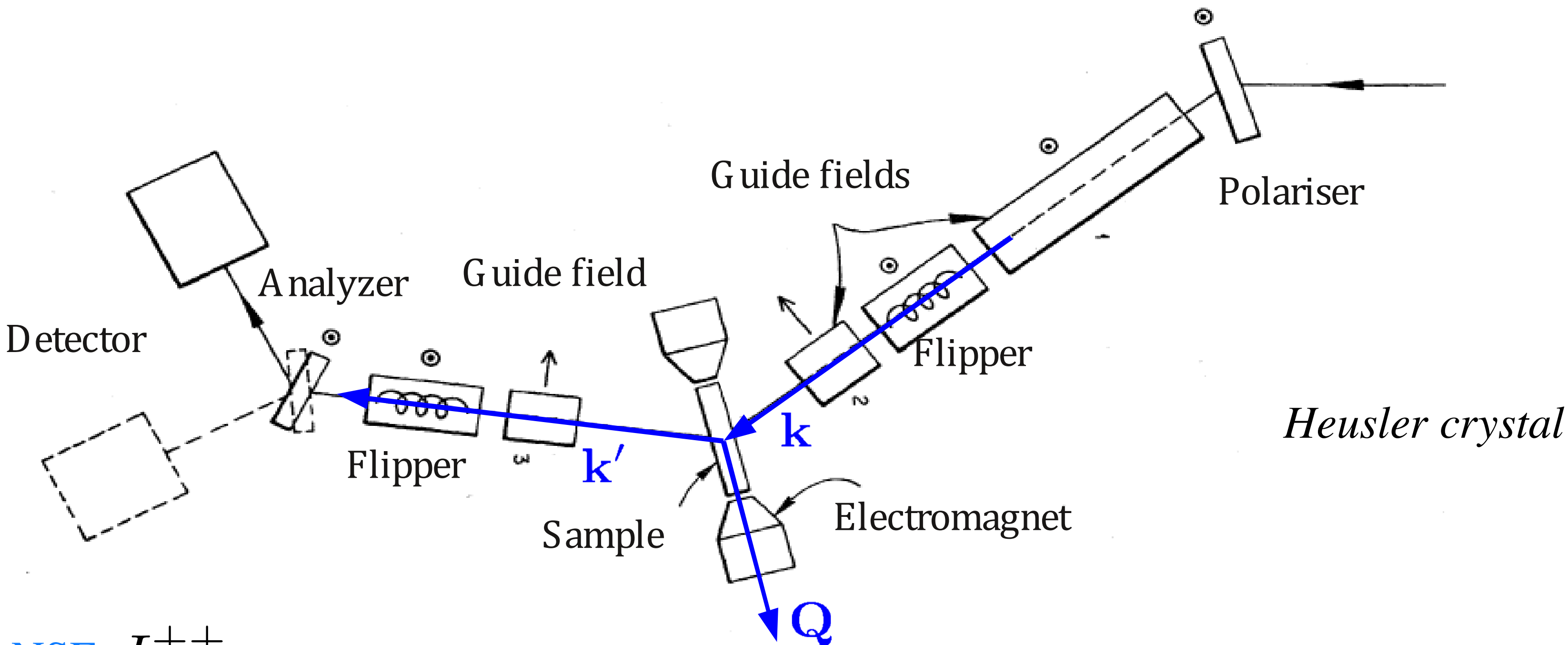
with $\gamma/2\pi = -2916\,\mathrm{Hz/Oe}$ and $mv = h/\lambda$

$$B = \frac{\pi}{d}(3956\,\mathrm{m/s}\cdot\text{Å}/\lambda)/(2916\cdot 2\pi\mathrm{Hz/Oe}) = \frac{67.83}{d\lambda}\,\mathrm{cm}\,\text{Å}\,\mathrm{Oe}$$

PHYSICAL REVIEW VOLUME 181, NUMBER 2 10 MAY 1969

Polarization Analysis of Thermal-Neutron Scattering*

R. M. Moon, T. Riste,† and W. C. Koehler
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

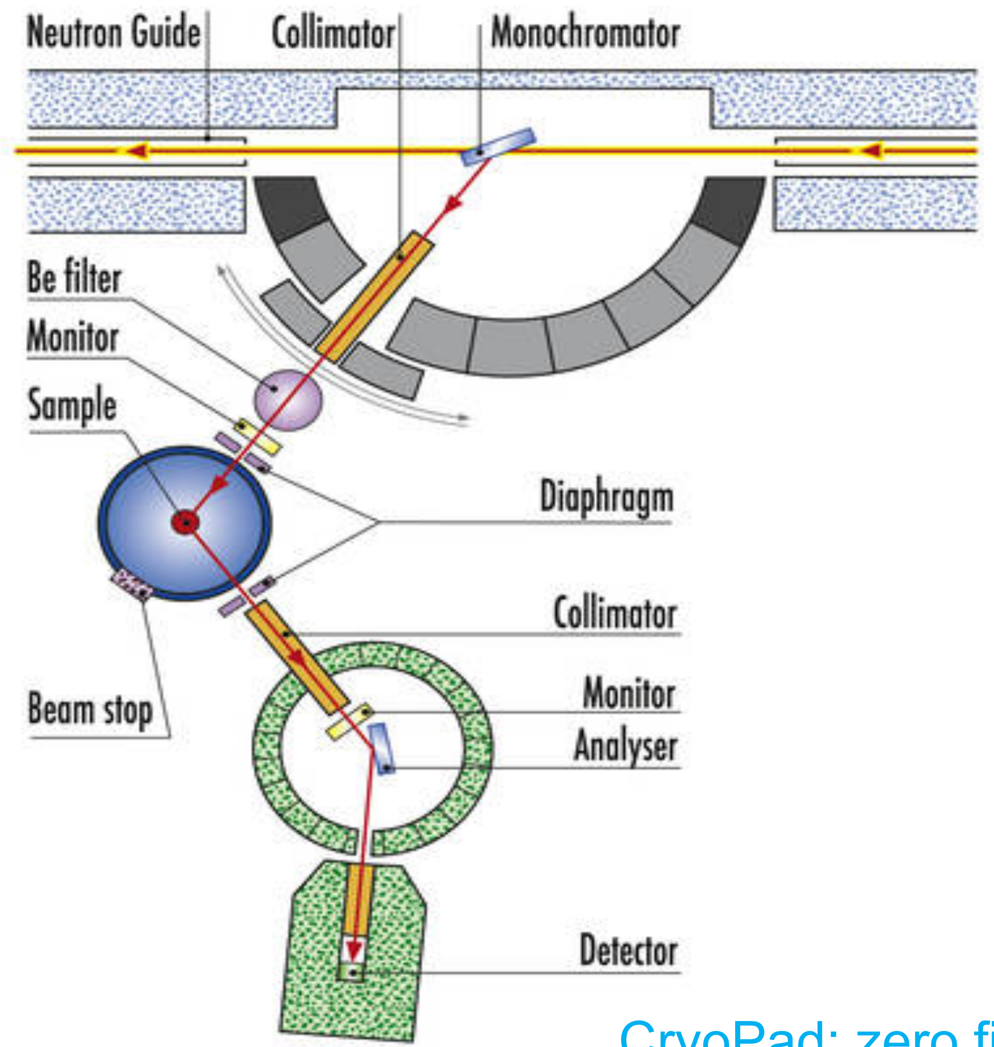


NSF I^{++}

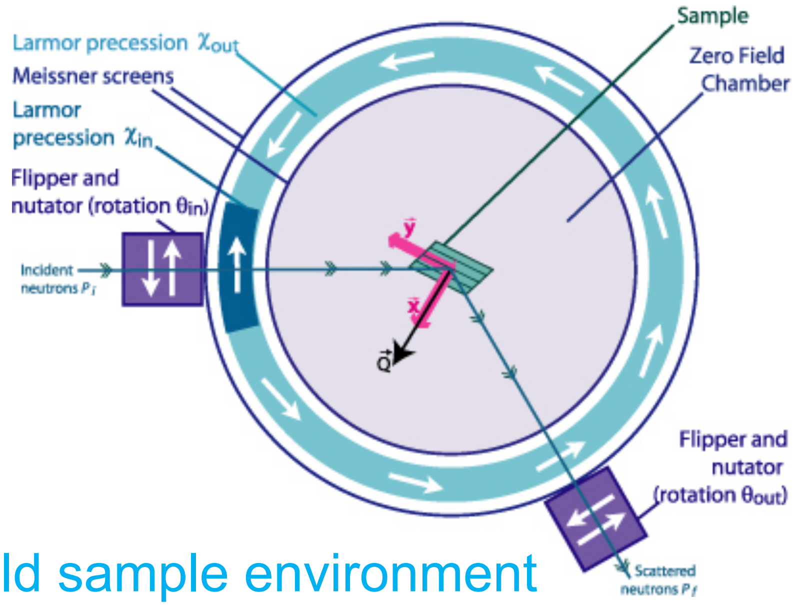
SF I^{+-}

Triple axis instrument
with *longitudinal* polarization analysis P || H

Triple axis instrument with *spherical* polarization analysis



IN12 @ ILL



CryoPad: zero field sample environment

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  - toolbox of 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

including initial and final spin states

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

1st Born approximation

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

Point like nucleus

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

Conservation of momentum and plane wave scattering

$$= b(\mathbf{Q})$$

assuming the nuclei have no spin

Scattering amplitude – transition matrix element

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

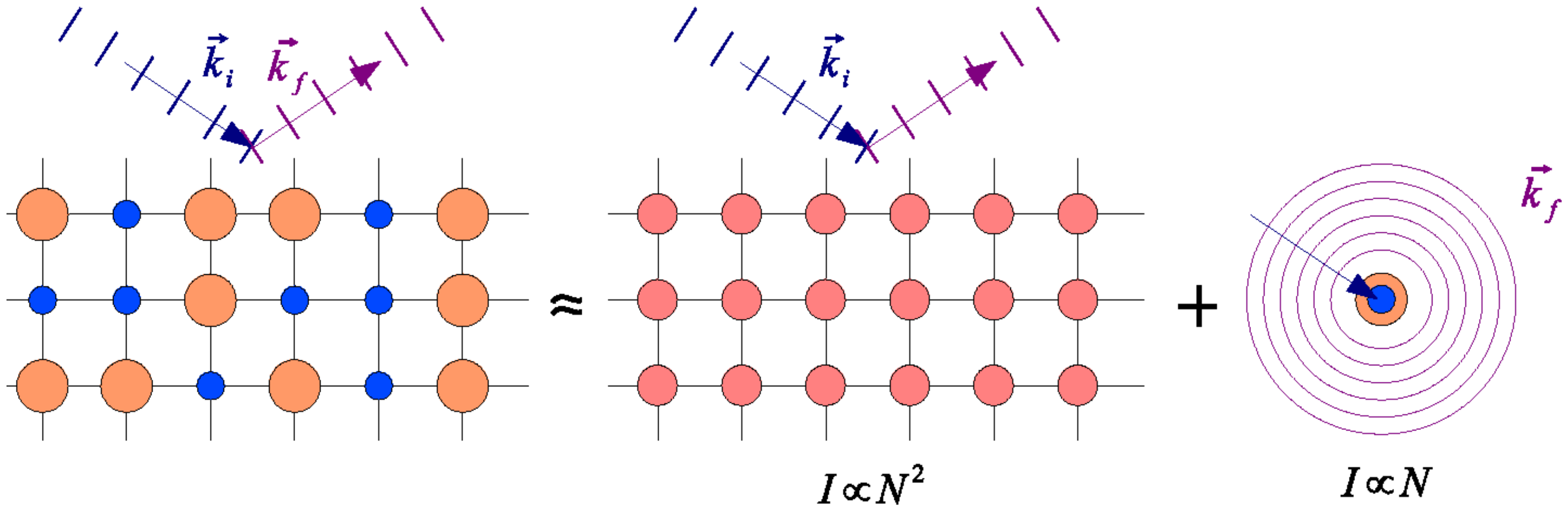
$$= b(\mathbf{Q}) \quad \text{no spin-flip}$$

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

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

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 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 $|+\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ $|-\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$

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

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

2/3 spinflip

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

1/3 non-spinflip

Spin dependent nuclear scattering amplitude

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

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

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

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

No spin flip in absence of a nuclear spin

$I \neq 0$ $$A(\mathbf{Q})^{\mathrm{NSF}} = A + BI_z \quad \text{for the } ++ \text{ and } -- \text{ case}$$

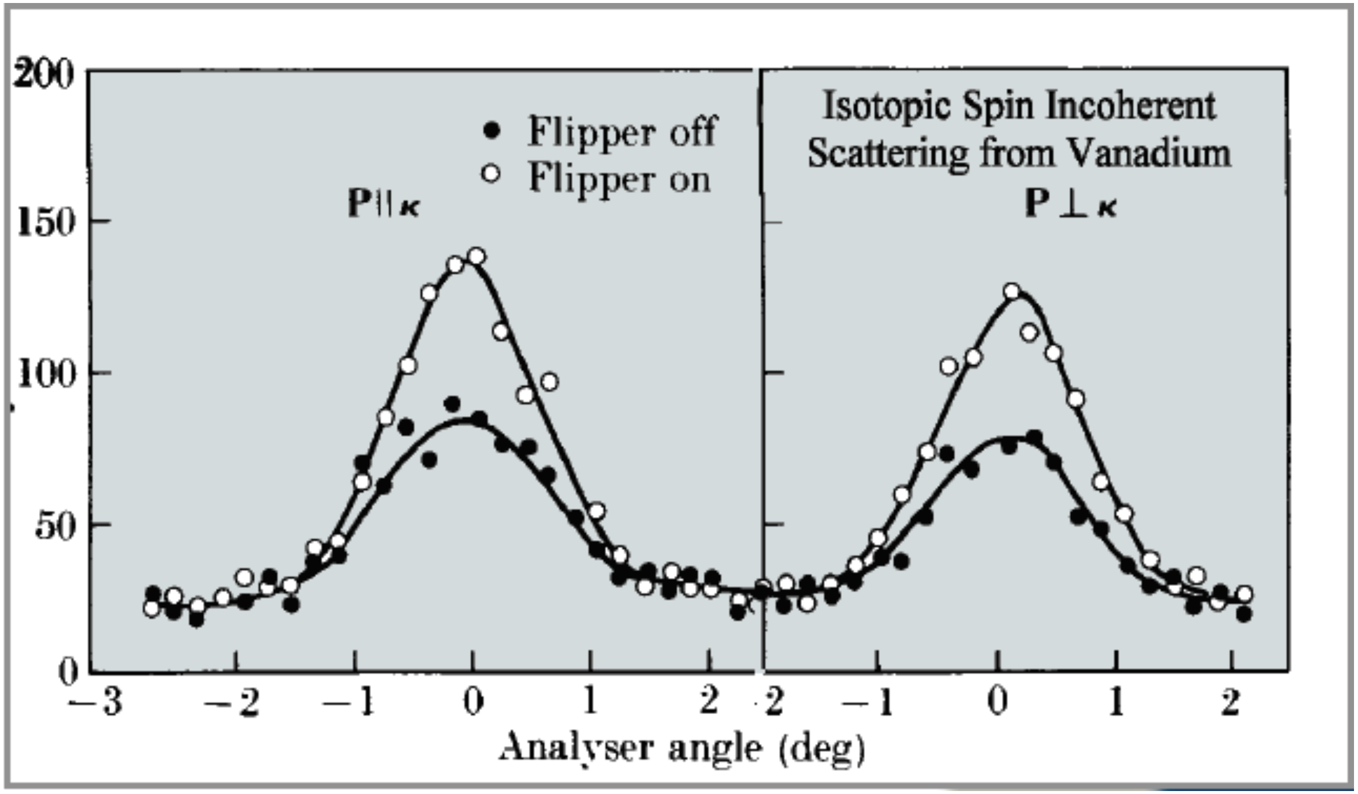
$$A(\mathbf{Q})^{\mathrm{SF}} = B(I_x + \mathrm{i}I_y) \quad \text{for the } +- \text{ and } -+ \text{ case}$$

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

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

Separation of spin-incoherent and coherent nuclear scattering

Applications to hydrogeneous materials, soft matter, etc.

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

small

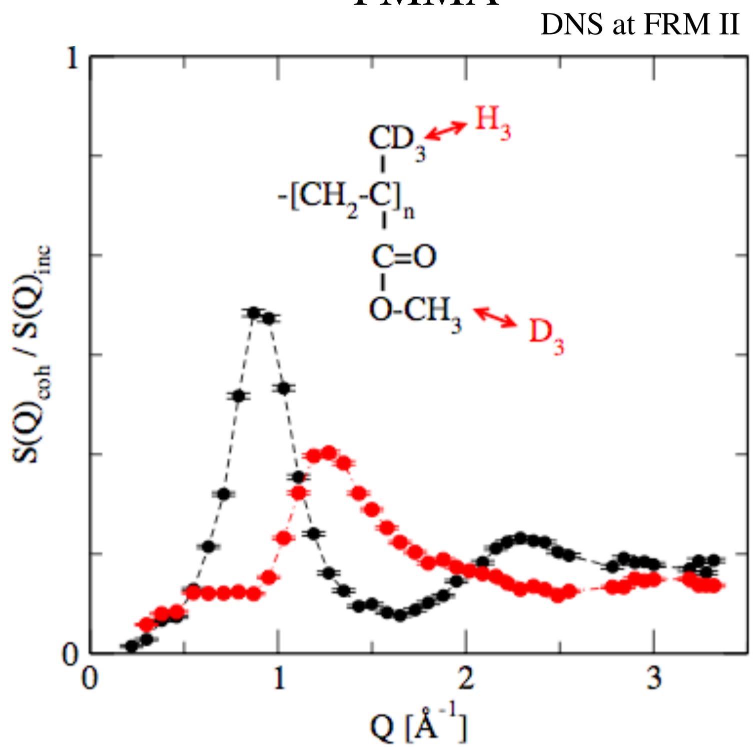
$$\frac{d\sigma}{d\Omega}_{spin-inc} = \frac{3}{2}\frac{d\sigma}{d\Omega}^{SF}$$

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

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

* Separating huge incoherent background of H
* Intrinsic calibration

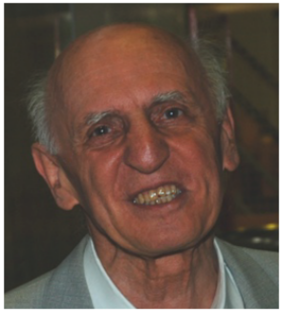
PMMA



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

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

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

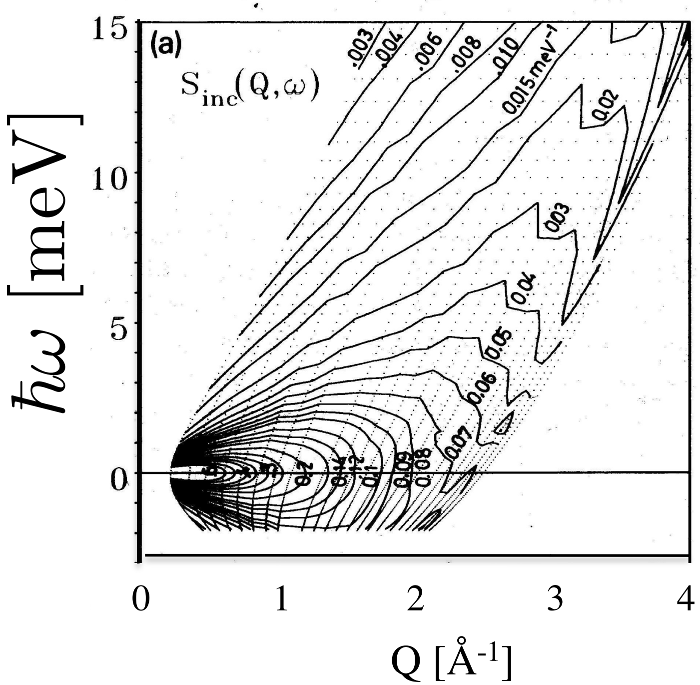
a good motivation to think about how to separate scattering

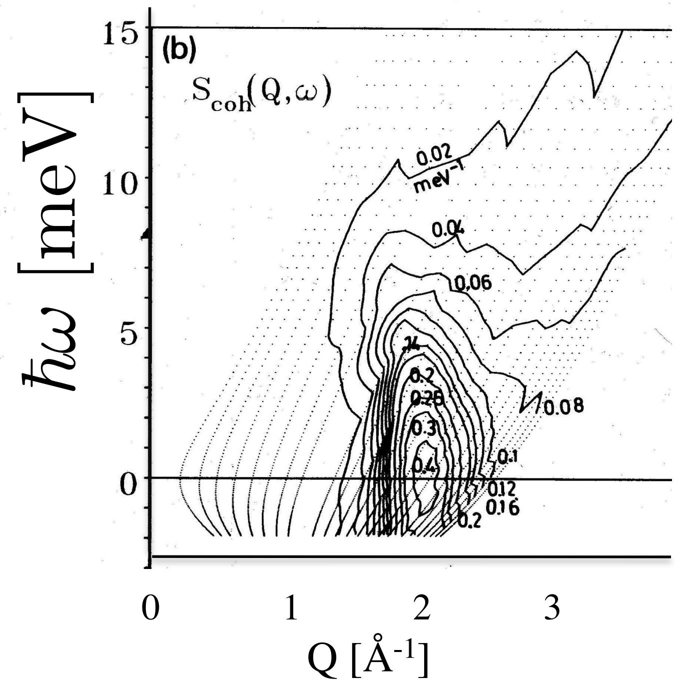
spin-incoherent

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

coherent

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





FT (self correlation)

single particle diffusion

FT (pair correlation)

collective behavior

precursors of Bragg scattering

Magnetic scattering cross section

initial and final spin states

$$A(\mathbf{Q}) = \langle S_z'| -\frac{\gamma_n r_0}{2\mu_B}\hat{\boldsymbol{\sigma}}\cdot\mathbf{M}_\perp(\mathbf{Q})|S_z\rangle$$
$$= -\frac{\gamma_n r_0}{2\mu_B}\sum_\alpha \langle S_z'|\underline{\hat{\boldsymbol{\sigma}}}_\alpha|S_z\rangle \mathbf{M}_{\perp\alpha}(\mathbf{Q}) \qquad \alpha = x, y, \text{ or } z$$

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

Choosing *z* as quantization axis

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

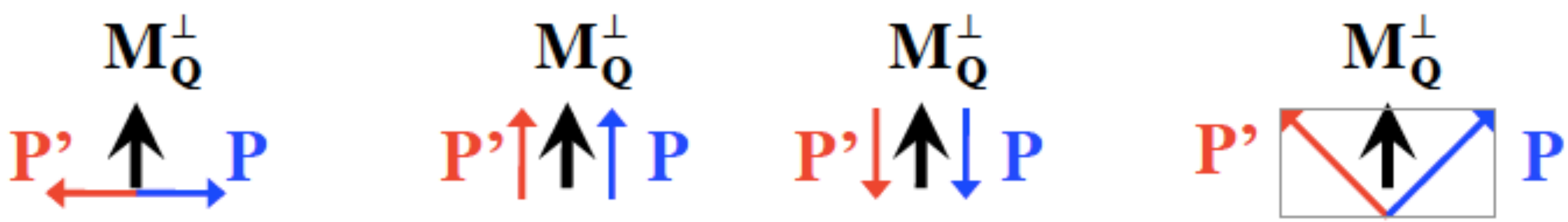
coordinate system say x || Q

we have seen this before:

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

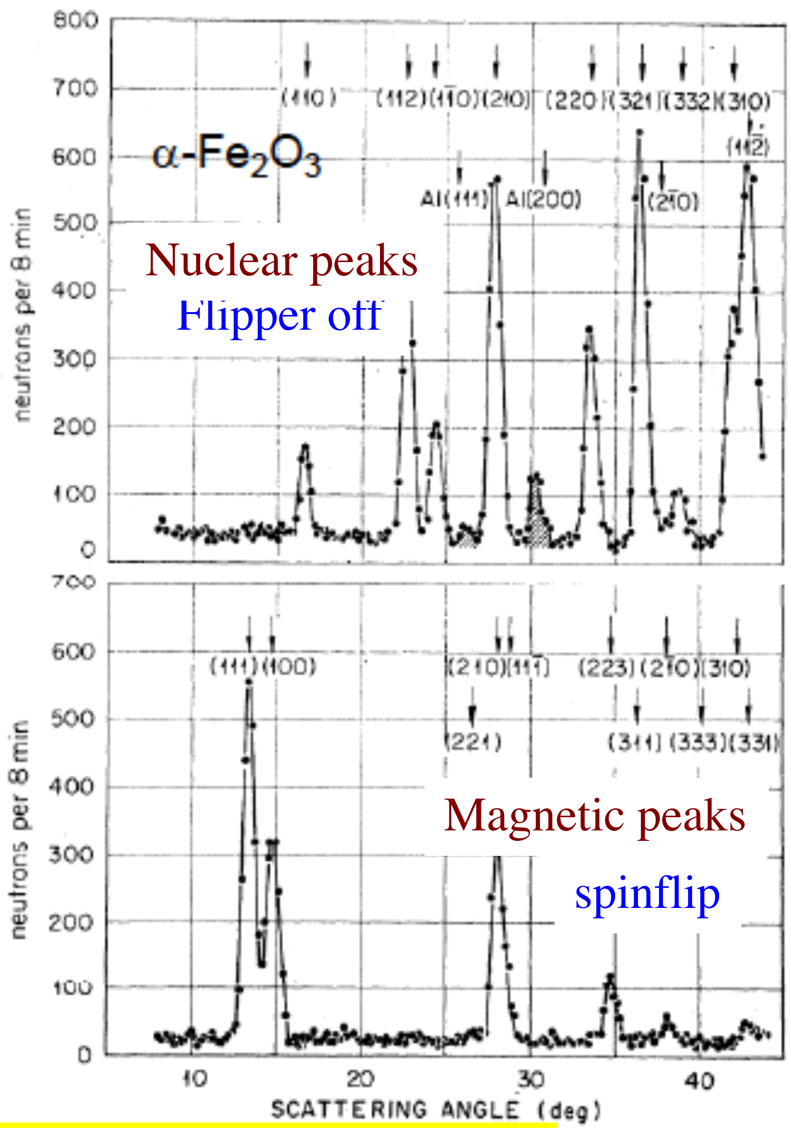
direction of **P, M, Q** matters!

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



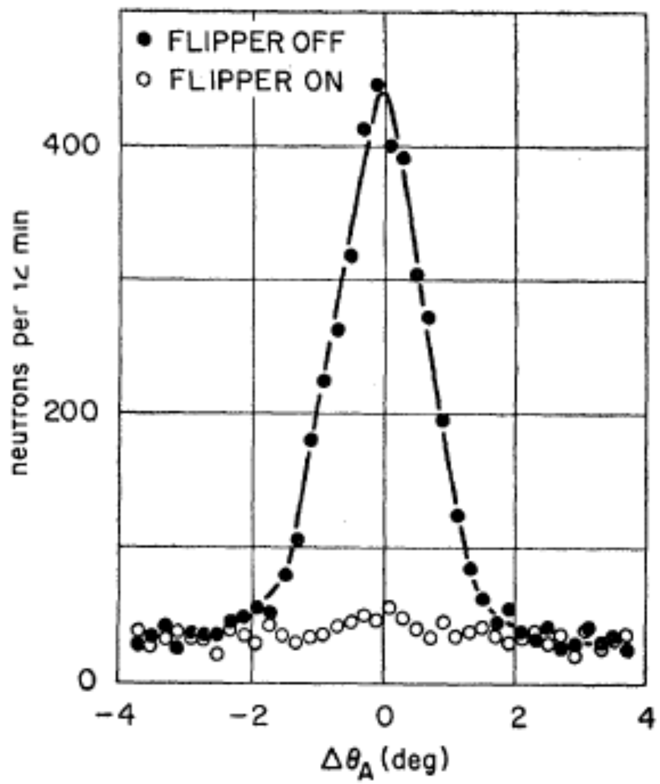
Separating nuclear and magnetic scattering

- Antiferromagnet: Bragg scattering



Moon, Riste and Koehler (1969)

Ni – Ferromagnet



Isotopic incoherent scattering

P || Q

no magnetic scattering
for **M || Q**

Typically, FM need saturating fields

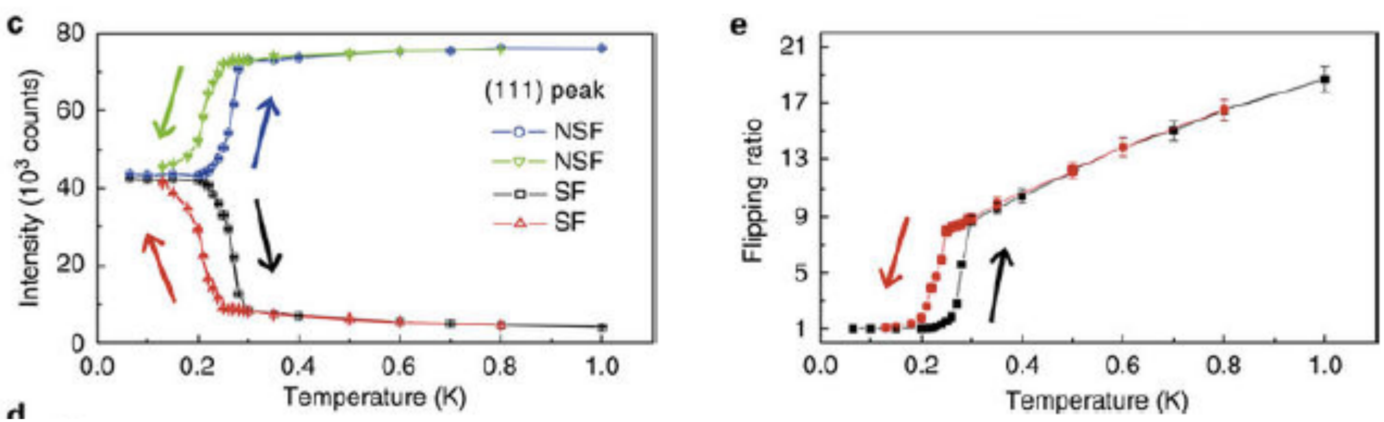
Note: thermocouple alloy $Ni_{89}Cr_{11}$ is non-magnetic

Depolarisation of the neutron spins are observed …

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

LJ Chang et al, Nature Communications 2012

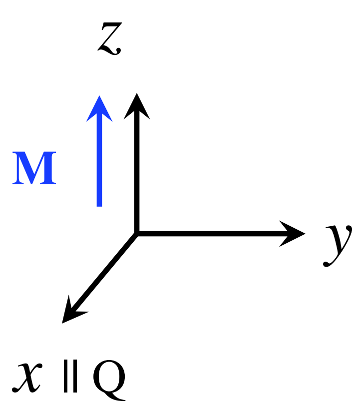
depolarization of the neutron spins are observed with thermal hysteresis, indicating a first-order ferromagnetic transition. Our results are explained on the basis of a quantum spin-ice model, whose high-temperature phase is effectively described as a magnetic Coulomb liquid, whereas the ground state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



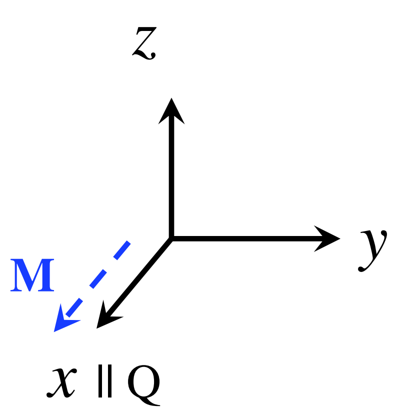
Ferromagnets: Separation by the || - ⊥ method, case of strong field H

FM

$\mathbf{P} \parallel \mathbf{H} \perp \mathbf{Q}$



nsf	$\lvert \mathrm{M}_z \rvert^2 + \lvert \mathrm{N} \rvert^2 \pm 2\mathrm{M}_z N$	$+\frac{1}{3}\sigma_{si}$
sf		$+\frac{2}{3}\sigma_{si}$



$\mathbf{H} \parallel \mathbf{Q}$

nsf	$\lvert \mathrm{N} \rvert^2$	$+\frac{1}{3}\sigma_{si}$
sf		$+\frac{2}{3}\sigma_{si}$

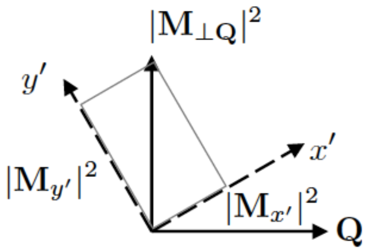
Note: For ferromagnets, the magnetic scattering can be separated by the difference due to field variation even without polarization analysis.
The interference term M_zN requires P, however, no polarization analysis

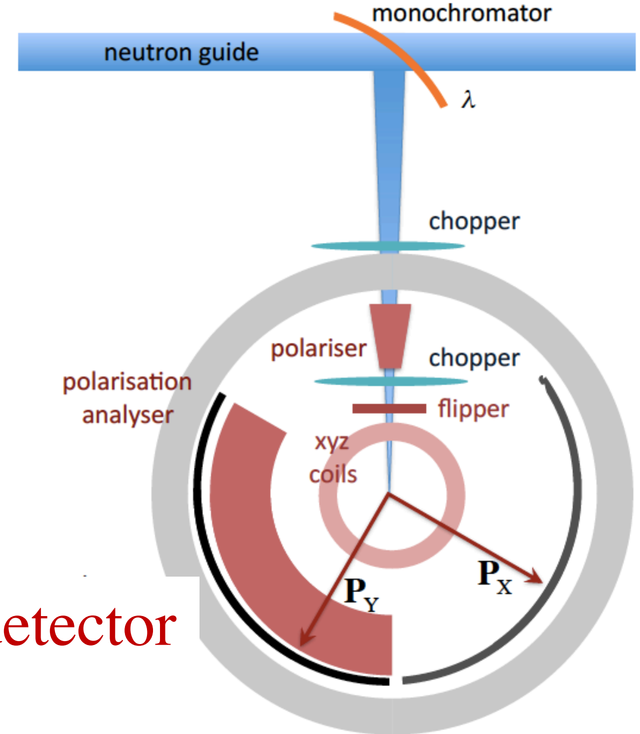
Powder diffraction with "XYZ" polarization analysis

⇒ **Separation of magnetic and nuclear scattering and spin-incoherent background**

O. Schärpf and H. Capellmann,
Phys. Status Solidi A 135, 359 (1993).

DNS at MLZ, Munich
(~ D7 at ILL, Grenoble)





multi-detector

$$\frac{d\sigma}{d\Omega}_{magn} = 2\left(\frac{d\sigma}{d\Omega}_{x}^{SF} + \frac{d\sigma}{d\Omega}_{y}^{SF} - 2\frac{d\sigma}{d\Omega}_{z}^{SF}\right) = -2\left(\frac{d\sigma}{d\Omega}_{x}^{NSF} + \frac{d\sigma}{d\Omega}_{y}^{NSF} - 2\frac{d\sigma}{d\Omega}_{z}^{NSF}\right)$$

Powder diffraction with "XYZ" polarization analysis

DNS at MLZ, Munich
(~ D7 at ILL, Grenoble)

⇒ **Separation of magnetic and nuclear scattering and spin-incoherent background**

Non-stoichiometry and the magnetic structure of Sr_2CrO_3FeAs

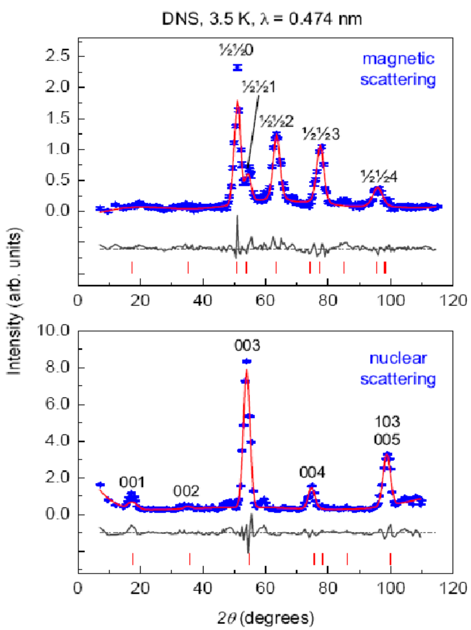
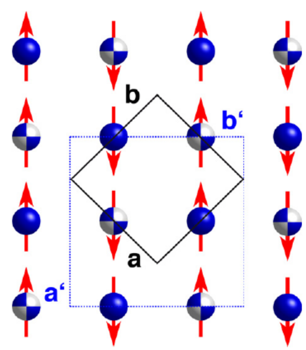


FIG. 3: Magnetic and nuclear reflections of Sr_2CrO_3FeAs (blue) and Rietveld fit (red) at 3.5 K measured at DNS [3].



M. Tegel et al, Europhysics Lett. 2010

new dedicated powder diffractometers
WOMBAT, ECHNIDA at ANSTO, Sidney

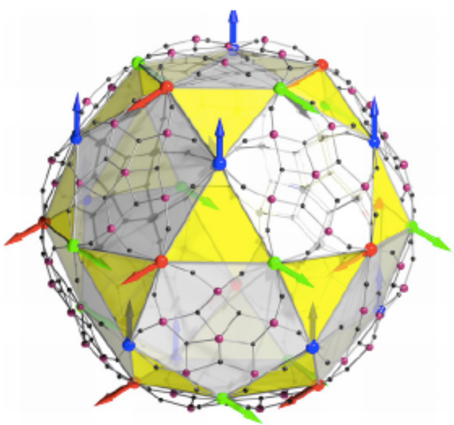
Diffuse magnetic scattering
with “XYZ” polarization analysis

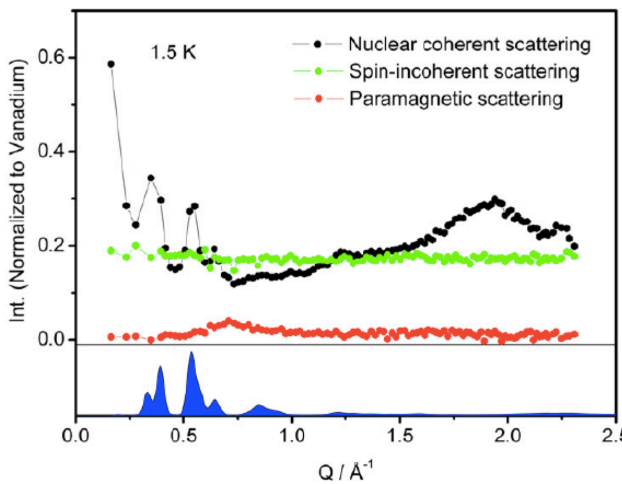
DNS at MLZ, Munich
(~ D7 at ILL, Grenoble)

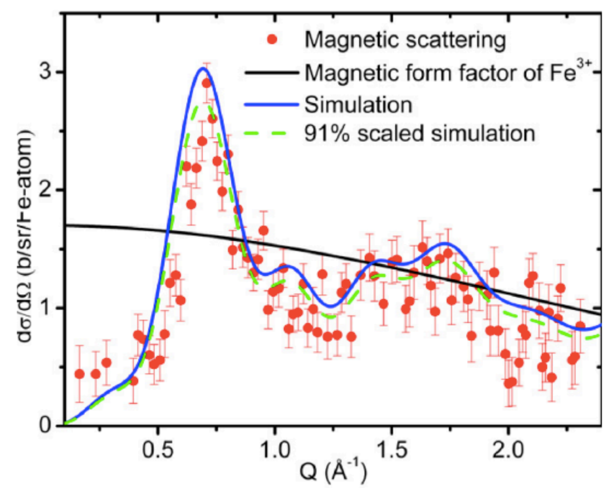
⇒ **Separation of**
magnetic and nuclear scattering
and spin-incoherent background

Magnetic ground state of the molecular magnet [$Mo_{72}Fe_{30}$]

Zhendong Fu et al, New Journal of Physics **12** (2010)

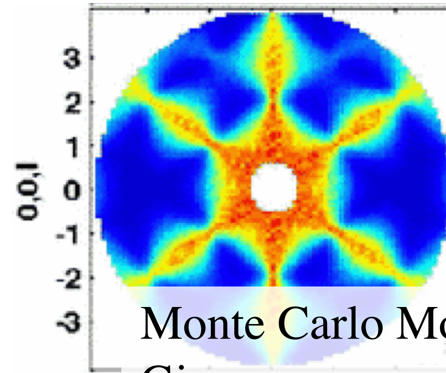






Single crystal diffuse magnetic scattering with “XYZ” polarization analysis

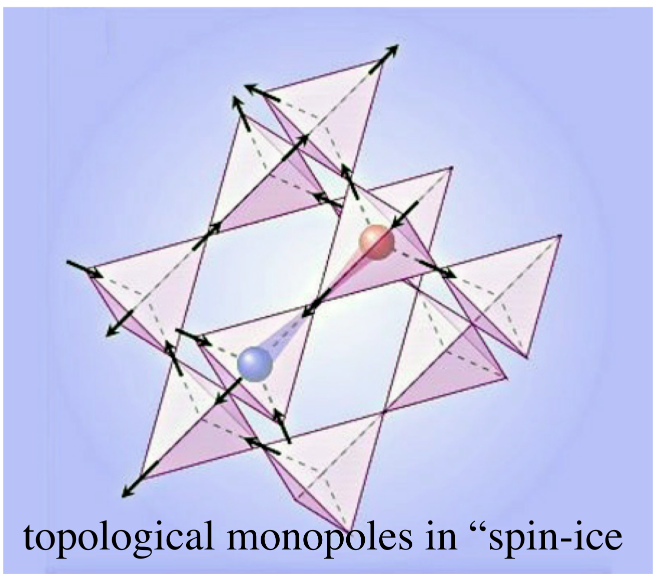
⇒ **Separation of magnetic and nuclear scattering and spin-incoherent background**



Monte Carlo Model Gingras

Magnetic Coulomb Phase in the Spin Ice $Ho_2Ti_2O_7$

T. Fennell et al. Science 2009

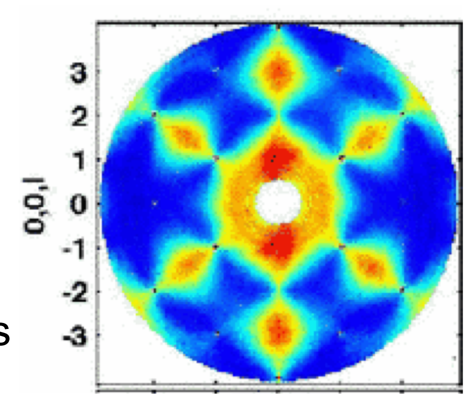


topological monopoles in “spin-ice

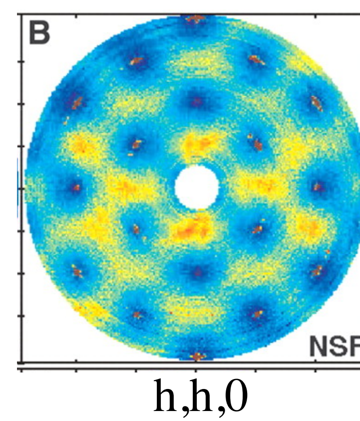
Poles apart. A pyramid with three ions pointing in (blue) acts as a north monopole; one with one ion pointing in (red) acts as a south monopole. By flipping other spins, the monopoles can be moved apart.

P || z

Spin-flip Scattering shows pinch-points



NSF



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**

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

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

magnetic *magnetic-nuclear interference* *chirality*

and **final polarized intensity**

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

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

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

Final polarisation

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

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**

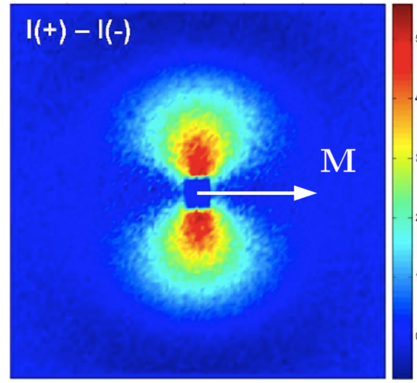
$$\mathbf{P}'\sigma_{\mathbf{Q}} = \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{coh}} + \mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{isotop\text{-}inc}} - \tfrac{1}{3}\mathbf{P}\sigma^{\mathbf{N}}_{\mathbf{Q},\mathrm{spin\text{-}inc}}$$

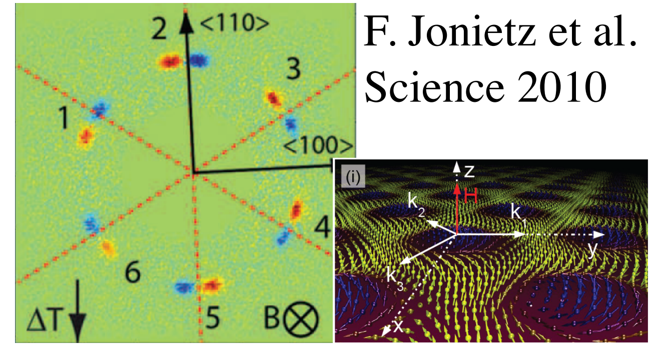
$$+\mathbf{M}^{\perp}_{\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{-\mathbf{Q}}) + \mathbf{M}^{\perp}_{-\mathbf{Q}}(\mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}) - \mathbf{P}\mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}$$

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

Blume – Maleyev
general theory for polarized neutron scattering

… yields two expressions





F. Jonietz et al. Science 2010

Skyrmions

Polarization reversal

for **scattering intensity**

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

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

magnetic *magnetic-nuclear interference* *chirality*

and **final polarized intensity**

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

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

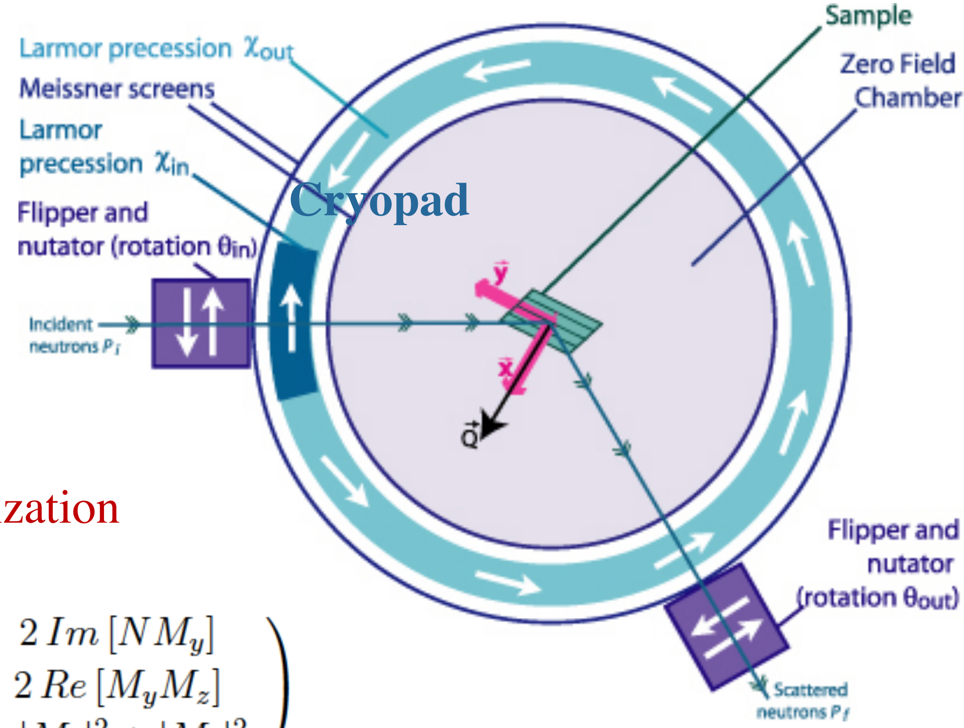
$$+\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} + \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}} + i(\mathbf{M}^{\perp}_{\mathbf{Q}}N_{-\mathbf{Q}} - \mathbf{M}^{\perp}_{-\mathbf{Q}}N_{\mathbf{Q}}) \times \mathbf{P} + i\mathbf{M}^{\perp}_{\mathbf{Q}} \times \mathbf{M}^{\perp}_{-\mathbf{Q}}$$

creates **P’**

Spherical neutron polarimetry

Jane Brown, Francis Tasset

Cryopad



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

P" created polarization

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

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

Half-polarized experiments – polarization reversal

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

Single crystal diffuse magnetic scattering
with "XYZ" polarization analysis + polarisation reversal

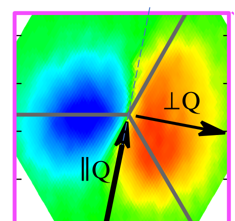
⇒ **Separation of**
all terms in the Blume – Maleyev equations

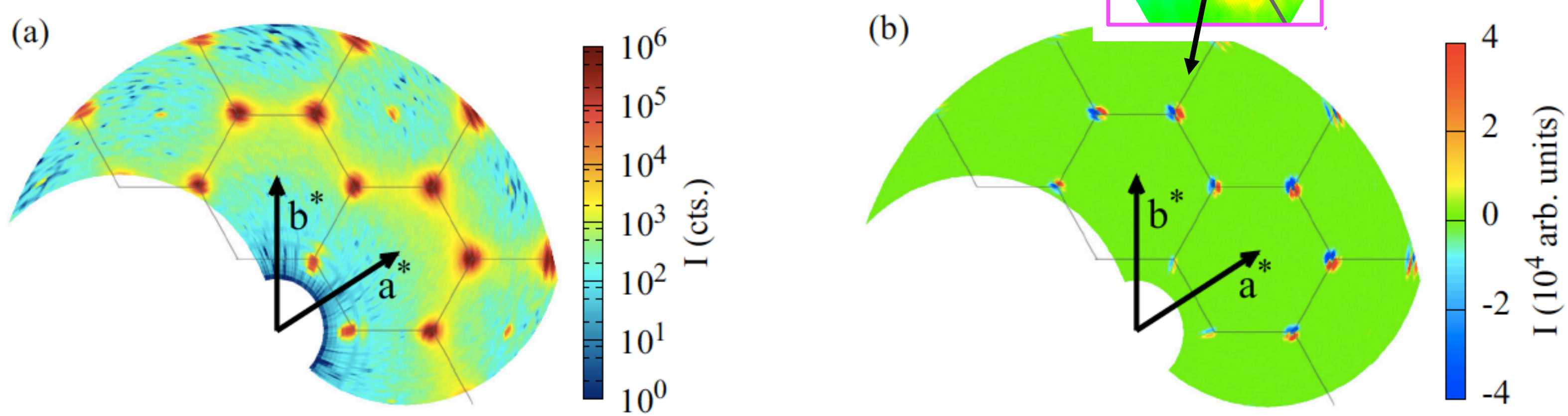
W. Schweika 2010 J. Phys.: Conf. Ser. **211** 012026

PHYSICAL REVIEW B **97**, 144402 (2018)

Neutron diffraction study and theoretical analysis of the antiferromagnetic order and the scattering in the layered kagome system $CaBaCo_2Fe_2O_7$

J. D. Reim,[1,2,*] E. Rosén,[2] O. Zaharko,[3] M. Mostovoy,[4] J. Robert,[5] M. Valldor,[6] and W. Schweika[2,7,†]

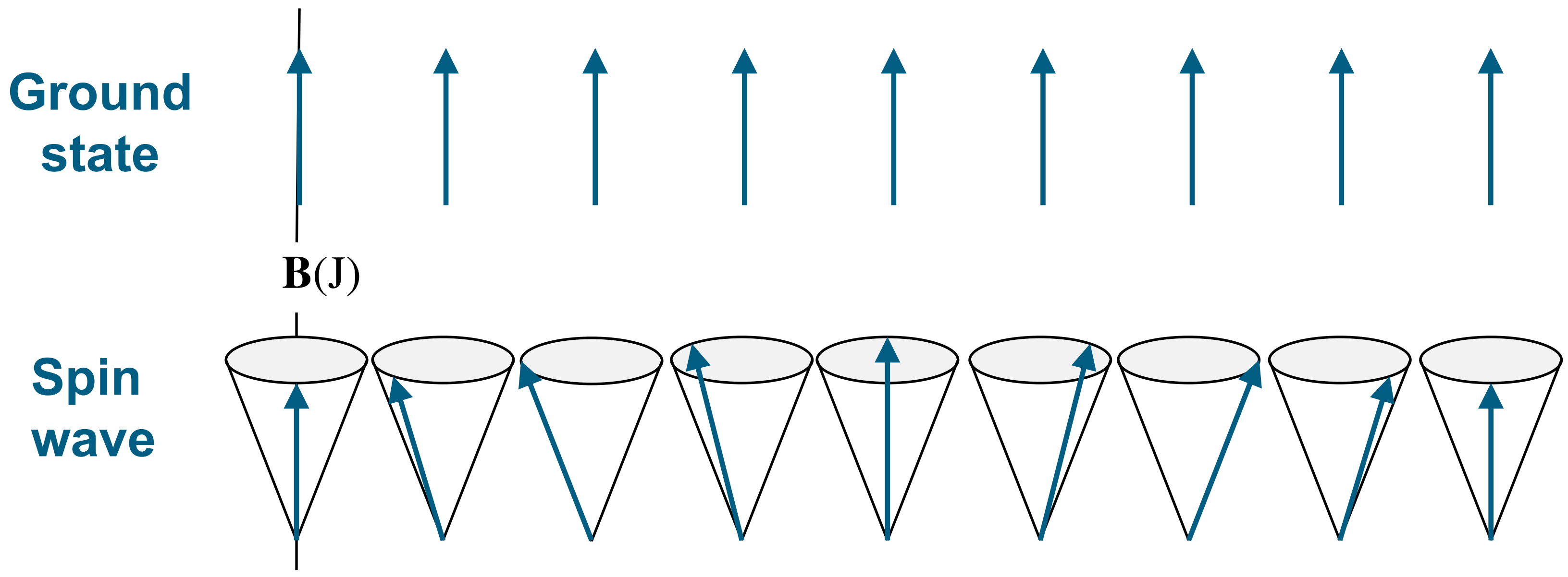




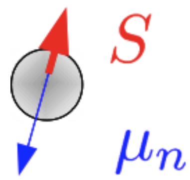
total magnetic scattering

Interference due to chirality

Spin wave excitations



Spin waves, aren‘t they chiral?
left or right handiness?
is the picture correct?
helix or cycloid?



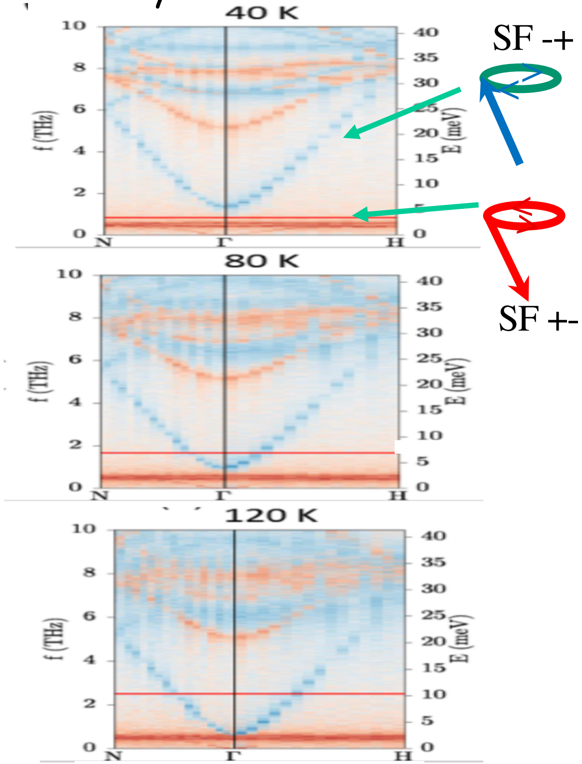
and what about neutrons
aren‘t they chiral, too?

The Spin Seebeck effect

is caused by thermally excited spin dynamics that are converted to a voltage by the inverse spin Hall effect at the interface to a heavy metal contact.

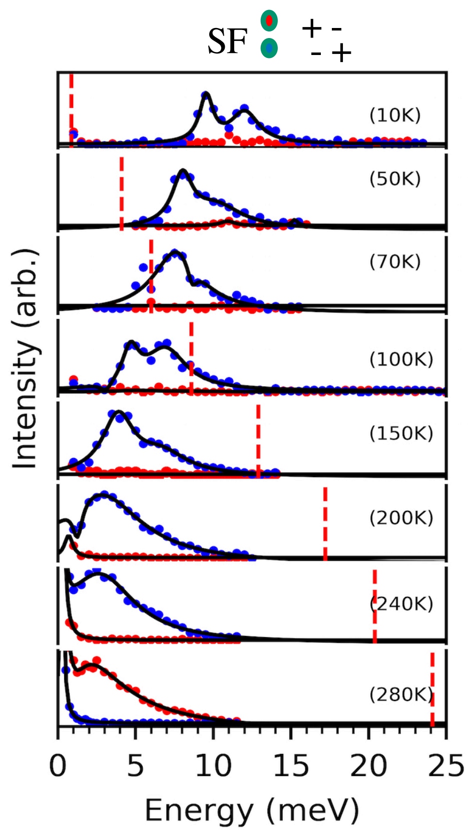
Neutron Scattering Results

Theory

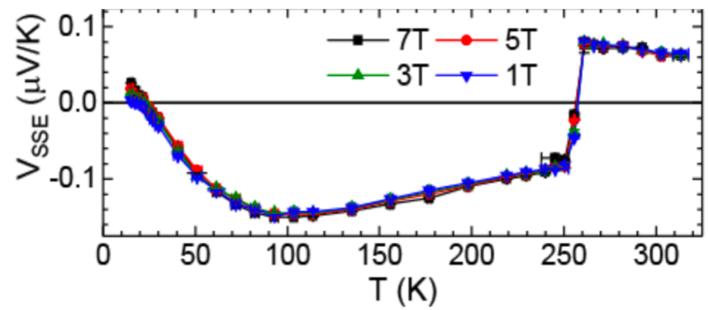


Landau-Lifshitz-Gilbert

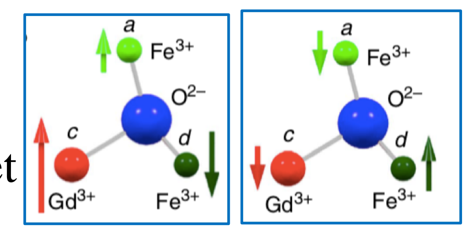
Nature Comms. 7 10452 (2016)

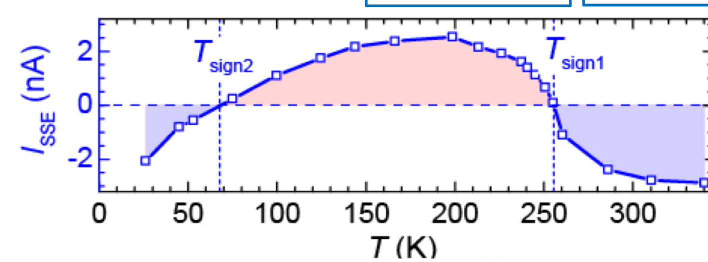


Tb-iron garnet



Gd-iron garnet





magnons are chiral, of course ...

courtesy of Dan Mannix, ESS

Magnetization and spin densities

wide angle Bragg diffraction for atomic resolution

I^+ (**P** *parallel* **H**) and I^- (**P** antiparallel **H**)

"flipping ratio" measurements sensitive to small **M**

$$R = \frac{I^+}{I^-} = \frac{N_{\mathbf{Q}}N_{-\mathbf{Q}} + (N_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}} + N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}}) + \mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}}{N_{\mathbf{Q}}N_{-\mathbf{Q}} - (N_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}} + N_{-\mathbf{Q}}\mathbf{M}^{\perp}_{\mathbf{Q}}) + \mathbf{M}^{\perp}_{\mathbf{Q}}\mathbf{M}^{\perp}_{-\mathbf{Q}}}.$$

example $M_z = 0.1N$

$$I = 1.01|N|^2, \quad \text{and} \quad \frac{I^+}{I^-} = \frac{(1+0.1)N^2}{(1-0.1)N^2} = \frac{1.21}{0.81} \approx 1.5$$

solve nuclear structure first

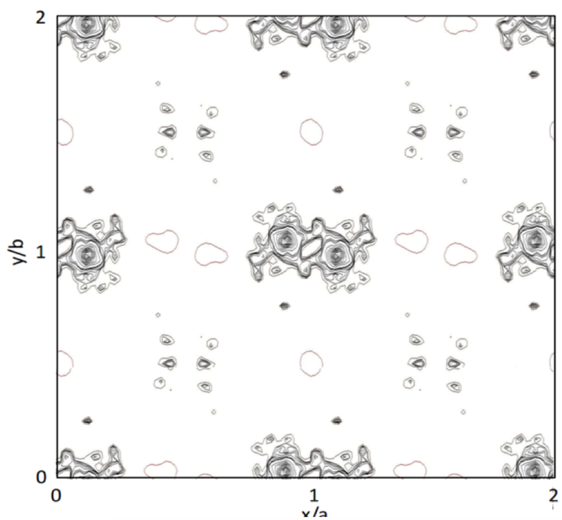
– collecting Bragg peaks from single crystal samples

D3 ILL Grenoble, Navid Qureshi

now developed for 2D powder diffraction

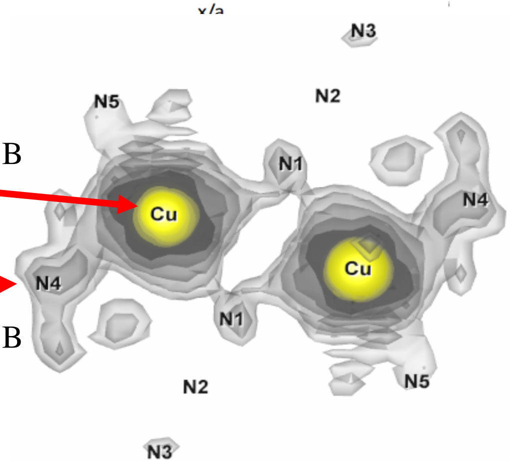
Examples of
using nuclear magnetic interference
(no polarization analysis)

Spin densities in molecular magnets



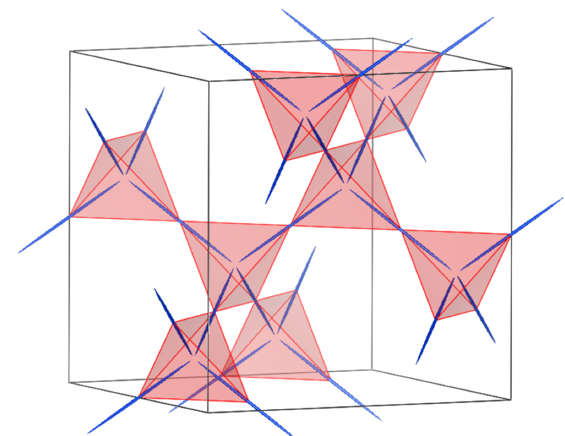
Aebersold
JACS 1998

$0.75(1)\ \mu_B$



$0.04(1)\ \mu_B$

Local susceptibilities anisotropies



magnetisation ellipsoids on Ho
(axis along <111>)

spin-ice
$Ho_2Ti_2O_7$

at 5K 1T:

$\chi_{\|} = 11.3(1)\ \mu_B$

$\chi_{\perp} = -0.1(1)\ \mu_B$

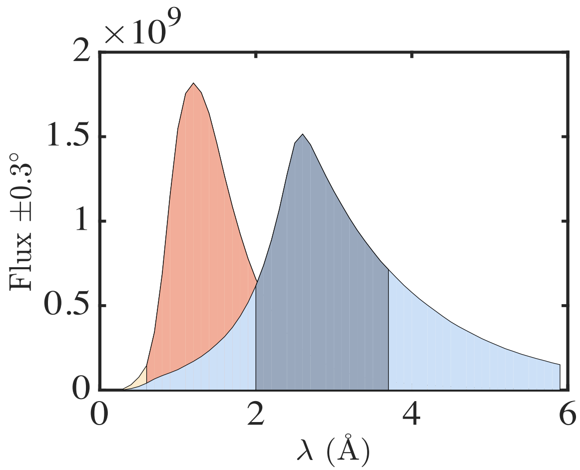
Iurii Kibalin & Arsen Gukasov
PRB 2019 *(from powder)*

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**

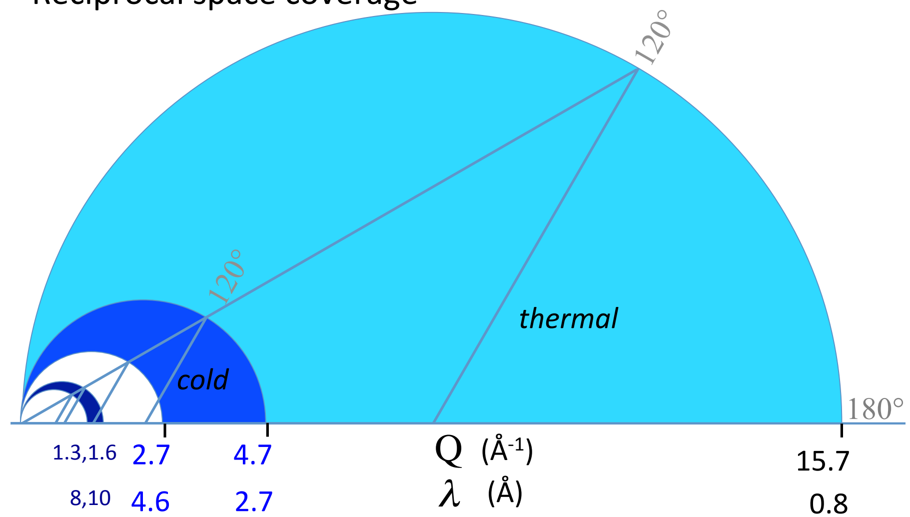
ESS polarised single crystal instrument MAGiC

in construction LLB, JCNS, PSI
User operation 2023

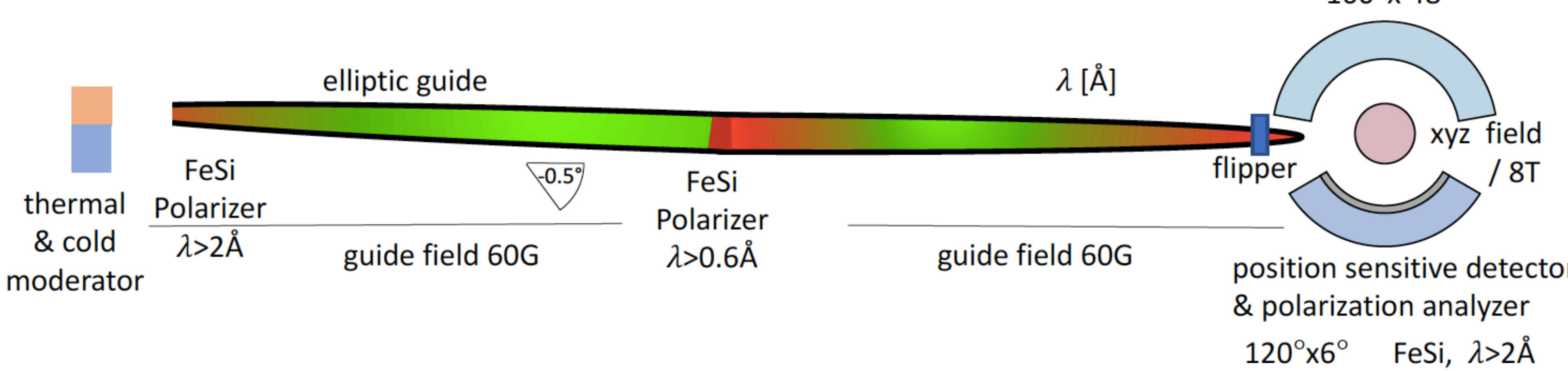


Neutron time-of-flight Laue instrument

Reciprocal space coverage



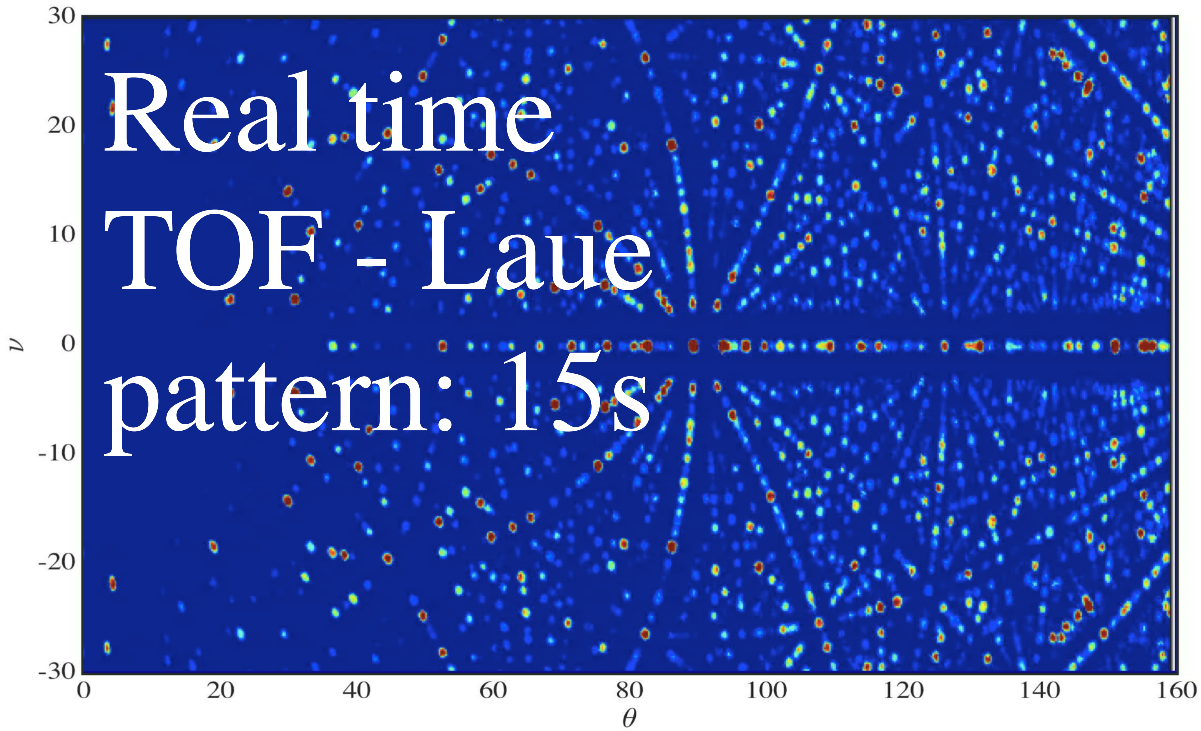
intense polarized white beam

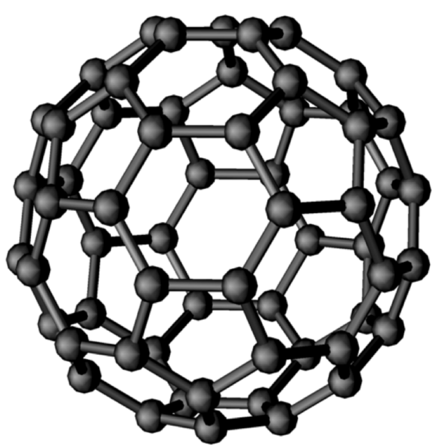


virtual MAGIC experiments

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

Cases

HoMnO$_3$

BiFeO$_3$

Spin ice

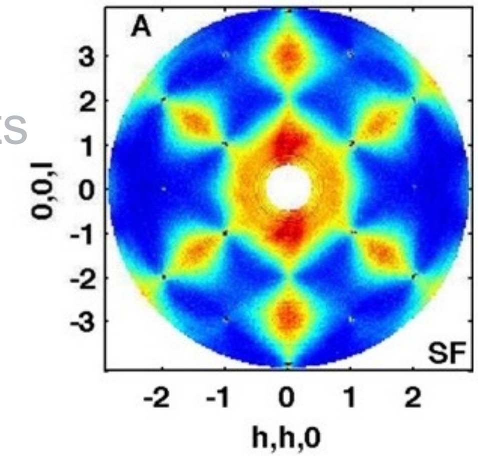
Bucky ball

Molecular magnets

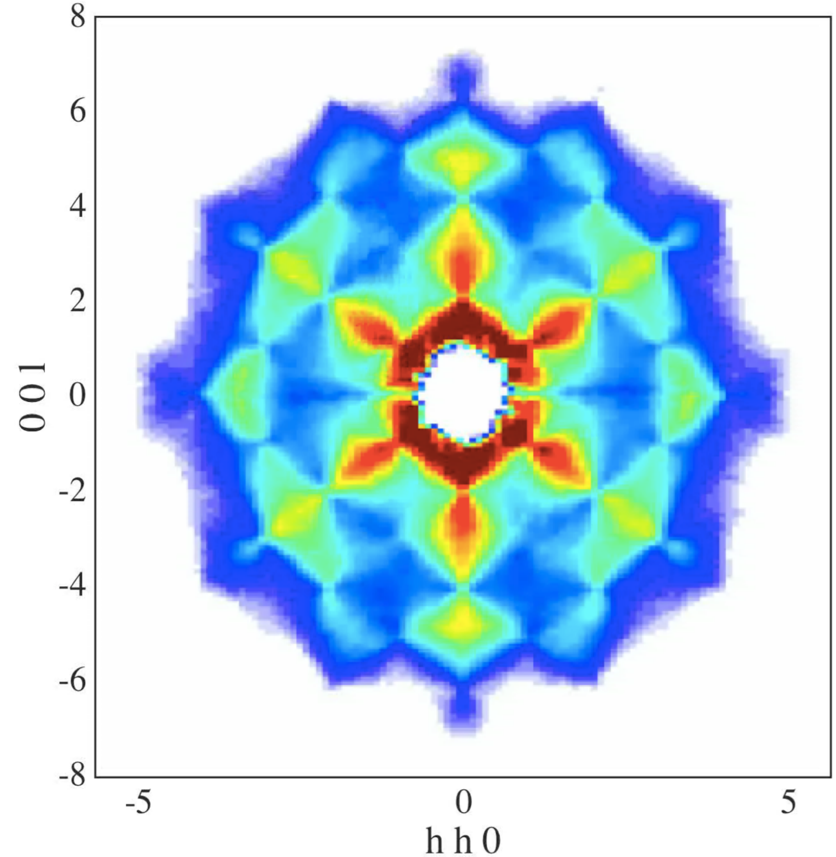
$Ho_2Ti_2O_7$

T. Fennell *et* al.
Science 2009

D7: $2x10^6$ n/s/cm^2



MAGiC: $2x10^{\mathbf{9}}$ n/s/cm^2



10 min & 10 mm^3

virtual MAGIC experiments

Cases

HoMnO$_3$

BiFeO$_3$

Spin ice

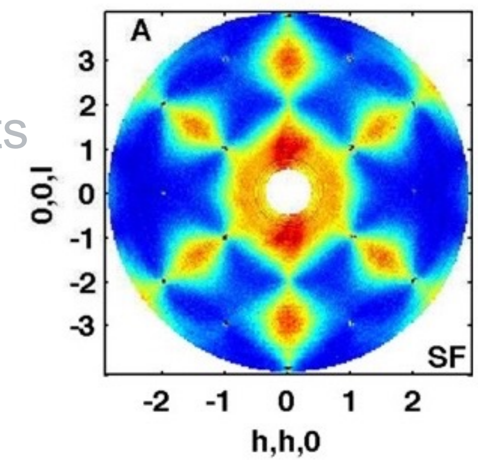
Bucky ball

Molecular magnets

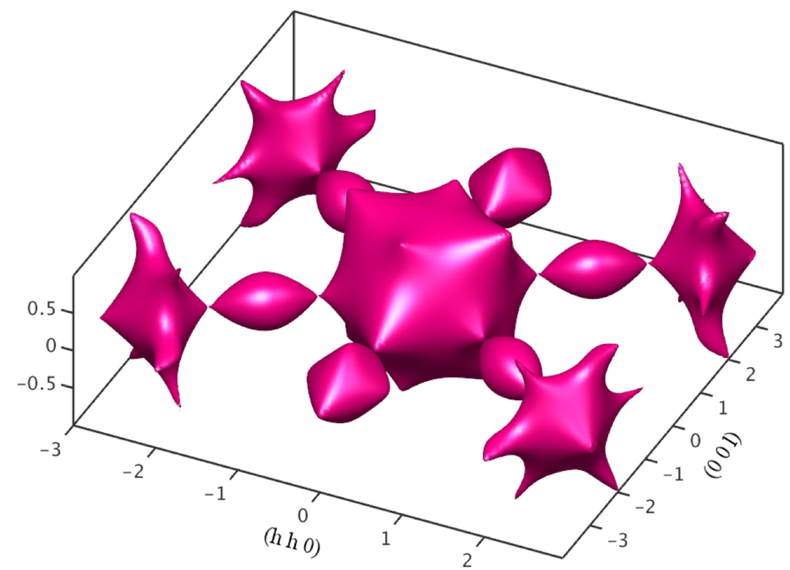
$Ho_2Ti_2O_7$

T. Fennell *et* al.
Science 2009

D7: 2x10^6 n/s/cm^2



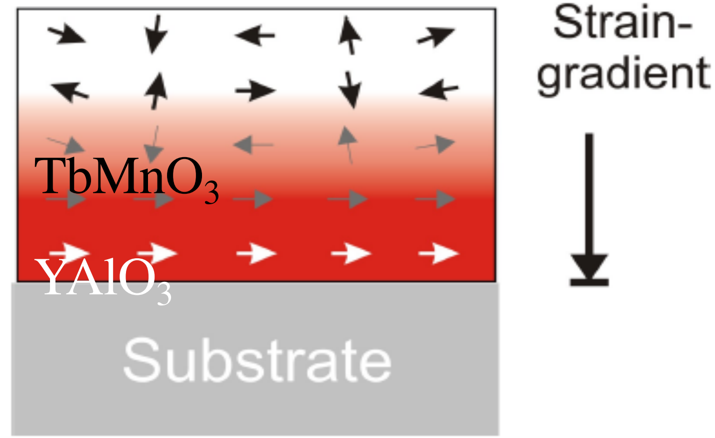
MAGiC: 2x10^{9} n/s/cm^2



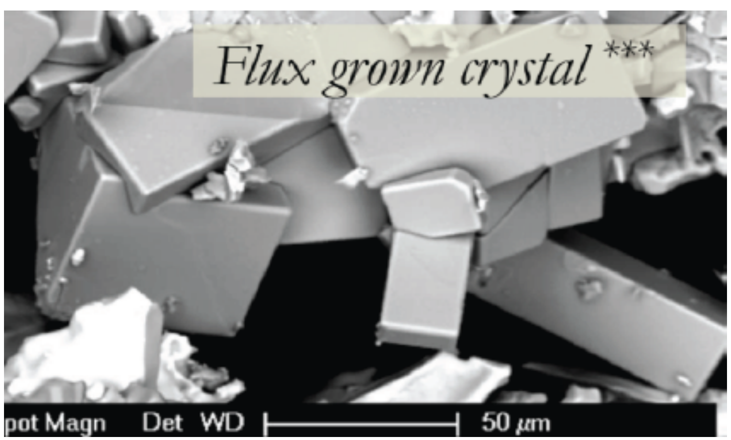
10 min & 10 mm^3

Future:
Small moments, small samples, or heterostructures

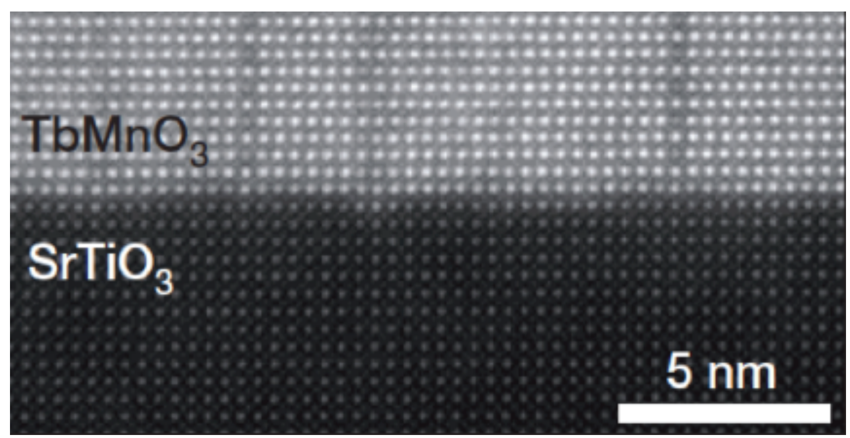
Many single crystalline materials are only available in very small quantities



Adapted from J. White et al., Phys. Rev. Lett. **111**, 037201 (2013)



Courtesy Dr. M. Valldor



S. Farokhipoor et al, Nature Materials **515** , 379 (2015)

Summary

Neutron Spin

$\mathbf{S}$

nuclear interaction

N — coherent — nsf

$\mathbf{I} \neq 0$ — spin-incoherent, isotropic — 1/3 nsf, 2/3 sf

hydrogenous materials

magnetic moment

$\boldsymbol{\mu}$

magnetic dipolar interaction

$\mathbf{M}_{\mathbf{Q}}^{\perp}$ — anisotropic

$\mathrm{I}, \mathbf{M}_{\mathbf{Q}}^{\perp}$

complex frustrated magnetism

Polarisation

$$2\langle \mathbf{S} \rangle = \mathbf{P}$$

probing vector properties and interference
separation of scattering terms