# Magnetic neutron diffraction

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# Magnetic moment-Rare earths

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- **Progressive filling of 4f levels**
  - Strong Hund's rules
  - Strong spin-orbit interaction
  - Weak CEF
- **Unpaired electrons**
  - Total angular momentum

$$J = L + 2S$$

$$\mu = g_J \mu_B J \approx g_J J \frac{e\hbar}{2m_e}$$

*f*-shell ($l = 3$)

| $n$ | $l_z = 3$, | 2, | 1, | 0, | $-1$, | $-2$, | $-3$ | $S$ | $L = \lvert\Sigma l_z\rvert$ | $J$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | ↓ | | | | | | | 1/2 | 3 | 5/2 | | $^2F_{5/2}$ |
| 2 | ↓ | ↓ | | | | | | 1 | 5 | 4 | | $^3H_4$ |
| 3 | ↓ | ↓ | ↓ | | | | | 3/2 | 6 | 9/2 | | $^4I_{9/2}$ |
| 4 | ↓ | ↓ | ↓ | ↓ | | | | 2 | 6 | 4 | $J = \lvert L - S\rvert$ | $^5I_4$ |
| 5 | ↓ | ↓ | ↓ | ↓ | ↓ | | | 5/2 | 5 | 5/2 | | $^6H_{5/2}$ |
| 6 | ↓ | ↓ | ↓ | ↓ | ↓ | ↓ | | 3 | 3 | 0 | | $^7F_0$ |
| 7 | ↓ | ↓ | ↓ | ↓ | ↓ | ↓ | ↓ | 7/2 | 0 | 7/2 | | $^8S_{7/2}$ |
| 8 | ↓↑ | ↑ | ↑ | ↑ | ↑ | ↑ | ↑ | 3 | 3 | 6 | | $^7F_6$ |
| 9 | ↓↑ | ↓↑ | ↑ | ↑ | ↑ | ↑ | ↑ | 5/2 | 5 | 15/2 | | $^6H_{15/2}$ |
| 10 | ↓↑ | ↓↑ | ↓↑ | ↑ | ↑ | ↑ | ↑ | 2 | 6 | 8 | | $^5I_8$ |
| 11 | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↑ | ↑ | ↑ | 3/2 | 6 | 15/2 | $J = L + S$ | $^4I_{15/2}$ |
| 12 | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↑ | ↑ | 1 | 5 | 6 | | $^3H_6$ |
| 13 | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↑ | 1/2 | 3 | 7/2 | | $^2F_{7/2}$ |
| 14 | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | ↓↑ | 0 | 0 | 0 | | $^1S_0$ |

[a] ↑ = spin $\frac{1}{2}$; ↓ = spin $-\frac{1}{2}$.

# Transition metals

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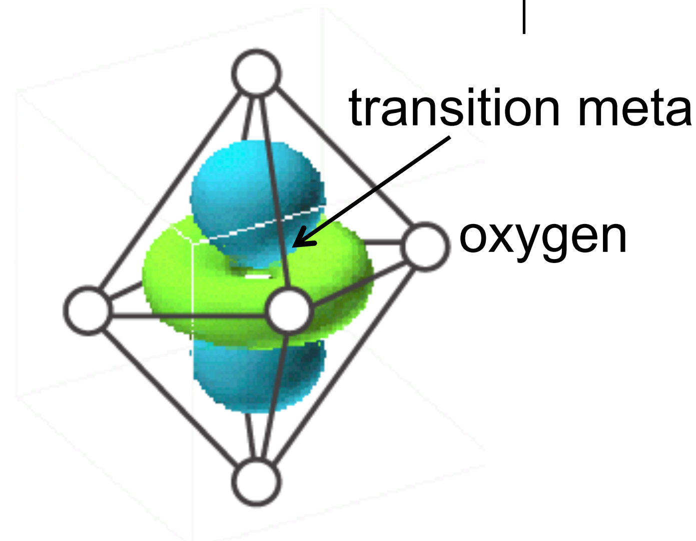
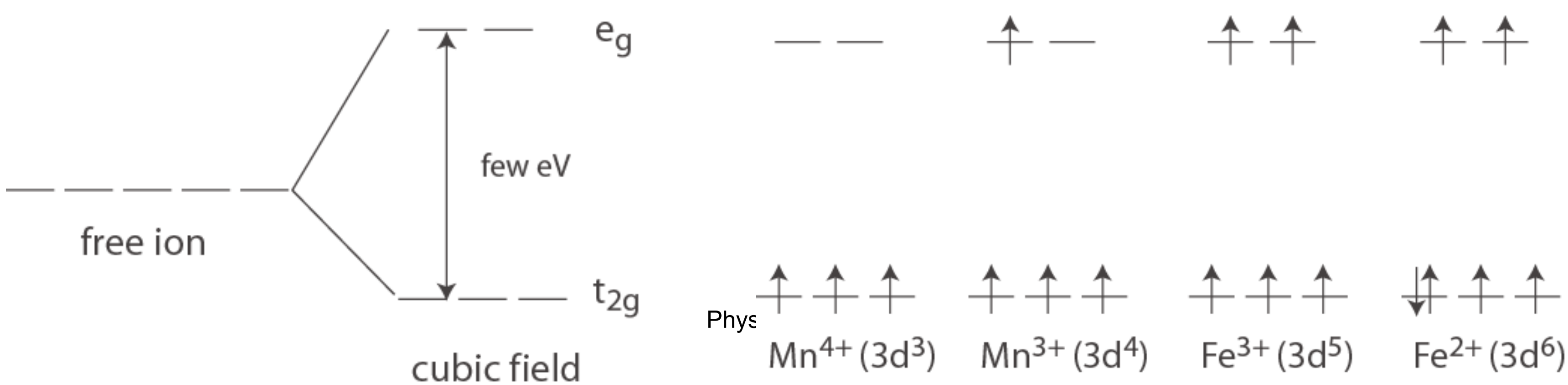
- **Progressive filling of 3d levels**
  - Strong Hund's rules interactions
  - Strong CEF
  - Weak spin-orbit interaction
- **Unpaired electrons**
  - Spin moment
  - Orbital moment (quenched)




$$\mu = g\mu_B S \approx 2S\frac{e\hbar}{2m_e}$$



# Magnetic structures

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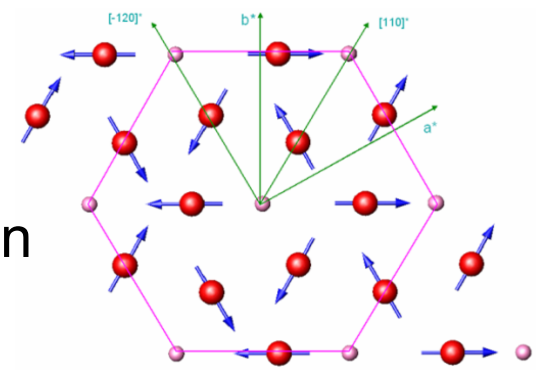
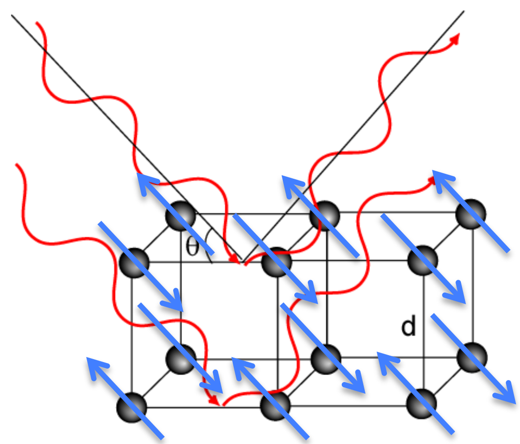
- Exchange coupling between moments leads to ordering
  - Direct exchange
  - Superexchange (insulators)
  - RKKY (metals)
  - Dipolar
- Magnetic anisotropy leads to moment direction
- Magnetic structures defined by
  - Propagation vector(s)
  - Moment size
  - Moment direction(s)

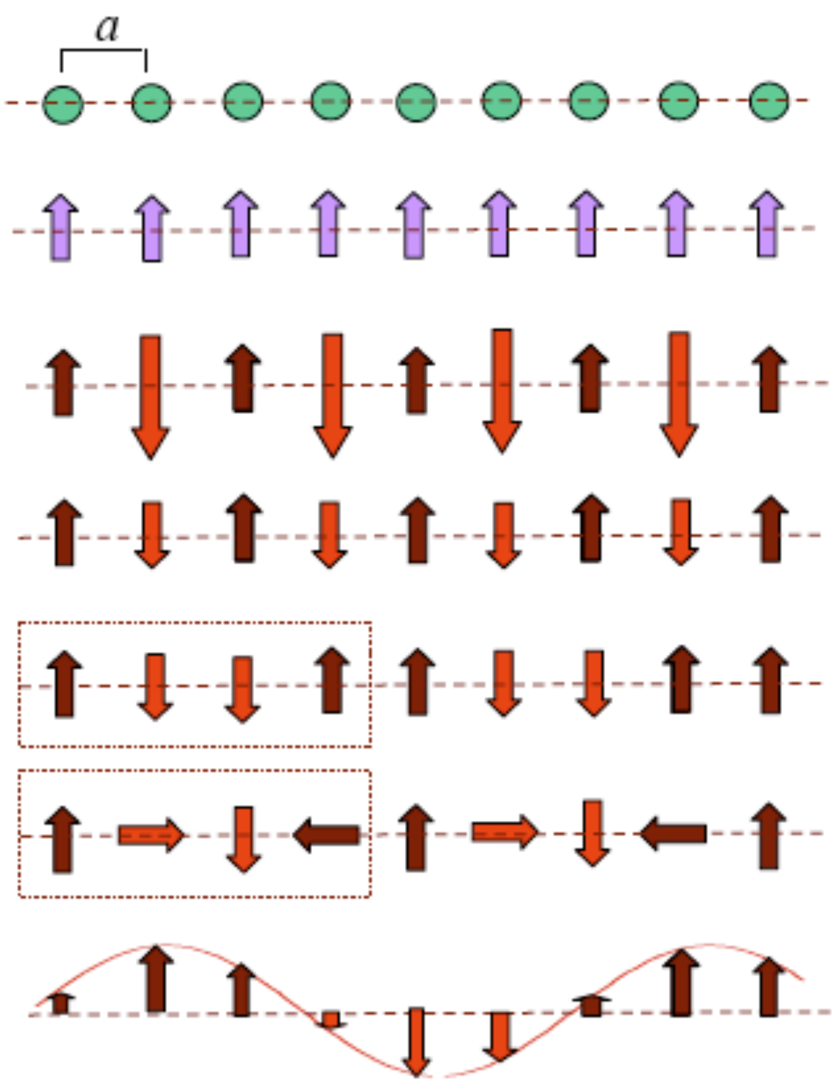



Elastic scattering - Bragg's Law

$2d\sin\theta = n\lambda$

# 1-D cartoons




**nuclear structure**
atoms separated by lattice spacing *a*

**ferromagnet**
collinear moments; commensurate

**simple ferrimagnet**

**simple antiferromagnet**

**antiferromagnet with larger unit cell**

**non-collinear antiferromagnet**

**incommensurate antiferromagnet**

# Neutron magnetism

- Spin-1/2 particle
- Magnetic moment


$$\mu_n = -\gamma\mu_N = -1.913\frac{e\hbar}{2m_p}$$

$$\mu_n / \mu_e \approx m_e / m_p = 1/2000$$

# Dipole interaction

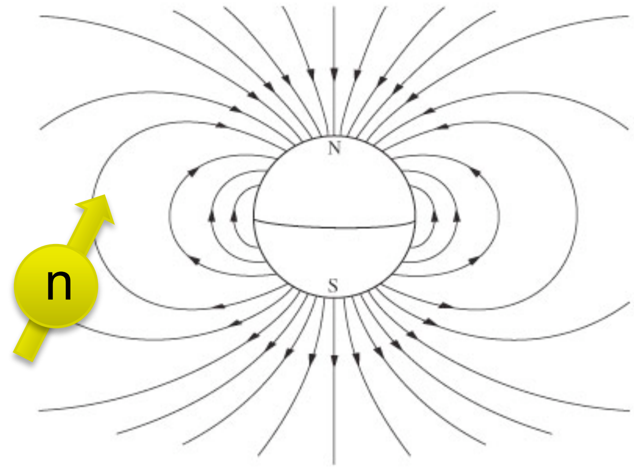
Interaction between neutron and electron

$$U = -\boldsymbol{\mu}_n \cdot \mathbf{B} = \frac{\mu_0}{4\pi}\frac{\gamma e^2}{m_e}\boldsymbol{\sigma}\cdot\mathbf{B} = \gamma r_0 \boldsymbol{\sigma}\cdot\mathbf{B}$$

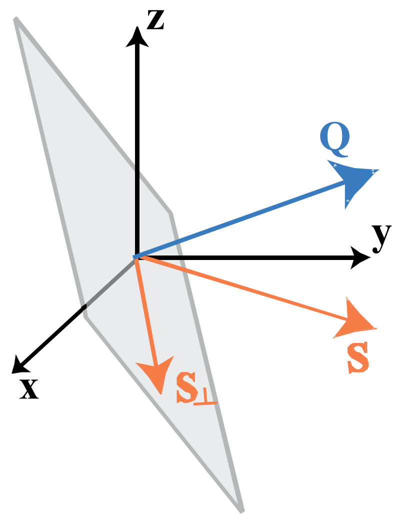
$$U^{uv} = \langle u | b - p\mathbf{S}_\perp \cdot \boldsymbol{\sigma} | v \rangle$$

$$p = \gamma r_0 g S f(\mathbf{Q}) \qquad \mathbf{S}_\perp = \hat{S} - (\hat{S}\cdot\hat{Q})\hat{S}$$

strength




$$\mathbf{B} = \mathbf{B}_L + \mathbf{B}_S$$




$$U^{++} = b - pS_{\perp z}$$

$$U^{--} = b + pS_{\perp z}$$

$$U^{+-} = -p(S_{\perp x} + iS_{\perp y})$$

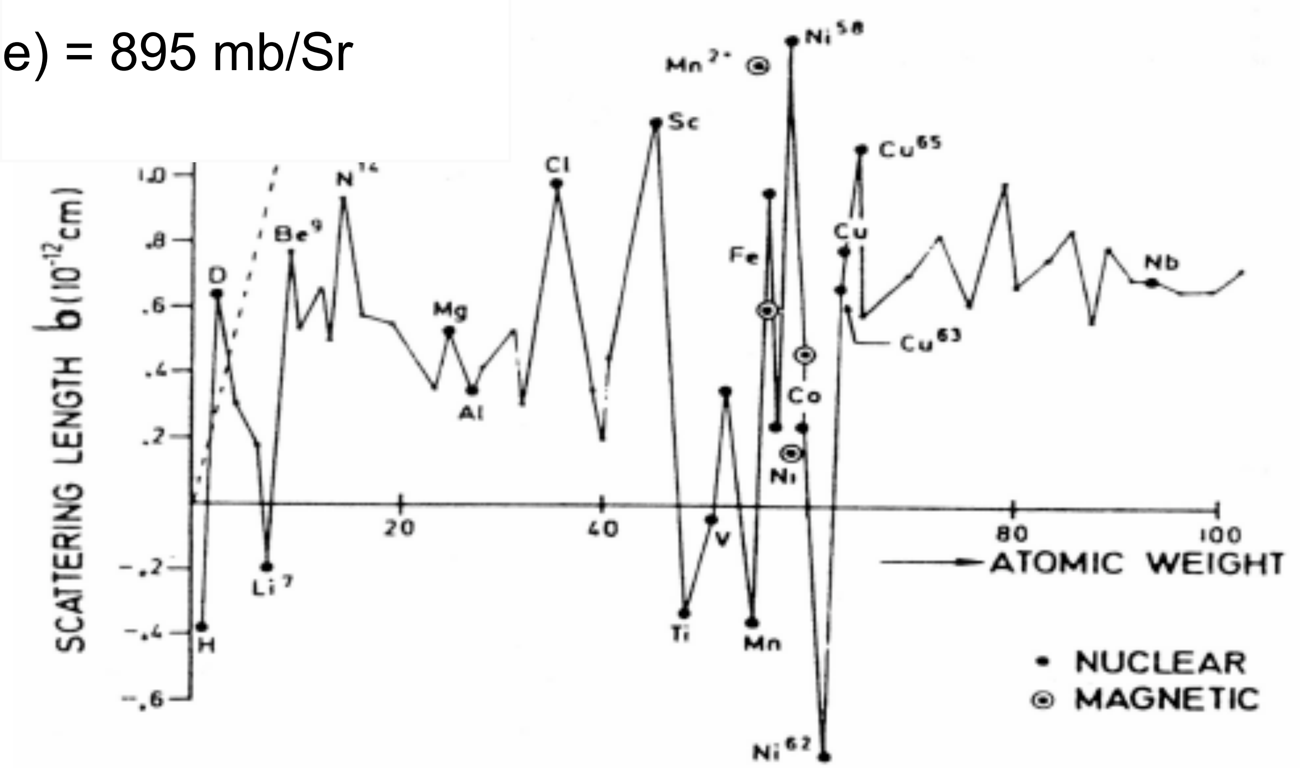
$$U^{-+} = -p(S_{\perp x} - iS_{\perp y})$$

Only moment projection perp. to **Q** will scatter neutrons

# Magnetic cross-section

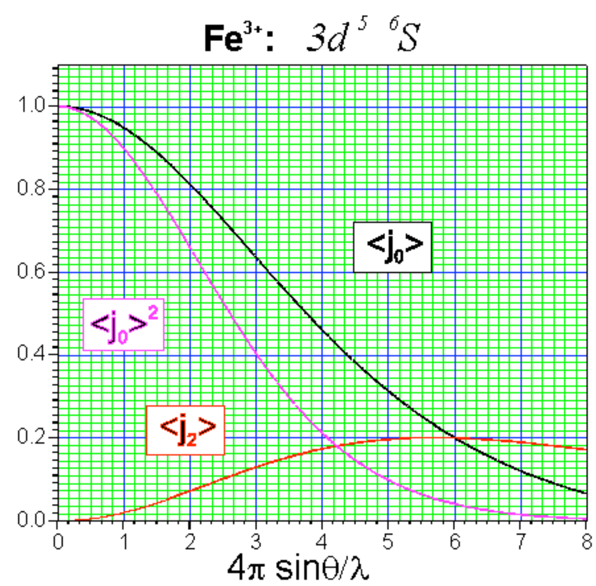
$$(\gamma r_0)^2 = 291 \text{ millibarns/steradian}$$

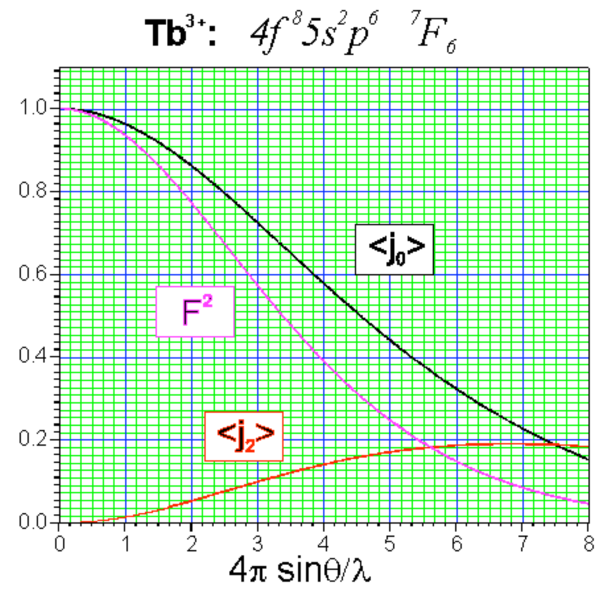
$b^2(\text{Fe}) = 895$ mb/Sr



# Magnetic form factor

f(**Q**): Fourier transform of the atomic magnetization density



# Magnetic structure factor



Magnetic structure factor is actually a vector quantity, but for collinear structure, can be simplified

$$F_M(\boldsymbol{\tau}) = \sum_d \frac{1}{2} \underbrace{g_d \langle S_d \rangle}_{\text{ordered moment}} \underset{\substack{\uparrow \\ \text{moment direction}}}{\sigma_d} \underbrace{F_d(Q)}_{\text{form factor}} \underbrace{\exp(-W_d)}_{\text{DW factor}} \underbrace{\exp(i\boldsymbol{\tau}\cdot\mathbf{d})}_{\text{phase factor}}$$

Scattering differential cross-section for *unpolarized* beam

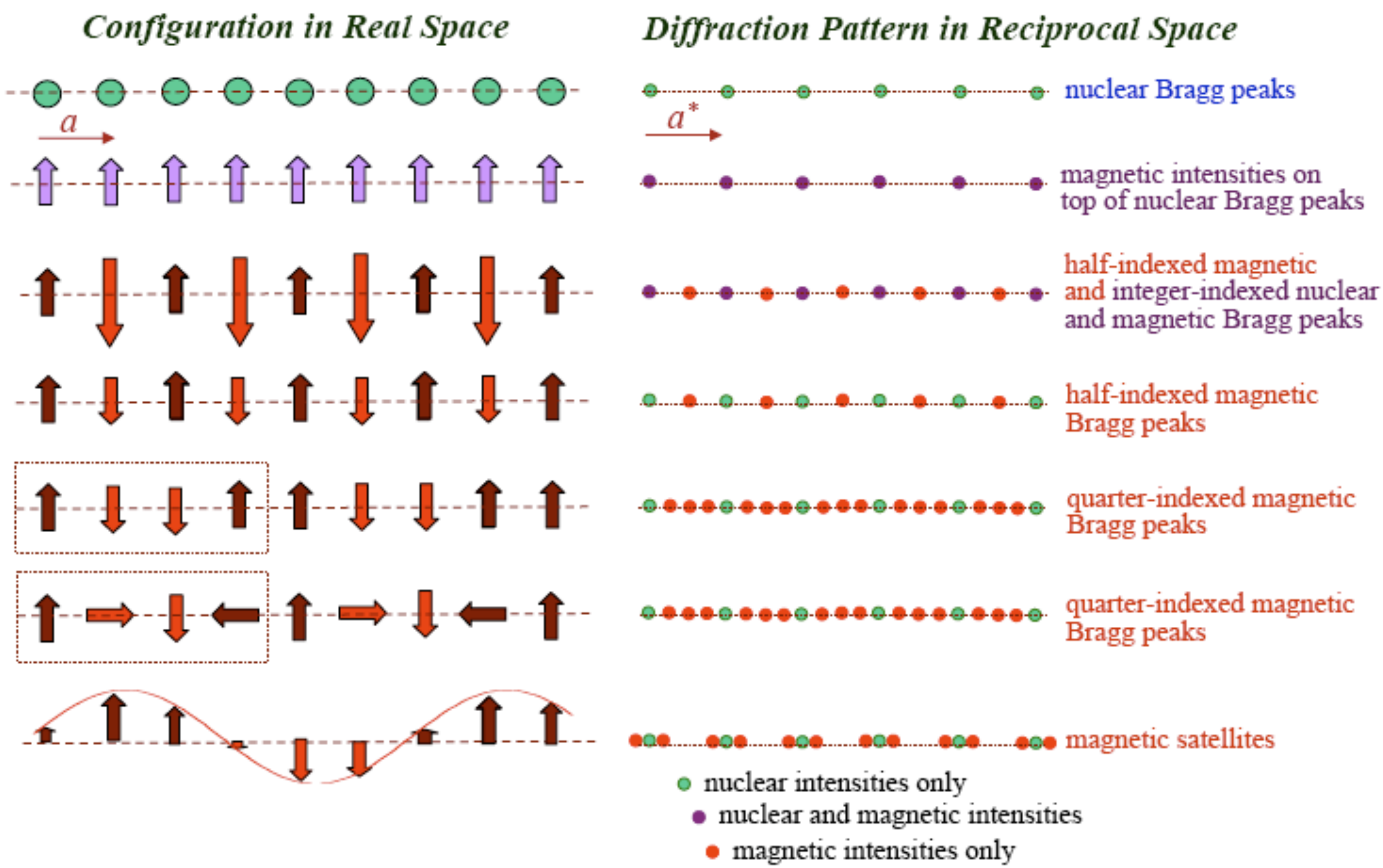
$$\frac{d\sigma}{d\Omega} = N \underset{\substack{\nearrow \\ \text{strength}}}{r_0^2} \underbrace{(1-\hat{\tau}_z^2)}_{\text{dipole scattering}} \left|F_M(\boldsymbol{\tau})\right|^2$$

More generally

$$\frac{d\sigma}{d\Omega} = N r_0^2 \sum_\tau \delta(\mathbf{Q}-\boldsymbol{\tau}) \left|\hat{\mathbf{Q}} \times \{\mathbf{M}(\boldsymbol{\tau}) \times \hat{\mathbf{Q}}\}\right|^2$$

Fourier transform of magnetization density

# 1-D Cartoons



Configuration in Real Space
Diffraction Pattern in Reciprocal Space
a
a*
nuclear Bragg peaks
magnetic intensities on top of nuclear Bragg peaks
half-indexed magnetic and integer-indexed nuclear and magnetic Bragg peaks
half-indexed magnetic Bragg peaks
quarter-indexed magnetic Bragg peaks
quarter-indexed magnetic Bragg peaks
magnetic satellites
nuclear intensities only
nuclear and magnetic intensities
magnetic intensities only

# Determine magnetic structure

- **Prescription**
  - Measure the magnetic propagation vector(s)
  - Magnetic space group
    - Limits the possible structures
    - You need to know the crystal structure
  - Detemine moment direction(s) (refinement)
- **Potential problems**
  - Magnetic domains
  - Crystallographic twinning
  - Multiple wavevectors/multi-q structures

# Confirmation of AF structure

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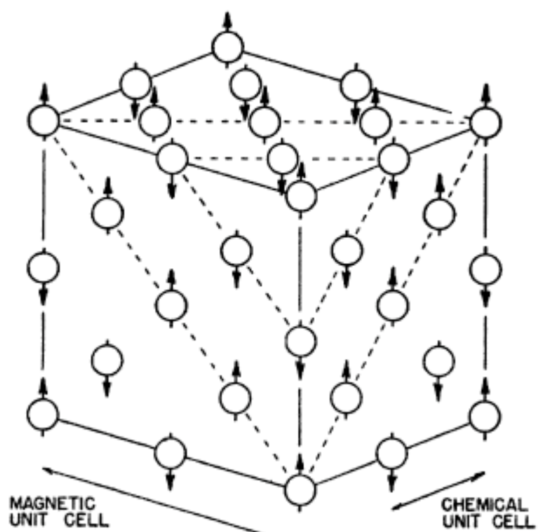
In 1949, Clifford Shull observed additional magnetic reflections in MnO which led to the confirmation of antiferromagnetism




Mn ATOMS IN MnO

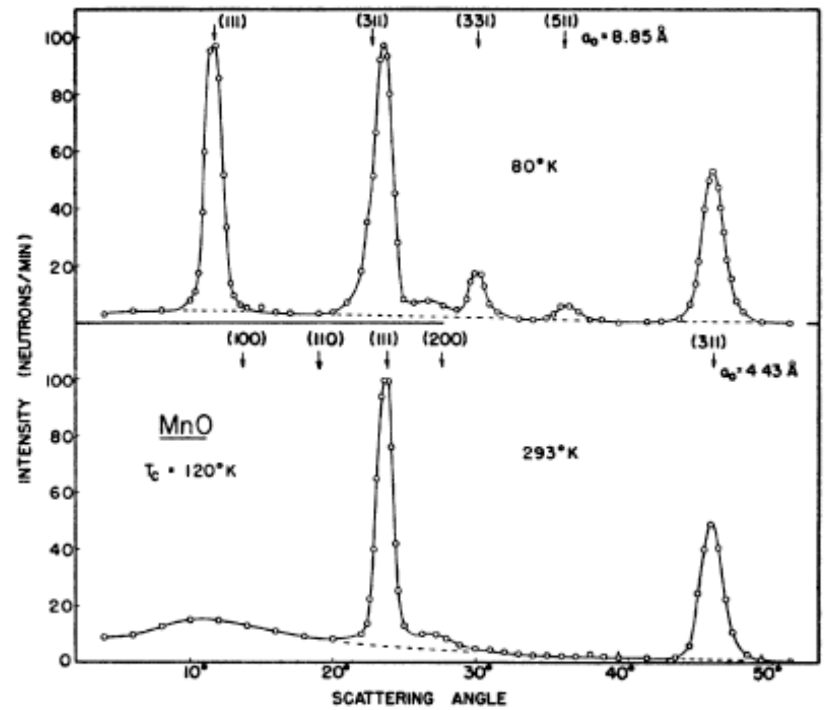



TABLE II. Comparison between observed MnO antiferromagnetic intensities and those calculated for various models of magnetic orientation with respect to crystallographic axes.

| | Calculated for various oriented models (a) | (b) | (c) | Observed (neutrons/min) |
|---|---|---|---|---|
| (111) | 1038 | 0 | 1560 | 1072 |
| (311) | 460 | 675 | ... | 308 |
| (331) | 129 | 109 | ... | 132 |
| (511), (333) | 54 | 24 | ... | 70 |

Shull and J. S. Smart, Phys Rev **76**, 1256 (1949).

C. G. Shull et al., Phys. Rev. **83,** 333 (1951).

# Cone structure of Er

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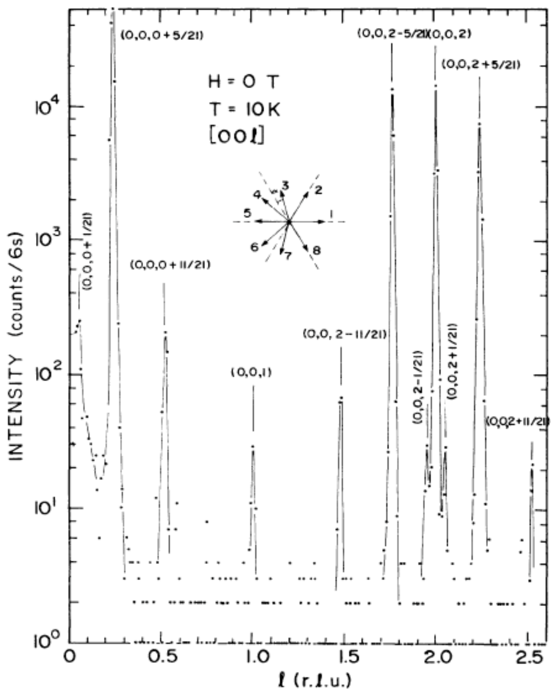
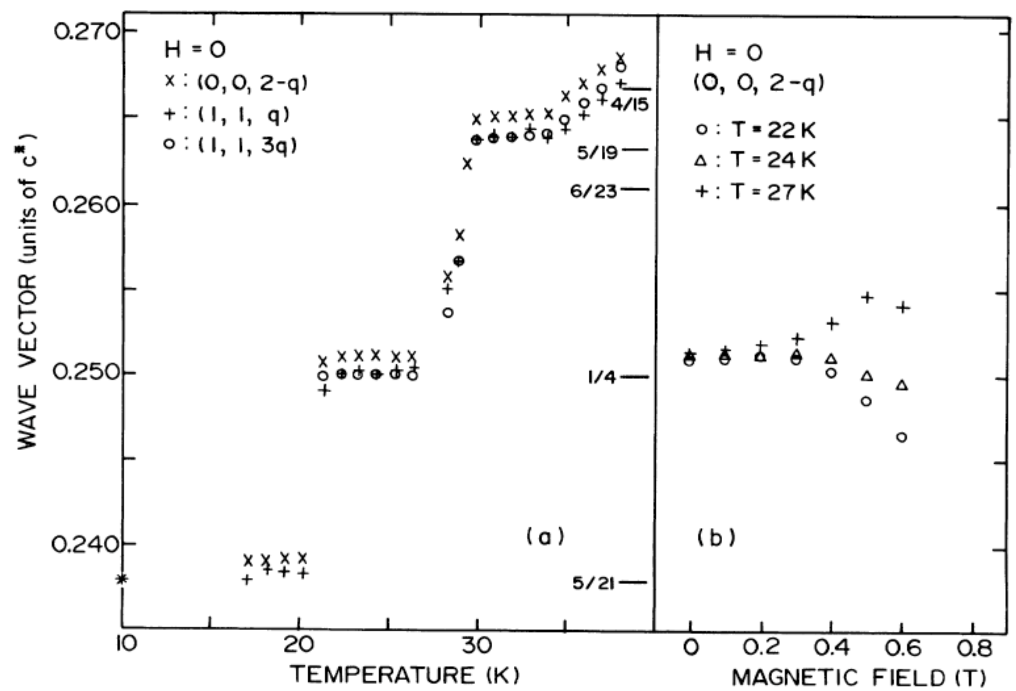



FIG. 6. Diffraction pattern from the $\mathbf{q}=(5/21)\mathbf{c}^*$ phase at 0 T and 10 K along the [00$l$] direction. The insert shows the first eight layers of the basal-plane spin-slip model for this structure.




- incommensurate
- Alternating cone structure
- Spin slips from magnetoelastic effect

H. Lin *et al., Phys. Rev B* ***45,*** *12873 (1992)*.

# Cone structure of Er

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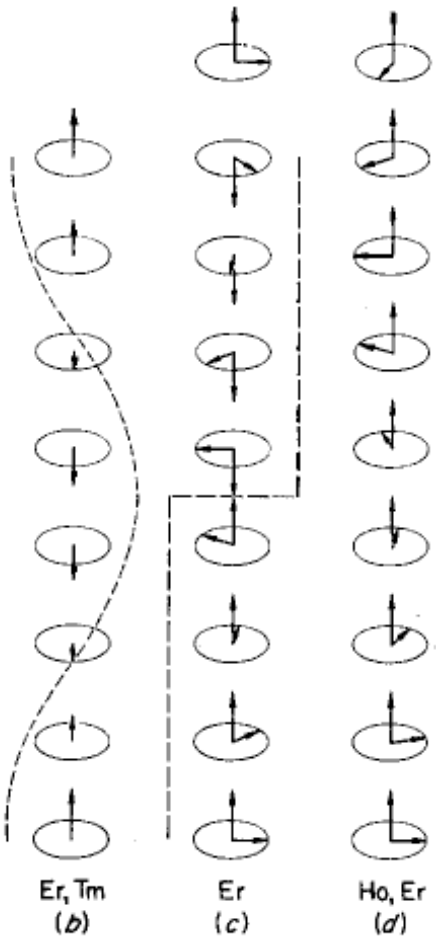
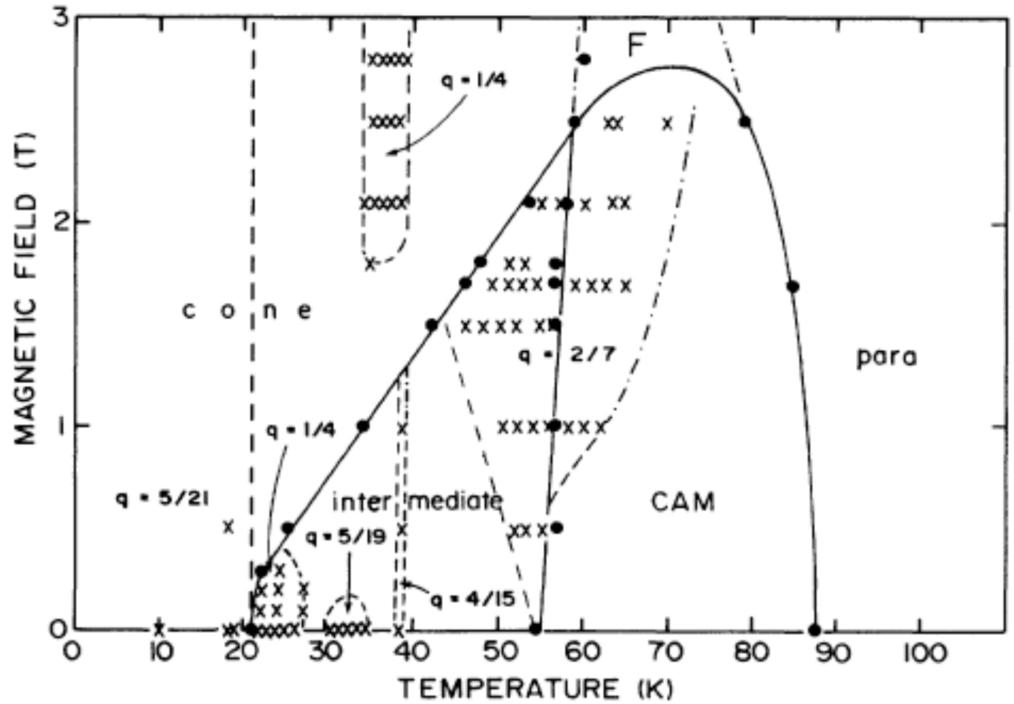
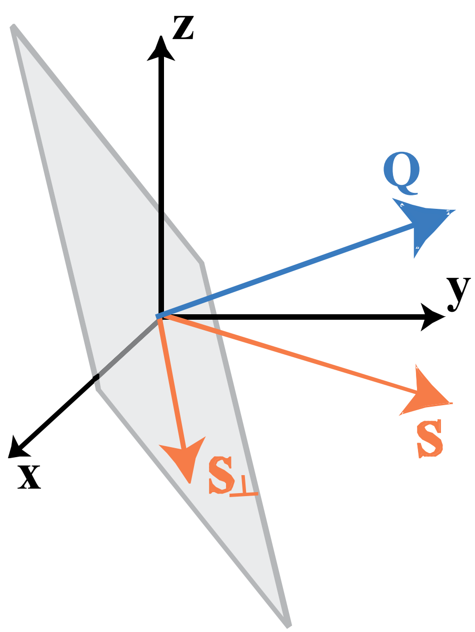



CAM Slip cone Cone



# Neutron polarization analysis

- **Why use polarization?**
  - Separate magnetic/nuclear scatt. (q=0 structures)
  - Refine structure determination (eg. canting)
  - Separate coherent/incoherent (diffuse scattering, mag. densities)



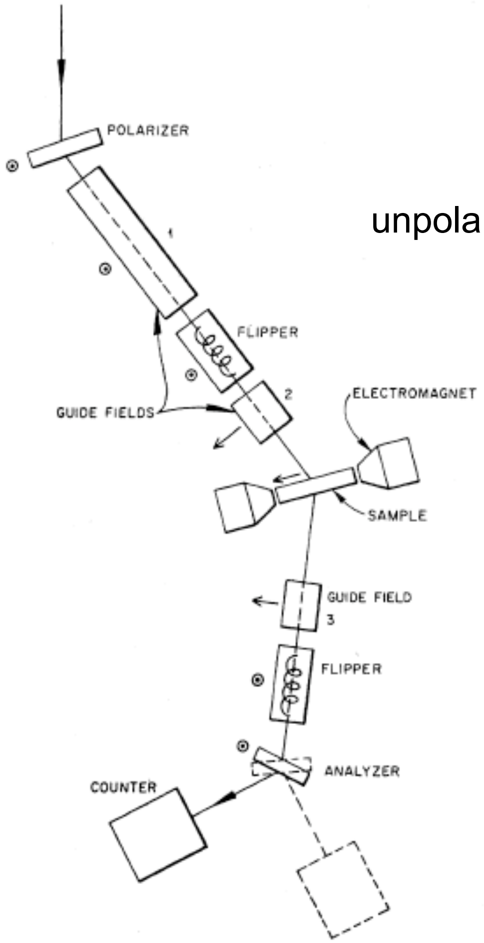

$$U^{++} = b - pS_{\perp z}$$

$$U^{--} = b + pS_{\perp z}$$

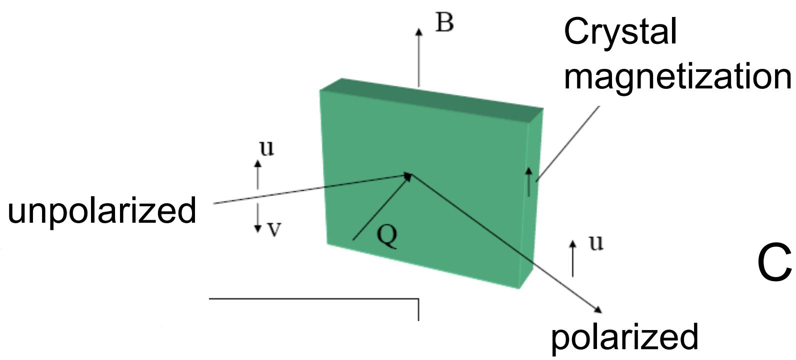
$$U^{+-} = -p(S_{\perp x} + iS_{\perp y})$$

$$U^{+-} = -p(S_{\perp x} - iS_{\perp y})$$

# Instrumentation




## Monochromator

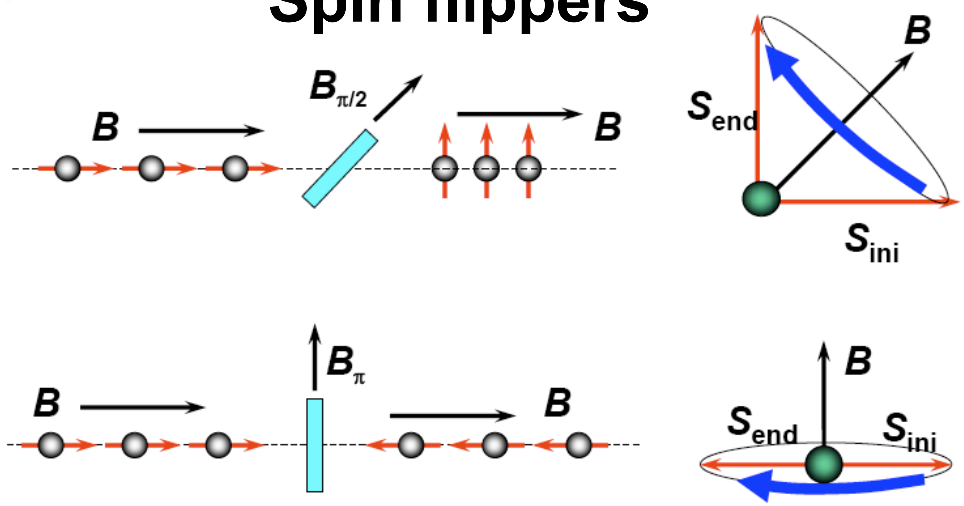



$$U^{++} = b - pS_{\perp z}$$

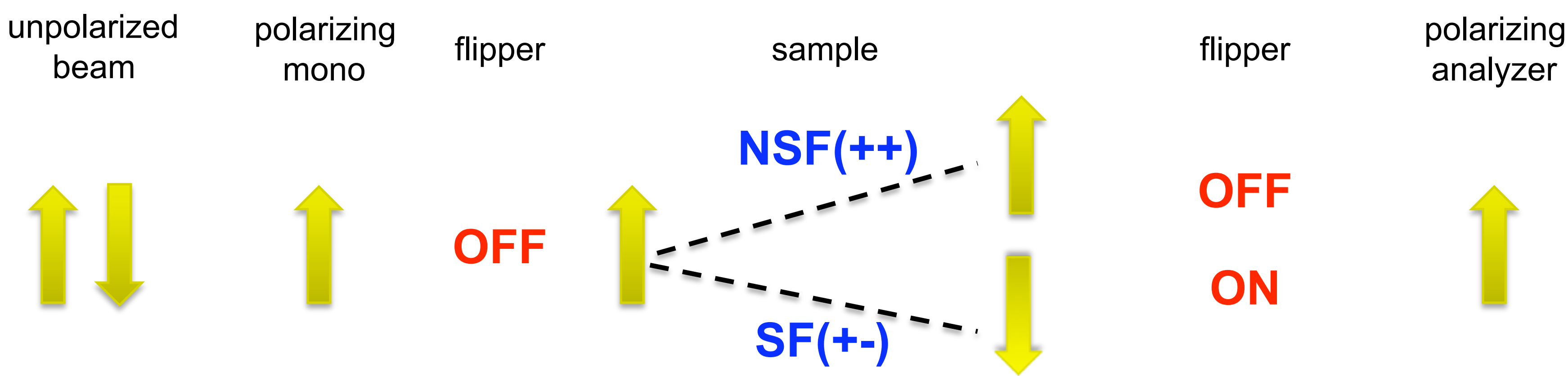
$$U^{--} = b + pS_{\perp z} \approx 0$$

$Cu_2MnAl$ (111) (Heusler)

## Spin flippers



# Spin-flip vs. Non-spin-flip




- **Useful modes**
  - **P || Q** (in-plane polarization): All magnetic scattering is SF
  - **P ⊥ Q** (vertical polarization): magnetic scattering can be SF & NSF

# Polarized experiments

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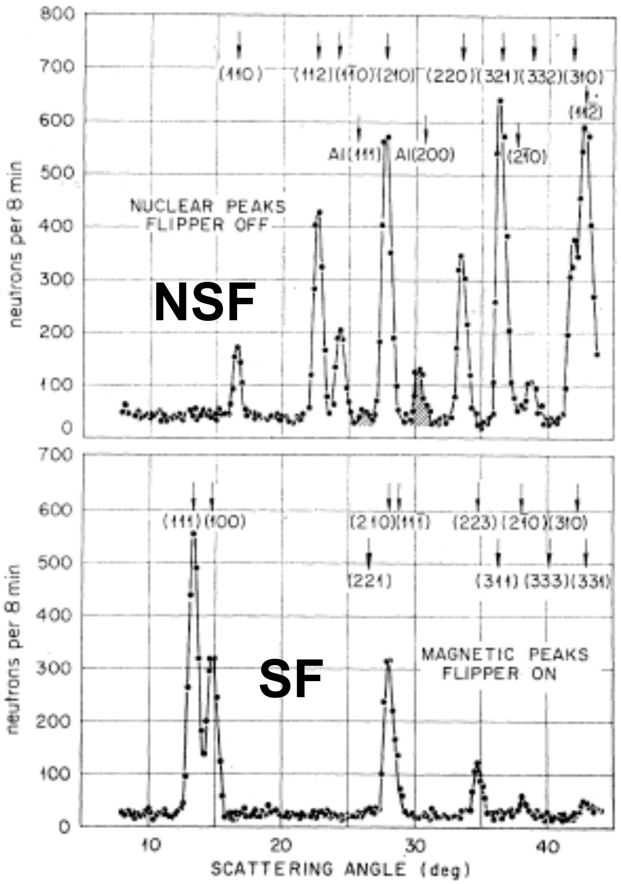


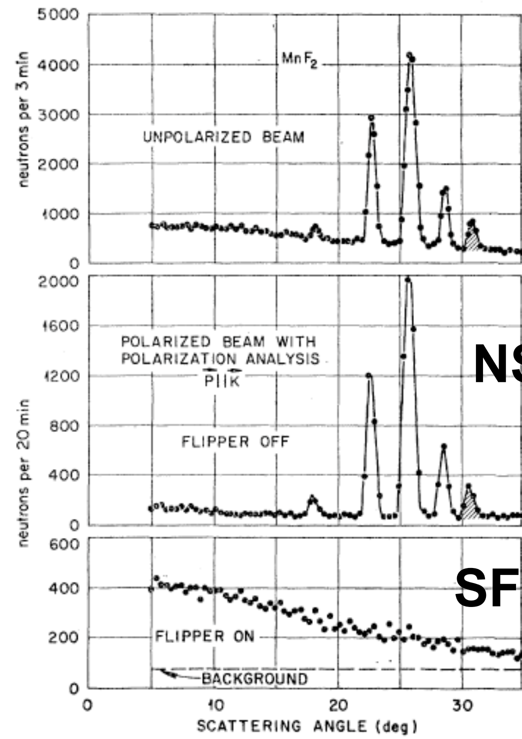
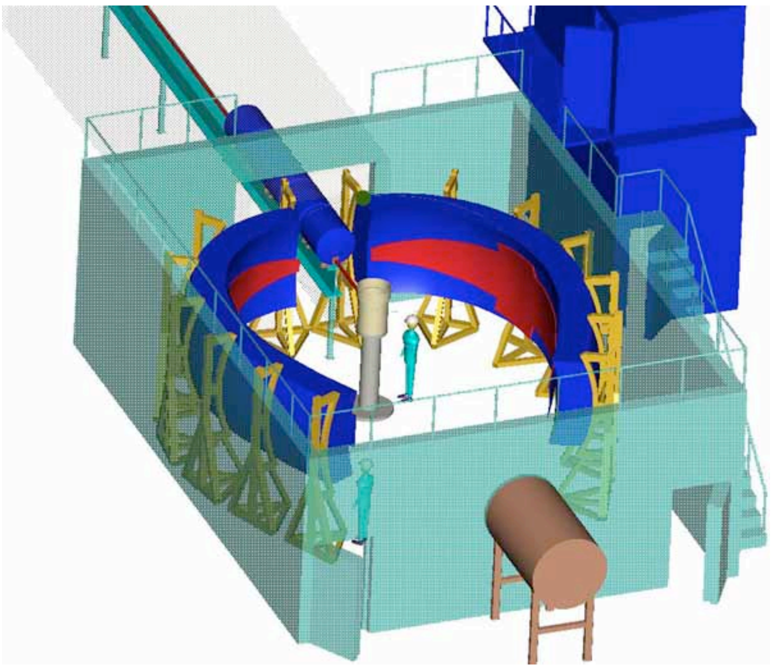




FIG. 5. $MnF_2$ powder pattern—separation of paramagnetic scattering through polarization analysis. No analyzer was used in the unpolarized-beam experiment. Note the loss of intensity in the polarization analysis experiment.

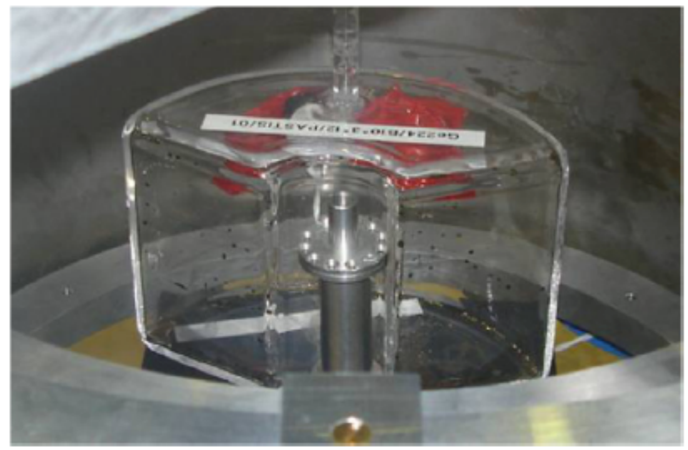
**Separation of magnetic/nuclear** **Paramagnetic scattering**

Moon, Koehler, Riste, Phys. Rev **181**, 920 (1969).

# Polarization @ pulsed source



Heusler mono won't work for wide angle scattering



$^3$He polarizers

# Further references

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  - G. Squires, “Intro to theory of thermal neutron scattering”, Dover, 1978.
  - S. Lovesey, “Theory of neutron scattering from condensed matter”, Oxford, 1984.
  - Moon, Koehler, Riste, Phys. Rev **181**, 920 (1969).
  - R. Pynn, http://www.mrl.ucsb.edu/~pynn/.
- **Structural refinements**
  - GSAS http://www.ncnr.nist.gov/xtal/software/gsas.html
  - FullProf http://www.ill.eu/sites/fullprof/
- **Magnetic space groups**
  - Izyumov, Ozerov, “Neutron diffraction of magnetic materials”
  - Sarah program (representational analysis)