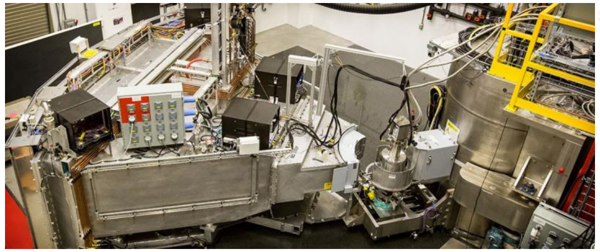
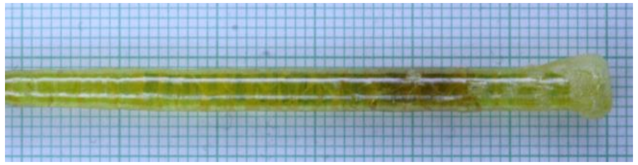
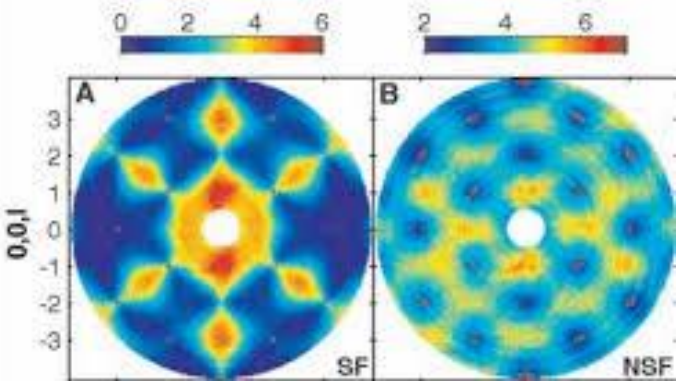
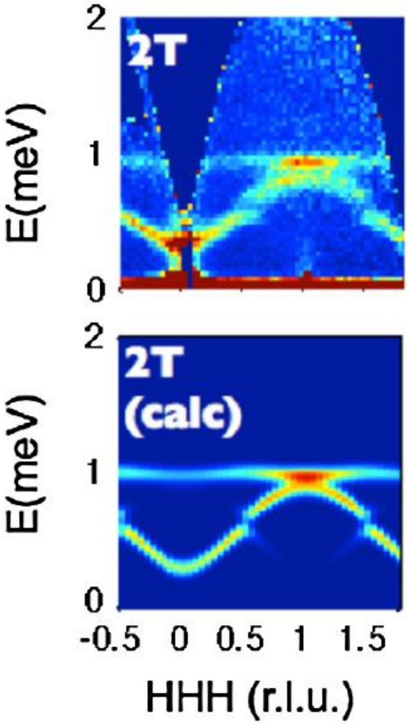
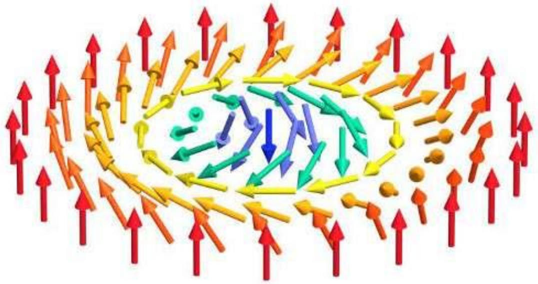
Magnetic Scattering

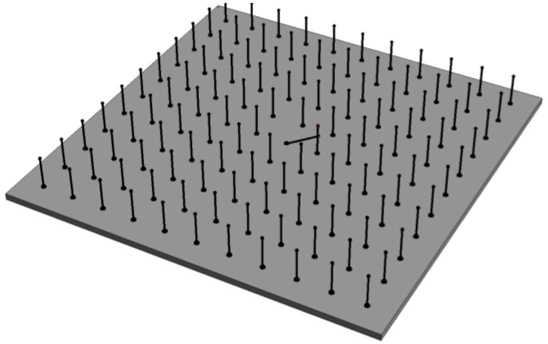












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Introduction to Magnetic Scattering

- Magnetic Scattering with **Neutrons**
- Essential tool for the study of magnetic materials
- Elastic Scattering (diffraction) – magnetic structure, phase transitions
- Inelastic Scattering (spectroscopy) – magnetic dynamics, excitations, interactions

- Magnetic Scattering with **X-rays**
- How does it work?
- When is it a good idea?

Suggestions for Further Reading…

- Magnetic Scattering with **Neutrons**:

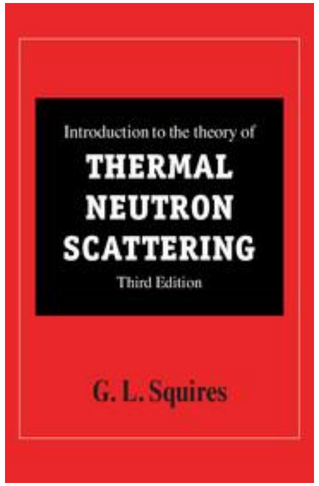
Introduction to the Theory of Thermal Neutron Scattering, G. L. Squires (2012)

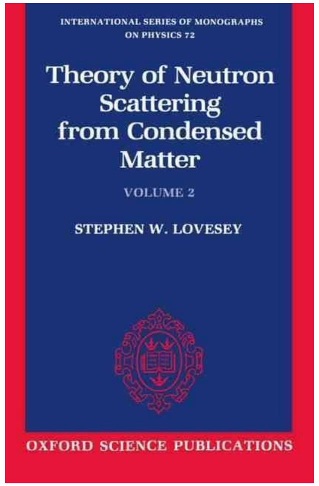
Theory of Neutron Scattering from Condensed Matter (Vol. 2), S. W. Lovesey (1984)

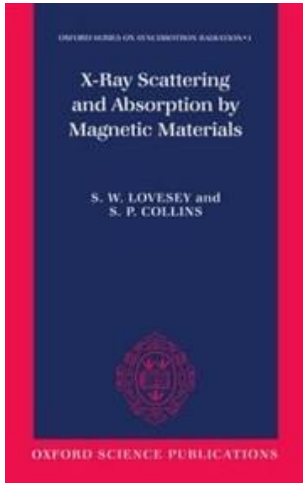
- Magnetic Scattering with **X-rays**:

X-ray Scattering and Absorption by Magnetic Materials, S. W. Lovesey & S. P. Collins (1996)

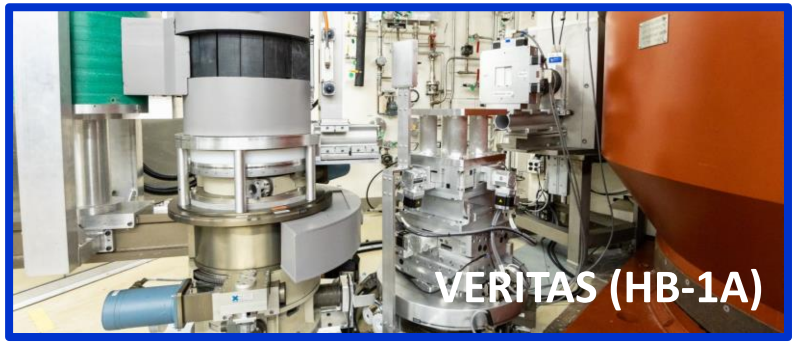
"*Magnetic Scattering*" by J. W. Lynn and B. Keimer in *Handbook of Magnetism* (arXiv:1910.01218)

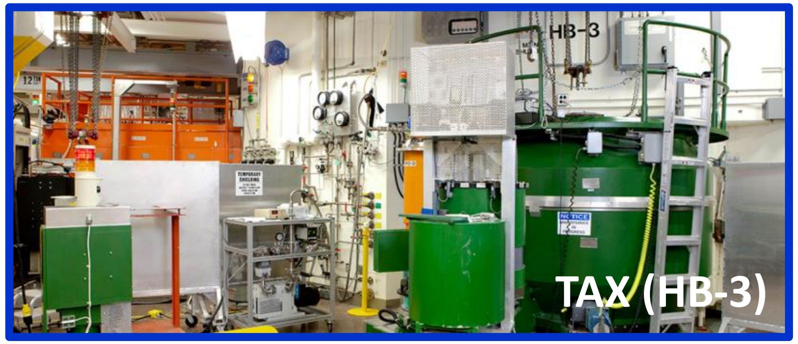


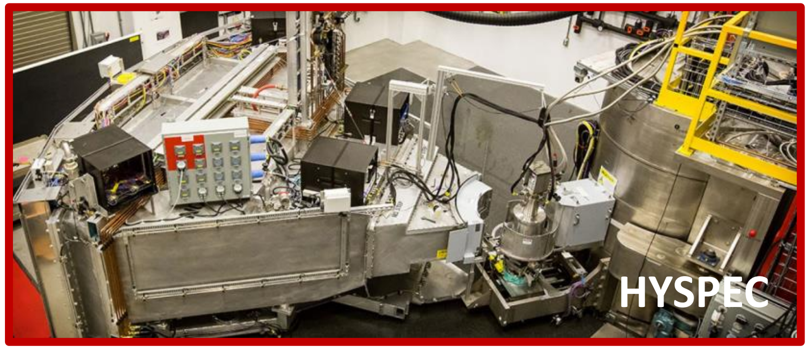


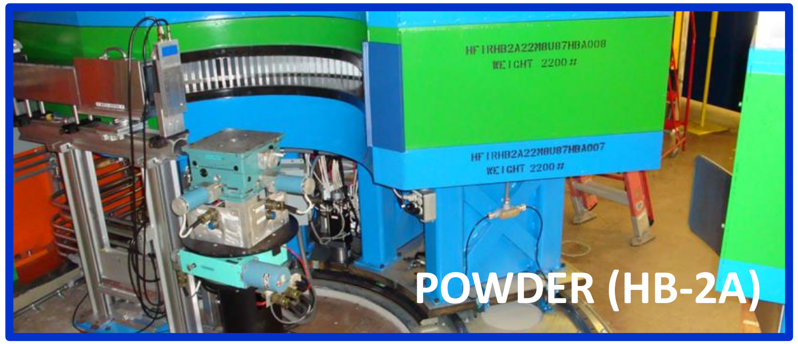


Magnetic Scattering with Neutrons

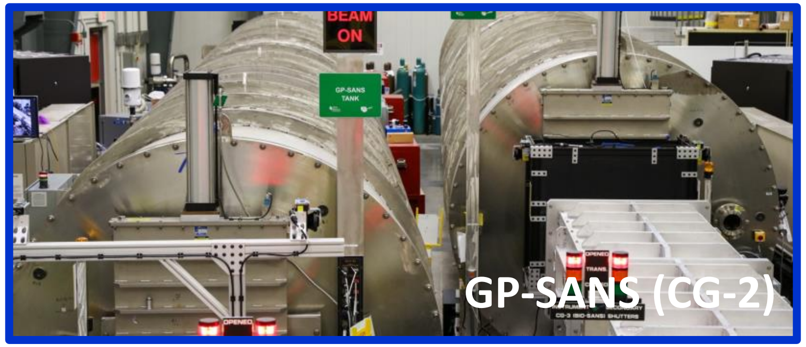

VERITAS (HB-1A)


TAX (HB-3)


HYSPEC


POWDER (HB-2A)


WAND^2 (HB-2C)


GP-SANS (CG-2)


POWGEN


DEMAND (HB-3A)


MAGREF

Magnetic Scattering with Neutrons

- Neutrons are spin ½ particles
- They carry no charge, but do carry a **magnetic dipole moment**:

$$\boxed{\mu_n = -\gamma \mu_N \boldsymbol{\sigma}}$$

(Pauli spin operator)

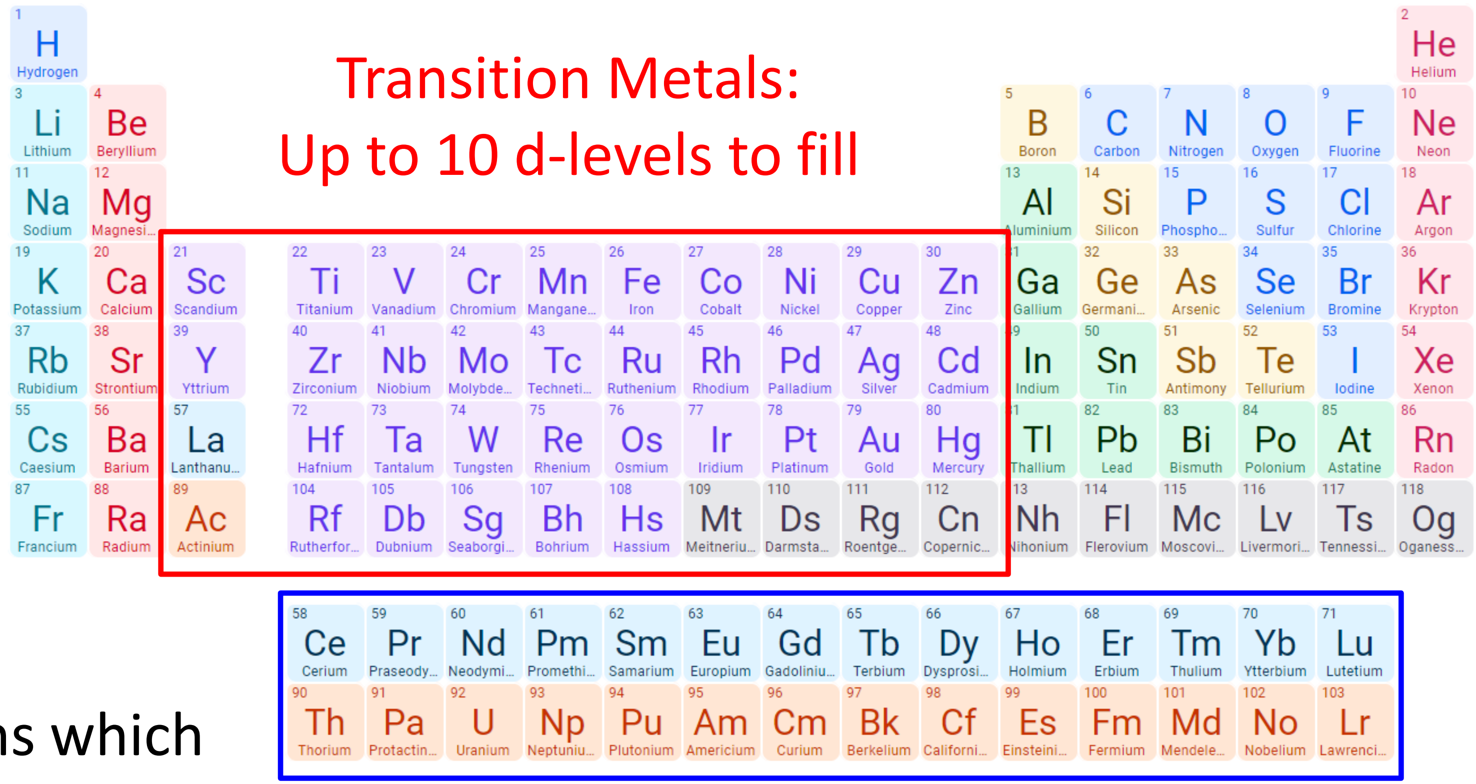
$\gamma = 1.913$
(Gyromagnetic ratio)

$$\mu_N = \frac{e\hbar}{2m_n}$$

(Nuclear magneton)

- μ_n can interact with the electrons in a material via magnetic potentials
- **Scattering from these potentials can be comparable in strength to nuclear scattering**

Magnetic Materials



Lanthanides/Rare Earths and Actinides:
Up to 14 f-levels to fill

- **Magnetic moments** arise on atoms which have **unpaired electrons** in partially filled electronic orbitals

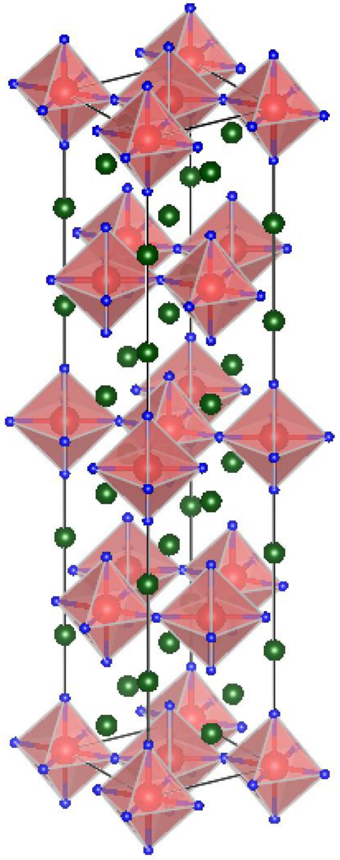
- Most common families of magnetic materials tend to be based on elements with partially filled d- or f-shells (e.g. **transition metals** or **rare earth/lanthanides**)

Magnetic Materials

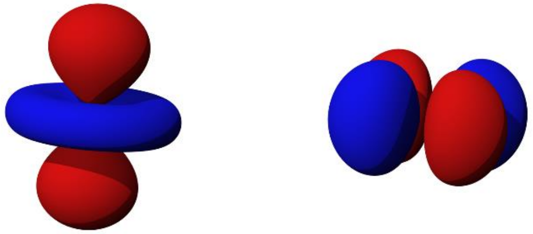
- Size of magnetic moments is determined by Hund's Rules:

e.g. Mn^{2+} ($3d^5$)

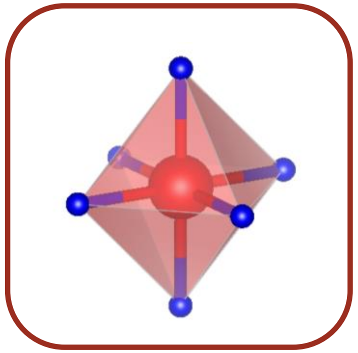
$\boxed{S = 5/2}$

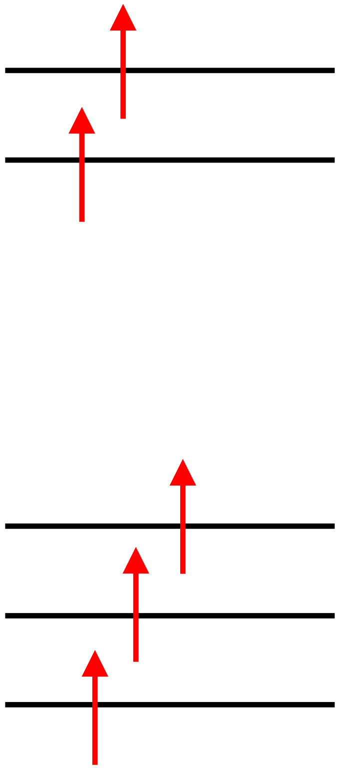


$3z^2 - r^2$ $x^2 - y^2$

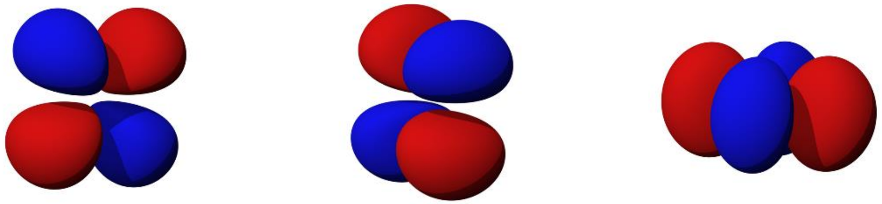


e_g orbitals

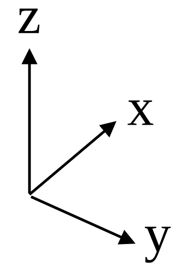




zx yz xy



t_{2g} orbitals

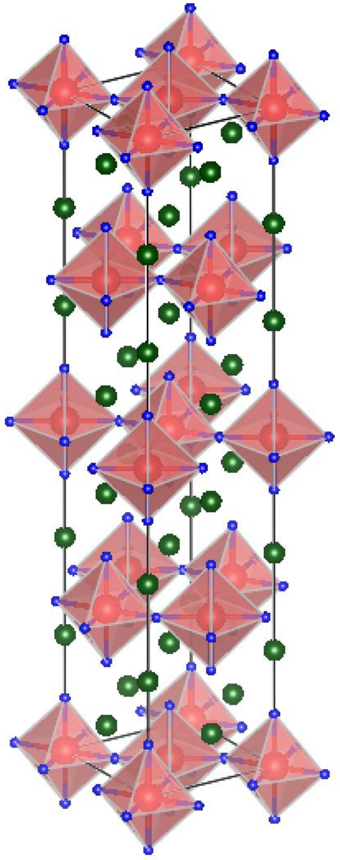


Magnetic Materials

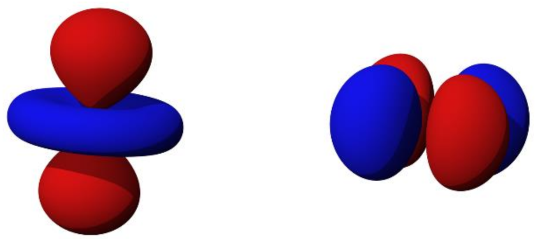
- Size of magnetic moments is determined by Hund's Rules:

e.g. Cu^{2+} ($3d^9$)

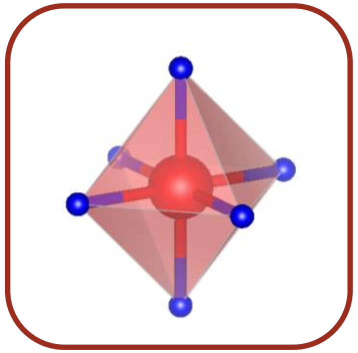
$\boxed{S = 1/2}$

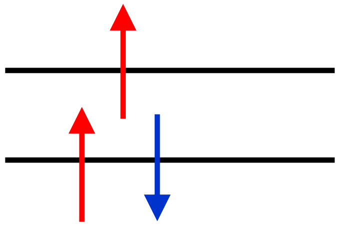


$3z^2 - r^2$ $\quad$ $x^2 - y^2$

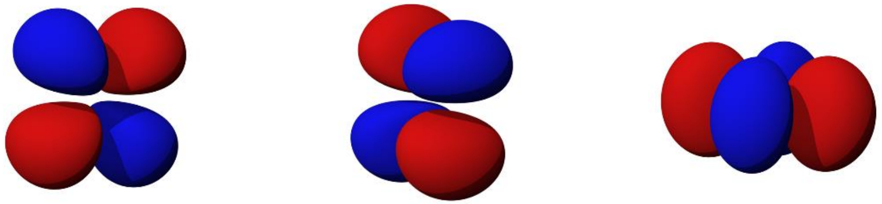


e_g orbitals





zx $\quad$ yz $\quad$ xy



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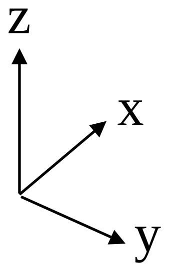
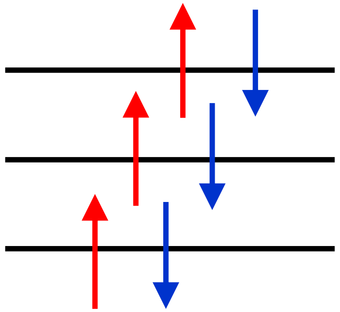
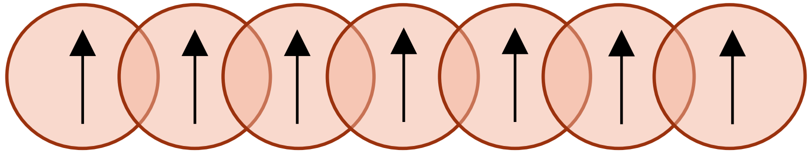


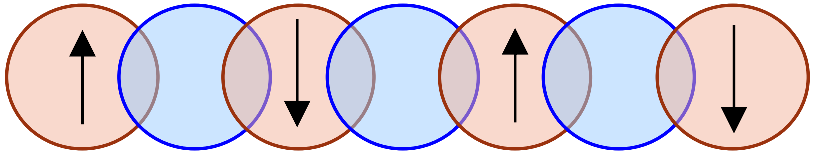


Magnetic Interactions

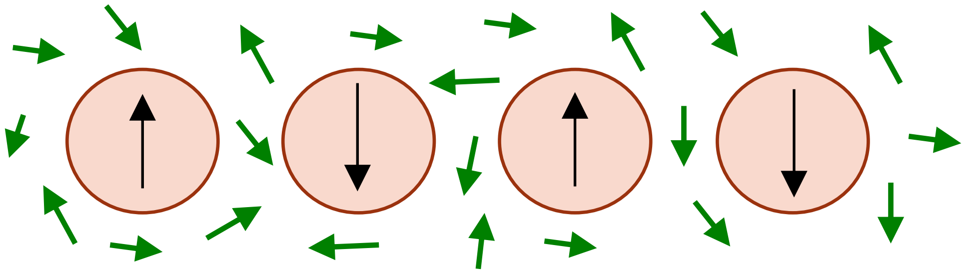
- Direct exchange:



- Superexchange:



- RKKY exchange:

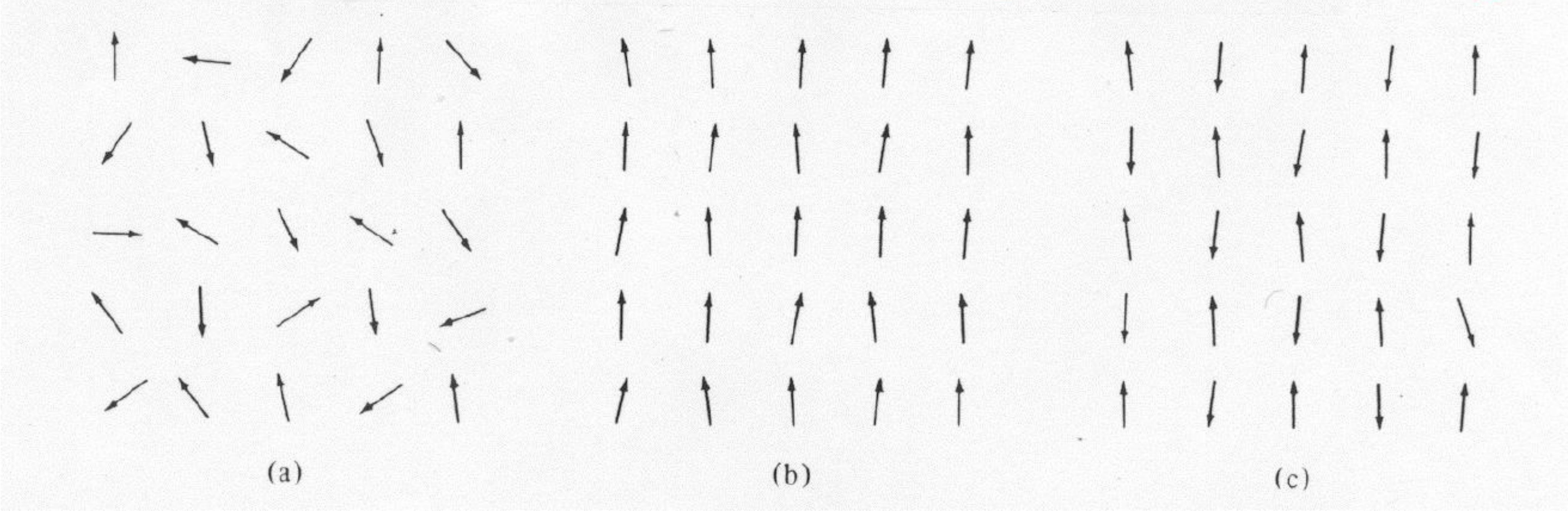


Describe interactions by a magnetic Hamiltonian:

e.g. $H = J \sum_{i,j} S_i \cdot S_j$

(exchange parameter)

Magnetic Order



Paramagnet

($T > T_C$)

Ferromagnet

($T < T_C$)

Antiferromagnet

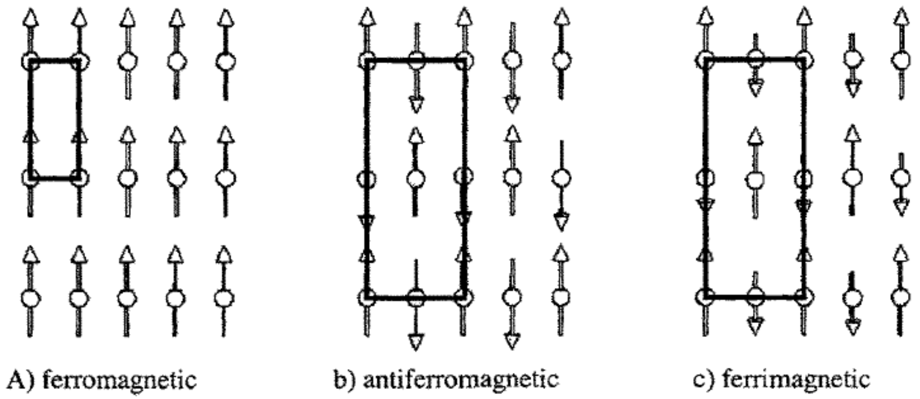
($T < T_N$)

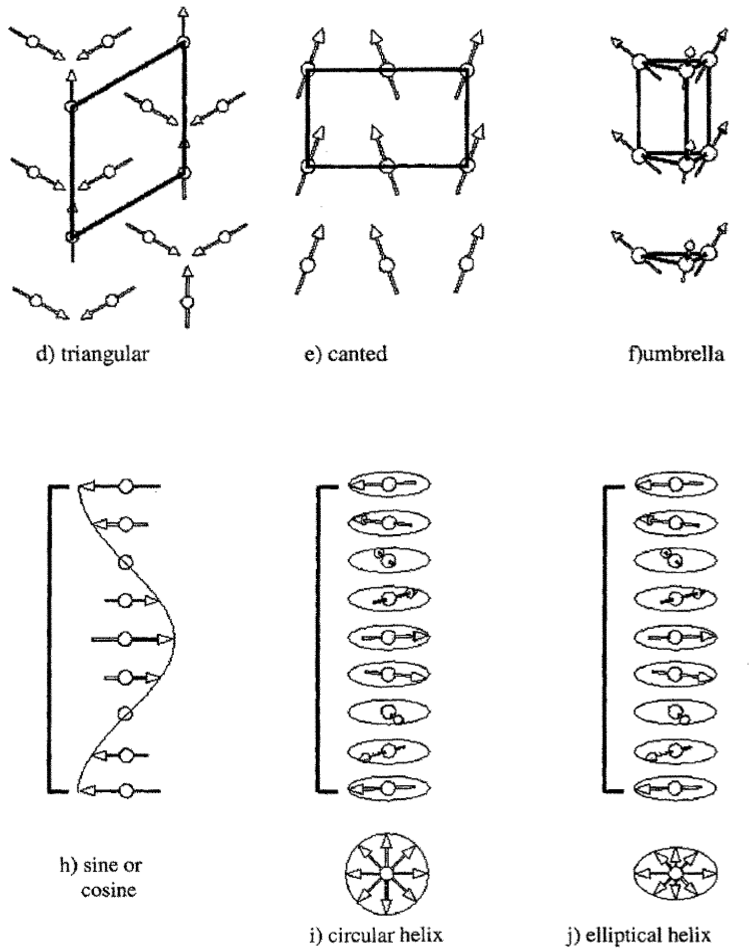
Magnetic Structures

... or more complex

- Magnetically ordered structure that develops in a material depends on nature of underlying magnetic interactions

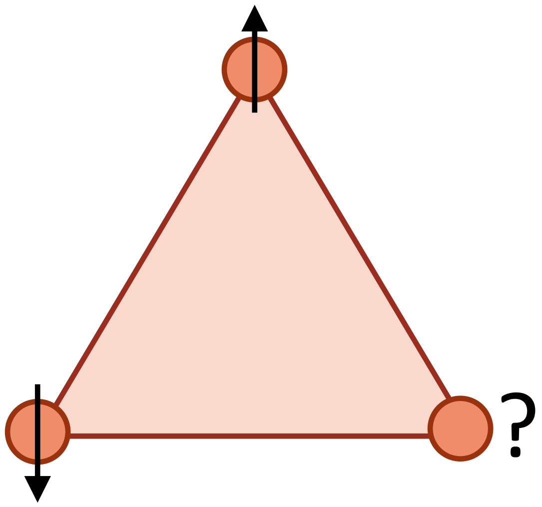
Structures can be relatively simple…

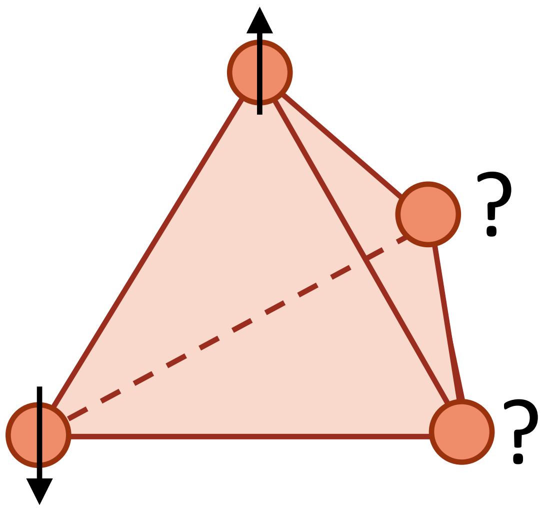


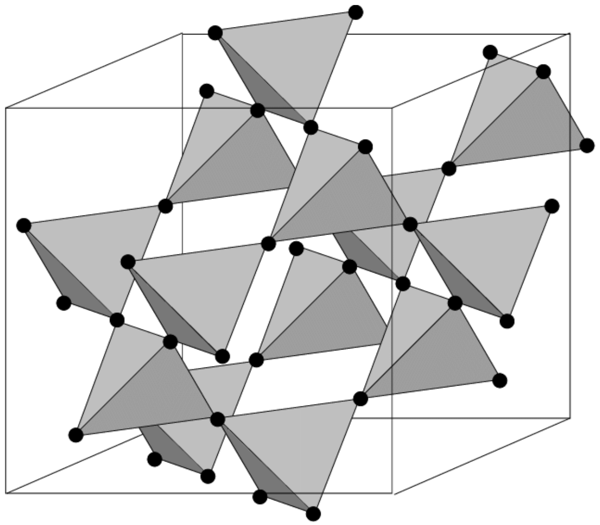


Geometric Frustration

- We can also try to design magnetic materials which don’t order at all:







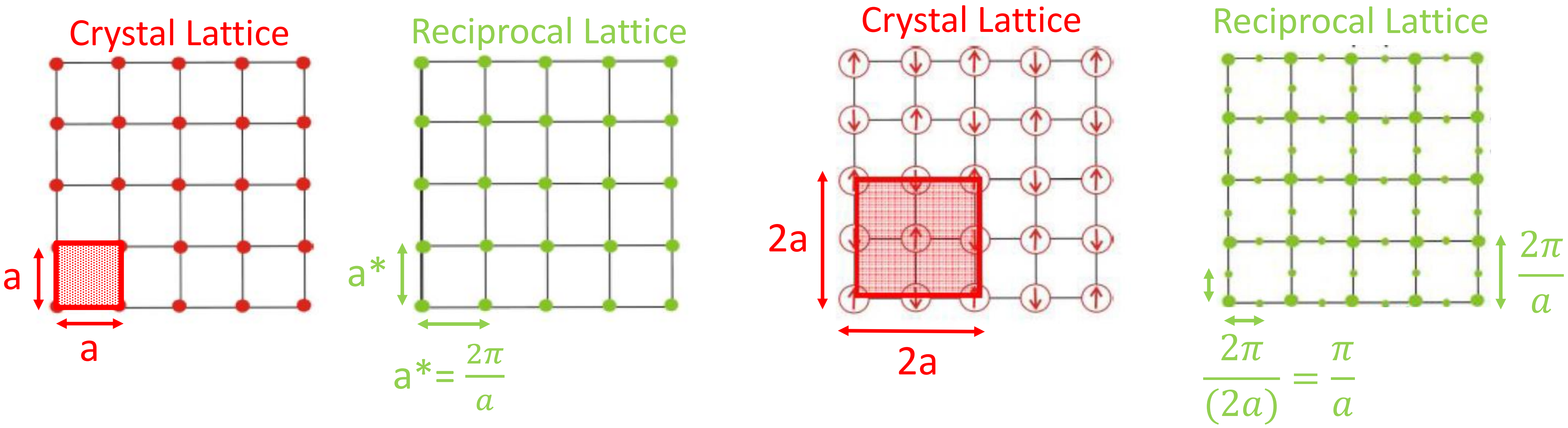
The Pyrochlore Lattice

- Geometrically frustrated magnets can display exotic quantum ground states at low temperatures, e.g. **quantum spin liquids**, **spin ices**, **spin glasses**…

Scattering from a Magnetically Ordered Crystal

- How can we detect magnetic order in a neutron scattering experiment?

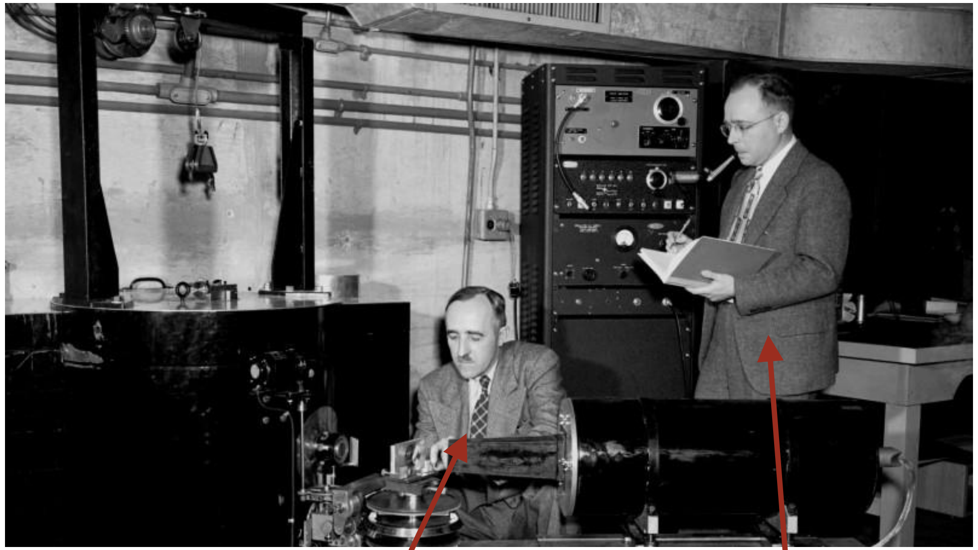
Paramagnetic State ($T > T_N$) **Antiferromagnetic State ($T < T_N$)**



- Development of AF order increases size of unit cell → **new magnetic Bragg peaks appear**

First Observation: Magnetic Neutron Scattering from MnO

- Early neutron diffraction experiments at the ORNL X-10 Graphite Reactor:



Ernest Wollan   Clifford Shull

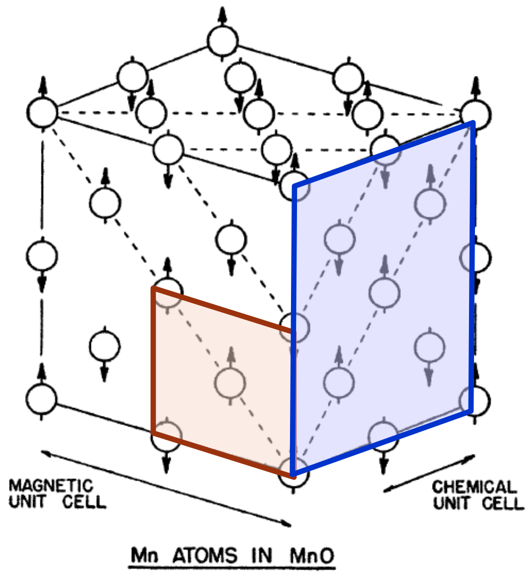


FIG. 5. Antiferromagnetic structure existing in MnO below its Curie temperature of 120°K. The magnetic unit cell has twice the linear dimensions of the chemical unit cell. Only Mn ions are shown in the diagram.

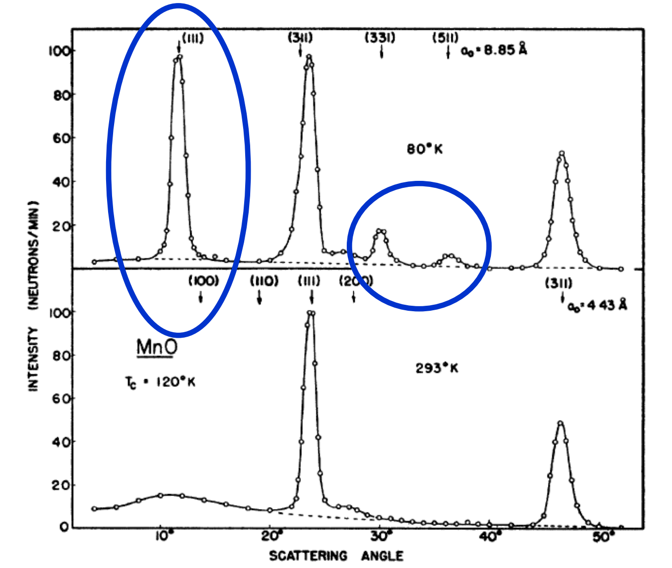


FIG. 4. Neutron diffraction patterns for MnO taken at liquid nitrogen and room temperatures. The patterns have been corrected for the various forms of extraneous, diffuse scattering mentioned in the text. Four extra antiferromagnetic reflections are to be noticed in the low temperature pattern.

First direct evidence of antiferromagnetism

Shull, Strauser, and Wollan, Phys. Rev. ***83****, 333 (1951)*

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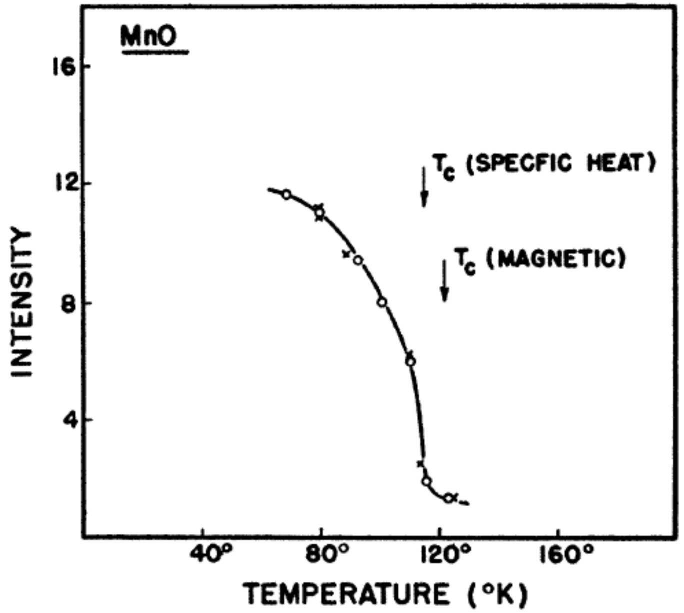
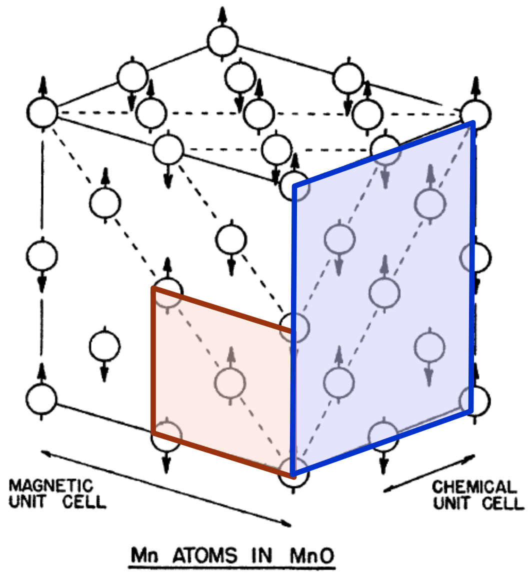


FIG. 7. Temperature dependence of magnetic intensity for MnO. The Curie temperatures suggested by specific heat and magnetic susceptibility data are shown.



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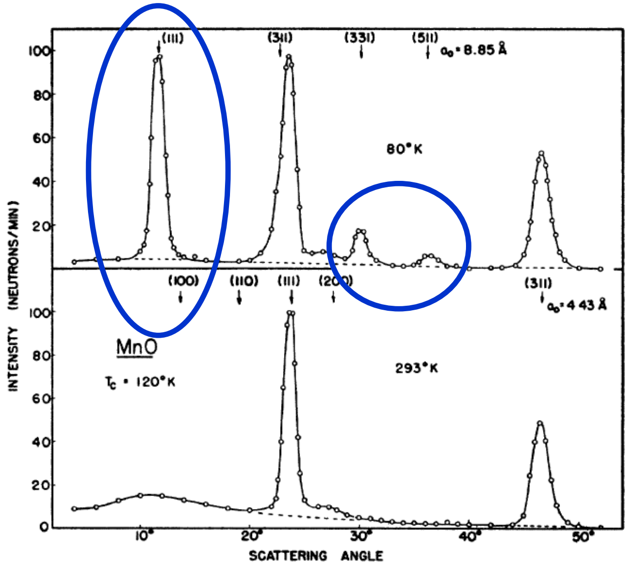

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First direct evidence of antiferromagnetism

Peak intensity ∝ staggered magnetization

Shull, Strauser, and Wollan, Phys. Rev. ***83****, 333 (1951)*

Magnetic Scattering Cross Section

- What fraction of neutrons will scatter off a sample with a particular change in energy and momentum?
- Change in momentum: $\vec{Q} = \vec{k} - \vec{k}'$
- Change in energy: $\Delta E = \hbar\omega = \frac{\hbar^2 k^2}{2m} - \frac{\hbar^2 k'^2}{2m}$
- Apply Fermi's Golden Rule (1st order perturbation theory):

$$\frac{d^2\sigma}{d\Omega\, dE'}_{k,\sigma,\lambda \to k',\sigma',\lambda'} = \boxed{\left(\frac{m}{2\pi\hbar^2}\right)^2 \frac{k'}{k}} \boxed{|\langle k'\ \sigma'\ \lambda' | V_m | k\ \sigma\ \lambda\rangle|^2} \boxed{\delta\left(E_\lambda - E_{\lambda'} + \hbar\omega\right)}$$

(Kinematics) (Interaction Term) (Energy Conservation)

The Magnetic Potential

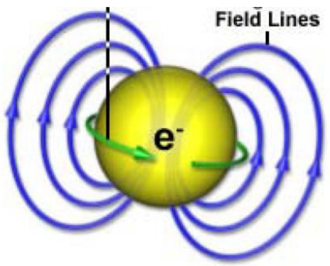
- In order to evaluate the matrix element in the interaction term, we need to determine the magnetic potential produced by all of the unpaired electrons in the material:

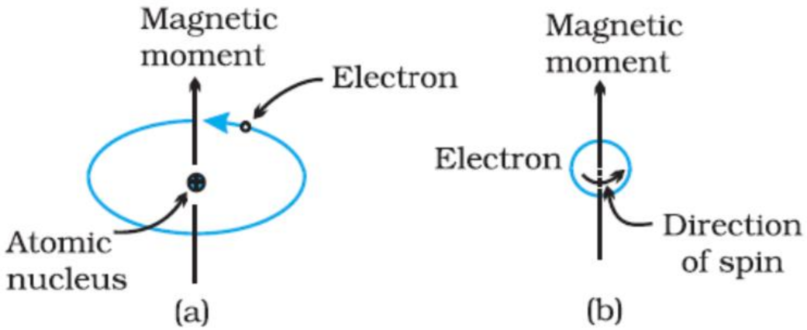
$$\boxed{V_m = \vec{\mu}_n \cdot \vec{B}}$$

(Magnetic Potential)

(Magnetic Dipole Moment of Neutron)

(Magnetic Field Produced by Unpaired Electrons)





Must consider:

B_l = Magnetic field from **orbital motion** of an electron

B_s = Magnetic field from **spin** of an electron

Magnetic Scattering Cross Section

- Evaluating the interaction term $|\langle k'\ \sigma'\ \lambda'|V_m|k\ \sigma\ \lambda\rangle|^2$ can be quite complicated.
- Jumping to the final result:

$$\frac{d^2\sigma}{d\Omega\, dE'} = \boxed{\frac{(\gamma r_0)^2}{2\pi\hbar}} \frac{k'}{k} N \boxed{\left[\frac{1}{2} gF\left(\vec{Q}\right)\right]^2} \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right)$$

$$\times \sum_l e^{i\vec{Q}\cdot\vec{l}} \int \left\langle e^{-i\vec{Q}\cdot\vec{u}_0(0)} e^{i\vec{Q}\cdot\vec{u}_l(t)} \right\rangle \left\langle S_0^\alpha(0)\, S_l^\beta(t) \right\rangle e^{-i\omega t} dt$$

Key features:

1. From constants – magnetic scattering comparable in strength to nuclear scattering ($\sim {r_0}^2$)
2. Proportional to square of **magnetic form factor**, $F\left(\vec{Q}\right)^2$

Magnetic Scattering Cross Section

- Evaluating the interaction term $|\langle k' \ \sigma' \ \lambda' | V_m | k \ \sigma \ \lambda \rangle|^2$ can be quite complicated.
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Key features:

3. **Polarization factor** – describes dependence on spin direction. Term vanishes if components of spin are parallel to scattering vector $\vec{Q} \longrightarrow$ only sensitive to $S \perp \vec{Q}$

Magnetic Scattering Cross Section

- Evaluating the interaction term $|\langle k' \; \sigma' \; \lambda' | V_m | k \; \sigma \; \lambda \rangle|^2$ can be quite complicated.
- Jumping to the final result:

$$\frac{d^2\sigma}{d\Omega \, dE'} = \boxed{\frac{(\gamma r_0)^2}{2\pi\hbar}} \frac{k'}{k} N \boxed{\left[\frac{1}{2} g F\left(\vec{Q}\right)\right]^2} \boxed{\sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right)}$$

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Key features:

4. **Dynamic spin pair correlation function** – measures correlation between spin α at origin and t = 0 and spin β at position l and time t. The Fourier transform of this term is the **dynamic structure factor**, $\boldsymbol{S(\vec{Q}, \omega)}$

Magnetic Form Factor

- $F\left(\vec{Q}\right)$ = Fourier transform of the spin distribution in real space

$$\boxed{F\left(\vec{Q}\right) = \int S(\vec{r})\, e^{i\vec{Q}\cdot\vec{r}} d^3r}$$

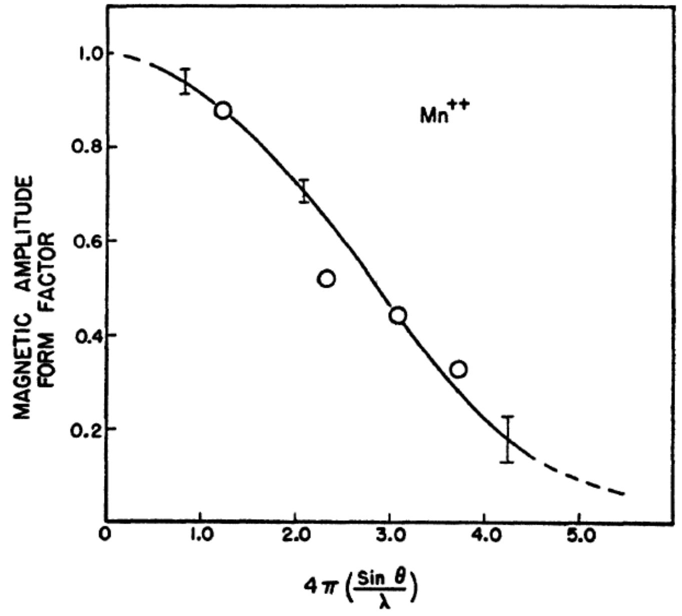
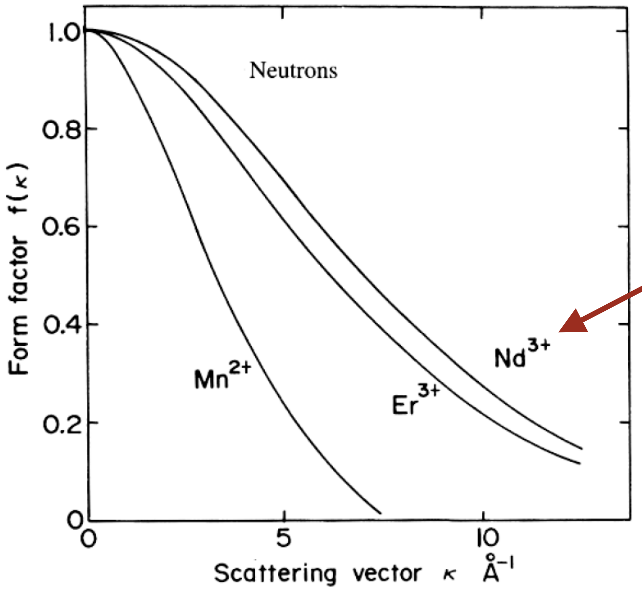


FIG. 2. Magnetic amplitude form factor for Mn^{++} ions. The curve is that obtained from paramagnetic diffuse scattering with estimated error as shown. The points represent values of the form factor obtained from the low temperature antiferromagnetic reflections of MnO.



$F\left(\vec{Q}\right)$ decreases faster as wavefunctions become more spatially extended

- Analogous to chemical form factor for x-ray scattering
- Typically drops off monotonically as $\vec{Q}$ increases

Shull, Strauser, and Wollan, Phys. Rev. ***83****, 333 (1951)*

Elastic Magnetic Scattering

- For elastic scattering (i.e. diffraction), we have: $\Delta E = \hbar\omega = \frac{\hbar^2 k^2}{2m} - \frac{\hbar^2 {k'}^2}{2m} = 0$
- What we measure is the **time-independent** structure factor, S($\vec{Q}$)

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \ \frac{k'}{k} \ N \left[\frac{1}{2} gF\left(\vec{Q}\right)\right]^2 e^{-2W} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) \times \sum_l e^{i\vec{Q}\cdot\vec{l}} \int \langle S_0^\alpha \rangle \left\langle S_l^\beta \right\rangle$$

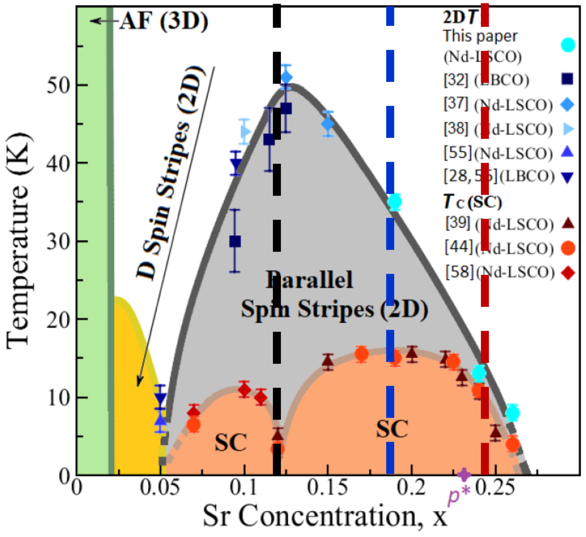
Debye-Waller Effect

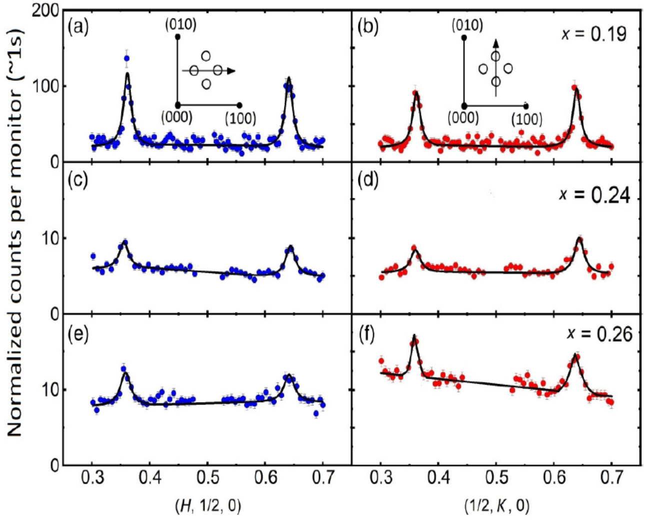
Polarization Factor:
Only sensitive to $S \perp \vec{Q}$

Add up spins with a phase factor of $e^{i\vec{Q}\cdot\vec{l}}$

Elastic Magnetic Scattering: Examples

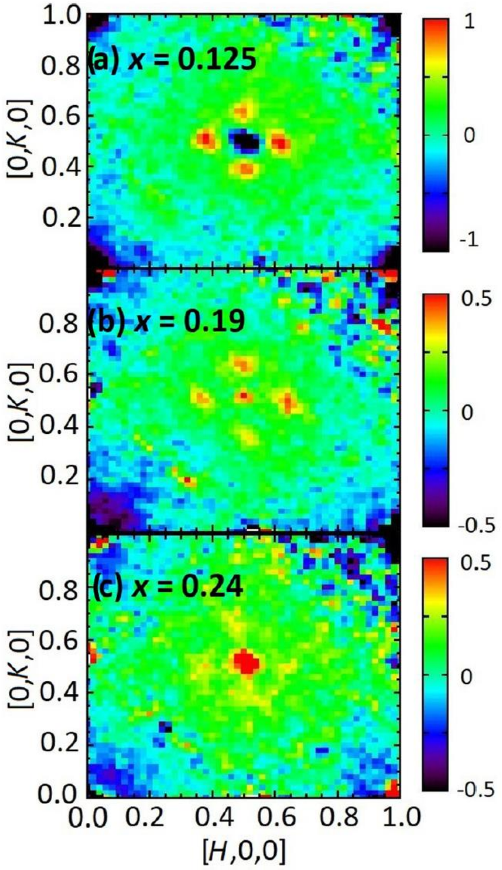
- **High temperature superconductors**
- **e.g. $Nd_{0.4}La_{1.6-x}Sr_xCuO_4$ Single Crystal**
- Measured on SEQUOIA at SNS, and HB3 at HFIR

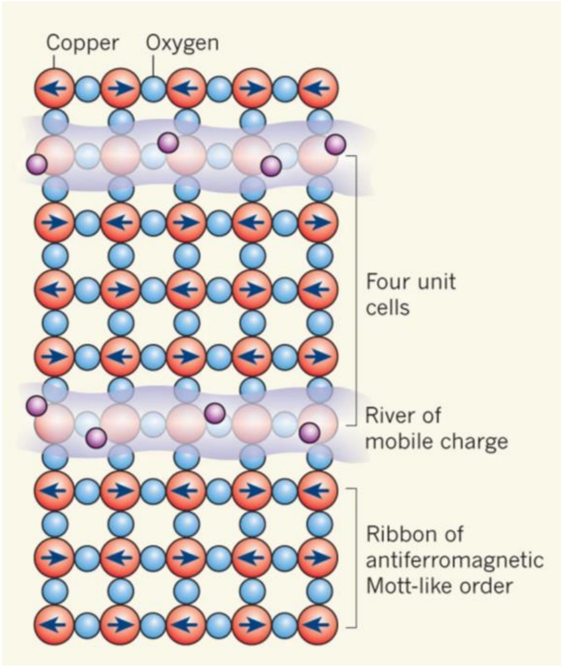




Q. Ma et al, Phys. Rev. Res. **3**, 023151 (2021)

T = 5K – 60K





*K. A. Moler, Nature **468**, 643 (2010)*

- Magnetic scattering is **incommensurate**, arising at $(0.5 \pm \delta, 0.5, \mathrm{L})$ and $(0.5, 0.5 \pm \delta, \mathrm{L})$ due to stripe order

Elastic Magnetic Scattering: Examples

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- **e.g. $Nd_{0.4}La_{1.6-x}Sr_xCuO_4$ Single Crystal**
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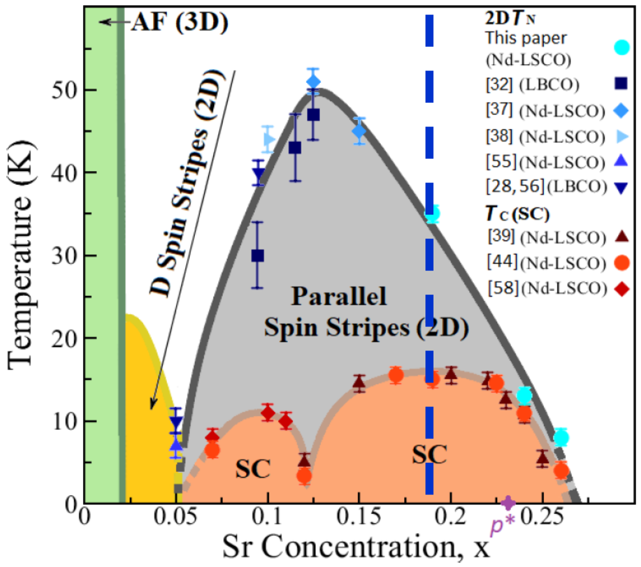
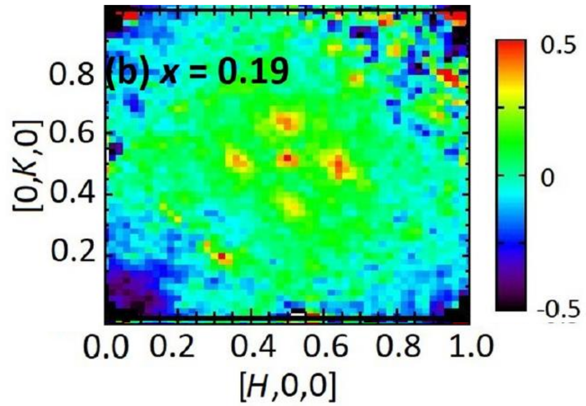
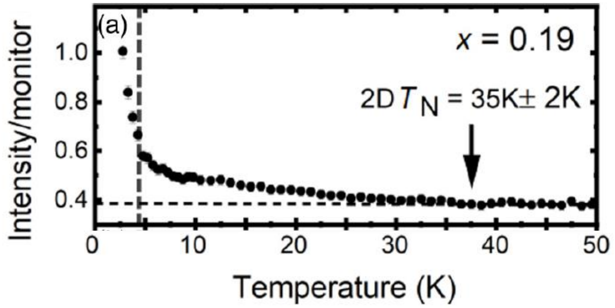






Q. Ma et al, Phys. Rev. Res. ***3****, 023151 (2021)*

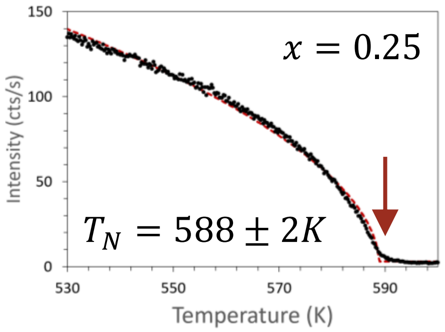
- Above T = 5K: **Rods** of diffuse scattering along the L-direction, indicates shorter-range **quasi-2D** magnetic correlations
- Below T = 5K: Well-defined **Bragg peaks**, indicates **3D long-range** magnetic order

$\xi \propto \frac{1}{Q}$ = correlation length

- More conventionally:

$$I \propto M^2 = {M_0}^2 \left(1 - \frac{T}{T_C}\right)^{2\beta}$$

$Ca_2Fe_{2-x}Mn_xO_5$



J. Greedan et al (2023)

Inelastic Magnetic Scattering

- For inelastic scattering (i.e. spectroscopy), we have: $\Delta E = \hbar\omega = \frac{\hbar^2 k^2}{2m} - \frac{\hbar^2 k'^2}{2m} \neq 0$
- This implies that $|\vec{k}| \neq |\vec{k}'| \rightarrow$ change in both $\vec{Q}$ and ω
- What we measure is the **dynamical structure factor** $S(\vec{Q}, \omega)$

- Key points:
- Study *dynamic* magnetic moments (on time scales of 10^{-9} to 10^{-12} sec)

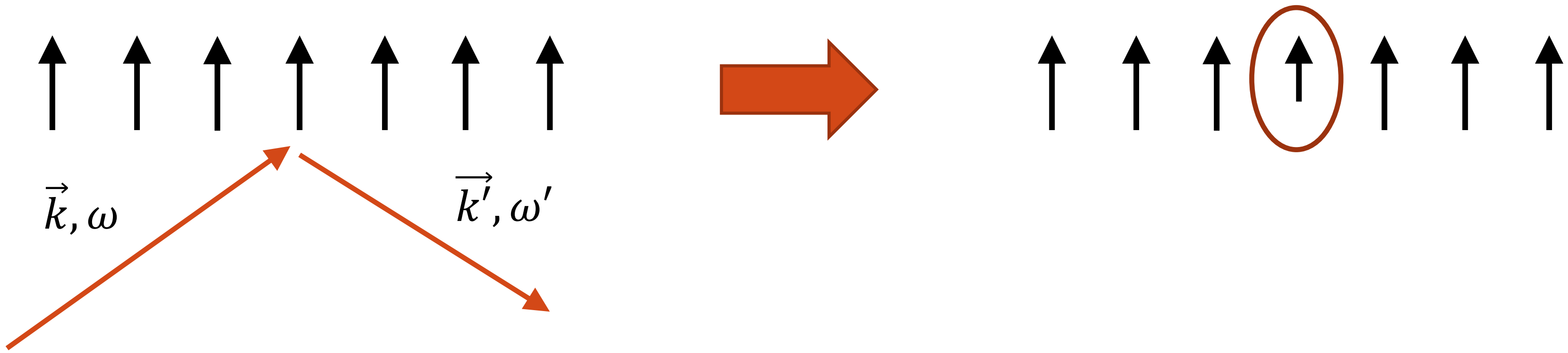
Bose (Temperature) Factor Imaginary part of dynamic susceptibility

- $S(\vec{Q}, \omega) = \frac{1}{1-e^{-\beta\hbar\omega}} \frac{\chi''(\vec{Q},\omega)}{\pi (g\mu_B)^2} = \boxed{n(\omega)}\, \boxed{\chi''(\vec{Q}, \omega)}$ *(Fluctuation-Dissipation Theorem)*
- Intensity integrated over all $\vec{Q}, \omega$ is constant: $\int d\omega \int_{BZ} d\vec{Q}\, S(\vec{Q}, \omega) \sim S(S+1)$

(Total Moment Sum Rule)

Inelastic Magnetic Scattering: Spin Waves

- When a neutron scatters off a sample it can create or destroy an excitation
- If sample is magnetically ordered (e.g. a FM spin chain), the incident neutron can create a spin "defect" which is distributed over all possible sites
- We call this collective excitation a **spin wave** or **magnon**



Spins are coupled through magnetic Hamiltonian: $H = J \sum_{i,j} S_i \cdot S_j$

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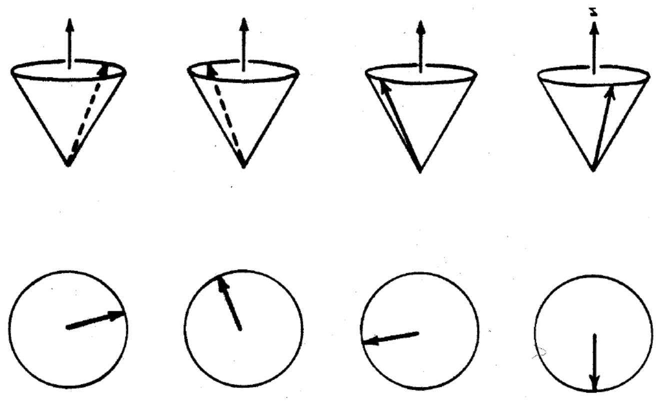

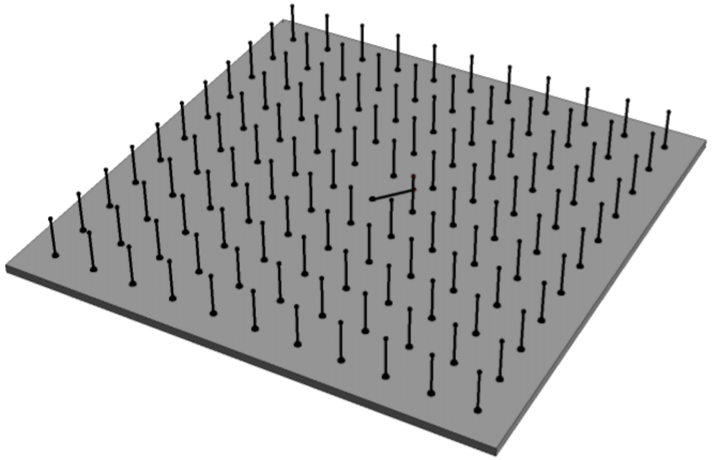

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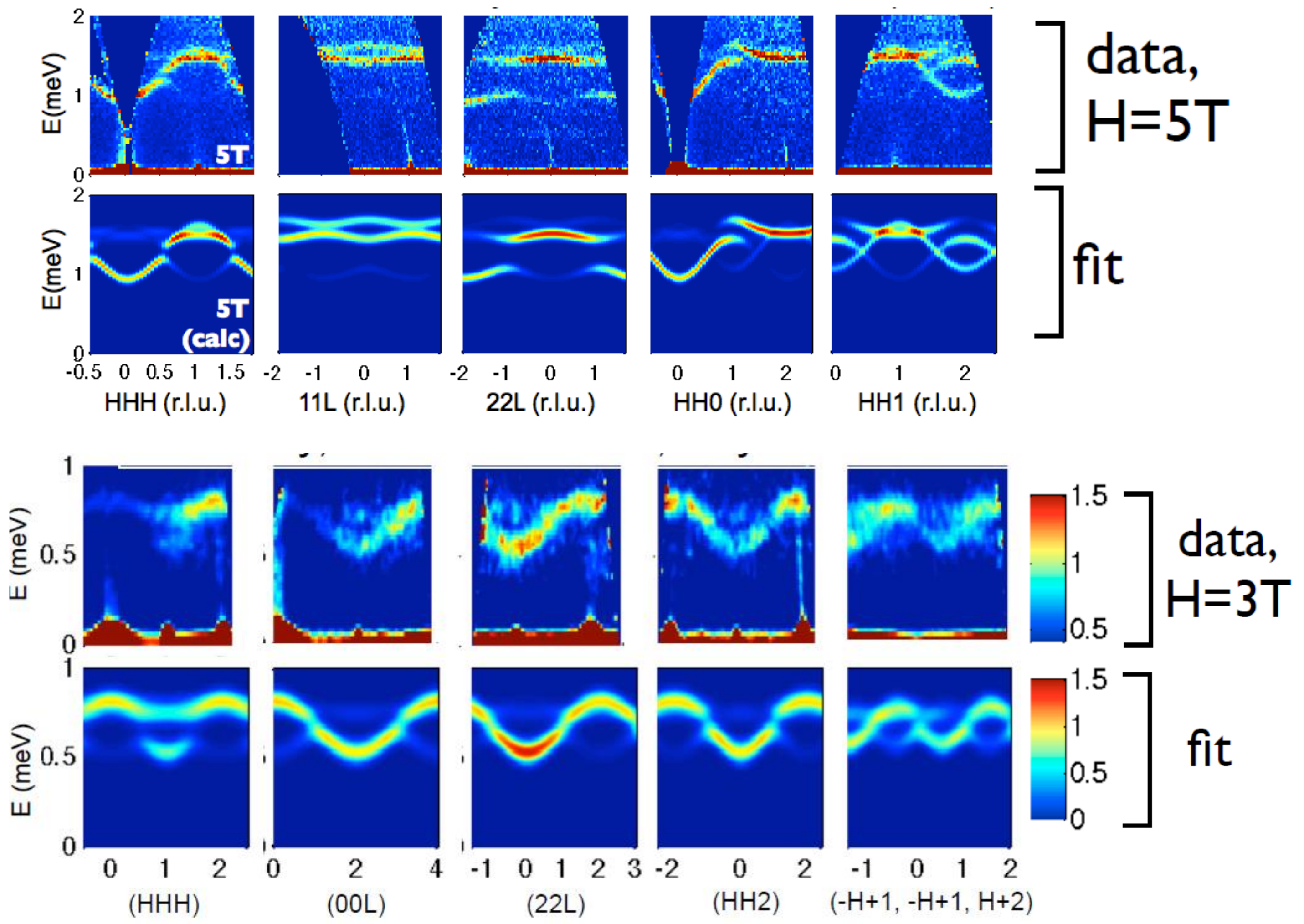
Inelastic Magnetic Scattering: Examples



- **Frustrated magnetism in pyrochlores**
- **$Yb_2Ti_2O_7$** (top) and **$Er_2Ti_2O_7$** (bottom) Single Crystals
- Measured on DCS at NIST
- Fit spin wave dispersion to theoretical model and extract detailed exchange parameters (J_1, J_2, J_3, J_4)
- Magnetic interactions explain low temperature magnetic ground states

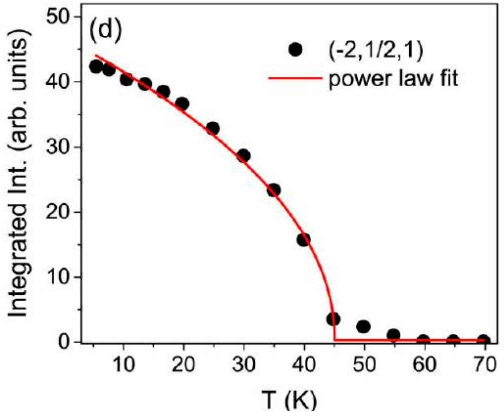
K. A. Ross et al, Phys. Rev. X **1**, 021002 (2011)

L. Savary et al, Phys. Rev. Lett. 109, 167201 (2012)

How can we distinguish magnetic scattering?

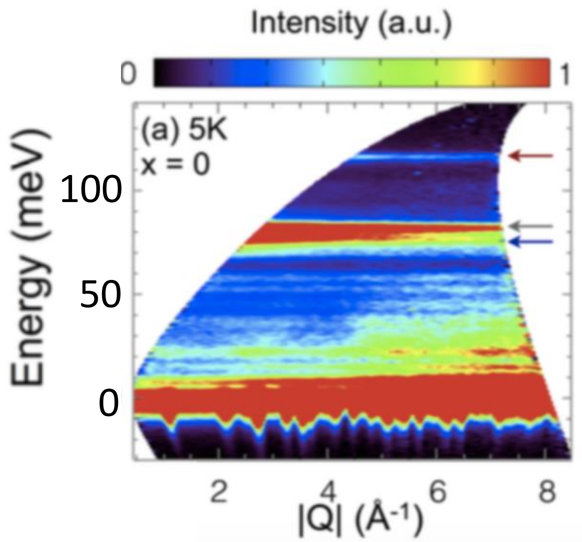
(1) Temperature dependence:

- Magnetic scattering **decreases** with increasing T (disappears at T > T_C)
- Phonon scattering **increases** with increasing T ($\propto$ thermal population)



(2) Momentum dependence:

- Magnetic scattering **decreases** with increasing |Q| ($\propto |F(Q)|^2$)
- Phonon scattering **increases** with increasing |Q| ($\propto |e \cdot Q|^2$)

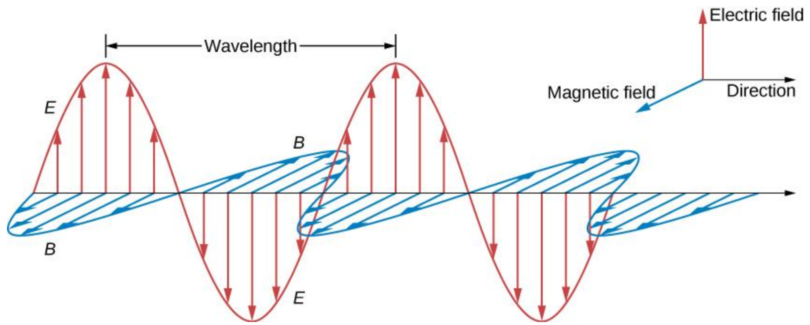


(3) Polarization dependence (with polarized beam):

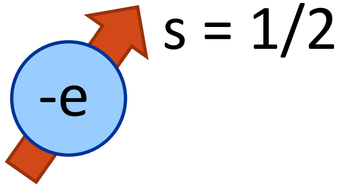
- Magnetic scattering **mostly spin flip**
- Nuclear scattering **mostly non-spin flip**

More on polarized neutrons in experiments N2 (HYSPEC) and N20 (MAGREF), and in Barry Winn's lecture on Wednesday

Magnetic Scattering with X-rays



- X-rays carry no magnetic moment
- Primary interaction with matter: **E-field** of x-ray + **charge** of electrons
- Also interacts through: **B-field** of x-ray + **spin** of electrons



- Unlike neutrons:

1. Magnetic scattering is MUCH weaker than charge scattering

Amplitude ratio: $$\frac{A(magnetic)}{A(charge)} = \frac{\hbar\omega}{mc^2}$$ (*for **single** electron*)

At Ei ~5 keV:
Amplitude ratio ~10^{-2}
Intensity ratio ~10^{-4}

2. X-ray photon energies (**~0.5 to 50 keV**) are orders of magnitude larger than typical energy scales for magnetic excitations (**~0.5 to 500 meV**)

(More on magnetic x-ray scattering in lectures and experiments from Week 1 at ANL)

First Observation: Magnetic X-ray Scattering from NiO

NiO: Antiferromagnet (T_N ~ 250 °C)

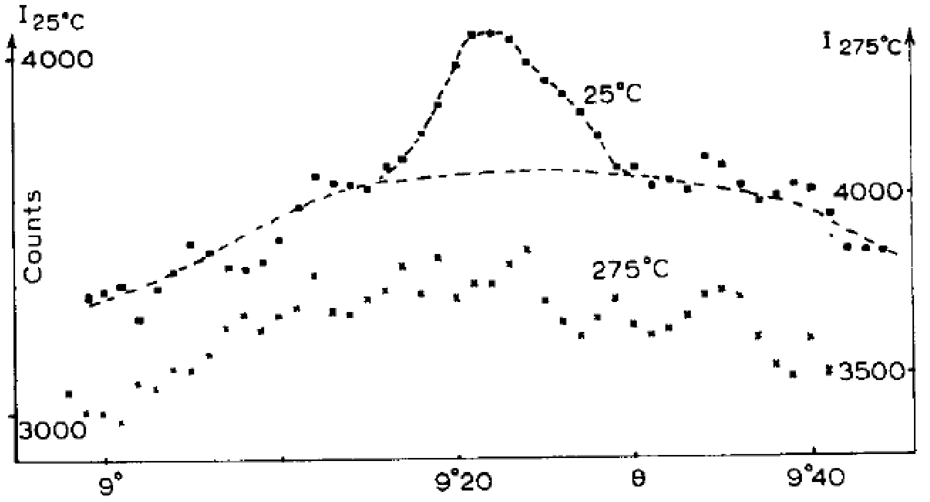
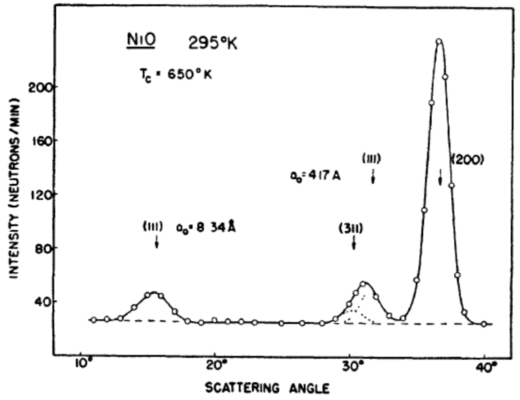


Fig. 1. Intensity $I_t(\theta)$ near the $(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ position at $t = 25°$ C and 275° C in counts/225 min. The hump which cover the interval could be due to some impurity.

De Bergevin and Brunel, Phys. Lett. ***39A****, 141 (1972)*

- **NON-RESONANT magnetic x-ray scattering**
- Lab-based experiment carried out using x-ray tube source (Cu Kα, λ = 1.54 Å)
- Hard!
- Counting time: 3 days/scan
(~2 cts/min signal on ~18 cts/min bkgd)
- Compare to magnetic neutron scattering:

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



To the Synchrotron: Magnetic X-ray Scattering from Ho

Ho: Incommensurate spiral antiferromagnet (T_N ~ 131 K)

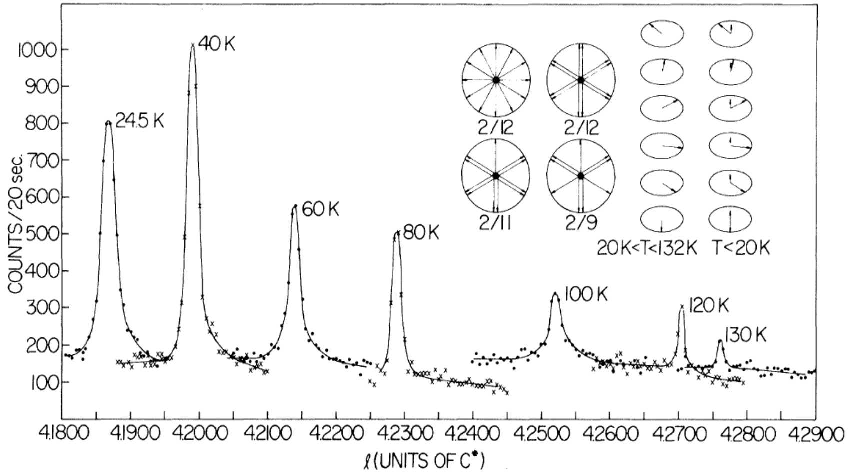


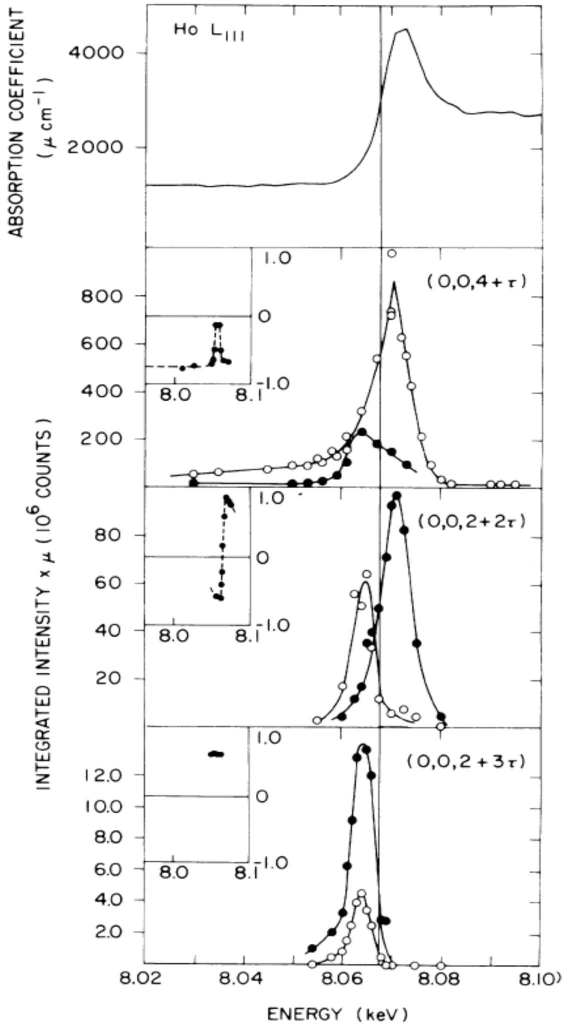
FIG. 1. Temperature dependence of the Ho(004)$^+$ magnetic satellite taken with synchrotron radiation (lines drawn to guide the eye). Inset: Right, schematic representation of the magnetic structure of Ho (after Koehler[9]). Left, projections of the magnetic unit cell for different spin-slip structures. For simplicity the doublet has been drawn as two parallel spins.

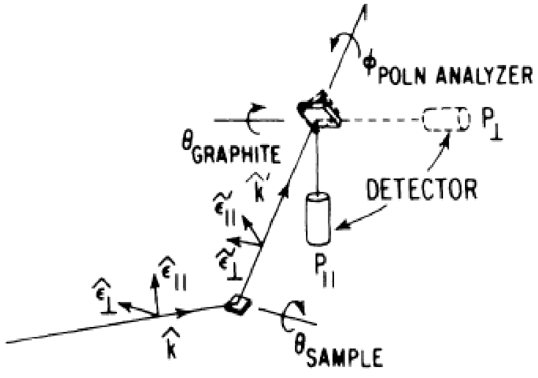
D. Gibbs et al, Phys. Rev. Lett. ***55****, 234 (1985)*

- **NON-RESONANT magnetic x-ray scattering**
- Synchrotron-based experiment (SSRL)
- Higher flux and higher momentum resolution

- Compare to magnetic neutron scattering:
- X-ray: 25 cts/s on 10 cts/s, FWHM = 0.001 $Å^{-1}$
- Neutron: 50 cts/s on 0.1 cts/s, FWHM = 0.005 $Å^{-1}$

On Resonance: Magnetic X-ray Scattering from Ho





Gibbs et al, Phys. Rev. Lett. ***61****, 1241 (1988)*

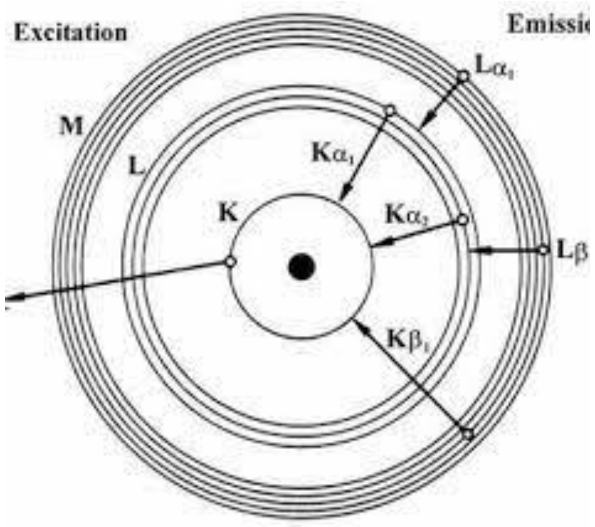
- **RESONANT MAGNETIC X-RAY SCATTERING**
- First predicted by M. Blume (1985)
- Synchrotron-based experiment (NSLS, CHESS)
- Tune incident energy to Ho L_3-edge (E_i = 8.067 keV, λ = 1.54 Å)
- Take advantage of polarized beam and resonant enhancement at absorption edge
- Magnetic peak intensity enhanced by **~50x**!

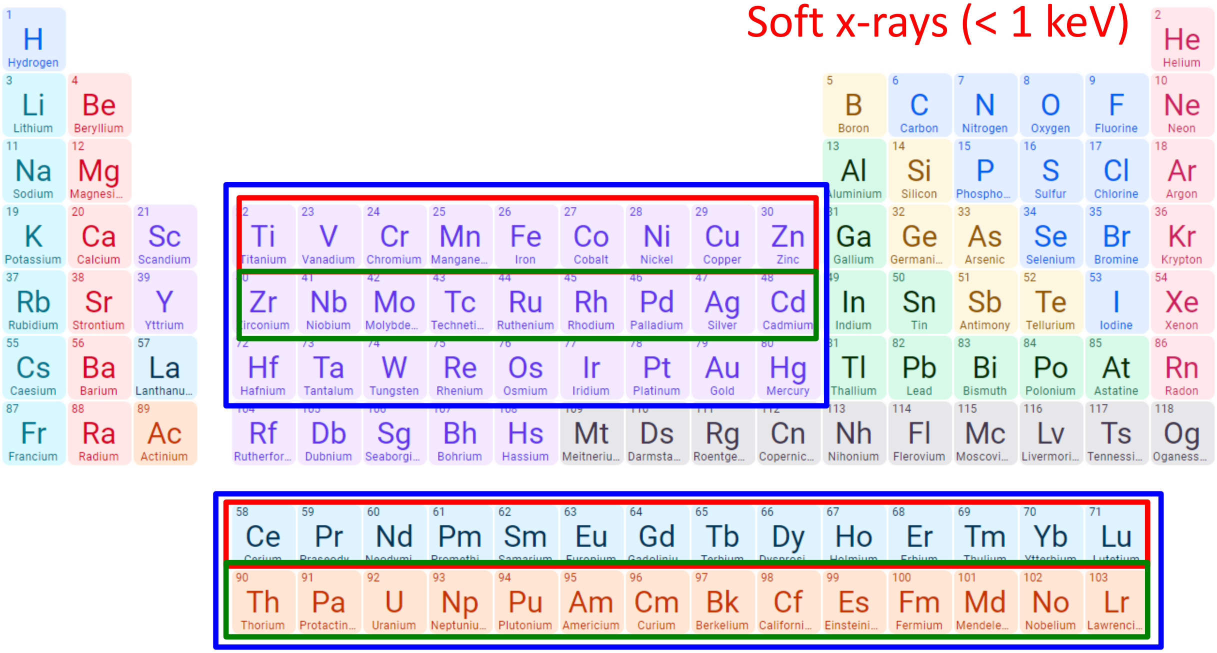
Resonant Magnetic X-ray Scattering

Hard x-rays (> 5 keV)

Tender x-rays (1-5 keV)

Soft x-rays (< 1 keV)





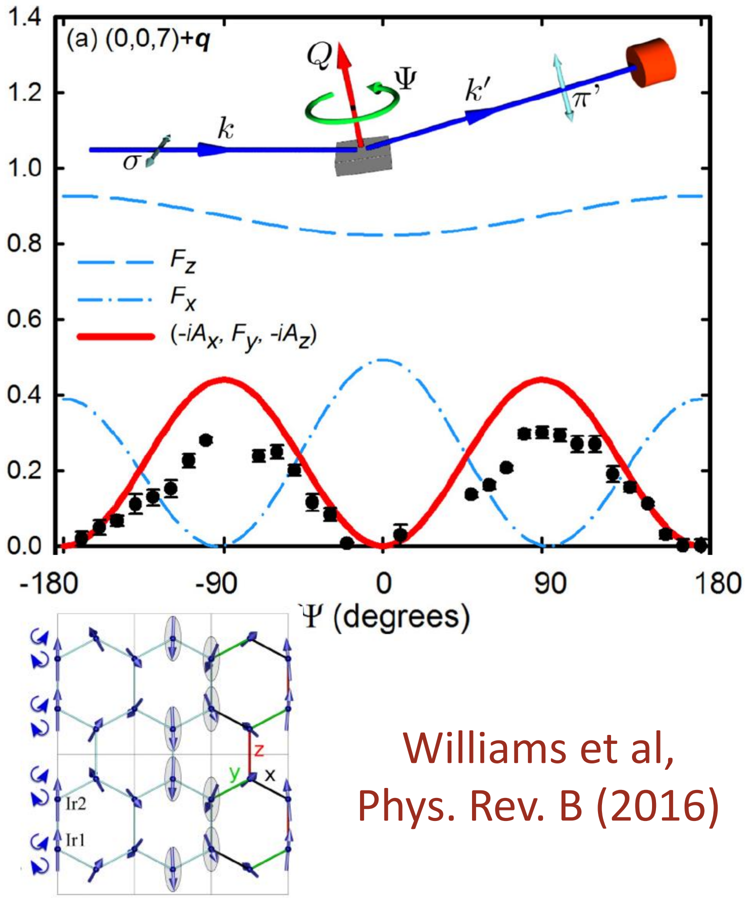
- Scattering tensor for magnetic x-ray scattering:

$$F_j(E) = \sigma^{(0)}(E)\varepsilon_i \cdot \varepsilon_0{}^* + \sigma^{(1)}(E)\ \varepsilon_i \times \varepsilon_0{}^* \cdot M_j + \sigma^{(2)}(E)\left[\left(\varepsilon_i \cdot M_j\right)\left(\varepsilon_0{}^* \cdot M_j\right) - \frac{1}{3}\varepsilon_i \cdot \varepsilon_0{}^*\right]$$

- Intensity of magnetic Bragg peaks: $I = \left|\sum_j e^{ig\cdot r_j}\,\sigma_j{}^{(1)}(E)\ \varepsilon_i \times \varepsilon_0{}^* \cdot M_j\right|^2$

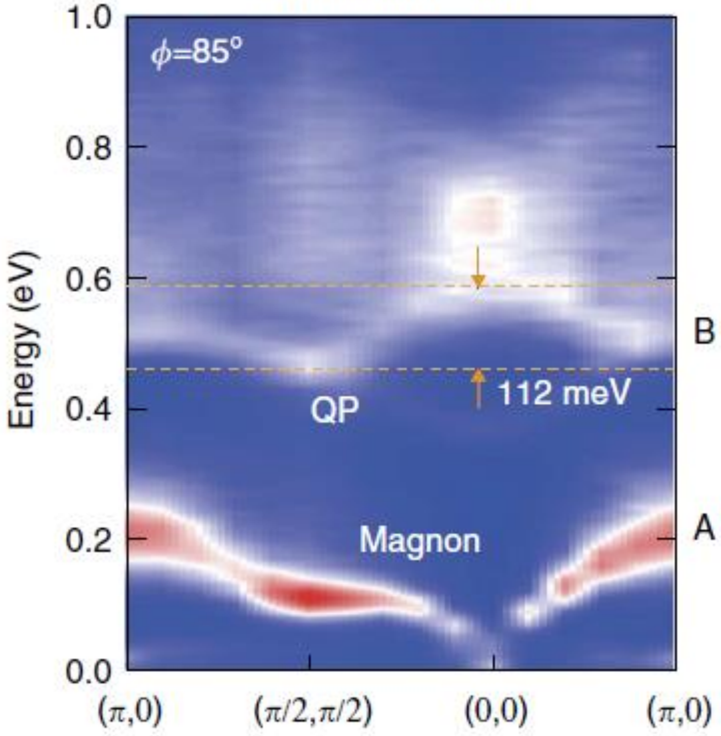
Resonant Magnetic X-ray Scattering: Examples

α-Li_2IrO_3 single crystal
(Kitaev model candidate)



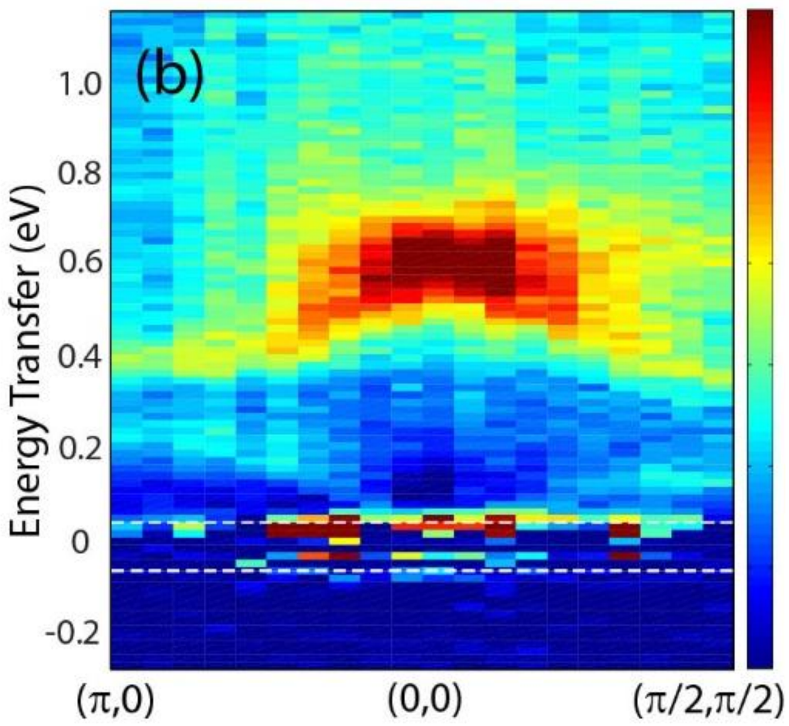
Williams et al,
Phys. Rev. B (2016)

Sr_2IrO_4 single crystal
(spin-orbital Mott insulator)



Kim et al, Nat. Comm. (2014)

Ba_2IrO_4 thin film
(13 nm thickness = 10 ng)



Clancy et al, Phys. Rev. B (2023)

Magnetic X-ray Scattering

Advantages:

- Element (and even orbital) specificity
- Smaller samples (ideal for thin films, high pressure diamond anvil cell experiments)
- Better resolution in momentum

Disadvantages:

- More complicated theory/modeling
- Magnetic scattering much weaker than charge scattering
- Worse resolution in energy
- Restricted momentum transfer (soft x-ray)

X-ray and neutron scattering are highly complementary techniques for the study of magnetic materials

Any Questions?

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