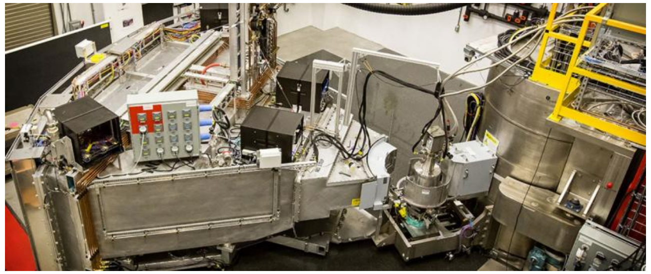
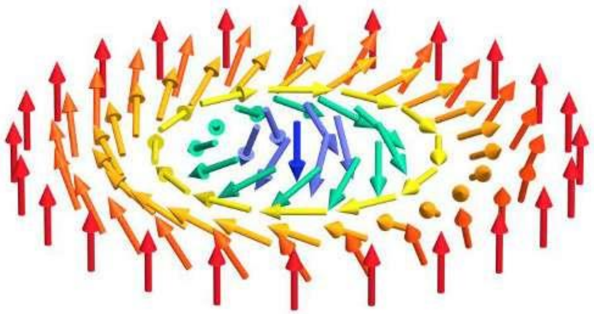
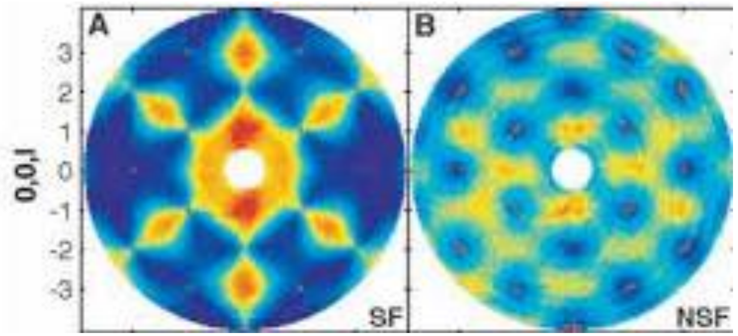
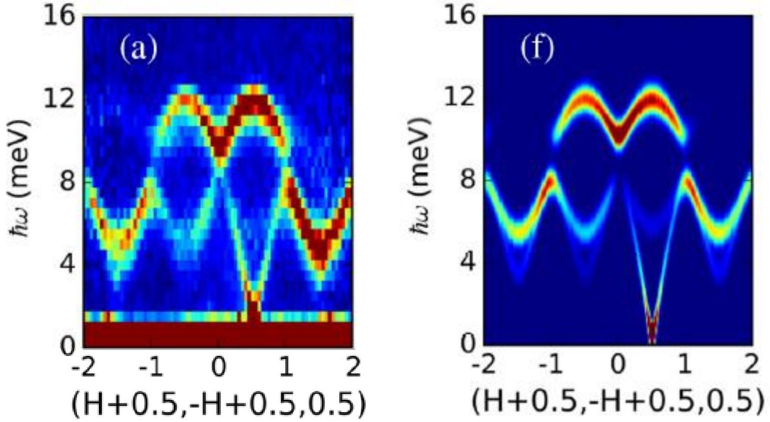
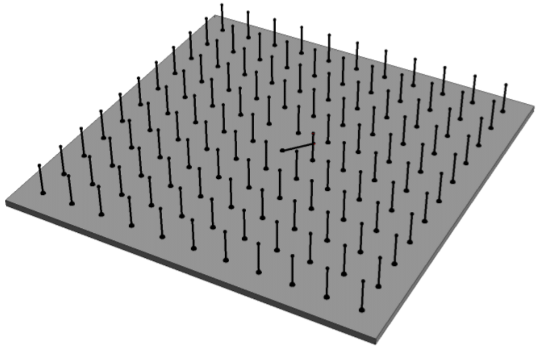
Magnetic Scattering











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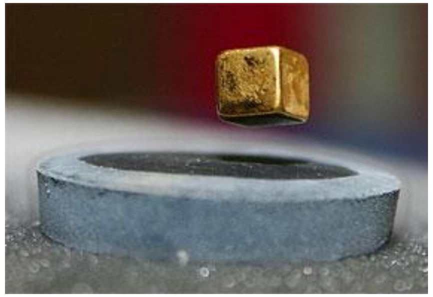
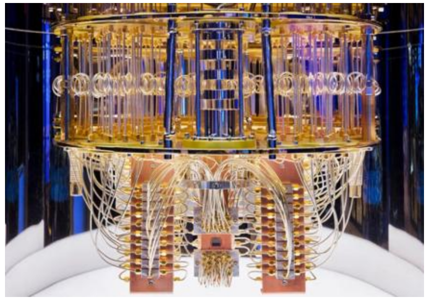
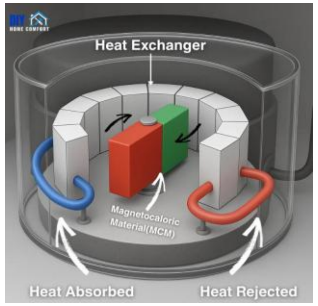
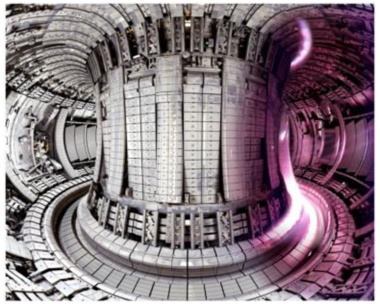
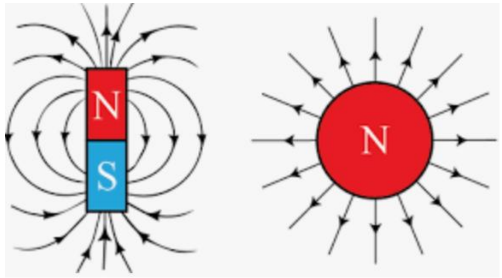
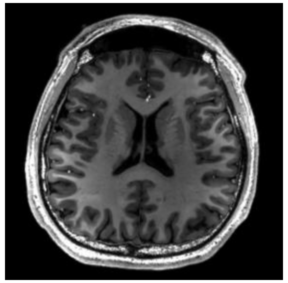
Introduction to Magnetic Scattering

- One of the “killer applications” of neutron scattering
- Neutron scattering is an essential tool for the study of magnetic materials
- Elastic Scattering (diffraction) – magnetic structure, phase transitions
- Inelastic Scattering (spectroscopy) – magnetic dynamics, excitations, interactions

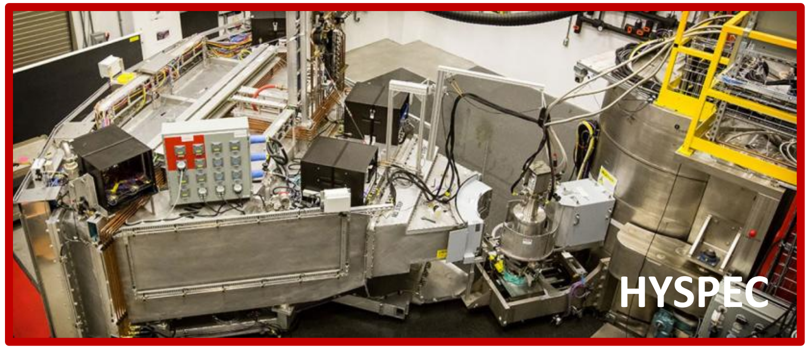



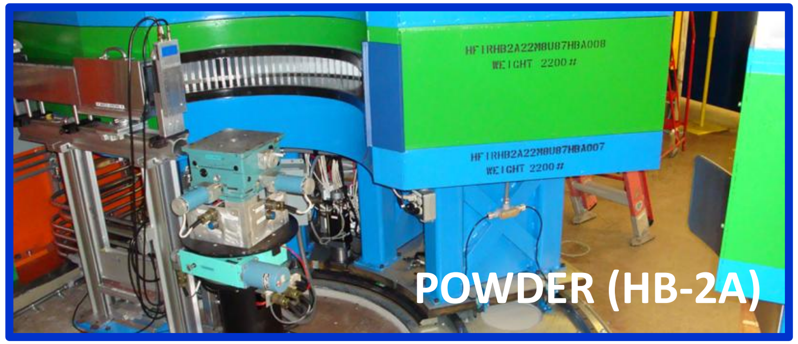


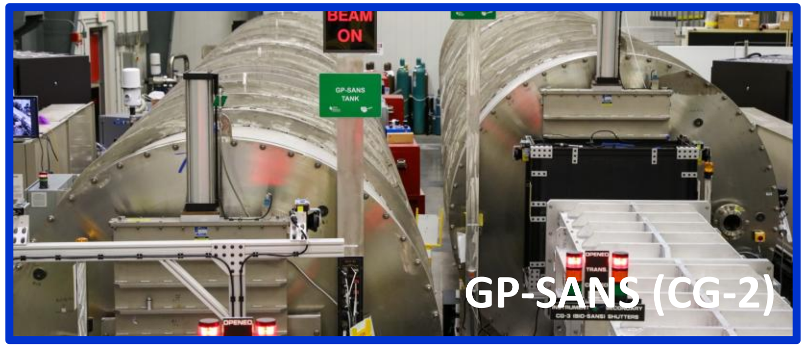



Magnetic Scattering with Neutrons







Magnetic Scattering with Neutrons

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

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

(Pauli spin operator)

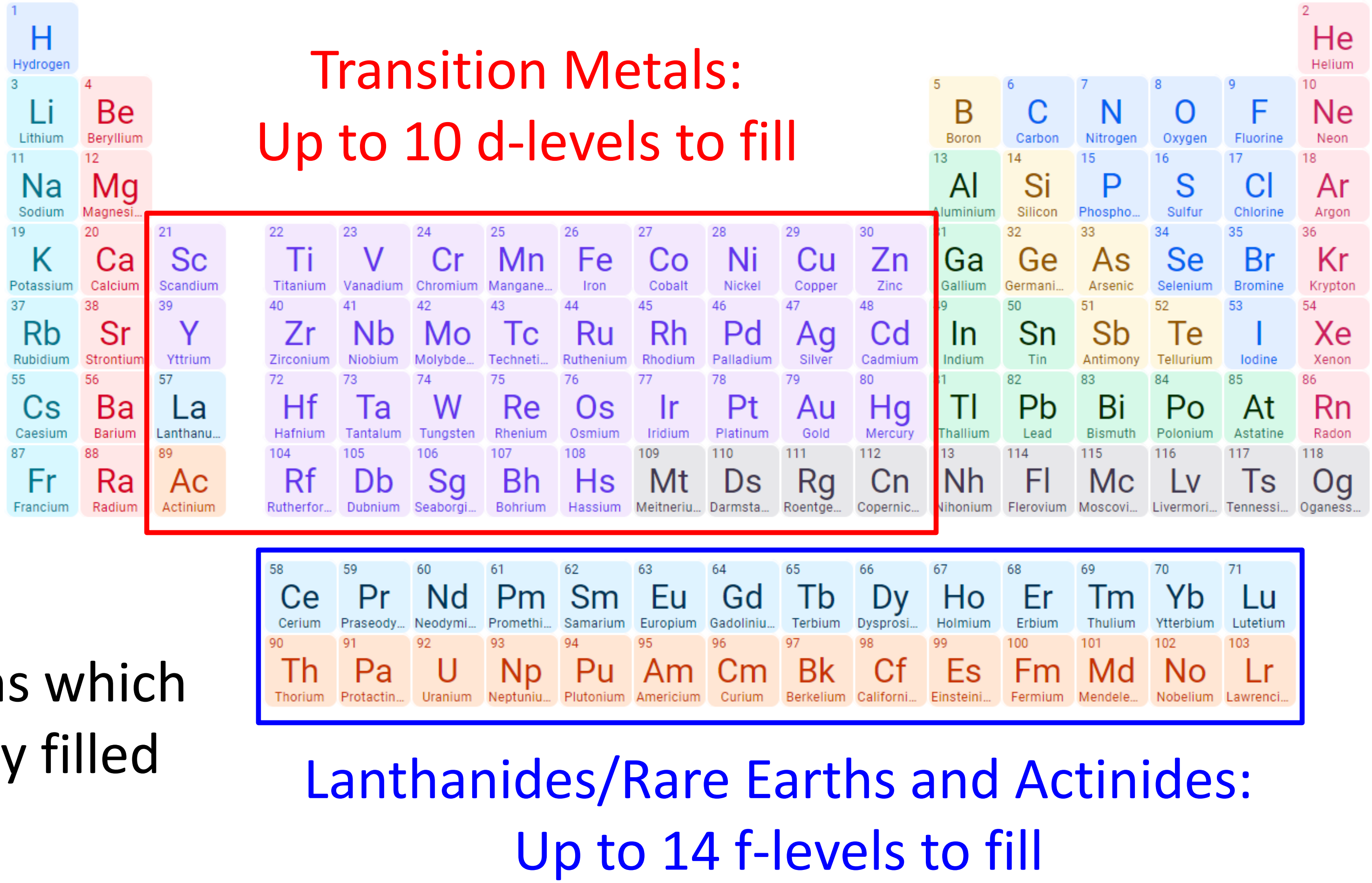
$\gamma = 1.913$
(Gyromagnetic ratio)

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

(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



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

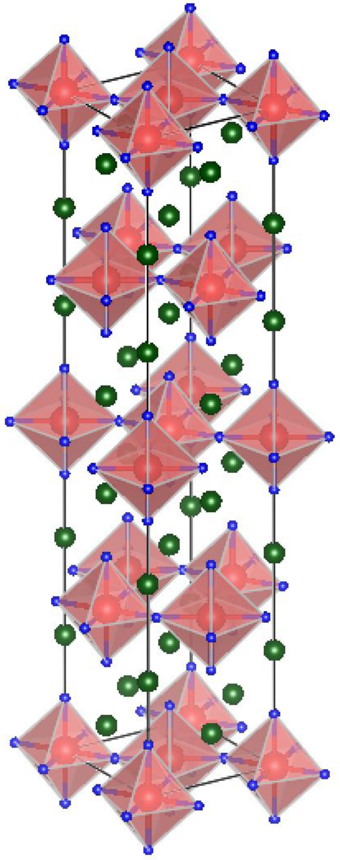
- Most common families of magnetic materials tend to be based on elements with partially filled d- or f-shells (e.g. **transition metals** or **rare earth/lanthanides**)

Magnetic Materials

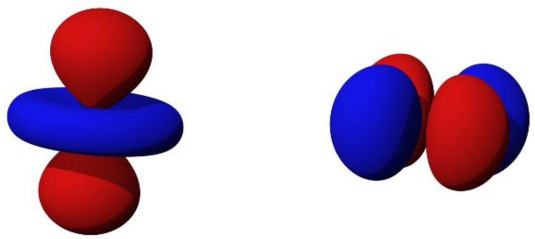
- Size of magnetic moments is determined by Hund's Rules:

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

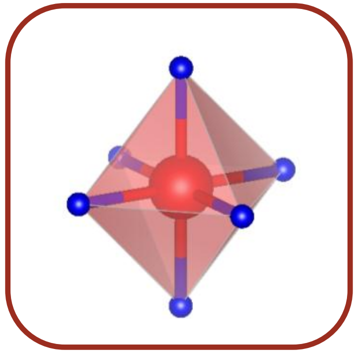
$S = 5/2$

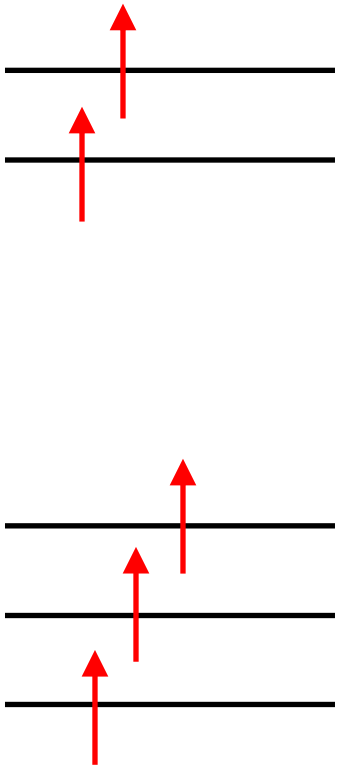


$3z^2 - r^2$ $x^2 - y^2$

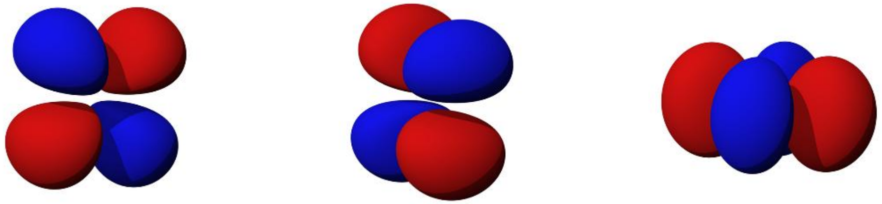


e_g orbitals

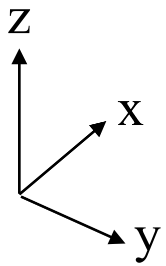




zx yz xy



t_{2g} orbitals



Magnetic Materials

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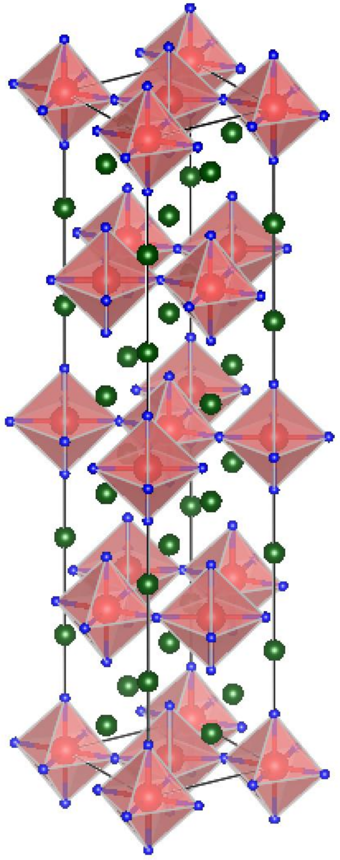
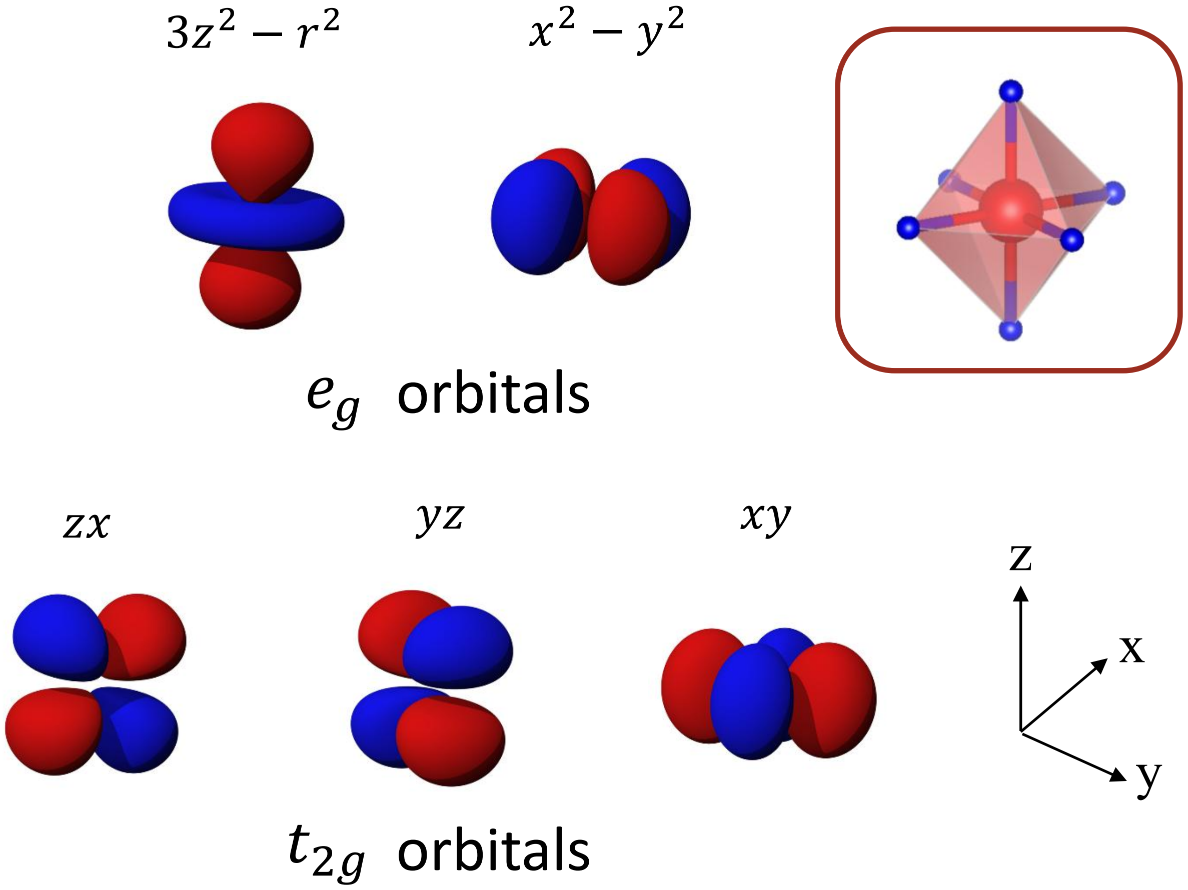
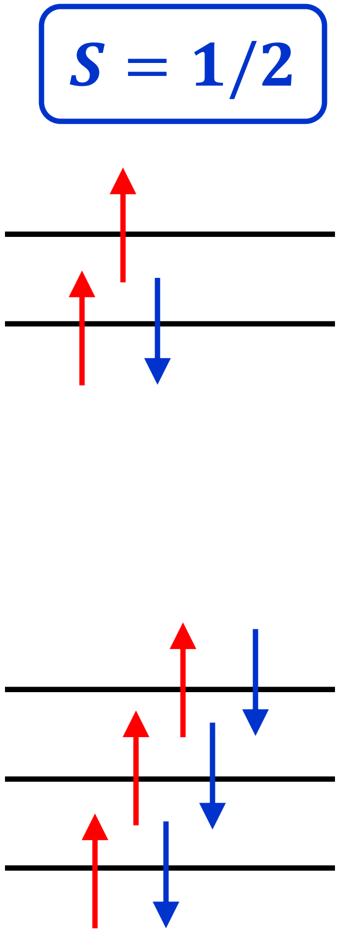


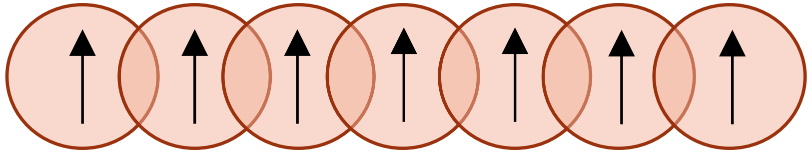


e.g. Cu^{2+} ($3d^9$)

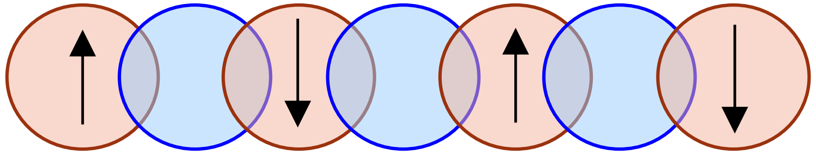


Magnetic Interactions

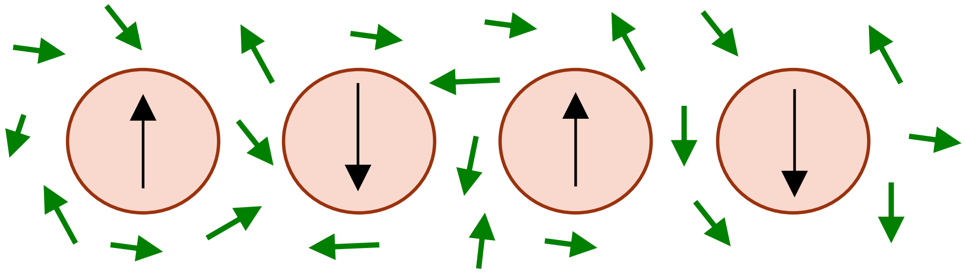
- Direct exchange:



- Superexchange:



- RKKY exchange:

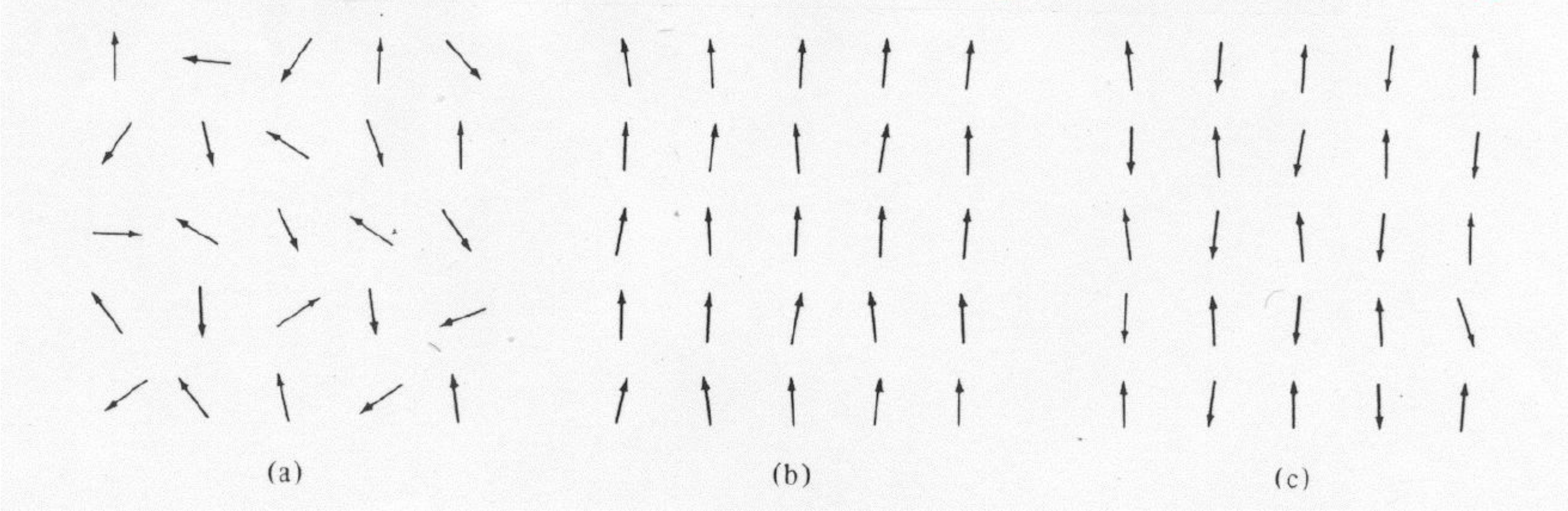


Describe interactions by a magnetic Hamiltonian:

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

(exchange parameter)

Magnetic Order



Paramagnet

($T > T_C$)

Ferromagnet

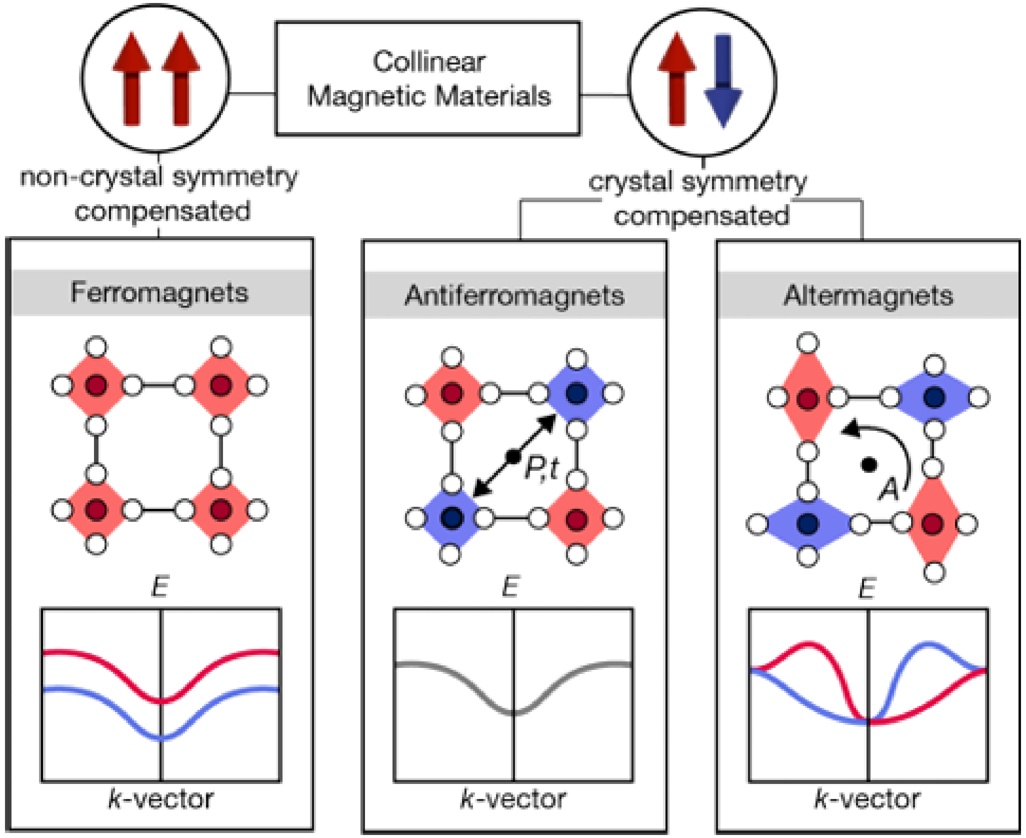
($T < T_C$)

Antiferromagnet

($T < T_N$)

A New Form of Magnetism?

Altermagnetism: Initially proposed in 2019-2021 (*Smejkal/Hayami/Yuan/Mazin et al*)



S. S. Fender et al, JACS (2025)

First experimental observations:

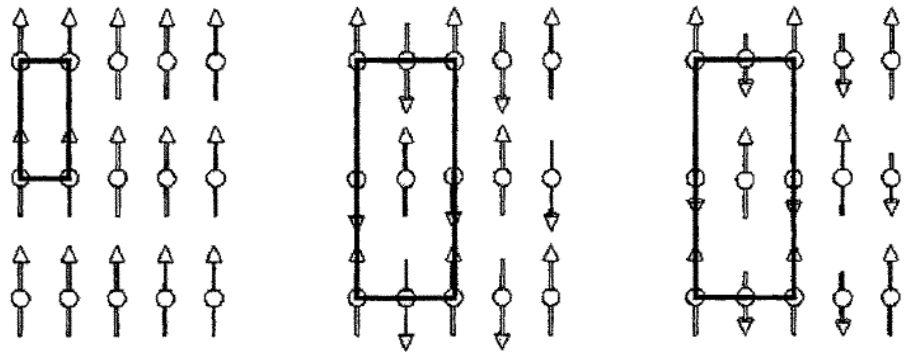
MnTe – *J. Krempasky et al, Nature (2024)*

RuO_2 – *O. Fedchenko et al, Sci. Adv. (2024)*

Magnetic Structures

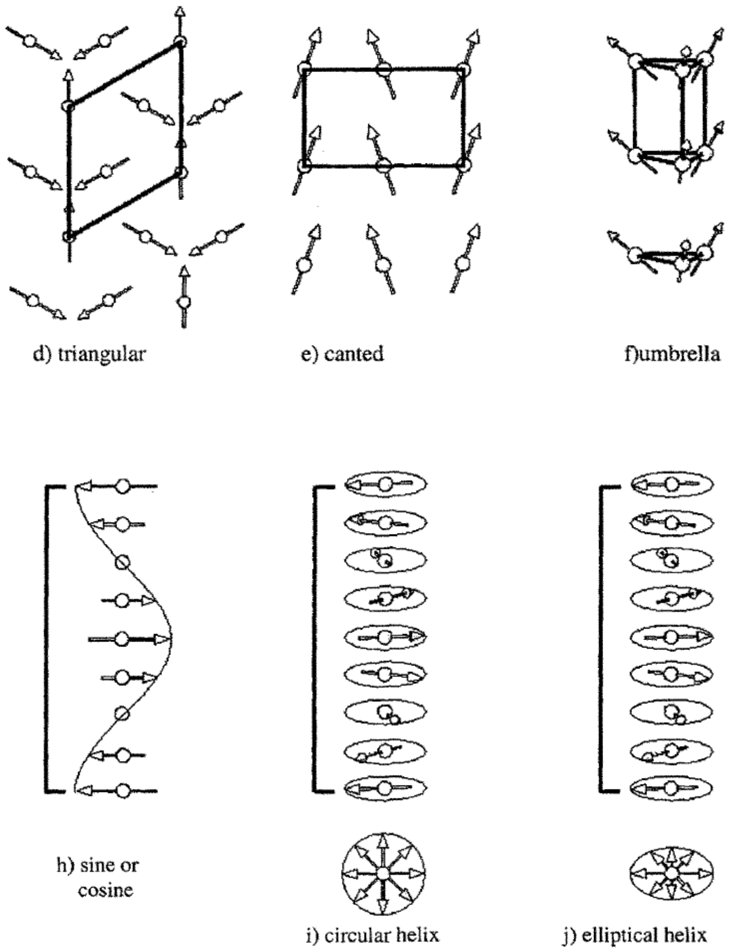
- Magnetically ordered structure that develops in a material depends on nature of underlying magnetic interactions

Structures can be relatively simple…



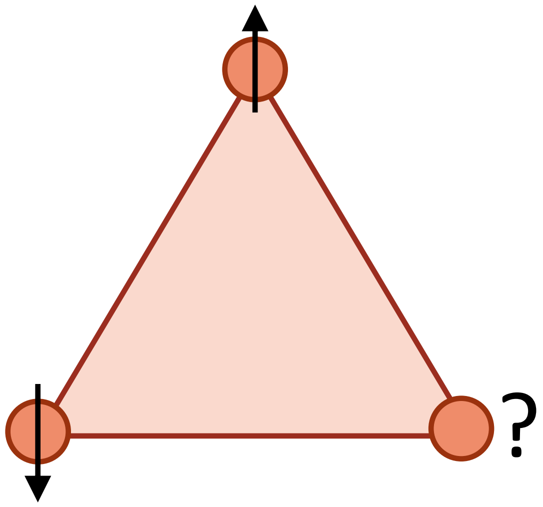
A) ferromagnetic b) antiferromagnetic c) ferrimagnetic

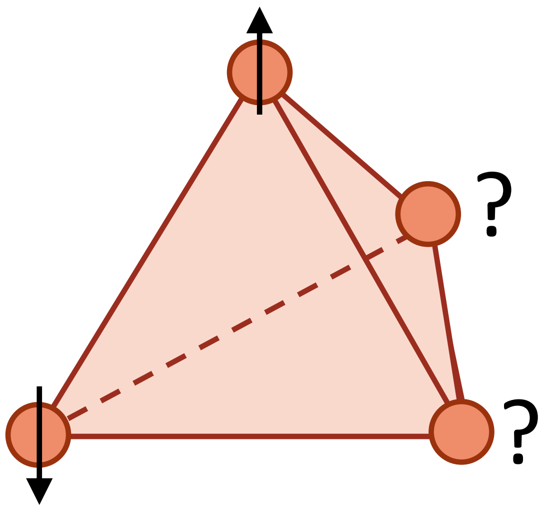
… or more complex

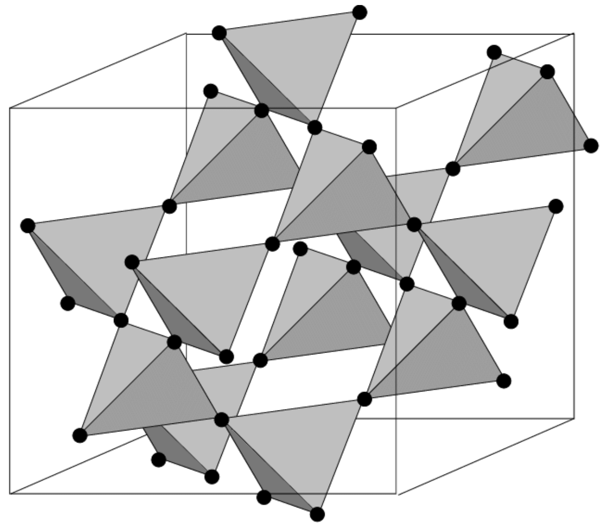


Geometric Frustration

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







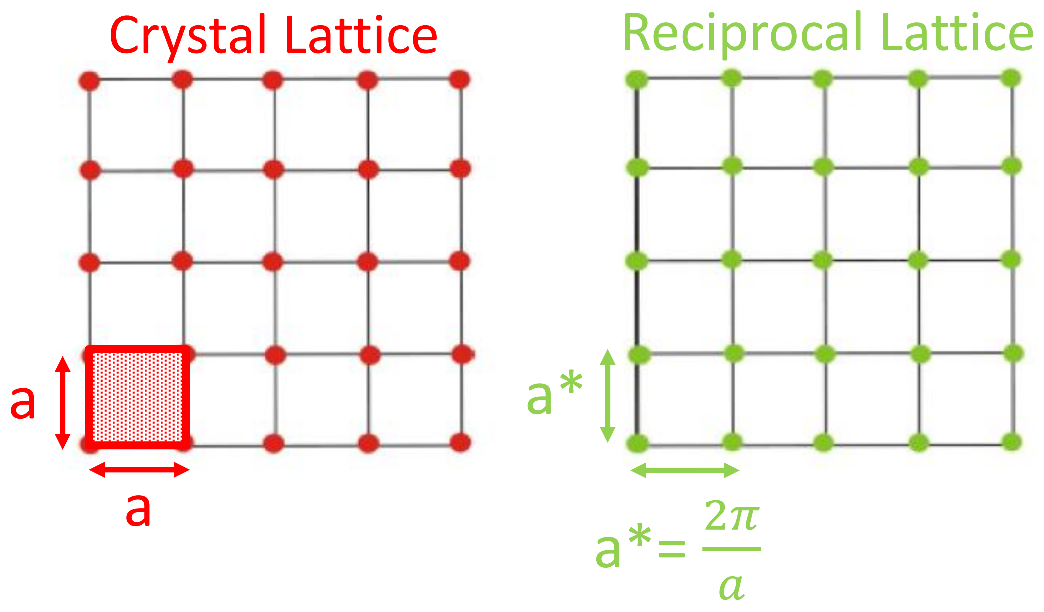
The Pyrochlore Lattice

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

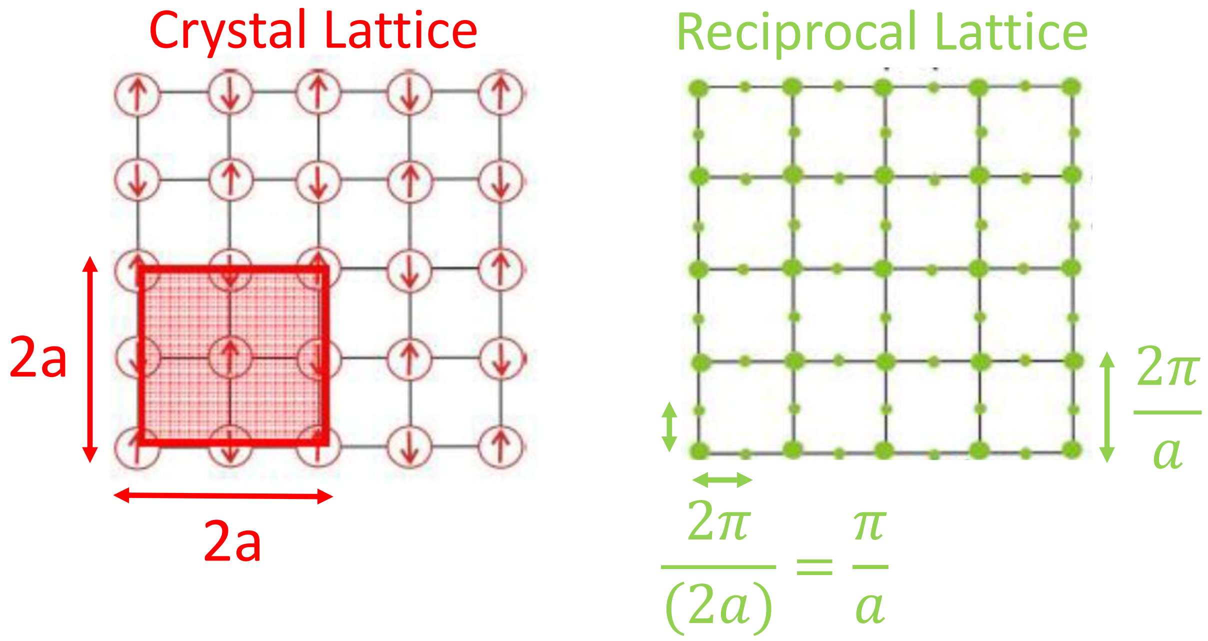
Scattering from a Magnetically Ordered Crystal

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

Paramagnetic State ($T > T_N$)



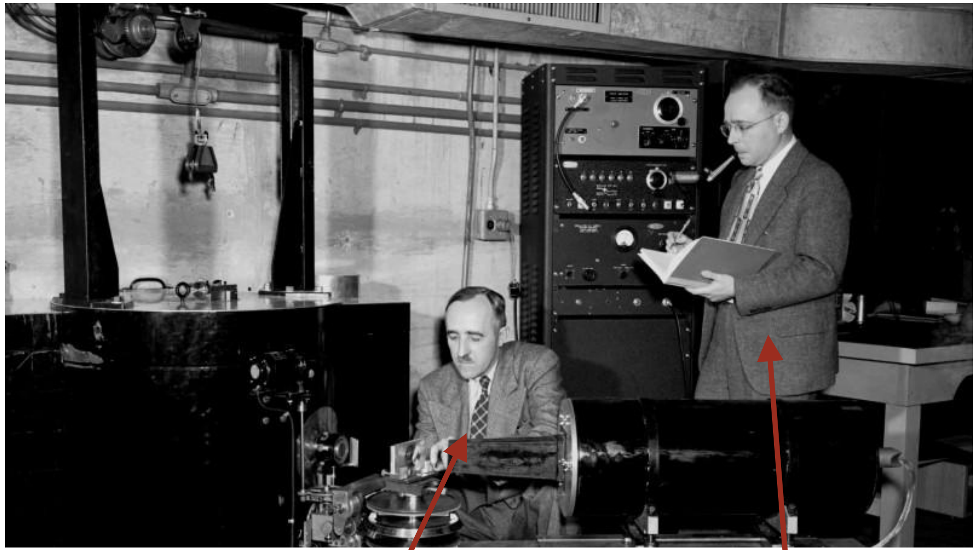
Antiferromagnetic State ($T < T_N$)



- 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

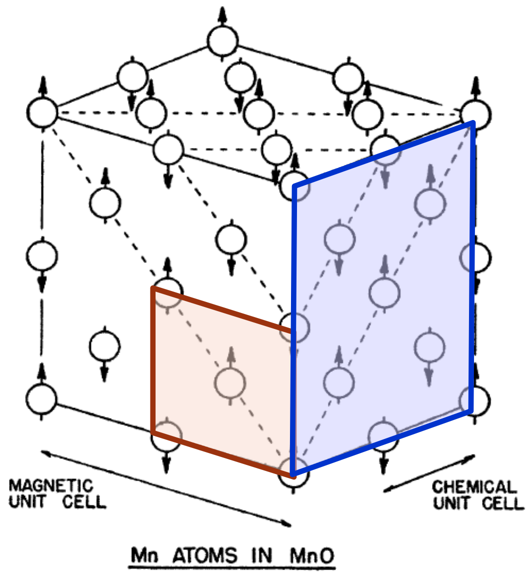


FIG. 5. Antiferromagnetic structure existing in MnO below its Curie temperature of 120°K. The magnetic unit cell has twice the linear dimensions of the chemical unit cell. Only Mn ions are shown in the diagram.

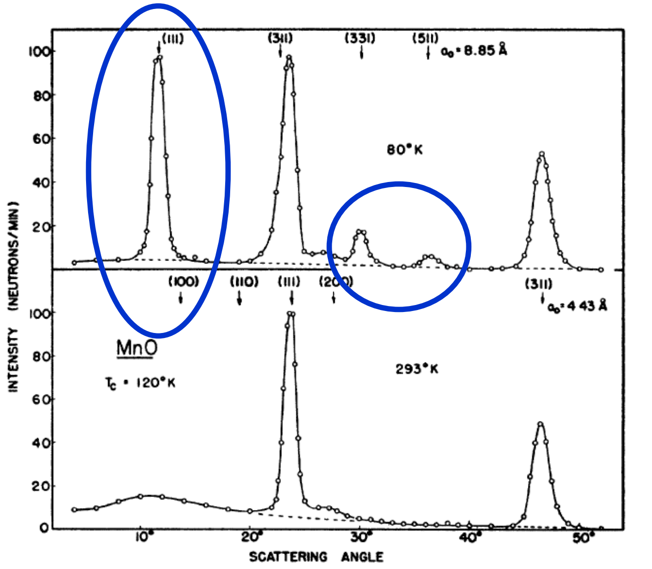


FIG. 4. Neutron diffraction patterns for MnO taken at liquid nitrogen and room temperatures. The patterns have been corrected for the various forms of extraneous, diffuse scattering mentioned in the text. Four extra antiferromagnetic reflections are to be noticed in the low temperature pattern.

First direct evidence of antiferromagnetism

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

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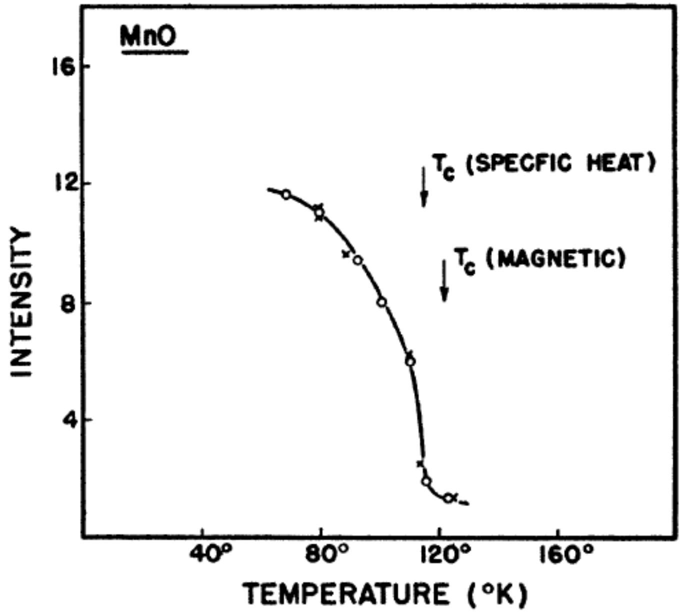
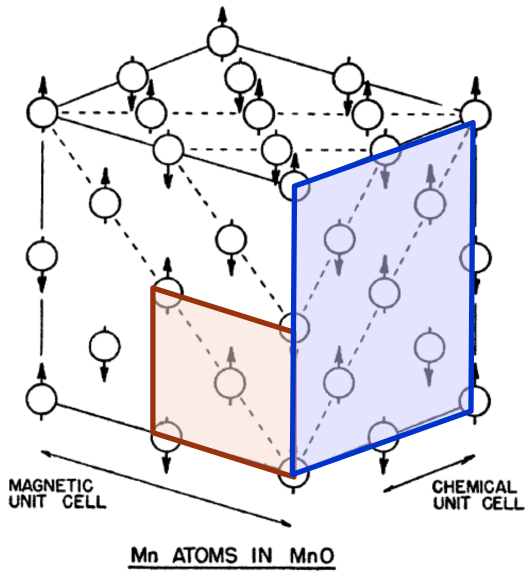


FIG. 7. Temperature dependence of magnetic intensity for MnO. The Curie temperatures suggested by specific heat and magnetic susceptibility data are shown.



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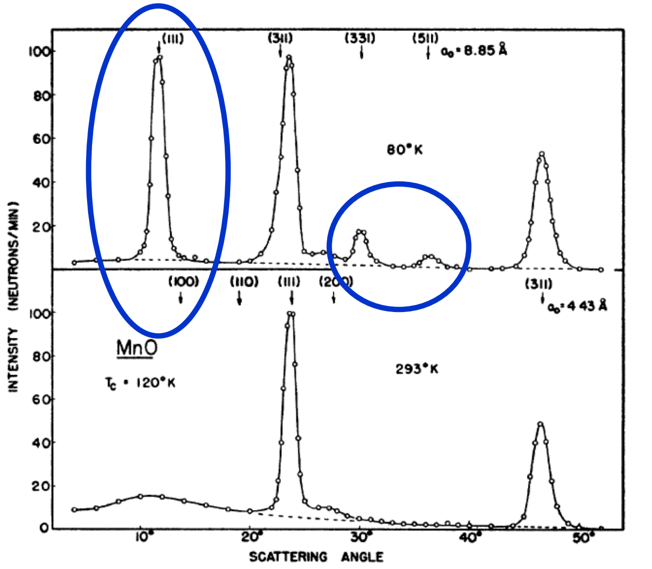

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Peak intensity ∝ staggered magnetization

First direct evidence of antiferromagnetism

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

(More on this in Experiment N7 on HB-2A)

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):

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

(Kinematics) (Interaction Term) (Energy Conservation)

The Magnetic Potential

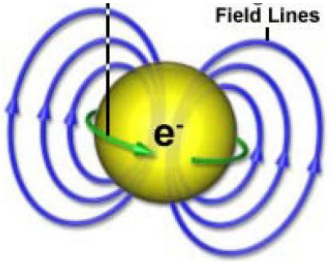
- In order to evaluate the matrix element in the interaction term, we need to determine the magnetic potential produced by all of the unpaired electrons in the material:

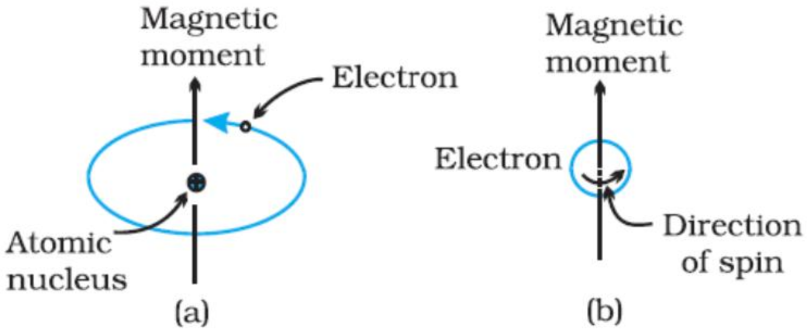
$$V_m = \vec{\mu}_n \cdot \vec{B}$$

(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:

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

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

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 $\vec{Q}$ ⟶ 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:

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

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

Key features:

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**, $S(\vec{Q},\omega)$

Magnetic Form Factor

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

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

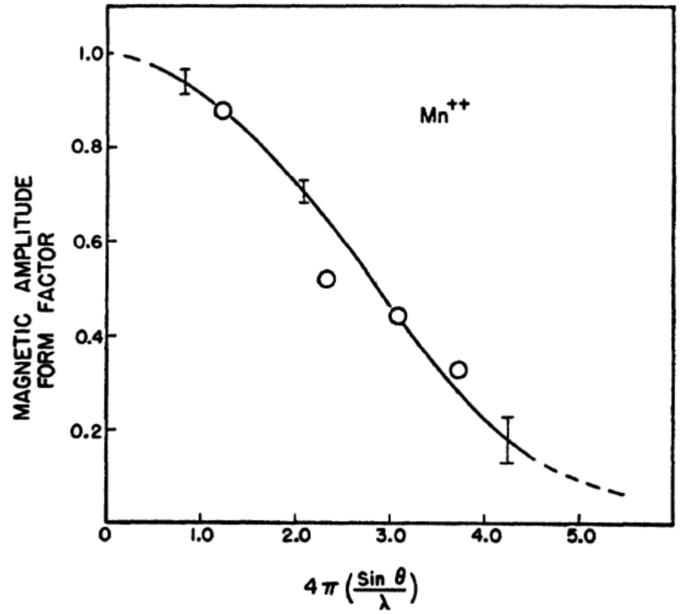
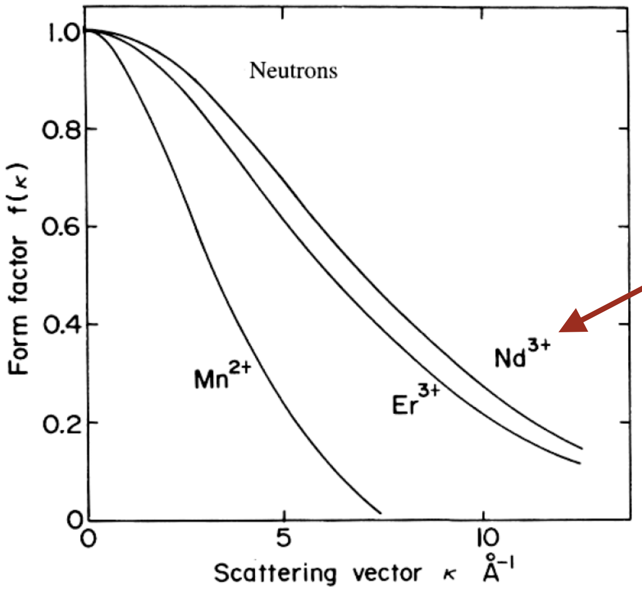


FIG. 2. Magnetic amplitude form factor for Mn^{++} ions. The curve is that obtained from paramagnetic diffuse scattering with estimated error as shown. The points represent values of the form factor obtained from the low temperature antiferromagnetic reflections of MnO.



$F\left(\vec{Q}\right)$ 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($\vec{Q}$)

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

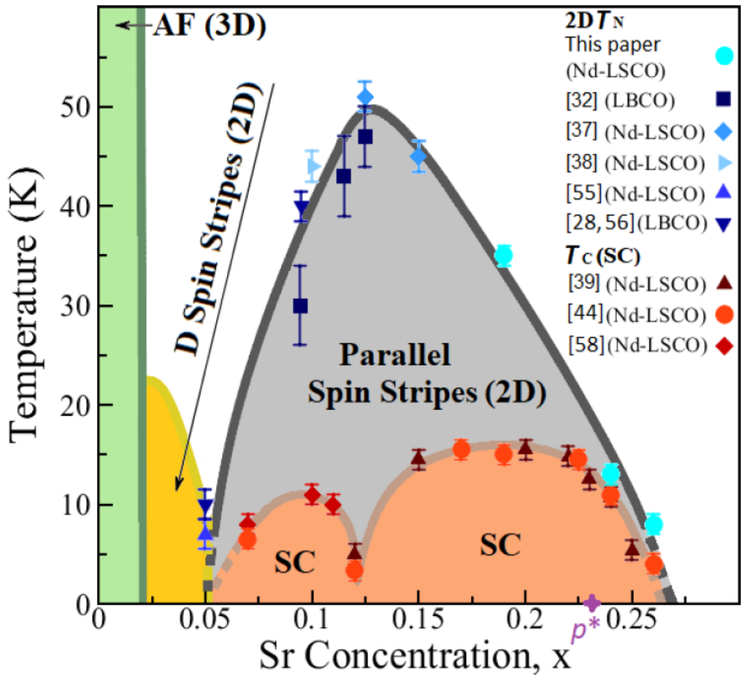
Debye-Waller Effect

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

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

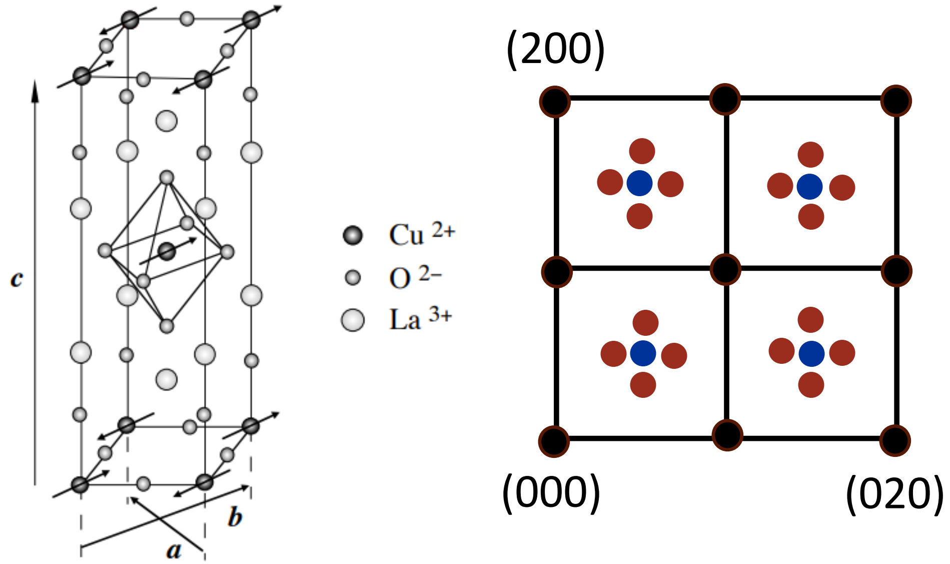
Elastic Magnetic Scattering: Examples

- **Magnetic order in High T_C cuprate superconductors (e.g. $Nd_{0.4}La_{1.6-x}Sr_xCuO_4$)**

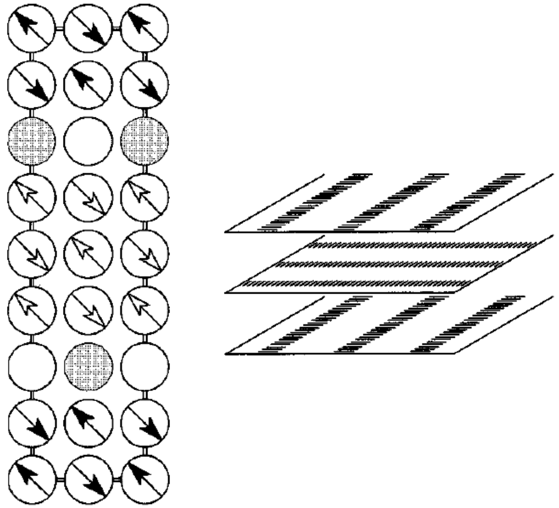


Q. Ma et al, Phys. Rev. Res. (2021)

La_2CuO_4



$Nd_{0.4}La_{1.6-x}Sr_xCuO_4$

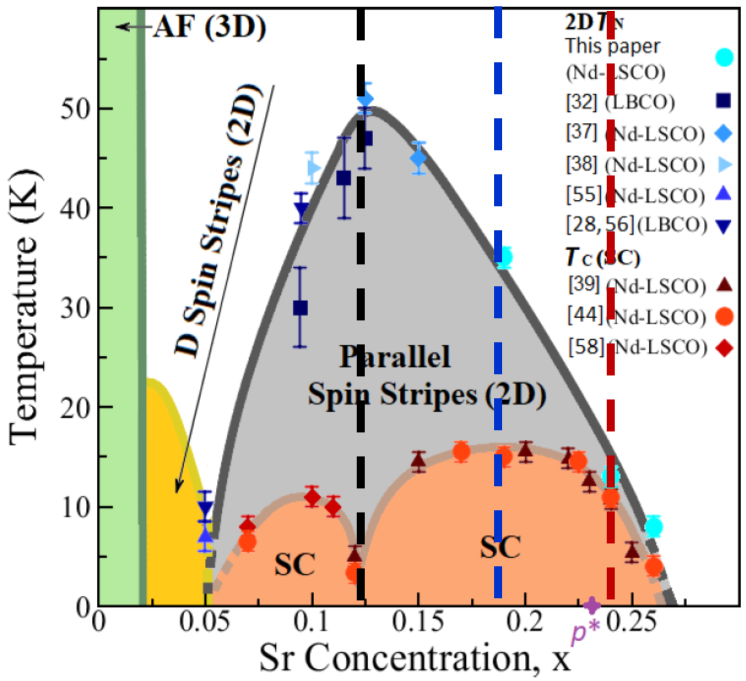


J. Tranquada et al, Phys. Rev. B (1996)

- Undoped parent compound: **commensurate** antiferromagnetic order, $\mathbf{Q_{AF}} = (0.5, 0.5, L)$
- For x > 0.02: **incommensurate "stripe"** order, $\mathbf{Q_{AF}} = (0.5 \pm \delta, 0.5, L)$ **and** $(0.5, 0.5 \pm \delta, L)$

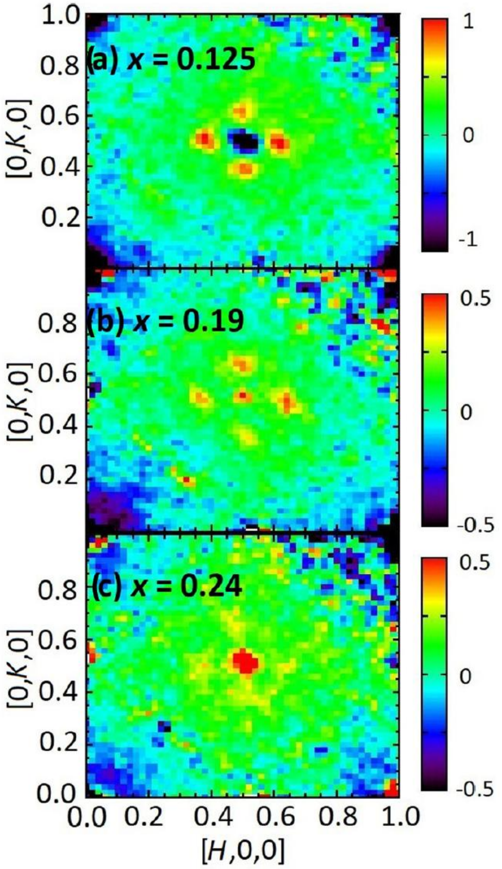
Elastic Magnetic Scattering: Examples

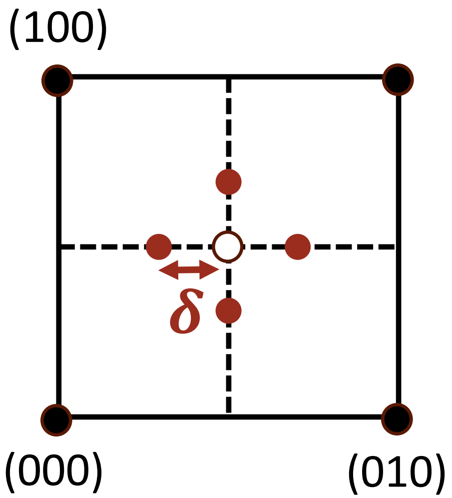
- **Magnetic order in High T_C cuprate superconductors (e.g. $Nd_{0.4}La_{1.6-x}Sr_xCuO_4$)**



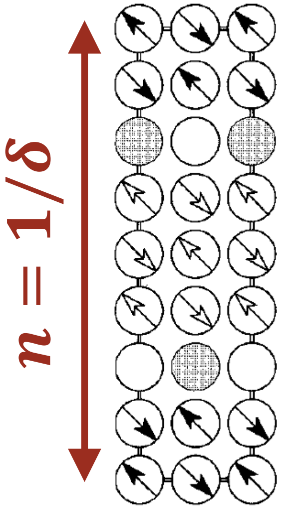
Q. Ma et al, Phys. Rev. Res. (2021)

T = 5K – 60K





$Nd_{0.4}La_{1.6-x}Sr_xCuO_4$



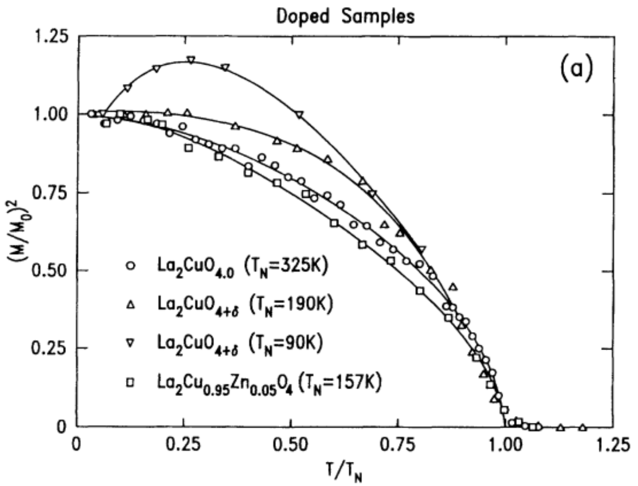
Peaks at $\mathbf{Q_{AF}} = (\mathbf{0.5 \pm \delta, 0.5, L})$ imply modulation of magnetic structure along [100] with period of $n = 1/\delta$ unit cells

Elastic Magnetic Scattering: Examples

La_2CuO_4

Well-defined Bragg peaks:

Long-range 3D magnetic order



Keimer et al, PRB (1992)

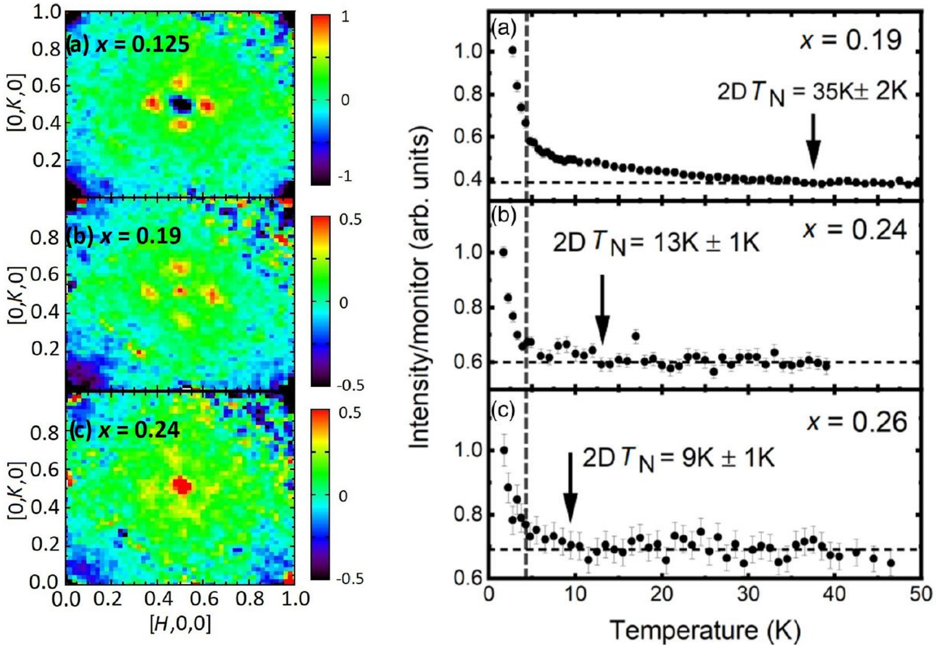
$$I \propto M^2 = {M_0}^2\left(1 - \frac{T}{T_C}\right)^{2\beta}$$

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

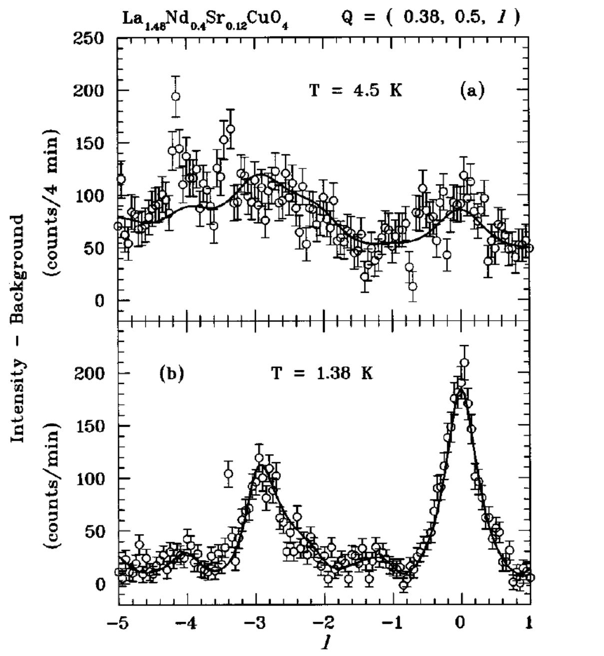
$Nd_{0.4}La_{1.6-x}Sr_xCuO_4$

Diffuse "rods" of magnetic scattering:

Quasi-2D magnetic order



Ma et al, PRR (2021)



Tranquada et al, PRB (1996)

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 $\left|\vec{k}\right| \neq \left|\vec{k}'\right| \rightarrow$ change in both $\vec{Q}$ and ω
- What we measure is the **dynamical structure factor** $S\left(\vec{Q},\omega\right)$

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

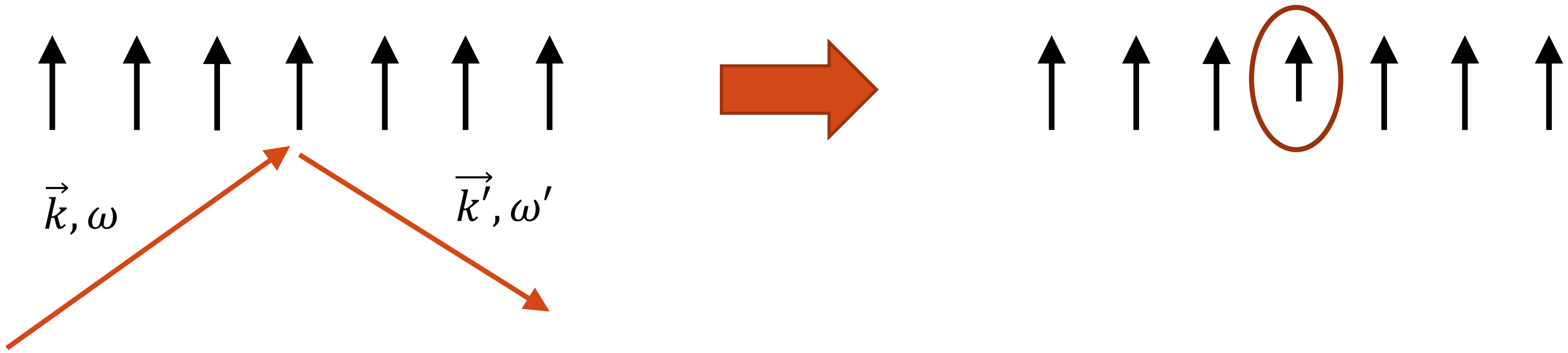
Bose (Temperature) Factor — Imaginary part of dynamic susceptibility

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

(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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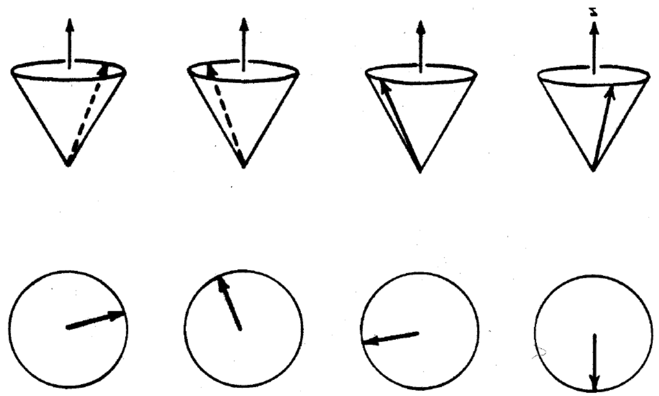

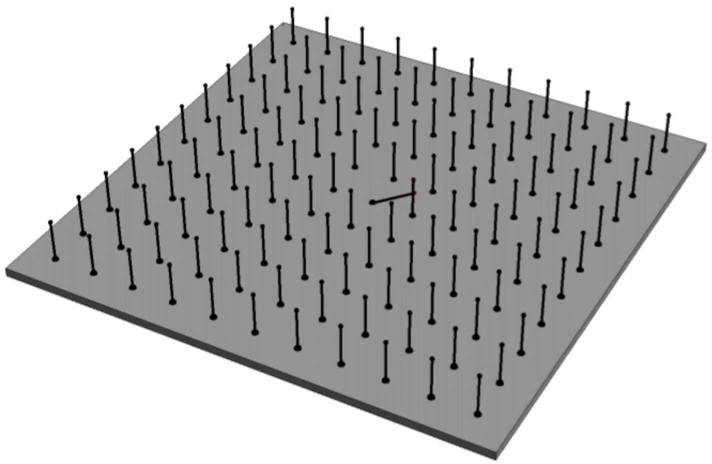

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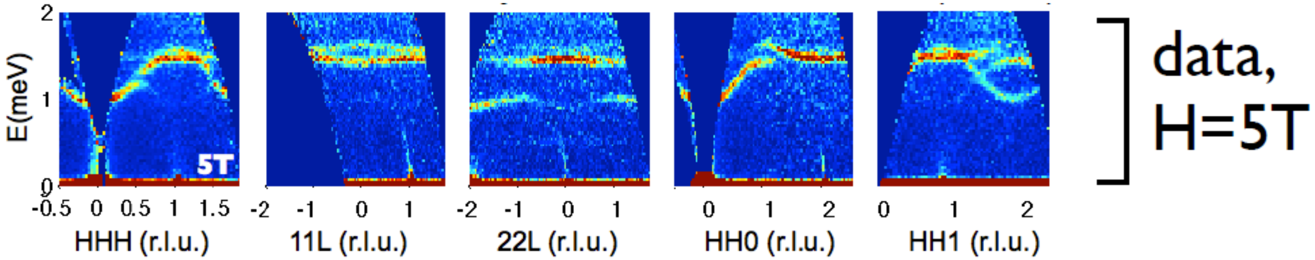
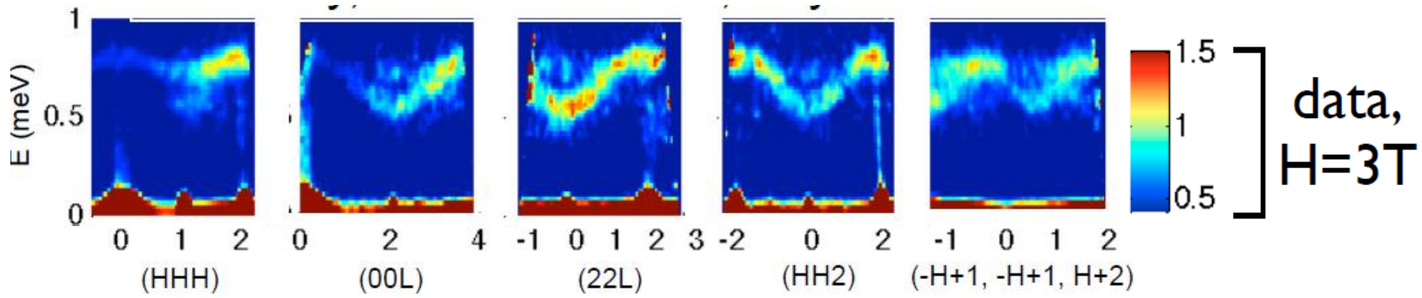
Inelastic Magnetic Scattering: Examples



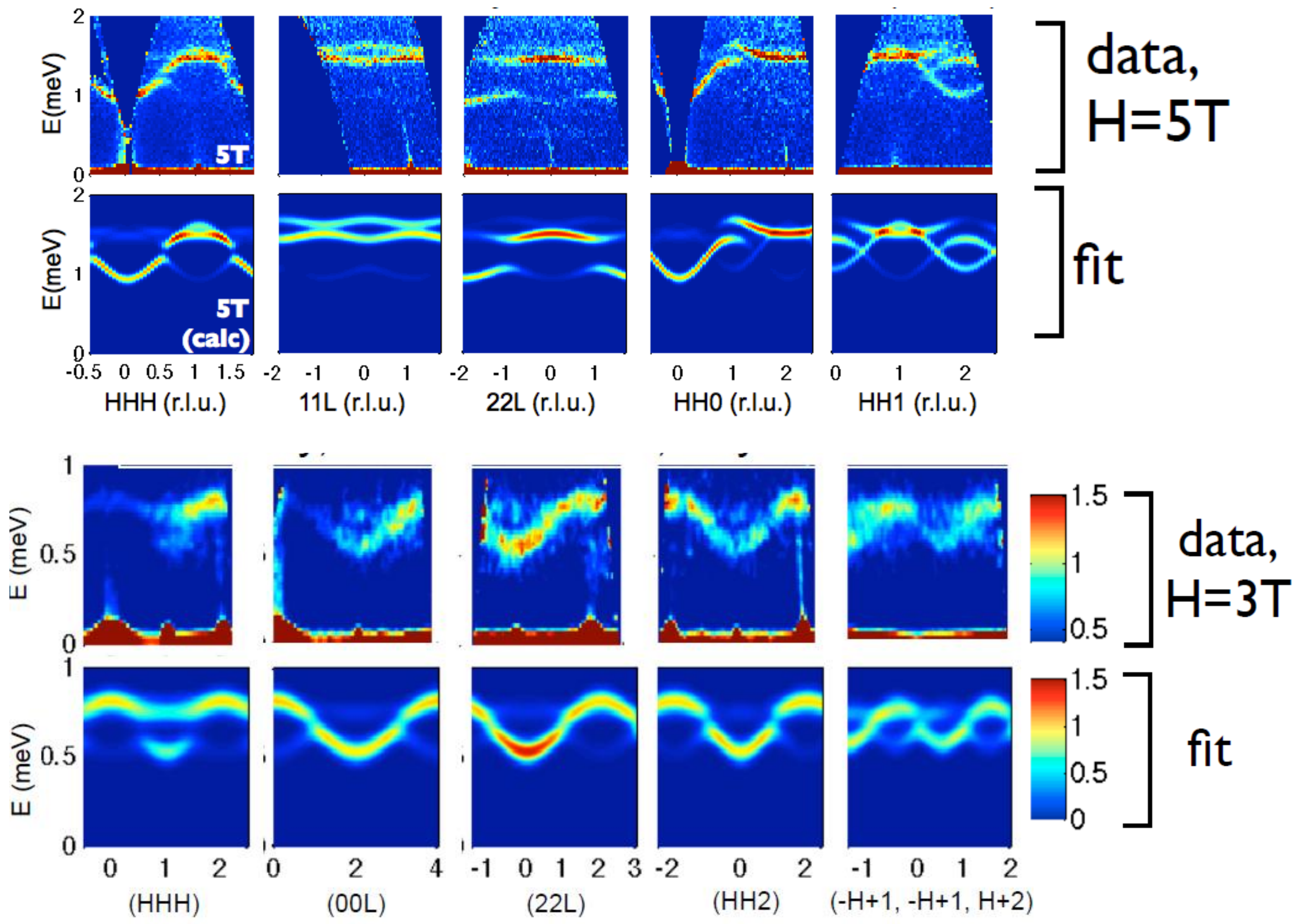
- **Frustrated magnetism in pyrochlores**
- **$Yb_2Ti_2O_7$** (top) and **$Er_2Ti_2O_7$** (bottom) Single Crystals
- Measured on DCS at NIST



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

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

Inelastic Magnetic Scattering: Examples

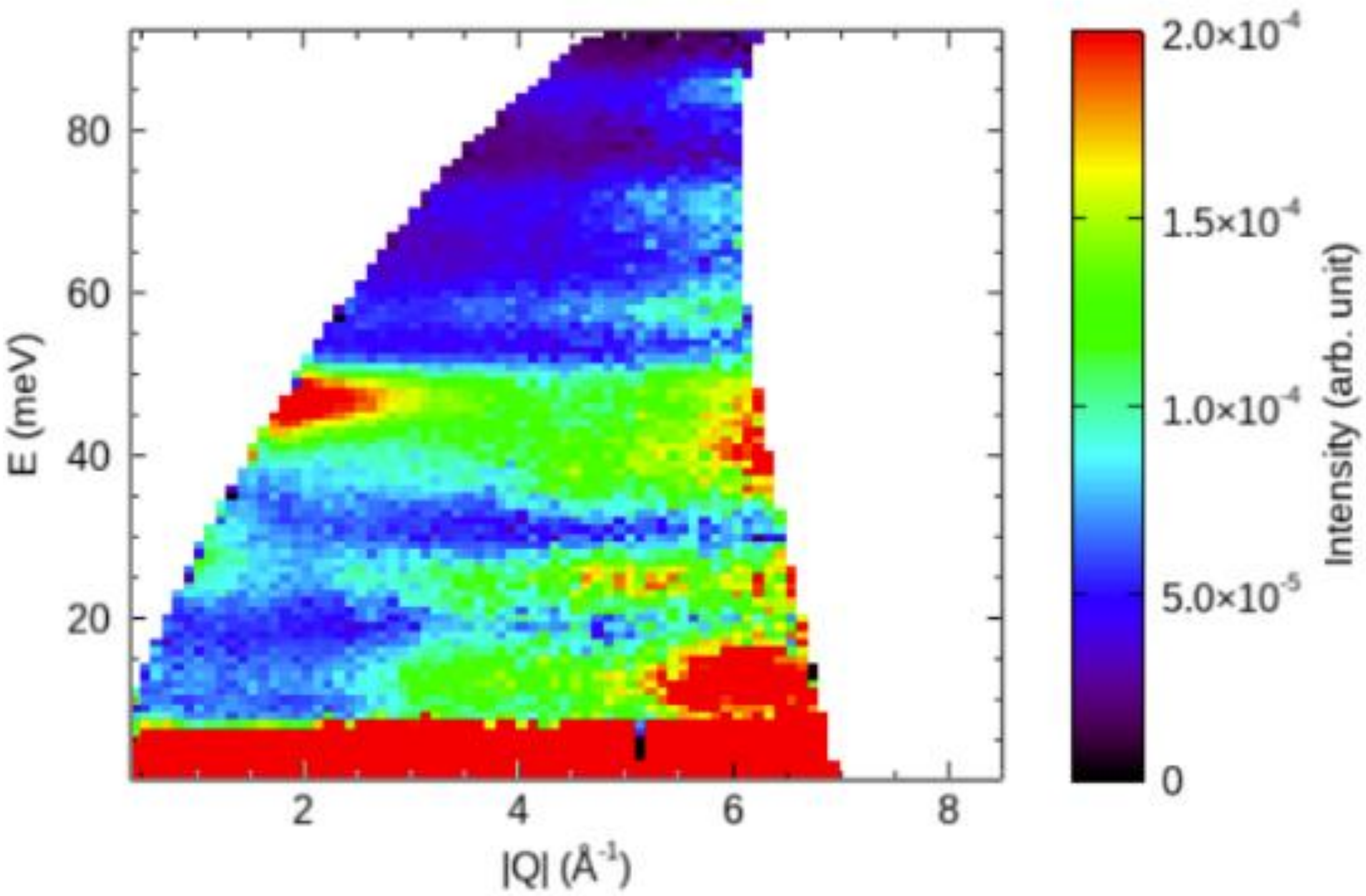


- **Frustrated magnetism in pyrochlores**
- **$Yb_2Ti_2O_7$** (top) and **$Er_2Ti_2O_7$** (bottom) Single Crystals
- Measured on DCS at NIST
- Fit spin wave dispersion to theoretical model and extract detailed exchange parameters (J_1, J_2, J_3, J_4)
- Magnetic interactions explain low temperature magnetic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

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

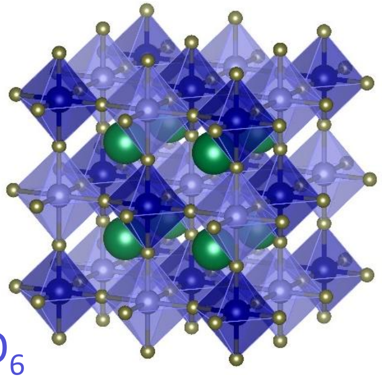
L. Savary et al, Phys. Rev. Lett. 109, 167201 (2012)

How can we distinguish magnetic scattering?



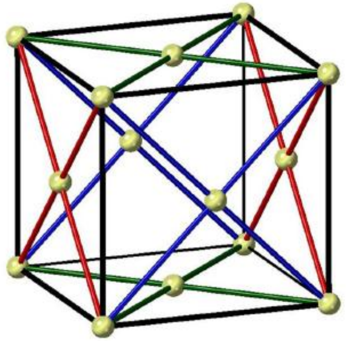
(Measurements by B. Yuan et al)

- **Spin and Orbital Excitations in Ru Double Perovskites**
- Powder samples of A_2BRuO_6
- Measured on SEQUOIA at SNS



RuO_6

BO_6



fcc lattice

How can we distinguish magnetic scattering?

(1) Momentum dependence:

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

(2) Temperature dependence:

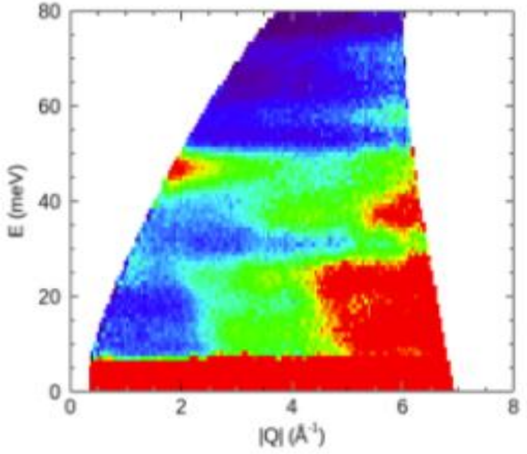
- Magnetic scattering **decreases** with increasing T (disappears at $T > T_C$)
- Phonon scattering **increases** with increasing T (∝ thermal population)

(3) Polarization dependence (with polarized beam):

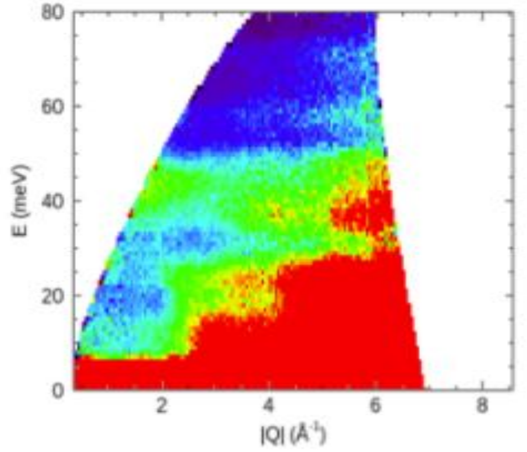
- Magnetic scattering **mostly spin flip**
- Nuclear scattering **mostly non-spin flip**

More on polarized neutrons in experiments N2 (HYSPEC) and N23 (MAGREF), and Barry Winn's lecture on Wednesday

Low Temp (T=6K)



High Temp (T=200K)



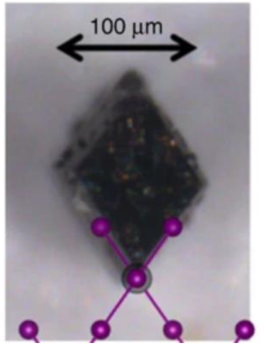
(Measurements by B. Yuan et al)

What if neutrons aren't an (easy) option?

- Magnetic neutron scattering can be challenging for:
- Very small samples (e.g. sub-mm sized crystals, thin films, heterostructures, diamond anvil or strain cells)
- Highly absorbing elements (e.g. Gd, Sm, Eu, B, Ir)
- Very expensive elements (e.g. >$1000/g)

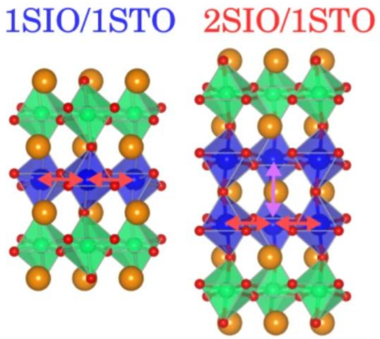
Magnetic scattering is also possible with **x-rays**…

Harmonic honeycomb β-Li_2IrO_3



K.A. Modic et al, Nat. Comm. (2014)

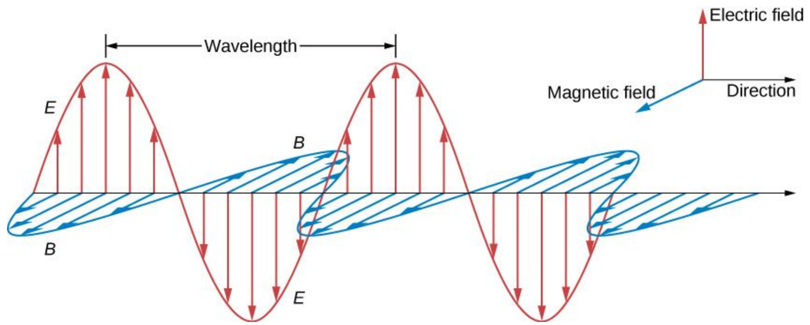
(n$SrIrO_3$/$SrTiO_3$) heterostructures



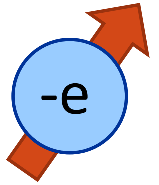
D. Meyers et al, Sci. Rep. (2019)

α-Li_2IrO_3 in diamond anvil cell

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:

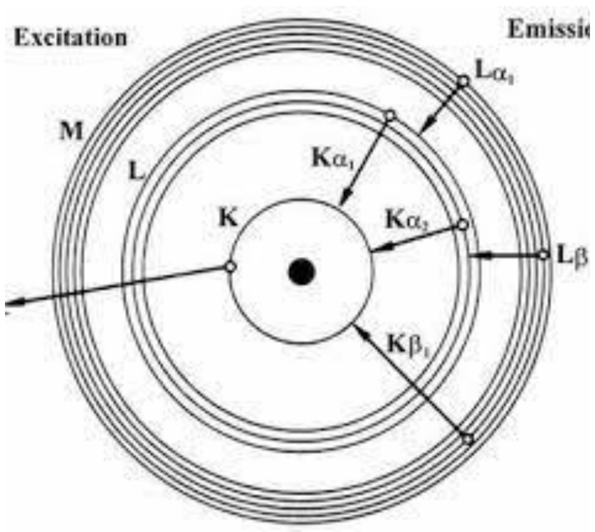
1. Magnetic scattering is MUCH weaker than charge scattering

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**)

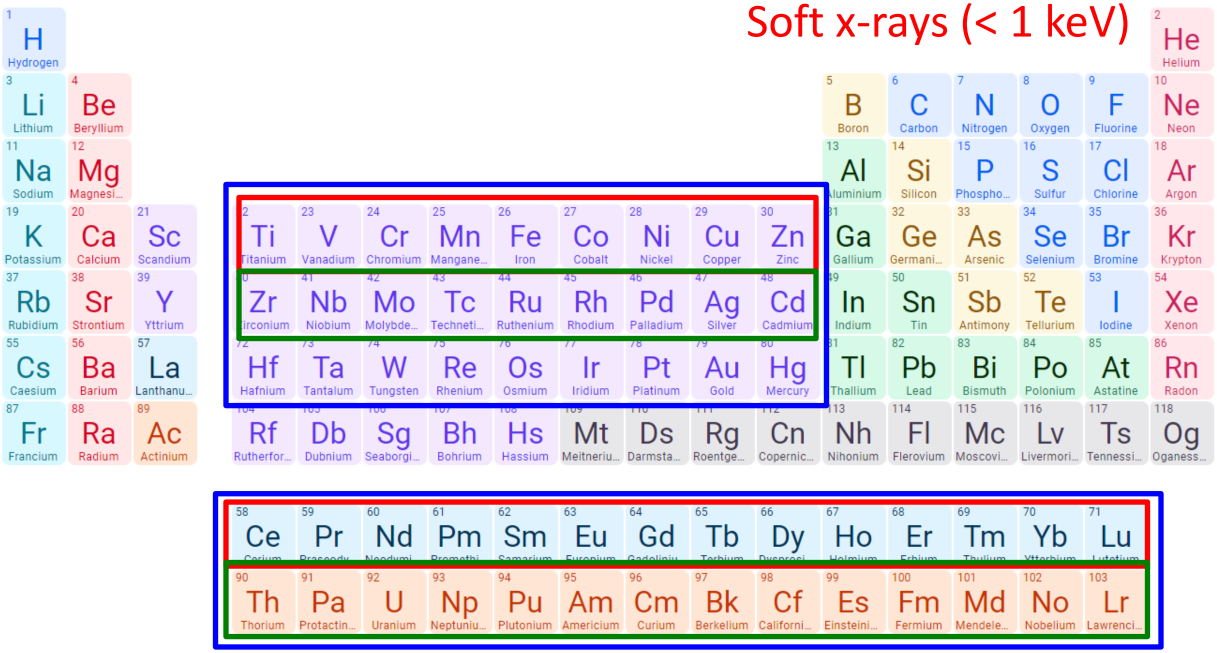
Resonant Magnetic X-ray Scattering



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

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

(But **neutrons** should almost always be your 1st choice)

Suggestions for Further Reading…

Textbooks:

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

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

T. Chatterji – *Neutron Scattering from Magnetic Materials* (2006)

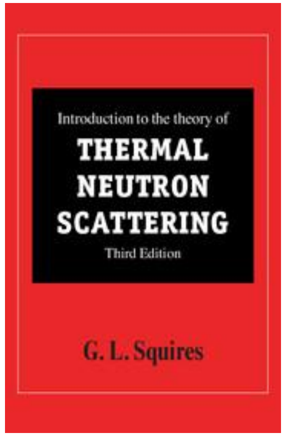
Reviews:

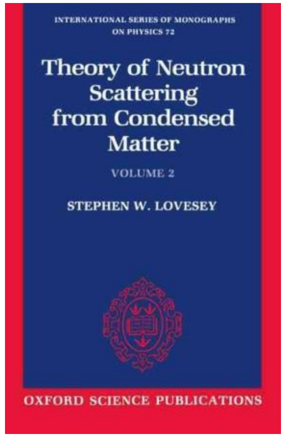
J.W. Lynn – *Magnetic Neutron Scattering* from *Characterization of Materials, Vol. 2* (2012)

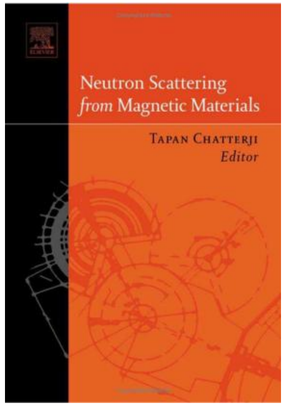
I.A. Zaliznyak and S.-H. Lee – *Magnetic Neutron Scattering* from *Modern Techniques for Characterizing Magnetic Materials* (2005)

S.M. Yusuf and A. Kumar – *Neutron Scattering of Advanced Magnetic Materials*, Appl. Phys. Rev. (2017)

S. Mühlbauer et al – *Magnetic Small-Angle Neutron Scattering*, Rev. Mod. Phys. (2019)







Any Questions?

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