Outline

- Antiferromagnetic ordering: order parameters, Néel temperature, susceptibility below and above T_N
- Frustrated antiferromagnets

Antiferromagnetic ordering: neutron diffraction

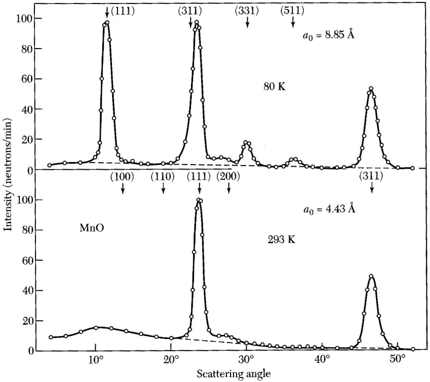
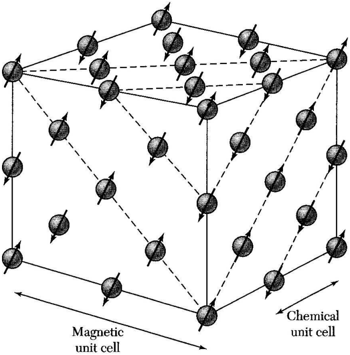


Figure 17 Neutron diffraction patterns for MnO below and above the spin-ordering temperat of 120 K, after C. G. Shull, W. A. Strauser, and E. O. Wollan. The reflection indices are basec an 8.85 Å cell at 80 K and on a 4.43 Å cell at 293 K. At the higher temperature the Mn^{2+} ions still magnetic, but they are no longer ordered.

Looks like at lower T-s, the unit cell just gets twice bigger.

Antiferromagnetic ordering, MnO example

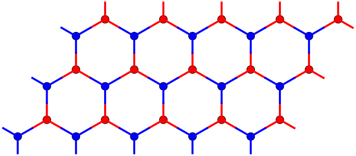


(Oxygen atoms not shown.)

Symmetry breaking at AFM phase transitions

- Rotational invariance
- Time inversion symmetry
- Translational invariance (period doubling)

Order parameters for AFM state



Two sublattices: even/odd, red/blue, etc
Can have two averages: $\langle \mathbf{S}_{\mathrm{even}} \rangle$ and $\langle \mathbf{S}_{\mathrm{odd}} \rangle$, or, equivalently

$$\mathbf{s} = \langle \mathbf{S}_{\mathrm{even}} \rangle + \langle \mathbf{S}_{\mathrm{odd}} \rangle \qquad \text{(total spin)}$$

$$\mathbf{p} = \langle \mathbf{S}_{\mathrm{even}} \rangle - \langle \mathbf{S}_{\mathrm{odd}} \rangle \qquad \text{(sublattice magnetization)}$$

Our goal: construct a Ginzburg-Landau type theory for AFM phases and phase transitions.

Interatomic exchange in solids III: Heisenberg model

$$\widehat{H} = -J_{\mathrm{ex}} \sum_{i=1}^{N} \sum_{\alpha=1}^{z} \widehat{\mathbf{S}}_i \cdot \widehat{\mathbf{S}}_{i+\alpha}$$

Where

$\sum_i$	runs over the lattice sites
$\sum_\alpha$	runs over the nearest neighbours of site i

At low T-s:

$\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow$

$J_{\mathrm{ex}} > 0$: ferromagnetic order

$\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow$

$J_{\mathrm{ex}} < 0$: antiferromagnetic order

AFM Heisenberg model

$$E = J \sum_{i:\text{even}} \mathbf{S}_i^{(\text{even})} \cdot \sum_{\alpha} \mathbf{S}_{i+\alpha}^{(\text{odd})} + J \sum_{i:\text{odd}} \mathbf{S}_i^{(\text{odd})} \cdot \sum_{\alpha} \mathbf{S}_{i+\alpha}^{(\text{even})}$$

Mean-field type approximation:

$$\begin{aligned} E/N &\approx zJ \langle \mathbf{S}_{\text{even}} \rangle \cdot \langle \mathbf{S}_{\text{odd}} \rangle \\ &= \frac{zJ}{4} \left(\mathbf{s}^2 - \mathbf{p}^2 \right) \qquad \text{since } \mathbf{s}^2 - \mathbf{p}^2 = 4 \langle \mathbf{S}_{\text{even}} \rangle \cdot \langle \mathbf{S}_{\text{odd}} \rangle \end{aligned}$$

$$\text{entropy per spin} = -2k_B \langle \mathbf{S} \rangle^2 - (4/3) k_B \langle \mathbf{S} \rangle^4$$

$$\begin{aligned} \text{entropy per unit cell} = &- 2k_B \langle \mathbf{S}_{\text{even}} \rangle^2 - \frac{4}{3} k_B \langle \mathbf{S}_{\text{even}} \rangle^4 \\ &- 2k_B \langle \mathbf{S}_{\text{odd}} \rangle^2 - \frac{4}{3} k_B \langle \mathbf{S}_{\text{odd}} \rangle^4 \end{aligned}$$

AFM Heisenberg model, cont'd

Using $2\langle \mathbf{S}_{\rm even}\rangle = \mathbf{s}+\mathbf{p}$ and $2\langle \mathbf{S}_{\rm odd}\rangle = \mathbf{s}-\mathbf{p}$:

$$\langle \mathbf{S}_{\rm even}\rangle^2 + \langle \mathbf{S}_{\rm odd}\rangle^2 = \frac{1}{4}\left(2s^2+2p^2\right)$$
$$\langle \mathbf{S}_{\rm even}\rangle^4 = \left[\langle \mathbf{S}_{\rm even}\rangle^2\right]^2 = \frac{1}{16}\left[s^2+p^2+2(\mathbf{s}\cdot\mathbf{p})\right]^2$$
$$\langle \mathbf{S}_{\rm odd}\rangle^4 = \left[\langle \mathbf{S}_{\rm odd}\rangle^2\right]^2 = \frac{1}{16}\left[s^2+p^2-2(\mathbf{s}\cdot\mathbf{p})\right]^2$$

$$\langle \mathbf{S}_{\rm even}\rangle^4 + \langle \mathbf{S}_{\rm odd}\rangle^4 = \frac{1}{16}\left[2s^4+2p^4+4s^2p^2+8(\mathbf{s}\cdot\mathbf{p})^2\right]$$

Finally, entropy density

$$S = -k_B(s^2+p^2) - \frac{1}{6}k_B\left[s^4+p^4+2s^2p^2+4(\mathbf{s}\cdot\mathbf{p})^2\right]$$

Ginzburg-Landau theory

Free energy =

interaction energy − temperature × entropy

$$F - F_0 = \left(k_B T + \frac{Jz}{4}\right) s^2 + \left(k_B T - \frac{Jz}{4}\right) p^2 + \frac{1}{6} k_B T \left[s^4 + p^4 + 2s^2 p^2 + 4(\mathbf{s}\cdot\mathbf{p})^2\right]$$

$\mathbf{s} = 0\,, \mathbf{p} \neq 0:$	AFM
$\mathbf{s} \neq 0\,, \mathbf{p} = 0:$	FM (need external field)
$\mathbf{s} = 0\,, \mathbf{p} = 0:$	unordered

NEEL TEMPERATURE

LOOK FOR $\mathbf{s} = 0\,, \mathbf{p} \neq 0$

$$F = \left(k_B T - \frac{Jz}{4}\right) p^2 + \frac{1}{6} k_B T \left[p^4\right]$$

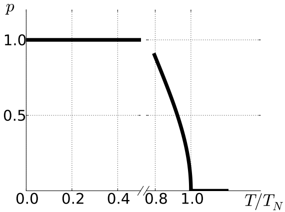
In thermodynamic equilibrium $F = \min \Rightarrow$

$$0 = \frac{\partial F}{\partial p} = \left(-\frac{Jz}{4} + k_B T\right) 2p + \frac{1}{6} k_B T 4p^3$$

NÉEL TEMPERATURE: $T_N = zJ/4k_B$

For $T > T_N$: $\mathbf{p} = 0$

For $T < T_N$: $p = \left(3\frac{T_N - T}{T}\right)^{1/2}$



Susceptibility of AFM for $T > T_N$

For $T > T_N$ take $\mathbf{p} = 0$, $\mathbf{s} \upuparrows \mathbf{H}$ and s small (neglect the s^4 term):

$$F = -\mu \mathbf{s} \cdot \mathbf{H} + k_B(T + T_N)\,\mathbf{s}^2 + \frac{1}{6} k_B T\,\mathbf{s}^4$$
$$\approx -\mu\, sH + k_B(T + T_N)\, s^2$$

$$0 = \frac{\partial F}{\partial s} = -\mu H + 2s\, k_B(T + T_N) \qquad \Rightarrow \qquad \mu s = \frac{\mu^2}{2k_B} \frac{H}{T + T_N} \equiv \chi H$$

$\chi(T)$:

	Paramagnet	Ferromagnet ($T > T_c$)	Antiferromagnet ($T > T_N$)
$\chi \propto$	$\frac{1}{T}$	$\frac{1}{T - T_c}$	$\frac{1}{T + T_N}$

Weak fields: Susceptibility of AFM for $T < T_N$

For $T < T_N$
$\mathbf{p} \neq 0$ *spontaneously*, $p = \sqrt{3(T_N - T)/T}$ and is directed along an easy axis.
$\mathbf{s} \neq 0$ because of the applied field $\mathbf{H}$ and is directed along it.

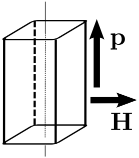
Need to distinguish two situations:

$$\chi_{\parallel} : \quad \mathbf{H} \parallel \mathbf{s} \parallel \mathbf{p}$$
$$\chi_{\perp} : \quad \mathbf{H} \parallel \mathbf{s} \perp \mathbf{p}$$

As before, to calculate the susceptibility we need to only consider *weak fields*, hence will be neglecting higher-order terms where possible.

Perp susceptibility of AFM for $T < T_N$

$$\mathbf{H} \parallel \mathbf{s} \perp \mathbf{p} \quad \Rightarrow (\mathbf{s} \cdot \mathbf{p}) = 0$$



$$F = -\mu s H + k_B(T + T_N)s^2 + k_B(T - T_N)p^2 + \frac{1}{6}k_B T \left[2s^2p^2 + s^4 + p^4\right]$$

Using $p^2 = 3(T_N - T)/T$ and neglecting quartic terms

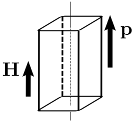
$$F = -\mu s H + k_B(T + T_N)s^2 + k_B(T_N - T)s^2 = -\mu s H + 2k_B T_N s^2$$

$$0 = \frac{\partial F}{\partial s} = -\mu H + 4k_B T_N s$$

$$\chi_\perp \propto \frac{1}{T_N} \qquad T\text{-independent}$$

Parallel susceptibility of AFM for $T < T_N$

$$\mathbf{H} \parallel \mathbf{s} \parallel \mathbf{p} \quad \Rightarrow (\mathbf{s}\cdot\mathbf{p})^2 = s^2\, p^2$$



$$F = -\mu s H + k_B(T+T_N)s^2 + k_B(T-T_N)p^2 + \frac{1}{6}k_B T\left[6s^2p^2 + s^4 + p^4\right]$$

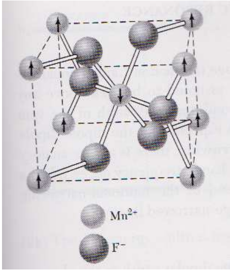
Using $p^2 = 3(T_N - T)/T$ and neglecting quartic terms

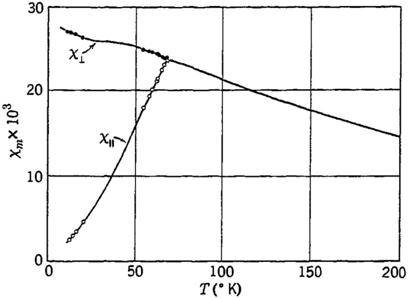
$$\begin{aligned} F &= -\mu s H + k_B(T+T_N)s^2 + 3k_B(T_N - T)s^2 \\ &= -\mu s H + 2k_B(T_N + |T - T_N|)s^2 \end{aligned}$$

$$0 = \frac{\partial F}{\partial s} = -\mu H + 4k_B(T_N + |T-T_N|)s$$

$$\chi_\parallel \propto \frac{1}{T_N + |T - T_N|}$$

How this looks in real life: MnF_2





both plots taken from Kittel's *ISSP*.

AFM in strong magnetic fields and $T < T_N$

Consider the parallel fields, $\mathbf{H} \parallel \mathbf{s} \parallel \mathbf{p}$, and define dimensionless variables

$$f = F/k_B T_N \qquad h = \mu H/k_B T_N \qquad \tau = T/T_N \quad (\tau < 1)$$

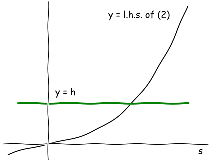
$$f = (1+\tau)s^2 - (1-\tau)p^2 + \frac{\tau}{6}(s^4 + p^4 + 6s^2p^2) - hs$$

Thermal equilibrium:

$$0 = \partial f/\partial p = 2p\left[s^2 - (1-\tau) + \frac{\tau}{3}p^2\right] \tag{1}$$

$$0 = \partial f/\partial s = 2s\left[p^2 + (1+\tau) + \frac{\tau}{3}s^2\right] - h \tag{2}$$

AFM in strong magnetic fields and $T < T_N$, II



Eq. (2): l.h.s. is monotonic, thus there is exactly one solution with $s \neq 0$.

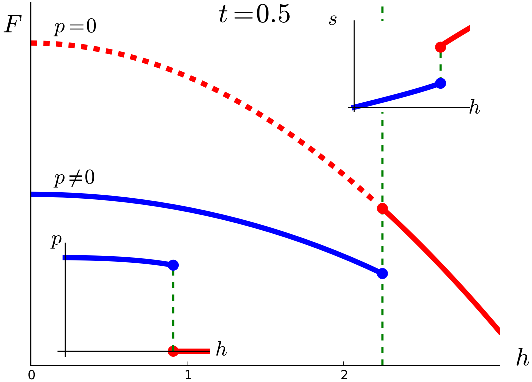
Solutions of Eq. (1)

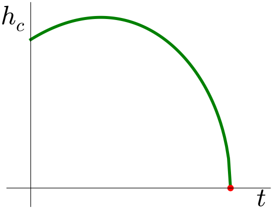
$s^2 > (1-\tau)$:	$p = 0$ only	(strong fields)
$s^2 < (1-\tau)$:	$p = 0$ and $p \neq 0$ possible	(weak fields)

At $h = 0$: $s = 0$, $p \neq 0$ is stable. At $h > 0$ the two states compete.

At a critical value of the field, the AFM arrangement of spins ($p \neq 0$), changes into the FM one ($s \neq 0$, $p = 0$) in a first order transition.

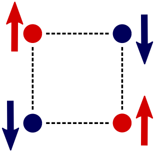
Solutions of Eqs. (1)–(2):



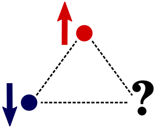


Geometric frustration

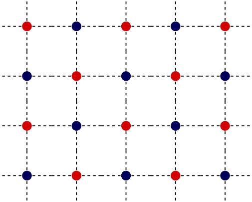
Ferromagnetic ordering is largely insensitive to the local structure of the lattice. On the contrary, *anti*ferromagnetic ordering is:

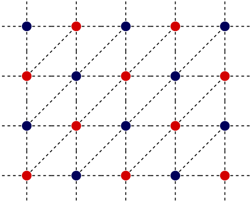


square lattice is *bipartite*



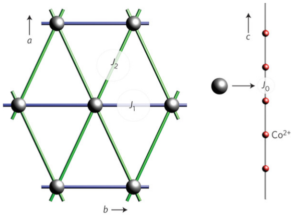
triangular lattice is not

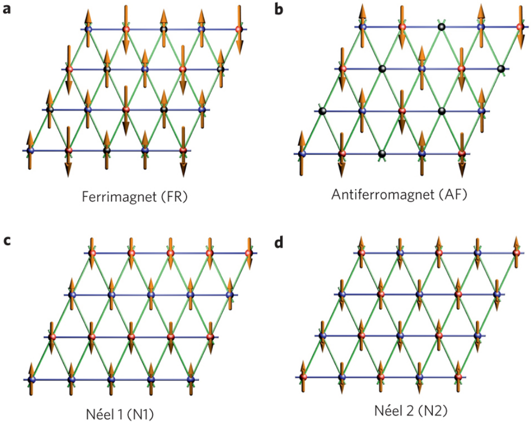




Example: columbite $CoNb_2O_6$

S. Lee, R.K. Kaul & L. Balents, “Interplay of quantum criticality and geometric frustration in columbite”, *Nature Physics* **6**, 702 (2010).





Example: Pyrochlore lattices

A lattice of corner-sharing tetrahedra. Tetrahedra themselves live on an fcc lattice.

$Cd_2Re_2O_7$

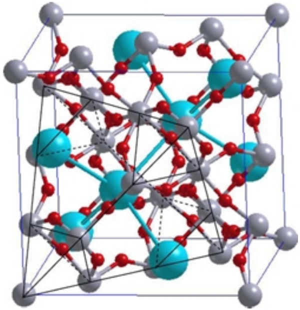


image (c) LPT Toulouse

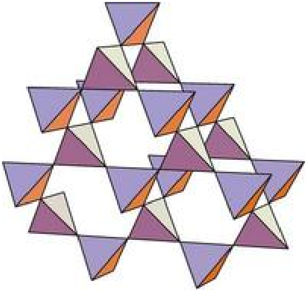


image (c) UCSB

Exotic superconductivity, spin liquids, spin ice, etc etc etc

Magnetic monopoles in spin ice: Dysprosim titanate $Dy_2Ti_2O_7$ and its relatives

Water ice: L. Pauling, 1935: ice rules
Residual entropy
$S/N = k_B \ln 3/2$ as $T \to 0$

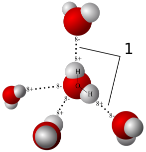


image (c) Wikipedia

Spin ice: C. Castelnovo, R. Moessner & S.L. Sondhi, "Magnetic monopoles in spin ice", *Nature* **451**, 42 (2008)