Frustrated Magnetism

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Lesson Plan*

- Introduction: basic magnetism, empirical signs of frustration
- Classical Ising systems and spin ice
- Heisenberg systems and quantum effects
- A biased materials survey
- Quantum spin liquids

*Subject to change according to my whims
Heisenberg Systems

- If spin-orbit is a weak perturbation, a magnet will have approximate Heisenberg symmetry.
- In 1/2-filled shell, the physics is described to a first approximation by a simple Heisenberg model,

\[ H = \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \]
Heisenberg Systems

\[ H = \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \]

- Unlike the Ising model, this is not intrinsically classical
- However, it turns out that the semi-classical “spin wave” expansion tends to work well even for small \( S \) (unfortunately!)
- We may still ask about the degeneracy in the classical limit
Classical degeneracies

• In the Heisenberg model, the degrees of freedom are continuous, so we cannot count discrete states

• Instead, we can e.g. count the number of continuous parameters that specify the ground state manifold (i.e. its dimension)
Example: triangular lattice

- Spins must sum to zero
Example: triangular lattice

Degrees of freedom: 2
Example: triangular lattice

Degrees of freedom: 2+1
Example: triangular lattice

Degrees of freedom: 2+1
Kagome lattice

Degrees of freedom: 3
Kagome lattice

Degrees of freedom: $3 + 1 + ...$
• Degrees of freedom: ~ N
Luttinger-Tisza Method

- A systematic method for finding classical ground states in many models

- Idea: $|S_i| = 1$ constraint makes minimization of $H$ difficult. So let’s relax this constraint to instead $\sum_i |S_i|^2 = N$ and minimize. Then afterward we can check our solutions for those which satisfy $|S_i| = 1$.

- Minimization of Heisenberg Hamiltonian is then simple because it is quadratic!
Luttinger-Tisza Method

- Lagrange multiplier
  \[
  \tilde{H} = \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j - \lambda \left( \sum_i |\vec{S}_i|^2 - N \right)
  \]

- Minimize
  \[
  \sum_j J_{ij} \vec{S}_j = \lambda \vec{S}_i
  \]

- Bloch theorem
  \[
  \vec{S}_i = \vec{u}_i e^{i\mathbf{q} \cdot \mathbf{r}_i}
  \]

- where \( u_i \) depends only upon the basis site
Luttinger-Tisza Method

- Simplest case: Bravais lattice
- $u_i$ independent of $i$
- get just a band $\lambda(q) = \lambda(-q)$
- It is easy to solve the $|S_i|=1$ constraints

$$\vec{S}_i = \hat{u} \cos[\mathbf{q} \cdot \mathbf{r}_i] + \hat{v} \sin[\mathbf{q} \cdot \mathbf{r}_i] \quad \hat{u} \cdot \hat{v} = 0$$

- This is a coplanar spiral with spins in the $u$-$v$ plane.
- The ground states are just those with minimal $\lambda(q)$
Luttinger-Tisza Method

- Examples

<table>
<thead>
<tr>
<th>lattice</th>
<th>minima</th>
</tr>
</thead>
<tbody>
<tr>
<td>FCC</td>
<td>lines (1d)</td>
</tr>
<tr>
<td>diamond*</td>
<td>surface (2d)</td>
</tr>
<tr>
<td>kagome</td>
<td>flat band (2d)</td>
</tr>
<tr>
<td>pyrochlore</td>
<td>flat band (3d)</td>
</tr>
</tbody>
</table>

*not a Bravais lattice but you can still make it work
What does this tell us?

- Fluctuations typically stronger if degeneracy is larger
- Can easily study thermal fluctuation effects via Monte Carlo
- But the physics is rather different in many ways from the Ising models
- Fluctuations out of the ground state manifold are always possible because they are smooth
- Quantum effects are not really very small at low $T$: formally they contribute to the energy at $O(JS)$ compared to the classical energy $O(JS^2)$. 
Quantum effects?

• Simplest approach is spin-wave theory: $1/S$ expansion for ground state properties

• e.g. triangular lattice

$$Ms = |\langle S_i \rangle| = S - 0.2613 + \frac{0.0055}{S} + \cdots$$

• correction is already pretty small for $S=1$

• Numerics: ED, DMRG, QMC, series

• Best estimates for triangular lattice $M_s \approx 0.205 \pm 0.015$

• compare to spin waves $M_s = 0.24$: pretty good!
Difficulties with spin waves

- Hard to use for $T>0$ properties
- (not too surprising since classical model is nontrivial at $T>0$)
- e.g. specific heat - where is the peak?

Suppressed by quantum fluctuations?

\[ T_p / |\Theta_{CW}| \approx 0.17 \quad S=\infty \]
\[ T_p / |\Theta_{CW}| \approx 0.27 \quad S=1/2 \]
Difficulties with spin waves

• In frustrated magnets, classical degeneracy leads to divergences in linear spin wave theory

• Naively, there are zero energy spin wave modes, in which the spins fluctuate into nearby degenerate ground states

• This leads to vanishing energy denominators in perturbation theory
Difficulties with spin waves

• This can be fixed, formally, by nonlinear spin wave theory in which anharmonic terms are treated self-consistently (see C. Henley and others) - magnet is always ordered at large $S$

• As a result, spin-wave theory is non-analytic, e.g.

$$M_s = \begin{cases} 
S - c \ln (S) & \text{FCC} \\
S - c S^a, & 0 < a < 1/2 \\
S - c S, & \text{kagome/pyrochlore}
\end{cases}$$

• These corrections are large for $S=1/2$

• We may hope for more quantum ground state
What to do?

• We can discuss various methods for a very long time. The fact is that there is no simple recipe for solving this sort of hard problem.

• I will instead discuss some interesting physical examples and some of the ways in which we have understood them
Materials Survey

- B-site spinels: Heisenberg pyrochlores
- A-site spinels: non-geometrical frustration
- $\text{Cs}_2\text{CuCl}_4$: dimensional reduction in an anisotropic triangular system
- $\text{FeSc}_2\text{S}_4$: spin-orbital quantum criticality
What do we look for?

- Is it an insulator?
- Is it a magnet? Curie law
- Signs of frustration
  - $f >> 1$
    - $\Theta_{CW}(\chi)$
    - $T_N$: signs of transition in $\chi$, $C_v$, ...
  - low $T$ entropy, low energy excitations
  - $C_v$, $1/T_1$, ...
- Identify the states
- nature of correlations?
- ordering if it occurs
- Compare with some theoretical expectations
AB$_2$X$_4$ spinels

- One of the most common mineral structures
- Common valence:
  - $A^{2+}, B^{3+}, X^{2-}$
  - $X$ = O, S, Se

Cubic Fd$ar{3}$m
Deconstructing the spinel

- A atoms: diamond lattice
- Bipartite: not geometrically frustrated
Deconstructing the spinel

- B atoms: pyrochlore
- decorate the plaquettes of the diamond lattice
ACr$_2$O$_4$ spinels

- pyrochlore lattice
- S=3/2 Isotropic moment
- X=O spinels: B-B distance close enough for direct overlap
- dominant AF nearest-neighbor exchange
H=0 Susceptibility

- Frustration:

<table>
<thead>
<tr>
<th></th>
<th>Zn</th>
<th>Cd</th>
<th>Hg</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Theta_{CW}$ (K)</td>
<td>-390</td>
<td>-70</td>
<td>-32</td>
</tr>
<tr>
<td>$T_N$ (K)</td>
<td>12</td>
<td>7.8</td>
<td>5.8</td>
</tr>
<tr>
<td>$f$</td>
<td>33</td>
<td>9</td>
<td>6</td>
</tr>
</tbody>
</table>

H. Ueda et al
Degeneracy

- Heisenberg model

\[ H = \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j = \frac{1}{2} \sum_t \left( \sum_{i \in t} \vec{S}_i \right)^2 + \text{const.} \]

- Ground state constraint: total spin 0 per tetrahedron

- Quantum mechanically: not possible
Classical spin liquid

- No LRO (Reimers)
Classical spin liquid

- No LRO (Reimers)
- Dipolar correlations

(Youngblood+Axe, Henley, Isakov et al...)

\[ S^\mu_i = b^\mu_{ab} \]
Classical spin liquid

- Unusual “ring” correlations seen in CdCr$_2$O$_4$. Related?
- does not seem to be pinch point structure
- it has been proposed that other interactions are responsible

Broholm et al
Ordering

- Many perturbations important for ordering:
  - Spin-lattice coupling
  - Further exchange
  - Spin-orbit effects
  - Quantum corrections

ZnCr$_2$O$_4$

CdCr$_2$O$_4$

HgCr$_2$O$_4$

S.H. Lee + many others

JH Chung et al, 2005
Magnetization Plateaus

- Classically: $M = M_s \frac{H}{H_s}$
- Plateau indicates 3:1 structure

H. Ueda et al, 2005/6
Magnetization Plateaus

- Plateau mechanism:
  - spin-lattice coupling favors collinearity
- Order on plateau may be selected by
  - spin-lattice
  - quantum effects (nice application of Ising+XY model)
  - further neighbor coupling
- Prediction: all these mechanisms favor the same state
  - This is due to the high degree of constraints on the 3:1 states - different interactions projected into the 3:1 states give the same “pseudopotential” (familiar from LLL physics)
Magnetization Plateaus

- “R” (Ryuichi) state

HgCr$_2$O$_4$: Matsuda et al, 2007

CdCr$_2$O$_4$: Matsuda et al, 2009
A-site spinels

- Spectrum of materials

CoRh$_2$O$_4$  Co$_3$O$_4$  MnSc$_2$S$_4$  FeSc$_2$S$_4$

MnAl$_2$O$_4$  CoAl$_2$O$_4$  Naively unfrustrated

$s = 5/2$

$s = 3/2$

$s = 2$

Orbital degeneracy

Naively unfrustrated

V. Fritsch et al. PRL 92, 116401 (2004); N. Tristan et al. PRB 72, 174404 (2005); T. Suzuki et al. (2006)

$f = \frac{|\Theta_{CW}|}{T_N}$
Why frustration?

• Roth, 1964: 2nd and 3rd neighbor exchange not necessarily small
• Exchange paths: A-\(x\)-B-\(x\)-B comparable
• Minimal model
• \(J_1\)-\(J_2\) exchange
Ground state evolution

- Coplanar spirals

Spiral surfaces:

- $J_2/J_1 = 0.2$
- $J_2/J_1 = 0.4$
- $J_2/J_1 = 0.85$
- $J_2/J_1 = 20$
Monte Carlo

\[ f = 1 \text{ at } \frac{J_2}{J_1} = 0.85 \]
Phase Diagram

- Entropy and $J_3$ compete to determine ordered state
- Spiral spin liquid regime has intensity over entire spiral surface
Comparison to Expt.

- Diffuse scattering
- Ordered state
  - (qq0) spiral
- Specific heat?

A. Krimmel et al, 2006

agrees with theory for FM $J_1$
Cs$_2$CuCl$_4$

- Spatially anisotropic triangular lattice
- Cu$^{2+}$ spin-1/2 spins

\[
H = \frac{1}{2} \sum_{i,j} \left[ J_{ij} \vec{S}_i \cdot \vec{S}_j - D_{ij} \cdot (\vec{S}_i \times \vec{S}_j) \right]
\]

- couplings:
  \[ J = 0.37 \text{meV} \]
  \[ J' = 0.3J \]
  \[ D = 0.05J \]

R. Coldea et al.
Neutron scattering

- Coldea et al, 2001/03: a 2d spin liquid?

Very broad spectrum similar to 1d (in some directions of k space). Roughly fits power law.

Fit of “peak” dispersion to spin wave theory requires adjustment of J,J’ by 40% - in opposite directions!
Dimensional reduction?

- Frustration of interchain coupling makes it less “relevant”
- First order energy correction vanishes

- Leading effects are in fact $O[(J')^4/J^3]$!
• Frustration of interchain coupling makes it less “relevant”
• First order energy correction vanishes.
• Numerics: $J'/J < 0.7$ is “weak”
Excitations

• In a 1d spin chain, the elementary excitations are spinons.

• You can see that the spinon is like a domain wall.

• It has in this sense a “string”, but this does not confine the spinon because the string’s boundary is just its endpoint.
Excitations

- Build 2d excitations from 1d spinons

- Exchange:
  \[ \frac{J'}{2} (S_i^+ S_j^- + S_i^- S_j^+) \]

- Expect spinon binding to lower inter-chain kinetic energy

- Use 2-spinon Schroedinger equation
Broad lineshape: “free spinons”

- “Power law” fits well to free spinon result
- Fit determines normalization

$J'(k) = 0$ here
Bound state

- Compare spectra at $J'(k)<0$ and $J'(k)>0$:

- Curves: 2-spinon theory with experimental resolution
- Curves: 4-spinon RPA with experimental resolution
Transverse dispersion

Bound state and resonance

Solid symbols: experiment
Note peak (blue diamonds) coincides with bottom edge only for $J'(k)<0$
Spectral asymmetry

Vertical lines: $J'(k)=0$. 
Orbital degeneracy in FeSc$_2$S$_4$

- Chemistry:
  - Fe$^{2+}$: 3d$^6$
  - 1 hole in eg level
  - Spin $S=2$
  - Orbital pseudospin 1/2
  - Static Jahn-Teller does not appear

Refs: PRL 102, 096406 (2009); arXiv:0907.1692
Atomic Spin Orbit

- Separate orbital and spin degeneracy can be split!

\[ H_{SO} = -\lambda \left( \frac{1}{\sqrt{3}} \tau^x \left[ (S^x)^2 - (S^y)^2 \right] + \tau^z \left[ (S^z)^2 - \frac{S(S+1)}{3} \right] \right) \]

- Energy spectrum: singlet GS with gap = \( \lambda \)

- Microscopically,

\[ \lambda = \frac{6\lambda_0^2}{\Delta} \]

- Naive estimate \( \lambda \approx 25K \)
Spin orbital singlet

- Ground state of $\lambda > 0$ term:

$$|S^z=0\rangle - \frac{1}{\sqrt{2}} (|S^z=2\rangle + |S^z=-2\rangle)$$

- Due to gap, there is a stable SOS phase for $\lambda \gg J$. 
Inelastic neutrons show significant dispersion indicating exchange

Bandwidth $\approx 20$K is of similar order as $\Theta_{CW}$ and estimated $\lambda$

Gap (?) 1-2K

Small gap is classic indicator of incipient order

N. Büttgen et al, PRB 73, 132409 (2006)
Exchange

- Heisenberg interactions

\[ H = \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \]  

(\(J_1 - J_2\))

- Rather classical (S=2)
  - favors spiral ground states
  - but competes with spin-orbit interaction
Minimal Model

- Neutron scattering suggests peak close to $2\pi(100)$
- Indicates $J_2 \gg J_1$

$$H_{\text{min}} = J_2 \sum_{\langle\langle ij\rangle\rangle} S_i \cdot \langle S_j \rangle + H_{SO}$$

Expect MFT good in 3+1 dimensions

Quantum Critical Point

- Mean field phase diagram

\[ \frac{\lambda}{J_2} \]

\[ T = 2\pi(100) \text{ AF} \]

\[ \text{Ferro OO} \]

\[ \text{SO singlet} \]

\[ \text{FeSc}_2\text{S}_4 \]

\[ \lambda/J_2 \]
Predictions

- Large $T=0$ susceptibility (estimated) ✓
- Scaling form for $(T_1 T)^{-1} \sim f(\Delta/T)$ ✓
- Specific heat $C_v \sim T^3 f(\Delta/T)$ ✓
- Possibility of pressure-induced ordering
- Magnetic field suppresses order - opposite to dimer antiferromagnet (e.g. TlCuCl$_3$)
Next: Quantum Spin Liquids
Some other interesting materials