Degeneracy Breaking in Some Frustrated Magnets

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*Itzykson meeting*, Saclay, June 2006
Outline

• Motivation: Why study frustrated magnets?
• Chromium spinels and magnetization plateau
• Quantum dimer model and its phase diagram
• Constrained phase transitions and exotic criticality
Degeneracy “breaking”

• Origin of (most) magnetism: Hund’s rule
  – Splitting of degenerate atomic multiplet
  – Degeneracy “quenches” kinetic energy and makes interactions dominant

• Degeneracy on a macroscopic scale
  – Macroscopic analog of “Hund’s rule” physics
  – Landau levels $\Rightarrow$ FQHE
  – Large-U Hubbard model $\Rightarrow$ High-$T_c$?
  – Frustrated magnets $\Rightarrow$
    • Spin liquids ??
    • Complex ordered states
    • Exotic phase transitions ??
Spin Liquids?

- Anderson: proposed RVB states of quantum antiferromagnets

\[ |\psi\rangle = + + \ldots \]

- Phenomenological theories predict such states have remarkable properties:
  - topological order
  - deconfined spinons

- There are now many models exhibiting such states
Quantum Dimer Models

- Models of “singlet pairs” fluctuating on lattice (can have spin liquid states)

\[ \mathcal{H}_{QDM} \approx V \sum_P (|\bigcirc\bigcirc\rangle\langle\bigcirc\bigcirc| + |\bigcirc\bigcirc\rangle\langle\bigcirc\bigcirc|) - K \sum_P (|\bigcirc\bigcirc\rangle\langle\bigcirc\bigcirc| + |\bigcirc\bigcirc\rangle\langle\bigcirc\bigcirc|) \]

- Constraint: 1 dimer per site.
- Construction problematic for real magnets
  - non-orthogonality
  - not so many spin-1/2 isotropic systems
  - dimer subspace projection not controlled
  - We will find an alternative realization, more akin to spin ice
Chromium Spinels

\[ \text{ACr}_2\text{O}_4 \quad (A=\text{Zn}, \text{Cd}, \text{Hg}) \]

cubic \text{Fd}3\text{m}

- Spins form pyrochlore lattice
- Antiferromagnetic interactions

\[ \Theta_{\text{CW}} = -390\text{K}, -70\text{K}, -32\text{K} \]

for \( A=\text{Zn}, \text{Cd}, \text{Hg} \)

\[ \text{Cr}^{3+}(\text{d}^3; S=3/2) \]

- Spin \( S=3/2 \)
- No orbital degeneracy
- Isotropic

Takagi group
Pyrochlore Antiferromagnets

- Heisenberg

\[ H = \sum \frac{J}{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j = \frac{J}{2} \sum_t \left( \sum_{i \in t} \vec{S}_i \right)^2 \]

- Many degenerate classical configurations

- Zero field experiments (neutron scattering)
  - Different ordered states in ZnCr$_2$O$_4$, CdCr$_2$O$_4$
  - HgCr$_2$O$_4$?

- What determines ordering not understood

\[ \Theta_{CW} = -390K, -70K, -32K \text{ for } A = \text{Zn, Cd, Hg} \]
Magnetization Process

- Magnetically isotropic
- Low field ordered state complicated, material dependent
- Plateau at half saturation magnetization in 3 materials

H. Ueda et al, 2005
HgCr$_2$O$_4$ neutrons

- Neutron scattering can be performed on plateau because of relatively low fields in this material.

H. Ueda et al, unpublished

- We will try to understand this ordering
Collinear Spins

- Half-polarization = 3 up, 1 down spin?
  - Presence of plateau indicates no transverse order

- Spin-phonon coupling?
  - classical Einstein model
    - Penc et al

\[ H = k u_{ij}^2/2 - g u_{ij} \vec{S}_i \cdot \vec{S}_j \rightarrow -J b (\vec{S}_i \cdot \vec{S}_j)^2 \]

  - effective biquadratic exchange favors collinear states
  - But no definite order

- “Order by disorder”
  - in semiclassical S→∞ limit, thermal and quantum fluctuations favor collinear states (Henley…)
  - this alone probably gives rather narrow plateau if at all

large magnetostriction

H. Ueda et al
3:1 States

- Set of 3:1 states has thermodynamic entropy
  - Less degenerate than zero field but still degenerate
  - Maps to dimer coverings of diamond lattice

- Effective dimer model: **What splits the degeneracy?**
  - Classical:
    - further neighbor interactions
    - Lattice coupling beyond Penc et al?
  - Quantum?
Ising Expansion

• Strong magnetic field breaks SU(2) → U(1)
• Substantial polarization: $S_i^\perp < S_i^z$
• Formal expansion in $J_\perp/J_z$ reasonable (carry to high order)

$$\mathcal{H}_0 = \frac{J_z}{2} \sum_t \left[ (S_t^z - h)^2 - h^2 \right] - J_z \sum_i (S_i^z)^2$$
$$\mathcal{H}_1 = \frac{J_\perp}{2} \sum_{\langle ij \rangle} (S_i^+ S_j^- + h.c.)$$

3:1 GSs for $1.5<h<4.5$

• Obtain effective hamiltonian by DPT in 3:1 subspace
  - First off-diagonal term at 9th order! [(6S)th order]
  - First non-trivial diagonal term at 6th order!
Effective Hamiltonian

\[ \mathcal{H}_{\text{eff}} = \alpha^6 J_z \sum_P \hat{E}_P - c\alpha^9 J_z \sum_P \left( \begin{pmatrix} | & | \\ \downarrow & \downarrow \end{pmatrix} \begin{pmatrix} | & | \\ \downarrow & \downarrow \end{pmatrix} \right) + \text{h.c.} \]

\[ \alpha = J_\perp / J_z \]

Diagonal term:

\[ \hat{E}_P \]

\[ \begin{array}{c|c|c}
 \text{State} & V_1 & V_2 = \\
 \hline
 \uparrow \uparrow \uparrow \uparrow & \frac{3s^4 (98304s^5 - 139648s^4 + 79136s^3 - 22040s^2 + 3006s - 165)}{32(2s-1)(4s-1)^5(8s-3)^2(12s-5)} & \frac{s^3 (256s^3 - 51s + 9)}{32(4s-1)^3(8s-3)^2} \\
 \uparrow \uparrow \uparrow \uparrow \uparrow & V_2 = & \frac{s^4 (272s^2 - 136s + 15)}{16(4s-1)^5(8s-3)^2} \\
 \hline
 \end{array} \]

- Two independent techniques to sum 6\textsuperscript{th} order DPT
- Agrees exactly with large-s calculation (Hizi+Henley) in overlapping limit
Quantum Dimer Model on diamond lattice

\[ \mathcal{H}_{\text{QDM}} \approx V \sum_P (|\bigcirc\rangle\langle\bigcirc| + |\bigcirc\rangle\langle\bigcirc|) - K \sum_P (|\bigcirc\rangle\langle\bigcirc| + |\bigcirc\rangle\langle\bigcirc|) \]

- Expected phase diagram (various arguments)
  
  \[ v = \frac{V}{K} \approx -2.3 \]

  \[ -2.3 \quad S=1 \quad 0 \quad 1 \quad v \]

  - Maximally "resonatable" R state
  - U(1) spin liquid
  - "frozen" state

- Interesting phase transition between R state and spin liquid! Will return to this.

Quantum dimer model is expected to yield the R state structure

Caveat: other diagonal terms can modify phase diagram for large negative \( v \)
R state

- Unique state saturating upper bound on density of resonatable hexagons
- Quadrupled (simple cubic) unit cell
- Still cubic: P4\(_3\)32
- 8-fold degenerate

- Quantum dimer model predicts this state uniquely.
Is this the physics of HgCr$_2$O$_4$?

• Probably not:
  – Quantum ordering scale \( \sim |V| \sim 0.02J \)
  – Actual order observed at \( T \gtrsim T_{\text{plateau}}/2 \)

• We should reconsider classical degeneracy breaking by
  – Further neighbor couplings
  – Spin-lattice interactions
    • C.f. “spin Jahn-Teller”: Tchernyshyov et al

Considered identical distortions of each tetrahedral “molecule”
Einstein Model

• Site phonon

\[ \delta J_{ij} = -\gamma J e_{ij} \cdot (u_i - u_j) \]

• Optimal distortion:

\[ u^*_j \propto - \sum_{i \in N(j)} \left( S_i \cdot S_j \right) e_{ij} \]

• Lowest energy state maximizes \( u^* \):

• “bending rule”
Bending Rule States

• At 1/2 magnetization, only the R state satisfies the bending rule globally
  - Einstein model predicts R state!

• Zero field classical spin-lattice ground states?

![Diagram showing collinear states with bending rule satisfied for both polarizations]

<table>
<thead>
<tr>
<th>Unit cells</th>
<th>states</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>12</td>
</tr>
<tr>
<td>16</td>
<td>36</td>
</tr>
<tr>
<td>32</td>
<td>82</td>
</tr>
<tr>
<td>64</td>
<td>216</td>
</tr>
</tbody>
</table>

• ground state remains degenerate

- Consistent with *different* zero field ground states for A=Zn,Cd,Hg
- Simplest “bending rule” state (weakly perturbed by DM) appears to be consistent with CdCr$_2$O$_4$

*Chern et al*, cond-mat/0606039
Constrained Phase Transitions

- Schematic phase diagram:

- Local constraint *changes* the nature of the “paramagnetic” state
  - Youngblood+Axe (81): *dipolar correlations* in “ice-like” models

- Landau-theory *assumes* paramagnetic state is disordered
  - Local constraint in many models implies *non-Landau classical criticality*

  Bergman *et al*, PRB 2006
Dimer model = gauge theory

- Can consistently assign direction to dimers pointing from A → B on any bipartite lattice

\[ \vec{E} = \pm S^z \]

- Dimer constraint \( \Rightarrow \) Gauss’ Law

\[ \left( \text{div } \vec{E} \right)_i = \varepsilon_i = \pm 1 \]

- Spin fluctuations, like polarization fluctuations in a dielectric, have power-law dipolar form reflecting charge conservation
A simple constrained classical critical point

- Classical cubic dimer model
  \[ n_{r,r'} = \begin{cases} 
  1 & \text{dimer} \\
  0 & \text{no dimer} 
\end{cases} \]

- Hamiltonian
  \[ H = V \sum_r (-1)^{\tilde{z}} n_{r,r+\tilde{z}} \]

- Model has unique ground state – no symmetry breaking.
- Nevertheless there is a continuous phase transition!
  - Analogous to SC-N transition at which magnetic fluctuations are quenched (Meissner effect)
  - Without constraint there is only a crossover.
Numerics (courtesy S. Trebst)

Specific heat

“Crossings”
Conclusions

- Quantum and classical dimer models can be realized in some frustrated magnets
  - This effective model can be systematically derived by degenerate perturbation theory
- Spin-lattice coupling probably is dominant in HgCr$_2$O$_4$, and a simple Einstein model predicts a unique and definite state (R state), consistent with experiment
  - Probably spin-lattice coupling plays a key role in numerous other chromium spinels of current interest (multiferroics).
- Local constraints can lead to exotic critical behavior even at classical thermal phase transitions.
  - It would be interesting to explore the possibility of observing such exotic criticality in a “spin ice” type material