Disorder as a probe for spin liquids

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Physics Colloquium @ IIT Madras

17 January 2018
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Matter exhibits different phases

- Matter at equilibrium usually organize themselves into well-defined phases that are distinguished by macroscopic properties.
  - Solids, liquids and gases, superconductors, magnets, · · ·
- Solids are rigid but liquids/gases flow

Typically, low temperature states have less symmetry than the Hamiltonian.

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Example of crystals

- Lower symmetry (ONLY discrete translations+rotations)
- Easier to cut crystal along cleavage planes
- Internal pattern often appears at surface (angles representative of the internal geometry)
- Specific heat $C_v \sim T^3$ for low $T$ due to acoustic phonons
Magnetism inherently quantum phenomenon (Bohr-Van Leeuwen theorem) \([\mathbf{p} \rightarrow \mathbf{p} - q\mathbf{A}\) leaves \(\mathcal{Z}\) unchanged]\)

Essential components—permanent (atomic) magnetic moments and \([\mathbf{p}, \mathbf{A}] \neq 0\)

Exchange energy \(E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j\) due to Coulomb interactions and Pauli exclusion.

- Magnetism in insulators

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Ising model: Simple model for ferromagnetism

\[ H = -J \sum_{\langle ij \rangle} S_i S_j \] where \( S_i = \pm 1 \) on sites of a square lattice

\[ T_c = \frac{2J}{\ln(1+\sqrt{2})} \approx 2.269J \] (Lars Onsager, 1944)

- For \( T > T_c \) paramagnet, \( T < T_c \) ferromagnet (breaks \( S_i \rightarrow -S_i \) symmetry spontaneously), \( T = T_c \) continuous phase transition
Experimental discovery of antiferromagnetism

- MnO data ⇒ Shull and Smart, 1949 using neutron scattering
- Historical note: ferromagnets known from at least 600 B.C.!
- Most magnetic materials have antiferromagnetic interactions.
What is a spin liquid?

- **Spin solid** — e.g. A ferromagnet or an antiferromagnet

Fig shows dispersion in La$_2$CuO$_4$ [from Coldea et. al, PRL 86, 5377 (2000)] – shows existence of magnons

- **Spin gas** — e.g. an uncorrelated paramagnet
- **Spin liquid** — strongly “correlated” paramagnet
These break no symmetries of the underlying Hamiltonian but possess *completely different* excitations from conventional ordered states.

- \( H = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j \) with \( S = 1/2 \) spins in 1D
- Excitations not magnons \( (S = 1) \) but spinons \( (S = 1/2) \)
Search for new phases in magnets

Smoking gun signatures of fractionalization not yet found for quantum spin liquids in $d \geq 2$ (unlike $\nu = 1/3$ FQH where shot noise exps gave $e^* = e/3$ for quasiparticles)

Simple classical models: $H = \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j$ can present liquidity and fractionalization at low-$T$! of experimental relevance for “large” $S \rightarrow$ classical regime
Frustration: a possible route

What would probably favor a low-$T$ classical SL phase?
Frustration.

$$H = \sum_{i,j} J_{ij} S_i S_j$$

But still no guarantee: plethora of ways for conventional order.
(Weak) Diagnostics

- No magnetic ordering even well below $|\Theta_{CW}| \sim JzS^2$
  unlike unfrustrated magnets
- $T_f \ll T \ll |\Theta_{CW}|$—spin liquid regime (Ramirez)

**Frustration parameter** $f = |\Theta_{CW}| / T_f$

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What causes glassiness?
Which classes of spin-liquids exist?
Some experimental examples

1. **SCGO (SrCr$_{9-x}$Ga$_{3+x}$O$_9$)**
   - **Magnetic lattice**: Pyrochlore slab (quasi two-dim lattice)
   - Heisenberg spins ($S = 3/2$) with $\Theta_{CW} \approx -500$ K.

2. **Spin Ice materials Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7**
   - **Magnetic lattice**: Pyrochlore lattice (3 D)
   - Ising spins with $\Theta_{CW} \approx 2$ K (ferromagnetic!)

3. **herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2**
   - **Magnetic lattice**: Kagome lattice (2 D)
   - Heisenberg spins ($S = 1/2$) with $\Theta_{CW} \approx -300$ K [no ordering till $T \sim mK$]

4. **κ-ET κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3**
   - **Magnetic lattice**: Triangular lattice (2 D)
   - Heisenberg spins ($S = 1/2$) with $\Theta_{CW} \approx -400$ K
   - $C \sim \gamma T$ and $\chi \sim const$ at low $T$
Disorder inevitably present in solid state experiments (substitution, random strains, vacancies, .....)

- Can be useful as a probe for a phase e.g. nature of the superconducting phase Davis et. al.(2000)

Response to defects and new states emerging out of disorder.
In a nutshell

- Disorder induced spin textures with fractionalized moments (1/2 or 1/3) in a class of classical spin liquids.

- Development of hybrid field theory to treat disorder in a class of spin liquids.

- Topological spin glass in diluted spin ice.
  - First microscopic Hamiltonian exhibiting partial freezing and coexistence of spin liquid + glass
Coulomb spin liquids I

\[ H = J \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j = \frac{J}{2} \sum_{\Box} (\vec{S}_\Box)^2 \]

Experimentally relevant: Dy\(_2\)Ti\(_2\)O\(_7\), Ho\(_2\)Ti\(_2\)O\(_7\), Er\(_2\)Ti\(_2\)O\(_7\), SrCr\(_8\)Ga\(_4\)O\(_{19}\)

Units must share corners and occupy bipartite lattice

Local constraints of $S_{\square} = 0$ become $\nabla \cdot \vec{B} = 0$

Flippable loops have low average flux $\tilde{B}$

$\mathcal{Z} = \int \mathcal{D}\tilde{B} \exp \left( -\frac{K}{2} \int d^d x \tilde{B}^2 - \beta J \int d^d x (\nabla \cdot \tilde{B})^2 \right)$

Henley (2004); Isakov, Moessner, Sondhi (2004)

$\langle \tilde{B}_\mu(-\mathbf{k}) \tilde{B}_\nu(\mathbf{k}) \rangle = \frac{1}{K} \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2 + \xi - 2} \right)$ where $\xi \sim \sqrt{\beta J/K}$. 

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Spin ice and Dumbbell model I

\[ H = \frac{J}{3} \sum \langle ij \rangle S_i S_j + \frac{\mu_0 \mu^2}{4\pi} \sum_{ij} \left( \frac{\hat{e}_i \cdot \hat{e}_j}{|r_{ij}|^3} - \frac{3(\hat{e}_i \cdot r_{ij})(\hat{e}_j \cdot r_{ij})}{|r_{ij}|^5} \right) S_i S_j \]

- \( J = -1.56 \) K, \( D = \frac{\mu_0 \mu^2}{4\pi a^3} = 1.41 \) K for \( \text{Ho}_2\text{Ti}_2\text{O}_7 \)
- Replace dipoles by dumbbells that live on the ends of the dual diamond lattice. Charge \( \pm \mu / a_d \). Castelnovo, Moessner, Sondhi (2008)

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Spin ice and Dumbbell model II

- $H = \frac{J}{3} \sum_{\langle ij \rangle} S_i S_j + D a^3 \sum_{(ij)} \left( \frac{\hat{e}_i \cdot \hat{e}_j}{|r_{ij}|^3} - \frac{3(\hat{e}_i \cdot \hat{r}_{ij})(\hat{e}_j \cdot \hat{r}_{ij})}{|r_{ij}|^5} \right) S_i S_j$

- Value of the charge $q_i = \pm \mu / a_d$;
  $Q_\alpha = q_{1\alpha} + q_{2\alpha} + q_{3\alpha} + q_{4\alpha}$

- $V(r_{\alpha\beta}) = \frac{\mu_0}{4\pi} \frac{Q_\alpha Q_\beta}{r_{\alpha\beta}}$ if $\alpha \neq \beta$; else $\frac{1}{2} v_0 Q_\alpha^2$ “regularized” Coulomb interaction

- $v_0 \left( \frac{\mu}{a_d} \right)^2 = \frac{J}{3} + \frac{4}{3} \left( 1 + \sqrt{\frac{2}{3}} \right) D$

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Consequences of dipolar interactions

Glassiness in spin liquids?

Exp in $\text{Cu}_3\text{Ba(VO}_5\text{H)}_2$ which is $S = 1/2$ kagome antiferromagnet.

No sign of transition to magnetic long range order.

Coexistence of both dynamical and small frozen moments from $\mu$SR without any phase separation.
Introducing disorder

At defective tetrahedra, $S \neq 0$ even at $T = 0$.

However, dipoles are energetically favored compared to monopoles. $\delta = \frac{4\sqrt{2}D}{3\sqrt{3}}$

Impurity monopoles cheaper and dominate below $T \sim -\frac{\Delta - \delta}{\ln x}$.

Can use effective theory (of Sen, Moessner, Sondhi (2013)) to calculate disorder-averaged spin correlators above glass transition.
Disorder effects

Phase diagram of diluted spin ice

\[ \Theta_w \sim \Delta = \frac{2J}{3} + \frac{8}{3} \left( 1 + \sqrt{\frac{2}{3}} \right) D; \quad \delta = \frac{4\sqrt{2D}}{3\sqrt{3}}; \quad T_\delta = \frac{\delta - \Delta}{\ln x}; \quad T_c \sim 0.95 Dx \]

- **Glass**: Freezing of ghost spins below \( T_c(x) \).
- **Liquid**: Pinch points persist below \( T_c(x) \).
- **Interplay** shows up in gradual but complete loss of Pauling entropy as \( T \) is lowered below \( T_c(x) \).

Very recent exp. on Tb\(_2\)Hf\(_2\)O\(_7\) (Sibille et. al, Nat. Commun. (2017)) show signatures of both liquidity and glassiness.
Excitations: $\vec{S} \neq 0$ can be induced:

- Thermally.
- From quenched disorder. Robust down to $T = 0$!

Hybrid field theory\(^1\):

- Orphan fractionalization (unusual in classical systems) $\rightarrow$ direct signature of liquidity.

Orphans in $\text{SrCr}_{9p}\text{Ga}_{12-9p}\text{O}_{19}$

Ideal SCGO unrealizable

Fractionalized $1/2$ orphans!

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Replace $\vec{S}_i$ by $\vec{\phi}_i$ which are \textbf{soft} spins.

$Z \propto \int \mathcal{D}\vec{\phi} \exp (-F)$, where

$$F = \frac{1}{2} \sum_i \rho_i \vec{\phi}_i^2 + \frac{\beta J}{2} \sum_{\Delta, \Box} (\vec{\phi}_\Delta, \vec{\phi}_\Box - \vec{h}/2J)^2$$

Orphan spins need to be retained as fixed-length vectors.

These couple to the bulk spin liquid represented by $\vec{\phi}$. 

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New types of spin liquids

More diversity than Coulomb spin liquids!!

<table>
<thead>
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<th>type</th>
<th>lattice</th>
<th>pinch-points?</th>
<th>$\xi$</th>
<th>orphan fract.</th>
<th>orphan inter. ($T=0$)</th>
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<tr>
<td>Coulomb bipartite</td>
<td>corner-sharing</td>
<td>Yes</td>
<td>$1/\sqrt{T}$</td>
<td>1/2</td>
<td>long-ranged</td>
</tr>
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<td>edge-sharing</td>
<td>Yes</td>
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<td>1/3</td>
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<tr>
<td>non-Coulomb</td>
<td>corner-sharing</td>
<td>No</td>
<td>$1/\sqrt{T+\gamma}$</td>
<td>1/2</td>
<td>short-ranged</td>
</tr>
</tbody>
</table>

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Disorder as a probe for spin liquids
The maximally frustrated honeycomb lattice

“Edge-sharing octahedra” on triangular lattice

\[ H = \frac{J}{2} \sum_\bigodot (\vec{S}_\bigodot)^2, \]
\[ J = J_1 = 2J_2 = 2J_3 \]

Fractionalized $\frac{1}{3}$ orphans with long ranged Coulomb interactions at $T = 0$!

Further interesting lattices
Now, corner-sharing, but non-bipartite premedial lattice.

\[ H = \frac{J}{2} \sum_{\square} (\vec{S}_{\square})^2 \]

\[ H = \frac{J}{2} \sum_{\triangle} (\vec{S}_{\triangle})^2 \]

Monte Carlo: Heisenberg and Ising

Ruby lattice

H'berg, $\beta J = 20$, $N = 7776$ spins

Ising, $T = 0$, $N = 10584$ spins

Qualitative agreement with large $-n$. No signs of ordering.

No pinch points!
Orphans: non-trivial correlations

Orphans fractionalize $\frac{1}{2}$ at low-$T$.

⇒ correlations of pure system are not of a trivial paramagnet.
Play many roles: may be a nuisance in some cases but may be really interesting in others
Topological glass in spin ice
Orphan physics in classical spin liquids
Examples beyond the classical Coulomb spin liquids

Thanks for your attention