Experimental Candidates of Quantum Spin Liquids

CURRENT STATUS

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Experimental Candidates of Quantum Spin Liquids

Where do they stand

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OUTLINE

• What is a Quantum Spin Liquid?
• Experimental candidates
  Why they were considered as QSL
  Are their alternative explanations for observed behavior?
• Discussion: What would be convincing of a Quantum Spin Liquid? ---Challenge for experiments and numerical studies
What is a Quantum Spin Liquid?

- A simple-minded perspective
- Liquid: Condensed Yet Fluid (Dynamical) Phase
- Quantum Liquid: Coherent Quantum Dynamics
- Not necessarily Macro Quantum Phenomena?
- Translational Motion of Atoms: (He)

Phase Diagram
Assembly of Interacting Quantum Spins

• Nearly ideal paramagnet is like a gas
• Ordered FM/AFM is like a solid
• In between one can have a spin-liquid
  Strong Short-range order, yet not frozen
  Not FM/AFM/Helical/Spiral/Spin-glass
• Is there a Quantum Spin Liquid?
  Must exist down to low temperatures possibly T=0
  Likely associated with small spin (1/2 OR 1)
Spin Liquids


Spin Ice are examples of classical spin-liquids where theory and experimental signatures are on much firmer footing.

We focus on Quantum Spin Liquids
Exclusions from QSL

Pair of AFM coupled spins form fully entangled rotationally invariant singlet states
--cluster of even number of spin-1/2
--which decouple from the rest

Let us exclude systems that by material geometry decouple into (adiabatically connect to) finite systems (decoupled spin clusters)

Cu(NO3)2, CuHpCl, CaV4O9, SrCu(BO3), …

Note: Excitations will be extended/dispersive (Triplons) (BEC)
Shastry-Sutherland Lattice SrCu(BO3)
Kageyama et al

Exact singlet GS with no broken symmetry
Lots of interesting behavior but not what we are looking for
More Exclusions

• Valence Bond Solids that Break Lattice Symmetry (analogous to Molecular Solids)
  --- End up by SSB as decoupled spin clusters
  --Must have a finite-T transition

Majumdar-Ghosh Models and spin-Peierls Systems (many quasi-one-d examples)

Higher Dimensional Examples?
VBS Pinwheel in a Kagome system

Nature Physics
Matan et al
Distortions are present upto high $T$
Also exclude
Valence Bond States that mesh with the lattice

AKLT Models
Spin-3/2 on
Honeycomb

Haldane Chains,
Spin-Ladders
Quantum Spin Liquids
What are theorists looking for?

• Resonating Valence Bonds (Anderson, Fazekas and Anderson 1975)
• Allows delocalization of fractional excitations – spinons (Fermions/Bosons) (Sachdev, ……)
• Quantum Dimer Models – Reduced Hilbert Space (Kivelson-Rokhsar-Sethna, Moessner-Sondhi, Misguich et al, ….)– Short-range \( \Rightarrow \) Gapped Topological Spin Liquids
• Gapless, Algebraic Spin-Liquids – Examples: Flux phases, Gutzwiller Projected Fermi Sea or BCS states (Affleck-Marston, …. PA Lee, …. MPA Fisher)
• Gauge Structure, Chiral spin-liquid, ………
Interesting properties of RVB states ("spin liquids")

Topological order
And Degeneracy

Free ("deconfined")
$S=1/2$ spinon excitations

Example of confined spinons in valence-bond ordered state
Where can one find such QSL?

- **Low-dimensions** (One-D Bethe Ansatz solution satisfies all criterion- motivation for Anderson)— **we will exclude that too**--- 1D is special in too many ways— AFM LRO can only be Algebraic, Domain walls are points, ...

- **Low spin** (spin-half)

- **Geometric Frustration** – possibly leading to extensive ground state degeneracy at classical level

- **Itinerant systems** (Near Mott limit): Ring Exchanges
Handful of Proposed Candidates
many more lately

- He3 Adsorbed on Graphite
- Cs-Cu-Cl
- Variety of Organic Molecular Crystals
- Herbertsmithite Kagome Materials
- 3D Hyperkagome system Na-Ir-O
- Spin-one system Ni-Ga-S
- Spin-Orbital Liquid Fe-Sc-S

a. Frustration parameter   b. Gap/ power-laws in temperature
Helium-3—Nuclear Magnet

- In Bulk (BCC) nuclear moments known to form uudd phase as a result of ring exchanges
- Very high purity and no spin-orbit coupling
- For He-3 absorbed on graphite --- second layer forms a commensurate triangular-lattice
- Shows no long range order and no spin-gap

Osheroff, Godfrin, …., Greywall, … Fukuyama
Gapless spin-liquid?

Specific Heat and Susceptibility of second layer Helium-3 on Graphite
---Review by Fukuyama (JPSJ)
MSE Model Misguich et al PRL 1998
Two types of sites: Elser

Kagome-Triangular Lattice with Kagome-sites more mobile

Susceptibility can be explained quantitatively by a 2 component system
Kagome system may form a gapped spin-liquid
Weakly coupled spins also form a triangular lattice
Very Well Characterized
Layered Triangular
Know Js from high-field
Has LRO ---
But shows exotic physics as well

Experimental Realization of a 2D Fractional Quantum Spin Liquid

Balents and Starykh have convincingly argued that the spin-liquid physics is essentially one-d (spinons are confined in chains)

$$J_2/J_1 = 1/3$$

Series exp. Zheng et al
Careful neutron spectroscopy may well reveal a lot more about QSL (even in Cuprates)

Anomalous High-Energy Spin Excitations in $\text{La}_2\text{CuO}_4$

N. S. Headings, S. M. Hayden, R. Coldea, and T. G. Perring

Intermediate $U/t$

Non quasi-particle dynamics is a hallmark of QSLs
Variety of organic molecular solids

Molecular Dimers in Triangular Geometry

Held together by various ligands

\[ \kappa-(\text{BEDT-TTF})_2X \]

Molecular Solid Susceptibility well described by Heisenberg Model with $J=250$ K but no sign of LRO at 32 mK

Zheng et al HTE
Gapless spin-liquid with spinon fermi-surface?
Caused by finite t/U (O. Motrunich)

- Not fully insulating, U is B-AB splitting (small)
- Susceptibility, 1/T_1, specific heat
- Can one observe the spinon FS???
- What about thermal conductivity?
- Are other degrees of freedom? Impurities?
Highly Mobile Gapless Excitations in a Two-Dimensional Candidate Quantum Spin Liquid

Is this a better candidate for a QSL? More Insulating, Linear kappa
Herbertsmithites (Helton et al) structurally perfect kagome planes

J=200K, no LRO 100mK, Large chi, field-dependent specific heat
Is Curie impurity causing upturn?

Rigol+RRPS

c=0.04 fits to 0.3 J

Misguich+sindzingre

FM CW constant 6.5K
Fits to 0.15 J
Impurities dilute the planes (NMR) (Bert and Mendels JPSJ)

Intersite substitution exceeds 6%
U(1) Dirac Spin-liquid? (Y. Ran et al)

After subtracting for impurities Susceptibility and specific heat can be fitted to power-laws as expected for a U(1) Dirac spin-liquid

What is the ground state of KLHM?

Or a Valence Bond Glass?
RRPS (PRL)
Dimers freeze around impurities
Dommange (Mila) et al
VB Glass can explain Dimer-like form factors (deVries et al) and scaling of susceptibility (Helton et al) Assuming power-law distribution of Js
Other systems
Large frustration parameter, power-law C/T

3D Hyperkagome (Takagi)

Spin-one system (Nakatsuji)
Summary

• **GOOD NEWS**: Experimental groups (especially in Japan) are actively searching for QSL
• Many more materials with large frustration parameter and power-law specific heat are likely to show up.
• What is the best way to identify QSL?
• Are there smoking-gun signatures?
• Are QSL truly robust to disorder, Lattice and other degrees of freedom?
The End