

Experimental Candidates of Quantum Spin Liquids

CURRENT STATUS

RRP Singh

UC DAVIS

Experimental Candidates of Quantum Spin Liquids

Where do they stand

RRP Singh

UC DAVIS



OUTLINE

- What is a Quantum Spin Liquid?
- Experimental candidates

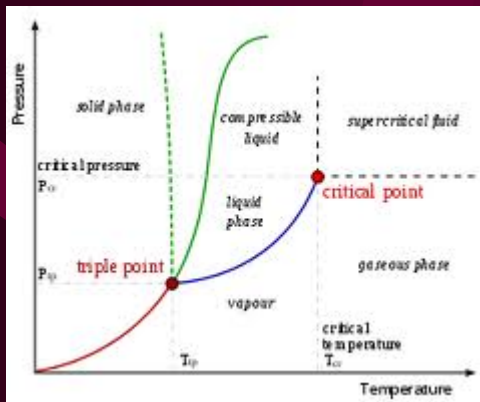
Why they were considered as QSL

Are there alternative explanations for observed behavior?

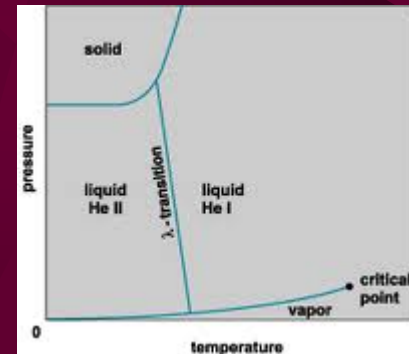
- Discussion: What would be convincing of a Quantum Spin Liquid? ---Challenge for experiments and numerical studies

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

Classical and QSL L. Balents Nature 464 199 (2010)

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

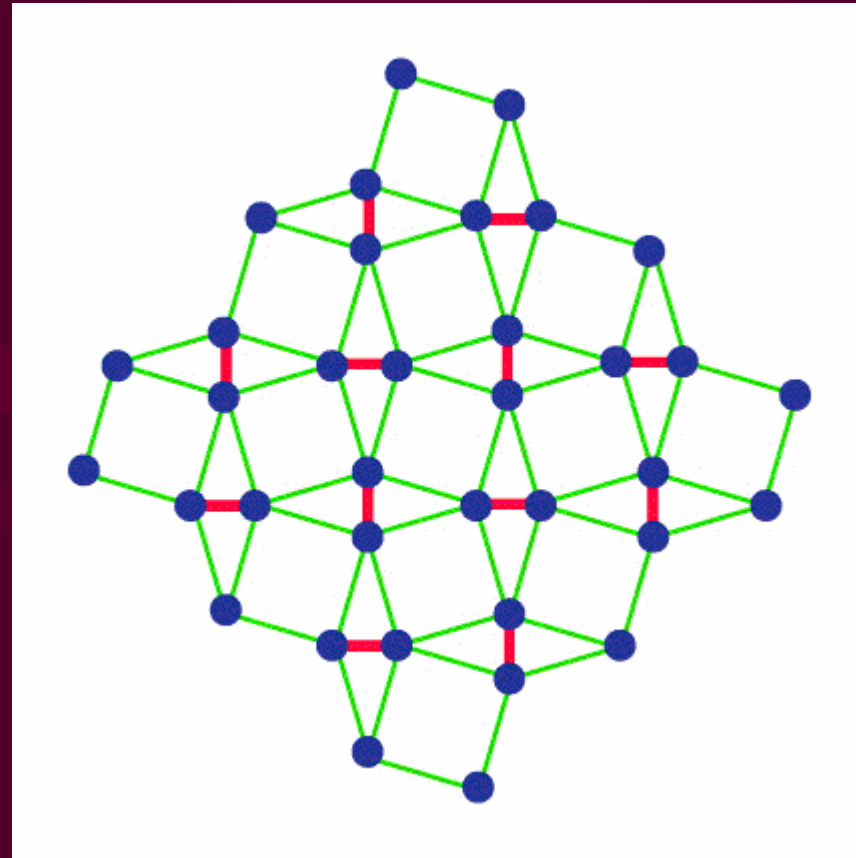
--which decouple from the rest

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

$\text{Cu}(\text{NO}_3)_2$, CuHpCl , CaV_4O_9 , $\text{SrCu}(\text{BO}_3)$,.....

Note: Excitations will be extended/dispersive (Triplons) (BEC)

Shastry-Sutherland Lattice $\text{SrCu}(\text{BO}_3)$
Kageyama et al



Exact singlet GS with no broken symmetry

Lots of interesting behavior but not **what we are looking for**

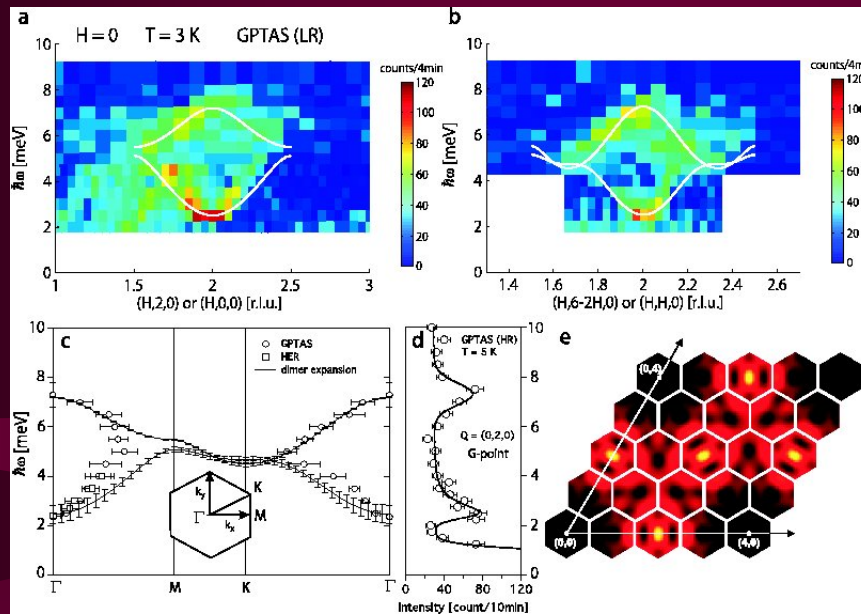
More Exclusions

- **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

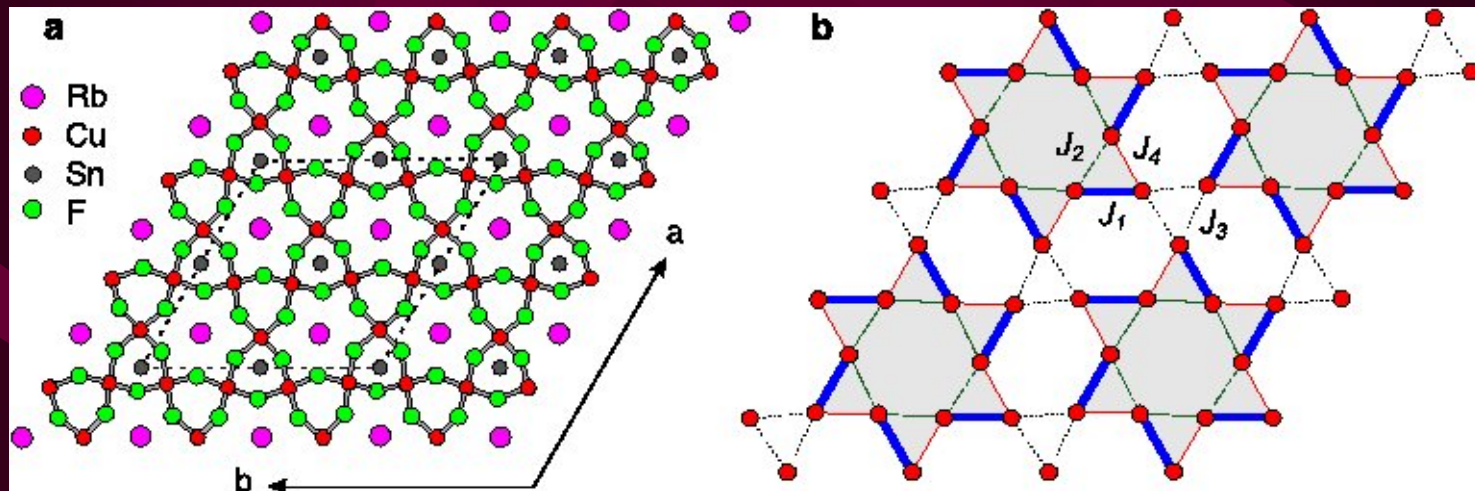
Majumdar-Ghosh Models and spin-Peierls Systems (**many quasi-one-d examples**)

Higher Dimensional Examples?

VBS Pinwheel in a Kagome system



Nature Physics
 Matan et al
 Distortions are
 present upto high T



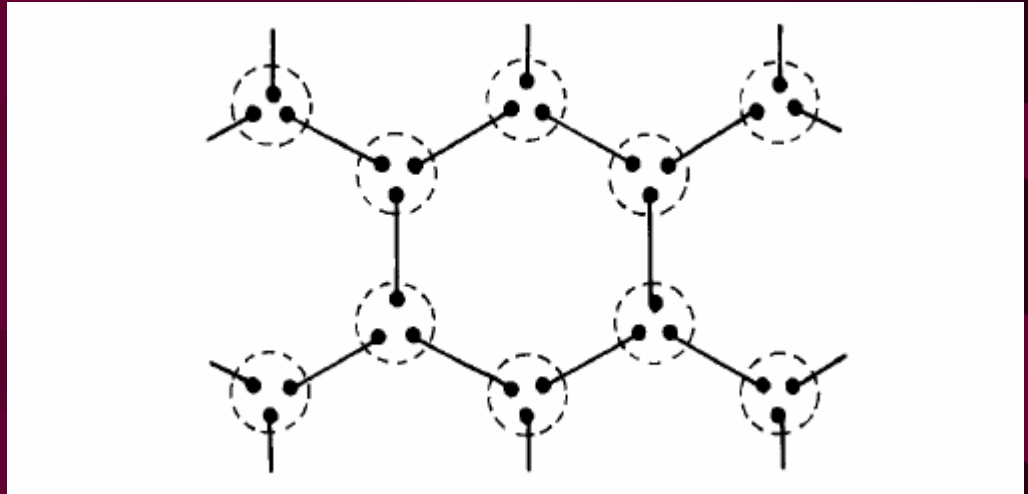
Also exclude

Valence Bond States that mesh with the lattice

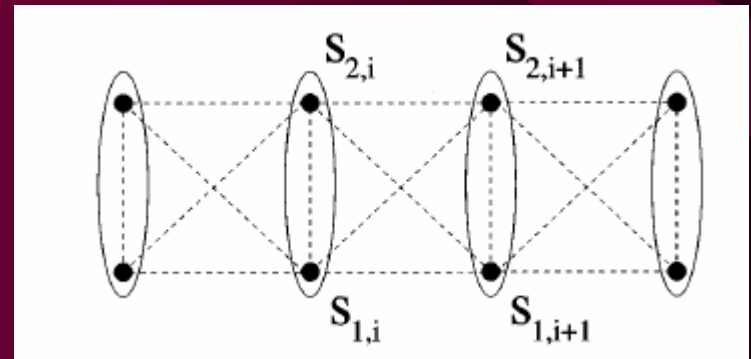
AKLT Models

Spin-3/2 on

Honeycomb



Haldane Chains,
Spin-Ladders



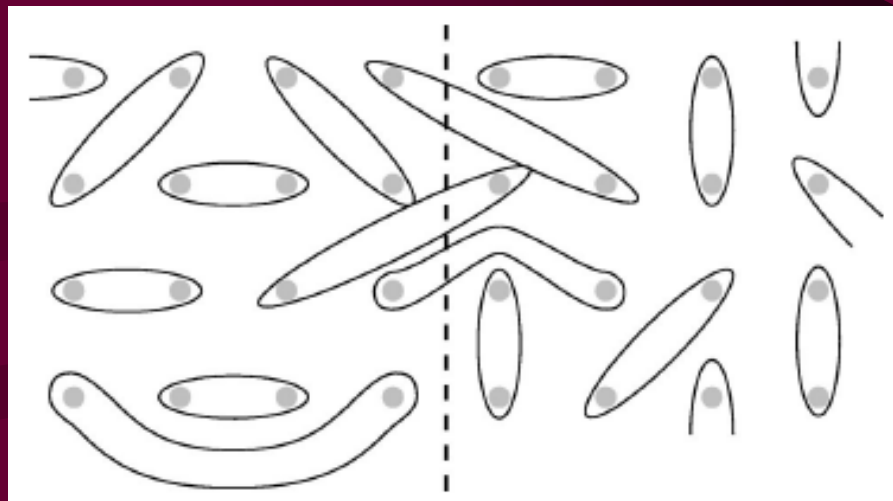
Quantum Spin Liquids

What are theorists looking for?

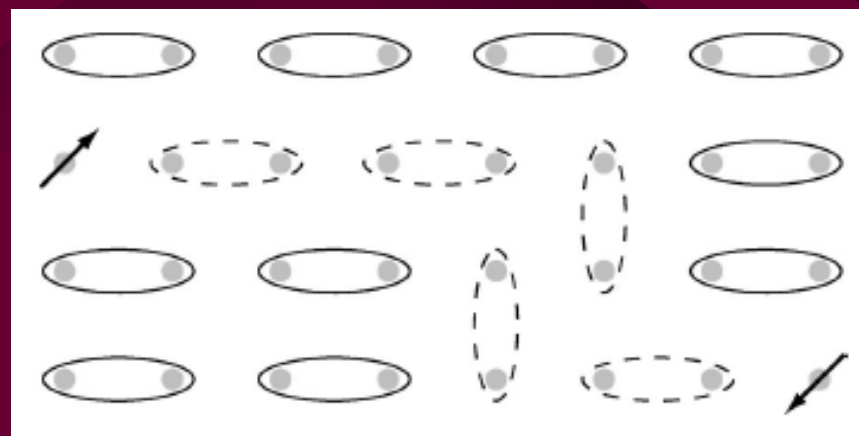
- Resonating Valence Bonds (Anderson, Fazekas and Anderson 1975)
- Allows delocalization of fractional excitations – spinons (Fermions/Bosons) (Sachdev,
- Quantum Dimer Models –Reduced Hilbert Space (Kivelson-Rokhsar-Sethna, Moessner-Sondhi, Misguich et al,)– Short-range→Gapped Topological Spin Liquids
- 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

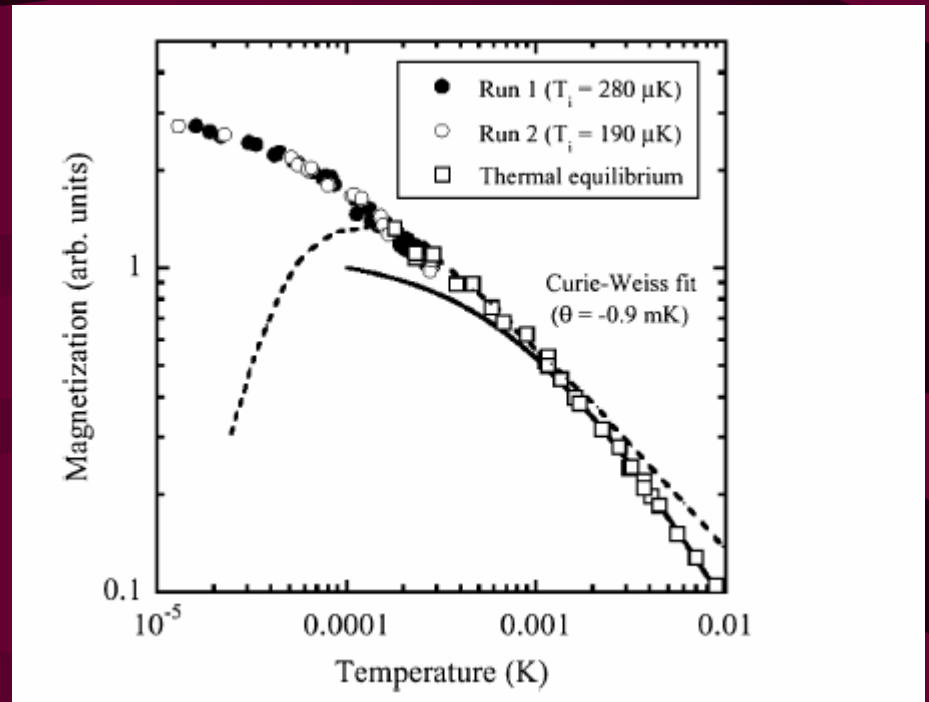
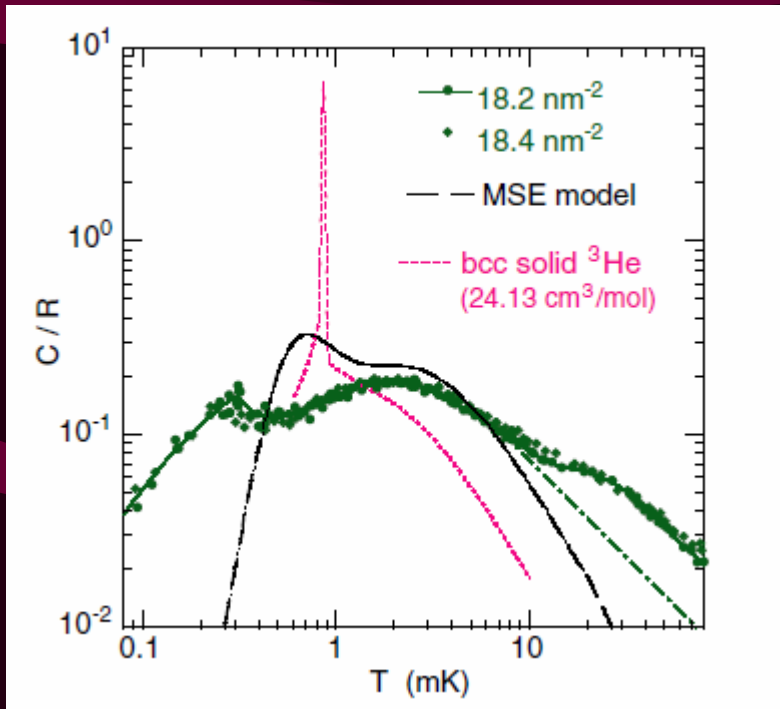
a. Frustration parameter b. Gap/ power-laws in temperature

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?



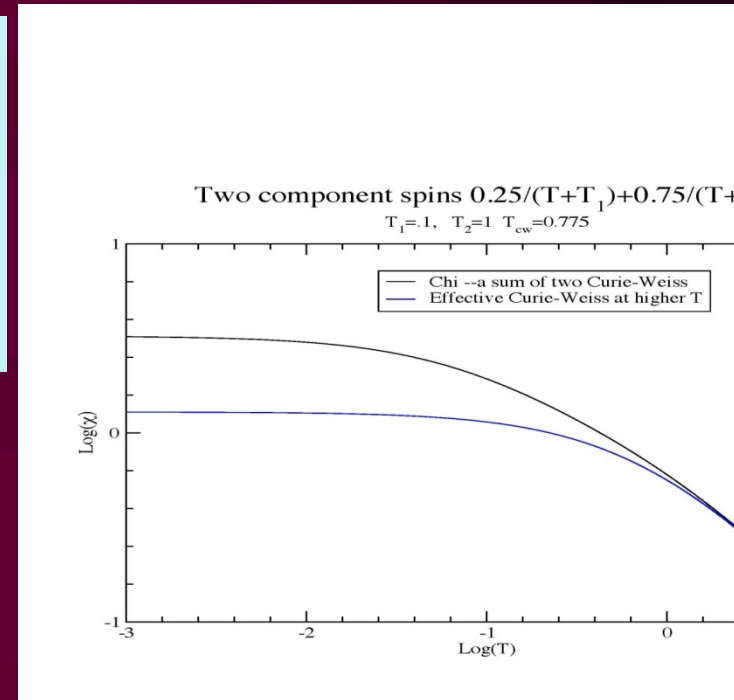
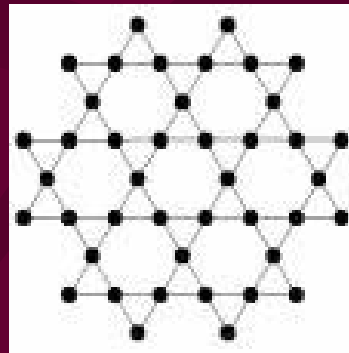
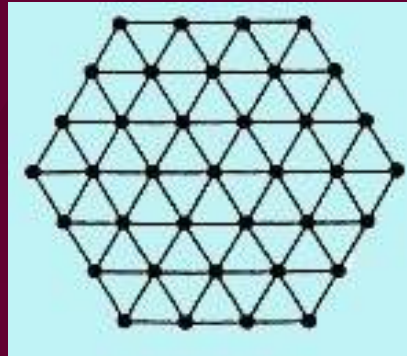
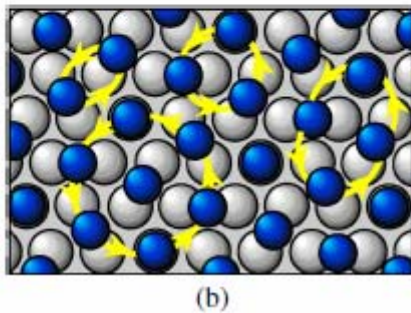
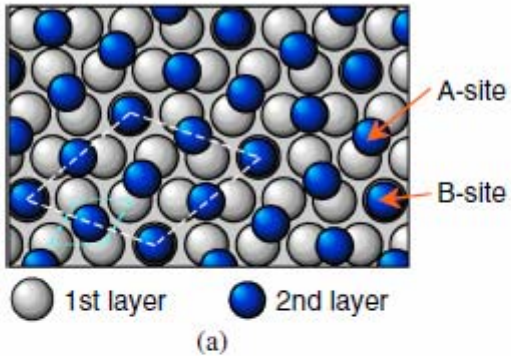
Specific Heat and Susceptibility of
second layer Helium-3 on Graphite

---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

Experimental Realization of a 2D Fractional Quantum Spin Liquid

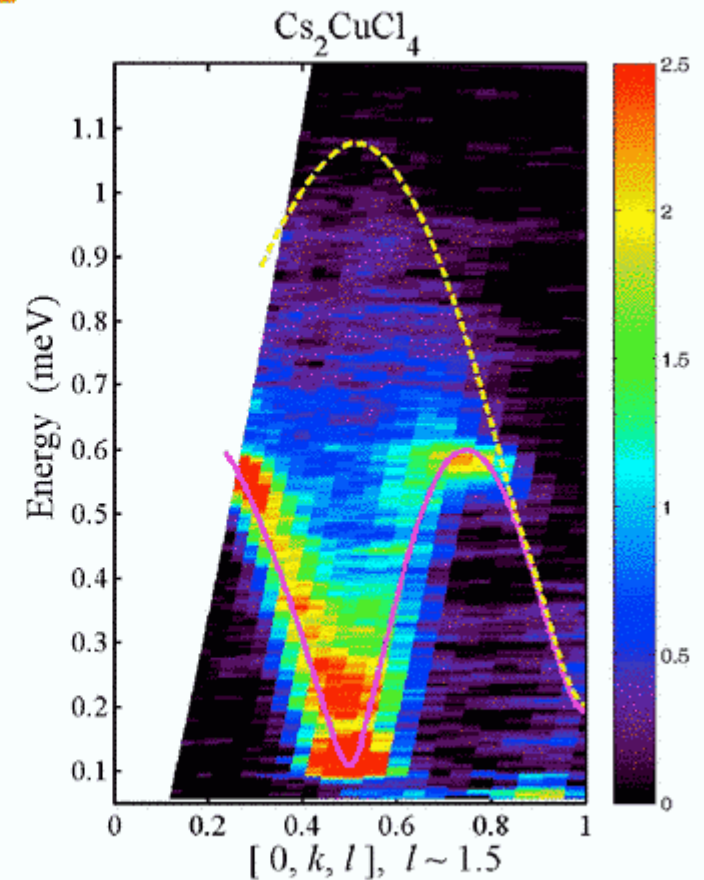
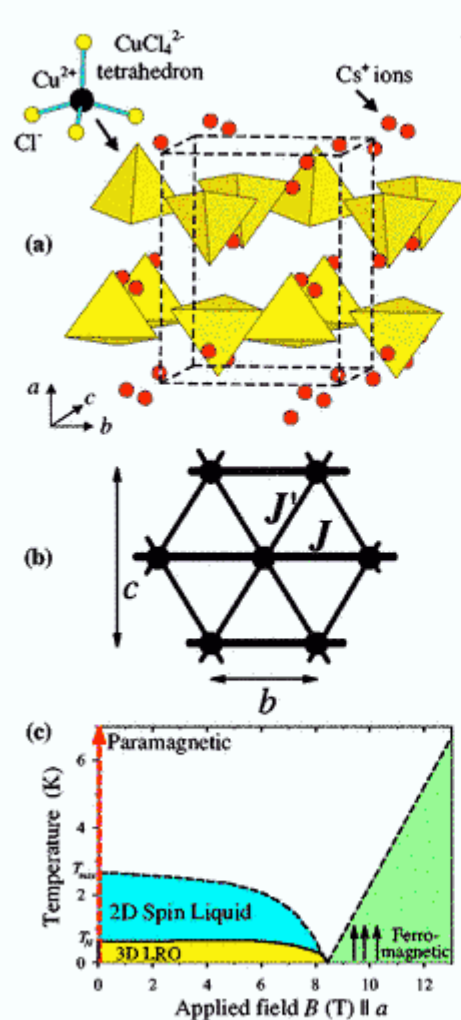
Very Well
Characterized

Layered
Triangular

Know Js from
high-field

Has LRO ---

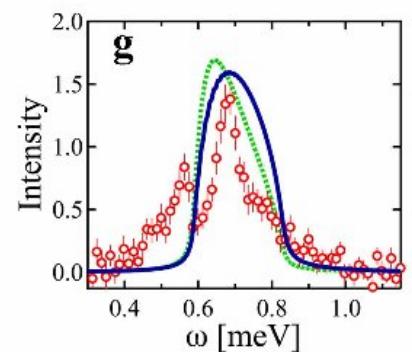
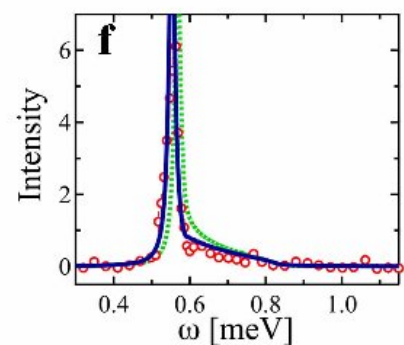
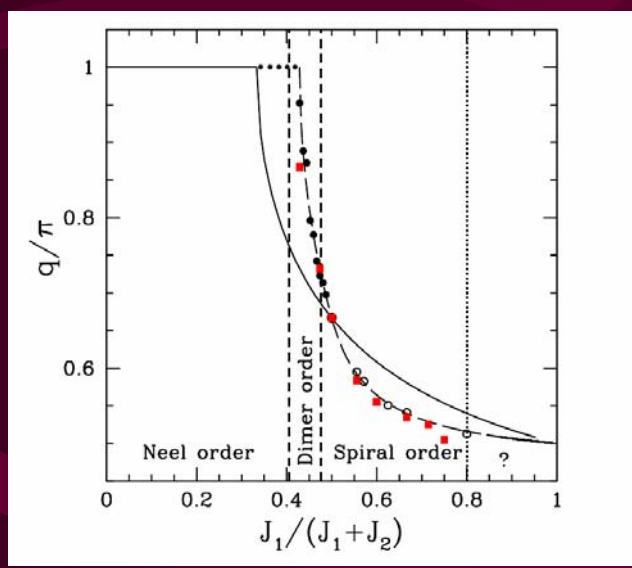
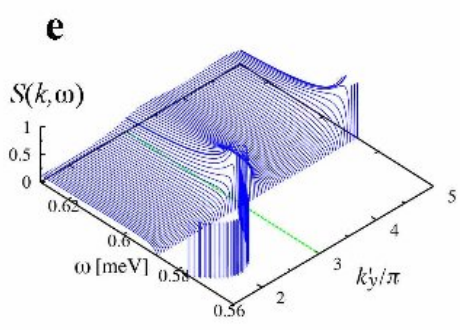
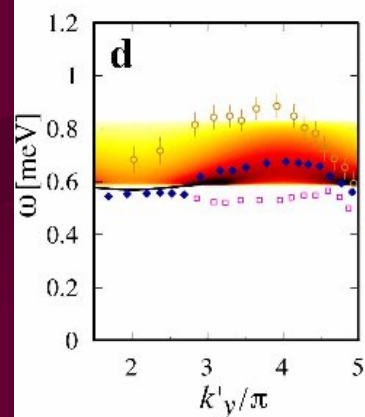
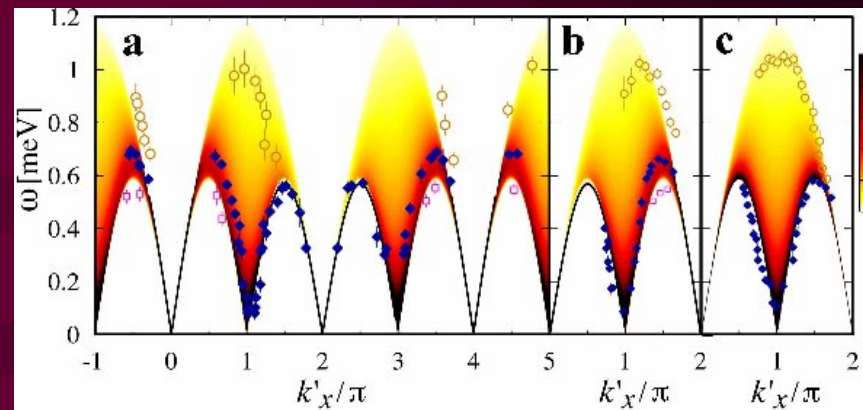
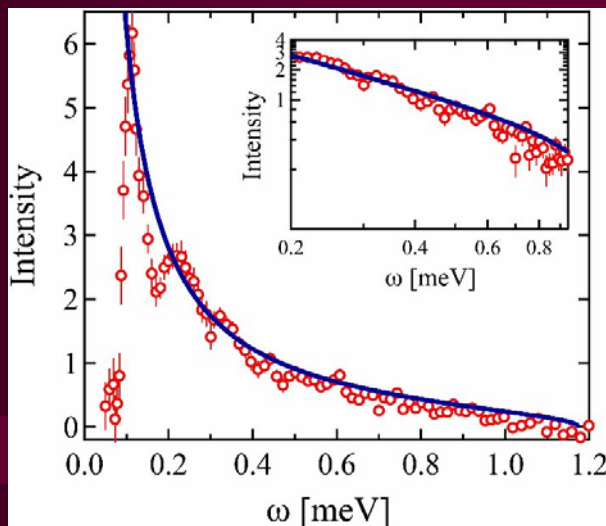
But shows
exotic physics
as well



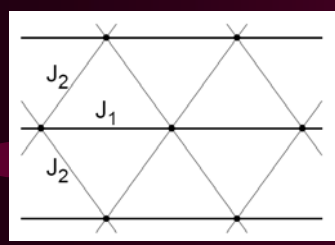
Coldea, Tennant, Tylczynski, PRB68, 134424 (2003)

Balents and Strykh 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

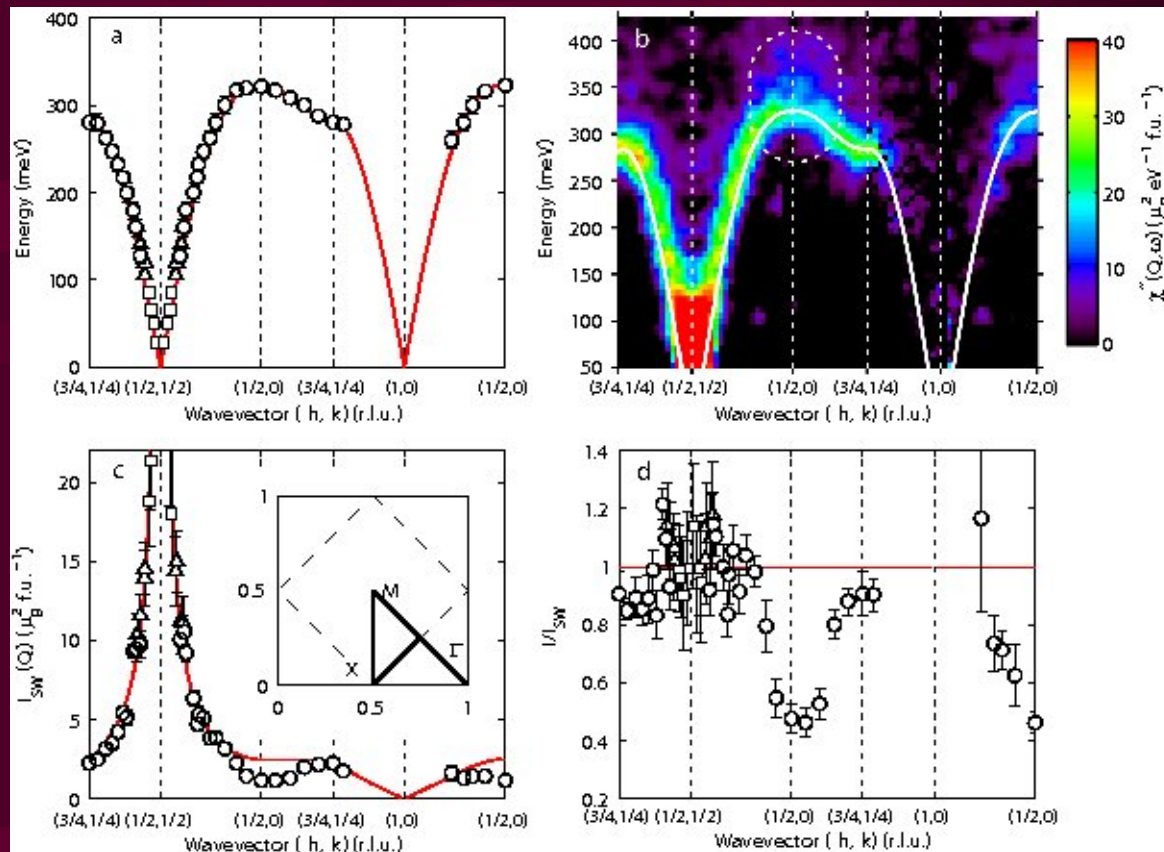


Careful neutron spectroscopy may well reveal a lot more about QSL (even in Cuprates)

Anomalous High-Energy Spin Excitations in La_2CuO_4

N. S. Headings,¹ S. M. Hayden,^{1,*} R. Coldea,^{1,2} and T. G. Perring³

Intermediate
 U/t



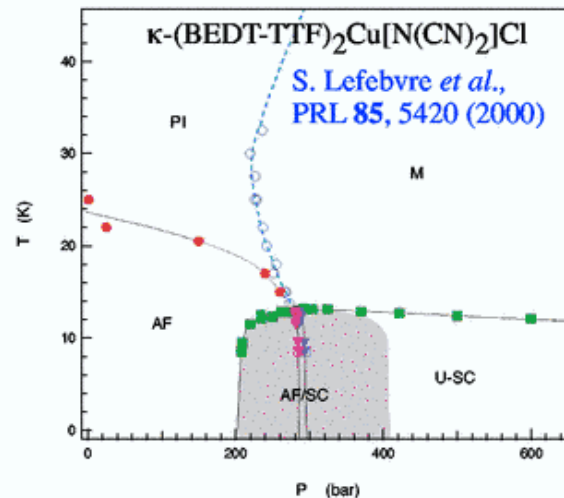
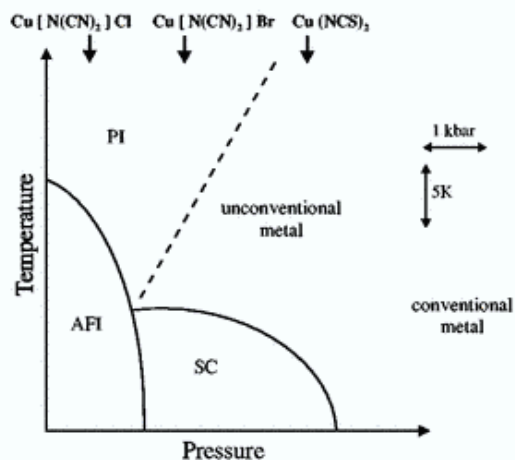
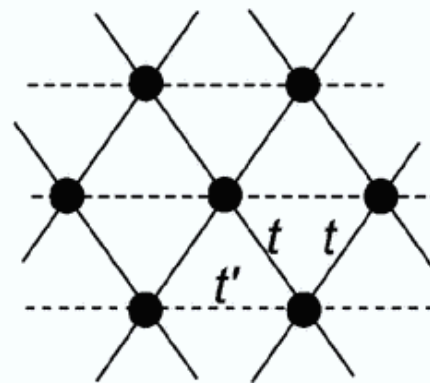
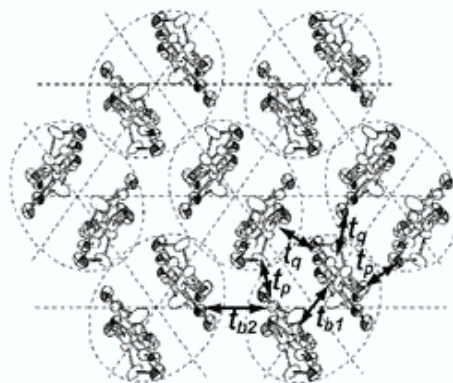
Non quasi-particle dynamics is a hallmark of QSLs

Variety of organic molecular solids

Molecular Dimers in Triangular Geometry

Held together by various ligands

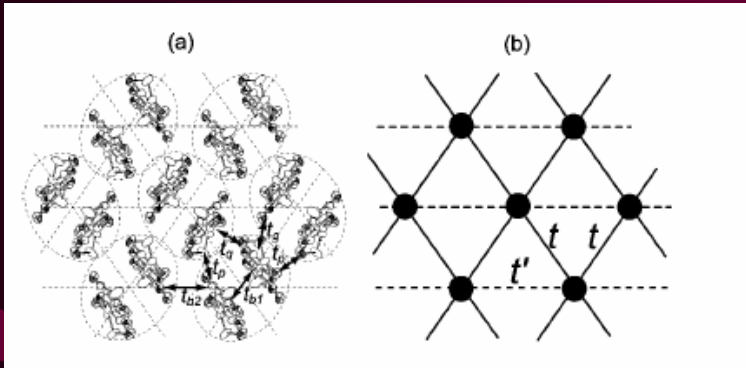
κ -(BEDT-TTF)₂X



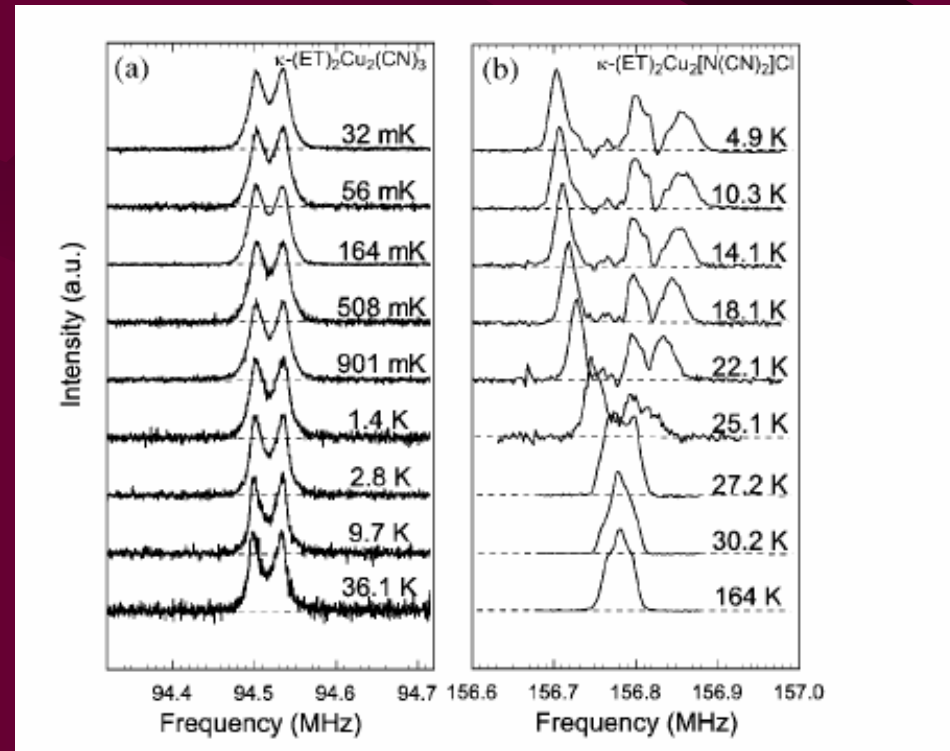
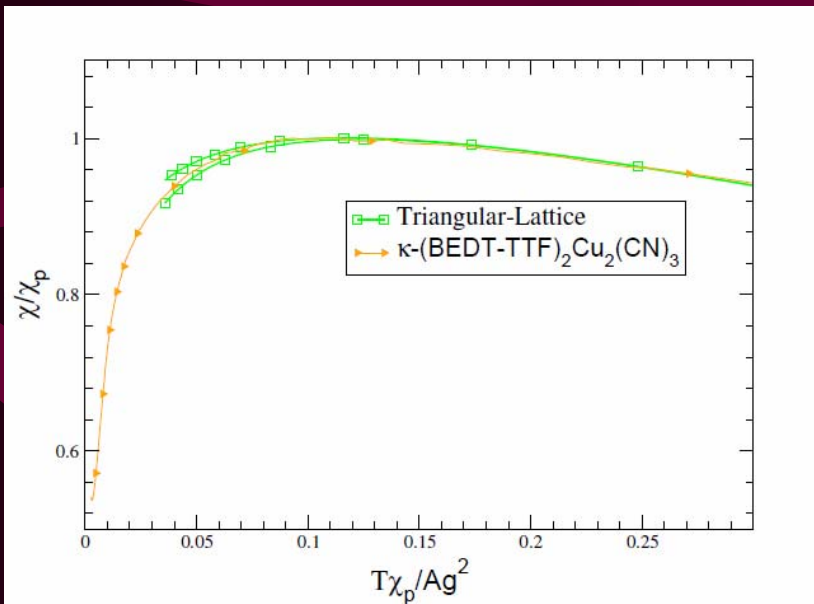
Ross McKenzie, *Science* 278, 820 (1997)

Spin Liquid State in an Organic Mott Insulator with a Triangular Lattice

Y. Shimizu,^{1,2} K. Miyagawa,² K. Kanoda,^{2,3} M. Maesato,¹ and G. Saito¹



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



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?

Lattice Effects and Entropy Release at the Low-Temperature Phase Transition
in the Spin-Liquid Candidate κ -(BEDT-TTF)₂Cu₂(CN)₃

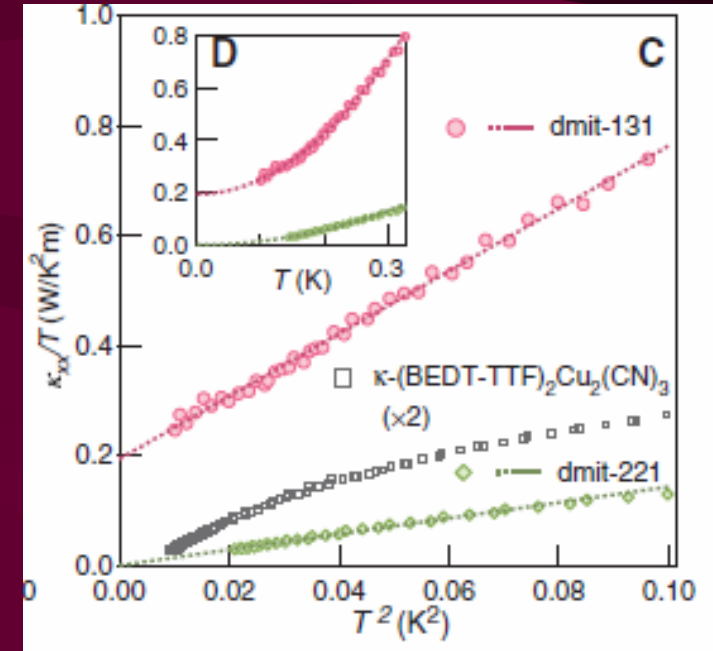
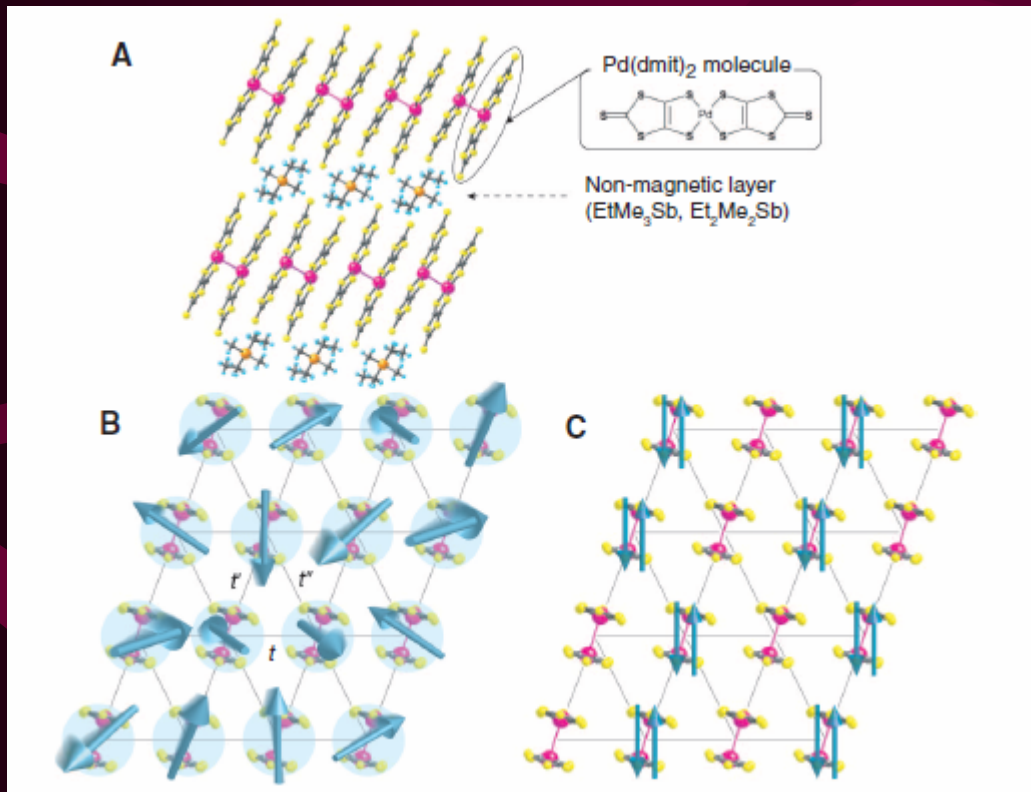
R. S. Manna,¹ M. de Souza,¹ A. Brühl,¹ J. A. Schlueter,² and M. Lang¹

¹Physikalisches Institut, J. W. Goethe-Universität Frankfurt(M), SFB/TR49, D-60438 Frankfurt(M), Germany

²Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

Highly Mobile Gapless Excitations in a Two-Dimensional Candidate Quantum Spin Liquid

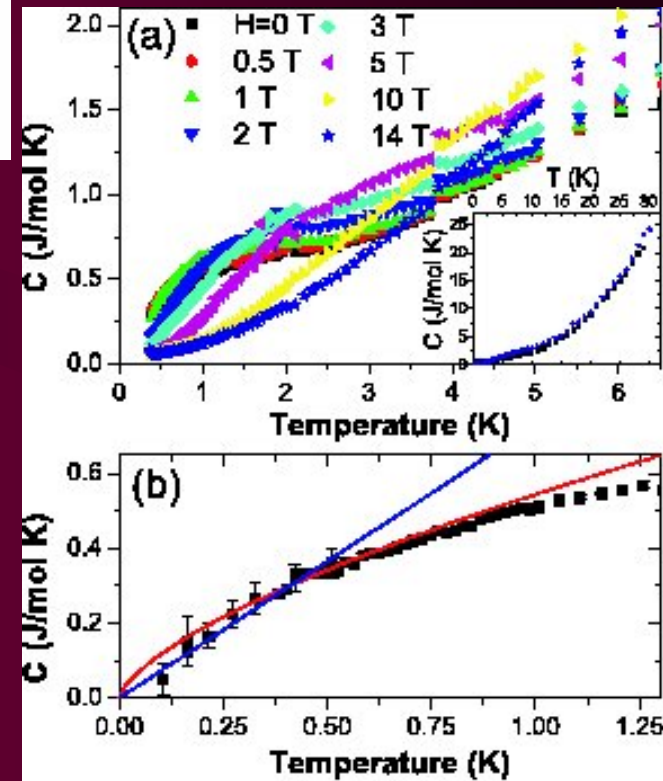
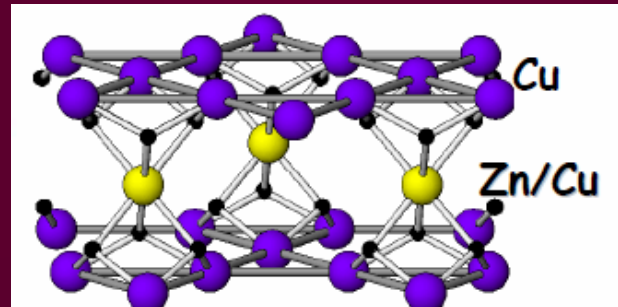
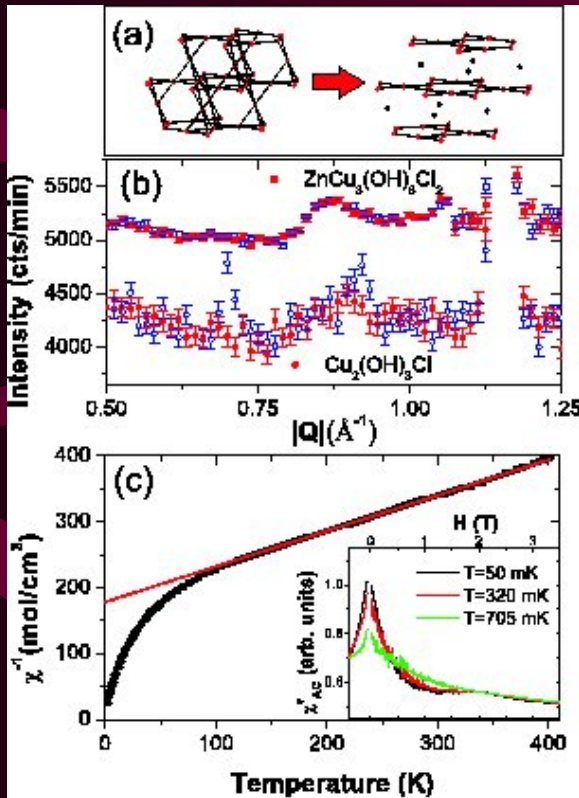
Minoru Yamashita,^{1*} Norihito Nakata,¹ Yoshinori Senshu,¹ Masaki Nagata,¹
Hiroshi M. Yamamoto,^{2,3} Reizo Kato,² Takasada Shibauchi,¹ Yuji Matsuda^{1*}



Is this a better candidate for a QSL? More Insulating, Linear kappa

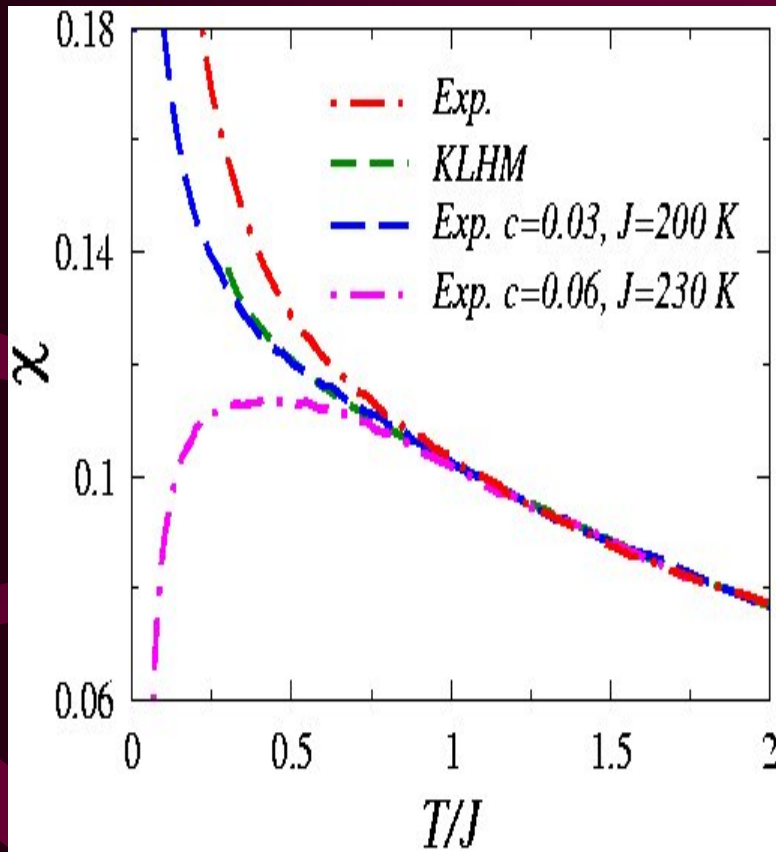
Herbertsmithites (Helton et al)

structurally perfect kagome planes



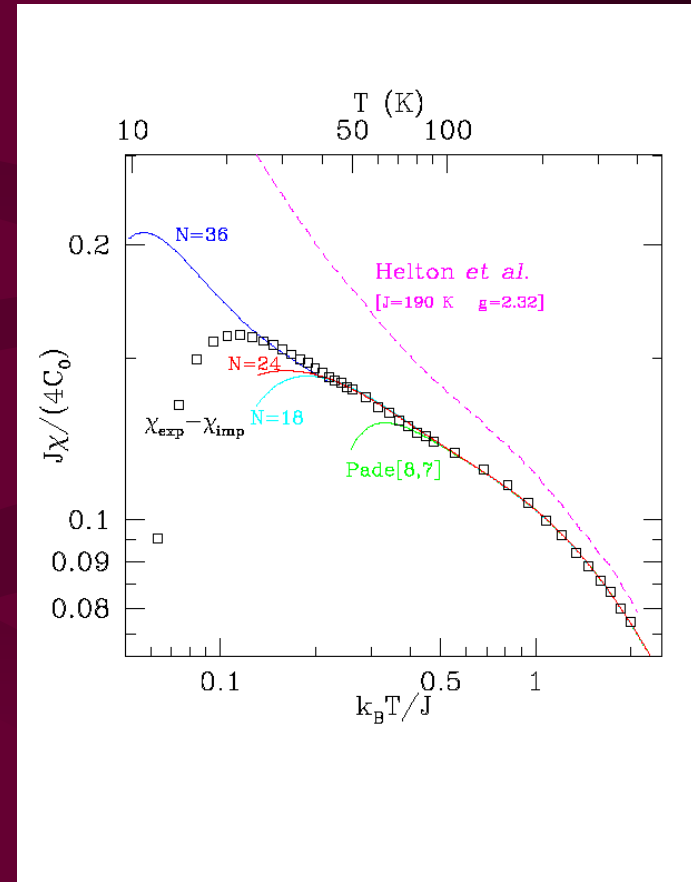
$J=200\text{K}$, no LRO 100mK, Large χ , field-dependent specific heat

Is Curie impurity causing upturn?



Rigol+RRPS

$c=0.04$ fits to 0.3 J

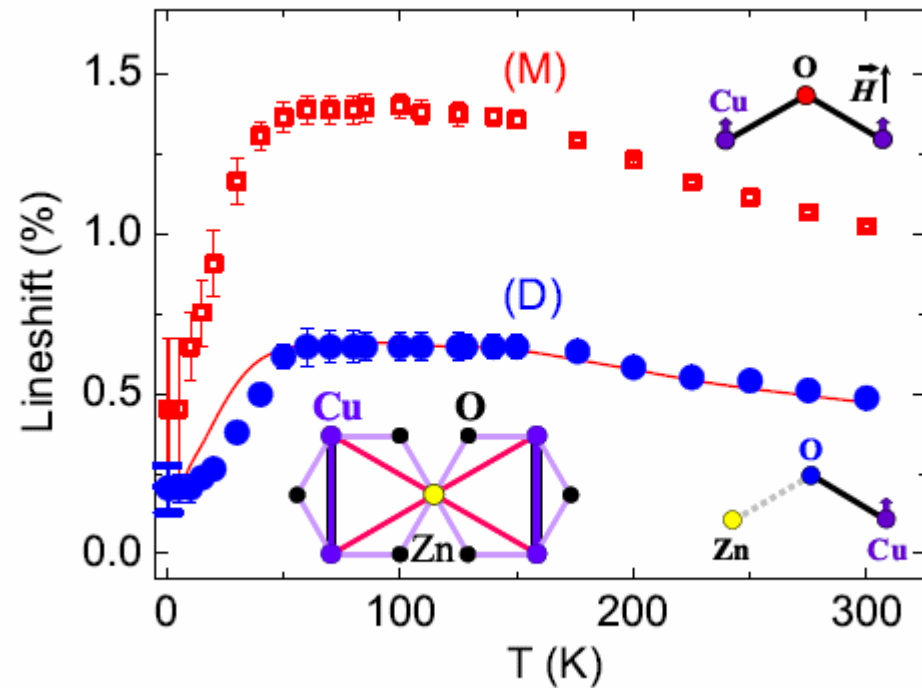
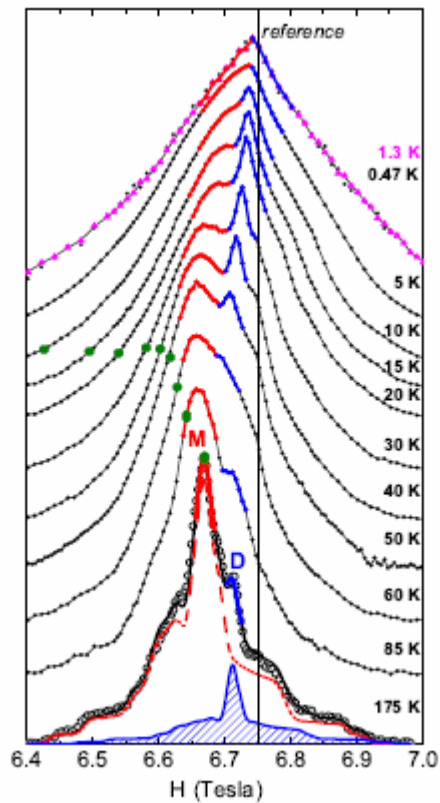


Misguich+sindzingre

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

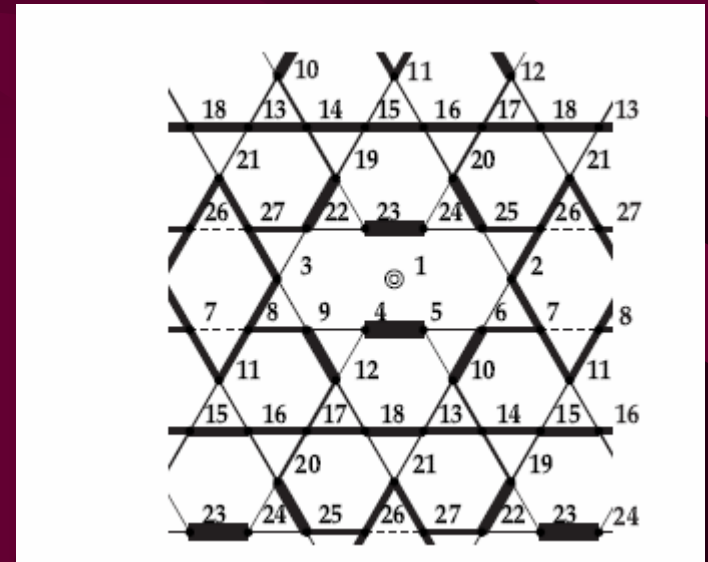
What is the ground state of KLHM?

Or a Valence Bond Glass?

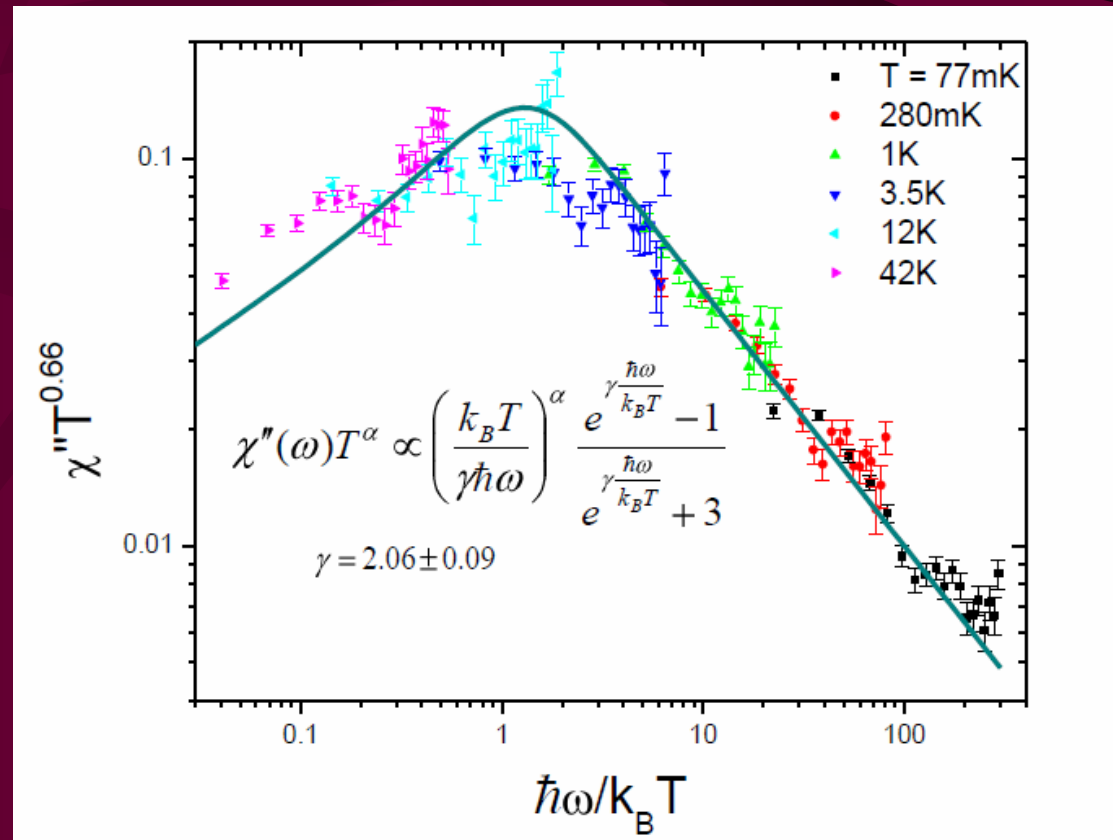
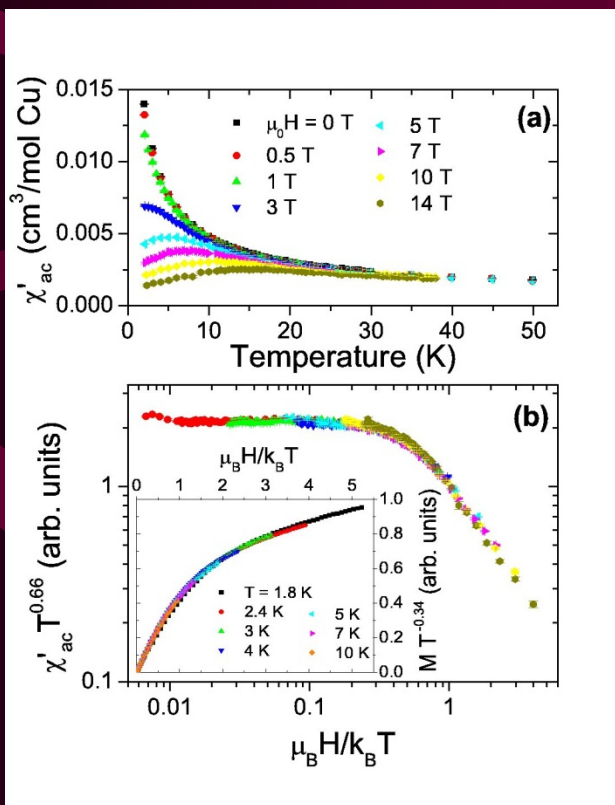
RRPS (PRL)

Dimers freeze around impurities

Dommange (Mila) et al

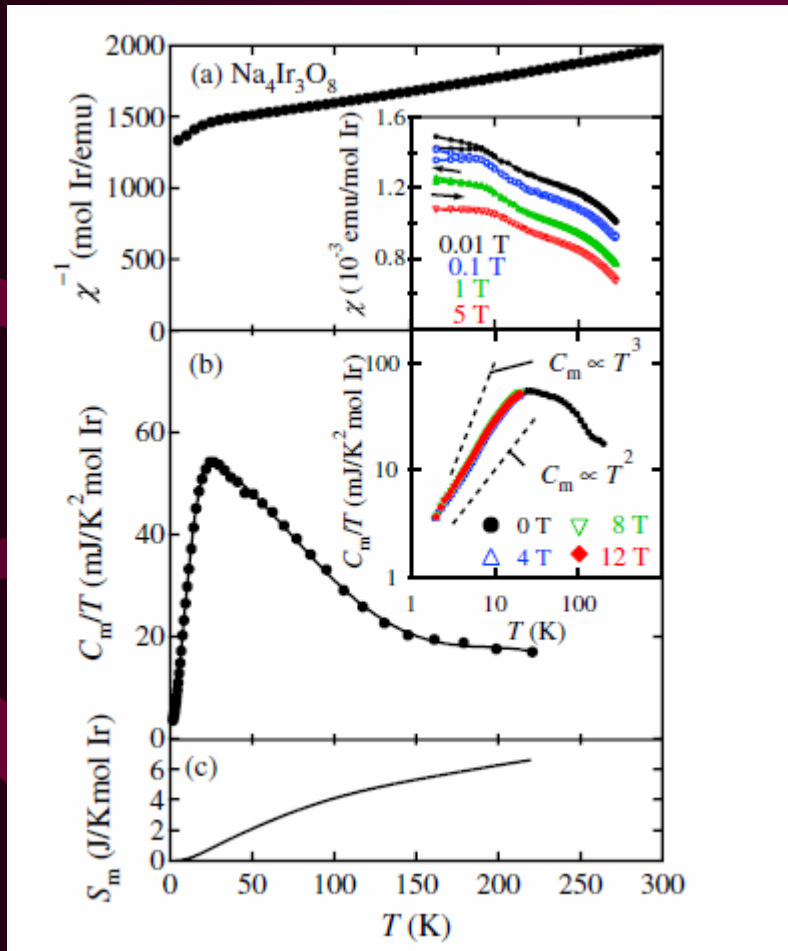


VB Glass can explain Dimer-like form factors (deVries et al) and scaling of susceptibility (Helton et al) Assuming power-law distribution of Js

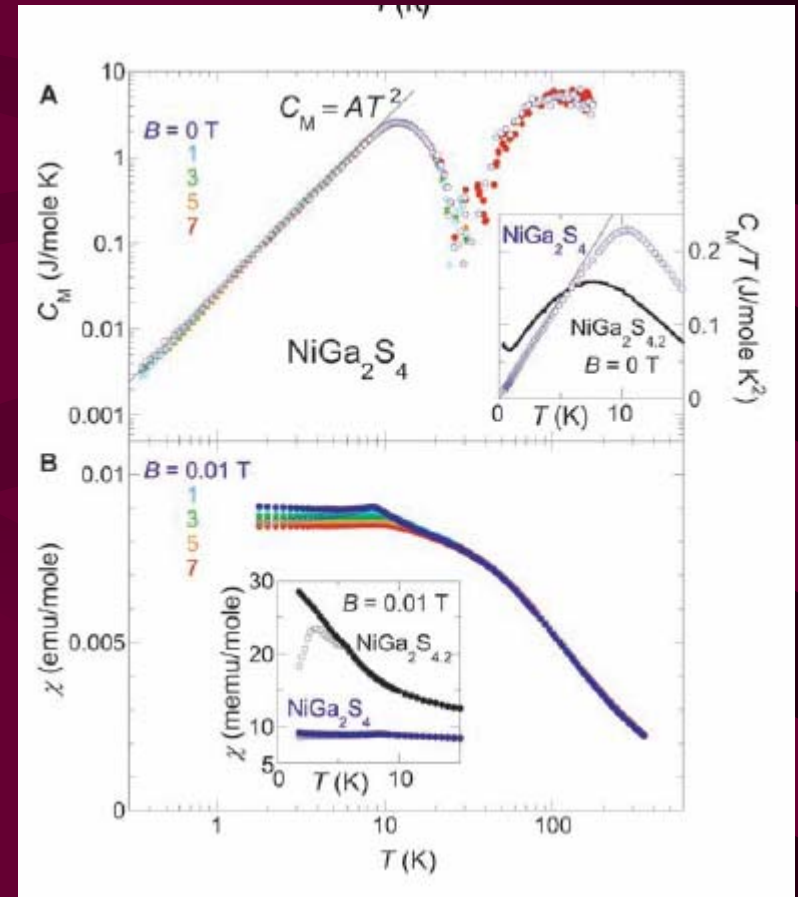


Other systems

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