Title: Lightning Review on Quantum Spin Liquid

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Abstract: We provide a brief introduction to quantum spin liquid and review current status of theoretical and experimental progresses on this subject. Spin liquid phases that arise in different situations are examined in the light of both theoretical models and experimental systems.

Lightning Review on Quantum Spin Liquid

Yong Baek Kim

University of Toronto,
Center for Quantum Materials

HFM "Background Methods",
Perimeter Institute, June 3, 2012

Outline

What is "Quantum" Spin Liquid ?

Overview of Experiments

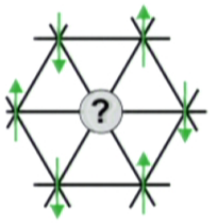
Overview of Theories

Interpretation of Experiments

What is Quantum Spin Liquid ?

1) Different from Cooperative Paramagnet

Large number of equally unhappy (classical) ground states ~ $e^{\alpha N}$



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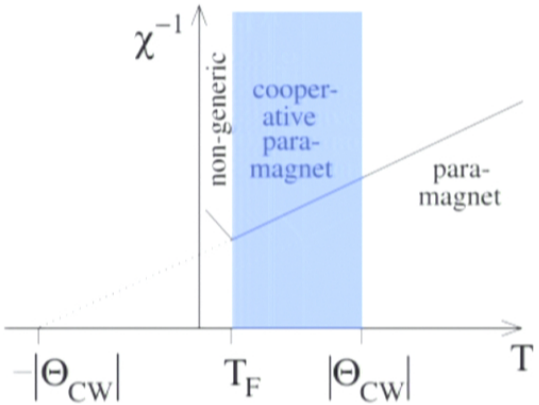

Θ_{CW} : Curie-Weiss temperature

$$\chi \sim \frac{1}{T - \Theta_{CW}} \quad T \gg |\Theta_{CW}|$$

$$f = \frac{|\Theta_{CW}|}{T_F}$$ useful diagnostic of frustration

$f \gg 1$ strong frustration

highly degenerate classical ground state manifold

"classical spin liquid"

What is Quantum Spin Liquid ?

2) Different from boring incoherent (high temp) Paramagnet

Spins fluctuate independently; don't talk to each other

No coherent excitations

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No coherent excitations

3) Spin Liquid in 1D

Spin-1/2 Heisenberg Model in 1D: Spin Liquid Ground State

Quantum paramagnet, No translational symmetry breaking

Coherent Excitations; S=1/2 spinons

What is Quantum Spin Liquid ?

Neutron Scattering -- Spin-1 excitations

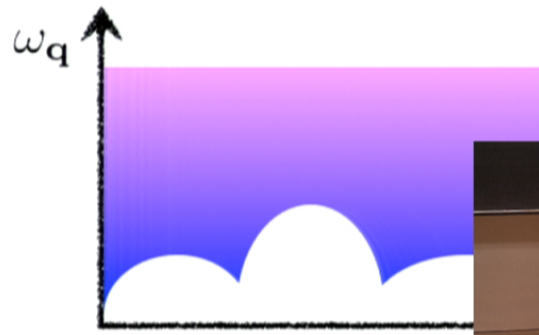
Spinon-Antispinon pair excitations

Well-defined dispersion -->

Threshold energy for pair excitations

$$\omega_{\mathbf{q}} \sim \min\left[\varepsilon_{\frac{\mathbf{q}}{2}+\frac{\mathbf{p}}{2}} + \varepsilon_{\frac{\mathbf{q}}{2}-\frac{\mathbf{p}}{2}}\right]$$

for all possible $\mathbf{p}$



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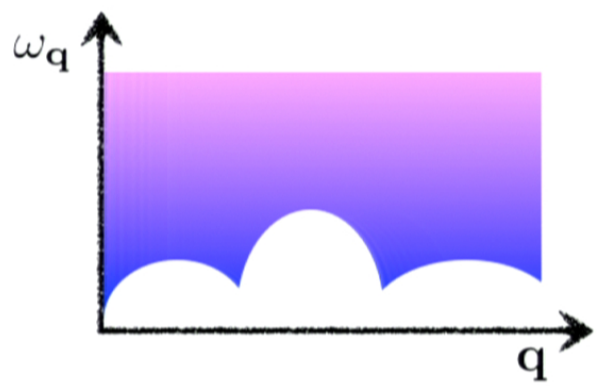

Higher dimensions ?

Quantum Paramagnet $\langle \mathbf{S} \rangle = 0$

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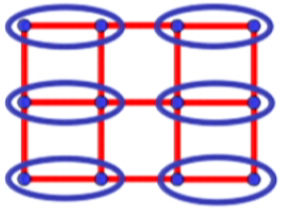
i) Translational symmetry breaking; Valence Bond Solid (VBS)



Valence Bond

$$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$$

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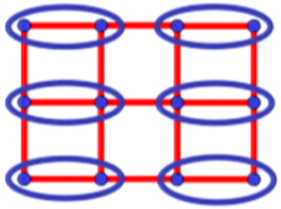

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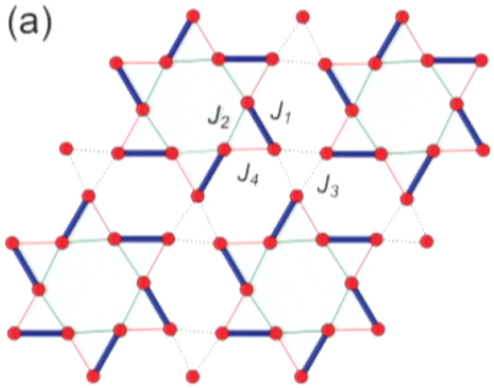

$Rb_2Cu_3SnF_{12}$

Tanaka; Thursday
HFM conference

Higher dimensions ? Quantum Paramagnet $\langle \mathbf{S} \rangle = 0$

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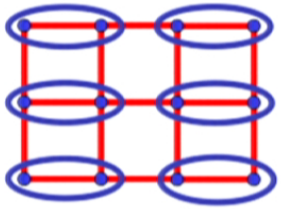

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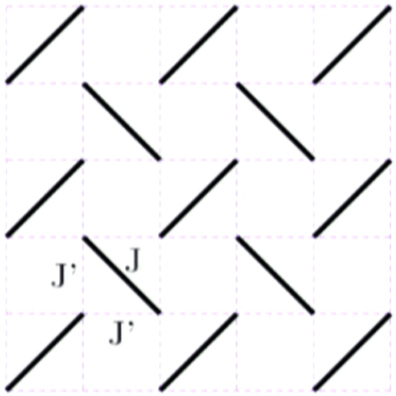


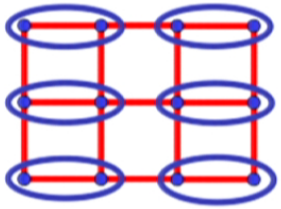
Shastry-Sutherland Lattice

$(\mathrm{CuCl})\mathrm{LaNb_2O_7}$ Kageyama; Wednesday

$\mathrm{SrCu_2(BO_3)_2}$ Takigawa; Wednesday

Higher dimensions ? Quantum Paramagnet $\langle \mathbf{S} \rangle = 0$

i) Translational symmetry breaking; Valence Bond Solid (VBS)



Valence Bond

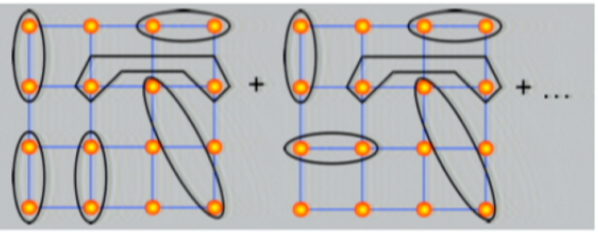
$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$

ii) No broken translational symmetry in Mott insulator

Spin Liquid: chargeless spin-1/2 excitations with/without spin gap

Resonating Valence Bond state (RVB); Superposition of Valence Bond coverings

$\Psi =$

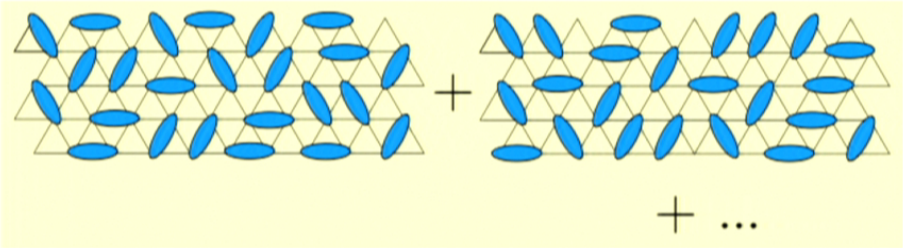


P.W.Anderson

Rokhsar-Kivelson

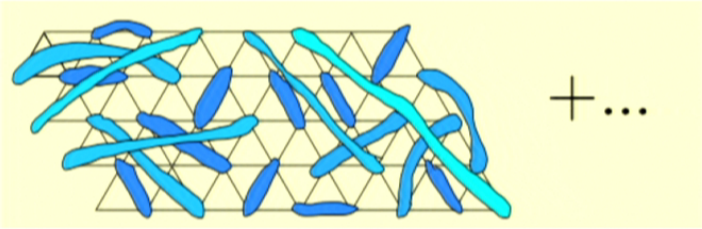
Many Flavors of Spin Liquid States

Short-range RVB



Gapped spinons, Finite spin gap $\langle \mathbf{S}(\mathbf{r}) \cdot \mathbf{S}(\mathbf{0}) \rangle \sim e^{-r/\xi}$

Long-range RVB



Gapless spinons; "Critical" Spin Liquid
Zero spin gap

$$\langle \mathbf{S}(\mathbf{r}) \cdot \mathbf{S}(\mathbf{0}) \rangle \sim \frac{1}{r^{\alpha}}$$

S=1/2 Candidate Materials for Spin Liquid

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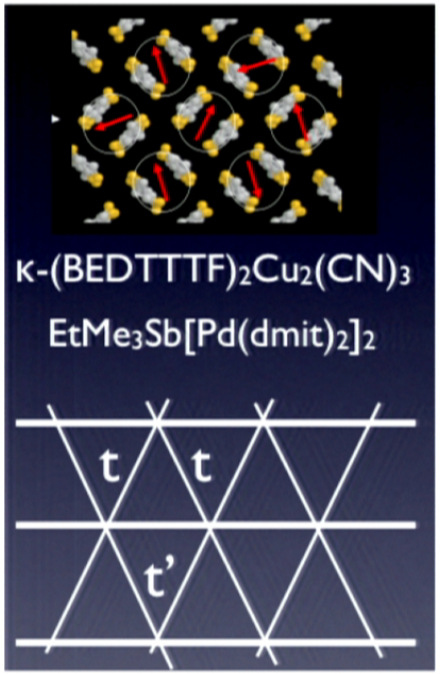
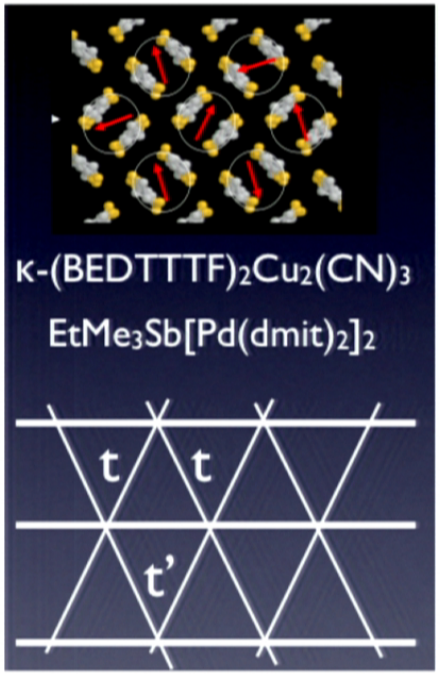


K. Kanoda
R. Kato

S=1/2 Candidate Materials for Spin Liquid

D. G. Nocera, Y. S. Lee



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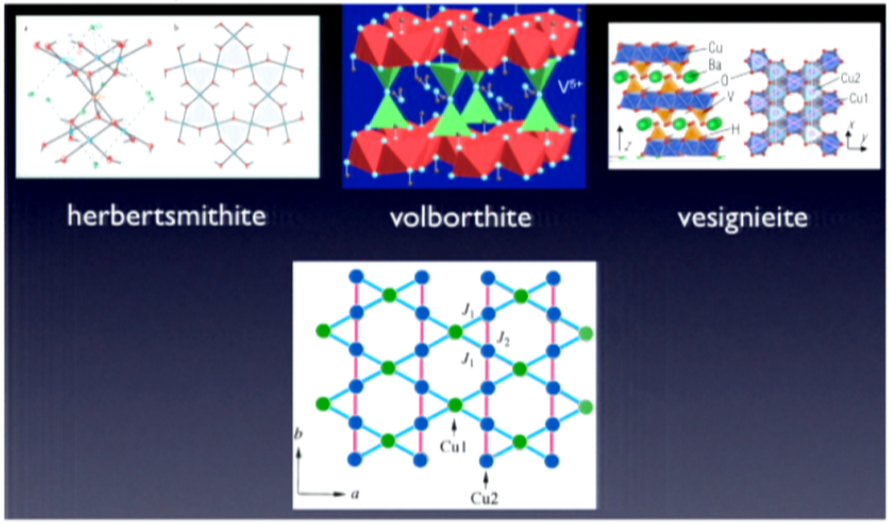

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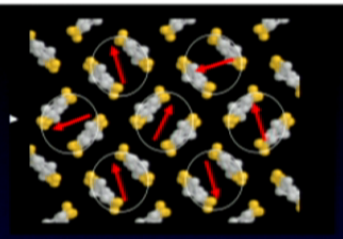
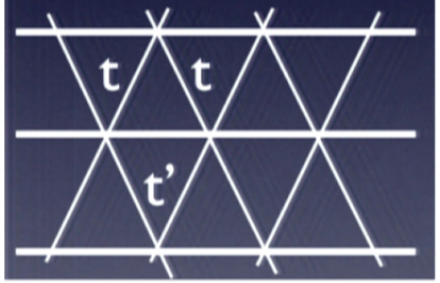


κ-$(BEDTTTF)_2Cu_2(CN)_3$

$EtMe_3Sb[Pd(dmit)_2]_2$



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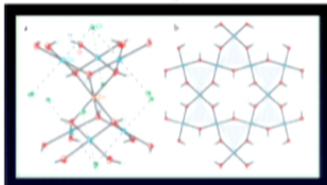
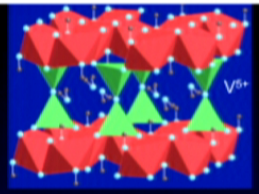
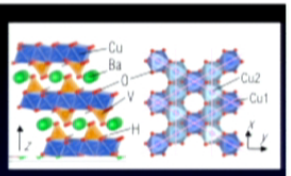


herbertsmithite



volborthite



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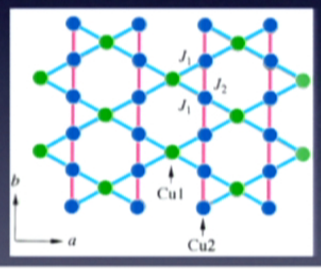

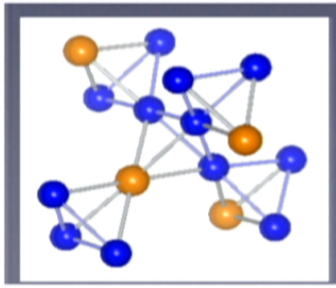

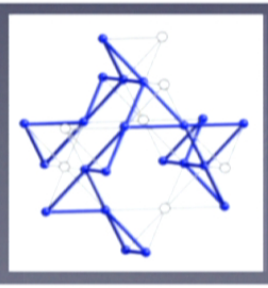

$Na_4Ir_3O_8$

H. Takagi

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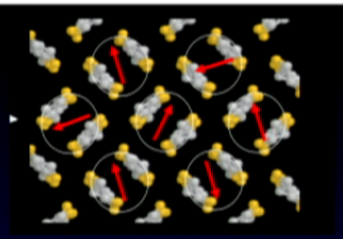

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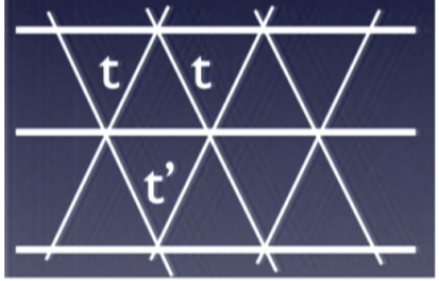

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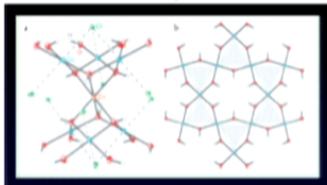
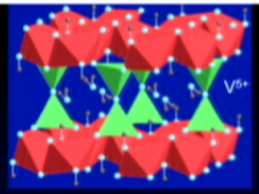


herbertsmithite



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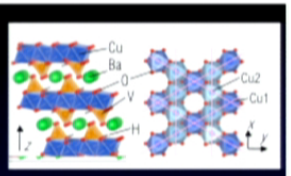
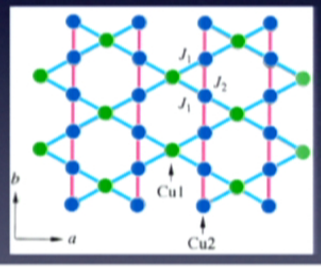
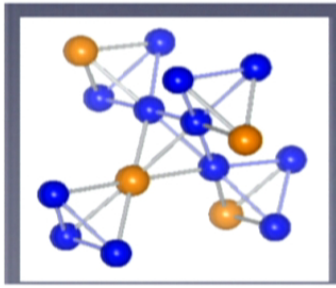


vesignieite

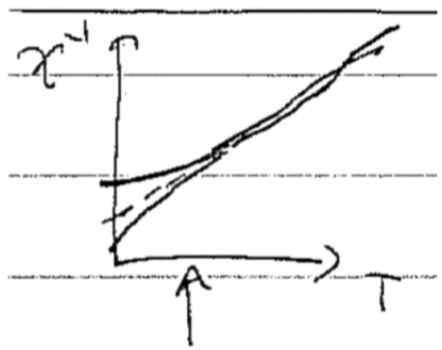


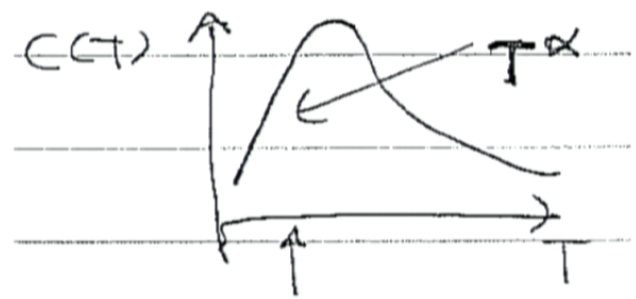


$Na_4Ir_3O_8$

H. Takagi

Typical Behaviors





Charge Insulator

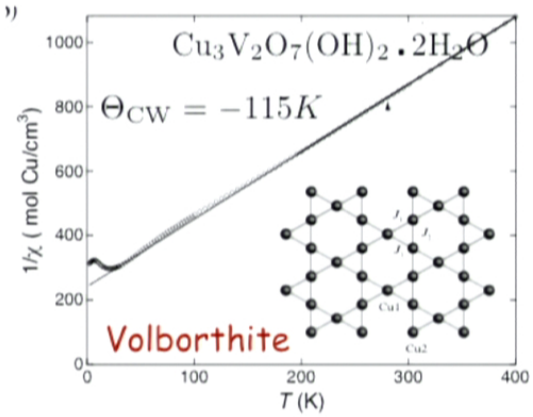
Constant susceptibility at low temperatures

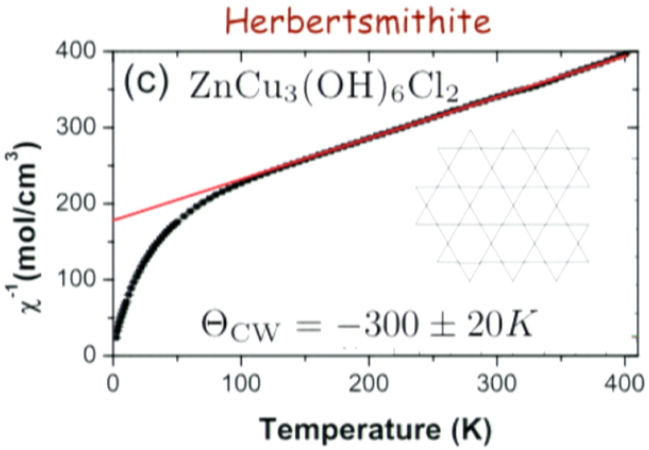
Power-law specific heat

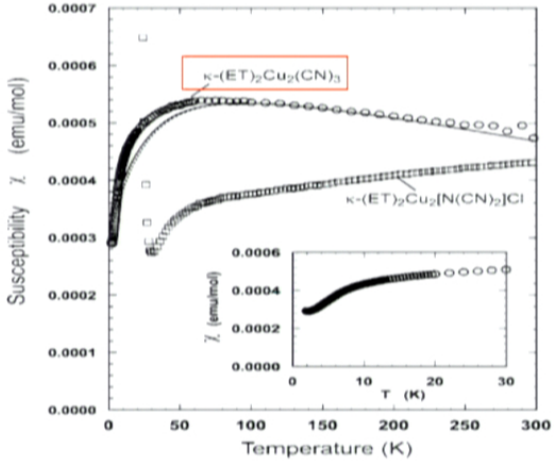
A lot of low energy (charge-neutral) excitations that carry spin quantum number

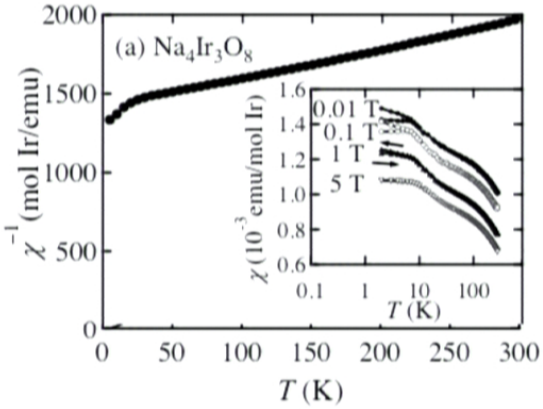
--> Spin Liquid with gapless spinon excitations ?

Spin Susceptibility

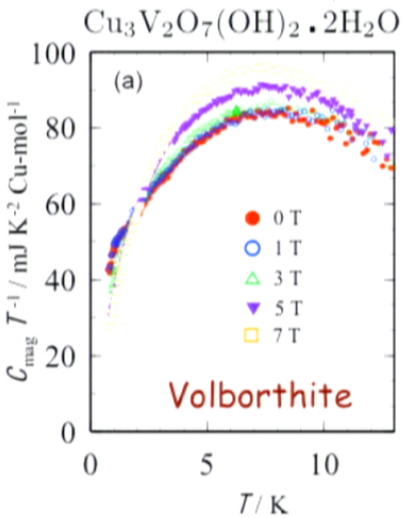


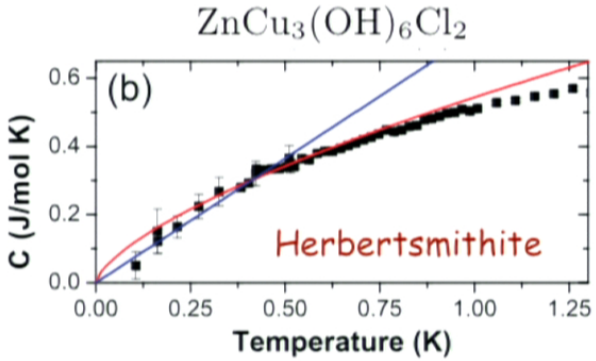


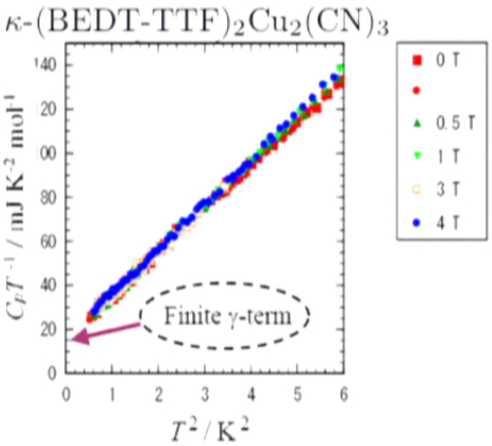


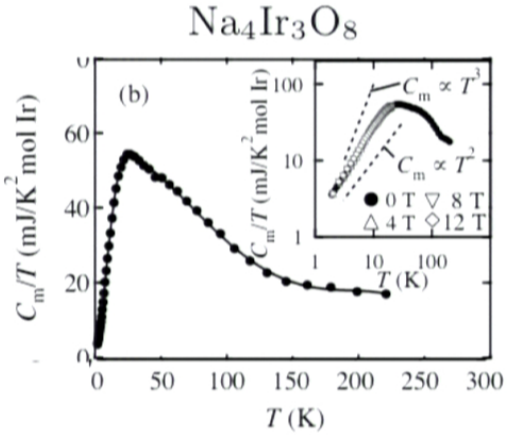


Specific Heat









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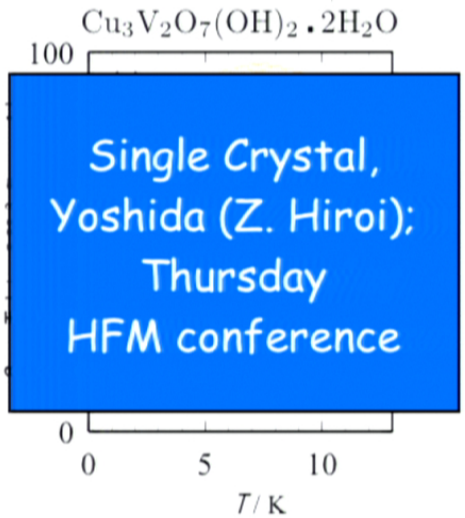
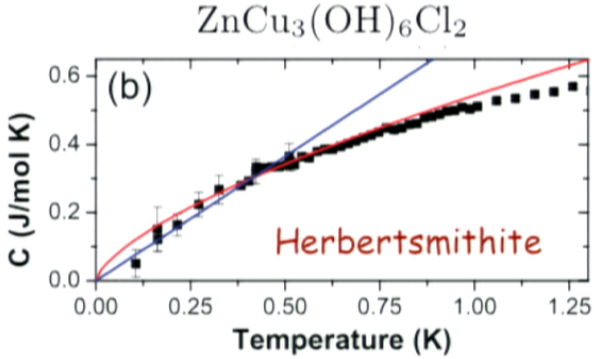
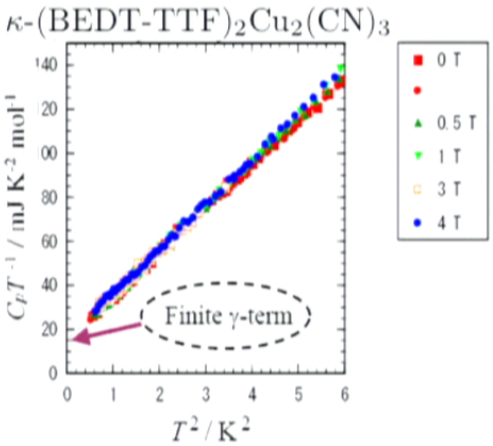
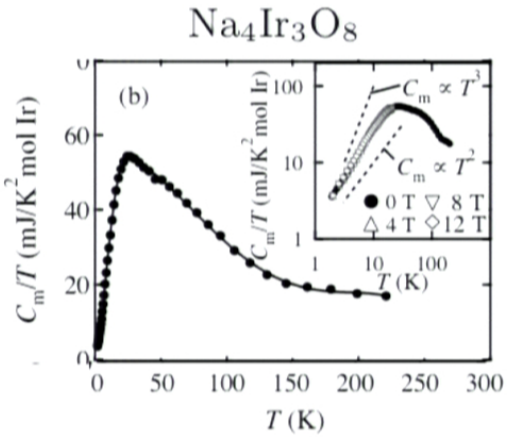


Single Crystal,
Yoshida (Z. Hiroi);
Thursday
HFM conference







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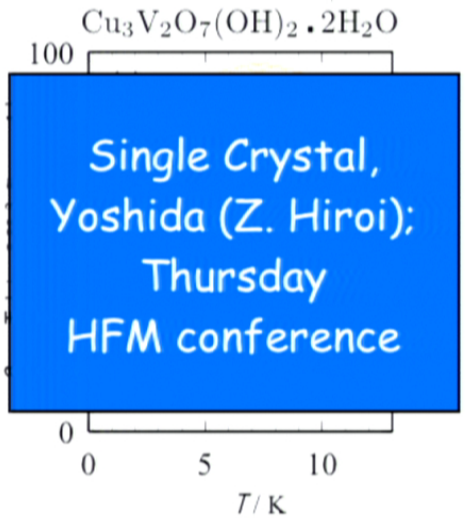

Single Crystal,
Yoshida (Z. Hiroi);
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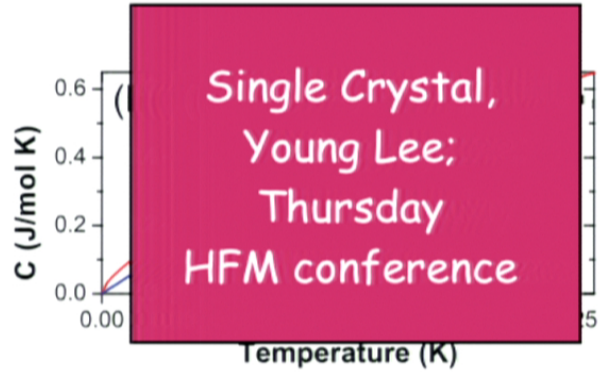
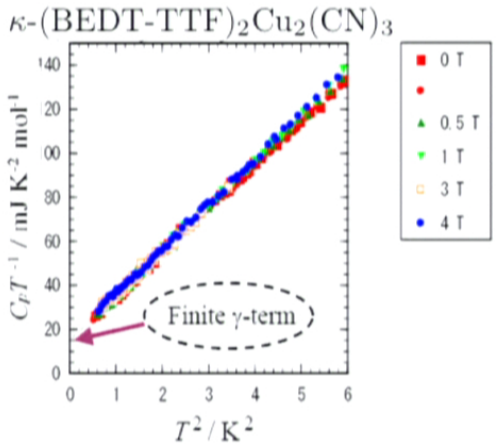
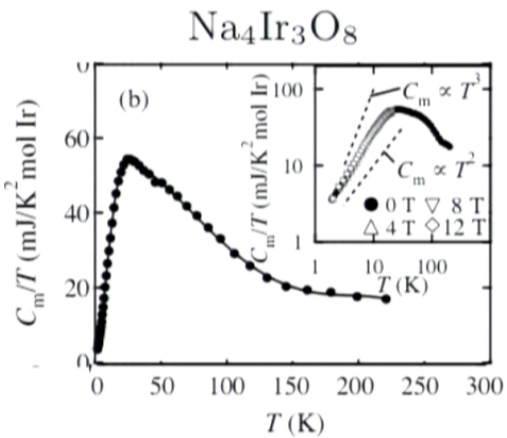


Single Crystal,
Young Lee;
Thursday
HFM conference





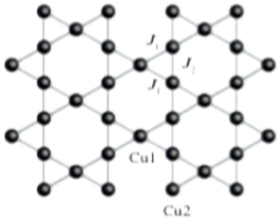
Search for Quantum Spin Liquid ("S=1/2")

Herbertsmithite "Ideal" Kagome lattice

$ZnCu_3(OH)_6Cl_2$ D. G. Nocera, Y. S. Lee

Volborthite Distorted Kagome lattice

$Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ Z. Hiori



κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ K. Kanoda

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Organic Material
Triangular Lattice

Hyperkagome $Na_4Ir_3O_8$

H. Takagi

Search for Quantum Spin Liquid ("S=1/2")

Herbertsmithite "Ideal" Kagome lattice

$ZnCu_3(OH)_6Cl_2$ D. G. Nocera, Y. S. Lee

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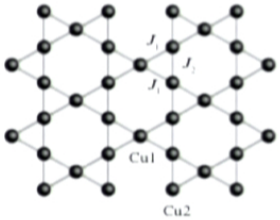

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Weak Mott Insulator ?

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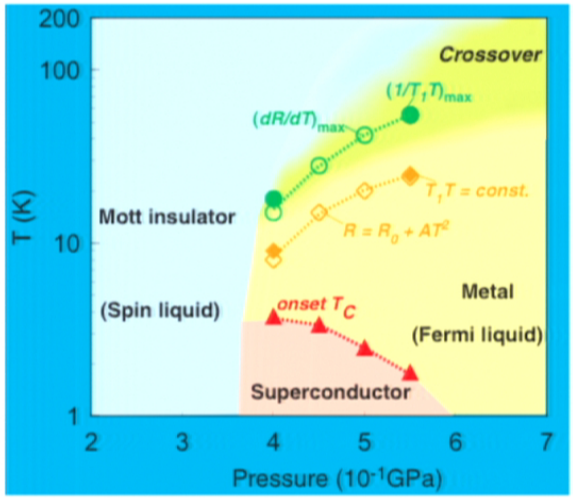

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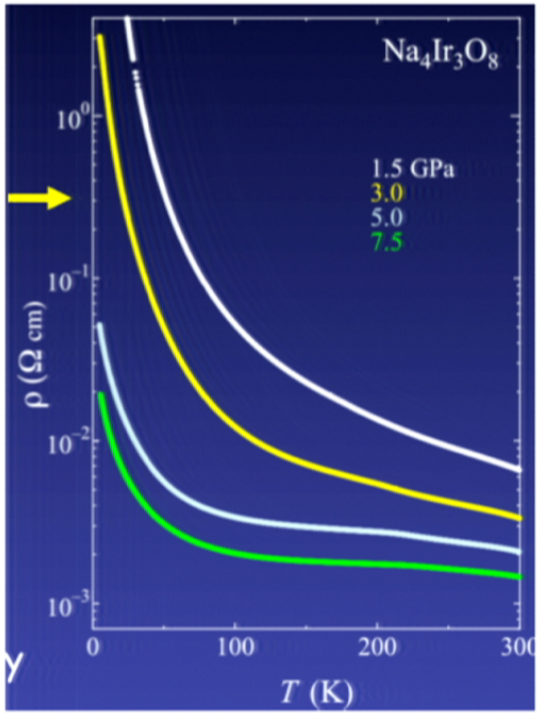

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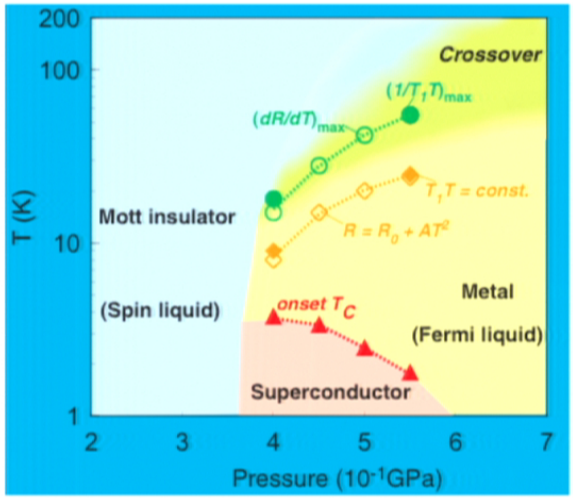

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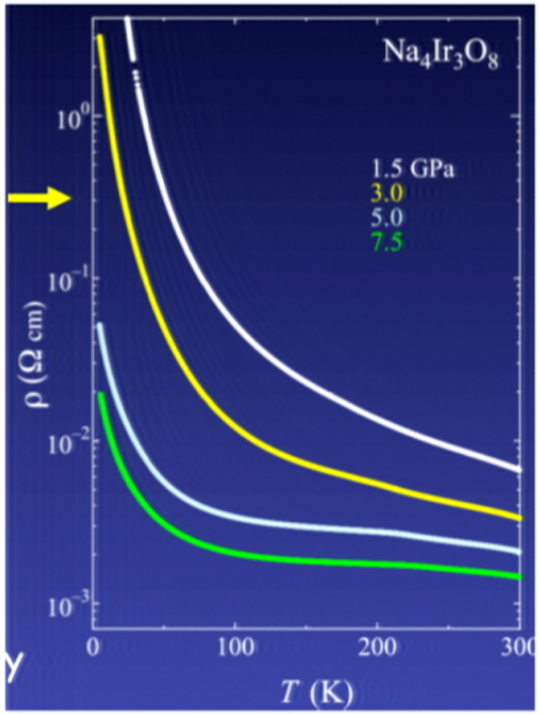
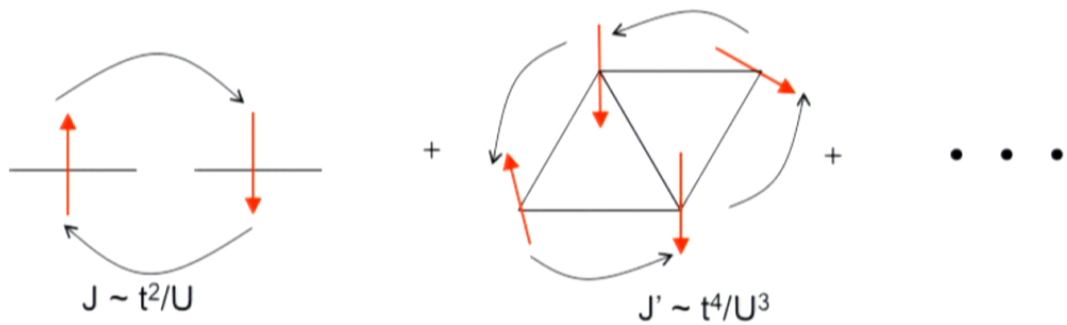


Importance of Charge Fluctuations

$$H \sim H_{\text{heisenberg}} + H_{\text{ring}} + \cdots$$



Charge fluctuations are important near
the Mott transition even in the insulating phase

Imada (2003)

Motrunich (2005)

S. S. Lee and P. A. Lee (2005)

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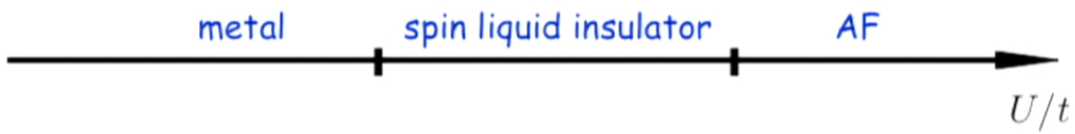
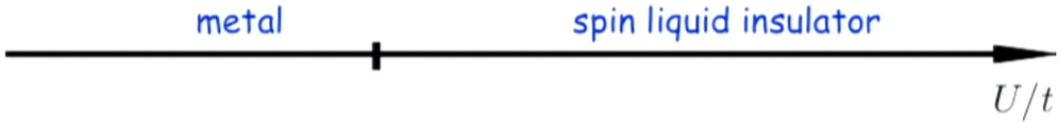
Triangular Lattice; near Mott transition



Hyper-Kagome $\mathrm{Na_4Ir_3O_8}$ H. Takagi

5d transition metal oxides; intermediate coupling



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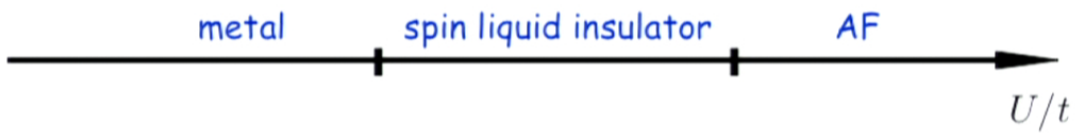

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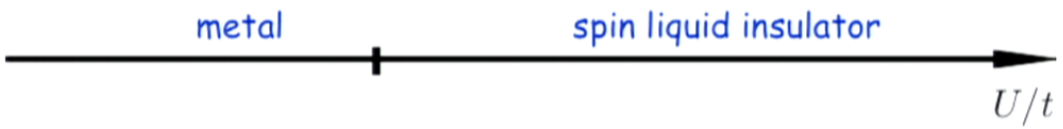
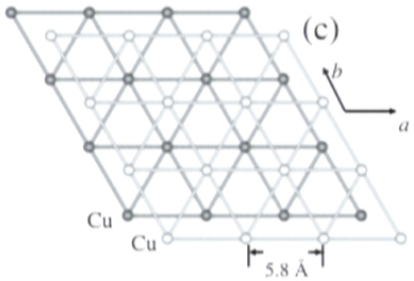


Spinons inherit the Fermi Surface of Electrons

PRL **106,** 147204 (2011) PHYSICAL REVIEW LETTERS week ending 8 APRIL 2011

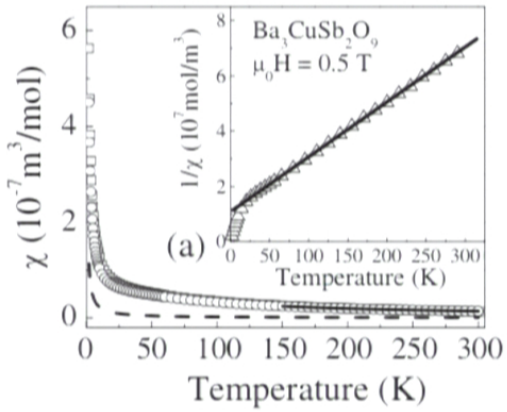
Spin Liquid State in the $S = 1/2$ Triangular Lattice $Ba_3CuSb_2O_9$

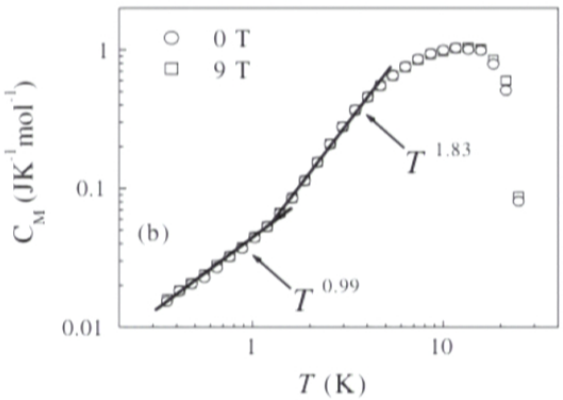
H. D. Zhou,[1,*] E. S. Choi,[1] G. Li,[1] L. Balicas,[1] C. R. Wiebe,[1,2,3] Y. Qiu,[4,5] J. R. D. Copley,[4] and J. S. Gardner[4,6]



$$\theta_{CW} = -55 \text{ K.}$$

$$\gamma = 43.4 \text{ mJ K}^{-2} \text{ mol}^{-1}$$



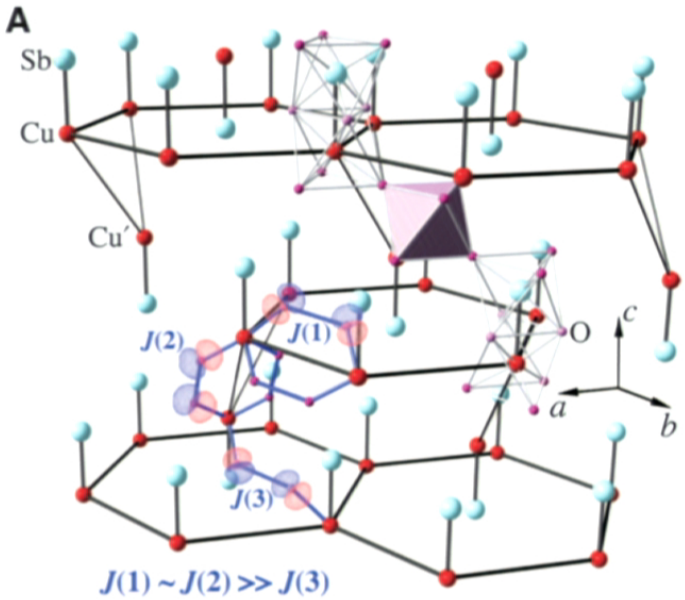


Recent Experiments: $Ba_3CuSb_2O_9$

Spin-Orbital Short-Range Order on a Honeycomb-Based Lattice

SCIENCE VOL 336 4 MAY 2012

S. Nakatsuji,[1*] K. Kuga,[1] K. Kimura,[1] R. Satake,[2] N. Katayama,[2] E. Nishibori,[2] H. Sawa,[2] R. Ishii,[3] M. Hagiwara,[3] F. Bridges,[4] T. U. Ito,[5] W. Higemoto,[5] Y. Karaki,[6] M. Halim,[7] A. A. Nugroho,[7] J. A. Rodriguez-Rivera,[8,9] M. A. Green,[8,9] C. Broholm[8,10]



S. Nakatsuji, ... C. Broholm et al, Science (2012)

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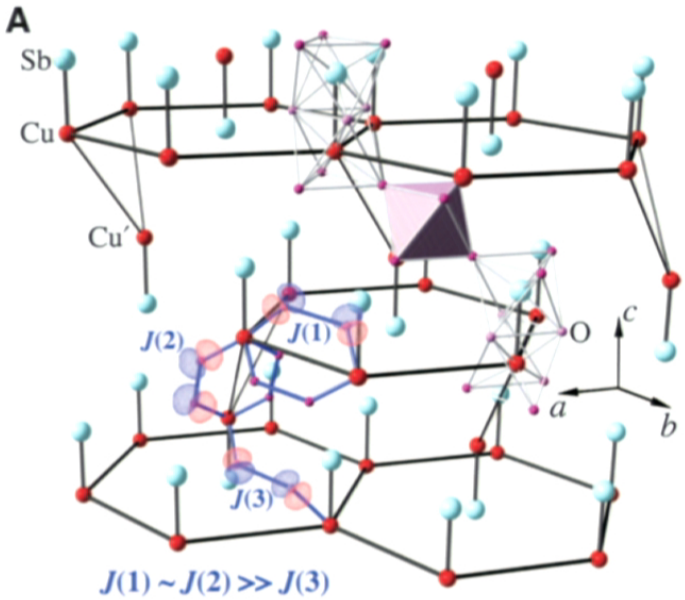


Nakatsuji; Tuesday HFM conference

S. Nakatsuji, ... C. Broholm et al, Science (2012)

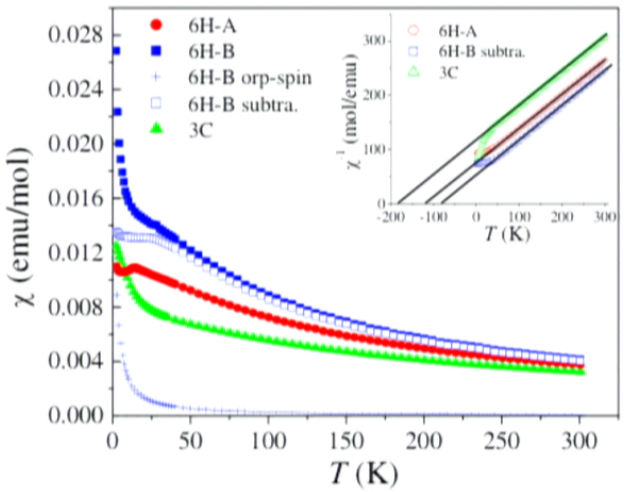
PRL 107, 197204 (2011) PHYSICAL REVIEW LETTERS week ending 4 NOVEMBER 2011

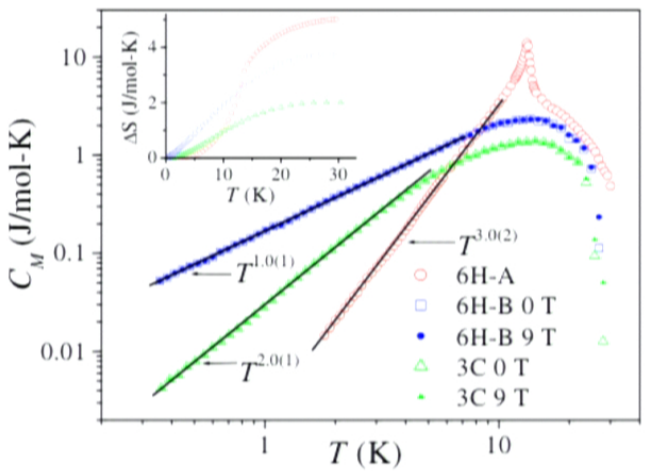
High-Pressure Sequence of $Ba_3NiSb_2O_9$ Structural Phases: New $S = 1$ Quantum Spin Liquids Based on Ni^{2+}

J. G. Cheng,[1] G. Li,[2] L. Balicas,[2] J. S. Zhou,[1] J. B. Goodenough,[1] Cenke Xu,[3] and H. D. Zhou[2,*]

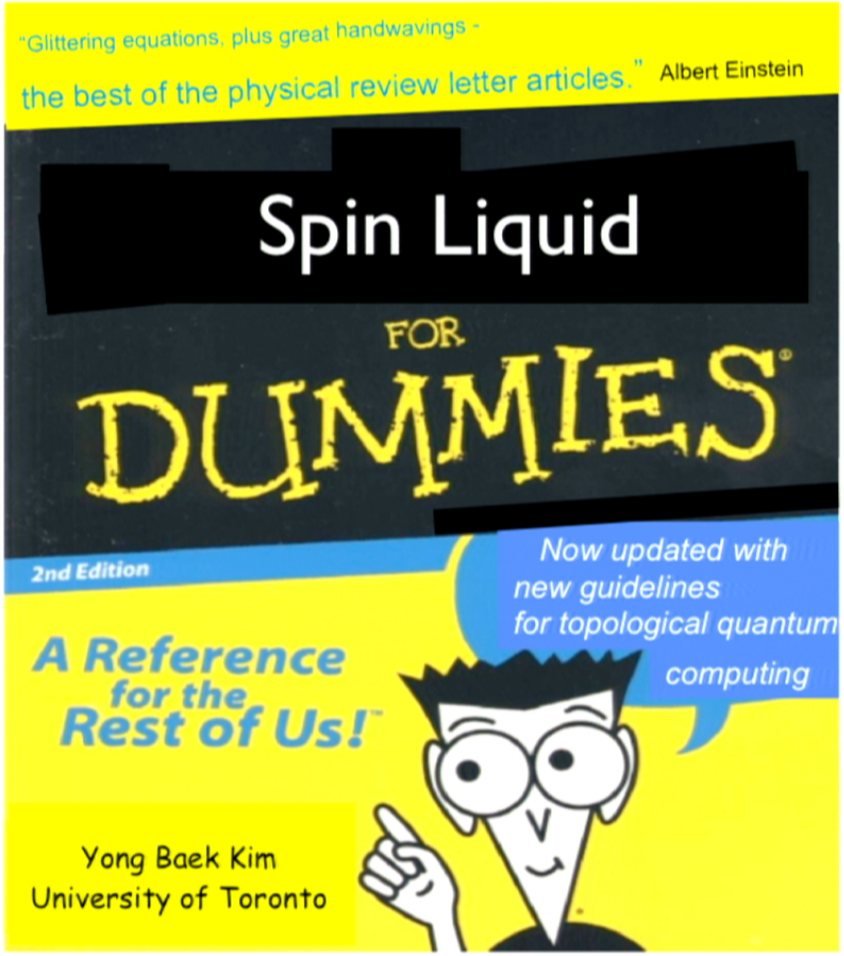
θ_{CW} of -75.5 (6*H*-B) K

$\gamma = 168$ mJ/mol K^2





A Micky Mouse version of "Theory"


"Glittering equations, plus great handwavings -
the best of the physical review letter articles." Albert Einstein
Spin Liquid
FOR
DUMMIES
2nd Edition
Now updated with
new guidelines
for topological quantum
computing
A Reference
for the
Rest of Us!
Yong Baek Kim
University of Toronto

Variational Wavefunctions for Spin Liquids

Variational Wavefunctions for Spin Liquids

Slave-Particle Approach

$$\vec{S}_i = \frac{1}{2} f^\dagger_{i\alpha} \vec{\sigma}_{\alpha\beta} f_{i\beta}$$ with the constraint $$\sum_\alpha f^\dagger_{i\alpha} f_{i\alpha} = 1 \quad \alpha, \beta = \{\uparrow, \downarrow\}$$

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$$H = \sum_{ij} J_{ij} \; \vec{S}_i \cdot \vec{S}_j \quad \Longrightarrow \quad H = \sum_{ij} J_{ij} \; f^\dagger_{i\alpha} f^\dagger_{i\beta} f_{j\beta} f_{j\alpha} + \cdots$$

fermion-fermion interaction

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Mean-Field Theory

$\chi_{ij} \sim \langle f^\dagger_{i\alpha} f_{i\alpha} \rangle$ fermion "kinetic" energy dynamically generated

$\Delta_{ij} \sim \langle \epsilon_{\alpha\beta} f_{i\alpha} f_{j\beta} \rangle$ possible pairing correlation

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$$H_{MF} = \sum_{ij} [\chi_{ij} f^\dagger_{j\alpha} f_{i\alpha} + \Delta_{ij} \epsilon_{\alpha\beta} f^\dagger_{j\beta} f^\dagger_{i\alpha} + h.c.]$$

$\sum_\alpha \langle f^\dagger_{i\alpha} f_{i\alpha} \rangle = 1$ Constraint imposed only on average

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Not every choice of χ_{ij} and Δ_{ij} is independent

The mean-field Hamiltonian is invariant under certain gauge and lattice symmetry transformation

Classification requires "equivalence" study --
PSG (projected symmetry Group) analysis by X. G. Wen

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Topological Order and Quantum Order

(gapped) (gapless)

Variational Wavefunctions for Spin Liquids

Projected Wavefunction (project out double-occupancy)

$$\Psi_{proj} = P_G \Psi_{MF}$$

χ_{ij} and Δ_{ij} can be taken as variational parameters

Find χ_{ij} and Δ_{ij} that give the lowest energy solution

Ranking of ground state energy for Ψ_{MF} and Ψ_{proj} changes quite often

Possible Spin Liquid Phases; Why Gauge Theory ?

Possible Spin Liquid Phases; Why Gauge Theory ?

1) U(1) Spin Liquid $\Delta_{ij} = 0 \quad \chi_{ij} \neq 0$

$$H \sim \sum_{ij} \chi_{ij} f^\dagger_{i\alpha} f_{j\alpha} \sim \sum_{ij} |\chi| e^{i a_{ij}} f^\dagger_{i\alpha} f_{j\alpha}$$

$$f_{i\alpha} \to f_{i\alpha} e^{i\theta_i}$$
$$a_{ij} \to a_{ij} + \theta_i - \theta_j$$

U(1) gauge invariance

$f_{i\alpha}$ spinons interact with a U(1) gauge field

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2) Z_2 Spin Liquid $\Delta_{ij} \neq 0 \quad \chi_{ij} \neq 0$ (a BCS state of the spinons)

Only $f_{i\alpha} \to \pm f_{i\alpha}$ is the gauge symmetry Only $a_{ij} = 0, \pi$ are allowed

Ising gauge field $e^{ia_{ij}} = \sigma_{ij} = \pm 1$

$$f_{i\alpha} \to \epsilon_i f_{i\alpha} \qquad \epsilon_i = \pm 1$$
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Possible Spin Liquid Phases; Wavefunctions

1) U(1) Spin Liquid

$$\Psi_{proj} \sim P_G \Psi_{\text{“free” fermions}}$$

2) Z_2 Spin Liquid

$$\Psi_{proj} \sim P_G \Psi_{BCS}$$

Wavefunction of a Z_2 Spin Liquid

- BCS superconductor (L x L lattice)

average number of electrons per site = one (Half-filled)

$g(\mathbf{r}-\mathbf{r}')$ ⇔ Cooper pair wave function

BCS wave function $|BCS\rangle \propto e^{(\sum_{\mathbf{r},\mathbf{r}'} g(\mathbf{r}-\mathbf{r}') c^\dagger_{\mathbf{r}\uparrow} c^\dagger_{\mathbf{r}'\downarrow})^{\frac{L^2}{2}}} |0\rangle$

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- RVB Spin Liquid wave function $|RVB\rangle = P_G |BCS\rangle$

P_G projection: exactly-one-particle per site

Insulator: valence bond ~ incoherent Cooper pair

Elementary Excitations

- Elementary excitations in superconductors

  Bogoliubov quasiparticles (zero average charge, S=1/2)

- Elementary excitations in the spin liquid state

  P_G (Bogoliubov quasiparticles) = Spinons (Q=0,S=1/2)

- Fractionalization of electrons !

Non-trivial excitations

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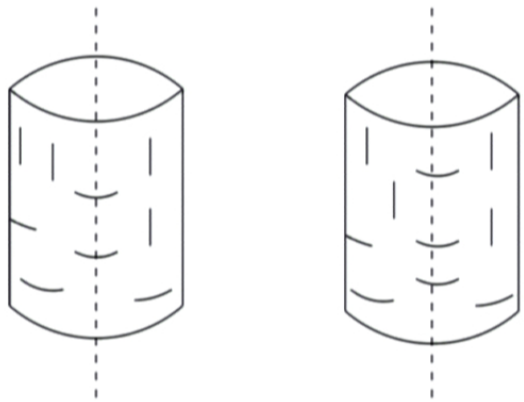
Non-trivial excitations

Short 'Coherence Length' Limit

TWO topologically distinct valence bond coverings

intersecting even number of dimers



intersecting odd number of dimers

Non-trivial ground state degeneracy

Properties of Spin Liquid Phases

1) U(1) Spin Liquid

	Gapped	Gapless	Specific Heat
2D	Not stable	Stable Fermi Surface Dirac point	$C_{MF}(T) \sim T$ $C_{MF}(T) \sim T^2$
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Gauge field fluctuation effect

Gapless Cases

2D Fermi surface case: $C(T) \sim T^{2/3}$

3D Fermi surface case: $C(T) \sim T \ln(1/T)$

Gapped Case ("photons")

3D $C(T) \sim T^3$

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2) Z_2 Spin Liquid

Stable in 2D and 3D

Can support Dirac point (d-wave gap) and line nodes etc.
(just like superconductor)

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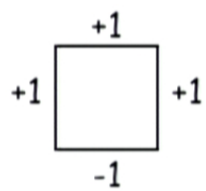
Stable in 2D and 3D

Can support Dirac point (d-wave gap) and line nodes etc.
(just like superconductor)

i) Fully gapped case: $C(T) \sim e^{-\Delta_{\rm vison}/T}$

$\Delta_{\rm vison}$ ~ energy gap for visons
- topological defect in σ_{ij}



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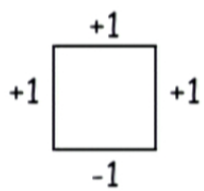
Can support Dirac point (d-wave gap) and line nodes etc.
(just like superconductor)

i) Fully gapped case: $C(T) \sim e^{-\Delta_{\rm vison}/T}$

$\Delta_{\rm vison}$ ~ energy gap for visons
- topological defect in σ_{ij}

charge-neutral spinless;
only carries entropy



Properties of Spin Liquid Phases

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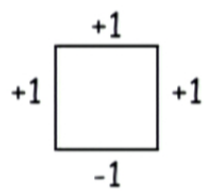

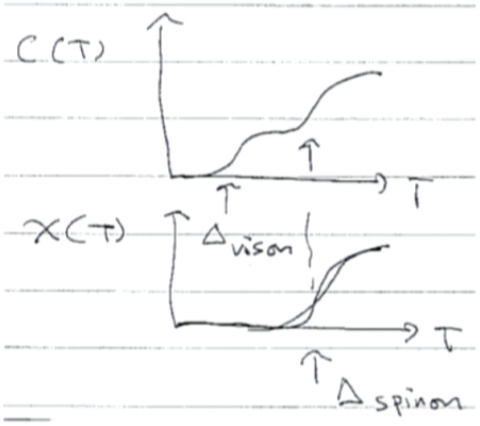

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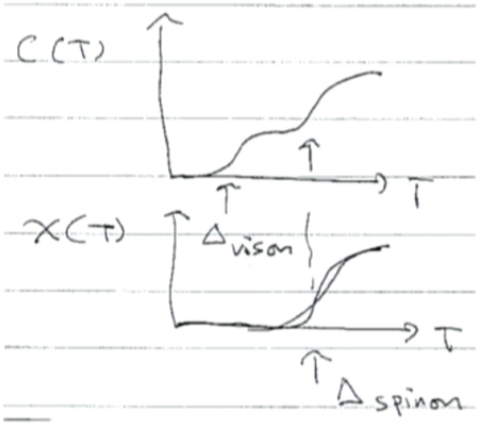

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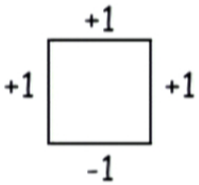


ii) Gapless case: $C(T) \sim T^2$ Dirac point in 2D (d-wave);
Line Nodes in 3D

Properties of Spin Liquid Phases

Notice C(T) ~ T^{α} often in candidate spin liquid materials

indicates the presence of low energy spin-carrying excitations ?

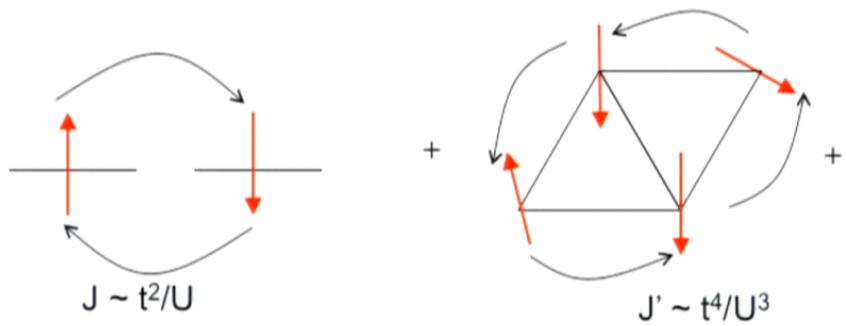
Spinons ?

Organic Materials

Organics: Triangular Lattice

Motrunich $H \sim H_{\text{heisenberg}} + H_{\text{ring}} + \cdots$

S. S. Lee + P. A. Lee,
M. Imada,
A.-M. Tremblay,



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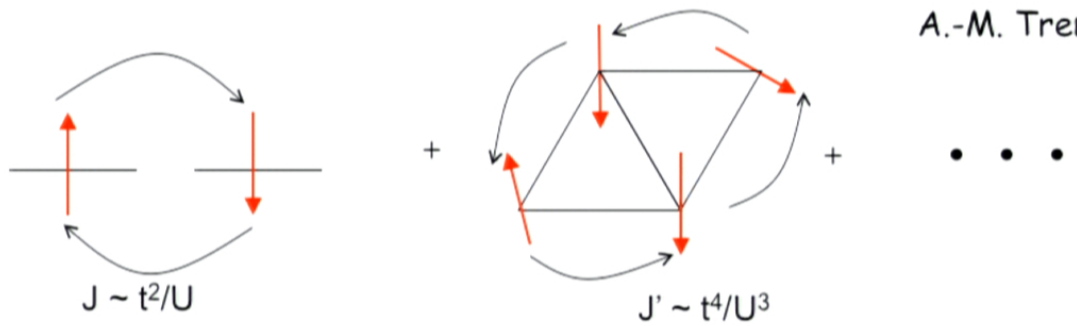

Variational wavefunction - Uniform RVB: $\chi_{ij} = \chi \quad \Delta_{ij} = 0$

Fermi surface of spinons $C_{\text{MF}}(T) \sim \gamma T$ but ... $C_{\text{fluc}}(T) \sim T^{2/3}$

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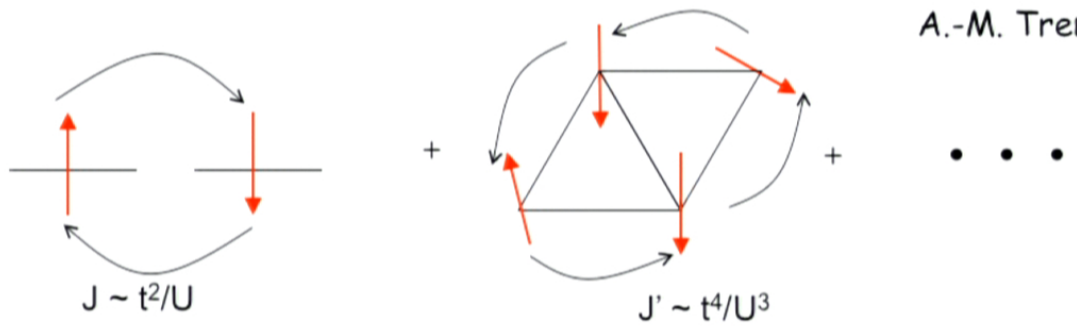

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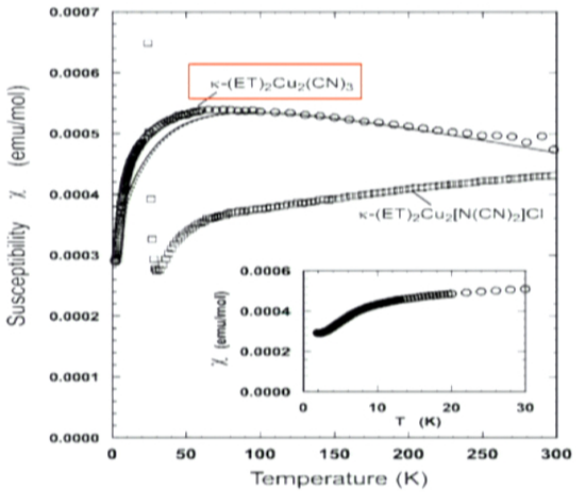
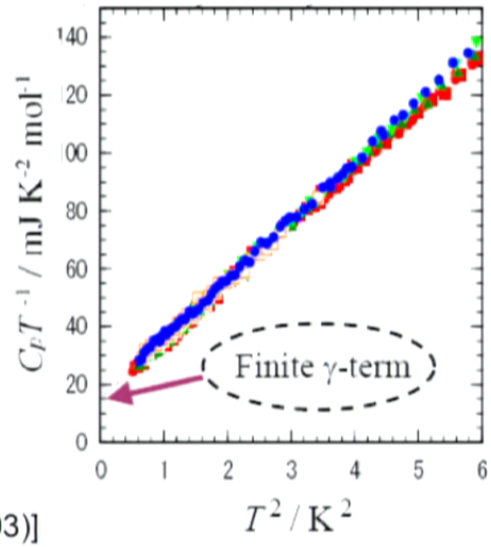
T. Senthil: Charge fluctuations near Metal-Insulator transition

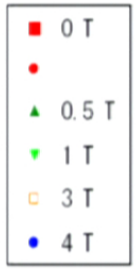
$$C_{\text{fluc}}(T) \sim T^{2/3} \rightarrow C_{\text{fluc}} \sim T \ln(1/T)$$

Organics: Triangular Lattice

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$







[Y. Shimizu et al., PRL 91, 107001 (03)]

No magnetic ordering;
Constant T=0 susceptibility;
C(T) ~ T;
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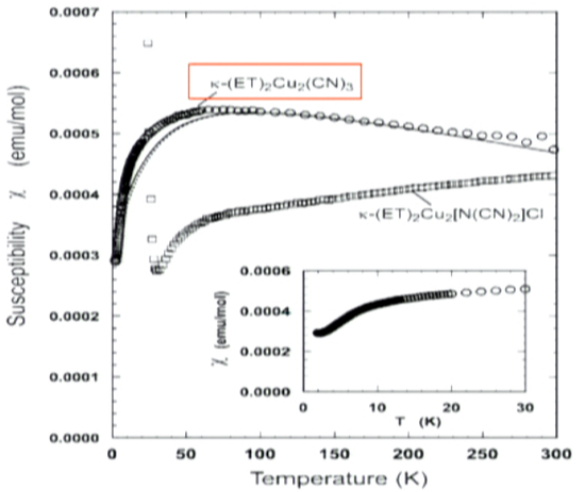

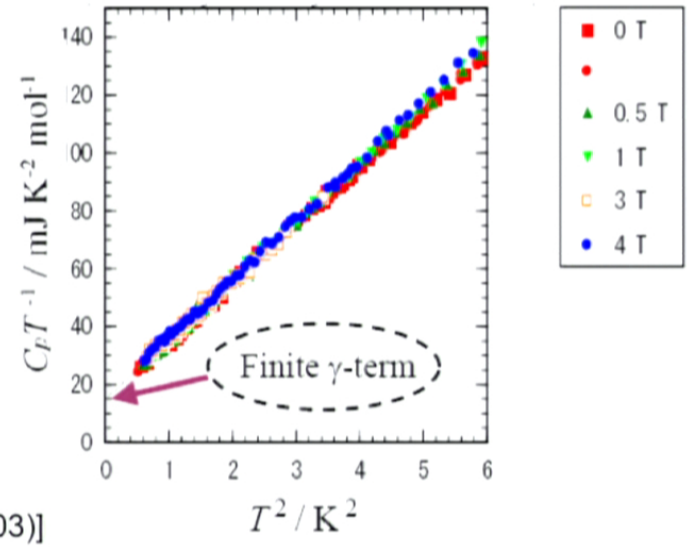

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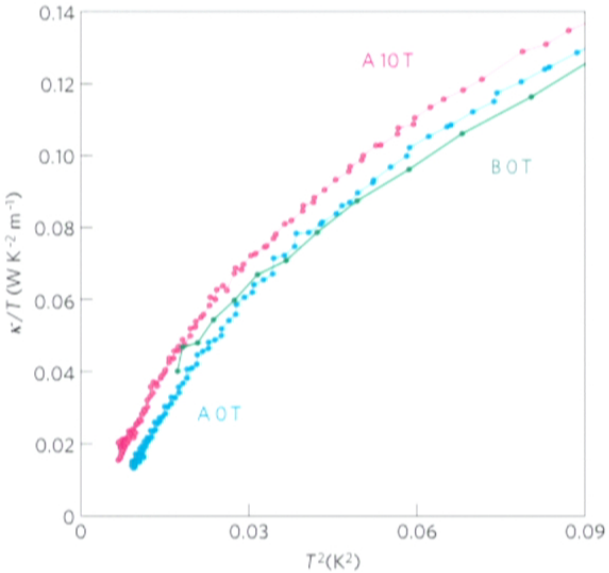
$C_{fluc}(T) \sim T^{2/3}$ vs $T \ln(1/T)$?

$1/T_1$ inhomogeneous ?
more than one relaxation time ?

Organics: Triangular Lattice; Further Challenges



κ/T goes to zero as T -> 0 ?

(Y. Matsuda, Nature Physics)

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C(T) shows a kink around 5K ?
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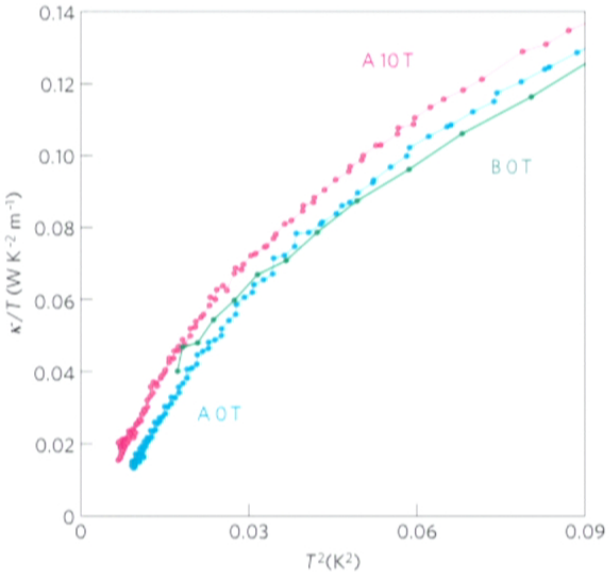

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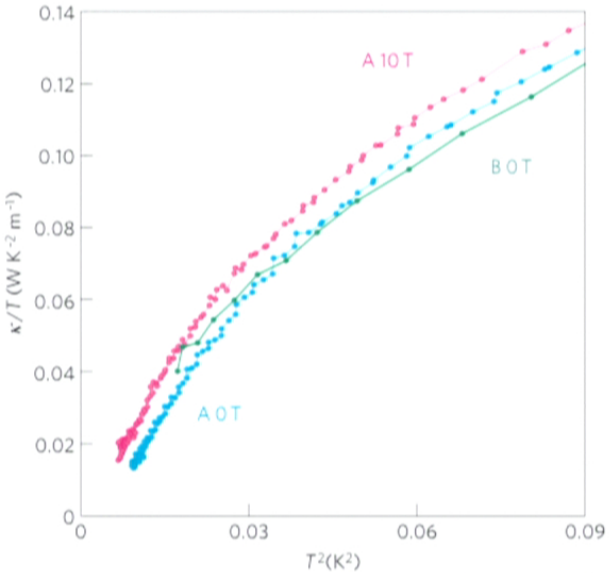

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Z_2 Spin Liquid ?

T. Senthil + P. A. Lee -- d-wave nematic spin liquid

S. S. Lee + P. A. Lee -- Finite momentum Pairing via amperian pairing

V. Galitski + Y. B. Kim -- spin-triplet pairing (no change in Knight shift)

Organics: Triangular Lattice

$EtMe_3Sb[Pd(dmit)_2]_2$

C(T) ~ T

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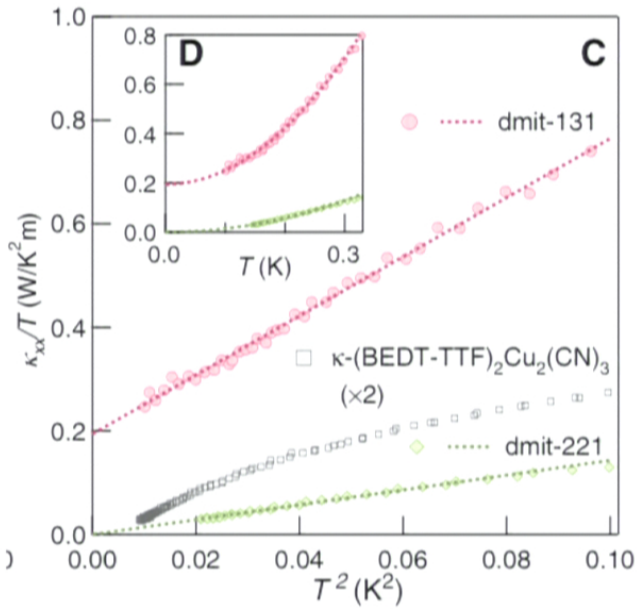

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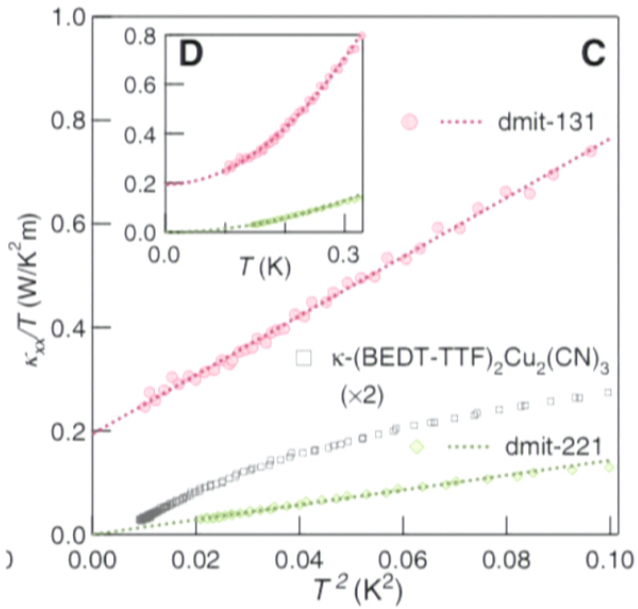

NMR data ?
Itou; Wednesday
HFM conference

Herbertsmithite: Kagome

Kagome Lattice; Herbertsmithite

$$ZnCu_3(OH)_6Cl_2$$

$C(T) \sim T^{\alpha} \quad \alpha = ?$ constant susceptibility (extrinsic?)

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Ying Ran + P. A. Lee + X. G. Wen; U(1) Dirac Spin Liquid

spinons have Dirac spectrum $C(T) \sim T^2 \quad \chi \sim T$

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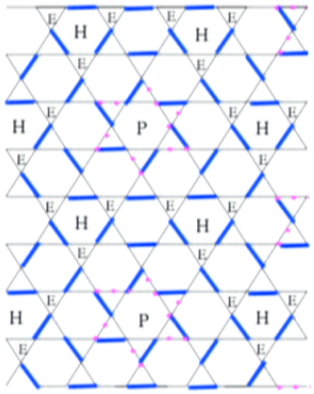
G. Vidal



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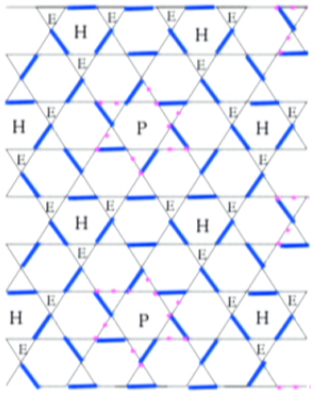

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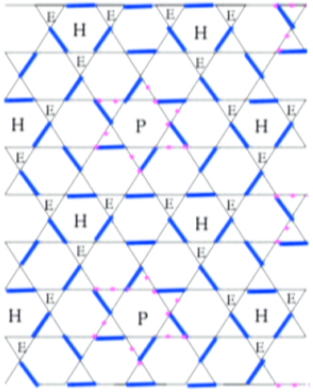


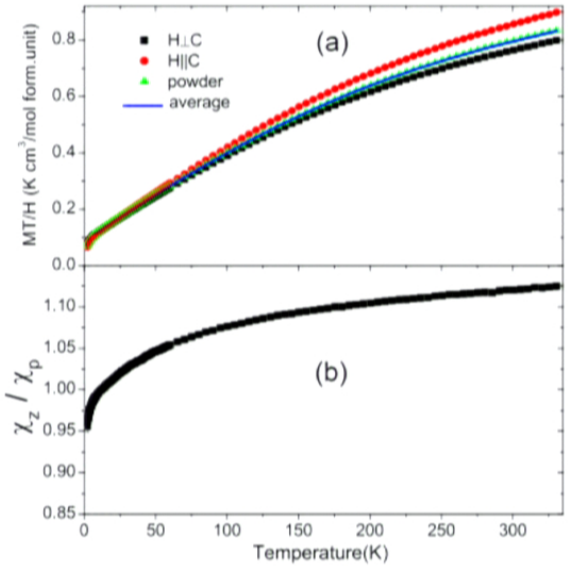
Recent DMRG study by White + Huse;
Spin Liquid with finite spin gap

Z_2 Spin Liquid ?

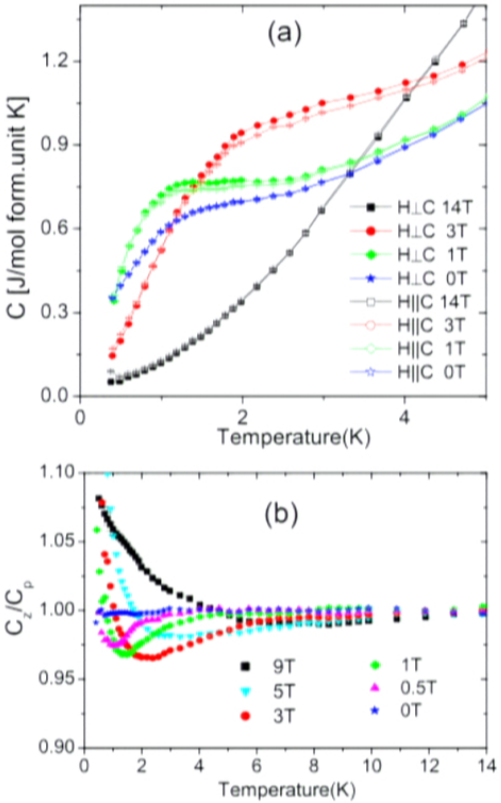
White; Thursday
HFM conference

Recent Single Crystal Data: Herbertsmithite

T. Han, S. Chu, Young S. Lee (2012)



$$MT/H = \chi T$$



Heat Capacity

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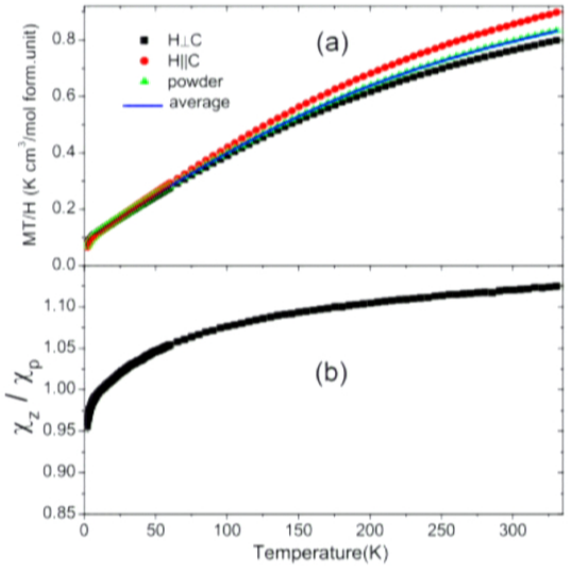
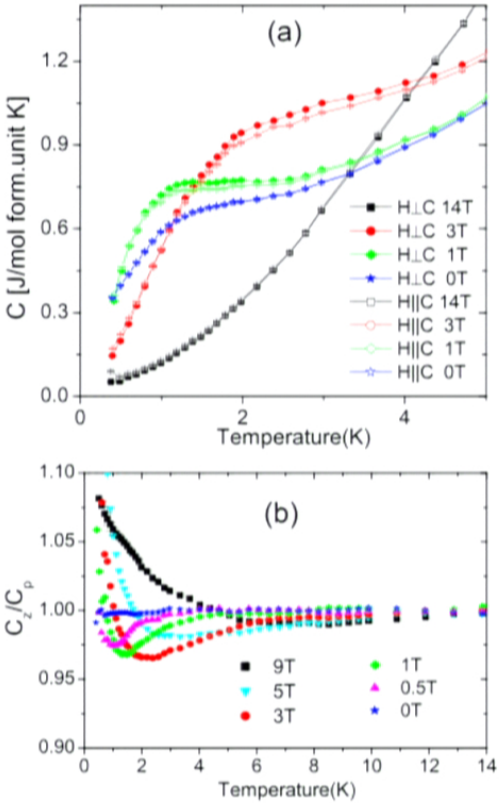


$$MT/H = \chi T$$

Young Lee; Thursday

Observation of two-spinon continuum !

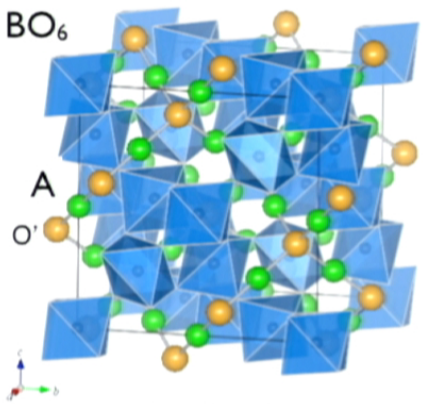


Heat Capacity

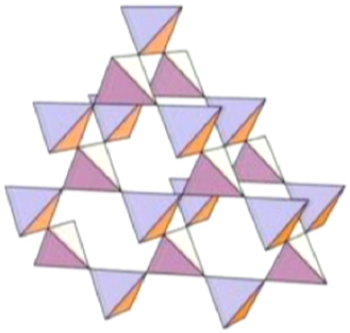
$Na_4Ir_3O_8$: Hyperkagome

“Quantum Spin Ice”

Rare earth pyrochlores



$A_2B_2O_7$



A (B) sublattice

$(A^{3+})_2(B^{4+})_2(O^{2-})_7$

Rare earth A-site element

Local atomic picture

4f electrons are well localized

Strong spin-orbit coupling:
local J eigenstates split by crystal field

Ground state is often a doublet

Effective S=1/2 description with
local quantization axis

Model for rare earth magnetism

$$H = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z$$

Classical spin ice

$$-J_{\pm} \sum_{\langle i,j \rangle} \left(S_i^+ S_j^- + S_i^- S_j^+ \right)$$

$$+ J_{z\pm} \sum_{\langle i,j \rangle} \left[S_i^z \left(\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^- \right) + i \leftrightarrow j \right]$$

$$+ J_{\pm\pm} \sum_{\langle i,j \rangle} \left(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^- \right)$$

Quantum fluctuations

= "Quantum Spin Ice"

S. Curnoe 2008,
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Savary + Balents 2011

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Effective Hamiltonian

$$H_0 = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z$$

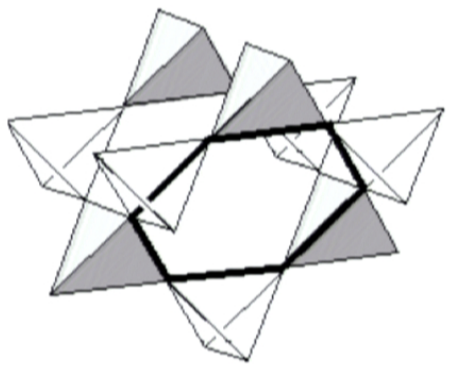
spin ice Hamiltonian

$$H_{pert} = -J_{\pm} \sum_{\langle i,j \rangle} \left(S_i^+ S_j^- + S_i^- S_j^+ \right)$$

quantum fluctuations

$$H_{ring} = -\frac{12 J_{\pm}^3}{J_{zz}^2} \sum_{\{1,..6\} \in \hexagon} \left(S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + \text{h.c.} \right)$$

Ring exchange



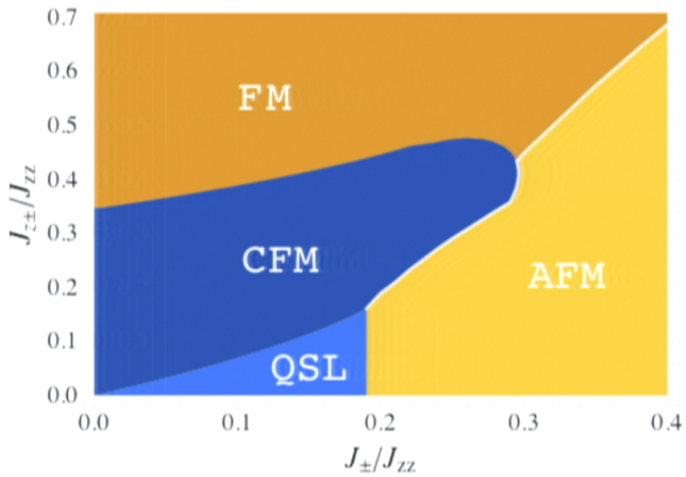
Gauge Theory

- Perturbation in J_{+-}
- The effective Hamiltonian can be mapped to a gauge theory

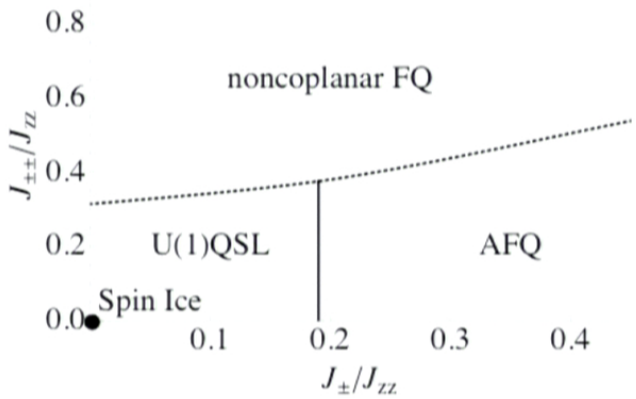
$$S_i^z = E_{ab} \qquad S_i^{\pm} = e^{\pm i A_{ab}}$$

$$H_{\text{ring}} = -K \sum_{\text{hex}} \cos(\nabla \times A) + U \sum_{\langle ab \rangle} \left(E_{ab}^2 - \frac{1}{4} \right)$$

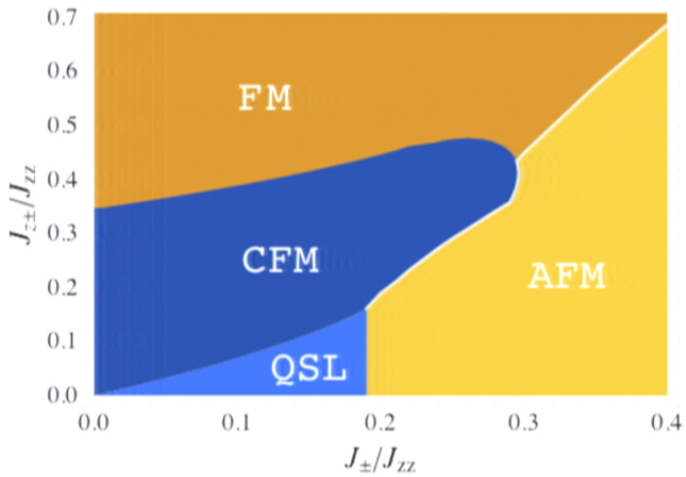
compact U(1) gauge theory



Savary, Balents 2011

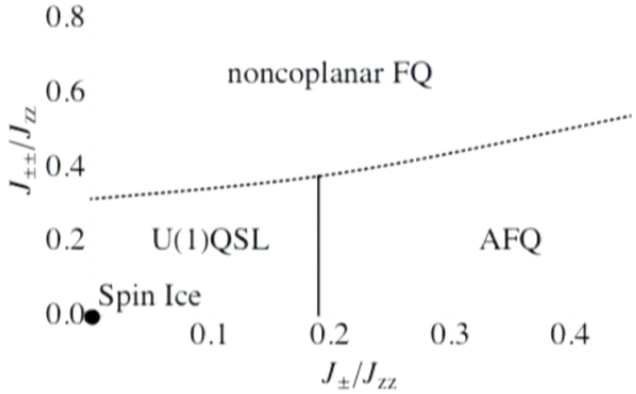


S.-B.Lee, Onoda, Balents, 2012



Savary, Balents 2011

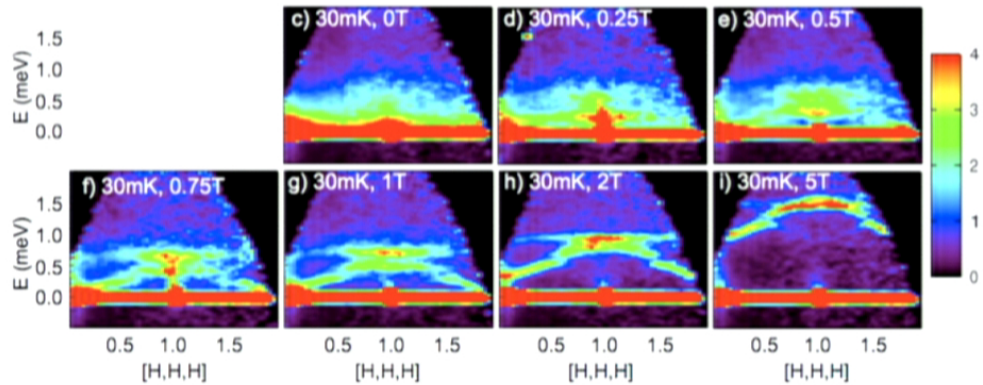
Savary; Monday
HFM conference



S.-B.Lee, Onoda,
Balents, 2012

Onoda (SungBin Lee);
Monday (Poster)
HFM conference

$Yb_2Ti_2O_7$ K.A. Ross, et al, 2009, 2011



- Spin waves appear absent in low field, but emerge for B>0.5T
  - a low field spin liquid state?

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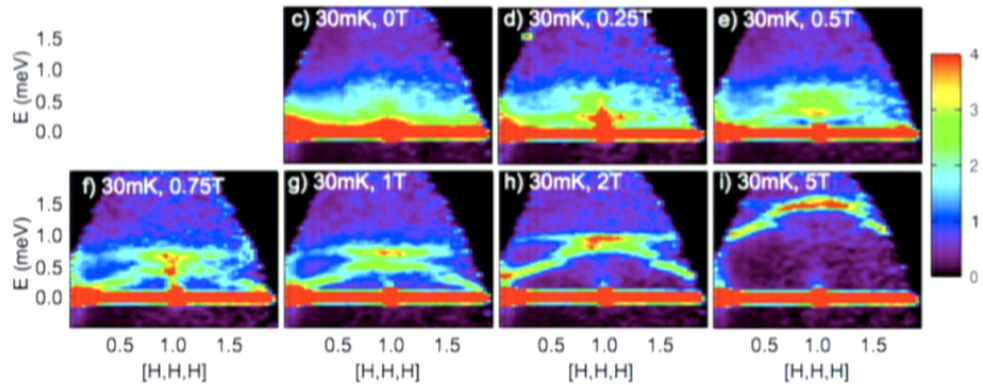

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Ross, Singh; Monday
Experiment, Theory

Broholm; Tuesday
Pr-based pyrochlore

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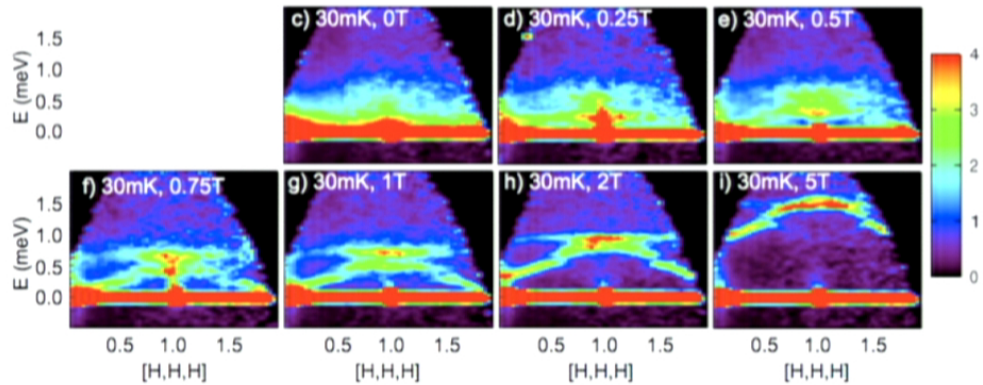

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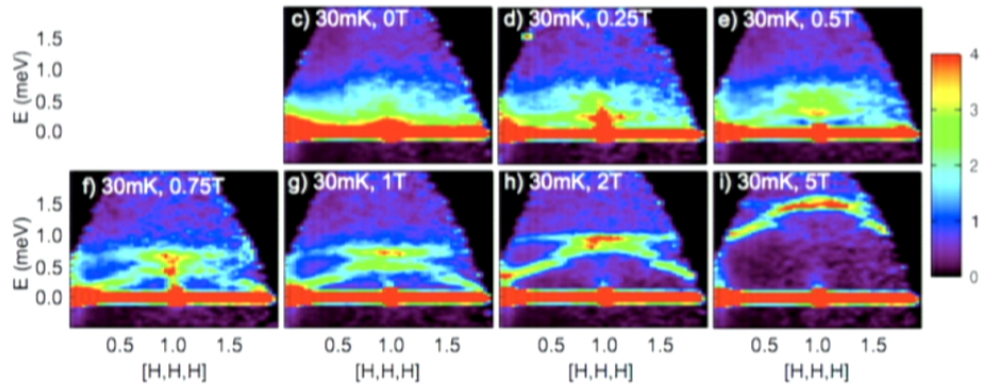

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