

Title: Where Are We in Spin Liquid Research?

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URL: <http://pirsa.org/12020095>

Abstract: <span>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.</span>



# Where are we in spin liquid research ?

Yong Baek Kim  
University of Toronto

Perimeter Institute, March 16, 2012



"Glittering equations, plus great handwavings -  
the best of the physical review letter articles." Albert Einstein

# Spin Liquid

FOR  
**DUMMIES**<sup>®</sup>

2nd Edition

**A Reference  
for the  
Rest of Us!**

Now updated with  
new guidelines  
for topological quantum  
computing

Yong Baek Kim  
University of Toronto



# Outline

What is "Quantum" Spin Liquid ?

Overview of Experiments

Overview of Theories

Interpretation of Experiments

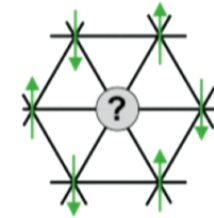
Topological Mott Insulator



# What is Quantum Spin Liquid ?

1) **Different** from Cooperative Paramagnet

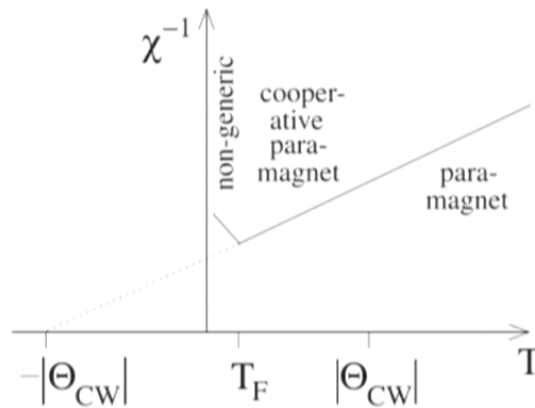
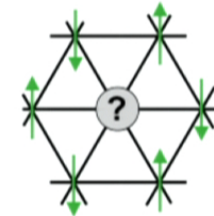
Large number of equally unhappy (classical) ground states  $\sim e^{\alpha N}$



# What is Quantum Spin Liquid ?

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



$\Theta_{CW}$ : Curie-Weiss temperature

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

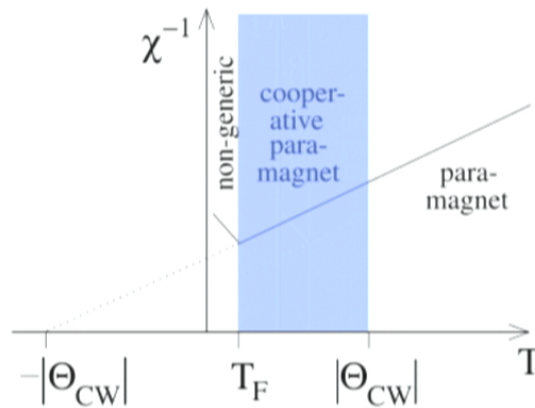
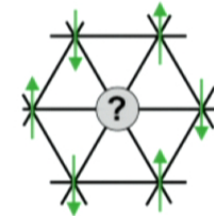
$$f = \frac{|\Theta_{CW}|}{T_F} \quad \text{useful diagnostic of frustration}$$

$f \gg 1$  **strong frustration**

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highly degenerate classical ground state manifold

"classical spin liquid"

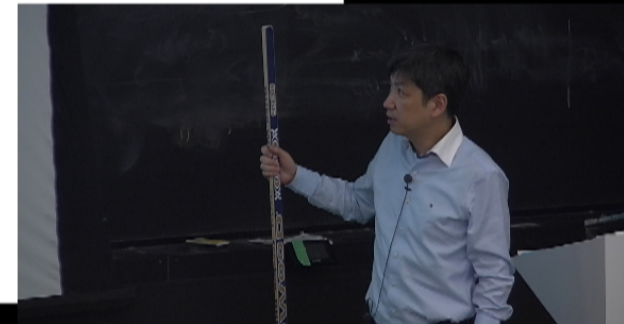


# What is Quantum Spin Liquid ?

2) **Different** from boring incoherent (high temp) Paramagnet

Spins fluctuate independently; don't talk to each other

No coherent excitations



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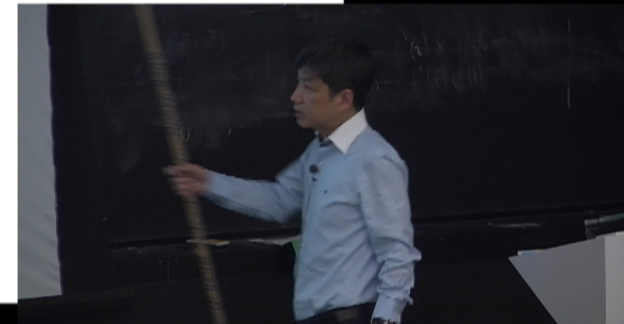
No coherent excitations

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 ?

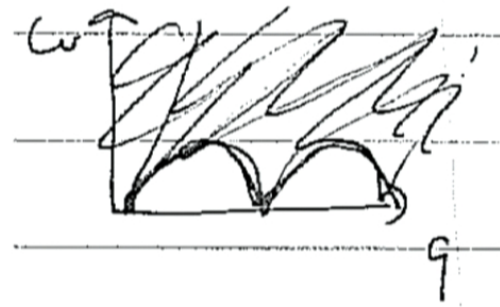
Neutron Scattering -- Spin-1 excitations

Spinon-Antispinon pair excitations

Well-defined dispersion --> Threshold energy for pair excitations

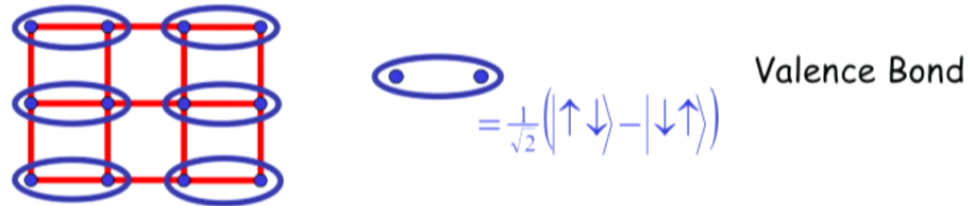
$$\omega_{\mathbf{q}} \sim \min [\varepsilon_{\frac{\mathbf{q}}{2} + \mathbf{p}} + \varepsilon_{\frac{\mathbf{q}}{2} - \mathbf{p}}]$$

↑  
for all possible  $\mathbf{p}$



# Higher dimensions ? Quantum Paramagnet $\langle S \rangle = 0$

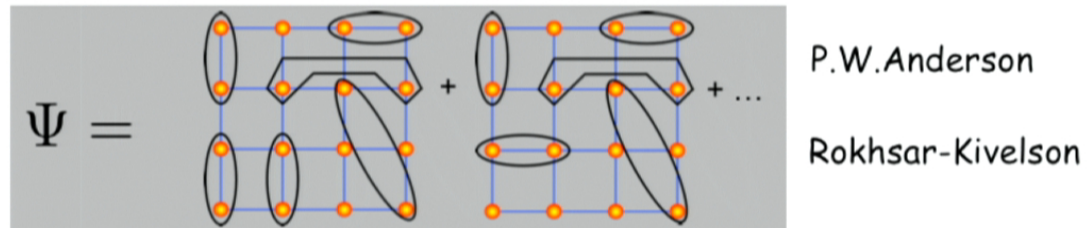
i) Translational symmetry breaking; **Valence Bond Solid (VBS)**



ii) No broken translational symmetry in Mott insulator

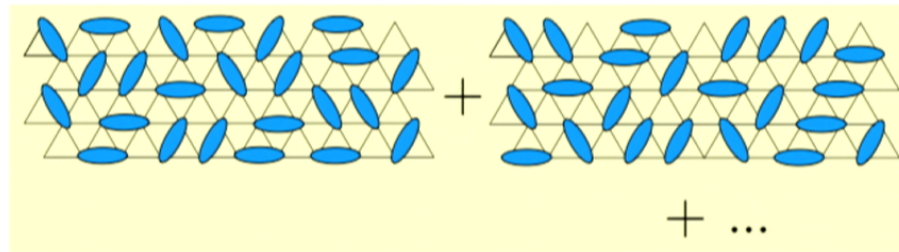
**Spin Liquid:** charge neutral spin-1/2 excitations with/without spin gap

**Resonating Valence Bond state (RVB):** Superposition of Valence Bond coverings



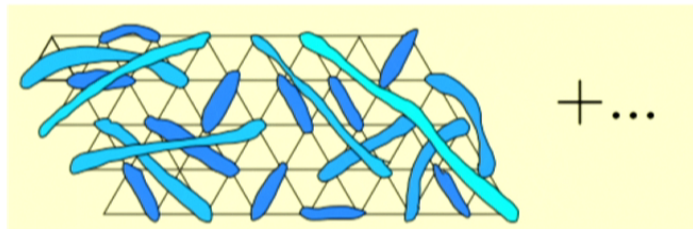
## Many Flavors of Spin Liquid States

Short-range  
RVB



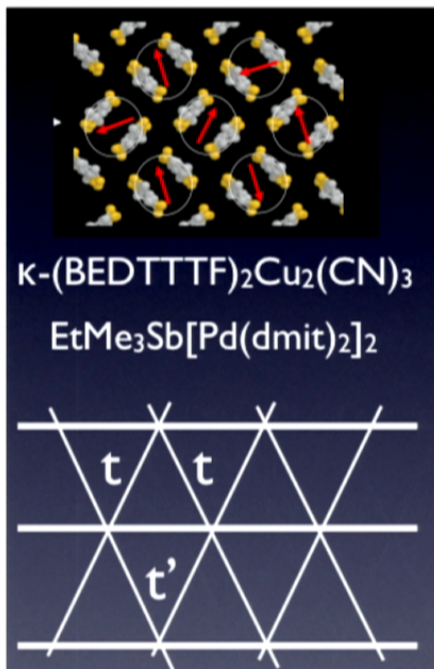
Gapped spinons, Finite spin gap  $\langle \mathbf{S}(\mathbf{r}) \cdot \mathbf{S}(\mathbf{0}) \rangle \sim e^{-r/\xi}$

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

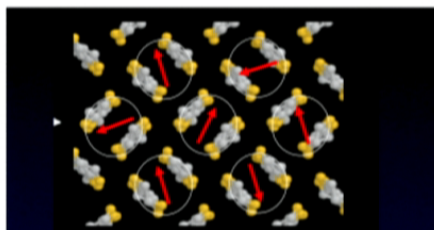


K. Kanoda  
R. Kato

# S=1/2 Candidate Materials for Spin Liquid

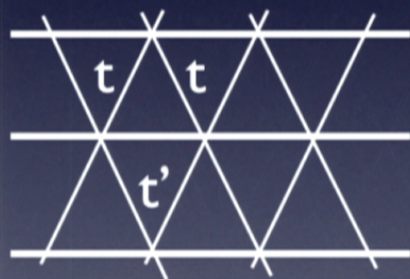
D. G. Nocera, Y. S. Lee

Z. Hiori



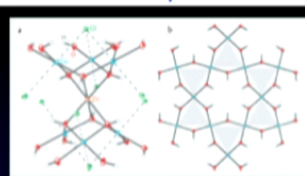
$\kappa$ -(BEDTTTF)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub>

EtMe<sub>3</sub>Sb[Pd(dmit)<sub>2</sub>]<sub>2</sub>

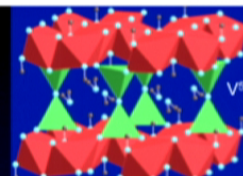


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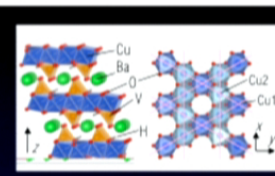
R. Kato



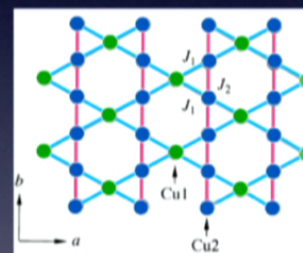
herbertsmithite



volborthite



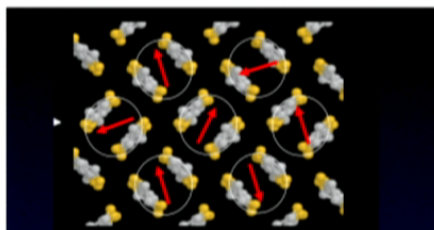
vesignieite



# S=1/2 Candidate Materials for Spin Liquid

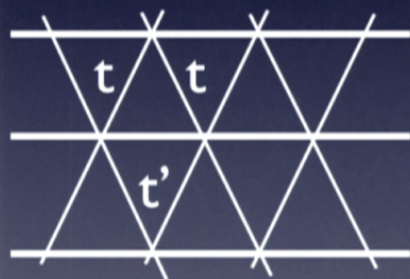
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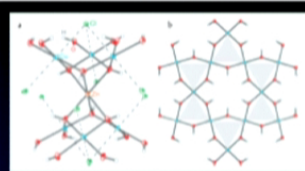
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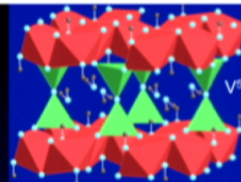


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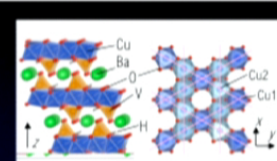
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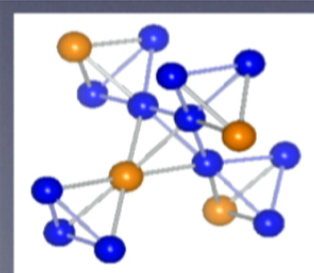
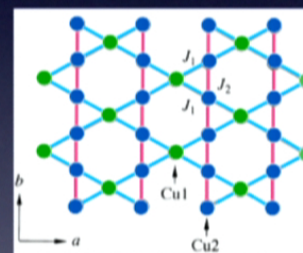
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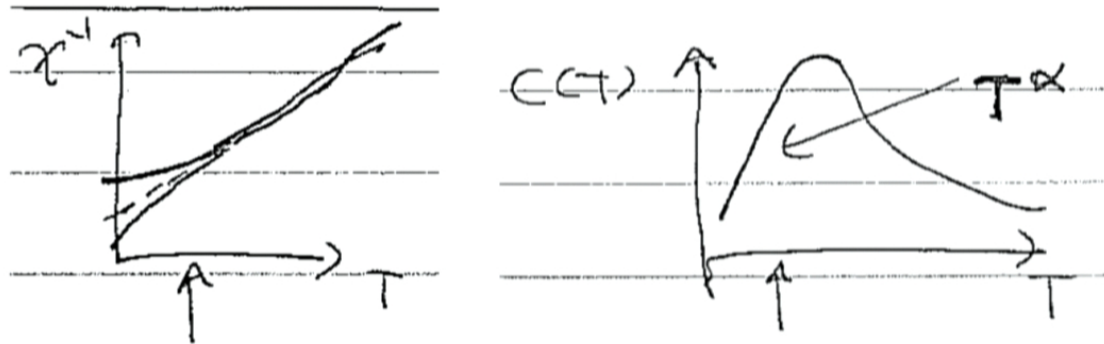


Na<sub>4</sub>Ir<sub>3</sub>O<sub>8</sub>

H. Takagi



## Typical Behaviors



### Charge Insulator

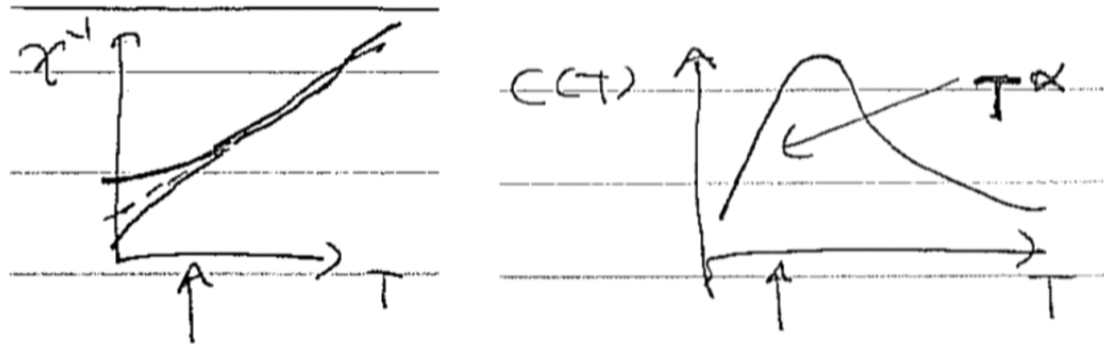
Constant susceptibility at low temperatures

Power-law specific heat

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

--> Spin Liquid with gapless spinon excitations ?

## Typical Behaviors



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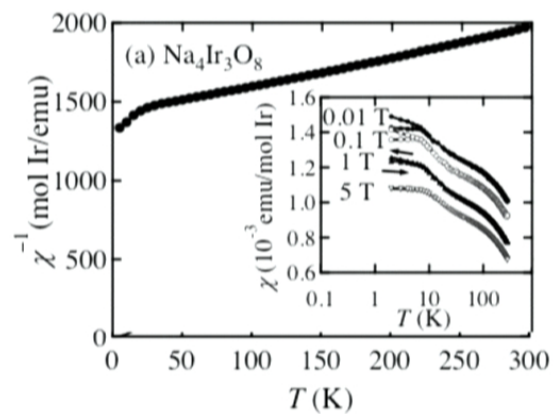
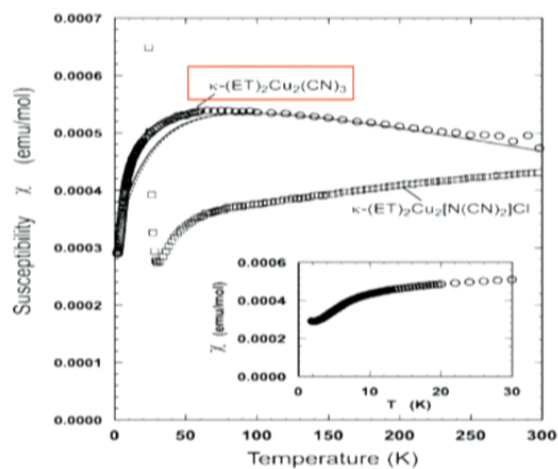
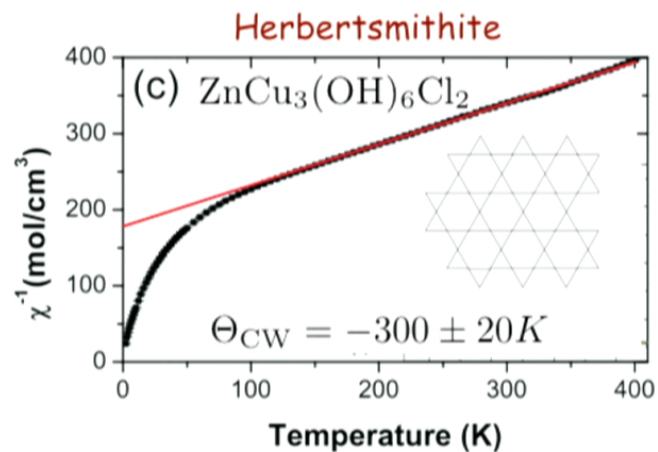
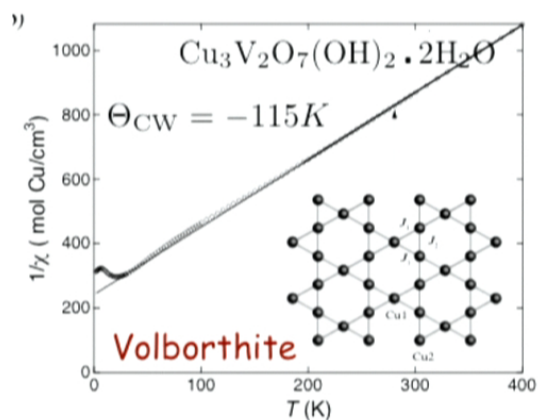
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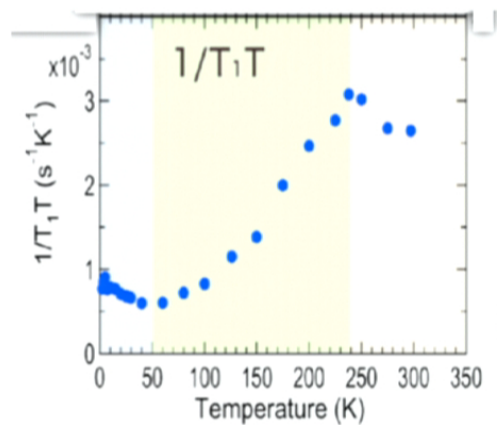
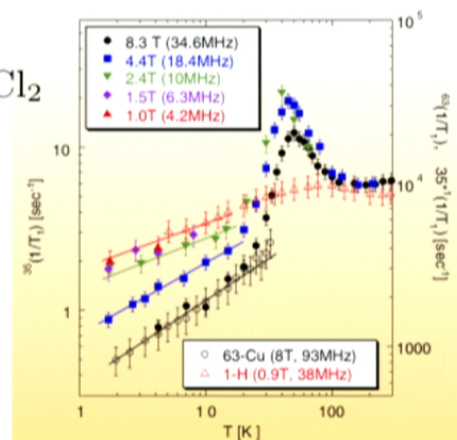
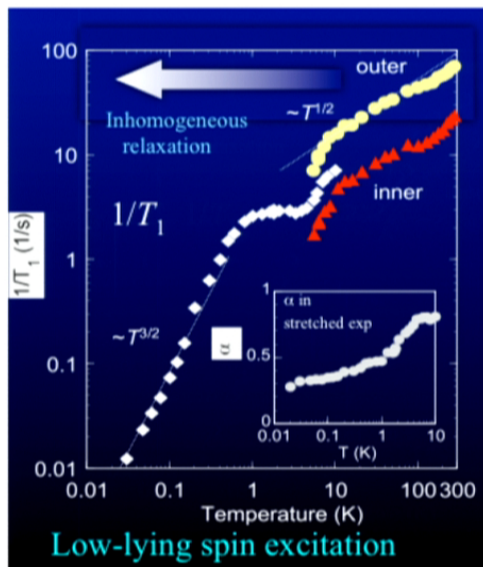
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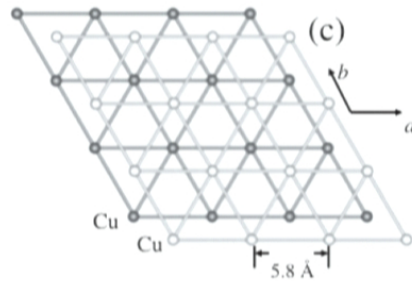
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# Spin Susceptibility



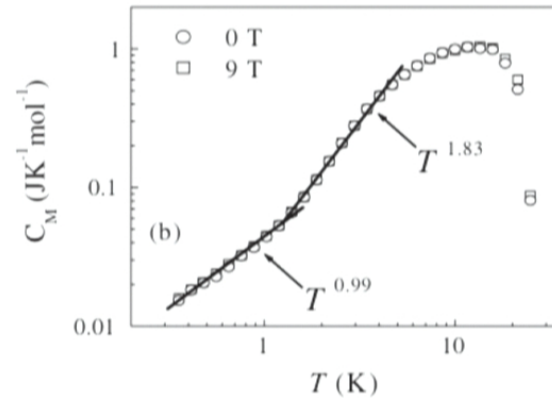
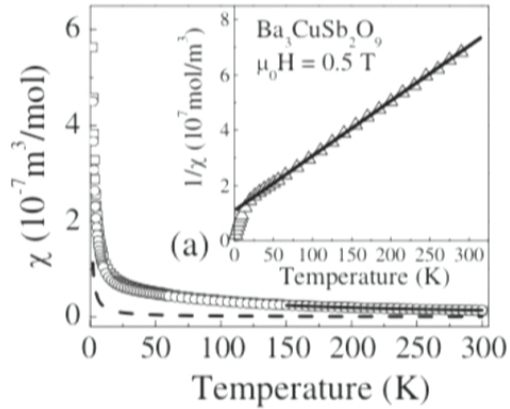
# Abundance of low energy spin excitations

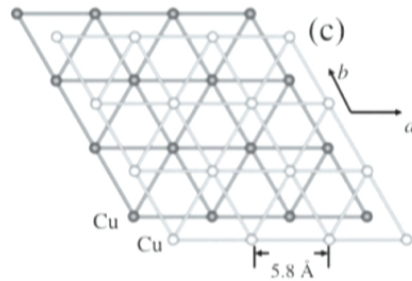


Spin Liquid State in the  $S = 1/2$  Triangular Lattice  $\text{Ba}_3\text{CuSb}_2\text{O}_9$ H. D. Zhou,<sup>1,\*</sup> E. S. Choi,<sup>1</sup> G. Li,<sup>1</sup> L. Balicas,<sup>1</sup> C. R. Wiebe,<sup>1,2,3</sup> Y. Qiu,<sup>4,5</sup> J. R. D. Copley,<sup>4</sup> and J. S. Gardner<sup>4,6</sup>

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

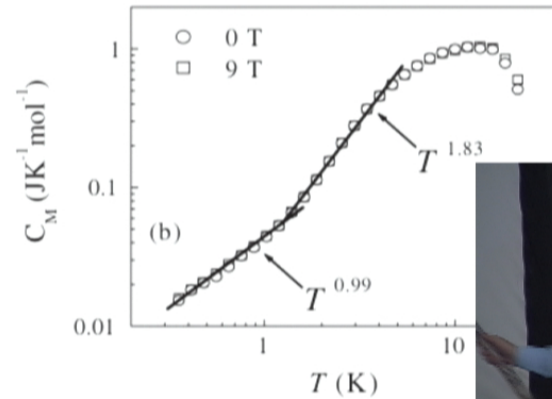
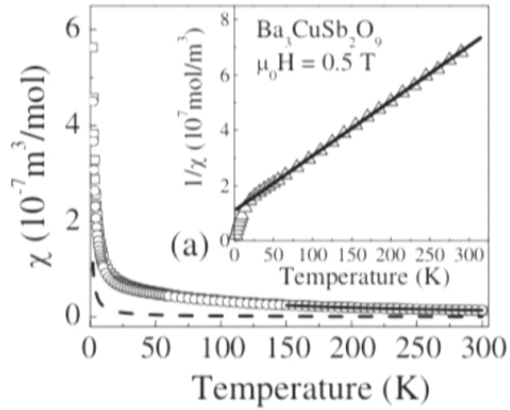
$$\gamma = 43.4 \text{ mJ K}^{-2} \text{ mol}^{-1}$$



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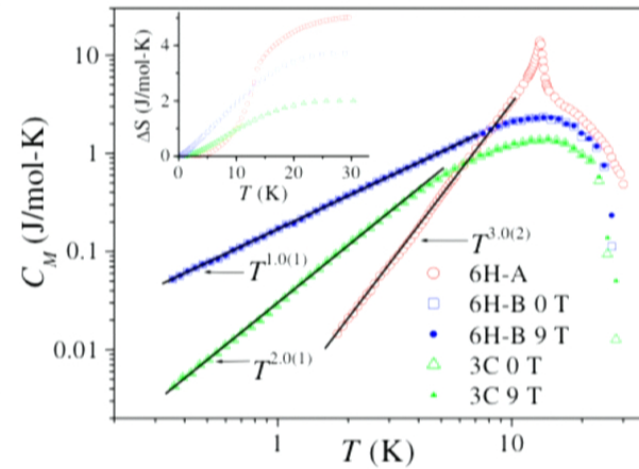
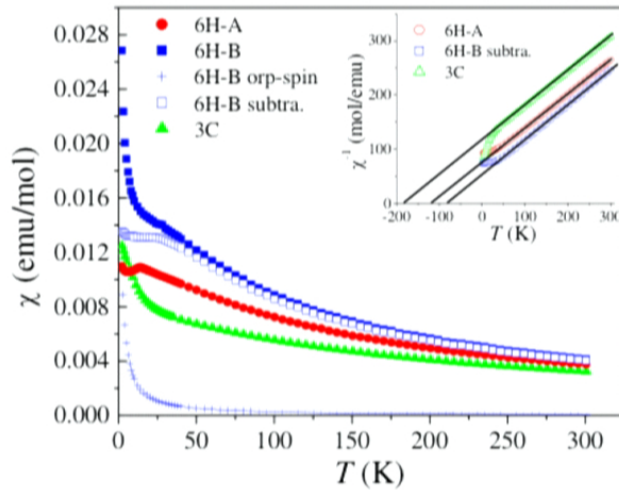


## High-Pressure Sequence of $\text{Ba}_3\text{NiSb}_2\text{O}_9$ Structural Phases: New $S = 1$ Quantum Spin Liquids Based on $\text{Ni}^{2+}$

J. G. Cheng,<sup>1</sup> G. Li,<sup>2</sup> L. Balicas,<sup>2</sup> J. S. Zhou,<sup>1</sup> J. B. Goodenough,<sup>1</sup> Cenke Xu,<sup>3</sup> and H. D. Zhou<sup>2,\*</sup>

$\theta_{\text{CW}}$  of  $-75.5$  (6H-B) K

$\gamma = 168$  mJ/mol K<sup>2</sup>



## S=1/2 Candidate Materials for Spin Liquid

Material	Lattice	Ground State	$f= \Theta _{CW}/T_F$
$\kappa$ -(BEDTTTF) <sub>2</sub> Cu <sub>2</sub> (CN) <sub>3</sub>	triangular	QSL?	$> 10^3$
ETMe <sub>3</sub> Sb[Pd(dmit) <sub>2</sub> ] <sub>2</sub>	triangular	QSL?	$> 10^3$
ZnCu <sub>3</sub> (OH) <sub>6</sub> Cl <sub>2</sub> (Herbertsmithite)	kagome	QSL?	$> 10^3$
Cu <sub>3</sub> V <sub>2</sub> O <sub>7</sub> (OH) <sub>2</sub> • 2H <sub>2</sub> O (Volborthite)	anisotropic- kagome	AF? Glassy?	$\approx 100$
BaCu <sub>3</sub> V <sub>2</sub> O <sub>8</sub> (OH) <sub>2</sub> (vesigniete)	anisotropic- kagome	QSL?	$> 100$
Na <sub>4</sub> Ir <sub>3</sub> O <sub>8</sub>	hyperkagome	QSL?	$> 10^3$



# Search for Quantum Spin Liquid (“S=1/2”)

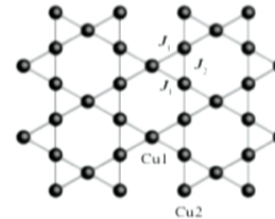
**Herbertsmithite** “Ideal” Kagome lattice

$\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$  D. G. Nocera, Y. S. Lee



**Volborthite** Distorted Kagome lattice

$\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$  Z. Hiori



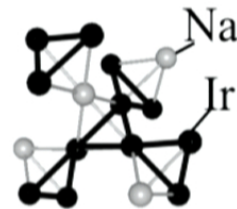
$\kappa$ -(BEDT-TTF) $_2\text{Cu}_2(\text{CN})_3$  K. Kanoda

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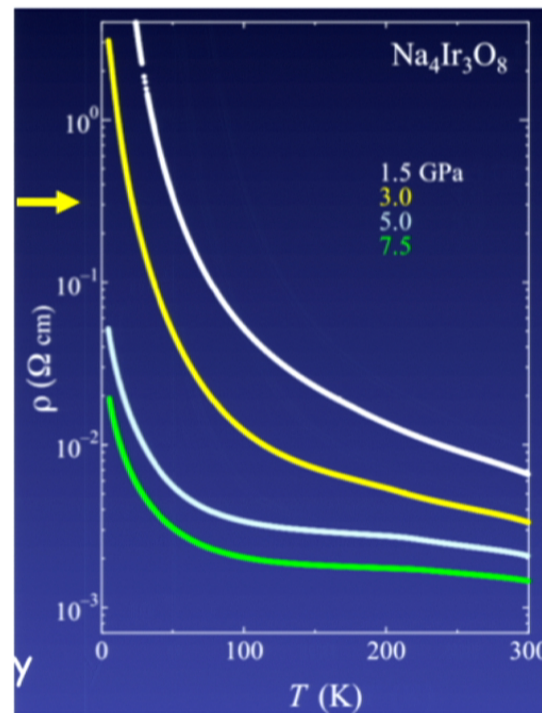
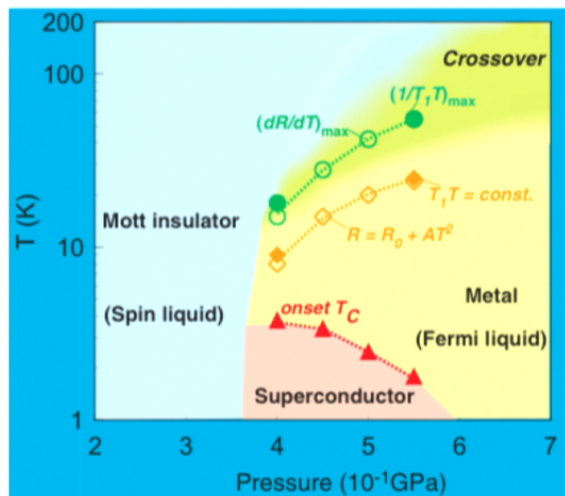
Organic Material  
Triangular Lattice

**Hyperkagome**  $\text{Na}_4\text{Ir}_3\text{O}_8$

H. Takagi

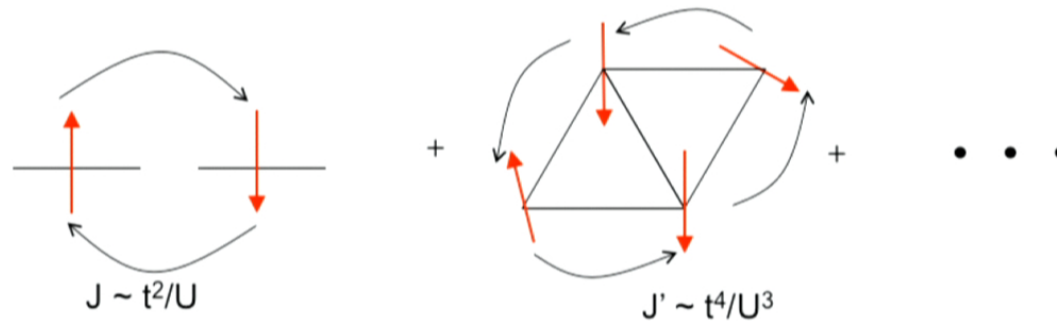


## Weak Mott Insulator ?



# Importance of Charge Fluctuations

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



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)

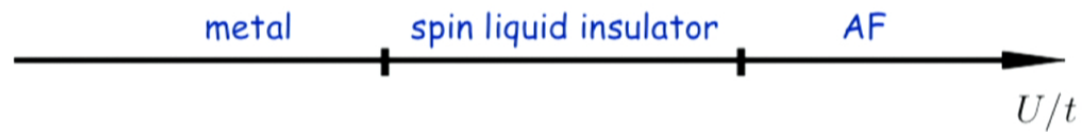


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$\kappa$ -(BEDT-TTF)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub> K. Kanoda

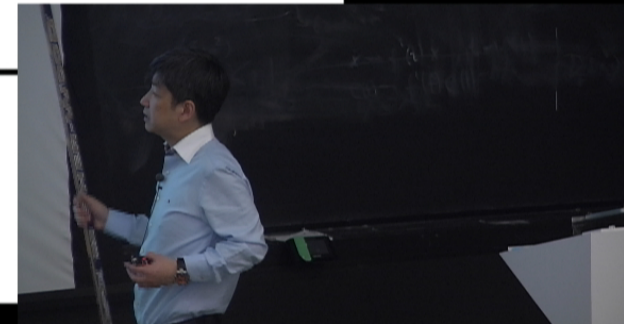
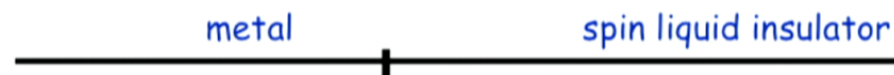
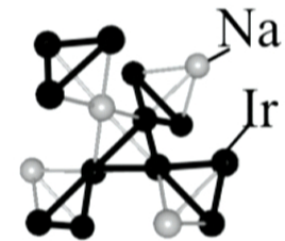
EtMe<sub>3</sub>Sb[Pd(dmit)<sub>2</sub>]<sub>2</sub> R. Kato, Y. Matsuda

Triangular Lattice;  
near Mott transition



Hyper-Kagome Na<sub>4</sub>Ir<sub>3</sub>O<sub>8</sub> H. Takagi

5d transition metal oxides;  
intermediate coupling

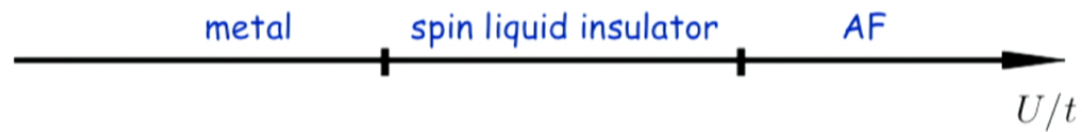


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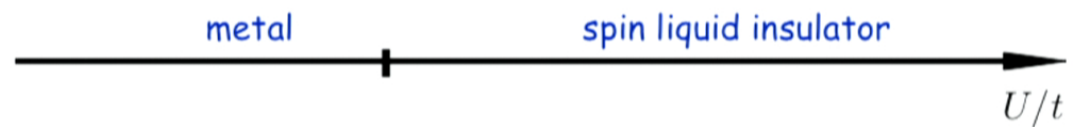
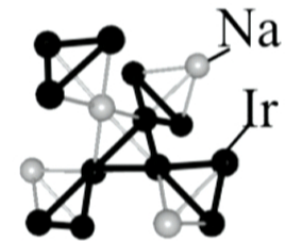
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Hyper-Kagome Na<sub>4</sub>Ir<sub>3</sub>O<sub>8</sub> H. Takagi

5d transition metal oxides;  
intermediate coupling



Spinons inherit the Fermi Surface of Electrons

# Variational Wavefunctions for Spin Liquids

## Slave-Particle Approach

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

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$$H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \quad \longrightarrow \quad H = \sum_{ij} J_{ij} f_{i\alpha}^\dagger f_{i\beta}^\dagger f_{j\beta} f_{j\alpha} + \dots$$

fermion-fermion interaction

# Variational Wavefunctions for Spin Liquids

## Slave-Particle Approach

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

$$H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \quad \longrightarrow \quad H = \sum_{ij} J_{ij} f_{i\alpha}^\dagger f_{i\beta}^\dagger f_{j\beta} f_{j\alpha} + \dots$$

fermion-fermion interaction



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## Mean-Field Theory

$$\chi_{ij} \sim \langle f_{i\alpha}^\dagger f_{i\alpha} \rangle \quad \text{fermion "kinetic" energy dynamically generated}$$

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

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

$$\sum_{\alpha} \langle f_{i\alpha}^\dagger f_{i\alpha} \rangle = 1 \quad \text{Constraint imposed only on average}$$

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Not every choice of  $\chi_{ij}$  and  $\Delta_{ij}$  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)

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

$\chi_{ij}$  and  $\Delta_{ij}$  can be taken as variational parameters

Find  $\chi_{ij}$  and  $\Delta_{ij}$  that give the lowest energy solution

Ranking of ground state energy for  $\Psi_{MF}$  and  $\Psi_{proj}$  changes quite often

## Possible Spin Liquid Phases; Why Gauge Theory ?

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

$$H \sim \sum_{ij} \chi_{ij} f_{i\alpha}^\dagger f_{j\alpha} \sim \sum_{ij} |\chi| e^{ia_{ij}} f_{i\alpha}^\dagger f_{j\alpha}$$

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

U(1) gauge invariance

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

2) **Z<sub>2</sub> Spin Liquid**  $\Delta_{ij} \neq 0$   $\chi_{ij} \neq 0$  (a BCS state of the spinons)

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

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

$$f_{i\alpha} \rightarrow \epsilon_i f_{i\alpha} \quad \epsilon_i = \pm 1$$
$$\sigma_{ij} \rightarrow \epsilon_i \sigma_{ij} \epsilon_j$$

Z<sub>2</sub> gauge invariance



## Possible Spin Liquid Phases; Wavefunctions

### 1) U(1) Spin Liquid

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

### 2) Z<sub>2</sub> Spin Liquid

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

## Properties of Spin Liquid Phases

### 1) U(1) Spin Liquid

	Gapped	Gapless	Specific Heat
2D	Not stable	Stable Fermi Surface Dirac point	$C_{MF}(T) \sim T$ $C_{MF}(T) \sim T^2$
3D	Stable	Stable Fermi Surface Line Node	$C_{MF}(T) \sim T$ $C_{MF}(T) \sim T^2$



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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  $C(T) \sim T^3$

## Properties of Spin Liquid Phases

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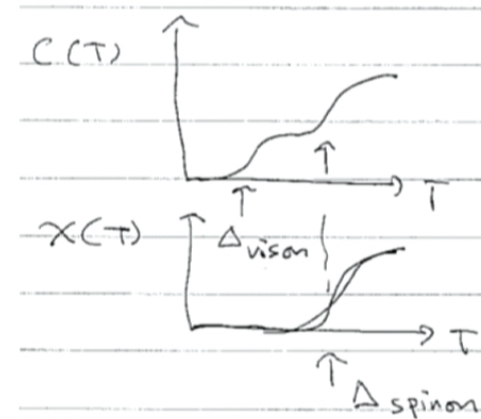
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i) Fully gapped case:  $C(T) \sim e^{-\Delta_{\text{vison}}/T}$

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

charge-neutral spinless;  
only carries entropy



## Properties of Spin Liquid Phases

Notice  $C(T) \sim T^\alpha$  often in candidate spin liquid materials

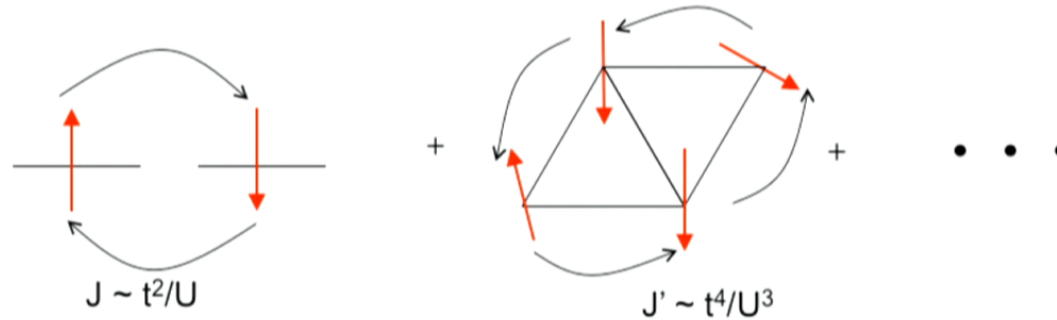
indicates the presence of low energy spin-carrying excitations ?

Spinons ?



# Organics: Triangular Lattice

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

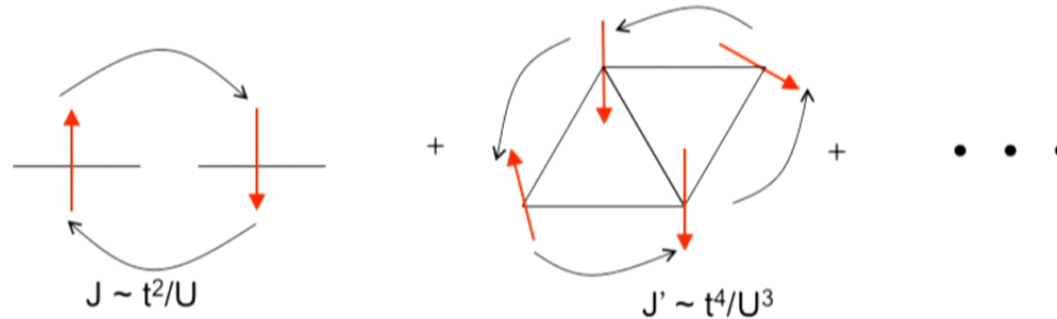


Variational wavefunction - Uniform RVB:  $\chi_{ij} = \chi \quad \Delta_{ij} = 0$

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

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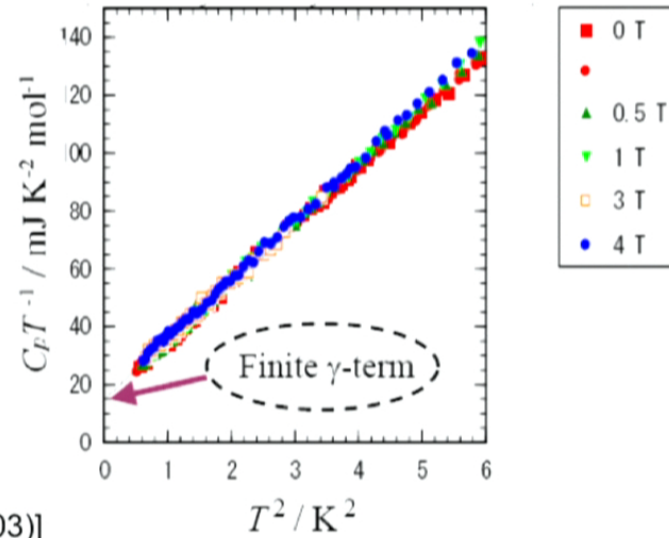
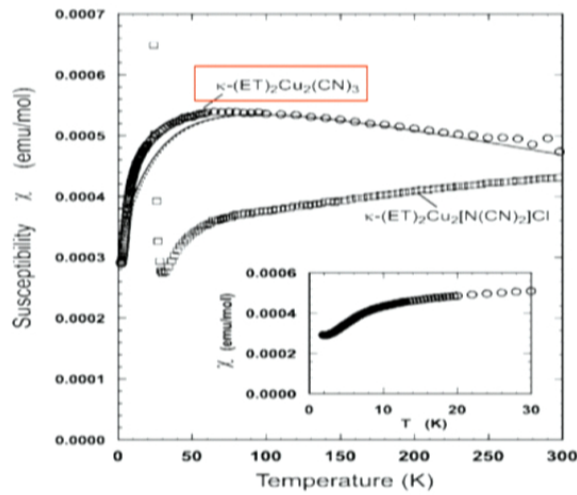
**Fermi surface of spinons**  $C_{\text{MF}}(T) \sim \gamma T$  but ...  $C_{\text{fluc}}(T) \sim T^{2/3}$

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

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

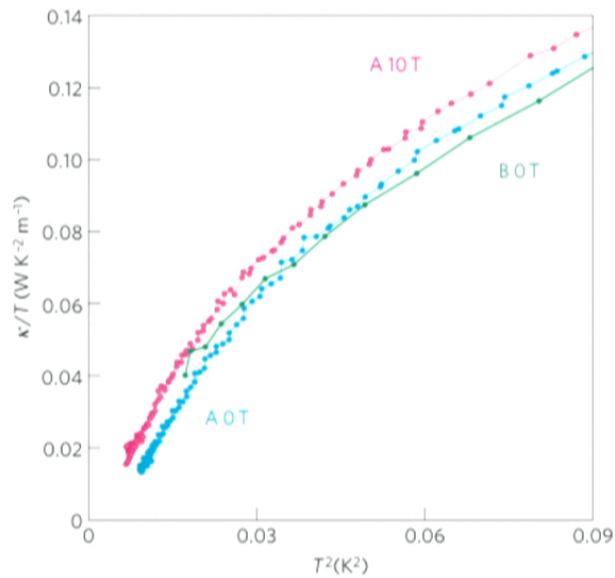


[Y. Shimizu et al., PRL 91, 107001 (03)]

- No magnetic ordering;
- Constant  $T=0$  susceptibility;
- $C(T) \sim T$ ;
- $1/T_1$  Power-law in  $T$ ;
- Wilson Ratio: Order  $O(1)$



## Organics: Triangular Lattice; Further Challenges

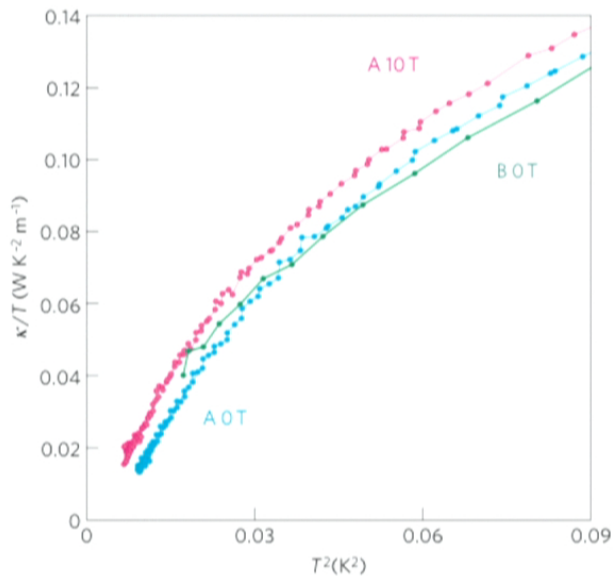


$\kappa/T$  goes to zero as  $T \rightarrow 0$  ?  
(Y. Matsuda, Nature Physics)

inconsistent with  
"Fermi surface" state  
of spinons

$C(T)$  shows a kink around 5K ?  
(K. Kanoda)

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### $Z_2$ Spin Liquid ?

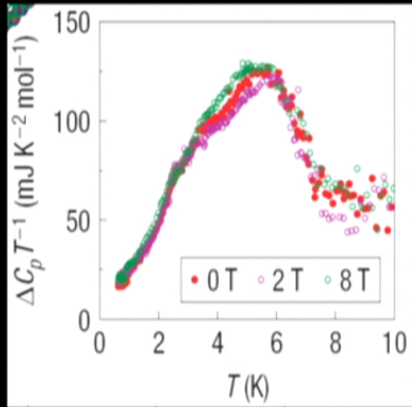
T. Senthil + P. A. Lee -- d-wave nematic spin liquid

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)

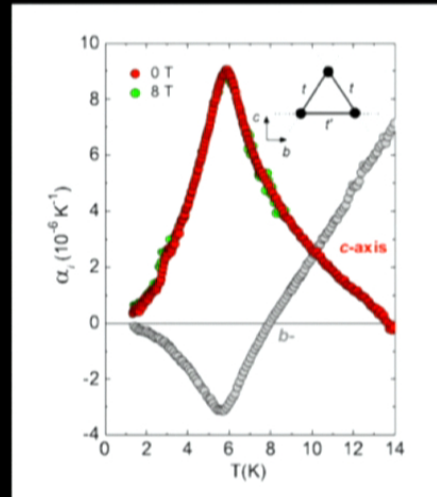
# Thermodynamic anomaly at 5-6K in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

**Specific heat** S. Yamashita *et al.*, *Nature Phys.* 4 (2008) 459



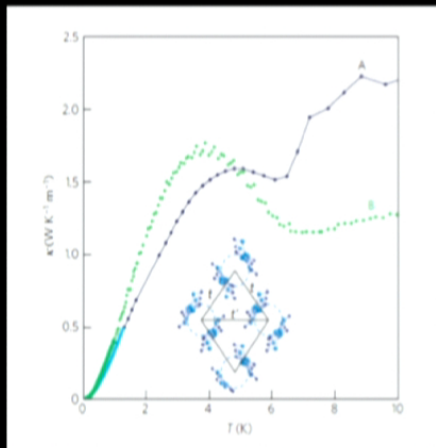
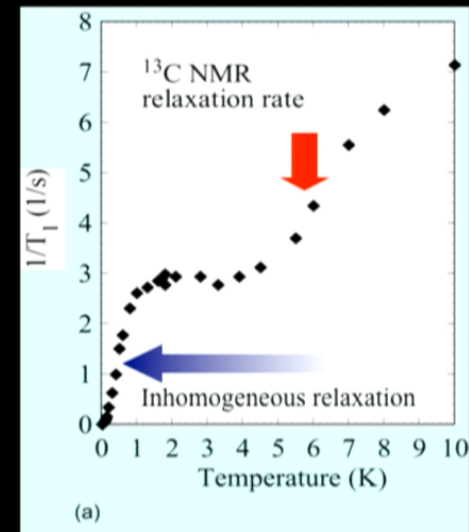
**Thermal expansion coefficient**

Manna *et al.*, *PRL* 104 (2010) 016403



**NMR Relaxation rate**

Shimizu *et al.*, *PRB* 70 (2006) 060510



**Thermal conductivity**

M. Yamashita *et al.*, *Nature Phys.* 5 (2009) 44

# Organics: Triangular Lattice

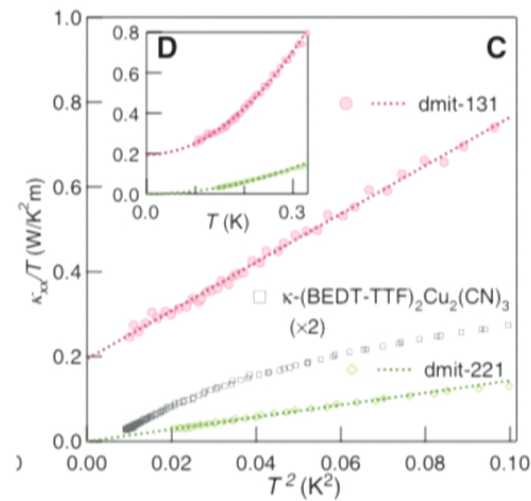


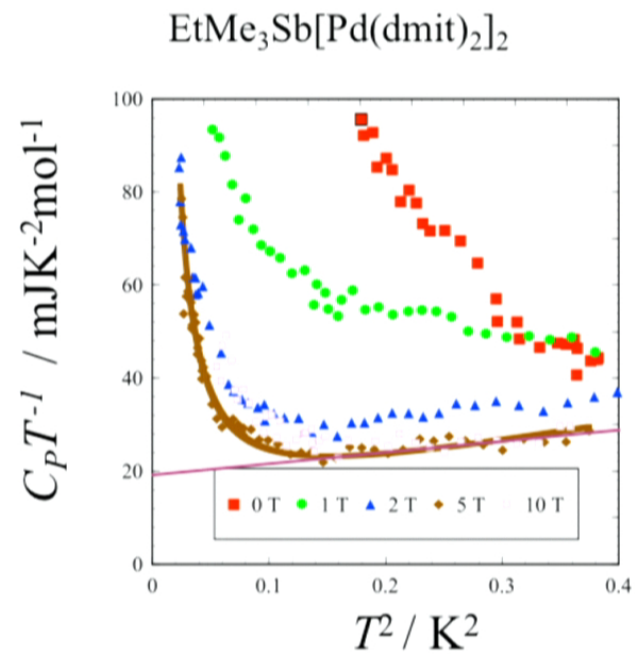
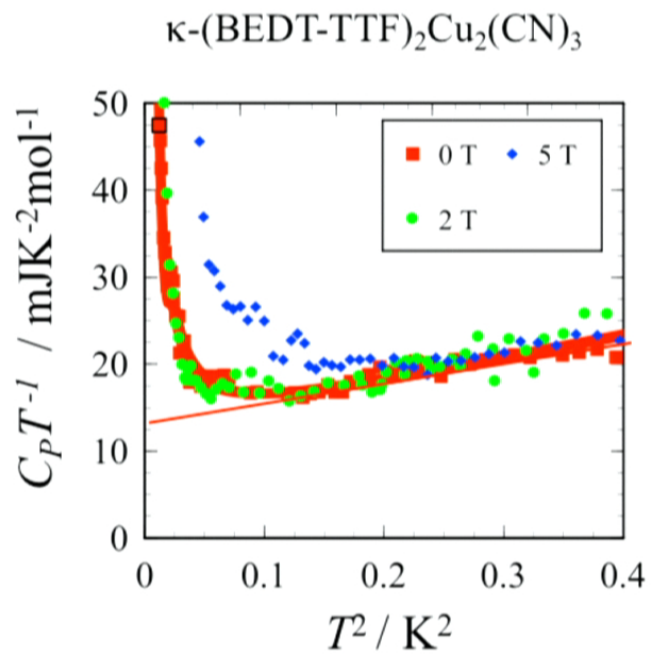
$C(T) \sim T$  (but  $C(T)$  at very low temperatures may be complicated...)

$\kappa/T$  is **finite** as  $T \rightarrow 0$  (Y. Matsuda, Science)

$1/T_1 \sim T^3$  in another experiment; d-wave spinon gap ?

but it was obtained from  $e^{-c(t/T_1)^\alpha}$  with  $\alpha \neq 1$



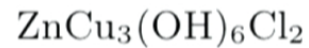


$$\gamma = 12.6 \text{ mJK}^{-2}\text{mol}^{-1} (\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3)$$

$$\gamma = 19.9 \text{ mJK}^{-2}\text{mol}^{-1} (\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2)$$

Existence of  $T$ -linear contribution suggests the realization of gap-less quantum spin liquid state.

# Kagome Lattice; Herbertsmithite



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

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

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

need "disorder" to be consistent with the experiments

**VBS state with 36-site unit cell; small spin gap**

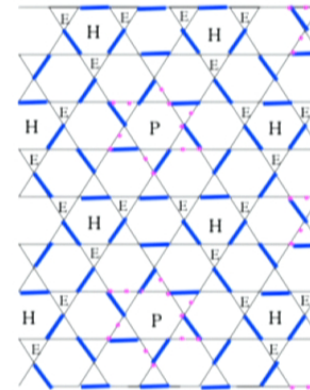
Marston+Zeng, Huse+Singh ...  $C(T) \sim e^{-\Delta/T}$

G. Vidal

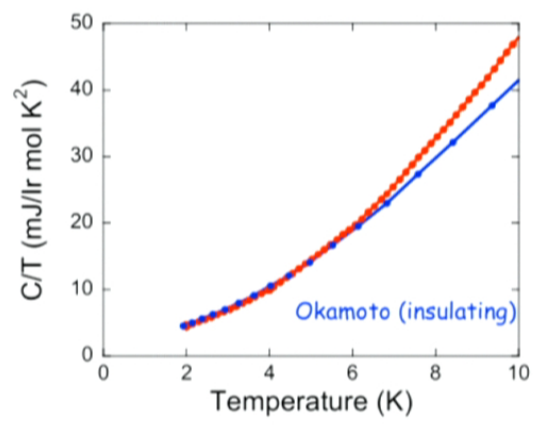
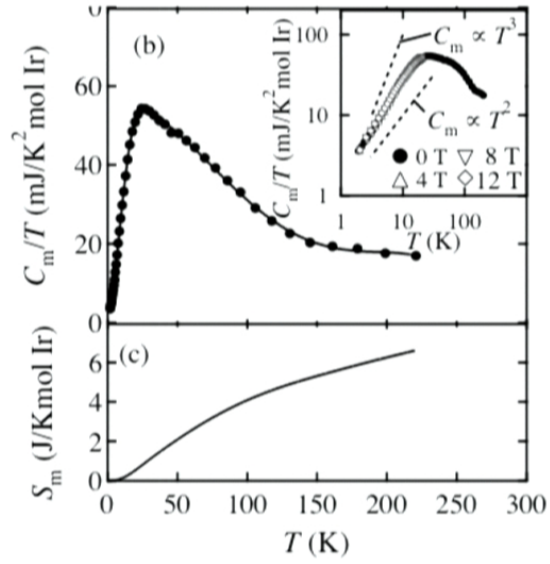
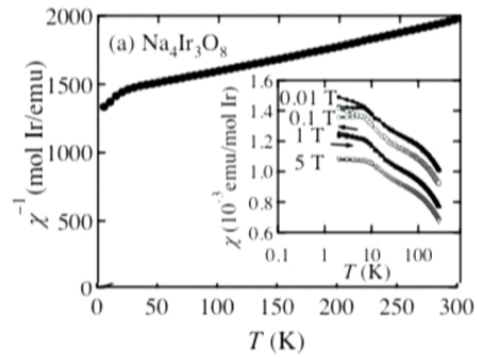
Recent DMRG study by White + Huse;

Spin Liquid with finite spin gap

$\mathbb{Z}_2$  Spin Liquid ?



# Hyper-Kagome Lattice; $\text{Na}_4\text{Ir}_3\text{O}_8$



- $C(T) \sim \gamma T$       Wilson ratio  $\sim 40-50$
- $\chi \sim \text{constant}$
- $1/T_1 \sim T$       Large spin-orbit coupling?

# Hyper-Kagome Lattice; $\text{Na}_4\text{Ir}_3\text{O}_8$

Spin Liquid with small particle-like and hole-like pockets  $C_{\text{MF}}(T) \sim \gamma T$

M. Lawler, A. Paramakanti, Y. B. Kim, L. Balents

Y. Zou, T.-K. Ng, F.-C. Zhang, P. A. Lee

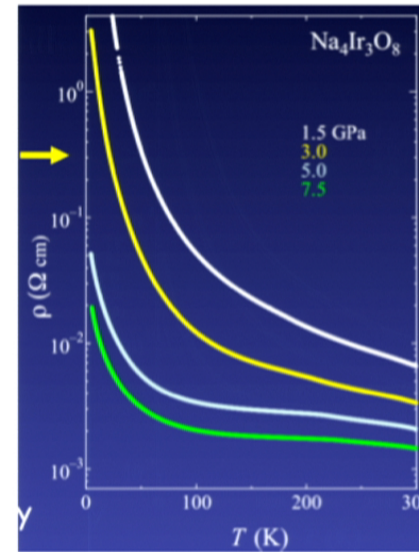
Charge-fluctuations due to the proximity to Metal-Insulator transition

D. Podolsky, A. Paramakanti, Y. B. Kim, T. Senthil

$$C_{\text{fluc}}(T) \sim T \ln \ln(1/T)$$

instead of

$$C_{\text{fluc}}(T) \sim T \ln(1/T)$$





# Topological Mott Insulator



# Type I Topological Phases (Gapped Phases)

Topological Phases



No "Path"  
(Local unitary transformations)  
without closing the bulk gap

"simple phases" (fully characterized by local order  
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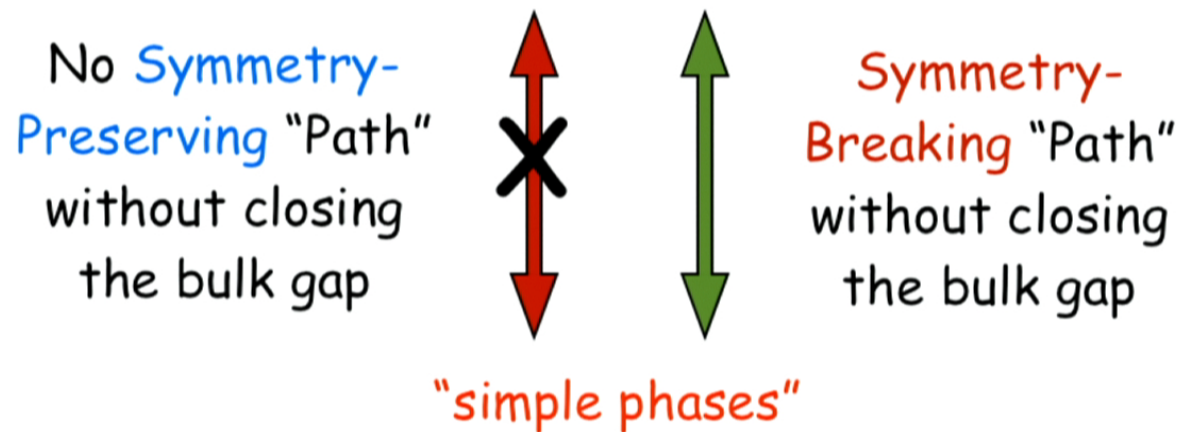
Quantum Hall States

Spin Liquids

(correlated quantum paramagnetic state)

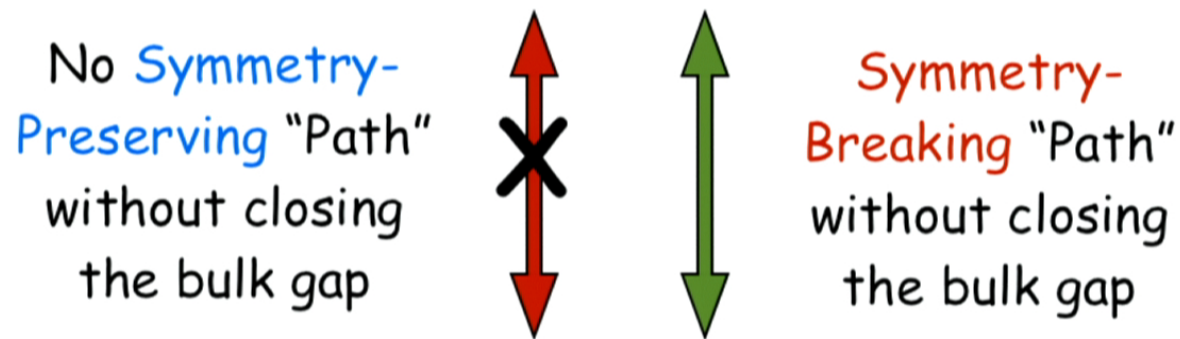
## Type II “Symmetry-Protected” Topological Phases (Gapped Phases)

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Topological Phases



“simple phases”

Topological Band Insulator  
(e.g. time-reversal symmetry)

Can we have topological phases with both Type I and Type II characters ?



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Three-dimensional spin liquid with **spinons**  
having topological band structure



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spinon topological band insulator;  
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Fractionalized topological insulator;  
time-reversal symmetry

Such phases in 2D is unstable

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$J_1$ - $J_2$  model on the pyrochlore lattice  $J_1 > 0$  and  $J_2 < 0$

S. Bhattacharjee, S.-S.Lee, Y.B.Kim, D.H.Lee, arXiv:1202.0291

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spin liquid with  
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Mean-field theory ( $J_1 \gg |J_2|$ );  
spin liquid with  
"strong topological insulator" for spinons

Such phases also have **bond-spin-nematic order**

$$\langle Q_{ij}^{ab} \rangle = -\frac{1}{2} \left[ E_{ij,a} E_{ij,b}^* - \frac{\delta_{ab}}{3} |\mathbf{E}_{ij}|^2 \right] \neq 0 \quad \text{R. Shindou and T. Momoi (2009)}$$

S. Bhattacharjee, S.-S.Lee, Y.B.Kim, D.H.Lee, arXiv:1202.0291

# Contemplation

Our theory of spin liquid is incomplete; Standard Model ?

Shall we call it *S-theory* ??

Landau Fermi Liquid theory v.s. *Spinon Fermi Liquid theory*

density-density interaction  $\longleftrightarrow$  current-current interaction

Instability of electron FS  $\rightarrow$  Broken Symmetry States

Instability of spinon FS  $\rightarrow$  A variety of Mott insulators ?

But *the spinons do not have to be well-defined*;  
gauge-invariant response functions still know about them

*Is gauge theory necessary ?*

long-range current-current interaction of spinons

non-locality; consequence of requiring fermions/bosons

# Contemplation

We need **more spectroscopic tools**: Theory and Experiments

Neutron ? Magnetic X-ray ? **Direct probe of spinon FS ?**

We need better single crystals, thermal transport, and  
specific heat data

Control and understanding of disorder effect