Spin-metal phase in Mott insulator with two-dimensional triangular antiferromagnets

Minoru Yamashita, Kyoto Univ.

1. Quantum spin liquid in 2D
   - ☐ Recipe.
   - ☐ Gapless? Gapped?
2. Thermal-transport study of $EtMe_3Sb[Pd(dmit)_2]_2$
   - ☐ Finite κ/T in the zero-temperature limit

     M. Y. et al., *Science*, **328**, 1246 (2010)

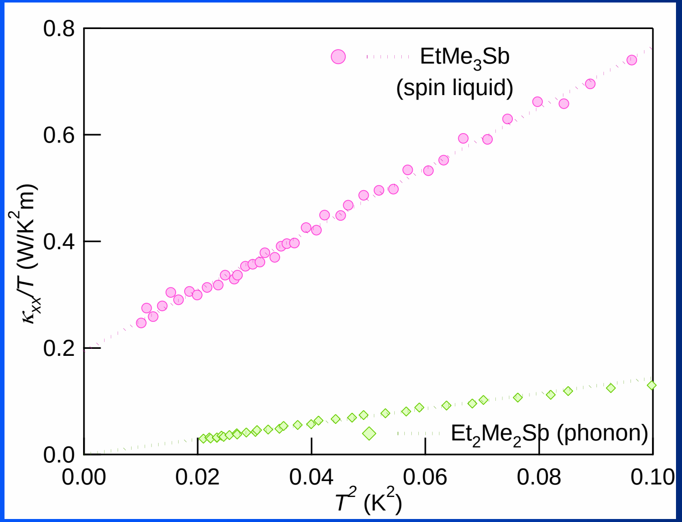
   → Gapless excitation with long mean free path
3. Magnetic torque
   - ☐ $\Delta\chi$ remains finite down to 30 mK, 0.5 T

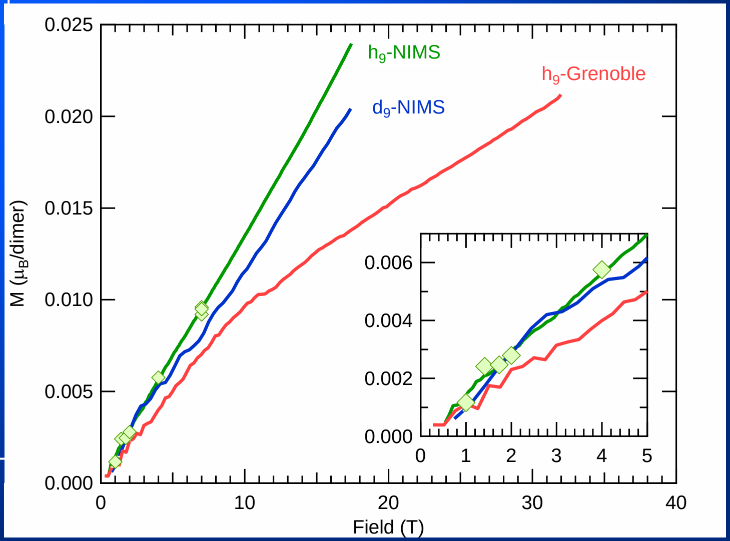
   →Gapless **magnetic** excitation

4. Discussion

   Spin-metal phase?

   Quantum critical phase?





Collaborators

Kyoto University

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Takasada Shibauchi

Yuji Matsuda

Riken

Yugo Oshima

Hiroshi M. Yamamoto

Reizo Kato

$EtMe_3Sb[Pd(dmit)_2]_2$

NIMS

Taichi Terashima

Shinya Uji

CNRS, Grenoble

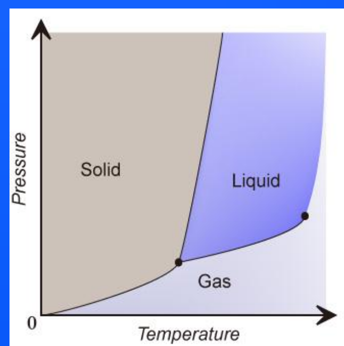
Ilya Sheikin

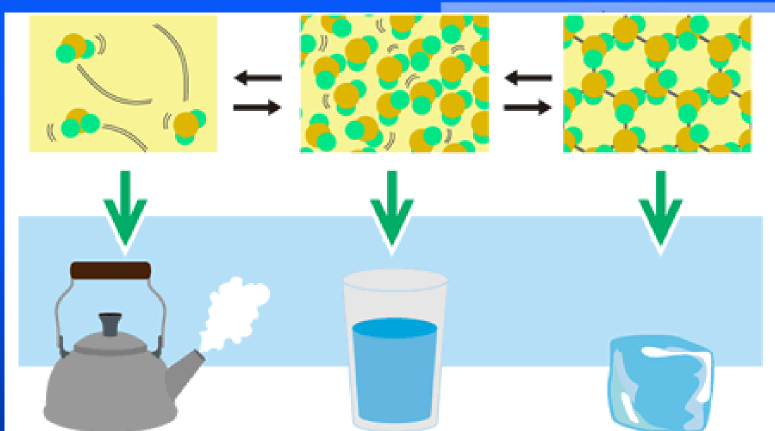
Quantum Liquid

Conventional material

- ✓ Frozen at the absolute zero temperature
- ✓ Ordering in conventional way

e.g. crystallization

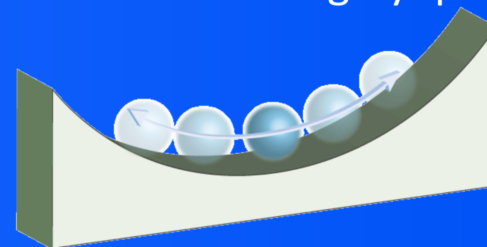




Steam Water Ice

Quantum Liquids

- No freezing by quantum fluctuation

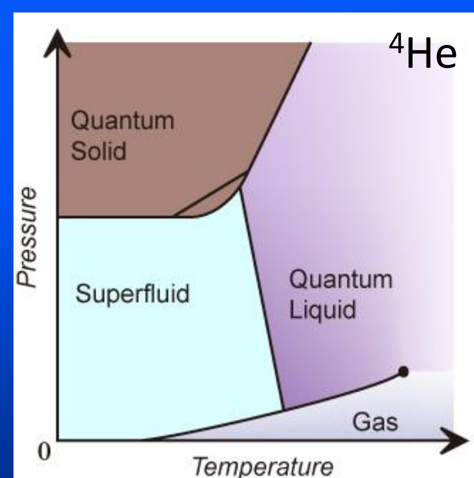


Zero point oscillation
> Interaction

Difference of quantum statics explicitly emerges

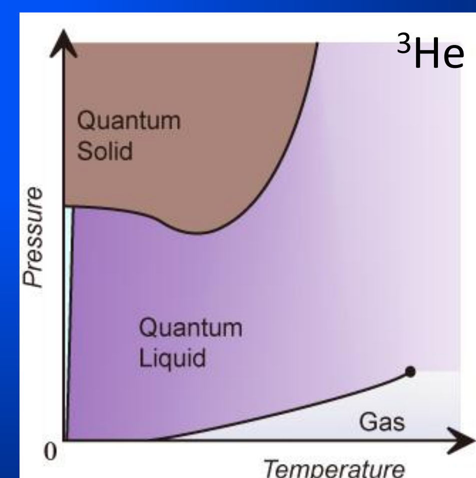
Bose statics

BEC, superfluid ^{4}He



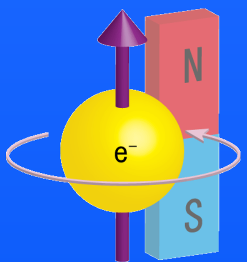
Fermi statics

Fermi liquid, Cooper pairing



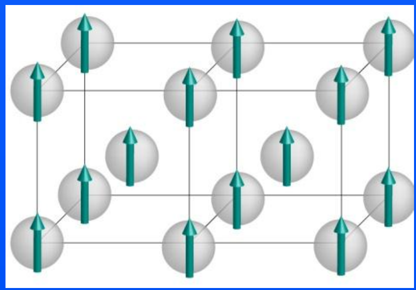
Quantum **Spin** Liquid
A new quantum state of matter

Spins (classical/quantum) in 3D



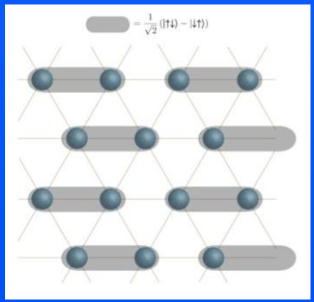
Long-range order for $T < J$
(J: interaction energy)

Ferromagnetic order



Broken rotational symmetry

Valence bond solid (spin singlet)



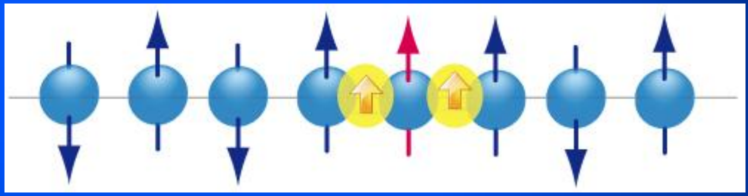
Broken translational symmetry

(exception: spin ice, 3D hyper kagomé)

1D spin chain

Strong quantum fluctuation
No long-range order

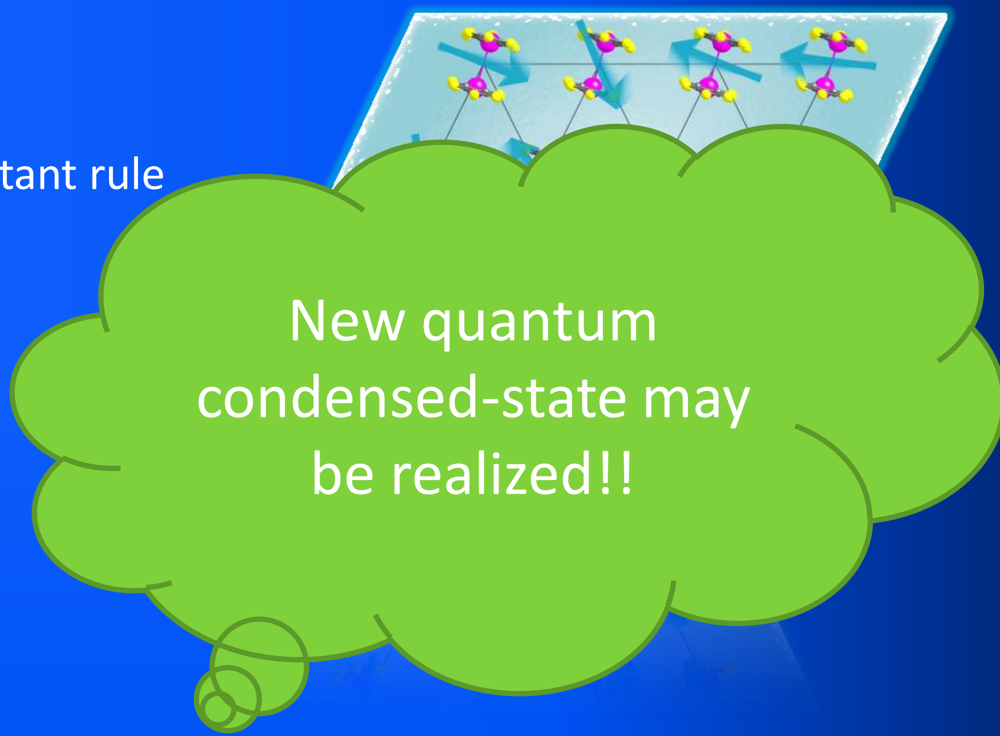
- Gapless spin liquid (S = 1/2)
  fractionalized excitation
  spinon (domain wall)
- Gapped spin liquid (S = 1)
  Haldane gap



Quantum Spin Liquid in Low-dimensions

How about in 2D？

Quantum **Spin** Liquid in 2D



Order or not order?

- geometrical frustration plays important rule

Many theories proposed

- Resonating-valence-bond liquid
- Chiral spin liquid
- Quantum dimer liquid
- Z_2 spin liquid
- Algebraic spin liquid
- Spin Bose Metal
- Etc.,,

Elementary exciation not yet identified

- Spinon with Fermi surface
- Vison
- Majorana fermions

- Recent discoveries of QSL candidates
- No LRO found down to experimentally reachable low temperature
  → New frontier of condensed-matter

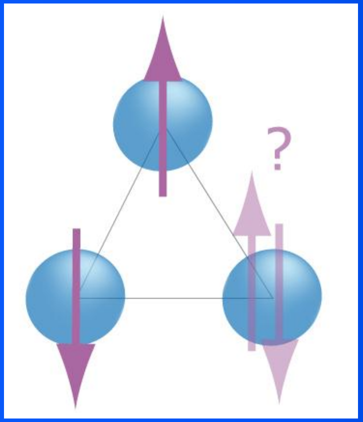
Geometrical Frustration

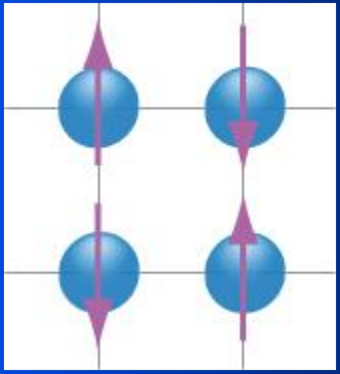
How can we stabilize QSL in 2D？

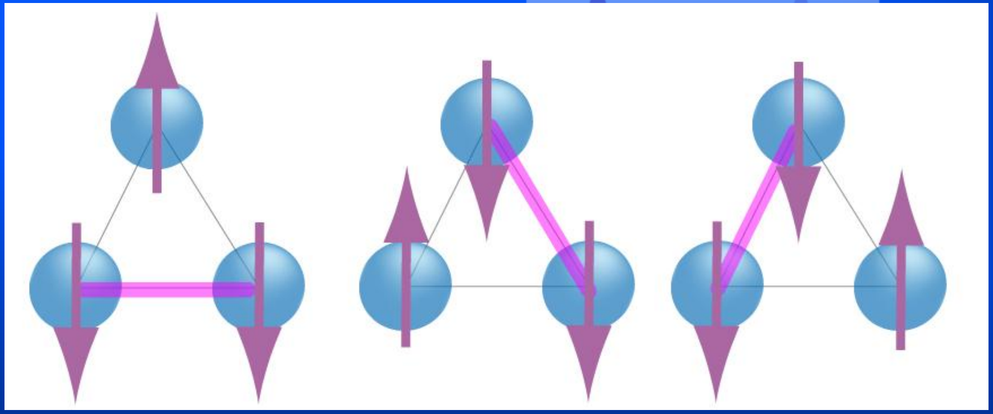
P.W. Anderson (1973)

Geometrical frustration
in AF Ising

<u>“Triangular relation of spins”</u>
Unsatisfied bond







Geometrical Frustration

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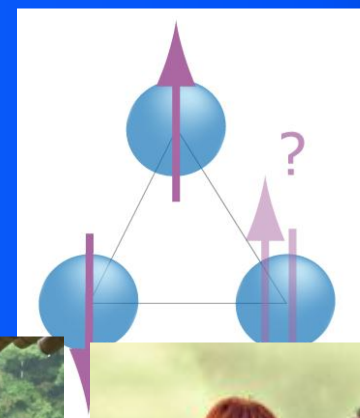
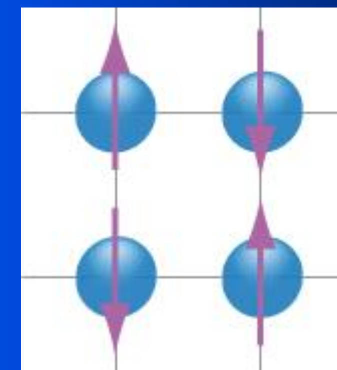




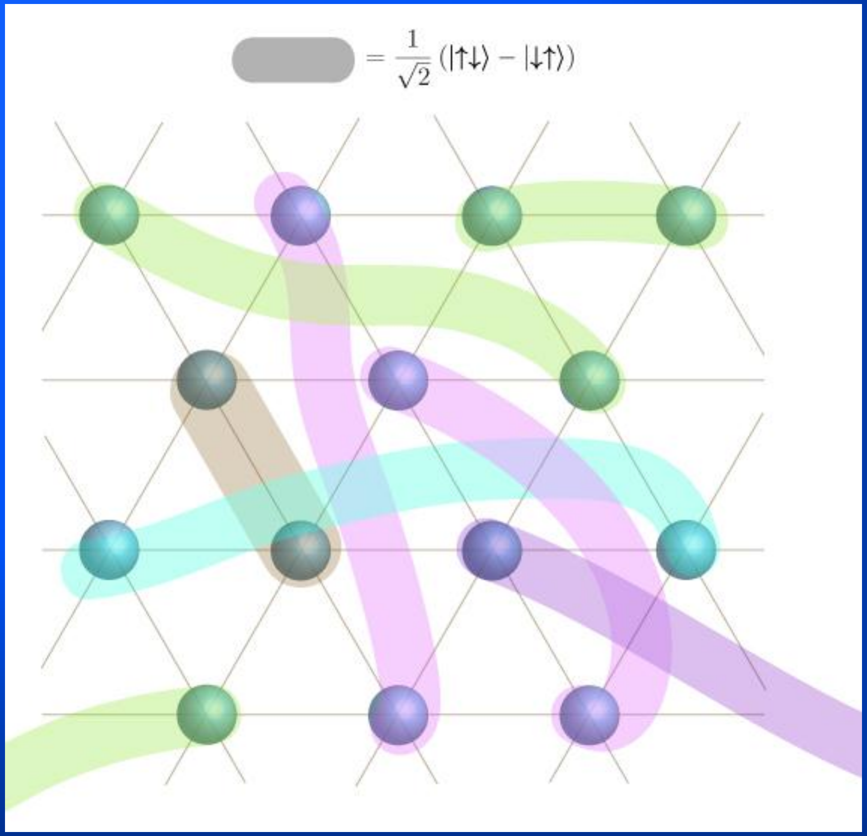
Triangular relation of love

Geometrical frustration: Heisenberg case

P.W. Anderson (1973)

Heisenberg Spins on 2D triangular lattice

$$\mathcal{H} = J \sum_{(i,j)} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$



$+ \cdots$

Quantum spin liquid state

Resonating-valence-bond state

- No long-range order
- No symmetry breaking
- Fractionalized excitation (spinon)
- Long correlation length

Geometrical frustration: Heisenberg case

P.W. Anderson (1973)

Heisenberg Spins on 2D triangular lattice

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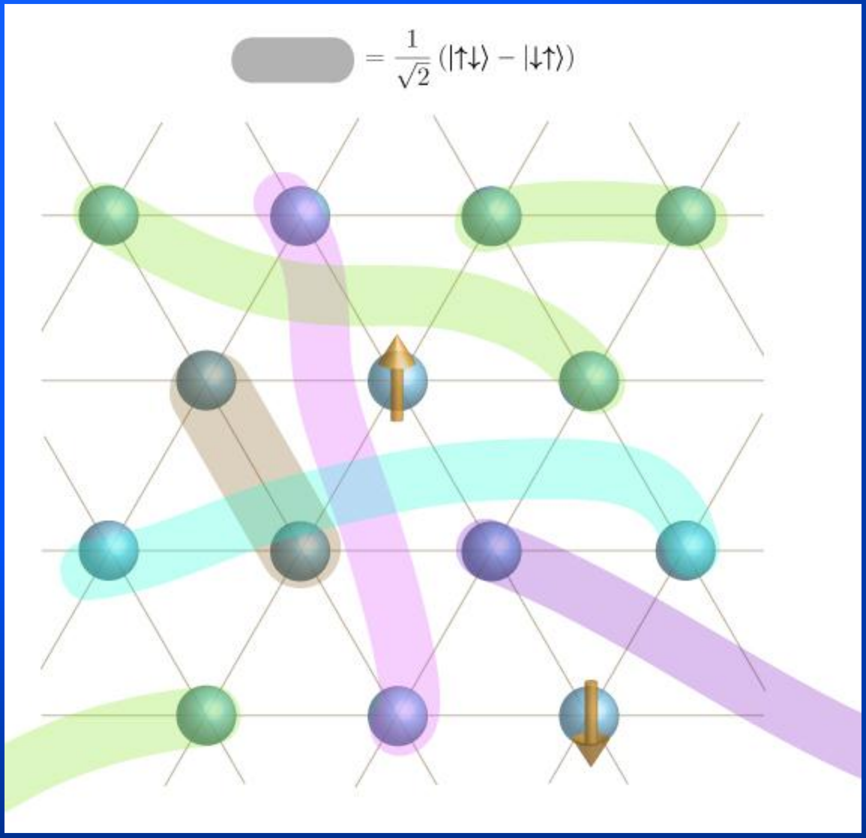

$+ \cdots$

Quantum spin liquid state

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- No long-range order
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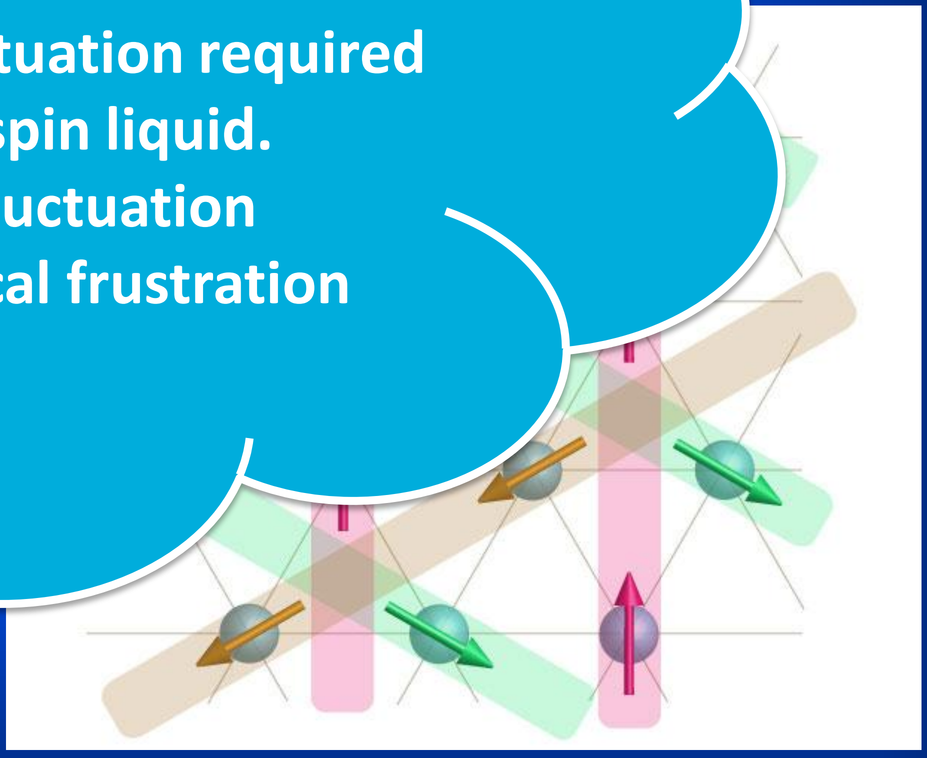
Geometrical frustration: Heisenberg case

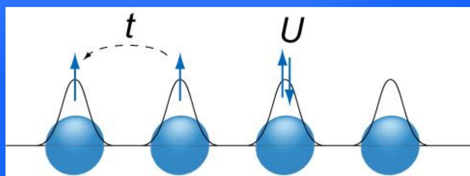
Heisenberg Spins on 2D triangular lattice

Further quantum fluctuation required to stabilize quantum spin liquid.

1. Further quantum fluctuation
2. Stronger geometrical frustration



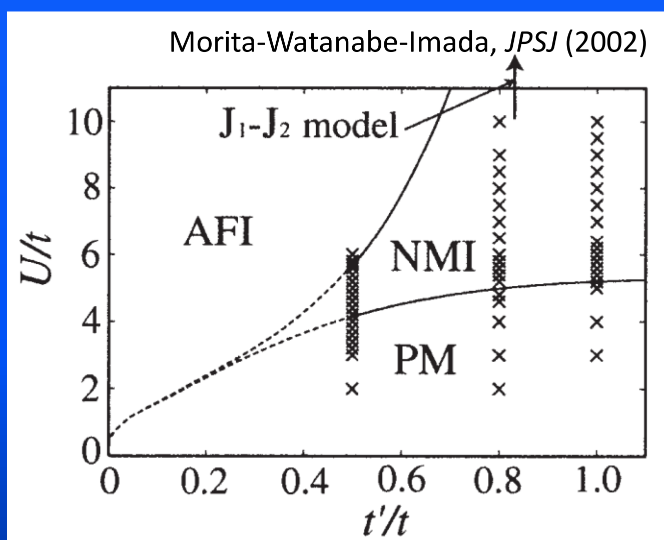
QSL under geometrical frustration



Hubbard model

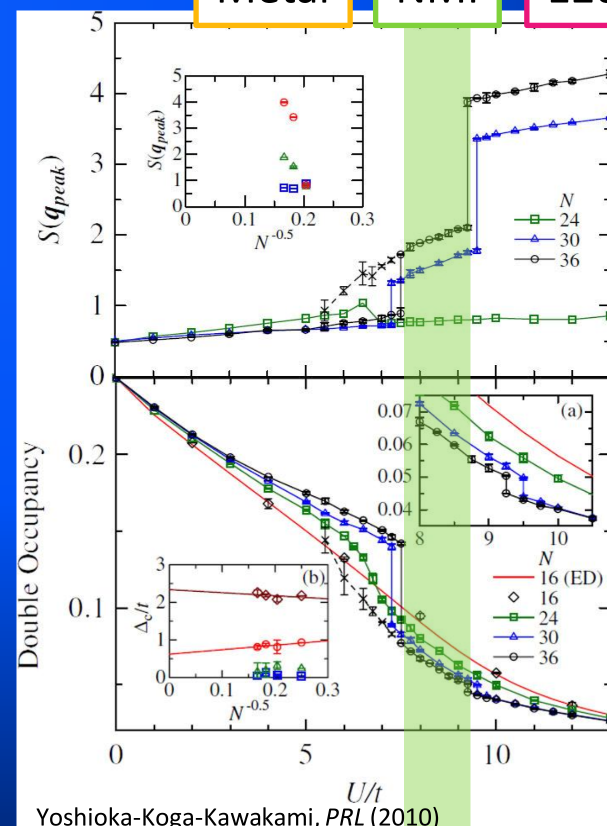
$$\mathcal{H} = -\sum_{(i,j),\sigma} t\left(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.C.}\right) + U\sum_{i=1}^{N} n_{i\uparrow}n_{i\downarrow}$$

Non-Magnetic Insulating phase
(Quantum spin liquid)

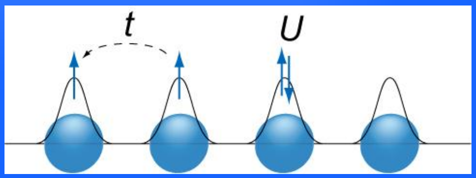


t'/t = 1

Metal | NMI | 120º Néel

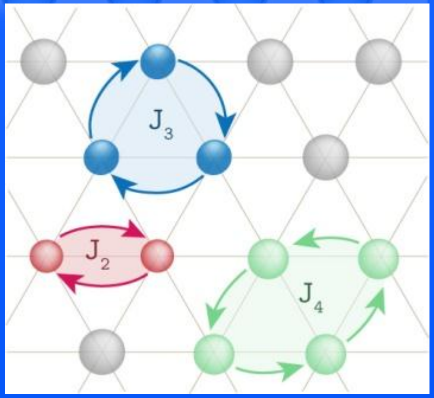


QSL under geometrical frustration



Hubbard model

$$\mathcal{H} = -\sum_{(i,j),\sigma} t\left(c_{i\sigma}^{\dagger}c_{j\sigma} + \mathrm{H.C.}\right) + U\sum_{i=1}^{N} n_{i\uparrow}n_{i\downarrow}$$

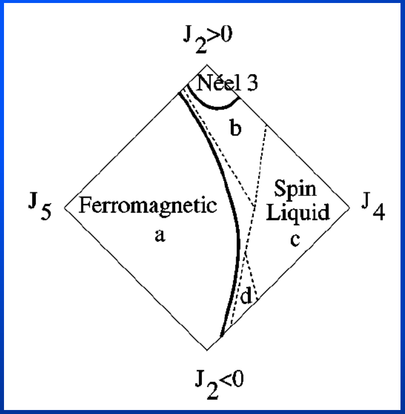


Ring-exchange model

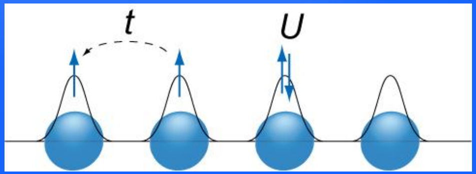
$$\begin{aligned}\mathcal{H} &= J_2^{eff}\sum P_2 + J_4\sum\left(P_4 + P_4^{-1}\right)\\ &\quad - J_5\sum\left(P_5 + P_5^{-1}\right) + J_6\sum\left(P_6 + P_6^{-1}\right)\end{aligned}$$

Quantum spin liquid for large J_4

Misguich-Bernu-Lhuillier-Waldtmann (PRL `98, PRB `99)
LiMing-Misguich-Sindzingre-Lhuillier (PRB `00)
O.I. Motrunich, PRB (2005)

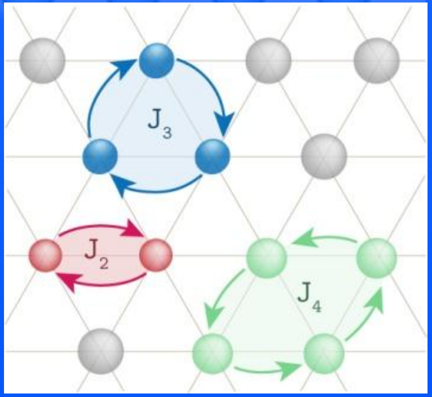


QSL under geometrical frustration



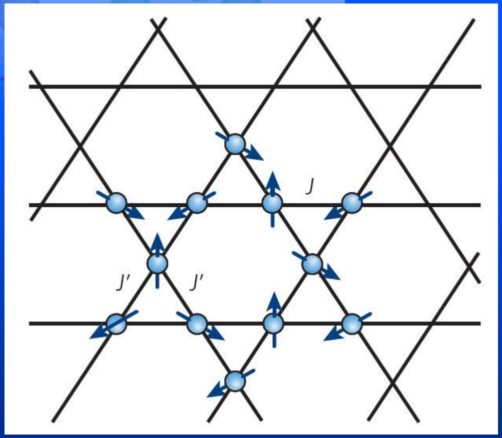
Hubbard model

$$\mathcal{H} = -\sum_{(i,j),\sigma} t\left(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.C.}\right) + U\sum_{i=1}^{N} n_{i\uparrow}n_{i\downarrow}$$



Ring-exchange model

$$\begin{aligned}\mathcal{H} \;=\; & J_2^{eff}\sum P_2 + J_4\sum\left(P_4 + P_4^{-1}\right) \\ & -J_5\sum\left(P_5 + P_5^{-1}\right) + J_6\sum\left(P_6 + P_6^{-1}\right)\end{aligned}$$



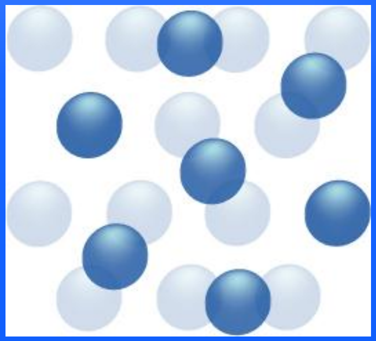
2D kagomé lattice

- Corner-sharing triangular lattice
- Stronger geometrical frustration
- Quantum spin liquid

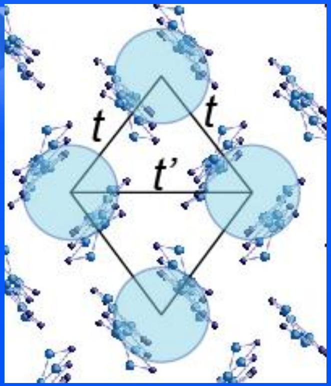
  Yang-Huse-White, Science (2011)

  Nakano-Sakai, JPSJ (2011)

Some of promising candidates of 2D quantum spin liquids



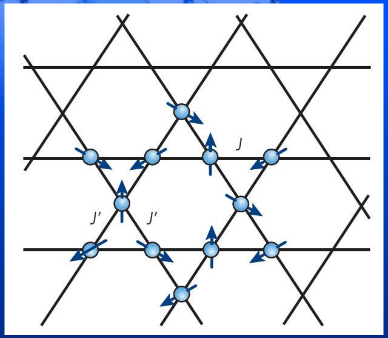
^{3}He absorbed on Graphite



Organic compounds

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$

$EtMe_3Sb[Pd(dmit)_2]_2$

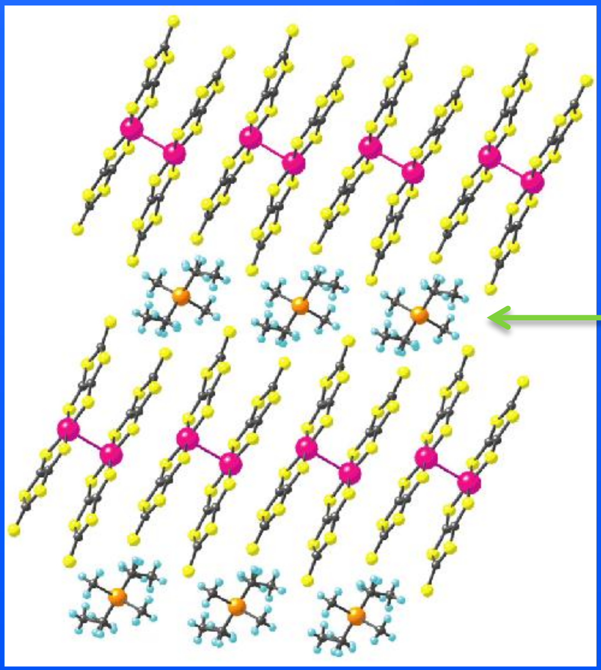


Kagomé

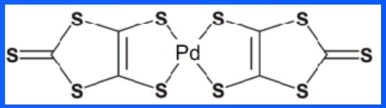
Herbertsmithite, Volborthite, $Rb_2Cu_3SnF_{12}$, $Na_3Ir_3O_8$

(Cation)[Pd(dmit)$_2$]$_2$

SIDE VIEW



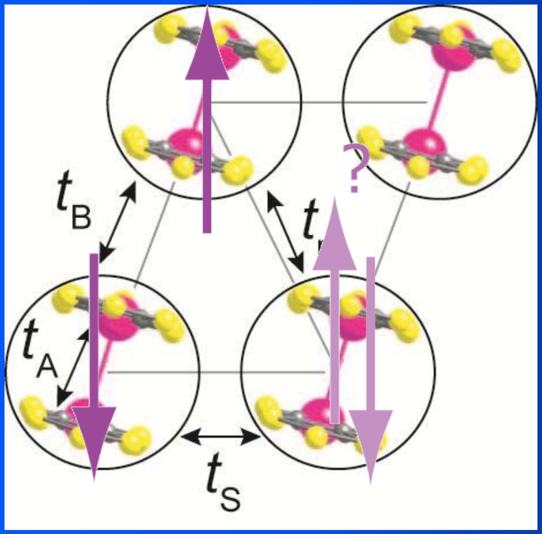
2D layer of Pd(dmit)$_2$ molecule



Cation layer
Non-magentic
X = EtMe$_3$Sb,
Et$_2$Me$_2$Sb,
etc.

2D spin system

TOP VIEW



$t_A \sim 0.5\ \mathrm{eV} \gg t_B, t_S, t_r \sim 30\ \mathrm{mV}$

Dimerization → Half-filled Mott insulator

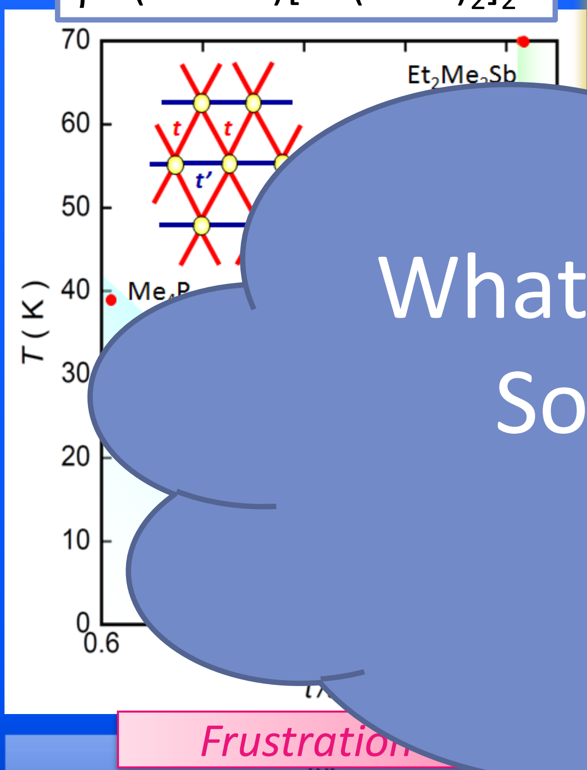
Insulator with *S* = 1/2
Triangular lattice

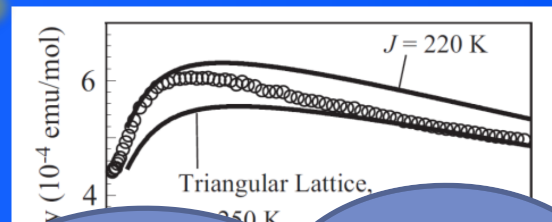
✓Very clean single crystal available
✓"Weak" Mott insulator (small *U*/*t*)
✓Variety of material syntheses available

New spin liquid Material: $EtMe_3Sb[Pd(dmit)_2]_2$

K. Kanoda and R. Kato
Annu. Rev. Condens. Matter Phys. (2011)

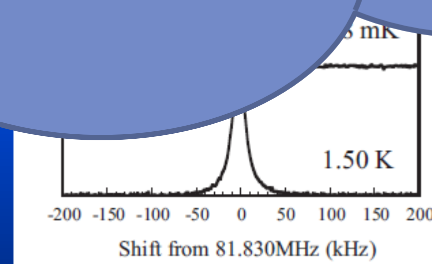
β'-(Cation)$[Pd(dmit)_2]_2$





① χ(T) : 2D triangular

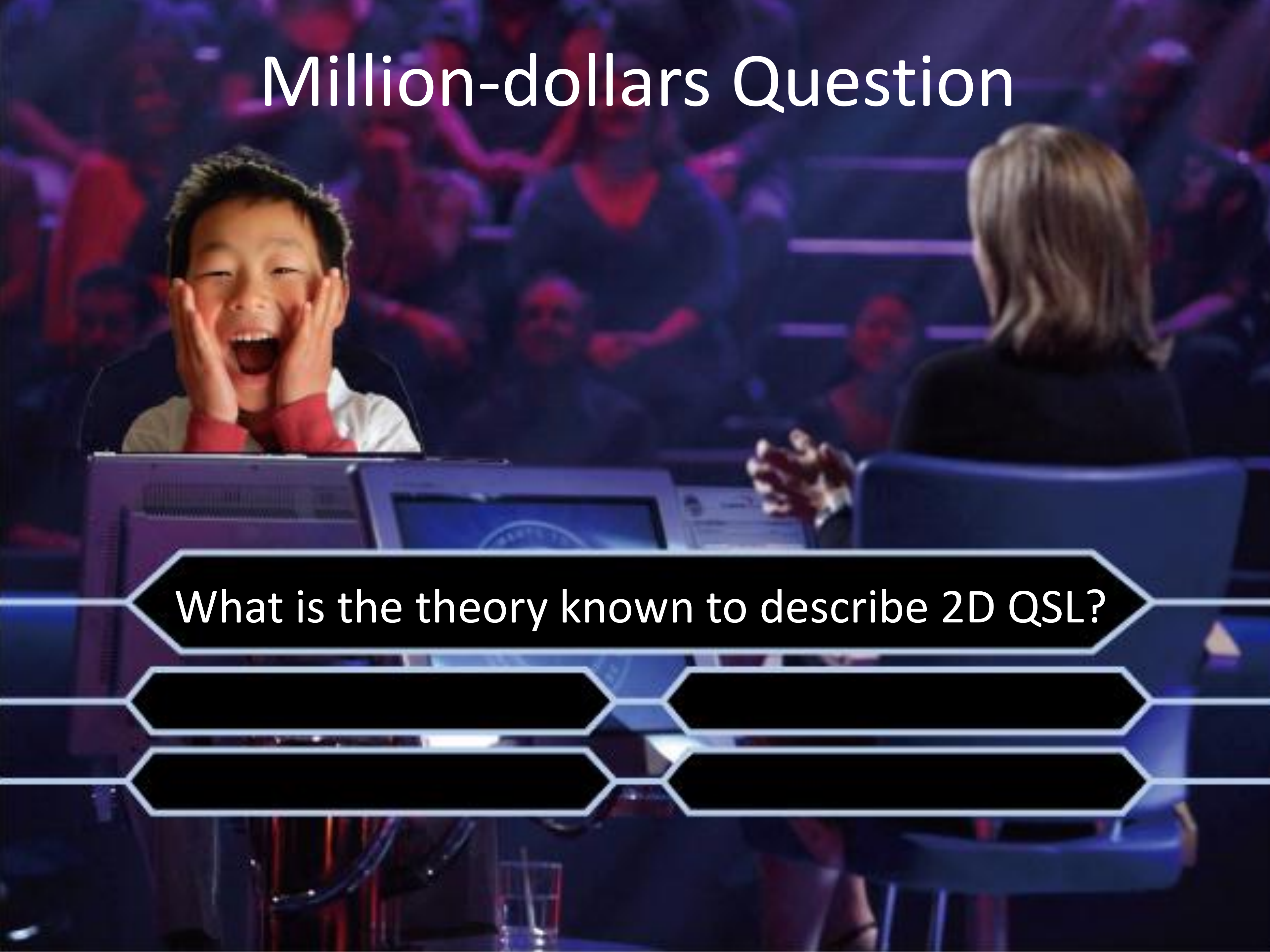
What kind of spin liquid??
So many theoretical candidates,,,



Itou et al., J. Phys. Conf. Ser. **145**, 012039 (2009)

Million-dollars Question

Million-dollars Question



What is the theory known to describe 2D QSL?

Million-dollars Question

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR?*

P. W. Anderson
Bell Laboratories, Murray Hill, New Jersey 07974
and
Cavendish Laboratory, Cambridge, England

(Received December 5, 1972; Invited**)

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond

Million-dollars Question

VOLUME 59, NUMBER 18 PHYSICAL REVIEW LETTERS 2 NOVEMBER 1987

Equivalence of the Resonating-Valence-Bond and Fractional Quantum Hall States

V. Kalmeyer
Department of Physics, Stanford University, Stanford, California 94305

and

R. B. Laughlin
Department of Physics, Stanford University, Stanford, California 94305, and
University of California, Lawrence Livermore National Laboratory, Livermore, California 94550

(Received 24 July 1987)

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond
- **B:** chiral spin liquid

Million-dollars Question

VOLUME 86, NUMBER 9 PHYSICAL REVIEW LETTERS 26 FEBRUARY 2001

Resonating Valence Bond Phase in the Triangular Lattice Quantum Dimer Model

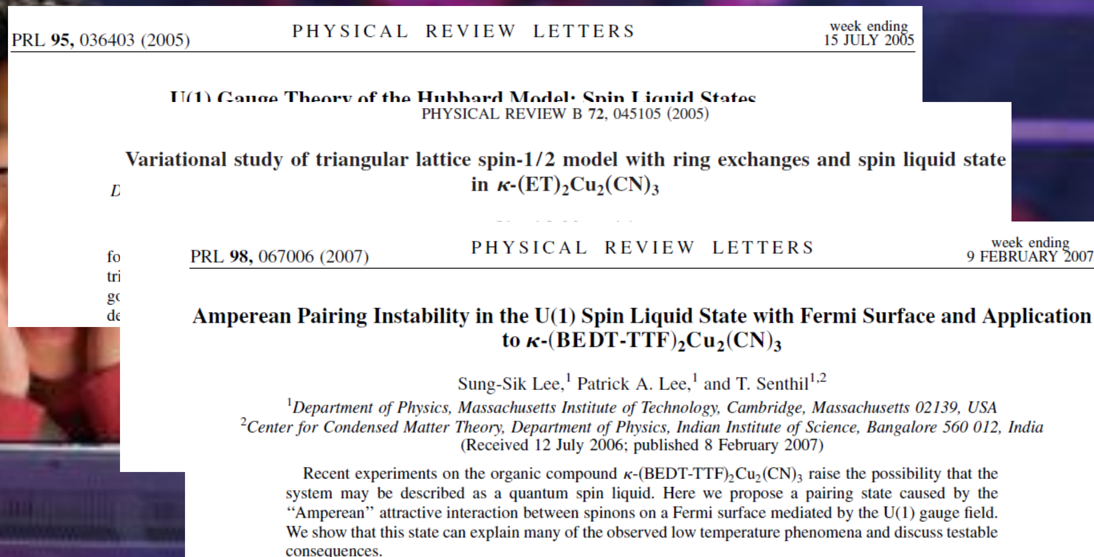
R. Moessner and S. L. Sondhi
Department of Physics, Princeton University, Princeton, New Jersey 08544
(Received 3 August 2000)

We study the quantum dimer model on the triangular lattice, which is expected to describe the singlet dynamics of frustrated Heisenberg models in phases where valence bond configurations dominate their physics. We find, in contrast to the square lattice, that there is a truly short ranged resonating valence bond phase with no gapless excitations and with deconfined, gapped, spinons for a *finite* range of parameters. We also establish the presence of crystalline dimer phases.

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond
- **B:** chiral spin liquid
- **C:** Quantum dimer liquid

Million-dollars Question



PRL **95,** 036403 (2005) PHYSICAL REVIEW LETTERS week ending 15 JULY 2005

U(1) Gauge Theory of the Hubbard Model: Spin Liquid States

PHYSICAL REVIEW B 72, 045105 (2005)

Variational study of triangular lattice spin-1/2 model with ring exchanges and spin liquid state in κ-$(ET)_2Cu_2(CN)_3$

PRL **98,** 067006 (2007) PHYSICAL REVIEW LETTERS week ending 9 FEBRUARY 2007

Amperean Pairing Instability in the U(1) Spin Liquid State with Fermi Surface and Application to κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$

Sung-Sik Lee,[1] Patrick A. Lee,[1] and T. Senthil[1,2]

[1]*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
[2]*Center for Condensed Matter Theory, Department of Physics, Indian Institute of Science, Bangalore 560 012, India*

(Received 12 July 2006; published 8 February 2007)

Recent experiments on the organic compound κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ raise the possibility that the system may be described as a quantum spin liquid. Here we propose a pairing state caused by the "Amperean" attractive interaction between spinons on a Fermi surface mediated by the U(1) gauge field. We show that this state can explain many of the observed low temperature phenomena and discuss testable consequences.

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond
- **B:** chiral spin liquid
- **C:** Quantum dimer liquid
- **D:** QSL with spinon Fermi surface

Million-dollars Question

PHYSICAL REVIEW B, VOLUME 65, 165113

Quantum orders and symmetric spin liquids

Xiao-Gang Wen*
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 3 June 2001; revised manuscript received 21 December 2001; published 10 April 2002)

A concept—quantum order—is introduced to describe a new kind of orders that generally appear in quantum states at zero temperature. Quantum orders that characterize the universality classes of quantum states (described by *complex* ground-state wave functions) are much richer than classical orders that characterize the universality classes of finite-temperature classical states (described by *positive* probability distribution functions). Landau's theory for orders and phase transitions does not apply to quantum orders since they cannot be described by broken symmetries and the associated order parameters. We introduced a mathematical object—projective symmetry group—to characterize quantum orders. With the help of quantum orders and projective symmetry groups, we construct hundreds of symmetric spin liquids, which have SU(2), U(1), or Z_2 gauge

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond
- **B:** chiral spin liquid
- **C:** Quantum dimer liquid
- **D:** QSL with spinon Fermi surface
- **E:** Algebraic spin liquid

Million-dollars Question

PRL **102,** 176401 (2009) PHYSICAL REVIEW LETTERS week ending 1 MAY 2009

Dynamics and Transport of the Z_2 Spin Liquid: Application to κ-(ET)$_2$Cu$_2$(CN)$_3$

Yang Qi, Cenke Xu, and Subir Sachdev

Department of Physics, Harvard University, Cambridge Massachusetts 02138, USA

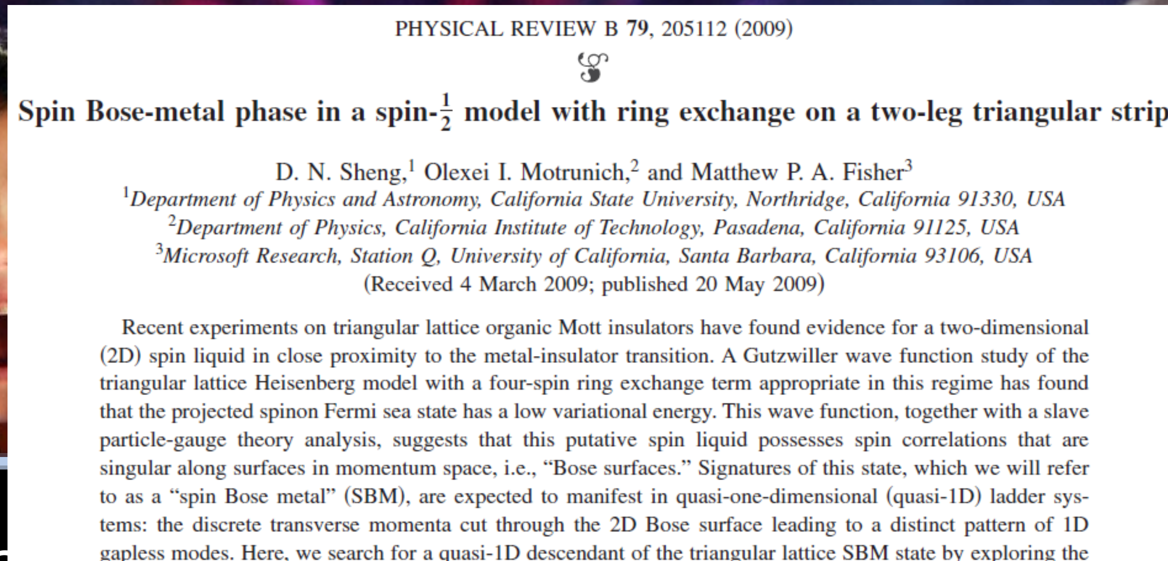
(Received 6 September 2008; published 29 April 2009; publisher error corrected 30 April 2009)

We describe neutron scattering, NMR relaxation, and thermal transport properties of Z_2 spin liquids in two dimensions. Comparison to recent experiments on the spin $S = 1/2$ triangular lattice antiferromagnet in κ-(ET)$_2$Cu$_2$(CN)$_3$ shows that this compound may realize a Z_2 spin liquid. We argue that the topological "vison" excitations dominate thermal transport, and that recent thermal conductivity experiments by M. Yamashita *et al.* have observed the vison gap.

What is the theory known to describe 2D QSL?

- **A:** resonating-valence-bond
- **B:** chiral spin liquid
- **C:** Quantum dimer liquid
- **D:** QSL with spinon Fermi surface
- **E:** Algebraic spin liquid
- **F:** Z_2 spin liquid

Million-dollars Question


PHYSICAL REVIEW B **79**, 205112 (2009)

Spin Bose-metal phase in a spin-$\frac{1}{2}$ model with ring exchange on a two-leg triangular strip

D. N. Sheng,[1] Olexei I. Motrunich,[2] and Matthew P. A. Fisher[3]
[1]*Department of Physics and Astronomy, California State University, Northridge, California 91330, USA*
[2]*Department of Physics, California Institute of Technology, Pasadena, California 91125, USA*
[3]*Microsoft Research, Station Q, University of California, Santa Barbara, California 93106, USA*
(Received 4 March 2009; published 20 May 2009)

Recent experiments on triangular lattice organic Mott insulators have found evidence for a two-dimensional (2D) spin liquid in close proximity to the metal-insulator transition. A Gutzwiller wave function study of the triangular lattice Heisenberg model with a four-spin ring exchange term appropriate in this regime has found that the projected spinon Fermi sea state has a low variational energy. This wave function, together with a slave particle-gauge theory analysis, suggests that this putative spin liquid possesses spin correlations that are singular along surfaces in momentum space, i.e., "Bose surfaces." Signatures of this state, which we will refer to as a "spin Bose metal" (SBM), are expected to manifest in quasi-one-dimensional (quasi-1D) ladder systems: the discrete transverse momenta cut through the 2D Bose surface leading to a distinct pattern of 1D gapless modes. Here, we search for a quasi-1D descendant of the triangular lattice SBM state by exploring the

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- **C:** Quantum dimer liquid
- **D:** QSL with spinon Fermi surface
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- **F:** Z_2 spin liquid
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Million-dollars Question

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Gapless spin liquid

Gapped spin liquid

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Elementary excitation characterizing QSL

What is the elementary excitation characterizing QSL?

- □ Gapless (Algebraic spin liquid)

  S = 1/2 1D spin chain: Gapless
  Algebraic spin correlation $\langle S(r)S(0)\rangle \sim r^{-\nu}$
  Quantum critical state

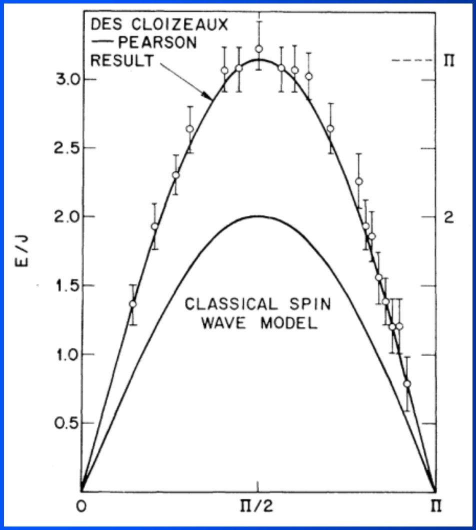
- □ Gapped (Topological spin liquid)

  S = 1, 1D spin chain: Haldane gap
  Exponentially decaying spin correlation $\langle S(r)S(0)\rangle \sim e^{-r/\xi}$

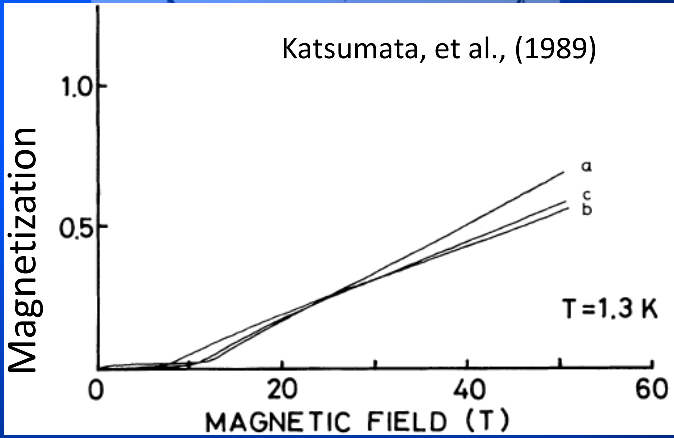
Experiment

Neutron scattering:
Not available for organic compound
Heat capacity, magnetization
Impurity problems in low temperature

Endoh, et al., (1974)



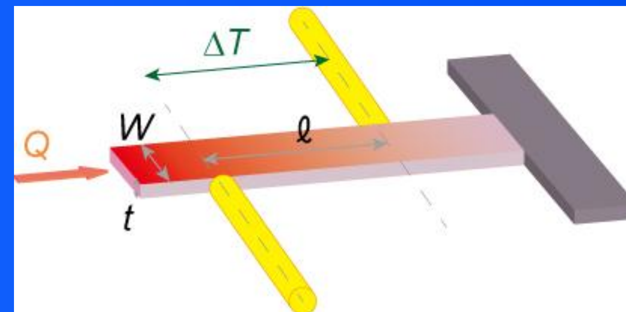
Katsumata, et al., (1989)

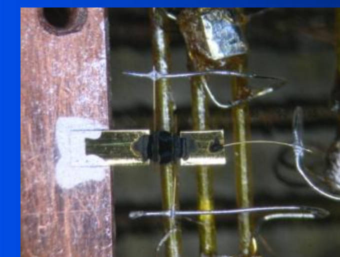


Thermal-transport & Magnetic torque

☐ Thermal conductivity

$$\frac{1}{wt}Q = \kappa\frac{\Delta T}{\ell}$$



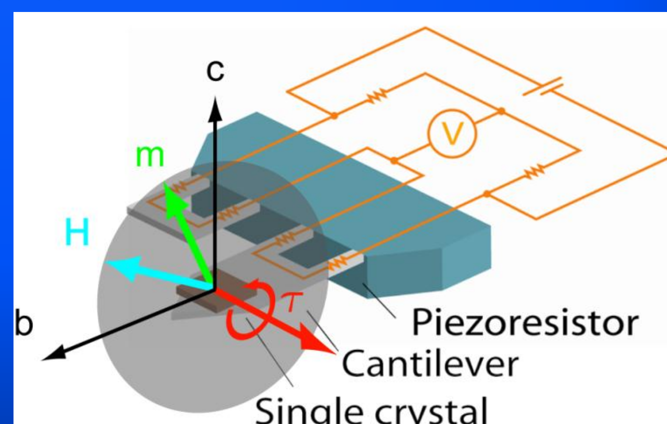


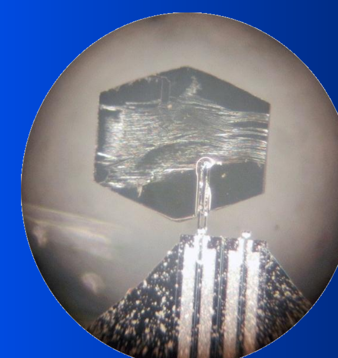
Selectively sensitive to itinerant excitations.
Not affected by localized impurity (Schottky anomaly).

κ_{xx} Measurement

☐ Magnetic torque measurement

$$\tau = \frac{1}{2}\mu_0 H^2 V \Delta\chi \sin 2\theta$$





- Only anisotropic susceptibility detected.
- Isotropic impurity (free spins) cancelled.
- High sensitivity. ONE single crystal measurement available.

Thermal conductivity in low temperature

- Enhancement of κ in spin liquid state

$$\kappa = \boldsymbol{\kappa_{spin}} + \kappa_{phonon}$$

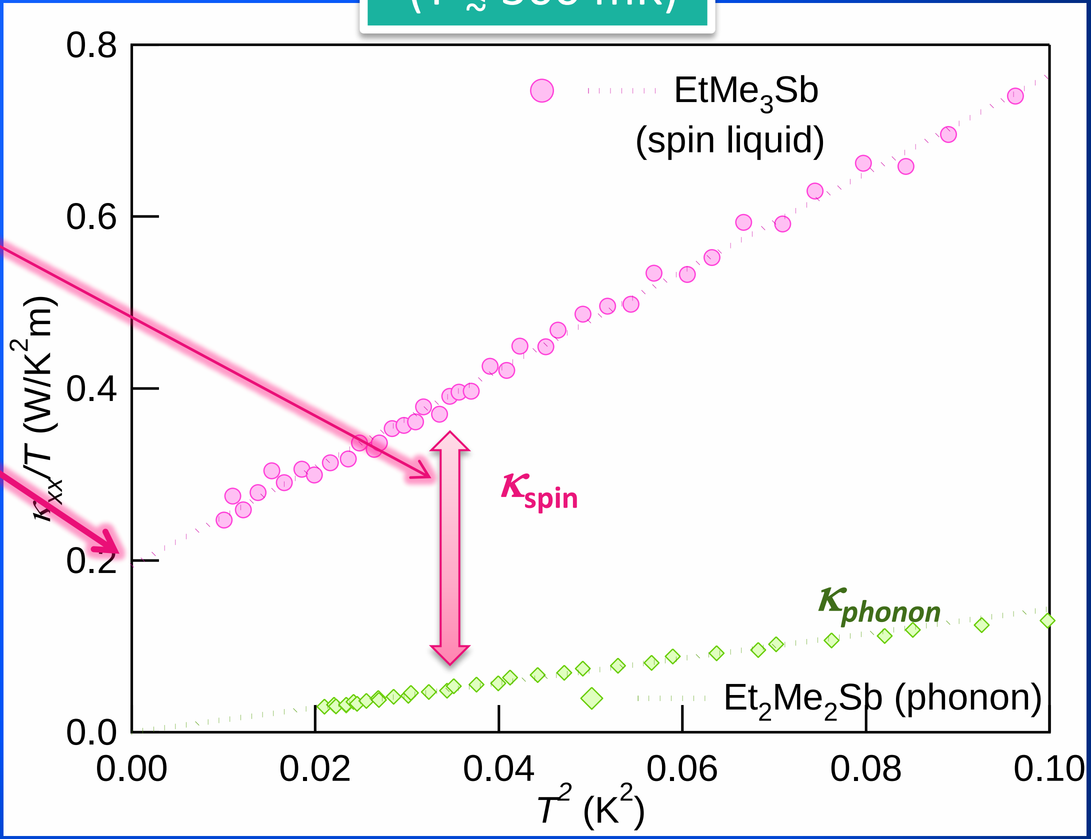
- Clear residual of *κ/T* !

$$\frac{\kappa_{spin}}{T} \longrightarrow 0.19\ \mathrm{W/K^2m}$$

Normally property of metals.
(comparable to κ of Brass
WF raw → ρ_0 = 13 μΩ•cm)
But, this is INSULATOR!!

Evidence for a gapless excitation, like electrons in normal metals.

κ/T vs T^2 plot
(T ≲ 300 mK)



M. Y., Science (2010)

Magnetic torque at High field

Magnetic torque under high magnetic field
@ LNCMI Grenoble & NIMS

M ∝ H from zero field

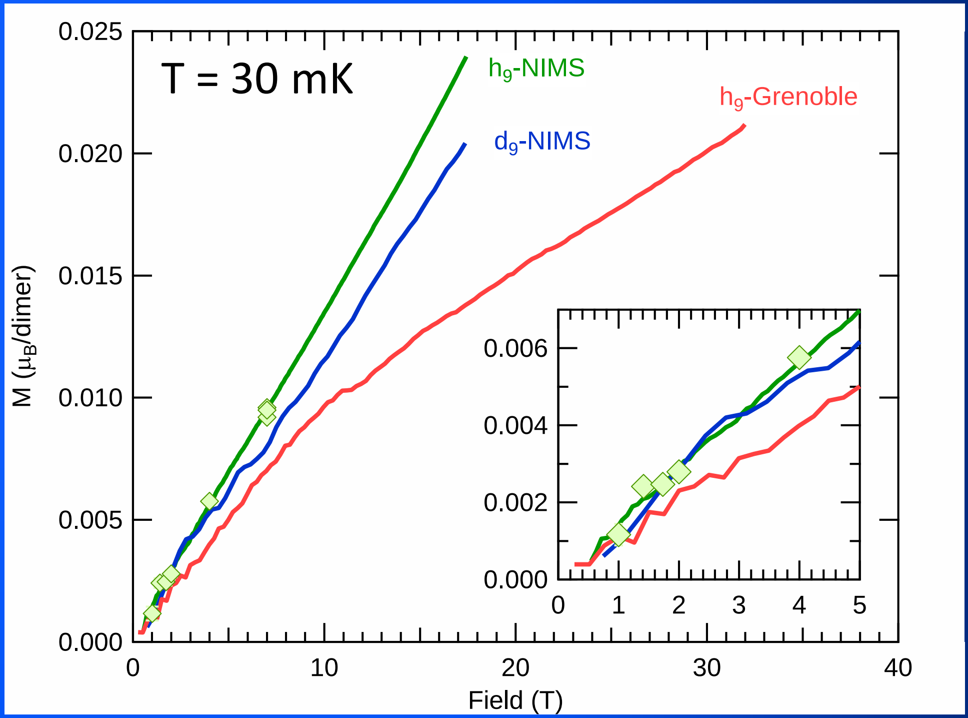
Gapless magnetic excitation

Property of magnetically LRO state
(Goldstone theorem)

$$\Delta \propto 1/\xi$$

Spin liquid with an algebraic correlation

$$\langle S(r)S(0)\rangle \sim r^{-\nu}$$

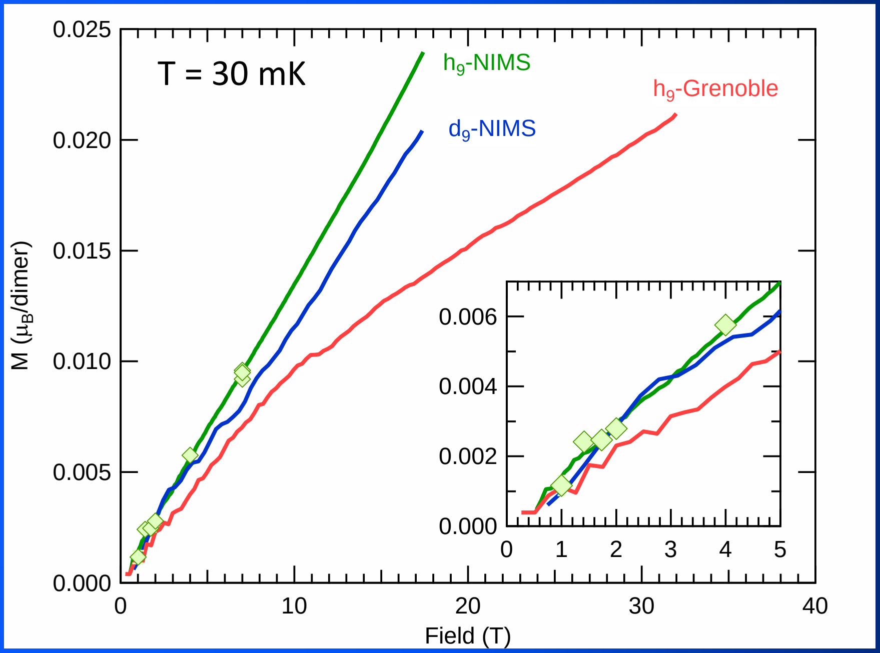


Magnetic torque at High field

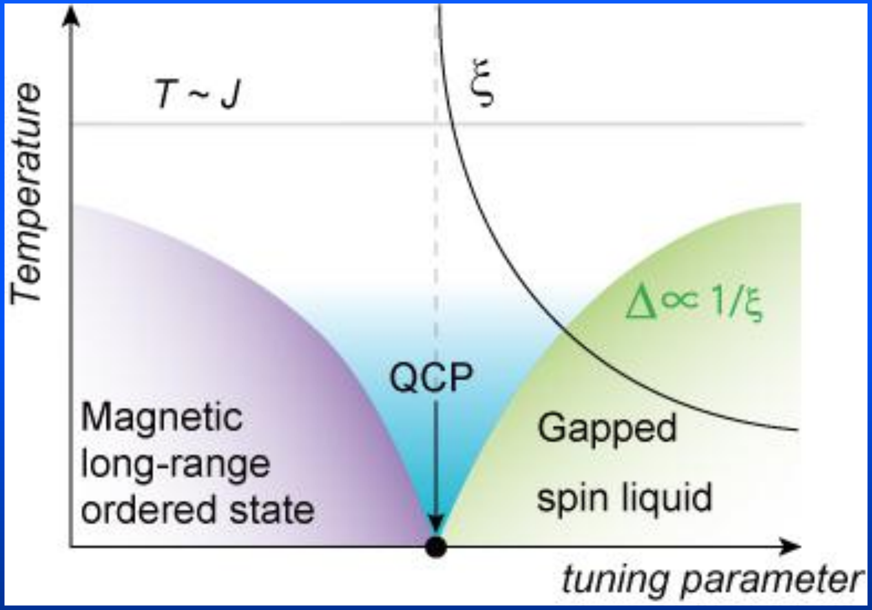
Gapless <u>magnetic</u> excitation

➡ ***Algebraic spin liquid***

Gapless magnetic excitation in Quantum spin liquid !!



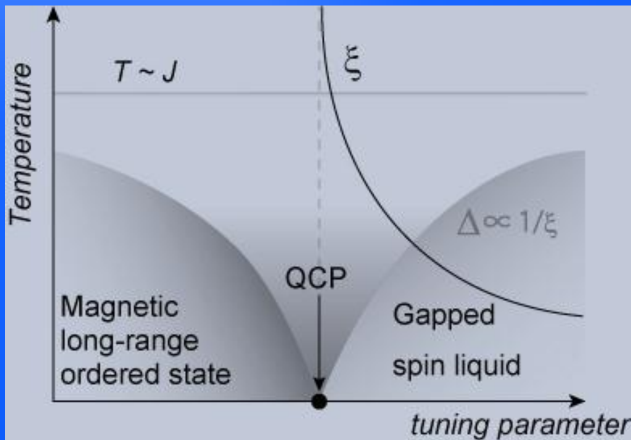
No LRO, No Gap down to ~*J*/10,000

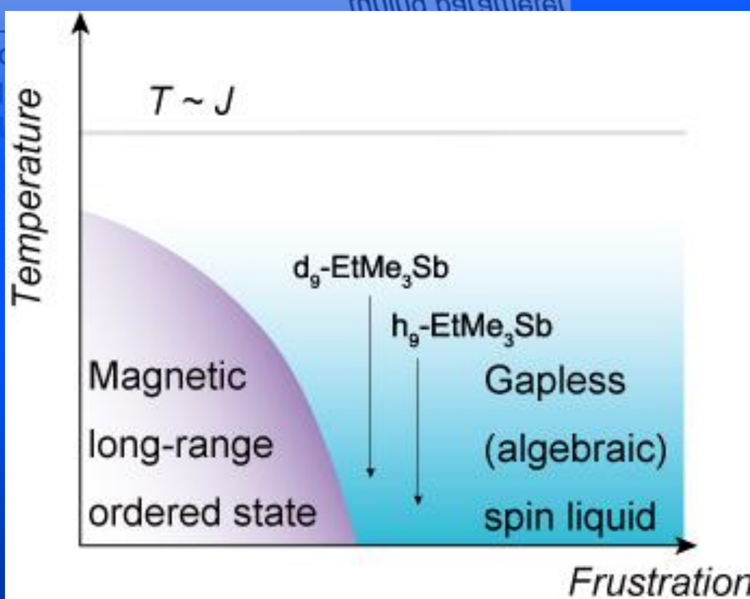


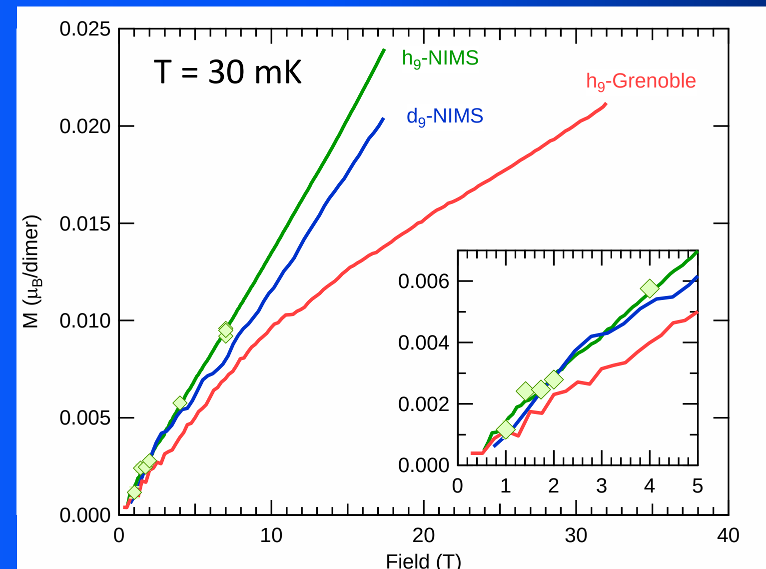
EXACTLY on QCP? Too nice to believe!
For both pristine and deuterated samples?

Chakravarty-Halperin-Nelson PRB (1989)
Sachdev & Keimer, *Physics Today* (2011)

Quantum Critical Phase?



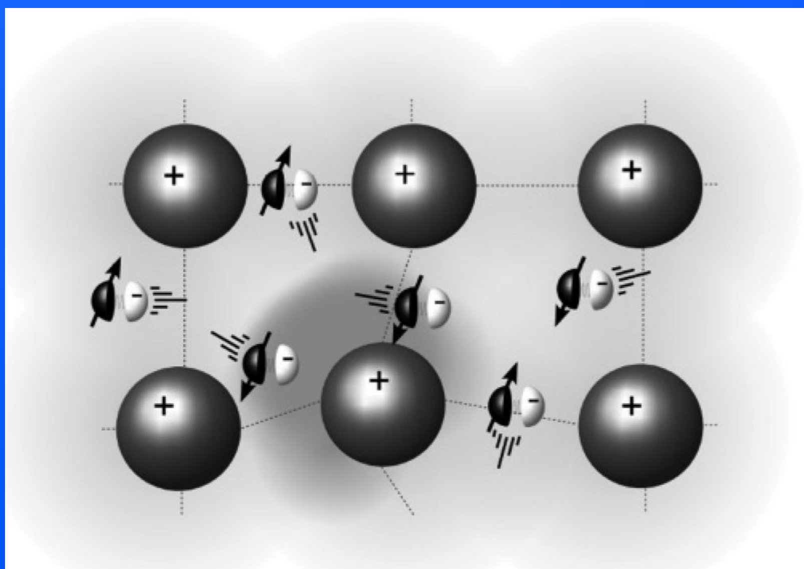




Stable gapless magnetic phase (quantum critical phase), rather than on QCP.

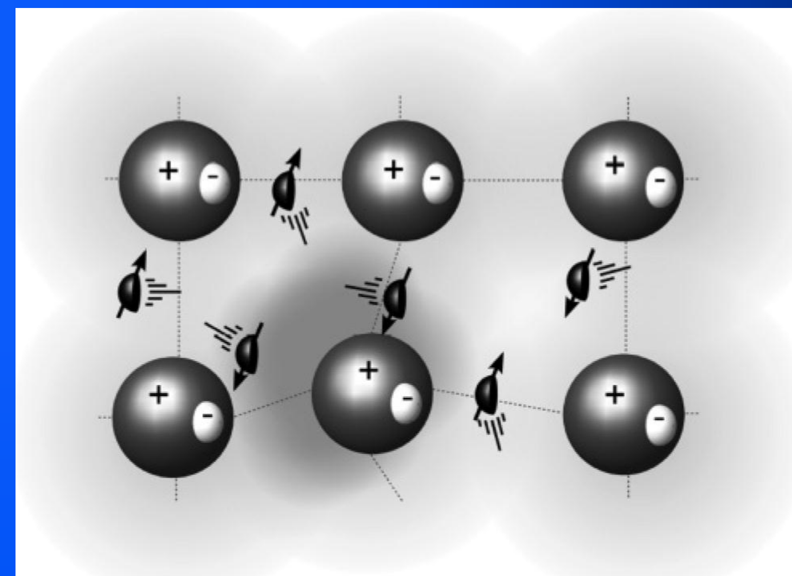
Spin-metal phase in Mott insulator

Metal



Both charge and spin can swim.

Spin-metal phase (spin liquid)



Only charges are frozen .

Mott-insulating transition

Spins can still swim

Geometrical frustration

Mross-Senthil, PRB **84**, 041102(R) (2010)

Summary

Minoru Yamashita et al.
Science **328**, 1246 (2010)

Thermal conductivity and magnetic torque measurement of $EtMe_3Sb[Pd(dmit)_2]_2$

- Finite κ/T and long mean free path
- Finite $\Delta\chi$ down to 30 mK.
- $\Delta M \propto H$ from almost zero field up to high field

Highly-entangled QSL with gapless magnetic excitation

Spin-metal phase (algebraic spin liquid)

Quantum critical phase, rather than on QCP?
Fermionic excitation? (Exotic) bosons?