



# Quantum Spin Liquids

Leon Balents, KITP

PSSCMP 2020

# Influenced by

Lucile Savary

SungBin Lee

Shigeki Onoda

Gang Chen

Oleg Starykh

Hong-Chen Jiang

Yi-Zhuang You

Xueyang Song

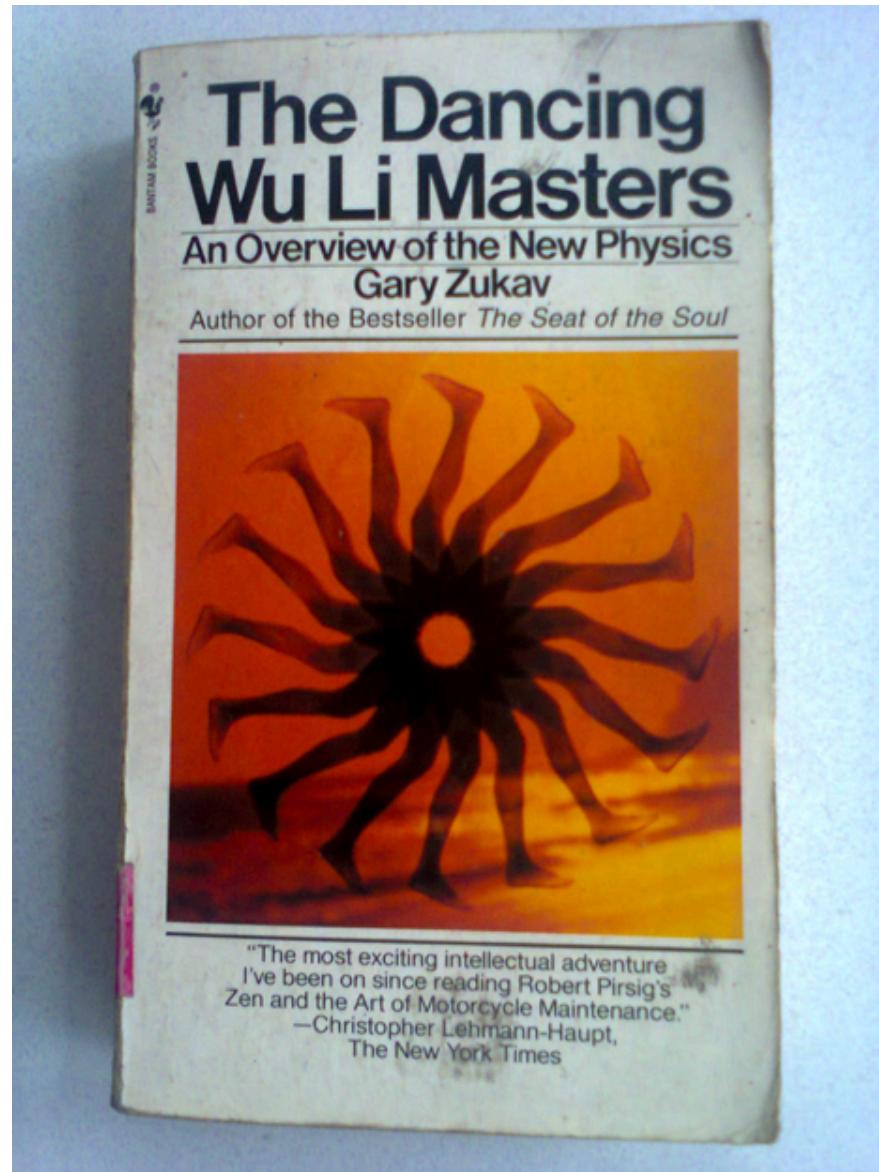
Jason Iaconis

Chunxiao Liu

Bruce Gaulin

Kate Ross

# Quantum Spin Liquid



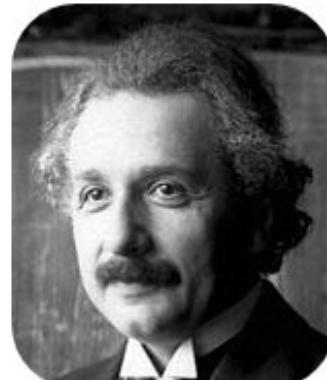
# Quantum non-locality

EPR

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



??where is the information??



A. Einstein

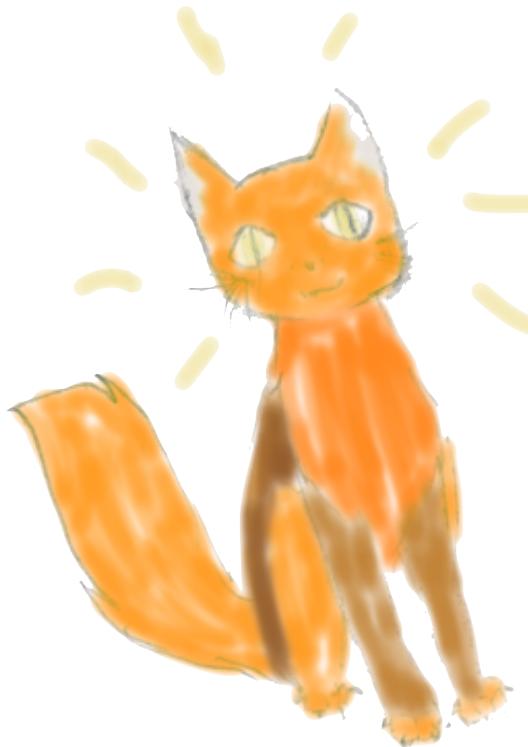


B. Podolsky

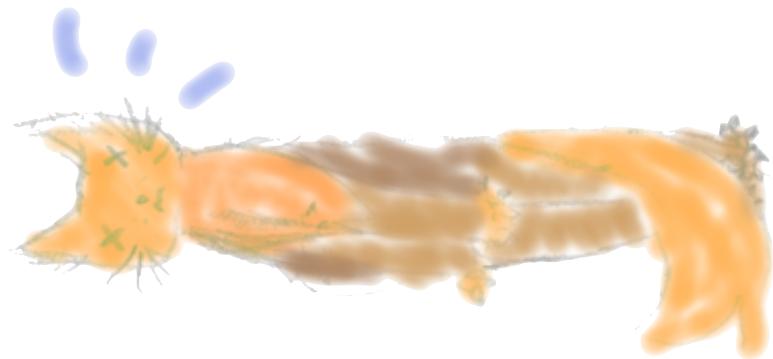


N. Rosen

# Schrödinger's Cat

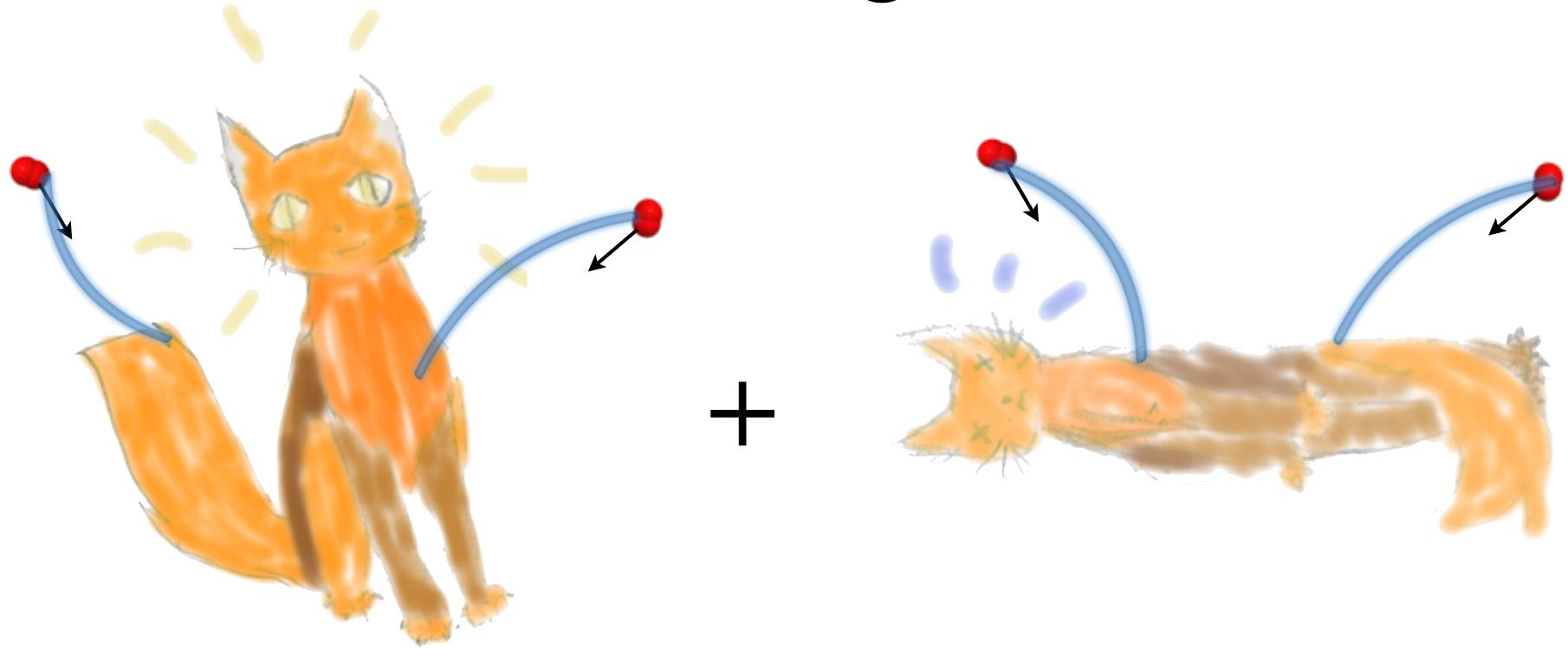


+



© Megan Balents

# Schrödinger's Cat



UNSTABLE to decoherence - uncontrolled entanglement with the environment



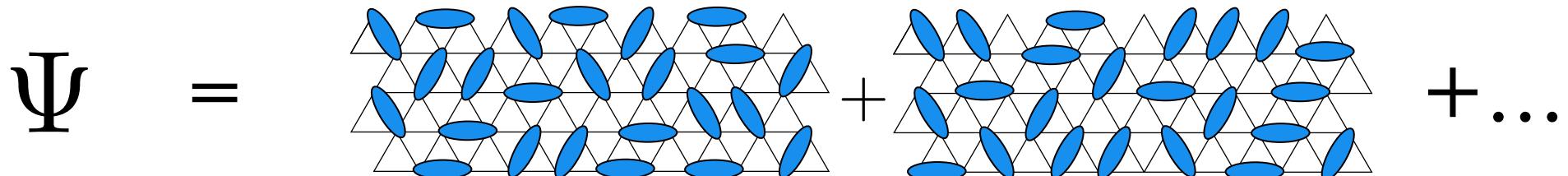
# Strange Stuff



Phil Anderson, 1973

a “quantum liquid” of spins

$$\text{blue oval} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



Resonating Valence Bond state

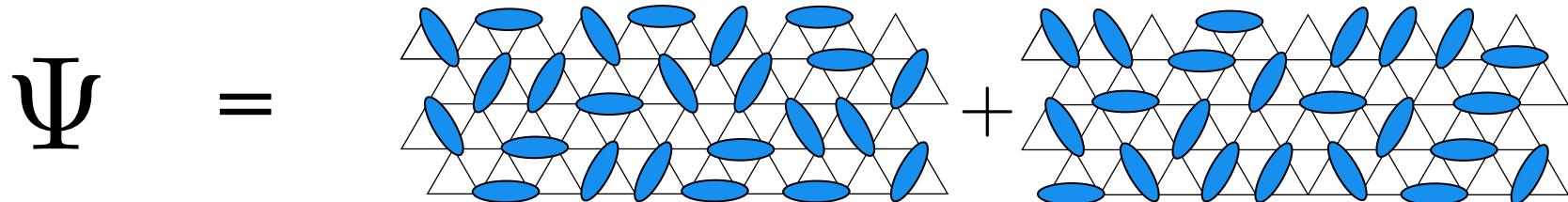
# Strange Stuff



Phil Anderson, 1973

a “quantum liquid” of spins

$$\text{blue oval} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



Resonating Valence Bond state



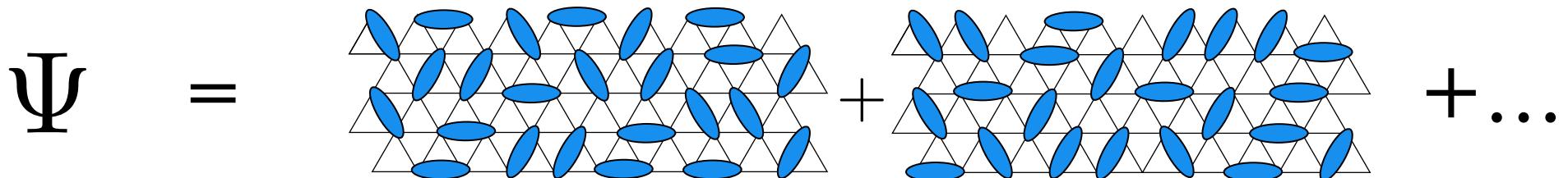
# Strange Stuff



Phil Anderson, 1973

a “quantum liquid” of spins

$$\text{blue oval} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

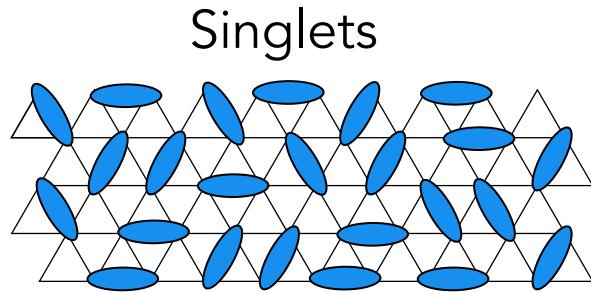
$$\Psi = \text{Diagram 1} + \text{Diagram 2} + \dots$$
A diagram representing the wavefunction  $\Psi$  as a sum of two components. Each component is a triangular lattice of sites, each containing a blue oval representing a spin. The first component (Diagram 1) shows a regular pattern of ovals. The second component (Diagram 2) shows a staggered pattern where ovals are only at specific lattice sites. The two diagrams are separated by a plus sign, followed by an ellipsis.

Two features:

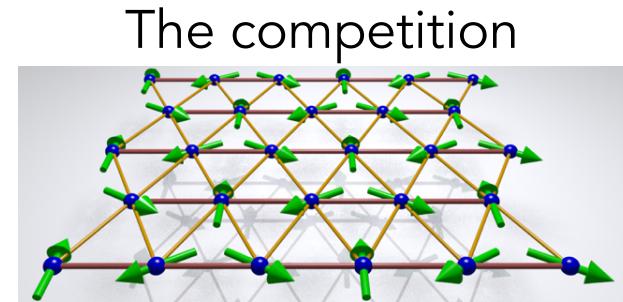
1. Spin-zero pairs
2. Massive superposition

# When do we expect RVB?

- Compare singlet energy to ordered energy:



versus



caveats:

Neglects superposition

Neglects zero point fluctuations

# When do we expect RVB?

- Compare singlet energy to ordered energy:

$$\vec{S}_i \cdot \vec{S}_j = \frac{1}{2} \left( \vec{S}_{\text{tot}}^2 - \vec{S}_i^2 - \vec{S}_j^2 \right)$$

$$\left\langle \vec{S}_i \cdot \vec{S}_j \right\rangle_{\text{AF}} = -S^2 x \quad \left\langle \vec{S}_i \cdot \vec{S}_j \right\rangle_{\text{singlet}} = -S(S+1)$$

# When do we expect RVB?

- Compare singlet energy to ordered energy:

$$\vec{S}_i \cdot \vec{S}_j = \frac{1}{2} \left( \vec{S}_{\text{tot}}^2 - \vec{S}_i^2 - \vec{S}_j^2 \right)$$

$$\left\langle \vec{S}_i \cdot \vec{S}_j \right\rangle_{\text{AF}} = -S^2 x \quad \left\langle \vec{S}_i \cdot \vec{S}_j \right\rangle_{\text{singlet}} = -S(S+1)$$

$$E_{\text{AF}}/\text{bond} = -S^2 x$$

angular factor

$$E_{\text{singlet}}/\text{bond} = -S(S+1)f_{\text{singletbonds}}$$

$$= -\frac{1}{z}S(S+1)$$

Coordination number

- Favorable for small  $S$ , small  $z$ , small  $x$  (frustration)

To understand what is strange about spin liquids, we should understand

# Ordinary (local) Matter

We can consistently assign local properties (elastic moduli, etc.) and obtain all large-scale properties



- Measurements far away do not affect one another
- From local measurements we can deduce the global state

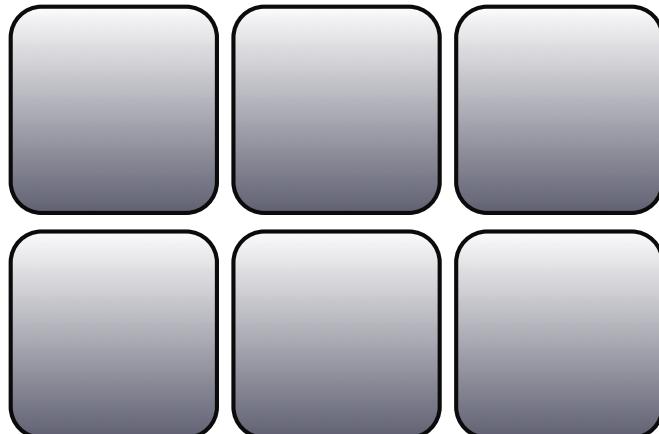
# Ordinary (local) Matter

Hamiltonian is local

$$H = \sum_x \mathcal{H}(x) \quad \mathcal{H}(x) \text{ has local support near } x$$

Ground state is “essentially”  
a product state

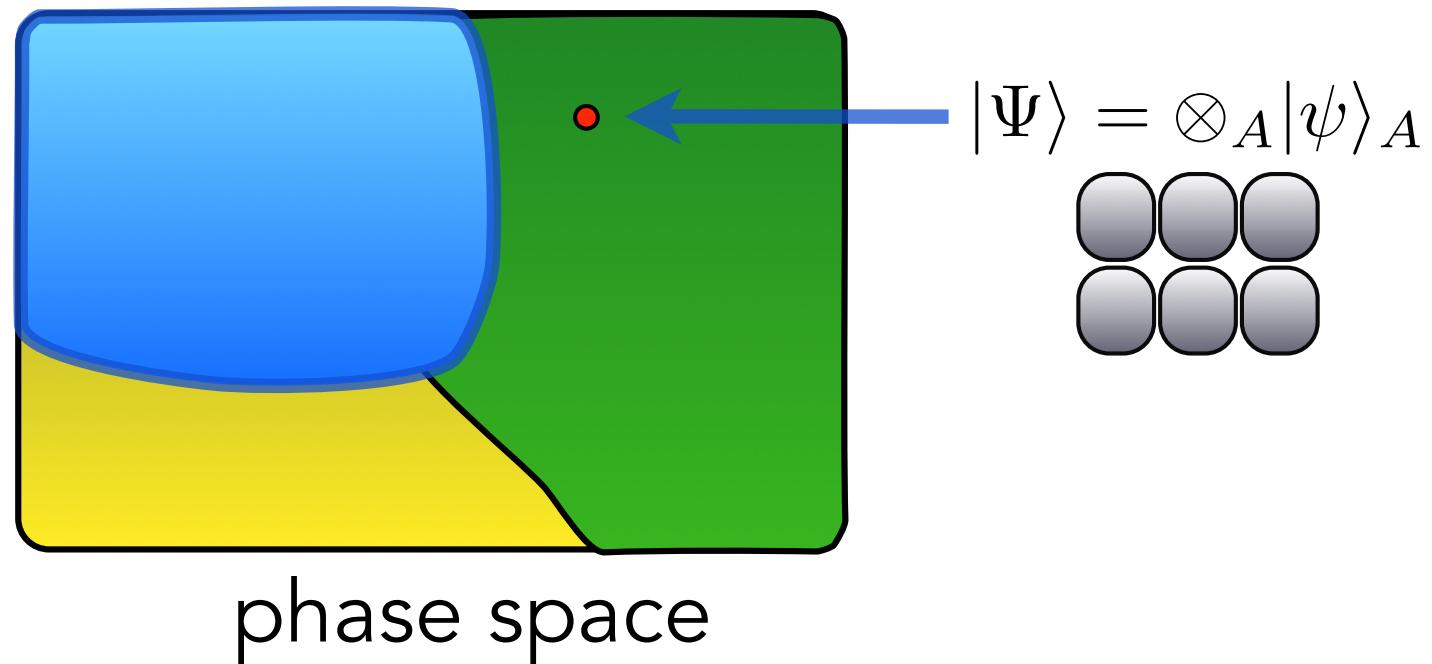
$$|\Psi\rangle = \otimes_A |\psi\rangle_A$$



no entanglement  
between blocks

# “Essentially” a product state?

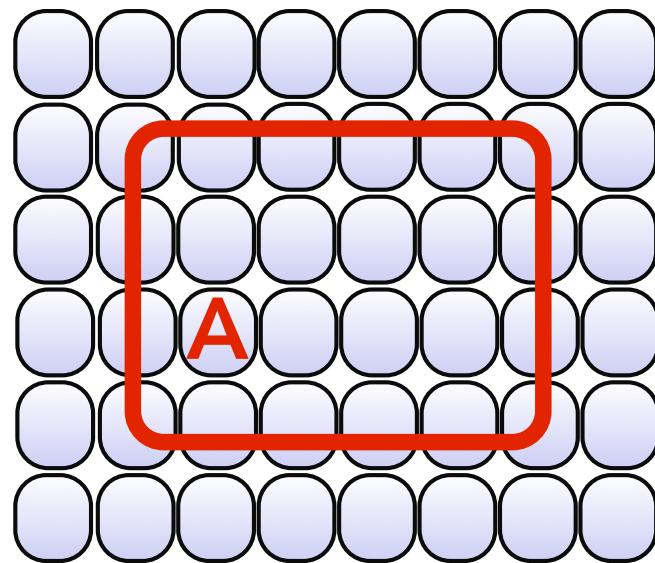
- Adiabatic continuity



n.b. This is not true for gapless fermi systems

# “Essentially” a product state?

- Entanglement scaling



$$\rho_A = \text{Tr}_{\bar{A}} |\Psi\rangle\langle\Psi|$$

$$S(A) = -\text{Tr}_A (\rho_A \ln \rho_A)$$

$$S(A) \sim \sigma L^{d-1} \text{ area law}$$

satisfied with exponentially small corrections

# Best example: ordered magnet

Hamiltonian

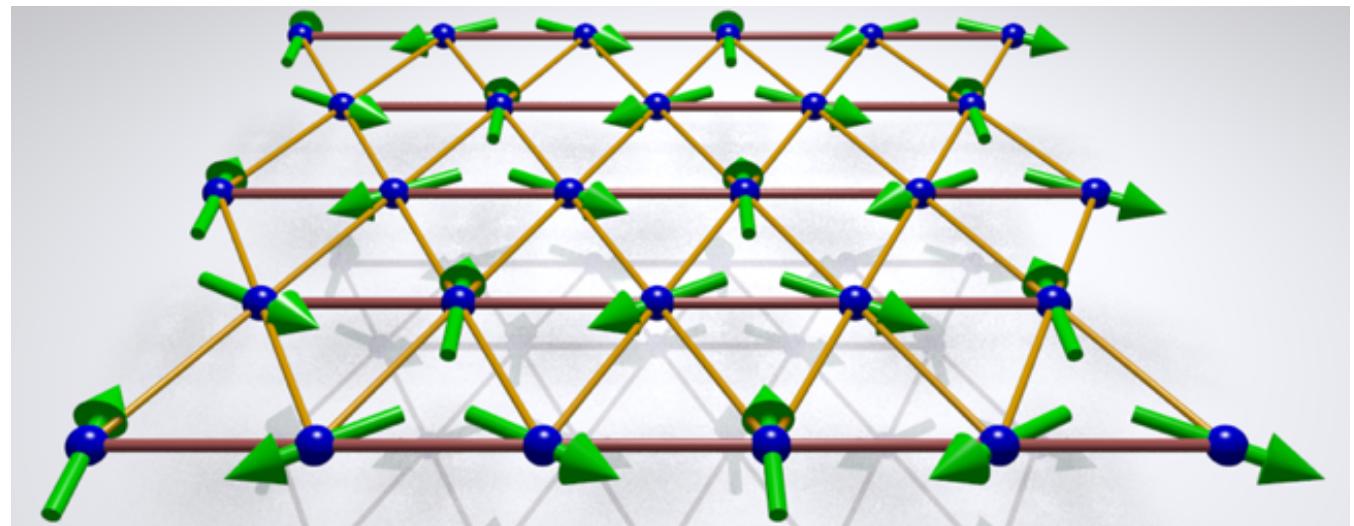
$$H = \sum_{(ij)} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

exchange is short-  
range: local

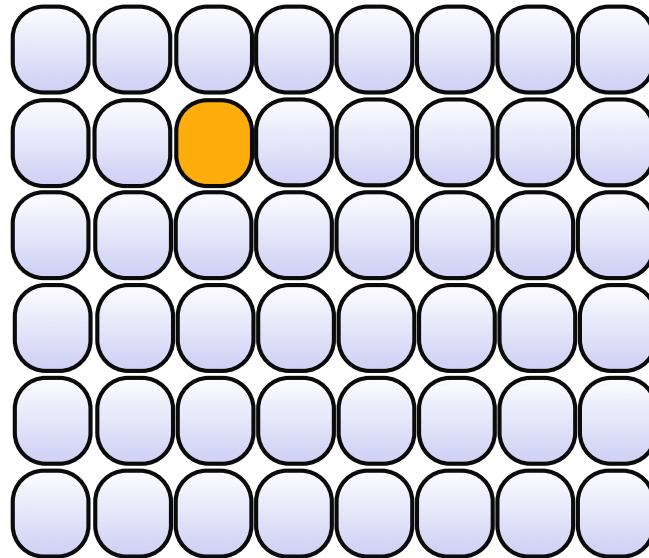
ordered state

$$|\Psi\rangle \approx \bigotimes_i |\mathbf{S}_i \cdot \hat{\mathbf{n}}_i = +S\rangle$$

block is a single  
spin



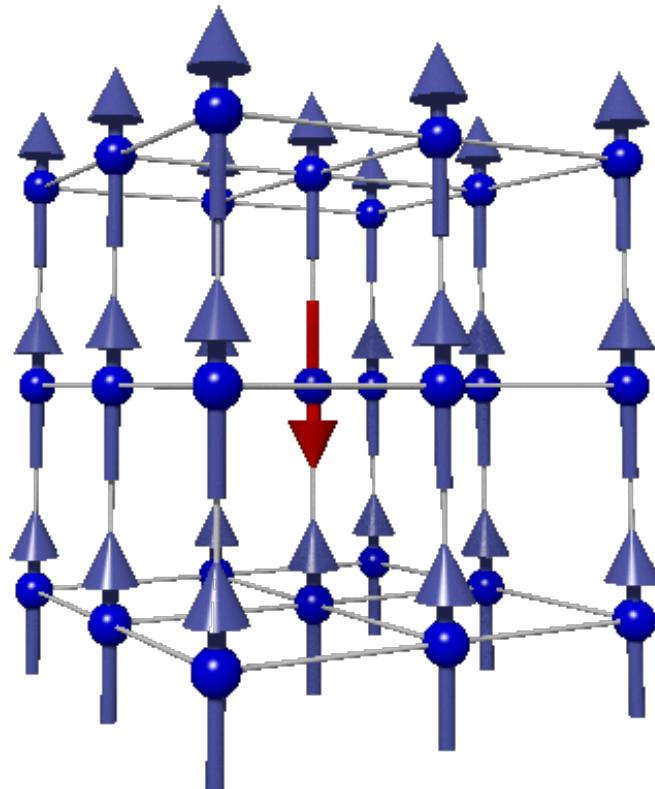
# Quasiparticles



excited states  $\sim$  excited  
levels of one block

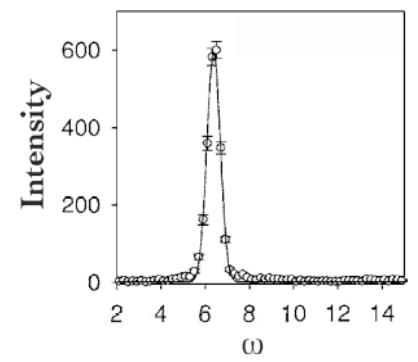
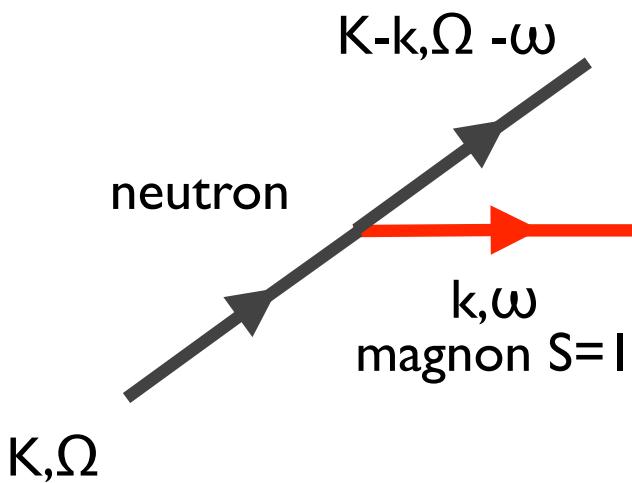
- local excitation can be created with operators in one block
- localized excitation has discrete spectrum with non-zero gap, and plane wave forms sharp band
- quantum numbers consistent with finite system: no emergent or fractional quantum numbers

# Spin wave



$$\omega(k) \approx \Delta - 2t \cos k_x a - \dots$$

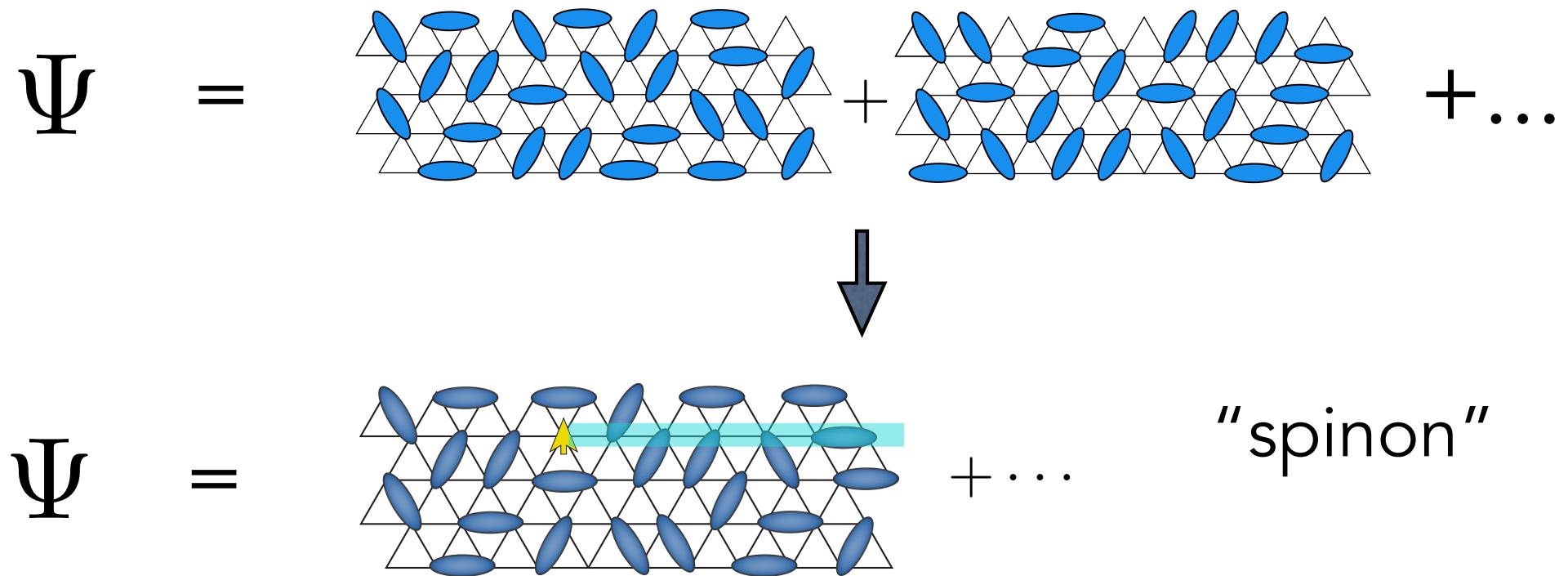
$$|f\rangle = S_k^+ |i\rangle$$



Line shape in  $\text{Rb}_2\text{MnF}_4$

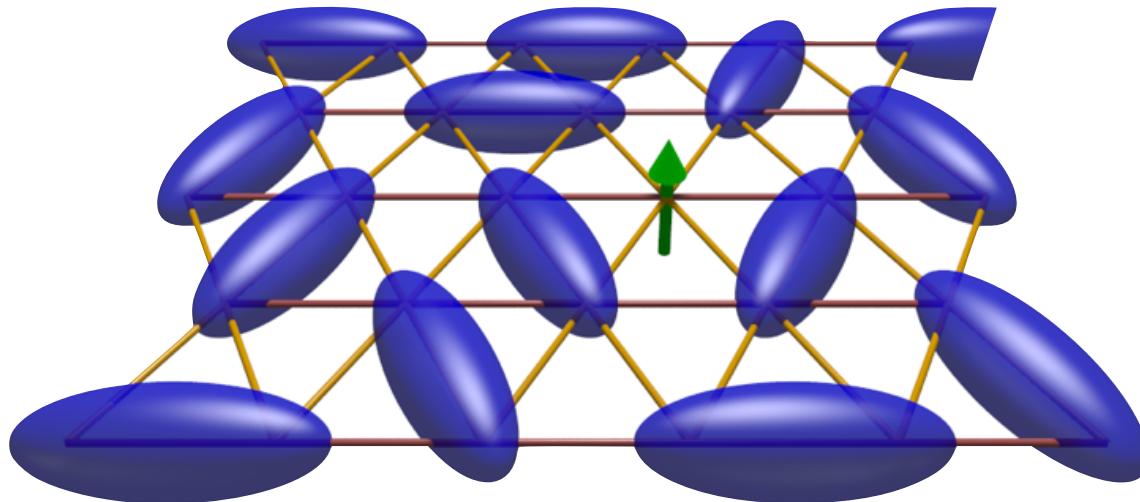
# Quantum spin liquid

Entanglement -> non-local excitation



“quasiparticle” above a non-zero gap

# Fractional quantum number

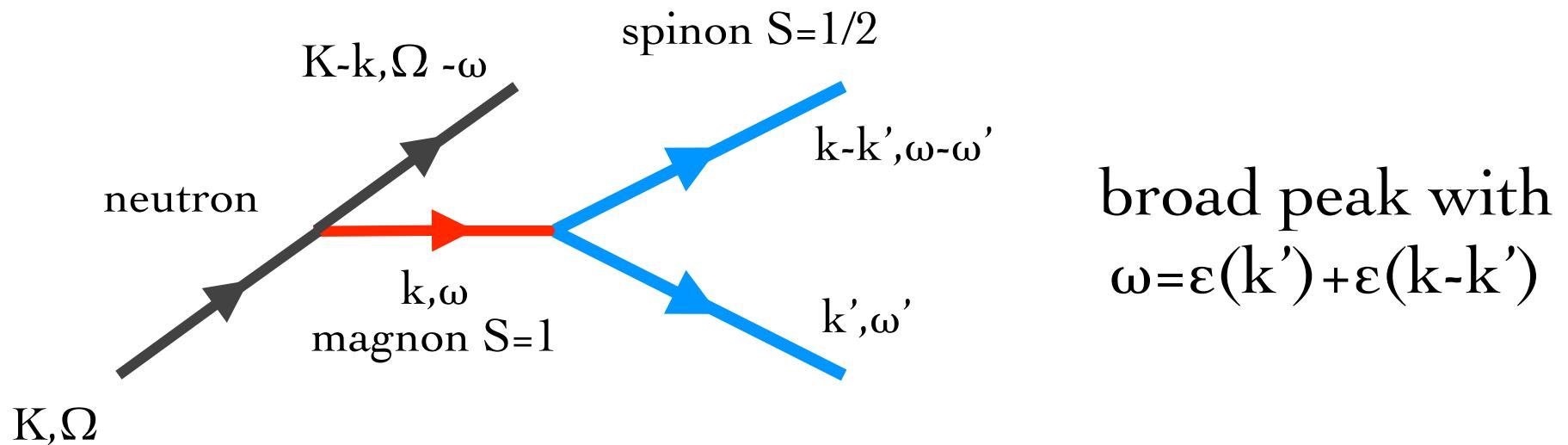


excitation with  $\Delta S = 1/2$   
not possible for any finite  
cluster of spins

always created in pairs by any  
local operator

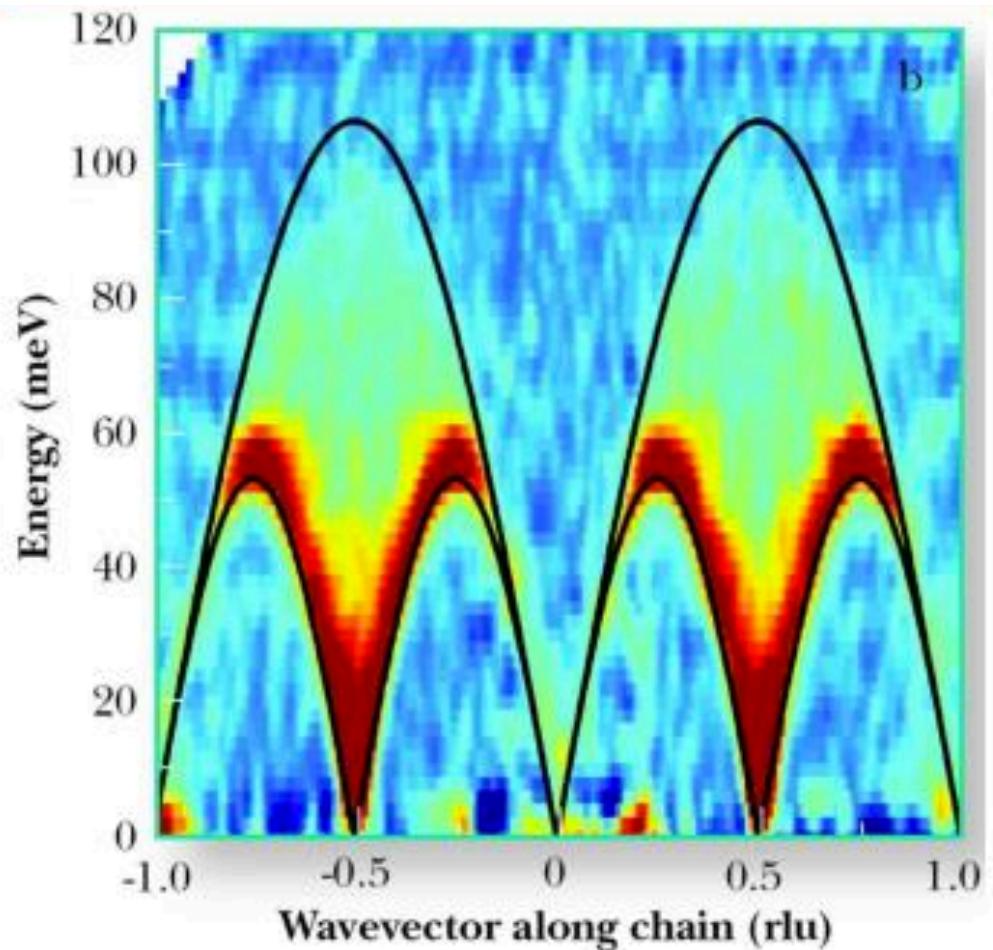
# No spin waves

- In a quantum spin liquid, the elementary spin excitations are *fractional*,  $S=1/2$  spinons



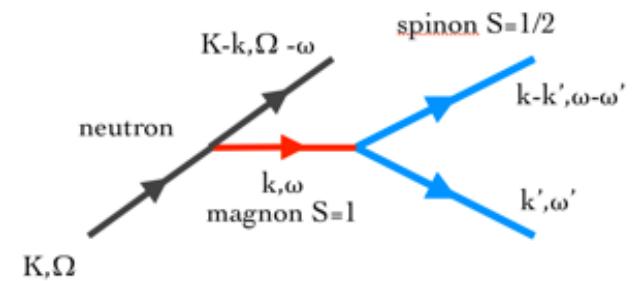
- Sharp peaks should be reduced or absent in the spin structure factor

# c.f. One dimension

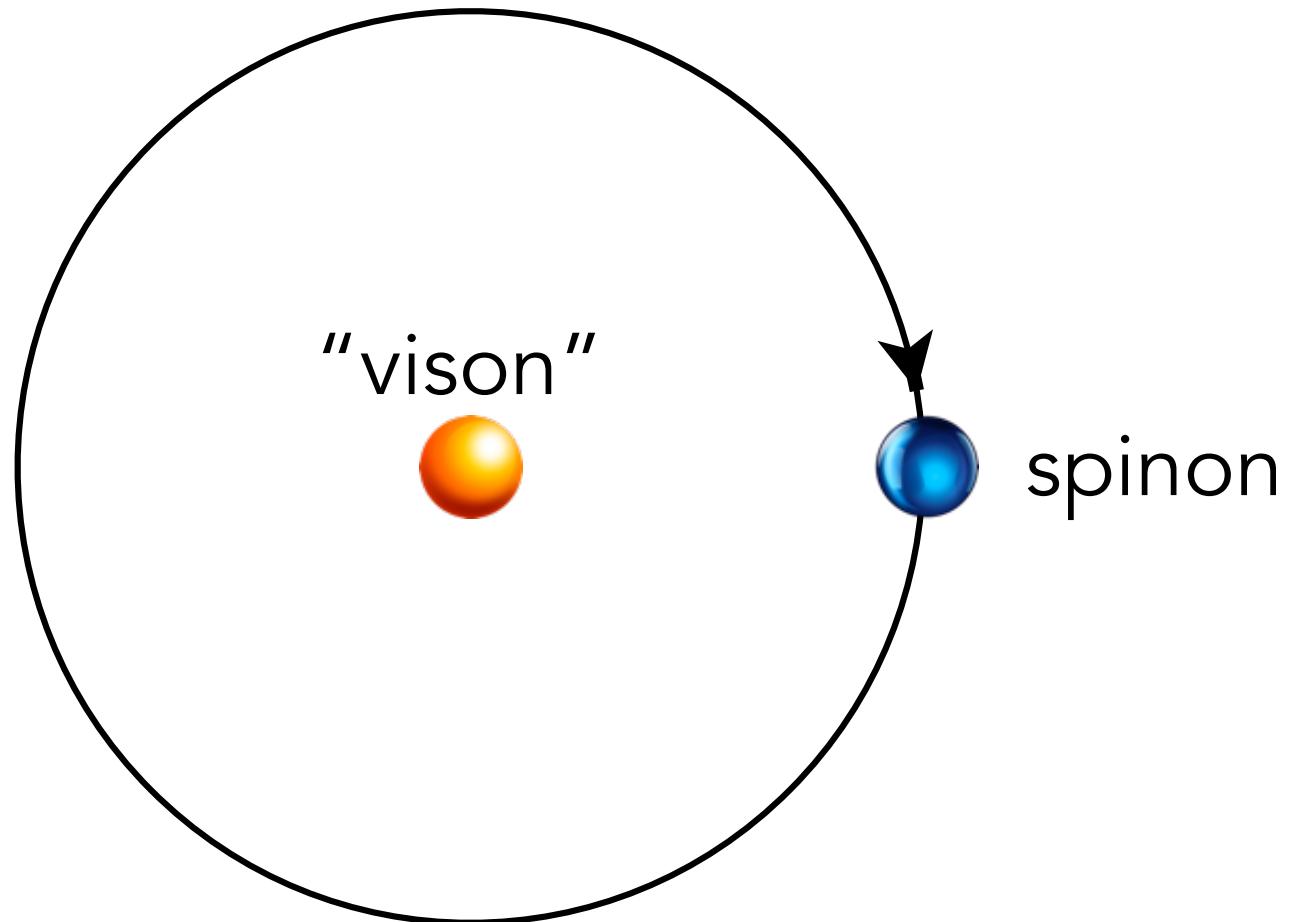


A. Tennant *et al*, 2001

$\text{KCuF}_3$



# Anyons



$$\Psi \rightarrow -\Psi$$

"mutual semions"

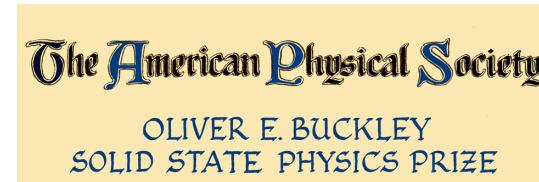


X.-G. Wen



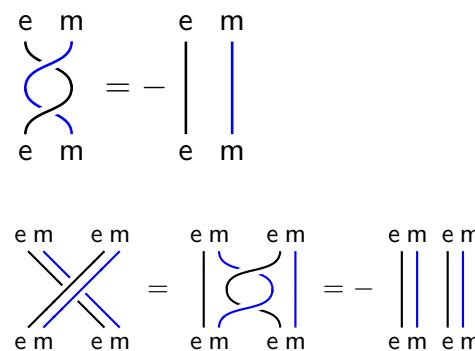
A. Kitaev

# Topological phases



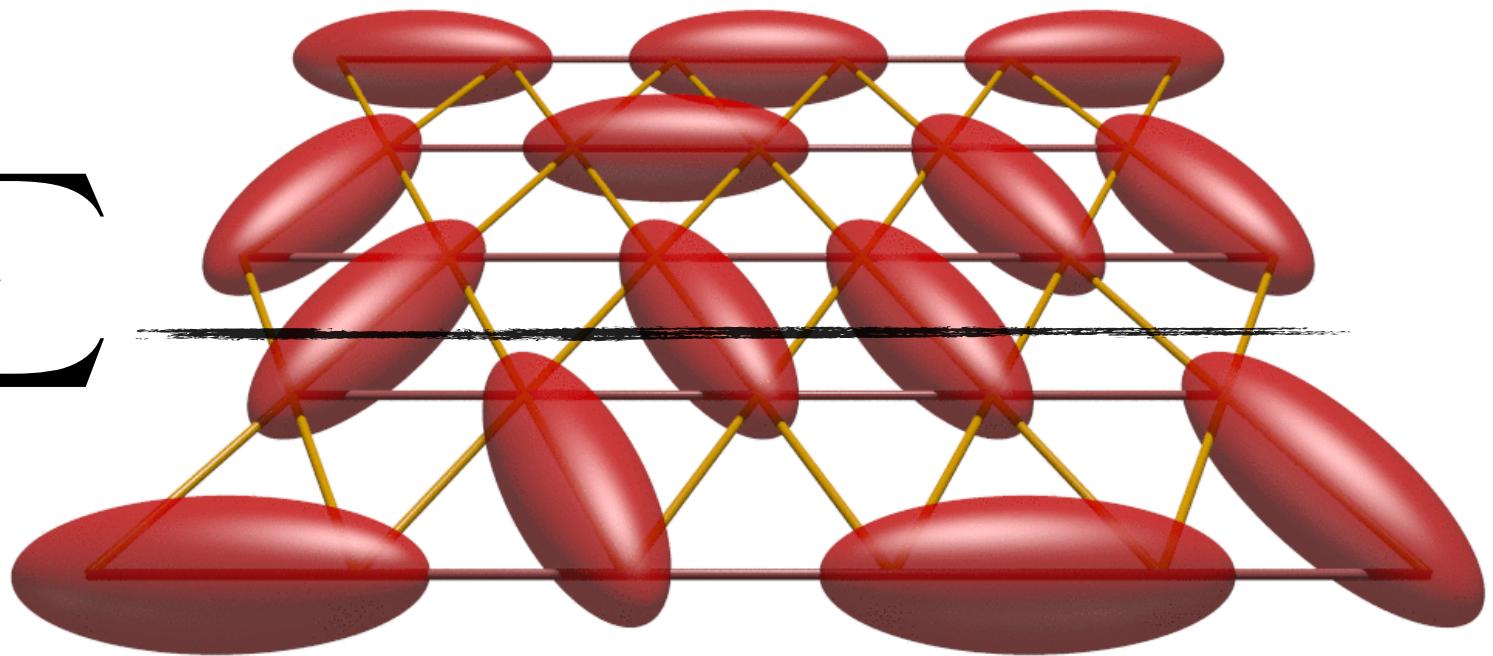
Anderson's RVB state is thus an example of a “topological phase” - the best understood sort of QSL

Understood and classified by anyons and their braiding rules in 2d



# Stability

$$\Psi = \sum$$



Robustness arises from topology: a QSL is a stable phase of matter (at T=0)

# Many kinds of QSLs

$$\Psi = \begin{array}{c} \text{Diagram of a triangular lattice with blue ovals representing spins, with a red arrow pointing to it labeled '#'} \\ + \end{array} \begin{array}{c} \text{Diagram of a triangular lattice with blue ovals representing spins, with a red arrow pointing to it labeled '#'} \\ + \dots \end{array}$$

For  $\sim 500$  spins, there are more amplitudes than there are atoms in the visible universe!

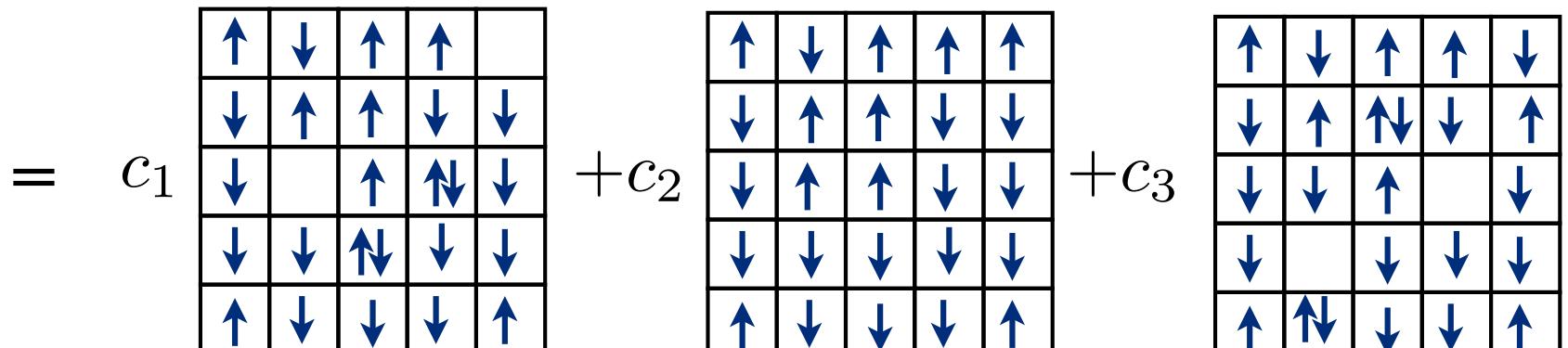
Different choices of amplitudes can realize different QSL phases of matter.

# Gutzwiller Construction

- Construct QSL state from free fermi gas with spin, with 1 fermion per site ( $S=0$ )

$$|\Psi_0\rangle = \prod_{k \in FS} c_{k\uparrow}^\dagger c_{k\downarrow}^\dagger |0\rangle$$

“partons”  
“spinons”

$$= c_1 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & & \uparrow & \uparrow \downarrow & \downarrow \\ \hline \downarrow & \downarrow & \uparrow \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_2 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline \downarrow & \uparrow & \uparrow & \uparrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_3 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow \downarrow & \downarrow & \uparrow \\ \hline \downarrow & \downarrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \uparrow \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + \dots$$


# Gutzwiller Construction

- Project out any components with empty or doubly occupied sites

$$|\Psi\rangle = \hat{P}_G |\Psi_0\rangle$$

“partons”  
“spinons”

$$= c_1 \cancel{\begin{array}{|c|c|c|c|c|} \hline & \uparrow & \downarrow & \uparrow & \uparrow \\ \hline & \downarrow & & \uparrow & \downarrow \\ \hline & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline & \uparrow & \downarrow & \uparrow & \downarrow \\ \hline & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline & \uparrow & \downarrow & \downarrow & \downarrow \\ \hline \end{array}} + c_2 \begin{array}{|c|c|c|c|c|} \hline & \uparrow & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline & \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline & \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline & \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline & \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_3 \cancel{\begin{array}{|c|c|c|c|c|} \hline & \uparrow & \downarrow & \uparrow & \uparrow & \downarrow \\ \hline & \downarrow & \uparrow & \uparrow & \downarrow & \uparrow \\ \hline & \downarrow & \downarrow & \uparrow & \downarrow & \downarrow \\ \hline & \downarrow & \uparrow & \downarrow & \downarrow & \downarrow \\ \hline & \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array}} + \dots$$

# Gutzwiller Construction

- Can build many QSL states by choosing different free fermion states

$$|\Psi\rangle = \hat{P}_G |\Psi_0\rangle$$

“partons”  
“spinons”

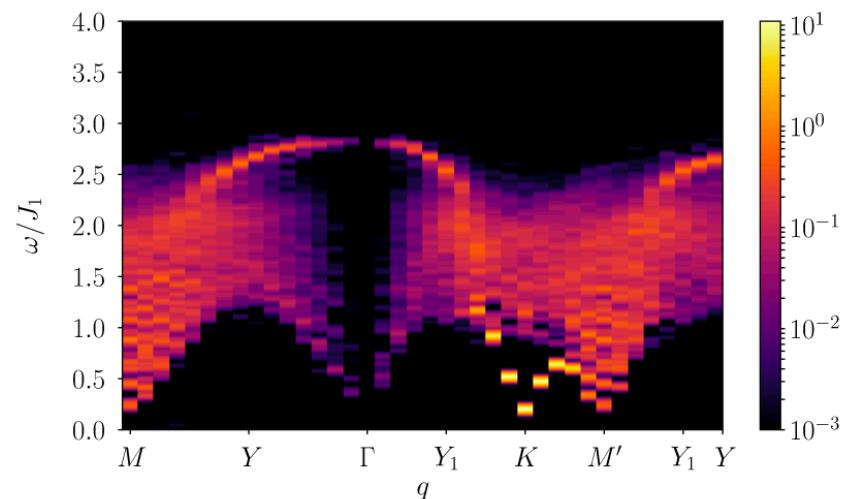
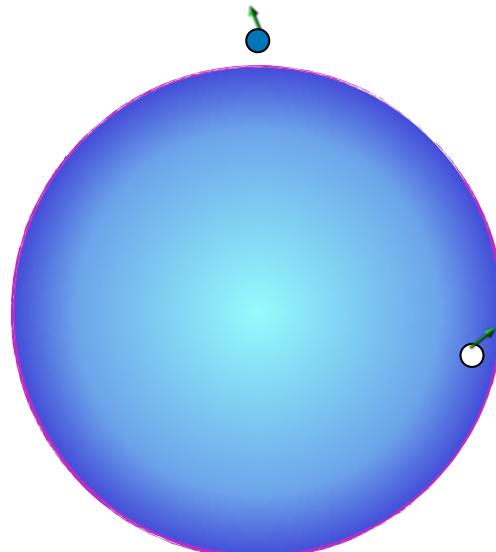
$$= c_1 \cancel{\begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline \downarrow & & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow & \uparrow & \uparrow \\ \hline \downarrow & \downarrow & \uparrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \uparrow & \downarrow & \uparrow \\ \hline \end{array}} + c_2 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_3 \cancel{\begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \uparrow \\ \hline \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \uparrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array}} + \dots$$

# Gutzwiller Construction

- Partons/spinons as quasiparticles

$$|\Psi'\rangle = \hat{P}_G \left( c_{k\alpha}^\dagger c_{q\beta} |\Psi_0\rangle \right)$$

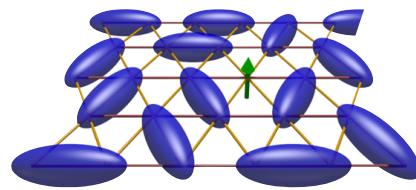
Two spinons but independent  
(before projection)



Applied to triangular lattice  
to compute  $S(q, \omega)$ ,  
F. Ferrari + F. Becca, 2019

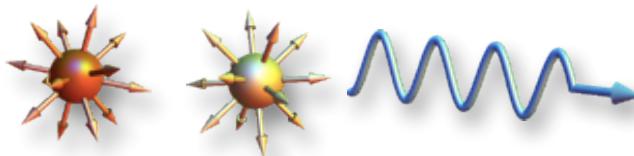
# Classes of QSLs

- Topological QSLs



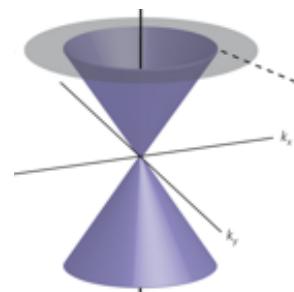
projected  
superconductor

- $U(1)$  QSL



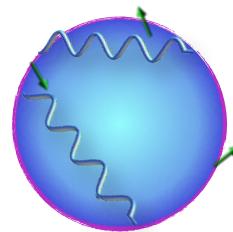
projected 3d band  
insulator

- Dirac QSLs



projected  
graphene

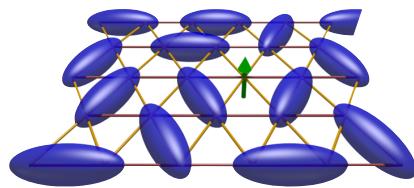
- Spinon Fermi surface



projected  
metal

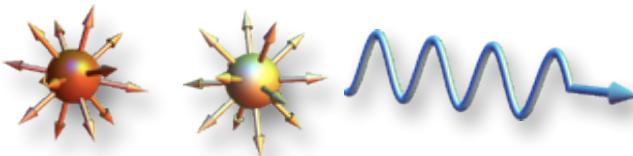
# Classes of QSLs

- Topological QSLs



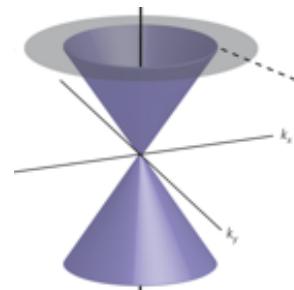
anyonic  
spinons

- $U(1)$  QSL



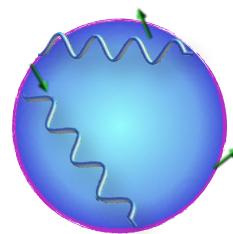
electric+magnetic  
monopoles, photon

- Dirac QSLs



strongly  
interacting  
Dirac fermions

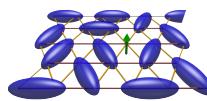
- Spinon Fermi surface



non-Fermi  
liquid "spin  
metal"

# Classes of QSLs

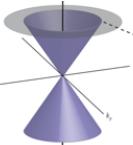
- Topological QSLs



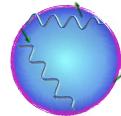
- U(1) QSL



- Dirac QSLs

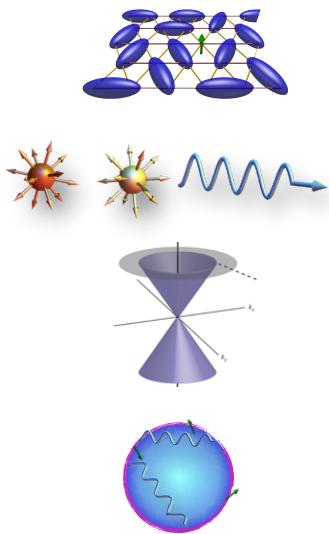


- Spinon Fermi surface



These spin liquids are all different phases of matter, and are rather different from one another. Like the corresponding unprojected states, their phenomenology can be quite distinct. Too naive to look for a single identifying feature for all QSLs.

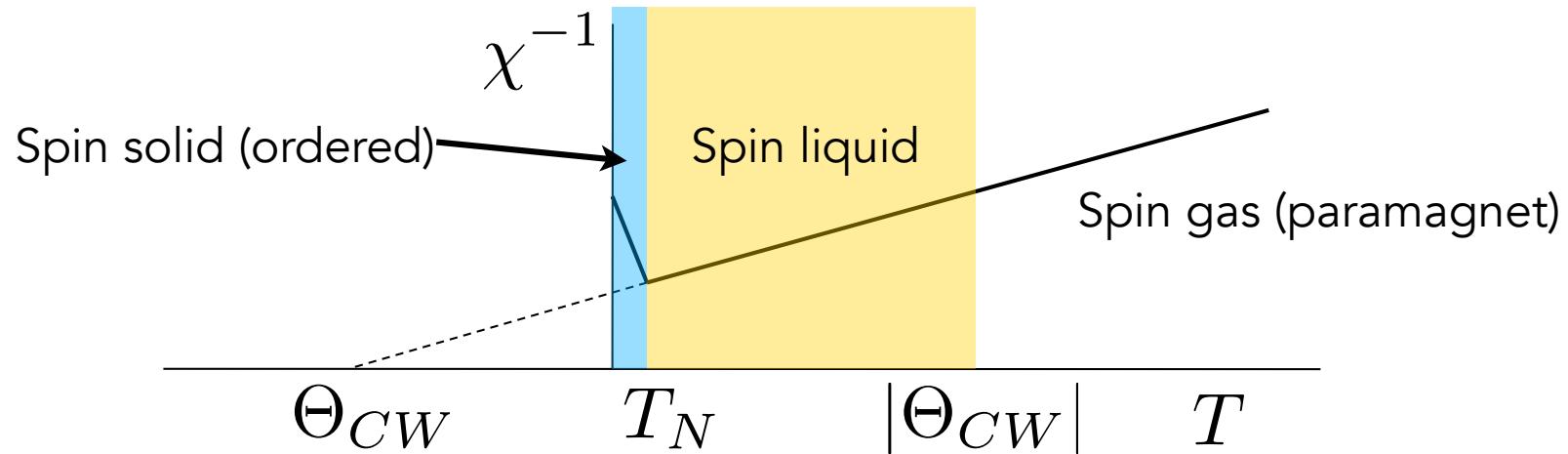
# Strange stuff



where do we find it?



# Ramirez Plot

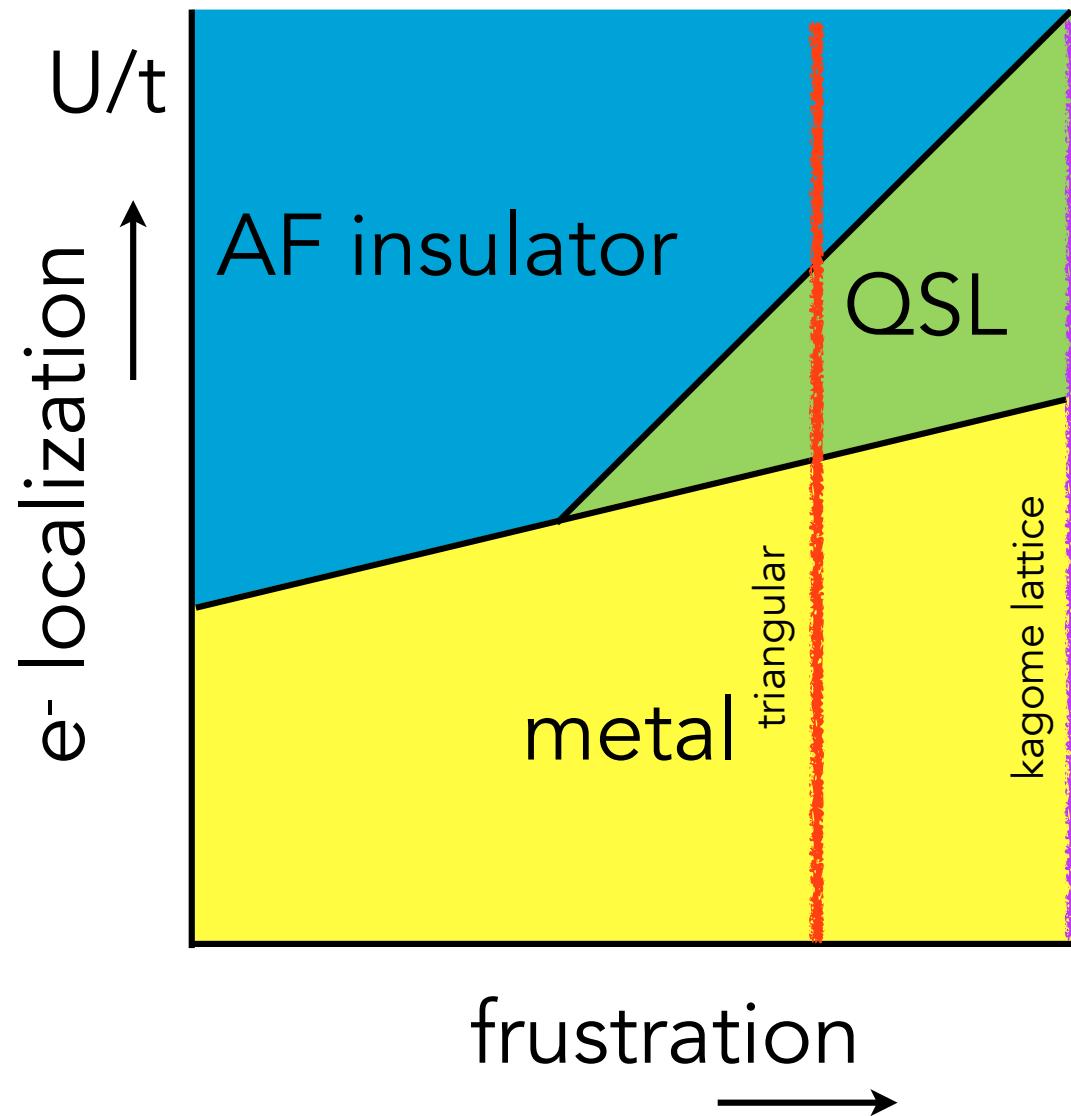


- Local moments: Curie-Weiss law at high T
$$\chi \sim \frac{A}{T - \Theta_{CW}}$$
- Frustration parameter:  $f = |\Theta_{CW}|/T_N$
- Larger  $f \gg 1$  is better.  $f = \infty$  for true QSL

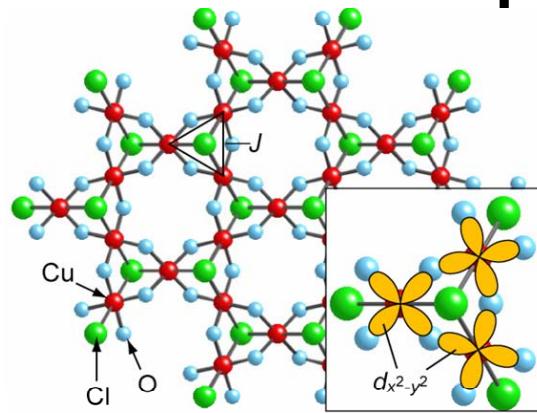
# Materials criteria

- $S=1/2$  spins
- Geometrical or exchange frustration
- Significant charge fluctuations
- Exotic interactions (c.f. Spin-orbit coupling)

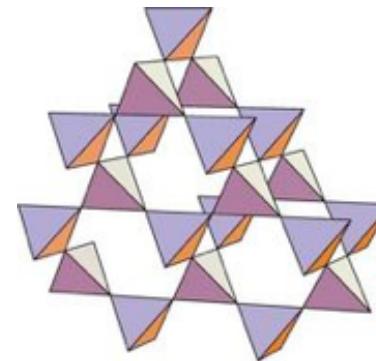
# Where to look?



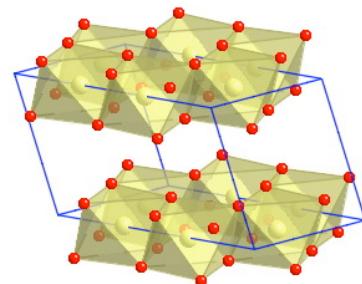
# Top experimental platforms



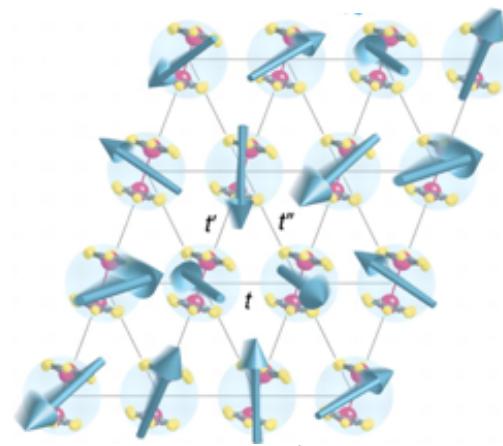
kagomé



Quantum spin ice



Kitaev materials



organics

# A rough guide to experiments on QSLs

## Does it order?

- NMR line splitting
- muSR oscillation
- thermodynamic transition via specific heat, susceptibility
- Bragg peak in neutron/ x-ray

## Delocalized excitations?

- thermal conductivity
- INS

## Is there a gap?

- Specific heat
- NMR  $1/T_1$
- Dynamic susceptibility
- T-dependence of  $\chi$

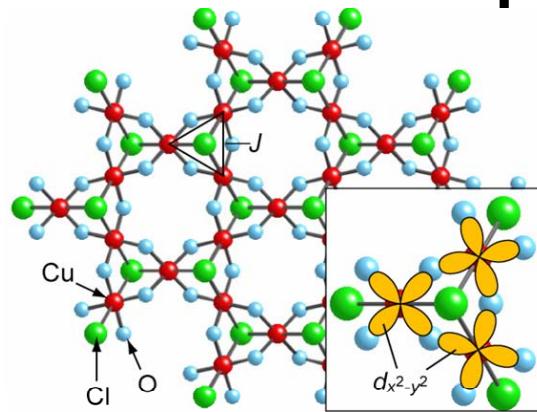
## Exotica

- Local measurements
- thermal Hall
- ARPES (on insulator!)
- Proximity effects

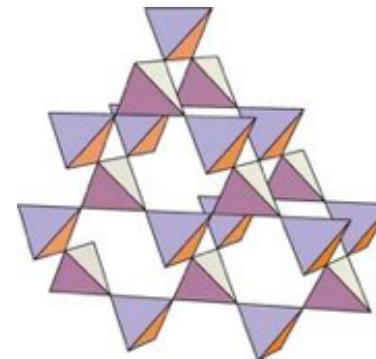
## Structure of excitations?

- $E(k)$  from INS, RIXS
- optics, Raman

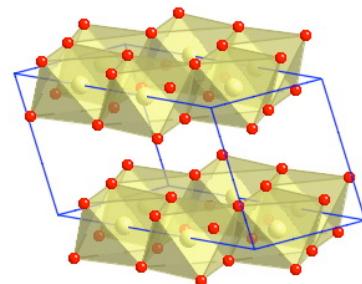
# Top experimental platforms



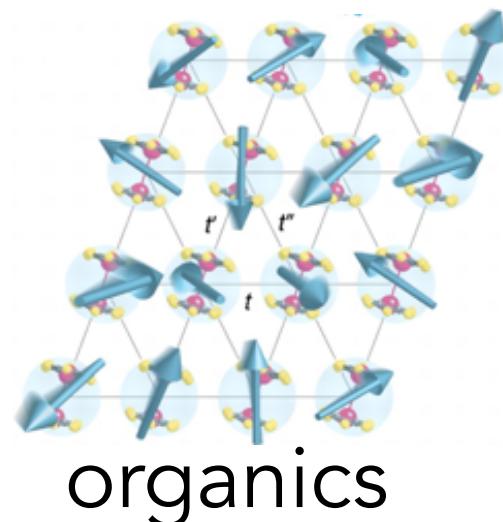
kagomé



Quantum spin ice

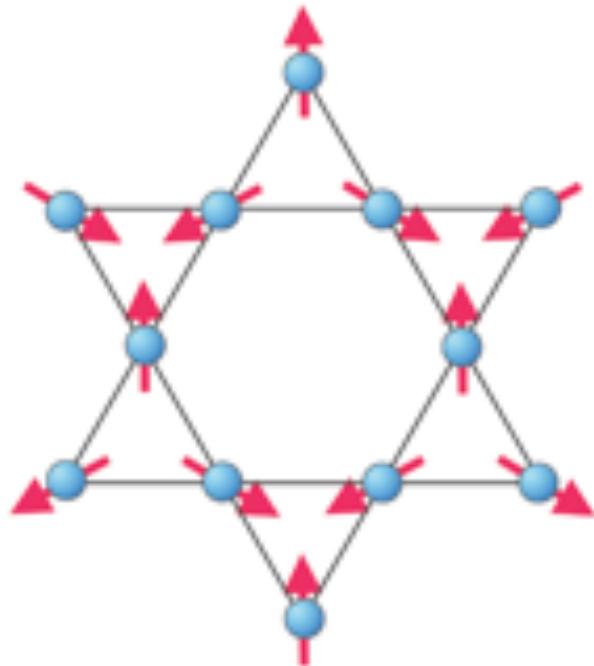


Kitaev materials



organics

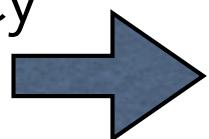
# Kagomé antiferromagnet



$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \dots$$

Very large classical degeneracy

Small  $z=4$ ,  $x=1/2$

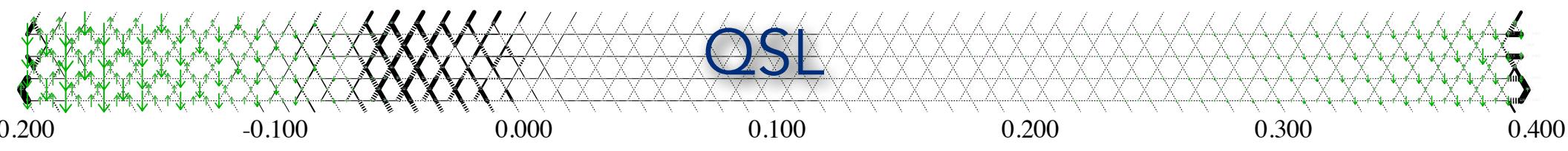


likely to be a QSL

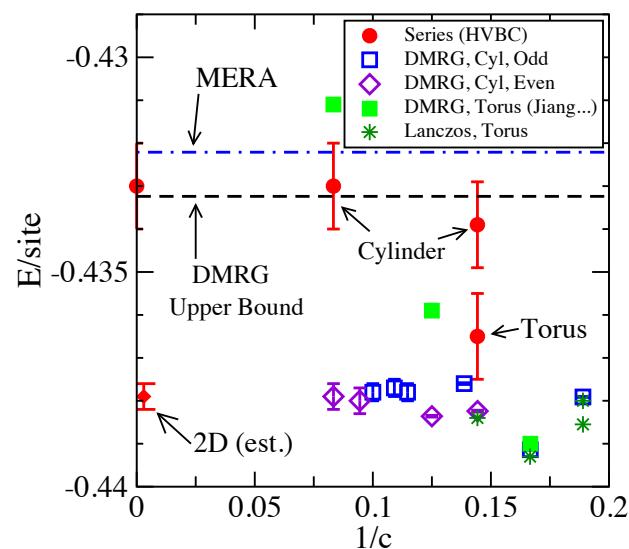
V. Elser, 1989 + many many others

# $S=1/2$ kagomé AF

- Rather definitive evidence for QSL by DMRG



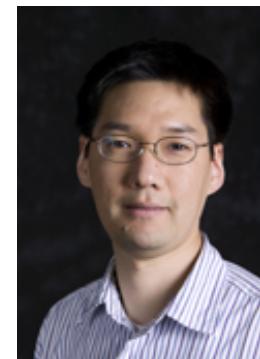
© Steve White



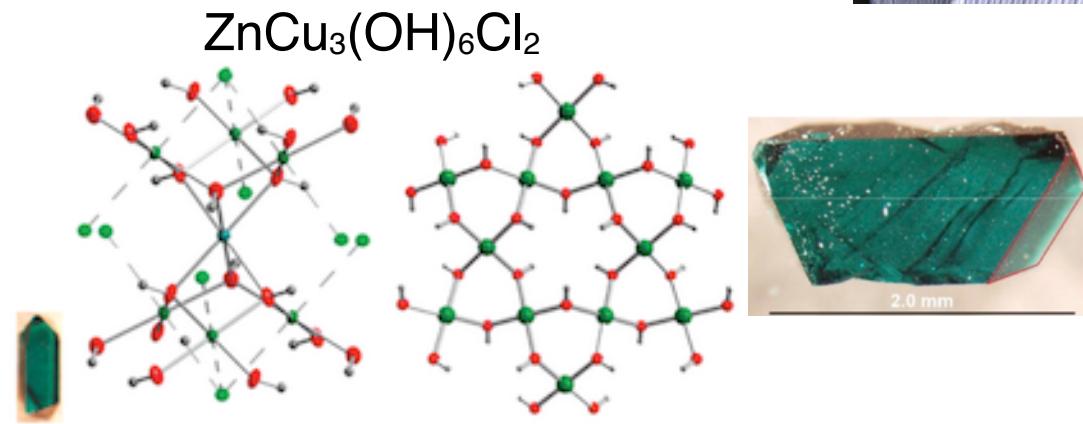
S. Yan *et al*, 2010

many other studies support  
existence of some QSL phase

# Herbertsmithite

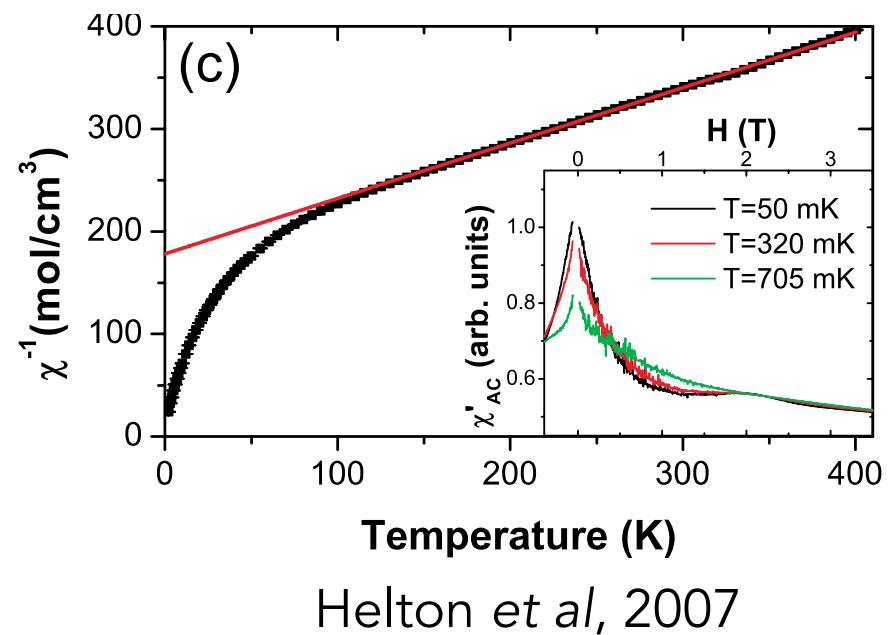


kagomé layers of Cu  
S=1/2 spins, separated  
by non-magnetic Zn



Heisenberg-like  
with  $J \sim 200\text{K}$

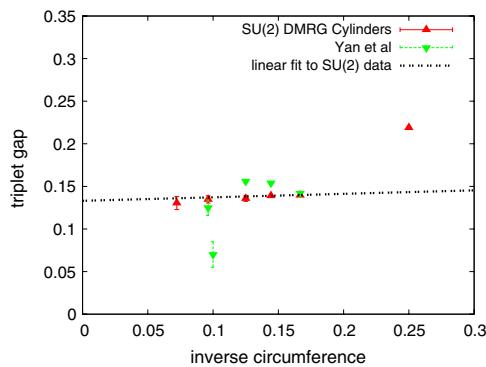
no order down to  
50mK



Helton *et al*, 2007

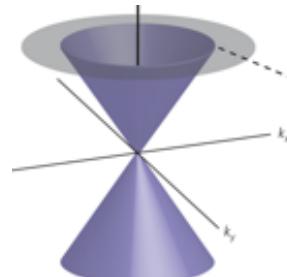
# Theory

- What kind of QSL?



S. Depenbrock *et al*, 2012

gapped,  
topological QSL



Y. Ran *et al*, 2007  
F. Becca...

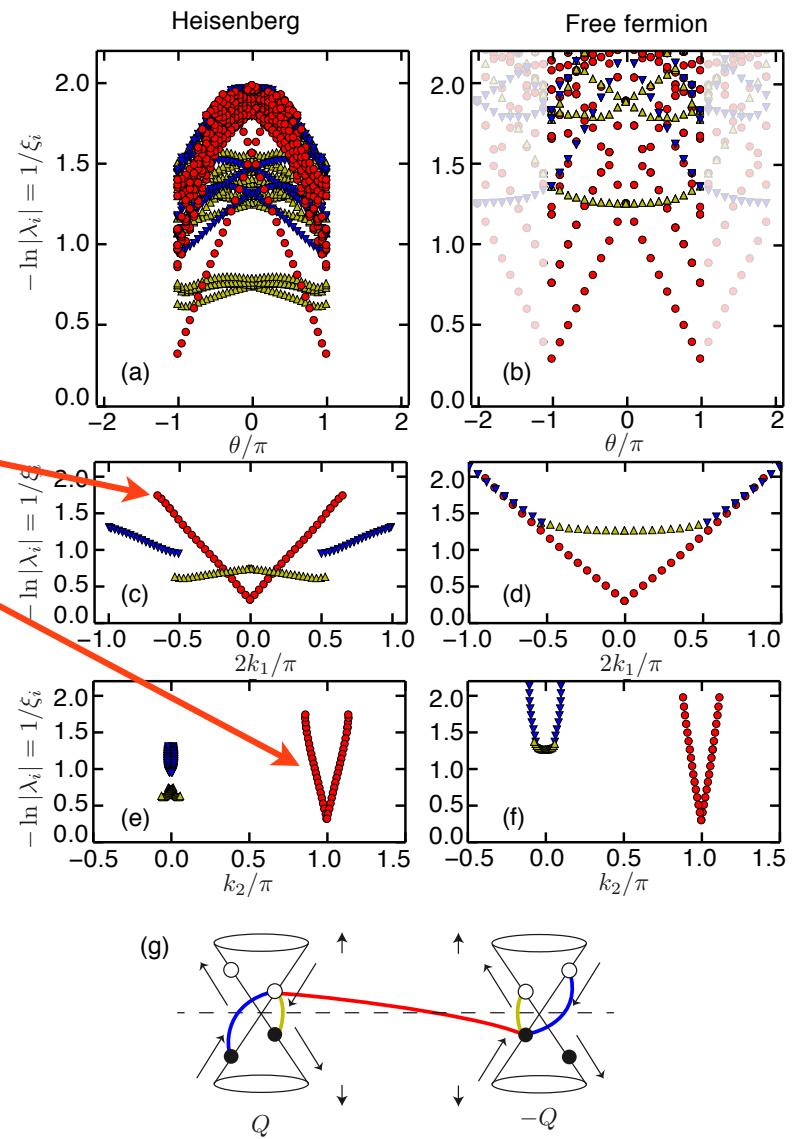
gapless  
Dirac QSL



+ various other  
proposals with  
weaker  
quantitative  
support

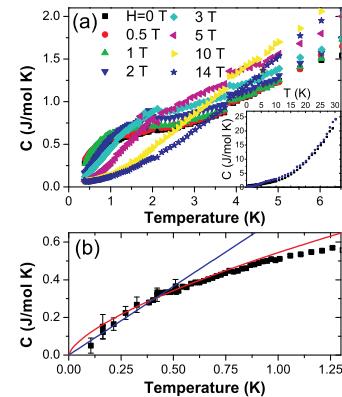
# DMRG (2016)

Y.-C. He et al:  
evidence for  
Dirac QSL

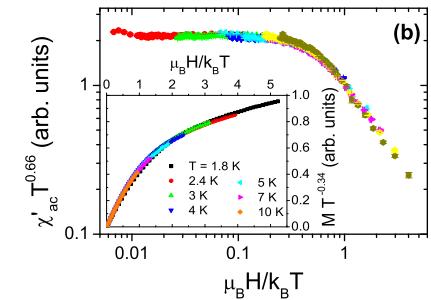


# Herbertsmithite

Lots of early evidence  
for gaplessness



Helton et al, 2007

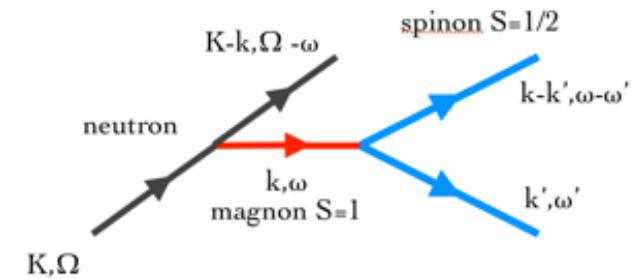
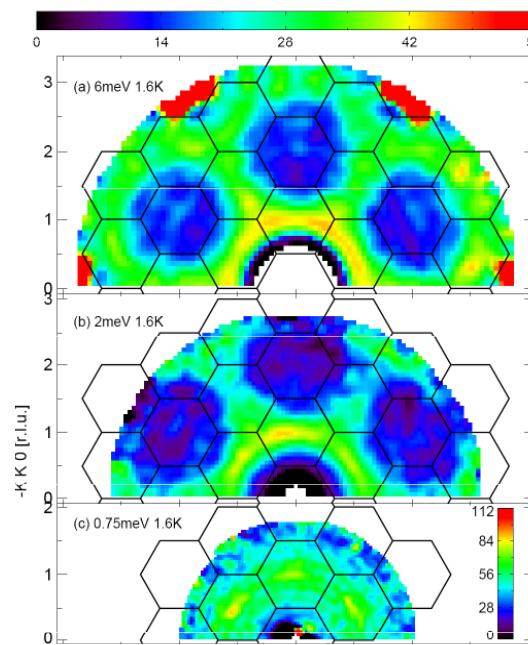


Helton et al, 2010

Single crystal INS

smooth continuum  
scattering

T-H Han et al, 2012



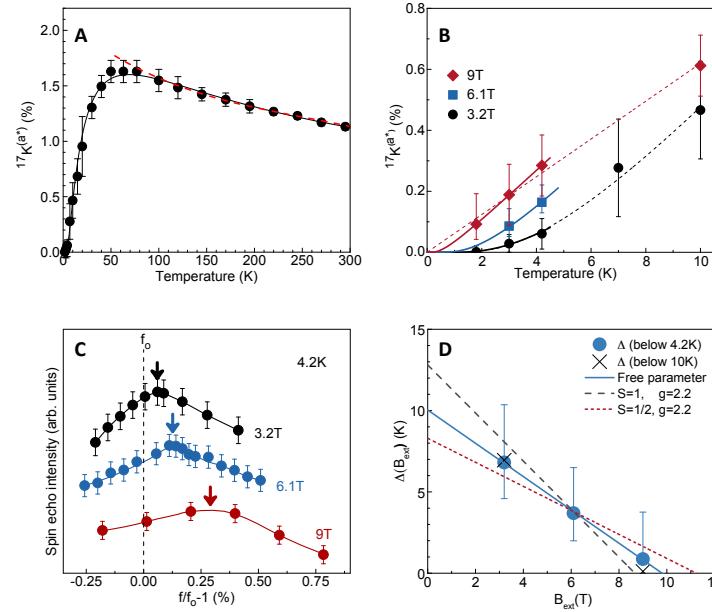
continuum scattering  
expected  
...but probably with more  
structure?

# Herbertsmithite

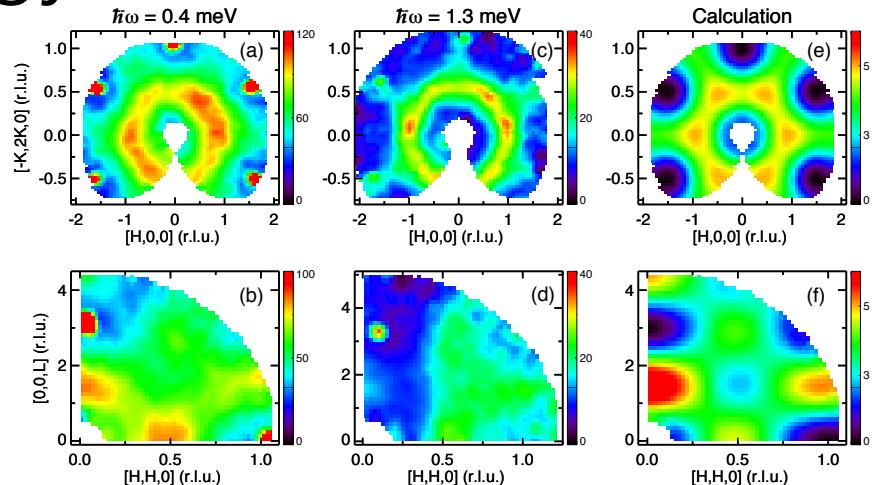
## Single crystal NMR

M. Fu *et al*, 2015

McMaster



## Low energy INS

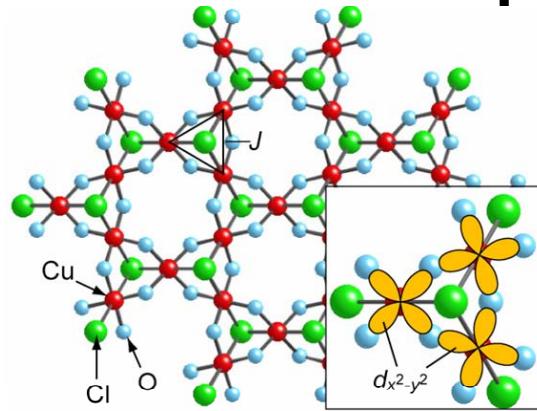


T-H Han *et al*, 2015

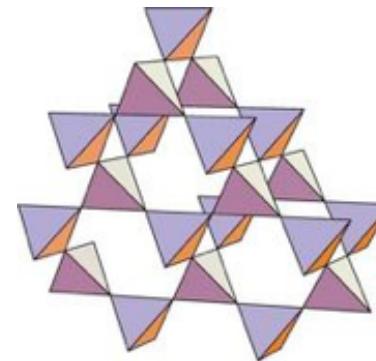
estimate gap  $\sim$   
10K

claim to separate  
impurity signal  
below 0.7meV

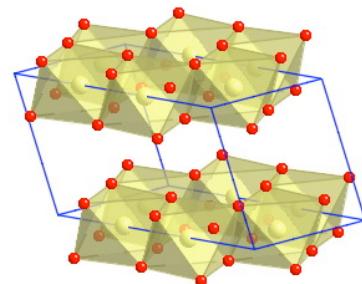
# Top experimental platforms



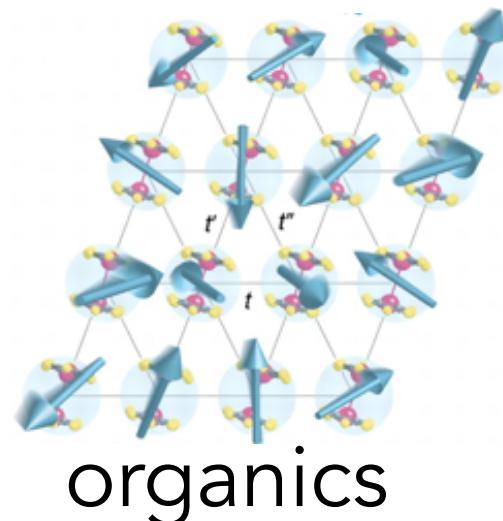
kagomé



Quantum spin ice



Kitaev materials



organics

# Kitaev model



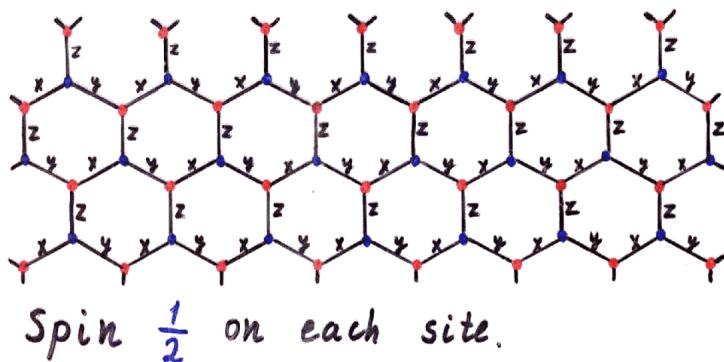
Kitaev's honeycomb model

c.f. Kitaev, Annals of Physics, 2006

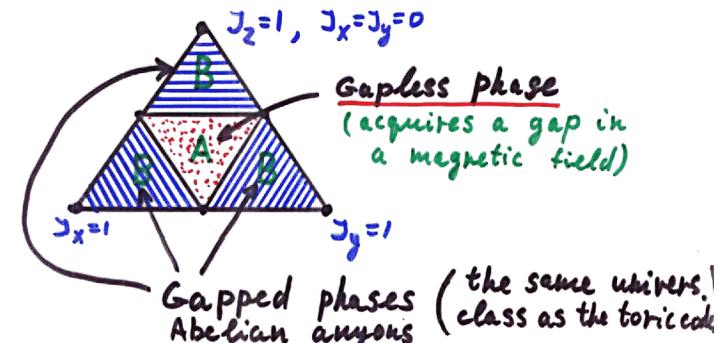
$$H = \sum_{i,\mu} K_\mu \sigma_i^\mu \sigma_{i+\mu}^\mu$$

KITP, 2003

## 1. The model

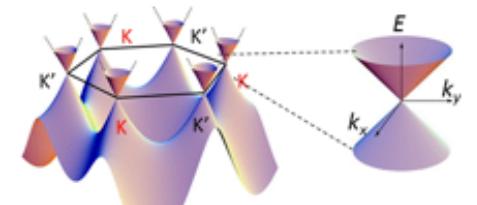


## Phase diagram



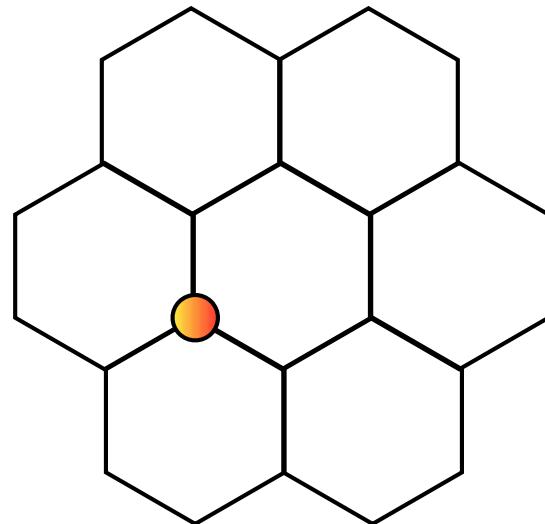
exact parton construction  $\sigma_i^\mu = i c_i c_i^\mu \quad c_i c_i^x c_i^y c_i^z = 1$

physical Majoranas  $H_m = K \sum_{\langle ij \rangle} i c_i c_j$

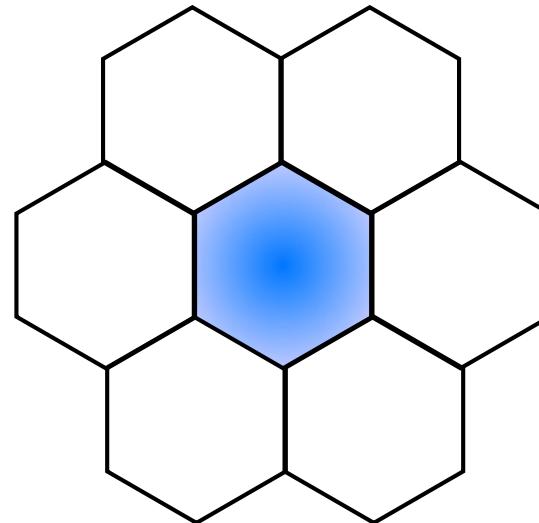


No S=0 singlets, but highly entangled.

# Non-local excitations



Majorana  $\epsilon$



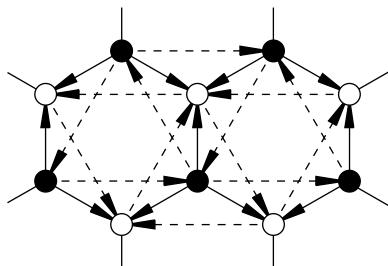
Flux  $e, m$

In Kitaev's model:

- Majorana's dispersion  $\sim K$  and Dirac-like
- Fluxes are localized and gapped

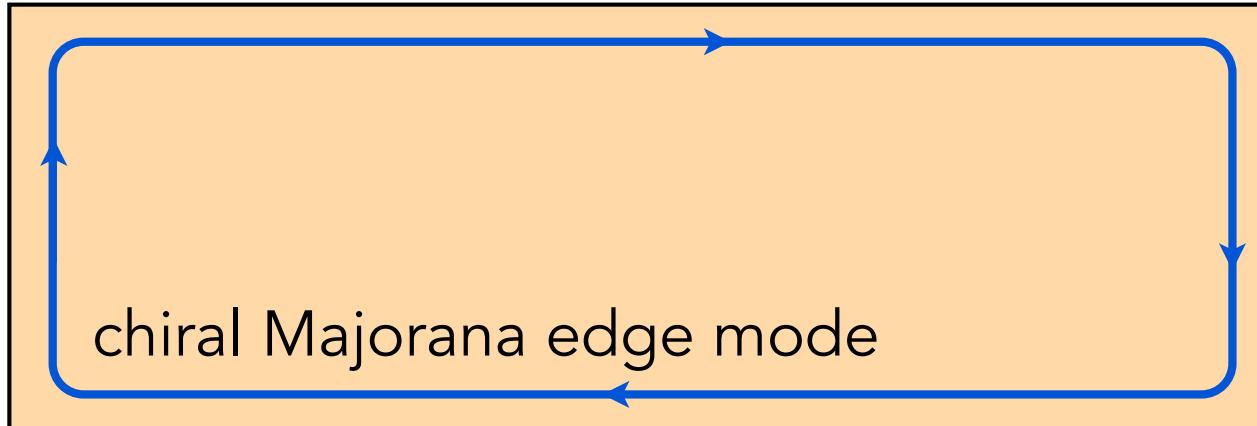
# Non-Abelian Phase

- In an applied magnetic field, the Majoranas acquire a gap



$$H_{\text{eff}} = \frac{i}{4} \sum_{j,k} A_{jk} c_j c_k,$$
$$A = 2J(\text{---}) + 2\kappa(\text{---}),$$
$$\kappa \sim \frac{h_x h_y h_z}{J^2}.$$

field induces a fermion mass, very similar to the Haldane model (except Majorana)



$$H_e = -\frac{iv}{4} \int dx \eta \partial_x \eta$$

# Quantum Hall Effect?

- No charge. Have to study heat transport!

T



$$I = \int_0^\infty \frac{dq}{2\pi} v^2 q f(vq) = \frac{c\pi k_B^2}{12\hbar} T^2$$

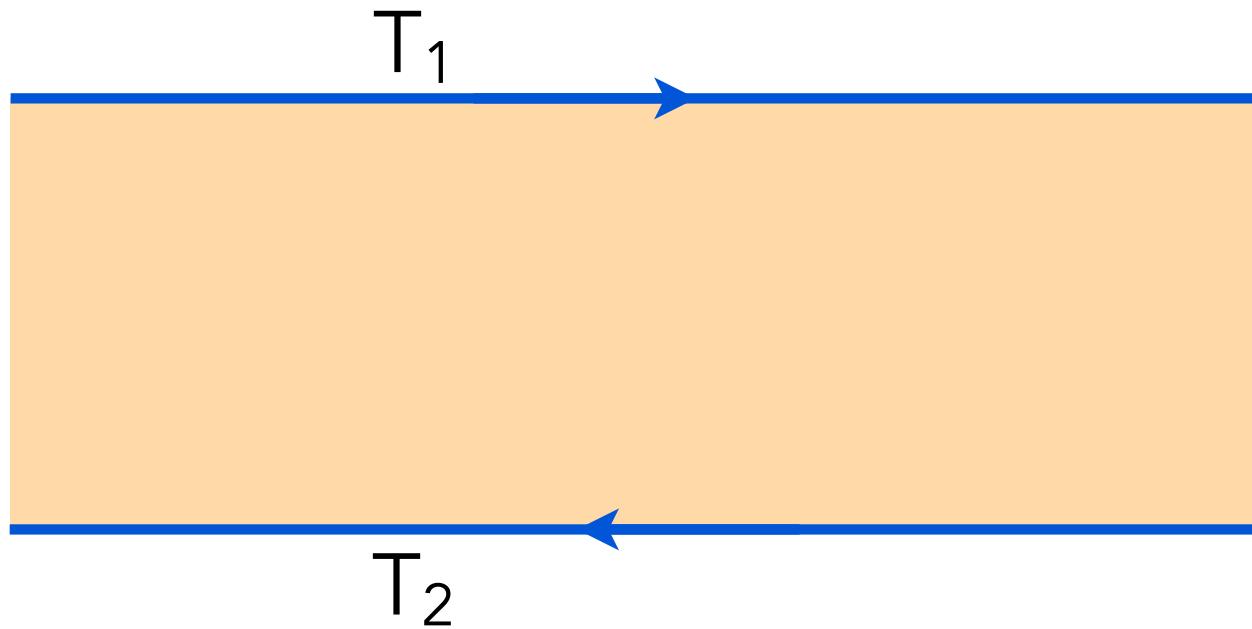
central charge  $c=1/2$

c.f.  $c=1$  for both IQHE and FQHE abelian states

implies the existence of bulk non-abelian excitations (the fluxes, bound to MZMs)

# Quantum Hall Effect?

- No charge. Have to study heat transport!

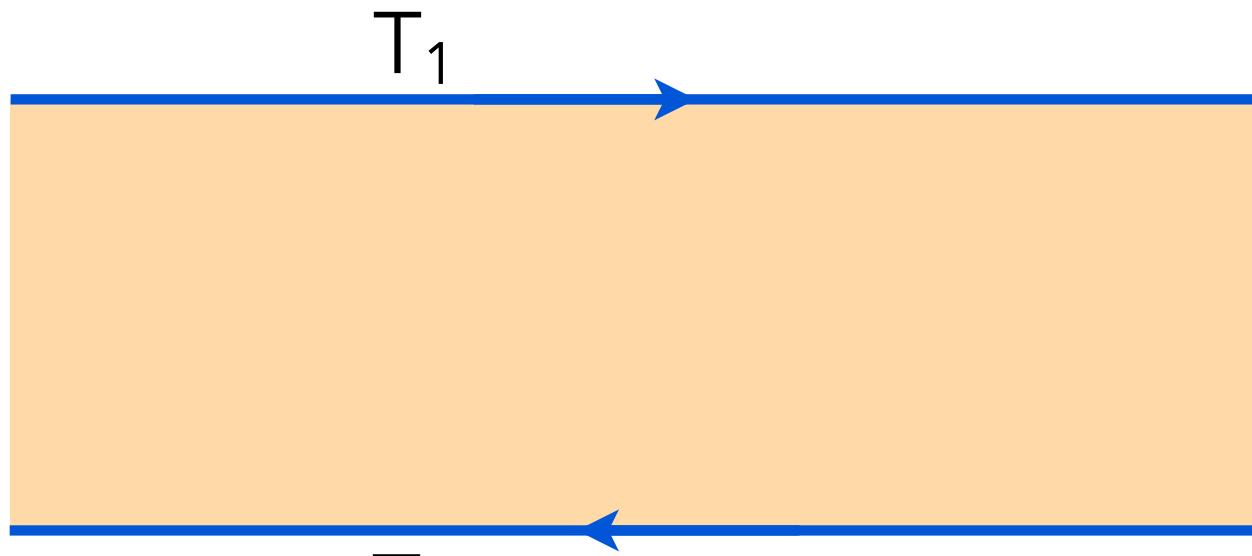


$$I = \frac{c\pi k_B^2}{12\hbar} (T_1^2 - T_2^2)$$

$$\approx \frac{c\pi k_B^2 T}{6\hbar} (T_1 - T_2)$$

# Quantum Hall Effect?

- No charge. Have to study heat transport!



$$I_x = \kappa_H \Delta T_y$$

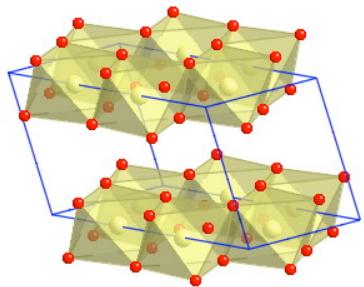
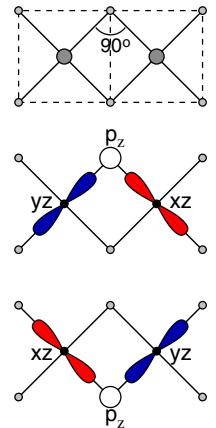
$$\kappa_H = \frac{\pi c k_B^2 T}{6 \hbar}$$

a universal prediction for chiral  
“Ising anyon” phase: agnostic to  
microscopic spin interactions

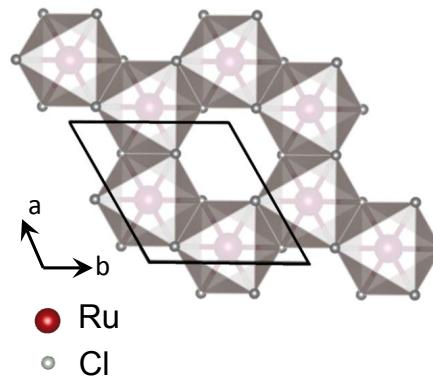
# Kitaev Materials

Jackeli, Khaliullin  
2009

Showed that Kitaev interaction can be large in edge-sharing octahedra with large spin-orbit-coupling



$\text{Na}_2\text{IrO}_3$ ,  
 $(\alpha, \beta, \gamma)$ -  
 $\text{Li}_2\text{IrO}_3$

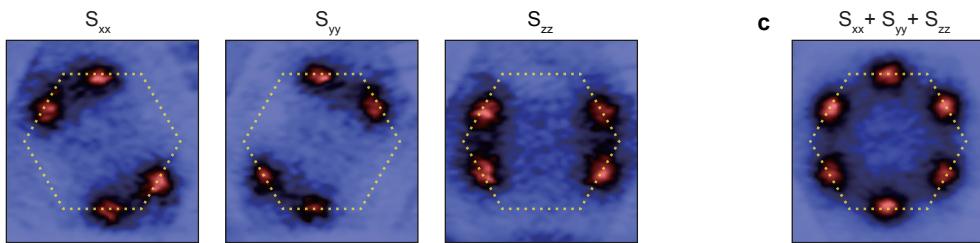


$\alpha\text{-RuCl}_3$

Y.-J. Kim...

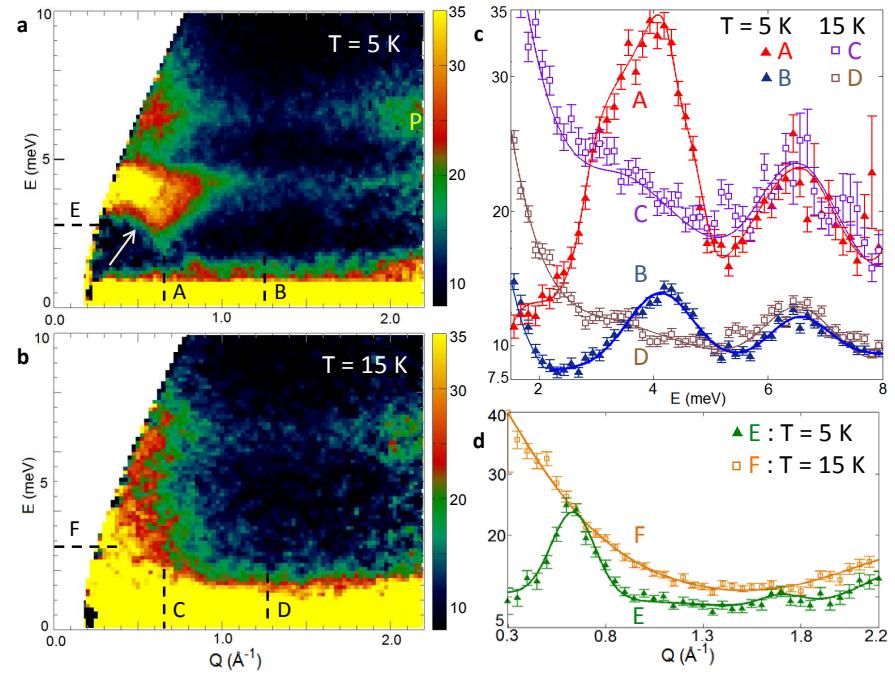
Honeycomb and hyper-honeycomb structures

# Kitaev Materials



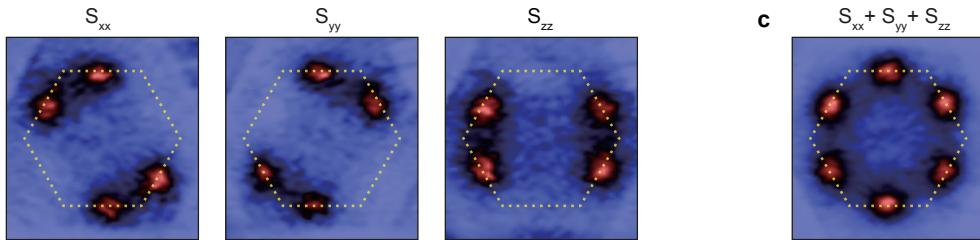
direct evidence for  
direction-dependent  
anisotropic exchange  
from diffuse magnetic  
x-ray scattering in  
 $\text{Na}_2\text{IrO}_3$  (BJ Kim group)

there is pretty strong evidence  
of substantial Kitaev exchange  
in quite a few materials



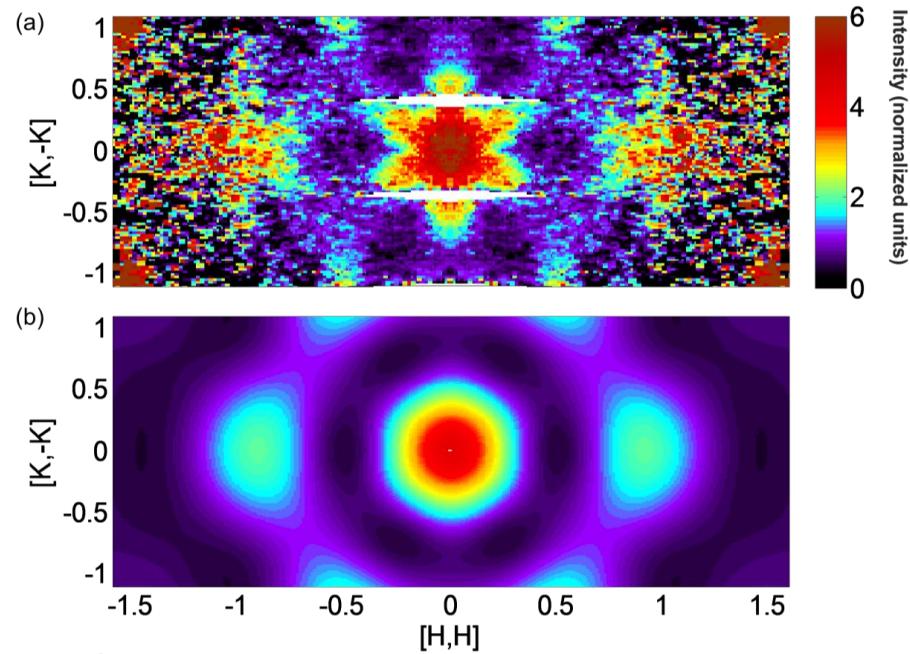
Observation of gapped  
continuum mode persisting  
above  $T_N$  in  $\alpha\text{-RuCl}_3$   
consistent with Majoranas  
(A. Banerjee *et al*)

# Kitaev Materials



direct evidence for  
direction-dependent  
anisotropic exchange  
from diffuse magnetic  
x-ray scattering in  
 $\text{Na}_2\text{IrO}_3$  (BJ Kim group)

there is pretty strong evidence  
of substantial Kitaev exchange  
in quite a few materials



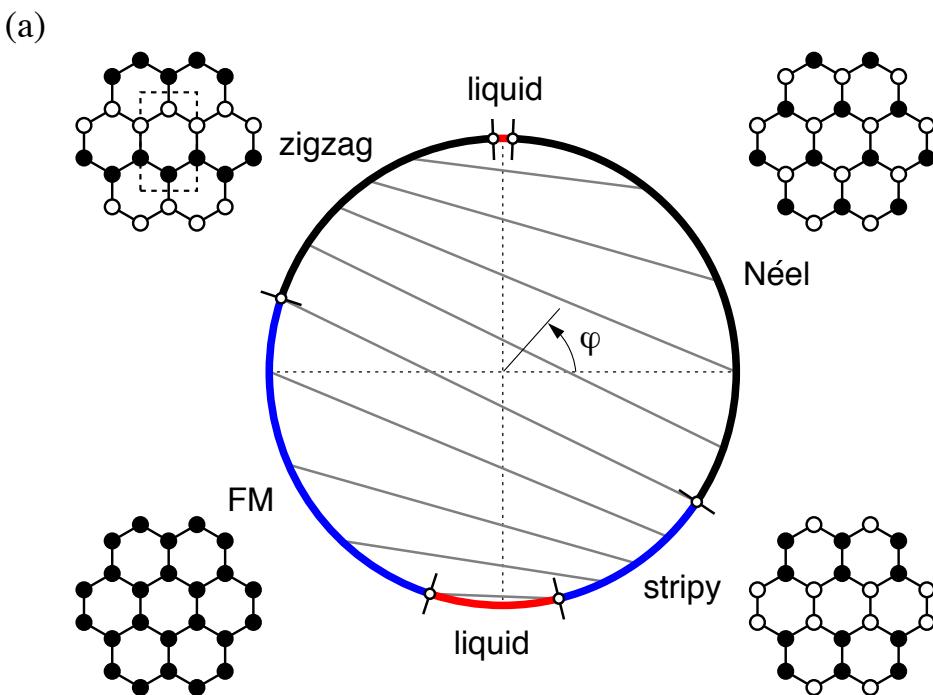
single-crystal data in  $\alpha\text{-RuCl}_3$   
compared to Kitaev's soluble  
model (A. Banerjee et al)

# Magnetism

- But...they all order so far

due to additional interactions,  
e.g. Heisenberg

$$H = \sum_{i,\alpha} K S_i^\alpha S_{i+\alpha}^\alpha + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j$$



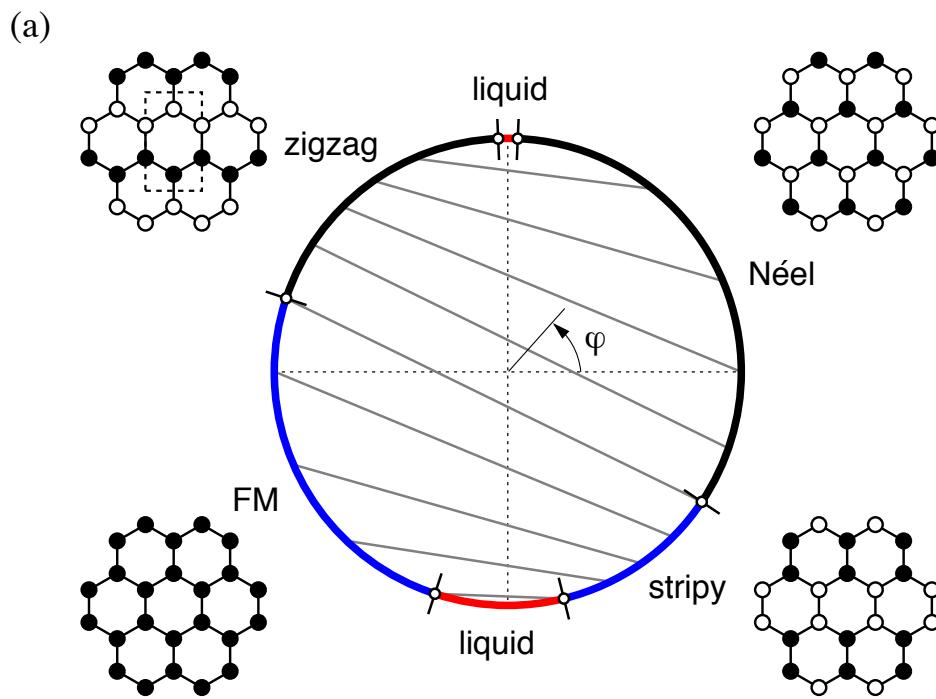
zigzag ordered state  
has been observed in  
 $\text{Na}_2\text{IrO}_3$  and  $\alpha\text{-RuCl}_3$  ;  
incommensurate order  
in  $\text{Li}_2\text{IrO}_3$

# Magnetism

- But...they all order so far

due to additional interactions,  
e.g. Heisenberg

$$H = \sum_{i,\alpha} K S_i^\alpha S_{i+\alpha}^\alpha + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j$$

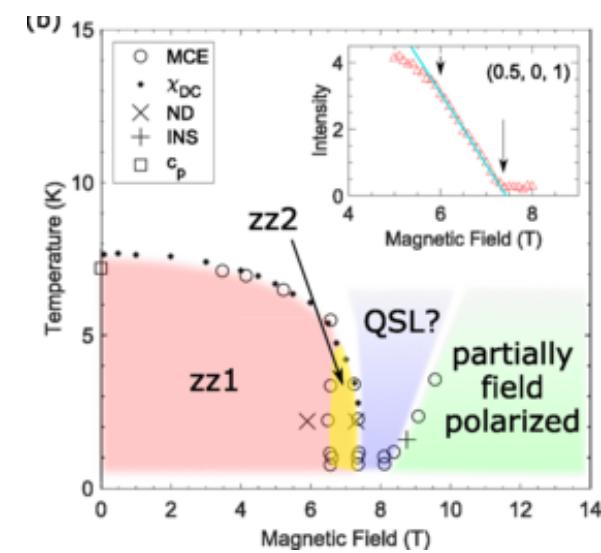


so far no QSL!

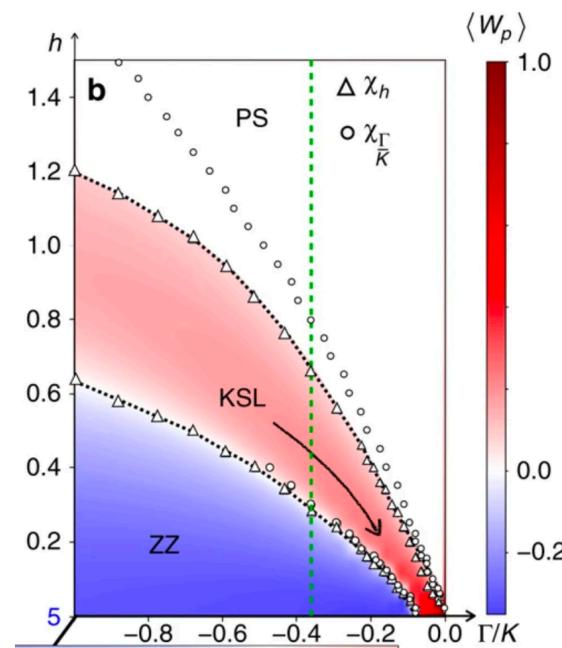


# $\alpha$ -RuCl<sub>3</sub>

Huge effort to understand field-induced paramagnetic state



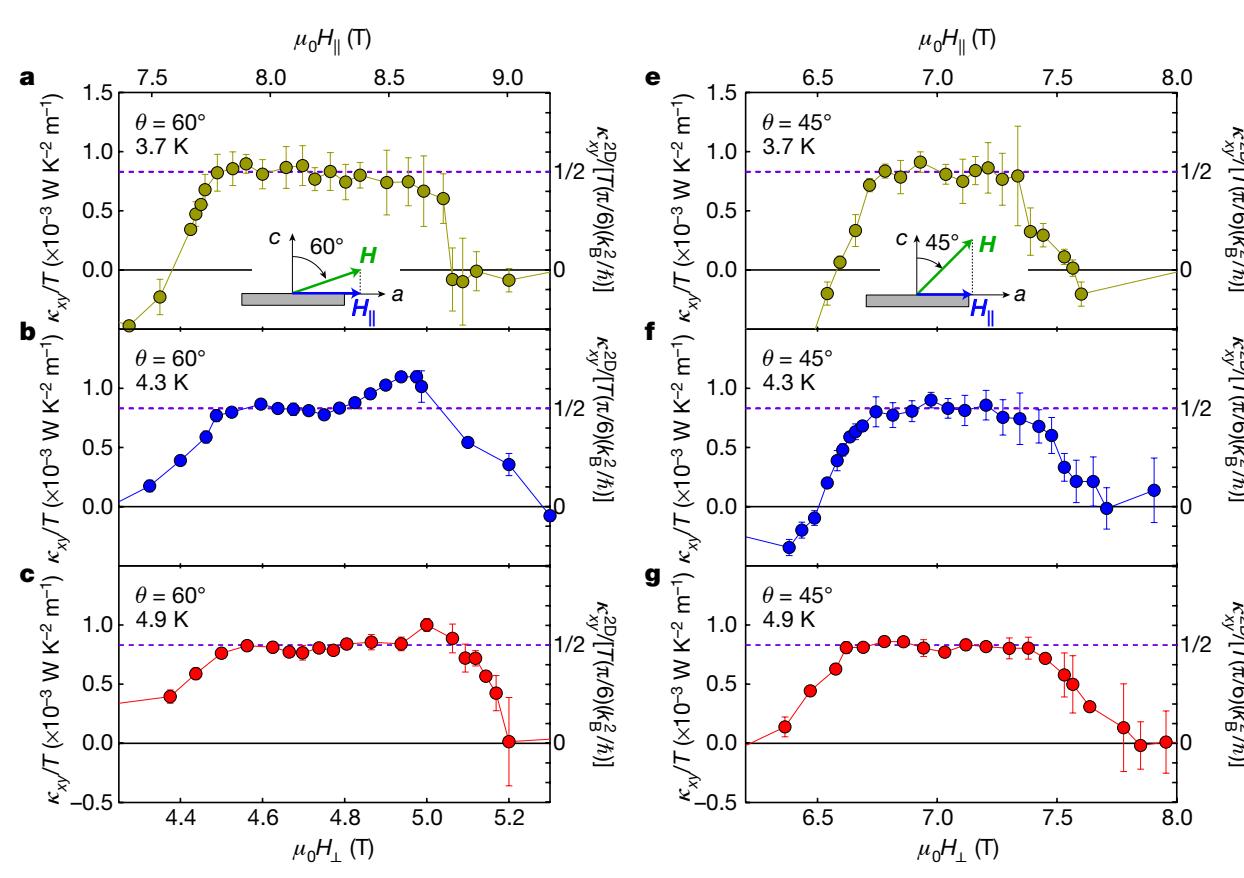
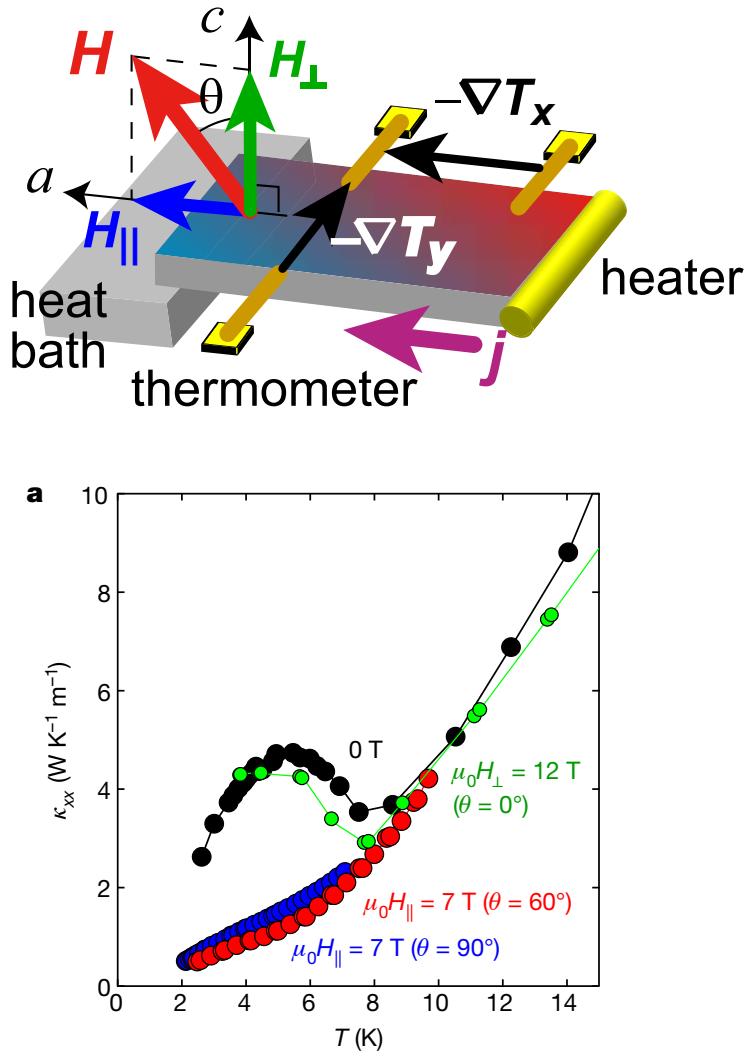
C. Balz *et al*, 2019



J. Gordon *et al*, 2019

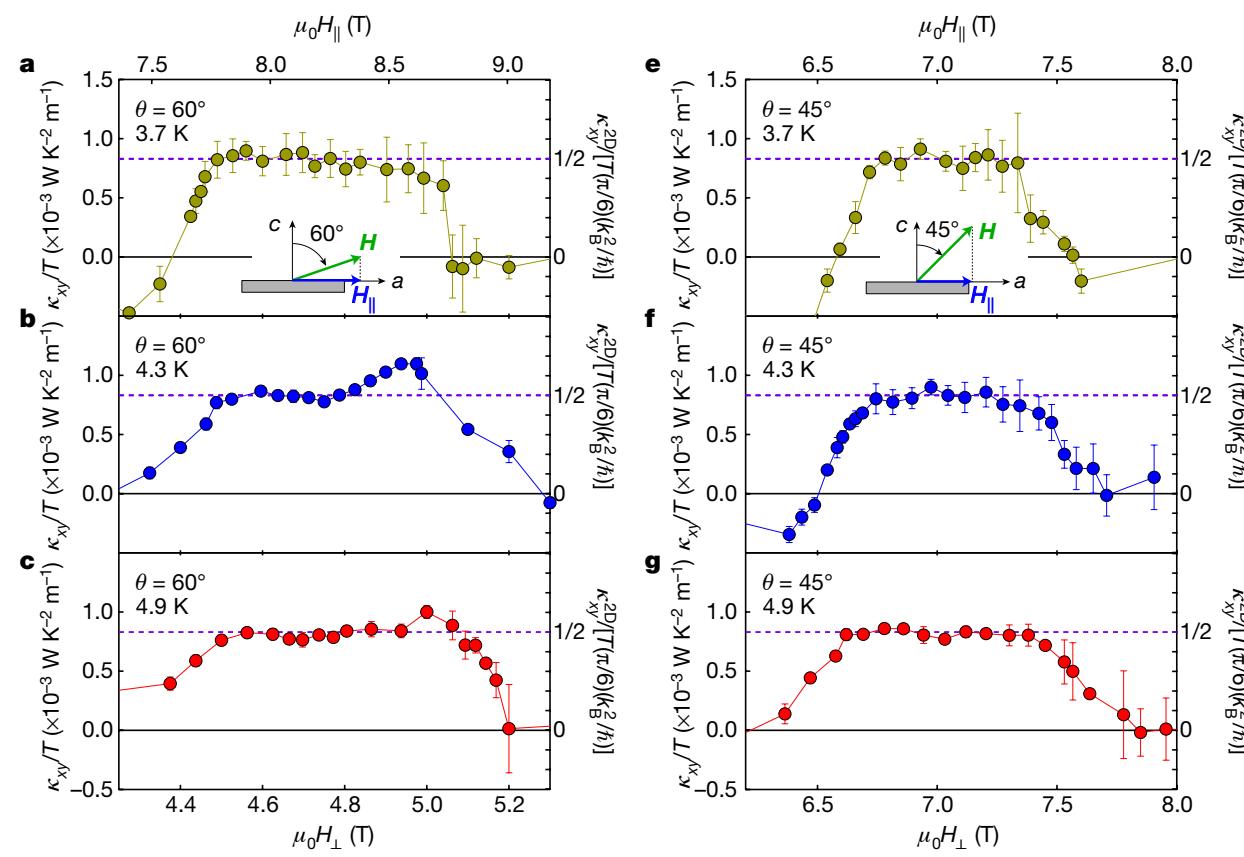
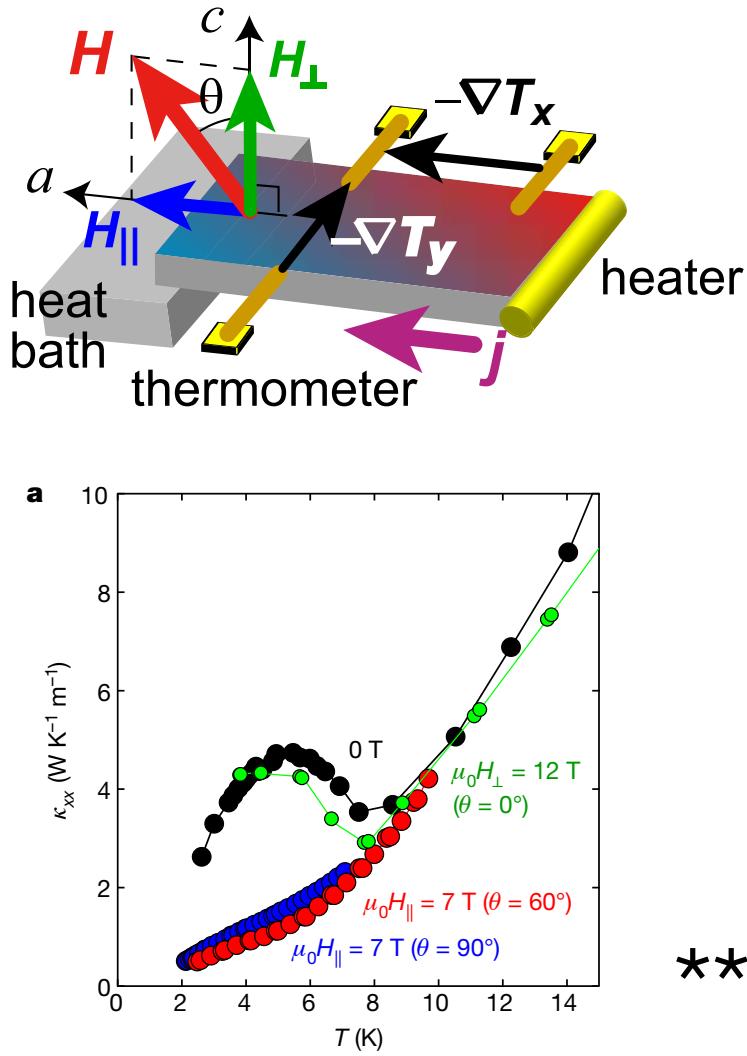
# Thermal Hall Effect?

Y. Kasahara *et al*, Nature 2018



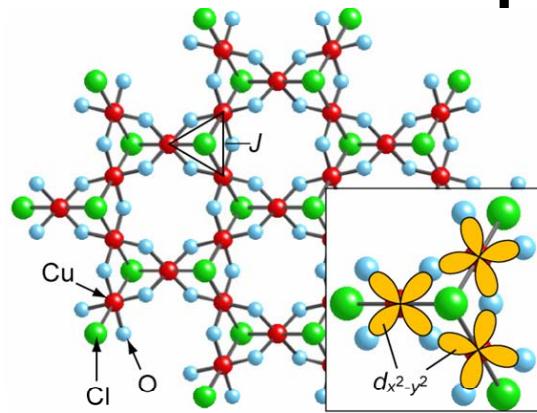
# Thermal Hall Effect?

Y. Kasahara *et al*, Nature 2018

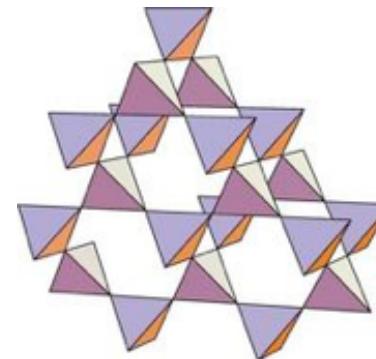


\*\* Not reproduced. c.f. Ong talk?

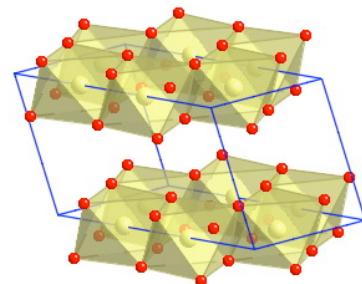
# Top experimental platforms



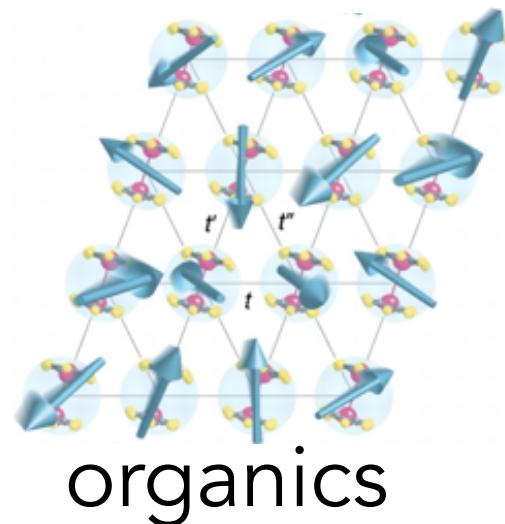
kagomé



Quantum spin ice



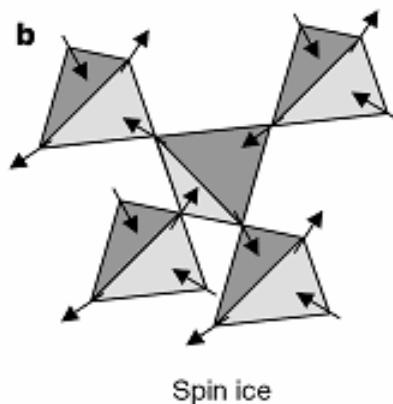
Kitaev materials



organics

# Spin ice

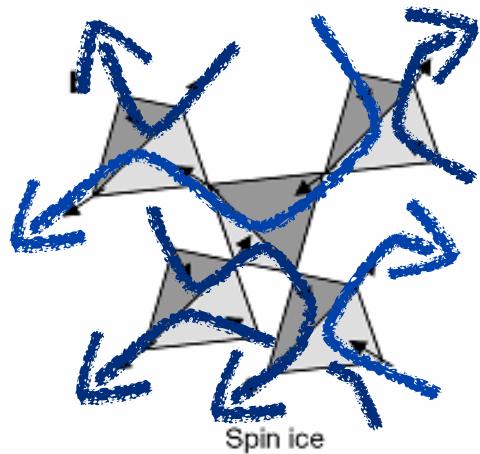
- Spins in  $\text{Ho}_2\text{Ti}_2\text{O}_7$ ,  $\text{Dy}_2\text{Ti}_2\text{O}_7$  have dominant NN Ising coupling  $J_{zz}$  enforcing classical 2in-2out “ice rules” for  $T < 1\text{K}$



thermal fluctuations  
through many  
degenerate states

# Spin ice

- Spins in  $\text{Ho}_2\text{Ti}_2\text{O}_7$ ,  $\text{Dy}_2\text{Ti}_2\text{O}_7$  have dominant NN Ising coupling  $J_{zz}$  enforcing classical 2in-2out “ice rules” for  $T < 1\text{K}$



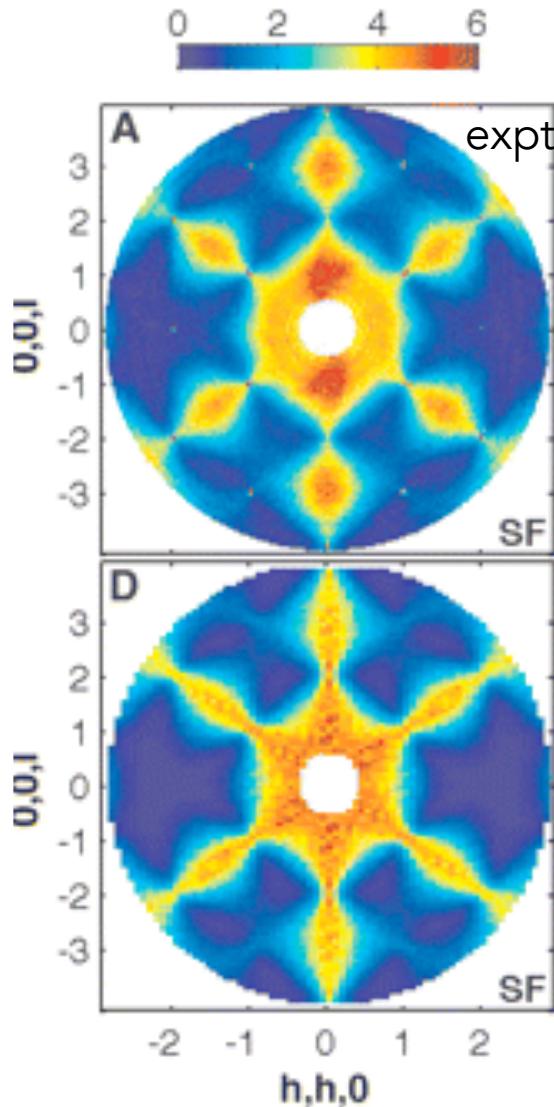
$$\vec{S} \sim \vec{b}$$

$$\vec{\nabla} \cdot \vec{b} = 0$$

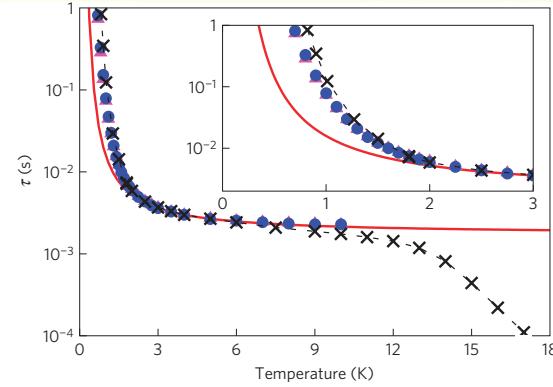
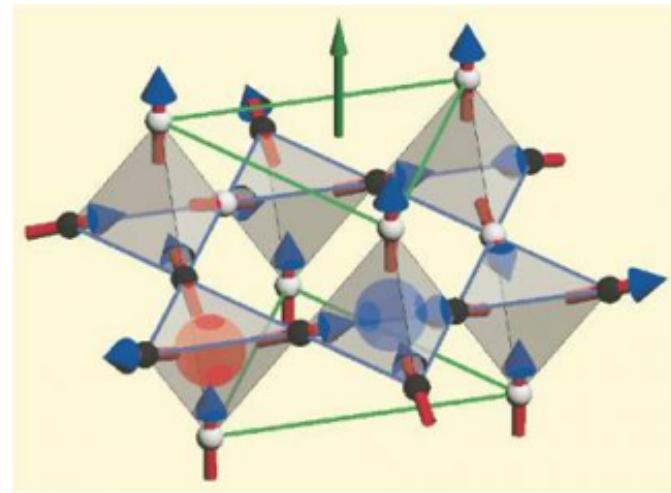
artificial magnetostatics: spins map to field lines

# Classical spin liquid

T. Fennell et al, 2009



pinch  
points  
 $\vec{\nabla} \cdot \vec{b} = 0$



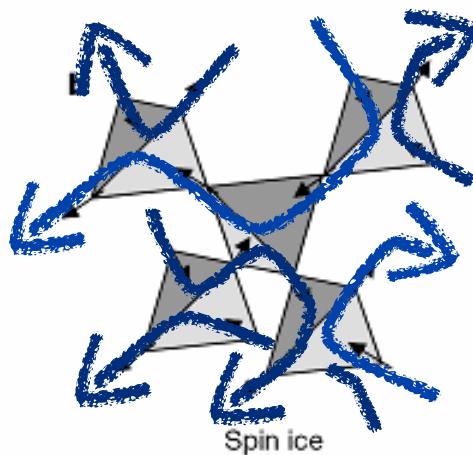
magnetic monopoles  
behave like diffusing  
ions in a polyelectrolyte

# Quantum spin ice

$$H = H_{CSI} + J_{\pm} \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.})$$

quantum dynamics creates  
superposition state

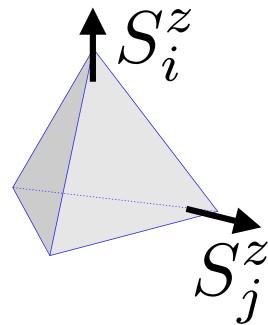
$$|\Psi\rangle = \sum_{\text{loops}}$$



QSL which “simulates”  
quantum  
electrodynamics -  
vacuum fluctuations

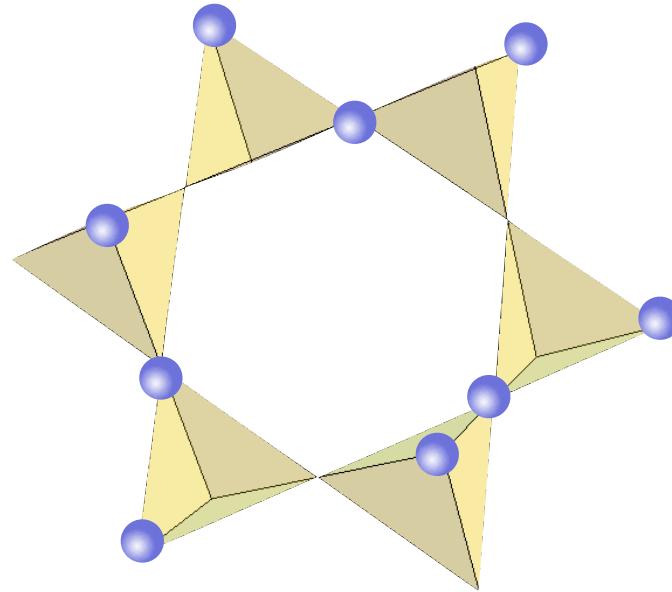
# Mapping to E+M

## 1. Degenerate perturbation theory



$$H_0 = \frac{J_{zz}}{2} \sum_t \left( \sum_{i \in t} S_i^z \right)^2$$

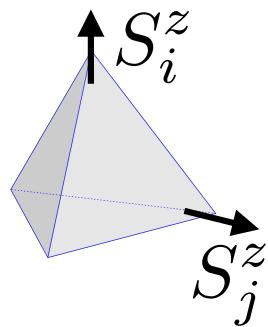
$$H_1 = J_{\pm} \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.})$$



- $S_i^z = +\frac{1}{2}$

# Mapping to E+M

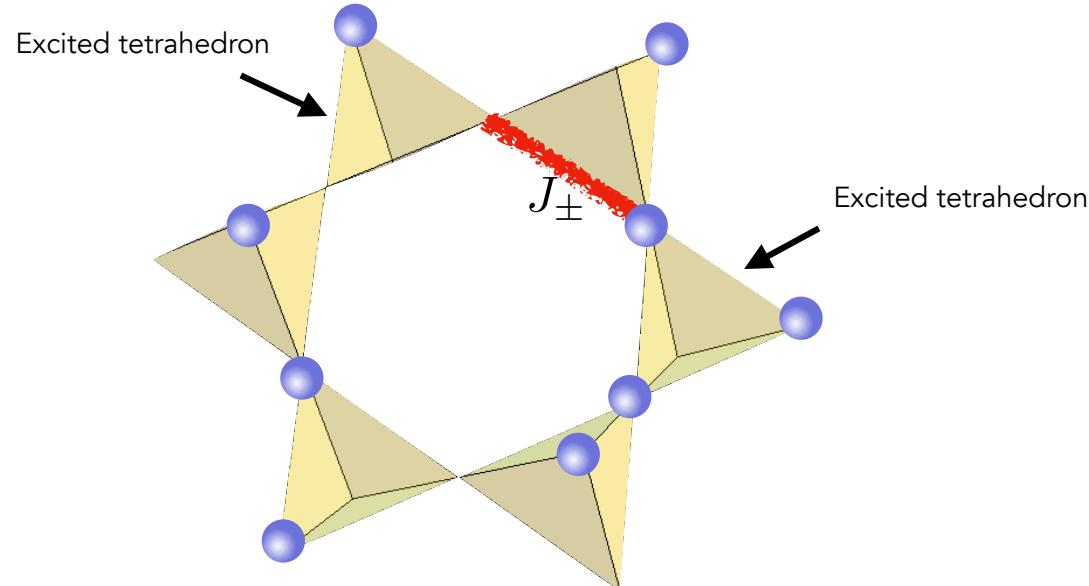
## 1. Degenerate perturbation theory



$$H_0 = \frac{J_{zz}}{2} \sum_t \left( \sum_{i \in t} S_i^z \right)^2$$

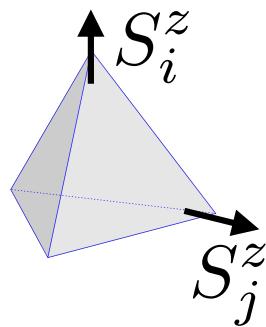
$$H_1 = J_{\pm} \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.})$$

1. Act once with  $H_1$



# Mapping to E+M

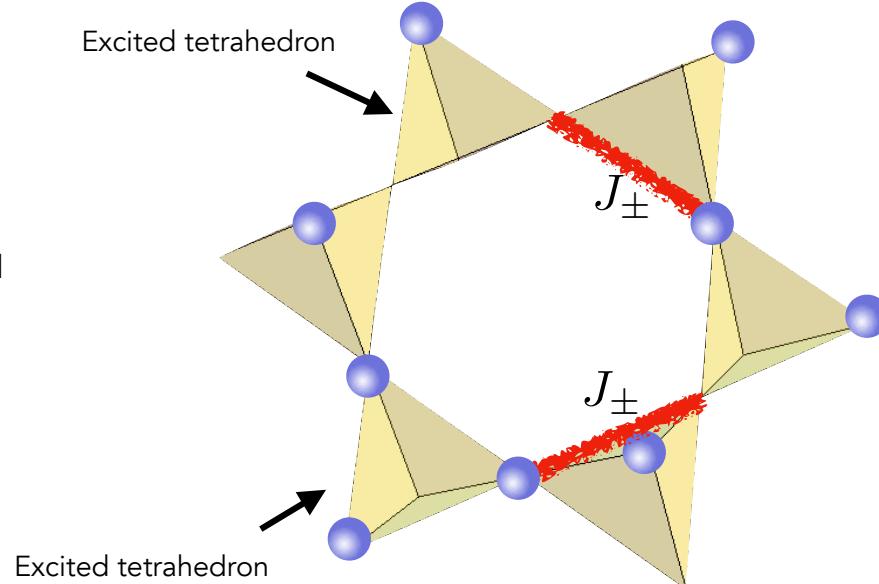
## 1. Degenerate perturbation theory



$$H_0 = \frac{J_{zz}}{2} \sum_t \left( \sum_{i \in t} S_i^z \right)^2$$

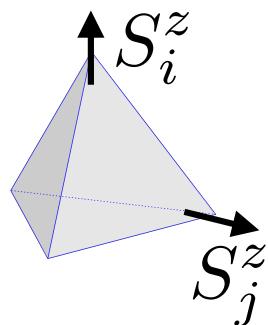
$$H_1 = J_{\pm} \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.})$$

1. Act once with  $H_1$
2. Act twice with  $H_1$



# Mapping to E+M

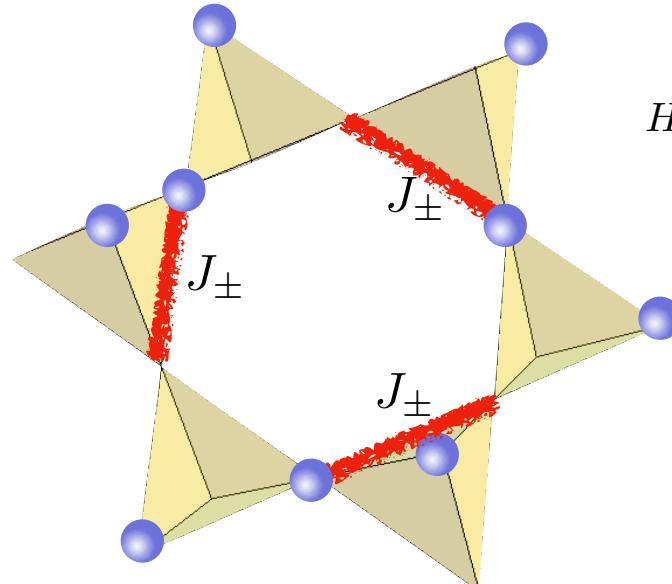
## 1. Degenerate perturbation theory



$$H_0 = \frac{J_{zz}}{2} \sum_t \left( \sum_{i \in t} S_i^z \right)^2$$

$$H_1 = J_{\pm} \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.})$$

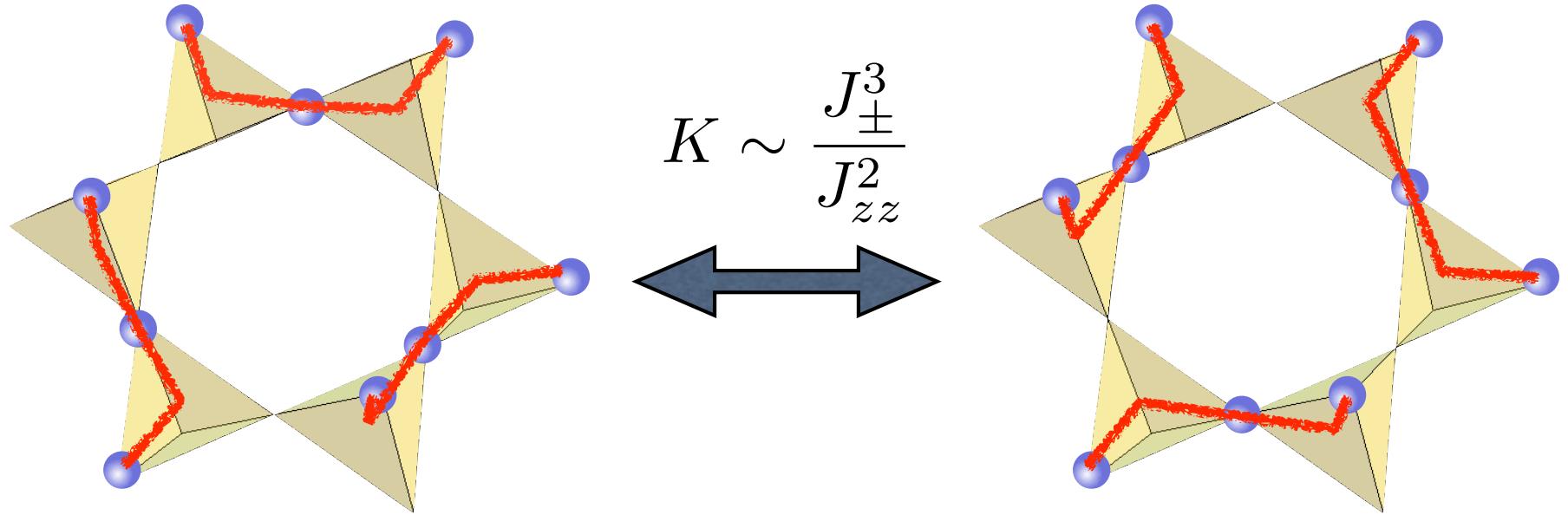
1. Act once with  $H_1$
2. Act twice with  $H_1$
3. Act thrice with  $H_1$



$$H_{\text{eff}} \sim \frac{J_{\pm}^3}{J_{zz}^2} \sum_h (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + \text{h.c.})$$

“ring exchange”

# Ring exchange

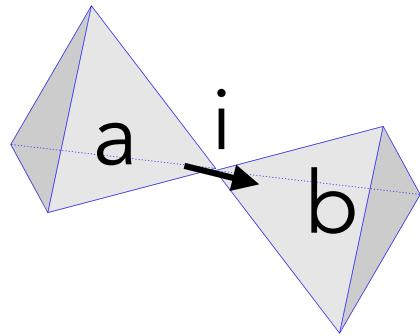


$$H_{\text{eff}} \sim \frac{J_{\pm}^3}{J_{zz}^2} \sum_h (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + \text{h.c.})$$

Tunneling reconnects field lines

# Mapping to E+M

## 2. Introduce gauge fields



$$S_i^z = E_{ab}$$

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

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

“odd” lattice **compact** U(1) gauge theory

(means  $E$  is half integer)

(means  $A_{ab}$  is a  $2\pi$  periodic phase)

# Mapping to E+M

## 3. Deconfined phase

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

$\downarrow$   
 $B^2$

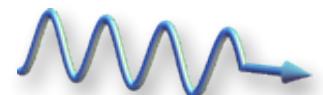
$$\approx U \sum_{\langle ab \rangle} \left( E_{ab}^2 - \frac{1}{4} \right) + \frac{K}{2} \sum_h (\nabla \times A)^2$$

Based on extensive study of lattice gauge theory, numerics, this is a qualitatively good approximation (gauge fields are the “right” choice of variables)

The rest is textbook E+M

# Excitations

- fully coherent propagating monopoles  
= “spinons” (charges in gauge theory)
- dual monopoles (dual charges)
- artificial photon: a gapless protected collective excitation which is *not* a Goldstone mode

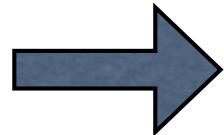


# Artificial photon

- gapless, linear, *non-Goldstone* mode

Mapping to EM:  $S_i^z \sim \mathbf{E} \cdot \hat{n}_i$

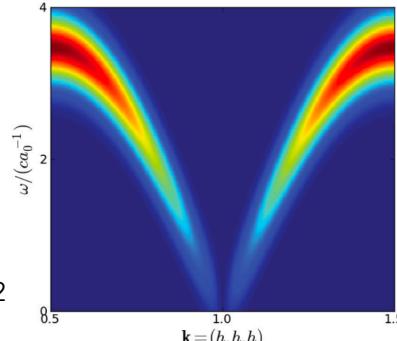
Quantization of SHO:  $E_\mu \sim \frac{1}{\sqrt{V}} \sum_{k,\sigma} i\sqrt{k} (a_{k\sigma} e^{ik \cdot r} - a_{k\sigma}^\dagger e^{-ik \cdot r}) \epsilon_{\mu\sigma}(k)$



$$\langle S_k^z S_{-k}^z \rangle \sim |k| \delta(\omega - v|k|)$$

K.Ross *et al*, 2011

- Linearly dispersion mode at Bragg point but vanishing weight at low energy
- Completely robust to anisotropy, magnetic field, etc: does not arise from breaking any physical symmetry



Plot from O. Benton *et al*, 2012

Not yet observed -  
challenge is narrow  
bandwidth due to small  
exchange in candidate  
materials

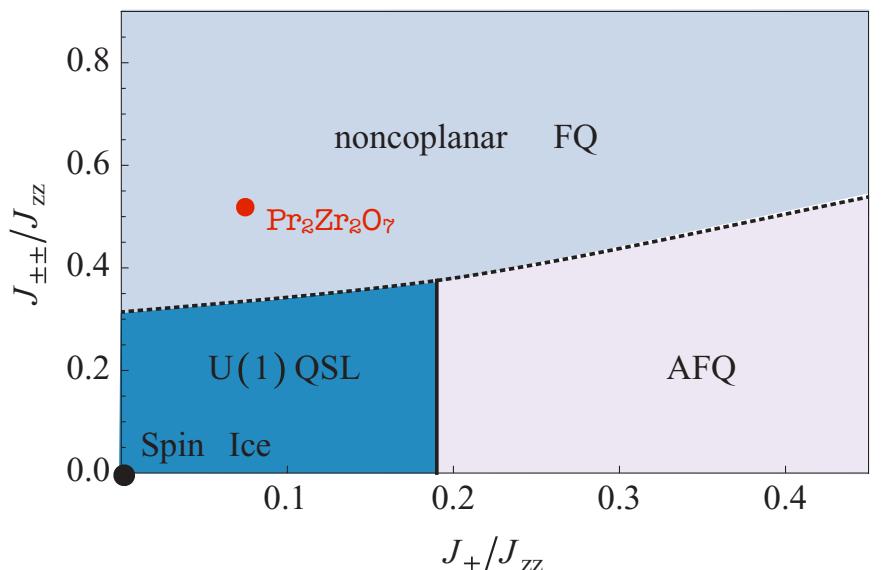
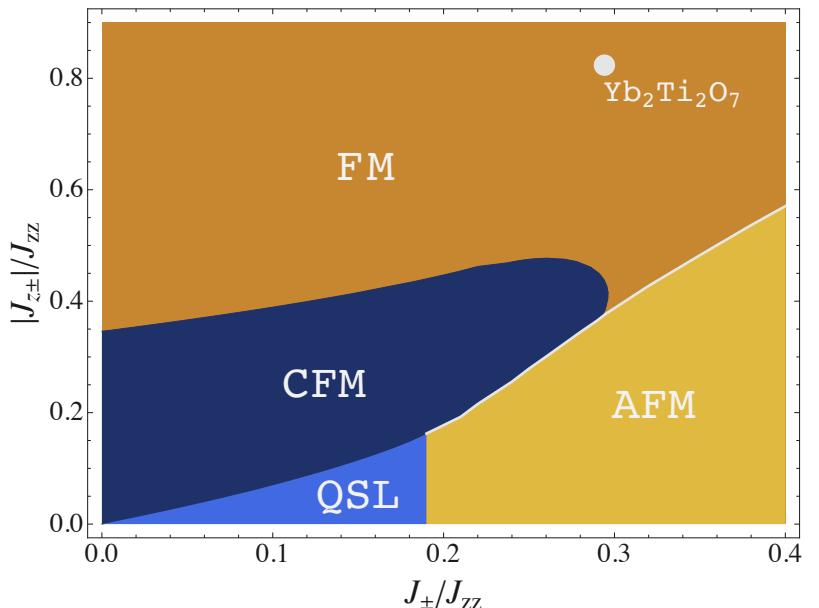
# Quantum spin ice

Realistic theory for  
quantum rare earth  
pyrochlores

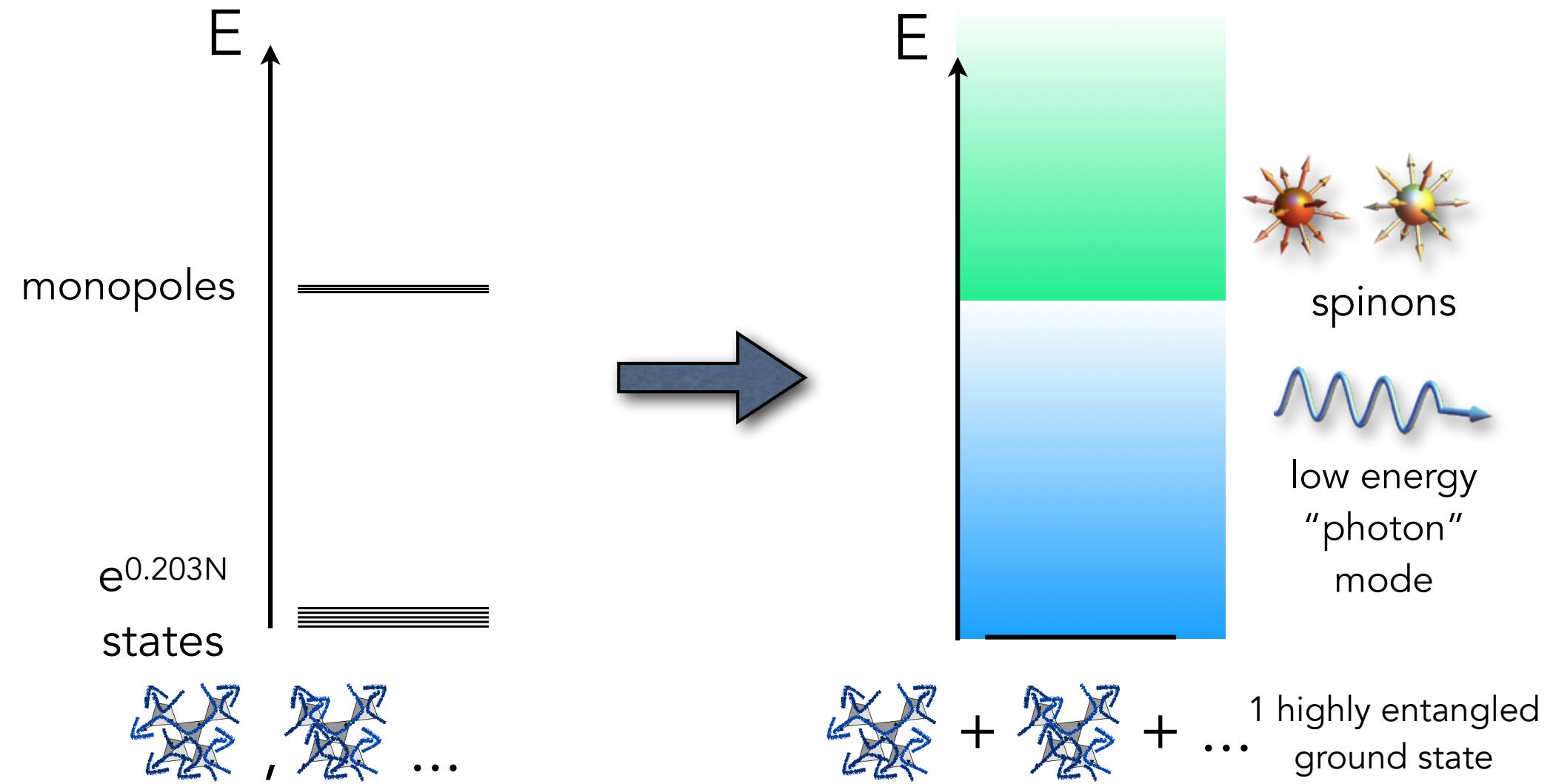
L. Savary + LB, 2012

S.B. Lee, S. Onoda + LB, 2012  
+ Many subsequent numerical  
and analytical works

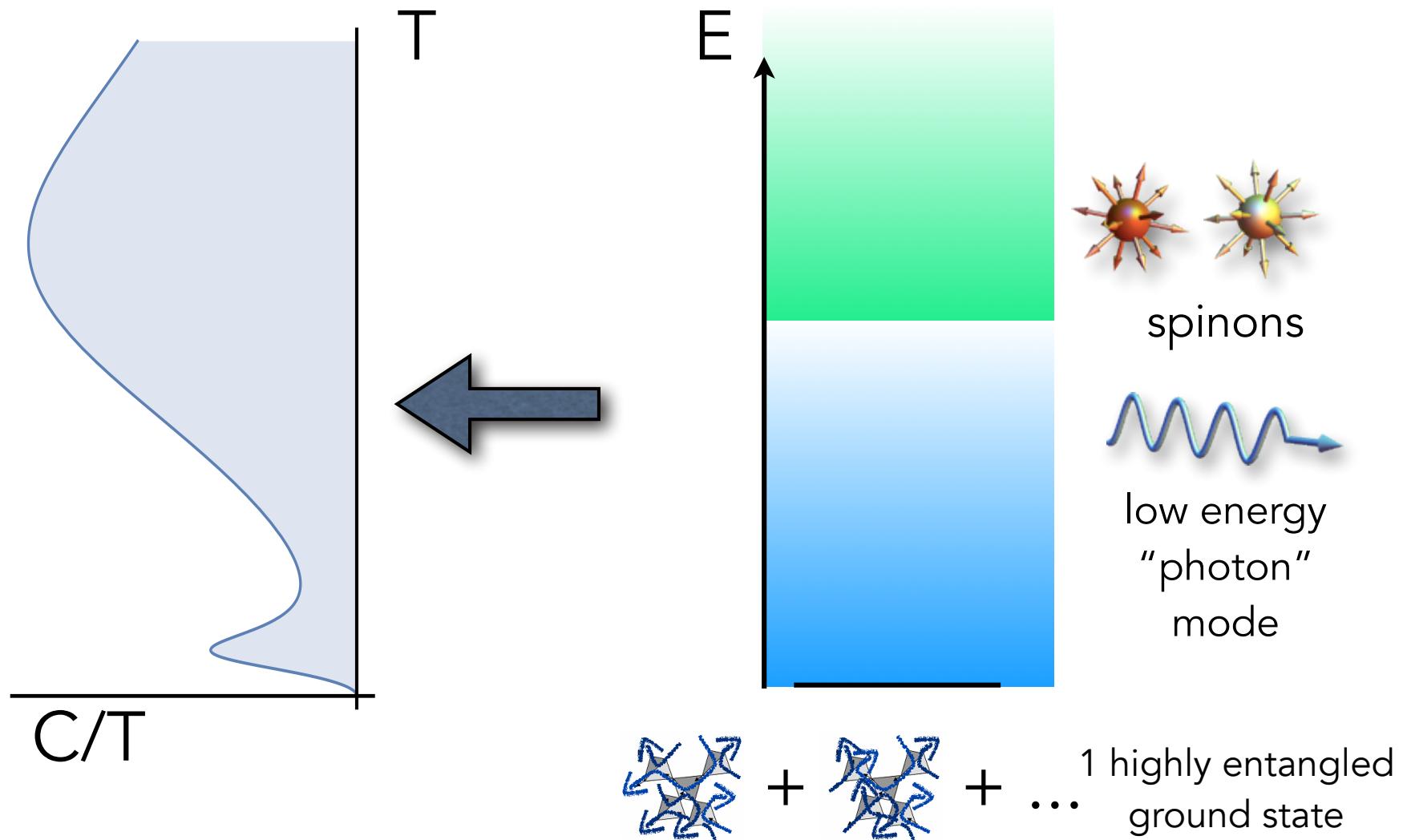
Possible application to  
 $\text{Yb}_2\text{Ti}_2\text{O}_7$ ,  $\text{Pr}_2\text{Zr}_2\text{O}_7$ ,  
others...



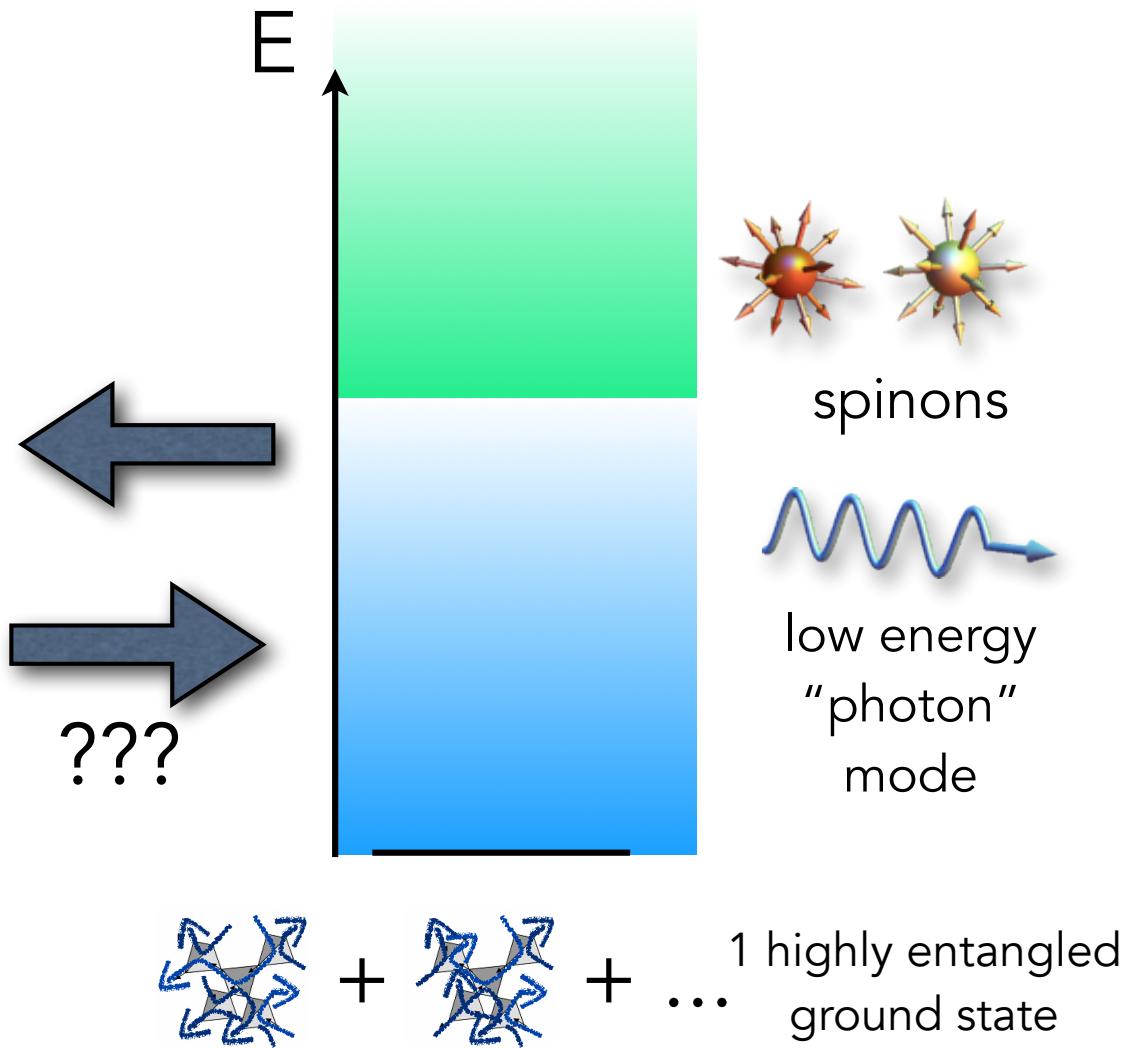
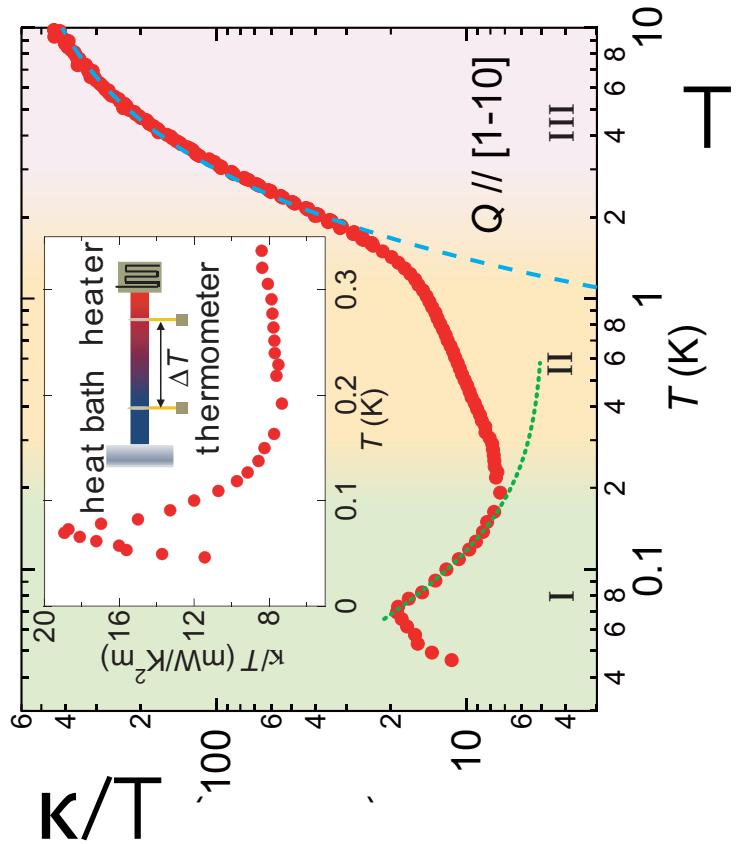
# Quantum versus classical



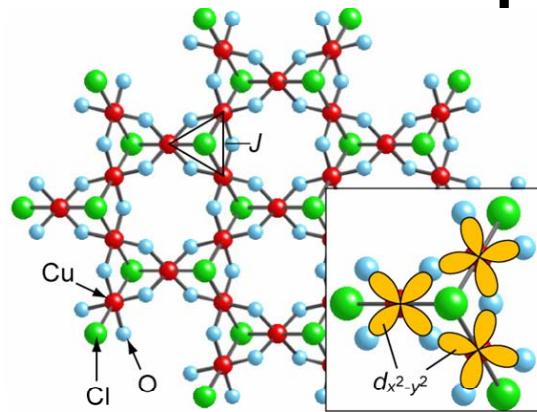
# Quantum spin ice



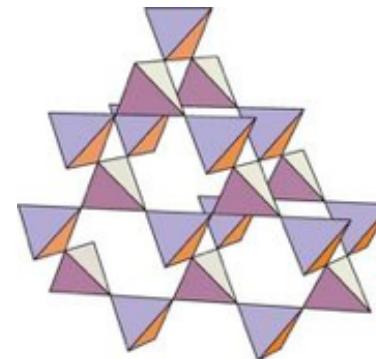
# Quantum spin ice



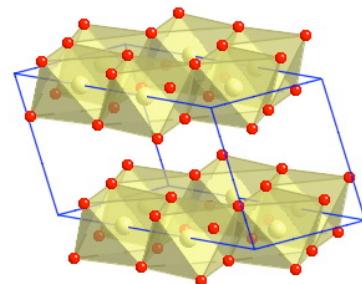
# Top experimental platforms



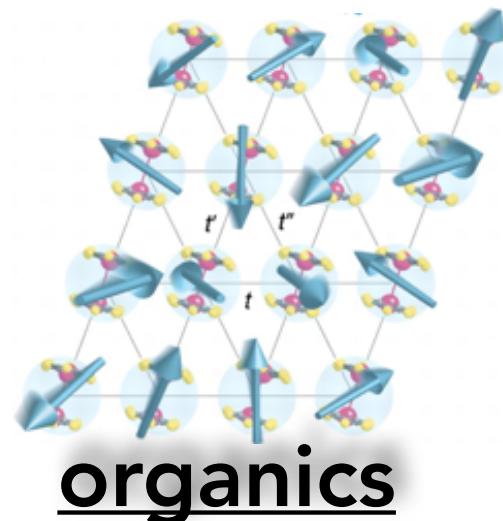
kagomé



Quantum spin ice

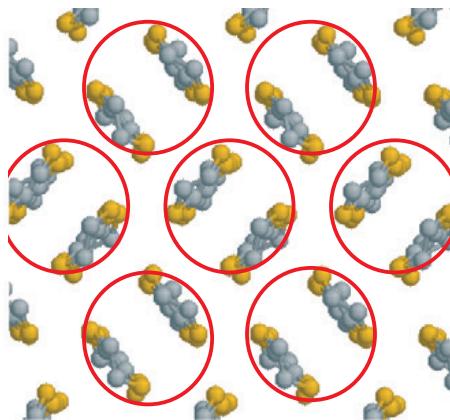


Kitaev materials

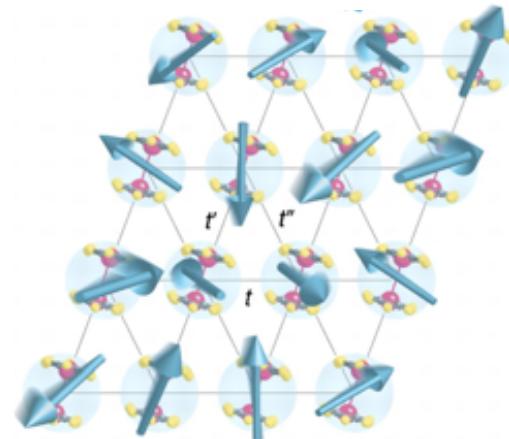


organics

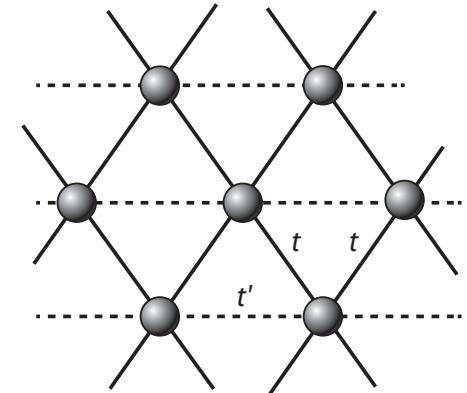
# Organics



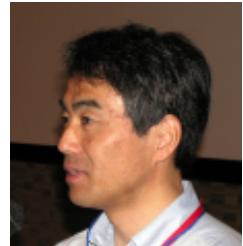
$\kappa$ -(ET)<sub>2</sub>X



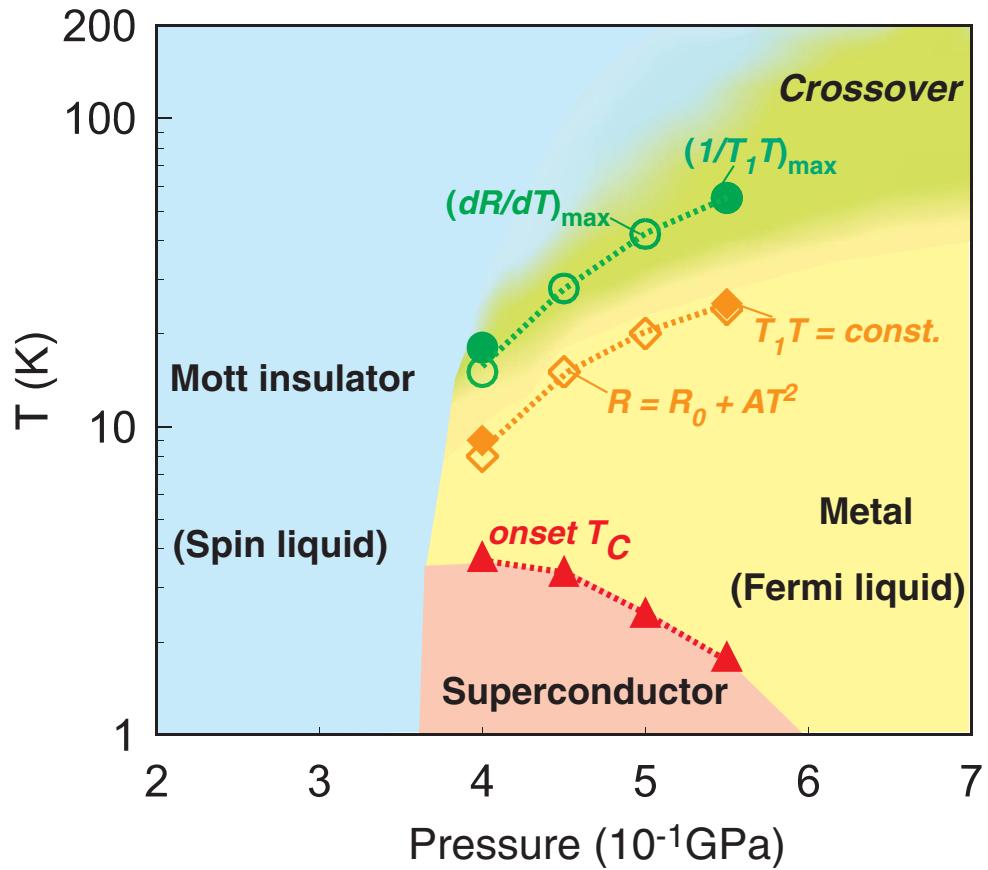
$\beta'$ -Pd(dmit)<sub>2</sub>



- Molecular materials which behave as effective triangular lattice  $S=1/2$  antiferromagnets with  $J \sim 250\text{K}$
- significant charge fluctuations

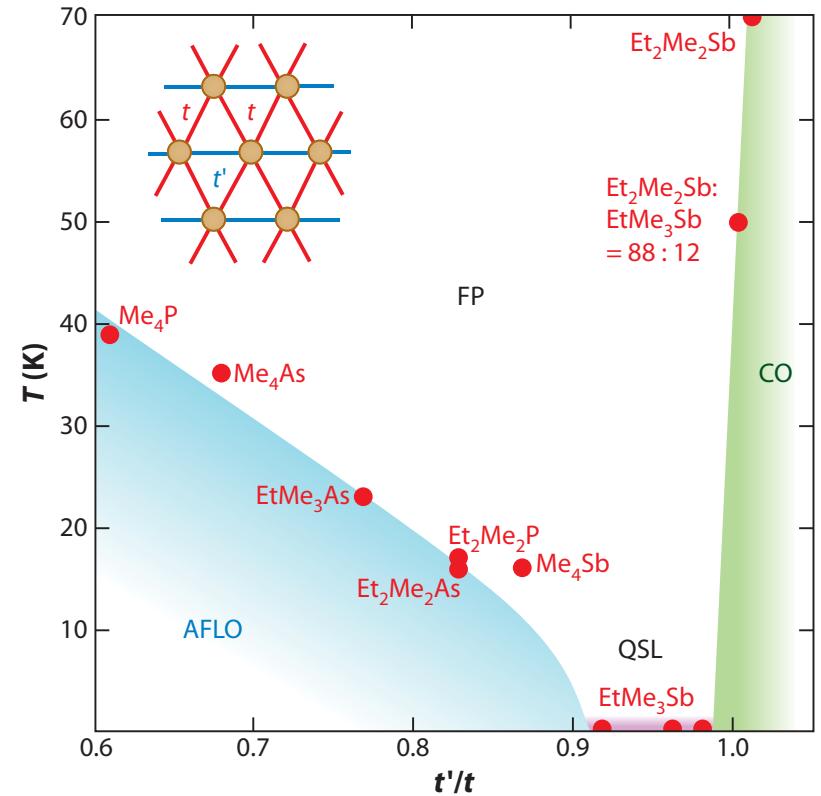


# Organics



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

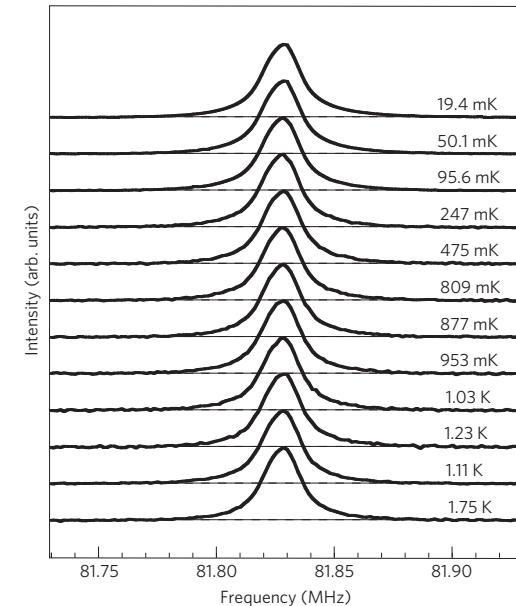
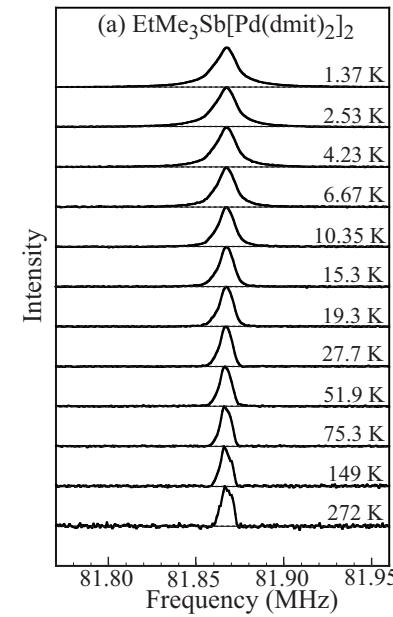
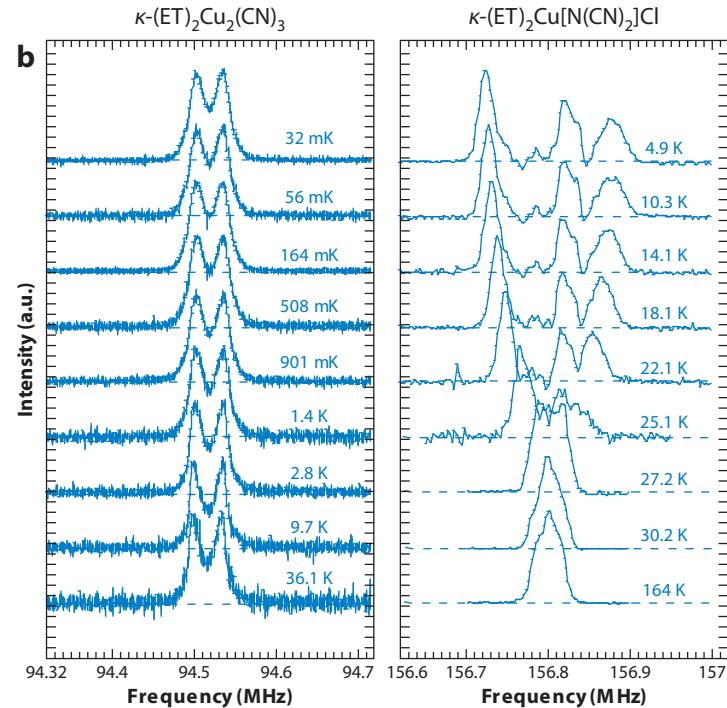
K. Kanoda group (2003-)



$\beta'$ -Pd(dmit)<sub>2</sub>

R. Kato group (2008-)

# NMR lineshapes



$\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

Y. Shimizu  
et al, 2003

$^1\text{H}$  NMR

$\beta'\text{-Pd}(\text{dmit})_2$

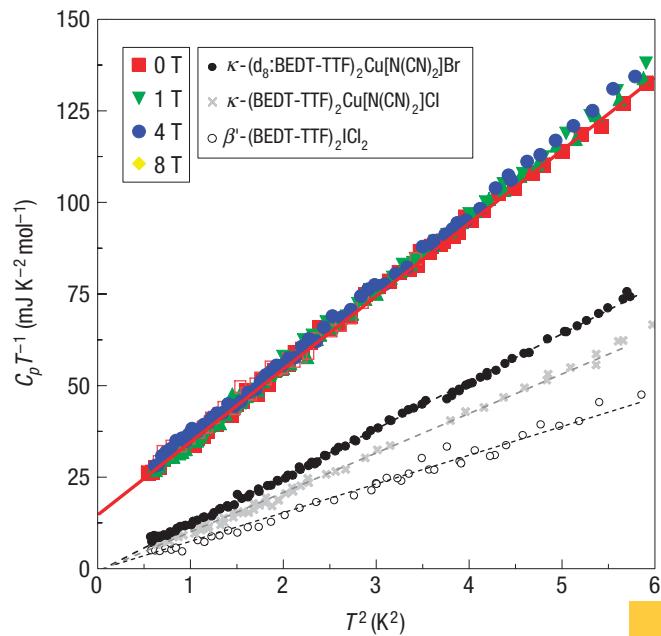
T. Itou et  
al,  
2008, 2010

$^{13}\text{Cs}$  NMR

Evidence for lack of static moments:  $f > 1000!$

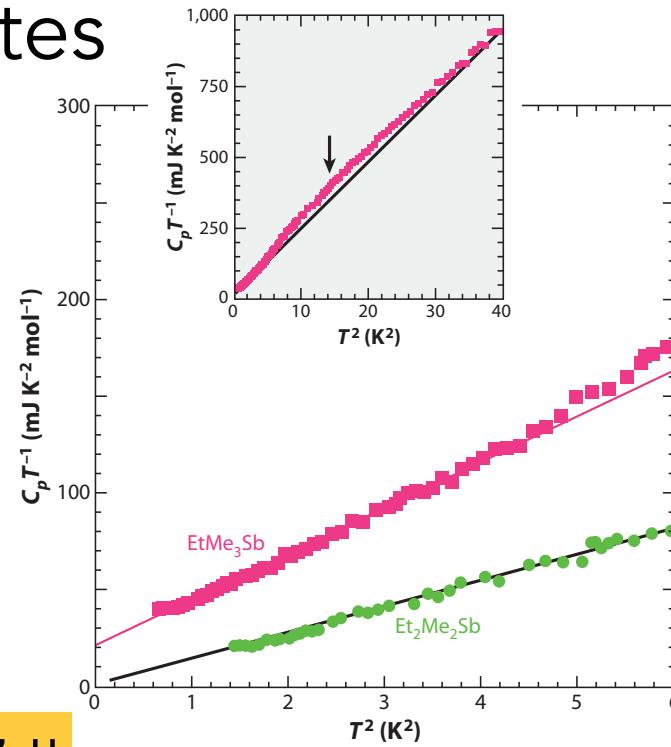
# Specific Heat

- $C \sim \gamma T$  indicates gapless behavior with large density of states



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

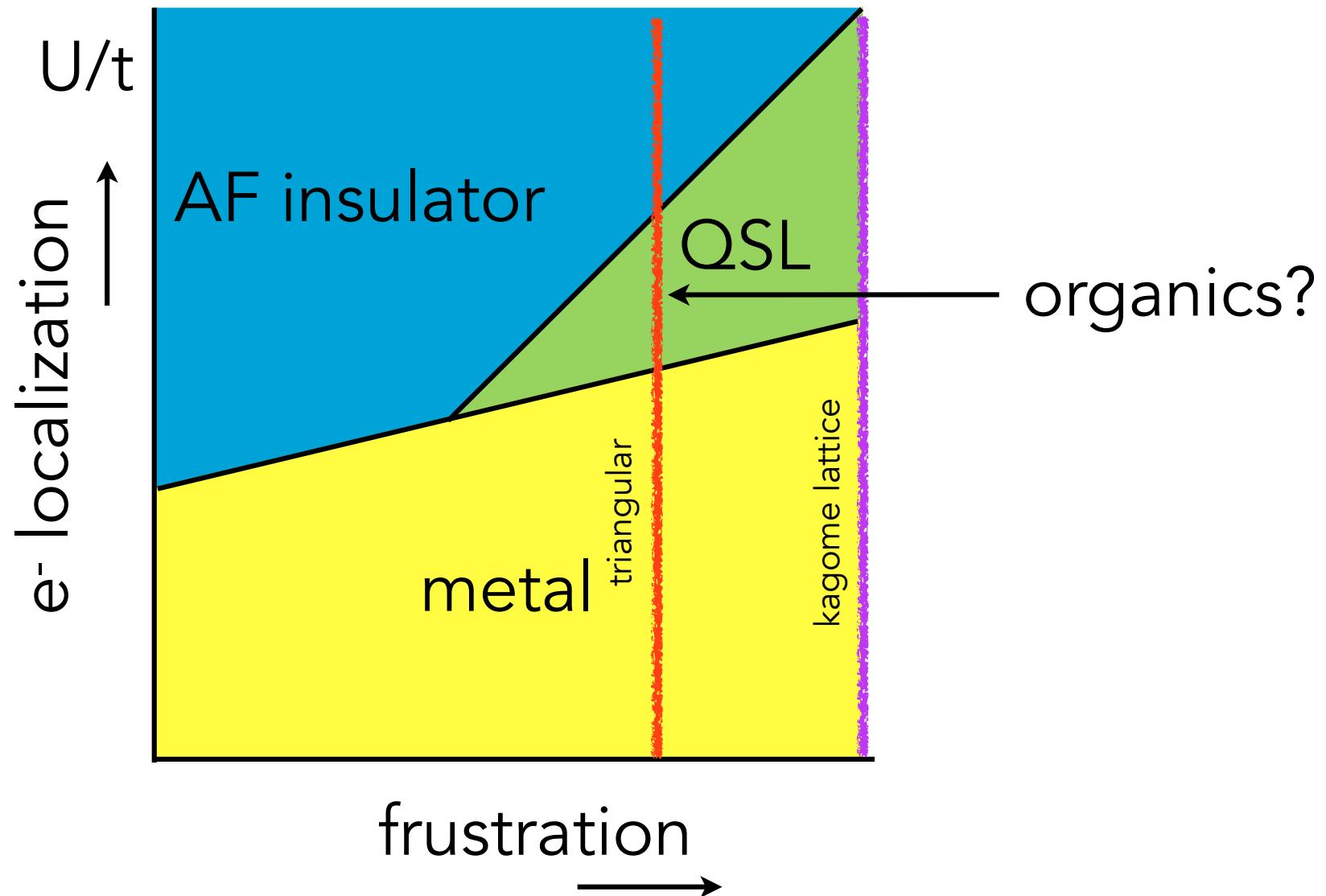
$$\gamma_{\text{Cu}} \sim 0.7 !!$$



$\beta'$ -Pd(dmit)<sub>2</sub>

S. Yamashita *et al*, 2008

# Charge fluctuations

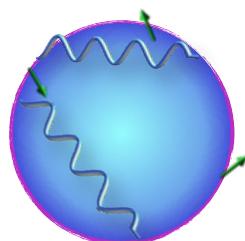


# Organics - Theory



- RVB/QSL state:
  - Motrunich, Lee+Lee: (2005) “uniform RVB”
  - It is described by a **“Fermi sea” of spinons** coupled to a U(1) gauge field

$$|\Psi\rangle = \prod_i \hat{n}_i (2 - \hat{n}_i) \prod_{k < k_F} c_{k\uparrow}^\dagger c_{k\downarrow}^\dagger |0\rangle$$



- The most gapless/highly entangled QSL state
- Like a “metal” of neutral fermions w/ a U(1) gauge field
- Prototype “non-Fermi liquid” state of great theoretical interest

# Spinon Fermi surface

Calculations based on effective field theory “uniform RVB”

- Fermions w/ U(1) gauge field

Zeeman term

$$S_\psi = \int d^3x \psi^\dagger \left( \partial_\tau - \mu - \frac{1}{2m} (\nabla_{\mathbf{r}} - i\mathbf{A})^2 - \omega_B \sigma^3 \right) \psi,$$

Kinetic energy      Emergent gauge field

$$S_A = \int \frac{d^3q}{(2\pi)^3} \frac{1}{2} (\gamma |\omega_n|/q + \chi q^2) |A(q)|^2, \quad \text{Landau damping}$$

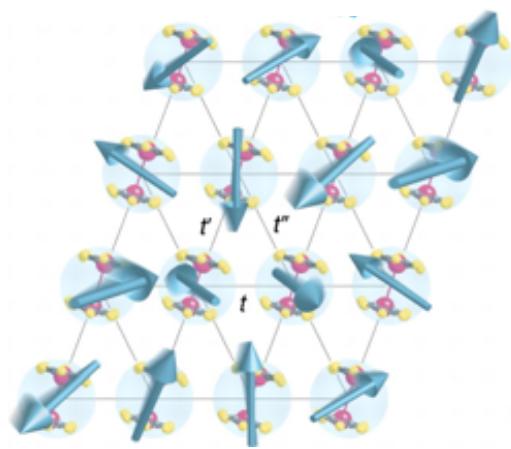
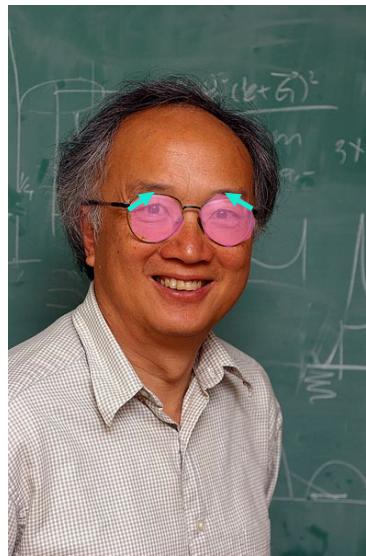
$$S_u = \int d^3x u \psi_\uparrow^\dagger \psi_\uparrow \psi_\downarrow^\dagger \psi_\downarrow. \quad \text{Short-range repulsion (from } a_0 \text{)}$$

Ioffe,Larkin 1989      Nagaosa 1999

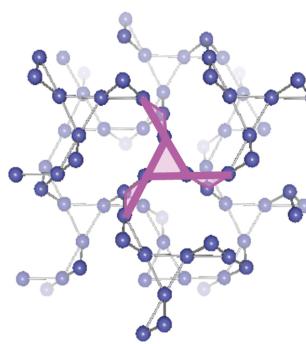
Sachdev, Metlitski, Senthil, McGreevy...

Kim, Furusaki, Lee, Wen 1994

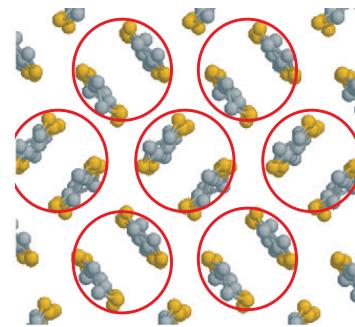
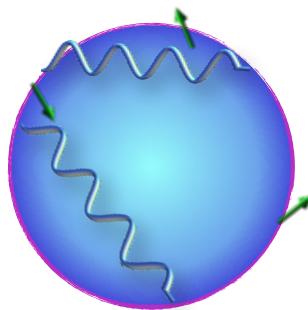
# Spinon Fermi surface



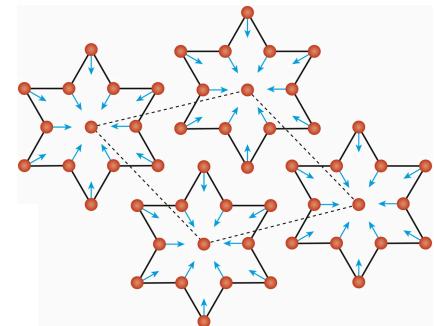
$k$ -ET



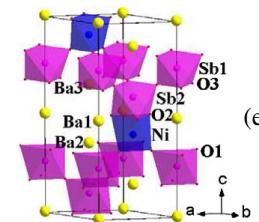
$\text{Na}_4\text{Ir}_3\text{O}_8$



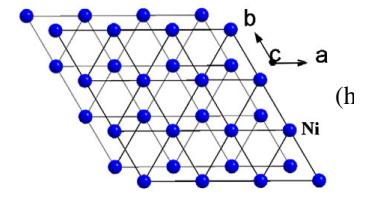
dmit



1T-TaS<sub>2</sub>



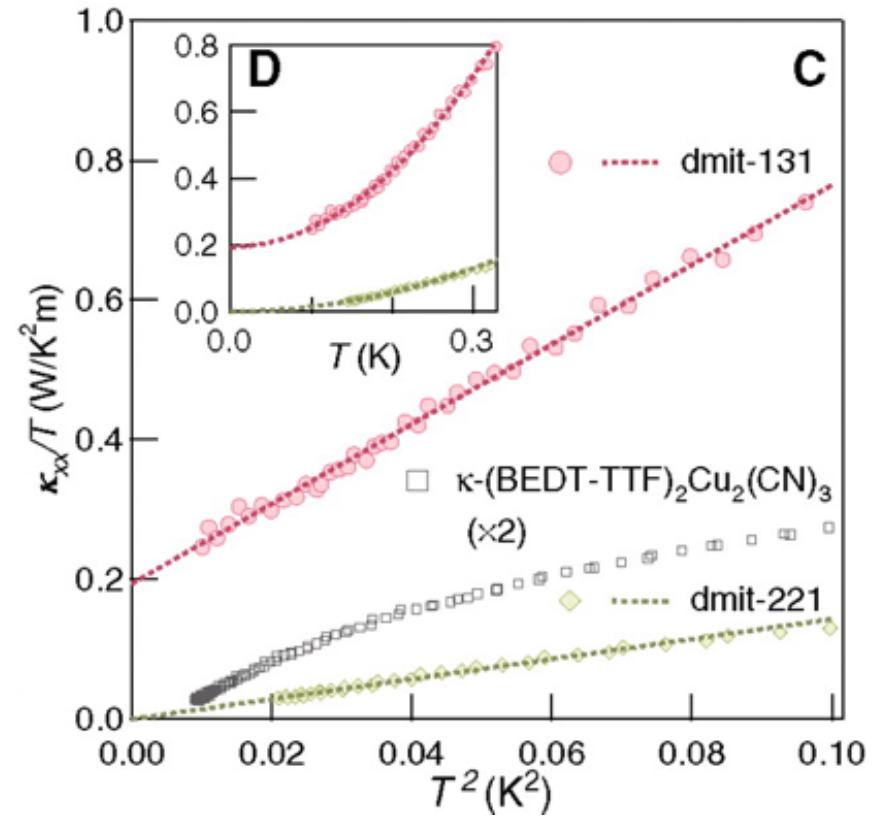
$\text{Ba}_3\text{Ni}\text{Sb}_2\text{O}_9$



(e)  $\text{Ba}_3\text{Ni}\text{Sb}_2\text{O}_9$

# Thermal conductivity

- Huge linear thermal conductivity indicates the gapless excitations are propagating
- Consistent with spinon Fermi surface?
- Estimate for a metal would correspond to a mean free path  $l \sim 1 \mu\text{m}$   
 $\approx 1000 \text{ \AA}$  !



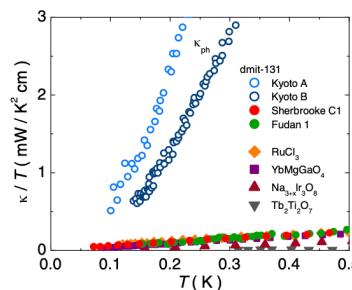
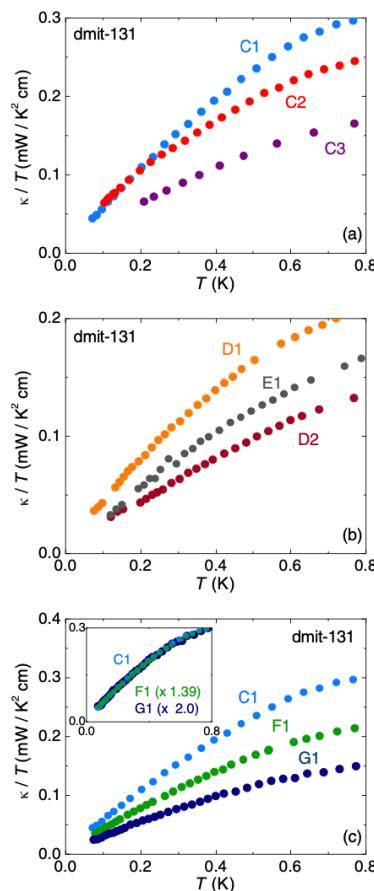
M. Yamashita *et al*, 2010

# 9 years later...

PHYSICAL REVIEW X 9, 041051 (2019)

## Thermal Conductivity of the Quantum Spin Liquid Candidate $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$ : No Evidence of Mobile Gapless Excitations

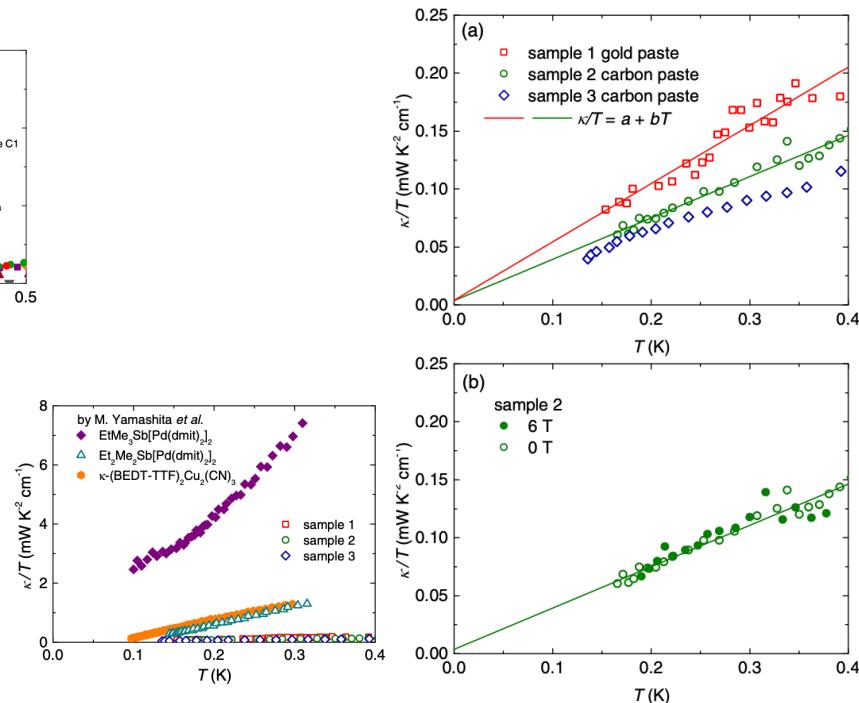
P. Bourgeois-Hope,<sup>1</sup> F. Laliberté,<sup>1</sup> E. Lefrançois,<sup>1</sup> G. Grissonnanche,<sup>1</sup> S. René de Cotret,<sup>1</sup> R. Gordon,<sup>1</sup> S. Kitou,<sup>2</sup> H. Sawa,<sup>2</sup> H. Cui,<sup>3</sup> R. Kato,<sup>3</sup> L. Taillefer,<sup>1,4,\*</sup> and N. Doiron-Leyraud<sup>1</sup>



PHYSICAL REVIEW LETTERS 123, 247204 (2019)

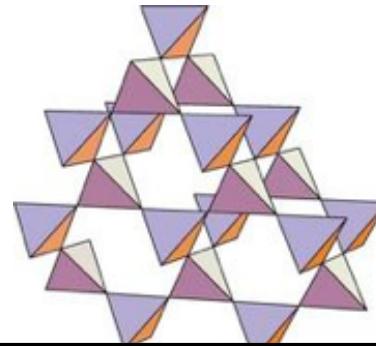
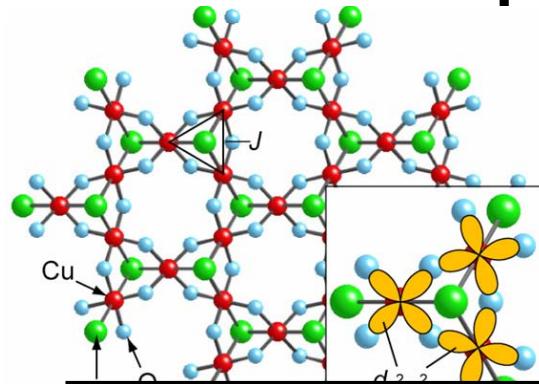
## Absence of Magnetic Thermal Conductivity in the Quantum Spin Liquid Candidate $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$

J. M. Ni,<sup>1</sup> B. L. Pan,<sup>1</sup> B. Q. Song,<sup>1</sup> Y. Y. Huang,<sup>1</sup> J. Y. Zeng,<sup>1</sup> Y. J. Yu,<sup>1</sup> E. J. Cheng,<sup>1</sup> L. S. Wang,<sup>1</sup> D. Z. Dai,<sup>1</sup> R. Kato,<sup>2</sup> and S. Y. Li<sup>1,3,\*</sup>

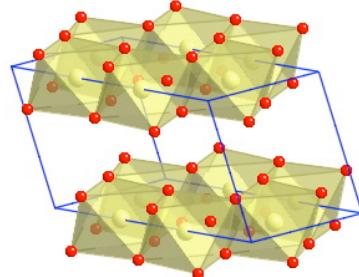


## Controversy!

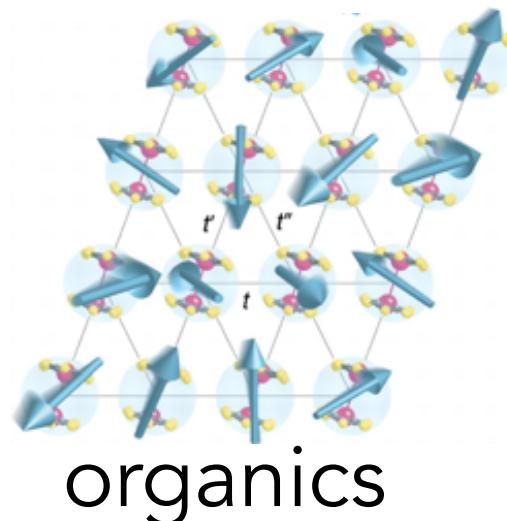
# Top experimental platforms



This just scratches the surface: many more materials being studied



Kitaev materials



organics

# Frontiers

## New phases

- Fractons
- Quenched disorder

## Fundamental problems

- QSLs with strongly coupled matter-gauge theory
- QCPs to/from QSL phases
- Out of equilibrium
- Doping - QSL induced SCivity?

## Reality

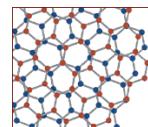
- New Materials! Maybe QSLs in VdW crystals?
- Definitive experimental signatures
  - Thermal Hall? Non-linear spectroscopy?
- Computational methods: less bias, reliability of variational methods, beyond ground states

# Thanks for your attention

References here: <https://spinsandelectrons.com/pedagogy/>



Office of  
Science



Simons Collaboration on  
Ultra-Quantum Matter

