

Leon Balents  
CIFAR school, Toronto, April 2016

# Quantum Spin Liquids

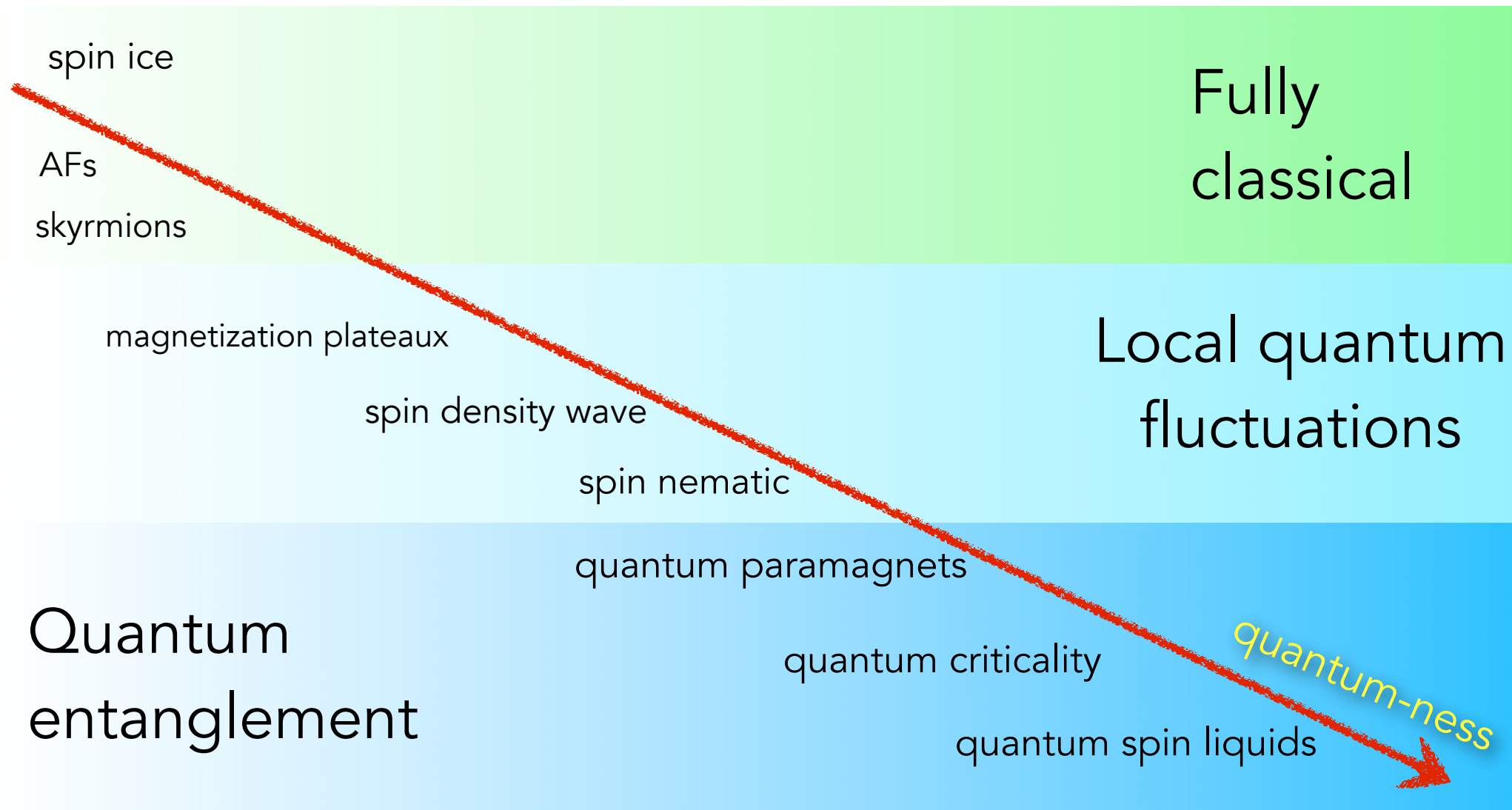


# Plan

- The bird's eye view: QSLs as *ultra-quantum matter*
  - What is different from ordinary stuff?
- Review experimental status and recent developments

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

# Quantum Magnetism



# Quantum non-locality

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



A. Einstein



B. Podolsky



N. Rosen



# Schrödinger Cat

$$\frac{1}{\sqrt{2}}|\text{cat}\rangle + \frac{1}{\sqrt{2}}|\text{dead cat}\rangle$$

Cat states - superposition of a small number of macroscopically distinct components - are exponentially unstable: any local measurement collapses the superposition. They also require an exponentially long time to assemble with local unitary operators

# How quantum can dense matter *stably* be?

Quantum spin liquids are ground states that retain long-distance entanglement and are robust to perturbations



“Ultra-quantum matter”: stable *phases* of matter that retain some degree of quantum non-locality

# 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

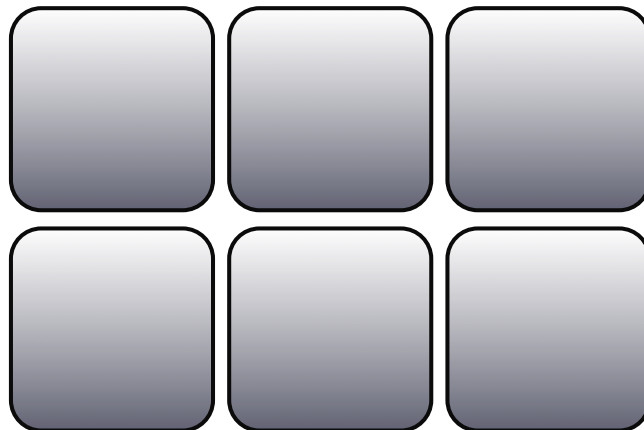
# Ordinary (local) Matter

Hamiltonian is local

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

Ground state is “essentially”  
a product state

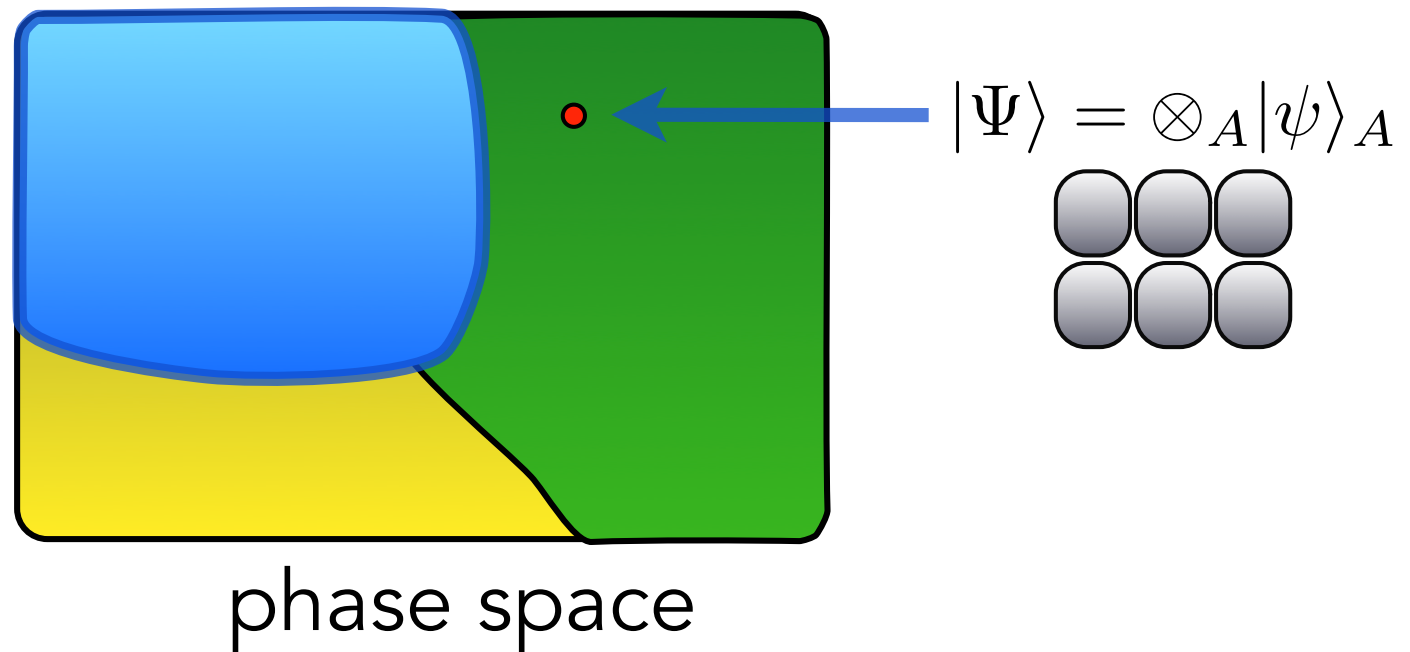
$$|\Psi\rangle = \bigotimes_A |\psi\rangle_A$$



no entanglement  
between blocks

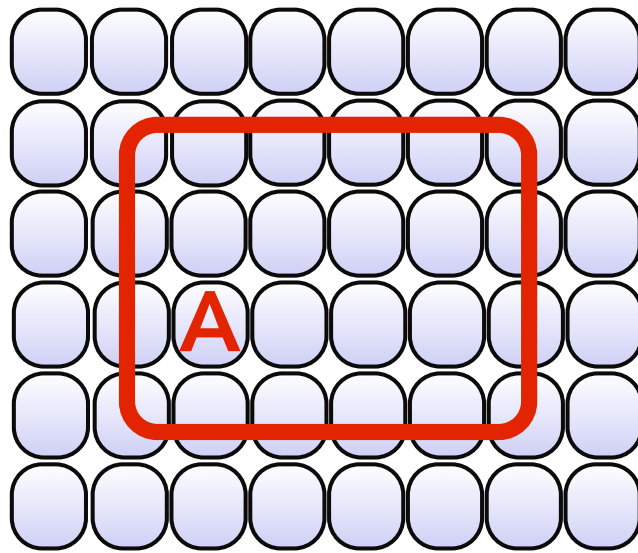
# "Essentially" a product state?

- Adiabatic continuity



# “Essentially” a product state?

- Entanglement scaling



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

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

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

satisfied with exponentially small corrections

# Best example: ordered magnet

Hamiltonian

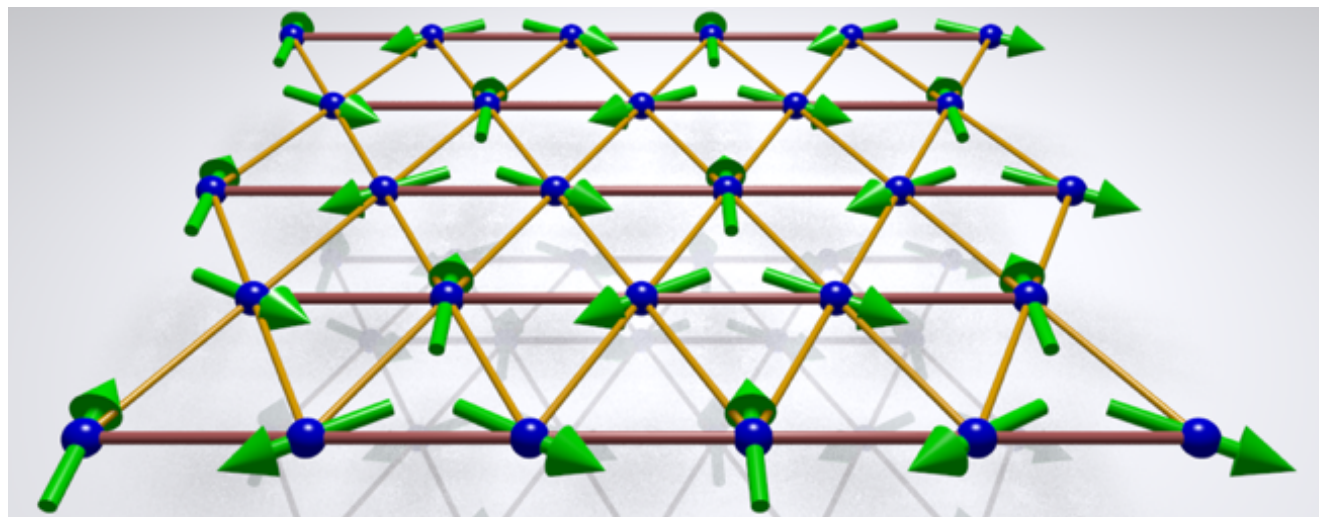
$$H = \sum_{(ij)} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

exchange is short-  
range: local

ordered state

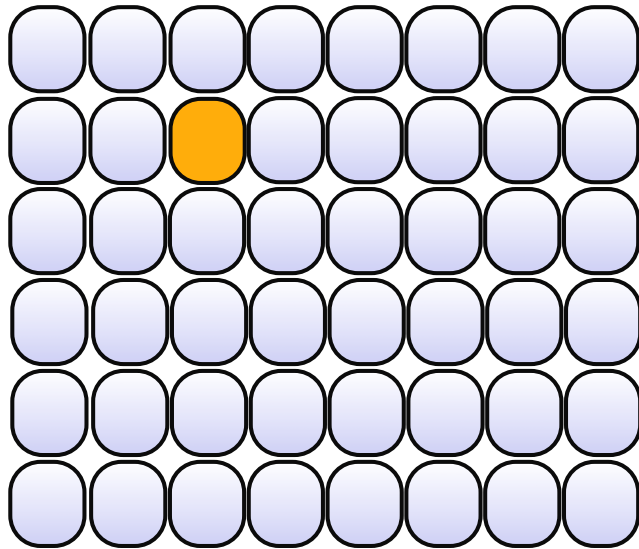
$$|\Psi\rangle \approx \bigotimes_i |\mathbf{S}_i \cdot \hat{n}_i = +S\rangle$$

block is a single  
spin





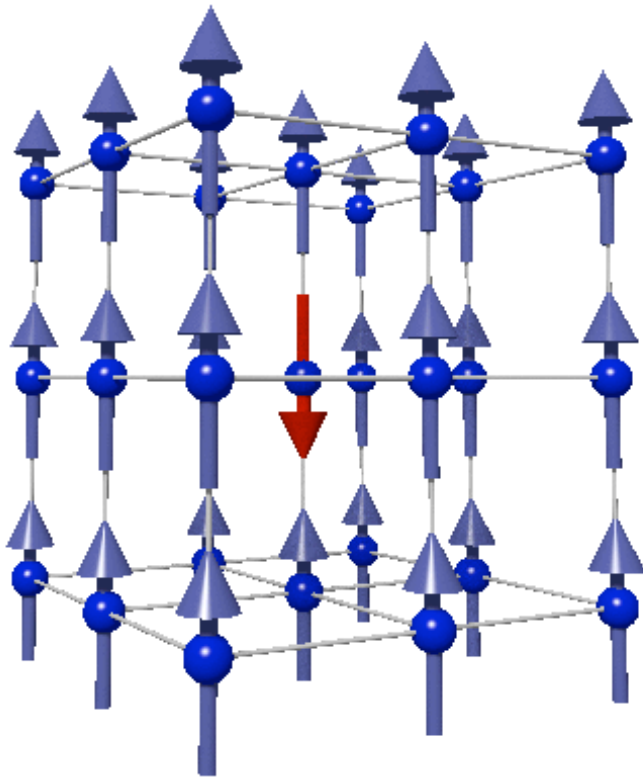
# Quasiparticles



excited states  $\sim$  excited levels of one block

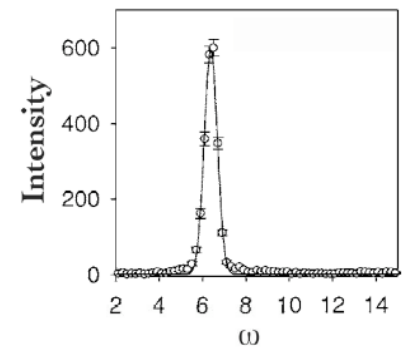
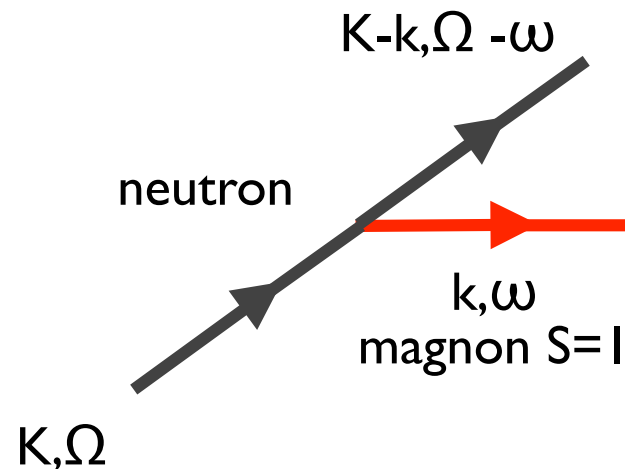
- local excitation can be created with operators in one block
- localized excitation has discrete spectrum with non-zero gap, and plane wave forms sharp band
- quantum numbers consistent with finite system: no emergent or fractional quantum numbers

# Spin wave



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

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



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

# How quantum can dense matter *stably* be?

Quantum spin liquids are ground states that retain long-distance entanglement and are robust to perturbations



©Bruce Gaulin

“Ultra-quantum matter”: stable *phases* of matter that retain some degree of quantum non-locality

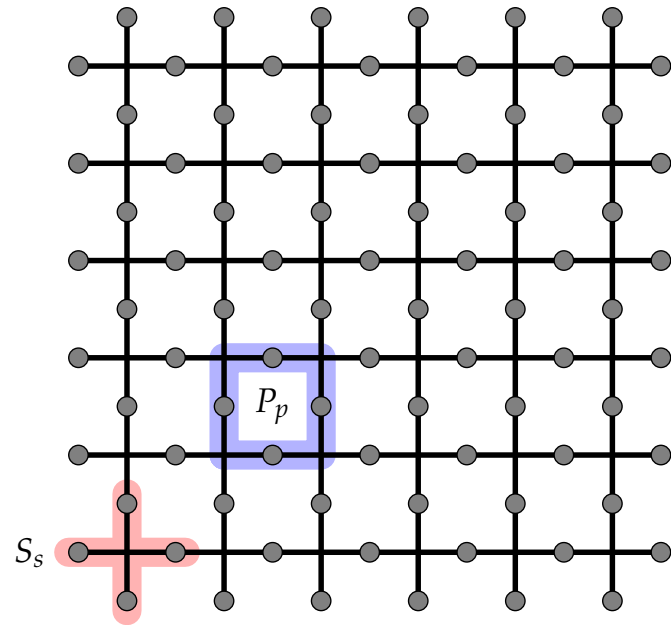
# Example: toric code



A. Kitaev

$$H_{\text{tc}} = -K \sum_p P_p - K' \sum_s S_s,$$

$$P_p = \prod_{i \in p} \sigma_i^z \quad S_s = \prod_{i \in s} \sigma_i^x$$



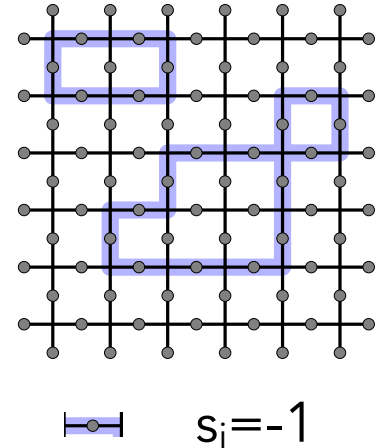
Everything commutes!

➡ In ground state simply  $P_p = S_s = +1$

But what is the state?

# Ground state

Product basis  $|\{s_i\}\rangle = \bigotimes_i |\sigma_i^x = s_i\rangle$



Solve  $S_s = +1$ :

$$|\psi_0\rangle = \bigotimes_i |\sigma_i^x = +1\rangle$$

Now project to  $P_p = +1$ :  $Q_p = \frac{1 + P_p}{2} = \frac{1}{2} \sum_{q_p=0,1} P_p^{q_p}$

$$|0\rangle = \prod_p Q_p |\psi_0\rangle = 2^{-N} \sum_{q_1 \dots q_N=0,1} \prod_p P_p^{q_p} |\psi_0\rangle$$

# Ground state

$$|0\rangle = 2^{-N} \sum_{q_1 \dots q_N=0,1} \prod_p P_p^{q_p} |\psi_0\rangle = \sum_{\text{loops}} \text{[Diagram of a 6x6 grid with blue loops and labels } P_p \text{]} \quad \begin{array}{l} \text{massive} \\ \text{superposition} \\ \text{state} \end{array}$$

(similar form in Z variables)

All spins are uncertain

$$\begin{aligned}\langle 0 | \sigma_i^x | 0 \rangle &= \langle 0 | S_s \sigma_i^x S_s | 0 \rangle = -\langle 0 | \sigma_i^x | 0 \rangle = 0 \\ \langle 0 | \sigma_i^z | 0 \rangle &= \langle 0 | P_s \sigma_i^z P_p | 0 \rangle = -\langle 0 | \sigma_i^z | 0 \rangle = 0\end{aligned}$$

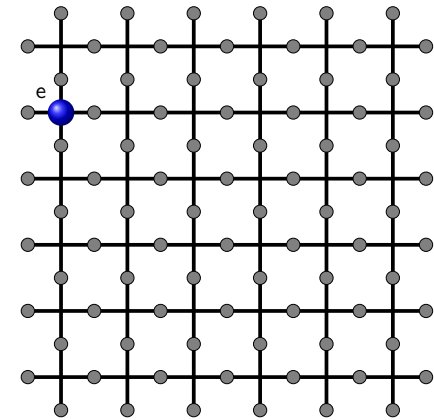
And yet there is some structure...

# Excitations

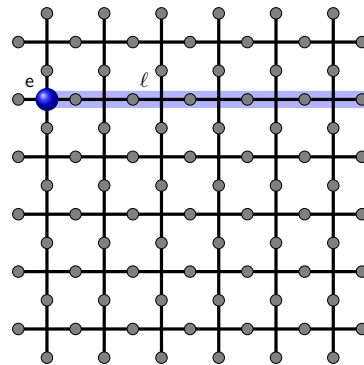
$$H_{\text{tc}} = -K \sum_p P_p - K' \sum_s S_s,$$

Consider state with  
just one  $S_s = -1$

$$|e_s\rangle =$$



$$|e_s\rangle = \prod_{i \in \ell} \sigma_i^z |0\rangle$$



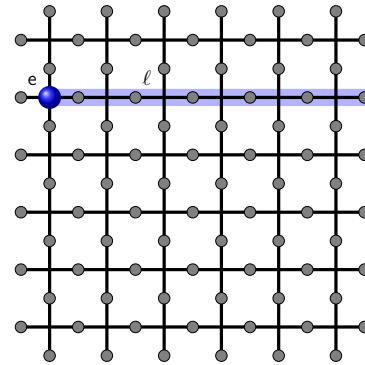
non-local excitation!

local operators, like finite  
product of  $Z$  operators,  
create **e** particles in pairs

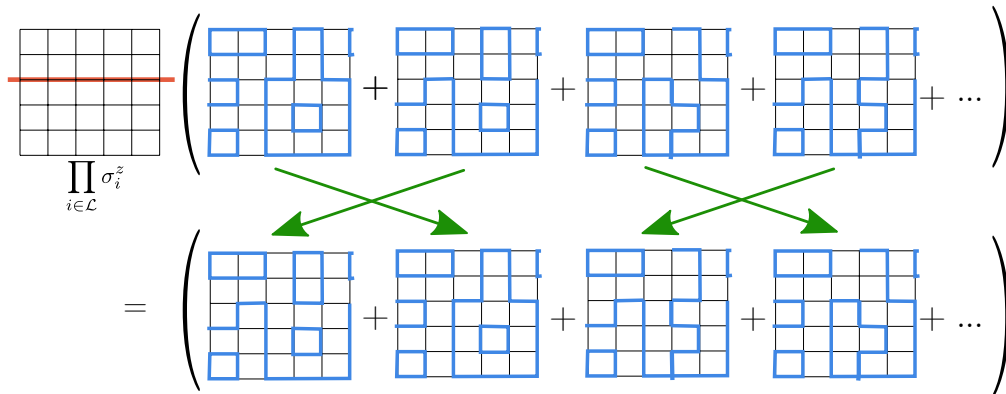


# Non-local excitations and entanglement

$$|e_s\rangle = \prod_{i \in \ell} \sigma_i^z |0\rangle$$



Why does this have finite energy?      $\langle e_s | H | e_s \rangle \propto L_\ell ??$



away from the ends, the  
string just reshuffles  
elements of the  
superposition

# Excitations

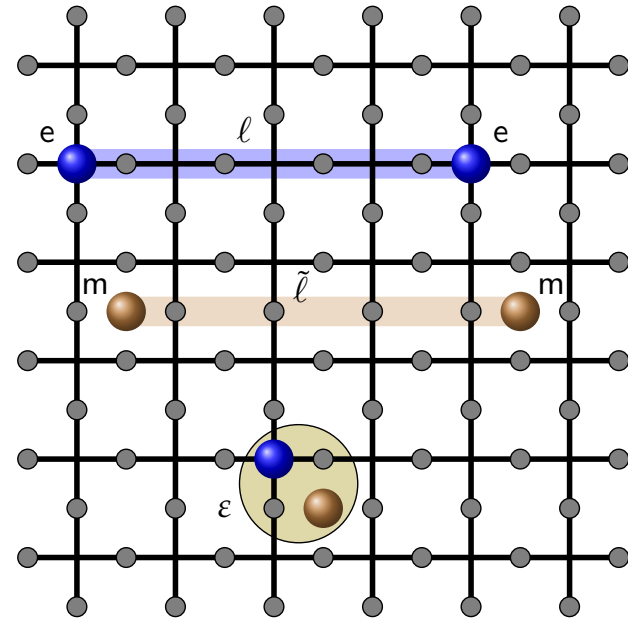
There is also an **m** excitation

$$P_p = -1$$

And we can consider two of these together

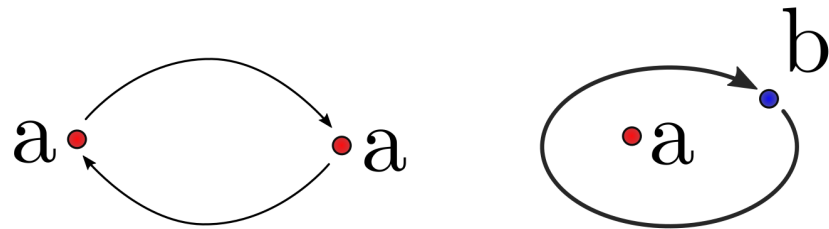
$$\varepsilon = e \times m$$

**e, m,  $\varepsilon$**  are the *emergent non-local quasiparticles* of the toric code



# Non-locality

For the toric code, and in general for *topological phases* (i.e. gapped QSLs), the non-locality of excitations manifests as *statistics*



the excitations are *anyons*, i.e. the state acquires a unitary rotation (e.g. phase) when one excitation is taken around another

$$\begin{array}{c} e \quad m \\ \diagdown \quad \diagup \\ e \quad m \end{array} = - \begin{array}{c} e \quad m \\ | \quad | \\ e \quad m \end{array}$$

**e** and **m** see each other as “pi flux”

$$\begin{array}{c} e \quad m \quad e \quad m \\ \diagdown \quad \diagup \quad \diagdown \quad \diagup \\ e \quad m \quad e \quad m \end{array} = \begin{array}{c} e \quad m \quad e \quad m \\ | \quad \diagdown \quad | \quad \diagup \\ e \quad m \quad e \quad m \end{array} = - \begin{array}{c} e \quad m \quad e \quad m \\ | \quad | \quad | \quad | \\ e \quad m \quad e \quad m \end{array}$$

**$\epsilon$**  is a fermion

# Stability

One can show that the toric code phase is *absolutely stable* to arbitrary (small) local perturbations, even those which break all symmetries.

This is because the “order” of the toric code is purely a type of entanglement, not any symmetry breaking. Only by bringing the gap of a quasiparticle to zero can one “unwind” the entangled ground state.

rigorous proofs by Hastings, Bravyi



# RVB states

Historically the first proposal of a QSL by Anderson in 1973

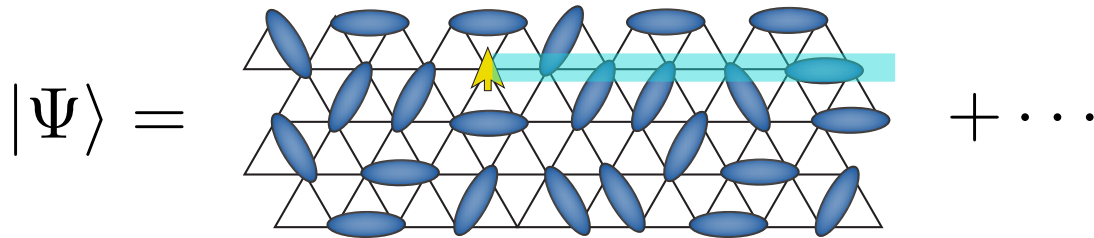
$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \quad \text{blue oval} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

$$|\Psi\rangle = \text{[Diagram 1]} + \text{[Diagram 2]} + \dots$$

The diagram shows a superposition of two states. Each state is represented by a triangular lattice of sites. In each state, blue ovals (representing singlets) are placed on the bonds between sites. In the first state, the ovals form a specific pattern. In the second state, the ovals are shifted. The states are added together, as indicated by the plus signs and ellipsis.

superposition of singlets is quite similar to sum of loops in toric code, and indeed when made precise such a nearest-neighbor RVB state is typically *in the same phase as the toric code*

# Spinon



create a spinon by rearranging valence bonds to expose a single free spin

New feature:  $SU(2)$  spin symmetry

- spinon excitation has  $S=1/2$ , a *fractional quantum number* (spin flips are  $S=1$ )
- this "enriches" the topological label  **$\mathbf{e}, \mathbf{m}, \boldsymbol{\varepsilon}$**

many efforts to understand Symmetry Enriched Topological order

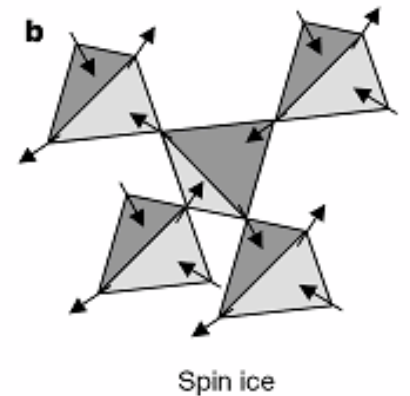
# Quantum spin ice

Minimal XXZ model on *pyrochlore* lattice

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

$$\frac{J_{zz}}{2} \sum_t \left( \sum_{i \in t} S_i^z \right)^2 \Rightarrow J_{\pm} \ll J_{zz} \text{ is the spin ice limit}$$

$$\sum_{i \in t} S_i^z = 0 \text{ in the classical ground state: "2in-2out"}$$



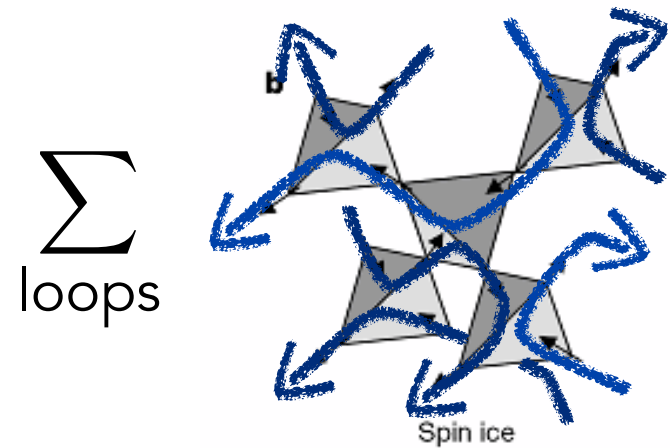
With a lot more work one can show that  $J_{\pm}$  selects a massive superposition of these states

M. Hermele, MPA Fisher, L.B., 2004;  
A. Banerjee et al, 2008



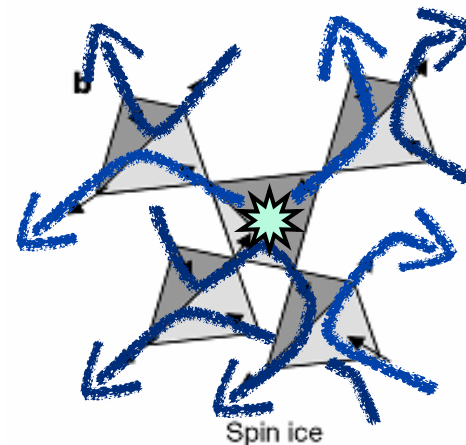
# Quantum spin ice

like the toric code, the QSL state can be viewed as a sum of loops - follow the arrows!



There are also non-local excitations

$$\sum_{i \in t} S_i^z = \pm 1$$



"spinons"  
"monopoles"

# Gauge theory

These QSL states are all conveniently described mathematically by *gauge theory*

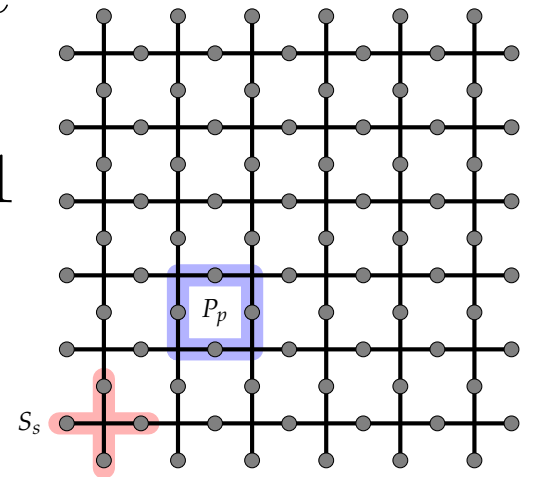
- Local constraints:  $S_s = +1 \quad \sum_{i \in t} S_i^z = 0$
- Generate local unitaries

$$U = U^\dagger = \prod_s S_s^{(1-q_s)/2} \quad q_s = \pm 1$$

$$\sigma_{ss'}^z \rightarrow U^\dagger \sigma_{ss'}^z U = q_s q_{s'} \sigma_{ss'}^z$$

$\sigma_{ss'}^z$  are  $\mathbb{Z}_2$  gauge fields

toric code = " $\mathbb{Z}_2$  QSL"



# Gauge theory

These QSL states are all conveniently described mathematically by *gauge theory*

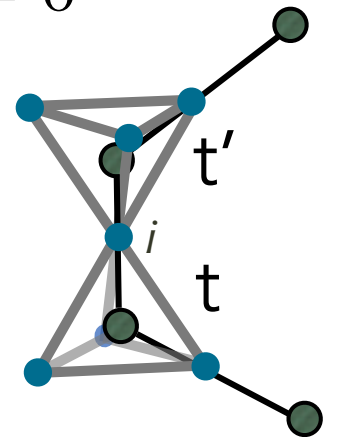
- Local constraints:  $S_s = +1 \quad \sum_{i \in t} S_i^z = 0$
- Generate local unitaries

$$U = \prod_t e^{i\chi_t S_t^z} \quad \chi_t \in U(1)$$

$$S_{tt'}^\pm \rightarrow U^\dagger S_{tt'}^\pm U = e^{i(\chi_t - \chi_{t'})} S_{tt'}^\pm$$

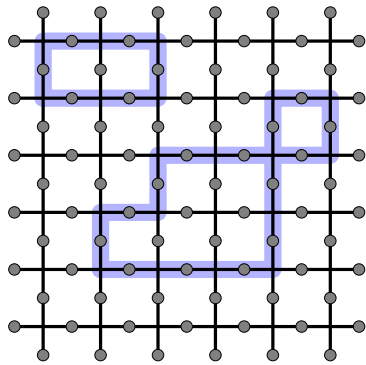
$S_{tt'}^\pm \sim e^{\pm i A_{tt'}}$  is a U(1) gauge connection

QSI hosts a U(1) QSL

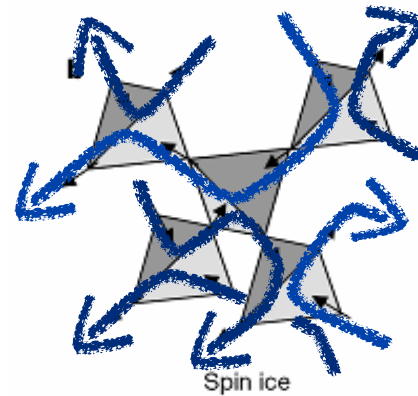


# Gauge theory

- Loops  $\sim$  field lines describing "vacuum fluctuations"



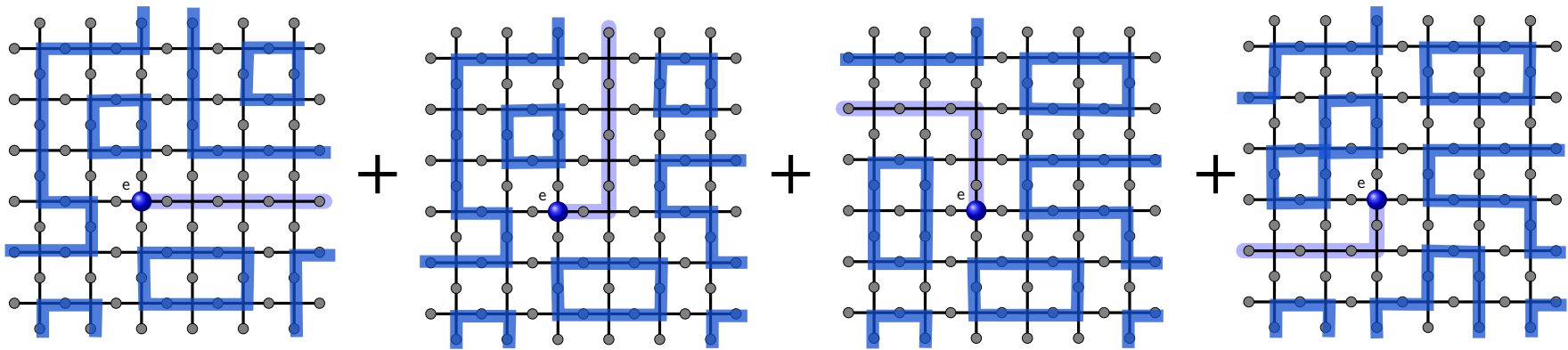
toric code:  $Z_2$



QSI:  $U(1)$

# Deconfinement

$$|e_s\rangle =$$

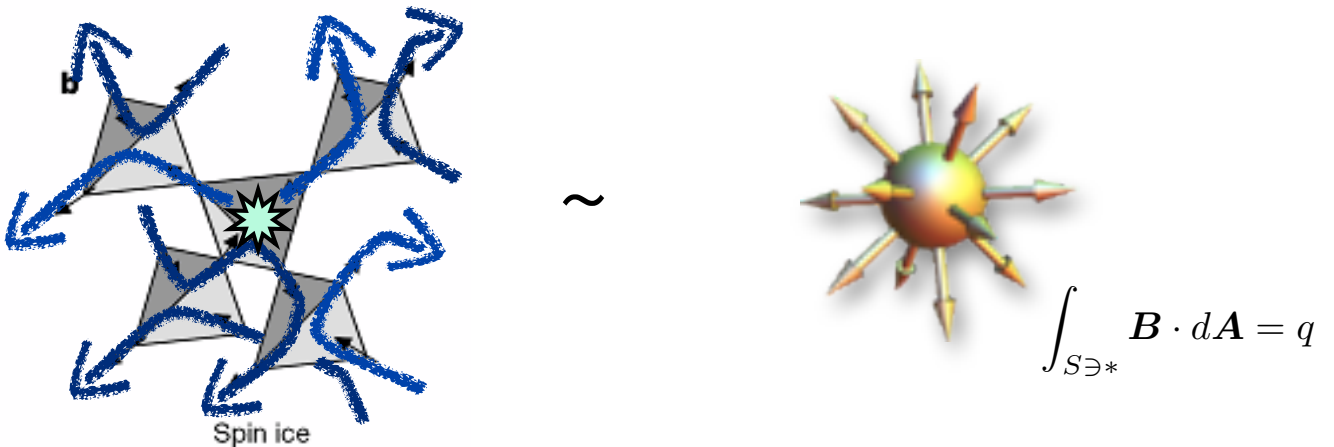


The superposition of different string configurations "smears out" the flux emanating from **e**, so that it cannot be detected by any *local* measurement away from the quasiparticle

- consequently the string tension is zero = deconfinement
- the complete local undetectability of the flux is characteristic of a *topological* QSL

# Deconfinement

U(1) QSL

$$|\text{spinon}\rangle = \sum_{\text{flux lines}} \sim$$


Here the flux is a number, not a parity, and can be added.  
The superposition of many field lines smears the flux into a  
“uniform” dispersed magnetic/electric field

$$\mathbf{B} \sim \frac{\hat{r}}{r^2}$$

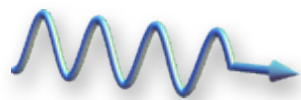
This leads to the usual  $1/r$  Coulomb interaction between charges. It decays with distance so charges are “deconfined”, but the flux is locally detectable

# Photon

Since there is a detectable average field, there is energy density associated with the field lines

Consequently, there are “pure gauge” excitations. These are not non-local particles but emergent collective excitations. They are exactly analogous to the photons of electromagnetism.

$$\omega \sim ck$$

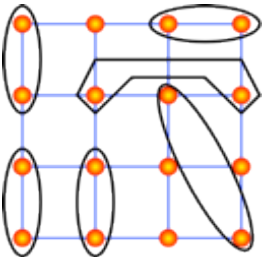
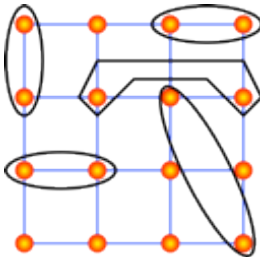


U(1) QSL is a gapless, not topological  
example of ultra-quantum matter



# Variational method

- Up to now I described results out of soluble or otherwise tractable models
- Variational wavefunctions are an attractive way to approach models when no other handle is available

RVB:  $\Psi = c_1$    $+ c_2$    $+ \dots$

but how to keep track of so many coefficients??

# Free Fermions

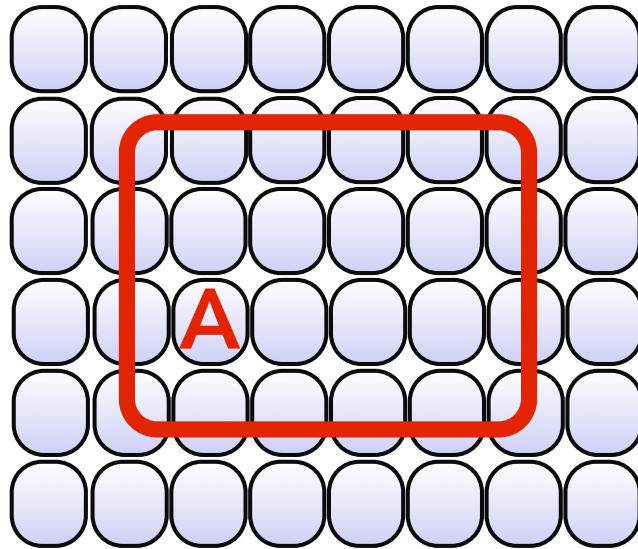
- One useful construction uses a Fermi gas: a product in momentum space rather than real space

$$\Psi = \prod_{k < k_F} c_k^\dagger |0\rangle$$

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

# Entanglement Entropy

- Free fermions  $S \sim \sigma L^{d-1} \log L$



D. Gioev+I. Klich, 2006  
M.M. Wolf, 2006

- Very large entanglement is generic. A metal is in this sense “ultraquantum”  
however, it has local quasiparticles

# Gutzwiller Construction

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

$$\Psi_0 = c_1 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & & \uparrow & \uparrow\downarrow & \downarrow \\ \hline \downarrow & \downarrow & \uparrow\downarrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_2 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \uparrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow & \downarrow & \downarrow \\ \hline \downarrow & \downarrow & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + c_3 \begin{array}{|c|c|c|c|c|} \hline \uparrow & \downarrow & \uparrow & \uparrow & \downarrow \\ \hline \downarrow & \uparrow & \uparrow\downarrow & \downarrow & \uparrow \\ \hline \downarrow & \downarrow & \uparrow & & \downarrow \\ \hline \downarrow & & \downarrow & \downarrow & \downarrow \\ \hline \uparrow & \uparrow\downarrow & \downarrow & \downarrow & \uparrow \\ \hline \end{array} + \dots$$

# Gutzwiller Construction

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- Projection removes empty and doubly occupied sites  $\Psi = P_G \Psi_0$

"like" a gauge constraint  $n_i = 1$

# Gutzwiller Construction

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

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

- Such wavefunctions can be efficiently simulated using Monte Carlo methods

# Partons

- Gutzwiller-type variational wavefunction uses a reference Hamiltonian

$$H_{ref} = \sum_{ij} \left[ t_{ij} c_{i\alpha}^\dagger c_{j\alpha} + \text{h.c.} + \Delta_{ij} c_{i\uparrow}^\dagger c_{j\downarrow}^\dagger + \text{h.c.} \right]$$

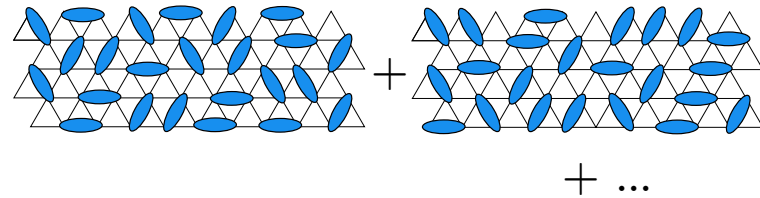
- Project

$$|\Psi_{var}\rangle = \prod_i \hat{P}_{n_i=1} |\Psi_{ref}\rangle$$

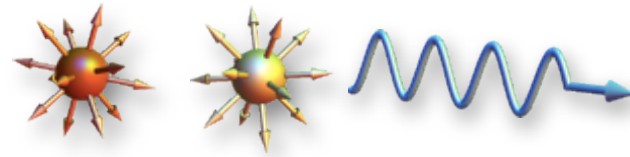
- The fermions are “partons”  $\vec{S}_i = c_{i\alpha}^\dagger \frac{\vec{\sigma}_{\alpha\beta}}{2} c_{i\beta}$
- Standard (MIT) belief: each such projected wavefunction represents a true QSL phase, in which the partons become the non-local quasiparticles - “spinons” - and they are coupled to an effective gauge field

# Classes of QSLs

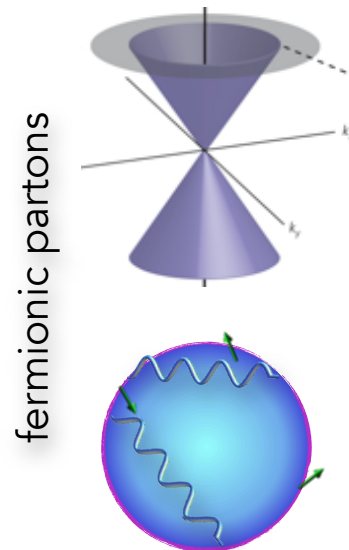
- Topological QSLs
  - full gap
- U(1) QSL
  - gapless emergent "photon"
- Algebraic QSLs
  - Relativistic CFT (power-laws)
- Spinon Fermi surface QSL



TQFT



compact  
U(1)  
gauge  
theory



QED<sub>3</sub>

QED<sub>3</sub>  
w/  $\mu > 0$



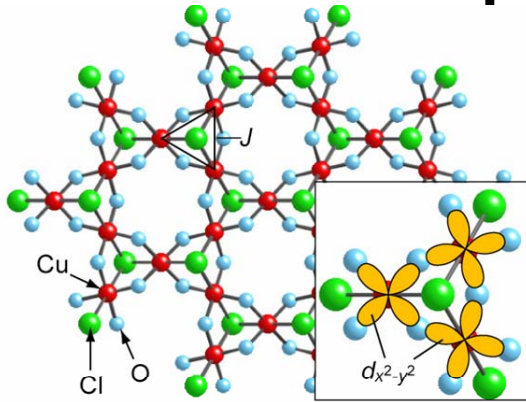
# Summary so far

- QSLs are examples of “ultra-quantum matter” whose ground states are massive superpositions and exhibit long-range entanglement
- Characteristically they support *non-local* excitations, which might be anyons or other exotic particles
- The natural theoretical description of many QSLs is gauge theory
- Many QSLs are absolutely stable to all small perturbations, irrespective of symmetry. “Highly gapless” QSLs are less stable.

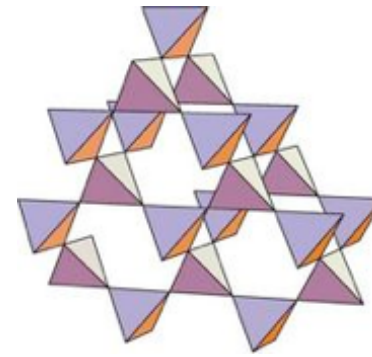
# Spin liquid candidates



# Top experimental platforms

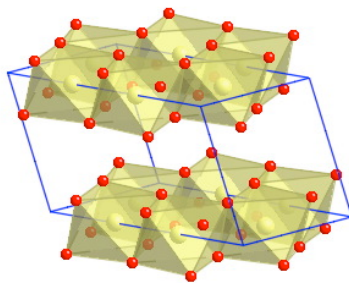


Herbertsmithite



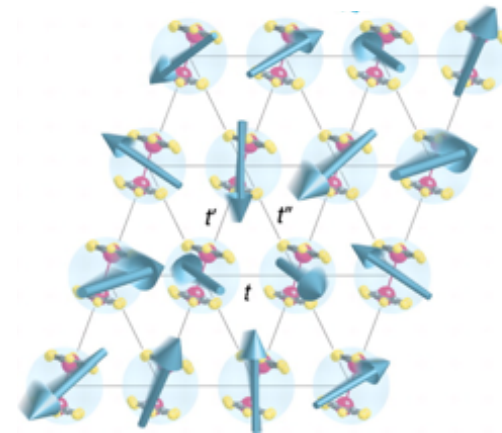
Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>  
...

Quantum spin ice



Na<sub>2</sub>IrO<sub>3</sub>,  
( $\alpha, \beta, \gamma$ )-  
Li<sub>2</sub>IrO<sub>3</sub>  
 $\alpha$ -RuCl<sub>3</sub>

Kitaev materials



organics



# A rough guide to experiments on QSLs

## Does it order?

- NMR line splitting
- $\mu$ SR oscillation
- thermodynamic transition via specific heat, susceptibility
- Bragg peak in neutron/x-ray

## Is there a gap?

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

## Delocalized excitations?

- thermal conductivity
- INS

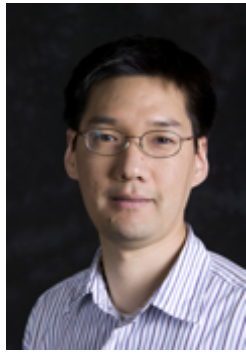
## Structure of excitations?

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

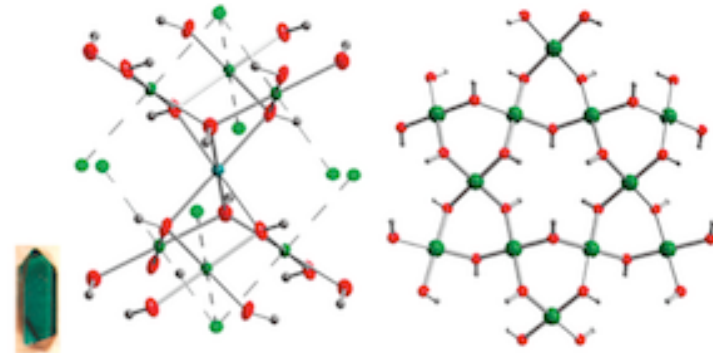
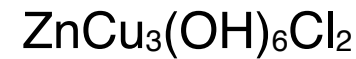
## Exotica

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

# Herbertsmithite

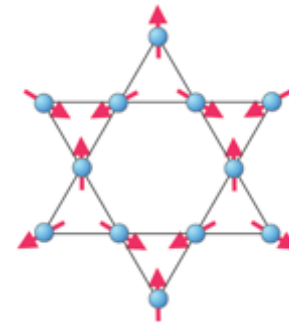


kagomé layers of Cu  
 $S=1/2$  spins, separated  
by non-magnetic Zn



Hamiltonian

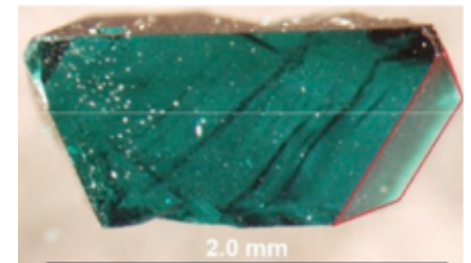
$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \dots$$



$J \sim 200\text{K}$

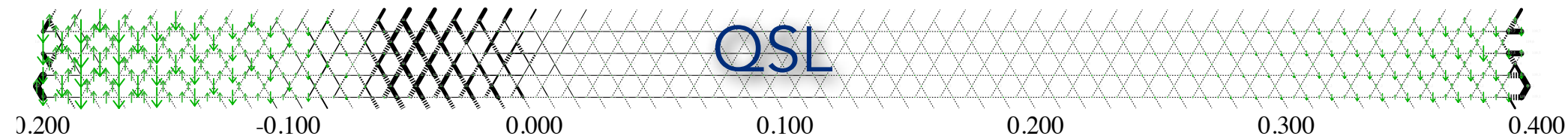
Long efforts by Nocera, Young Lee  
groups produced crystals

beautiful material, but complicated by Cu/Zn site defects

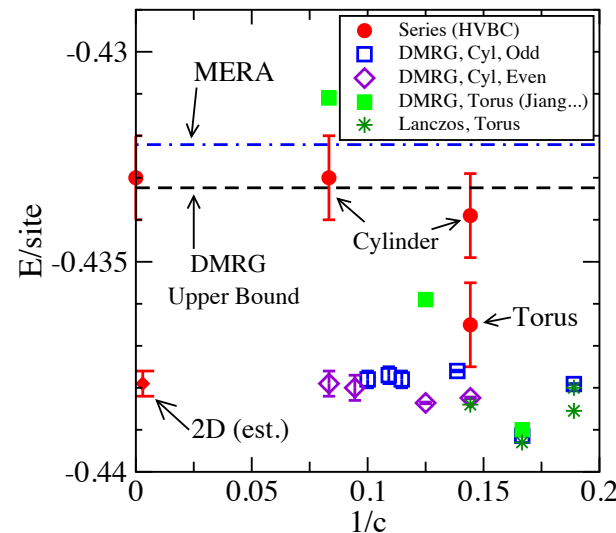


# $S=1/2$ kagomé AF

- Long history - but definitive evidence for QSL by DMRG



© Steve White



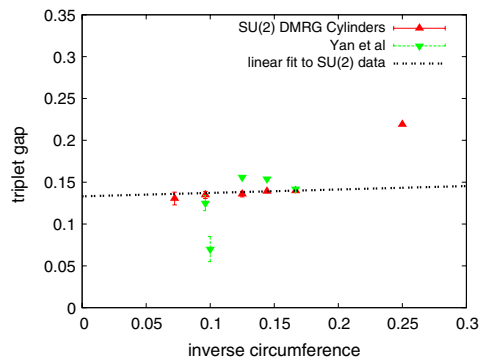
*S. Yan et al, 2010*

many other studies support  
existence of some QSL phase

# $S=1/2$ kagomé AF

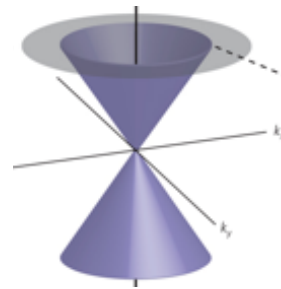
- What kind of QSL?

DMRG



S. Depenbrock *et al*, 2012

partons



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

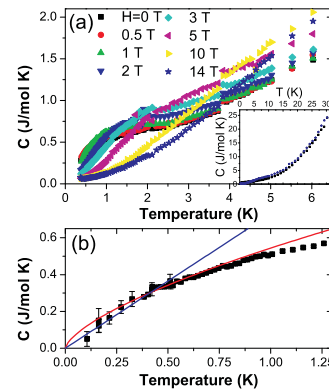
gapped,  
presumably  $Z_2$   
topological QSL

gapless U(1)  
Dirac QSL

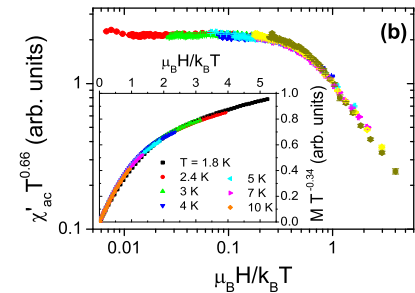
+ various other  
proposals with  
weaker  
quantitative  
support

# Herbertsmithite

Lots of early evidence  
for gaplessness



Helton et al, 2007

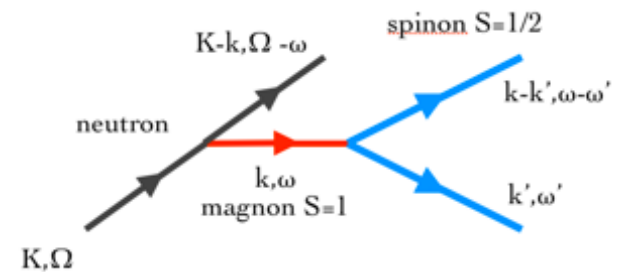
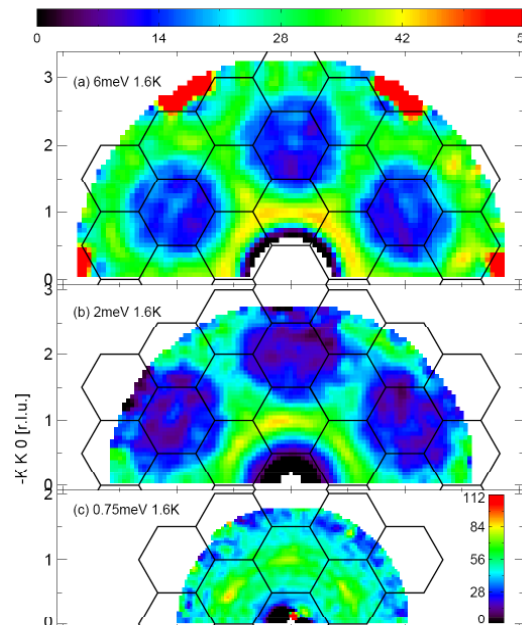


Helton et al, 2010

Single crystal INS

smooth continuum  
scattering

T-H Han et al, 2012



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

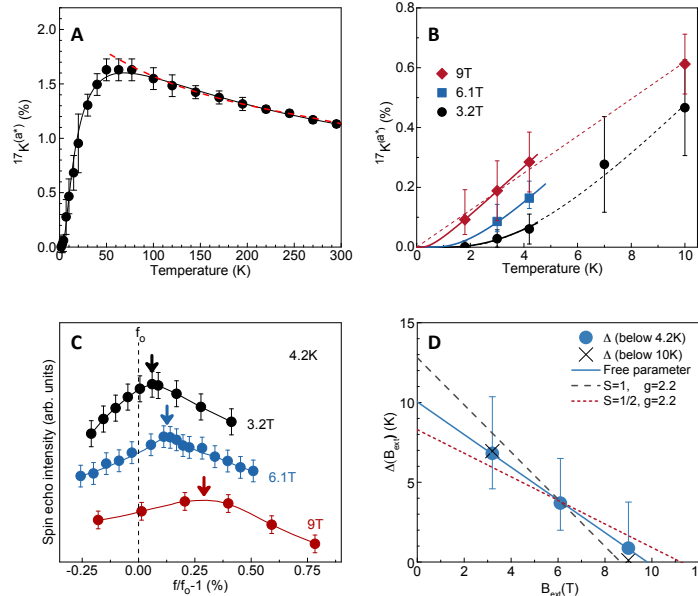


# Herbertsmithite

Single  
crystal NMR

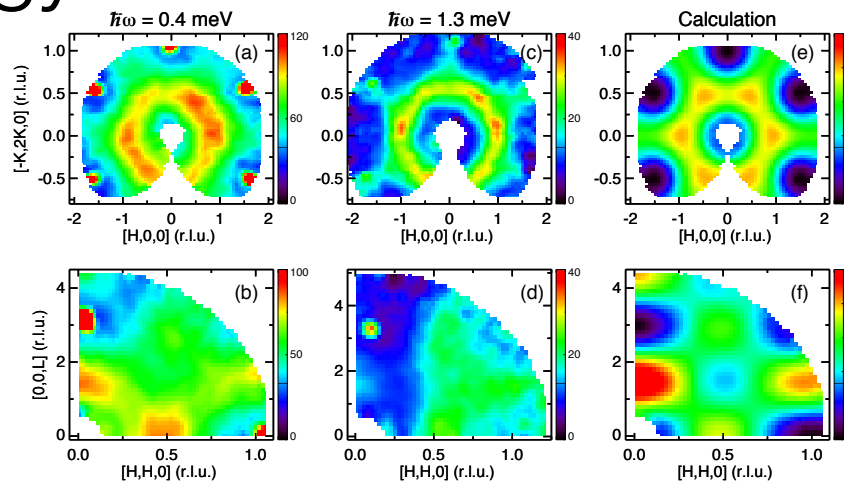
M. Fu *et al*, 2015

c.f. T. Imai lecture



estimate gap  $\sim$   
10K

Low energy INS



claim to separate  
impurity signal  
below 0.7meV

T-H Han *et al*, 2015



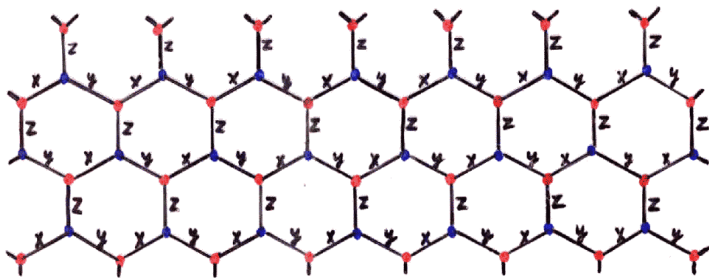
# Kitaev model

Kitaev's honeycomb model

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

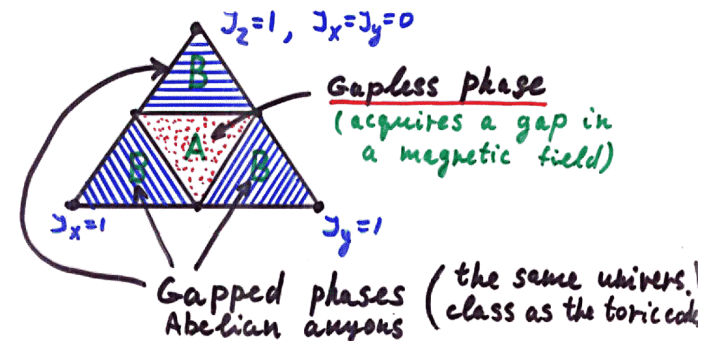
KITP, 2003

## 1. The model



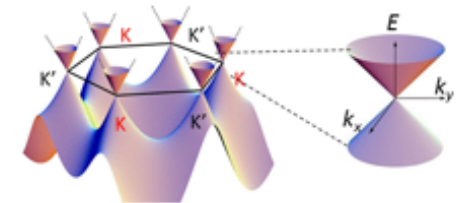
Spin  $\frac{1}{2}$  on each site.

## Phase diagram

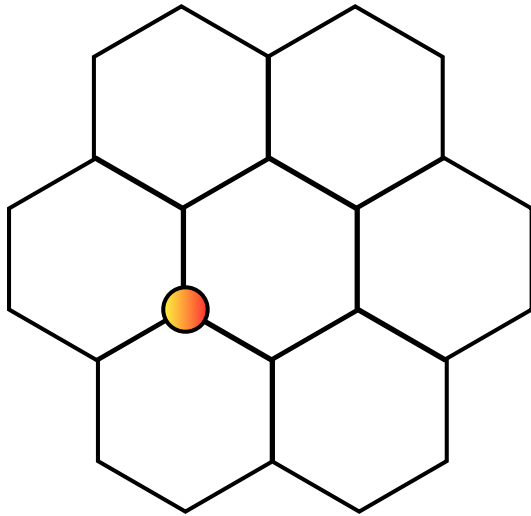


exact parton construction  $\sigma_i^{\mu} = i c_i c_i^{\mu} \quad c_i c_i^x c_i^y c_i^z = 1$

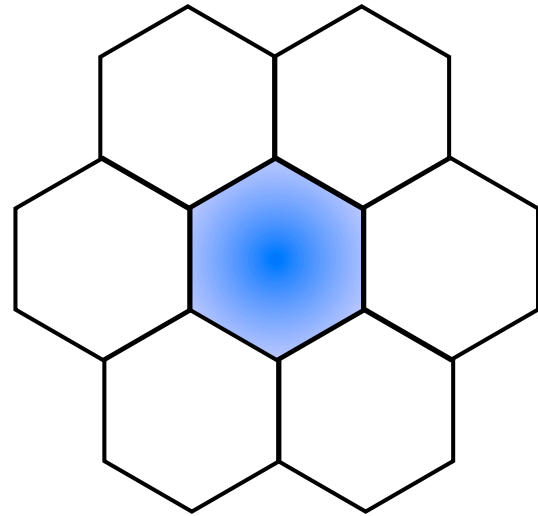
physical Majoranas  $H_m = K \sum_{\langle ij \rangle} i c_i c_j$



# Non-local excitations



Majorana  $\varepsilon$



Flux  $e, m$

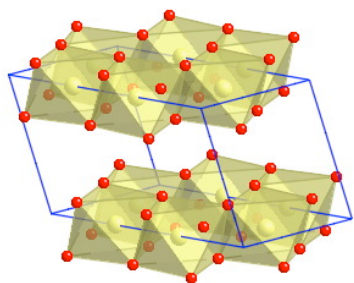
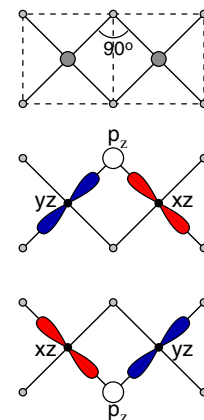
In Kitaev's model:

- Majorana's dispersion  $\sim k$  and gapless
- Fluxes are localized and gapped

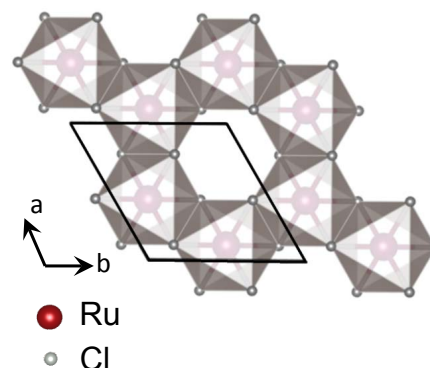
# Kitaev Materials

Jackeli, Khaliullin

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



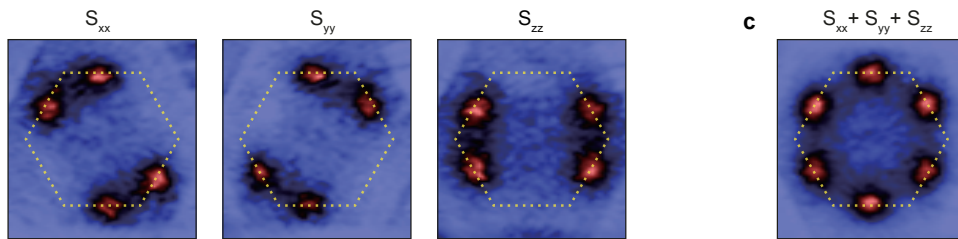
$\text{Na}_2\text{IrO}_3$ ,  
( $\alpha, \beta, \gamma$ )-  
 $\text{Li}_2\text{IrO}_3$



$\alpha\text{-RuCl}_3$

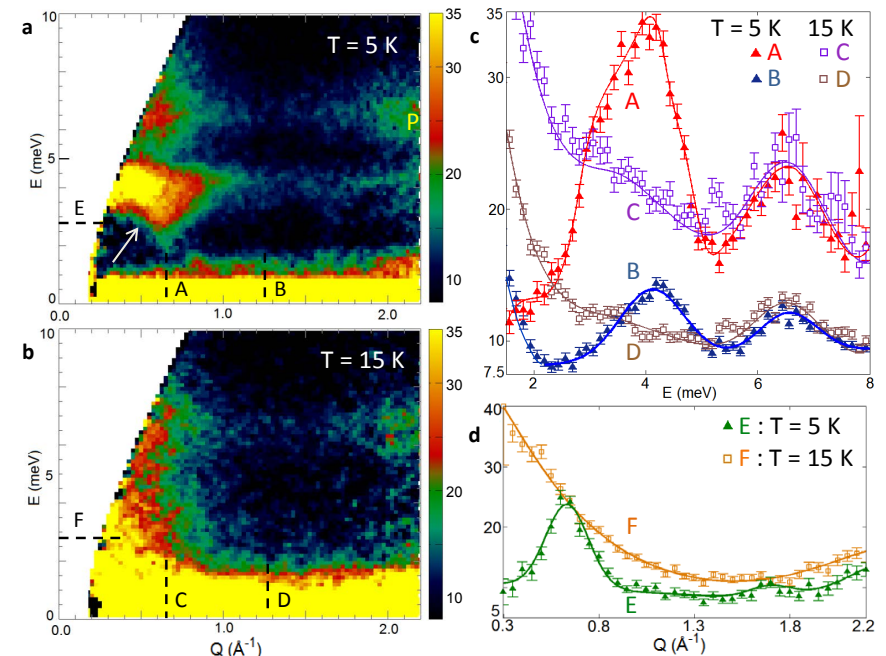
Honeycomb and hyper-honeycomb structures

# Kitaev Materials



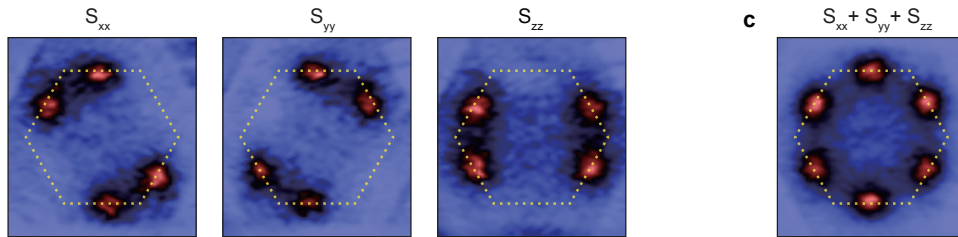
direct evidence for  
direction-dependent  
anisotropic exchange  
from diffuse magnetic  
x-ray scattering in  
 $\text{Na}_2\text{IrO}_3$  (BJ Kim group)

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



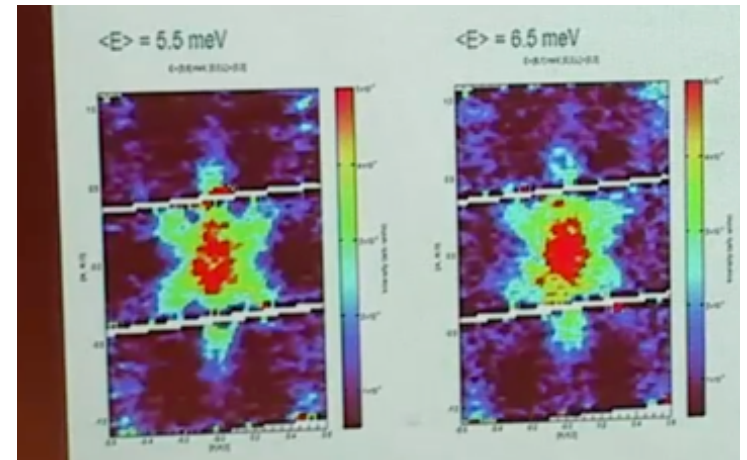
Observation of gapped  
continuum mode persisting  
above  $T_N$  in  $\alpha\text{-RuCl}_3$   
consistent with Majoranas  
(A. Banerjee et al)

# 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)

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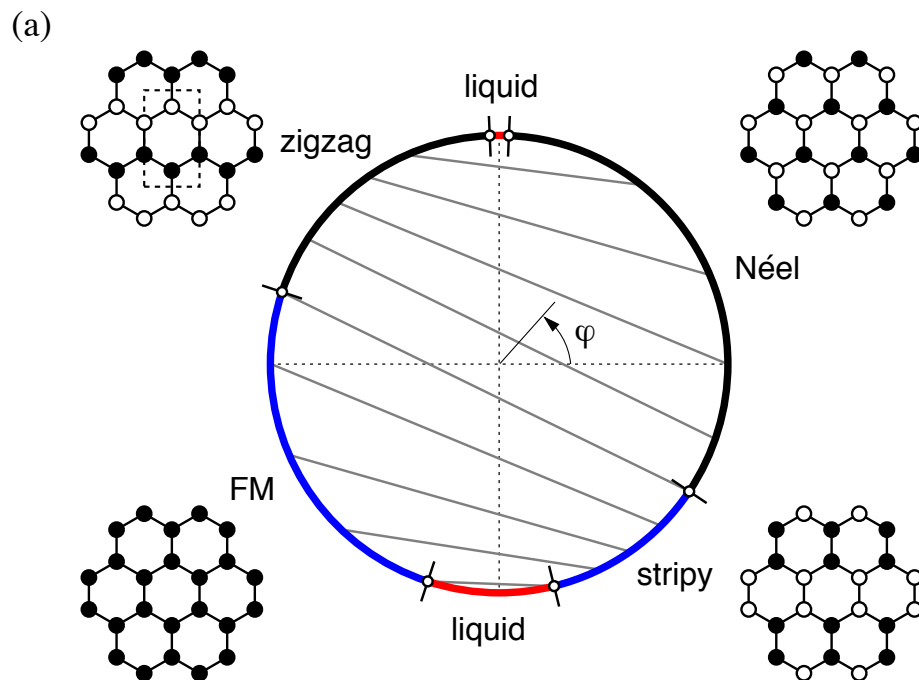
Unpublished single-crystal  
data (A. Banerjee *et al*) in  $\alpha\text{-}$   
 $\text{RuCl}_3$  has expected  
momentum structure for  
Kitaev QSL

# Magnetism

- But...they all order so far

due to additional interactions,  
e.g. Heisenberg

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



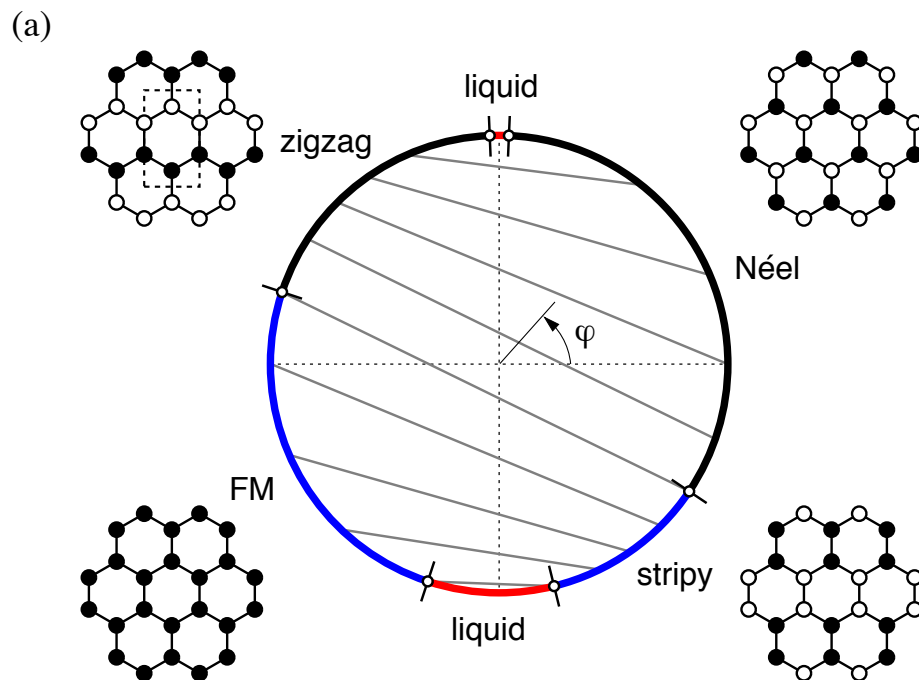
zigzag ordered state  
has been observed in  
 $\text{Na}_2\text{IrO}_3$  and  
incommensurate order  
in  $\text{Li}_2\text{IrO}_3$

# Magnetism

- But...they all order so far

due to additional interactions,  
e.g. Heisenberg

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



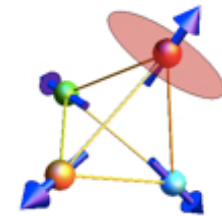
so far no QSL!





# Real quantum spin ice

- Symmetry constrains form of generic Hamiltonian for



local z  
axes

S. Curnoe, 2008 Kramer's doublets

$$H = \left. \begin{aligned} &J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z \\ &-J_{\pm} \sum_{\langle i,j \rangle} (S_i^+ S_j^- + S_i^- S_j^+) \end{aligned} \right\} \begin{array}{l} \text{these terms give the} \\ \text{earlier model} \end{array}$$

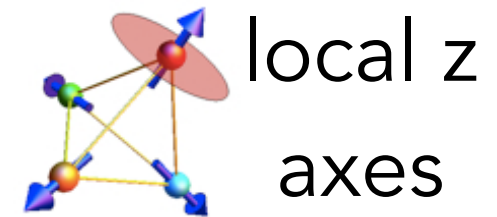
$$+ J_{z\pm} \sum_{\langle i,j \rangle} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j]$$

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

# Hamiltonian

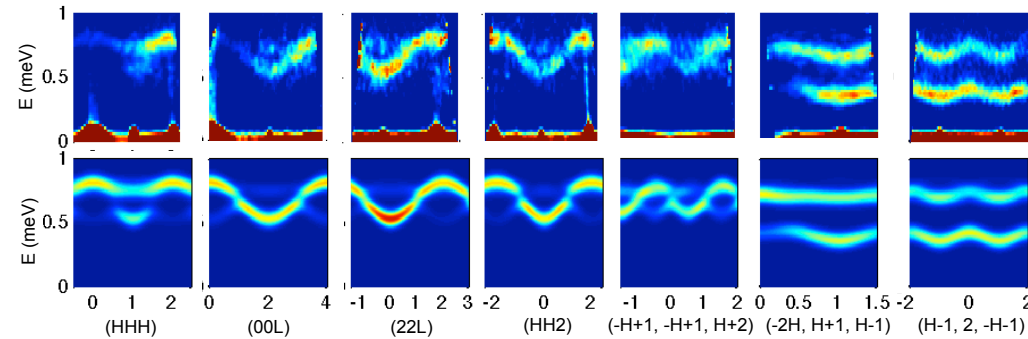


- Symmetry constrains form of generic Hamiltonian for Kramer's doublets



L. Savary et al, 2012

$$\begin{aligned}
 H = & J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z \\
 & - J_{\pm} \sum_{\langle i,j \rangle} (S_i^+ S_j^- + S_i^- S_j^+) \\
 & + J_{z\pm} \sum_{\langle i,j \rangle} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \\
 & + J_{\pm\pm} \sum_{\langle i,j \rangle} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-)
 \end{aligned}$$



$\text{Er}_2\text{Ti}_2\text{O}_7$

XY-like

$$J_{zz} = -2.5 \pm 1.8 \times 10^{-2} \text{ meV}$$

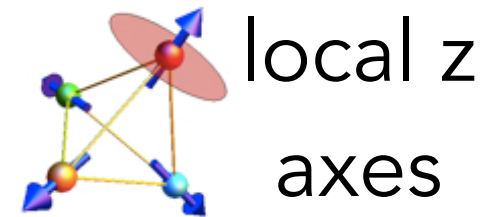
$$J_{z\pm} = -0.88 \pm 1.5 \times 10^{-2} \text{ meV}$$

$$J_{\pm} = 6.5 \pm 0.75 \times 10^{-2} \text{ meV}$$

$$J_{\pm\pm} = 4.2 \pm 0.5 \times 10^{-2} \text{ meV}$$

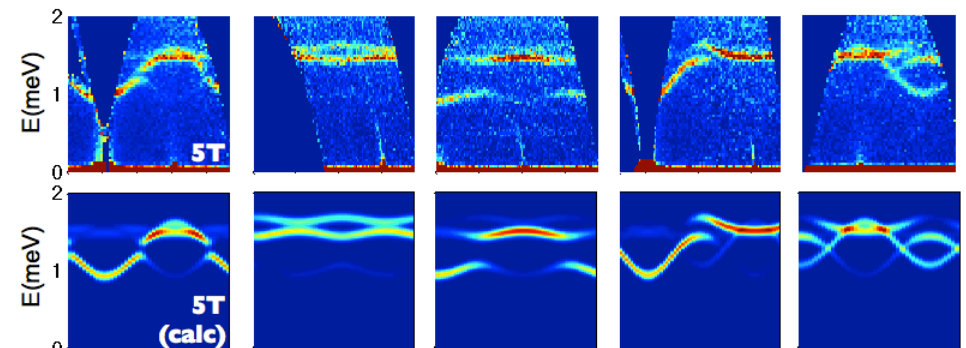
# Hamiltonian

- Symmetry constrains form of generic Hamiltonian for Kramer's doublets



$$\begin{aligned}
 H = & J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z \\
 & - J_{\pm} \sum_{\langle i,j \rangle} (S_i^+ S_j^- + S_i^- S_j^+) \\
 & + J_{z\pm} \sum_{\langle i,j \rangle} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \\
 & + J_{\pm\pm} \sum_{\langle i,j \rangle} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-)
 \end{aligned}$$

K. Ross et al, 2011



$\text{Yb}_2\text{Ti}_2\text{O}_7$   
QSI

$$J_{zz} = 0.17 \pm 0.04 \text{ meV}$$

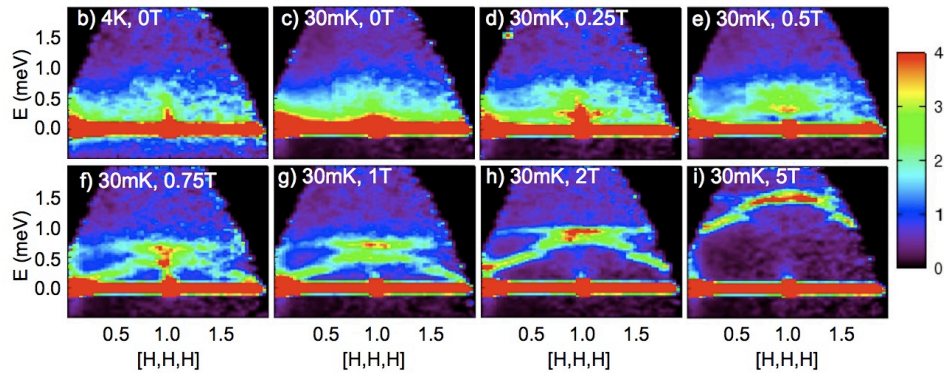
$$J_{z\pm} = 0.14 \pm 0.01 \text{ meV}$$

$$J_{\pm} = 0.05 \pm 0.01 \text{ meV}$$

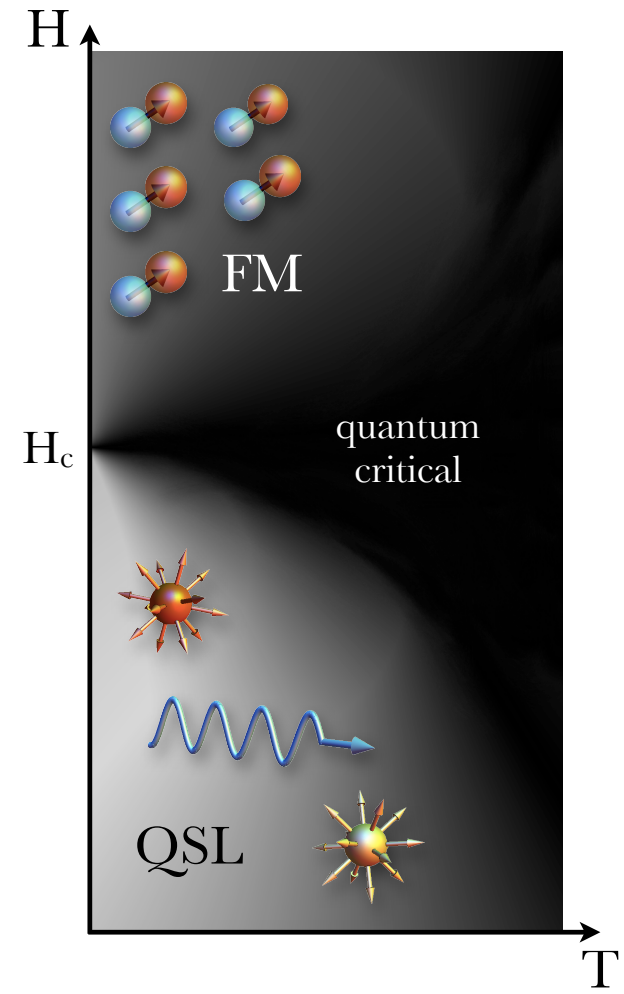
$$J_{\pm\pm} = 0.05 \pm 0.01 \text{ meV}$$

# $\text{Yb}_2\text{Ti}_2\text{O}_7$ ?

missing  
magnon?

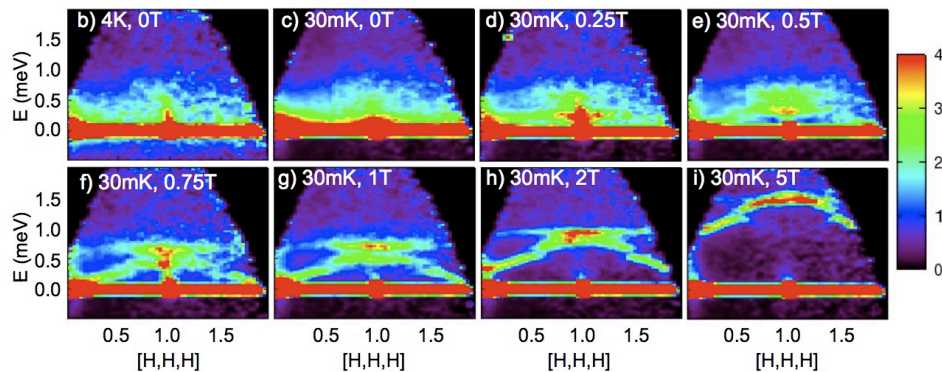


K.A. Ross *et al* (2009)



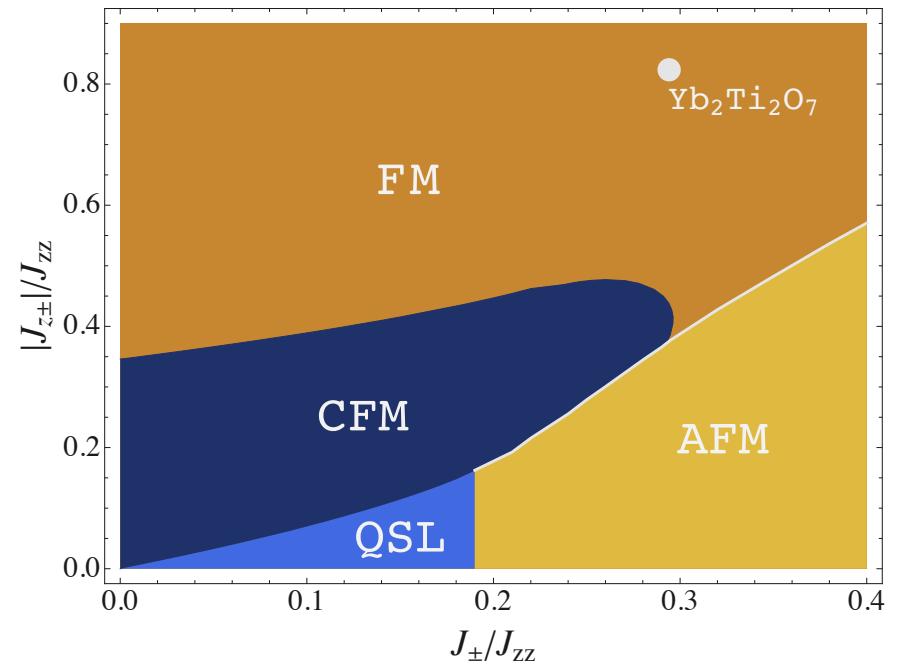
# Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>?

missing  
magnon?



K.A. Ross *et al* (2009)

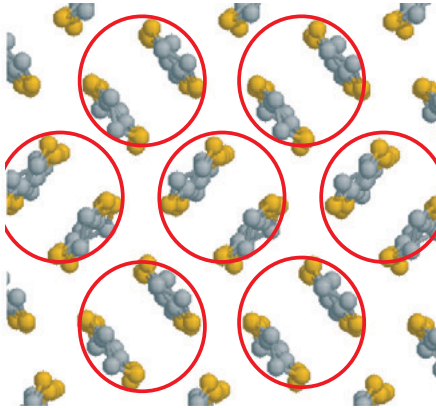
L. Savary + LB, 2012



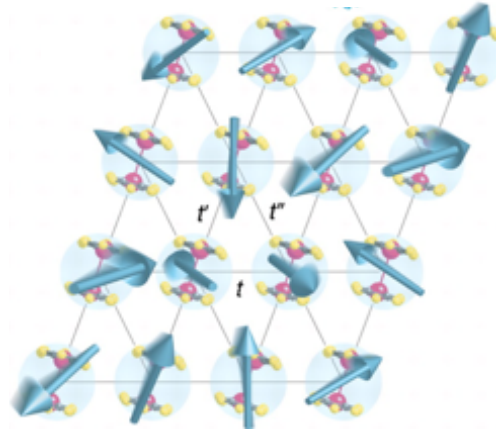
I tend to think recent evidence points away from the U(1) QSL and toward an unconventional FM. But this is still an unfinished story.

$J_{\pm\pm}$  might help?  
disorder effects?  
other materials?

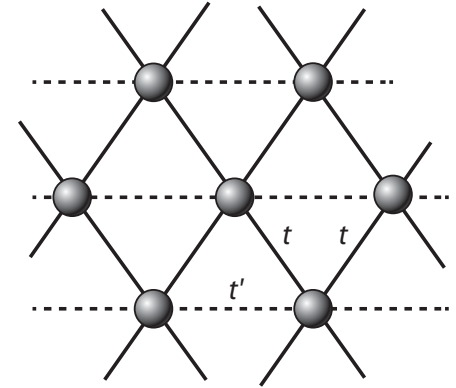
# Organics



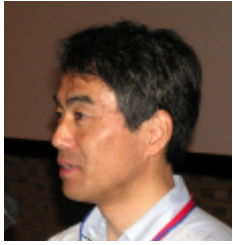
$\kappa\text{-(ET)}_2\text{X}$



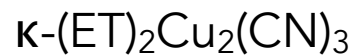
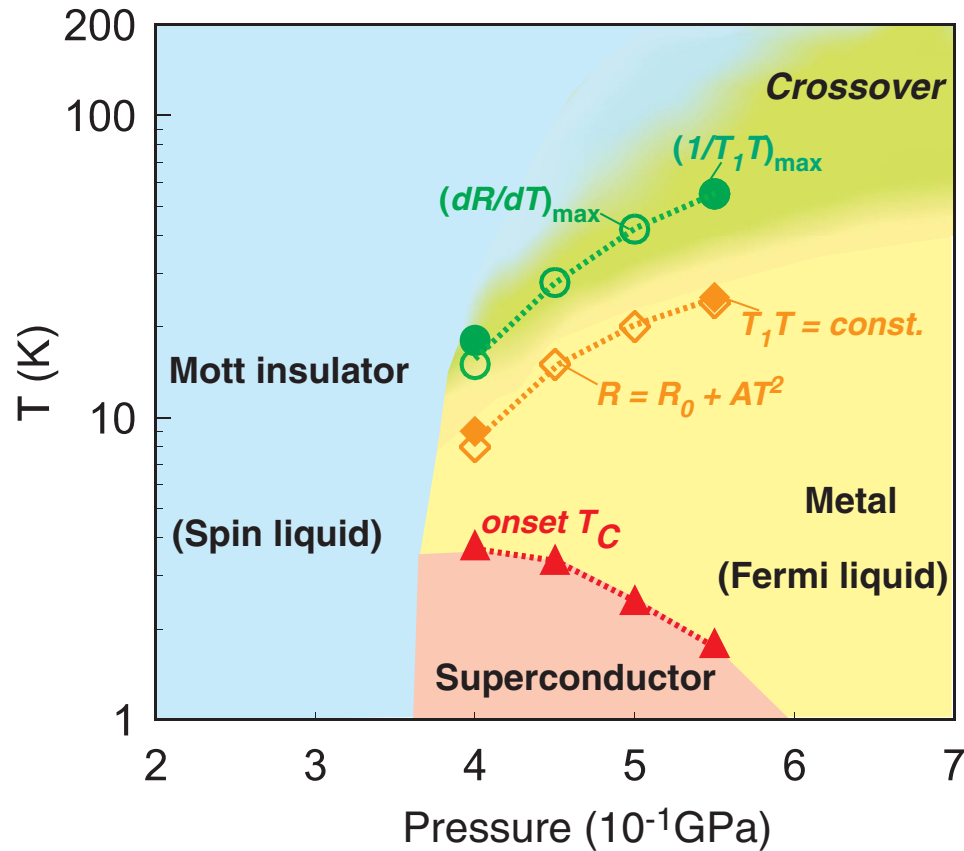
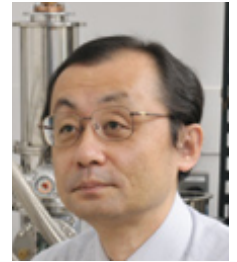
$\beta'\text{-Pd(dmit)}_2$



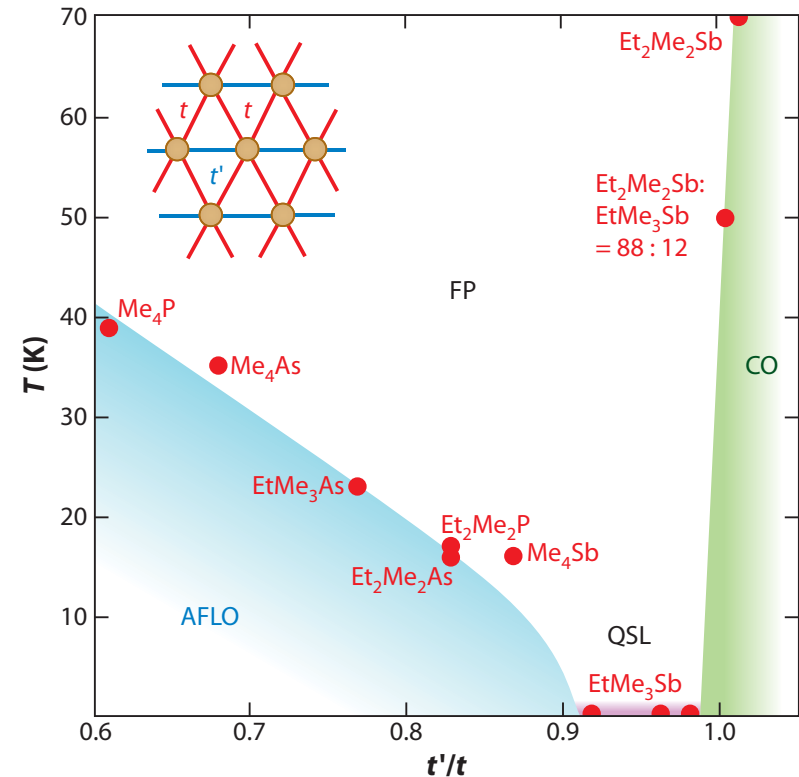
- Molecular materials which behave as effective triangular lattice  $S=1/2$  antiferromagnets with  $J \sim 250\text{K}$
- significant charge fluctuations



# Organics

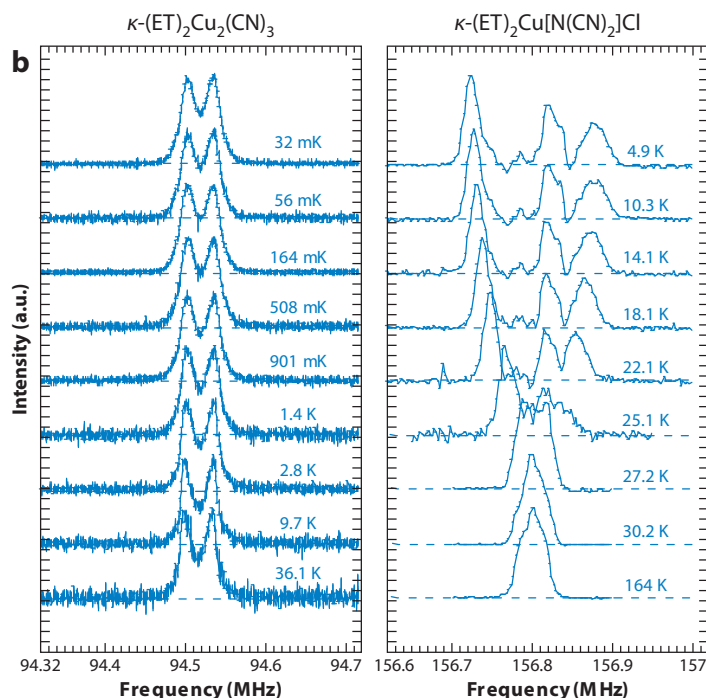


K. Kanoda group (2003-)

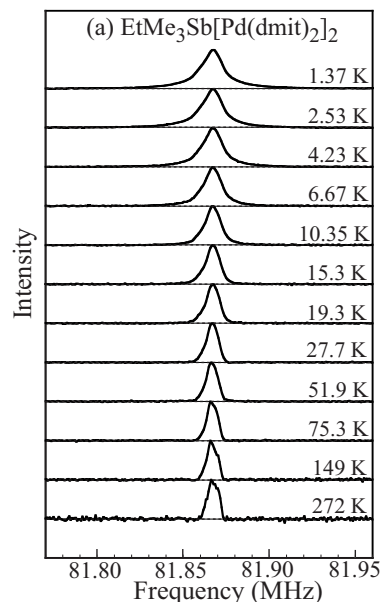


R. Kato group (2008-)

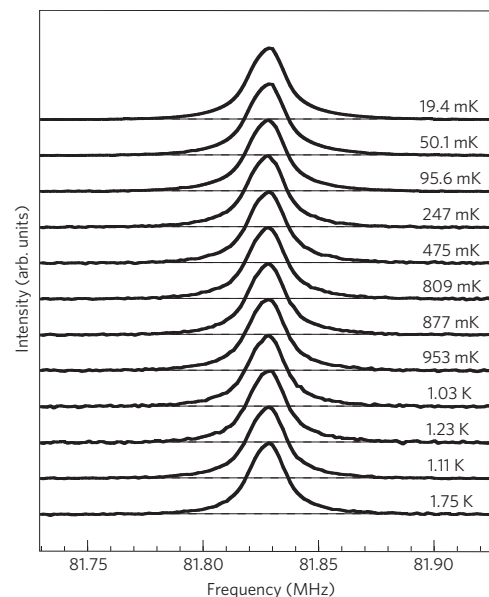
# NMR lineshapes



Y. Shimizu  $^1\text{H}$  NMR  
*et al*, 2003



T. Itou *et al*,  
 2008, 2010



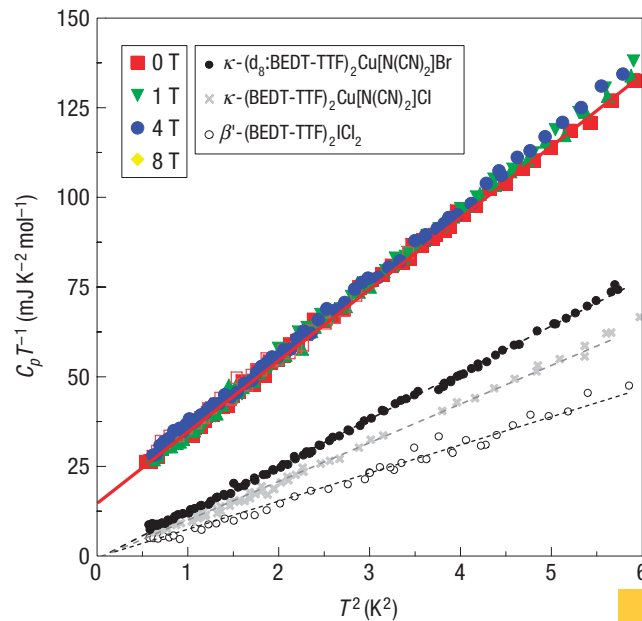
$^{13}\text{Cs}$  NMR

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

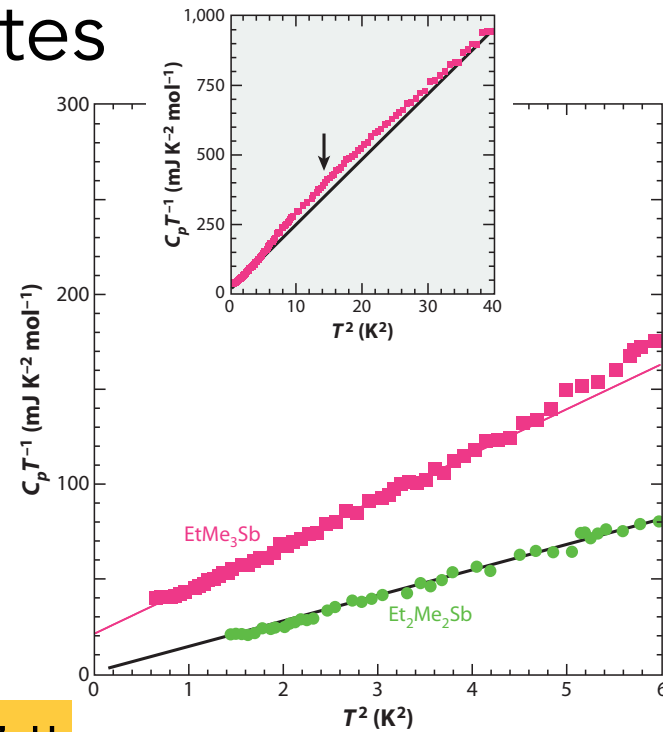


# Specific Heat

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



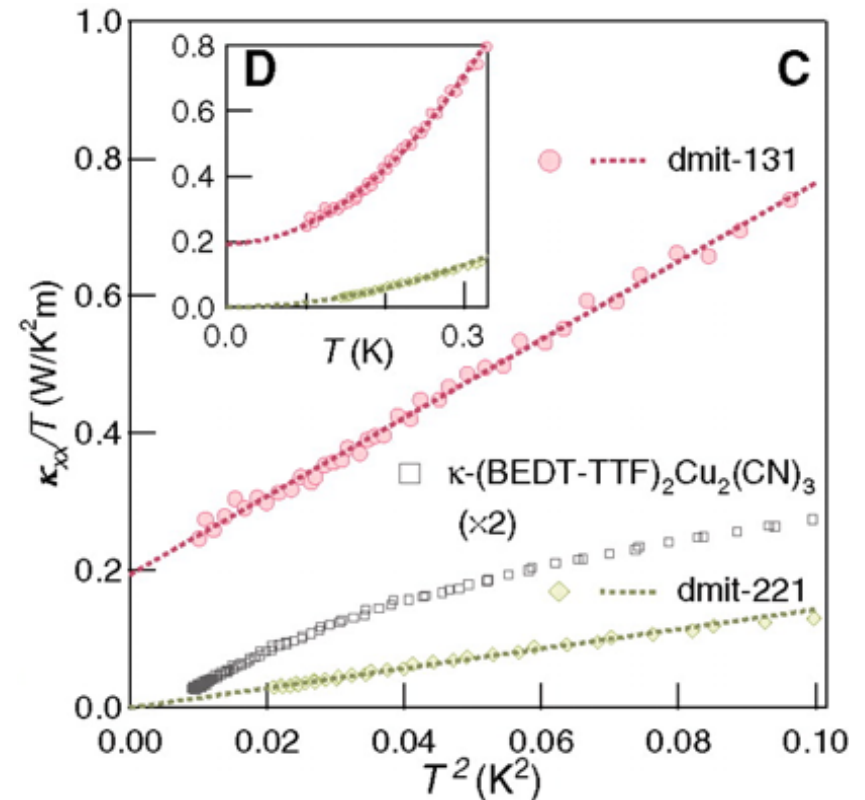
$\gamma_{Cu} \sim 0.7$  !!



S. Yamashita *et al*, 2008

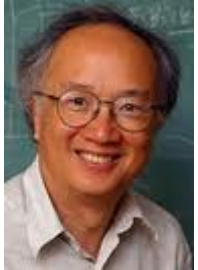
# Thermal conductivity

- Huge linear thermal conductivity indicates the gapless excitations are propagating, at least in dmit
- Estimate for a *metal* would correspond to a mean free path  $l \sim 1 \mu\text{m} \approx 1000 a$  !

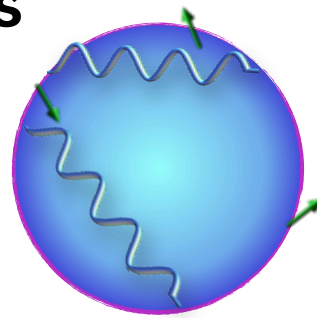


M. Yamashita *et al*, 2010

# Organics - Theory



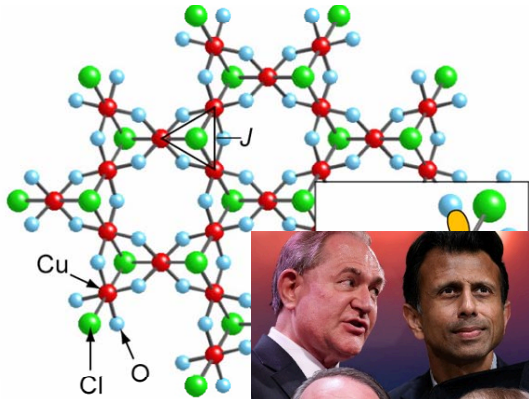
- RVB/QSL state:
  - Motrunich, Lee+Lee: (2005) “uniform RVB”
  - this is a kind of RVB state with very many (maybe a maximal number of?) long-range VBs
  - It is described by a “**Fermi sea**” of spinons coupled to a U(1) gauge field
- Good variational energy for triangular lattice Hubbard model



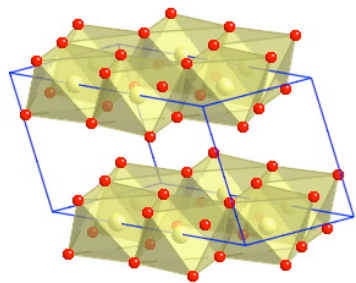
# Organics: issues

- Why these very small set of materials?
- Spatial homogeneity?
- Indications of phase transitions. Gaps opening?  
Charge ordering?
- Quantitative inconsistencies with expected scaling behavior from theory (c.f.  $C \sim T^{2/3}$  etc.)
- Large isotope effects. Role of molecular rotations?
- Almost all experimental checks of QSL are limited to  $T < 5\text{K}$ , and many are not directly tied to spins, while  $J \sim 200\text{-}300\text{K}$ . Hampered by nature of materials.

# New candidates are desirable



Herbert



Kitaev materials



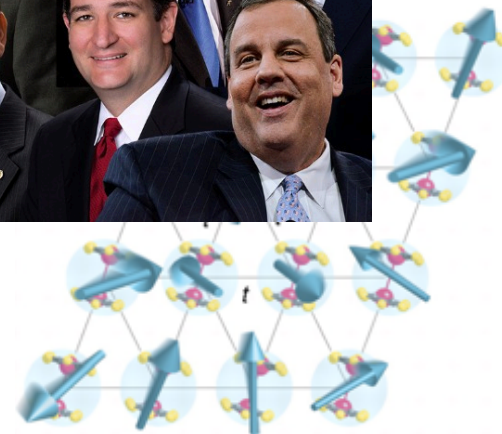
$\text{Li}_2\text{IrO}_3$   
 $\alpha\text{-RuCl}_3$



$\text{Yb}_2\text{Ti}_2\text{O}_7$

...

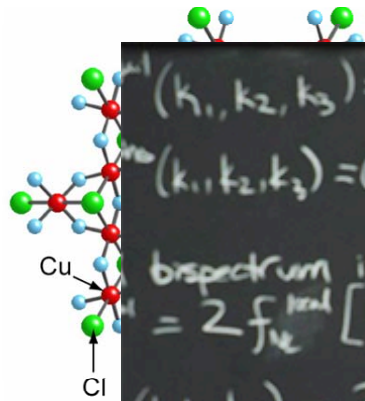
spin ice



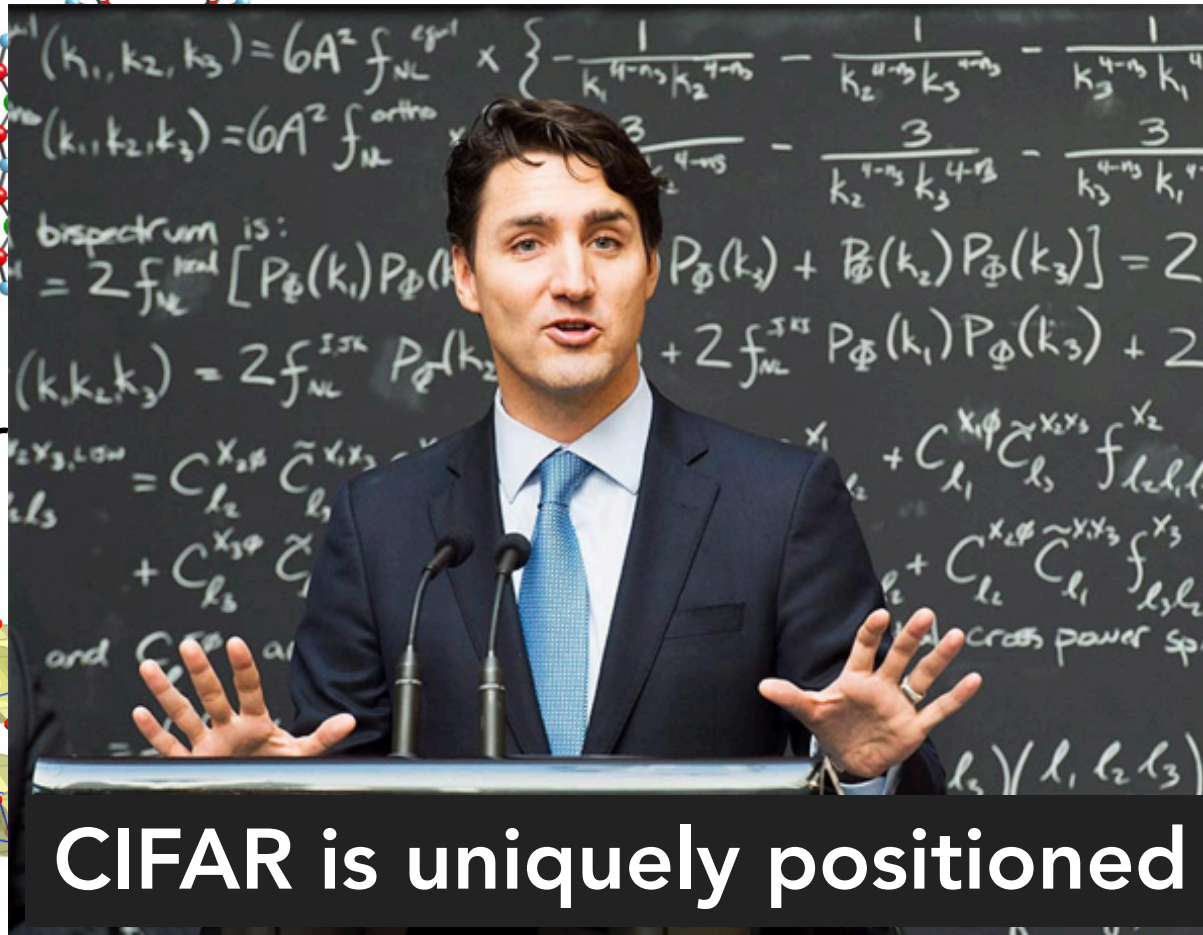
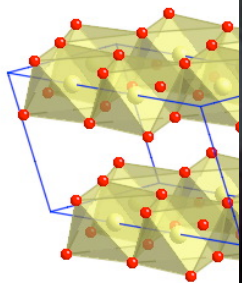
organics



# New candidates are desirable



Here



**CIFAR is uniquely positioned**

$\text{Yb}_2\text{Ti}_2\text{O}_7$

...

spin ice

Kitaev materials

organics

# Theory: Frontiers

## New phases

- Fractal spin liquids (Haah++) in 3d
- SPT phases
- Quenched disorder

## Fundamental problems

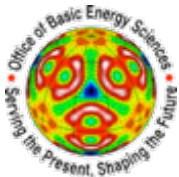
- QSLs with strongly coupled matter-gauge theory
- QCPs to/from QSL phases
- Out of equilibrium
- Doping

## Reality

- Devise experimental protocols to reveal quantum non-locality of QSLs
- Computational methods: less bias, reliability of variational methods, beyond ground states

# Thanks for your attention

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



GORDON AND BETTY  
**MOORE**  
FOUNDATION

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