# Recent development of quantum spin liquids

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### Opportunity for students and postdocs

- My research group is looking for graduate students and postdocs
- Our postdocs and visiting professors are generously funded.





### Outline

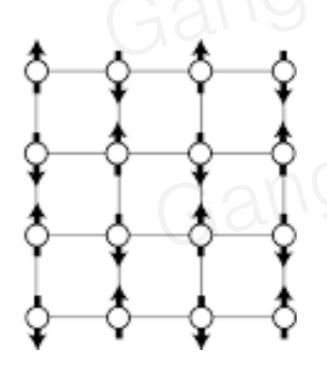
- A general introduction to quantum spin liquids
- Spinon Fermi surface U(1) quantum spin liquid
- Rare earth triangular lattice quantum spin liquid and experiment prediction
- Control spinons in a quantum spin ice U(1) quantum spin liquid

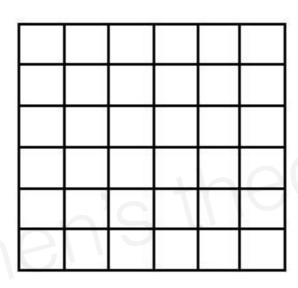


## Neel vs Landau (1930-40s)



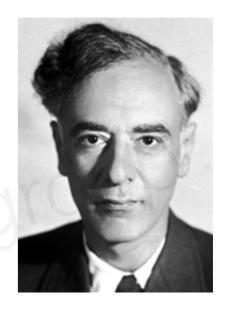
Neel





$$H = J \sum_{\langle ij 
angle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Spin singlet 
$$=\frac{1}{\sqrt{2}}\left[\uparrow \downarrow - \downarrow \uparrow \right]$$



Landau





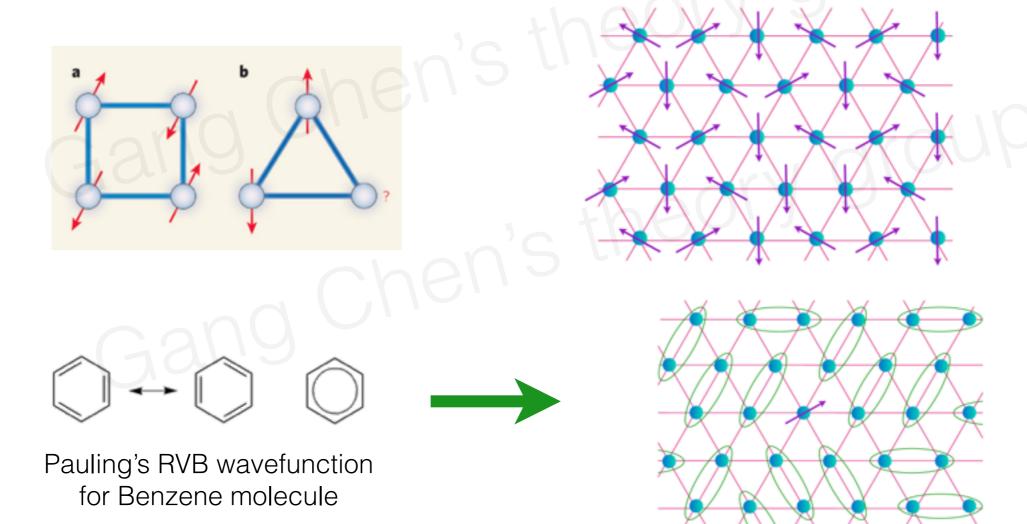
P. W. Anderson

### The idea of quantum spin liquid (1973)

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR?\*

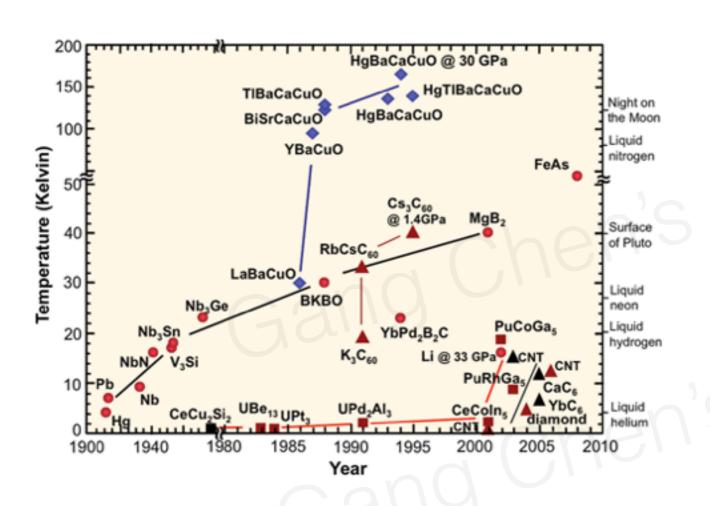
P. W. Anderson

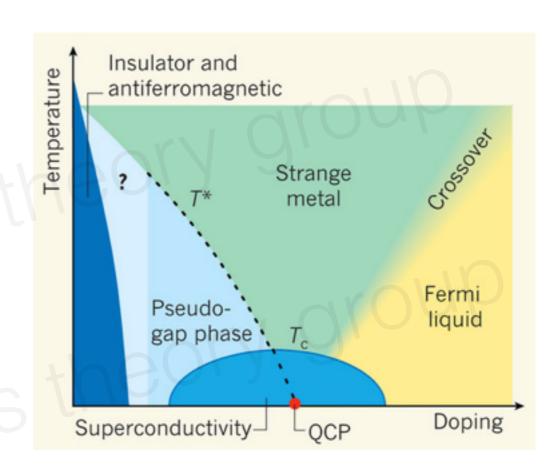
type would be insulating; it would represent an alternative state to the Néel antiferromagnetic state for S = 1/2. An estimate of





### High temperature superconductivity (1986)





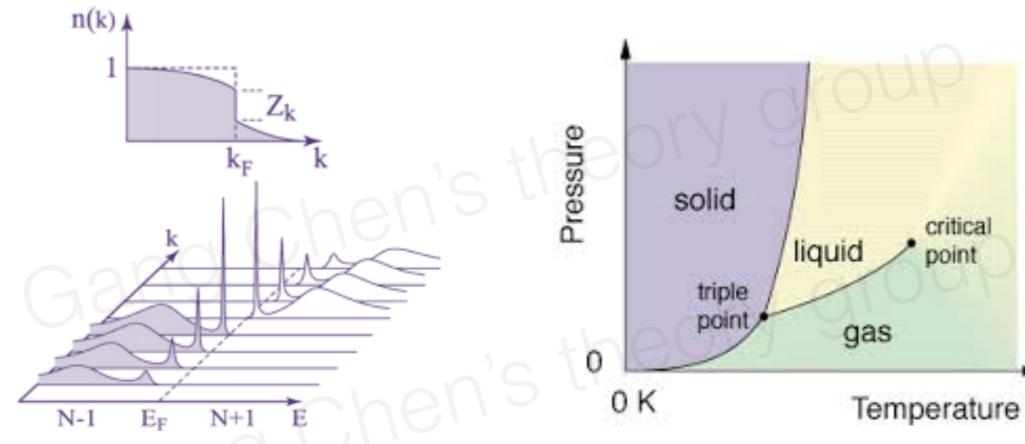
The idea is to view Mott insulator (QSL) as the parent state of high-temperature superconductor. In the QSL, there are preformed Cooper pairs. Doping it allows to Cooper pairs to condense and lead to superconductivity.





Landau

## Two milestones of 20th century condensed matter physics



Landau Fermi liquid theory

Landau symmetry breaking theory

These two paradigms break down after the discovery of fractional quantum Hall effect in 1980s.



### Quantum spin liquid

 Quantum spin liquid is a new quantum phase of matter, and cannot be characterized by Landau symmetry breaking, instead by emergent gauge structure and deconfined fractionalized excitations.

- QSL, its existence, is very clear, at least at the level of theory.
  - Exactly solvable model with QSL ground state: e.g. Kitaev model and extension.
  - Classification of QSLs: many distinct symmetry enriched QSLs (XG Wen etc).
  - Numerical solutions: DMRG, QMC, exact diagonalization, etc.

QSL is **robust** against any local perturbation. So it should exist in Nature!



### QSL: existing experiments

2D triangular and Kagome lattice

organics: kappa-(BEDT-TTF)2Cu2(CN)3, EtMe3Sb[Pd(dmit)2]2, kappa-H3(Cat-EDT-TTF)2 herbertsmithite (ZnCu3(OH)6Cl2), Ba3NiSb2O9, Ba3CuSb2O9, LiZn2Mo3O8, ZnCu3(OH)6Cl2 volborthite (Cu3V2O7(OH)2), BaCu3V2O3(OH)2, [NH<sub>4</sub>]<sub>2</sub>[C<sub>7</sub>H<sub>14</sub>N][V<sub>7</sub>O<sub>6</sub>F<sub>18</sub>], Na2IrO3, CsCu2Cl4, CsCu2Br4, NiGa2S4, He-3 layers on graphite, etc

- 3D pyrochlore, hyperkagome, FCC lattice, diamond lattice, etc
   Na4Ir3O8, IrO2, Ba2YMoO6, Yb2Ti2O7, Pr2Zr2O7, Pr2Sn2O7, Tb2Ti2O7, Nd2Zr2O7, FeSc2S4, etc
- Ultracold atom and molecules on optical lattices: temperature is too high now.

Some candidate materials have already been ruled out.

Not being a QSL does not necessarily mean the physics is not interesting!



• Spinon Fermi surface U(1) quantum spin liquid



### Any guiding rule to find QSL? Not really.

Frustrated lattice? Honeycomb Kitaev model.

Frustrated interaction? We do not really know unless we identify the interaction.

Low dimensionality? 3D lattice also has QSL.

Odd electrons per cell? Many QSLs have even electrons per cell.



Lieb



Oshikawa



Hastings



Vishwanath

- Hastings-Oshikawa-Lieb-Shultz-Mattis theorem.
- Recent extension to spin-orbit coupled insulators (Watanabe, Po, Vishwanath, Zaletel, PNAS 2016).



### A rare-earth triangular lattice quantum spin liquid: YbMgGaO4



Dr. Yuesheng Li
Renmin Univ -> MPI, Germany

This part is in collaboration with experimentalists

Dr. Yuesheng Li (Renmin Univ, Beijing)
Prof. Qingming Zhang (Renmin Univ, Beijing)

Wei Tong (High Magnetic field Lab, Hefei)

Pi Li (High Magnetic field Lab, Hefei)

Juanjuan Liu (Renmin Univ, Beijing)

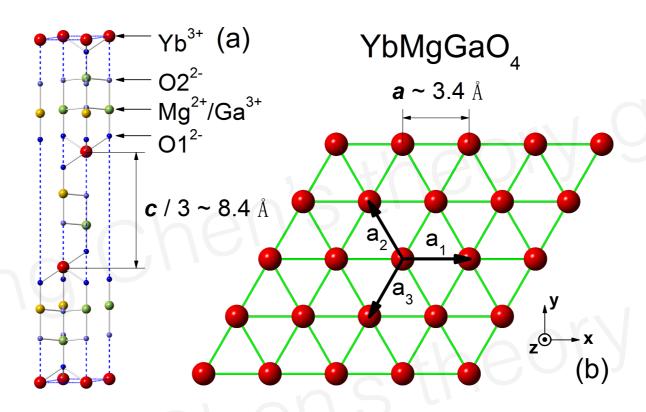
Zhaorong Yang (Institute of Solid-State Physics, Hefei)

Xiaoqun Wang (Renmin, Shanghai Jiaotong)

YS Li, **GC**\*,...., QM Zhang\* PhysRevLett 2015



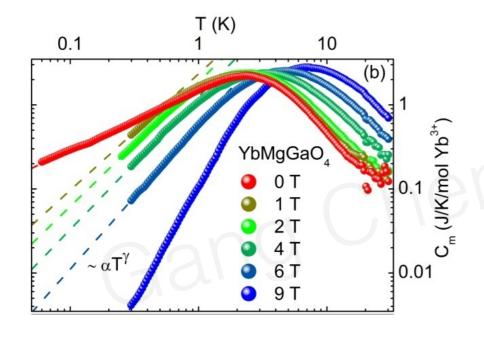
### A rare-earth triangular lattice quantum spin liquid: YbMgGaO4

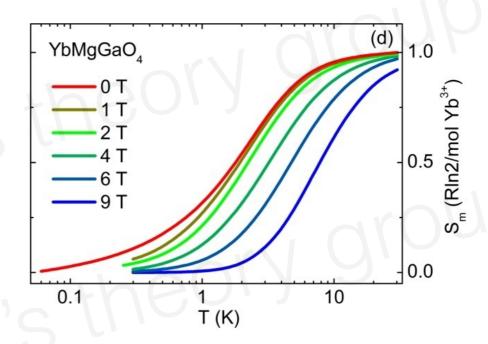


- This is the **first** strong spin-orbit coupled QSL with odd number of electrons and effective spin-1/2.
- It is the **first** clear observation of T<sup>2/3</sup> heat capacity.
- We understand the microscopic Hamiltonian and the physical mechanism.



### YbMgGaO4





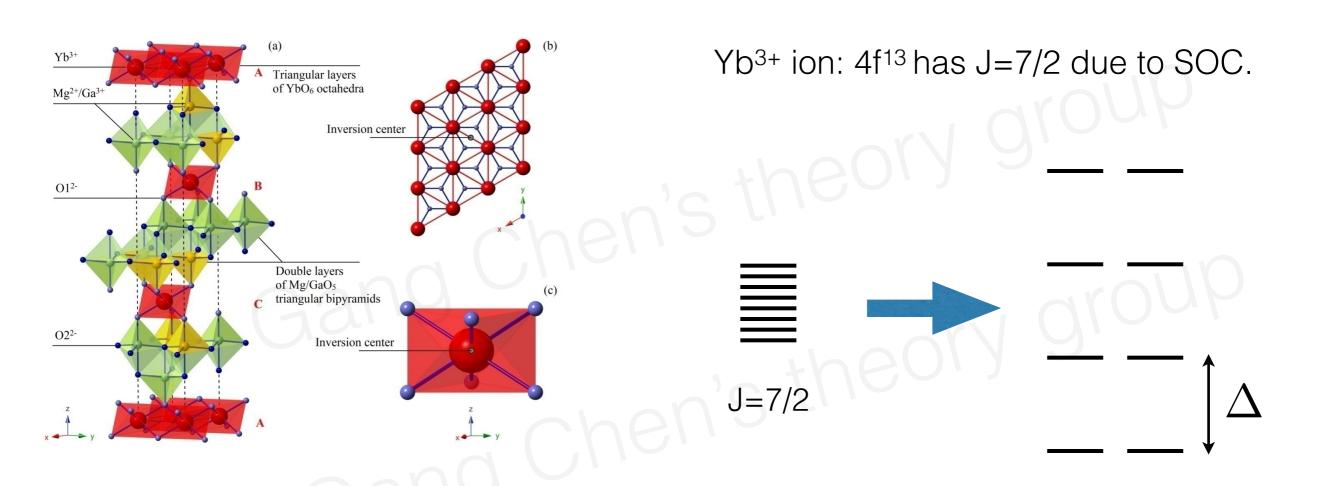
observation of T<sup>2/3</sup> heat capacity

• Entropy: effective spin-1/2 local moments

My proposal for ground state: spinon Fermi surface U(1) QSL.



## Microscopics



At  $T \ll \Delta$ , the only active DOF is the ground state doublet that gives rise to an effective spin-1/2.



### Can this kind of system support a QSL ground state? Yes.

### Filling constraints for spin-orbit coupled insulators in symmorphic and non-symmorphic crystals

Haruki Watanabe,<sup>1</sup> Hoi Chun Po,<sup>1</sup> Ashvin Vishwanath,<sup>1,2</sup> and Michael P. Zaletel<sup>3</sup>

May 2015

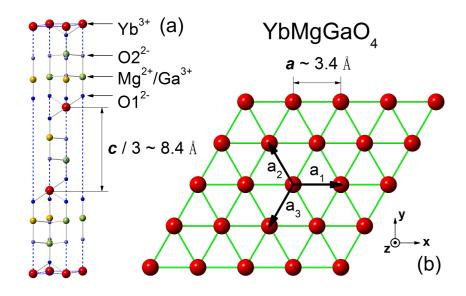
and a crystalline lattice or a magnetic field. Mott insulators are a particularly interesting class, with an odd number of electrons in each unit cell. Their low energy physics is captured by a spin model with an odd number of S=1/2 moments in the unit cell. A powerful result due to Lieb, Schultz, and Mattis in  $1D^1$ , later extended to higher dimensions by Hastings and Oshikawa<sup>2,3</sup>, holds that if all symmetries remain unbroken, the ground state must be 'exotic' - such as a Luttinger liquid in 1D, or a quantum spin liquid in higher dimensions, with fractional 'spinon' excitations. These exotic states cannot be represented as simple product states, as a consequence of long ranged quantum entanglement. This general re-

tirely different theoretical approaches are needed. We argue that if a spin-orbit coupled insulator at odd filling is time-reversal symmetric, its ground state must, in a precise sense, be exotic. We introduce two theoreti-



### What is the physical origin of the QSL?

4f electron is very localized, and dipolar interactions weak.



PhysRevLett, 2015

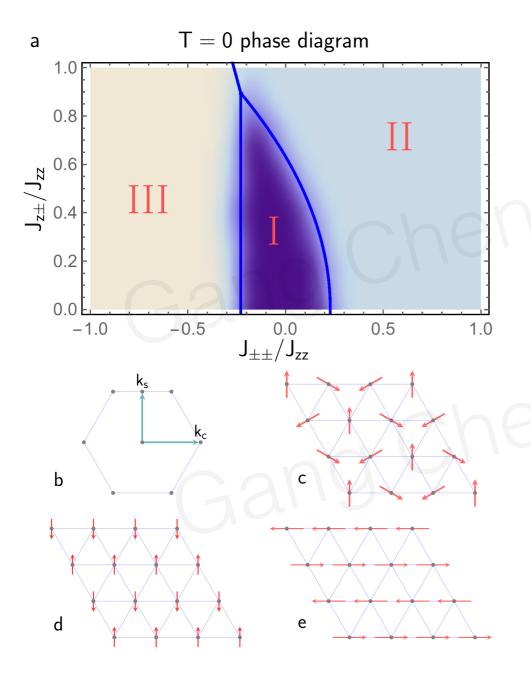
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ J_{zz} S_i^z S_j^z + J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm\pm} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-) - \frac{iJ_{z\pm}}{2} (\gamma_{ij}^* S_i^+ S_j^z - \gamma_{ij} S_i^- S_j^z + \langle i \leftrightarrow j \rangle) \right], \quad (1)$$

where  $S_i^{\pm} = S_i^x \pm i S_i^y$ , and the phase factor  $\gamma_{ij} = 1, e^{i2\pi/3}, e^{-i2\pi/3}$  for the bond ij along the  $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$  direction (see Fig. 1), respectively. This generic Hamil-

The spin-1/2 XXZ model supports conventional order. (Yamamoto, etc, PRL 2014)

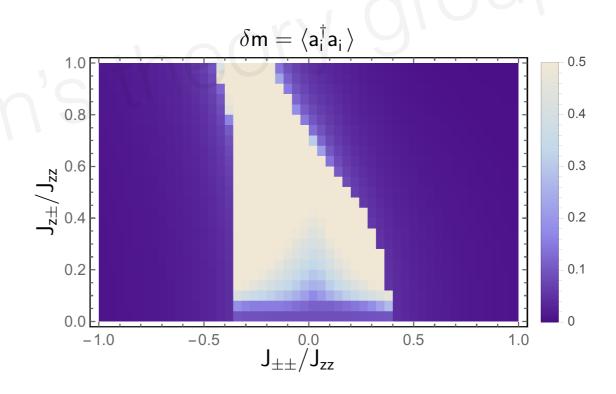


## Anisotropic spin interaction could potentially stabilize QSL.





Yao-Dong Li Dept of Computer Sciences Fudan University

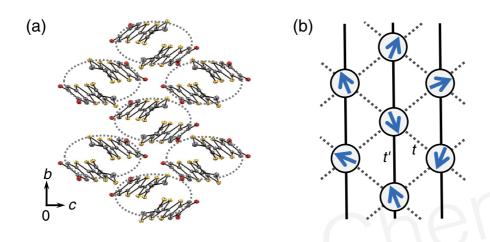




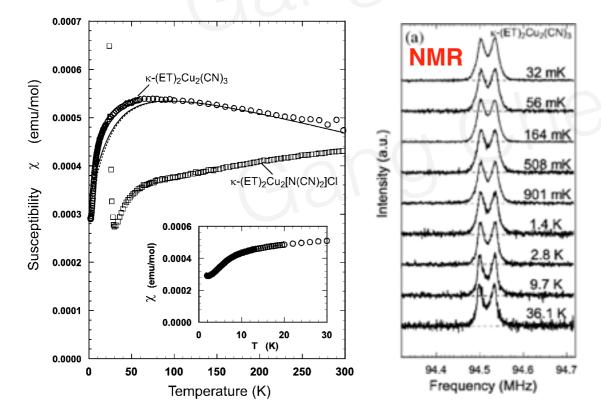
## Spinon Fermi surface U(1) QSL in organic magnets?



Kanoda



kappa-(BEDT-TTF)2Cu2(CN)3, EtMe3Sb[Pd(dmit)2]2, kappa-H3(Cat-EDT-TTF)2 **a new one!** 



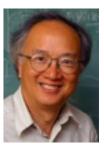
- \* No magnetic order down to 32mK
- \* Constant spin susceptibility at zero temperature

Other experiments: transport, heat capacity, optical absorption, etc, Unfortunately, **no neutron scattering** so far. Theoretical understanding: expected phase diagram

$$H = -t \sum_{\langle ij \rangle, \sigma} c_{i\sigma}^{\dagger} c_{j\sigma} + h.c. + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$



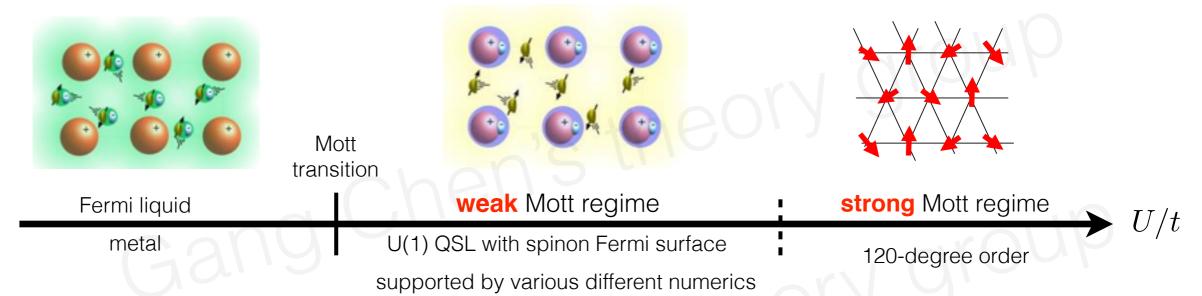




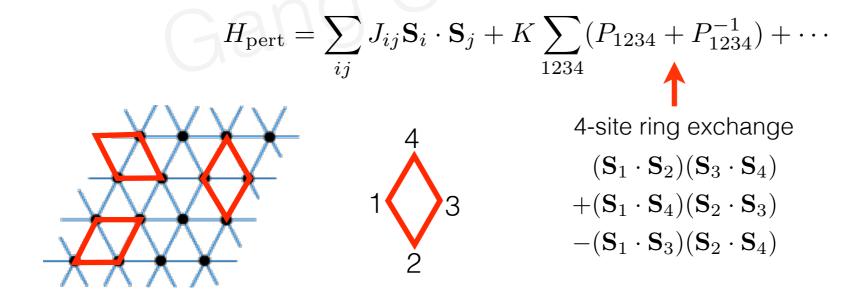
Sung-Sik Lee

T Senthil Patrick Lee

#### Senthil's cartoon



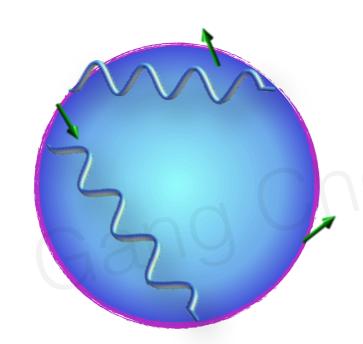
Physical mechanism for weak Mott insulator spin liquids: perturbation in t/U

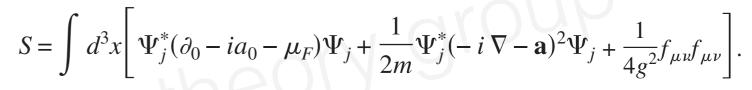




Motrunich

## Low energy property of spinon Fermi surface U(1) QSL: spinon non-Fermi liquid

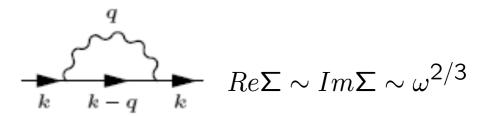






gauge photon is overly Landau-damped.

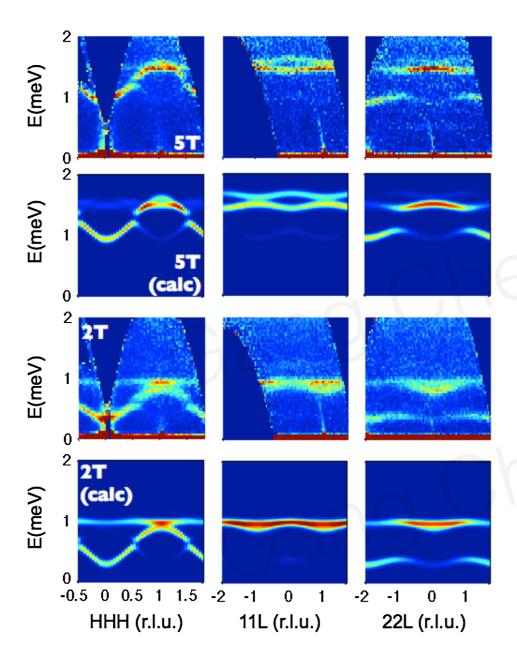
dual to extremal/charged black hole?



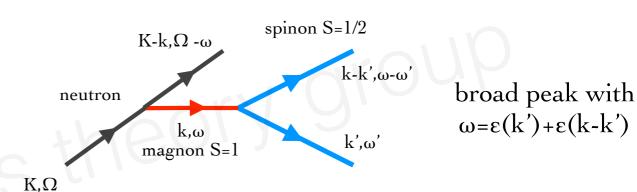


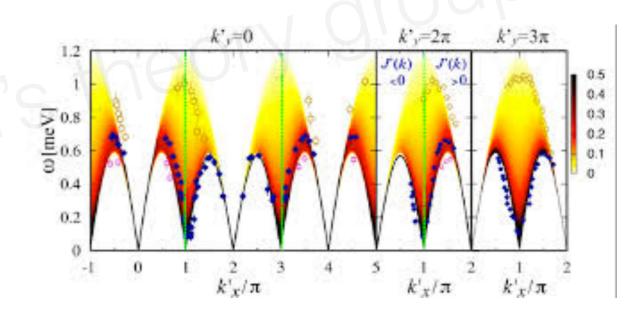
Hermele et al., PRB 70, 214437 (04) Sung-Sik Lee, PRB 78, 085129(08).

### Spin wave vs (fractionalized) spinon continuum



spin wave in Yb2Ti2O7 L Savary, et al, PRX 2011

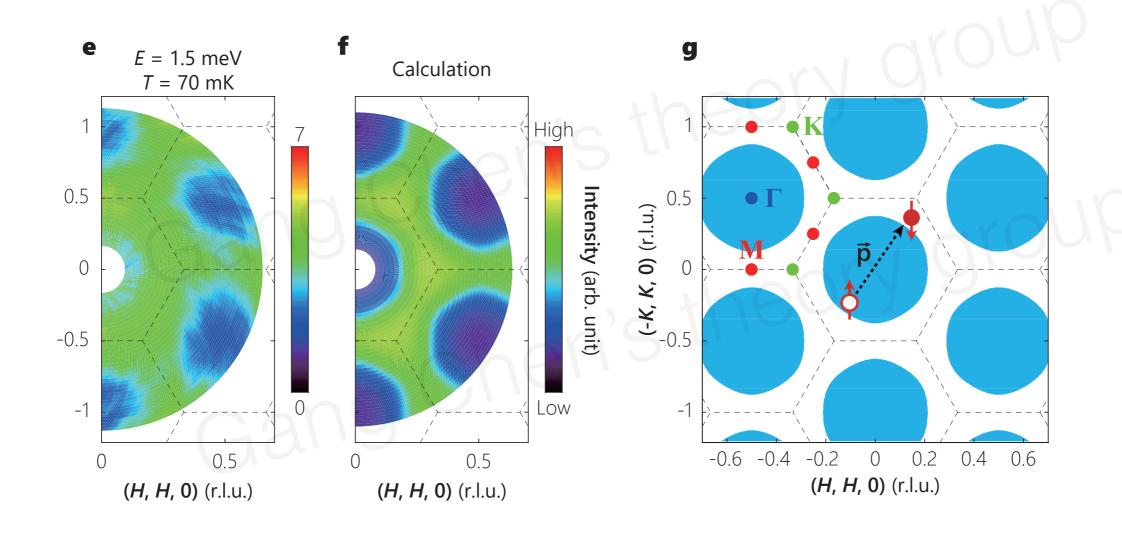




spinon continuum in Cs2CuCl4 Masanori, etc NatPhys 2009 but these are **1d spinons**!

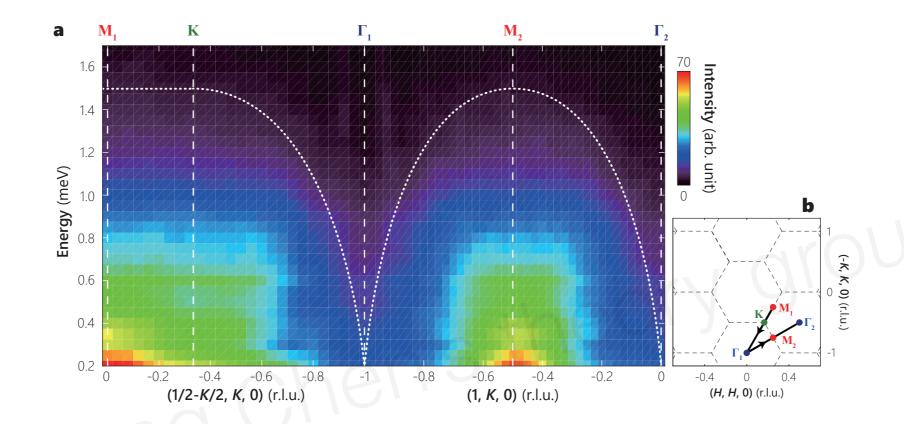


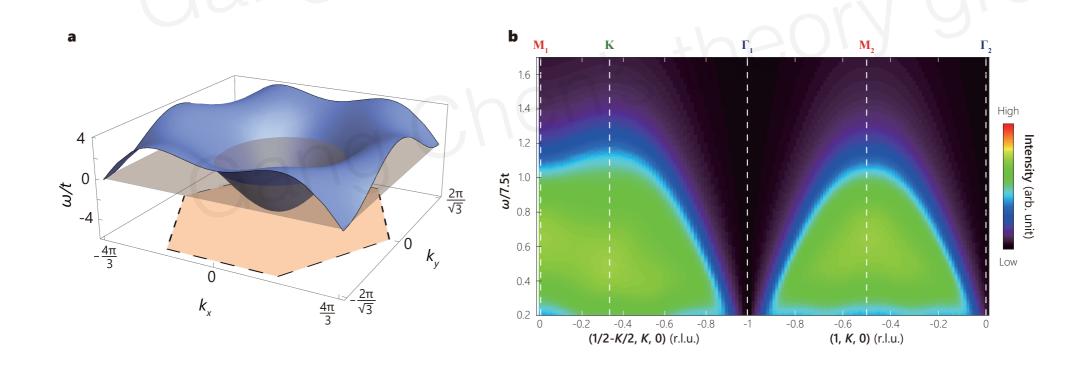
### Huge spinon continuum at all energies



Yao Shen, ...GC, Jun Zhao arxiv 2016







### Summary

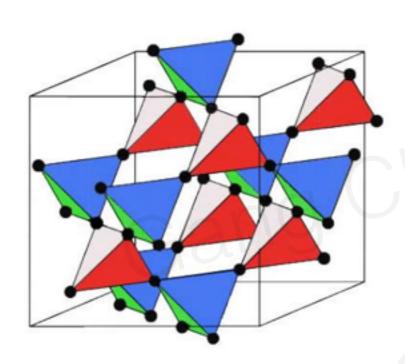
1. QSL is a field that bridges the fundamental ideas with the frontier experiments, it provides exciting opportunities for both theorists and experimentalists.

- 2. Rare-earth triangular lattice quantum spin liquid: YbMgGaO4
  - To our best knowledge, this is the **first strong spin-orbit coupled** quantum spin liquid candidate with odd number of electrons per unit cell and effective spin-1/2 moment.

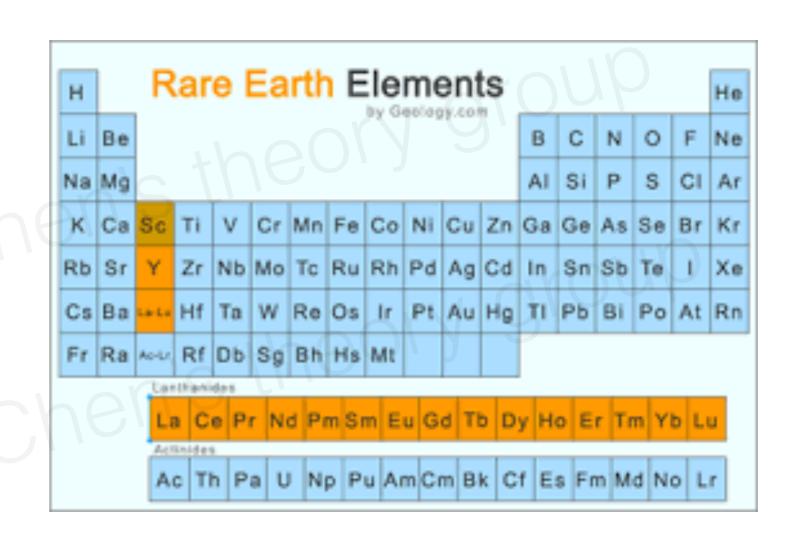
 Octupolar U(1) quantum spin liquid of quantum spin ice



### Rare-earth pyrochlores

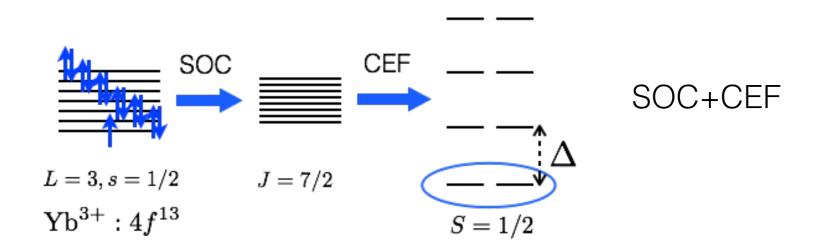


RE<sub>2</sub>M<sub>2</sub>O<sub>7</sub>

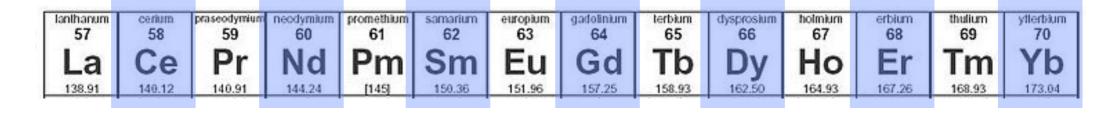




### Rare-earth local moments: a crude classification



Kramers' doublet: R3+ with **odd** number of electrons



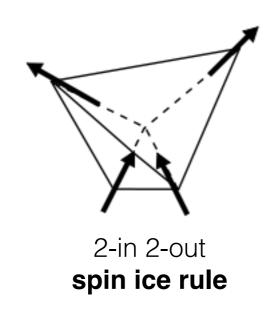
Non-Kramers' doublet / singlet: R3+ with **even** number of electrons

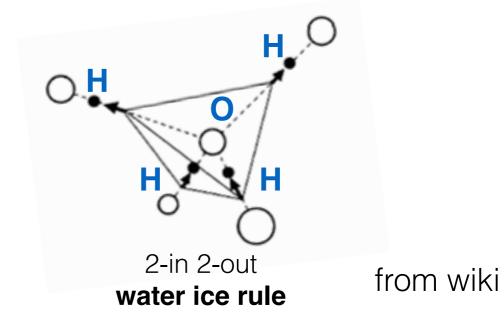
lanthanum 57	cerium 58	praseodymium 59	neodymium 60	promethium 61	samarium 62	europium 63	gadolinium 64	terbium 65	dysprosium 66	holmium 67	erbium 68	thulium 69	ytlerbium 70
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb
138.91	140.12	140.91	144.24	[145]	150.36	151.96	157.25	158.93	162.50	164.93	167.26	168.93	173.04



## Spin ice (Ising) limit

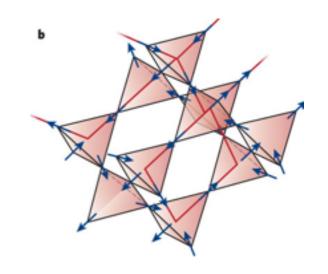
$$H = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z + \dots$$







## Classical spin ice



- The "2-in 2-out" states are extensively degenerate.
- At temperature T < Jzz, the system thermally fluctuates within the ice manifold, leading to classical spin ice and interesting experimental discoveries.





Pinch points in spin correlation



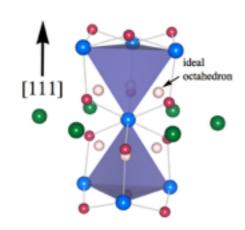
# Dipole-octupole doublet

The early classification of local moments is a bit crude!

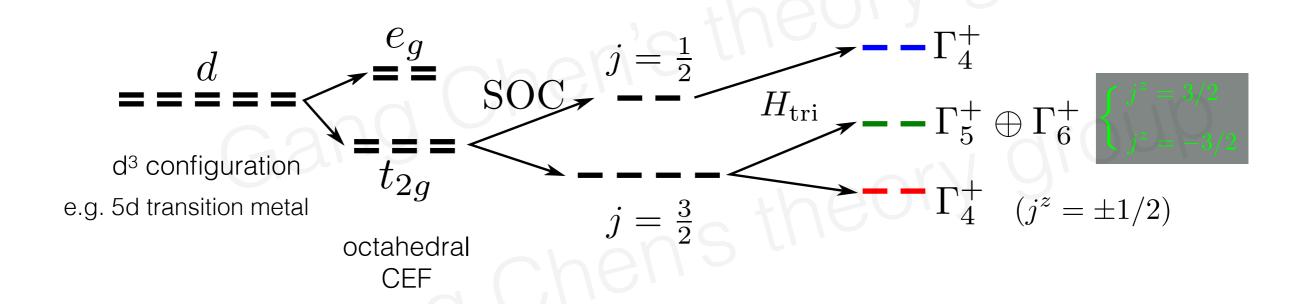
One should carefully examine the wavefunction of the local doublet.







Local moments on pyrochlore lattice: effective spin-1/2

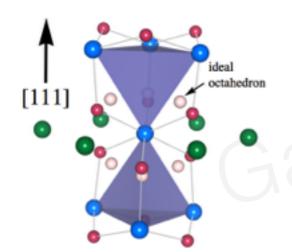


d electrons under D3d point group crystal field



Why is this Kramers doublet so special?

ONE-dimensional representations of the point group!



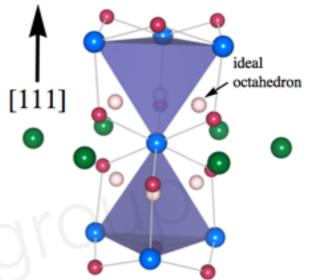
$$R(2\pi/3)|J^z = \pm 3/2\rangle = -|J^z = \pm 3/2\rangle$$

$$R(2\pi/3) \equiv e^{-i\frac{2\pi}{3}J^z} = e^{-i\frac{2\pi}{3}\times(\pm\frac{3}{2})} = e^{\mp i\pi} = -1$$

$$J^z = +3/2\rangle$$
 time reversal  $|J^z = -3/2\rangle$ 

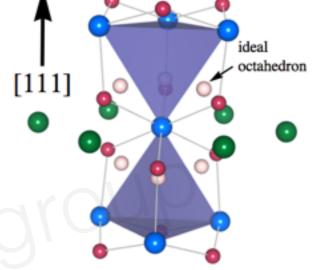


## More generally, ...



Also applies to 4f electron moments on pyrochlore

$$J = \frac{3}{2}, \frac{9}{2}, \frac{15}{2}, \dots$$



with the local crystal field Hamiltonian

$$H_{
m cf} = 3B_2^0 (J^z)^2 + \cdots \quad {
m if } B_2^0 < 0.$$

local doublet wavefunction of  $\mathrm{Dy^{3+}}$   $(J = \frac{15}{2})$  in  $\mathrm{Dy_2Ti_2O_7}$ e.g.

$$|\phi_0^{\pm}\rangle = 0.981|\pm\frac{15}{2}\rangle \pm 0.190|\pm\frac{9}{2}\rangle - 0.022|\pm\frac{3}{2}\rangle \mp 0.037|\mp\frac{3}{2}\rangle + 0.005|\mp\frac{9}{2}\rangle \pm 0.001|\mp\frac{15}{2}\rangle$$



### Emphasis: what matters is the wavefunction, not the spin value!

may generally apply to any Kramers' doublets with J > 1/2!

e.g, Ce: Ce2Sn2O7

PRL 115, 097202 (2015)

PHYSICAL REVIEW LETTERS

week ending 28 AUGUST 2015

#### Candidate Quantum Spin Liquid in the Ce<sup>3+</sup> Pyrochlore Stannate Ce<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>

Romain Sibille, 1,\* Elsa Lhotel, Vladimir Pomjakushin, Chris Baines, Tom Fennell, and Michel Kenzelmann

 $4f^1$  ion in  $D_{3d}$  local symmetry to the susceptibility was realized between T=1.8 and 370 K, and the resulting calculation of the single ion magnetic moment is shown in Fig. 2(c). The wave functions of the ground state Kramers doublet correspond to a linear combination of  $m_J=\pm 3/2$  states. The fitted coefficients result in energy levels at  $50 \pm$ 

Ce<sup>3+</sup> 
$$(4f^1, {}^2F_{5/2}).$$
  
$$J = \frac{5}{2}$$



### Realistic XYZ model

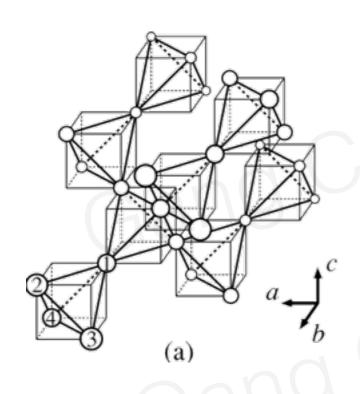
and

Symmetry Enriched U(1) topological order



## Symmetry properties

Effective spin-1/2 under lattice symmetry Tetrahedral Group



$$T_d \times \mathcal{I} \times translations$$
 and  $T_d = \{C_3, M\}$ 

$$\begin{cases} S^{z} = \frac{1}{2} |\frac{3}{2}\rangle \langle \frac{3}{2}| - \frac{1}{2}| - \frac{3}{2}\rangle \langle -\frac{3}{2}| \\ S^{+} = |\frac{3}{2}\rangle \langle -\frac{3}{2}|, \ S^{-} = |-\frac{3}{2}\rangle \langle \frac{3}{2}| \end{cases}$$

$$C_3: S^{\mu} \to S^{\mu}$$

$$M: S^{x,z} \to -S^{x,z}, S^y \to S^y$$

$$\mathcal{I}: S^{\mu} \to S^{\mu}$$

Important:  $S^x$  and  $S^z$  transform identically (as a dipole), while  $S^y$  transforms as an octupole moment under *mirror*.

## Generic model: XYZ model

$$H = \sum_{\langle ij \rangle} J_z S_i^z S_j^z + J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_{xz} \left( S_i^x S_j^z + S_i^z S_j^x \right)$$

**Uniform** Spatially!

**VS** 

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

Anisotropic Spatially!



#### A small transformation into XYZ model

$$H = \sum_{\langle ij \rangle} J_z S_i^z S_j^z + J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_{xz} \left( S_i^x S_j^z + S_i^z S_j^x \right)$$

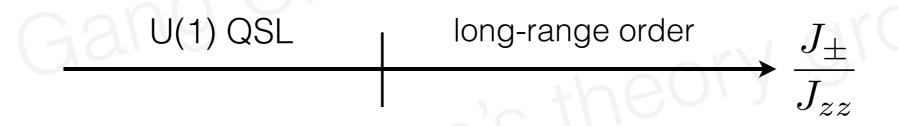


Rotation around the y axis in the effective spin space

$$H_{ ext{XYZ}} = \sum_{\langle ij \rangle} ilde{J}_z ilde{S}_i^z ilde{S}_j^z + ilde{J}_x ilde{S}_i^x ilde{S}_j^x + ilde{J}_y ilde{S}_i^y ilde{S}_j^y$$
 XYZ model

## XXZ model can lead to U(1) QSL

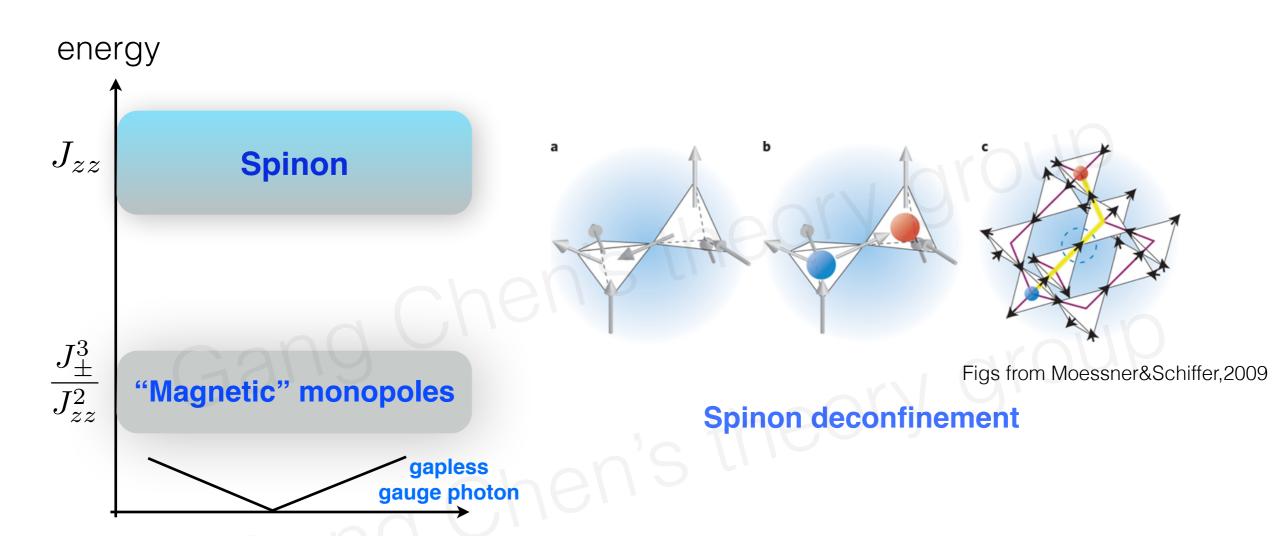
$$H = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z \left( -J_{\pm} \sum_{\langle i,j \rangle} \left( S_i^+ S_j^- + S_i^- S_j^+ \right) \right) + \cdots$$
 Hermele, Fisher, Balents, Moessner, Isakov, ....



• Pretty much one can add any term to create **quantum** tunneling, as long as it is not too large to induce magnetic order, the **ground state** is a U(1) QSL!



# Emergent Quantum Electrodynamics



Emergent electric field

 $S^z \sim E$ 

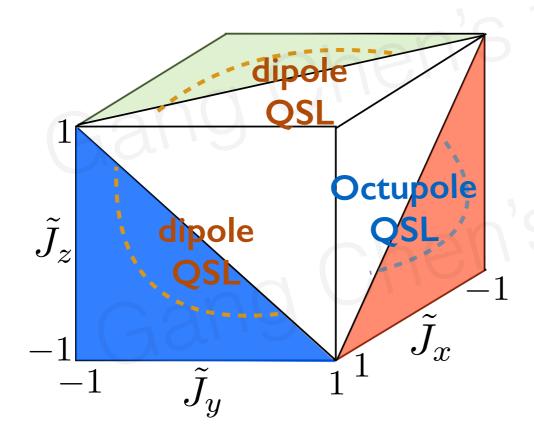
Emergent vector potential

 $S^{\pm} \sim e^{\pm iA}$ 



# XYZ model is the generic model that describes the interaction between DO doublets.

$$H_{XYZ} = \sum_{\langle ij \rangle} \mathcal{J}_x \tau_i^z \tau_j^z + \mathcal{J}_y \tau_i^y \tau_j^y + \mathcal{J}_z \tau_i^z \tau_j^z$$



3D phase diagram

Each component (not just Sz) can be emergent electric field, depending on the parameters!

Study phase on a cube:  $-1 \leq \tilde{J}_{x,y,z} \leq 1$ .

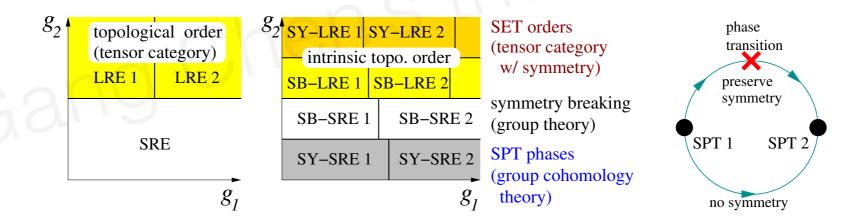




Xiaogang Wen

## Gapped phases w/ symmetry $\rightarrow$ SET and SPT phases

- there are LRE symmetric states  $\rightarrow$  Symm. Enriched Topo. phases
  - 100s symm. spin liquid through the PSG of topo. excit. Wen 02
  - 8 trans. symm. enriched  $Z_2$  topo. order in 2D, 256 in 3D Kou-Wen 09
  - 1000,000s symm.  $Z_2$  spin liquid through  $[\mathcal{H}^2(SG,Z_2)]^2 \times$  Hermele 12
  - Classify SET phases through  $\mathcal{H}^3[SG \times GG, U(1)]$  Ran 12
- there are SRE symmetric states → many different phases
   We may call them symmetry protected trivial (SPT) phase





 Control spinons in a quantum spin ice U(1) quantum spin liquid

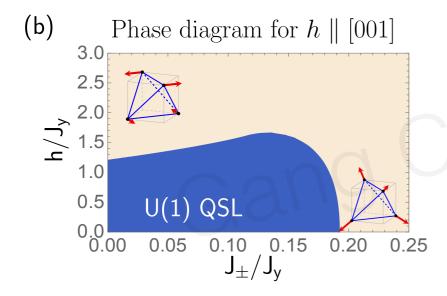


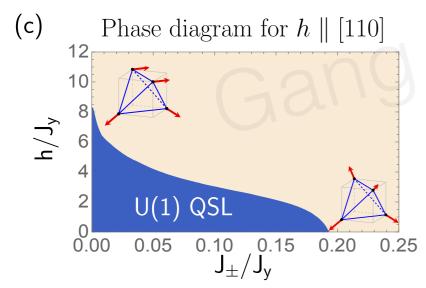
Yao-Dong Li (Fudan)

arxiv 1607



# (a) Phase diagram for $h \parallel [111]$ 3.0 2.5 2.0 1.5 1.0 0.5 0.00 0.05 0.00 0.05 0.10 0.15 0.20 0.25





# Field-driven Higgs transition for octupolar U(1) QSL

How to tell if Ce2Sn2O7 is an octupolar U(1) QSL or not?

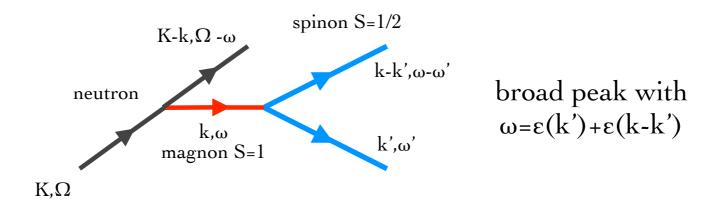
The idea to use a little knob that could simply lead to some clear experimental consequence, very much like the isotope effect of BCS superconductors.

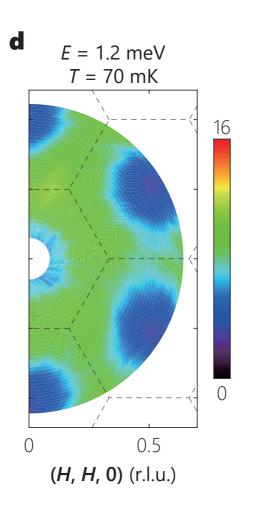
Here we apply external magnetic field, and expect a field-driven Higgs transition to magnetic ordering as the field only couples to the matter field (spinons).

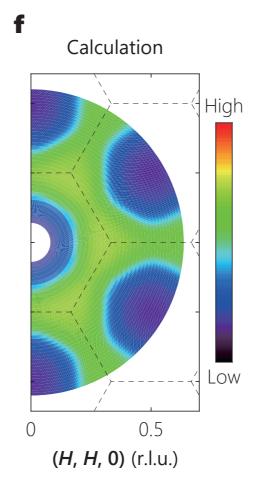
$$H_{\text{sim}} = \sum_{\langle ij \rangle} J_y \tau_i^y \tau_j^y - J_{\pm}(\tau_i^+ \tau_j^- + h.c.)$$
$$- \sum_i h \left( \hat{n} \cdot \hat{z}_i \right) \tau_i^z,$$
$$\tau_i^{\pm} = \tau_i^z \pm i \tau_i^x$$



# Inelastic neutron scattering and spinon continuum





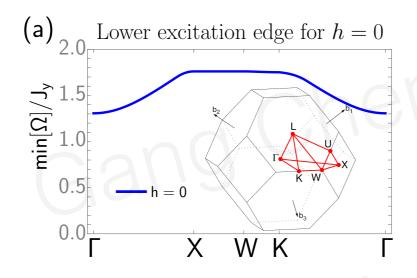


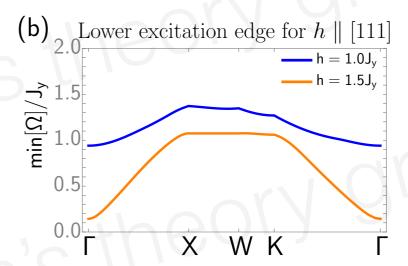
Spinon continuum in YbMgGaO4 (today's arXiv)

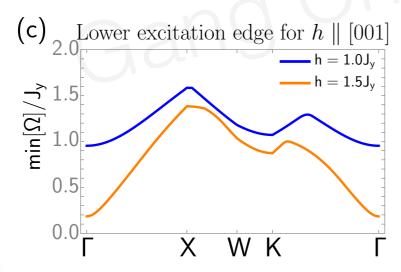


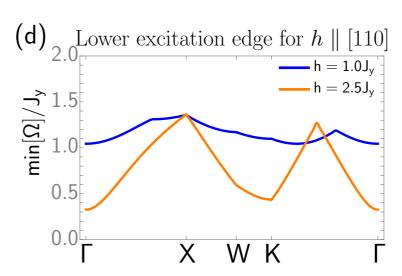
## Lower excitation edge

$$\mathbf{q} = \mathbf{k}_1 + \mathbf{k}_2,$$
  
$$\Omega(\mathbf{q}) = \omega_i(\mathbf{k}_1) + \omega_j(\mathbf{k}_2),$$











## Neutron scattering and thermal transport

Different U(1) QSLs	Heat capacity	Inelastic neutron scattering measurement
Octupolar $U(1)$ QSL for DO doublets	$C_v \sim T^3$	Gapped spinon continuum
Dipolar U(1) QSL for DO doublets	$C_v \sim T^3$	Both gapless gauge photon and gapped spinon continuum
Dipolar $U(1)$ QSL for non-Kramers' doublets	$C_v \sim T^3$	Gapless gauge photon
Dipolar U(1) QSL for usual Kramers' doublets	$C_v \sim T^3$	Both gapless gauge photon and gapped spinon continuum
Thermal transport		

#### Thermal transport

see both contribution, but there is a big separation of energy scales in spinon and gapless photons.



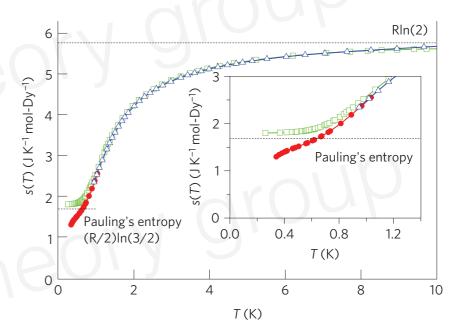
# Material survey: other DO doublet systems

Our doublet can potentially be realized for any Kramers spin moment with J > 1/2.

Two well-known systems:

Pyrochlores A<sub>2</sub>B<sub>2</sub>O<sub>7</sub>,

e.g., Nd<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, Nd<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Nd<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub>, etc Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Cd<sub>2</sub>Os<sub>2</sub>O<sub>7</sub>, etc Ce<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>,



Prof Gaulin's group, Dy2Ti2O7, Nat Phys, 2013

Spinels AB<sub>2</sub>X<sub>4</sub>, B=lanthanide?

CdYb<sub>2</sub>S<sub>4</sub>

e.g. CdEr2Se4



## Conclusion

- We propose a new doublet dubbed "dipole-octupole" doublet.
- We propose a generic XYZ model for our new doublet.
- This XYZ model supports both exotic (octupolar) order and symmetry enriched U(1) quantum spin liquid (quantum spin ice) ground states.
- There exist a large class of materials (not just pyrochlore, any other lattices with the same point group) that can support such doublets.
- The remarkable properties of the doublet allows a direct comparison between numerics and experiments. We propose a way to detect the consequence of symmetry enrichment.

