

Recent development of quantum spin liquids

Gang Chen (陈 钢)
Fudan University



Opportunity for students and postdocs

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



Some kind of center is forming. we are hiring people for all field of physics, and theoretical physics.

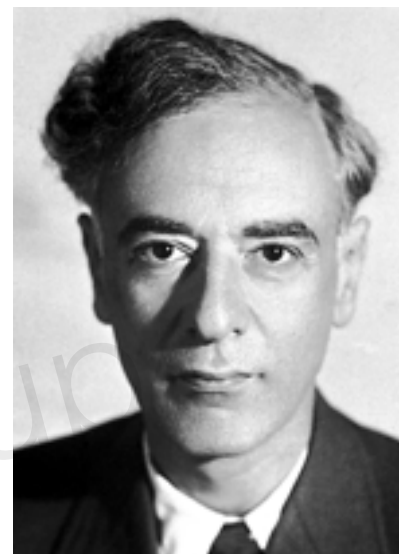
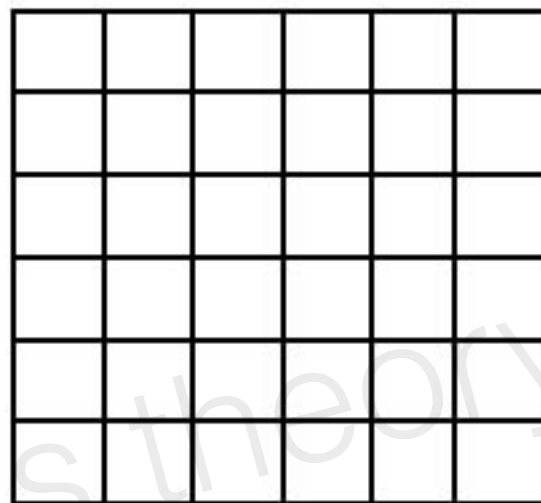
Outline

- 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

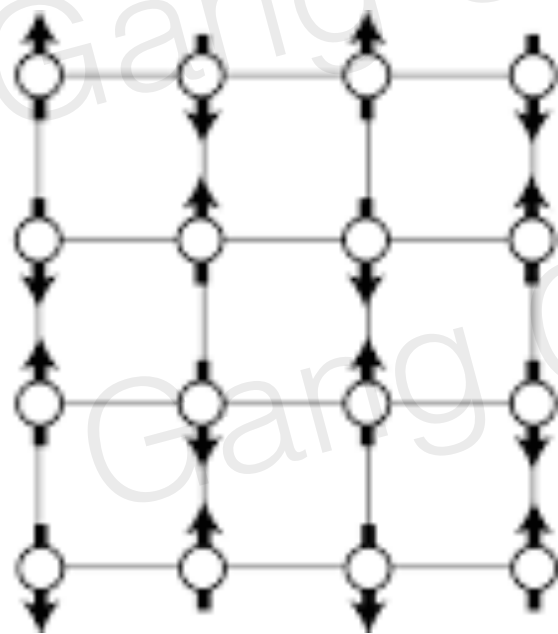
Neel vs Landau (1930-40s)



Neel



Landau



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

$$\text{Spin singlet} = \frac{1}{\sqrt{2}} \left[\begin{array}{c} \uparrow \text{---} \downarrow \\ \downarrow \text{---} \uparrow \end{array} \right]$$



P. W. Anderson

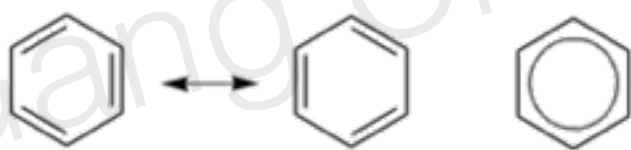
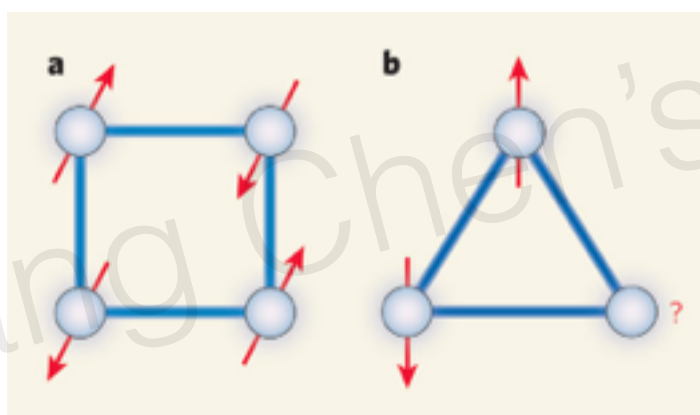
The idea of quantum spin liquid (1973)

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR?*

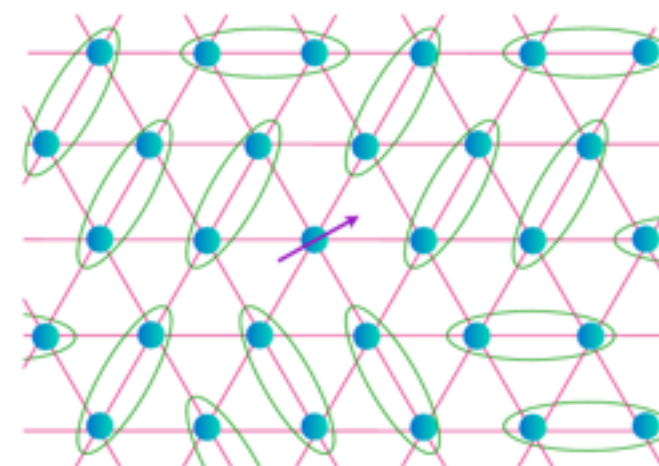
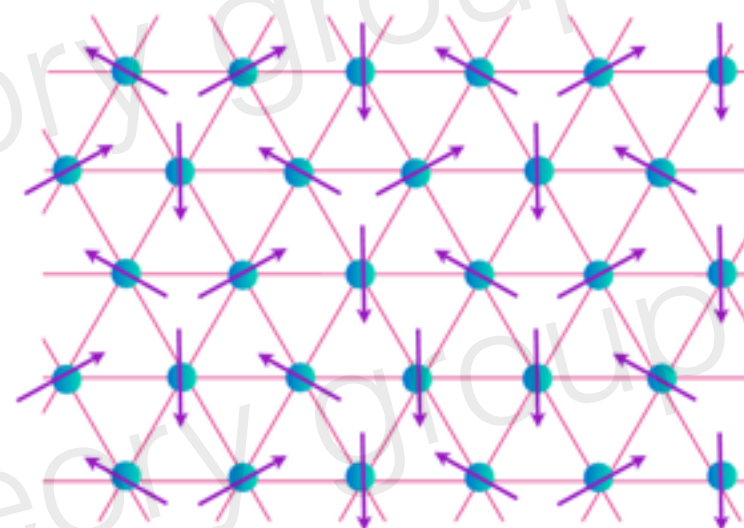
P. W. Anderson

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

it is very il
motivation

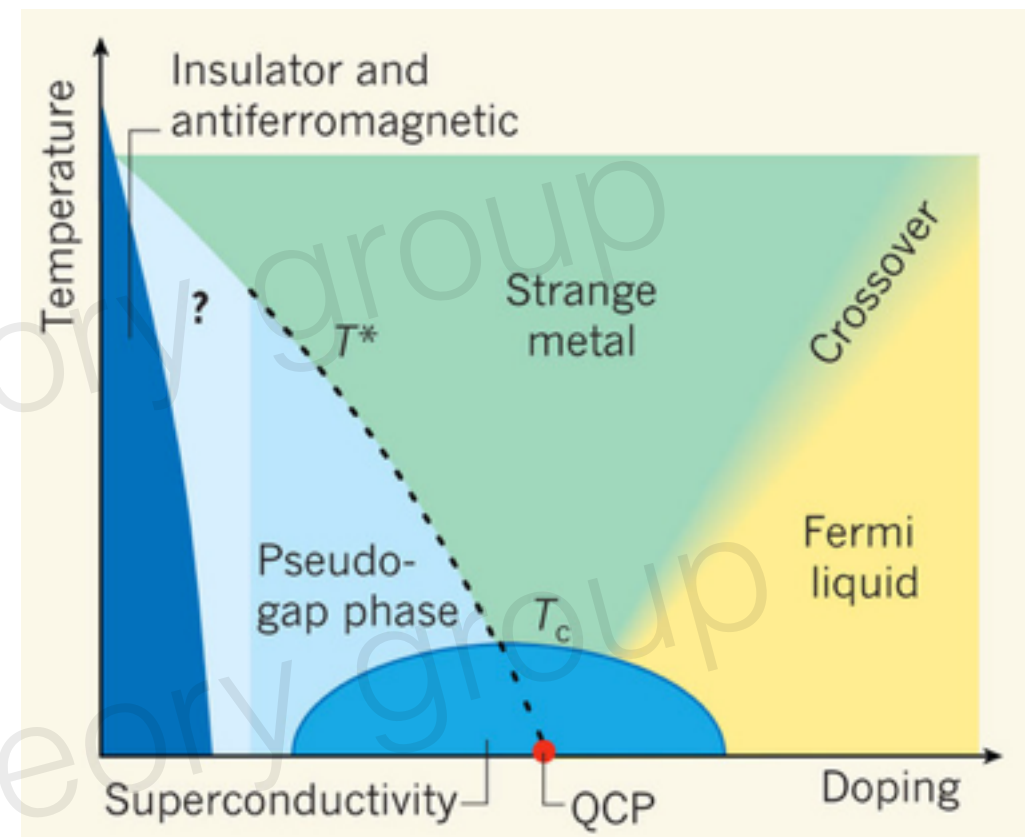
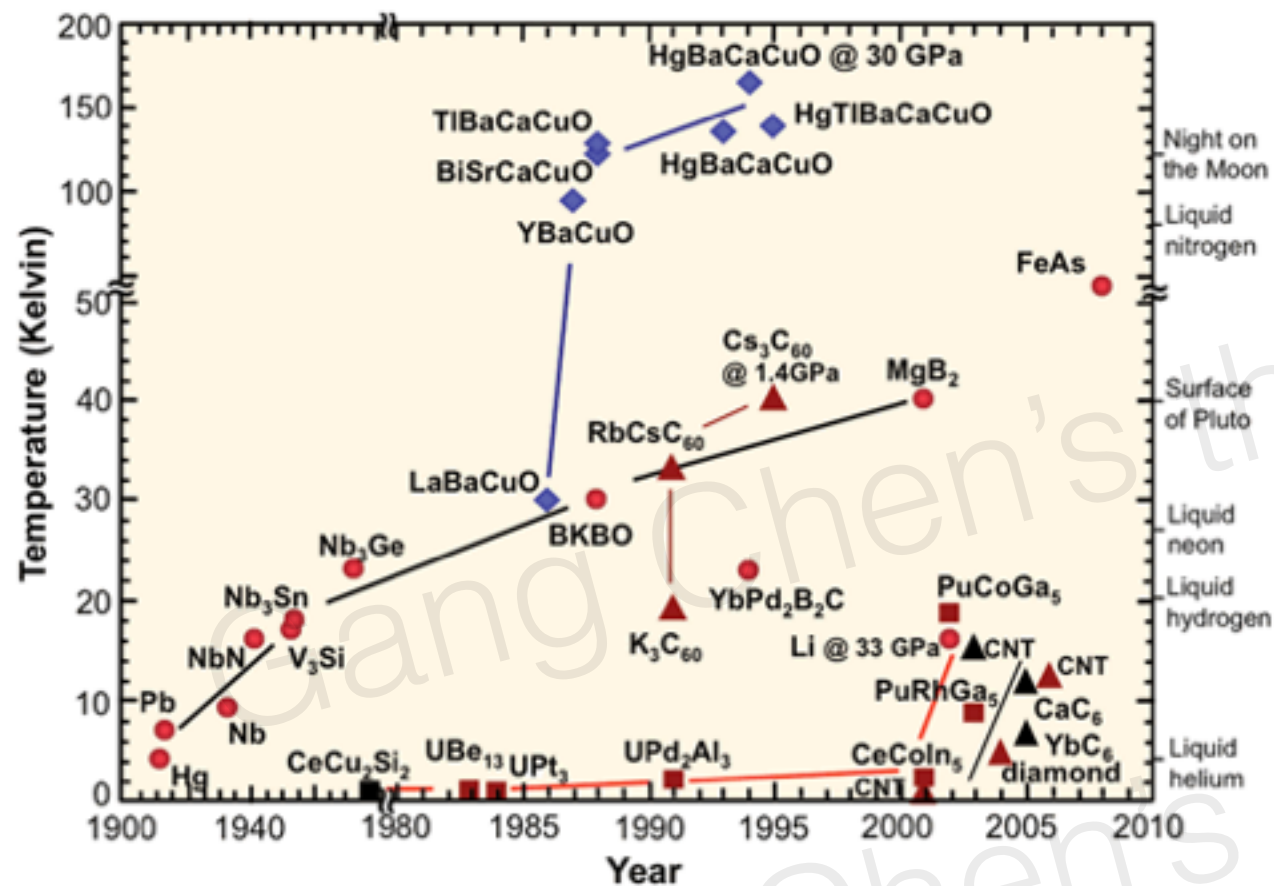


Pauling's RVB wavefunction
for Benzene molecule



$$\text{Diagram of a pair of spheres in a green oval} = \frac{1}{\sqrt{2}} \left[\begin{array}{c} \uparrow \text{---} \downarrow \\ \downarrow \text{---} \uparrow \end{array} \right]$$

High temperature superconductivity (1986)

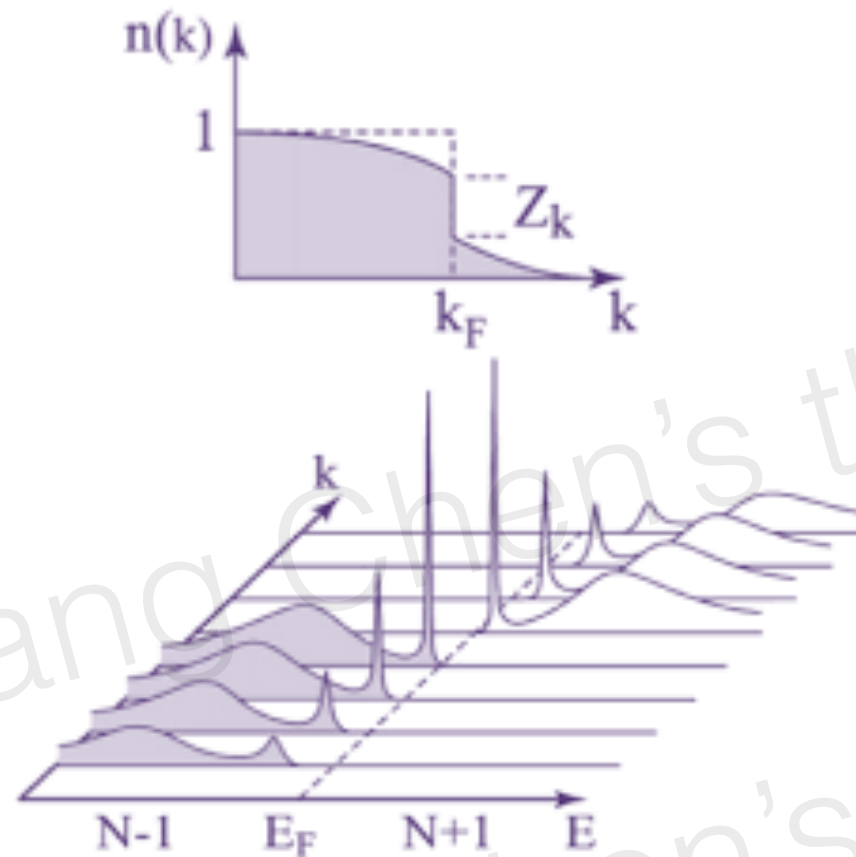


The idea is to view Mott insulator (QSL) as the parent state of high-temperature superconductor. In the QSL, there are preformed Cooper pairs. Doping it allows to Cooper pairs to condense and lead to superconductivity.

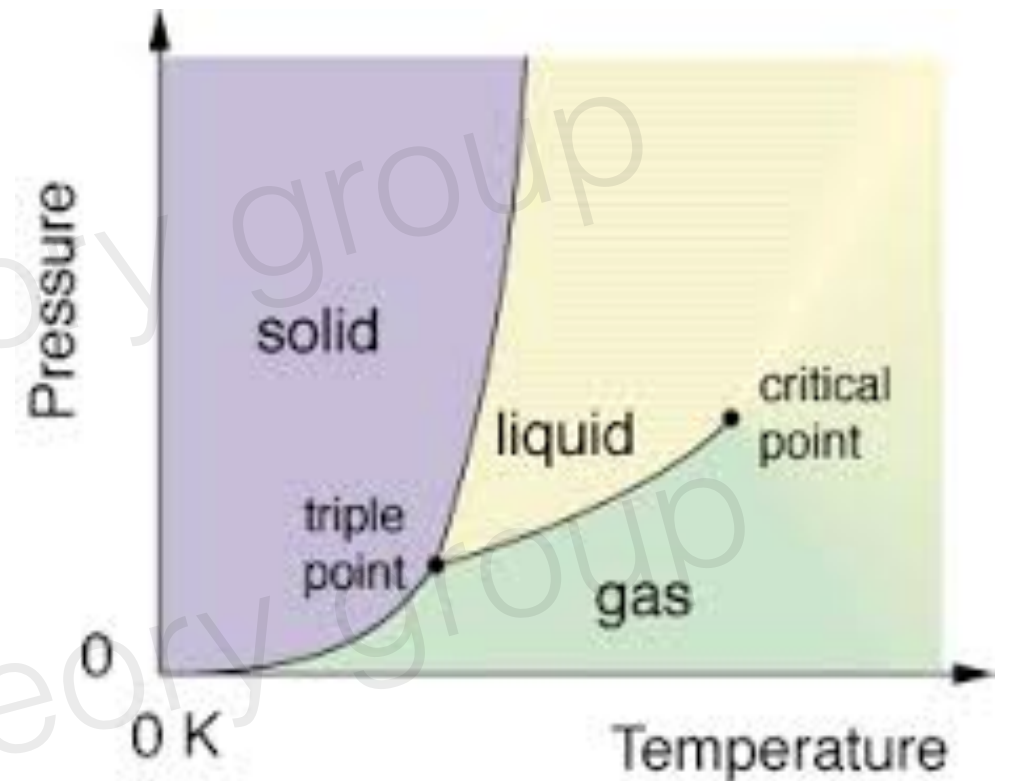


Landau

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: κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$, EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$, κ -H $_3$ (Cat-EDT-TTF) $_2$
herbertsmithite (ZnCu $_3$ (OH) $_6$ Cl $_2$), Ba $_3$ NiSb $_2$ O $_9$, Ba $_3$ CuSb $_2$ O $_9$, LiZn $_2$ Mo $_3$ O $_8$, ZnCu $_3$ (OH) $_6$ Cl $_2$
volborthite (Cu $_3$ V $_2$ O $_7$ (OH) $_2$), BaCu $_3$ V $_2$ O $_3$ (OH) $_2$, [NH $_4$] $_2$ [C $_7$ H $_{14}$ N][V $_7$ O $_6$ F $_{18}$], Na $_2$ IrO $_3$, CsCu $_2$ Cl $_4$, CsCu $_2$ Br $_4$, NiGa $_2$ S $_4$, He-3 layers on graphite, etc
- 3D pyrochlore, hyperkagome, FCC lattice, diamond lattice, etc
Na $_4$ Ir $_3$ O $_8$, IrO $_2$, Ba $_2$ YMoO $_6$, Yb $_2$ Ti $_2$ O $_7$, Pr $_2$ Zr $_2$ O $_7$, Pr $_2$ Sn $_2$ O $_7$, Tb $_2$ Ti $_2$ O $_7$, Nd $_2$ Zr $_2$ O $_7$, FeSc $_2$ S $_4$, 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)

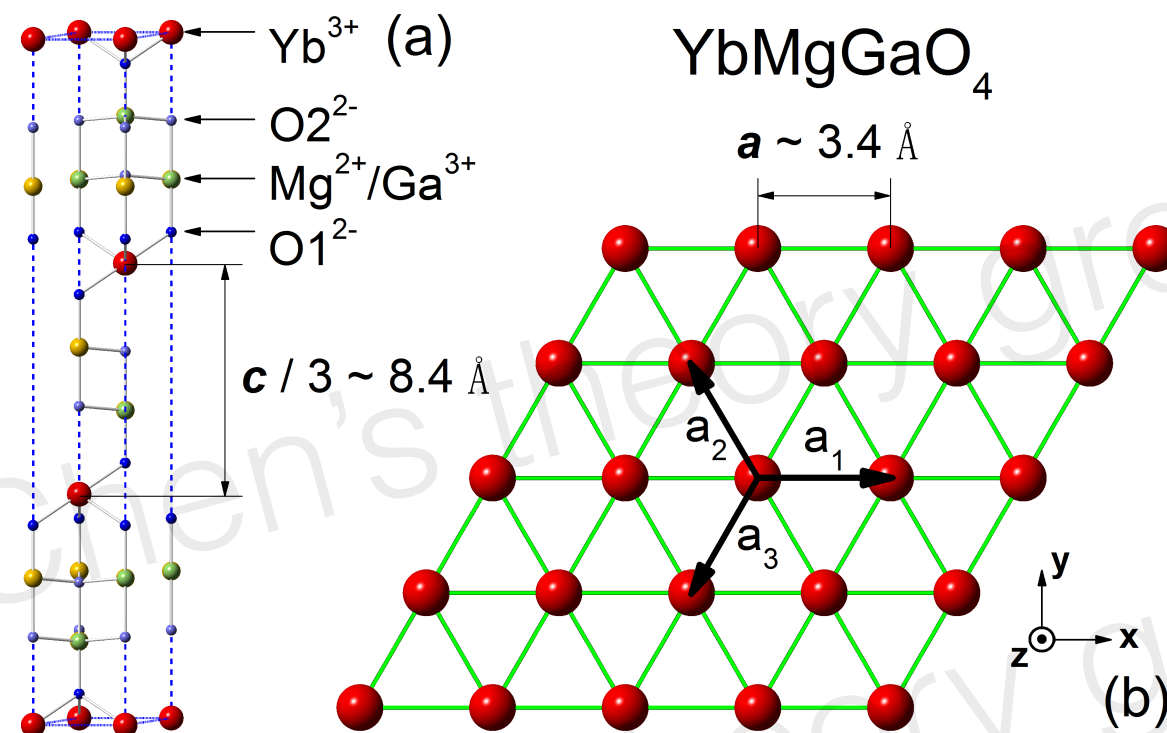
Zhaorong Yang (Institute of Solid-State Physics, Hefei)

Xiaoqun Wang (Renmin, Shanghai Jiaotong)

YS Li, **GC***,, QM Zhang*

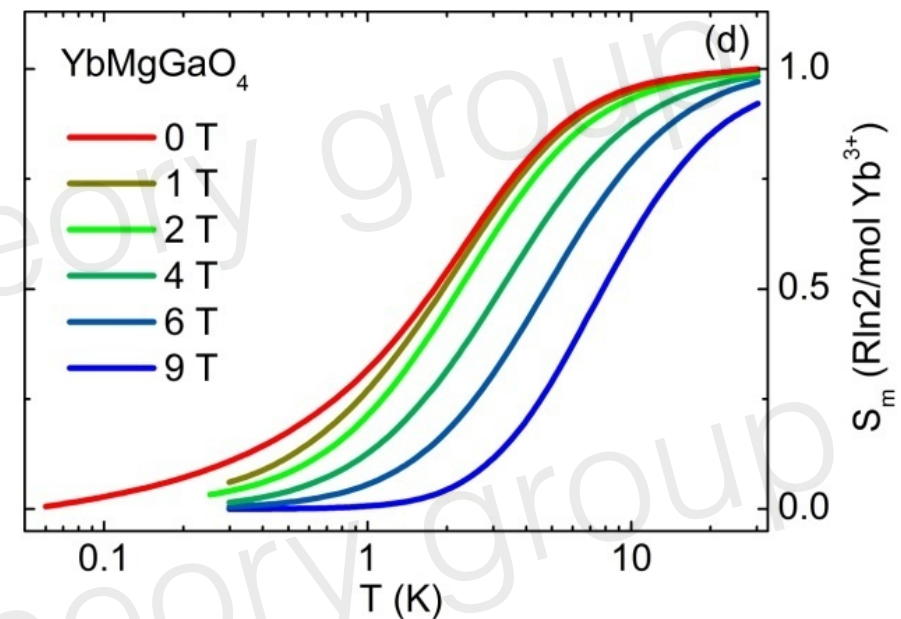
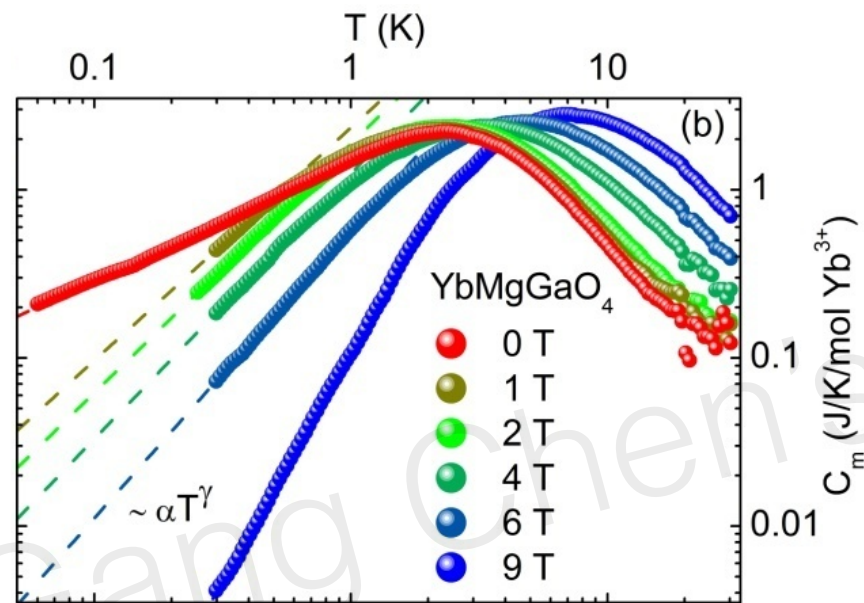
PhysRevLett 2015

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 $T^{2/3}$ heat capacity.
- We understand the microscopic Hamiltonian and the physical mechanism.

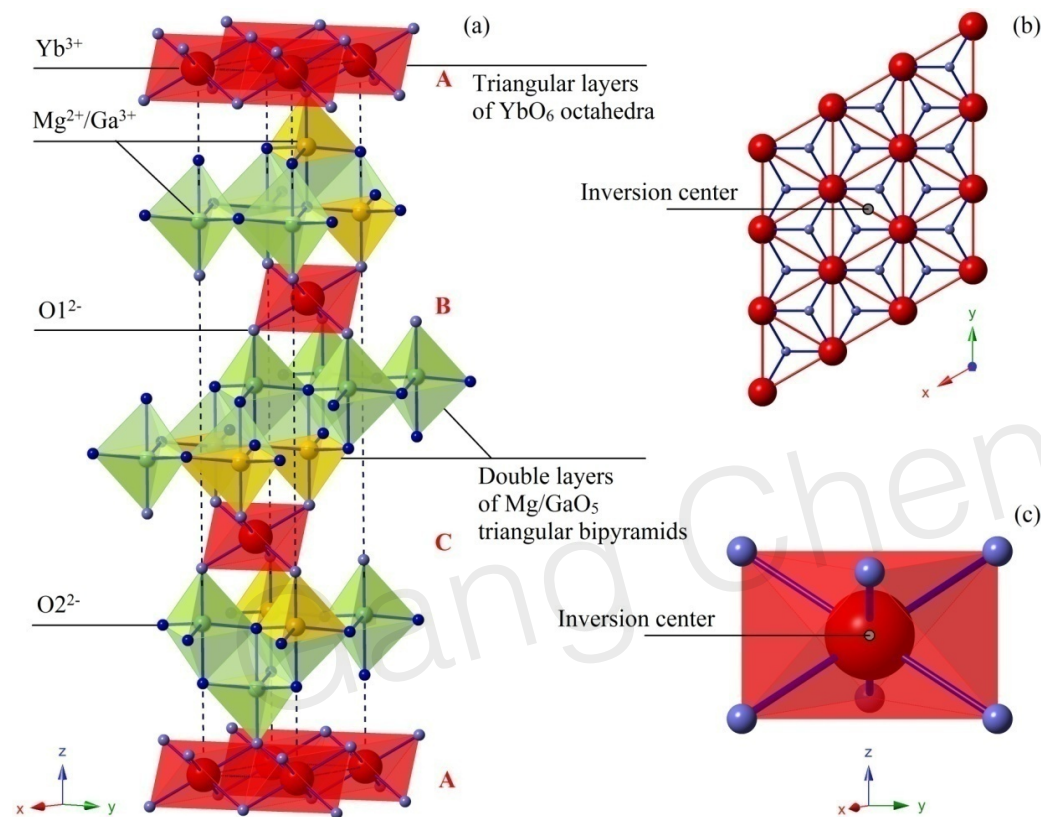
YbMgGaO₄



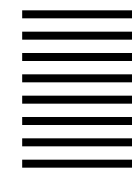
- observation of $T^{2/3}$ heat capacity
- Entropy: effective spin-1/2 local moments

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

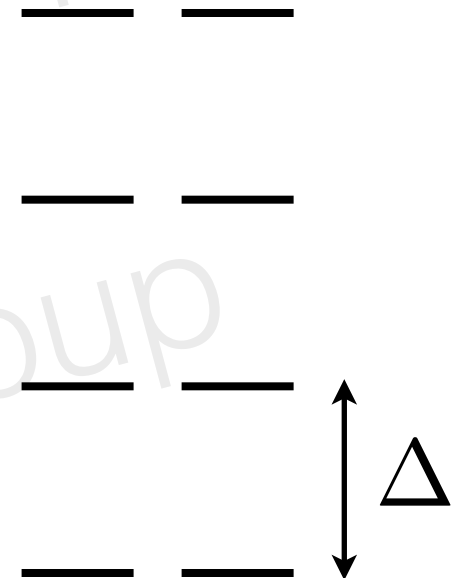
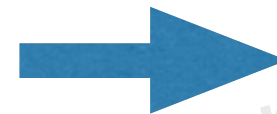
Microscopics



Yb³⁺ ion: 4f¹³ has $J=7/2$ due to SOC.



$J=7/2$



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,¹ Hoi Chun Po,¹ Ashvin Vishwanath,^{1,2} and Michael P. Zaletel³ 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 1D¹, later extended to higher dimensions by Hastings and Oshikawa^{2,3}, 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-

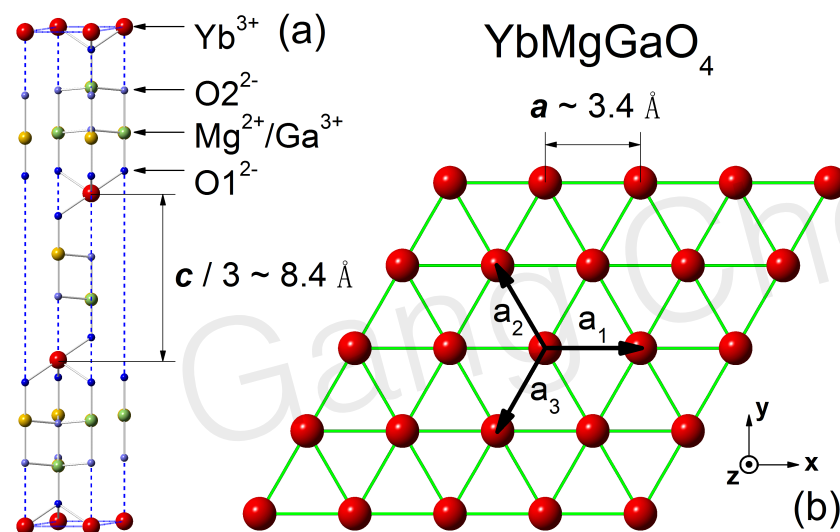
tirely different theoretical approaches are needed. We argue that if a spin-orbit coupled insulator at odd filling is time-reversal symmetric, its ground state must, in a precise sense, be exotic. We introduce two theoretic-

“this kind of system” means effective spin-1/2, spin-orbit coupling, odd number of electron per cell.

as you may know, there are many theoretical works trying to constrain the possible states from being existing.

What is the physical origin of the QSL?

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



PhysRevLett, 2015

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

where $S_i^{\pm} = S_i^x \pm iS_i^y$, 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-

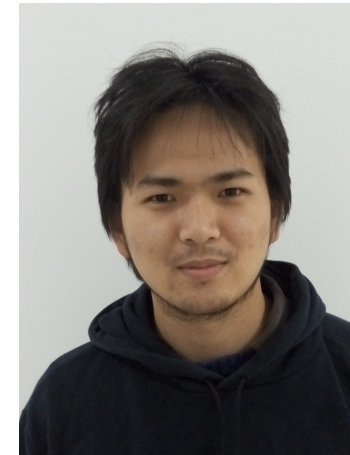
The spin-1/2 XXZ model supports conventional order.

(Yamamoto, etc, PRL 2014)

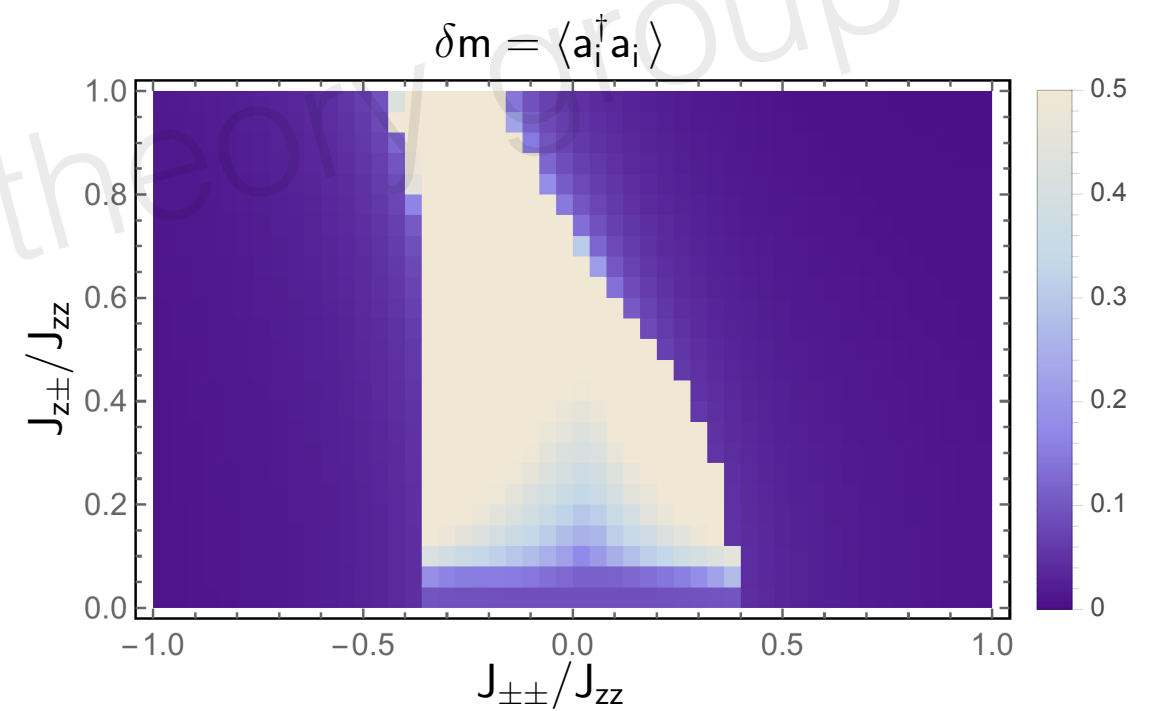
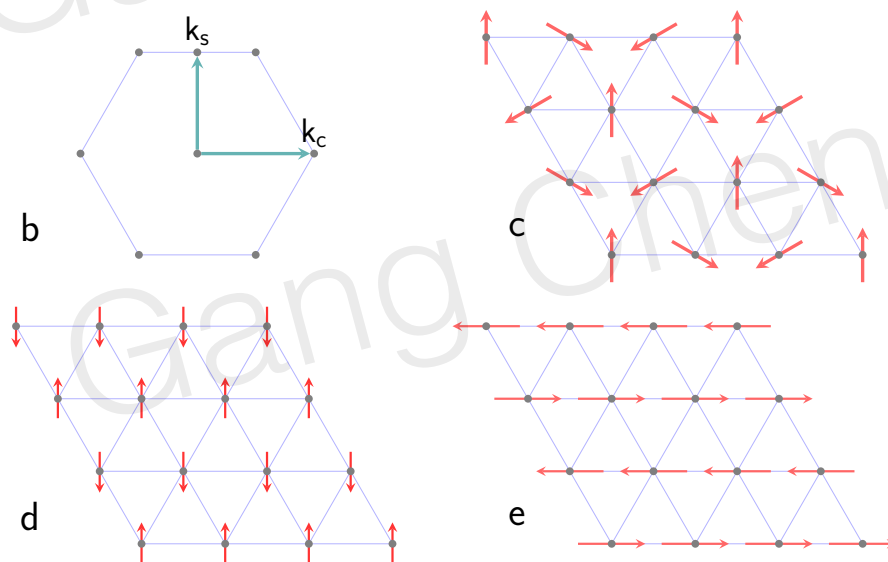
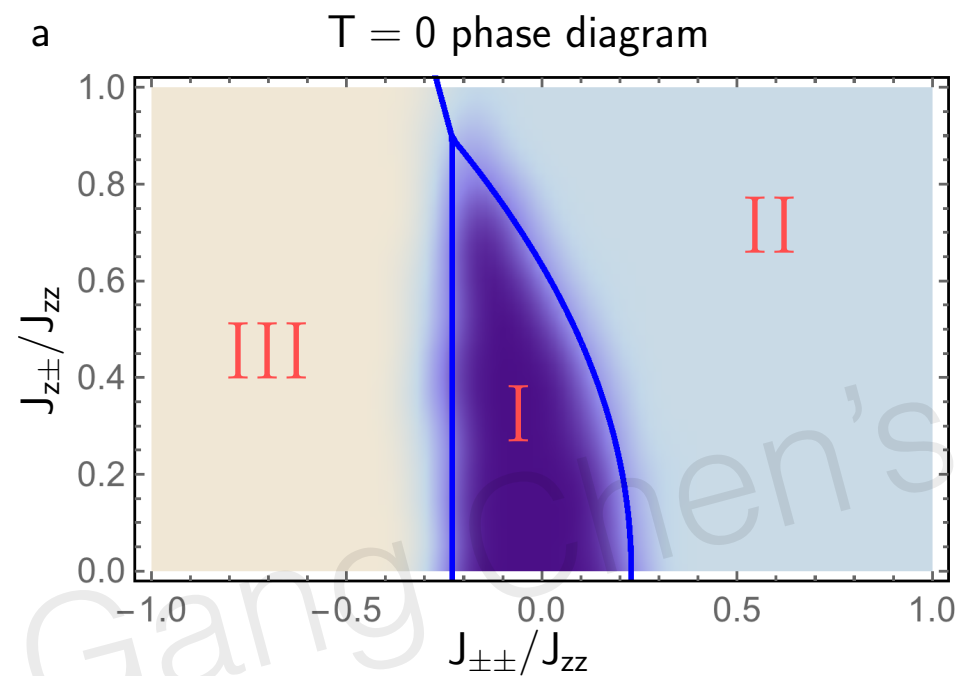
This will makes George happy.

This model

Anisotropic spin interaction
could potentially stabilize QSL.



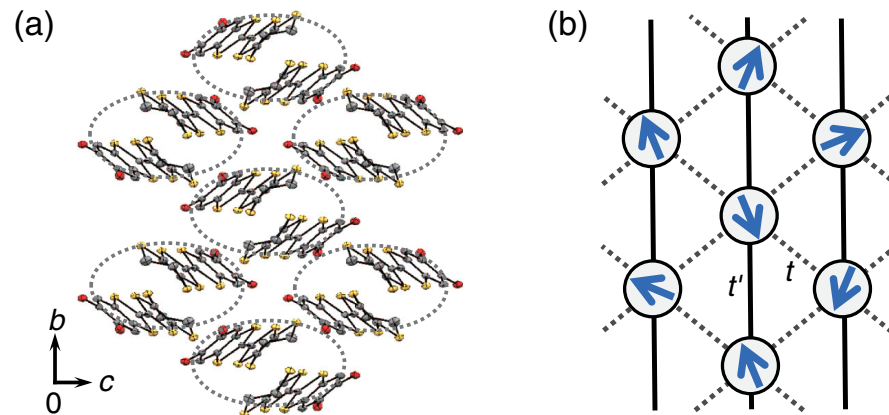
Yao-Dong Li
Dept of Computer Sciences
Fudan University



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

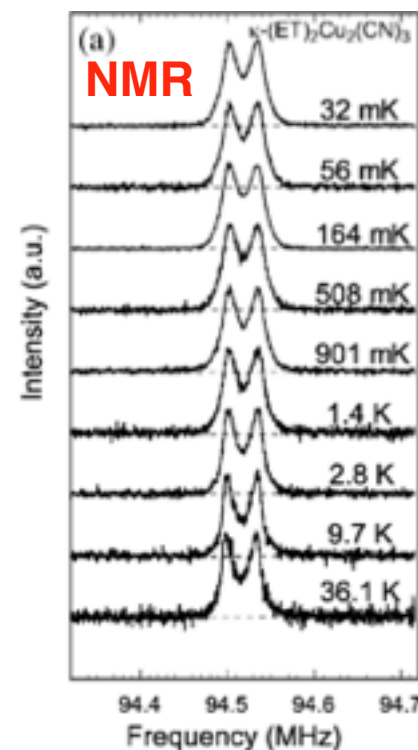
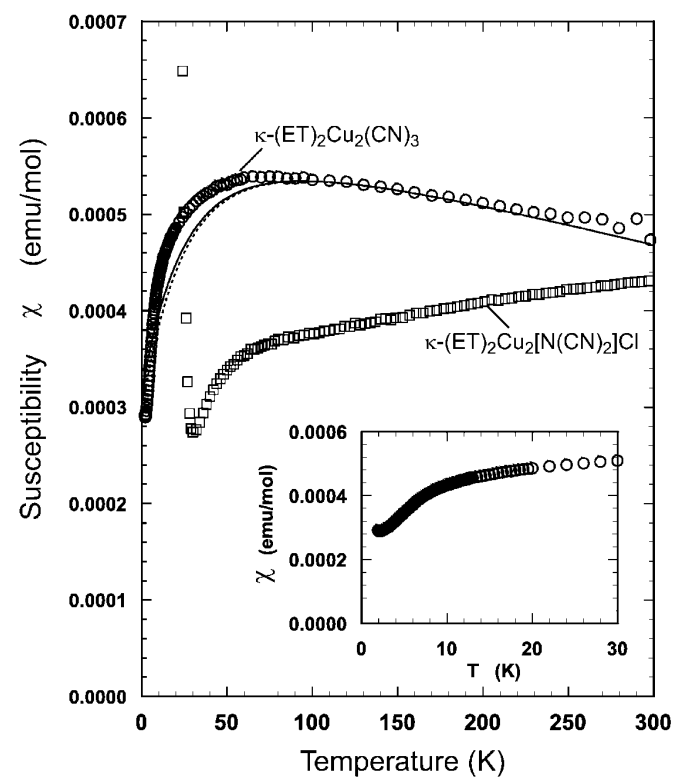


Kanoda



κ -(BEDT-TTF)₂Cu₂(CN)₃,
EtMe₃Sb[Pd(dmit)₂]₂,
 κ -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

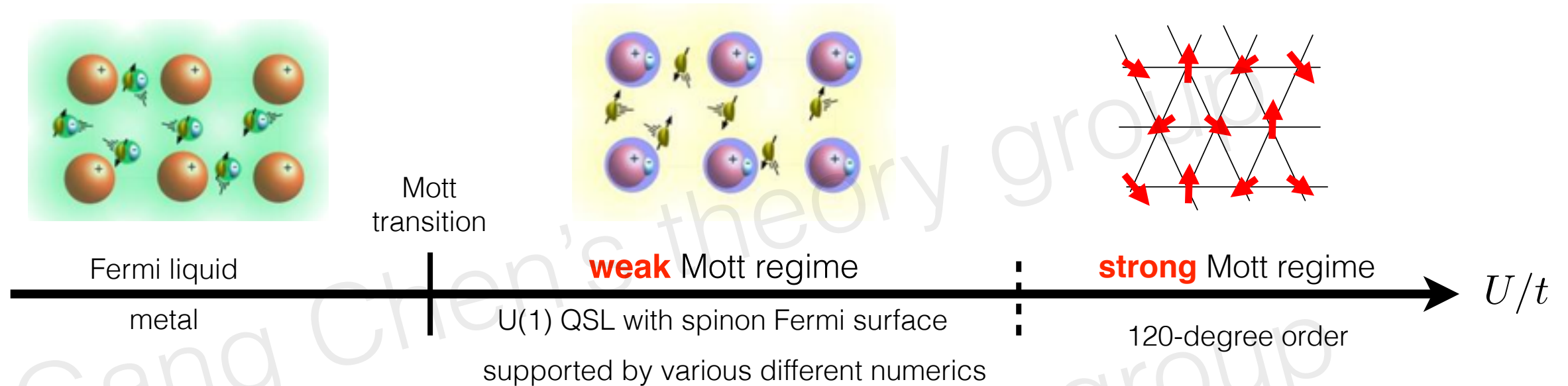


T Senthil



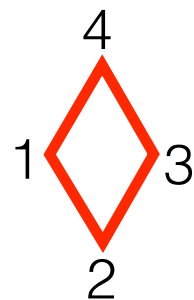
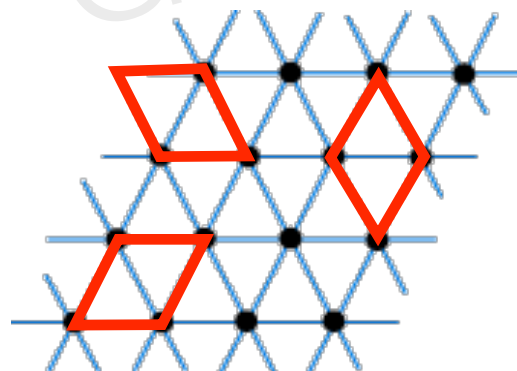
Patrick Lee

Senthil's cartoon



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

$$H_{\text{pert}} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{1234} (P_{1234} + P_{1234}^{-1}) + \dots$$



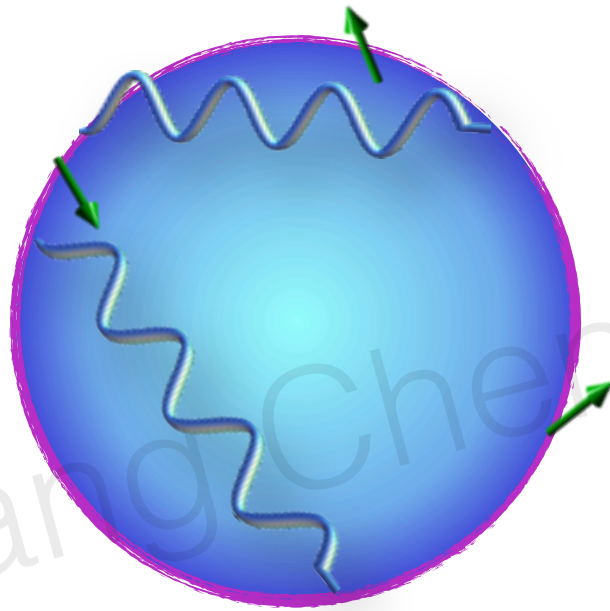
4-site ring exchange

$$(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_4)(\mathbf{S}_2 \cdot \mathbf{S}_3) - (\mathbf{S}_1 \cdot \mathbf{S}_3)(\mathbf{S}_2 \cdot \mathbf{S}_4)$$



Motrunich

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



Spinon Fermi surface coupled
with dynamical U(1) gauge field:
instanton event is suppressed.

dual to extremal/charged black hole?

$$S = \int d^3x \left[\Psi_j^* (\partial_0 - ia_0 - \mu_F) \Psi_j + \frac{1}{2m} \Psi_j^* (-i \nabla - \mathbf{a})^2 \Psi_j + \frac{1}{4g^2} f_{\mu\nu} f_{\mu\nu} \right].$$



gauge photon is overly Landau-damped.

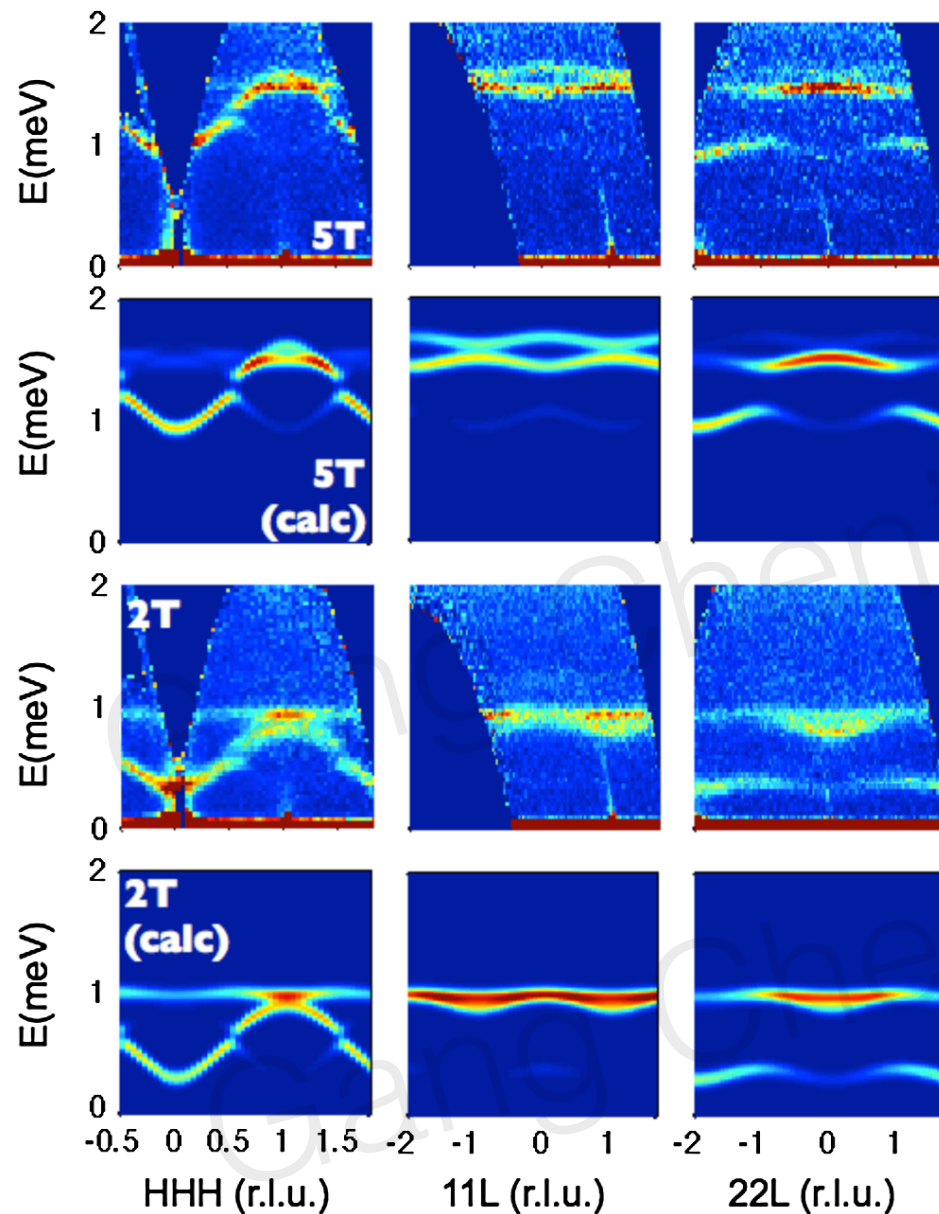


$$Re\Sigma \sim Im\Sigma \sim \omega^{2/3}$$

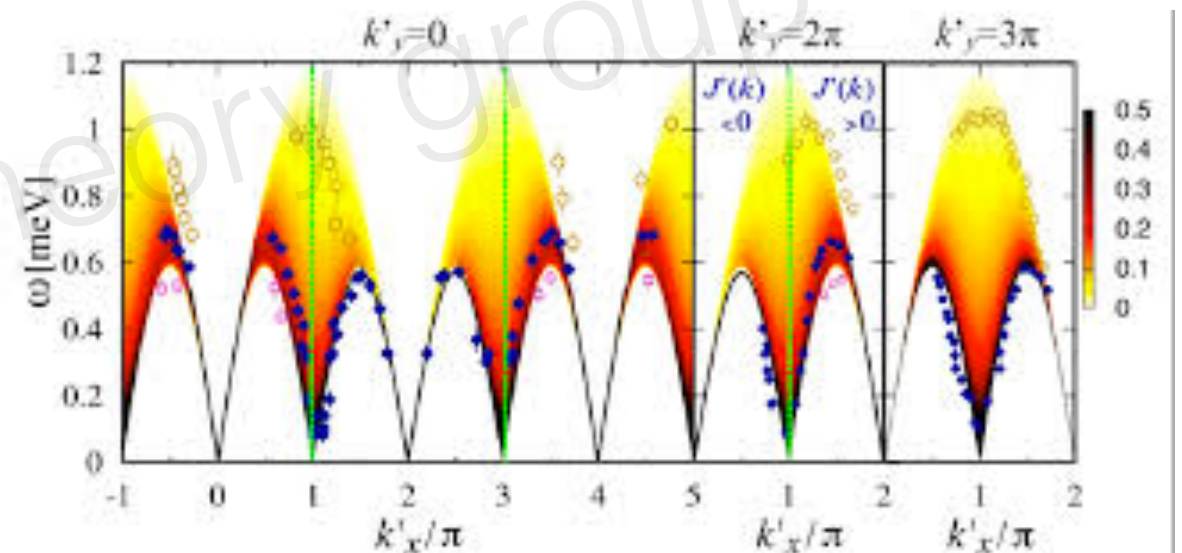
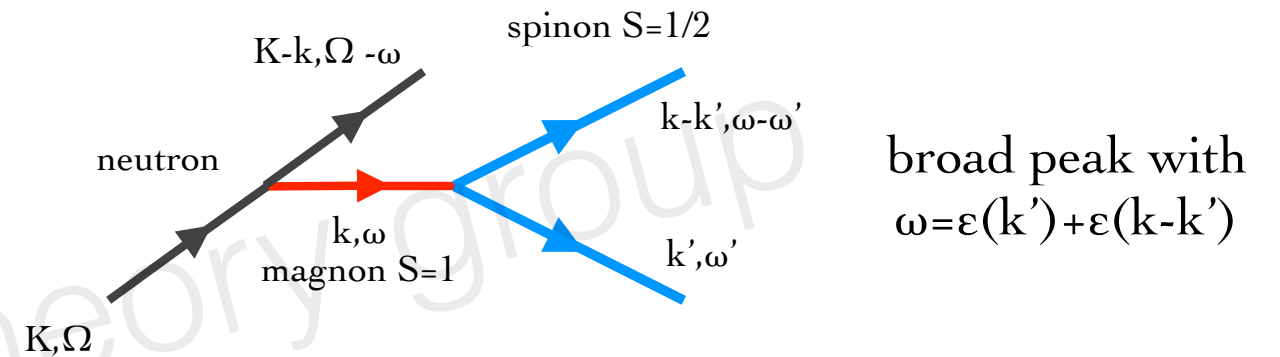
Hermele et al., PRB 70, 214437 (04)

Sung-Sik Lee, PRB 78, 085129(08).

Spin wave vs (fractionalized) spinon continuum

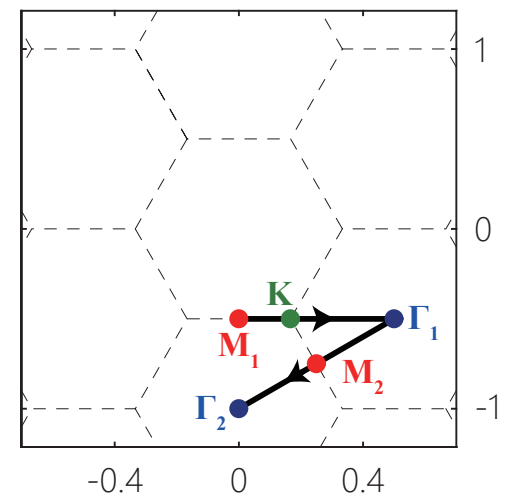
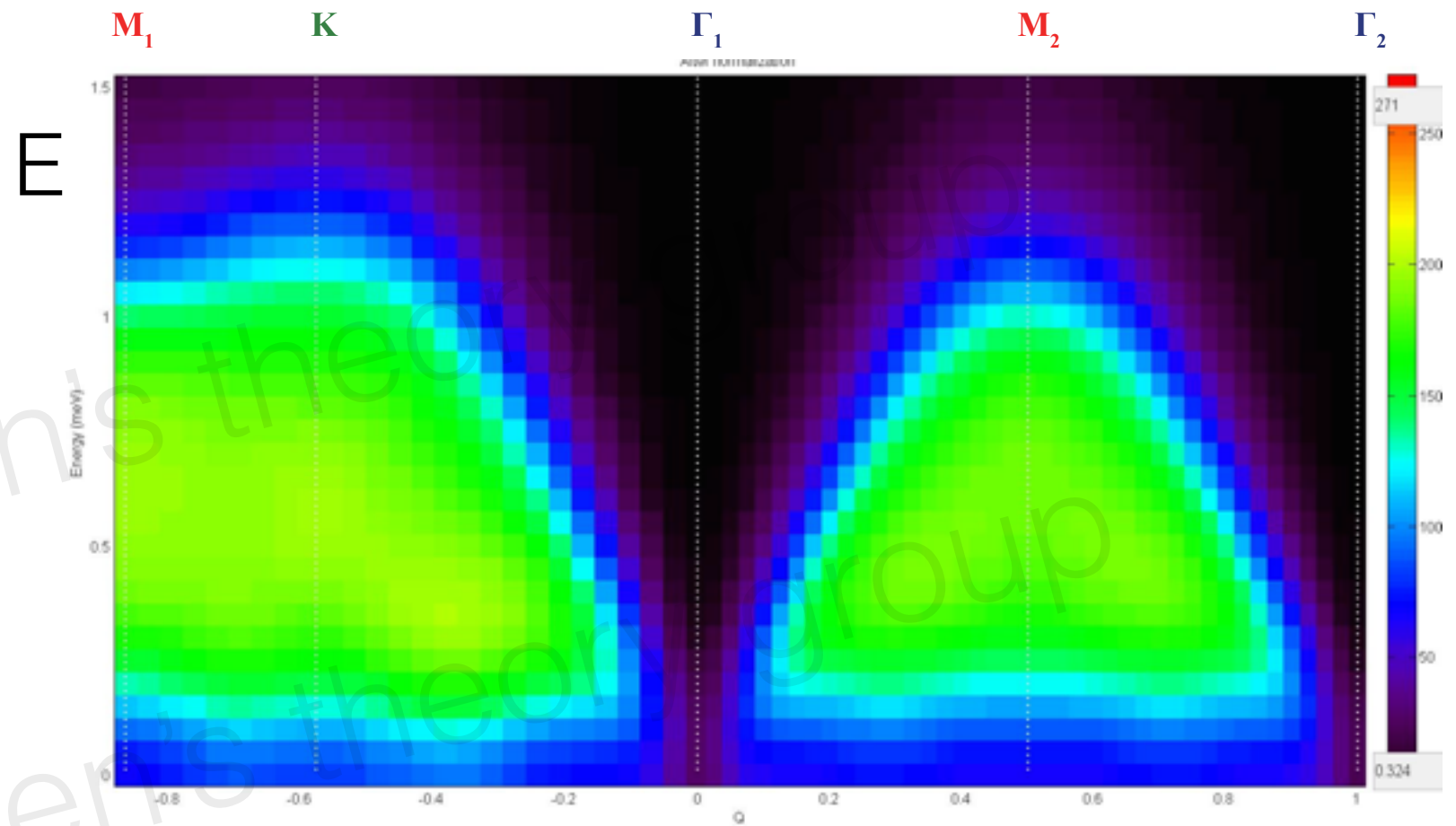
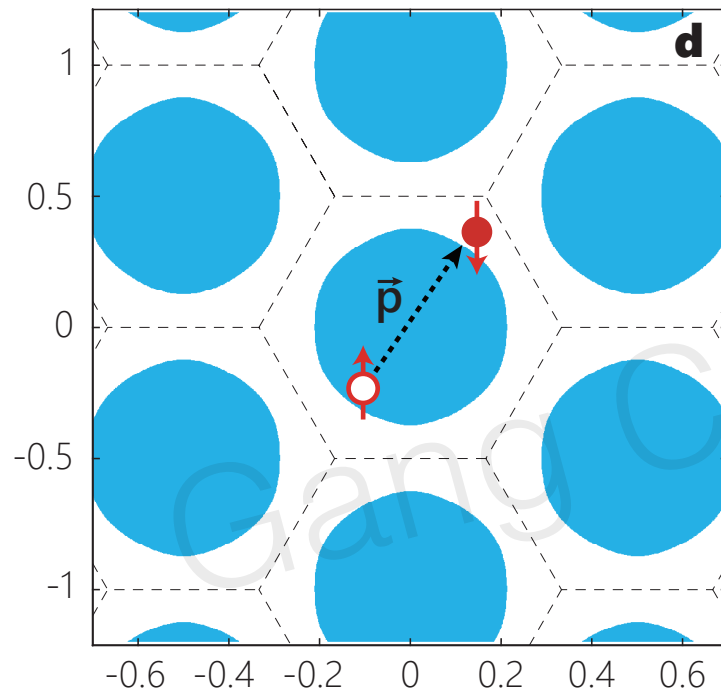


spin wave in $\text{Yb}_2\text{Ti}_2\text{O}_7$
L Savary, et al, PRX 2011



spinon continuum in Cs_2CuCl_4
Masanori, etc NatPhys 2009
but these are **1d spinons** !

Huge spinon continuum at all energies



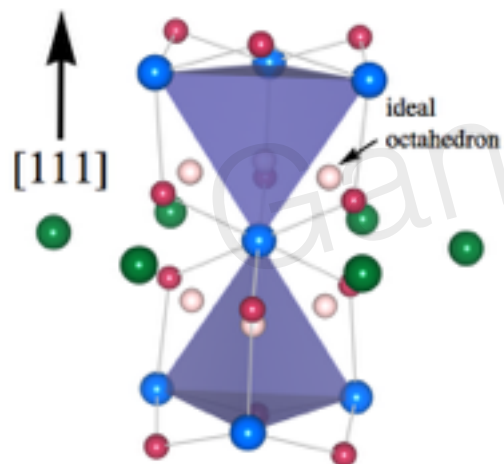
Summary

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.

Dipole-octupole doublet

- Why is this Kramers doublet so special ?

ONE-dimensional representations of the point group



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

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

$$|J^z = +3/2\rangle \xrightarrow{\text{time reversal}} |J^z = -3/2\rangle$$

why special,

they are one dimensional rep of po group.

in particular, if you look at the 3-fold rotation operation, under this rotation each state stay invariant, except a minus sign. they do not transform into each other under the point group transformation.

simple algebra

this is very different from 2-dim irrep where these two states of the doublet are mixed.

These two state are degenerate under time reversal, deg protected by time reversal.

More generally, ...

- Also applies to 4f electron moments on pyrochlore

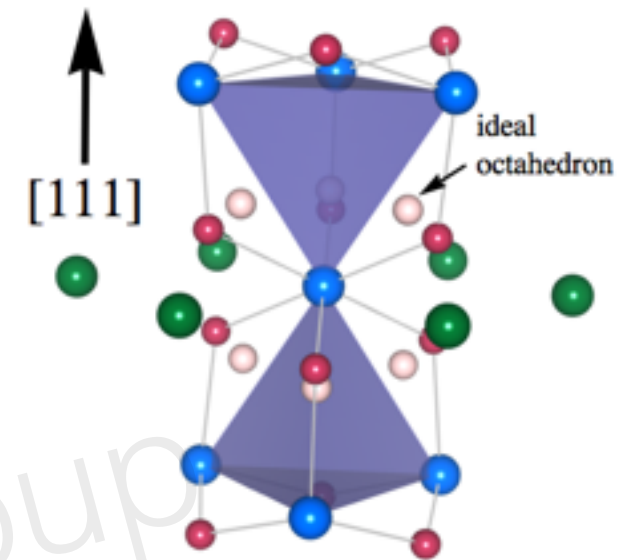
$$J = \frac{3}{2}, \frac{9}{2}, \frac{15}{2}, \dots$$

with the local crystal field Hamiltonian

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

e.g. local doublet wavefunction of Dy^{3+} ($J = \frac{15}{2}$) in $\text{Dy}_2\text{Ti}_2\text{O}_7$

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

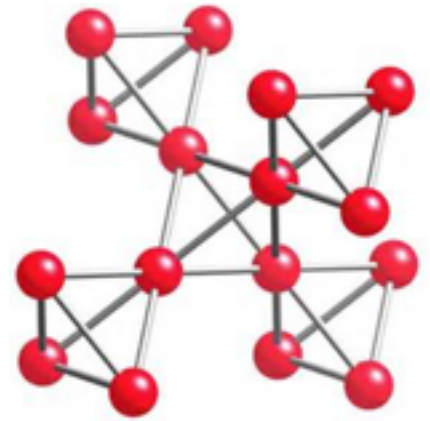


more generally, it applies to xxxx.
if the local crystal field hamiltnian is
easy axis like.

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 Ce³⁺ Pyrochlore Stannate Ce₂Sn₂O₇

Romain Sibille,^{1,*} Elsa Lhotel,² Vladimir Pomjakushin,³ Chris Baines,⁴ Tom Fennell,^{3,†} and Michel Kenzelmann¹

$4f^1$ 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 $50 \pm$

$$\text{Ce}^{3+} (4f^1, {}^2F)$$
$$J = \frac{5}{2}$$

the wavefunction should be of states with odd integer $3/2$.

this really because of the fact that they are one-dimensional, even in the YbTiO₂, such as the excited doublets.

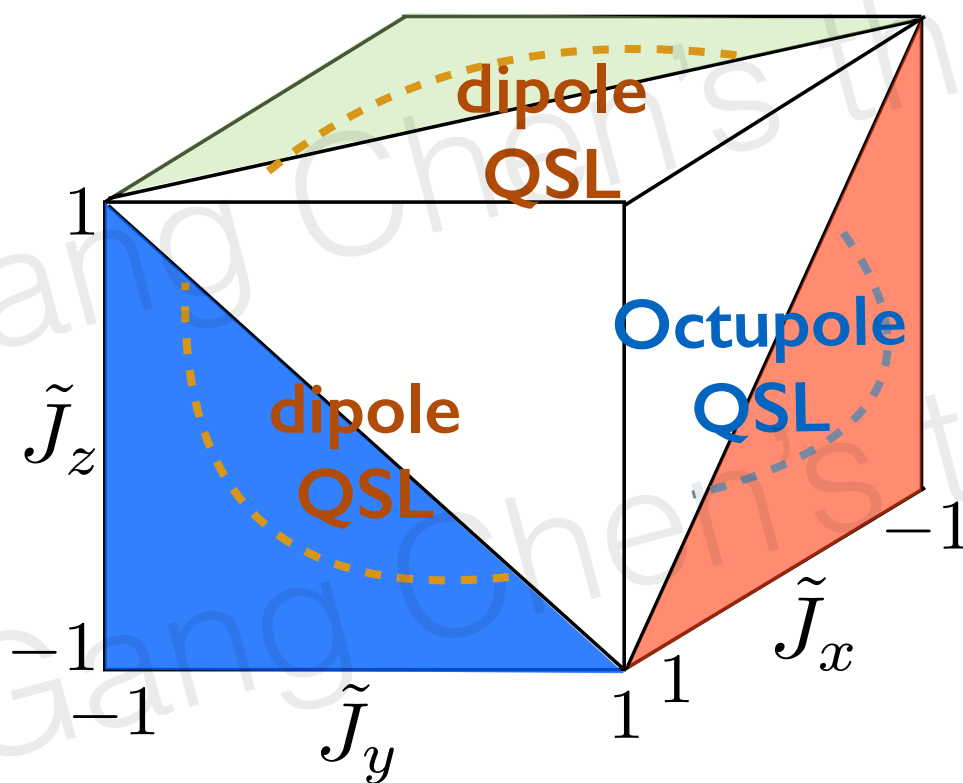
experimentalists of this paper do not know what they really talk about.

But this sentence means a lot to us. It means that gs doublet is a DO doublet, and the model is described by XX

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

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

unlike XXZ model, XYZ model is richer



3D phase diagram

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

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

Emergent Quantum Electrodynamics of U(1) Q

as quantum spin ice is a disordered state, there is no long range order, it is a new phase of matter and cannot be described in the Landau's paradigm of symmetry breaking.

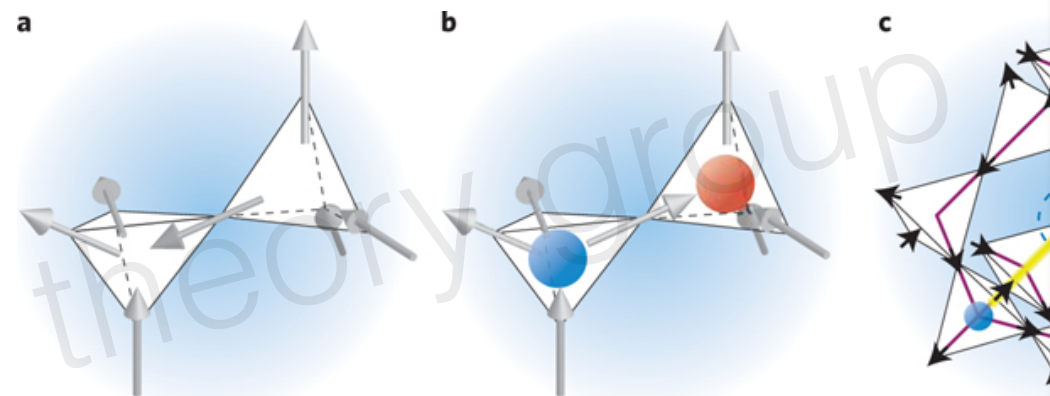
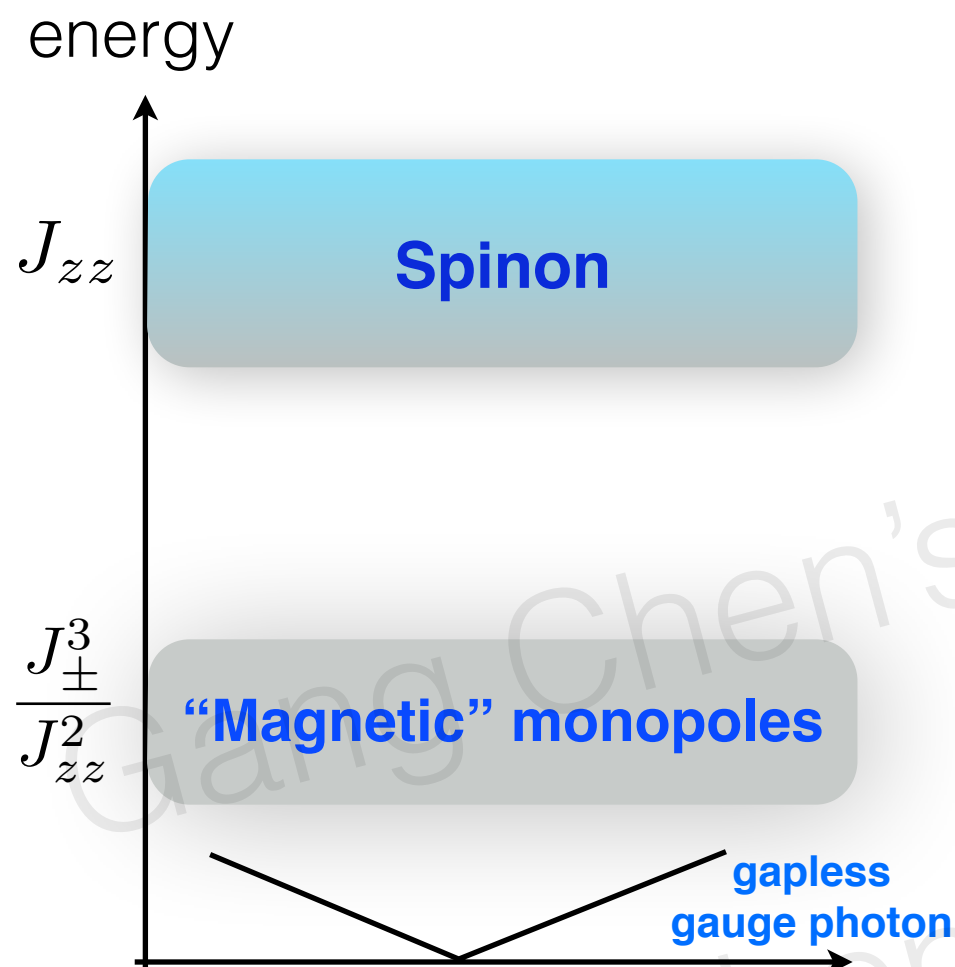
the right description of quantum spin ice is fractionalization and emergence of emergent gauge structure.

there are 3 elementary excitations: spinon, gauge photon, and magnetic monopole. it is not a Goldstone boson, without symmetry breaking, there is no symmetry breaking of emergent gauge structure.

there are deconfined spinons, you can create 2 spinons, you can flip the further spins at an arbitrary distance, it is only a string of spinons. in the image there is a string connecting two spinons, the spin is strongly fluctuating, the spinons are deconfined.

the 3rd is magnetic monopole, a defect of the emergent U(1) gauge field.

It is an exotic phase of matter, it has properties as topological order.



Figs from Mo

Spinon deconfinement

Emergent electric field

$$S^z \sim E$$

Emergent vector potential

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

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

Field-driven Higgs transition

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

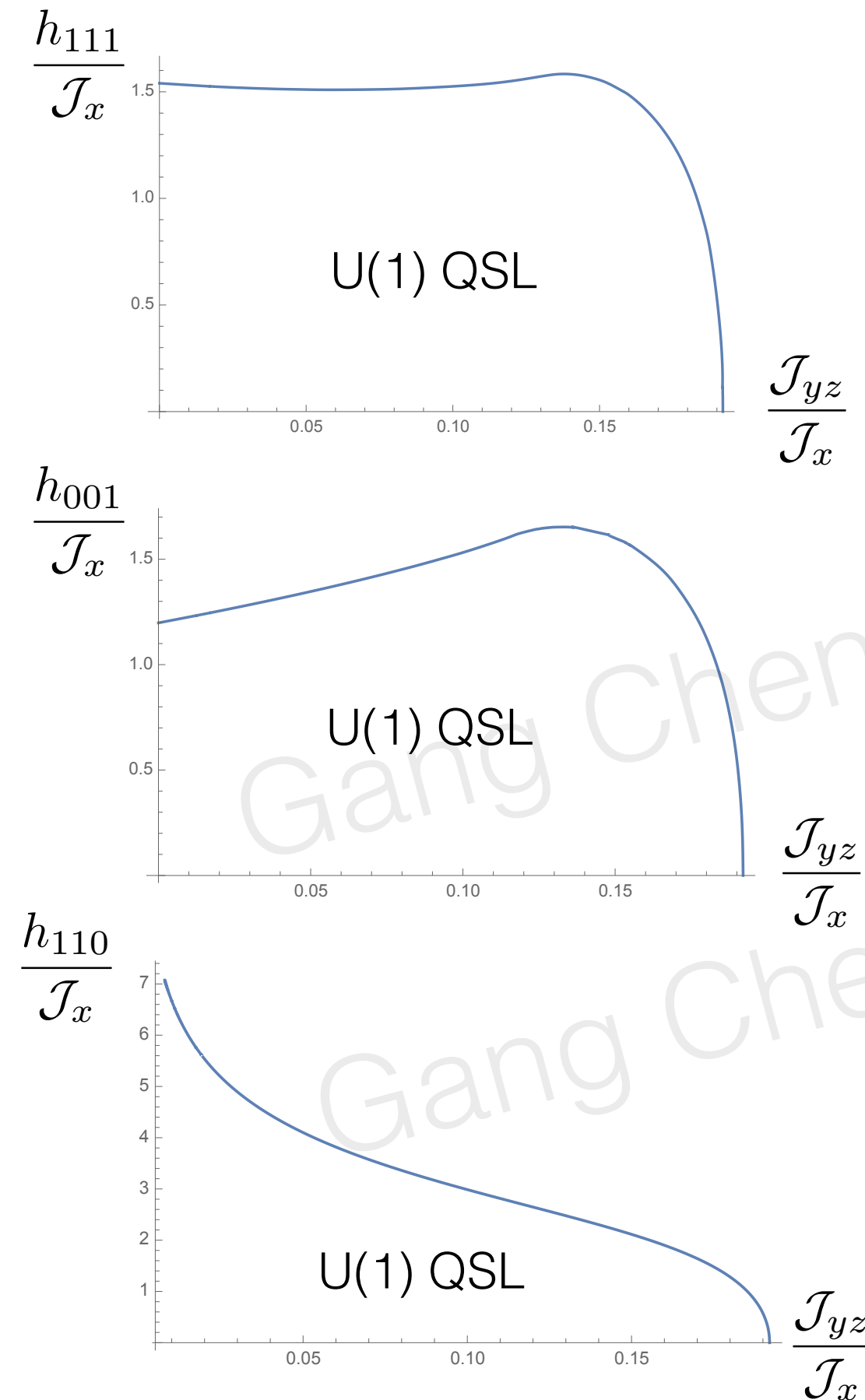
The idea to use a little knob that could simply lead to some clear experimental consequence, very much like the isotope effect of BCS supercond.

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).

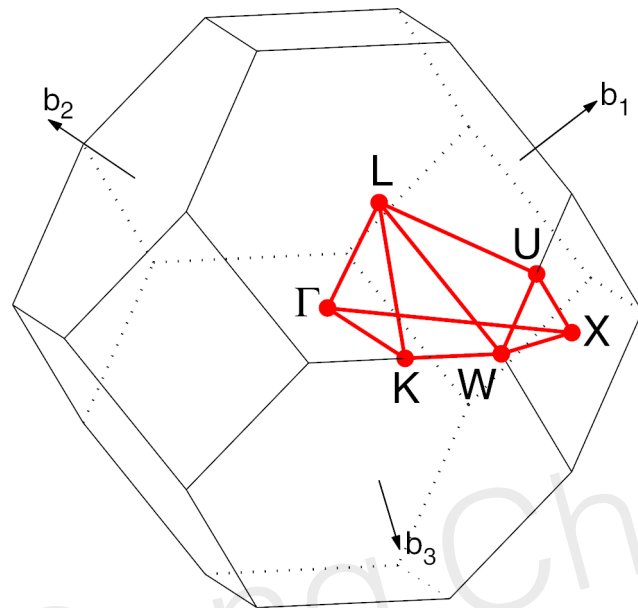
$$H = \sum_{\langle ij \rangle} \mathcal{J}_x \tau_i^x \tau_j^x - \mathcal{J}_{yz} (\tau_i^y \tau_j^y + \tau_i^z \tau_j^z) - h \sum_i \tau_i^z (\hat{n}_i)$$

without losing generality

Higgs transition is very much like
meissner effect in superconductor



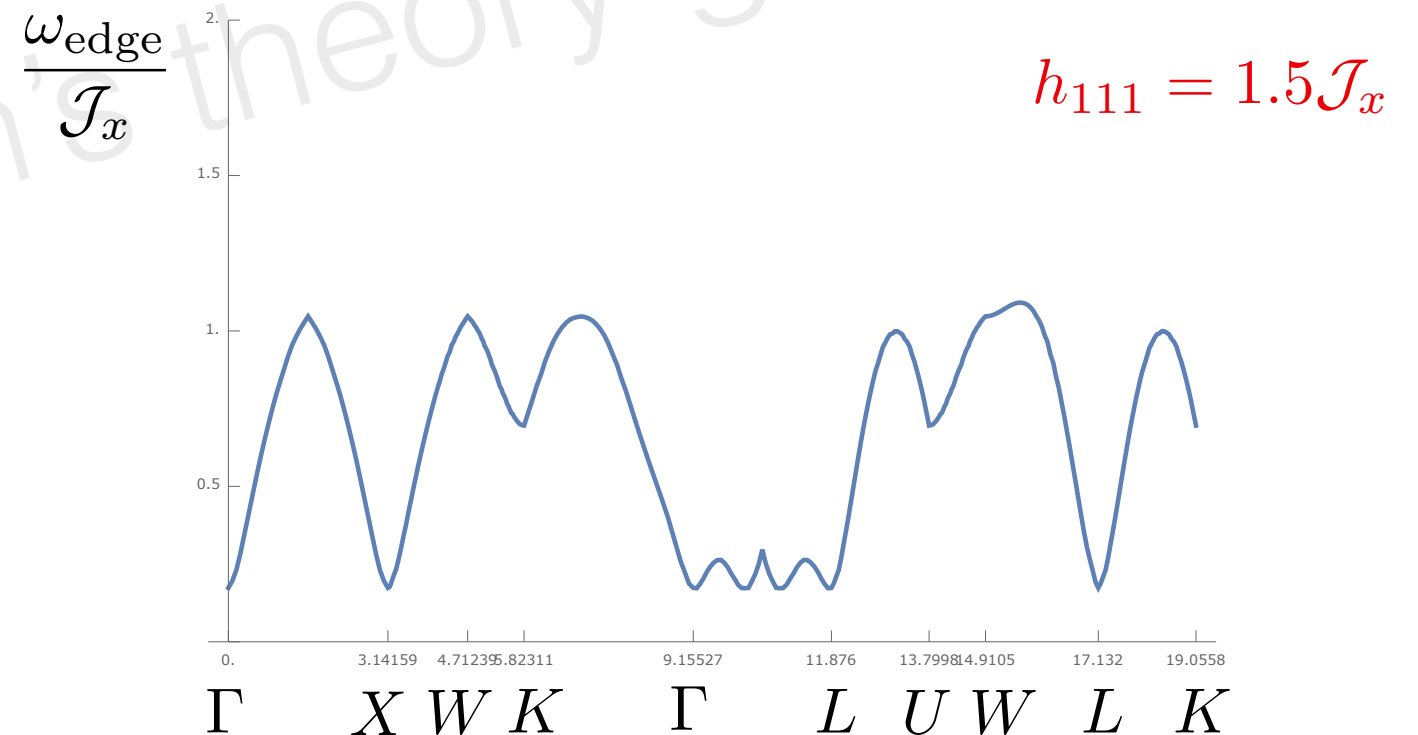
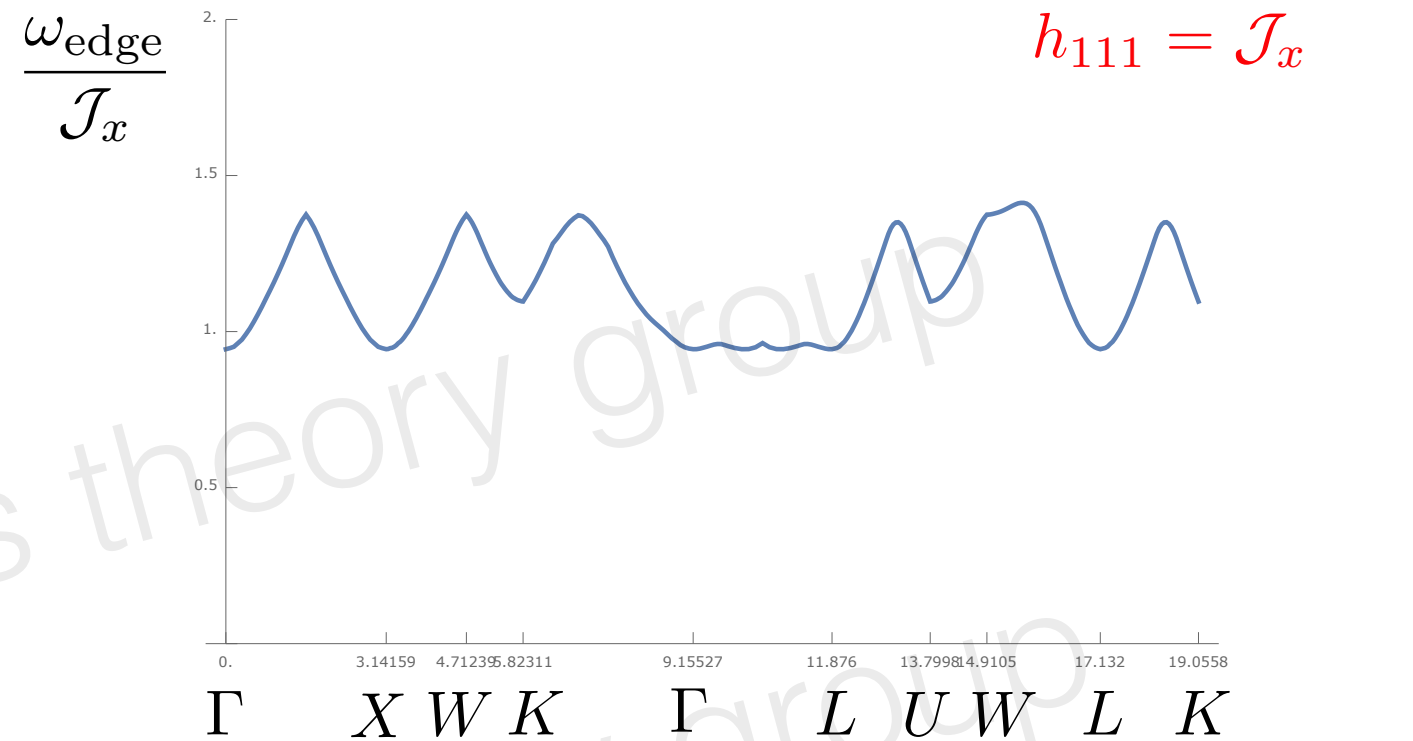
Lower excitation edge



FCC path: Γ -X-W-K- Γ -L-U-W-L-K|U-X

[Setyawan & Curtarolo, DOI: 10.1016/j.commatsci.2010.05.010]

Brioullin zone of
FCC lattice



Neutron scattering and thermal transport

Dipolar- $U(1)$ QSL

neutron spin couples to both gauge field and matter field, observe both gapless gauge photon and gapped spinon continuum

Octupolar- $U(1)$ QSL

neutron spin only couples to the matter field (spinons), observes only the gapped spinon continuum. External magnetic field can manipulate the spinon continuum, which can be confirmed by neutron scattering.

Thermal transport

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

It can be beneficial to observe the low temperature peak in the thermal transport.