SYMMETRY ENRICHED QUANTUM SPIN ICES ON THE PYROCHLORE LATTICE

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Collaborators

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- Michael Hermele (CU Boulder)
- Ref: arXiv 1311.1231 (more stuff)

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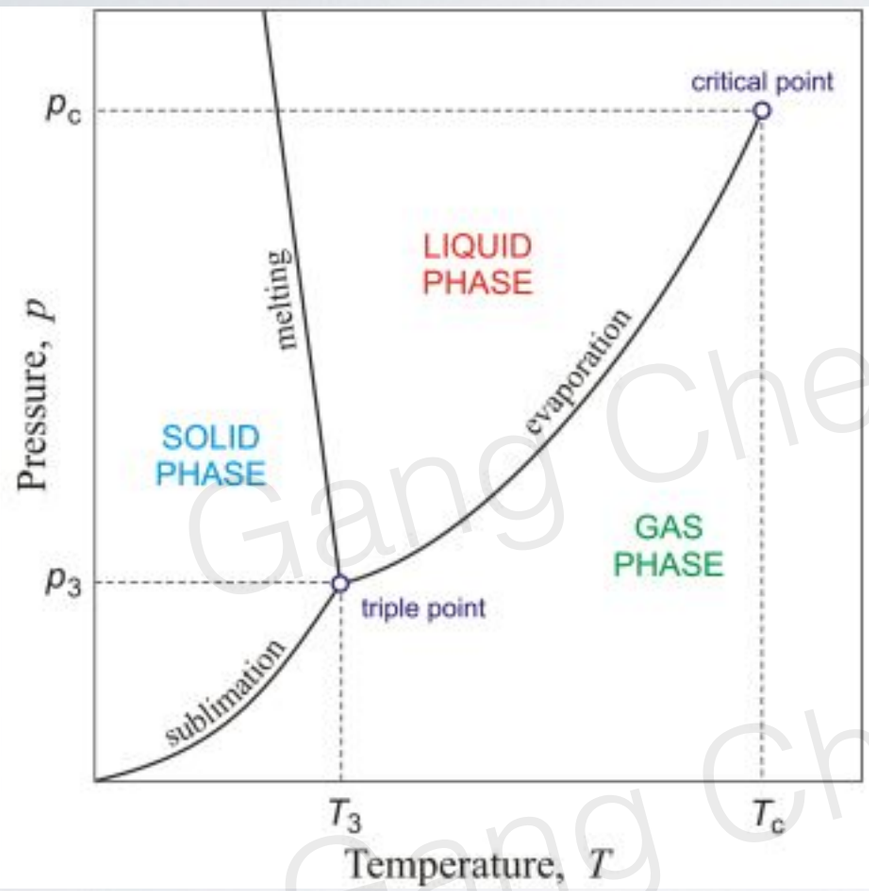
OUTLINE

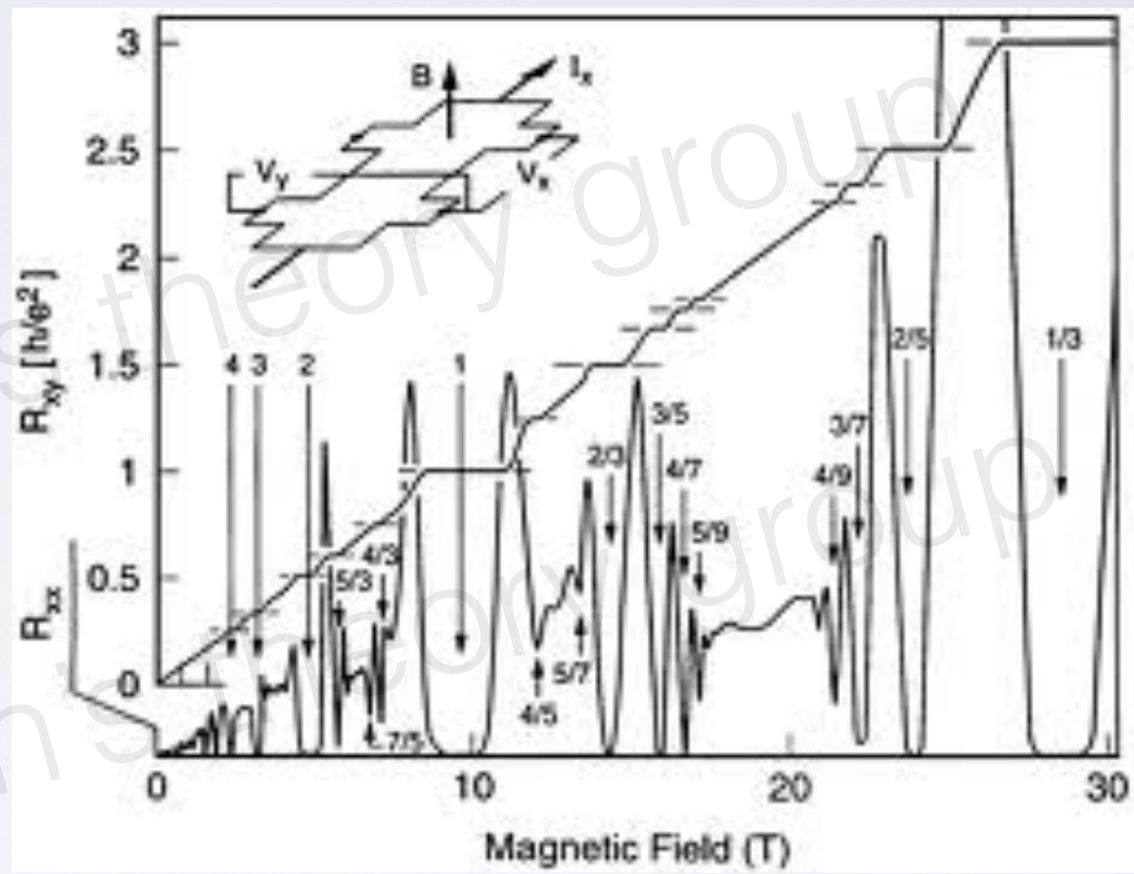
- Quantum spin liquids: review of history, theories, experiments
- Classical spin ice and quantum spin ice
- A realistic spin model on the pyrochlore lattice
- Symmetry enrichment and other prediction

WHY DO WE CARE ABOUT QSLs?

It breaks the Landau paradigm of phases and phase transitions.



Liquid-gas transition: Landau symmetry breaking theory.

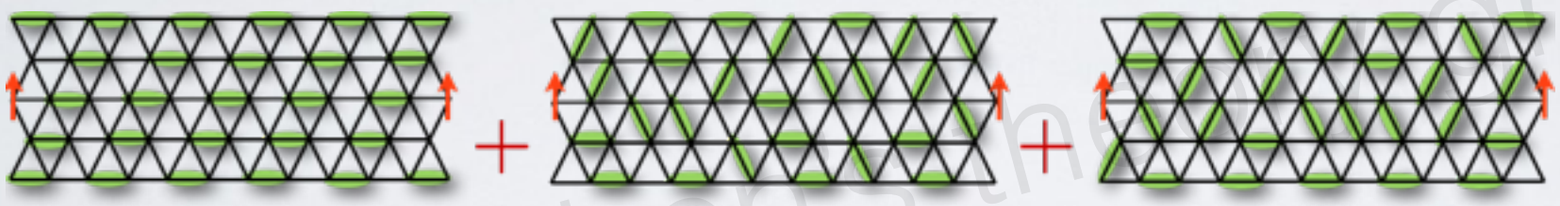


FQHE: no symmetry breaking, ground state degeneracy not protected by symmetry, fractional excitation, emergent Chern-Simons gauge theory…

We want to look for exotic phases beyond FQHE. QSLs provide such a possibility both in theory and experiments.

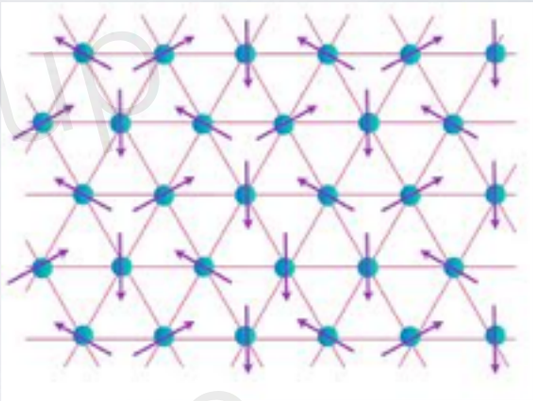
HISTORY

- 1973, Anderson's RVB wavefuc for triangle lattice Heisenberg model to suppress AFM Neel order



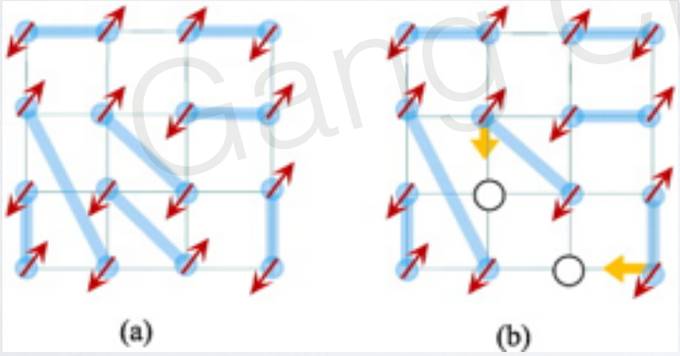
+ all other dimer covering

each bond=spin singlet, quantum superposition



noncollinear order

- 1987, high temperature superconductor without phonons



RVB state becomes a superconductor upon doping with mobile holes.
RVB wavefct is a superconductor wavefct after doping.

THEORY (INCOMPLETE)

- Definition of QSL

  historical/experimental definition
  Properties: states w/ fractional excitations (quantum number, e.g. spin-1/2 spinons, statistics), states w/ emergent gauge structure (to "deconfine" the fractional excitation.

- Classification

  2002, Wen's PSG: an example of $H^2(SG, GG)$ for Spinons.
  Recent SET: classify symmetry fractionalization patterns of fractional excitations $H^d(SG,GG)$.

- Exact solvable models

  Kitaev's toric code
  Kitaev model on honeycomb lattice,
  many new ones……

- Numerics (and EE): ED (small system), QMC (sign problem), DMRG (1d, quasi-1d), etc

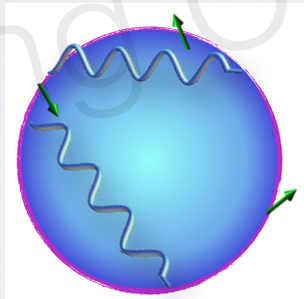
  Heisenberg model on kagome lattice,
  J_1-J_2 model on square lattice, honeycomb lattice
  ……

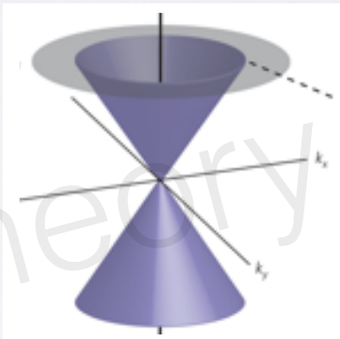
VARIOUS QSLS (OFTEN DISCUSSED ONES)

- Critical spin liquids

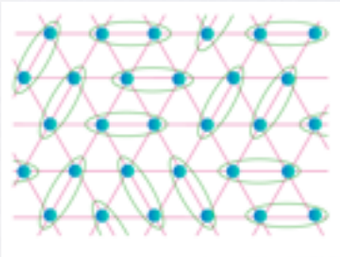
  e.g. spinon non-FL in 2D: spinon Fermi sea+U(1) gauge

  Dirac spin liquid: Dirac spinon+U(1) gauge





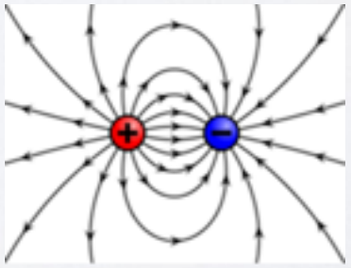
Anderson's RVB



- Topological spin liquids (gapped spinons)

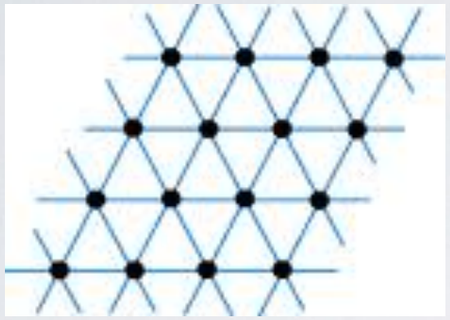
  e.g. Z_2 spin liquid in 2d or 3d

  QSI: U(1) spin liquid in 3d



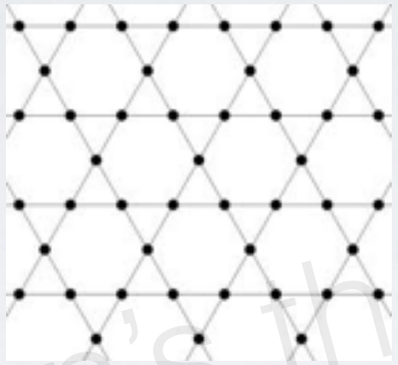
QSL CANDIDATES (INCOMPLETE)

- Solid-state systems (explain one)



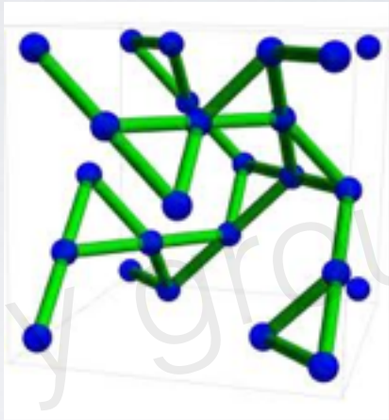
2d-triangle

materials
$EtMe_3Sb[Pd(dmit)_2]_2$
κ-$(ET)_2Cu_2(CN)_3$
$Ba_3CuSb_2O_9$
$Ba_3NiSb_2O_9$

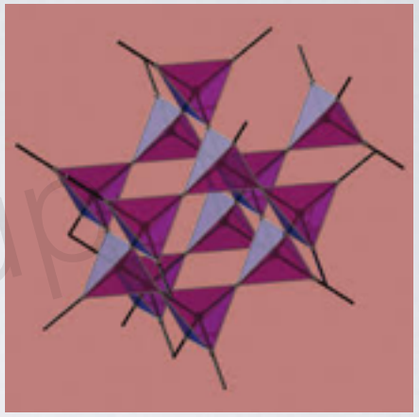


2d-kagome

materials
$ZnCu_3(OH)_6Cl_2$
$Cu_3V_2O_7(OH)_2{\cdot}2H_2O$
$BaCu_3V_2O_3(OH)_2$



3d-hyperkagome



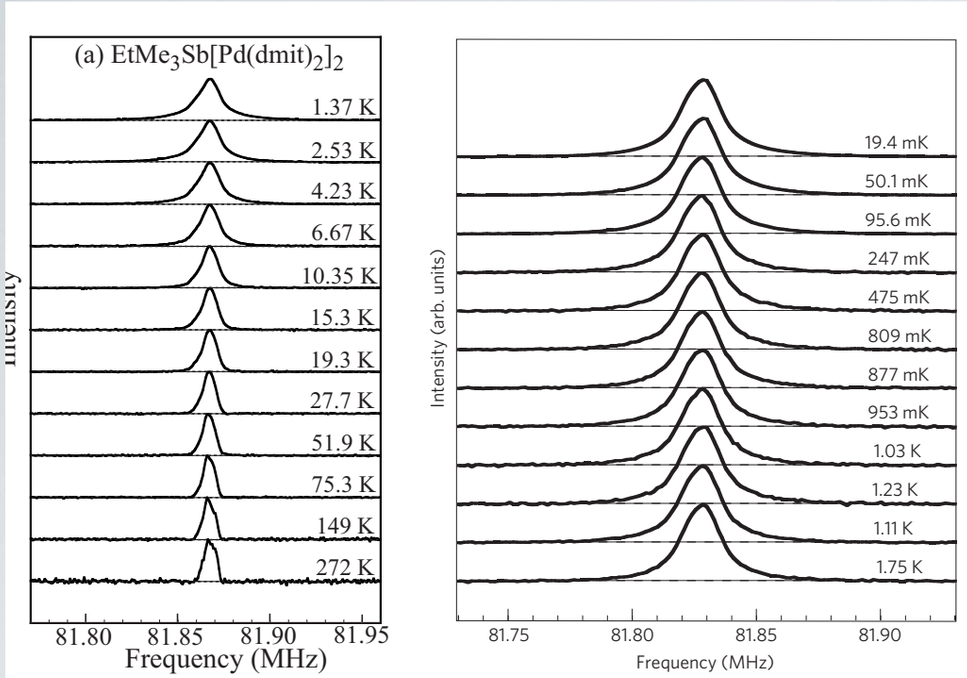
3d-pyrochlore

strong SOC material
$Na_4Ir_3O_8$
Ba_2YMoO_6
$Pr_2Zr_2O_7$
$\ldots$

- "Ultracold" atoms: dipolar magnetism, large quantum group [SP(N) or SU(N) with reasonably large N !]

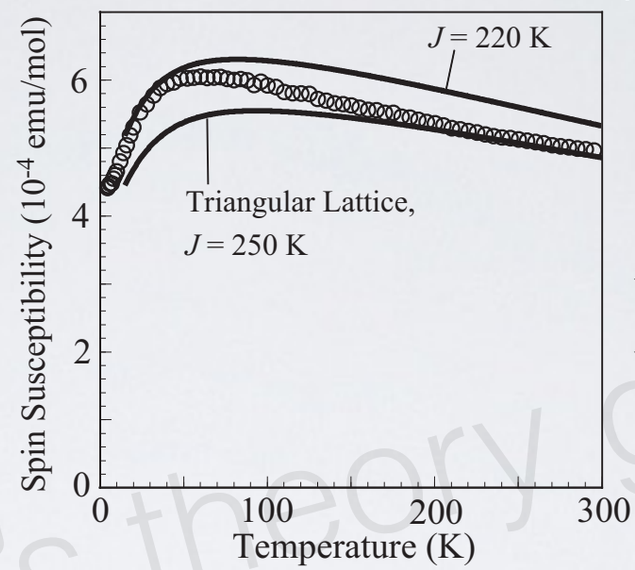
Issue: prepare/design, cooling, probing

AN EXPERIMENTAL ONE $EtMe_3Sb[Pd(dmit)_2]_2$



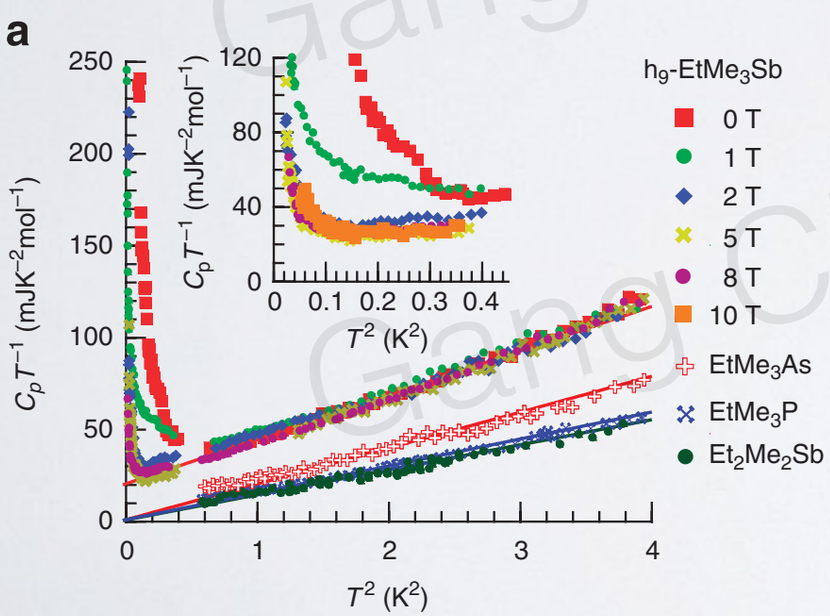
no magnetic ordering/freezing from NMR

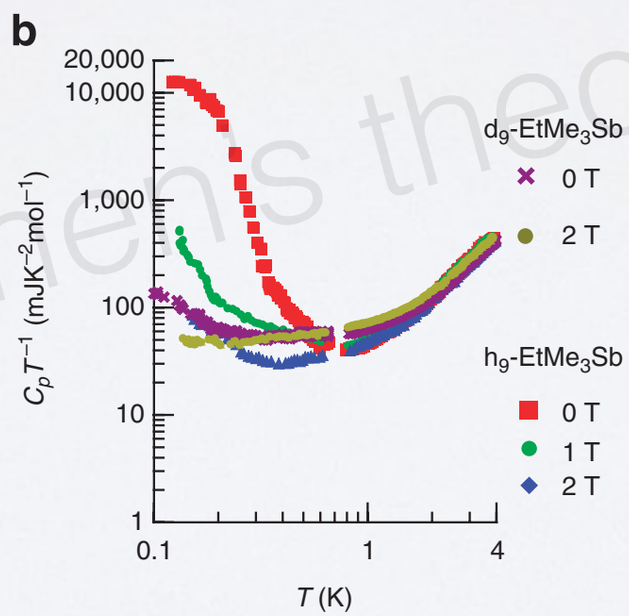
Spinon Fermi Surface ??



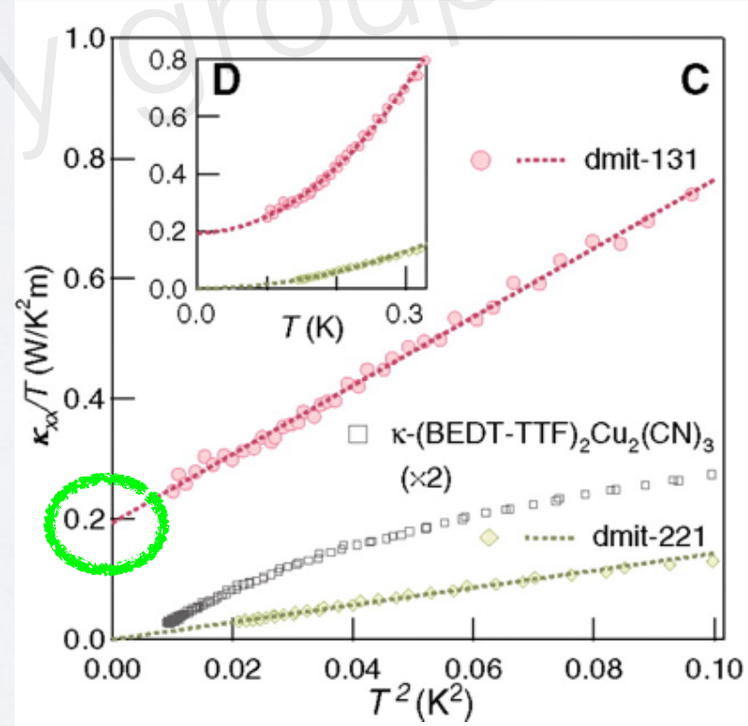
Itou, etc, PRB 2008
Yamashita, etc Science 2011
Yamamoto etc, NatComm 2011

constant spin susceptibility





some complication: but consistent with large density of states at low energies



metallic thermal conductivity

CHALLENGES: CONNECT THEORY WITH EXPERIMENTS

- Modelling:
  Given a QSL candidate, want to know if it is QSL and what type of QSL. Require the Hamiltonian and "solving" it. Both are hard.

- Measurement:
  How to unambiguously confirm QSLs of any type by experiments on a given QSL candidate. Not just absence of ordering at low Ts. More direct probe for the "defining" properties of QSL, e.g. emergent gauge structure, deconfined (fractionalized) degrees of freedom.

OUTLINE

- Quantum spin liquids: review of history, theories, experiments
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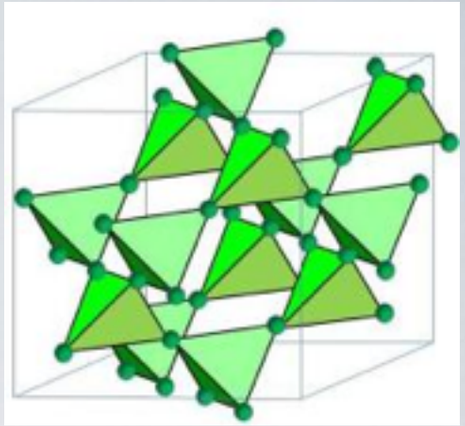
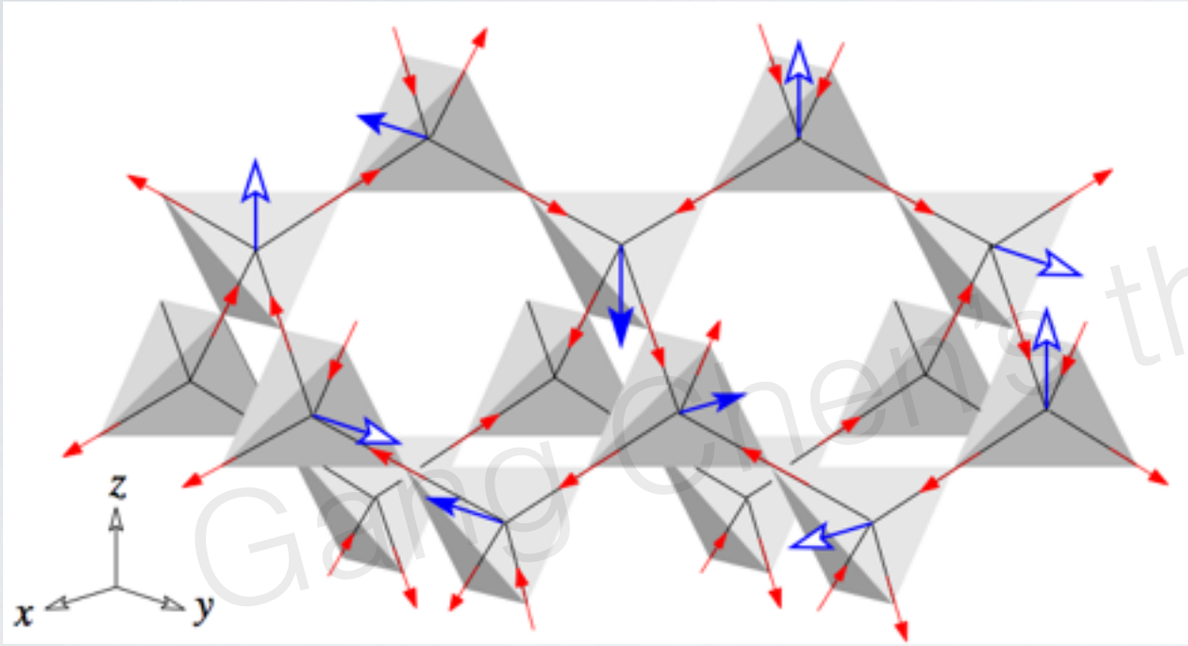
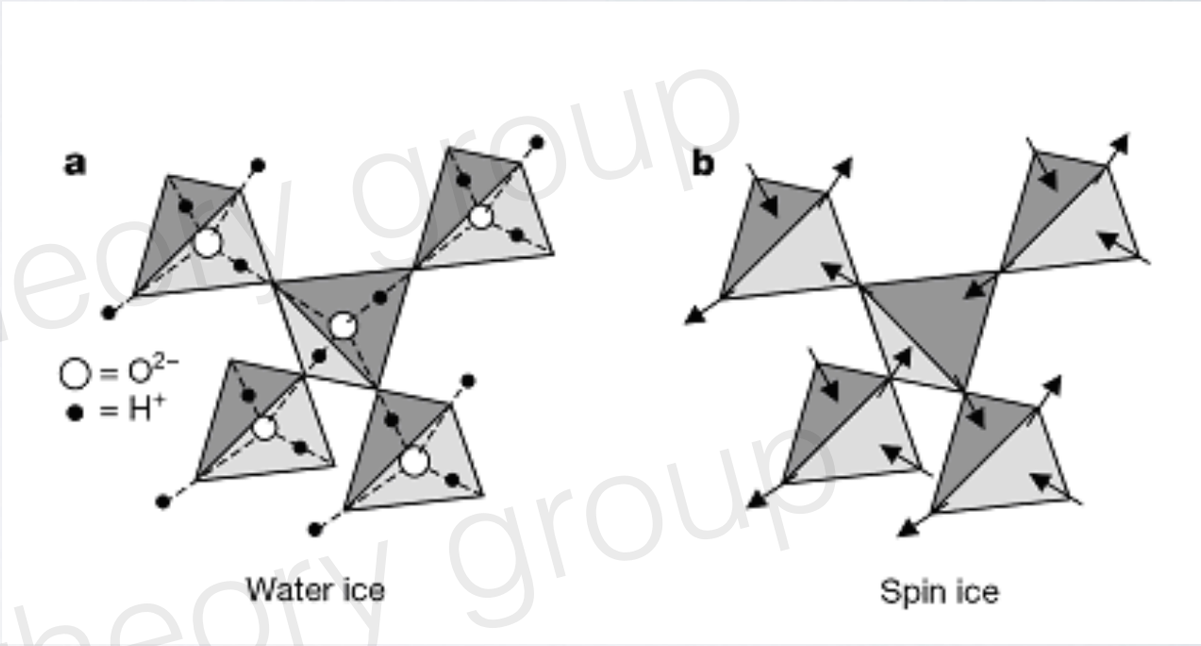
- A realistic spin model on the pyrochlore lattice
- Symmetry enrichment and other prediction

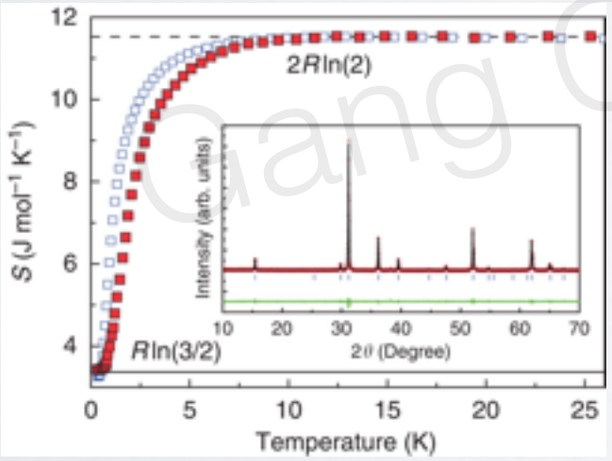
CLASSICAL SPIN ICE
(WHAT WE UNDERSTOOD VERY WELL)



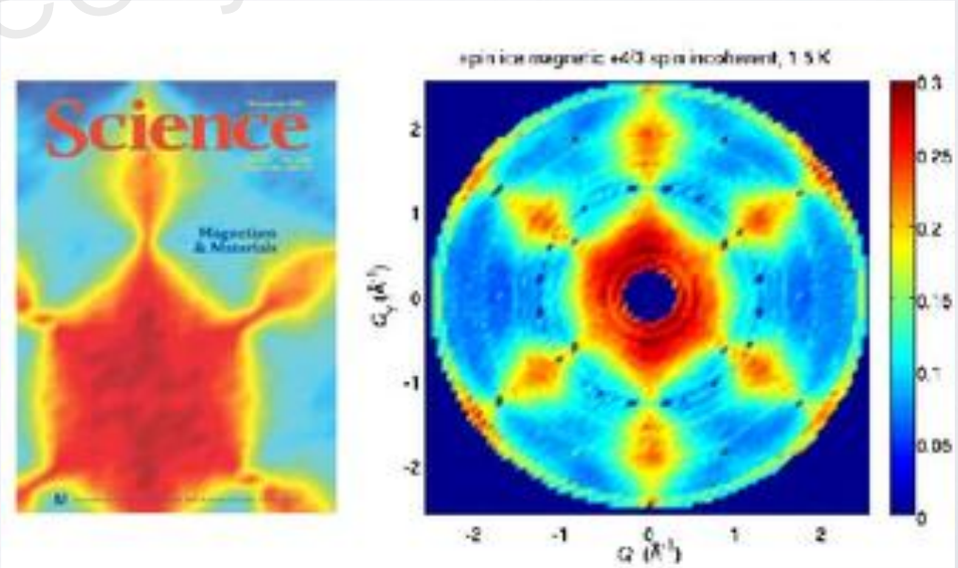
- Ising spins on pyrochlore lattice







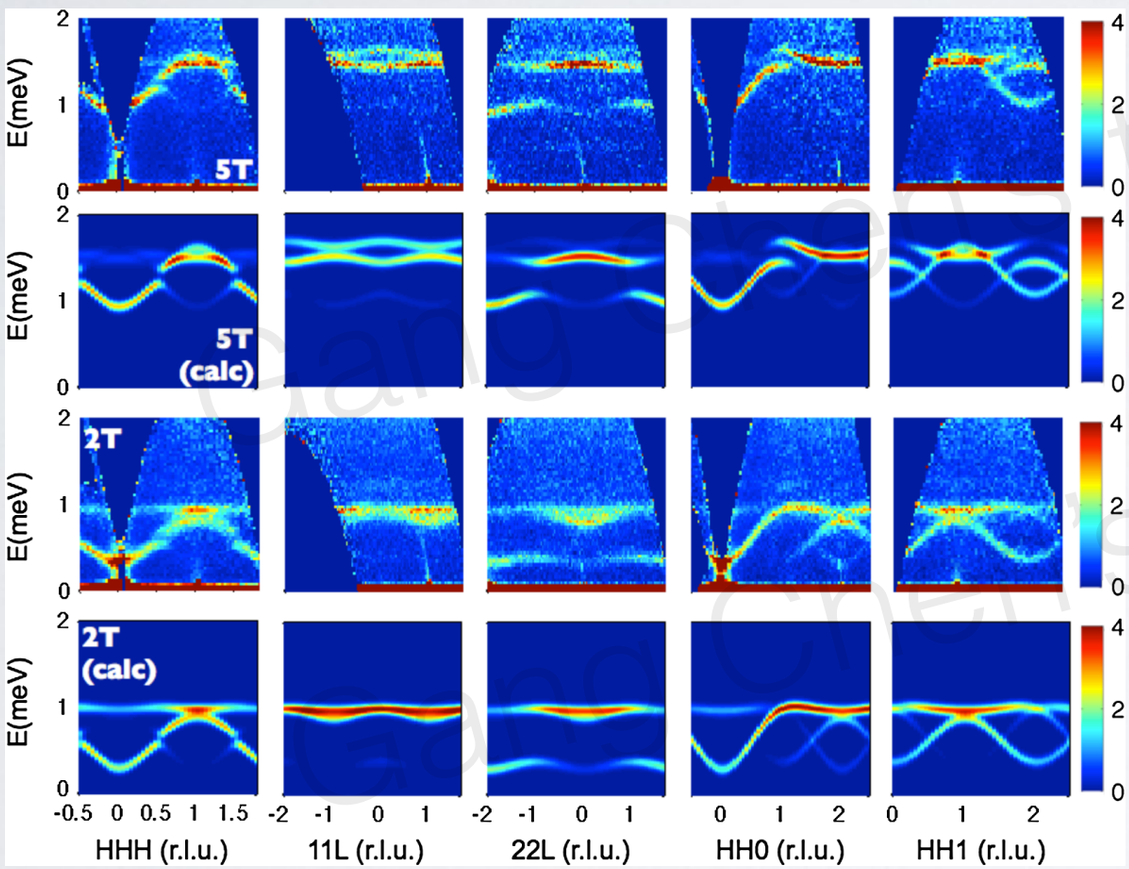
Residual Pauling entropy
Zhou, etc, nat. comm. 2011



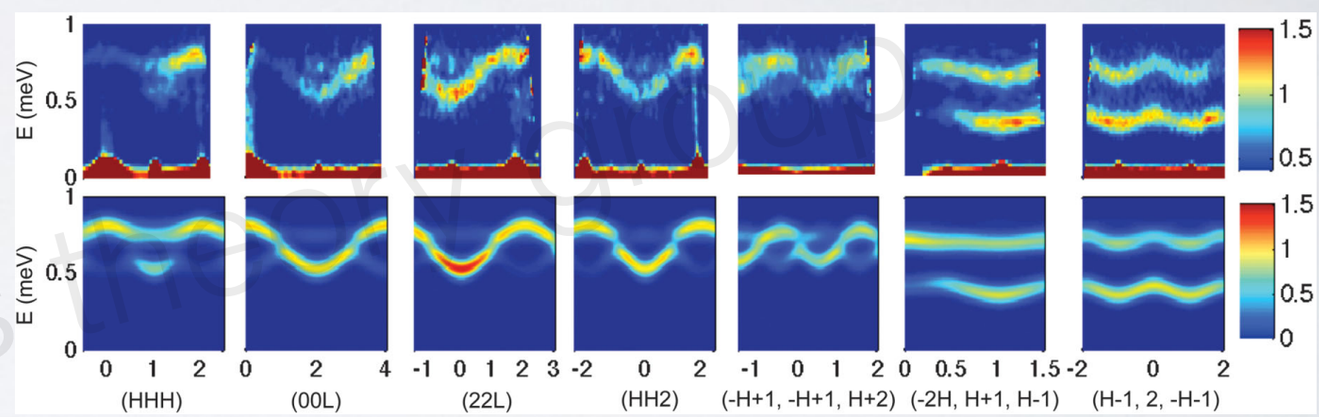
pinch points in neutron scattering:
dipolar like spin correlation

QUANTUM MECHANICS IS IMPORTANT!

- Experiments



Well-defined spin waves in $Yb_2Ti_2O_7$ by Prof B. Gaulin's group



Spin waves and quantum order-by-disorder in $Er_2Ti_2O_7$ also by Prof B. Gaulin's group

Prof Gingras' review 2013

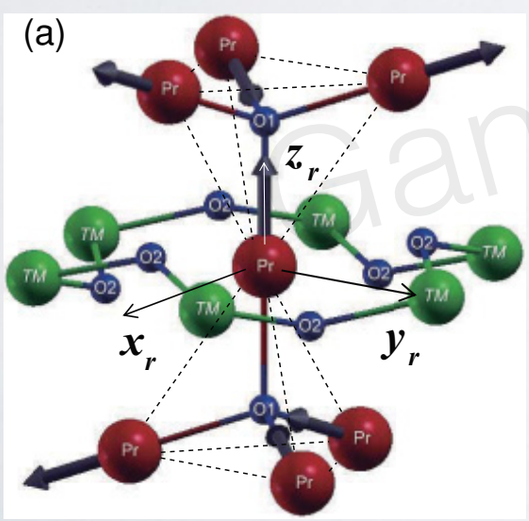
also in many other experiments…

ISING SPIN IS NOT ENOUGH

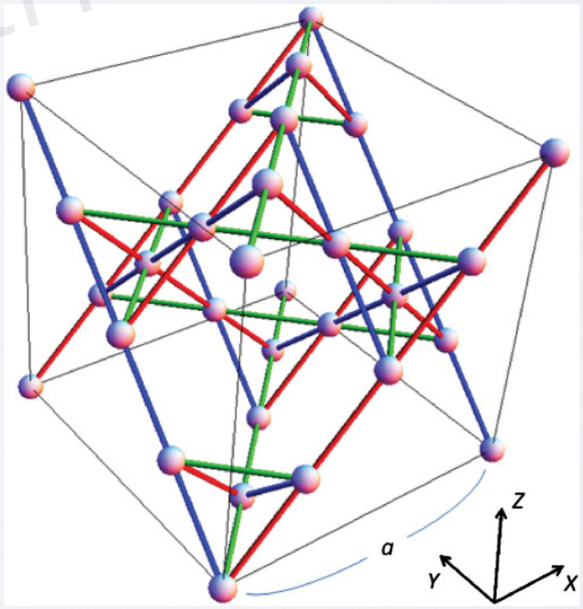
- **A** generic Hamiltonian for effective spin-1/2 (S. Onoda etc)

$$H = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{\pm\pm} (\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^-) + J_{z\pm} [\mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j] \},$$

Complicated!



spins are defined in local coordinate system



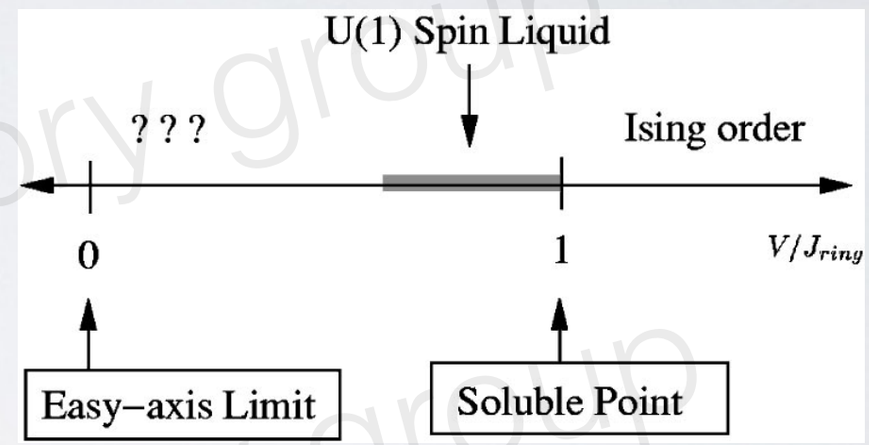
exchange has bond-dependent phase factors

Prof Gingras' review 2013

QUANTUM SPIN ICE

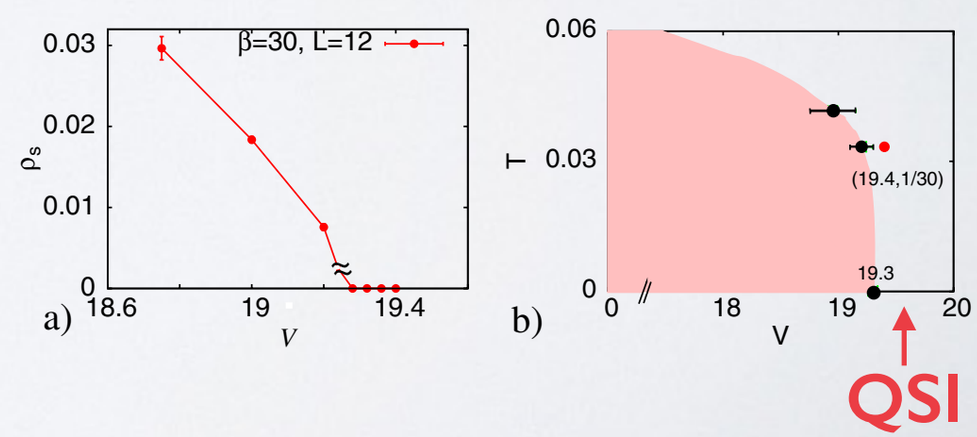
- "Toy" XXZ model (Hermele etc 2004)

$$H = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+)$$



- Quantum Monte Carlo (Isakov etc 2008)

$$H = \sum_{\langle ij \rangle} [V(n_i - 1/2)(n_j - 1/2) - t(b_i^\dagger b_j + b_i b_j^\dagger)]$$
$$+ \sum_i [U(n_i - 1/2)^2 - \mu n_i].$$



DIFFERENCE BETWEEN CLASSICAL AND QUANTUM

- Classical spin ice: basically classical stat mech with local constraints (ice rule), i.e. a thermal/entropy effect

  Residual Pauling entropy
  Dipolar spin correlation ($1/r^3$)

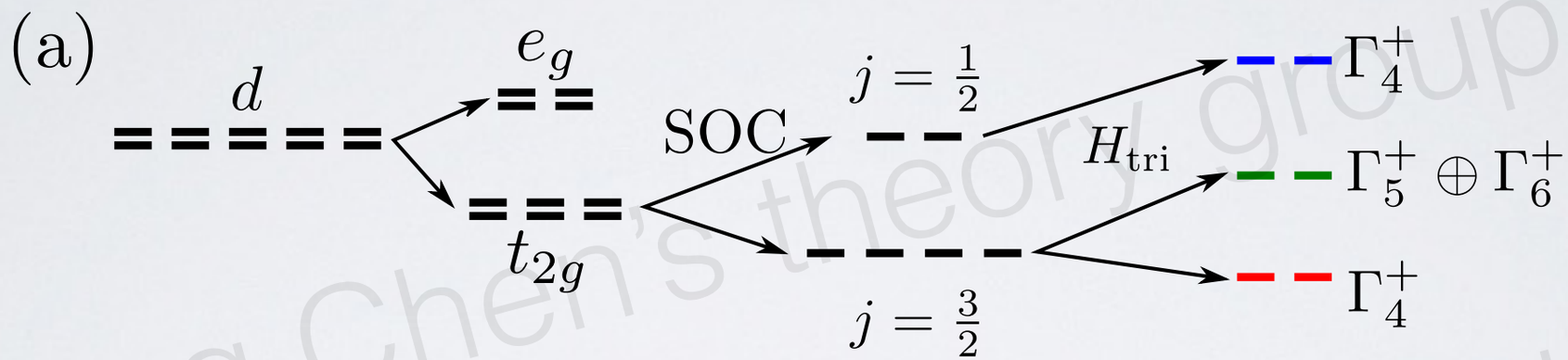
- Quantum spin ice: string-net condensation, emergent quantum electrodyna
  a novel quantum phase of matter.

  Deconfined (coherent) spinons
  Emergent photons

  No residual Pauling entropy
  T^3 heat capacity
  Power-law spin (S^z) correlation
  Defects induced ground state degeneracy

OUR DOUBLET: LOCAL PHYSICS

- Local moments on pyrochlore lattice: pseudospin 1/2



$$\begin{cases} j^z = 3/2 \\ j^z = -3/2 \end{cases}$$

a simple picture for d electrons
under D_{3d} point group crystal field

- Why is this Kramers doublet so special?

1-dimensional representations of the point group!

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

OUR DOUBLET: LOCAL PHYSICS

- Also applies to f electron moments on pyrochlore

$$j = 3/2, 9/2, 15/2, \cdots$$

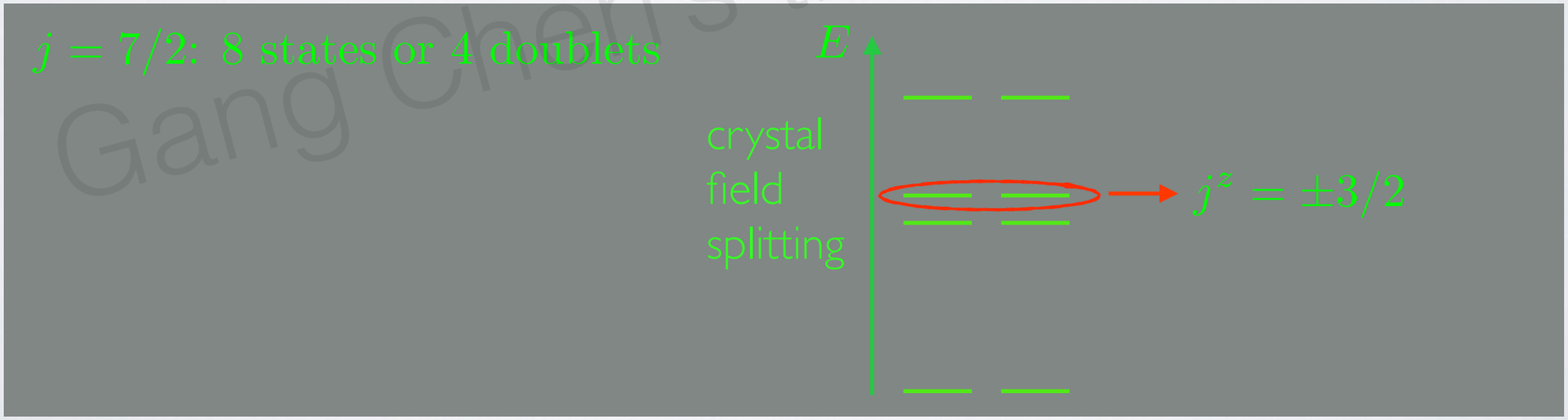
Local crystal field Hamiltonian

$$H_{\rm cf} = 3B_2^0 (J^z)^2 + \cdots$$

$$\text{if } B_2^0 < 0.$$

- may generally apply to any Kramers' spin (j>1/2)

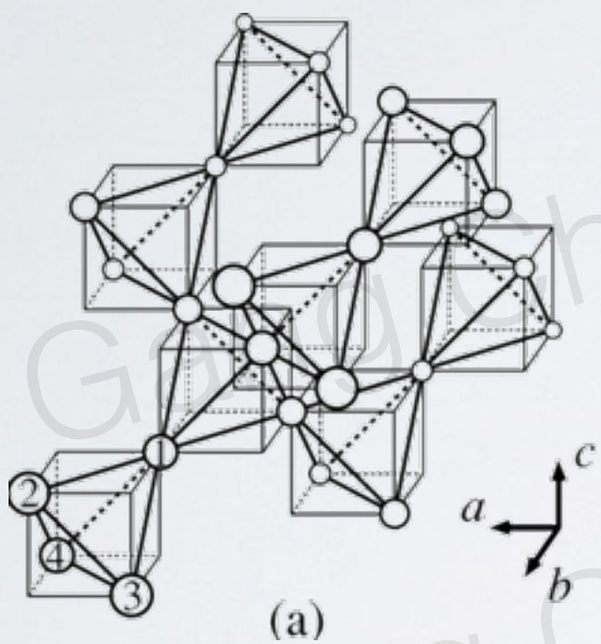
For example, Yb:$Yb_2Ti_2O_7$



Bertin, etc, J. Phys: cond.mat 2012

SYMMETRY PROPERTIES

- Space group symmetry



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

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

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

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

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

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

OUR MODEL

- Nearest neighbour exchange from symmetry

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

Same on every bond !

Versus S. Onoda, L. Balents' model

$$H = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm}(\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{\pm\pm}(\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^-) + J_{z\pm}[\mathsf{S}_i^z(\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j] \},$$

Apply a global rotation around y axis in the pseudospin space and obtain XYZ model

$$H = \sum_{\langle ij \rangle} \tilde{J}_z \tilde{S}_i^z \tilde{S}_j^z + \tilde{J}_x \tilde{S}_i^x \tilde{S}_j^x + \tilde{J}_y \tilde{S}_i^y \tilde{S}_j^y$$

UNFRUSTRATED REGIME: MAGNETIC ORDER

1. $\tilde{J}_z < 0$ and $|\tilde{J}_z| \gg \tilde{J}_{x,y}$, then $\langle \tilde{S}_i^z \rangle \neq 0$.

   This is an "all-in all-out" AFM state with magnetic dipolar order.

2. $\tilde{J}_x < 0$ and $|\tilde{J}_x| \gg \tilde{J}_{y,z}$, then $\langle \tilde{S}_i^x \rangle \neq 0$.

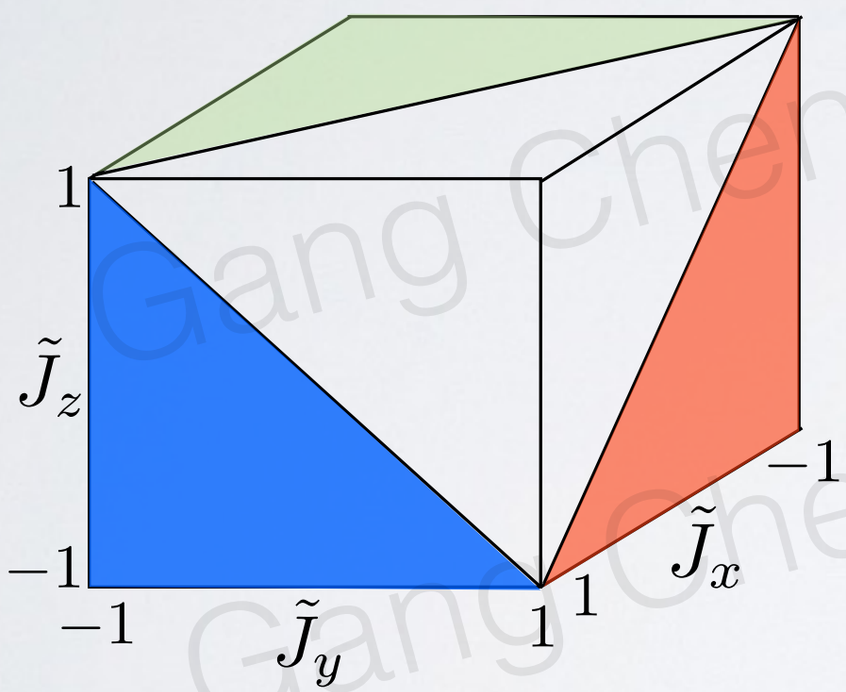
   This state is not distinct from the first state.

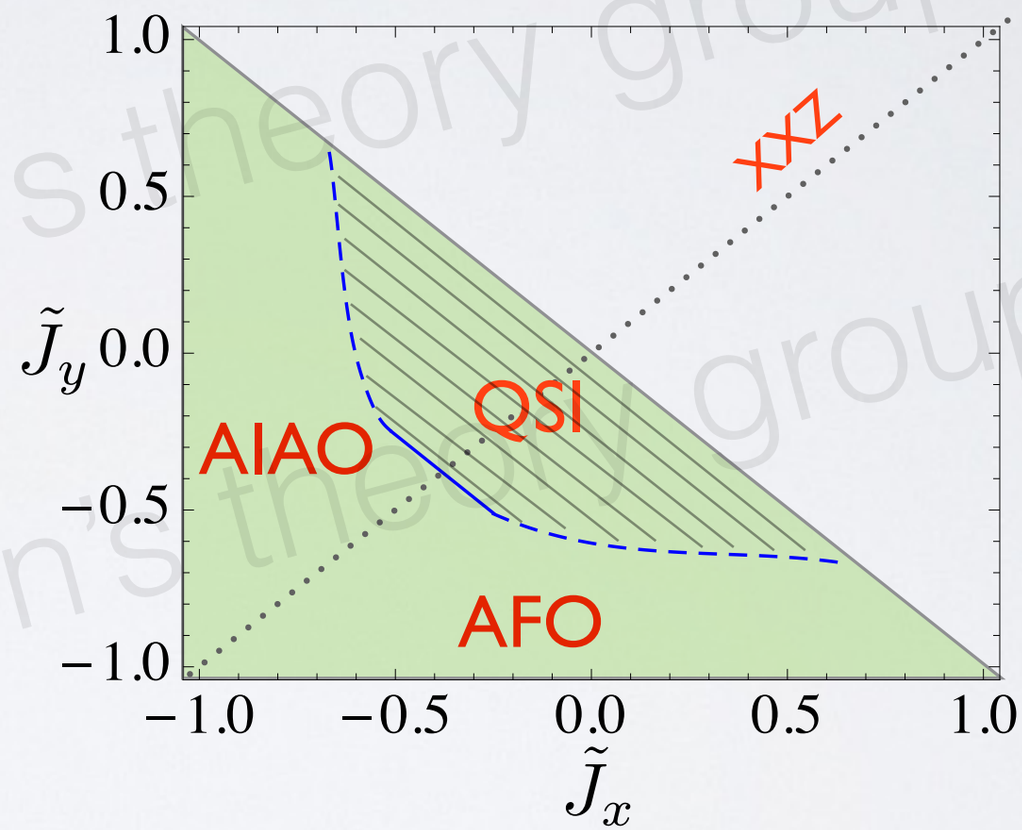
3. $\tilde{J}_y < 0$ and $|\tilde{J}_y| \gg \tilde{J}_{x,z}$, then $\langle \tilde{S}_i^y \rangle \neq 0$.

   This state is distinct from the above two states!
   It has an antiferro-octupolar order but no dipolar order.

FRUSTRATED REGIME: MAGNETIC ORDER

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





$\tilde{J}_z = 1$: phase diagram by gauge mean field theory.

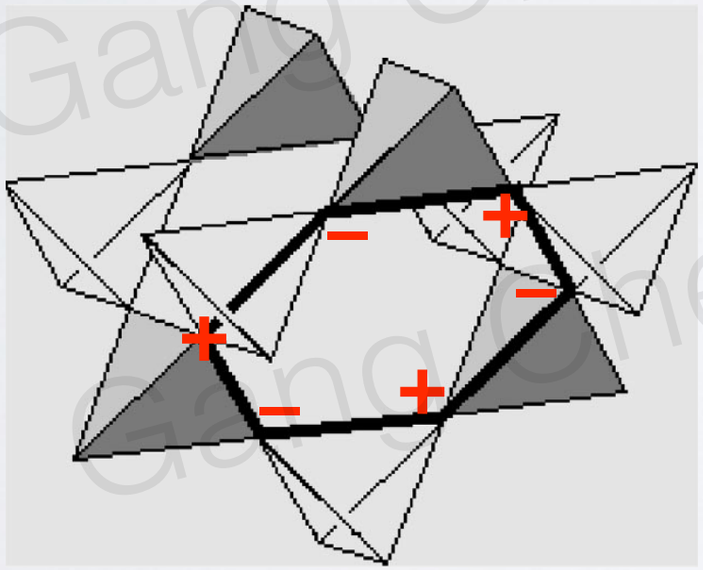
No sign problem in the shaded region !

PERTURBATIVE ANALYSIS

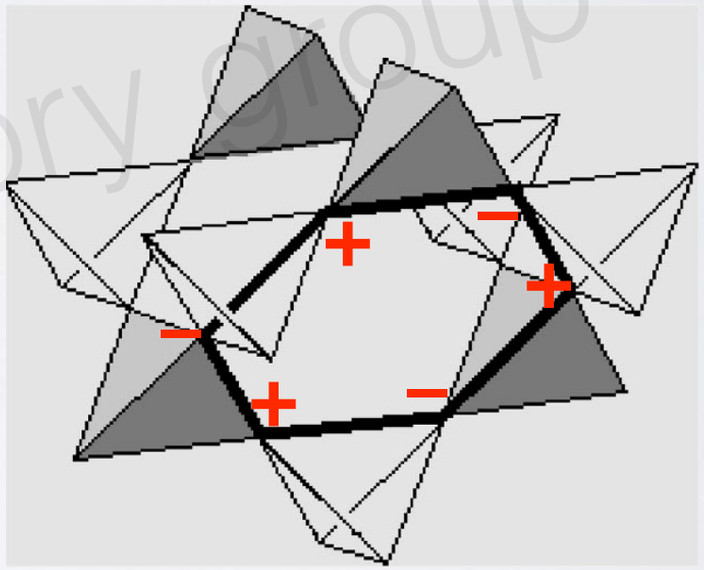
Hermele etc 2004: $J_z \gg J_\pm$

$$H_0 = J_z \sum_{\langle ij \rangle} S_i^z S_j^z$$ gives ice-ruled manifold

$$H_1 = -J_\pm \sum_{\langle ij \rangle} (S_i^+ S_j^- + h.c.)$$ generates quantum tunnelling among ice-rule states



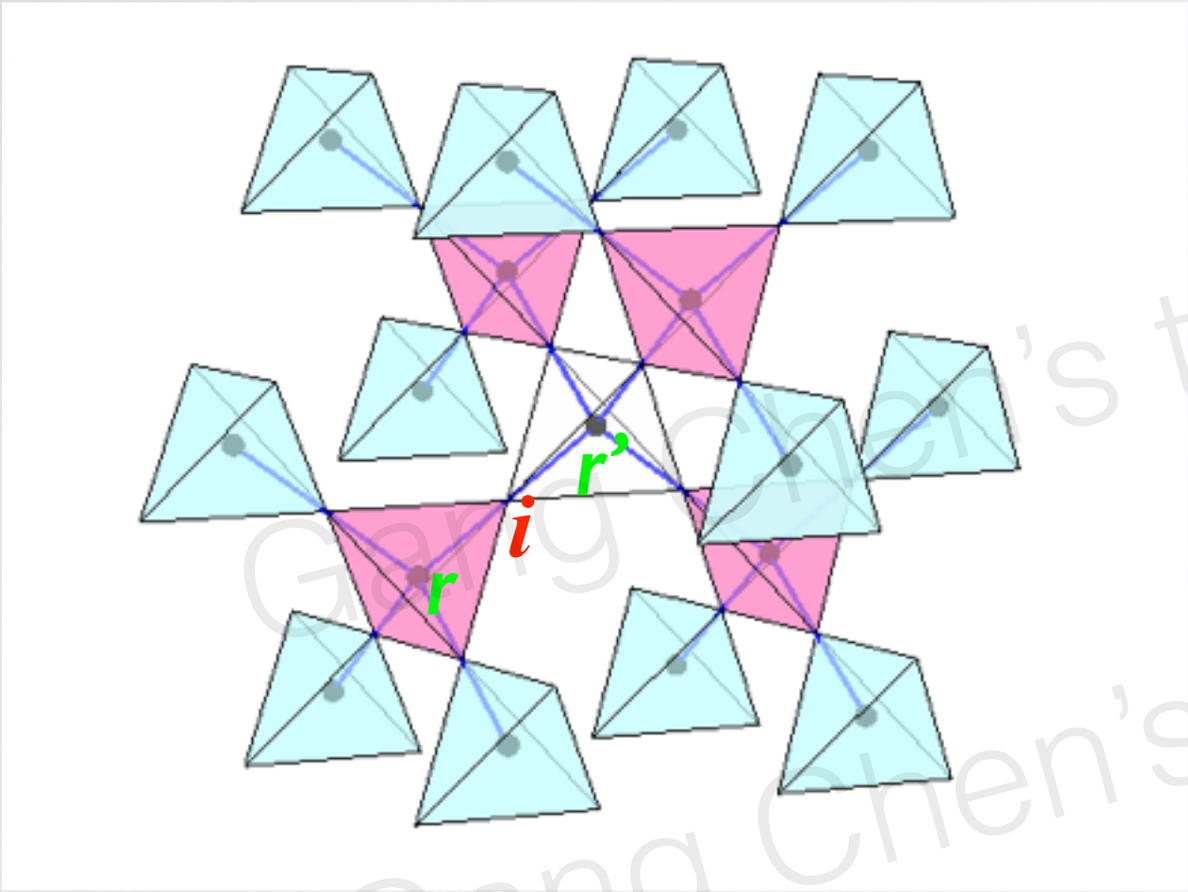
apply H_1
3 times



Effectively given by a 6-site ring exchange

$$H_{\text{ring}} = -\frac{12 J_\pm^3}{J_z^3} \sum_{\text{hexagon}} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.)$$

MAP TO U(1) LATTICE GAUGE THEORY



Define

$$E_{\boldsymbol{rr'}} = S_i^z$$

$$e^{\pm i A_{\boldsymbol{rr'}}} = S_i^{\pm}$$

Compact U(1) lattice gauge theory

$$H_{\text{eff}} = -K \sum_{\text{hexagon}} cos(\nabla \times A)$$

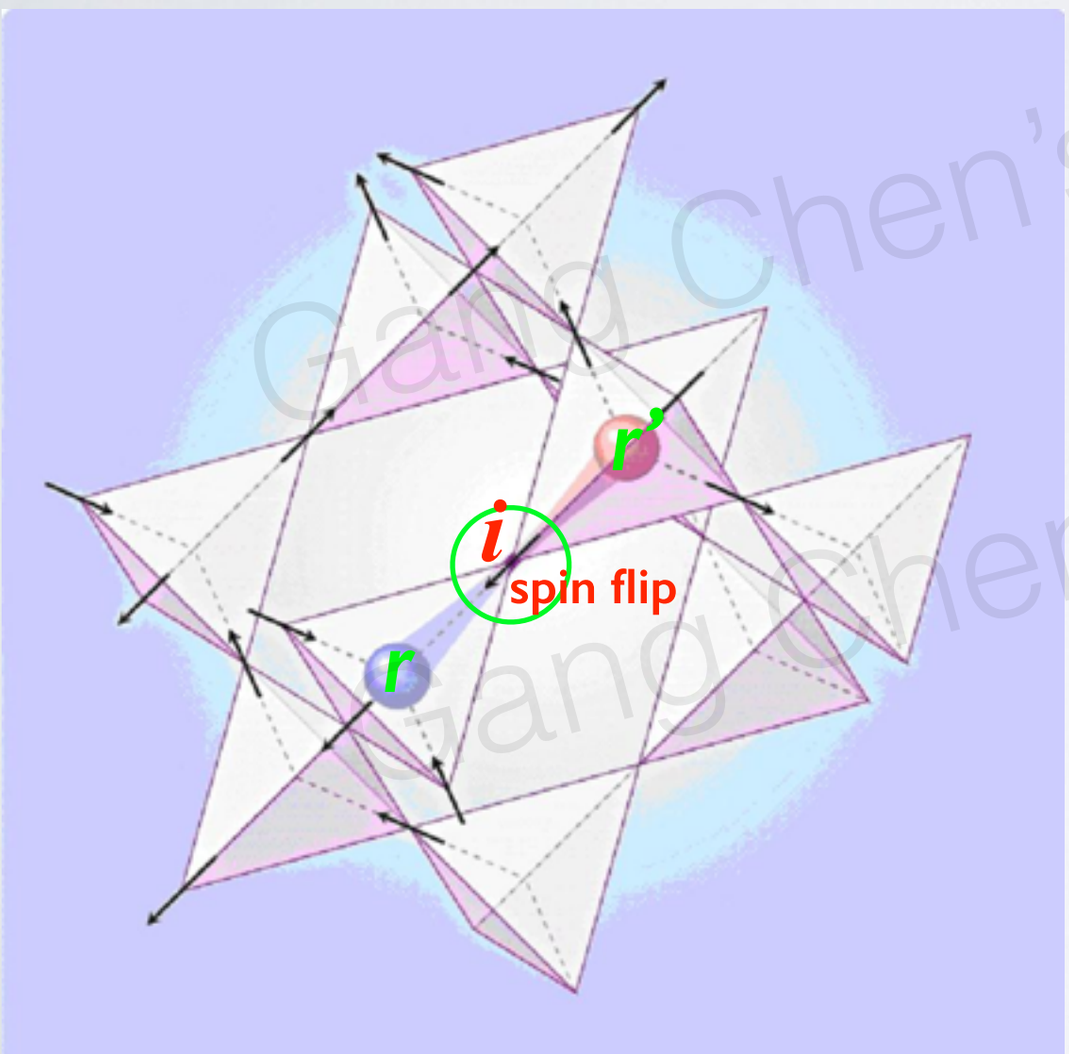
hexagons on the dual diamond lattice

$K > 0$ favours a zero-flux state with $\nabla \times A = 0$

$K < 0$ favours a π-flux state with $\nabla \times A = \pi$

OPEN STRING

Similar as any other slave particle approach, but here is more physically justified.



Spin flip creates spinon-antispinon pair on neighboring diamond sites. (Balents etc 2012)

$$S_i^{\pm} = \Phi_{\boldsymbol{r}}^{\dagger}\Phi_{\boldsymbol{r}'}s_{\boldsymbol{r}\boldsymbol{r}'}^{\pm}$$

where $s_{\boldsymbol{r}\boldsymbol{r}'}^{\pm} = e^{\pm iA_{\boldsymbol{r}\boldsymbol{r}'}}$ is the gauge field.

and gauge charge is defined as

$$Q_{\boldsymbol{r}} = (-1)^{\boldsymbol{r}}\sum_{i\in\boldsymbol{r}} S_i^z$$

invariant under local U(1) gauge transformation

$$\Phi_{\boldsymbol{r}} \to \Phi_{\boldsymbol{r}}e^{i\chi_{\boldsymbol{r}}}$$

$$s_{\boldsymbol{r}\boldsymbol{r}'}^{\pm} \to s_{\boldsymbol{r}\boldsymbol{r}'}^{\pm}e^{i\chi_{\boldsymbol{r}}-i\chi_{\boldsymbol{r}'}}$$

APPLY TO XYZ

Rewrite the XYZ model to manifest the gauge structure

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

$$= \sum_{\langle ij \rangle} J_{zz} \tilde{S}_i^z \tilde{S}_j^z - J_{\pm}(\tilde{S}_i^+ \tilde{S}_j^- + h.c.) + J_{\pm\pm}(\tilde{S}_i^+ \tilde{S}_j^+ + \tilde{S}_i^- \tilde{S}_j^-)$$

with $J_{zz} = \tilde{J}_z$, $J_{\pm} = -\frac{1}{4}(\tilde{J}_x + \tilde{J}_y)$ and $J_{\pm\pm} = \frac{1}{4}(\tilde{J}_x - \tilde{J}_y)$.

show the spinon-gauge coupling

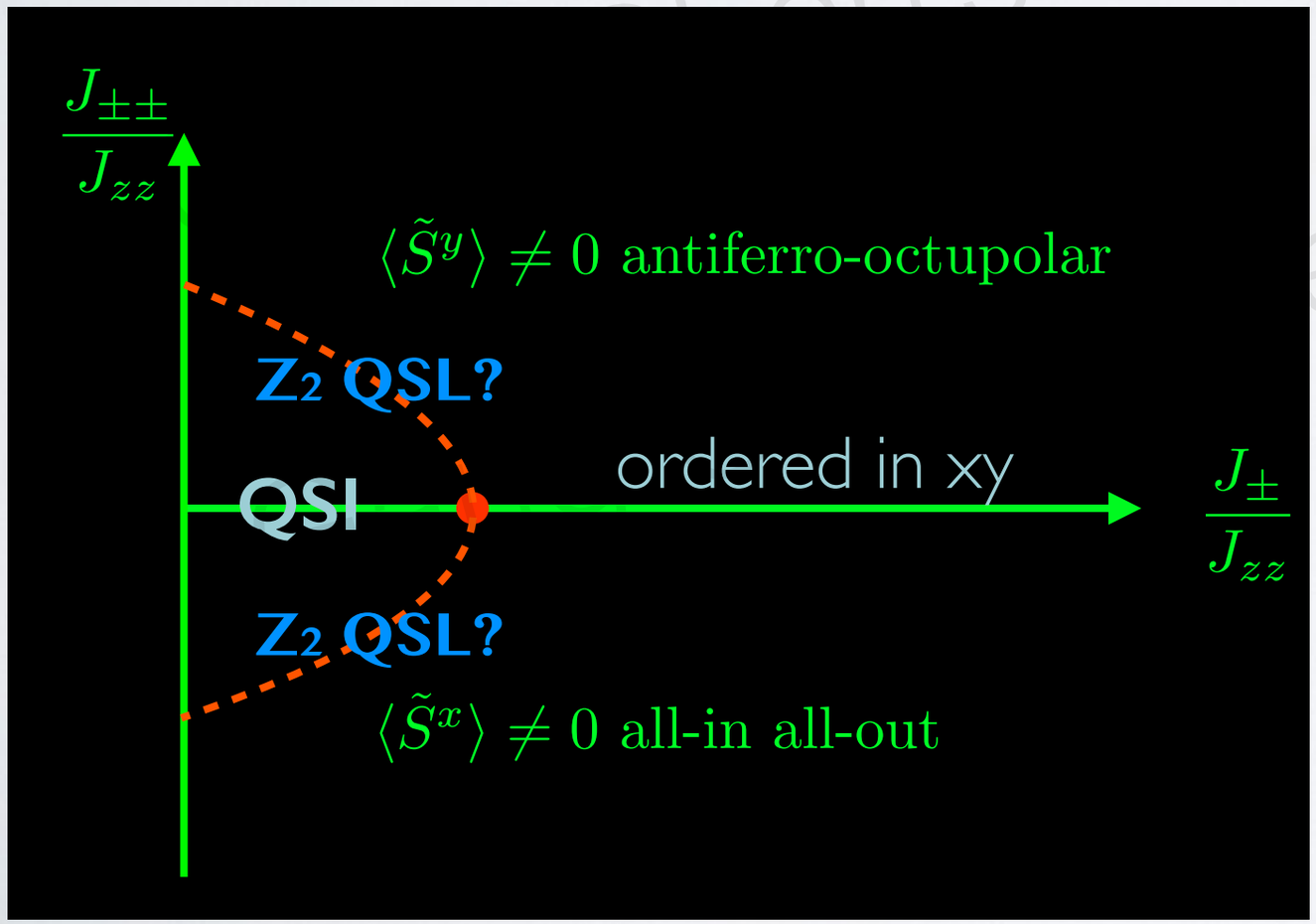
$$H_{\mathrm{XYZ}} = \frac{J_{zz}}{2} \sum_{\mathbf{r}} Q_{\mathbf{r}}^2 - J_{\pm} \sum_{\mathbf{r}} \sum_{i \neq j} \Phi^\dagger_{\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_i} \Phi_{\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_j} s^{-\eta_{\mathbf{r}}}_{\mathbf{r},\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_i} s^{+\eta_{\mathbf{r}}}_{\mathbf{r},\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_j}$$

$$+ \frac{J_{\pm\pm}}{2} \sum_{\mathbf{r}} \sum_{i \neq j} \left(\Phi^\dagger_{\mathbf{r}} \Phi^\dagger_{\mathbf{r}} \Phi_{\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_i} \Phi_{\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_j} s^{\eta_{\mathbf{r}}}_{\mathbf{r},\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_i} s^{\eta_{\mathbf{r}}}_{\mathbf{r},\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_j} + h.c. \right)$$

CONDENSE SPINONS

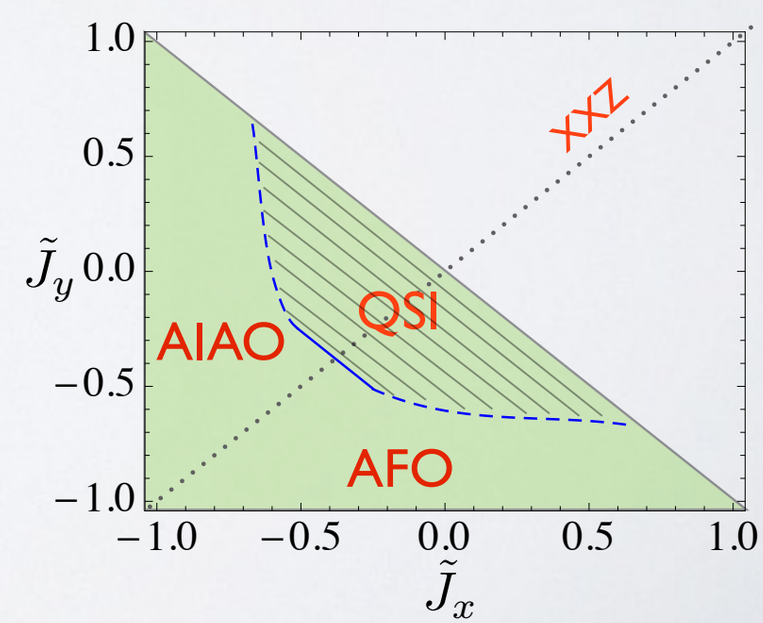
$$H_{\mathrm{XYZ}} = \sum_{\langle ij \rangle} \tilde{J}_z \tilde{S}_i^z \tilde{S}_j^z + \tilde{J}_y \tilde{S}_i^x \tilde{S}_j^x + \tilde{J}_x \tilde{S}_i^y \tilde{S}_j^y$$

$$= \sum_{\langle ij \rangle} J_{zz} \tilde{S}_i^z \tilde{S}_j^z - J_{\pm}(\tilde{S}_i^+ \tilde{S}_j^- + h.c.) + J_{\pm\pm}\underbrace{(\tilde{S}_i^+ \tilde{S}_j^+ + \tilde{S}_i^- \tilde{S}_j^-)}_{= 2(\tilde{S}_i^x \tilde{S}_j^x - \tilde{S}_i^y \tilde{S}_j^y)}$$



Z_2 QSL: $\langle \Phi \rangle = 0$ but $\langle \Phi\Phi \rangle \neq 0$

not found in my gauge MFT

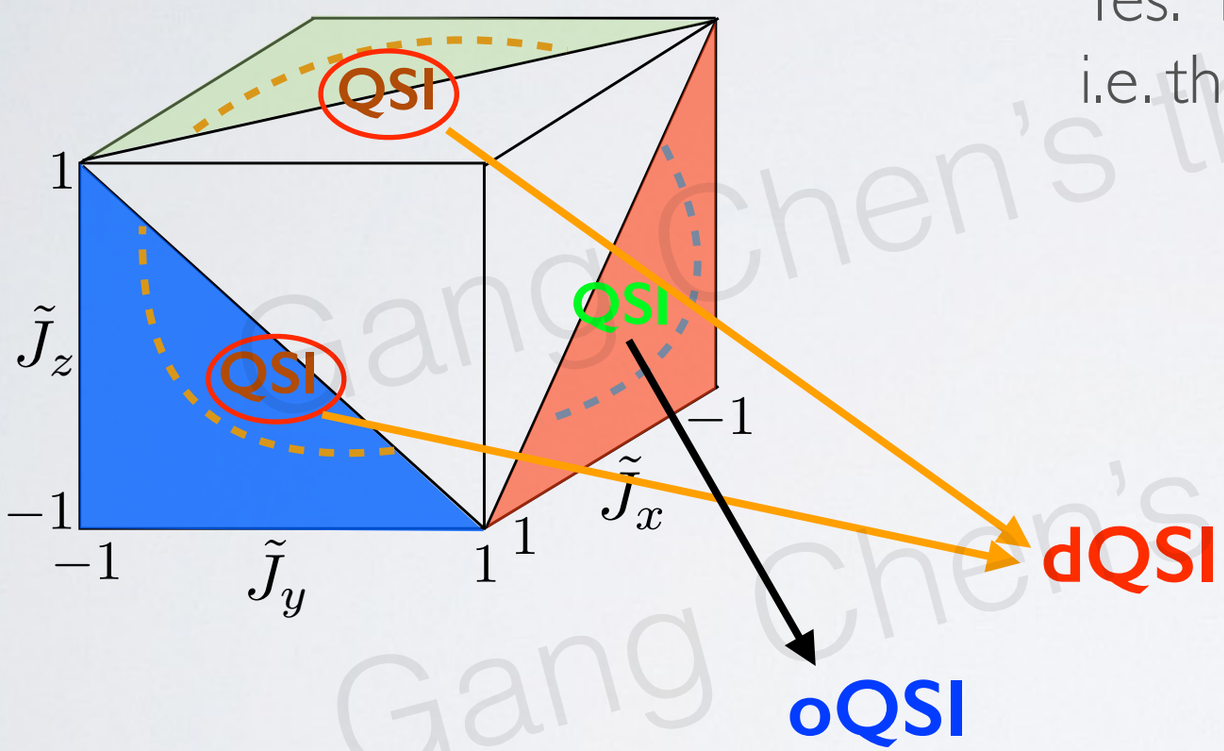


$\tilde{J}_z = 1$

SYMMETRY ENRICHMENT

Are they different?

Yes. They are distinct by symmetry,
i.e. they are symmetry enriched QSI**s**



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

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

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

In fact, Pi-flux state is also also example of symmetry enrichment.
This is similar as the 3 symmetry enriched Z_2 QSLs in Kitaev's toric code model.

dQSI vs oQSI

Transformation of continuum E/B field under O_h point group

	dQSI	oQSI
E-field	T_1^+ (pseudovector)	T_2^+
B-field	T_1^- (vector)	T_2^-

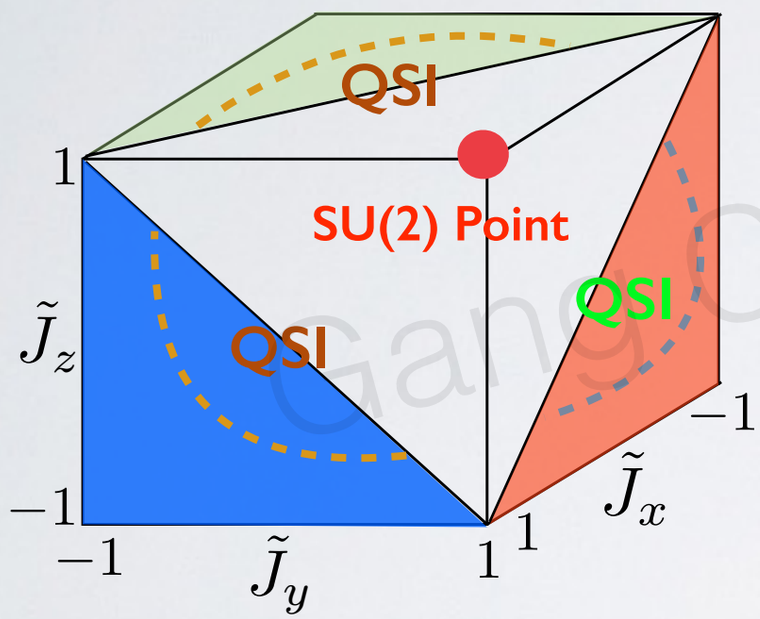
- Both phases have identical thermodynamical properties, e.g. T^3 heat capacity
- Different dipolar static spin correlation:

  dQSI: $< S_z(0)\, S_z(r) > \sim 1/r^4$.

  oQSI: $< S_z(0)\, S_z(r) > \sim 1/r^8$,

  with $Z_2 x Z_2$ symmetry, decay exponentially.

OPEN QUESTION



It is expected that, QSIs are more stable in the (frustrated) white region. But how are the QSIs connected with each other?

What is the ground state of SU(2) Heisenberg model on the pyrochlore lattice?
My conjecture: multicritical point or critical region with emergent non-Abelian gauge structure,
i.e. SU(2) quantum spin liquid

MATERIAL SURVEY

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

Two well-known systems:

- Pyrochlores $A_2B_2O_7$,

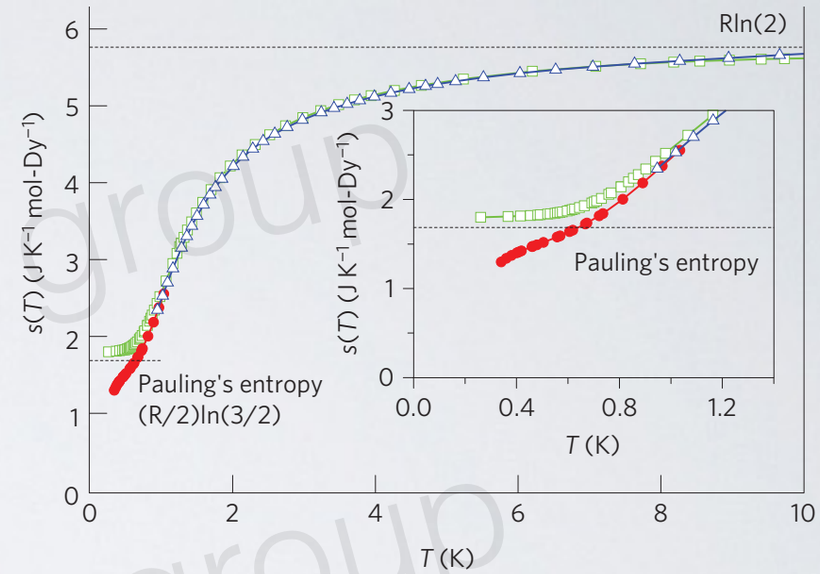
  A = Nd, Er, Dy, . . . ?

  e.g. ,

  $Nd_2Ir_2O_7$, $Nd_2Sn_2O_7$, $Nd_2Zr_2O_7$, etc

  $Dy_2Ti_2O_7$,

  $Cd_2Os_2O_7$, etc

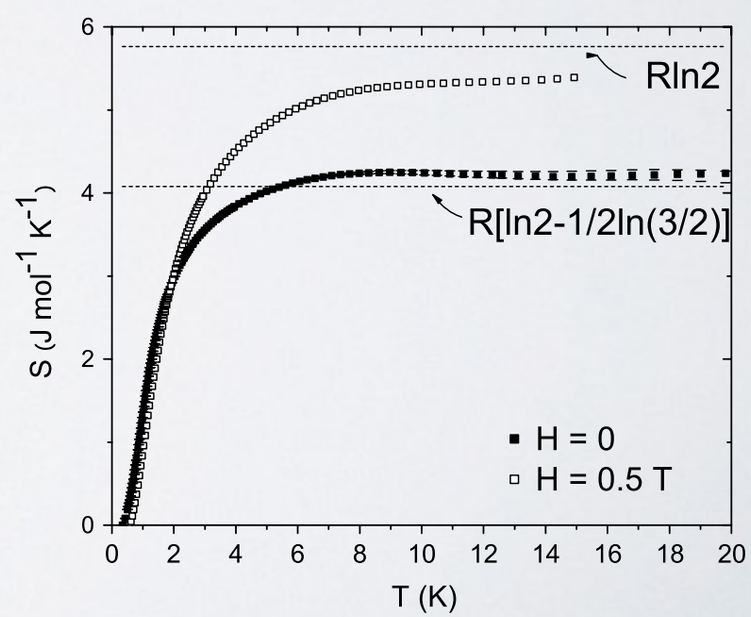


Prof Gaulin's group, Nat Phys, 2013

- Spinels AB_2X_4, B=lanthanide?

  e.g. $CdEr_2Se_4$

  $CdYb_2S_4$



J Lago, etc, PRL 2010

SUMMARY

- We propose a realistic XYZ model based on a singlet Kramers doublet on the pyrochlore lattice.

- This realistic model supports two distinct symmetry enriched quantum spin ice phases.

- This model can be well understood by quantum Monte Carlo simulation.