

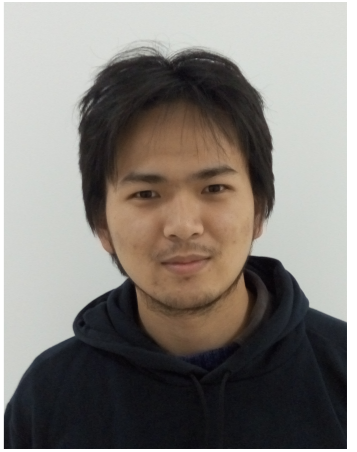
Hidden multipolar orders from **dipole-octupole doublets** on a triangular lattice

Gang Chen

Fudan University, May 14, 2016



Outline

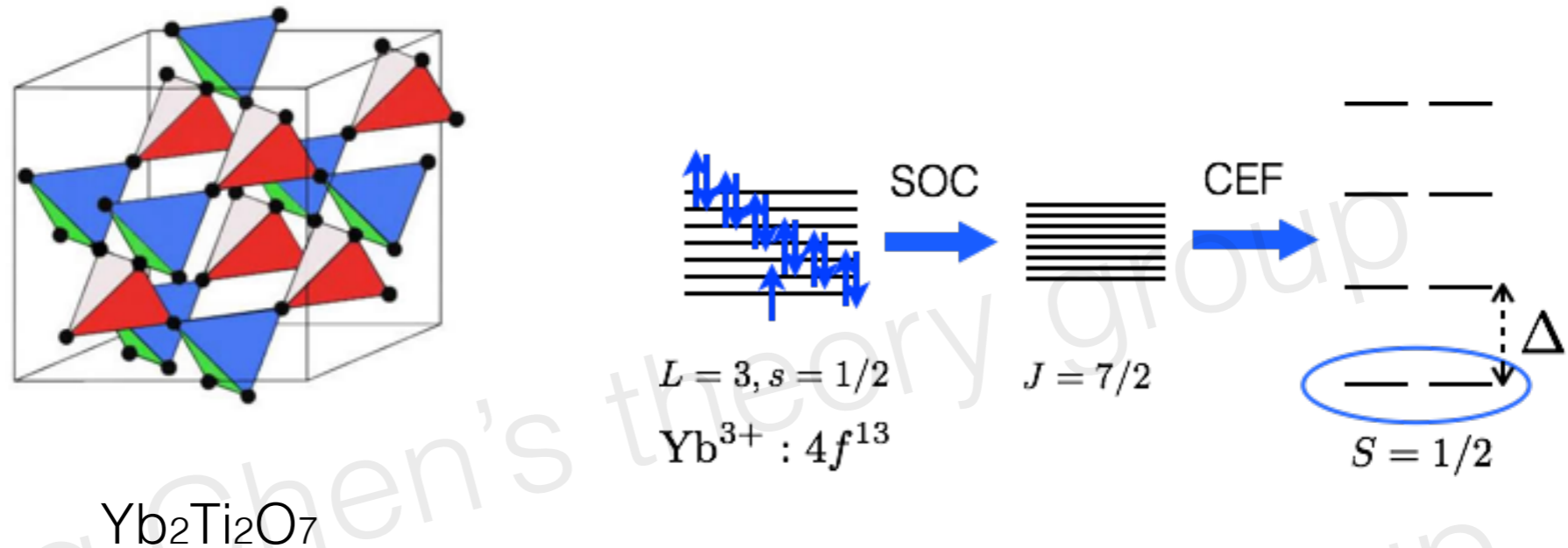


Yao-Dong Li

- Dipole-octupole doublet and pyrochlore spin ice
- Triangular lattice XYZ model
- Hidden multipolar orders
- Summary

Yao-Dong Li, **GC***, in preparation

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



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

Lanthanum 57 La 138.91	Cerium 58 Ce 140.12	Praseodymium 59 Pr 140.91	Neodymium 60 Nd 144.24	Promethium 61 Pm [145]	Samarium 62 Sm 150.36	Europium 63 Eu 151.96	Gadolinium 64 Gd 157.25	Terbium 65 Tb 158.93	Dysprosium 66 Dy 162.50	Holmium 67 Ho 164.93	Erbium 68 Er 167.26	Thulium 69 Tm 168.93	Ytterbium 70 Yb 173.04
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Non-Kramers' doublet / singlet: R³⁺ with **even** number of electrons

Lanthanum 57 La 138.91	Cerium 58 Ce 140.12	Praseodymium 59 Pr 140.91	Neodymium 60 Nd 144.24	Promethium 61 Pm [145]	Samarium 62 Sm 150.36	Europium 63 Eu 151.96	Gadolinium 64 Gd 157.25	Terbium 65 Tb 158.93	Dysprosium 66 Dy 162.50	Holmium 67 Ho 164.93	Erbium 68 Er 167.26	Thulium 69 Tm 168.93	Ytterbium 70 Yb 173.04
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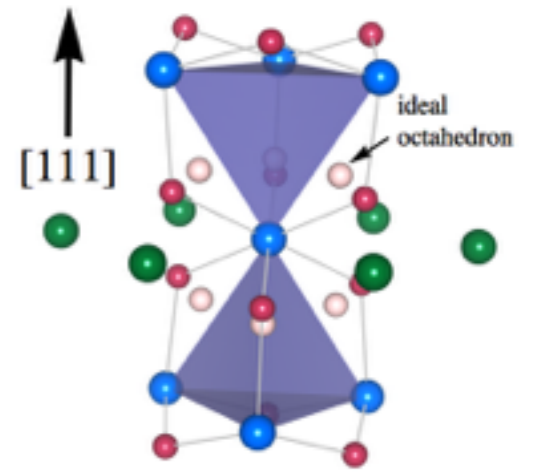
Dipole-octupole doublet

The classification of local moments is a bit crude !

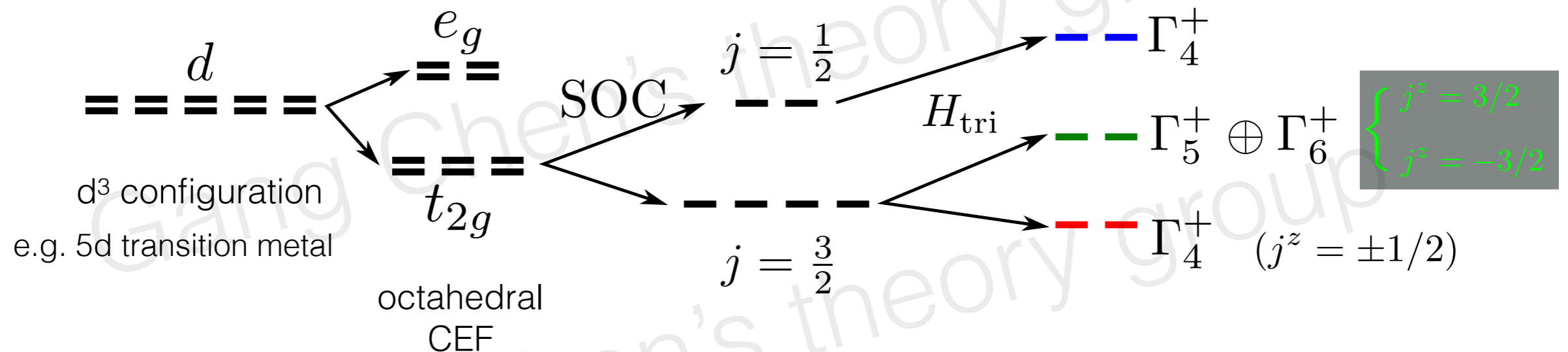
One should carefully examine the wavefunction of the local doublet.

Yi-Ping Huang, [GC*](#), Michael Hermele
arXiv 1311.1231, **Phys. Rev. Lett.** 112, 167203 (2014)

Local physics: start with t_{2g} electrons



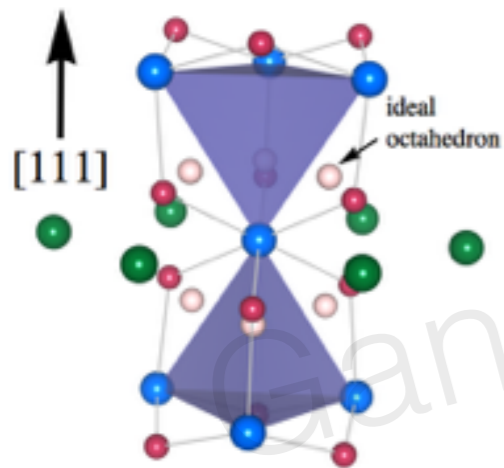
- Local moments on pyrochlore lattice: effective spin-1/2



d electrons under D_{3d}
point group crystal field

- Why is this Kramers doublet so special ?

ONE-dimensional representations of the point group !



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

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

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

More generally, ...

- Also applies to 4f electron moments on pyrochlore

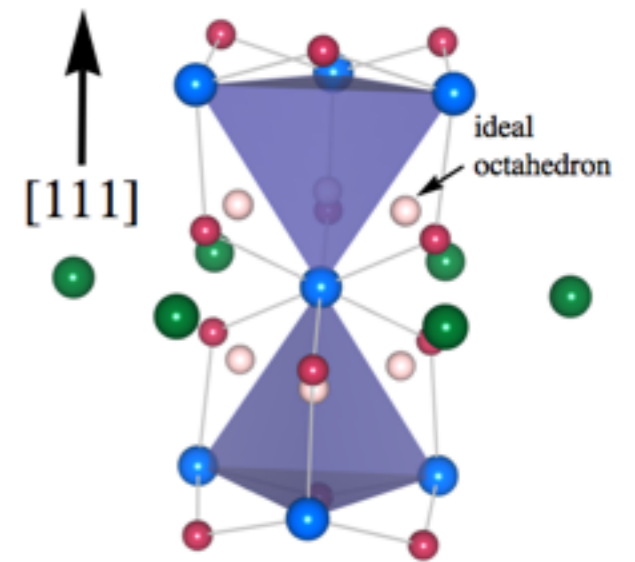
$$J^z = \frac{3}{2}, \frac{9}{2}, \frac{15}{2}, \dots$$

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

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

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

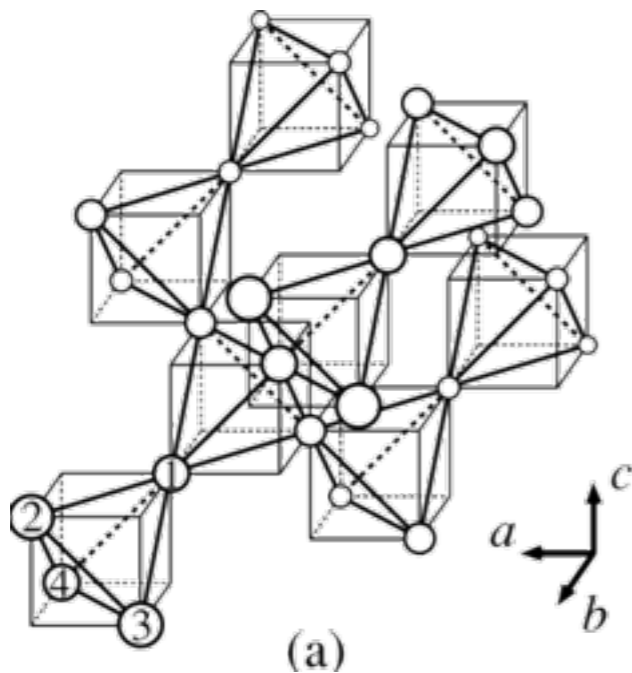
Many Nd pyrochlores have been experimentally confirmed to be DO doublets.



Symmetry properties

- Effective spin-1/2 under lattice symmetry Tetrahedral Group

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



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

$$C_3 : S^\mu \rightarrow S^\mu$$

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

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

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

A small transformation into XYZ model

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



Rotation around the y axis
in the effective spin space

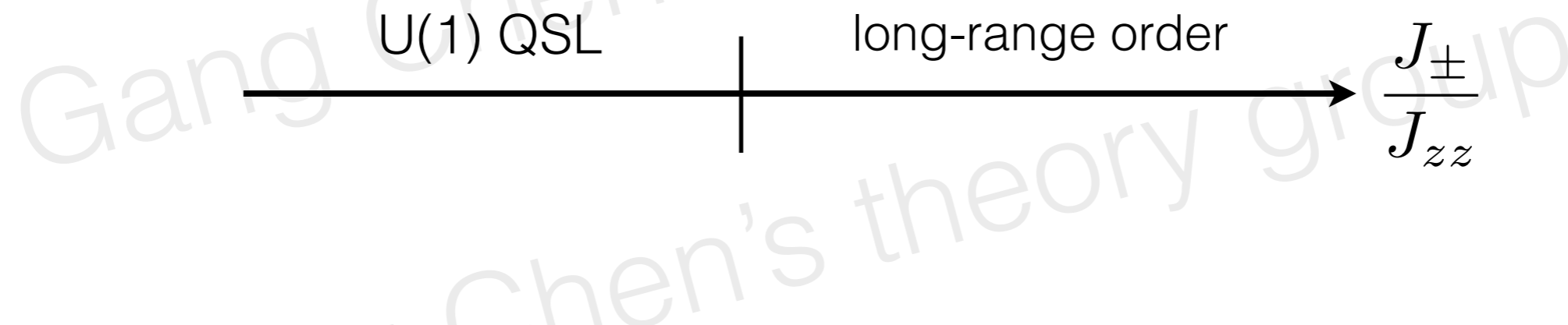
$$H_{\text{XYZ}} = \sum_{\langle ij \rangle} \tilde{J}_z \tilde{S}_i^z \tilde{S}_j^z + \tilde{J}_x \tilde{S}_i^x \tilde{S}_j^x + \tilde{J}_y \tilde{S}_i^y \tilde{S}_j^y$$

XYZ model

XXZ model can lead to U(1) QSL

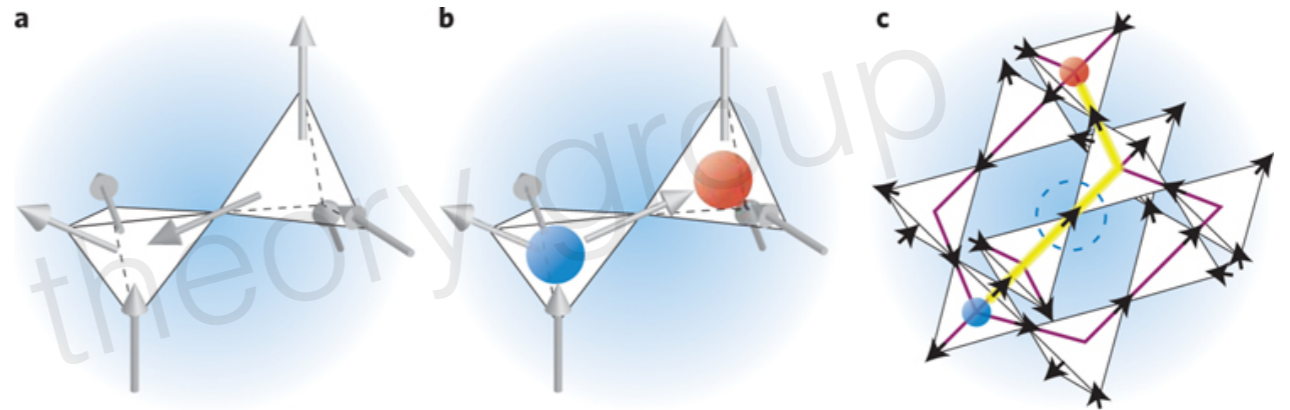
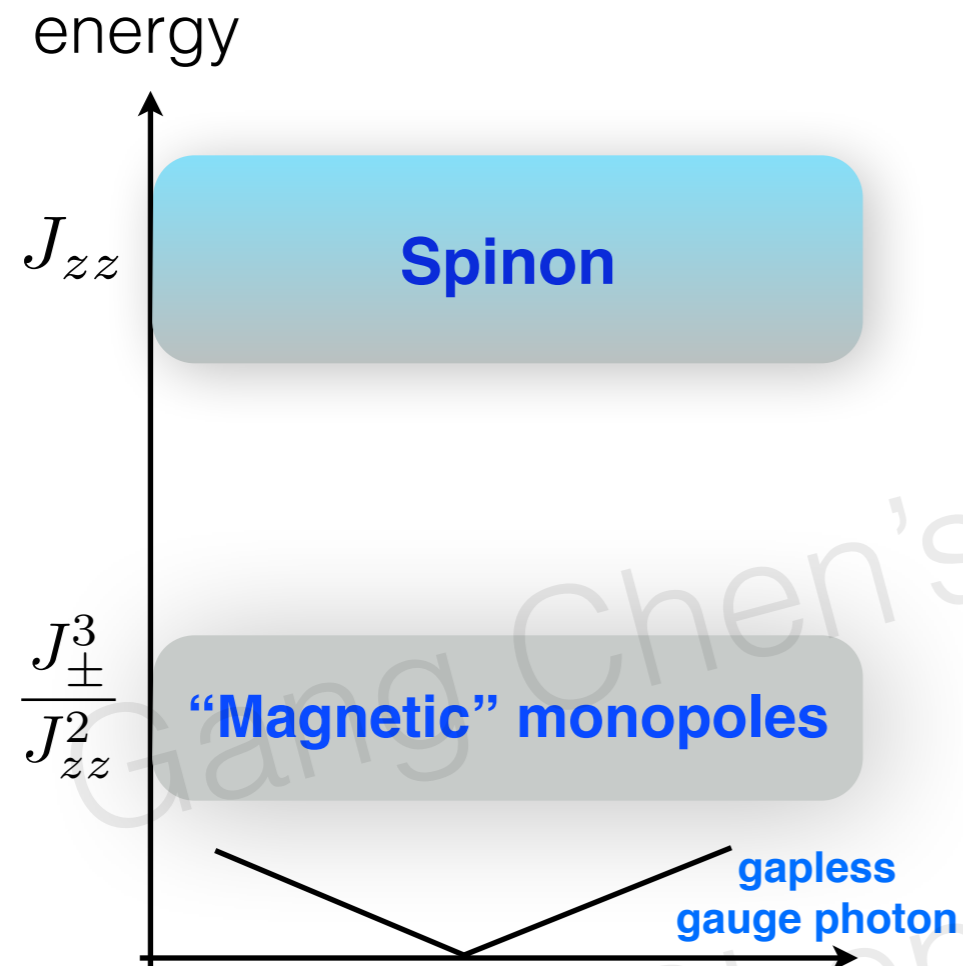
$$H = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z - J_{\pm} \sum_{\langle i,j \rangle} (S_i^+ S_j^- + S_i^- S_j^+) + \dots$$

Hermele, Fisher, Balents, Moessner, Isakov, YB Kim....



- 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



Figs from Moessner&Schiffer,2009

Spinon deconfinement

Emergent electric field

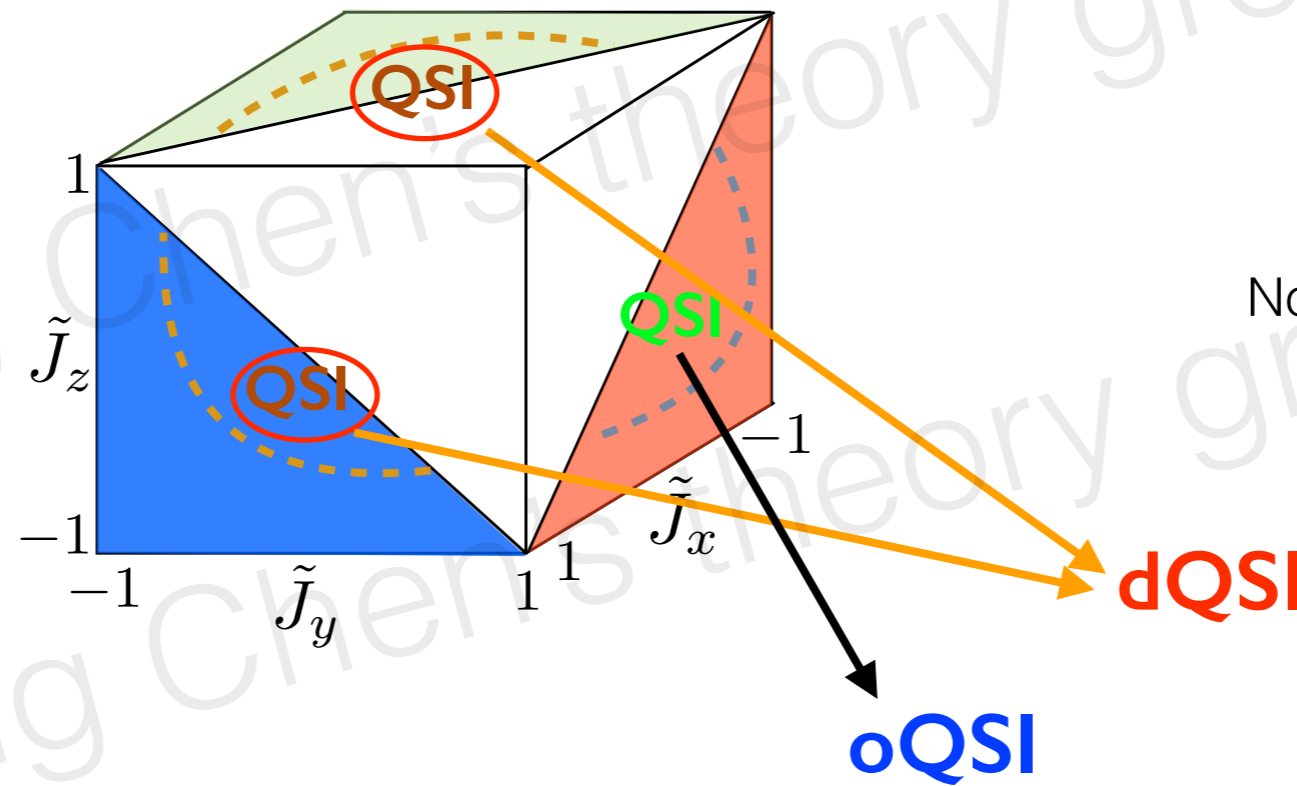
$$S^z \sim E$$

Emergent vector potential

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

Symmetry enriched quantum spin ice on pyrochlore

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



Yi-Ping Huang, [GC*](#), Michael Hermele
arXiv 1311.1231, **Phys. Rev. Lett.** 112, 167203 (2014)

dQSI vs oQSI

Transformation of continuum E/B field under Oh 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: $\langle S_z(0) S_z(r) \rangle \sim 1/r^4$.
 oQSI: $\langle S_z(0) S_z(r) \rangle \sim 1/r^8$,
 with nearest-neighbor $Z_2 \times Z_2$ symmetry, decay exponentially.

What makes a DO doublet is the wavefunction, not the J 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

4f¹ 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 $50 \pm$

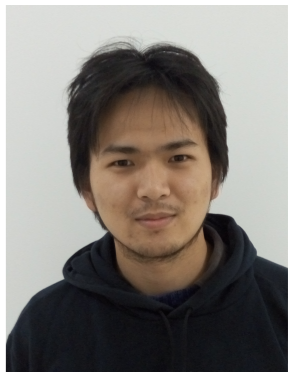
Ce³⁺ ($4f^1, {}^2F_{5/2}$).

$$J = \frac{5}{2}$$

How to differentiate dQSI and oQSI?

Yao-Dong Li, **GC***, in preparation

- Triangular lattice XYZ model and hidden order



Yao-Dong Li

Triangular lattice antiferromagnets

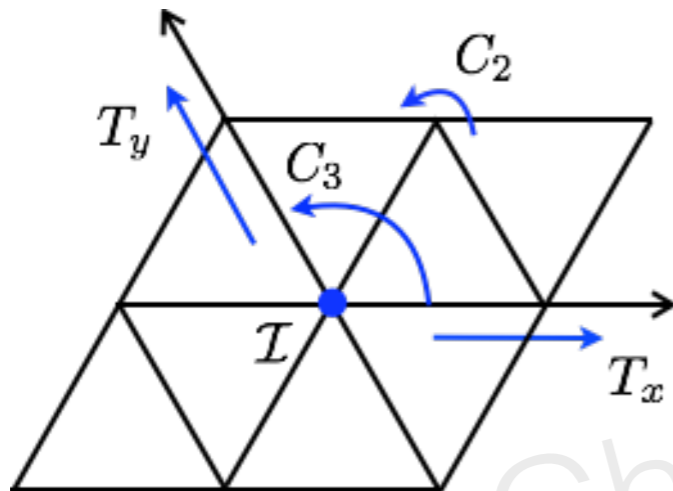
Compound	Magnetic ion	Space group	Local moment	Θ_{CW} (K)	Magnetic transition	Frustration para. f	Ref
YbMgGaO ₄	Yb ³⁺ ($4f^{13}$)	R $\bar{3}m$	Kramers doublet	−4	PM down to 60mK	$f > 66$	4
CeCd ₃ P ₃	Ce ³⁺ ($4f^1$)	P6 ₃ / mmc	Kramers doublet	−60	PM down to 0.48K	$f > 200$	5
CeZn ₃ P ₃	Ce ³⁺ ($4f^1$)	P6 ₃ / mmc	Kramers doublet	−6.6	AFM order at 0.8K	$f = 8.2$	7
CeZn ₃ As ₃	Ce ³⁺ ($4f^1$)	P6 ₃ / mmc	Kramers doublet	−62	unknown	unknown	8
PrZn ₃ As ₃	Pr ³⁺ ($4f^2$)	P6 ₃ / mmc	Non-Kramers doublet	−18	unknown	unknown	8
NdZn ₃ As ₃	Nd ³⁺ ($4f^3$)	P6 ₃ / mmc	Kramers doublet	−11	unknown	unknown	8
Nd ₂ O ₂ CO ₃	Nd ³⁺ ($4f^3$)	P6 ₃ / mmc	Kramers doublet	−21.7	AFM order at 1.25K	$f = 17.4$	9
Sm ₂ O ₂ CO ₃	Sm ³⁺ ($4f^5$)	P6 ₃ / mmc	Kramers doublet	−18	AFM order at 0.61K	$f = 31$	9
Dy ₂ O ₂ CO ₃	Dy ³⁺ ($4f^9$)	P6 ₃ / mmc	Kramers doublet	−10.6	AFM order at 1.21K	$f = 8.8$	9

- * All of them have rare-earth elements, and involve strong spin-orbit coupling.
- * It is likely that some of them realize dipole-octupole doublet on the triangular lattice.

Yuesheng Li, [GC*](#),Qingming Zhang, PhysRevLett 2015

Yao-Dong Li, Xiaoqun Wang, [GC*](#), ArXiv 1512.02151

Triangular lattice antiferromagnets



$$\left\{ \begin{array}{l} C_3 : \tau_{\mathbf{r}}^x \rightarrow \tau_{C_3(\mathbf{r})}^x, \quad \tau_{\mathbf{r}}^y \rightarrow \tau_{C_3(\mathbf{r})}^y, \quad \tau_{\mathbf{r}}^z \rightarrow \tau_{C_3(\mathbf{r})}^z, \\ C_2 : \tau_{\mathbf{r}}^x \rightarrow \tau_{C_2(\mathbf{r})}^x, \quad \tau_{\mathbf{r}}^y \rightarrow -\tau_{C_2(\mathbf{r})}^y, \quad \tau_{\mathbf{r}}^z \rightarrow -\tau_{C_2(\mathbf{r})}^z, \\ I : \tau_{\mathbf{r}}^x \rightarrow \tau_{I(\mathbf{r})}^x, \quad \tau_{\mathbf{r}}^y \rightarrow \tau_{I(\mathbf{r})}^y, \quad \tau_{\mathbf{r}}^z \rightarrow \tau_{I(\mathbf{r})}^z, \\ T_x : \tau_{\mathbf{r}}^x \rightarrow \tau_{T_x(\mathbf{r})}^x, \quad \tau_{\mathbf{r}}^y \rightarrow \tau_{T_x(\mathbf{r})}^y, \quad \tau_{\mathbf{r}}^z \rightarrow \tau_{T_x(\mathbf{r})}^z, \\ T_y : \tau_{\mathbf{r}}^x \rightarrow \tau_{T_y(\mathbf{r})}^x, \quad \tau_{\mathbf{r}}^y \rightarrow \tau_{T_y(\mathbf{r})}^y, \quad \tau_{\mathbf{r}}^z \rightarrow \tau_{T_y(\mathbf{r})}^z. \end{array} \right.$$

$$H_0 = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} [J_x \tau_{\mathbf{r}}^x \tau_{\mathbf{r}'}^x + J_y \tau_{\mathbf{r}}^y \tau_{\mathbf{r}'}^y + J_z \tau_{\mathbf{r}}^z \tau_{\mathbf{r}'}^z + J_{yz} (\tau_{\mathbf{r}}^y \tau_{\mathbf{r}'}^z + \tau_{\mathbf{r}}^z \tau_{\mathbf{r}'}^y)]$$

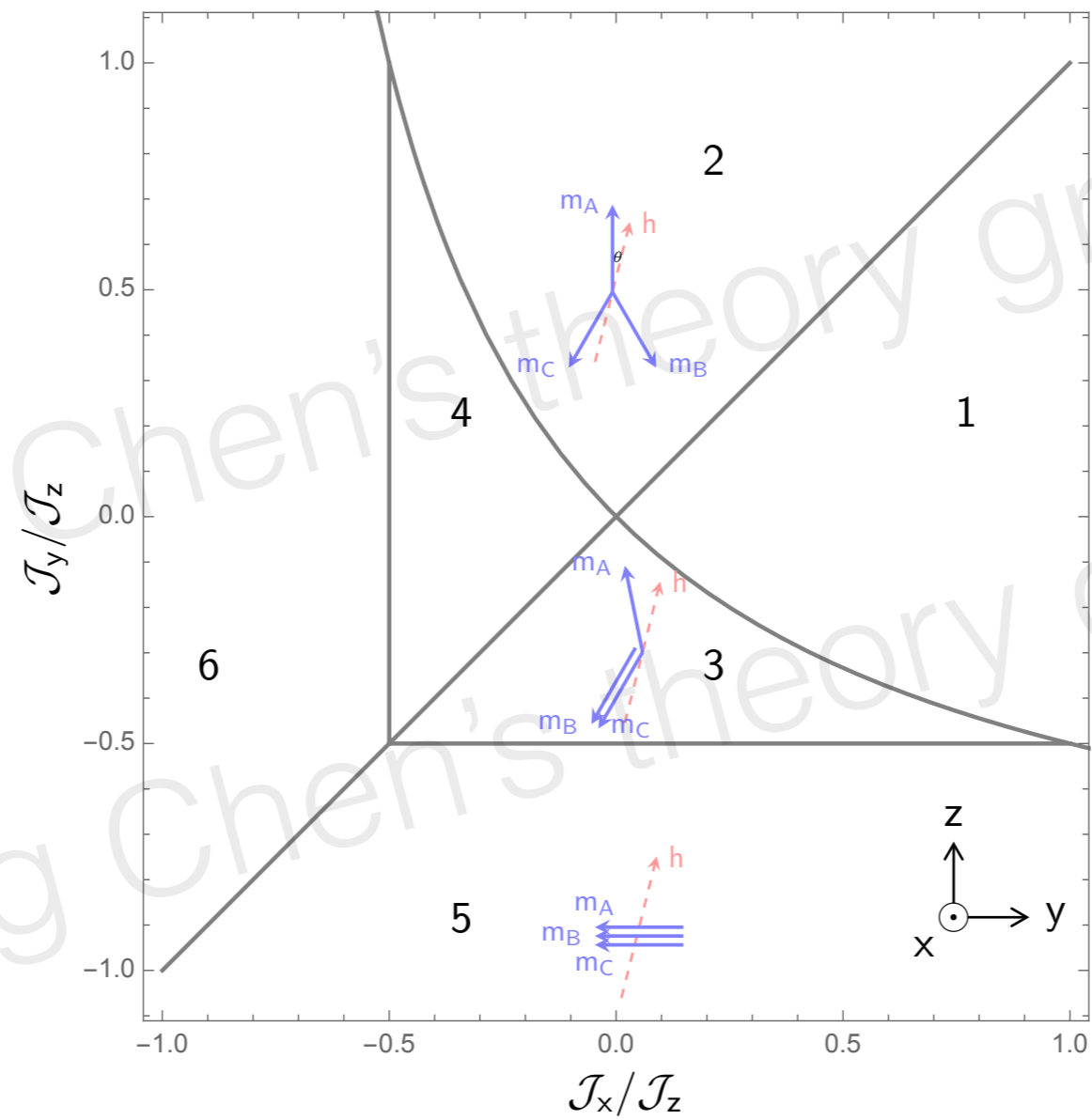


$$H = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} [\mathcal{J}_x T_{\mathbf{r}}^x T_{\mathbf{r}'}^x + \mathcal{J}_y T_{\mathbf{r}}^y T_{\mathbf{r}'}^y + \mathcal{J}_z T_{\mathbf{r}}^z T_{\mathbf{r}'}^z] - h \sum_{\mathbf{r}} [\cos \theta T_{\mathbf{r}}^z + \sin \theta T_{\mathbf{r}}^y],$$

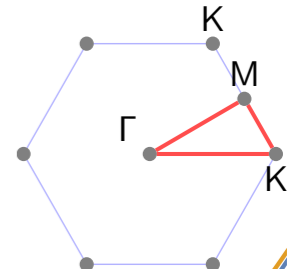
spatially uniform,
rather unusual for strong SOC system

τ^x is a pure octupolar moment !

Rich phase diagram



“Hidden” ferro-octupolar order

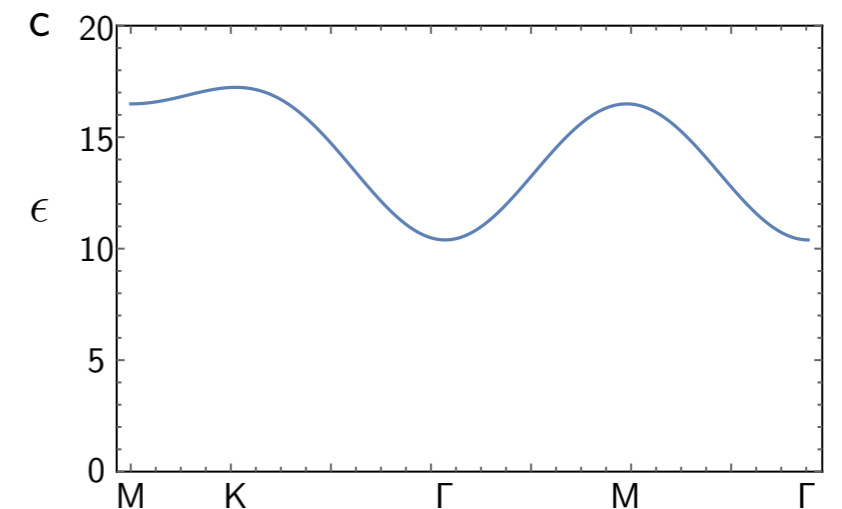
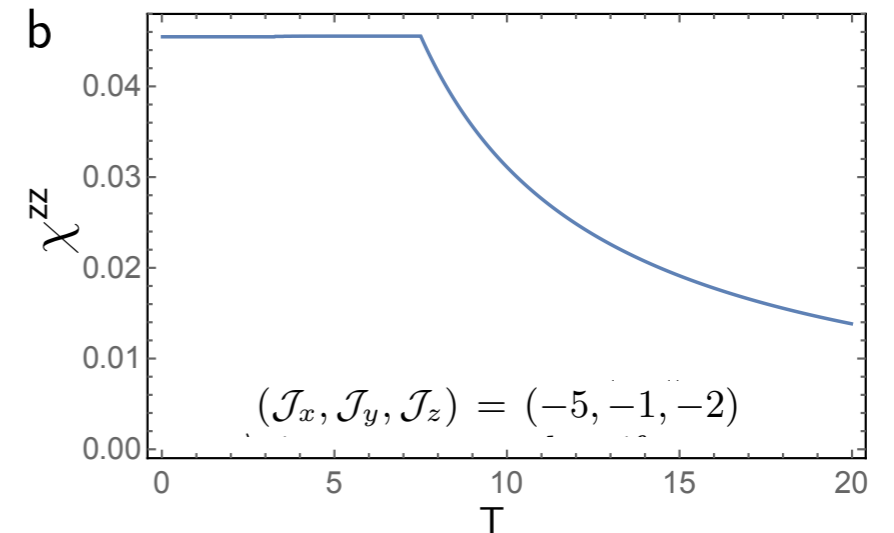


$$H = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} [\mathcal{J}_x T_{\mathbf{r}}^x T_{\mathbf{r}'}^x + \mathcal{J}_y T_{\mathbf{r}}^y T_{\mathbf{r}'}^y + \mathcal{J}_z T_{\mathbf{r}}^z T_{\mathbf{r}'}^z] - h \sum_{\mathbf{r}} [\cos \theta T_{\mathbf{r}}^z + \sin \theta T_{\mathbf{r}}^y],$$



$\mathcal{J}_x < 0$ and dominates
Mean-Field Theory

$$H = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} [\mathcal{J}_x T_{\mathbf{r}}^x \langle T_{\mathbf{r}'}^x \rangle + \mathcal{J}_y T_{\mathbf{r}}^y \langle T_{\mathbf{r}'}^y \rangle + \mathcal{J}_z T_{\mathbf{r}}^z \langle T_{\mathbf{r}'}^z \rangle] - h \sum_{\mathbf{r}} [\cos \theta T_{\mathbf{r}}^z + \sin \theta T_{\mathbf{r}}^y]$$



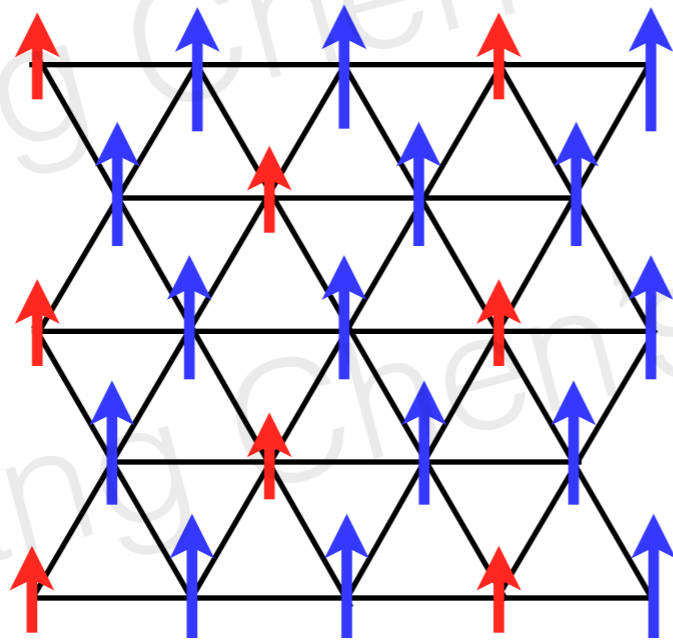
Well-defined spin wave should be observed by inelastic neutron.

“Hidden” antiferro-octupolar order

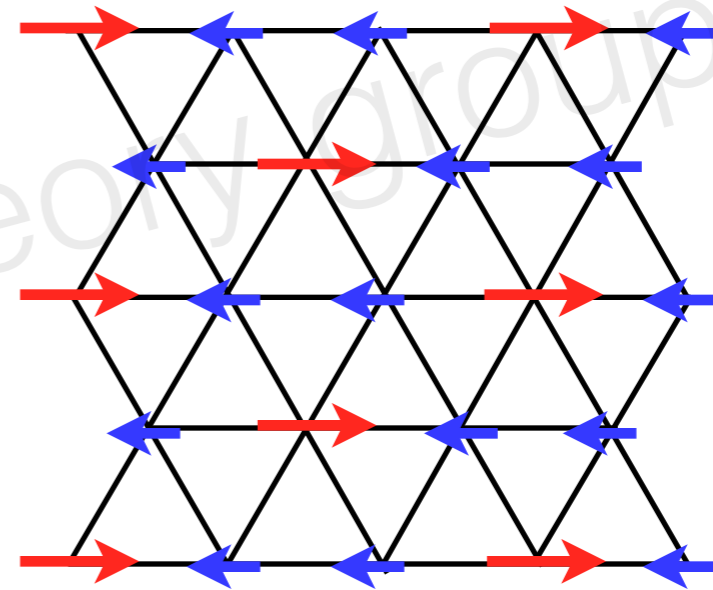
$$H = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} [\mathcal{J}_x T_{\mathbf{r}}^x T_{\mathbf{r}'}^x + \mathcal{J}_y T_{\mathbf{r}}^y T_{\mathbf{r}'}^y + \mathcal{J}_z T_{\mathbf{r}}^z T_{\mathbf{r}'}^z]$$

$\mathcal{J}_x > 0$ and dominates, $\mathcal{J}_x = \mathcal{J}_y$, “XXZ” model with a global U(1) symmetry

R Melko, etc PRL 2005: XXZ model on triangular lattice has supersolid ground state (break translation).



dipolar order



octupolar order

muSR, NMR, neutron sensitive

Summary

- We introduced a new Kramers doublet: dipole-octupole doublet.
- Motivated by rare earth triangular materials, we propose XYZ model and point out the hidden multipolar orders.
- Various experimental consequences are suggested and proposed.