Detecting Hidden Orders in frustrated magnets

Gang Chen
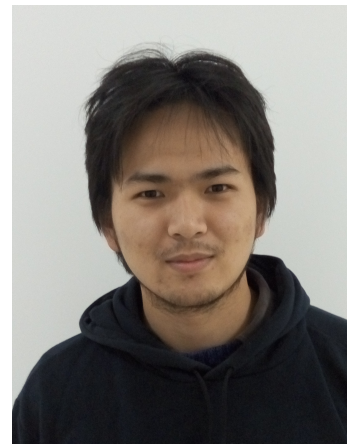
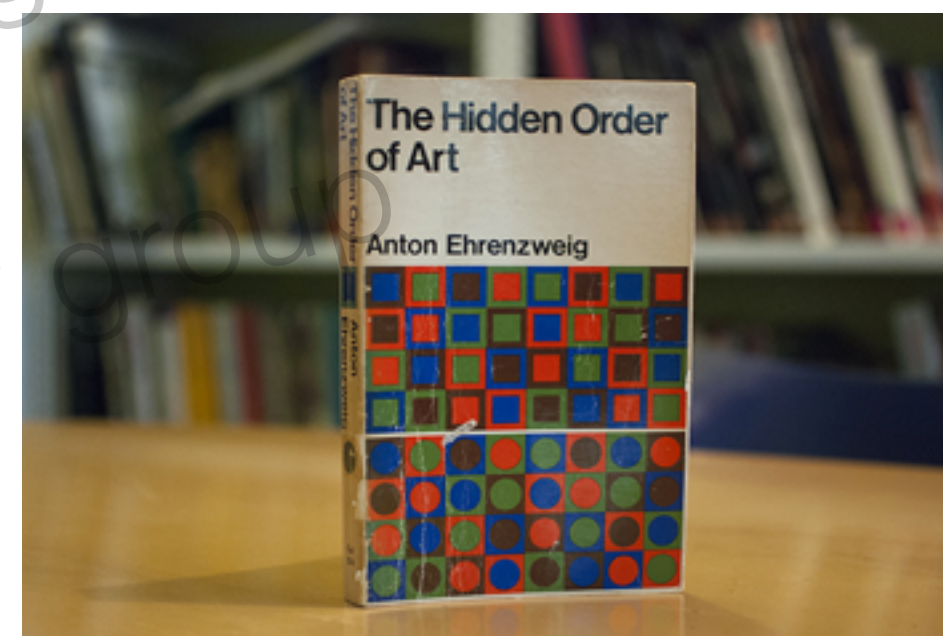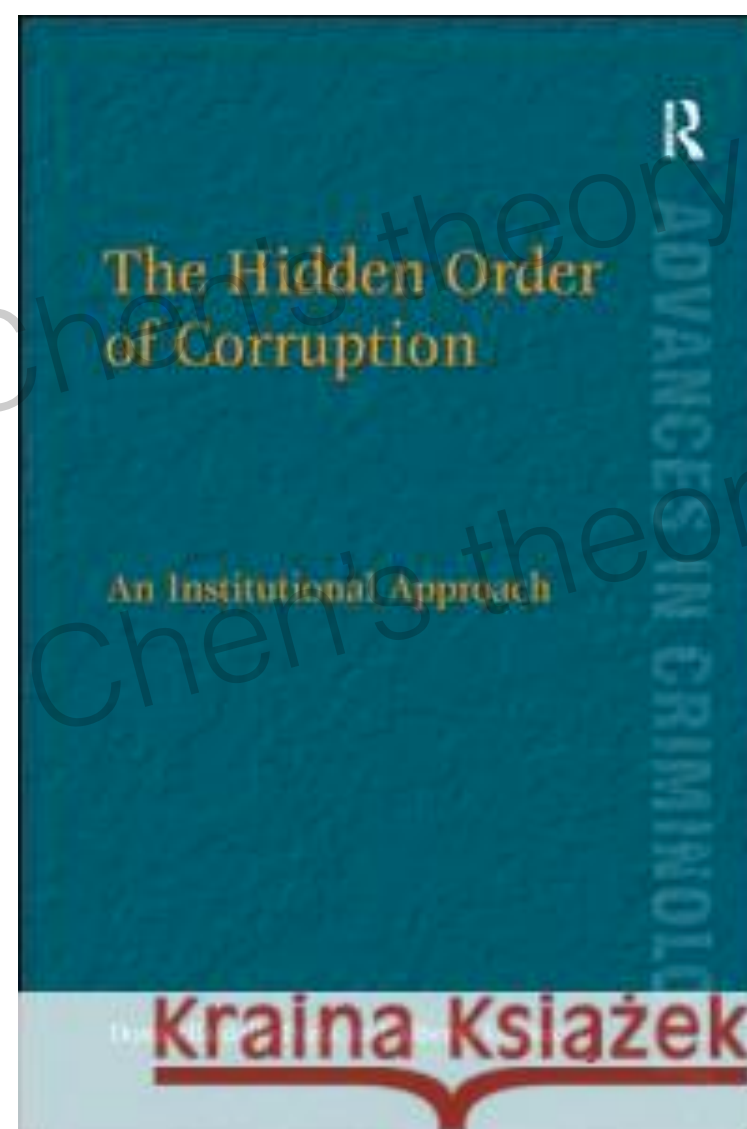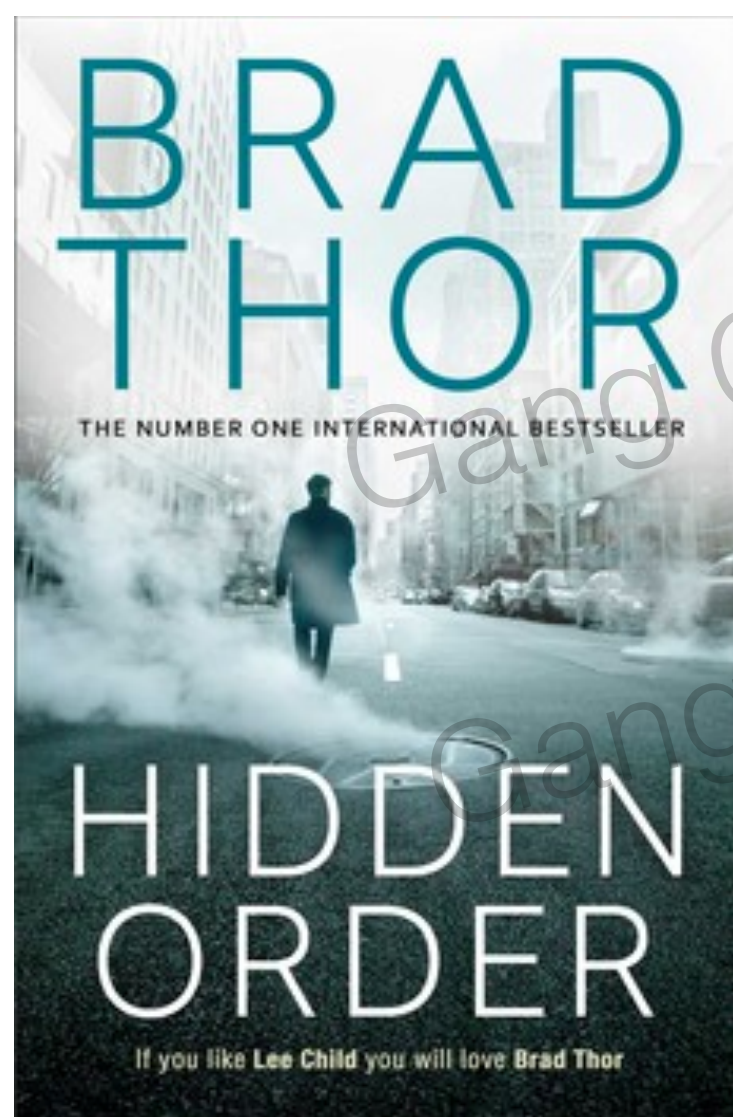Fudan University, Shanghai

Outline

Changle Liu
Fudan

Yaodong Li
Fudan -> UCSB

1. Hidden order in condensed matter physics.

2. Hidden orders with intertwined multipolar structure in rare-earth triangular lattice magnets.

3. Discovery of intertwined multipolar order in TmMgGaO4
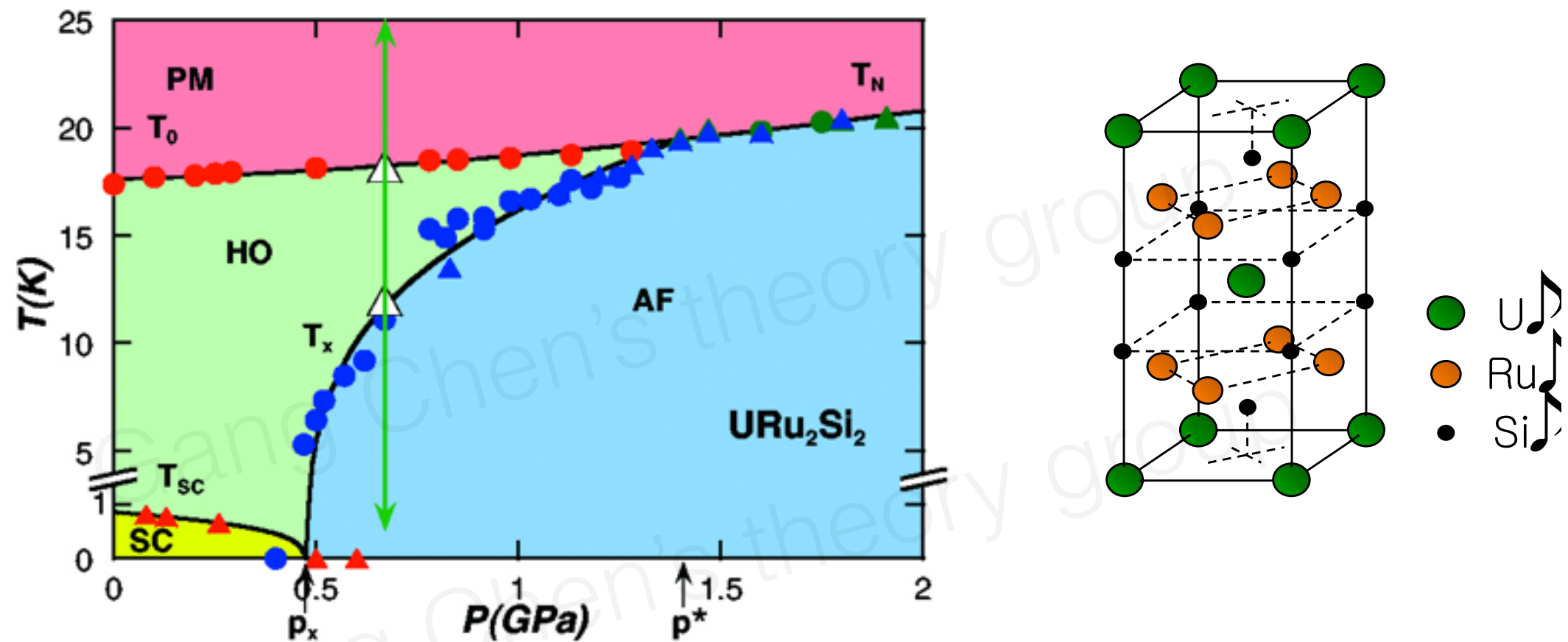   ("**dry product**" with Yao Shen, Jun Zhao)

Changle Liu, Yaodong Li, Gang Chen*, Phys. Rev. B, 98, 045119 (2018)
Yaodong Li, Xiaoqun Wang, Gang Chen*, Phys. Rev. B, 94, 201114(R) (2016)
Yao Shen, Changle Liu, ..., Gang Chen*, Jun Zhao*, arXiv 1810.05054 (2018)

There is no field theory, no exotic phenomenon, no fractionalization, no topological order, etc in this talk.

HIDDEN ORDER

BRAD THOR

THE NUMBER ONE INTERNATIONAL BESTSELLER

HIDDEN ORDER

If you like Lee Child you will love Brad Thor

The Hidden Order of Corruption

An Institutional Approach

ADVANCES IN CRIMINOLOGY

Kraina Książek

The Hidden Order of Art

Anton Ehrenzweig

Hidden order in condensed matter

- Hidden order: "dark matter" in CMT
- URu_2Si_2
  - Second order transition at ~17K, $\Delta \mathcal{S} \sim 0.42$ Rln2
  - Order parameters unknown after decades

Nature of hidden orders

1. Magnetic multipolar order
   Quadrupolar order
   Octupolar order
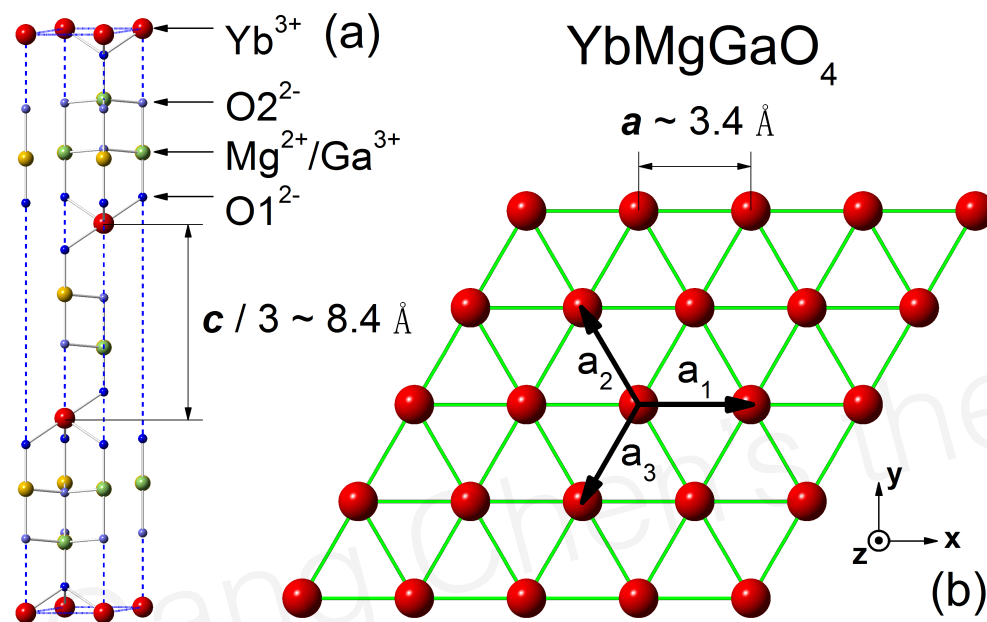
2. Electric multipolar order

3. Orbital order

…

How to probe these hidden orders?

Rare-earth triangular lattice magnets

A rare-earth triangular lattice quantum spin liquid: **YbMgGaO$_4$**

collaboration with QM Zhang, Jun Zhao, Yuesheng Li, Yaodong Li

Qingming Zhang
(Renmin)

- Hastings-Oshikawa-Lieb-Shultz-Mattis theorem.

- Recent extension to spin-orbit coupled insulators (Watanabe, Po, Vishwanath, Zaletel, PNAS 2015).

- This is likely the **first strong spin-orbit coupled QSL with odd electron filling** and effective spin-1/2.

- It is the first clear observation of $T^{2/3}$ heat capacity. (needs comment.)

- Inelastic neutron scattering is consistent with spinon Fermi surface results.

- We think it is a spinon Fermi surface U(1) QSL.

Inelastic neutron scattering performed by Jun Zhao's group and M Mourigal's group

YMGO is not alone: **lots of isostructural materials**

| Compound	Magnetic ion	Space group	Local moment	Θ_{CW} (K)	Magnetic transition	Frustration para. f	Refs.
$YbMgGaO_4$	$Yb^{3+}(4f^{13})$	$R\bar{3}m$	Kramers doublet	-4	PM down to 60 mK	$f > 66$	[4]
$CeCd_3P_3$	$Ce^{3+}(4f^1)$	$P6_3/mmc$	Kramers doublet	-60	PM down to 0.48 K	$f > 200$	[5]
$CeZn_3P_3$	$Ce^{3+}(4f^1)$	$P6_3/mmc$	Kramers doublet	-6.6	AFM order at 0.8 K	$f = 8.2$	[7]
$CeZn_3As_3$	$Ce^{3+}(4f^1)$	$P6_3/mmc$	Kramers doublet	-62	Unknown	Unknown	[8]
$PrZn_3As_3$	$Pr^{3+}(4f^2)$	$P6_3/mmc$	Non-Kramers doublet	-18	Unknown	Unknown	[8]
$NdZn_3As_3$	$Nd^{3+}(4f^3)$	$P6_3/mmc$	Kramers doublet	-11	Unknown	Unknown	[8]

YD Li, XQ Wang, GC*, PRB 94, 035107 (2016)

Magnetism in the $KBaRE(BO_3)_2$ (RE=Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) series: materials with a triangular rare earth lattice

M. B. Sanders, F. A. Cevallos, R. J. Cava
Department of Chemistry, Princeton University, Princeton, New Jersey 08544

Many ternary chalcogenides NaRES2 , NaRESe2 , KRES2 , KRESe2 , KRETe2 , RbRES2 , RbRESe2 , RbRETe2 , CsRES2 , CsRESe2, etc.)

C Liu, YD Li, GC*, PRB 98, 045119 (2018)

$KBaRE(BO_3)_2$

RE Magnetic Lattice

RE   K/Ba   B
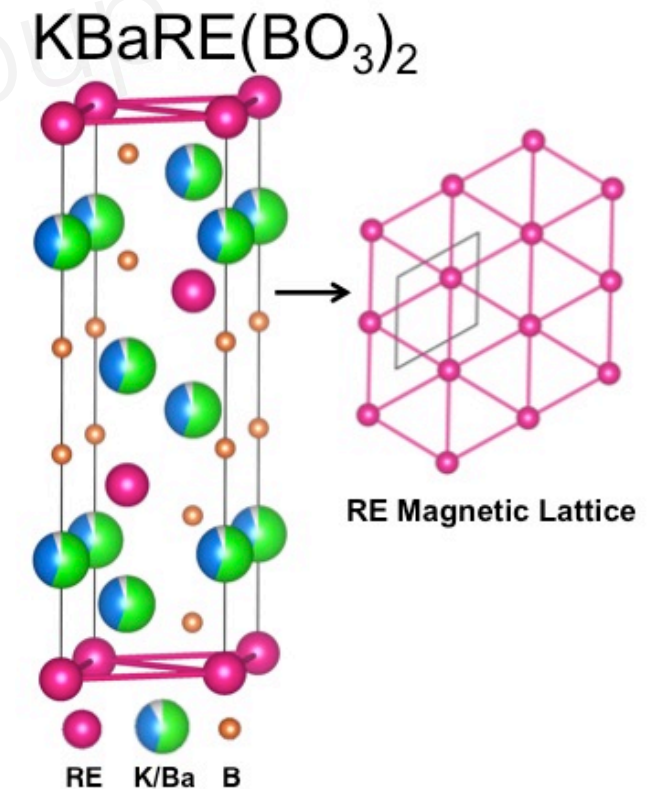
Model Hamiltonian for all cases

TABLE I. The relevant spin Hamiltonians for three different doublets on the triangular lattice. The models for th
and the dipole-octupole doublet have been obtained in the previous works.

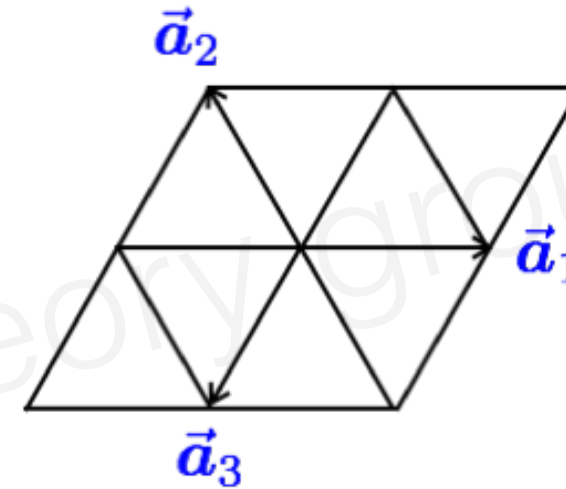| Local doublets	The nearest-neighbor spin Hamiltonians on the triangular lattice
Usual Kramers doublet	$H = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-) - \frac{iJ_{z\pm}}{2}$ $[(\gamma_{ij}^* S_i^+ - \gamma_{ij} S_i^-)S_j^z + S_i^z(\gamma_{ij}^* S_j^+ - \gamma_{ij} S_j^-)]$
Dipole-octupole doublet	$H = \sum_{\langle ij \rangle} J_z S_i^z S_j^z + J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_{yz}(S_i^z S_j^y + S_i^y S_j^z)$
Non-Kramers doublet	$H = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-)$

Model for non-Kramers doublets
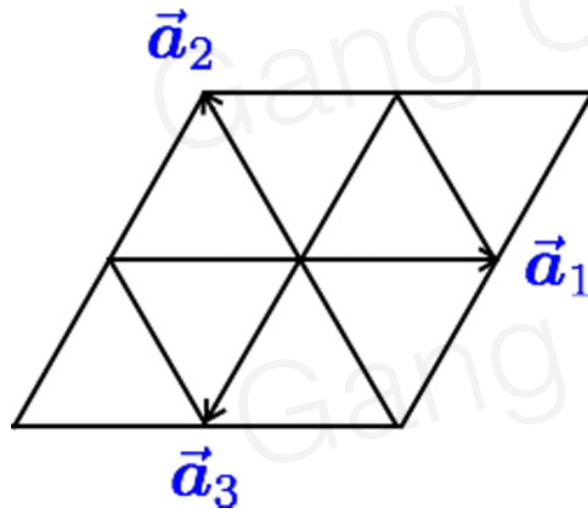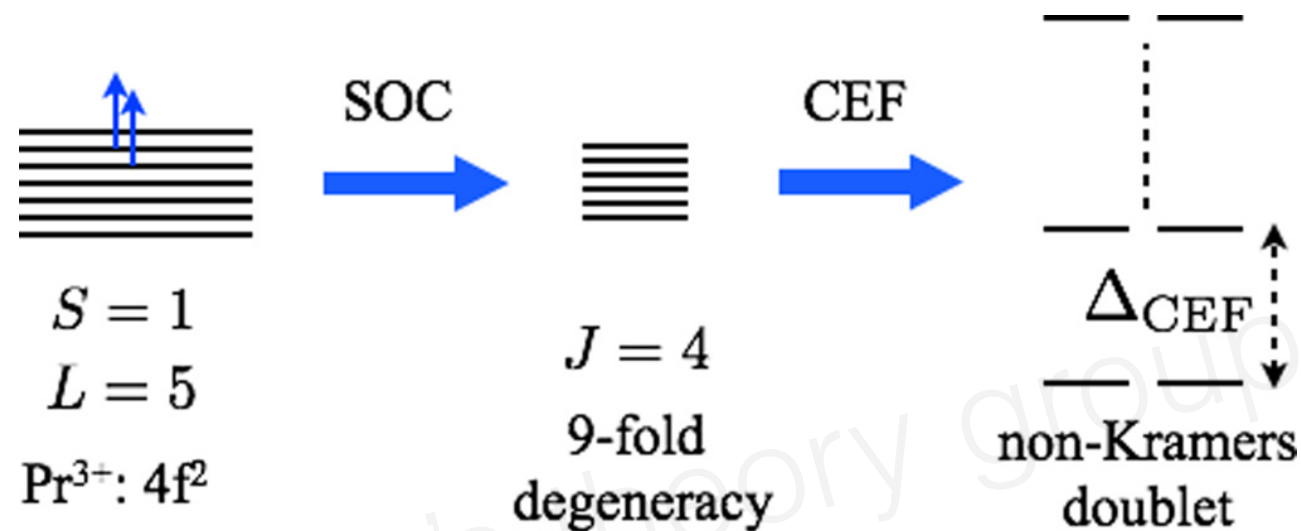
$$H = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+)$$

$$+ J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-), \tag{1}$$

in which, γ_{ij} is a bond-dependent phase factor, and takes 1, $e^{i2\pi/3}$ and $e^{-i2\pi/3}$ on the a_1, a_2, and a_3 bond (see Fig. 2),

Time reversal symmetry forbids the coupling between transverse and Ising components.

Kitaev interaction

$$S_i^a \equiv \sqrt{\frac{2}{3}} S_i^x + \sqrt{\frac{1}{3}} S_i^z,$$

$$S_i^b \equiv \sqrt{\frac{2}{3}} (-\frac{1}{2} S_i^x + \frac{\sqrt{3}}{2} S_i^y) + \sqrt{\frac{1}{3}} S_i^z,$$

$$S_i^c \equiv \sqrt{\frac{2}{3}} (-\frac{1}{2} S_i^x - \frac{\sqrt{3}}{2} S_i^y) + \sqrt{\frac{1}{3}} S_i^z,$$
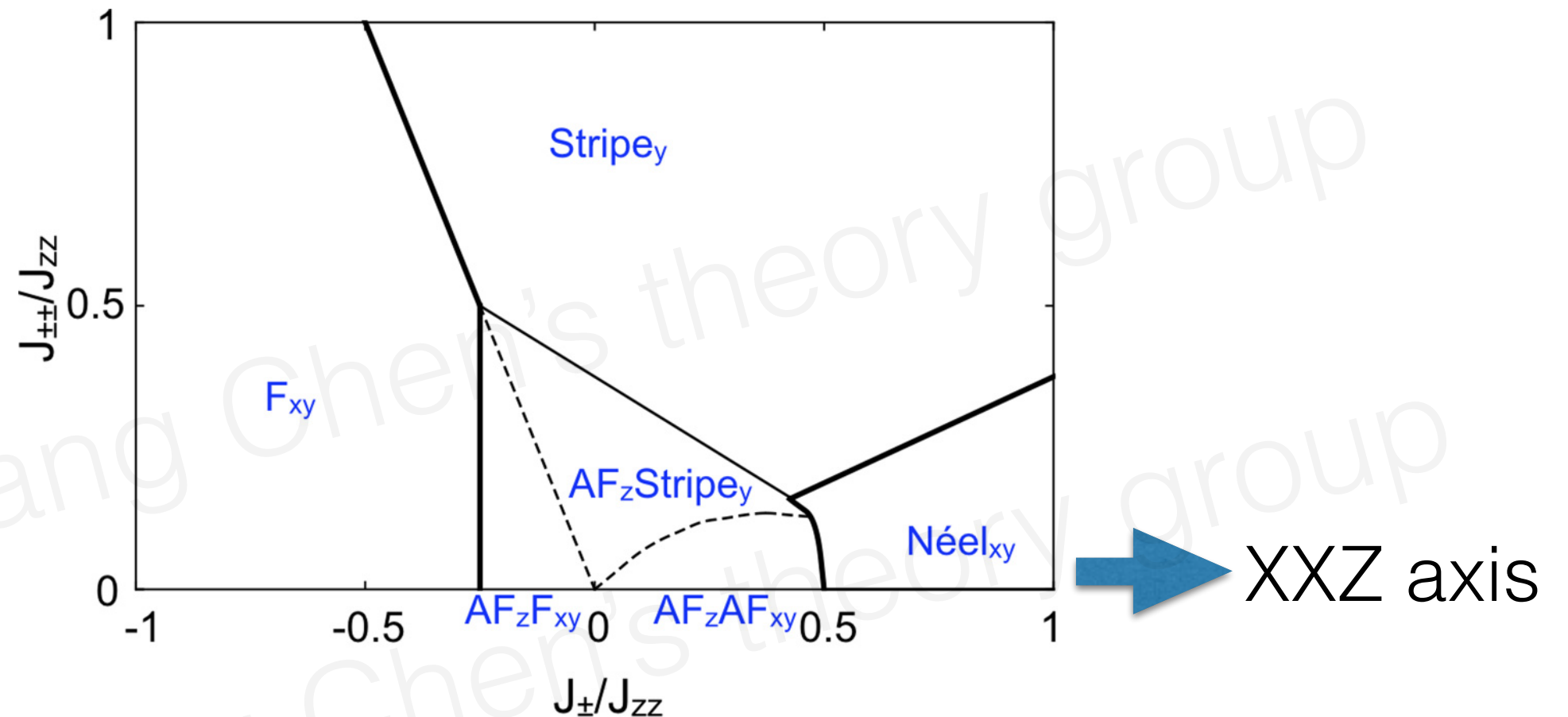

$$
\begin{aligned}
H = \sum_{\langle ij \rangle \in \alpha} & \Big[J \boldsymbol{S}_i \cdot \boldsymbol{S}_j + K S_i^\alpha S_j^\alpha \\
& + \sum_{\beta,\gamma \neq \alpha} \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha) \\
& + \sum_{\beta,\gamma \neq \alpha} (K + \Gamma)(S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta) \Big],
\end{aligned}
$$

Kitaev material beyond iridates: the advantage of f electrons.

pointed out in Fei-Ye Li, YD Li, ..., GC, arXiv 2016, PRB 2017

Non-Kramers doublets: intertwined multipolar orders

We first notice that in the model, the spin rotation around the z direction by $\pi/4$ transforms $S^{\pm} \to \mp i\, S^{\pm}$ and the couplings in the model transform as

$$J_{zz} \to J_{zz}, \quad J_{\pm} \to J_{\pm}, \quad J_{\pm\pm} \to -J_{\pm\pm}. \qquad (2)$$

combined consequence of geometrical frustration
and multipolar nature of the local moments

TABLE II. The list of ordered phases in the phase diagram of Fig. 3.

States	Order types	Elastic neutron
F_{xy}	pure quadrupolar	no Bragg peak
$120°$ Néel	pure quadrupolar	no Bragg peak
$Stripe_y$	pure quadrupolar	no Bragg peak
AF_zF_{xy}	intertwined multipolar	Bragg peak at K
AF_zAF_{xy}	intertwined multipolar	Bragg peak at K
$AF_zStripe_y$	intertwined multipolar	Bragg peak at K

(a) F_{xy} (b) $Stripe_y$ (c) $Néel_{xy}$

(d) AF_zF_{xy} (e) AF_zAF_{xy} (f) $AF_zStripe_y$
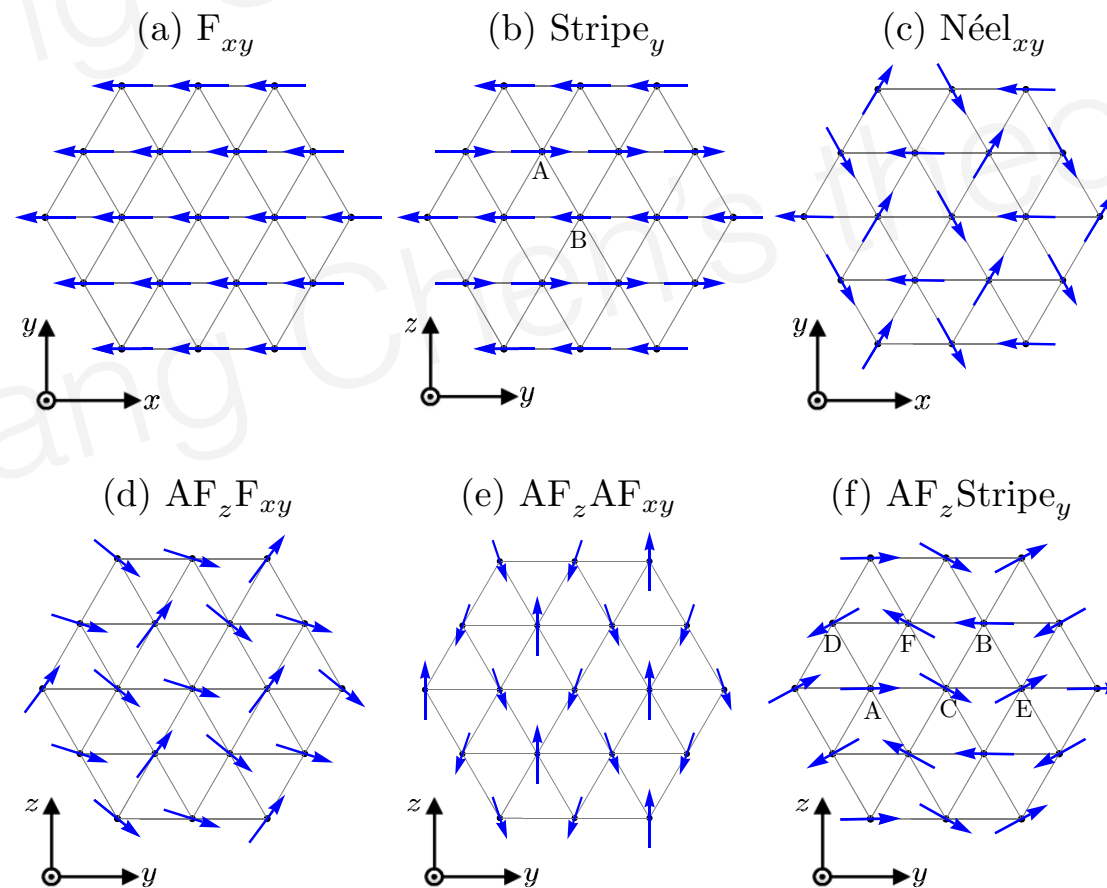
Quantum order by disorder
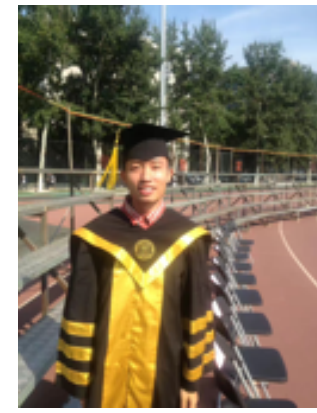
Changle Liu
Fudan

s. Assuming spins with sublattice index s has the direction pointing along the unit vector $\mathbf{n}_s$, one can always associate two unit vectors $\mathbf{u}_s \cdot \mathbf{n}_s = 0$ and $\mathbf{v}_s = \mathbf{n}_s \times \mathbf{u}_s$ so that $\mathbf{n}_s$, $\mathbf{u}_s$ and $\mathbf{v}_s$ are orthogonal with each other. Then we perform Holstein-Primakoff transformation for the spin operator $\mathbf{S}_{\mathbf{r}s}$,

$$\mathbf{n}_s \cdot \mathbf{S}_{\mathbf{r}s} = S - b_{\mathbf{r}s}^\dagger b_{\mathbf{r}s}, \tag{4}$$

$$(\mathbf{u}_s + i\mathbf{v}_s) \cdot \mathbf{S}_{\mathbf{r}s} = (2S - b_{\mathbf{r}s}^\dagger b_{\mathbf{r}s})^{\frac{1}{2}} b_{\mathbf{r}s}, \tag{5}$$

$$(\mathbf{u}_s - i\mathbf{v}_s) \cdot \mathbf{S}_{\mathbf{r}s} = b_{\mathbf{r}s}^\dagger (2S - b_{\mathbf{r}s}^\dagger b_{\mathbf{r}s})^{\frac{1}{2}}. \tag{6}$$

After performing Fourier transformation

$$b_{\mathbf{r}s} = \sqrt{\frac{M}{N}} \sum_{\mathbf{k}\in\overline{\mathrm{BZ}}} b_{\mathbf{k}s} e^{i\mathbf{R}_{\mathbf{r}s}\cdot\mathbf{k}}, \tag{7}$$

the spin Hamiltonian can be rewritten in terms of boson bilinears as

$$H_{\mathrm{sw}} = E_0 + \frac{1}{2} \sum_{\mathbf{k}\in\overline{\mathrm{BZ}}} \left[\Psi(\mathbf{k})^\dagger h(\mathbf{k})\Psi(\mathbf{k}) - \frac{1}{2}\mathrm{tr}\, h(\mathbf{k}) \right], \tag{8}$$

where E_0 is the mean-field energy,

$$\Psi(\mathbf{k}) = [b_{\mathbf{k}1}, \dots, b_{\mathbf{k}M}, b_{-\mathbf{k}1}^\dagger, \dots, b_{-\mathbf{k}M}^\dagger]^T, \tag{9}$$

and $h(\mathbf{k})$ is a $2M \times 2M$ Hermitian matrix, and $\overline{\mathrm{BZ}}$ is the magnetic Brillouin zone. H_{sw} can be diagonalized via a standard Bogoliubov transformation $\Psi(\mathbf{k}) = T_{\mathbf{k}}\Phi(\mathbf{k})$ where

$$\Phi(\mathbf{k}) = [\beta_{\mathbf{k}1}, \dots, \beta_{\mathbf{k}M}, \beta_{-\mathbf{k}1}^\dagger, \dots, \beta_{-\mathbf{k}M}^\dagger]^T, \tag{10}$$

and $T_{\mathbf{k}} \in SU(M,M)$. Here $SU(M,M)$ refers to indefinite special unitary group that is defined as [43]

$$SU(M,M) = \{g \in \mathbb{C}_{2M\times 2M} : g^\dagger \Sigma g = \Sigma, \det g = 1\}, \tag{11}$$

where Σ is the metric tensor and given as

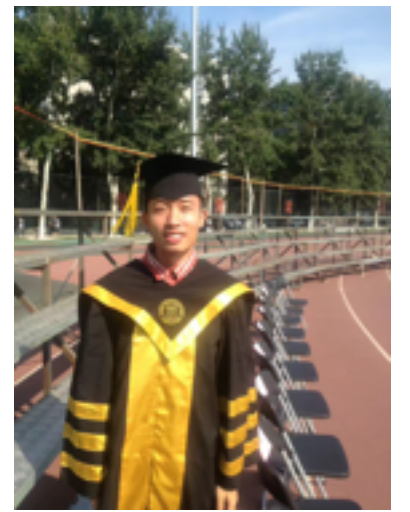$$\Sigma = \begin{pmatrix} I_{M\times M} & 0 \\ 0 & -I_{M\times M} \end{pmatrix}. \tag{12}$$

It is straightforward to prove that such transformation preserves the boson commutation rules. The diagonalized Hamiltonian reads

$$\begin{aligned} H_{\mathrm{sw}} &= E_0 + \frac{1}{2} \sum_{\mathbf{k}\in\overline{\mathrm{BZ}}} \left[\Phi(\mathbf{k})^\dagger E(\mathbf{k})\Phi(\mathbf{k}) - \frac{1}{2}\mathrm{tr}\, h(\mathbf{k}) \right] \\ &= E_0 + E_r + \sum_{\mathbf{k}\in\overline{\mathrm{BZ}}} \omega_{\mathbf{k}s} \beta_{\mathbf{k}s}^\dagger \beta_{\mathbf{k}s}, \end{aligned} \tag{13}$$

where $E(\mathbf{k}) = \mathrm{diag}[\omega_{\mathbf{k}1}, \dots, \omega_{\mathbf{k}M}, \omega_{-\mathbf{k}1}, \dots, \omega_{-\mathbf{k}M}]$ and

$$E_r = \frac{1}{4} \sum_{\mathbf{k}\in\overline{\mathrm{BZ}}} \mathrm{tr}\,[E(\mathbf{k}) - h(\mathbf{k})] \tag{14}$$
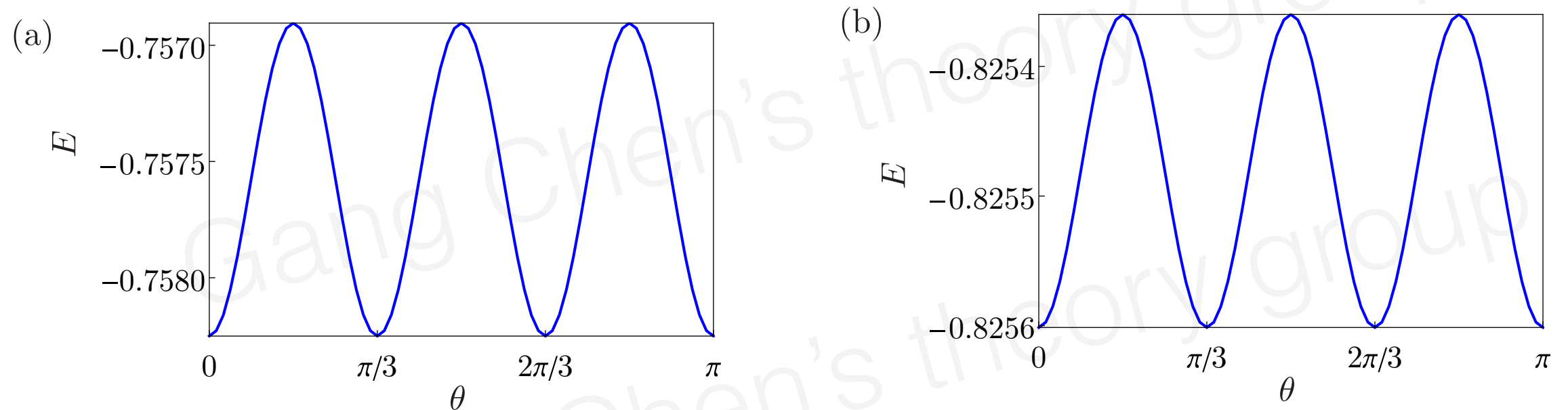
Quantum order by disorder

Changle Liu
Fudan

FIG. 5. Energy per spin taking into account quantum zero-point energy vs the azimuth angle θ of spins for (a) the F_{xy} state and (b) the 120° Néel$_{xy}$ state. Here we take the parameter $J_{\pm} = 0.4J_{zz}$, $J_{\pm\pm} = 0.4J_{zz}$ for the F_{xy} state and parameter $J_{\pm} = 0.9J_{zz}$, $J_{\pm\pm} = 0.2J_{zz}$ for the Néel$_{xy}$ state. The zero-point energy is calculated within the linear spin-wave method.
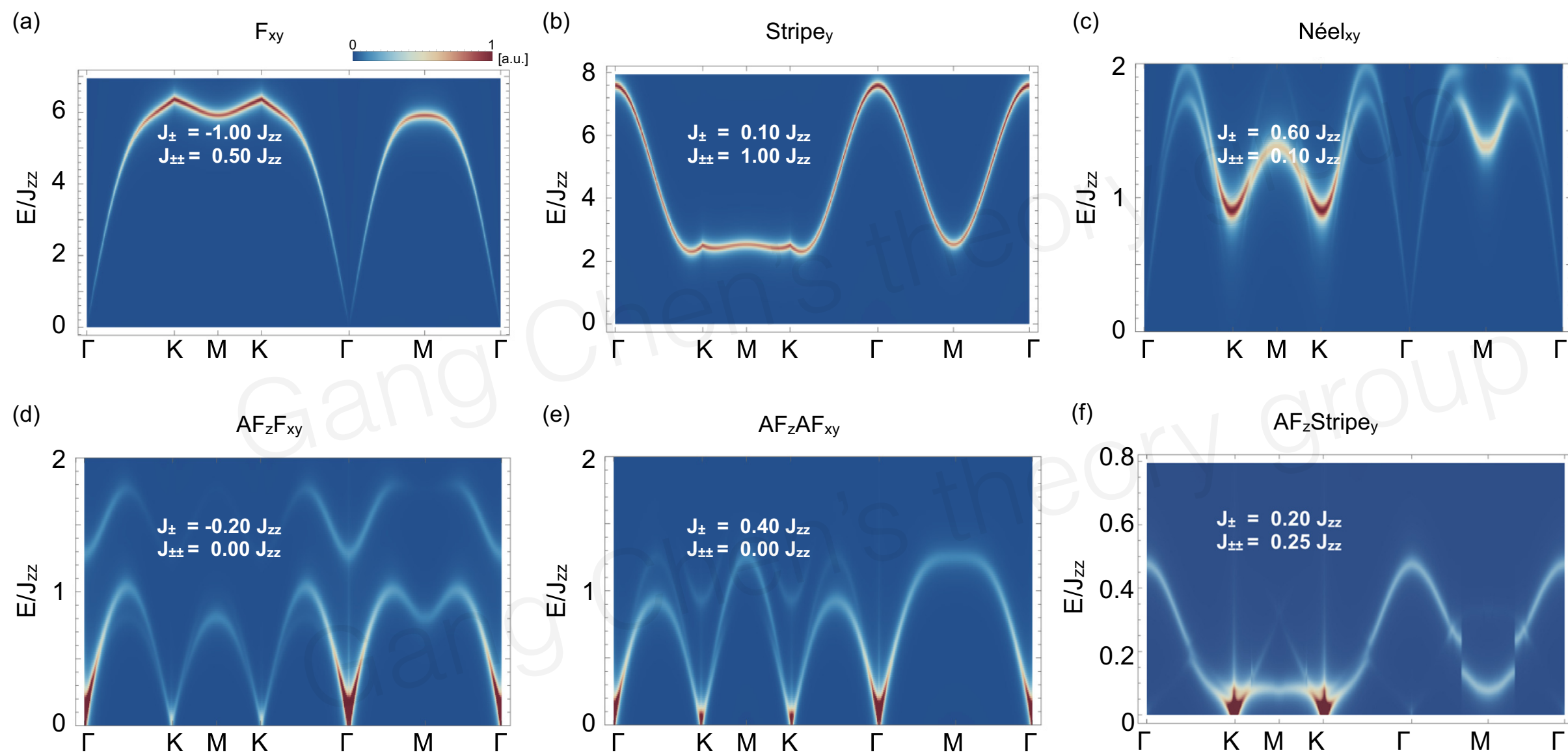
The idea of non-commutative observables

To detect intertwined multipolar orders, one can combine both elastic and inelastic neutron scattering measurements.

$$\mathcal{S}^{zz}(\mathbf{q}, \omega > 0)$$

$$= \frac{1}{2\pi N} \sum_{ij} \int_{-\infty}^{+\infty} dt \, e^{i\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j) - i\omega t} \langle S_i^z(0) S_j^z(t) \rangle.$$

as if one is doing polarized neutron scattering measurements.

Detection of the intertwined multipolar orders: excitations

(a) F_{xy}
$J_{\pm} = -1.00\ J_{zz}$
$J_{\pm\pm} = 0.50\ J_{zz}$

(b) Stripe$_y$
$J_{\pm} = 0.10\ J_{zz}$
$J_{\pm\pm} = 1.00\ J_{zz}$

(c) Néel$_{xy}$
$J_{\pm} = 0.60\ J_{zz}$
$J_{\pm\pm} = 0.10\ J_{zz}$

(d) AF$_z$F$_{xy}$
$J_{\pm} = -0.20\ J_{zz}$
$J_{\pm\pm} = 0.00\ J_{zz}$

(e) AF$_z$AF$_{xy}$
$J_{\pm} = 0.40\ J_{zz}$
$J_{\pm\pm} = 0.00\ J_{zz}$

(f) AF$_z$Stripe$_y$
$J_{\pm} = 0.20\ J_{zz}$
$J_{\pm\pm} = 0.25\ J_{zz}$

Selection rules

Yaodong Li
Fudan -> UCSB

Changle Liu
Fudan

selection rule associated with the symmetry generated by

$$\hat{W} = T_{-\mathbf{a}_1+\mathbf{a}_2} \otimes e^{i\pi \sum_j S_j^z}, \qquad (18)$$

where $T_{-\mathbf{a}_1+\mathbf{a}_2}$ denotes the lattice translation by $-\mathbf{a}_1 + \mathbf{a}_2$. The Hamiltonian stays invariant under $\hat{W}$, $[\hat{W}, H] = 0$.

From now on, we introduce the notation s and $\bar{s}$ to denote the sublattice pair that is interchanged under the action of $\hat{W}$. In the labeling of Fig. 4, we find that $\bar{A} = B, \bar{C} = D, \bar{E} = F$.

For the elementary excitations, the effect of $\hat{W}$ is such that

$$\text{Stripe}_y : \hat{W} b_{\mathbf{k},s} \hat{W}^\dagger = e^{i\phi(\mathbf{k})} b_{\mathbf{k},\bar{s}}, s = A, B, \qquad (19)$$

$$\text{Stripe}_y \text{AF}_z : \hat{W} b_{\mathbf{k},s} \hat{W}^\dagger = e^{i\phi(\mathbf{k})} b_{\mathbf{k},\bar{s}}, s = A, \ldots, F, \qquad (20)$$

where $\phi(\mathbf{k}) = -k_x + k_y$.

The eigenmodes of $\hat{W}$ take bonding/antibonding form,

$$\alpha_{\mathbf{k},s,\pm} = b_{\mathbf{k},s} \pm b_{\mathbf{k},\bar{s}}, \qquad (21)$$

whose eigenvalues are

$$\hat{W} \alpha_{\mathbf{k},s,\pm} \hat{W}^\dagger = \pm e^{i\phi(\mathbf{k})} \alpha_{\mathbf{k},s,\pm}. \qquad (22)$$
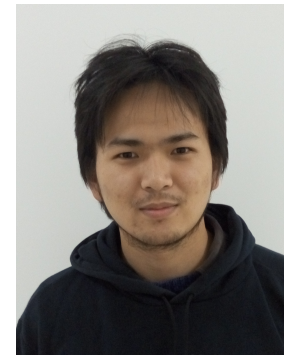
Since $\hat{W}$ is a symmetry of the Hamiltonian, the energy eigenmodes are separate linear combinations of $\alpha_{\mathbf{k},s,\pm}$,

$$\beta_{\mathbf{k},t,\pm} = \sum_s c_{t,s} \alpha_{\mathbf{k},s,\pm} + d_{t,s} \alpha^\dagger_{-\mathbf{k},s,\pm}, \qquad (23)$$
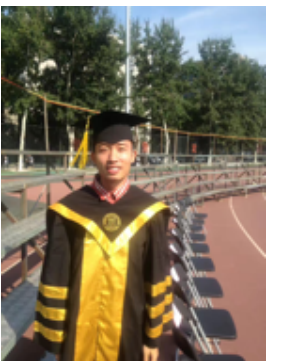
and

$$\hat{W} \beta_{\mathbf{k},t,\pm} \hat{W}^\dagger = \pm e^{i\phi(\mathbf{k})} \beta_{\mathbf{k},t,\pm}. \qquad (24)$$

The $\pm$ branches do not mix, since they have distinct eigenvalues under $\hat{W}$.

$$S^{zz}(\mathbf{q}, \omega > 0) = \sum_n \langle 0| \sum_{s=1} S_s^z(-\mathbf{q}, -\omega)|n\rangle \langle n| \sum_{s=1} S_s^z(\mathbf{q}, \omega)|0\rangle$$

$$\propto \sum_n \delta(\omega - (\epsilon_n - \epsilon_0)) \langle 0| \sum_{s=1}^M (b_{\mathbf{q},s} + b^\dagger_{-\mathbf{q},s})|n\rangle \langle n| \sum_{s=1}^M (b_{-\mathbf{q},s} + b^\dagger_{\mathbf{q},s})|0\rangle$$

$$\propto \sum_n \delta(\omega - (\epsilon_n - \epsilon_0)) \langle 0| \sum_{s=1}^M (\alpha_{\mathbf{q},s,+} + \alpha^\dagger_{-\mathbf{q},s,+})|n\rangle \langle n| \sum_{s=1}^M (\alpha_{-\mathbf{q},s,+} + \alpha^\dagger_{\mathbf{q},s,+})|0\rangle.$$

It is thus obvious that the contribution is nonzero if and only if $|n\rangle$ is created by the $\beta_{\mathbf{k},t,+}$ operators. The $\beta_{\mathbf{k},t,-}$ states are not accessible. As a result, the S^z-S^z correlation function only measures coherent excitations with even parity. The odd parity excitations, instead, are present in S^x-S^x and S^y-S^y correlation functions.

Discovery of intertwined multipolar order in TmMgGaO4
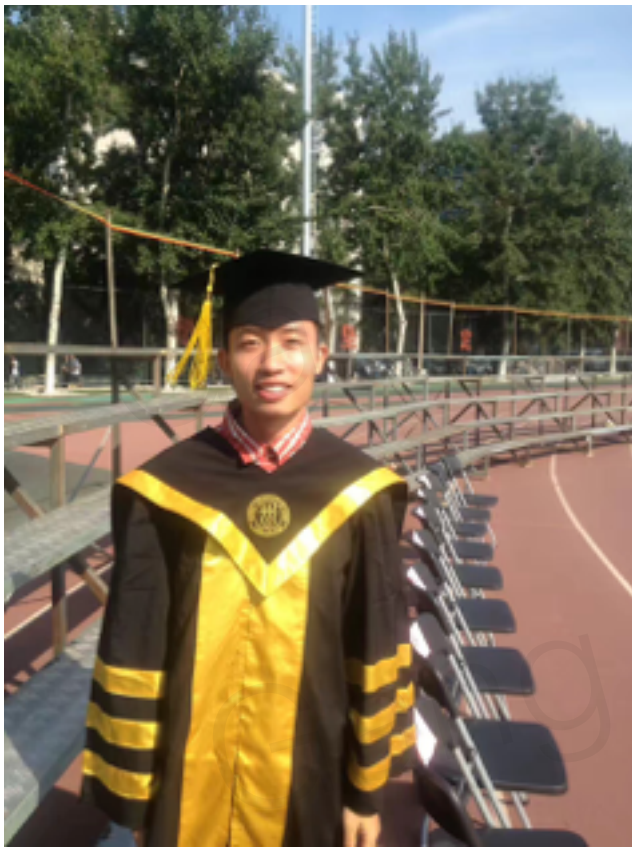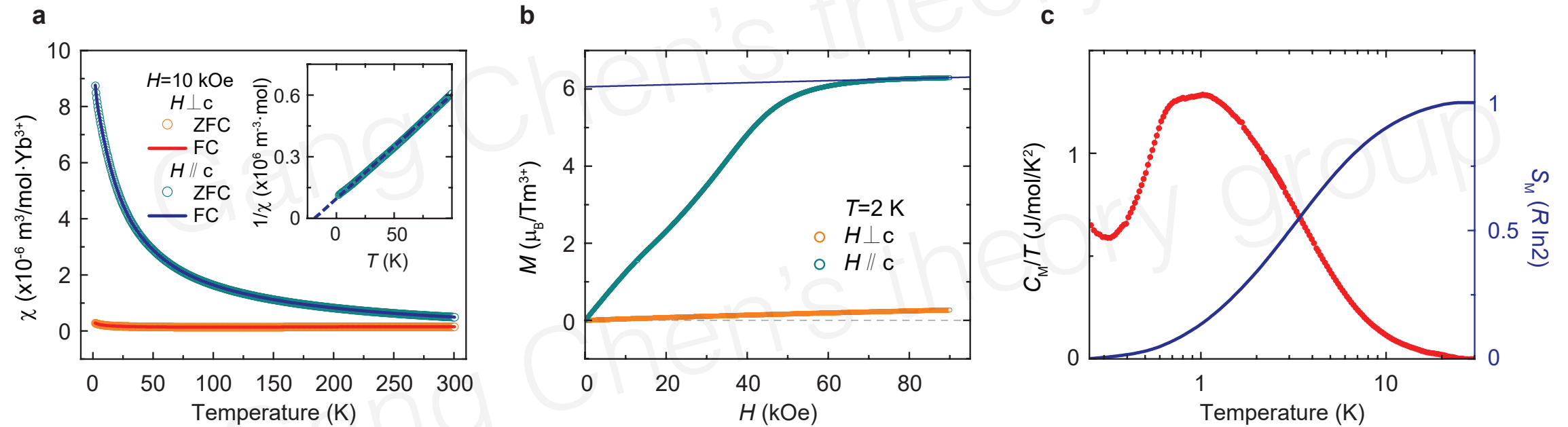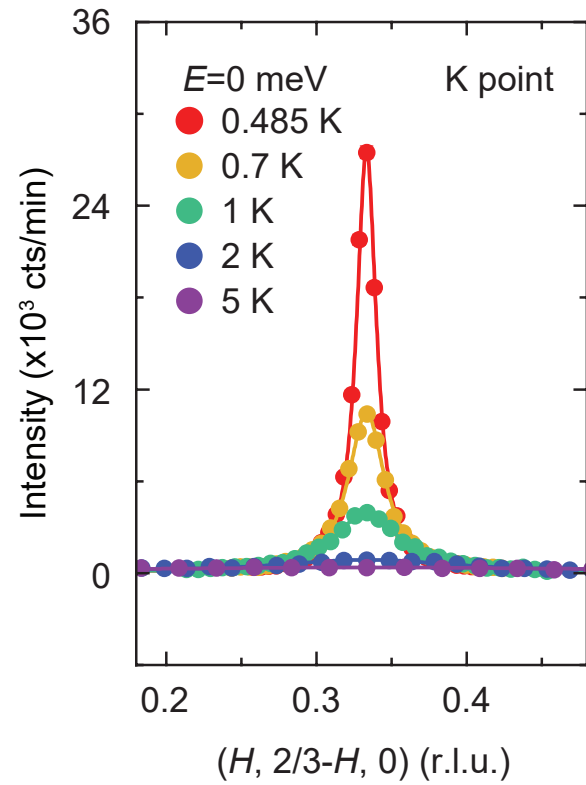

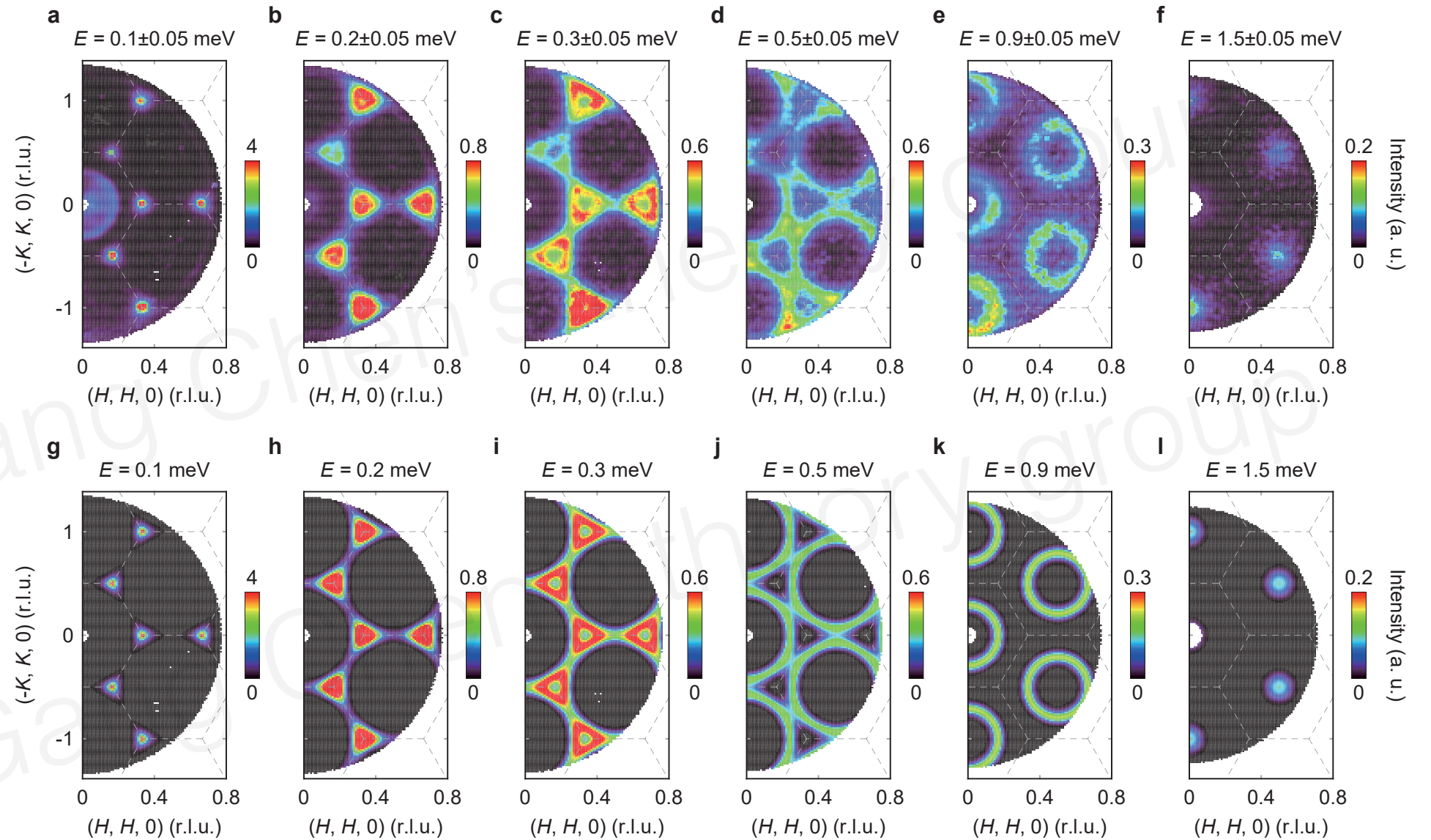
Changle Liu
(Fudan)
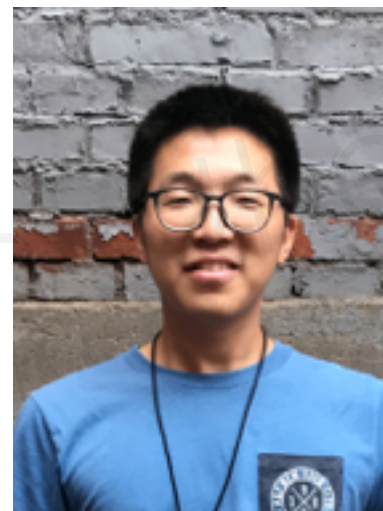
**Yao Shen
(Fudan)**

**Jun Zhao
(Fudan)**

approximately thought as non-Kramers doublets
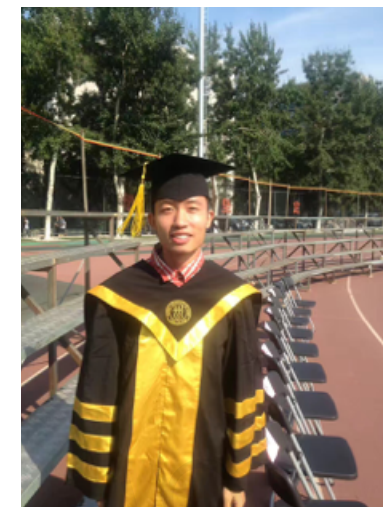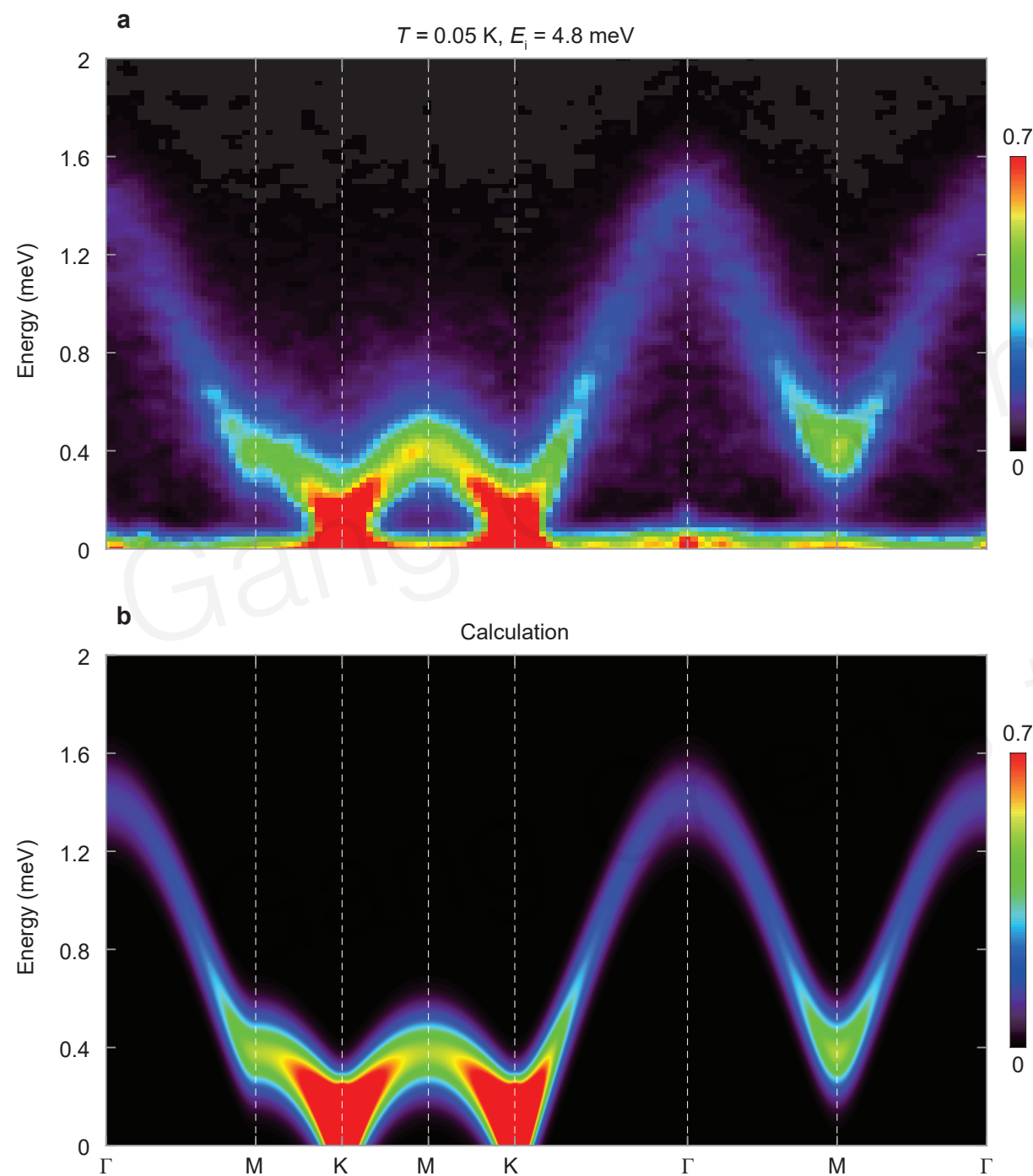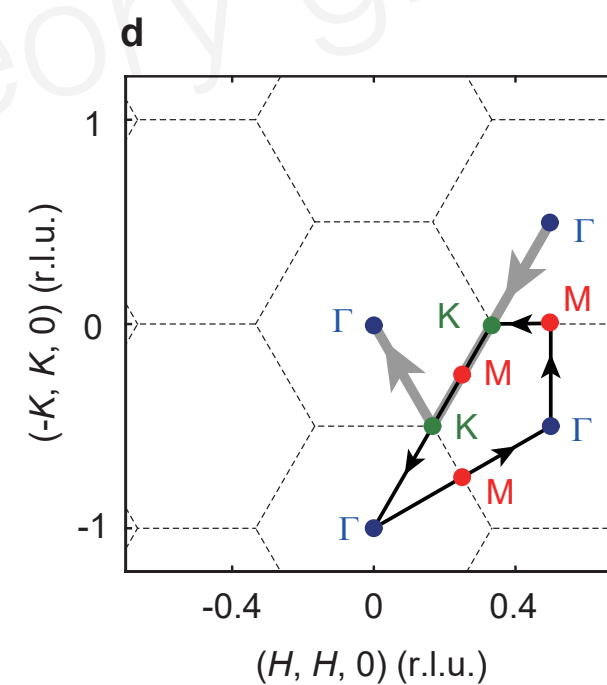
Well-defined spin wave

The presence of well-defined spin wave indicates the presence of the hidden order !

Comparison with theory

Changle Liu

Summary

1. The interplay between geometrical frustration and multipolar local moments leads to rich phases and excitations.

2. The manifestation of the hidden multipolar orders is rather non-trivial, both in the static and dynamic measurements.

3. The non-commutative observables/operators can be used to reveal the dynamics of hidden orders. This is general and can be adapted to many other hidden order systems.

4. Finally, the non-trivial Berry phase effect has not yet been discussed. This thought has been hinted in Kivelson's recent work (PNAS 2018).