Hidden multipolar orders of dipole-octupole doublets on a triangular lattice

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Motivated by the recent development in strong spin-orbit-coupled materials, we consider the dipole-octupole doublets on the triangular lattice. We propose the most general interaction between these unusual local moments. Due to the spin-orbit entanglement and the special form of its wavefunction, the dipole-octupole doublet has a rather peculiar property under the lattice symmetry operation. As a result, the interaction is highly anisotropic in the pseudospin space, but remarkably, is uniform spatially. We analyze the ground state properties of this generic model and emphasize the hidden multipolar orders that emerge from the dipolar and octupolar interactions. We clarify the quantum mutual modulations between the dipolar and octupolar orders. We predict the experimental consequences of the multipolar orders and propose the rare-earth triangular materials as candidate systems for these unusual properties.

Introduction.—In recent years, there has been an intensive interest in exploring electron systems that involve both strong spin-orbit coupling (SOC) and substantial electron correlations, especially in materials with heavy elements such as 5$d$ transition metal elements and 4$f$ rare-earth elements [1–4]. Because of the spatial orientation of the orbitals, the spin-orbit entanglement in strongly correlated Mott insulators often gives rise to rather complicated models that involve both spatial and spin anisotropies [2 5–9]. While this is true for most spin-orbit-entangled moment, in this Letter, we propose a remarkably simple model for a peculiar spin-orbit-entangled doublet, namely “dipole-octupole doublet” (DO doublet) [10, 11] on a triangular lattice, and connect this model with the rare-earth based triangular lattice materials. Due to the multipolar nature of the interaction, this simple but realistic model, in a large parameter regime, realizes hidden magnetic multipolar orders and leads to unexpected experimental consequences.

The search for hidden order is an active field in the $f$ electron systems [12]. The magnetic multipolar order has been proposed for URu$_2$Si$_2$ and NpO$_2$, and various experimental evidence has been found [12–14]. Nevertheless, the precise nature of the multipolar orders in URu$_2$Si$_2$ and NpO$_2$ has not come to a consensus. It is partly because the complication and multitudes of the degree of freedom often prohibit the precise modeling of the multipolar interactions in these systems. In contrast, our model is a precise modeling of the multipolar interactions for the DO doublet systems and might be the simplest such model in the strong spin-orbit-coupled materials that realizes hidden multipolar orders [11, 12].

Three families of triangular lattice materials, MgYbGaO$_4$ [15, 20], the isostructural ternary family R Cd$_3$P$_3$, R Zn$_3$P$_3$, R Cd$_3$As$_3$, R Zn$_3$As$_3$ (R = Ce, Pr, Nd, Sm) [21–23], and R$_2$O$_2$CO$_3$ (R = Nd, Sm, Dy) [24], have recently been discovered. These materials contain the rare-earth elements, whose 4$f$ electrons involve strong SOC and strong correlations. The strong SOC entangles the total electron spin $S$ with the orbital angular momentum $L$ and leads to a total moment $J$. Like the case in the rare-earth pyrochlores [25], the local D$_{3d}$ crystal electric field (CEF) splits the $(2J + 1)$ states into the crystal field states [16]. For a half-integer (integer) $J$, the CEF ground state is a Kramers’ doublet (either a singlet or a non-Kramers’ doublet). The ground state doublets define the low-temperature magnetic properties of the system. In the previous work, we proposed a generic anisotropic spin model for non-Kramers’ doublets and the usual Kramers’ doublets on a triangular
lattice \[16\]. Here we introduce a generic model for the DO Kramers’ doublet on the triangular lattice \[15\] \[16\] and predict the experimental consequences of the hidden multipolar orders.

Dipole-octupole doublet.—The DO doublet is a special type of Kramers’ doublet. It occurs when the crystal field ground state wavefunctions \( |\Psi_\pm\rangle \) are linear superpositions of the states with \( J^z = 3n/2 \) where \( n \) is an odd integer. Unlike the usual Kramers doublets that transform as a two-dimensional irreducible representation of the \( D_{3d} \) point group \[16\], each state of the DO doublet transforms as a one-dimensional irreducible representation \((\Gamma^+_3 \text{ or } \Gamma^+_6)\) of the \( D_{3d} \) point group \[10\]. This crucial difference is most easy to be understood if one applies the 3-fold rotation along the \( z \) axis to these states. Under the 3-fold rotation, we have \( \exp(-i\frac{2\pi}{3}J^z) |J^z = 3n/2\rangle = -|J^z = 3n/2\rangle \). Therefore, the wavefunctions of the DO doublet, \( |\Psi_\pm\rangle \), stay invariant under this rotation except getting an overall minus sign, i.e.,

\[
\exp(-i\frac{2\pi}{3}J^z) |\Psi_\pm\rangle = -|\Psi_\pm\rangle.
\]

In contrast, for the usual Kramers’ doublet, the two states would mix with each other under this rotation. The degeneracy of the DO doublet is protected by time reversal symmetry that switches the two states. This special doublet has been found in various neodymium (Nd) pyrochlores \[29\] \[32\], dysprosium (Dy) pyrochlore \[33\], osmium (Os) pyrochlore \[34\] \[35\], erbiurn (Er) and ytterbium (Yb) spinels \[32\] \[36\], and \( \text{Ce}_2\text{Sn}_2\text{O}_7 \) \[37\]. We expect the DO doublet should occur in some of the rare-earth triangular materials, especially since these rare-earth ions experience the same \( D_{3d} \) crystal field environment.

Generic pseudospin model on a triangular lattice.— Here we explain the interaction between the DO doublets on a triangular lattice. Due to the two-fold degeneracy of the DO doublet, we introduce the pseudospin operators that act on this DO doublet, \( \tau^+ = |\Psi_+\rangle \langle \Psi_-| \), \( \tau^- = |\Psi_-\rangle \langle \Psi_+| \), \( \tau^z = \frac{1}{2} |\Psi_+\rangle \langle \Psi_+| - \frac{1}{2} |\Psi_-\rangle \langle \Psi_-| \), where \( \tau^z \equiv \tau^x + i\tau^y \). To obtain the exchange interaction, we start with the symmetry properties of the pseudospins under the space group symmetry.

For all the three families of rare-earth triangular lattice materials \[15\] \[24\], the space group is either R3m or P6_3mmc. As all rare-earth ions in these materials have a layered triangular structure and the interlayer separation is much larger than the intralayer lattice constant, it is sufficient to just keep the interaction within the triangular layer and ignore the interlayer couplings. As far as the space group symmetry is concerned, we only need to retain the symmetry generators that operate within each triangular layer. It turns out that, for a single triangular layer, both \( \text{R}3\text{m} \) and \( \text{P}6_3\text{mmc} \) space groups give a three-fold rotation around the \( z \) axis, \( C_3 \), a two-fold rotation about the diagonal direction, \( C_2 \), a site inversion symmetry \( I \), and two lattice translations, \( T_x \) and \( T_y \). The symmetry operation on \( \tau^x \) is given as \[38\]

\[
\begin{align*}
C_3 : & \quad \tau^x \rightarrow \tau^y_{C_3(r)} , \quad \tau^y \rightarrow \tau^x_{C_3(r)} , \quad \tau^z \rightarrow \tau^z_{C_3(r)} , \\
C_2 : & \quad \tau^x \rightarrow \tau^y_{C_2(r)} , \quad \tau^y \rightarrow \tau^y_{C_2(r)} , \quad \tau^z \rightarrow \tau^z_{C_2(r)} , \\
I : & \quad \tau^x \rightarrow \tau^x_{I(r)} , \quad \tau^y \rightarrow \tau^y_{I(r)} , \quad \tau^z \rightarrow \tau^z_{I(r)} , \\
T_x : & \quad \tau^x \rightarrow \tau^z_{T_x(r)} , \quad \tau^y \rightarrow \tau^y_{T_x(r)} , \quad \tau^z \rightarrow \tau^y_{T_x(r)} , \\
T_y : & \quad \tau^x \rightarrow \tau^z_{T_y(r)} , \quad \tau^y \rightarrow \tau^y_{T_y(r)} , \quad \tau^z \rightarrow \tau^y_{T_y(r)} .
\end{align*}
\]

Since the 4f electron wavefunction is very localized, we only need to keep the nearest-neighbor interactions. The most general nearest-neighbor model, allowed by the above symmetries, is given as

\[
H_0 = \sum_{\langle rr' \rangle} \left[ J_x \tau^x_{rr} \tau^x_{rr'} + J_y \tau^y_{rr} \tau^y_{rr'} + J_z \tau^z_{rr} \tau^z_{rr'} \\
+ J_y (\tau^x_{rr} \tau^y_{rr'} + \tau^y_{rr} \tau^x_{rr'}) \right].
\]

Here we give a few comments on this model. First of all, the pseudospin interaction is anisotropic in the pseudospin space because of the spin-orbit entanglement in the DO doublet. What is surprising is that the interaction is spatially uniform and is identical for every bond orientation. This is unusual since the orbitals have orientations. This remarkable spatial property comes from the peculiar symmetry property of the DO doublet in Eq. (2). Secondly, there exists a crossing coupling between \( \tau^y \) and \( \tau^z \) because \( \tau^y \) and \( \tau^z \) transform identically and behave like the magnetic dipole moments under the space group. Thirdly, there is no crossing coupling between \( \tau^x \) and \( \tau^y \) or \( \tau^z \) because \( \tau^x \) transforms as an octupole moment under the space group. This holds even for further neighbor interactions \[39\]. The \( J_x \) interaction is the interaction between the octupole moments.

Another remarkable property of the DO doublet is the infinite anisotropy in the Landé \( g \)-factor when it couples to an external magnetic field. After including the Zeeman term, we have the full Hamiltonian \( H = H_0 - \hbar \sum_r \tau^x_r \). Due to the spatial uniformity of the interaction, we are able to implement a rotation by an angle \( \theta \) around the \( x \) direction in the pseudospin space and eliminate the crossing coupling between \( \tau^y \) and \( \tau^z \). The reduced model is given as

\[
H = \sum_{\langle rr' \rangle} \left[ J_x T_x^r T_x^{r'} + J_y T_y^r T_y^{r'} + J_z T_z^r T_z^{r'} \right] \\
- \hbar \sum_r \left[ \cos \theta T_z^r + \sin \theta T_y^r \right],
\]

where \( T_x = \tau^x, T_y = \tau^z \sin \theta + \tau^y \cos \theta, T_z = \tau^z \cos \theta - \tau^y \sin \theta, \) and \( J_x, J_y, J_z \) are defined in the Supplementary information. Note both \( T^y \) and \( T^z \) behave like dipole moments. Like the XYZ model on the pyrochlore lattice \[10\] \[11\], this model does not have a sign problem for quantum Monte Carlo simulation in a large parameter regime, and this is valid on any other lattices such as the 3D FCC lattice where DO doublets could exist \[8\].
Hidden ferro-octupolar orders.—We now explain the hidden multipolar orders of the model in Eq. (4). We start with the parameter regime on the I_a surface with \( J_z = -1 \) (see Fig. 1). This regime simply gives a conventional ferromagnetic ground state with a uniform \( \langle T^z \rangle \). Since \( T^z \) is a dipole moment, this state is dubbed ferro-dipolar (FD_y) state, where the subindex \( y \) refers to the direction of the dipole moment. With a ferromagnetic dipole moment, this state can be readily confirmed in a magnetization measurement.

The reduced model in Eq. (4) has an interesting permutation structure. Using the result on the I_x surface, we can generate the ground states on the I_y surface with \( J_y = -1 \) and the I_z surface with \( J_z = -1 \). As the FD_y order of the I_y surface shares the same symmetry as the FD_x order of the I_x surface, we do not give a repeated discussion here. Although the permutation trick to relate different regimes seems simple, the physics on the I_x surface is rather special and unconventional, and it is this distinction that we clarify below. Clearly, as \( \langle T^x \rangle \) is uniform and non-zero on the I_x surface, time reversal symmetry is explicitly broken and the ground state is a ferromagnetic state. As we compute within the mean-field theory in the Supplementary Information and show in Fig. 1, however, the magnetic susceptibility does not show any divergent behavior. This is very different from what we would naively expect for an usual ferromagnetic state. The order parameter \( \langle T^x \rangle \) is an octupole moment and does not couple linearly to the external magnetic field. Therefore, it is hidden in the usual magnetization measurement.

Despite its invisibility in the usual thermodynamic measurements, one could instead search for the evidence of the octupolar order by other experimental probes. Since the octupolar order explicitly breaks time reversal symmetry, polar Kerr effect could be used to detect the time reversal symmetry breaking [40]. Moreover, inside the FO phase, the dipole moment \( \tau^z \) flips the octupole moment and creates octupolar-wave excitations. As \( \tau^z \) directly couples to the neutron spin, the octupolar-wave excitation can be directly detected by an inelastic neutron scattering experiment. Using the Holstein-Primakoff boson transformation [38], we obtain the octupolar-wave dispersion,

\[
\omega_k = \left[ J_y \sum_i \cos |\mathbf{k} \cdot \mathbf{a}_i| - 3J_z \right]^{1/2} \times \left[ J_z \sum_i \cos |\mathbf{k} \cdot \mathbf{a}_i| - 3J_x \right]^{1/2},
\]

where the summation is over the three nearest neighboring vectors \( \mathbf{a}_1 = (1, 0) \), \( \mathbf{a}_2 = (-1/2, \sqrt{3}/2) \), and \( \mathbf{a}_3 = (-1/2, -\sqrt{3}/2) \). One should observe a well-defined octupolar wave excitation below the FO transition despite the absence of ordering in the magnetization measurement. This mode is generically gapped because of the low symmetry of the model. We depict the octupolar wave excitation in Fig. 1.

Hidden antiferro-octupolar orders.—Here we consider the parameter regimes where the dominant interaction is antiferromagnetic. We focus on the O_x surface where the octupolar exchange coupling \( J_x \) is antiferromagnetic and dominant. For the O_x and the O_y surfaces, one can apply the permutation on the O_x surface and generate the phase diagrams and the relevant phases. In the absence of the exchange couplings \( J_y \) and \( J_z \), the Ising exchange interaction \( J_x \) is highly frustrated on the triangular lattice. Any state that satisfies the “2-plus 1-minus” or “2-minus 1-plus” condition for the \( T^x \) configuration on every triangle is the ground state. Therefore, the ground state is extensively degenerate.

In the XXZ limit of the model with \( J_y = J_z \), the weak \( J_y \) and \( J_z \) exchanges allows the system to tunnel quantum mechanically within the degenerate ground state manifold and lifts the degeneracy via an order by quantum disorder effect [41–44]. It is well established that the system develops a supersolid order in a large quantum disorder effect [41–44]. We focus on the O_x surface and generate the phase diagrams and the relevant phases. In the absence of the exchange couplings \( J_y \) and \( J_z \), the Ising exchange interaction \( J_x \) is highly frustrated on the triangular lattice. Any state that satisfies the “2-plus 1-minus” or “2-minus 1-plus” condition for the \( T^x \) configuration on every triangle is the ground state. Therefore, the ground state is extensively degenerate. In the XXZ limit of the model with \( J_y = J_z \), the weak \( J_y \) and \( J_z \) exchanges allows the system to tunnel quantum mechanically within the degenerate ground state manifold and lifts the degeneracy via an order by quantum disorder effect [41–44]. It is well established that the system develops a supersolid order in a large quantum disorder effect [41–44]. We focus on the O_x surface and generate the phase diagrams and the relevant phases. In the absence of the exchange couplings \( J_y \) and \( J_z \), the Ising exchange interaction \( J_x \) is highly frustrated on the triangular lattice. Any state that satisfies the “2-plus 1-minus” or “2-minus 1-plus” condition for the \( T^x \) configuration on every triangle is the ground state. Therefore, the ground state is extensively degenerate.

To obtain the phase diagram away from the XXZ limit, we implement a self-consistent mean-field theory by assuming a 3-sublattice structure for the mean-field ansatz [38]. Via the mean-field decoupling, we have

\[
H_{MF} = 3 \sum_{r \in A} \sum_{\mu} \left[ J_{x \mu} (m^\mu_B + m^\mu_C) T^r_{x \mu} \right] + 3 \sum_{r \in B} \sum_{\mu} \left[ J_{z \mu} (m^\mu_C + m^\mu_B) T^r_{z \mu} \right]
\]
where \( m_\alpha^r = \langle T_\alpha^r \rangle \) is determined self-consistently for \( r \in \text{A-th sublattice with } \Lambda = \text{A, B, C} \). Such a mean-field theory captures both the uniform state and the 3-sublattice state. The mean-field phase diagram is depicted in Fig. 2. The FD\(_x\) and the FD\(_y\) phases are the previously mentioned ferro-dipolar orders with an uniform \( \langle T^y \rangle \neq 0 \) and \( \langle T^z \rangle \neq 0 \), respectively. There is no octupolar order here. It is the considerable ferro-dipolar interaction in these regions that competes with the antiferro-octupolar interaction and competently suppresses any octupolar order.

In region AFO-FD\(_x\) (AFO-FD\(_y\)) where the transverse exchange \( J_y (J_z) \) is reduced, the octupole moment \( T^x \) orders antiferromagnetically and develops a 3-sublattice structure while the dipole moment \( T^y (T^z) \) remains ferromagnetically ordered (see Fig. 3b). Therefore, the phase is listed as AFO-FD\(_x\) (AFO-FD\(_y\)). In these regions, the weak ferro-dipolar interaction allows the system to fluctuate within the extensively degenerate ground state manifold of the predominant antiferro-octupolar interaction and breaks the degeneracy, leading to the 3-sublattice octupolar order. The background 3-sublattice octupolar order further modulates the ferro-dipolar order and renders the 3-sublattice structure to the ferro-dipolar order. Such a mutual modulation between unfrustrated ferro-dipolar and the frustrated antiferro-octupolar interactions is in fact a quantum effect, and cannot occur in a classical spin system with the same model.

The 3-sublattice structure of the ferro-dipolar order is a direct consequence of the underlying antiferromagnetic octupolar order. This 3-sublattice structure, however, is completely hidden in the magnetization measurement that merely gives a finite net magnetization. To reveal the underlying 3-sublattice structure, one would need local probes such as NMR and \( \mu \)SR. The nuclear spin and muon spin only couple to the dipolar moment, and probe the local dipolar orders of different sublattices. Alternatively, the elastic neutron scattering directly probes the structure of the dipolar orders, and would observe the magnetic Bragg peaks at the \( \Gamma \) point that corresponds to the uniform part of the dipolar order as well as the K points that correspond to the 3-sublattice modulation of the dipolar order. Besides the static properties, the system supports three bands of excitations because of the 3-sublattice structure of the octupolar order. This can be well-observed in an inelastic neutron scattering measurement. We plot the the magnetic excitations in Fig. 3c.

In region AFO-AFD\(_y\) (AFO-AFD\(_x\)), the transverse coupling \( J_y (J_z) \) is antiferromagnetic. The system is therefore frustrated, and due to frustration the 3-sublattice structure persists for rather large \( J_y \) and \( J_z \). Besides the antiferromagnetic order of the octupolar moment \( T^x \), the dipolar moments are also antiferromagnetically ordered (Fig. 3d). The ordering of the local moments is constrained to either \( xy \)- or \( xz \)-plane depending on the magnitude of \( J_y \) and \( J_z \), as in AFO-FD\(_x\) and AFO-AFD\(_x\) phases. The net magnetization of the dipolar moments in AFO-AFD phases is always zero, hence hidden to the thermodynamic measurements; but the 3-sublattice structure can manifest itself in the spin-wave excitations with 3 bands (see Fig. 3d). The gapless modes at \( \Gamma \) in Fig. 3c and d are accidental due to the extended degeneracy in the Ising limit and should be gapped when the magnon interactions are included.

**Discussion.**—It has been realized that a strong SOC could create a significant interaction between the magnetic multipole moments. The magnetic multipolar orders have been proposed in several strong spin-orbit-coupled systems, e.g. the quadrupolar orders and the octupolar orders in ordered double perovskites [8]. The magnetic dipolar orders, being time reversally odd, are often concomitant with the magnetic octupolar orders. Since the former plays a dominant role in many magnetic measurements, it could complicate the interpretation of many experiments and the identification of the underlying octupolar orders. For the DO doublet on the triangular lattice, the lattice symmetry naturally distinguishes the octupole moments from the dipole ones and allows them to have independent structures.

The peculiar property of the DO doublets arises from
the wavefunction, and has little to do with the value of the total moment $J$. Any moment with $J > 1/2$ can potentially support a DO doublet as the CEF ground state doublet. There is no need to restrict $J$ to be odd integer multiples of 3/2. It gives a lot more room for the experimental discovery of DO doublets in the rare-earth triangular lattice materials. The experimental studies of the rare-earth triangular lattice materials have just started. The CEF ground states of most magnetic ions have not been understood. A systematic study of the CEFs will be of great interest. The magnetic properties of many materials in these families are not yet known, and a careful experimental investigation is highly needed.

To summarize, we propose a peculiar Kramers’ doublet, namely, the dipole-octupole doublet, on a triangular lattice. We propose a rather simple model to describe the interaction between the dipole-octupole doublets and predict the hidden magnetic multipolar order and various unexpected properties associated with the multipolar order. In the future, we expect the unprecedented simplicity of the model and the absence of Monte Carlo sign problem will allow a direct comparison between theories, numerics, and experiments on these peculiar doublets.

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I. Dipole-octupole doublet

We consider the general wavefunctions of a DO doublet that are linear superpositions of the $J^z$ states with odd integer multiples of $3/2$,

$$\Psi_+ = \sum_{n_1>0} a_{n_1}|J^z = \frac{3n_1}{2}\rangle + \sum_{n_2<0} a_{n_2}|J^z = \frac{3n_2}{2}\rangle, \quad (7)$$

$$\Psi_- = \sum_{n_1>0} (-)^{n_1+1} a_{n_1}|J^z = -\frac{3n_1}{2}\rangle + \sum_{n_2<0} (-)^{n_2+1} a_{n_2}|J^z = -\frac{3n_2}{2}\rangle, \quad (8)$$

in which $|\Psi_-\rangle$ is simply obtained from $|\Psi_+\rangle$ by a time reversal operation. Here, both $n_1$ and $n_2$ are odd integers by definition, and we assume the wavefunctions have been properly normalized. Using the definition of the effective spin operator in the main text, we can relate the effective spin $\tau^\mu$ with the total moment $J^\mu$ as follows

$$\tau^z \propto PJ^z P, \quad (9)$$

$$\tau^+ \propto P(J^+)^{3n_1} P \quad \text{or} \quad \propto P(J^-)^{3n_1} P, \quad (10)$$

$$\tau^- \propto P(J^-)^{3n_1} P \quad \text{or} \quad \propto P(J^+)^{3n_1} P, \quad (11)$$

where $P = |\Psi_+\rangle\langle\Psi_+| + |\Psi_-\rangle\langle\Psi_-|$ is the projection operator that projects onto the DO doublet manifold. In Eq. (10) and Eq. (11), the lowest order in $J^z$ is $(J^z)^3$. Although the magnetic field couples linearly to $J^\mu$, only $\tau^z$ component survives after we restrict the magnetic field coupling to the DO doublet. The octupole moment $\tau^z$, however, can couple to the magnetic field in the cubic order.
II. Space group symmetry

As we have explained in the main text, we only need to keep the space group symmetry generators of the R3m or P63mmc space group. Within the triangular layer, both R3m and P63mmc space groups give the same list of symmetry generators. As we show in Fig. 4 we have the three-fold rotation, \( C_3 \), the two-fold rotation, \( C_2 \), the inversion, \( I \), and two lattice translations, \( T_x \) and \( T_y \). Under the symmetry operation, the total moment \( J^\mu \) transforms as

\[
\begin{align*}
C_3 & : \quad J^z_r \rightarrow J^z_{C_3(r)}, \quad J^x_r \rightarrow e^{-i \frac{2\pi}{3}} J^x_{C_3(r)}, \quad J^y_r \rightarrow e^{i \frac{2\pi}{3}} J^y_{C_3(r)}, \\
C_2 & : \quad J^z_r \rightarrow -J^z_{C_2(r)}, \quad J^x_r \rightarrow e^{i \frac{2\pi}{3}} J^x_{C_2(r)}, \quad J^y_r \rightarrow e^{-i \frac{2\pi}{3}} J^y_{C_2(r)}, \\
I & : \quad J^x_r \rightarrow J^x_{I(r)}, \quad J^y_r \rightarrow J^y_{I(r)}, \quad J^z_r \rightarrow J^z_{I(r)}, \\
T_x & : \quad J^x_r \rightarrow J^x_{T_x(r)}, \quad J^y_r \rightarrow J^y_{T_x(r)}, \quad J^z_r \rightarrow J^z_{T_x(r)}, \\
T_y & : \quad J^x_r \rightarrow J^x_{T_y(r)}, \quad J^y_r \rightarrow J^y_{T_y(r)}, \quad J^z_r \rightarrow J^z_{T_y(r)},
\end{align*}
\]  

(12)

Using the relations in Eqs. (9)–(11), we obtain the symmetry properties of the pseudospin \( \tau^\mu \).

III. The transformation for the pseudospin

In the transformation that we did to eliminate the crossing coupling between \( \tau^y \) and \( \tau^z \), we choose the \( \theta \) variable such that

\[
\sin 2\theta = \frac{2J_{yz}}{[(J_y - J_z)^2 + (2J_{yz})^2]^{1/2}},
\]

(13)

\[
\cos 2\theta = \frac{J_y - J_z}{[(J_y - J_z)^2 + (2J_{yz})^2]^{1/2}},
\]

(14)

and, the new couplings in the reduced model are given as

\[
J_x = J_x,
\]

(15)

\[
J_y = \frac{1}{2} [J_y + J_z + (J_y - J_z) \cos(2\theta)],
\]

\[
J_z = \frac{1}{2} [J_y + J_z - (J_y - J_z) \cos(2\theta)] + 2J_{yz} \sin(2\theta),
\]

(16)

\[
J_z = \frac{1}{2} [J_y + J_z - (J_y - J_z) \cos(2\theta)] - 2J_{yz} \sin(2\theta).
\]

(17)

IV. Mean field theory in the ferro-octupolar ordered regime

Starting with the model in Eq. (4), we apply mean field decoupling of terms quadratic in \( T^\mu \) by neglecting their fluctuations,

\[
T^\mu T^\nu \rightarrow \langle T^\mu_T^\nu \rangle T^\mu_T^\nu + T^\mu_T^\nu \langle T^\nu_T^\mu \rangle - \langle T^\mu_T^\nu \rangle \langle T^\nu_T^\mu \rangle.
\]

(18)

For the ferromagnetic order, we can assume a site-independent ansatz, and define \( m^\mu \equiv \langle T^\mu_T^\mu \rangle \). This gives us the mean-field Hamiltonian,

\[
H_{MF} = 6 \sum_r [J_x m^x T^x_T^x + J_y m^y T^y_T^y + J_z m^z T^z_T^z]
\]
This Hamiltonian can be diagonalized, and \( m^\mu \) can be solved self-consistently. For dominant \( J_x \), we may further assume \( m^y = m^z = 0 \).

At \( T = 0 \) and \( h = 0 \), it is obvious that \( m^x = 1/2 \). For finite \( T \), the self-consist equation is given by \( m^x = \frac{1}{2} \tanh \left( \frac{3m^x}{T} \right) \). Since \( T^x \) does not couple to \( h \) linearly, an infinitesimal \( h \) would not alter the form of this self-consistent equation. It can be shown that \( m^h \sim \frac{\tanh (3m^x/T)h}{2m^x} \) and \( m^z \sim \frac{\tanh (3m^x/T)h}{2m^x} \), hence a constant \( \chi^{zz} \) below \( T_c \).

V. Mean field theory in the antiferro-octupolar ordered regime

\[
-h \sum_r \left[ \cos \theta T^x_r + \sin \theta T^y_r \right]. \tag{19}
\]

\( T^x \) is consistent with results in the XXZ model. As a result, this Hamiltonian can be diagonalized, and \( m^\mu \) can be solved self-consistently. For dominant \( J_x \), we may further assume \( m^y = m^z = 0 \).

At \( T = 0 \) and \( h = 0 \), it is obvious that \( m^x = 1/2 \). For finite \( T \), the self-consist equation is given by \( m^x = \frac{1}{2} \tanh \left( \frac{3m^x}{T} \right) \). Since \( T^x \) does not couple to \( h \) linearly, an infinitesimal \( h \) would not alter the form of this self-consistent equation. It can be shown that \( m^h \sim \frac{\tanh (3m^x/T)h}{2m^x} \) and \( m^z \sim \frac{\tanh (3m^x/T)h}{2m^x} \), hence a constant \( \chi^{zz} \) below \( T_c \).

To reduce the number of free parameters, we further constraint the magnetization vectors to form patterns depicted in Fig. 5b,c. The Hamiltonian on each sublattice can now be diagonalized separately, and we solve for \( \mathbf{m}_i \) self-consistently. We determine the phase diagram by comparing the mean-field ground state energy between the two possible patterns of orderings and measuring the suppression of \( \langle T^z \rangle \).

VI. Linear spin wave theory

Our mean field theory gives the magnetization vectors for different parameter regimes. Within such phases, there is a stable magnetic ordering, therefore spin wave excitations are well-defined. Using neutron scattering one can measure the spin wave spectrum, as an indirect probe of the ground state.

Suppose the magnetization on site \( i \) is given by \( \mathbf{m}_i \), we introduce the Holstein-Primakoff representation for the pseudospin-\( \frac{1}{2} \) operators,

\[
T_i \cdot \hat{m}_i = \frac{1}{2} - a_i^\dagger a_i, \tag{21}
\]

\[
T_i \cdot \hat{z}_i = \frac{1}{2} (a_i + a_i^\dagger), \tag{22}
\]

\[
T_i \cdot [\hat{m}_i \times \hat{z}_i] = \frac{1}{2i} (a_i - a_i^\dagger), \tag{23}
\]

where \( \hat{m}_i \) is the unit vector parallel to \( \mathbf{m}_i \), and \( \hat{z}_i \) is a unit vector perpendicular to \( \hat{m}_i \). In this representation, the Bloch Hamiltonian has the form

\[
H_{HP} = \sum_{k \in BZ'} \left( A_k^\dagger \begin{pmatrix} F_k & G_k^\dagger \\ G_k & F_{-k} \end{pmatrix} A_k^\dagger \right), \tag{24}
\]

where \( A_k = (a_{1k}, \ldots, a_{nk}) \) is the vector of boson annihilation operators, the subindices \( 1 \ldots n \) label the \( n \) sublattices of the magnetic unit cell, and \( BZ' \) is the magnetic Broullin zone. \( F_k \) and \( G_k \) are \( 2 \times 2 \) matrices and depend on the mean field magnetizations. The Bloch Hamiltonian is diagonalized by the standard Bogoliubov transformation, giving the spectrum of Holstein-Primakoff bosons plotted in Fig. 1 and Fig. 3.