Rare earth Kitaev materials

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We continue the extension of the Kitaev physics and Kitaev interaction to the rare-earth magnets, and study the physical properties of the rare-earth based honeycomb lattice Kitaev materials. We are particularly interested in the experimental consequences of the highly anisotropic spin interaction due to the spin-orbit entanglement. We perform a high-temperature series expansion using a generic nearest-neighbor Hamiltonian with anisotropic interactions, and obtain the heat capacity, the parallel and perpendicular spin susceptibilities, and the magnetic torque coefficients. We further examine the electron spin resonance linewidth as an important signature of the anisotropic spin interactions. Due to the small interaction energy scale of the rare-earth moments, it is experimentally feasible to realize the strong field regime. Therefore, we perform the spin wave analysis and study the possibility of topological magnons when a strong field is applied to the system. The application and relevance to the rare-earth honeycomb Kitaev materials is discussed.

I. INTRODUCTION

The Kitaev honeycomb model\textsuperscript{3} is an exactly solvable model and is defined on a honeycomb lattice with the pairwise and bond dependent spin exchange interactions. It can realize both abelian and non-abelian anyons in different ranges of the parameters with and without the external magnetic field. Apart from its signficance as a theoretical model to illustrate the general properties of topological orders, its possible experimental connection to the honeycomb iridates was later pointed by G. Jackeli and G. Khaliullin\textsuperscript{2}. Their result was based on the spin-orbit-entangled $J_{\text{eff}} = 1/2$ moment of the Ir$^{4+}$ ion where the exchange interaction between the effective moments inherits the orientational dependence of the orbitals. For the nearly 90-degree Ir-O-Ir exchange path\textsuperscript{33}, the principal Heisenberg-like microscopic superexchange interactions become rather small due to the cancellation effect; while the Ir-Ir exchange path gives a Heisenberg interaction\textsuperscript{22} G. Jackeli and G. Khaliullin introduced the subdominant Hund’s coupling in Ref.\textsuperscript{2} and found that the Kitaev interaction can be significant. Other approximations of microscopic calculations bring different and/or additional anisotropic exchange interactions between the local moments\textsuperscript{34}. The essence for the presence of Kitaev interaction is the combination of the spin-orbital entanglement and the three-fold rotational symmetry of the honeycomb lattice. Nevertheless, spin-orbital entanglement only guarantees anisotropic effective spin interaction, does not necessarily prefer a dominant Kitaev interaction. Other competing interactions are present and could drive the system away from the Kitaev spin liquid regime\textsuperscript{24}. Apparently, all the proposed honeycomb Kitaev materials (Na\textsubscript{2}IrO\textsubscript{3}, Li\textsubscript{2}IrO\textsubscript{3}, Li\textsubscript{2}RhO\textsubscript{3} and α-RuCl\textsubscript{3}) develop magnetic orders or spin freezing at low temperatures, even though α-RuCl\textsubscript{3} probably supports a non-magnetic state in a finite magnetic field\textsuperscript{25,26}. It might eventually be possible that some 4$d$/5$d$ honeycomb magnets are located in the parameter regimes with a dominant Kitaev interaction, but there is no conclusive example yet.

Since being 4$d$/5$d$ magnets is not the necessary condition to generate the Kitaev interactions, part of the authors and the collaborators have extended the Kitaev materials to the rare-earth magnets\textsuperscript{16}. The advantages of the rare-earth magnets are the much stronger spin-orbit coupling and much more localized $4f$ orbitals than $4d$/5$d$ magnets\textsuperscript{19}. For the latter ones, one may worry about the further neighbor exchange interactions. Apparently, there exist vast families of rare-earth magnets, and most of them have never been seriously considered along the lines of Kitaev materials. Some of them do form the honeycomb lattice structure\textsuperscript{17}, even though other structures can potentially be interesting for their own rights. Inspired by these thoughts, we study the rare-earth based honeycomb lattice Kitaev material in this paper. We are not analyzing the stability of the Kitaev spin liquid in the phase diagram nor the novel physical properties of Kitaev spin liquid. The later goal is probably more important for the actual confirmation and detection of the Kitaev spin liquid once a relevant physical system is proposed, although spin liquids other than Kitaev spin liquid can also be stabilized\textsuperscript{18}. Instead, our goal here is less mighty. We simply pursue an understanding of the experimental consequence of the spin-orbital entanglement in the rare-earth honeycomb magnets.

For our purpose, we first explore the thermodynamic properties of a rare-earth honeycomb magnet with a generic model for the nearest neighbor interactions. It is known that the anisotropic exchange couplings could appear in the temperature dependence of the thermodynamic quantities such as the specific heat, spin susceptibility and magnetic torque. Especially for the spin susceptibility and magnetic torque, magnetic fields along dif-
ferent directions induce magnetization of different magnitudes, leading to the anisotropic spin susceptibility and the angular dependence of the magnetic torque and providing a natural detection of the intrinsic spin anisotropy in the system. To go beyond the thermodynamic properties, we further consider the electron spin resonance (ESR) measurement of the system. The ESR measurement turns out to be a very sensitive probe of the magnetic anisotropy and is especially useful for the study of the strong spin-orbit-coupled quantum materials, and we compute the ESR linewidth to reveal the intrinsic spin anisotropy of the spin interactions.

Due to the small energy scale of the interaction between the rare-earth local moment, it is ready to apply a small magnetic field in the laboratory to change the magnetic state into a fully polarized one. For such a simple product state, the magnetic excitation can be readily worked out from the linear spin wave theory. We further consider the spin wave spectrum and Berry curvatures of these magnon bands. We find the magnon spectrum supports non-trivial topological band structure. This feature can be manifested in thermal Hall transport measurements.

The remaining of the paper is organized as follows. In Sec. II we introduce the nearest-neighbor spin Hamiltonian, followed by the high-temperature analysis of heat capacity, spin susceptibilities and magnetotropic coefficients in Sec. III. Then we consider the ESR and calculate the influence of anisotropy on the ESR linewidth in Sec. IV. Next the linear spin wave theory of the system is exploited under strong external fields in Sec. V and the aspect of topological magnons is discussed. Finally in Sec. VI we comment on a possible material YbCl₃ and other rare-earth magnets.

II. MODEL

We begin with the following microscopic spin model, that is the most general nearest neighbor Hamiltonian on a honeycomb lattice with the (usual) Kramers doublet effective spin-1/2 local moments,

\[ H = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z + J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \]

\[ \quad + J_{\pm \pm} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij} S_i^- S_j^-) \]

\[ \quad + J_{\pm z} (\gamma_{ij} S_i^z S_j^z + \gamma_{ij} S_i^- S_j^+) + \langle i \leftrightarrow j \rangle \]  

(1)

with \( \gamma_{ij} \) taking \( e^{2i\pi/3} \), \( e^{-2i\pi/3} \), and 1 on the bonds along \( a_1, a_2, a_3 \) directions respectively, as shown in Fig. 1. The spin components are defined in the local coordinate system in Fig. 1. This is possible because the system is planar and has an unique rotational axis. This differs from the rare-earth pyrochlore materials where the spins are often defined in the local coordinate system for each sublattice. This model applies to the rare-earth local moment such as the Yb\(^{3+} \) ion. For non-Kramers doublet like Pr\(^{3+} \) or Tb\(^{3+} \) ion, the \( J_{\pm z} \) term is not allowed by symmetry, and the model becomes further simplified. In fact, a non-Kramers doublet based rare-earth honeycomb magnet arises from the triangular lattice magnet TbInO\(_3\) after 1/3 of the Tb\(^{3+} \) ions becomes inactive magnetically\(^{19} \).

For the rare-earth local moments, the 4f electrons are much localized, and most often, one only needs to consider nearest-neighbor interactions, and occasionally, one would like to include the further neighbor dipole-dipole interactions. In contrast, for the 4d/5d systems, one may need to worry about further neighbor exchange interactions because of the large spatial extension of the electron wavefunctions.

An alternative and often used parametrization of the Hamiltonian is that of the \( J-K-\Gamma-\Gamma' \) model\(^{20} \):

\[ H = \sum_{\langle ij \rangle} J S_i S_j + K S_i^z S_j^z + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \]

\[ \quad + \Gamma' \sum_{\langle ij \rangle} (S_i^z S_j^z + S_i^y S_j^y + S_i^x S_j^x). \]

(2)

where \( \alpha, \beta, \gamma \in \{x, y, z\} \). For instance, \( (xy)z \) specifies the \( z \)-bond. These two expressions are related by the following transformations

\[ J = \frac{4}{3} J_\pm - \frac{\sqrt{2}}{3} J_{\pm z} - \frac{2}{3} J_{\pm \pm} + \frac{1}{3} J_{zz}, \]

\[ K = \sqrt{2} J_{\pm \pm} + 2 J_{\pm z}, \]

\[ \Gamma = -\frac{2}{3} J_\pm - \frac{\sqrt{2}}{3} J_{\pm z} + \frac{4}{3} J_{\pm \pm} + \frac{1}{3} J_{zz}, \]

\[ \Gamma' = -\frac{2}{3} J_\pm + \frac{\sqrt{2}}{6} J_{\pm z} - \frac{2}{3} J_{\pm \pm} + \frac{1}{3} J_{zz}. \]

The Hamiltonian in Eq. (1) can also be used to describe the general exchange interaction between the higher spin local moments for the honeycomb magnets after some modification. The differences are explained in details in the Appendix A.

III. THERMODYNAMICS

The highly anistropic nature of the exchange interaction first impacts the thermodynamic properties of...
and $\chi_O$ compute the parallel and perpendicular spin susceptibilities
Again using high-temperature series expansion, we com-
blets, For the rare-earth local moments with non-Kramers dou-
anisotropic. The Landé factors are different for the in-
the local moment to the external magnetic field is also
\[ C = \frac{3J_0^2}{2k_B T^2} - \frac{27J_0^4}{8k_B^2 T^4}, \tag{4} \]
where we have
\[ J_0^2 = \frac{1}{16} J_{zz}^2 + \frac{1}{2} (J_{\pm}^2 + J_{\pm\pm}^2 + J_{\pm\mp}^2). \tag{5} \]
Due to the spin-orbit entanglement, the coupling of the local moment to the external magnetic field is also anisotropic. The Landé factors are different for the in-
plane and out-plane magnetic fields, and the Zeeman coupling is given as
\[ H_Z = -\mu_0 \mu_B \sum_i \left[ g_\perp (h_x S_i^x + h_y S_i^y) + g_\parallel h_z S_i^z \right]. \tag{6} \]
Again using high-temperature series expansion, we compute the parallel and perpendicular spin susceptibilities up to $O(T^{-3})$
\[ \chi_\parallel = \frac{\mu_0 \mu_B^2 g_\parallel^2}{4k_B T} \left( 1 - \frac{3J_{zz}}{4k_B T} - \frac{J_{\pm\pm}^2}{2k_B^2 T^2} - \frac{J_{\pm\mp}^2}{2k_B^2 T^2} \right), \tag{7} \]
\[ \chi_\perp = \frac{\mu_0 \mu_B^2 g_\parallel^2}{4k_B T} \left( 1 - \frac{3J_{\pm}}{2k_B T} + \frac{5J_{\pm\pm}^2}{4k_B^2 T^2} - \frac{J_{\pm\mp}^2}{k_B^2 T^2} \right), \]
\[ -\frac{3J_{\pm\mp}^2}{4k_B^2 T^2} - \frac{J_{\pm\pm}^2}{16k_B^2 T^2} \right). \tag{7} \]
For the rare-earth local moments with non-Kramers dou-
bles, $g_\perp = 0$ so $\chi_\perp = 0$. In Fig. 2 we plot the magnetic susceptibilities and show the deviation from the simple Curie-Weiss law due to the high order anisotropic terms.
In addition to the simple thermodynamics such as $C_v$
and $\chi$, the magnetic torque measurement is proved to
be quite useful in revealing the magnetic anisotropy. In-
trinsically, there is because the induced magnetization is
generically not parallel to the magnetic field. Thus, when
the sample has an anisotropic magnetization, the system
would experience a torque $\tau = M \times H = -\partial F/\partial \theta$ in an
external magnetic field. The magnetotropic coefficient $k = \partial^2 F/\partial \theta^2$, defined as the second derivative of the free energy to the angle $\theta$ between the sample and the ap-
plicated magnetic field, can be introduced to quantify such
anisotropy. It can be directly measured using the resonant
torsion magnetometry. Under the high-temperature ex-
expansion, we find the magnetotropic coefficient $k$ is
given as
\[ k = \mu_0 \mu_B^2 \frac{G^2}{k_B^2 T^2} \cos 2\theta \left[ \frac{3}{16} (J_{zz} - 2J_\pm) \right. \]
\[ + \left. \frac{1}{64k_B T} (28J_{\pm\pm}^2 - 8J_{\pm\mp}^2 + 4J_{\pm\perp}^2 - 7J_{\pm\mp}^2) \right] \] \tag{8}
where we have defined $G = \sqrt{g_\parallel^2 (h_x^2 + h_y^2) + g_\parallel^2 h_z^2}$. The coefficient $k$ vanishes in the Heisenberg limit: $J_{zz} = 2J_\pm$, $J_{\pm\pm} = J_{\pm\mp} = 0$. More details of the calculation can be found in Appendix B.

IV. ELECTRON SPIN RESONANCE

In the thermodynamic properties, the leading contribu-
tions come from the $J_{zz}$ and $J_\pm$ terms, while the $J_{\pm\pm}$
and $J_{\pm\mp}$ terms are subleading. Arising from spin-orbital
entanglement and completely breaking the U(1) rota-
tional symmetry, these terms play important roles in the
potential quantum spin liquid behavior. To resolve them,
we now turn to the electron spin resonance.

Electron spin resonance measures the absorption of
electromagnetic radiation by a sample subjected to an
external static magnetic field. For a SU(2) invariant sys-
tem, the absorption is completely sharp, i.e. described by
a delta function located exactly at the Zeeman energy. Therefore, the broadening of the resonance spectrum has to arise from the magnetic anisotropy. To understand the contribution of the anisotropy of the nearest-neighbor spin interaction to the ESR linewidth, we decompose the Hamiltonian Eq. (1) into the isotropic Heisenberg part and the anisotropic exchange part
\[ H = J \sum_{(i,j)} S_i \cdot S_j + H', \tag{9} \]
where the Heisenberg coupling $J$ can be found in Eq. (2)
and the anisotropic part
\[ H' = \sum_{(i,j)} S_i^\mu \Gamma_{ij,\mu\nu} S_j^\nu. \tag{10} \]
Here $\Gamma_{ij}$ a traceless and symmetric exchange coupling
matrix. Under the Zeeman term of Eq. (6), the ESR
The linewidth \( \Delta \) for a Lorentzian-shaped spectrum is

\[
\Delta H(\theta) = \frac{\sqrt{2\pi}}{\mu_B g(\theta)} \left( \frac{M_2^2}{M_A} \right)^{1/2},
\]

where \( \theta \) is again the angle between the external field and the sample, and

\[
g(\theta) = \sqrt{g_i^2 \sin^2 \theta + g_i^2 \cos^2 \theta},
\]

\[
M_2 = \frac{\langle [H', M^+] [M^-, H'] \rangle}{\langle M^+ M^- \rangle},
\]

\[
M_4 = \frac{\langle [H, [H', M^+]] [H, [H', M^-]] \rangle}{\langle M^+ M^- \rangle}.
\]

\( M_2 \) and \( M_4 \) are the second and the fourth moments, respectively, and \( M^{\pm} = \sum_i S_i^\pm \). The expectation \( \langle \cdot \cdot \cdot \rangle \) in the above equations is taken with respect to high temperatures. Specifically, we find that

\[
M_2 = \frac{1}{8}(6J_{zz}^2 + 29J_{\pm}^2 + 24J_{\pm\pm}^2 + 55J_{\pm\pm\pm}^2 - 24J_{\pm}J_{zz}),
\]

\[
M_4 = \frac{63}{4}J_{zz}^4 - \frac{249}{2}J_{zz}^2 J_{\pm}^2 + \frac{1129}{32}J_{\pm\pm}^2 J_{\pm\pm\pm}^2 + \frac{63}{16}J_{\pm\pm}^2 J_{\pm\pm\pm}^2 + \frac{1021}{32}J_{\pm\pm}^2 + \frac{21}{4}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}\]

\[
- \frac{1087}{8}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm} - \frac{9}{8}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}\]

\[
- \frac{33}{16}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}^2 - \frac{4091}{16}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm^2} + \frac{3}{16}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}^2 J_{\pm\pm\pm\pm}\]

\[
+ \frac{1849}{32}J_{\pm\pm\pm}\]

\[
+ \frac{1111}{16}J_{\pm\pm\pm}J_{\pm\pm\pm}\]

\[
+ \frac{39}{8}J_{\pm\pm\pm}J_{\pm\pm\pm\pm}\]

\[
+ \frac{2081}{16}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}\]

\[
+ \frac{9}{4}J_{\pm\pm\pm}^2 J_{\pm\pm\pm\pm}\]

\[
+ \frac{51}{4}J_{\pm\pm\pm}^4\]

\[
+ \frac{193}{8}J_{\pm\pm\pm}J_{\pm\pm\pm\pm}\]

\[
+ \frac{3}{8}J_{\pm\pm\pm}^3 J_{\pm\pm\pm\pm} + \frac{8509}{32}J_{\pm\pm\pm}^2.
\]

Our result for ESR linewidths can be compared to the future ESR experiments on the rare-earth based honeycomb magnets. In Fig. 3, we further depict the three-dimensional plots that explicitly demonstrate the dependence of the ESR linewidth on the anisotropic couplings \( J_{zz} \) and \( J_{\pm\pm} \) for three different choices of \( J_{\pm} \).

V. POLARIZED PHASES

A. Strong field normal to the honeycomb plane

To further explore the effect of the anisotropic exchange interaction, we study the spin wave excitation with respect to the polarized states under the strong magnetic fields. This is clearly feasible in the current laboratory setting for the rare-earth magnets as the energy scales for them are usually rather small. For the 4d/5d magnets, there can be difficulty to achieve as the energy scale over there is much higher. Our results here are relevant to the inelastic neutron scattering and thermal Hall transport measurements.

We first consider the case of a strong magnetic field in the direction normal to the honeycomb plane such that the system is in the fully polarized paramagnetic phase and all the spins are aligned along the z direction. We expand about this fully polarized state using the conventional Holstein-Primakoff transformations of the spin variables, which are \( S_i^z = S - a_i^\dagger a_i, S_i^+ = a_i, S_i^- = a_i^\dagger \) for sublattice A, and substitute \( a \rightarrow b \) for sublattice B.

Keeping only the bilateral terms of bosonic operators and taking the Fourier transformation, we arrive at

\[
H = \frac{3N}{4}J_{zz} - 2N\mu_B g(\theta) h_z + \frac{1}{2} \sum_{k} \mathcal{H}_k \mathbf{Y}_k.
\]

with \( \mathbf{Y} = (a_k, b_k, a_k^\dagger, b_k^\dagger)^T \). Here we have denoted

\[
k_1 = -\frac{1}{2}k_x + \frac{\sqrt{3}}{2}k_y, k_2 = -\frac{1}{2}k_x - \frac{\sqrt{3}}{2}k_y, \text{ and } k_3 = k_x
\]

that correspond to the \( y \)-, \( z \)- and \( x \)-directions, respectively. We further define

\[
f(k) = \sum_i e^{i k_i}, \quad g_1(k) = \sum_i e^{-i k_i} \gamma_i, \quad g_2(k) = \sum_i e^{i k_i} \gamma_i \quad \text{and} \quad u = (g_1 \mu_B h_y - 3J_{zz}/2), \]

we then have for \( \mathcal{H}_k \) a block form

\[
\mathcal{H}_k = \begin{bmatrix}
A(k) & B(k) \\
B^\dagger(k) & A^T(-k)
\end{bmatrix},
\]

where we have

\[
A(k) = \begin{bmatrix}
u \\
J_{\pm} f \end{bmatrix},
\]

\[
B(k) = \begin{bmatrix}
0 \\
J_{\pm\pm} g_1
\end{bmatrix}.
\]

All the \( J_{\pm\pm} \) terms are not present.
The spin wave dispersion relation for $\mathcal{H}_k$ follows as

$$\epsilon(k)^2 = u^2 + [f]^2 J_{\pm}^2 \pm \frac{|g_1|^2 + |g_2|^2}{2} J_{\pm}^2$$

$$\pm [4|^2 J_{\pm}^2 + \frac{|g_1|^2 - |g_2|^2}{4} J_{\pm}^4$$

$$+ (f^* g_1^* - f g_2^*)(f^* g_2 - f g_1) J_{\pm}^2 J_{\pm}^2]^{1/2},$$

where only the positive square root of $\epsilon(k)^2$ is taken.

Several simple limits of this expression can be checked: (1) in the Heisenberg limit $J_{\pm} = J$ then $\epsilon(k) = \sqrt{\frac{g_1^2 + g_2^2}{2} h_\parallel - 3 J} \pm |f/J|^{1/2}$; (2) when only $J_{\pm}$ is finite, it reduces to the Ising case $\epsilon(k) = \frac{g_1^2}{2 J} h_\parallel - \frac{J_{\pm}}{2} J_{\pm}$; if only $J_{\pm}$ is present, we have a graphene-like dispersion $\epsilon(k) = \frac{g_1^2}{2 J} h_\parallel \pm |f/J|$. At high fields, the results can be simplified by the Schrieffer-Wolff transformation,

$$\hat{\mathcal{H}}_k = e^{iW} \mathcal{H}_k e^{-iW}$$

$$= \mathcal{H}_k + [W, \mathcal{H}_k] + \frac{1}{2} [W, [W, \mathcal{H}_k]] + \cdots,$$

with the commutator understood as

$$[X, Y] = XY - YX,$$

and $\eta$ is a diagonal matrix with entries $(1, 1, 1, -1)$. Following the treatment of Ref. [27] we choose the transformation to be

$$W = \frac{1}{2u} \begin{pmatrix} 0 & B(k) \\ -B^*(k) & 0 \end{pmatrix},$$

so that up to $O(h_\parallel^{-2})$, we have the $\hat{\mathcal{H}}_k$ to become $A(k) \rightarrow \tilde{A}(k)$, $B(k) \rightarrow \tilde{B}(k)$,

$$\tilde{A}(k) = \left( u - \frac{J_{\pm}^2}{2u} |g_1|^2 \right) f^* J_{\pm} \left( u - \frac{J_{\pm}^2}{2u} |g_2|^2 \right),$$

$$\tilde{B}(k) = -\frac{J_{\pm}^2}{2u} \begin{pmatrix} f^* g_1 + g_2^* f & 0 \\ 0 & f^* g_1 + g_2^* f \end{pmatrix}.$$ 

At high fields, we can thus ignore $\tilde{B}(k)$ and focus on the $\tilde{A}(k)$ term. Writing $\tilde{A}(k) = d_0(k) 1 + \frac{1}{2} d(k) \cdot \sigma$, with the three components being

$$d_1(k) = 2 J_{\pm} Re(f),$$

$$d_2(k) = 2 J_{\pm} Im(f),$$

$$d_3(k) = \frac{J_{\pm}^2}{2u} (|g_1|^2 - |g_2|^2),$$

$$d_0(k) = u - \frac{J_{\pm}^2}{4u} (|g_1|^2 + |g_2|^2).$$

At each momentum $k$ we have the eigenvalues

$$\omega_{\pm}(k) = d_0(k) \pm \frac{1}{2} |d(k)|.$$ 

The above spin wave bands Eq. [29] do not touch unless $J_{\pm} = J_{\pm} = 0$, as we have depicted in Fig. 4. We further compute the Berry curvature as follows

$$F_{xy}^T(k) = \pm \frac{i}{2} \left[ \frac{d(k)}{|d(k)|^2} \times \left( \frac{\partial d(k)}{\partial k_y} \times \frac{\partial d(k)}{\partial k_x} \right) \right].$$

This is negative semi-definite in the Brillouin zone. The Chern numbers follow as

$$C_\pm = \frac{1}{2 \pi i} \int_{BZ} dk_x dk_y F_{xy}^T = \mp 1.$$

This implies the presence of chiral magnon edge states and thermal Hall effect, resulting from the presence of magnon number non-conserving terms $B(k)$ in the Hamiltonian. The edge state for the open boundary condition is depicted in Fig. 5.

**B. Strong field in the honeycomb plane**

We now turn to a strong in-plane field, in the $x$-direction. This is relevant for the rare-earth local moments with the usual Kramers doublet, and does not apply to the non-Kramers doublet. The Holstein-Primakoff transformation for sublattice A is modified as $S^x_i = \frac{1}{2} - a_i^\dagger a_i$, $S^y_i = \frac{1}{2} (a_i + a_i^\dagger)$, $S^z_i = \frac{1}{2} (a_i - a_i^\dagger)$, and that for sublattice B is obtained by substituting $a$ by $b$. These are
again bosonic operators satisfying $[a_i, a_j^\dagger] = [b_i, b_j^\dagger] = \delta_{ij}$. Keeping only the bilinear terms of bosonic operators and taking the Fourier transformation, we obtain

$$H = \frac{3N}{2} J_\pm - 2N\mu_B g_\perp h_x + \frac{1}{2} \Upsilon_k^\dagger \mathcal{H}_k \Upsilon_k, \quad (32)$$

$$\Upsilon = (a_{k}, b_{k}, a_{-k}^\dagger, b_{-k}^\dagger)^T. \quad (33)$$

Define $g_3(k) = e^{ik_1} + e^{ik_2} - 2e^{ik_3}$, $g_4(k) = e^{ik_1} - e^{ik_2}$ and recall $f(k) = \sum_\epsilon e^{\epsilon k}$. The $\mathcal{H}_k$ is of the familiar form

$$\mathcal{H}_k = \begin{bmatrix} A(k) & B(k) \\ B^\dagger(k) & A^T(-k) \end{bmatrix},$$

but now with the $A, B$ matrices given by

$$A(k)_{11} = A(k)_{22} = v = \mu_B g_\perp h_x - 3J_\pm, \quad (34)$$

$$A(k)_{21} = A(-k)_{12} = \left(\frac{1}{4} J_{zz} + \frac{1}{2} J_\pm \right) f + \frac{3}{4} J_{\pm \pm} \quad (35)$$

$$B(k)_{11} = B(k)_{22} = 0, \quad (36)$$

$$B(k)_{21} = B(-k)_{12} = \left(\frac{1}{4} J_{zz} + \frac{1}{2} J_\pm \right) f + \frac{1}{4} J_{\pm \pm} + i\frac{\sqrt{3}}{2} J_{zz}g_4. \quad (37)$$

Appealing again to the Schrieffer-Wolff transformation with

$$W = \frac{1}{2v} \begin{bmatrix} 0 & B(k) \\ -B^\dagger(k) & 0 \end{bmatrix}, \quad (38)$$

then up to $O(h_\perp^2)$, we have the effective $\tilde{\mathcal{H}}_k$ to be

$$\tilde{A}(k) = \begin{bmatrix} v - \frac{1}{2v^2} |B(k)_{12}|^2 & \frac{1}{4} A(k)_{12} \\ \frac{1}{4} A(k)_{21} & v - \frac{1}{2v^2} |B(k)_{21}|^2 \end{bmatrix}, \quad (39)$$

$$\tilde{B}(k) = -\frac{1}{2v} [A(k)_{21} B(k)_{12} + A(k)_{12} B(k)_{21}]. \quad (40)$$

At high fields $h_\perp$, we can ignore $\tilde{B}(k)$ and focus on the $\tilde{A}(k)$ term. Rewrite $\tilde{A}(k) = d_0(k)\mathbb{1} + \frac{1}{2} \mathbf{d}(k) \cdot \mathbf{s}$, with each component being

$$d_1 = \left(\frac{1}{4} J_{zz} + \frac{1}{2} J_\pm \right) f + \frac{1}{4} J_{\pm \pm} \quad (41)$$

$$d_2 = \left(\frac{1}{4} J_{zz} + \frac{1}{2} J_\pm \right) f + \frac{1}{4} J_{\pm \pm} \quad (42)$$

$$d_3 = -\frac{1}{2v} \left[\frac{1}{2} \sqrt{3} g_4 J_{zz} \left[\frac{1}{4} J_{zz} + \frac{1}{2} J_\pm \right] f + \frac{1}{4} J_{\pm \pm} g_4 \right] + c.c. \quad (43)$$

$$d_0 = v - \frac{1}{2v} \left[\frac{1}{4} J_{zz} + J_{\pm \pm} \right]^2 |f|^2 + \frac{3}{4} J_{\pm \pm} g_4^2. \quad (44)$$

We then arrive at the dispersions $\omega \pm(k) = d_0(k) \pm \frac{1}{2} |\mathbf{d}(k)|$. The spectrum is plotted in Fig. 5. We find both bands have zero Chern numbers, and we have checked for many other parameter choices and also obtained trivial zero Chern number. Thus, the in-plane field magnon band structure is quite distinct from the topological magnon band structure for the normal-plane field case.

**VI. DISCUSSION**

The rare-earth based honeycomb Kitaev materials have not been carefully explored yet. These systems can be equally interesting as the 4$d$/5$d$ magnets with the $J_{\text{eff}} = 1/2$ local moments, and the anisotropic spin interaction can be more significant than the 4$d$/5$d$ magnets due to the much stronger spin-orbit couplings in these materials. In our work here, we have carefully studied the experimental consequences of the spin-orbit entanglement and the anisotropic spin exchange interactions. These results can be directly compared with the experiments on the rare-earth honeycomb magnets and provide a useful guidance for the future experiments. One potential candidate material in the rare-earth honeycomb magnets is YbCl$_3$ which has a similar crystal structure to that of RuCl$_3$. The Yb$^{3+}$ ions have nearly filled $f$-orbitals, which, combined with the large crystal fields lead to very simple ground state manifold. Furthermore, its edge-shared octahedral structure gives simple exchange physics that is relatively well-understood. However, there is limited information about this material in the literature.

Finally, we emphasize that most rare-earth magnets have not been discussed along the line of Kitaev interactions. In this paper, as one of the first few works on this topic [16,17,30], we merely focus the analysis on the honeycomb lattice rare-earth magnets. In the previous work [15], we illustrate our observation with the FCC rare-earth magnets. As many non-honeycomb lattice iridates are claimed to be Kitaev materials, it is thus reasonable to consider the rare-earth magnets with other structures to be candidate Kitaev materials [31,32]. In a more serious manner, one actually needs to have a local three-fold rotation to permute the three components of the effective spin such that a Kitaev interaction can be well present.

To summarize, we have focused on rare-earth Kitaev materials with nearest-neighbor interactions and computed the high-temperature thermodynamic properties, ESR linewidth, and spin-wave behaviors as the experimental consequences of the anisotropic spin interaction.
Due to the spin-orbital entanglement and the spin-lattice combined back to a spin-3/2 local moments, the spin-1/2 states and three onsite spin-1/2 spins nearest neighbor bonds are covered with spin singles of the ALKT states on the honeycomb lattice where the spin-1/2 local moments, the pairwise spin interaction is given as

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

$$+ J_{\pm \pm} (\gamma_{ij} S_i^+ S_j^z + \gamma_{ij} S_i^z S_j^z)$$

$$+ J_{\pm \mp} (\gamma_{ij} S_i^z S_j^+ + \gamma_{ij} S_i^+ S_j^-) + (i \leftrightarrow j)$$

$$+ \sum_i D(S_i^z)^2.$$ 

Because of the larger Hilbert space, a single-ion anisotropy is allowed and new states such as the quantum paramagnet can be favored here. Thus, the phase transitions between quantum paramagnet and other ordered phases can be interesting. Further neighbor exchange interaction, if included, could bring more frustration channel than the spin-orbit entanglement induced frustration. It is known that, simple $J_1$-$J_2$ (first neighbor and second neighbor Heisenberg) model on honeycomb lattice could induce spiral spin liquids in two dimensions where the spiral degeneracy has a line degeneracy in the momentum space rather than the surface degeneracy. The presence of the anisotropic interaction in Eq. (A1) would overcome the quantum/classical order by disorder effect and lift the degeneracy. In addition to the spin-1 local moments, the model in Eq. (A1) also applies to the spin-3/2 systems. Since the honeycomb lattice contains three nearest neighbor bonds, one may consider the possibility of the ALKT states on the honeycomb lattice where the three nearest neighbor bonds are covered with spin singles of the spin-1/2 states and three onsite spin-1/2 spins are combined back to a spin-3/2 local moments.

Here, we have only listed the pairwise spin interactions. Due to the spin-orbital entanglement and the spin-lattice coupling, the effective interaction for the spin-1 and spin-3/2 magnets can contain significant multipolar interactions. A simple example would be the biquadratic exchange $-(S_i \cdot S_j)^2$ that is induced effectively by the spin-lattice coupling. The presence of these multipolar interactions can significantly enhance quantum fluctuation by allowing the system to tunnel more effectively within the local spin Hilbert space and thus create more quantum states such as multipolar ordered phases and quantum spin liquid.\cite{331041}

The relevant physical systems for the spin-1 and spin-3/2 moments would contain the $4d^2, 5d^2, 4d^4, 5d^4$ and $4d^1, 5d^1, 4d^3, 5d^3$ magnetic ions, respectively. The relevant ions can arise from Ru, Mo and even V atoms, where spin-orbit coupling in the partially filled $t_{2g}$ shell is active.\cite{331041}

Appendix A: Generic spin models and candidate states for higher spins

This spin model in Eq. (1) is designed for effective spin-1/2 local moments. It can be well extended to the high spin local moments. For the honeycomb lattice with spin-1 local moments, the pairwise spin interaction is given as

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

$$+ J_{\pm \pm} (\gamma_{ij} S_i^+ S_j^z + \gamma_{ij} S_i^z S_j^z)$$

$$+ J_{\pm \mp} (\gamma_{ij} S_i^z S_j^+ + \gamma_{ij} S_i^+ S_j^-) + (i \leftrightarrow j)$$

$$+ \sum_i D(S_i^z)^2.$$ 

Appendix B: Details of high temperature expansion

The high temperature expansion requires to take into account the commutation relations between different spin operators on a same site. To this end, we define a vertex function $\nu(n_x, n_y, n_z)$ at each site $i$ following Ref.\cite{35} where $n_x, n_y$ and $n_z$ have to be even integers for the function to be nonzero. Its explicit form can be calculated by introducing the generating function

$$\psi(\xi, \eta, \zeta) = \text{Tr} \left[ \exp(\xi S^x + \eta S^y + \zeta S^z) \right].$$

Expanding the exponential and using the definition of $\nu$, we have

$$\psi(\xi, \eta, \zeta) = 2 \sum_{n_x=0}^{\infty} \sum_{n_y=0}^{\infty} \sum_{n_z=0}^{\infty} \nu(n_x, n_y, n_z) \xi^{n_x} \eta^{n_y} \zeta^{n_z} n_x! n_y! n_z!.$$ 

On the other hand, by diagonalizing the matrix of the exponential, we have

$$\psi(\xi, \eta, \zeta) = 2 \cosh \left( \sqrt{\xi^2 + \eta^2 + \zeta^2/2} \right).$$

Expanding this and comparing with the previous equation,

$$\nu(n_x, n_y, n_z) = \frac{[(n_x + n_y + n_z)/2]!}{(n_x/2)! (n_y/2)! (n_z/2)!} \frac{1}{(n_x + n_y + n_z)!} n_x! n_y! n_z!.$$ 

We note this function is symmetric under the permutation of $n_x, n_y, n_z$.\footnote{Alexei Kitaev. Anyons in an exactly solved model and}

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