Electron quasi-itinerancy intertwined with quantum order by disorder in pyrochlore iridate magnetism

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We point out the emergence of magnetism from the interplay of electron quasi-itinerancy and quantum order by disorder in pyrochlore iridates. Like other Mott insulating iridates, the Ir$^{4+}$ ion in pyrochlore iridates develops an effective $J = 1/2$ moment from the on-site spin-orbit coupling. We consider the generic symmetry-allowed exchange between these local moments on a pyrochlore lattice and obtain the mean-field phase diagram. Assuming the superexchange is mediated by direct and/or indirect electron hopping via intermediate oxygens, we derive the exchange interactions in the strong coupling regime from the Hubbard model. This exchange has a degenerate classical ground state manifold and quantum fluctuation selects a non-coplanar ground state, known as quantum order by disorder. Extending to the intermediate coupling regime, the same non-coplanar order is selected from the degenerate manifold by the kinetic energy, which is dubbed “electron quasi-itinerancy”. We discuss the experimental relevance of our results and electron quasi-itinerancy among other iridates and 4$d$/5$d$ magnets.

I. INTRODUCTION

In recent years, there have been a lot of activities on the Ir-based transition metal oxides. Due to the strong spin-orbit coupling (SOC) of its 5$d$ electrons, many novel phases, theoretical models and experiments have been proposed and discovered in these Ir-based materials. Among them, for example, a quantum spin liquid phase was proposed for an Ir-based hyperkagomé lattice in Na$_4$Ir$_3$O$_8$. A ferromagnetic ground state with a large ferromagnetic moment was discovered in Sr$_2$IrO$_4$ with the Ir$^{4+}$ ions forming a square lattice. In these Mott insulating systems, the presence of strong SOC drastically changes the local spin physics. The local moment of the magnetic ion Ir$^{4+}$ is an effective $J = 1/2$ moment describing local spin-orbital doublets rather than usual electron spin $S = 1/2$ for systems with a weak SOC. The existence of local spin-orbital doublets has been detected by resonant X-ray scattering experiment in Sr$_2$IrO$_4$. As a consequence, the non-trivial exchange interaction can arise due to the mixing of spin and orbitals.

Even though the superexchange interaction between the Ir local moments is often used to describe most iridates, most well-known Mott insulating iridates are actually weak Mott insulators with quasi-itinerant 5$d$ electrons and small charge gaps. This weak Mott insulating nature was not really emphasized in the literature, and we think this may be important in understanding some of the physical properties of iridates and the related materials. What is electron quasi-itinerancy? Quasi-itinerancy is the key property of the electrons in the weak Mott regime where the Mott gap is not large enough to fully localize the electron to one single lattice site, and the electron can still be delocalized to a finite extent spatially due to the small charge gap. Electron quasi-itinerancy is believed to be the driving force for the possible spin liquid phase in the weak Mott regime for $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$. Over there, the electron quasi-itinerancy generates the frustrated ring exchange interactions that suppresses the magnetic orders. Similar kind of electron quasi-itinerancy that emphasizes different outcomes of the charge fluctuations has been discussed in various spinels and osmate pyrochlores. Thus, besides the prevailing strong coupling perspectives, the weak to intermediate coupling perspective is found to be both complementary and exciting. Ref. applied a slave-rotor mean field theory to study the Mott transition in a series of rare-earth based pyrochlore iridates R$_2$Ir$_2$O$_7$. They discovered topological band insulator in the non-interacting limit and a novel topological Mott insulator in intermediate coupling regime. Several other groups re-examined the problem with more realistic Hamiltonian and discovered various magnetic ordered phases and an interesting Weyl semi-metal phase that is located in the narrow regime separating the topological band insulator or metal phase from strong coupling Mott insulating phase. Aligned with the above theoretical efforts, the experiments discovered that a metal-insulator transition in R$_2$Ir$_2$O$_7$ (R = Nd, Sm, Y and Eu) involves a magnetic ordering produced by the 5$d$ electrons in Ir. Moreover, an exotic spin liquid metallic phase was also proposed experimentally for Pr$_2$Ir$_2$O$_7$. Now, the Ir
electrons are proposed as Luttinger semimetal$^{60,19}$ while the Pr spin is proposed to be proximate to a transition between a U(1) spin liquid and an ordered spin ice$^{19,52}$.

Based on the existing theoretical$^{19,51,53,54,55}$ and experimental works, the true magnetic state of these Ir-based pyrochlore systems remains open. In the present paper, we address this problem and provide some understanding. We primarily focus on the magnetic properties and avoid touching the band structure topology that has been invoked in early works. We first explore the magnetic properties of the Ir-based pyrochlore lattice in the strong coupling regime. Physically, since the 5$d$ electron orbitals of the Ir$^{4+}$ is spatially extended, which enhances the electron bandwidth, therefore these Ir-based systems are usually considered to be in the intermediate coupling regime. Nevertheless, the SOC could enhance the correlation by suppressing the bandwidth.$^{16}$ Moreover, certain magnetic properties in the strong coupling limit could persist to the intermediate coupling regime even if the system is located in the intermediate coupling regime. In the strong coupling limit, the effective moments $J = 1/2$ of the Ir$^{4+}$ ions are coupled by the superexchange interaction. We analyze the symmetry-allowed exchange Hamiltonian, that includes three types of pairwise terms, Heisenberg exchange, antisymmetric Dzyaloshinskii-Moriya (DM) interaction and the symmetric pseudo-dipolar (PD) interaction. This model is equivalent to the one that was used for interacting Kramers doublet for the rare-earth pyrochlores. In the mean-field phase diagram, we find five different ordered phases (see Sec. II): 4-in-4-out state, a continuously degenerate state spanned by two basis vectors $(v_1, v_2)$, a weakly ferromagnetic state (FM) and two coplanar states with spin orient along the particular [110] directions. Almost all these ordered states have the magnetic wavevector $q = \mathbf{0}$. For the realistic exchange model obtained from an extended Hubbard model relevant for R$_2$Ir$_2$O$_7$, there are only two ordered phases, that are the “4-in-4-out” state and the continuously degenerate manifold spanned by two basis vectors $(v_1, v_2)$. For the latter, we find that the quantum fluctuation selects a non-coplanar spin configuration by a linear spin-wave expansion. This is the mechanism of quantum order by disorder. For the intermediate coupling regime, we apply the self-consistent mean-field theory to study the microscopic Hubbard model and assume a general magnetic configuration except having the same magnetic cell as the crystallographic cell (or $q = \mathbf{0}$) order. Again, we find the system is “fluctuating” within the continuously degenerate manifold spanned by $(v_1, v_2)$, and the electron kinetic energy selects the magnetic orders. The electron kinetic energy is important here due to the quasi-itinerancy in the weak Mott regime. It is found that the magnetic orders in the strong coupling regime persist into the intermediate coupling regime. Since it is unclear which regime the actual system is proximate to, it is reasonable to think the electron quasi-itinerancy is intertwined with the quantum order by disorder here.

In the following, we outline the main content of this paper. In Sec. II we study a generic symmetry-allowed exchange Hamiltonian on the pyrochlore lattice with the effective spin-1/2 originating from Kramers’ degeneracy, that is relevant for R$_2$Ir$_2$O$_7$ in the strong coupling regime. In the exchange Hamiltonian, there are four symmetry allowed coupling parameters, Heisenberg exchange $J_0$, DM interaction $D$, and $\Gamma_1, \Gamma_2$ for PD interaction. We analyze this Hamiltonian with the mean-field method in different parameter regimes. In many parts of phase diagram, the ground state can be understood as simultaneously optimizing different terms of the Hamiltonian. In Sec. III we derive a realistic exchange from the extended Hubbard model. Two limits with the dominant direct or indirect electron tunneling via intermediate oxygens are considered. In these two cases, we find there is only one mean-field phase, which is the continuously degenerate manifold $(v_1, v_2)$. We then implement the linear spin-wave theory and a non-coplanar ground state is favored by this quantum order by disorder mechanism. For the certain intermediate regime with comparable direct and indirect electron tunnelings, the “4-in-4-out” state is favored. We further explore the magnetic properties of the Hubbard model in the intermediate coupling regime. By assuming a $q = \mathbf{0}$ magnetic structure, we implement a Hartree-Fock type of self-consistent mean-field theory for the interaction. Finally in Sec. IV we discuss the relevant experiments and other related works.

II. THE GENERIC EXCHANGE HAMILTONIAN

In this section, we analyze the Ir-based pyrochlore lattice in strong coupling regime. In the strong coupling limit, the local effective spin moments are coupled by an exchange Hamiltonian. For the effective spin-1/2 moment describing the local Kramers’ doublets, the exchange interaction is guaranteed to be pairwise. The generic exchange Hamiltonian has the following form

$$H_{\text{ex}} = \sum_{\langle ij \rangle} J_0 (\mathbf{J}_i \cdot \mathbf{J}_j) + D_{ij} (\mathbf{J}_i \times \mathbf{J}_j) + \Gamma_{ij}^{\mu\nu} J_{\mu i}^* J_{\nu j}, \quad (1)$$

where the nearest-neighbor interaction is assumed, and $J_0$ is the isotropic Heisenberg exchange, $D_{ij}$ describes the antisymmetric Dzyaloshinskii-Moriya (DM) interaction and $\Gamma_{ij}^{\mu\nu}$ is the symmetric pseudo-dipolar (PD) interaction. This form of decomposition is well-known to the much older literature of magnetism$^{59}$ but is not quite popular among newer ones. Kitaev or any other anisotropic exchange interactions can be well cast into this form, as long as they are pairwise interactions. As a general rule of thumb, for systems with a weak SOC, the DM interaction is weaker than to the Heisenberg part, and PD interaction is even weaker than DM interaction. For systems with strong SOC such as iridates here, there is no general thumb of rule, and all the interactions could be of similar magnitudes. Thus, for most
magnetic systems composed of 3d transition metal ions, the DM interaction and PD interaction are expected to be much weaker than the Heisenberg exchange and hence can be neglected at lowest order approximation. For Ir-based magnets or other magnetic systems formed by 4d/5d transition metal ions, SOC is quite strong and local moment is a mixture of spin and orbitals. As a result, the exchange interaction is usually very non-Heisenberg-like and the anisotropic exchanges (such as DM and PD interactions) can be quite significant.

Throughout this section, we assume an antiferromagnetic Heisenberg part with $J_0 > 0$. Since most $\text{R}_2\text{Ir}_2\text{O}_7$ (and also spinel $\text{AB}_2\text{X}_4$) compounds have a space group $\text{Fd}_{3} \text{~m}$, this space group symmetry further restricts the allowed forms of the DM interaction and PD interaction. Therefore, for the bond connecting the sublattice 0 with the sublattice 1 (see Fig. 1), we have,

$$D_{01} = D(0, \frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}),$$

$$\Gamma_{01} = \begin{bmatrix} -2\Gamma_1 & 0 & 0 \\ 0 & \Gamma_1 & -\Gamma_2 \\ 0 & -\Gamma_2 & \Gamma_1 \end{bmatrix},$$

where the matrix $\Gamma_{01}$ is demanded to be symmetric and traceless as the traceful part of the full interaction is taken care of by the Heisenberg interaction, and antisymmetric one is from the DM interaction. Exchange interactions on other bonds can be simply generated by cubic permutations.

Although the exchange Hamiltonian in Eq. (4) is introduced for Ir-based pyrochlore lattice, it is widely applicable to other pyrochlore systems with the same symmetry properties as long as the local moment is a Kramer spin-1/2 doublet. Our results would also apply to these contexts as well. In fact, this model is equivalent to the one that was used for the rare-earth pyrochlore material $\text{Yb}_2\text{Ti}_2\text{O}_7$ where some detailed analysis were given in Ref. [61] and [62]. Over there, a local coordinate system was used for each pyrochlore sublattice and the local moment is the Kramers doublet of the $\text{Yb}^{3+}$ ion, while here we are using a global cubic coordinate for the $\text{Ir}^{4+}$ effective spin-1/2 moments. In the next subsection, we analyze the mean-field ground states of this general Hamiltonian and understand the role of different anisotropic interactions.

### A. Role of Dzyaloshinskii-Moriya interaction

Here we consider the role of Dzyaloshinskii-Moriya interaction on top of the Heisenberg interaction and set $\Gamma_1 = \Gamma_2 = 0$. Classically, it is well-known that the pyrochlore lattice is the most frustrated lattice by having a macroscopic number of ground state degeneracies with the nearest-neighbor Heisenberg model. The presence of the anisotropic exchange surely lifts the classical ground state degeneracy. Ref. [63] has already studied the role of DM interaction by mean-field theory and classical Monte-Carlo simulation. Our mean-field analysis below by treating the effective spin $\mathbf{J}$ as a classical vector is consistent with their results. With a direct DM interaction that corresponds to $D < 0$ in the present work, the ground state is 2-fold degenerate (related by time reversal) with the magnetic ordering wavevector $\mathbf{q} = 0$. The magnetic unit cell coincides with the crystallographic one and the four spins on the unit cell are

$$\Psi \equiv (\mathbf{J}_0, \mathbf{J}_1, \mathbf{J}_2, \mathbf{J}_3) = \frac{1}{\sqrt{3}}(1\bar{1}1, 1\bar{1}\bar{1}, \bar{1}1\bar{1}, \bar{1}1\bar{1}).$$

Here we define a vector $\Psi$ for the four spin vectors on the elementary tetrahedron. This is the simple 4-in-4-out state.

For the indirect DM interaction with $D > 0$, DM interaction only partially lifts the ground state degeneracy. There are two sets of ground states, coplanar and non-coplanar states, both of which have a magnetic wavevector $\mathbf{q} = 0$. The 4-spin vector $\Psi$ of the coplanar ground states can be constructed as linear superpositions of the following two basis vectors $\mathbf{u}_1$ and $\mathbf{u}_2$ (or their equivalence under discrete symmetry operations)

$$\mathbf{u}_1 = (100, 010, 010, \bar{1}0\bar{1}),$$

$$\mathbf{u}_2 = (010, \bar{1}0\bar{1}, 100, 0\bar{1}0).$$

The non-coplanar states are constructed from the following two basis vectors $\mathbf{v}_1$ and $\mathbf{v}_2$ (or their symmetry equivalence)

$$\mathbf{v}_1 = \frac{1}{\sqrt{2}}(\bar{1}10, \bar{1}1\bar{1}, 1\bar{1}0, 1\bar{1}\bar{1}),$$

$$\mathbf{v}_2 = \frac{1}{\sqrt{6}}(\bar{1}\bar{1}2, \bar{1}\bar{1}2, 1\bar{1}\bar{2}, 1\bar{1}\bar{2}).$$
FIG. 2. (Color online.) The spin configuration on each sublattice for different phases.

Here, when only the first basis vector $v_1$ is chosen, the ground state is a special coplanar state with spin oriented along different [110] lattice directions. Both the coplanar and non-coplanar degenerate ground state manifolds have an accidental U(1) degeneracy with one continuous degree of freedom. This degenerate spin manifold is actually identical to the one that was proposed for the rare-earth pyrochlore $\text{Er}_2\text{Ti}_2\text{O}_7$, and are selected via the quantum order by disorder mechanism.\(^{23}\)

**B. Role of pseudo-dipolar interaction: case 1**

In this and next subsections, we study the role of the PD interaction. We first consider the regime with $D = 0$, $\Gamma_1 \neq 0, \Gamma_2 = 0$. For $\Gamma_1 > 0$, we find that optimal spin configurations have magnetic wavevector $q = 0$. Even though the Hamiltonian breaks the spin rotation symmetry completely, the ground state manifold has an accidental $O(3)$ degeneracy. The 4-spin vector $\Psi$ of the ground states is an arbitrary linear superposition of the following three basis vectors $w_1, w_2$ and $w_3$,

$$w_1 = (100, 100, 010, 010), \quad (9)$$
$$w_2 = (010, 010, 010, 010), \quad (10)$$
$$w_3 = (001, 001, 001, 001). \quad (11)$$

For $\Gamma_1 < 0$, to simultaneously optimize the energy and satisfy the hard spin constraint, there only exist two sets of ground states. One has the magnetic wavevector $q = 0$. Similar to the case with $\Gamma_1 > 0$, the ground state spin configuration has an accidental $O(3)$ degeneracy. The 4-spin vector $\Psi$ is constructed from the following three basis vectors $z_1, z_2$ and $z_3$ (or their symmetry equivalence),

$$z_1 = (100, 100, 100, 100), \quad (12)$$
$$z_2 = (010, 010, 010, 010), \quad (13)$$
$$z_3 = (001, 001, 001, 001). \quad (14)$$

The other set of ground states has the magnetic wavevector $q = 2\pi(100)$ or its cubic equivalences. Although the magnetic unit cell doubles the size of crystallographic cell, the spin configuration can still be fully described within one tetrahedron and the 4-spin vector $\Psi$ is given as

$$\Psi = (100, 100, 100, 100), \quad (15)$$

and the spin configuration of other sites is generated from this and the ordering wavevector.

**C. Role of pseudo-dipolar interaction: case 2**

Here we consider the parameter regime with $D = 0$, $\Gamma_1 = 0, \Gamma_2 \neq 0$. For $\Gamma_2 < 0$, the ground state is the same as the case for $D < 0$, which is the 4-in-4-out state. For $\Gamma_2 > 0$, the anisotropy does not lift the classical degeneracy of the nearest-neighbor Heisenberg model on pyrochlore lattice.

**D. With both Dzyaloshinskii-Moriya and pseudo-dipolar interactions**

In this subsection, we study the classical phase diagram when both two of the anisotropic exchanges are present. We start from the $D-\Gamma_1$ plane with $\Gamma_2 = 0$. The phase diagram is depicted in Fig. 2. In all the parts of the phase diagram, the magnetic wavevector is $q = 0$. Most parts of the phase diagram can be understood as the intersection of two different ground state manifolds separately favored by $D$ and $\Gamma_1$, which have already been discussed in detail in the previous subsections.

For $D < 0, \Gamma_1 > 0$, the 4-in-4-out state is favored. For $D > 0, \Gamma_1 > 0$, we have the classical ground states con-
structured as the linear superpositions of the same two basis vectors $v_1$ and $v_2$ that are introduced in Eq. (7) and (8) for the case of $D > 0$. For $D > 0, \Gamma_1 < 0$, the ground state is a coplanar state with the spins pointing along different [110] directions (denoted as “coplanar-[110]” in Fig. 3), whose 4-spin vectors $\Psi$ can be constructed from the basis vectors $u_1$ and $u_2$ in Eq. (5) and Eq. (6),

$$\Psi = \frac{1}{\sqrt{2}} (110, 110, 110, 110).$$

(16)

For $D < 0, \Gamma_1 < 0$, the $D$-demanded and $\Gamma_1$-demanded ground state manifolds have no overlap. We find that, when $D < 3\sqrt{2}\Gamma_1$, DM interaction has a more weight in the Hamiltonian and the ground state is the 4-in-4-out state, and in the opposite case, the ground state is a coplanar state (denoted as “coplanar-[110]” in Fig. 3) whose 4-spin vector is given

$$\Psi = \frac{1}{\sqrt{2}} (1\bar{1}0, 1\bar{1}0, 1\bar{1}0, 1\bar{1}0).$$

(17)

Note this coplanar state is distinct from the “coplanar-[110]” state found for $D > 0, \Gamma_1 < 0$.

Now we discuss the ground states in $D-\Gamma_2$ plane with $\Gamma_1 = 0$. The phase diagram is depicted in Fig. 4. The magnetic wavevector is $q = 0$ everywhere in the phase diagram.

For $D < 0, \Gamma_2 < 0$, the ground state is simply the 4-in-4-out state. For $D > 0, \Gamma_2 > 0$, the ground state is an arbitrary linear superposition of the basis vectors $v_1$ and $v_2$ in Eq. (7) and Eq. (8). In the regime of $D > 0, \Gamma_2 < 0$, there exist two phases. When $D > D_{c1}(\Gamma_2)$ with

$$D_{c1}(\Gamma_2) = \frac{\sqrt{2}}{6} (3J_0 - 2\Gamma_2 - \sqrt{9J_0^2 - 6J_0\Gamma_2 + 4\Gamma_2^2}),$$

(18)

the ground state turns out to be weakly ferromagnetic and denoted as “weak FM” in Fig. 4. The 4-spin vectors of the magnetic unit cell are parameterized as

$$\Psi = \cos \theta y_1 + \sin \theta y_2$$

(19)

with

$$y_1 = \frac{1}{\sqrt{2}} (1\bar{1}0, 1\bar{1}0, 1\bar{1}0, 1\bar{1}0),$$

(20)

$$y_2 = (001, 001, 001, 001),$$

(21)

and the angular variable $\theta$ satisfies

$$\cos 2\theta = \frac{4J_0 + \sqrt{2}D - \Gamma_2}{\sqrt{(4J_0 + \sqrt{2}D - \Gamma_2)^2 + 8\Gamma_2^2}},$$

(22)

$$\sin 2\theta = \frac{-2\sqrt{2}\Gamma_2}{\sqrt{(4J_0 + \sqrt{2}D - \Gamma_2)^2 + 8\Gamma_2^2}}.$$  

(23)

When $D < D_{c1}(\Gamma_2)$, the ground state is the 4-in-4-out state.

In the region of $D < 0, \Gamma_2 > 0$, there also exist two phases. When $D < D_{c2}(\Gamma_2)$ with $D_{c2}(\Gamma_2)$ given by

$$D_{c2} = \frac{-3\sqrt{2}\Gamma_2}{2}.$$  

(24)

When the DM interaction is dominant and negative, the ground state is the 4-in-4-out state. When $D > D_{c2}(\Gamma_2)$, a coplanar state with spins pointing along various [110] directions is favored and the 4-spin vector $\Psi$ is the same as the one introduced in Eq. (17) and its symmetry equivalence. Hence, we also denote this coplanar state as “coplanar-[110]” in Fig. 3.

III. MAGNETISM FROM ELECTRON QUASI-ITINERANCY

Having understood the role of each anisotropic exchange for the generic exchange Hamiltonian in the previous section, in this section we discuss the physical exchange Hamiltonian derived perturbatively from the microscopic parent Hubbard model and from there approach the magnetic states in the intermediate coupling regime. We analyze the possible magnetic ground states for the compound $R_2Ir_2O_7$.

A. Hubbard model and exchange

We assume the on-site SOC is strong enough so that the lower $J = 3/2$ bands are completely filled and the

FIG. 5. (Color online.) The direct electron tunneling between the Ir atoms. Left: the $\sigma$-bonding with tunneling amplitude $t_1$. Right: the $\pi$-bonding with tunneling amplitude $t_2$. 
upper \( J = 1/2 \) bands are half filled. This approximation misses the hybridization between the \( J = 1/2 \) and the \( J = 3/2 \) bands, and this process may lead to some interesting properties and will be addressed in later works. The electrons can tunnel from one Ir\(^{4+}\) ion to neighboring Ir\(^{4+}\) ions either directly or indirectly via the \( p \) orbitals of the intermediate oxygen ions. Since 5d electron orbitals are spatially extended, therefore the direct tunneling of electrons might be equally important as the indirect tunneling. With electrons locally projected onto the \( J = 1/2 \) basis, one can write down a minimal Hubbard model

\[
\mathcal{H} = \sum_{\langle ij \rangle} \left[ (T_{ij}^d + T_{ij}^{dd})d_{i\alpha}^\dagger d_{j\beta} + h.c. \right] + \sum_i U n_{i\uparrow} n_{i\downarrow},
\]

in which, only the nearest-neighbor tunneling term is included, \( d_{i\alpha}^\dagger \) (\( d_{i\alpha} \)) is the creation (annihilation) operator for an electron on effective spin state \( |J = 1/2, J^z = \alpha \rangle \) at site \( i \), and \( n_{i\sigma} \equiv d_{i\sigma}^\dagger d_{i\sigma} \) measures the electron number with the spin \( \sigma \) at site \( i \). In Eq. (25), \( T^d \) and \( T^{dd} \) are the tunneling matrices for the direct and indirect processes, respectively.

For the direct tunneling processes, there exist two types of tunneling amplitudes: the \( \sigma \)-bonding \( t_1 \), and the \( \pi \)-bonding \( t_2 \) (see Fig. 3). Moreover, it is expected from the orbital overlaps that \( t_2 \) has a different sign from \( t_1 \). In the limit of dominant direct tunneling, standard second order perturbation yields the exchange couplings introduced in Eq. (1).

\[
J_0 = \frac{603^2 t_1^2 - 58296 t_1 t_2 + 248369 t_2^2}{2834352 U},
\]

\[
D = \frac{5\sqrt{2}(153 t_1^2 - 1356 t_1 t_2 + 2528 t_2^2)}{118098 U},
\]

\[
\Gamma_1 = \frac{50(9 t_1^2 - 48 t_1 t_2 + 64 t_2^2)}{177147 U},
\]

\[
\Gamma_2 = 3\Gamma_1.
\]

It turns out that, the DM interaction has the most weight in the exchange Hamiltonian. As \( J_0 \) is assured to be positive in Eq. (29), we depict the ratios of \( D/J_0, \Gamma_1/J_0 \) and \( \Gamma_2/J_0 \) in Fig. 4.

In contrast, the indirect tunneling process is described by one single tunneling amplitude \( t_1^2 \). When it is dominant, the exchange couplings are given by

\[
J_0 = \frac{49132 t_1^2}{59049 U},
\]

\[
D = \frac{7280 \sqrt{2} t_1^2}{59049 U},
\]

\[
\Gamma_1 = \frac{1568 t_1^2}{177147 U},
\]

\[
\Gamma_2 = 3\Gamma_1.
\]

It is important to note that, although we find \( \Gamma_2 = 3\Gamma_1 \) for both limits studied above, this relation is not protected by symmetry and will break down if a more realistic model is assumed. Although we find that \( J_0, D, \Gamma_1, \Gamma_2 \) are all positive for the two limits studied above, this result actually breaks down when both direct and indirect tunnelings are included. As plotted in Fig. 4 for the case of \( t_2 = -t_2/3 \), the Heisenberg exchange \( J_0 \) and DM interaction \( D \) both change sign for certain intermediate ranges of \( t_1/t \). This indicates that different magnetic order may emerge in the intermediate regimes of \( t_1/t \).

### B. Ground states of the exchange Hamiltonian

In the previous subsection, we have explicitly derived the exchange Hamiltonian from the Hubbard model. For both exchanges in the limit of the dominant direct or indirect tunneling, the coupling parameters \( J_0, D, \Gamma_1, \Gamma_2 \) are found to be positive. For this parameter regime, It is ready to show by the mean-field theory and/or directly
observe from the phase diagram depicted in Fig. 7 and 4 that, the mean-field classical ground state manifold is continuously degenerate and is spanned by the two basis vectors \( v_1 \) and \( v_2 \) (see Eq. 7 and Eq. 8). As shown in Fig. 7, there is a region that the DM interaction \( D \) changes sign, that may favor the “4-in-4-out” state as the classical ground state in that region. After a complete calculation, we find the phase diagram that is depicted in Fig. 7. Region II develops the “4-in-4-out” ground state. Region I and III have the degenerate ground state manifold \( (v_1, v_2) \). Remarkably, the phase boundary between region II and region I and III is exactly the same as the one obtained from a self-consistent mean field calculation for the intermediate coupling regime in the calculation below and the one in Ref. 18.

This continuous degeneracy of \( (v_1, v_2) \) ground state manifold will be lifted if the quantum fluctuation is included. We study this quantum order-by-disorder effect by the linear spin-wave theory. We express the classical spin wave Hamiltonian,  

\[
\mathcal{H}_sw = \sum_{\mathbf{k}} [A_{ij}(\mathbf{k})a_i^\dagger(\mathbf{k})a_j(\mathbf{k}) + B_{ij}(\mathbf{k})a_i(-\mathbf{k})a_j(\mathbf{k})]
\]

(35)

and are given in Appendix A. From the quadratic spin-wave Hamiltonian, we obtain the quantum zero-point energy, that is found to be optimized by the non-coplanar spin configuration \( v_2 \) (see Eq. 5) with \( \phi = \pi/2 \) (and its symmetry equivalences). We also find that, the magnon dispersion below and the one in Ref. 18.

FIG. 8. The magnon dispersion along the high symmetry momentum direction \( \Gamma - X - W - L - \Gamma \). The parameters in this figure are set to be \( D = 0.5J_0, \Gamma_1 = 0.2J_0, \Gamma_2 = 0.3J_0 \). The gapless mode at the \( \Gamma \) point is an artifact of the linear spin-wave theory.

C. Hubbard model and electron quasi-itinerancy in the intermediate coupling regime

In the previous subsections, we have analyzed the magnetic ground states of the Ir-based pyrochlore lattice for \( R_2\text{Ir}_2\text{O}_7 \) in the strong coupling regime. We find that, even though the classical mean-field ground states are continuously degenerate for the exchange derived from the Hubbard model, all the ground states have a magnetic wavevector \( \mathbf{q} = 0 \). It is known that, the SOC twists the electron motion and reduces the electron bands. Although the large spatial extension of the 5d electrons reduces the electron correlation, as the bandwidth is also reduced, it is then not quite obvious where the actual physical system is located. Thus, it is legitimate for us to tackle the system from the strong correlation to the intermediate correlation by reducing the correlation strength. The knowledge that we have learned from the strong coupling regime may be extended to the intermediate regime. Moreover, the existing experiments on \( \text{Eu}_2\text{Ir}_2\text{O}_7, \text{Nd}_2\text{Ir}_2\text{O}_7, \text{Tb}_2\text{Ir}_2\text{O}_7 \text{ and Sm}_2\text{Ir}_2\text{O}_7 \) suggest a \( \mathbf{q} = 0 \) magnetic order.\(^\text{29-32}\) In this subsection, we study the magnetic properties of the Hubbard model in the intermediate coupling regime by a self-consistent mean field theory. Based on the results from the strong correlation regime, we assume the magnetic order in this regime also have a magnetic wavevector \( \mathbf{q} = 0 \). To implement the mean-field theory, we decouple the Hubbard-\( U \) interaction as,

\[
Un_i,\uparrow n_{i,\downarrow} = -\frac{2U}{3}J_i^2 + \frac{U}{6}n_i
\]

(37)

protected by any continuous symmetry of the Hamiltonian. A mini-gap would appear if a full calculation is performed.

\[
\rightarrow -\frac{2U}{3}(2\langle J_i \rangle \cdot \mathbf{J}_i - \langle J_i \rangle^2) + \frac{U}{6}n_i
\]

(38)
in which, \( n_i \) is the electron number at site \( i \) and \( \mathbf{J}_i = \sum_{\alpha\beta} d_{i\alpha}^\dagger \sigma_{\alpha\beta} d_{i\beta}/2 \) is the operator for the effective spin moment. With this decoupling, the mean-field Hamiltonian is quadratic with

\[
H_{\text{MF}} \equiv \sum_{\langle ij \rangle} \left[ (T^d_{ij,\alpha\beta} + T^\text{id}_{ij,\alpha\beta}) d_{i\alpha}^\dagger d_{j\beta} + h.c. \right] - \sum_i 4U/3 (\mathbf{J}_i \cdot \mathbf{J}_i + \cdots),
\]

(42)

where “\( \cdots \)” refers to the unessential terms that do not involve the electron operators. We then diagonalize the mean-field Hamiltonian and solve for the magnetic order of each sublattice self-consistently. Our results of the magnetic orders can be found in Fig. 2. In the region II, the calculation quickly converges to the 4-in 4-out magnetic order. For the region I and the region III, the calculation does not quickly converge. After a few steps, the magnetic order from the self-consistent calculation actually drops into the continuous manifold that is spanned by the 4-spin vectors \( \mathbf{v}_1 \) and \( \mathbf{v}_2 \) and then fluctuate within this manifold without seeing a quick convergence. To resolve the magnetic orders in these two regions, we perform a different calculation below that may be illuminating. The self-consistent calculation tells us that the magnetic orders can be parameterized as

\[
\Psi(\phi) = (\mathbf{J}_0, \mathbf{J}_1, \mathbf{J}_2, \mathbf{J}_3) = M(\cos \phi \mathbf{v}_1 + \sin \phi \mathbf{v}_2),
\]

(43)

where the order parameter \( M \) depends on the dimensionless parameter \( U/t \) that measures the strength of the interaction. For a given \( U/t \), the magnetic order parameter \( M \) is fixed. The self-consistent calculation was unable to quickly converge the angular parameter \( \phi \) which is the task to be fulfilled. It is ready to see that the task boils down to optimize the kinetic energy in the mean-field Hamiltonian \( H_{\text{MF}} \), i.e.

\[
\langle \Psi(\phi) \mid \sum_{\langle ij \rangle} \left[ (T^d_{ij,\alpha\beta} + T^\text{id}_{ij,\alpha\beta}) d_{i\alpha}^\dagger d_{j\beta} + h.c. \right] \Psi(\phi) \rangle. \quad (44)
\]

The spirit of this calculation scheme is a bit similar to the double exchange. Over there, the itinerant electron is coupled with the local moments with ferromagnetic Kondo/Hund’s coupling, and the magnetic order is established by optimizing the kinetic energy of the itinerant electrons and the exchange energy of the local moments. In the doped maganites, to gain the kinetic energy, the local moments twist themselves from the spin configuration favored by the exchange energy. Another possible electron kinetic energy driven magnetism was proposed for the doped van der Waals antiferromagnet CeTe\(_3\) and was refereed as fermionic order by disorder. For our case here, the electron kinetic energy is optimized within the background of the magnetism that operates on the continuously degenerate manifold. Our calculation suggests the selection of the angle \( \phi = \pi/2 \) for all \( U > 0 \). We find that, the kinetic energy stabilizes the non-coplanar state with \( \phi = \pi/2 \). Although this mechanism of breaking the continuous degeneracy by optimizing the kinetic energy is qualitatively different from the quantum order by disorder discussed in the previous subsection, the magnetic order from both mechanisms turns out to be identical, the phase boundaries separating different ordered phases are also remarkably identical for both mechanisms. These results suggest that the magnetic orders in the intermediate and the strong coupling regimes may be continuously connected.

IV. DISCUSSION

To summarize, we have studied the magnetic ground states for the Ir-based pyrochlore lattice in both intermediate and strong coupling regimes. Various classical ground states are identified for the generic exchange Hamiltonian in the strong coupling limit. These results can be further applied to other magnetic systems on the pyrochlore lattice. We find that, the magnetic orders in the intermediate and strong coupling regimes for the pyrochlore iridates turn out to be identical.

The experiments on the pyrochlore iridates have rapidly evolved\(^{20,38}\). There exists a large body of experimental works, and the review papers on this topic can be more useful to the interested readers\(^{11,14}\). Instead of delving on a few specific experimental results and details, we here make some experimental suggestion based on the theoretical calculations in our work. In the strong coupling analysis, there exists a broad parameter regime that the magnetic order is realized from the quantum order by disorder mechanism. Once the particular magnetic order with the ordering wavevector \( \mathbf{q} = 0 \) and the spins orientating along the vector \( \mathbf{v}_2 \) in Eq. (6) is realized, one can check if the excitation spectrum and thermodynamic properties are consistent with the theoretical results. A qualitative feature in the magnetic excitation spectrum is the almost gapless mode at the \( \Gamma \) point (see Fig. 8 and the explanation in Sec. III C). A consequence on the thermodynamics is the a nearly \( T^3 \) temperature dependence in the specific heat at the temperatures above the mini-gap energy. In the intermediate coupling scenario, the interaction and the charge gap are not very large compared to the bandwidth. Although the same magnetic order persists to the intermediate coupling regime, the quantum order by disorder mechanism is expected to break down. If one uses the local moment language and relies on the exchange interaction, one necessarily needs to invoke further neighbor exchanges and even the ring exchange interactions. These extra interactions modify the original pairwise nearest-neighbor exchange model and will break the original applicability of the quantum order by disorder here. A surprising result in our self-consistent calculation in Sec. III C is that the magnetic order quickly falls into the degenerate manifold spanned by \( \mathbf{v}_1 \) and \( \mathbf{v}_2 \), and then we use the electron kinetic energy to break the degeneracy and select the magnetic order. This indicates
that the degenerate manifold could be readily accessible if the system is activated by a small energy. A pump-probe measurement of the magnetic properties of the system would be helpful in this regards.

Finally, the weak Mottness with quasi-itinerant electrons might be relevant for many other 4$d$/5$d$ materials. The effect should be considered if the charge gap is not very large. It is very likely that many 4$d$/5$d$ magnets would be located in this regime. Even the square lattice material Sr$_2$IrO$_4$ was believed to be proximate to a Mott transition. The well-known α-RuCl$_3$ has a relatively weak charge gap, even though the existing theoretical analysis mostly starts from a pairwise superexchange interaction between the effective spin-1/2 moments. The interlayer ring exchange, due to the weak Mott gap and the electron quasi-itinerancy, could be responsible for the anomalous thermal Hall effect in α-RuCl$_3$ for the magnetic field in the honeycomb plane and parallel to the zig-zag ordering axis where the interlayer magnetic flux could be experienced by the material.

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Appendix A: The linear spin-wave theory

In Sec. III B of the main text, the couplings $A_{ij}(k)$ and $B_{ij}(k)$ in the spin wave Hamiltonian for the magnetic orders given by the basis vector $v_2$ are listed as follows,

$$A_{00}(k) = A_{11}(k) = A_{22}(k) = A_{33}(k) = c_1,$$

$$A_{12}(k) = \frac{1}{24}(1 + e^{-i(k_y+k_z)^2})c_2,$$

$$A_{13}(k) = \frac{1}{24}(1 + e^{-i(k_y+k_x)^2})c_2,$$

$$A_{14}(k) = \frac{1}{12}(1 + e^{-i(k_y+k_x)^2})c_3,$$

$$A_{23}(k) = \frac{1}{12}(1 + e^{-i(k_y-k_z)^2})c_3,$$

$$A_{24}(k) = \frac{1}{24}(1 + e^{-i(k_y-k_x)^2})c_2,$$

$$A_{34}(k) = \frac{1}{24}(1 + e^{-i(k_y-k_z)^2})c_2,$$

and

$$B_{12}(k) = \frac{1}{24}(1 + e^{-i(k_y+k_x)^2})c_4,$$

$$B_{13}(k) = \frac{1}{24}(1 + e^{-i(k_y+k_x)^2})c_4^*,$$

$$B_{14}(k) = \frac{1}{12}(1 + e^{-i(k_y+k_x)^2})c_5,$$

$$B_{23}(k) = \frac{1}{12}(1 + e^{-i(k_y-k_z)^2})c_5,$$

$$B_{24}(k) = \frac{1}{24}(1 + e^{-i(k_y-k_x)^2})c_4^*,$$

$$B_{34}(k) = \frac{1}{24}(1 + e^{-i(k_y-k_z)^2})c_4,$$

in which, we have set $J = 1/2$ and the coefficients are given as

$$c_1 = J_0 + \sqrt{2}D + 4\Gamma_1 + \Gamma_2,$$

$$c_2 = -2J_0 + \sqrt{2}D - 17\Gamma_1 + 4\Gamma_2,$$

$$c_3 = -4J_0 + 2\sqrt{2}D - 7\Gamma_1 - \Gamma_2,$$

$$c_4 = -(2 + 4i\sqrt{6})J_0 + (7\sqrt{2} - 2i\sqrt{3})D + (1 + 2i\sqrt{6})\Gamma_1 + (4 + 2i\sqrt{6})\Gamma_2,$$

$$c_5 = 2J_0 + 2\sqrt{2}D - \Gamma_1 + 5\Gamma_2.$$
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