RKKY interactions and anomalous Hall effect in metallic rare-earth pyrochlores

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Motivated by experiments on Pr2Ir2O7, we consider metallic pyrochlore systems A2B2O7, where the A-sites are occupied by rare-earth local moments and the B-sites host 5d transition metal ions with itinerant strongly spin-orbit coupled electrons. Assuming non-Kramers doublets on the A-site, we derive the RKKY interaction between them mediated by the B-site itinerant electrons and find extended non-Heisenberg interactions. Analyzing a simplified model of the RKKY interaction, we uncover a local moment phase with coexisting spiral Ising-like magnetic dipolar and XY-like quadrupolar ordering. This state breaks time-reversal and lattice symmetries, and reconstructs the B-site electronic band structure, producing a Weyl Metallic phase with an intrinsic anomalous Hall effect and an undetectably small magnetization. We discuss implications of our results for Pr2Ir2O7.

The metallic pyrochlores, A2B2O7, with a rare-earth A-site ion and a 5d transition metal B-site ion, lie at the intersection of exciting recent developments in condensed matter physics. The rare-earth moments on the A-site pyrochlore sublattice could lead to quantum spin ice physics [11,2] from Ising anisotropy and geometric frustration, a feature they share with the well-studied insulating pyrochlore oxides. The strongly spin-orbit coupled 5d conduction electrons, on the other hand, contain the seeds of topological phases like Weyl semimetals or topological insulators [3–8]. The interplay of these two effects could pave the way for new emergent phenomena. The pyrochlore iridate Pr2Ir2O7 provides an example of such a metallic frustrated system, with a significant Curie-Weiss temperature $\theta_{\text{CW}} \approx -20$K, much larger than in the insulating compounds Pr2Sn2O7 or Pr2Ti2O7 [9,12]. For $T \lesssim 1.7$K, even when an applied magnetic field along the $\langle 111 \rangle$ direction is switched off, it exhibits a significant anomalous Hall effect (AHE) which grows upon cooling [13–15], although, in contrast to Nd2M02O7, the uniform magnetization is undetectably small over a range of temperatures [16]. This raises the key issue of mechanisms underlying the AHE [17] in materials with geometric frustration and strong spin-orbit coupling.

For metallic pyrochlores, the RKKY interaction between the local $f$-moments induced by the conduction electrons is expected to be important. When the $f$-moment is a non-Kramers doublet, such as in Pr$^{3+}$, the pseudospin component $\tau^z$ along the local $\langle 111 \rangle$ direction carries a magnetic dipole moment and couples to the conduction electron spin density, while the $\tau^{x,y}$ components carry a quadrupole moment and couple to the electronic charge density. We show that the resulting RKKY coupling has a highly non-Heisenberg form and extends beyond the nearest neighbor term. We propose and study a simplified such extended XXZ model for non-Kramers doublets using a variational analysis, finding a rich phase diagram, shown in Fig. [1] which includes incommensurate spiral ‘coexistence’ states (CE1, CE2), with wavevec-

![Variational phase diagram of the pseudospin Hamiltonian in Eq.(2)](image)

FIG. 1: Variational phase diagram of the pseudospin Hamiltonian in Eq.(2) relevant to RKKY coupled non-Kramers doublets as a function of anisotropy $w$ and $\theta = \tan^{-1}(J_2/J_1)$. $w < 0$: We find ferro-XY (FM-XY), antiferro-XY (AF-XY), where spins order in the plane transverse to the local $\langle 111 \rangle$ axes. These are time-reversal invariant states with modulated quadrupolar order. $w > 0$: We find Ising all-in/out (AIO), Ising two-in-out (2IO) and states with coexisting modulated Ising-XY orders (CE1,CE2). The CE2 state breaks time-reversal and lattice symmetries, producing an anomalous Hall effect for the spin-orbit coupled conduction electrons.

tors $(0, q, \pi)$ or $(0, 0, q)$, having spatially modulated, magnetic (dipolar) and quadrupolar order, i.e., ‘magnetoquadrupolar supersolids’. We confirm that such a CE2 state also exists within a classical Monte Carlo simulation. Although the CE2 state has no net local moment magnetization, it reconstructs the band structure of the spin-orbit coupled 5d-electrons, producing Weyl points as well as small Fermi pockets, leading to a measurable AHE and an extremely small conduction electron magnetization [29]. We discuss possible implications and predictions for Pr2Ir2O7, for which our proposal appears to be distinct from the more widely discussed spin chirality scenario [15,18,20].
Basic microscopics and RKKY coupling.— Since we are motivated by experiments on Pr$_2$Ir$_2$O$_7$, we briefly review the relevant microscopics of the local moments and conduction electrons in this material. The Pr$^{3+}$ ion is in a $4f^2$ configuration, with electron-electron interactions and strong spin-orbit coupling favoring a total angular momentum state with $J=4$. This ninefold degenerate manifold is split by crystal fields with $D_{3h}$ point group symmetry around the Pr$^{3+}$ ion, arising from a cage of eight O$^{2-}$ ions stretched along the local (111) axis. The splitting is captured by an effective time-reversal invariant crystal field Hamiltonian expressed in terms of $J=4$ angular momentum operators, $H_{\text{CEF}} = -\alpha J_z^2 + \beta (J^3_{x} + J^3_{y}) + \gamma (J^6_{x} + J^6_{y})$, with $\alpha > 0$ and $\beta, \gamma \ll \alpha$, leading to a ground state non-Kramers doublet with a dominant $|J_z = \pm 4\rangle$ component.\cite{21} Projection to this low energy doublet allows us to define pseudospin-1/2 operators $\mathbf{\tau}$ with $\mathbf{\tau} \propto J_2$, while $\mathbf{\tau} \propto \{J_\pm, J_y\}$. Under time-reversal, $\mathbf{\tau} \to -\mathbf{\tau}$, transforming as the magnetic dipole moment, while $\mathbf{\tau} \cdot \mathbf{\tau}$ are left invariant, transforming like a quadrupole moment.

For Ir$^{4+}$ electrons, strong spin-orbit coupling in the $t_{2g}$ manifold leads to a half-filled effective $j=1/2$ band. The metallic character of Pr$_2$Ir$_2$O$_7$ suggests that a tight binding model which ignores strong electronic correlations would be an adequate starting point,

$$H_{\text{tb}} = \sum_{ij} \sum_{\alpha \beta} c_{i \alpha}^\dagger (t_{ij} \delta_{\alpha \beta} + i v_{ij} \cdot \sigma_{\alpha \beta}) c_{j \beta},$$

where $c_{i \alpha}^\dagger (c_{j \beta})$ denotes the electron creation (annihilation) operator at site $i$ with the Kramers pseudospin index $\alpha$ corresponding to $j_z = \pm 1/2$, and $\sigma = (\sigma^x, \sigma^y, \sigma^z)$ are the Pauli matrices. We assume $t_{ij} = t$ ($t'$) for nearest (next-nearest) neighbors, and $v_{ij} \neq 0$ only for nearest-neighbors and is constrained by lattice symmetries.\cite{22}

How does the A-site non-Kramers doublet couple to the B-site conduction electrons? Unlike the usual Kondo coupling to a magnetic Kramers doublet, time-reversal invariance dictates that the Ising component of the A-site pseudospin $\tau^z_{j}$ at site $j$ (which points along the local (111) axis) couples to spin density $j^z_m = c_{i \alpha}^\dagger n_i \sigma^z_{\alpha \beta} c_{j \beta}$ of electrons on the six neighboring B-sites, while the planar components of the pseudospin $\tau^\pm_{j}$ couple to the charge density $n_i = c_{i \alpha}^\dagger c_{i \alpha}$ on the neighboring B-sites. Keeping $j^z_m$ and $n_i \tau^\pm_{j}$ terms, we find the symmetry allowed Kondo coupling $H_{AB}$ with three parameters $c_1, c_2, c_3$. (See Supplementary Materials for details.) Integrating out the conduction electrons, we find that the resulting RKKY interaction has two important features which are insensitive to the details of the Ir band structure: (i) it allows for significant couplings beyond the nearest-neighbor interaction, but is negligible beyond the third neighbor; (ii) it is highly anisotropic in spin-space since $\tau^+ \cdot \tau^-$ and $\tau^z$ interact very differently with the spin-orbit coupled conduction band. A complete microscopic set of Kondo couplings should include $\tau^z_{j} c_{i \alpha}^\dagger \sigma^z_{\alpha \beta} c_{j \beta}$ or $\tau^\pm_{j} c_{i \alpha}^\dagger c_{i \alpha}$ with $i \neq j$, terms which we have omitted, and the full characterization may require multiple parameters. Rather than dealing with such a complex model, we would like to focus on general and robust features of the RKKY interactions. We therefore turn to a study of a simplified RKKY Hamiltonian which retains the two key features described above.

Local Moment Model and Phase Diagram. — Motivated by our observation that the RKKY interaction between local moments is highly anisotropic in spin space, and has beyond nearest-neighbor terms, we study a simplified model in the basis with a sublattice-dependent quantization axis along the local (111) direction,

$$H = \sum_{r, r'} \left[ (1 - w) \tau^z_r (r) \tau^z_{r'} (r') + \tau^+_r (r) \cdot \tau^+_r (r') \right]$$

Here $J_{r, r'} = J_1 (J_2)$ for nearest (next-nearest) neighbor sites, $\tau^z_{r}(r)$ is $z$-component of the pseudospin on sublattice $s$ at site $r$, $\tau^+_r (r)$ denotes the transverse component of the pseudospin which lies in the local $XY$ plane, and $w$ quantifies the exchange anisotropy. We set $J_1 = J \cos \theta$ and $J_2 = J \sin \theta$, and explore the phase diagram of this model as a function of $(\theta, w)$.

Treating the spins as classical unit vectors, we minimize the energy using a variational ansatz

$$\tau_s (r) = d_s \hat{e}_3 + \sqrt{1 - d^2_s} \Re e [i(\hat{e}_1 + i \hat{e}_2) e^{i(Q \cdot r + \varphi_s)}],$$

where $\hat{e}_{1,2,3}$ form a triad of orthonormal vectors, so that $|\tau_s (r)| = 1$. In the local coordinate system, this ansatz allows, (i) for $d_s = 0$, a coplanar spiral with wavevector $Q$ with spins in the ($\hat{e}_1, \hat{e}_2$) plane, and, (ii) for $d_s^2 = 1$, a collinear state with spins along $\pm \hat{e}_3$. In the isotropic limit, $w = 0$, this ansatz recovers (0, 0, q) spirals\cite{23}, while in the Ising limit it allows for 2-in-2-out or all-in all-out states. The complete ground state phase diagram from this variational analysis is shown in Fig.\cite{1}

For $w > 0$, by contrast, we find in addition to well known states like the ferromagnetic Ising (all-in all-out), and Ising spin ice (2-in 2-out), large parameter regimes which support coexistence phases (CE$_1$, CE$_2$) with coplanar order involving spatially modulated Ising (magnetic dipolar) and XY (quadrupolar) order. The CE$_1$ and CE$_2$ states order at the wavevectors $(0, q, \pi)$ and $(0, 0, q)$ respectively (or their symmetry related momenta). Such
CE states are ‘magneto-quadrupolar’ super solids which break time-reversal symmetry (defined by $\tau^z \rightarrow -\tau^z$), and most lattice symmetries. Below, we focus on the remarkable physical properties including the AHE of the CE2 state which is (i) robustly present in an unbiased numerical energy minimization using a simulated annealing approach [30], and (ii) stable to the addition of weak perturbations such as $(\tau_i^x \tau_j^x + \tau_i^y \tau_j^y)$ to the simple Hamiltonian in Eq. [2].

AHE and Magnetization in the CE2 state. — Once the CE2 state is stabilized on the A-site, it implements effective spatially varying magnetic fields and chemical potentials on the B-site conduction electrons due to the Kondo-type coupling between them. For an ordering wave vector $Q$, this mixes electrons with wave vectors $k$ and $k + Q$, leading to a Hamiltonian of the form

$$H_{\text{th}}(k) = \left( \begin{array}{cc} H_{\text{th}}(k) & H_{AB}(Q) \\ H_{AB}(-Q) & H_{\text{th}}(k + Q) \end{array} \right)$$

(4)

For the CE2 state in our anisotropic $J_1$-$J_2$ pseudospin model, $Q = (00q)$, and time-reversal symmetry and all lattice symmetries except $C_{2z}$ ($\pi$ rotation along $\hat{z}$ direction) are broken. Once these broken symmetries are inherited by the resulting 5d electron band structure, one can argue for an intrinsic AHE. The $C_{2z}$ symmetry leads to $\sigma_{yz} = \sigma_{xz} = 0$ since the current operator $J_z$ is invariant under $C_{2z}$ rotation $(x, y, z) \rightarrow (-x, -y, z)$ while the currents $J_y$ and $J_x$ change sign. However, $\sigma_{xy}$ remains unchanged under this rotation and can thus be nonzero.

To explicitly compute the Hall conductivity, we assume the tight-binding parameterization for the Ir 5d electrons in Eq. [1], as relevant to Pr$_2$Ir$_2$O$_7$, choosing $t = 1$, $t' = -0.1$, and $|v_{ij}| = 0.2$. These values are close to those determined from the Slater-Koster parameters [22].

Fig. 2 shows an example of the reconstructed B-site band structure based on Eq. [4]. (Red line indicates the Fermi level.) We consider the A-site incommensurate CE2 local moment order with wave vector $Q \approx 1.2\pi(001)$, which is appropriate for $\theta = 1.72\pi$, $w = -0.5$ in Eq. [2] (marked in Fig. 1), and the Kondo coupling constant $c/t = 0.1$ where $c_1 = 0, c_2 = c_3 = c$ in $H_{AB}(Q)$. (See Supplementary material) The reconstructed band structure generates both Weyl points and Fermi pockets near $\Gamma, \Gamma + Q$. There are four pairs of Weyl points in total (two pairs near $\Gamma, \Gamma + Q$ and other two pairs at their $C_{2z}$ symmetry related points) and the inset of Fig. 2 shows one of those Weyl points and Fermi pockets near $k \approx (0, 0.12\pi, 0.02\pi)$.

Fig. 3 shows the explicitly calculated Hall conductivity using the Kubo formula as a function of Kondo coupling constant $c$, based on the reconstructed B-site band structures. The strength of Kondo coupling $c$ determines the magnitude of the AHE response. $\sigma_{xy}$ initially increases with increasing $c$, acquiring contributions from small electron-like and hole-like Fermi pockets as well as four pairs of Weyl points induced by the CE2 order. For $c/t \gtrsim 0.7$, a band gap opens up and $\sigma_{xy} = 0$.

On symmetry grounds, the AHE must be accompanied by a nonzero uniform magnetization, with $M_x = M_y = 0$ but $M_z \neq 0$. Remarkably, although the CE2 state has no net magnetization from the local moments, we find that it induces a nonzero magnetization $M_z$ for the 5d electrons, where $M_z = \frac{1}{N} \sum_i \sum_{\alpha \beta} \gamma_{\alpha \beta} \left( \frac{1}{2} \delta_{\alpha z} \sigma^z_{\beta} \chi \right)$. For small $c$, the net magnetization $M_z$ gets larger, proportional to the density of states (DOS) near the Fermi level, with the same sign and a trend qualitatively similar to $\sigma_{xy}$. For large $c/t \gtrsim 0.7$, we find that $M_z \neq 0$ although $\sigma_{xy} = 0$, signalling a magnetized band insulator.

This dichotomy of a large $\sigma_{xy}$ but a small $M$ can be argued for as follows. On dimensional grounds, the AHE signal $\sigma_{xy} \sim \frac{c}{\hbar} \Delta k$, where the momentum scale $\Delta k$ must be induced by an effective ‘internal magnetic field’ $B^{\text{int}}$ due to the spontaneous breaking of time-reversal symmetry [23, 24]. If the $\tau^{x,y}$ order in the CE2 state reconstructs the band structure to produce a small Fermi pocket, with a Fermi wavevector $k_F$ and an effective mass $m^*$, we expect $\Delta k \sim B^{\text{int}} m^*/k_F$ resulting in a large $\sigma_{xy}$ due to a small $k_F$ Fermi pocket, while the magnetization $M \sim B^{\text{int}} m^* k_F$ stays small. The Weyl point contribution from splitting a quadratic band touching, with a curvature $m^{**}$, on the other hand leads to $\Delta k \sim \sqrt{m^{**} B^{\text{int}}}$ [27], and a magnetization $M \sim B^{\text{int}}$ again...
leads to a large $\sigma_{xy}$ and a small $M$. The presence of both contributions would lead to a non-linear $\sigma_{xy}(M)$.

**Application to Pr$_2$Ir$_2$O$_7$:** The CE$_2$ state in our simple model of RKKY coupled non-Kramers ions and spin-orbit coupled conduction electrons captures a key aspect of the experimental data on Pr$_2$Ir$_2$O$_7$: an AHE accompanied by a negligible magnetization. However, since our ordering wavevector is along the (001) direction, the spontaneous AHE is produced in the $xy$-plane, whereas the spontaneous AHE in Pr$_2$Ir$_2$O$_7$ is seen for fields along the ⟨111⟩ direction. This discrepancy may be resolved if such coexisting spiral order had a wavevector along the ⟨111⟩ direction, or if our CE order gave way to a multimode spiral formed by superposing (0,0,q), (0,q,0), (q,0,0) spirals while preserving $C_3$ rotation along ⟨111⟩ direction but breaking all other lattice symmetries. The Hall conductivity in that case will naturally be in the plane perpendicular to ⟨111⟩, in agreement with experiment. We estimate the magnitude of the AHE (at $c/t = 0.1$, which is $c \approx 5$meV) in our CE$_2$ state to be $\sigma_{xy} \approx -15(\Omega^{-1} \text{cm}^{-1})$ and $M_z \approx -0.002(\mu_B/Ir)$. This is consistent with the magnitude of the experimentally measured Hall signal, and the absence of any measurable magnetization over a range of temperatures. At lower temperatures, experiments detect a nonzero remnant magnetization, which may arise in our model from the feedback of the magnetized conduction electrons on the local moments, an effect we have not taken into account.

The Curie-Weiss temperature is sensitive to $J_2/J_1$, changing sign as a function of $\theta$ even within the CE$_2$ state. For couplings $c/t \sim 0.1$, we can obtain $\theta_{CW}$ to be of the right sign and magnitude, $\theta_{CW} \sim -20K$ as observed experimentally, for parameter values closer to the all-in-all-out phase boundary.

The CE$_2$ state should exhibit two distinct thermal transitions, associated with the onset of Ising and XY orders, with this splitting being smaller for a weaker anisotropy $w$. However, this simple expectation gets confounded by two issues. (i) Terms which we have omitted, such as $(\tau_i^+ \tau_j^- + \tau_i^- \tau_j^+)$, will break the (staggered) U(1) invariance of the XY terms, and modify the 3d-XY universality class of the quadrupolar ordering transition. (ii) Oxygen vacancy defects will produce strong electric fields which break the $D_{3d}$ point group symmetry around the Pr$^{3+}$ ion, splitting the non-Kramers doublet due to extra random terms $\Delta H_{\text{vac}} \propto \{J_z, J_{xy}\} \sim \tau^z$ in the effective crystal field Hamiltonian. This would lead to a time-reversal invariant strong random field on $\tau^{z,y}$. These effects might conspire smear or destroy the XY ordering transition. However, the time-reversal symmetry breaking Ising transition is expected to survive, which would be consistent with the single specific heat “peak” seen at the onset of the spontaneous AHE in Pr$_2$Ir$_2$O$_7$. Monte Carlo studies of the thermal properties and disorder effects will be discussed elsewhere [24].

The most direct evidence for the scenario advocated here would be a probe which can detect the modulated Ising order using neutron diffraction. Landau theory arguments predict that entering such a coexistence phase in a clean system would also lead to a weak charge density wave of the Ir electrons at the ordering wavevector of the spiral, from a term $\propto \tau^{z,y}(q)\rho(-q)$, and which we find small but finite in our calculations; such charge order may be weak but can be probed, in principle, using X-ray diffraction. In the presence of an induced charge order, Landau theory arguments also predict a nonzero $d$-band magnetization, from a term $\propto M^z(q)\rho(-q)$, which is indeed present as discussed above. Quantum oscillation measurements to detect the ordering-induced Fermi pockets are desirable, but might be difficult due to the significant AHE and the small size of the pockets.

**Conclusion.**—In conclusion, we have proposed a mechanism of intrinsic AHE in metallic pyrochlore systems, such as Pr$_2$Ir$_2$O$_7$, arising from spiral order of local moments driven by their extended anisotropic RKKY exchange interactions, and the resulting reconstruction of the electronic band structure to form small Fermi pockets and pairs of Weyl points. This ordering could occur proximate to an all-in-all-out state of the local moments, and appears to be distinct from previously proposed spin-chirality scenarios for the AHE in Pr$_2$Ir$_2$O$_7$.

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[29] The presence of Fermi pockets does not allow us to separate the band and Weyl point contributions to $\sigma_{xy}$.
[30] Although we have not mapped out the entire parameter space, we find that the variational ansatz captures most of the phases found in the simulated annealing approach. Our ansatz does not capture regimes where multiple-$Q$ orders are favored - such states are well known to occur in the isotropic case $w = 0$ in the regime where we find CE$_1$ order, and will be discussed separately in future work for $w \neq 0$. 



Supplementary Material

Local coordinates and primitive vectors in A$_2$B$_2$O$_7$

First, we define the local coordinates for every A-site pyrochlore lattice. The local spin quantization axis ($\hat{z}$) is always pointing towards the center of each tetrahedron and $\hat{x}, \hat{y}$ axes are defined on its basal plane. Table I shows their local coordinates frames depending on the four sublattices of the pyrochlore lattice. The A-site pseudospin in global coordinates, say $\hat{\tau}_s(r)$, can be represented as

$$\hat{\tau}_s(r) = \tau^x_s(r)\hat{x}_s + \tau^y_s(r)\hat{y}_s + \tau^z_s(r)\hat{z}_s$$  

(5)

where $\tau_s(r) = (\tau^x_s(r), \tau^y_s(r), \tau^z_s(r))$ is the pseudospin at site $r$ and sublattice $s$ defined in the local coordinates. Both A and B-sites form pyrochlore lattice and each can be viewed as FCC lattice with a four-site basis. We define the primitive vectors ($b_i$) and the position of four sublattices ($A_i, B_i$) for A and B-sites in Table II.

| $i$ | 0   | 1   | 2   | 3   |
|-----|-----|-----|-----|-----|
| $x_i$ | $\frac{1}{\sqrt{2}}(011)$ | $\frac{1}{\sqrt{2}}(011)$ | $\frac{1}{\sqrt{2}}(011)$ | $\frac{1}{\sqrt{2}}(011)$ |
| $y_i$ | $\frac{1}{\sqrt{2}}(211)$ | $\frac{1}{\sqrt{2}}(211)$ | $\frac{1}{\sqrt{2}}(211)$ | $\frac{1}{\sqrt{2}}(211)$ |
| $z_i$ | $\frac{1}{\sqrt{2}}(111)$ | $\frac{1}{\sqrt{2}}(111)$ | $\frac{1}{\sqrt{2}}(111)$ | $\frac{1}{\sqrt{2}}(111)$ |

TABLE I: Local coordinate frames for the four sublattices on the pyrochlore lattice.

Kondo-like coupling between A and B-sites

In this section, we derive the explicit form of the Kondo-like coupling between the A-site non-Kramers doublet and the B-site Kramers doublet. As we argued in the main text, the Ising component of the A-site pseudospin $\tau^z_i$ couples to their six neighboring B-site magnetic moments $j^\mu_i = c_i^\alpha a_i^{\alpha \mu} c_{i\beta}$, whereas, the A-site planar components $\tau^\pm_i$ couple to the B-site electron density $n_i = c_i^\alpha c_i^\alpha$. Hence, we separate the former and the latter cases and derive the Kondo-like coupling term allowed by lattice symmetry. First of all, let’s consider the former case where $\tau^z_i$ couples to $j^\mu_i$. We note that this magnetic coupling term is already derived in Ref. 28.

$$H_{A/B}^{zz} = \sum_r c_1 \left[ \{ j^z_i(r) + j^z_i(r - b_2 + b_3) + j^z_i(r + b_1 - b_2) + j^z_i(r - b_2 + b_3) + j^z_i(r) + j^z_i(r + b_1 - b_2) \} \tau^z_0(r) 

+ \{ j^z_i(r + b_1) + j^z_i(r + b_1 - b_2 + b_3) - j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2 + b_3) 

- j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) \} \tau^z_1(r) + \{ j^z_i(r + b_1) + j^z_i(r + b_1 - b_2 + b_3) 

- j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) \} \tau^z_2(r) + \{ j^z_i(r + b_1) + j^z_i(r + b_1 - b_2 + b_3) 

- j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) \} \tau^z_3(r) \right] 

+ c_2 \left[ \{ j^z_i(r) + j^z_i(r - b_2 + b_3) + j^z_i(r - b_2 + b_3) + j^z_i(r + b_1 - b_2) + j^z_i(r) + j^z_i(r + b_1 - b_2) \} \tau^z_0(r) 

+ \{ -j^z_i(r + b_1) - j^z_i(r + b_1 - b_2 + b_3) - j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2 + b_3) 

+ j^z_i(r + b_1 - b_2) + j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) + j^z_i(r + b_1 - b_2) \} \tau^z_1(r) + \{ j^z_i(r + b_1) + j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) \} \tau^z_2(r) 

+ \{ j^z_i(r + b_1) + j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) - j^z_i(r + b_1 - b_2) \} \tau^z_3(r) \right] 

+ \{ \} \right]$$

Here, $\tau^z_i(r)$ is the Ising component of the pseudospin at sublattice $i (= 0, 1, 2, 3)$ in unit cell labeled by $r$ which is pointing along local $\hat{z}_i$ direction. (See Table I for their

| $i$ | 0   | 1   | 2   | 3   |
|-----|-----|-----|-----|-----|
| $A_i$ | (0, 1, 0) | (0, 0, 1) | (1/2, 1, 0) | (1/2, 1, 0) |
| $B_i$ | (0, 0, 0) | (0, 0, 1) | (1/2, 0, 1/2) | (1/2, 1/2, 0) |
| $b_i$ | (0, 1, 1) | (1/2, 1/2, 1) | (1/2, 1/2, 1) | (1/2, 1/2, 1) |

TABLE II: Primitive vectors ($b_i$) and four basis vectors on each A-site ($A_i$) and B-site ($B_i$).
local coordinate frames for the four sublattices on A-site pyrochlore lattice) In a similar way, $j^\mu_i(r)$ is $\mu = (x, y, z)$ component of B-site magnetic moments in global cubic coordinates. Primitive vectors ($b_i$) and four basis vectors on each A-site ($A_i$) and B-site ($B_i$) are shown in Table II. There are two independent parameters, $c_1$ and $c_2$ which cannot be determined by symmetry grounds. However, two independent parameters can be easily understood as follows. For every given site $i$ (on the A-site), there are one local $\hat{z}_i$ axis and its basal plane formed by $\hat{x}_i$ and $\hat{y}_i$. (See Table II) The magnetic coupling can be written as $\tau_1^z (c_j^x j^y_j + c_j^y j^x_j + c_j^z j^z_j)$ assuming that $j^z_j$ is aligned along their local $\hat{z}_i$ axis. For its basal plane, we have freedom to choose $\hat{x}_j$ and $\hat{y}_j$, which leaves $c_j^x = c_j^y$. Once the coupling between A-site $i$ and one of their six neighboring B-site $j$ is determined, the coupling with all the other five neighbors are determined by lattice $c_3$ rotation and $\sigma_h$ mirror symmetries, with two independent parameters $c_j^z$ and $c_j^x = c_j^y$.

On the other hand, the planar components of the A-site pseudospin are time reversal invariant and they do not couple to B-site magnetic moments but couple to the B-site electron density $n_i = c_{i\alpha} c_{i\alpha}$. In this case, there is only one independent parameter and one can write the coupling term as following

\[
H^{\pm}_{AB} = c_3 \sum_r \left[ \tau_0^+(r) \{ \omega^2 n_2 (b_1 - b_2 + r) + \omega^2 n_2 (-b_2 + b_3 + r) + \omega n_3 (b_1 - b_2 + r) + \omega n_3 (b_1 - b_2 + r) + \omega n_3 (b_2 - b_2 + r) \} + n_1 (-b_2 + b_2 + r) + n_1 (r) \right] + \tau_1^+(r) \{ \omega^2 n_2 (b_1 + r) + \omega^2 n_2 (b_1 - b_2 + r) + \omega n_2 (b_1 - b_2 + r) + n_0 (b_1 + r) + n_0 (b_1 - b_2 + b_3 + r) \} + \tau_2^+(r) \{ \omega^2 n_2 (b_1 + r) + \omega^2 n_2 (b_1 + r) + \omega n_2 (b_1 + r) + n_0 (b_1 + r) + n_0 (b_1 - b_2 + b_3 + r) \} + \tau_3^+(r) \{ \omega^2 n_1 (b_1 + r) + \omega^2 n_1 (b_1 - b_2 + r) + n_0 (b_1 - b_2 + b_3 + r) + n_0 (b_1 - b_2 + b_3 + r) \} + n_2 (-b_2 + b_3 + r) + n_2 (b_1 - b_2 + b_3 + r) \} + h.c \]  

(7)

Extra phases $1, \omega = e^{i2\pi/3}, \omega^2$ are present depending on the different sublattices due to their local coordinates on the A-site pseudospin. Combining $H^{\pm}_{AB}$ and $H^{\pm}_{AB}$, the Kondo-like coupling with three independent coupling parameters $c_1, c_2, c_3$ is written as

\[
H_{AB}(c_1, c_2, c_3) = H^{\pm}_{AB}(c_1, c_2) + H^{\pm}_{AB}(c_3). \quad (8)
\]

In the presence of magnetic ordering on the A-site, we take $\mathbf{\tau}_s(r) \rightarrow (\mathbf{\tau}_s(r))$ which acts as an effective magnetic field or chemical potential at every B-site itinerant electron.