Fluctuation-Driven Selection at Criticality in a Frustrated Magnetic System: the Case of Multiple-\(k\) Partial Order on the Pyrochlore Lattice

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We study the problem of partially ordered phases with periodically arranged disordered (paramagnetic) sites on the pyrochlore lattice, a network of corner-sharing tetrahedra. The periodicity of these phases is characterized by one or more wave vectors \(k = \{2\ 2\ 2\}\). Starting from a general microscopic Hamiltonian including anisotropic nearest-neighbor exchange, long-range dipolar interactions and second- and third-nearest neighbor exchange, we identify using standard mean-field theory (s-MFT) an extended range of interaction parameters that support partially ordered phases. We demonstrate that thermal fluctuations ignored in s-MFT are responsible for the selection of one particular partially ordered phase, e.g. the “4-\(k\)” phase over the “1-\(k\)” phase. We suggest that the transition into the 4-\(k\) phase is continuous with its critical properties controlled by the cubic fixed point of a Ginzburg-Landau theory with a 4-component vector order-parameter. By combining an extension of the Thouless-Anderson-Palmer method originally used to study fluctuations in spin glasses with parallel-tempering Monte-Carlo simulations, we establish the phase diagram for different types of partially ordered phases. Our results elucidate the long-standing puzzle concerning the origin of the 4-\(k\) partially ordered phase observed in the Gd\(_2\)Ti\(_2\)O\(_7\) dipolar pyrochlore antiferromagnet below its paramagnetic phase transition temperature.

FIG. 1. (Color online) The yellow (arrowless) sites are disordered (paramagnetic). \(a\) a 1-\(k\) state with \(k\) along [111]. The spins on the (blue) ordered sites form a 120° pattern on a triangle that does not share a corner with a disordered site. These ordered sites form a kagome plane (blue-shaded plane) perpendicular to the [111] direction. \(b\) 4-\(k\) state arising from the superposition of four 1-\(k\) states.

Highly frustrated magnetism is one of the paradigms of modern condensed matter physics [1]. In frustrated magnets, the combination of lattice geometry and competing interactions often leads to degenerate classical states. The degeneracies are generally accidental as they are not protected by the symmetries of the spin Hamiltonian. Yet, the degenerate states may be related by transformations that form an emergent symmetry group. Near a continuous phase transition, these approximate symmetries provide “organizing principles” in determining the critical properties by distinguishing relevant perturbations from irrelevant ones. In the most interesting case, the leading degeneracy-lifting perturbations, which may be relevant or irrelevant in the renormalization group sense, are thermal or quantum fluctuations – a phenomenon called order-by-disorder (ObD) [2–5]. The competition among diverse degeneracy-lifting effects can result in a modulated long-range ordered state at nonzero wave vector \(k\), which may or may not be commensurate with the lattice [6–12]. In some cases, a number of superposed symmetry-related \(k\) modes within the first Brillouin zone form a so-called multiple-\(k\) order [6, 7, 13, 14]. A particular interesting form of such modulated magnetism is a partially ordered state (POS) with periodically arranged “paramagnetic” sites [15–18]. These fluctuating magnetic moments decimate a fraction of the energy-costly frustrated bonds while retaining an extensive entropy, hence lowering the free energy.

In this Letter, we study the convergence of the aforementioned phenomena (emergent symmetry, multiple-\(k\) POS and fluctuation-induced degeneracy-lifting) in an extensively studied class of frustrated magnetic materials, the insulating \(R_2M_2O_7\) pyrochlore oxides [19]. In these, the \(R^{3+}\) magnetic rare-earth ions (e.g. Gd, Tb, Er, Yb) occupy the vertices of a network of corner-sharing tetrahedra – the “pyrochlore” lattice (see Fig. 1). \(M^{4+}\) (\(M=\text{Ti, Sn, Zr, Ge}\)) is nonmagnetic. The competition between four types of nearest-neighbor anisotropic interactions and the nature of the single-ion magnetic anisotropy are largely responsible for the wealth of phenomena displayed by the \(R_2M_2O_7\) materials [19]. In this paper, we focus on a general description for the perplexing yet rich physics of multi-\(k\) partial magnetic ordering in pyrochlore oxides and not on any material-specific issues.

A multi-\(k\) POS (see Fig. 1b) is believed to exist in Gd\(_2\)Ti\(_2\)O\(_7\) [20] for temperature \(0.7 \, \text{K} \lesssim T \lesssim T_c \sim 1 \, \text{K}\) in which the POS is a superposition of spin density waves with wave vectors \(k = \{1\ 1\ 1\}\) [20–26]. While this compound has been the subject of a number of investigations [20–22, 27–36], the mechanism responsible for the selection of 4-\(k\) order has not been identified. Further, after fifteen years of research
on the Tb$_2$Ti$_2$O$_7$ spin liquid candidate [37], evidence has recently begun accumulating that short-range magnetic correlations develop below $T \sim 0.4$ K in the form of broad elastic neutron intensities at $k = \{\frac{1}{2} \frac{1}{2} \frac{1}{2}\}$ [38–42]. No theory has explained the origin of these correlations. As Tb$_2$Ti$_2$O$_7$ and Gd$_2$Ti$_2$O$_7$ constitute two out of the six magnetic $R_2$Ti$_2$O$_7$ pyrochlores compounds that exist ($R$=Gd, Tb, Dy, Ho, Er and Yb), it may be that $k = \{\frac{1}{2} \frac{1}{2} \frac{1}{2}\}$ is not unusual among the plethora of $R_2$Ti$_2$O$_7$ materials, or even all $AR_2B_4$ rare-earth spinels ($A=$Cd, Mg; $B=$S, Se and $R$ is a rare-earth) [43–45].

In our work, we identify an extensive range of exchange parameters able to support $k = \{\frac{1}{2} \frac{1}{2} \frac{1}{2}\}$ partial order through a standard mean-field theory (s-MFT) study. The s-MFT free energy displays, up to quartic order in the order parameters, an emergent $O(4)$ symmetry. The transition into the POS is thus described by the Ginzburg-Landau free-energy, $\mathcal{F}$, of an $n$-component vector model ($n=4$) [46, 47]. The most relevant perturbation is a “cubic anisotropy” which breaks the $O(4)$ symmetry. We thus identify the physical origin of cubic anisotropy as thermal fluctuations beyond s-MFT. The $O(4)$ symmetry-breaking and selection of either 1- or 4- dimensional order in this model is an example of thermal OdB. We stress, however, that our problem conceptually departs significantly from the more common cases of OdB where “small” thermal or quantum fluctuations are typically considered. This is valid for temperatures much lower than the critical temperature where the harmonic approximation is often justified. For example, in Gd$_2$Ti$_2$O$_7$, where another transition occurs at $T \sim 0.7$ K $< T_c$ [27, 32, 33, 35, 36], the low temperature fluctuations have no bearing on the state-selection at $T_c$.

Given the above considerations, we develop an extension of the Thouless-Anderson-Palmer (TAP) method, E-TAP, to obtain the phase diagram of 1-k and 4-k orders. We use Monte Carlo simulations to confirm the E-TAP predictions that 1-k and 4-k POSs are selected in different portions of the phase diagram. We suggest that the phase transition at $T_c \sim 1$ K in Gd$_2$Ti$_2$O$_7$ belongs to the above $n = 4$ cubic universality class. From a broader methodological perspective, our E-TAP method for spin models with anisotropic interactions could be applied to other problems in frustrated magnetism where the question of state selection at $T_c$ is of interest.

**Model** – We consider the general Hamiltonian, $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{dip}} + \mathcal{H}_2 + \mathcal{H}_3$, for classical spins, $S_i$, on the pyrochlore lattice:

$$\mathcal{H}_0 \equiv \sum_{\langle i,j \rangle} \left[ J_{\text{Ising}} S_i \cdot S_j + J_{\text{DM}} \hat{d}_{ij} \cdot (S_i \times S_j) + J_{\text{pd}} S_i^a A_{ij}^{(\alpha \beta)} S_j^\beta \right].$$

$$\mathcal{H}_{\text{dip}} = \sum_{i>j} S_i^a A_{ij}^{(\alpha \beta)} \delta_{ij}^\alpha \delta_{ij}^\beta.$$  

$$\mathcal{H}_2 \equiv \sum_{\langle\langle ij \rangle\rangle} J_2 S_i \cdot S_j,$$

where $\mathcal{H}_0$ includes all possible symmetry-allowed nearest-neighbor (n.n.) bilinear interactions: isotropic ($J$), Dzyaloshinskii-Moriya (DM) ($J_{\text{DM}}$), Ising ($J_{\text{Ising}}$) and pseudo-dipolar ($J_{\text{pd}}$) [48, 49]. Of exchange origin, all these couplings can be positive or negative. Unit vectors $\hat{d}_{ij}$ are chosen such that positive and negative $J_{\text{DM}}$ correspond to direct and indirect DM interactions, respectively [50]. $\delta$ is the local cubic [111] direction at site $i$ and $A_{ij}^{(\alpha \beta)} \equiv \langle \delta_{\alpha \beta} r_{ij}^3 - 3r_{ij} r_{ij} r_{ij}^2 \rangle$. $\mathcal{H}_{\text{dip}}$ is the long-range magnetostatic dipole-dipole interaction with strength $J_{\text{dip}} = \frac{\mu_0 r_n^3 a^2}{4 \pi m_n}$, where $r_m$ is the nearest-neighbor distance. $r_n$ is measured in units of $r_m$, and $\mu$ is the magnetic moment. Our convention for $\mathcal{H}_0$ [48, 49] differs from that used by other groups [51, 52] but the two are related by a linear-transformation. $\mathcal{H}_2$ and $\mathcal{H}_3$ are second and third n.n. exchange interactions, respectively.

We begin by studying $\mathcal{H}_0$ using s-MFT [9, 23, 53, 54]. For a large region of parameter space, a degenerate line of modes with momenta $\{hhh\}$ first becomes critical at $T_c$ [21, 23–25]. s-MFT calculations show that $\mathcal{H}_{\text{dip}}$ lifts the degeneracy by weakly selecting soft modes at the four $L = \{\frac{1}{2} \frac{1}{2} \frac{1}{2}\}$ points within the first Brillouin zone, which we label $k_a$ ($a = 0, 1, 2, 3$) [23, 24]. Other perturbations to $\mathcal{H}_0$, such as $\mathcal{H}_2$ and $\mathcal{H}_3$, can have similar effects [23–26, 53]. Here, we focus on $\mathcal{H}_{\text{dip}}$ since it can be of prominence in rare-earth pyrochlore oxides [19]. In particular, we choose the Gd$_2$Ti$_2$O$_7$ value $J_{\text{dip}}/J \sim 0.2$ [21] as an exemplar. For completeness, and of possible relevance to magnetic pyrochlores with ions having a small magnetic moment, we present some results in the Supplemental Material (Sup. Mat.) [53] that use the notation of Ref. [52] for $\mathcal{H}_0$ and where the $\{hhh\}$ degeneracy-lifting originates from $\mathcal{H}_2$ as opposed to $\mathcal{H}_{\text{dip}}$.

The direct space spin configurations corresponding to a 1-k state, with $k = k_0 \equiv \{\frac{1}{2} \frac{1}{2} \frac{1}{2}\}$, is illustrated in Fig. 1a. Denoting the corresponding spin direction at site $r$, as $\hat{e}_{k_0}(r)$, with $\hat{e}_{k_a}(r_i)$ ($a = 1, 2, 3$) defined similarly [53], we introduce the order parameter $\psi_a$ for a particular ordering wave vector $\mathbf{k}_a$ as $\psi_a = \frac{1}{N} \sum_{r} S_i \cdot \hat{e}_{k_a}(r_i)$, where the summation goes over all $N$ sites of the pyrochlore lattice and $\psi = \{\psi_0, \ldots, \psi_3\}$ defines a 4-dimensional vector.

For a wide range of ($J$, $J_{\text{DM}}$, $J_{\text{Ising}}$, $J_{\text{pd}}$, $J_{\text{dip}}$, $J_2$, $J_3$) couplings, s-MFT predicts a second order transition with ordering at momenta $k_a$. Interestingly, we find that up to quartic order in the $\{\psi_a\}$ order parameters, the s-MFT-free energy displays an emergent $O(4)$ symmetry [53]. While the $O(4)$ symmetry is spoiled at higher order in $\{\psi_a\}$, no selection of 1-k (which has only one $\psi_a \neq 0$) vs 4-k (which has four $\psi_a$ with the same nonzero amplitude) occurs at any order [53].

The observation of an emergent $O(4)$ symmetry at quartic order in s-MFT helps us to recognize that a “cubic anisotropy” in the $\{\psi_a\}$ is the most relevant symmetry-allowed perturbation. We then write the Ginzburg-Landau free energy $\mathcal{F}(\{\psi_a\})$ [53] of the system, up to quartic order in the $\{\psi_a\}$, that includes a cubic anisotropy term ($v \neq 0$):

$$\mathcal{F} = \frac{1}{2} \sum_a \psi_a^2 + u \left( \sum_a \psi_a^2 \right)^2 + v \sum_a \psi_a^4.$$  

(2)
Eq. (2) is the celebrated n-vector model \((n = 4)\) with cubic anisotropy \([46]\) with \(r = a_0(T - T_c)\) and \(a_0 > 0\). This model, a cornerstone of the theory of critical phenomena \([46, 47]\), has been extensively investigated with numerous methods \([55–58]\). In 3\(D\), for \(n > n_c \approx 2.89\) and \(u > 0\) \([58]\), the model undergoes a second-order transition into a phase where all \(\psi_n\) have the same amplitude if \(v > 0\). The universality class is controlled by the cubic fixed point \([46]\) with distinct critical exponents from those of the isotropic \(O(n)\) fixed point \([47, 58]\). For \(v < 0\), the phase transition is a fluctuation-induced first-order transition to a state with only one nonzero \(\psi_n\). Therefore, we conclude that for \(v > 0\) the magnetic order is defined by a superposition of four \(k_0\) spin states (4-\(k\) state) while for \(v < 0\) the order is defined by a single \(k_0\) spin structure (1-\(k\) state). Both states are POSs since the spins on 1/4 of the sites remain disordered (see Fig. 1). As stated above, s-MFT predicts that 1-\(k\) and 4-\(k\) have the same \(F\) (i.e. \(v = 0\)). We thus identify thermal fluctuations as the mechanism for generating \(v \neq 0\) leading to a selection of 1-\(k\) vs 4-\(k\). To expose how fluctuations may lead at the microscopic level to a selection directly at \(T_c\), we devise and then use an E-TAP method \([59–61]\).

**Extended TAP Method** – A magnetic moment at a particular lattice site experiences a local field due to its neighbors. At the s-MFT level, the presence of the spin at the site of interest affects its local field indirectly. This is an artifact of s-MFT. The Onsager reaction field (ORF) introduces a term that cancels this unphysical effect. The TAP approach provides a systematic way to implement the ORF correction \([62]\). In this work, we devise an extension of the TAP method, E-TAP, where the ORF is the first term of a series originating from nonzero on-site fluctuations, e.g. \(\langle S_i^S S_i^H \rangle \neq \langle S_i^S \rangle \langle S_i^H \rangle\) \([53, 60, 61]\).

To compute the E-TAP corrections, we consider a perturbative expansion of the Gibbs free-energy, \(G\), in inverse temperature \(\beta\) \([53, 60]\):

\[
\beta G = -\ln \left( \text{Tr}[\exp(-\beta H + \sum_i \lambda_i \cdot (S_i - m_i))] \right). \tag{3}
\]

Here \(m_i\) is the local magnetization and the \(\lambda_i\) vector is a Lagrange multiplier; different from the local mean field at site \(i\) by a factor of \(\beta\) \([53, 60]\). Defining \(\tilde{G}(\beta) \equiv \beta G\), the first and second terms in the expansion, \(\tilde{G}(0)/\beta\) and \(\tilde{G}'(0)\), where the prime represents differentiation with respect to \(\beta\), are the s-MFT entropy and energy, respectively. The third term, \(\Omega \equiv \tilde{G}''(0)/\beta^2/2\), is the first correction beyond s-MFT, arising from fluctuations \([53, 60]\):

\[
\Omega = -\frac{\beta}{4} \sum_{i,j} \sum_{\alpha,\beta,\gamma,\delta} J_{ij}^{\alpha\beta} j_{ij}^{\gamma\delta} \chi_{ij}^{\alpha\beta} \chi_{ij}^{\gamma\delta}. \tag{4}
\]

Here, \(\chi_{ij}^{\alpha\beta} = \langle S_i^\alpha S_j^\beta \rangle - \langle S_i^\alpha \rangle \langle S_j^\beta \rangle\) is the on-site susceptibility. Since \(\chi_{ij}^{\alpha\beta}\) is quadratic in \(\{\psi_a\}\) \([53]\), \(\Omega\) in Eq. (54) is therefore of quartic order in \(\{\psi_a\}\). The first order E-TAP correction, \(\Omega\), can thus, in principle, generate a finite cubic anisotropy term in \(F\) and select 1-\(k\) or 4-\(k\) depending on the bilinear spin-spin interaction matrix \(J_{ij}^{\alpha\beta}\) defined through

\[
\mathcal{H} = \sum_{(i>j)} \alpha,\beta J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta
\]

in Eq. (1). We calculate \(\Omega\) \([53]\) for the region in parameter space with \((1/2, 1, 1)\) ordering (the dark-shaded 1-\(k/4-k\) wedge in Fig. 2). Specifically, we compute \(\delta \Omega \equiv \Omega_{1k} - \Omega_{4k}\). \(\delta \Omega > 0\) indicates a 1-\(k\) selection and conversely for \(\delta \Omega < 0\), with the \(\Omega\) contribution splitting the wedge into 1-\(k\) and 4-\(k\) sectors. In particular, for a dipolar Heisenberg model with \(J_{\text{sing}} = J_{\text{DM}} = J_{\text{pol}} = 0\), relevant to Gd\(_2\)Ti\(_2\)O\(_7\) \([21]\), E-TAP calculations predict a 4-\(k\) state selection at \(T_c\), as observed in this compound \([20]\).

**Monte Carlo Simulations** – We performed parallel tempering classical Monte Carlo simulations to check the E-TAP predictions. We pick, somewhat arbitrarily, two sets of interaction parameters corresponding to the 1-\(k\) and 4-\(k\) regions in Fig. 2. One set is the simplest model for a spin-only \(^5\)\(S_{7/2}\) \((L = 0, S = 7/2, J = L + S)\) state for Gd\(^{3+}\) in Gd\(_2\)Ti\(_2\)O\(_7\), with \(J_{\text{dip}}/J = 0.18\) \([21]\). We include, as in a previous Monte Carlo work \([25]\), weak ferromagnetic second n.n. interaction (e.g. \(-0.02J < J_2 < 0\)) to stabilize a temperature range wide enough to numerically resolve \((1/2, 1, 1)\) order \([25, 53]\). We measure the magnitude of \(\psi\):

\[
|\psi|^2 = \langle \sum_{a=0}^3 \psi_a^2 \rangle, \tag{5}
\]

where \(\langle \ldots \rangle\) denotes a thermal average.

In addition to its magnitude, we identify the orientation of \(\psi\) at \(T < T_c\) through two additional order parameters, \(d_{ik}\) \((i = 1, 4)\) defined below. On the surface of a four-dimensional unit hyper-sphere, there are eight “1-\(k\) points” corresponding.
to 1-\(k\) states including \((\pm 1, 0, 0, 0), (0, \pm 1, 0, 0)\), etc. Similarly, there are sixteen “4-\(k\) points” on the hyper-sphere [53]. Given a certain spin configuration, we calculate \(\hat{\psi} = \psi / |\psi|\). The \(d_{ik}\) (\(i = 1, 4\)) are then defined as the minimum Euclidean distance between point \(\hat{\psi}\) and all of the \(i-k\) points. The thermal average of \(d_{1k}\) (\(d_{4k}\)) is expected to decrease if the system enters a 1-\(k\) (4-\(k\)) state for \(T < T_c\).

The results for two sets of interaction parameters in the 1-\(k\) and 4-\(k\) regions of Fig. 2 are shown in the left and right columns of Fig. 3, respectively. The growth of \(|\psi|^2\) at \(T_c / J \sim 5.5\) and \(T_c / J \sim 0.15\) shows that the system orders with \(k = (\frac{1}{2} \frac{1}{2} \frac{1}{2})\). In the left column, the 1-\(k\) state is selected at \(T_c\), as indicated by a minimum for \(d_{1k}\) and a maximum for \(d_{4k}\). The system orders in a 4-\(k\) state in the right column. The separation of \(d_{1k}\) and \(d_{4k}\) for both cases in Fig. 3 accentuates as the linear dimension \(L\) of the system increases, indicating that the selection of either 1-\(k\) or 4-\(k\) survives in the thermodynamical limit. These results are consistent with the predictions from the E-TAP calculations at \(T_c\). Unfortunately, the large computational resources required for simulations with long-range dipolar interactions prevent us from investigating the order of the phase transitions in Fig. 3.

The kinks in \(|\psi|^2\) and merging of \(d_{1k}\) and \(d_{4k}\) indicate the system enters into a distinct phase at \(T / J \lesssim 4\) and \(T / J \lesssim 0.1\) in the left and right columns of Fig. 3. Since in a 2-\(k\) state \(d_{1k} \equiv d_{4k} = \sqrt{2 - \sqrt{2}} \approx 0.765\) [53], the results for \(d_{4k}\) suggest that the low-temperature region may be a 2-\(k\) state. We note that the latest single-crystal neutron diffraction results [63] indicate that the low-temperature state \((T < 0.7\) K) of \(\text{Gd}_2\text{Ti}_2\text{O}_7\) may not be the previously suggested 4-\(k\) structure [20]. The results of a more in-depth numerical investigation of the low-temperature regime will be reported elsewhere.

**Discussion** – Considering a general symmetry-allowed anisotropic Hamiltonian, we found that \(k = (\frac{1}{2} \frac{1}{2} \frac{1}{2})\) partial order can occur over a wide range of anisotropic exchange and long-range dipolar interactions in pyrochlore magnets. We argued that fluctuations beyond s-MFT are responsible for the stabilization of a 1-\(k\) or 4-\(k\) partially ordered structure. This conclusion is based on results from E-TAP calculations where on-site fluctuations are included. We used Monte Carlo simulations to illustrate that different values of the magnetic exchange interactions can, as anticipated on the basis of the E-TAP calculations, lead to either 1-\(k\) or 4-\(k\) order.

From our work, we have exposed a likely mechanism for the establishment of 4-\(k\) order in \(\text{Gd}_2\text{Ti}_2\text{O}_7\) below its paramagnetic transition [20]. Further quantitative progress on this problem will require a better estimate of the material exchange parameters [48, 52, 64]. From this work we conclude that the transition from the paramagnetic state to the 4-\(k\) phase should be second order and belong to the 3D \(n = 4\) cubic universality class. Experimental evidence [33] suggests that this transition is second order in \(\text{Gd}_2\text{Ti}_2\text{O}_7\). Determining the critical exponents for this system could confirm our prediction but will be a challenge, given that the exponents for the 3D \(n = 4\) cubic and 3D Ising, XY and Heisenberg universality classes are proximate to one another [47, 58].

Finally, the E-TAP method for frustrated magnets formulated here could prove useful for other systems where state selection at \(T_c\) is an open question. This is particularly so if the low-temperature state selected via (thermal or quantum) ObD is separated by a phase transition from the state selected at \(T_c\). In such a case, an understanding of ObD at \(T = 0^+\) can not be leveraged to explain the selection at \(T_c\). Examples include the transition to long-range order in the pyrochlore Heisenberg antiferromagnet with indirect DM interactions [50, 65, 66], the problem of magnetization direction selection in face-centered cubic dipolar ferromagnets [67, 68] and the topical issue of state selection in XY pyrochlore antiferromagnets [45, 49, 64, 69–73].

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**DERIVATION OF THE GINZBURG-LANDAU FREE ENERGY**

To proceed, we first recall the definition of the order parameters of the theory. With the spin direction at site \( r_i \) given by \( \mathbf{e}_{k_a}(r_i) \) (Table I), we introduce the order parameter \( \psi_a \) for a particular ordering wavevector \( k_a \) as:

\[
\psi_a = \frac{1}{N} \sum_i S_i \cdot \mathbf{e}_{k_a}(r_i). \tag{6}
\]

The Ginzburg-Landau free energy is constructed in terms of the four order parameters \( \psi_a \) \((a = 0, 1, 2, 3)\). Each 1-\( k \) order is a spin-density wave and \( \psi_a \) is the amplitude of the wave. The four order parameters \( \psi_a \) form the components of a four-dimensional vector which fully describes the long-range order at the four \( \{ 111 \} \) momenta.

To construct the free energy, we study how \( \psi_a \) transforms under the space group symmetry of the pyrochlore lattice. Based on the real space spin configurations illustrated in Fig. 1 of the main text, we obtain how \( \psi_a \) transforms. Under three fold rotations about the local \([111]\) direction of, say, the 0th sublattice (see Eq. (10)), three of \( \psi_a \)'s are permuted: \( \psi_1 \rightarrow \psi_2, \psi_2 \rightarrow \psi_3 \) and \( \psi_3 \rightarrow \psi_1 \). Under a primitive FCC lattice translation, \( r \rightarrow r + \hat{x}/2 + \hat{y}/2 \) for example, two \( \psi_a \)'s, in this case \( \psi_0 \) and \( \psi_3 \), reverse sign. All \( \psi_a \)'s reverse sign under time-reversal transformation.

Lastly, the point inversion about a site on sublattice \( a \) reverses the sign of \( \psi_a \) while leaving other three order parameters intact. Since the symmetry operation is responsible for the elimination of an additional term in free energy invoked in Ref. [25], we demonstrate its consequence in detail here. Considering the 1-\( k \) structure with momentum \( k_0 \), the point inversion about a site on sublattice \( 0 \) exchanges the spin configurations on adjacent kagome layers, which have opposite directions. As a result, \( \psi_0 \rightarrow -\psi_0 \). On the other hand, the point inversion about a site on any other sublattice than \( 0 \) leaves the 1-\( k \) structure of \( k_0 \) intact. In other words, such a transformation leaves \( \psi_0 \) unchanged. Similar arguments can be made for \( k_a \) with \( a = 1, 2, 3 \).

We now construct the Ginzburg-Landau free energy to quartic order. At the quadratic order, the only invariant term is \( \sum_{a=0}^{3} \psi_a^2 \). At quartic order, there are two terms that are invariant under the above symmetry transformations: \( \sum_{a=0}^{3} \psi_a^4 \) and \( \sum_{a=0}^{3} \psi_a^3 \). We note that the quartic term of the form

**SUPPLEMENTAL MATERIAL**

In this Supplementary Material we provide the reader with some of the technical details to assist with the reading of the main body of the paper. In Section I, we derive the Ginzburg-Landau theory presented in the main text. In Section II, we show that the two types of partially ordered states, 1-\( k \) and 4-\( k \) states, have the same free energy in standard mean-field theory (s-MFT) without the presence of crystal electric field. Then, in Section III, we show that the Ginzburg-Landau theory has an emergent \( O(4) \) symmetry at the s-MFT level. In Section IV, we present the details of the extended TAP method (E-TAP) used in deriving the on-site fluctuation corrections needed to go beyond s-MFT. In Section V, we go over the technical details of our Monte Carlo simulations. Finally, in Section VI, we consider the problem of \( \{ hhh \} \) degeneracy-lifting when the selection of \( \{ \frac{1}{2} \frac{1}{2} \frac{1}{2} \} \) is induced by second nearest-neighbor exchange, \( \mathcal{H}_2 \), as opposed to long-range dipolar interaction (\( \mathcal{H}_{\text{dip}} \)). For completeness sake, we consider a formulation of the spin Hamiltonian with anisotropic exchange couplings as expressed in Ref. [52, 74–76].
\[ \psi_0 \psi_1 \psi_2 \psi_3 \] invoked in Ref. [25] is odd under the aforementioned point inversion symmetry and this term should not be present in the free energy. Given these considerations, we write down the Ginzburg-Landau free energy:
\[ F = \frac{1}{2} \sum_{\alpha=0}^{3} \psi_\alpha^2 + u \left( \sum_{\alpha=0}^{3} \psi_\alpha^2 \right)^2 + v \sum_{\alpha=0}^{3} \psi_\alpha^2. \] (7)

In this work, we make use of the following quantities:
\[ \psi \equiv (\psi_0, \psi_1, \psi_2, \psi_3), \] (a)
\[ |\psi| = \sqrt{\sum_{\alpha=0}^{3} \psi_\alpha^2}, \] (8b)
\[ \hat{\psi} \equiv \frac{\psi}{|\psi|}. \] (8c)

Here, \( \psi \) is the 4-component vector order parameter and \(|\psi|\) is its magnitude. \( \hat{\psi} \) is a 4-component unit vector parallel to \( \psi \) which is convenient to parametrize the behavior of the system below the critical temperature, \( T_c \), in the Monte Carlo simulations discussed in Section V below.

### FREE-ENERGY DEGENERACY OF 1-k AND 4-k AT THE S-MFT LEVEL

In this section we show that, at the s-MFT level, the 1-k and 4-k states have the same free energy with and without considering the effect of crystal electric field in pyrochlore lattice.

#### No Crystal Electric Field

We begin by defining the orientation of the moments, \( \hat{e}_\alpha \), on a typical tetrahedron that makes up a 1-k state. Here, \( \alpha = 0, \cdots , 3 \) corresponds to one of the four bases (sublattices) of the pyrochlore structure (see Eq. (10)) and \( k_a \) refers to ordering wave vector of the corresponding 1-k state. \( \hat{e}_\alpha \)'s are given in Table I and are expressed in global Cartesian coordinates.

| \( k_a \) | sublattice 0 | sublattice 1 | sublattice 2 | sublattice 3 |
|-------|-------------|-------------|-------------|-------------|
| \( k_0 \) | (0, 0, 0) | \( \frac{1}{2} \) | (0, \( \frac{1}{2} \)) | (0, \( \frac{1}{2} \)) |
| \( k_1 \) | (0, \( \frac{1}{2} \), 0) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |
| \( k_2 \) | (0, \( \frac{1}{2} \), \( \frac{1}{2} \)) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |
| \( k_3 \) | (0, \( \frac{1}{2} \), \( \frac{1}{2} \)) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |

TABLE I. Spin configurations of a single tetrahedron in 1-k states

The orientation of the rest of the moments in a 1-k state can be obtained from a single tetrahedron configuration using the following equation:
\[ \hat{e}_\alpha (r_i) = \hat{e}_\alpha (k_a) \cos (k_a \cdot R_i) \] (9)

where \( R_i \equiv r_i - r^\alpha \) are the FCC lattice position vectors, \( r_i \) are pyrochlore lattice position vector. The vector \( r^\alpha \) specifies the bases’ positions in pyrochlore lattice and are given by:
\[ r^0 = (0, 0, 0) \] (10a)
\[ r^1 = \left( \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right) \] (10b)
\[ r^2 = \left( \frac{1}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}} \right) \] (10c)
\[ r^3 = (0, 1, \frac{1}{\sqrt{2}}). \] (10d)

The above coordinates are expressed in units of \( r_{nn} \), the nearest-neighbor distance in pyrochlore structure. The spin orientations corresponding to the 4-k states can be expressed in terms of linear combinations of the four 1-k states:
\[ \hat{e}_{4k}(r_i) \equiv \sum_{\alpha=0}^{3} \hat{\psi}_\alpha \hat{e}_\alpha (r_i). \] (11)

Here \( \hat{\psi}_\alpha = \pm 1/2 \) conveniently keeps \( \hat{e}_{4k}(r_i) \) normalized (except for the sites with zero moments) and generate all possible 4-k states.

Having defined the 1-k and 4-k states, we now proceed to show that both the 1-k and 4-k states have the same s-MFT free energy. The general form of the s-MFT free energy for a classical 3-component spin, \( S_i \) at site \( i \) reads[23]:
\[ F = -\frac{1}{2} \sum_{i,j} m_i^\mu J_{ij}^{\mu\nu} m_j^\nu - \frac{1}{\beta} \sum_i \ln \left( 4 \pi \sinh (\beta h_i) \right). \] (12)

\( J_{ij}^{\mu\nu} \) is the bilinear spin-spin exchange coupling matrix defined through the Hamiltonian of Eq. (1) in the main text. The mag-

```markdown
| \( k_a \) | sublattice 0 | sublattice 1 | sublattice 2 | sublattice 3 |
|-------|-------------|-------------|-------------|-------------|
| \( k_0 \) | (0, 0, 0) | \( \frac{1}{2} \) | (0, \( \frac{1}{2} \)) | (0, \( \frac{1}{2} \)) |
| \( k_1 \) | (0, \( \frac{1}{2} \), 0) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |
| \( k_2 \) | (0, \( \frac{1}{2} \), \( \frac{1}{2} \)) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |
| \( k_3 \) | (0, \( \frac{1}{2} \), \( \frac{1}{2} \)) | (0, 0, 0) | (0, 0, 0) | (0, 0, 0) |
```

TABLE I. Spin configurations of a single tetrahedron in 1-k states
nentic moment, $m_i$ for both 1-$k$ and 4-$k$ states at site $r_i$ is obtained from the s-MFT self-consistent (Langevin function) equation,

$$m_i = -\frac{h_i}{\hbar_i} [\coth(\beta h_i) - \frac{1}{\beta h_i}]$$

where

$$h_i \equiv |h_i| = \sum_{j,\nu} J_{ij} m_{j\nu}^\nu.$$  \hspace{1cm} (14)

In what follows, we compare the free energy, $F$, within standard mean-field theory (s-MFT), of a 1-$k$ state, with 4-component order parameter, $\psi = (|\psi|, 0, 0, 0)$, with the free energy of a 4-$k$ state, with 4-component order parameter $\psi = (|\psi|/2, |\psi|/2, |\psi|/2, |\psi|/2)$. We thus have $m_i = |\psi|$ which is also confirmed numerically by solving the Eq. (13).

We define for a 1-$k$ state:

$$m_i = |\psi| \hat{e}_{k_a}(r_i) \equiv m_{k_a}(r_i)$$

while we have for a 4-$k$ state:

$$m_i = |\psi| \hat{e}_{4k}(r_i) = \sum_{a=1}^{4} \hat{\psi}_a m_{k_a}(r_i) \equiv m_{4k}(r_i).$$

In Eq. (12), we are employing implicit summation convention for repeated Greek superscripts $\mu$ and $\nu$ which represent Cartesian coordinates.

We now show that both terms in Eq. (12) are the same for 1-$k$ and 4-$k$ states and thus, at a given temperature the free-energy of 1-$k$ and 4-$k$ is the same at the s-MFT level.

**First Term of Eq. (12):** --- The first term, $F_1 \equiv -\frac{1}{2} \sum_{i,j} m_i^\mu J_{ij} m_j^\nu$, reads for a typical 1-$k$ state:

$$F_1(1k) \equiv -\frac{1}{2} \sum_{i,j} m_{k_a}(r_i) J_{ij} m_{k_a}(r_j)$$

Using Eqs. (9, 15) we have:

$$F_1(1k) = -\frac{1}{2} \sum_{\alpha,\beta} m_{k_a}(r^\alpha) J_{\alpha\beta}(k_a) m_{k_a}(r^\beta).$$

where $\alpha$ and $\beta$ are sublattice labels and $J_{\alpha\beta}(k_a)$ is the Fourier transform of $J_{ij}$. Similarly, using Eqs. (16, 17), $F_1$ for a 4-$k$ state can be written as:

$$F_1(4k) = -\frac{1}{2} \sum_{i,j} \sum_{a,b} \hat{\psi}_a \hat{\psi}_b m_{k_a}(r_i) J_{ij} m_{k_a}(r_j).$$

Using Eq. (9) we obtain:

$$F_1(4k) = -\frac{1}{2} \sum_{a,b,\alpha,\beta} \hat{\psi}_a \hat{\psi}_b m_{k_a}(r^\alpha) J_{\alpha\beta}(k_a) m_{k_a}(r^\beta) \delta_{ab}.$$  \hspace{1cm} (20)

Consequently,

$$F_1(4k) = -\frac{1}{2} \sum_{\alpha,\beta} (\hat{\psi}_a)^2 m_{k_a}(r^\alpha) J_{\alpha\beta}(k_a) m_{k_a}(r^\beta)$$

and using Eq. (18), we find:

$$F_1(4k) = \sum_{\alpha} (\hat{\psi}_a)^2 F_1(1k).$$

(22)

Considering that for all $a$, $\hat{\psi}_a = \pm \frac{1}{2}$, and all $F_1(1k)$ are the same, we thus have:

$$F_1(4k) = F_1(1k).$$

(23)

**Second Term of Eq. (12):** --- In this term, the only variable that depends on different spin configurations is the magnitude of local field, $h_i$, in Eq. (14), which we focus on. Using Eq. (15), the local field, $h_i$, experienced by a moment at site $r_i$ in a 1-$k$ state, is given by:

$$h_{k_a}(r_i) \equiv \sum_{j,\nu} J_{ij} m_{k_a}(r_j).$$

while for a 4-$k$ state we have

$$h_{4k}(r_i) \equiv \sum_{j,\nu} J_{ij} m_{4k}(r_j) = \sum_{a} \hat{\psi}_a h_{k_a}(r_i).$$

(24)

where $m_{4k}(r_j) = \sum_a \hat{\psi}_a m_{k_a}(r_j)$. Within s-MFT, the local field at each site is antiparallel to the moment at that site. As a result, $h_{k_a}(r_i)$ can be written as

$$h_{k_a}(r_i) = C m_{k_a}(r_i).$$

(26)

Here, $C$ is the same constant for each of the 1-$k$ states with $k \equiv k_a (a = 0, 1, 2, 3)$, since all of 1-$k$ states have the same free energy by definition.

Similarly, for the 4-$k$ state, using Eqs. (25, 26), we find

$$h_{4k}(r_i) = C \sum_{a} \hat{\psi}_a m_{k_a}(r_i) = C m_{4k}(r_i).$$

(27)

Since we have $|\psi|$ the same for both 1-$k$ and 4-$k$ states by construct, Eq. (27) shows that the magnitude of the local field is the same for 1-$k$ and 4-$k$ at the lattice sites with nonzero moments at a given temperature $T$. We note that, the theory is internally consistent, since we have taken $|\psi|$ to be the same for both 1-$k$ and 4-$k$ states which is confirmed numerically as mentioned earlier. As a result, the second term in Eq. (12) is the same for 1-$k$ and 4-$k$.

Based on Eqs. (23, 27) and Eq. (12), we therefore find that the s-MFT free energy is the same for the 1-$k$ and 4-$k$ states.

**With Crystal Electric Field**

The discussion in the previous subsection, as well as in the main text, has assumed for simplicity that the spin degrees of freedom, $S_i$, at each site $i$, is a classical vector of fixed length. As a result, the s-MFT treatment leads for the self-consistent equation for $m_i$, to the Langevin function in Eq. (13). Had we assumed a quantum $S_i$, Eq. (13) would be replaced $m_i = -\frac{h \hat{S}_i}{\hbar} B_S(h_i)$, where $B_S(x) = \frac{2x+1}{2x} \coth(\frac{2x+1}{2x}) - \frac{1}{2x}$.
same constant in Eqs. (26, 27). As a result, Eq. (32) can be
trivially written as:
\[ \hat{G}(h_i, \theta_i, \phi_i), \]
where \(\theta_i\) and \(\phi_i\) are the polar and azimuthal angles of \(h_i\), in the
local \{111\} frame of coordinates. We dropped \(\beta\) from the
argument of \(\hat{G}\) for simplicity. Now we show the \(\hat{G}\) is the same for
both 1-\(k\) and 4-\(k\) states. According to Eq. (27), \(h_i\) is the same for
both 1-\(k\) and 4-\(k\) states. Considering Table I and Eq. (11), \(\forall i, \theta_i = \pi/2\) for both of these states. On the other hand, \(\phi_i\) can only have the following values: \(\{\pi/6, 2\pi/6, 5\pi/6, \ldots, 11\pi/6\}\) for both 1-\(k\) and 4-\(k\) states. Although for 1-\(k\) and 4-\(k\) states, these angles are distributed differently over the lattice, all of
them are present in a typical 1-\(k\) and 4-\(k\) states. Since there
is a sum over all lattice sites in Eq. (30), the outcome of this
equation is the same for the 1-\(k\) and 4-\(k\) states. Consequently,
including the crystal electric field in the s-MFT Hamiltonian
does not differentiate between 1-\(k\) and 4-\(k\) states, free-energy-
wise.

We numerically confirmed this result by minimizing Eq. (30)
and solving the resulting s-MFT self-consistent equation where \(\hat{S}_i\) were spin-7/2 quantum mechanical operators and \(B_{n}^{m}\) coefficients in Eq. (29), were chosen in accordance with
the experimental values for Gd$_2$Ti$_2$O$_7$ from Ref. [77]. We
found that the 1-\(k\) and 4-\(k\) states have identical paramagnetic
critical transition temperature, \(T_c\), and identical free energy below \(T_c\).

**EMERGENT O(4) SYMMETRY UP TO QUARTIC ORDER IN S-MFT**

In this section we demonstrate that the free energy within the
s-MFT treatment is \(O(4)\) symmetric up to quartic order in the \(\psi\)’s. To do this, we expand the s-MFT free energy, Eq.
(12), in powers of \(h_i\):
\[
F = \sum_{n=0}^{\infty} \sum_{i} \frac{1}{n!} \frac{\partial^n F}{\partial h_i^n} h_i^n
\]  
(35)
where \(h_i\) is defined in Eq. (14). Considering the pyrochlore
structure, \(\sum_i\) is equivalent to \(\sum_{\alpha=0}^{\alpha=2} R_{\alpha}\), where \(\alpha\) is the sub-
lattice label and \(R_{\alpha}\) is the FCC lattice position vector. As a result, \(h_i\) can be relabelled as \(h^\alpha(R_{\alpha})\). This notation makes the
book-keeping clearer in what follows. We will focus on the
\(n = 4\) term because, based on our Ginzburg-Landau symme-
try analysis in Section , the quadratic term is \(O(4)\) invariant.

For \(T \lesssim T_c\) and in a region of parameter space with \(\{111\\}\)
ordering wave vectors, \(h^\alpha(R_{\alpha})\) can be written as
\[
h^\alpha(R_{\alpha}) = \sum_{\alpha} \hat{\psi}^\alpha h_k(R_{\alpha}),
\]  
(36)
where \(r_t = R_{\alpha} + r^\alpha\). Using Eqs. (9, 15, 24) we have
\[
h_k(R_{\alpha}) = |h_k(R_{\alpha})| e^{i\psi_k} \cos(k_{\alpha} \cdot R_{\alpha}),
\]  
(37)
where according to the arguments presented in Section ,
\(|h_k(R_{\alpha})|\) is the same for all sites with nonzero moments. So
\(|h_k(R_{\alpha})| \equiv h\). The fourth order term in \(h_i\), arising in Eq. (35),
can be written as \(\propto \sum_{R_{\alpha}} \sum_{\alpha=0}^{\alpha=3} |h^\alpha(R_{\alpha})|^4\). We now proceed
to show that this term is \(O(4)\) invariant.

Using Eqs. (36, 37), we have:
\[
\sum_{R_i} \sum_{\alpha=0}^{3} |h^\alpha(R_i)|^4 = \sum_{R_i} \sum_{\alpha} h^4 \left( \sum_{b=0}^{3} \psi_b^2 + \sum_{b_1 < b_2} 2\hat{\psi}_{b_1} \hat{\psi}_{b_2} \left( \hat{e}_{k_{b_1}}^{\alpha} \cdot \hat{e}_{k_{b_2}}^{\alpha} \cos(k_{b_1} \cdot R_i) \cos(k_{b_2} \cdot R_i) \right) \right)^2 
\]

\[
\sum_{R_i} \sum_{\alpha=0}^{3} |h^\alpha(R_i)|^4 = \frac{Nh^4}{4} \sum_{\alpha=0}^{3} \left( \sum_{b=0}^{3} \psi_b^2 + \sum_{b_1 < b_2} \psi_{b_1}^2 \psi_{b_2}^2 \right). 
\]

\[
\sum_{R_i} \sum_{\alpha=0}^{3} |h^\alpha(R_i)|^4 = \frac{3Nh^4}{4|\psi|^4} \sum_{b=0}^{3} \psi_b^4. 
\]

where
\[
\delta S_{j}^{\nu} \equiv S_{j}^{\nu} - m_{j}^{\nu}. 
\]

It can be shown that the derivatives of \( \tilde{G}(\beta) \) with respect to \( \beta \) can be evaluated in terms of expectation value of powers of \( U[60] \). We find:
\[
\frac{\partial \tilde{G}(\beta)}{\partial \beta} = \langle H \rangle 
\]
\[
\frac{\partial^2 \tilde{G}(\beta)}{\partial \beta^2} = -\langle U^2 \rangle 
\]
\[
\frac{\partial^2 \tilde{G}(\beta)}{\partial \beta^3} = \langle U^3 \rangle 
\]

The \( \langle \cdots \rangle \) above denotes a thermal average which, for a general observable \( O \), is given by:
\[
\langle O \rangle = \frac{\text{Tr}[O \exp \left( -\beta H + \sum_i \lambda_i \cdot (S_i - m_i) \right)]}{\text{Tr}[\exp \left( -\beta H + \sum_i \lambda_i \cdot (S_i - m_i) \right)].} 
\]

According to Eq. (43), all the averages are calculated at \( \beta = 0 \). The limit \( \beta = 0 \) in Eq. (47) corresponds to the s-MFT approximation and, consequently, \( \langle \cdots \rangle \) is changed to \( \langle \cdots \rangle_{\text{MF}} \). The first two terms in Eq. (43) correspond to the s-MFT free energy while the higher order terms in \( \beta \) provide the corrections beyond s-MFT and introduce the fluctuations.

Calculating the expectation value of powers of \( U \) reduces to the evaluation of averages of the following form:
\[
\langle \delta S_{i_1}^{\alpha_1} \delta S_{i_2}^{\alpha_2} \cdots \delta S_{i_n}^{\alpha_n} \rangle_{\text{MF}} 
\]

where \( i_n \) represents the site label, \( \alpha_n \) represents a Cartesian coordinate and \( n \) is the number of \( \delta S \)’s in the Eq. (48). We henceforth drop the MF subscript for simplicity. For \( n = 1 \), the expectation value in Eq. (48) is zero due to the relation \( m_i = \langle S_i \rangle \). For \( n \geq 2 \), however, this expectation value is nonzero only if no site label appears only once. For example, averages of the following form have a nonzero contribution:
\[
\langle \delta S_{i_1}^{\alpha_1} \delta S_{i_2}^{\alpha_2} \delta S_{i_3}^{\alpha_3} \delta S_{i_4}^{\alpha_4} \delta S_{i_5}^{\alpha_5} \rangle = \langle \delta S_{i_1}^{\alpha_1} \delta S_{i_2}^{\alpha_2} \rangle \langle \delta S_{i_3}^{\alpha_3} \delta S_{i_4}^{\alpha_4} \rangle \langle \delta S_{i_5}^{\alpha_5} \rangle 
\]
The expectation values above can be calculated using the self-consistent s-MFT equation which, for the case of 3-component classical spins, is given by,

\[ m_i = -\frac{\lambda_i}{|\lambda_i|} [\coth(|\lambda_i|) - 1] \]. \hspace{1cm} (50)

Consequently

\[ \langle \delta S_i^\alpha \delta S_i^\beta \rangle = \frac{\partial m_i^\alpha}{\partial \lambda_i^\beta} = \chi_i^{\alpha\beta}, \] \hspace{1cm} (51)

\[ \langle \delta S_i^\alpha \delta S_i^\beta \delta S_i^\gamma \rangle = \frac{\partial \chi_i^{\alpha\beta}}{\partial \lambda_i^\gamma}, \] \hspace{1cm} (52)

and, generally,

\[ \langle \delta S_i^\alpha \delta S_i^\beta \cdots \delta S_i^{\alpha_n} \rangle = \frac{\partial \langle \delta S_i^{\alpha_1} \delta S_i^{\alpha_2} \cdots \delta S_i^{\alpha_{n-1}} \rangle}{\partial \lambda_i^{\alpha_n}}. \] \hspace{1cm} (53)

Referring to Eq. (43), the lowest order correction beyond s-MFT originates from the third term. Considering Eq. (46b), the result reads:

\[ \Omega \equiv \frac{\beta}{2!} \frac{\partial^2 \tilde{G}}{\partial \beta^2} \bigg|_{\beta=0} = -\beta \sum_{i=\alpha,\beta,\gamma} \sum_{\alpha,\beta,\gamma} J^\alpha_{ij} J^\beta_{ij} \lambda_i^{\alpha\beta} \lambda_j^{\gamma}. \] \hspace{1cm} (54)

In our expansion, we are interested in a temperature range close to \( T_c \). In this case, Eq. (50) can be expanded for small \( |\lambda_i| \):

\[ m_i^\alpha = -\frac{\lambda_i^\alpha}{3} \left[ 1 - \frac{|\lambda_i|^2}{15} + \frac{2}{315} |\lambda_i|^4 + \cdots \right]. \] \hspace{1cm} (55)

To calculate \( \chi_i^{\alpha\beta} \) in Eq. (51), we differentiate Eq. (55) with respect to \( \lambda_i^\alpha \). This gives:

\[ \chi_i^{\alpha\beta} = -\frac{\delta \lambda_i^{\beta}}{3} \left[ 1 - \frac{|\lambda_i|^2}{15} + \frac{2}{25} \lambda_i^{\alpha\beta} \lambda_i^{\alpha} \lambda_i^{\beta} + \cdots \right], \] \hspace{1cm} (56)

where \( \cdots \) means higher order terms. According to the general discussion pertaining to the Ginzburg-Landau theory presented in the main text, we are foremost interested in the quartic terms in \( |\lambda_i| \) and \( \lambda_i^\alpha \); these are the ones that lead to a selection between 1- and 4-k states on which we henceforth focus. We substitute Eq. (56) for \( \chi_i^{\alpha\beta} \) and \( \chi_i^{\gamma\delta} \) in Eq. (54). The following subset of terms are found to potentially be able to differentiate between 1-hop and 4-hop states at the quartic order:

\[ \Omega_1 = -\beta \frac{1}{4! \langle 45 \rangle^2} \sum_{ij} |\lambda_i|^2 |\lambda_j|^2 \sum_{\gamma\delta} J_{ij}^{\gamma\delta} J_{ij}^{\gamma\delta}, \] \hspace{1cm} (57)

\[ \Omega_2 = -\frac{1}{4 \langle 45 \rangle^2} \sum_{ij} \sum_{\alpha,\beta,\gamma,\delta} J_{ij}^{\alpha\gamma} J_{ij}^{\alpha\delta} |\lambda_i|^2 \lambda_i^{\alpha} \lambda_j^{\beta} \lambda_j^{\delta}, \] \hspace{1cm} (58)

\[ \Omega_3 = -\frac{1}{4 \langle 45 \rangle^2} \sum_{ij} \sum_{\alpha,\beta,\gamma,\delta} J_{ij}^{\alpha\beta} J_{ij}^{\alpha\delta} \lambda_i^{\alpha} \lambda_i^{\gamma} \lambda_j^{\beta} \lambda_j^{\delta}. \] \hspace{1cm} (59)

To calculate these terms, we used a computer program which directly calculates the sums by calculating \( \lambda_i \) for 1- and 4-k. \( J_{ij}^{\gamma\delta} \) is known through the Hamiltonian in Eq. (1) of the main text and the Ewald summation method has been considered to compute the contribution from the long-range dipolar interaction. The reason these terms can distinguish between 1-k and 4-k is because they can not be reduced to onsite terms, an effect that happens with the lower order terms in the expansion with respect to \( \beta \), i.e., the first two terms in Eq. (43), which constitute the s-MFT free energy. Fig. 2 in the main text presents the corrections to free energy beyond s-MFT arising from the computation of \( \sum_{i=1}^{3} (\Omega_i(1-k) - \Omega_i(4-k)) \).

**MONTE CARLO SIMULATION DETAILS**

To further investigate the 1-k versus 4-k selection mechanism in the Hamiltonian of Eq. (1) in the main text, we performed Monte Carlo simulations of three-component classical spins on the pyrochlore lattice. We used the parallel tempering[78] method to assist with thermal equilibration. The simulations were performed for three system sizes \( L = 4, 6, 8 \) where the system is composed of \( L^3 \) conventional cubic unit cells and each cell contains 16 lattice sites (pyrochlore structure). Even system sizes \( L \) are required for the simulation cell to be commensurate with the \( \frac{4}{\sqrt{2}} \) order. To account for the long-range dipolar interaction, we employed the Ewald summation method [54, 79]. For each temperature considered, about \( 10^8 \) spin flips per spin were attempted while maintaining an average acceptance of approximately 50%.

As described in the main text, we define a distance parameter, \( d_{ik}, i = 1, 4 \), in order to distinguish between the 1-k and 4-k states. To do so, we consider the 4-dimensional (4D) Euclidean space spanned by the four 1-k states. In this 4D space, there are eight points, that we denote as \( \{ \tilde{p}_{1k} \} \), corresponding to 1-k states (4 of them are defined in Table 1 and 4 are the time-reversal of those), which can be represented as:

\[ \tilde{p}_{1k} = (\pm 1, 0, 0, 0), (0, \pm 1, 0, 0), (0, 0, \pm 1, 0), (0, 0, 0, \pm 1). \] \hspace{1cm} (60)

These points reside on the 4 axes of this 4D space. Since the 4-k states are linear combinations of the 1-k states with coefficients \( \psi_a = \pm \frac{1}{2} \) (see Eq.(11)), there are sixteen points corresponding to them

\[ \{ \tilde{p}_{4k} \} = \left( \pm \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2} \right). \] \hspace{1cm} (61)

In the simulation, we measured \( \psi_a \) defined in the main text and Eq. (6):

\[ \psi_a(\tau) = \frac{1}{N} \sum_i S_i(\tau) \cdot \hat{e}_{ka}(r_i), \] \hspace{1cm} (62)

where \( \psi_a(\tau) \) corresponds to the value of \( \psi_a \) at the Monte Carlo step \( \tau \) and \( a = 0, \cdots, 3 \). \( \hat{e}_{ka}(r_i) \) is given by Eq. (9).
Consequently, the order parameter corresponding to \( \{ \frac{1}{2} \} \) ordering wave vectors, can be represented in the 4D space as:
\[
|\psi(\tau)|^2 = \left\langle \sum_{a=0}^{3} \psi(a(\tau))^2 \right\rangle_{\tau}, \quad (63)
\]
where \( \langle \cdots \rangle_{\tau} \) represents Monte Carlo average. Since we foremost care about the orientation of this vector in the 4D space, once \( |\psi(\tau)| \) acquires a finite value at \( T < T_c \), we define:
\[
\hat{\psi}(\tau) = \frac{1}{\sqrt{\sum_{a=0}^{3} |\psi(a(\tau))|^2}} (\psi(0(\tau), \psi(1(\tau)), \psi(2(\tau)), \psi(3(\tau))). \quad (64)
\]
from which we define:
\[
d\hat{1}_{k/4}(\tau) \equiv |\hat{\psi}(\tau) - \hat{p}_{1k/4}|, \quad (65)
\]
At a particular temperature, at each \( \tau \) instance, eight and sixteen values are obtained for \( d\hat{1}_{k}(\tau) \) and \( d\hat{4}_{k}(\tau) \), respectively. The minimum value among the eight/sixteen determines the \( d\hat{1}_{k}(\tau) \) and \( d\hat{4}_{k}(\tau) \). We then define \( d\hat{1}_{k} \) at temperature \( T \) as
\[
d\hat{1}_{k} = \left\langle d\hat{1}_{k}(\tau) \right\rangle_{\tau}, \quad (66)
\]
which we refer to as the distance parameter at the temperature considered. The results for \( |\hat{\psi}|^2 \) and \( d\hat{1}_{k} \) are presented in Fig. 3 of the main text. We note that the low temperature distances show a different feature from the intermediate temperature regime.

We close this section with the following observation. Consider the four-dimensional unit vector \( \hat{\psi} \). The \( 4-k \) state corresponds to \( \hat{\psi}^{(4)} = (1/2, 1/2, 1/2, 1/2) \) while the \( 1-k \) state has \( \hat{\psi}^{(1)} = (1, 0, 0, 0) \). It is straightforward algebra to show the 2-k state, \( \hat{\psi} = (1/\sqrt{2}, 1/\sqrt{2}, 0, 0) \) has \( d\hat{1}_{k} = d\hat{4}_{k} = \sqrt{2} - \sqrt{2} \). The low-temperature behavior of the \( d\hat{1}_{k} \) in Fig. 3 of the main text may therefore suggests that the system is entering a 2-k state at low-temperature.

\[
\{ \frac{1}{2} \} \text{ AND } 1-k \text{ VS } 4-k \text{ FROM SECOND NEAREST-NEIGHBOR EXCHANGE}
\]

Because they are often of considerable strength, we focused in the main body on the dipole-induced \( \{ hhh \} \) degeneracy-lifting into \( \{ \frac{1}{2} \} \). Yet, there are some rare-earth pyrochlore oxides, such as Yb\(_2\)Ti\(_2\)O\(_7\) and Pr\(_2\)(Sn,Zr)\(_2\)O\(_7\), where the magnetic moment is sufficiently small that the dipolar interactions, \( \mathcal{H}_{\text{dip}} \), may not be the leading perturbation to \( \mathcal{H}_0 \). Rather, the second nearest-neighbors interactions, \( \mathcal{H}_2 \) might well be. Furthermore, it is of interest to consider the problem of the pyrochlore lattice with magnetic transition metal ions where dipolar interactions are definitely negligible compared to superexchange. We thus consider below the case \( J_{\text{dip}} = 0 \), but \( J_2 \neq 0 \). Also, for completeness, we consider a different, but equivalent, representation of the anisotropic exchange couplings in \( \mathcal{H}_0 \) [52].

The nearest-neighbor Hamiltonian, Eq. (1.a) in the main text, can be written in terms of local coordinates, \( (lc) \), and the couplings used in Ref. [52]
\[
\mathcal{H}_{0}^{lc} = \sum_{\langle i,j \rangle} J_{zz} S_{i}^{z} S_{j}^{z} - J_{\pm} (S_{i}^{+} S_{j}^{-} + S_{i}^{-} S_{j}^{+}) + \gamma_{ij} S_{i}^{+} S_{j}^{+} + \gamma_{ij} S_{i}^{-} S_{j}^{-} + J_{\pm} \sum_{i,j} (\zeta_{ij} S_{i}^{+} + \zeta_{ij} S_{j}^{+}) + i \leftrightarrow j \}, \quad (67)
\]
where the \( 4 \times 4 \) matrix \( \zeta_{ij} \) is given in Ref.[52] and \( \gamma = -\zeta^{*} \).

This representation has appeared in a number of recent publications on pyrochlore magnets [52, 74–76]. In what follows, first, we consider only a nearest-neighbor Hamiltonian, \( \mathcal{H}_{0}^{lc} \). We identify the critical modes at \( T_c \) [54]. These modes specify the ordering wave vector that first become soft (critical) as the temperature is decreased. As discussed in the main body of the paper, we obtain a line-degeneracy with ordering wave vectors \( \{ hhh \} \) with arbitrary \( h \) in a certain region of the parameter space colored dark in Fig. (4). Adding the beyond-nearest-neighbor interactions such as ferromagnetic second- and/or antiferromagnetic third-nearest-neighbors and/or long-range dipolar interaction, lifts this degeneracy and favors \( \{ \frac{1}{2} \} \) as an ordering wave vector [23–26].

As stated above, we considered in the main text long-range dipole-dipole interactions as a source of the selection of a \( \{ \frac{1}{2} \} \) ordering wave vector. Here, we instead consider a ferromagnetic second nearest-neighbor interaction as perturbation. We repeat all calculations leading to the computation
of the TAP correction $\Omega$ in Eq. (54). The results are illustrated in Fig. 4. In this case, we observe regions corresponding to $k = 0$ and $k = (\frac{1}{2} \frac{1}{2} \frac{1}{2})$ ordering wave vectors. Using the E-TAP method, we identify subregions corresponding to $1-k$ and $4-k$ states in the $k = (\frac{1}{2} \frac{1}{2} \frac{1}{2})$ region (see Fig. 4 caption).

The terms obtained in Eqs. (57, 58, 59) can raise/lower the free energy of the $1-k$ state with respect to $4-k$ thus breaking the degeneracy. We also, observe a region in parameter space in which the ordering wave vector is incommensurate (specified in Fig. 4). The study of this region is beyond the scope of the present work.