Ising-like order by disorder in the pyrochlore antiferromagnet with Dzyaloshinskii-Moriya interactions

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It is shown that the mechanism of order out of disorder is at work in the antisymmetric pyrochlore antiferromagnet. Quantum as well as thermal fluctuations break the continuous degeneracy of the classical ground-state manifold and reduce its symmetry to $\mathbb{Z}_3 \times \mathbb{Z}_2$. The role of anisotropic symmetric exchange is also investigated, and we conclude that this discrete-like ordering is robust with respect to these second-order-like interactions. The antisymmetric pyrochlore antiferromagnet is therefore expected to order at low temperatures, whatever the symmetry type of its interactions, in both the classical and semiclassical limits.

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I. INTRODUCTION

The role of geometrical frustration in magnetic systems is at present one of the open question in the physics of strongly correlated systems since it can lead to novel low-temperature behaviors. One of the most studied model is the nearest-neighbor antiferromagnetic Heisenberg model on the pyrochlore lattice (Fig. 1). For classical spins, the ground state is known to have a macroscopic degeneracy, preventing any magnetic ordering at $T=0$.[1] In real systems, this degeneracy is often removed, partially or totally, by additional interactions such as dipolar interactions,[2,3] second-neighbor exchange,[4] spatially anisotropic interactions,[5,6] single ion anisotropy,[7] and magnetoelastic coupling.[8,9] This explains why pyrochlore compounds often order at low temperature, but their ordering temperature is usually much smaller than the paramagnetic Curie temperature Θ_p since T_N is not related to the exchange interaction J but to the additional small interaction. Another type of process which can remove the degeneracy is order by disorder: as proposed first by Villain[10] the classical degeneracy can be lifted by fluctuations if the fluctuations around these classical ground states select one of these states. Both classical and quantum fluctuations are able to produce order by disorder through their contribution to entropy or zero-point energy, respectively [see Refs. 11–14 for examples of (spin, structural, and orbital)-fluctuation-driven degeneracy breaking in different models]. In this paper we study the effect of Dzyaloshinsky-Moriya (DM) interactions (DMIs) in the Heisenberg pyrochlore model. In an earlier paper,[15] we have shown that the $T=0$ K degeneracy is completely removed only for one particular choice of the DM vectors. Here we show that if the DM vectors are such that the $T=0$ degeneracy is not removed, both classical and quantum fluctuations select six equivalent ordered configurations, leading to a well-defined magnetic structure.

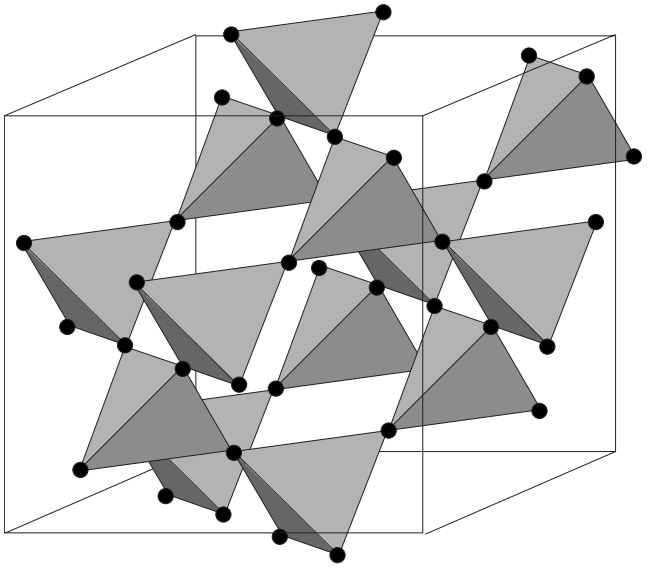

FIG. 1. The pyrochlore lattice. The interplay of the frustration of the tetrahedral unit cell with the weak connectivity of the (corner sharing) tetrahedra provides peculiar magnetic properties to the pyrochlore antiferromagnet.

II. DMI AND MORIYA'S RULES FOR THE PYROCHLORE LATTICE

Dzyaloshinsky[16] showed that, in crystals with no inversion center, the usual isotropic exchange $J\vec{S}_i \cdot \vec{S}_j$ is not the only magnetic interaction and antisymmetric exchange $\vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$ is allowed. In magnetic oxides, exchange interactions are usually attributed to superexchange mechanism, which involves virtual hopping between two neighboring magnetic ions. Taking into account Coulomb repulsion and Pauli's principle, Anderson proposed a microscopic mechanism which leads to isotropic superexchange interactions. Later, Moriya[17,18] showed that inclusion of spin-orbit coupling on the magnetic ions in first and second orders leads to antisymmetric exchange and anisotropic exchanges, respectively. Moriya's microscopic derivation[19,20] of the DMI is only valid for insulators but other possible microscopic mechanism relevant for other materials have been proposed, for instance in systems with RKKY interactions.

Whatever the microscopic origin of the DMI is, there are always symmetry constraints on the possible **D** vectors which may appear in the Hamiltonian. Indeed, the Hamiltonian must be invariant under the symmetry operations of the crystal, and this will restrict the possible **D** vectors to those for which $\vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$is invariant under these symme-

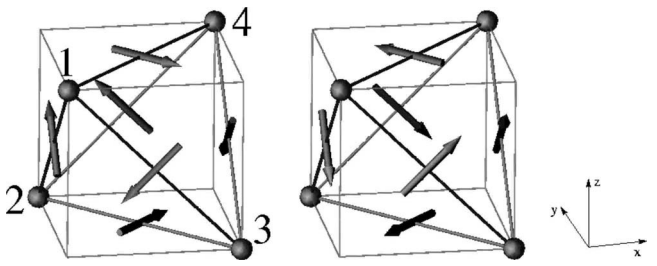

FIG. 2. **D** vectors for the DMI in the pyrochlore lattice. The convention is taken to fix the order for the cross products (always $\mathbf{D}\cdot\mathbf{S}_i\times\mathbf{S}_j$, with $j>i$). The two possible DMIs are those obtained by varying the direction of the **D** vectors ($\mathbf{D}\rightarrow-\mathbf{D}$). The case with the **D** as represented on the left is referred to as the direct case and the other case (on the right) is the indirect case. Once the DMI between two spins is fully specified, the others DMI are also fixed and obtained by applying the different $\frac{2\pi}{3}$ rotations around the cube's diagonals which leave the tetrahedron invariant. The DMIs in the rest of the lattice are also fixed and obtained by applying appropriate symmetry operations of the pyrochlore lattice.

try operations. The way of constraining the **D** vectors was known as Moriya's rules. Using these rules, we have shown[15] that two types of DM's interactions are compatible with the pyrochlore space-group symmetry. The two patterns of **D** vectors were denoted as "direct" and "indirect" cases and are described in Fig. 2.

As shown in our previous work, the classical ground state is always $q=0$. It is nondegenerate in the direct case and is described by the so-called all-in all-out magnetic phases; this case will not be discussed in this paper. From now on, we will focus on the second case of indirect DMI, where the ground-state manifold is reduced, compared to the case with no DMI, but is still continuously degenerate as shown in Ref. 15. The role of anisotropic exchange will also be discussed in this paper.

III. ROLE OF FLUCTUATIONS WITHIN THE GROUND-STATE MANIFOLD

From now on, we will focus on the second case of indirect DMI, where the ground-state manifold is $q=0$ but is continuously degenerate. There are two kinds of ground states.[15] (i) Phases can be coplanar, belonging either to the xy, yz, or zx plane and consists in perpendicular pairs of anticollinear spins (see Fig. 3) which can rotate freely within these planes. (ii) There are also noncoplanar phases, parametrized by one angle φ as follows:

$$\mathbf{S}_1=\begin{cases}\cos\,\theta\cos\left(\varphi-\dfrac{\pi}{4}\right)\\ \cos\,\theta\sin\left(\varphi-\dfrac{\pi}{4}\right)\\ \sin(\theta),\end{cases}\qquad \mathbf{S}_2=\begin{cases}\cos\,\theta\cos\left(-\varphi+\dfrac{\pi}{4}\right)\\ \cos\,\theta\sin\left(-\varphi+\dfrac{\pi}{4}\right)\\ -\sin(\theta),\end{cases}$$

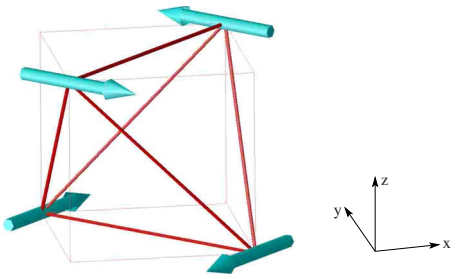

FIG. 3. (Color online) One possible ground state in the case of indirect DMI. The ground state for the whole pyrochlore lattice is a $\mathbf{q}=\mathbf{0}$ structure so that only one tetrahedron is represented. Similar structures in the zx and yz planes are degenerate.

$$\mathbf{S}_3=\begin{cases}\cos\,\theta\cos\left(-\varphi-\dfrac{3\pi}{4}\right)\\ \cos\,\theta\sin\left(-\varphi-\dfrac{3\pi}{4}\right)\\ -\sin(\theta),\end{cases}$$

$$\mathbf{S}_4=\begin{cases}\cos\,\theta\cos\left(\varphi+\dfrac{3\pi}{4}\right)\\ \cos\,\theta\sin\left(\varphi+\dfrac{3\pi}{4}\right)\\ \sin(\theta),\end{cases}$$

where the spins are labeled as in Fig. 2 and where φ and θ are *not* independent,

$$\theta=\arctan(\sqrt{2}\sin\,\varphi). \tag{1}$$

Consequently, it is possible to picture the classical ground-state manifold by following the paths defined by the arrow of say, spin 1, in all-allowed configurations (actually, fixing any spin determines the configuration of the three other spins). Such paths are depicted in Fig. 4.

The ground state is therefore a multiply arcwise connected one-dimensional manifold, represented by the thick black lines on Fig. 4. Six configurations play a particular role since they are at the intersection of two lines, i.e., they belong to both kinds of ground states defined above, where spins are coplanar, either in the xy, yz, or zx plane (see Fig. 3). In these phases, take for instance the xy plane, two spins are antiparallel [aligned along the (110) axis] and perpendicular to the two remaining spins [aligned along the $(1\bar{1}0)$ axis]. These configurations, related by time-reversal symmetry and rotations, are globally invariant under $\mathbb{Z}_3\times\mathbb{Z}_2$ and will be referred to this way from now on.

It is of practical and theoretical interest to test whether fluctuations operate on that manifold and reduce the degeneracy. Before getting into a quantitative analysis, it is worth pointing out the qualitative arguments toward an order out of disorder mechanism. The main ingredient for fluctuations to be efficient resides in the spectrum of the system: if two degenerate phases have different spectra and in particular if one has softer modes, it is common that this one is selected at low temperatures, where entropic effects and/or quantum

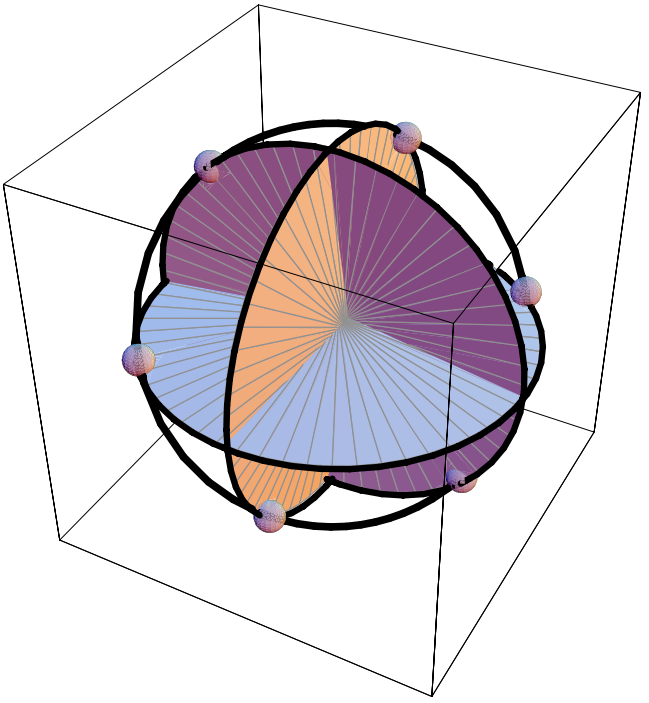

FIG. 4. (Color online) Description of the one-dimensional ground-state manifold. Any move within the ground state can be described by the possible path followed by only one of the four spins as they are all parametrized by the same angle [see Eq. (1)]. Here, only spin 1 is described. It can belong either to the *xy*, *yz*, or *zx* plane or be in a noncoplanar phase. These noncoplanar phases intersect coplanar phases at six points and the *only* way to go from one plane to another is to pass through these intersections, depicted by spheres.

fluctuations may take place. The measure of the ground-state manifold also plays an important role and should be sufficiently small. Schematically, if one phase is to be selected by fluctuations but is "drowned" in a huge ground-state manifold, the system cannot statistically pick up that phase and fluctuations will therefore not operate. In the present case, as it is shown hereafter, both preceding conditions are fulfilled: there are obviously peculiar points expected to have spectra with softer modes (zero modes actually) and the ground-state manifold, because it is one dimensional, has a null measure in configuration space and allows order out of disorder to be efficient, both at thermal and quantum levels.

A. Thermal fluctuations

As emphasized in the previous paragraph we can guess that the peculiar points of the configuration space around which thermal fluctuations could be efficient are the above mentioned six points because they are the only one within the ground-state manifold having two obvious "escape lines," e.g., two zero-energy modes. It is therefore natural to define a parameter which is maximum and equal to one only when spin configurations are among the previous six. In each of these configurations, spins all belong to the same plane, either *xy*, *yz*, or *zx*. When they belong to the *xy* plane, spins 1 and 4 are collinear as are spins 2 and 3, with the 1–4 pair

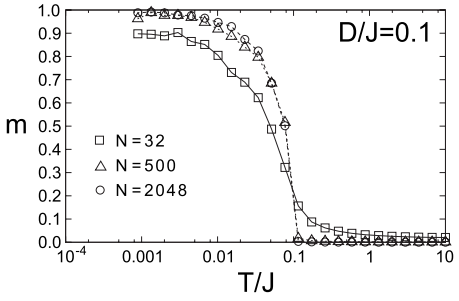

FIG. 5. Thermal average of m [see Eq. (3)] as a function of temperature for three lattice sizes, $N=32$, 500, and 2048 and for $D/J=0.1$.

being perpendicular to the 2–3 pair (see Fig. 3). The same occurs for the two other planes. Therefore, for each plane selection, a global index α can be defined to label a vector e_i^α on each site of the lattice so that the quantity

$$m_\alpha = \frac{1}{N}\sum_i^N \vec{S}_i \cdot \vec{e}_\alpha \tag{2}$$

equals ± 1 for the two time-reversal-related phases of each plane selection $\alpha = xy, yz, zx$. If the spins belong to the *xy* plane, they point along the $(1,-1,0)$ direction, thus $e_{xy} = \frac{1}{\sqrt{2}}(1,-1,0)$ and similarly for the two other planes. Defining then

$$m = \max_\alpha (m_\alpha^2), \tag{3}$$

allows characterizing any spin configuration and its proximity to one of the six peculiar phases because $M=1$ for these configurations and these configurations only.

In order to test whether thermal fluctuations may entropically select peculiar phases of the ground-state manifold, we have performed classical Monte Carlo simulations of finite-size lattices with periodic boundary conditions. The clusters we investigated had 32, 500, and 2048 sites. For each simulations, we used a single-flip metropolis algorithm with a modified update so that the acceptance rate stayed roughly above 40%, associated to a local rotation of each spin around its local molecular field. During the simulation, the autocorrelation time is calculated on the fly in order to adapt the number of Monte Carlo steps between two measures and ensure that measures are uncorrelated. Results are reported in Figs. 5 and 6, respectively, for $D/J=0.1$ and $D/J=1$. The measure of $\langle m \rangle$ as a function of the temperature clearly indicates that this parameter is an order parameter although we cannot discuss the order of the transition so far.

Both cases support that at low temperature, the system orders in one of the six previously described states as was expected from qualitative arguments. Moreover, it can be seen that T_C is of the order of D, which confirms that DMIs are responsible for this ordering.

B. Quantum fluctuations: Semiclassical approach

The same investigation has been done for quantum fluctuations. Contrary to the classical case, it is not necessary to

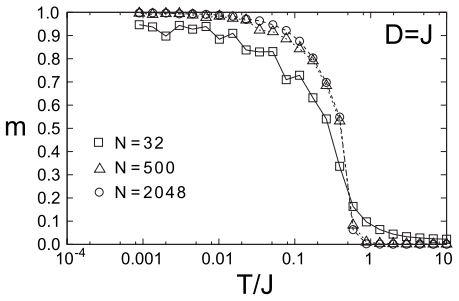

FIG. 6. Thermal average of m [see Eq. (3)] as a function of temperature for three lattice sizes, $N=32$, 500, and 2048 and for $D/J=1.0$.

guess which points will play a particular role. This is because it is possible to compute at zero temperature the energy renormalized by quantum fluctuations for all phases of the ground-state manifold. To do so, we have performed a linear spin-wave expansion of the Hamiltonian around each state of the ground-state manifold, e.g., around each state parametrized by the angle φ. The resulting bosonic Hamiltonian is then diagonalized which allows us to obtain the normal modes and calculate the renormalized energy and magnetization by quantum fluctuations.

The first result is that the $\mathbb{Z}_3\times\mathbb{Z}_2$ phases are unambiguously selected by quantum fluctuations, as shown in Fig. 7. For planar phases, they correspond to $\alpha=0$ or $\alpha=\pi$, with α being the angle of the spins with one of the six peculiar phases (two for each plane). The same calculation was performed for conic phases (indexed by φ), confirming that the six phases are selected by quantum fluctuations.

This selection is also illustrated by the computation of the renormalized magnetization, as shown in Fig. 8. As confirmed by the calculation of the energy, there are softer modes near the six selected phases. Consequently, transverse fluctuations are much stronger giving a larger renormalization of magnetization around these phases.

Whether the magnetization, which is zero for $D/J=0$, could be stabilized by taking DM's interactions into account is also of interest because it quantifies how easily this model becomes "classical." We have therefore computed for one of the six selected phases, the value of the quantum renormalized magnetization as a function of D/J. Results are reported

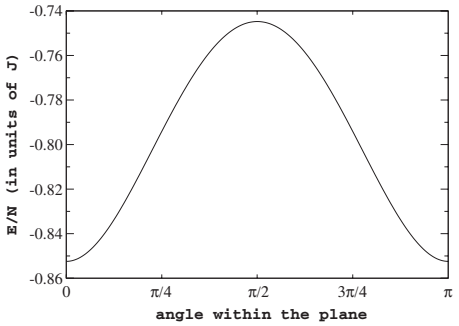

FIG. 7. Renormalized energy per site as a function of the angle α, for planar phases (all planes are equivalent), with $D/J=1$. $\alpha=0$ and $\alpha=\pi$ correspond to one time-reversal related pair of states belonging to the $\mathbb{Z}_3\times\mathbb{Z}_2$ phases.

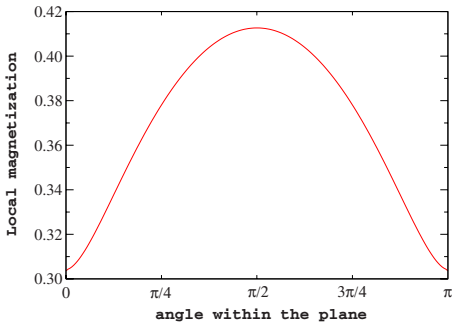

FIG. 8. (Color online) Renormalized magnetization for $S=1/2$ as a function of the angle α, for planar phases, with $D/J=1$. $\alpha=0$ and $\alpha=\pi$ correspond to one time-reversal related pair of states belonging to the $\mathbb{Z}_3\times\mathbb{Z}_2$ phases. At these two points, fluctuations have softer spectra, hence local magnetization is much more renormalized.

in Fig. 9. It is worth noting that apart from the case $S=1/2$, a very small value of D/J ($D/J\gtrsim 0.01$) stabilizes the magnetic ground state of the pyrochlore antiferromagnet. Such a result is very similar to what was obtained with the same model on the kagome lattice.[21] We must therefore emphasize that these critical thresholds, especially for low spin values, are underestimated by the semiclassical expansion, as shown by the $S=1/2$ exact diagonalization study of the antisymmetric kagome antiferromagnet.[22] What is the nature of the quantum ground state in the disordered region and whether there still is a symmetry breaking is beyond the scope of this work.

Finally, we can quantify how efficient DM's interactions are by fitting the gain in energy as a function of D/J. For small values of D/J, quantum fluctuations contribute to the renormalization of the energy as $\sqrt{D/J}$. This is an important point as discussed in Sec. IV.

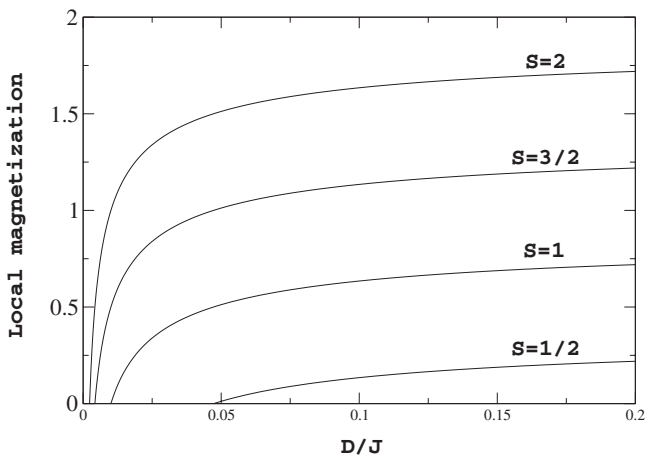

FIG. 9. Renormalized magnetization of one of the symmetry related $\mathbb{Z}_3\times\mathbb{Z}_2$ phases as a function of D/J for $S=1/2$, 1, 3/2, and 2. For $S\geq 1$, a DM interaction of 1% is sufficient to stabilize a Néel-type long-range order. From that point of view, DM interactions are very efficient in destroying the classical spin-liquid behavior and driving this model to a classical behavior.

IV. ROLE OF ANISOTROPIC SYMMETRICLIKE EXCHANGE

So far, only DM's interactions have been taken into account. Whatever the type of fluctuations, thermal or quantum, the order out of disorder mechanism is efficient and reduces the continuous degeneracy to a discrete one. But is this selection robust *vis-à-vis* next order (in spin-orbit coupling) interactions present in the Hamiltonian, the anisotropic symmetric exchange? Unfortunately, we must lose generality to address this question because many kind of symmetry-allowed interactions are possible. It is nevertheless possible to argue quantitatively on that matter.

In order to be specific, we choose to consider interactions that keep all symmetries of the pure pyrochlore lattice. They correspond to nearest-neighbor truncated dipolar interactions,[23]

$$K\frac{(\hat{n}^{\alpha}R_{ij}^{mn})(\hat{n}^{\beta}R_{ij}^{mn})}{|R_{ij}^{mn}|^5}, \tag{4}$$

where indices i,j are related to the fcc Bravais lattice while indices m,n correspond to one of the four sites of the tetrahedral unit cell. The set of unit vectors $\hat{n}^{\alpha}$ is (1,0,0), (0,1,0), and (0,0,1). We allow the sign of K to vary as there is no obvious reason why one peculiar type, $K>0$ or $K<0$, should prevail. The full Hamiltonian we are left with read now as

$$\mathcal{H}=-\frac{1}{2}\sum_{i,j}\sum_{m,n}\sum_{\alpha,\beta}\mathcal{J}(R_{ij}^{mn})^{\alpha\beta}S_i^{m,\alpha}S_j^{n,\beta}, \tag{5}$$

where $\mathcal{J}(R_{ij}^{mn})^{\alpha\beta}$ are the coefficients of the coupling matrix $\mathcal{J}(\boldsymbol{q})$ which contains the isotropic symmetric exchange $-JS_iS_j$, the antisymmetric exchange $D_{ij}S_i\times S_j$ and the anisotropic symmetric exchange K [see Eq. (4)]. Stabilized low-temperature phases are investigated through a mean-field analysis. The Hamiltonian is rewritten in reciprocal space using the following transformations:

$$S_i^{m,\alpha}=\frac{1}{\sqrt{N}}\sum_{\boldsymbol{q}}S_{\boldsymbol{q}}^{m,\alpha}e^{-\imath\boldsymbol{q}\cdot\boldsymbol{R}_i^m}, \tag{6}$$

$$\mathcal{J}(R_{ij}^{mn})^{\alpha\beta}=\frac{1}{N}\sum_{\boldsymbol{q}}\mathcal{J}_{mn}^{\alpha\beta}(\boldsymbol{q})e^{\imath\boldsymbol{q}\cdot\boldsymbol{R}_{ij}^{mn}}, \tag{7}$$

where N is the number of Bravais lattice points. The resulting interaction matrix $\mathcal{J}(\boldsymbol{q})$ is a 12×12 nondiagonal Hermitian matrix. Hence, to completely diagonalize $\mathcal{J}(\boldsymbol{q})$ one must transform the $\boldsymbol{q}$-dependent variables, $\boldsymbol{S}_{\boldsymbol{q}}^m$, to normal-mode variables. In component form, the normal-mode transformation is given by

$$S_{\boldsymbol{q}}^{n,\alpha}=\sum_{p=1}^{4}\sum_{\gamma=1}^{3}U_{n,p}^{\alpha,\gamma}(\boldsymbol{q})\phi_{\boldsymbol{q}}^{p,\gamma}, \tag{8}$$

where the indices (p,γ) label the normal modes (12 for Heisenberg spins) and $\{\phi_{\boldsymbol{q}}^{p,\gamma}\}$ are the amplitudes of these normal modes. $U(\boldsymbol{q})$ is the unitary matrix that diagonalizes $\mathcal{J}(\boldsymbol{q})$ with eigenvalues $\lambda(\boldsymbol{q})$. Finally, the mean-field free energy to quadratic order in the normal modes reads as, up to an irrelevant constant,[3]

$$\mathcal{F}(T)=\frac{1}{2}\sum_{\boldsymbol{q},p,\gamma}[nT-\lambda_p^{\gamma}(\boldsymbol{q})]|\phi_{\boldsymbol{q}}^{p,\gamma}|^2, \tag{9}$$

where $\mathcal{F}(T)$ is the mean-field free energy per unit cell, T is the temperature in units of k_B, and $n=3$ for Heisenberg spins. Therefore, the mean-field low-temperature phase is defined by the corresponding wave vector $\boldsymbol{q}_{\mathrm{ord}}$ associated with the extremal eigenvalue $\max_{p,\gamma,\boldsymbol{q}}[\lambda_p^{\gamma}(\boldsymbol{q})]$.

When $K=0$, it was shown in Sec. II that $\boldsymbol{q}_{\mathrm{ord}}=0$ and that the ground-state manifold is continuously degenerate. Including nonzero K, whatever its sign, the degeneracy is lifted already at the mean-field level. The new ground states are, for both positive and negative K, $\boldsymbol{q}=0$ slightly distorted version of the $\mathbb{Z}_3\times\mathbb{Z}_2$ states. The energy decrease, for small K, is always quadratic, $\Delta E\propto K^2$.

At this point, we can discuss whether anisotropic symmetric interactions may interfere with quantum fluctuations induced by DM's interactions. The first remark is that at the mean-field level, K breaks the degeneracy but select phases which are continuous deformation of quantum-selected phase. Therefore, we expect that taking into account anisotropic exchange would not change the behavior of this system. The second point is related to how strong K is efficient in selecting a particular phase. Suppose that the mean-field selected phase is *not* one of the $\mathbb{Z}_3\times\mathbb{Z}_2$ phases or a slightly distorted version of those. Because the energy gain is quadratic in K, it appears that quantum fluctuations are much more efficient as they induce an energy gain proportional to $\sqrt{D}$ (see Sec. III B). This clearly shows that even if anisotropic interactions are taken into account, we expect a lifting of degeneracy corresponding to the $\mathbb{Z}_3\times\mathbb{Z}_2$ phases or slightly distorted versions of those. The only mechanism which could work against such scenario would be that quantum fluctuations around K selected phases be larger than $\sqrt{D}$. Because $K\propto D^2$, this would mean that one should expect the energy gain induced by K-like interactions to behave like $\sqrt[4]{K}$, which is highly improbable.

V. CONCLUSION

This work investigates the role of thermal and quantum fluctuations on the antisymmetric pyrochlore antiferromagnet. When the symmetry type of the allowed DMI leads to a one-dimensional degenerate ground state, it is shown that these two types of fluctuations are efficient in reducing the degeneracy and drives the model to the same ordered ground state with a discrete global degeneracy of the $\mathbb{Z}_3\times\mathbb{Z}_2$ type. Including higher-order terms in the Hamiltonian the anisotropic symmetric exchange is likely to leave thermal and quantal fluctuations dominating the low-temperature behavior of this system. It is therefore concluded that whatever the type of symmetry-allowed DMI, direct or indirect, they should drive the pyrochlore Heisenberg antiferromagnet to a low-temperature ordered phase. When the model is essentially classical, the critical temperature is of order **D**. If quantum fluctuations are present, the value of the ordering tem-

perature results from a balance between the strength of the DMI and the magnitude of the fluctuations. As we have shown that for moderately large DMI the local magnetization is quite stable, the value of the ordering temperature should not be very different from the one of the classical case. Finally, it is worth pointing out that most of studied pyrochlore compounds are rare-earth oxides. It is therefore the next natural step to investigate the mechanism of DMI within 5*f* alloys and determine whether those interactions could be in some compounds large enough to drive the low-temperature behavior of these frustrated systems.

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