

# Dzyaloshinsky-Moriya induced ordering in the spin-liquid phase of the pyrochlore antiferromagnet

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We show that the S=1/2 pyrochlore lattice with both Heisenberg and antisymmetric, Dzyaloshinsky-Moriya (DM) interactions, can order antiferromagnetically into a state with chiral symmetry, dictated by the distribution of the DM interactions. This state is characterized by a small magnetic moment induced by the DM interaction. An external magnetic field can also lead to magnetic order, and a quantum transition separates the field-induced and chiral ordered phases.

*Introduction.* The behavior of many-body systems involving quantum spins has been one of the central topics in recent years since the properties of such systems are relevant to a great variety of materials, mostly oxides. The structure of the ground state and the various symmetry broken phases that emerge are issues of special interest, especially in systems of low-dimensionality and/or where frustration is present [1]. In this context the Heisenberg model on the three-dimensional pyrochlore lattice consisting of corner sharing tetrahedra, shown in Fig. 1(a), is in a league of its own. The pyrochlore lattice is strongly geometrically frustrated and is relevant to numerous compounds. It has been argued that no magnetic order is present in the ground state [2, 3, 4]. The effects of various additional interactions have also been studied, such as magnetoelastic couplings [5], long-range dipolar interactions [6], and orbital degeneracy [7]. These interactions (in addition to various anisotropies) can generally lead to bond, magnetic and/or orbital order, and which of them is dominant depends on the details of the model relevant to the specific class of materials.

In the present work we study a new mechanism for magnetic order in the S=1/2 pyrochlore lattice, driven by the Dzyaloshinsky-Moriya (DM) interactions. In the pyrochlore such interactions are always expected to be present by symmetry. For the S=1/2 Heisenberg model on the pyrochlore lattice it has been suggested [2, 4] that the ground state is dimerized (non-magnetic), but macroscopic degeneracy still remains. We show that the DM interactions lift the degeneracy, leading to a chiral antiferromagnetic state with a small magnetic moment. In an external magnetic field quantum transitions between weakly ordered states with different symmetries, depending on the field direction, are possible.

The spin Hamiltonian (S=1/2) is

$$\hat{\mathcal{H}} = \sum_{i,j} J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} \mathbf{D}_{i,j} \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (1)$$

where  $\mathbf{D}_{i,j}$  are the DM vectors, to be specified later. We start by summarizing the results for  $\mathbf{D}_{i,j} = 0$ , i.e. the Heisenberg case. Our starting point is the weak-coupling approach, similar to that of Refs. [2, 4], with the lat-

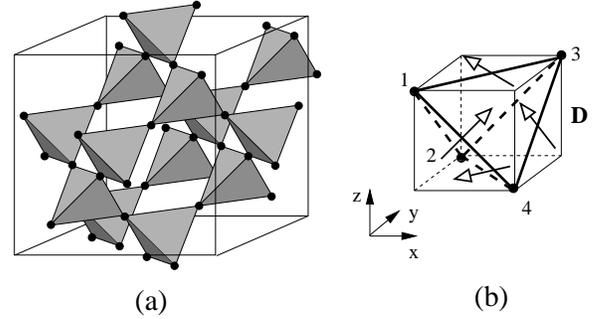


FIG. 1: (a) Pyrochlore lattice. (b) Distribution of DM vectors on a single tetrahedron (four of the six shown, see text).

tice divided into two interpenetrating sub-lattices, one of them formed by “strong” tetrahedra (with exchange  $J$ ), connected by “weak” tetrahedra (exchange  $J'$ ). The “strong” tetrahedra then form a fcc lattice, as shown in Fig. 2(a), where every site represents a tetrahedron, and one can attempt to analyze the structure of the ground state starting from the limit  $J' \ll J$ .

For  $J' = 0$  the tetrahedra are disconnected, and on a single tetrahedron the ground state is a singlet and is twofold degenerate. We choose the two ground states as:  $|s_1\rangle = \frac{1}{\sqrt{3}}\{[1, 2][3, 4] + [2, 3][4, 1]\}$ ,  $|s_2\rangle = \{[1, 2][3, 4] - [2, 3][4, 1]\}$ , where  $[k, l]$  denotes a singlet formed by the nearest-neighbor spins  $k$  and  $l$ , labeled as in Fig. 1(b). In the pseudo-spin  $T = 1/2$  representation, so that  $T_z = 1/2$  corresponds to  $|s_1\rangle$  and  $T_z = -1/2$  corresponds to  $|s_2\rangle$ , one finds that third order is the lowest one contributing to the effective Hamiltonian in the singlet sub-space:  $\hat{\mathcal{H}}_{\text{eff}} = (J^3/J^2)(1/8)[\hat{\mathcal{H}}_{\text{eff}}^{(2)} + \hat{\mathcal{H}}_{\text{eff}}^{(3)}] + \text{Const.}$ , where [8]

$$\hat{\mathcal{H}}_{\text{eff}}^{(2)} = \sum_{\langle i,j \rangle} \left\{ \Omega_{i,j}^x T_i^x T_j^x + \Omega_{i,j}^z T_i^z T_j^z + \Omega_{i,j}^{xz} (T_i^x T_j^z + T_i^z T_j^x) \right\}, \quad (2)$$

$$\hat{\mathcal{H}}_{\text{eff}}^{(3)} = \sum_{\langle i,j,k \rangle} \left\{ \frac{1}{3} T_i^z T_j^z T_k^z - T_i^z T_j^x T_k^x + \frac{T_i^z}{\sqrt{3}} (T_j^x T_k^z - T_j^z T_k^x) \right\} \quad (3)$$

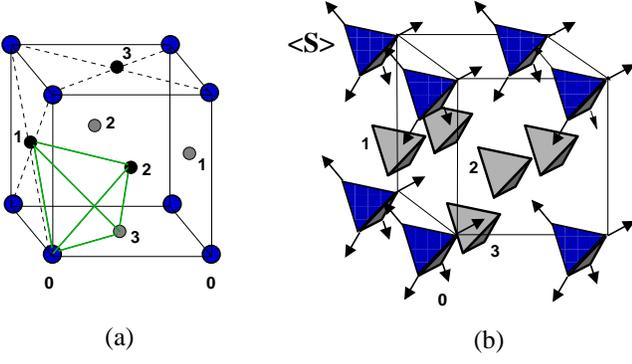


FIG. 2: (a) Fcc lattice of tetrahedra (tetrahedron = dot) with interactions  $J'$  between them. (b) Antiferromagnetic chiral order with magnetic moment  $\langle \mathbf{S} \rangle \sim \tilde{D}$ , induced by the DM interactions

In the two-body part  $\Omega_{03}^x = \Omega_{12}^x = 1/2$ ;  $\Omega_{23}^z = \Omega_{01}^z = \Omega_{02}^z = \Omega_{13}^z = 1/3$ ,  $\Omega_{03}^z = \Omega_{12}^z = -1/6$ ;  $\Omega_{23}^{xz} = \Omega_{01}^{xz} = -\Omega_{02}^{xz} = -\Omega_{13}^{xz} = 1/(2\sqrt{3})$ , and all remaining  $\Omega_{ij} = 0$ ,  $i < j$ . The site indexes refer to the fcc lattice made of individual tetrahedra, Fig. 2(a), and it is sufficient to know the interactions on one "supertetrahedron", shown in green. In the three-body interaction the indexes run over the values:  $(\mathbf{i}, \mathbf{j}, \mathbf{k}) = \{(3, 2, 1), (1, 0, 3), (2, 3, 0), (0, 1, 2)\}$ .

On a mean-field level the ground state of  $\hat{\mathcal{H}}_{\text{eff}}$  is defined by the following averages:  $\langle T_1^x \rangle = -\sqrt{3}/4$ ,  $\langle T_1^z \rangle = 1/4$ ,  $\langle T_2^x \rangle = \sqrt{3}/4$ ,  $\langle T_2^z \rangle = 1/4$ ,  $\langle T_3^x \rangle = 0$ ,  $\langle T_3^z \rangle = -1/2$ ,  $\langle T_0^x \rangle = \langle T_0^z \rangle = 0$ . This means that while a dimerization pattern sets in on sites 1,2,3, the pseudospins on the "0" sites, shown in blue in Fig. 2(a) remain "free", i.e. there is no fixed dimer pattern on those sites and consequently a macroscopic degeneracy remains [4].

One should certainly keep in mind that the weak-coupling approach breaks artificially the lattice symmetry and while one hopes that the structure of the ground state is correct even in the isotropic limit  $J' = J$ , it is very difficult to assess this by other means (e.g. exact diagonalizations) at the present time. Fluctuations around the mean-field solution can lift the degeneracy, leading to unique dimer order. However the corresponding energy scale is very small, of the order of  $10^{-3}\beta$ ,  $\beta \equiv J'^3/(48J^2)$  [4]. A unique (singlet) ground state is also produced if one starts the expansion from a larger cluster of 16 sites, with an ordering scale of  $10^{-2}J$  [9] (extrapolated to the limit when all couplings are the same). In what follows we will take the mean-field solution as a starting point and discuss a physical mechanism, based on the presence of interactions beyond Heisenberg exchange, that can lead to the lifting of degeneracy and consequently to (magnetic) order in the ground state.

*Antiferromagnetic order induced by the Dzyaloshinsky-Moriya interactions.* Now we consider the effect of the DM interactions [10, 11] on the ground state properties. On a single tetrahedron the DM vectors are dis-

tributed as shown in Fig. 1(b)  $\mathbf{D}_{13} = \frac{D}{\sqrt{2}}(-1, 1, 0)$ ,  $\mathbf{D}_{24} = \frac{D}{\sqrt{2}}(-1, -1, 0)$ ,  $\mathbf{D}_{43} = \frac{D}{\sqrt{2}}(0, -1, 1)$ ,  $\mathbf{D}_{12} = \frac{D}{\sqrt{2}}(0, -1, -1)$ ,  $\mathbf{D}_{14} = \frac{D}{\sqrt{2}}(1, 0, 1)$ ,  $\mathbf{D}_{23} = \frac{D}{\sqrt{2}}(1, 0, -1)$ . Here  $D$  is the magnitude of the (all equal) DM vectors. The directions of the DM vectors respect the pyrochlore lattice symmetry and thus the DM interactions are expected to be always present in the system [12]. Since  $\mathbf{D}_{ij}$  originate from the spin-orbit coupling [10, 11], one expects  $D \ll J, J'$ .

Following the weak-coupling approach outlined above for the purely Heisenberg case, we have to determine how the singlet ground states  $|s_1\rangle, |s_2\rangle$  on a single tetrahedron are modified by the presence of  $D$ . Since the DM interactions break the rotational invariance, they admix triplets to the two ground states, not lifting their degeneracy [13]. We will also be interested in effects in the presence of an external magnetic field, and in this case the field (in combination with the DM interactions) also mixes certain triplet states with  $|s_1\rangle, |s_2\rangle$ . In order to determine the additional contributions to  $\hat{\mathcal{H}}_{\text{eff}}$ , it is convenient to express the spin operators on a tetrahedron, labeled as in Fig. 1(b), in terms of the pseudospin operators. For magnetic field  $\mathbf{H} = \frac{H}{\sqrt{2}}(1, 1, 0)$  (along the 1-3 bond), assuming  $D \ll J, H \ll J$ , we obtain

$$\begin{aligned} S_{1,3}^x &= \mp \frac{2\tilde{D}}{\sqrt{6}}T^y - \frac{\tilde{D}\tilde{H}}{\sqrt{3}}T^x, & S_{2,4}^x &= \mp \frac{2\tilde{D}}{\sqrt{6}}T^y + \frac{\tilde{D}\tilde{H}}{\sqrt{3}}T^x \\ S_{1,3}^y &= \mp \frac{2\tilde{D}}{\sqrt{6}}T^y + \frac{\tilde{D}\tilde{H}}{\sqrt{3}}T^x, & S_{2,4}^y &= \pm \frac{2\tilde{D}}{\sqrt{6}}T^y - \frac{\tilde{D}\tilde{H}}{\sqrt{3}}T^x \\ S_{1,3}^z &= \frac{2\tilde{D}}{\sqrt{6}}T^y \mp \tilde{D}\tilde{H}T^z, & S_{2,4}^z &= -\frac{2\tilde{D}}{\sqrt{6}}T^y \pm \frac{\tilde{D}\tilde{H}}{\sqrt{3}}T^x \end{aligned} \quad (4)$$

where we have defined, and from now on use the notation  $\tilde{D} \equiv D/J$ ,  $\tilde{H} \equiv H/J$ . These expressions are obtained by using the ground state wave-functions, written explicitly in Ref. [13], to lowest order in  $\tilde{D}$  and  $\tilde{D}\tilde{H}$ .

First we analyze the case of zero magnetic field ( $H = 0$ ). Taking into account the connections between the tetrahedra (green bonds in Fig. 2(a)), and using Eq. (4), we obtain the following additional term in the effective Hamiltonian ( $\hat{\mathcal{H}}_{\text{eff}} \rightarrow \hat{\mathcal{H}}_{\text{eff}} + \hat{\mathcal{H}}_{\text{eff}}^{(DM)}$ )

$$\hat{\mathcal{H}}_{\text{eff}}^{(DM)} = -J'\tilde{D}^2 \frac{2}{3} \sum_{i < j} T_i^y T_j^y \quad (5)$$

The above term is of the lowest, first order in  $J'$ . While extra terms of the same power  $J'(D'/J)^2$  also arise from the DM interactions  $D'$  on the inter-tetrahedral bonds, we find that they only give a small renormalization of the energy scale  $J'^3/(8J^2)$  in front of Eqs. (2,3) and are, therefore, neglected.

We have performed mean-field calculations of the Hamiltonian defined by Eqs. (2,3,5) in the unit cell of Fig. 2(a). The results can be particularly simply summarized in the limit  $\tilde{D} \ll 1$ , which is also the case of

physical relevance. It is physically clear that ferromagnetic order in the  $T_i^y$  component is generated on the “0” sites, since no order in the  $T_i^{x,z}$  components (dimer order) was present on those sites without DM interactions (on mean-field level). Indeed we find  $\langle T_0^y \rangle = 1/2$ , while for the other sites we have, to lowest non-trivial order in  $D$ ,  $\langle T_i^y \rangle \approx 1.8(D/J)^2$ ,  $i = 1, 2, 3$ . From Eq. (4) it is then clear that a non-zero average of the operator  $T_i^y$  corresponds to a finite moment in the ground state, with magnitude  $|\langle \mathbf{S} \rangle_i| = \tilde{D}\sqrt{2}\langle T_i^y \rangle$ :

$$|\langle \mathbf{S} \rangle_i| = \frac{\tilde{D}}{\sqrt{2}}, \quad i = 0; \quad |\langle \mathbf{S} \rangle_i| \approx \frac{3.6}{\sqrt{2}}\tilde{D}\frac{D^2}{J^2}, \quad i = 1, 2, 3. \quad (6)$$

From (4) it follows that the moments point out of the cube’s center (the cube is defined in Fig. 1(b)), leading to formation of sublattices and the order shown in Fig. 2(b). Since from Eq. (6)  $|\langle \mathbf{S} \rangle_i|/|\langle \mathbf{S} \rangle_0| \sim (D/J)^2 \ll 1$ ,  $i = 1, 2, 3$ , we have neglected the magnetic order on those tetrahedra.

The antiferromagnetic order of Fig. 2(b) corresponds to non-zero scalar chirality  $\chi = \langle \mathbf{S}_m \cdot (\mathbf{S}_n \times \mathbf{S}_l) \rangle \neq 0$ , where  $m, n, l$  are any three spins on the “blue” tetrahedra. The Ising symmetry  $T_i^y \rightarrow -T_i^y$  is broken in the ground state, which in terms of real spins corresponds to the time-reversal symmetry broken state of Fig. 2(b). The energy gain (per site of Fig. 2(a)) from the formation of the ordered state is  $E^{(DM)} = \langle \hat{\mathcal{H}}_{\text{eff}}^{(DM)} \rangle \approx -1.8J\tilde{D}^2(D/J)^2$ . The order we have just discussed is in competition with other mechanisms for lifting of the degeneracy that could originate from the Heisenberg interactions themselves (e.g. fluctuations beyond the mean-field), typically also leading to very small energy scales.

*Magnetic order induced by external magnetic fields and DM interactions.* In the presence of an external magnetic field other possibilities for lifting of the degeneracy exist. We will consider two (quite symmetric) field directions, for which the results are particularly transparent. The magnetic field leads to splitting of the ground states [13], which in the language of the pseudospin effective Hamiltonian produces an on-site “effective magnetic field”  $h$  in the pseudospin  $z$  direction:

$$\hat{\mathcal{H}}_{\text{eff}}^{(H)} = \hat{\mathcal{H}}_{\text{eff}}^{(H=0)} + h \sum_i T_i^z + \dots \quad (7)$$

We consider fields in the  $(1, 1, 0)$  and  $(0, 0, 1)$  directions (the axes are defined in Fig. 1(b)), and using the wavefunctions in a field we obtain

$$h = \begin{cases} \frac{1}{2}D^2H^2/J^3, & \mathbf{H} = \frac{H}{\sqrt{2}}(1, 1, 0) \\ -D^2H^2/J^3, & \mathbf{H} = H(0, 0, 1) \end{cases} \quad (8)$$

The dots in (7) denote lattice contributions, originating from the various combinations in Eq. (4) once the tetrahedra are coupled, and also producing terms of order  $D^2H^2$ . These terms are cumbersome and are not explic-

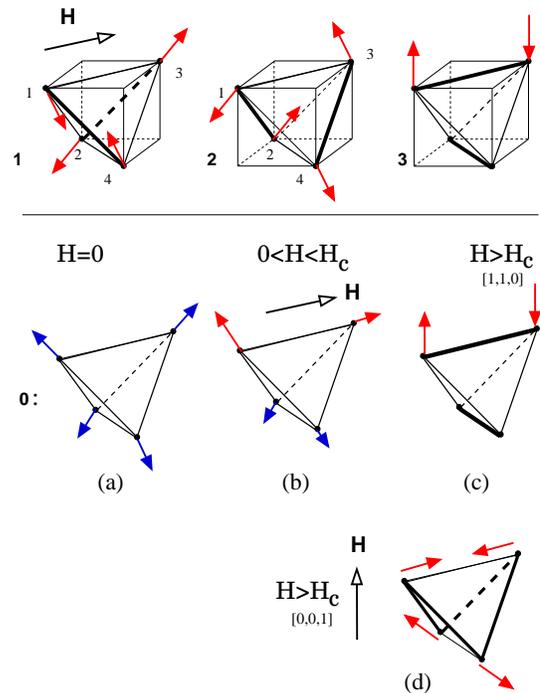


FIG. 3: (a,b,c) Evolution of magnetic order on the blue tetrahedra in an external magnetic field in the  $(1, 1, 0)$  direction. Upper row: field-induced order on the rest of the tetrahedra. (d) Representative pattern for a large field in the  $z$  direction.

itly written, but their effect is taken into account in the (numerical) mean-field implementation within the unit cell of Fig. 2(a).

The on-site  $h$  term in Eq. (7) is responsible for the main effect, namely competition between order in the  $T_i^z$  pseudospin component and order in the “chiral”  $T_i^y$  component favored by Eq. (5). Therefore the physics is that of the transverse field Ising model (although in our case the unit cell is larger). It is also clear that the mentioned competition is most effective on the “0” (blue) sites, while the non-zero averages of  $T_i^{z,x}$  on the other sites are not much affected by the presence of small  $D$  and  $H$ . We have found that a quantum transition takes place between a state with  $\langle T_0^y \rangle \neq 0$ ,  $H < H_c$  and  $\langle T_0^y \rangle = 0$ ,  $H \geq H_c$ . The result for  $\tilde{D} \ll 1$  can be written in an explicit way, and we have for the field  $\mathbf{H} = \frac{H}{\sqrt{2}}(1, 1, 0)$

$$\langle T_0^y \rangle^2 = \frac{1}{4} \left[ 1 - \left( \frac{\tilde{H}}{\tilde{H}_c} \right)^4 \right], \quad \tilde{H} \leq \tilde{H}_c \approx 5.3\sqrt{\frac{J}{J'}}\tilde{D} \quad (9)$$

while  $\langle T_0^z \rangle^2 = 1/4 - \langle T_0^y \rangle^2$  (and  $\langle T_0^z \rangle < 0$  since  $h > 0$ ). The values of the spin moments for given values of  $\langle T_i^{x,y,z} \rangle$  on a tetrahedron can be determined directly from Eq. (4). On the “0” (blue) sites this leads to evolution of the magnetic order as shown in Fig. 3(a,b,c). For  $H = 0$  there is only chiral order (blue arrows) with moment  $|\langle \mathbf{S} \rangle| \sim \tilde{D}$ , changing, for  $H > 0$  into a combination of chiral and field induced order (red arrows) with

$|\langle \mathbf{S} \rangle| \sim \tilde{D}\tilde{H}$ . Gradually, as  $H$  approaches  $H_c$  the chiral order diminishes (Eq. (9)), leaving for  $H > H_c$  only the field-induced component, equal to  $|\langle \mathbf{S} \rangle| = \tilde{D}\tilde{H}|\langle T_0^z \rangle| = \tilde{D}\tilde{H}/2$ ,  $H > H_c$ .

On the tetrahedra 1, 2, 3 labeled as in Fig. 2(a,b) there is virtually no evolution as a function of the field, and the order is determined by Eq. (4) with  $\langle T_1^{x,z} \rangle$  fixed by the Heisenberg exchanges (see discussion after Eq. (3)). This leads to the magnetic moments (proportional to  $\tilde{D}\tilde{H}$ ) shown in Fig. 3, upper row. On tetrahedra 1 and 2 the spins point along the internal diagonals of the cube perpendicular to the field. Dimerization is also present in the ground state (bolder lines = stronger bonds) and coexists with the magnetic order.

Finally, a similar quantum transition takes place for magnetic field in the  $z$  direction  $\mathbf{H} = H(0, 0, 1)$ . Without presenting explicit formulas we show the pattern on the blue sites for  $H > H_c$  in Fig. 3(d), while for small fields the order is a combination of Fig. 3(d) and Fig. 3(a). The critical field is also somewhat smaller in this case  $\tilde{H}_c \approx 3.8(J/J')^{1/2}\tilde{D}$ . For other, less symmetric field directions, the form of the effective Hamiltonian, and consequently the field-induced patterns can be quite complex.

In addition to the field-induced ordered patterns of Fig. 3, determined mostly by the inter-tetrahedral interactions, a single tetrahedron with DM interactions also possesses a finite moment in the direction of the field [13], meaning that the spins in Fig. 3 tend to tilt in that direction. However the moment along the field is proportional to  $\tilde{D}^2\tilde{H}$  and consequently has not been taken into account in Eq. (4), valid to lowest order in  $\tilde{D}, \tilde{H}$ . Finally, we emphasize that while we have assumed  $\tilde{D}, \tilde{H}$  to be small, the ratio  $\tilde{D}/\tilde{H}$  can be arbitrary, meaning that the quantum transitions in a field are within the limit of validity of our approach.

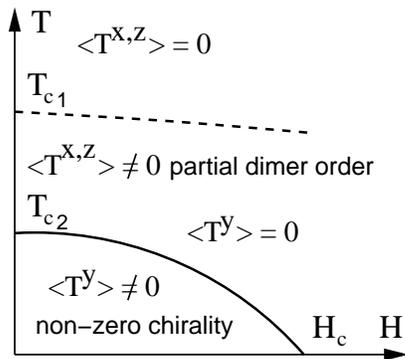


FIG. 4: Schematic phase diagram at non-zero temperature in the presence of small magnetic field ( $H \ll J$ ) and DM interactions.

At finite temperature we expect the phase diagram to look as presented in Fig. 4. The higher transition temperature  $T_{c1} \sim J^3/J^2$  corresponds to the scale below which the translational symmetry is broken (dimeriza-

tion occurs), and is determined by the energy scale in  $\tilde{\mathcal{H}}_{\text{eff}}$  for  $D = 0$ . We expect  $T_{c1}$  to decrease weakly when DM interactions are present. At a lower scale  $T_{c2}$  the Ising  $T^y \rightarrow -T^y$  symmetry is spontaneously broken by Eq. (5). For  $H = 0$  we can estimate  $T_{c2} \sim J'\tilde{D}^2(D/J')^2$ . At fixed field this finite-temperature transition is in the 3D Ising universality class, and the specific heat diverges as  $C \sim |T - T_{c2}(H)|^{-\alpha}$ ,  $\alpha \approx 0.11$  [14]. We emphasize that Fig. 4 shows only the low-field part of the phase diagram (since  $H_c \sim D \ll J$ ), while the physics at high fields cannot be determined within the effective Hamiltonian framework presented here. In certain pyrochlores, such as the gadolinium titanium oxides with  $S=7/2$ , field-driven phase transitions have been observed [15], although in this material the magnetic order is typically explained as originating from the long-range dipolar interactions. For such large value of the spin the DM mechanism for magnetic order, at least the way it is developed in this work, should not be effective since our calculations were based on strong singlet correlations in the ground state.

In conclusion, we have shown that DM interactions can induce weak antiferromagnetic order characterized by non-zero chirality. In an external magnetic field quantum transitions between the chiral state and field-induced ordered states take place. We have used an expansion around a configuration which breaks the lattice symmetry [4] and leaves a macroscopic degeneracy, subsequently lifted by the DM interactions. Full restoration of lattice symmetry within such an approach seems impossible to achieve, as it is impossible for example in the large- $N$  approach [16]. Nevertheless we expect that without DM interactions the ground state properties and the inherent degeneracy present in this strongly frustrated magnet are well accounted for. In this situation the DM interactions “push” the pyrochlore lattice towards the ordered states analyzed in the present work. More generally, the DM interactions can be relevant and lead to weak magnetism in strongly frustrated systems, where the Heisenberg exchanges on their own fail to produce long-range order.

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