Dzyaloshinsky-Moriya-Induced Order in the Spin-Liquid Phase of the S=1/2 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. The chiral antiferromagnetic state is characterized by a small staggered magnetic moment induced by the DM interaction. An external magnetic field can also lead to characteristic field-induced ordering patterns, strongly dependent on the field direction, and generally separated by a quantum phase transition from the chiral ordered phase. The phase diagram at finite temperature is also discussed.

I. 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. 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. The effects of various additional interactions have also been studied, such as magnetoelastic couplings, long-range dipolar interactions, and orbital degeneracy. 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 expected to be present by symmetry. For the S=1/2 Heisenberg model on the pyrochlore lattice it has been suggested that the ground state is dimerized (non-magnetic), but macroscopic degeneracy still remains. For certain other lattices, such as the 2D pyrochlore and related models, the ground state is a unique valence bond solid, and while the DM interactions (if present) can lead to non-trivial order in the ground state, such DM induced order can only occur above a critical threshold, due to its inherent competition with the underlying dimer order. In this work we show that in the 3D pyrochlore antiferromagnet, where a macroscopic degeneracy is present, the DM interactions have a more profound effect and can lift the degeneracy, leading to a chiral antiferromagnetic state with a small staggered magnetic moment. In an external magnetic field quantum transitions between weakly ordered states with different symmetries, depending on the field direction, are possible. We determine the field-induced patterns for several field orientations, generally pointing in highly-symmetric crystal directions. The phase diagram at finite temperature is also briefly discussed.

![Fig. 1: (a) Pyrochlore lattice. (b) Distribution of DM vectors on a single tetrahedron (four of the six shown, see text).](image)

The spin Hamiltonian (S=1/2) is

\[ \hat{\mathcal{H}} = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} D_{ij} (\mathbf{S}_i \times \mathbf{S}_j), \]

where \( D_{ij} \) are the DM vectors, to be specified later. We start by summarizing the results for \( D_{ij} = 0 \), i.e. the Heisenberg case. Our starting point is the strong-coupling approach, similar to that of Refs. [2,4], with the lattice 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: \( |s_1\rangle = \frac{1}{\sqrt{2}} \{|1, 2, 3, 4\} + |2, 3, 4, 1\rangle \), \( |s_2\rangle = \{|1, 2, 3, 4\} - |2, 3, 4, 1\rangle \)
[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 inter-tetrahedron Hamiltonian in the singlet sub-space:\(^7\)

\[
\hat{H}_{\text{eff}} = \frac{J'^3}{8J^2} \left[ \hat{H}_{\text{eff}}^{(2)} + \hat{H}_{\text{eff}}^{(3)} \right] + \text{Const.}, \quad (2)
\]

where

\[
\hat{H}_{\text{eff}}^{(2)} = \sum_{(i,j)} \left( \Omega_{ij}^x T_i^x T_j^x + \Omega_{ij}^y T_i^y T_j^y + \Omega_{ij}^z (T_i^z T_j^z + T_j^z T_i^z) \right),
\]

\[
\hat{H}_{\text{eff}}^{(3)} = \sum_{(i,j,k)} \left( \frac{1}{3} T_i^z T_j^z T_k^z - T_i^z T_j^z T_k^z + \frac{T_i^z}{\sqrt{3}} (T_j^z T_k^z - T_j^z T_k^z) \right)
\]

(3)

In the two-body part we have defined

\[
\Omega_{01}^x = \Omega_{12}^x = 1/2, \quad \Omega_{03}^x = \Omega_{23}^x = -1/6, \quad \Omega_{24}^x = \Omega_{01}^x = \Omega_{02}^x = \Omega_{13}^x = 1/3, \quad \Omega_{03}^z = \Omega_{24}^z = -\Omega_{02}^z = -\Omega_{13}^z = 1/(2\sqrt{3}).
\]

(4)

All remaining \( \Omega_{ij} = 0, \) \( i < j \). The site indexes \( i,j \) 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 (containing the sites 0,1,2,3). In the three-body interaction the indexes run over the values: \( (i,j,k) = \{(3, 2, 1), (1, 0, 3), (2, 3, 0), (0, 1, 2)\} \).

On a mean-field level the ground state of \( \hat{H}_{\text{eff}} \) is defined by the following averages:

\[
\langle T_i^x \rangle = -\sqrt{3}/4, \quad \langle T_i^y \rangle = 1/4, \quad \langle T_i^z \rangle = \sqrt{3}/4, \quad \langle T_i^2 \rangle = 1/4, \quad \langle T_i^3 \rangle = 0, \quad \langle T_i^4 \rangle = 0, \quad \langle T_i^5 \rangle = 0.
\]

(6)

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.\(^8\)

One should certainly keep in mind that the strong-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. Nevertheless this approach is expected to provide reliable description of the ground state properties as long as the relevant physics remains in the singlet subspace, i.e. the triplet modes stay high in energy and no magnetic order is generated, as might be the case for the pyrochlore antiferromagnet due to the strong frustration. Fluctuations around the mean-field solution, Eq. (6), can lift the degeneracy, leading to unique dimer order. However the corresponding degeneracy lifting energy scale is very small of the order of \( 10^{-3} \beta, \beta \equiv J'^3/(48J^2) \). A unique (singlet) ground state is also produced if one starts the expansion from a larger cluster of 16 sites, with an ordering energy scale (energy gain) of \( 10^{-2}J \), extrapolated to the limit where all couplings are equal.\(^9\)

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.

\[\text{FIG. 2: (Color online.) (a) Fcc lattice of tetrahedra (tetrahedron = dot) with interactions \( J' \) between them. (b) Antiferromagnetic chiral order on the blue (dark gray) tetrahedra, with magnetic moment \(|\langle S \rangle| \sim D\), induced by the DM interactions. On the gray tetrahedra (labeled as 1,2,3) the order has the same symmetry, but is much weaker \(|\langle S \rangle| \sim D^3 \sim D\), Eq. (9), and is not shown.}\]

II. CHIRAL ANTIFERROMAGNETIC ORDER INDUCED BY THE DZYALOSHINSKY-MORIYA INTERACTIONS

Now we consider the effect of the DM interactions\(^11,12\) on the ground state properties. On a single tetrahedron the DM vectors are distributed as shown in Fig. 1(b), or explicitly: \( \mathbf{D}_{13} = \frac{D}{\sqrt{2}}(-1,1,0), \mathbf{D}_{24} = \frac{D}{\sqrt{2}}(-1,-1,0), \mathbf{D}_{13} = \frac{D}{\sqrt{2}}(0,-1,1), \mathbf{D}_{12} = \frac{D}{\sqrt{2}}(0,1,-1), \mathbf{D}_{14} = \frac{D}{\sqrt{2}}(1,0,1), \mathbf{D}_{23} = \frac{D}{\sqrt{2}}(1,0,-1)\). 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.\(^11,12\) Since \( \mathbf{D}_{ij} \) originate from the spin-orbit coupling\(^11,12\) we have \( D \ll J, J' \), and typically the values of the DM interactions are several percent of the Heisenberg couplings. There are two DM distribution patterns that are equally acceptable on symmetry grounds - the one shown in Fig. 1(b), and one with all directions of the vectors \( \mathbf{D}_{ij} \) reversed (\( \mathbf{D}_{ij} \rightarrow -\mathbf{D}_{ij} \).}
These two cases were named, respectively, "indirect" and "direct" in Ref. 13. The reader is referred to that paper for more details on Moriya’s rules as applied to the pyrochlore lattice. In the extreme quantum case of S=1/2, and within our approach, we have found that the two allowed (by symmetry) DM distributions lead to qualitatively the same physics (see discussion following Eq. (8)).

Following the strong-coupling approach outlined above for the purely Heisenberg case, we have to determine how the singlet ground states |s1⟩, |s2⟩ on a single tetrahedron are modified by the presence of D. Since the DM interactions break the spin rotational invariance, they admix triplets to the two ground states, not lifting their degeneracy. We will also be interested in effects of 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 |s1⟩, |s2⟩. In order to determine the additional contributions to $\hat{H}_{\text{eff}}$, it is convenient to express the spin operators on a singlet tetrahedron, labeled as in Fig. 1b, in terms of the pseudospin operators. For magnetic field $\mathbf{H} = \mathbf{H}/\sqrt{2}(1, 1, 0)$ (along the 1 – 3 bond), assuming $D \ll J$ and $H \ll J$, we obtain (defining the rescaled quantities $\tilde{D}, \tilde{H}$ along the way):

$$\begin{align*}
H & = \frac{H}{\sqrt{2}}(1, 1, 0); \quad \tilde{D} = D/J, \quad \tilde{H} = H/J, \\
S_{1,3}^x & = \tilde{D}\frac{\sqrt{2}}{4} - \frac{\tilde{D}}{3} \tilde{H} T^x, \\
S_{1,4}^y & = \tilde{D}\frac{\sqrt{2}}{4} + \frac{\tilde{D}}{3} \tilde{H} T^y, \\
S_{1,4}^z & = \tilde{D}\frac{2}{4} T^z + \frac{\tilde{D}}{3} \tilde{H} T^z.
\end{align*}$$

(7)

The notation $\mathbf{S}_{i,j}$ simply combines in one line the formulas for both $\mathbf{S}_i$ and $\mathbf{S}_j$, where $i$ and $j$ label sites on a tetrahedron (as defined in Fig. 1b), while the left index in $\mathbf{S}_{i,j}$ corresponds to the upper sign on the right hand side, and the right index to the lower sign. The formulas Eq. (7) are obtained by using the ground state wave-functions, written explicitly in Ref. 5 (Eqs. (2.5)), to lowest order in $\tilde{D}$ and $\tilde{H}$. For magnetic field in the z direction, the corresponding expressions are given in Appendix A.

First we analyze the case of zero magnetic field ($\mathbf{H} = 0$). Taking into account the connections between the tetrahedra (green bonds in Fig. 2a), and using Eq. 6, we obtain an additional interaction term, so that the full effective Hamiltonian $\hat{H}_{\text{eff}}^{(DM)}$ becomes

$$\begin{align*}
\hat{H}_{\text{eff}}^{(DM)} & = \hat{H}_{\text{eff}} - J'\tilde{D}\frac{2}{3} \sum_{i,j} T_{ij}^y T_{ij}^y, \\
& = \hat{H}_{\text{eff}} - J'\tilde{D}\frac{2}{3} \sum_{i,j} T_{ij}^y T_{ij}^y.
\end{align*}$$

(8)

where $\hat{H}_{\text{eff}}$ is the part originating from the Heisenberg exchanges, Eq. (2). The above result is obtained in lowest, first order in $J'$. While extra terms of the same power $J'(D^2/J)^2$ 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 Eq. (2) and are, therefore, neglected.

We would like to also point out that in general the coupling constant in $\hat{H}_{\text{eff}}^{(DM)}$ is not symmetric under $D \to -D$. To verify this requires a calculation of the next to leading order in $\tilde{D}$ in Eq. (7). We have found that, in the case of zero field $H = 0$, the next order present is $\tilde{D}^2$, and one has to substitute in all formulas $\tilde{D} \to \tilde{D} - \frac{1}{4} \tilde{D}^2$. Consequently the same substitution has to be made in the coefficient $-J'\tilde{D}\frac{2}{3}$ in Eq. (8). Most importantly however the $T_{ij}^y T_{ij}^y$ structure of the interaction is not affected by increasing the strength of $D \ll 1$, and from now on we will work with the leading order in $\tilde{D}$. Therefore the physics (ground state structure) associated with the two DM distribution patterns will be the same. This conclusion might be connected with the fact that we have kept only the lowest non-trivial order in the coupling $J'$ in the effective Hamiltonian - we have used this as our guiding principle as the difficulties associated with the derivation and analysis of higher orders seem insurmountable.

We have performed mean-field calculations of the Hamiltonian defined by Eqs. (2) and (6) in the unit cell of Fig. 2a, as represented by the four sites connected by green lines. The results can be particularly simply summarized in the limit $D \ll 1$, which is also the case of physical relevance. It is physically clear that ferromagnetic order in the $T_{ij}^z$ component is generated on the “0” sites, since no order in the $T_{ij}^z$ components (dimer order) was present on those sites without DM interactions (on mean-field level), Eq. (6). Indeed we find $\langle T_{ij}^y \rangle = 1/2$, while for the other sites we have, to lowest non-trivial order in $D$, $\langle T_{ij}^y \rangle \approx 1.8(D/J')^2, i = 1, 2, 3$. From Eq. (8) it is then clear that a non-zero average of the operator $T_{ij}^y$ corresponds to a finite moment in the ground state, with magnitude $\langle |\mathbf{S}| \rangle = D\sqrt{2} \langle T_{ij}^y \rangle$. To summarize:

$$\begin{align*}
\langle T_{ij}^y \rangle & = 1/2, \quad \langle T_{ij}^y \rangle \approx 1.8(D/J')^2, i = 1, 2, 3 \\
|\mathbf{S}| & = \frac{D}{\sqrt{2}} i = 0; \quad |\mathbf{S}| \approx \frac{3.6}{\sqrt{2}} D\frac{D^2}{J^2} i = 1, 2, 3.
\end{align*}$$

(9)

Here $|\mathbf{S}|$ stands for the magnitude of the moment on each site of pyrochlore lattice, belonging to a tetrahedron labeled by the index i. From (7) it follows that the moments point out of the cube’s center (the cube is defined in Fig. 1b)), leading to formation of sublattices and the order shown in Fig. 2b). Since from Eq. 9 $|\mathbf{S}|/|\langle \mathbf{S} \rangle| \approx (D/J')^2 \ll 1, i = 1, 2, 3$, we have neglected the magnetic order on those tetrahedra.

The antiferromagnetic order of Fig. 2b corresponds to non-zero scalar chirality $\chi = (S_m \times S_i \times S_l) \neq 0$, where $m, n, l$ are any three spins on a given tetrahedron. The Ising symmetry $T_{ij}^y \to -T_{ij}^y$ is spontaneously broken in the ground state, which in terms of real spins corresponds to the time-reversal symmetry broken state of Fig. 2b). In this state the two ground state wave...
functions $|\Phi\rangle$ and $|\Psi\rangle$ (see (A1)) form linear combinations in the "chiral" sector: $\alpha|\Phi\rangle + i\beta|\Psi\rangle$, where $\alpha, \beta$ are real coefficients ($\alpha^2 + \beta^2 = 1$). This combination is ferromagnetically repeated on every $T^0$ ordered tetrahedron. A straightforward calculation shows that both $\langle T^0_i \rangle \propto \alpha \beta$, $\chi \propto \alpha \beta$. The energy gain (per site of Fig. 2(a)) from the formation of the ordered state is $\Delta E = \langle \hat{H}^{(DM)}_{\text{eff}} \rangle - \langle \hat{H}_{\text{eff}} \rangle \approx -1.8J'\tilde{D}^2(D/J')^2$. 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.

### III. MAGNETIC ORDER INDUCED BY EXTERNAL MAGNETIC FIELDS IN THE PRESENCE OF DM INTERACTIONS

In the presence of an external magnetic field other possibilities for lifting of the degeneracy exist. We will consider three symmetric field orientations, for which the results are particularly transparent. The magnetic field generally leads to splitting of the ground states, which in the language of the pseudospins produces an on-site "effective magnetic field" $h$ in the pseudospin $z$ direction. The effective Hamiltonian has the form

$$\hat{H}^{(H)}_{\text{eff}} = \hat{H}^{(DM)}_{\text{eff}} + h \sum_i T^z_i + \delta\hat{H}^{(H)}_{\text{eff}}.$$ (10)

We consider fields in the $(1,1,0)$ and $(0,0,1)$ directions, as well as comment on the case $(1,1,1)$, where the axes are defined in Fig. 4(b). Using the wave-functions in a field we obtain (see Eq. (A3)):

$$h = \left\{ \begin{array}{ll} \frac{1}{2}D^2H^2/J^3, & H = \frac{H}{\sqrt{2}}(1,1,0) \\ -D^2H^2/J^3, & H = \frac{H}{\sqrt{2}}(0,0,1) \\ 0, & H = \frac{H}{\sqrt{2}}(1,1,1) \end{array} \right.$$ (11)

$\delta\hat{H}^{(H)}_{\text{eff}}$ in (10) represents lattice contributions, originating from the various combinations in Eq. (11) once the tetrahedra are coupled, and also producing terms of order $D^2H^2$. These terms are cumbersome and are not explicitly written, but their effect is taken into account in the (numerical) mean-field implementation within the unit cell of Fig. 2(a). A further discussion appears in Appendix B.

We will mostly discuss the two cases with $h \neq 0$. Then the on-site $h$ term in Eq. (10) is responsible for the main effect, namely competition between order in the $T^z_i$ pseudospin component and order in the "chiral" $T^x_i$ 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 $T^x_i$ 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 $\langle T^0_0 \rangle \neq 0, H < H_c$ and $\langle T^0_0 \rangle = 0, H \geq H_c$. The result for $D \ll 1$ can be written in an explicit way, and we have for the field $H = \frac{H}{\sqrt{2}}(1,1,0)$

$$\langle T^0_0 \rangle^2 = \frac{1}{4} - \left( \frac{\tilde{D}H}{H_c} \right)^4, \quad \tilde{H} \leq \tilde{H}_c \approx 5.3\sqrt{\frac{J}{J'}} \tilde{D}$$ (12)

(13) and $\langle T^0_i \rangle^2 < 0$ since $h > 0$. The values of the spin moments for given values of $\langle T^z_i \rangle$ on a tetrahedron can be determined directly from Eq. (12). 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 $\langle |\langle S \rangle| \rangle \sim \tilde{D}$, changing, for $H > 0$ into a combination of chiral and field induced order (red arrows) with $\langle |\langle S \rangle| \rangle \sim \tilde{D}H$. Gradually, as $H$ approaches $H_c$ the chiral order diminishes (Eq. (13)), leaving for $H > H_c$ only the field-induced component, equal to $\langle |\langle S \rangle| \rangle = \tilde{D}H/\langle T^0_0 \rangle = \tilde{D}H/2$, $H > H_c$.

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. (13) with $\langle T^z_i \rangle$ fixed by the Heisenberg exchanges, see Eq. (6). This leads to the magnetic moments (proportional to $\tilde{D}H$) 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 co-exists with the magnetic order.

\[ H = H(0,0,1) \]

A similar quantum transition takes place for magnetic field in the \( z \) direction. In this case the field-induced order is shown in Fig. 4. The critical field is also somewhat smaller in this case \( H_c \approx 3.8(J/J')^{1/2}D \), mainly due to the fact that \( h \) is larger by a factor of 2, see Eq. (11). For other, less symmetric field directions, the form of the effective Hamiltonian, and consequently the field-induced patterns can be quite complex. Finally, in the case of a field \( H \sim (1,1,1) \), when \( h = 0 \), the quantum transition described above does not take place, and the chiral order of Fig. 5(a) essentially does not evolve. In this case the various additional terms similar to the ones described in Appendix B may lead to small, sub-leading deviations from the perfect chiral state.

In addition to the field-induced ordered patterns of Fig. 4 and determined mostly by the inter-tetrahedral interactions, a single tetrahedron with DM interactions also possesses a finite moment in the direction of the field, meaning that the spins in Fig. 4 would also tend to tilt in that direction. However the moment along the field is proportional to \( D^2\tilde{H} \), as can be deduced from the fact that the ground state energy varies as \( D^2\tilde{H}^2 \) from (A3). Consequently this component has not been taken into account in Eqs. (7), (A2), 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 \( D/\tilde{H} \) can be arbitrary, meaning that the quantum transitions in a field are within the limit of validity of our approach.

**IV. PHASE DIAGRAM AND DISCUSSION**

At finite temperature we expect the phase diagram to look as presented in Fig. 5 (it is assumed that \( h \neq 0 \)). The higher transition temperature \( T_{c1} \sim J'^3/J^2 \) corresponds to the scale below which the translational symmetry is broken (dimerization occurs), and is determined by the energy scale in \( \mathcal{H}_{\text{eff}} \) for \( D = 0 \), Eq. (2). We expect \( T_{c1} \) to have weak dependence on magnetic field when DM interactions are present. At a lower scale \( T_{c2} \) the Ising transition \( T^y \to -T^y \) symmetry is spontaneously broken by Eq. (8). 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 \( C \sim (T - T_{c2}(H))^{-\alpha} \), \( \alpha \approx 0.11 \). We emphasize that Fig. 5 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.

![Phase Diagram](attachment:image.png)

**FIG. 5:** Schematic phase diagram at non-zero temperature in the presence of small magnetic field \( (H \ll J) \) and DM interactions.

In certain pyrochlores, such as the gadolinium titanates with \( S = 7/2 \), field-driven phase transitions have been observed, 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 in a long range dipolar interactions. For such large value of the spin the DM mechanism for magnetic order, at least in a long range interaction, should not be effective since our calculations were based on strong singlet correlations in the ground state. At the moment it is hard to point out a class of materials where the DM interactions are definitely expected to be dominant with respect to other anisotropies capable to produce ordering; some possible examples are given in Ref. [13]. In particular, our results are specific to the case \( S = 1/2 \), while most
currently known pyrochlores have higher $S$. Let us also point out the main differences between the present work and the purely classical model. (a.) We have found that the antiferromagnetic order is weak, determined by the DM interaction scale itself. (b.) The "chiral" non-coplanar pattern is the stable one for $S = 1/2$ in the absence of a field. Field-induced patterns then dominate for $H > H_c \sim D$. Let us also give some estimates: if we take the optimistic viewpoint and apply our formulas to the case $J = J'$, and take $J \sim 100 K, D/J \sim 10^{-1}$, then the characteristic critical fields for onset of chiral order would be $T_{c2} \sim 10 mK$, and the magnitude of the moment $|S| \sim 0.1$. The characteristic critical fields would be $H_c \sim 10 K$. These should be viewed as order of magnitude estimates. Inevitably the scale $T_{c2}$ falls into the $mK$ range, which, in combination with the smallness of $|S|$ itself, would probably make the chiral state rather hard to observe with neutron scattering techniques used to probe the spin structure. The spectral weight of magnetic excitations in such neutron scattering measurements would be determined by $D$ itself. However the scale of $H_c$ of around $10T$ or smaller (also determined by $D$), means that the various field-induced patterns, strongly dependent on the magnetic field direction, might be accessible. 

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. Field-induced patterns with different symmetries depending on the direction of the field are very characteristic of the presence of DM interactions. We have used an expansion around a configuration which breaks the lattice symmetry 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. 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 antiferromagnetism in strongly frustrated systems, where the Heisenberg exchanges on their own fail to produce long-range order. We also emphasize that the physics behind the weakly antiferromagnetic states with different symmetries discussed in this work is very different from the phenomenon of weak ferromagnetism, usually associated with DM interactions.

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APPENDIX A: SPIN OPERATORS AND ENERGY LEVELS IN MAGNETIC FIELD

Here we present the expressions for the spin operators for magnetic field in the $z$ direction. The two ground state wave-functions $|s_1\rangle, |s_1\rangle$ are modified in the following way in the presence of DM interactions ($|s_1\rangle \rightarrow |\Phi\rangle$, $|s_2\rangle \rightarrow |\Psi\rangle$):

$$H = H(0, 0, 1);$$

$$|\Phi\rangle = |s_1\rangle + \frac{i\sqrt{6}D}{4} \left(|p_x| - |q_y| + |q_x| + |q_y|\right) + \frac{\sqrt{6}D H}{4} \left(|p_x| + |q_y| - |q_x| + |q_y|\right),$$

$$|\Psi\rangle = |s_2\rangle + \frac{i\tilde{D}}{2\sqrt{2}} \left(|p_x| + |p_y| + |q_x| - |q_y|\right) + \frac{\tilde{D}H}{2\sqrt{2}} \left([-p_x] + |p_y| + |q_x| + |q_y|\right),$$

(A1)

where $|p_x|, |q_y|, |q_x|$, $\mu = x, y, z$ are the three excited triplet states on a tetrahedron. From these equations we obtain

$$S_{1,3}^{y} = \frac{2D}{\sqrt{6}} T_{y} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{z} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{x}$$

$$S_{1,3}^{x} = \frac{2D}{\sqrt{6}} T_{y} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{z} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{x}$$

$$S_{1,3}^{z} = \frac{2\tilde{D}}{\sqrt{6}} T_{y}$$

$$S_{2,4}^{z} = \frac{2\tilde{D}}{\sqrt{6}} T_{y} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{z} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{x}$$

$$S_{2,4}^{x} = \frac{2D}{\sqrt{6}} T_{y} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{z} \pm \frac{\tilde{D}H}{\sqrt{6}} T_{x}$$

$$S_{2,4}^{y} = -\frac{2D}{\sqrt{6}} T_{y}.$$  

(A2)

We also give the ground state energy splitting on a single tetrahedron, in (weak) external magnetic field with arbitrary direction

$$H = (H_x, H_y, H_z);$$

$$E_0^{(1,2)} = -\frac{3J}{2} - \frac{3D^2}{2J} - \frac{D^2|H|^2}{J^3}$$

$$+ \frac{D^2}{2J^3} \sqrt{|H|^4 - 3(H_x^2 H_y^2 + H_x^2 H_z^2 + H_y^2 H_z^2)}$$

(A3)

From here the "effective magnetic field" $h$ appearing in the pseudospin Hamiltonian (10) is: $h = E_0^{(2)} - E_0^{(1)}$.

APPENDIX B: EFFECTIVE HAMILTONIAN CONTRIBUTIONS IN MAGNETIC FIELD

We briefly discuss the structure and treatment of the term $\delta H_{\text{eff}}^{(H)}$ in (10). As is clear from (7) and (A2), po-
tentially contributions of two types appear: (a.) terms of order $D^2 \tilde{H}$, and (b.) terms of order $D^2 \tilde{H}^2$. While treating these terms we will assume that the unit cell structure of Fig. 2(a) does not change. Indeed, since we are interested in the case of weak fields $\tilde{H} \ll 1$, the above coupling constants are of sub-leading order with respect to the case without a field, and therefore one does not expect the unit cell to change. Under this assumption we find that the terms of order $D^2 \tilde{H}$ vanish identically for the field directions considered in this work. The remaining contribution, e.g. for a field $\mathbf{H} = \frac{\tilde{H}}{\sqrt{2}}(1,1,0)$, written (per site) within the unit cell of Fig. 2(a) is

$$\langle \delta \hat{H}^{(H)}_{\text{eff}} \rangle = \frac{J}{6} D^2 \tilde{H}^2 \delta \hat{H}, \quad (\text{B1})$$

where

$$\delta \hat{H} = -4(T^x_0 + T^x_3)(T^x_1 + T^x_2) + \sqrt{3}(T^x_0 - T^x_3)(T^x_1 - T^x_2) + \sqrt{3}(T^x_0 - T^x_3)(T^x_1 - T^x_2) + 3(T^x_0 T^x_3 - T^x_0 T^x_3 + T^x_0 T^x_3 - T^x_0 T^x_3). \quad (\text{B2})$$

These terms were taken into account in our numerical solution of the mean-field equations corresponding to (10).