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## Noncollinear Ground State from a Four-Spin Chiral Exchange in a Tetrahedral Magnet

I. A. Ado, O. Tchernyshyov, and M. Titov

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## Non-collinear ground state from a four-spin chiral exchange in a tetrahedral magnet

I. A. Ado, 1 O. Tchernyshyov, 2 and M. Titov 1

<sup>1</sup>Institute for Molecules and Materials, Radboud University, Heyendaalseweg 135, 6525AJ Nijmegen, The Netherlands <sup>2</sup>Institute for Quantum Matter and Department of Physics and Astronomy, Johns Hopkins University, Baltimore, MD 21218, USA (Dated: June 4, 2021)

We propose a quartic chiral term  $m_x m_y m_z \nabla \cdot \mathbf{m}$  for the energy density of a cubic ferromagnet with broken parity symmetry (point group  $T_d$ ). We demonstrate that this interaction causes a phase transition from a collinear ferromagnetic state to a non-collinear magnetic cone ground state provided its strength exceeds the geometric mean of magnetic exchange and cubic anisotropy. The corresponding non-collinear ground state may also be additionally stabilized by an external magnetic field pointing along certain crystallographic directions. The four-spin chiral exchange does also manifest itself in peculiar magnon spectra and favors spin waves with the wave vector that is perpendicular to the average magnetization direction.

Chiral spin textures, such as spin-spirals and skyrmion crystals, are expected to play an important role in novel information technologies [1–6]. The appearance of non-collinear chiral spin states is often understood as the result of an interplay between Dzyaloshinskii-Moriya interaction (DMI) and magnetic anisotropy [7]. Indeed, the role of DMI in stabilizing non-collinear magnetic order has been well established since the first observations of helical spin-density waves in 1976 [8]. More recently, various non-collinear textures including magnetic cones, helixes, vortices or skyrmion crystals have been routinely observed in a variety of magnetic systems with broken inversion symmetry (for instance, in MnSi, FeGe, Ir/Co/Pt, or Pt/CoFeB/MgO [8–13]), where DMI is expected to be strong.

DMI has been first proposed [14, 15] as an indirect asymmetric Heisenberg exchange between neighboring spins. Theoretical understanding of non-collinear magnetic order is, however, normally achieved within Ginzburg-Landau functional approach that resorts to micromagnetic energy functional of a magnet. On the level of micromagnetic energy, DMI is often defined more broadly by terms that are quadratic in magnetization but linear with respect to magnetization gradients – the so-called Lifshitz invariants (LI) [16]. From a symmetry point of view, terms linear in gradients are only allowed for systems with broken inversion symmetry.

In conducting systems, LI terms may also originate from long-range magnetic interactions mediated by conduction electrons with strong spin-orbit coupling (e. g. from contributions to a long range asymmetric exchange due to RKKY type of processes [17–24]). The importance of this mechanism is supported by the fact that long-range non-collinear order is indeed mostly observed in conducting magnets.

In 1989 Bogdanov and Yablonskii [25] determined possible combinations of LIs in micromagnetic energy for several important crystalline symmetry classes. These LI terms lead to instability of collinear order. More recently, Ado *et al.* showed that for three specific point groups for crystals with broken inversion symmetry:  $T_d$ ,  $D_{3h}$  and  $C_{3h}$  all LI terms are forbidden by symmetry. The natural question to ask

	2-spin (LI)	4-spin (non-LI)
0	$\mathbf{m} \cdot (\nabla \times \mathbf{m})$	$\sum_{\alpha} m_{\alpha}^{3} (\nabla \times \mathbf{m})_{\alpha}$ $m_{x}^{2} \mathcal{L}_{yx}^{(x)} + m_{y}^{2} \mathcal{L}_{zx}^{(y)} + m_{z}^{2} \mathcal{L}_{xy}^{(z)}$
 $T_d$	None	$m_x m_y m_z (\nabla \cdot \mathbf{m})$

TABLE I: Energy density from two-spin (LI) and four-spin (non-LI) chiral interactions in the point group T (chiral tetrahedral symmetry) that is a subgroup of O (chiral octahedral symmetry) and  $T_d$  (full tetrahedral symmetry). The notation  $\mathcal{L}_{\alpha\beta}^{(\gamma)} = m_\alpha \partial_\gamma m_\beta - m_\beta \partial_\gamma m_\alpha$  denotes the Lifshitz invariant (LI).

is whether the broken inversion symmetry may still destroy the collinear order in such crystals despite the absence of LI terms.

In this Letter we answer this question positively for the tetrahedral point group  $T_d$  (which is the most symmetric group out of the three). In particular, we demonstrate that a lack of inversion symmetry in this group does lead to the appearance of a non-LI type contribution  $w_{4\rm S} \propto m_x m_y m_z \nabla \cdot {\bf m}$  in the micromagnetic energy density, where the vector  ${\bf m}$  is a unit vector in the direction of local magnetization.

We use the term 4-spin chiral interaction for such a non-LI type of contribution and refer to standard LI terms as 2-spin chiral interactions. Below we demonstrate that the 4-spin chiral interaction  $w_{4\rm S}$  destroys collinear magnetic order provided cubic crystal anisotropy is sufficiently weak. We also demonstrate that 4-spin chiral interaction can be revealed in a collinear magnetic state by asymmetry of the magnon spectra.

Our analysis remains fairly general and applies to a variety of magnetic systems with tetrahedral point group symmetry of magnetic atoms such as half-metal halcogenides, spinels, pyrochlores and Heusler alloys including Cu<sub>3</sub>FeTe<sub>4</sub>, GaV<sub>4</sub>S<sub>8</sub>, Lu<sub>2</sub>V<sub>2</sub>O<sub>7</sub>, Cr<sub>x</sub>Zn<sub>1-x</sub>Te, Mn<sub>x</sub>Zn<sub>1-x</sub>S and related materials [26–32]. We expect the proposed 4-spin chiral interaction to be especially strong in conducting magnets with large spin-orbit coupling of charge carriers. We also note that the cubic magnetic anisotropy itself is also fourth order in magnetization and is often very substantial. It is therefore natural to expect that

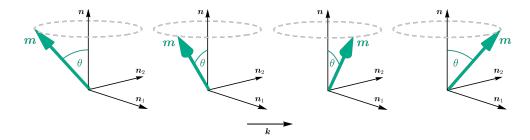


FIG. 1: Schematic illustration of the magnetic cone state that minimize the energy of Eq. (6). The state wave-vector is perpendicular to the average magnetization,  $\mathbf{k} \cdot \mathbf{n} = 0$ , that is characteristic for the 4-spin chiral interaction  $w_{4\mathrm{S}} \propto m_x m_y m_z \nabla \cdot \mathbf{m}$ .

the 4-spin chiral interaction may be equally important, while, to the best of our knowledge, it has never been previously proposed or analyzed.

In Table I we list the results of the symmetry analysis of micromagnetic energy functional  $E[\mathbf{m}]$  for a lattice with the point group T (chiral tetrahedral symmetry). This is a common subgroup of the point groups O and  $T_d$ . One can readily see that a two-spin chiral interaction, the bulk DMI with the energy density  $w_{\rm DMI} \propto \mathbf{m} \cdot (\nabla \times \mathbf{m})$ , arises in point group O but not in  $T_d$ . The bulk DMI is represented by a particular combination of Lifshitz invariants:  $w_{\rm DMI} \propto$  $\mathcal{L}_{yz}^{(x)} + \mathcal{L}_{zx}^{(y)} + \mathcal{L}_{xy}^{(z)}$ , where  $\mathcal{L}_{\alpha\beta}^{(\gamma)} = m_{\alpha}\partial_{\gamma}m_{\beta} - m_{\beta}\partial_{\gamma}m_{\alpha}$ . The key role of  $w_{\rm DMI}$  on the formation of helical spin density waves is well established [33–36]. This interaction is responsible for skyrmion crystal and helical spin phases in MnSi, MnFeSi, FeCoSi, FeGe and in many other magnetic materials [8–11]. It has been recently suggested that 4-spin (and in general multi-spin) chiral interactions may also play an important role in conducting magnets if spin-orbit induced splitting of conduction electron bands becomes comparable with s-d exchange energy [37].

From Table I one can also see that there exist two possible 4-spin chiral interactions in point group O and only one in  $T_d$ , where 2-spin chiral terms are forbidden [38]. We will see that, despite the absence of 2-spin chiral interactions, the collinear state may become unstable also in point group  $T_d$ .

Let us formulate a universal energy functional of  $T_d$  ferromagnet,  $E[\mathbf{m}] = \int d^3\mathbf{r} \ [w(\mathbf{r}) - \mathbf{H} \cdot \mathbf{m}]$ , where  $\mathbf{H}$  stands for external magnetic field measured in energy units, while the energy density of the magnet reads,

$$w = A \sum_{\alpha} (\nabla m_{\alpha})^2 + 8B m_x m_y m_z \nabla \cdot \mathbf{m} + K \sum_{\alpha} m_{\alpha}^4, (1)$$

where we collected all possible terms up to the forth order in local magnetization.

The first term in Eq. (1) represents the usual symmetric exchange, A>0, the second term corresponds to the newly proposed 4-spin chiral interaction discussed above, and the last term is the cubic anisotropy. Throughout the Letter we assume that the ferromagnet is well below the Curie temperature, hence  $|\mathbf{m}|=1$ .

In order to see how the 4-spin chiral interaction may induce an instability of the collinear state, we consider a generalized conical ansatz for magnetization vector,

$$\mathbf{m}(\mathbf{r}) = \mathbf{n}\cos\theta + [\mathbf{n}_1\cos(\mathbf{k}\cdot\mathbf{r}) + \mathbf{n}_2\sin(\mathbf{k}\cdot\mathbf{r})]\sin\theta, (2)$$

where  $\mathbf{n_1}$ ,  $\mathbf{n_2}$  and  $\mathbf{n} = \mathbf{n_1} \times \mathbf{n_2}$  are mutually orthogonal unit vectors; the wave vector reversal,  $\mathbf{k} \to -\mathbf{k}$ , is equivalent to  $\mathbf{n_2} \to -\mathbf{n_2}$  (helicity reversal);  $\theta = 0$  corresponds to a collinear state, while  $\theta = \pi/2$  corresponds to a pure helix. Remarkably, the brute force numerical minimization of the energy functional of Eq. (1) performed recently by the other authors [39] does indeed correspond to magnetic cone ground state that is described by Eq. (2).

The translation  $\mathbf{r} \mapsto \mathbf{r} + \Delta \mathbf{r}$  is equivalent in Eq. (2) to a rotation of the reference frame through the angle  $\mathbf{k} \cdot \Delta \mathbf{r}$  about  $\mathbf{n}$  direction, which is the direction of averaged magnetization in the cone. Translational symmetry implies the existence of a Goldstone mode involving the rotation of spins about  $\mathbf{n}$ .

We further substitute Eq. (2) into Eq. (1) and average the result over the phase  $\mathbf{k} \cdot \Delta \mathbf{r}$  to obtain a Landau energy density  $\mathcal{E} = E/V$ . The latter becomes a function of the parameters  $\mathbf{k}$ ,  $\mathbf{n}$  and  $\theta$  of the conical state (2)

$$\mathcal{E} = A k^2 \sin^2 \theta - B \mathbf{k} \cdot \mathbf{v}(\mathbf{n}) \sin^2 \theta \left(1 - 5 \cos^2 \theta\right) + K \left[u_1(\theta) + u_2(\theta)c(\mathbf{n})\right] - \mathbf{n} \cdot \mathbf{H} \cos \theta, \tag{3}$$

where we introduced

$$\mathbf{v}(\mathbf{n}) = (n_x(n_y^2 - n_z^2), n_y(n_z^2 - n_x^2), n_z(n_x^2 - n_y^2)), \quad (4a)$$

$$c(\mathbf{n}) = 3(n_y^2 n_z^2 + n_z^2 n_x^2 + n_x^2 n_y^2), \tag{4b}$$

$$u_1(\theta) = \cos^4 \theta + (3/4)\sin^4 \theta,$$
 (4c)

$$u_2(\theta) = 2\cos^2\theta \sin^2\theta - (2/3)\cos^4\theta - (1/4)\sin^4\theta.$$
 (4d)

Note that the transverse polarization condition of the conical spiral  $\mathbf{n} \cdot \mathbf{v}(\mathbf{n}) = 0$  follows directly from Eq. (4a).

The four-spin interaction sets the energy scale  $B^2/A$  that defines the non-collinear order. After convenient rescaling

$$\mathbf{k} = B\tilde{\mathbf{k}}/A, \quad \mathbf{H} = B^2\tilde{\mathbf{H}}/A, \quad K = B^2\tilde{K}/A,$$
 (5)

the energy density of Eq. (3) takes the form

$$\mathcal{E} = \frac{B^2}{A} \left[ (\tilde{\mathbf{k}} - \tilde{\mathbf{k}}_0)^2 \sin^2 \theta + u(\mathbf{n}, \theta) - \mathbf{n} \cdot \tilde{\mathbf{H}} \cos \theta \right], \quad (6)$$

where  $\tilde{\mathbf{k}}_0 = \mathbf{v}(\mathbf{n}) (1 - 5\cos^2\theta)/2$  is a characteristic wavevector and  $u(\mathbf{n}, \theta) = \tilde{K} [u_1(\theta) + u_2(\theta)c(\mathbf{n})] - \tilde{k}_0^2 \sin^2\theta$  is

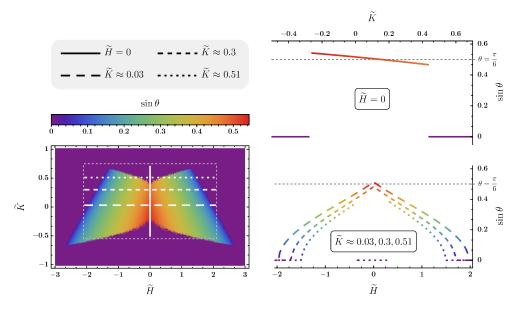


FIG. 2: The color plot is obtained by numerical minimization of the function  $\mathcal{E}(\mathbf{k},\mathbf{n},\theta)$  of Eq. (6) and represents the value of  $\sin\theta$  (the span of magnetic cone) at the global minimum, provided external magnetic field is directed as  $\tilde{\mathbf{H}} = \tilde{H}(0,1,1)/\sqrt{2}$ . Non-collinear magnetic cone state (finite  $\theta$  and  $\mathbf{k}$ ) is realized for moderate values of  $\tilde{K}$  and  $\tilde{H}$ . The upper left panel shows the horizontal crosssection with  $\tilde{H} = 0$ , while the lower left panel shows three vertical crosssections for  $\tilde{K} = 0.03, 0.3,$  and 0.51. The angle  $\theta$  smoothly deviates from zero across the lines  $\tilde{K} = 2 - |\tilde{H}|$ , which correspond to the second order phase transition. Noisy borders for  $\tilde{K} \approx \pm 0.5$  correspond to the first order phase transition from collinear to a non-collinear state with a finite  $\theta$ . The corresponding jumps are also seen in the left panels.

an effective potential. Thus, the vector  $\mathbf{v}$  (if non vanishing) defines the propagation direction of the conical spiral, while the angle  $\theta$  (if it deviates from  $\theta=0$ ) defines the opening angle of the cone.

The energy density (6) has an absolute minimum at  $\mathbf{k} = \mathbf{k}_0 = B\tilde{\mathbf{k}}_0/A$ . The wavevector  $\mathbf{k}_0$  is always perpendicular to  $\mathbf{n}$  since  $\mathbf{v} \cdot \mathbf{n} = 0$ . For the non-collinear phase (i. e. for finite  $k_0$  and  $\theta$  at the minimum), the resulting conical magnetic order is illustrated schematically in Fig. 1. This is in contrast to the bulk DMI  $\propto \mathbf{m} \cdot (\nabla \times \mathbf{m})$  that stabilizes conical or helical states with  $\mathbf{k}_0$  parallel to  $\mathbf{n}$ .

The result of numerical energy minimization in Eq. (6) is illustrated in Fig. 2 by plotting the dependence of  $\sin\theta$  on both  $\tilde{K}$  and  $\tilde{H}$  at the absolute energy minimum. One can see from the minimization procedure that the opening angle  $\theta$  may, at best, only slightly exceed the value  $\pi/6$ , while the pure helix,  $\theta=\pi/2$ , is never reached.

For zero field and small anisotropy,  $-0.28 < \tilde{K} < 0.44$ , we find a non-collinear conical state with  $\mathbf{k} = \mathbf{k}_0$  and  $\theta \approx \pi/6$ . The minimum is reached for  $\mathbf{n} = (0,1,1)/\sqrt{2}$ ,  $\mathbf{v} = (0,1,-1)/2\sqrt{2}$ , and for the other 11 equivalent directions of  $\mathbf{n}$  that are related by the rotation symmetries of the  $T_d$  point group (see Table ?? of the Supplemental material [40]).

In the limit of large anisotropy, the ground state is collinear. For example, for zero field one finds the minimal energy density,  $\mathcal{E}=K/3$  for  $\tilde{K}>0.44$  with the magnetization along a body diagonal such as  $\mathbf{n}=(1,1,1)/\sqrt{3}$ , and  $\mathcal{E}=K$  for  $\tilde{K}<-0.28$  with the magnetization along  $\mathbf{n}=(0,0,1)$  and symmetry equivalents. An external magnetic field applied in  $\langle 011 \rangle$  (or any equivalent) direction can additionally stabilize

the non-collinear state as can be indeed seen in Fig. 2. For example, increasing magnetic field in  $\langle 011 \rangle$  direction for a system with  $\tilde{K}=0.51$  induces the first order phase transition to a non-collinear phase as it is illustrated in the right bottom panel in Fig. 2.

Generally, the angle  $\theta$  deviates smoothly from zero across the lines  $\tilde{K}=2-|\tilde{H}|$  indicating a second order phase transition. The noisy borders of the color plot in Fig. 2 correspond to the first order transition that is characterized by the competition of minima at finite  $\theta$  and  $\theta=0$  (see also the left panels).

Let us now investigate how the four-spin chiral interaction may affect the magnon spectra. To that end we linearize Landau-Lifshitz equation  $\partial \mathbf{m}/\partial t = \mathbf{H}_{\rm eff} \times \mathbf{m}$  with respect to a small variation  $\delta \mathbf{m}$ . We consider a collinear phase, where the unit vector  $\mathbf{n}$  yields the equation  $(\mathbf{H} - 4K\mathbf{n}^{o3}) \times \mathbf{n} = 0$  with  $\mathbf{n}^{o3} = (n_x^3, n_y^3, n_z^3)$ . Instead of solving the resulting cubic equation we introduce the Lagrange multiplier  $\lambda = \lambda(\mathbf{H}, K)$  that is set by the algebraic equation

$$\mathbf{H}_{\text{eff}} = \mathbf{H} - 4K\mathbf{n}^{o3} - \lambda\mathbf{n} = 0, \tag{7}$$

alongside with two independent components of the vector  $\mathbf{n}$ . Using the ansatz  $\mathbf{m} = \mathbf{n} + \delta \mathbf{m} \exp(i\omega_{\mathbf{q}}t - i\mathbf{q} \cdot \mathbf{r})$  with  $\mathbf{n} \cdot \delta \mathbf{m} = 0$ , we, then, obtain the magnon dispersion [40]

$$\omega_{\mathbf{q}} = \sqrt{(\Omega_q + 4cK)^2 + 16K^2(d^2 - c^2)} - 8B\mathbf{v} \cdot \mathbf{q},$$
 (8)

where  $\Omega_q = 2A q^2 + \lambda$ , v and c are defined in Eqs. (4a) and (4b), correspondingly, and  $d = 3\sqrt{3} n_x n_y n_z$ .

For  $H \gg |K|$ , one finds  $\mathbf{n} = \mathbf{H}/H$ , hence  $\lambda = H$  and

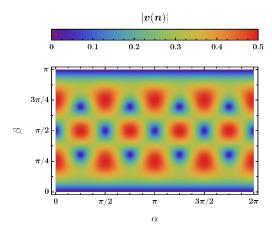


FIG. 3: Absolute value of the vector  $\mathbf{v}$  that defines the asymmetry of the magnon dispersion  $\delta \omega_{\mathbf{q}} = -16B\mathbf{v} \cdot \mathbf{q}$  as the function of the magnetization direction  $\mathbf{n} = (\cos \alpha \sin \beta, \sin \alpha \sin \beta, \cos \beta)$ .

Eq. (8) is reduced to

$$\omega_{\mathbf{q}}|_{H\gg K} = 2A(\mathbf{q} - \mathbf{q}_0)^2 + H - 8B^2v^2/A,$$
 (9)

where  $\mathbf{q}_0 = (2B/A)\mathbf{v}$ .

The wave-vector  $\mathbf{q}_0$  defines the effect of the four-spin chiral interaction on magnon spectra. For  $\theta=0$  one formally finds  $\mathbf{q}_0=-\mathbf{k}_0$ , even though the wave vector  $\mathbf{k}_0$  of the spiral is irrelevant in the collinear phase.

Thus, the coefficient B can be quantified by measuring the difference  $\delta\omega_{\bf q}=\omega_{\bf q}-\omega_{\bf -q}=-16B{\bf v}\cdot{\bf q}$  for the wave-vector  ${\bf q}$  that is orthogonal to the magnetization direction  ${\bf n}$ , provided the vector  ${\bf v}$  is finite. This clarifies the meaning of the vector  ${\bf v}$  in collinear phase as the vector that defines the asymmetry of magnon dispersion.

The largest value of  $\delta\omega_{\mathbf{q}}$  is observed for magnon wave vectors  $\mathbf{q}$  that are parallel to  $\mathbf{v}$ , and, consequently, perpendicular to  $\mathbf{n}$ . This is again in sharp contrast to the effect of the bulk DMI for which  $\mathbf{q}_0 \propto \mathbf{n}$ .

To maximize the effect of the four-spin term one needs to drive the length of the vector  ${\bf v}$  to its maximal value v=1/2. This can be achieved again by applying an external field in the direction  $\langle 011 \rangle$  or in any other equivalent crystallographic direction. In this case, one finds a particularly simple result  $\delta \omega_{\bf q} \propto q_y - q_z$  for any K>0.

The absolute value of the vector  $\mathbf{v}$  is illustrated in Fig. 3 for different magnetization directions  $\mathbf{n}$ .

It is worth noting that the coefficient B does not enter the magnon dispersion in the absence of external field. Indeed, for  $\mathbf{H}=0$ , the ground state magnetization  $\mathbf{n}$  is set by the sign of the anisotropy constant only. For K>0 one finds  $\mathbf{n}=(1,1,1)/\sqrt{3}$ , which corresponds to  $\lambda=-4K/3$ ,  $\mathbf{v}=0$ , c=d=1. Therefore, the magnon dispersion reads  $\omega_q=2A\,q^2+8K/3$ . For K<0 one finds  $\mathbf{n}=(0,0,1)$ ,  $\lambda=-4K$ ,  $\mathbf{v}=0$ , c=d=0, hence  $\omega_q=2A\,q^2+4|K|$ .

Thus, the new 4-spin chiral interaction term in cubic crystals with broken inversion symmetry does indeed lead to non-reciprocal magnon dispersion. Similarly to the bulk DMI, it

breaks the symmetry with respect to the wave vector reversal  $\mathbf{q} \to -\mathbf{q}$ , but in a direction of  $\mathbf{q}$  that is orthogonal to magnetization. The bulk DMI leads to  $\mathbf{q} \to -\mathbf{q}$  non-reciprocity in the direction parallel to magnetization.

It is evident from Eqs. (8,9) that the four spin chiral interaction shifts the minimum of magnon energy  $\mathbf{q} \propto \mathbf{q}_0$ . Moreover, the results suggest that the frequency  $\omega_{\mathbf{q}}$  becomes negative at least for  $H \simeq 2B^2/A$ , provided anisotropy is sufficiently weak,  $|K| \lesssim B^2/A$ . Such negative values of  $\omega_{\mathbf{q}}$  are unphysical and indicate an instability of the collinear order. Low-energy magnons in the presence of non-collinear periodic ground state form a banded spectrum that we do not analyze in this Letter.

So far we have discussed the 4-spin chiral interaction in the continuum theory limit. One possible Heisenberg equivalent of this interaction can be constructed on a pyrochlore lattice. Let us consider the four vertices of a regular tetrahedron with coordinates  $\mathbf{r}_0 = (0,0,0)$ ,  $\mathbf{r}_1 = (0,-a/4,-a/4)$ ,  $\mathbf{r}_2 = (-a/4,0,-a/4)$ , and  $\mathbf{r}_3 = (-a/4,-a/4,0)$ , where a is the cubic lattice constant of the pyrochlore lattice. We further define the four unit vectors pointing from the center of the tetrahedron to the respective sites:

$$\mathbf{n}_0 = (+1, +1, +1)/\sqrt{3}, \quad \mathbf{n}_1 = (+1, -1, -1)/\sqrt{3},$$
  
 $\mathbf{n}_2 = (-1, +1, -1)/\sqrt{3}, \quad \mathbf{n}_3 = (-1, -1, +1)/\sqrt{3}, \quad (10)$ 

which satisfy  $\mathbf{n}_i \cdot \mathbf{n}_j = (4\delta_{ij} - 1)/3$ .

With these notations, the four-spin chiral exchange interaction is given by the following energy:

$$U_{4} = (\mathbf{n}_{0} \cdot \mathbf{S}_{0})(\mathbf{e}_{x} \cdot \mathbf{S}_{1})(\mathbf{e}_{y} \cdot \mathbf{S}_{2})(\mathbf{e}_{z} \cdot \mathbf{S}_{3})$$

$$+ (\mathbf{e}_{x} \cdot \mathbf{S}_{0})(\mathbf{n}_{1} \cdot \mathbf{S}_{1})(-\mathbf{e}_{z} \cdot \mathbf{S}_{2})(-\mathbf{e}_{y} \cdot \mathbf{S}_{3})$$

$$+ (\mathbf{e}_{y} \cdot \mathbf{S}_{0})(-\mathbf{e}_{z} \cdot \mathbf{S}_{1})(\mathbf{n}_{2} \cdot \mathbf{S}_{2})(-\mathbf{e}_{x} \cdot \mathbf{S}_{3})$$

$$+ (\mathbf{e}_{z} \cdot \mathbf{S}_{0})(-\mathbf{e}_{y} \cdot \mathbf{S}_{1})(-\mathbf{e}_{x} \cdot \mathbf{S}_{2})(\mathbf{n}_{3} \cdot \mathbf{S}_{3}), \quad (11)$$

where  $\mathbf{e}_{\alpha}$  stand for the unit vectors in the chosen coordinate frame,  $\alpha = x, y, z$ , while  $\mathbf{S}_i$  stand for spins on respective lattice cites. The gradient expansion of  $U_4$  to the lowest order,

$$\mathbf{S}_{i}(\mathbf{r}_{i}) = S\left[\mathbf{m}(0) + (\mathbf{r}_{i} \cdot \nabla)\mathbf{m}(\mathbf{r})|_{\mathbf{r}=0} + \ldots\right], \tag{12}$$

and subsequent integration by parts yields the chiral 4-spin term with  $B=-a/8S^4$ .

In conclusion, we suggest the existence of a four-spin chiral magnetic interaction that may be responsible for the appearance of non-collinear magnetic order in ferromagnets with the  $T_d$  point-group symmetry. Even though, the DMI interaction between pairs of spins is possible in  $T_d$  magnets it does not lead to linear in gradient terms in micromagnetic energy [28–31]. We demonstrate that, in this case, four-spin chiral interactions become more important. Similar situation arises in crystals with  $D_{3h}$  and  $C_{3h}$  point group symmetries that are rather common among two dimensional magnets. Thus, taking into account possible four-spin chiral exchange interactions is important for understanding non-collinear magnetic order in these systems.

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