Spiral plane flops in frustrated helimagnets in external magnetic field

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We discuss theoretically frustrated Heisenberg spiral magnets in magnetic field **H**. We demonstrate that small anisotropic spin interactions (single-ion biaxial anisotropy or dipolar forces) select the plane in which spins rotate (spiral plane) and can lead to the spiral plane flop upon in-plane field increasing. Expressions for the critical fields H_{flop} are derived. It is shown that measuring of H_{flop} is an efficient and simple method of quantifying the anisotropy in the system (as the measurement of spin-flop fields in collinear magnets with axial anisotropy). Corresponding recent experiments are considered in spiral magnets some of which are multiferroics of spin origin.

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I. INTRODUCTION

Multiferroics with coexisting magnetic and ferroelectric orders have attracted a lot of attention recently [1]. The possibility to realize cross control between magnetism and electricity in such compounds would lead to many desirable applications. For instance, strong enough magnetoelectric coupling would allow us to manage magnetic memory by electric field [2]. In so-called multiferroics of spin origin ferroelectricity is induced by some types of magnetic ordering and magnetoelectric coupling in such compounds is discovered to be strong [1,3,4]. There are three main mechanisms of ferroelectricity of spin origin: exchange-striction mechanism, inverse Dzyaloshinskii-Moriya (DM) mechanism, and spin-dependent *p-d* hybridization mechanism [1]. Noncollinear spin ordering induced, e.g., by frustration is indispensable for the second and the third mechanisms.

While the appearance of noncollinear magnetic textures in frustrated helimagnets is mainly caused by the competition between different exchange couplings, fine details of the spin ordering depend on usually weak low-symmetry relativistic interactions (anisotropy and dipolar forces). In particular, they fix the plane in which spins rotate (spiral plane) and, in turn, the direction of the electric polarization **P** which is related with the spiral plane orientation [1]. The smallness of the anisotropic interactions opens a way to handle orientation of the spiral plane and **P** by, e.g., small magnetic field [4].

It is well known that in collinear antiferromagnet a spin-flop transition of the first-order type takes place in magnetic field **H** applied along the easy axis [5,6]. Sublattices magnetizations stay parallel to **H** at $H < H_{flop}$ and they become nearly perpendicular to the field after the flop at $H > H_{flop}$ forming a canted antiferromagnetic spin arrangement. Well known relations are $H_{flop} \sim S\sqrt{DJ} \ll H_s \sim SJ$, where S is the spin value, $D \ll J$ is the anisotropy value, J is the exchange coupling constant, and H_s is the saturation field [5,6].

A similar phenomenon has been observed both experimentally (see, e.g., Refs. [7–9]) and numerically (see, e.g., Refs. [10,11]) in frustrated Heisenberg spiral magnets. Without anisotropy, the spiral plane is perpendicular to any finite **H**. On the other hand, the spiral plane can be fixed by anisotropic interactions so that the spiral order is only slightly deformed by small in-plane magnetic field. However the spiral plane flops at some critical field H_{flop} and becomes perpendicular to **H** at $H > H_{flop}$ as it is illustrated by Fig. 1. To the best of our knowledge, the spiral plane flops have not been described analytically so far. It is the aim of the present paper to fill up this gap.

In Sec. II we discuss in details a simple model of frustrated Heisenberg magnet with small single-ion biaxial anisotropy. At $H = 0$, a slightly distorted spiral ordering arises in the classical ground state of the system, where spins rotate in the plane containing easy and middle axes. Spin arrangement and expressions for H_{flop} are found analytically for field directed along principal axes. We show that similar to collinear magnets $H_{flop} \sim S\sqrt{DJ} \ll H_s \sim SJ$.

It is well known that dipolar forces can be the main anisotropic interaction in helimagnets containing magnetic ions with $L = 0$ (e.g., Mn^{2+} and Eu^{2+}) in which anisotropy of spin-orbit origin is strongly suppressed. In particular, magnetodipolar interaction was shown to be important for the description of transitions in many multiferroics of spin origin [12]. Then, we discuss in Sec. III the spiral plane flops in frustrated Heisenberg magnets with dipolar interaction. The results obtained are qualitatively similar to those observed for the system with biaxial anisotropy.

In Sec. IV we analyze the systems in arbitrary directed magnetic field. We find that spiral plane flops can happen only if the external magnetic field lies in the spiral plane stabilized at $H = 0$. In contrast to collinear magnets, where the spin flop takes place only for a very narrow range of field directions along the easy axis [13], the spiral plane flop occurs for any field direction within the plane. Corresponding expressions for H_{flop} are derived.

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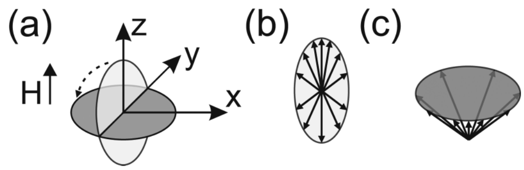

FIG. 1. (a) Illustration of the flop of the plane in which spins rotate (spiral plane) upon in-plane field $\mathbf{H}$ increasing. The spiral plane containing the easy z and the middle y axes flops to the xy plane. (b) Spins lie in the yz plane at small field and form a helix slightly deformed by the field and anisotropy. (c) Spin arrangement after the flop (conical spiral).

In Sec. V using our theory we describe experimentally observed field-induced spiral plane reorientations in several compounds including multiferroics of spin origin. We believe that our results could be useful for interpreting experimental data in many frustrated helimagnets in external magnetic field. We point out in conclusion (Sec. VI) that measurement of H_{flop} provides an easy and efficient way to quantify the anisotropy in frustrated helical magnets.

II. SPIRAL PLANE FLOP IN FRUSTRATED HELIMAGNET WITH BIAXIAL ANISOTROPY

In this section we consider a simple model containing frustrating exchange interaction and a small single-ion biaxial anisotropy. We assume that the frustration leads to a spiral in the classical ground state.

A. Model and general consideration

The system Hamiltonian reads as

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_{ex} + \mathcal{H}_{an} + \mathcal{H}_z,\\ \mathcal{H}_{ex} &= -\frac{1}{2}\sum_{i,j} J_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j),\\ \mathcal{H}_{an} &= -\sum_i\left[D\left(S_i^z\right)^2 + E\left(S_i^y\right)^2\right],\\ \mathcal{H}_z &= -\sum_i(\mathbf{h}\cdot\mathbf{S}_i),\end{aligned}\tag{1}$$

where $D > E > 0$, $\mathbf{h} = g\mu_B\mathbf{H}$, x and z axes are the hard and the easy ones, respectively, there is one spin in a unit cell, and the lattice is assumed arbitrary in all general derivations below. After the Fourier transform

$$\mathbf{S}_j = \frac{1}{\sqrt{N}}\sum_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} e^{i\mathbf{q}\mathbf{R}_j},\tag{2}$$

where N is the number of spins in the lattice, Hamiltonian (1) acquires the following form:

$$\mathcal{H}_{ex} = -\frac{1}{2}\sum_{\mathbf{q}} J_{\mathbf{q}}(\mathbf{S}_{\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{q}}),\tag{3}$$

$$\mathcal{H}_{an} = -\sum_{\mathbf{q}}\left[DS_{\mathbf{q}}^zS_{-\mathbf{q}}^z + ES_{\mathbf{q}}^yS_{-\mathbf{q}}^y\right],\tag{4}$$

$$\mathcal{H}_z = -\sqrt{N}(\mathbf{h}\cdot\mathbf{S}_0).\tag{5}$$

We assume that $J_{\mathbf{q}}$ has two equivalent maxima at $\mathbf{q} = \pm\mathbf{k}$ so that a plane spiral arises in the classical ground state at $h = D = E = 0$. The plane at which spins lie can be fixed by small anisotropy and/or magnetic field which can also distort the spiral order.

For theoretical description of a cone helix, we introduce the local right-hand orthogonal coordinate frame at the jth site (see Ref. [14])

$$\begin{aligned}\hat{\zeta}_j &= (\hat{a}\cos\mathbf{kR}_j + \hat{b}\sin\mathbf{kR}_j)\cos\alpha + \hat{c}\sin\alpha,\\ \hat{\eta}_j &= -\hat{a}\sin\mathbf{kR}_j + \hat{b}\cos\mathbf{kR}_j,\\ \hat{\xi}_j &= -(\hat{a}\cos\mathbf{kR}_j + \hat{b}\sin\mathbf{kR}_j)\sin\alpha + \hat{c}\cos\alpha,\end{aligned}\tag{6}$$

where $\hat{a}$, $\hat{b}$, and $\hat{c}$ are some mutually orthogonal unit vectors, and α is a cone angle ($\alpha = 0$ in the plane spiral). Then, the spin at the jth site is expressed as

$$\mathbf{S}_j = S_j^\zeta\hat{\zeta}_j + S_j^\eta\hat{\eta}_j + S_j^\xi\hat{\xi}_j,\tag{7}$$

where

$$\begin{aligned}S_j^\zeta &= S - a_j^\dagger a_j,\\ S_j^\eta &\simeq \sqrt{\frac{S}{2}}(a_j + a_j^\dagger),\\ S_j^\xi &\simeq i\sqrt{\frac{S}{2}}(a_j^\dagger - a_j)\end{aligned}\tag{8}$$

is the Holstein-Primakoff transformation [15] in which square roots are replaced by unity. It is convenient to rewrite local basis vectors (6) as

$$\begin{aligned}\hat{\zeta}_j &= (\mathbf{A}e^{i\mathbf{kR}_j} + \mathbf{A}^*e^{-i\mathbf{kR}_j})\cos\alpha + \hat{c}\sin\alpha\\ \hat{\eta}_j &= i\mathbf{A}e^{i\mathbf{kR}_j} - i\mathbf{A}^*e^{-i\mathbf{kR}_j}\\ \hat{\xi}_j &= -(\mathbf{A}e^{i\mathbf{kR}_j} + \mathbf{A}^*e^{-i\mathbf{kR}_j})\cos\alpha + \hat{c}\cos\alpha,\end{aligned}\tag{9}$$

where auxiliary vectors $\mathbf{A} = (\hat{a} - i\hat{b})/2$ and $\mathbf{A}^* = (\hat{a} + i\hat{b})/2$ are introduced. Then, we have from Eqs. (7) and (9) after Fourier transform (2)

$$\mathbf{S}_{\mathbf{q}} = S_{\mathbf{q}}^A\mathbf{A} + S_{\mathbf{q}}^{A^*}\mathbf{A}^* + S_{\mathbf{q}}^c\hat{c},\tag{10}$$

where

$$\begin{aligned}S_{\mathbf{q}}^A &= S_{\mathbf{q-k}}^\zeta\cos\alpha + iS_{\mathbf{q-k}}^\eta - S_{\mathbf{q-k}}^\xi\sin\alpha,\\ S_{\mathbf{q}}^{A^*} &= S_{\mathbf{q+k}}^\zeta\cos\alpha - iS_{\mathbf{q+k}}^\eta - S_{\mathbf{q+k}}^\xi\sin\alpha,\\ S_{\mathbf{q}}^c &= S_{\mathbf{q}}^\zeta\sin\alpha + S_{\mathbf{q}}^\xi\cos\alpha.\end{aligned}\tag{11}$$

Substituting Eqs. (10) and (11) into Eqs. (3) and (4), one obtains

$$\begin{aligned}\mathcal{H}_{ex} = -\frac{1}{2}\sum_{\mathbf{q}}\Big[&(\sin^2\alpha J_{\mathbf{q}} + \cos^2\alpha J_{\mathbf{q},\mathbf{k}})S_{\mathbf{q}}^\zeta S_{-\mathbf{q}}^\zeta\\ &+ J_{\mathbf{q},\mathbf{k}}S_{\mathbf{q}}^\eta S_{-\mathbf{q}}^\eta + (\cos^2\alpha J_{\mathbf{q}} + \sin^2\alpha J_{\mathbf{q},\mathbf{k}})S_{\mathbf{q}}^\xi S_{-\mathbf{q}}^\xi\\ &+ \sin\alpha\cos\alpha(J_{\mathbf{q}} - J_{\mathbf{q},\mathbf{k}})\big(S_{\mathbf{q}}^\zeta S_{-\mathbf{q}}^\xi + S_{\mathbf{q}}^\xi S_{-\mathbf{q}}^\zeta\big)\\ &+ i\cos\alpha N_{\mathbf{q},\mathbf{k}}\big(S_{\mathbf{q}}^\eta S_{-\mathbf{q}}^\zeta - S_{\mathbf{q}}^\zeta S_{-\mathbf{q}}^\eta\big)\\ &+ i\sin\alpha N_{\mathbf{q},\mathbf{k}}\big(S_{\mathbf{q}}^\xi S_{-\mathbf{q}}^\eta - S_{\mathbf{q}}^\eta S_{-\mathbf{q}}^\xi\big)\Big],\end{aligned}\tag{12}$$

where $J_{\mathbf{q},\mathbf{k}} = (J_{\mathbf{q}+\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}})/2$ and $N_{\mathbf{q},\mathbf{k}} = (J_{\mathbf{q}+\mathbf{k}} - J_{\mathbf{q}-\mathbf{k}})/2$, and

$$\begin{aligned}\mathcal{H}_{an} = &-D\sum_{\mathbf{q}}\left(S^A_{\mathbf{q}}A_z + S^{A^*}_{\mathbf{q}}A^*_z + S^c_{\mathbf{q}}c_z\right)\\ &\times\left(S^A_{-\mathbf{q}}A_z + S^{A^*}_{-\mathbf{q}}A^*_z + S^c_{-\mathbf{q}}c_z\right)\\ &- E\sum_{\mathbf{q}}\left(S^A_{\mathbf{q}}A_y + S^{A^*}_{\mathbf{q}}A^*_y + S^c_{\mathbf{q}}c_y\right)\\ &\times\left(S^A_{-\mathbf{q}}A_y + S^{A^*}_{-\mathbf{q}}A^*_y + S^c_{-\mathbf{q}}c_y\right).\end{aligned} \tag{13}$$

B. Ground-state energy of the plane helix at finite anisotropy and $h = 0$

At zero field, the spin texture in the classical ground state is a slightly distorted (due to the anisotropy) spiral in which spins lie in the yz plane. Then, we take $\hat{a} = \mathbf{e}_y$, $\hat{b} = \mathbf{e}_z$, and $\hat{c} = \mathbf{e}_x$ in Eq. (6), where $\mathbf{e}_{x,y,z}$ are unit vectors directed along corresponding axes. To find the ground-state energy and the spin arrangement, we substitute Eqs. (8) into Eqs. (12) and (13) and put $\alpha = 0$. One obtains the Hamiltonian in the following form:

$$\mathcal{H} = \mathcal{E}^{yz}_0 + \mathcal{H}_1 + \mathcal{H}_2. \tag{14}$$

Henceforth $\mathcal{E}_0$ denotes part of the Hamiltonian without bosonic operators (constant term), $\mathcal{H}_1 = \mathcal{H}_{1an} + \mathcal{H}_{1z}$ contains linear in bosonic operators terms which arise from the anisotropy and Zeeman energy, and $\mathcal{H}_2$ is bilinear in bosonic operators. At $h = 0$ ($\mathcal{H}_{1z} = 0$) one obtains

$$\frac{1}{N}\mathcal{E}^{yz}_0 = -\frac{S^2 J_{\mathbf{k}}}{2} - \frac{S^2(D+E)}{2}, \tag{15}$$

$$\frac{1}{\sqrt{N}}\mathcal{H}_{1an} = i(D-E)\left(\frac{S}{2}\right)^{3/2}\left(a_{-2\mathbf{k}} - a_{2\mathbf{k}} + a^\dagger_{2\mathbf{k}} - a^\dagger_{-2\mathbf{k}}\right), \tag{16}$$

$$\mathcal{H}_2 = \sum_{\mathbf{q}}\left(C_{\mathbf{q}}a^\dagger_{\mathbf{q}}a_{\mathbf{q}} + B_{\mathbf{q}}\frac{a_{\mathbf{q}}a_{-\mathbf{q}} + a^\dagger_{\mathbf{q}}a^\dagger_{-\mathbf{q}}}{2}\right), \tag{17}$$

where

$$C_{\mathbf{q}} = \frac{S}{2}(2J_{\mathbf{k}} - J_{\mathbf{q},\mathbf{k}} - J_{\mathbf{q}} + D + E), \tag{18}$$

$$B_{\mathbf{q}} = -\frac{S}{2}(J_{\mathbf{q},\mathbf{k}} - J_{\mathbf{q}} + D + E). \tag{19}$$

We omit the so-called Umklapp terms in $\mathcal{H}_2$ which have the form $a^\dagger_{\mathbf{q}}a_{\mathbf{q}\pm2\mathbf{k}}$, $a_{\mathbf{q}}a_{-\mathbf{q}\pm2\mathbf{k}}$, and $a^\dagger_{\mathbf{q}}a^\dagger_{-\mathbf{q}\pm2\mathbf{k}}$ and which are proportional to $D-E$. As it is explained below, their contribution to the ground-state energy and the spin arrangement is small.

Terms linear in Bose operators $\mathcal{H}_{1an}$ arise in Hamiltonian (14) because we assume in derivation of Eqs. (12) and (13) that the spiral ordering is undisturbed [see Eq. (6)]. To eliminate the linear terms from the Hamiltonian (14), we perform the following shift in operators:

$$\begin{aligned}&a_{2\mathbf{k}} \mapsto \rho_+ e^{i\varphi_+} + a_{2\mathbf{k}}, \quad a^\dagger_{2\mathbf{k}} \mapsto \rho_+ e^{-i\varphi_+} + a^\dagger_{2\mathbf{k}},\\ &a_{-2\mathbf{k}} \mapsto \rho_- e^{i\varphi_-} + a_{-2\mathbf{k}}, \quad a^\dagger_{-2\mathbf{k}} \mapsto \rho_- e^{-i\varphi_-} + a^\dagger_{-2\mathbf{k}},\end{aligned} \tag{20}$$

where $\rho_\pm$ and $\varphi_\pm$ are real constants. Linear terms vanish if the following equalities hold:

$$\begin{aligned}&-i\frac{D-E}{2}S\sqrt{\frac{S}{2}}\sqrt{N} + C_{2\mathbf{k}}\rho_+ e^{-i\varphi_+} + B_{2\mathbf{k}}\rho_- e^{i\varphi_-} = 0,\\ &i\frac{D-E}{2}S\sqrt{\frac{SN}{2}} + C_{2\mathbf{k}}\rho_- e^{-i\varphi_-} + B_{2\mathbf{k}}\rho_+ e^{i\varphi_+} = 0.\end{aligned} \tag{21}$$

A solution of Eqs. (21) has the form

$$\begin{aligned}&\varphi_+ = -\varphi_- = \pi/2,\\ &\rho_+ = \rho_- = -\sqrt{N}\sqrt{\frac{S}{2}}\frac{D-E}{J_{\mathbf{k}} - J_{3\mathbf{k}}}.\end{aligned} \tag{22}$$

A correction $\Delta\mathcal{E}^{yz}_{an}$ to the constant $\mathcal{E}^{yz}_0$ also arises after shift (20) which has the form $-N(C_{2\mathbf{k}}\rho_+^2 + C_{-2\mathbf{k}}\rho_-^2 + (B_{2\mathbf{k}} + B_{-2\mathbf{k}})\rho_+\rho_-)/2$. Substituting Eqs. (22) to this formula, one obtains

$$\frac{1}{N}\Delta\mathcal{E}^{yz}_{an} = -\frac{S^2(D-E)^2}{2(J_{\mathbf{k}} - J_{3\mathbf{k}})}. \tag{23}$$

One has for the spin arrangement from Eqs. (8)–(11) after taking into account shift (20) and Eqs. (22)

$$\begin{aligned}\mathbf{S}_j = S\Bigg[&\mathbf{e}_z\left(1 + \frac{D-E}{J_{\mathbf{k}} - J_{3\mathbf{k}}}\right)\sin\mathbf{k}\mathbf{R}_j\\ &+ \mathbf{e}_y\left(1 - \frac{D-E}{J_{\mathbf{k}} - J_{3\mathbf{k}}}\right)\cos\mathbf{k}\mathbf{R}_j\\ &+ \frac{D-E}{J_{\mathbf{k}} - J_{3\mathbf{k}}}(\mathbf{e}_z\sin3\mathbf{k}\mathbf{R}_j + \mathbf{e}_y\cos3\mathbf{k}\mathbf{R}_j)\Bigg].\end{aligned} \tag{24}$$

Then, we obtain that the in-plane anisotropy leads to an elliptical distortion of the spiral and to the third harmonic of $\mathbf{k}$.

Umklapp terms would complicate considerably the above analysis. In particular, one would have to consider shifts of the form (20) for momenta $2n\mathbf{k}$, where n is any integer. As a result, an infinite set of equations would arise instead of Eqs. (21). Fortunately, Umklapp terms are proportional to $D-E$. Then, it is easy to realize that their contribution to Eqs. (23) and (24) is of the third order in small parameter $(D-E)/J$ which can be safely neglected.

C. Ground-state energy of the plane helix at finite anisotropy and in-plane magnetic field

Let us take into account the in-plane magnetic field directed along the z axis. One obtains from Eqs. (5) and (8)–(11) the following contribution to $\mathcal{H}_1$:

$$\frac{1}{\sqrt{N}}\mathcal{H}_{1z} = -\frac{h}{2}\sqrt{\frac{S}{2}}\left(a_{\mathbf{k}} + a_{-\mathbf{k}} + a^\dagger_{\mathbf{k}} + a^\dagger_{-\mathbf{k}}\right) \tag{25}$$

which contains Bose-operators on momenta $\pm\mathbf{k}$ rather than $\pm2\mathbf{k}$ [cf. Eq. (16)]. To eliminate $\mathcal{H}_{1z}$, we perform a shift similar to Eq. (20)

$$\begin{aligned}&a_{\mathbf{k}} \mapsto \tilde{\rho}_+ e^{i\tilde{\varphi}_+} + a_{\mathbf{k}}, \quad a^\dagger_{\mathbf{k}} \mapsto \tilde{\rho}_+ e^{-i\tilde{\varphi}_+} + a^\dagger_{\mathbf{k}},\\ &a_{-\mathbf{k}} \mapsto \tilde{\rho}_- e^{i\tilde{\varphi}_-} + a_{-\mathbf{k}}, \quad a^\dagger_{-\mathbf{k}} \mapsto \tilde{\rho}_- e^{-i\tilde{\varphi}_-} + a^\dagger_{-\mathbf{k}}.\end{aligned} \tag{26}$$

Linear terms vanish when

$$-\frac{h}{2}\sqrt{\frac{SN}{2}}+C_{\mathbf{k}}\tilde{\rho}_+e^{-i\tilde{\varphi}_+}+B_{\mathbf{k}}\tilde{\rho}_-e^{i\tilde{\varphi}_-}=0,$$
$$-\frac{h}{2}\sqrt{\frac{SN}{2}}+C_{\mathbf{k}}\tilde{\rho}_-e^{-i\tilde{\varphi}_-}+B_{\mathbf{k}}\tilde{\rho}_+e^{i\tilde{\varphi}_+}=0 \tag{27}$$

that gives

$$\tilde{\varphi}_+=\tilde{\varphi}_-=0,$$
$$\tilde{\rho}_+=\tilde{\rho}_-=\frac{h\sqrt{NS/2}}{S(2J_{\mathbf{k}}-J_{\mathbf{0}}-J_{2\mathbf{k}})}. \tag{28}$$

The correction to the ground-state energy appearing as a result of the shift (26) reads as

$$\frac{1}{N}\Delta\mathcal{E}_z^{yz}=-\frac{h^2}{2(2J_{\mathbf{k}}-J_{\mathbf{0}}-J_{2\mathbf{k}})}. \tag{29}$$

It can be shown that the correction to the ground-state energy for the field directed along y axis is also given by Eq. (29). Thus, we obtain from Eqs. (15), (23), and (29) for the energy of the spiral in which all spins lie in the yz plane

$$\frac{1}{N}\mathcal{E}^{yz}=-\frac{S^2J_{\mathbf{k}}}{2}-\frac{S^2(D+E)}{2}-\frac{S^2(D-E)^2}{2(J_{\mathbf{k}}-J_{3\mathbf{k}})}$$
$$-\frac{h^2}{2(2J_{\mathbf{k}}-J_{\mathbf{0}}-J_{2\mathbf{k}})}. \tag{30}$$

D. Ground-state energy of conical helix

We calculate now the ground-state energy of the conical spiral in which spins rotate in the xy plane [see Fig. 1(c)]. In this case, all spins are canted towards magnetic field direction (i.e., z axis) and $\alpha \neq 0$. It is convenient to take $\hat{a}=\mathbf{e}_x$, $\hat{b}=\mathbf{e}_y$, and $\hat{c}=\mathbf{e}_z$ in Eq. (6). The angle α is to be chosen to eliminate linear in $a_{\mathbf{0}}$ and $a_{\mathbf{0}}^\dagger$ terms in the Hamiltonian. As usual, these α values minimize the system classical energy having the form

$$\frac{1}{N}\mathcal{E}_0^{xy}=-\frac{S^2(J_{\mathbf{0}}\sin^2\alpha+J_{\mathbf{k}}\cos^2\alpha)}{2}-S^2D\sin^2\alpha$$
$$-\frac{S^2E\cos^2\alpha}{2}-hS\sin\alpha. \tag{31}$$

The minimum of $\mathcal{E}_0^{xy}$ is achieved at

$$\sin\alpha=\frac{h}{S(J_{\mathbf{k}}-J_{\mathbf{0}}-2D+E)}\approx\frac{h}{S(J_{\mathbf{k}}-J_{\mathbf{0}})}. \tag{32}$$

We obtain from Eqs. (31) and (32) in the leading orders in small parameters E/J, D/J, and h/J

$$\frac{1}{N}\mathcal{E}_0^{xy}=-\frac{S^2J_{\mathbf{k}}}{2}-\frac{S^2E}{2}-\frac{h^2}{2(J_{\mathbf{k}}-J_{\mathbf{0}})}. \tag{33}$$

One has also to eliminate terms in the Hamiltonian linear in $a_{\pm2\mathbf{k}}$ and $a_{\pm2\mathbf{k}}^\dagger$ stemming from the anisotropy. Calculations similar to those performed above in Sec. II B lead to the following correction to the ground-state energy [cf. Eq. (23)]:

$$\frac{1}{N}\Delta\mathcal{E}_{an}^{xy}=-\frac{S^2E^2}{2(J_{\mathbf{k}}-J_{3\mathbf{k}})}. \tag{34}$$

Thus, we obtain from Eqs. (33) and (34) for the energy of the conical spiral in which spins rotate in the xy plane

$$\frac{1}{N}\mathcal{E}^{xy}=-\frac{S^2J_{\mathbf{k}}}{2}-\frac{S^2E}{2}-\frac{S^2E^2}{2(J_{\mathbf{k}}-J_{3\mathbf{k}})}$$
$$-\frac{h^2}{2(J_{\mathbf{k}}-J_{\mathbf{0}})}. \tag{35}$$

E. Spiral plane flop in magnetic field

Let us compare now energies $\mathcal{E}^{yz}$ and $\mathcal{E}^{xy}$ of the plane and the conical spirals given by Eqs. (30) and (35), respectively. It is seen that $\mathcal{E}^{yz}<\mathcal{E}^{xy}$ at $h=0$. However, the field correction in Eq. (35) is smaller than that in Eq. (30) because $J_{\mathbf{k}}>J_{2\mathbf{k}}$ (remember, $J_{\mathbf{q}}$ is maximized at $\mathbf{q}=\pm\mathbf{k}$). Thus, $\mathcal{E}^{xy}$ becomes smaller than $\mathcal{E}^{yz}$ at $h>h_{flop}$, where h_{flop} is determined in the leading order in small parameters by the equation

$$S^2D=\frac{h_{flop}^2}{J_{\mathbf{k}}-J_{\mathbf{0}}}-\frac{h_{flop}^2}{2J_{\mathbf{k}}-J_{\mathbf{0}}-J_{2\mathbf{k}}}. \tag{36}$$

Then, the spiral plane flop takes place at the critical field h_{flop} for which we have from Eq. (36)

$$h_{flop}=S\sqrt{D\tilde{J}}, \tag{37}$$

where

$$\tilde{J}=\frac{(J_{\mathbf{k}}-J_{\mathbf{0}})(2J_{\mathbf{k}}-J_{\mathbf{0}}-J_{2\mathbf{k}})}{J_{\mathbf{k}}-J_{2\mathbf{k}}}. \tag{38}$$

Notice that $h_{flop}\sim S\sqrt{DJ}$ is much smaller than the saturation field

$$h_s=S(J_{\mathbf{k}}-J_{\mathbf{0}}) \tag{39}$$

found from Eq. (32) because $h_s\sim SJ$.

The critical field h_{flop} given by Eqs. (37) and (38) is related to h_s as

$$h_{flop}=\sqrt{2SDh_s} \tag{40}$$

if the exchange interaction satisfies the condition $J_{\mathbf{0}}\approx J_{2\mathbf{k}}$ in which case

$$\tilde{J}\approx 2(J_{\mathbf{k}}-J_{\mathbf{0}})=2h_s/S. \tag{41}$$

One expects that the latter equality is fulfilled not so rare as soon as points $\mathbf{q}=\mathbf{0}$ and $\mathbf{q}=2\mathbf{k}$ are symmetric according to $\mathbf{q}=\mathbf{k}$ at which $J_{\mathbf{q}}$ is maximized. Equation (40) may be very useful in determination of the anisotropy value from experimentally obtained values of h_{flop} and h_s. Interestingly, Eq. (40) coincides with the spin-flop field in collinear magnets with small easy-axis anisotropy D. As it follows from the above discussion, one should substitute D by E in Eqs. (37) and (40) if the magnetic field is directed along the y axis.

III. SPIRAL PLANE FLOP IN FRUSTRATED HELIMAGNET WITH DIPOLAR FORCES

In this section, we show that small magnetodipolar interaction has a similar impact on the spiral ordering as the biaxial anisotropy discussed above. The system Hamiltonian has the form (1), where $\mathcal{H}_{an}$ should be replaced by

$$\mathcal{H}_d = \frac{1}{2}\sum_{i,j} D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta,$$
$$\mathcal{D}_{ij}^{\alpha\beta} = \omega_0 \frac{v_0}{4\pi}\left(\frac{1}{R_{ij}^3} - \frac{3R_{ij}^\alpha R_{ij}^\beta}{R_{ij}^5}\right), \tag{42}$$

where v_0 is the unit cell volume and

$$\omega_0 = 4\pi \frac{(g\mu_B)^2}{v_0} \ll J \tag{43}$$

is the characteristic dipolar energy. We have after Fourier transform (2)

$$\mathcal{H}_d = \frac{1}{2}\sum_{\mathbf{q}} \mathcal{D}_{\mathbf{q}}^{\alpha\beta} S_{\mathbf{q}}^\alpha S_{-\mathbf{q}}^\beta. \tag{44}$$

Tensor $\mathcal{D}_{\mathbf{q}}^{\alpha\beta}/2$ has three eigenvalues $\lambda_1(\mathbf{q}) \geqslant \lambda_2(\mathbf{q}) \geqslant \lambda_3(\mathbf{q})$ corresponding to three orthogonal eigenvectors $\mathbf{v}_1(\mathbf{q})$, $\mathbf{v}_2(\mathbf{q})$, and $\mathbf{v}_3(\mathbf{q})$.

At $h = 0$, the classical ground-state energy per spin $-J_{\mathbf{q}} + (\lambda_2(\mathbf{q}) + \lambda_3(\mathbf{q}))/2$ is minimized at an incommensurate vector $\mathbf{k}$ which is close to the momentum maximizing $J_{\mathbf{q}}$. Then, $\mathbf{v}_1(\mathbf{k})$, $\mathbf{v}_2(\mathbf{k})$, and $\mathbf{v}_3(\mathbf{k})$ are the hard, the middle, and the easy axis for magnetization along which we direct x, y, and z axes, respectively. Notice that $\mathcal{D}_{\mathbf{k}}^{\alpha\beta}$ is diagonal in this basis. One obtains from Eqs. (8)–(11) for terms linear in bosonic operators, which arise in Eq. (44) only at $\mathbf{q} = \pm 2\mathbf{k}$

$$\frac{1}{\sqrt{N}}\mathcal{H}_{1d} = i[\lambda_2(\mathbf{k}) - \lambda_3(\mathbf{k})]\left(\frac{S}{2}\right)^{3/2} \times (a_{-2\mathbf{k}} - a_{2\mathbf{k}} + a_{2\mathbf{k}}^\dagger - a_{-2\mathbf{k}}^\dagger). \tag{45}$$

Linear terms (45) have the same form as those arisen in the case of biaxial anisotropy [see Eq. (16)]. Corrections to the ground state energies can be calculated in much the same way as it is done above for the biaxial anisotropy.

As a result, one has to compare the following ground-state energies if the magnetic field is directed along the z axis [cf. Eqs. (30) and (35)]:

$$\frac{1}{N}\mathcal{E}^{yz} = -\frac{S^2 J_{\mathbf{k}}}{2} - \frac{S^2[2\lambda_1(\mathbf{k}) - \lambda_2(\mathbf{k}) - \lambda_3(\mathbf{k})]}{2} - \frac{h^2}{2(2J_{\mathbf{k}} - J_{\mathbf{0}} - J_{2\mathbf{k}})}, \tag{46}$$
$$\frac{1}{N}\mathcal{E}^{xy} = -\frac{S^2 J_{\mathbf{k}}}{2} - \frac{S^2[\lambda_1(\mathbf{k}) - \lambda_2(\mathbf{k})]}{2} - \frac{h^2}{2(J_{\mathbf{k}} - J_{\mathbf{0}})}. \tag{47}$$

The critical field value at which the spiral plane flop takes place reads as [cf. Eq. (37)]

$$h_{flop} = S\sqrt{[\lambda_1(\mathbf{k}) - \lambda_3(\mathbf{k})]\tilde{J}}, \tag{48}$$

where $\tilde{J}$ is given by Eq. (38). If the external magnetic field is along the y axis, the spiral plane flop occurs at

$$h_{flop} = S\sqrt{[\lambda_1(\mathbf{k}) - \lambda_2(\mathbf{k})]\tilde{J}}. \tag{49}$$

Equations (48) and (49) can be related to h_s using Eq. (41) if $J_{\mathbf{0}} \approx J_{2\mathbf{k}}$.

IV. FLOPS AT ARBITRARY FIELD DIRECTION

Let us assume now that the external magnetic field

$$\mathbf{h} = h(\sin t \cos f, \sin t \sin f, \cos t) \tag{50}$$

is directed arbitrary. For definiteness, we consider the system with the biaxial anisotropy (1). An extension to the system with dipolar forces can be made straightforwardly as in Sec. III. Let us characterize the spiral plane by the vector normal to it

$$\mathbf{n}(\theta, \varphi) = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta). \tag{51}$$

It is convenient to introduce two components of the magnetic field: perpendicular to the spiral plane $\mathbf{h}_n$ and the in-plane component $\mathbf{h}_\tau$ whose values read as

$$h_n = h[\sin\theta \sin t \cos(\varphi - f) + \cos\theta\cos t], \tag{52}$$
$$h_\tau = \sqrt{h^2 - h_n^2}. \tag{53}$$

In terms of these quantities, the system energy has the form

$$\frac{\mathcal{E}(\theta,\varphi)}{NS^2} \simeq -\frac{E(\cos^2\varphi + \cos^2\theta\sin^2\varphi) + D\sin^2\theta}{2} - \frac{h_n^2}{2\tilde{J}S^2}, \tag{54}$$

where the angle-independent term $-J_{\mathbf{k}}/2 - h^2/2S^2(2J_{\mathbf{k}} - J_{\mathbf{0}} - J_{2\mathbf{k}})$ is omitted and $\tilde{J}$ is given by Eq. (38).

We analyze now the stability of the spiral planes with respect to small variations in θ and φ using Eq. (54). Let us start with spin rotation in the yz plane (i.e., $\theta = \pi/2$, $\varphi = 0$). In particular, energy (54) is minimal in this case at $h = 0$. Let us discuss the stability of such spin texture at finite magnetic field by considering angle variations of the form

$$\theta = \frac{\pi}{2} - \delta\theta, \quad \varphi = \delta\varphi. \tag{55}$$

The energy variation reads as

$$\begin{aligned}\frac{\delta\mathcal{E}(\theta,\varphi)}{NS^2} = {}& \frac{E(\delta\varphi)^2 + D(\delta\theta)^2}{2} \\ & - \frac{h^2}{\tilde{J}S^2}(\delta\theta\cos t + \delta\varphi\sin t\sin f)\sin t\cos f \\ & - \frac{h^2}{2\tilde{J}S^2}[(\delta\varphi)^2\sin^2 t(\sin^2 f - \cos^2 f) \\ & + 2\delta\theta\delta\varphi\cos t\sin t\sin f \\ & + (\delta\theta)^2(\cos^2 t - \sin^2 t\cos^2 f)].\end{aligned} \tag{56}$$

Notice that there are field-dependent terms in Eq. (56) linear in $\delta\theta$ and $\delta\varphi$. They vanish if the magnetic field lies in the yz plane (i.e., at $f = \pi/2$) and if $\mathbf{h}$ is parallel to the x axis (i.e., at $t = 0$). In other cases, linear terms lead only to a continuous rotation of the spiral plane by the external magnetic field [$\mathbf{n}(\theta,\varphi)$ rotates towards the magnetic field direction]. No spiral plane flops can happen also if the the magnetic field is oriented along the x axis because h-dependent terms in

Eq. (56) read in this case as

$$\frac{h^2}{2\tilde{J}S^2}[(\delta\varphi)^2+(\delta\theta)^2] \tag{57}$$

that results in a stable energy minimum for the spin texture in the yz plane.

If $\mathbf{h}$ lies in the yz plane (i.e., at $f=\pi/2$), we have for h-dependent terms in Eq. (56)

$$-\frac{h^2}{2\tilde{J}S^2}[(\delta\varphi)^2\sin^2 t+2\delta\theta\delta\varphi\cos t\sin t+(\delta\theta)^2\cos^2 t]. \tag{58}$$

The energy minimum at $\theta=\pi/2$ and $\varphi=0$ is stable until $\delta\mathcal{E}(\theta,\varphi)$ remains a positively defined quadratic form, i.e., if the following inequality holds:

$$ED-\frac{h^2}{\tilde{J}S^2}(E\cos^2 t+D\sin^2 t)>0. \tag{59}$$

The field value at which the spiral plane flop takes place can be found from Eq. (59) with the result

$$h_{flop}=S\sqrt{\tilde{J}\frac{ED}{E\cos^2 t+D\sin^2 t}} \tag{60}$$

which is a generalization of Eq. (37) for arbitrary t. The generalization of Eqs. (48) and (49) has the form

$$h_{flop}=S\sqrt{\tilde{J}\frac{[\lambda_1(\mathbf{k})-\lambda_2(\mathbf{k})][\lambda_1(\mathbf{k})-\lambda_3(\mathbf{k})]}{[\lambda_1(\mathbf{k})-\lambda_2(\mathbf{k})]\cos^2 t+[\lambda_1(\mathbf{k})-\lambda_3(\mathbf{k})]\sin^2 t}}. \tag{61}$$

The generalization of Eq. (40) reads as

$$h_{flop}=\sqrt{2Sh_s\frac{ED}{E\cos^2 t+D\sin^2 t}}. \tag{62}$$

Let us discuss now the orientation of the spiral plane after the flop when $\mathbf{h}$ lies in the yz plane. We have carried out an analysis of the stability of the configuration with $\theta=t$ and $\varphi=f$ similar to that performed above. We have found that the anisotropy provides terms in the energy linear in angles variations if the field is not directed along y or z axes. Thus, we make a conclusion that if the external magnetic field is in the yz plane but $\theta\neq 0$ or $\pi/2$, $\mathbf{n}$ is not parallel to $\mathbf{h}$ after the flop and it smoothly rotates towards $\mathbf{h}$ upon further field increasing.

V. POSSIBLE APPLICATIONS

We discuss in this section application of the theory proposed above to particular spiral materials. Co-doped $MnWO_4$ with the dopant concentration 0.05 is thoroughly investigated experimentally in Ref. [16]. $Mn_{0.95}Co_{0.05}WO_4$, in contrast to pure $MnWO_4$ [17], is in a multiferroic cycloidal phase at small T. Application of in-plane magnetic field leads to a spontaneous flop of the spin rotation plane perpendicular to the field at $h=h_{flop}\approx 10$ T $\ll h_s\approx 60$ T [16]. If $\mathbf{h}$ is directed along the hard axis, the spin rotation plane stays intact. This picture is very similar to that we obtain above theoretically. The difference is that for $\mathbf{h}$ directed along the medium axis the flop is replaced by a rather rapid but continuous rotation of the spiral plane in a field interval of about 4 T. The latter may be attributed to local anisotropy of Co ions and requires more careful consideration. Since Mn^{2+} ions are in a spherically symmetric state with $L=0$ and $S=5/2$, it is expected that the anisotropy of the spin-orbit origin is strongly suppressed and the main anisotropic interaction in the system is the dipolar one. We have calculated eigenvalues of the dipolar tensor $\mathcal{D}_{\mathbf{q}}^{\alpha\beta}$ for pure $MnWO_4$ and substituted them to Eqs. (48) and (49) for h_{flop} estimation in $Mn_{0.95}Co_{0.05}WO_4$. Values of $J_{\mathbf{0}}$, $J_{\mathbf{k}}$, and $J_{2\mathbf{k}}$ arisen in Eqs. (48) and (49) have been calculated using exchange coupling constants found from fitting of neutron experimental data in Ref. [18]. For magnetic field along the easy axis, we find $h_{flop}=8$ T while the experimentally observed [16] value is ≈10 T. For magnetic field directed along the medium axis, we obtain $h_{flop}=6.5$ T which lies in the middle of the field interval, where the continuous rotation of the spiral plane is observed experimentally [16]. Notice also that h_{flop} found using Eq. (41) via experimentally obtained h_s is only 20% smaller than that obtained above although $J_{\mathbf{0}}$ is 1.5 times as large as $J_{2\mathbf{k}}$.

$EuNiGe_3$ is a helimagnet with equally possible spiral vectors $\mathbf{k}=(\frac{1}{4},\delta,0)$, $(\frac{1}{4},-\delta,0)$, and $(\delta,\frac{1}{4},0)$ allowed by the tetragonal symmetry, where $\delta=0.05$ [19]. Magnetodipolar interaction is expected to be very important in this material because exchange constants are rather small and Eu^{2+} ions are in a spherically symmetric state with $L=0$ and $S=7/2$ [19]. It can be shown [20] that dipolar forces make the spiral plane be perpendicular to $\mathbf{k}$ in agreement with experimental observations. It is believed that a small Dzyaloshinskii-Moriya interaction is responsible for the finite δ [21]. Magnetic field directed along a and b tetragonal axes results in the spiral plane flop accompanied with changing $\mathbf{k}$ by another equivalent spiral wave vector [19]. Then, the theory presented above should be modified to describe such flops (as it is done in Ref. [22] for a collinear antiferromagnet). However, $\mathbf{k}$ does not change significantly during the flop if $\mathbf{h}$ is parallel to the c axis and our theory can work in this case. Calculations show that $\lambda_a(\mathbf{k})-\lambda_c(\mathbf{k})=0.135$ K in Eq. (48). To estimate $\tilde{J}$ given by Eq. (38) and appearing in Eq. (48), we assume that $J_{2\mathbf{k}}\approx J_{\mathbf{0}}$ in which case $\tilde{J}$ is related to h_s [see discussion after Eq. (41)]. It was found experimentally that the saturation field $h_s\approx 6$ T in $EuNiGe_3$ [19]. As a result, we obtain $h_{flop}=2.05$ T which matches excellently the experimentally observed value ≈2 T [19].

Spiral plane flops have been reported recently also in many others spiral magnets many of which are multiferroics: $LiCu_2O_2$ [23–25], $NaCu_2O_2$ [26], $CuCrO_2$ [27–29], $CuCl_2$ [30], $LiCuVO_4$ [8,31,32], and $KCu_3As_2O_7(OD)_3$ [33] to mention just a few. In all of them the anisotropy of spin-orbit origin is expected to overcome significantly the dipolar forces. On the other hand, values of anisotropy have not been determined yet in these compounds so that we cannot check our theory in these cases.

VI. SUMMARY AND CONCLUSION

To conclude, we present a theory of field-induced flops of plane in which spins rotate in frustrated Heisenberg helimagnets with small anisotropic interactions, biaxial anisotropy, and dipolar forces. We find that flops occur upon the field increasing if the field lies in the spiral plane stabilized at $h = 0$. The spiral plane becomes perpendicular to the field after the flop (see Fig. 1). The critical fields h_{flop} are given by Eqs. (60) and (61) for biaxial anisotropy and dipolar interaction, respectively. In the case of biaxial anisotropy, if $J_{\mathbf{0}} \approx J_{2\mathbf{k}}$, where $\mathbf{k}$ is the helix vector, h_{flop} is expressed via the saturation field h_s [see Eq. (62)] that opens a simple way to determine the anisotropy value if h_{flop} and h_s are known. Notice also that if the field is directed along the easy axis Eq. (62) is identical to that for the spin-flop field in collinear axial magnets. In contrast to the spin flop in collinear magnets, where the flop takes place only at a very narrow interval of the field directions along the easy axis [13], flops of the spiral plane happen at any orientation of the field in the spiral plane.

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