From spiral to ferromagnetic structure in B20 compounds: Role of cubic anisotropy

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The cubic noncentrosymmetric structure of the B20 compounds produces the helical (homochiral) structure with the wave vector $k_s = D/J$ balanced by the competition of two interactions: the large ferromagnetic exchange interaction J and small antisymmetric Dzyaloshinskii-Moriya interaction D. The difference in the energies between the ferromagnetic collinear and helical states can be experimentally measured by the critical magnetic field H_{c2} needed to transform the helix into the field-induced ferromagnet. We show that the cubic anisotropy, first, can limit the stability of the helix phase in the range of small k_s and, second, makes its own contribution to the value of the critical field H_{c2}. We illustrate our findings taking an example of a transformation of the helix structure to the ferromagnet at $x \to x_c$ in the solid solutions $Fe_{1-x}Co_xGe$. We demonstrate that the mechanism of the transformation is realized via the competition between the cubic anisotropy and the Dzyaloshinskii-Moriya interaction.

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I. INTRODUCTION

The cubic B20 compounds have a noncentrosymmetric crystallographic structure described by the $P2_13$ space group. Lack of the symmetry center of the crystal structure produces the chiral spin-spin Dzyaloshinskii-Moriya (DM) interaction [1,2]. The DM interaction results in appearance of the spin helix with the certain chirality γ_m rigorously determined by the structural chirality Γ_c (left or right) [3–8]. However, the relation between two chiralities ($\Gamma_c \leftrightarrow \gamma_m$) is found to be different for various B20 compounds. For Mn-based compounds ($Mn_{1-x}Fe_xSi$ and $Mn_{1-x}Co_xSi$) the crystalline and magnetic chiralities have the same sense ($\Gamma_c \cdot \gamma_m = 1$), while for the Fe based ones ($Fe_{1-x}Co_xSi$) the chiralities Γ_c and γ_m are opposite to each other ($\Gamma_c \cdot \gamma_m = -1$).

The experimental evidence was recently given for the magnetic transition in the $Mn_{1-x}Fe_xGe$ compounds, where the helix chirality can be altered by mixing the two types of magnetic atoms (Fe and Mn) [9,10]. Provided that left-handed crystals are considered, the left-handed helix is observed for the compounds with $x < x_c$, while the right-handed helix appears in the compounds with $x > x_c$. The ferromagnetlike system with the wave vector $k_s \to 0$ is realized at $x \to x_c = 0.75$. Further experiments have shown that another FeGe type of compound ($Fe_{1-x}Co_xGe$) demonstrates the same phenomenon of the flip of spin chirality at the critical concentration $x_c = 0.6$ [11]. It was found in both cases that the change of the chirality undergoes through the ferromagnetic state, characterized by the zero value of the wave vector $k = 0$.

In accord with the model suggested by Bak and Jensen [12] and independently by the group led by Kataoka [13], the major ferromagnetic exchange interaction J together with the DM interaction D stabilize the helical (homochiral) structure in these systems below T_c. These two interactions are balanced in the value of the helix wave vector,

$$k_s = D/J. \tag{1}$$

The anisotropic exchange interaction had been introduced into the model since it can both change slightly the value and fix the direction of the wave vector $\mathbf{k}$ along the principal cubic axes. Kataoka and co-authors also noticed the importance of the cubic anisotropy, which can make the entire helix structure unstable and stabilize the ferromagnetic state instead, if the anisotropy energy is comparable with the DM interaction [13].

Upon application of the magnetic field the helix, first, is transformed to the single-domain conical state with $\mathbf{k} \parallel \mathbf{H}$ at the first critical field H_{c1}. Then, the conical state is transformed to the collinear ferromagnetic state at the second critical field $H_{c2} > H_{c1}$. According to [14,15], the difference in energies between the ferromagnetic collinear and helical states can be experimentally measured by the critical field H_{c2}. This energy difference is equal to

$$g\mu_B H_{c2} \approx Ak_s^2, \tag{2}$$

where $A = J \cdot S \cdot a^2$ is the spin wave stiffness, S is the ordered spin, and a is the lattice constant. Thus, the set experimental parameters k_s, H_{c2}, and S describes completely the magnetic system of such compounds and allows one to estimate values of the ferromagnetic exchange interaction J and the DM interaction D.

In this paper we address the question of the applicability of the above-mentioned model for the $Fe_{1-x}Co_xGe$ compounds close to the critical concentration x_c. The manuscript is organized in the following way. Section II summarizes the experimental finding of Ref. [11]. We show inconsistencies of the values of the ferromagnetic exchange interaction J and the DM interaction D estimated using Eqs. (1) and (2) close to the critical concentration x_c. We discuss the essence of the model [12,13] in Sec. III. We describe the role of the cubic anisotropy in the possible transition from the helical to the ferromagnetic state at small values of the helix wave vector k. Particularly, we show in a simple phenomenological calculation that it is the cubic anisotropy that is responsible for the instability of the helical state and for the transformation of the system to the ferromagnet close to x_c. Section IV presents the concluding remarks.

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II. TRANSITION FROM HELICAL TO FERROMAGNETIC STATE

The $Fe_{1-x}Co_xGe$ compounds with x running from 0.0 to 1.0 had been synthesized under high pressure as described in [16]. Neutron diffraction and magnetization measurements have shown that these compounds are ordered into the spin helix structure below the critical temperature T_c. The x dependence of the critical temperature T_c is shown in Fig. 1(a). T_c decreases monotonically with increase of x approaching 0 at $x \to 0.9$. The value of the ordered spin S per magnetic atom taken from SQUID measurements are also presented in Fig. 1(a). It also decreases with x but shows a "little shoulder" in the range of x between 0.3 and 0.5. Figure 1(b) shows the x dependence of the helix wave vector k_s. In the Fe-rich part of compounds

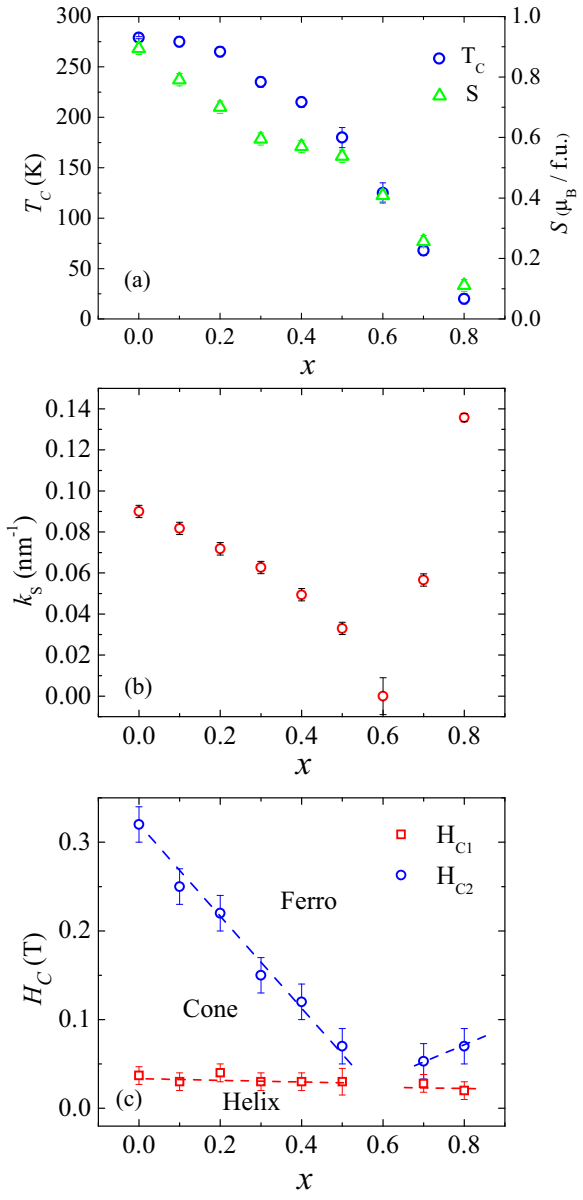

FIG. 1. (Color online) (a) Critical temperature T_C and ordered spin value S. (b) Helix wave vector k_s. (c) Critical magnetic fields H_{c1} and H_{c2}. All experimental parameters are plotted in dependence on the concentration x of $Fe_{1-x}Co_xGe$ compounds.

k_s decreases for $x \in [0 \div 0.5]$, then it falls down to zero at $x \approx 0.6$, and increases again up to the value of $k_s = 0.14\ \text{nm}^{-1}$ for $x = 0.8$. Accounting for the fact that the value of k_s approaches zero at $x_c \approx 0.6$ and the ordering temperature is as high as $T_c = 130$ K, we conclude that this compound is a ferromagnet. This can be only possible if the macroscopic Dzyaloshinskii constant D is effectively vanishing. In any case, the x dependence of the wave vector k_s in the Co-rich part of compounds [Fig. 1(b)] can be now interpreted as a change or chirality at $x \approx 0.6$.

The helix is transformed into the ferromagnet by application of the magnetic field above the critical value H_{c2} [Fig. 1(c)]. The values of the critical field H_{c2} depends strongly on concentration x, first, decreasing from pure FeGe to its minimum (accompanied by $|\mathbf{k}_s| \to 0$) at $x_c \approx 0.6$, and, then increasing again at higher x. Thus, we observe a transformation of the helix structure to the ferromagnet at $x \to x_c$ at zero field. Additionally, the weak anisotropic exchange and/or cubic anisotropy should fix the direction of the magnetic helix along the principal axes of the cubic symmetry. Upon application of the magnetic field the helix wave vector $\mathbf{k}$ rotates toward the field axis at the field H_{c1}, which is the measure for the anisotropic interactions. The field H_{c1} changes little with the concentration x and is roughly equal to 0.03 T.

Figure 1(c) represents the $H - x$ phase diagram of the $Fe_{1-x}Co_xGe$ compounds. The magnetic system is ordered in the plane spin helix at zero field. The structure of the spin helix is changed upon application of a field even smaller than H_{c1}. The helix wave vector is fixed along the easy anisotropic axis but its structure is distorted by the field. This distortion is well recorded in the small-angle neutron diffraction measurements as the high-order harmonics appeared in the scattering picture [17]. This distorted helix is transformed into the cone structure with the wave vector $\mathbf{k}$ along the field axis at H_{c1}. The cone structure is stable in the field range between H_{c1} and H_{c2}. It becomes a ferromagnet with the spins aligned along the field axis above H_{c2}.

In accord with the model [12,13], Eq. (1) gives k_s as a ratio between the isotropic spin exchange interaction J and the DMI constant D. The sign of the wave vector $\mathbf{k}_s$ describes the chirality of the helical structure and is determined by the sign of the Dzyaloshinskii constant sgn(D). The value of k_s is also bound to the critical field H_{c2} via Eq. (2) [14,15]. Thus, using Eqs. (1) and (2) and the experimental data from Figs. 1(a)–1(c), one is, seemingly, able to estimate the major driving interactions $J = A/(\text{Sa}^2)$ and D/a. In this approach, J is determined by the critical field H_{c2} and k_s. We denote the exchange constants obtained in this approach as $J(H_{c2},k)$ and $D(H_{c2},k)$. They are shown in Figs. 2(a) and 2(b) as a function of x (here a is the lattice parameter).

The exchange constant J [quadratic symbols in Fig. 2(a)] changes strongly with x. It shows a divergentlike behavior at $x \to 0.6$ that is certainly related to zero value of k. As it is shown in Fig. 2(b) the constant D is independent on x at $x < 0.6$ and then it changes the sign at $x \to 0.6$.

It is worthy to note that the measured characteristic parameters of the system (H_{c2} and k_s) show tendency to zero at $x = x_c$, what is interpreted as a transition from the spiral to the ferromagnetic phase. At the same time the calculated parameters, $J(H_{c2},k_s)$ and $D(H_{c2},k_s)$, have a little physical

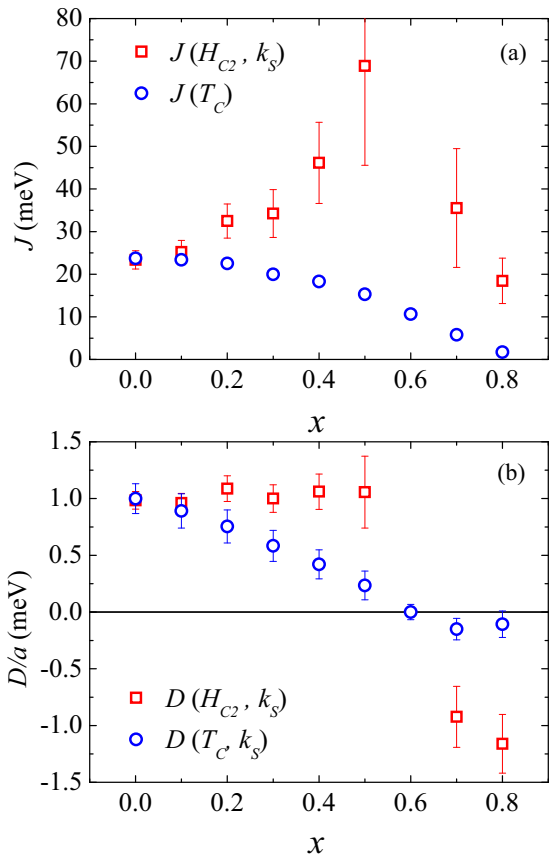

FIG. 2. (Color online) (a) Exchange constant J and (b) DM constant D estimated using the model ([12,13] + [14,15]) and using the model ([12,13] + [10]) in dependence on concentration x of $Fe_{1-x}Co_xGe$ compounds.

meaning: The exchange constant $J(H_{c2},k_s)$ increases drastically at x_c, while the DM constant $D(H_{c2},k_s)$ does not vanish at x_c but shows steplike change only! The very fact of J tending to infinity in the vicinity of x_c, accompanying steplike abrupt change of D shows that the model, characterized by Eqs. (1) and (2), is unapplicable to the mixed $Fe_{1-x}Co_xGe$ compounds.

Another approach to estimate the major energy constants was used in [10]. The magnetic transition temperature k_BT_c is roughly equal to J. Since the wave vector k is proportional to D/J, therefore, the change of J and D with the concentration x can be recalculated. In this approach the value of $J(T_c)$ changes smoothly with x similar to the x dependence observed for the critical temperature T_c. The DM interaction constant $D(T_c,k_s)$ shows a steep change with x in the range of $x \leqslant 0.5$. It crosses the zero line, i.e., changing the sign around $x = 0.6$ and keeps a small negative value above x_c. This behavior of the effective constants of $J(T_c)$ and $D(T_c,k_s)$ is consistent to the hypothesis on the averaged exchange constants for Fe and Co atoms in the mixed compounds.

It is interesting to note that these two approaches give the same values of J and D for the MnSi compound [17]. We note also that they work fairly well for the small values of x (Fig. 2). This range of x differs from the whole range only by the condition that the critical field H_{c2}, which is related to the helix energy, is much larger than the critical field H_{c1}, which is related to the anisotropy [Fig. 1(c)]. Thus one can conclude that the anisotropy can at least interfere with DM interaction upon formation of the spin helix.

It is shown in the following section (Sec. III) that Eq. (2) must be corrected in order to make the first approach working. The cubic anisotropy competes to the weakened DM interaction. Moreover, it is able to suppress the formation of the helix structure. This competition also introduces a new term in the relation between H_{c2} and k_s [Eq. (2)], which can be rewritten in the form given by Eqs. (27) and (28).

III. SPIN HELIX AND CUBIC ANISOTROPY

A. Bak-Jensen model

Let us outline the results of the previous studies obtained for the helical state in ferromagnets distorted by the DM interaction [12,14]. The corresponding Hamiltonian is a sum of the exchange energy H_{EX}, energy of the DM interaction H_{DM}, anisotropic exchange energy H_{AE}, and Zeeman energy H_Z. They can be written as

$$\begin{aligned} H_{EX} &= -\frac{1}{2}\sum J_{\mathbf{R}-\mathbf{R}'}\mathbf{S}_{\mathbf{R}}\cdot\mathbf{S}_{\mathbf{R}'};\\ H_{DM} &= \frac{1}{2}\sum D_{\mathbf{R}-\mathbf{R}'}(\nabla-\nabla')[\mathbf{S}_{\mathbf{R}}\times\mathbf{S}_{\mathbf{R}'}];\\ H_{AE} &= \frac{1}{2}\sum F_{\mathbf{R}-\mathbf{R}'}\{(\nabla_x S^x_{\mathbf{R}})(\nabla'_x S^x_{\mathbf{R}'}) + (\nabla_y S^y_{\mathbf{R}})(\nabla'_y S^y_{\mathbf{R}'})\\ &\quad + (\nabla_z S^z_{\mathbf{R}})(\nabla'_z S^z_{\mathbf{R}'})\};\\ H_Z &= -\mathbf{H}\sum \mathbf{S}_{\mathbf{R}}, \end{aligned} \tag{3}$$

where $\mathbf{H} = g\mu_B\mathbf{B}_{in}$ and $\mathbf{B}_{in}$ is an internal magnetic field [14]. In general form the spin helix can be represented as

$$\mathbf{S}_{\mathbf{R}} = S[\hat{c}\sin\alpha + (\mathbf{A}e^{i\mathbf{k}\cdot\mathbf{R}} + \mathbf{A}^*e^{-i\mathbf{k}\cdot\mathbf{R}})\cos\alpha], \tag{4}$$

where $\mathbf{k}$ is the helix wave vector and α is the angle between the spins and the spin rotation plane. Vector $\mathbf{A}$ is determined as $\mathbf{A} = (\hat{a} - i\hat{b})/2$. Unit vectors $\hat{a},\hat{b},\hat{c}$ form the right-handed orthogonal frame and $[\hat{a}\times\hat{b}] = \hat{c}$. Thus, one has $(\mathbf{A}\cdot\mathbf{A}) = 0, (\mathbf{A}\cdot\mathbf{A}^*) = 1/2, [\mathbf{A}\times\mathbf{A}^*] = i\hat{c}/2$.

We use spherical coordinates with angles ϕ,θ bound to the spin helix. The basic vectors $\hat{a},\hat{b}$, and $\hat{c}$ can be given as

$$\begin{aligned} \hat{a} &= (\cos\phi, \sin\phi, 0);\\ \hat{b} &= (-\sin\phi\cos\theta, \cos\phi\cos\theta, -\sin\theta);\\ \hat{c} &= (-\sin\phi\sin\theta, \cos\phi\sin\theta, \cos\theta). \end{aligned} \tag{5}$$

Inserting Eq. (4) in Eq. (3) we obtain in k^2 approximation an expression for the classical energy of the helical state,

$$\begin{aligned} E_{BJ} = -\frac{S^2J_0}{2} + \Bigg[\frac{Ak^2}{S} + \frac{F_0 I(\mathbf{k})}{2}\\ - 2D_0(\mathbf{k}\cdot\hat{c})\Bigg]\frac{S^2\cos^2\alpha}{2} - SH_{\parallel}\sin\alpha, \end{aligned} \tag{6}$$

where $A = S(J_0 - J_{\mathbf{k}})/k^2$ is the spin-wave stiffness [14], the $\mathbf{k}$-dependent functions are determined as $J_{\mathbf{k}} = \sum J_{\mathbf{R}}\exp(\mathbf{k}\cdot\mathbf{R})$, $I(\mathbf{k}) = \sum k_j^2(\hat{a}_j^2 + \hat{b}_j^2)$ is a cubic invariant, and $H_{\parallel} = (\mathbf{H}\cdot\hat{c})$. The subscript notation in E_{BJ} stands to refer to the Bak-Jensen (BJ) model [12].

The energy $E_{\rm BJ}$ is minimal with respect to the value of $\mathbf{k}$ if

$$\begin{aligned} Ak_j + SF_0k_j\left(\hat{a}_j^2+\hat{b}_j^2\right)/2 = SD_0\hat{c}_j;\\ A\mathbf{k}^2 + SF_0I(\mathbf{k})/2 = SD_0(\mathbf{k}\cdot\hat{c}),\end{aligned} \tag{7}$$

here the first line demonstrates a weak dependence of the orientation of $\mathbf{k}$ on the crystal structure. Neglecting it we obtain a helix wave vector:

$$\mathbf{k}_s = SD_0\hat{c}/A. \tag{8}$$

Substituting $D_0 = Ak_s/S$ into Eq. (6) we have as a result the final expression for $E_{\rm BJ}$:

$$E_{\rm BJ} = -\frac{S^2J_0}{2} - \frac{SAk_s^2\cos^2\alpha}{2}\left[1-\frac{SF_0I(\hat{c})}{2A}\right] - SH_\parallel\sin\alpha. \tag{9}$$

The second term corresponds to DM interaction and presents the energy gain of the helical state as compared with the ferromagnetic one. The cubic invariant $I(\hat{c})$ has minimum $I = 0$ and maximum $I = 2/3$ at $\hat{c} = (1,0,0)$ and $\hat{c} = (1,1,1)/\sqrt{3}$, respectively. As a consequence, at small magnetic fields $H \ll |F_0|k_s^2$ the vector $\mathbf{k}_s$ points along the cubic edge or diagonal for $F_0 > 0$ and $F_0 < 0$, respectively [12,14]. In the opposite limiting case $H \gg |F_0|k_s^2 \sim H_{c1}$ one finds the helix wave vector directed along the field axis $\mathbf{k}_s \parallel \mathbf{H}$. Neglecting in Eq. (9) the F_0 term connected to the exchange anisotropy and minimizing the energy with respect to the angle α, we obtain

$$\sin\alpha = H/Ak_s^2. \tag{10}$$

Equation (10) describes the effect of the field H on the spin helix. When the field $H = 0$, then $\alpha = 0$ implying existence of the plane helix. When the field is applied $H \neq 0$, then $\alpha \neq 0$ and the cone phase appears. At the critical field H_{c2}, when $\alpha = \pi/2$ and $H_{c2} = Ak_s^2$ the cone transforms to the ferromagnet. Finally, when $H > H_{c2}$ we have the ferromagnetic state [14].

B. Cubic anisotropy

As the anisotropic exchange is small in comparison with the isotropic one, we neglect the F_0 term in Eq. (9). Another anisotropic term—the cubic anisotropy—should be considered.

The cubic anisotropy can be represented as

$$H_{\rm CA} = K\sum\left\{\left(S_{\mathbf{R}}^x\right)^4 + \left(S_{\mathbf{R}}^y\right)^4 + \left(S_{\mathbf{R}}^z\right)^4\right\}. \tag{11}$$

In ferromagnets $\mathbf{S_R} = \mathbf{S}$ and a contribution of the cubic anisotropy to the classical energy can be written,

$$E_{\rm CA} = G\begin{cases}1;\mathbf{S}\parallel(1,0,0),\\ 1/3;\mathbf{S}\parallel(1,1,1).\end{cases} \tag{12}$$

Here $G = KS^4$ and the extrema of $E_{\rm CA}$ are shown in the right-handed side of Eq. (12). So directions (1,1,1) and (1,0,0) are the easy axes for $K > 0$ and $K < 0$, respectively.

Inserting Eq. (4) into Eq. (11) and adding to Eq. (9) we get the expression for the helix energy in the magnetic field, which takes into account the cubic anisotropy:

$$\begin{aligned} E = -\left(SAk_s^2/2\right)\cos^2\alpha + G[C\sin^4\alpha + (3/8)B\cos^4\alpha\\ + 3I\sin^2\alpha\cos^2\alpha] - SH_\parallel\sin\alpha.\end{aligned} \tag{13}$$

The cubic invariants C, B, I are given by

$$C = \sum\hat{c}_j^4;\quad B = \sum\left(\hat{a}_j^2+\hat{b}_j^2\right)^2;\quad I = \sum\hat{c}_j^2\left(\hat{a}_j^2+\hat{b}_j^2\right). \tag{14}$$

They depends on angles ϕ,θ determining $\hat{c}$ direction and considered in the Appendix.

Taking into account the relations given by Eq. (A6) we rewrite Eq. (13) in the form which is more convenient for further analysis:

$$\begin{aligned} E = -\frac{SAk_s^2}{2}(1-\sin^2\alpha) + \frac{G}{8}[(3-5C)(-7\sin^4\alpha\\ + 6\sin^2\alpha + 1) + 8C] - SH_\parallel\sin\alpha.\end{aligned} \tag{15}$$

Here the first term is the energy of the spiral with wave vector k_s which appears as a result of the competition between ferromagnetic exchange constant J and DM interaction D. The second term represents the anisotropic energy of the spiral and depends on the angles θ and ϕ of orientation of vector $\hat{c}$. The energy of the cubic anisotropy in this form is a function of one cubic invariant C only and of powers of sine of conical angle α.

The minimum of this energy determines the spin structure and magnetic field behavior of the B20 helimagnets in this classical approximation.

C. Stability of spiral in zero field

In zero field the energy Eq. (15) is minimal for the planar helix ($\alpha = 0$) and $\hat{c} \parallel (1,1,1)$ in case of $G > 0$. Using Eq. (A3) we obtain for the helix energy $E = -SAk^2/2 + G/2$. The helix exists if this energy is smaller than the anisotropic energy $G/3$ of the ferromagnetic state. The corresponding condition is given, by

$$SAk^2 > G/3. \tag{16}$$

Otherwise the helix is unstable and the system is stabilized in the ferromagnetic state.

In the case of $G < 0$ the spiral points along the (1,0,0) axis and we have the following condition for the stability of the helical state in zero magnetic field:

$$SAk^2 > |G|/2. \tag{17}$$

D. Magnetic field behavior

Let us consider the case of the positive anisotropy $G > 0$. Suppose the field is arbitrary oriented in the range of ϕ from 0 to $\pi/4$ and θ from 0 to $\pi/2$, then we have

$$\begin{aligned} E = \frac{SAk_s^2}{2}\sin^2\alpha + \frac{G}{16}W(-7\sin^4\alpha + 6\sin^2\alpha + 1)\\ - SH\cos(\theta-\theta_H)\cos(\phi-\phi_H)\sin\alpha - \frac{SAk_s^2}{2} + GC,\end{aligned} \tag{18}$$

where θ_H and ϕ_H denote the angles of the magnetic field direction. We put cubic invariant $W(\theta,\phi) \equiv 10C - 6$ [Eq. (A7)]. The planar spiral ($\alpha = 0$) aligned along the (1,1,1) axis, i.e., $(\theta,\phi) = (\pi/2 - \arctan(1/\sqrt{2}),\pi/4)$, will turn toward the (θ_H,ϕ_H) with increasing the field. This rotation

of the helix axis is followed by the increase of the conical angle α. The minimum of the energy E determines the helix configuration. The corresponding condition is given by

$$\partial E/\partial\alpha = 0, \quad \partial E/\partial(\theta,\phi) = 0. \tag{19}$$

To estimate the critical field H_{c1} (of orientation of the helix along the field direction) consider the limiting case of the magnetic field along the hard axis $(0,0,1)$. Thus $\theta_H = 0, \phi_H = \pi/4$ in Eq. (18) and θ runs from $\pi/2 - \arctan(1/\sqrt{2})$ (diagonal of the cube) to 0 with constant $\phi = \pi/4$. Solving the system Eq. (19) with respect to α and substituting $\theta = 0$ one can obtain the conical angle at the field H_{c1}:

$$\sin^2\alpha_{c1} = \frac{4}{7}\left(1 + r - \sqrt{r^2 + 2r + \frac{9}{16}}\right), \quad r > \sqrt{\frac{1}{30}}, \tag{20}$$

where $r = SAk_s^2/12G$ is a ratio of the DM interaction and the cubic anisotropy. Using Eqs. (19) and (20) we have the critical magnetic field H_{c1} which depends on the cubic anisotropy G and on the ratio r:

$$H_{c1} = \frac{G}{S}\sin\alpha_{c1}(7\sin^2\alpha_{c1} + 12r - 3), \quad r > \sqrt{\frac{1}{30}}. \tag{21}$$

Obviously, critical magnetic field H_{c1} so much less how much the orientation of the magnetic field is closer to the easy axis.

To obtain the critical field of the phase transition H_{c2} we can consider the equation,

$$SH = \frac{7}{4}GW(\theta_H,\phi_H)\sin^3\alpha + \left(SAk_s^2 - \frac{3}{4}GW(\theta_H,\phi_H)\right)\sin\alpha, \tag{22}$$

where (θ,ϕ) are already equal to (θ_H,ϕ_H). Equation (22) describes the helix behavior in the field above H_{c1}. The value of the magnetic field H_{c2} of the ferromagnetic transition depends on the orientation of the field. As one can see the cubic invariant W is negative for those (θ,ϕ) which are close to the cube diagonal and is positive for those (θ,ϕ) close to the cube edge.

If $W(\theta_H,\phi_H) \geqslant 0$, then Eq. (22) has a solution for $\sin\alpha = 1$ at

$$H_{c2} = Ak_s^2 + W(\theta_H,\phi_H)\frac{G}{S}. \tag{23}$$

If $W(\theta_H,\phi_H) < 0$, then the result depends on the ratio of the DMI and the cubic anisotropy. In the case of $SAk_s^2/G \geqslant 18|W|/4$, Eq. (22) has a solution for $\sin\alpha = 1$ at

$$H_{c2} = Ak_s^2 - |W(\theta_H,\phi_H)|\frac{G}{S}. \tag{24}$$

In the opposite case of $SAk_s^2/G < 18|W|/4$, we have the first-order phase transition to the ferromagnetic state at the critical field,

$$H_{c2} = \sin\alpha_{c2}\left(\frac{2}{3}Ak_s^2 + \frac{1}{2}|W(\theta_H,\phi_H)|\frac{G}{S}\right), \tag{25}$$

and the corresponding critical cone angle,

$$\sin\alpha_{c2} = \sqrt{\frac{4}{21|W|}\frac{SAk_s^2}{G} + \frac{1}{7}}. \tag{26}$$

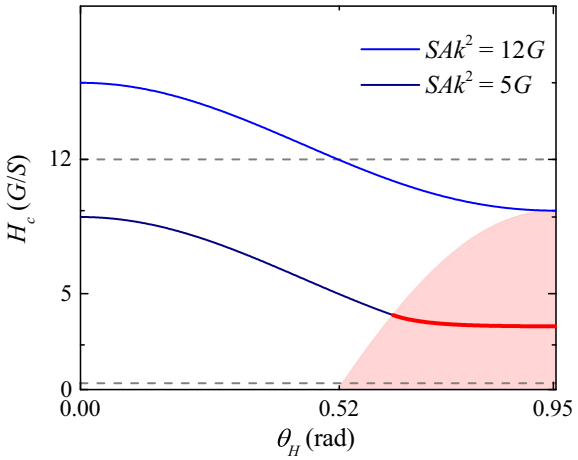

FIG. 3. (Color online) Dependence of the critical magnetic field H_{c2} (in units G/S) on the orientation of the magnetic field within the $\phi = \pi/4$ plane changing from $(0,0,1)$ to $(1,1,1)$ directions for two variants of the ratio of the DM interaction and the cubic anisotropy. The top dashed line corresponds to the ratio SAk_s^2/G, which does not imply any first-order phase transition in any field direction. The bottom dashed line corresponds to the minimal stable value of the SAk_s^2/G equals $1/3$. The red area shows the condition of the first-order phase transition, which depends on the angle θ and the ratio SAk_s^2/G.

Figure 3 shows the dependence of the critical field H_{c2} on the field direction in a particular case of the field oriented between the hard $(0,0,1)$ axis and the easy $(1,1,1)$ axis. Equations (21)–(23) are plotted, for instance, for the case $Ak_s^2 = 12G/S$ and $Ak_s^2 = 5G/S$. The ratio SAk_s^2/G determines the position of the curve along the H_{c2} axis and the cubic invariant W [Eq. (A8)] determines its form. For $Ak_s^2 < 12G/S$ the curve has a part corresponding to the first-order phase transition to the ferromagnet described by Eq. (25). The critical field H_{c2} and critical cone angle [Eq. (26)] are also plotted in Fig. 4 as functions of the ratio r in the case of the field oriented along the $(1,1,1)$ axis.

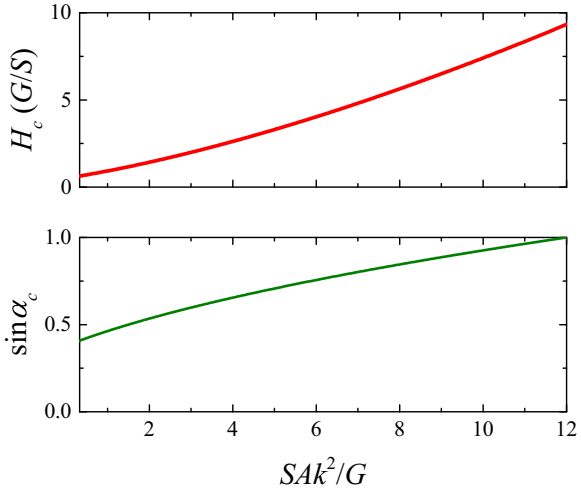

FIG. 4. (Color online) Dependence of the sine of the critical cone angle α_{c2} and the critical magnetic field [along the $(1,1,1)$ direction] of the first-order phase transition H_{c2} on the ratio SAk_s^2/G.

For polycrystalline samples we have to average the action of the magnetic field applied to all directions. So $W(\theta,\phi)$ [Eq. (A7)] should be averaged on θ,ϕ and thus we obtain

$$H_{c2} = Ak_s^2 + \frac{9}{16}\frac{G}{S}, \quad G > 0. \tag{27}$$

If $G < 0$, the first critical field has the same form as Eq. (21) but for $r = SAk_s^2/8|G|$. Since Eq. (22) includes G only in terms $G \cdot W$, it is easy to see that in the case of negative anisotropy $G < 0$ replacement $G \to -|G|$ is equivalent to replacement $W \to -|W|$. Obviously, after averaging of W we will get for the second critical field:

$$H_{c2} = Ak_s^2 - \frac{9}{16}\frac{|G|}{S}, \quad G < 0. \tag{28}$$

IV. CONCLUDING REMARKS

The phenomenological model [12] resulting in the helix spin structure is built on the hierarchy of interactions: ferromagnetic exchange interaction, antisymmetric Dzyaloshinskii-Moriya interaction, and the anisotropic exchange interaction. The cubic anisotropy was not initially included in this model, nevertheless, as it was noticed in [13] and is shown in Sec. III, it plays an important role in cases when the value of the helix wave vector k_s becomes relatively small.

Particularly, the model [12] does not impose any limitations on the value k_s that can be infinitively small. Similar to it, there is no limitation on the value of the critical field H_{c2}, which can go infinitively small as well. Note that the anisotropic exchange interaction, being part of the exchange interaction, cannot impose any limitation on these values of k_s and H_{c2}. On the contrary, the cubic anisotropy, first, leads to the conditions limiting the stability of the helix phase in the range of small k_s [Eqs. (16) and (17)] and, second, makes its own contribution to the value of the critical field H_{c2} [Eqs. (27) and (28)].

Moreover, it was shown that the critical field H_{c2} depends on the orientation of the magnetic field relative to the crystallographic axes, due to the contribution of the cubic anisotropy in Eqs. (27) and (28). The critical field increases (decreases) in the case when the energy of cubic anisotropy is positive (negative). It is also shown that, when DM interaction is weakened, the first-order phase transition occurs from the conical to ferromagnetic phase for certain orientations of magnetic field. The critical cone angle for these cases are given by Eq. (26).

As an illustration of the role of the cubic anisotropy in the magnetic system of the B20 compounds, we take an example of a transformation of the helix structure to the ferromagnet at $x \to x_c$ in the solid solutions $Fe_{1-x}Co_xGe$. This transformation is a result of the different signs of the Dzyaloshinskii-Moriya interaction and the different helix chiralities related to the different magnetic atoms (Fe and Mn). In accord with the hypothesis on the averaged exchange constants in the mixed compounds, the DM constant is effectively vanishing at x_c, while the isotropic exchange is positive and finite. We have shown that the mechanism of the transformation is realized via the competition between the Dzyaloshinskii-Moriya interaction and the cubic anisotropy. In accord with the Eqs. (16) and (17) there should be a range of the concentrations close to x_c, where the $Fe_{1-x}Co_xGe$ compounds should be ferromagnetic. The competition is well noticeable in the helical phase as soon as the second critical field H_{c2} becomes comparable to the first critical field H_{c1}.

In general, we have proven the possibility for existence of the ferromagnetic phase in the noncentrosymmetric cubic magnets with Dzyaloshinskii-Moriya interaction.

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APPENDIX: CUBIC INVARIANTS

For completeness we give explicit expressions for the cubic invariants. Thus we have for the cubic anisotropy,

$$E_{CA} = G[(\cos^4\phi + \sin^4\phi)\sin^4\theta + \cos^4\theta], \tag{A1}$$

and Eq. (12) is a result of the standard calculations.

Inserting the expressions Eq. (5) in $I(\hat{c})$ of Eq. (9), we obtain

$$I(\hat{c}) = [2\sin^2\phi\cos^2\phi + (1 + \sin^4\phi + \cos^4\phi)\cos^2\theta]\sin^2\theta. \tag{A2}$$

Extrema of this function are obtained from this expression and given below Eq. (9).

We have two invariants in Eq. (14). The first one is given by

$$C = \sum c_j^4 = \begin{cases} 1; \hat{c} \parallel (0,0,1) \\ 1/3; \hat{c} \parallel (1,1,1). \end{cases} \tag{A3}$$

Here the extreme values of C are given in the right-handed side.

The second invariant is determined as

$$B = \sum \left(a_j^2 + b_j^2\right)^2 = (\cos^2\phi + \sin^2\phi\cos^2\theta)^2 + (\sin^2\phi + \cos^2\phi\cos^2\theta)^2 + \sin^4\theta, \tag{A4}$$

and we get for the extrema,

$$B = \begin{cases} 2; \hat{c} \parallel (0,0,1) \\ 4/3; \hat{c} \parallel (1,1,1). \end{cases} \tag{A5}$$

There are simple relations between cubic invariants:

$$C - B = 1, \quad I + C = 1, \quad I + B = 2. \tag{A6}$$

The extrema of cubic invariant $W \equiv 10C - 6$:

$$W = \begin{cases} 4; \hat{c} \parallel (0,0,1) \\ -8/3; \hat{c} \parallel (1,1,1). \end{cases} \tag{A7}$$

At $\phi = \pi/4$ W has the simple form:

$$W(\theta, \phi = \pi/4) = 15\cos^4\theta - 10\cos^2\theta - 1. \tag{A8}$$

[1] I. E. Dzyaloshinskii, Zh. Eksp. Teor. Fiz. **46**, 1420 (1964) [Sov. Phys. JETP **19**, 960 (1964)].
[2] T. Moriya, Phys. Rev. **120**, 91 (1960).
[3] M. Tanaka, H. Takayoshi, M. Ishida, and Ya. Endoh, J. Phys. Soc. J. **54**, 2970 (1985).
[4] M. Ishida, Y. Endoh, S. Mitsuda, Y. Ishikawa, and M. Tanaka, J. Phys. Soc. J. **54**, 2975 (1985).
[5] S. V. Grigoriev, D. Chernyshov, V. A. Dyadkin, V. Dmitriev, S. V. Maleyev, E. V. Moskvin, D. Menzel, J. Schoenes, and H. Eckerlebe, Phys. Rev. Lett. **102**, 037204 (2009).
[6] S. V. Grigoriev, D. Chernyshov, V. A. Dyadkin, V. Dmitriev, E. V. Moskvin, D. Lamago, T. Wolf, D. Menzel, J. Schoenes, S. V. Maleyev, and H. Eckerlebe, Phys. Rev. B **81**, 012408 (2010).
[7] V. A. Dyadkin, S. V. Grigoriev, D. Menzel, D. Chernyshov, V. Dmitriev, J. Schoenes, S. V. Maleyev, E. V. Moskvin, and H. Eckerlebe, Phys. Rev. B **84**, 014435 (2011).
[8] D. Morikawa, K. Shibata, N. Kanazawa, X. Z. Yu, and Y. Tokura, Phys. Rev. B **88**, 024408 (2013).
[9] S. V. Grigoriev, N. M. Potapova, S.-A. Siegfried, V. A. Dyadkin, E. V. Moskvin, V. Dmitriev, D. Menzel, C. D. Dewhurst, D. Chernyshov, R. A. Sadykov, L. N. Fomicheva, and A. V. Tsvyashchenko, Phys. Rev. Lett. **110**, 207201 (2013).
[10] K. Shibata, X. Z. Yu, T. Hara, D. Morikawa, N. Kanazawa, K. Kimoto, S. Ishiwata, Y. Matsui, and Y. Tokura, Nature Nanotechnology **8**, 723 (2013).
[11] S. V. Grigoriev, S.-A. Siegfried, E. V. Altynbayev, N. M. Potapova, V. Dyadkin, E. V. Moskvin, D. Menzel, A. Heinemann, S. N. Axenov, L. N. Fomicheva, and A. V. Tsvyashchenko, Phys. Rev. B **90**, 174414 (2014).
[12] P. Bak and M. H. Jensen, J. Phys. C **13**, L881 (1980).
[13] O. Nakanishi, A. Yanase, A. Hasegawa, and M. Kataoka, Solid State Commun. **35**, 995 (1980).
[14] S. V. Maleyev, Phys. Rev. B **73**, 174402 (2006).
[15] A. N. Bogdanov, U. K. Roessler, and C. Pfleiderer, Physica B **359-361**, 1162 (2005).
[16] A. Tsvyashchenko, J. Less Common Metals **99**, L9 (1984).
[17] S. V. Grigoriev, S. V. Maleyev, A. I. Okorokov, Y. O. Chetverikov, P. Böni, R. Georgii, D. Lamago, H. Eckerlebe, and K. Pranzas, Phys. Rev. B **74**, 214414 (2006).