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Ground State of a Two-Sublattice Anisotropic Ferromagnet in a Magnetic Field

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Abstract—The ground state of a classical two-sublattice ferromagnet with the noncollinear single-ion anisotropy axes of the sublattices and the antisymmetric and anisotropic symmetric exchanges between them has been investigated in a magnetic field applied along the hard magnetization directions in the crystal. The threshold relations for the parameters of the anisotropic interactions have been obtained, which determine the choice of the ground state among the three possible magnetic phases. Depending on the ground state type and the field direction, the transition between the phases is a first- or second-order phase transition. The antisymmetric exchange value above which the reorientation between the noncollinear phases ends with a second-order transition depends on the angle between the local easy axes and the single-ion anisotropy value. Field dependences of the magnetization and susceptibility for different ground states have been calculated. A comparison with the results of the magnetic measurements in the highly anisotropic $PbMnBO_4$ ferromagnet has been made.

Keywords: ferromagnetism, single-ion anisotropy, antisymmetric exchange, orientational phase transition

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1. INTRODUCTION

Noncollinearity of the moments of magnetic sublattices is a wide-spread phenomenon in magnetically ordered crystals. It is caused by the fact that magnets are characterized, along with the isotropic Heisenberg exchange, by the anisotropic interactions tending to differently orient spins. The resulting competition between the interactions leads to a compromise noncollinear orientation of the moments. In this case, even a relatively slight deviation from the parallel or antiparallel orientation of the moments not only significant changes the magnetic properties of the crystal (a classic example is weak ferromagnetism), but also induces fundamentally new magnetoelectric, magnetoelastic, magnetocaloric, and other multiferroic properties [1–5]. The bright manifestation of the magnetoelectric properties should be expected in magnetodielectrics with a large total magnetic moment at the abrupt change of the magnetization direction in a magnetic field (the spin-flop transition). In noncollinear antiferromagnets, the value of the weak ferromagnetic moment is determined by the ratio between the anisotropic interactions and the dominant isotropic exchange; as a rule, this value is low. The situation is different in a ferromagnet with the noncollinear magnetic sublattices, i.e., in a weak antiferromagnet [6–8]. In this case, the spin-flop transition along the weak antiferromagnetism vector leads to the stepwise variation in the direction of the total ferromagnetic moment, which is comparable to the ion saturation moment. In this case, the field inducing the reorientation is determined by the value of the anisotropic interactions, which is much weaker than the reorientation field of a weak ferromagnet.

A first-order orientational transition was observed in the highly anisotropic ferromagnet $PbMnBO_4$ in a magnetic field applied along the orthorhombic **b** axis of the crystal [9]. The recent intense studies of its properties [10–14] have been stimulated not only by the rare type of ferromagnetic ordering in dielectrics in general, but also by the high magnetocaloric effect in this crystal [15]. In addition, the existence of isostructural crystals with different magnetic and nonmagnetic ions and different types of magnetic ordering provides a unique opportunity for controlling the magnetic and multiferroic properties by replacing both magnetic Mn^{3+} and nonmagnetic Pb^{2+} ions [15–18]. It was shown [19] that the completion of the magnetization reorientation in a two-sublattice ferromagnet by a first-order phase transition is explained by the noncollinearity of the single-ion anisotropy (SIA) easy axes of the sublattices. This mechanism of noncollinearity of the magnetic sublattices was proposed first by Bozorth to explain weak ferromagnetism in orthoferrites [20] and used by Moriya to describe the magnetic properties of the NiF_2 antiferromagnet [21]. The conditions for the occurrence of the SIA-induced noncollinearity were studied by Bertaut in the symmetry analysis of orthorhombic perovskites [22]. In real crystals, the orientation of local moments is governed, as a rule, by several anisotropic mechanisms. The description of the magnetic properties most frequently takes into account the SIA, antisymmetric exchange (the Dzyaloshinskii–Moriya (DM) interaction) [23, 24], and anisotropic symmetric exchange. Among the last two mechanisms, the DM interaction is usually preferred, since the absolute value of the constant of this exchange in the Hamiltonian is proportional to the first degree of the deviation of the g factor from the purely spin value: $|D| \propto (\Delta g/g)J$ [24]. At the same time, the anisotropic symmetric exchange constant A is proportional to the second power of this deviation $|A| \propto (\Delta g/g)^2 J$, which is less important for $3d$ ions. However, D^2/J and A contribute to the energy of the ground state, which is comparable in value. As a result, the joint account for these interactions can even restore the isotropic symmetry [25–28]. Thus, all the three main anisotropy mechanisms yield the first corrections to the energy of the ground state in the second order of the theory of perturbation in the spin-orbit interaction. Ignoring any interaction without additional examination can lead to the quantitatively incorrect determination of the remaining ones in the interpretation of experimental data.

The aim of this study is to accurately analytically calculate the ground state of the model of a classical two-sublattice ferromagnet with the noncollinear SIA axes of the sublattices and the anisotropic antisymmetric and symmetric exchanges between them. The threshold relations between the parameters of the anisotropic interactions are obtained, which determine three possible magnetic phases in zero magnetic field. The change in each ground state in a magnetic field applied along the hard magnetization axes of the crystal is investigated and the field dependences of the energy, magnetization, and susceptibility are calculated. A change in the type of an orientational phase transition is considered depending on the relation between the SIA parameters and the DM exchange.

2. MODEL

Let us consider a model of a unisite magnet, i.e., a crystal in which the lattice sites with magnetic ions transform into each other during the symmetry transformations. As such symmetry elements, we take the mirror plane and the twofold axis located between ions. The choice of symmetry determines the distribution of the anisotropic interactions and is important for solving the problem accurately. The Hamiltonian of the model has the form

$$\begin{aligned} H &= J\sum_{ij}\mathbf{S}_i\mathbf{S}_j + \mathbf{D}\sum_{ij}[\mathbf{S}_i \times \mathbf{S}_j] + A\sum_{ij}S_i^{\alpha}S_j^{\alpha} \\ &+ K_1\left(\sum_i S_i^{z1^2} + \sum_j S_j^{z2^2}\right) + K_2\left(\sum_i S_i^{y1^2} + \sum_j S_j^{y2^2}\right) \\ &+ g\mu_B\mathbf{H}_0\left(\sum_i \mathbf{S}_i + \sum_j \mathbf{S}_j\right). \end{aligned} \quad (1)$$

The ferromagnetic exchange $J < 0$ relates spins of different sublattices $i \in 1, j \in 2$. The isotropic ferromagnetic exchange between spins inside the sublattices, which, at their identical orientation, leads to a minor constant addition to the energy, is omitted. The twofold symmetry axis is chosen as a crystal **a** axis (2_a); the symmetry plane coincides with the ac ($\mathbf{m}_b$) plane. The noncollinear easy SIA axes ($K_1 < 0$) $z1$ and $z2$ lie in the ab plane. The second axes of the twofold SIA (K_2) $y1$ and $y2$ are parallel to the **c** axis. The symmetry of the problem determines also the directions of the axes of two-ion anisotropic interactions: the axis α of the symmetric anisotropic exchange and the Dzyaloshinskii vector **D** are also parallel to the **c** axis [4, 5, 22, 29, 30].

The ground state of the model is found by minimizing the energy

$$E = \frac{N}{2}z|J|S^2 \cdot \epsilon(\theta_1, \theta_2, \varphi_1, \varphi_2)$$

along the angles of orientation of the sublattice moments. Here, z is the number of magnetic neighbors and $\epsilon(\theta_1, \theta_2, \varphi_1, \varphi_2)$ is the normalized energy of the classical unit sublattice moments $\mathbf{m}_{1,2} = -\mathbf{S}_{1,2}/S$. In the absence of an external magnetic field, depending on signs and values of the anisotropic interactions, three ground states can exist with the orientation of the total ferromagnetic moment $\mathbf{M} = \mathbf{m}_1 + \mathbf{m}_2$ along each crystal axis with different magnetic symmetries: phase A ($\mathbf{M} \parallel \mathbf{a}$) with symmetry $2_a m_b'$, phase B ($\mathbf{M} \parallel \mathbf{b}$) with symmetry $2_a' m_b$, and phase C ($\mathbf{M} \parallel \mathbf{c}$) with symmetry $2_a' m_b'$. The symmetry constraints reduce the determination of the energy of noncollinear phases A and B with the moments lying in the easy axes plane ab to minimization in only one variable and the energy of collinear phase C is found trivially

$$\epsilon_0^A = a - \sqrt{(1 - a\cos\delta_K)^2 + (d - a\sin\delta_K)^2}, \quad (2)$$

$$\epsilon_0^B = a - \sqrt{(1 + a\cos\delta_K)^2 + (d + a\sin\delta_K)^2}, \quad (3)$$

$$\epsilon_0^C = -1 + 2a_2 + a_2', \quad (4)$$

where $d = D/|J|$, $a = K_1/z|J|$, $a_2 = K_2/z|J|$, and $a_2' = A/|J|$ are the normalized parameters of the anisotropic interactions and δ_K is the angle between the local easy

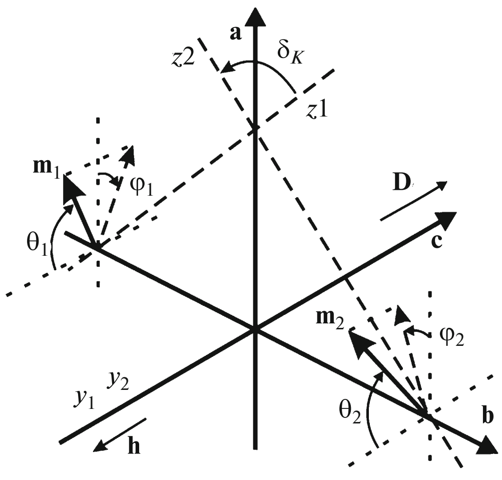

Fig. 1. Orientation of sublattice moments m_1 and m_2 in a field applied in the specular symmetry axis.

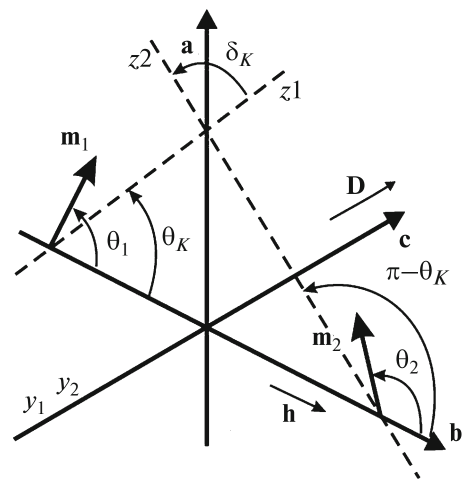

Fig. 2. Orientation of sublattice moments m_1 and m_2 in a field applied in the plane containing the noncollinear easy SIA axes $z1$ and $z2$.

SIA axes $z1$ and $z2$ of the sublattices. The ground state is the phase with the minimum energy. Thus, the threshold conditions for the anisotropy parameters are set by inequalities (2)–(4) between the energies.

3. $\mathbf{H} \parallel \mathbf{c}$

The magnetic field directed along the hard magnetization axis has a symmetry that does not coincide with the symmetry of the ground state. Let us consider the change in the orientation of the moments for the case

$$\epsilon_0^A < \epsilon_0^B, \epsilon_0^C \tag{5}$$

in the field applied in the symmetry plane m_b ($\mathbf{H} \parallel \mathbf{c}$) (Fig. 1). In this case, the magnetic field symmetry $2'_a m'_b$ preserves the symmetry m'_b of the magnetic structure. In the system of coordinates with the polar **c** axis, the angles of orientation of the moments and SIA axes have the form

$$\theta_2 = \theta_1 = \theta, \quad \varphi_2 = -\varphi_1 = \varphi,$$
$$\theta_K = \pi/2, \quad \varphi_K = \delta_K/2.$$

The energy minimization

$$\begin{aligned}\epsilon(\theta,\varphi) = &-\cos^2\theta - \sin^2\cos 2\varphi - d\sin^2\theta\sin 2\varphi \\ &+ 2a\sin^2\theta\cos^2(\varphi - \delta_K/2) \\ &+ (2a_2 + a'_2)\cos^2\theta - 2h\cos\theta\end{aligned} \tag{6}$$

over polar angle θ yields two solutions:

(I) $\theta = 0$ is phase C with the energy $\epsilon^C = -1 + 2a_2 + a'_2 - 2h$, where $h = g\mu_B H_0/|J|Sz > 0$.

(II) $0 < \theta \le \pi/2$. At $h = 0$, we have $\theta = \pi/2$ and this solution passes to phase A. Minimization of Eq. (6) over angle φ yields its value independent of h and θ

$$\tan 2\varphi = \frac{d - a\sin\delta_K}{1 - a\cos\delta_K}. \tag{7}$$

At $0 < h < h_c$, we obtain an explicit dependence of the normalized energy values, the projection of the magnetization onto the field direction, and the constant susceptibility upon reorientation from phase A to phase C

$$\begin{aligned}\epsilon(h) &= \epsilon_0^A - h^2/h_c^C, \\ m(h) &= \cos\theta = h/h_c^C, \\ \chi(h) &= dm/dh = 1/h_c^C,\end{aligned} \tag{8}$$

where h_c^C is the field at which the reorientation ends with phase C:

$$\begin{aligned}h_c^C = &\sqrt{(1 - a\cos\delta_K)^2 + (d - a\sin\delta_K)^2} \\ &- 1 - a + 2a_2 + a'_2.\end{aligned} \tag{9}$$

4. $\mathbf{H} \parallel \mathbf{b}$

The external magnetic field applied orthogonally to the symmetry plane and twofold axis ($\mathbf{H} \parallel \mathbf{b}$) has the symmetry $2'_a m_b$ and, consequently, violates the initial symmetry of phase A. When threshold condition (5) is met, the moments $m_{1,2}$ remain in the plane containing the easy SIA axes and the external field; the problem remains coplanar with polar angles θ_1 and θ_2 as independent variables in the system of coordinates with the polar **b** axis (Fig. 2).

The minimization of the normalized energy

$$\begin{aligned}\epsilon(\theta_1,\theta_2) &= -\cos(\theta_1-\theta_2) + d\sin(\theta_1-\theta_2)\\ &+ a(\cos^2(\theta_1-\theta_K)+\cos^2(\theta_2+\theta_K))\\ &- h(\cos\theta_1+\cos\theta_2)\end{aligned} \tag{10}$$

yields the system of equation

$$\begin{aligned}&a\sin(\theta_1+\theta_2)\cos(\theta_1-\theta_2-2\theta_K)\\ &- h\sin\frac{\theta_1+\theta_2}{2}\cos\frac{\theta_1-\theta_2}{2} = 0,\\ &\sin(\theta_1-\theta_2) + d\cos(\theta_1-\theta_2) - a\cos(\theta_1+\theta_2)\\ &\times\sin(\theta_1-\theta_2-2\theta_K) + h\cos\frac{\theta_1+\theta_2}{2}\sin\frac{\theta_1-\theta_2}{2} = 0,\end{aligned} \tag{11}$$

which has the solutions corresponding to the two magnetic phases

$$\begin{aligned}A:\quad &\cos\frac{\theta_1+\theta_2}{2} = \frac{h\cos((\theta_1-\theta_2)/2)}{2a\cos(\theta_1-\theta_2-2\theta_K)},\\ B:\quad &\theta_2 = -\theta_1.\end{aligned} \tag{12}$$

The angles θ_K and $\pi-\theta_K$ of the easy SIA axes are related to δ_K as $\pi - 2\theta_K = \delta_K$. Taking into account Eq. (12), we exclude the half-sum of the angles and obtain, for each phase, the equations of state that depend on only one parameter: the difference between the sublattice angles

$$\delta = \theta_2 - \theta_1.$$

In zero external field, for each phase we obtain

$$\begin{aligned}A:\quad &\tan\delta_0^A = \frac{d-a\sin\delta_K}{1-a\cos\delta_K},\quad \delta_0 = \theta_2-\theta_1\big|_{h=0},\\ B:\quad &\tan\delta_0^B = \frac{d+a\sin\delta_K}{1+a\cos\delta_K}.\end{aligned} \tag{13}$$

The corresponding energies are equal to (2) and (3). In the presence of a magnetic field, phase A becomes asymmetric relative to the symmetry plane and its solution is expressed through parameter δ as

$$\begin{aligned}h^A(\delta) &= 2\cos(\delta-\delta_K)\\ &\times\sqrt{\frac{a(\sin\delta - a\sin(\delta-\delta_K) - d\cos\delta)}{\sin\delta_K - \sin(\delta-\delta_K)}},\\ \epsilon^A(\delta) &= -\cos\delta - d\sin\delta + a(1+\cos(\delta-\delta_K))\\ &+ \frac{(h^A(\delta))^2\cos^2(\delta/2)}{2a\cos(\delta-\delta_K)},\\ m^A(\delta) &= -\frac{\cos^2(\delta/2)}{2a\cos(\delta-\delta_K)}h^A(\delta).\end{aligned} \tag{14}$$

Phase B contains the magnetic symmetry in the field and its solution has the form

$$\begin{aligned}h^B(\delta) &= -2\cos(\delta/2) + \frac{d\cos\delta - a\sin(\delta-\delta_K)}{\sin(\delta/2)},\\ \epsilon^B(\delta) &= -\cos\delta - d\sin\delta + a(1-\cos(\delta-\delta_K))\\ &- 2h^B(\delta)\cos(\delta/2),\\ m^B(\delta) &= \cos(\delta/2).\end{aligned} \tag{15}$$

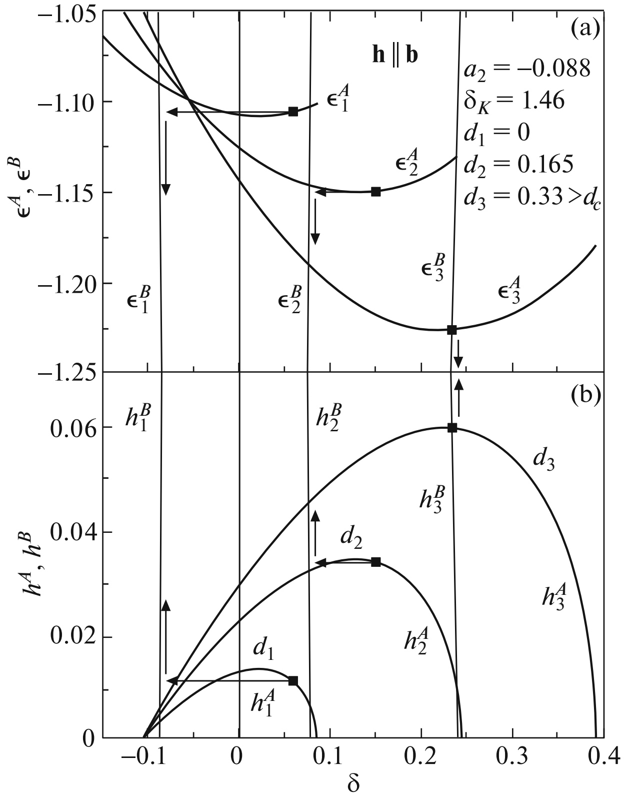

Fig. 3. Dependences of (a) energy $\epsilon^{A,B}$ and (b) field $h^{A,B}$ in phases A and B on angle δ between the sublattice moments at **h** ∥ **b** at different DM exchange values d_{1-3}. Horizontal arrows show the jump in the values of $\delta \to \delta'$ at the first-order orientational phase transition for d_1 and d_2. Vertical arrows show the energy and field variation in phase B.

The obtained parametric solutions for both phases determine the character of reorientation of the sublattice moments from phase A to phase B in a magnetic field (Fig. 3). With an increase in the field, the angle between the sublattice moments decreases and when the conditions

$$\begin{aligned}h^A(\delta) &= h^B(\delta'),\\ e^A(\delta) &= \epsilon^B(\delta')\end{aligned} \tag{16}$$

are met, the reorientation ends with phase B. Figure 3 shows the energy and field changes for three values of the DM exchange parameter d. At $d = 0$ and $d = 0.165$, conditions (13) are satisfied at different angles ($\delta \neq \delta'$) between the sublattices and the reorientation ends with a first-order phase transition with a jump in the parameter δ and, consequently, in the projection of

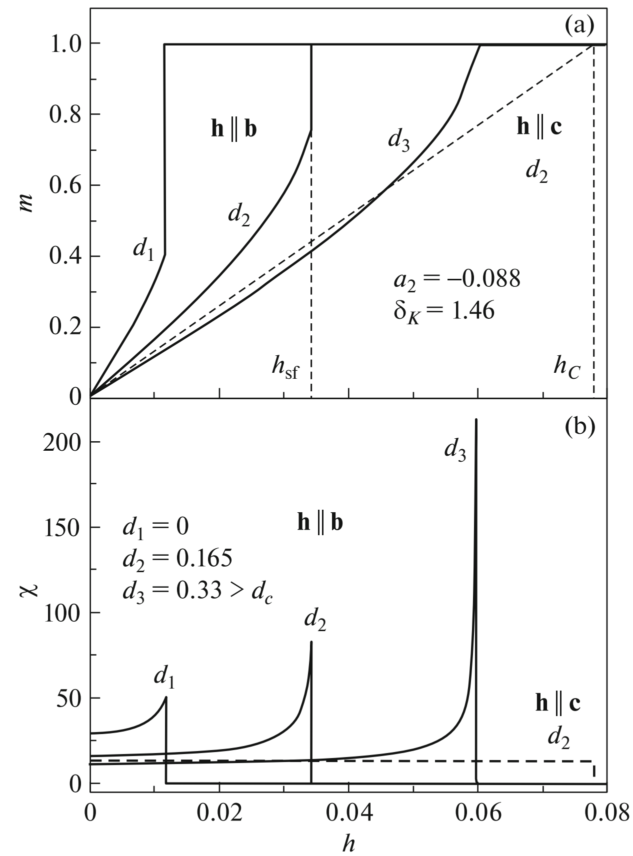

Fig. 4. Field dependences of (a) magnetization m and (b) susceptibility χ in a field applied along the **b** axis (solid lines) at different DM exchange values d and in a field applied along the **c** axis (dashed lines) at $d = d_2$. The values of anisotropy parameters a, δ_K, and d_2 correspond to the $PbMnBO_4$ crystal, in which the reorientation ends with a spin-flop transition in field h_{sf}.

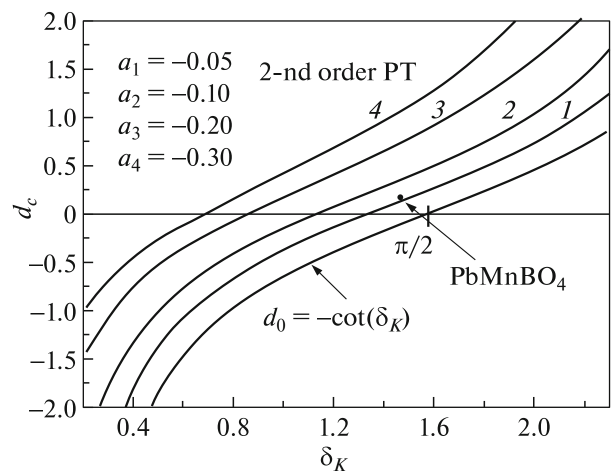

Fig. 5. Dependence of limit DM exchange value d_c on angle δ_K between the easy SIA axes of the sublattices at different values of the easy-axis anisotropy parameter $a < 0$. The reorientation of the sublattice moments from phase A to phase B in a field applied along the **b** axis ends with a first-order transition at $d_0(\delta_K) < d < d_c(a, \delta_K)$ and a second-order transition at $d_c(a, \delta_K) < d$. The dot $PbMnBO_4$ marks the anisotropy parameters corresponding to the magnetization measurements on this crystal [9].

the magnetization onto the applied field direction. At $d = 0.33$, $\delta = \delta'$ and the transition from phase A to phase B has a continuous character of two second-order phase transitions. Thus, there is a limit value of the DM exchange parameter d_c depending on the SIA parameters above which a first-order transition does not occur. Figure 4 shows field dependences of the magnetization and differential susceptibility for three d values in a field applied along the **b** axis. For comparison with the data of the magnetic measurements on the $PbMnBO_4$ crystal, the angle between the long axes of the oxygen octahedra surrounding the neighboring magnetic Mn^{3+} ions was taken as angle δ_K between the easy axes [10, 11]. The numerical values of anisotropy parameter a and DM exchange parameter $d_2 = 0.165$ in Figs. 3, 4 correspond to the best agreement between calculated field dependence of magnetization (14) and the results of the magnetic measurements in a field applied along the orthorhombic crystal **b** axis [9]. The field h_{sf} corresponds to the field of the spin-flop transition along the weak antiferromagnetism vector $\mathbf{L} = \mathbf{m}_1 - \mathbf{m}_2$. At the symmetry of model (1), the second SIA constant a_2 and the anisotropic symmetric exchange a_2' do not affect the energy of the moments lying in the ab plane during the transition $A \to B$. Therefore, the resulting value $2a_2 + a_2' = -0.051$ of these parameters is determined from the comparison of linear dependence of magnetization (8) (the dashed line in Fig. 4) with the experimental dependence in the field $\mathbf{H} \parallel \mathbf{c}$. The susceptibility remains constant upon reorientation in this field, in contrast to the characteristic nonlinear dependence with a sharp maximum in the field of completion of the reorientation $A \to B$.

Comparing energies of phases A (2) and B (3), we obtain a simple relation between the DM exchange value and the noncollinearity of the easy SIA axes at which the above-considered reorientation occurs

$$d > d_0 = -\cot \delta_K. \quad (17)$$

Note that Eq. (17) is independent of the anisotropy value a.

The DM exchange value d_c above which the reorientation ends with a second-order phase transition depends on both the angle between the anisotropy axes and the SIA value a. Figure 5 shows angular dependences for different values of this quantity. Thus, the range of DM exchange values at which the transition is first-order lies between the $d_c(a, \delta_K)$ curve and lower $d_0(\delta_K)$ curve (17) passing through $\pi/2$. Above the d_c curves, the reorientation ends with a second-order phase transition (the 2nd order PT region). The dot marks the d_2 and δ_K values corresponding to $PbMnBO_4$.

5. H ‖ a

If phase B is the ground state in zero external field, inequality (17) changes its sign. The symmetry of model (1) relative to the permutation $i \leftrightarrow j(1 \leftrightarrow 2) \Rightarrow A \leftrightarrow B$ allows us, at $\mathbf{H} \parallel \mathbf{a}$, to use the results of (10)–(13) with the replacement

$$d \to -d, \quad \delta_K \to \pi - \delta_K.$$

Figure 5 is symmetrical relative to such transformations. The field dependence of the magnetization remains nonlinear and is also accompanied by a jump at $d_0 < d < d_c$.

If phase C is the ground state in zero external field, the field applied along the a axis will cause a reorientation similar to the transition $A \to C$. To determine the field dependences of the magnetization projection onto the **a** axis and the energy, we use Eq. (6) for the energy with the replacement of the Zeeman term by

$$\epsilon_h = -2h\sin\theta\cos\varphi.$$

The energy minimization in the reorientation region $h < h_c$ yields a constant azimuth angle of orientation of the sublattice moments in the system of coordinates with the polar **c** axis

$$\tan\varphi = \frac{d - a\sin\delta_K}{2 + a(1-\cos\delta_K) - 2a_2 - a_2'}, \qquad (18)$$

the linear field dependence of the magnetization projection

$$m(h) = \sin\theta\cos\varphi = m_c^A h/h_c^A, \qquad (19)$$

and the quadratic dependence of the energy

$$\epsilon(h) = \epsilon_0^C - m_c^A h^2/h_c^A. \qquad (20)$$

After obtaining the field of the reorientation completion

$$h_c^A = \frac{(a(1+\cos\delta_K) - 2a_2 - a_2')U - V^2}{\sqrt{U^2+V^2}}, \qquad (21)$$

where $U = 2 + a(1 - \cos\delta_K) - 2a_2 - a_2'$ and $V = d - a\sin\delta_K$, the magnetization projection

$$m_c^A = \frac{U}{\sqrt{U^2+V^2}} \qquad (22)$$

does not saturate and continues to asymptotically tend to saturation at $h \to \infty$. At the exact compensation of two noncollinearity mechanisms $d = a\sin\delta_K$, we obtain a particular case of a collinear one-sublattice ferromagnet with the noncollinear SIA axes and the anisotropy field $h_c^A = 2a\cos^2\delta_K/2 - 2a_2 - a_2' = -h_c^C$ (9). At $\delta_K = 0$, we obtain the case of a ferromagnet with the collinear axes and DM exchange. However, in this particular case (as in the case $a = 0$), model (1) describes a magnet with the inversion center between the sublattice moments; therefore, it is necessary to set $d = 0$.

6. DISCUSSION

The exact solution of model (1) in relatively simple analytical expressions is caused by its symmetry. The question arises about the criteria for its applicability to real magnets with lower symmetry. In particular, the crystal structure of the ferromagnet $PbMnBO_4$ (sp. gr. *Pnma*) used in the comparison and estimation of the anisotropic interactions has four translationally nonequivalent sites of magnetic ions and does not contain a second-order axis between them. In this four-sublattice magnet, the planes of the easy anisotropy axes in chains with the maximum isotropic exchange are rotated by an angle of 30° relative to the orthorhombic **a** axis. The ferromagnetic exchange between chains brings spins out of the anisotropy planes. As a result, in the field applied along chains (the **b** axis), the problem is no longer coplanar, even for each individual chain. At the maintained mirror plane, the axes of the two-ion anisotropic interactions remain in the symmetry plane. However, the absence of a twofold axis means that the direction of the second axes of the biaxial SIA of each sublattice does not coincide with the **c** axis and may lie beyond the symmetry plane. This leads to a change in the equations for the moment orientation angles in a field applied along this axis. The azimuthal angle does not remain constant upon reorientation and, consequently, the linearity of the field dependence of the magnetization is violated. Despite these differences between exactly solved model (1) and the ferromagnet $PbMnBO_4$, the linear field dependence of the magnetization in a field applied along the **c** axis and the nonlinear dependence with a magnetization jump along the **b** axis are in good quantitative agreement with the results of the magnetic measurements on this crystal. This allows us to conclude that the considered model is adequate to the main features of the anisotropic interactions in this magnet, i.e., the noncollinearity of the local easy SIA axes and the significant effect of the DM exchange.

The field dependences of the magnetization in the investigated model are highly sensitive to the changes in the parameters of the anisotropic interactions. The interactions differently affect the parameters of the field dependences of the magnetization, including the initial susceptibility for both field orientations, the field of the spin-flop transition, and the value of the magnetization jump upon completion of the reorientation in a field applied along the **b** axis. This makes it possible to find the anisotropy parameters with the higher accuracy as compared with the experiment. Using the value of the ferromagnetic exchange between the sublattices obtained from the paramagnetic Curie temperature and the $PbMnBO_4$ ordering temperature [14], we obtain $K_1 = -3.6$ K. The large SIA value is related to the strong Jahn–Teller distortion of the oxygen octahedron surrounding the Mn^{3+} ions. Mn–O distances of 2.225, 1.885, and 1.99 Å are similar to the distances in distorted octahedra in the rare-earth $RMnO_3$ perovskites [3]. Let us compare the obtained K_1 value with the SIA of the most well-studied manganite $LaMnO_3$ (the Mn–O distances are 2.181, 1.914, and 1.966 Å, respectively). The SIA energy of model (1) is counted from the energy of spins at the orientation along the local hard axes x_1 and x_2. Such a form is convenient to clearly distinguish the easy plane in which reorientation occurs in the external field $\mathbf{H} \parallel \mathbf{b}$. The transition to the most widespread notation of the anisotropy in the local axes for each sublattice ($i = 1, 2$) has the form

$$K_1 S_i^{zi^2} + K_2 S_i^{yi^2} = D S_i^{yi^2} + E(S_i^{zi^2} - S_i^{xi^2}) + \text{const},$$

where $D = K_2 - K_1/2$, $E = K_1/2$. Depending on the splitting of the orbital levels t_{2g} and e_g, the value of the spin-orbit interaction and the degree of mixing of the electronic functions $d_{3z^2-r^2}$ and $d_{x^2-y^2}$, the calculation of constant E along the local axes in the second order of the perturbation theory in the spin-orbit interaction yields the values ranging between −1 K and −3.5 K [3, 31, 32]. The SIA values used in the interpretation of the experimental field dependence of the magnetization, the value of the weak ferromagnetic moment, neutron scattering, and resonance investigations on $LaMnO_3$ depend on the models used and the DM interaction taken into account. In the simplest and most widespread model of a two-sublattice antiferromagnet, all spins with the local noncollinear easy SIA axes in the ferromagnetically ordered planes are considered to be parallel [32–36]. In this case, the SIA in the plane is determined by one term $D_b S_b^2$, where $\mathbf{b}$ is the easy orthorhombic axis of the crystal. This approximation is uncontrollable, since the nondiagonal SIA terms arising during the transition from the local to orthorhombic axes are ignored [37]. This can be easily shown using model (1) by rotating the local sublattice axes by angle $\pm\delta_K/2$ around the $\mathbf{c}$ axis. If the sublattice moments remain in the easy plane ab, then, in addition to the diagonal SIA term, the nondiagonal ones occur:

$$K_1 \cos\delta_K S_i^{a2} \pm K_1 \sin\delta_K S_i^a S_i^b. \quad (23)$$

The latter determine the noncollinearity of the ferromagnetic sublattices, which exists at any ratio between the anisotropy and exchange [19]. It can be seen from (23) that it is incorrect to use simple projections of the local anisotropy at the transition to the orthorhombic axes [32]. If we compare diagonal term (23) with the anisotropy constant $D_b = -1.92$ K along the $\mathbf{b}$ axis obtained from neutron study of the spin-wave spectrum [33], then, at $\delta_K \approx 70°$ [32], we obtain a value of $E \approx -2.8$ K, which lies in the above-mentioned range of theoretical values. The K_1 value for $PbMnBO_4$ coincides with the $2E$ value following from the parameters for Mn^{3+} used in the analysis of the resonance spectrum in the mixed manganites $La_{1-x}Ca_xMnO_3$ with the charge ordering of Mn^{3+} and Mn^{4+} ions [38].

The above-made comparison of the SIA values of Mn^{3+} ions in $PbMnBO_4$ and $LaMnO_3$ is qualitative. In the first compound, the DM exchange strongly affects the magnetic anisotropy (in (1), the constant is $D =$ 3.4 K). In the investigated two-sublattice model, the contributions of the second anisotropy constant and symmetric anisotropic exchange to the field dependences have the same form. The comparison with the experiment allows one to determine only their total value. The possibility of finding these parameters separately appears during the numerical analysis of the four-sublattice model, where the absence of a twofold rotary axis is taken into account. This analysis will be presented elsewhere; we only note that the resulting parameters are similar to the values for the two-sublattice model and confirm the conclusion about the need for taking into account the single- and two-ion anisotropy mechanisms in $PbMnBO_4$.

7. CONCLUSIONS

Depending on the fulfillment of the threshold relations between the parameters of the anisotropic interactions, the ground state of a two-sublattice ferromagnet with the noncollinear easy SIA axes of the sublattices and the anisotropic symmetric and antisymmetric exchanges between the sublattices is one of the three states with the total magnetic moment $\mathbf{M} = \mathbf{m}_1 + \mathbf{m}_2$ directed along the principal crystal axes

$$A: \quad \mathbf{M} \parallel \mathbf{a}$$

$$d > -\cot\delta_K,$$

$$a - \sqrt{(1 - a\cos\delta_K)^2 + (d - a\sin\delta_K)^2} < -1 + 2a_2 + a_2';$$

$$B: \quad \mathbf{M} \parallel \mathbf{b}$$

$$d < -\cot\delta_K,$$

$$a - \sqrt{(1 + a\cos\delta_K)^2 + (d + a\sin\delta_K)^2} < -1 + 2a_2 + a_2';$$

$$C: \quad \mathbf{M} \parallel \mathbf{c}$$

$$-1 + 2a_2 + a_2' < a - \sqrt{(1 \mp a\cos\delta_K)^2 + (d \mp a\sin\delta_K)^2}.$$

The anisotropy parameters normalized to the isotropic exchange between the sublattices are determined in Section 2 and d_K is the angle between the local easy axes of the sublattices. In phases A and B, the noncollinear moments of the sublattices lie in the plane of the noncollinear SIA axes; in phase C, the collinear

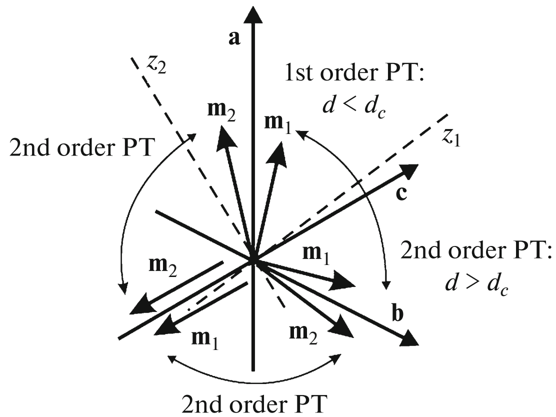

Fig. 6. Evolution of the magnetic phases of a two-sublattice ferromagnet with the noncollinear SIA axes in a magnetic field applied along the hard magnetization axes of the crystal.

moments of the sublattices are orthogonal to the plane of these axes (Fig. 6). The orientational transition A, $B \leftrightarrow C$ between phases in a magnetic field applied along the hard magnetization axes occurs in the form of two second-order phase transitions with a linear field dependence of the magnetization projection onto the field direction. The reorientation between the noncollinear states A and B, depending on the antisymmetric exchange value d, ends with a phase transition of either the first ($d < d_c$) or second ($d > d_c$) order. The threshold value of the DM exchange parameter d_c depends on the angle between the easy SIA axes and the SIA value. The field dependence of the magnetization projection is nonlinear and the first-order transition is accompanied by a magnetization jump. The differential susceptibility has a sharp maximum in the field of the reorientation completion.

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CONFLICT OF INTEREST

The author declares that he has no conflicts of interest.

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