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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

# The Incommensurate Magnetic Structure of a Tetragonal Antiferromagnet with Antisymmetric Exchange

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**Abstract**—Analysis of the incommensurate magnetic structure that emerges for two coexisting types of the antisymmetric Dzyaloshinski–Moriya exchange interaction (the weakly ferromagnetic component of vector **D** along the tetragonal axis and the helicoidal component distributed in the tetragonal plane) is carried out for the first time for a tetragonal antiferromagnet. The helicoidal component for each pair of interacting spins has a 2D distribution; its direction in the tetragonal plane depends on the direction of the exchange bond in each pair. The Lifshits invariant of the Ginzburg–Landau functional is obtained, which is responsible for the formation of an incommensurate magnetic structure for such a distribution. It is shown in the mean field approximation that the incommensurate magnetic structure that forms in this case is a nonlinear double helicoid with a modulation vector lying in the tetragonal plane and with a varying angle between the polarization planes of quasi-antiferromagnetic sublattices. The ground state of the magnet is degenerate in the orientation of the modulation vector in the tetragonal plane. The rate of variation in the orientations of moments in the polarization planes passing through the tetragonal axis is controlled by the angle between the directions of the moments and the tetragonal axis. The local weakly ferromagnetic moment remaining in the polarization plane varies in magnitude and sign. The relation between the orientations of the modulation and polarization vectors is derived for the cases of simple and inversion tetragonal axes in the space symmetry group of the crystal.

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## 1. INTRODUCTION

The symmetry of the crystal structure of a magnet determines the magnetic anisotropy and (to a considerable extent) the type of magnetic ordering. Combinations of the order parameter (magnetic moment) in the free energy expansion (Ginzburg–Landau functional), which are invariant to specific magnetic symmetry transformations, make it possible to analyze the form of the possible magnetic structure and the conditions of its formation [1, 2]. The invariance of energy relative to time inversion in zero magnetic field makes it possible to confine the symmetry analysis of magnetic anisotropic interaction to the symmetry elements of a crystallographic point group. In a microscopic description of a magnet, magnetic symmetry is taken into account in the form of the *g*-factor, one-ion anisotropy, and anisotropy of spin–spin interaction even at the stage of writing the spin Hamiltonian. The microscopic Hamiltonian must also be invariant to symmetry transformations (anisotropic Hamiltonian components associated with these transformations and distributed in space must be transformed onto one another). An example of the effect of a spatial distribution of anisotropic interaction on the type of magnetic ordering is the antisymmetric Dzyaloshinski–Moriya exchange [3, 4]:

$$H_{\mathrm{D}} = \sum_{\mathbf{r},\,\mathbf{r}'} \mathbf{D}_{\mathbf{r},\,\mathbf{r}'}[\mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}'}]. \qquad (1)$$

It emerges as a result of spin–orbit interaction and hence reflects the symmetry properties of the spatial distribution of orbitals of magnetic ions and surrounding ligands; it is transformed upon a transition from one pair of interacting spins to another depending on the local symmetry operation generating this transformation. If the transition is performed via an inversion operation that does not change the components of the axial vectors of magnetic moments and has a center at a magnetic ion **r**, such an invariant pair of anisotropic terms of the Hamiltonian has the form

$$\mathbf{D} \cdot ([\mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}'}] + [\mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}''}]).$$

If the rotational binary axis ($2_z$) changing the signs of the vector components perpendicular to this axis passes through magnetic ion **r** instead of the inversion center, the sum of the vector products will be preserved only for the *z* components of scalar triple product (1). For the remaining two components (*x*, *y*), we obtain the difference

$$D_{xy}(\mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}'} - \mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}''})_{xy}.$$

If we consider the spatial distribution for antisymmetric exchange in one direction (labeling spins in this direction and taking into account the transposition rules of the vector product), we obtain the corresponding Hamiltonians differing in the $x$ and $y$ components:

$$H_{xy}^{S} = D_S \sum_i (-1)^i [\mathbf{S}_i \times \mathbf{S}_{i+1}]_{xy},$$
$$H_{xy}^{U} = D_U \sum_i [\mathbf{S}_i \times \mathbf{S}_{i+1}]_{xy}. \quad (2)$$

The first of these Hamiltonians describes the antisymmetric staggered Dzyaloshinski–Moriya (SDM) exchange and leads to alternation of magnetic moment canting and, as a consequence, to weakly ferromagnetic ordering of the magnet with predominant antiferromagnetic exchange [5, 6]. The second type of antisymmetric exchange is the uniform Dzyaloshinski–Moriya (UDM) exchange, which leads to a deviation of magnetic moments in the same direction (fixed rotation of magnetization) [7–9]. A magnetic structure formed in this case has a magnetic lattice period that generally does not coincide with the multiple period of the crystal lattice (incommensurate magnetic structure). For $d$ ions with "frozen" orbital moments, the value of $D$ is much smaller as a rule than the symmetric Heisenberg exchange, and such a structure has a long period. A necessary condition for the existence of antisymmetric exchange (1) in both cases is the absence of an inversion center between interacting ions [10]. However, in contrast to the SDM exchange, a necessary condition for the UDM exchange is the absence of an inversion center at the magnetic ion as well. If the inversion center and the rotational axis are simultaneously preset at a magnetic ion, the components of "vector" $\mathbf{D}_S$, which are transverse to the direction of the axis, must be zero. Only the component parallel to the axis may differ from zero.

We must make the following general remark concerning the analysis of the $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ transformation by symmetry operations. The representation of the antisymmetric exchange Hamiltonian in the form of scalar triple product (1) is just a convenient analytic form. The transformation of components of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ is defined by a transformation of invariants constructed on magnetic moment components. Consequently, it is convenient to use the available expressions for invariants relative to symmetry transformations [10] connecting individual pairs of interacting spins, to transform these components with the help of the symmetry operations transforming pairs of spins into one another and then to determine the components of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ corresponding to invariants of the new pair. This procedure can be replaced by the transformation of vector $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ itself if we take into account its axiality and antisymmetry relative to spatial inversion $\mathbf{r} \longleftrightarrow \mathbf{r}'$ [2]. Both methods for determining the spatial distribution of antisymmetric exchange lead to the same results. However, the application of symmetry transformations to both cofactors of the scalar product in (1) leads to incorrect results. For example, the inversion center between a pair of spins leaves the product invariant since the sign reversal due to spatial inversion occurs twice.

In the presence of symmetry elements transforming pairs of interacting spins with different orientations in space into one another, a distribution over the directions of vector $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ for different pairs appears. The symmetry of such a distribution is controlled by crystallographic symmetry and is generally three-dimensional. The existence of an incommensurate magnetic structure formed as a result of UDM exchange occurring in highly symmetric crystals was established first for cubic crystals of MnSi and FeGe [11, 12]. A characteristic feature of a highly symmetric distribution of the UDM exchange is the nearly isotropic behavior of the helicoid in a magnetic field. However, analysis of the helicoid structure was performed for only one component even in the case when the 2D distribution of the $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ components of a tetragonal crystal in the microscopic Hamiltonian was written in explicit form [13]. Such a generally accepted approach to describing an incommensurate magnetic structure, which is based on the 1D distribution of antisymmetric exchange, can be treated only as a model simplification for a low-symmetry case. The existence of a tetragonal rotational axis in a tetragonal crystal, which transforms pair interactions in the basal plane into one another, imposes constraints on the transverse components of antisymmetric exchange: a 2D spatial distribution of the UDM exchange is formed. In the general case, the longitudinal weakly ferromagnetic component of the SDM exchange is preserved. The magnetic moment distribution in such a system is much more complicated than in the 1D case.

It is convenient to describe the magnetization distribution in an incommensurate magnetic structure by introducing modulation vector $\mathbf{k}$ and polarization vector $\mathbf{m}$ [14] since it is these two vectors that are detected in experiments on neutron scattering. In the case of a simple spiral structure formed in the case of a 1D distribution of the UDM exchange in the absence of anisotropy in the polarization plane, one vector $\mathbf{k}$ and one vector $\mathbf{m}$ are sufficient since the magnetic moment components vary as harmonic functions of coordinates, and the polarization plane in space remains unchanged. The presence of any type of magnetic anisotropy that removes degeneracy in the polarization plane of an incommensurate magnetic structure complicates the spatial distribution of magnetization (necessitates the introduction of a set of vectors $\mathbf{k}$ defining the law of this distribution). In experiments, this is manifested in the emergence of additional peaks in the neutron scattering spectrum. Traditionally, such a structure is referred to as a "soliton lattice" [1, 14]. In the case of an isotropic mechanism of formation of an incommensurate magnetic structure as a result of

the competition of Heisenberg exchanges between different magnetic neighbors, the direction of propagation of the incommensurate magnetic structure (**k**) and the polarization plane are not interrelated (the latter is fixed by magnetic anisotropy, e.g., one-ion anisotropy). In the case when an incommensurate magnetic structure is formed by anisotropic mechanisms (in particular, antisymmetric exchange), the polarization plane of the magnetic structure and the spatial modulation vector are interrelated, and the formation of a complex structure with a set of vectors **k** must also lead to spatial modulation **m**.

The goal of this study is to analyze the ground state of a system of spins coupled by predominant antiferromagnetic exchange with different types of antisymmetric exchange existing simultaneously in a tetragonal crystal (2D distribution of uniform component $D_U$ and 1D weakly ferromagnetic component $D_S$).

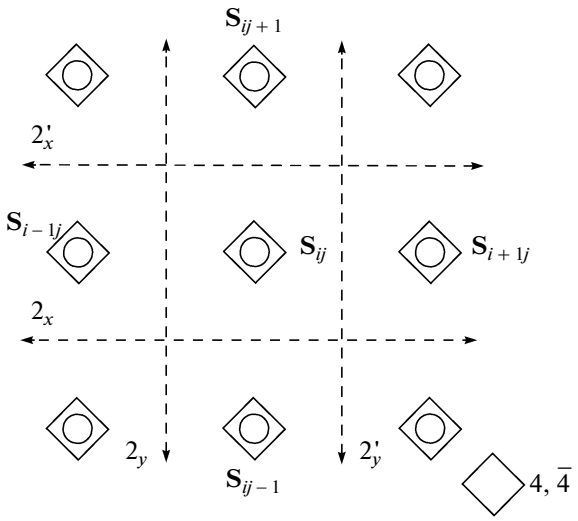


**Fig. 1.** Symmetry elements of the tetragonal crystal structure: the tetrad axis is directed along the tetragonal axis on magnetic ions and simple binary axes are parallel to the tetragonal plane.

## 2. HAMILTONIAN AND INVARIANTS

The importance of studying the effect of spatial distribution of antisymmetric exchange in a magnet with a tetragonal crystalline structure on the ground state is obvious. The existence of a tetragonal plane with mutually perpendicular equivalent directions ensures the distribution of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ with a varying orientation of the $D_U$ component. The symmetry element defining the spatial distribution (generating element) in our case is the tetragonal axis. It is also convenient to single out the symmetry operation limiting possible antisymmetric combinations of the components of interacting pairs of spins (limiting operation). The latter is not necessary for determining nonvanishing components of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$: the combination of two generating elements can also preserve only quite definite components of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ for each bond. In a tetragonal crystal, the role of generating elements can be played by the helical axis and slip and reflection planes, as well as other operations transforming various pairs of interacting spins into one another. Limiting symmetry operations can be simple binary axes or reflection symmetry planes passing between interacting spins.

Let us consider the simplest case with a generating tetragonal axis and limiting binary axes parallel to the tetragonal plane (Fig. 1). Since simple and inversion tetragonal axes transform the spatial indices of $\mathbf{D}_{\mathbf{r},\mathbf{r}'}$ in different ways, this leads to different relative orientations of tetragonal components of $D_U$, which in turn leads to its own peculiarities of the incommensurate magnetic structure in these cases. Consequently, following the proposed method for obtaining invariants of the antisymmetric exchange Hamiltonian [15], we will consider both these cases simultaneously, although incommensurate magnetic structures with a simple tetragonal axis at an ion are not known (as a rule, such an axis exists simultaneously with an inversion center). Invariant antisymmetric combinations of the second-order magnetic components for binary axes (see Fig. 1) have the form [10]

$$\begin{aligned} &2_x\colon\ a)\ \ m_x l_y - m_y l_x, \quad b)\ \ m_x l_z - m_z l_x;\\ &2_y\colon\ a)\ \ m_x l_y - m_y l_x, \quad b)\ \ m_y l_z - m_z l_y, \end{aligned} \tag{3}$$

where $m_\alpha$ and $l_\alpha$ are the local components of the ferro- and antiferromagnetism vectors constructed on the magnetic moments connected with each axis separately. For example, in the notation used in Fig. 1, we have

$$\begin{aligned} 2_x\colon\ &\mathbf{m}(\mathbf{r}) = \mathbf{S}_{ij} + \mathbf{S}_{ij-1}; \quad \mathbf{l}(\mathbf{r}) = \mathbf{S}_{ij} - \mathbf{S}_{ij-1},\\ 2'_x\colon\ &\mathbf{m}(\mathbf{r}) = \mathbf{S}_{ij} + \mathbf{S}_{ij+1}; \quad \mathbf{l}(\mathbf{r}) = \mathbf{S}_{ij} - \mathbf{S}_{ij+1},\\ 2_y\colon\ &\mathbf{m}(\mathbf{r}) = \mathbf{S}_{ij} + \mathbf{S}_{i-1j}; \quad \mathbf{l}(\mathbf{r}) = \mathbf{S}_{ij} - \mathbf{S}_{i-1j},\\ 2'_y\colon\ &\mathbf{m}(\mathbf{r}) = \mathbf{S}_{ij} + \mathbf{S}_{i+1j}; \quad \mathbf{l}(\mathbf{r}) = \mathbf{S}_{ij} - \mathbf{S}_{i+1j}. \end{aligned} \tag{4}$$

The tetragonal axis at magnetic ion $\mathbf{S}_{ij}$ leaves the combination of groups *a* from formula (3) invariant:

$$\begin{aligned} &S^x_{i-1j}S^y_{ij} - S^y_{i-1j}S^x_{ij} + S^x_{i+1j}S^y_{ij} - S^y_{i+1j}S^x_{ij}\\ &+ S^x_{ij-1}S^y_{ij} - S^y_{ij-1}S^x_{ij} + S^x_{ij+1}S^y_{ij} - S^y_{ij+1}S^x_{ij}. \end{aligned} \tag{5}$$

The second invariant constructed on combinations of groups *b* from formula (3) has the form

$$\begin{aligned} &S^y_{i-1j}S^z_{ij} - S^z_{i-1j}S^y_{ij} - S^y_{i+1j}S^z_{ij} + S^z_{i+1j}S^y_{ij}\\ &\mp (S^x_{ij-1}S^z_{ij} - S^z_{ij-1}S^x_{ij} - S^x_{ij+1}S^z_{ij} + S^z_{ij+1}S^x_{ij}). \end{aligned} \tag{6}$$

Here and below, the upper and lower signs of the antisymmetric *y* component correspond to the cases with a simple and inversion tetragonal axis, respectively. The absence of an inversion center at a magnetic ion can be ensured by alternating displacement of groups

of ligand ions, paired displacement of ligands or ligand groups along the tetragonal $z$ axis, or the corresponding distortions of the ligand surroundings. In the case of the tetragonal inversion axis, a tetrahedral surrounding in the nearest magnetic neighbors is formed (coordination number $Z = 4$). The strongest interaction in the system, viz., the Heisenberg exchange,

$$J \gg D_{U,S}, \tag{7}$$

is assumed to be isotropic so that the anisotropic properties obtained for the ground state completely reflect the action of antisymmetric exchange. In the actual case, tetragonal anisotropy must also be present in the Heisenberg exchange anisotropy, but its magnitude for the $S$ ions of the $3d$ group is much smaller as a rule than the antisymmetric exchange (if the latter is permitted by the symmetry):

$$\begin{aligned} H = H_J + H_U + H_S &= J\sum_{ij}(\mathbf{S}_{ij}\cdot\mathbf{S}_{i+1,j} + \mathbf{S}_{ij}\cdot\mathbf{S}_{i,j+1}) \\ &+ D_U\sum_{ij}(\mathbf{e}_x\cdot[\mathbf{S}_{ij}\times\mathbf{S}_{i+1,j}] \pm \mathbf{e}_y\cdot[\mathbf{S}_{ij}\times\mathbf{S}_{i,j+1}]) \\ &+ D_S\sum_{ij}(-1)^{i+j}\mathbf{e}_z\cdot(\mathbf{S}_{ij}\times\mathbf{S}_{i+1,j} + \mathbf{S}_{ij}\times\mathbf{S}_{i,j+1}). \end{aligned} \tag{8}$$

It will be shown below that a collinear structure is formed along the tetragonal axis $z$; consequently, the third index of summation over sites is omitted only to simplify the form of Hamiltonian (8), but the magnet remains three-dimensional.

The invariants of the expansion of the thermodynamic potential (Ginzburg–Landau functional) can be obtained for $T = 0$ by passing from discrete Hamiltonian (8) to a continual distribution of the magnetic energy density, which was used earlier in analysis of incommensurate magnetic structures [16]. Local ferro- and antiferromagnetism vectors $\mathbf{m}(\mathbf{r})$ and $\mathbf{l}(\mathbf{r})$, as well as $\mathbf{S}_{ij}$ (4), will indicate the average values of operators. In the continual transition, it is convenient to introduce for an antiferromagnet two magnetic sublattices $\alpha$ and $\beta$ with a smooth (infinitely differentiable) dependence of magnetization on $\mathbf{r}$. This allows us to omit higher-order derivatives in the expansion of the moments of sublattices in the description of a long-periodic incommensurate magnetic structure:

$$\mathbf{S}_{\mathbf{r}+\mathbf{a}} = \mathbf{S}_\mathbf{r} + \mathbf{a}\frac{\partial\mathbf{S}_\mathbf{r}}{\partial\mathbf{r}} + \frac{a^2}{2}\frac{\partial^2\mathbf{S}_\mathbf{r}}{\partial r^2} + \ldots. \tag{9}$$

Here, $\mathbf{a}$ are the vectors connecting the nearest magnetic neighbors (see Fig. 1).

The magnetic energy density for the UDM exchange at a site of each sublattice can be written in the form

$$\begin{aligned} \varepsilon_U &= \frac{D_U}{2}\sum_\mathbf{a}\{\mathbf{e}_x\cdot(\mathbf{S}_r\times\mathbf{S}_{r+a} - \mathbf{S}_r\times\mathbf{S}_{r-a}) \\ &\pm (x\longrightarrow y)\} \approx \frac{Z}{2}D_U a\left\{\mathbf{e}_x\cdot\left[\mathbf{S}_r\times\frac{\partial\mathbf{S}_r}{\partial x}\right] \pm (x\longrightarrow y)\right\} \end{aligned} \tag{10}$$

or in the form symmetrized in both sublattices:

$$\varepsilon_U \approx \frac{Z}{4}D_U a\left\{\mathbf{e}_x\cdot\left(\mathbf{S}_\alpha\times\frac{\partial\mathbf{S}_\beta}{\partial x} + \mathbf{S}_\beta\times\frac{\partial\mathbf{S}_\alpha}{\partial x}\right) \pm (x\longrightarrow y)\right\}.$$

Here and below, index $\mathbf{r}$ on the spin variables is omitted to simplify notation. Passage to ferro- and antiferromagnetism vectors $\mathbf{m} = \mathbf{S}_\alpha + \mathbf{S}_\beta$ and $\mathbf{l} = \mathbf{S}_\alpha - \mathbf{S}_\beta$ completes the derivation of the UDM exchange invariant in standard form:

$$\begin{aligned} I_U &= m_y\frac{\partial m_z}{\partial x} - m_z\frac{\partial m_y}{\partial x} - l_y\frac{\partial l_z}{\partial x} + l_z\frac{\partial l_y}{\partial x} \\ &\pm\left(m_x\frac{\partial m_z}{\partial y} - m_z\frac{\partial m_x}{\partial y} - l_x\frac{\partial l_z}{\partial y} + l_z\frac{\partial l_x}{\partial y}\right). \end{aligned} \tag{11}$$

This invariant is a particular (tetragonal) case of the Lifshits generalized invariant [1]. The SDM exchange components give the sum of vector products instead of their difference (10); as a result, their contribution to the magnetic energy is given by the spatially homogeneous invariant

$$I_S = m_x l_y - m_y l_x. \tag{12}$$

For finite temperatures, the expansion of the free energy contains terms with higher powers of the order parameter; in addition, the coefficients of quadratic invariants are themselves functions of temperature. However, the coefficients of inhomogeneous and homogeneous invariants are proportional to parameters $D_U$ and $D_S$ as before. In the limit $T \longrightarrow 0$, in which the free energy entropy term vanishes, the coefficients of these invariants must tend to $ZaD_U/4$ and $ZD_S/2$, respectively, defining together with (11) and (12) the energy of the volume containing only one spin for each interaction.

## 3. MEAN FIELD APPROXIMATION

We will determine the equilibrium orientation of sublattice spins in the mean field approximation. If all spins are in identical (in magnitude) mean fields (one-subsystem case), the minimization of free energy can be reduced to determining the mean field extremum [16]. For the spatially nonuniform magnetization distribution over directions we are interested in, this boils down to solving differential equations. The number of

such equations is determined by the number of independent variables over which the energy is varied. In determining the ground state ($T = 0$), the absolute values of spins can be fixed, assuming that the spins are equal to the saturation moment. If we confine our analysis to the form of Hamiltonian (8) quadratic in spin variables, the equivalence condition for all spins relative to the mean field makes it possible to avoid the self-consistency procedure for the equilibrium amplitude (absolute value) of the spin for $T > 0$ as well, since all fields from paired interactions are proportional to the same value of the average spin. Thus, we are left with four equations connecting the four functions defining the distribution of orientations of the moments of two sublattices. Instead of the variation of the longitudinal field, we can use an equivalent procedure at making the transverse fields vanish at the spins of each sublattice. The order of derivatives in the expansion of the terms of Hamiltonian (8) is limited by condition (7); for antisymmetric exchange, we retain only the first derivatives with respect to coordinates.

In the mean field approximation, we can write antisymmetric Hamiltonians (8) in the form

$$H_{U,S} = \sum_{ij} \mathbf{h}_{ij}^{U,S} \cdot \mathbf{S}_{ij}, \tag{13}$$

where

$$\begin{aligned} \mathbf{h}_{ij}^{U} &= \frac{D_U}{2}\{\mathbf{e}_x \times (\mathbf{S}_{i-1,j} - \mathbf{S}_{i+1,j}) \\ &\pm \mathbf{e}_y \times (\mathbf{S}_{i,j-1} - \mathbf{S}_{i,j+1})\}, \\ \mathbf{h}_{ij}^{S} &= (-1)^{i+j+1}\frac{D_S}{2}\{\mathbf{e}_z \times (\mathbf{S}_{i-1,j} \\ &+ \mathbf{S}_{i+1,j} + \mathbf{S}_{i,j-1} + \mathbf{S}_{i,j+1})\}. \end{aligned} \tag{14}$$

In the continual representation, for sublattice α with even sum $i + j$, we have

$$\begin{aligned} h_\alpha^{U,x} &= \mp\frac{Z}{2}D_U a\frac{\partial S_\beta^z}{\partial y}, \quad h_\alpha^{S,x} = \frac{Z}{2}D_S S_\beta^y, \\ h_\alpha^{U,y} &= \frac{Z}{2}D_U a\frac{\partial S_\beta^z}{\partial x}, \quad h_\alpha^{S,y} = -\frac{Z}{2}D_S S_\beta^x, \\ h_\alpha^{U,z} &= -\frac{Z}{2}D_U a\left(\frac{\partial S_\beta^y}{\partial x} \mp \frac{\partial S_\beta^x}{\partial y}\right), \quad h_\alpha^{s,z} = 0. \end{aligned} \tag{15}$$

Fields $h^U$ are even, while fields $h^S$ are odd relative to transpositions of indices of sublattices α and β. The absence of derivatives with respect to coordinate $z$ directed along tetragonal axis $c$ indicates that the antisymmetric exchange in our case does not lead to a spatially nonuniform spin density distribution in this direction.

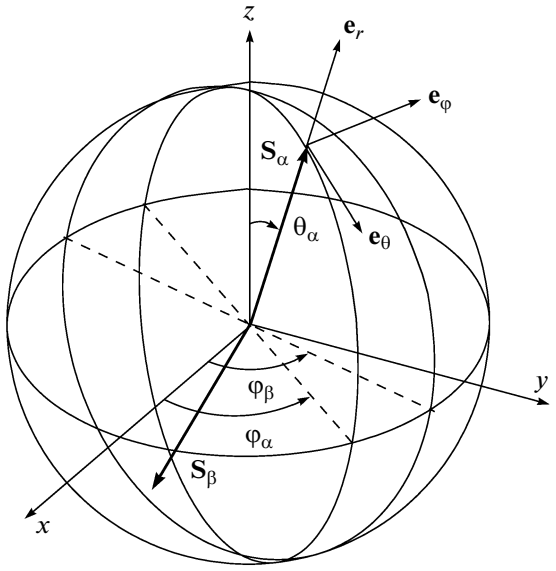


**Fig. 2.** Polar system of coordinates. Unit vectors are shown only for one quasi-antiferromagnetic sublattice α.

It is convenient to carry out the separation into transverse and longitudinal components in the polar system of coordinates $\{r, \theta, \varphi\}$ (Fig. 2):

$$\begin{aligned} S_{\alpha,\beta}^x &= S\sin\theta_{\alpha,\beta}\cos\varphi_{\alpha,\beta}, \\ S_{\alpha,\beta}^y &= S\sin\theta_{\alpha,\beta}\sin\varphi_{\alpha,\beta}, \quad S_{\alpha,\beta}^z = S\cos\theta_{\alpha,\beta}, \\ h_{\alpha,\beta}^{\varphi} &= -h_{\alpha,\beta}^x\sin\varphi_{\alpha,\beta} + h_{\alpha,\beta}^y\cos\varphi_{\alpha,\beta}, \\ h_{\alpha,\beta}^{\theta} &= h_{\alpha,\beta}^x\cos\theta_{\alpha,\beta}\cos\varphi_{\alpha,\beta} \\ &+ h_{\alpha,\beta}^y\cos\theta_{\alpha,\beta}\sin\varphi_{\alpha,\beta} - h_{\alpha,\beta}^z\sin\theta_{\alpha,\beta}, \\ h_{\alpha,\beta}^{r} &= h_{\alpha,\beta}^x\sin\theta_{\alpha,\beta}\cos\varphi_{\alpha,\beta} \\ &+ h_{\alpha,\beta}^y\sin\theta_{\alpha,\beta}\sin\varphi_{\alpha,\beta} + h_{\alpha,\beta}^z\cos\theta_{\alpha,\beta}. \end{aligned} \tag{16}$$

For sublattice α, we have

$$\begin{aligned} h_\alpha^{U,\varphi} &= -\frac{Z}{2}D_U a S\sin\theta_\beta \\ &\times\left(\pm\sin\varphi_\alpha\frac{\partial\theta_\beta}{\partial y} + \cos\varphi_\alpha\frac{\partial\theta_\beta}{\partial x}\right), \end{aligned} \tag{17}$$

$$h_\alpha^{U,\theta} = \frac{Z}{2} D_U a S$$
$$\times \left\{ \cos\theta_\alpha \sin\theta_\beta \left( \pm \cos\varphi_\alpha \frac{\partial \theta_\beta}{\partial y} - \sin\varphi_\alpha \frac{\partial \theta_\beta}{\partial x} \right) \right.$$
$$+ \sin\theta_\alpha \cos\theta_\beta \left( \mp \cos\varphi_\beta \frac{\partial \theta_\beta}{\partial y} + \sin\varphi_\beta \frac{\partial \theta_\beta}{\partial x} \right) \quad (18)$$
$$\left. + \sin\theta_\alpha \sin\theta_\beta \left( \cos\varphi_\beta \frac{\partial \varphi_\beta}{\partial x} \pm \sin\varphi_\beta \frac{\partial \varphi_\beta}{\partial y} \right) \right\},$$

$$h_\alpha^{U,r} = \frac{Z}{2} D_U a S$$
$$\times \left\{ \sin\theta_\alpha \sin\theta_\beta \left( \pm \cos\varphi_\alpha \frac{\partial \theta_\beta}{\partial y} - \sin\varphi_\alpha \frac{\partial \theta_\beta}{\partial x} \right) \right.$$
$$- \cos\theta_\alpha \cos\theta_\beta \left( \mp \cos\varphi_\beta \frac{\partial \theta_\beta}{\partial y} + \sin\varphi_\beta \frac{\partial \theta_\beta}{\partial x} \right) \quad (19)$$
$$\left. - \cos\theta_\alpha \sin\theta_\beta \left( \cos\varphi_\beta \frac{\partial \varphi_\beta}{\partial x} \pm \sin\varphi_\beta \frac{\partial \varphi_\beta}{\partial y} \right) \right\},$$

$$h_\alpha^{S,\varphi} = -\frac{Z}{2} D_S S \sin\theta_\beta \cos(\varphi_\alpha - \varphi_\beta), \quad (20)$$

$$h_\alpha^{S,\theta} = \frac{Z}{2} D_S S \cos\theta_\alpha \sin\theta_\beta \sin(\varphi_\beta - \varphi_\alpha), \quad (21)$$

$$h_\alpha^{S,r} = \frac{Z}{2} D_S S \sin\theta_\alpha \sin\theta_\beta \sin(\varphi_\beta - \varphi_\alpha). \quad (22)$$

The field components of the Heisenberg exchange with the nearest neighbors can be written (correct to the second derivatives inclusively) in the form

$$h_\alpha^{J,\varphi} = \frac{Z}{2} JS$$
$$\times \{ \sin(\varphi_\beta - \varphi_\alpha)((1 - d_1 - d_1')\sin\theta_\beta + d_2 \cos\theta_\beta)$$
$$+ \cos(\varphi_\beta - \varphi_\alpha)(d_2' \sin\theta_\beta + 2d_3 \cos\theta_\beta) \}, \quad (23)$$

$$h_\alpha^{J,\theta} = \frac{Z}{2} JS$$
$$\times \{ \cos\theta_\alpha [\cos(\varphi_\beta - \varphi_\alpha)((1 - d_1 - d_1')\sin\theta_\beta$$
$$+ d_2 \cos\theta_\beta)$$
$$- \sin(\varphi_\beta - \varphi_\alpha)(d_2' \sin\theta_\beta + 2d_3 \cos\theta_\beta)]$$
$$- \sin\theta_\alpha ((1 - d_1)\cos\theta_\beta + d_2 \sin\theta_\beta) \}, \quad (24)$$

$$h_\alpha^{J,r} = \frac{Z}{2} JS$$
$$\times \{ \sin\theta_\alpha [\cos(\varphi_\beta - \varphi_\alpha)((1 - d_1 - d_1')\sin\theta_\beta$$
$$+ d_2 \cos\theta_\beta)$$
$$- \sin(\varphi_\beta - \varphi_\alpha)(d_2' \sin\theta_\beta + 2d_3 \cos\theta_\beta)]$$
$$+ \cos\theta_\alpha ((1 - d_1)\cos\theta_\beta - d_2 \sin\theta_\beta) \}, \quad (25)$$

where

$$d_1 = a^2 \left[ \left( \frac{\partial \theta_\beta}{\partial x} \right)^2 + \left( \frac{\partial \theta_\beta}{\partial y} \right)^2 \right],$$
$$d_1' = a^2 \left[ \left( \frac{\partial \varphi_\beta}{\partial x} \right)^2 + \left( \frac{\partial \varphi_\beta}{\partial y} \right)^2 \right],$$
$$d_2 = a^2 \left( \frac{\partial^2 \theta_\beta}{\partial x^2} + \frac{\partial^2 \theta_\beta}{\partial y^2} \right), \quad (26)$$
$$d_2' = a^2 \left( \frac{\partial^2 \varphi_\beta}{\partial x^2} + \frac{\partial^2 \varphi_\beta}{\partial y^2} \right),$$
$$d_3 = a^2 \left( \frac{\partial \theta_\beta}{\partial x} \frac{\partial \varphi_\beta}{\partial x} + \frac{\partial \theta_\beta}{\partial y} \frac{\partial \varphi_\beta}{\partial y} \right).$$

Writing the field components in the polar system of coordinates, we can take into account the following peculiarity of this system. The single-valuedness condition for the description necessitates that the domain of one of the angles (polar $\theta$ or azimuthal $\varphi$) be confined to interval $\{0, \pi\}$. In the description of spatial variation of angle $\theta$ in standard form, this leads to a stepwise jump in the values of angles and to sign reversal in the rate of variation of angle $\theta$ upon crossing the boundary of interval $\theta = \pi$. Since it is the polar angle that mainly experiences a variation in space (see below), we impose a constrain on azimuthal angles $0 \le \varphi_{a,\beta} < \pi$. These angles will define the position of the vertical planes in which angles $\theta_{\alpha,\beta}$ change (polarization planes) and not the angles proper (see Fig. 2). The azimuthal angles of spin orientation of quasi-antiferromagnetic sublattices $\alpha$ and $\beta$ coincide with these angles for $0 \le \theta_{\alpha,\beta} < \pi$ and differ from these angles for $\pi \le \theta_{\alpha,\beta} < 2\pi$. As a result, expressions (17), (20), and (23) for fields $h^\varphi$ of the sublattices acquire an additional factor $(-1)$ if the corresponding angle $\theta$ becomes larger than $\pi$. Therefore, with allowance for the change in sign of functions $\theta_{\alpha,\beta}$, the signs of the fields for each sublattice remain unchanged and display no singularities upon a change in polar angles $\theta_{\alpha,\beta}$.

It is convenient to classify possible magnetic structures according to the form of the functional dependence of angular functions on spatial variables. Homogeneous structures in the ground state have no

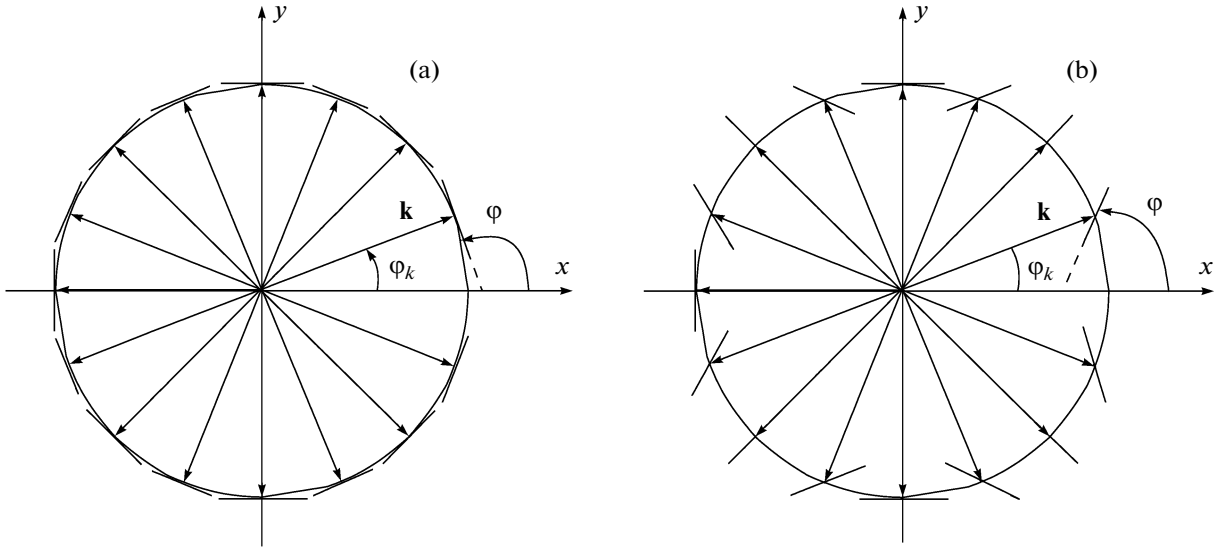


**Fig. 3.** Mutual orientation of vector **k** of the incommensurate magnetic structure and the polarization plane in the cases with (a) simple and (b) inversion tetragonal axes.

derivatives with respect to coordinates (all $d_i \equiv 0$ in formulas (21)). Structures of a simple helicoid type with a linear dependence of angles on the coordinates of the tetragonal plane contain only the first derivatives of angles in formula (21) ($d_{i>1} \equiv 0$). The emergence of second-order derivatives indicates that an incommensurate magnetic structure has a more complex coordinate dependence of the spin orientation (nonlinear modulation of the antiferromagnetic state).

## 4. PARTICULAR CASES

Let us consider particular cases, in which only one type of antisymmetric exchange exists.

1. $D_S \neq 0$, $D_U = 0$. From expression $h^\varphi = h^{J,\varphi} + h^{S,\varphi} = 0$, we obtain a homogeneous solution, viz., weakly ferromagnetic canting of sublattices:

$$\tan(\varphi_\alpha - \varphi_\beta) = -\frac{D_S}{J}. \quad (27)$$

2. $D_S = 0$, $D_U \neq 0$. The ground state with the maximum value of $h^r$ corresponds to the antiparallel orientation of the moments of the sublattices in the vertical plane, passing through the tetragonal axis,

$$\varphi = \varphi_\alpha = \varphi_\beta, \quad \theta_\alpha - \theta_\beta = \pi,$$

with a linear modulation of the initial antiferromagnetic structure,

$$\frac{\partial \theta_{\alpha,\beta}}{\partial \mathbf{r}} = \mathbf{k} - \text{const}.$$

In this case, $h^{J,\varphi} = 0$ and conditions $h^\varphi = h^{U,\varphi} = 0$ gives a simple relation between the modulations plane φ of the helicoid and the direction of vector **k** of the incommensurate magnetic structure in the plane:

$$\frac{\partial \theta_{\alpha,\beta}}{\partial x}\cos\varphi \pm \frac{\partial \theta_{\alpha,\beta}}{\partial y}\sin\varphi = k_x\cos\varphi \pm k_y\sin\varphi = k\cos(\varphi \pm \varphi_k) = 0, \quad (28)$$

$$\varphi \pm \varphi_k = \frac{\pi}{2}, \quad k = \sqrt{k_x^2 + k_y^2},$$

where $\varphi_\mathbf{k}$ is the angle between the direction of vector **k** and the tetragonal $x$ axis. The absolute value of the helicoid vector can easily be obtained from the equation for the longitudinal field extremum:

$$\frac{\partial h^r}{\partial k} = 0, \quad k = k_0 = -\frac{D_U}{2aJ}. \quad (29)$$

The value of the vector is independent of the orientation of the helicoid plane (and, accordingly, the direction of the vector)—the ground state is degenerate in angle φ. Figure 3 shows the mutual orientation of the polarization plane and the wavevector of the incommensurate magnetic structure for various tetragonal axes. In the case of a simple axis, the polarization plane and the vector of propagation of the incommensurate magnetic structure are always orthogonal; in the case of an inversion axis, the rotation of the vector of the incommensurate structure leads to the rotation of the polarization plane in the opposite direction.

## 5. GENERAL CASE $D_S \neq 0$, $D_U \neq 0$

Substitution of variables

$$\theta_{\alpha,\beta} = \theta^0_{\alpha,\beta} + \mathbf{k}_{\alpha,\beta} \cdot \mathbf{r}$$

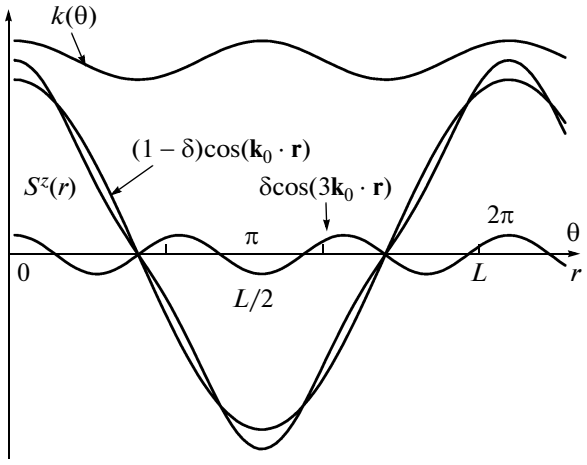


**Fig. 4.** Nonlinear modulation of the helicoid by the SDM exchange. The rate $k(\theta)$ of variation of the polar angle and the projection of the magnetic moment onto the tetragonal axis as functions of the polar angle and coordinate, respectively. The second harmonic in the rate leads to the third harmonic in the projection of the magnetic moment.

makes it possible to single out the main linear part of the functional dependence of angles θ on tetragonal coordinates. The remaining nonlinear part of this dependence is transferred to new variable $\mathbf{k}_{\alpha,\beta}$. For $D_S = 0$, the variation of longitudinal field $h^r$ in this new variable gives its value that is independent of the angles; i.e., the rate of variation of θ remains unchanged, which justifies the use of the linear approximation $d_{i>1} \equiv 0$ (21). If both types of antisymmetric exchange are simultaneously present in the tetragonal magnet, we can use this approximation to find the qualitative difference from a simple helicoid. The algorithm of solution remains unchanged: we determine the relation between angles $\varphi_\alpha$ and $\varphi_\beta$ defining the orientation of the polarization planes of quasi-antiferromagnetic sublattices, discarding second-order derivatives in exchange field $h^J$ and setting $\partial\varphi/\partial r = 0$; then we determine the extremum of the longitudinal field in $k$. If the value of $k$ corresponding to the longitudinal field extremum contains the dependence of angular variables, the form of this dependence defines the type of the nonlinearity. The latter can be taken into account as the corresponding correction to the form of the solution. Naturally, the amplitudes of all next corrections must contain higher powers of smallness parameter $\varepsilon = D_{U,S}/J$.

The first relation from four constraints defining two pairs of angular variables of the sublattices obviously remains unchanged for the linear approximation:

$$\mathbf{k}_\alpha = \mathbf{k}_\beta = \mathbf{k}. \tag{30}$$

This relation is a consequence of the general relation between the angles $\theta_\alpha - \theta_\beta = \pi$, which follows from the symmetry of the problem in angle θ (the existence of particular solution $\theta_\alpha = 0$, $\theta_\beta = \pi$ and invariance of fields $h^\theta$ and $h^r$ (13), (14), (16), (17), (19), and (20) relative to the shift $\theta \longrightarrow \theta + \pi$). The following two equations for angular variables $h^\varphi_{\alpha,\beta} = 0$ fix the cant of the polarization planes and their relation with the direction of **k**:

$$J(1 - a^2k^2)\sin R + D_S\cos R - D_U ak\sin\frac{R}{2} = 0, \tag{31}$$

$$\cos\left(\varphi_k + \frac{\varphi_\alpha + \varphi_\beta}{2}\right) = 0, \tag{32}$$

where $R = \varphi_\alpha - \varphi_\beta$. The helicoid becomes a double helicoid (each quasi-antiferromagnetic sublattice has its own polarization plane). The fourth equation (the extremum of $h_r$) gives

$$\begin{aligned} k &= -\frac{D_U}{2aJ}\cos\frac{R}{2}\left(1 + 2\sin^2\frac{R}{2}\sin\theta_\alpha\sin\theta_\beta\right) \\ &= -\frac{D_U}{2aJ}\cos\frac{R}{2}\left(1 - 2\sin^2\frac{R}{2}\sin^2\theta_{\alpha,\beta}\right) \\ &= -\frac{D_U}{2aJ}\cos\frac{R}{2}\left(\cos^2\frac{R}{2} + \sin^2\frac{R}{2}\cos 2\theta_{\alpha,\beta}\right). \end{aligned} \tag{33}$$

It can be seen that in the vicinity of the tetragonal plane ($\theta \longrightarrow \pi/2$), the rate of variation of angle θ decreases, i.e., the SDM exchange lowering the energy of the system due to canting of the components transverse to the tetragonal axis ensures the total easy-plane anisotropy of the magnet. The absence of a dependence of vector **k** on the average azimuthal angle $(\varphi_\alpha + \varphi_\beta)/2$ indicates that the ground state remains degenerate relative to the orientation of the modulation vector in the plane.

## 6. RESULTS AND DISCUSSION

The emergence of the dependence of the rate of variation of polar angle θ on the doubled value of this angle (second harmonic (28)) even in the first linear approximation leads to the emergence of odd harmonics in the momentum representation of spin variables

$$S = \sum_n C_{2n+1}\exp i(2n+1)\mathbf{k}_0\cdot\mathbf{r},$$

$$k_0 = -\frac{D_U}{2aJ\cos R/2}.$$

Figure 4 shows this relation qualitatively, in which

$$L = \frac{4\pi Ja}{D_U\cos R/2}$$

is the wavelength of the modulated helicoid. Axial anisotropy of symmetric exchange leads to the same

result. As a result of the action of the total axial anisotropy, the inelastic neutron scattering spectrum contains odd harmonics of the principal vector of the incommensurate structure. Analysis of the effect of anisotropy and determination of the relative amplitude of higher (e.g., third) harmonics is beyond the scope of this article. However, the linear approximation shows that the next correction (e.g., for axial component $S^z$) can be sought in the form

$$S^z = S((1-\delta)\cos\mathbf{k}_0\cdot\mathbf{r} + \delta\cos 3\mathbf{k}_0\cdot\mathbf{r}),$$

where $\delta$ is the smallness parameter in which successive approximations can be carried out.

Two more qualitative results of the mutual effect of different types of antisymmetric exchange are worth noting.

(i) It follows from relation (31) that the emergence of a dependence of the rate $k$ of variation of angle $\theta$ on this angle leads to the emergence of a dependence of the angle between the polarization planes of sublattices $R$ on angle $\theta$ as well. Consequently, we also have $\partial\varphi/\partial r \neq 0$. The polarization planes of the sublattices are transformed into curvilinear surfaces (Fig. 5).

(ii) Local weakly ferromagnetic moment **M** emerging as a result of canting of neighboring spins by the SDM exchange changes its magnitude and sign in the interval $\{-M_{max}, M_{max}\}$ upon a variation in angle $\theta$, remaining in the tetragonal plane. As a consequence, the total weakly ferromagnetic moment in the system in zero magnetic field is zero.

An example of a tetragonal antiferromagnet with a helicoidal incommensurate magnetic structure is $Ba_2CuGe_2O_7$ with space group $P\bar{4}2_1m$ [13, 17–20]. The generating symmetry element for this antiferromagnet is the inversion tetragonal axis, while the limiting element is the reflection symmetry plane passing between interacting spins. The plane singles out another antisymmetric combination of second-order spin components. As a result, the modulation vector lies in the tetragonal plane of the crystal and simultaneously in the vertical polarization plane of the helicoid.

Another tetragonal antiferromagnet with the crystal structure corresponding to the necessary symmetry requirements is copper metaborate $CuB_2O_4$ with space group $I\bar{4}2d$. Two nonequivalent positions of $Cu^{2+}$ ions in this substance form two magnetic subsystems, which strongly differ from one another in the distribution and strength of exchange bonds in the subsystems [21–25]. Strong antiferromagnetic exchange between $Cu^{2+}$ ions in the 4*b* crystallographic positions leads to the emergence of a spontaneous moment in this subsystem at $T_N = 20$ K. As the temperature decreases, the moment rapidly decreases to a value close to saturation, indicating the classical "3D" nature of magnetic ordering. A magnetic moment in the second subsystem of ions in the 8*d* positions emerges at $T \approx 11$ K

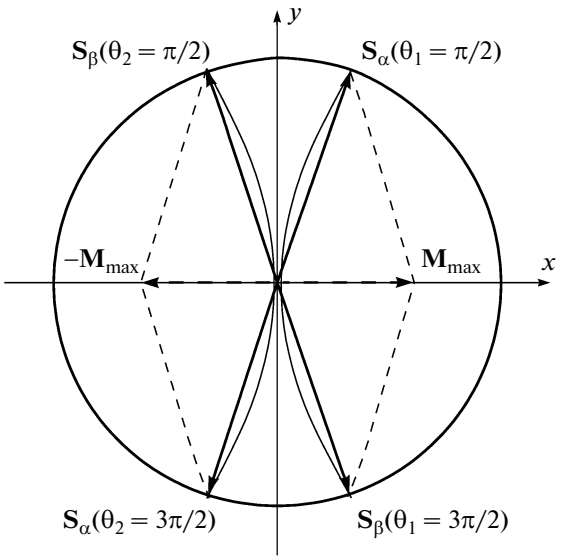


**Fig. 5.** Projection of the polarization surfaces of the sublattices and their moments for two values of angles $\theta_\alpha = \theta_\beta - \pi = \pi/2$ and $3\pi/2$ onto the tetragonal plane.

according to neutron diffraction data. A decrease in temperature to $T \approx 2$ K leads to an increase in the magnetic moment at the sites of this subsystem only to $m \sim 0.5\ \mu_B$, indicating a weak and probably quasi-one-dimensional exchange in this subsystem [26]. At $T < 9.5$ K, an incommensurate magnetic structure with a modulation vector directed along the tetragonal axis is formed in $CuB_2O_4$ due to removal of frustration in the intersubsystem symmetrical exchange [16, 27]. It was shown [28] that the high-temperature phase existing at $9.5\ \text{K} < T < 20.0$ K in zero external field is modulated. An external magnetic field destroys this phase, transforming the system into the weakly ferromagnetic state [21, 29–33]. A small critical field of the high-temperature incommensurate magnetic structure with its weak temperature dependence [29] indicates that a weak interaction in this subsystem is responsible for the emergence of this structure simultaneously with the emergence of a magnetic order in the "strong" subsystem. The symmetry of the 4*b* subsystem [15] ensures the coexistence of component $D_S$ along the tetragonal axis and the required distribution of uniform antisymmetric exchange $D_U$ in the tetragonal plane. The presence of satellite peaks in the inelastic neutron scattering spectrum [23] and stage-by-stage suppression of incommensurability by the field [34] at $T < 9.5$ K indicate the mutual effect (superposition) of two different incommensurate magnetic structures in the transition region at $T \approx 9.5$ K and at the phase boundary of the low-temperature incommensurate magnetic structure. All these facts lead to the conclusion that two incommensurate magnetic structures of different types and mechanismsof formation exist in $CuB_2O_4$. The high-temperature incommensurate

magnetic structure at $T > 9.5$ K is formed by the 2D distribution of the antisymmetric uniform Dzyaloshinski–Moriya exchange in the presence of the weakly ferromagnetic component of this exchange. Its modulation vector lies in the tetragonal plane, and the polarization planes of the sublattices pass through the tetragonal axis.

The importance of determining the types of all magnetic phases in $CuB_2O_4$ (both commensurate and incommensurate) is enhanced primarily due to strong magnetoelectric effects detected in this material in recent years [30–33]. Analysis of the spatial distribution of anisotropic magnetic interactions is an indispensable part of the description of physical properties of multiferroics since the magnetoelectric effects directly depend on the direction of magnetization in homogeneous phases, as well as on the orientation of the modulation vector and the polarization plane in an incommensurate magnetic structure.

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