Microscopic description of twisted multiferroic Cu$_2$OSeO$_3$

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In the frame of the Heisenberg model, the twist and canting of magnetic moments are studied for multiferroic Cu$_2$OSeO$_3$. Two kinds of local spin cantings are distinguished: the first one, induced by the gradients of the magnetization, and the second one managed by the Dzyaloshinskii–Moriya (DM) interactions. The macroscopic parameters $J$ and $D$ in the free energy and the helix wave number $k$ are expressed through the interatomic exchange constants $J_{ij}$ and the DM vectors $D_{ij}$ of individual bonds. These expressions gain the simplest form with the use of fictitious (“exchange”) coordinates of the magnetic atoms, which also describe the cantings due to the magnetization gradients. In the first approximation, the spin cantings due to the DM interactions is given by a vector product $[\rho \times \mu]$, where $\mu$ is the mean spin of the equivalent atoms, and $\rho$ are tilt vectors. The tilt vectors and the exchange coordinates of the magnetic atoms in equivalent positions are connected by corresponding transformations of the crystal point group. It is found that the macroscopic parameters $J$ and $D$ are very sensitive to the values of interatomic parameters $J_{ij}$ and $D_{ij}$ up to the second magnetic shell. It is predicted that the magnetic helix should be left-handed ($k = -0.074$) for the enantiomorph with the right-handed atomic structure.

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I. INTRODUCTION

Most of the known twisted magnetics possess symmetry lower than cubic. Their strong anisotropy orients magnetic helices in selected crystallographic directions. As opposed to them, in cubic magnetics without center of inversion the binding between magnetic and crystal structures can be so subtle, that the helix can be easily reoriented along arbitrary direction without essential change of its pitch and energy. Nevertheless, in microscopic level this seeming freedom is achieved by correlated tilts of discrete magnetic moments tightly bound with atoms in the crystal. Besides, the easier seems to be the phenomenology of the isotropic system as compared with anisotropic one, the more complex appears its description in terms of discrete spins, particularly when the magnetic helix is oriented by the field in some arbitrary direction. As it has been shown recently for the helimagnetics with $B20$ structure (MnSi-type), this freedom of the helix rotations is achieved by extra tilts (or canting) of the local magnetic moments, which always accompany global spiralling, but do not fit in the frame of the phenomenological theory. Moreover, the canting still remains, when the spiralling disappears in strong magnetic field. In the present work, a microscopical theory will be developed of the twisted cubic magnetics with weakly noncollinear spins. The latter means that the angle between neighboring spins is close to 0 or to $\pi$, i.e. both the spiralling and canting are small. It is just the case of the majority of itinerant magnetics with $B20$ structure (exemplified by MnSi) and twisted ferrimagnetic Cu$_2$OSeO$_3$.

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The magnetic properties of the cubic crystal Cu$_2$OSeO$_3$ are of great interest for several reasons. First of all, having the space group $P2_13$ without center of inversion, this crystal becomes a cubic helimagnet below the critical temperature of about 58 K. Moreover, it is a first cubic crystal out of the class of itinerant magnetics with $B20$ crystal structure [1–4], for which A-phase, associated with Skyrmion lattice [5, 6], has been recently observed [7–12]. Secondly, being an insulator, Cu$_2$OSeO$_3$ has magnetoelectric properties [12–18]. The interconnection between magnetization gradients and electric polarization makes this crystal potentially applicable for data storing devices and spintronics [13]. And thirdly, but not finally, the more complex than $B20$ structure (16 magnetic copper atoms in two nonequivalent positions in the unit cell) makes it an interesting object for studying the spin textures.

As a rule, when someone talks about the spin canting, some class of canted antiferromagnetics is implied [20], where small unidirectional slants of magnetic moments belonging to different sublattices result in appearing of weak magnetization. The phenomenon is called weak ferromagnetism, and it serves as direct evidence of the canting, because the tilt angles of individual spins are directly proportional to the observed spontaneous magnetization. The canting in ferromagnetics is less elaborated, because it is more difficult to observe a little decrease of strong magnetization, and, therefore, a refined experiment is needed, for example, on observation of forbidden Bragg reflections of antiferromagnetic origin [21].

Repeated small rotations (cantings) of the planes of magnetization can result in the global twist of the magnetic moment. Therefore, in chiral ferromagnetics, the twist and the canting are often mistaken for the same thing. Only recently, we suggested an adequate conception how to distinguish between canting and twist in ferromagnetics [22, 23]. It was shown that the canting should always appear when there are more than one magnetic atom in the unit cell. It leads to coexistence of several spin helices within the helimagnet, which differ by phases and rotational planes (Fig. 1). It was shown by example of cubic helimagnets with $B20$ structure that the canting makes contribution to the magnetic energy comparable with that of global spiralling. Therefore, it should be taken into account when calculating propagation number of spin helices from the first principles, i.e. starting from the Heisenberg microscopical model of ferromagnetism. Even more intriguing is the fact that, if the Dzyaloshinskii–Moriya (DM) vectors are perpendicular to the bonds between magnetic atoms (which is predicted by many quantum mechanical models), both twist and canting in MnSi-type crystals are determined by the same components of the DM vectors. The study of the Cu$_2$OSeO$_3$ crystal, with its structure different from $B20$, also can help us to distinguish between general behavioral regularities of twisted cubic ferromagnetics and particularities of the MnSi-type crystals.

In this paper, a general theory of the spin canting in cubic helimagnetics is developed, which unifies the cases of the MnSi-type and Cu$_2$OSeO$_3$ crystals. We confine only to the case of the twist induced by the DM interactions; the case of the twist owing to the frustrated exchange one is beyond the scope of our consideration. In Sec. II, the definition of canting is given. Then, the transition to continuous approximation is performed (Sec. III), and the density of the magnetic energy is calculated (Sec. IV). In Sec. V it is shown which simplifications can be made in the case, when, in the absence of the DM interaction, all the spins of the system remain collinear. We assert that two different kinds of the canting exist in the twisted magnetics. The first of them is connected with the gradients of the magnetization, and it arises even in the absence of the DM interaction. In order to take this kind of canting into account, it is convenient to use the method of the “exchange” coordinates (Sec. VI). Then, the simple expressions can be found for the macroscopic constants $J$ and $D$, and the helix propagation number $k = D/2J$ (Sec. VII). The second kind of canting is induced by the DM interactions. It can be described in terms of the “tilt” vectors, whose symmetry is
found to be closely connected with the symmetry of the crystal (Sec. V). The canting of this kind still remains even in the ferromagnetic state, when the helical spin structure is unwound by the external magnetic field. This residual canting can be measured, using neutron and x-ray diffractions (see discussion in Section IX). In Secs. VII, VIII, the theory is applied to the cases of MnSi and Cu$_2$OSeO$_3$.

II. BALANCE OF MAGNETIC MOMENTS AND CANTING

In the classical Heisenberg theory of magnetics the energy of a magnetic structure is written as

$$E = \sum_i \left\{ \frac{1}{2} \sum_j \left( -J_{ij}s_i \cdot s_j + D_{ij} \cdot [s_i \times s_j] \right) - \mu_B g_i H \cdot s_i \right\},$$

(1)

where $i$ enumerates all the magnetic atoms, the inner sum ($j$) is taken over close neighbors of $i$th atom, the coefficient $\frac{1}{2}$ is needed because each bond is included twice in the sum, $\mu_B g_i s_i$ is the magnetic moment of $i$th atom ($s_i$ is the direction of classical spin, $|s_i| = 1$), $J_{ij}$ are the isotropic exchange constants, whereas the antisymmetrical exchange is characterized by the Dzyaloshinskii–Moriya (DM) vectors $D_{ij}$ ($J_{ji} = J_{ij}$, $D_{ji} = -D_{ij}$), $H$ is an external magnetic field.

The equation of $i$th spin balance can be easily found copying out the part of Eq. (1) associated with the $i$th spin,

$$E_i = - \left\{ \sum_j (J_{ij}s_j + [D_{ij} \times s_j]) + \mu_B g_i H \right\} \cdot s_i \equiv -\varepsilon_i \cdot s_i,$$

(2)

(notice the absence of $1/2$ in comparison with Eq. (1)). It is evident that $E_i$ is minimal, when

$$s_i = \frac{\varepsilon_i}{|\varepsilon_i|},$$

(3)

Being of relativistic origin, the DM interaction is considerably weaker than the isotropic exchange,

$$|D_{ij}|/J_{ij} \ll 1.$$  

(4)

Therefore it is convenient to develop a perturbation theory with a small parameter of the order of $D/J$. In the case of twisted magnetics we can also neglect the influence of the external magnetic field on canting, supposing that

$$H \leq H_{c2} \sim \left( \frac{D}{J} \right)^2 \frac{J}{\mu_B}.$$  

(5)

where $H_{c2}$ is the field of “full unwinding” of the magnetic helix.

Neglecting the DM interaction and external magnetic field, Eq. (3) determines an equilibrium system of spins

$$\mathbf{\mu}_i \equiv s_i^{(0)} = \frac{\varepsilon_i^{(0)}}{|\varepsilon_i^{(0)}|},$$

(6)

with

$$\varepsilon_i^{(0)} = |\varepsilon_i^{(0)}|\mu_i = \sum_j J_{ij}\mu_j.$$  

(7)

The isotropy of the exchange interaction results in that both the structure energy and spin balance remain unchanged, if all the spins rotate as a unit by arbitrary angle relative to immovable atomic structure, or, which is
practically the same, if the structure rotates preserving initial spin directions. The latter understanding is important for cubic crystal structure, possessing equivalent atomic positions connecting to each other by rotational symmetry transformations. It is obvious that a spin balance can be achieved, when \( \mu_i \) are equal for all the equivalent positions (Fig. 2). Here we suppose that the twist is induced only by the DM interactions, and leave the possibility of the pure exchange spiralling outside the scope of our consideration.

The non-isotropic DM interaction breaks the symmetry of \( \mu_i \), because the DM vectors of equivalent bonds rotate with the bond directions. Therefore the spin \( s_i \) deviates from \( \mu_i \) by a small angle of order of \( D/J \). Taking into account that \( |\mu_i| = 1 \), this spin change, hereinafter referred to as “canting”, in the first approximation is perpendicular to \( \mu_i \). Introducing correction \( \varepsilon_i^{(1)} \) of first order on \( D/J \), we find

\[
s_i \approx \frac{\varepsilon_i^{(0)} + \varepsilon_i^{(1)} + \varepsilon_i^{(1)}}{|\varepsilon_i^{(0)} + \varepsilon_i^{(1)} + \varepsilon_i^{(1)}|} \approx \mu_i + \mu_i^{(1)} - \frac{|\mu_i^{(1)}|^2}{2} \mu_i,
\]

where \( \varepsilon_i^{(1)} \) and \( \varepsilon_i^{(1)} \) are the parallel and perpendicular to \( \mu_i \) components of \( \varepsilon_i^{(1)} \), correspondingly; \( \mu_i^{(1)} = \varepsilon_i^{(1)}/|\varepsilon_i^{(0)}| \) is the canting of \( i \)th spin in the first approximation, and

\[
- \frac{|\mu_i^{(1)}|^2}{2} \mu_i \equiv \mu_i^{(2)}
\]

is proportional to \( (D/J)^2 \) change of the spin along \( \mu_i \) (Fig. 3).

III. CONTINUOUS SPIN FUNCTIONS

In cubic crystal without center of inversion, a constant direction of magnetic moment density vector is energetically disadvantageous, and magnetic structure becomes twisted. Besides, the strength of the twist is characterized by the same ratio \( D/J \) as the canting does. If the magnetic moment density changes slowly along the crystal, a transition can be performed from discrete spins to smooth continuous spin functions \( \hat{s}_i(r) \), whose number coincides with the number of magnetic atoms in the crystal unit cell. Then, the magnetic energy can be rewritten in integral form [23],

\[
E = \int dr \sum_i \left\{ \frac{1}{2} \sum_j \left[ -J_{ij} \hat{s}_i \cdot \exp(\mathbf{b}_{ij} \cdot \nabla) \hat{s}_j + \mathbf{D}_{ij} \cdot [\hat{s}_i \times \exp(\mathbf{b}_{ij} \cdot \nabla) \hat{s}_j] - \mu_B g_i \mathbf{H} \cdot \hat{s}_i \right] \right\},
\]

where the sum \( (i) \) is now taken over all the magnetic atoms in the unit cell, and \( \mathbf{b}_{ij} \) is directed bond connecting \( i \)th and \( j \)th atoms, measured in cubic cell parameters \( a = 1 \); the values of the spin functions and their spatial derivatives in the integrand are taken at the same point.

Using Eq. 2, the vector \( \hat{\varepsilon}_i^{(1)}(r) \) can be written in continuous approximation,

\[
\hat{\varepsilon}_i^{(1)} = \sum_j \left( J_{ij} \hat{u}_j^{(1)} + J_{ij} (\mathbf{b}_{ij} \cdot \nabla) \hat{\mu}_j + [\mathbf{D}_{ij} \times \hat{\mu}_j] \right),
\]

where we neglect the magnetic field and replace \( \hat{u}_j^{(1)} \) and \( \hat{\mu}_j \) with continuous functions \( \hat{u}_j^{(1)}(r) \) and \( \hat{\mu}_j(r) \). Then,

\[
|\varepsilon_i^{(0)}| |\varepsilon_i^{(1)}| = \left\{ \sum_j \left[ J_{ij} \hat{u}_j^{(1)} + J_{ij} (\mathbf{b}_{ij} \cdot \nabla) \hat{\mu}_j + [\mathbf{D}_{ij} \times \hat{\mu}_j] \right] \right\}. 
\]
where the subscript "⊥" designates vector projection on the plane perpendicular to \( \hat{\mu}_i \). Because \( \hat{u}_j^{(1)} \perp \hat{\mu}_j \) and \( |\hat{\mu}_j| = 1 \), then each summand in the sum over \( j \) is a vector perpendicular to \( \hat{\mu}_j \). Eq. (12) shows that the canting is determined by both the DM interaction and spatial derivatives of \( \hat{\mu} \).

Let us average out Eq. (12) using the crystal symmetry. For any cubic point group

\[
\langle h_{ij} \rangle_{eq} = \langle D_{ij} \rangle_{eq} = 0,
\]

where index "eq" means averaging over equivalent bonds and positions. Thus, we obtain

\[
|\hat{\epsilon}_i^{(0)}|\langle \hat{u}_i^{(1)} \rangle_{eq} = \sum_j J_{ij} \langle \hat{u}_j^{(1)} \rangle_{eq,⊥}.
\]

It is evident from comparison with Eq. (7), that Eq. (14) is satisfied, when

\[
\langle \hat{u}_i^{(1)} \rangle_{eq} = [\varphi \times \hat{\mu}_i],
\]

with \([\varphi \times \hat{\mu}_i]\) being the change of spin \( \hat{\mu}_i \), induced by rotation of all the spins as a unit by a small angle \( \varphi \). As it is mentioned above, the rotation does not change the isotropic exchange energy and results in new functions \( \hat{\mu}_i \), also satisfying Eq. (7). Therefore it is possible to include \( \langle \hat{u}_i^{(1)} \rangle_{eq} \) into the definition of \( \hat{\mu}_i \) and assume without loss of generality that

\[
\langle \hat{u}_i^{(1)} \rangle_{eq} = 0.
\]

In this case, \( \hat{\mu}_i \) is the average spin over all the positions in the unit cell equivalent to the \( i \)th one.

**IV. ENERGY OF TWIST AND CANTING**

Let us write out the contributions to the energy (10) up to the second order terms on \( D/J \). The zeroth order energy is

\[
E^{(0)} = -\frac{1}{2} \sum_i \sum_j J_{ij} \hat{\mu}_i \cdot \hat{\mu}_j.
\]

The 1st order contribution

\[
E^{(1)} = \frac{1}{2} \sum_i \sum_j \left\{ J_{ij} \left( \hat{\mu}_i \cdot (b_{ij} \cdot \nabla) \hat{\mu}_j + \hat{u}_i^{(1)} \cdot \hat{\mu}_j + \hat{\mu}_i \cdot \hat{u}_j^{(1)} \right) + D_{ij} \cdot [\hat{\mu}_i \times \hat{\mu}_j] \right\}
\]

becomes zero in the equilibrium due to Eqs. (13), (16).

It is convenient to divide the 2nd order contribution into two parts: the first one, associated with the change of the magnetic moment \( \hat{\mu} \) only,

\[
E^{(2)}_{\hat{\mu}} = \frac{1}{2} \sum_i \sum_j \left\{ -\frac{1}{2} J_{ij} \hat{\mu}_i \cdot (b_{ij} \cdot \nabla)^2 \hat{\mu}_j + D_{ij} \cdot [\hat{\mu}_i \times (b_{ij} \cdot \nabla) \hat{\mu}_j] \right\},
\]

and the second one, depending on canting,

\[
E^{(2)}_u = \frac{1}{2} \sum_i \sum_j \left\{ -J_{ij} \left( \hat{u}_i^{(2)} \cdot \hat{\mu}_j + \hat{\mu}_i \cdot \hat{u}_j^{(2)} + \hat{u}_i^{(1)} \cdot (b_{ij} \cdot \nabla) \hat{\mu}_j - \hat{u}_j^{(1)} \cdot (b_{ij} \cdot \nabla) \hat{\mu}_i + \hat{u}_i^{(1)} \cdot \hat{u}_j^{(1)} \right) + D_{ij} \cdot \left( [\hat{u}_i^{(1)} \times \hat{\mu}_j] + [\hat{\mu}_i \times \hat{u}_j^{(1)}] \right) \right\}.
\]
The expression for $\mathcal{E}_\mu^{(2)}$ can be averaged over equivalent bonds, using equation

$$\langle a_\alpha b_\beta \rangle_{eq} = \frac{1}{3} \delta_{\alpha\beta} (a \cdot b)$$

(21)

for the vectors $a$ and $b$ transformed with the elements of a cubic group. Then

$$\mathcal{E}_\mu^{(2)} = \frac{1}{6} \sum_i \sum_j \left\{ \frac{1}{2} J_{ij} |b_{ij}|^2 \hat{\mu}_i \cdot \Delta \hat{\mu}_j + (D_{ij} \cdot b_{ij}) \hat{\mu}_i \cdot [\nabla \times \hat{\mu}_j] \right\}.$$  

(22)

Notice that all the summands corresponding to equivalent bonds are equal, and, therefore, we can calculate them only once, multiplying then by the bond multiplicities.

Changing summation order and using relations $b_{ji} = -b_{ij}$ and $D_{ji} = -D_{ij}$, Eq. (20) can be rewritten as

$$\mathcal{E}_u^{(2)} = \sum_i \sum_j \left\{ -J_{ij} \left( \hat{u}_i^{(2)} \cdot \hat{\mu}_j + \hat{u}_j^{(1)} \cdot (b_{ij} \cdot \nabla) \hat{\mu}_j + \frac{1}{2} \hat{u}_i^{(1)} \cdot \hat{u}_j^{(1)} \right) + D_{ij} \cdot [\hat{u}_i^{(1)} \times \hat{\mu}_j] \right\}.$$  

(23)

Using Eqs. (7), (9) and (12), the latter expression can be transformed to

$$\mathcal{E}_u^{(2)} = -\frac{1}{2} \sum_i |\hat{e}_i^{(0)}| |\hat{u}_i^{(1)}|^2 + \frac{1}{2} \sum_i \sum_j J_{ij} \hat{u}_i^{(1)} \cdot \hat{u}_j^{(1)}.$$  

(24)

V. THE CASE OF COLLINEAR MEAN SPINS

In frustrated magnetic structures, the spins $\hat{\mu}_i$ can be noncollinear. Besides, in a number of practically important cases, including ferro-, antiferro-, and ferrimagnetic orders, all $\hat{\mu}_i$ can be aligned along a line, and several considerable simplifications can be made. Indeed, assume that

$$\hat{\mu}_i = c_i \hat{\mu}, \quad c_i = \pm 1, \quad |\hat{\mu}| = 1.$$  

(25)

Here, $\hat{\mu}$ is the unit vector directed along the local magnetization. Then all $\hat{u}_i^{(1)}$ belong to the plane perpendicular to $\hat{\mu}$, and we can discard the index “⊥” in Eq. (12):

$$\sum_j J_{ij} c_j \hat{u}_i^{(1)} = \sum_j \left( J_{ij} \hat{u}_j^{(1)} + J_{ij} c_j (b_{ij} \cdot \nabla) \hat{\mu} + c_j (D_{ij} \times \hat{\mu}) \right),$$  

(26)

where is used that

$$|\hat{e}_i^{(0)}| = \sum_j J_{ij} c_j.$$  

(27)

Another simplification is the use of condition

$$\sum_j J_{ij} c_j b_{ij} = 0.$$  

(28)

The system (28) contains several independent vector equations (whose number coincides with the number of nonequivalent magnetic positions in the unit cell) and can be considered as a condition imposed on the atomic coordinates. Indeed, the energy (11) does not depend explicitly on the atomic coordinates, as opposed to Eq. (10), where they are included in vectors $b_{ij}$. Obviously, the coordinates should disappear after minimization of the energy, therefore we
can choose them arbitrarily, e.g. using Eq. (28). It is shown in Ref. [23], that the possibility of intentional choice of ideal atomic coordinates is connected with the ambiguity of transition from discrete spins to continuous density of magnetic moment. Notice that, because Eq. (28) contains only exchange interaction parameters, these fictitious coordinates depend only on $J_{ij}$. So, hereinafter we will refer to the positions as “exchange” ones. In Sec. IX the sense and properties of the exchange coordinates are discussed in details.

The atomic “shift” from real to fictitious positions results in disappearing of the part of the canting induced by derivatives of $\hat{\mu}$:

$$\sum_j J_{ij} c_j \left| \hat{u}_i^{(1)} \right| = \sum_j \left( J_{ij} \hat{u}_j^{(1)} + c_j [D_{ij} \times \hat{\mu}] \right).$$  \hspace{1cm} (29)

Now the canting can be found in form

$$\hat{u}_i^{(1)} = c_i [\rho_i \times \hat{\mu}],$$  \hspace{1cm} (30)

where “tilt” vectors $\rho_i$ possess the following properties. (i) The vector $\rho_i$ has the symmetry of the $i$th atomic position, e.g. the tilt vector of an atom in position 4a of the space group $P2_13$ is directed along 3-fold axis of symmetry passing through the atom. (ii) The tilt vectors in equivalent positions are connected to each other by the corresponding transformation of the point group.

Now the system (29) can be rewritten as

$$\sum_j J_{ij} c_j = \sum_j \left( J_{ij} c_j \rho_j + c_j D_{ij} \right).$$  \hspace{1cm} (31)

The system (31) contains as many independent equations as many nonequivalent magnetic atoms are in the unit cell.

**VI. PHENOMENOLOGICAL CONSTANTS**

The energy density expressions are also simplified in the case of collinear mean spins:

$$\mathcal{E}^{(0)} = -\frac{1}{2} \sum_i \sum_j J_{ij} c_i c_j,$$  \hspace{1cm} (32)

$$\mathcal{E}^{(2)} = \mathcal{E}^{(2)}_{\mu} = -\frac{1}{12} \left( \sum_i \sum_j J_{ij} c_i c_j |b_{ij}|^2 \right) \hat{\mu} \cdot \Delta \hat{\mu} - \frac{1}{6} \left( \sum_i \sum_j c_i c_j \hat{D}_{ij} \times \hat{b}_{ij} \right) \hat{\mu} \cdot [\nabla \times \hat{\mu}],$$  \hspace{1cm} (33)

Using equation

$$\langle [\rho_i \times \hat{\mu}] \cdot [\rho_j \times \hat{\mu}] \rangle_{eq} = \frac{2}{3} \rho_i \cdot \rho_j,$$  \hspace{1cm} (34)

we find

$$\mathcal{E}^{(2)}_u = -\frac{1}{3} \sum_i \left| \sum_j J_{ij} c_j \right| |\rho_i|^2 + \frac{1}{3} \sum_i \sum_j J_{ij} c_i c_j \rho_i \cdot \rho_j,$$  \hspace{1cm} (35)
or, using Eq. 31,

\[ \mathcal{E}_u^{(2)} = \frac{1}{3} \sum_i \sum_j c_i c_j D_{ij} \cdot \rho_i. \]  

Notice that in the latter expression as well as in Eq. 33 the summands corresponding to equivalent bonds are equal.

Because, as it is seen from Eq. 31, \( \rho_i \) are independent of spatial derivatives of \( \hat{\mu} \), the energy \( \mathcal{E}_u^{(2)} \) does not contain the derivatives as well. On the other hand, \( \mathcal{E}_\mu^{(2)} \) does not depend on \( \rho_i \), and this part of the energy can be rewritten in the following form conventional for the macroscopic phenomenological theory:

\[ \mathcal{E}_\mu^{(2)} = J \frac{\partial \hat{\mu}_i}{\partial x_k} \frac{\partial \hat{\mu}_i}{\partial x_k} + D \hat{\mu} \cdot [\nabla \times \hat{\mu}], \]  

where \( J \) and \( D \) are the phenomenological constants expressed now through the parameters of the microscopical Heisenberg model:

\[ J = \frac{1}{12} \sum_i \sum_j J_{ij} c_i c_j |b_{ij}|^2, \]  

\[ D = -\frac{1}{6} \sum_i \sum_j c_i c_j D_{ij} \cdot b_{ij}, \]

and \( \hat{\mu} \) is a unit vector oriented along the local magnetic moment density \( M \). Parameters \( J \) and \( D \) determine spiralling of the spin structure. In particular, the minimization of Eq. 37 gives as a result the helix with propagation number

\[ k = \frac{D}{2J} \]

and the vector \( \hat{\mu} \) rotating in the plane perpendicular to the helix axis.

The absence of canting in Eq. 37 does not mean that the canting does not affect the spiralling. As a matter of fact, the contribution of canting is taken into account, when choosing the fictitious atomic coordinates with Eq. (28). The simple additive expressions 38 and 39 can appear only because the contributing bonds \( b_{ij} \) are the functions of the exchange coordinates. Notice that, due to the dependence of the vectors \( b_{ij} \) on the exchange constants, both \( J \) and \( D \) are nonlinear functions of \( J_{ij} \).

**VII. THE EXAMPLE OF THE MnSi-TYPE CRYSTALS**

Let us illustrate the theory developed by the simple example of the MnSi-type crystals with the B20 structure. The cubic crystal MnSi has the space group \( P2_13 \). Its unit cell contains four magnetic manganese atoms in the position 4a (at threefold axes) with \( x = 0.138 \). In accordance with Ref. [23], we define four magnetic shells by the non-equivalent bonds \( b_1 = (-2x, \frac{1}{2}, \frac{1}{2} - 2x) \), \( b_2 = (1 - 2x, \frac{1}{2}, \frac{1}{2} - 2x) \), \( b_3 = (-2x, \frac{1}{2}, -\frac{1}{2} - 2x) \), and \( b_4 = (1, 0, 0) \), with corresponding exchange constants \( J_1 - J_4 \) and DM vectors \( D_1 - D_4 \).

The first atom \((x, x, x)\) has 6 bonds of each kind: \((-2x, \pm \frac{1}{2}, \frac{1}{2} - 2x) \), \((1 - 2x, \pm \frac{1}{2}, \frac{1}{2} - 2x) \), \((-2x, \pm \frac{1}{2}, -\frac{1}{2} - 2x) \), \((\pm 1, 0, 0) \) (the symbol \( \circ \) means possible cyclic permutations of the coordinates). The corresponding DM vectors can be obtained from \( D_1 - D_4 \) by the same sign changes and cyclic permutations of the components.
We assign $c_i = 1$ for all the manganese atoms in the equivalent positions. Then, the condition (28) for the atom $(x, x, x)$ can be rewritten as

$$\sum_j J_{ij} b_{ij} = 0 \quad \text{(41)}$$

or, using the symmetry 3 of the position,

$$J_1 \left( \frac{1}{2} - 4x \right) + J_2 \left( \frac{3}{2} - 4x \right) + J_3 \left( \frac{1}{2} - 4x \right) + J_4 \cdot 0 = 0 \quad \text{(42)}$$

and

$$x_{\text{exch}} = \frac{J_1 + 3J_2 - J_3}{8(J_1 + J_2 + J_3)} \quad \text{(43)}$$

in accordance with Ref. [23]. The 4th neighbors do not influence the exchange coordinate, because the bond $b_4$ connect the atoms belonging to the same magnetic sublattice.

From Eqs. (38), (39) the macroscopic parameters can be found,

$$J = 2(J_1b_1^2 + J_2b_2^2 + J_3b_3^2 + J_4b_4^2) \quad \text{(44)}$$

$$D = -4(D_1 \cdot b_1 + D_2 \cdot b_2 + D_3 \cdot b_3 + D_4 \cdot b_4) \quad \text{(45)}$$

After substitution of the exchange coordinate $x_{\text{exch}}$ into the $b$ vectors, the macroscopic exchange parameter can be expressed through the exchange constants of the bonds,

$$J = \frac{3J_1^2 + 3J_2^2 + 3J_3^2 + 10J_1J_2 + 10J_1J_3 + 22J_2J_3}{4(J_1 + J_2 + J_3)} + 2J_4 \quad \text{(46)}$$

which coincides with the result from Ref. [23].

The atom $(x, x, x)$, belonging to the 1st magnetic sublattice of the crystal, has two atoms of each other sublattice (2, 3, 4) in 1st, 2nd and 3rd magnetic shells, and six atoms of the same sublattice (1) in the 4th magnetic shell. Therefore, the condition (31) on tilt vectors can be written for the atom $(x, x, x)$ as

$$6(J_1 + J_2 + J_3 + J_4)\rho_1 = 2(J_1 + J_2 + J_3)(\rho_2 + \rho_3 + \rho_4) + 6J_4\rho_1 + 2D_+(1, 1, 1) \quad \text{(47)}$$

with $D_+ = D_{1x} + D_{1z} + D_{2x} + D_{2z} + D_{3x} + D_{3z}$. Using the symmetry equation

$$\rho_1 + \rho_2 + \rho_3 + \rho_4 = 0 \quad \text{(48)}$$

we find

$$\rho_1 = (\rho_x, \rho_x, \rho_x) \quad \text{(49)}$$

$$\rho_x = \frac{D_+}{4(J_1 + J_2 + J_3)} \quad \text{(50)}$$

in accordance with Ref. [23]. The tilt vectors $\rho_2, \rho_3$ and $\rho_4$ of other manganese atoms in the unit cell can be obtained from $\rho_1$ by the corresponding symmetry transformations. As we see from Eq. (50), the 4th magnetic neighbors, belonging to the same sublattice, do not influence the tilt vectors.
VIII. THE CASE OF Cu$_2$OSeO$_3$

The cubic crystal Cu$_2$OSeO$_3$ has the space group P2$_1$3, the lattice constant $a = 8.925$ Å. Its unit cell contains 16 magnetic copper atoms: four in the position 4$a$ with coordinates $r_I = (x_I, x_I, x_I)$ = (0.8860, 0.8860, 0.8860) (at threefold axes) and twelve in the general position 12$b$ with coordinates $r_{II} = (0.1335, 0.1211, 0.8719)$ $^{24}$. The nearest magnetic environment is determined by four types of nonequivalent bonds, but following Ref. [18] we will take into account an additional bond with the atoms of the second magnetic shell (Table I).

### TABLE I: Cu$_2$OSeO$_3$ crystal: five nonequivalent bonds between copper atoms with the corresponding exchange constants $J$ and DM vectors $\mathbf{D}$, taken from Ref. [18]. The lattice parameter $a = 8.925$Å and atomic coordinates from Ref. [24].

| $b$ | $|b|$,Å | $J$, meV | $\mathbf{D}$, meV | $\angle(\mathbf{b}, \mathbf{D})$ |
|-----|--------|---------|------------------|------------------|
| $-\frac{1}{3} - x_{II} + z_{II}, \frac{1}{3} - x_{II} - y_{II}, 1 - y_{II} - z_{II}$ | 3.054 | $J_1 = 1.132$ | $D_{1x}, D_{1y}, D_{1z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $-1 + x_{II} - x_{II}, -1 + x_{II} - y_{II}, x_{II} - z_{II}$ | 3.049 | $J_2 = -6.534$ | $D_{2x}, -D_{2y}, -D_{2z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $-1 - x_{II} + z_{II}, x_{II} - y_{II}, 1 + y_{II} - z_{II}$ | 3.226 | $J_3 = 3.693$ | $D_{3x}, -D_{3y}, D_{3z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $1 - x_{II} - x_{II}, -\frac{1}{3} + x_{II} - y_{II}, 1 - 3 - x_{II} - z_{II}$ | 3.304 | $J_4 = -0.900$ | $D_{4x}, D_{4y}, -D_{4z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $-\frac{1}{3} - x_{II} - x_{II}, -1 - x_{II} - y_{II}, -\frac{1}{3} + x_{II} - z_{II}$ | 6.349 | $J_5 = -0.984$ | $D_{5x}, D_{5y}, -D_{5z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |

The magnetic neighbors of the atom $r_I$ are nine copper atoms in positions 12$b$. Three of them are listed in Table II. Because the atom $r_I$ is on the 3-fold axis [111], the remaining six bonds can be obtained by cyclic permutations of the coordinates of the vectors.

### TABLE II: Magnetic neighbors of the copper $r_I$, energy parameters of the bonds and tilt vectors $\rho$. Only three neighbors are listed. Other six bonds can be obtained by threefold rotations, i.e. by cyclic permutations of the coordinates of the vectors.

| $r_I = r_I + b_{ij}$ | $J_{ij}$ | $\mathbf{D}_{ij}$ | $\mathbf{\rho}$ |
|----------------------|---------|------------------|------------------|
| $1 + x_{II}, 1 + y_{II}, z_{II}$ | $J_2$ | $-D_{2x}, -D_{2y}, -D_{2z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $1 - x_{II}, \frac{1}{3} + y_{II}, \frac{3}{2} - z_{II}$ | $J_3$ | $D_{3x}, -D_{3y}, D_{3z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $-\frac{1}{3} - x_{II}, 1 - y_{II}, \frac{3}{2} + z_{II}$ | $J_5$ | $D_{5x}, D_{5y}, -D_{5z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |

The magnetic environment of the atom $r_{II}$ contains seven copper atoms: four in positions 12$b$ and three in positions 4$a$ (Table III).

### TABLE III: Magnetic neighbors of the copper $r_{II}$, energy parameters of the bonds and tilt vectors $\mathbf{\rho}$. 

| $r_I = r_{II} + b_{ij}$ | $J_{ij}$ | $\mathbf{D}_{ij}$ | $\mathbf{\rho}$ |
|----------------------|---------|------------------|------------------|
| $-\frac{1}{3} + z_{II}, \frac{1}{2} - x_{II}, 1 - y_{II}$ | $J_1$ | $D_{1x}, D_{1y}, D_{1z}$ | $\rho_{11,z}, -\rho_{11,x}, -\rho_{11,y}$ |
| $\frac{1}{2} - y_{II}, 1 - z_{II}, \frac{1}{2} + x_{II}$ | $J_1$ | $D_{1y}, D_{1z}, -D_{1x}$ | $-\rho_{11,y}, -\rho_{11,z}, \rho_{11,x}$ |
| $-1 + x_{II}, -1 + x_{II}, x_{II}$ | $J_2$ | $D_{2x}, D_{2y}, D_{2z}$ | $\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $-1 + z_{II}, x_{II}, 1 + y_{II}$ | $J_3$ | $D_{3x}, D_{3y}, D_{3z}$ | $\rho_{11,z}, \rho_{11,x}, \rho_{11,y}$ |
| $y_{II}, -1 + z_{II}, 1 + x_{II}$ | $J_3$ | $-D_{3y}, -D_{3z}, -D_{3x}$ | $\rho_{11,y}, \rho_{11,z}, \rho_{11,x}$ |
| $1 - x_{II}, -\frac{1}{3} + x_{II}, \frac{3}{4} - x_{II}$ | $J_4$ | $D_{4x}, D_{4y}, D_{4z}$ | $-\rho_{11,x}, \rho_{11,y}, \rho_{11,z}$ |
| $\frac{1}{3} - x_{II}, 1 - x_{II}, -\frac{1}{4} + x_{II}$ | $J_5$ | $D_{5x}, D_{5y}, D_{5z}$ | $-\rho_{11,x}, -\rho_{11,y}, \rho_{11,z}$ |

All 16 copper positions in the unit cell are characterized by “sense” numbers $c_i$ and tilt vectors $\mathbf{\rho}_i$ determining...
canting. Owing to ferrimagnetic order, the spins of the copper atoms in positions 4a are opposite to those in positions 12b and to the summary magnetic moment \( r \), so we can assign the values \( c_I = -1 \) and \( c_{II} = 1 \). The tilt vectors of the atoms \( r_I \) and \( r_{II} \) are chosen as \( (\rho_{I,x}, \rho_{I,y}, \rho_{I,z}) \) and \( (\rho_{II,x}, \rho_{II,y}, \rho_{II,z}) \), correspondingly. Tilt vectors of other copper atoms can be obtained from these two vectors using symmetry transformations of the point group 23.

Let us use the condition (28) for the atoms \( r_I \) and \( r_{II} \), in order to find exchange coordinates of the positions. Then, the system of linear equations can be easily obtained and solved:

\[
A \begin{pmatrix}
    x_I \\
    x_{II} \\
    y_{II} \\
    z_{II}
\end{pmatrix} = \begin{pmatrix}
    -2J_2 - 3J_4 - 2J_5 \\
    -J_2 + J_3 + J_4 + \frac{1}{2}J_5 \\
    -\frac{3}{2}J_1 - J_2 + J_3 - \frac{3}{2}J_4 + J_5 \\
    -\frac{3}{2}J_1 - 2J_3 + \frac{3}{2}J_4 - \frac{1}{2}J_5
\end{pmatrix},
\]

with

\[
A = \begin{pmatrix}
    -3J_2 - 3J_4 - 3J_5 & J_2 - J_4 - J_5 & J_2 + J_4 - J_5 & J_2 - J_4 + J_5 \\
    -J_2 + J_4 + J_5 & -2J_1 + J_2 - 2J_3 + J_4 + J_5 & -J_1 + J_3 & J_1 + J_3 \\
    -J_2 - J_4 + J_5 & -J_1 + J_3 & -2J_1 + J_2 - 2J_3 + J_4 + J_5 & -J_1 + J_3 \\
    -J_2 + J_4 - J_5 & J_1 + J_3 & -J_1 + J_3 & -2J_1 + J_2 - 2J_3 + J_4 + J_5
\end{pmatrix}.
\]

(51)

General solution is rather cumbersome. Using the values of \( J_1-J_5 \) from Table I we find

\[
\begin{align*}
    x_{I,exch} &= 0.9417, \\
    x_{II,exch} &= -0.0042, \\
    y_{II,exch} &= 0.0202, \\
    z_{II,exch} &= 0.8969.
\end{align*}
\]

(52)

Just these ideal coordinates, rather than the real ones, should be used in calculations of the phenomenological constants \( J, D \) by the formulae (28), (29). The calculation with the data from Table I gives \( J = 2.565 \text{ meV}, D = -0.379 \text{ meV} \). Therefore, the helix propagation number \( k = D/2J = -0.0738 \) (\( q = k/2\pi = -0.0118 \)). The sign “minus” means that the magnetic helix is expected to be left-handed in the crystals with the given set of atomic positions. In the enantiomorphs with opposite values of all atomic positions the helix should be also opposite (right-handed). This prediction is rather easy for experimental proof (the corresponding procedure is well developed for MnSi-type crystals [25, 26]).

In order to find the tilt vectors, we write the system (31) for the atoms \( r_I \) and \( r_{II} \),

\[
A \begin{pmatrix}
    \rho_{I,x} \\
    \rho_{II,x} \\
    \rho_{II,y} \\
    \rho_{II,z}
\end{pmatrix} = \begin{pmatrix}
    D_{2x} + D_{2y} + D_{2z} - D_{4x} + D_{4y} - D_{4z} - D_{5x} - D_{5y} + D_{5z} \\
    -D_{1x} - D_{1y} + D_{2x} - D_{3x} + D_{3y} + D_{4x} + D_{5x} \\
    -D_{1y} - D_{1z} + D_{2y} - D_{3y} + D_{3z} + D_{4y} + D_{5y} \\
    D_{1x} - D_{1z} + D_{2x} + D_{3x} - D_{4x} + D_{4z} + D_{5x}
\end{pmatrix}.
\]

(54)

Using the data from Table I we find

\[
\begin{align*}
    \rho_{I,x} &= -0.000887, \\
    \rho_{II,x} &= 0.0285, \\
    \rho_{II,y} &= 0.0105, \\
    \rho_{II,z} &= 0.0586.
\end{align*}
\]

(55)
The canting angles are $|\rho_I| = 0.00154 \ (0.088^\circ)$, $|\rho_{II}| = 0.0660 \ (3.784^\circ)$.

The tilt vectors $\mathbf{u}^{(1)}$ determine the spin cantings $\mathbf{u}(1)$ of the first approximation, both in arbitrary twisted phases, including helicoids and A-phase, and in the state unwound by magnetic field $H > H_{c2}$. For example, Fig. 4 shows the canting arrangement in the periodic structure, if the external magnetic field is along the [001] axis.

The absolute atomic configuration of Cu$_2$OSeO$_3$ was determined in Ref. [24]. We have found that it is very close to $P4_132$ cubic symmetry [29], see Table IV where the idealized structure with the $P4_132$ symmetry is compared with real structure. Thus, similar to the case of $B20$ structures [25], we can say that this atomic structure is right-handed because the space group $P4_132$ contains only the right-handed screw axes $4_1$. And in this right-handed structure the magnetic helix is expected to be left-handed according to above calculations. For another enantiomorph with inverse values of all atomic coordinates, the idealized atomic structure is left-handed ($P4_33$ space group). The corresponding inverted real structure (it has exactly the same energy as the structure determined in Ref. [24]) can be also considered as left-handed and its magnetic helix is expected to be right-handed, but its space group remains $P2_13$.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|}
\hline
atoms & $P2_13$, Ref. [24] & $P4_132$ & \\
\hline
& type & $x, y, z$ & type & $x, y, z$ & \\
\hline
Cu-I & 4a & 0.8860,0.8860,0.8860 & 4b & $\frac{7}{8}, \frac{7}{8}, \frac{7}{8}$ & \\
Cu-II & 12b & 0.1335,0.1211,-0.1281 & 12d & $y + \frac{1}{4}, \frac{1}{4}, y \ (y \approx -0.12)$ & \\
Se-I & 4a & 0.4590,0.4590,0.4590 & 4c & $x, x, x \ (x \approx 0.5)$ & \\
Se-II & 4a & 0.2113,0.2113,0.2113 & & & \\
O-I & 4a & 0.0105,0.0105,0.0105 & 8c & $x, x, x \ (x \approx 0.0)$ & \\
O-II & 4a & 0.7621,0.7621,0.7621 & & & \\
O-III & 12b & 0.2699,0.4834,0.4706 & 12d & $-y, y + \frac{1}{4}, \frac{1}{4} \ (y \approx -0.27)$ & \\
O-IV & 12b & 0.2710,0.1892,0.0313 & 12d & $y + \frac{1}{4}, \frac{1}{4}, y \ (y \approx 0.03)$ & \\
\hline
\end{tabular}
\caption{The real atomic structure of Cu$_2$OSeO$_3$ [24] with $P2_13$ symmetry and the idealized structure with $P4_132$ symmetry.}
\end{table}

\section*{IX. DISCUSSION}

As it can be seen from Eq. (53), the maximal deviation of the exchange coordinate from the real one is for $x_{II}$, $|x_{II,exch} - x_{II,real}| = 0.14$. In Ref. [23] it is shown that for itinerant magnetics of the MnSi-type in the frame of RKKY model, $x_{exch}$ can have practically any value (we would remind that the ideal coordinates are nothing but some functions of the exchange parameters $J_{ij}$). Nevertheless, some limitation can result from the condition of validity of the theory. In order to study this we should first understand the origin of the exchange coordinates. When the transition is performed from discrete spins to continuous magnetic moment, some frustrations appear from the fact that the spins correspond poorly to the values of smooth function (e.g. sinusoid) in their physical positions. It is just the cause why, accordingly to Eq. (12), two kinds of canting can be distinguished: the first one determined by the DM interaction, and the second one connected with spatial derivatives. The latter is due to existence of several magnetic helices, with small phase shifts from that of the average single helix. The appropriate displacement of the magnetic atoms into the fictitious positions makes all the individual helices confluent, and the canting due to spatial derivatives disappears (Fig. 5). In fact, the small deviation of the exchange coordinates from the real ones means
that the phase shifts of the individual helices are also small, which is corresponding to the assumption about small cantings between neighboring spins. Greater phase shifts would mean that we could not already consider the spins as weak noncollinear, and the simplifications of Sec. \[V\] would impossible.

There is another interesting property of the exchange coordinates relating to the fact, known from the superexchange theory, that the DM vectors are approximately perpendicular to the bonds (see also last column in Table \[I\]). It is seen from Eq. \(39\), that the phenomenological parameter \(D\) determining spiralling of the magnetic structure is composed of the scalar products \(D \cdot b\). This means that, were the perpendicularity of the DM vectors to the bonds exact, the spiralling would be absent on condition that Eq. \(39\) contained vectors \(b\) with real coordinates. However, the expression depends on the exchange ones, and, consequently, \(D\) is a linear combination of differences \(x_{\text{exch}} - x_{\text{real}}\) with coefficients from the DM vectors components. Therefore, the exact numerical values of \(x_{\text{exch}}\) can strongly affect the spiralling, determining both pitch and sense of the magnetic helix.

In the first approximation, the cantings, being only small corrections to the spins, are perpendicular to the latters. Consequently, the cantings of all \(N\) magnetic atoms in the unit cell are determined by \(2N\) variables. Thus, for Cu$_2$OSeO$_3$ the number of canting components amounts to 32 per unit cell. Surprisingly, that, owing the (cubic) symmetry of the crystal, we can make with a considerably lower number of parameters in order to describe the local magnetic structure: four exchange coordinates \(x_{I,\text{exch}}, x_{II,\text{exch}}, y_{II,\text{exch}}, z_{II,\text{exch}}\), and four tilt vector components \(\rho_{I,x}, \rho_{II,x}, \rho_{II,y}, \rho_{II,z}\). For MnSi-type helimagnetics the number of the parameters is two: \(x_{\text{exch}}\) and \(\rho_x\).

In Eq. \(55\), the values of the tilt vectors \(\rho\) are calculated with the use of the energetic parameters \(J, D\) from Ref. \[18\]. In addition to the propagation number \(k\), given by Eq. \(10\), these, characterizing the canting, vectors are the experimentally observed parameters of the magnetic structure. In Ref. \[21\] the conditions were proposed of a diffraction experiment to find the canting in the MnSi-type helimagnets. Because the magnetic atoms in these crystals are situated at the special positions 4\(a\), the tilt vectors are defined by only one parameter \(\rho_x\), which is proposed to be found in the measurement of the “forbidden” Bragg reflection \(00\ell, \ell = 2n + 1\) in the unwound by magnetic field structure. In the general case of cubic helimagnets with almost collinear spins, the antiferromagnetic (canting) part of the structure factor is determined by the vector

\[
\Phi = \sum_i c_i \rho_i \exp(iQ \cdot r_i),
\]

where \(Q = 2\pi(hk\ell)\) is the reflection vector, and summation is taken over all magnetic atoms in the unit cell. In particular, for the copper atoms in the general positions 12\(b\) and the pure magnetic reflection \(00\ell, \ell = 2n + 1\), this sum has the view

\[
\Phi = 4(\rho_y \cos 2\pi \ell x + \rho_z \cos 2\pi \ell y + \rho_x \cos 2\pi \ell z, i\rho_y \sin 2\pi \ell x + i\rho_x \sin 2\pi \ell y + i\rho_z \sin 2\pi \ell z, 0).
\]

It is obvious that, in order to find all components of the tilt vectors \(\rho\) for magnetic atoms in all nonequivalent positions, several forbidden reflections with different \(\ell\) should be measured.

Notice that, as far as the measurable parameters \(k, \rho\) are determined by greater number of constants \(J_{ij}, D_{ij}\), the problem of finding of the latter from experimental data is unsolvable in general. At the best, only some combinations of \(J\) and \(D\) can be calculated by inversion of Eqs. \(10\), \(14\). Nevertheless, the comparison of theoretical predictions with measurements can confirm (or refute) reliability of \textit{ab initio} calculations of the energetic parameters.

We have shown that the canting is not only an independent effect of the antisymmetric Dzyaloshinskii–Moriya
exchange, but it also influences the spiralling, even in the absence of the DM interaction. Particularly, taking the
canting into account results in more correct estimation of the propagation number $k = 2\pi q$. Nevertheless, the obtained
here value $|q| = 0.0118$ is closer to the calculated in Ref. [18] $|q| = 0.0104$ than to the experimentally measured value
0.014 [8, 9].

### Table V: The estimation of the significance of the individual bond contributions into the macroscopic constants $J$ and $D$, and the propagation number $k = 2\pi q$.

| Bond | $|\mathbf{D}|$ | $J$, meV | $D$, meV | $k$ |
|------|---------|--------|--------|-----|
| all 5 bonds | | 2.565 | −0.379 | −0.074 |
| $J_1 = |\mathbf{D}_1| = 0$ | 1.310 | −0.967 | −0.369 |
| $J_2 = |\mathbf{D}_2| = 0$ | 2.166 | 0.286 | 0.066 |
| $J_3 = |\mathbf{D}_3| = 0$ | 2.170 | −0.948 | −0.218 |
| $J_4 = |\mathbf{D}_4| = 0$ | 1.942 | 0.027 | 0.007 |
| $J_5 = |\mathbf{D}_5| = 0$ | 1.677 | −0.179 | −0.053 |

Another important property is the sense of the magnetic chirality. In the case when the twist arises from a
spontaneous break of symmetry, the helix sense can be plus or minus with equal probability and in each case it is
casual. In contrast, for the magnetics without center of inversion, exemplified by MnSi, Cu$_2$OSeO$_3$ and other crystals,
the sense of the magnetic chirality correlates with the structural one. Thus, in pure MnSi, the left-handed atomic
structure results in the left-handed magnetic helix [26, 27]. Besides, the chiral interlink in MnSi-type helimagnetics
is found to be strongly dependent on their atomic composition [23, 25, 28]. For Cu$_2$OSeO$_3$ the question about
its magnetic chirality still remains open. In the present work, using the magnetic data from Ref. [18], we obtain
$q = −0.0118$, i.e. the helix is left for the structure described in Ref. [24] and used in simulations of Ref. [18]. In
order to understand the importance of contributions from different interatomic bonds to the helix sense, we can
study the influence of individual bonds on the macroscopic constants $J$ and $D$, and the helix propagation number
$k = 2\pi q$ just by excluding the bonds one by one (Table [V]). It is seen from the Table, that removing of any of the
five considered bonds causes an essential change of the helicity. It is noteworthy, that even the 5th bond, belonging
to the second magnetic shell ($d_5 = 6.35\text{Å}$), makes the phenomenological parameters two times less, which seems to
be in contradiction with our intuitive expectation. Indeed, it is usually assumed, that the contribution from the
2nd and next shells is small due to the exponential decay of the exchange constants with growing distance between
interacting atoms. In the case of Cu$_2$OSeO$_3$ the exchange parameter $J_5$ of the 5th bond is comparable with those
of the bonds belonging to the first magnetic shell (Table [I]). Besides, the contribution of an individual bond into
the condition [28] on the exchange coordinates is proportional to $Jd$, with $d$ being the distance between interacting
magnetic atoms, whereas the bond contributions into the macroscopic parameters $J$ and $D$, accordingly to Eqs. [18],
[39], are proportional to $Jd^2$ and $Dd$, correspondingly. It is most probable that the contributions from other bonds
from the 2nd and next magnetic surroundings should be taken into account, which can change not only the helix pitch
(Fig. [6]) but also its sense.
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FIG. 1: (color online). The twist and canting. The canting of spin sublattices leads to the combination of two helices with the same propagation vector and a constant phase shift.

FIG. 2: (color online). Three magnetic atoms (green) in positions connected by rotational symmetry elements, and their nearest magnetic environments. In absence of nonsymmetric DM interaction, a balance is achieved, when the spins in the equivalent positions (equally colored circles) have the same directions.
FIG. 3: Deviation of the spin $s$ from the equilibrium state $\mu$ in presence of non-isotropic DM interaction. The tilt angle $\phi \sim D/J$. In a simple approximation the deviation can be decomposed into two parts, $u^{(1)} \perp \mu$ and $u^{(2)} \parallel \mu$.

FIG. 4: (color online). The spin canting in the ferrimagnetic state of Cu$_2$OSeO$_3$, induced by the external magnetic field applied in the [001] direction. The green darts and cyan arrows show directions and magnitudes of the spin cantings of 4 Cu-I and 12 Cu-II atoms, correspondingly. In the first approximation all cantings lie in the plane perpendicular to the field and magnetization. It is seen that the canting arrangement is symmetrical relative to the 2$_1$ screw axes directed along the field.
FIG. 5: (color online). (a) In the real spin structure there are several magnetic helices, corresponding to different magnetic atoms in the unit cell, with phase shifts between them. (b) When the atoms “move” from the real positions to the fictitious (exchange) ones, the phase shifts disappear and the helices become confluent.

FIG. 6: (color online). The propagation number $k$ dependencies on the isotropic exchange constants of unconsidered in Ref. [18] bonds with 5.35Å < $d$ < 5.54Å, 1-8: $(-x_{II} - y_{II}, -\frac{1}{2} - y_{II} + z_{II}, z_{I} - x_{II} - z_{II})$, $(\frac{1}{2}, \frac{1}{2} - 2y_{II}, 2 - 2z_{II})$, $(\frac{1}{2} - 2x_{II}, -2y_{II}, -2z_{II})$, $(-2x_{II}, \frac{1}{2} - 2y_{II}, 2z_{II})$, $(\frac{1}{2} - x_{II} - z_{II}, -x_{II} - y_{II}, \frac{1}{2} + y_{II} - z_{II})$, $(-\frac{1}{2} + x_{I} - x_{II}, \frac{1}{2} - x_{II} - y_{II}, 2 - x_{I} - z_{II})$, correspondingly. Solid, dash, and dash-dot lines indicate Cu-II/Cu-II, Cu-II/Cu-I, and Cu-I/Cu-I bonds, correspondingly. It is seen from the graphs, that even relatively small exchange interaction between atoms at this distance ($d < d_5$) is sufficient in order to explain the discrepancy between theoretical predictions and the experimental data ($|k_{real}| = 0.088$).