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Electric polarization evolution equation for antiferromagnetic multiferroics with the polarization proportional to the scalar product of the spins

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Abstract

The spin current model of electric polarization in multiferroics is justified via the quantum hydrodynamic method and the mean-field part of the spin-orbit interaction. The spin current model is applied to derive the electric polarization proportional to the scalar product of the spins of the nearby ions, which appears to be caused by the Dzylaoshinskii-Moriya interaction. The symmetric tensor spin structure of the polarization is discussed as well. We start our derivations for the ferromagnetic multiferroic materials and present further generalizations for the antiferromagnetic multiferroic materials. We rederive the operator of the electric dipole moment, which provides the macroscopic polarization obtained via the spin current model. Finally, we use the quantum average of the found electric dipole moment operator to derive the polarization evolution equation for the antiferromagnetic multiferroic materials. The possibility of spiral spin structures is analyzed.

1. Introduction

The Landau–Lifshitz–Gilbert equation happens to be a highly effective method for the theoretical study of the macroscopic processes of the magnetization evolution in the magnetically ordered materials. However, if we dial with the multiferroic materials, it requires an equation for the polarization evolution. Therefore, the problem of the derivation of the polarization evolution equation is formulated [1, 2]. There are three mechanisms of the polarization formation in II-type multiferroics [3], where the magnetic properties related to the dielectric properties, while the I-type multiferroics are the materials, where the magnetic properties and the ferroelectric properties coexist without strong interference. These mechanisms are the exchange-striction mechanism (symmetric-parallel components of spins gives the polarization), the spin-current/inverse Dzylaoshinskii-Moriya model (antisymmetric-perpendicular components of spins gives the polarization), the spin dependent p-d hybridization related to the spin of single magnetic ion. Described mechanisms of ferroelectricity of spin origin are summarized in figure 2 of [3].

Particularly, there is the inverse Dzylaoshinskii-Moriya model associated with the spin current, which is also called the spin current model. In this paper, we are focused on the analytical justification of the spin current model, which basically states that the polarization is proportional to the spin-current $P^{\alpha} \sim \varepsilon^{\alpha\beta\gamma} J^{\beta\gamma}$, where P^{α} is the polarization, $\varepsilon^{\alpha\beta\gamma}$ is the three-dimensional Levi-Civita symbol, $J^{\beta\gamma}$ is the spin current tensor. We show that it can be derived with no relation to the particular mechanism of the polarization formation. Further application of the spin current caused by different mechanisms leads to the electric dipole moment being proportional to the scalar product of the spins, or the electric dipole moment being proportional to the vector product of the spins. The analytical derivation of polarization (its macroscopic form and corresponding microscopic electric dipole moment) allows to specify the interaction leading to the physical mechanisms of the polarization formation in two mentioned regimes.

The polarization evolution equation for multiferroics with ferromagnetic order of spins for two of described mechanisms (the symmetric and antisymmetric regimes) are considered in [1, 2]. Here, we consider the polarization evolution equation for the antiferromagnetic multiferroics, where the electric polarization is proportional to the scalar product of the spins of the neighboring ions.

The Landau–Lifshitz–Gilbert equation contains a number of terms representing major phenomena existing in the magnetically ordered materials. One of the major interactions in the ferromagnetic materials is the exchange interaction, which is modeled by the Heisenberg Hamiltonian.

Multiferroic materials are the magnetically ordered materials, so similar mechanisms can affect the dynamics of polarization in the multiferroic materials. It is especially related to the II-type multiferroics, where the magnetic and the dielectric phenomena are deeply related, in contrast to the I-type multiferroics, where the magnetic and the dielectric phenomena coexist. Therefore, we consider the role of exchange interaction in the evolution of polarization in the regime, where the electric dipole moment is proportional to the scalar product of the spins.

A classification of ferroelectrics is also presented in [4]. It shows the relation between materials and mechanism of the polarization formation (mechanism of inversion symmetry breaking) in this material (see table 1). It shows five mechanisms with relation to particular materials, but no analytical formalization is given in contrast with [3]. Nevertheless, [4] also includes the spin-atomic structure for a-b plane of perovskite $YNiO_3$ (see figure 1(d)), where we see antiparallel spins for the net of $Ni^{3+\delta}$ (smaller value of magnetic moment), we also see antiparallel spins for the net of $Ni^{3-\delta}$ (larger value of magnetic moment), while pairs of nearest $Ni^{3+\delta}$ and $Ni^{3-\delta}$ have parallel spins. This antiferromagnetic system of parallel/antiparallel spins can be associated with the results obtained in this paper. The fourth-order term in energy $\sim -P^2M^2$ is associated with $YMnO_3$ and $BiMnO_3$ [4] (see the text before equation (1) on page 16). It corresponds to the structural transition 'Geometric ferroelectrics.' It can be associated with the homogeneous magnetic ordering [4].

In contrast, the polarization can be described by the following equation for the inhomogeneous magnetic ordering [5–7]

$$\mathbf{P} \sim (\mathbf{S} \cdot \nabla)\mathbf{S} - \mathbf{S}(\nabla \cdot \mathbf{S}), \tag{1}$$

where the inhomogeneous magnetization can be associated with the rotation of the magnetic moments (see figures 4 and 5 in [4]). The antiferromagnetic analog of equation (1) can be found in [8, 9].

A model of the microscopic origin of electric polarization is given in [10]. The focus is made on the noncollinear magnetic order and formation of the electric polarization in Mott insulators. However, it is also shown that the form of the magnetoelectric coupling allows additional constructions in comparison with known configurations [3].

A slow change/rotation of the spin direction in a sample (see figure 4 in [4]) creates conditions for the noncollinear mechanisms of the polarization formation (see figures 2(d)–(f) of [3]). However, the collinear part of the relative spin orientation is nonzero, so the mechanism of the polarization related to the collinear spins can contribute in these systems. Coefficients giving the polarization can differs for different mechanisms, so one of mechanisms can be suppressed.

The area of the spin rotation leading to the noncollinear and the collinear spin formations appears in the domain walls. Magnon-induced domain wall motion in ordinary ferromagnets is considered in [11]. The Dzyaloshinskii-Moriya interaction as a mechanism of the magnon-driven domain-wall motion is considered in [12]. Antiferromagnetic domain wall motion induced by spin waves is considered in [13]. Magnon induced magnetization dynamics in multiferroics is considered in [14]. It includes the magnon-induced domain wall motion. Particularly, the Landau–Lifshitz–Gilbert equation is applied for the analysis, where an additional term is included to describe the coupling between the electric field and the magnetization in a manner associated with equation (1) (see equations (3) and (4)). Magnonic spin-transfer torque can efficiently drive a domain wall to propagate in the opposite direction to that of the spin wave, as demonstrated in [15]. An analytical derivation of the magnon-driven Dzyaloshinskii-Moriya torque is developed in [16]. It is also shown that the Dzyaloshinskii-Moriya interaction mediated by spin waves can generate a torque on a homogeneous magnetization that resembles the Rashba torque. The magnetization dynamics in a thin-film ferromagnet deposited on a topological insulator is studied [17]. It is focused on domain-wall motion via current and the possibility of a spin-wave torque acting on the magnetization. The coupling between the magnetic domain wall and the topological insulator removes the degeneracy of the wall profile with respect to its chirality and topological charge, as shown analytically [17]. The dynamics of a multiferroic domain wall in which an electric field can couple to the magnetization via the inhomogeneous magnetoelectric interaction is studied in [18]. It also demonstrates that in the stationary regime, the chirality of the domain wall can be efficiently reversed when the electric field is applied along the direction of the magnetic field. Some discussion of the structure of the Dzyaloshinskii constant and the Dzyaloshinskii-Moriya interaction can be found in [19] in context of the experimental analysis of magnetized Fe/Ni bilayers, where a new type of domain wall structure is reported.

Another object with the noncollinear spin structure is Skyrmions. Let us mention that the skyrmion-like spin structures formed by the thermal fluctuations in the ferromagnetic materials are studied in [20]. In [20] it is also specified that the uniaxial anisotropy K ($K > 0$, favoring perpendicular magnetic anisotropy with an energy of $-KM_z^2$) and 'effective stiffness' K_0 of the conical phase can be calculated from the in-plane and out-of-plane saturation fields.

In this paper we mainly consider the single-phase AFM multiferroics. However, important novel phenomena are discovered in multilayered, where the film is grown on the substrate. Application of our model depends on the properties of the materials. The current form of our model includes the connection between magnetic and dielectric properties of the materials, while multiferroics are well-known due to relation of the magnetic and dielectric properties to the elastic properties of the medium. However, the elasticity has not been considered in terms of our model. As an illustration we mention AFM/ferroelectric multiferroic heterostructures, such as experimentally demonstrated Mn-Pt/PMN-PT heterostructures [21]. Application of the electric field to $BaTiO_3$ leads to the strain appearance. As the consequence the strain appears in the intermetallic Mn_3Pt film. It causes changes in the magnetic properties of Mn_3Pt film. Particularly it changes properties of the topological anomalous Hall effect in a non-collinear phase of the antiferromagnet. So, the elastic properties play essential role in this phenomenon. Another example is the tunneling magnetoresistance effect [22]. An effect similar to the ferromagnet-antiferromagnet exchange-bias system is discovered for a room-temperature exchange-bias effect between a collinear antiferromagnet and a non-collinear antiferromagnet. The N*é* el pair anisotropy term between the nearest Mn-Pt pairs (equation (1) in section Methods of Supplementary materials of [22]) is applied at the numerical modeling. It structure is similar to the polarization in the spin dependent p-d hybridization mechanism.

Describing the magnetization and its dynamics, we usually focused on the spin density, while the spin of atom/ion is formed by the spins and orbital motion of elements of the atomic structure. The superposition of the spin angular momentum density and the orbital angular momentum density is obtained in [23] (see equation (14)). The conservation of the z component of the total angular momentum is demonstrated for a Heisenberg ferromagnet with isotropic exchange interaction [23].

We consider the Dzyaloshinskii-Moriya interaction to complete the spin current model for collinear spins (it is assumed that the collinear spin structure is formed by the Heisenberg exchange interaction, while further formation of spin related electric polarization is formed by the Dzyaloshinskii-Moriya interaction). So, we want to mention some continuous approaches based on the Dzyaloshinskii-Moriya Hamiltonian. The model used in [12] is based on the total free energy with a nonexplicit account of the Dzyaloshinskii-Moriya interaction. Its explicit contribution is given in the Landau–Lifshitz-Gilbert equation in the form of the torque $\mathbf{T} = \gamma\tilde{D}\mathbf{M} \times (\nabla \times \mathbf{M})$, with γ is the gyromagnetic ratio. It corresponds to the Dzyaloshinskii-Moriya interaction with a chiral energy of $\tilde{D}\mathbf{M} \cdot (\nabla \times \mathbf{M})$. In our work, we address the Dzyaloshinskii-Moriya interaction given by Hamiltonian $\hat{H} = (-1/2)\mathbf{D}_{ij} \cdot [\hat{\mathbf{s}}_i \times \hat{\mathbf{s}}_j]$ and derive a different form of the torque in the Landau–Lifshitz equation for the ferromagnetic and antiferromagnetic materials. In our derivation, we include the vector nature of the Dzyaloshinskii constant and its analytical structure.

The Dzyaloshinskii-Moriya interaction generates the torque on the magnetization. The Dzyaloshinskii-Moriya interaction mediated by spin waves is considered in [16] for systems displaying the interfacial Dzyaloshinskii-Moriya interaction. A thin magnetic film with the magnetization $\mathbf{m}$ aligned along the in-plane easy axis, the magnetic energy associated with Dzyaloshinskii-Moriya interaction reads [16] $W = -D\mathbf{m} \times [(\mathbf{z} \times \nabla) \times \mathbf{m}]$, where system with an interfacial inversion asymmetry along the normal z [24] (films and multilayers with in-plane and out-of-plane magnetization are considered with the prediction of the two-dimensional localized patterns), [25] (domain walls in ultrathin magnetic films are considered there). It leads to an effective fieldlike torque of the form $T \sim (\mathbf{m} \times \mathbf{z}) \times \mathbf{j}_m$, with $\mathbf{j}_m$ is the spin-wave current.

The many-particle quantum hydrodynamic method has been developed for the structureless mediums such as the quantum ultracold gases [26, 27] and plasma-like mediums However, it has been shown that it is possible to capture some features of solid state. Particularly, the material field form of the Landau–Lifshitz equation is derived from the many-particle Pauli equation in the coordinate representation [28]. It opened a possibility for the anlysis of the multiferroic materials in terms of the quantum hydrodynamic method. Therefore, in order to study the antiferromagnets, we present the derivation of the antiferromagnet analog of the Landau–Lifshitz equation. Next, in this paper, we develop a microscopically justified macroscopic spin current model. We also derive and apply the spin current caused by the Dzylaoshinskii-Moriya interaction to complete the derivation of the polarization. Obtained macroscopic polarization is compared with earlier suggested electric dipole moments of the pair of magnetic ions [3]. The polarization evolution equation is derived for the found form of polarization caused by the Dzylaoshinskii-Moriya interaction.

This paper is organized as follows. In section 2 the microscopic derivation of the spin evolution equation for the antiferromagnetic materials is developed within the many-particle quantum hydrodynamic method. In section 3 the approximate form of the polarization is considered for ferromagnetic and antiferromagnetic

multiferroics, if the electric dipole moment is proportional to the scalar product of the spins. In section 4 the spin current model is derived from the momentum balance equation with the spin-orbit interaction. Furthermore, in section 4, the spin-current caused by the Dzylaoshinskii-Moriya interaction is presented in order to obtain corresponding polarization. In section 5 the polarization evolution equation is derived as the evolution of the quantum average of the electric dipole moment operator under the influence of the Zeeman energy and the Coulomb exchange interaction, both for ferromagnetic and antiferromagnetic multiferroics. In section 6 the equilibrium solutions for the obtained model of multiferroics are discussed, including the spiral structures. In section 7 a brief summary of obtained results is presented.

2. The spin evolution equation for the antiferromagnetic material: the microscopic–based derivation with the exchange interaction

If we consider the spin evolution equation in the ferromagnetic materials with primarily exchange interaction, we find $\partial_t \mathbf{S} = (1/6) g_u [\mathbf{S}, \triangle \mathbf{S}]$, where $g_u = \int \xi^2 U(\xi) d^3\xi$. Firstly, it is based on the Heisenberg Hamiltonian $\hat{H} = -\frac{1}{2}\sum_{i=1}^{N}\sum_{j=1, j\neq i}^{N} U_1(r_{ij})(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j)$. Function $U_1(r_{ij})$ is the exchange integral. It depends on the distance between interaction particles. In combination with the spin operators, it gives an effective potential energy. Function $U_1(r_{ij})$ drops at the distance larger average interparticle distance. So, it includes the interaction of neighboring atoms or ions. It can include the interaction of the atoms separated by an atom, but it does not include the influence of atoms located at the further distances.

In order to get a systematic derivation of the spin evolution equation, we need to define the spin density of the system. For the quantum systems, it is defined as the quantum average of the spin operator $\hat{\mathbf{s}}_i$

$$\mathbf{S}(\mathbf{r},\, t) = \int \Psi^{\dagger}(R,\, t) \sum_i \delta(\mathbf{r} - \mathbf{r}_i)(\hat{\mathbf{s}}_i \Psi(R,\, t)) dR, \tag{2}$$

where i is the number of atoms. The spin operators obey the commutation relation

$$[\hat{s}_i^{\alpha},\, \hat{s}_j^{\beta}] = \imath\hbar\delta_{ij}\varepsilon^{\alpha\beta\gamma}\hat{s}_i^{\gamma}, \tag{3}$$

where α, β, γ are the tensor indexes, so each of them is equal to x, y, z, summation on the repeating Greek symbol is assumed, $\imath$ is the imaginary unit, δ_{ij} is the three-dimensional Kronecker symbol, $\varepsilon^{\alpha\beta\gamma}$ is the three-dimensional Levi-Civita symbol.

We consider systems of atoms or ions. Hence, we deal with structured objects or particles. There is the exchange interaction of valence electrons in each ion. It makes a contribution to the properties at the ion as the particle under consideration. However, there is a short-range correlation between neighboring ions due to the exchange interaction of valence electrons belonging to different ions. This interaction is included in the model presented below via the Heisenberg Hamiltonian.

Evolution of the wave function of the system of ions is described by the Pauli equation

$$\imath\hbar\, \partial_t \Psi(R,\, t) = \hat{H}\Psi(R,\, t). \tag{4}$$

In this paper, we mostly focused on the evolution of spin density and polarization under influence of the Zeeman energy and the Coulomb exchange interaction

$$\hat{H} = -\sum_{i=1}^{N} \hat{\boldsymbol{\mu}}_i \cdot \mathbf{B}_i - \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1, j\neq i}^{N} U(r_{ij})(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j), \tag{5}$$

$\hbar$ is the Planck constant, N number of atoms/ions in the system, $\Psi(R, t)$ many-particle wave function of the system, $R = \{\mathbf{r}_1,...,\mathbf{r}_N\}$, $\mathbf{B}_i$ is the magnetic field, acting on i-th atom, $\hat{\boldsymbol{\mu}}_i$ magnetic moment operator, which is proportional to the spin operator $\hat{\boldsymbol{\mu}}_i = \gamma_i \hat{\mathbf{s}}_i$ with the gyromagnetic ratio γ_i, $U_{ij} = U(\mathbf{r}_i - \mathbf{r}_j)$ is the exchange integral of the Heisenberg Hamiltonian as the function of the interparticle distance, (the exchange integral). Let us repeat that N is the full number of atoms under consideration. It can be considered as the sum of numbers of particles in each of two species of the magnetic atoms $N = N_A + N_B$. Formally, we have interaction between all pairs of atoms in the system in the second term of Hamiltonian (5), but the short range of function $U(r_{ij})$ leads to the contribution of the neighboring atoms only.

If we consider two subsystems for the antiferromagnetic material, we need to define the spin density for each subsystem

$$\mathbf{S}_s(\mathbf{r},\, t) = \int \Psi^{\dagger}(R,\, t) \sum_{i\in s} \delta(\mathbf{r} - \mathbf{r}_i)(\hat{\mathbf{s}}_i \Psi(R,\, t)) dR, \tag{6}$$

where index s refers to the number of the species of atoms, or, in the simplest case, it can be atoms of the same isotope with opposite spin projection. We focus on the system of two subspecies $s = A$ and $s = B$.

Next, we consider the spin evolution equation. The time derivative acts on the wave function, while the time derivative of the wave function is replaced with a Hamiltonian in accordance with the Pauli equation

$$\partial_t \mathbf{S}_s(\mathbf{r},\, t) = \frac{\imath}{\hbar}\int \sum_{i\in s} \delta(\mathbf{r} - \mathbf{r}_i)\Psi^{\dagger}(R,\, t)[\hat{H},\, \hat{\mathbf{s}}_i]\Psi(R,\, t)dR. \tag{7}$$

The partial contribution in the spin evolution equation from the Zeeman energy $-\sum_{i=1}^{N}\hat{\boldsymbol{\mu}}_i\cdot\mathbf{B}_i$ in Hamiltonian (5) leads to $\partial_t\mathbf{S}_s = \frac{2\mu}{\hbar}[\mathbf{S}_s,\, \mathbf{B}]$. If we consider the interaction of the nearest neighbors, which corresponds to the interaction of the different subspecies, we find the following contribution of the second term of Hamiltonian (5) in the spin evolution equation

$$\partial_t\mathbf{S}_s = g_{0u,AB}[\mathbf{S}_s,\, \mathbf{S}_{s'\neq s}] + \frac{1}{6}g_{u,AB}[\mathbf{S}_s,\, \triangle\mathbf{S}_{s'\neq s}]. \tag{8}$$

If we consider the interaction second row neighbors, which corresponds to the interaction of atoms of the same species we find

$$\partial_t\mathbf{S}_s = \frac{1}{6}g_u[\mathbf{S}_s,\, \triangle\mathbf{S}_s]. \tag{9}$$

If we consider antiferromagnetic material composed of atoms of the same species, we have same form of potential of interaction between atoms of the same subspecies and atoms of different subspecies. However, the signs of the potentials are different since the antiferromagnetic order corresponds to the negative exchange integral $g_{u,AB} < 0$ while atoms with the same spin direction have a positive exchange integral $g_u > 0$. So we can assume the following relation between the interaction constants $g_{u,AB} = -g_u$. Similarly, we have a relation for the zeroth order constant $g_{0u,AB} = -g_{0u}$. It allows us to combine equations (8) and (9) in order to get the spin evolution equation under the exchange interaction of two subspecies

$$\begin{aligned}\partial_t\mathbf{S}_s = \frac{2\mu}{\hbar}[\mathbf{S}_s,\, \mathbf{B}] - g_{0u}[\mathbf{S}_s,\, \mathbf{S}_{s'\neq s}]\\ + \frac{1}{6}g_u[\mathbf{S}_s,\, \triangle(\mathbf{S}_s - \mathbf{S}_{s'\neq s})].\end{aligned} \tag{10}$$

It is well known that the modeling of the antiferromagnetic materials includes superpositions of the partial magnetizations [29], in our case spin densities. Hence, we introduce $\boldsymbol{\Sigma} = \mathbf{S}_A + \mathbf{S}_B$ and $\mathbf{L} = \mathbf{S}_A - \mathbf{S}_B$. In literature, $\mathbf{L}$ is used for the difference of the magnetizations [29], hopefully it would not confuse the readers. It leads to the following equations

$$\partial_t\,\Sigma = \frac{2\mu}{\hbar}[\boldsymbol{\Sigma},\, \mathbf{B}] + \frac{1}{6}g_u[\mathbf{L},\, \triangle\mathbf{L}], \tag{11}$$

and

$$\partial_t\mathbf{L} = \frac{2\mu}{\hbar}[\mathbf{L},\, \mathbf{B}] + \frac{1}{6}g_u[\boldsymbol{\Sigma},\, \triangle\mathbf{L}] + g_{0u,AB}[\mathbf{L},\ \Sigma], \tag{12}$$

where we used simple representation $[\mathbf{S}_A, \mathbf{S}_B] = [\mathbf{S}_A - \mathbf{S}_B, \mathbf{S}_B] = [\mathbf{S}_A - \mathbf{S}_B, \mathbf{S}_A + \mathbf{S}_B - \mathbf{S}_A] = [\mathbf{L}, \boldsymbol{\Sigma}] - [\mathbf{S}_A - \mathbf{S}_B, \mathbf{S}_A] = [\mathbf{L}, \boldsymbol{\Sigma}] - [\mathbf{S}_A, \mathbf{S}_B]$. So, we get $2[\mathbf{S}_A, \mathbf{S}_B] = [\mathbf{L}, \boldsymbol{\Sigma}]$. Here we have $|\mathbf{S}_A| \approx |\mathbf{S}_B|$, and $\mathbf{S}_A \approx -\mathbf{S}_B$. So, the sum of partial spin densities $\boldsymbol{\Sigma} = \mathbf{S}_A + \mathbf{S}_B$ is a small value in the antiferromagnetic material, and $|\mathbf{L}| \approx 2|\mathbf{S}_A| \approx 2|\mathbf{S}_B|$.

Equation (11) is obtained for a small vector $|\boldsymbol{\Sigma}| \ll |\mathbf{L}|$. The first (second) term on the right-hand side is proportional to the small vector $\boldsymbol{\Sigma}$ (to the small combination of parameters $g_u\triangle\mathbf{L}$). In equation (12) we see the first term with no small parameters, the third term containing the small vector $\boldsymbol{\Sigma}$, and the second term containing the product of the small parameters $\boldsymbol{\Sigma}$ and $g_u\triangle\mathbf{L}$. So, the second term can be dropped in further applications. Equations (11) and (12) are well-known for the antiferromagnetic materials [29]. However, our derivation allows us to establish the explicit form of coefficients in this equation in relation to the microscopic nature of the interaction entering the Pauli equation (5). Our major goal in this paper is the derivation of the polarization evolution equation for the multiferroic materials. However, the derivation of equations (11) and (12) shows the usefulness of our method of derivation of the macroscopic equations from the microscopic theory. Details of the derivation are not considered here, but the method of derivation can be found in [2] and [26]. This work on the microscopic justification of the macroscopic equations for the magnetization and the polarization is a part of an interdisciplinary field, where similar justifications are made for the classical and quantum systems [26–28, 30, 31].

In this paper we focused on a partial microscopic derivation of the Landau–Lifshitz–Gilbert equation. Here we presented the contribution of the Heisenberg Hamiltonian. Below we consider the Zeeman energy and the Dzylaoshinskii-Moriya interaction. We also consider the spin-orbit interaction as the key element for the spin current model. However, the anisotropy energy and the Gilbert damping are consciously ignored, while these terms are crucial for the complete model of the magnetically ordered materials.

3. Microscopic definition of the macroscopic polarization in the exchange striction model

The electric dipole moment related to a pair of neighboring magnetic ions in the exchange striction regime is proportional to the scalar product of spins of these ions [3, 32, 33]

$$\mathbf{d}_i \sim \mathbf{\Pi}(\mathbf{s}_i \cdot \mathbf{s}_{i+1}). \tag{13}$$

The nonmagnetic ions contributing to the dipole moment are not considered explicitly in this equation. Equation (13) is useful to analyze a linear structure. Hence, we give the representation to the electric dipole moment of two ions

$$d_{ij}^{\alpha} = \overset{\alpha}{\Pi}_{ij}(r_{ij})(\mathbf{s}_i \cdot \mathbf{s}_j), \tag{14}$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the interparticle distance, and dependence of vector function $\Pi_{ij}^{\alpha}(r_{ij})$ ensures that the nearest neighbors give contribution in dipole moment d_{ij}^{α}.

We develop the quantum theory of multiferroics, so we need to make a transition to the operator form of the electric dipole moment via the consideration of the spin operators $\hat{d}_{ij}^{\alpha} = \Pi_{ij}^{\alpha}(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j)$. Considering all nearest neighbors of the i-th atom/ion, we get the full electric dipole moment related to this atom

$$\hat{d}_i^{\alpha} = \sum_{j \neq i} \Pi_{ij}^{\alpha}(r_{ij})(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j). \tag{15}$$

We can use the operator of the electric dipole moment (15) in order to find an approximate representation via the spin density, which can be an analog of the Mostovoy [7], but for different physical regime. Therefore, we present the quantum average of operator $\hat{\mathbf{d}}_i$, which gives the macroscopic polarization

$$\mathbf{P}(\mathbf{r},\, t) = \int \Psi^{\dagger}(R,\, t) \sum_i \delta(\mathbf{r} - \mathbf{r}_i)(\hat{\mathbf{d}}_i \Psi(R,\, t))\, dR. \tag{16}$$

Substituting operator (15) in the polarization definition (16) and account of the formation of the electric dipole moment by the nearest neighbors allows to get the required approximate form of the polarization. We need to explicitly introduce the interparticle distance in all functions in definition (16) as follows: $\mathbf{r}_i = \mathbf{R}_{ij} + (1/2)\mathbf{r}_{ij}$, and $\mathbf{r}_j = \mathbf{R}_{ij} - (1/2)\mathbf{r}_{ij}$, where $\mathbf{R}_{ij} = (\mathbf{r}_i + \mathbf{r}_j)/2$, and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$. This substitution includes the arguments of the wave function $\Psi(R, t) = \Psi(...,\mathbf{r}_i,...,\mathbf{r}_j,...,t)$.

3.1. The polarization of the ferromagnetic multiferroics

In this paper, we mostly focused on the antiferromagnetic materials. However, for comparison, it can be useful to obtain the polarization of the ferromagnetic multiferroics in the 'exchange striction' regime [3]. We use equation (16) with the operator of the electric dipole moment (15) and make the expansion on the interparticle distance (some details of the method are described in [2]). It allows us to get an approximate expression of the polarization (16) in terms of the spin density:

$$P^{\alpha} = g_{0\Pi}^{\alpha}\mathbf{S}^2 + \frac{1}{6} g_{\Pi}^{\alpha}(\mathbf{S} \cdot \triangle \mathbf{S}), \tag{17}$$

where $g_{0\Pi}^{\alpha} = \int \Pi^{\alpha}(r)\, d\mathbf{r}$, and $g_{(\Pi)}^{\alpha} = \int \xi^2 \Pi^{\alpha}(\xi)\, d\boldsymbol{\xi}$. Equation (17) can be also represented in the following form $P^{\alpha} = g_{0\Pi}^{\alpha}\mathbf{S}^2 + \frac{1}{3}\frac{1}{2^2} g_{\Pi}^{\alpha}[\triangle \mathbf{S}^2 - 2(\partial_{\mu} S^{\nu})(\partial_{\mu} S^{\nu})]$.

Equation (17) is the result of the expansion on the relative distance. We included three terms of expansion. The first and major term is proportional to $g_{0\Pi}^{\alpha}$. The second term is equal to zero. The third term is proportional to $g_{(\Pi)}^{\alpha}$ appears as a correction.

3.2. The polarization of the antiferromagnetic multiferroics

Formation of dipole (13) happens due to the presence of the nonmagnetic ion with the opposite charge between ions i and $i + 1$. If we consider the antiferromagnetic material, we find the alternation of the 'spin-up' and 'spin-down' ions. So, if ion i is 'spin-up' then ion $i + 1$ is 'spin-down'. It shows that if we consider a line of magnetic ions, we have the nonmagnetic ion after each magnetic ion. It has no relation to the spin direction of the magnetic ion. Hence, the effective dipole moment (15) can be associated with each magnetic ion ('spin-up' or 'spin-down')

$$\hat{d}_{i \in s}^{\alpha} = \sum_{j \in s'} \Pi_{ij}^{\alpha}(r_{ij})(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j), \tag{18}$$

where index s or s' refers to the subsystem of spin-up or spin-down ions. Moreover, s and s' refer to different subsystems. It leads to the polarization definition in the antiferromagnetic multiferroics

$$\mathbf{P}(\mathbf{r},\,t)=\int\Psi^{\dagger}(R,\,t)\sum_{i\in A\bigcup B}\delta(\mathbf{r}-\mathbf{r}_i)(\hat{\mathbf{d}}_i\Psi(R,\,t))dR, \tag{19}$$

where $s=A$ refers to the subsystem of spin-up ions, and $s=B$ refers to the subsystem of spin-down ions. The summation in equation (19) explicitly specifies that index s in operator (18) belongs to both subspecies A and B. The same is true for the index s', but they cannot belong to the same subspecies. Definition (18) can be splitted on two partial polarizations $\mathbf{P}=\mathbf{P}_A+\mathbf{P}_B$

$$\mathbf{P}_A(\mathbf{r},\,t)=\int\Psi^{\dagger}(R,\,t)\sum_{i\in A}\delta(\mathbf{r}-\mathbf{r}_i)(\hat{\mathbf{d}}_i\Psi(R,\,t))dR, \tag{20}$$

with $\hat{d}^{\alpha}_{i\in A}=\sum_{j\in B}\Pi^{\alpha}_{ij}(r_{ij})(\hat{\mathbf{s}}_i\cdot\hat{\mathbf{s}}_j)$, and

$$\mathbf{P}_B(\mathbf{r},\,t)=\int\Psi^{\dagger}(R,\,t)\sum_{i\in B}\delta(\mathbf{r}-\mathbf{r}_i)(\hat{\mathbf{d}}_i\Psi(R,\,t))dR, \tag{21}$$

with $\hat{d}^{\alpha}_{i\in B}=\sum_{j\in A}\Pi^{\alpha}_{ij}(r_{ij})(\hat{\mathbf{s}}_i\cdot\hat{\mathbf{s}}_j)$. In order to make the splitting given by equations (20) and (21), we specify the subspecies $s=A$ or $s=B$ in operator (18).

We can calculate $\mathbf{P}_A$ and $\mathbf{P}_B$ separately

$$\begin{aligned}P^{\alpha}_A=&\,g^{\alpha}_{0\Pi,AB}\mathbf{S}_A\mathbf{S}_B+\frac{1}{3}\frac{1}{2^3}g^{\alpha}_{\Pi}[\triangle(\mathbf{S}_A\mathbf{S}_B)\\&+2\partial_{\mu}[S^{\nu}_A\partial_{\mu}S^{\nu}_B-S^{\nu}_B\partial_{\mu}S^{\nu}_A]\\&+S^{\nu}_A(\triangle S^{\nu}_B)+S^{\nu}_B(\triangle S^{\nu}_A)-2(\partial_{\mu}S^{\nu}_A)(\partial_{\mu}S^{\nu}_B)],\end{aligned} \tag{22}$$

and

$$\begin{aligned}P^{\alpha}_B=&\,g^{\alpha}_{0\Pi,AB}\mathbf{S}_A\mathbf{S}_B+\frac{1}{3}\frac{1}{2^3}g^{\alpha}_{\Pi}[\triangle(\mathbf{S}_A\mathbf{S}_B)\\&+2\partial_{\mu}[S^{\nu}_B\partial_{\mu}S^{\nu}_A-S^{\nu}_A\partial_{\mu}S^{\nu}_B]\\&+S^{\nu}_A\triangle S^{\nu}_B+S^{\nu}_B\triangle S^{\nu}_A-2(\partial_{\mu}S^{\nu}_A)(\partial_{\mu}S^{\nu}_B)].\end{aligned} \tag{23}$$

Equations (22) and (23) have similar structure. The difference between them is in the replacement of subindexes $A\leftrightarrow B$. We obtain the major contribution appearing from the first order of the expansion on the relative distance. It is indicated by the coefficient $g^{\alpha}_{0\Pi,AB}$. We also find the correction to the major term. These corrections contain the coordinate derivatives of the spin density. These terms are indicated by the coefficient $g^{\alpha}_{\Pi,AB}$. The definitions of $g^{\alpha}_{0\Pi,AB}$ and $g^{\alpha}_{\Pi,AB}$ are identical to the definitions of $g^{\alpha}_{0\Pi}$ and g^{α}_{Π} presented after equation (17).

Next, we combine equations (22) and (23) to get an expression for polarization of the whole system

$$P^{\alpha}=2g^{\alpha}_{0\Pi,AB}\mathbf{S}_A\mathbf{S}_B+\frac{1}{6}g^{\alpha}_{\Pi,AB}(S^{\nu}_A\triangle S^{\nu}_B+S^{\nu}_B\triangle S^{\nu}_A). \tag{24}$$

Equation (24) shows that the direction of polarization is not related to the direction of the spin or the direction of change of the spin density in space. The direction is defined by the coefficients $g^{\alpha}_{0\Pi,AB}$ and $g^{\alpha}_{\Pi,AB}$. Below, we show that both constants have the same direction. It is the direction of the shift of the nonmagnetic ion from the line of the magnetic ions.

We also represent the major term in the polarization of via vectors $\mathbf{L}$ and $\mathbf{\Sigma}$:

$$P^{\alpha}=\frac{1}{2}g^{\alpha}_{0\Pi,AB}(\mathbf{\Sigma}^2-\mathbf{L}^2)\approx-\frac{1}{2}g^{\alpha}_{0\Pi,AB}\mathbf{L}^2. \tag{25}$$

4. Derivation of spin current model

4.1. The spin current model as the consequence of the momentum balance equation with spin-orbit interaction

It is possible to use the electric dipole moment operator (15) for the further derivation of the macroscopic polarization evolution equation as an addition to the Landau–Lifshitz–Gilbert equation for the study of perturbations and structures in the multiferroics. However, we are going to derive and justify the electric dipole moment operator (15) starting from the quantum microscopic theory. Therefore, we consider the many-particle Pauli equation (4) with the following Hamiltonian

$$\hat{H} = \sum_{i=1}^{N}\Bigg[-\hat{\mathbf{d}}_i \cdot \mathbf{E}_i - \hat{\boldsymbol{\mu}}_i \cdot \mathbf{B}_i - \frac{1}{2mc}(\hat{\boldsymbol{\mu}}_i \cdot [\mathbf{E}_i \times \hat{\mathbf{p}}_i]) - \frac{1}{2}\sum_{j=1,j\neq i}^{N}(U_{ij}\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j + \mathbf{D}_{ij} \cdot [\hat{\mathbf{s}}_i \times \hat{\mathbf{s}}_j])\Bigg], \tag{26}$$

where m is the mass of atom/ion, c is the speed of light in the vacuum, $\hat{\mathbf{d}}_i$ is the electric dipole moment operator, being defined via the displacement of ions with different charges, its relation to the spins of ions will be found below, $\mathbf{E}_i$ is the electric field, acting on the i-th dipole, $U_{ij} = U(\mathbf{r}_i - \mathbf{r}_j)$ is the exchange integral of the Heisenberg Hamiltonian as the function of the interparticle distance, $\mathbf{D}_{ij}$ is the Dzylaoshinskii vector constant. The Dzylaoshinskii vector has the structure related to the relative position of two magnetic ions and one nonmagnetic ion (the ligand ion) [34, 35]. It can be presented via the vector product of the radius-vectors of magnetic ions relatively nonmagnetic ion [34]. Overwise, it can be presented as the vector product of the relative position of two magnetic ions and the shift of the nonmagnetic ion from the line connecting two magnetic ions [35] $\mathbf{D}_{ij} \sim \mathbf{r}_{ij} \times \boldsymbol{\delta}$. This simple formula is useful, if we discuss one cell. However, if we consider the whole crystal macroscopically, we need to specify that we consider two neighboring ions. Hence, we introduce a function, which decreases (drops to zero) at the distances beyond the period of the crystal cell. So, we have the following structure $\mathbf{D}_{ij} = \beta(r_{ij})\mathbf{r}_{ij} \times \boldsymbol{\delta}$.

The single-ion anisotropy is considered in [3] (see p. 34, equations (39), (40), but we do not include it in our model at this stage. Equation (41) of [3] also presents the biquadratic interaction (see also the model in [36]), which is partially considered within our model for the spin evolution equation [37].

Let us also describe the physical meaning of terms in the Hamiltonian (26). The first term is the action of the electric field on the electric dipole moment. The second term is the action of the magnetic field on the magnetic moment. The third term is the spin-orbit interaction showing the action of the electric field on the moving magnetic moment. The fourth term is the Coulomb exchange interaction presented by the Heisenberg Hamiltonian. The last term is the Dzylaoshinskii-Moriya interaction.

It is essential to specify that the electric dipole moment is related to the group of magnetic and nonmagnetic ions. However, in section 3, we contracted the electric dipole moment operator associated with each magnetic ion. This meaning of the operator is implicated in the Hamiltonian.

4.2. The spin current model in ferromagnetic materials

Here, we derive the macroscopic polarization corresponding to the dipole moment operator (15) via the spin current model. Here, we also show that the spin current model follows from the momentum balance equation. Therefore, we derive the momentum balance equation corresponding to the Hamiltonian (26).

To derive the momentum balance equation, we need to define the momentum density via the many-particle wave function as the quantum average of the momentum operator of each particle

$$\mathbf{p}(\mathbf{r},\, t) = \frac{1}{2}\int\sum_i \delta(\mathbf{r} - \mathbf{r}_i)(\Psi_S^{\dagger}(R,\, t)\hat{\mathbf{p}}_i\Psi_S(R,\, t) + h.c.)dR, \tag{27}$$

where $h.\,c.$ is the Hermitian conjugation, and $\mathbf{p}_i = -\imath\hbar\nabla_i$ is the momentum operator of i-th particle.

We consider the time derivative of the momentum density (27). The time derivative acts on the wave function, while the time derivative of the wave function is replaced with Hamiltonian in accordance with the Pauli equation

$$\partial_t\mathbf{p}(\mathbf{r},\, t) = \frac{1}{2}\frac{\imath}{\hbar}\int\sum_i \delta(\mathbf{r} - \mathbf{r}_i) \times (\Psi^{\dagger}(R,\, t)[\hat{H},\, \hat{\mathbf{p}}_i]\Psi(R,\, t) + h.c.)dR. \tag{28}$$

Further calculation depends on the explicit form of the Hamiltonian. Moreover, different interaction terms are considered in different approximations. The first three terms in the Hamiltonian (26) can be considered straightforwardly. However, two last terms can be considered with the account of the short-range nature of these interactions. It leads to the following momentum balance equation

$$\partial_t\mathbf{p} = P^{\beta}\nabla E^{\beta} + \gamma S^{\beta}\nabla B^{\beta} + \frac{\gamma}{2mc}\varepsilon^{\beta\gamma\delta}J^{\delta\gamma}(\nabla E^{\beta}) + g_{0u}S^{\beta}\nabla S^{\beta} + \mathbf{F}_{DM}, \tag{29}$$

where $g_{0u} = \int U(r)d\mathbf{r}$, $J^{\delta\gamma}$ is the spin-current tensor, P^{β} is the polarization or the electric dipole moment density (16), $\mathbf{F}_{DM}$ is the force density of the Dzylaoshinskii-Moriya interaction:

$$\mathbf{F}_{DM} = \frac{1}{3} g_{(\beta)}((\boldsymbol{\delta} \cdot \mathbf{S})\nabla(\nabla \cdot \mathbf{S}) - (\mathbf{S} \cdot \nabla)\nabla(\boldsymbol{\delta} \cdot \mathbf{S})), \tag{30}$$

with $g_{(\beta)} = \int \xi^2 \beta(\xi) d\boldsymbol{\xi}$. So, physical meaning of terms in equation is the same like in Hamiltonian (26), they are also placed in the same order.

Let us consider the stationary regime, where the momentum density does not change in time $\partial_t \mathbf{p} = 0$. Therefore, the right-hand side of the momentum balance equation (29) should be equal to zero. The balance of the second and fourth terms gives an equilibrium magnetic field formed by magnetic moments due to the exchange interaction $\mathbf{B} = -g_{0u}\mathbf{S}/\gamma$. The last term can be equal to zero in equilibrium if the spin polarization is perpendicular to the shift of the ligand ion from the line connecting neighboring magnetic ions $(\boldsymbol{\delta} \cdot \mathbf{S}) = 0$ or it can be equal to zero at the nontrivial balance of two terms in equation (30). However, our goal is to obtain the spin current model, which follows from the balance of the first and third terms. It can happen in the arbitrary electric field if polarization is balanced by the spin current appearing in the spin-orbit interaction

$$P^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} J^{\alpha\beta}. \tag{31}$$

We obtain the spin-current model of the polarization with no particular relation to the form of the spin-current. Hence, equation (31) can be applied for the derivation of the polarization caused by different mechanisms. The mean-field part of the spin-orbit interaction plays main role in the derivation of the spin current model, i.e. in the connection between the polarization and the antisymmentric part of the spin-current tensor. To the best of our knowledge we are not familiar with the contribution of the symmetric part of the spin current tensor in the polarization of multiferroics.

4.3. The spin current model in antiferromagnetic materials

The first three terms in the Hamiltonian (26) and the momentum balance equation (29) have the same form for the ferromagnetic and antiferromagnetic materials. A difference appears for the Coulomb exchange interaction and the Dzylaoshinskii-Moriya interaction. The force fields for these interactions are obtained in the following forms

$$\mathbf{F}_{HH,s} = g_{0u,ss'} S_s^{\beta} \nabla S_{s'}^{\beta}, \tag{32}$$

and

$$\mathbf{F}_{DM,s} = \frac{1}{3} g_{(\beta),ss'}((\boldsymbol{\delta} \cdot \mathbf{S}_s)\nabla(\nabla \cdot \mathbf{S}_{s' \neq s}) - (\mathbf{S}_s \cdot \nabla)\nabla(\boldsymbol{\delta} \cdot \mathbf{S}_{s' \neq s})). \tag{33}$$

Hence, we derive the momentum balance equation for each subspecies

$$\partial_t \mathbf{p}_s = P_s^{\beta} \nabla E^{\beta} + \gamma S_s^{\beta} \nabla B^{\beta} + \frac{\gamma}{2mc} \varepsilon^{\beta\gamma\delta} J_s^{\delta\gamma} (\nabla E^{\beta}) + \mathbf{F}_{HH,s} + \mathbf{F}_{DM,s}. \tag{34}$$

The electric dipole moment is related to the group of ions, but the operator definition is recontracted to the form of operator associated with each magnetic ion. Hence, we get the partial polarization of each subspecies from the momentum balance equation of each subspecies. It gives us the representation of the partial polarization via the partial spin current $P_s^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} J_s^{\alpha\beta}$. Complete polarization of the sample is the combination of partial polarizations $P^{\mu} = P_A^{\mu} + P_B^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} (J_A^{\alpha\beta} + J_B^{\alpha\beta})$. Therefore, equation (31) is reestablished for the antiferromagnetic materials.

4.4. Dzylaoshinskii-Moriya spin current and related polarization in ferromagnetic materials

We presented the derivation of the spin current model. It appears due to the relativistic spin-orbit interaction. Its further application requires an example of the spin current related to a specific physical mechanism. The explicit form of the spin-current tensor can be found in the spin evolution equation (Landau–Lifshitz–Gilbert equation). In this work, we focus on the justification of operator (15), which contains the vector coefficient of proportionality between the combination of the spin operators and the electric dipole moment operator. In Hamiltonian (26) we have two interactions containing inexplicitly defined space dependencies. They are the two last terms corresponding to the exchange Coulomb interaction given by the Heisenberg Hamiltonian and the Dzylaoshinskii-Moriya interaction, correspondingly. The Heisenberg Hamiltonian contains a scalar function, so we expect that it is not related to the considering regime. In contrast, the Dzylaoshinskii-Moriya interaction is proportional to the vector Dzylaoshinskii constant, so it can give a mechanism for the vector constant appearance in operator (15). To check the described suggestion, we need to consider the Dzylaoshinskii-Moriya interaction contribution in the spin evolution equation. Similarly to section 2, we use the quantum hydrodynamic method and include the short-range nature of the Dzylaoshinskii-Moriya interaction. As the

result, we find the partial contribution of the Dzylaoshinskii-Moriya interaction in the spin evolution equation

$$\partial_t \mathbf{S}_s = \mathbf{T}_{DM}, \tag{35}$$

where

$$\mathbf{T}_{DM} = \frac{1}{3} g_{(\beta)}((\mathbf{S}_s \cdot [\boldsymbol{\delta} \times \nabla])\mathbf{S}_s - S_s^{\beta}[\boldsymbol{\delta} \times \nabla] S_s^{\beta}), \tag{36}$$

with $g_{(\beta)} = \int \xi^2 \beta(\xi) d\boldsymbol{\xi}$.

Reference [12] is based on the total free energy with a nonexplicit account of the Dzylaoshinskii-Moriya interaction. Its explicit contribution is given in the Landau–Lifshitz-Gilbert equation [38] in the form of the torque $\mathbf{T} = \gamma \tilde{D} \mathbf{M} \times (\nabla \times \mathbf{M})$ (see equation (2) and text after equation (2) of [12]). The expression we obtain in this paper (36) shows a similar structure at the replacement $g_{(\beta)}[\boldsymbol{\delta} \times \nabla]$ on $\tilde{D}\nabla$. The difference partially appears due to the account of structure of the Dzylaoshinskii vector constant $\mathbf{D}_{ij} = \beta(r_{ij})\mathbf{r}_{ij} \times \boldsymbol{\delta}$. These expressions give different directions of the spin torque vector.

The spin wave is considered described in [12] as a small fluctuation of the static domain-wall profile. As a limit, a simple dispersion dependence is found for location away from the domain-wall center, where the magnetization is uniform in the domains (see equation (9) of [12]). The dispersion relations appears to be asymmetric outside the domain-wall in accordance with works [39, 40]. Let us specify that in [39] experimentally demonstrated (on an Fe double layer grown on W(110)) the Dzyaloshinskii-Moriya leads to an asymmetric spin-wave dispersion relation.

Reference [40] presents a translation of the Dzyaloshinskii-Moriya interaction Hamiltonian (like the last term in equation (26)) to a continuum model of the energy density with magnetization direction and symmetry breaking in the y-direction.

In order to compare our model with equation (2) of [40], we calculate the correlationless limit of the energy density. The definition of the energy density is

$$E_{DM}(\mathbf{r}, t) = -\frac{1}{2} \int \sum_{i,j=1, j\neq i}^{N} \delta(\mathbf{r} - \mathbf{r}_i) \times \Psi_S^{\dagger}(R, t) \mathbf{D}_{ij} \cdot [\hat{\mathbf{s}}_i \times \hat{\mathbf{s}}_j] \Psi_S(R, t) dR. \tag{37}$$

It gives us the following approximate expression

$$E_{DM} \approx \frac{1}{6} g_{(\beta)} \delta^{\alpha} (S^{\alpha}(\nabla \cdot \mathbf{S}) - (\mathbf{S} \cdot \nabla) S^{\alpha}), \tag{38}$$

where we use $\mathbf{D}_{ij} = \beta(r_{ij})\mathbf{r}_{ij} \times \boldsymbol{\delta}$. Let us repeat here the equation (2) [40] for the comparison $E_{DM} = -D[(\mathbf{m} \times \partial_x \mathbf{m})_z - (\mathbf{m} \times \partial_z \mathbf{m})_x]$, where $\mathbf{m}$ is the magnetization direction, $(\mathbf{b})_x$ is the x-projection of vector $\mathbf{b}$, and the y-direction is chosen in [40] as the anisotropy direction. For the fixed concentration we have $\mathbf{S} \sim \mathbf{m}$ (otherwise the change of concentration would give contribution in the derivatives of the spin density $\mathbf{S}$). We see rather different vector structures in these expressions. To continue our comparison, we would assume $\mathbf{D}_{ij} = \tilde{\beta}(r_{ij})\mathbf{r}_{ij}$. It leads to $E_{DM} = -(1/6) g_{(\tilde{\beta})} \mathbf{S} \cdot [\nabla \times \mathbf{S}]$, but it also differs from even if we assume that there is no dependence on the chosen direction y. The described difference can be caused by the different form of structure of the Dzyaloshinskii constant $\mathbf{D}_{ij}$. Our expression (38) corresponds to equation (2) in [25], up to the details of the coefficient. Here, we presented the derivation for the arbitrary three-dimensional sample, while the ultrathin films with perpendicular easy axis, grown on a substrate with a capping from the different material so that the structural inversion symmetry is broken along the film normal, are considered in [25].

The Dzylaoshinskii constant has a known particular numerical value for materials. In contrast, constants like $g_{(\beta)}$ are novel parameters. Hereby, we need to give some comment on the estimation of these parameters for better comparison with other works. To get an approximate expression for $g_{(\beta)}$ if we consider explicit form for the function $\beta(r)$. We choose a model expression for the function $\beta(r)$: $\beta(r) = (D_0/(4\pi a \delta))\theta(r - a)$, where a is the distance between magnetic ions, δ is the module of the vector of the ligand shift. It leads to $g_{(\beta)} = D_0 a^4/5\delta$. Similar analysis for the constant like g_{0u} is given in [28].

Equation (36) shows that the spin-torque caused by the Dzylaoshinskii-Moriya interaction cannot be represented as the divergence of the spin-current tensor. However, the second term in (36) can be represented in the required form $S_s^{\delta} \varepsilon^{\alpha\beta\gamma} \delta^{\beta} \nabla^{\gamma} S_s^{\delta} = \nabla^{\gamma}[(1/2)\varepsilon^{\alpha\beta\gamma}\delta^{\beta}\mathbf{S}_s^2]$. We can use this part for the calculation of the polarization. However, the first term in (36) gives some freedom in the interpretation of the spin current since it allows to get an additional term.

Let us start the analysis of the polarization using the simplest form of the Dzylaoshinskii-Moriya spin current $\mathbf{T}_{DM} = -\partial_{\beta} J_{DM}^{\alpha\beta}$

$$J_{DM}^{\alpha\beta} = -\frac{1}{6} g_{(\beta)} \varepsilon^{\alpha\beta\gamma} \delta^{\gamma} \mathbf{S}_s^2. \tag{39}$$

It leads to the following form of the macroscopic polarization

$$P_{DM}^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} J_{DM}^{\alpha\beta} = -\frac{1}{6} \frac{\gamma}{mc} g_{(\beta)} \delta^{\mu} \mathbf{S}_s^2, \tag{40}$$

which corresponds to the first term on the right-hand side of equation (17), which is derived from operator (13). Let us to point out that the collinear spin structure is formed by the Heisenberg exchange interaction, while further formation of spin related electric polarization is formed by the combination of the spin-orbit and the Dzyaloshinskii-Moriya interactions.

As a matter of discussion, let us represent the spin torque (36) in a form, where the spin current is extracted from the first term in addition to the spin current following from the second term

$$\mathbf{T}_{DM} = -\frac{1}{3} g_{(\beta)} ((\mathbf{S}_s (\boldsymbol{\delta} \cdot [\nabla \times \mathbf{S}_s]) + \nabla^{\beta} \{[\boldsymbol{\delta} \times \mathbf{S}_s]^{\beta} \mathbf{S}_s\} + \frac{1}{2} [\boldsymbol{\delta} \times \nabla] \mathbf{S}_s^2 \Big). \tag{41}$$

It shows that we can choose the spin current in the following alternative form

$$\tilde{J}_{DM}^{\alpha\beta} = \frac{1}{3} g_{(\beta)} \delta^{\gamma} \left[S_s^{\alpha} \varepsilon^{\beta\gamma\delta} S_s^{\delta} - \frac{1}{2} \varepsilon^{\alpha\beta\gamma} \mathbf{S}_s^2 \right]. \tag{42}$$

This extended spin current leads to the following form of polarization

$$\tilde{P}_{DM}^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} \tilde{J}_{DM}^{\alpha\beta} = -\frac{1}{6} \frac{\gamma}{mc} g_{(\beta)} (\boldsymbol{\delta} \cdot \mathbf{S}_s) S_s^{\mu}. \tag{43}$$

This form of the polarization completely differs from the structure following from the well-known electric dipole moment of the pair of ions (13) and (17). We want to mention that the spin torque following from the Heisenberg exchange interaction appears as the divergence of the spin current tensor with no additional terms.

In the comparison of expressions (17) and (40), we see that they are found in different approximations. Polarization (40) is obtained in the main order of the expansion, while polarization (17) contains corrections related to the second space derivative of the spin density. To complete our comparison, we can find corrections to the spin-torque caused by the Dzylaoshinskii-Moriya interaction (36) in the next order of expansion. Our calculation gives the following spin-torque

$$T_{DM,3}^{\mu} = \frac{1}{30} g_{2(\beta)} [(\mathbf{S}_s \cdot [\boldsymbol{\delta} \times \nabla]) \triangle \mathbf{S}_s - S_s^{\beta} [\boldsymbol{\delta} \times \nabla] \triangle S_s^{\beta}], \tag{44}$$

with $g_{2(\beta)} = \int \xi^4 \beta(\xi) d\boldsymbol{\xi}$.

We need to extract a part of the spin torque, which appears as the divergence of the spin current $\partial^{\beta} J^{\alpha\beta}$. In equation (36) we used the term containing the scalar product of the spin densities $S_s^{\beta} [\boldsymbol{\delta} \times \nabla] S_s^{\beta}$ (the second term on the right-hand side). In equation (44) we follow the same approach and consider the term containing the scalar product of the spin densities $S_s^{\beta} [\boldsymbol{\delta} \times \nabla] \triangle S_s^{\beta}$ (the second term on the right-hand side). However, the term under consideration does not appear as the divergence of the second rank tensor. We need to split it into two parts $S_s^{\beta} [\boldsymbol{\delta} \times \nabla] \triangle S_s^{\beta} = [\boldsymbol{\delta} \times \nabla] (S_s^{\beta} \triangle S_s^{\beta}) - (\triangle S_s^{\beta}) [\boldsymbol{\delta} \times \nabla] S_s^{\beta}$ and use the first of them to get the effective spin current

$$J_{DM3}^{\alpha\beta} = -\frac{1}{5} \frac{1}{6} g_{2(\beta)} \varepsilon^{\alpha\beta\gamma} \delta^{\gamma} (\mathbf{S}_s \cdot \triangle \mathbf{S}_s). \tag{45}$$

It leads to the following polarization

$$P_{DM}^{\mu} = \frac{\gamma}{2mc} \varepsilon^{\mu\alpha\beta} J_{DM}^{\alpha\beta} = -\frac{1}{6} \frac{1}{5} \frac{\gamma}{mc} g_{2(\beta)} \delta^{\mu} (\mathbf{S}_s \cdot \triangle \mathbf{S}_s). \tag{46}$$

We see the spin structure in polarization (46) corresponds to the spin structure in the second term on the right-hand side in equation (17). Hence, we justify equation (17) found from the electric dipole moment (13) using the spin current model with the spin-current related to the Dzylaoshinskii-Moriya interaction. Moreover, our calculations in two major orders of expansion give the interpretation of the direction of the vector constant $\Pi^{\alpha}(\xi)$. We see that it is parallel to the shift of the ligand ion from the line connecting neighboring magnetic ions δ^{α}. The complite polarization is also parallel to this direction. It also corresponds to the microscopic meaning of the electric dipole moment as a shift of ions of opposite charges.

In section 3, we found the macroscopic polarization (17) corresponding to the electric dipole moment (13). Here, we found macroscopic polarization using the momentum balance equation with the spin-orbit interaction and the spin-current caused by the Dzylaoshinskii-Moriya interaction. These expressions have the same

structure. It allows us to give a physical interpretation of the vector constant in the electric dipole moment (13). Let us compare the polarization given by the first term in equation (17) with equation (40). It gives the following relation $g^{\alpha}_{0\Pi}\mathbf{S}_s^2 = -\frac{1}{6}\frac{\gamma}{mc}g_{(\beta)}\delta^{\mu}\mathbf{S}_s^2$, where we can drop the square of the spin density $\mathbf{S}_s^2$ and compare the coefficients. Basically, we need to compare the functions under the integrals. We have two options. First, we equate the functions under integrands and find $\Pi^{\alpha}(\xi) = -(1/6)(\gamma/mc)\xi^2\beta(\xi)\delta^{\alpha}$. Second, we transform the left-hand side by integration by parts, so we obtain $g^{\alpha}_{0\Pi} = -(1/3)\int\xi(\partial\Pi^{\alpha}(\xi)/\partial\xi)d^3\xi$. Next, we equate the functions under integrands and obtain $\partial\Pi^{\alpha}(\xi)/\partial\xi = (\gamma/2mc)\xi\beta(\xi)\delta^{\alpha}$.

We found a relation between the empirically introduced function $\Pi^{\alpha}(\xi)$, which is the coefficient of proportionality in the electric dipole moment (13) and the function $\beta(\xi)$ appearing in the Dzylaoshinskii-Moriya interaction. The Dzylaoshinskii-Moriya interaction is the exchange part of the spin-orbit interaction, and the coefficient $\mathbf{D}_{ij}$ is the exchange integral. It is similar to the exchange integral in the Heisenberg exchange interaction, where the exchange part of the Coulomb interaction is considered.

In order to prove the found expression for function $\Pi^{\alpha}(\xi)$ we considered the next order of expansion for the polarization definition (17) and the Dzylaoshinskii-Moriya spin-torque giving the effective spin-current and corresponding polarization (46). Hence, we compare the second term on the right-hand side of equation (17) with polarization (46). Dropping equal spin structures, we find $\frac{1}{6}g^{\alpha}_{\Pi} = -\frac{1}{6}\frac{1}{5}\frac{\gamma}{mc}g_{2(\beta)}\delta^{\mu}$. Here we need to compare the functions under integral and prove the relation between $\partial\Pi^{\alpha}(\xi)/\partial\xi$ and $\beta(\xi)$ obtained above. We transform the left-hand side by integration by parts, so we obtain $g^{\alpha}_{\Pi} = -(1/5)\int\xi^3(\partial\Pi^{\alpha}(\xi)/\partial\xi)d^3\xi$. It leads to relation

$$\frac{\partial\Pi^{\alpha}(\xi)}{\partial\xi} = \frac{\gamma}{2mc}\xi\beta(\xi)\delta^{\alpha}, \tag{47}$$

which is presented above from the first order of expansion.

The spin torque caused by the Heisenberg exchange interaction can be presented as the divergence of the corresponding spin current tensor [29] $g_u\varepsilon^{\alpha\beta\gamma}S_s^{\beta}\triangle S_s^{\gamma} = \partial_{\delta}(g_u\varepsilon^{\alpha\beta\gamma}S_s^{\beta}\partial_{\delta}S_s^{\gamma}) = -\partial_{\delta}J^{\alpha\delta}_{HH}$. This spin current tensor can be placed in the polarization obtained in the spin current model $P^{\mu} = \frac{\gamma}{2mc}\varepsilon^{\mu\alpha\beta}J^{\alpha\beta}$. It gives polarization coinciding with the result of Mostovoy [7]. The method demonstrated in section 3 can be applied to the operator $\mathbf{d}_{ij} = \alpha_{ij}[\mathbf{r}_{ij}\times[\mathbf{s}_i\times\mathbf{s}_j]]$ in order to rederive the result of Mostovoy [7]. So, we can conclude that this result follows from the Heisenberg exchange interaction. This comment is placed here for comparison with the results of our paper.

4.5. Dzylaoshinskii-Moriya spin current and related polarization in antiferromagnetic materials

In this section, we need to consider the spin-torque caused by the Dzylaoshinskii-Moriya interaction. Particularly, we need to consider the interaction between different subspecies in the antiferromagnetic samples. Our calculations give the following form of the s subspecies spin evolution equation under the Dzylaoshinskii-Moriya interaction with s' subspecies:

$$\partial_t\mathbf{S}_s = \mathbf{T}_{DM,s'\neq s}, \tag{48}$$

where

$$\mathbf{T}_{DM,s'\neq s} = \frac{1}{3}g_{(\beta)AB}((\mathbf{S}_s\cdot[\boldsymbol{\delta}\times\nabla])\mathbf{S}_{s'\neq s} - S_s^{\beta}[\boldsymbol{\delta}\times\nabla]S^{\beta}_{s'\neq s}). \tag{49}$$

The general structure of the obtained spin torque is similar to the torque existing under interaction of the ions of the same subspecies (36). However, there is an essential difference related to the appearance of two kinds of subindexes, s and s'. So, we find no term, which can be rewritten as the spin current.

In order to solve the described problem, we suggest the following step. We need to consider the sum of spin torques $\mathbf{T}_{DM,A}$ and $\mathbf{T}_{DM,B}$ instead of the sum of partial spin currents:

$$\begin{aligned}\mathbf{T}_{DM} = \frac{1}{3}g_{(\beta)AB}&((\mathbf{S}_A\cdot[\boldsymbol{\delta}\times\nabla])\mathbf{S}_B\\ &+(\mathbf{S}_B\cdot[\boldsymbol{\delta}\times\nabla])\mathbf{S}_A - [\boldsymbol{\delta}\times\nabla](S_A^{\beta}S_B^{\beta})).\end{aligned} \tag{50}$$

So, a part of combined spin torque can be presented as a 'combined' spin current. So, we would be able to derive the spin polarization of the full system instead of partial polarizations. Anyway, the partial polarizations are intermediate theoretical constructions, which have no physical meaning since the polarization formation is related to magnetic ions of both subspecies (and ions of the nonmagnetic subspecies). Similarly to the last term in equation (36), we see that the last term in equation (50) gives us the effective spin current caused by the intersubspecies Dzylaoshinskii-Moriya interaction

$$J^{\alpha\beta}_{DM} = -\frac{1}{3}g_{(\beta)AB}\varepsilon^{\alpha\beta\gamma}\delta^{\gamma}(\mathbf{S}_A\cdot\mathbf{S}_B). \tag{51}$$

It leads to the polarization of antiferromagnetic materials

$$P^{\mu}_{DM} = \frac{\gamma}{2mc}\varepsilon^{\mu\alpha\beta}J^{\alpha\beta}_{DM} = -\frac{1}{3}\frac{\gamma}{mc}g_{(\beta)AB}\delta^{\mu}(\mathbf{S}_A \cdot \mathbf{S}_B). \quad (52)$$

4.6. On the other types of mechanisms of the polarization formation

In this paper we are focused on the contribution of the Dzylaoshinskii-Moriya spin current in the spin current model in order to derive the symmetric form of the electric polarization in multiferroics. However, this is one of three known forms of the polarization. One of them is the antisymmetric form of the electric polarization. Corresponding polarization can be obtained via the magnon spin current existing due to the Heisenberg exchange interaction (it is briefly described in the final part of section 4.4.). The biquadrtic exchange can give a correction to this result. The third type of the polarization is the spin dependent p-d hybridization mechanism. At the current state of our research we cannot suggest any interaction which provides a suitable spin current. Here we demonstrated that the spin current model can be applied for the symmetric form of the electric polarization in addition to the well-known explanation of the antisymmetric form of polarization, but it is not extended to the third mechanism of the polarization formation.

5. Polarization evolution equation

For the derivation of the polarization evolution equation, we use the Hamiltonian (5), where we include no relativistic interactions. The polarization itself is caused by the relativistic effects, hence, relativistic interactions (the spin-orbit interaction, the Dzylaoshinskii-Moriya interaction, and the evolution of the dipole moment under the action of the external electric field due to the relativistic nature of the electric dipole moment) give the relatively small effect.

In order to derive the polarization evolution equation, we consider the definition of the polarization in terms of the microscopic many-particle wave function (16), with operator (15). We consider the time derivative of this definition. The time derivative acts on the wave functions under the integral. We find for Hamiltonian (5) the following intermediate form of the polarization evolution equation

$$\partial_t \mathbf{P}(\mathbf{r}, t) = \frac{\imath}{\hbar}\int \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\Psi^{\dagger}(R, t)[\hat{H}, \hat{\mathbf{d}}_i]\Psi(R, t)dR. \quad (53)$$

The first term contains dependence on two particles i and j in the functions $\delta(\mathbf{r} - \mathbf{r}_i)$, B_i^{α} and Π_{ij}^{α} placed under the integral in equation (53). In the following calculations of this term, we need to include the strong decrease of function Π_{ij}^{α} with the increase of the interparticle distance. We need to make the transition to relative interparticle distance similarly to section 3, where we made analysis of the definition of polarization (16).

The second term has a more complex structure. It depends on three particles i, j and k in the functions placed under the integral in equation (53). Therefore, we need to introduce the center of mass and the relative distances for three particles. In our calculations, we use the following substitution $\mathbf{r}_i = \mathbf{R}_{ijn} + (2/3)\mathbf{r}_{in} - (1/3)\mathbf{r}_{jn}$, $\mathbf{r}_j = \mathbf{R}_{ijn} - (1/3)\mathbf{r}_{in} + (2/3)\mathbf{r}_{jn}$, and $\mathbf{r}_n = \mathbf{R}_{ijn} - (1/3)\mathbf{r}_{in} - (1/3)\mathbf{r}_{jn}$, where $\mathbf{R}_{ijn} = (\mathbf{r}_i + \mathbf{r}_j + \mathbf{r}_n)/3$, $\mathbf{r}_{in} \equiv \mathbf{r}_1 = \mathbf{r}_i - \mathbf{r}_n$, $\mathbf{r}_{jn} \equiv \mathbf{r}_2 = \mathbf{r}_j - \mathbf{r}_n$, and $\mathbf{r}_{ij} \equiv \mathbf{r}_3 = \mathbf{r}_1 - \mathbf{r}_2$. It leads to the change of the element of volume in the configuration space $dR = dR_{N-3}d\mathbf{R}_{ijk}d\mathbf{r}_{in}d\mathbf{r}_{jn}$. We use these substitutions in the delta function $\delta(\mathbf{r} - \mathbf{r}_i)$ and in the many-particle wave function $\Psi(R, t) = \Psi(...,\mathbf{r}_i,...,\mathbf{r}_j,...,\mathbf{r}_n,...,t)$.

After the described change of notations under the integral in equation (53) we make an expansion on the relative distances. It is possible due to the strong dependence of functions Π_{ij}^{α} and $U(r_{kj})$ on the relative distance. Below, we present the results of our calculations for two regimes: the ferromagnetic materials and the antiferromagnetic materials.

5.1. Polarization evolution for the ferromagnetic materials

In this subsection, we present the results of our derivation of the polarization evolution for the ferromagnetic materials

$$\partial_t P^{\alpha} = \frac{1}{3}\gamma g_{\Pi}^{\alpha}\varepsilon^{\beta\gamma\delta}(\partial^{\mu}B^{\beta})S^{\gamma}\nabla^{\mu}S^{\delta} + G^{\alpha}(\mathbf{S} \cdot [\nabla^{\mu}\mathbf{S} \times \partial^{\mu}\triangle\mathbf{S}]), \quad (54)$$

where the first term is the contribution of the Zeeman energy (see the first term in Hamiltonian (5)), it is obtained in this paper, the last term is caused by the Coulomb exchange interaction and obtained in [2]. The following notations are used in equation (54): γ is the gyromagnetic ratio, $\hat{\mu}_i^{\alpha} = \gamma\hat{s}_i^{\alpha}$ is the magnetic moment, and the vector interaction constant

$$G^{\alpha} = \frac{1}{3!}\frac{1}{3} g_u g_{\Pi}^{\alpha}, \tag{55}$$

which is a combined interaction constant with $g_u = \int r^2 U(r) d^3 r$, $g_{\Pi}^{\alpha} = \int r^2 \Pi^{\alpha}(r) d^3 r$. Here we see one interaction constant related to the exchange integral $g_u = \int r^2 U(r) d^3 r$, and the interaction constant related to the function describing formation of the electric dipole moment $g_{\Pi}^{\alpha} = \int r^2 \Pi^{\alpha}(r) d^3 r$. The contribution of the Zeeman energy in the polarization evolution equation, in the regime, where the electric dipole moment is proportional to the vector product of the spin operators, is found in [1]. It is related to another mechanism of the polarization formation in the multiferroic material.

5.2. Polarization evolution for the antiferromagnetic materials

In this section, we consider the time evolution of polarization given by equation (20), which includes the structure of antiferromagnetic materials. Here we have two subspecies A and B, so we derive the polarization evolution equation for each of them. Let us present the result for one of the subspecies

$$\begin{aligned}\partial_t P_A^{\alpha} = {} & \frac{1}{6}\varepsilon^{\beta\gamma\delta} g_{\Pi}^{\alpha} \gamma (\partial^{\mu} B^{\gamma}) [S_B^{\delta} \partial^{\mu} S_A^{\beta} - S_A^{\beta} \partial^{\mu} S_B^{\delta} - \partial^{\mu}(S_A^{\beta} S_B^{\delta})] \\ & + \frac{1}{6}\varepsilon^{\beta\gamma\delta} [(g_{\Pi}^{\alpha} g_{0u} + 2 g_{0\Pi}^{\alpha} g_u) S_B^{\beta} S_A^{\gamma} \triangle S_A^{\delta} \\ & + (-g_{\Pi}^{\alpha} g_{0u} + 2 g_{0\Pi}^{\alpha} g_u) S_A^{\beta} S_B^{\gamma} \triangle S_B^{\delta} \\ & + 2 g_{\Pi}^{\alpha} g_{0u} (\partial^{\mu} S_B^{\beta}) S_A^{\gamma} \partial^{\mu} S_A^{\delta}].\end{aligned} \tag{56}$$

The first term in this equation is proportional to the space derivative of the magnetic field ($\partial^{\mu} B^{\gamma}$). It appears from the Zeeman energy (the first term in Hamiltonian (5)), like the first term in equation (54) obtained for the ferromagnetic regime. Other terms in equation (56) contain the interaction constants of the Heisenberg-Coulomb exchange interaction, since their appearance is caused by this interaction from the second term in Hamiltonian (5). The result for the second subspecies can be obtained via the exchange of subindexes $A \leftrightarrow B$.

It has been mentioned above that the polarization appears in the complex of two neighboring ions that belong to different subspecies. So, the partial polarization (20) and equation for its evolution (56) are the intermediate theoretical tools. We need to combine the partial polarizations in the full polarization $\partial_t P^{\alpha} = \partial_t P_A^{\alpha} + \partial_t P_B^{\alpha}$ and obtain the equation for its evolution

$$\begin{aligned}\partial_t P^{\alpha} = {} & -\frac{1}{3}\varepsilon^{\beta\gamma\delta} g_{\Pi}^{\alpha} \gamma (\partial^{\mu} B^{\gamma}) \partial^{\mu} (S_A^{\beta} S_B^{\delta}) \\ & + \frac{1}{3}\varepsilon^{\beta\gamma\delta} [2 g_{0\Pi}^{\alpha} g_u S_B^{\beta} S_A^{\gamma} \triangle (S_A^{\delta} - S_B^{\delta}) \\ & + g_{\Pi}^{\alpha} g_{0u} (\partial^{\mu} S_B^{\beta}) (S_A^{\gamma} - S_B^{\gamma}) \partial^{\mu} S_A^{\delta}],\end{aligned} \tag{57}$$

where the meaning of different terms is the same as the physical meaning of the terms in equation (56).

The spin evolution equations (10) are combined in the evolution equations for functions $\mathbf{\Sigma} = \mathbf{S}_A + \mathbf{S}_B$ and $\mathbf{L} = \mathbf{S}_A - \mathbf{S}_B$, which are traditionally used in the theory of the antiferromagnetic materials [29]. Therefore, it is essential to represent equation (57) in terms of these functions

$$\begin{aligned}\partial_t P^{\alpha} = {} & -\frac{1}{6}\varepsilon^{\beta\gamma\delta} g_{\Pi}^{\alpha} \gamma (\partial^{\mu} B^{\gamma}) \partial^{\mu} [L^{\beta} \Sigma^{\delta}] \\ & + \frac{1}{6}\varepsilon^{\beta\gamma\delta} [2 g_{0\Pi}^{\alpha} g_u \Sigma^{\beta} L^{\gamma} \triangle L^{\delta} + g_{\Pi}^{\alpha} g_{0u} (\partial^{\mu} \Sigma^{\beta}) L^{\gamma} \partial^{\mu} L^{\delta}],\end{aligned} \tag{58}$$

where we include $\varepsilon^{\beta\gamma\delta}[(\Sigma^{\beta} + L^{\beta})(\Sigma^{\delta} - L^{\delta})] = 2\varepsilon^{\beta\gamma\delta} L^{\beta} \Sigma^{\delta}$. Physical meaning of terms in the obtained equations can be traced via the coefficients. The presence of the magnetic field shows the appearance of this term from the Zeeman energy, and the presence of g_{0u} or g_u shows its appearance from the exchange interaction, similarly to the equations shown above.

6. Equilibrium solutions set of spin-polarization evolution equations

Let us consider equilibrium structures obeying a system of equations for the spin evolution and polarization evolution. For the simplicity of derivation of the equation obtained above, we considered the external magnetic field. It is possible to include the magnetic field created by the magnetic moments of the medium. Hence, we need to include the Maxwell equations: $\nabla \cdot \mathbf{B} = 0$ and $\nabla \times \mathbf{B} = 4\pi\gamma \nabla \times \mathbf{S}$, where we included the zero time derivative of the electric field due to our focus on the static regime.

6.1. Ferromagnetic multiferroics

We start this analysis with the ferromagnetic materials. In this case, we consider several configurations of the spin density.

6.1.1. Parallel spins, transverse change of spin magnitude

Let us consider the regime, where we consider the spin density directed in the z-direction $\mathbf{S}_0 = S_0\mathbf{e}_z$. We also assume that its module changes in the x-direction $S_0 = S_0(x)$. Since we assume $\partial_t\mathbf{S}_0 = 0$, we need to check that the right-hand side of the spin evolution equation

$$\partial_t\mathbf{S} = \gamma[\mathbf{S}\times\mathbf{B}] + \frac{1}{6}g_u[\mathbf{S}\times\triangle\mathbf{S}] \tag{59}$$

is equal to zero. We see $\triangle\mathbf{S}\|\mathbf{e}_z$, hence the last term is equal to zero. To check the first term, we need to find the corresponding magnetic field, assuming that the external magnetic field is equal to zero. We find $\nabla\times\mathbf{S}_0 = -(\partial_x S_0(x))\mathbf{e}_y$. Hence, $\nabla\times\mathbf{B}_0 = -4\pi\gamma(\partial_x S_0(x))\mathbf{e}_y$. It gives $\mathbf{B}_0 = -4\pi\gamma S_0(x)\mathbf{e}_z$. The condition $\nabla\times\mathbf{B}_0 = 0$ is also satisfied. It is parallel to $\mathbf{S}_0$, so the first term in equation (59) is equal to zero as well.

The equilibrium condition means that the polarization does not depend on time. Hence, the right-hand side of equation (54). Estimations given above show that this condition is satisfied. We also need to find the corresponding polarization (nonzero value) via the first term on the right-hand side of equation (17): $\mathbf{P}_0 = \mathbf{g}_{0\Pi}\mathbf{S}_0^2$. Let us remind that constant $\mathbf{g}_{0\Pi}$ is parallel to the shift of the ligand ion $\boldsymbol{\delta}$. We present a simple equilibrium spin structure leading to nonzero polarization.

6.1.2. Parallel spins, longitudinal change of spin magnitude

We consider the spin density directed in the z-direction $\mathbf{S}_0 = S_0\mathbf{e}_z$, where its module changes in the z-direction $S_0 = S_0(z)$ as well. We require $\partial_t\mathbf{S}_0 = 0$ and check the value of the right-hand side of the spin evolution equation (59). We see $\triangle\mathbf{S}\|\mathbf{e}_z$, hence the last term is equal to zero. We also find the zero magnetic field $\mathbf{B} = 0$. Hence, both equations (54) and (59) are satisfied. We also find the corresponding polarization $\mathbf{P}_0(z) = \mathbf{g}_{0\Pi}\mathbf{S}_0^2(z)$.

6.1.3. Cycloidal spiral spin structure

Let us consider the spiral spin structure that was earlier presented in works [7, 41]:

$$\mathbf{S}_0(\mathbf{r}) = s_b\mathbf{e}_y\cos(\mathbf{r}\cdot\mathbf{q}) + s_c\mathbf{e}_z\sin(\mathbf{r}\cdot\mathbf{q}) + s_a\mathbf{e}_x, \tag{60}$$

where $\mathbf{q} = q\mathbf{e}_y$. It is a spiral shifting in the direction being in the rotation plane. It can be represented in the following form

$$\mathbf{S}_0(y) = s_y\mathbf{e}_b\cos(yq) + s_c\mathbf{e}_z\sin(yq) + s_a\mathbf{e}_x, \tag{61}$$

which can be substituted in the spin evolution equation to find the magnetic field corresponding to the equilibrium condition.

The right-hand side of equation (59) should be equal to zero for the static regime. In addition to the magnetic field parallel $\mathbf{B}_1 = \chi\mathbf{S}_0$ to the equilibrium spin density, we need to include an additional field since the second term $[\mathbf{S}_0\times\triangle\mathbf{S}_0] = -q^2[\mathbf{S}_0\times(\mathbf{S}_0 - s_a\mathbf{e}_x)] = s_aq^2[\mathbf{S}_0\times\mathbf{e}_x]$ has a nonzero value. It leads to the following structure of the magnetic field $\mathbf{B}_0 = \mathbf{B}_1 + \mathbf{B}_2$ with the additional constant field $\mathbf{B}_2 = -\frac{g_u\hbar}{12\gamma}q^2s_a\mathbf{e}_x$. To complete the solution, we need to find the coefficient χ. If we assume $\chi = const$ we find that equation $\nabla\cdot\mathbf{B}_0 = 0$ cannot be satisfied. So we consider coefficient χ s a function of coordinates $\chi(\mathbf{r})$. However, the x and z projections of equation $\nabla\times\mathbf{B}_0 = 4\pi\gamma\nabla\times\mathbf{S}_0$ can be satisfied at $\chi = 4\pi\gamma$ or $q = 0$. We conclude that two interactions entering the spin evolution equation (59) cannot support structure (60). Possibly, one can find a consistent solution in form (62) by extending the range of interactions included in the model.

6.1.4. Screw spiral spin structure

Here we consider a spiral shifting in the direction perpendicular to the rotation plane, so substitute $\mathbf{q} = q\mathbf{e}_x$ in equation (60). In the chosen regime, the structure simplifies to

$$\mathbf{S}_0(x) = s_b\mathbf{e}_y\cos(xq) + s_c\mathbf{e}_z\sin(xq) + s_a\mathbf{e}_x. \tag{62}$$

Let us consider the right-hand side of equation (59) under assumption (62) for the spin structure. The balance of two terms leads to the following form of magnetic field $\mathbf{B}_0 = \mathbf{B}_1 + \mathbf{B}_2$ with $\mathbf{B}_1 = \chi\mathbf{S}_0$ and

$$\mathbf{B}_2 = -\frac{g_u\hbar}{12\gamma}q^2s_a\mathbf{e}_x. \tag{63}$$

We need to check that the found magnetic field satisfies equation $\nabla\cdot\mathbf{B}_0 = 0$. It shows that function χ depends on coordinates y and z ($\chi(y,z)$) or to be a constant $\chi = const$. Next, we need to consider the second

Maxwell equation $\nabla \times \mathbf{B}_0 = 4\pi\gamma\nabla \times \mathbf{S}_0$ (its static regime). Assuming $\chi = const$, we find the explicit expression for $\chi = 4\pi\gamma$.

We can check that the right-hand side of equation (54) is equal to zero, since we consider the equilibrium state. We see that it is satisfied. Let us present the corresponding polarization $P_0^\alpha = g_{0\Pi}^\alpha \mathbf{S}^2 = g_{0\Pi}^\alpha (s_a^2 + s_b^2 \cos^2 qx + s_c^2 \sin^2 qx)$. It can be a constant under conditions $s_b = \pm s_c$. It gives polarization P_0^α in the following form $P_0^\alpha = g_{0\Pi}^\alpha (s_a^2 + s_b^2)$. Let us remind that the constant $g_{0\Pi}^\alpha$ is parallel to the shift of the ligand ion δ^α (47).

6.2. Antiferromagnetic multiferroics

We presented a spiral spin structure in the a-direction for ferromagnetic multiferroics in terms of the model based on the Zeeman energy and the Heisenberg-Coulomb exchange interaction. So, we are focused on the same regime for the antiferromagnetic multiferroics, but we also briefly mention the uniform regime.

6.2.1. Uniform regime

For the uniform regime, we have parallel partial spin densities, and therefore we have parallel vectors $\mathbf{L}_0$ and $\mathbf{\Sigma}_0$. However, we can consider different modules of the partial spin densities in their opposite directions, so $\mathbf{\Sigma}_0$ has a nonzero equilibrium value. It corresponds to the constant magnetic field parallel to vectors $\mathbf{L}_0$ and $\mathbf{\Sigma}_0$.

6.2.2. On a form of screw spiral spin structure

In the uniform case, we consider the parallel partial spin densities with different modules. Here, we can consider two regimes of spirals in a-direction. One corresponds to the parallel partial spin densities with different modules. So, we see spirals for $\mathbf{L}_0$ and $\mathbf{\Sigma}_0$ with the space phase shift on π. Another case is the regime, where the partial spin densities have approximately equal modules, but they are directed at the angle to each other. It leads to perpendicular directions of $\mathbf{L}_0$ and $\mathbf{\Sigma}_0$ at each point. It corresponds to the space phase shift on $\pi/2$ for $\mathbf{L}_0$ and $\mathbf{\Sigma}_0$.

Let us start the analysis with the screw spiral structure for $\mathbf{L}_0$ vector

$$\mathbf{L}_0(x) = l_b \mathbf{e}_y \cos(xq) + l_c \mathbf{e}_z \sin(xq) + l_a \mathbf{e}_x, \tag{64}$$

while other characteristics we retrieve from equilibrium regime of equations of motion.

Next, we need to find the magnetic field corresponding to both the spin evolution equations (11), (12), and the Maxwell equations $\nabla \cdot \mathbf{B}_0 = 0$ and $\nabla \times \mathbf{B}_0 = 4\pi \sum_{i=1}^{2} \nabla \times (\gamma_i \mathbf{S}_{0i})$. In the chosen case, we have $\gamma_1 = \gamma_2$ and $\mathbf{S}_{01} + \mathbf{S}_{02} = \mathbf{\Sigma}_0$.

We consider the equilibrium form of equation (12), where we dropped the second term on the right-hand side. It gives $(2\mu/\hbar)\mathbf{B}_0 = \chi \mathbf{L}_0 - g_{0u,AB}\mathbf{\Sigma}_0$, where χ is an unknown coefficient. We substitute this magnetic field in equation (11) and obtain vector $\mathbf{\Sigma}_0$:

$$\Sigma_0(x) = \frac{\alpha}{\chi}\mathbf{L}_0 + \frac{1}{6} g_u q^2 l_a \mathbf{e}_x, \tag{65}$$

where α is another unknown coefficient. It also leads to the expression for the magnetic field $\mathbf{B}_0 = (\hbar/2\mu)[(\chi - g_{0u,AB}\alpha/\chi)\mathbf{L}_0 - \frac{1}{6} g_{0u,AB} g_u q^2 l_a \mathbf{e}_x$.

Equation $\nabla \cdot \mathbf{B} = 0$ can be satisfied if $(\chi - g_{0u,AB}\alpha/\chi)$ is a constant or a combination of functions equal to zero. equation $\nabla \times \mathbf{B} = 4\pi\gamma\nabla \times \mathbf{\Sigma}_0$ can be satisfied at the following relation between two introduced coefficients

$$\chi^2 = \alpha(4\pi\gamma\mu/\hbar + g_{0u,AB}). \tag{66}$$

Hence, $(\chi - g_{0u,AB}\alpha/\chi)$ is a nonzero constant. So, α and χ are constants connected via equation (66).

To complete our analysis, we need to check that the polarization evolution equation (58) also corresponds to the equilibrium regime, so its right-hand side is equal to zero. The direct substitution of found $\mathbf{B}_0$, $\mathbf{L}_0$, and $\mathbf{\Sigma}_0$ shows that it is satisfied.

In this case, the polarization is mostly defined by vector $\mathbf{L}$ in accordance with equation (25). If we need to get a constant value of polarization, we need to choose $l_b = \pm l_c$ and find $\mathbf{P}_0 = (1/6)(\gamma/2mc) g_{(\beta)} \boldsymbol{\delta} (l_a^2 + l_b^2)$.

The spiral spin structures are the periodic magnetic structures, which appears to be one of nontrivial spin structures along with skyrmions, magnetic helix, magnetic vortex, chiral domain walls. Spatial variation of the spin density is the key property for the polarization formation. Hence, the spiral spin structure is one of structures which allows to obtain the electric polarization of the medium. If we consider the antisymmetric form of the electric polarization following [7], the spiral spin structures are necessary for electric polarization in multiferroics. The nonzero symmetric form of the electric polarization can be obtained for the collinear spin density in accordance with equation (40). However, this is a formal result, since experimental estimation of the polarization is made up to normalization constant. Nontrivial space dependence of the polarization can be

found for spiral structures. Moreover, the spiral spin structures allow to obtain the periodic change of the electric polarization in multiferroics.

7. Conclusion

The Landau–Lifshitz–Gilbert equation can be called the main macroscopic equation for the evolution of the magnetization in the magnetically ordered materials. Multiferroic materials show the existence of the electric polarization in addition to magnetization. Hence, the study of the multiferroics requires a couple of connected equations for the magnetization and the electric polarization. The problem of the derivation of described set of equations for the antiferromagnetic materials has been formulated in this paper. The II-type of multiferroics with the electric dipole moment proportional to the scalar product of the neighboring spins has been chosen for this research. The polarization evolution equation has been found under the action of the Zeeman energy and the Heisenberg-Coulomb exchange interaction. The similar equation for the ferromagnetic regime has been demonstrated as well. The many-particle quantum hydrodynamic method has been applied for the derivation of the required polarization evolution equation. Before, the application of this method to this derivation, the method has been successfully tested on the derivation of the spin/magnetization evolution equation.

However, the chosen definition of the electric dipole moment has been required an analytical justification. The justification has been made in several steps. First, the spin-current model is justified for the ferromagnetic materials via the momentum balance equation (the hydrodynamic Euler equation) containing the spin-orbit interaction. Second, the spin-current caused by the Dzylaoshinskii-Moriya interaction has been found from the spin/magnetization evolution equation and placed in the spin-current model to find the required polarization. Finally, the same steps have been made for the antiferromagnetic materials.

Therefore, it has been analytically derived that there is the electric dipole moment proportional to the scalar product of the neighboring spins caused by the Dzylaoshinskii-Moriya. The interpretation of the direction of the vector coefficient of proportionality in the electric dipole moment has been interpreted as being parallel to the shift of the ligand ion from the line connecting neighboring magnetic ions (this vector is the well-known part of the Dzylaoshinskii vector constant).

Some equilibrium spin configurations have been considered for the ferromagnetic and antiferromagnetic multiferroics. Regimes of parallel and spiral spin structures have been discussed, and corresponding electric polarizations have been calculated.

Overall, the Landau–Lifshitz–Gilbert equation contains the contribution of a number of physical mechanisms. Their systematic account in the polarization evolution equation is the research program demonstrated in this paper. The account of the Zeeman energy and the Heisenberg-Coulomb exchange interaction for the antiferromagnetic materials with the electric dipole moment proportional to the scalar product of the neighboring spins has been one of the initial steps towards the realization of this program.

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Data availability statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study, which is a purely theoretical one. The data cannot be made publicly available upon publication because no suitable repository exists for hosting data in this field of study. The data that support the findings of this study are available upon reasonable request from the authors.

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