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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED MEDIA

EVOLUTION EQUATION OF ELECTRIC POLARIZATION IN MULTIFERROICS PROPORTIONAL TO THE VECTOR PRODUCT OF CELL ION SPINS UNDER THE INFLUENCE OF THE HEISENBERG HAMILTONIAN

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Abstract. An evolution equation for polarization (electric dipole moment density) has been derived for type II multiferroics, where polarization is proportional to the vector product of cell ion spins. A regime is considered in which the main evolution mechanism is exchange Coulomb interaction, modeled by the Heisenberg Hamiltonian. The obtained polarization evolution equation contains spin density and nematic tensor density, which appears as an anticommutator of spins for particles with $S = 1$ and higher (for particles with spin $S = 1/2$ it degenerates into particle concentration). Also, to construct a closed model of spin and polarization evolution in multiferroics, equations for the above-mentioned physical quantities were obtained. The spin-current model is justified using the momentum balance equation and spin evolution equation, derived from the microscopic many-particle Pauli equation taking into account spin-orbit interaction. To analyze the mechanism of electric dipole moment formation proportional to the vector product of magnetic ion spins, the spin-current model was used, within which the relationship between the proportionality coefficient and the exchange integral was obtained. The mean-field approximation is used in the work, where the many-particle wave function of the ion system is approximated by the product of single-particle functions.

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1. INTRODUCTION

The diversity of phenomena in magnetically ordered and dielectrically ordered media has been attracting researchers' attention over the past decades. One example of theoretical research is the famous Landau-Lifshitz-Gilbert equation for the evolution of magnetic moment in magnetically ordered media, the concept of which was proposed by Landau and Lifshitz in 1935. Of particular interest is the simultaneous manifestation of magnetic and dielectric ordering that occurs in media called multiferroics. Moreover, these phenomena can coexist relatively independently (type I multiferroics) or exhibit interconnection (type II) [1]. In this paper, we consider type II multiferroics, in which the polarization of the crystal cell is formed proportionally to the vector product of the spins of its constituent magnetic ions.

Usually, three mechanisms of polarization emergence in type II multiferroics are distinguished [2], for each of which a relationship between the electric dipole moment of the crystal cell and the spins of its magnetic ions was proposed [2]. The features of the crystal lattice structure for the formation of multiferroics can also be found in [2]. For our case under consideration, the relationship between the dipole moment and spins of magnetic ions was derived in [3] based on the spin-current model. We use it below to derive a macroscopic expression for polarization, which coincides with the result of [4], where polarization was obtained from symmetry considerations applied to thermodynamic potentials. We also consider the justification of the spin-current model from the perspective of the quantum hydrodynamics method [5–9], which is the main research method in this work. Using effective spin current in the spin field evolution equation, caused by exchange interaction, allows not only to reproduce the result of [4] but also to establish a connection between the coefficient determining the cell dipole moment and the exchange integral. Moreover, a generalization of the result from [4] is provided, taking into account the contribution of quantum spin current associated with the Bohm potential. Such generalization also helps to clarify the applicability range of the result obtained in [4]. Our proposed approach to justifying the form of polarization and, one might say, to justifying the spin-current model, allows establishing the proportionality coefficient between spin current and polarization.

The results described above serve as preliminary steps before deriving the polarization evolution equation, for which a set of possible interactions can be included in the initial Hamiltonian in the microscopic non-stationary Schrödinger-Pauli equation. However, in this work, we limit ourselves to considering Coulomb exchange interaction in the form of the Heisenberg Hamiltonian $\widehat{H}_H = -J\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$, where $\hat{\mathbf{S}}_1$ and $\hat{\mathbf{S}}_2$ are the spins of two interacting particles (in our case, ions), and J is the exchange integral related to the overlap of electron wave functions.

In most cases, when analyzing magnetic phenomena, the analysis is limited to using the magnetization of the medium, which is proportional to spin density (for a system of magnetic particles of the same type). However, such a simplified representation is valid only for particles with spin 1/2 [10]. When considering atoms/ions with large spin, which is true for most magnetics forming magnetically ordered states (ferromagnetic phases, antiferromagnetic phases, etc.), quantum average values of products of spin operator projections of a single particle yield new physical quantities. The simplest example is the nematic tensor, proportional to the quantum average of the spin operators' anticommutator [11, 12]. When examining individual domains of a ferromagnet, we see a system of parallel spins. In this case, the nematic tensor can be, at least approximately, expressed through a single non-zero spin projection. However, when disturbance propagates, the picture becomes more complex, and the degree of inaccuracy in the approximate transition from the nematic tensor to the combination of spin projections increases and requires systematic evaluation. A similar situation arises in the domain wall region. In this paper, we focus on discussing the nematic tensor because it enters the evolution equation of electric polarization. Therefore, below we derive the evolution equation of the nematic tensor along with the evolution equation of spin density and the evolution equation of polarization. Note that the exchange interaction for a system of ions with spin greater than 1/2 is not limited to the term $\widehat{H}_H = -J\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$, and at least for particles with spin $S = 1$ gives biquadratic exchange $\widehat{H}_{H2} = -\tilde{J}(\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2)^2$. Its contribution to the Landau-Lifshitz equation and the necessity of using the nematic tensor are discussed in work [13]. However, in this paper, we focus on the contribution of exchange interaction described by the Heisenberg Hamiltonian $\widehat{H}_H = -J\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2$.

The article is organized as follows. Section 2 examines the fundamentals of the quantum hydrodynamics method and uses the momentum balance equation for approximate justification of the spin-current polarization model. Section 3 investigates the relationship between microscopic electric dipole moment and macroscopic electric dipole moment density. Section 4 derives the polarization evolution equation under the influence of exchange interaction described by the Heisenberg Hamiltonian. The evolution equation derivation is performed based on microscopic theory. Section 5 presents a brief discussion of the obtained results.

2. QUANTUM HYDRODYNAMICS METHOD — MOMENTUM BALANCE EQUATION, EQUILIBRIUM STATE, JUSTIFICATION OF POLARIZATION STRUCTURE

The simplest definition in quantum theory of material fields is the particle number concentration (we only consider magnetic ions, but a more general model allows accounting for concentration of particles of different type — non-magnetic ions)

$$n(\mathbf{r}, t) = \int \Psi_S^\dagger(R, t) \sum_i \delta(\mathbf{r} - \mathbf{r}_i) \Psi_S(R, t)\, dR. \quad (1)$$

The evolution of concentration leads to the continuity equation, in which particle flux density appears $\mathbf{j}$, coinciding with momentum density for non-relativistic systems. In this work, we partially consider spin-orbital interaction for analyzing the spin-current model, i.e., we account for relativistic effects. Here it is important to distinguish between particle flux density and particle momentum density. The structure of particle flux density was used in work [14] to justify the spin-current model. In this work, we consider the evolution equation of momentum density and force balance in it to reinterpret the result of work [14] within the quantum hydrodynamics method.

Following the logic of concentration definition (1), we define momentum density

$$\mathbf{p}(\mathbf{r},t) = \int(\Psi_S^{\dagger}(R,t)\sum_i \delta(\mathbf{r}-\mathbf{r}_i)\hat{\mathbf{p}}_i\Psi_S(R,t) + \\ +\text{H.c.})dR, \qquad (2)$$

and spin density

$$\mathbf{S}(\mathbf{r},t) = \int\Psi_S^{\dagger}(R,t)\sum_i \delta(\mathbf{r}-\mathbf{r}_i)\times(\hat{\mathbf{s}}_i\Psi(R,t))_S dR, \qquad (3)$$

where H.c. is used to denote the Hermitian conjugate term.

2.1. Momentum and Spin Balance Equations

The main purpose of this work is to examine the influence of exchange interaction on the evolution of electrical polarization in type II multiferroics. However, we will begin with a preliminary analysis of polarization definition. For this, let us consider the evolution of momentum and spin density of the medium. We will take into account several other interactions, namely, spin-orbital interaction, Dzyaloshinskii-Moriya interaction, and electric dipole moment energy in an external electric field. Eventually, we will use the microscopic many-particle Schrödinger-Pauli equation

$$i\hbar\partial_t\Psi(R,t) = \hat{H}\Psi(R,t) \qquad (4)$$

with the interaction Hamiltonian of the form

$$\hat{H} = \sum_{i=1}^{N}\Bigg[-\hat{\mathbf{d}}_i\cdot\mathbf{E}_i - \frac{1}{2mc}(\hat{\mu}_i\cdot[\mathbf{E}_i\times\hat{\mathbf{p}}_i]) - \\ -\hat{\mu}_i\cdot\mathbf{B}_i - \frac{1}{2}\sum_{\substack{j=1,\\ j\neq i}}^{N}\left(U_{ij}\hat{\mathbf{s}}_i\cdot\hat{\mathbf{s}}_j + \mathbf{D}_{ij}\cdot\left[\hat{\mathbf{s}}_i\times\hat{\mathbf{s}}_j\right]\right)\Bigg]. \qquad (5)$$

Here N is the total number of particles/ions, $\Psi(R,t)$ is the wave function of the particle system, $R = \{\mathbf{r}_1,\ldots,\mathbf{r}_N\}$, $\hat{\mathbf{d}}_i$ is the electric dipole moment operator, which is defined through ion displacement, its further connection with ion spins [2] will be determined during the analysis of the obtained evolution equations of macroscopic functions, $\hat{\mathbf{p}}_i = -i\hbar\hat{\nabla}$ is the momentum operator of the i-th particle, $\hbar$ is the reduced Planck constant, m_i is the particle mass, c is the speed of light in vacuum, $\mathbf{E}_i$ and $\mathbf{B}_i$ are the electric field strength and magnetic field induction acting on the i-th particle/ion, $\hat{\mathbf{s}}_i$ is the dimensional spin operator, $\hat{\mu}_i$ is the magnetic moment operator, proportional to the spin operator $\hat{\mu}_i = \gamma_i\hat{\mathbf{s}}_i$ through the gyromagnetic ratio $\gamma_i, U_{ij} = U(\mathbf{r}_i - \mathbf{r}_j)$ is the scalar coefficient of exchange Coulomb interaction in the Heisenberg Hamiltonian (exchange integral $\mathbf{D}_{ij} = \mathbf{D}(\mathbf{r}_i - \mathbf{r}_j) = -\mathbf{D}_{ji}$ is the vector coefficient of Dzyaloshinskii-Moriya exchange spin-orbital interaction. Further in the work, the mean field approximation is used, where the many-particle wave function of the ion system is approximated by the product of functions.

The Hamiltonian (5) contains five terms corresponding to different interactions, namely (in the order of terms in the Hamiltonian): the potential energy of dipole moments in an external electric field, spin-orbital interaction corresponding to the effect of electric field on magnetic moments of ions [15] (see sections 33 and 83), potential energy of magnetic moment in an external magnetic field, exchange Coulomb interaction in the form of Heisenberg Hamiltonian, and Dzyaloshinskii-Moriya exchange spin-orbital interaction. A more detailed description allows accounting for the electric field created by the system's dipoles and acting on dipole moments along with the external electric field. It can also be noted that the electric field in spin-orbital interaction can be caused by electric dipole moments of the medium. Including exchange interaction in the Hamiltonian for the Schrödinger equation is not an example of a fundamental microscopic approach. In this work, we consider materials with strongly pronounced magnetic and dielectric properties. These properties are formed in groups of ions located at crystal lattice sites. Thus, some interactions leading to the formation of an ion or crystal lattice are not explicitly taken into account. The exchange part of the electromagnetic interaction of valence electrons is indirectly accounted for by corresponding terms in the Hamiltonian, which reflects the transition to the scale of distances and energies at which ions and their compositions are "elementary" objects of our theory.

Following the quantum hydrodynamics method, we obtain the corresponding momentum balance equation

$$\partial_t \mathbf{p} = g_{0u} S^\beta \nabla S^\beta + \mu S^\beta \nabla B^\beta + P^\beta \nabla E^\beta +$$

$$+ \frac{\gamma}{2mc} \varepsilon^{\beta\gamma\delta} J^{\delta\gamma} (\nabla E^\beta) + \mathbf{F}_{DM}, \qquad (6)$$

where

$$g_{0u} = \int U(r) d\mathbf{r}$$

is the exchange interaction constant, arising as an integral characteristic of the interaction between pairs of neighboring particles, it can be expressed through the exchange integral, for a specific model form of the exchange integral [16], $J^{\delta\gamma}$ is the spin current tensor, P^β is the electric dipole moment density (formula presented below), $\mathbf{F}_{DM}$ is the Dzyaloshinskii-Moriya interaction force density, and the spin evolution equation

$$\partial_t \mathbf{S} = \frac{2\gamma}{\hbar} [\mathbf{S}, \mathbf{B}] + \frac{1}{6} g_u [\mathbf{S}, \Delta \mathbf{S}] + \mathbf{T}_{DM} + \mathbf{T}_{SO}, \qquad (7)$$

where Δ is the Laplace operator, $g_u = \int r^2 U(r) d\mathbf{r}$ is the second exchange interaction constant, being the second "moment" of the exchange integral, while constant g_{0u} can be called the zero "moment" of the exchange integral, $\mathbf{T}_{DM}$ is the Dzyaloshinskii-Moriya interaction torque density,

$$T^\alpha_{SO} = -\frac{\gamma}{\hbar c} \varepsilon^{\alpha\beta\gamma} \varepsilon^{\beta\mu\nu} E^\mu J^{\gamma\nu} - \partial_\beta J^{\alpha\beta}_{SO}$$

is the spin-orbit interaction torque density. It contains the total spin current $J^{\gamma\nu}$ and the relativistic part of the spin current due to spin-orbit interaction, which for a particle with spin $S = 1/2$ has the form

$$J^{\alpha\beta}_{SO} = \frac{\mu\hbar}{4mc} \varepsilon^{\alpha\beta\gamma} n E^\gamma.$$

For particles with large spin, the spin current is expressed through the nematic tensor $\pi^{\alpha\beta}$ (defined below by formula (29)):

$$J^{\alpha\beta}_{SO} = (\gamma/mc) \varepsilon^{\beta\mu\nu} \pi^{\alpha\nu} E^\mu.$$

A special case of the spin evolution equation is the Landau-Lifshitz-Gilbert equation. Equations (6) and (7) are derived from the Schrödinger-Pauli equation with Hamiltonian (5). Consequently, equations (6) and (7) contain the same interactions as Hamiltonian (5). Note that the contribution of the Heisenberg Hamiltonian is evidently the second term on the right side of equation (7), which can be represented as the divergence of the spin current tensor [17]:

$$(1/6) g_u \varepsilon^{\alpha\beta\gamma} S^\beta \Delta S^\gamma =$$

$$= \partial_\delta ((1/6) g_u \varepsilon^{\alpha\beta\gamma} S^\beta \partial_\delta S^\gamma) = -\partial_\delta J^{\alpha\delta}_{HH},$$

where index *HH* emphasizes that the spin current is associated with the Heisenberg Hamiltonian.

In work [18], an example of the coefficient $\mathbf{D}_{ij}$ for perovskites is provided. In perovskites, the following picture of interaction between the nearest magnetic ions emerges. Exchange interaction occurs through superexchange via a ligand, which is a nonmagnetic ion (for example, an oxygen ion) located between magnetic ions but away from the direct line connecting these ions. This mechanism leads to the following structure of the Dzyaloshinsky coefficient:

$$\mathbf{D}_{ij} \sim \mathbf{r}_{ij} \times \delta,$$

where δ is the displacement vector of the ligand from the center of the segment connecting the magnetic ions. Additionally, we need to introduce a proportionality coefficient that accounts for the decrease in interaction with increasing distance between magnetic ions r_{ij}. As a result, we obtain

$$\mathbf{D}_{ij} = \beta(r_{ij}) \mathbf{r}_{ij} \times \delta,$$

where the proportionality coefficient $\beta(r_{ij})$ depends only on the magnitude of the distance between ions.

For the presented model of the Dzyaloshinsky coefficient in perovskites, we arrive at the following expressions for the density of the Dzyaloshinsky-Moriya interaction force:

$$F^\sigma_{DM} = \frac{1}{2} \hat{g}^{\alpha\lambda\mu\sigma}_{4D} \delta^\nu \varepsilon^{\beta\mu\nu} \varepsilon^{\beta\gamma\delta} \partial^\alpha (S^\gamma \partial^\lambda S^\delta), \qquad (8)$$

where

$$\hat{g}^{\alpha\lambda\mu\sigma}_{4D} = \int \xi^\alpha \xi^\lambda (\beta(\xi) \delta^{\mu\sigma} + \xi^\mu \xi^\sigma \frac{1}{\xi} \frac{\partial \beta(\xi)}{\partial \xi})$$

is the unreduced form of the interaction constant, and for the torque density

$$T^\mu_{DM} = \frac{1}{3} g_{2(\beta)} \delta^\nu (-\varepsilon^{\mu\delta\nu} S^\beta \partial^\delta S^\beta + \varepsilon^{\alpha\delta\nu} S^\alpha \partial^\delta S^\mu), \qquad (9)$$

where

$$g_{2(\beta)} = \int \xi^2 \beta(\xi) d^3\xi.$$

The first term in the torque (9) can be transformed in the form of spin current divergence.

As a result, we get

$$T^{\mu}_{DM} = -\partial_{\beta} J^{\mu\beta}_{DM} + \frac{1}{3} g_{2(\beta)} \delta^{\nu} \varepsilon^{\alpha\delta\nu} S^{\alpha} \partial^{\delta} S^{\mu},$$

where

$$J^{\mu\beta}_{DM} = \frac{1}{3} g_{2(\beta)} \delta^{\nu} \frac{1}{2} \varepsilon^{\mu\beta\nu} \mathbf{S}^2. \qquad 10)$$

Let’s once again focus on the structure of the Dzyaloshinskii-Moriya interaction force density (8), for which we write the final expression in vector form:

$$\mathbf{F}_{DM} = \frac{1}{3} g_{(\beta)} ((\delta \cdot \mathbf{S}) \nabla (\nabla \cdot \mathbf{S}) - (\mathbf{S} \cdot \nabla) \nabla (\delta \cdot \mathbf{S})), \quad (11)$$

where

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

The Dzyaloshinskii-Moriya vector, like the exchange integral, is a function of distance that can be replaced with a specific value when considering a fixed distance between atoms in a crystal. Exchange (primarily Coulomb) interaction is the mechanism of interaction between neutral atoms, as the interaction of valence electrons (electrons from outer shells). It also contributes to the interaction of ions. In gases, the distance between atoms varies significantly. However, the interaction manifests at small distances (compared to the average distance between atoms). This leads to the fact that in macroscopic equations, short-range interaction is described by a set of interaction constants (mainly by one constant – the integral of effective interaction potential, in exchange interaction associated with the overlap of valence electron wave functions of nearby atoms and therefore significantly dependent on distance). From this perspective, the Heisenberg exchange integral and the Dzyaloshinskii-Moriya vector are integrable functions of the distance between interacting atoms. In crystals, when neglecting thermal vibrations of atoms/ions around the equilibrium position, there is a fixed distance between atoms/ions. When considering such systems, we can choose a specific form of spatial dependence of the considered functions in the form of a narrow “step”:

$$U(r) = U_0 \theta(r - a) \theta(a + \delta a - r),$$

where θ is the Heaviside function, U_0 is the value of the exchange integral in the considered crystal, a is the average distance between atoms, δa is the amplitude of thermal vibrations.

Note that if we obtain the integral only from the Dzyaloshinskii-Moriya vector (as a function of r_{ij}), it will turn to zero. However, when expanding wave functions and the delta function, additional factors containing r_{ij} arise, so that some expressions become non-zero.

2.2. Equilibrium State and Spin-Current Model

A multiferroic is a system with magnetic and dielectric ordering. As a consequence, there exist macroscopic equilibrium electric and magnetic fields inside the system. Therefore, it is necessary to consider the equilibrium state of the system with non-zero field values. From equation (6), it is evident that for the considered interactions, non-zero field values are possible in the presence of field inhomogeneity, as all terms in the right-hand side contain spatial derivatives of fields. The third and fourth terms in the right-hand side are proportional to ∇E^{β}. This provides an opportunity for the formation of a balance of these forces in the equilibrium state with various types of electric field inhomogeneity. This leads to the formation of equilibrium polarization due to spin-orbital interaction:

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

Note that here we used the self-consistent part of the spin-orbital interaction corresponding to the part of the many-particle wave function consisting of the product of single-particle wave functions of individual magnetic ions.

The combination of the first and second terms in the right-hand side of the momentum balance equation (6) gives an equilibrium magnetic field proportional to the spin density $B^{\beta} = -g_{0u} S^{\beta}/\gamma$ and caused by exchange Coulomb interaction.

The force density of the Dzyaloshinskii-Moriya interaction for an odd Dzyaloshinskii coefficient has a structure different from other terms in the momentum balance equation. Moreover, it contains two spatial derivatives, which indicates a relatively small contribution compared to other terms.

Analysis of the equilibrium state based on the momentum balance equation (6) allowed us to obtain a relationship between polarization and spin current. Obviously, the obtained relationship is a special case of force balance, and in general, other interactions may contribute to this relationship. The next question that arises in the development of the spin-current model is the expression for spin current caused by various effects. We take expressions for spin currents from the spin evolution equation. As shown by the analysis above, the contribution of exchange Coulomb interaction, taken in the form of the Heisenberg Hamiltonian in the spin evolution equation, can be represented as a divergence of spin current. This gives us one of the partial spin currents. The contribution of the Dzyaloshinskii-Moriya interaction can also be represented as a divergence of spin current, but under conditions that are also formulated above.

2.3. Macroscopic polarization within the spin-current model

Let's consider the application of the spin-current model to two types of partial spin currents. For the spin current caused by the Heisenberg Hamiltonian, we obtain the following expression for macroscopic polarization:

$$P^{\mu}_{HH} = \frac{\gamma}{2mc}\varepsilon^{\mu\alpha\beta}J^{\alpha\beta}_{HH} = $$

$$= \frac{\gamma}{12mc}g_u(S^{\beta}\partial_{\beta}S^{\mu} - S^{\mu}\partial_{\beta}S^{\beta}). \quad (12)$$

This corresponds to the result obtained by M. Mostovoy [4] (see also work [19], p. 533). For the spin current caused by the Dzyaloshinskii-Moriya interaction, we also find an expression for macroscopic polarization:

$$P^{\mu}_{DM} = \frac{\gamma}{2mc}\varepsilon^{\mu\alpha\beta}J^{\alpha\beta}_{DM} = -\frac{\gamma}{12mc}g_{2(\beta)}\delta^{\mu}\mathbf{S}^2. \quad (13)$$

We use these expressions below when analyzing the microscopic structure of the electric dipole moment considered in the literature [2].

Equations (9) and (10) lead to expression (13). The main contribution to polarization does not depend on spatial derivatives. In this case, the presence of δ gives a non-zero spin current $J^{\alpha\beta}_{DM}$, which gives a non-zero value of the spin-orbital interaction force (the fourth term in the right-hand side of equation (6)). And the balance of this force with the third term, proportional to polarization $\mathbf{P}$ (where $\mathbf{P}$ is proportional to δ), gives a consistent equilibrium condition. From formula (13), it follows that in such equilibrium, there exists a relationship between the medium polarization $\mathbf{P}$ and spin density in the presented form. Expression (13) corresponds to the electric dipole moment operator for collinear spins presented in work [2] (see Figs. 2*a*–2*c*).

Depending on the structure and symmetry of crystals, different mechanisms of polarization formation are realized. It is shown that the displacement of charge density can occur due to spin-orbital interaction. The latter manifests itself in the presence of spin current in the system of ions forming an inhomogeneous electric field. If the spin current due to Coulomb exchange interaction predominates, then we arrive at formula (12). Expression (12) coincides with the expression for polarization (23), which was used in literature for magnetically ordered structures with noncollinear spins realized in perovskites of type $\mathrm{RMnO_3}$, where $\mathrm{R = Tb, Dy}$ [20]. Formula (13) arises for M–X–M-structure, where M represents magnetic ions, X represents a ligand ion. This structure is realized in rare-earth perovskite-type manganites (or orthorhombic manganites) $\mathrm{RMnO_3}$ at $\mathrm{R = Ho, Er, Tm, Yb}$ [21, 22]. Another multiferroic is the family of mixed-valence manganese oxides $\mathrm{RMnO_3}$, where R = Y, Tb, Ho, Er or Tm [23]. The magnetic structure in such substances is antiferromagnetic in the ab plane, forming a collinear spin order of magnetic ions $\mathrm{Mn_4^+}$ and $\mathrm{Mn_3^+}$.

2.4. Spin current and Bohm potential

Accounting for small oscillations of ions in the vicinity of equilibrium position would require considering the kinetic energy of ions in the initial Hamiltonian (5). This would also lead to terms describing particle flows, which are absent in this case when considering crystals, and systematic exclusion of flows from equations would complicate the given equations. However, the consideration of kinetic energy also manifests in the emergence of quantum effects, particularly in the appearance of the quantum Bohm potential, which contributes to the spin current in the spin evolution equation [24] (see equation (9)):

$$J^{\alpha\beta}_{Bohm} = -\frac{\hbar\gamma}{2m}\varepsilon^{\alpha\mu\nu}S^{\mu}\partial^{\beta}\left(\frac{S^{\nu}}{n}\right). \quad (14)$$

If we consider the quasi-classical dynamics of polarization, then the partial currents obtained and used above should exceed the spin current caused by the Bohm potential.

Furthermore, the Bohm potential is associated with the quantum motion of particles, which can lead to deformations and the formation of an electric dipole moment. Let us calculate it within the framework of the spin-current model:

$$P^{\mu}_{Bohm} = \frac{\gamma}{2mc}\varepsilon^{\mu\alpha\beta}J^{\alpha\beta}_{Bohm} =$$

$$= \frac{\hbar\gamma^2}{4m^2c}\left[S^{\mu}\partial^{\beta}\left(\frac{S^{\beta}}{n}\right) - S^{\beta}\partial^{\beta}\left(\frac{S^{\mu}}{n}\right)\right]. \quad (15)$$

Let us compare the polarizations (12) and (15). They have a similar structure, but expression (15) contains concentration derivatives. Let us consider the constant concentration regime and obtain an estimate for the constant g_u (without considering the sign): $g_u \gg \hbar\gamma/2m$.

2.5. Discussion of the spin-current model based on the probability current structure

For the considered Hamiltonian, the continuity equation $\partial_t n + \nabla\cdot\mathbf{j} = 0$ leads to the particle current density **j** of the following form:

$$\mathbf{j} = \int(\Psi^{\dagger}_{S}(R,t)\sum_{i}\delta(\mathbf{r}-\mathbf{r}_i)\hat{\mathbf{j}}_i\Psi_S(R,t) +$$

$$+\text{H.c.})dR, \quad (16)$$

where

$$\hat{\mathbf{j}}_i = \frac{1}{2m_i}\left(\hat{\mathbf{p}}_i + \frac{\mu}{2m_i c}\left[\mathbf{E}_i\times\hat{\mathbf{s}}_i\right]\right). \quad (17)$$

To give a more familiar form to the particle current operator, we included the momentum operator $\hat{\mathbf{p}}_i$, which may arise from the kinetic energy operator in the original Hamiltonian, but it is not considered in our case. Moreover, in work [14], the momentum contribution is also discarded since ions are considered whose motion is negligibly small. Additionally, the particle/probability current is defined ambiguously, and we can add the curl of an arbitrary vector to definition (16), which will lead to a change in the structure of operator (17) (see [14, 25]). Usually, the curl of spin density/magnetization is added to the probability current definition

$$\tilde{\mathbf{j}} = \mathbf{j} + (\hbar/2ms)\text{rot}\mathbf{S},$$

where s is the spin of the considered ions. The balance between the spin curl and the term caused by spin-orbit interaction provides the relationship between the electric field strength and spin density, and the further connection between polarization and spin density:

$$\mathbf{P} \sim (\mathbf{S}\times(\nabla\times\mathbf{S}))/\mathbf{S}^2,$$

see [14]. We also write a simplified form of this formula without vector products

$$\mathbf{P} \sim [(\nabla\mathbf{S}^2/2 - (\mathbf{S}\cdot\nabla)\mathbf{S}]/\mathbf{S}^2$$

for further comparison with the results of microscopic structure analysis of the electric dipole moment [2].

3. ON THE MICROSCOPIC DETERMINATION OF MULTIFERROIC POLARIZATION

In the previous section, we presented an analysis of the polarization structure associated with spin-orbital interaction. This provides partial justification for the spin-current model and the possibility for its further generalization. Furthermore, such analysis provides macroscopic expressions for polarization formed by different types of exchange interaction (within the spin-current model). This allows us to reconstruct the microscopic structure of polarization leading to the obtained macroscopic expressions. However, these microscopic structures are already known in the literature [2], and we can choose the one we need to transition to the macroscopic expression and compare it with the one obtained above.

Let's consider the spins of two magnetic ions $\mathbf{s}_i$ and $\mathbf{s}_j$ and, following the works, write the expression for the electric dipole moment of the crystal unit cell

$$\mathbf{d}_{ij} = \alpha_{ij}[\mathbf{r}_{ij}\times[\mathbf{s}_i\times\mathbf{s}_j]], \quad (18)$$

where we use the relative distance between particles $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, in this case magnetic ions. Further, let's transition to the operator form of this equality

$$\hat{\mathbf{d}}_{ij} = \alpha_{ij}[\mathbf{r}_{ij}\times[\hat{\mathbf{s}}_i\times\hat{\mathbf{s}}_j]] \quad (19)$$

for developing a quantum model of type II multiferroics polarization. Let's note for certainty the commutation properties of spin operators

$$[\hat{s}^{\alpha}_i, \hat{s}^{\beta}_j] = i\hbar\delta_{ij}\varepsilon^{\alpha\beta\gamma}\hat{s}^{\gamma}_i, \quad (20)$$

where α, β, γ are tensor indices taking values of Cartesian coordinates x, y, z. In this work, we imply summation over repeated indices (Greek letters). The symbol i is used for the imaginary unit $i^2 = -1$, δ_{ij} – three-dimensional Kronecker symbol, $\varepsilon^{\alpha\beta\gamma}$ is the three-dimensional Levi-Civita symbol.

From formula (18), it is evident that we are considering ions with non-parallel spins in systems with spiral magnetization structures [18]. However, there are non-trivial examples of systems with parallel spins [26], which are not described by this model.

The main element of consideration is the electric dipole moment of the crystal cell. Therefore, in definition (19), we transition from a pair of ions to a cell with number i. To account for the fact that the dipole moment is created by ion i in conjunction with the neighboring ion, we introduce coefficient $\alpha_{ij}(r_{ij})$, which rapidly decreases with increasing distance, and arrive at the following modification of definition (19):

$$\hat{\mathbf{d}}_i = \sum_{j\neq i}\alpha_{ij}(r_{ij})[\mathbf{r}_{ij}\times[\hat{\mathbf{s}}_i\times\hat{\mathbf{s}}_j]], \qquad (21)$$

where, for example, $\alpha_{ij}(r_{ij}) = \alpha_{ij}$ at $r < a_{eff}$, $\alpha_{ij}(r_{ij}) = 0$ at $r > a_{eff}$ coefficient $\alpha_{ij}(r_{ij})$ can be represented as a step function in three-dimensional space.

Above, when deriving the momentum balance equation (6), we used the definition of polarization through the electric dipole moment operator $\hat{\mathbf{d}}_i$:

$$\mathbf{P}(\mathbf{r},t) = \int\Psi_S^\dagger(R,t)\sum_i\delta(\mathbf{r}-\mathbf{r}_i)(\hat{\mathbf{d}}_i\Psi(R,t))_S dR. \qquad (22)$$

The initial “bare” electric dipole moment $\hat{\mathbf{d}}_i$ is associated with ion displacement $\hat{\mathbf{d}}_i = q_i\mathbf{r}_i$, where q_i is the ion charge. Further, we will consider the evolution of polarization (22) with operator (21). Before moving on to the polarization evolution equation, let’s examine the relationship between macroscopic polarization $\mathbf{P}$ and spin density associated with operator (21). Let’s take into account the rapid decay of function $\alpha_{ij}(r_{ij})$ with increasing distance r_{ij} (following the method described in [27]). To account for this property, let’s introduce the center of mass variables of the particle pair $\mathbf{R}_{ij} = (\mathbf{r}_i + \mathbf{r}_j)/2$ and their relative distance $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$. Let’s express the coordinates of the particles under consideration through new coordinates $\mathbf{r}_i = \mathbf{R}_{ij} + (1/2)\mathbf{r}_{ij}$ and $\mathbf{r}_j = \mathbf{R}_{ij} - (1/2)\mathbf{r}_{ij}$. Let’s make the corresponding substitution in the delta function $\delta(\mathbf{r}-\mathbf{r}_i)$ and in the wave function $\Psi(R,t) = \Psi(...,\mathbf{r}_i,...,\mathbf{r}_j,...,t)$, which are included in the definition of polarization (22). Then, let’s perform the expansion in terms of relative motion coordinates r_{ij}. In the lowest non-zero order of expansion from (22), with substitution of (21), we find

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{3}g_{(\alpha)}[(\mathbf{S}\cdot\nabla)\mathbf{S} - \mathbf{S}(\nabla\cdot\mathbf{S})], \qquad (23)$$

where

$$g_{(\alpha)} = \int\xi^2\alpha(\xi)d\xi.$$

Up to a proportionality coefficient, the obtained expression coincides with the result of M. Mostovoy [4] (see also [19], p. 533), which was derived by him using symmetry considerations for polarization, magnetization, and thermodynamic potentials. Comparing the coefficients in formula (23) and formula (2) of work [4], we obtain

$$g_{(\alpha)} = 3\gamma\chi_e\mu^2,$$

where χ_e is the dielectric susceptibility in the absence of magnetism, γ is the undetermined proportionality coefficient used in work [4] for constructing the thermodynamic potential (see formula (1) of work [4]).

Let’s continue comparing formula (23) with the results discussed above. We can see that the result of work [14], presented by us in section 2.4, does not correspond to work [4] and our result based on formula (21). However, the application of the spin-current model with partial spin current due to the Heisenberg exchange interaction gives us an expression for $\mathbf{P}_{HH}$ (see section 2.3), which coincides with formula (23) up to a proportionality coefficient. Moreover, the coefficient in $\mathbf{P}_{HH}$ and is related to the exchange integral included in the Heisenberg Hamiltonian and is proportional to g_u. This allows us to interpret the function $\alpha_{ij}(r_{ij})$ in (18) and operator (21). As a result, we obtain the following relation at $\frac{1}{12}(3\gamma/mc)g_u = g_{(\alpha)}$:

$$\alpha_{ij}(r_{ij}) = \frac{1}{12}\frac{3\gamma}{mc}U(r_{ij}). \qquad (24)$$

In works [3, 18], it is stated that exchange Coulomb interaction leads to the formation of polarization with parallel spins, while the structure of type (18) corresponds to the Dzyaloshinskii-Moriya interaction. *Our analysis shows that considering the exchange Coulomb interaction between ions in the form of the Heisenberg Hamiltonian leads to the structure of the electric dipole moment defined by expression (18). Furthermore, our analysis allows us to analytically calculate the proportionality coefficient that determines the dipole moment in formula (18).*

3.1. On physical mechanisms leading to the formation of spin polarization structure

For comparison, let’s consider the polarization structure arising in a system of parallel spins, following review [2] (see Fig. 2, case 1): $\hat{\mathbf{d}}_i \sim \pi(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_{i+1})$. This leads to the following macroscopic polarization: $P^\alpha = g^\alpha_{0\Pi}\mathbf{S}^2$, where $g^\alpha_{0\Pi} = \int \Pi^\alpha(r)d\mathbf{r}$. Analyzing case (13), in the mode of ligand ion displacement perpendicular to the spin polarization direction, we obtain $P^\alpha = g^\alpha_{0\Pi}\mathbf{S}^2$ and with additional condition $g^\alpha_{0\Pi} = -(1/6)g_{2(\beta)}\delta^\alpha$. From the microscopic description perspective, we can conclude that $\Pi^\alpha_{ij}(r_{ij}) = r^2_{ij}\beta(r_{ij})\delta^\alpha$.

The presented analysis allows us to reinterpret Fig. 2 in review [2] (the first two cases). Paper [2] states that the electric dipole moment $\hat{\mathbf{d}}_i \sim \pi(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_{i+1})$ is caused by symmetric exchange interaction, or, in other words, by the Coulomb exchange interaction represented by the Heisenberg Hamiltonian. Paper [2] also states that the electric dipole moment

$$\hat{\mathbf{d}}_i \sim \alpha_{ij}[\mathbf{r}_{ij} \times [\mathbf{s}_i \times \mathbf{s}_j]]$$

is caused by antisymmetric exchange interaction, i.e., the Dzyaloshinskii-Moriya interaction. Our analysis, based particularly on equations (12) and (13), leads to an opposite interpretation of the nature of these dipole moment operators. Note that the spin-current model is usually tied to explaining the structure

$$\hat{\mathbf{d}}'_i \sim \alpha_{ij}[\mathbf{r}_{ij} \times [\mathbf{s}_i \times \mathbf{s}_j]].$$

This is evident even from Fig. 2*f* in paper [2]. The connection between the polarization structure and the interaction leading to its formation is not quite clear in paper [2]. One might assume that there is a qualitative interpretation based on comparing spin structures in the Heisenberg Hamiltonian (or Dzyaloshinskii-Moriya) and in the electric dipole moment. If both structures contain scalar products of spins, they have a common mechanism, i.e., symmetric Heisenberg exchange. Otherwise, if both structures contain vector products of spins, the common mechanism is the Dzyaloshinskii-Moriya interaction. But the analogy described here is not correct. Manipulation with spin operators often leads to the emergence of a commutator, which transforms the scalar product of spin operators into a vector product and vice versa. What needs to be monitored as an invariant in qualitative analysis is the mathematical or tensor structure of the interaction coefficient in the original Hamiltonian. In the Heisenberg Hamiltonian, this is a scalar quantity – the exchange integral. Therefore, one can expect that the corresponding polarization will be related to spin operators through a scalar coefficient (see α in formula (18)). The Dzyaloshinskii-Moriya Hamiltonian contains the Dzyaloshinskii vector constant. Therefore, one can expect that polarization will be related to spin operators through a vector coefficient $\hat{\mathbf{d}}_i \sim \pi(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_{i+1})$. Here, as we have shown, the coefficient is not the Dzyaloshinskii constant itself, but the ligand displacement vector that enters into the Dzyaloshinskii constant [18] $\mathbf{D}_{ij} \sim \mathbf{r}_{ij} \times \delta$. The described discrepancy between the mechanism and interpretation may be the reason for criticism of the spin-current model [28]. However, the reasons for this noted inconsistency may be deeper.

4. POLARIZATION EVOLUTION EQUATION

The analysis presented above shows that different types of electric dipole moment of a multiferroic cell can be obtained based on the quantum hydrodynamics method. Despite the emphasis on the evolution of macroscopic functions, the quantum hydrodynamics method contains a connection with microscopic description. Based on the obtained definition of the electric dipole moment for the regime when it is proportional to the vector product of spins (18) (previously known from works [2, 3]), and its transformation to form (21), we proceed to derive the evolution equation for multiferroic polarization. Let us specify the form of the Hamiltonian used for the proposed derivation:

$$\hat{H} = -\frac{1}{2}\sum_{i=1}^{N}\sum_{\substack{j=1,\\ j\neq i}}^{N} U(r_{ij})(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j), \tag{25}$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$.

Further, according to the quantum hydrodynamics method, we differentiate the polarization definition (22) with operator (21) with respect to time and use the time-dependent Schrödinger equation to transform the obtained expression to the form

$$\partial_t \mathbf{P}(\mathbf{r},t) =$$
$$= \frac{i}{\hbar}\int \Psi^{\dagger}(R,t)\sum_i \delta(\mathbf{r}-\mathbf{r}_i)[\widehat{H},\hat{\mathbf{d}}_i]\Psi(R,t)dR. \quad (26)$$

Let us represent the commutator appearing in formula (26) as

$$[\widehat{H},\hat{d}_i^{\alpha}] = -i\hbar\left\{\frac{1}{2}\sum_{j\neq i}U_{ij}\alpha_{ij}\varepsilon^{\alpha\beta\gamma}r_{ij}^{\beta}(2\hat{\mathbf{s}}_i^2\hat{s}_j^{\gamma} - 2\hat{\mathbf{s}}_j^2\hat{s}_i^{\gamma} +\right.$$
$$+\{\hat{s}_j^{\gamma},\hat{s}_j^{\sigma}\}s_i^{\sigma} - \{\hat{s}_i^{\gamma},\hat{s}_i^{\sigma}\}\hat{s}_j^{\sigma}) + \sum_{n\neq i,j}\sum_{j\neq i}\alpha_{ij}\varepsilon^{\alpha\beta\gamma}r_{ij}^{\beta}\times$$
$$\left.\times((\hat{s}_n^{\gamma}\hat{s}_j^{\sigma}\hat{s}_i^{\sigma} - \hat{s}_n^{\sigma}\hat{s}_j^{\sigma}\hat{s}_i^{\gamma})U_{ni} + (\hat{s}_n^{\sigma}\hat{s}_j^{\gamma}\hat{s}_i^{\sigma} - \hat{s}_n^{\gamma}\hat{s}_j^{\sigma}\hat{s}_i^{\sigma})U_{nj})\right\}, \quad (27)$$

where

$$\{\hat{s}_i^{\alpha},\hat{s}_i^{\beta}\} = 2\overset{?}{\pi}_i^{\alpha\beta} = \hat{s}_i^{\alpha}\hat{s}_i^{\beta} + \hat{s}_i^{\beta}\hat{s}_i^{\alpha}$$

is the anticommutator of spin operators, proportional to the nematic tensor operator $\pi_i^{\alpha\beta}$.

Let's use the property of rapid decrease of functions U_{ij} and α_{ij} with increasing distance between particles. For this, we introduce the coordinates of the center of mass and relative distance. We have two groups of terms. In one, we consider pairs of particles, and in the other – groups of three particles. For particle pairs, we have already introduced the required coordinates when analyzing the polarization structure (23). Let's present the required variables for a system of three particles. Let's write the center of mass coordinate

$$\mathbf{R}_{ijn} = (\mathbf{r}_i + \mathbf{r}_j + \mathbf{r}_n)/3,$$

as well as two coordinates of relative motion

$$\mathbf{r}_{in} \equiv \mathbf{r}_1 = \mathbf{r}_i - \mathbf{r}_n, \qquad \mathbf{r}_{jn} \equiv \mathbf{r}_2 = \mathbf{r}_j - \mathbf{r}_n.$$

When necessary, we can use the coordinates of relative motion of the third pair of particles

$$\mathbf{r}_{ij} \equiv \mathbf{r}_3 = \mathbf{r}_1 - \mathbf{r}_2.$$

Also, let's present the inverse transformation of individual particle coordinates through the introduced variables:

$$\mathbf{r}_i = \mathbf{R}_{ijn} + (2/3)\mathbf{r}_{in} - (1/3)\mathbf{r}_{jn},$$
$$\mathbf{r}_j = \mathbf{R}_{ijn} - (1/3)\mathbf{r}_{in} + (2/3)\mathbf{r}_{jn},$$
$$\mathbf{r}_n = \mathbf{R}_{ijn} - (1/3)\mathbf{r}_{in} - (1/3)\mathbf{r}_{jn}.$$

Next, we'll use the property of rapid decay of functions U_{ij} and α_{ij} with increasing distance and perform decomposition of all functions in the integral expression (delta functions and two wave functions) by the relative distance between particles. Let's note the necessary elements of the wave function argument structure

$$\Psi(R,t) = \Psi(\ldots,\mathbf{r}_i,\ldots,\mathbf{r}_j,\ldots,\mathbf{r}_n,\ldots,t).$$

The derivation method is discussed in more detail in the Appendix. Note that a similar method is used in deriving other equations in the presented work.

The expression obtained in the first order of the relative distance between particles leads to the following expression for polarization evolution (in zero order, the polarization derivative equals zero):

$$\partial_t P^{\alpha} = \frac{1}{3}\varepsilon^{\alpha\beta\gamma}g_{u(\alpha)}[S^{\gamma}\partial_{\beta}\pi^{\sigma\sigma} - \pi^{\sigma\sigma}\partial_{\beta}S^{\gamma} +$$
$$+\pi^{\gamma\sigma}\partial_{\beta}S^{\sigma} - S^{\sigma}\partial_{\beta}\pi^{\gamma\sigma}], \quad (28)$$

which is valid for atoms/ions with arbitrary spin. Equation (28) contains the following interaction constant:

$$g_{u(\alpha)} = \int \xi^2 U(\xi)\alpha(\xi)d\xi.$$

The calculation method can be found in work [27], where it is considered for another type of physical systems but contains an analogy in the main stages. Furthermore, equation (28) contains the nematic tensor $\pi^{\alpha\beta}$. For ions with spin different from 1/2, the anticommutator of spin operators differs from the Kronecker symbol. The quantum average of this operator gives an independent physical quantity called the nematic tensor $\pi^{\alpha\beta}(\mathbf{r},t)$, which exists alongside spin density:

$$\pi^{\alpha\beta} = \frac{1}{2}\int \Psi_S^{\dagger}\sum_i \delta(\mathbf{r}-\mathbf{r}_i)[(\hat{s}_i^{\alpha}\hat{s}_i^{\beta} + \hat{s}_i^{\beta}\hat{s}_i^{\alpha})\Psi]_S dR. \quad (29)$$

For spin 1/2 the nematic tensor becomes proportional to concentration

$$\pi^{\alpha\beta} = (\hbar^2/4)\delta^{\alpha\beta}n,$$

and equation (28) simplifies to

$$\partial_t P^{\alpha} = \frac{\hbar^2}{6}\varepsilon^{\alpha\beta\gamma} g_{u(\alpha)}[S^{\gamma}\partial_{\beta} n - n\partial_{\beta} S^{\gamma}]. \qquad (30)$$

Note that when considering Hamiltonian (25), the concentration does not change with time according to the continuity equation but can be inhomogeneous in space.

4.1. Spin Density Evolution Equation

When justifying the spin-current model, we considered a Hamiltonian containing several types of interactions (5), while the main result of this work (28) and (30) was obtained for evolution under the Heisenberg Hamiltonian. Here is the corresponding spin evolution equation

$$\partial_t \mathbf{S} = \frac{1}{6} g_u [\mathbf{S}, \Delta\mathbf{S}], \qquad (31)$$

where

$$g_u = \int \xi^2 U(\xi) d\xi.$$

Note that the rigorous derivation of the motion equation for magnetization/spin density (31) in the continuum approximation was performed using the quantum hydrodynamics method in work [16].

4.2. Nematic Tensor Evolution Equation

To obtain a closed system of quantum hydrodynamics equations, we need to derive the evolution equation for the nematic tensor (29) under the exchange interaction (25):

$$\partial_t \pi^{\alpha\beta} = g_{0u}[\pi^{\alpha\gamma}\varepsilon^{\beta\gamma\sigma} + \pi^{\beta\gamma}\varepsilon^{\alpha\gamma\sigma}]S^{\sigma}, \qquad (32)$$

where the coefficient

$$g_{0u} = \int U(\xi) d\xi.$$

For spin 1/2 we have $\pi^{\alpha\beta} = (\hbar^2/4)\delta^{\alpha\beta} n$, the left side reduces to $\partial_t n$, and the right side of equation (32) becomes zero in accordance with the continuity equation for the considered Hamiltonian.

5. CONCLUSIONS

An evolution equation for electric polarization in type II multiferroics is proposed, where the crystal cell polarization is formed proportionally to the vector product of spins of its magnetic ions. The equation is derived using the quantum hydrodynamics method as part of a closed system of equations, including evolution equations for spin density, nematic tensor, and concentration.

Justification of the spin-current model for multiferroic polarization is provided based on the quantum hydrodynamics method. The force density acting from the electric field on the electric dipole moment and the spin-orbital interaction force density have the same structure, leading to the possibility of balance between these forces. This allows introducing an effective electric dipole moment density caused by spin-orbital interaction and proportional to the spin current. Using the spin current from the spin evolution equation gives the final macroscopic expression. Based on this, one can reconstruct the operator corresponding to this type of polarization. As a result, we obtain the microscopic expression used to derive the polarization evolution equation.

Furthermore, an interpretation of the scalar coefficient defining the microscopic electric dipole moment through the exchange integral included in the Heisenberg Hamiltonian was obtained. This conclusion is based on using the spin-current model with spin current caused by exchange interaction in the Heisenberg Hamiltonian approximation.

6. DATA AVAILABILITY

The question of data availability is not applicable to this article, as this purely theoretical study did not generate or analyze any new data.

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APPENDIX. ON THE METHOD OF DERIVING THE POLARIZATION EVOLUTION EQUATION

When averaging the commutator (27) over wave functions (26), we perform substitution of spatial variables in the *i*-th, *j*-th, and *n*-th arguments when considering the function of three coordinates under the integral (*i*-th and *j*-th when considering the function of two coordinates). Variable substitution in the delta function is also performed. Further, we use the fact that the integrand functions rapidly decay with increasing relative distance between ions, which corresponds to the nearest-neighbor interaction approximation. This property of functions $U(r)$ and $\alpha(r)$ gives us the possibility to expand the wave function and δ-function in terms of the relative distance between particles. Let us write the corresponding wave function expansion, limiting ourselves to the case when the integrand functions $U(r)$ and $\alpha(r)$ depend on coordinates of two particles. This corresponds to the first group of terms in commutator (27). Further, we obtain

$$\begin{gathered}\Psi(R,t) = \Psi(\ldots,\mathbf{r}_i,\ldots,\mathbf{r}_j,\ldots,t) = \\ = \Psi(R,t) = \Psi\left(\ldots,\mathbf{R}_{ij} + \frac{1}{2}\mathbf{r}_{ij},\ldots,\mathbf{R}_{ij} - \frac{1}{2}\mathbf{r}_{ij},\ldots,t\right) \approx \\ \approx \Psi(R,t) = \Psi(\ldots,\mathbf{R}_{ij},\ldots,\mathbf{R}_{ij},\ldots,t) + \\ + \frac{1}{2}\mathbf{r}_{ij}\left(\frac{\partial}{\partial \mathbf{R}_{ij,1}} - \frac{\partial}{\partial \mathbf{R}_{ij,2}}\right)\Psi(\ldots,\mathbf{R}_{ij,1},\ldots,\mathbf{R}_{ij,2},\ldots,t) + \\ + \frac{1}{2^3} r_{ij}^{\alpha} r_{ij}^{\beta}\left(\frac{\partial}{\partial R_{ij,1}^{\alpha}} - \frac{\partial}{\partial R_{ij,2}^{\alpha}}\right)\times \\ \times\left(\frac{\partial}{\partial R_{ij,1}^{\beta}} - \frac{\partial}{\partial R_{ij,2}^{\beta}}\right)\Psi(\ldots,\mathbf{R}_{ij,1},\ldots,\mathbf{R}_{ij,2},\ldots,t), \quad (33) \\ \mathbf{R}_{ij,1} = \mathbf{R}_{ij,2} = \mathbf{R}_{ij}\end{gathered}$$

are defined for ions located in the i-th and j-th cells respectively. Additional indices 1 and 2 are used to specify which argument of the many-particle wave function is being differentiated, since as a result of Taylor series expansion, we obtained that these arguments contain the same variable.

Note the presence of spin indices in the considered many-particle wave function (wave spinor)

$$\begin{gathered}\Psi(R,t) = \Psi_S(R,t) = \\ = \Psi_{\ldots,s_i,\ldots,s_j,\ldots,s_n,\ldots}(\ldots,\mathbf{r}_i,\ldots,\mathbf{r}_j,\ldots,\mathbf{r}_n,\ldots,t)\end{gathered}$$

and the action of spin operators on this wave spinor

$$\begin{gathered}\hat{s}_i^{\alpha}\Psi(R,t) = (\hat{s}_i^{\alpha}\Psi)_S(R,t) = \\ = \hat{s}^{\alpha}_{s_i s_{i'}}\Psi_{\ldots,s_{i'},\ldots,s_j,\ldots,s_n,\ldots}(\ldots,\mathbf{r}_i,\ldots,\mathbf{r}_j,\ldots,\mathbf{r}_n,\ldots,t),\end{gathered}$$

as well as the formula for calculating average values taking into account the spinor structure

$$F = \int \Psi_S^{\dagger}\sum_{i,j\neq i}\delta(\mathbf{r}-\mathbf{r}_i)(\hat{F}_{ij}\Psi)_S dR. \quad (34)$$

Changes in the form of arguments in the coordinate part of the wave function do not result in changes in the form of spin operators' action.

Separately, we note the result of the expansion of δ-function:

$$\begin{gathered}\delta(\mathbf{r}-\mathbf{r}_i) = \delta(\mathbf{r}-\mathbf{R}_{ij}-(1/2)\mathbf{r}_{ij}) \approx \\ \approx \delta(\mathbf{r}-\mathbf{R}_{ij}) - \frac{1}{2}\mathbf{r}_{ij}\frac{\partial}{\partial \mathbf{r}}\delta(\mathbf{r}-\mathbf{R}_{ij}) + \\ + \frac{1}{2^3} r_{ij}^{\alpha} r_{ij}^{\beta}\frac{\partial}{\partial r^{\alpha}}\frac{\partial}{\partial r^{\beta}}\delta(\mathbf{r}-\mathbf{R}_{ij}) + \ldots \quad (35)\end{gathered}$$

Let's also present how this expansion manifests in the structure of the macroscopic function:

$$\begin{gathered}F(\mathbf{r},t) = \int \Psi_S^{\dagger}\sum_{i,j\neq i}\delta(\mathbf{r}-\mathbf{r}_i)(\hat{F}_{ij}\Psi)_S dR \approx \\ \approx \int \Psi_S^{\dagger}\sum_{i,j\neq i}\delta(\mathbf{r}-\mathbf{R}_{ij})(\hat{F}_{ij}\Psi)_S dR - \\ - \frac{1}{2}\frac{\partial}{\partial r^{\alpha}}\int \Psi_S^{\dagger}\sum_{i,j\neq i}\delta(\mathbf{r}-\mathbf{R}_{ij})r_{ij}^{\alpha}(\hat{F}_{ij}\Psi)_S dR + \\ + \frac{1}{2^3}\frac{\partial}{\partial r^{\alpha}}\frac{\partial}{\partial r^{\beta}}\int \Psi_S^{\dagger}\sum_{i,j\neq i}\delta(\mathbf{r}-\mathbf{R}_{ij})r_{ij}^{\alpha}r_{ij}^{\beta}(\hat{F}_{ij}\Psi)_S dR, \quad (36)\end{gathered}$$

where derivatives with respect to coordinate $\mathbf{r}$ can be taken outside the integral. Note that formula (36) represents a partial result, and the final expression for the function expansion $F(\mathbf{r},t)$ appears when considering the expansion of wave functions.

After taking into account the short-range nature of interaction, there arises a need for approximate consideration of the many-particle wave function to obtain a closed mathematical apparatus.

In general, for a many-particle system with complex spin configuration, the exact wave function cannot be represented as a product of a function depending on spatial coordinates and a function depending only on spin variables. The general form of the function is determined by the superposition of such products, with separate groups corresponding to different values of the total spin of a pair or triple of considered particles. This is especially significant for systems with spin-orbital interaction, which is considered in Hamiltonian (5). Nevertheless, we limit ourselves to representing the wave function as a product of single-particle wave functions, assuming that a more detailed consideration of the wave function structure will give corrections to the proposed "main field" approximation. Such approximation proved sufficient when deriving the contribution of Heisenberg exchange interaction to the Landau-Lifshitz equation [16]. Therefore, this approximation is used despite accounting for spin-orbital interaction, which is a small relativistic effect. Note that, as shown above, polarization is caused by spin-orbital interaction. However, the evolution of polarization and other macroscopic functions occurs under the influence of a set of interactions, and spin-orbital interaction can be considered small compared to them.

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