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Toroidal ordering in crystals

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Contents

Abstract. Phase transitions to ordered states, which correspond to the toroidal family of multipoles known in electrodynamics, are discussed. The ordering of toroidal moments may either occur simultaneously with their formation (as in superconductivity) or may follow it (at lower temperatures). In addition to electrodynamic toroidal moments corresponding to either poloidal charge currents or a spin configuration, a toroidal state corresponding to poloidal spin currents is possible.

1. Introduction

A review of theoretical works on spontaneous currents and appropriate toroidal states is given for both the weak electron–electron interaction with a nesting of Fermi surfaces (model of an excitonic insulator) and an extremely strong interaction (Hubbard model). For the intermediate case, a number of studies are considered in which states with spontaneous currents have been obtained within the framework of the Fermi-liquid theory.

The review describes the magnetoelectric properties of the toroidal state which is characterized by anomalously high values of magnetoelectric coefficients. It is mainly this property that drew the attention of specialists in magnetoelectricity and led to the detection of such a class of systems.

Collective toroidal modes are characterized by the proximity or even coincidence of resonance frequencies in the dielectric and magnetic susceptibilities. As a result, a region of frequencies exists in which the refractive index becomes negative, i.e., the toroidal state belongs to the class of metamaterials.

This review also describes the behavior of toroidal states in a magnetic field. A homogeneous toroidal state has an anomalous response near the point of phase transition to a nonuniform magnetic field or to an external current which is a thermodynamically conjugate field with respect to it. As to an anomalous diamagnetic response to a uniform magnetic field, it is exhibited by an inhomogeneous toroidal state, in particular, antitoroidal.

Two different reasons for the appearance of a large diamagnetism of inhomogeneous toroidal states are possible.

In the case of the predominance of the first spatial derivatives of the toroidal moment, an effective magnetic field arises in the system (even in the absence of an external magnetic field, in contrast to the case of the fractional quantum Hall effect). If the effective magnetic length applicable to this field is less than the characteristic scale of inhomogeneity, then the response to the external field corresponds to the differential susceptibility of the de Haas–van Alphen regime for a large effective magnetic field. In this case, the response can correspond to ideal diamagnetism.

If the second derivatives are predominant, in which case the effective magnetic length proves to be greater than the characteristic scale of inhomogeneity, the response is determined by the precession of the current loops of a large correlation radius, tightly nested into each other. In this case, ideal diamagnetism is also possible at the point of the phase transition.

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2. Groups of time reversal and space inversion

Phase transitions in crystals are characterized by a reduction in the macroscopic symmetry with the appearance of an appropriate order parameter.

Apart from the earlier known spatial operations of symmetry, including the operation of space inversion, $r \to -r$, in 1932 Wigner [1] introduced the operation of time reversal. The axial vectors of the magnetic field (**H**) and magnetization (**M**) reverse their sign in the operation $t \to -t$, whereas the polar vectors of the electric field (**E**) and polarization (**P**) do not change their signs. In 1957, in connection with the parity-nonconservation reactions accompanying weak interactions in nuclear physics, Ya B Zel'dovich [2] introduced a classical image of a toroidal solenoid (anapole), where the polar vector changes sign both as $t \to -t$ and $r \to -r$. In the physics of the condensed state, Ascher [3, 4] suggested that it is the current density **j** that can play the role of such a vector. Vectors of such a symmetry can exist in crystals of 31 classes (out of 122 magnetic classes) of the Shubnikov point group; in 13 classes out of these 31 classes, a vector **M** can also exist. Ascher also indicated other possible vectors (momentum **p**, velocity **v**, etc.) with such a symmetry. On the basis of closed loops of such spontaneous currents, Ascher erroneously attempted to explain the basic properties of superconductors. It should be noted that earlier [5–8] it was supposed that the current **j** can be used as the order parameter for superconductivity. The existence of a uniform current for the ground state of an equilibrium system contradicts the known Bloch theorem [9]. The situation with nonuniform currents that was considered by Ascher [3, 4] contradicts the condition of the gauge invariance, since a variation in the term **jA** (**A** is the vector potential) in the expression for free energy with respect to the order parameter **j** leads to the appearance in the induced parameter and in the energy of a contribution proportional to the vector potential, which has not to take place. As was shown by Ginzburg et al. [10], the role of this order parameter can be played by the polar *t*-asymmetric vector **T** of the toroidal moment. The existence of an appropriate multipole in electrodynamics was discovered in the 1970s by the group of V M Dubovik [11, 12]. The manifestation of the toroidal moment in atomic physics was first investigated in Ref. [13].

Thus, for reasons of symmetry, four order parameters can be introduced for the space–time inversion:

(1) a *t*- and *r*-even axial vector **G**. As examples, vectors such as the vector **n** in liquid crystals [14], the order parameter in spin nematics [15], the axial toroidal moment caused by poloidal spin currents (see Section 4.2 and Ref. [16]), and the vector of spontaneous mechanical stresses in ferroelastics can serve;

(2) *t*-even and *r*-odd polar vector **P** of the electric polarization, which describes a seignettoelectric (ferroelectric) state. The role of a thermodynamically conjugate field is played by an electric field **E** which aligns the domains in one direction; the response to the electric field diverges at the point of the phase transition;

(3) *t*-odd and *r*-even axial magnetization vector **M**. The role of the thermodynamically conjugate field is played by the magnetic field **H** which aligns the magnetic domains of the ferromagnet; the response to the magnetic field diverges at the point of the phase transition;

(4) *t*-odd and *r*-odd polar vector **T** which describes a toroidal state. The role of the thermodynamically conjugate field is played by an external current **j** which aligns toroidal domains; the response to the current diverges at the point of the corresponding phase transition.

3. Multipole expansions in classical electrodynamics

Let us examine the problem of finding the time-averaged magnetic field $\langle \mathbf{H}(\mathbf{R}) \rangle$ at the point **R** in the case of a stationary distribution of classical electric currents [18]

$$\mathbf{j}(\mathbf{r}) = \sum_\alpha e_\alpha \dot{\mathbf{r}}_\alpha \, \delta(\mathbf{r} - \mathbf{r}_\alpha)$$

of electrons moving with velocities $\dot{\mathbf{r}}_\alpha$ at the points $\mathbf{r}_\alpha$.

For an electrodynamic vector potential $\langle \mathbf{A} \rangle$ that satisfies the conditions $\langle \mathbf{H} \rangle = \nabla \times \langle \mathbf{A} \rangle$ and $\nabla \langle \mathbf{A} \rangle = 0$, we obtain [17]

$$\langle \mathbf{A}(\mathbf{R}) \rangle = \frac{1}{c} \int \mathrm{d}^3 r \, \frac{\langle \mathbf{j}(\mathbf{r}) \rangle}{|\mathbf{R} - \mathbf{r}|} = \frac{1}{c} \left\langle \sum_\alpha \frac{e_\alpha \dot{\mathbf{r}}_\alpha}{|\mathbf{R} - \mathbf{r}_\alpha|} \right\rangle , \tag{1}$$

and its multipole expansion takes on the form

$$\langle \mathbf{A}(\mathbf{R}) \rangle = \frac{1}{c} \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \left\langle \sum_\alpha e_\alpha \dot{\mathbf{r}}_\alpha \, (\mathbf{r}_\alpha \nabla)^n \frac{1}{\mathbf{R}} \right\rangle . \tag{2}$$

We have

$$\langle \mathbf{A} \rangle^0 = \frac{1}{c} \left\langle \sum_\alpha e_\alpha \dot{\mathbf{r}}_\alpha \right\rangle = 0 ,$$

since the average of the time derivative is equal to zero:

$$\langle \mathbf{A} \rangle^{(1)} = -\mathbf{m} \times \nabla \left(\frac{1}{R} \right) ,$$

where

$$\mathbf{m} = \frac{1}{2c} \left\langle \sum_\alpha \left[\mathbf{r}_\alpha \times \dot{\mathbf{r}}_\alpha \right] \right\rangle$$

is the magnetic dipole moment of the system.

The second-order multipole expansion represents a sum of a quadrupole (q_{ij}) term

$$\langle A_i \rangle^{(2)}_{\mathrm{quad}} = -\varepsilon_{ijk} q_{kl} \nabla_j \nabla_l \frac{1}{R} ,$$

where

$$q_{ij} = \frac{1}{6c} \left\langle \sum_\alpha e_\alpha \left(\left[\mathbf{r}_\alpha \times \dot{\mathbf{r}}_\alpha \right]_i r_{\alpha j} + \left[\mathbf{r}_\alpha \times \dot{\mathbf{r}}_\alpha \right]_j r_{\alpha i} \right) \right\rangle , \tag{3}$$

and a toroidal term

$$\langle \mathbf{A} \rangle^{(2)}_{\mathrm{tor}} = \nabla (\mathbf{t} \nabla) \frac{1}{R} + 4\pi \, \mathbf{t} \, \delta(\mathbf{R}) ,$$

$$\mathbf{t} = \frac{1}{6c} \left\langle \sum_\alpha e_\alpha \left(\mathbf{r}_\alpha \times [\mathbf{r}_\alpha \times \dot{\mathbf{r}}_\alpha] \right) \right\rangle = -\frac{1}{4c} \left\langle \sum_\alpha e_\alpha r_\alpha^2 \, \dot{\mathbf{r}}_\alpha \right\rangle$$

$$= -\frac{1}{4c} \left\langle \int \mathrm{d}^3 r r^2 \, \mathbf{j}(\mathbf{r}) \right\rangle . \tag{4}$$

The following equivalent form is encountered more frequently:

$$\mathbf{t} = \frac{1}{10c} \left\langle \sum_\alpha \left(\mathbf{r}_\alpha (\mathbf{r}_\alpha \, \mathbf{j}_\alpha) - 2 r_\alpha^2 \, \mathbf{j}_\alpha \right) \right\rangle . \tag{5}$$

For systems in which the magnetic fields are induced by spins rather than by moving charges, the *electric* current (but not the current of *spins*) is defined as the variational derivative of the energy with respect to the vector potential:

$$\mathbf{j}(r) = c\frac{\delta}{\delta \mathbf{A}}\left\{-g\mu_{\mathrm{B}}\int \mathrm{d}^3 r\, \mathbf{S}[\nabla\times\mathbf{A}]\right\}, \tag{6}$$

where g is the gyromagnetic ratio, μ_{B} is the Bohr magneton, and $\mathbf{S}(\mathbf{r})$ is the spin density.

Substituting $\mathbf{j}(r)$ defined by formula (6) into Eqn (5), we obtain

$$\mathbf{t} = \frac{g\mu_{\mathrm{B}}}{2}\left\langle \int \mathrm{d}^3\mathbf{r}\,\left[\mathbf{r}\times\mathbf{S}(\mathbf{r})\right]\right\rangle .$$

The interaction of H_{int} with the magnetic field $\mathbf{H}(\mathbf{r})$ at the point $\mathbf{r} = 0$ is described as follows [17]:

$$H_{\mathrm{int}} = -\mathbf{m}\,\mathbf{H}(0) - \mathbf{t}\,[\nabla\times\mathbf{H}]_{r=0} - q_{ij}(\partial_i H_j + \partial_j H_i)_{r=0}\,. \tag{7}$$

Thus, the interaction of the toroidal moment occurs with rot $\mathbf{H}$, i.e., it is nonzero only in the presence of an electric current. Just as for the electric polarization, there are some subtle details in the determination of the toroidal moment of periodic systems, as well [17].

In the case of an inhomogeneous toroidal moment $\mathbf{T}(r)$, the spontaneous macroscopic current has the form [12]

$$\mathbf{j}(r) = \mathrm{rot}\,\mathrm{rot}\,\mathbf{T}(\mathbf{r})\,. \tag{8}$$

In the conclusive part of the article [12], a review is given of works (as of 1990) devoted to the manifestation of toroidal moments in atomic and nuclear physics, and the physics of elementary particles, including leptons and quarks, and Z and W bosons.

4. Microscopic models of toroidal ordering

4.1 Model of an excitonic insulator

The question of the appearance of ordered current states was first examined in Ref. [19] on the basis of a two-band model, with the Fermi surfaces of electrons and holes coinciding in the momentum space (nesting of Fermi surfaces) — the model of an excitonic insulator [20].

A specific feature of this model is the fact that at temperatures lower than a certain critical point (T_{c}), pairs consisting of an electron and a hole are formed simultaneously with Bose condensation, which leads to the formation of an insulating (semiconducting) state and the appearance of an insulator gap Δ as the order parameter. In the case of ideal nesting, this solution appears even in the presence of an infinitely weak Coulomb interaction and is asymptotically exact. Formally, this solution is analogous to that obtained in the Bardeen–Cooper–Schrieffer (BCS) theory [21] for superconducting electron–electron (Cooper) pairing due to the electron–phonon (rather than Coulomb) interaction, but hindered by the Coulomb interaction weakened by Tolmachev's logarithm [22].

In addition, there are some other fundamental distinctions compared to Cooper superconductive pairing:

(1) since the behavior of an electron relative to a hole does not obey the Pauli exclusion principle, there is a degeneracy relative to singlet and triplet pairings;

(2) since pairing occurs between states from different bands and, generally speaking, with Bloch functions of different symmetries, the physical properties of the resultant insulating state will depend on the relative symmetry of the wave functions of these bands or, to be more exact, on the type of nonzero interband matrix elements;

(3) the interband matrix elements (hybridization) can fix the phase of the insulating order parameter [23], thereby affecting the physical properties of the system.

Notice that in the case of superconducting pairing the phase of the order parameter remains arbitrary (which, by the way, is of primary importance for the manifestation of the superconducting properties), and the gradient of the phase manifests itself, which determines the magnitude of superconducting current.

As a result, the order parameter can be represented in the form of a matrix $\hat{\Delta}$ in spin space:

$$\hat{\Delta} = \Delta^{\mathrm{s}}_{\mathrm{Re}} + \mathrm{i}\Delta^{\mathrm{s}}_{\mathrm{Im}} + \boldsymbol{\sigma}\left(\boldsymbol{\Delta}^{\mathrm{t}}_{\mathrm{Re}} + \mathrm{i}\boldsymbol{\Delta}^{\mathrm{t}}_{\mathrm{Im}}\right),$$

where $\boldsymbol{\sigma}$ is a vector constructed from Pauli matrices, $\Delta^{\mathrm{s}}_{\mathrm{Re,\,Im}}$ are the singlet real and imaginary components of the order parameter, and $\boldsymbol{\Delta}^{\mathrm{t}}_{\mathrm{Re,\,Im}}$ are the triplet real and imaginary components.

Let us note once again that the physical properties of the system will depend not only on which of these four parameters or their combination corresponds to the ground state, but also on the type of nonzero interband matrix elements. It was emphasized in Ref. [19] that $\Delta^{\mathrm{s}}_{\mathrm{Im}}$ and $\boldsymbol{\Delta}^{\mathrm{t}}_{\mathrm{Im}}$ can be connected with current states, but the nature of these current states and the structure of the matrix elements have not been investigated.

Which of these four components of the order parameter or their combination will be realized depends on the relationship between the intraband and interband Coulomb and electron–phonon interactions [24, 25], scattering by impurities [26], spin–orbit interaction [27], violation of the condition of ideal nesting, for example, due to doping [28], and the structure of domain walls in a ferroelectric [29–31].

The issue of the structure of current states in the case of a nonzero interband matrix element of the momentum p_{12} was examined in Ref. [32]. In that study, the expression for the current took into account only interband components (this seemed natural, since the order parameter is determined by interband electron–hole pairing), which led to the presence of a uniform current component.

This result, on the one hand, was like a microscopic confirmation of the phenomenological results obtained in Refs [3–8], where the current (in this case, through the combination $p_{12}\Delta^{\mathrm{s}}_{\mathrm{Im}}$) was identified with the order parameter $\Delta^{\mathrm{s}}_{\mathrm{Im}}$. But, on the other hand, this result contradicts (as was noted above) both the Bloch theorem and the condition of gauge invariance. There even arose a certain discussion [33–35] concerning the elimination of the uniform current. In Ref. [33], a procedure was proposed for redefining the operator of current at the expense of the exchange energy. It was shown in Ref. [28] that the exchange correction does not contribute to the average current.

The authors of Ref. [35] used a model differing from that employed in Ref. [32] for eliminating the uniform current, in which the interband momentum operator reverses sign while moving over the Brillouin zone.

In reality, the compensation for the uniform interband component obtained in Ref. [32] occurs depending on the

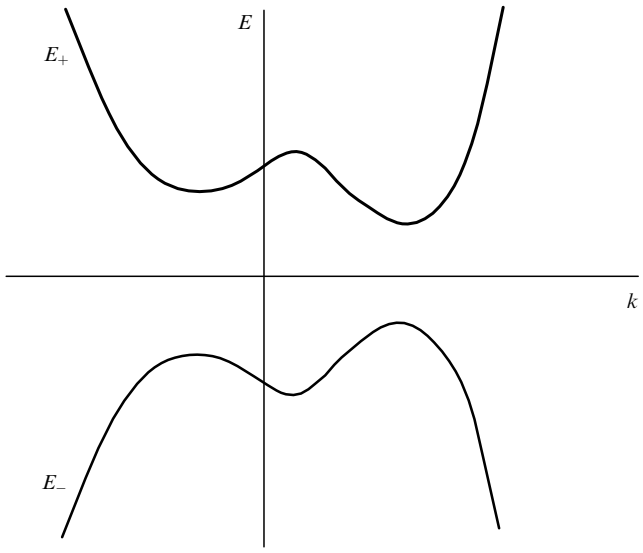

Figure 1.

magnitude of the intraband current components [36] which appear as a result of the 'skew' nature of the electron spectrum containing a momentum-asymmetrical component $\mathbf{p}_{12}\mathbf{p}\Delta^{\rm s}_{\rm Im}$, which was overlooked in Ref. [32].

The spectrum of electronic excitations in the insulating phase takes the following form (see Fig. 1):

$$E(p) = \pm\sqrt{\varepsilon^2 + \left|\,\mathrm{i}\,\mathbf{p}_{12}\,\mathbf{p} - \Delta^{\rm s}_{\rm Im}(T)\right|^2}\,, \tag{9}$$

where $\varepsilon_1(p) = -\varepsilon_2(p) \equiv \varepsilon(p)$.

In Ref. [36], an expression was obtained for the current with the availability of an inhomogeneous order parameter $\Delta^{\rm s}_{\rm Im}(r)$:

$$\mathbf{j}(\mathbf{r}) = \mathrm{rot}\,\mathrm{rot}\,\mathbf{p}_{12}\Delta^{\rm s}_{\rm Im}\,. \tag{10}$$

One can readily see from a comparison of the expression for the current (10) with expression (8) that it is precisely the combination $\mathbf{p}_{12}\Delta^{\rm s}_{\rm Im}$ that takes the part of the toroidal order parameter (this was first inferred by V L Ginzburg):

$$\mathbf{T} = \mathbf{p}_{12}\,\frac{\Delta^{\rm s}_{\rm Im}}{m_0}\,. \tag{11}$$

A more general proof of the absence of a uniform current in the state with $\mathbf{T}(\mathbf{r}) = \mathrm{const}$ was given in Ref. [28]. It was shown that for the gauge-invariant local Hamiltonian with an interaction that depends only on the difference in the coordinates, all three methods of determining the operator of current $\hat{\mathbf{j}}$, i.e.,

• from the equation of motion for the coordinate operator $\hat{\mathbf{r}}$:

$$\dot{\hat{\mathbf{r}}} = \mathrm{i}[\hat{H}, \hat{\mathbf{r}}]\,; \tag{12}$$

• from the continuity equation for the charge-density operator $e\hat{n}(\mathbf{r})$:

$$\frac{\partial e\hat{n}(\mathbf{r})}{\partial t} = \mathrm{i}\big[\hat{H}, e\hat{n}(\mathbf{r})\big] \equiv -\mathrm{div}\,\hat{\mathbf{j}}\,, \tag{13}$$

• and from the variation of $\hat{H}$ over the vector potential $\hat{\mathbf{A}}$:

$$\hat{\mathbf{j}} = c\,\frac{\delta\hat{H}}{\delta\hat{\mathbf{A}}}\,, \tag{14}$$

lead to one and the same result (10).

It was shown that in the case of doping a contribution to the current also appears from the real part $\Delta^{\rm s}_{\rm Re}$ of the order parameter, which is antisymmetric with respect to the electron momentum.

Until now, we have been considering the case where the wave functions of the electron (subscript 1) and hole (subscript 2) zones have the opposite parity and the interband matrix element p_{12} of the momentum is nonzero.

However, in the case of zones with identical parity, we have $\mathbf{p}_{12} \equiv 0$, and the interband matrix element of the angular momentum operator is nonzero.

In this situation, the electron spectrum proves to be symmetric in momentum, the intraband components of current are $\mathbf{j}_{11} = \mathbf{j}_{22} = 0$, and the expression for the current has the form [37]

$$\mathbf{j} = \mathbf{j}_{12} + \mathbf{j}_{21} = \mathrm{rot}\,\mathbf{M}(\mathbf{r})\,, \tag{15}$$

$$\mathbf{M} = 2\mathbf{I}\Delta^{\rm s}_{\rm Im}\,\frac{1}{g^{\rm s}_{\rm Im}}\,, \tag{16}$$

$$\mathbf{I} = \frac{1}{2}\,\mu_{\rm B}\sum_{n\neq 1,2}\left(\frac{1}{E_1 - E_n} + \frac{1}{E_2 - E_n}\right)\frac{[\mathbf{p}_{1n}\times\mathbf{p}_{n2}]}{m_0}\,, \tag{17}$$

where m_0 is the mass of free electron; E_1 is the position of the point of the minimum of band 1; E_2 is the position of the maximum of band 2; $\mathbf{p}_{1n}$ and $\mathbf{p}_{n2}$ are the matrix elements of the momentum operator of bands 1 and 2, respectively, with other bands n, and $g^{\rm s}_{\rm Im}$ is the effective coupling constant corresponding to the parameter $\Delta^{\rm s}_{\rm Im}$.

Thus, a ferromagnetic state of the wholly filled band then appears in the insulating phase.

4.2 Structure of the spin current state

In the case of the imaginary triplet order parameter $\mathbf{\Delta}^{\rm t}_{\rm Im}$, nonzero interband matrix elements $\mathbf{p}_{12}$, and $\Delta = \mathrm{i}\boldsymbol{\sigma}\mathbf{\Delta}^{\rm t}_{\rm Im}$, the spectrum of the excited electronic states for $\mathbf{p}_{12}||0Z||\mathbf{\Delta}^{\rm t}_{\rm Im}$ takes the following form [38] (see Fig. 2):

$$E^2_{\pm}(p) = \varepsilon^2(p) + \left(\frac{1}{m_0}\,p_{12}\,p_z \pm \Delta^{\rm t}_{\rm Im}\right)^2\,, \tag{18}$$

where the plus and minus signs correspond to opposite spin orientations.

The flux of spins is described by the second-rank tensor

$$\mathbf{J}^{\rm s}_a = \langle\sigma_a\,\hat{\mathbf{j}}\rangle\,, \tag{19}$$

where σ_a is the Pauli matrix ($a = x, y, z$), and $\hat{\mathbf{j}}$ is the current-density operator.

The macroscopic symmetry of a state with spin flux density is characterized by the symmetry of the spatial distribution of the tensor $J^{\rm s}_a$ in the unit cell of the crystal rather than by its local magnitude. The magnitude of $J^{\rm s}_a$ is invariant (in contrast to the current of charges) relative to time reversal. However, the complete symmetry group here is not the ordinary space group of symmetry, but a 'color' group which additionally includes the operations of rotation and

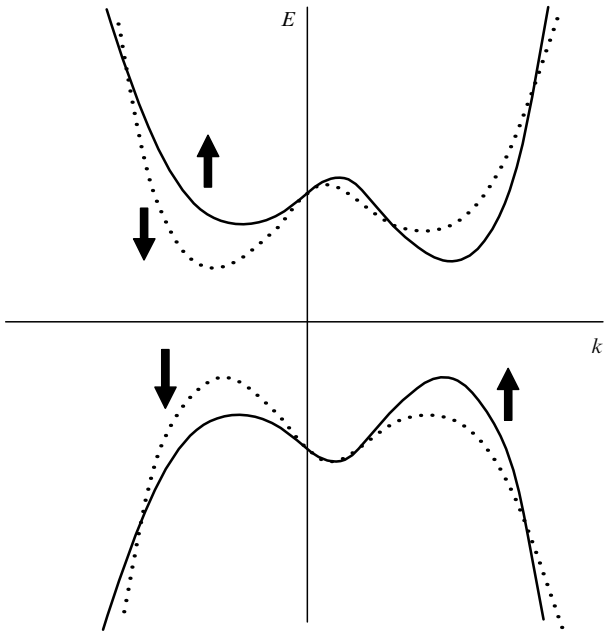

Figure 2.

reflection in spin space. If the spin component of the tensor J_a^s is coordinate-independent (i.e., the spins of all the particles that make contributions to the spin flux are parallel), then the symmetry of a state with $J_a^s \neq 0$ is described especially simply. Let the spin axes be directed along the symmetry axis of the crystal. Then, an additional symmetry operation is rotation in the spin space through an angle π. This operation is completely equivalent to the inversion of coordinates in usual space. The spin flux can be conveniently represented in the form of two equal-in-magnitude fluxes of particles with the same sign of charge, but with the opposite spin, which flow toward each other. Therefore, as in the case of the above-examined current of charges, it is easy to ascertain that the expression for the uniform current of spins reduces to the integral of the total derivative in the momentum space. Thus, the uniform spin current is equal to zero.

For a nonuniform spin current, just as for the current of charges, it is convenient to isolate the toroidal and poloidal components. The poloidal configuration is associated with the symmetry group of a pseudoscalar $\boldsymbol{\sigma}\mathbf{T}$ (where $\mathbf{T}$ is the time-odd polar vector of the toroidal magnetic moment).

Thus, the toroidal moment T_s of spin currents is expressed through Δ_{Im}^t as follows [see formula (11)]:

$$T_s = \boldsymbol{\sigma}\mathbf{T} = \frac{p_{12}}{m_0}\,\Delta_{Im}^t\,. \tag{20}$$

Consequently, T_s, due to the order parameter Δ_{Im}^t, is one additional example of the axial vector $\mathbf{G}$ examined in Section 2.

In Ref. [38], it was shown that in this state in the presence of an electric current I a momentum asymmetry arises in the distribution of carriers, which leads to the appearance of a total magnetization $M \sim Ip_{12}\Delta_{Im}^t$ — a manifestation of the current-magnetic effect (an analog of the magnetoelectric effect for the toroids with a current of charges, considered in Section 5).

The case of a real singlet parameter Δ_{Re}^s for $\mathbf{p}_{12} \neq 0$ corresponds to the appearance of an electric polarization, i.e., to ferroelectricity [39].

In the case of noncoincident (differing by $\mathbf{q}$ in momentum space) electron (subscript 1) and hole (subscript 2) bands at $\varepsilon_1(\mathbf{p}) = -\varepsilon_2(\mathbf{p}+\mathbf{q})$ or in the one-band scheme at $\varepsilon(\mathbf{p}) = -\varepsilon(\mathbf{p}+\mathbf{q})$, the order parameter Δ_{Re}^s is associated with the initiation of a charge-density wave (CDW), and the parameter $\boldsymbol{\Delta}_{Re}^t$, with the initiation of a spin-density wave (SDW), the latter being responsible for spin antiferromagnetism. Chromium and its alloys are the examples of such a state.

Numerous examples of the realization of CDWs in quasi-one-dimensional and layered systems are well known.

The coexistence of CDWs and SDWs [40] ensures the realization of a ferromagnetic spin state, which is exemplified by hexaborides [42].

An explanation of the nature of these ferromagnets, based on the model described in Ref. [40], was given in Refs [43, 44]. There are grounds to believe that in such systems the toroidal state can compete with the ferromagnetic one.

The above examples of the realization of electron–hole pairing in the case of real order parameters Δ_{Re}^s and $\boldsymbol{\Delta}_{Re}^t$ can prove to be useful in searching for systems with toroidal orderings, keeping in mind the conditions mentioned in Section 4.1, which are favorable to the realization of current states. In this connection, a recent article [41] should be noted, which reports on the discovery of an excitonic insulator state in $TiSe_2$. In Ref. [45], for explaining the pseudogap state of cuprates, a model was suggested with spontaneous currents in the one-band scheme with $\varepsilon(p) = -\varepsilon(p+q)$ and with an insulator parameter of the d type (d-density wave, DDW), with a doubling of the period.

With the same purpose, a similar model with spontaneous currents flowing over adjacent bonds without a change in the lattice period was examined in Refs [46, 47]. Experiments exist which confirm this point of view [48, 49].

The authors of Ref. [50] indicated the magnetic classes in which toroidal states appear as a result of electron–hole pairing in the two-band model against the background of an antiferromagnetic state caused by localized spins. The appropriate examination against the background of a ferromagnetic ordering of localized spins was carried out in Ref. [51].

Kopayev et al. [52] gave a classification of magnetic materials that allow toroidal ordering near the crystal surface. An example of such an ordering is chromium in which the asymmetry of the electron spectrum, $E(\mathbf{k}) \neq E(-\mathbf{k})$, characteristic of the toroidal state was observed in experiments on the angle-resolved photoemission [53].

The occurrence of toroidal ordering near the surfaces of ferromagnets Fe and EuO follows from the asymmetry of the spectrum of spin waves, which was observed in optical experiments on Mandelstam–Brillouin scattering [54].

4.3 Hubbard model

In the limiting case of strong interaction, which is opposite to the above-considered model of an excitonic insulator, the problem of obtaining an exact solution is substantially more complicated. However, in the last 20 years huge efforts have been undertaken in the development of different methods, both analytical and numerical, of describing the ground state and the basic physical properties of systems in the limiting case of strong interaction. These efforts arose due to the need to understand the nature of both the superconductivity of cuprates and the magnetic properties of manganates which possess colossal magnetoresistance.

A solution with spontaneous currents (flux phase) was first obtained in the Hubbard model by Affleck and Marston [55]. Later, there was suggested a whole series of methods, mainly in terms of separation of charge and spin degrees of freedom, the solutions in which were nonuniform currents of charges and spins. The appropriate literature can be found in the review [56].

Notice specially the work by Bulaevskii et al. [57], in which the authors obtained a solution for a Mott insulator with spontaneous currents circulating over nearest chemical bonds. In contrast to other work with spontaneous currents, a structure which allows an exact solution was suggested for the Mott insulator. Systems have been considered based on a triangular structure with a nonzero average scalar chirality:

$$\langle \chi_{123} \rangle = \left\langle [\mathbf{S}_1 \times \mathbf{S}_2]\, \mathbf{S}_3 \right\rangle , \tag{21}$$

where 1, 2, 3 are the order numbers of the sites of this triangular structure, and $\mathbf{S}_i$ is the operator of spin on the ith site.

In Ref. [57], one can find references to articles in which the presence of such an average leads to the emergence of spontaneous currents in another (not-Hubbard) subsystem with conduction electrons, which by itself is of interest in connection with the problem under consideration. In the same work, the scalar chirality and orbital currents are caused by one and the same electrons.

If we introduce an operator of spin at the ith site as

$$S_i^{\eta} = \sum_{\mu\nu} C_{i\mu}^{+} \sigma_{\mu\nu}^{\eta} C_{i\nu} , \tag{22}$$

where $\eta = \{x, y, z\}$, $\sigma_{\mu\nu}^{\eta}$ are the Pauli matrices, and $C^{+}(C)$ are the operators of the creation (annihilation) of an electron, then the Hamiltonian in such a representation (one electron at a site, and the degree of freedom only in spin) will take the form

$$H = \sum_{ij} J_{ij} \left(\mathbf{S}_i \mathbf{S}_j - \frac{1}{4} \right) , \tag{23}$$

where $J_{ij} = 4t_{ij}^2/U$, t_{ij} is the integral of hopping between the ith and jth sites, and U is the electron–electron interaction at a site.

Then, the operator of the current flowing between sites i and j may be represented in the form

$$\hat{I}_{ij} = \frac{\sqrt{-1} e t_{ij} \mathbf{r}_{ij}}{\pi r_{ij}} \sum_{\sigma} (C_{j\sigma}^{+} C_{i\sigma} - C_{i\sigma}^{+} C_{j\sigma}) , \tag{24}$$

and we obtain the following expression for the contribution from the bond 1–2 to the current over the triangle 1–2–3:

$$\hat{\tilde{I}}_{123} = \frac{\mathbf{r}_{12} 24 e t_{12} t_{23} t_{31}}{r_{12} \hbar U^2} \, [\mathbf{S}_1 \times \mathbf{S}_2]\, \mathbf{S}_3 . \tag{25}$$

Equation (25) shows that the presence of spin chirality on a triangular structure provides orbital current. This current results in a nonzero magnetic moment L_z directed perpendicularly to the plane of the triangle, with the average moment $\langle L_z \rangle$ being proportional to the angle formed by vectors $\langle \mathbf{S}_1 \rangle$, $\langle \mathbf{S}_2 \rangle$, and $\langle \mathbf{S}_3 \rangle$. Note that this state arises even at an arbitrarily small parameter t_{ij}/U.

The above conditions are like an analog of the nesting condition in the model of an excitonic insulator at a nonzero interband matrix element of the angular momentum operator, which leads to the emergence of magnetization in the opposite limiting case of arbitrarily weak interaction for $\Delta_{\mathrm{Im}}^{\mathrm{s}} \neq 0$ and $L_{12} \neq 0$ [37].

It has been recently shown [17] that a linear combination with a complex coefficient of states with opposite scalar spin chiralities results in the appearance of a toroidal moment. The phase transition to an ordered state with a toroidal moment will differ from the phase transition considered in Section 4.1 in the model of an excitonic insulator for $\Delta_{\mathrm{Im}}^{\mathrm{s}} \neq 0$ and $p_{12} \neq 0$, where the formation of toroidal moments and their ordering (Bose condensation) should occur simultaneously at the point of the phase transition.

In Ref. [57], it was also shown that for another spin structure in the triangle there can arise a seignettoelectric (ferroelectric) state in the limit of an infinitely small t/U.

If we introduce an operator $\delta n_i = n_i - 1$ of the deviation of the number of electrons at a site from unity (notice that this deviation is usually assumed to be zero in the limit of $t/U \ll 1$), we obtain, for example, for $i = 1$ that

$$\delta n_1 = n_1 - 1 = 8 \, \frac{t_{12} t_{23} t_{31}}{U^3} \left[\mathbf{S}_1 (\mathbf{S}_2 + \mathbf{S}_3 - 2\mathbf{S}_2\, \mathbf{S}_3) \right] . \tag{26}$$

Thus, if the average of the expression in brackets differs from zero, then at some sites there will be $\langle n_i \rangle > 1$, and at some others $\langle n_j \rangle < 1$ (certainly, under the condition that $\sum_{i=1}^{3} \delta n_i = 0$).

Then, an electric polarization originates in the structure, the ordering in which leads to a phase transition to a ferroelectric state. It should be noted that in the model of an excitonic insulator this state appears for $\Delta_{\mathrm{Re}}^{\mathrm{s}} \neq 0$ and $p_{12} \neq 0$.

In Ref. [58], a mechanism for the emergence of a spin nondissipative current (spin supercurrent) in a spin noncollinear magnet was suggested.

A possible analogy between the magnetic and superconducting orderings has been demonstrated. As is known, the operators of the number of particles n_i and of the Josephson phase φ_i are canonically conjugate for superconductors: $[n_i, \varphi_j] = \sqrt{-1}\delta_{ij}$, where i, j are the indices of sites.

A similar relationship also exists for spin operators: $[S_i^z, \Theta_j] = \sqrt{-1}\delta_{ij}$, where S^z is the z-component of the spin operator, and Θ is the angle of the vector (S^x, S^y). This leads to a connection between the XY spin model and the superconductivity.

The Hamiltonian in the XY model assumes the form

$$H_{XY} = \sum_{\langle ij \rangle} \frac{J_{\perp ij}}{2} \, (S_i^{+} S_j^{-} + S_i^{-} S_j^{+}) . \tag{27}$$

From the expression

$$\frac{\partial S_i^z}{\partial t} = \frac{1}{\sqrt{-1}\hbar} \, [S_i^z, H_{XY}] = -\sum_j j_{ji}^{\mathrm{s}} \tag{28}$$

we obtain the following expression for the spin current j_{ij}^{s}:

$$j_{ij}^{\mathrm{s}} = \sqrt{-1} J_{\perp ij} (S_i^{+} S_j^{-} - S_i^{-} S_j^{+}) . \tag{29}$$

By expressing $(S_j^x, S_j^y) = S(\cos\Theta_j, \sin\Theta_j)$, we arrive at

$$j_{ij}^{\mathrm{s}} = J_{ij} S^2 \sin(\Theta_i - \Theta_j) . \tag{30}$$

Equation (30) represents an analog of the Josephson equation for the superconducting current flowing between

two superconductors with a fixed phase difference. The quantity $J_\perp S^2$ plays the role of spin stiffness. In this case, an electric polarization emerges, as well. In an applied electric field, a magnetization appears. Thus, the system constitutes a magnetoelectric.

4.4 Theory of a Fermi liquid

It was shown in Sections 4.1 and 4.3 that states with spin currents and toroidal ordered states can occur both within the framework of an extremely weak interaction with a special form (nesting) of the electron spectrum and in the reverse limit of strong interaction. With strengthening interaction, the requirements for the special form of the electron spectrum become less and less rigid. Therefore, it is clear that in the intermediate situation such ordered states remain possible.

This is supported by the results of a number of studies where similar states were obtained within the framework of the Landau theory of a Fermi liquid. Pomeranchuk's work [59] proved to be at the center of attention; this work was devoted to phenomena now called Landau–Pomeranchuk instabilities of Fermi surfaces, caused by the violation of the symmetries of rotation in electronic liquid-crystal states, which were connected with the 'hidden' ordering in systems with heavy fermions. Within this approach, Varma [60] has demonstrated the transition to an insulating state with an anisotropic gap. One type of instability or another and the corresponding type of ordered state are determined by the exceeding of the critical value of a certain coefficient in the Fermi-liquid theory. It should be noted that the appropriate coefficients in the electron–hole scattering channel under the condition of nesting of the Fermi surface diverge, even at an arbitrarily weak interaction. An analogous situation takes place in the superconducting channel of scattering with a zero momentum of the electron–hole pairs for an arbitrary shape of the Fermi surface, and in the case of a finite momentum of superconducting pairs, upon fulfillment of the condition of a mirror nesting of the Fermi-surface segments [61].

All types of the ordered states examined in Section 4.1 can naturally be obtained within the framework of the Fermi-liquid theory. In connection with the manifestations of the Landau–Pomeranchuk instabilities, let us single out Ref. [62] in which, in particular, an ordered state was obtained in a triplet channel with an angular momentum equal to unity, the state which corresponds to the violation of a 'spin-orbit symmetry', when the spin–orbit interaction appears as a result of electron–electron correlations already in the non-relativistic limit, rather than as a relativistic effect. Previously, such a state was known only in the theory of superfluidity of ^{3}He.

The operator of spin current in this case assumes the following form (cf. formula (19) in Section 4.2):

$$Q^{\mu a}(\mathbf{r}) = \Psi_\alpha^+(\mathbf{r})\,\sigma_{\alpha\beta}^{\mu}(-\mathrm{i}\hat{\nabla}^a)\,\Psi_\beta(\mathbf{r})\,, \tag{31}$$

where the indices α, β, and μ indicate the direction in the spin space, and the index a, in the orbital space.

The Hamiltonian in the channel F_1^a with $l=1$ has the form

$$H = \int \mathrm{d}^3 r \Psi_\alpha^+(\mathbf{r})\left(\varepsilon(\nabla)-\mu\right)\Psi_\alpha(\mathbf{r}) + h_{\mu a}Q^{\mu a}(\mathbf{r}) + \frac{1}{2}\int \mathrm{d}^3 r\,\mathrm{d}^3 r' f_1^a(\mathbf{r}-\mathbf{r}')\,Q^{\mu a}(\mathbf{r})\,Q^{\mu a}(\mathbf{r}')\,, \tag{32}$$

where μ is the chemical potential, $\varepsilon(\nabla)$ is the operator of kinetic energy, and $h_{\mu a}$ is the external (spin-orbital) field thermodynamically conjugate to the spin current $Q^{\mu a}$, i.e., as if an infinitely small 'external' relativistic interaction induces a large nonrelativistic spin–orbit interaction at the point T_c of the corresponding phase transition.

A similar role belongs to the magnetic field in the case of ferromagnets, to the electric field in ferroelectrics, and to the electric current (as was noted in Section 4.1) in the toroidal magnetic state.

For a further discussion, it is important to retain in the dispersion relation $\varepsilon(\mathbf{k})$ not only the linear term, but also the cubic term:

$$\varepsilon(\mathbf{k}) = v_\mathrm{F}\Delta k\left[1 + b\left(\frac{\Delta k}{k_\mathrm{F}}\right)^2\right],$$

where $\Delta k = k - k_\mathrm{F}$, and k_F and v_F are the momentum and velocity of an electron on the Fermi surface, respectively.

The Fourier component $f_1^a(\mathbf{q})$ of the function $f_1^a(\mathbf{r})$ takes the form

$$f_1^a(\mathbf{q}) = \frac{f_1^a}{1+\chi|f_1^a|\,q^2}\,, \tag{33}$$

the dimensionless Landau parameter $F_1^a = Nf_1^a$, and N is the density of states at the Fermi level.

Let us define the spin-orbital susceptibility

$$\chi_{\mu a,\,\nu b} = \frac{\langle Q_{\mu a}\rangle}{h_{\nu b}}$$

in the limit of $h_{\nu b}\to 0$ as a diagonal part:

$$\chi_{\mu a,\,\nu b} = \chi_0\delta_{\mu a}\delta_{\nu b}\,.$$

Then, the Fermi-liquid correction χ_FL assumes the form

$$\chi_\mathrm{FL} = \chi_0\,\frac{m^*}{m_0}\,\frac{1}{1+F_1^a/3}\,.$$

The susceptibility χ_FL diverges at $F_1^a = -3$.

If we introduce the order parameter as

$$n^{\mu a}(\mathbf{r}) = -\int \mathrm{d}^3 r' f_1^a(\mathbf{r}-\mathbf{r}')\left\langle Q^{\mu a}(\mathbf{r}')\right\rangle, \tag{34}$$

we obtain, at $h_{\mu a}=0$, a mean-field Hamiltonian H_MF in the following form

$$H_\mathrm{MF} = \int \mathrm{d}^3 r \Psi^+(\mathbf{r})\left(\varepsilon(\nabla) - n^{\mu a}\sigma^\mu(-\mathrm{i}\nabla^a) - \mu\right)\Psi(\mathbf{r}) + Vn^{\mu a}\,\frac{n^{\mu a}}{2|f_1^a|}\,, \tag{35}$$

and the equation for the self-consistent order parameter then takes on the form

$$n^{\mu a} = |f_1^a|\int \frac{\mathrm{d}^3 k}{(2\pi)^3}\left\langle \Psi^+(k)\,\sigma^\mu \hat{k}^a \Psi(k)\right\rangle. \tag{36}$$

The last equation has two solutions:

$$n^{\mu a} = \begin{cases} \bar{n}\hat{d}_\mu\hat{e} & \text{for the } \alpha\text{-phase}\,, \\ \bar{n}D_{\mu a} & \text{for the } \beta\text{-phase}\,, \end{cases} \tag{37}$$

where $\hat{d}$ and $\hat{e}$ are the unit vectors in the spin and orbital spaces, respectively; $D_{\mu a}$ is the SO(3) matrix of rotation [62], and $\bar{n}$ is a real number.

Thus, the expression

$$\left\langle Q^{\mu a}(\mathbf{r})\, Q^{\nu b}(\mathbf{r}')\right\rangle \to \delta_{\mu\nu}\delta_{ab}\,\frac{\bar{n}^2}{|f_1^a|^2}\begin{cases}\hat{d}_\mu\hat{e}_a & \text{in the } \alpha\text{-phase},\\ D_{\mu a} & \text{in the } \beta\text{-phase},\end{cases} \tag{38}$$

for $|\mathbf{r}-\mathbf{r}'|\to\infty$ determines the off-diagonal long-range order.

At $\bar{n}^{\mu a}=\bar{n}\delta_{\mu z}\delta_{az}$, we have the following dispersion relation

$$E^A(k)_{12}=\varepsilon(k)-\mu\pm\bar{n}\cos\Theta\,, \tag{39}$$

where Θ is the angle between the vector $\mathbf{k}$ and the z-axis.

Expression (39) formally coincides with the above expression (18) for the dispersion law in the case of the toroid of spin currents, which was obtained for a triplet imaginary order with $l=0$, but now for $p_{12}\neq 0$ [38]. This solution also coincides with the spin-split state [63] which was sufficiently reasonably used for explaining the specific features of the phase transition in chromium at the Néel temperature $T_{\rm N}=311$ K instead of a transition that is usually treated as a phase transition to the spin-wave state for the real triplet order parameter $\mathbf{\Delta}^{\rm t}_{\rm Re}$.

It should be also noted that Hirsch's assertion [63] about the presence of a uniform spin current in this state due to a 'skew' spectrum (39) is incorrect. The proof to this assertion consists, as in the case with $\Delta^{\rm s}_{\rm Im}$ for $p_{12}\neq 0$ (see Section 4.1), in reducing the expression for the current of each spin component to the integral of the total derivative. A similar situation arose in the problem of ^{3}He.

The solution for the β-phase is completely new.

In this state, neither spatial inversion, nor time reversal, nor rotational symmetry are broken. For the case of $n^{\mu\alpha}=\bar{n}\delta_{\mu\alpha}$, the expression for the mean-field Hamiltonian $H_{\rm MF}$ is reduced to the following:

$$H_{\rm MF}=\sum_{\bar{k}}\Psi^+(\mathbf{k})\left(\varepsilon(\mathbf{k})-\mu-\bar{n}\,\boldsymbol{\sigma}\hat{\mathbf{k}}\right)\Psi(\mathbf{k})\,. \tag{40}$$

The dispersion law $E^B_{12}(\mathbf{k})$ for the eigenstates ±1 of the scalar chirality of the operator $\boldsymbol{\sigma}\hat{\mathbf{k}}$ assumes the form

$$E^B_{12}(\mathbf{k})=\varepsilon(\mathbf{k})-\mu\pm\bar{n}\,. \tag{41}$$

Similar to the state of the B-phase of ^{3}He, the dispersion law is isotropic. In Fig. 3, the dashed line depicts the Fermi contour of the normal phase ($\bar{n}=0$) in accordance with expression (41). For the state with $k_{\rm F2}$, the direction of the electron spin coincides with the direction of the momentum (positive chirality), while for the state with $k_{\rm F1}$, these directions are opposite (negative chirality). In spite of an outward similarity to the ferromagnetic state, the total spin for both cases ($k_{\rm F1}$ and $k_{\rm F2}$) is equal to zero.

The last term in expression (40) for $H_{\rm MF}$ corresponds to spin–orbit interaction; it is determined only by electron–electron correlations and is in no way connected with the relativity. Therefore, it can be large, especially near the corresponding transition temperature.

The α-phase is realized for the coefficient $b<1/3$ in the bare dispersion law (i.e., at $b=0$, as well).

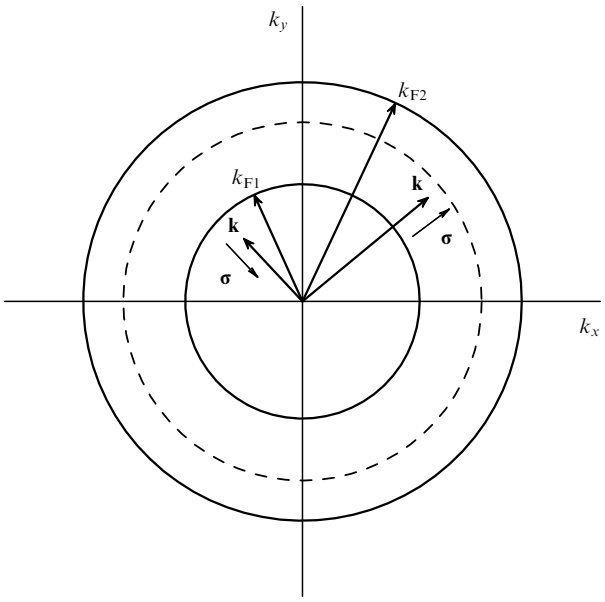

Figure 3.

The β-phase can be realized for $b>1/3$, i.e., at a significant deviation of the bare dispersion law from a linear one.

Naturally, this state can be obtained at an arbitrarily weak interaction in the model of an excitonic insulator for the triplet imaginary order parameter with an angular momentum $l=1$. The problem of the competition with other states requires detailed study. However, this problem also exists in the framework of the Fermi-liquid theory.

5. Magnetoelectric properties of the toroidal state

The family of magnetoelectrics comprises substances in which the application of a magnetic field $\mathbf{H}$ results in an electric polarization $\mathbf{P}$, and the electric field $\mathbf{E}$ generates magnetization $\mathbf{M}$ (see the review [64]).

It follows from the expression for the free energy $F(\mathbf{E},\mathbf{H})$, namely

$$F(\mathbf{E},\mathbf{H})=F_0-\frac{\varepsilon_{ij}E_iE_j}{8\pi}-\frac{\mu_{ij}H_iH_j}{8\pi}-\alpha_{ij}E_iH_j\,, \tag{42}$$

that

$$\begin{aligned}P_i&=\chi^{\rm e}_{ij}E_j+\alpha_{ij}H_j\,,\\ M_i&=\alpha_{ij}E_j+\chi^{\rm m}_{ij}H_j\,,\end{aligned} \tag{43}$$

where $\chi^{\rm e}_{ij}=(\varepsilon_{ij}-\delta_{ij})/4\pi$ and $\chi^{\rm m}_{ij}=(\mu_{ij}-\delta_{ij})/4\pi$ are the dielectric and magnetic susceptibility tensors, respectively, and α_{ij} is the magnetoelectric tensor. The term magnetoelectricity was introduced by P Debye [65]. The symmetry-based classification of crystals from the viewpoint of magnetoelectricity was performed by P Curie [66]. The tensor $\alpha_{ij}\neq 0$ is nonzero for 58 out of 122 magnetic classes.

In 1959, Dzyaloshinskii [67] predicted the possibility of the occurrence of magnetoelectricity in crystalline Cr_2O_3; in the next year, Astrov [68] revealed this effect experimentally.

From the symmetry perspective, the magnetoelectricity must exist in the toroidal state:

$$\mathbf{P}=[-\mathbf{T}\times\mathbf{H}]\,,\quad \mathbf{M}=[\,\mathbf{T}\times\mathbf{E}\,]\,, \tag{44}$$

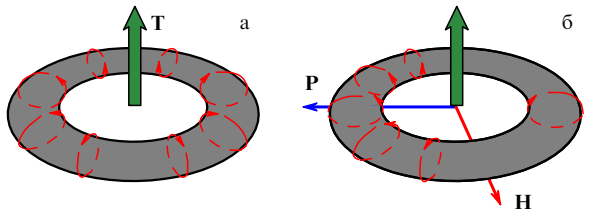

Figure 4.

i.e., the off-diagonal component of the magnetoelectric tensor must be nonzero and antisymmetric ($\alpha_{ij} = -\alpha_{ji}$) [69].

The physical nature of the occurrence of a magnetoelectric effect in the toroidal state, as well as the antisymmetry and the off-diagonal nature (transversity) of the magnetoelectric effect [70], are illustrated in Fig. 4.

Figure 4a illustrates a uniform distribution of the poloidal-current rings in the absence of a magnetic field. Figure 4b corresponds to the switching-on of a magnetic field **H** in the plane of the torus. For topological reasons, the rotation of poloidal rings with a magnetic moment oriented against the field **H** must be accompanied by their flowing from right to left.

As a result of the existence of an excess of electrons on the left-hand side, electric polarization **P** emerges. Its emergence limits the paramagnetic component of the magnetic susceptibility in the response to the field **H**; this problem will be discussed in Section 6 devoted to an analysis of the diamagnetism of the toroidal state.

From the condition for thermodynamic stability [17], we obtain the following upper bound on the magnitude of α_{ij}:

$$\alpha_{ij} \leqslant \sqrt{\chi_{ii}^{\mathrm{e}} \chi_{jj}^{\mathrm{m}}}\,. \tag{45}$$

An increase in α_{ij} should be expected near the temperature of phase transitions to the ferroelectric and (or) ferromagnetic state, where the quantities χ_{ii}^{e} and χ_{jj}^{m} diverge, respectively.

The same refers to the temperature range near the point of transition to the toroidal state, which is observed in boracites [71].

All the above results to an equal extent relate to toroidal states caused by an appropriate spin ordering.

The components of the vector **T** and, consequently, the components of the magnetoelectric tensor are transformed [72] according to:

(a) one of the one-dimensional irreducible representations, if the vector **T** is directed along the principal symmetry axis;

(b) a two-dimensional irreducible representation, if the vector **T** lies in the plane perpendicular to the principal axis, and

(c) a three-dimensional irreducible representation, for example, in cubic crystals.

In Ref. [69], it was shown that in the presence of dissipation the vector $[\mathbf{E} \times \mathbf{H}]$, which in the absence of dissipation is the source (together with current **j**) for the toroidal order parameter, responsible for the magnetoelectric effect, serves also as a source for electric polarization and, consequently, for the ferroelectric order parameter.

At the same time, the vector **E**, which in the absence of dissipation is a source of electric polarization **P**, in the presence of dissipation serves, because of a change in the electron distribution function, as a source of the toroidal order parameter **T**. As a result, we have a diverging photoconductivity at the point of the phase transition to the toroidal state. In the nonlinear response to an electric field, there appears a term in the conductivity σ, which is linear in E (instead of the usual quadratic term). Some other anomalous nonlinear effects have also been considered in Ref. [69].

In Refs [73, 74], it was shown that a sufficiently large magnetoelectric effect can occur in asymmetric semiconductor heterostructures. A review of experimental work in this field is given in Ref. [75].

6. Diamagnetic anomalies in an inhomogeneous toroidal state

As far back as 1952, V L Ginzburg [76] posed the question of the possible existence of states which possess the properties of an ideal diamagnet (superdiamagnetism) similar to superconductors, but having normal resistance.

It should be noted that as long ago as before the creation of the microscopic theory of superconductivity [21] there were attempts to explain the nature of the superconductive state by the introduction of a current **j** as an order parameter. Precession of macroscopic loops with current in a magnetic field could be considered to be responsible for strong diamagnetism; however, this requires a hardness of wave functions, which would exclude the paramagnetic component. As is known, this is precisely the case that takes place in superconductors.

The very existence of a current **j** (including a nonuniform one) as an order parameter contradicts (as was already mentioned in Section 2) the condition of gauge invariance. In Section 5, it was shown that the poloidal configuration of current loops in the unit cell of the toroidal state [see formula (5)] leads to a weakening of the paramagnetic component because of the emergence of electric polarization (see Fig. 4). Simultaneously, an elementary but physically transparent analysis [10] shows that, because of a quadratic dependence of the toroidal moment [see formula (5)] on **r** under the summation sign instead of a linear dependence on **r** in the expression for the magnetic moment, a twofold increase is observed in the diamagnetic component.

6.1 Suppression of the paramagnetic component in the response to a magnetic field

For a loop with a current density I and an area S, the magnetization equals $M = AIS$, where A is a dimensionless constant. The paramagnetic component of the response to a magnetic field, $\delta M_{\mathrm{s}} = AI\delta S$, is connected with a change in the projection of the loop onto the direction perpendicular to the magnetic field. The diamagnetic component $\delta M_I = AS\delta I$ is caused by a change in the current flowing in the loop. In the case of the magnetic moment $M \approx \int \mathbf{j}\mathbf{r}\,\mathrm{d}^3 r$ or the inhomogeneous ferromagnetic state, when $\mathbf{I} = \mathrm{rot}\,\mathbf{M}(\mathbf{r})$, the current density can be expressed through the characteristic dimension of the loop: $I \sim M/\rho$.

Then, $\delta M_I \sim -AM\delta\rho$ and $\delta M_{\mathrm{s}} \sim 2AM\delta\rho$ ($\delta\rho$ is the change in the characteristic dimension under the effect of the field). The total variation in magnetization is given by $\delta M = \delta M_I + \delta M_{\mathrm{s}} \sim AM\delta\rho$, i.e., the response of the loop is paramagnetic.

In the case of the toroidal moment [see formula (5)] or inhomogeneous toroidal ordering, when $\mathbf{I} \sim \mathrm{rot}\,\mathrm{rot}\,\mathbf{T}(\mathbf{r})$, the current density **I** is expressed through the characteristic

dimension of the loop as $\mathbf{I} \sim \mathbf{T}/\rho^2$. In this case, we arrive at

$$\delta M_I \sim -\frac{2AT}{\rho}\,\delta\rho\,, \quad \delta M_{\rm s} \sim \frac{2AT}{\rho}\,\delta\rho\,,$$

i.e., $\delta M = \delta M_I + \delta M_{\rm s} \equiv 0$.

Although the above discussion also touched on an inhomogeneous toroidal state, the estimation performed corresponds in fact to the homogeneous state $\mathbf{T}(\mathbf{r}) = \text{const}$, since the characteristic dimension ρ and the magnitude of $\mathbf{T}$ in the above estimates were considered independent of the coordinate. In this case, the anomalous susceptibility near the point of the phase transition to the toroidal state is observed only in the response to the external current, just as to the external field thermodynamically conjugate to the toroidal ordering.

This also refers to $\operatorname{rot}\mathbf{B} = \mathbf{j}$, i.e., an anomalous response exists only in the case of a nonuniform magnetic field.

The effect of a weakening of the paramagnetic component because of the emerging electric polarization, which was neglected in our estimates, must lead to the predominance of the diamagnetic component. A rigorous treatment of this problem on the basis of the balance of forces near the phase transition temperature and on the basis of the excitonic insulator model at a zero temperature (see Section 6.2) confirms this conclusion.

Notice first of all that the macroscopic model with the observance of gauge invariance confirmed that the response of a homogeneous toroidal state to a nonuniform magnetic field exhibits an anomalous behavior of the paramagnetic sign [77].

6.2 Current precession of inhomogeneous toroidal states (condition of the balance of forces)

Let us now turn to a strict study of the problem of the response of an inhomogeneous toroidal state $\mathbf{T}(\mathbf{r})$ with a macroscopic current $\mathbf{I} \sim \operatorname{rot}\operatorname{rot}\mathbf{T}(\mathbf{r})$ to a uniform magnetic field.

The procedure of minimization of the functional of free energy, used in Ref. [77], correctly describes the response of a toroidal state to a nonuniform magnetic field. In fact, it has a static nature and ignores the dynamic effects of the magnetic field on the current loops and current lines as the whole entities. Correspondingly, the precession in the loop and related diamagnetic component δM_I of the response are also neglected in the classical sense. From the formal viewpoint, the absence of reaction to a uniform field is connected with the necessity of integration-by-parts of the gradient terms from $\mathbf{T}(\mathbf{r})$ upon minimization of the functional. In this procedure, the surface contribution is rejected, which in fact contains information about the response of the system to the external uniform field. This follows from the expression

$$\delta F_a \sim -\int \mathrm{d}^3 r\, \mathbf{B}\operatorname{rot}\mathbf{T}$$

for the interaction with the field, which at $\mathbf{B} = \text{const}$ is the total derivative and is reduced to the integral over the surface. The scheme of balance of forces was suggested in Ref. [78]. It makes it possible to locally and explicitly consider boundary conditions. The basic idea of the approach proposed is the description of the interaction of the system with the field in terms of force, rather than the usually employed energy, characteristics. The switch-on of a field leads to the appearance of an external force. The equilibrium value of the order parameter is determined from the condition of the balance of external and internal forces. The latter arise as a result of deformation of the system, i.e., of the appearance of an induced component $\mathbf{T}_{\rm ind}$ in the order parameter. Both the external and internal forces arise only through the derivatives of the order parameter $\mathbf{T}(\mathbf{r})$ with respect to the coordinate.

In the expression for the free energy $F(\mathbf{R})$ in the absence of a field, we restrict ourselves to only first-order derivatives $\nabla\mathbf{T}(\mathbf{R})$:

$$F(\mathbf{R}) \equiv F\big(\mathbf{T}(\mathbf{R}), \nabla\mathbf{T}(\mathbf{R})\big)\,.$$

For describing the reaction of the system to external force, it is necessary to exclude the energy $f_{\rm int}$ of the internal stresses from the free energy. These stresses exist in the absence of external force as well; they are caused by one of the reasons for the spatial inhomogeneity of $\mathbf{T}(\mathbf{R})$:

$$f_{\rm int}(\mathbf{R}) = \nabla F(\mathbf{R})\,. \tag{46}$$

The minimum of $F(\mathbf{R})$ is determined from the Euler equation

$$\frac{\partial F}{\partial \mathbf{T}} - \frac{\mathrm{d}}{\mathrm{d}x}\,\frac{\partial F}{\partial \nabla\mathbf{T}} = 0\,. \tag{47}$$

By determining $F(\mathbf{R})$ from equation (47) and then $f_{\rm int}$ from formula (46), we obtain the equilibrium condition in the absence of an external force:

$$U(\mathbf{R}) \equiv F - \nabla\mathbf{T}\left(\frac{\partial F}{\partial \nabla\mathbf{T}}\right) = \text{const}\,. \tag{48}$$

Equality (48) means that the free-energy density minus the energy of the inhomogeneity is invariant with respect to $\mathbf{R}$.

It is convenient to represent Eqn (48) in the form

$$\nabla U(\mathbf{R}) = 0\,. \tag{49}$$

The allowance for an external force leads to the replacement of expression (49) by the following one:

$$\nabla U(\mathbf{R}) = f_{\rm ext}\,, \tag{50}$$

which is the basic relationship for the scheme of the balance of forces.

For the case we are interested in, Eqn (50) takes the form

$$\frac{\mathrm{d}U}{\mathrm{d}\mathbf{R}} = \frac{e}{m_0}\,\tilde{\gamma}\,[\mathbf{B}\operatorname{rot}\operatorname{rot}\mathbf{T}]\,, \tag{51}$$

where the coefficient $\tilde{\gamma}$ is determined by the fine details of the microscopic model.

The left-hand side of equation (51) involves the force that appears as a result of a displacement of the internal coordinates by $\delta\mathbf{R}$ in the direction of the external force, i.e., in the direction in which the work is done over the body. The right-hand side of equation (51) represents the Lorentz force which supplies no work. The apparent contradiction is resolved by the fact that in the element of volume moving during the displacement of internal coordinates under the action of the Lorentz force an electric field $\mathbf{E} = [\mathbf{V} \times \mathbf{B}]$ (where $V = \partial\mathbf{R}/\partial t$ is the velocity of motion) is induced,

which in time δt delivers work [79]

$$\delta A = \mathbf{E}\,\mathbf{j}\,\delta t = -[\mathbf{j}\times\mathbf{B}]\,\delta\mathbf{R}\,.$$

It is this fact that explains the above-mentioned manifestation of dynamic effects. The system with a current is reconstructed until the Lorentz force is balanced by the internal stresses (excluding the internal stresses that existed in the system because of the inhomogeneity of $\mathbf{T}(\mathbf{r})$ in the absence of the external force). Thus, condition (51) describes the situation that was established as a result of the dynamic effect, i.e., a reconstruction of current loops, which is reduced to the emergence of diamagnetic surface current in the system.

The authors of Ref. [78] considered, on the basis of the above approach, three types of inhomogeneities of $\mathbf{T}(\mathbf{R})$ near the temperature of the phase transition to the toroidal state.

The first case corresponds to the inhomogeneities connected with the antisymmetric boundary conditions along the direction x for the equilibrium parameter: $\mathbf{T}_0(-\infty) = -\mathbf{T}_0(\infty)$.

The solution to equation (47) in this case takes on the form

$$T_0(x) = T_0\tanh\left(\frac{x}{\xi_{\rm tor}\sqrt{2}}\right),\ \xi_{\rm tor}^2 = -\frac{\gamma}{\alpha},\ T_0^2 = -\frac{\alpha}{2\beta}\,. \tag{52}$$

The coefficients α, β, and γ define the expression for the free-energy density without an external field:

$$F(\mathbf{T},\nabla\mathbf{T}) = \alpha|\mathbf{T}|^2 + \beta|\mathbf{T}|^4 + \gamma|\nabla\mathbf{T}|^2\,. \tag{53}$$

A domain wall corresponds to solution (52); the spontaneous current and the Lorentz force that acts on this current are equal to zero at $\pm\infty$ in this case.

The first integral of equation (51) has the form

$$\gamma(\nabla T)^2 - \left(\frac{e}{m_0}\right)\tilde{\gamma}B\nabla T - \beta(T_0^2 - T^2) = 0\,. \tag{54}$$

Hence, we obtain the following implicit expression for $T(x)$:

$$x = 2\gamma\int_0^T \mathrm{d}T\left\{\tilde{\gamma}\,\frac{e}{m_0}\,B - \left[\frac{e^2}{m_0^2}\,\tilde{\gamma}^2B^2 + 4\beta\gamma(T_0^2 - T^2)^2\right]^{1/2}\right\}^{-1/2}.$$

For the contribution $T_1(x)$ which is linear in B, we have $T_1(\pm\infty) = 0$, $T_1(x) = -T_1(-x)$, and $T(0) = 0$.

From expression (54), we can obtain the following asymptotics at zero point and at infinity:

$$T_1(x\to 0) \sim \frac{e\tilde{\gamma}}{2\gamma m_0}\,xB,\ \max T_1(x \gg \xi_{\rm tor}) \sim \frac{e\tilde{\gamma}}{m_0(\gamma\alpha)^{1/2}}\,B\,. \tag{55}$$

Then, we arrive at the following estimate for the susceptibility χ':

$$\chi' = c\left(\frac{m^*}{m_0}\right)^2|\mathbf{p}_{12}|^2\xi_0^2\chi_{\rm L}\,, \tag{56}$$

where c is the concentration of domain walls, $\gamma \sim \xi_0^2$, ξ_0 is the correlation length of the order parameter, $\chi_{\rm L} = -e^2k_{\rm F}/12\pi^2m_0$ is the Landau diamagnetic susceptibility, and $k_{\rm F}$ is the Fermi momentum.

If we introduce a characteristic scale $\lambda_{\rm tor} = \xi_0|\mathbf{p}_{12}|/k_{\rm F}$ of current correlations, then for χ' we obtain the following formula

$$\chi' = \chi_{\rm L}\left(\frac{\lambda_{\rm tor}}{r_{\rm a}}\right)^2, \tag{57}$$

where $r_{\rm a}$ is on the order of interatomic distance.

According to the well-known Langevin formula, the diamagnetic susceptibility of an individual electron is proportional to the area bounded by its orbit; however, the number of orbits per unit area is, in turn, inversely proportional to the area of orbits. Therefore, a large radius of an orbit cannot ensure a high value of susceptibility per unit volume. In the model under consideration ($\xi_0 \gg r_{\rm a}$), the situation is analogous to the BCS theory [21], where there is an embedding of a large number of electron–hole pairs into each other.

At the same time, the mutual induction effect also suppresses an increase in the diamagnetism in the case of a mechanical enclosure of one orbit into another. An anomalously large diamagnetism for $\lambda_{\rm tor} \gg r_{\rm a}$, obtained for the inhomogeneous toroidal state, corresponds to such an embedding of orbits of radius $\lambda_{\rm tor}$ at which the electron–electron correlations suppress the mutual induction effect.

As the second example in Ref. [78], a case was examined where the inhomogeneous state of $\mathbf{T}(\mathbf{r})$ is connected with the change in the sign of the coefficient γ of the gradient term, i.e., near the Lifshitz point, rather than with the boundary conditions. In the model of an excitonic insulator, this situation is realized when the condition of nesting, $\varepsilon_1(p) = -\varepsilon_2(p)$, is violated, for example, because of alloying [80, 81].

The spatial scale of the corresponding inhomogeneity can change over wide limits, beginning from the doubling of the period. Such states are in fact antitoroidal: the toroidal moment periodically reverses sign in them. The antitoroidal states are just the examples of systems with anomalously strong diamagnetism.

From the viewpoint of magnetoelectric properties, the antitoroidal states are of no interest.

The expression for the susceptibility in this case coincides with formula (57) to an accuracy of a numerical coefficient on the order of unity, and $c \equiv 1$. Special attention should be given to the case with so-called embedded inhomogeneity, when the scale of the inhomogeneity is specified rather than found self-consistently, unlike both cases above.

In the case of an embedded inhomogeneity, we have $\lambda_{\rm tor} \sim \xi(T)$, where

$$\xi(T) = \xi_0\left(\frac{T_{\rm c}}{T_{\rm c} - T}\right)^{1/2} \tag{58}$$

and the current correlation length $\lambda_{\rm tor}$ diverges together with $\xi(T)$ as T approaches the critical temperature $T_{\rm c}$, i.e., the system becomes an ideal diamagnet near the phase transition temperature.

There are grounds for hoping that it is precisely this regime that can be realized in a periodically repeating semiconductor heterostructure [70, 74, 75], i.e., such a system will possess an antitoroidal ordering.

In $LiCoPO_4$ crystals with toroidal ordering, a domain structure is observed [82]. However, the proportion of

domain walls that draw a spontaneous current and therefore could be responsible for the anomalous diamagnetism is sufficiently small for them to manifest themselves in the total susceptibility.

Thus, the inhomogeneous toroidal, in particular, antitoroidal, state is an example of the realization of V L Ginzburg's ideas of a situation where the anomalously large diamagnetism (superdiamagnetism) has a collective but not superconductive nature. The above-given phenomenological description of the diamagnetic susceptibility near T_c is correct in the case of any of the above mechanisms of formation of the toroidal state.

In Ref. [78], the problem of the magnetic susceptibility in a constant magnetic field at a zero temperature in the model of an excitonic insulator has also been solved.

The nontriviality of the problem lies in the fact that to strictly control the fulfillment of the condition of gauge invariance, which guarantees the reliability of the result obtained, the electron spectrum for the inhomogeneous toroidal state should be known.

The results obtained for χ' at a zero temperature agree qualitatively with those obtained above for $T \leqslant T_c$ [83].

6.3 De Haas–van Alphen oscillations in an effective internal magnetic field — a diamagnetic giant differential susceptibility in a weak external magnetic field

In Ref. [84], another reason was analyzed for the anomalously large diamagnetism caused by the manifestation in the inhomogeneous toroidal state of the effective magnetic (pseudomagnetic) field B_{eff} determined by the first-order derivatives of $\mathbf{T}(\mathbf{r})$ instead of the cases of current precession (examined in Section 6.2), which are determined by the second derivatives.

A quasi-two-dimensional two-band model with an anisotropic interband hybridization **Pk** was presented. In the dispersion law for each of the bands $i = 1, 2$, $\varepsilon_i(\mathbf{k}) = \varepsilon_i(k_x k_z) + \varepsilon_i(k_y)$, the dispersion in k_y is disregarded in comparison with the dispersion of hybridization, which is nonzero only in the direction k_y: $|\varepsilon_i(k_y)| \ll |Pk_y|/m_0$.

In this case, the effective reduced Hamiltonian for the singlet imaginary parameter $\Delta(r)$ has the form

$$\hat{H}_{\text{red}} = \begin{pmatrix} \varepsilon_1(\hat{k}_x \hat{k}_z) & \mathrm{i}\left(\dfrac{1}{m_0} P\hat{k}_y + \Delta(\mathbf{r})\right) \\ -\mathrm{i}\left(\dfrac{1}{m_0} P\hat{k}_y + \Delta(\mathbf{r})\right) & \varepsilon_2(\hat{k}_x, \hat{k}_z) \end{pmatrix}. \quad (59)$$

The parameter $\Delta(\mathbf{r})$ enters into $\hat{H}_{\text{red}}$ quite analogously to the case of the vector potential directed along the y-axis.

After the transformation

$$\Delta(\mathbf{r}) = \frac{Pe}{m_0 c} A_{\text{eff}}, \quad \mathbf{A}_{\text{eff}} = (0, A_{\text{eff}}, 0),$$

the Hamiltonian $\hat{H}_{\text{red}}$ coincides with the Hamiltonian of the two-band model in the pseudomagnetic field B_{eff}:

$$\mathbf{B}_{\text{eff}} = \operatorname{rot} \mathbf{A}_{\text{eff}} = \left(-\frac{cm_0}{eP} \nabla_z \Delta(\mathbf{r}),\, 0,\, \frac{cm_0}{eP} \nabla_x \Delta(\mathbf{r})\right). \quad (60)$$

In neglecting the dependence of $B_{\text{eff}}(\mathbf{r})$ on $\mathbf{r}$ (which corresponds to the neglect of spontaneous currents $\mathbf{j} \sim \operatorname{rot}\operatorname{rot} \mathbf{P}\Delta(\mathbf{r})$ which determine the diamagnetic susceptibility (56)), one can use the methods of calculating macroscopic characteristics of a homogeneous system in a uniform external magnetic field $\mathbf{B}$. The true magnetic field $\mathbf{B} = \operatorname{rot} \mathbf{A}$ is introduced into Hamiltonian (59) through the replacement $\mathbf{A}_{\text{eff}} \to \mathbf{A}_{\text{eff}} + \mathbf{A}$.

The magnetic susceptibility is determined by the combined action of the true and pseudomagnetic field: $\mathbf{B}_\Sigma = \mathbf{B}_{\text{eff}} + \mathbf{B}$. The magnitude of the pseudomagnetic field on the scale of the true magnetic field can be sufficiently large.

Then, the magnetic susceptibility of the system with an inhomogeneous $\Delta(\mathbf{r})$ in a weak magnetic field coincides with the differential susceptibility of a system with $\Delta = 0$ placed in a strong field B_{eff}. In the case of a low temperature T, when $k_B T \ll \mu_B B_{\text{eff}}$ (k_B is the Boltzmann constant, and μ_B is the Bohr magneton), the system is under the conditions corresponding to the de Haas–van Alphen effect. In this situation, the differential susceptibility (equal to the sought-for susceptibility χ'), as is known [85], can take arbitrarily large values in modulus, both positive (paramagnetism) and negative (diamagnetism). True, in contrast to the standard situation, the field B_{eff}, generally speaking, is nonuniform (in fact, it is precisely its nonuniformity that corresponds to the existence of a current and its precession). This resembles the situation with the de Haas–van Alphen effect in the presence of Shoenberg diamagnetic domains. However, there can exist a situation where the nonuniformity of B_{eff} can be neglected [86]. For this to be possible, the scale of the nonuniformities (in our case, this is the scale of changes in $\Delta(\mathbf{r})$), three examples of which were considered in Section 6.2, must be large in comparison with the effective magnetic length of the pseudomagnetic field:

$$\lambda = \frac{ck_F}{eB_{\text{eff}}}.$$

Then, if the condition

$$\frac{\mu_B B_{\text{eff}}}{E_F} \ll 1$$

is fulfilled, the oscillating (with changing B_{eff}) part of the susceptibility proves to be much greater than the smooth part, and the response of the susceptibility χ' (which is of interest for us) to the external field B will be equal to the oscillating component:

$$\chi' = a \cos \frac{E_F}{\mu_B B_{\text{eff}}}, \quad a = \left(\frac{e}{c}\right)^{1/2} \frac{E_F^2 m_0}{(2\pi^5 B_{\text{eff}})^{1/2}} \frac{\varkappa}{\sin \varkappa}, \quad (61)$$

where

$$\varkappa = \frac{2\pi^2 T}{\mu_B B_{\text{eff}}}.$$

It follows from expression (61) that for some values of B_{eff} corresponding to the positive half-period of the cosine function, the susceptibility is positive (paramagnetic), while for other values of B_{eff} which correspond to the negative half-period, it is negative (diamagnetic). In the limit $a \to \infty$, the susceptibility χ' approaches the susceptibility of an ideal diamagnet: $\chi' \to -\infty$ ($\chi \to 1/4\pi$).

In contrast to the problem concerning the system placed in a real strong magnetic field, in the case under consideration the magnitude of B_{eff} depends on the parameters of the system, which determine the sign of the half-period and,

therefore, the sign of the susceptibility. In principle, there can exist a situation where a change in the temperature will change the sign of χ' as a result of a change in $\Delta(\mathbf{r})$.

The conditions of the realization of anomalous diamagnetism due to the pseudomagnetic field B_{eff} [first spatial derivatives of $\Delta(\mathbf{r})$ in expression (60)] appear to be more hardly fulfilled in comparison with the manifestation of the current precession [second spatial derivatives of $\Delta(\mathbf{r})$ in expression (10)].

6.4 Co-existence of the toroidal and superconducting states

An original manifestation of a toroidal state takes place in the case of its co-existence with superconductivity [84]. This co-existence can be strictly described within the framework of the same model of an excitonic insulator; this was first performed for a real singlet electron–hole parameter $\Delta^{\text{s}}_{\text{Re}}$ in Refs [87, 88]. The co-existence of superconductivity with an insulating phase with spontaneous currents was studied in connection with the superconductivity of cuprates [45–47, 55, 56, 61].

In Ref. [84], a number of specific features were revealed in the interaction of the superconductive and toroidal states that have not been previously studied.

In superconductors, the ordinary condition of gauge invariance is known to be violated. The role of the gauge-invariant quantity is played by the combination $\nabla\varphi - (2e/c)\,\mathbf{A}$ (where φ is the phase of the superconducting order parameter Δ_{c}), which can explicitly be present in macroscopic relationships.

For the superconducting current $\mathbf{j}_{\text{c}}$, the following expression is obtained in this case:

$$\mathbf{j}_{\text{c}} = \eta\left(\nabla\varphi - \frac{2e}{c}\,\mathbf{A} - e\xi\,\mathbf{T}\right), \tag{62}$$

where the quantity $\eta \sim |\Delta_{\text{c}}|^2$ is proportional to the density of the superconducting electrons, and $\xi \sim 1/v_{\text{F}}$.

Therefore, the following term appears in the expression for the free energy:

$$\delta F \sim \text{i}\mathbf{T}\left(\Delta_{\text{c}}\left(\nabla - \text{i}\,\frac{2e}{c}\,\mathbf{A}\right)\Delta_{\text{c}}^{*} - \text{c.c.}\right). \tag{63}$$

Calculation in the model of an excitonic insulator with $P \neq 0$ and $\Delta^{\text{s}}_{\text{Im}} \neq 0$ results in the following additional term in the expression for the current:

$$\mathbf{j} = \frac{7}{8}\,\xi(3)\,\frac{ek_{\text{F}}|\Delta_{\text{c}}|^2}{\pi^4 T^2}\,\mathbf{P}\Delta^{\text{S}}_{\text{Im}}, \tag{64}$$

where $\xi(3)$ is the Riemann zeta function.

From expression (62), we derive the following equation for the phase:

$$\nabla^2\varphi(\mathbf{r}) = e\xi\,\text{div}\,\mathbf{T}. \tag{65}$$

Then, an additional phase factor appears in Δ_{c}:

$$\Delta_{\text{c}}(\mathbf{r}) = |\Delta_{\text{c}}(\mathbf{r})|\exp\left\{\text{i}\xi e\int_0^r \mathbf{T}_{||}(\mathbf{r}')\,\text{d}\mathbf{r}'\right\}, \tag{66}$$

where $\mathbf{T}_{||}(\mathbf{r})$ is the vortex-free component of the toroidal order parameter ($\text{rot}\,\mathbf{T}_{||}(\mathbf{r}) \equiv 0$); at $\mathbf{T}(\mathbf{r}) = \text{const}$, one should assume that $\mathbf{T}_{||} = \mathbf{T}$.

In the open-loop system, this phase completely compensates for the additional term in the expression for the current, without changing the parameters of the Meissner effect.

For the closed loop, which consists of a usual superconductor and a superconductor with a coexisting toroidal ordering, the following expression is obtained for the magnetic flux through the loop:

$$\Phi = \Phi_0\left\{n - \frac{e\xi}{2\pi}\int \mathbf{T}_{||}(\mathbf{r})\,\text{d}\mathbf{r}\right\}, \tag{67}$$

where n is an integer.

If the second term in the curly brackets is not equal to n, then, because of $\Phi \neq 0$, a spontaneous current $I = cL^{-1}\Phi$ arises in the loop, where L is the loop inductance.

Of great interest is the manifestation of the vortex component of the toroidal moment $\mathbf{T}$ with $\text{rot}\,\mathbf{T} \neq 0$ in the superconducting state.

If we apply a rot operation to expression (62) for the current, we obtain a modified London equation

$$\text{rot}\,\Lambda^2\mathbf{j}_{\text{c}} = -c^{-1}\mathbf{B} - \frac{1}{2}\,\xi\,\text{rot}\,\mathbf{T}, \tag{68}$$

where $\Lambda^2 = 2e\eta$, and $\chi_{\text{L}} = (\Lambda c)/(4\pi)^{1/2}$.

The Maxwell equation takes the form

$$\text{rot}\,\mathbf{B} = \frac{4\pi}{c}\,\mathbf{j}_{\text{c}} + 4\pi\,\text{rot}\,\text{rot}\,\mathbf{T}. \tag{69}$$

Then, from Eqns (68) and (69) we arrive at

$$-\nabla^2\mathbf{j}_{\text{c}} - \frac{\mathbf{j}_{\text{c}}}{\lambda_{\text{L}}^2} = \frac{c}{\lambda_{\text{L}}^2}\,\text{rot}\,\text{rot}\,\mathbf{T} + \frac{c^2\xi}{\lambda_{\text{L}}^2}\,\text{rot}\,\text{rot}\,\mathbf{T}. \tag{70}$$

The second term on the right-hand side of equation (70) is due to the direct interaction of the superconducting and toroidal order parameters; since $c\xi \sim c/v_{\text{F}} \gg 1$, its contribution substantially exceeds the first term, which is an analog of the term with $\text{rot}\,\mathbf{M}$ in the case of the co-existence of superconductivity and ferromagnetism [89].

If the temperatures of the superconducting (T_{c}) and toroidal (T_{tor}) phase transitions are close, the linearized equation for Δ_{c} takes on the form

$$\frac{1}{2m^*}\left(-\text{i}\nabla - \frac{2e}{c}\,\mathbf{A}(\mathbf{r}) - e\xi\,\mathbf{T}(\mathbf{r})\right)^2\Delta_{\text{c}} = -\alpha\Delta_{\text{c}}, \tag{71}$$

where $\alpha \sim (T - T_{\text{c}})$.

This leads to a specific structure of the penetration of the magnetic field into the superconductor, and to a nonmonotonic temperature dependence of the second critical magnetic field H_{c2} [84].

7. Collective oscillations and optical properties of toroidal states

To describe the interaction of toroidal states with an electromagnetic field, it is necessary to investigate the collective modes of toroidal oscillations, which was performed at the phenomenological level in Refs [90, 91]. The interaction with the magnetic component of the electromagnetic wave being proportional to the term $\mathbf{T}\,\text{rot}\,\mathbf{B}$ in the expression for the free energy, then the interaction with the electric component $\mathbf{E}$ is proportional to $\mathbf{T}\,\text{d}\mathbf{E}/\text{d}t$. This corresponds to the emergence of electric polarization $\mathbf{P}$ proportional to $\text{d}\mathbf{T}/\text{d}t$. As a result, a resonance appears in the expression for the dielectric polarization $\varepsilon(\omega)$, which corresponds to one of the modes of toroidal oscillations. In

Refs [90, 91], we predicted a characteristic feature in the temperature dependence of the absorption coefficient, which is observed in the $TlGaSe_2$ compound [92].

To the extent of interaction $\mathbf{T}\,\mathrm{rot}\,\mathbf{B}$, the toroidal oscillations also manifest themselves in the frequency dependence of the magnetic permeability $\mu(\omega)$. The authors of Ref. [93] have studied the effect of toroidal oscillations on the angle of rotation of the plane of polarization of an electromagnetic wave (see also Ref. [94]). Similar problems were also studied in the above-mentioned work [57] on the basis of the Hubbard model.

Since $\varepsilon(\omega)$ and $\mu(\omega)$ can have resonances that are close or even coinciding in frequency, this circumstance, besides the above-noted possibility of exhibiting a high value of the magnetoelectric coefficient α, can lead to the existence of a region of frequencies with a negative refractive index $n(\omega)$. Thus, the toroidal states are of interest from the viewpoint of the realization of metamaterials [95].

In the case of an inhomogeneous toroidal state ($\mathrm{rot}\,\mathbf{T}(\mathbf{r}) \neq 0$), in the equation that describes the trajectory of a light beam in the framework of geometric optics, there appears a term proportional to $\mathrm{rot}\,\mathbf{T}(\mathbf{r})$, which leads to the Hall effect for the light: the direction of the beam deflection changes with a change in the beam direction [96].

Detailed information concerning substances with a toroidal ordering and the experimental methods of their study can be found in the reviews [17, 64]. Special attention should be given to Ref. [97] in which toroidal ordering was observed for the first time.

8. Conclusion

Thus, the anomalous diamagnetic component in the response to a uniform magnetic field exists only in the region of the inhomogeneity of the toroidal state, i.e., in the region of domain walls. Although the latter (as was indicated above) have been observed in the toroidal state [82], the domain walls occupy only a small fraction of the sample volume and, therefore, their contribution to the total susceptibility is negligible. As to the antitoroidal state, in which large diamagnetism must occur, it has not yet been revealed.

To date, we have proposed (and have started studying) heterostructures analogous to those in which a toroidal state with a large magnetoelectric effect was observed [74, 75], but with a periodically repeating structure. In these structures, the fraction of domain walls proves to be on the order of unity.

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References

1. Wigner E P *Nachr. Ges. Wiss. Göttingen Math.-Phys. Kl.* 546 (1932)
2. Zel'dovich Ya B *Zh. Eksp. Teor. Fiz.* **33** 1531 (1957) [*Sov. Phys. JETP* **6** 1184 (1958)]
3. Ascher E *Helv. Phys. Acta* **39** 40 (1966)
4. Ascher E *Int. J. Magn.* **5** 287 (1974)
5. Landau L D *Phys. Z. Sowjetunion* **4** 43 (1933)
6. Heisenberg W *Z. Naturforsch. A* **2** 185 (1947)
7. Heisenberg W *Ann. Physik* **3** 289 (1948)
8. Koppe H *Ergebnisse exakten Naturwissensch.* **23** 283 (1950)
9. Kittel *Quantum Theory of Solids* (New York: Wiley, 1963) [Translated into Russian (Moscow: Nauka, 1967)]
10. Ginzburg V L et al. *Solid State Commun.* **50** 339 (1984)
11. Dubovik V M, Cheshkov A A, Preprint R2-5283, R2-5284 (Dubna: JINR, 1970)
12. Dubovik V M, Tugushev V V *Phys. Rep.* **187** 145 (1990)
13. Apenko S M, Lozovik Yu E *J. Phys. B* **15** L57 (1982)
14. Landau L D, Lifshitz E M *Statisticheskaya Fizika* Vol. 1 (Statistical Physics) (Moscow: Nauka, 1976) [Translated into English (Oxford: Pergamon Press, 1980)]
15. Andreev A F, Grishchuk I A *Zh. Eksp. Teor. Fiz.* **87** 467 (1984) [*Sov. Phys. JETP* **60** 267 (1984)]
16. Gorbatsevich A A, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **39** 558 (1984) [*JETP Lett.* **39** 684 (1984)]
17. Spaldin N A, Fiebig M, Mostovoy M *J. Phys.: Condens. Matter* **20** 434203 (2008)
18. Landau L D, Lifshitz E M *Teoriya Polya* (The Classical Theory of Fields) (Moscow: Nauka, 1976) [Translated into English (Oxford: Pergamon Press, 1983)]
19. Halperin B I, Rice T M *Solid State Phys.* **21** 115 (1968)
20. Keldysh L V, Kopaev Yu V *Fiz. Tverd. Tela* **6** 2791 (1964) [*Sov. Phys. Solid State* **6** 2219 (1965)]
21. Bardeen J, Cooper L N, Schrieffer J R *Phys. Rev.* **108** 1175 (1957)
22. Bogolyubov N N, Tolmachev V V, Shirkov D V *Novyi Metod v Teorii Sverkhprovodimosti* (A New Method in the Theory of Superconductivity) (Moscow: Izd. AN SSSR, 1958) [Translated into English (New York: Consultants Bureau, 1959)]
23. Guseinov R R, Keldysh L V *Zh. Eksp. Teor. Fiz.* **63** 2255 (1972) [*Sov. Phys. JETP* **36** 1193 (1973)]
24. Volkov B A, Gorbatsevich A A, Kopaev Yu V *Usp. Fiz. Nauk* **143** 331 (1984) [*Sov. Phys. Usp.* **27** 464 (1984)]
25. Volkov B A, Kopaev Yu V, Nunuparov M S *Fiz. Tverd. Tela* **21** 2733 (1979) [*Sov. Phys. Solid State* **21** 1571 (1979)]
26. Kopaev Yu V, Nunuparov M S *Fiz. Tverd. Tela* **22** 3599 (1980)
27. Gorbatsevich A A, Kopaev Yu V, Prokopov V I *Fiz. Tverd. Tela* **26** 1155 (1986)
28. Volkov B A et al. *Zh. Eksp. Teor. Fiz.* **81** 729 (1981) [*Sov. Phys. JETP* **54** 391 (1981)]
29. Gorbatsevich A A, Tugushev V V *Zh. Eksp. Teor. Fiz.* **77** 2104 (1979) [*Sov. Phys. JETP* **50** 1006 (1979)]
30. Gorbatsevich A A, Tugushev V V *Zh. Eksp. Teor. Fiz.* **78** 1945 (1980) [*Sov. Phys. JETP* **51** 977 (1980)]
31. Artamonov Yu A, Gorbatsevich A A, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **40** 290 (1984) [*JETP Lett.* **40** 1076 (1984)]
32. Volkov B A, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **27** 10 (1978) [*JETP Lett.* **27** 7 (1978)]
33. Batyev E G *Pis'ma Zh. Eksp. Teor. Fiz.* **29** 381 (1979) [*JETP Lett.* **29** 345 (1979)]
34. Fenton E W *Solid State Commun.* **38** 457 (1981)
35. Nozières P, Saint-James D *J. Physique Lett.* **41** L197 (1980)
36. Volkov B A et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **30** 317 (1979) [*JETP Lett.* **30** 293 (1979)]
37. Volkov B A, Kantser V G, Kopaev Yu V *Zh. Eksp. Teor. Fiz.* **76** 1856 (1979) [*Sov. Phys. JETP* **49** 943 (1979)]
38. Gorbatsevich A A, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **39** 558 (1984) [*JETP Lett.* **39** 684 (1984)]
39. Elesin V F, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **24** 78 (1976) [*JETP Lett.* **24** 66 (1976)]
40. Volkov B A, Kopaev Yu V, Rusinov A I *Zh. Eksp. Teor. Fiz.* **68** 1899 (1975) [*Sov. Phys. JETP* **41** 952 (1975)]
41. Cercellier H et al. *Phys. Rev. Lett.* **99** 146403 (2007)
42. Young D P et al. *Nature* **397** 412 (1999)
43. Balents L, Varma C M *Phys. Rev. Lett.* **84** 1264 (2000)
44. Barzykin V, Gor'kov L P *Phys. Rev. Lett.* **84** 2207 (2000)
45. Chakravarty S et al. *Phys. Rev. B* **63** 094503 (2001)
46. Varma C M *Phys. Rev. B* **55** 14554 (1997)
47. Varma C M *Phys. Rev. Lett.* **83** 3538 (1999)
48. Kaminski A et al. *Nature* **416** 610 (2002)
49. Fauque B et al. *Phys. Rev. Lett.* **96** 197001 (2006); cond-mat/0509210
50. Krotov S S, Tugushev V V *Zh. Eksp. Teor. Fiz.* **91** 1708 (1986) [*Sov. Phys. JETP* **64** 1010 (1986)]

51. Krotov S S, Tugushev V V *Dokl. Akad. Nauk SSSR* **295** 852 (1987) [*Sov. Phys. Dokl.* **32** 658 (1987)]
52. Kopayev Yu V, Krotov S S, Tugushev V V *Solid State Commun.* **61** 737 (1987)
53. Klebanoff L E et al. *J. Mag. Magn. Mater.* **54–57** 728 (1986)
54. Mills D "Surface spin waves on magnetic crystals", in *Surface Excitations* (Modern Problems in Condensed Matter Sciences, Vol. 9, Eds V M Agranovich, R London) (Amsterdam: North-Holland, 1984) p. 379
55. Affleck I, Marston J B *Phys. Rev. B* **37** 3774 (1988)
56. Lee P A, Nagaosa N, Wen X-G *Rev. Mod. Phys.* **78** 17 (2006)
57. Bulaevskii L N et al. *Phys. Rev. B* **78** 024402 (2008)
58. Katsura H, Nagaosa N, Balatsky A *Phys. Rev. Lett.* **95** 057205 (2005); cond-mat/0412319
59. Pomeranchuk I Ya *Zh. Eksp. Teor. Fiz.* **35** 524 (1958) [*Sov. Phys. JETP* **8** 361 (1959)]
60. Varma C M, cond-mat/0311145
61. Belyavskii V I, Kopaev Yu V *Usp. Fiz. Nauk* **176** 457 (2006) [*Phys. Usp.* **49** 441 (2006)]
62. Wu C, Zhang S-C *Phys. Rev. Lett.* **93** 36403 (2004); cond-mat/0312632
63. Hirsch J E *Phys. Rev. B* **41** 6820 (1990)
64. Fiebig M *J. Phys. D* **38** R123 (2005)
65. Debye P *Z. Phys.* **36** 300 (1926)
66. Curie P *J. Physique 3* 393 (1894)
67. Dzyaloshinskii I E *Zh. Eksp. Teor. Fiz.* **37** 881 (1959) [*Sov. Phys. JETP* **10** 628 (1960)]
68. Astrov D N *Zh. Eksp. Teor. Fiz.* **38** 984 (1960) [*Sov. Phys. JETP* **11** 708 (1960)]
69. Gorbatsevich A A, Kopaev Yu V, Tugushev V V *Zh. Eksp. Teor. Fiz.* **85** 1107 (1983) [*Sov. Phys. JETP* **58** 643 (1983)]
70. Gorbatsevich A A, Kopaev Yu V *Ferroelectrics* **161** 321 (1994)
71. Sannikov D G *Ferroelectrics* **219** 177 (1998)
72. Ascher E, in *Magnetoelectric Interaction Phenomena in Crystals*: *Symp., Seattle, 1973* (Eds A J Freeman, H Schmid) (London: Gordon and Breach Sci. Publ., 1975)
73. Gorbatsevich A A, Kapaev V V, Kopaev Yu V *Ferroelectrics* **161** 303 (1994)
74. Gorbatsevich A A, Kapaev V V, Kopaev Yu V *Pis'ma Zh. Eksp. Teor. Fiz.* **57** 565 (1993) [*JETP Lett.* **57** 580 (1993)]
75. Gorbatsevich A A, Omel'yanovskii O E, Tsebro V I *Usp. Fiz. Nauk* **179** 887 (2009) [*Phys. Usp.* **52** 835 (2009)]
76. Ginzburg V L *Usp. Fiz. Nauk* **48** 25 (1952)
77. Volkov B A et al. *Zh. Eksp. Teor. Fiz.* **81** 1904 (1981) [*Sov. Phys. JETP* **54** 1008 (1981)]
78. Volkov B A, Gorbatsevich A A, Kopaev Yu V *Zh. Eksp. Teor. Fiz.* **86** 1870 (1984) [*Sov. Phys. JETP* **59** 1087 (1984)]
79. Landau L D, Lifshitz E M *Elektrodinamika Sploshnykh Sred* (Electrodynamics of Continuous Media) (Moscow: Nauka, 1982) p. 184 [Translated into English (Oxford: Pergamon Press, 1984)]
80. Kopaev Yu V *Fiz. Tverd. Tela* **12** 3 (1970) [*Sov. Phys. Solid State* **12** 1 (1970)]
81. Rice T M *Phys. Rev. B* **2** 3619 (1970)
82. Van Aken B B et al. *Nature* **449** 702 (2007)
83. Gorbatsevich A A, Kopaev Yu V, in *Superconductivity, Superdiamagnetism, Superfluidity* (Ed. V L Ginzburg) (Moscow: Mir Publ., 1987)
84. Gorbatsevich A A *Zh. Eksp. Teor. Fiz.* **95** 1467 (1989) [*Sov. Phys. JETP* **68** 847 (1989)]
85. Shoenberg D *Magnetic Oscillations in Metals* (Cambridge: Cambridge Univ. Press, 1984) [Translated into Russian (Moscow: Mir, 1986)]
86. Privorotskii I A *Zh. Eksp. Teor. Fiz.* **52** 1755 (1967) [*Sov. Phys. JETP* **25** 1167 (1967)]
87. Rusinov A I, Kat D Ch, Kopaev Yu V *Zh. Eksp. Teor. Fiz.* **65** 1984 (1973) [*Sov. Phys. JETP* **38** 991 (1974)]
88. Kopaev Yu V, in *Problema Vysokotemperaturnoi Sverkhprovodimosti* (High-Temperature Superconductivity) (Eds V L Ginzburg, D A Kirzhnits) (Moscow: Nauka, 1977) Ch. 5 [Translated into English (New York: Consultants Bureau, 1982) Ch. 5]
89. Ginzburg V L *Zh. Eksp. Teor. Fiz.* **31** 202 (1956) [*Sov. Phys. JETP* **4** 153 (1957)]
90. Kopaev Yu V, Tugushev V V *Pis'ma Zh. Eksp. Teor. Fiz.* **41** 320 (1985) [*JETP Lett.* **41** 392 (1985)]
91. Kopaev Yu V, Tugushev V V *Zh. Eksp. Teor. Fiz.* **88** 2244 (1985) [*Sov. Phys. JETP* **61** 1327 (1985)]
92. Volkov B A et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **39** 293 (1984) [*JETP Lett.* **39** 351 (1984)]
93. Tugushev V V, Fursa T N *Fiz. Tverd. Tela* **28** 1805 (1986)
94. Zvezdin A K *Kratk. Soobshch. Fiz. FIAN* (6) 35 (2000)
95. Veselago V G *Usp. Fiz. Nauk* **92** 517 (1967) [*Sov. Phys. Usp.* **10** 509 (1968)]
96. Sawada K, Nagaosa N *Phys. Rev. Lett.* **95** 237402 (2005)
97. Popov Yu F et al. *Zh. Eksp. Teor. Fiz.* **114** 263 (1998) [*JETP* **87** 146 (1998)]