Toroidal oscillations in crystals

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Systems with low frequency toroidal-moment density oscillations are investigated in the vicinity of the phase transition point. Dynamical equations describing the toroidal oscillations and their interaction with the photons and phonons are derived within the framework of a model Lagrangian. The behavior of the permittivity and permeability is investigated, and the singularities of the law of dispersion of the toroidal oscillations in noncentrosymmetric crystals are considered. The possibility of experimental observation of the predicted effects is discussed.

§1. INTRODUCTION

The investigation of a new type of long-range order in crystals that is characterized by the appearance of a macroscopic toroidal moment ("toroidal order") has lead to the discovery of new and quite unusual properties of these systems.[1–4] From the purely phenomenological standpoint, to describe the type of order in question, we introduce a vectorial order parameter with transformation properties similar to those of the velocity vector **v** (or, which is the same, of the electric current **j**). The crystal transformation group under consideration is classified within the framework of the magnetic symmetry groups, and belongs to one of the 31 magnetoelectric classes that admit of the existence of an antisymmetric component of the magnetoelectric tensor.[5,6] It is natural that the introduction of an order parameter with the indicated transformation properties does not by itself reveal the physical nature of the phenomena occurring in the system. The main achievement of the theory[1–4] consists in the fact that it has, for the first time, been possible to analyze within the framework of a quantum-mechanical model the genesis of a new collective electron state in a solid, and establish the characteristics of the behavior of this state in external fields. It has also been possible to establish a definite analogy between it and the classical system of distributed electric-charge fluxes, which admits, within the framework of a formal multipole expansion, of a description in terms of the classical toroidal moments.[7] The term "toroidal state" itself arose precisely on account of this analogy.

It is natural to relate the establishment of the toroidal long-range order with the softening of some collective model of the electronic oscillations. In the microscopic model[1,2] such a mode is the transverse excitonic oscillation mode that, for a definite relationship $\Delta\varphi$ between the phases of the wave functions of the electron and hole in the electron-hole pair ($\Delta\varphi = \pi/2$), characterizes the oscillations of the toroidal-moment density $\mathbf{T}(\mathbf{r},t)$. For a different phase relation ($\Delta\varphi = 0$) the corresponding excitonic mode characterizes in this model the oscillations of the electron-polarization density. The softening of a specific excitonic mode implies the occurrence of a phase transition into a state with a spontaneous toroidal moment or with spontaneous polarization.

It is clear that, in such a formulation of the problem, it is reasonable to describe the toroidal oscillations in the unreconstructed phase as collective excitations in a background of the "normal" ground state. In the present paper we investigate systems with low-frequency toroidal oscillations in the vicinity of the toroidal instability threshold. We assume that the magnetic-symmetry group of the unreconstructed (high-temperature) phase has as a subgroup one of the above-indicated magnetoelectric groups, and that the order parameter **T** transforms according to the corresponding irreducible vector representation. Furthermore, we consider only those systems in which the toroidal ordering can occur as a second-order transition (i.e., we assume that the symmetry of the high-temperature phase does not admit of invariants of the Lifshitz type[8]). These assumptions allow us to use in the analyze of the low-amplitude low-frequency toroidal oscillations the effective-Lagrangian method, in which a power series expansion in the order parameter **T** and its derivatives is carried out. It is shown that these oscillations interact in an unusual manner with light, and, thus, can manifest themselves in the optical properties of crystals that are prone to toroidal instability.

The paper is organized as follows:

In §2 we introduce a Lagrangian for the description of the toroidal oscillations in crystals with cubic symmetry, investigate the spectrum of the longitudinal and transverse oscillations without allowance for the retardation of the interaction with the electromagnetic field, and find the frequency dependence of the dynamical permittivity $\varepsilon(\omega)$. We also show here that, as the frequency of the toroidal soft mode tends to zero, the static permittivity $\varepsilon(0)$ does not pass through any singularities.

In §3 we analyze the effect of a homogeneous magnetic field (homogeneous magnetization in the case of a ferromagnetic crystal) on the dynamical permittivity $\varepsilon(\omega)$. We find the natural frequencies of the toroidal oscillations in a magnetic field. Of importance is the result that the transverse static-permittivity components $\varepsilon_{xx}(0)$ and $\varepsilon_{yy}(0)$ diverge at the toroidal transition point (the z axis is oriented along the direction of the magnetic field), below which the system exhibits spontaneous polarization in the xy plane.

In §4 we investigate the singularities of the dispersion law for the toroidal oscillations in noncentrosymmetric crystals, and demonstrate the possibility of the appearance in such crystals of an incommensurate toroidal structure (and, consequently, of a spontaneous current[1]) below the phase

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transition point. This possibility is due to the existence of a minimum in the toroidal-mode dispersion law $\omega_T(\mathbf{q})$ at some quasimomentum value $\mathbf{q}\neq 0$, which leads to the vanishing at the phase transition point of the soft mode frequency, i.e., to a situation in which $\omega_T \to 0$ at $\mathbf{q} = \mathbf{q}_{min} \neq 0$.

In §5 we study the interaction of the transverse toroidal oscillations with light, and derive an expression for the law of dispersion of the toroidal-photon oscillations (we are in fact talking about new branches in the polariton spectrum).

In §6 we investigate the relationship between the toroidal and lattice vibrations in polar crystals. We pay particular attention to systems that are close to a second-order structural transition point. We find that the toroidal oscillations, by intermixing with the phonons, induce the softening of the frequency of the polar lattice vibrations, but do not cause a shift of the structural transition point in the absence of a magnetic field. If on the other hand an external uniform magnetic field is applied to the system (or if the system had earlier undergone a transition into the ferromagnetic state), then there is a rise in the structural transition temperature.

In §7 we consider the behavior of the dynamical magnetic susceptibility tensor $\chi(\omega)$ in noncentrosymmetric crystals (in particular, in pyroelectric crystals, or in crystals located in an external electric field). One of the components of the static magnetic susceptibility diverges at the toroidal transition point, and a magnetic order occurs below this point.

In conclusion (§8) we state the main results obtained in the investigation, and discuss the possibility of an experimental observation of the toroidal oscillations. In the Appendix we compute the coefficients of the Lagrangian for the two-band semiconductor model.

§2. DYNAMICAL PERMITTIVITY OF SYSTEMS WITH LOW-FREQUENCY TOROIDAL OSCILLATIONS

We shall describe the toroidal oscillations in crystals for the case in which there is no macroscopic toroidal moment in the ground state, and the system has not undergone any type of magnetic or ferroelectric ordering. It is assumed that, in the frequency region under consideration, the toroidal oscillations are well-defined, weakly damped collective excitations occurring above the ground state (in the microscopic semiconductor model considered in the Appendix, these conditions are fulfilled, and the eigenfrequencies of the toroidal oscillations lie within the forbidden band in the single-electron spectrum).

It is most natural to use the Lagrangian formalism, within the framework of which we must write down the "kinetic" and "potential" energies of the oscillations. The kinetic energy in the case of low-amplitude, low-frequency toroidal oscillations can be written in the form

$$K_T=\beta_2(\dot{\mathbf{T}})^2+\beta_4(\ddot{\mathbf{T}})^2, \tag{1}$$

where $\mathbf{T}(\mathbf{r},t)$ is the toroidal-moment density, $\beta_2, \beta_4>0$, and it is under certain conditions absolutely necessary to retain the second term in (1) (see below). For the two-band semiconductor model, the coefficients β_2 and β_4 are computed in the Appendix, where it is found that $\beta_4/\beta_2\sim E_g^{-2}$, E_g being the forbidden-band width. The expansion (1) has meaning at oscillation frequencies $\omega \ll E_g$. Let us introduce the quantity $M_T = (^2\beta_2)^{-1}$, which plays the role of the "mass" of the toroidal oscillation.

The potential energy in zero external electric and magnetic fields can be written in the simplest form (in this section we consider only systems with cubic symmetry):

$$U_{\mathrm{T}}=\alpha\mathbf{T}^2+\gamma[(\operatorname{div}\mathbf{T})^2+(\operatorname{rot}\mathbf{T})^2], \tag{2}$$

where we have retained only the lowest-order coordinate derivatives and the terms that are quadratic in the amplitude. Here and below we assume that $\alpha, \gamma>0$. Varying the Lagrangian of the system in the absence of external fields

$$L_{\mathrm{T}}=K_{\mathrm{T}}-U_{\mathrm{T}} \tag{3}$$

with respect to $\mathbf{T}(\mathbf{r},t)$, we obtain the equation of motion, from which we find for the eigenfrequency of the toroidal oscillations in the case when the second term in (1) is neglected the expression

$$\omega_{0T}^2(\mathbf{q})=2M_T(\alpha+\gamma q^2). \tag{4}$$

A characteristic feature of the interaction of the toroidal moment with external fields is that the source conjugate to the toroidal-moment density is the current,[7] and that in uniform stationary fields the toroidal moment is not induced in the absence of dissipation (in the approximation linear in the electric field $\mathbf{E}$ and the magnetic field $\mathbf{B}$). Let us write the field-related corrections to the Lagrangian of the system in form

$$\Delta L_{\mathrm{E}}=\lambda_{\mathrm{E}}\mathbf{T}\dot{\mathbf{E}}, \tag{5}$$

$$\Delta L_{\mathrm{B}}=(\lambda_{\mathrm{B}}/c)\mathbf{T}\operatorname{rot}\mathbf{B}. \tag{6}$$

The formula (6) clearly exhibits the relativistic smallness (c is the velocity of light) of the strength of the interaction with a magnetic field.

Let a variable electric field $\mathbf{E}$ be applied to the system. Neglecting the effects of the retardation (i.e., assuming that $c\to\infty$), and varying the Lagrangian $L = L_T + \Delta L_E$ with respect to $\mathbf{T}$, we find, when only the first term in (1) is taken into account, that

$$-\frac{1}{2M_{\mathrm{T}}}\ddot{\mathbf{T}}-\alpha\mathbf{T}+\gamma\Delta\mathbf{T}+\frac{\lambda_E}{2}\dot{\mathbf{E}}=0. \tag{7}$$

In the presence of toroidal oscillations there arises the dynamical polarization

$$\mathbf{P}=\frac{\delta\Delta L_{\mathrm{E}}}{\delta\mathbf{E}}=-\lambda_{\mathrm{E}}\dot{\mathbf{T}}. \tag{8}$$

The toroidal-oscillation-related correction to the dynamical permittivity has the form

$$\Delta\varepsilon(\omega,\ \mathbf{q})=-4\pi\lambda_{\mathrm{E}}^2\omega^2M_{\mathrm{T}}/(\omega^2-\omega_{0\mathrm{T}}^2(\mathbf{q})). \tag{9}$$

We shall consider below only the quantity $\Delta\varepsilon(\omega, 0)$, i.e., the response to a uniform electric field. Introducing a special symbol for the eigenfrequency, i.e., setting $\omega_{0\mathrm{T}}^2(\mathbf{q}=0)=\Omega_{\mathrm{T}}^2$, we rewrite (9) for $\mathbf{q}=0$ in somewhat different form:

$$\Delta\varepsilon(\omega,\ 0)=\varepsilon(\omega)-\varepsilon'=-4\pi\lambda_{\mathrm{E}}^2\omega^2M_{\mathrm{T}}/(\omega^2-\Omega_{\mathrm{T}}^2), \tag{10}$$

where ε' is that contribution to the permittivity which is not

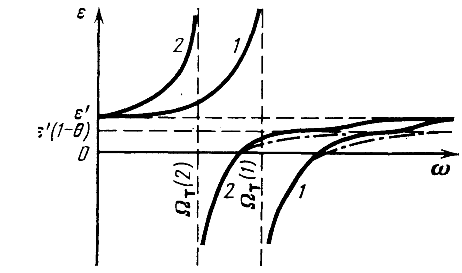

FIG. 1

connected with the toroidal degrees of freedom. It is clear that the expression (10) is invalid in the region of high frequencies ω, where we must retain the second term in (1). It is not difficult to verify that allowance for this term in the region $\omega \gg \Omega_T$ yields $\Delta\varepsilon(\omega) \propto -\omega^{-2} \to 0$, which ensures the fulfillment of the sum rules for the oscillator strengths.[1)]

In the case of the semiconductor model considered in the Appendix allowance for the second term in (1) is essential in the computation of $\varepsilon(\omega)$ in the region of frequencies $\omega \sim E_g \gg \Omega_T$.

Thus, the toroidal oscillations do not make a contribution to the static permittivity $\varepsilon(0)$. In the case when the toroidal oscillation mode softens (i.e., when $\Omega_T \to 0$) the behavior of the frequency dependence is quite distinctive, differing essentially from the behavior in the case when ferroelectric phase transitions occur. Figure 1 qualitatively depicts the variation of the dependence $\varepsilon(\omega)$ as Ω_T decreases (the curves 1 and 2 correspond to the values $\Omega_T(1) > \Omega_T(2) > 0$).

The interaction with the electric self-field produced during the toroidal oscillations leads to the renormalization of the frequencies of these oscillations.

When the retardation is ignored, the transverse toroidal modes are not renormalized (since the transverse electric field is equal to zero), but the longitudinal modes undergo substantial modifications. In fact let us add to the Lagrangian of the system the term

$$\Delta L_E' = \varepsilon' E^2 / 8\pi \qquad (11)$$

connected with the energy of the longitudinal electric field produced by the longitudinal toroidal oscillations.

We shall be interested only in uniform longitudinal oscillations. Varying the total Lagrangian $L = L_T + \Delta L_E + \Delta L_{E'}$ with respect to $\mathbf{T}$ and $\mathbf{E}$, we obtain the equation of motion for the toroidal-moment density,

$$-\frac{1}{2M_T}\ddot{\mathbf{T}} + \beta_4 \ddot{\ddot{\mathbf{T}}} - \alpha\mathbf{T} + \gamma\Delta\mathbf{T} + \frac{\lambda_E}{2}\dot{\mathbf{E}} = 0\,, \qquad (12)$$

and the Maxwell equation

$$\varepsilon'\mathbf{E} - 4\pi\lambda_E\dot{\mathbf{T}} = 0, \qquad (13)$$

the simultaneous solution of which leads to the following expression for the longitudinal-oscillation frequency $\omega_T^{\parallel}(\mathbf{q})$ for $\mathbf{q} = 0$:

$$\omega_T^{\parallel 2} = \frac{1}{2\beta_4}\left[\left(\frac{1}{4M_\parallel^2} + 4\beta_4\alpha\right)^{1/2} - \frac{1}{2M_\parallel}\right], \qquad (14)$$

$$\frac{1}{2M_\parallel} = \frac{1}{2M_T}\left(1 - \frac{4\pi\lambda_E^2 M_T}{\varepsilon'}\right). \qquad (15)$$

The frequency of the transverse toroidal oscillation is not, as has already been noted, renormalized:

$$\omega_T^{\perp 2} = \Omega_T^2 = 2\alpha M_T. \qquad (16)$$

The region of parameter values in which we must retain the term with the coefficient β_4 in (1) can be seen at once from the formulas (14) and (15). Indeed, if $M_\parallel > 0$ and, moreover, $16M_\parallel^2\beta_4\alpha \ll 1$, then we have from the (14) the relation

$$\omega_T^{\parallel 2} \approx \omega_T^{\perp 2}(1 - 4\pi\lambda_E^2 M_T/\varepsilon')^{-1}. \qquad (17)$$

It is clear that the expression (17) is not valid when the quantity in the parentheses has a small positive or negative value. As soon as the condition ${}^4\beta_4\alpha \gg (4M_\parallel^2)^{-1}$ begins to be fulfilled, the expression for $\omega_T^{\parallel}$ changes:

$$\omega_T^{\parallel 2} \approx (\alpha/\beta_4)^{1/2}. \qquad (18)$$

Finally, when $M_\parallel < 0$ and ${}^4\beta_4\alpha \ll (4M_\parallel^2)^{-1}$, we have

$$\omega_T^{\parallel 2} \approx (2\beta_4|M_\parallel|)^{-1}. \qquad (19)$$

The region of applicability of the relations obtained is, in principle, limited by the stipulation that the term with the coefficient β_4 should be small in comparison with the term with the coefficient β_2 in the expression (1) (otherwise the power series expansion $\dot{T}$ becomes invalid, and we fall within the region of high-frequency oscillations). This condition is met when $\omega_T^{\parallel 2} \ll (2\beta_4 M_T)^{-1}$, and can be violated in the case of an arbitrary relation between the coefficients of the Lagrangian (this applies especially to the expression (19), which is not applicable when $|1 - 4\pi\lambda_E^2 M_T/\varepsilon'| > 1$). The relations (17) and (18) satisfy the condition for the frequency of the toroidal oscillations to be low at low values of the coefficient α (i.e., at values not too far from the toroidal instability threshold).

Thus, when $4\pi\lambda_E^2 M_T/\varepsilon' \gtrsim 2$, the low longitudinal toroidal oscillations cannot, in principle, be low-frequency oscillations (in the semiconductor model the frequencies of such oscillations are of the order of E_g). Therefore, below all the investigations are carried out under the assumption that $4\pi\lambda_E^2 M_T/\varepsilon' \ll 1$. The case of high-frequency oscillations requires special treatment within the framework of a specific microscopic model.

§3. EFFECT OF A CONSTANT MAGNETIC FIELD ON THE DYNAMICAL PERMITTIVITY OF CRYSTALS WITH TOROIDAL OSCILLATIONS

Let us consider a nonmagnetic cubic crystal[2)] located in a constant external magnetic field $\mathbf{H}$ and a variable electric field $\mathbf{E}(t)$. The system's Lagrangian, as compared with the Lagrangian in §2, should include additional terms connected with the effect of the magnetic field (we consider the approximation linear in $\mathbf{H}$):

$$\Delta L_H^{(1)} = \nu\mathbf{H}[\mathbf{T}\times\dot{\mathbf{T}}], \qquad (20)$$

$$\Delta L_H^{(2)} = \mu[\mathbf{H}\mathbf{E}\times]\mathbf{T}. \qquad (21)$$

The terms (20) and (21) are connected with the magnetoelectric effect in the system ($\mathbf{E}$ is the external electric field and

$\mathbf{P} \propto \dot{\mathbf{T}}$ is the polarization of the system in the presence of toroidal oscillations).

The "equation of motion" for the toroidal-moment density in the electric and magnetic fields has the form

$$\delta L_T/\delta \mathbf{T}+\lambda_E\dot{\mathbf{E}}+\mu[\mathbf{H}\times\mathbf{E}]+2\nu[\mathbf{H}\times\dot{\mathbf{T}}]=0. \tag{22}$$

It is not difficult to find the eigenfrequencies of the toroidal oscillations in a magnetic field form (22) after setting $\mathbf{E}=0$. After simple computations we find for the frequencies of the oscillations in the (x, y) plane and along the z axis the expressions

$$(\omega_T)_{xy}^{1,2}=[\Omega_T^2+(M_T\nu H)^2]^{1/2}\pm|M_T\nu H|, \tag{23}$$

$$(\omega_T)_z=\Omega_T. \tag{24}$$

In a weak magnetic field, i.e., for $M_T\nu H \ll \Omega_T$, the toroidal-oscillation frequency shift turns out to be linear in the field $\mathbf{H}$, and, moreover, the dengeneracy of the clockwise- and counterclockwise-polarized oscillations in the (x, y) plane is lifted.

The dynamical permittivity tensor is calculated in much the same way as in §2. After tedius computations we arrive at the following result:

$$\Delta\varepsilon_{zz}(\omega)=-4\pi\lambda_E^2\frac{\omega^2 M_T}{\omega^2-\Omega_T^2}, \tag{25}$$

$$\Delta\varepsilon_{xx}(\omega)=\Delta\varepsilon_{yy}(\omega)=-4\pi(\omega^2-\Omega_T^2)(\omega^2\lambda_E^2+\mu^2H^2)M_T/D(\omega), \tag{26}$$

$$\Delta\varepsilon_{xy}(\omega)=\Delta\varepsilon_{yx}^*(\omega)=4\pi i L(\omega)/D(\omega), \tag{27}$$

$$L(\omega)=2\omega H[\lambda_E^2M_T^2\omega^2\nu-\mu^2H^2\nu M_T^2-(\omega^2-\Omega_T^2)M_T\lambda_E\mu],$$
$$D(\omega)=(\omega^2-\Omega_T^2)^2-4M_T^2\nu^2\omega^2H^2. \tag{28}$$

In the static limit $\omega \to 0$ there remain nonzero corrections only for the following components:

$$\Delta\varepsilon_{xx}(0)=\Delta\varepsilon_{yy}(0)=4\pi M_T\mu^2H^2/\Omega_T^2. \tag{29}$$

Thus, in an external magnetic field H the components $\varepsilon_{xx}(0)$ and $\varepsilon_{yy}(0)$ of the static permittivity tensor diverge in the vicinity of the toroidal transition point (i.e., as $\Omega_T \to 0$), but the component $\varepsilon_{zz}(0)$ does not exhibit any anomalies. The dynamical permittivity is characterized by the fact that the $\varepsilon_{xx}(\omega)$, $\varepsilon_{yy}(\omega)$, and $\varepsilon_{xy}(\omega)$ bands are split, and also by the fact that the dependence $\varepsilon_{xy}(\omega)$ is linear in the region of small $\omega \ll \Omega_T$. Let us emphasize that only the totality of all the indicated anomalies connected with the presence of the invariants (20) and (21) could unambiguously indicate the detection of toroidal oscillations in optical experiments.

§4. SINGULARITIES OF THE SPATIAL DISPERSION OF TOROIDAL OSCILLATIONS IN NONCENTROSYMMETRIC CRYSTALS

As has already been noted in the Introduction, inhomogeneous (incommensurate) toroidal structures are interesting first and foremost because of the occurrence of a macroscopic spontaneous current $\mathbf{j} \propto \text{curl curl } \mathbf{T}$ in them. The possibility in principle of realizing such an incommensurate structure as a result of the variation of the sign of the second-order gradient term in the functional (2) is considered in Ref. 1 in the model proposed there. Here we shall analyze another possibility connected with a specific contribution to the functional (2) in noncentrosymmetric systems. Let the crystal symmetry of the normal phase admit of the existence of a pseudoscalar η (e.g., in the T_d class). Then to the functional (2) must be added the term

$$\Delta U_T^{(1)}=\eta\mathbf{T}\,\text{rot}\,\mathbf{T}. \tag{30}$$

A similar situation obtains in magnetic materials,[9] and the analysis of the contribution (30) is entirely similar to the analysis performed by Dzyaloshinskiĭ[10] for the helical magnetic structure. In systems with an invariant of the type (20) the eigenfrequencies of the transverse toroidal oscillations have the form

$$\omega_{T\pm}^{\perp 2}=\omega_{0T}^2(\mathbf{q})\pm|2\eta M_T q|, \quad \mathbf{q}\perp\mathbf{T}. \tag{31}$$

The minimum toroidal-oscillation frequency is attained for the lower branch $(\omega_T^{\perp})_-$ at $q_0=\eta/2\gamma$:

$$\omega_{T\,min}^{\perp 2}=2M_T(\alpha-\eta^2/4\gamma). \tag{32}$$

In the case when the expression (32) vanishes, the transverse structure of the toroidal moment is helicoidal in the region below the transition point:

$$T_x \propto T_0\cos q_0 z, \qquad T_y \propto T_0 \sin q_0 z, \tag{33}$$

where T_0 is the amplitude of the toroidal moment; correspondingly, the spontaneous current $\mathbf{j}(\mathbf{r})$ also has a helicoidal character.

Somewhat more complicated is the case of systems that admit the existence of a polar vector $\mathbf{u}_0$ (these may, in particular, be crystals of the pyroelectric classes, or crystals located in an external homogeneous electric field $\mathbf{E}$). Let us add to the expression (2) the invariant

$$\Delta U_T^{(2)}=\lambda\mathbf{u}_0[\mathbf{T}\times\text{rot}\,\mathbf{T}], \tag{34}$$

and let us, for simplicity, neglect the terms of the type $\lambda'(\mathbf{u}_0\mathbf{T})^2$, assuming that $\lambda' \ll \lambda^2/\gamma$, where γ is the coefficient of the gradient term in (2).

Let us consider the eigenfrequencies of the toroidal oscillations under the assumption that $4\pi\lambda_E^2/\varepsilon' \ll 1$, under which condition a strong stiffening of the longitudinal modes does not occur (see §2) and $M_\parallel \approx M_T$. It is easy to show that the dispersion law for $\omega_T(\mathbf{q})$ contains three branches:

$$(\omega_T^2)_1=\omega_{0T}^2(q),$$
$$(\omega_T^2)_{2,3}=\omega_{0T}^2(\mathbf{q})\pm|\lambda\mathbf{u}_0 M_T q_\perp|, \tag{35}$$
$$q_\perp=(q_x^2+q_y^2)^{1/2}, \qquad q^2=q_\perp^2+q_z^2,$$

where the z axis is oriented along $\mathbf{u}_0$. The minimum of the frequency $(\omega_T)_3$ is attained on the (q_x, q_y) line in the (x, y) plane at

$$q_{0\perp}=|\lambda u_0|/4\gamma. \tag{36}$$

It is given by the expression

$$\omega_{T\,min}=\Omega_T^2-(\lambda u_0)^2M_T/8\gamma. \tag{37}$$

The toroidal moment has a mixed longitudinal-transverse structure in the region below the phase transition point. In the simplest variant, for which $q_y=q_z=0$ and $q_x \neq 0$, we have

$$T_x \propto T_0 \cos q_\perp{}^0 x, \qquad T_z \propto T_0 \sin q_\perp{}^0 x, \qquad T_y=0. \tag{38}$$

In another variant, for which $q_x = q_y = q_\perp^0/\sqrt{2}$ and $q_z = 0$, we have

$$T_x=T_y \propto T_0 \cos[q_\perp{}^0(x+y)/\sqrt{2}], \qquad T_z \sim T_0 \sin[q_\perp{}^0(x+y)/\sqrt{2}]. \tag{39}$$

The choice of a particular structure in the region below the transition point should be made with allowance for the higher-order terms in the functional (2), and the corresponding analysis is not carried out in the present paper. The spontaneous current $\mathbf{j}$ in the case of the configurations (38) and (39) flows along the polar axis $\mathbf{u}_0$, but in more complicated configurations it may turn out to be nonzero in the transverse plane as well.

It should be especially noted that the coefficients η and λ do not contain insignificant relativistic contributions, since the toroidal moments in crystals (i.e., the toroidal moments under investigation here) are due to the orbital motion of the electrons. Therefore, the considered mechanisms of the formation of the nonhomogeneous structures can be quite effective (thus, in the microscopic two-band model the quantities η and λ are connected with the electron-phonon interaction). Naturally, the analysis carried out above is valid only when the toroidal inhomogeneities have macroscopic dimensions ($q^0 \ll a^{-1}$, where a is the lattice constant).

§5. TOROIDAL POLARITONS

In the nonrelativisitic approximation the transverse toroidal oscillations do not interact with the electromagnetic field. Allowance for the retardation leads to the mixing up of the transverse toroidal and electromagnetic oscillations. To find the eigenfrequencies of the mixed toroidal-photon modes, let us write down the Lagrangian of the "field + medium" system in the lowest approximation in the vector potential $\mathbf{A}(\mathbf{r}, t)$ and the toroidal-moment density $\mathbf{T}(\mathbf{r},t)$:

$$L=L_T+L_A+L_A', \tag{40}$$

where L_T is given by the formula (3),

$$L_A=-\lambda_E \mathbf{T}\frac{1}{c}\ddot{\mathbf{A}}+\lambda_B \mathbf{T}\frac{1}{c}\operatorname{rot}\operatorname{rot}\mathbf{A}, \tag{41}$$

$$L_A'=\frac{\varepsilon'}{8\pi c^2}(\dot{\mathbf{A}})^2-\frac{(\operatorname{rot}\mathbf{A})^2}{8\pi}, \quad \mathbf{E}=-\frac{1}{c}\dot{\mathbf{A}}, \quad \mathbf{B}=\operatorname{rot}\mathbf{A}, \tag{42}$$

and $\mathbf{A}(\mathbf{r}, t)$ is the vector potential, given in the transverse gauge $\operatorname{div}\mathbf{A} = 0$, of the electromagnetic field.

Varying the Lagrangian (40) with respect to $\mathbf{T}$ and $\mathbf{A}$, we obtain the system of equations

$$\frac{\delta L}{\delta \mathbf{T}}-\lambda_E\frac{1}{c}\ddot{\mathbf{A}}+\lambda_B\frac{1}{c}\operatorname{rot}\operatorname{rot}\mathbf{A}=0, \tag{43}$$

$$-\frac{\varepsilon'}{4\pi c^2}\ddot{\mathbf{A}}-\frac{1}{4\pi}\operatorname{rot}\operatorname{rot}\mathbf{A}-\lambda_E\frac{1}{c}\dddot{\mathbf{T}}+\lambda_B\frac{1}{c}\operatorname{rot}\operatorname{rot}\mathbf{T}=0. \tag{44}$$

We can, under the assumption that $M_\parallel > 0$ and $\theta \equiv 1 - M_T/M_\parallel \ll 1$, discard the term with β_4 in the kinetic energy (1). For the eigenfrequencies of the transverse toroidal-photon oscillations we find from (43) and (44) the equation

$$(\omega^2-\omega_{0T}^2)\left(\omega^2-\frac{c^2q^2}{\varepsilon'}\right)-\frac{4\pi M_T}{\varepsilon'}(\lambda_E\omega^2+\lambda_B q^2)^2=0, \tag{45}$$

whence we find in the lowest approximation in the parameter θ that

$$(\omega_T^{\perp 2})_{1,2}=\frac{1}{2(1-\theta)}\left\{\omega_{0T}^2+\frac{c^2q^2}{\varepsilon'}\pm\left[\left(\omega_{0T}^2-\frac{c^2q^2}{\varepsilon'}\right)^2+4\theta q^2\left(\frac{c^2}{\varepsilon'}\omega_{0T}^2+\frac{\lambda_B^2}{\lambda_E^2}q^2\right)\right]^{1/2}\right\}. \tag{46}$$

In the region of strongest intermixing of the toroidal and electromagnetic oscillations (i.e., for $q_1{}^2 \sim \omega_{0T}^2\, \varepsilon'/c^2$) the second term in the square brackets under the radical sign is small compared to the first. We find from dimensional considerations that $\lambda_B/\lambda_E \sim v^2$, where v is the characteristic velocity in the electron subsystem (for the two-band model of semiconductors $v = (2E_g/m^*)^{1/2}$, where m^* is the effective mass). Since $v \ll c$, we find that the second term is comparable to the first in the region of momenta

$$q_2{}^2 \sim c^2\omega_{0T}{}^2/v^4\varepsilon' = (c/v)^4 q_1{}^2/\varepsilon'^2 \gg q_1{}^2.$$

In the momentum region $q \sim q_1$ we can, with a high degree of accuracy, neglect the gradient terms in (3) and assume that $\omega_{0T} \approx \Omega_T$.

The asymptotic expressions for the transverse-oscillation frequencies (46) in the case when $c^2q^2 \gg \varepsilon'\Omega_T^2$ have the form

$$(\omega_T{}^{\perp 2})_1 \approx c^2q^2/\varepsilon'(1-\theta), \qquad (\omega_T{}^{\perp 2})_2 \approx \Omega_T{}^2. \tag{47}$$

For $\mathbf{q} \to 0$ we obtain

$$(\omega_T{}^{\perp})_1 \approx 0, \qquad (\omega_T{}^{\perp 2})_2 \approx \Omega_T{}^2/(1-\theta) = \omega_T{}^{\parallel 2}. \tag{48}$$

The plot of the function $\omega_T^\perp(q)$ is qualitatively shown[3)] in Fig. 2.

Thus, the combined transverse toroidal-photon oscillations can be described in terms of distinctive polaritons with the dispersion law $\omega_T^\perp(\mathbf{q})$. This indicates the possibility of observing toroidal oscillations in Raman light scattering experiments, as well as in reflection or absorption experiments. But actually the toroidal mode can be confused with the

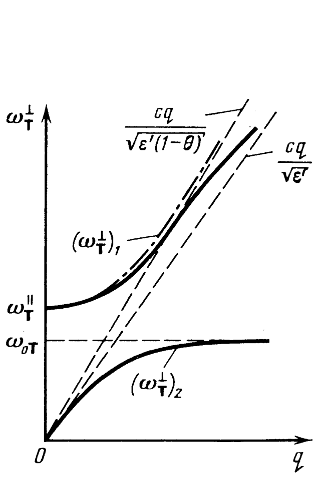

FIG. 2.

phonon modes in the infrared frequency region, and it is necessary to construct a theory that takes account of this interrelationship. The simplest variant of such a theory is considered in the following section.

§6. TOROIDAL-PHONON OSCILLATIONS IN POLAR CRYSTALS

Let us consider the situation in which the transverse toroidal mode of the electronic oscillations falls within the region of optical phonon frequencies of a polar crystal. Within the framework of the Lagrangian formalism, we must write down the following expression for the effective Lagrangian of the system:

$$L=L_T+L_{ph}+L_{T-ph}, \tag{49}$$

where L_T is given by the expression (3), while

$$L_{ph}=\frac{1}{2M}[(\dot{\mathbf{u}})^2-\Omega_{ph}^2\mathbf{u}^2] \tag{50}$$

is the usual phonon Lagrangian, M is the mass of the optical phonon, Ω_{ph} is the frequency of this phonon (with allowance for the renormalization resulting from the interaction with the electron-polarization density oscillations, which we assume to have sufficiently high frequencies), and $\mathbf{u}(\mathbf{r}, t)$ is the optical displacement of the sublattices in the polar crystal. The third term in (49) has the form

$$L_{T-ph}=\lambda_u[\mathbf{T}\dot{\mathbf{u}}-\mathbf{u}\dot{\mathbf{T}}]. \tag{51}$$

The term (51) has its origin in the interaction between the toroidal moment $\mathbf{T}$ and the displacement current $\mathbf{j}\propto\dot{\mathbf{u}}$ produced as a result of the polar vibrations of the lattice (the second term has been added in order to give (51) a symmetric form, and reduces, up to a total time derivative, to the first term).

Let us consider the transverse toroidal-phonon vibrations without allowance for the spatial dispersion. After varying the Lagrangian (49) with respect to $\mathbf{T}$ and $\mathbf{u}$, we arrive at the following equation for the determination of the eigenfrequencies:

$$(\omega^2-\Omega_T^2)(\omega^2-\Omega_{ph}^2)=\lambda_u^2\omega^2MM_T, \tag{52}$$

$$\omega_{1,2}^2=\tfrac{1}{2}\{\Omega_T^2+\Omega_{ph}^2+\lambda_u^2MM_T\pm[[(\Omega_T+\Omega_{ph})^2+\lambda_u^2MM_T][(\Omega_T-\Omega_{ph})^2+\lambda_u^2MM_T]]^{1/2}\}. \tag{53}$$

Let us note that, for crystals that undergo second-order structural (ferroelectric) phase transitions, the condition $\omega_2=0$ for the vanishing of the soft mode coincides with the condition $\Omega_{ph}=0$, i.e., does not depend on the coupling between the toroidal and phonon modes. This is natural, since the toroidal and phonon modes do not intermix in the static limit. But the rate of softening of the initial stable mode ω_2 changes, and from (53) we find that for $\Omega_{ph}\to 0$,

$$\omega_2^2=\Omega_{ph}^2\frac{\Omega_T^2}{\Omega_T^2+\lambda_u^2MM_T}<\Omega_{ph}^2,\quad \Omega_{ph}^2\ll\Omega_T^2,\quad \lambda_u^2MM_T. \tag{54}$$

Figure 3 shows a plot of the function $\omega(\Omega_{ph})$ describing the solution (53). The toroidal and phonon modes get uncoupled in the frequency region where $\Omega_{ph}^2\gg\lambda_u^2MM_T$:

$$\omega_1^2\to\Omega_T^2,\qquad \omega_2^2\to\Omega_{ph}^2. \tag{55}$$

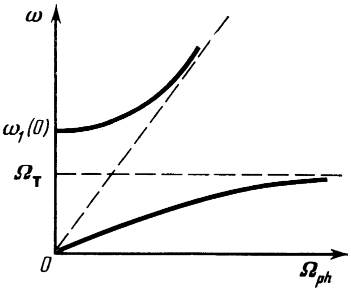

FIG. 3.

Thus, there occurs in the slope $\partial\omega_2/\partial\Omega_{ph}$ of the function $\omega(\Omega_{ph})$ for the low-frequency branch of the toroidal-phonon excitation a characteristic change that can be detected by the methods of *IR* spectroscopy or in Raman light scattering experiments. Of special interest is the $\Omega_{ph}\sim\Omega_T$ case, in which strong intermixing of the two types of vibration occurs, and a gap exists in the frequency spectrum (Fig. 3). If, for example, Ω_{ph} varies with temperature according to the law $\Omega_{ph}^2\propto|T-T_c|$, then a splitting of the "soft" mode will occur in the spectral characteristic at certain temperatures $T^*>T_c$ where $\Omega_{ph}\sim\Omega_T$; one of the branches will continue to soften right down to the transition point ($T=T_c$), though with a different slope, but the frequency of the other branch will remain finite, and will attain the value $\omega_1=(\Omega_T^2+\lambda_u^2MM_T)^{1/2}$ at $T\to T_c$.

Within the framework of the toroidal-oscillational concept, it is possible[11] to explain the unusual temperature dependences of the spectral characteristics in the $TlGaSe_2$ compound.[12]

Another important effect connected with the intermixing of the toroidal and phonon modes manifests itself upon the application of an external magnetic field $\mathbf{H}$. As noted in Ref. 4, the toroidal ordering in crystals should be accompanied by the occurrence of the magnetoelectic effect. In our case the application of a magnetic field $\mathbf{H}$ leads, in the approximation linear in $\mathbf{H}$, to the appearance in the system's Lagrangian of the mixed term

$$\Delta L_{u,H}=\xi[\mathbf{T}\times\mathbf{H}]\mathbf{u}, \tag{56}$$

which is similar in structure to the "magnetoelectric" term in Ref. 4.

Taking (56) into account, we obtain for the eigenfrequencies of the toroidal-phonon vibrations in the plane, (x, y), perpendicular to the direction of the vector $\mathbf{H}$, which is assumed to be oriented along the z axis, the relation

$$[\omega^{\perp 2}-\Omega_{ph}^2][\omega^{\perp 2}-\Omega_T^2]=(\lambda_u\omega^\perp\pm\xi H)^2MM_T; \tag{57}$$

the relation (52) remains valid for the vibrations along the z axis. In the region above the transition point we obtain for the transverse (with respect to the z axis) low-frequency mode in the case when $\lambda_u\omega^\perp\ll\xi H$ the expression

$$\omega^{\perp 2}=[(\Omega_{ph}^2\Omega_T^2-\xi^2H^2MM_T)/2\lambda_u\xi HMM_T]^2, \tag{58}$$

i.e., a structural transition with the displacement vector lying in the (x, y) plane occurs provided

$$\Omega_{ph}^2=\xi^2H^2MM_T/\Omega_T^2. \tag{59}$$

Thus, an anisotropy of the "easy plane" type arises in the system, and the transition with the displacement vector in the (x, y) plane occurs at a higher temperature $T_c(\mathbf{H})$ (i.e., $\Omega_{ph}^2 \sim T - T_c^0$, where T_c^0 is the transition temperature in the absence of a magnetic field), which can be determined from (59). Let us note that a rise in $T_c(\mathbf{H})$ has been observed[13] in some narrow-band ferroelectric semiconductors, although we do not as yet have sufficient reasons to relate this fact only to the influence of the toroidal oscillations.

The transition temperature shift predicted by the expression (59) can be appreciable even in relatively weak fields, since the expression for the coefficient ξ does not contain the small relativistic factor characteristic of the magnon-phonon coupling.

We must specially discuss magnetoferroelectrics, in which the rise in the strucutral transition temperature can be many times greater (in place of $\mathbf{H}$ we should substitute $\mathbf{B} \gg \mathbf{H}$). It is clear that in this case the ferroelectric transition temperature T_c should be lower than the Curie temperature T_C. It would have been quite interesting to investigate of the long-range magnetic order on the ferroelectric transition temperature shift in magnetoferroelectrics with $T_C > T_c$.

§7. THE MAGNETIC SUSCEPTIBILITY IN NONCENTROSYMMETRIC CRYSTALS

Let us consider the contribution of the toroidal oscillations to the dynamical magnetic susceptibility $\chi(\omega)$ of noncentrosymmetric crystals without allowance for the spatial dispersion. For definiteness, let the crystal belong to one of the pyroelectric classes, and let $\mathbf{u}_0$ be a vector in the direction of the polar axis. Because of the presence of the invariant

$$\Delta U_{\mathrm{T}}^{(3)} = \lambda_1 \mathbf{u}_0 [\mathbf{T} \times \mathbf{H}], \tag{60}$$

where $\mathbf{H}$ is the magnetic field intensity, there arise additional contributions to the components χ_{xx}, χ_{yy}, and χ_{xy}, of the susceptibility tensor (the z axis is oriented along $\mathbf{u}_0$). The corresponding unwieldy expressions are similar to the expressions derived in §3, and we do not give them here. In the static limit ($\omega \to 0$), we have

$$\Delta\chi_{xx}(0) = \Delta\chi_{yy}(0) = \lambda_1^2 \mathbf{u}_0^2 M_{\mathrm{T}}/\Omega_{\mathrm{T}}^2, \quad \Delta\chi_{xy}(0) = \Delta\chi_{zz}(0) = 0. \tag{61}$$

The components $\chi_{xx}(0)$ and $\chi_{yy}(0)$ diverge at the toroidal transition point (i.e., as $\Omega_{\mathrm{T}} \to 0$), and a ferromagnetic order with the easy plane type of anisotropy arises below the transition point together with the toroidal order. It is clear that the results will remain valid in the case of centrosymmetric crystals located in an external electric field $\mathbf{E}$.

Thus, in pyroelectric crystals, the toroidal phase transition should be accompanied by ferromagnetic ordering. And, conversely, ferromagnetic ordering in pyroelectric (in particular, ferroelectric) crystals should induce toroidal ordering.[14]

If the absence of a center of inversion is not due to the appearance of a polar aixs, but the symmetry admits of the existence of a pseudoscalar η, then the magnetic susceptibility χ will also diverge at the toroidal transition point because of the presence of the invariant

$$\Delta U_{\mathrm{T}}^{(4)} = \eta \mathbf{T}\mathbf{H}. \tag{62}$$

In the static limit

$$\Delta\chi(0) = \eta^2 M_{\mathrm{T}}/\Omega_{\mathrm{T}}^2 \to \infty \quad \text{as} \quad \Omega_{\mathrm{T}} \to 0. \tag{63}$$

Thus, the softening of the toroidal mode of the vibrations can be the cause of the appearance of the ferromagnetic order in noncentrosymmetric crystals. It is possible that the ferromagnetism observed in the $\mathrm{GaMo_4S_8}$ crystal[15] with the imperfect spinel structure is precisely of the nature (for greater detail, see the discussion in Ref. 14).

The singularities of the toroidal-oscillation dispersion law (see §4) may give rise to a helicoidal, and not a ferromagnetic, structure in noncentrosymmetric crystals. In this connection, an interesting potential object for investigation could be the itinerant-electron ferromagnet *MnSi*, although there are for the present not enough experimental data to confirm the toroidal nature of the helicoidal[16] magnetic structure in this compound.

§8. CONCLUSION

In the present paper we have predicted a number of toroidal-oscillation-related effects that can be detected in optical experiments.

1. An unusual frequency dependence of the dynamical permittivity $\varepsilon(\omega)$, that can be determined from reflection or absorption spectra.

2. The divergence of the static permittivity at the toroidal transition point and, as a result, the ferroelectric ordering in crystals located in an external magnetic field, or possessing a ferromagnetic order.

3. The existence in the infrared and Raman-scattering spectra of additional peaks due to the presence of the soft toroidal modes. First order frequencies should appear in the Raman scattering spectrum at the toroidal phase transition (just as happens at the instability threshold of a Raman active phonon mode), but a structural transition does not occur then, i.e., no additional reflections occur in the x-ray spectra.

4. The "splitting" of the soft phonon mode and a change in the temperature dependence of the mode as the structural transition point is approached in crystals with intermixed toroidal and lattice vibrations.

5. A rise in the structural transition temperature in a magnetic field. This effect can be estimated within the framework of the two-band semiconductor model. The effective field H_{eff} in which the critical width of the forbidden band of the semiconductor undergoes a relative change $\Delta E_g / E_g^* \sim 10^{-2}$ is of the order of 100 kOe ($E_g^* \sim E_{\mathrm{ex}}$, where E_{ex} is the exciton binding energy). Similar fields are required for comparable transition temperature shifts.

6. The narrowing of the transmission band $\omega_{0\mathrm{T}} \leqslant \omega \leqslant \omega_{\mathrm{T}}^{\parallel}$) in the spectrum of the toroidal polaritons as we approach the toroidal transition temperature T_t. Notice that $\omega^{\parallel} \approx$ const in the case of structural and ferroelectric transitions, and the gap in the polariton spectrum widens as $\omega^{\perp} \to 0$. In our case (when $M_{\parallel} > 0$), $\omega_{\mathrm{T}}^{\parallel} \to 0$ as $\omega_{0\mathrm{T}} \to 0$, i.e., the gap in the polariton spectrum decreases.

7. The divergence of the static magnetic susceptibility and the appearance of magnetic ordering upon the occurrence of a toroidal transition in a noncentrosymmetric crys-

tal, or in a crystal located in an external electric field.

Naturally, the indicated effects do not exhaust the wealth of phenomena connected with toroidal oscillations. In particular, the toroidal-magnon oscillations, which can occur in, for example, ferromagnets in the case when $T_t < T_C$, deserve to be specially investigated.

Suitable materials for the detection of toroidal oscillations may turn out to be substances in which the electric and magnetic properties are strongly interrelated (magnetoelectric[17] and magnetoferroelectric[18] materials) and, possibly, certain narrow-band semiconductors: ferroelectrics.

Thus authors express their gratitude to N. E. Alekseeviskiĭ, A. S. Borovik-Romanov, B. A. Volkov, and V. L. Ginzburg for a discussion of the results and valuable comments.

APPENDIX

As an illustration of the general relations obtained in §§2–5, let us compute the parameters of the toroidal oscillations $M_{\mathbf{T}}$, λ_E, λ_B, β_4, and μ) in the microscopic two-band **kP** model a superconductor with a forbidden-band with $E_g \gtrsim E_{ex}$, where E_{ex} is the exciton binding energy (the analysis is being performed for zero temperature). We choose the model Hamiltonian in the form

$$\hat{H}=\begin{bmatrix} \frac{1}{2m_1}\left(\hat{\mathbf{k}}-\frac{e}{c}\mathbf{A}\right)^2+\frac{E_g}{2}+e\Phi & \frac{1}{m}\mathbf{P}_{12}\left(\hat{\mathbf{k}}-\frac{e}{c}\mathbf{A}\right)-\Delta_{12} \\ \frac{1}{m}\mathbf{P}_{21}\left(\hat{\mathbf{k}}-\frac{e}{c}\mathbf{A}\right)-\Delta_{21} & -\frac{1}{2m_2}\left(\hat{\mathbf{k}}-\frac{e}{c}\mathbf{A}\right)^2-\frac{E_g}{2}+e\Phi \end{bmatrix}, \tag{A.1}$$

where m_1 and m_2 are the effective electron masses in the bands 1 and 2, $\mathbf{P}_{12}$ is the interband matrix element of the momentum, m is the free electron mass, $\mathbf{A}$ and Φ are the vector and scalar potentials of the electromagnetic field, and Δ_{12} is the order parameter,[1–4] which we assume below to be purely imaginary ($\Delta_{12}=\Delta_{21}^*=i\Delta_{\mathrm{Im}}$).

Let us write the self-consistency equation for the quantity $\Delta_{\mathrm{Im}}(\mathbf{r},t)$ in the approximation linear in Δ_{Im} and $\mathbf{P}_{12}$ and in the lowest approximations in $\mathbf{A}$ and Φ, replacing, for simplicity, the interelectron interaction potential by an effective constant g_{Im}. This yields

$$\alpha\Delta_{\mathrm{Im}}=-\beta_2\ddot{\Delta}_{\mathrm{Im}}+\beta_4\ddddot{\Delta}_{\mathrm{Im}}-\frac{\lambda_E}{2}\dot{\mathbf{E}}\mathbf{n} -\frac{\lambda_B}{2c}\mathbf{n}\,\mathrm{rot}\,\mathbf{B}-\frac{\mu}{2}[\mathbf{E}\times\mathbf{B}]\,\mathbf{n}, \tag{A.2}$$

$$\mathbf{E}=-\frac{1}{c}\dot{\mathbf{A}}-\mathrm{grad}\,\Phi,\quad \mathbf{B}=\mathrm{rot}\,\mathbf{A},$$

$$\alpha=\frac{1}{g_{\mathrm{Im}}\overline{N}}-\ln\frac{2W}{E_g}=\ln\frac{E_g}{E_g^\cdot}>0, \tag{A.3}$$

$$\beta_2=\frac{1}{16E_g^2},\quad \beta_4=\frac{5}{256E_g^4}, \tag{A.4}$$

$$\lambda_E=\frac{\pi}{6}\frac{e}{E_g^3}\frac{|\mathbf{P}_{12}|}{m},\quad \lambda_B=\frac{\pi}{5}\frac{e}{M}\frac{|\mathbf{P}_{12}|}{E_g^2 m}, \tag{A.5}$$

$$\mu=-\frac{\pi}{48}\frac{e^2\delta|\mathbf{P}_{12}|}{M^2cE_g^2m}, \tag{A.6}$$

where $\overline{N}=M^{3/2}E_g^{1/2}/2\pi^2$, W is the cutoff energy, which is of the order of the forbidden-band width, $\delta=m_1-m_2$, $M=(m_1+m_2)/2$, and it is assumed that $\delta\ll M$; $\mathbf{n}$ is the unit vector in the direction of $\mathbf{P}_{12}$. The toroidal moment for the uniaxial system (A.1) is introduced in the following manner:

$$\mathbf{T}=\mathbf{n}\Delta_{\mathrm{Im}}, \tag{A.7}$$

after which we obtain for the component T_z (the z axis is oriented along $\mathbf{P}_{12}$) a dynamical equation (similar to (A.2)) for which the Lagrangian (3) can be recovered.

[1)]The authors thank B. A. Volkov, who drew their attention to this circumstance. In Fig. 1 the continuous and dot-dash lines respectively depict the dependence $\varepsilon(\omega)$ with and without allowance for the second term in the kinetic energy (1).

[2)]The results obtained in §3 can be generalized for a ferromagnet with $T_C T_t$, where T_C is the Curie temperature and T_t is the toroidal transition temperature, by making the substitution $\mathbf{H}\to\mathbf{H}+4\pi\mathbf{M}$ ($\mathbf{M}$ is the magnetic moment of the crystal).

[3)]Allowance for the higher time derivatives in (1) leads in the high-frequency region to the correct asymptotic form $(\omega_{\mathbf{T}}^{\perp})_1{}^2\to c^2q^2/\varepsilon'$ (the continuous curve).

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