

# Toroidal collective excitations and the optical properties of crystals

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A new type of collective excitations in solids is investigated which is connected with the oscillations of the toroidal-moment density of the band electrons. The natural frequencies and logarithmic decrements of these oscillations are computed for different types of ground state (specifically, for ferroelectric, toroidal-orbital-magnetic, and spin-antiferromagnetic substances) according to the two-band model. Optical effects in which toroidal oscillations manifest themselves are investigated.

## §1. INTRODUCTION

A toroidal magnetic substance is a peculiar type of band electron antiferromagnet, characterized by a nonzero macroscopic toroidal moment  $\mathbf{T}_0$  in the ground state. Characteristic of this class of materials are a strong (without a small relativistic parameter) interrelationship between the magnetic and electric phenomena, a considerable reconstruction of the single-electron spectrum of the quasiparticles in the ordered phase [with  $E(\mathbf{k}) \neq E(-\mathbf{k})$  even in the exchange approximation], and unusual magnetic and electric properties.<sup>1–3</sup> In toroidal magnetic materials special collective oscillations (toroidal oscillations) can propagate whose properties differ quite sharply from those of the well-known types of excitation in crystals.

Kopaev and one of the present authors<sup>4</sup> have already constructed a phenomenological theory of toroidal oscillations, and have investigated by means of the effective Lagrangian method certain general properties of this new type of oscillations in crystals in the region close to the toroidal phase transition point. But a number of important questions (the damping of the toroidal oscillations, their contribution to the optical properties of toroidal magnetic substances at points far from the transition point, the characteristics of the high-frequency branches of the toroidal oscillations) that are beyond the scope of the purely phenomenological approach need to be investigated independently, using specific microscopic models.

Indeed, the spectra of toroidal oscillations, generally speaking, exhibit a gap; therefore, the hydrodynamic approach developed in Ref. 4 is correct only in the low-frequency region (this condition holds, for example, in the vicinity of the instability point, where the toroidal mode is anomalously soft).

Thus, the present paper is a direct continuation of Ref. 4. The geometrical form of toroidal oscillations can be depicted by considering a variable poloidal current flowing around a toroidal coil (Figs. 1a and 1b). The toroidal moment of the coil varies with the current density  $\mathbf{j}(\mathbf{r}, t)$  (Ref. 5):

$$\mathbf{T}(t) = \frac{1}{10c} \int [\mathbf{r}(\mathbf{r}\mathbf{j}) - 2r^2\mathbf{j}] d^3\mathbf{r},$$

and the  $\mathbf{T}(t)$  oscillations about the “zero” position  $\mathbf{T}_0$  [for  $\mathbf{j} = \mathbf{j}_0(\mathbf{r})$ ] can be either longitudinal or transverse. In the

general case both the magnitude and the direction of the vector  $\mathbf{T}$  vary. For example, we obtain longitudinal oscillations of the toroidal moment when we vary the current density  $\mathbf{j}(\mathbf{r}, t)$  with the position in space and the geometrical dimensions of the coil fixed (Fig. 1a). A rigid coil oscillating about its equilibrium position and carrying a current of density  $\mathbf{j}(\mathbf{r}, t)$  fixed in magnitude produces a geometrical image of transverse toroidal oscillations, which in this case take the form of precessions (Fig. 1b).

The toroidal oscillations can, within the framework of the microscopic model,<sup>1</sup> be related to the amplitude and phase oscillations of the order parameter  $\hat{\Delta}(t)$  describing the electron-hole pairing (for more details, see §2). It is essential to emphasize that here we are dealing with the case of “soft” systems, when the form of the wave function of the electron-hole pair can change. This “softness” gives rise, in particular, to the mix-up of toroidal and ordinary polar oscillations, and to the absence of a small relativistic parameter in the Hamiltonian for the interaction between the toroidal oscillation and light.<sup>4,6</sup>

In the present paper we consider the simplest case, when here is only one branch of toroidal oscillations (i.e., when the vector  $\mathbf{T}$  transforms according to a one-dimensional irreducible representation). There can, in principle, be more complicated toroidal structures (two- and three-dimensional representations), but the absence of well-developed microscopic models makes the investigation of these systems difficult.

The paper is organized as follows. In §2 we classify the oscillations in systems with electron-hole pairing according

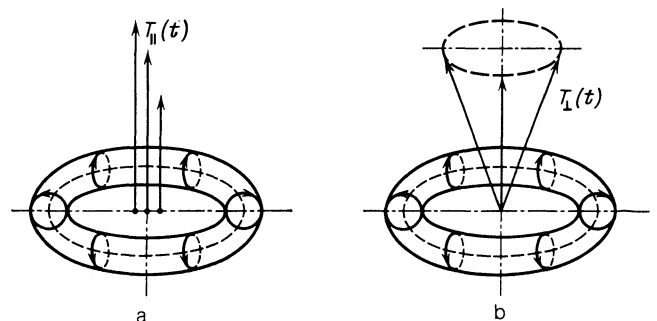


FIG. 1.

to the order-parameter symmetry class and the pair wave function structure. In §3 we find the natural frequencies and the logarithmic decrements of the toroidal excitations in ferroelectric and orbital toroidal magnetic materials at zero temperature. In §4 we find the contribution of the toroidal excitations to the dynamic dielectric susceptibility  $\chi(\omega)$  at zero temperature. In §5 we consider the characteristics of the toroidal order and of the collective excitations in band antiferromagnets with spin-density waves. In the Conclusion (§6) we formulate some problems that need to be investigated further.

## §2. CLASSIFICATION OF THE COLLECTIVE EXCITATIONS IN SYSTEMS WITH ELECTRON-HOLE PAIRING

It is well known<sup>7</sup> that, in models of the "exciton dielectric" type, the system can have four types of ground state, which are characterized respectively by the four order parameters  $\Delta_{Re}^s$ ,  $\Delta_{Im}^s$ ,  $\Delta_{Re}^i$ , and  $\Delta_{Im}^i$  and the corresponding effective interaction constants  $g_{Re}^s$ ,  $g_{Im}^s$ ,  $g_{Re}^i$ , and  $g_{Im}^i$ . Each of these states exhibits one of the possible structures of the wave function of the electron-hole pair in phase and spin space. Thus, for example, the state with a real singlet order parameter  $\Delta_{Re}^s$  corresponds to zero phase difference between the electron and hole wave functions, i.e., to  $\Delta\varphi = 0$ , while the state with an imaginary singlet order parameter  $\Delta_{Im}^s$  corresponds to a phase difference  $\Delta\varphi = \pi/2$  between these wave functions. The phase relations are similar for the triple pair structures with  $\Delta_{Re}^i$  and  $\Delta_{Im}^i$ . In its general form the order parameter  $\hat{\Delta}$  has the form of a spinor:

$$\hat{\Delta}_{12} = \hat{\Delta}_{21}^* = (\Delta_{Re}^s + i\Delta_{Im}^s) \hat{I} + (\Delta_{Re}^i + i\Delta_{Im}^i) \hat{\sigma}, \quad (1)$$

where  $\hat{I}$  is the unit matrix,  $\hat{\sigma}$  is a vector composed of the Pauli matrices, and 1 and 2 are the band indices.

It is, however, not only the phase and spin structures of the pair wave function that are by themselves important for the determination of the macroscopic properties of the system, but also the specific crystal symmetry reflected in the electron- and hole-dispersion laws and in the matrix elements of the interelectron interaction. Thus, for example, when the interband matrix elements  $P_{12}$  of the momentum is nonzero, the state with  $\Delta_{Re}^s \neq 0$  possesses ferroelectric properties,<sup>8</sup> while the state with  $\Delta_{Im}^s \neq 0$  is a toroidal state.<sup>1</sup> At the same time when the interband matrix element  $L_{12}$  of the orbital angular momentum is nonzero, the state with  $\Delta_{Im}^s \neq 0$  corresponds to orbital ferromagnetism,<sup>9</sup> while the one with  $\Delta_{Re}^s \neq 0$  exhibit the so-called orientational state.<sup>10</sup> The triplet pair structure also can exhibit different types of ordering, depending on the symmetry properties of the system (see, for example, Ref. 10).

The Hamiltonian of the two-band model, within the framework of which the investigation of the collective excitations in systems with electron-hole pairing will be carried out, has the form<sup>1)</sup>

$$\hat{H} = \hat{H}_1 + \hat{H}_2 + \frac{(\Delta_{Re}^s)^2}{g_{Re}^s} + \frac{(\Delta_{Im}^s)^2}{g_{Im}^s} + \frac{(\Delta_{Re}^i)^2}{g_{Re}^i} + \frac{(\Delta_{Im}^i)^2}{g_{Im}^i}. \quad (2)$$

Here

$$\hat{H}_1 = \begin{pmatrix} \frac{1}{2m_1} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)^2 - \varepsilon_F + e\Phi & \frac{i\mathbf{P}}{m} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) - \hat{\Delta}_{12} \\ -\frac{i\mathbf{P}}{m} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) - \hat{\Delta}_{21} & -\frac{1}{2m_2} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)^2 + \varepsilon_F + e\Phi \end{pmatrix} \quad (3)$$

where  $m_1$  and  $m_2$  are the effective electron and hole masses [the signs in (3) have been chosen such that  $m_1, m_2 > 0$ ];  $m$  is the free-electron mass;  $\varepsilon_F$  is the Fermi energy (for a semiconductor  $\varepsilon_F > 0$  and for a metal  $\varepsilon_F = -E_g/2 < 0$ );  $\mathbf{P}_{12} = \mathbf{P}_{21}^* = i\mathbf{P}$ ; and  $\mathbf{A}$  and  $\Phi$  are the vector and scalar potentials of the electromagnetic field. In the computations carried out below we assume, for simplicity, that  $m_1 = m_2 = m^*$ , and also that  $\gamma_0 = |(P/m)k_F| \ll \Delta$ , where  $\Delta$  is the characteristic value of the order parameter in the ground state. Further,

$$\hat{H}_2 = \begin{pmatrix} 0 & \left[ \lambda_{12} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) \right] \\ \left[ \lambda_{21} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) \right] & 0 \end{pmatrix} \hat{\sigma}, \quad (4)$$

where

$$\lambda_{12} = \lambda_{21}^* = \lambda = \langle \varphi_1 | \text{grad } V | \varphi_2 \rangle,$$

$V(\mathbf{r})$  is the crystal potential, and the  $\varphi_{1,2}$  are the Bloch wave functions.

In the present paper we shall treat the interaction con-

stants  $\Delta_{Re}^{s,i}$  and  $g_{Im}^{s,i}$  as model parameters whose magnitudes determine the type of ground state the system has. Similar investigations of the conditions under which a particular ground state is realized are carried out in Refs. 1 and 11-14. The situations most easily achieved are the ones with  $\Delta_{Re}^{Os} \neq 0$  and  $\Delta_{Re}^{Oe} \neq 0$ , whereas the conditions for the appearance of the states with  $\Delta_{Im}^{Os} \neq 0$  and  $\Delta_{Im}^{Oe} \neq 0$  are rather rigid.

The collective excitations in two-band systems with electron-hole pairing can be classified on the basis of (1) according to the microscopic pair structure. There are four possible types of structure: charge- and spin-density waves (CDW and SDW) and electric-current- and spin-current-density waves (ECDW and SCDW). On the other hand, to study the macroscopic properties of the system we need to classify the oscillations according to the type of space-time symmetry connected with these physical-quantity oscillations. In Table I we present such a classification using the following notation:  $\mathbf{P}$  denotes the dipole moment;  $\mathbf{T}$ , the orbital toroidal moment;  $\mathbf{M}$ , the magnetic moment; and  $\mathbf{G}$ , the orientational moment. The indices  $\parallel$  and  $\perp$  indicate the ori-

TABLE I.

	Microstructure of pair	Symmetry type	
		$P_{12} \neq 0$	$L_{12} \neq 0$
$\Delta_{\text{Re}}^s$	CDW	$P(\parallel)$	$G(\parallel)$
$\Delta_{\text{Im}}^s$	ECDW	$T(\parallel)$	$M(\parallel)$
$\Delta_{\text{Re}}^t$	SDW	$T(\perp)$	$M(\perp)$
$\Delta_{\text{Im}}^t$	SCDW	$P(\perp)$	$G(\perp)$

entations of the vectors  $\mathbf{P}$ ,  $\mathbf{T}$ ,  $\mathbf{M}$ , and  $\mathbf{G}$  relative to  $\mathbf{P}_{12}$  or  $\mathbf{L}_{12}$ . Let us make a few comments by way of explanation of the table.

In uniaxial systems with  $\mathbf{P}_{12} \neq 0$  the correspondence between the type of microscopic structure possessed by the electron-hole pair and the type of symmetry possessed by the macroscopic physical vector quantity connected with this structure is specified as follows:

$$\begin{aligned} \mathbf{P}(\parallel) &\leftrightarrow n\Delta_{\text{Re}}^s, \\ \mathbf{T}(\parallel) &\leftrightarrow n\Delta_{\text{Im}}^s, \\ \mathbf{P}(\perp) &\leftrightarrow [n\Delta_{\text{Im}}^t], \\ \mathbf{T}(\perp) &\leftrightarrow [n\Delta_{\text{Re}}^t], \end{aligned} \quad (5)$$

where  $\mathbf{n}$  is the unit vector in the direction of the principal axis ( $\mathbf{n} \parallel \mathbf{P}_{12}$ ). The vectors  $\mathbf{P}(\parallel, \perp)$  and  $\mathbf{T}(\parallel, \perp)$  describe the oscillations of the polarization and the toroidal moment along the principal axis  $\mathbf{n}$  or in the plane perpendicular to  $\mathbf{n}$ . In this case the relations connecting  $\mathbf{P}(\perp)$  with  $\Delta_{\text{Im}}^t$  and  $\mathbf{T}(\perp)$  with  $\Delta_{\text{Re}}^t$  contain a small relativistic parameter proportional to the spin-orbit interaction (see Ref. 10 and §5 of the present paper), while for  $\mathbf{P}(\parallel)$  and  $\mathbf{T}(\parallel)$  the relations with  $\Delta_{\text{Re}}^s$  and  $\Delta_{\text{Im}}^s$  do not contain such a small parameter (see Refs. 1 and 8).

The genesis of a particular type of oscillations depends essentially on the structure of the ground state of the system. For example, in the case of the singlet ferroelectric ground state ( $\Delta_{\text{Re}}^{\text{Os}} \neq 0$ )<sup>8</sup> the  $\mathbf{P}(\parallel)$  polar oscillations are largely of an amplitude nature (the natural frequency of this oscillation  $\omega_{\text{Re}}^s \approx 2\Delta_{\text{Re}}^{\text{Os}}$  in the low-temperature region  $\theta \ll \Delta_{\text{Re}}^{\text{Os}}$ ). The remaining [i.e., the  $\mathbf{T}(\parallel, \perp)$  and  $\mathbf{P}(\perp)$ ] oscillations are of a phase (in the case of the triplet oscillations we mean the phase in spin space) and, generally speaking, display a gap.

For  $\Theta \ll \Delta_{\text{Re}}^{\text{Os}}$  the corresponding frequencies

$$\omega_{\text{Im}(\text{Re}, \text{Im})}^{s(t)} \approx 2\eta_{\text{Im}(\text{Re}, \text{Im})}^{s(t)} \Delta_{\text{Re}}^{\text{Os}},$$

where the  $\eta$  coefficients are of the order of the corresponding differences between the effective interaction constants  $g_{\text{Re}}^s$  and  $\omega_{\text{Im}(\text{Re}, \text{Im})}^{s(t)}$  (Refs. 15–17). In principle, if the differences between the constants are small, then we can have  $\omega_{\text{Im}(\text{Re}, \text{Im})}^{s(t)} \ll \omega_{\text{Re}}^s$ .

If the system has a single toroidal ground state ( $\Delta_{\text{Im}}^{\text{Os}} \neq 0$ ), then the  $\mathbf{T}(\parallel)$  toroidal oscillations are largely of an amplitude nature, while the  $\mathbf{P}(\parallel)$ ,  $\mathbf{P}(\perp)$ , and  $\mathbf{T}(\perp)$  oscillations concerning the natural frequencies of the oscillations

hold good here when we make the appropriate change of designation  $\text{Re} \leftrightarrow \text{Im}$ .

The situation in the case of the  $\Delta_{\text{Re}}^{\text{Or}} \neq 0$  (or  $\Delta_{\text{Im}}^{\text{Or}} \neq 0$ ) triplet ground state is peculiar. Besides the gap, phase, and amplitude modes already discussed above, there occurs a gapless (Goldstone) mode corresponding to SDW (or SCDW) oscillations with orientation  $\Delta_{\text{Re}}^t \perp \Delta_{\text{Re}}^{\text{Or}}$  (or  $\Delta_{\text{Im}}^t \perp \Delta_{\text{Im}}^{\text{Or}}$ ) (see, for example, Ref. 18).

We thus can have, depending on the type of ground state, quite different toroidal oscillations in crystals (relatively high-frequency oscillations in orbital toroidal magnetic materials at temperatures far from the transition point, low-frequency oscillations in electronic ferroelectrics, gapless oscillations in itinerant antiferromagnets in which the magnetic anisotropy is ignored).

Similarly, we can classify the collective excitations in systems with  $\mathbf{L}_{12} \neq 0$ . Since such systems are not considered in the present paper, further comments on the table are unnecessary.

### §3. THE NATURAL FREQUENCIES AND LOGARITHMIC DAMPING RATES OF THE COLLECTIVE EXCITATIONS IN ELECTRONIC-FERROELECTRIC AND ORBITAL-TOROIDAL-MAGNETIC MATERIALS

Let us consider the transverse, optically active collective oscillations that occur in a system with the Hamiltonian (2) at zero temperatures as a result of the fluctuations of the singlet order parameters  $\Delta_{\text{Re}}^s$  and  $\Delta_{\text{Im}}^s$  [when the spin-orbit interaction (4) is ignored, the triplet branches split off, and do not contribute to the optical properties of the system; here we shall not be interested in them, and shall set  $\lambda \equiv 0$ ]. Let the ground state of the system be toroidal, i.e., let  $\Delta_{\text{Im}}^{\text{Os}} \neq 0$ , in the absence of external fields. An alternating electromagnetic field  $\mathbf{A}(\mathbf{r}, t)$  induces a nonequilibrium correction  $\delta\Delta_{12}^s(\mathbf{r}, t)$  to the order parameter:

$$\delta\Delta_{12}^s(\mathbf{r}, t) = \delta\Delta_{\text{Re}}^s(\mathbf{r}, t) + i\delta\Delta_{\text{Im}}^s(\mathbf{r}, t). \quad (6)$$

Let us consider the problem under conditions when we can neglect the spatial dispersion and the retardation (the latter stipulation allows us to ignore the polariton effects). In the approximation linear in the vector potential  $\mathbf{A}(\mathbf{r}, t)$  the Green's function of the system acquires the correction.

$$\hat{G} - \hat{G}^0 = \delta\hat{G}(x, x') = \hat{G}^0(x, x') [-\delta\hat{\Delta}(x'') + \hat{H}_A] \hat{G}^0(x'', x'), \quad (7)$$

$$x = (\mathbf{r}, t),$$

where  $\hat{G}^0$  and  $\hat{H}_A$  are  $2 \times 2$  matrices with indices corresponding to the band indices:

$$\hat{G}^0 = \begin{pmatrix} G_{11}^0 & G_{12}^0 \\ G_{21}^0 & G_{22}^0 \end{pmatrix}, \quad (8)$$

$$\hat{H}_A = \begin{pmatrix} -\frac{e}{cm^*} \mathbf{A} \frac{\nabla}{i} & -i\mathbf{P} \frac{e}{mc} \mathbf{A} \\ i\mathbf{P} \frac{e}{mc} \mathbf{A} & \frac{e}{cm^*} \mathbf{A} \frac{\nabla}{i} \end{pmatrix}. \quad (9)$$

The explicit form of the Green's functions  $G_{ij}^0$  is known (see, for example, Ref. 7), and we shall not give it here. After substituting (7) into the self-consistency equations

$$\Delta_{\text{Re}}^s = g_{\text{Re}}^s \text{Re } G_{12}(x, x), \quad (10)$$

$$\Delta_{\text{Im}}^s = g_{\text{Im}}^s \text{Im } G_{12}(x, x), \quad (11)$$

we obtain a system of equations for the corrections  $\delta\Delta$ . The formalism for correctly introducing the vector potential  $\mathbf{A}(x)$  into the self-consistency equations, i.e., one that guarantees gauge invariance, is set forth in Ref. 1; therefore, we omit the intermediate expressions. For the Fourier transforms  $\delta\Delta_{\text{Re,Im}}^s(\omega)$  and  $\mathbf{A}(\omega)$  we have, when no allowance is made for the spatial dispersion (see the Appendix), the equations

$$\begin{aligned} \bar{\Pi}_{\text{Re}}(\omega) \delta\Delta_{\text{Re}}^s(\omega) - i\omega\tilde{L}(\omega) \delta\Delta_{\text{Im}}^s(\omega) &= (i\omega/c) \tilde{\Gamma}_{\text{Re}}(\omega) \mathbf{A}(\omega), \\ i\omega\tilde{L}(\omega) \delta\Delta_{\text{Re}}^s(\omega) + \bar{\Pi}_{\text{Im}}(\omega) \delta\Delta_{\text{Im}}^s(\omega) &= (\omega^2/c) \tilde{\Gamma}_{\text{Im}}(\omega) \mathbf{A}(\omega), \end{aligned} \quad (12)$$

where

$$\bar{\Pi}_{\text{Re}}(\omega) = \frac{1}{g_{\text{Re}}^s} - \frac{1}{g_{\text{Im}}^s} + \sum_{\mathbf{k}} \left\{ \frac{\omega^2}{2E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)} - \frac{\gamma_{\mathbf{k}}}{2\Delta E_{\mathbf{k}}} \right\}, \quad (13)$$

$$\bar{\Pi}_{\text{Im}}(\omega) = \sum_{\mathbf{k}} \left\{ \frac{\omega^2 - 4(\gamma_{\mathbf{k}} - \Delta)^2}{2E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)} - \frac{\gamma_{\mathbf{k}}}{2\Delta E_{\mathbf{k}}} \right\}, \quad (14)$$

$$\tilde{L}(\omega) = \sum_{\mathbf{k}} \frac{\zeta_{\mathbf{k}}}{E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (15)$$

$$\tilde{\Gamma}_{\text{Re}}(\omega) = e \sum_{\mathbf{k}} \frac{-(\gamma_{\mathbf{k}} - \Delta)(\mathbf{k}/m^*) + \zeta_{\mathbf{k}}(\mathbf{P}/m)}{E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (16)$$

$$\tilde{\Gamma}_{\text{Im}}(\omega) = e \sum_{\mathbf{k}} \frac{(\gamma_{\mathbf{k}} - \Delta)\zeta_{\mathbf{k}}(\mathbf{k}/m^*) - \zeta_{\mathbf{k}}^2(\mathbf{P}/m)}{2E_{\mathbf{k}}^3(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (17)$$

$$E_{\mathbf{k}} = [\zeta_{\mathbf{k}}^2 + (\gamma_{\mathbf{k}} - \Delta)^2]^{1/2}, \quad \Delta = \Delta_{\text{Im}}^{0s}, \quad \gamma_{\mathbf{k}} = (P/m)k. \quad (18)$$

Notice that mixing of  $\delta\Delta_{\text{Re}}^s$  and  $\delta\Delta_{\text{Im}}^s$  occurs once the coefficient  $\tilde{L}(\omega)$  is nonzero, and this happens only when allowance is made for the weak energy dependence of the density of states in the vicinity of the Fermi surface:  $N(\zeta_{\mathbf{k}}) \approx \bar{N}(0)(1 + \zeta_{\mathbf{k}}/2\varepsilon_F)$ . Generally speaking, this result is peculiar to the semimetallic model; in the case of the semiconductor model  $\tilde{L}(\omega)$  does not contain any smallness parameter.<sup>19</sup>

The natural frequencies of the transverse oscillations can, when the retardation is ignored, be found from (12) with  $\mathbf{A}$  set equal to zero. To lowest order in  $\Delta/\varepsilon_F \ll 1$  we can ignore the mixing of the amplitude and phase branches. The natural frequency of the phase branch corresponding to the  $\delta\Delta_{\text{Re}}^s$  oscillations in the case of weak hybridization, i.e., in the case when  $\gamma_0^2 \ll \Delta^2$ , has the form

$$\omega_{\text{Re}}^s = 2\Delta \left[ \frac{1}{\bar{g}_{\text{Re}}^s} - \frac{1}{\bar{g}_{\text{Im}}^s} - \frac{1}{3} \frac{\gamma_0^2}{\Delta^2} \right]^{1/2}, \quad (19)$$

while for the amplitude branch corresponding to the  $\delta\Delta_{\text{Im}}^s$  oscillations we have

$$\omega_{\text{Im}}^s = 2\Delta(1 + i\gamma_0/\sqrt{3}\Delta), \quad (20)$$

$$\bar{g}_{\text{Re,Im}}^s = g_{\text{Re,Im}}^s \bar{N}(0). \quad (21)$$

In deriving (19) and (20), we assumed that

$$(\bar{g}_{\text{Re}}^s)^{-1} > (\bar{g}_{\text{Im}}^s)^{-1} + \gamma_0^2/3\Delta^2.$$

For the ferroelectric ground state, i.e., for  $\Delta_{\text{Re}}^{0s} \neq 0$ , the system of equations for  $\delta\Delta_{\text{Re,Im}}^s$  is derived in Ref. 20 [see the formulas (23) and (25)], and has a form similar to (12), but with different coefficients:

$$\Pi_{\text{Re}}(\omega) = \sum_{\mathbf{k}} \frac{\omega^2 - 4\Delta^2}{2E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (22)$$

$$\Pi_{\text{Im}}(\omega) = \frac{1}{g_{\text{Im}}^s} - \frac{1}{g_{\text{Re}}^s} + \sum_{\mathbf{k}} \frac{\omega^2 - 4\gamma_{\mathbf{k}}^2}{2E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (23)$$

$$L(\omega) = \sum_{\mathbf{k}} \frac{\zeta_{\mathbf{k}}}{E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (24)$$

$$\Gamma_{\text{Re}}(\omega) = e \sum_{\mathbf{k}} \frac{-\gamma_{\mathbf{k}}(\mathbf{k}/m^*) + \zeta_{\mathbf{k}}(\mathbf{P}/m)}{E_{\mathbf{k}}(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (25)$$

$$\Gamma_{\text{Im}}(\omega) = e \sum_{\mathbf{k}} \frac{\zeta_{\mathbf{k}}\gamma_{\mathbf{k}}(\mathbf{k}/m^*) - (E_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2)(\mathbf{P}/m)}{2E_{\mathbf{k}}^3(\omega^2 - 4E_{\mathbf{k}}^2)}, \quad (26)$$

$$E_{\mathbf{k}} = (\zeta_{\mathbf{k}}^2 + \Delta^2 + \gamma_{\mathbf{k}}^2)^{1/2}, \quad \Delta = \Delta_{\text{Re}}^{0s}. \quad (27)$$

In contrast to Ref. 20, here  $\Gamma_{\text{Im}}$  is not neglected, since it is the source of the toroidal oscillations of interest to us (for the purposes of Ref. 20 the retention of the source  $\Gamma_{\text{Im}}$  was unessential).

The expressions for the natural frequencies for the case of the ferroelectric ground state have the same form as those found in Ref. 20 when  $g_{\text{Im}}^s$  is set identically equal to  $g_{\text{Re}}^s$ :

$$\omega_{\text{Im}}^s = 2\Delta \left[ \frac{1}{\bar{g}_{\text{Im}}^s} - \frac{1}{\bar{g}_{\text{Re}}^s} + \frac{1}{3} \frac{\gamma_0^2}{\Delta^2} \right]^{1/2}, \quad (28)$$

$$\omega_{\text{Re}}^s = 2\Delta \left[ 1 + \frac{2\gamma_0^2}{\Delta^2} \left( \frac{\alpha}{\ln \alpha} \right)^2 \left( 1 + \frac{i\pi}{\ln \alpha} \right) \right], \quad (29)$$

$$\alpha = \Delta(2\pi\bar{g}_{\text{Re}}^s\varepsilon_F)^{-1}. \quad (30)$$

Notice the significant difference in the attenuation of the amplitude modes (19) and (20) (the small parameter  $\alpha$  does not occur in the last case). The cause of this lies in the violation of the parity of the single-electron spectrum in the toroidal state, and is evident from Figs. 2a and 2b. In the case of a ferroelectric crystal

$$E_{\pm}(\mathbf{k}) = (\zeta_{\mathbf{k}}^2 + \gamma_{\mathbf{k}}^2 + \Delta^2)^{1/2},$$

and the amplitude-excitation energy which satisfies  $\omega_{\text{Re}}^s(\mathbf{q}) \approx 2\Delta$  at low  $q$  values, lies inside the gap (touches in

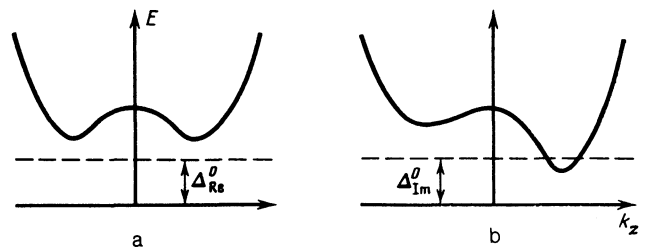


FIG. 2.

the case when  $\mathbf{k} \perp \mathbf{P}$ ). In the toroidal state

$$E_{\pm}(\mathbf{k}) = \pm [\zeta_{\mathbf{k}}^2 + (\gamma_{\mathbf{k}} - \Delta)^2]^{1/2},$$

and in a broad region of energies  $E(\mathbf{k})$  the frequency  $\omega_{\text{im}}^s(\mathbf{q})$  lies within the single-electron spectrum.

Evidently, the conclusion that the toroidal oscillations in toroidal magnetic materials are strongly damped is quite general. Therefore, their detection in infrared optical experiments may present some difficulty. At the same time toroidal oscillations of the type (28) occurring in ferroelectric materials and lying inside the dielectric gap are weakly damped. Such oscillations make a specific contribution to the spectral dependence of the dielectric susceptibility  $\chi(\omega)$  and the optical-absorption coefficient<sup>6</sup>  $K_T(\omega)$ .

#### §4. CONTRIBUTION OF THE TOROIDAL EXCITATIONS TO THE DYNAMIC DIELECTRIC SUSCEPTIBILITY OF ORBITAL-TOROIDAL-MAGNETIC AND ELECTRONIC-FERROELECTRIC MATERIALS

It follows from the equations (12) that an alternating electric field induces order parameters  $\delta\Delta_{\text{Re}}^s$  and  $\delta\Delta_{\text{Im}}^s$ , and that near the frequencies  $\omega \approx \omega_{\text{Re, Im}}^s$  the field interacts resonantly with the collective excitations. Let us consider the contribution of the toroidal excitations to the dynamic dielectric susceptibility  $\chi(\omega)$ ; evidently, it is most important in the region  $\omega \approx \omega_{\text{im}}^s$ . Neglecting the spin-orbit interaction, we define the orbital-current operator  $\hat{\mathbf{j}}$  in the usual manner:

$$\hat{\mathbf{j}} = -c \frac{\delta \hat{H}_1}{\delta \mathbf{A}} = \begin{pmatrix} \frac{e}{m^*} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) & i \frac{e}{m} \mathbf{P} \\ -i \frac{e}{m} \mathbf{P} & -\frac{e}{m^*} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right) \end{pmatrix}. \quad (31)$$

The mean value of the current density is given by the relation

$$\langle \hat{\mathbf{j}} \rangle = \text{Sp } \hat{\mathbf{j}} \hat{G}, \quad (32)$$

where the total Green's function  $\hat{G}$  includes both the direct contribution of the electromagnetic field  $\mathbf{A}$  and the contribution due to the change that occurs in the order parameter in the field. We must, in computing the current (32), make sure that gauge invariance is maintained at all stages of the calculation, so as to preclude the appearance of unphysical contributions to  $\langle \hat{\mathbf{j}} \rangle$  (this question is discussed in detail in Ref. 1).

In the case of the ferroelectric ground state we have the formula (24) from Ref. 20:

$$\mathbf{j}(\omega) = \mathbf{j}_{\text{im}}^{\Delta}(\omega) + \mathbf{j}_{\text{re}}^{\Delta}(\omega) + \mathbf{j}^{\Delta}(\omega), \quad (33)$$

$$\begin{aligned} \mathbf{j}_{\text{im}}^{\Delta}(\omega) &= 2\tilde{\Gamma}_{\text{im}}(\omega) \omega^2 \delta\Delta_{\text{im}}^s(\omega), \\ \mathbf{j}_{\text{re}}^{\Delta}(\omega) &= -2i\omega \tilde{\Gamma}_{\text{re}}(\omega) \delta\Delta_{\text{re}}^s(\omega), \end{aligned} \quad (34)$$

$$\begin{aligned} \mathbf{j}^{\Delta}(\omega) &= -\frac{e^2}{c} \omega^2 \sum_{\mathbf{k}} \left\{ \frac{\mathbf{k}(\mathbf{A}\mathbf{k})}{(m^*)^2} (\Delta^2 + \gamma_{\mathbf{k}}^2) \right. \\ &\quad \left. + \frac{\mathbf{P}}{m} \left( \frac{\mathbf{P}\mathbf{A}}{m} \right) (\zeta_{\mathbf{k}}^2 + \Delta^2) \right\} [E_{\mathbf{k}}^s(\omega^2 - 4E_{\mathbf{k}}^2)]^{-1}. \end{aligned}$$

$$-\frac{\mathbf{k}\gamma_{\mathbf{k}}(\mathbf{P}\mathbf{A})\zeta_{\mathbf{k}}}{mm^*} - \frac{\mathbf{P}\gamma_{\mathbf{k}}(\mathbf{A}\mathbf{k})\zeta_{\mathbf{k}}}{mm^*} \left\{ [E_{\mathbf{k}}^s(\omega^2 - 4E_{\mathbf{k}}^2)]^{-1} \right\}. \quad (35)$$

Here  $\mathbf{j}_{\text{im}}^{\Delta}$  and  $\mathbf{j}_{\text{re}}^{\Delta}$  are respectively the contributions of the toroidal and polar excitations and  $\mathbf{j}^{\Delta}$  is the usual single-particle contribution to the current for a fixed order-parameter structure.

For a toroidal ground state ( $\Delta = \Delta_{\text{im}}^{\text{Os}}$ ) the structure of the total current  $\mathbf{j}(\omega)$  is similar to (33), but now

$$\mathbf{j}_{\text{im}}^{\Delta}(\omega) = 2\tilde{\Gamma}_{\text{im}}(\omega) \omega^2 \delta\Delta_{\text{im}}^s(\omega), \quad (36)$$

$$\mathbf{j}_{\text{re}}^{\Delta}(\omega) = -2i\omega \tilde{\Gamma}_{\text{re}}(\omega) \delta\Delta_{\text{re}}^s(\omega),$$

$$\begin{aligned} \mathbf{j}^{\Delta}(\omega) &= -\frac{e^2}{c} \omega^2 \sum_{\mathbf{k}} \left\{ \frac{\mathbf{k}(\mathbf{A}\mathbf{k})}{(m^*)^2} (\gamma_{\mathbf{k}} - \Delta)^2 + \frac{\mathbf{P}}{m} \left( \frac{\mathbf{P}\mathbf{A}}{m} \right) \zeta_{\mathbf{k}}^2 \right. \\ &\quad \left. - \frac{\mathbf{P}(\mathbf{A}\mathbf{k})\zeta_{\mathbf{k}}}{mm^*} (\gamma_{\mathbf{k}} - \Delta) \right. \\ &\quad \left. - \frac{\mathbf{k}(\mathbf{P}\mathbf{A})\zeta_{\mathbf{k}}}{mm^*} (\gamma_{\mathbf{k}} - \Delta) \right\} [E_{\mathbf{k}}^s(\omega^2 - 4E_{\mathbf{k}}^2)]^{-1}. \end{aligned} \quad (37)$$

Let us draw particular attention to the expressions (34) and (36) for the current  $\mathbf{j}_{\text{re, im}}^{\Delta}(\omega)$  due to the collective modes. Since the total current  $\mathbf{j}$  enters into the Lagrangian of the system in the combination  $\mathbf{j} \cdot \mathbf{A}$ , the form (34) and (36) uniquely determines the structure of the sources in the equations (12) for the dynamical corrections  $\delta\Delta_{\text{Re, Im}}^s$ , since these equations are themselves obtained through by varying the Lagrangian  $\mathcal{L}$  in  $\delta\Delta_{\text{Re, Im}}^s$ .

In the low-frequency region  $\omega \ll 2\Delta$ , we can obtain the following effective Lagrangian describing the low-frequency excitations in a toroidal magnetic material:

$$\mathcal{L} = \mathcal{H} - U, \quad (38)$$

$$\mathcal{H} = \frac{1}{2M_{\text{Re}}} (\delta\dot{\Delta}_{\text{Re}}^s)^2 + \frac{1}{2M_{\text{Im}}} (\delta\dot{\Delta}_{\text{Im}}^s)^2, \quad (39)$$

$$\frac{1}{2M_{\text{Re}}} = \frac{1}{4\Delta^2}, \quad \frac{1}{2M_{\text{Im}}} = \bar{N}(0) \frac{1}{12\Delta^2}, \quad (40)$$

$$\begin{aligned} U &= \bar{\Pi}_{\text{Re}}(0) (\delta\Delta_{\text{Re}}^s)^2 + \bar{\Pi}_{\text{Im}}(0) (\delta\Delta_{\text{Im}}^s)^2 - L(0) [\delta\Delta_{\text{Re}}^s \delta\dot{\Delta}_{\text{Im}}^s \\ &\quad - \delta\Delta_{\text{Im}}^s \delta\dot{\Delta}_{\text{Re}}^s] - \frac{2}{c} \tilde{\Gamma}_{\text{Re}}(0) \delta\Delta_{\text{Re}}^s \dot{\mathbf{A}} + \frac{2}{c} \tilde{\Gamma}_{\text{Im}}(0) \delta\Delta_{\text{Im}}^s \ddot{\mathbf{A}}. \end{aligned} \quad (41)$$

The term  $(\mathbf{j}_{\text{im}}^{\Delta} + \mathbf{j}_{\text{re}}^{\Delta}) \cdot \mathbf{A}$  is obtained from (41) in its explicit form by adding the total time derivative. The Lagrangian for a ferroelectric has a similar form in the frequency region  $\omega \ll 2\Delta$ .

The dielectric susceptibility  $\chi_{ij}$  can be found from the expressions (33) and (36) with allowance for the coupling of the current  $\mathbf{j}$  to the polarization  $\mathbf{P}$ , but without allowance for the spatial dispersion:

$$j_i = \dot{P}_i, \quad i = x, y, z. \quad (42)$$

We are interested only in the contribution of the collective excitations and, in particular, that of the toroidal collective excitations. It is clear from the structure of the sources in (12) that the contribution will be only to the component  $\chi_{zz}^{\Delta}$ . For a toroidal magnetic crystal we have

$$\begin{aligned}\chi_{zz}^{\Delta} &= \chi_{\text{Im}}^{\Delta} + \chi_{\text{Re}}^{\Delta}, \\ \chi_{\text{Re}}^{\Delta}(\omega) &= 2 \frac{\Gamma_{\text{Re}}^2 \bar{\Pi}_{\text{Im}} + \bar{L} \omega^2 \Gamma_{\text{Re}} \Gamma_{\text{Im}}}{\bar{\Pi}_{\text{Re}} \bar{\Pi}_{\text{Im}} - \bar{L}^2 \omega^2}, \\ \chi_{\text{Im}}^{\Delta}(\omega) &= 2 \omega^2 \frac{\Gamma_{\text{Im}}^2 \bar{\Pi}_{\text{Re}} + \bar{L} \Gamma_{\text{Re}} \Gamma_{\text{Im}}}{\bar{\Pi}_{\text{Re}} \bar{\Pi}_{\text{Im}} - \bar{L}^2 \omega^2}.\end{aligned}\quad (43)$$

In the frequency region  $\omega \approx 2\Delta$  the contribution of the toroidal excitations is resonant when  $\omega \approx \omega_{\text{Im}}^s$ , and has the form

$$\begin{aligned}\chi_{\text{Im}}^{\Delta}(\omega) &\approx \bar{A}_{\text{Im}} \left(1 - \frac{\omega^2}{4\Delta^2}\right)^{-1/2}, \\ \bar{A}_{\text{Im}} &= \frac{1}{24} \left(\frac{eP}{3m}\right)^2 \frac{\pi}{\Delta^2} \bar{N}(0), \quad 1 \gg 1 - \frac{\omega^2}{4\Delta^2} \gg 2 \frac{\gamma_0}{\Delta}.\end{aligned}\quad (44)$$

In the low-frequency region  $\omega \ll 2\Delta$  the polar excitations make the dominant contribution:

$$\chi_{\text{Re}}^{\Delta}(\omega) \approx \bar{A}_{\text{Re}} [\omega^2 - (\omega_{\text{Re}}^s)^2]^{-1}, \quad \bar{A}_{\text{Re}} = -8 \left(\frac{eP}{3m}\right)^2 \frac{\epsilon_F^2}{\Delta^2} \bar{N}(0).\quad (45)$$

Let us, in the ferroelectric case, take account of the contribution of the toroidal excitations in the low-frequency region  $\omega \approx \omega_{\text{Im}}^s \ll 2\Delta$ , where it has a resonance form:

$$\chi_{\text{Im}}^{\Delta}(\omega) \approx \frac{\omega^2 A_{\text{Im}}}{\omega^2 - (\omega_{\text{Im}}^s)^2}, \quad A_{\text{Im}} = \left(\frac{eP}{3m}\right)^2 \frac{\bar{N}(0)}{\Delta^2 \bar{g}_{\text{Re}}^s}.\quad (46)$$

The contribution of the polar excitations, which was computed earlier in Ref. 20, is greater than (46) by a factor of  $(\bar{g}_{\text{Re}}^s)^{-1} \gg 1$ . But its existence and the unusual frequency dependence, which corroborate at the microscopic level the phenomenological results obtained in Refs. 4 and 6, are what is of importance to us. Notice that, in the static limit,  $\chi_{\text{Re}}^{\Delta} \neq 0$ , whereas  $\chi_{\text{Im}}^{\Delta} = 0$ . The formulas (43) also allow us to analyze those optical properties of toroidal magnetic and ferroelectric materials which are connected with the excitation of toroidal oscillations in the region of relatively high frequencies, where the effective-Lagrangian scheme proposed in Ref. 4 does not work.

## §5. THE TOROIDAL MOMENTS IN ANTIFERROMAGNETS WITH SPIN DENSITY WAVES

Thus far we have been discussing only the singlet ground states and the collective excitations. In the absence of spin-orbit interaction the triplet excitations do not get mixed up with the singlet excitations, and do not make any contribution to the dielectric susceptibility  $\chi(\omega)$ .

Let us now consider itinerant-electron antiferromagnets with spin density waves (SDW), where we have in the ground state a nonzero real triplet order parameter  $\Delta_{\text{Re}}^{\text{Or}}(\mathbf{r})$  describing an antiferromagnetic structure that does not cause a lattice-constant doubling<sup>21</sup> [here we retain the coordinate dependence of  $\Delta_{\text{Re}}^{\text{Or}}(\mathbf{r})$ , bearing in mind the possibility of the appearance of modulated structures]. Let us find the single-electron spectrum in the case of a commensurate SDW structure with allowance for the spin-orbit interaction (4):

$$E_{\sigma^{\pm}}(\mathbf{k}) = \pm [\zeta_{\mathbf{k}}^2 + \gamma_{\mathbf{k}}^2 + ([\lambda \mathbf{k}] - \Delta_{\text{Re}}^{\text{Or}})^2]^{1/2},\quad (47)$$

i.e., time-reversal symmetry is broken.

As we can see, the spectrum (47) exhibits a certain similarity with the spectrum (18) for orbital toroidal magnetic materials, and this similarity is not just formal, but is in fact connected with the appearance of toroidal order in the system of band electrons. To verify this, let us consider the structure of the orbital current that arises when the band-electron spin density is uniformly distributed. To do this, let us add to (31) the contribution due to the spin-orbit interaction:

$$\hat{\mathbf{j}}_{\text{L}} = -c \left[ \frac{\delta \hat{H}_1}{\delta \mathbf{A}} + \frac{\delta \hat{H}_2}{\delta \mathbf{A}} \right].\quad (48)$$

Let us consider the region of temperatures  $\Theta$  close to the phase transition point, where  $\Theta \ll \Delta_{\text{Re}}^{\text{Or}}$ . After calculations similar to those carried out in Ref. 1, we obtain in first order in  $\lambda$  and  $\Delta_{\text{Re}}^{\text{Or}}$  the expression

$$\mathbf{j}_{\text{L}}(\mathbf{r}) = \text{rot rot } \mathbf{T}_{\text{L}}(\mathbf{r}),\quad (49)$$

$$\mathbf{T}_{\text{L}} = A_{\text{L}} [\mathbf{n} \Delta_{\text{Re}}^{\text{Or}}], \quad A_{\text{L}} = \frac{e}{3} \frac{\lambda k_{\text{F}}^2}{(m^*)^2} \frac{7\zeta(3)}{8\pi^2 \Theta^2} \bar{N}(0).\quad (50)$$

Besides the orbital contribution (52), made by  $\mathbf{T}_{\text{L}}$  to the toroidal moment, there exists another (spin or inductive<sup>5</sup>) contribution due to the distribution of the nonuniform band-electron spin magnetic moment density:

$$\mathbf{M}_{\text{s}}(\mathbf{r}) = \text{Sp} \sum_{\mathbf{r}} \hat{\sigma} \hat{G}_{ii}(x, x).\quad (51)$$

Evaluating (51) in the case of the transverse SDW structure, when  $\text{div} \Delta_{\text{Re}}^{\text{Or}} \equiv 0$ , we find to first order in  $\mathbf{P}$  and  $\Delta_{\text{Re}}^{\text{Or}} \ll \Theta$  that

$$\mathbf{M}_{\text{s}}(\mathbf{r}) = \text{rot } \mathbf{T}_{\text{s}}(\mathbf{r}),\quad (52)$$

$$\mathbf{T}_{\text{s}} = A_{\text{s}} [\mathbf{n} (\Delta_{\text{Re}}^{\text{Or}})^{\perp}],\quad (53)$$

$$A_{\text{s}} = -\frac{7}{6} \frac{P \epsilon_{\text{F}} \mu_{\text{B}} \zeta(3)}{m \pi^2 \Theta^2} \bar{N}(0).\quad (54)$$

Introducing the "induction" current  $\mathbf{j}_{\text{s}}(\mathbf{r}) = \text{rot} \mathbf{M}_{\text{s}}(\mathbf{r})$ , we have for the total toroidal moment the expression

$$\mathbf{T}_{\text{x}} = \mathbf{T}_{\text{L}} + \mathbf{T}_{\text{s}}, \quad \mathbf{T}_{\text{x}} \perp \mathbf{n},\quad (55)$$

The toroidal moment in systems with SDW, in contrast to those with ECDW, which are considered in Ref. 1, lies in the plane perpendicular to the principal axis  $\mathbf{n}$ . Notice that there is no spin contribution to the toroidal moment in the case of the longitudinal SDW, when  $\text{rot} \Delta_{\text{Re}}^{\text{Or}} \equiv 0$ :

$$\mathbf{M}_{\text{s}}(\mathbf{r}) = \text{grad } \varphi_{\text{s}}(\mathbf{r}),\quad (56)$$

$$\varphi_{\text{s}} = -A_{\text{s}} \mathbf{n} (\Delta_{\text{Re}}^{\text{Or}})^{\parallel},\quad (57)$$

i.e., the  $\mathbf{M}_{\text{s}}$  field is purely longitudinal, in this case there remains only the orbital contribution.

We shall not dwell at length on the "toroidal" properties of systems with SDW, which in many respects are similar to the properties of systems with ECDW, and differ only in the magnitudes of the effects (by a factor on the order of the spin-orbit interaction). Of greatest interest, in our opinion, is the existence of a specific branch of transverse (with

respect to  $\Delta_{\text{Re}}^{\text{or}}$  SDW oscillations of the acoustic (Goldstone) type. Let us compute the dispersion law for these oscillations at zero temperature, i.e., at  $\Theta = 0$ . Using the scheme employed in §3, we find for the transverse oscillations of the triplet order parameter  $[\delta\Delta_{\text{Re}}^{\text{or}}(\mathbf{q}, \omega)]_{\perp}$  the system of equations

$$\begin{aligned} N_{\text{Re}}(\delta\Delta_{\text{Re}}^{\text{or}})_{\perp} + iK[(\delta\Delta_{\text{Im}}^{\text{or}})_{\perp}\Delta_{\text{Re}}^{\text{or}}] &= 0, \\ iK[(\delta\Delta_{\text{Re}}^{\text{or}})_{\perp}\Delta_{\text{Re}}^{\text{or}}] + N_{\text{Im}}(\delta\Delta_{\text{Im}}^{\text{or}})_{\perp} &= 0, \end{aligned} \quad (58)$$

where  $N_{\text{Re}}$ ,  $N_{\text{Im}}$ , and  $K$  are coefficients that have in the frequency region  $\omega \ll 2\Delta$  the form

$$\begin{aligned} N_{\text{Re}} \approx \left[ 1 - \frac{1}{3} \left( \frac{\gamma_0}{\Delta} \right)^2 \right] \omega^2 - \frac{(v_F q)^2}{3} - \frac{2}{45} \left( \frac{\epsilon_F}{m\Delta} \right)^2 \\ \times [P^2 q^2 + 2(\mathbf{P}\mathbf{q})^2], \end{aligned} \quad (59)$$

$$N_{\text{Im}} \approx -4\Delta^2 [1 + (\bar{g}_{\text{Im}}^{\text{or}})^{-1} - (\bar{g}_{\text{Re}}^{\text{or}})^{-1}], \quad K = 2 \frac{\mathbf{P}\mathbf{q}}{m}.$$

Equating the determinant of the system (59) to zero, we find that the low-frequency transverse branch of the SDW excitations has an acoustic dispersion law:

$$\begin{aligned} [\omega_{\text{Re}}^{\text{or}}(\mathbf{q})]_{\perp} \approx \left[ \frac{(v_F q)^2}{3} + aP^2 q^2 + b(\mathbf{P}\mathbf{q})^2 \right]^{1/2}, \\ a \approx \frac{22}{45} \left( \frac{\epsilon_F}{m\Delta} \right)^2, \quad b \approx \frac{4}{45} \left( \frac{\epsilon_F}{m\Delta} \right)^2. \end{aligned} \quad (60)$$

The contribution of the mode (60) to the dielectric susceptibility of the system is seen only to the extent that the spin-orbit interaction manifest itself. Notice that in systems with the hybrid type of antiferromagnetism, where there are localized magnetic moments besides the SDW,<sup>22</sup> the mode (60) gets mixed up with the magnons, this effect being strongest in the region of quasimomenta  $\mathbf{q}^*$  such that  $\omega'_{\text{Re}}(\mathbf{q}^*)_{\perp} \approx \Omega(0)$ , where  $\Omega(0)$  is the antiferromagnetic resonance frequency.

In these systems a peculiar magnon-light interaction mechanism arises (namely, interaction via toroidal oscillations of the band electrons). The question of the optical properties of such antiferromagnets is (among these are, in particular, certain magneto-electric crystals) requires further investigation.

## §6. CONCLUSION

The present paper is devoted to the investigation of the dynamics and optical properties of "soft" toroidal systems. The toroidal excitations can, depending on the structure of the ground state, be either high- or low-frequency excitations. And both the magnitude and the direction of the vector  $\mathbf{T}$  undergo oscillations.

For the investigation of the dynamics of "hard" toroidal structures (e.g., structures in which the magnitude of the vector  $\mathbf{T}_j$  at each lattice site  $j$  is fixed) the equations obtained in Ref. 4 and in the present paper are not suitable. The precession of the vector  $\mathbf{T}$  in "hard" toroidal magnetic crystals should be described by equations of the Landau-Lifshitz

type, but this problem has thus far not been solved—not at the phenomenological level, and certainly not within the framework of a microscopic model.

So far models for toroidal ordering have been discussed in which the characteristic spatial scale of the toroidal-moment density is much greater than the atomic scale (of order the correlation length for the electron-hole pair). It would be of great interest to consider systems with localized toroidal moments and compare the properties of such magnetic materials with those of Ising and Heisenberg magnetic substances. The mechanism underlying the establishment of long-range toroidal order in such systems has a number of distinctive features, the discussion of which is beyond the scope of the present paper.

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## APPENDIX

Let us discuss the derivation of the dynamical equations (12) for the toroidal ground state. Substituting the Green's function (7) into (10) and (11), and taking account of the fact that in the ground state

$$\Delta_{12}^* = \Delta_{21}^{**} = i\Delta_{1m}^{0*} = i\bar{g}_{1m}^* \text{Im} G_{12}^0(x, x), \quad (A1)$$

we find, after integrating over the internal frequency, that

$$\bar{M}(\omega) \begin{pmatrix} \delta\Delta_{\text{Re}}^{\text{or}}(\omega) \\ \delta\Delta_{\text{Im}}^{\text{or}}(\omega) \end{pmatrix} = \hat{Q}(\omega) \mathbf{A}_\omega, \quad (A2)$$

$$\bar{M}(\omega) = \begin{pmatrix} \bar{\Pi}_{\text{Re}}(\omega) & -i\omega\bar{L}(\omega) \\ i\omega\bar{L}(\omega) & \bar{\Pi}_{\text{Im}}(\omega) \end{pmatrix}. \quad (A3)$$

Let us discuss the structure of the source  $\hat{Q}$  on the right-hand side of (A2). The explicit expression for  $\hat{Q}$  in terms of the Green's functions of the Hamiltonian (3) in the absence of a field is the following:

$$\hat{Q} = \begin{pmatrix} \mathbf{Q}_{\text{Re}}(\omega) \\ \mathbf{Q}_{\text{Im}}(\omega) \end{pmatrix}, \quad (A4)$$

$$\begin{aligned} \mathbf{Q}_{\text{Re}(\text{Im})}(\omega) \\ = \text{Re}(\text{Im}) \sum_{\mathbf{k}} \frac{-i}{(2\pi)^4} \int d\epsilon \left\{ \left[ G_{11}^{0\mathbf{k}} \left( \epsilon - \frac{\omega}{2} \right) G_{12}^{0\mathbf{k}} \left( \epsilon + \frac{\omega}{2} \right) \right. \right. \\ \left. \left. - G_{12}^{0\mathbf{k}} \left( \epsilon - \frac{\omega}{2} \right) G_{22}^{0\mathbf{k}} \left( \epsilon + \frac{\omega}{2} \right) \right] \left( -\frac{e\mathbf{k}}{m'c} \right) \right. \\ \left. + \left[ G_{11}^{0\mathbf{k}} \left( \epsilon - \frac{\omega}{2} \right) G_{22}^{0\mathbf{k}} \left( \epsilon \right. \right. \right. \\ \left. \left. + \frac{\omega}{2} \right) - G_{12}^{0\mathbf{k}} \left( \epsilon - \frac{\omega}{2} \right) G_{12}^{0\mathbf{k}} \left( \epsilon + \frac{\omega}{2} \right) \right] \left( -\frac{i\epsilon}{mc} \mathbf{P} \right) \right\}. \end{aligned} \quad (A5)$$

The structure of (A5) is such that at  $\omega = 0$  the entire integral turns into a total derivative with respect to the quasimomentum  $\mathbf{k}$ , i.e., such that

$$Q_{\text{Re}(\text{Im})}(\omega=0) = \text{Re}(\text{Im}) \left[ -\frac{e}{c} \sum_{\mathbf{k}} \frac{\partial}{\partial \mathbf{k}} \left[ \frac{i\Delta_{1m}^{0*} - i\mathbf{P}\mathbf{k}/m}{E_{\mathbf{k}}} \right] \right]. \quad (A6)$$

A detailed analysis of this problem is carried out in the second paper cited in Ref. 1. Evaluating the integral over the frequency  $\varepsilon$  in (A5) with allowance for (A6), we obtain

$$\bar{Q}(\omega) = \begin{pmatrix} \frac{i\omega}{c} \Gamma_{\text{Re}}(\omega) \\ \frac{\omega^2}{c} \Gamma_{\text{Im}}(\omega) \end{pmatrix}, \quad (\text{A7})$$

where  $\tilde{\Gamma}_{\text{Re}}(\omega)$  and  $\tilde{\Gamma}_{\text{Im}}(\omega)$  are given by the formulas (16) and (17). Notice that the result (A6) is a consequence of the gauge invariance of the Hamiltonian (3). Therefore, only the space and time derivatives of the vector potential  $\mathbf{A}$  enter into all the expressions for the order parameters.

<sup>1)</sup>Here and below the interaction constants are assumed to be momentum-independent.

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