

# Axial toroidal moments in electrodynamics and solid-state physics

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We have found a family of multipole moments (axial toroidal moments) that differ in their space-time symmetry from the Maxwell–Lorentz moments known from electrodynamics. We consider the realization of this family in a system of magnetic charges, in electric dipole media, and also in media with magnetic fluxes. We investigate within the framework of a microscopic model a phase transition in a crystal with formation of an axial toroidal moment, and discuss certain interesting properties of the axial toroidal state.

## §1. INTRODUCTION

In the study of electromagnetic properties of systems with distributed charges and currents it becomes necessary to choose macroscopic characteristics that describe adequately the interaction of these systems with external fields and currents. A convenient mathematical procedure for this purpose is the formalism of multipole expansions of the microscopic charge density  $\rho(\mathbf{r}, t)$  and current density  $\mathbf{j}(\mathbf{r}, t)$ . For classical electrodynamics, this formalism is consistently developed, e.g., in Ref. 1. It is also shown there that besides the known families of charge and magnetic multipole moments there is produced a third family of toroidal multipole moments. The formal cause of the toroidal moments is the representation of vortex field (in this case, of the transverse current-density component  $\mathbf{j}_\perp(\mathbf{r}, t)$ , i.e., of a function for which  $\text{div } \mathbf{j}_\perp = 0$ ), in the form of the sum

$$\mathbf{j}(\mathbf{r}, t) = \text{rot}[\psi(\mathbf{r}, t)] + \text{rot rot}[\chi(\mathbf{r}, t)], \quad (1)$$

where  $\psi(\mathbf{r}, t)$  and  $\chi(\mathbf{r}, t)$  are respectively pseudoscalar and scalar functions. We shall call this formula the Neumann-Debye representation<sup>2–4</sup>. The expansion of  $\psi(\mathbf{r}, t)$  in radial spherical harmonics generates a family of magnetic multipole functions, among which the generating one is the dipole magnetic moment  $\mathbf{M}$ , since it can be used to construct all the higher moments of this family:  $M_{ik} = r_i M_k + r_k M_i$ , etc. A similar expansion of  $\chi(\mathbf{r}, t)$  generates a family of toroidal multipole moments.<sup>1,2</sup> The generating moment in the toroidal family is the toroidal dipole moment  $\mathbf{T}$ . In addition, expansion of the charge density  $\rho(\mathbf{r}, t)$  generates a family of charge multipole moments, of which the vector characteristic is the charge dipole moment  $\mathbf{P}$ . In accordance with their symmetry properties (the behavior under the coordinate inversion and time reversal operations  $\hat{I}$  and  $\hat{R}$ ) the vector quantities  $\mathbf{P}$ ,  $\mathbf{M}$ , and  $\mathbf{T}$  are different and are transformed in accordance with Table I.

It can be seen, however, that to construct the complete vector basis of multipole representations of the space-time inversion groups  $\hat{R} \otimes \hat{I}$  the set of vectors  $\mathbf{P}$ ,  $\mathbf{M}$ , and  $\mathbf{T}$  is insufficient, and in principle we need one more axial vector  $\mathbf{G}$  whose symmetry properties are indicated in the lower row of Table I. In the Maxwell-Lorentz classical electrodynamics, where

$$\mathbf{j}(\mathbf{r}, t) = \sum e_i \dot{\mathbf{r}}_i \delta(\mathbf{r} - \mathbf{r}_i(t)),$$

there is no place for realization of a vector with such properties. It will be shown nonetheless in the present paper that there are many physical applications of the mathematical formalism of multipole expansions, where the presence of the generating dipole moment  $\mathbf{G}$  of a family of its multipoles is indispensable.

In the modification of the multiple-expansion scheme for problems of electrodynamics with a magnetic charge (§2) the vector  $\mathbf{G}$  is the toroidal moment of the magnetic-charge current. It is similar in a certain sense to the toroidal moment  $\mathbf{T}$  of the electric charges. Owing to the pseudoscalar properties of the magnetic-charge density, however, it turns out that  $\mathbf{G}$  is an axial vector whereas  $\mathbf{T}$  is a polar vector.

In the electrodynamics of continuous media,  $\mathbf{G}$  can be introduced to describe systems with charge dipole moments (§3). In this case  $\mathbf{G}$  is the analog of the induction toroidal moment  $\mathbf{T}_{ind}$  in media with distributed magnetic dipole moments.<sup>1,6–8</sup>

Starting from the symmetry properties and also from the analogy with toroidal multipoles in the Maxwell-Lorentz electrodynamics, we shall hereafter call the vector  $\mathbf{G}$  the axial toroidal moment, and the vector  $\mathbf{T}$  the polar toroidal moment.

Introduction of the spin makes it necessary to classify the toroidal moments  $\mathbf{T}$  and  $\mathbf{G}$  with respect to the inversion transformation  $\hat{R}_\sigma$  in spin space. Where necessary, we shall label the even (singlet) vectors by a subscript  $s$ , and the odd (triplet) by  $t$ .

To describe phase transitions in crystals, the vectors  $\mathbf{T}$  and  $\mathbf{G}$  (or their higher multipoles) can be naturally be chosen to be order parameters that transform in accordance with certain irreducible representations of the magnetic group of a high-symmetry phase. We shall examine the distinguishing features of toroidal types of ordering and the systems for which introduction of these terms makes sense.

From the viewpoint of formal symmetry, many of the known order parameters are transformed in analogy with  $\mathbf{T}$  and  $\mathbf{G}$ . Thus, for example, multipole order parameters (spin densities) were introduced to describe spin magnets, and some of these parameters have transformation properties similar to those of  $\mathbf{T}$ .<sup>9,10</sup> The simplest case is that of a two-

TABLE I

	$\hat{I}$	$\hat{R}$
$P$	-	+
$M$	+	-
$T$	-	-
$G$	+	+

sublattice antiferromagnet.

The vector  $\omega$ , which is the dual of the antisymmetric part of the strain tensor,<sup>11</sup> transforms in analogy with  $\mathbf{G}_s$ , as does also the director  $\mathbf{n}$  that characterizes the orientational ordering in liquid crystals.<sup>12</sup> In the theory of "spin nematics"<sup>13</sup> are introduced order parameters similar in symmetry to  $\mathbf{G}_{s,t}$  and  $\mathbf{T}_{s,t}$ . A description of the properties of these systems in the language of toroidal distributions, however, is not particularly instructive physically, although it may be helpful when it comes to describe an interaction with an electromagnetic field.

The introduction of toroidal moments in a special group of order parameters is much more justified in the case of system with itinerant electrons. In particular, the polar toroidal moment  $\mathbf{T}_s$  describes orbital antiferromagnetic ordering,<sup>14</sup> the vector  $\mathbf{T}_t$  can be used to describe a number of spin itinerant antiferromagnets, the axial toroidal moment  $\mathbf{G}$  describes a peculiar charge ordering of the type of itinerant antiferroelectricity (*vide infra*), and the vector  $\mathbf{G}_t$  describes the orientational ordering in spin itinerant magnets.<sup>15</sup>

The multipole-expansion scheme permits a very effective description of the general macroscopic properties of many systems with complex distributions of the charges and currents (in particular, the response to an electric field). No less important a task, however, is the investigation of actual quantum-mechanical models that realize various types of multipole structures. A very clear example is the theory developed in Refs. 14 for polar toroidal ordering in crystals.

In this paper, along with a general phenomenological analysis of axial toroidal ordering (§4), we propose a microscopic model of a phase transition with formation of  $\mathbf{G}_s$  (§5). Another microscopic model with formation of  $\mathbf{G}_t$  was considered earlier,<sup>15</sup> and we deemed it unnecessary to pay attention to its specific features (the general derivations are given in §4). These models illustrate quite clearly the physical meaning of  $\mathbf{G}$  and permit a better understanding of the nature of its formation in crystals.

## §2. MULTIPOLE EXPANSION OF A SYSTEM OF MAGNETIC CHARGES

It was noted in Ref. 1 that in the dual-invariant scheme of electrodynamics there is complete symmetry between the multipole source forms and the types of fields, especially radiation fields.<sup>11</sup> We note that in an electromagnetic theory invariant to  $\hat{R}$  and  $\hat{I}$  reflections the electric-charge current should be a vector, and the magnetic-charge current a pseudovector, if the customary convention concerning the space-charge properties of the field  $\mathbf{E}$  and  $\mathbf{H}$  is adhered to (see, e.g., Ref. 17). Obviously (see Ref. 16), formulation of an electromagnetic theory with magnetic point charges is difficult

(see, however, Ref. 18) since, e.g., the relation  $\text{div } \mathbf{H} = g\sigma(\mathbf{r})$  either the charge is not simply a number, or else the charge is a number and there is no parity conservation in the theory. This difficulty does not arise in the macroscopic formulation, since the function  $\rho_g(\mathbf{r}, t)$  in the equation  $\text{div } \mathbf{H} = \rho_g(\mathbf{r}, t)$  can be always assumed to be odd, and  $j_g(\mathbf{r}, t)$  in the equation  $\text{curl } \mathbf{E} = -\mathbf{H} - \mathbf{j}_g(\mathbf{r}, t)$  can be regarded as an axial vector. Taking into account this difference between the world of magnetic charges and the world of electric charges, the problem of multipole expansion of the densities  $\rho_g(\mathbf{r}, t)$  and  $\mathbf{j}_g(\mathbf{r}, t)$  can be easily solved by simply making the replacements  $\rho_e \rightarrow \rho_g$  and  $\mathbf{j}_e \rightarrow \mathbf{j}_g$  in the corresponding equations of Ref. 1. We write out these equations and note the identical dual symbolism of  $g/e$  in the "electric" and "magnetic" worlds. Thus, the expansion of the charge is written in the form

$$\rho_{g/e}(\mathbf{r}, t) = \frac{1}{(2\pi)^3} \int_0^\infty \sum_{lm} (-ik)^l \frac{[4\pi(2l+1)]^{1/2}}{(2l+1)!!} \times F_{lmk}(\mathbf{r}) Q_{lm}^{g/e}(k^2, t) k^2 dk, \quad (2)$$

$$F_{lmk} = f_l(kr) Y_{lm}(\hat{\mathbf{r}}),$$

where the charge multipole distributions  $Q_{lm}^{g/e}(k^2, t)$  (Ref. 1) are defined as

$$Q_{lm}^{g/e}(k^2, t) = \frac{(2l+1)!!}{(-ik)^l [4\pi(2l+1)]^{1/2}} \int \rho_{g/e}(\mathbf{r}, t) F_{lmk}^*(\mathbf{r}) d^3\mathbf{r}. \quad (3)$$

This leads to definitions of the charge multipole moments

$$Q_{lm}^{g/e}(0, t) = \left(\frac{4\pi}{2l+1}\right)^{1/2} \int r^l Y_{lm}(\hat{\mathbf{r}}) \rho_{g/e}(\mathbf{r}, t) d^3\mathbf{r} \quad (4)$$

and their radii raised to the power  $2n$

$$\overline{r_{lm}^{2n}}(t) = \left(\frac{4\pi}{2l+1}\right)^{1/2} \int r^{l+2n} Y_{lm}(\hat{\mathbf{r}}) \rho_{g/e}(\mathbf{r}, t) d^3\mathbf{r}, \quad (5)$$

which complete the multipole parametrization of the initial function  $\rho_{g/e}(\mathbf{r}, t)$ .

The multipole expansion of the current density of the magnetic (electric) charges with the toroidal part singled out takes the form<sup>1</sup>

$$\mathbf{j}_{g/e}(\mathbf{r}, t) = \frac{1}{(2\pi)^3} \int \sum_{lm} (-ik)^{l-1} \frac{[4\pi(2l+1)(l+1)/l]^{1/2}}{(2l+1)!!} \times \left\{ k \mathbf{F}_{lmk}^{(0)} M_{lm}^{g/e}(k^2, t) + \mathbf{F}_{lmk}^{(+)} [Q_{lm}^{g/e}(0, t) + k^2 T_{lm}^{g/e}(k^2 t)] + \left(\frac{l}{l+1}\right)^{1/2} \mathbf{F}_{lmk}^{(-)} Q_{lm}^{g/e}(k^2, t) \right\} k^2 dk. \quad (6)$$

The basis of the expansion (6) is introduced as follows:

$$\mathbf{F}_{lmk}^{(-)} = -\frac{i}{k} \nabla F_{lmk}, \quad \mathbf{F}_{lmk}^{(0)} = \frac{i}{[l(l+1)]^{1/2}} \text{rot} \{ \mathbf{r} F_{lmk} \}; \quad (7)$$

$$\mathbf{F}_{lmk}^{(+)} = \frac{i}{k[l(l+1)]^{1/2}} \text{rot rot} \{ \mathbf{r} F_{lmk} \}.$$

The spherical vector are introduced as in Ref. 19:

$$\{ \mathbf{Y}_{l'm'}(\hat{\mathbf{r}}) \}^\mu = \sum_{m'} \langle l' m' | \mu | l m \rangle Y_{l'm'}(\hat{\mathbf{r}}).$$

The magnetic multipole distributions are

$$M_{lm}^{g/e}(k^2, t) = \frac{-i(2l+1)!!}{(-ik)^l [4\pi(2l+1)(l+1)/l]^{1/2}} \int \mathbf{F}_{lmk}^{(0)} \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}; \quad (8)$$

the magnetic multipole moments are

$$M_{lm}^{g/e}(0, t) = \left[ \frac{4\pi}{(2l+1)(l+1)^2} \right]^{1/2} \int r^l \mathbf{Y}_{ilm} \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}; \quad (9)$$

the toroidal multipole distributions are given by

$$T_{lm}^{g/e}(k^2, t) = \frac{(2l+1)!!}{(-ik)^{l+1} [4\pi(2l+1)(l+1)/l]^{1/2}} \times \int \left\{ \mathbf{F}_{lmk}^{(+)} - \frac{4\pi(-ikr)^{l-1}}{(2l+1)^{1/2}} \frac{(l+1)^{1/2}}{(2l-1)!!} \mathbf{Y}_{il-lm}^* \right\} \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}; \quad (10)$$

the toroidal multipole moments are

$$T_{lm}^{g/e}(0, t) = \frac{-(4\pi l)^{1/2}}{2(2l+1)} \int r^{l+1} \left\{ \mathbf{Y}_{il-lm}^* + \frac{2[l(l+1)]^{1/2}}{2l+3} \mathbf{Y}_{il+l+1m}^* \right\} \times \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}; \quad (11)$$

the longitudinal charge multipole distributions are

$$Q_{lm}^{g/e}(k^2, t) = \frac{[4\pi(2l+1)]^{1/2}}{(2l+1)!!} \int \mathbf{F}_{lmk}^{(-)} \cdot \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}; \quad (12)$$

and the longitudinal charge multipole moments are

$$Q_{lm}^{g/e}(0, t) = (4\pi l)^{1/2} \int r^{l-1} \mathbf{Y}_{il-lm}^*(\hat{\mathbf{r}}) \cdot \mathbf{j}_{g/e}(\mathbf{r}, t) d^3\mathbf{r}. \quad (13)$$

When the conditions for spectral expansion of  $\rho_{g/e}(\mathbf{r}, t)$  and  $\mathbf{j}_{g/e}(\mathbf{r}, t)$  are satisfied (the simplest case is that of harmonic sources) the last definitions reduce to Eqs. (3) and (4), and the expressions (12), (13) are functionally dependent on  $\dot{Q}_{lm}$  in all cases, in view of the conservation of the 4-current. The question of how the "longitudinal" moments  $\dot{Q}_{lm}(0, t)$  "turn up" in the expansion of the transverse part of the current ( $\text{div } \mathbf{j} = 0$ ) [see Eq. (6)] is more complicated and is answered in Ref. 1 (see also the literature cited there).

It is easy to ascertain, by starting from the properties of the vector spherical functions  $F_{lmk}$  relative to  $\hat{I}$  reflections, that the distributions  $M_{lm}^g$  now produce  $E_l$ -type fields (in particular, they emit  $E_l$  multipoles), while the charge  $Q_{lm}^g$  and toroidal  $T_{lm}^g$  distributions produce the  $M_l$ -type fields (in particular,  $\dot{Q}_{lm}^g(0, t)$  and  $T_{lm}^g(0, t)$  are responsible for the emission of  $M_l$  multipoles).

### §3. MULTIPOLE REPRESENTATIONS OF DIPOLE MEDIA

Since no free magnetic charges (and their currents) have been observed so far (the history of the problem can be found, e.g., in Ref. 20). It might seem that the results of the multipole expansion in dual electrodynamics, which are reported in §2, are purely of scholastic character. We shall show, however, that the dual symmetry  $M_{lm}^e \leftrightarrow Q_{lm}^g$ ,  $M_{lm}^g \leftrightarrow Q_{ml}^e$  has a rather profound meaning, and the formalism expounded in §2 is quite useful for the description of dipole structures in the electrodynamics of continuous media.

We describe a magnetic medium as an aggregate  $\{\mu_i\}$  of elementary dipole moments ( $i = 1, 2, \dots, N$ ). The magnetic-dipole current density  $\mathbf{j}_\mu$  is introduced in known fashion<sup>21</sup>:

$$\mathbf{j}_\mu(\mathbf{r}, t) = \sum_i [\mu_i \nabla \delta(\mathbf{r} - \mathbf{r}_i(t))] \rightarrow \text{rot } \mathbf{M}_\perp(\mathbf{r}, t), \quad (14)$$

where the arrow denotes a transition to a continuous distribution of the magnetic-dipole-moment density (magnetization)  $\mathbf{M}_\perp$ . It is convenient to introduce besides (14) the formal quantity  $\rho_\mu$ , which is the pseudoscalar distribution

density of the "magnetic charges":

$$\rho_\mu(\mathbf{r}, t) = -\text{div } \mathbf{M}_\parallel(\mathbf{r}, t). \quad (15)$$

Note the usual Maxwell-Lorentz equations do not contain the quantity  $\mathbf{M}_\parallel(\mathbf{r}, t)$  (the longitudinal component of the magnetization).

Replacing now in Eqs. (3)–(5) the density  $\rho_e$  by  $\rho_\mu$ , we get an almost complete analogy with the multipole expansions of these quantities (the only difference is that we must put

$$M_{00} = \int \rho_\mu(\mathbf{r}, t) d^3\mathbf{r} \equiv 0.$$

Clearly, the corresponding equations give the "charge" multipole moments of the system of magnetic dipoles. We note that no fully symmetric scheme of multipole expansion is produced, since  $\text{div } \mathbf{j}_\mu \neq \dot{\rho}_\mu$  and  $M_{00} = 0$ .

Thus, the vortical current  $\mathbf{j}_\mu$  makes no contribution to  $\dot{Q}_{lm}$ , but its contribution to  $T_{lm}$  does not vanish. Note that historically it is just this contribution that initiated in fact the introduction of toroidal distributions in electromagnetism (see Ref. 6 and the references therein). The induction part of the toroidal moment

$$T_{lm}^\mu(t) = i \left[ \frac{4\pi l}{(2l+1)(l+1)} \right]^{1/2} \int r^l \mathbf{Y}_{ilm} \mathbf{M}_\perp(\mathbf{r}, t) d^3\mathbf{r} \quad (16)$$

was named earlier "induced electric moments" (Ref. 8). This equation is given here in the normalization of Ref. 6. It can be seen that (16) differs from the definition of  $M_{lm}^e$  for free current by the substitution  $\mathbf{j}^e \rightarrow \mathbf{M}_\perp$ . The elementary dipole  $\mathbf{T}_\mu$  can thus be written, in analogy with  $\mathbf{M}$ , in the form

$$\mathbf{T}_\mu = \frac{1}{2} \sum_i [\mathbf{r}_i \mu_i]. \quad (17)$$

The geometric representation of the induction (toroidal) dipole is a closed circular chain of elementary dipole moments  $\{\mu_i\}$  (Fig. 1a).

We shall describe electric dipole media by a set of elementary dipoles  $\{d_i\}$ . In this description, the characteristic

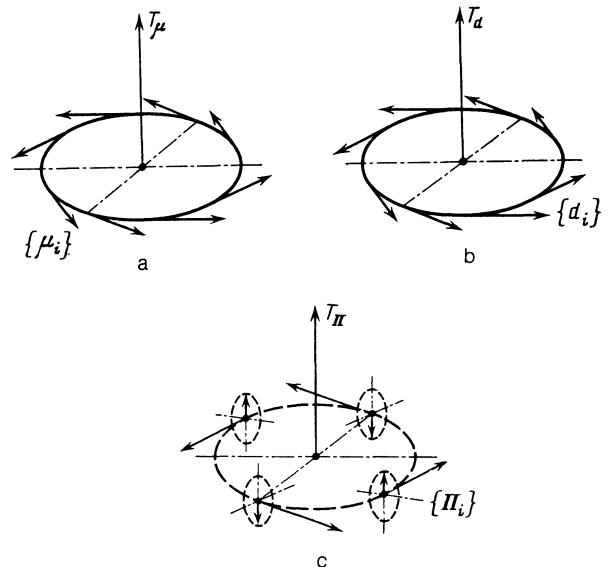


FIG. 1.

scales over which the multipole expansion is carried out are large compared with the characteristic dimensions of the dipoles themselves, so that the latter can be regarded as pointlike. The electric polarization of the medium is introduced in the usual manner:

$$\mathbf{P}(\mathbf{r}, t) = \sum_i \mathbf{d}_i \delta(\mathbf{r} - \mathbf{r}_i(t)). \quad (18)$$

Obviously, the electric polarization  $\mathbf{P}$  can imitate, by virtue of the dual symmetry, multipole moments of magnetic charges. We introduce an axial "current" that is a pseudo-vector with respect to time reversal in a medium of distributed electric dipoles:

$$\mathbf{j}_a^{(a)}(\mathbf{r}, t) = \sum_i [\mathbf{d}_i \nabla \delta(\mathbf{r} - \mathbf{r}_i(t))] \rightarrow \text{rot } \mathbf{P}_\perp(\mathbf{r}, t), \quad (19)$$

where  $\mathbf{P}_\perp$  is the transverse part of the electric-dipole-moment density (polarization). The longitudinal part  $\mathbf{P}_\parallel$  of the polarization is described by the scalar distribution density of the electric charges

$$\rho_a(\mathbf{r}, t) = \text{div } \mathbf{P}_\parallel(\mathbf{r}, t). \quad (20)$$

We emphasize that the axial current  $\mathbf{j}_a^{(a)}$  differs in its nature from the polar current  $\mathbf{j}_a$  in the Maxwell-Lorentz equation:

$$\mathbf{j}_a(\mathbf{r}, t) = \sum_i \dot{\mathbf{d}}_i \delta(\mathbf{r} - \mathbf{r}_i(t)) \rightarrow \dot{\mathbf{P}}(\mathbf{r}, t). \quad (21)$$

In contrast to the magnetization components  $\mathbf{M}_\perp$  and  $\mathbf{M}_\parallel$ , both polarization components  $\mathbf{P}_\parallel$  and  $\mathbf{P}_\perp$  enter in the Maxwell-Lorentz equations, and  $\mathbf{P}_\perp$  drops out of them only in the static limit.

Returning to Eq. (19), we see that substitution of the effective current  $\mathbf{j}_a^{(a)}$  in the definition of  $\mathbf{M}_{lm}^g$  transforms, after appropriate integration by parts, the definition of  $\mathbf{M}_{lm}^g$  into the usual definition of the electric part, in which  $\mathbf{j}_g$  is replaced by  $\mathbf{P}_\perp$ . In this case, of course,  $E_{00}^d \equiv 0$ . On the other hand, the situation with  $T_{lm}^g$  is more curious. Substitution of (19) in the definition (11) and transfer of the derivative lead to an equation similar to (16):

$$T_{lm}^d(t) = i \left[ \frac{4\pi l}{(2l+1)(l+1)} \right]^{1/2} \int r^l \mathbf{Y}_{lm} \mathbf{P}_\perp(\mathbf{r}, t) d^3r. \quad (22)$$

It follows directly that the elementary "induced" axial toroidal dipole moment is

$$\mathbf{T}_d = \frac{1}{2} \sum_i [\mathbf{r}_i \mathbf{d}_i] \quad (23)$$

and its geometric representation is a closed chain of electric charge dipoles (Fig. 1b). The last equations demonstrate the simplest possibility of imitating, in the dipole representation, symmetry elements that are absent from a system of electric point charges. (In principle, completeness of the properties under  $\hat{R}$  and  $\hat{I}$  reflections can be obtained also in media made up of elementary higher multiples of the usual type.)

We indicate one more possibility of realizing the  $T^g$  symmetry in media. Recall that in the problem of particle motion in a centrosymmetric potential one encounters a correlation between the angular-momentum and momentum vectors  $\hat{\mathbf{L}}$  and  $\hat{\mathbf{P}}$ . This correlation is described by the Runge-Lenz operator

$$\Pi = [\hat{\mathbf{L}} \hat{\mathbf{P}}]. \quad (24)$$

The Runge-Lenz operator appears formally even in the analysis of the dynamic symmetry of the nonrelativistic Kepler problem, but it plays a more substantial role in the analysis of the dynamic symmetry of the relativistic Coulomb problem<sup>22</sup> or of the motion of a free relativistic particle that satisfies the Dirac equation.<sup>23</sup> The presence in a medium of a distribution of a vector of type  $\Pi$  (of orbital or spin origin) also requires introduction of axial toroidal moments.

Consider now a medium with distributed moment fluxes described in the general case by the second-rank tensor (dyad)

$$\Pi_{ij} = \langle \hat{\mathbf{L}}_i \otimes \hat{\mathbf{P}}_j \rangle, \quad i, j = x, y, z. \quad (25)$$

We represent (25) as a sum of a symmetric  $\Pi_{ij}^{(s)}$  and antisymmetric  $\Pi_{ij}^{(as)}$  parts. The latter is the dual of the polar vector  $\Pi$  and can be described in analogy with the preceding analysis of the vector  $\mathbf{P}$  in electro-dipole media. We introduce a transverse induction (axial) current

$$\mathbf{j}_\Pi^{(a)} = \text{rot } \Pi_\perp(\mathbf{r}, t) = \text{rot } \langle [\hat{\mathbf{L}} \hat{\mathbf{P}}] \rangle. \quad (26)$$

Obviously, substitution of this current in Eqs. (10) and (11), just as substitution of  $\mathbf{j}_a$ , will give rise to a multipole family  $T_{lm}^\Pi$ . The elementary is in this case

$$\mathbf{T}_\Pi = \frac{1}{2} \sum_i [\mathbf{r}_i \Pi_\perp(\mathbf{r}_i, t)]. \quad (27)$$

The ideal geometric picture of this dipole consists of local moments precessing on a circle (Fig. 1c).

#### §4. PHENOMENOLOGICAL THEORY OF AXIAL TOROIDAL ORDERING IN CRYSTALS

Consider a system of itinerant electrons, in which a second-order phase transition produces a unique type of long-range order describable by an axial vector that is even with respect to time reversal. Before turning to actual microscopic models that explain the mechanisms of this ordering, let us dwell on some of its phenomenological consequences that involve only formal symmetry considerations.

Among all magnetic symmetry classes, the following 43 allow existence of an axial vector that is even with respect to time reversal  $\hat{R}$ : 1) thirteen ordinary crystal classes that do not contain  $\hat{R}$  at all:

$$C_1, C_i, C_s, C_2, C_{2h}, C_4, S_4, C_{4h}, C_6, S_6, C_{3h}, C_{6h}, C_3; \quad (28)$$

2) the same classes supplemented by the operation  $R$ : 3) seventeen proper magnetic classes:

$$C_1(C_1), C_2(C_1), C_{2h}(C_i), C_{2h}(C_2), C_{2h}(C_s), C_s(C_1), C_4(C_2), S_4(C_2), C_{4h}(C_4), C_{4h}(C_{2h}), C_{4h}(S_4), S_6(C_3), C_{3h}(C_3), C_6(C_3), C_{6h}(C_6), C_{6h}(S_6), C_{6h}(C_{3h}). \quad (29)$$

We consider hereafter only systems that do not contain nontrivial translations, in which specification of the magnetic class is the necessary and sufficient condition that determines the existence of a vector  $\mathbf{G}$ . As in the case of the polar vector  $\mathbf{T}$ , all the foregoing classes can be easily obtained from the tables of irreducible representations of point groups.

The establishment of an axial toroidal order in a crystal can be associated with relaxation of some collective electron

oscillation mode. We name this an axial toroidal mode, in analogy with the previously considered polar toroidal oscillation modes.<sup>24</sup> Assume that for some reason the frequency of a toroidal mode became anomalously small and a tendency to establish a toroidal long-range order set in.

It is convenient to analyze the general properties of such systems by the effective-Lagrangian method. Assuming that the symmetry group of the high-symmetry phase admits as a subgroup one of the axial magnetic groups listed above, we consider small low-frequency axial toroidal oscillations above the phase-transition point. In the absence of external field, the Lagrangian of the system takes the form

$$\mathcal{L} = K - U, \quad (30)$$

$$K = \frac{1}{2M_G} (\dot{\mathbf{G}})^2 + D_G (\ddot{\mathbf{G}})^2, \quad (31)$$

$$U = \alpha \mathbf{G}^2 + \beta \mathbf{G}^4 + \gamma (\text{rot } \mathbf{G})^2, \quad (32)$$

where the coefficients  $M_G$ ,  $D_G$ ,  $\alpha$ ,  $\beta$ ,  $\gamma > 0$ , and the system symmetry above the transition point is assumed for the sake of argument to be cubic. We have retained in the kinetic energy (31) a term of type  $(\mathbf{G})^2$ , allowance for which will be justified presently. From symmetry consideration, we express the interaction with the external field in the form

$$\Delta \mathcal{L}_A = (-\lambda/c) \mathbf{G} \text{rot } \dot{\mathbf{A}}, \quad (33)$$

where  $\mathbf{A}$  is the vector potential,  $c$  the speed of light, and  $\lambda$  a coefficient. Equation (33) can be represented also in the form of two equivalent expressions

$$\Delta \mathcal{L}_B = (-\lambda/c) \mathbf{G} \dot{\mathbf{B}}, \quad (34)$$

$$\Delta \mathcal{L}_E = \lambda \mathbf{G} \text{rot } \mathbf{E}, \quad (35)$$

where  $\mathbf{B} = \text{curl } \mathbf{A}$ ,  $\mathbf{E} = -\dot{\mathbf{A}}/c$ , and we used the Maxwell equation for the solenoidal component of the electric field  $\mathbf{E}$ :

$$\text{rot } \mathbf{E} = -\dot{\mathbf{B}}/c. \quad (36)$$

It can be seen from (33) and (34) that the addition to the dynamic magnetic susceptibility  $\Delta\chi(\omega)$  takes at low frequencies the form

$$\Delta\chi(\omega) = -\left(\frac{\lambda}{c}\right)^2 \frac{M_G \omega^2}{\omega^2 - \Omega_G^2}, \quad (37)$$

where  $\Omega_G^2 = 2M_{G\alpha}$  is the natural frequency of the axial toroidal oscillations. The vanishing of  $\Omega_G$  corresponds to a second-order phase transition. Note that we are dealing throughout only with transverse axial toroidal oscillations, while the longitudinal ones do not interact with the electric and magnetic fields. In principle, axial toroidal modes could react to a current of magnetic charges (were they to exist), just as polar toroidal modes react to an ordinary electric current.<sup>14</sup>

What is noteworthy is the nontrivial frequency dependence of  $\Delta\chi(\omega)$  [the numerator is proportional to  $\omega^2$ , just as in the case of polar toroidal oscillations, where the dynamic dielectric constant has an anomaly similar to (37), Ref. 24]. Expression (37) is valid only at low frequencies, when the second term of (31) can be neglected. To obtain the correct

asymptotic form at  $\omega \gg \Omega_G$  it is necessary to take this term into account, and then the contribution  $\Delta\chi(\omega)$  vanishes at high frequencies, as it should. In the microscopic model (§5) the second term becomes appreciable at frequencies  $\omega \sim E_g$ , where  $E_g$  is a characteristic one-electron energy of the order of the semiconductor band gap.

Curious effects can take place in systems in which the axial toroidal ordering is accompanied by some other type of magnetic long-range order. For example, in the case of antiferromagnets with spin density waves (SDW) the appearance of axial toroidal order leads to "weak" ferromagnetism.<sup>15</sup>

In the case of antiferromagnets that contain localized moments besides itinerant electrons, the axial toroidal order also introduces in the Lagrangian of the system a term responsible for the weak ferromagnetism of the local moments:

$$\Delta \mathcal{L}_F = \xi \mathbf{G} [\mathbf{L} \mathbf{M}], \quad (38)$$

where  $\mathbf{L}$  is the antiferromagnetism vector,  $\mathbf{M}$  the average magnetic moment of the cell, and  $\xi$  a coefficient. We note that in the model of Ref. 15 the entire effect is of purely exchange origin and does not contain a relativistic smallness. On the other hand, the usual Dzyaloshinskii-Moriya weak-ferromagnetism mechanism is connected with spin-orbit or magnetodipole interaction.<sup>25</sup>

If the axial toroidal moment  $\mathbf{G}$  has an incommensurate structure below the phase-transition point, inhomogeneous spontaneous polarization sets in:

$$\mathbf{P} = -\lambda \text{rot } \mathbf{G}, \quad (39)$$

as a direct result of writing the term representing the interaction with the electric field in the form (35). The transverse static dielectric constant diverges at the phase-transition point<sup>2)</sup>

$$\Delta \epsilon_{\perp}(\mathbf{q}) = 4\pi\lambda^2 q^2 M_G / \Omega_G^2(\mathbf{q}). \quad (40)$$

On a certain wave vector  $\mathbf{q} = \mathbf{q}_0$ , obtained when the frequency of the natural transverse oscillations vanishes, we have  $\Omega_G(\mathbf{q}_0) \rightarrow 0$ ,  $\Theta \epsilon_{\perp}(\mathbf{q}_0) \rightarrow \infty$  (at  $\mathbf{q}_0 \neq 0$ ).

Axial toroidal oscillations can interact with other collective excitations in crystals. Consider, for example, a ferromagnet with local moments, in which axial toroidal ordering is not realized in the ground state. At the same time, the collective toroidal oscillations are intermixed with the ordinary magnons, since the effective Hamiltonian of the system contains terms of the type

$$\Delta U_{eff}^{(G)} = -\mathbf{M} \mathbf{H}_{eff}(\mathbf{G}), \quad (41)$$

$$\mathbf{H}_{eff}(\mathbf{G}) = \lambda_1 \mathbf{G}, \quad (42)$$

where  $\lambda_1$  is a proportionality coefficient and  $\mathbf{M}$  is the magnetic moment. We write down the Bloch equation, with (41) and (42) taken into account, for small deviations  $\mathbf{m}(\mathbf{r}, t)$  of the magnetic moment  $\mathbf{M}$  from the equilibrium value  $\mathbf{M}_0$ :

$$\dot{\mathbf{m}} = \gamma_0 [\mathbf{H}_{eff} \mathbf{M}], \quad (43)$$

$$\mathbf{M} = \mathbf{M}_0 + \mathbf{m}, \quad \mathbf{H}_{eff}^{(0)} + \mathbf{H}_{eff}(\mathbf{G}) = \mathbf{H}_{eff}, \quad (44)$$

$$\mathbf{H}_{eff}^{(0)} = \alpha_{ik} \frac{\partial^2 \mathbf{m}}{\partial x_i \partial x_k} + \mathbf{H}_{an}^{(0)}. \quad (45)$$

Here  $\gamma_0 = g|e|/2mc$ , where  $g$  is the gyromagnetic ratio and  $\mathbf{H}_{an}^{(0)}$  is the magnetic anisotropy contribution, which we shall not write down here explicitly (see Ref. 12). Putting  $\tilde{\alpha} = \alpha_{ik} n_i n_k$ , where  $\mathbf{n}$  is a unit vector in the direction of the wave vector  $\mathbf{q}$ ,  $\mathbf{m} \perp \mathbf{M}_0$ , and  $g = 2$ , we obtain for the Fourier components of  $\mathbf{m}_\omega$

$$i\omega \mathbf{m}_\omega = \gamma_0 \{ [\mathbf{H}_{eff}^{(0)} \mathbf{M}_0]_\omega + i\omega \lambda_1 [\mathbf{G}_\omega \mathbf{M}_0] \}, \quad (46)$$

and use for  $\mathbf{G}_\omega$  an equations that follows from the variation of the effective Lagrangian with allowance for (33) and (34):

$$\left( \frac{\omega^2}{2M_G} - \alpha - \gamma q^2 \right) \mathbf{G}_\omega + \frac{i\omega \lambda_1}{2} \mathbf{m}_\omega = 0. \quad (47)$$

The equation for the dispersion law of the magnon-toroidal oscillations is of the form

$$\omega = \gamma_0 \left[ \omega_H(0) + \tilde{\alpha} q^2 + \frac{\lambda_1^2 \omega^2}{2D_\omega} \right] M_0, \quad (48)$$

$$D_\omega = \frac{\omega^2}{2M_G} - \alpha - \gamma q^2, \quad (49)$$

where  $\omega_H(0)$  is the ferromagnetic-resonance frequency. The solutions of the cubic equation (48) in general form are too unwieldy to write down here. It is clear that the mixing of the toroidal oscillations with the magnons is a maximum at the quasimomenta  $\mathbf{q}_0$  determined from the approximate relation

$$\begin{aligned} \gamma_0 M_0 [\tilde{\alpha} q_0^2 + \omega_H(0)] &\approx \Omega_G(\mathbf{q}_0), \\ \Omega_G^2(\mathbf{q}_0) &= (\alpha + \gamma q_0^2) 2M_G. \end{aligned} \quad (50)$$

Entanglement of the magnons with axial toroidal oscillations is possible also in antiferromagnets at the proper parity of the antiferromagnetic structure. The generalization of Eqs. (46)–(49) to include antiferromagnets is obvious.

Axial toroidal oscillations interact in a rather distinct manner with light; this interaction produces new polariton branches. In fact, the Lagrangian of the system in a magnetic field  $\mathbf{A}(\mathbf{r}, t)$  is

$$\mathcal{L} = \mathcal{L}_G + \frac{\lambda}{c} \mathbf{A} \operatorname{rot} \dot{\mathbf{G}} + \frac{1}{8\pi} \left[ \frac{\varepsilon_\infty}{c^2} (\dot{\mathbf{A}})^2 - (\operatorname{rot} \mathbf{A})^2 \right]. \quad (51)$$

Varying (51) with respect to  $\mathbf{G}$  and  $\mathbf{A}$  we obtain a system of equations for the axial toroidal moment and the Maxwell equation:

$$\begin{aligned} -\frac{1}{2M_G} \ddot{\mathbf{G}} + \gamma \nabla^2 \mathbf{G} - \frac{\lambda}{c} \operatorname{rot} \dot{\mathbf{A}} &= 0, \\ -\frac{\varepsilon_\infty}{c^2} \ddot{\mathbf{A}} - \operatorname{rot} \operatorname{rot} \mathbf{A} + \frac{4\pi\lambda}{c} \operatorname{rot} \dot{\mathbf{G}} &= 0. \end{aligned} \quad (52)$$

For the natural oscillation frequencies we get from (51) and (52)

$$\hat{H} = \left[ \frac{1}{2m_1} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)^2 + \frac{E_g}{2} + e\mathcal{D} \eta_{\alpha\beta}^{12} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)_\alpha \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)_\beta - \hat{\Delta}_{12} \right. \\ \left. \eta_{\alpha\beta}^{21} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)_\alpha \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)_\beta - \hat{\Delta}_{21} - \frac{1}{2m_2} \left( \frac{\nabla}{i} - \frac{e}{c} \mathbf{A} \right)^2 - \frac{E_g}{2} + e\mathcal{D} \right], \quad (59)$$

$$\omega_{1,2}^2 = \frac{1}{2\varepsilon_\infty} \{ q^2 \tilde{c}^2 + \varepsilon_\infty \Omega_G^2 \pm [ (q^2 \tilde{c}^2 + \varepsilon_\infty \Omega_G^2)^2 - 4\varepsilon_\infty \Omega_G^2 q^2 c^2 ]^{1/2} \}, \quad (53)$$

$$\tilde{c}^2 = c^2 + 4\pi M_G \lambda^2.$$

As  $q \rightarrow 0$  Eq. (53) takes the simpler form

$$\omega_1^2 \rightarrow \Omega_G^2, \quad \omega_2^2 \rightarrow q^2 c^2 / \varepsilon_\infty, \quad (54)$$

and at  $q^2 c^2 \gg \varepsilon_\infty \Omega_G^2$  we have the asymptotic relations

$$\omega_1^2 \rightarrow \Omega_G^2 (c^2 / \tilde{c}^2), \quad \omega_2^2 \rightarrow q^2 \tilde{c}^2 / \varepsilon_\infty. \quad (55)$$

Note that Eqs. (51)–(55) are valid only at low frequencies and small momenta ( $\omega, c_q \ll E_g$  in the microscopic model of §5). If  $\omega, c_q \sim E_g$  we must retain in the Lagrangian (31) the terms with higher derivatives of the order parameter  $\mathbf{G}$ , in analogy with the case of polar toroidal oscillations.<sup>24</sup> As a result we have at high energies and momenta the correct asymptotic forms

$$\omega_1^2 \rightarrow \Omega_G^2, \quad \omega_2^2 \rightarrow q^2 c^2 / \varepsilon_\infty. \quad (56)$$

Interesting nonlinear optical effects can take place in systems with axial toroidal ordering. Below the transition point, in particular, an anomalous contribution proportional to the electric field  $\mathbf{E}$  is made to the components of the gyration tensor  $g_{ik}$ :

$$g_{ik} = \gamma_{ikln} G_l E_n. \quad (57)$$

Note that the tensor  $g_{ik}$  in systems with polar toroidal order can also acquire an anomalous contribution, but one proportional to the magnetic field  $\mathbf{H}$ :

$$g_{ik} = \gamma'_{ikln} T_l H_n. \quad (58)$$

The anomalous behavior of the electrooptic and magneto-optic characteristics of crystals can be of help in the identification of toroidal transitions.

## §5. MACROSCOPIC MODEL OF AXIAL TOROIDAL ORDERING

Consider now a two-band model of a semiconductor or a semimetal with straight extrema at the point  $\mathbf{k}_0$  of the Brillouin zone. Assume that the matrix element of the interband dipole transition is zero at the point  $\mathbf{k}_0$  (the wave functions of bands 1 and 2 have like parity but belong to different irreducible representations of the group of the wave vector  $\mathbf{k}_0$ ) and the matrix element of the interband transition with respect to the orbital momentum differs from zero. A model with this symmetry was considered in Ref. 26, where it was shown that realization of electron-hole pairing with imaginary singlet order parameter gives rise to orbital ferromagnetism of the occupied-band electrons. We write the Hamiltonian of the system in the  $\mathbf{k} \cdot \mathbf{p}$  approximation in an external electromagnetic field<sup>27</sup>

where  $m_1$  and  $m_2$  are the effective masses of the electrons and holes in bands 1 and 2;  $E_g$  is the band gap of the semiconductor ( $E_g < 0$  for semimetals):  $\mathbf{A}(\mathbf{r}, t)$  and  $\Phi(\mathbf{r}, t)$  are the vector and scalar potentials of the electromagnetic field, and  $\hat{\Delta}_{ij}(\mathbf{r}, t)$  is the order parameter that describes the ordered state below the phase-transition point in a two-band model of the excitonic-dielectric type and has in the general case a tensor structure<sup>28</sup>:

$$\hat{\Delta}_{ij} = \Delta_{ij}^s \hat{I} + \Delta_{ij}^t \hat{\sigma}, \quad (60)$$

where  $\hat{I}$  is a unit matrix and  $\hat{\sigma}$  is a vector made up of Pauli matrices. It is assumed that the effective interaction constant  $g_{\text{Re}}^s$  is a maximum in the case of a transition into a state with  $\Delta_{\text{Re}}^s$ , so that the corresponding transition temperature (or the critical value of the band gap  $E_g^*$  in the semiconductor model at  $T = 0$ ) is also a maximum, and the state with  $\Delta_{\text{Re}}^s$  is energywise most favored. Explicit forms of the effective interaction constants for all possible structures of the order parameter  $\hat{\Delta}_{ij}$  can be found, e.g., in Ref. 28.

The tensors  $\eta_{\alpha\beta}^{ij}$  in the one-electron part of the Hamiltonian  $\hat{H}$  are given by

$$\eta_{\alpha\beta}^{12} = \eta_{\alpha\beta}^{21} = -\frac{1}{2} \sum_{s \neq 1,2} \left[ \frac{1}{E_1 - E_s} + \frac{1}{E_2 - E_s} \right] \frac{P_{1s}^\alpha P_{2s}^\beta}{m^2}, \quad (61)$$

where  $E_s$  is the  $s$ -band energy at the point  $\mathbf{k}_0$ , while  $P_{is}$  is the momentum matrix element between the and  $i = 1, 2$  and the remote band  $s \neq 1, 2$  and  $m$  is the electron mass. We consider next the case when the tensor  $\eta_{\alpha\beta}^{12}$  is pure real (this occurs, for example, when the Bloch wave functions  $\varphi_n k_0(r)$  at the point  $\mathbf{k}_0$  can be chosen real).

The system with Hamiltonian (59) is analyzed by the standard Green's function method, and we shall not dwell on the calculation technique (a detailed exposition of the general calculation procedure in models of the excitonic-dielectric type can be found in Ref. 28). We note only the singularities connected with the reaction to an external magnetic field, since it just these singularities which explain the type of electronic ordering that is produced in the system below the phase-transition point. We write down the effective Lagrangian that describes the transition into the state with  $\varphi_n k_0(r)$  at  $T = 0$  for a semiconductor model with a small band gap  $E_g \approx E_g^*$ , where  $E_g^*$  is of the order of the exciton band energy. Accurate to lower terms relative to the parameter  $\Delta_{\text{Re}}^s/E_g \ll 1$ , when  $\Delta_{\text{Re}}^s = (\Delta_{12}^s + \Delta_{21}^s)/2$ , we obtain in a weak and slowly varying transverse field, after laborious calculations,

$$\begin{aligned} \mathcal{L}_{\text{eff}} &= K - U, \\ K &= \bar{N} \left[ \frac{1}{2M_G} (\dot{\Delta}_{\text{Re}}^s)^2 + D_G (\ddot{\Delta}_{\text{Re}}^s)^2 \right], \\ U &= \bar{N} \left[ \alpha (\Delta_{\text{Re}}^s)^2 - \frac{\lambda}{c} \text{rot } \dot{\Delta}_{\text{Re}}^s \right], \end{aligned} \quad (62)$$

$$M_G = 8E_g^2, \quad D_G = 5/256E_g^4, \quad \alpha = \ln(E_g/E_g^*), \quad \lambda = e\pi l_{12}/4E_g m,$$

$$l_{12} = -\frac{1}{4m} \sum_{s \neq 1,2} \left[ \frac{1}{E_1 - E_s} + \frac{1}{E_2 - E_s} \right] [\mathbf{P}_{1s} \mathbf{P}_{2s}],$$

$$\bar{N} = m^{*h} E_g^h / 2\pi^2, \quad m^* = m_1 = m_2.$$

Denoting  $\mathbf{G} = l_{12} \Delta_{\text{Re}}^s$ , we obtain an expression equivalent to the phenomenological Lagrangian (30).

Thus, axial toroidal ordering of the singlet type is realized in a microscopic model with Hamiltonian (59) below the point of the transition to a state with  $\Delta_{\text{Re}}^s$ . This allows us not only to illustrate the general phenomenological scheme considered above, but also to consider some more specific properties of the system. One of the most interesting, in our opinion, is the influence of collective excitations in a system with Hamiltonian (59) on its optical and magneto-optical properties in the restructured phase. In the case of a ground state with  $\Delta_{\text{Re}}^s$  the amplitude excitations are in fact longitudinal toroidal oscillations, while the phase excitations are magnons, inasmuch as at small deviations from equilibrium we have in such a system

$$\Delta_{12}^s(t) = |\Delta(t)| \exp(i\varphi(t)) \approx |\Delta| (1 + i\varphi), \quad (63)$$

$$\Delta_{\text{Re}}^s(t) = \Delta_0 + \delta |\Delta(t)|,$$

$$\Delta_{\text{Im}}^s(t) = \Delta_0 \varphi(t), \quad (64)$$

$$\delta G(t) \propto l_{12} \delta |\Delta(t)|, \quad \delta M \propto l_{12} \Delta_0 \varphi(t),$$

where  $\mathbf{G}(t)$  is the density of the axial toroidal moment and  $\mathbf{M}(t)$  is the density of the orbital magnetic moment. Both oscillations make resonant contributions to the dielectric constant and the magnetic permeability of the system at the corresponding frequencies.

An interesting situation can arise in the case of a non-commensurate structure  $\Delta_{\text{Re}}^s$  (soliton lattice). In accordance with the general conclusions of §4, a spontaneous inhomogeneous transverse polarization  $\mathbf{P}_\perp(\mathbf{r}) \propto \text{curl } \mathbf{G}(\mathbf{r})$  is produced in the system. In the semimetal model with Hamiltonian (59) (where the Fermi energy is  $\varepsilon_F = -E_g/2$ ), in the region of the incommensurate structure<sup>28</sup> at  $T \lesssim T_G$ , where  $T_G$  is the transition temperature, we have

$$\begin{aligned} \mathbf{P}_\perp(\mathbf{r}) &= \xi \text{rot} (l_{12} \Delta_{\text{Re}}^s), \\ \xi &= -\frac{eN(0)}{4\varepsilon_F \bar{g}_{\text{Re}}}, \quad N(0) = \frac{m p_F}{2\pi^2}, \end{aligned} \quad (65)$$

$$\Delta_{\text{Re}}^s(\mathbf{r}) \approx \Delta_0 \cos \mathbf{q}_0 \mathbf{r},$$

and  $\mathbf{q}_0$  is the wave vector of the superstructure ( $\mathbf{q}_0 \rightarrow 0$  near the Lifshitz point). With decreasing temperature, one more transition can occur and produce an order parameter  $\Delta_{\text{Im}}^s(\mathbf{r})$  against the background of  $\Delta_{\text{Re}}^s(\mathbf{r})$ , with the spatial distribution of  $\Delta_{\text{Im}}^s(\mathbf{r})$  shifted by  $\pi/2$  relative to  $\Delta_{\text{Re}}^s(\mathbf{r})$  (for details see, e.g., Ref. 29):

$$\Delta_{\text{Im}}^s(\mathbf{r}) \approx \Delta_{\text{Im}}^s \sin \mathbf{q}_0 \mathbf{r}. \quad (66)$$

As already noted in Ref. 26, the appearance of  $\Delta_{\text{Im}}^s(\mathbf{r})$  in an orbital-momentum-allowed interband transition ( $l_{12} \neq 0$ ) means the onset of orbital magnetic ordering with a magnetic-moment density

$$\mathbf{M}(\mathbf{r}) \propto l_{12} \Delta_{\text{Im}}^s(\mathbf{r}). \quad (67)$$

It follows from (66) and (67) that in a system with a commensurate structure of the parameters  $\Delta_{\text{Re}}^s$  and  $\Delta_{\text{Im}}^s$  there is produced a unique ordering ("ferroelectromagnetic," a magnetic analog of ferroelectricity) in the domain-wall re-

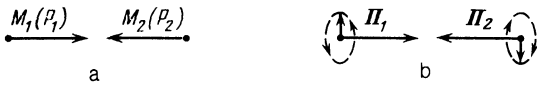


FIG. 2.

gion, with  $\mathbf{P} \perp \mathbf{M}$ ,  $\mathbf{P} \perp \mathbf{q}_0$ ,  $\mathbf{M} \perp \mathbf{q}_0$ , if  $\mathbf{q}_0 \perp \mathbf{l}_{12}$ . If, however,  $\mathbf{q}_0 \perp \mathbf{l}_{12}$ , then  $\mathbf{P} = 0$  but  $\mathbf{M}$  need not be zero, and we arrive at the case of orbital long-period ferromagnetism.

Thus various types of electronic ordering (ferroelectric, ferromagnetic, or "ferroelectromagnetic") are produced in the region of domain walls of incommensurate structures with axial toroidal moments. A detailed analysis of the regions in which various types of structure are realized call for a special treatment outside the scope of the present article.

## §6. CONCLUSION

The universality of the scheme of multipole expansions allows in principle to consider structures that are even more complicated than those discussed above. It would be of great interest, in particular, to study dipole toroidal media that feature sets of elementary toroidal dipoles  $\{ti\}$ . While seemingly exotic, such a model might be useful for the study of phase transformations in various molecular crystals.

Generally speaking, by using the scheme considered above, one can "construct" also media consisting of higher multipoles (the hierarchy of the multipole distributions is considered in Ref. 30). These include, in particular, systems with distributed fluxes of magnetic moments, characterized by a symmetric tensor  $\Pi_{ij}^{(s)}(\mathbf{r}, t)$  (see §3). We regard, however, the microscopic description and the discussion of such multipole media as premature.

In this paper we have only casually touched upon the question of the longitudinal components  $\mathbf{M}_{\parallel}$ ,  $\mathbf{P}_{\parallel}$ , and  $\mathbf{\Pi}_{\parallel}$ . Without going into details, we indicate only a geometric illustration of the distributions of these quantities (Figs. 2a and 2b): a pair of oppositely directed magnetic or electric dipoles ( $\mathbf{M}_{\parallel}$  or  $\mathbf{P}_{\parallel}$ ), and a pair of spins that precess about a common axis but in opposite directions ( $\mathbf{\Pi}_{\parallel}$ ).

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<sup>1)</sup>Not only the field equations, but also the equations of the charged-particle dynamics should be dual-invariant. The latter is achieved by introducing into the theory particles that have simultaneously electric and

magnetic charges.<sup>16</sup> If the  $e/g$  ratio is constant for particles of all type, this scheme can be reduced by a dual transformation to the usual single-charge electrodynamics.

<sup>2)</sup>A similar result in the polar toroidal state was obtained in Ref. 14 for the magnetic susceptibility.

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