Anomalous nonlinear effects at phase transitions to ferroelectric and magnetoelectric states

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A study is made of a new type of phase transition due to the onset of the magnetoelectric effect in crystals. A general phenomenological model is used to explore the anomalies which occur in the nonlinear conductivity and in several other kinetic characteristics of the system. A connection is established between the general theory of the magnetoelectric state and the microscopic model of the current state in crystals [B. A. Volkov, A. A. Gorbatsevich, Yu' V. Kopaev, and V. V. Tugushev, Sov. Phys. JETP **54**, 391 (1981)]. The kinetic Green function method is used to investigate the nonlinear effects which arise in this model under the action of external electric and magnetic fields.

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1. INTRODUCTION

Structural and electronic phase transitions can be accompanied by various anomalies in the responses to external influences and in the kinetic characteristics of the system. In the case of a proper ferroelectric the linear dielectric susceptibility diverges at a second-order phase-transition point, and the magnetic susceptibility diverges at the transition to a ferromagnetic state. The electrical conductivity σ has a kink (at a second-order transition) or a discontinuity (at a first-order transition), and so forth.

Similar anomalies occur in the case of transitions to states with charge-density waves. The state with a charge-density wave describes a structural transition, and the state with a spin-density wave describes an antiferromagnetic transition. It is usual to consider only effects of first order in the external field (i.e., linear responses and linear kinetic characteristics). In the present paper it is shown for the illustrative case of the two-band model,[1] which describes, in particular, the transition to the ferroelectric (FE) and magnetoelectric (ME) states, that the study of the nonlinear responses and kinetic characteristics yields most important information on the nature of the phase transition, especially in cases where the linear effects are not anomalous. The meaning of the term "magnetoelectric state" will be revealed in Sec. 2, where we examine the phenomenological model of this state and establish its connection to the "current" state introduced previously in Ref. 1. It is shown in Sec. 2 that the transition to the ME state is accompanied by anomalies in such physical quantities as the magnetoelectric tensor, the nonliear conductivity, and the photovoltaic tensor. These anomalies should occur in ferroelectric semiconductors under the action of electric and magnetic fields. We show that the ME state and the FE state in conducting systems are intimately interrelated, and in this sense the ME state is by no means exotic. In Secs. 3–5 we construct a microscopic model which confirms the general phenomenological deductions of Sec. 2. Using the model of Ref. 1 and the mathematical apparatus of the kinetic Green functions,[2] we derive expressions for a number of the nonlinear kinetic characteristics.

2. PHENOMENOLOGICAL THEORY OF THE MAGNETOELECTRIC STATE

It was demonstrated previously[1] in the framework of the two-band model with hybridization that a new type of state with an imaginary order parameter Δ_{Im} can arise in systems which are unstable with respect to electron–hole pairing. It is known that a state with a real order parameter Δ_{Re} corresponds to the FE phase. In the case of the FE state with $\Delta_{\mathrm{Re}} \neq 0$, $\Delta_{\mathrm{Im}} = 0$ the electric spectrum of the system remains an even function of the momentum, $\varepsilon(\mathbf{k}) = \varepsilon(-\mathbf{k})$, whereas in the case of a state with $\Delta_{\mathrm{Im}} \neq 0$ the spectrum $\varepsilon(\mathbf{k}) \neq \varepsilon(-\mathbf{k})$, i.e., the time-inversion symmetry is lost. On the other hand, in the case of a homogeneous imaginary order parameter a spontaneous current does not arise[1] in spite of the lack of time inversion, symmetry and in this sense the term "current state" introduced in Ref. 1 does not reflect the essence of the symmetry change which has occurred. In the case of an incommensurate inhomogeneous structure a spontaneous current does arise, but it is related to the imaginary order parameter in a nonlocal way, and to describe the system using the current as the order parameter one would have to consider a nonlocal free-energy functional.

Let us consider systems which suffer a loss of time inversion symmetry from a general phenomenological standpoint. Let us suppose that the spontaneous symmetry breaking in these systems is characterized by a vector order parameter. With respect to spatial transformations of the point group, vectors can be separated into polar and axial vectors (the first change sign under space inversion, while the second do not). With respect to the operation of time inversion, vectors can be either odd or even (do or do not change sign, respectively). There are thus four possible types of vector order parameters. Two of them are familiar. The polar vector **P**, which is even with respect to time inversion, is identical with the polarization vector in the theory of proper ferroelectricity; this vector thus transforms under all symmetry operations of a given crystallographic group according to one of the irreducible representations of the components of the polar vector. The axial vector **M**, which changes sign under time inversion, is identical with the mag-

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netization vector (the magnetic moment), which transforms according to an irreducible representation of the components of the axial vector of one of the magnetic symmetry groups. The other two vector order parameters in the theory of phase transitions remain practically uninvestigated. One of them—the polar vector $\boldsymbol{\pi}$—changes sign under the operation of time inversion; the other—the axial vector $\boldsymbol{\mu}$—does not. A formal symmetry classification of the four vector order parameters is given by Ascher.[3] The vector $\boldsymbol{\pi}$ transforms like the current density $\mathbf{j}$, while the vector $\boldsymbol{\mu}$ transforms like the moment current $\partial\boldsymbol{\mu}/\partial t$. In the microscopic model of Ref. 1 the state with the vector $\mathbf{P}$ is associated with the quantity $|\mathbf{P}_{12}|\Delta_{\mathrm{Re}}$ (here Δ_{Re} is the real order parameter and $\mathbf{P}_{12}$ is the interband hybridization vector, i.e., the interband matrix element of the momentum operator). The state with vector $\boldsymbol{\pi}$ is associated with the quantity $|\mathbf{P}_{12}|\Delta_{\mathrm{Im}}$, where Δ_{Im} is the imaginary order parameter. The state with vector $\mathbf{M}$ (ferromagnetism) is associated with the quantity $|\mathbf{L}_{12}|\Delta_{\mathrm{Im}}$ [here $\mathbf{L}_{12}$ is the interband matrix element of the orbital angular momentum operator).[4] Finally, the state with vector $\boldsymbol{\mu}$ is associated with the quantity $|\mathbf{L}_{12}|\Delta_{\mathrm{Re}}$. Thus, depending on the symmetry of the crystal (whether $\mathbf{P}_{12}$ or $\mathbf{L}_{12}$ is nonzero), all four types of vector order parameters are possible in the model of Ref. 1. The phase of the order parameter in this model[1] determines the properties of the system with respect to the operation of time inversion.

Structures with the vector $\boldsymbol{\mu}$ are no doubt very interesting objects of study in their own right, but in the present paper we shall deal with structures with the vector $\boldsymbol{\pi}$; for the reasons indicated just below, we shall call these "magnetoelectric" (ME) structures. For simplicity we restrict discussion to the case in which the period of the ME structure (and also of the FE structure, which we shall also consider) coincides with the initial period of the crystal lattice in the high-symmetry phase. Such a restriction enables one to work with the magnetic and crystal classes instead of the symmetry space groups. We note straightaway that the ME structures appear in the familiar symmetry classification of magnetic structures[5] (as, by the way, do structures with the vector $\boldsymbol{\mu}$), so that the vector $\boldsymbol{\pi}$ (and, analogously, $\boldsymbol{\mu}$) transforms according to one of the irreducible representations of the corresponding magnetic (crystallographic) symmetry groups. The study of ME structures whose periods do not coincide (or are entirely incommensurate) with the original period of the crystal lattice is of significant interest in its own right. This question will not be taken up here except for a single remark concerning the appearance of current in incommensurate (inhomogeneous) ME structures.

Let us consider the case in which the components of the vector $\boldsymbol{\pi}$ transform according to one of the one-dimensional irreducible representations[3] (for example, the case of uniaxial crystals, where the vector $\boldsymbol{\pi}$ is directed along the principal axis of symmetry). In this case the order parameter is the projection π_z of the vector $\boldsymbol{\pi}$ onto the corresponding axis. In principle, situations are possible in which the components of the vector $\boldsymbol{\pi}$ transform according to a two-dimensional irreducible representation (for example, if the vector $\boldsymbol{\pi}$ lies in the plane perpendicular to the principal symmetry axis of a uniaxial crystal) or three-dimensional irreducible representation (in crystals with cubic symmetry). Let us suppose that, in addition to the polar vector $\boldsymbol{\pi}$, a polar vector $\mathbf{P}$ can also spontaneously arise, i.e., there is competition between the FE and ME states (just such a situation is possible in the microscopic two-band model[1]). Let us also assume that the high-symmetry phase does not have either magentic or ferroelectric ordering, and that the system belongs to one of the nonpyroelectric classes (in uniaxial systems with tetragonal, rhombohedral, and hexagonal symmetry we exclude the classes $\mathbf{C}_n$ and $\mathbf{C}_{nv}$, $n = 3, 4, 6$). The effective equilibrium Hamiltonian U (free-energy functional) in the presence of an external electric field $\mathbf{E}$ and magnetic field $\mathbf{H}$ is of the following form in the first nonvanishing approximations:

$$U=\alpha_1 P_z^2+\alpha_2\pi_z^2-\lambda_1 P_z E_z-\lambda_2\pi_z(E_x H_y-H_x E_y)$$
$$+\beta_1 P_z^4+\beta_2\pi_z^4+\beta_3 P_z^2\pi_z^2. \qquad (1)$$

We note that the magnetic field $\mathbf{H}$ is not of itself a source of the order parameter π_z, unlike the electric field $\mathbf{E}$, which is a source of P_z. From expression (1) the physical meaning of the order parameter π_z is perfectly clear: With accuracy up to the constant λ_2, it is the antisymmetric component of the magnetoelectric tensor; this component becomes nonzero below the second-order phase-transition point ($\alpha_2 = 0$). In the general case the three components of the vector $\boldsymbol{\pi}$ are proportional to the three components of the antisymmetric magnetoelectric tensor, while the source for the vector $\boldsymbol{\pi}$ is the vector $\mathbf{S}$ with components $S_i \sim (\mathbf{E}\times\mathbf{H})_i$.

The onset of the magnetoelectric effect below the phase-transition point thus defines the meaning of the term "magnetoelectric state." The symmetry properties of this state place it in the magnetic classes which admit the magnetoelectric effect.[5,6] In our case, because of the vector character of the order parameter $\boldsymbol{\pi}$ only the antisymmetric components of the ME tensor are nonzero. A similar situation also obtains in certain ferroelectric magnets (see Ref. 7). It is easily seen, however, that the symmetry classifications of the magnetoelectric and ferroelectromagnetic states do not correspond (see Ref. 7; see also Ref. 3).

Let us dwell briefly on the connection between the ME state and the appearance of spontaneous current. In the case of a homogeneous order parameter $\boldsymbol{\pi}_0$ the symmetry generally admits the appearance of a homogeneous spontaneous current $\mathbf{j}_0 \sim \boldsymbol{\pi}_0$. However, energy considerations (Bloch's theorem) prohibit the appearance of a homogeneous spontaneous current (this is also confirmed by direct calculation in the microscopic model[1]). In other words, the coefficient of proportionality between $\mathbf{j}_0$ and $\boldsymbol{\pi}_0$ in the homogeneous case is identically equal to zero. Upon the transition to an incommensurate structure [to a state with a small wave vector $\mathbf{q}$ characterizing the spatial dependence of $\boldsymbol{\pi}(\mathbf{r})$] one must take into account in the Ginzburg-Landau functional the invariants containing the derivatives of the vector $\boldsymbol{\pi}$. The dependence of the current $\mathbf{j}_\mathbf{q}$ on $\boldsymbol{\pi}_\mathbf{q}$ in this case (with accuracy up to the first nonvanishing terms in $\mathbf{q}$) should be as follows:

$$\mathbf{j}_\mathbf{q}=\eta[\mathbf{q}\times[\mathbf{q}\times\boldsymbol{\pi}_\mathbf{q}]]\equiv\eta(\mathbf{q}(\mathbf{q}\boldsymbol{\pi}_\mathbf{q})-\mathbf{q}^2\boldsymbol{\pi}_\mathbf{q}), \qquad (2)$$

where $\eta \neq 0$ is the coefficient of proportionality. The current determined in accordance with (2) satisfies the transversality condition $\mathbf{q}\cdot\mathbf{j}_\mathbf{q} = 0$ and transforms as a polar vector, chang-

ing sign upon inversion of the time. In coordinate form

$$\mathbf{j}(\mathbf{r})=-\eta\ \mathrm{rot}\ \mathrm{rot}\ \boldsymbol{\pi}(\mathbf{r}).$$

The incommensurate ME state can therefore also be called a current state, although, as we mentioned earlier in this paper, the current cannot be the order parameter if the local form of the Ginzburg-Landau functional is to be preserved. We stress that we are discussing macroscopic currents which vary slowly on the scale of the dimensions of the unit cell (intracel currents are, of course, always present, but they cannot be described in the framework of the Ginzburg-Landau expansion). Let us discuss the role of dissipative processes in systems which undergo transitions to the FE and ME states. Although the present paper does not include a detailed study of dynamical effects, for the sake of generality let us write the Lagrangian of the model containing the parameters $\mathbf{P}_z$ and π_z in the first nonvanishing approximations.

$$\mathscr{L}=T-U,$$
$$T=A_1\left(\frac{\partial P_z}{\partial t}\right)^2+A_2\left(\frac{\partial \pi_z}{\partial t}\right)^2+A_3\left(P_z\frac{\partial \pi_z}{\partial t}-\pi_z\frac{\partial P_z}{\partial t}\right), \tag{3}$$

where T is the "kinetic" energy. We note that the nontrivial temporal "Lifshitz invariant" provides a linear dynamic coupling of P_z and π_z, intermixing the corresponding modes of oscillation. The role of this invariant in the microscopic model was mentioned in Ref. 8. We shall ignore the spatial dependence of P_z and π_z in (3). The equations of motion of the "generalized invariants" P_z and π_z are obtained by variation of Lagrangian (3):

$$\begin{aligned}-2A_1\frac{\partial^2 P_z}{\partial t^2}+2A_3\frac{\partial \pi_z}{\partial t}-\frac{\partial U}{\partial P_z}=0,\\ -2A_2\frac{\partial^2 \pi_z}{\partial t^2}-2A_3\frac{\partial P_z}{\partial t}-\frac{\partial U}{\partial \pi_z}=0.\end{aligned} \tag{4}$$

We retain in U only the terms quadratic and linear in P_z and π_z (assuming that all of the analysis is carried out above the transition point, $\alpha_1, \alpha_2>0$). Then the quantities $-\partial U/\partial P_z$ and $-\partial U/\partial \pi_z$ play the role of generalized forces in the equations of motion (4) (the term proportional to A_3 could also be attributed to generalized forces, in which case it should be included in the "potential" energy U, but we prefer to regard this term as part of the kinetic energy). In the presence of dissipative processes the "generalized forces" in (4) should be supplemented with "thermodynamic forces" F containing the Onsager coefficients.[9] Denoting these as F_1 and F_2 [for the first and second equations in (4), respectively], we have

$$F_1=\sigma_1\frac{\partial P_z}{\partial t}+\varepsilon_1(E_xH_y-H_xE_y),\qquad F_2=\sigma_2\frac{\partial \pi_z}{\partial t}+\varepsilon_2E_z. \tag{5}$$

Formulas (5) follow from the properties of the coefficients $\sigma_{1,2}$ and $\varepsilon_{1,2}$, which are proportional to the dissipative constants and change sign upon inversion of the time. Thus, in the presence of dissipation the polar vector $\mathbf{S}$ serves as a source for the vector $\mathbf{P}$ and the vector $\mathbf{E}$ serves as a source for π. Dissipative processes intermix the sources of $\mathbf{P}$ and π, and so the field $\mathbf{E}$ can now simultaneously induce the FE and ME order parameters, but the latter only insofar as the distribution function changes in the electric field. Similarly, $\mathbf{S}$ induces both order parameters, but the FE order parameter only in the case of a nonequilibrium distribution function. The terms in (5) containing σ_1 and σ_2 are the ordinary fricitional forces.

In the static case the equations of motion (4) with allowance for (5) give the self-consistency equations for the order parameters P_z and π_z. Above the phase-transition point these equations are of the form

$$\begin{aligned}-2\alpha_1P_z+\lambda_1E_z+\varepsilon_1(E_xH_y-H_xE_y)=0,\\ -2\alpha_2\pi_z+\lambda_2(E_xH_y-H_xE_y)+\varepsilon_2E_z=0.\end{aligned} \tag{6}$$

The electric current arising in the system in the presence of dissipative processes, with accuracy to terms of higher order in $\mathbf{E}$ and $\mathbf{H}$, is of the form

$$\begin{aligned}j_i(\mathbf{E},\mathbf{H})=\sigma_{ij}^{(0)}(\mathbf{E},\mathbf{H})E_j+P_z^2[\sigma_1^{(1)}E_i+\sigma_2^{(1)}\delta_{ij}\delta_{jz}E_z]\\ +\pi_z^2[\sigma_1^{(2)}E_i+\sigma_2^{(2)}\delta_{ij}\delta_{jz}E_z]+P_z\pi_z[\sigma_1^{(3)}E_i+\sigma_2^{(3)}\delta_{ij}\delta_{jz}E_z]\\ +P_z[\gamma_1^{(1)}E_zE_i+\gamma_2^{(1)}\delta_{ij}\delta_{jz}E^2]+\pi_z[\gamma_1^{(2)}E_zE_i+\gamma_2^{(2)}\delta_{ij}\delta_{jz}E^2]\\ +P_z^2[\mathbf{E}\times\boldsymbol{\eta}^{(1)}]_i+\pi_z^2[\mathbf{E}\times\boldsymbol{\eta}^{(2)}]_i+P_z\pi_z[\mathbf{E}\times\boldsymbol{\eta}^{(3)}]_i.\end{aligned} \tag{7}$$

Here $\boldsymbol{\eta}^{(1)}$, $\boldsymbol{\eta}^{(2)}$, and $\boldsymbol{\eta}^{(3)}$ are axial vectors having the components

$$\begin{aligned}\eta_i^{(1)}=\eta_1^{(1)}H_i+\eta_2^{(1)}\delta_{iz}H_z,\qquad \eta_i^{(2)}=\eta_1^{(2)}H_i+\eta_2^{(2)}\delta_{iz}H_z,\\ \eta_i^{(3)}=\eta_1^{(3)}H_i+\eta_2^{(3)}\delta_{iz}H_z,\end{aligned} \tag{8}$$

and $\sigma_m^{(n)}$, $\gamma_m^{(n)}$, and $\eta_m^{(n)}$ are the nonzero components of the tensors. The nonlinear (in the external field) components of the conductivity tensor and Hall coefficient are anomalous near phase transitions to the FE and ME states. The terms quadratic in P_z and π_z diverge as α_1^{-2} and α_2^{-2}, respectively, the terms linear in P_z and π_z diverge as α_1^{-1} and α_2^{-1}, and the terms bilinear in P_z and π_z diverge as $(\alpha_1\alpha_2)^{-1}$. We shall not write out here the awkward expressions for the conductivity and Hall-conductivity tensors.

From expression (7) one can also obtain an expression for the components of the photovoltaic tensor. Placing the system in an alternating electric field of frequency Ω can give rise to a homogeneous current[10]

$$j_i=\beta_{ijk}(\Omega)E_j^*(\Omega)E_k(\Omega). \tag{9}$$

Let us consider the case of a linearly polarized E field. The tensor $\beta_{ijk}(\Omega)$ in our model is purely antisymmetric and real:

$$\begin{aligned}\beta_{ijk}(\Omega)=P_z[\beta_1^{(1)}(\Omega)\delta_{ik}\delta_{jz}+\beta_2^{(1)}(\Omega)\delta_{jk}\delta_{iz}]\\ +\pi_z[\beta_1^{(2)}(\Omega)\delta_{ik}\delta_{jz}+\beta_2^{(2)}(\Omega)\delta_{jk}\delta_{iz}].\end{aligned} \tag{10}$$

The photovoltaic effect can thus arise at the transition to the FE or ME state. Substituting the expression for P_z and π_z from (6) into (10), we immediately find that the photoconductivity diverges as α_1^{-1} or α_2^{-1} at the transition point. Below we carry out a concrete analysis of the anomalies in the kinetic characteristics in terms of the model of Ref. 1.

3. KINETIC GREEN FUNCTIONS IN THE TWO-BAND MODEL WITH HYBRIDIZATION

Let us consider the Hamiltonian of the two-band model with coincident band extreme in momentum space:

$$H=H_0+V_i. \tag{11}$$

Here H_0 is the Hamiltonian of the ideal crystal, which was investigated previously,[1] and V_i is the interaction Hamiltonian of the electrons with the impurity; we assume that this is the leading interaction for momentum-relaxation processes. The Hamiltonian H_0 is of the form

$$H_0=\begin{bmatrix} \varepsilon_1(\hat{\mathbf{p}}-e\mathbf{A})+e\Phi & \gamma_{12}(\hat{\mathbf{p}}-e\mathbf{A})-\Delta_{12} \\ \gamma_{21}(\mathbf{p}-e\mathbf{A})-\Delta_{21} & \varepsilon_2(\hat{\mathbf{p}}-e\mathbf{A})+e\Phi \end{bmatrix}, \tag{12}$$

where $\mathbf{A}$ and Φ are the vector and scalar potentials, ε_1 and ε_2 are the energies of bands 1 and 2, and γ_{12} is the interband hybridization operator, which describes single-particle electronic transitions between bands 1 and 2. Assuming that the momentum region of interest is near the extrema of the bands, one can use the $\mathbf{k}\cdot\mathbf{P}$ approximation, and then the operator γ_{12} is of the form

$$\gamma_{12}(\hat{\mathbf{p}})=\mathbf{P}_{12}\hat{\mathbf{p}}/m, \tag{13}$$

where $\mathbf{P}_{12}$ is the interband momentum matrix element at the extremum. If the wave functions of bands 1 and 2 are real, then $\mathbf{P}_{12}=-\mathbf{P}_{21}$ is a purely imaginary vector. In principle, one can also consider more complicated functions $\gamma_{12}(\hat{\mathbf{p}})$ is the symmetry of the crystal is such that $\mathbf{P}_{12}=0$.

We note that by introducing the operator $\gamma_{12}(\hat{\mathbf{p}})$ we can also take into account effects due to single-particle interband transitions and impurity scattering involving an interband transition. The symmetry structure of the "Coulomb" and "impurity" contributions to γ_{12} are the same as that of the single-particle contribution. Upon averaging over angles we find $\langle\gamma_{12}(\hat{\mathbf{p}})\rangle=0$, i.e., hybridization is not a source of the homogeneous (in momentum space) order paramater Δ_{12} which arises below the temperature T_c on account of the phase transition.

The electron-impurity interaction Hamiltonian is

$$V_i=\sum_{\mathbf{R}_j} V(\mathbf{r}-\mathbf{R}_j), \tag{14}$$

where $\mathbf{R}_j$ are the coordinates of the impurity atom. We assume for simplicity that $V(\mathbf{r}-\mathbf{R}_j)$ are point potentials, i.e., we neglect the difference between the transport and momentum relaxation times. The matrix elements of the potentials $V(\mathbf{r}-\mathbf{R}_j)$ are

$$V_{mn}^{\mathbf{kk}'}=\int \varphi_{m\mathbf{k}}^{*}(\mathbf{r})\,V(\mathbf{r}-\mathbf{R}_i)\,\varphi_{n\mathbf{k}'}(\mathbf{r})\,d\mathbf{r}, \tag{15}$$

where m and n are the band indices, and $\mathbf{k}$ and $\mathbf{k}'$ are the quasimomenta. Clearly, the momentum region of the electrons involved in the scattering is much greater than that of the electrons involved in the formation of the order parameter Δ_{12}. We assume that the congruent regions represent only a small fraction of the Fermi surface, so that the total density of states $N_p(0)$ is much larger than the density of states $N(0)$ of the congruent regions. In this case it can be shown that in calculating the impurity-scattering amplitude one may neglect the corrections due to the onset of the order parameter Δ_{12} [these corrections are proportionally smaller by the ratio $N(0)/N_p(0)$]. Such a simplification of the problem does not qualitatively alter the results even in the case when $N(0)\sim N_p(0)$, but in view of the awkwardness of the calculations involved in "dressing" with impurity lines the vertices containing Δ_{12}, γ_{12}, and the external fields, let us restrict discussion to the case $N(0)\ll N_p(0)$. For three-dimensional systems this restriction is not excessively stringent [in contrast to the case of the one-dimensional systems which support charge-density waves and undergo dielectric transitions of the Peierls type, where the role of impurity renormalization is rather important, since $N(0)/N_p(0)=1$].

In the present paper the relationships of the parameters are assumed to be as follows. First, $\tau T_c\gg 1$ (τ is the momentum relaxation time), corresponding to low impurity concentrations. The temperature region of study is $T\gtrsim T_c$. Second, the hybridization parameter is assumed to be small in the sense that

$$|\gamma_{12}(p_F)|\ll T_c,\ \tau^{-1}.$$

This condition allows us to keep only the first nonvanishing terms containing γ_{12}. The electric field $\mathbf{E}$ is assumed small in the sense that $e\tau E v_F\ll T_c\tau^{-1}$. There is one additional specific parameter in our problem, stemming from the presence of band hybridization: $eT|\gamma_{12}|v_F E\ll\varepsilon_F T_c$. As T_c is approached from the high-temperature side, the parameter Δ_{12} induced by the external field also changes. It turns out that the external field induces a real Δ_{Re} and an imaginary Δ_{Im} part of the order parameter, so that for $|\Delta|\tau\ll 1$ we have $\Delta\sim e|\mathbf{P}_{12}|(E/T)\ln(T/T_c)$, where T_c is different for the real and imaginary parts (T_{Re} and T_{Im}, respectively). As $T\to T_c$ the quantity Δ_{12} grows, so that at some point the relation $\tau^{-1}\ll|\Delta|\ll T_c$ begins to be satisfied, and, finally, $|\Delta|\gg T_c$. This last situation will not be considered in the present paper, since it can occur only in an extremely narrow region near T_c. Calculations have been done for the other two situations ($|\Delta|\tau\ll 1$ and $\tau^{-1}\ll|\Delta|\ll T_c$).

Following Keldysh,[2] we introduce the matrix of Green functions:

$$\hat{\check{G}}_{ij}=\begin{pmatrix} 0 & \check{G}_{ij}^{R} \\ \check{G}_{ij}^{A} & \check{F}_{ij} \end{pmatrix}, \quad i,j=1,2. \tag{16}$$

The self-energy part, describing the electron-impurity interaction, in matrix form is

$$\hat{\check{\Sigma}}_{ij}=\begin{pmatrix} \Gamma_{ij} & \check{\Sigma}_{ij}^{A} \\ \check{\Sigma}_{ij}^{R} & 0 \end{pmatrix}, \tag{17}$$

and the averaging over the positions of the impurities is done by the usual "crossover" technique. The system of equations for the Green function $\hat{\check{G}}_{ij}(x,x')$, where $x=(\mathbf{r},t)$, is of the standard form.[11] Since we shall be interested only in the static homogeneous fields, it is convenient to choose the gauge of the potentials $\mathbf{A}$ and Φ in the form

$$\mathbf{A}(\mathbf{r})=\tfrac{1}{2}[\mathbf{H}\times\mathbf{r}],\quad \Phi(\mathbf{r})=-\mathbf{E}\mathbf{r}. \tag{18}$$

Accordingly, we seek the Green functions $\hat{\check{G}}_{ij}$ in the form

$$\hat{G}_{ij}(x,x')=\hat{G}_{ij}(x-x')\exp\left[ieA\left(\frac{\mathbf{r}+\mathbf{r}'}{2}\right)[\mathbf{r}-\mathbf{r}']-i\Phi\left(\frac{\mathbf{r}+\mathbf{r}'}{2}\right)[t-t']\right]. \quad (19)$$

The functions $\hat{G}_{ij}(x-x')$ depend only on the differences of the coordinates and times, and one can therefore transform to the Fourier components $\hat{G}_{ij}(\omega,\mathbf{p})$, which obey the following system of equations:

$$\left[\omega-\varepsilon_1\left(\mathbf{p}-\frac{e}{2}i\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-\frac{ie}{2}\mathbf{E}\frac{d}{d\omega}\right)+\frac{ie}{2}\mathbf{E}\frac{d}{d\mathbf{p}}\right]\hat{G}_{11}(\omega,\mathbf{p})$$
$$+\left[\Delta_{12}-\gamma_{12}\left(\mathbf{p}-\frac{ie}{2}\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-\frac{ie}{2}E\frac{d}{d\omega}\right)\right]\hat{G}_{21}(\omega,\mathbf{p})=\sigma_x+\sigma_x\hat{\Sigma}_{1j}\hat{G}_{j1}(\omega,\mathbf{p}),$$
$$\left[\omega-\varepsilon_2\left(\mathbf{p}-i\frac{e}{2}\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-\frac{ie}{2}\mathbf{E}\frac{d}{d\omega}\right)+\frac{ie}{2}\mathbf{E}\frac{d}{d\mathbf{p}}\right]\hat{G}_{21}(\omega,\mathbf{p})$$
$$+\left[\Delta_{21}-\gamma_{21}\left(\mathbf{p}-i\frac{e}{2}\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-\frac{ie}{2}\mathbf{E}\frac{d}{d\omega}\right)\right]\hat{G}_{11}(\omega,\mathbf{p})=\sigma_x\hat{\Sigma}_{2j}\hat{G}_{j1}(\omega,\mathbf{p}). \quad (20)$$

Here σ_x is the Pauli matrix in the indices of matrix (16). The equations for $\hat{G}_{22}$ and $\hat{G}_{12}$ are obtained by interchanging the indices $1\leftrightarrow 2$. In Eqs. (20) we have neglected the energy and momentum dependence of the self-energy parts Σ_{ij}. The off-diagonal (in the band indices) matrix elements Σ_{ij} can be regarded as being already included in the hybridization term (this was discussed above). By virtue of the assumptions indicated above, the nonlocal field corrections to the collision integral in the remaining intraband matrix elements can be ignored, and the matrix elements Σ_{11} and Σ_{22} themselves can be regarded as independent of Δ_{12}. Further, it is convenient to include all the terms diagonal in the band index in the "zeroth-order" Green functions on which the perturbation theory is based. We assume that $\varepsilon_i=p^2/2m_i$, where m_i is the effective mass of the electrons. The zeroth-order Green functions for $\mathbf{P}_{12}=0$, $\Delta_{12}=0$ are of the form

$$\hat{G}_{ij}^{0}(\omega,p)=\delta_{ij}\begin{pmatrix}0 & G_{ii}^{0R}(\omega,p)\\ G_{ii}^{0A}(\omega,p) & F_{ii}^{0}(\omega,p)\end{pmatrix},$$
$$G_{ii}^{0R,A}(\omega,p)=\mathcal{G}_{ii}^{0R,A}-\frac{E^2}{m_i}(\mathcal{G}_{ii}^{0R,A})^3+\frac{[\mathbf{E}\times\mathbf{H}]\mathbf{v}_i}{m_i}(\mathcal{G}_{ii}^{0R,A})^3, \quad (21)$$
$$F_{ii}^{0}(\omega,p)$$
$$=-i\exp\left(\frac{-ie\mathbf{E}\hat{\Pi}_E+ie\mathbf{H}\hat{\Pi}_H}{2}\right)[\{S_i,G_{ii}^{0A}\}-\{G_{ii}^{0R},S_i\}], \quad (22)$$
$$\mathcal{G}_{ii}^{R,A}(\omega,p)=(\omega-\varepsilon_i\pm\nu)^{-1},\qquad \varepsilon_i=\varepsilon_i(p),$$

where $\nu=(2\tau)^{-1}=c\langle V^2\rangle N_p(0)$, c is the impurity concentration,

$$\hat{\Pi}_E\{f,g\}=\frac{\partial f}{\partial\mathbf{p}}\frac{\partial g}{\partial\omega}-\frac{\partial f}{\partial\omega}\frac{\partial g}{\partial\mathbf{p}},$$
$$\hat{\Pi}_H\{f,g\}=\left[\frac{\partial f}{\partial\mathbf{p}}\times\frac{\partial g}{\partial\mathbf{p}}\right], \quad (23)$$

and the function $S_i(\omega,\mathbf{p})$ is the solution of the kinetic equation

$$\left[e\mathbf{E}\mathbf{v}_i\frac{d}{d\omega}+e(\mathbf{E}+[\mathbf{v}_i\times\mathbf{H}])\nabla_{\mathbf{p}}\right]S_i=\frac{S^0-S_i}{\tau},$$
$$S^0=2n^0(\omega)-1=-\operatorname{th}\frac{\omega}{2T}. \quad (24)$$

Only the first nonvanishing terms in powers of the magnetic field $\mathbf{H}$ are retained everywhere, but it is necessary to retain also nonlinear terms in powers of $\mathbf{E}$. Perturbation theory based on the functions $\hat{G}_{ij}^{0}$ of (21) works for $|\Delta|\tau\ll 1$. If, on the other hand, $|\Delta|\tau\gtrsim 1$, then it is necessary to use as zeroth-order functions the solutions of Eqs. (20) with nonzero Δ_{12} (recall that $|\gamma_{12}|\tau\ll 1$):

$$G_{ii}^{A(R)}=\frac{G_{ii}^{0A(R)}}{\mathrm{Det}^{A(R)}},\qquad G_{12}^{A(R)}=-\Delta_{12}\frac{G_{11}^{0A(R)}G_{22}^{0A(R)}}{\mathrm{Det}^{A(R)}},$$
$$F_{11}=\frac{F_{11}^{0}+|\Delta|^2F_{22}^{0}G_{11}^{0A}G_{11}^{0R}}{\mathrm{Det}^{A}\,\mathrm{Det}^{R}},\quad F_{12}=-\Delta_{12}\frac{F_{22}^{0}G_{11}^{0R}+F_{11}^{0}G_{22}^{0A}}{\mathrm{Det}^{A}\,\mathrm{Det}^{R}},$$
$$F_{22}=\frac{F_{22}^{0}+|\Delta|^2F_{11}^{0}G_{22}^{0A}G_{22}^{0R}}{\mathrm{Det}^{A}\,\mathrm{Det}^{R}},\quad \mathrm{Det}^{A(R)}=1-|\Delta|^2G_{11}^{0A(R)}G_{22}^{0A(R)}. \quad (25)$$

Returning to the case $|\Delta|\tau\ll 1$, which obtains when T is sufficiently far from T_c (but with $T-T_c\ll T_c$), we note that one should take into account in (22) the terms linear and quadratic in $\mathbf{E}$. It will become clear from what follows that for finding the order parameter Δ_{12} it is sufficient to include the terms linear in $\mathbf{E}$, while for evaluating the current one should also take the quadratic terms into account.

We shall henceforth be interested only in the kinetic Green functions, since it is in terms of these functions that the physical quantities Δ_{12} and current $\mathbf{j}$ are expressed. The procedure for applying the perturbation theory is given in Ref. 12. To the desired degree of accuracy we have

$$F_{12}=G_{11}^{0R}H_{12}F_{22}^{0}+F_{22}^{0}H_{12}G_{22}^{0A},\qquad F_{21}=F_{12}^{*},$$
$$F_{11}=F_{11}^{0}+G_{11}^{0R}H_{12}G_{22}^{0R}H_{21}F_{11}^{0}+G_{11}^{0R}H_{12}F_{22}^{0}H_{21}G_{11}^{0A}$$
$$+F_{11}^{0}H_{12}G_{22}^{0A}H_{21}G_{11}^{0A}. \quad (26)$$

The expression for F_{22} is obtained by interchanging $1\leftrightarrow 2$:

$$H_{12}=\frac{\mathbf{P}_{12}}{m}\left(\mathbf{p}-i\frac{e}{2}\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-i\frac{e}{2}\mathbf{E}\frac{d}{d\omega}\right)-\Delta_{12},$$
$$H_{21}=\frac{\mathbf{P}_{21}}{m}\left(\mathbf{p}-i\frac{e}{2}\left[\mathbf{H}\times\frac{d}{d\mathbf{p}}\right]-i\frac{e}{2}\mathbf{E}\frac{d}{d\omega}\right)-\Delta_{21}. \quad (27)$$

In (27) the $\mathbf{k}\cdot\mathbf{P}$ approximation was used for the hybridization γ_{12}. The order parameter is $\Delta_{12}=\Delta_{\mathrm{Re}}+i\Delta_{\mathrm{Im}}$, $|\Delta_{12}|=|\Delta|$.

4. THE ORDER PARAMETER IN AN EXTERNAL FIELD NEAR THE TRANSITION TEMPERATURE

The complex order parameter Δ_{12} can be found from the self-consistency equation

$$\Delta_{12}=\frac{1}{2}\left[g_{\mathrm{Re}}\,\mathrm{Re}\sum_{\omega,\mathbf{p}}F_{12}(\omega,\mathbf{p})+ig_{\mathrm{Im}}\,\mathrm{Im}\sum_{\omega,\mathbf{p}}F_{12}(\omega,\mathbf{p})\right]. \quad (28)$$

The exact expressions for the functions F_{12} for arbitrary relationships of the quantities τ, T, and $|\Delta|$ are extremely complicated, and the integrals in (28) cannot be evaluated. Let us study the region of temperatures T close to the transition temperatures T_{Re} or T_{Im} (in the absence of external fields). The temperatures T_{Re} and T_{Im} are regarded as given, and we shall not write out the explicit expressions for them in this paper (see, e.g., Ref. 1). In addition, we shall assume that the scattering by impurities is weak $(\tau T)^{-1}\ll 1$. In the "gapless" regime ($|\Delta|\tau\ll 1$) the equations for Δ_{Re} and Δ_{Im} separate into two independent equations in the approximation linear in Δ_{Re} and Δ_{Im}:

$$\Delta_{\mathrm{Re}}\ln\frac{T}{T_{\mathrm{Re}}}=i\left(\frac{\mathbf{P}_{12}\mathbf{E}}{m}\varphi_1+\frac{\mathbf{P}_{12}}{m}[\mathbf{E}\times\mathbf{H}]\Psi_1\right),$$
$$i\Delta_{\mathrm{Im}}\ln\frac{T}{T_{\mathrm{Im}}}=\frac{\mathbf{P}_{12}\mathbf{E}}{m}\varphi_2+\frac{\mathbf{P}_{12}}{m}[\mathbf{E}\times\mathbf{H}]\Psi_2. \quad (29)$$

Before writing out the explicit expressions for $\varphi_{1,2}$ and $\Psi_{1,2}$, let us discuss the origin of the terms on the right-hand side of (29). The kinetic Green function F_{12} is expressed, according to Sec. 3, in terms of the bare (zeroth-order) functions F^0_{ii} ($i=1,2$), which contain the electric and magnetic field. In the functions F^0_{ii} one can distinguish two types of terms:

$$F_{ii}^0=F_{ii}^{0\prime}+F_{ii}^{0\prime\prime}. \quad (30)$$

The first term in (30),

$$F_{ii}^{0\prime}=-iS_i[G_{ii}^{0A}(\omega,\mathbf{p})-G_{ii}^{0R}(\omega,\mathbf{p})], \quad (31)$$

is due to the change in the distribution function S_i in the external fields. The second term in (30) describes nonlocal field corrections.[11] In the lowest nonvanishing approximations in the fields $\mathbf{E}$ and $\mathbf{H}$ we have

$$F_{ii}^{0\prime\prime}=\left(\frac{e}{2}\mathbf{E}\mathbf{v}_i+\frac{e^2\tau}{2m_i}[\mathbf{E}\times\mathbf{H}]\mathbf{v}_i\right)\frac{dS^0}{d\omega}[(G_{ii}^{0A})^2+(G_{ii}^{0R})^2]. \quad (32)$$

Generally speaking, one should also take into account that the mass operators in the functions $G_{ii}^{0A,R}$ change with $\mathbf{E}\times\mathbf{H}$ [see formula (22)]. It can be shown, however, that in our case this effect is small [containing the parameter $(\tau T)^{-1}\ll 1$] in comparison with the kinetic contribution (31) and "nonlocal" contribution (32). The distribution function $S_i(\omega,\mathbf{p},\mathbf{E},\mathbf{H})$ is written in the form

$$S_i(\omega,\mathbf{p},\mathbf{E},\mathbf{H})=S^0(\omega)-e\tau\mathbf{E}\cdot\mathbf{v}_i\frac{dS^0}{d\omega}-\left(\frac{e\tau}{m_i}\right)^2\mathbf{E}\times\mathbf{H}\cdot\mathbf{p}\frac{dS^0}{d\omega}, \quad (33)$$

where $\mathbf{v}_i$ is the velocity and m_i the effective mass of the electrons in bands $i=1,2$. For $\Delta_{\mathrm{Re}}\tau\ll 1$ and $\Delta_{\mathrm{Im}}\tau\ll 1$ the nonlocal contributions turn out to be smaller than the kinetic contributions by a factor containing $(\tau T)^{-1}$. In the case of ideally congruent Fermi surfaces the coefficients φ_1 and φ_2 for $m_1=|m_2|$ are

$$\varphi_1=\frac{4e\tau\varepsilon_F}{3\pi T}A_2,\qquad \varphi_2=\frac{e}{2\pi T}A_2, \quad (34)$$

where from here on

$$A_s=\sum_{k=0}^{\infty}\frac{1}{(2k+1)^s}. \quad (35)$$

The coefficients Ψ_1 and Ψ_2 are strictly zero in the approximation of ideally congruent electron and hole Fermi surfaces. We therefore introduce the small quantities

$$\delta=\frac{m_1-|m_2|}{m_1+|m_2|}\ll 1,\qquad \frac{\mu}{T}\ll 1, \quad (36)$$

which describe respectively the difference of the effective masses in bands 1 and 2 and the difference of the electron and hole densities. Neglecting the corrections proportional to δ and μ/T in φ_1 and φ_2, we obtain for Ψ_1 and Ψ_2 the expressions

$$\Psi_1=-\frac{(e\tau v_F)^2}{3}\frac{\delta}{\pi T}A_2,\qquad \Psi_2=\frac{(e\tau v_F)^2}{3}\frac{\mu}{(\pi T)^2}A_3. \quad (37)$$

Thus the electric field $\mathbf{E}$ simutaneously induces real and imaginary order parameters. Here if $\mathbf{H}=0$ and $\mathbf{E}\|\mathbf{P}_{12}$, then $\Delta_{\mathrm{Im}}\ll\Delta_{\mathrm{Re}}$ by a factor containing $(\varepsilon_F\tau)^{-1}\ll 1$. However, in crossed fields for $\mathbf{E}\perp\mathbf{H}\perp\mathbf{P}_{12}$ the situation changes, and $\Delta_{\mathrm{Re}}\ll\Delta_{\mathrm{Im}}$ by a factor containing $T/\varepsilon_F\ll 1$ (it is assumed that the temperatures T_{Re} and T_{Im} are of the same order of magnitude). The induction of order parameters Δ_{Re} and Δ_{Im} by the electric and magnetic fields is essentially a kinetic effect. For φ_1, Ψ_1, and Ψ_2 this is clear simply from the proportionality of these quantities to the momentum relaxation time τ. The coefficient φ_2 does not formally contain τ, but this is only the result of our neglect of corrections of order $(\tau T)^{-1}\ll 1$ and the condition $\Delta_{\mathrm{Im}}\tau\ll 1$, $\Delta_{\mathrm{Re}}\tau\ll 1$. Equations (29) are the microscopic realization of the general phenomenological system of equations (6). To see this one need only introduce the notation $\mathbf{P}=|\mathbf{P}_{12}|\Delta_{\mathrm{Re}}$, $\boldsymbol{\pi}=|P_{12}|\Delta_{\mathrm{Im}}$. Relationships of this sort between the phenomenological quantities $\mathbf{P}$ and $\boldsymbol{\pi}$ and the order parameters Δ_{Re} and Δ_{Im} become extremely obvious if one considers a system with Hamiltonian (13) under conditions of thermodynamic equilibrium ($\nu\to 0$), when there is no change in the distribution function. In this case we have

$$\varphi_1=e\frac{2}{3}\frac{\varepsilon_F}{(\pi T)^2}A_3,\qquad \varphi_2\equiv 0,$$
$$\Psi_1\equiv 0,\qquad \Psi_2=\frac{4}{3}e^2v_F^2\frac{\mu}{(\pi T)^4}A_5. \quad (38)$$

In other words, under conditions of thermodynamic equilibrium an electric field $\mathbf{E}\|\mathbf{P}_{12}$ induces only a real order parameter Δ_{Re}, while electric and magnetic fields $\mathbf{E}\perp\mathbf{H}\perp\mathbf{P}_{12}$ induce only an imaginary order parameter. Equations (29) with coefficients (38) are the microscopic realization of system (6).

Let us now consider the case $|\Delta|\tau\gg 1$. This case can formally be realized either very close to the transition point [when the field-induced order parameter becomes large (compared to τ^{-1}) because the logarithm is small] or below the transition point, when the order parameter $|\Delta|$ is already large even without an external field. For this second case one must make the following replacements on the left-hand side of (29): Δ_{Re} or Δ_{Im} by δ_{Re} or δ_{Im}, respectively, and $\ln[T/$

$T_{\mathrm{Re}}(T_{\mathrm{Im}})$ by $2|\ln[T/T_{\mathrm{Re}}(T_{\mathrm{Im}})]|$ (the "law of two"); here δ_{Re} and σ_{Im} are the field-induced components of the order parameter. On the right-hand side of (29) the coefficients φ_i and Ψ_i depend on the total order parameters Δ_{Re} and Δ_{Im}.

To evaluate the coefficients φ_i and Ψ_i for $|\Delta|\tau \gg 1$ one must take into account the nonlocal corrections to the functions F_{ii}, which become quantities of the same order as the kinetic terms. In addition, still another order parameter arises, which we shall assume to be small: $|\Delta|^2\tau/T \ll 1$. In this case we have

$$\varphi_1 = -\frac{8}{3} e \frac{\varepsilon_F \nu}{|\Delta|^2 \pi T}\left(1-2\frac{\Delta_{\mathrm{Im}}^2}{|\Delta|^2}\right)A_2,$$

$$\varphi_2 = \frac{8}{3}\frac{\varepsilon_F \nu}{|\Delta|^2\pi T}\frac{2\Delta_{\mathrm{Re}}\Delta_{\mathrm{Im}}}{|\Delta|^2}A_2 + \frac{5}{8}\frac{e\nu}{|\Delta|\pi T}A_2, \tag{39}$$

$$\Psi_1 = \frac{2(ev_F\tau)^2\mu\tau}{(\pi T)^3}\Delta_{\mathrm{Re}}\Delta_{\mathrm{Im}}A_4 - \frac{2(ev_F)^2\delta}{\pi T|\Delta|^2}\left(1-2\frac{\Delta_{\mathrm{Im}}^2}{|\Delta|^2}\right)A_2,$$

$$\Psi_2 = -\frac{(ev_F\tau)^2\mu\tau}{(\pi T)^3}\left(1+2\frac{\Delta_{\mathrm{Im}}^2}{|\Delta|^2}\right) + \frac{(ev_F)^2\delta}{\pi T|\Delta|^4}4\Delta_{\mathrm{Re}}\Delta_{\mathrm{Im}}A_2.$$

Finally, if $(|\Delta|\tau)^{-1} \gg |\Delta|/T$, all the kinetic corrections become small and, letting $\nu \to 0$, we arrive at the purely "thermodynamic" equilibrium coefficients (38).

Thus, as $|\Delta|$ increases, the coefficients φ_i and Ψ_i change in an extremely complex way: Substantial simplifications can be made in (39) if it is assumed that only one type of order parameter is realized (this, in fact, corresponds to the case in which one of the temperatures T_{Re} or T_{Im} is much larger than the other). Let $T_{\mathrm{Re}} \gg T_{\mathrm{Im}}$. Then, assuming $\Delta_{\mathrm{Im}} \ll \Delta_{\mathrm{Re}}$, we obtain for φ_i and Ψ_i:

$$\varphi_1 = -\frac{8}{3} e \frac{\varepsilon_F \nu}{\Delta_{\mathrm{Re}}^2 \pi T}A_2, \qquad \Psi_1 = -\frac{2e^2 v_F^2 \delta}{\pi T \Delta_{\mathrm{Re}}^2}A_2. \tag{40}$$

Substituting (40) into the self-consistency equation for Δ_{Re} we find, for example, that for $\mathbf{E}\|\mathbf{P}_{12}$,

$$(\Delta_{\mathrm{Re}})_{\mathbf{E}} \sim E_z^{2/3}, \tag{41}$$

and for the case $\mathbf{E}\perp\mathbf{H}\perp\mathbf{P}_{12}$,

$$(\Delta_{\mathrm{Re}})_{\mathbf{E},\mathbf{H}} \sim (E_x H_y - E_y H_x)^{1/3}. \tag{42}$$

In the case of a purely imaginary order parameter $(T_{\mathrm{Im}} \gg T_{\mathrm{Re}})$ we have, respectively

$$(\Delta_{\mathrm{Im}})_{\mathbf{E}} \sim E_z^{1/2}, \qquad (\Delta_{\mathrm{Im}})_{\mathbf{E},\mathbf{H}} \sim (E_x H_y - E_y H_x)^{-1}. \tag{43}$$

The second formula in (43) may seem strange at first glance, since Δ_{Im} grows as the external fields become weaker. This growth, however, is limited to the region $|\Delta_{\mathrm{Im}}| \ll (T/\tau)^{1/2}$, and when this value is exceeded, Δ_{Im} begins to fall off in accordance with (38). Result (43) simply means that Δ_{Im} can behave nonmonotonically with change in field (and, generally speaking, temperature) in the vicinity of T_{Im}. A more rigorous analysis of (39) is needed to assess the applicability of our approximations in the immediate vicinity of the temperatures T_{Re} and T_{Im} for $T_{\mathrm{Re}} \sim T_{\mathrm{Im}}$. As regards the region of temperatures below the point T_{Re} (or T_{Im}), Eq. (39) is valid in this region if in the absence of field the equilibrium values of Δ_{Re} and Δ_{Im} satisfy the criteria indicated above.

5. THE CURRENT AND THE ANOMALIES OF THE NONLINEAR CONDUCTIVITY NEAR THE TRANSITION TEMPERATURE

The current operator in our model was investigated in detail in Ref. 1. It has the following form:

$$\hat{\mathbf{j}} = e\begin{vmatrix} \dfrac{\partial\varepsilon_1}{\partial\mathbf{k}} & \dfrac{1}{m}\mathbf{P}_{12} \\ \dfrac{1}{m}\mathbf{P}_{21} & \dfrac{\partial\varepsilon_2}{\partial\mathbf{k}} \end{vmatrix}. \tag{44}$$

Averaging the operator (44) over the basis of the kinetic Green functions, we obtain for the average value of the current density the expression

$$\mathbf{j} = e\sum_{\omega,\mathbf{k}} \mathrm{Sp}\,\hat{\mathbf{j}}\hat{F}, \tag{45}$$

where the spur is taken over the band indices 1,2.

In the present paper we shall consider only the case $\mathbf{H} = 0$ and $|\Delta|\tau \ll 1$ and concentrate our attention on the effects of the nonlinear conductivity [both the linear and quadratic terms in the electric field are retained in $\mathbf{j}(\mathbf{E})$]. We note that the corrections to the inear conductivity of the system in the absence of interband transitions ($\mathbf{P}_{12} = 0$) were found in Ref. 13, and the corrections to the linear Hall conductivity for $\mathbf{P}_{12} = 0$ were found in Ref. 14. These corrections are proportional to $|\Delta|^2\tau/T$ for $|\Delta|\tau \ll 1$ and to $|\Delta|/T$ for $|\Delta|\tau \gg 1$. Of course, one type of nonlinear correction to the conductivity arises simply through allowance for the dependence of Δ on E in the case $\mathbf{P}_{12} \neq 0$ [see (29)]. However, there is another specific type of nonlinear correction. It was shown previously[1] that in the absence of electric field the expression for the current $\mathbf{j}$ reduces to the total derivative with respect to momentum of a certain function, and in the case of a distribution function depending only on the energy, $\mathbf{j} = 0$. This result also obtains in the case in which there is no dissipation and the electric field $\mathbf{E}$ alters only the energy spectrum of the system. This can easily be shown for the local and nonlocal terms, since all the changes in this case affect only the functions G^R and G^A.

A contribution to the current will thus arise only to the extent that the distribution function is distorted in the electric field. Incorporating the field corrections to order E^2 in the functions $S_i(\omega,\mathbf{p})$, we obtain the desired result.

Let us consider the case $|\Delta|\tau \ll 1$. The current $\mathbf{j}$ in this case is of the form

$$\mathbf{j} = \mathbf{j}_{11} + \mathbf{j}_{22} + \mathbf{j}_{12} + \mathbf{j}_{21} + \mathbf{j}_0, \tag{46}$$

where $\mathbf{j}_0$ is the normal current in the absence of Δ_{12}. The matrix elements $\mathbf{j}_{ij}$ are given by

$$\mathbf{j}_{11} = e\sum_{\omega,\mathbf{k}} \frac{\partial\varepsilon_1}{\partial\mathbf{k}}\left[G_{11}^{0R}H_{12}G_{22}^{0R}H_{21}F_{11}^{0} + G_{11}^{0R}H_{12}F_{22}^{0}H_{21}G_{11}^{0A} + F_{11}^{0}H_{12}G_{22}^{0A}H_{21}G_{11}^{0A}\right], \tag{47}$$

$$\mathbf{j}_{12} = e\sum_{\omega,\mathbf{k}} \frac{\mathbf{P}_{12}}{m}\left(G_{22}^{0R}H_{21}F_{11}^{0} + F_{22}^{0}H_{21}G_{11}^{0A}\right),$$

and $\mathbf{j}_{22}$ and $\mathbf{j}_{21}$ are obtained by interchanging $1 \leftrightarrow 2$. The operator H_{12} was defined earlier [see (27)]. Retaining only the terms linear in $\mathbf{P}_{12}$, we obtain the following expression for the current:

$$\mathbf{j}=\mathbf{j}_{|\Delta|}+\mathbf{j}_{\mathrm{Im}}+\mathbf{j}_0, \tag{48}$$

$$\mathbf{j}_{|\Delta|}=-\sigma^0\mathbf{E}\frac{\tau|\Delta|^2}{\pi T}8A_2, \tag{49}$$

$$\mathbf{j}_{\mathrm{Im}}=-\sigma^0\mathbf{E}\frac{i\tau^2(e\mathbf{P}_{12}\cdot\mathbf{E})}{m\pi T}\Delta_{\mathrm{Im}}8A_2, \tag{50}$$

$$\mathbf{j}_0=\sigma^0\mathbf{E},\quad \sigma^0=\frac{2}{3}e^2v_F^2N(0)\tau. \tag{51}$$

In obtaining (48) it was assumed that for a momentum-independent distribution function the expression for the current reduces to the total derivative

$$\mathbf{j}=\frac{e}{m}\sum_{\mathbf{k},\omega}\frac{\partial}{\partial\mathbf{k}}[\mathbf{P}_{12}\cdot\mathbf{k}\Delta_{21}(G_{22}{}^{0R}F_{11}{}^0+F_{22}{}^0G_{11}{}^{0A}) + \mathbf{P}_{21}\cdot\mathbf{k}\Delta_{12}(G_{11}{}^{0R}F_{22}{}^0+F_{11}{}^0G_{22}{}^{0A})]\equiv 0. \tag{52}$$

Formula (52) expresses in the language of the kinetic Green functions the condition of "cancellation" of the intraband and interband components of the current (this condition was derived by another method in Ref. 1).

The term $\mathbf{j}_{|\Delta|}$ describes the ordinary contribution, proportional to $\mathbf{E}$. The term $\mathbf{j}_{\mathrm{Im}}$ is specific to systems having an imaginary order parameter and is quadratic in the electric field $\mathbf{E}$. We see immediately that $|\mathbf{j}_{\mathrm{Im}}|\ll|\mathbf{j}_{|\Delta|}|$ in weak fields, but for describing nonlinear effects the term $\mathbf{j}_{\mathrm{Im}}$ should be taken into account because of its specific symmetric structure.

Generally speaking, there exists still another current contribution, due to the imaginary order parameter. This contribution arises because the function F_{ii}^0 changes on account of the interference of the kinetic and nonlocal corrections in second order in the electric field. To find this contribution it is necessary to substitute the function $S_i(\omega,\mathbf{p})$ to first order in $\mathbf{E}$ into the nonlocal correction $F_{ii}^{0''}$. As a result, one obtains a current contribution which is small compared to (50) [it contains the small parameter $(\tau T)^{-1}$], but differing from (50) by the symmetry structure. A calculation shows that

$$\mathbf{j}_{\mathrm{Im}}{}'=-\sigma^0\frac{2ie\mathbf{P}_{12}\Delta_{\mathrm{Im}}}{m(\pi T)^3}E^2A_4. \tag{53}$$

Interestingly, the current corrections quadratic in E contain only Δ_{Im}, but not Δ_{Re}. The contribution (53) becomes important in fields $\mathbf{E}\perp\mathbf{P}_{12}$, when $\mathbf{j}_{\mathrm{Im}}=0$, while for $\mathbf{E}\|\mathbf{P}_{12}$ we have $\mathbf{j}'_{\mathrm{Im}}\ll\mathbf{j}_{\mathrm{Im}}$. Let us now discuss some of the consequences of our results. It is seen that upon the transition to a phase with an imaginary order parameter (i.e., to the magnetoelectric state), a qualitatively new effect appears—the conductivity becomes a linear function of the electric field. The third-rank tensor describing the current contribution of quadratic order in the field is of the form

$$\tilde{\sigma}_{ijk}=(\alpha\delta_{ij}|\mathbf{P}_{12}|_k+\beta|\mathbf{P}_{12}|_i\delta_{jk})\Delta_{\mathrm{Im}}. \tag{54}$$

It is easy to see that expression (54), with allowance for (50) and (53), is the microscopic realization of the general phenomenological result for the current upon the transition to the magnetoelectric state with vector $\boldsymbol{\pi}=|\mathbf{P}_{12}|\Delta_{\mathrm{Im}}$. The appearance of an imaginary component of the order parameter also leads to the onset of the photovoltaic effect. The photovoltaic-effect tensor $\lambda_{ijk}(\Omega)$ coincides for $\Omega\to 0$ (in our case for $\Omega\tau\ll 1$) with the tensor $\tilde{\sigma}_{ijk}$. We note that it is pointless to follow the usual practice[15] of separating the current into the parts due to the diagonal and off-diagonal components of the density matrix.[15] In the Keldysh technique (20) used here both contributions are taken into account automatically. We stress that the photovoltaic effect in our case is due to the momentum asymmetry of the spectrum and is already described in the relaxation-time approximation.

If we substitute the expressions for Δ_{Re} and Δ_{Im} obtained in Sec. 4 into the expressions (47)–(50) for the current, we obtain an anomalous growth of the nonlinear conductivity near the transition temperature. We note that the leading contribution to the nonlinear conductivity above the transition temperature is given by the term $\mathbf{j}_{|\Delta|}$, while the contribution of the term $\mathbf{j}_{\mathrm{Im}}$ is smaller by a factor $T\ln(T/T_{\mathrm{Im}})/\varepsilon_F^2\tau$ for $T_{\mathrm{Re}}\sim T_{\mathrm{Im}}$. The specific corrections thus begin to play an important role in the nonlinear effects due to the imaginary order parameter in the region below the transition point, where the third-rank tensor (54) and the photovoltaic-effect tensor appear.

6. CONCLUSION

The linear magnetoelectric effect in our model is a consequence of the orbital antiferromagnetic ordering with no change in the unit cell of the crystal. Here the onset of the vector order parameter gives rise to only an antisymmetric component of the magnetoelectric tensor. We note that in addition to those examined in this paper, any flux vector S_0 (heat, diffusion, etc.) can be a source of the vector order parameter $\boldsymbol{\pi}$. Unfortunately, in this case we cannot illustrate the conclusions of the phenomenological theory in the microscopic model, since for nonequilibrium processes of this sort we do not as yet have a diagram technique. The corresponding phenomenological approach requires the use of the methods of nonequilibrium thermodynamics and is beyond the scope of this paper. A direct check can be made by measuring the reaction of the system to an external flux; the induced flux should experience anomalous growth as $T\to T_{\mathrm{Im}}$.

The onset of the photovoltaic effect can also serve as evidence (albeit inconclusive) of a transition to the ME state. Actually, the ME state evidently coexists with the FE state, and to separate the effects caused by these two types of ordering is rather complicated. In any case it is clear that only a comprehensive study of "suspicious" systems will enable one to state unequivocally whether the ME state is present or not (unlike the case of the FE state, where the very presence of a polarization is conclusive).

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