

Photovoltaic effect in an orbital antiferromagnet

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A microscopic theory of the bulk photovoltaic effect (PVE) in an orbital antiferromagnet (OAF) is constructed. The PVE in an OAF is caused by the asymmetry of the spectrum of elementary excitations as a function of the quasimomentum and exists even in the Born approximation for the scattering amplitude. It is shown that if the OAF state arises as a result of a second-order phase transition, photoconductivity in the neighborhood of the transition point conforms to the Curie-Weiss law.

The photovoltaic effect (PVE) consists of the appearance of an emf in a crystal when the crystal absorbs electromagnetic radiation.¹ The anomalous (or bulk) PVE is of special interest. It differs from the ordinary PVE in that a uniform electric current appears in a uniformly illuminated specimen in the absence of a gradient of the electrochemical potential.^{2,3} For sufficiently weak fields the constant component of the electric current can be expanded in powers of the electric field

$$j_i = \sigma_{ii} E_i + \sigma_{ijk} E_j E_k + \sigma_{ijkl} E_j E_k E_l + \gamma_{ijk} E_j E_k E_l E_n + \beta_{ijk} E_j E_k E_n, \quad (1)$$

where E_l is the l th component of the constant electric field and $\tilde{E}_l(\Omega) = \tilde{E}_l^*(-\Omega)$ is the l th component of the alternating field with frequency Ω . The first three terms describe static conductivity with nonlinear corrections, the fourth term describes photoconductivity, and the last term describes the PVE itself, i.e., a photocurrent in the medium in the absence of a constant field. The tensor β_{ijk} can differ from zero only in noncentrosymmetric media.

Two basic mechanisms for the appearance of the bulk PVE have been proposed and investigated. They are associated with the existence of two components in the photovoltaic current. The first component is described by the standard expression for the electric current:

$$j_i = \frac{e}{(2\pi\hbar)^3} \int d^3k \frac{\partial \epsilon_k}{\partial k} f_k, \quad (2)$$

where f_k is the electron distribution function (the component of the density matrix that is diagonal with respect to the band indices). For a t -invariant nonmagnetic system, when the particle spectrum is symmetric as a function of the momentum ($\epsilon_k = \epsilon_{-k}$), the current (2) can be different from zero only if the distribution function contains an asymmetric component: $f_k^a = -f_{-k}^a$. The appearance of asymmetry of the distribution function as a function of the momentum in the absence of a gradient of the electrochemical potential presupposes that the principle of detailed balance breaks down, which can happen in the presence of asymmetric scattering of charge carriers in a noncentrosymmetric crystal.² In order to describe this effect it is necessary to go beyond the Born approximation in the calculation of the scattering amplitude.

The other mechanism⁴⁻⁵ is associated with the contribution of the components of the density matrix $\rho_{n'n}(\mathbf{k})$ which are off-diagonal in terms of the band indices to the current:

$$j_z = \frac{e}{m(2\pi\hbar)^3} \sum_{n \neq n'} \int d^3k \mathbf{P}_{nn'}(\mathbf{k}) \rho_{n'n}(\mathbf{k}). \quad (3)$$

Here $\mathbf{P}_{nn'}$ is the interband matrix element of the momentum. The off-diagonal current (3) arises as a result of coherent interference of real and virtual quantum transitions induced by the electromagnetic field. It is related to the displacement of charge carriers in position when a photon is absorbed.⁶ Both the diagonal and off-diagonal contributions to the current exist only in the presence of dissipation.

In this paper we describe the bulk PVE in an orbital antiferromagnet (OAF). The OAF state was obtained^{7,8} in the model of an excitonic dielectric (ED). This model describes interelectronic correlations in the limit of weak interaction. The possibility of the existence of the OAF state was also recently demonstrated in a model with strong interaction (the so-called flux phases).⁹ If the transition into the OAF state is accompanied by loss of the center of inversion, then the energy spectrum of the charge carriers in the magnetic phase is asymmetric as a function of the momentum.^{8,10} As will be shown below, because of this asymmetry the diagonal component of the photocurrent arises even in the Born approximation for the scattering amplitude. The only significant point is that the distribution function should not be a function of the energy ϵ_k (otherwise the derivative in the integrand in Eq. (2) is a total derivative and the current vanishes). As will become evident from the microscopic model developed below, if the analysis is limited only to isotropic scattering by phonons and impurities, processes in which the number of particles in the band is conserved, then the distribution function f_k remains a quasi-Fermi distribution function. Under these conditions the PVE arises only to the extent that recombination, which does not conserve the number of charge carriers in a band and results in the appearance of a flux of quasiparticles in energy space, occurs.¹¹ The photocurrent is described by the formulas (47) ($\tau_{ph} \ll \hbar/\lambda \ll \tau_r$) or (59) ($\hbar/\lambda \ll \tau_r \ll \tau_{ph}$), depending on the ratio of the characteristic electron-phonon relaxation time τ_{ph} in a band, the interband recombination time τ_r , and the time \hbar/λ of field-induced interband transitions.

The expression (48) for the photocurrent in a weak alternating field for $\tau_{ph} \ll \tau_r \ll \hbar/\lambda$ can be rewritten in the following form:

$$j_A = -\frac{ea}{3} \frac{\kappa J}{\hbar\Omega} d \frac{2\Delta_I}{\alpha_{ph} E_g} k_0^2 a^2 \mathbf{n}.$$

Here Ω is the frequency of a linearly polarized electromagnetic wave.

netic wave with intensity J , κ is the absorption coefficient of light in a semiconductor with a lattice constant a and band gap E_g , $\hbar k_0 \equiv p_0 = [m(\hbar\Omega - E_g)]^{1/2}$ is the Fermi momentum of photoexcited electrons, and n is the unit vector along the polar axis of the crystal \mathbf{P}_{12} . The representation^{2,6}

$$\mathbf{P}_{12} = i \frac{amE_g}{16\hbar} d\mathbf{n},$$

where d is a dimensionless real constant, is used for the interband matrix element of the momentum. The electron-phonon interaction parameter α_{ph} can be represented in the form

$$\alpha_{ph}^{-1} = \frac{2\hbar\tau_{ph}}{ma^2} \text{th} \frac{\omega_{ph}}{2\theta},$$

where ω_{ph} is the characteristic phonon energy and θ is the temperature. The order parameter Δ_I of the OAF state arises either as a self-consistent potential owing to the interband Coulomb interaction as a result of the phase transition (this is the case that is studied in the present work using an excitonic dielectric model) or owing to spin-orbital interaction in a spin antiferromagnet;¹² in the last case the investigated contribution to the photocurrent has an additional relativistic smallness. The possibility of the appearance of a photocurrent along \mathbf{P}_{12} is governed by the asymmetry of the OAF spectrum as a function of the quasimomentum with $\mathbf{P}_{12}\Delta_I \neq 0$. The following conditions were used in the derivation of the expressions for the photocurrent: the condition of resonance

$$\frac{\hbar\Omega - E_g}{\hbar\Omega} \ll 1,$$

the smallness of the anisotropy and asymmetry of the spectrum of the OAF

$$\frac{P_{12}p_0}{mE_g}, \frac{\Delta_I}{E_g} \ll 1,$$

and the condition

$$\theta \ll \frac{\hbar\Omega - E_g}{2}.$$

In order to compare the magnitudes of the diagonal (2) and off-diagonal (3) contributions to the photocurrent in nonmagnetic media without a center of inversion and the photocurrent \mathbf{j}_Δ in an OAF we note that the structure of the expression for \mathbf{j}_Δ is identical to the structure of the inelastic contribution to the displacement scattering photocurrent.⁶ The difference lies in the fact that in piezoelectrics the asymmetric probability W_{as} of inelastic scattering of an electron by phonons, which is proportional to α_{ph} , appears in the displacement photocurrent instead of the symmetry parameter $2\Delta_I/E_g$ of the spectrum. As a result, \mathbf{j}_2 does not depend on the phonon coupling constant, but rather is determined directly by the asymmetry parameters of the crystal potential. The diagonal photocurrent \mathbf{j}_1 in a nonmagnetic medium also contains a contribution that is proportional to the product of $W_{as} \sim \alpha_{ph}$ and the relaxation time $\tau_{ph} \sim \alpha_{ph}^{-1}$ (Refs. 6, 2). In order of magnitude this contribution is equal to \mathbf{j}_2 . Thus the ratio of \mathbf{j}_Δ in OAF to the known contributions (2) and (3) to the photocurrent in piezoelectrics in weak fields is determined by the ratio of the spectrum asymmetry param-

eter $2\Delta_I/E_g$ of the OAF to the electron-phonon interaction parameter α_{ph} . If α_{ph} in the OAF is sufficiently small, then this ratio can be of the order of or greater than unity.

The situation when the necessary conditions for the existence of the PVE appear in the system as a result of a phase transition is unique. Under nonequilibrium conditions such a transition is accompanied by the appearance of spontaneous uniform current. When some macroscopic physical quantity arises spontaneously in thermodynamic equilibrium as a result of a second-order phase transition the corresponding response function (permittivity function in the case of a ferroelectric transition and magnetic susceptibility in the case of a magnetic transition) diverges at the point of the transition. In this paper it is shown that a similar situation also occurs at a transition into the OAF state with a loss of the center of inversion under nonequilibrium conditions. In this case, near the transition point the photoconductivity conforms to the Curie-Weiss law.

1. MODEL HAMILTONIAN AND THE EQUATIONS FOR THE KINETIC GREEN'S FUNCTIONS

We shall study the two-band model of an excitonic dielectric with a singlet order parameter with arbitrary phase structure ($\Delta = \Delta_R + i\Delta_I$) in an external field

$$\mathbf{A}(t) = \mathbf{A}_0 \exp(-i\Omega t) + \text{c.c.}$$

The scheme used to describe the behavior of an excitonic dielectric in external fields will thus be suitable for analysis of the kinetic properties of both an OAF (Δ_I) (Ref. 10) and a ferroelectric based on collectivized electrons (Δ_R) (Ref. 8). We start from the Hamiltonian

$$\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_{ext}(t) + \mathcal{H}_E + \mathcal{H}_{e-ph} + \mathcal{H}_{e-imp} + \mathcal{H}_r. \quad (4)$$

Here \mathcal{H}_0 is the Hamiltonian of the electron-hole subsystem taking into account dielectric pairing:

$$\mathcal{H}_0 = \int \frac{d^3p}{(2\pi)^3} H_{0ijp} a_{ip}^+ a_{jp}, \quad \hat{H}_{0p} = \begin{pmatrix} \xi_p & W_p \\ W_p^* & -\xi_p \end{pmatrix}. \quad (5)$$

For simplicity, we assume that the electron spectrum ($\xi_p = p^2/2m + E_g/2$) and the hole spectrum ($-\xi_p$) are isotropic and have the same effective mass m . Here E_g is the band gap ($E_g > 0$ for the semiconductor model and $E_g < 0$ for the semimetal model); a_{ip}^+ are the Schrödinger operators creating an electron with quasimomentum \mathbf{p} in the i th band ($i = 1, 2$); $W_p = \gamma_p + \Delta_p$, where $\gamma_p = \mathbf{P}_{12}\mathbf{p}/m_0 = -\gamma_p^*$ is the hybridization parameter, describing the single-particle interband transitions;⁸ $\mathbf{P}_{12} \equiv i\mathbf{P}$ is the interband matrix element of the momentum in the Luttinger-Kohn basis; m_0 is the free-electron mass; and, here and below the system of units $c = \hbar = -e = 1$ is employed.

We write the Hamiltonian describing the interaction with an alternating electric field

$$\tilde{\mathbf{E}}(t) = -\frac{\partial}{\partial t} \mathbf{A}(t)$$

as follows:

$$\mathcal{H}_{ext}(t) = \int \frac{d^3p}{(2\pi)^3} \{ \hat{\lambda}_p e^{-i\Omega t} + \text{h.c.} \}_{ij} a_{ip}^+ a_{jp}, \quad (6)$$

$$\hat{\lambda}_p = \begin{pmatrix} f_p & \tilde{\Delta}_{ep} \\ \tilde{\Delta}_{ep} & -f_p \end{pmatrix}.$$

For the components of the matrix $\hat{\lambda}_p$ we have

$$f_p = \mathbf{A}_0 \frac{\partial \xi_p}{\partial \mathbf{p}}, \quad \tilde{\Delta}_{ep} = \lambda + \tilde{\Delta}_p, \quad \tilde{\tilde{\Delta}}_{ep} = \tilde{\lambda}^* + \tilde{\tilde{\Delta}}_p,$$

where f_p describes interband motion; $\lambda = \mathbf{P}_{12} \mathbf{A}_0 / m_0$ and $\tilde{\lambda} = \mathbf{P}_{12} \mathbf{A}_0^* / m_0$ describe field-induced interband transitions; and $\tilde{\Delta}_p$ and $\tilde{\tilde{\Delta}}_p$ are the components of the dielectric order parameter

$$\Delta_p(t) = \Delta_p + \tilde{\Delta}_p e^{-i\Omega t} + \tilde{\tilde{\Delta}}_p e^{i\Omega t}, \quad (7)$$

which are induced by the external field.

The term \mathcal{H}_E in the complete Hamiltonian (4) describes the interaction with a weak constant external electric field \mathbf{E} :

$$\mathcal{H}_E = i \int \frac{d^3 p}{(2\pi)^3} \mathbf{E} \frac{\partial}{\partial \mathbf{p}} \delta_{ij} a_{i\mathbf{p}}^+ a_{j\mathbf{p}}, \quad (8)$$

where $i\mathbf{E}(\partial \delta_{ij} / \partial \mathbf{p})$ is the operator of the scalar potential Φ , whose gauge in the coordinate space is chosen in the form

$$\Phi(\mathbf{r}) = -\mathbf{E} \cdot \mathbf{r}.$$

The term \mathcal{H}_{e-ph} in Eq. (4) is the intraband electron-phonon interaction Hamiltonian

$$\mathcal{H}_{e-ph} = \int \frac{d^3 p d^3 q}{(2\pi)^6} g(\mathbf{q}) \varphi(\mathbf{q}) \delta_{ij} a_{i\mathbf{p}}^+ a_{j\mathbf{p}+\mathbf{q}}, \quad (9)$$

where $g(\mathbf{q})$ is the matrix element of the electron-phonon interaction and $\varphi(\mathbf{q}) = b_{\mathbf{q}}^+ + b_{-\mathbf{q}}$, where $b_{\mathbf{q}}^+$ is the operator creating a photon with momentum \mathbf{q} .

The term \mathcal{H}_{e-imp} describes intraband electron-impurity scattering. For simplicity we assume that the impurity potential is identical for both hands:

$$\mathcal{H}_{e-imp} = \int \frac{d^3 p d^3 q}{(2\pi)^6} V_{imp}(\mathbf{q}) \delta_{ij} a_{i\mathbf{p}}^+ a_{j\mathbf{p}+\mathbf{q}}, \quad (10)$$

$$V_{imp}(\mathbf{r}) = \sum_{\mathbf{r}_a} V_{imp}^a(\mathbf{r} - \mathbf{r}_a),$$

where the summation extends over impurity atoms distributed randomly in the crystal.

Finally, the last term in Eq. (4) describes recombination processes. For what follows, it is sufficient to take into account only radiative recombination:

$$\mathcal{H}_r = \int \frac{d^3 p d^3 k}{(2\pi)^6} \Gamma_{ij}^\alpha(\mathbf{p}, \mathbf{k}) d_{\mathbf{k}\alpha} a_{i\mathbf{p}+\mathbf{k}} a_{j\mathbf{p}} + \text{h.c.}, \quad (11)$$

$$\Gamma_{ij}^\alpha(\mathbf{p}, \mathbf{k}) = \left(\frac{2\pi}{\omega_{\mathbf{k}}} \right)^{1/2} \left(\frac{\mathbf{p}}{m} \hat{\sigma}_z + \frac{i}{m_0} \mathbf{P}_{12} \hat{\sigma}_y \right)_{ij} e^\alpha.$$

where $d_{\mathbf{k}\alpha}$ is an operator that annihilates an incoherent photon with momentum \mathbf{k} and polarization given by the vector e^α ; $\omega_{\mathbf{k}}$ is the dispersion relation of the photons; and $\hat{\sigma}_{z,y}$ are the Pauli matrices with respect to the band indices.

Following Ref. 13, we introduce the matrix of kinetic Green's functions:

$$\hat{G}_{\alpha\beta} = \begin{pmatrix} \mathbf{0}, & \hat{G}' \\ \hat{G}^a, & \hat{G} \end{pmatrix}_{\alpha\beta}. \quad (12)$$

The self-energy part, which describes the scattering of charge carriers, has the following matrix form:

$$\hat{\Sigma}_{\alpha\beta} = \begin{pmatrix} \hat{\Sigma}, & \hat{\Sigma}^a \\ \hat{\Sigma}^r, & \mathbf{0} \end{pmatrix}_{\alpha\beta}. \quad (13)$$

Here and below the circumflex designates a matrix with respect to the band indices. The following system of equations can be derived for the kinetic Green's functions in an alternating field:

$$\left(i \frac{\partial}{\partial t_1} I - H_{0p} \right) \hat{G}^{a,r}(t_1, t_2) = I \delta(t_1 - t_2) + \hat{\lambda}_p(t_1) \hat{G}^{a,r}(t_1, t_2) + \int dt' \hat{\Sigma}^{a,r}(t_1, t') \hat{G}^{a,r}(t', t_2), \quad (14)$$

$$\left(i \frac{\partial}{\partial t_1} I - H_{0p} \right) \hat{G}(t_1, t_2) = \hat{\lambda}_p(t_1) \hat{G}(t_1, t_2) + \int dt' [\hat{\Sigma}(t_1, t') \hat{G}^a(t', t_2) + \hat{\Sigma}^r(t_1, t') \hat{G}(t', t_2)]. \quad (15)$$

Here \hat{I} is the unit matrix, and

$$\hat{\lambda}_p(t) = \hat{\lambda}_p \exp(-i\Omega t) + \text{h.c.}$$

In the absence of an external field the solutions of Eqs. (14) and (15) can be represented in the following form:¹⁴

$$\hat{G}_0^{a,r} = \frac{\hat{x}}{\omega - \varepsilon \mp i\nu} + \frac{\hat{y}}{\omega + \varepsilon \mp i\nu} \equiv \hat{x} G_x^{a,r} + \hat{y} G_y^{a,r}, \quad (16)$$

$$G_0 = \hat{S} \hat{G}_0^a - \hat{G}_0^r \hat{S}, \quad \hat{S} = \hat{S}_1(\omega, \mathbf{p}) \hat{x} + \hat{S}_2(\omega, \mathbf{p}) \hat{y}.$$

In these relations

$$\varepsilon \equiv \varepsilon_p = (\xi_p^2 + |W_p|^2)^{1/2}$$

is the dispersion relation for the Hamiltonian of an excitonic dielectric (5), and the matrices $\hat{x}, \hat{y} \equiv \hat{x}_p, \hat{y}_p$ are constructed from the coefficients of the canonical transformation that diagonalizes the Hamiltonian (5):

$$\hat{x}_p = \hat{I} - \hat{y}_p = \begin{pmatrix} u_p^2, & -u_p v_p \\ -u_p v_p, & |v_p|^2 \end{pmatrix}, \quad (17)$$

$$u_p^2, \quad |v_p|^2 = \frac{\varepsilon_p \pm \xi_p}{2\varepsilon_p}, \quad u_p v_p = -\frac{W_p}{2\varepsilon_p}, \quad v_p = |v_p| \frac{W_p}{|W_p|}.$$

We note the following obvious properties of the matrices \hat{x} and \hat{y} :

$$\hat{x}_p^2 = \hat{x}_p, \quad \hat{y}_p^2 = \hat{y}_p, \quad \hat{x}_p \hat{y}_p = 0, \quad \text{Sp } \hat{x}_p = \text{Sp } \hat{y}_p = 1.$$

The momentum distribution functions $S_{1,2}(\mathbf{p}) = 2n_{1,2}(\mathbf{p}) - 1$ can be obtained from the relations

$$S_{1(2)}(\mathbf{p}) = \text{Sp} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \hat{x}_p(\hat{y}_p) G_0(\mathbf{p}, \omega), \quad (18)$$

Finally, in order to take into account the damping in the equilibrium Green's functions (16) we introduce the parameter $\nu \sim (2\tau)^{-1}$, which in the simplest case of scattering by impurities (10) can be assumed to be constant:^{8,14}

$$\nu = (2\tau_{imp})^{-1} = 1/2 n_{imp} V_{imp}^2 N(0).$$

where n_{imp} is the concentration of scattering centers and $N(0)$ is the density of states at the Fermi level. We note that if damping is neglected, the equality (18) holds identically. If, however, $\nu \neq 0$, then Eq. (18) holds to within the parameter $(\tau((E_g/2)^2 + |\Delta|^2)^{1/2})^{-1}$ in the semiconductor model

or the parameter $(\tau|\Delta|)^{-1}$ in the semimetal model. We assume that this parameter is small.

Averaging the current operator with respect to the basis of the kinetic Green's functions, we obtain for the average current density the expression

$$\mathbf{j}(t) = i \text{Sp} \int \frac{d^3 p d\omega}{(2\pi)^4} \hat{\mathbf{j}}(\mathbf{p}, t) \hat{G}(\mathbf{p}, \omega, t), \quad (19)$$

where $\hat{\mathbf{j}}(\mathbf{p}, t)$ is the current operator, whose time dependence is related to the dependence of the Hamiltonian on the vector potential of the electromagnetic field in the Luttinger-Kohn representation:

$$\hat{\mathbf{j}}(\mathbf{p}, t) = \frac{\partial \hat{H}_{0p}}{\partial \mathbf{p}} + \frac{\partial \hat{\lambda}_p}{\partial \mathbf{p}} e^{-i\omega t + \text{h.c.}}, \quad (20)$$

$\hat{G}(\mathbf{p}, \omega, t)$ is the Fourier transform of the kinetic Green's functions $\hat{G}(\mathbf{p}, t_1, t_2)$ (12) with respect to the time difference $t_1 - t_2$ ($t = (t_1 + t_2)/2$). We represent analogously the self-consistency equation for the dielectric order parameter in the following form:

$$\Delta_p(t) = \frac{i}{2} \int \frac{d^3 k d\omega}{(2\pi)^4} V(\mathbf{p}-\mathbf{k}) G_{12}(\mathbf{k}, \omega, t). \quad (21)$$

Where $V(\mathbf{p}-\mathbf{k})$ is the interband Coulomb interaction potential. Thus the presence of the oscillating components in the order parameter (7) is related to the fact that in an external field with frequency Ω the function $\hat{G}(\mathbf{p}, \omega, t)$ can be represented in the form of the series

$$\hat{G}(\mathbf{p}, \omega, t) = \hat{G}_p(\omega) + \hat{G}_p'(\omega) e^{-i\omega t} + \hat{G}_p''(\omega) e^{i\omega t} + \dots \quad (22)$$

and analogously for the functions $\hat{G}^{a,r}(\mathbf{p}, \omega, t)$.

The functions $S_{1,2}(\mathbf{p})$ in equilibrium, which are determined from Eq. (18), are quasi-Fermi distribution functions for the quasiparticles of an excitonic dielectric. Under non-equilibrium conditions, it is convenient to employ for the kinetic function $\hat{G}(\mathbf{p}, t_1, t_2)$ the representation proposed in Ref. 15:

$$\hat{G}(\mathbf{p}, t_1, t_2) = \int dt' [\hat{S}(\mathbf{p}, t_1, t') \hat{G}^a(\mathbf{p}, t', t_2) - \hat{G}^r(\mathbf{p}, t_1, t') \hat{S}(\mathbf{p}, t', t_2)]. \quad (23)$$

Therefore, a representation of the type (22) is also valid for the function $\hat{S}(\mathbf{p}, \omega, t)$. From the expression (23) we obtain for the terms in the series (22)

$$\begin{aligned} \hat{G}_p(\omega) &= \hat{S}_p(\omega) \hat{G}_p^a(\omega) + \hat{S}_p'(\omega - \Omega/2) \hat{G}_p^{a''}(\omega - \Omega/2) \\ &\quad + \hat{S}_p''(\omega + \Omega/2) \hat{G}_p^{a'}(\omega + \Omega/2) - [\hat{G}^r \hat{S}], \quad (24) \\ \hat{G}_p'(\omega) &= \hat{S}_p(\omega + \Omega/2) \hat{G}_p^{a'}(\omega) + \hat{S}_p'(\omega) \hat{G}_p^a(\omega - \Omega/2) - [\hat{G}^r \hat{S}], \\ \hat{G}_p''(\omega) &= \hat{S}_p(\omega - \Omega/2) \hat{G}_p^{a''}(\omega) + \hat{S}_p''(\omega) \hat{G}_p^a(\omega + \Omega/2) - [\hat{G}^r \hat{S}]. \end{aligned}$$

The bracketed terms in Eqs. (24) are obtained by writing the functions out explicitly by permuting the indices. The function $\hat{S}(\mathbf{p}, \omega, t)$ is determined from the kinetic equation. It can be shown that the oscillating components of the distribution functions \hat{S}' and \hat{S}'' are small, on the order of the parameter $\lambda/\Omega \ll 1$. For this reason, it is sufficient to seek the time-independent component of the function $\hat{G}(\mathbf{p}, \omega, t)$ (22) in the form (16) with

$$\hat{S} \equiv \hat{S}_p(\omega) = S_{1p}(\omega) \hat{x}_p + S_{2p}(\omega) \hat{y}_p, \quad (25)$$

where $S_{1,2p}(\omega)$ are as yet unknown distribution functions.

Treating as a perturbation the Hamiltonian (6) describing the interaction with an alternating external field we can write down the expansion in powers of $\hat{\lambda}$ for the oscillating components for the function \hat{G} . Separating the resonant part, containing the functions $G_x^{a,r}(\omega + \Omega)$ and $G_y^{a,r}(\omega - \Omega)$ [in the notation of Eq. (16)], which can be anomalously large at $\Omega \sim \varepsilon$, we obtain, for example, for \hat{G}_p' (the methodological details are presented in Ref. 14):

$$\begin{aligned} \hat{G}_p'(\omega) &= \hat{G}_0(\omega + \Omega) \hat{\lambda} \hat{G}_0^a(\omega) + \hat{G}_0^r(\omega + \Omega) \hat{\lambda} \hat{G}_0(\omega) + \dots \\ &= \frac{\hat{A} + \Lambda}{|1 - \eta|^2} [G_x^r(\omega + \Omega) G_y(\omega) + G_x(\omega + \Omega) G_y^a(\omega)]. \quad (26) \end{aligned}$$

Here the argument \mathbf{p} is omitted in order to simplify the equations and the following notation was introduced:

$$\hat{A} = \begin{pmatrix} uv, & -v^2 \\ u^2, & -uv \end{pmatrix}, \quad \eta = |\Lambda|^2 G_x^r(\omega + \Omega) G_y^r(\omega), \quad (27)$$

$$|\Lambda|^2 = \text{Sp}(\hat{\lambda} \hat{y} \hat{\lambda}^+ \hat{x}), \quad \Lambda = 2uvf + \tilde{\Delta}_e u^2 - \tilde{\Delta}_e v^2.$$

We note that the parameter Λ_p is essentially the renormalization of the matrix element of the interband dipole transition λ (6) owing to coherence factors of the excitonic dielectric and can be written in the form

$$\Lambda = 2i \left(v \frac{\partial u}{\partial \mathbf{p}} - u \frac{\partial v}{\partial \mathbf{p}} \right) \mathbf{A}_0 \equiv \frac{1}{m_0} \mathbf{P}_{12}(\mathbf{p}) \mathbf{A}_0, \quad (28)$$

where $\mathbf{P}_{12}(\mathbf{p})$ is the \mathbf{p} -dependent interband matrix element of the momentum in the basis in which the Hamiltonian of the excitonic dielectric (5) is diagonal.

2. ANOMALOUS PHOTOVOLTAIC EFFECT IN AN ORBITAL ANTIFERROMAGNET: WEAK-FIELD CASE

Using the notation introduced above we obtain from Eqs. (19) and (20) the following expression for the constant component of the current:

$$\begin{aligned} \mathbf{j} = i \text{Sp} \int \frac{d^3 p d\omega}{(2\pi)^4} \left\{ \frac{\partial \hat{H}_{0p}}{\partial \mathbf{p}} \hat{G}_p(\omega) + \frac{\partial \hat{\lambda}_p}{\partial \mathbf{p}} \hat{G}_p''(\omega) \right. \\ \left. + \frac{\partial \hat{\lambda}_p^+}{\partial \mathbf{p}} \hat{G}_p'(\omega) \right\}. \quad (29) \end{aligned}$$

We first study the case of a weak electromagnetic field: $\Lambda\tau \ll 1$, where τ is the shortest relaxation time. The field can be taken into account by means of perturbation theory. We note that in the weak-field limit the damping ν must be retained in the denominators of the Green's functions $\hat{G}_0^{a,r}$ and \hat{G}_0 in the expansion (26). In the limit $\nu \rightarrow 0$ all terms in the perturbation series in Λ are found to be of the same order of magnitude; this was not pointed out in Refs. 16 and 17. The limit $\nu \rightarrow 0$ actually means a transition to the strong-field limit, and in this case the entire perturbation series must be summed. Substituting into the formula (29) expansions of the type (26) for the kinetic Green's functions we obtain the following expressions for the constant component of the current (PVE) to second order in the field:

$$\mathbf{j} = - \int \frac{d^3p}{(2\pi)^3} (S_{1p} - S_{2p}) \left\{ \frac{\partial \varepsilon_p}{\partial \mathbf{p}} - \frac{\partial \varepsilon_p}{\partial \mathbf{p}} \frac{|\Lambda_p|^2}{(\varepsilon_p - \Omega/2)^2 + \nu^2} + \frac{\partial |\Lambda_p|^2}{\partial \mathbf{p}} \frac{\varepsilon_p - \Omega/2}{(\varepsilon_p - \Omega/2)^2 + \nu^2} + i\pi\delta(2\varepsilon_p - \Omega) \left[\Lambda_p \frac{\partial \Lambda_p^*}{\partial \mathbf{p}} - \Lambda_p^* \frac{\partial \Lambda_p}{\partial \mathbf{p}} + 2|\Lambda_p|^2 \left(v_p \cdot \frac{\partial v_p}{\partial \mathbf{p}} - v_p \frac{\partial v_p^*}{\partial \mathbf{p}} \right) \right] \right\}. \quad (30)$$

The second and third terms in braces correspond to renormalization of the diagonal part of the current, i.e., the first term [compare with Eq. (2)], owing to the interaction with the field. As one can see from Eq. (30), these terms are smaller by $(\Lambda\tau)^2$ than the first time. If the damping ν is neglected but the change in the distribution functions owing to renormalization of the spectrum in second-order perturbation theory in Λ (optical Stark effect) is taken into account

$$S_i(\varepsilon_p + \Delta\varepsilon_p) = S_i(\varepsilon_p) + \frac{\partial S_i(\varepsilon_p)}{\partial \varepsilon_p} \Delta\varepsilon_p, \quad \Delta\varepsilon_p = \frac{2|\Lambda_p|^2}{2\varepsilon_p - \Omega},$$

then the indicated terms reduce to a total derivative with respect to the momentum

$$\frac{\partial}{\partial \mathbf{p}} \left(\frac{|\Lambda_p|^2 (S_{1p} - S_{2p})}{\varepsilon_p - \Omega/2} \right)$$

and the corresponding contribution to the current (30) is identically equal to zero. The last term in the braces in Eq. (30) corresponds to the displacement PVE (3), studied in Refs. 3-6 in the Bloch basis. We note that in this technique the field-induced renormalization of the current operator is taken into account in a natural manner [second and third terms in Eq. (20)]; these terms were not taken into account in Refs. 3-6.

Below we study only the diagonal component of the current that is associated with the first term in Eq. (30). We take into account only the interactions in which the number of quasielectrons and quasiholes is conserved [such interactions include the intraband scattering processes (9) and (10) and radiative recombination (11)]. Then the expression for the photocurrent assumes the form

$$\mathbf{j} = -2 \int \frac{d^3p}{(2\pi)^3} \frac{\partial \varepsilon_p}{\partial \mathbf{p}} S_{1p}. \quad (31)$$

It is immediately evident that the photocurrent is different from zero if the distribution function S_{1p} is different from the quasi-Fermi distribution function or if the integrand in Eq. (31) contains a component that is symmetric as a function of the momentum. The last condition is realized in OAF, where the spectrum of the Hamiltonian (5)

$$\varepsilon_p = (\xi_p^2 + |\gamma_p + \Delta_I|^2)^{1/2}$$

is asymmetric as a function of the momentum ($\varepsilon_p \neq \varepsilon_{-p}$).

The derivation of the kinetic equation for determining S_{1p} in our model is standard.¹³ We add the equation for the kinetic Green's function \hat{G} (15) and the Hermitian conjugate equation. The result is

$$i \frac{\partial}{\partial t} \hat{G}(t_1 - t_2, t) + \hat{G}(t_1 - t_2, t) \hat{H}_{0p} - \hat{H}_{0p} \hat{G}(t_1 - t_2, t) = \hat{\lambda}_p(t_1) \hat{G}(t_1, t_2) - \hat{G}(t_1, t_2) \hat{\lambda}_p(t_2) + \int dt' [\hat{\Sigma}(t_1, t') \hat{G}^a(t', t_2) + \hat{\Sigma}^r(t_1, t') \hat{G}(t', t_2) - \hat{G}^r(t_1, t') \hat{\Sigma}(t', t_2) - \hat{G}(t_1, t') \hat{\Sigma}^a(t', t_2)], \quad t = (t_1 + t_2)/2. \quad (32)$$

The kinetic equation is obtained from Eq. (32) with the help of the relation (18). The first two terms on the right-hand side of Eq. (32) correspond to the source in the collision integral, describing field-induced interband transitions, and the integral terms describe scattering by phonons and impurities and radiative recombination. In the stationary case the kinetic equation has the form

$$\left(\frac{\partial S_{1,2p}}{\partial t} \right)_{cm} = \left(\frac{\partial S_{1,2p}}{\partial t} \right)_{ph} + \left(\frac{\partial S_{1,2p}}{\partial t} \right)_{imp} + \left(\frac{\partial S_{1,2p}}{\partial t} \right)_r, \quad \left(\frac{\partial S_{1,2p}}{\partial t} \right)_{ext} = 0. \quad (33)$$

Substituting the expressions for the self-energy matrices in the Born approximation into the integral term in Eq. (32) and using Eqs. (16) and (18), we obtain for the electron-phonon collision integral

$$\left(\frac{\partial S_{1,2p}}{\partial t} \right)_{ph} = \pi \int \frac{d^3k}{(2\pi)^3} g^2(\mathbf{p}-\mathbf{k}) S_p(\hat{x}_p, \hat{x}_k) \times \{ [(1+N_{\mathbf{p}-\mathbf{k}})(S_{1,2\mathbf{k}} \mp 1)(1 \pm S_{1,2\mathbf{p}}) + N_{\mathbf{p}-\mathbf{k}}(S_{1,2\mathbf{k}} \pm 1)(1 \mp S_{1,2\mathbf{p}})] \delta(\varepsilon_p - \varepsilon_k - \omega_{\mathbf{p}-\mathbf{k}}) + [(1+N_{\mathbf{p}-\mathbf{k}})(S_{1,2\mathbf{k}} \pm 1)(1 \mp S_{1,2\mathbf{p}}) + N_{\mathbf{p}-\mathbf{k}}(S_{1,2\mathbf{k}} \mp 1)(1 \pm S_{1,2\mathbf{p}})] \delta(\varepsilon_p - \varepsilon_k + \omega_{\mathbf{p}-\mathbf{k}}) \}, \quad (34)$$

where $N_p = (\exp(\omega_p/\theta) - 1)^{-1}$ is the Planck distribution function of the phonons. The term describing the recombination of quasiparticles through the gap is omitted in the collision integral. This is valid when $((E_g/2)^2 + |\Delta|^2)^{1/2} > \omega_{ph}$ in the semiconductor model or $|\Delta| > \omega_{ph}$ in the semi-metal model, where ω_{ph} is the characteristic energy of the phonons. The collision integral Eq. (34) vanishes for the quasi-Fermi distribution functions:

$$S_{1p}(\varepsilon_p) = -S_{2p}(-\varepsilon_p) = \text{th} \frac{\mu - \varepsilon_p}{2\theta}. \quad (35)$$

The first equality in Eq. (35) reflects the fact that the number of quasielectrons is equal to the number of quasiholes. The chemical potential μ in Eq. (35) can be arbitrary when recombination is neglected.

Analogously, the collision integral for intraband electron-impurity scattering has the form

$$\left(\frac{\partial S_{1,2p}}{\partial t} \right)_{imp} = 2\pi n_{imp} \int \frac{d^3q}{(2\pi)^3} |V_{imp}(\mathbf{p}-\mathbf{q})|^2 S_p(\hat{x}_p, \hat{x}_q) \times (S_{1,2q} - S_{1,2p}) \delta(\varepsilon_p - \varepsilon_q) \quad (36)$$

and vanishes for arbitrary energy distribution functions $S_{1,2p}(\pm \varepsilon_p) = S_{1,2}(\pm \varepsilon_p)$. In particular, it vanishes for the distributions (35).

Finally, the recombination collision integral can be written in the following form:

$$\begin{aligned} \left(\frac{\partial S_{1,2p}}{\partial t}\right)_r &= \pi \sum_{\alpha} \int \frac{d^3k}{(2\pi)^3} \text{Sp}(\hat{x}_p \hat{\Gamma} \hat{x}_p \hat{\Gamma}^+) (S_{2,1p} \mp 1) \\ &\quad \times (1 \pm S_{1,2p}) \delta(2\varepsilon_p - \omega_k) \\ &= \frac{1}{4\tau_r(\mathbf{p})} (S_{2,1p} \mp 1) (1 \pm S_{1,2p}). \end{aligned} \quad (37)$$

where the matrix element $\hat{\Gamma} \equiv \hat{\Gamma}^{\alpha}(\mathbf{p}, \mathbf{k})$ is defined in Eq. (11) and we have introduced the recombination time

$$\begin{aligned} \tau_r^{-1}(\mathbf{p}) &= \frac{4}{3} \Omega \left| \frac{\partial \xi_p}{\partial \mathbf{p}} \cdot 2u_p v_p + \frac{\mathbf{P}_{12}}{m_0} (u_p^2 + v_p^2) \right|^2 \\ &= \frac{4}{3} \Omega \left| \frac{\partial \Lambda_p}{\partial \mathbf{A}_0} \right|^2, \end{aligned} \quad (38)$$

which is inversely proportional to the probability of emission of an incoherent photon in an interband transition. The integral (37) vanishes if

$$S_{1p}(\varepsilon_p) = -1, \quad S_{2p}(-\varepsilon_p) = 1. \quad (39)$$

In this case, $n_{1p} = 0$ and $n_{2p} = 1$, i.e.,

$$\left(\frac{\partial S_{1,2p}}{\partial t}\right)_r = 0$$

in the case of a completely filled lower band and a completely empty upper band, which happens for very strong recombination, when the recombination time τ_r is much shorter than all other characteristic relaxation times in the system.

It is useful to calculate directly the source in the collision integral [the last term in Eq. (33)] for an arbitrary value of the parameter Λ without treating the field as a perturbation. We separate from the expression for the source in Eq. (32) the stationary part:

$$I_{ext}(\omega) = \lambda \hat{G}''(\omega) + \hat{\lambda}^+ \hat{G}'(\omega) - \hat{G}'(\omega - \Omega) \hat{\lambda}^+ - \hat{G}''(\omega + \Omega) \hat{\lambda}.$$

Using the relations (26) and (18) we obtain in the limit $\nu \rightarrow 0$:

$$\begin{aligned} \left(\frac{\partial S_1}{\partial t}\right)_{ext} &= - \left(\frac{\partial S_2}{\partial t}\right)_{ext} \\ &= \lim_{\nu \rightarrow 0} \frac{2\nu |\Lambda|^2 [S_2(-\Omega/2 - E) - S_1(\Omega/2 - E)]}{4E^2 + \nu^2}. \end{aligned} \quad (40)$$

In Eq. (40) we have introduced the function E_p defined as

$$E_p = [(\varepsilon_p - \Omega/2)^2 + |\Lambda_p|^2]^{1/2}, \quad (41)$$

which is the dispersion relation for quasiparticles in an excitonic dielectric in a strong electromagnetic field (see below). Passing to the limit $\nu \rightarrow 0$ we have

$$\begin{aligned} \left(\frac{\partial S_1}{\partial t}\right)_{ext} &= \begin{cases} 0, & \Lambda\tau \gg 1, \\ 2\pi |\Lambda_p|^2 [S_{2p}(\varepsilon_p - \Omega) - S_{1p}(\varepsilon_p)] \delta(\Omega - 2\varepsilon_p), & \Lambda\tau \ll 1. \end{cases} \end{aligned} \quad (42)$$

Thus in a strong field ($\Lambda\tau \gg 1$), when the renormalization of the spectrum is significant, there is no source in the kinetic equation. In a weak field ($\Lambda\tau \ll 1$) one can see from Eq. (42) that the source vanishes if $S_{1p}(\varepsilon_p) = S_{2p}(\varepsilon_p - \Omega)$. If the distribution function is chosen to be of the form Eq. (35), then the condition that the source (42) be equal to zero determines the chemical potential $\mu = \Omega/2$. As a result we find that when recombination is neglected the functions of the

form

$$S_{1p}(\varepsilon_p) = -S_{2p}(-\varepsilon_p) = \text{th} \frac{\Omega/2 - \varepsilon_p}{2\theta} \quad (43)$$

represent stationary distributions of quasiparticles in an excitonic dielectric in a weak field with frequency Ω , slightly exceeding the gap width [$(\Omega - |\Delta|)/\Omega \ll 1$ for the semimetal model or $(\Omega - E_g)/\Omega \ll 1$ for the semiconductor model; this condition made it possible to separate the resonance part of the series (26)]. The physical meaning of the distribution (43) is obvious: The external field transfers quasiparticles into the level $\varepsilon_p = \Omega/2$ in the upper band (the energy is measured from the center of the forbidden band) and then, owing to intraband relaxation processes, the quasiparticles fill states near the bottom of the band up to this level (the so-called "saturation state"¹¹).

It follows from Eq. (31) that the photocurrent vanishes for quasi-Fermi distribution functions (43) [this result is also true for the off-diagonal component of the photocurrent (30)]. The distribution function differs from the quasi-Fermi function and the photocurrent is nonzero only to the extent that recombination occurs, since the complete collision integral (33), including the recombination term, no longer vanishes for the distribution function (43). We note, however, that in the hypothetical limit of very strong recombination, when $\tau_r \ll \tau_{imp}, \tau_{ph}$ holds, there is no photocurrent in a weak field. The solution of the kinetic equation (39) shows that under these conditions the system simply does not contain any free carriers that could participate in the formation of the current. As will be shown below, the situation is different in the case of a strong field: A photocurrent is possible in the presence of strong recombination.

We consider only the electron-phonon mechanism of intraband relaxation. We seek an expression for the distribution function, differing from the quasi-Fermi function, from the kinetic equation $(\partial S_{1p}/\partial t)_{st} = 0$, including the electron-phonon (34) and recombination (37) collision integrals and the source from the external field (42). If recombination is weak, then owing to intraband relaxation a quasiequilibrium distribution is rapidly established for the created quasiparticles. In this case recombination merely shifts the Fermi level from $\Omega/2$ (43) by some amount η_p , so that

$$S_{1p}(\varepsilon_p) = \text{th} \frac{\Omega/2 - \varepsilon_p - \eta_p}{2\theta}. \quad (44)$$

Let $\eta_p \sim \tau_r^{-1} \ll \theta \ll \Omega/2 - \varepsilon_{min}$, $\varepsilon_{min} = [(E_g/2)^2 + |\Delta|^2]^{1/2}$. Then the perturbation theory in the parameter $(\theta\tau_r)^{-1} \sim \eta_p/\theta \ll 1$ can be used to solve the kinetic equation. The equation for the function η_p then has the form

$$\begin{aligned} &\int \frac{d^3k}{(2\pi)^3} g^2(\mathbf{p}-\mathbf{k}) \text{Sp}(\hat{x}_p \hat{x}_k) \\ &\quad \times \frac{(\eta_p - \eta_k) [\delta(\varepsilon_p - \varepsilon_k - \omega_{p-k}) + \delta(\varepsilon_p - \varepsilon_k + \omega_{p-k})]}{\text{ch} \frac{\Omega/2 - \varepsilon_p}{2\theta} \text{ch} \frac{\Omega/2 - \varepsilon_k}{2\theta} \text{sh} \frac{\omega_{p-k}}{2\theta}} \\ &+ \frac{4\nu}{\pi} \frac{|\Lambda_p|^2 \eta_p \text{ch}^{-2} \frac{\Omega/2 - \varepsilon_p}{2\theta}}{(2\varepsilon_p - \Omega)^2 + \nu^2} = \frac{\theta}{2\pi\tau_r(\mathbf{p})} \left(1 + \text{th} \frac{\Omega/2 - \varepsilon_p}{2\theta}\right)^2. \end{aligned} \quad (45)$$

We seek a solution of Eq. (45) which is approximately linear

in the hybridization in the form

$$\eta_{\mathbf{p}} = \eta + \alpha(\varepsilon_{\mathbf{p}}^0) \Delta_I |\gamma_{\mathbf{p}}|, \quad (46)$$

where $\gamma_{\mathbf{p}} = i\mathbf{P}\mathbf{p}/m_0$ is the hybridization and $\alpha(\varepsilon_{\mathbf{p}}^0)$ is the unknown isotropic spectrum function ($\varepsilon^0 = \varepsilon(\gamma = 0)$). For simplicity we replace the matrix element of the electron-phonon interaction by a constant: $g^2(\mathbf{p}) = g^2/p_0$, where $\varepsilon_{p_0}^0 = \Omega/2$. In addition, we replace the acoustic phonon spectrum $\omega_{\mathbf{p}}$ by the characteristic phonon energy $\omega_{\text{ph}} = 2p_0v_s$, where v_s is the velocity of sound. Let

$$\frac{|\gamma_{\mathbf{p}_0}| \tau_r}{\omega_{\text{ph}} \tau_{\text{ph}}} \ll 1, \quad \frac{\omega_{\text{ph}}}{\theta} \ll 1,$$

where $\tau_{\text{ph}}^{-1} = 2mg^2/\pi$. Then in Eq. (45) the quantity ν , characterizing the damping owing to the phonons, has the form

$$\nu = \frac{\pi\theta}{2\omega_{\text{ph}}\tau_{\text{ph}}},$$

and for the parameters appearing in the expression for $\eta_{\mathbf{p}}$ (46) we obtain the following values:

$$\eta = \frac{\pi\theta(\Omega/2 - \varepsilon_{\text{min}})}{24\omega_{\text{ph}}\tau_{\text{ph}}\tau_r\Lambda^2}, \quad \alpha\left(\frac{\Omega}{2}\right) = -\frac{\tau_{\text{ph}}\omega_{\text{ph}}}{\tau_r\Omega\theta}.$$

We note that the component in Eq. (46) that is asymmetric in the quasimomentum is present only in the OAF state ($\Delta = i\Delta_I$) and arises as a consequence of the anisotropy of the spectrum of the OAF with $\gamma_{\mathbf{p}} \neq 0$. Substituting into the expression for the current (31) the distribution function (44) with a shift of the chemical potential (46), we arrive at the final result to first order in Δ_I/E_g and $\eta_{\mathbf{p}}/\theta$ (we assume that the normalizing volume is equal to unity):

$$\begin{aligned} \mathbf{j} &= \frac{2\mathbf{P}\Delta_I}{3\pi^2 m_0} (m(\Omega - E_g))^{\nu} \alpha\left(\frac{\Omega}{2}\right) \\ &= -\frac{2\mathbf{P}\Delta_I \tau_{\text{ph}} \omega_{\text{ph}}}{3\pi^2 m_0 \tau_r \Omega \theta} (m(\Omega - E_g))^{\nu}, \end{aligned} \quad (47)$$

$$\Lambda = \tilde{\Delta}_e, \quad \tau_r^{-1} = \frac{4}{3} \Omega \left| \frac{\mathbf{P}_{12}}{m_0} \right|^2 = \frac{4}{3} \Omega \left| \frac{\partial \Lambda}{\partial \mathbf{A}_0} \right|^2.$$

From the inequality $\eta \ll (\Omega/2) - \varepsilon_{\text{min}}$ and the expression obtained above for η it follows that the expression (47) for the photocurrent is valid for the following ratio of the characteristic times in the system: $\tau_{\text{ph}} \ll \Lambda^{-1} \ll \tau_r$. Then the saturation state (43) is distorted only slightly owing to recombination [see Eq. (44)]. In the other limiting case, when $\tau_{\text{ph}} \ll \tau_r \ll \Lambda^{-1}$ holds (this is the weak-field case), it is natural to study a weak distortion of the distribution function (39) owing to the source from the external field:

$$S_{i\mathbf{p}}(\varepsilon_{\mathbf{p}}) = -1 + a(\varepsilon_{\mathbf{p}}) + b(\varepsilon_{\mathbf{p}}^0) \Delta_I |\gamma_{\mathbf{p}}|.$$

Here the functions $a(\varepsilon_{\mathbf{p}})$ and $b(\varepsilon_{\mathbf{p}}^0)$ arise to the extent that $(\partial S_{i\mathbf{p}}/\partial t)_{\text{ext}}$ is nonzero, i.e., $a, b \sim \Lambda^2$. Here the photocurrent also appears owing to the asymmetry of the distortion of the distribution function due to the asymmetry of the spectrum of the OAF with $\gamma_{\mathbf{p}} \neq 0$. From Eq. (31) we obtain the corresponding expression for the photocurrent, assuming $\omega_{\text{ph}}/\theta \ll 1$ and $\Delta_I/E_g \ll 1$:

$$\begin{aligned} \mathbf{j} &= -\frac{\mathbf{P}\Delta_I(2m)^{\nu/2}}{3\pi^2 m_0} \int_{\varepsilon_{\text{min}}}^{\infty} d\varepsilon (e - \varepsilon_{\text{min}})^{\nu/2} b(\varepsilon) \\ &= \frac{16\mathbf{P}\Delta_I \tau_{\text{ph}} \omega_{\text{ph}} \Lambda^2}{3\pi m_0 \theta \Omega^2} (m(\Omega - E_g))^{\nu/2}. \end{aligned} \quad (48)$$

The distortions $\eta_{\mathbf{p}}$ (46) of the stationary distribution function, which lead to the photocurrent (47) in the case of an "intermediate" field ($\tau_{\text{ph}} \ll \Lambda^{-1} \ll \tau_r$), like the distortions $a + b\Delta_I|\gamma|$ in the weak-field case ($\tau_{\text{ph}} \ll \tau_r \ll \Lambda^{-1}$), which lead to the photocurrent (48), are significant near the bottom of the conduction band in the region of momenta of the order of the quasi-Fermi momentum p_0 . In the semiconductor model of an excitonic dielectric, which we are studying here, states in a wide range of energies on the order of the binding energy of an exciton,¹⁸ which owing to the resonance condition significantly exceeds the interval $\Omega/2 - \varepsilon_{\text{min}}$ near the bottom of the conduction band, participate in the formation of the order parameter $\Delta_I(t)$. For this reason, in the indicated interval the function $\Delta_{i\mathbf{p}}(t)$ can be assumed to be a slowly varying function of the momentum $\Delta_{i\mathbf{p}}(t) \approx \Delta_I(t)$, as was done in the derivation of Eqs. (47) and (48).

3. ANOMALOUS PHOTOVOLTAIC EFFECT IN AN ORBITAL ANTIFERROMAGNETIC: STRONG-FIELD CASE

Let us study the semiconductor model of an OAF in a strong field ($\Delta\tau \gg 1$). In a standard semiconductor, coherent states of electrons and holes form in a strong alternating field under conditions of resonance (model of a photon dielectric¹¹):

$$\frac{\Omega - E_g}{\Omega} \ll 1.$$

We show that even in a semiconductor excitonic dielectric it is possible to introduce quasiparticles whose ground state is constructed taking into account both dielectric interband correlations and interband transitions induced by the strong wave. The canonical transformation with the coefficients (17), which accomplishes the transition from the operators $a_{i\mathbf{p}}$ (5) to new operators $\alpha_{i\mathbf{p}}$, diagonalizes the time-independent part (5) of the Hamiltonian (4). The time-dependent off-diagonal part of the transformed Hamiltonian can be expressed in terms of the renormalized interband matrix element of the momentum (28). In this form the problem is now analogous to the problem of a photon dielectric.¹¹ We now transform in the obtained Hamiltonian to an "oscillating" system of coordinates through a time-dependent unitary transformation,¹¹ which fixes the chemical potential of the quasiparticles of the excitonic dielectric $\alpha_{i\mathbf{p}}$ at the value $\Omega/2$:

$$U = \exp\left[-\frac{i\Omega t}{2} \int \frac{d^3 p}{(2\pi)^3} (\alpha_{1\mathbf{p}}^+ \alpha_{1\mathbf{p}} - \alpha_{2\mathbf{p}}^+ \alpha_{2\mathbf{p}})\right]. \quad (49)$$

After this transformation the matrix of the Hamiltonian $\mathcal{H}_0 + \mathcal{H}_{\text{ext}}(t)$ in the basis $\alpha_{i\mathbf{p}}$ is reduced to the form

$$\begin{aligned} \hat{H}(t) &= \hat{H}_0 + \hat{H}_1(t), \\ \hat{H}_0 &= \begin{pmatrix} \varepsilon - \Omega/2 & \Lambda \\ \Lambda^* & -\varepsilon + \Omega/2 \end{pmatrix}, \\ \hat{H}_1(t) &= \begin{pmatrix} F e^{-i\Omega t} + \text{c.c.}, & \tilde{\Lambda}^* e^{2i\Omega t} \\ \tilde{\Lambda} e^{-2i\Omega t}, & -F e^{-i\Omega t} + \text{c.c.} \end{pmatrix}. \end{aligned} \quad (50)$$

Here, in accordance with the notation (6), the following parameters were defined:

$$\begin{aligned}\tilde{\Lambda}_p &= \hat{2}u_p v_p^* f_p + \tilde{\Delta}_{ep} u_p^2 - \tilde{\Delta}_{ep} (v_p^*)^2, \\ F_p &= (u_p^2 - |v_p|^2) f_p - \tilde{\Delta}_{ep} u_p v_p^* + \tilde{\Delta}_{ep} u_p v_p \equiv \frac{\partial \varepsilon_p}{\partial p} A_0.\end{aligned}$$

The resonance part \hat{H}_0 of the Hamiltonian $\hat{H}(t)$ no longer depends on time and can be diagonalized by a canonical transformation with the coefficients \bar{u}_p and \bar{v}_p :

$$\begin{aligned}\bar{u}_p^2 &= 1 - |\bar{v}_p|^2 = \frac{E_p + \varepsilon_p - \Omega/2}{2E_p}, \\ \bar{u}_p \bar{v}_p &= -\frac{\Lambda_p}{2E_p}, \quad \bar{v}_p = |\bar{v}_p| \frac{\Lambda_p}{|\Lambda_p|},\end{aligned}\quad (51)$$

where E_p is the dispersion relation (41). The transformation (51) transforms the operators α_{ip} into the new operators β_{ip} , which describe quasiparticles that are coherent electron and hole states arising as a result of both Coulomb correlations and interband transitions induced by the strong field.¹⁴

In Eq. (29) for the current we go over to the operator basis α_{ip} , and then perform the transformation (49). Then the current can be expressed as follows in terms of the Green's function of the Hamiltonian (50):

$$\begin{aligned}j(t) &= i \text{Sp} \int \frac{d^3 p d\omega}{(2\pi)^4} \left\{ \begin{pmatrix} \partial \varepsilon_p & \frac{1}{m_0} \mathbf{P}_{12}(\mathbf{p}) e^{i\Omega t} \\ \frac{1}{m_0} \mathbf{P}_{12}^*(\mathbf{p}) e^{-i\Omega t} & -\partial \varepsilon_p \end{pmatrix} \right. \\ &+ \left. \begin{pmatrix} (\partial F_p - \delta F_p) e^{-i\Omega t} & \partial \Lambda_p - \delta \Lambda_p \\ (\partial \tilde{\Lambda}_p - \delta \tilde{\Lambda}_p) e^{-2i\Omega t} & -(\partial F_p - \delta F_p) e^{-i\Omega t} \end{pmatrix} \right. \\ &+ \left. \text{h.c.} \right\} \hat{G}(\mathbf{p}, \omega, t),\end{aligned}\quad (52)$$

where the renormalized interband matrix element of the momentum $\mathbf{P}_{12}(\mathbf{p})$ is defined in Eq. (28), and the following notation has been introduced:

$$\begin{aligned}\partial &= \partial / \partial \mathbf{p}, \\ \delta \Lambda_p &= 2f_p \partial(u_p v_p) + \tilde{\Delta}_{ep} \partial(u_p^2) - \tilde{\Delta}_{ep} \partial(v_p^2), \\ \delta \tilde{\Lambda}_p &= 2f_p \partial(u_p v_p^*) + \tilde{\Delta}_{ep} \partial(u_p^2) - \tilde{\Delta}_{ep} \partial(v_p^*), \\ \delta F_p &= f_p \partial(u_p^2 - |v_p|^2) - \tilde{\Delta}_{ep} \partial(u_p v_p^*) - \tilde{\Delta}_{ep} \partial(u_p v_p).\end{aligned}$$

In order to obtain the constant component of the current (52) we expand the kinetic Green's function in a series in the perturbation $\hat{H}_1(t)$ (50). Separating in Eq. (50) the principal (resonance) components we obtain

$$\hat{G}(\mathbf{p}, \omega, t) = \hat{G}_0(\mathbf{p}, \omega) + \hat{G}_0 \hat{H}_1(t) \hat{G}_0 + \hat{G}_0 \hat{H}_1(t) \hat{G}_0, \quad (53)$$

where the "zeroth" functions satisfy relations of the form (16), in which the matrices \hat{x} and \hat{y} are constructed from the coefficients (51) and the energy denominators contain instead of ε_p the spectrum E_p (41). The contribution to the stationary component of the current, determined by the first term in Eq. (53), has the form

$$\begin{aligned}j_1 &= \int \frac{d^3 p}{(2\pi)^3} \frac{\partial E_p}{\partial \mathbf{p}} (S_{2p} - S_{1p}) \\ &+ \int \frac{d^3 p}{(2\pi)^3} \left(\delta \Lambda_p \frac{\Lambda_p^*}{2E_p} + \text{c.c.} \right) (S_{1p} - S_{2p}).\end{aligned}\quad (54)$$

The other contribution to the current, associated with the second and third terms in Eq. (53), can be written as follows:

$$j_2 = \int \frac{d^3 p}{(2\pi)^3} \left[\frac{\Lambda_p^* F_p}{E_p \Omega} \frac{\mathbf{P}_{12}(\mathbf{p})}{m_0} + \text{c.c.} \right] (S_{1p} - S_{2p}).$$

Using the self-consistency equation for the dielectric order parameter (see below) it can be shown that the current j_2 exactly cancels the second term in Eq. (54). As a result, the expression for the constant component of the current in an excitonic dielectric in a strong field assumes the standard form

$$j = \int \frac{d^3 p}{(2\pi)^3} \frac{\partial E_p}{\partial \mathbf{p}} (S_{2p} - S_{1p}). \quad (55)$$

The distribution functions $S_{1,2p}$ in Eq. (55) must be sought as a solution of the corresponding kinetic equations. The method for obtaining these equations for an excitonic dielectric in a strong field is in principle analogous to the method employed above for the weak-field case (see also Ref. 14). It is significant that in a strong field a source does not arise in the complete collision integral [see Eq. (42)] and the effect of the field on the excitonic dielectric reduces to renormalization of the quasiparticle spectrum (41). The electron-phonon collision integral, which, in contrast with Eq. (34), must be supplemented with a term describing creation and annihilation of quasiparticles through the field gap Λ , vanishes for a quasi-Fermi distribution function with zero chemical potential (an analogous situation also occurs in a photon dielectric¹¹):

$$S_{1p} = -S_{2p} = -\text{th} \frac{E_p}{\theta}, \quad n_{ip} = \left(\exp \frac{E_p}{\theta} + 1 \right)^{-1}. \quad (56)$$

The quasiparticle-impurity collision integral vanishes for an arbitrary energy function $S_{1p} = S_1(E_p)$, in particular, the distribution (56). Thus intraband relaxation processes cannot distort the quasiparticle distribution away from the quasiequilibrium distribution (56). A distribution function different from the quasi-Fermi function and therefore a nonzero photocurrent (55) can be obtained only by taking into account recombination, which does not conserve the number of particles in a band. The recombination collision integral for the strong-field case has the form

$$\left(\frac{\partial S_{1p}}{\partial t} \right)_r = \frac{1}{2\tau_r(\mathbf{p})} [|\bar{v}_p|^4 (1 - S_{1p})^2 - \bar{u}_p^4 (1 + S_{1p})^2], \quad (57)$$

where the relaxation time $\tau_r(\mathbf{p})$ is defined in Eq. (38). In the absence of dielectric pairing and hybridization the results (38) and (57) transform into analogous relations of the theory of a photon dielectric.¹¹

The stationary distribution, for which Eq. (57) vanishes, does not reduce to the quasiequilibrium distribution (56) in any limit:

$$S_{1p} = |\bar{v}_p|^2 - \bar{u}_p^2, \quad n_{ip} = |\bar{v}_p|^2. \quad (58)$$

The function (58) is the solution of the kinetic equation in the limit $\tau_r \ll \tau_{ph}, \tau_{imp}$. However, even in the case $\tau_r \gg \tau_{ph}, \tau_{imp}$, which is usually realized in semiconductors, the complete collision integral does not vanish for the quasi-Fermi distribution function. It is not difficult to show (see, for ex-

ample, Refs. 11 and 14) that the dissipation of energy of the external field in the stationary state also arises in the system only to the extent that recombination occurs.

Since the dielectric order parameter $\Delta_p(t)$ is formed in a wide range of momenta, greatly exceeding the volume of the phase space occupied by nonequilibrium charge carriers, to calculate the photocurrent we assume that $\Delta_p(t)$ given by (7) does not depend on \mathbf{p} . As a result, in the limit of strong recombination (58) we obtain from Eq. (55) to first order in the hybridization and the imaginary order parameter Δ_I

$$\mathbf{j} = \frac{4\mathbf{P}\Delta_I\Delta_-\Lambda}{3\pi m_0\Omega^2} \left[\frac{(m(\Omega - E_g))^{3/2}}{\Lambda} + \frac{(mE_g)^{3/2}}{\Omega} \right]. \quad (59)$$

Here Δ_- is the interband transition amplitude λ renormalized by the Coulomb interaction [see the formula (67)] and the normalization volume has been set equal to unity.

Thus we have shown that in an OAF without a center of inversion ($\mathbf{P}_{12}\Delta_I \neq 0$) the photovoltaic effect, described by the formulas (47), (48), and (59) in the limiting cases of moderate, weak, and strong fields, occurs under nonequilibrium conditions in the presence of dissipation. It is significant that these contributions to the diagonal photocurrent arose even in the Born approximation when scattering processes were taken into account.

4. ORDER PARAMETER AND ANOMALIES OF THE PHOTOCONDUCTIVITY NEAR THE TRANSITION POINT

We first study the order parameter of an excitonic dielectric in the semimetal model, when the screened interelectron interaction potential can be assumed to be constant. The self-consistency equation for the complex order parameter [compare with Eq. (21)] has the form

$$\Delta = g_R \operatorname{Re} \int \frac{d^3p d\omega}{(2\pi)^4} iG_{12}(\mathbf{p}, \omega) + ig_I \operatorname{Im} \int \frac{d^3p d\omega}{(2\pi)^4} iG_{12}(\mathbf{p}, \omega), \quad (60)$$

where the integration constants g_R and g_I for the real and imaginary parts of Δ are, generally speaking, different. On the basis of the structure of the kinetic Green's functions (16) the self-consistency equation (60) can be rewritten, neglecting nonlocal corrections, as follows:

$$\Delta_R = g_R \int \frac{d^3p d\omega}{(2\pi)^4} [S_{1\mathbf{p}}(\omega) \operatorname{Re} A_{12\mathbf{p}}^*(\omega) + S_{2\mathbf{p}}(\omega) \operatorname{Re} A_{12\mathbf{p}}^y(\omega)], \quad (61)$$

$$\Delta_I = g_I \int \frac{d^3p d\omega}{(2\pi)^4} [S_{1\mathbf{p}}(\omega) \operatorname{Im} A_{12\mathbf{p}}^*(\omega) + S_{2\mathbf{p}}(\omega) \operatorname{Im} A_{12\mathbf{p}}^y(\omega)],$$

where we have introduced the spectral functions

$$\hat{A}^x = \hat{x}(G_x^a - G_x^r), \quad \hat{A}^y = \hat{y}(G_y^a - G_y^r).$$

According to Eq. (16), because of the presence of hybridization

$$\operatorname{Im} A_{12}^{x,y}(\Delta=0) = \operatorname{Im} A_{120}^{x,y} \neq 0, \quad \operatorname{Im} A_{120}^{x,y}(\mathbf{p}) = -\operatorname{Im} A_{120}^{x,y}(-\mathbf{p}).$$

For this reason, any asymmetric distortion of the distribution functions $S_{1,2\mathbf{p}}$ in the direction of the interband matrix element of the momentum vector \mathbf{P} results in the appearance of a source in the self-consistency equation for the imaginary (toroidal) order parameter [the macroscopic physical quantity that arising spontaneously at the phase transition into the OAF state in the model of an excitonic dielectric is

the toroid momentum density $\mathbf{T} \sim \mathbf{P}\Delta_I$ (Refs. 7 and 10)]. The asymmetric distortion of the distribution function can be caused by both a constant electric field \mathbf{E} and some entrainment effect and is accompanied by the appearance of a dissipative electric current. For this reason we can say that the ohmic current is a source for the toroid order parameter.

The corresponding microscopic calculation under conditions of weak impurity scattering ($\theta\tau_{\text{imp}} \gg 1$) was performed in Ref. 8. At temperatures θ near the transition temperatures θ_R or θ_I in the zero-gap state ($|\Delta|\tau_{\text{imp}} \ll 1$) it was found that to first order in Δ_R and Δ_I the equations for Δ_R and Δ_I become decoupled:

$$\Delta_R = \frac{(\mathbf{PE})\varphi_1}{\theta - \theta_R}, \quad \Delta_I = \frac{(\mathbf{PE})\varphi_2}{\theta - \theta_I},$$

where under certain simplifying assumptions

$$\varphi_1 = -\frac{4\tau_{\text{imp}}\varepsilon_F}{3\pi m} A_2, \quad \varphi_2 = -\frac{A_2}{2\pi m}, \quad A_n = \sum_{k=0}^{\infty} (2k+1)^{-n},$$

and ε_F is the Fermi energy of the semimetal. In thermodynamic equilibrium, when the distribution function does not change, it follows from Eq. (61) that

$$\varphi_1 = -\frac{2}{3} \frac{\varepsilon_F}{m(\pi\theta)^2} A_3, \quad \varphi_2 = 0,$$

which reflects ordering of the ferroelectric type when $\Delta_R \neq 0$. Moreover, in Ref. 8 an expression was derived for corrections to the current which arise owing to the OAF ordering and are nonlinear in the field \mathbf{E} :

$$\mathbf{j}_{\text{im}} = -8\sigma_0 \frac{\tau_{\text{imp}}^2 \mathbf{E}(\mathbf{PE}) A_2 \Delta_I}{m\pi\theta} + 2\sigma_0 \frac{\mathbf{P}(\mathbf{E})^2 A_4 \Delta_I}{m(\pi\theta)^3},$$

$$\sigma_0 = \frac{4\varepsilon_F}{3m} N(0) \tau_{\text{imp}},$$

and it was pointed out that the existence of a photovoltaic effect in OAF without a center of inversion ($\mathbf{P}\Delta_I \neq 0$) already follows directly from the expression for the current \mathbf{j}_{im} . In the limit of very low frequencies of the external electromagnetic field ($\Omega\tau_{\text{imp}} \ll 1$) the frequency Ω can be neglected when calculating the current. The PVE tensor β_{ilk} (1) in this limit is identical to the nonlinear conductivity tensor, which determines the current \mathbf{j}_{im} and has the form

$$\beta_{ilk} = \sigma_{ilk} = (\alpha\delta_{il}P_k + \beta P_i\delta_{lk}) \Delta_I.$$

Using the results of Ref. 8, which were presented above, for the induced order parameter Δ_I we can write the following expression for the photoconductivity tensor (1) in the case $\Omega\tau_{\text{imp}} \ll 1$ near the point of the transition into the OAF state:

$$\gamma_{ilkn} = \frac{\bar{\gamma}_{ilkn}}{\theta - \theta_I}, \quad (62)$$

where $\bar{\gamma}_{ilkn}$ is the dissipative tensor:

$$\bar{\gamma}_{ilkn} = -\sigma_0 \frac{A_2}{\pi^2 m^2 \theta} [-4\tau_{\text{imp}}^2 A_2 (\delta_{ik}P_n + \delta_{in}P_k) P_l + \frac{A_4}{\pi^2 \theta^2} \delta_{kn} P_i P_l].$$

From the expression for γ_{ilkn} (62) it follows that near the temperature of the transition into the OAF state the temperature dependence of the photoconductivity conforms to

the Curie-Weiss law. In reality, the singularity in Eq. (62) is smeared out to the extent that the phase transition is smeared out under nonequilibrium conditions. The presence of a source in the order parameter, associated with the external field responsible for the nonequilibrium, also leads to an analogous effect. For a toroid order parameter, for example, the Poynting vector can be such a source.

We return now to the semiconductor model, in which an alternating electromagnetic field with finite frequency gives rise to interband transitions of charge carriers and the phase transition into the excitonic-dielectric state occurs with $E_g = E_{ex}$ (E_{ex} is the binding energy of the exciton). The self-consistency equation for the imaginary dielectric order parameter ($\Delta = i\Delta_I$) has the following form in the weak-field limit:

$$\Delta_p + \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\Delta_{\mathbf{k}}}{2\varepsilon_{\mathbf{k}}} S_1(\varepsilon_{\mathbf{k}}) = h_p, \quad (63)$$

$$h_p = - \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\gamma_{\mathbf{k}}}{2\varepsilon_{\mathbf{k}}} S_{1\mathbf{k}}^1.$$

Here $S_1(\varepsilon_{\mathbf{p}})$ is the stationary distribution function (43) (we neglect the recombination corrections to the order parameter $\eta/\theta \ll 1$) and $S_{1\mathbf{p}}^1$ is the change induced in the distribution function by the constant external field and can be determined from the kinetic equation:

$$-\mathbf{E} \frac{\partial S_1(\varepsilon_{\mathbf{p}})}{\partial \mathbf{p}} = \frac{\delta}{\delta S_1(\varepsilon_{\mathbf{p}})} \left[\left(\frac{\partial S_1}{\partial t} \right)_{em} \right] S_{1\mathbf{p}}^1.$$

In the weak-field limit we neglect the alternating field in Eq. (63) according to the parameter $\Lambda\tau \ll 1$.

By means of the substitution

$$\Delta_p = 2\xi_p \Psi(\mathbf{p})$$

Eq. (63) can be reduced in the standard manner to the Schrödinger equation for the Coulomb problem with the effective potential¹⁷

$$V^*(\mathbf{p}-\mathbf{k}) = V(\mathbf{p}-\mathbf{k}) \operatorname{th} \frac{E_g}{4\theta}$$

and the right-hand side of Eq. (63), which plays the role of a source. The solution is sought with the help of the Green's function of the homogeneous equation

$$G(\omega, \mathbf{p}, \mathbf{k}) = \sum_n \frac{\varphi_n^*(\mathbf{k}) \varphi_n(\mathbf{p})}{\omega - E_n}. \quad (64)$$

Here $\varphi_n(\mathbf{p})$ are the eigenfunctions and E_n the eigenvalues of the homogeneous equation, containing, in particular, the exciton binding energy

$$E_{ex}(\theta) = E_{ex}^0 \operatorname{th}^2 \frac{E_g}{4\theta},$$

where in dimensionless units

$$E_{ex}^0 = \frac{me^4}{2\hbar^2 \varepsilon^2}.$$

On the basis of the assumptions made in deriving the expression for the current (47) and retaining in Eq. (64) only the singular term, we obtain for the particular solution $\Psi(\mathbf{p})$ of the inhomogeneous problem and of interest to us

$$\Psi(\mathbf{p}) = \frac{\Delta_p}{E_g} = \frac{\tau_{ph} \omega_{ph} \Omega(\mathbf{PE}) I \varphi(\mathbf{p})}{\theta^2 m_0 (E_g - E_{ex}(\theta))}.$$

where I is a dimensionless constant. As a result, the expression for the photocurrent component (47), which is related to the constant field \mathbf{E} and determines the photoconductivity, has the following form above the transition point ($E_g > E_{ex}(\theta)$):

$$\mathbf{j} = - \frac{2\tau_{ph}^2 \omega_{ph}^2 I \varphi(0)}{3\pi^2 m_0^2 \tau_r \theta^3} (m(\Omega - E_g))^{1/2} \frac{\mathbf{P}(\mathbf{PE})}{1 - E_{ex}(\theta)/E_g}. \quad (65)$$

Thus the photoconductivity in the OAF in the semiconductor model near the transition point conforms to the Curie-Weiss law as a function of the band gap. The photocurrent component in a very weak field (48) also behaves analogously near the transition point, if the order parameter of the OAF is induced by a constant field \mathbf{E} ; we shall not present the corresponding expression.

In the strong-field limit $\Lambda\tau \gg 1$ renormalization of the interband transition amplitude λ by the Coulomb interaction becomes significant.¹⁸ This renormalization is described by the self-consistency equation for the component $\tilde{\Delta}$ of the order parameter (7):

$$\tilde{\Delta}_{ep} = \lambda + \tilde{\Delta}_p = \lambda + \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\tilde{\Delta}_{e\mathbf{k}} (n_{2\mathbf{k}} - n_{1\mathbf{k}})}{2[(\varepsilon_{\mathbf{k}} - \Omega/2)^2 + |\tilde{\Delta}_{e\mathbf{k}}|^2]^{1/2}}. \quad (66)$$

For $\Omega > E_g - E_{ex}$ the coherent external field in Eq. (66) serves as a source of the Bose condensate of excitons with the order parameter formed at the frequency Ω .^{18,14}

For what follows it is convenient to redefine the order parameters $\tilde{\Delta}_p$ and $\tilde{\Delta}_p$, introducing the following quantities:

$$\Delta_+ = 1/2(\tilde{\Delta} + \tilde{\tilde{\Delta}}), \quad \Delta_- = \lambda + 1/2(\tilde{\Delta} - \tilde{\tilde{\Delta}}). \quad (67)$$

Since the number of quasiparticles is equal to the number of quasiholes, $S_{1\mathbf{p}} = -S_{2\mathbf{p}}$, the system of self-consistency equations assumes the form¹³

$$\Delta_p = \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) u_{\mathbf{k}} v_{\mathbf{k}} (\bar{u}_{\mathbf{k}}^2 - |\bar{v}_{\mathbf{k}}|^2) S_{1\mathbf{k}},$$

$$\Delta_{+p} = \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) (u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2) \bar{u}_{\mathbf{k}} \bar{v}_{\mathbf{k}} S_{1\mathbf{k}}, \quad (68)$$

$$\Delta_{-p} = \lambda + \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) (u_{\mathbf{k}}^2 + v_{\mathbf{k}}^2) \bar{u}_{\mathbf{k}} \bar{v}_{\mathbf{k}} S_{1\mathbf{k}}.$$

We consider the case of strong recombination (58). In this limit the self-consistency equation (68) can be written as follows:

$$\Delta_p = \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\gamma_{\mathbf{k}} + \Delta_{\mathbf{k}}}{2\varepsilon_{\mathbf{k}}} \left(1 - \frac{|\Lambda_{\mathbf{k}}|^2}{E_{\mathbf{k}}^2} \right),$$

$$\Delta_{+p} = \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\Lambda_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \Omega/2)}{4E_{\mathbf{k}}^2}, \quad (69)$$

$$\Delta_{-p} = \lambda + \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\xi_{\mathbf{k}}}{\varepsilon_{\mathbf{k}}} \frac{\Lambda_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \Omega/2)}{4E_{\mathbf{k}}^2}.$$

As follows from the equation for Δ_p in Eqs. (69), the transition to the state of an excitonic dielectric is suppressed in a strong field by the effective decrease of the interaction potential. In the range of energies which are significant for the formation of the order parameter Δ_p , however, the field cor-

rection is small ($|\Lambda|^2/E^2 \sim (\Lambda/\Omega)^2 \ll 1$) and can be neglected.

If, in addition, a weak constant field \mathbf{E} is superposed on the system, then from the kinetic equation with a recombination collision integral (57) we obtain for the correction to the stationary distribution function (58)

$$S_{1p}(\mathbf{E}) = S_{1p} + 2\delta n_p, \quad (70)$$

$$\delta n_p = \tau_r(\mathbf{p}) \frac{E_p^2}{|\Lambda_p|^2} \left(\mathbf{E} \frac{\partial}{\partial \mathbf{p}} \right) \left(\frac{\Omega/2 - \varepsilon_p}{E_p} \right).$$

The correction to the distribution function (70) leads to the appearance of a source in the equation for Δ_p :

$$\Delta_p - \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{\Delta_k}{2\varepsilon_k} = h_p, \quad (71)$$

$$h_p = \int \frac{d^3k}{(2\pi)^3} V(\mathbf{p}-\mathbf{k}) \frac{(\mathbf{P}\mathbf{k})(\mathbf{E}\mathbf{k})}{mm_0} \times \tau_r(\mathbf{k}) \frac{\xi_k - \Omega/2}{\xi_k [(\xi_k - \Omega/2)^2 + \Lambda^2]^{1/2}}.$$

As we have already pointed out above, in calculations of the photocurrent in the region of states near the bottom of the conduction band all Coulomb parameters can be assumed to depend weakly on the momentum. To first order in the hybridization and the order parameter $\Delta = \Delta_I$ the field gap parameter Λ also does not depend on the momentum and the constant field E :

$$\Lambda \approx \Delta_+ + \Delta_- = \bar{\Delta}_e = \text{const.}$$

In addition, in this approximation the recombination time can also be replaced by a constant [see Eq. (47)].

Substituting into Eq. (59) the solution Eq. (71) found with the help of the Green's function of the homogeneous equation, we obtain for the current contribution determining the photoconductivity

$$j = \frac{4\Delta_- \Lambda T_r J}{3\pi m^2 \Omega^2} \left[\frac{(m(\Omega - E_g))^{3/2}}{\Lambda} + \frac{(mE_g)^{3/2}}{\Omega} \right] \frac{\mathbf{P}(\mathbf{P}\mathbf{E})}{1 - E_{ex}/E_g}, \quad (72)$$

where J is a dimensionless constant. As in the weak-field case (65) the current (72) near the point of a toroidal excitonic transition conforms to the Curie-Weiss law in the parameter E_g . Of course, all remarks about the smearing of the singularity in the photoconductivity, i.e., replacement of the divergence by a local maximum depending on the decay and the presence of a source from the alternating field in the self-consistency equation, made above in application to the semiconductor model (62), are also valid for the semiconductor model (65), (72).

The obtained results can be given a simple phenomenological interpretation. We introduce the dissipative function

$$\tilde{\mathcal{F}}(\mathbf{T}) = \mathcal{F}_0(\mathbf{T}) - \tilde{\kappa}(\mathbf{T}\mathbf{j}_0) = \alpha T^2 + \beta T^4 - \tilde{\kappa}\mathbf{T}\mathbf{j}_0. \quad (73)$$

where $\mathcal{F}_0(\mathbf{T})$ is Landau's free energy functional for the toroidal order parameter $\mathbf{T} \sim \mathbf{P}\Delta_I$ and $\alpha = a(\theta - \theta_I)$, where θ is any parameter with respect to which a phase transition occurs (temperature, band gap), $a, \beta > 0$, and $\mathbf{j}_0 = \sigma_0\mathbf{E}$ is the ohmic current. The last term on the right-hand side of (73) reflects the fact, established in Ref. 8, that the ohmic current

is the source for the toroidal order parameter. By varying the function (73) with respect to \mathbf{T} and substituting the expression found for the induced toroidal order parameter

$$\mathbf{T}_{ind} = \frac{\kappa\mathbf{n}(\mathbf{n}\mathbf{E})}{\theta - \theta_I}, \quad \kappa = \frac{\tilde{\kappa}\sigma_0}{a}, \quad \mathbf{n} = \frac{\mathbf{P}}{|\mathbf{P}|} \quad (74)$$

into the expression for the dissipative toroidal current

$$j_i = \beta_{ij} T_j, \quad (75)$$

we obtain the Curie-Weiss law calculated in this work on the basis of the microscopic model. The obtained effect is universal and is independent of the special method, studied in this work, for creating nonequilibrium in an alternating electric field (PVE).

Finally, we note that the singularity of the photoconductivity at the point of the nonequilibrium phase transition is not a property specific to an OAF. The expression (75) for the photocurrent can contain, instead of the density of the toroidal moment, for example, the polarization $\vec{\mathcal{P}}$ (in the excitonic dielectric model $\vec{\mathcal{P}} \sim \mathbf{P}\Delta_R$). The photovoltaic effect is then governed by the coherent (displacement) mechanism (3).³⁻⁵ The polarization component induced by a constant electric field contains, as is well known, a singular denominator analogous to Eq. (74). The divergence of the photoconductivity at the point of the ferroelectric transition thus will be a simple consequence of the divergence of the permittivity, which also happens under equilibrium conditions.

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