Kinetic theory of the displacement photovoltaic effect in piezoelectrics

V. I. Belinicher, E. L. Ivchenko, and B. I. Sturman

Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk

(Submitted 15 January 1982)
Zh. Eksp. Teor. Fiz. **83**, 649–661 (August 1982)

It is shown that, besides the mechanisms of the photovoltaic effect (PVE) that are connected with the appearance of asymmetry in the electron velocity distribution [V. I. Belinicher and B. I. Sturman, Sov. Phys. Usp. **23**, 199 (1980); E. L. Ivchenko and G. E. Pikus, in: Problemy sovremennoĭ fiziki (Problems of Modern Physics), Nauka, Leningrad, 1980, p. 275], there exist independent mechanisms stemming from the displacement of the electrons in **R** space when they undergo quantum transitions. This displacement PVE is determined by the contributions to the current of the off-diagonal—in the band numbers—density matrix elements. A consistent kinetic theory of the displacement PVE is constructed for band-band and impurity-band transitions. It is shown that the magnitudes of the displacements can be expressed in terms of the phases of the transition matrix elements. The displacement photocurrent is equal to the sum of the ionization, recombination, and scattering currents, which do not depend on the electron and hole mean free paths. The magnitude of the off-diagonal contribution to the current can be comparable to the magnitude of the diagonal contribution.

PACS numbers: 72.40. + w, 77.60. + v

1. INTRODUCTION

A microscopic theory of the photovoltaic effect in noncentrosymmetric crystals was developed recently.[1,2] The photovoltaic effect (PVE) is determined by the following phenomenological relation for the current:

$$j_\alpha=\beta^{s}_{\alpha\beta\gamma}(e_\beta e_\gamma^*+e_\beta^* e_\gamma)J+i\beta_{\alpha\beta}{}^{a}[\mathbf{e}\times\mathbf{e}^*]_\beta J, \tag{1}$$

where **e** is the polarization vector, and J is the intensity, of the light wave. The tensors $\beta^{s}_{\alpha\beta\gamma}$ and $\beta^{a}_{\alpha\beta}$ describe respectively the linear and circular components of the photocurrent.

Until recently, the theory of the PVE was based on the following well-known formula for the current:

$$\mathbf{j}=\frac{e}{\hbar}\sum_{s\mathbf{k}} f_{s\mathbf{k}}\frac{\partial}{\partial\mathbf{k}}\varepsilon_{s\mathbf{k}}, \tag{2}$$

where $f_{s\mathbf{k}}$ is the distribution function for the electrons in the band s. In that theory this formula is obtained on the basis of intuitive arguments, according to which the current can be considered to be the sum of contributions from all the particles with velocities $\mathbf{v}_{s\mathbf{k}}=\hbar^{-1}\partial\varepsilon_{s\mathbf{k}}/\partial\mathbf{k}$. The momentum (i.e., k) distribution asymmetry, which is necessary for the existence of the current (2), arises, under the action of light, as a result of the asymmetry in the elementary electronic processes (photoexcitation, recombination, scattering). The magnitude of the asymmetric part of the distribution function (i.e., the part that is odd in **k**) and the current strengths themselves contain a kinetic parameter, and are proportional to the momentum relaxation time τ_p, i.e., to the electron mean free path. This relationship is general in that theory of the PVE.[1,2]

At the same time, it is well known that, besides the contribution (2) to the current, there exists, generally speaking, another contribution connected with the off-diagonal—in the band numbers—density matrix elements $\rho\mathbf{k}_1\mathbf{k}$:

$$\mathbf{j}^{\rho}=\frac{e}{m}\sum_{\substack{s\neq s_1\\ \mathbf{k},\mathbf{k}_1}}\mathbf{P}^{ss_1}_{\mathbf{k}\mathbf{k}_1}\rho^{s_1 s}_{\mathbf{k}_1\mathbf{k}}. \tag{3}$$

Here $\mathbf{P}^{ss_1}_{\mathbf{k}\mathbf{k}_1}=\langle s\mathbf{k}|-i\hbar\nabla|s_1\mathbf{k}_1\rangle$ is a matrix element of the momentum operator. As applied to the circular PVE, these contributions were first considered by Mednis.[3] But it is shown in Ref. 4 that the theory developed in Ref. 3 is incorrect, and that the "off-diagonal circular current" is completely canceled out by that part of the diagonal contribution which is due to the change that occurs in the distribution function $f_{s\mathbf{k}}$ as a result of the renormalization of the energy in the field of the circularly polarized wave. The off-diagonal contributions to the circular current at high light intensities are taken into account in Ref. 5.

A number of authors have made attempts to construct a theory of the linear PVE with allowance for the off-diagonal contributions to the current.[6-8] Kristofel' and Gulbis[6] and Krant and Baltz[7] proceeded from a well-known formula (similar to Kubo's formula) based on the computation of the current at the difference frequency $\omega'-\omega$ of two waves and the subsequent passage to the limit $\omega'\to\omega$:

$$j_\alpha=\frac{e^3E^2}{m^3\omega^2}\sum_{\substack{ss_1s_2\\ \mathbf{k},\xi=\pm\hbar\omega}}(f_{s_1\mathbf{k}}-f_{s_2\mathbf{k}})\frac{e_\beta e_\gamma P^{\gamma}_{s_2s_1}}{\varepsilon^{s_1s_2}_{\mathbf{k}}+\xi+i0}\left(\frac{P^{\alpha}_{s_1s}P^{\beta}_{ss_2}}{\varepsilon^{s_1}_{\mathbf{k}}-\varepsilon_{\mathbf{k}}{}^{s}}-\frac{P^{\beta}_{s_1s}P^{\alpha}_{ss_2}}{\varepsilon_{\mathbf{k}}{}^{s}-\varepsilon^{s_2}_{\mathbf{k}}}\right). \tag{4}$$

In this relation ω is the amplitude of the electric field of the wave. The computation of the current from this formula in the case of low intensities should be carried out with the equilibrium Fermi distribution functions for the electrons. Therefore, the current (4), in contrast to the corresponding expression in Ref. 1, does not depend on the electron mean free paths. As will be shown below, this photo-voltaic effect

0038-5646/82/080359-08$04.00

is due to the displacement of the electrons in **R** space when they undergo quantum transitions; therefore, we shall call it the displacement PVE. The displacement-PVE theory developed in Refs. 6–8 on the basis of (4) or similar relations is inconsistent, incomplete, and in many cases intrinsically contradictory. Thus, in Ref. 6 it is concluded that the PVE can occur in the absence of light absorption, which contradicts the law of conservation of energy. The expressions obtained in Refs. 6–8 for the current do not satisfy the requirement that they vanish under the conditions of thermodynamic equilibrium with the thermal photons, which contradicts the second law of thermodynamics. The main shortcomings of the indicated papers are connected with some procedural errors.1)

First, the formula (4) cannot be used to describe the diagonal contribution to the current. This can be seen from the existence in it of singularities at $s=s_{1,2}$. The diagonal contribution to the current should be found by solving the kinetic equation, and is described by the theory expounded in Refs. 1 and 2. And to describe the off-diagonal contributions to the direct current, we must eliminate from the formula (4) the terms containing the singularities, i.e., we must assume $s\neq s_1$ in the first term and $s\neq s_2$ in the second.

Second, besides the above-considered contribution to the current, there exist other off-diagonal contributions, and these contributions are not small. They arise when the interband (primarily spontaneous) recombination processes and the processes of scattering on the impurities and phonons are taken into account. Allowance for these contributions is not only necessary in quantitative computations, it is essential to the intrinsic self-consistency of the theory. In particular, the neglect of the spontaneous emission (the drift processes) leads to the existence of current under conditions of total thermodynamic equilibrium, to the existence of a direct current in the case when an electron on an impurity undergoes transitions between localized states.

Of great importance is the third shortcoming, which consists in the following. The direct use of finite-band models, i.e., two-band, three-band, etc., models in the j^{ρ} calculation is incorrent. It turns out that it is necessary and possible to carry out a single summation over all the bands of the crystal. This summation can be performed with the use of only the completeness property of the basis Bloch functions and the commutation relations for the coordinate and momentum operators, without recourse to model arguments. The performance of this intermediate summation results in a significant simplification of the general formulas for the current, which then easily admit of the use of particular models for the crystal.

The present paper is devoted to the exposition of a consistent theory of the displacement PVE in piezoelectrics for the case of interband and impurity-band transitions.

2. THE GENERAL RELATIONS FOR THE CURRENT

The consistent kientic theory taking account of both the diagonal and the off-diagonal density matrix elements should, as is well known,[9] be constructed as follows. The diagonal matrix elements, i.e., the distribution functions $\rho_{\mathbf{k}}^{ss}=f_{s\mathbf{k}}$, satisfy the kinetic equation. The off-diagonal elements $\rho_{\mathbf{k}\mathbf{k}_1}^{ss_1}$ $(s\neq s_1)$ can be expressed in terms of the distribution functions obtained from the equation of motion

$$i\hbar\,\partial\rho/\partial t=[H,\rho],\quad H=H_0+H^{\Phi\,\mathrm{ph}}+V,\tag{5}$$

where H_0 determines the energy spectrum of the crystal, $H^{\mathrm{ph}}=-e(\mathbf{P}\cdot\mathbf{A})/mc$ describes the interaction with the light, and V is the Hamiltonian for the interaction with the impurities and the phonons. Each of the interactions that induce real transitions then makes an additive contribution to the current. We shall, for simplicity, assume that the electron-phonon interaction is responsible for intraband transitions, i.e., we shall set $V=V_{\mathrm{ef}}$. The contribution made to the current by the electron-impurity interaction can easily be taken into account in the final formulas. To quantize the light, we must express the vector potential **A** in terms of the photon creation and annihilation operators with the aid of the well-known formulas.[10]

The region of applicability of such a kinetic theory coincides with the region of applicability of the kinetic equation. Although the final formulas for the current do not contain the kinetic parameters of the crystals, it is necessary that the conditions, $kl\gg 1$, where k and l are the characteristic wave vector and the electron mean free path respectively, be fulfilled.

Using the above procedure to find $\rho_{kk_1}^{ss_1}$, and neglecting the photon momentum q in comparison with the electron quasimomentum, we obtain for the off-diagonal contribution to the current j^{ρ} (we shall drop hereafter the superscript ρ when that will not lead to a misunderstanding) the expression

$$\begin{gathered}\mathbf{j}=\mathbf{j}^{\mathrm{ph}}+\mathbf{j}^{f};\\ j_{\alpha}^{\mathrm{ph}}=\frac{2\pi ie^3}{m^2}\sum_{\substack{\mathbf{n}_1\mathbf{n}_2\\ \mathbf{q},\lambda}}\frac{[(f_{\mathbf{n}_1}-f_{\mathbf{n}_2})N_{\mathbf{q}\lambda}{}^{\mathrm{ph}}-f_{\mathbf{n}_2}(1-f_{\mathbf{n}_1})]}{\varepsilon_{\mathbf{n}_1\mathbf{n}_2}+\hbar\omega_{\mathbf{q}}+i0}\\ \times\Bigl(\sum_{\substack{n\\ s\neq s_1}}X_{\mathbf{n}_1\mathbf{n}}^{\alpha}P_{\mathbf{n}\mathbf{n}_2}^{\beta}-\sum_{\substack{n\\ s\neq s_2}}P_{\mathbf{n}_1\mathbf{n}}^{\beta}X_{\mathbf{n}\mathbf{n}_2}^{\alpha}\Bigr)\frac{e_{\beta}e_{\gamma}P_{\mathbf{n}_2\mathbf{n}_1}^{\gamma}}{n^2\omega_{\mathbf{q}}}+\text{c.c.},\\ j_{\alpha}{}^{f}=ie\sum_{\mathbf{n}_1\mathbf{n}_2}\frac{[(f_{\mathbf{n}_1}-f_{\mathbf{n}_2})N_q{}^{f}-f_{\mathbf{n}_2}(1-f_{\mathbf{n}_1})]}{\varepsilon_{\mathbf{n}_1\mathbf{n}_2}+\hbar\Omega_{\mathbf{k}_1-\mathbf{k}_2}+i0}\\ \times\Bigl(\sum_{\substack{\mathbf{n}\\ s\neq s_1}}X_{\mathbf{n}_1\mathbf{n}}^{\alpha}V_{\mathbf{n}\mathbf{n}_2}-\sum_{\substack{\tilde{\mathbf{n}}\\ s\neq s_2}}V_{\mathbf{n}_1\mathbf{n}}X_{\mathbf{n}\mathbf{n}_2}^{\alpha}\Bigr)V_{\mathbf{n}_2\mathbf{n}_1}+\text{c.c.}\end{gathered}\tag{6}$$

Here $\mathbf{n}\equiv(\mathbf{k},s)$; $\omega_{\mathbf{q}}$ and $\Omega_{\mathbf{q}}$ are the frequencies, and $N_{\mathbf{q}\lambda}^{\mathrm{ph}}$ and $N_{\mathbf{q}}^{f}$ the occupation numbers, of the photons and phonons respectively; $\lambda=1,2$ numbers the light polarization2); n is the refractive index (we neglect its ω dependence and birefringence); $\varepsilon_{\mathbf{n}_1\mathbf{n}_2}=\varepsilon_{\mathbf{n}_1}-\varepsilon_{\mathbf{n}_2}$ and the $V_{\mathbf{n}_1\mathbf{n}_2}$ are the electron-phonon interaction matrix elements. The periodic Bloch amplitudes $u_n(\mathbf{r})\equiv u_{s\mathbf{k}}(\mathbf{r})$ in (6) are normalized to the volume of the unit cell:

$$\int_{a^3}u_{s\mathbf{k}}{}^{*}u_{s_1\mathbf{k}}\,d\mathbf{r}=\delta_{ss_1}a^3.\tag{7}$$

The relations (6) can be significantly simplified by using the exact sum rules derived in the Appendix. Assuming that the nonradiative interband transitions are forbidden in energy terms, we find finally that

$$\mathbf{j}^{\mathrm{ph}}=e\sum_{\lambda s_1 s_2}\int[(f_{s_1\mathbf{k}}-f_{s_2\mathbf{k}})N_{\mathbf{q}\lambda}^{\mathrm{Ph}}-f_{s_2\mathbf{k}}(1-f_{s_1\mathbf{k}})]$$
$$\times W_{s_2s_1}^{\mathrm{ph}}(\mathbf{q},\mathbf{k})\mathbf{R}_{s_2s_1}^{e}\delta(\hbar\omega_{\mathbf{q}}-\varepsilon_{\mathbf{k}}^{21})\,d\mathbf{k}\,d\mathbf{q},$$
$$\mathbf{j}^{f}=e\sum_{s}\int[(f_{s\mathbf{k}_1}-f_{s\mathbf{k}_2})N_{\mathbf{k}_1-\mathbf{k}_2}^{f}-f_{s\mathbf{k}_2}(1-f_{s\mathbf{k}_1})]\tag{8}$$
$$\times W_{s}^{f}(\mathbf{k}_2,\mathbf{k}_1)\mathbf{R}_{s}(\mathbf{k}_2,\mathbf{k}_1)\delta(\hbar\Omega_{\mathbf{k}_1-\mathbf{k}_2}-\varepsilon_{\mathbf{k}_2\mathbf{k}_1}^{s})\,d\mathbf{k}_1\,d\mathbf{k}_2.$$

Here the $W^{\mathrm{ph},f}$ are the differential probabilities for transitions in which photons and phonons participate:

$$W_{s_2s_1}(\mathbf{k})=\frac{e^2a^3}{(2\pi)^4n^2m^2\omega_{\mathbf{q}}}|\mathbf{e}\mathbf{P}^{s_2s_1}(\mathbf{k})|^2,$$
$$W_{s}^{f}(\mathbf{k}_2,\mathbf{k}_1)=\frac{a^6}{\hbar(2\pi)^5}|V_{\mathbf{k}_2\mathbf{k}_1}^{ss}|^2.\tag{9}$$

The vectors $\mathbf{R}^e$ and $\mathbf{R}$, which have the dimensions of length, are given by the relations

$$\mathbf{R}_{s_2s_1}^{e}(\mathbf{k})=-\frac{\partial}{\partial\mathbf{k}}\Phi_{s_2s_1}^{e}+\boldsymbol{\Omega}_{s_2}(\mathbf{k})-\boldsymbol{\Omega}_{s_1}(\mathbf{k}),$$
$$\mathbf{R}_{s}(\mathbf{k}_2,\mathbf{k}_1)=-\left(\frac{\partial}{\partial\mathbf{k}_1}+\frac{\partial}{\partial\mathbf{k}_2}\right)\Phi_{s}(\mathbf{k}_2,\mathbf{k}_1)+\boldsymbol{\Omega}_{s}(\mathbf{k}_2)-\boldsymbol{\Omega}_{s}(\mathbf{k}_1),\tag{10}$$

in which $\Phi_{s_2s_1}^{e}$ and Φ_s are the phases of the complex quantities $-(\mathbf{e}\cdot\mathbf{P}^{s_2s_1})$ and $V_{\mathbf{k}_2\mathbf{k}_1}^{ss}$, while $\Omega_s(\mathbf{k})$, is the diagonal—in the momentum and the band numbers—part of the coordinate matrix element[11]:

$$\boldsymbol{\Omega}_{s}(\mathbf{k})=\frac{i}{a^3}\int u_{s\mathbf{k}}^{*}\frac{\partial}{\partial\mathbf{k}}u_{s\mathbf{k}}\,d\mathbf{r}.\tag{11}$$

The physical nature of the off-diagonal contributions to the current is clear from the formulas (8). Each real transition event in **k** space is accompanied by a displacement in **R** space. If the initial state of the electron is described by a narrow wave packet in the band 1 with average momentum **k**, then a direct transition to a higher band 2 is accompanied by a distortion $\delta\langle\hat{\mathbf{r}}\rangle(t)$ of the unperturbed trajectory. Then, besides the usual contribution connected with the change that occurs in the velocity during the transition, there exists a contribution connected with the displacement of the electron in **R** space:

$$\delta\langle\hat{\mathbf{r}}\rangle(t)=W_{21}(\mathbf{k})t[{}^1/{}_2(\mathbf{v}_{2\mathbf{k}}-\mathbf{v}_{1\mathbf{k}})t+\mathbf{R}_{21}^{e}(\mathbf{k})].\tag{12}$$

It is remarkable that this formula can be derived directly on the basis of the elementary perturbation theory, and that only the initial and final states figure in this derivation. Indeed, the mean value of the coordinate operator has the form

$$\langle\hat{\mathbf{r}}\rangle(t)=\sum_{s\mathbf{k}}\left[|a_{s\mathbf{k}}|^2(\boldsymbol{\Omega}_{s\mathbf{k}}+\mathbf{v}_{s\mathbf{k}}t)+ia_{s\mathbf{k}}^{*}\frac{\partial}{\partial\mathbf{k}}a_{s\mathbf{k}}\right].$$

Using for $a_{2\mathbf{k}}$ the standard first-order-perturbation-theory formula, setting, on the basis of the normalization condition, $\mathrm{a}_{1\mathbf{k}}=1-\frac{1}{2}|\mathrm{a}_{2\mathbf{k}}|^2$ and taking into account the narrowness of the wave packet, we immediately arrive at (12).

The displacement $\mathbf{R}_s(\mathbf{k}_2,\mathbf{k}_1)$ associated with the intraband transition from the point $\mathbf{k}_1$ to the point $\mathbf{k}_2$, occurring as a result of the interaction with the phonons and impurities, has a similar meaning. The generalization of (8) to the case of the electron-impurity scattering is obvious:

$$\mathbf{j}^{imp}=e\sum_{s}W_{s}^{imp}(\mathbf{k}_2,\mathbf{k}_1)\mathbf{R}_{s}(\mathbf{k}_2,\mathbf{k}_1)\delta(\varepsilon_{\mathbf{k}_1}^{s}-\varepsilon_{\mathbf{k}_2}^{s})f_{s\mathbf{k}_1}\,d\mathbf{k}_1\,d\mathbf{k}_2;\tag{13}$$

$W_{s}^{\mathrm{imp}}(\mathbf{k}_2,\mathbf{k}_1)$ is the probability for elastic scattering from $\mathbf{k}_1$ to $\mathbf{k}_2$.

Let us discuss the general properties of the displacement PVE. The displacements $\mathbf{R}^e$ and $\mathbf{R}$ are nonzero in noncentrosymmetric crystals. They change sign upon the interchange of the initial and final states:

$$\mathbf{R}_{s_1s_2}(\mathbf{k})=-\mathbf{R}_{s_2s_1}(\mathbf{k}),\quad \mathbf{R}_{s}(\mathbf{k}_1,\mathbf{k}_2)=-\mathbf{R}_{s}(\mathbf{k}_2,\mathbf{k}_1).\tag{14}$$

It can already be seen from this that stationary currents do not arise in the absence in **k** space of fluxes connected with a state-of-nonequilibrium-maintaining source. On account of the fact that the Bloch functions possess the property $\psi_{\mathbf{k}}=\psi^{*}_{-\mathbf{k}}$, which reflects invariance under time reversal, the displacements do not change under the operation of spatial inversion:

$$\mathbf{R}^{e}(\mathbf{k})=\mathbf{R}^{e}(-\mathbf{k}),\quad \mathbf{R}(\mathbf{k}_2,\mathbf{k}_1)=\mathbf{R}(-\mathbf{k}_2,-\mathbf{k}_1).\tag{15}$$

It is not difficult to verify that, under conditions of thermodynamic equilibrium, i.e., when the photon and phonon distributions are Planckian, the integrands in (8) and (13) vanish. If, on the other hand, the distributions $N_{\mathbf{q}\lambda}^{\mathrm{ph}}$ and $N_{\mathbf{q}}^{f}$ are, generally speaking, nonzero. We shall, in investigating the PVE, assume that the phonon distribution function is an equilibrium distribution.

To anlayze the general properties of the PVE further, we must use the kinetic equation for f_{sk}:

$$\partial f_{s\mathbf{k}}/\partial t=I_{s\mathbf{k}},\quad I_{s\mathbf{k}}=I_{s\mathbf{k}}^{\mathrm{ph}}+I_{s\mathbf{k}}^{f}.\tag{16}$$

The collision integrals $I^{\mathrm{ph},f}$ describe the direct interband transitions (with allowance for the spontaneous emission) and the scattering on the phonons.

Using the explicit forms of the $I_{\mathbf{k}}^{\mathrm{ph},f}$, we can easily show that part of the total current which is connected with the diagonal matrix elements $\Omega_s(\mathbf{k})$ has the form

$$\delta\mathbf{j}^{\Omega}=e\sum_{s}\int\boldsymbol{\Omega}_{s}(\mathbf{k})I_{s\mathbf{k}}\,d\mathbf{k}.\tag{17}$$

Since under steady-state conditions $I_{s\mathbf{k}}=0$, the current $\delta\mathbf{j}^{\Omega}$ also vanishes exactly. This result can easily be generalized to the case of the electron-impurity scattering. Thus, in (10) we can eliminate the contributions connected with Ω. But let us note at once that the vanishing of $\delta\mathbf{j}^{\Omega}$ has quite a simple meaning. As can be seen from (10), the contributions $\delta\mathbf{R}_{\Omega}^{e}$ and $\delta\mathbf{R}_{\Omega}$ to the displacement that are connected with $\boldsymbol{\Omega}_s(\mathbf{k})$ can formally be regarded as the differences between the positions of the charge in the final and initial states. Under steady-state conditions such displacements, just like the currents due to the change in polarization, cannot lead to charge transport. As to those contributions to $\mathbf{R}^e$ and $\mathbf{R}$ which are connected with the phase gradients (see (10)), they cannot be represented in the form of the indicated differences, and are therefore not canceled out under steady-state conditions.

Let us further point out the following property of the currents $\mathbf{j}^{\mathrm{ph}}$, $\mathbf{j}^{f}$, $\mathbf{j}^{\mathrm{imp}}$. As is well known, the values of the observable physical quantities should not change when the wave function is multiplied by a constant phase factor, i.e., they should be invariant under the substitution

$$\Psi_{\mathbf{k}}(r)\rightarrow\Psi_{\mathbf{k}}(r)\exp(i\varphi_{\mathbf{k}}).$$

It can be seen from (10) that the displacements $\mathbf{R}^{e}$ and $\mathbf{R}$ (and, consequently, the currents $\mathbf{j}^{\mathrm{ph}}$, $\mathbf{j}^{f}$, and $\mathbf{j}^{\mathrm{imp}}$) are indeed phase-invariant. But each of the terms entering into $\mathbf{R}^{e}$ and $\mathbf{R}$ (in particular, $\delta\mathbf{R}_{\Omega}{}^{e}$ and $\delta\mathbf{R}_{\Omega}$) does not possess this property. Therefore, the contributions to the current that are connected with these terms individually do not have a physical meaning. Notice that none of the contributions to $\mathbf{R}^{e}$ and $\mathbf{R}$ is entirely eliminated by a phase shift. When $\delta\mathbf{j}^{\Omega}$ is discarded, the contributions $\mathbf{j}^{\mathrm{ph}}$, $\mathbf{j}^{f}$, and $\mathbf{j}^{\mathrm{imp}}$ individually lose the property of phase invariance, and the invariance of the sum of these contributions becomes unclear and should be established with the aid of the kinetic equation (16).

Thus far, we have not concretized the form of $N_{\mathbf{q}\lambda}^{\mathrm{ph}}$ in our discussions. We shall now assume that we have in the system, along with the thermal photons, a monochromatic, completely polarized wave of intensity J:

$$N_{\mathbf{q}\lambda}^{\mathrm{ph}}=N_{\mathbf{q}\lambda}{}^{0}+(2\pi)^{3}\frac{nJ}{\hbar\omega c}\delta(\mathbf{q}-\mathbf{q}_{0})\delta_{\lambda\lambda_{0}}. \tag{18}$$

We need to find the current $\mathbf{j}$ at low light intensities. It can be seen from (8) that $\mathbf{j}^{\mathrm{ph}}$ is a sum of two terms:

$$\mathbf{j}^{\mathrm{ph}}=\mathbf{j}^{i}+\mathbf{j}^{\mathrm{rec}}, \tag{19}$$

the first of which is proportional to J, and can be called ionization current. The second term (like $\mathbf{j}^{f}$ and $\mathbf{j}^{\mathrm{imp}}$) does not explicitly depend on the intensity, and has the meaning of recombination current. Thus, as in Ref. 1, the total current is equal to the sum of the ionization, recombination, and scattering-related currents.

In many cases the representation of the current (8) in terms of the moduli and phases of the matrix elements is inconvenient. Below we shall express the currents directly in terms of these complex quantities.

The ionization current should clearly be computed with the unperturbed (Planck) distribution functions:

$$\mathbf{j}_{\alpha}{}^{i}=-\frac{e^{3}Je_{\beta}e_{\gamma}}{2\pi nm^{2}\omega^{2}\hbar c}\sum_{s_{1}s_{2}}\int\mathrm{Im}\left(P_{s_{2}s_{1}}^{\gamma}\frac{\partial}{\partial k_{\alpha}}P_{s_{1}s_{2}}^{\beta}\right)$$
$$\times(\overset{0}{f}_{s_{2}\mathbf{k}}-\overset{0}{f}_{s_{1}\mathbf{k}})\delta(\hbar\omega-\varepsilon_{\mathbf{k}}^{s_{1}s_{2}})d\mathbf{k}. \tag{20}$$

To determined $\mathbf{j}^{\mathrm{rec}}$ and $\mathbf{j}^{f}$, we must find from the kinetic equation (16) the corrections to the equilibrium distribution functions for small J. Performing in (8) the averaging over the polarizations and momenta of the emitted photons, and neglecting the thermal radiation, we obtain

$$j_{\alpha}^{\mathrm{rec}}=\frac{4e^{3}n}{3(2\pi)^{3}m^{2}\hbar^{2}c^{3}}\sum_{s_{1}s_{2}}\int\mathrm{Im}\left(P_{s_{2}s_{1}}^{\gamma}\frac{\partial}{\partial k_{\alpha}}P_{s_{1}s_{2}}^{\gamma}\right)$$
$$\times[f_{s_{1}\mathbf{k}}(1-f_{s_{2}\mathbf{k}})-\overset{0}{f}_{s_{1}\mathbf{k}}(1-\overset{0}{f}_{s_{2}\mathbf{k}})]\varepsilon_{\mathbf{k}}^{s_{1}s_{2}}d\mathbf{k}. \tag{21}$$

As a rule, there exists in semiconductor crystals the hierarchy of times,[12]

$$\tau\gg\tau_{\varepsilon}\gg\tau_{p}; \tag{22}$$

τ is the characteristic time of the interband recombination (the lifetime in a band) and τ_{ε} is the energy relaxation time. Accordingly,

$$f_{s\mathbf{k}}=f_{s\mathbf{k}}{}^{0}+f_{s\mathbf{k}}{}'+\delta f_{s\mathbf{k}}, \tag{23}$$

where $f_{s\mathbf{k}}'(\varepsilon_{\mathbf{k}}^{s})$ describes the thermalized electrons, while $\delta f_{s\mathbf{k}}$ describes the nonequilibrium eletrons, i.e., those electrons which have not had time to relax in terms of momentum and energy after the photoexcitation. Clearly,

$$\delta f\sim f'\tau_{\varepsilon}/\tau\ll f'.$$

Therefore, we need, in computing $\mathbf{j}^{\mathrm{rec}}$, to take only the thermalized electrons into account in the formula (21). Just as in Ref. 1, the direction of $\mathbf{j}^{\mathrm{rec}}$ cannot depend on the polarization of the light. Therefore, this current is nonzero in pyroelectrics, where it is directed along the intrinsic polar directions of the medium.

Further, using the formula (8), we can easily verify that the scattering-related current $\mathbf{j}^{f}$ (as well as $\mathbf{j}^{\mathrm{imp}}$) vanishes identically when $\delta f_{\mathbf{k}}$ is neglected. In other words, only the nonequilibrium electrons make a contribution to the scattering-related current.[3)] Assuming that either $f_{s\mathbf{k}}\ll1$ or $1-f_{s\mathbf{k}}\ll1$ in the semiconductor, i.e., that the bands are either conduction or valence bands, we obtain for the photon contribution to the scattering-related current the expression

$$j^{f}=\frac{ea^{3}}{(2\pi)^{5}\hbar}\sum_{s}(\pm1)\int\mathrm{Im}\left[(V_{\mathbf{k}_{1}\mathbf{k}_{2}}^{s})^{*}\left(\frac{\partial}{\partial\mathbf{k}_{1}}+\frac{\partial}{\partial\mathbf{k}_{2}}\right)V_{\mathbf{k}_{1}\mathbf{k}_{2}}^{s}\right]$$
$$\times[(\delta f_{s\mathbf{k}_{1}}-\delta f_{s\mathbf{k}_{2}})N_{\mathbf{k}_{1}-\mathbf{k}_{2}}^{f}-\delta f_{s\mathbf{k}_{2}}]\delta(\varepsilon_{\mathbf{k}_{1}\mathbf{k}_{2}}^{s}-\hbar\Omega_{\mathbf{k}_{1}-\mathbf{k}_{2}})d\mathbf{k}_{1}d\mathbf{k}_{2}. \tag{24}$$

The upper sign pertains to the conduction bands; the lower sign, to the valence bands. We should, in computing the scattering-related current, bear in mind that the nonequilibrium distribution function contains, generally speaking, both an isotropic, and an anisotropic, part:

$$\delta f_{s\mathbf{k}}=\langle\delta f_{s\mathbf{k}}\rangle+(\delta f_{s\mathbf{k}}-\langle\delta f_{s\mathbf{k}}\rangle);$$

the angle brackets denote averaging over the constant-energy surface. The isotropic component describes the particles that have relaxed in terms of momentum, but have not had time to become thermalized. The corresponding contribution to $\mathbf{j}^{f}$ (just as $\mathbf{j}^{\mathrm{rec}}$) is nonzero only in pyroelectrics when allowance is made for the inelasticity of the scattering. As can be seen from (13) and (24), it strictly vanishes when the scattering is elastic. The second component $\delta f_{s\mathbf{k}}-\langle\delta f_{s\mathbf{k}}\rangle$ exists in the case of anisotropic photoexcitation, and describes the electrons that have not had time to relax in terms of momentum. The corresponding contribution to the current $\mathbf{j}^{f}$ is nonzero in the elastic-scattering case as well; it exists not only in pyroelectric, but also in piezoelectric, crystals.

It should be pointed out that the intraband-scattering-related displacement $\mathbf{R}_{s}(\mathbf{k}_{2},\mathbf{k}_{1})$ is due not only to the asymmetry of the periodic potential of the crystal, but also to the asymmetry of the electron-phonon interaction proper, i.e., to the noninvariance of V_{ef} under space inversion, as well as to the asymmetry of the impurity potential. The above-described general properties of the scattering-related current in the theory of the displacement PVE has a direct similarity to the theory expounded in Ref. 1 .

3. MODEL CALCULATIONS, ESTIMATES

Let the light-induced transitions occur between the valence band of the semiconductor and the conduction band. We shall neglect the anisotropy of the crystal, and assume the electron and hole dispersion laws to be quadratic with masses, m_e and m_h. Furthermore, we shall limit ourselves to the case of low momenta, i.e., to the case in which $ka \ll 1$, where a is the lattice constant. For this purpose, it is sufficient to require that the photoexcited-electron momentum

$$k_0 = \hbar^{-1}[2\mu(\hbar\omega - \Delta)]^{1/2};$$

(where Δ is the forbidden-band width and $\mu = m_e m_h/(m_e + m_h)$ is the reduced mass) be low. In the case of small k, we can use for the matrix element of $\mathbf{P}^{cv}$ the expansion (see also Ref. 1)

$$P_\alpha^{cv}(\mathbf{k}) = i\frac{am\Delta}{\hbar}(fc_\alpha + igak_\alpha + a^2 h_{\alpha\beta\gamma}k_\beta k_\gamma + \dots), \qquad (25)$$
$$h_{\alpha\beta\gamma} = h_{\alpha\gamma\beta} = h_{\beta\alpha\gamma},$$

in which f, g, and h are real dimensionless quantities and $\mathbf{c}$ is the unit vector along the polar direction of the crystal. In nonpyroelectric crystals $f = 0$, and $\hat{h}$ possesses the properties of a piezotensor: $h_{\alpha\beta\beta} = 0$.[4)] In pyroelectrics, $f \neq 0$, and the last term in (25) is unimportant.

We should, in computing $\mathbf{j}^i$ with the aid of the formula (25), take into consideration the fact that the electron-phonon interaction matrix element $V_{\mathbf{k}_1\mathbf{k}_2}$ can diverge at low momenta. Of greatest importance here is the Fröhlich interaction with the longitudinal optic phonons, for which[15]

$$V_{\mathbf{k}_1\mathbf{k}_2} = \frac{V_0}{|\mathbf{k}_1 - \mathbf{k}_2|}A_{\mathbf{k}_1\mathbf{k}_2}, \quad A_{\mathbf{k}_1\mathbf{k}_2} = \frac{1}{a^3}\int_{a^3} u_{\mathbf{k}_1}^* u_{\mathbf{k}_2}\,d\mathbf{r}. \qquad (26)$$

The intraband displacement $\mathbf{R}(\mathbf{k}_2, \mathbf{k}_1)$ can then be expressed in terms of the phase of the quantity $A_{\mathbf{k}_1\mathbf{k}_2}$, which is determined by the periodic crystal potential. Expanding $A_{\mathbf{k}_1\mathbf{k}_2}$ up to terms quadratic in $\mathbf{k}$, and neglecting the anisotropy, we obtain

$$\mathrm{Im}\left[(V_{\mathbf{k}_1\mathbf{k}_2}^{c,v})^*\left(\frac{\partial}{\partial\mathbf{k}_1} + \frac{\partial}{\partial\mathbf{k}_2}\right)V_{\mathbf{k}_1\mathbf{k}_2}^{c,v}\right]$$
$$= |V_0|^2 a^3 d^{c,v}\frac{(\mathbf{c}, \mathbf{k}_1 - \mathbf{k}_2)(\mathbf{k}_1 + \mathbf{k}_2)}{|\mathbf{k}_1 - \mathbf{k}_2|^2}; \qquad (27)$$

$d^{c,v}$ are dimensionless constants.

The corrections to the equilibrium distribution functions are found from the kinetic equation in the two-band approximation. In computing $\mathbf{j}^{\mathrm{rev}}$, we can consider the electrons and holes to be thermalized, i.e., assume that

$$f_{e\mathbf{k}}f_{h\mathbf{k}} - f_{e\mathbf{k}}^0 f_{h\mathbf{k}}^0 \propto \exp(-\varepsilon_\mathbf{k}/T), \qquad (28)$$

where $f_{e\mathbf{k}} = f_{c\mathbf{k}}$ and $f_{h\mathbf{k}} = 1 - f_{v\mathbf{k}}$ are the electron and hole distribution functions and $\varepsilon_\mathbf{k} = k^2\hbar^2/2\mu$ is the reduced energy. The coefficient of proportionality in (28) does not depend on the form of the electron-phonon interaction, and is determined by the interband recombination.

In computing the scattering-related current, we shall, for simplicity, limit ourselves to the inelastic contribution. The nonequilibrium corrections $\delta f_e(\varepsilon_\mathbf{k}^e)$ and $\delta f_h(\varepsilon_\mathbf{k}^h)$ necessary for this purpose are determined by the rate of intraband relaxation in terms of energy. If for the electrons and holes $\varepsilon_{e,h} \gg T$, $\hbar\Omega$, then we find from kinetic equation (16) in the case of the Fröhlich interaction that[16]

$$\alpha\Omega\frac{(\hbar\Omega)^{3/2}}{\varepsilon_{e,h}^{1/2}}\frac{\partial}{\partial\varepsilon_{e,h}}\left[\ln\left(\frac{4\varepsilon_{e,h}}{\hbar\Omega}\right)\delta f_{e,h}\right]$$
$$= \frac{(2\pi)^5 e^2 J}{m^2\omega^2\hbar c}|\mathbf{eP}_\mathbf{k}^{cv}|^2\delta(\varepsilon_\mathbf{k} - \varepsilon_{\mathbf{k}_0}), \qquad (29)$$

where α is the dimensionless electron-phonon interaction constant and $\varepsilon_{e,h}$ are measured from the band edges. Equation (29) describes the photoexcitation of particles with momentum k_0 and their transfer into the low-energy region as a result of inelastic scattering. It is not difficult to find that, for $k \leqslant k_0$,

$$\delta f_{e,h} = \left(\frac{\hbar}{2\Omega m_{e,h}}\right)^{3/2}\frac{(2\pi)^2}{\alpha\Omega}\frac{\varkappa J}{\hbar\omega}\ln^{-1}\left(\frac{4\varepsilon_{e,h}}{\hbar\Omega}\right), \qquad (30)$$

where $\varkappa$ is the light-absorption coefficient:

$$\varkappa = \frac{2n\mu e^2 k_0}{\hbar^2 m^2\omega c}\left[f^2(\mathbf{ce})^2 + \frac{1}{3}k_0^2 a^2 g^2\right]. \qquad (31)$$

In the region $k > k_0$ the quantity $f_{e,h} = 0$.

Taking the foregoing into account, we easily obtain explicit expressions for the current. Let us first consider the current $\mathbf{j}^{\mathrm{ph}}$. In piezoelectrics, $\mathbf{j}^{\mathrm{ph}} = \mathbf{j}^i$; in this case $f = 0$ in (25) and (31), and the expression for the current assumes an especially simple form:

$$j_\alpha^{\mathrm{ph}} = 2ea(\varkappa J/\hbar\omega)g^{-1}h_{\alpha\beta\gamma}e_\beta e_\gamma. \qquad (32)$$

Since $h_{\alpha\beta\beta} = 0$ in a piezoelectric, $\mathbf{j}^{\mathrm{ph}}$ vanishes upon being averaged over the polarizations and directions of the light.

In pyroelectric crystals

$$\mathbf{j}^{\mathrm{ph}} = -ea\frac{\varkappa_\| J}{\hbar\omega}fg\left[\frac{\mathbf{e}(\mathbf{ce})}{f^2 + {}^1/_3 g^2 k_0^2 a^2} - \frac{\varkappa}{\varkappa_\|}\frac{\mathbf{c}}{f^2 + {}^3/_2 g^2 k_T^2 a^2}\right]; \qquad (33)$$

where $\varkappa_\|$ is the value of the absorption coefficient $\varkappa$ for $\mathbf{e}\|\mathbf{c}$ and $k_T = \hbar^{-1}(2\mu T)^{1/2}$ is the thermal momentum. The recombination current (the second term in (33)) depends on the polarization only through the absorption coefficient $\varkappa$. The value of the current (33) averaged over the light polarizations and directions is nonzero: $\langle\mathbf{j}\rangle \propto \mathbf{c}$. In (33) we have retained the terms that are quadratic in $\mathbf{k}$. The point is that one of the parameters f and g in the expansion (25) characterizes the degree of asymmetry of the crystal, and should be small. If, for example, the c and v bands are close to the s bands, then $f \ll g$. Therefore, there can be situations in which $f/g \lesssim ka \ll 1$. The relation between $\mathbf{j}^i$ and $\mathbf{j}^{\mathrm{rec}}$ is different from the relation found in the earlier models for the PVE.[1] If k_T, $k_0 \lesssim f/ga$, then $\mathbf{j}^i \sim \mathbf{j}^{\mathrm{rec}}$, $\mathbf{j}^{\mathrm{rec}}$ being independent of the crystal temperature. As k_0 (or the light frequency) increases, the ionization contribution decreases, and the recombination current becomes dominant. Let us also note that, in the model considered, $\mathbf{j}^i$ does not possess a component $\delta\mathbf{j}^i \propto \mathbf{c}$, which is allowed by the symmetry. Such a contribution arises only when allowance is made for corrections of higher order in the parameter ka.

Using (24), (27), and (30), we can easily compute the inelastic contribution to the scattering-related current:

$$j_{\text{in}}{}^{f}=-\frac{ea}{3}\frac{\varkappa J}{\hbar\omega}(d^{v}+d^{c})k_0{}^2a^2\mathbf{c}. \tag{34}$$

The elastic contribution to $\mathbf{j}^f$ has, generally speaking, the same order of magnitude as the inelastic contribution, but its polarization dependence is more complicated. The scattering-related current strength does not depend on the electron-phonon interaction parameters (the phonon frequency and the coupling constant), and is determined only by the properties of the periodic crystal potential. In spite of the fact that $\mathbf{j}^f$ is due to the nonthermalized electrons and holes, it is not of the order of the parameter $\tau_\varepsilon/\tau \ll 1$ in smallness. But at low momentum values $\mathbf{j}^f$ is of the order of $k_0^2a^2 \ll 1$ as compared to $\mathbf{j}^{\text{ph}}$. This smallness is, in the final analysis, due to the smallness of the intraband-transition-related displacements $\mathbf{R}_e$ and $\mathbf{R}_h$ as compared to the photoexcitation-related displacement $\mathbf{R}^e$.

It can be seen from the explicit expressions (33) and (34) for the current that the displacement photocurrent has the form

$$\mathbf{j}=e\frac{\varkappa J}{\hbar\omega}\delta\mathbf{R}. \tag{35}$$

The quantity $\delta\mathbf{R}$ has the meaning of electron displacement per absorbed photon. For the purpose of making a rough estimate, we can set $j\sim g$, and then $\delta\mathbf{R}\sim a$, the lattice constant. There can, however, be situations in which $\delta R \gg a$. As can be seen from (33), when $k_0a\sim f/g$, the displacement δR can be of the order of the de Broglie wavelength of the electron: $\delta R_{\max}\sim 2\pi/k_0$. As $f/g\to 0$, the displacment $\delta R_{\max}\to\infty$; the total current then tends, of course, to zero.

Let us consider the question of the relative magnitudes of the diagonal and off-diagonal contributions to the current. Since under conditions when the kinetic equation is applicable the electron (hole) mean free path $l\gg 2\pi/k\gtrsim a$, the conclusion that the displacement PVE is weak suggests itself. Such a general conclusion is incorrect. Indeed, the current ratio

$$j^p/j\sim\delta R/\xi l, \tag{36}$$

where $\xi \ll 1$ is the asymmetry parameter, i.e., the fraction particles flying asymmetrically after the photoexcitation. The quantity ξ depends essentially on the mechanism underlying the PVE. In the case of interband transitions there are two principal mechanisms: the phonon and Coulomb mechanism.[1] The phonon mechanism makes, in its turn, two *independent* contributions to the current. One contribution, $\mathbf{j}_1^f$, is due to the asymmetry of the electron-phonon interaction; the second contribution, $\mathbf{j}_2^f$, is connected directly with the asymmetry of the crystal potential, and is determined by the same parameters that determine $\mathbf{j}^p$. The electron-phonon interaction is taken into consideration twice in the computation of $\mathbf{j}_2^f$: once in the computation of the asymmetric probabilities W^{as}; the second time, in the computation of the momentum relaxation time τ_p. Since $W^{\text{as}}\propto\alpha$ and $\tau_p\propto\alpha^{-1}$, the current $\mathbf{j}_2^f\propto W^{\text{as}}\tau_p$ does not depend at all on the electron-phonon coupling constant. Therefore, there are no general reasons why the displacement PVE should be weak. Computations based on the formulas of Ref. 17 show that, near the interband absorption edge,

$$j^p/j_2{}^f\approx {}^3/{}_{16}\pi,$$

i.e., the displacement photocurrent is smaller by roughly a factor of two than the current given by theory expounded in Ref. 1.

In the case of the Coulomb mechanism for the PVE the photoexcitation asymmetry is due to the Coulomb attraction of the electron-hole pair formed, whereas τ_p is determined as before by the collisions with the phonons. If the Coulomb mechanism predominates over the phonon mechanism, i.e., if $j^C > j^f$, then the displacment PVE is weak, i.e., $j^p \ll j$.

4. IMPURITY-BAND TRANSITIONS

The periodicity of the crystal potential was essentially not used in the derivation of the basic formulas (20), (21), and (24). Therefore, the theory can easily be generalized to the case of the impurity-band transitions. For definiteness, let the transitions occur between a deep-lying impurity level and the conduction band. Neglecting the scattering-related current, which is weak in the vicinity of the absorption edge, we find that at low impurity concentrations

$$\mathbf{j}_\alpha=-\frac{e^3a^3N_0J}{2\pi m^2\omega^2\hbar c}e_\beta e_\gamma\int \operatorname{Im}\left(P_\gamma^{*}\frac{\partial}{\partial k_\alpha}P_\beta\right)\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}_0})\,d\mathbf{k}$$
$$+\frac{4}{3}\frac{e^3a^3\omega(N-N_0)}{m^2\hbar c^3(2\pi)^3}\int \operatorname{Im}\left(P_\beta^{*}\frac{\partial}{\partial k_\alpha}P_\beta\right)(f_{\mathbf{k}}-f_{\mathbf{k}}{}^0)\,d\mathbf{k}. \tag{37}$$

Here N is the number of impurities, N_0 is the number of electrons on an impurity, and $\varepsilon_{\mathbf{k}}$ is the electrons energy, measured from the bottom of the band. The matrix element $\mathbf{P}(\mathbf{k})=\langle\mathbf{k}|\mathbf{P}|0\rangle$ should be computed with the exact wave functions, $\psi_0(\mathbf{r})$ and $\psi_{\mathbf{k}}(\mathbf{r})$, of the localized initial state and the continuous spectrum. As is well known, there exist two main sets of continuous-spectrum wave functions $\psi_{\mathbf{k}}^{\pm}(\mathbf{r})$, which contain diverging and converging waves at great distances from the defect.[18] The difference between ψ^+ and ψ^- stems from the fact that the poles in an integral equation of the Lippmann-Schwinger type are bypassed in different directions.[19] It can be shown that this difference is unimportant for the theory of displacment PVE: as $\psi_{\mathbf{k}}$, we can choose any of the functions $\psi_{\mathbf{k}}^{\pm}$. This circumstance is not accidental: it follows from the general transformation properties of the kinetic equations under time reversal.

In the case of small k we can again use the expansion (25), but the coefficients g and h in it are complex and (like $f=f^*$) characterize not only the properties of the crystal, but also the impurity potential. The expression (32) and (33) for the current preserve their form if we make the substitutions $g, h\to\operatorname{Re} g, \operatorname{Re} h$ in them and take $\varkappa$ as the absorption coefficient for the impurity-band transition.

In the general case the individual properties of the impurity cannot be separated, and f, g, and h should be regarded as phenomenological parameters. The situation changes in the case of shallow impurities, for the computation of which methods of the effective-mass type are available.[20] For example, the dipole-impurity model is valid for shallow donor levels in a pyroelectric.[14] In this case the asymmetry of the periodic potential does not make a contribution to the current, and the displacement turns out to be connected with the impurity potential. The quantity f in the expansion (25) is determined by the contribution from the principal values of the perturbation theory integrals. For the transitions from the s states of the impurity

$$f/g=4da/3ea_Br_0, \tag{38}$$

where d is the dipole moment of the impurity, $a_B=\hbar^2\bar{\varepsilon}/me^2$ is the Bohr radius, $\bar{\varepsilon}$ is the static dielectric constant, and r_0 is the radius (determined by the depth at which the level occurs) of localization of the electron on the impurity. A comparison of the magnitudes of the diagonal and off-diagonal contributions to the current shows that, in the present model, $j/j^p\sim k_0l\gg 1$.

5. CONCLUSION

It is safe to assert that, besides the PVE theory expounded in Refs. 1 and 2, there exists an independent intrinsically consistent displacement-PVE theory. Above this theory was constructed for the direct interband transitions and in part for the impurity-band transitions. Other cases of current interest—the effect connected with free carriers, indirect transitions, the influence of a magnetic field, etc.—are yet to be considered. There is also no doubt that the displacement contributions occur not only in the theory of the PVE, but also in the theory of other transport phenomena, e.g., the photon drag effect, the quadratic—in the field—electrical conductivity, etc.

Of great interest is the problem of the experimental detection of the displacement PVE. Since from the point of view of the phenomenological theory the two types of PVE are identical, the separation of the contributions to the current should be carried out on the basis of the characteristics of the microscopic mechanisms. The difference between the effects can manifest itself in the kinetics of the establishment of the currents in the case when the light is switched on instantaneously, as well as in the influence of the spatial light-intensity inhomogeneity and the properties of the crystal. Since j^p is not connected with the kinetic parameters of the crystal, its establishment (more precisely, the establishment of the ionization contribution) occurs in atomic times; as to the influence of the inhomogenity, it is insignificant right up to the point where the characteristic scales of the inhomogeneity are comparable to the dimension of the unit cell. The difference between the two effects can also manifest itself in their frequency dependences. Thus, for example, in the case of the interband transitions in GaAs the photocurrent j should undergo frequency oscillations connected with the characteristics of the processes of electron scattering on the optical phonons.[21] The exstence of these oscillations, whose experimental detection is reported in Ref. 21, indicates the collisionless character of the PVE. At present there are, apparently, no experimental data indicating the existence of the displacment PVE.

APPENDIX

Let us rewrite the commutation relation $[X_\alpha,P_\beta]=i\hbar\delta_{\alpha\beta}$ in the basis of the Bloch functions:

$$\sum_{\mathbf{n}}(\langle\mathbf{n}_1|X_\alpha|\mathbf{n}\rangle\langle\mathbf{n}|P_\beta|\mathbf{n}_2\rangle-\langle\mathbf{n}_1|P_\beta|\mathbf{n}\rangle\langle\mathbf{n}|X_\alpha|\mathbf{n}_2\rangle)=i\hbar\delta_{\alpha\beta}\delta_{\mathbf{n}_1\mathbf{n}_2}. \tag{A.1}$$

We can write down the identity $[X_\alpha,V_{\text{ef}}]=0$ in a similar fashion. For momentum and coordinate matrix elements we have the relations[11]

$$\begin{aligned}\langle s\mathbf{k}|X_\alpha|s_1\mathbf{k}_1\rangle&=i\delta_{ss_1}\frac{\partial}{\partial k_\alpha}\delta_{\mathbf{k}\mathbf{k}_1}+\Omega_\alpha^{ss_1}(\mathbf{k})\delta_{\mathbf{k}\mathbf{k}_1},\\ \langle s\mathbf{k}|P_\alpha|s_1\mathbf{k}_1\rangle&=P^\alpha_{ss_1}\delta_{\mathbf{k}\mathbf{k}_1},\quad P_{ss}{}^\alpha=\frac{m}{\hbar}\frac{\partial}{\partial k_\alpha}\varepsilon_{\mathbf{k}}{}^s.\end{aligned} \tag{A.2}$$

The off-diagonal (in the band numbers) elements of the matrix $\Omega_\alpha^{ss_1}$ can be expressed in terms of the momentum matrix elements:

$$\Omega_\alpha^{ss_1}=i\frac{\hbar}{m}\frac{P^\alpha_{ss_1}}{\varepsilon_{\mathbf{k}}{}^{s_1}-\varepsilon_{\mathbf{k}}{}^s}. \tag{A.3}$$

Separating out the terms with $s=s_1$, s_2 from the sums over the band numbers, using (A.2) and (A.3) and summing over $\mathbf{k}_2$ in (A.1), we easily obtain the following exact relations:

$$\begin{aligned}\sum_{s\neq s_1}X^\alpha_{s_1s}P^\beta_{ss_2}-\sum_{s\neq s_2}P^\beta_{s_1s}X^\alpha_{ss_2}&=-i\frac{\partial P^\beta_{s_1s_2}}{\partial k}+P^\beta_{s_1s_2}(\Omega_\alpha{}^{s_2}-\Omega_\alpha{}^{s_1})\\&\quad+i\hbar\delta_{\alpha\beta}\delta_{s_1s_2};\\ \sum_{\mathbf{k},s\neq s_1}X^\alpha_{\mathbf{n}_1\mathbf{n}}V_{\mathbf{n}\mathbf{n}_2}-\sum_{\mathbf{k},s\neq s_2}V_{\mathbf{n}_1\mathbf{n}}X^\alpha_{\mathbf{n}\mathbf{n}_2}&=V_{\mathbf{n}_1\mathbf{n}_2}(\Omega_\alpha{}^{n_2}-\Omega_\alpha{}^{n_1})\\&\quad-i\left(\frac{\partial}{\partial k_{1\alpha}}+\frac{\partial}{\partial k_{2\alpha}}\right)V_{\mathbf{n}_1\mathbf{n}_2}.\end{aligned} \tag{A.4}$$

The authors are grateful to G. E. Pikus for a discussion and valuable comments.

[1)] Reference 8 also contains an obvious error: the chosen basic Hamiltonian is not invariant under time reversal.

[2)] The field components with polarizations $\lambda=1,2$ are assumed to be incoherent.

[3)] Just as in the computation of the diagonal contribution.[1]

[4)] Strictly speaking, there can be situations in which $fc_\alpha\neq 0$ in piezoelectrics as well, and then g in the formula (25) is perforce a tensor.[13,14]

[1] V. I. Belinicher and B. I. Sturman, Usp. Fiz. Nauk **130**, 415 (1980) [Sov. Phys. Usp. **23**, 199 (1980].

[2] E. L. Ivchenko and G. E. Pikus, in: Problemy sovremennoĭ fiziki (Problems of Modern Physics), Nauka, Leningrad, 1980, p. 275.

[3] P. M. Mednis, Pis'ma Zh. Eksp. Teor. Fiz. **7**, 355 (1968) [JETP Lett. **7**, 278 (1968)].

[4] V. I. Belinicher, Zh. Eksp. Teor. Fiz. **75**, 641 (1978) [Sov. Phys. JETP **48**, 322 (1978)]; Fiz. Tverd. Tela (Leningrad) **23**, 1229 (1981) [Sov. Phys. Solid State **23**, 723 (1981)].

[5] B. I. Sturman, Zh. Eksp. Teor. Fiz. **75**, 673 (1978) [Sov. Phys. JETP **48**, 339 (1978)]; Fiz. Tverd. Tela (Leningrad) **23**, 3686 (1981) [Sov. Phys. Solid State **23**, 2143 (1981)].

[6] N. Kristofel' and A. Gulbis, Izv. Akad. Nauk Est. SSR **28**, 268 (1979).

[7] W. Krant and R. Von Baltz, Phys. Lett. A **79**, 364 (1980).

[8] Ya. G. Girshberg and N. N. Trunov, Doklad na IX Soveschchanii po segnetoélektrichestvy (Paper Presented *at the Ninth Conference on Ferroelectricity*), Rostov-on-Don, 1979; É. V. Bursian, Ya.G. Girshberg, and N. N. Trunov, Izv. Vyssh. Uchebn. Zaved. **24**, 94 (1981).

[9] Transl. in; Voprosy kvantovoĭ teorii neobratimykh protsessov (Collected Papers on the Problems of the Quantum Theory of Irreversible Processes), IIL, Moscow, 1961.

[10] V. B. Berestetskiĭ, E. M. Lifshitz, and L. P. Pitaevskiĭ, Relyativists-kaya kvantovaya teoriya (Relativistic Quantum Theory), Nauka, Moscow, 1968 (Eng. Transl., Pergamon, Oxford, 1971).

[11] L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics), Nauka, Moscow, 1964 (Eng. Transl., Pergamon, Oxford, 1969), p. 251.

[12] S. M. Ryvkin, Fotoélektricheskie yavleniya v poluprovodnikakh (Photo-electric Effects in Semiconductors), Fizmatgiz, Moscow, 1963 (Eng. Transl., Consultants Bureau, New York, 1964).

[13] Yu. I. Sirotin and M. P. Shaskol'skaya, Osnovy kristallofiziki (Principles of Crystal Physics), Nauka, Moscow, 1975.

[14] B. I. Sturman, Fiz. Tverd. Tela (Leningrad) **22**, 3084 (1980) [Sov. Phys. Solid State **22**, 1801 (1980)].

[15] O. Madelung, Introduction to Solid State Theory, Springer-Verlag, New York, 1978 (Russ. Transl., Nauka, Moscow, 1980, p. 200).

[16] V. K. Malinovskiĭ and B. I. Sturman, Fiz. Tverd, Tela (Leningrad) **22**, 3678 (1980) [Sov. Phys. Solid State **22**, 2153 (1980)].

[17] V. I. Belinicher and B. I. Sturman, Fiz. Tverd. Tela (Leningrad) **20**, 821 (1978) [Sov. Phys. Solid State **20**, 476 (1978)].

[18] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Nauka, Moscow, 1974 (Eng. Transl., Pergamon, Oxford, 1977), p. 643.

[19] A. Messiah, Quantum Mechanics, Vol. II, Wiley, New York, 1963 (Russ. Transl., Nauka, Moscow, 1979, p. 300).

[20] A. M. Stoneham, Theory of Defects in Solids: The Electronic Structure of Defects in Insulators and Semiconductors, Clarendon Press, Oxford, 1975 (Russ. Transl., Nauka, Moscow, 1978).

[21] V. L. Al'perovich, V. I. Belinicher, V. N. Novikov, and A. S. Terrekhov, Ferroelectrics (in press).

Translated by A. K. Agyei