Theory of the coherent photovoltaic effect and the method of nonequilibrium Green's functions

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(Submitted 22 July 1991)
Zh. Eksp. Teor. Fiz. **101**, 1749–1771 (June 1992)

An expression for the DC density generated by a coherent pump field and its second harmonic due to impurity-band transitions is derived. The method used in obtaining the expression is that of nonequilibrium Green's functions. The estimates given show that the coherent photovoltaic effect can play a key role in explaining the photoinduced second-harmonic generation in quartz-glass-based optical fiber guides.

INTRODUCTION

In 1986, Osterberg and Margulis reported on the first observation of effective second-harmonic generation in quartz-glass based optical fiber guides.[1] Since then this phenomenon has been under intense investigation since it is not only of interest for technological reasons but also is of fundamental value, because in glass such generation should not be present because the nonlinear second-order susceptibility vanishes. Experimental data fits the general model, in which it is assumed that under laser radiation a nonlinear second-order susceptibility grating is written in the guide.

There are several qualitatively different models for explaining the emergence of a nonzero second-order susceptibility in glass as an effect of laser radiation (see, e.g., the review paper by Sokolov and Sulimov[2]). Among these is the model of Dianov, Kazanskiĭ, and Stepanov,[3] according to which a strong transverse electrostatic field $\mathscr{E}_{DC}$ of the order of 10^4–10^5 V cm^{-1} is generated in the guide. The reason is the spatial redistribution of charges between traps, caused by the coherent photovoltaic effect initiated by the photoionization of impurities in the glass of the fiber core. The coherent photovoltaic effect consists in the appearance of DC when the pumping radiation is coherently superimposed on the radiation's second harmonic and may occur in centrally symmetric media such as, say, glass, in contrast to the usual photovoltaic effect, which is realized only in crystals without an inversion center.[4,5]

The possibility of such an effect manifesting itself in an insulator was considered theoretically by Èntin.[6] For semiconductors the theory of the effect was examined earlier by Shmelev *et al.*[7] and by Baskin and Èntin.[8] Zel'dovich *et al.*[9–12] considered the generation of DC by the interference of two photoionization channels: the single-photon channel in the second harmonic and the two-photon channel in the pumping radiation and gave the results of an experiment in observing this mechanism in the photoemissive effect.

The theory of the coherent photovoltaic effect developed by Èntin[6] is based on the kinetic equation. It appears that in considering impurity–band transitions Èntin was insufficiently thorough in separating the contributions to the effect from the field term and from the collision integral. In addition, the final expression for the DC is written in a rather complicated form, which makes it difficult to estimate the effect numerically.

The authors of Refs. 9–12 considered the interference of quantum mechanical transition amplitudes and the associated probability but failed to give the procedure for calculating DC proper in the photoionization of impurities.

These drawbacks of the cited papers devoted to the theory of the coherent photovoltaic effect, along with the growing interest in this effect among researchers studying second-harmonic generation in glass optical fiber guides,[13–20] prompted the present author to return once more to calculations of DC induced in a solid by the pumping radiation and its coherent second harmonic. He believes it expedient to employ not a kinetic equation but the method of nonequilibrium Green's functions introduced by Keldysh,[21] which can be applied to a much broader range of phenomena (say, when only localized states participate in the photoexcitation process and the kinetic-equation method does not work). Such a method also allows the contributions from the field and the generation to be described in a unified manner.

Let us consider a semiconductor or insulator that in addition to distributed band states contains localized states, which correspond to impurity or defect centers of some kind. Suppose that intense monochromatic laser light (pumping radiation) of frequency Ω is incident on this system. We will assume that because of the radiation the respective electron subsystem is transformed to a nonequilibrium time-independent state, and energy dissipation will be taken into account by electron–phonon interaction. Radiation of double frequency 2Ω (the second harmonic of the pumping radiation) is incident on this nonequilibrium system. Our goal is to calculate the time-constant DC density linear in the second-harmonic field.

LINEAR RESPONSE AND THE TWO-PARTICLE GREEN'S FUNCTION

The Hamiltonian of this system is

$$\hat{H}=\hat{H}_0+\hat{H}_i+\hat{H}_\Omega+\hat{H}_{2\Omega}. \tag{1}$$

Here $\hat{H}_0$ is the Hamiltonian of the electron system in the static ion field, $\hat{H}_i$ is the electron–photon interaction Hamiltonian,

$$\hat{H}_i=g\int d\mathbf{x}\psi^+\psi\chi, \tag{2}$$

where ψ^+ and ψ are the electron field operators, χ is the phonon-field Bose operator, g is the electron–phonon coupling constant, and $\hat{H}_\Omega$ and $\hat{H}_{2\Omega}$ are the Hamiltonians of the interaction between electrons and the pump field (of frequency Ω) and its second harmonic (of frequency 2Ω), re-

 0038-5646/92/060932-13$05.00

spectively, which we take in the form

$$\hat{H}_\Omega = -(e/c)(\mathbf{A}_\Omega, \mathbf{v}), \tag{3}$$

$$\hat{H}_{2\Omega} = -(e/c)(\mathbf{A}_{2\Omega}, \mathbf{v}), \tag{4}$$

with $\mathbf{A}_\Omega$ and $\mathbf{A}_{2\Omega}$ the vector potentials, $\mathbf{v}$ the electron velocity operator, e the electron charge, and c the speed of light.

The electromagnetic pump field and its second harmonic are assumed monochromatic and can be written in general form as

$$\mathbf{A}_\Omega(t) = (c/2\Omega i)[\vec{\mathscr{E}}_\Omega \exp(i\Omega t) - \vec{\mathscr{E}}_\Omega^* \exp(-i\Omega t)], \tag{5}$$

$$\mathbf{A}_{2\Omega}(t) = (c/4\Omega)[\vec{\mathscr{E}}_{2\Omega} \exp(i2\Omega t) + \vec{\mathscr{E}}_{2\Omega}^* \exp(-i2\Omega t)], \tag{6}$$

where $\vec{\mathscr{E}}_\Omega$ and $\vec{\mathscr{E}}_{2\Omega}$ are the complex-valued amplitudes of the electric pump field and the second harmonic, respectively.

By linearizing the total density matrix of the system in the second-harmonic radiation field we can easily arrive at an expression for the average value of the βth component ($\beta = x,y,z$) of the current density:

$$j^\beta(\mathbf{x}t) = (1/i\hbar)\int_{-\infty}^{+\infty} dt' \theta(t-t') \langle [\hat{j}^\beta(\mathbf{x}t), \hat{H}_{2\Omega}(t')]_- \rangle, \tag{7}$$

where $\hat{j}^\beta(\mathbf{x}t)$ is the operator of the βth component of the current density,

$$\hat{j}^\beta(\mathbf{x}t) = \frac{ie\hbar}{2m}\left(\frac{\partial\psi^+(x)}{\partial x^\beta}\psi(x) - \psi^+(x)\frac{\partial\psi(x)}{\partial x^\beta}\right) - \frac{e^2}{mc}A^\beta\psi^+(x)\psi(x), \tag{8}$$

$[...]_-$ stands for a commutator, $\langle ... \rangle$ for an average over the state of the system in the absence of second-harmonic radiation, $\theta(t-t')$ for the unit step function, and $\hbar$ for Planck's constant.

Since we are interested in the value of the current density averaged over a macroscopic volume V, we substitute (4) into (7) and transform to the λ-representation, in which the set of eigenfunctions $\{\varphi_\lambda(\mathbf{x})\}$ of $\hat{H}_0$ is used as the basis. Assuming summation over the repeated index α and setting $\hbar = 1$, we get

$$j^\beta(t) = (1/iV)\sum_{\substack{1,2\\3,4}}\int_{-\infty}^{\infty} dt' [j_{42}^\beta - (e^2/mc)A_\Omega^\beta(t)\delta_{42}] \times (-e/c)A_{2\Omega}^\alpha(t') \times [v_{31}^\alpha - (e/mc)A_\Omega^\alpha(t')\delta_{31}]\theta(t-t') \times \langle [a_4^+(t)a_2(t), a_3^+(t')a_1(t')]_- \rangle, \tag{9}$$

where $\alpha_\lambda^+(t)$ and $\alpha_\lambda(t)$ are the Heisenberg λ-state electron creation and annihilation operators, and $j_{42}^\beta = \langle\lambda_4|\hat{j}^\beta|\lambda_2\rangle$ and v_{31}^α the matrix elements of the respective components of the current density operator and the velocity operator that do not incorporate the dependence of the pump field on the vector potential [the corresponding terms are written explicitly in (9)]. For the sake of brevity, in (9) and in what follows we use the integers 1, 2, 3, and 4 instead of the indices $\lambda_1, \lambda_2, \lambda_3$, and λ_4.

In Refs. 22 and 23 it was shown that the term in Eq. (9) containing the step function of the time difference $t - t'$ and the average of the commutation relation for the creation and annihilation operators can be written as

$$\begin{aligned}\theta(t-t')\langle [a_4^+(t)a_2(t), a_3^+(t')a_1(t')]_- \rangle &= -(1/2)G^{II}(1t', 2t; 3t_3, 4t_4) \quad (t_3 \to t', t_4 \to t) \\ &= -(1/2)[G_{22}^{II}(1t', 2t; 3t_3, 4t_4) - G_{22}^{II^*}(3t_3, 4t_4; 1t', 2t)], \quad (t_3 \to t'+0,\ t_4 \to t-0) \quad (t_3 \to t'-0,\ t_4 \to t+0)\end{aligned} \tag{10}$$

where G_{22}^{II} is the two-particle nonequilibrium Green's function, introduced in the following manner.[22,23] In accordance with the Keldysh technique,[21] we introduce the causal two-particle Green's functions

$$G_{ij,kl}(x_1, x_2; x_3, x_4) = \langle T_C[\psi(x_1)\psi(x_2)\psi^+(x_3)\psi^+(x_4)]\rangle, \tag{11}$$

where the operator $T_C[...]$ performs T-ordering along the contour C, which passes along the time axis from $-\infty$ to $+\infty$ and back. The subscript i assumes values 1 or 2, depending on whether the temporal component of the first independent variable x_1 lies on the direct or the return branch of contour C. The same correspondence exists between indices j, k, and l and the positions of the temporal components of the other independent variables x_2, x_3, and x_4. From casual functions we go to retarded and advanced functions via the following transformation.[21-23]

$$G_{1n}^{II} = U_{1i}G_{1i,1j}U_{jn}^{-1}, \tag{12}$$

where the summation convention is assumed and the matrices U and U^{-1} have the form

$$U = \frac{1}{2^{1/2}}\begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix}, \quad U^{-1} = \frac{1}{2^{1/2}}\begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}.$$

The second equality in (10) defines function G^{II} and the exact meaning of function G_{22}^{II} for concurrent times.

Thus, in accordance with Eqs. (9) and (10), to calculate the current density we must find the two-particle Green's function in the nonequilibrium state created in the system by the pumping radiation. We start by considering the spectral representation of the nonequilibrium two-particle Green's function.[22,23]

SPECTRAL REPRESENTATIONS

An arbitrary two-particle function $G^{II}(t_1t_2;t_3,t_4)$ in an equilibrium system depends on any three time differences, say, $\tau = t_1 - t_4$, $\tau' = t_2 - t_3$, and $\tau'' = t_1 - t_3$. In the time-independent nonequilibrium state created in the electron subsystem by the monochromatic pumping radiation of frequency Ω, homogeneity in time is, apparently, violated only by periodic variations (with the period of the pumping radiation). In this case any two-particle function can be represented as a Fourier series in harmonics that are integer multiples of Ω:

$$G^{II}(t_1, t_2; t_3, t_4)=\sum_{n=-\infty}^{\infty} \exp(-in\Omega t_3)\, G^{II}(\tau, \tau'; \tau''; n). \quad (13)$$

In turn, we can represent the amplitude of nth harmonic, $G^{II}(\tau,\tau';\tau'';n)$, in the form of a triple Fourier integral:

$$G^{II}(t_1, t_2; t_3, t_4)$$
$$=\sum_{n=-\infty}^{+\infty} \exp(-in\Omega t_3) \int \frac{dE\, dE'\, d\xi}{(2\pi)^3} \exp(-iE\tau - iE'\tau' - i\xi\tau'')$$
$$\times G^{II}(E, E'; \xi; n). \quad (14)$$

We must now pass to the limit in (14), as required by Eqs. (9) and (10); $t_4 \to t_2 = t$ and $t_3 \to t_1 = t'$. As a result we arrive at the final expression for the spectral representation of the two-particle function:

$$G^{II}(1t_1, 2t_2; 3t_3, 4t_4)$$
$$(t_4 \to t_2 \to t,\ t_3 \to t_1 \to t')$$
$$=\sum_{n=-\infty}^{n=+\infty} \exp(-in\Omega t') \int_{-\infty}^{+\infty} \frac{dE\, dE'}{(2\pi)^2} \exp[-i(E-E')(t'-t)]$$
$$\times G^{II}_{12,34}(E, E'; n), \quad (15)$$

where we have introduced the notation

$$G^{II}_{12,34}(E, E'; n)=\int \frac{d\xi}{2\pi} G^{II}_{12,34}(E, E'; \xi; n). \quad (16)$$

In (15) and (16) the lower indices 1, 2, 3, and 4 stand for the one-particle states λ_1, λ_2, λ_3, and λ_4. In what follows we call $G^{II}_{12,34}(E,E';n)$ the spectral densities of the harmonics or simply the harmonics of the two-particle function. We will also find it expedient to use the conjugation condition

$$G^{II^*}_{12,34}(E, E'; n)=-G^{II}_{34,12}(E', E; -n), \quad (17)$$

which can easily be proved via the definition (10) and the spectral representation (15). Employing the Fourier transformation

$$G^{II}(t_1, t_2; t_3, t_4)$$
$$=\int \frac{dE_1\, dE_2\, dE_3\, dE_4}{(2\pi)^4} \exp(-iE_1t_1 - iE_2t_2 + iE_3t_3 + iE_4t_4)$$
$$\times G^{II}(E_1, E_2; E_3, E_4) \quad (18)$$

and the spectral representation (15), we can establish the relation between the spectral densities of the harmonics and the Fourier transform of the two-particle function:

$$G^{II}(E_1, E_2; E_3, E_4)$$
$$=\sum_n \int \frac{d\xi}{2\pi} 2\pi\delta(E_1 - E_4 - \xi) 2\pi\delta(E_2 - E_3 + \xi - n\Omega)$$
$$\times G^{II}(E_4, E_2; \xi; n). \quad (19)$$

A similar harmonic expansion can be obtained for one-particle functions and the mass operator,

$$G(E, E')=\sum_n 2\pi\delta(E - E' - n\Omega) G(E, E'; n), \quad (20)$$

where the Fourier transform $G(E,E')$ is related to the temporal function through the transformation

$$G(E, E')=\int dt\, dt' \exp(iEt - iE't') G(t, t'). \quad (21)$$

In Keldysh's method[21] the one-particle functions and the mass operator are 2×2 matrices:

$$G=\begin{pmatrix} 0 & G^a \\ G^r & F \end{pmatrix}, \quad M=\begin{pmatrix} M^F & M^r \\ M^a & 0 \end{pmatrix},$$

where G^r is the retarded function, G^a the advanced function, and F the correlation function. The matrix elements of the mass operator are defined as

$$M_{ij}(x, x')=-ig^2 \int dx_1\, dx_2 \gamma^k_{ii'} G_{i'j'}(x, x_1)$$
$$\times \Gamma^{k'}_{j'j}(x_1, x'; x_2) \mathscr{D}_{k'k}(x_2, x), \quad (22)$$

where the $\mathscr{D}(x,x')$ are phonon functions, $\Gamma^k_{ij}(x_1,x';x_2)$ is the complete vertex matrix, g is the electron–phonon coupling constant, and γ^k_{ij} is a matrix defined as

$$\gamma_{ij}{}^1=(1/\sqrt{2})\delta_{ij},\ \gamma_{ij}{}^2=(1/\sqrt{2})(\sigma_x)_{ij} \quad (23)$$

and σ_x is the first Pauli matrix.

The following conjugacy conditions can be shown to hold true:

$$G^{r^*}_{\lambda'\lambda}(E', E; -n)=G^a_{\lambda\lambda'}(E, E'; n),$$
$$F^*_{\lambda'\lambda}(E', E; -n)=-F_{\lambda\lambda'}(E, E'; n). \quad (24)$$

$$M^{r^*}_{\lambda'\lambda}(E', E; -n)=M^a_{\lambda\lambda'}(E, E'; n),$$
$$M^{F^*}_{\lambda'\lambda}(E', E; -n)=-M^F_{\lambda\lambda'}(E, E'; n). \quad (25)$$

AN EXPRESSION FOR DC IN TERMS OF HARMONICS OF THE TWO-PARTICLE GREEN'S FUNCTION

Combining Eq. (9) with (5), (6), (10), and (17) and taking into account the spectral representation (15), we get the following expression for the DC density (valid to within terms of the order of $\mathscr{E}^2_\Omega \mathscr{E}_{2\Omega}$):

$$j_{DC}{}^\beta = j^\beta(\mathrm{I}) + j^\beta(\mathrm{II}) + j^\beta(\mathrm{III}) + j^\beta(\mathrm{IV}), \quad (26)$$

where we have introduced the notation

$$j^\beta(\mathrm{I}) = \frac{e}{4V\Omega} \sum_{\substack{1,2\\3,4}} \mathrm{Im}\left[j_{42}{}^\beta(\mathbf{v}_{31}, \vec{\mathscr{E}}_{2\Omega}) \int \frac{dE}{2\pi} G^{II}_{12,34}(E, E; +2) \right], \quad (27)$$

$$j^\beta(\mathrm{II}) = -\frac{e^3}{8Vm\Omega^2} \sum_{1,3,4} \mathrm{Re}\left[\mathscr{E}_\Omega{}^{\beta^*}(\mathbf{v}_{31}, \vec{\mathscr{E}}_{2\Omega}) \times \int \frac{dE}{2\pi} G^{II}_{14,34}(E, E-\Omega; +1) \right], \quad (28)$$

$$j^\beta(\mathrm{III}) = -\frac{e^2}{8Vm\Omega^2} \sum_{1,2,4} \mathrm{Re}\left[j_{42}{}^\beta(\vec{\mathscr{E}}_\Omega{}^*, \vec{\mathscr{E}}_{2\Omega}) \times \int \frac{dE}{2\pi} G^{II}_{12,14}(E, E; +1) \right], \quad (29)$$

$$j^{\beta}(\mathrm{IV}) = -\frac{e^4}{16Vm^2\Omega^3}\sum_{1,4}\mathrm{Im}\Big[\mathscr{E}_{\Omega}{}^{\beta *}(\vec{\mathscr{E}}_{\Omega}{}^{*},\vec{\mathscr{E}}_{2\Omega}) \times\int\frac{dE}{2\pi}G^{\mathrm{II}}_{14,14}(E,E-\Omega;0)\Big]. \tag{30}$$

Appendix A shows that the contributions of $j^{\beta}(\mathrm{II})$, $j^{\beta}(\mathrm{III})$, and $j^{\beta}(\mathrm{IV})$ can be ignored, and the problem is reduced to determining the second harmonic of the two-particle function and then using the formula (27).

AN EQUATION FOR THE TWO-PARTICLE FUNCTION

The equation for the causal two-particle functions (11) in the ladder approximation has the form

$$\begin{aligned}G_{11,1n}(x',x;x_1',x'') &= G_{11}(x',x_1')G_{1n}(x,x'')-G_{1i}(x,x_1')G_{1n}(x',x'')\\ &+ig^2\int dx_1\,dx_2 G_{1i}(x,x_1)\gamma^{k}_{ii'}G_{1i',1j'}(x',x_1;x_1',x_2)\\ &\times\gamma^{k'}_{j'j}G_{jn}(x_2,x'')\mathscr{D}_{kk'}(x_1,x_2),\end{aligned} \tag{31}$$

where $G_{ij}(x,x')$ and $\mathscr{D}_{kk'}(x,x')$ are one-particle electron and phonon functions.

Appendix B shows that the ladder approximation can be used provided that

$$\gamma/E\ll 1, \tag{32}$$

where γ is the linewidth caused by the electron–phonon interaction, and E is the characteristic electron energy.

Applying transformations (12) and (10) to both sides of Eq. (31) and passing to the limit in (10) as $t_1'\to t'$, we find that the resulting equations contain, in addition to the desired function G^{II}, two Hermitian conjugate functions φ and φ^{+}:

$$\underset{(t_3=t_1)}{\varphi(x_1,x_2;x_3,x_4)} = \underset{(t_3\to t_1+0)}{G_{21}{}^{\mathrm{II}}(x_1,x_2;x_3,x_4)} + \underset{(t_3\to t_1-0)}{G_{12}^{\mathrm{II}*}(x_3,x_4;x_1,x_2)}. \tag{33}$$

Symbolically the equations for G^{II} and φ can be written as

$$\begin{aligned}G^{\mathrm{II}} &= -G^rFG^a+\{\Phi(G^rG^{\mathrm{II}}G^a+G^r\varphi F+F\varphi^{+}G^a)\\ &+\mathscr{D}^a(FG^{\mathrm{II}}G^a+G^r\varphi^{+}G^a)+\mathscr{D}^r(G^rG^{\mathrm{II}}F+G^r\varphi G^a)\},\end{aligned} \tag{34}$$

$$\varphi = -G^rG^r+\{\Phi G^r\varphi G^r+\mathscr{D}^rG^rG^{\mathrm{II}}G^r\}. \tag{35}$$

Here the ordering of the fermion cofactors is important. The independent variables of the functions and the integral operator, denoted by braces, in the space-time representation have the form

$$\begin{aligned}\varphi(\mathbf{x}'t',\mathbf{x}t;\mathbf{x}_1't',\mathbf{x}''t'') &= -G^r(\mathbf{x}t,\mathbf{x}_1't')G^r(\mathbf{x}'t',\mathbf{x}''t'')\\ &+(ig^2/2)\int dx_1\,dx_2\Phi(x_1,x_2)G^r(\mathbf{x}t,x_1)\\ &\times\varphi(\mathbf{x}'t',x_1;\mathbf{x}_1't',x_2)G^r(x_2\mathbf{x}''t'')+\ldots\end{aligned}$$

It is more convenient to use the λ-representation, however. Performing the Fourier transformations analogous to (18) and (21) and using the spectral representations (13)–(15) and (20) to go over to the harmonics of functions, we get

$$\begin{aligned}G^{\mathrm{II}}_{12,34}(E,E';n) &= -\sum_{\substack{l,m\\(l+m=n)}}[G_{23}{}^r(E',E'-l\Omega;l)F_{14}(E+m\Omega,E;m)\\ &+F_{23}(E',E'-l\Omega;l)G_{14}{}^a(E+m\Omega,E;m)]\\ &+\frac{ig^2}{2}\sum_{\substack{1',2'\\3',4'}}\int\frac{d\mathbf{k}\,dk_0}{(2\pi)^4}J_{2'3'}(\mathbf{k})J^{*}_{4'1'}(\mathbf{k})\Phi(\mathbf{k})\\ &\times\sum_{\substack{l,m,m'\\(n=l+m+m')}}G^r_{22'}(E',E'-m\Omega;m)\\ &\times G^{\mathrm{II}}_{13',31'}(E+l\Omega-k_0,E'-m\Omega-k_0;m')G^a_{4'4}(E+l\Omega,E;l)+\ldots\end{aligned} \tag{36}$$

The other terms on the right-hand side of Eq. (36), which for the sake of brevity are denoted by dots and have been written explicitly in the symbolic equations (34) and (35), have a similar form with appropriate replacement of phonon and electron one- and two-particle functions. In Eq. (36) we have set $J_{\lambda\lambda'}(\mathbf{k}) = \langle\lambda|\exp(i\mathbf{kx})|\lambda'\rangle$, and $\mathscr{D}^r$, $\mathscr{D}^a$, and Φ are the retarded, advanced, and correlation equilibrium phonon functions, which in the homogeneous-continuum approximation have the following form:[21,24]

$$\begin{aligned}\Phi(\mathbf{k},k_0) &= -i\pi|f_k|^2(1+2N_k)\delta(k_0\mp\omega_k),\\ \mathscr{D}^r(\mathbf{k},k_0) &= (\pm^1/_2)|f_k|^2/(k_0\mp\omega_k+i\delta),\ \delta\to+0,\\ \mathscr{D}^a(\mathbf{k},k_0) &= \mathscr{D}^{r*}(k),\end{aligned} \tag{37}$$

where summation over is implied, f_k is the electron–phonon interaction matrix element, ω_k specified the dispersion law for phonons, and

$$N_k = [\exp(\omega_k/T)-1]^{-1};$$

for acoustic phonons, for instance, we have

$$|f_k|^2 = |\mathbf{k}|/u_s,\ g^2 = E_D{}^2/d,$$

where u_s, E_D, and d are the speed of sound, the deformation potential constant, and the density of the substance, respectively.

Thus, the second harmonic of the two-particle function can be found by solving the system of equations (34) and (35). The latter equation incorporates harmonics of one-particle functions, and the next section is devoted to calculating such harmonics. It must be mentioned, however, that the contribution of φ and φ^{+} to the current density can be ignored.

DETERMINING THE HARMONICS OF ONE-PARTICLE FUNCTIONS

One-particle electron functions satisfy the following equation:[21]

$$\begin{aligned}\sigma_x(i\partial/\partial t-\hat{H}_0-\hat{H}_{\Omega})G(x,x') &= \delta(x-x')-\int dx_1M(x,x_1)G(x_1,x').\end{aligned} \tag{38}$$

Applying the Fourier transformation (21) to both sides of this equation and using the spectral representations (20), we arrive at a system of coupled equations for the harmonics of one-particle functions. Using these equations, in which we

ignore terms of the third and higher orders in the pump field, we find[24] ($G = G^r$ or G^a):

$$G_{\lambda\lambda'}(E, E-\Omega; +1) = -D^*_{\lambda'\lambda} G_\lambda(E) G_{\lambda'}(E-\Omega). \tag{39}$$

$$F_{\lambda\lambda'}(E, E-\Omega; +1) = -D^*_{\lambda'\lambda}[F_\lambda(E) G_{\lambda'}{}^a(E-\Omega) + G_\lambda{}^r(E) F_{\lambda'}(E-\Omega)], \tag{40}$$

$$G_{\lambda\lambda'}(E, E-2\Omega; +2) = \sum_{\lambda_1} D^*_{\lambda'\lambda_1} D^*_{\lambda_1\lambda} G_\lambda(E) G_{\lambda_1}(E-\Omega) G_{\lambda'}(E-2\Omega), \tag{41}$$

$$\begin{aligned} F_{\lambda\lambda'}(E, E-2\Omega; +2) = \sum_{\lambda_1} D^*_{\lambda'\lambda_1} D^*_{\lambda_1\lambda} [& G_\lambda{}^r(E) F_{\lambda_1}(E-\Omega) G_{\lambda'}{}^a(E-2\Omega) \\ & + G_\lambda{}^r(E) G_{\lambda_1}{}^r(E-\Omega) F_{\lambda'}(E-2\Omega) \\ & + F_\lambda(E) G_{\lambda_1}{}^a(E-\Omega) G_{\lambda'}{}^a(E-2\Omega)]. \end{aligned} \tag{42}$$

Here $D_{\lambda\lambda'} = (\omega_{\lambda\lambda'}/2\Omega)(e\mathbf{x}_{\lambda\lambda'}, \vec{\mathscr{E}}_\Omega)$,

$$\omega_{\lambda\lambda'} = (E_\lambda - E_{\lambda'}), \quad \mathbf{x}_{\lambda\lambda'} = \langle\lambda|\mathbf{x}|\lambda'\rangle,$$

$$G_\lambda{}^r(E) = (E - E_\lambda + i\gamma_\lambda)^{-1}, \tag{43}$$

$$\begin{aligned} F_\lambda(E) &= (1-2n_E)[G_\lambda{}^r(E) - G_\lambda{}^a(E)] \\ &= -G_\lambda{}^r(E) M_\lambda{}^F(E) G_\lambda{}^a(E), \end{aligned} \tag{44}$$

$n_E = \{\exp[(E - E_F)/T] + 1\}^{-1}$, T is the temperature in energy units, E_F is the Fermi level, and $\gamma_\lambda = \mathrm{Im}[M_\lambda^r(E_\lambda)]$ is the width of the energy level E_λ, the eigenvalue of the Hamiltonian H_0 renormalized with allowance for the electron–phonon interaction.

In the derivation of Eqs. (39)–(42) all terms containing the first and second harmonics of the mass operator were omitted, which can be done (see Appendix C) provided that

$$\gamma_\lambda/\Omega \ll 1. \tag{45}$$

Obviously, this condition is met if the electron–phonon interaction is fairly weak.

CALCULATING THE DC DENSITY: CONTRIBUTION OF PRODUCTS OF HARMONICS OF ONE-PARTICLE FUNCTIONS

For convenience we write (27) as

$$j_{DC}{}^\beta = (e/4V\Omega)\,\mathrm{Im}(I^\beta), \tag{46}$$

where

$$I^\beta = \sum_{\substack{1,2\\3,4}} j_{42}{}^\beta(\mathbf{v}_{31}, \vec{\mathscr{E}}_{2\Omega}) \int \frac{dE}{2\pi} G^{\mathrm{II}}_{12,34}(E, E; +2). \tag{47}$$

We start by calculating the contribution to the current from the abstract terms of Eq. (34):

$$\begin{aligned} G^{\mathrm{II}}_{12,34}(E, E; +2) = & -G_{23}{}^r(E, E-2\Omega; +2) F_{14}(E, E; 0) \\ & - G_{23}{}^r(E, E; 0) F_{14}(E+2\Omega, E; +2) \\ & - F_{23}(E, E; 0) G_{14}{}^a(E+2\Omega, E; +2) \\ & - G_{23}{}^r(E, E-\Omega; +1) F_{14}(E+\Omega, E; +1) \\ & - F_{23}(E, E-\Omega; +1) G_{14}{}^a(E+\Omega, E; +1). \end{aligned} \tag{48}$$

Now we can use (39)–(44). When calculating the integral with respect to E, we assume, in accordance with (47), that the following conditions hold true:

$$\gamma_\lambda/E_\lambda \ll 1 \text{ and } \gamma_\lambda/T \ll 1, \tag{49}$$

which makes it possible to ignore the contributions of the poles of $(1 - 2n_E)$. We find

$$\begin{aligned} I^\beta = & -i \sum_{1-4} j_{42}{}^\beta(\mathbf{v}_{34}, \vec{\mathscr{E}}_{2\Omega}) D_{31}{}^* D_{12}{}^* \frac{2}{(\omega_{42} + i\Gamma_{42})} \\ & \times \Big[\frac{n_4 - n_3}{(\omega_{41} - \Omega + i\Gamma_{41})(\omega_{43} - 2\Omega + i\Gamma_{43})} \\ & + \frac{n_1 - n_2}{(\omega_{21} - \Omega - i\Gamma_{21})(\omega_{23} - 2\Omega - i\Gamma_{23})} \\ & + \frac{n_3 - n_1}{(\omega_{13} - \Omega - i\Gamma_{13})(\omega_{41} - \Omega + i\Gamma_{41})} \\ & - \frac{n_3 - n_1}{(\omega_{13} - \Omega - i\Gamma_{13})(\omega_{23} - 2\Omega - i\Gamma_{23})} \Big] \\ & - i \sum_{1-4} j_{42}{}^\beta(\mathbf{v}_{21}, \vec{\mathscr{E}}_{2\Omega}) D_{43}{}^* D_{31}{}^* \frac{2}{(\omega_{42} + i\Gamma_{42})} \\ & \times \Big[\frac{n_4 - n_3}{(\omega_{41} + 2\Omega + i\Gamma_{41})(\omega_{43} + \Omega + i\Gamma_{43})} \\ & + \frac{n_1 - n_2}{(\omega_{23} + \Omega - i\Gamma_{23})(\omega_{21} + 2\Omega - i\Gamma_{21})} \\ & + \frac{n_3 - n_1}{(\omega_{13} - \Omega - i\Gamma_{13})(\omega_{41} + 2\Omega + i\Gamma_{41})} \\ & - \frac{n_3 - n_1}{(\omega_{13} - \Omega - i\Gamma_{13})(\omega_{23} + \Omega - i\Gamma_{23})} \Big] \\ & - i \sum_{1-4} j_{42}{}^\beta(\mathbf{v}_{31}, \vec{\mathscr{E}}_{2\Omega}) D_{32}{}^* D_{41}{}^* \frac{2}{(\omega_{42} + i\Gamma_{42})} \\ & \times \Big[\frac{n_4 - n_1}{(\omega_{43} - \Omega + i\Gamma_{43})(\omega_{41} + \Omega + i\Gamma_{41})} \\ & + \frac{n_3 - n_2}{(\omega_{21} + \Omega - i\Gamma_{21})(\omega_{23} - \Omega - i\Gamma_{23})} \\ & + \frac{n_3 - n_1}{(\omega_{31} + 2\Omega - i\Gamma_{31})} \Big(\frac{1}{(\omega_{34} + \Omega - i\Gamma_{34})} + \frac{1}{(\omega_{12} + \Omega - i\Gamma_{21})} \Big) \Big], \end{aligned} \tag{50}$$

where we have introduced the notation $\Gamma_{\lambda\lambda'} = \gamma_\lambda + \gamma_{\lambda'}$.

Up to this point no assumptions concerning the properties of the electronic states were made, with the result that (46) and (50) can be applied to any model of the coherent photovoltaic effect. But we are interested in the description of this effect for impurity–band transitions. Hence, we will assume that the indices 1, 2, 3, and 4 in (50), which denote one-electron states, can refer either to the conduction band or to an impurity state (excitation of holes in quartz glass is of less interest because of the low hole mobility).

Thus, in summing over the four indices in (50) we encounter sixteen types of terms corresponding to the distribution of two types of states (impurity "*i*" and band "**p**" states)

over four indices. To simplify matters, we assume that the intraband matrix elements are diagonal in momenta:

$$j^{\beta}_{\mathbf{pp}'}=\langle \mathbf{p}|\hat{j}^{\beta}|\mathbf{p}'\rangle=ep^{\beta}\Delta(\mathbf{p}-\mathbf{p}')/m. \tag{51}$$

$$v^{\alpha}_{\mathbf{pp}'}=\langle \mathbf{p}|\hat{v}^{\alpha}|\mathbf{p}'\rangle=p^{\alpha}\Delta(\mathbf{p}-\mathbf{p}')/m, \tag{52}$$

$$D_{\mathbf{pp}'}=(\omega_{\lambda\lambda'}/2\Omega)(e\mathbf{x}_{\lambda\lambda'}\vec{\mathscr{E}}_{\Omega})=\frac{e}{2im\Omega}(\mathbf{p}\vec{\mathscr{E}}_{\Omega})\Delta(\mathbf{p}-\mathbf{p}'), \tag{53}$$

where $\mathbf{x}_{\lambda\lambda'}=\langle\lambda|\mathbf{x}\lambda'\rangle$, and

$$\Delta(\mathbf{p}-\mathbf{p}')=\begin{cases}1, & \mathbf{p}=\mathbf{p}'\\ 0, & \mathbf{p}\neq\mathbf{p}'\end{cases}.$$

Formulas (51)–(53) have been obtained for the simple case where the delocalized states are described by plane waves. We also assume that

$$j^{\beta}_{ii'}=0, \quad v^{\alpha}_{ii'}=0, \quad D_{ii'}=0 \text{ for all } i \text{ and } i', \tag{54}$$

which simply reflects the fact that state $\langle i|$ is localized and that the concentration of such states is so low that the overlap of the wave functions belonging to different centers can be ignored. Conditions (54), obviously, also imply that the centers cannot have excited localized states.

Allowing for (54), we note that Eq. (50) can have only the following nonzero combinations of four indices: four combinations in which one index corresponds to a localized state and the others to delocalized states, and combinations in which two indices correspond to localized states and the other two to delocalized states, with the respective terms in (50) containing the products of four matrix elements of impurity–band transitions. Appendix D shows that the contribution of the latter terms to the current can be ignored if conditions (45) are met.

If conditions (51)–(53) are met, among the four combinations of indices in which one index corresponds to a localized state we first isolate the combinations in which either index 1 or index 3 corresponds to a localized state. In this case conditions (51)–(53) ensure that the other three indices coincide. Such combinations of indices have the characteristic feature that each term in (50) acquires a factor γ_p^{-1}, guaranteeing that the terms under consideration dominate all other terms if condition (45) is met (see Appendix D).

Thus, in (50) we put either $\lambda_1=i$ and $\lambda_2=\lambda_3=\lambda_4=\mathbf{p}$, or $\lambda_3=i$ and $\lambda_1=\lambda_2=\lambda_4=\mathbf{p}$. We also allow for the fact that summation over localized states in the case considered here can be replaced with multiplication by NV, where N is the concentration of localized states (i.e., the concentration of the respective defects), and V is the system volume. As a result, combining (46) and (51)–(53) with (50) yields

$$\begin{aligned}j_{DC}^{\beta}=&\frac{e^2\pi N}{4(\hbar\Omega)^2m^2}\sum_{\mathbf{p}}p^{\beta}\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_{2\Omega})D_{i\mathbf{p}}D_{\mathbf{p}i}^{*}]\\ &\times\gamma_p^{-1}(n_i-n_p)\delta(E_p-E_i-\hbar\Omega)\\ &+\frac{e^2\pi N}{4(\hbar\Omega)^2m^2}\sum_{\mathbf{p}}p^{\beta}\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_{\Omega}^{*})D_{i\mathbf{p}}^{*}(\omega_{i\mathbf{p}}/\Omega)\\ &\times(e\mathbf{x}_{i\mathbf{p}},\vec{\mathscr{E}}_{2\Omega})]\gamma_p^{-1}(n_i-n_p)\\ &\times[\delta(E_p-E_i-\hbar\Omega)-\delta(E_p-E_i-2\hbar\Omega)],\end{aligned} \tag{55}$$

where the delta function appears as a result of the passage to the limit

$$(\Gamma/\pi)/(x^2+\Gamma^2)\to\delta(x), \tag{56}$$

which is justified due to the first condition in (49).

Thus, formula (55) gives the contribution to the DC density from the abstract terms in Eq. (34) for the second harmonic of the two-particle function G^{II}.

CALCULATING THE DC DENSITY: CONTRIBUTION OF INTEGRAL TERMS

Appendix E shows that if condition (45) is met, of all the integral terms in Eq. (34) only the integral term containing the second harmonic of function G^{II} provides a contribution to the DC density comparable, in order of magnitude, with (55). In accordance with (46) and (47), let us introduce a new function

$$g_{24}^{\alpha}(E)=\sum_{1,3}v_{31}^{\alpha}G^{\mathrm{II}}_{12,34}(E,E;+2), \tag{57}$$

in terms of which the DC density will be calculated. An equation for this function can easily be derived from (36) (to within terms of order $\mathscr{E}_{\Omega}\mathscr{E}_{\Omega}\mathscr{E}_{2\Omega}$):

$$\begin{aligned}g_{24}^{\alpha}(E)=&C_{24}^{\alpha}(E)\\ &+\frac{i}{2}g^2\sum_{1',3'}\int\frac{d\mathbf{k}\,dk_0}{(2\pi)^4}J_{23'}(\mathbf{k})J_{41'}^{*}(\mathbf{k})g^{\alpha}_{3'1'}(E-k_0)\\ &\times\{\Phi(k)G_2^{r}(E)G_4^{a}(E)+\mathscr{D}^{a}(k)F_2(E)G_4^{a}(E)\\ &+\mathscr{D}^{r}(k)G_2^{r}(E)F_4(E)\},\end{aligned} \tag{58}$$

where

$$C_{24}^{\alpha}(E)=\sum_{1,3}v_{31}^{\alpha}A_{12,34}(E), \tag{59}$$

and the $A_{12,34}(E)$ stand for the abstract terms in (36), whose form is specified by the right-hand side of Eq. (48) with allowance for (39)–(44).

In the previous section we saw that if condition (45) is met, the main contribution to the current density, specified by formula (55), is provided by the abstract term of Eq. (53), $C^{\alpha}_{24}(E)$, in which the indices "2" and "4" coincide and belong to the set of delocalized states. If we allow for the general form of the solution of the inhomogeneous Fredholm integral equation of the second kind (see, e.g., Ref. 25), this suggests that [provided that condition (45) is met] the contribution to the current from the solution of Eq. (58) with identical indices "2" and "4" belonging to delocalized states, $g^{\alpha}_{22}(E)=g^{\alpha}_{\mathbf{p}}(E)$, considerably exceeds the contribution to the current from the functions $g^{\alpha}_{24}(E)$ with distinct indices.

Assuming that the electron–phonon interaction does not take charge carriers outside the conduction band and

$$J_{\mathbf{pp}'}(\mathbf{k})=\Delta(\mathbf{p}-\mathbf{k}-\mathbf{p}'), \tag{60}$$

which is true if the wave functions of the localized states are plane waves, we find that Eq. (58) leads to the following equation for $g^{\alpha}_{\mathbf{p}}(E)$:

$$g_{\mathbf{p}}^{\alpha}(E)=C_{\mathbf{p}}^{\alpha}(E)+\frac{i}{2}g^2\int\frac{d\mathbf{k}\,dk_0}{(2\pi)^4}g_{\mathbf{p}-\mathbf{k}}^{\alpha}(E-k_0)$$
$$\times\{\Phi(k)G_{\mathbf{p}}^{r}(E)G_{\mathbf{p}}^{a}(E)+\mathscr{D}^{a}(k)F_{\mathbf{p}}(E)G_{\mathbf{p}}^{a}(E)$$
$$+\mathscr{D}^{r}(k)G_{\mathbf{p}}^{r}(E)F_{\mathbf{p}}(E)\}. \tag{61}$$

The integral operator in this equation can be simplified by employing (37) and (44). As a result we get

$$g_{\mathbf{p}}^{\alpha}(E)=G_{\mathbf{p}}^{\alpha}(E)$$
$$+\frac{g^2}{4}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2[(1+2N_k)\mp(1-2n_p)]$$
$$\times G_{\mathbf{p}}^{r}(E)G_{\mathbf{p}}^{a}(E)g_{\mathbf{p}-\mathbf{k}}^{\alpha}(E\mp\omega_k), \tag{62}$$

where summation over double signs is implied.

Now, by taking into account the definition (59), the explicit form of the function $A_{12,34}(E)$ [the right-hand side of Eq. (48)], and Eqs. (39)–(44), we easily conclude that $G_{\mathbf{p}}^{\alpha}(E)$, as well as the integral operator in Eq. (62), contain a factor $G_{\mathbf{p}}^{r}(E)G_{\mathbf{p}}^{\alpha}(E)$, which becomes $\delta(E-E_p)$ as $\gamma_p\to 0$. Thus, the desired function $g_{\mathbf{p}}^{\alpha}(E)$ proves to be proportional to $\delta(E-E_p)$:

$$g_{\mathbf{p}}^{\alpha}(E)=g^{\alpha}(\mathbf{p})2\pi\delta(E-E_p). \tag{63}$$

If we integrate both sides of Eq. (62) with respect to E, we get

$$g^{\alpha}(\mathbf{p})=C^{\alpha}(\mathbf{p})+\frac{\gamma_p^{-1}\pi g^2}{4}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2[(1+2N_k)\mp(1-2n_p)]$$
$$\times g^{\alpha}(\mathbf{p}-\mathbf{k})\delta(E_{\mathbf{p}}-E_{\mathbf{p}-\mathbf{k}}\mp\omega_k). \tag{64}$$

Noting that the function

$$C^{\alpha}(\mathbf{p})=\int dE C_{\mathbf{p}}^{\alpha}(E)/2\pi$$

is actually present when we obtain formula (50) and the expression (55) for the current density, we find

$$C^{\alpha}(\mathbf{p})=p^{\alpha}f_1(\mathbf{p})/\gamma_p+mv_{ip}{}^{\alpha}f_2(\mathbf{p})/\gamma_p, \tag{65}$$

where

$$f_1(\mathbf{p})=\frac{\pi iVN}{m\Omega}D_{ip}{}^{*}D_{pi}{}^{*}(n_i-n_p)\delta(\omega_{pi}-\Omega), \tag{66}$$

$$f_2(\mathbf{p})=\frac{\pi iVN}{m\Omega}D_{ip}{}^{*}D_{pi}{}^{*}(n_i-n_p)[\delta(\omega_{pi}-2\Omega)-\delta(\omega_{pi}-\Omega)]. \tag{67}$$

Here we have omitted, as we did in deriving (55), the terms containing $\delta(\omega_{pi}+\Omega)$ and $\delta(\omega_{pi}+2\Omega)$, since in the case under investigation E_p is higher than E_i and, in addition, summation over localized states has been replaced with multiplication by NV.

For quasielastic scattering, the solution to Eq. (64), where the free term is taken to be the first term on the right-hand side of (65), has the form

$$g^{\alpha}(\mathbf{p})=p^{\alpha}f_1(\mathbf{p})/\Gamma_1(\mathbf{p}), \tag{68}$$

with $\Gamma_1(\mathbf{p})$ independent of the direction of vector $\mathbf{p}$:

$$\Gamma_1(\mathbf{p})$$
$$=\gamma_p-\frac{\pi g^2}{2}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2(1+2N_k)\delta(E_{\mathbf{p}}-E_{\mathbf{p}-\mathbf{k}})$$
$$\times\frac{(p^{\alpha}-k^{\alpha})}{p^{\alpha}}f_1(\mathbf{p}-\mathbf{k})/f_1(\mathbf{p}). \tag{69}$$

This equation can be simplified if we take into account the explicit form of the phonon level width $\gamma_\lambda=\mathrm{Im}[M_{\lambda\lambda}^{r}(E_\lambda,E_{\lambda;0})]$. Using the general expression (22) for the mass operator and taking the complete vertex matrix in the zeroth approximation in the electron–phonon coupling constant,

$$\Gamma_{j'j}^{kk'}(x_1,x';x_2)=\gamma_{jj'}^{kk'}\delta(x_1-x')\delta(x_2-x'),$$

we get[24]

$$\gamma_\lambda=\frac{\pi g^2}{4}\sum_{\lambda'}\int\frac{d\mathbf{k}}{(2\pi)^3}|J_{\lambda\lambda'}(\mathbf{k})|^2|f_k|^2[(1+2N_k)\pm(1-2n_{\lambda'})]$$
$$\times\delta(E_\lambda-E_{\lambda'}\mp\omega_k).$$

For the quasielastic approximation and delocalized λ and λ' states this yields

$$\gamma_p=\frac{\pi g^2}{2}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2(1+2N_k)\delta(E_{\mathbf{p}}-E_{\mathbf{p}-\mathbf{k}}), \tag{70}$$

which can easily be used to transform (70) to

$$\Gamma_1(\mathbf{p})=\frac{\pi g^2}{2}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2(1+2N_k)\delta(E_{\mathbf{p}}-E_{\mathbf{p}-\mathbf{k}})$$
$$\times\left[1-\frac{(p^{\alpha}-k^{\alpha})}{p^{\alpha}}\frac{f_1(\mathbf{p}-\mathbf{k})}{f_1(\mathbf{p})}\right]. \tag{71}$$

Function $\Gamma_1(\mathbf{p})$ is independent of the direction of momentum $\mathbf{p}$, say, in the event of depolarized pumping. This leads to an average of $f_1(\mathbf{p})$ over the various directions of pump radiation the polarizations, as a result of which this function ceases to depend on the direction of vector $\mathbf{p}$ and in (71) the factor $f_1(\mathbf{p}-\mathbf{k})/f_1(\mathbf{p})$ becomes equal to unity. Clearly, the fraction $(p^{\alpha}-k^{\alpha})/p^{\alpha}$ in the integrand in (71) coincides with the cosine of the angle θ between vectors $\mathbf{p}$ and $\mathbf{p}-\mathbf{k}$, and Eq. (71) assumes the form

$$\Gamma_1(p)=\frac{\pi g^2}{2}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2(1+2N_k)[1-\cos(\theta)]\delta(E_{\mathbf{p}}-E_{\mathbf{p}-\mathbf{k}}). \tag{72}$$

This quantity is well-known from solid-state physics, the inverse transport relaxation time (see, e.g., Refs. 26 and 27) in quasielastic scattering on acoustic phonons.

In the case of polarized radiation, to find the inverse relaxation time we must use the general expression (71) containing the fraction $f_1(\mathbf{p}-\mathbf{k})/f_1(\mathbf{p})$, which it is advisable to rewrite in a form containing the polarization of the pump radiation explicitly. For this, as Eq. (66) shows, we must calculate the product of matrix elements, $D_{ip}^{*}D_{pi}^{*}$, expressed in terms of the dipole-transition matrix elements $\mathbf{x}_{ip}$. Obviously, if the wave function of a localized state $|i\rangle$ is isotropic (an s state), the only preferred direction in the expression for $\mathbf{x}_{ip}$ is that of the momentum $\mathbf{p}$, and we arrive at the following expression for the plane wave describing a delocalized state:

$$\mathbf{x}_{ip}=b_p\mathbf{p}\exp[i(\mathbf{p},\mathbf{R}_i)], \tag{73}$$

where $\mathbf{R}_i$ is the radius vector of a localization center, and the scalar b_p is independent of the direction of the vector $\mathbf{p}$. Here also

$$D_{ip}^{*}D_{pi}^{*}=-e^2(\omega_{pi}/2\Omega)^2|b_p|^2(\mathbf{p},\vec{\mathscr{E}}_\Omega^{*})^2, \tag{74}$$

$$f_i(\mathbf{p}-\mathbf{k})/f_i(\mathbf{p})=(\mathbf{p}-\mathbf{k},\vec{\mathscr{E}}_\Omega^{*})^2/(\mathbf{p},\vec{\mathscr{E}}_\Omega^{*})^2. \tag{75}$$

In the case of directed orbitals of localized states, the vectors $\mathbf{x}_{ip}$ and $\mathbf{p}$ may not be collinear, but in obtaining expression (66) an average over the various directions occurs for random orientations of different defects, and we can again use the formula (73).

Similarly, we solve Eq. (64), where the second term on the right-hand side of (65) serves as the abstract term. As in (68), the solution has the form

$$g^\alpha(\mathbf{p})=mv_{ip}^{\alpha}f_2(\mathbf{p})/\Gamma_2(\mathbf{p}), \tag{76}$$

where

$$\Gamma_2(\mathbf{p})=\frac{\pi g^2}{2}\int\frac{d\mathbf{k}}{(2\pi)^3}|f_k|^2(1+2N_k)\delta(E_p-E_{\mathbf{p}-\mathbf{k}}) \times[1-(v_{ip-k}^{\alpha}/v_{ip}^{\alpha})f_2(\mathbf{p}-\mathbf{k})/f_2(\mathbf{p})]. \tag{77}$$

From $v_{ip}^{\alpha}=i\omega_{ip}x_{ip}^{\alpha}$, if condition (73) is met we have

$$mv_{ip}^{\alpha}D_{ip}^{*}D_{pi}^{*}=-e^2p^{\alpha}(\omega_{pi}/2\Omega)^2|b_p|^2(\mathbf{p},\vec{\mathscr{E}}_\Omega^{*})^2. \tag{78}$$

Here, obviously, $\Gamma_1(\mathbf{p})=\Gamma_2(\mathbf{p})=\Gamma(\mathbf{p})$.

Thus, the solution to the linear integral equation (64) in the quasielastic approximation has the form

$$g^\alpha(\mathbf{p})=p^{\alpha}f_1(\mathbf{p})/\Gamma_1(\mathbf{p})+mc_{ip}^{\alpha}f_2(\mathbf{p})/\Gamma_2(\mathbf{p}), \tag{79}$$

where $f_1(\mathbf{p})$ and $f_2(\mathbf{p})$ have been defined by (66) and (67), respectively. Taking into account the definition (57) and Eqs. (46) and (47), we conclude that an expression for the DC density allowing for the integral operator in Eq. (58) is actually Eq. (55) with γ_p^{-1} replaced with $1/\Gamma_1(\mathbf{p})$ in the first term and with $1/\Gamma_2(\mathbf{p})$ in the second:

$$j_{DC}^{\beta}=(e^2\pi N/4(\hbar\Omega)^2m^2)\sum_{\mathbf{p}}p^{\beta}\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_{2\Omega})D_{ip}^{*}D_{pi}^{*}] \times(n_i-n_p)/\Gamma_1(p) \times\delta(E_p-E_i-\hbar\Omega)+(e^2\pi N/4(\hbar\Omega)^2m^2) \times\sum_{\mathbf{p}}p^{\beta}\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_\Omega^{*})D_{ip}^{*}(\omega_{ip}/\Omega)\cdot \times(e\mathbf{x}_{ip},\vec{\mathscr{E}}_{2\Omega})](n_i-n_p)/\Gamma_2(p) \times[\delta(E_p-E_i-\hbar\Omega)-\delta(E_p-E_i-2\hbar\Omega)]. \tag{80}$$

If condition (73) is met, we have

$$(\mathbf{p},\vec{\mathscr{E}}_\Omega^{*})D_{ip}^{*}(\omega_{ip}/\Omega)(e\mathbf{x}_{ip},\vec{\mathscr{E}}_{2\Omega})=-2D_{ip}^{*}D_{pi}^{*}(\mathbf{p},\vec{\mathscr{E}}_{2\Omega}), \tag{81}$$

owing to which formula (80) is transformed into

$$j_{DC}^{\beta}=(e^2\pi N/4(\hbar\Omega)^2m^2) \times\sum_{\mathbf{p}}p^{\beta}\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_{2\Omega})D_{ip}^{*}D_{pi}^{*}](n_i-n_p)/\Gamma(p) \times[2\delta(E_p-E_i-2\hbar\Omega)-\delta(E_p-E_i-\hbar\Omega)]. \tag{82}$$

PHYSICAL INTERPRETATION OF THE RESULTS

Let us now interpret the results physically. Each of the terms in (80) can easily be associated with the common expression for a drift current-density component which, however, contains a nonequilibrium distribution function for the electrons excited by light from localized states to the conduction band. This leads to the following interpretation of the phenomenon.

The probability $P(t)$ of the transition between two states, $n\to m$, stimulated by a biharmonic perturbation

$$\hat{V}(t)=\hat{h}\exp(-i\Omega t)+\hat{h}^{+}\exp(i\Omega t) +\hat{H}\exp(-i2\Omega t)+\hat{H}^{+}\exp(i2\Omega t), \tag{83}$$

where $\hat{h}$ and $\hat{H}$ are operators representing the interaction of an electron with the pump field of frequency Ω and with the second-harmonic field of frequency 2Ω, respectively, contains not only a time-independent term but also terms that vary with time harmonically with frequencies Ω and 2Ω:

$$P(t)\propto\frac{h_{mn}h_{mn}^{*}}{(\omega_{mn}\mp\Omega)^2+\delta^2}+\frac{h_{mn}h_{nm}}{(\omega_{mn}\pm\Omega-i\delta)(\omega_{mn}\mp\Omega+i\delta)} \times\exp(\pm i2\Omega t) +\left[\frac{h_{mn}^{*}H_{mn}}{(\omega_{mn}\mp\Omega-i\delta)(\omega_{mn}\mp2\Omega+i\delta)} +\frac{H_{nm}^{*}h_{nm}}{(\omega_{mn}\pm\Omega+i\delta)(\omega_{mn}\pm2\Omega-i\delta)}\right]\exp(\pm i\Omega t). \tag{84}$$

Here summation over repeated indices and double signs is implied and terms proportional to H^2 and to $\exp(\pm i3\Omega t)$ are discarded. We assume that the perturbation (83) increases adiabatically (δ is the adiabaticity parameter) starting at time $t=-\infty$, when the system was in state n. Formula (84) shows that the distribution function for electrons in the conduction band must contain time-dependent terms:

$$\Delta n_p(t)=\Delta n_1(p)\cos(\Omega t)+\Delta n_2(p)\cos(2\Omega t+\varphi). \tag{85}$$

As formula (84) shows, the first term in (85) appears because of the photoionization of defects caused by the interference of the second-harmonic radiation and the pumping radiation. The second term in (85) emerges because of the photoionization of electrons solely by the pumping radiation. The time-constant current can be obtained from the general expression

$$\mathbf{j}=en(t)\mu\vec{\mathscr{E}}(t), \tag{86}$$

where μ is the electron mobility, in two cases. First, when the second-harmonic electric field is the attractive field $\vec{\mathscr{E}}$ in (86) and the density $n(t)$ is determined by the second term on the right-hand side of (85), and, second, when the electric field in the pump wave is the attractive field and the

density is determined by the first term on the right-hand side of (85). Thus, we arrive at (80), where the first term describes the drift of nonequilibrium electrons, photoionized by the pump wave, in the electric field of the second-harmonic wave, while the second describes the drift of nonequilibrium electrons, photoionized owing to the interference of the pump wave and the second-harmonic wave, in the electric field of the pump wave. From Eq. (84) we see that the second mechanism operates when the energy difference between the electron levels amounts to either $\hbar\Omega$ or $2\hbar\Omega$. This agrees with the form of the second term on the right-hand side of (80).

Electron excitation caused by the interference of the pump wave and the second-harmonic wave and accompanied by absorption of an amount of energy equal to $2\hbar\Omega$ was discussed in Refs. 9–12, 16, 18, and 20. The first term on the right-hand side of (80) was obtained in Refs. 2, 28, and 29. The expression for the current density in a coherent photovoltaic effect obtained in Ref. 6 also contains terms describing processes with energy absorption amounting to either $\hbar\Omega$ or $2\hbar\Omega$ (apparently, because of a misprint the process thresholds listed in Ref. 6 are incorrect).

All the above mechanisms of the coherent photovoltaic effect, obviously, may yield contributions to DC similar in order of magnitude, differing, perhaps, only in the probabilities of the corresponding impurity-band dipole transitions.

ESTIMATING THE CURRENT DENSITY IN THE COHERENT PHOTOVOLTAIC EFFECT

To be definite, let us estimate the size of the first term in the expression (80) for the current density. Since the coherent photovoltaic effect is used to explain second-harmonic generation in quartz-glass based optical fiber guides, we will carry out this estimate for SiO_2, whose conduction band can be assumed to be isotropic and parabolic (see, e.g., the calculations done in Ref. 30) and the effective mass to be close to the mass of a free electron, $m^* \approx m$ (see Ref. 31). For simplicity we take the wave function of the delocalized state with momentum p in the form of a plane wave,

$$\psi_p(\mathbf{x}) = \exp[i(\mathbf{p}, \mathbf{x})/\hbar] V^{-1/2}, \tag{87}$$

and use a hydrogenlike model with an s-type wave function for the localized state,

$$\psi_i(\mathbf{x}) = \frac{1}{(\pi\rho^3)^{1/2}} \exp\left(-\frac{x}{\rho}\right), \tag{88}$$

with a localization radius ρ whose magnitude can be estimated by $\rho = \hbar(2mI)^{-1/2}$, where $I = \hbar\Omega - E_p$ is the ionization potential of the defect.

We now use the wave functions (87) and (88) to calculate the dipole-transition matrix element $\mathbf{x}_{ip}$:

$$(\mathbf{x}_{ip}, \vec{\mathscr{E}}_\Omega) = 4\pi i (\mathbf{p}, \vec{\mathscr{E}}_\Omega)(\hbar^4/p^5)\eta/(\pi\rho^3 V)^{1/2}, \tag{89}$$

where $\eta = 8s^5(1+s^2)^{-3}$, with $s = \rho p/\hbar$.

Taking into account the well-known relation $\mu = e\tau_r/m$ existing between the transport relaxation time τ_r for electrons and their mobility μ in the conduction band ($\mu \approx 20$ $cm^2 V^{-1} s^{-1}$ for SiO_2; see Ref. 32), we can easily calculate the first term on the right-hand side of (80). As a result, for $\mathscr{E}_\Omega \approx 3\times 10^8$ Vm^{-1} and $\mathscr{E}_{2\Omega} \approx 3\times 10^7$ Vm^{-1} (such fields correspond to the power of laser radiation used in experiments in second-harmonic generation in light guides) at $\lambda = 1064$ nm, $E_p \approx 0.1$ eV, and $N \approx 2\times 10^{11}$ cm^{-3} we get $\rho \approx 0.19$ nm and $j_{DC} \approx 10^{-6}$ A cm^{-2}. This value of the current density is sufficient to explain, in accordance with the model of Ref. 3, the appearance of a strong electrostatic field $E_{DC} \approx 10^4$–10^5 V cm^{-1} in the fiber core in the process of preparing the guide for second-harmonic generation. Such an estimate was first done in Ref. 33 but can probably be better substantiated along different lines.

For instance, in planar geometry the equation for the total current in the direction transverse to the guide's axis (along the x axis) is

$$j_{dr}^x + j_{dif}^x + j_{DC}^x + \varepsilon\varepsilon_0 \partial E^x/\partial t = 0, \tag{90}$$

where j_{dr}^x and j_{dif}^x are the drift and diffusion currents, respectively, and ε is the low-frequency dielectric constant ($\varepsilon = 3.8$ for SiO_2). Obviously, only the current j_{DC}^x of the coherent photovoltaic creates an asymmetric charge distribution and, hence, a corresponding nonzero average electrostatic field E_{DC}. To estimate the order of magnitude of this field it is thus sufficient to put $j_{DC}^x \approx \varepsilon\varepsilon_0 E_{DC}/t_w$, where t_w is the time required to record the nonlinear susceptibility in the process of preparing the guide for second-harmonic generation, that is, the average time during which the pump field and the initial second-harmonic field are nonzero in the process of preparing the guide. Following Ref. 33, we put $t_w \approx 10^{-3}$ s, and for $j_{DC} \sim 10^{-6}$ A cm^{-2} we obtain $E_{DC} \approx t_w j_{DC}^x (\varepsilon\varepsilon_0)^{-1} \approx 10^4$ V cm^{-1}. An electrostatic field of this strength ($E_{DC} \geqslant 10^4$ V cm^{-1}) was observed in the experiment reported in Ref. 34.

Within the same approximation scheme and for the same values of the above parameters, the absorption coefficient α of the pumping radiation in defect–band transitions proves to be approximately equal to $N\rho^2 \approx 10^{-4}$ $cm^{-1} \approx 10^2$ dB km^{-1}. We see that the absorption of the pumping radiation over distances of several centimeters is extremely low (it is over lengths like this that second-order nonlinear susceptibility is recorded in optical fiber guides[2]). Thus, at least in essence the model is noncontradictory: the processes leading to optical generation of an asymmetric current of the necessary magnitude are not accompanied by such a strong absorption of the pumping radiation that no radiation travels along the guide. This consistency feature is retained up to defect densities N of approximately 10^{14} cm^{-3}; that is, it has a "safety factor" of about a thousand.

On the one hand, the above defect density $N \approx 2\times 10^{11}$ cm^{-3} at which a current density necessary for explaining second-harmonic generation appears is extremely low. On the other, the majority of defects in SiO_2 have filled levels deep in the forbidden bands.[35–37] Hence, the localized electrons on energy levels close to the conduction band (at a distance of 1–2 eV from its bottom) may appear in quartz glass only due to electron excitation (because of multiphoton absorption) from deep-lying localized states. Such impurity complexes are suggested in Refs. 2, 28, and 29, say, a pair of germanium atoms and a neighboring phosphorous atom at the silicon sites of quartz glass. Not only phosphorous atoms but also triply coordinated silicon atoms can act as optical donors.

CONCLUSION

We have derived an expression for the density of DC generated by coherent pumping radiation and second-harmonic radiation and found two mechanisms of current generation: the first due to the absorption of pumping radiation and the drift of photoelectrons in the field of the second-harmonic wave (one photon from the pump field is absorbed), and the second due to the drift of photoelectrons in the field created by the interference of the pump field and the second-harmonic field (either one photon from the pump field is absorbed or two). The latter differs from the mechanism suggested by Zel'dovich *et al.*,[9-12] although it also belongs to photoionization by the pumping and second-harmonic radiations.

The expressions obtained in this paper make it possible to estimate the photocurrent and show that the coherent photovoltaic effect may quite reasonably be considered responsible for second-harmonic generation in glass optical fiber guides.

The author is grateful to all participants of the seminars arranged by the Fiber Optics Department and the Vibrations Department and of the theoretical seminar held at the Institute of General Physics of the Russian Academy of Sciences, the staff of the Semiconductor Theory Department at Moscow State University, and B. A. Veklenko, E. M. Dianov, V. P. Makarov, and V. O. Sokolov for critical remarks and fruitful discussions.

APPENDIX A

Let us show that the current-density contribution determined by (28)–(30) vanishes when the two-particle function can be represented as the sum of products of one-particle functions. Obviously, if we wish to remain within the required accuracy in the field amplitudes, the two-particle function in (30) must be the equilibrium one:

$$G^{II}_{14,14}(E,E-\Omega;0) = -\{G_4^r(E-\Omega)F_4(E)+F_4(E-\Omega)G_4^a(E)\}\delta_{41}. \quad (A1)$$

Then integrating in (30) with respect to E yields zero.

Using Eq. (36), for $G^{II}_{12,14}(E,E;+1)$ we find that

$$\begin{aligned} G^{II}_{12,14}(E,E;+1) = &-G_{21}^r(E,E-\Omega;+1)F_{14}(E,E;0) \\ &-F_{21}(E,E-\Omega;+1) \\ &\times G_{14}^a(E,E;0)-G_{21}^r(E,E;0)F_{24}(E+\Omega,E;+1) \\ &-F_{21}(E,E;0)G_{14}^a(E+\Omega,E;+1)\}. \end{aligned} \quad (A2)$$

Remaining within the required accuracy in the field amplitudes, we can assume the zeroth harmonics to be the equilibrium harmonics and, hence, diagonal in λ. As a result, Eq. (A2) assumes the form

$$\begin{aligned} G^{II}_{12,14}(E,E;+1) = &-\delta_{14}[G_{21}^r(E,E-\Omega;+1)F_4(E) \\ &+F_{21}(E,E-\Omega;+1)G_4^a(E)] \\ &-\delta_{12}[G_2^r(E)F_{14}(E+\Omega,E;+1)+F_2(E)G_{14}^a(E+\Omega,E;+1)]. \end{aligned} \quad (A3)$$

Substituting (A3) into (29), employing for the first harmonics of one-particle functions the expressions (39) and (40), and integrating with respect to E, we get zero.

Remaining within the required accuracy in the field amplitudes, from Eq. (36) we find for the function $G^{II}_{14,34}(E,E-\Omega;+1)$ the following expression:

$$\begin{aligned} G^{II}_{14,34}(E,E-\Omega;+1) = &-[G_{43}^r(E-\Omega,E-2\Omega;+1)F_4(E) \\ &+F_{43}(E-\Omega,E-2\Omega;+1)G_4^a(E)]\delta_{14} \\ &-[G_4^r(E-\Omega)F_{14}(E+\Omega,E;+1) \\ &+F_4(E-\Omega)G_{14}^a(E+\Omega,E;+1)]\delta_{43}. \end{aligned} \quad (A4)$$

Substituting (A4) into (28), employing the expressions (39) and (40), and integrating with respect to E, we get

$$\begin{aligned} j^\beta(II) = \frac{e^3}{4Vm\Omega^2}\sum_{3,4}\mathrm{Re}\Big\{&\mathscr{E}_\alpha^{\beta*}v_{34}^\alpha\mathscr{E}_{2\alpha}^\alpha\int\frac{dE}{2\pi}D_{34}^*(n_3-n_4) \\ &\times\Big[\frac{1}{(\omega_{34}+2\Omega-i\Gamma_{34})}\Big(\frac{1}{(\Omega-2i\gamma_4)}-\frac{1}{(\Omega-2i\gamma_3)}\Big) \\ &+\frac{1}{(\omega_{43}-\Omega-i\Gamma_{34})}\Big(\frac{1}{(\Omega-2i\gamma_4)}-\frac{1}{(\Omega-2i\gamma_3)}\Big)\Big]\Big\}. \end{aligned} \quad (A5)$$

Obviously, when condition (45) is met, that is, when γ_λ can be neglected in comparison to Ω in the denominators of the fractions in the parentheses in (A5), this expression for $j^\beta(II)$ becomes equal to zero.

Thus, we have demonstrated that the abstract term of the integral equation (34) contributes nothing to the DC density if we use formulas (28)–(30). This implies that the solution to the integral equation (34) also contributes nothing to the DC density if we use formulas (28)–(30). Indeed, physical considerations suggest that the solution to Eq. (34) must be unique for any function (belonging to a certain class) that is the abstract term. Here, in accordance with the Fredholm alternative,[25] the homogeneous equation corresponding to Eq. (34) has no nontrivial solutions.

APPENDIX B

Let us compare the contributions to DC from cross and ladder diagrams. The contribution to the causal two-particle function from a cross diagram with two phonon lines is

$$\begin{aligned} \overset{\times}{G}_{1n,1n'}(x',x;x_1',x'') = &(g/\hbar)^4\int dx_1\,dx_2\,dx_3\,dx_4 G_{ni}(x,x_1)\gamma_{ii'}^k \\ &\times G_{i'l}(x_1,x_3)\gamma_{ll'}^{k'}G_{l'1}(x_3,x_1')G_{1m}(x',x_2)\gamma_{mm'}^{k''}G_{m'j'}(x_2,x_4)\gamma_{j'j}^{k'''} \\ &\times G_{jn'}(x_4,x'')\mathscr{D}_{kk''}(x_1,x_2)\mathscr{D}_{k'k'''}(x_3,x_4). \end{aligned} \quad (B1)$$

A ladder diagram yields

$$\begin{aligned} \overset{=}{G}_{1n,1n'}(x',x;x_1',x'') = &(g/\hbar)^4\int dx_1\,dx_2\,dx_3\,dx_4 G_{ni}(x,x_1)\gamma_{ii'}^k \\ &\times G_{i'l}(x_1,x_3)\gamma_{ll'}^{k'}G_{l'1}(x_3,x_1')G_{1m}(x',x_4)\gamma_{mm'}^{k'''}G_{m'j'}(x_4,x_2)\gamma_{j'j}^{k''} \\ &\times G_{jn'}(x_2,x'')\mathscr{D}_{kk''}(x_1,x_2)\mathscr{D}_{k'k'''}(x_3,x_4) \end{aligned} \quad (B2)$$

with summation over repeated indices implied.

It is easily shown that to compare the contributions to the current density from (B1) and (B2) one need only compare the following quantities (for the sake of simplicity the phonon energy is ignored):

$$\overset{\times}{I}=\int\frac{d\mathbf{k}\,d\mathbf{k}'}{[E(\mathbf{p})-E(\mathbf{p}-\mathbf{k})+i\gamma][E(\mathbf{p})-E(\mathbf{p}-\mathbf{k}')-i\gamma][E(\mathbf{p})-E(\mathbf{p}-\mathbf{k}-\mathbf{k}')+i\gamma]^2},$$

$$\overset{=}{I}=\int\frac{d\mathbf{k}\,d\mathbf{k}'}{[E(\mathbf{p})-E(\mathbf{p}-\mathbf{k})+i\gamma][E(\mathbf{p})-E(\mathbf{p}-\mathbf{k})-i\gamma][E(\mathbf{p})-E(\mathbf{p}-\mathbf{k}-\mathbf{k}')+i\gamma]^2},$$

where $E(\mathbf{p})=p^2/2m$ specifies the dispersion law for electrons in the conduction band. A direct estimate of these expressions shows that their ratio is equal, in order of magnitude, to γ/E, with γ the phonon width of the energy levels, and E the characteristic energy of electrons participating in photoconductivity.

APPENDIX C

Let us estimate the contribution to the DC density from terms in expressions for one-particle functions containing the first and second harmonics of the mass operator. From (38) we easily establish that such a term in the first harmonic of the retarded one-particle function has the form

$$\tilde{G}^r_{\lambda\lambda'}(E,E-\Omega;+1)$$
$$=-\sum_{\lambda_1}M^r_{\lambda\lambda_1}(E,E-\Omega;+1)G_{\lambda_1\lambda'}(E-\Omega,E-\Omega;0)G_{\lambda'}{}^r(E), \tag{C1}$$

where the "tilde" indicates that instead of writing the entire term we have written only the part of interest to us. The zeroth harmonic on the right-hand side of (C1) can be assumed to be the equilibrium one. Obviously, the contribution to the current density is provided by the term next to the last on the right-hand side of (48), in which for $F_{14}(E+\Omega,E;+1)$ we can use (40). As a result the corresponding additional term to the current density is

$$\tilde{j}_{DC}\simeq\frac{e^2\pi N}{4(\hbar\Omega)^2m^2}\sum_{\mathbf{p}}p^\beta\,\mathrm{Re}[(\mathbf{p},\vec{\mathscr{E}}_\Omega^*(\omega_{ip}/2\Omega)(e\mathbf{x}_{ip},\vec{\mathscr{E}}_{2\Omega})]\gamma_p{}^{-1}$$
$$\times M_{pi}{}^r(E_p,E_p-\hbar\Omega;+1)\delta(E_p-E_i-\hbar\Omega). \tag{C2}$$

The ratio of the additional term (C2) to the current density determined by the second term in (55) in order of magnitude is evidently

$$\left|\frac{\tilde{j}_{DC}}{j_{DC}}\right|\approx\left|\frac{M_{pi}{}^r(E_p,E_p-\hbar\Omega;+1)}{D_{pi}}\right|, \tag{C3}$$

and is small if condition (45) is met.

Similarly, it is easy to show that the corrections from the first harmonic of the mass operator to the first harmonics of the correlation and retarded functions lead, in accordance with the last two terms on the right-hand side of (48), to additional terms in the DC density whose expressions coincide with (C2), in order of magnitude.

Next, from (38) we can find the corrections from the first harmonic of the mass operator to the second harmonics of one-particle functions:

$$F_{\lambda\lambda'}(E,E-2\Omega;+2)$$
$$=-\sum_{\lambda_1}F_{\lambda\lambda_1}(E,E-\Omega;+1)M^a_{\lambda_1\lambda'}(E-\Omega,E-2\Omega;+1)$$
$$\times G_{\lambda'}{}^a(E-2\Omega)-\sum_{\lambda_1}G^r_{\lambda\lambda_1}(E,E-\Omega;+1)M^F_{\lambda_1\lambda'}$$
$$\times(E-\Omega,E-2\Omega;+1)G_{\lambda'}{}^a(E-2\Omega), \tag{C4}$$

$$\tilde{G}^r_{\lambda\lambda'}(E,E-2\Omega;+2)$$
$$=-\sum_{\lambda_1}M^r_{\lambda\lambda_1}(E,E-\Omega;+1)G^r_{\lambda_1\lambda'}(E-\Omega,E-2\Omega;+1)G^r_{\lambda}(E-2\Omega). \tag{C5}$$

If for the first harmonics of one-particle functions we use (39) and (40), we can easily calculate the additional terms in the DC density. It appears that under condition (45) in the model used in deriving formula (55) these additional terms also coincide with (C3), in order of magnitude.

Similarly, it is easily shown that the ratio of the additional term in the DC density from the second harmonic of the mass operator to the current density given by formula (55) is, in order of magnitude,

$$\left|\frac{\tilde{j}_{DC}}{j_{DC}}\right|\approx\left|\frac{M_{pp}{}^r(E_p,E_p-2\hbar\Omega;+2)\Gamma_{pi}}{D_{pi}D_{ip}}\right|, \tag{C6}$$

where $\Gamma_{pi}=\gamma_p+\gamma_i$. Obviously, the right-hand side of (C6) is small if condition (45) is met.

The harmonics of the mass operator, which appear on the right-hand sides of (C3) and (C6), can be calculated explicitly for each specific case via the general expression (22). For instance, if we take the complete vertex matrix in the zeroth approximation in the electron–phonon coupling constant and the phonon functions in their equilibrium form in the homogeneous continuum approximation (37), the right-hand sides of (C3) and (C6) are equal, in order of magnitude, to the fraction γ_p/Ω, which in accordance with condition (45) is much lower than unity.

APPENDIX D

Let us show that the contribution to the current density from terms containing products of four matrix elements of impurity–band transitions is negligible in comparison to the value given by (55), provided that condition (45) is met. Such terms appear when two of the four indices over which summation is done in (50) refer to a localized state and the other two to delocalized states. As an example, here is one such term in (50):

$$I^\beta=-2i\sum_{i,\mathbf{p},\mathbf{p}'}j^\beta_{p'i}(\mathbf{v}_{ip'},\vec{\mathscr{E}}_{2\Omega})D_{ip}{}^*D_{pi}{}^*/(\omega_{p'i}+i\Gamma_{p'i})$$
$$\times\left[\frac{(n_{p'}-n_i)}{(\omega_{p'p}-\Omega+i\Gamma_{p'p})(\omega_{p'i}-2\Omega+i\Gamma_{p'i})}\right.$$
$$+\frac{(n_i-n_p)}{(\omega_{p'p}-\Omega+i\Gamma_{p'p})(\omega_{pi}-\Omega-i\Gamma_{pi})}$$
$$+\frac{(n_i-n_p)}{(\omega_{pi}-\Omega-i\Gamma_{pi})(2\Omega+2i\gamma_i)}$$
$$\left.+\frac{(n_i-n_p)}{(\omega_{ip}-\Omega-i\Gamma_{ip})(2\Omega+2i\gamma_i)}\right]+\ldots, \tag{D1}$$

where the dots indicate that there are several other similar terms. All the terms of this form at $\mathbf{p}=\mathbf{p}'$ provide the following contribution to the DC density [if condition (73) is met]:

$$j_{DC}^{\beta}=\frac{eN}{\hbar^3\Omega^3}\sum_{\mathbf{p}} j_{pi}^{\beta}(\mathbf{v}_{ip}, \operatorname{Re}\vec{\mathscr{E}}_{2\Omega})(n_i-n_p)D_{ip}^{*}D_{pi}^{*}$$
$$\times\operatorname{Re}\left[\frac{1}{(\omega_{pi}+\Omega+i\Gamma_{pi})}+\frac{1}{(\omega_{pi}-\Omega-i\Gamma_{pi})}-\frac{1}{(\omega_{pi}-2\Omega+i\Gamma_{pi})}\right.$$
$$\left.-\frac{1}{(\omega_{pi}+2\Omega-i\Gamma_{pi})}\right]. \qquad \text{(D2)}$$

Obviously, the ratio of expression (D2) to expression (55) is equal to γ/Ω, in order of magnitude. The more general expression (D1) provides a contribution to the current density which is no greater than (D2) in order of magnitude. Thus, if condition (45) is met, the contribution to the current from terms containing products of four matrix elements of impurity–band transitions can be ignored.

We now consider the contribution to the current density from the terms in (50) that contain the product of two matrix elements of impurity–band transitions with different values of indices "2" and "4." This occurs when one of these indices refer to a localized state and the other to a delocalized state. Assuming that (51)–(54) and (73) are valid, we easily find from (46) and (50) that

$$j_{DC}^{\beta}=\frac{e^2N}{2\hbar^3\Omega^2m^2}\sum_{\mathbf{p}} p^{\beta}(\mathbf{p}, \operatorname{Re}\vec{\mathscr{E}}_{2\Omega})(n_i-n_p)D_{ip}^{*}D_{pi}^{*}$$
$$\times\operatorname{Re}\left[\frac{1}{(\omega_{pi}-\Omega+i\Gamma_{pi})(\omega_{pi}-2\Omega+i\Gamma_{pi})}\right.$$
$$\left.-\frac{1}{(\omega_{pi}+\Omega+i\Gamma_{pi})(\omega_{pi}+2\Omega+i\Gamma_{pi})}\right].$$

The ratio of this expression to (55) is clearly equal to γ/Ω, in order of magnitude, where γ is a typical value of the inverse time for nonequilibrium electrons to scatter on phonons. We see that if condition (45) is met, the terms considered here are indeed small in comparison to the value specified by (55).

APPENDIX E

Let us estimate the contribution to the DC density from integral terms in the equation for the two-particle function G^{II} containing functions φ and φ^{+} [see Eq. (34)]. In symbolic form the integral term in question has the form

$$\bar{G}^{II}=\{\Phi(G^r\varphi F+F\varphi^{+}G^a)+\mathscr{D}^r(G^r\varphi G^a)+\mathscr{D}^a(G^r\varphi^{+}G^a)\}, \qquad \text{(E1)}$$

where the bar over function $\bar{G}^{II}$ implies that the right-hand side constitutes only a part of the full function G^{II}. It can be demonstrated (in a way similar to that in Ref. 23) that when the electron-phonon interaction is fairly small, Eq. (35) for function φ can be solved iteratively and only the first approximation need be retained, with $\varphi=-G^rG^r$. Then the M th harmonic of $\bar{G}^{II}$ has the form

$$\bar{G}^{II}_{12,34}(E,E';M)=-\frac{g^2}{4}\sum_{\substack{1',2'\\3',4'}}\int\frac{d\mathbf{k}}{(2\pi)^3}J_{2'3'}(\mathbf{k})J^{*}_{4'1'}(\mathbf{k})|f_k|^2$$
$$\times\{(1+2N_k)[G_{22'}^{r}(E',E'-m\Omega;m)G_{3'3}^{r}$$
$$\times(E'\mp\omega_k-m\Omega,E'\mp\omega_k-m\Omega-m'\Omega;m')$$
$$+G_{11'}^{r}(E\mp\omega_k+n\Omega+n'\Omega,E\mp\omega_k+n\Omega;n')$$
$$\times F_{4'4}(E+n\Omega,E;n)+\ldots]+\ldots\}, \qquad \text{(E2)}$$

where summation over all the integers m, m', n, and n' satisfying the condition $m+m'+n+n'=M$ is implied, and the dots refer to other terms that are similar products of harmonics of other combinations of one-particle functions (with the same independent variables and indices). The full expression (E2) can be written symbolically

$$\bar{G}^{II}_{12,34}(E,E';M)=\{(1+2N_k)[G^rG^rG^rF+FG^aG^aG^a]$$
$$\pm[G^rG^rG^rG^a-G^rG^aG^aG^a]\},$$

where the double sign $\pm$ corresponds to the double signs in (E2), over which summation is implied.

In accordance with formula (27) for the DC density, we must consider the second harmonic of the two-particle function. Then we have $M=2$ in (E2). Let us examine in greater detail one of the terms corresponding in (E2) to the set $m=2$ and $m'=n=n'=0$ (for all other terms the estimate remains valid). For the second harmonic of the retarded function we use (41) and for the zeroth harmonics we use the equilibrium functions. Employing (E2), we calculate the value of (47), denoting it by $\bar{I}^{\beta}$. As in the derivation of expression (55) for the current density, we consider the case where the indices 1, 2, 3, and 4 in (E2) pertain to delocalized states, and we also assume that $4=2=\mathbf{p}$ and $1=3=\mathbf{q}$ and allow for (51)–(53) and (60). Here the term explicitly written in (E2) leads to the following expression for $\bar{I}^{\beta}$:

$$\bar{I}^{\beta}=(g^2/4m)\sum_{\mathbf{p}} j_{pp}^{\beta}\int\frac{d\mathbf{k}}{(2\pi)^3}(\mathbf{p}-\mathbf{k},\vec{\mathscr{E}}_{2\Omega})|f_k|^2(1+2N_k)(1-2n_p)(1/2\Omega)(1/2\gamma_p)$$
$$\times\sum_i\frac{|D_{pi}|^2}{(\omega_{pi}-\Omega-i\Gamma_{pi})(\omega_{p,p-k}-2\Omega\mp\omega_k+i\Gamma_{p,p-k})(\omega_{p,p-k}\mp\omega_k+i\Gamma_{p,p-k})},$$

where, as before, the index i denotes a localized state (not to be confused with the unit imaginary number in the denominators). Obviously, integration in (E3) with respect to $\mathbf{k}$ leads to the appearance in the numerator of a quantity of the order of γ_p. For this reason the ratio of the contribution to the DC density from (E3) to the current density given by formula (55) is equal, in order of magnitude, to the small quantity γ/Ω.

Similarly, it is easy to show that the integral terms on the right-hand side of Eq. (34) containing the zeroth har-

monic of the two-particle function G^{II} yield the same value of the current density.

[1] U. Osterberg and W. Margulis, Opt. Lett. **11**, 516 (1986).
[2] V. O. Sokolov and V. B. Sulimov, Izv. Akad. Nauk SSSR, Ser. Fiz. **54**, 2313 (1990) [Bull. Acad. Sci. USSR, Phys. Ser **54** (1990)].
[3] E. M. Dianov, P. G. Kazanskiĭ, and D. Yu. Stepanov, Kvantovaya Elektron. (Moscow) **16**, 887 (1989) [Sov. J. Quantum Electron. **19**, 575 (1989)].
[4] V. M. Fridkin, *Photoferroelectrics*, Springer, Berlin (1979).
[5] V. I. Belinicher and B. I. Sturman, Usp. Fiz. Nauk **130**, 415 (1980) [Sov. Phys. Uspekhi **130**, 199 (1980)].
[6] M. B. Èntin, Fiz. Tekh. Poluprovodn. **23**, 1066 (1989) [Sov. Phys. Semicond. **23**, 664 (1989)].
[7] G. M. Shmelev, Nguen Hong Shon, and G. I. Tsurkan, Izv. Vyssh. Uchebn. Zaved. Fiz. **28**, 84 (1985).
[8] È. M. Baskin and M. B. Èntin, Pis'ma Zh. Eksp. Teor. Phys. **48**, 554 (1988) [JETP Lett. **48**, 601 (1988)].
[9] B. Ya. Zel'dovich and A. N. Chudinov, Pis'ma Zh. Eksp. Teor. Fiz. **50**, 405 (1989) [JETP Lett. **50**, 439 (1989)].
[10] N. B. Baranova, A. N. Chudinov, and B. Ya. Zel'dovich, in *Conference on Lasers and Electro-Optics*, CLEO'90, 21–25 May 1990, Anaheim, Calif., Paper CFJ2, p. 518.
[11] N. B. Baranova, A. N. Chudinov, and B. Ya. Zel'dovich, Opt. Commun. **79**, 116 (1990).
[12] N. B. Baranova, B. Ya. Zel'dovich, A. N. Chudinov, and A. A. Shul-'ginov, Zh. Eksp. Teor. Fiz. **98**, 1857 (1990) [Sov. Phys. JETP **71**, 1043 (1990)].
[13] E. V. Anoĭkin, E. M. Dianov, P. G. Kazanskiĭ *et al.*, Opt. Lett. **15**, 834 (1990).
[14] E. M. Dianov, P. G. Kazanskiĭ, and D. Yu. Stepanov, Kvantovaya Elektron. (Moscow) **17**, 926 (1990) [Sov. J. Quantum Electron. **20**, 849 (1990)].
[15] E. M. Dianov, P. G. Kazanskiĭ, and D. Yu. Stepanov, *Technical Digest of the OSA Annual Meeting*, 4–9 November 1990, Boston, Paper TuU4.
[16] D. Z. Andersen, V. Mizrahi, and J. E. Sipe, *Tech. Digest of OSA Annual Meeting*, 4–9 November 1990, Boston, Paper PDP24, p. 57.
[17] B. Ya. Zel'dovich, *Conference on Quantum Electronics Laser Sciences*, 12–17 May 1991, Paper JTuAl, p. 38.
[18] D. Z. Anderson, V. Mizrahi, and J. E. Sipe, *Conference on Quantum Electronics Laser Science*, 12–17 May 1991, Paper JTuA2, p. 38.
[19] V. M. Churicov, Yu. E. Kapitzky, and B. Ya. Zel'dovich, *Conference on Quantum Electronics Laser Science*, 12–17 May 1991, Paper JTuB4, p. 48.
[20] N. M. Lawandy, C. L. Adler, J. P. Bernardin *et al.*, *Conference on Quantum Electronics Laser Science*, 12–17 May 1991, Postedeadline papers, Paper QPDP14, p. 324.
[21] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys. JETP **20**, 1307 (1964)].
[22] V. B. Sulimov, Candidate's dissertation, Moscow University, Moscow (1979) (in Russian).
[23] V. B. Sulimov, Deposited in VINITI (All-Union Institute of Scientific and Technical Information), No. 2588-78, Moscow (1978) (in Russian); Referativnyĭ Zh. No. 11E, 257 (1978).
[24] V. B. Sulimov, Phys. Status Solidi B **96**, 507 (1979).
[25] V. S. Vladimirov, *Equations of Mathematical Physics*, Mir Publishers, Moscow (1984).
[26] A. I. Ansel'm, *Introduction to Semiconductor Theory*, Mir Publishers, Moscow (1981).
[27] V. L. Bonch-Bruevich and S. G. Kalashnikov, *Semiconductor Physics*, Nauka, Moscow (1977) (in Russian).
[28] E. M. Dianov, V. O. Sokolov, and V. B. Sulimov, *Sixth Europhysical Topical Conference: Lattice Defects in Ionic Material*, 3–7 September 1990, Groningen, Holland, p. 338.
[29] E. M. Dianov, V. O. Sokolov, and V. B. Sulimov, *Abstracts of the Conference on Quantum Chemistry of Solids*, 26–30 November 1990, Riga, Latvia, p. 190.
[30] J. R. Chelikowsky and M. Schluter, Phys. Rev. B **15**, 4020 (1977).
[31] V. B. Sulimov, *Abstracts of the All-Union Conference on Quantum Chemistry of the Solid State*, 8–10 October 1985, Lielupe, Latvia (Latvian University, Riga), p. 60 (in Russian).
[32] R. C. Huges, Phys. Rev. Lett. **30**, 1333 (1973).
[33] E. M. Dianov, P. G. Kazarskiĭ, D. Yu. Stepanov, and V. B. Sulimov, *Proc. of the Conference on Integrated Photoionics Research*, 26–28 March 1990, Hilton Head, S. C., Paper MJ1, p. 46.
[34] A. Kamal, L. Stock, A. Szpak *et al.*, *Technical Digest of the OSA Annual Meeting*, 4–9 November 1990, Boston, Paper TuU5, p. 85.
[35] E. M. Dianov, V. O. Sokolov, and V. B. Sulimov, in *Proceedings (Trudy) of the Institute of General Physics, vol. 23: Fiber Optics,* Moscow (1990), p. 122 (in Russian).
[36] E. M. Dianov, V. O. Sokolov, and V. B. Sulimov, Phys. Status Solidi B **162**, 173 (1990).
[37] E. M. Dianov, V. O. Sokolov, and V. B. Sulimov, Sov. Lightwave Commun. **1**, 1 (1991).

Translated by Eugene Yankovsky