

Surface photovoltaic effect in solids. Theory and experiment for interband transitions in gallium arsenide

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It is shown that when a solid is illuminated an electric current flows along the surface. This surface photocurrent (SPC) appears in the case of oblique incidence of the light on an isotropic semi-infinite crystal, or at normal incidence if the anisotropy of the medium is taken into account. The surface photocurrent is due, on the one hand, to the anisotropy of the momentum distribution of the carriers produced in the bands in interband optical transitions, and on the other to the difference between the momentum relaxation rates of the electrons and holes in the interior of the crystal and on its surface. A general theory of the SPC is constructed, as well as a quantitative theory for GaAs, with account taken of the band structure, of the momentum-scattering mechanisms in this material, and of the Coulomb interaction of the electrons and holes. Surface photocurrent was experimentally observed in GaAs, and its angular, polarization, and spectral characteristics were investigated. The SPC spectrum consisted of oscillations due to partial relaxation of the carrier momentum upon emission of optical phonons. A comparison of the theory with experiments on GaAs has made it possible to determine the degree of momentum relaxation of the electrons and holes scattered by the surface. It has turned out that the scattering is close to specular. The observed discrepancy between theory and experiment can be attributed to the influence of the surface electric field.

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1. INTRODUCTION

It is known that illumination of a medium without a symmetry center gives rise to an electric current (review¹ and the references therein). A related effect in films subject to the classical size effect over the electron mean free path was considered in Ref. 2. The effect was caused by the difference between the specular coefficients of the film surfaces, i.e., the difference between the rates of momentum loss by the carriers at the film surfaces. It is shown in the present paper that a photocurrent can be produced near a surface of a bulky solid. This surface photocurrent (SPC) is due to anisotropy of the momentum distribution of the electrons¹⁾ produced in the conduction band when optical transitions take place in solids. The anisotropy of the photoelectrons can be due both to polarization of the light and to the anisotropy of the crystal. If account is taken of the diffusion and drift of the electrons in a direction perpendicular to the surface, as well as of scattering by the surface, then the momentum loss by the electron is asymmetrical, and this is the cause of the SPC.

The onset of the current is easiest to explain when the electrons are scattered by a rough surface. Assume that the electrons produced by the light have an anisotropic momentum distribution, as shown in Fig. 1. Because of the additional scattering by the surface, the electrons traveling to the right lose their momentum more rapidly than the electrons moving to the left and scattered only in the interior. As a result, an electron flux towards the left is produced. For a specular surface, the photocurrent is equal to zero.

Thus, the SPC is a combination of two effects: the anisotropy of the photoexcitation and the diffuse character of the surface scattering. The anisotropy of the electron photoexcitation (optical alignment) was first

revealed in solids by the linear hot-luminescence polarization.³ The diffuseness of the surface scattering of the electrons comes into play in the high-frequency properties of metals,⁴ as well as in certain transport phenomena in thin semiconductor layers.⁵

We develop in this paper a general theory of the SPC and construct a quantitative theory of the effect for GaAs, with account taken of the peculiarities of the band spectrum, of the momentum scattering processes, and of the properties of the particular semiconductor. The SPC in GaAs was experimentally investigated and the theory compared quantitatively with experiment.

Preliminary theoretical calculations and a report of the first observation of SPC were published in Refs. 6 and 7.

2. EXPERIMENTAL RESULTS AND THEIR QUALITATIVE ANALYSIS

1. The surface photocurrent \mathbf{J} is quadratic in the field of the light wave \vec{E} :

$$J_i = \theta_{ij} (\gamma_{jkm}^* E_k E_l^* + i \gamma_{jkm}^* [\vec{E} \times \vec{E}^*]_k) n_m. \quad (1)$$

Here $\theta_{ij} = \delta_{ij} - n_i n_j$, $\gamma_{jkm}^* = \gamma_{jkm}^*$, and \mathbf{n} is a vector normal to the surface. The current \mathbf{J} differs from

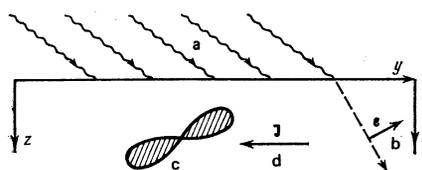


FIG. 1. Illustrative mechanism of the onset of SPC: a—incident light, b—light-polarization vector, c—angular distribution of photoelectrons, d—direction of the total electron flux.

zero in media of any symmetry, including isotropic media. The general form of current along the boundary of an isotropic body is given by two constants:

$$\mathbf{J} = a(\omega) [(\mathbf{E} - n(\mathbf{n}\mathbf{E}))(\mathbf{n}\mathbf{E}) + \text{H.c.}] + ib(\omega) [\mathbf{n} \times (\mathbf{E} \times \mathbf{E}^*)], \quad (2)$$

where $a(\omega)$ and $b(\omega)$ are two scalar functions of the light frequency ω . Relation (1) is not invariant to time reversal, therefore $\gamma_{j_{k_{lm}}^s}$ is an odd function of the dissipative constants, and $\gamma_{j_{k_{lm}}^a}$ is an even function.

The different parities of $\gamma_{j_{k_{lm}}^s}$ and $\gamma_{j_{k_{lm}}^a}$ under time reversal causes the contributions to $\gamma_{j_{k_{lm}}^s}$ and $\gamma_{j_{k_{lm}}^a}$ to have different microscopic properties. The quantity $\gamma_{j_{k_{lm}}^s}$ differs from zero in second order when the kinetic equation is solved by iteration, while the contribution to $\gamma_{j_{k_{lm}}^a}$ arises only when account is taken of the distortion of the probabilities of the transition by the surface, by the spatial inhomogeneity, or by the external field. For convenience in the analysis of the experiments, it is useful to express the photocurrent in terms of the light intensity I incident on the crystal, the incidence angle θ , and the angle φ between the light-polarization vector and the incidence plane. Such an expression can be obtained on the basis of (2) by using the Fresnel formulas. For linearly polarized light, assuming that the square of the refractive index $n^2 \gg 1$, we have

$$\mathbf{J} = -\frac{16\pi a(\omega) I \sin \theta \cos^2 \theta \cos \varphi}{c(n \cos \theta + 1)^2 (n + \cos \theta)} [\mathbf{a}(n + \cos \theta) \cos \varphi + \mathbf{b}(n \cos \theta + 1) \sin \varphi], \quad (3)$$

here c is the speed of light, \mathbf{a} is a unit vector in the incidence plane, $\mathbf{n} \cdot \mathbf{a} = 0$, and \mathbf{b} is a unit vector normal to \mathbf{n} and \mathbf{a} .

2. To calculate the contribution to $\gamma_{j_{k_{lm}}^s}$ we use the kinetic equation for the electron distribution function $f(z, \mathbf{k})$:

$$\frac{k_z}{m} \frac{\partial f}{\partial z} + e\mathbf{E} \frac{\partial f}{\partial \mathbf{k}} = -\Gamma(f - \bar{f}) + W_k \exp(-\kappa z). \quad (4)$$

Here z is the coordinate in the direction normal to the surface, Γ is the frequency of momentum isotropization, W_k is the density of the photoproduced electrons, κ is the light absorption coefficient, and \bar{f} is the distribution function averaged over the angles. We assume the electron dispersion to be quadratic and isotropic, and disregard thermalization and recombination processes. The function $f(z, \mathbf{k})$ must satisfy the usual boundary condition on the surface^{5,8}:

$$f(0, \mathbf{k}) = (1-P)f(0, \mathbf{k}') + P' \frac{(\theta(\mathbf{k}_z) f(0, \mathbf{k}') + \theta(\mathbf{k}_z) f(0, \mathbf{k}'))}{\theta(\mathbf{k}_z)}, \quad k_z > 0. \quad (5)$$

The momentum \mathbf{k}^* differs from \mathbf{k} in that the sign of k_z is reversed, and $\theta(x)$ is the unit step function.

The diffuseness coefficient is $P=0$ for a specular surface and $P=1$ for a surface that scatters the electrons diffusely. $P'=P$ if there is no surface recombination, and $P' < P$ if surface recombination is present.

We put initially $\mathbf{E}=0$. Then the distribution function has a nonisotropic increment that satisfies the boundary condition (5), is equal to zero as $z \rightarrow \infty$, and is given by

$$\varphi(z, \mathbf{k}) = \frac{W_k}{\Gamma} [T_- \exp(-\kappa z) - \theta(k_z) (T_- + (1-P)T_+) \exp(-z/\Lambda_z)]. \quad (6)$$

Here

$$\Lambda_z = k_z/m\Gamma, \quad T_{\pm} = (1 \pm \kappa\Lambda_z)^{-1}.$$

The first term in (6) is due to the diffusion process, and the second is due to scattering by the surface. Integrating $\varphi(z, \mathbf{k})$ with respect to the coordinate, we obtain the distribution-function correction responsible for the current:

$$\varphi(\mathbf{k}) = W_k P \theta(k_z) \Lambda_z T_+ / \Gamma. \quad (7)$$

The quantity W_k^{an} which contributes to the current can be written in the form

$$W_k^{\text{an}} = \frac{(3|e\mathbf{k}|^2 - \mathbf{k}^2) \kappa_{\text{an}} I}{4\pi k^2 m \hbar \omega} \delta(\epsilon_k + E_g - \hbar\omega). \quad (8)$$

Here ϵ_k is the kinetic energy of the photoelectrons, E_g is the transition threshold energy, and $\kappa_{\text{an}}/\kappa$ is the fraction of the anisotropically produced electrons. For example, $\kappa_{\text{an}}/\kappa \sim 1$ for transitions between two s -bands at the center of the Brillouin zone ($\kappa \sim 10^2 \text{ cm}^{-1}$), and also for the transition of Γ_3^+ into Γ_6^+ in the well known III-IV semiconductors ($\kappa \approx 10^4 \text{ cm}^{-1}$).^{9,10} Substituting (7) and (8) in the standard expression for the current, we obtain the total photocurrent

$$\mathbf{J} = (e - n(\mathbf{n}\mathbf{e})) (\mathbf{n}\mathbf{e}) P \frac{\kappa_{\text{an}} e I}{\kappa \hbar \omega} \Lambda_f(\kappa\Lambda), \quad (9)$$

where

$$f(x^{-1}) = (12x^2(1-x^2) \ln \left(\frac{1+x}{x} \right) + 3-8x-6x^2+12x^3)/8.$$

In the limiting cases we have

$$\mathbf{J} = P \frac{\kappa_{\text{an}} e I}{\kappa \hbar \omega} \Lambda \begin{cases} \kappa\Lambda/5 & \text{for } \kappa\Lambda \ll 1, \\ 1/8 & \text{for } \kappa\Lambda \gg 1. \end{cases} \quad (10)$$

Here $\eta = [1 - (\mathbf{n} \cdot \mathbf{e})^2]^{1/2} (\mathbf{n} \cdot \mathbf{e})$. From (9) we see that the photocurrent depends strongly on the polarization of the light. The surface photocurrent vanishes at normal incidence. This is no longer the case if the anisotropy of the medium is taken into account.²

It is of interest to discuss the surface-photocurrent density distribution as a function of the coordinate z . The current density $j(z)$ is either parallel or antiparallel to the total current, and the value of the current, for example at $\kappa\Lambda \ll 1$, is given by

$$j = \eta \frac{e I}{\hbar \omega} \left[\frac{2}{5} (\kappa\Lambda)^2 e^{-\kappa z} - \frac{2-P}{16} e^{-z} (6 - 10\xi - \xi^2 + \xi^3 - \xi^2)(12 - \xi^2) e^{\xi} \text{Ei}(-\xi) \right]. \quad (11)$$

Here $\xi = z/\Lambda$ and $\text{Ei}(x)$ is the integral exponential function. In the cases $z \gg \Lambda$ and $z \ll \Lambda$ these expressions for the photocurrent are simpler:

$$j = \eta \frac{e I}{\hbar \omega} \begin{cases} \frac{2}{5} (\kappa\Lambda)^2 e^{-\kappa z}, & -\frac{3}{8} (2-P) \kappa\Lambda & \text{for } \kappa\Lambda \ll 1, \\ 3P \left(\frac{\Lambda}{z} \right)^2 e^{-z/\Lambda}, & P - 2e^{-\kappa z} & \text{for } \kappa\Lambda \gg 1. \end{cases} \quad (12)$$

$z \gg \Lambda \quad z \ll \Lambda$

It follows from (11) that the current near the surface is antiparallel to the total current. At $z \sim \kappa^{-1}$ or Λ , the current reverses direction. The sign of the SPC is determined by the contribution of the large z to the

total current. We call attention to the fact that the total SPC is proportional to the difference between the specular coefficient and unity, whereas the current density differs from zero also for an absolutely specular surface.

3. We estimate now the order of magnitude of the SPC for $P=1$. Let $\kappa=10^4 \text{ cm}^{-1}$ (interband transition) and $\Lambda=10^{-3} \text{ cm}$. At a light intensity $I=1 \text{ W}$ and $\hbar\omega=1 \text{ eV}$, we have $J\approx 10^{-4} \text{ A/cm}$. The current (9) is larger by approximately two orders of magnitude than the photocurrent in media without a symmetry center.¹ At $\Lambda\approx 10^{-6} \text{ cm}$ the photocurrents are of the same order of magnitude.

In experiments one frequently measures the no-load photo-emf. The value of V depends on the geometry of the experiment. At a sample thickness $b\ll L$, where L is the length of the crystal, V is easy to determine from the condition that the total current through the section of the sample be zero:

$$\int (j(x)+\sigma(x)E)dx=0. \quad (13)$$

If the photocurrent is not uniform over the sample cross section the current density is, naturally, different from zero, i.e., annular currents are present in the crystal. The condition (13) is rewritten in the form well known from elementary physics: $V=JR$, where R is the total resistance of the crystal and J is the total photocurrent. The value of R depends strongly on the type of the crystal. Let us estimate V in the case when $b\approx\max(\kappa^{-1},\Lambda)$ and the photoconductivity is much larger than the dark conductivity of the crystal. In this case

$$R\approx\frac{m\Gamma\gamma\hbar\omega}{e^2\kappa l b}.$$

Here γ^{-1} is the lifetime of the electrons in the conduction band. At $\Gamma>\gamma$ we have for V the estimate

$$V\approx\frac{m\gamma v_T l}{e}(\kappa\Lambda)^{\pm 1} \text{ for } \kappa\Lambda\lesssim 1. \quad (14)$$

Here v_T is the thermal velocity. Putting $l=1 \text{ cm}$, $\kappa\Lambda\approx 1$, $m\approx m_0$ (m_0 is the free-electron mass), $v_T\approx 10^7 \text{ cm/sec}$, and $\gamma=10^9 \text{ sec}^{-1}$, we obtain the estimate $V\approx 1$ volt, in agreement with the estimate given in Ref. 2.

3. EXPERIMENTAL RESULTS AND THEIR QUALITATIVE ANALYSIS

1. Measurements of the SPC were made on epitaxial layers of n -GaAs (with mobility $\mu\sim 10^5 \text{ cm}^2/\text{V}\cdot\text{sec}$ and with electron density $n\sim 2\times 10^{14} \text{ cm}^{-3}$ at $T=78 \text{ K}$), grown on semi-insulating substrates. Two indium contacts were brazed on the surface of the sample, in the form of parallel strips (just as in the measurement of photoconductivity). The light source was a halogen incandescent lamp. The spectral dependences were measured with a DFS-24 monochromator. The spectral resolution was $2\times 10^{-3} \text{ eV}$, and the irradiance was $\sim 10^{-6} \text{ W/cm}^2$. The contacts were not exposed to the light. The measurements were performed at temperatures 1.6 and 4.2 K. In the case of oblique incidence of the light (the incidence plane was perpendicular to the contacts), an emf was produced between the contacts and was measured with an amplifier and a Unipan 232B

synchronous detector. The spectra of this emf were plotted with an automatic recorder. The emf depended on the angle φ between the light-incidence plane and polarization plane, but did not vanish at the angle $\varphi=\pi/2$ at which, according to (3), the SPC vanishes. The origin of the residual (at $\varphi=\pi/2$) emf V_{\perp} is not completely clear. Part of it can apparently be attributed to inhomogeneities in the doping of the sample, in its illumination, etc., and part to the dragging effect,¹¹ to the anisotropic increment to the SPC, or to the photovoltaic effect. The electromotive force V_{SPC} due to the SPC was taken to be the difference between the values of V measured at arbitrary φ and at $\varphi=\pi/2$. Account was taken of the polarization-induced changes in the intensity of the light absorbed by the sample, by normalizing the emf to the value of the photoconductivity. The value of $V_{\text{SPC}}(\omega)$ as a function of $\hbar\omega$ was calculated point by point from the spectra of the emf at $\varphi=0$ and $\varphi=\pi/2$, in steps of 4 meV. The ratio V_{SPC}/V_{\perp} at $\hbar\omega=1.55 \text{ eV}$ varied from sample to sample in the range from 0.15 to 5.

2. We investigated in the experiment the dependences of V_{SPC} on the polarization of the light, on the angle θ of incidence of the light on the sample, and also on the frequency of the light. The dependence of V_{SPC} on the angle φ at $\hbar\omega=1.55 \text{ eV}$ was shown in Fig. 1 of Ref. 7. It is seen that the experiment (points) is well described by the relation $V_{\text{SPC}}=v(\omega,\theta)\cos^2\varphi$ (solid line), which follows from (3). In all the samples, the sign of the SPC at $\hbar\omega=1.55 \text{ eV}$ corresponded to more rapid scattering of the electrons traveling towards the surface, compared with those traveling into the volume of the sample. The sign of the SPC was reversed when the sign of the angle of incidence of the light on the sample was reversed. This sign was determined by comparing the phase of the emf signal from the synchronous detector with the phase of the photoconductivity signal. We note that at normal incidence the polarization-dependent component of the emf always decreased by several times, not to zero as predicted by the SPC theory but to a certain value exceeding the random error. At normal incidence, other relations between the polarization and the emf were observed, depending on the point of the spectrum.²⁾ The measured dependence of the V_{SPC} on the angle θ of light incidence on

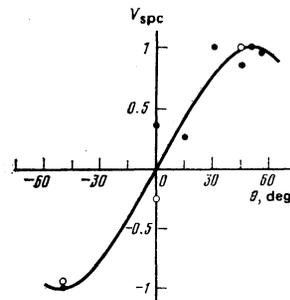


FIG. 2. Dependence of SPC on the angle of incidence of the light on the sample: \bullet sample M243, $N\approx 7\times 10^{14} \text{ cm}^{-3}$; \circ sample C52, $N\approx 5\times 10^{14} \text{ cm}^{-3}$. The experiment and theory are normalized to unity at $\theta=50^\circ$. The value of V_{SPC} at $\theta=0$ was defined as the difference between the emf when the light is polarized perpendicular and parallel to the contacts.

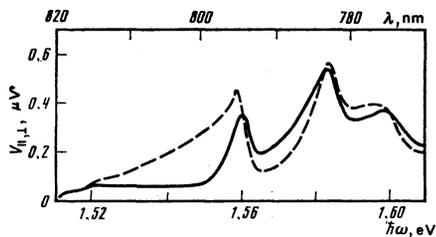


FIG. 3. Spectral dependences of the emf between the contacts in two polarizations: V_{\parallel} ($\varphi=0$, solid line) and V_{\perp} ($\varphi=90^\circ$, dashed line) in sample C52 (with allowance for normalization to the value of the photoconductivity); $\theta=50^\circ$, $T=4.2$ K.

the sample is shown in Fig. 2. The circles show the $V_{SPC}(\omega)$ dependence measured on the sample M243 having the most unfavorable ratio $V_{SPC}/V_{\perp}=0.15$. It is seen nevertheless that the SPC exceeds substantially the scatter of the experimental points and is satisfactorily described by the theoretical relation that follows from (3). According to (3), the effect has a maximum at $\theta \approx 50^\circ$.

Figure 3 shows a spectra of the emf produced when sample C52 was illuminated in two polarizations: with the SPC maximal ($\varphi=0$) and with no SPC ($\varphi=90^\circ$). The spectral dependence of $V_{SPC}(\omega)$ was shown in Fig. 2 of Ref. 7. It could be seen from that figure that the SPC spectrum consists of damped alternating-sign oscillations due to the stepwise thermalization of the electrons with emission of optical phonons. Similar spectra were obtained for all the investigated samples. The most substantial difference between the spectra of the different samples, and also between the spectra measured at different temperatures, was that the magnitude and sign of the SPC were changed at large ($\hbar\omega \geq 1.6$ eV) photon energies. In the indicated spectrum, $V_{SPC}(\omega)$ tends to zero at large $\hbar\omega$. In certain spectra, $V_{SPC}(\omega)$ assumed a constant value at large $\hbar\omega$. The value of this "dc component" was always smaller than V_{SPC} at the maximum of the first oscillation.

3. We present now a qualitative analysis of the results of the experiment. The dependences of V_{SPC} on the polarization and on the light-incidence angle are simple and are well described by the theory, so that principal attention will be paid to the SPC spectra. Figure 4 shows the band structure of GaAs near $k=0$ and two channels of interband transitions under the influence of light with $\hbar\omega > E_g$: 1—from the heavy-hole band and 2—from the light-hole band. Thus, for light with $E_g < \hbar\omega < E_g + \Delta$, contributions to the SPC can be made by four "species" of particles (electrons and holes from each channel). We shall consider the contribution of particles of one species, say electrons from the heavy channel. The form of the SPC spectrum is closely related with the energy dependence of the dominant electron-momentum scattering mechanisms. We propose that in GaAs at helium temperatures, for electrons with $E < \hbar\Omega_{LO}$ (passive band), momentum scattering by charged impurities predominates. Since the mean free path increases with increasing energy, the SPC increases with increasing $\hbar\omega$. At an energy $E = \hbar\Omega_{LO}$, rapid emission of a longitudinal opti-

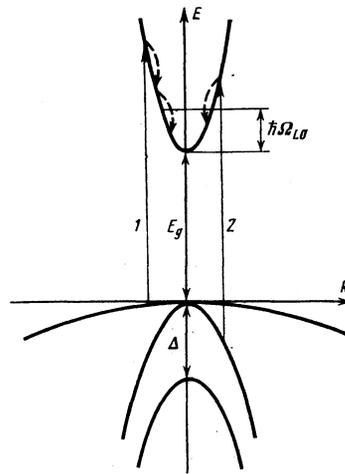


FIG. 4. Band structure of GaAs. The horizontal lines in the conduction band limit the passive band for the electrons. 1 and 2—interband transitions respectively from the band of heavy and light holes. The dashed lines show the processes of thermalization with emission of optical phonons.

cal phonon takes place and makes the electron distribution isotropic, and a jumplike decrease of the SPC is observed as a result. At $E > \hbar\Omega_{LO}$, the phonon emission makes the electron distribution function only partially isotropic.¹² The isotropization terminates completely in the passive zone. Thus, the SPC spectrum of the electrons from the heavy channel should consist of damped oscillations that are connected with emission of several optical phonons. The energies of the minima of these oscillations are described by the formula

$$\hbar\omega = E_g + n\hbar\Omega_{LO}(1 + m_e/m_h), \quad (15)$$

where n is an integer.

The form of the first oscillation is determined by the processes of isotropization of the electron distribution in the passive zone, and that of the succeeding oscillations is determined by these processes as well as by the process of isotropization upon emission of the optical phonons. The contribution of the electrons from the light channel are of opposite sign and have a larger oscillation period. The largest period is possessed by the contribution of the heavy holes ($\approx 9\hbar\Omega_{LO}$). The resultant SPC spectrum is a superposition of all four contributions and should have the form of damped alternating-sign oscillations. This is precisely the form of the experimentally measured spectrum (Fig. 2 of Ref. 7). At $1.52 < \hbar\omega < 1.56$ eV, the contribution of the electrons from the heavy channel predominates; at $\hbar\omega \approx 1.56$, these electrons emit one optical phonon, and the contribution of the electrons from the light channel becomes predominant, therefore V_{SPC} reverses sign, etc.

4. QUANTITATIVE THEORY OF SURFACE PHOTOCURRENT IN GALLIUM ARSENIDE

1. For a quantitative description of the experimental results, the SPC theory should take into account the distinguishing features of the band spectrum and of the

momentum scattering mechanisms in GaAs. In the calculation of the probability $W_{\mathbf{k}}$ of production of an electron and a hole in interband absorption of light in GaAs, we shall assume that the energies of the photons are insufficient for the production of detached holes: $\hbar\omega < E_g + \Delta$. Calculation of the anisotropy of the probability of electron and hole production in this situation is not trivial, even though all the parameters of the band structure are well known. To obtain a quantitative result it is necessary to take into account the interaction of the electron and of the hole in the phototransition process (the excitonic effect). The degeneracy of the band structure complicates the problem considerably.

The probability of production of an electron and a hole is

$$W_{\mathbf{k},\nu} = (2\pi)^{-1} \sum_{\alpha,\beta} |D_{\alpha,\nu\beta}(\mathbf{k}) \tilde{\mathbf{E}}|^2 \delta(\epsilon_{\mathbf{k}}^{\nu} + E_{\beta} - \hbar\omega). \quad (16)$$

Here $\epsilon_{\mathbf{k}}^{\nu} = \hbar^2/2m_{\nu}$ is the kinetic energy of the electron and hole of species ν ($\nu = +1$ for light holes, $\nu = -1$ for heavy holes), $\alpha, \beta = \pm 1/2$ are the spin indices of the electron and of the holes, $D_{\alpha,\nu\beta}(\mathbf{k})$ is the matrix element of the dipole-moment operator between the valence band and the conduction band. Taking the Coulomb interaction into account, $[D_{\alpha,\nu\beta}(\mathbf{k})]_i$ can be written in the form

$$(D_{\alpha,\nu\beta}(\mathbf{k}))_i = \frac{ieP}{\omega} \psi_{i\alpha\mathbf{k}}^{\nu\beta}(0), \quad (17)$$

where $\psi_{i\alpha\mathbf{k}}^{\nu\beta}(\mathbf{r})$ is the wave function of the continuous spectrum of the electron-hole system with zero total momentum, which goes over at large r into the free hole function $\exp(i\mathbf{k} \cdot \mathbf{r}) u_{i\alpha\mathbf{k}}^{\nu\beta}$ (the set of subscripts $i\alpha$ numbers here six states in the valence band). In its transformation properties, $\psi_{i\alpha\mathbf{k}}^{\nu\beta}(0)$ is similar to $u_{i\alpha\mathbf{k}}^{\nu\beta}$, therefore these functions differ by a factor (the Sommerfeld factor) which is a function³⁾ of $|\mathbf{k}|$:

$$\psi_{i\alpha\mathbf{k}}^{\nu\beta}(0) = Z_{\mathbf{k},\nu} u_{i\alpha\mathbf{k}}^{\nu\beta}. \quad (18)$$

For the function $u_{i\alpha\mathbf{k}}^{\nu\beta}$ we can obtain the relation:

$$\sum_{\alpha,\beta} u_{i\alpha\mathbf{k}}^{\nu\beta} u_{j\beta\mathbf{k}}^{\nu\beta} = \frac{1}{4} \left[(3-\nu + (1+\nu) \cos \psi) \delta_{ij} - \frac{k_i k_j}{k^2} (1-3\nu + 3(1+\nu) \cos \psi) \right], \quad (19)$$

$\cos \psi = -D^{-1}(\Delta - 3Lk^2)/3$; $D = (\Delta^2 - 2Lk^2\Delta/3 + \bar{\Delta}^2 k^4)^{1/2}$;

$\sin \psi > 0$; $\bar{L} = (M - L) \approx -N$; L , M , and N are the standard parameters of the Kane theory (Ref. 10).⁴⁾ The dispersion laws of the light and heavy holes are in this case

$$E_i(k) = Mk^2 - Lk^2/2 + (D - \Delta)/2, \quad E_h(k) = Mk^2.$$

An effective method of obtaining relations of the type (19) was developed in Ref. 13. In the derivation of such relations it is assumed that the kinetic energy of the electrons and holes is much less than E_g . Substituting (18) and (19) in (16), we obtain $W_{\mathbf{k},\nu}$ in the form

$$W_{\mathbf{k},\nu} = \frac{3\kappa_{\nu} I}{32\pi\hbar\omega} \frac{1}{k^2 m_{\nu}} [(3-\nu + (1+\nu) \cos \psi) k^2 - |k\mathbf{e}|^2 (1-3\nu + 3(1+\nu) \cos \psi)] \delta(\epsilon_{\mathbf{k}}^{\nu} + E_{\beta} - \hbar\omega), \quad (20)$$

$$\kappa_{\nu} = \frac{4}{3} \frac{e^2}{\hbar c} \frac{\mu_{\nu} P^2 k_{\nu}}{\hbar^2 \omega n} |Z_{\mathbf{k},\nu}|^2 \quad (21)$$

is the partial coefficient of the light absorption, and

$\kappa = \kappa_{+} + \kappa_{-}$. Without allowance for the Coulomb interaction, an expression equivalent to (20) was obtained for $W_{\mathbf{k},\nu}$ in Ref. 9. If $\epsilon_{\mathbf{k}}^{\nu} \ll \Delta$, then $\cos \psi \rightarrow -1/3$ and expression (21) goes over into the corresponding formulas of Refs. 9 and 12 for $W_{\mathbf{k},\nu}$.

In the Luttinger theory¹⁴ it is possible to obtain for $Z_{\mathbf{k},\nu}$ the expression $Z_{\mathbf{k},\nu} = \psi_{\mathbf{k}3/2\nu}(0)$, where $\psi_{\mathbf{k}3/2\nu}(\mathbf{r})$ is the wave function of an s -wave with total angular momentum $F = 3/2$. The normalization of $\psi_{\mathbf{k}3/2\nu}(\mathbf{r})$ is fixed by the asymptotic condition $\psi_{\mathbf{k}3/2\nu}(\mathbf{r}) \rightarrow \sin(\mathbf{k} \cdot \mathbf{r}) + \varphi/kr$ as $r \rightarrow \infty$. We do not know at present, however, the analytic form of $\psi_{\mathbf{k}3/2\nu}(0)$. We note that if the electron-hole interaction is neglected we get $Z_{\mathbf{k},\nu} = 1$, and if the degeneracy effect is neglected (the Elliott theory¹⁰), then $|Z_{\mathbf{k},\nu}|^2 = f(1 - e^{-f})^{-1}$; $f = 2\pi/k_{\nu} a_{\nu}$; k_{ν} and a_{ν} are respectively the momentum and the Bohr radius of the electron and hole.

To calculate the SPC we can use formulas (6) and (9). When (20) is taken into account, this leads to the following expression for the constant $a(\omega)$:

$$a(\omega) = \frac{ec}{16\pi\hbar\omega} \sum_{\nu=\pm 1} (3\nu - 1 - 3(1+\nu) \cos \psi) \frac{\kappa_{\nu}}{\kappa} [P_{\nu} \Lambda_{\nu}^{\nu} f(\kappa \Lambda_{\nu}^{\nu}) - P_{\nu} \Lambda_{\nu}^{\nu} f(\kappa \Lambda_{\nu}^{\nu})], \quad (22)$$

$$\Lambda_{\nu}^{\nu} = k_{\nu}/m_{\nu} \Gamma_{\mathbf{k},\nu}^{\nu},$$

m_{ν} , $\Gamma_{\mathbf{k},\nu}^{\nu}$ and m_{ν} , $\Gamma_{\mathbf{k},\nu}^{\nu}$ are the masses and momentum isotropization frequencies of the electron and ν -holes (the mass of the light hole depends on the momentum).

2. When optical phonons are emitted, partial isotropization of the photoproduced electrons and holes takes place. The decrease of the anisotropy of the distribution of the photoelectron upon emission of optical phonons was considered in Ref. 12. We consider this effect for an arbitrary anisotropic distribution of the electrons and holes, and take into account the processes of conversion of light and heavy holes, as well as the nonparabolicity of the light-hole band. The problem is formulated mathematically as follows. The generation term for the electrons (holes) is of the form

$$W_{\mathbf{k}}^0 = \sum_{l,m} a_{l,m}^0 Y_{lm}(\theta, \varphi) (km)^{-1} \delta(E_{\mathbf{k}} - E_0). \quad (23)$$

Here $Y_{lm}(\theta, \varphi)$ are spherical functions with angular momentum l and its projection m . In the chosen normalization, the coefficient $a_{l,m}$ has the meaning of the "number of particles" with specified angular distribution $Y_{lm}(\theta, \varphi)$. After emission of p optical phonons, the generation term goes over into

$$W_{\mathbf{k}}^p = \sum_{l,m} a_{l,m}^p Y_{lm}(\theta, \varphi) (km)^{-1} \delta(E_{\mathbf{k}} - E_0 - p\Omega_{LO}). \quad (24)$$

The connection between the coefficients $a_{l,m}^p$ and $a_{l,m}^0$ is given by the transition matrix

$$a_{l,m}^p = \sum_{l',m'} U_{l,m,l',m'}^p a_{l',m'}^0.$$

In the case of holes, the matrix U depends on the hole indices ν and ν' . To find the matrices U , it is necessary to write down and solve a system of balance equations for $W_{\mathbf{k}}^{\nu}$ and $W_{\mathbf{k}}^{\nu'}$. For electrons we have the following equation:

$$\int \frac{\delta(E_k - E_{k'} - \Omega_{LO})}{|k - k'|^2} dk' W_k^p - \int \frac{\delta(E_k - E_{k'} + \Omega_{LO})}{|k - k'|^2} W_k^{p-1} dk' = 0.$$

For holes

$$\sum_{\nu'} \left[\int \frac{\delta(E_{\nu k} - E_{\nu k'} - \Omega_{LO})}{|k - k'|^2} \Gamma_{\nu\nu'}(k', k) dk' W_k^{p\nu'} - \int \frac{\delta(E_{\nu k} - E_{\nu k'} + \Omega_{LO})}{|k - k'|^2} \Gamma_{\nu\nu'}(k, k') W_k^{p\nu'} dk' = 0. \quad (25) \right.$$

Here E_k and $E_{\nu k}$ are the dispersion laws of the electron and of the ν -holes, $\Gamma_{\nu\nu'}(k', k) = \Gamma_{\nu\nu'}(k, k')$ are the averaged squares of the hole functions⁵⁾:

$$\Gamma_{\nu\nu'}(k', k) = \sum_{\beta, \beta'} \left| \sum_{\alpha, \alpha'} u_{\alpha k'}^{\nu\nu'} u_{\alpha k}^{\nu\nu'} \right|^2 = (1 + \nu\nu') + \nu_{\nu\nu'}(\psi) (1 - |kk'|^2/k^2 k'^2), \quad (26)$$

$$\nu_{\nu\nu'}(\psi) = \frac{1}{64} [8(1 - \nu\nu') (3 - \cos \psi + 2\sqrt{2} \sin \psi - 24(1 - \nu)(1 - \nu') - (1 + \nu)(1 + \nu') (14 - 4 \cos \psi + 8\sqrt{2} (1 + \cos \psi) \sin \psi + 14 \cos^2 \psi)].$$

From the invariance of these equations to rotations, it follows that the matrix A is diagonal in l and m , and does not depend on m :

$$U_{lm, l'm'}^p = \delta_{ll'} \delta_{mm'} U_l^p.$$

After quite straightforward but cumbersome operations, we obtain explicit expressions for the isotropization functions U_l^n and $U_{l\nu}^n$. For the electrons we have

$$U_l^n = U_l^n(E_0, m_e, \Omega_{LO}) = \prod_{p=1}^n (Q_l(q_p)/Q_0(q_p)), \quad U_l^0 = 1. \quad (27)$$

Here

$$q_p = (k_p^2 + k_{p-1}^2)/2k_p k_{p-1}, \quad E_{k_p} = E_0 - p\Omega_{LO}, \quad n = \text{Int}(E_0/\Omega_{LO}),$$

$Q_l(x)$ are Legendre functions of the second kind. In the case $l=2$ are resolved for $U_2^2(E_0)$ coincides with the result of Ref. 12. For holes, the expression is more complicated:

$$U_{l\nu}^n = U_{l\nu}^n(E_0, m_h, \Delta, \Omega_{LO}) = \left(\prod_{p=1}^n Z_{lp} \right)_{\nu\nu'}, \quad U_{l\nu}^0 = \delta_{\nu\nu}. \quad (28)$$

The matrix Z_{lp} is of the form

$$(Z_{lp})_{\nu\nu'} = A_{p\nu}^{-1} m_{\nu} T_{l\nu\nu'}(q_p^{\nu\nu'}).$$

Here

$$q_p^{\nu\nu'} = (k_p^2 + k_{p-1}^2)/2k_p k_{p-1}, \quad E_{k_p} = E_0 - p\Omega_{LO},$$

$$T_{l\nu\nu'} = (1 + \nu\nu') Q_l(q_p^{\nu\nu'}) + \nu_{\nu\nu'}(\psi) [(2l+3)(2l+1)(2l-1)]^{-1}$$

$$\times [2(2l^2 + 3l^2 - l - 1) Q_l(q_p^{\nu\nu'}) - (2l+3)l(l-1) Q_{l-2}(q_p^{\nu\nu'}) - (2l-1)(l+1)(l+2) Q_{l+2}(q_p^{\nu\nu'})],$$

$$A_{p\nu} = \sum_{\nu'=-1}^{\nu'+1} m_{\nu'} T_{l\nu\nu'}(q_p^{\nu\nu'}).$$

Figure 5 shows plots of the isotropization function $U_l^n(E_0)$ for $l=0, 1, 2$. The index n is determined in this case by the relation $n = \text{Int}(E_0/\hbar\Omega_{LO})$.

We now obtain an expression for the SPC in the active zone of GaAs. To this end it suffices to replace in (22) the quantities $\Lambda_{\nu}^e(\kappa, \Lambda_{\nu}^e)$ by the corresponding quantities in the passive zone, with allowance for the partial isotropization of the electron and hole generation function, and also for the process of hole conver-

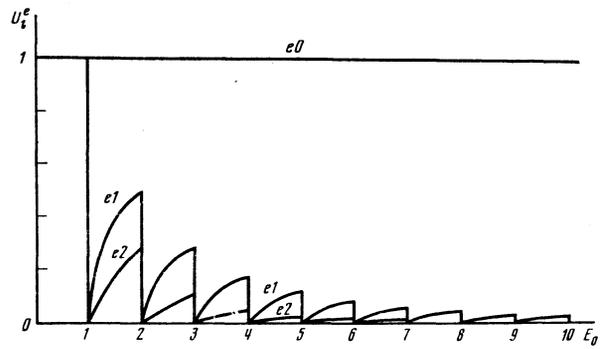


FIG. 5. Electron isotropization function $U_l^n(E_0)$ at $l=0, 1, 2$. The energy E_0 is marked in units of the energy of the optical phonon $\hbar\Omega_{LO}$. The symbols el label the plots of the functions U_l^n .

sion. For the electrons we have

$$[\Lambda_{\nu}^e f(\kappa, \Lambda_{\nu}^e)]_{\text{act}} + U_2^e(\omega) [\Lambda_{\nu}^e f(\kappa, \Lambda_{\nu}^e)]_{\text{pas}},$$

and for the holes

$$[\Lambda_{\nu}^h f(\kappa, \Lambda_{\nu}^h)]_{\text{act}} + \sum_{\nu'=-1}^{\nu'+1} U_{2\nu\nu'}^h(\omega) [\Lambda_{\nu}^h f(\kappa, \Lambda_{\nu}^h)]_{\text{pas}}. \quad (29)$$

The expression obtained for the photocurrent as a result of a combination of formulas (2), (22), and (29) at $\cos \psi = -1/3$ goes over into the expression for the SPC used in Ref. 7.

3. In the passive zone of GaAs, we took into account the loss of electron and hole momentum following scattering by charged impurities and upon emission of acoustic phonons. The frequencies Γ_n of electron and hole momentum losses in scattering by charged impurities were calculated by the Rutherford formula⁶⁾

$$\Gamma_n = \frac{4\pi N m e^4}{e^2 k^2 \hbar} \ln(k r_{\text{scr}}), \quad (30)$$

where N is the density of the charged impurities, ϵ_0 is the static dielectric constant, and r_{scr} is the screening radius.

The frequency of electron momentum loss upon emission of acoustic phonons is of the form

$$\Gamma_{\text{ac}} = 4\sigma^2 m k^2 / 5\pi \rho \hbar^2 c_s. \quad (31)$$

Here σ is the deformation-potential constant (for electrons $\sigma_e \sim 7$ eV, Refs. 15 and 16), ρ is the crystal density, and c_s is the speed of sound. An estimate based on formulas (30) and (31) shows that the contribution of the acoustic phonons to the momentum loss by electrons in the passive zone must be taken into account at $N \leq 10^{14} \text{ cm}^{-3}$.

For a heavy hole, the principal process of momentum loss is emission of an acoustic phonon, while for a light hole it is the conversion into a heavy one upon emission of an acoustic phonon. Formula (31) can be used to calculate $\Gamma_{\text{ac}}^{h,l}$ by using in (31) the momentum and mass of the heavy hole. The constants σ_e^2 and σ_h^2 are bilinear combinations of the hole deformation constants a , b , and d (the notation of Bir and Pikus¹⁰). The values of b and d are known from optical measurements.¹⁵ The constant a , just as the electron constant σ_e ,¹⁶ was ap-

parently determined from kinetic phenomena only in order of magnitude, and amounts to several electron volts. We used for the calculations $\sigma_e \approx \sigma_h \approx 5$ eV. For holes, the contribution of the acoustic phonons to the momentum loss process becomes substantial at $N \leq 10^{16}$ cm⁻³. The difference by two orders of magnitude between the densities of the charged impurities, for which account must be taken of the contribution of the acoustics to the electron and hole momentum losses, is due to the fact that at a fixed particle energy we have $\Gamma_{ac}^e/\Gamma_{ac}^{h,1} \sim m_e^2/m_h^2$.

5. QUANTITATIVE COMPARISON OF THEORY AND EXPERIMENT FOR GaAs AND DISCUSSION OF RESULTS

We have carried out a numerical calculation of the SPC on the basis of Eqs. (22) and (29). The momentum isotropization frequencies were calculated from formulas (30) and (31). The following parameters of GaAs were used: $m_e = 0.067m_0$, $m_l = 0.086m_0$, $m_h = 0.550m_0$, $m_\Delta = 0.183m_0$, $\Delta = 0.34$ eV; $E_g = 1.518$ eV, and $\hbar\Omega_{LO} = 0.0367$ eV. The parameters L , M , N , and P are uniquely determined by the values of the effective masses and energy gaps. Since the accuracy of the parameter P was insufficient, we have determined P from the experimental data on the absorption coefficient κ .¹⁷ The value of $|Z_w|^2$ in (21) was chosen in accordance with the Elliott theory. A least-squares approximation of the experimental $\kappa(\omega)$ curve yielded $m_e P^2/E_g = 10.3$, from which P_Δ was obtained. The diffuseness coefficients P_e and P_h are free parameters of the theory and characterize a concrete sample. These parameters were determined by us by a best fit of the theory to the experiment. The theory describes well the emission thresholds of the optical phonons, the sign reversal, and the ratio of the amplitudes of the oscillations. The theoretical curve with allowance for the scattering of the holes by the deformation potential of the acoustic phonons, as well as with allowance for the nonparabolicity of the light-hole band, does not differ substantially from the theoretical spectrum shown in Fig. 2 of Ref. 7. The scattering of the electrons by the surface turned out to be close to specular, $P_e \approx 0.2$. The hole contribution to the SPC is substantially lower than the electron contribution, therefore the corresponding diffuseness coefficients P_h can be roughly estimated as comparable with P_e in order of magnitude.⁷⁾

Generally speaking, the diffuseness coefficient of the carrier scattering by the surface depends on the carrier energy.⁸ Allowance for this dependence would improve the agreement between the theory and experiment at least in the region of the first oscillation. However, this procedure is apparently not advisable, since the form of the first oscillation varies from sample to sample and furthermore varies with temperature for the same sample. It is possible that the interaction of the electrons with the surface, and consequently the form of the oscillations, is strongly influenced by the surface electric field, the value of which was not monitored in the experiment. At low temperatures, total or partial rectification of the bands

is possible in GaAs,¹⁸ and the degree of this rectification depends on the properties of the surface states, on the temperature, and on the intensity and duration of the illumination. By varying the intensity of the surface field and the regions covered by it in various samples and in the same sample, depending on the experimental conditions, it is apparently possible to explain the difference between the contributions of the surface field to the SPC. We note that an electric field perpendicular to the sample surface can itself lead to the appearance of current along the surface (anisotropic photoconductivity).^{19,20} As shown by a qualitative analysis, the "dc component" in the V_{SPC} spectrum can be attributed to a heavy-hole contribution whose magnitude and sign vary with changing surface field. It is more probable, however, that the "dc component" is an instrumental effect connected with superposition of a polarization-dependent inhomogeneity of the illumination field of the sample on the inhomogeneity of the sample itself.

It should be noted that the accuracy with which the diffuseness P_e is determined depends on exact knowledge of the electron mean free path Λ_e , the intensity of the light absorbed by the sample, and the resistance of the illuminated part of the sample. The error in Λ_e can be connected with the error in the determination of the density of the charged particles N from hole measurements. In addition, we note that the value of Λ_e at the top of the passive band, estimated from (30) for $N = 5 \times 10^{14}$ cm⁻³, turned out to be $\Lambda_e \approx 20$ μ m. The process of electron scattering in semiconductors at such large Λ_e was not investigated previously in experiment. It is not excluded that under these conditions other weaker momentum relaxation mechanisms become significant, such as scattering by neutral impurities or macroscopic defects. This can also lead to corrections to the value of the diffuseness.

6. CONCLUSION

The SPC theory developed in this paper describes quite well the main features of this phenomenon, which was first observed and experimentally investigated in GaAs. The magnitude and spectrum of the surface photocurrent are governed by an aggregate of momentum relaxation mechanisms of the photoexcited electrons and holes. The presence of oscillations and their damping are due to partial isotropization of the distribution of the particles upon emission of optical phonons. The form of the first oscillation is determined to a considerable degree by the dependence of the momentum mean free path in scattering by the impurity ions on the particle energy, and the total amplitude of the spectrum is determined by the degree of diffuseness of the surface scattering and by the absolute magnitude of the momentum mean free path. Thus, the theoretical and experimental results obtained in the paper make it possible to investigate process of carrier momentum relaxation in the bulk and near the surface of the crystal, and also near interfaces of various types.

¹⁾For the sake of argument, we are discussing the electron contribution to the current; the holes make an analogous

contribution to the SPC.

²⁾This emf can also be due to inhomogeneities of the samples and of its illumination, to anisotropic additions to the SPC, and to the dragging effect and volume photovoltaic effects.

³⁾This relation is valid if one neglects the contribution of the band of detached holes, which is small in GaAs relative to the parameter $(Ry^*/\Delta)^{1/2} \approx 0.1$, where Ry^* is the exciton binding energy in GaAs.

⁴⁾We neglect corrugation effects, which in GaAs necessitate a correction smaller than 10% to the isotropic effects.

⁵⁾In the limit $E_{ph} \ll \Delta$, $\Gamma_{ph} = 1 - \nu\nu' (1 - |\mathbf{k} \cdot \mathbf{k}'|^2/k^2k'^2)/2$.

⁶⁾We note that the hole scattering cross section is not described by the Rutherford formula, since the band structure of the valence band is degenerate. The error should not be too large, inasmuch as in the perturbation-theory calculation the cross section of the holes differs from the Rutherford cross section by a factor on the order of unity, and in the quasiclassical approximation the degeneracy effects are negligible.

⁷⁾The value of P_s differs from the one given by us in Ref. 7. The difference is due to a refinement of the absolute intensity of the light flux emerging from the monochromator. The statement made in Ref. 7 that the surface scattering of the hole is more specular than that of the electrons is due to failure to take into account the scattering of the holes by the acoustic phonons.

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