Linear photovoltaic effect in p-type gallium arsenide

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A theory of the intraband linear photovoltaic effect (PVE) is developed for semiconductors with degenerate bands. It is found experimentally that a PVE due to free carriers arises in p-GaAs at $T \geq 250$ K as the result of optical transitions between the sublevels of heavy and light holes, accompanied by scattering by phonons. The temperature dependences of the photovoltaic current can be satisfactorily described by a PVE theory in which scattering by optical phonons is taken into account.

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1. INTRODUCTION

A number of recently discovered new photovoltaic effects (PVE) are, unlike the Dember effect, not connected with the separation of a carrier pair as a result of inhomogeneity of the sample or inhomogeneity of its illumination. The first of these effects was dragging due to momentum transfer from the photon to the electron (see the reviews[1,2]). In crystals without inversion center, two the other observed new effects were named the linear and circular PVE (see the reviews[3-5]). As is now clear, these effects are due to asymmetry of the elementary interactions between the electrons and photons, phonons, or impurities in crystals without inversion center. The linear PVE is produced in piezoelectrics illuminated by linearly polarized light, and under certain conditions also by unpolarized light. The circular photocurrent is produced in gyrotropic crystals illuminated by circularly polarized light and changes direction when the sign of the circular polarization is reversed.

In semiconductors, the linear PVE was first observed in Te (Refs. 6, 7) and next in GaAs (Ref. 8) and GaP (Ref. 9). It is possible that this effect was already observed in dielectrics in the past century (see Ref. 10). As a new effect, however, it was identified only in 1974 in Refs. 11 and 12. It was in these references that one of the possible mechanisms of the linear photogalvanic effect was first proposed, namely the asymmetry of the distribution of the carriers excited by light from impurities having an oriented dipole moment. [The mechanisms proposed earlier in Refs. 6, 7, and 9 turned out to be wrong (see Refs. 3–5).] A detailed theory of this and other possible mechanisms of the linear PVE was developed in Refs. 13–15 and in other studies reported in detail in the reviews[3-5]. Thus, it was shown in Ref. 15 that when light is absorbed by free carriers the appearance of the linear PVE is accounted for by two-photon processes or multiple scattering from impurities. It was established in Ref. 16 that in a semiconductor with a complex band consisting of several branches the linear PVE is produced in transitions between branches also in the case of one-phonon processes or in the case of single scattering by impurities, in analogy with the situation in interband transitions.[15] It was also suggested in Ref. 16 that it is precisely this mechanism which is responsible for the PVE observed in GaP, GaAs, and possibly also in Te.

Depending on the experimental conditions (temperature, frequency of exciting light, doping level), different mechanism can predominate in one and the same crystal. An experimental investigation of the density and temperature dependence of the PVE[17] has shown that a PVE in p-GaAs illuminated by a CO_2 laser at low temperatures is due to photoexcitation of carriers from impurity levels. At room and higher temperatures, phonon mechanisms predominate, as is evidenced by the independence of the photo-emf of the impurity density under these conditions.

The aim of the present paper is to ascertain which of the possible phonon mechanisms predominates in p-type III-IV compounds at high temperatures. To this end, we examine first the photocurrent temperature dependences that result from the one-phonon and two-phonon mechanisms of the PVE, and analyze the asymmetry of which of the elementary processes makes the predominant contribution to these mechanisms and the cause of this asymmetry. We present next results of an experimental investigation of the temperature dependence of the photocurrent in p-GaAs samples with different hole densities and compare them with the theoretical results.

2. TEMPERATURE DEPENDENCE OF THE PHOTOCURRENT FOR THE ONE- AND TWO-PHONON MECHANISMS

The photocurrent in crystals of symmetry T_d is described by the following phenomenological formula:

$$j_\alpha = I[\chi|\delta_{\alpha\beta\gamma}|e_\beta e_\gamma^* + (T_1+T_2|e_\alpha|^2)q_\alpha], \tag{1}$$

where I the intensity of the light; e_α and q_α are the projections of the polarization vector $\mathbf{e}$ and of the wave vector $\mathbf{q}$ of the light on the principal axes of the crystal x, y, and z, and $\delta_{\alpha\beta\gamma}$ is a unit antisymmetrical tensor of third rank. The coefficients T_1 and T_2 determine the dragging effect, and the coefficient χ determines the linear PVE. By orienting the crystal in a definite manner, it is possible to measure these effects separately. Thus, when the light propagates along the [110] axis the photocurrent in the transverse directions [$1\bar{1}0$] and [001] is determined only by the linear PVE:

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$$j_{[1\bar{1}0]}=I\chi\sin 2\theta, \quad j_{[001]}=I\chi\sin^2\theta, \tag{2}$$

where θ is the angle between the vector **e** and the [001] axis.

Excitation with unpolarized light produces no current along $[1\bar{1}0]$, and $j_{[001]}=I\chi/2$. Under conditions when the frequency ω of the exciting light exceeds the reciprocal carrier momentum relaxation time τ_{nk}^{-1}, the photocurrent is given by

$$\mathbf{j}=e\sum_{nn'\mathbf{kk}'}[\mathbf{v}_{n'}(\mathbf{k}')\tau_{n'\mathbf{k}'}-\mathbf{v}_n(\mathbf{k})\tau_{n\mathbf{k}}]W^{(as)}_{n'\mathbf{k}',n\mathbf{k}}. \tag{3}$$

Here $W_{n'\mathbf{k}',n\mathbf{k}}$ is the probability of an optical transition of the carriers from the state $(n,\mathbf{k})$ into $(n',\mathbf{k}')$; $v_n(\mathbf{k})$ is their group velocity; e is the elementary charge ($e<0$ for an electron and $e>0$ for a hole). A contribution to the current is made only by the component $W^{(as)}_{n'\mathbf{k}',n\mathbf{k}}$ that is odd with respect to the replacement of $(n'\mathbf{k}',n\mathbf{k})$ by $(\bar{n}',-\mathbf{k}',\bar{n},-\mathbf{k})$, where the states $(n,\mathbf{k})$ and $(\bar{n},-\mathbf{k})$ are connected by the time-reversal operation. As shown in Appendix 1, in the case of Boltzmann statistics, when the distribution function $f_{nk}\ll 1$, no asymmetry of the momentum distribution arises in the departure term, and after summation over all the final states the second term of (3) vanishes and

$$\mathbf{j}=e\sum_{n'\mathbf{k}'}\mathbf{v}_{n'}(\mathbf{k}')\tau_{n'\mathbf{k}'}g^{as}_{n'\mathbf{k}'}, \tag{4}$$

where

$$g^{as}_{n'\mathbf{k}'}=\sum_{n\mathbf{k}}W^{(as)}_{n'\mathbf{k}',n\mathbf{k}}.$$

For holes in the Γ_8 band of III-V crystals the indices n or n' in (3) and (4) number the subbands $l=1$ and 2 of the valence band ($l=1$ for heavy holes and $l=2$ for light ones) and the spin states in these subbands.

In the calculation of $W^{(as)}_{n'\mathbf{k}',n\mathbf{k}}$ one can use the usual perturbation-theory equations for the continuous spectrum. It is known that light absorption in intraband transitions (within the limits of one branch) can occur only if a phonon is simultaneously absorbed or emitted, or else in the case of scattering by impurities. In semiconductors with degenerate bands, direct transitions between the branches are allowed. Neither process contributes to $W^{(as)}$. Such a contribution appears in interbranch transitions accompanied by absorption or emission of a phonon. The effect is due to interference of two second-order matrix elements (type A transitions) or between matrix elements of first and third order. In the latter case the final state corresponds only either to absorption of a photon (type B transitions) or only to absorption (or emission) of a phonon (type C transitions).[1)] According to Ref. 16, $W^{(as)}\neq 0$ only under the additional condition that the total energies in the initial state and in one of the intermediate states are equal. As a result, the contribution to the PVE is made by transitions of nine types, as shown in Fig. 1. The energy and the momentum conservation laws and the condition that the energies be

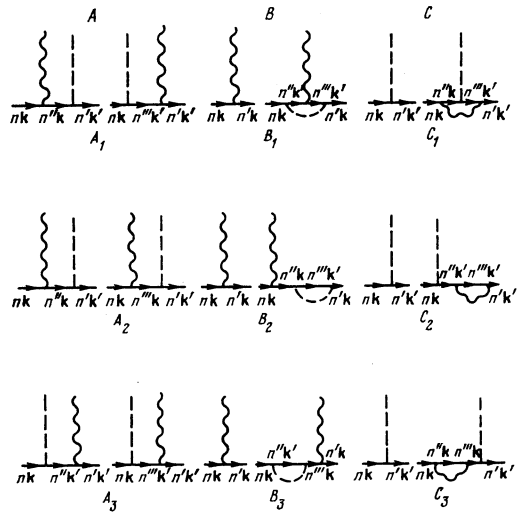

FIG. 1. Processes whose interference leads to a linear PVE in semiconductors with complex bands: solid line—hole (or electron), dashed—phonon, wavy—photon.

equal in the initial and in one of the intermediate states for each type of transition specify uniquely the initial ($\varepsilon_{l\mathbf{k}}$) and the final ($\varepsilon_{l'\mathbf{k}'}$) energies of the holes that participate in the transition, as well as the energies of the holes in the intermediate states (at definite l'', l'''). Therefore the only temperature-dependent quantities in the generation rate $g^{as}_{n\mathbf{k}}$ are the distribution function of the holes in the initial state

$$f(\varepsilon_{l\mathbf{k}})=\exp[(\mu-\varepsilon_{l\mathbf{k}})/k_BT]$$

and the phonon occupation numbers N_Ω. Table I lists the values of the initial and final energies of the holes for all possible types of transitions shown in Fig. 1. The corrugation of the valence band is neglected here, so that

$$\varepsilon_{l\mathbf{k}}=\hbar^2k^2/2m_l, \tag{5}$$

TABLE I.

Type of transitions	l	l''	l'''	l'	$\varepsilon_{l\mathbf{k}}$	$\varepsilon_{l'\mathbf{k}'}$	Absorption (+) and emission (−) of phonons	$g^{as}(T)$
A_1	1	2	1,2	1,2	E^*	$E^*+\hbar\omega\pm\hbar\Omega$	+	$N_\Omega\varphi(T)$
A_2	1	2	1	1,2	E^*	$E^*+\hbar\omega\pm\hbar\Omega$	−	$(N_\Omega+1)\varphi(T)$
A_1	1,2	1,2	1	2	$E^*\mp\hbar\Omega$	$E^*+\hbar\omega$	+	$(N_\Omega+1)\varphi(T)$
A_3	1,2	2	1	2	$E^*\mp\hbar\Omega$	$E^*+\hbar\omega$	−	$N_\Omega\varphi(T)$
B_1	1	1,2	1,2	2	E^*	$E^*+\hbar\omega$		$N_\Omega\varphi(T)$
B_2	1	1	1,2	2	E^*	$E^*+\hbar\omega$		$(N_\Omega+1)\varphi(T)$
B_3	1	1,2	2	−				
C_1	1	2	1,2	1,2	E^*	$E^*\pm\hbar\Omega$	+	$N_\Omega\varphi(T)$
C_3	1	2	2	1,2	E^*	$E^*\pm\hbar\Omega$	−	$(N_\Omega+1)\varphi(T)$
C_1	1,2	1,2	2	1	$E^*\mp\hbar\Omega$	E^*	+	$(N_\Omega+1)\varphi(T)$
C_2	1,2	2	2	1	$E^*\mp\hbar\Omega$	E^*	−	$N_\Omega\varphi(T)$

Note. Here $\varphi(T)=T^{-3/2}\exp(-E^*/k_BT)$. The states in which the energy coincides with the total energy of the initial and final states are underlined. (For the B_1 transitions this condition is satisfied either for the state l'' or for the state l'''). In the last column, allowance is made for the fact that $N_\Omega\exp(\hbar\Omega/k_BT)=N_\Omega+1$.

where m_1 and m_2 are the effective masses of the heavy and light holes, respectively.

The last column of Table I gives the temperature dependence of g^{as} for each type of transition following absorption and emission of a phonon (for the transitions A and C). Account is taken here of the fact that $\exp(\mu/k_BT) \sim pT^{-3/2}$, where p is the density of the free holes. Unless otherwise stipulated, we consider hereafter interaction of holes with LO phonons. For p-GaAs excited by a CO_2 laser, the energy $E^* = \hbar\omega m_2/(m_1 - m_2)$ is less than the energy $\hbar\Omega$ of the LO phonon, and in this case there are no transitions into or from a state with energy $E^* - \hbar\Omega$.

It is seen from Table I that for each of the transitions the $g^{as}(T)$ dependence is described by a single equation

$$g_{nk}^{as} \sim [aN_\Omega + b(N_\Omega+1)]\, T^{-3/2} p \exp(-E^*/k_BT). \quad (6)$$

It follows from (4) and (6) that the temperature dependence of $\chi(T)$ is given by

$$\chi(T) = pT^{-3/2} \exp(-E^*/k_BT) \sum_{l,i} [a_i^{(l)} N_\Omega + b_i^{(l)}(N_\Omega+1)]\, \tau_l(\varepsilon_i^{(l)}). \quad (7)$$

Here $\tau_l(\varepsilon)$ is the effective momentum relaxation time of the holes of branch l with energy ε. The index i numbers the different values of the energy $\varepsilon_l^{(i)}$ in the final states, as determined by Table I.

In Appendix 2 is analyzed the temperature dependence of the time $\tau_l(\varepsilon)$ where carrier scattering by LO phonons. Account is taken here of the fact that after the emission or absorption of several phonons the directional momentum of the carrier is completely lost. It follows from the foregoing estimates that the temperature dependence of the time $\tau_l(\varepsilon)$ practically coincides with the temperature dependence of the time of emission or absorption of one phonon by a hole, so that

$$\tau_l^{-1}(\varepsilon) \sim [N_\Omega + \beta_l(N_\Omega+1)], \quad (8)$$

where β_l is a positive coefficient. For hot holes with energy $\varepsilon_l \approx \hbar\omega \gg k_BT$ this coefficient is close to unity and for their contribution to χ we obtain from (7)

$$\chi(T) = p(k_BT/E^*)^{-3/2} \exp(-E^*/k_BT)\, [\bar{a}N_\Omega + \bar{b}(N_\Omega+1)]/(2N_\Omega+1). \quad (9)$$

For transitions into the state with energy $E^* < \hbar\Omega$ the coefficient $\beta_l = 0$. Since, however, this state can be reached only upon emission of the phonon (see Table I), in this case the coefficient $b^{(l)}$ in (6) is also equal to zero. Therefore, $\chi(T)$ for such transitions is also described by Eq. (9), with $\bar{a} = \bar{b}$. At $\varepsilon_l = E^* + \hbar\Omega$ the coefficient β_l in (8) differs noticeably from unity. However, the contribution of such states to the current (4) is relatively small, since the relaxation time and the group velocity in a state with energy $E^* + h\Omega$ are less than for hot holes, and this state is reached only via processes C_1. For the one-phonon mechanism of the linear PVE the dependence of the photocurrent on the temperature is therefore well described by the general equation (9).

If the predominant contributions to g^{as} and $\tau(\varepsilon)$ were to be made by interaction with acoustic phonons, then the $\chi(T)$ dependence would be determined also by Eq. (9), in which one must put $N_\Omega \gg 1$, i.e., in this case

$$\chi(T) \sim K_0(T) \sim pT^{-3/2} \exp(-E^*/k_BT), \quad (10)$$

where $K_0(T)$ is the coefficient of light absorption for direct optical transitions between the branches.

In the two-phonon mechanism, the contribution to χ is the consequence of interference of two processes with participation of two phonons. The transition can take place here from a state with any initial energy, and the main contribution to the current is made by hot holes with energy $\varepsilon \approx \hbar\omega \gg k_BT$, $\hbar\Omega$. Consequently, for this mechanism

$$g^{as}(T) \sim p(2N_\Omega+1)^2, \qquad \chi(T) \sim p(2N_\Omega+1) = p \operatorname{cth}(\hbar\Omega/2k_BT). \quad (11)$$

There is a qualitative difference between the temperature dependences of the functions (9) and (10). The function (9) first increases rapidly, reaches a maximum at $k_BT \sim E^*$ ($E^*/k_B = 300$ K at the CO_2-laser frequency) and then decreases slowly. The function (11) increases monotonically with increasing temperature (almost linearly starting with $T = 400$ K). Thus, measurement of the $\chi(T)$ dependence at high temperatures makes it possible to distinguish between the one- and two-phonon mechanisms.

3. ASYMMETRY OF ELEMENTARY PROCESSES IN p-TYPE III-V CRYSTALS

The cause of asymmetry in the transition probabilities $W_{n'k',nk}$ is the presence of terms of different parity in the operators for the interaction of the holes with phonons and with photons. The operator D_q, which describes the interaction of the holes of Γ_8 with optical phonons, is given by (Ref. 18, Sec. 32)

$$D_q = iC \frac{\mathbf{q}\mathbf{u}_q}{q^2} + \frac{d_0}{\sqrt{3}} |\delta_{\alpha\beta\gamma}| [J_\alpha J_\beta] u_{q\gamma}, \quad (12)$$

where $\mathbf{u}_q$ is the amplitude of the relative displacement of the two sublattices in optical oscillations,

$$C = e\Omega \left(\frac{4\pi\rho}{\varepsilon^*}\right)^{1/2}, \quad \frac{1}{\varepsilon^*} = \frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0},$$

ε_0 and ε_∞ are the static and high-frequency dielectric constants; ρ is the density and J_α are the matrices of the projection operator of the angular momentum in the basis

$$Y_m^{(3/2)}(m = \pm 3/2, \pm 1/2); \quad 2[J_\alpha J_\beta] = J_\alpha J_\beta + J_\beta J_\alpha.$$

In crystals with inversion centers, one of the constants in (12) vanishes: in a lattice of the NaCl type, $d_0 = 0$, i.e., there is no deformation interaction, while in a diamond-type lattice there is no long-range interaction, i.e., $c = 0$.

The eigenfunctions ψ_{nk} of the holes are determined by

the matrix $F_n(\mathbf{k})$:

$$\psi_{nk}=e^{i\mathbf{kr}}\sum_m F_{n,m}(\mathbf{k})\,Y_m(\mathbf{r}). \tag{13}$$

The matrix $\mathbf{F}_n$ diagonalizes the Hamiltonian $\mathcal{H}(\mathbf{k})$ of the holes. In the spherical approximation,

$$\mathcal{H}(\mathbf{k})=-[(A+{}^5/_4B)k^2-B(\mathbf{Jk})^2], \tag{14}$$
$$\hbar^2/2m_1=-(A-B),\quad \hbar^2/2m_2=-(A+B),\quad A<B<0.$$

In this basis, the matrix element $d^{(-)}_{n'\mathbf{k}',n\mathbf{k}}$ of a transition with absorption of an LO phonon $\mathbf{q}$ is of the form

$$d^{(-)}_{n',\mathbf{k}+\mathbf{q},n\mathbf{k}}=(\hbar/2\rho\Omega)^{1/2}(\mathbf{qe}^L_{\mathbf{q}}/q^2)\mathbf{F}_{n'}{}^+(\mathbf{k}+\mathbf{q})\,(iC+3^{-1/2}d_0|\delta_{\alpha\beta\gamma}|[J_\alpha J_\beta]q_\gamma)\mathbf{F}_n(\mathbf{k}), \tag{15}$$

where $\mathbf{e}^L_q=\mathbf{e}^L_{-q}$ is the LO-phonon polarization vector. For transitions with phonon emission

$$d^{(+)}_{n'\mathbf{k}',n\mathbf{k}}=(d^{(-)}_{n\mathbf{k},n'\mathbf{k}'})^*.$$

A contribution to $W^{(as)}$ is made also by a product of terms of different parity in the matrix element of the velocity operator $\hat{v}$, which determines the probability of the optical transitions:

$$v_{n'n}(\mathbf{k})=\hbar^{-1}\mathbf{F}_{n'}{}^+(\mathbf{k})\,(\nabla_{\mathbf{k}}\mathcal{H}(\mathbf{k}))\,\mathbf{F}_n(\mathbf{k}). \tag{16}$$

The operator $\mathcal{H}(\mathbf{k})$ includes, besides the principal term (14) that is quadratic in k, also a relativistic term linear in k

$$\mathcal{H}_k=\frac{4}{\sqrt{3}}k_0\sum_\alpha k_\alpha[J_\alpha(J^2_{\alpha+1}-J_{\alpha+2})] \tag{17}$$

and three terms cubic in k, the largest of which is nonrelativistic:

$$\mathcal{H}_{k^3}=\mathcal{D}\sum_\alpha J_\alpha k_\alpha(k^2_{\alpha+1}-k^2_{\alpha+2}). \tag{18}$$

The transition probability $W^{(as)}$ in (3) and (4) contains only terms that are odd in k and result from the product of the first and second terms in (15), or else from the product of the term linear in k and the term quadratic in k or independent of k in (16). Therefore a measure of the contribution due to the asymmetry of the scattering by optical phonons is the quantity

$$Z_1=d_0\bar{k}/C, \tag{19}$$

while the measures of the contributions connected with the asymmetry of the optical transitions on account of the terms (17) and (18) are respectively the quantities

$$Z_2=k_0/B\bar{k},\quad Z_3=\mathcal{D}\bar{k}/B, \tag{20}$$

where $\bar{k}\approx\bar{q}$ is the mean value of the wave vector of the holes with energy $\varepsilon_{1k}\approx\hbar\omega$. For GaAs at $\hbar\omega=0.12$ eV we have $Z_1=0.8\cdot10^{-2}(d_0a_0)$, where (d_0a_0) is the value of the corresponding constant of the deformation potential in (12) (in eV), and a_0 is the lattice constant.

The constant k_0 for different crystals lies in the range $(1-6)\cdot10^{-10}$ eV (see Refs. 19 and 20), in agreement with the estimates of k_0 obtained for GaAs, from the values of the hole spin-relaxation time in strongly deformed crystals.[21] Corresponding to these values of k_0 are $Z_2=(0.6-4)\cdot10^{-3}$. The constant $\mathcal{D}$ in (18) can be estimated from the splitting, cubic in k, of the conduction band, which is given by an equation similar to (18)

$$\mathcal{H}_{c,k^3}=\frac{1}{2}\alpha_c(2m_c{}^3E_g)^{-1/2}\hbar^3\sum_\alpha\sigma_\alpha k_\alpha(k^2_{\alpha+1}-k^2_{\alpha+2}), \tag{21}$$

where m_c is the effective mass of the electron, E_g is the width of the forbidden band, and σ_α are Pauli matrices.

The main contribution to (18) and (20) is made by a product of terms linear and quadratic in k in the interband matric elements [see Ref. 18, Eq. (26.37)]

$$\mathcal{H}_{cv}=\frac{\hbar^2}{m_{cv}}\sum_\alpha\mathcal{P}_\alpha k_{\alpha+1}k_{\alpha+2}.$$

When account is taken of only this contribution

$$\mathcal{D}=\alpha_v(2m_c{}^3E_g)^{-1/2}\hbar^3,$$
$$\alpha_v=-\frac{\alpha_c}{2}\frac{E_g+\Delta}{\Delta}=-2\frac{m_c}{m_{cv}}\left(\frac{E_g+\Delta}{3E_g+2\Delta}\right)^{1/2}. \tag{22}$$

Here Δ is the spin-orbit splitting of the valence band.

According to Ref. 22, the value of $|\alpha_c|$ determined from the electron spin-relaxation time in GaAs is $2.2\cdot10^{-2}$. It follows therefore that $|\alpha|=5.8\cdot10^{-2}$ and $m_{cv}=0.8\,m_0$. At this value of α_v we have $Z_3=4.7\times10^{-2}$. These estimates show that the contribution made to $W^{(as)}$ by the asymmetry of the photon processes on account of the $\mathcal{H}$ terms cubic in k is larger by at least one order of magnitude than the contribution due to the terms linear in k. The asymmetry of the phonon processes, which is determined by the value of Z_1, prevails over the photon asymmetry at $d_0a_0>8$ eV. We do not know the value of d_0 for GaAs. (We note that for Ge the corresponding constant d_0a_0 is 42 eV according to Refs. 23 and 24.)

A linear photovoltaic effect can arise also in the case of scattering by acoustic phonons as a result of interference between piezoelectric and deformation scatterings. According to estimates, at $\omega>\Omega$ or $k_BT>\hbar\Omega$ the contribution of the acoustic phonons is approximately one-thirtieth of that of the optical phonons.

We now estimate the constant χ. We take into account for this purpose only transitions of type A_1 from the state $\mathbf{1k}$ into $\mathbf{2k'}$. In this case, according to (2), (3), (15), and (16)

$$\chi^{\pm}=\frac{4\pi^3e^3}{\hbar\omega^2cn}|\delta_{\alpha\beta\gamma}|\sum_{\mathbf{kk}'}v_{2'}{}^z(\mathbf{k}')\tau_{2\mathbf{k}'}f_{1\mathbf{k}}\left(N_\Omega+\frac{1}{2}\pm\frac{1}{2}\right)$$
$$\times\,\mathrm{Im}[d^{(\pm)}_{2\mathbf{k}',2\mathbf{k}}v_{21}{}^\alpha(\mathbf{k})\,d^{(\mp)}_{1\mathbf{k},1\mathbf{k}'}v_{12}{}^\beta(\mathbf{k}')]\left[\frac{\delta(\varepsilon_{2\mathbf{k}}-\varepsilon_{1\mathbf{k}'}-\hbar\omega)}{\varepsilon_{2\mathbf{k}}-\varepsilon_{1\mathbf{k}}-\hbar\omega}\right.$$
$$\left.+\frac{\delta(\varepsilon_{2\mathbf{k}}-\varepsilon_{1\mathbf{k}}-\hbar\omega)}{\varepsilon_{2\mathbf{k}'}-\varepsilon_{1\mathbf{k}'}-\hbar\omega}\right]\delta(\varepsilon_{2\mathbf{k}'}-\varepsilon_{1\mathbf{k}}-\hbar\omega\pm\hbar\Omega). \tag{23}$$

The upper and lower signs in (23) correspond to phonon emission and absorption, respectively. The δ function

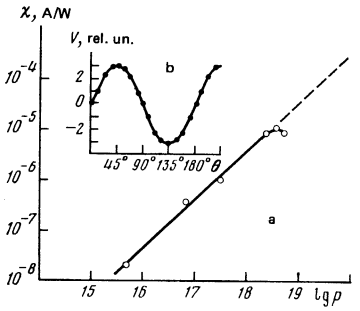

FIG. 2. a) Dependence of the PVE constant χ on the hole density. $T = 300$ K, $\lambda = 10.6$ μm; b) dependence of the PVE signal in a transverse geometry on the angle θ.

in the square brackets corresponds to the condition that the energies in the initial (final) state and in one of the intermediate states be equal. An estimate in accordance with (23) at τ_{2k} determined from (A2.5) yields at $m_1 \gg m_2$, $\omega \gg \Omega$, and $N_\Omega \sim 1$

$$\chi = Z \frac{e^3\hbar}{cn\omega} \frac{p \exp(-E^*/k_BT)}{m_1 m_2^{1/2} (k_BT)^{3/2}}, \tag{24}$$

where Z is one of the parameters Z_{1-3} defined by the equations (19), (20).[2)]

In the considered case A_1, just as in B_1 and C_1, when both optical transitions are interband, one of the intermediate states can be located in other bands, say in the conduction band. In this case the optical-transition matrix element $v_{cv} = p_{cv}/m_0 \approx (E_g/m_2)^{1/2}$ is larger than for the interband transition, but the energy denominator in the composite matrix element is equal to E_g in place of $\hbar\Omega$ in (23). Therefore the contribution of the transitions through the conduction band is small compared with (24) in the ratio $\hbar\Omega/(\hbar\omega E_g)^{1/2}$.

4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

The PVE was investigated for p-GaAs samples with impurity (Zn) density from $5 \cdot 10^{17}$ cm^{-3} to $4 \cdot 10^{18}$ cm^{-3}. The radiation source was a Q-switched CO_2 laser. The pulse duration is 150 nsec, the pulse radiation power was 6 kW, and the radiation wavelengths were 10.6 and 9.6 μm. The samples were of narrow plates cut so that the radiation was incident in the [110] direction. On the end faces of the samples, along the [1$\bar{1}$0] directions, were two ohmic contacts. The contacts were prepared by fusing-in indium containg 2.5% nickel and 1.5% zinc in a hydrogen atmosphere at a temperature 600 °C.

As seen from Fig. 2, when the plane of polarization was rotated the emf on the contacts varied in accordance with (2) like

$$V = D \frac{P_{in}}{h} \sin 2\theta, \tag{25}$$

where P_{in} is the radiation power incident on the sample; h is the width of the sample. The signal picked off the sample duplicated exactly the shape of the laser pulse and had a linear dependence on the light intensity.

The investigations of the PVE were carried out also at other sample orientations, in both longitudinal and transverse geometry. We have performed a number of experiments that provided an approximate check on the PVE behavior that follows from the phenomenological expression (1). Thus, at the transverse experimental geometry indicated above, rotation of the sample around the [110] axis by 180° at a constant direction of the light propagation leads to a change in the polarity of the signal. The reason is that in such a rotation the angle between the polarization vector $\mathbf{e}$ and the [001] axis changes from θ to $\pi - \theta$, which causes in accordance with (2) and (25) a reversal of the sign of the PVE.

In a longitudinal geometry, when the light propagates along [100] and the emf of the PVE is registered in the same direction, a linear PVE and the dragging effect are observed. According to (1), the emf of the linear PVE varies in this case like $\sin 2\theta$, where θ is the angle between the vector $\mathbf{e}$ and the [001] axis, while the dragging emf does not depend on the polarization. When the sample is rotated 180° around the [001] axis, the sign of the dragging effect is reversed, and the linear PVE remains unchanged, so that the angle θ is not changed by this rotation.

In all the experimental geometries, reflection, with the aid of a mirror, of the radiation passing through the sample increased the signal of the linear PVE and decreased the dragging signal.

In transverse geometry, the quantity D in (25), which is determined from experiment, is connected with the constant χ in (1) and (2) by the relation

$$\chi = \frac{P_{in}}{\bar{P}_{in}} \frac{D}{R_0} \frac{l}{dh}, \tag{26}$$

where d is the sample thickness, l is the distance between the electrodes, R_0 is the sample resistance, and $\bar{P}_{in}$ is the average power of the radiation in the sample.

With allowance for the absorption and reflection of the light, the ratio of $\bar{P}_{in}$ to the incident flux P_{in} is

$$\frac{\bar{P}_{in}}{P_{in}} = \frac{(1-R)(1-e^{-\alpha d})}{\alpha d (1 - Re^{-\alpha d})}, \tag{27}$$

where α is the absorption coefficient and R is the reflection coefficient. For the dragging effect, which depends on the light propagation direction, the minus sign preceding the exponential in the denominator is replaced by a plus.

As already noted in Ref. 17, at room temperature the constant D, with allowance for the absorption and reflection of the radiation, is practically independent of the degree of doping, and consequently χ, as seen from Fig. 2, increases linearly with increasing hole density up to $p = 4 \cdot 10^{18}$ cm^{-3}. This shows that at room temperature the linear PVE in p-GaAs is connected with the absorption of light by the free carriers and is caused by the phonon mechanism. If the effect were connected with carrier scattering by impurity centers, then the photocurrent, meaning the constant χ, would

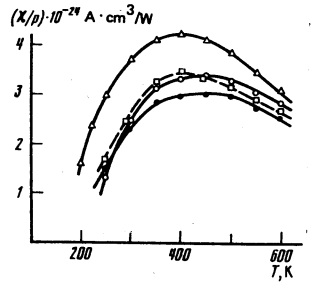

FIG. 3. Dependence of the constant χ/p on the temperature ($\lambda = 10.6$ μm): Δ—$p = 7.4 \cdot 10^{16}$ cm^{-3}, O—$p = 3.4 \cdot 10^{17}$ cm^{-3}; ●—$p = 2.36 \cdot 10^{18}$ cm^{-3}; □—$p - 4 \cdot 10^{18}$ cm^{-3}.

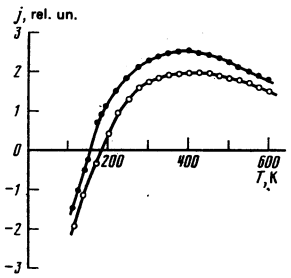

FIG. 5. Temperature dependence of the photovoltaic current: $p = 7.4 \cdot 10^{16}$ cm^{-3}, ●) $\lambda = 10.6$ μm; O) $\lambda = 9.5$ μm.

depend quadratically on p, inasmuch as at such densities ($\lesssim 10^{18}$ cm^{-3}) and temperatures the hole mobility depends weakly on p. The deviation of $\chi(p)$ from linearity at $p > 4 \cdot 10^{18}$ cm^{-3} is apparently due to the contribution made by the scattering of the photoexcited carriers from the impurities.

To clarify the role of one- and two-phonon processes, as well as the contribution of the optical and acoustic phonons, we investigated the temperature dependence of the PVE current in a wide range of temperatures. Inasmuch as at low temperatures, as shown in Ref. 17, the PVE current is due to transitions from the impurity, the investigation was carried out in the temperature range from 100 to 600 K. In samples with different impurity density, as seen from Fig. 3, the value of χ for all samples increases with increasing T, reaching a maximum in the region $T = 400$–450 K, after which it decreases slowly. The ratio χ/p in the temperature range 300–600 K changes in this case by not more than 1.3 times when the density changes by almost two orders of magnitude. Then the temperature drops below 300 K, χ/p decreases rapidly, goes through zero, and reverses sign. The temperature corresponding to the reversal of the sign decreases with decreasing hole density. In a sample with $p = 5.2 \cdot 10^{15}$ cm^{-3} the phonon mechanism predominates in the temperature range $T \gtrsim 250$ K, where the carrier density increases noticeably with increasing T on account of the loss of the deep impurities. As seen from Fig. 4, in this sample χ increases with increasing T in the entire investigated interval, whereas the ratio χ/p, as in the other samples, goes through a maximum at $T = 400$ K.

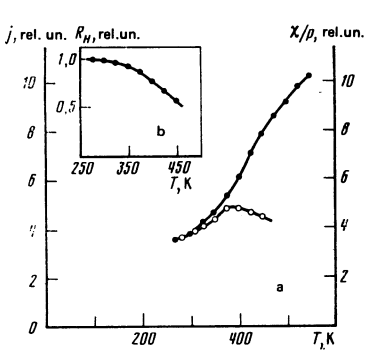

FIG. 4. a) Temperature dependence of the experimentally measured photocurrent (●) and the value of χ/p (O) for a sample with hole density p (300 K) = $5.2 \cdot 10^{15}$ cm^{-3}; b) temperature dependence of the Hall coefficient.

The photocurrent changes somewhat with changing frequency of the exciting light but, as seen from Fig. 5, the temperature dependence of χ/p is the same at $\lambda = 10.6$ as at $\lambda = 9.5$ μm.

The indicated temperature dependence of $\chi(T)/p$ agrees with that expected for the one-phonon mechanism in accord with Eq. (9). In Fig. 6 are compared the experimental and theoretical plots at $\chi(T)/p(T)$. The sample chosen for the comparison was the one with $p = 7.4 \cdot 10^{16}$ cm^{-3}. The best agreement between theory and experiment takes place at $a = 3.4 \cdot 10^{-24}$ A·cm^{-3}/W and $\bar{b} = 0$.

When two-phonon processes predominate, χ/p should increase monotonically with temperature in accordance with (11), but this does not agree with the experimental relation.

We note that in n-GaAs, where there is no one-phonon contribution from transitions between branches, a monotonic increase of the photocurrent with increasing T is observed at a constant electron density.

We compare now the observed value of χ with the estimate (24). According to (24), $\chi_{teor} = 6 \cdot 10^{-3} Z$ (A/W) at $p = 7.4 \cdot 10^{16}$ cm^{-3}. The experimental value of χ at this density is $\chi_e = 5 \cdot 10^{-7}$ A/W, which agrees with the theoretical estimate at $Z \approx 10^{-2}$. This value is close to the estimates of Sec. 3. Since Eq. (24) is only a rough estimate, it is impossible to conclude unequivocally from this value of Z which of the components, Z_1 or Z_2 predominates in (19) and (20). According to (23), χ should decrease with increasing like

$$\omega^{-s} \exp\left(-\frac{\hbar\omega}{k_B T} \frac{m_2}{m_1 - m_2}\right),$$

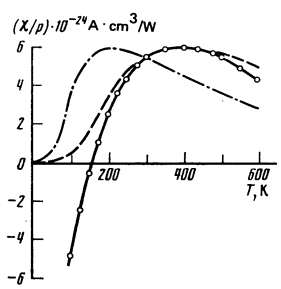

FIG. 6. Temperature dependence of χ/p for the sample with $p = 7.4 \cdot 10^{16}$ cm^{-2}, $\lambda = 10.6$ μm; points—experiment, dash-dot—theory (scattering of holes by acoustic phonons); dashed—theory (scattering of holes by optical phonons).

where $s = 1-2$. Indeed, as seen from Fig. 5, the variation of χ with frequency agrees with this relation.

Thus, our investigations show that in p-GaAs with impurity density $10^{15}-10^{19}$ cm^{-3} at $T > 200$ K, excited with light from a CO_3 laser, the predominant mechanism of the linear PVE constitutes transitions between the branches of the heavy and light holes of the valence band, with simultaneous emission or absorption of a longitudinal optical phonon. It appears here that the principal role is played by the asymmetry of the interaction of the electron with the optical phonon.

APPENDIX I

We shall prove that in the case of Boltzmann statistics, i.e., at $f_{n\mathbf{k}} \ll 1$, the departure of the electrons does not produce an asymmetry in the momentum distribution. We use for this purpose the diagram technique for nonequilibrium systems.[25,26] Assume that a plane linearly polarized light wave propagates in the crystal. We introduce nonequilibrium time-averaged Green's functions[15]:

$$G(\tau) = \lim_{T\to\infty} \frac{1}{2T} \int_{-T}^{T} dt\, G(t+\tau, t). \tag{A1.1}$$

The Fourier components of the causal (G^{--}) and anticausal (G^{++}) Green's functions satisfy the relation

$$G^{++}_{n\mathbf{k}\omega} = -(G^{--}_{n\mathbf{k}\omega})^*. \tag{A1.2}$$

At $f_{n\mathbf{k}} \ll 1$, contributions to $G^{--}_{n\mathbf{k}\omega}$ (or $G^{++}_{n\mathbf{k}\omega}$) are made by diagrams whose vertices are located on the upper (or lower) part of the Keldysh contour. In this case, these functions are expressed only in terms of one self-energy function ($\Sigma^{--}_{n\omega}$ or $\Sigma^{++}_{n\mathbf{k}\omega}$, respectively):

$$\begin{aligned} (G^{--}_{n\mathbf{k}\omega})^{-1} &= (G^{(0)--}_{n\mathbf{k}\omega})^{-1} + \Sigma^{--}_{n\mathbf{k}\omega}, \\ (G^{++}_{n\mathbf{k}\omega})^{-1} &= (G^{(0)++}_{n\mathbf{k}\omega})^{-1} + \Sigma^{++}_{n\mathbf{k}\omega}, \end{aligned} \tag{A1.3}$$

where the index 0 labels the equilibrium Green's functions. Taking (A1.2) and (A1.3) into account, we can express the contribution of the optical transitions to the damping of an electron whose state is $(n, \mathbf{k})$ in the form

$$\gamma^{(\mathrm{rad})}_{n\mathbf{k}\omega} = \operatorname{Im}\Sigma^{--}_{n\mathbf{k}\omega} = \operatorname{Im}\Sigma^{++}_{n\mathbf{k}\omega} = {}^1\!/_2 \operatorname{Im}(\Sigma^{--}_{n\mathbf{k}\omega} + \Sigma^{++}_{n\mathbf{k}\omega}). \tag{A1.4}$$

At $f_{n\mathbf{k}} \ll 1$ any diagram for $\Sigma^{++}_{n\mathbf{k}\omega}$ can be obtained from the corresponding (mirror-symmetry) diagram for $\Sigma^{--}_{n\mathbf{k}\omega}$ by simple replacement of the causal Green's functions by anticausal ones. Taking into account the diagram-technique rules, according to which a vertex on the upper or lower part of the contour is set in correspondence with a matrix element and a factor $\mp ih$, it is easy to show that the contribution of two mirror-symmetrical diagrams for $\Sigma^{--}_{n\mathbf{k}\omega}$ and $\Sigma^{++}_{n\mathbf{k}\omega}$ to $\gamma^{(\mathrm{rad})}_{n\mathbf{k}}$ does not contain the imaginary part of the product of matrix elements corresponding to the vertices of the diagram. We designate this product by $\Pi(n\mathbf{k}, \{\mathbf{k}_r\})$, where $\{\mathbf{k}_r\}$ is the aggregate of the wave vectors of the electron in the intermediate states (the spin indices in these stages are left out for brevity). Owing to the symmetry of the system to time reversal (the light is linearly polarized and the crystal is nonmagnetic), the energy spectrum $\varepsilon_{n\mathbf{k}}$ and the quantities $\operatorname{Re}\Pi(n\mathbf{k}, \{\mathbf{k}_r\})$ satisfy the relations (see, e.g., Ref. 18, Sec. 18)

$$\varepsilon_{n\mathbf{k}} = \varepsilon_{\bar{n},-\mathbf{k}}, \quad \operatorname{Re}\Pi(n\mathbf{k}, \{\mathbf{k}_r\}) = \operatorname{Re}\Pi(\bar{n}, -\mathbf{k}, \{-\mathbf{k}_r\}), \tag{A1.5}$$

where the states $(n, \mathbf{k})$ and $(\bar{n}, -\mathbf{k})$ are connected by the time-reversal operation. Since the damping $\gamma^{(\mathrm{rad})}_{n\mathbf{k}\omega}$ is connected precisely with these quantities, it follows that if the filling of the electronic states is neglected

$$\gamma^{(\mathrm{rad})}_{n\mathbf{k}} = \sum_{n'\mathbf{k}'} W_{n'\mathbf{k}',n\mathbf{k}} = \gamma^{(\mathrm{rad})}_{\bar{n},-\mathbf{k}} = \sum_{n'\mathbf{k}'} W_{n'\mathbf{k}',\bar{n},-\mathbf{k}}. \tag{A1.6}$$

At $f_{n\mathbf{k}}$ comparable with unity, relation (A1.6) does not hold. For example, in the phonon interband mechanism of the PVE (Ref. 14) the contribution made to the photocurrent by the departure of electrons from the valence band differs from zero.

APPENDIX 2

Let us estimate the effective momentum relaxation time for photoexcited electrons. We consider for simplicity a degenerate spherical parabolic band. Assume that at the energy level ε there are generated electrons with average velocity $\mathbf{v}_0$ at the instant of production. These photoelectrons interact with the LO phonons, so that the probability of a $\mathbf{k} \to \mathbf{k}'$ transition with emission or absorption of the phonon takes in accordance with (12) the form

$$W^{(\pm)}_{\mathbf{k}'\mathbf{k}} = \frac{\pi}{\rho\Omega} \frac{C^2}{|\mathbf{k}'-\mathbf{k}|^2} \left(N_\Omega + \frac{1}{2} \pm \frac{1}{2}\right) \delta(\varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}} \pm \hbar\Omega). \tag{A2.1}$$

The ratio of the average velocity per particle for electrons going over to level $\varepsilon_{\pm 1} = \varepsilon \pm \hbar\Omega$, and the velocity v_0 is

$$\varkappa_{\pm 1,0} = \left(\frac{\varepsilon_{\pm 1}}{\varepsilon}\right)^{1/2} \left[1 + \frac{(k-k_\pm)^2}{2kk_\pm} - \ln^{-1}\left|\frac{k+k_\pm}{k-k_\pm}\right|\right], \tag{A2.2}$$

where

$$k = (2m_c\varepsilon/\hbar^2)^{1/2}, \quad k_\pm = (2m_c\varepsilon_{\pm 1}/\hbar^2)^{1/2}.$$

It is seen that the quantity $\varkappa$ is not very small, and that if scattering of carriers by LO phonons predominates it is necessary, generally speaking, to sum the contributions made to j_{PV} from electrons on the energy levels ε, $\varepsilon \pm \hbar\Omega$, $\varepsilon \pm 2\hbar\Omega$, etc, including also the returns. At not too large values of ε, i.e., at ε on the order of several $\hbar\omega$, it suffices to take into account only one or two steps. If the expression for j_{PV} is represented in the form

$$j_{\mathrm{PV}} = eG_0 v_0 \tau(\varepsilon), \tag{A2.3}$$

where G_0 is the rate of electron generation on the level ε, then, taking two steps into account, we obtain for the effective momentum relaxation time $\tau(\varepsilon)$

$$\frac{\tau(\varepsilon)}{\tau_0(\varepsilon)}=1+\varkappa_{1,0}\frac{\tau_1}{\tau_{1,0}}\left(1+\varkappa_{0,1}\frac{\tau_0}{\tau_{0,1}}+\varkappa_{2,1}\frac{\tau_2}{\tau_{2,1}}\right)$$
$$+\varkappa_{-1,0}\frac{\tau_{-1}}{\tau_{-1,0}}\left(1+\varkappa_{0,-1}\frac{\tau_0}{\tau_{0,-1}}+\varkappa_{-2,-1}\frac{\tau_{-2}}{\tau_{-2,-1}}\right), \qquad (A2.4)$$

where $\tau^{-1}_{n\pm1,n}$ is the probability of the transition of an electron with energy $\varepsilon_n=\varepsilon+n\hbar\Omega$ to the level $\varepsilon_{n\pm1}=\varepsilon+(n\pm1)\hbar\Omega$:

$$\frac{1}{\tau_{n\pm1\,n}}=\frac{n_e e^2\Omega}{\varepsilon^*\hbar^2}\left(N_a+\frac{1}{2}\pm\frac{1}{2}\right)\frac{1}{k_n}\ln\left|\frac{k_n+k_{n\pm1}}{k_n-k_{n\pm1}}\right|, \qquad (A2.5)$$

τ_n is the lifetime of the level ε_n:

$$\tau_n^{-1}=\tau^{-1}_{n+1,n}+\tau^{-1}_{n-1,n}. \qquad (A2.6)$$

At $\varepsilon_{n-1}<0$, the quantity $\tau^{-1}_{n-1,n}$ must be set equal to zero. Equation (A2.4) is easily generated to the case of two branches of light and heavy holes in p-GaAs. In this case the quantities τ, τ_n, and $\tau_{n\pm1,n}$ depend also on the subband indices $l=1$, and 2.

Estimates show that although the ratio $(\tau-\tau_0)/\tau_0$ is comparable with unity, in the region $T=(300-600)$ K the temperature dependence of the ratio τ/τ_0 is very weak and this dependence can be neglected compared with the temperature dependence of the time τ_0.

[1)]We note that the contribution of the C-type processes was not taken into account in Ref. 16. Allowance for these processes leads to a reversal of the sign of the product $A_1\tau_{p1}$ in Eq. (6) of that reference. The estimate given in Ref. 16 for the coefficient χ_{max} remains the same if the coefficient D is increased by three times.

[2)]An exact calculation with allowance for all the possible transitions is extremely cumbersome and is the subject of a separate study.

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