

ON THE THEORY OF THE DRAG EFFECT UPON NONLINEAR LIGHT ABSORPTION IN SEMICONDUCTORS WITH COMPLICATED VALENCE BAND

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The photon drag effect (PDE) of current carriers upon a nonlinear absorption of polarized light in semiconductors with a complex band structure consisting of two closely located or two degenerate branches has been studied theoretically. The components of the drag current, provided two-photon light absorption, have been analyzed phenomenologically. A contribution of the Rabi effect for one-photon optical transitions, caused by the saturation of the final states of photoexcited holes, to the drag current has been taken into account in quantum-mechanical calculations. Theoretical results are compared with experimental data concerning the PDE in *p*-Ge.

transition probability was supposed to depend on the photon momentum not only in accordance with the conservation laws of energy and momentum but also through the dependence of the matrix element square on \vec{q} . In this work, we have demonstrated that the account of this contribution is essential when considering transitions between the branches of degenerate bands.

The PDE in semiconductors is connected with the transfer of a photon momentum to current carriers. The photocurrent, which arises at that, is described by the phenomenological formula

$$j_{\alpha} = I \zeta_{\alpha\beta\gamma\mu} e_{\beta} e_{\gamma} q_{\mu} \quad (1)$$

where \vec{e} is the vector of polarization, \vec{q} the wave vector, I the intensity of light, \vec{j} the density of the DPE current, and $\zeta_{\alpha\beta\gamma\mu}$ the PDE tensor ($\alpha, \beta, \gamma, \mu = x, y, z$).¹

The photon momentum is transferred to a system of current carriers not only at the one-photon absorption of light but at the multiphoton absorption as well, with the photocurrent depending on the degree of light polarization [1]. In work [1], the PDE was considered in the range of small light intensities, where perturbation theory is valid. Below, we will consider the case of an arbitrary radiation intensity.

We note that the photon momentum transfer can induce an ordered motion of current carriers either along or across the crystal surface depending on its orientation with respect to the specimen's crystallographic axes. In this work, we have calculated only the longitudinal component of the drag current in semiconductors of the *p*-Ge type. The optical

The PDE is caused by the photon momentum transfer to electrons and holes at the event of light absorption by them and by the subsequent relaxation of their ordered motion owing to the processes of their scattering by the nonhomogeneities of the crystal structure. We note that the PDE possesses many interesting properties. The drag current can change its sign depending on the light frequency. Such an opportunity is explained by the fact that the light momentum is transferred to the "crystal + electrons" or "holes + phonons" system. The distribution of the photon-transferred momentum (and its sign) between the neutral and charged subsystems of the crystal depends on the light frequency.

In a number of cases, the change of the geometry of experiment results in a current, which is produced by either the linear or the circular photo-galvanic effect (PGE)² and is quadratic in the intensity of light. The physical nature of this current has not been investigated enough until now [1–3]. Our report is devoted to this issue.

To analyze the symmetry properties of the effect, we rewrite Eq. (1) taking into account that the symmetrized, $(e_{\beta} e_{\gamma}^* + e_{\gamma} e_{\beta}^*)/2$, and antisymmetrized, $(e_{\beta} e_{\gamma}^* - e_{\gamma} e_{\beta}^*)/2$, products are transformed by

¹In the case where the intensity of radiation varies in time, a non-stationary current proportional to the value of $\frac{\partial \vec{P}}{\partial t}$, where \vec{P} is the specimen polarization vector under the action of an external electric field, emerges in the medium due to the effect of optical rectification.

²see, e.g., [3].

independent representations. Then, we have

$$\begin{aligned}
 j_{\alpha}^{(2)} = & I^2 \left(D_{\alpha\beta\gamma\mu\nu\lambda} \frac{e_{\beta}e_{\gamma}^{*} + e_{\beta}^{*}e_{\gamma}}{2} \frac{e_{\mu}e_{\nu}^{*} + e_{\mu}^{*}e_{\nu}}{2} + \right. \\
 & + iF_{\alpha\beta\gamma\lambda\mu} \times (\vec{e} \times \vec{e}^{*})_{\mu} \frac{e_{\beta}e_{\gamma}^{*} + e_{\beta}^{*}e_{\gamma}}{2} + \\
 & \left. + G_{\alpha\beta\gamma\lambda} (\vec{e} \times \vec{e}^{*})_{\beta} (\vec{e} \times \vec{e}^{*})_{\gamma} \right) q_{\lambda}. \quad (2)
 \end{aligned}$$

Since the values of the quantities $[e_m e_v^*]$ and $i(\vec{e}^* \times \vec{e})$ and of the stationary current $\vec{j}^{(2)}$ are real, the same is true for the tensors $D_{\alpha\beta\gamma\mu\nu\lambda}$, $F_{\alpha\beta\gamma\mu\lambda}$, and $G_{\alpha\beta\gamma\lambda}$ in Eq. (2). The tensor of the fifth rank $F_{\alpha\beta\gamma\mu\lambda}$ has non-zero components in crystals without the center of symmetry. Tensors $D_{\alpha\beta\gamma\mu\nu\lambda}$ and $G_{\alpha\beta\gamma\lambda}$ are non-zero in crystals of arbitrary symmetry, including those without the center of inversion.

The photocurrent, which is connected with the third term in Eq. (2), differs from zero for the elliptically polarized light only and does not emerge at illuminating the specimen with the linearly polarized or non-polarized light. Therefore, it is convenient to call the effect, described by $G_{\alpha\beta\gamma\lambda}$, the two-photon circular drag effect. The effect, described by the tensor $D_{\alpha\beta\gamma\mu\nu\lambda}$, is usually observed provided a linearly polarized exciting radiation, so that it is convenient to call it the two-photon linear drag effect.

The photocurrent, which corresponds to the second term in Eq. (2), differs from zero if the specimen is simultaneously illuminated with elliptically and linearly polarized light. It is this contribution to the total photocurrent that distinguishes two-photon polarization photogalvanic effects from one-photon ones.

Taking into account the symmetry properties of the tensors $D_{\alpha\beta\gamma\mu\nu\lambda}$, $F_{\alpha\beta\gamma\mu\lambda}$, and $G_{\alpha\beta\gamma\lambda}$ with respect to the operation of time inversion, we can analyze whether the effects described by them are connected to the processes of dissipation. For example, the linear PDE is connected, while the circular one is not (see the table). Drawing this conclusion, we took into account that the current \vec{j} , wave vector of a photon \vec{q} , and quantity $i(\vec{e}^* \times \vec{e})$ do not change their signs.

1. Quantum-mechanical Description

The expression for the density of the N -photon drag current $\vec{j}^{(N)}$ in semiconductors with a cubic symmetry and a hole conductivity can be presented in the

framework of the electron representation and in the approximation of the relaxation time as follows [1]:

$$\begin{aligned}
 \vec{j}^{(N)} = & -e \sum_{\vec{k} m m'} \left[\vec{v}_{1\vec{k}} \tau_{\vec{p}}(1, \vec{k}) W_{1m'\vec{k}; 2m, \vec{k}-N\vec{q}}^{(1)} - \right. \\
 & \left. - \vec{v}_{2\vec{k}} \tau_{\vec{p}}(2, \vec{k}) W_{1m', \vec{k}+N\vec{q}; 2m\vec{k}}^{(1)} \right], \quad (3)
 \end{aligned}$$

$$\begin{aligned}
 W_{1m'\vec{k}; 2m, \vec{k}-N\vec{q}}^{(1)} = & \frac{2\pi}{\hbar} \left| M_{1m'\vec{k}; 2m, \vec{k}-N\vec{q}}^{(N)} \right|^2 \times \\
 & \times \left(f_{2, \vec{k}-N\vec{q}}^{(e)} - f_{1\vec{k}}^{(e)} \right) \delta \left(E_{\vec{k}} - E_{2, \vec{k}-N\vec{q}} - N \hbar \omega \right), \quad (4)
 \end{aligned}$$

$$\begin{aligned}
 W_{1m', \vec{k}+N\vec{q}; 2m\vec{k}}^{(1)} = & \frac{2\pi}{\hbar} \left| M_{1m', \vec{k}+N\vec{q}; 2m\vec{k}}^{(N)} \right|^2 \times \\
 & \times \left(f_{2\vec{k}}^{(e)} - f_{1, \vec{k}+N\vec{q}}^{(e)} \right) \delta \left(E_{1, \vec{k}+N\vec{q}} - E_{2\vec{k}} - N \hbar \omega \right), \quad (5)
 \end{aligned}$$

$$f_{l\vec{k}}^{(e)} = 1 - e^{(\chi + E_{l\vec{k}})/k_{\text{B}}T}. \quad (6)$$

Here, $N = 1$ or 2 , $e > 0$, $E_{l\vec{k}} = -\frac{\hbar^2 k^2}{2m_l}$ is the energy spectrum, $\vec{v}_{l\vec{k}} = -\frac{\hbar \vec{k}}{2m_l}$ the velocity, $\tau_p(l, \vec{k})$ the momentum relaxation time of electrons of the l -th branch, l the index of the heavy ($l = 1$) or light ($l = 2$) hole subzone, m' and m the indices of the degenerate states of heavy ($m' = \pm 3/2$) and light ($m = \pm 1/2$) holes or, to put it more precisely, of the electron states in the corresponding subzones; $M_{1m'\vec{k}; 2m\vec{k}}^{(N)}$ the matrix element of the direct N -photon transition, and χ the chemical potential of holes.

According to work [1], if the intensity of light with the wavelength $\lambda = 90.55 \mu\text{m}$ increases, the longitudinal photon drag current, $\vec{j} \parallel \vec{q}$, changes its sign, notwithstanding the light polarization, linear or circular. Following [1], it can be explained if one assumes different signs of the one- and N -photon contributions to the total drag current and takes into account that the role of multiphoton contributions grows with the intensity. As was shown in work [1], the main contribution to the drag current at room temperature and $\lambda = 90.55 \mu\text{m}$ was given by direct optical transitions, in spite of the fact that the light absorption coefficient was governed by indirect transitions under those experimental conditions.

Tensor	Name of effect	Time inversion parity	Availability in crystals with the center of inversion
$D_{\alpha\beta\gamma\mu\nu\lambda}$	Two-photon linear photon drag effect	+	Yes
$G_{\alpha\beta\gamma\lambda}$	Two-photon circular photon drag effect	-	Yes

In this work, we will not concentrate our attention at the details of calculations of the two-photon current $j_{\text{circ}}^{(2)}$ excited by the circularly polarized light, because, when calculating it, we used the procedure described in detail earlier in work [1]. Therefore, only the final formulae for $j_{\text{circ}}^{(2)}$ and $j_{\text{lin}}^{(2)}$ are reported here. We shall explain the introduced notations and discuss an expectable distinction between the contributions to the total drag current.

We have to consider another probable contribution to the nonlinear drag current. The matter concerns the effect of resonant saturation (or the Rabi effect) of one and two-photon light absorption (see, e.g., work [2], where the Rabi effect was considered for a gyrotropic medium). Let us consider the contribution of the Rabi effect to the PDE, which is connected with a saturation of one-photon light absorption. In order to take the Rabi effect into account, the substitution

$$\begin{aligned} f_2^{(e)} - f_1^{(e)} &\rightarrow f_2^{(e)} - f_1^{(e)} + \delta f_2^{(e)} - \delta f_1^{(e)} = \\ &= \left(f_2^{(e)} - f_1^{(e)} \right) \left[1 - (T_1 + T_2) \frac{2\pi}{\hbar} \left| M_{12}^{(1)} \right|^2 \times \right. \\ &\times \left. \frac{1}{\pi} \hbar \Gamma \left((\Delta E)^2 + \hbar^2 \Gamma^2 \right)^{-2} \right], \end{aligned} \quad (7)$$

is to be executed under the sum sign in the expression for the one-photon transition probability $W^{(1)}$, where Γ is the attenuation factor [1], T_1 and T_2 are the times of escape of heavy and light holes, respectively, from the region of saturation, and ΔE is the energy difference depending on the type of optical transitions (as a rule, it is equal to the energy difference between the initial and one of the intermediate states). Then, after cumbersome but not very difficult mathematical transformations, we obtain the expression for the current of the N -photon drag effect, where the dependence of the probability of optical transitions on the photon momentum is neglected,

$$\begin{aligned} j^{(N)} &= e \frac{K^{(N)} J}{N} \frac{\hbar}{m_1} N \frac{\mu_-}{m_2} q \left\{ [\tau_p(1, k_N) - \tau_p(2, k_N)] \times \right. \\ &\times \left\langle \cos^2 \varphi \frac{\partial}{\partial k} \left(\frac{k^3 |M_{m', m}|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} |M_{m', m}|^2}} \right) \right\rangle_{k_N} + \\ &+ k_N \left\langle \cos^2 \varphi \left(\frac{k^3 |M_{m', m}|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} |M_{m', m}|^2}} \right) \right\rangle_{k_N} \times \\ &\times \left[e^{\beta \frac{\hbar^2 k_N^2}{2m_1}} \frac{\partial}{\partial k} \left(\tau_p(1, k_N) e^{-\beta \frac{\hbar^2 k^2}{2m_1}} \right) - \right. \end{aligned}$$

$$\left. - e^{\beta \frac{\hbar^2 k_N^2}{2m_2}} \frac{\partial}{\partial k} \left(\tau_p(2, k_N) e^{-\beta \frac{\hbar^2 k^2}{2m_2}} \right) \right] \left. \right\} \frac{1}{\langle |M_{m', m}|^2 \rangle_{k_N}}, \quad (8)$$

and the correction to the one-photon drag current owing to the Rabi effect

$$\begin{aligned} \delta j^{(1)} &= e \frac{\hbar K^{(1)} J}{m_1} \frac{\mu_-}{m_2} q \left\{ [\tau_p(1, k_1) - \tau_p(2, k_1)] \times \right. \\ &\times \left\langle \cos^2 \varphi \left(\frac{5|e'_+|^2}{\sqrt{1 + a|e'_+|^2}} \right) \right\rangle - \langle 5 \cos^2 \varphi |e'_+|^2 \rangle - \\ &- \left\langle \frac{a \cos^2 \varphi |e'_+|^2}{(1 + a|e'_+|^2)^{3/2}} \right\rangle + \\ &+ \left[\left\langle \cos^2 \varphi \left(\frac{|e'_+|^2}{\sqrt{1 + a|e'_+|^2}} \right) \right\rangle - \langle \cos^2 \varphi |e'_+|^2 \rangle \right] \times \\ &\times k_1 e^{\beta \frac{\hbar^2 k_1^2}{2m_1}} \frac{\partial}{\partial k} \left(\tau_p(1, k_1) e^{-\beta \frac{\hbar^2 k^2}{2m_1}} \right) - \\ &- e^{\beta \frac{\hbar^2 k_1^2}{2m_2}} \frac{\partial}{\partial k} \left(\tau_p(2, k_1) e^{-\beta \frac{\hbar^2 k^2}{2m_2}} \right) \left. \right\} \frac{1}{\langle |e'_+|^2 \rangle}, \end{aligned} \quad (9)$$

where $\beta = 1/k_B T$, $\alpha_\omega = 6\omega^2 T_1^{(1)} T_2^{(1)} \frac{I}{I_0}$, $I_0 = \frac{cn_\omega \hbar^3 \omega^3}{2\pi |B|}$ ($I_0 = 13420 \frac{\text{kW}}{\text{cm}^2}$ for p -GaAs at $\hbar\omega = 17 \text{ meV}$), $m_2 = 0.045m_0$, and φ is the angle between the hole and photon wave vectors; the other notations are standard.

In the range of small light intensity, where the methods of perturbation theory are applicable, relation (9) becomes

$$\begin{aligned} \delta j^{(1)} &= -e K^{(1)} J \frac{\hbar q}{m_1 - m_2} \frac{2T_1 T_2}{\hbar^2} \omega^2 \frac{9}{10\sqrt{2}} \times \\ &\times \left(\frac{1}{\frac{11}{9}} \right) \frac{e^{\beta E_1^{(1)}}}{1 + e^{-\beta \hbar \omega}} \left\{ \tau_p(1, k_1) - \tau_p(2, k_2) + \right. \\ &+ \frac{2}{7} \tau_p(1, k_1) \left[\left(\frac{\partial \ln(\tau_1(E))}{\partial \ln E} \right)_{k=k_1} - \frac{\hbar \omega}{k_B T} \frac{\mu_-}{m_1} \right] - \\ &- \frac{2}{7} \tau_p(2, k_2) \left[\left(\frac{\partial \ln(\tau_2(E))}{\partial \ln E} \right)_{k=k_2} - \frac{\hbar \omega}{k_B T} \frac{\mu_-}{m_2} \right] \left. \right\}, \end{aligned} \quad (10)$$

where $\tau_l = \tau_p(l, E)$, $K^{(N)}$ is the coefficient of N -photon light absorption, and μ the reduced effective hole mass.

The expression for the one-photon drag current, derived neglecting the dependences of the matrix elements of the examined optical transitions on \vec{q} , reads

$$\delta j^{(1)} = \frac{3}{2} - \frac{\hbar q}{m_1 - m_2} eK^{(1)} J \frac{1}{10} \{D_1 Q_1 + D_2 Q'_1\}, \quad (11)$$

where

$$D_1 = \tau_p(1, k_1) - \tau_p(2, k_1),$$

$$D_2 = k_1 \left(e^{\beta \hbar^2 k_1^2 / (2m_1)} \frac{\partial}{\partial k} \left(\tau_p(1, k_1) e^{-\beta \hbar^2 k_1^2 / (2m_1)} \right) - e^{\beta \hbar^2 k_1^2 / (2m_2)} \frac{\partial}{\partial k} \left(\tau_p(2, k_1) e^{-\beta \hbar^2 k_1^2 / (2m_2)} \right) \right)_{k_1},$$

$$Q_1 = 5L_2 + 2a \frac{\partial L_2}{\partial a} - \frac{2}{3},$$

and

$$Q'_1 = aL_2 - \frac{2}{15}.$$

For the linear polarization,

$$L_2 = \frac{3+a}{8a^2} \left[1 + \frac{1+a}{\sqrt{a}} \frac{a-3}{a+3} \arcsin \sqrt{\frac{a}{1+a}} \right],$$

while, for the circular one,

$$L_2 = \frac{1}{2\sqrt{2a}} (R_1 + R_2 + R_3),$$

where

$$R_1 = \frac{\sqrt{2}}{\sqrt{a}} \left[\sqrt{1+2a} + \frac{1}{2a} \ln \left(\sqrt{1+2a} - \sqrt{2a} \right) \right],$$

$$R_2 = 4 \frac{\sqrt{2}}{3\sqrt{a^3}} (1-2a) (2a-1 + \sqrt{1+2a}),$$

and

$$R_3 = \frac{\sqrt{2}}{\sqrt{a^3}} \left[(4a - \sqrt{3}) \sqrt{1+2a} - \frac{3}{\sqrt{2a}}, \ln(\sqrt{1+2a} - \sqrt{2a}) \right].$$

In the range of small light intensity, expression (11) takes the form

$$\delta j^{(1)} = eK^{(1)} J \frac{T_1 T_2}{\hbar^2} \hbar \omega \left(\frac{eA_0}{\hbar} \right)^2 |B| \frac{12}{35} \frac{\hbar q}{m_1 - m_2} \times$$

$$\times \left\{ \tau_1^{(1)} \left(\begin{array}{c} 5 - 9 \frac{m_2}{m_1} + 7 \frac{m_2}{\mu_-} \\ 8 - 20 \frac{m_2}{m_1} + 14 \frac{m_2}{\mu_-} \gamma \end{array} \right) + \tau_2^{(1)} \left(\begin{array}{c} 9 - 5 \frac{m_2}{m_1} - 7 \frac{m_2}{\mu_-} \\ 20 - 8 \frac{m_2}{m_1} - 14 \frac{m_2}{\mu_-} \gamma \end{array} \right) \right\},$$

where

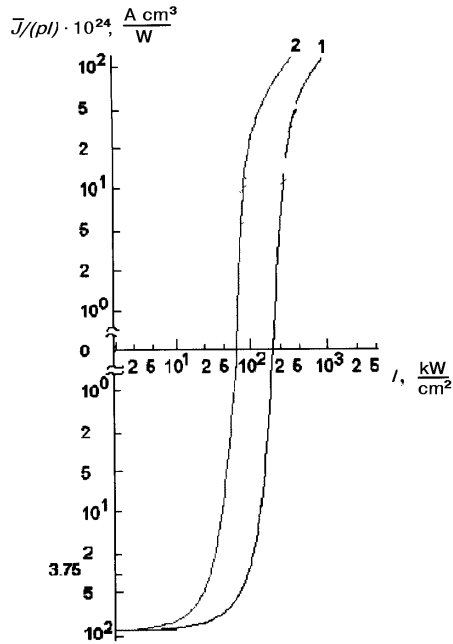
$$\gamma = g\hbar/(2m_0B) = -2g\mu_-/m_0,$$

and g is the hole g -factor.

Thus, possessing the temperature and energy dependences of photohole relaxation times and the equilibrium distributions of photoholes in the final state, as well as the corresponding absorption coefficients, it is easy to compare the experimental and theoretical results obtained for semiconductors with certain symmetry.

For the calculation of the PDE in p -Ge, the following parameters were used: $m_1 = 0.33m_0$, $m_2 = 0.045m_0$, $g = -6.8$, the acoustic length $l_0 = 4.3 \times 10^{-3}$ cm, the constant of interaction with optical phonons $E_{\text{opt}} = 13$ eV, and the energy of optical phonon $\hbar\omega_{\text{opt}} = 37$ meV. In this case, the theoretical value of the quantity $\chi = \bar{J}/(pI)$ for one-, two-, and three-photon light absorption was $\chi_{\text{theor}} = 83 \times 10^{-24} \frac{\text{A cm}^3}{\text{W}}$, while the experimental one $\chi_{\text{exp}} = \bar{J}^{(1)}/pI_0 = 80 \times 10^{-24} \frac{\text{A cm}^3}{\text{W}}$, where \bar{J} is the average value of the resulting photocurrent. We note that χ_{theor} at $E_{\text{opt}} = 11.4$ eV is 1.7 as large as χ_{exp} . When calculating the contribution $\delta J^{(1)}$ of the Rabi effect to the photocurrent, it is necessary also to set the value of $T_1^{(1)} T_2^{(1)}$, which, as was marked in work [1], can be smaller than the product $\tau_1^{(1)} \tau_2^{(1)}$. The best agreement with the experimental values $I_i(\text{lin.}) = 114$ kW/cm² and $I_i(\text{circ.}) = 80$ kW/cm² was achieved for $\omega^2 T_1^{(1)} T_2^{(1)} = 24$, namely, $I_i(\text{lin.}) = 129$ kW/cm² and $I_i(\text{circ.}) = 76$ kW/cm². In this case, the contribution of saturation to the one-photon drag current $\delta J^{(1)}$ is 2.82 times the sum of the two- and three-photon contributions, $J^{(2)}$ and $J^{(3)}$, respectively, with the signs of the latter two currents being the same and opposite to the sign of $J^{(1)}$.

The theoretical dependences of the resulting PDE signal $\chi_{\text{PDE}} = \bar{J}/pI$ on the light intensity, calculated for hole germanium at $m_2 = 0.045m_0$ and $\omega = \sqrt{T_1 T_2}$, and taking the Rabi effect into account, are presented in the figure both for the linear and circular light polarization. Those dependences qualitatively describe the results of experimental researches [1] of the PDE in p -Ge at $T = 300$ K, where an NH₃-laser optically pumped by a CO-one was used as a source of emission. The emission wavelength and the pulse duration were 90.6 μm and 40 ms, respectively.



Theoretical dependences of the photon drag effect signal on the intensity of exciting light with $\lambda = 90.6 \mu\text{m}$ for $p\text{-Ge}$ at room temperature: 1 — linear, 2 — circular polarization

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ДО ТЕОРІЇ ЕФЕКТУ ЗАХОПЛЕННЯ
У НАПІВПРОВІДНИКАХ ЗІ СКЛАДНОЮ ВАЛЕНТНОЮ
ЗОНОЮ ЗА НЕЛІНІЙНОГО ПОГЛИНАННЯ СВІТЛА

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Резюме

Теоретично досліджено ефект захоплення фотонами у напівпровідниках зі складною зонною структурою, що складається з двох близько розташованих або двох вироджених гілок, при нелінійному поглинанні поляризованого світла. Феноменологічно проаналізовано складові струму захоплення для випадку двофоновного поглинання. У квантово-механічних розрахунках враховано внесок у струм захоплення ефекту Рабі для однофоновних оптичних переходів, зумовлений насиченням кінцевих станів фотозбуджених дірок. Теоретичні результати порівняно з експериментальними даними для $p\text{-Ge}$.