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POLARIZATION-DEPENDENT PHOTOCURRENT IN *p*-GaAs

An expression for the spectral and temperature dependences of a photocurrent arising as a result of the linear photovoltaic effect in such semiconductors as gallium arsenide with the hole conduction has been derived. The photocurrent is shown to arise owing to the presence of terms with different parities in the effective hole Hamiltonian. Theoretical and experimental results have been compared.

Keywords: photovoltaic effect, semiconductor, polarization, photocurrent, Hamiltonian, momentum operator, energy spectrum, light absorption coefficient.

1. Introduction

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The development of the powerful laser technique gave rise to the emergence of a number of problems concerning the formation mechanisms of optical and photovoltaic effects, which are to be studied in detail. Of those two, the mechanism of photovoltaic effect (PVE) in noncentrosymmetric semiconductor crystals is a consequence of an asymmetry among elementary electronic processes inherent to noncentrosymmetric media, and the direction of the PVE current is completely determined by light polarization and crystal orientation [1–5]. In spite of the fact that the PVE was observed in a number of semiconductors [6–9], the mechanism of its formation has not been elucidated yet.

The presented work fills this gap to a great extent. It is devoted to a theoretical research of the photon mechanism of the linear photovoltaic effect (LPVE). The latter is related to the absorption of stationary linearly polarized radiation in homogeneous semiconductors of the *p*-GaAs type, but involves the asymmetry of the interaction between holes and photons, which is not associated with the momentum transfer from photons to holes, as it occurs in the effect of hole drag by photons [3].

Note that now we distinguish between the ballistic (BLPVE) and shift (SLPVE) linear PVEs [2–5]. The latter is associated with a shift of the center-of-mass of a hole wave packet at quantum transitions. In semiconductors with an involved valence band, the

SLPVE can arise at both direct optical transitions (the photon mechanism) and asymmetric hole scattering by phonons (the phonon mechanism) [9, 10]. In what follows, the contribution of an asymmetry in the electron-photon interaction to the current of SLPVE is considered. The asymmetry arises owing to the account for the terms in the effective hole Hamiltonian that are linear and cubic (relativistic) in the wave vector. The corresponding contribution was qualitatively estimated in works [10, 11], but only for the ballistic linear PVE.

2. General Relationships

The current of the shift linear PVE is determined by the formula [3]

$$j_{\alpha} = -\frac{e^{3}Ie_{\beta}e_{\gamma}|\delta_{\alpha\beta\gamma}|}{2\pi m_{0}^{2}\omega^{2}\hbar cn_{\omega}} \times \int d\mathbf{k} \operatorname{Im}[P_{21}^{(\beta)^{*}}\frac{\partial}{\partial k_{\alpha}}P_{21}^{(\alpha)}]f_{1\mathbf{k}}\partial(E_{2}-E_{1}-\hbar\omega).$$
(1)

Here, e is the elementary charge, m_0 the free electron mass, I the light intensity, \mathbf{e} the light polarization vector, ω the light frequency, n_{ω} the refractive index, \mathbf{p}_{21} the matrix element of the momentum operator

$$\mathbf{p} = \frac{m_0}{\hbar} \nabla_\mathbf{k} \hat{H},\tag{2}$$

 \hat{H} the effective Hamiltonian, $E_l = \hbar^2 k^2 / (2m_l)$ the energy spectrum, m_l the effective mass of holes in the *l*-th branch (l = 1 for the subband of light holes,

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and l = 2 for the subband of heavy ones), and $\delta_{\alpha\beta\gamma}$ the antisymmetric unit tensor $(\alpha, \beta, \gamma = x, y, z)$.

The hole Hamiltonian, besides the terms that are quadratic in \mathbf{k} [13, 14],

$$\hat{H}^{(2)} = \left(A + \frac{5}{4}B\right)k^2 - B(\mathbf{J}\mathbf{k})^2,$$
(3)

also includes the cubic and linear terms [14],

$$\hat{H}^{(3)} = D'\mathbf{J}\mathbf{K},\tag{4}$$

$$\hat{H}^{(1)} = \frac{4}{\sqrt{3}} k_0 \mathbf{V} \mathbf{k},\tag{5}$$

where $V_{\alpha} = [J_{\alpha}(J_{\alpha+1}^2 - J_{\alpha+2}^2)]$, J_{α} are the matrices of the angular momentum projection operator in the Γ_8 representation [13, 14], and $A \pm B = \hbar^2/(2m_{1,2})$. In further calculations, we take into account that

$$\frac{\partial}{\partial k_{\alpha}} \mathbf{e} \mathbf{p}_{2m\mathbf{k},1n\mathbf{k}} = \hat{F}^{+}_{2m\mathbf{k}} (\frac{\partial}{\partial k_{l}} \mathbf{e} \mathbf{p}) \hat{F}_{1n\mathbf{k}} + \tilde{P}^{(\alpha)}_{2m\mathbf{k},1n\mathbf{k}}, \quad (6)$$

where

$$\tilde{P}_{2m\mathbf{k},1n\mathbf{k}}^{(z)} = i \frac{1-\mu^2}{k} \times \left(\sum_{l_1m_1} (J_y)_{l_1m_1,2m_2} (\mathbf{ep})_{2m_2\mathbf{k},l_1m_1\mathbf{k}} - \sum_{l_1m_1} (J_y)_{2m_2\mathbf{k},l_1m_1\mathbf{k}} \mathbf{ep}_{l_1m_1\mathbf{k},1n\mathbf{k}} \right).$$
(7)

Here, we took into consideration that

$$\frac{\partial \hat{F}_{lmk}}{\partial k_z} = i \frac{1 - \mu^2}{k} \sum_{l_1 m_1} (J_y)_{l_1 m_1 lm} \hat{F}_{l_1 m_1 k}, \tag{8}$$

where $\mu = \mathbf{k}_z \mathbf{k}/k^2$, $\hat{F}_{lm\mathbf{k}}$ is the characteristic matrix of the Hamiltonian \hat{H} , and m the projection of the hole angular momentum onto the vector \mathbf{k} $(m = \pm 1/2)$ in the case l = 2, and $\pm 3/2$ in the case l = 1).

3. Current Calculation for the Shift Linear PVE

Substituting Eq. (2) into Eq. (1) taking Eqs. (4)-(6) into account, summing up over the degenerate hole states, and integrating over the hole wave vector, we obtain the following expression for the current of the shift linear PVE:

$$j_{\alpha}^{(\nu)} = e \frac{I}{\hbar \omega} K(\omega, T) L^{(\nu)} e_{\beta} e_{\gamma} |\delta_{\alpha\beta\gamma}|, \qquad (9)$$

where

$$K(\omega,T) = \frac{e^2}{c\hbar n_{\omega}} f_0(E_1^*) \sqrt{\frac{\hbar\omega}{2B}}$$
(10)

is the coefficient of one-photon light absorption, which is associated with the direct optical transitions of charge carriers between the branches of light and heavy holes in the valence band of *p*-GaAs, $f_0(E_1^*)$ the distribution function of holes over their energy $E_1^* = m_2 \hbar \omega / (m_1 - m_2)$, $L^{(\nu)}$ are the quantities (with the dimension of length)

$$L^{(3)} = -\frac{D'}{2B} \left(1 + \frac{21}{16\pi} I_0 \right), \tag{11}$$
 and

$$L^{(1)} = -\frac{k_0}{\hbar\omega} \left(1 + \frac{4}{\sqrt{3}} \frac{m_1 - m_2}{m_1 + m_2} \right)$$
(12)

used below to quantitatively evaluate the photocurrent, and

$$I_0 = \int_{-1}^{+1} d\mu \int_{0}^{2\pi} \left[O_y^2 O_z^2 \sum_{\alpha} O_{\alpha}^2 \left(O_{\alpha+1}^2 - O_{\alpha+2}^2 \right)^2 \right]^{1/2},$$

where $\mathbf{O} = \mathbf{k}/k$.

It should be noted that the contribution to the SLPVE current arises not only owing to the interference between those terms of the matrix element of the momentum operator that are linear in \mathbf{k} , on the one hand, and the terms that are either \mathbf{k} -independent or quadratic in \mathbf{k} , on the other hand, but also as a result of the appearance of perturbation components in the wave function and in the distribution functions of light and heavy holes. According to our estimation, the account for this circumstance makes an insignificant contribution to the total photocurrent, because it is proportional to the band parameters that are relativistically small with respect to A and B.

The account for Eq. (4) in the Hamiltonian \hat{H} results in the splitting of light-hole subbands, i.e.

$$E_2^{\pm} = \frac{\hbar^2 k^2}{2m_1} \pm D' k^3 \left[\sum_{\alpha} O_{\alpha}^2 (O_{\alpha+1}^2 - O_{\alpha+2}^2) \right]^{1/2}.$$
 (13)

At the same time, the account for Eq. (5) gives rise to the splitting of both subbands, i.e.

$$E_l^{\pm} = \frac{\hbar^2 k^2}{2m_l} \pm \sqrt{3}k_0 k \sqrt{0_x^2 + 0_y^2}.$$
 (14)

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In the former case, the account for the splitting in the energy conservation law (in the argument of the δ -function) brings about an additional contribution to the SLPVE current. In the latter case, according to the results of calculations, no hole shift emerges in the real space at the optical transitions $E_1^{\pm} \rightarrow E_2^{\pm}$, if the splitting of heavy hole subbands is taken into account. Here, as the hole shift, we understand the coordinate matrix element calculated with the use of hole wave functions in the Luttinger representation [13, 14].

From Eq. (9), one can see that the temperature dependence of the current driven by the photon mechanism of the shift linear PVE is determined by the temperature dependence of the light absorption factor K(T) [see Eq. (10)].

In Fig. 1, the theoretical results calculated for the photon mechanism and the experimental data obtained for the temperature dependence of the linear PVE current in *p*-GaAs with the hole concentration $p = 7.4 \times 10^{16}$ cm⁻³ are compared. The theoretical calculations were carried out following the method proposed in works [10, 12]. Note that the shift of the theoretical dependence with respect to the experimental one occurs due to the contribution of other mechanisms, which were considered in works [10, 12] in detail. However, the mechanism proposed here was not taken into consideration in the cited works.

4. Evaluation of Photocurrent $L^{(\nu)}$

The values of the constant k_0 for various crystals fall within the limits of $(1\div 6) \times 10^{-10}$ eV cm (see, e.g., work [12]). Accepting those values of k_0 for p-GaAs, we obtain that, when illuminating it by a CO₂ laser ($\hbar\omega = 0.12 \text{ eV}$) at room temperature, the quantity $L_1^{(1)} = (0.2\div 1.4) \times 10^{-8} \text{ cm} = (0.15\div 1)X_{\text{exp}}$, where $X_{\text{exp}} = 0.17 \times 10^{-7} \text{ cm}$. This evaluation testifies that the contribution to the current of the shift linear PVE, which is associated with the asymmetry of electron-photon interactions emerging when the relativistic linear in **k** terms in \hat{H} are taken into consideration, is comparable with the experimental value.

Again, in the case $\mathbf{k} \parallel [011]$ for *p*-GaAs with the parameters $|D'| = 3.25 \times 10^{-23} \text{ eV cm}^3$, $B = 3.25 \times 10^{-15} \text{ eV cm}^2$, $m_2 = 0.68m_0$, and $m_1 = 0.12m_0$, we obtain $L^{(3)} = 4.2 \times 10^{-8} \text{ cm} = 2.7X_{\text{exp}}$.

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Fig. 1. Temperature dependences of the linear PVE current for p-GaAs at the hole concentration $p = 7.4 \times 10^{16}$ cm⁻³. Experimental results for the light wavelengths $\lambda = 10.6$ (squares) and 9.5 μ m (circles) [12]. Theoretical results for the photon mechanism of shift linear PVE in p-GaAs: $\lambda = 0.5$ (solid curve) and 10.6 μ m (diamonds)

It should be noted that the analyzed literature sources contain no information about the signs of band parameters before the linear and cubic (in the wave vector) terms in the effective Hamiltonian. Therefore, it is impossible to talk about those contributions to the photocurrent that depend on the constants D' and k_0 . However, if we adopt the quantity D' to be positive, and k_0 negative, then the total contribution to the photocurrent approaches the experimental value. At the same time, if those quantities are of the same sign, it is quite natural that the magnitude of the resulting contribution to the photocurrent is strongly different from the experimental value.

The results of calculations demonstrate that if the values of band parameters do not change, the growth of the photon energy is accompanied by an increase of the photocurrent maximum and its shift toward higher temperatures (here, as a photocurrent, we understand the current driven by the photon mechanism of the shift linear PVE in p-GaAs). For example, in the case $m_2 = 0.68m_0$ and $m_1 = 0.12m_0$, if $\hbar\omega = 117$ meV, the photocurrent maximum associated with the phonon emission takes place at T = 120 K, and that associated with the phonon absorption is observed at T = 360 K. At the same time, if $\hbar\omega = 130$ meV, the corresponding values equal T = 180 and 420 K. With the growth of the photon energy, the maximum value of photocurrent corresponding to the phonon absorption almost does

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Fig. 2. Feynman–Keldysh diagrams, whose interference gives a contribution to the current of the linear photovoltaic effect in structures with quantum wells: processes with the participation of real phonons and virtual photons (A), and processes with the participation of real photons and virtual phonons (B)

not change, and that corresponding to the phonon emission becomes 1.135 times larger.

It is worth noting that if the effective mass of light holes increases (for example, by a factor of 4/3), provided that the effective mass of heavy holes ($m_1 =$ $= 0.12m_0$) and the light frequency ($\hbar\omega = 117 \text{ meV}$) remain the same, the maximum of the photocurrent associated with the phonon emission grows (by approximately a factor of 1.6) and shifts toward higher temperatures. In this case, the maximum of the photocurrent associated with the phonon absorption almost does not change: it slightly – more precisely, by a factor of 1.08 – decreases, but also shifts toward higher temperatures.

Finally, we note that the experimental [12] and presented theoretical temperature dependences of photocurrent are comparable in the case where the processes associated with the phonon absorption prevail at $m_1/m_2 = 1.135$ as well. Note also that the SLPVE can arise in nanostructures of a noncentrosymmetric semiconductor. In this case, the contribution to the SLPVE current is given by the interference of the processes shown in Fig. 2. In this figure, the processes, in which real phonons and photons participate simultaneously, are not depicted. Here, we imply that charge carriers are asymmetrically scattered at both phonons (the phonon mechanism of SLPVE) and photons (the photon mechanism of SLPVE) [14–17].

On the basis of the energy conservation relations between the initial, final, and one of the transient states of photo-excited charge carriers, it is not difficult to obtain the following expressions for the wave vectors of charge carriers participating in the formation of the SLPVE current:

$$\begin{split} k_{\omega}^{2} &= \sqrt{\frac{2}{\hbar^{2}} \mu_{21}^{(n_{2}n_{1})} \hbar \omega_{21}^{(n_{2}n_{1})}}, \\ k_{1}^{\prime 2} &= \frac{2}{\hbar^{2}} m_{1}^{(n_{1}')} \times \\ &\times \left[\frac{\mu_{21}^{(n_{2}n_{1})}}{m_{1}^{(n_{1})}} \hbar \omega_{21}^{(n_{2}n_{1})} + \frac{\hbar^{2} \pi^{2}}{2m_{1} a^{2}} \left(n_{1}^{2} - n_{1}^{\prime 2} \right) \mp \hbar \Omega \right], \\ k_{2}^{\prime 2} &= \frac{2m_{2}^{(n_{2}')}}{\hbar^{2}} \times \\ &\times \left\{ \frac{\mu_{21}^{(n_{2}n_{1})}}{m_{1}^{(n_{1})}} \hbar \omega_{21}^{(n_{2}n_{1})} \mp \hbar \Omega + \frac{\hbar^{2} \pi^{2}}{2m_{1}} \left(n_{1}^{2} - n_{2}^{\prime 2} \right) \right\}, \\ k_{3}^{\prime 2} &= \frac{2m_{1}^{(n_{2}')}}{\hbar^{2}} \times \\ &\times \left\{ \frac{\mu_{21}^{(n_{2}n_{1})}}{m_{1}^{(n_{1})}} \hbar \omega_{21}^{(n_{2}n_{1})} + \hbar \omega \mp \hbar \Omega + \frac{\hbar^{2} \pi^{2}}{2a^{2}} \left(\frac{n_{1}^{2} - n_{2}^{\prime 2}}{m_{1}} \right) \right\}, \\ k_{4}^{\prime 2} &= \frac{2m_{2}^{(n_{2}')}}{\hbar^{2}} \times \\ &\times \left\{ \frac{\mu_{21}^{(n_{2}n_{1})}}{m_{2}^{(n_{2})}} \hbar \omega_{21}^{(n_{2}n_{1})} + \hbar \omega \mp \hbar \Omega + \frac{\hbar^{2} \pi^{2}}{2a^{2}} \left(\frac{n_{1}^{2} - n_{2}^{\prime 2}}{m_{1}} \right) \right\}, \\ k_{5}^{\prime 2} &= \frac{2}{\hbar^{2}} \mu_{21}^{(n_{2}n_{1})} \left[\hbar \omega - \frac{\hbar^{2} \pi^{2}}{2a^{2}} \left(\frac{n_{1}^{2}}{m_{2}} - \frac{n_{1}^{\prime 2}}{m_{1}} \right) \right], \\ k_{6}^{\prime 2} &= \frac{\mu_{21}^{(n_{2}n_{1})}}{a^{2}} \left(\frac{n_{1}^{2}}{m_{1}} - \frac{n_{2}^{2}}{m_{2}} \right) \pi^{2}. \end{split}$$

Here, ω is the light frequency, *a* the quantum well width, n_l the number of the size-quantized state, m_l the effective bulk mass of holes in the *l*-th branch (l = 2 or lh for light holes and l = 1 or hh for heavy ones in semiconductors with the hole conduction and a complicated valence band), $m_l^{(n)}$ the effective mass of charge carriers in the *l*-th branch in the direction transverse to the size-quantization axis,

$$\begin{split} \hbar\omega_{21}^{(n_2n_1)} &= \hbar\omega - \frac{\hbar^2\pi^2}{2a^2} \left(\frac{n_2^2}{m_2} - \frac{n_1^2}{m_1}\right)\!\!,\\ \hbar\omega_{l'l}^{(n'n)} &= \hbar\omega - \frac{\hbar^2\pi^2}{2a^2} \left(\frac{n'^2}{m_{l'}} - \frac{n^2}{m_l}\right)\!\!, \end{split}$$

where the upper sign corresponds to the absorption and the lower one to the emission of a longitudinal optical phonon.

While deriving the expressions given above, it was taken into account that the quantum well favors the

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appearance of an additional mechanism of intraband light absorption. This mechanism is connected with the direct optical transitions of free charge carriers between the states that emerge owing to the size quantization. These transitions, when running at the center of the Brillouin zone, are described by the corresponding effective bulk masses. If they run at the Brillouin zone periphery, they are also described by effective transverse masses. The latter, in turn, depend on the numbers of size-quantization bands for both the initial and final states. When calculating the contribution of those transitions to the photocurrent, the partial filling of states has to be taken into account in the formula for the SLPVE current in bulk crystals. Therefore, the calculation of the photocurrent includes not only the summation over the identical numbers of size-quantized states n_1 and n_2 (this procedure involves the optical transitions between the light and heavy hole subbands in the *p*-GaAs valence band), but also over different n_1 and n_2 , which corresponds to making allowance for optical transitions within the same subband. This issue will be considered elsewhere.

It is worth noting that a similar situation arises, while calculating the photon-drag current in the nanostructure of a semiconductor with complicated band consisting of two or several closely located or degenerate subbands. This problem should also be considered separately.

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ПОЛЯРИЗАЦІЙНІ ЗАЛЕЖНОСТІ ФОТОСТРУМУ В *p*-GaAs

Резюме

Отримано вираз для спектральної і температурної залежностей струму фотонного механізму лінійного фотогальванічного ефекту в напівпровідниках типу арсеніду галію діркової провідності, зумовлений наявністю доданків різної парності в ефективному гамільтоніані дірок. Зіставлені теоретичні та експериментальні результати.