

# Multiphoton absorption in semiconductors at submillimeter wavelengths

S. D. Ganichev, S. A. Emel'yanov, E. L. Ivchenko, E. Yu. Perlin, Ya. V. Terent'ev, A. V. Fedorov, and I. D. Yaroshetskii

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A theory is developed of nonlinear absorption of high-power submillimeter radiation that induces direct multiphoton transitions of free carriers between the subbands of a complex band. A detailed analysis is made of a strongly nonlinear regime in which a new type of nonlinear absorption of light takes place in crystals, as found recently, for *p*-type Ge. This absorption represents a set of simultaneous *n*-photon transitions making comparable contributions to the total absorption coefficient. The results of a theory of such absorption are compared with the experimental data on the dependences of the optical transmission of *p*-type Ge samples of different thickness on the intensity of the incident light ( $\lambda = 90.55 \mu$ ). It is shown that the change in the sign of the current representing the drag of electrons by photons in *p*-type Ge, observed experimentally on increase in the intensity of the incident light, is associated with the difference between the signs of the one-photon and many-photon ( $n \geq 2$ ) components of the photocurrent. It is shown that the intensity of light  $I^{(0)}$  at which the sign of the drag current in *p*-type Ge changes at room temperature can be calculated simply by determining the one- and two-photon drag currents in the lowest order of perturbation theory. The theoretical value of  $I^{(0)}$  and the nature of the dependence of the drag current on the intensity  $I$  in the region  $I \sim I^{(0)}$  are in good agreement with the experimental results.

## 1. INTRODUCTION

Experimental investigations of *n*-photon absorption in crystals are usually made under conditions when optical transitions with fewer photons are forbidden. For example, in studies of two- or three-photon interband absorption of light in semiconductors the photon energy  $\hbar\omega$  is selected so that for  $n = 2$  and  $n = 3$  the inequalities  $(n - 1)\hbar\omega < E_g < n\hbar\omega$  are satisfied ( $E_g$  is the band gap of the semiconductor). In this case the coefficient of *n*-photon absorption  $K^{(n)}$  representing interband transitions due to incident light of intensity  $\sim 1 \text{ MW/cm}^2$  may be considerably greater than the contribution made to the absorption coefficient by intraband optical transitions or band-impurity level transitions. Then, many-photon transitions of higher order can be ignored because in the case of radiation with the wavelengths in the visible, near infrared, and middle infrared ranges the value of  $K^{(n)}$  is much higher than the coefficient  $K^{(n+1)}$  of  $(n + 1)$ -photon absorption right up to light intensities corresponding to the damage threshold of the investigated materials. For the same reason, the main contribution in the case of intraband absorption of light comes from one-photon processes, and many-photon transitions with intermediate virtual states make a negligible contribution.

The appearance of lasers capable of generating high-power submillimeter radiation<sup>1</sup> has revealed new aspects of the interaction of electromagnetic radiation with solids. In fact, if the calculation of the *n*-photon absorption coefficients  $K^{(n)}$  is limited to the lowest order of perturbation theory for each of these processes, it is found that the ratio  $\eta_n = K^{(n)}/K^{(n+1)}$  (which can be conveniently called the many-photon parameter of order *n*) is proportional to  $I\omega^{-3}$

in the case of intraband absorption of light, where  $I$  is the intensity of light in a crystal. In the case of submillimeter radiation of wavelength  $\lambda \approx 100 \mu$  and intensity  $I \sim \text{MW/cm}^2$  the parameter  $\eta_n$  is comparable with unity for the first few values of *n*. A new type of nonlinear absorption of light due to a set of simultaneous *n*-photon transitions with comparable contributions to the total absorption coefficient has been observed recently for *p*-type Ge crystals subjected to high-power pulses from an optically pumped  $\text{NH}_3$  laser ( $\lambda = 90.55 \mu$ ,  $\hbar\omega = 13.7 \text{ meV}$ ).<sup>2,3</sup> Experimental investigations of the dependence of the optical transmission<sup>3</sup> and of the drag photocurrent<sup>4</sup> have been reported.

We shall propose a general approach to the problem of many-photon absorption as a result of intraband transitions and concentrate our attention on the strong nonlinearity regime when the many-photon parameter is  $\eta_n \sim 1$ . We shall use this approach to study also a new type of nonlinear absorption of light in semiconductors, which was reported, as already mentioned, in Refs. 3 and 4.

In Sec. 2 we shall calculate the two- and three-photon absorption coefficients for the case when  $\eta_n \ll 1$ , using the standard form of perturbation theory. In Sec. 3 we shall use a method which goes beyond the perturbation theory framework and calculate the values of  $K^{(n)}$  for any value of *n*. The expressions which are then obtained are valid both for  $\eta_n < 1$  and  $\eta_n \gtrsim 1$ . Preliminary results of these calculations were published earlier in a brief communication.<sup>3</sup> The current due to the drag of holes in the case of two-photon absorption by intersubband transitions is calculated in Sec. 4. Since the drag effect in *p*-type semiconductors can be considered conveniently not in the hole but in the electron representation, this representation will be used throughout the present paper in calculations of the matrix elements and of the probabi-

lities of optical transitions. In Secs. 3 and 4 we shall also analyze the experimental data on the absorption of light and on the drag effect, respectively, using the theory developed in these sections.

## 2. PERTURBATION-THEORY CALCULATION OF ABSORPTION COEFFICIENTS OF LIGHT

In this section we shall calculate the two- and three-photon absorption coefficients for linearly polarized light when direct transitions take place between the heavy- and light-hole subbands of the band  $\Gamma_8^+$  in Ge-type semiconductors (Fig. 1). In accordance with the experimental conditions in Refs. 3 and 4, we shall assume that the photon energy  $\hbar\omega$  satisfies the inequalities

$$\hbar\omega \ll \Delta_{so}, E_g, \quad (1)$$

where  $\Delta_{so}$  is the spin-orbit splitting of the valence band. Under these conditions the absorption of light can be considered using the effective Hamiltonian  $H^{(2)}(\vec{\mathcal{H}})$ , where  $\vec{\mathcal{H}} = -i\nabla + (eA/c\hbar)$ ,  $-e$  is the electron charge,

$$A = 2A_0 e \cos(\mathbf{q}\mathbf{r} - \omega t)$$

is the vector potential of the light wave, and  $H^{(2)}(\mathbf{k})$  is the effective Hamiltonian of electrons in the energy band  $\Gamma_8^+$  in the absence of illumination [this Hamiltonian is a  $4 \times 4$  matrix quadratic in the electron wave vector  $\mathbf{k}$  (see, for examples, Ref. 5)]. The calculations will be carried out in the spherical approximation when

$$H^{(2)}(\mathbf{k}) = (A + 5/4 B) k^2 - B(\mathbf{J}\mathbf{k})^2, \quad (2)$$

where  $J_\alpha$  ( $\alpha = x, y, z$ ) are the matrices of the angular momentum operator in the basis of states with the total momentum  $3/2$ ;  $A$  and  $B$  are the parameters of the energy band structure. In this case the energy of an electron in the  $l$ th band is

$$E_{l\mathbf{k}} = -\frac{\hbar^2 k^2}{2m_l}, \quad m_{1,2} = -\frac{\hbar^2}{2(A \mp B)}. \quad (3)$$

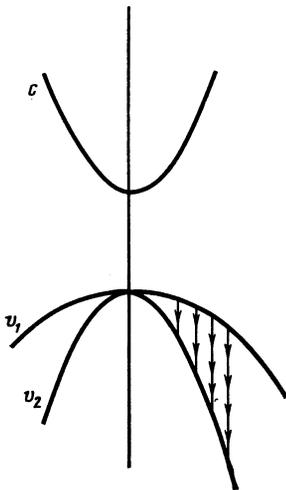


FIG. 1. Direct  $n$ -photon transitions of electrons between the subbands  $v_2$  and  $v_1$  of the valence band  $\Gamma_8^+$  ( $n = 1, 2, 3, 4, \dots$ ).

The electrons in the heavy-hole subband  $v_1$  ( $l = 1$ ) correspond to states with the projection  $m = \pm 3/2$  of the angular momentum along the direction of the vector  $\mathbf{k}$ , whereas the electrons in the light-hole subband  $v_2$  ( $l = 2$ ) correspond to the states with  $m = \pm 1/2$ . In view of the smallness of the wave vector of light  $\mathbf{q}$  compared with the wave vectors of electrons participating in optical transitions, we can calculate  $K^{(n)}$  assuming that  $\mathbf{q} = 0$  (in Sec. 4 we shall allow for the terms linear in  $\mathbf{q}$ , which occur in the optical transition rates). In the spatially homogeneous approximation the matrix elements of the interaction operator

$$H^{int} = H^{(2)}(\vec{\mathcal{H}}) - H^{(2)}(-i\nabla) = H_1^{int} + H_2^{int} \quad (4)$$

can be written as follows:

$$H_{1,m'm}^{int}(\mathbf{k}) = 2 \cos \omega t M_{m'm}^{(1)}(\mathbf{k}), \quad (5)$$

$$H_{2,m'm}^{int}(\mathbf{k}) = 4 \cos^2 \omega t \left( \frac{eA_0}{c\hbar} \right)^2 H_{m'm, \mathbf{k}, \mathbf{m}\mathbf{k}}^{(2)}(\mathbf{e}). \quad (6)$$

Here, the index  $m$  labels the states in the subbands  $v_1$  ( $m = \pm 3/2$ ) and  $v_2$  ( $m = \pm 1/2$ ), whereas the matrix element of a one-photon transition  $M_{m'm}^{(1)}$  is related to the matrix elements of the velocity operator  $\hat{v}$ :

$$M_{m'm}^{(1)}(\mathbf{k}) = \frac{e}{c} A_0 \mathbf{e} \mathbf{v}_{m'm}(\mathbf{k}), \quad (7)$$

$$H_{m'm, \mathbf{k}, \mathbf{m}\mathbf{k}}^{(2)}(\mathbf{e}) = \hat{C}_{m'm, \mathbf{k}}^+ H^{(2)}(\mathbf{e}) \hat{C}_{m\mathbf{k}}, \quad (8)$$

where  $\hat{C}_{m\mathbf{k}}$  are the singular columns of the matrix  $H^{(2)}(\mathbf{k})$  and  $\mathbf{e}$  is the polarization vector of light. It should be noted that the second term on the right-hand side of Eq. (4) describes in the effective-mass method the simultaneous interaction of an electron with two photons. In the many-band model, when the conditions of Eq. (1) are met, this term describes the two-photon transitions with virtual states in the other bands, particularly in the conduction band.

In the spherical approximation and for a suitable selection of the phase factors of the columns  $\hat{C}_{m\mathbf{k}}$  the matrix elements  $\mathbf{e} \mathbf{v}_{m'm}$  and  $H_{m'm, \mathbf{k}, \mathbf{m}\mathbf{k}}^{(2)}(\mathbf{e})$  can be represented in the form

$$\mathbf{e} \mathbf{v}_{m'm}(\mathbf{k}) = -\frac{\hbar k}{m_l} e_z \delta_{m'm} + \frac{3^{1/2} B k}{\hbar} \delta_{|m-m'|,1} [e_{x'} + i(m-m') e_{y'}], \quad (9)$$

$$H_{m'm, \mathbf{k}, \mathbf{m}\mathbf{k}}^{(2)}(\mathbf{e}) = H_{m'm}^{(2)}(\mathbf{e}'), \quad (10)$$

where  $e_\alpha$  are the Cartesian coordinates of the polarization vector of light  $\mathbf{e}$  in the coordinate system  $x', y', z'$ , with the  $z'$  axis parallel to  $\mathbf{k}$ ,  $\mathbf{e}'$  is the vector with the projections  $e_{x'}$ ,  $e_{y'}$ ,  $e_{z'}$  along the axes  $x, y, z$  of a fixed coordinate system.

In the effective-Hamiltonian method, when the composite matrix element  $M_{m'm}^{(n)}(\mathbf{k})$  is calculated for an electron transition accompanied by the absorption of  $n$  photons, it is sufficient to allow for intermediate virtual states within the same energy band  $\Gamma_8^+$ . The absorption coefficient  $K^{(n)}$  can then be represented in the form

$$K^{(n)} = \frac{2\pi}{\hbar} \frac{n\hbar\omega}{I} \rho(n\hbar\omega) F(\beta, n, \omega) \overline{(|M_{3/2, 1/2}^{(n)}|^2 + |M_{-3/2, 1/2}^{(n)}|^2)}, \quad (11)$$

$$F(\beta, n, \omega) = [1 - \exp(-n\beta\hbar\omega)] \exp[\beta(\chi - nE_0)], \quad (12)$$

where the intensity of light is  $I = n\omega^2 A_0^2 (2\pi c)^{-1}$ ;  $n\omega$  is the refractive index of light;  $\chi$  is the chemical potential of holes;

$E_0 = \hbar\omega m_2 / (m_1 - m_2)$ ,  $\beta = (k_B T)^{-1}$ ,  $\rho(E)$  is the reduced density of states described by

$$\rho(E) = 2 \sum_{\mathbf{k}} \delta(E_{1\mathbf{k}} - E_{2\mathbf{k}} - E) = \mu k_x (\pi \hbar)^{-2}, \quad (13)$$

where

$$k_x = (2\mu E / \hbar^2)^{1/2}, \quad \mu = \hbar^2 / 4 |B| = m_1 m_2 / (m_2 - m_1),$$

$|\overline{M_{m'm}^{(n)}}|^2$  is the square of the modulus of the matrix element  $M_{m'm}^{(n)}(\mathbf{k})$  and is averaged over all the directions of the wave vector  $\mathbf{k}$  (it is assumed that the volume of the crystal is unity). We shall omit the intermediate steps and give the final expressions for  $|\overline{M_{m'm}^{(n)}}|^2$  when  $n = 1, 2, 3$ :

$$\begin{aligned} |\overline{M_{3/2, 1/2}^{(1)}}|^2 &= \hbar\omega\varepsilon, & |\overline{M_{-3/2, 1/2}^{(1)}}|^2 &= 0, \\ |\overline{M_{3/2, 1/2}^{(2)}}|^2 &= 18/5 \varepsilon^2, & |\overline{M_{-3/2, 1/2}^{(2)}}|^2 &= 2/5 \varepsilon^2, \\ |\overline{M_{3/2, 1/2}^{(3)}}|^2 &= 9.82 \varepsilon^3 / \hbar\omega, & |\overline{M_{-3/2, 1/2}^{(3)}}|^2 &= 1.37 \varepsilon^3 / \hbar\omega, \end{aligned} \quad (14)$$

where

$$\varepsilon = |B| (eA_0 / c \hbar)^2. \quad (15)$$

In Eq. (14) an allowance is made for the fact that in the case of an  $n$ -photon transition we have  $E_{1\mathbf{k}} - E_{2\mathbf{k}} = n\hbar\omega$ . For compactness, the expression describing  $|\overline{M_{3/2, 1/2}^{(3)}}|^2$  does not include the terms containing a small factor  $(m_2 / m_1)^2$ .

The value of  $K^{(2)}$  for carrier transitions in the  $\Gamma_8^+$  band was calculated in Ref. 6, but no allowance was made for the term proportional to  $H_{m'm}^{(2)}(\mathbf{e}')$  in  $M_{m'm}^{(2)}(\mathbf{k})$ . This resulted in an overestimate of the value of  $K^{(2)}$  given in Ref. 6 by a factor of 1.6. It should be noted that  $1/2 \rightarrow -3/2$  and  $-1/2 \rightarrow 3/2$  transitions appear in the spherical approximation only because of the contribution made to  $M_{m'm}^{(n)}(\mathbf{k})$  and proportional to  $H_{m'm}^{(2)}(\mathbf{e}')$ . According to Eq. (14), the relative contribution of these transitions to  $n$ -photon absorption for  $n = 2$  and 3 is small (it is 0.1 for  $n = 2$  and 0.12 for  $n = 3$ ). Therefore, in developing a theory of many-photon absorption in Sec. 3 we shall ignore the  $\pm 1/2 \rightarrow \mp 3/2$  transitions [this naturally does not mean neglect of the matrix elements  $H_{\pm 3/2, \pm 3/2}^{(2)}(\mathbf{e}')$ ,  $H_{\pm 1/2, 1/2}^{(2)}(\mathbf{e}')$ ,  $H_{3/2, 1/2}^{(2)}(\mathbf{e}')$ ,  $H_{-3/2, -1/2}^{(2)}(\mathbf{e}')$ , which are included in Sec. 3].

In the case of radiation with a photon energy  $\hbar\omega = 13.7$  meV and with an intensity  $I = 1$  MW/cm<sup>2</sup>, when  $T = 78$  K,  $n_\omega = 4$ ,  $m_1 = 0.33m_0$ , and  $m_2 = 0.045m_0$ , we find from Eqs. (11)–(14) that  $\eta_2 = K^{(2)} / K^{(1)} = 0.62$ ,  $\eta_3 = K^{(3)} / K^{(2)} = 0.25$ .

In the experiments reported in Refs. 3 and 4 the maximum values of the power density of the laser radiation incident on Ge crystal and transmitted into the crystal were respectively  $I_{0, \max} = 2$  MW/cm<sup>2</sup> and  $I_{\max} = I_{0, \max} \times 4n_\omega / (n_\omega + 1)^2 = 1.28$  MW/cm<sup>2</sup>. In this case the quantities  $\eta_2$  and  $\eta_3$  calculated from Eqs. (11)–(14) assume the values 0.75 and 0.3, respectively, and the results of the conventional perturbation theory are inapplicable.

### 3. MANY-PHOTON ABSORPTION OF HIGH-POWER RADIATION

In the calculation of the coefficients of many-photon absorption as a result of intersubband transitions under con-

ditions when  $\eta_n \sim 1$  we cannot use the standard perturbation theory, as already pointed out in Secs. 1 and 2. Difficulties are also encountered on application of asymptotic methods of the type described in Refs. 7 and 8 and utilizing smallness of the ratio  $\hbar\omega / (E_{1\mathbf{k}} - E_{2\mathbf{k}})$ , because the energy gap  $E_{1\mathbf{k}} - E_{2\mathbf{k}}$  between the states in the light- and heavy-hole subbands vanishes at  $\mathbf{k} = 0$ . Higher orders of perturbation theory can be applied to these effects if exact allowance is made for the diagonal (in the indices  $m$ ) part of the interaction  $H_d^{\text{int}}$  and if in the first or second order of perturbation theory the nondiagonal part  $H_{nd}^{\text{int}}$  is included. However, this approximation is unsatisfactory, because the contributions of  $H_d^{\text{int}}$  and  $H_{nd}^{\text{int}}$  of  $n$ th order in the amplitude are comparable.

We shall describe a method for calculating the  $n$ -photon absorption coefficients which makes it possible to include, for an arbitrary value of  $n$ , the effects of higher orders, including the shift of the energy bands of a crystal in the field of strong radiation. The general expressions obtained for  $K^{(n)}$  in the limit of low intensities ( $\eta_n \ll 1$ ) when  $n = 1, 2$  or 3 reduce to Eqs. (11)–(14) of perturbation theory. On the other hand, if  $I$  is high (and  $\eta_n \sim 1$ ), the convergence of the series used to find  $K^{(n)}$  is sufficiently rapid, so that the values of  $K^{(n)}$  necessary for comparison with the experimental results can be obtained by relatively simple numerical calculations. As pointed out in Sec. 2, the  $\pm 1/2 \rightarrow \mp 3/2$  transitions will be ignored, so that we can reduce our analysis of a system of two doubly degenerate levels  $(v_1, m, \mathbf{k})$  and  $(v_2, m', \mathbf{k})$  with  $m = \pm 3/2$  and  $m' = \pm 1/2$  to two independent two-level systems  $(v_1, 3/2, \mathbf{k})$ ,  $(v_2, 1/2, \mathbf{k})$  and  $(v_1, -3/2, \mathbf{k})$ ,  $(v_2, -1/2, \mathbf{k})$ . States of such a two-level system will be denoted briefly by the indices 1 and 2.

We shall transform the Hamiltonian  $H^{(2)}(\vec{\mathcal{X}})$ :

$$\tilde{H} = \exp(-\hat{S}) H^{(2)}(\vec{\mathcal{X}}) \exp(\hat{S}) - i \frac{\partial \hat{S}}{\partial t}. \quad (16)$$

The operator  $\hat{S}$  will be selected in such a way that  $\tilde{H}$  has no linear (in the field) nondiagonal terms:

$$[H^{(2)}(\mathbf{k}, \hat{S})]_{nd} + H_{1,nd}^{\text{int}} - i \frac{\partial \hat{S}_{nd}}{\partial t} = 0. \quad (17)$$

This condition is satisfied by the matrix

$$S_{12} = -S_{21}^* = u_+ e^{-i\omega t} + u_- e^{i\omega t}, \quad S_{11} = S_{22} = 0, \quad (18)$$

$$u_\pm = M_{12}^{(1)} (E_{2\mathbf{k}} - E_{1\mathbf{k}} \pm \hbar\omega)^{-1}. \quad (19)$$

The transformation (16)–(19) makes it possible to include in the diagonal part  $\tilde{H}_d$  of the transformed  $\tilde{H}$  the nondiagonal part of the initial Hamiltonian  $H^{(2)}(\vec{\mathcal{X}})$ . Then,  $\tilde{H}_d$  is allowed for exactly. In Ref. 9, where a similar method was used to calculate the probabilities of many-photon transitions between the valence and conduction bands, the terms up to the second order in respect of the field were retained in  $\tilde{H}_d$  and up to the third order in  $\tilde{H}_{nd}$ . We shall show later that this approximation is valid if  $|u_\pm| \ll 1$ , whereas in the case of interest to us ( $I \gtrsim 1$  MW/cm<sup>2</sup>,  $\hbar\omega \sim 10^{-2}$  eV), we have  $|u_\pm| \sim 1$ .<sup>1)</sup> We shall therefore utilize the circumstance that in the case of the two-level system under consideration the

transformation described by Eqs. (16)–(19) can be performed exactly (see the Appendix). This makes it possible to express the matrix elements of  $\tilde{H}$  in the form of an expansion in terms of harmonics of the field.

$$\tilde{H}_{11} - \tilde{H}_{22} = E_1 - E_2 + \hbar\omega$$

$$\times \left[ \frac{1}{2} \xi_0(u_+, u_-) + \sum_{p=1}^{\infty} \xi_p(u_+, u_-) \cos p\omega t \right], \quad (20)$$

$$\tilde{H}_{12} = \hbar\omega e^{i\psi_0} \sum_{p=-\infty}^{\infty} e^{ip\omega t} \zeta_p(u_+, u_-), \quad (21)$$

where  $\psi_0 = \arg M_{12}$  and the coefficients  $\xi_p(u_{\pm})$  and  $\zeta_p(u_{\pm})$  are given in the Appendix. At low values of  $I$ , when  $|u_{\pm}| \ll 1$ , these coefficients are proportional to  $I^{p/2}$  ( $p \geq 1$ ). If  $|u_{\pm}| \gtrsim 1$ , we find that  $\xi_p$  and  $\zeta_p$  are complex oscillatory functions of the intensity. Calculations indicate that for moderate values  $|u_{\pm}| \lesssim 3$  the coefficients  $\xi_p$  and  $\zeta_p$  decrease rapidly on increase in  $p$  and it is in practice sufficient to include only the first few harmonics in the summation over  $p$  in Eqs. (20) and (21).

The probability that an electron which is at a moment  $\tau = 0$  in a state  $|2\rangle$  can find itself at  $\tau = t$  in a state  $|1\rangle$  is given by:

$$W_{21}(t) = \text{Tr} \left[ \exp\left(-\frac{iHt}{\hbar}\right) \mathbf{P} \exp\left(\frac{iHt}{\hbar}\right) (\mathbf{I} - \mathbf{P}) \right]$$

$$= \text{Tr} [\tilde{T} + \tilde{\mathbf{P}}(t) \tilde{T} (\mathbf{I} - \tilde{\mathbf{P}})], \quad (22)$$

where  $\mathbf{I}$  is a unit ( $2 \times 2$ ) matrix,  $H \equiv H^{(2)}(\vec{\mathcal{H}})$ ,

$$\mathbf{P} = \begin{vmatrix} 0 & 0 \\ 0 & 1 \end{vmatrix}, \quad \tilde{\mathbf{P}} = \exp(-\delta) \mathbf{P} \exp(\delta), \quad (23)$$

$$\tilde{\mathbf{P}}(t) = \exp\left(-\frac{i}{\hbar} H_{at} t\right) \tilde{\mathbf{P}} \exp\left(\frac{i}{\hbar} H_{at} t\right),$$

$$\tilde{T} = T \exp \left\{ \frac{i}{\hbar} \int_0^t \exp\left(-\frac{iH_{ad}\tau}{\hbar}\right) H_{nd} \exp\left(\frac{iH_{ad}\tau}{\hbar}\right) d\tau \right\}. \quad (24)$$

We shall confine ourselves to the lowest order in  $\tilde{H}_{nd}$  using Eqs. (23), (24), (A.3), and (A.4); after some transformations, we find from Eq. (22) that

$$W_{21}(t) = |v_0^{(1)}(u_+, u_-)|^2 T_{21}^{+(1)} T_{12}^{(1)}, \quad (25)$$

where  $T_{ij}^{(1)}$  are the matrix elements of the operator  $\hat{T}$  calculated in the first order in  $\tilde{H}_{nd}$ . The quantity  $v_0^{(1)}$  is described by Eqs. (A.4)–(A.8) and is a complex oscillatory function of the intensity (in the limit  $I \rightarrow 0$ , we have  $v_0^{(1)} \rightarrow 1$ ).<sup>2)</sup>

Substituting Eqs. (20) and (21) into Eqs. (24) and (25), and applying the well-known expansion

$$\exp(iz \sin \varphi) = \sum_{n=-\infty}^{\infty} J_n(z) \exp(in\varphi), \quad (26)$$

where  $J_n(z)$  is a Bessel function, we can obtain expressions for the  $n$ -photon absorption coefficients, i.e., the contribution made to the absorption coefficient by the processes in which an electron energy changes by  $n\hbar\omega$  and which in the

expression for  $W_{21}$  correspond to the terms containing the  $\delta$  function with the argument

$$\Delta_n = E_{1k} - E_{2k} + 1/2 \hbar\omega \xi_0 - n\hbar\omega.$$

It is convenient to represent  $K^{(n)}$  in a form analogous to Eq. (11):

$$K^{(n)} = \frac{2\pi}{\hbar} \frac{n\hbar\omega}{I} \rho(\tilde{n}\hbar\omega) F(\beta, \tilde{n}, \omega) |v_0^{(1)} \tilde{M}_{\nu_0, \nu_0}^{(n)}|^2, \quad (27)$$

where  $\tilde{M}_{\nu_0, \nu_0}^{(n)}$  is the effective matrix element of  $n$ -photon absorption:

$$\tilde{M}_{\nu_0, \nu_0}^{(n)} = \hbar\omega \sum_{k=-\infty}^{\infty} \xi_k \sum_{\{r_{j,s}\}} \prod_{j=1}^n J_{r_{j,s}}(j^{-1} \xi_j), \quad (28)$$

and  $\sum_{\{r_{j,s}\}}$  denotes summation over various sets of integers  $\{r_{j,s}\}$ , satisfying the condition

$$\sum_j jr_{j,s} = n - k.$$

In contrast to Eq. (11), Eq. (27) includes only the  $\pm 1/2 \rightarrow \pm 3/2$  transitions, contains a factor  $|v_0^{(1)}|^2$ , and the arguments of the functions  $\rho$  and  $F$  contain now in place of  $n$  the quantity

$$\tilde{n} = \frac{1}{\hbar\omega} (E_{1\tilde{k}_n} - E_{2\tilde{k}_n}), \quad (29)$$

where  $\hbar\mathbf{k}_n$  is a quasimomentum satisfying the law of conservation of energy  $\Delta_n = 0$ , which together with Eq. (29) can also be represented in the form

$$\Delta_n = \tilde{n} - n = -1/2 \xi_0 [u_{\pm}(\tilde{n})]. \quad (30)$$

In  $n \geq 2$  and the intensity of light is low, the quantity  $1/2 \xi_0 \hbar\omega$  reduces exactly to the difference, which is linear in  $I$ , between the Stark shifts of the bands. If  $|u_{\pm}| \gtrsim 1$ , then  $\Delta_n$  becomes a complex nonmonotonic function of  $I$ . The quantities  $\tilde{n}(n)$  are found by numerical solution of the transcendental equation (30). In the investigated range of intensities the values of  $|\Delta_n/n|$  reach  $(2-3) \times 10^{-1}$ .

If  $n = 1$ , an analysis (we shall omit its details) shows that for  $\eta_n \ll 1$  the splitting of the spectrum of the light wave is linear in the field intensity:  $\Delta_n \approx \pm 0.7147 |M_{3/2, 1/2}^{(1)}| / \hbar\omega$ . The similarity of this result to that obtained in the resonance approximation<sup>10</sup> ( $\Delta_{nR} = \pm |M_{3/2, 1/2}^{(1)}| / \hbar\omega$ ) is evidence that the method provides a good approximation for the contributions of higher orders (in respect of the field) to the band energy.

The above analysis is valid in the case when the nonlinear absorption can be described in terms of the probabilities of  $n$ -photon intersubband transitions per unit time, which is true if

$$|M_{12}^{(n)}| |\tau_p(1, \tilde{\mathbf{k}}_n) \tau_p(2, \tilde{\mathbf{k}}_n)|^{1/2} / \hbar \ll 1, \quad \omega \tau_p(l, \tilde{\mathbf{k}}_n) \gg 1, \quad (31)$$

where  $\tau_p(l, \mathbf{k})$  is the relaxation time of the carrier momentum in the subband  $l$ . The inequalities of Eq. (31) lead also to the inequality

$$\kappa^{(n)} = |M_{12}^{(n)}| / \hbar\omega \ll 1. \quad (32)$$

A contribution to the  $n$ -photon absorption coefficient of Eq.

(27) is made not only by the process of the  $n$ th but also of higher  $(n + 2m)$ th orders when  $(n + m)$  photons are absorbed and  $m$  photons are emitted in different sequences in each elementary event. These different  $n$ -photon absorption channels interfere and partly suppress one another. It should be noted that in a fixed order of perturbation theory the interference of various channels is very strong. For example, the composite matrix element  $M_{m'm}^{(3)}$  calculated from perturbation theory is an order of magnitude less than a typical contribution of each of the channels. For similar reasons the quantities  $\kappa^{(n)}$  in Eq. (32) are small not only compared with the product of  $n$  factors of the type  $M_{m'm}^{(1)}/\hbar\omega$ , contained in the composite matrix element in the  $n$ th order of perturbation theory, but also compared with unit. Numerical calculations carried out for Ge when  $\hbar\omega = 13.7$  meV indicate that the values of  $\kappa^{(n)}$  do not exceed  $\sim 10^{-1}$ . Estimates of the relaxation time  $\tau_p(l, k)$  indicate that when the ionized acceptor concentration exceeds  $10^{16}$  cm $^{-3}$ , the inequalities of Eq. (31) are also satisfied. Therefore, the absorption of submillimeter radiation is due to a set of simultaneous  $n$ -photon interband transitions with different values of  $n$ . Figure 2a shows the dependences of  $K^{(n)}/F(\beta, \tilde{n}, \omega)$  on  $I_0$  calculated

for  $n = 2, 3, 4$ , and  $5$  on a computer using Eqs. (27), (38), and (A.6)–(A.14) for  $p$ -type Ge and  $\hbar\omega = 13.7$  meV. We can see that already for  $I_0 \sim 1$ – $2$  MW/cm $^2$  the coefficient  $K^{(n)}$  is a complex nonmonotonic function of the intensity, which differs strongly from the power law  $I_0^{n-1}$ . For  $I_0 = 2$  MW/cm $^2$  a significant contribution (representing a few percent) to the total absorption coefficient comes from the transitions with  $n = 7$ , whereas when  $I_0 = 5$  MW/cm $^2$ , a significant contribution comes also from 11-photon intersubband transitions.

As pointed out already, the approach developed in the present section can describe formally also the case of one-photon absorption. However, if  $n = 1$ , the condition (31) is violated at intensities lower than those at which higher-order effects described by Bessel functions can be observed. A consistent analysis of a one-photon resonance in this situation represents a problem on its own which is very difficult to solve. However, it should be pointed out that when  $I_0$  is increased, the contribution of  $K^{(1)}$  to the total absorption coefficient can only decrease,<sup>3)</sup> enhancing the role of many-photon processes.

Figure 2b gives the total absorption coefficient

$$K = \sum_n K^{(n)}$$

as a function of the intensity of light incident on a crystal for different dependences of the effective hole temperature  $T_h$  on  $I_0$  when this temperature differs from the lattice temperatures  $T_{\text{latt}}$  for  $I_0 \neq 0$  (for all the curves the lattice temperature is  $T_{\text{latt}} = 78$  K). We can see that by itself an increase in  $T_h$  can only reduce the absorption, i.e., it can mask the contribution of many-photon transitions. However, variation within reasonable limits of the function

$$\Delta T(I_0) = T_h - T_{\text{latt}} \quad (33)$$

does not alter significantly the  $K(I_0)$  dependence. We can also see from Fig. 2b that for  $I_0 < 1$  MW/cm $^2$  the value of  $K$  rises linearly on increase in the intensity. In this range the slowing down of the rise of  $K^{(2)}$  (compared with the linear increase) due to processes of higher orders is compensated by "enhancement" of the three- and four-photon transitions. In the range from 1 to 2 MW/cm $^2$  there is a flat region in the dependence of  $K$  on  $I_0$ . Here,  $K^{(2)}$  still makes a major contribution to  $K$ , but its value decreases (Fig. 2a). At high intensities the two-photon absorption process plays a relatively minor role and the probabilities of  $n$ -photon transitions that dominate  $K$  continue to rise on increase in  $I_0$ . Moreover, in view of the considerable "pushing apart" of the energy bands in the radiation field, the positions of the  $\tilde{k}_n$  points of an  $n$ -photon resonance shift toward the ceiling of the valence band, where the density of holes in the initial states is higher. All this results in a new rise of the dependence  $K(I_0)$ .

We shall now compare the theory and experiment. In the measurements carried out on samples of  $n$ - and  $p$ -type Ge at temperatures  $T = 78$  and 300 K use was made of a high-power pulses NH $_3$  laser pumped optically with CO $_2$  TEA laser radiation.<sup>2,3</sup> The duration of the radiation pulses with the wavelength  $\lambda = 90.55 \mu$  was 40 nsec. The dependences of

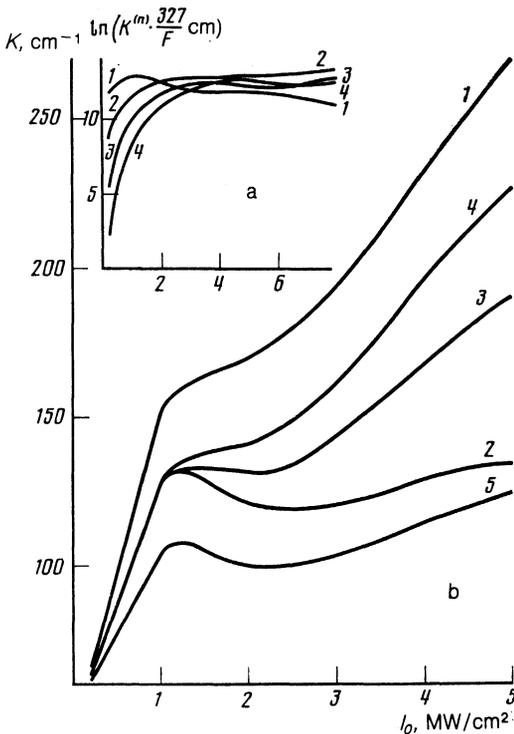


FIG. 2. Dependences of the  $n$ -photon absorption coefficients (a) and of the total absorption coefficient (b) for transitions between the light- and heavy-hole subbands in  $p$ -type Ge on the intensity of the incident light  $I_0$ . The values of  $\hbar\omega$ ,  $n_\omega$ , and  $m_l$  used in the calculations are given in the text (Sec. 2). a) Curves 1, 2, 3, and 4 correspond to  $n = 2, 3, 4$ , and  $5$ , respectively; the ordinate gives the values of  $\ln[K^{(n)} \times 327 \text{ cm}/F(\beta, \tilde{n}, \omega)]$ . b) Curves 1–5 correspond to different dependences of  $\Delta T$  on  $I_0$ : 1)  $\Delta T = 0$ ; 2)–4)  $\Delta T = 20I_0$  K  $\cdot$  cm $^2 \cdot$  MW $^{-1}$  for  $I_0 < 1$  MW/cm $^2$ . In the range  $I_0 > 1$  MW/cm $^2$  the dependences are as follows: 2)  $\Delta T \propto I_0$ ; 3)  $\Delta T \propto I_0^{1/2}$ ; 4)  $\Delta T = 20$  K. For curve 5 we have  $\Delta T = 50I_0^{1/2}$  K  $\cdot$  cm  $\cdot$  MW $^{-1/2}$  for all the values of  $I_0$ . Hole density  $p = 5.8 \times 10^{16}$  cm $^{-3}$ .

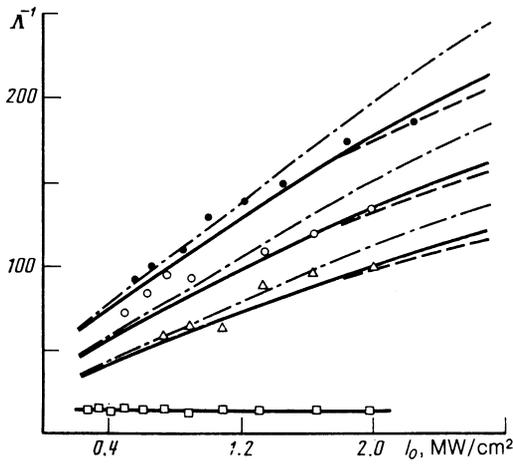


FIG. 3. Dependences of the reciprocal of the transmission on the intensity of the incident light  $I_0$  at  $T = 78$  K: (●, ○, △) results of measurements on  $p$ -type Ge samples ( $p = 5.8 \times 10^{16} \text{ cm}^{-3}$ ) of thicknesses  $d = 0.0695$ ,  $0.064$ , and  $0.058$  cm, respectively; (□)  $n$ -type Ge ( $3.5 \times 10^{16} \text{ cm}^{-3}$ ,  $d = 0.17$  cm). The theoretical dependences for each of the thicknesses of the  $p$ -type samples are as follows: the continuous, dashed, and dash-dot curves correspond to curves 4, 2, and 1 in Fig. 2b.

the reciprocal of the transmission  $\Lambda^{-1}$  of Ge samples on the intensity of light were determined employing a fast-response ( $\sim 10^{-9}$  sec) detector based on the drag of electrons by photons<sup>12</sup> and made of  $n$ -type Ge. It was found that operation of this detector was linear throughout the investigated range of intensities both at  $T = 78$  K and 300 K. Calibrated polyethylene filters were used as attenuators.

Figure 3 shows the dependences of the reciprocal of the transmission  $\Lambda^{-1}(I_0) = I_0/I(d)$  on the intensity  $I_0$  of the radiation incident at  $T = 78$  K on crystals with a hole density  $p = 5.8 \times 10^{16} \text{ cm}^{-3}$  and with different thicknesses  $d$  [ $I(d)$  is the intensity of the radiation transmitted by a crystal]. An increase in  $I_0$  resulted in darkening of the samples and the dependence of  $\Lambda^{-1}$  on  $I_0$  was nearly linear. Clearly, the value of  $\Lambda^{-1}$  at the maximum intensity  $I_0$  was several times higher than  $\Lambda^{-1}$  at relatively low values of  $I_0$ . The mechanism responsible for this nonlinearity was identified by investigating also the transmission as a function of  $I_0$  using  $n$ -type Ge samples at  $T = 78$  K and 300 K and  $p$ -type Ge at  $T = 300$  K. Throughout the investigated range  $I_0 \leq 2 \text{ MW/cm}^2$  there was no linearity in the investigated cases. These experiments indicated that the nonlinearity observed for  $p$ -type Ge at  $T = 78$  K was due to direct intraband transitions between the subbands  $v_1$  and  $v_2$ . We shall show later that at the wavelength  $\lambda = 90.55 \mu$  used in our experiments the absorption in  $n$ -type Ge ( $T = 300$  and  $78$  K) is governed by indirect intraband transitions, whereas in  $p$ -type Ge ( $T = 78$  K) the main role in the absorption is played by direct transitions in the  $v_1$  and  $v_2$  subbands.<sup>4)</sup>

We shall now identify the reason why the nonlinearity is not manifested in the absorption involving indirect transitions in the conduction and heavy-hole bands when  $I_0 \leq 2 \text{ MW/cm}^2$ . In such cases the nonlinearity parameter  $\eta_n$  is governed by the relatively large masses of electrons ( $m_e \approx 3m_2$ ) and of heavy holes ( $m_1 \approx 8m_2$ ), and not by the

mass of light holes  $m_2$ , as in the case of direct intersubband transitions. These estimates confirm that in the investigated range  $I_0$  the dependence  $\Lambda^{-1}(I_0)$  should not be manifested in practice for the indirect transitions.

We shall now consider in greater detail the problem of the dominant intraband absorption mechanisms in the case of Ge at low intensities when  $\eta_n \ll 1$ . We determined the absorption coefficients of  $n$ - and  $p$ -type germanium at  $T = 78$  and  $T = 300$  K with carrier densities from  $10^{13} \text{ cm}^{-3}$  to  $10^{16} \text{ cm}^{-3}$ . The result together with an allowance for multiphoton absorption at  $\lambda = 90.55 \mu$  (Ref. 13) were used to find the absorption cross sections in all four cases. In experiments on  $n$ -type Ge, for which the absorption was entirely due to indirect intraband transitions at the lattice, a relationship was found between the cross sections for the absorption of light by electrons in the case of indirect transitions  $\sigma_i(T)$  at two parameters:  $\sigma_i(300 \text{ K}) = 3.85\sigma_i(78 \text{ K})$ . In the case of  $p$ -type Ge the total absorption cross section was  $\sigma(T) = \sigma_i(T) + \sigma_d(T)$ , where  $\sigma_d(T)$  is the absorption cross section due to direct transitions. Bearing in mind that a similar temperature dependence  $\sigma_i(T)$  was exhibited by  $p$ -type Ge and using the dependence of the cross section for direct transitions  $\sigma_d(T)$  from Ref. 14, according to which  $\sigma_d(300 \text{ K}) \approx 0.07\sigma_d(78 \text{ K})$ , we obtained the relationship between the individual contributions to the total absorption cross section, which for  $p$ -type Ge at  $T = 78$  K was  $\sigma_d \approx 10\sigma_i$ , whereas at  $T = 300$  K it was  $\sigma_d \approx 0.17\sigma_i$ .

Figure 3 shows the theoretical dependences of the reciprocal of the transmission  $\Lambda^{-1}(I_0)$  calculated for samples of a given thickness using the  $K(I_0)$  plots shown in Fig. 2b. We can see that the experimental data are described satisfactorily by the results of a calculation of the nonlinear absorption representing a "fence" of simultaneous direct  $n$ -photon transitions for different values of  $n$  occurring at different points in  $k$  space between the subbands of light and heavy holes.

#### 4. DEPENDENCE OF THE DRAG CURRENT IN $p$ -TYPE Ge ON THE RADIATION INTENSITY

According to Ref. 4, the longitudinal current due to the drag of electrons by photons  $\mathbf{j} \parallel \mathbf{q}$  at the wavelength of  $\lambda = 90.55 \mu$  exhibits a reversal of its sign on increase in the radiation intensity. This can be explained by postulating a difference between the signs of the one-photon and  $n$ -photon ( $n \geq 2$ ) contributions to the photocurrent and bearing in mind that the  $n$ -photon contribution increases on increase in the radiation intensity. In the present section we shall calculate the dependence of the drag current on the intensity  $I_0$  in the case of  $p$ -type Ge at room temperature in the region of  $I_0 \sim I_0^{(0)} = 0.33 \text{ MW/cm}^2$ , where  $I_0^{(0)}$  is the intensity of the incident radiation at which the drag current changes its sign (Fig. 4). We note, as found experimentally in Ref. 15, that at room temperature and for  $\lambda = 90.55 \mu$  the main contribution to the drag current comes from direct transitions in spite of the fact that the absorption coefficient of light is then determined by indirect transitions. Therefore, we shall calculate the drag current allowing only for direct transitions. The value of  $I_0^{(0)}$  given above corresponds to the transmitted light intensity  $I^{(0)} = 0.21 \text{ MW/cm}^2$ ,  $\eta_2 = 0.13$ , and  $K^{(3)}/$

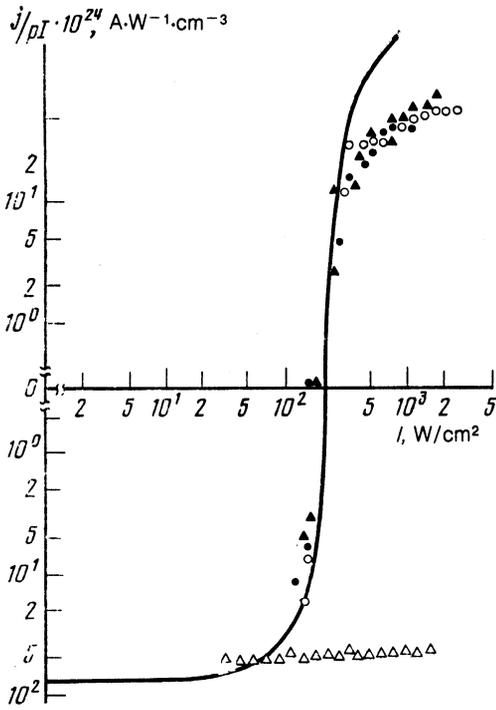


FIG. 4. Dependence of the drag current, normalized to the light intensity  $I$  and to the free carrier density  $p$ , on  $I$  at  $T = 300$  K: (●), (▲), (○), (Δ) experimental data for  $p$ -type and  $n$ -type Ge; (●)  $p = 3 \times 10^{14} \text{ cm}^{-3}$ ; (▲)  $p = 10^{15} \text{ cm}^{-3}$ ; (○)  $p = 10^{16} \text{ cm}^{-3}$ ; (Δ)  $n$ -type Ge,  $n = 3 \times 10^{15} \text{ cm}^{-3}$ . The continuous curve is theoretical and it applies to  $p = 10^{15} \text{ cm}^{-3}$ , with the current given by the sum  $j = j_1 + j_2 + j_s$ , where  $j_s = \text{const} = 1.1 \times 10^{-23} \text{ A} \cdot \text{cm}^3 \cdot \text{W}^{-1}$ .

$K^{(1)} = \eta_2 \eta_3 < 10^{-2}$ . Therefore, in the case under discussion ( $T = 300$  K,  $I \sim I^{(0)}$ ) it is sufficient to calculate in the lowest order of perturbation theory the one-photon and two-photon drag currents  $j = j^{(1)} + j^{(2)}$ , where  $j^{(1)} \propto I$  and  $j^{(2)} \propto I^2$ , and to show that the photocurrents  $j^{(1)}$  and  $j^{(2)}$  have opposite signs and compensate each other at the intensity  $I^{(0)}$ , although at this value of the intensity we have  $K^{(2)} \ll K^{(1)}$ . A calculation of the drag current in the case when  $\eta_2, \eta_3 \sim 1$  requires a separate study.

The expression for the density of the two-photon current  $j^{(2)}$  in the electron approximation, obtained using the electron representation and the relaxation time approximation is as follows:

$$j^{(2)} = -e \sum_{\mathbf{k}, \mathbf{m}', \mathbf{m}} [\mathbf{v}_{1\mathbf{k}} \tau_p(1, \mathbf{k}) w_{2\mathbf{m}, \mathbf{k}-2\mathbf{q}; 1\mathbf{m}', \mathbf{k}}^{(2)} - \mathbf{v}_{2\mathbf{k}} \tau_p(2, \mathbf{k}) w_{2\mathbf{m}, \mathbf{k}; 1\mathbf{m}', \mathbf{k}+2\mathbf{q}}^{(2)}], \quad (34)$$

where  $\mathbf{v}_{i\mathbf{k}} = -\hbar \mathbf{k} / m_i$ . The probability that the  $(v_{2\mathbf{m}, \mathbf{k}} - 2\mathbf{q}) \rightarrow (v_{1\mathbf{m}', \mathbf{k}})$  optical transition occurs in a unit time is given by the expression

$$w_{2\mathbf{m}, \mathbf{k}-2\mathbf{q}; 1\mathbf{m}', \mathbf{k}}^{(2)} = \frac{2\pi}{\hbar} |M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(2)}|^2 \exp[\beta(\chi + E_{i\mathbf{k}})] \times [1 - \exp(-2\beta\hbar\omega)] \delta(E_{1\mathbf{k}} - E_{2, \mathbf{k}-2\mathbf{q}} - 2\hbar\omega). \quad (35)$$

The transition probability  $w_{2\mathbf{m}, \mathbf{k}; 1\mathbf{m}', \mathbf{k}+2\mathbf{q}}^{(2)}$  is found in a similar manner. The matrix element  $M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(2)}$  is described by the formula

$$M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(2)} = \sum_{m_1} \frac{M_{\mathbf{m}', \mathbf{k}; m_1, \mathbf{k}-\mathbf{q}}^{(1)} M_{m_1, \mathbf{k}-\mathbf{q}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(1)}}{\hbar\omega + E_{m_1, \mathbf{k}-2\mathbf{q}} - E_{m_1, \mathbf{k}-\mathbf{q}}} + \left(\frac{eA_0}{c\hbar}\right)^2 H_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(2)}(e). \quad (36)$$

Using  $\mathbf{q}$ , we can calculate the matrix element of a one-photon transition from

$$M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-\mathbf{q}}^{(1)} = \frac{eA_0}{c\hbar} \hat{C}_{\mathbf{m}'\mathbf{k}}^+ \times \left[ e \frac{\partial}{\partial \mathbf{k}} H^{(2)}\left(\mathbf{k} - \frac{\mathbf{q}}{2}\right) + ig \frac{\hbar^2}{2m_0} [\mathbf{q}e] \mathbf{J} \right] \hat{C}_{\mathbf{m}, \mathbf{k}-\mathbf{q}}, \quad (37)$$

where  $g$  denotes the  $g$  factor of an electron in the band  $\Gamma_8^+$  considered in the spherical approximation. In the calculation of the components of the matrix elements  $M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-\mathbf{q}}^{(1)}$  and  $H_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-2\mathbf{q}}^{(2)}$  linear in respect of  $\mathbf{q}$  it is convenient to introduce a coordinate system with  $\mathbf{z} \parallel \mathbf{q}$  and to apply the formula

$$\frac{\partial}{\partial k_z} \hat{C}_{\mathbf{m}\mathbf{k}} = \frac{i}{k} \sum_{m'} [J_{x, m'm} \cos(\widehat{z, y'}) - J_{y, m'm} \cos(\widehat{z, x'})] \hat{C}_{m'\mathbf{k}}, \quad (38)$$

where  $(\widehat{\alpha, \beta'})$  is the angle between the  $\alpha$  and  $\beta'$  axes;  $J_{\alpha, m'm}$  are the components of the matrix  $J_\alpha$  in the fixed basis of the  $\Gamma_8^+$  band. Then the quantities  $\hat{C}_{\mathbf{m}\mathbf{k}}$  [the proper columns of the matrix  $H^{(2)}(\mathbf{k})$ ] satisfy the relationships

$$(\mathbf{J}\mathbf{k}) \hat{C}_{\mathbf{m}\mathbf{k}} = m\mathbf{k} \hat{C}_{\mathbf{m}\mathbf{k}}, \quad \hat{C}_{\mathbf{m}'\mathbf{k}}^+ \left( \sum_{\alpha} J_{\alpha} \cos(\widehat{\alpha, \beta'}) \right) \hat{C}_{\mathbf{m}\mathbf{k}} = J_{\beta, m'm}.$$

The contribution of free carriers in the  $v_l$  subband to the density of the drag current under  $n$ -photon absorption conditions can be rewritten for convenience in the form

$$j_l^{(n)} = e \frac{\hbar q}{m_1 - m_2} \tau_p(l, E_l^{(n)}) (-1)^{l+1} \frac{K^{(n)} I}{\hbar\omega} (u_{n,l} + u'_{n,l}), \quad (39)$$

where  $E_l^{(n)} = n\hbar\omega\mu/m_l$ . The quantity  $u_{n,l}$  describes the drag effect when the dependence of the matrix element  $M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-n\mathbf{q}}^{(n)}$  on  $\mathbf{q}$  is ignored. The following formula describes this quantity:

$$u_{n,l} = a_n \left\{ b_n + \frac{2}{2n+3} \left[ \left( \frac{\partial \ln \tau_p(l, E)}{\partial \ln E} \right)_{E=E_l^{(n)}} - \beta E_l^{(n)} \right] \right\}, \quad (40)$$

We shall give the values of  $a_n$  and  $b_n$  for  $n = 1$  and  $2$ :  $a_1 = 2$ ,  $b_1 = 1$ ;  $a_2 = 21/5$ ,  $b_2 = 53/49$ . The term  $u'_{n,l}$  appears on the right-hand side of Eq. (39) because the matrix element  $M_{\mathbf{m}', \mathbf{k}; \mathbf{m}, \mathbf{k}-n\mathbf{q}}^{(n)}$  depends on  $\mathbf{q}$ . We shall write down the explicit expressions for  $u'_{n,l}$  for  $N = 1$  and  $n = 2$ :

$$u'_{1,1} = -\frac{1}{20} \left( 3 - 7 \frac{m_2}{m_1} + 5 \frac{m_2}{\mu} \gamma \right), \quad u'_{1,2} = \frac{1}{20} \frac{m_1}{m_2} \left( 7 - 3 \frac{m_2}{m_1} - 5 \frac{m_2}{\mu} \gamma \right), \quad (41)$$

$$u_{2,1} = -\frac{1}{700} \frac{m_2}{\mu} \left[ 9 \left( 76 + 71 \frac{m_2}{m_1 - m_2} + 14\gamma \right) + 10(12 - 14\gamma) \right], \quad (42)$$

$$u'_{2,2} = \frac{1}{700} \frac{m_1}{\mu} \left[ -9 \left( 18 + 54 \frac{m_2}{m_1 - m_2} + 14\gamma \right) + 10(8 - 14\gamma) \right],$$

where  $\gamma = g\hbar^2/2m_0B = -g\mu/m_0$  and  $m_0$  is the mass of a free electron. The first term in the square brackets of Eq. (42) corresponds to the transitions  $\pm 1/2 \rightarrow \pm 3/2$  and the second terms to the transitions  $\pm 1/2 \rightarrow \mp 3/2$ . The first calculations of the drag current in the case of optical transitions between the heavy- and light-hole subbands were reported in Ref. 16. For  $\gamma = 0$ , Eqs. (39)–(41) are identical with the results obtained in Ref. 17, where the one-photon drag of free carriers was calculated for the first time with allowance for the dependence of  $M_{m',k;m,k-q}^{(1)}$  on  $\mathbf{q}$ . The terms (41) and (42) proportional to the coefficient  $\gamma$  are related to the contribution made to the matrix element by an optical transition in which the magnetic field of a light wave interacts with the angular momentum of electrons in the energy band  $\Gamma_8^+$  [as represented by the second term in brackets of Eq. (37)]. The need to allow for this contribution to the one-photon drag current was first pointed out in Ref. 18. The two-photon drag effect in the band  $\Gamma_8^+$  was discussed for the first time theoretically in Ref. 19, but no allowance was made there for the second term on the right-hand side of Eq. (37) or for the dependence of the matrix element  $M_{m',k;m,k-q}^{(1)}$  on  $\mathbf{q}$ .

The theoretical dependence of the drag current on the light intensity presented in Fig. 4 was calculated allowing for three mechanisms of the dissipation of the momentum of holes: by the scattering on acoustic phonons,<sup>20,21</sup> on optical phonons,<sup>14,22</sup> and on ionized impurities.<sup>20</sup> We selected the following values of the parameters of Ge:  $m_1 = 0.33m_0$ ,  $m_2 = 0.045m_0$ ,  $g = -6.8$ , acoustic length  $l_0 = 4.3 \times 10^{-3}$  cm, velocity of sound  $s = 5.2 \times 10^5$  cm/sec, density  $\rho = 5.3$  g/cm<sup>3</sup>, interaction constant  $E_{\text{opt}} = 11.4$  eV,  $\hbar\omega_{\text{opt}} = 37$  meV, static permittivity  $\epsilon_0 = 16$ . A calculation indicated that the relative contribution to  $\tau_p^{-1}$  by the scattering of holes on impurities is small in the investigated range of the impurity concentrations  $N_i$  and an increase from  $N_i = 3 \times 10^{14}$  cm<sup>-3</sup> to  $N_i = 10^{16}$  cm<sup>-3</sup> shifts the theoretical curve in the region of  $I \sim I^{(0)}$  by just 50 kW/cm<sup>2</sup> (the continuous curve in Fig. 4 corresponds to  $N_i = 10^{15}$  cm<sup>-3</sup>). It is clear from Fig. 4 that the agreement between the theory and experiment is good.

It should be noted that the equality  $j^{(1)} = -j^{(2)}$  and the inequality  $K^{(2)} \ll K^{(1)}$  are satisfied simultaneously when  $I = I^{(0)}$  because of the strong mutual compensation of the contributions of the heavy and light holes to  $j^{(1)}$ , whereas in the case of the two-photon drag current there is no such compensation ( $j_1^{(2)} \gg j_2^{(2)}$ ).

We have thus demonstrated that when a crystal of  $p$ -type Ge is excited by high-power laser radiation of submillimeter wavelengths, comparable contributions to the observed optical effects are made by  $n$ -photon transitions with different values of  $n$ .

## APPENDIX

The transformation of Eq. (16) gives, after some additional simple calculations, the following expressions for the matrix elements of the Hamiltonian:

$$\begin{aligned} \hat{H}_{11} = & (H^{(2)}(\mathbf{k}))_{11} + \frac{1}{2} (H_{11}^{\text{int}} + H_{22}^{\text{int}}) + \frac{1}{2} (H_{11}^{\text{int}} - H_{22}^{\text{int}}) f_1(z) \\ & + [H_1^{\text{int}}, \hat{S}]_{11} f_4(z) + [H_2^{\text{int}}, \hat{S}]_{11} f_2(z), \end{aligned} \quad (\text{A.1})$$

$$\begin{aligned} \hat{H}_{12} = & [H_2^{\text{int}}, \hat{S}]_{12} f_2(z) + [[H_1^{\text{int}}, \hat{S}], \hat{S}]_{12} f_3(z) \\ & + (H_2^{\text{int}})_{12} \frac{1+f_1(z)}{2} - 2S_{12}^2 (H_2^{\text{int}})_{21} f_5(z). \end{aligned} \quad (\text{A.2})$$

The transformation of the projection operation (23) gives

$$\tilde{\mathbf{P}} = \left\| \begin{array}{cc} \frac{1}{2}[1-f_1(z)] & -f_2(z)S_{12} \\ f_2(z)S_{21} & \frac{1}{2}[1+f_1(z)] \end{array} \right\|. \quad (\text{A.3})$$

The following notation is used in Eqs. (A.1)–(A.3):

$$\begin{aligned} f_i(z) = & z^{i-2} j_{i-2}(z) \quad |_{i=1, 2, 3}, \\ f_4(z) = & -[z^{-2} + j_{-2}(z)], \quad f_5 = f_4 - f_2, \end{aligned} \quad (\text{A.4})$$

where  $j_i(z)$  are spherical Bessel functions<sup>23</sup> and

$$z = 2(-S_{12}S_{21})^{1/2} = 2(|u_-|^2 + |u_+|^2 + 2u_-u_+^* \cos 2\omega t)^{1/2}. \quad (\text{A.5})$$

Using the Gegenbauer addition theorem<sup>23</sup> and carrying out some very time-consuming calculations, we find that

$$f_l = v_0^{(l)} + 2 \sum_{p=1}^{\infty} v_p^{(l)} \cos 2p\omega t, \quad (\text{A.6})$$

$$\begin{aligned} v_p^{(l)} = & \frac{(-1)^p}{\pi} (u_+ u_-)^{2-l} \\ & \times \sum_{m=0}^{\infty} v_{pm}^{(l)} j_{p+2m-2+l}(2u_-) j_{p+2m-2+l}(2u_+), \end{aligned} \quad (\text{A.7})$$

$$\begin{aligned} v_{pm}^{(l)} = & (2p+4m-2+l) \text{sign}(l-3/2) \\ & \times \frac{\Gamma(m-3/2+l) \Gamma(m+p-3/2+l)}{\Gamma(m+1) \Gamma(m+p+1)} \quad (l=1, 2, 3) \end{aligned} \quad (\text{A.8})$$

$$\begin{aligned} v_p^{(4)} = & \frac{(-1)^p}{2} \sum_{m=0}^{\infty} \frac{(2u_+^* u_-)^{p+2m}}{m!(p+m)!(2p+4m-1)!!} \\ & \times {}_1F_2 \left( \begin{array}{c} p+2m \\ p+2m+2, p+2m+1/2 \end{array} \middle| u_+^2 + u_-^2 \right), \end{aligned} \quad (\text{A.9})$$

where  $\Gamma(a)$  is the Euler gamma function,  ${}_1F_2(|)$  is the generalized hypergeometric function.<sup>24</sup> Figure 5 shows typical dependences  $v_p^{(l)}(u_+, u_-)$ .

We shall introduce the notation

$$V_{12} = M_{1/2, 1/2}^{(4)} (\hbar\omega)^{-1} = -\sqrt{\frac{3}{2}} e_- (\tilde{n}\epsilon')^{1/2},$$

$$V_1 = 2(M_{3/2, 3/2}^{(4)} - M_{1/2, 1/2}^{(4)}) (\hbar\omega)^{-1} = 4\sqrt{2} e_z (\tilde{n}\epsilon')^{1/2},$$

$$\begin{aligned} V_2 = & 2 \left( \frac{eA_0}{c\hbar} \right)^2 [H_{3/2, \tilde{k}_n; 3/2, \tilde{k}_n}^{(2)}(\mathbf{e}) - H_{1/2, \tilde{k}_n; 1/2, \tilde{k}_n}^{(2)}(\mathbf{e})] (\hbar\omega)^{-1} \\ = & 2(1 - 3e_z^2) \epsilon', \end{aligned} \quad (\text{A.10})$$

$$V_{12}^{(2)} = 2 \left( \frac{eA_0}{c\hbar} \right)^2 H_{3/2, \tilde{k}_n; 1/2, \tilde{k}_n}^{(2)}(\mathbf{e}) (\hbar\omega)^{-1} = 2\sqrt{3} e_z e_- \epsilon',$$

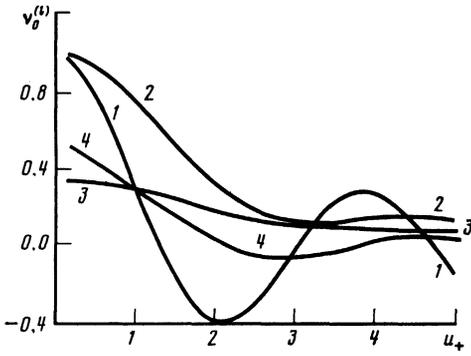


FIG. 5. Functions  $v_0^{(l)}(u_+, 9u_+/11)$ . Curves 1–4 correspond to  $l = 1, 2, 3$ , and 4, respectively.

$$\varepsilon' = \varepsilon(\hbar\omega)^{-1}, \quad e_{\pm} = e_{x'} \pm ie_{y'},$$

where  $\varepsilon$  is given by Eq. (15). In Eq. (A.10) an allowance is made for the law of conservation in the case of  $n$ -photon transitions [Eq. (30)]. Equations (A.1)–(A.6) and (A.10) yield the following expressions for the functions  $\xi_p$  and  $\zeta_p$  which occur in Eqs. (20), (21), and (28):

$$\xi_{2p} = 8s(\tilde{n}) |V_{12}|^2 v_{p-1,2}^{4,0} - V_2 v_{p-1,2}^{1,0}, \quad (\text{A.11})$$

$$\xi_{2p+1} = V_1 v_{p,1}^{1,0} - 4V_{12} V_{21}^{(2)} s(\tilde{n}) v_{p-1,3}^{2,0}, \quad (\text{A.12})$$

$$\begin{aligned} \zeta_{2p} = & \frac{1}{4} V_{12} V_{1s}(\tilde{n}) v_{p-1,2}^{2,1} \\ & + \frac{1}{4} V_{12}^{(2)} v_{p-1,2}^{2,0} - 2V_{12}^{(2)} |V_{12}|^2 v_{p-1,4}^{5,2} s^2(\tilde{n}), \end{aligned} \quad (\text{A.13})$$

$$\zeta_{2p+1} = V_{12} V_2 s(\tilde{n}) v_{p-1,2}^{2,1} - 4 |V_{12}|^2 V_{12} s^2(\tilde{n}) v_{p-1,3}^{3,1}, \quad (\text{A.14})$$

$$s(n) = \frac{n}{n^2 - 1}, \quad v_{l,j}^{i,t} = \sum_{h=0}^j \binom{j}{h} v_{l+h}^{(t)} g_{jh}^i, \quad (\text{A.15})$$

$$v_{l,j}^{i,0} = v_{l,j}^{i,0} + \sum_{h=0}^j \binom{j}{h} \delta_{p,l+h}. \quad (\text{A.16})$$

For all values of  $j$  and  $k$ , we have  $g_{jk}^0 = 1$ . The other coefficients  $g_{jk}^i$  are given by

$$\begin{aligned} g_{2k}^1 |_{h=1,2,3} = & (1 + \tilde{n}^{-1}, 1, 1 - \tilde{n}^{-1}), \\ g_{3k}^1 |_{h=1-4} = & (1 + \tilde{n}^{-1}, 1 + (3\tilde{n})^{-1}, 1 - (3\tilde{n})^{-1}, 1 - \tilde{n}^{-1}), \\ g_{4k}^2 |_{h=1-5} = & ((1 + \tilde{n}^{-1})^2, 1 + \tilde{n}^{-1}, 1 - (3\tilde{n}^2)^{-1}, 1 - \tilde{n}^{-1}, (1 - \tilde{n}^{-1})^2). \end{aligned} \quad (\text{A.17})$$

<sup>11</sup>The same approximation was used by the authors in Ref. 3. Although a qualitatively correct description of many-photon absorption was then obtained for interband transitions in  $p$ -type Ge, the quantitative results

of a more detailed analysis given below differ considerably from those in Ref. 3.

<sup>2</sup>The factor  $|v_0^{(1)}|^2$ , which occurs in Eq. (25) because of the transformation of the projection operator given by Eq. (23), ensures (when a simultaneous allowance is made for the Stark band shifts) that for  $n = 1$  and low values of  $I$  we go over to the "golden mean rule" of quantum mechanics.

<sup>3</sup>A reduction in the contribution  $K^{(1)}$  is due to the bleaching effect<sup>11</sup> characterized by the saturation parameter  $I_s$  and due to an increase in the temperature of holes. In the case under discussion the two factors lower  $K^{(1)}$ .

<sup>4</sup>Since under the experimental conditions we have  $\hbar\omega \approx E_g/65$ , in the investigated range of intensities  $I_0$  the transitions between the valence and conduction bands are quite unimportant, as demonstrated by estimates in Refs. 7–9.

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