Multiphoton interband absorption with participation of free carriers in crystals

E. Yu. Perlin, A. V. Fedorov, and M. B. Kashevnik

(Submitted 17 February 1983; resubmitted 4 May 1983)
Zh. Eksp. Teor. Fiz. **85**, 1357–1365 (October 1983)

Expressions are obtained for the probabilities of l-photon interband transitions ($l = 2$ and 3) under conditions when the energy of l photons is less than the forbidden band width and the energy deficit is covered by free carriers that are nonequilibrium by virtue of the action of an intense pump. The obtained dependences of the effect on the light intensity, polarization (linear or circular), dopant, and energy deficit are in qualitative agreement with the experimental data.

PACS numbers: 78.40. – q

1. Many known processes produce in semiconductors or dielectrics nonequilibrium electron-hole pairs following nonlinear absorption of intense light with quantum energy $\hbar\omega$ less than the forbidden-band width E_g. These include, besides cascade transitions via intermediate local levels in the forbidden band, also multiphoton transitions (MPT). Just as ordinary single-photon transitions, MPT can be direct[1–4] or indirect, e.g., with participation of phonons[5–7] or free carriers. An important difference between indirect MPT and analogous single-photon processes is that the high-power radiation that generates the MPT alters simultaneously the distribution functions of the vibrational subsystem or of the free carriers. This manifests itself substantially both in the MPT amplitudes and in the dependences of the probability of the process on the pump intensity I, the temperature T, and others.

Assume that the material contains a sufficient number of free electrons in the conduction band or holes in the valence band (the calculations that follow show that densities $n, p \gtrsim 10^{16}\ \mathrm{cm}^{-3}$ are usually needed if the process considered here is to predominate over other nonequilibrium-carrier generation processes).

Let l photons satisfy also the condition $0 < \Delta = E_g - l\hbar\omega < \hbar\omega$. The free carriers (we refer for the sake of argument to electrons unless otherwise stipulated) can, after giving up part of their kinetic energy E to cover the deficit Δ, participate together with the l photons in the production of an electron-hole pair (Fig. 1). It follows from energy and quasimomentum conservation that the process can take place only if $E > E_{\min}$. For $E_{\min}$ we easily obtain in the parabolic-band approximation

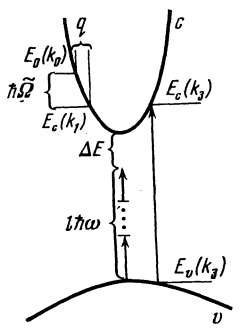

FIG. 1. Band scheme of MPT with participation of free electrons.

$$E_{min} = \frac{2\gamma+1}{\gamma+1}\Delta, \quad \gamma = \frac{m_c}{m_v}. \tag{1}$$

A similar process in single-photon absorption was investigated in Refs. 8 and 9.

Mention can be made of several situations in which the mechanism of interband MPT with participation of free carriers is important: a) The material is so doped that a sufficient number of free carriers is present even before the high-power pump is turned on—this situation is realized in the experiments of Ref. 10, and the qualitative features of the observed effect agree with the results of the theory expounded here (see Sec. 4). b) Free carriers are produced via impurity-band transitions induced by the pump radiation. c) The carriers are produced in interband $(l+1)$-photon absorption. The last situation is realized at pre-breakdown pump intensities, when free-carrier densities $\sim 10^{18}$–$10^{19}\ \mathrm{cm}^{-1}$ are produced (see, e.g., Ref. 11). It is clear therefore that even below the damage threshold and prior to the start of the "usual" avalanche lattice ionization[12] direct MPT can give way to the proposed process with participation of free carriers.[1)]

The dependence of the nonequilibrium-carrier density δn on the pump intensity I is now different than for usual MPT with the same l. Indirect MPT are stimulated mainly by excited electrons from the high-energy tail of the transitions, since they can transfer momentum to the produced pair in smaller portions than equilibrium electrons:

$$q = |\mathbf{k}_0 - \mathbf{k}_1| \geqslant q_{min} = \frac{m_c\Delta}{\hbar^2 k_0} = \frac{\tilde{\Delta}}{2k_0} \tag{2}$$

($\hbar\mathbf{k}_0$ and $\hbar\mathbf{k}_1$ are the stimulating-electron momenta in the initial and final states, see Fig. 1). In the simplest case when $\delta n \ll n_0$ (n_0 is the equilibrium density of the electrons) and the carrier excitation is due to the intraband indirect pump absorption, we have $\delta n^{(l)} \propto I^{l+1}$ at $\Delta > T$. At $\Delta \lesssim T$, a noticeable contribution to the process can be made also by the equilibrium carriers. Then $\delta n^{(l)} \propto I^{\nu} (l < \nu < l+1)$. It will be shown in Sec. 4 that in semiconductors, if $l > 2$, the dependence of δn on I is stronger for circular polarization than for linear.

2. We represent the Hamiltonian of the electron-photon system in the form

$$H = H_e^0 + H_p^0 + H_{ep}' + H_{ep}'' + H_{ee}', \tag{3}$$

 0038-5646/83/100787-05$04.00

where

$$H_e^0=\sum_{\mathbf{k}i}E_i(\mathbf{k})\xi_{\mathbf{k}i}^+\xi_{\mathbf{k}i},\qquad H_p^0=\sum_{\varkappa}\hbar\omega_\varkappa C_\varkappa^+C_\varkappa, \tag{4}$$

$E_i(\mathbf{k})$ are the Bloch energies of the ith band; $\xi_{\mathbf{k}i}^+$ and $\xi_{\mathbf{k}i}$ are the Fermi creation and annihilation operators acting in the electron subsystem; $C_\varkappa^+$ and $C_\varkappa$ are the creation and annihilation operators for photons with wave vector $\varkappa$,

$$H_{ep}'=\sum_{\mathbf{k},i,j}\xi_{\mathbf{k}i}^+\xi_{\mathbf{k}j}(U_{ij,\mathbf{k}}C_\varkappa+U_{ji,\mathbf{k}}^+C_\varkappa^+),$$
$$U_{ij,\mathbf{k}}=-\frac{ie\hbar}{m}\left(\frac{2\pi\hbar}{V}\right)^{1/2}\frac{\langle\mathbf{k}i|\mathbf{e}\nabla|\mathbf{k}j\rangle}{[\omega_\varkappa\varepsilon_1(\omega_\varkappa)]^{1/2}}. \tag{5}$$

V is the normalization volume, $\varepsilon_1(\omega_\varkappa)$ is the high-frequency dielectric constant, and $\mathbf{e}$ is the unit vector of the light polarization. The part of the electron-photon interaction H_{ep}'' which is quadratic in the operators is hereafter disregarded. The electron-electron interaction operator is of the form

$$H_{ee}'=\sum\langle\mathbf{k}_0i,\mathbf{k}_3j|\hat V^{ee}|\mathbf{k}_1i',\mathbf{k}_2j'\rangle\xi_{\mathbf{k}_0i}^+\xi_{\mathbf{k}_3j}^+\xi_{\mathbf{k}_1i'}\xi_{\mathbf{k}_2j'}, \tag{6}$$

$$\langle\mathbf{k}_0i,\mathbf{k}_3j|\hat V^{ee}|\mathbf{k}_1i',\mathbf{k}_2j'\rangle=\frac{4\pi\delta_{\mathbf{k}_0+\mathbf{k}_3,\mathbf{k}_1+\mathbf{k}_2}}{\varepsilon_1(\mathbf{k}_0-\mathbf{k}_1,\widetilde\Omega)|\mathbf{k}_0-\mathbf{k}_1|^2V}\beta_{\mathbf{k}_0\mathbf{k}_1}^{ii'}\beta_{\mathbf{k}_3\mathbf{k}_2}^{jj'},$$

$$\beta_{\mathbf{k},\mathbf{k}-\mathbf{q}}^{ii'}=\frac{1}{V_0}\int u_{i\mathbf{k}}^*u_{i'\mathbf{k}-\mathbf{q}}d\mathbf{r}\approx\begin{cases}1, & i=i'\\ \beta(q), & i\neq i'\end{cases}, \tag{7}$$

where $\beta(q)=\hbar q(2m_rE_g)^{-1/2}\ll1$, $m_r^{-1}=m_c^{-1}+m_v^{-1}$, $u_{i\mathbf{k}}$ are the Bloch amplitudes, V_0 is the unit-cell volume, the summation is over $\mathbf{k}_0i$, $\mathbf{k}_1i'$, $\mathbf{k}_3j$, and $\mathbf{k}_2j'$, and the band indices i, i', j, and j' run over the values of v and c. The estimate (7) for the overlap integrals of the Bloch amplitudes is obtained within the framework of the $\mathbf{k}\cdot\mathbf{p}$ perturbation theory, with the aid of which it is also easy to verify that the ratio of the matrix elements H_{ep}' to the interband ones is proportional to $\beta(k_2)$ [or $\beta(k_2+q)$]. At the values $q\sim\widetilde\Delta k_0^{-1}$, $k_2\sim\widetilde\Delta^{1/2}$ which are of importance at large $\mathbf{k}_0$ we have

$$\beta(k_2)\sim(\Delta/E_g)^{1/2}\ll1,\quad \beta(q)/\beta(k_2)\sim k_0^{-1}\widetilde\Delta^{1/2}\ll1.$$

For the process considered by us, the frequency transfer in the argument of the longitudinal dielectric constant $\varepsilon_1(\mathbf{q},\widetilde\Omega)$ is $\Omega=\hbar^{-1}[E_c(\mathbf{k}_0)-E_c(\mathbf{k}_0-\mathbf{q})]$.

We express the probability of l-photon transition between the valence band v and the conduction band c, with participation of the free electrons in the band c, in the form

$$W_{ee}^{(l)}=\frac{2\pi}{\hbar}\sum f_{c\mathbf{k}_0}\langle\!\langle|M_{\mathrm d}^{(l)}+M_{\mathrm{exch}}^{(l)}|^2\rangle\!\rangle_{phot}\delta[E_c(\mathbf{k}_0)-E_c(\mathbf{k}_1)$$
$$-E_c(\mathbf{k}_2)-E_v(\mathbf{k}_3)-E_g+l\hbar\omega], \tag{8}$$

(the summation is over $\mathbf{k}_0$, $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$); here $M_{\mathrm d}^{(l)}$ and $M_{\mathrm{exch}}^{(l)}$ is a composite matrix element of the l-photon transition and is the sum of direct and exchange contributions; $f_{c\mathbf{k}_0}$ is the electron distribution function in the band c; $E_c(\mathbf{k})$ and $E_v(\mathbf{k})$ are positive energies reckoned from the edges of the bands v and c. No account is taken in (8) of the occupation of the final states $|c\mathbf{k}_1\rangle$, $|c\mathbf{k}_2\rangle$, and $|v\mathbf{k}_3\rangle$. $\langle\langle\ldots\rangle\rangle_{phot}$ denotes averaging over the states of the phonon subsystem. The expressions given below for W_{ee} were written out for the case of single-mode coherent radiation, described by the Glauber-Sudershan δ-like weighting function.[1] The dependences of the MPT probabilities on the radiation statistics are quite substantial. Thus, for example in the case of a random (Gaussian) source, the probabilities of l-photon transitions are $l!$ times larger than for a δ-like source.[14,15] We shall not dwell on this in detail, for in this respect the process considered here does not differ from ordinary MPT.

Since $M^{(l)}$ contains the Coulomb interaction (6), (7) and the frequency transfer is $\widetilde\Omega\sim\hbar^{-1}\Delta\gg qu_T$, the amplitude of the effect increases as Δ approaches the plasma frequency Ω_e of the electron gas in the band c, owing to the decrease of $\varepsilon_l(\mathbf{q},\widetilde\Omega)$. This plasma resonance can play a substantial role at densities $n\gtrsim10^{17}$ cm^{-3} and at $\Delta<10^{-2}$ eV. The pertinent calculation, which calls for substituting in (8) the function $\varepsilon_l(\mathbf{q},\widetilde\Omega)$ calculated with allowance for the electron distribution in the field of the strong pump, meets with great difficulties and is outside the scope of the present paper, where we confine ourselves to the case $\Delta>\hbar\Omega_e$.

3. We shall calculate the matrix elements $M^{(l)}$ in the $(l+1)$st order of perturbation theory (l orders in H_{ep}' and one in H_{ee}'. The expansion in powers of H_{ep}' is applicable at practically all pump intensities below the damage threshold, except for some special cases (single-photon band-band resonance[16,17] or impurity-band resonance,[18,19] as well as small $\hbar\omega\lesssim10^{-2}$ eV.[1-3]

For the electrons with $k_0\gg\widetilde\Delta^{1/2}$, which make the largest contribution to the amplitude of the process, the criterion for the applicability of the Born approximation in H_{ee}' takes the simple form

$$e^2m_c/\varepsilon_l\hbar^2k_0\ll1 \tag{9}$$

(the relative velocity of the colliding particles practically coincides in this case with the velocity of the initial electron). The condition (9) is valid at $\gamma\lesssim1$. The criterion has a somewhat simpler form at $\gamma\gg1$. We shall not dwell on this, since the case of greatest interest will be shown below to be that of small γ.

We confine ourselves for brevity to a situation typical of many materials, when a direct single-photon transition is allowed between the bands v and c. From among the set of diagrams of order $l+1$ we separate those which make the main contribution to $M^{(l)}$. Assuming next throughout that $\Delta\ll\hbar\omega$, we retain only the resonant diagrams, i.e., those whose contribution contains the factors $\sim\Delta^{-1}$. It is easy to verify that in these diagrams all the photon lines enter into one of the electron lines and there are no vertices with interband Coulomb interactions [the contributions that include the interband part H_{ee}' are also small in the parameter $\beta(q)$]. Allowance for the nonresonant diagrams would lead only to a correction $\sim(\Delta/\hbar\omega)^2$ since, first, the contribution of each such diagram differs by a factor $\sim\Delta/\hbar\omega$ from the resonant one, and second, the contributions of the nonresonant diagrams are completely canceled by the terms of opposite signs; this yields one more factor $\sim\Delta/\hbar\omega$.

Using the smallness of the parameter $\beta(k_2)$, we simplify further, retaining only the diagrams that contain the minimum number of intraband matrix elements of the operator H_{ep}' needed for the given process. If the pump is linearly

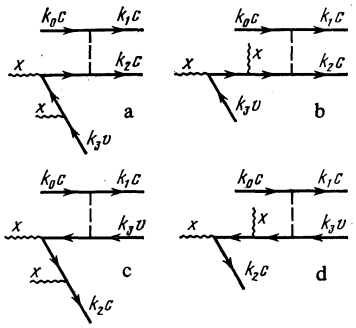

FIG. 2. Diagrams for $M^{(2)}_{d,L}$.

polarized, $M^{(2l+1)}$ contains in the indicated approximation only the interband matrix elements H'_{ep}, while $M^{(2l)}$ contains $2l-1$ interband and one intraband matrix elements $[M^{(2l)} \propto \beta(k_2)]$.

We consider now the cases $l=2$ and $l=3$, which are most frequently encountered in experimental investigations of MPT. The results can be easily generalized to include transitions with participation of an arbitrary number of photons. Figures 2 and 3 show the diagrams that make the main contribution to the amplitudes of two- and three-photon transitions. The corresponding expressions for the matrix elements are

$$M^{(2)}_{d\cdot L} = \frac{2\pi m\hbar\mu_2}{m_r\omega}[\mathbf{e}(\mathbf{k}_2-\mathbf{q})\Delta_2^{-1}(\mathbf{k}_2-\mathbf{q}) - \mathbf{e}\mathbf{k}_2\Delta_2^{-1}(\mathbf{k}_2)], \quad (10)$$

$$M^{(3)}_{d\cdot C} = \frac{1}{2}\frac{(2\pi)^{3/2}(\mathbf{e}\mathbf{p}_{cv})^2}{\hbar^{1/2}\omega^2}\mu_3[\Delta_3^{-1}(\mathbf{k}_2-\mathbf{q}) - \Delta_3^{-1}(\mathbf{k}_2)], \quad (11)$$

$$\mu_l = \frac{4\pi e^{l+2}\bar{n}_\varkappa(\mathbf{e}\mathbf{p}_{cv})}{Vm^l\omega^{l/2}\varepsilon_t^{l/2}(\omega)\varepsilon_l(\mathbf{q},\tilde{\Omega})\mathbf{q}^2},$$
$$\Delta_l(\mathbf{k}) = E_g + \frac{\hbar^2\mathbf{k}^2}{2m_r} - l\hbar\omega, \quad (12)$$

where $\bar{n}_\varkappa$ is the average photon-number density in the mode and $\mathbf{p}_{cv}$ are the interband matrix elements of the momentum operator.

We shall consider transitions induced by circularly polarized pumping for the case typical of cubic semiconductors, when the conduction and valence bands are made up of s-type and p-type functions. By virtue of the distinctive properties of the selection rules,[20,21] $M^{(l)}$ contains now only on interband matrix element H'_{ep} and $l-1$ intraband ones $(M^{(l)}_C \propto [\beta(k_2)]^{l-1})$. We confine ourselves here to the case $l=3$. The matrix elements $M^{(l)}_{d\cdot C}$ is determined by the diagrams of Fig. 4. We have

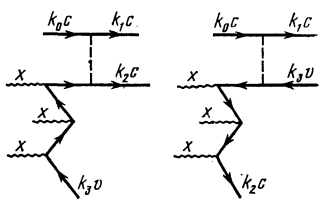

FIG. 3. Diagrams for $M^{(3)}_{d,L}$.

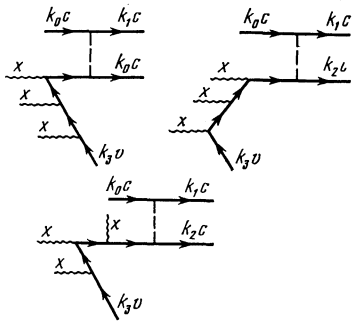

FIG. 4. Diagrams for $M^{(3)}_{d,C}$. Altogether there are six diagrams, three of which are obtained from those obtained in this figure in the same manner that diagrams c and d are obtained from a and b in Fig. 2.

$$M^{(3)}_{d\cdot C} = \frac{2(2\pi)^{3/2}\hbar m^2}{\omega^2 m_r^2}\mu_3[(\mathbf{e}(\mathbf{k}_2-\mathbf{q}))^2\Delta_3^{-1}(\mathbf{k}_2-\mathbf{q}) - (\mathbf{e}\mathbf{k}_2)^2\Delta_3^{-1}(\mathbf{k}_2)], \quad (13)$$

where $\mathbf{e} = 2^{-1/2}(\mathbf{e}_x + i\mathbf{e}_y)$, $\mathbf{e}_x$, and $\mathbf{e}_y$ are unit vectors along the corresponding axes in the plane perpendicular to the light-propagation direction.

In all cases that are close to Emin, except $E_c(\mathbf{k}_0)$ (the amplitude of the effect is in this case negligible), the interference between $M^{(l)}_d$ and $M^{(l)}_{\text{exch}}$ can be neglected:

$$|M^{(l)}_d + M^{(l)}_{\text{exch}}|^2 \approx 2|M^{(l)}_{\text{exch}}|^2.$$

We estimate now the ratio η_l of the probability of an l-photon transition with participation of free carriers to the probability of a direct $(l+1)$-photon transition. We take, e.g., the case of linear polarization and $l=3$. We substitute (11) in (8) and use a δ-function to eliminate the integration with respect to the angle between $\mathbf{k}_2$ and $\mathbf{q}$. Analysis of the integrands, with allowance for the energy and momentum conservation laws, shows that the main contribution is made by $q \sim \tilde{\Delta} k_0^{-1}$, $k_2 \sim [2\tilde{\Delta}/(\gamma+1)]^{1/2}$. Using this to estimate the integrals with respect to $\mathbf{k}_2$ and $\mathbf{q}$, and recognizing that the number of excited carriers that stimulate the MPT is $n_{\text{ex}} \sim \sigma_n n I\tau/\hbar\omega$, where σ_n is the cross section for single-photon intraband light absorption and τ is the energy relaxation times, we get[2)]

$$\eta_3 \sim e^2 n\sigma_n\tau(m_r\hbar\omega^3)^{1/2}\tilde{c}(1+\gamma^{-1})/\varepsilon_l^2\Delta^2, \quad (14)$$

where $\tilde{c}$ is the speed of light in the material. Obviously, $\eta_{2l+1} \sim \eta_l$ at all l. Estimates such as (14) are quite crude. More accurate values of η_l can be obtained by comparing Eqs. (15–(18) below with the probabilities of direct MPT (see, e.g., Ref. 3). At the parameter values typial of III-V semiconductors and at $\Delta \sim 10^{-2}$ eV we have $\eta_3 > 1$ starting with $n \gtrsim 10^{16}$ cm^{-3}.

Substituting (10)-(13) in (8) we obtain after quite lengthy calculations, for two-photon transitions (linear polarization of the pump)

$$W^{(2)}_{ee,L} = \frac{m^2 c_c^{3/2}\tilde{\mu}_2^2}{3\pi\omega^2\hbar^6}\tilde{\Phi}^{(2)}_{-1/2,L}, \quad \tilde{\mu}_l = \frac{1}{\pi}Vq^2\mu_l, \quad (15)$$

and for three-photon transitions (linear polarization)

$$W_{ee,L}^{(3)}=\frac{m_c^{5/2}p_{cv}^4\tilde{\mu}_3^2}{2^{7/2}\hbar^7\omega^4}\tilde{\Phi}_{-3/2,L}^{(3)}. \tag{16}$$

For three-photon transitions and circular polarization of the pump

$$W_{ee,C}^{(3)}=\frac{m^4m_c^{1/2}\tilde{\mu}_3^2}{30\sqrt{2}\,\hbar^7\omega^4}\tilde{\Phi}_{1/2,C}^{(3)}, \tag{17}$$

where

$$\tilde{\Phi}_{\nu,L,C}^{(I)}=\int_{E_{min}}dE\,E^{\nu}f_c(E)\int_{x_1}^{x_2}dxP_{L,C}^{(I)}(x,\gamma,\tilde{E}), \tag{18}$$

$$\tilde{E}=\frac{E}{\Delta},\quad x_{1,2}=\frac{\gamma+1}{2\gamma+1}\left[1\mp\left(1-\frac{2\gamma+1}{\gamma+1}\tilde{E}^{-1}\right)^{1/2}\right]. \tag{19}$$

Explicit expressions for the functions $P_L^{(I)}(x,\gamma,\tilde{E})$ are given in the Appendix. The equation for $P_C^{(3)}$ is extremely unwieldy, and we confine ourselves to a graphic representation of $\Phi_{1/2,C}^{(3)}$ (see Sec. 4).

4. Analysis of the functions $P_{L,C}^{(I)}$ in (15-(18) shows that the predominant contribution to $W_{ee}^{(I)}$ is made by electrons with $E\gg\Delta$. We consider in this connection the distribution function $f_C^I(E)$ in the energy region $\hbar\Omega_0<E\lesssim E_1\approx\hbar\omega$ ($\hbar\Omega_0$ is the optical phonon). The electrons land in this region (which we call band I) from the bottom of the conduction band as a result of direct intraband absorption of light ($\hbar\omega\gg\hbar\Omega_0$) and subsequent relaxation. If the principal energy-loss channel in band I is emission of optical phonons owing to the polarization mechanism of the interaction, the relaxation time is given by the formula

$$\tau_{p0}=\frac{1}{2\alpha\Omega_0}\frac{y^{1/2}}{\mathrm{Arch}\,y^{1/2}},\quad y=\frac{E}{\hbar\Omega_0}, \tag{20}$$

where α is the Fröhlich coupling constant. The averaged distribution function is of the form[22]

$$g(E)f^I(E)=\frac{g_1}{\hbar\Omega_0}\tau_{p0}(E), \tag{21}$$

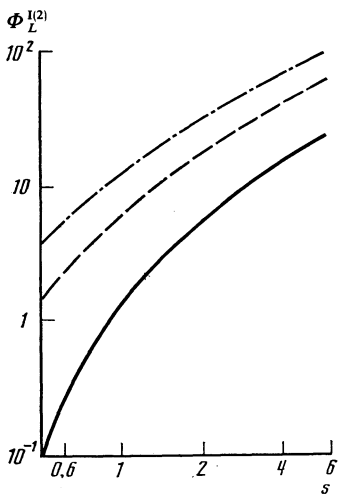

FIG. 5. The function $\Phi_L^{I(3)}(s,\gamma,y_1)$. The solid lines here and in Figs. 6 and 7 correspond to $y_1=5$, the dashed, to $y_1=10$, and the dash-dot, to $y_1=15$. The dependence on γ is weaker in this case, so that only the curves for $\gamma=1$ are shown.

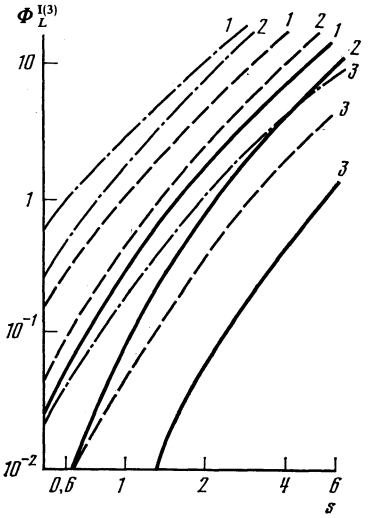

FIG. 6. The function $\Phi_L^{I(3)}(s,\gamma,y_1)$: 1—$\gamma=0.0575$ (corresponds to InAs); 2—$\gamma=1$; 3—$\gamma=17.39$.

where $g(E)$ is the state density and g_1 is the total number of intraband single-photon transitions per cm^3 and per second. Expressing g_1 in terms of σ_n and substituting (20) and (21) in (18) we obtain

$$\tilde{\Phi}_{\nu,L,C}^{(I)}=\frac{\pi^2\sigma_n n\bar{n}_x c\hbar^4}{\sqrt{2}\,\alpha m_c^{1/2}(\hbar\Omega_0)^{1/2-\nu}}\Phi_{L,C}^{I(I)}(\gamma,y_1,s), \tag{22}$$

$$y_1=E_1/\hbar\Omega_0,\quad s=\hbar\Omega_0/\Delta.$$

The functions $\Phi_L^{I(2)}(\gamma,y_1,s)$, $\Phi_L^{I(3)}(\gamma,y_1,s)$ and $\Phi_C^{I(3)}(\gamma,y_1,s)$ are plotted in Figs. 5–7.

Let us consider in greater detail three-photon absorption of linearly polarized light. As seen from Fig. 6, the amplitude of the effect decreases rapidly with increasing Δ, but at small γ this decrease is smoother. At the same time, $W_{ee,L}^{(3)}$ increases with decreasing γ. This means that more favorable conditions for observing the tail of the three-photon fundamental absorption in typical semiconductors, such as III-V,

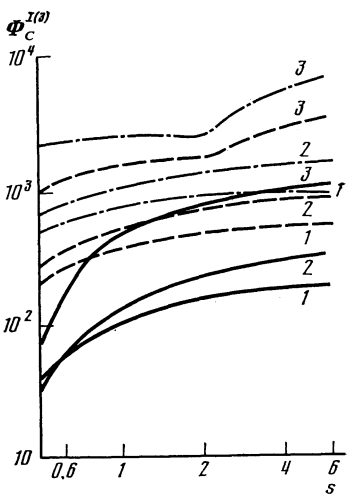

FIG. 7. The function $\Phi_C^{I(3)}(s,\gamma,y_1)$: 1—$\gamma=0.1$; 2—$\gamma=1$; 3—$\gamma=10$.

are realized in n-type samples, for in this case the amplitude of the effect is determined by transitions from the heavy-hole band to the conduction band ($\gamma \ll 1$). When considering the transitions that are stimulated by free holes, it is necessary to replace m_c by m_v and γ by γ^{-1} in all the equations. Since almost all the excited holes are in the heavy subband, we arrive at a situation analogous to the case $\gamma \gg 1$ for stimulating electrons, when the tail is more difficult to observe.

If the pump is circularly polarized, since the three-photon transition is forbidden, the amplitude of the process increases more rapidly than for linear polarization with increasing energy E of the stimulating electron. At sufficient I a situation may arise wherein the magnitude of the effect is determined by electrons with $E_1 < E < E_2 \approx 2\hbar\omega$ (band II). With the aid of the balance equations it is easy to obtain for the averaged distribution function in band II

$$g(E) f^{II}(E) = \frac{\tau_{p0}(E)}{\hbar\Omega_0}\left[g_2 + \frac{8\pi}{3}\frac{e^2\bar{n}_x}{\hbar\omega^{1/2}\Omega^{1/2}m_c} g_1 F\left(\frac{E}{\hbar\omega}\right)\right], \quad (23)$$

where g_2 is the number of two-photon intraband transitions per cm^3 and per second, and

$$F(x) = \int_x^2 \frac{t^{1/2}[\alpha^{-1}(t-1+\alpha)]^{1/2}}{\mathrm{Arch}[\alpha^{-1}(t-1+\alpha)]^{1/2}} dt. \quad (24)$$

A numerical calculation of $\tilde{\phi}^{II(3)}_{1/2,C}$ with a distribution function $f^{II}(E)$ shows, in particular, that at the parameter values corresponding to InAs and to $\hbar\omega = 0.117$ eV, the contribution of band II for $s = 1$ becomes predominant at $g_2/g_1 \gtrsim 4\cdot 10^{-2}$. The corresponding $I \gtrsim 10^7$ W·cm^{-2}. The number of nonequilibrium carriers is then $\delta n \propto I^5$ (as against $\delta n \propto I^4$ for linear pump polarization).

A detailed examination of the experimental data on nonlinear polarization in InAs is outside the scope of this article. We note here only that all the aforementioned qualitative features of three-photon absorption with participation of free carriers were observed in this material even in the first experiments,[10] where absorption of CO_2 laser radiation was investigated under conditions when $0 < E_g - 3\hbar\omega \ll \hbar\omega$.

The authors thank A. M. Danishevskiĭ, E. L. Ivchenko, V. A. Kovarskiĭ, V. A. Mashkov, S. I. Pekar, V. K. Subashiev, and I. D. Yaroshetskiĭ for interest in the work and for helpful discussions.

APPENDIX

We present explicit expressions for the functions $P_L^{(l)}(x,\gamma,\tilde{E})$ in the right-hand side of (18):

$$P_L^{(2)}(x,\gamma,\tilde{E}) = \frac{\gamma_1}{x^3}\left[\frac{C(^2/_3)B(1)}{(2-x)C(4)} - \frac{B(\gamma)}{2x\tilde{E}} - \frac{AC(-^8/_3)}{\tilde{E}_{-1}C(4)}\right], \quad (A.1)$$

$$P_L^{(3)}(x,\gamma,\tilde{E}) = \frac{1}{\gamma\gamma_1 x^4}\left\{\frac{1}{2}B(\gamma) - \gamma A + \frac{2\gamma x}{3\gamma_1}\left[\frac{B(1)}{x}\left(\frac{2}{\tilde{E}_1 C(4)} - \frac{1}{2-x}\right) + \frac{2A}{\tilde{E}C(4)}\right]\right\}, \quad (A.2)$$

where

$$\gamma_n = 1+n\gamma, \quad \tilde{E}_n = \tilde{E}\gamma_1^n, \quad \nu_n = \gamma^{2-n}, \quad C(u) = x^2 + u\tilde{E}_1^{-1},$$

$$A = \sum_{i=1}^{2}(A_i^{(+)} + A_i^{(-)}), \quad A_i^{(\pm)} = \tilde{E}_{-1}^{1/2}\,\mathrm{arctg}[\tilde{E}_{-1}^{1/2}(z \pm \nu_i x)^2],$$

$$B(u) = \sum_{i=1}^{2} u^{i-1}(B_i^{(+)} - B_i^{(-)}), \quad B_i^{(\pm)} = \ln|(z \pm \nu_i x)^2 + \tilde{E}_{-1}^{-1}|.$$

[1)] In some cases the electron system can "attune itself" to a resonance $\Delta \ll \hbar\omega$ because of the heating of the crystal by the pump radiation and the corresponding decrease of E_g.

[2)] It must be borne in mind that the equations given here are not directly applicable at small Δ ($\Delta \lesssim 10^{-3}$ eV). Besides the causes discussed in Sec. 2, we indicate the following. As $\Delta \to 0$ damping must be taken into account in the resonant energy denominators Δ_l in (10)–(13). Inasmuch as at small Δ a contribution can be made to the effect by electrons with small E_c, for which $\tilde{\Omega} < qv_T$, allowance must be made for the Debye screening, although the tendency of q to zero, as seen from (10)–(13), does not by itself lead to a divergence.

[1] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[2] Yu. A. Bychkov and A. M. Dykhne, Zh. Eksp. Teor. Fiz. **58**, 1734 (1970) [Sov. Phys. JETP **31**, 928 (1970)].
[3] V. A. Kovarskiĭ and P. Yu. Perlin, Phys. Stat. Sol. (b) **45**, 47 (1971).
[4] I. M. Catalano, A. Cingolani, and A. Minafra, Phys. Rev. **B5**, 1629 (1972).
[5] I. A. Chaĭkovskiĭ, Fiz. Tekh. Poluprovodn. **6**, 229 (1972) [Sov. Phys. Semicond. **6**, 198 (1972)].
[6] V. A. Kovarskiĭ and E. V. Vitiu, Fiz. Tekh. Poluprovodn. **3**, 421 (1969) [Sov. Phys. Semicond. **3**, 354 (1969)].
[7] F. Bassani and A. R. Hassan, Nuovo Cimento sec. 11, **7B**, 313 (1972).
[8] S. M. Ryvkin, Fiz. Tverd. Tela (Leningrad) **7**, 389 (1965) [sic]; S. M. Ryvkin, A. A. Grinberg, and N. I. Kramer, Fiz. Tverd. Tela (Leningrad) **7**, 2195 (1965) [Sov. Phys. Solid State **7**, 1766 (1965)].
[9] A. A. Rogachev and S. M. Ryvkin, Fiz. Tverd. Tela (Leningrad) **7**, 1278 (1965) [Sov. Phys. Solid State **7**, 1038 (1965)].
[10] A. M. Danishevskiĭ, S. F. Kochegarov, E. Yu. Perlin, V. K. Subashiev, and A. V. Fedorov, Proc. All-Union Conf. on Semiconductor Physics, Vol. I. Baku: Elm, p. 152 (1982).
[11] A. Schmid, P. Kelly, and P. Braunlich, Phys. Rev. **B16**, 4569 (1977).
[12] A. S. Epifanov, A. A. Manenkov, and A. M. Prokhorov, Trudy FIAN **101**, 87 (1978).
[13] J. R. Klauder and E. C. G. Sudarshan, Fundamentals of Quantum Optics, Benjamin, 1968.
[14] V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. **57**, 1217 (1969) [Sov. Phys. JETP **30**, 663 (1970)].
[15] G. S. Agarval, Phys. Rev. **A1**, 1445 (1970).
[16] V. M. Galitskiĭ, S. P. Goreslavskiĭ, and V. F. Elesin, Zh. Eksp. Teor. Fiz. **57**, 207 (1969) [Sov. Phys. JETP **30**, 117 (1970)].
[17] E. Yu. Perlin and V. A. Kovarskiĭ, Fiz. Tverd. Tela (Leningrad) **12**, 3105 (1970) [Sov. Phys. Solid State **12**, 2512 (1971)].
[18] E. Yu. Perlin, Fiz. Tverd. Tela (Leningrad) **15**, 66 (1973) [Sov. Phys. Solid State **15**, 44 (1973)].
[19] S. E. Kumekov and V. I. Perel', Zh. Eksp. Teor. Fiz. **81**, 1693 (1981) [Sov. Phys. JETP **54**, 899 (1981)].
[20] S. B. Arifzhanov, A. M. Danishevskiĭ, E. L. Ivchenko, S. F. Kochegarov, and V. K. Subashiev, Zh. Eksp. Teor. Fiz. **74**, 112 (1978) [Sov. Phys. JETP **47**, 58 (1978)].
[21] E. L. Ivchenko and E. Yu. Perlin, Fiz. Tverd. Tela (Leningrad) **15**, 2871 (1973) [Sov. Phys. Solid State **15**, 1921 (1974)].
[22] I. B. Levinson and B. N. Levinskiĭ, Zh. Eksp. Teor. Fiz. **71**, 300 (1976) [Sov. Phys. JETP **44**, 156 (1976)].

Translated by J. G. Adashko