Theory of linear IR absorption by semiconductors with degenerate bands

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A theory of nonlinear IR absorption by semiconductors with degenerate bands, such as p-Ge, is developed. It is shown that there are two nonlinearity mechanisms. The first is coherent saturation of the absorption in the vertical transition. The second is due to the finite rate of energy relaxation of the heavy holes. The nonlinearity of the first type sets in as a rule earlier with increase of intensity, but at high intensities the asymptote of the absorption coefficient is always determined by the second mechanism. It is shown that the first mechanism is highly sensitive to the presence of charged impurities in the semiconductor and to the degree of coherence of the radiation. Incoherence of the radiation and fluctuating electric fields of the impurities suppress the coherent Rabi oscillations when a hole is periodically moved between the two subbands. The Rabi oscillations are suppressed also by a weak electric (or magnetic field) applied to the semiconductor. The theory is compared with the available experimental data.

INTRODUCTION

Nonlinear absorption of IR radiation in degenerate-band semiconductors of the p-Ge type is now being intensively investigated both experimentally[1–9] and theoretically.[7,8,10–13] There is as yet, however, no meeting of minds concerning the nonlinearity mechanism. Thus, one group of workers[2–5,9–13] assumes that the absorption nonlinearity is due to coherent saturation of the vertical transition between the subbands of the heavy and light holes, and that at high intensities the absorption coefficient decreases with intensity I like $I^{-1/2}$.

A different nonlinearity mechanism was proposed in the papers of another group,[6–8] where it was shown to decrease the absorption coefficient at high intensity like I^{-1}.

We develop in the present paper a theory that makes simultaneous allowance for both mechanisms, and indicate when each of them predominates.

Absorption of IR radiation in degenerate-band semiconductors of the p-Ge type is due to vertical transitions of holes between subbands of the valence band (Fig. 1). We consider a case when the radiation frequency ω is such that the final energy of the light hole ε_f is lower than the $\hbar\omega_0$ of the optical phonon,[1)] and the impurity density is low, so that the holes are scattered only by acoustic phonons.

We show that there are two nonlinearity mechanisms. The first is coherent saturation of the absorption. It becomes important when the vertical-transition matrix element $A_{\mathbf{p}}/2$ (which is proportional to the amplitude of the electric field) becomes of the same order or larger than $\hbar/\tau_{\mathbf{p}}$ (the "spectral width" of the absorption line), where

$$\frac{1}{\tau_{\mathbf{p}}}=\frac{1}{2}\left(\frac{1}{\tau_{1\mathbf{p}}}+\frac{1}{\tau_{2\mathbf{p}}}\right); \tag{1}$$

$\mathbf{p}$ is the momentum corresponding to the vertical transition, while $\tau_{1\mathbf{p}}$ and $\tau_{2\mathbf{p}}$ are the departure times of the heavy and light holes from the initial and final states, respectively. This nonlinearity mechanism is identical with the well-known mechanism of saturation absorption in a two-level system.[14] The role of the two levels is assumed in our case by the states 1 and 2 (see Fig. 1). At $A_p \gg \hbar/\tau_{\mathbf{p}}$ the hole goes over periodically, with frequency $A_{\mathbf{p}}/\hbar$, from one subband to the other—the Rabi oscillations. The absorption coefficient decreases then with increase of the IR radiation intensity I like

$$\alpha\approx\alpha_0(I_1/I)^{1/2},\quad I\gg I_1, \tag{2}$$

where α_0 is the linear absorption coefficient and I_1 is the characteristic intensity at which $A_{\mathbf{p}}\approx\hbar/\tau_{\mathbf{p}}$. This occurs if no account is taken of the second nonlinearity mechanism, i.e., if $I_2=\infty$ [see Eq. (5)].

The cause of this asymptotic behavior can be explained in the following manner. The absorption coefficient of a two-level system with distance ΔE between the levels is given by[14]

$$\alpha\sim\frac{\hbar/\tau}{(\hbar\omega-\Delta E)^2+A^2/2+(\hbar/\tau)^2}, \tag{3}$$

where τ is the corresponding relaxation time.

It follows from (3) that the absorption line contour is Lorentzian with a height (at $A\gg\hbar/\tau$) inversely proportional to A^2 and with a width proportional to A (so-called dynamic broadening of the spectral line). The coefficient of absorption by an ensemble of such systems with different E, distributed over a sufficiently broad interval, is proportional to the area under this curve, i.e., $\alpha\sim 1/A\sim I^{-1/2}$.

To understand the cause of the second nonlinearity mechanism, we turn to Fig. 1. After absorbing a photon, a heavy hole goes over into the light subband. Next, absorbing or emitting an acoustic phonon, it returns to the heavy subband (in view of th higher density of states in it). Since the scattering by an acoustic phonon is quasielastic, the energy change in such a transition is small and the transition is al-

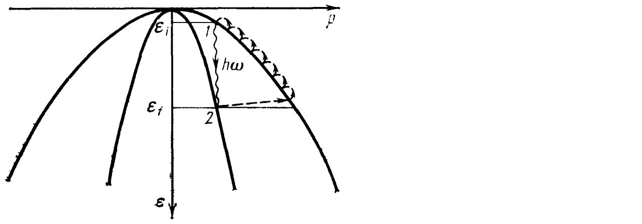

FIG. 1. Scheme of optical transitions and subsequent energy relaxation of heavy holes.

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most horizontal. This is followed by a start of a slow energy relaxation of the hole in the heavy band, from an energy ε_f to an energy ε_i, due to quasielastic scattering by the acoustic phonons.

The second nonlinearity mechanism becomes important when the rate of the vertical transitions of the heavy holes to the light subband (which is proportional to the radiation intensity) becomes of the order of the rate of their energy relaxation in the heavy hole from an energy ε_f to ε_i. We designate the corresponding intensity by I_2.

At $I \gg I_2$, the hole populations become equalized in the initial and final states corresponding to the vertical transition, and the absorption coefficient decreases with increase of intensity like

$$\alpha \approx \alpha_0 \frac{I_2}{I}, \tag{4}$$

if the first nonlinearity mechanism is disregarded, i.e., $I_1 = \infty$.

Relations of type (2) and (4) for the coefficient of linear absorption, and these two mechanisms, were separately known earlier (Refs. 7, 8, and 13). In the present paper we consider both mechanisms jointly. We show that if both are taken into account simultaneously the dependence of the absorption coefficient on the intensity (if the IR radiation is linearly polarized) is given by

$$\alpha = \frac{\alpha_0}{\zeta^{-1}(I/I_1) + I/I_2}, \tag{5}$$

where

$$\zeta(x) = \frac{3}{4x}\left[1 + \frac{x-1}{x^{1/2}} \arcsin\left(\frac{x}{1+x}\right)^{1/2}\right] = \begin{cases} 1, & x \ll 1 \\ \dfrac{3\pi}{8x^{1/2}} & x \gg 1 \end{cases}. \tag{6}$$

The relation between the characteristic intensities I_1 and I_2 depends on the IR frequency, the temperature, and the parameters of the semiconductor. The ratio I_1/I_2 is proportional to the hole-phonon coupling constant which depends on the characteristic energy of holes and increases with the latter. In principle, therefore, a situation with either $I_1 \ll I_2$ or its converse can be realized. It is interesting that in both cases the asymptotic form of the absorption coefficient coincides with (4) at $I \gg I_2(1 + I_2/I_1)$. At $I_1 \ll I_2$ and $I_1 \ll I \ll I_2^2/I_1$, however it is preceded by the asymptote (2).

1. FUNDAMENTAL EQUATIONS

The absorption coefficient α is determined from the off-diagonal component of the hole density matrix

$$\hat{F}_{\mathbf{p}} = \begin{pmatrix} f_{1\mathbf{p}} & \Psi_{\mathbf{p}} e^{i\omega t} \\ \Psi_{\mathbf{p}}^* e^{-i\omega t} & f_{2\mathbf{p}} \end{pmatrix} \tag{7}$$

to be

$$\alpha = -\frac{2\omega}{IV} \sum_{\mathbf{p}} A_{\mathbf{p}} \operatorname{Im} \Psi_{\mathbf{p}}, \tag{8}$$

where $A_{\mathbf{p}}/2$ is the matrix element of the vertical transition, I the intensity of the electromagnetic wave, ω its frequency, and V the volume of the crystal. The off-diagonal component of the density matrix $\Psi_{\mathbf{p}}$ and the distribution functions $f_{1\mathbf{p}}$ and $f_{2\mathbf{p}}$ of the heavy and light holes are determined from the system of kinetic equations

$$\frac{df_{1\mathbf{p}}}{dt} = \frac{A_{\mathbf{p}}}{\hbar} \operatorname{Im} \Psi_{\mathbf{p}} + I_{11}^+ + I_{11}^- + I_{21}^+ + I_{12}^-, \tag{9}$$

$$\frac{df_{2\mathbf{p}}}{dt} = -\frac{A_{\mathbf{p}}}{\hbar} \operatorname{Im} \Psi_{\mathbf{p}} + I_{22}^+ + I_{22}^- + I_{12}^+ + I_{21}^-, \tag{10}$$

$$\frac{d\Psi_{\mathbf{p}}}{dt} = \frac{i}{\hbar}\left[(\varepsilon_{1\mathbf{p}} - \varepsilon_{2\mathbf{p}} - \hbar\omega)\Psi_{\mathbf{p}} + \frac{A_{\mathbf{p}}}{2}(f_{2\mathbf{p}} - f_{1\mathbf{p}})\right] - \frac{\Psi_{\mathbf{p}}}{\tau_{\mathbf{p}}}. \tag{11}$$

This system was obtained in the so-called resonance approximation, when $A_{\mathbf{p}} \ll \hbar\omega$. It coincides with the system of equations of Ref. 11.

In Eqs. (9) and (10), $I_{jk}^{\pm}$ $(j,k = 1,2)$ represents the arrival (+) and departure (−) integrals of the collisions between the holes and the acoustic phonons, and describes their transition from the subband j to the subband k; $\tau_{\mathbf{p}}$ is the relaxation time of the off-diagonal component of the density matrix and is given by Eq. (1). Since the resonance region is narrow, we have left out of (11) the term integral with respect to $\Psi_{\mathbf{p}}$ (the arrival term). It describes "arrival of coherency" from other points of momentum space on account of hole interaction with acoustic phonons. Allowance for this term necessitates corrections small in the parameter $(\omega_{\mathbf{p}}\tau_{\mathbf{p}})^{-1} \ll 1$, where $\omega_{\mathbf{p}}$ is the frequency of an acoustic phonon with momentum $2\mathbf{p}$ [see (15)].

We consider next for simplicity the case when the mass m_1 of the heavy holes is much larger than the mass m_2 of the light ones. We take into account in Eqs. (9) and (10) only the preferred collision integrals that describe the hole transitions to the light subband (since the density of states is substantially lower there). An exception is a narrow energy region near ε_f, where such transition must be taken into account [the terms I_{12}^- and I_{12}^+ in (9) and (19)], since they can substantially influence the difference of the distribution functions of the heavy and light holes at the point of the vertical transition, and by the same token also influence the absorption coefficient.

2. DISTRIBUTION FUNCTIONS

Under stationary conditions we have from (11) the following expression for $\operatorname{Im}\Psi_{\mathbf{p}}$:

$$\operatorname{Im} \Psi_{\mathbf{p}} = \frac{\hbar A_{\mathbf{p}}}{2\tau_{\mathbf{p}}} \frac{f_{2\mathbf{p}} - f_{1\mathbf{p}}}{(\varepsilon_{1\mathbf{p}} - \varepsilon_{2\mathbf{p}} - \hbar\omega)^2 + (\hbar/\tau_{\mathbf{p}})^2}. \tag{12}$$

Substituting $\operatorname{Im}\Psi_{\mathbf{p}}$ in Eqs. (9) and (10), we obtain a system of coupled equations for the distribution functions $f_{1\mathbf{p}}$ and $f_{2\mathbf{p}}$ of the heavy and light holes. These functions can be represented in the form

$$f_{j\mathbf{p}} = f_{j\mathbf{p}}^{(si)} + f_{j\mathbf{p}}^{(sm)}. \tag{13}$$

Here $f_{j\mathbf{p}}^{(si)}$ is the "singular" (abrupt) part of the distribution function and is relatively large only if $\mathbf{p}$ satisfies the resonance condition

$$|\varepsilon_{1\mathbf{p}} - \varepsilon_{2\mathbf{p}} - \hbar\omega| \lesssim \hbar/\tau_{\mathbf{p}},$$

$f_{j\mathbf{p}}^{(sm)}$ is the smooth part of the distribution function and varies over energy scales on the order of max $(T, \hbar\omega)$, where T is the lattice temperature. Both quantities are considerably larger than $\hbar/\tau_{\mathbf{p}}$.

The kinetic equations (9)–(11) have thus two types of terms, abrupt and smooth functions of $\mathbf{p}$, respectively. Clearly, they must cancel one another separately. Equations (9) and (10) are therefore transformed near resonance:

$$\frac{A_p}{\hbar}\operatorname{Im}\Psi_p-\frac{f_{1p}^{(si)}}{\tau_{1p}}=0,\quad -\frac{A_p}{\hbar}\operatorname{Im}\Psi_p-\frac{f_{2p}^{(si)}}{\tau_{2p}}=0. \tag{14}$$

These equations have no terms that are integral (arrival) with respect to $f_{jp}^{(si)}$, since the resonance region has a width $\hbar/\tau_p$, and the "arrival" is from an energy region of order $h\omega_p$, where ω_p is the frequency of an acoustic phonon with momentum $2p$. We confine ourselves in the present paper to a case when

$$\omega_p\tau_p\gg 1; \tag{15}$$

this inequality holds in practically the entire temperature interval in which the interaction between the holes and the optical phonons can be neglected (see Sec. 5).

We express with the aid of (12)–(14) the singular parts of the hole distribution functions in terms of the smooth parts:

$$f_{1p}^{(si)}=A_p\hbar^{-1}\tau_{1p}\operatorname{Im}\Psi_p, \tag{16}$$

$$f_{2p}^{(si)}=-A_p\hbar^{-1}\tau_{2p}\operatorname{Im}\Psi_p, \tag{17}$$

where

$$\operatorname{Im}\Psi_p=-\frac{\hbar A_p}{2\tau_p}\frac{f_{1p}^{(sm)}-f_{2p}^{(sm)}}{(\varepsilon_{1p}-\varepsilon_{2p}-\hbar\omega)^2+(\hbar/\tau_p)^2+A_p^2(\tau_{1p}+\tau_{2p})/2\tau_p}. \tag{18}$$

Since the functions $f_{jp}^{(sm)}$ are smooth (in the present case they depend only on energy), they should be regarded in (18) as constants taken at the point **p** corresponding to the vertical transition: $\varepsilon_{1p}-\varepsilon_{2p}-\hbar\omega=0$.

It follows from (10) that

$$f_{1p}^{(sm)}=f_{2p'}^{(sm)}, \tag{19}$$

where $\mathbf{p}'=(m_2/m_1)^{1/2}\mathbf{p}$ (for the case of isotropic parabolic bands). This reflects the equality of the smooth parts of the distribution functions of the light and heavy holes at one and the same energy ε, which is a consequence of the quasielastic character of the scattering of the holes by acoustic phonons.

To determine the absorption coefficient α we must thus find the values of the smooth part of the heavy-hole distribution function $f_{1p}^{(sm)}$ at two points: at the initial energy ε_i and at the final one ε_f.

Far from the energies ε_i and ε_f the equation for the smooth part of the heavy-hole distribution function $f_{1p}^{(sm)}$ reduces to

$$I_{11}^{+}+I_{11}^{-}=0.$$

Owing to the quasielastic character of the hole scattering, this equation can be written in differential form.[15] At $T\gg\hbar\omega_p$ we have

$$\frac{1}{g_1(\varepsilon)}\frac{\partial}{\partial\varepsilon}\left[\varepsilon g_1(\varepsilon)\nu(\varepsilon)\left(T\frac{\partial f_{1p}^{(sm)}}{\partial\varepsilon}+f_{1p}^{(sm)}\right)\right]=0, \tag{20}$$

where $g_1(\varepsilon)=m_1^{3/2}\varepsilon^{1/2}/2^{1/2}\pi^2\hbar^3$ is the density of states in the heavy band. The function $f_{1p}^{(sm)}$ is assumed dependent on **p** via ε ($\varepsilon=p^2/2m_1$), since the quasielastic scattering establishes a distribution that depends only on energy. The quantity

$$\nu(\varepsilon)=\frac{2^{1/2}m_1^{5/2}\varepsilon^{1/2}}{\pi\hbar^4\rho}\left[(a-b)^2+\frac{3}{4}b^2\right] \tag{21}$$

characterizes the rate of the energy relaxation of the heavy hole; c and b are the constants of the deformation potential and describe the interaction of the holes with the acoustic phonons,[16–18] and ρ is the density of the crystal. In the calculation of (21) we have left out small terms proportional to the mass ratio m_2/m_1 of the light and heavy holes.[19]

Equation (20) reflects the constancy of the heavy-hole flux $j_1(\varepsilon)$ along the energy axis:

$$j_1(\varepsilon)=-\varepsilon g_1(\varepsilon)\nu(\varepsilon)\left(T\frac{\partial f_{1p}^{(sm)}}{\partial\varepsilon}+f_{1p}^{(sm)}\right)=c. \tag{22}$$

A nonzero flux exists only in region II: $\varepsilon_f\leqslant\varepsilon\leqslant\varepsilon_i$ between the drain (at $\varepsilon=\varepsilon_i$) and source (at $\varepsilon=\varepsilon_f$) of the heavy holes.[8] The flux under stationary conditions should be equal to the total drain power:

$$c=\frac{1}{V}\sum_p\frac{A_p}{\hbar}\operatorname{Im}\Psi_p=-J[f_{1p}^{(sm)}(\varepsilon_i)-f_{1p}^{(sm)}(\varepsilon_f)], \tag{23}$$

where the quantity J, which is independent of the radiation intensity I, is equal to

$$J=\frac{1}{V}\sum_p\frac{A_p^2/2\tau_p}{(\varepsilon_{1p}-\varepsilon_{2p}-\hbar\omega)^2+(\hbar/\tau_p)^2+A_p^2(\tau_{1p}+\tau_{2p})/2\tau_p}. \tag{24}$$

Within the limits of the accuracy of our calculation ($A_p\ll\hbar\omega$—the resonance approximation), the function in the sum over **p** in (24) can be approximated by a delta function[2)]

$$J=\frac{\pi}{V}\sum_p\frac{A_p^2/2\hbar}{(1+(A_p^2/\hbar^2)\tau_{1p}\tau_{2p})^{1/2}}\delta(\varepsilon_{1p}-\varepsilon_{2p}-\hbar\omega). \tag{25}$$

We have used here Eq. (1).

In the case of linear polarization of an electromagnetic wave, to which we confine ourselves, in the isotropic model the matrix element of the vertical transition depends on p and on the angle θ between the vector **p** and the polarization direction[20]:

$$A_p=\frac{3^{1/2}}{4}\frac{e\mathscr{E}_0}{\omega}\frac{m_1-m_2}{m_1m_2}p\sin\theta, \tag{26}$$

where e is the electron charge and $\mathscr{E}_0$ the amplitude of the electric field of the wave.

Substituting (26) in (25) and calculating, we obtain in the limit as $m_2/m_1\to 0$

$$J=\frac{e^2m_2^{1/2}I}{2^{1/2}\hbar^2c\varepsilon_0^{1/2}(\hbar\omega)^{1/2}}\zeta\left(\frac{I}{I_1}\right), \tag{27}$$

where $\mathscr{E}_0$ is the dielectric constant; the function $\zeta(x)$ is defined by Eq. (6), and the characteristic intensity

$$I_1=\frac{\hbar c\varepsilon_0^{1/2}\omega m_2}{3\pi e^2\tau_{1p}\tau_{2p}}. \tag{28}$$

In regions I ($\varepsilon\leqslant\varepsilon_i$) and III ($\varepsilon\geqslant\varepsilon_f$) the flux j_1 is zero and the heavy holes have a Boltzmann distribution.

Solving Eq. (22) in region II an joining the distribution functions at the boundaries of the regions at the points ε_i and ε_f we get (cf. Ref. 8)

TABLE I. The functions $\phi(x)$ and $\Phi(x)$ for $0.1 \leqslant x \leqslant 10$.

x	$\varphi(x)$	$\Phi(x)$	x	$\varphi(x)$	$\Phi(x)$
0.1	−13.3	13.17	2.2	1.05	1.62
0.2	−7.51	7.63	2.4	1.43	1.54
0.3	−5.38	5.67	2.6	1.82	1.48
0.4	−4.20	4.64	2.8	2.23	1.42
0.5	−3.42	3.99	3.0	2.66	1.37
0.6	−2.84	3.54	3.5	3.89	1.26
0.7	−2.39	3.21	4.0	5.40	1.17
0.8	−2.01	2.95	4.5	7.35	1.1
0.9	−1.69	2.74	5.0	9.93	1.04
1.0	−1.40	2.57	6.0	18.2	0.944
1.2	−0.902	2.3	7.0	34.3	0.871
1.4	−0.467	2.1	8.0	67.2	0.812
1.6	−0.0675	1.94	9.0	137	0.764
1.8	+0.312	1.82	10.0	289	0.723
2.0	0.682	1.71			

$$
\begin{aligned}
&\mathrm{I}\ (\varepsilon<\varepsilon_i) && f_{1p}^{(sm)}=\frac{n}{n_0}\frac{1+ZB(x_i)}{1+Z}e^{-x},\\
&\mathrm{II}\ (\varepsilon_i<\varepsilon<\varepsilon_f) && f_{1p}^{(sm)}=\frac{n}{n_0}\frac{1+ZB(x)}{1+Z}e^{-x},\\
&\mathrm{III}\ (\varepsilon>\varepsilon_f) && f_{1p}^{(sm)}=\frac{n}{n_0}\frac{1+ZB(x_f)}{1+Z}e^{-x},
\end{aligned}
\tag{29}
$$

where $x=\varepsilon/T$, $x_i=\varepsilon_i/T$, $x_f=\varepsilon_f/T$, n is the density of the heavy holes, and n_0 is determined from the normalization condition

$$n_0=m_1^{3/2}T^{3/2}/2^{1/2}\pi^{3/2}\hbar^3, \tag{30}$$

$B(x)$ is defined as

$$B(x)=\frac{\varphi(x)-\varphi(x_i)+N_\omega[\varphi(x_f)-\varphi(x_i)]}{\Phi(x_i)-\Phi(x_K)+N_\omega[\varphi(x_f)-\varphi(x_i)]}, \tag{31}$$

where $N_\omega=(e^{\hbar\omega/T}-1)^{-1}$ is the Planck function, and the expressions for the functions $\varphi(x)$ and $\Phi(x)$ are given in the Appendix. These functions were tabulated by us for the most interesting region of the argument values (see Table I). At other values of x they can be calculated with sufficient accuracy from asymptotic equations that are likewise given in the Appendix. Finally, the intensity-dependent quantity Z is equal to

$$Z=(I/I_2)\zeta(I/I_1), \tag{32}$$

where the characteristic intensity is

$$I_2=\frac{2^{1/2}c\varepsilon_0^{1/2}(\hbar\omega)^{1/2}m_1^4T^2}{\pi^3e^2\hbar^5\rho m_2^{1/2}\beta}\left[(a-b)^2+\frac{3}{4}b^2\right], \tag{33}$$

and

$$\beta=e^{-x_K}\{\varphi(x_f)-\varphi(x_i)+N_\omega^{-1}[\Phi(x_i)-\Phi(x_f)]\}. \tag{34}$$

It follows from (29), with (32) and (6) taken into account, that the smooth part of the heavy-hole distribution function does not tend to an intensity-dependent limit with increase of the intensity. The difference $f_{1p}^{(sm)}(\varepsilon_i)-f_{1p}^{(sm)}(\varepsilon_f)$, however, tends to zero with increase of intensity, i.e., the smooth parts of the distribution functions of the light and heavy holes become equalized at the vertical-transition point.

3. THE ABSORPTION COEFFICIENT

According to (8) and (23), the absorption coefficient is

$$\alpha=\hbar\omega\frac{2J}{I}[f_{1p}^{(sm)}(\varepsilon_i)-f_{1p}^{(sm)}(\varepsilon_K)]. \tag{35}$$

Substituting here the distribution function (29), we arrive at Eq. (5), where

$$\alpha_0=\frac{n}{n_0}\frac{2^{1/2}e^2m_2^{1/2}(\hbar\omega)^{1/2}}{\hbar^2c\varepsilon_0^{1/2}}e^{-x_f}N_\omega^{-1} \tag{36}$$

is the linear absorption coefficient, while the characteristic intensities I_1 and I_2 are given respectively by Eqs. (28) and (33). The asymptote of the absorption coefficient at high intensities depends on the ratio of these intensities.

The definition (28) of I_1 contains the relaxation times τ_{1p} and τ_{2p}. These satisfy (at $T\gg\hbar\omega_p$) the following expressions (at the point of the vertical transition)[16,19]:

$$
\begin{aligned}
\frac{1}{\tau_{1p}}&=\frac{2^{1/2}m_1m_2^{1/2}(\hbar\omega)^{1/2}T}{\pi\hbar^4\rho s_t^2}\left[\frac{3}{8}b^2+\frac{s_t^2}{2s_l^2}\left(a^2-ab+\frac{3}{2}b^2\right)\right],\\
\frac{1}{\tau_{2p}}&=\frac{2^{1/2}m_1^{3/2}(\hbar\omega)^{1/2}T}{\pi\hbar^4\rho s_t^2}\left[\frac{3}{4}b^2+\frac{s_t^2}{2s_l^2}(a-b)^2\right],
\end{aligned}
\tag{37}
$$

where s_t and s_l are respectively the velocities of the transverse and longitudinal sounds.

4. INFLUENCE OF WEAK ELECTRIC FIELD ON COHERENT SATURATION OF THE ABSORPTION

As follows from numerical estimates given in the next section, the case realized as a rule is one in which

$$I_1<I_2. \tag{38}$$

With increase of intensity, the absorption of the IR radiation exhibits first a nonlinearity of the first type, due to coherent equalization of the level populations of the two-level system at the vertical-transition point. If

$$I_1<I<I_2^2I_1 \tag{39}$$

the absorption coefficient decreases with increase of intensity in accordance with Eq. (2). The hole moves then periodically from the heavy subband to the light one and back, with a frequency $A_p/\hbar$ (Rabi oscillations). The relaxation time τ_p of the phase of the oscillations is long ($\tau_p>\hbar/A_p$) and is due

TABLE II. Characteristic intensities I_1 and I_2 calculated for p-Ge at various temperatures and wavelengths.

λ, μm	T, K	I_1 W/cm^2	I_2 W/cm^2	I_2/I_1
100	20	2.61	81.3	31.1
	30	5.87	97.0	16.5
	40	10.4	116	11.1
	50	16.3	138	8.46
	60	23.5	160	6.81
	70	32	186	5.82
	77.4	39.1	198	5.06
	80	41.8	204	4.88
	90	52.9	229	4.33
	100	65.3	259	3.97
	110	79	280	3.55
	120	94	291	3.10
	130	110	310	2.81
	140	128	332	2.60
	150	147	359	2.44
200	77.4	9.78	66	6.75
50	77.4	156	645	4.12

to the interaction of the holes with acoustic phonons, an interaction that takes them out of the resonance region; the width of the latter is [see (18)]

$$\Delta p_{res} \approx \frac{A_p}{v_{2p}}\left(\frac{\tau_{1p}+\tau_{2p}}{2\tau_p}\right)^{1/2}, \tag{40}$$

where $v_{2p} = p/2m_2$ is the velocity of a light hole with momentum $\mathbf{p}$ at the vertical-transition point.

It is possible to take holes out of resonance and by the same token suppress the Rabi oscillations by applying to the semiconductor a constant electric field $\mathbf{E}$.[21,22] The hole momentum changes in the electric field after a time t by an amount $\Delta \mathbf{p}_E = e\mathbf{E}t$. The coherent oscillations are suppressed if the change $\Delta \mathbf{p}_E$ of the hole momentum during a time equal to the period of the oscillations $\hbar/A_p$ in the electric field exceed Δp_{res}; this occurs if

$$E \geqslant \frac{A_p^2}{e\hbar v_{2p}}\left(\frac{\tau_{1p}+\tau_{2p}}{2\tau_p}\right)^{1/2}. \tag{41}$$

The difference between the population increases in this case, as does the absorption coefficient. The minimal electric field that can influence the absorption in this manner occur on the nonlinearity threshold at $A_p \sim \hbar/\tau_p$, and the field E corresponding to this A_p is

$$E_{min} \approx \frac{\hbar}{e v_{2p}\tau_p^2}\left(\frac{\tau_{1p}+\tau_{2p}}{2\tau_p}\right)^{1/2}. \tag{42}$$

Note that $E_{\min}$ is substantially weaker than the electric field capable of heating, for in a time τ_p it alters the light-hole energy by only $\hbar/\tau_p$, which is of the order of the energy uncertainty and much less than the energy itself.

5. DISCUSSION OF RESULTS AND NUMERICAL ESTIMATES

We present in this section, with p-Ge as the example, numerical estimates of the quantities involved in the theory, and estimate the region of its validity. We begin with an analysis of the characteristic intensities I_1 and I_2. They are given by Eqs. (28) with (37) and (33) with (34), respectively. Table II lists the values of I_1 and I_2 and of their ratios calculated for different temperatures and wavelengths. It can be seen that I_1 is always less than I_2 and their ratio (I_2/I_1) decreases with rise of temperature. We used in the table the following values for the constants: $a = -8.5$ eV, $b = -2.6$ eV, $\rho = 5.35$ g/cm^3, $S_t = 3.25\times 10^5$ cm/s, $s_l = 5.35\times 10^5$ cm/s, $\varepsilon_0 = 16$, $m_1 = 0.33m$, $m_2 = 0.042m$ (m is the mass of the free electron). These values of the deformation potential agree with the experimental conductivity of holes scattered by acoustic phonons.[23]

Figure 2 shows the dependence of the smooth part $f_{1p}^{(sm)}$ of the heavy-hole distribution function on the energy referred to the optical-phonon energy and calculated from Eqs. (29) for liquid-nitrogen temperature ($T = 77.4$ K), a radiation wavelength 100 μm, and $Z = 5$, corresponding to an intensity $I \approx 15$ kW/cm^2. It can be seen that in contrast of Ref. 8 the maximum of this function occurs not at the energy ε_f, but is strongly shifted towards lower energies close to ε_i. The reason is that in our problem it is important to take into account the arrival of holes from the heavy to the light subband at $\varepsilon = \varepsilon_f$, which influences strongly the difference between the distribution functions of the light and heavy holes at the vertical-transition point.

The presence of a maximum in the smooth part of the distribution function of the heavy holes leads to the following interesting phenomenon. As follows from (19), the distribution functions $f_{1p}^{(sm)}$ and $f_{2p}^{(sm)}$ of the light and heavy holes coincide at one and the same energy. Therefore the presence of a maximum in the distribution function of the heavy holes leads to a similar maximum in the light-hole

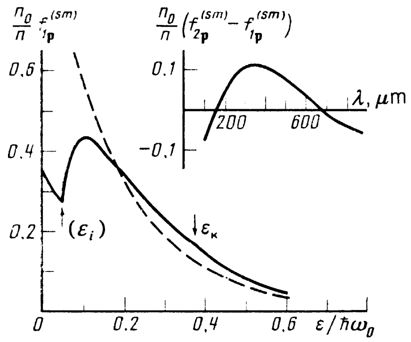

FIG. 2. Smooth part of heavy-hole distribution function vs the hole energy (referred to the optical-phonon energy $\hbar\omega_0$), calculated from Eq. (29) for p-Ge, $\lambda = 100\ \mu$m, $T = 77.4$ K, and $Z = 5$. The dashed curves show the equilibrium distribution ($Z = 0$). The inset shows the dependence of $f_{2p}^{(sm)} - f_{1p}^{(sm)}$ on the radiation wavelength λ corresponding to a vertical transition with momentum $\mathbf{p}$.

distribution function. In view of the difference between the masses m_1 and m_2, these maxima have different locations in momentum space: $P_{2max} = (m_2/m_1)^{1/2} P_{1max}$ (the maximum of the light–hole distribution function is located to the left of the one for the heavy holes). The difference $f_{2\mathbf{p}}^{(sm)} - f_{1\mathbf{p}}^{(sm)}$ in this region can therefore become positive, and population inversion takes place for the corresponding vertical transition. The inset of Fig. 2 shows the dependence of the difference between the distribution functions of the light and heavy holes on the wavelength corresponding to the vertical transition with momentum **p**. This difference turns out to be positive in the region $160 < \lambda < 660\,\mu$m, i.e., emission with such wavelengths can become amplified by nonequilibrium holes rather than be absorbed. A similar phenomenon occurs when the pumping wavelength is 10.6 μm (Ref. 8).

We estimate now the relaxation times contained in the problem, and discuss the role of the impurities and of the hole-hole collisions. At IR wavelength $\lambda = 100\,\mu$m and nitrogen temperature $T = 77.4$ K the relaxation times $\tau_{1\mathbf{p}}, \tau_{2\mathbf{p}}$ (37) and $\tau_{\mathbf{p}}$ (1) in p-Ge are respectively 1.58×10^{-11} s, 0.67×10^{-11} s and 0.95×10^{-11} s. The energy-relaxation times $\tau_\varepsilon = \nu^{-1}(\varepsilon)$ determined from (21) for the initial ε_f and final ε_f energy are respectively 1.7×10^{-9} and 0.6×10^{-9} s.

To estimate the frequency of the collisions of the heavy holes with one another, which describes the rate of energy relaxation, we use the expression[17]

$$\frac{1}{\tau_{hh}} = \frac{\pi e^4 n}{2^{1/2}\varepsilon_0^2 m_1^{1/2}\varepsilon^{3/2}}\left(\ln\frac{8m_1\varepsilon}{\hbar^2\varkappa^2} - \frac{5}{2}\right), \quad (43)$$

where $\varkappa^{-1} = (\varepsilon_0 T/4\pi e^2 n)^{1/2}$ is the screening radius. We take the characteristic heavy-hole energy to be the energy corresponding to the maximum of the distribution functions of the heavy holes, shown in Fig. 2, viz., $\varepsilon_{\max} = 0.64\times10^{-14}$ erg. We find then from (43) that $\tau_{hh} = 1.7\times10^{-9}$ s $= \nu^{-1}(\varepsilon_i)$ at $n = 10^{12}$ cm^{-3}. Thus, the energy relaxation is determined by the hole interaction with the acoustic phonons, at hole densities $\lesssim 10^{12}$ cm^{-3} and by the collisions between the holes at densities $> 10^{12}$ cm^{-3}.

As indicated in the preceding section, even a rather weak constant electric field can influence the coherent saturation of the absorption, taking the holes out of the resonance region. An estimate in accordance with Eq. (42) gives for p-Ge a value $E_{\min} \approx 0.23$ V/cm at $T = 77.4$ K and $\lambda = 100\,\mu$m. Such weak electric fields can be produced by charged impurities. Thus, at an impurity density $N_i = 10^{12}$ cm^{-3} the electric field at an average distance $\bar{r} = (\frac{4}{3}\pi N_i)^{1/3}$ between impurities is of the order of $E_i = e/\varepsilon_0\bar{r}^2 = 2.3$ V/cm. This leads to the important conclusion that electric fields due to charge impurities can hinder coherent saturation of the absorption. Let us estimate the impurity density at which this takes place.

The time required for a light particle to travel the distance $\bar{r}$ between impurities is equal to $\tau_f = \bar{r}/v_{2\mathbf{p}}$, and the particle momentum changes during that time by $\Delta P = eE_i\tau_f$. The ensuing change of kinetic energy is of the order of the potential energy $\Delta\varepsilon = \mathbf{v}_{2\mathbf{p}}\Delta\mathbf{p} \approx e^2/\varepsilon_0\bar{r}$.

The electric fields of the impurities will take the hole out of the resonance region if $\Delta\varepsilon > \hbar/_{\tau\mathbf{p}}$ ($\hbar/_{\tau\mathbf{p}}$ is the width of the resonance region). Thus, impurities hinder coherent saturation of the absorption if their density $N_i > N_i^{\min}$, where

$$N_i^{min} = \frac{3}{4\pi}\left(\frac{\hbar\varepsilon_0}{e^2\tau_{\mathbf{p}}}\right)^3. \quad (44)$$

For $\tau_{\mathbf{p}} = 0.95\cdot10^{-11}$ s we have $N_i = 1.1\cdot10^{11}$ cm^{-3}. Thus, very pure p-Ge specimens are needed for an experimental observation of coherent saturation of absorption in this wavelength and temperature region. The absorption coefficient due to the hole vertical transitions is then small and can be discerned in experiment by its dependence on the intensity.

To conclude this section, we estimate the parameter $(\omega_{\mathbf{p}}\tau_{\mathbf{p}})^{-1}$ (whose smallness was used by us to develop the theory [see Eq. (15)]), and the possibility of using the resonance approximation.

The characteristic value of the acoustic-frequency $\omega_{\mathbf{p}}$ of acoustic phonons emitted by backward-scattered holes is $\omega_{\mathbf{p}} = (2S_l/\hbar)\times(2m_1\varepsilon)^{1/2}$ (for longitudinal phonons and heavy holes.) The product $(\omega_{\mathbf{p}}\tau_{\mathbf{p}})^{-1}$, as follows from (1) and (37) is independent at the vertical-transition point of the radiation photon energy $\hbar\omega$ and is determined only by the temperature, increasing in proportion to T. For $T = 77.4$ K we have in p-Ge $(\omega_{\mathbf{p}}\tau_{\mathbf{p}})^{-1} = 0.08$. This parameter is small in the entire temperature range of interest to us (so long as the influence of optical phonons can be neglected).

The condition $A_{\mathbf{p}} \ll \hbar\omega$ for the applicability of the resonance approximation is met at intensities $I \ll I_r$, where the characteristic intensity is

$$I_r = \frac{c\varepsilon_0^{1/2} m_2(\hbar\omega)^3}{3\pi e^2\hbar^2}. \quad (45)$$

In p-Ge we have $I_r = 1.5$ MW/cm^2 at $\lambda = 100\,\mu$m. At higher intensities account must be taken of multiphoton processes.[24]

6. NONLINEAR ABSORPTION OF SHORT-WAVE IR RADIATION IN *p*-Ge

The theory developed by us can be generalized to include absorption of radiation of shorter wavelength, when the final energy of the light hole exceeds that of the optical phonon. This is precisely the situation realized when radiation of wavelength 10.6 μm is absorbed.[1–13]

We consider here only the case of low temperatures, when absorption of optical phonons can be neglected. When excited by 10.6-μm light the holes go over from a state with energy $\varepsilon_i = 23$ meV of the heavy-hole subband into a state with energy $\varepsilon_f' = 140$ meV of the light-hole subband. The light hole emits next one optical phonon ($\hbar\omega = 37$ meV) and goes over to the heavy subband. It emits there in succession two more optical phonons and ends up in a state of energy $\varepsilon_f = \varepsilon_f' - 3\hbar\omega_0 = 29$ meV. This is followed by energy relaxation of the heavy hole from ε_f to ε_i on acoustic phonons (as shown in Ref. 8, this takes place at a hole density lower than $7\cdot10^{13}$ cm^{-3}).

By the method described in the preceding sections we obtain in this case Eq. (5) for the absorption coefficient. The only difference is that owing to the almost instantaneous emission of optical phonons and to the absence of processes with absorption of optical phonons, the smooth part of the light-hole distribution function $f_{2\mathbf{p}}^{(sm)}$ at the vertical transition point was set equal to zero. The equation for the characteristic intensity I_1 coincides then with (28), where the departure time for the light holes is

$$\tau_{2p}^{-1}=(\tau_{2p}^{-1})_{ac}+(\tau_{2p}^{-1})_{opt}.$$

where $(\tau_{2p})_{ac}$ is determined by Eq. (37), and $(\tau_{2p})_{opt}$ by the interaction of light holes with optical phonons[16,17]:

$$\left(\frac{1}{\tau_{2p}}\right)_{opt}=\frac{3d_0^2 m_1^{1/2}(\hbar\omega-\hbar\omega_0)^{1/2}}{2^{1/2}\pi\hbar^2\rho\hbar\omega_0}, \tag{46}$$

d_0 is the constant of the coupling with the optical phonons [for p-Ge (Ref. 19) we have $d_0 = 32$ eV/5.65 Å $= 9.07\cdot 10^{-4}$ erg/cm]. The departure time τ_p of the heavy holes is determined as before by their interaction (37) with the acoustic phonons. Calculation for p-Ge at $T = 77.4$ K and $\lambda = 10.6\,\mu$m yields for τ_{1p}, $(\tau_{2p})_{ac}$, $(\tau_{2p})_{opt}$ and τ_{2p} the values $5.1\cdot10^{-12}$, $2.2\cdot10^{-12}$, $0.68\cdot10^{-12}$ and $0.52\cdot10^{-12}$ s respectively (from (1) we have $\tau_p = 0.94\cdot10^{-12}$ s). The value of I_1 is then[3)] 15 kW/cm^2.

The expression for the characteristic intensity I_2 is determined by Eq. (33), where $\beta = e^{-x}i[\Phi(x_i) - \Phi(x_f)]$ and coincides with expression (11) of Ref. 8, where the values of I_s, which plays thus the role of the characteristic intensity I_2 in our notation. The numerical value of I_2 at $T = 77.4$ K is 590 kW/cm^2. The numerical value of I_s in Ref. 8, however turned out to be of the order of 600 kW/cm^2. The measured absorption coefficient α varied in this case with intensity like $\alpha = \alpha_0(1 + I/I_s)^{-1}$, which agrees with Eq. (5) at $I_1 = \infty$.

The nonlinearity connected with the coherent saturation of the vertical transition was thus not observed in the experiment of Ref. 8. One might think that here, just as in the preceding case of $\lambda = 100\,\mu$m, the coherent saturation of the absorption is destroyed by the electric field of the impurities. However, an estimate based on Eq. (44) with $\tau_p = 0.94\cdot10^{-12}$ s yields $N_i^{\min} = 1.1\cdot10^{14}$ cm^{-3}, whereas the measurements in Ref. 8 were made with specimens having hole densities from 10^2 to $3\cdot10^{15}$ cm^{-3}.

The most probable reason for this is the insufficient coherence of the radiation used in Ref. 8. Thus, the radiation phase loss can destroy the Rabi coherent oscillations.[14] Estimates show that the phase-loss time τ_α (which should replace τ_p in the expression for I_1) turns out to be of the order of $2.4\cdot10^{-14}$ s if I_1 is to be of the order of I_2. Thus, the phase loss should occur once during one period. Such a phase loss can take place if a small admixture of radiation with $\lambda = 9.6$ μm is mixed in with the 10.6-μm radiation.

It is easiest to observe in experiment coherent saturation of the absorption by studying the effect exerted on it by a weak electric field. Thus, for the considered case with $\lambda = 10.6\,\mu$m and $T = 77.4$ K the field $E_{\min}$ (42) turns out to be of the order of 12 V/cm.

In conclusion, a few words concerning the influence of "corrugation" and of the magnetic field on the nonlinear-absorption coefficient. In our opinion (this was also noted in Ref. 11), allowance for corrugations at $\lambda = 10.6\,\mu$m is important, since the energy region of the heavy holes that execute vertical transitions extends from 11 to 34 meV, while the final energies range from 0 to 3 and from 17 to 37 meV, corresponding to the energy of the optical phonon, i.e., the energy regions of departure and arrival of heavy holes overlap strongly and the use of the isotropic model for the calculation can yield only an estimate of the characteristic intensity I_2.

Another result of corrugation is that now a magnetic field can also dephase the coherent Rabi oscillations. The holes, moving over the quasi-energy surface, go off-resonance under the influence of the magnetic field. Since this region is narrow, the magnetic fields needed for this purpose are also weak.

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APPENDIX

Calculation of the heavy-hole distribution function involves the integrals[8]

$$s(x)=\int_{x_i}^{x}\frac{e^y}{y}\,dy. \tag{A1}$$

and

$$d=\frac{2}{\pi^{1/2}}\left[\int_{x_i}^{x_\kappa}x^{1/2}e^{-x}s(x)\,dx+s(x_\kappa)\int_{x_\kappa}^{\infty}x^{1/2}e^{-x}\,dx\right]. \tag{A2}$$

By simple transformations they can be reduced to the form

$$s(x)=\varphi(x)-\varphi(x_i), \tag{A3}$$

$$\varphi(x)=-\frac{e^x}{x}+\ln x+\int_0^x\frac{e^y-1}{y}\,dy, \tag{A4}$$

$$d=\Phi(x_i)-\Phi(x_f), \tag{A5}$$

where

$$\Phi(x)=-\ln x+4\left(\frac{x}{\pi}\right)^{1/2}+\frac{2}{\pi^{1/2}}\int_0^\infty dt$$
$$\times e^{-t}\left[\left(\frac{1}{x}-2\right)(t+x)^{1/2}+2t^{1/2}\ln\left((t+x)^{1/2}+t^{1/2}\right)\right]. \tag{A6}$$

The representation of these functions in the form (A4) and (A6) is convenient for computer calculations which were used to compile Table I. At those values of the argument x which are not contained in Table I, they can be determined with accuracy $\sim 10\%$ from the asymptotic expressions at small and large x:

$$\varphi(x)=\begin{cases}-\dfrac{1}{x}+\ln x, & x\to 0\\[2mm] \dfrac{e^x}{x^2}\left(1+\dfrac{2!}{x}+\dfrac{3!}{x^2}+\dots\right), & x\to\infty,\end{cases} \tag{A7}$$

$$\Phi(x)=\begin{cases}\dfrac{1}{x}-\ln x, & x\to 0\\[2mm] \dfrac{4}{(\pi x)^{1/2}}, & x\to\infty.\end{cases} \tag{A8}$$

[1)]In the last section of this article we consider the opposite case of short wavelengths, when the final energy of the light hole exceeds that of the optical phonon.

[2)]The approximation used in Ref. 7 corresponds to neglect of $A_p^2\tau_{1p}\tau_{2p}/\hbar$ compared with unity in the radicand of (25).

[3)]We note in this connection that numerical calculations[13] for p-Ge at the same values of temperature and wavelength the value obtained for I_1 was ≈ 200 kW/cm^3, which exceeds our value by an order of magnitude. At the same time, the employed deformation-potential constant $E_{ac} = 3.5$ eV (Ref. 11) yields for $\tau_{1_p}^{-1}$ on acoustic phonons a value an order of magnitude smaller than ours.

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