Mechanisms of energy relaxation under conditions of nonlinear absorption of light in *p*-type Ge

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An investigation was made of the nonlinear absorption of light as a result of intraband transitions in *p*-type Ge. A change in the mechanism of relaxation of hot holes took place in the interval of the densities of free holes $7 \cdot 10^{13}$–10^{14} cm^{-3} in such a way that in the range $p > 10^{14}$ cm^{-3} the relaxation process was due to hole-hole collisions, whereas in the range $p < 7 \cdot 10^{13}$ cm^{-3} the main energy relaxation mechanism was the interaction with long-wavelength acoustic phonons. The distribution function of free carriers was derived theoretically for various intensities of the exciting light. The calculated relaxation time for the interaction with acoustic phonons was $0.73 \cdot 10^{-10}$ sec, in good agreement with the experimental results ($0.9 \cdot 10^{-10}$ sec).

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Earlier studies[1–4] have revealed nonlinear absorption of light in the case of intraband transitions under the action of CO_2 laser radiation on p-type germanium. It has been shown[3,4] that at low temperatures an increase in the transmission is controlled by hole–hole collisions and depends consequently on the equilibrium hole density. Under these conditions the absorption coefficient α is a function of the intensity of light I:

$$\alpha(I) = \frac{\alpha_0}{1 + (I/I_s)}, \tag{1}$$

where α_0 is the absorption coefficient of light at low intensities. The saturation parameter I_S is governed by the hole–hole collision time

$$I_s = \frac{1}{\sigma \langle \tau_{hh} \rangle}. \tag{2}$$

Here, σ is the cross section for the absorption of light by holes and $\langle \tau_{hh} \rangle$ is the hole–hole collision time averaged over the nonequilibrium distribution function. The exact expression for $\langle \tau_{hh} \rangle$ obtained by solving the transport equation and allowing for optical transitions can be found in Ref. 4. Since the saturation parameter I_S is proportional to the free hole density p_0, it is possible to lower considerably the value of I_S by doping and thus make highly efficient fast-response nonlinear filters of p-type Ge operating at T = 78°K and suitable for mode locking in the case of CO_2 lasers.[5]

We investigated the nonlinear absorption of light as a result of intraband transitions in p-type Ge at relatively low hole densities in order to find that range of densities in which the increase in the transmission was governed by the acoustic scattering and not by electron–electron collisions. The results obtained made it possible to determine for the first time by direct experiment the density of free holes at which there was a change in the mechanism of energy relaxation of hot holes from electron–electron collisions to collisions with acoustic phonons. Moreover, these experiments provided an independent method for determining the constant governing the energy relaxation time in the case of interaction with acoustic phonons. Our investigation was also of practical importance because it enabled us to find the minimum possible value of I_S for p-type Ge.

1. An investigation was made of the nonlinear absorption in p-type Ge with a free hole density 10^{12}–$3 \cdot 10^{15}$ cm^{-3} at T = 78°K. The radiation source was a tunable CO_2 laser emitting at 10.6 μ, corresponding to a photon energy $\hbar\omega = 0.117$ eV. Excitation with such light causes transitions of holes from an initial state of energy ε_i = 23 meV in the heavy hole subband V_1 to a state $\varepsilon_0 = \varepsilon_i + \hbar\omega$ in the light hole subband V_2 (Fig. 1), which is 140 meV.

During the first stage the relaxation of a photohole is due to the emission of discrete optical phonons of energy $\hbar\omega_0 = 37$ meV, because the energy relaxation time for the interaction with optical phonons $\tau_{opt} \sim 10^{-13}$ sec is considerably shorter than other relaxation times in our case. The emission of optical phonons occurs until the energy of a photohole becomes less than the energy of an optical phonon. Since the density of states in the heavy subband is considerably greater than in the light subband, the final state of a photohole is in the heavy subband and its energy is $\varepsilon_f = \varepsilon_0 - 3\hbar\omega_0$, amounting to 29 meV for the laser photons used. The second stage of the relaxation occurs because of the interaction with the equilibrium holes and acoustic phonons. Since the relaxation time during the second stage is considerably longer than during the first, the total relaxation time representing the sum of the two and governing[3,4] the value of I_S is determined by the rate of relaxation during the second stage. The saturation parameter I_S is always given by Eq. (2) where in the case of low hole densities the time $\langle \tau_{hh} \rangle$ should be replaced by the energy relaxation time due to the interaction

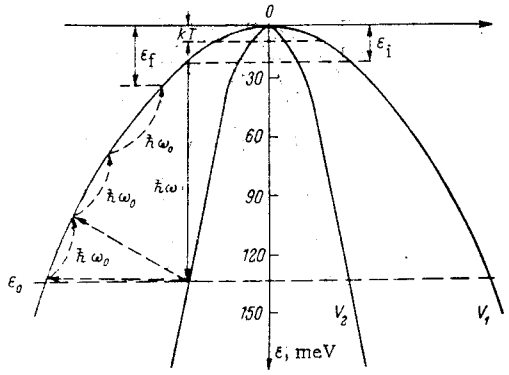

FIG. 1. Optical transitions and subsequent relaxation of photohole energy.

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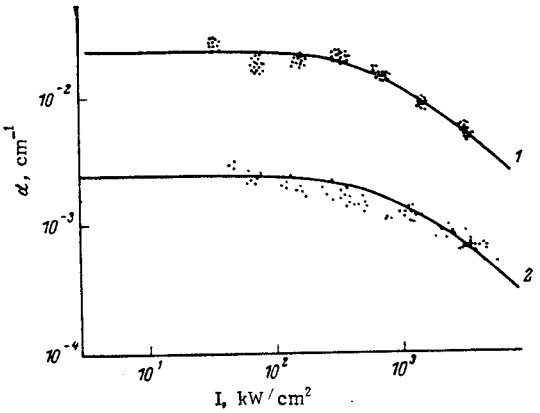

FIG. 2. Dependences of the absorption coefficient α on the intensity of light I at the wavelength λ = 10.6 μ. The points are the experimental results and the curves are calculated from Eq. (1). Hole density p (cm^{-3}): 1) $6.7 \cdot 10^{13}$; 2) $6.6 \cdot 10^{13}$. I_s (MW/cm^2): 1) 0.7; 2) 0.6.

with acoustic phonons $\langle \tau_{ac} \rangle$ averaged over the relevant distribution functions. Naturally, in this case there should be no dependence of I_s on the density of free holes. It follows that the following dependence of I_s on the free hole density p should be observed: at low densities the value of I_s should be independent of p, but beginning from a certain density it should rise and follow an almost linear dependence.

It is difficult to determine directly the absorption coefficient at low densities because the coefficient is then small. Therefore, the dependence of the absorption coefficient on the illumination intensity was determined using the photoelectric drag of carriers by photons, proportional to the absorption coefficient.[6] The experimental dependences $\alpha = f(I)$ obtained in this way are shown in Fig. 2. The same figure gives the theoretical dependences $\alpha \propto \alpha_0/1 + (I/I_s)$ with I_s as a variable parameter. We can see that the experiment and theory agree well. The values of I_s obtained for samples with different impurity concentrations are plotted in Fig. 3.

As predicted, the dependence of I_s on the carrier density consists in practice of two regions. In the region $p < 7 \cdot 10^{13}$ cm^{-3} the value of I_s is independent on the density and amounts to 0.6 MW/cm^2, whereas for $p > 10^{14}$ cm^{-3} right up to $2 \cdot 10^{15}$ cm^{-3} the value of I_s increases on increase in p. Application of the formula for I_s was used to find the experimental values of $\langle \tau \rangle$ plotted in Fig. 4. A comparison of the theory and experiment in the case of dominion of the energy relaxation mechanism by the hole–hole collisions was made by us earlier.[3]

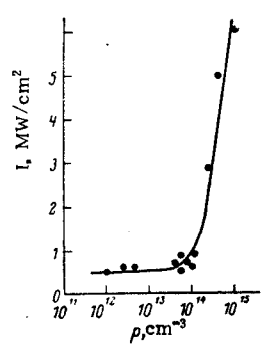

FIG. 3. Hole-density dependence of the saturation parameter I_s found experimentally for λ = 10.6 μ.

2. We shall consider here in detail the case when the main mechanism governing the behavior of I_s is the energy relaxation by the interaction with acoustic phonons.

In order to calculate $\langle \tau_{ac} \rangle$ we have to know the distribution function established under the influence of light. If the dominant energy relaxation mechanism involves acoustic phonons, the transport equation is

$$\frac{\alpha(I) I}{\rho(\varepsilon)} [\delta(\varepsilon - \varepsilon_k) - \delta(\varepsilon - \varepsilon_i)] + \frac{1}{\rho(\varepsilon)} \frac{dj_\varepsilon}{d\varepsilon} = 0, \quad (3)$$

where $\rho(\varepsilon) = m_h^{3/2}(2\varepsilon)^{1/2}/\pi^2\hbar^3$ is the density of states,

$$\alpha(I) = \frac{\alpha_0 f(\varepsilon_i)}{f_p(\varepsilon_i)} \quad (4)$$

is the absorption coefficient corresponding to a given light intensity, α_0 and $f_p(\varepsilon_i)$ are the absorption coefficient and the distribution function of holes under thermodynamic equilibrium conditions. The quantity $j(\varepsilon)$ is given by

$$j(\varepsilon) = \frac{\varepsilon}{\tau_\varepsilon} \rho(\varepsilon) \left[f(\varepsilon) + kT \frac{df(\varepsilon)}{d\varepsilon} \right] \quad (5)$$

and it represents a flux directed toward lower energies and resulting from the interaction with acoustic phonons (see, for example, Ref. 7). The energy relaxation time for the interaction with acoustic phonons can be written conveniently in the form

$$\tau_\varepsilon = l_0 \sqrt{\frac{m_h}{2\varepsilon}}, \quad (6)$$

where l_0 is a characteristic length related to the deformation potential constant. The mobility data are used in Ref. 7 to find l_0 for p-type Ge and this gives $l_0 = 4.3 \cdot 10^{-3}$ cm.

In the derivation of Eq. (3) it is assumed that all the holes are excited from an initial state ε_i and drop to a final state ε_f after emitting an optical phonon. The outgoing and incoming terms can be written in the form of δ functions provided we ignore the width of the exciting laser line, corrugations of the heavy hole subband, and dispersion of optical phonons. Dividing the domain of integration with respect to ε into three regions [1) $\varepsilon < \varepsilon_i$, 2) $\varepsilon_i < \varepsilon < \varepsilon_f$, 3) $\varepsilon_f < \varepsilon$] and integrating Eq. (3), bearing in mind that $j(\varepsilon)$ vanishes for $\varepsilon > \varepsilon_f$ because there is no flux from infinity as a result of the interaction with acoustic phonons, we find that if $\varepsilon < \varepsilon_i$, then $j(\varepsilon) = j_1(\varepsilon) = 0$, whereas for $\varepsilon_i < \varepsilon < \varepsilon_f$, we have $j(\varepsilon) = j_2(\varepsilon) = \alpha I$ and for $\varepsilon > \varepsilon_f$, we obtain $j(\varepsilon) = j_3(\varepsilon) = 0$. Integrating these ex-

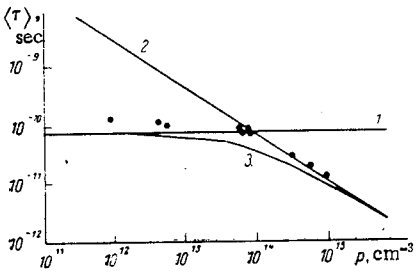

FIG. 4. Dependences of the average energy relaxation times on the hole density p. The points are the experimental values and the curves are theoretical: 1) $\langle \tau_{ac} \rangle$; 2) $\langle \tau_{hh} \rangle$; 3) $\langle \tau \rangle = [\langle \tau_{ac} \rangle^{-1} + \langle \tau_{hh} \rangle^{-1}]^{-1}$.

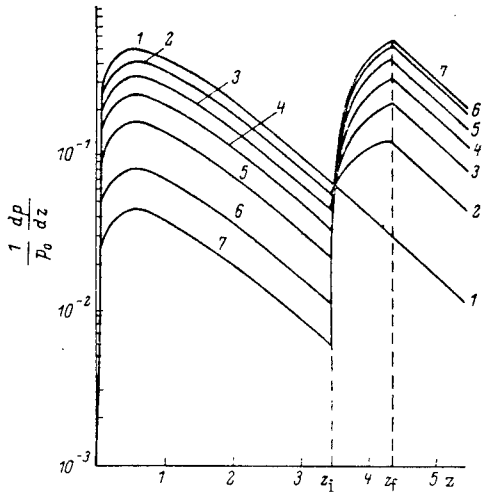

FIG. 5. Normalized distribution function of holes $(1/p_0)(dp/dz)$ obtained for different light intensities I: 1) 0; 2) $0.2I_S$; 3) $0.5I_S$; 4) I_S; 5) $2I_S$; 6) $5I_S$; 7) $10I_S$.

pressions for the fluxes and matching the distribution functions at the points ε_i and ε_f, we obtain

$$\left.\begin{aligned} f_1 &= Ce^{-\varepsilon/kT}, \quad \varepsilon < \varepsilon_i, \\ f_2 &= Ce^{-\varepsilon/kT} + \frac{\alpha I}{kT} e^{-\varepsilon/kT} \int_{\varepsilon_i}^{\varepsilon} \frac{\tau(\varepsilon') e^{\varepsilon'/kT}}{\varepsilon' \rho(\varepsilon')} d\varepsilon', \quad \varepsilon_i < \varepsilon < \varepsilon_f, \\ f_3 &= Ce^{-\varepsilon/kT} + \frac{\alpha I}{kT} e^{-\varepsilon/kT} \int_{\varepsilon_i}^{\varepsilon_f} \frac{\tau(\varepsilon') e^{\varepsilon'/kT}}{\varepsilon' \rho(\varepsilon')} d\varepsilon', \quad \varepsilon > \varepsilon_f, \end{aligned}\right\} \tag{7}$$

where C is the constant of integration. It should be noted that the absorption coefficient $\alpha(I)$ itself depends on the distribution function. However, in the case of a δ-like source it follows from Eq. (4) that this coefficient depends only on the values of the distribution functions for an energy ε_i. Using Eqs. (4) and (7), we obtain

$$\alpha = \frac{\alpha_0 C}{C_0}, \tag{8}$$

where $C_0 = (p_0/2)(2\pi\hbar/m_h kT)^{3/2}$ is the normalization coefficient of the equilibrium distribution. Substituting Eq. (8) into Eq. (7), we can now determine this constant from the normalization condition

$$\int_0^\infty f(\varepsilon) \rho(\varepsilon) d\varepsilon = p_0, \tag{9}$$

which gives

$$C = \frac{C_0}{1 + I/I_s}. \tag{10}$$

Here, I_S is given by the expression

$$I_s^{-1} = \alpha l_0 \sqrt{\frac{m_h}{2kT}} \left\{ \int_{z_i}^{z_f} e^{-z}\sqrt{z}\, dz \int_{z_i}^{z} \frac{e^{z'} dz'}{(z')^2} + \int_{z_f}^{\infty} e^{-z}\sqrt{z}\, dz \int_{z_i}^{z_f} \frac{e^{z'} dz'}{(z')^2} \right\}. \tag{11}$$

In the above equation we have $z = \varepsilon/kT$, where z_i and z_f are the corresponding values for the initial ε_i and final ε_f energies. It follows from Eq. (11) that $\langle \tau_{ac} \rangle$ corresponding to $l_0 = 4.3 \cdot 10^{-3}$ cm^{-3} (Ref. 7) is $0.73 \cdot 10^{-10}$ sec, which is close to the experimental values of $\langle \tau \rangle$ found in the region of the plateau of the carrier-density dependence (Fig. 4).

We shall conclude by considering the form of the distribution function of holes under the optical excitation conditions considered here (Fig. 5). Clearly, optical excitation gives rise to a fall of the distribution function at energies $\varepsilon < \varepsilon_i$ and to a rise in the range $\varepsilon > \varepsilon_f$. The energy distribution of holes changes and becomes of the two-humped nature, which is a specific feature of monochromatic optical excitation. At high illumination intensities $(I \geqslant 2I_S)$ the bulk of the holes is located in the second hump. Under these conditions we can expect various instabilities in the hole system.

3. Figure 4 gives the experimental data for the average relaxation time $\langle \tau \rangle$. It includes also the theoretical values $\langle \tau_{ac} \rangle$ (curve 1) and $\langle \tau_{hh} \rangle$ (curve 2) obtained from the calculation formulas given above and in Ref. 4. Curve 3 corresponds to the interpolation formula $\tau = [\langle \tau_{ac} \rangle^{-1} + \langle \tau_{hh} \rangle^{-1}]^{-1}$, which allows us to describe approximately the transition range. We can see that the agreement between the theoretical and experimental times is good.

The above results enabled us to use the experimental data to show directly that a change in the mechanism of relaxation of hot holes occurs in the interval $7 \cdot 10^{13}$–10^{14} cm^{-3} so that in the range $p > 1 \cdot 10^{14}$ cm^{-3} the energy relaxation process is governed by the hole–hole collisions, whereas in the range $p < 7 \cdot 10^{13}$ cm^{-3} the main energy relaxation mechanism is the interaction with long-wavelength acoustic phonons.

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