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CONTRIBUTION TO THE THEORY OF OPTICAL EXCITATION OF SEMICONDUCTORS. ABSORPTION AND DISPERSION IN SINGLE-PHOTON AND TWO-PHOTON PROCESSES

Yu. L. KLIMONTOVICH and É. V. Pogorelova

Moscow State University

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Interband absorption and dispersion in semiconductors subjected to the action of a strong electromagnetic field is calculated. The calculation is performed for single-photon and two-photon processes with account taken of electron deceleration in the bands. It is shown that under the action of a field of frequency ω_1 ($\hbar\omega_1$ is greater than the forbidden band width in single-photon processes and $\hbar\omega_1 > \Delta/2$ for two-photon processes) the absorption-band edge shifts towards higher frequencies. For frequencies $\Delta < \hbar\omega < \Delta + \Delta_E$ the absorption is negative. The quantity Δ_E determines the position of the band edge in the presence of the field and increases with increasing field strength. The value of the saturating field strength at which the Δ_E maximal is calculated to be $\hbar\omega_1 - \Delta$ for single-photon processes and $2\hbar\omega_1 - \Delta$ for two-photon processes. The absorption coefficient vanishes in this case at a frequency ω_1 (or $2\omega_1$) and hence further increase of the field strength does not lead to a shift of the band edge. It is shown that with increasing field the dispersion-curve maximum also shifts towards higher frequencies, i.e., in the same direction as the absorption-band-edge shift. The results are used for explaining some processes in semiconductor lasers with optical pumping and the transparency effect in semiconductor filters.

INTRODUCTION

WHEN sufficiently strong electromagnetic radiation from a laser acts on a semiconductor, a number of effects are produced, which depend essentially on the magnitude of the field and cannot be described within the framework of ordinary perturbation theory. These include effects due to occurrence of states with negative temperature in semiconductors (semiconductor laser with optical pumping)[1,2], bleaching of semiconductor filters in a strong field, reduction in the absorption at a given frequency (saturation effect), and others. The present paper is devoted to the investigation of the foregoing phenomena with allowance for single-photon and two-photon processes.

The calculations of two-photon absorption presented in[3-5] have been obtained by perturbation theory and do not describe the aforementioned effects. The saturation effect in semiconductors with electron injection through a p-n junction is the subject of a paper by Krokhin[6]. Basov and Krokhin[7,8] investigated the optical excitation of semiconductors. The saturation in optical pumping was first described in[7]. The expressions presented in[6-8] for the difference of the Fermi quasilevels and for the gain, with account of saturation, are valid near the maximum of the gain and consequently do not describe the course of the absorption (positive or negative) in the entire frequency region. We shall present below a more detailed comparison with the results of these papers.

1. DISTRIBUTION OF ELECTRONS AMONG THE BANDS IN OPTICAL EXCITATION WITH ACCOUNT OF DECELERATION

We investigated earlier[9] single-photon absorption processes and the dispersion under conditions when the deceleration time of the electrons in the band exceeds the lifetime in the conduction band (the recombination time). In this case, just as in a quantum system with a discrete spectrum, the absorption coefficient and the dispersion at frequency ω in a strong field are determined by the value of the field at the same frequency. In the opposite case of short deceleration time, the situation is radically altered. Owing to the deceleration within a time shorter than the recombination time, a quasi-equilibrium distribution of the electrons in the bands is established.[10] At the same time, the electron density increases at the lower edge of the conduction band, as a result of which a radiation is produced at frequencies lower than the frequencies of the given field—the pump field. This process is used in semiconductor lasers with optical pumping, which were described in the papers of Basov et al.[1,2]

To describe the process of polarization of a semiconductor in a strong field, we cannot confine ourselves to the single-particle approximation if we take deceleration into account. We therefore assume that the intensity of the electric field is determined by the expression

$$E(t) = {}^1\!/_2(\mathrm{E}e^{-i\omega t} + \mathrm{E}_1e^{-i\omega_1 t} + \mathrm{c.c.}). \tag{1.1}$$

Here ω_1 is the frequency of the given field—the pump field—and ω is an arbitrary frequency at which the emission of absorption is considered.

The field at the frequency ω_1 is strong, so that account must be taken of the nonlinear dependence of the polarization on the field. To describe the saturation effect in single-photon processes it is sufficient to include the first harmonic of the polarization (relative to ω_1), at which resonance takes place, and the first harmonic of the population difference between the bands. Inclusion of the second harmonic of the population difference leads to a change in the width of the forbidden band[9].

We shall use an expression derived by us earlier[9]:

$$\mathbf{P} = -\frac{e^2n}{3}\int \times \frac{|r_\mathbf{p}|^2D(\mathbf{p})\{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega]\cos\omega t + (\hbar/\tau_2)\sin\omega t\}}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega]^2 + \hbar^2/\tau_2{}^2}d\mathbf{p}. \tag{1.2}$$

Here $D(\mathbf{p}) = \rho_c(\mathbf{p}) - \rho_v(\mathbf{p})$ is the difference in the electron populations in bands with given momentum $\mathbf{p}$. When account is taken of the deceleration, the function $D(\mathbf{p})$ is no longer determined by formula (2.3) of[9]. We shall denote it by $D_1(\mathbf{p})$. To determine $D_1(\mathbf{p})$, we proceed as follows: deceleration causes quasi-equilibrium distributions to be established in the bands. We write them in the form

$$f_c(\mathscr{E}) = \left[\exp\frac{m^*}{m_c}\frac{\mathscr{E} - \Delta_E}{kT} + 1\right]^{-1}, \tag{1.3}$$

$$f_v(\mathscr{E}) = \left[\exp\frac{m^*}{m_v}\frac{\Delta_E - \mathscr{E}}{kT} + 1\right]^{-1}. \tag{1.4}$$

The subscripts v and c denote quantities pertaining respectfully to the valence and conduction bands. The quantity $\mathscr{E}$ is defined by the expression

$$\mathscr{E} = \frac{\mathbf{p}^2}{2m_c} + \frac{\mathbf{p}^2}{2m_v} = \frac{\mathbf{p}^2}{2m^*}; \quad m^* = \frac{m_cm_v}{m_c + m_v}. \tag{1.5}$$

The quantity Δ_E is connected with the Fermi quasi-levels introduced in the papers of Krokhin and Basov[6-8] by the relations

$$\Delta_E = \frac{m_c}{m^*}\mu_c = \frac{m_v}{m^*}(|\mu_v| - \Delta) \tag{1.6}$$

and depends on the intensity and frequency of the pump field. On the other hand, the quantity Δ_E is connected with the number n_1 of electrons in the conduction band:

$$n_1 = \frac{2}{(2\pi\hbar)^3}\int f_cd\mathbf{p} = n - \frac{2}{(2\pi\hbar)^3}\int f_vd\mathbf{p}, \tag{1.7}$$

$n = N/V$, N—total number of electrons.

At zero temperature we have

$$f_c(\mathscr{E}) = 1 \text{ for } 0 < \mathscr{E} < \Delta_E; \quad f_c(\mathscr{E}) = 0 \text{ for } \mathscr{E} > \Delta_E;$$
$$f_v(\mathscr{E}) = 1 \text{ for } \Delta_E < \mathscr{E} < \Delta_v; \ f_v(\mathscr{E}) = 0 \text{ for } 0 < \mathscr{E} < \Delta_E. \tag{1.8}$$

Δ_v is determined by the width of the valence band and is connected with the true width of the valence band by the relation

$$\Delta_v = (m_v/m^*)\Delta_{v,\mathrm{true}}. \tag{1.9}$$

The connection between Δ_E and n_1 at T = 0 is determined by the expression

$$\Delta_E = (3\pi^2)^{2/3}(\hbar^2/2m^*)n_1^{2/3}. \tag{1.10}$$

The population difference is equal to

$$D_1(\mathbf{p}) = \frac{2}{(2\pi\hbar)^3}(f_c(\mathbf{p}) - f_v(\mathbf{p})) \tag{1.11}$$

and consequently is determined entirely by the value of Δ_E.

To determine Δ_E we proceed in the following manner: We write the equation for D(p) without account of deceleration. Its form is (see (2.3) in[9])

$$\frac{1}{\tau_2}\frac{e^2|\mathbf{r}_\mathbf{p}\mathbf{E}_1|^2D(\mathbf{p})}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega_1]^2 + \hbar^2/\tau_2{}^2} = -\frac{1}{\tau_1}(D(\mathbf{p}) - D^0(\mathbf{p})), \tag{1.12}$$

D^0 is the equilibrium distribution, $\mathbf{E}_1$ the field intensity at the frequency ω_1, τ_1 the recombination time, and τ_2 the relaxation time of the off-diagonal elements of the density matrix. It follows therefore that the increase occurring in the time τ_1 in the population difference summed over all the momenta is given by the expression

$$\int Dd\mathbf{p} = \int D^0(\mathbf{p})\frac{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega_1]^2 + \hbar^2/\tau_2{}^2}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega_1]^2 + \hbar^2/\tau_2{}^2 + R_1{}^2}d\mathbf{p},$$
$$R_1{}^2 = \frac{\tau_1}{\tau_2}e^2|\mathbf{r}_\mathbf{p}\mathbf{E}_1|^2. \tag{1.13}$$

We use the normalization condition

$$\int[\rho_c(\mathbf{p}) + \rho_v(\mathbf{p})]d\mathbf{p} = \int[\rho_c{}^0(\mathbf{p}) + \rho_v{}^0(\mathbf{p})]d\mathbf{p} = n \tag{1.14}$$

and obtain an expression for the number of electrons that gathered by the field into the conduction band in the time τ_1:

$$\int \rho_c d\mathbf{p} - \int \rho_c^0 d\mathbf{p} \tag{1.15}$$

$$= -\frac{1}{2}\int \frac{(\rho_c^0 - \rho_v^0) R_1^2}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega_1]^2 + \hbar^2/\tau_2^2 + R_1^2}\, d\mathbf{p}.$$

Owing to deceleration, the quasi-equilibrium distributions (1.3) and (1.4) are established after a time $\tau_3 \ll \tau_1$ or τ_2. From (1.7) and (1.15) we obtain an expression for the number n_1 of electrons in the conduction band:

$$n_1 = \int \rho_c^0 d\mathbf{p} \tag{1.16}$$

$$-\frac{1}{2}\int \frac{(\rho_c^0 - \rho_v^0) R_1^2}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega_1]^2 + \hbar^2/\tau_2^2 + R_1^2}\, d\mathbf{p}.$$

The second term in the right side of (1.16) determines the increase in the electron density in the conduction band, due to the action of the field at the frequency ω_1. It vanishes at $\mathbf{E}_1 = 0$.

Let us obtain an expression for Δ_E at zero temperature. In this case $\rho_c^0 = 0$ and $\rho_v^0 = 2/(2\pi\hbar)^3$ at $\Delta_v > \mathscr{E} > 0$. For n_1 we obtain from (1.16)

$$n_1 = \frac{\sqrt{2}}{2}\frac{(m^*)^{3/2} R_1^2}{\pi^2\hbar^3}\int_0^{\Delta_v} \frac{\sqrt{\mathscr{E}}\, d\mathscr{E}}{(\mathscr{E} + \Delta - \hbar\omega_1)^2 + \hbar^2/\tau_2^2 + R_1^2} \tag{1.17}$$

Since $R_1^2 \ll \Delta_v$, the upper limit in this expression can be set equal to infinity. Carrying out the integration and using formula (1.10), we obtain the following expression for Δ_E:

$$\Delta_E = \Delta_E^0 \left(\left[1 + \frac{(\hbar\omega_1 - \Delta)^2}{\hbar^2/\tau_2^2 + R_1^2}\right]^{1/2} - \frac{\hbar\omega_1 - \Delta}{\sqrt{\hbar^2/\tau_2^2 + R_1^2}}\right)^{-1/3} \tag{1.18}$$

for $\hbar\omega_1 - \Delta \geq \Delta_E$. Here

$$\Delta_E^0 = \frac{1}{2}\left(\frac{3}{2}\pi\right)^{2/3} \frac{R_1^{4/3}}{(\hbar^2/\tau_2^2 + R_1^2)^{1/6}}. \tag{1.19}$$

The condition $\hbar\omega_1 - \Delta \geq \Delta_E$ in formula (1.18) denotes that when a field of frequency ω_1 is applied, the filling of the conduction band increases with increasing field only until Δ_E becomes equal to $\hbar\omega_1 - \Delta$. Under this condition (see the next section), the absorption coefficient of the field at the frequency $(\Delta + \Delta_E)/\hbar$ vanishes. Thus, if $\hbar\omega_1 - \Delta = \Delta_E$ and the frequency Δ_1 is specified, expression (1.18) turns into an equation for the limiting value of the field—the saturation field. We denote it by $\mathbf{E}_S$ and the corresponding $\Delta_E = \hbar\omega_1 - \Delta$ by $\Delta_{\max}$.

To find the value of the saturation field let us consider the equation

$$x^6 - 2cx^4 - c^2 = 0, \tag{1.20}$$

$$x = \Delta_{max}(\hbar^2/\tau_2^2 + R_1^2)^{-1/2}; \quad c = (\Delta_E^0)^3(\hbar^2/\tau_2^2 + R_1^2)^{-1/6},$$

which follows from (1.18) when $\hbar\omega_1 - \Delta = \Delta_{\max}$. We present a solution of this equation for two extreme cases.

A. The saturation occurs in a weak field:

$$\left(\frac{\tau_1}{\tau_2}\right)^{1/2} \frac{erE_S}{\sqrt{3}} \ll \frac{\hbar}{\tau_2}.$$

In this case it follows from (1.20) that

$$\Delta_{max} \approx 1.4(\hbar/\tau_2)^{2/3} R_1^{1/3}. \tag{1.21}$$

Hence, recognizing that $\Delta_{\max} = \hbar\omega_1 - \Delta$, we get

$$\left(\frac{\tau_1}{\tau_2}\right)^{1/2} \frac{erE_S}{\sqrt{3}} \approx \left(\frac{\hbar\omega_1 - \Delta}{\hbar/\tau_2}\right)^3 \frac{\hbar}{\tau_2}. \tag{1.22}$$

Using the criterion for the weakness of the field, we find from (1.22) that saturation is reached in a weak field only near threshold, when

$$\hbar\omega_1 - \Delta \ll \hbar/\tau_2. \tag{1.23}$$

B. Saturation occurs in a strong field:

$$\left(\frac{\tau_1}{\tau_2}\right)^{1/2} \frac{erE_S}{3} \gg \frac{\hbar}{\tau_2}$$

In this case we get from (1.20)

$$\Delta_{max} \approx 6{,}3 R_1 \text{ or } erE_S \approx \frac{\sqrt{3}}{6{,}3}\left(\frac{\tau_1}{\tau_2}\right)^{1/2} (\hbar\omega_1 - \Delta). \tag{1.24}$$

Thus, the saturating field increases with increasing distance from the edge of the band, i.e., with increasing $\hbar\omega_1 - \Delta$.

We have considered so far the case $T = 0$. When the temperature differs from zero, it is necessary to use (1.16) in lieu of (1.17). When $T \neq 0$, the integration can be carried out only if

$$\hbar\omega_1 - \Delta \gg (\hbar^2/\tau_2^2 + R_1^2)^{1/2}, \tag{1.25}$$

i.e., far from the edge of the band. Under this condition, the expression for n_1 takes the form

$$n_1 = n_c^0 - \frac{\sqrt{2}}{2}\frac{(m^*)^{3/2} R_1^2 (\hbar\omega_1 - \Delta)^{1/2}}{\pi\hbar^3(\hbar^2/\tau_2^2 + R_1^2)^{1/2}}$$

$$\times\, (f_c^0(\hbar\omega_1 - \Delta) - f_v^0(\hbar\omega_1 - \Delta)). \tag{1.26}$$

Here n_c^0 is the equilibrium concentration of the electrons in the conduction band.

In order to find the value of the saturating field when $T \neq 0$, we must put $\hbar\omega_1 - \Delta = \Delta_{\max}$ in (1.26) and use the expression for n_1 at $\Delta_E = \Delta_{\max}$, which follows from (1.3)—(1.6):

$$n_1 = \frac{2}{(2\pi\hbar)^3}\int \left[\exp\left(\frac{m^*}{m_c}\frac{\mathscr{E} - \Delta_{max}}{kT}\right) + 1\right]^{-1} d\mathbf{p}. \tag{1.27}$$

We do not present here any concrete results for $T \neq 0$. To obtain these results we must take into account the heating of the electrons in interactions with the lattice. With this, the electron temperature T in (1.25) and (1.26) depends itself on the field.

2. ABSORPTION COEFFICIENT

The absorption coefficient is determined by the imaginary part of the polarizability $\alpha''(\omega)$. From (1.2) we get

$$\alpha''(\omega) = -\frac{\sqrt{2}}{6}\frac{e^2r^2}{\pi^2\hbar^3}(m^*)^{3/2}I, \tag{2.1}$$

where

$$I = \frac{\hbar}{\tau_2} \times \left\{2\int_0^{\Delta_E}\frac{\sqrt{\mathscr{E}}\,\Delta\mathscr{E}}{[\mathscr{E}-(\hbar\omega-\Delta)]^2+\hbar^2/\tau_2^2} - \int_0^{\infty}\frac{\sqrt{\mathscr{E}}\,d\mathscr{E}}{[\mathscr{E}-(\hbar\omega-\Delta)]^2+\hbar^2/\tau_2^2}\right\}. \tag{2.2}$$

We have already taken into account here the fact that $D(p) = 2(2\pi\hbar)^{-3}[f_c(p) - f_v(p)]$, and used expressions (1.8) and (1.9).

After integration of (2.2) we get

$$I = \frac{\hbar}{\tau_2}\left\{\frac{1}{p}\ln\frac{|\Delta_E - p\sqrt{\Delta_E}+q|}{\Delta_E+p\sqrt{\Delta_E}+q} - \frac{\pi}{2\sqrt{q-p^2/4}} + \frac{1}{\sqrt{q-p^2/4}}\left(\operatorname{arctg}\frac{\sqrt{\Delta_E}+p/2}{(q-p^2/4)^{1/2}} + \operatorname{arctg}\frac{\sqrt{\Delta_E}-p/2}{(q-p^2/4)^{1/2}}\right)\right\} \tag{2.3*}$$

where

$$p = \sqrt{2}\{[(\hbar\omega-\Delta)^2+\hbar^2/\tau_2^2]^{1/2} + (\hbar\omega-\Delta)\}^{1/2},$$
$$q = [(\hbar\omega-\Delta)^2+\hbar^2/\tau_2^2]^{1/2}. \tag{2.4}$$

The quantity Δ_E in (2.3) is itself a function of the frequency and of the pump field and is determined by expression (1.8), while ω is the running frequency at which the absorption is determined.

Let us consider several particular cases.

1. $(\hbar\omega - \Delta) \gg \hbar/\tau_2$ or Δ_E, i.e., the frequency is far from the absorption edge. From (2.2) and (2.3) we get

$$\alpha''(\omega) = \frac{\sqrt{2}}{6}\frac{e^2r^2(m^*)^{3/2}}{\pi\hbar^3}\sqrt{\hbar\omega-\Delta}. \tag{2.5}$$

Thus, at a frequency much higher than the frequency of the pump field ω_1 the absorption coefficient is not sensitive to this field at all; ordinary absorption takes place in this case.

2. In the other limiting case, when $\hbar\omega - \Delta \ll \Delta_E$ but far from resonance, as before, $\hbar\omega - \Delta \gg \hbar/\tau_2$, we have

$$\alpha''(\omega) = -\frac{\sqrt{2}}{6}\frac{e^2r^2(m^*)^{3/2}}{\pi\hbar^3}\sqrt{\hbar\omega-\Delta}. \tag{2.6}$$

In this frequency band, inversion exists and the absorption coefficient becomes negative.

3. The quantum energy $\hbar\omega$ coincides with the width of the forbidden band Δ: $\hbar\omega - \Delta = 0$. With this: a) in the strong-field approximation, i.e., when $\Delta_E \gg \hbar/\tau_2$

$$\alpha'' = -\frac{e^2r^2(m^*)^{3/2}}{6\pi\hbar^3}\left(\frac{\hbar}{\tau_2}\right)^{1/2} < 0 \tag{2.7}$$

and b) in the weak-field approximation, when $\Delta_E \ll \hbar/\tau_2$,

$$\alpha'' = \frac{e^2r^2(m^*)^{3/2}}{6\pi\hbar^3}\left(\frac{\hbar}{\tau_2}\right)^{1/2} > 0. \tag{2.8}$$

4. When the pump field $\mathbf{E}_1$ tends to the value $\mathbf{E}_s$ defined by expressions (1.22) and (1.24), Δ_E tends to $\hbar\omega_1 - \Delta$ and $\alpha''(\omega_1) = 0$. Thus, a strong external field of frequency ω_1 exerts different influences on the absorption coefficient in different frequency ranges. At frequencies $\hbar\omega > \Delta + \Delta_E$, ordinary absorption takes place, when $\hbar\omega = \Delta + \Delta_E$ we have for the imaginary part $\alpha''(\omega) = 0$, and for frequencies $\hbar\omega < \Delta + \Delta_E$, owing to the action of the field $\mathbf{E}_1$, a state with negative temperature is produced, and amplification is possible for these frequencies.

The qualitative variation of $\alpha''(\omega)$ is shown in Fig. 1. The dashed curve shows $\alpha''(\omega)$ when $\hbar/\tau_2 \to 0$. An increase of the field at the frequency ω_1 leads to saturation. In a weak field $\alpha''(\omega_1) > 0$, but when $\mathbf{E}_1$ tends to $\mathbf{E}_s$ the absorption coefficient decreases and becomes equal to zero when $\mathbf{E}_1 = \mathbf{E}_s$, i.e., this system does not absorb the electromagnetic radiation incident on it. As seen from Fig. 1, this effect takes place when Δ_E shifts to the point A where $\hbar\omega = \hbar\omega_1 - \Delta$.

It follows from the foregoing reasoning that Δ_E characterizes the edge of the absorption band. Since Δ_E shifts to the right along the abscissa axis with increasing $\mathbf{E}_1$, the absorption edge itself also shifts towards larger values of the energy. This mechanism is the basis of bleaching of semiconductor filters. This phenomenon was observed long ago in InSb[11,12].

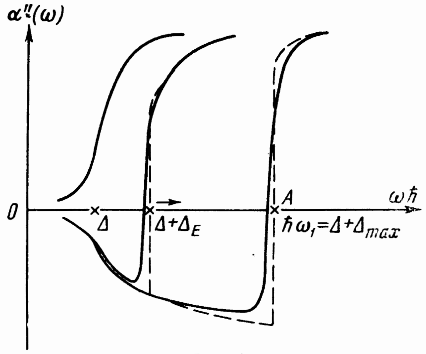

FIG. 1

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*arctg ≡ tan⁻¹.

Let us compare the obtained results with those of[6-8]. Krokhin[6] obtained expressions for the difference of the Fermi quasi-levels and for the gain k(ω) (formulas (24) and (25) of[6]). These results are valid near the maximum of the gain, under conditions when a negative temperature state is produced by electron injection through a p-n junction, i.e., it is not due to optical pumping. Consequently, the results of[6-8] have no direct bearing on the question considered here.

Saturation in optical excitation was described by Basov and Krokhin[7]. Saturation means that the quantity $\mu_C - \mu_V$ approaches $\hbar\omega_1$ when the field is increased. In our notation this corresponds to $\Delta_E \rightarrow \Delta_{max} = \hbar\omega_1 - \Delta$. In this respect, the saturation mechanism considered here coincides with that assumed in[7]. However, the character of the dependence of Δ_E on $\mathbf{E}_1$ and ω_1, and the dependence of the absorption coefficient on ω, ω_1, and $\mathbf{E}_1$ differs greatly in the present work from that obtained in[7]. In particular, when $\hbar\omega_1 - \Delta$ decreases (i.e., on approaching the edge of the absorption band in the absence of a field) the magnitude of the saturating field decreases (see (1.22) and (1.24)), whereas in[7] it increases. The results of[6-8] enable us only to indicate the direction of the change in the difference between the quasi-level energies, but the magnitude of the saturating field cannot be determined. The reason is that no account was taken in[6-8] of the fact that the time τ_2 is finite. Consequently no correct results can be obtained near the critical points when $\omega\hbar = \Delta$ (the edge of the band at $\mathbf{E}_1 = 0$) and $\hbar\omega - \Delta = \Delta_E$ (edge of the band in the presence of a field).

We now consider the effect of the electron deceleration on the dispersion in a strong field. This question has apparently not been considered heretofore.

3. THE DISPERSION CURVE

From (1.2) we can obtain not only the absorption coefficient but also the dispersion curve. It is determined by the real part of the polarizability $\alpha'(\omega)$:

$$\alpha'(\omega) = -\frac{e^2 n}{3}\int \frac{|r_{\mathbf{p}}|^2 D(\mathbf{p})[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega]}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - \hbar\omega]^2 + \hbar^2/\tau_2^2}\, d\mathbf{p}. \tag{3.1}$$

After integration, under the same assumptions as in Sec. 2, we obtain

$$\alpha'(\omega) = -\frac{\sqrt{2}e^2 r^2 (m^*)^{3/2}}{6\pi^2\hbar^3}\Big\{4\sqrt{\Delta_E} + \frac{p}{2}\ln\left|\frac{\Delta_E - p\sqrt{\Delta_E} + q}{\Delta_E + p\sqrt{\Delta_E} + q}\right|$$

$$-2\left(q - \frac{p^2}{4}\right)^{1/2}\left(\operatorname{arctg}\frac{\sqrt{\Delta_E} + p/2}{\sqrt{q - p^2/4}} + \operatorname{arctg}\frac{\sqrt{\Delta_E} - p/2}{\sqrt{q - p^2/4}}\right)$$

$$-2\sqrt{\Delta_v} - \frac{p}{4}\ln\left|\frac{\Delta_v - p\sqrt{\Delta_v} + q}{\Delta_v + p\sqrt{\Delta_v} + q}\right| \tag{3.2}$$

$$+\left(q - \frac{p^2}{4}\right)^{1/2}\left(\operatorname{arctg}\frac{\sqrt{\Delta_v} + p/2}{\sqrt{q - p^2/4}} + \operatorname{arctg}\frac{\sqrt{\Delta_v} - p/2}{\sqrt{q - p^2/4}}\right)\Big\};$$

p and q are defined in (2.4), and Δ_V and Δ_E are defined in Sec. 1.

In the absence of an external field, the dispersion curve is described by expression (4.2) of[9], provided we put A = 1 in the expressions for p and q, which enter in (4.2). The function $\alpha'(\omega)$ has a maximum at $\omega\hbar = \Delta$ and a minimum at $\omega\hbar = \Delta + \Delta_V$. $\alpha'(\omega)$ increases in the frequency range from zero to $\Delta/\hbar$. In the frequency range from $\omega = \Delta/\hbar$ to $\omega = (\Delta + \Delta_V)/\hbar$ there is a region of anomalous dispersion, and when $\hbar\omega \rightarrow \infty$ we get $\alpha'(\omega) \rightarrow 0$ (see Fig. 2).

In the presence of a field, the course of the dispersion curve changes. The position of the minimum is determined as before by the width of the valence band, but the function $\alpha'(\omega)$ reaches a maximum value at the frequency $\omega\hbar = \Delta + \Delta_E$ at which the absorption coefficient vanishes. The magnitude of the maximum is determined by the parameters Δ_E and $\hbar/\tau_2$. In a strong field (i.e., when $\Delta_E \gg \hbar/\tau_2$), we have

$$\alpha'(\omega\hbar = \Delta + \Delta_E) \approx \frac{e^2(m^*)^{3/2}}{\pi^2\hbar^3}\left(\sqrt{\Delta_v} + \frac{8}{3}\frac{\Delta_E^{5/2}}{\hbar^2/\tau_2^2}\right).$$

Depending on the value of Δ_E, a change takes place also in the frequency at which $\alpha'(\omega) = 0$. Allowance for the finite nature of τ_2, both without a field and in the presence of the field $\mathbf{E}_1$, eliminates the discontinuities at the maximum and minimum points of the dispersion curve.

We must note here the following. The expression for the conductivity of the system contains the matrix element of the velocity (or the momentum). It

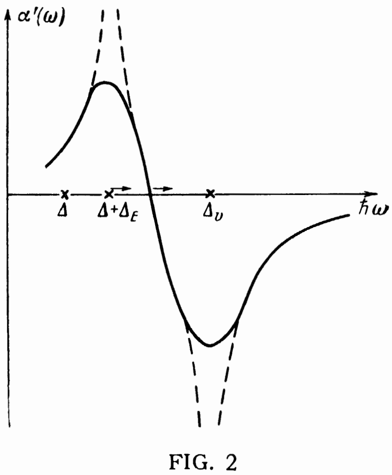

FIG. 2

is connected with the matrix element of the displacement by the relation $r_{pp'} = iv_{pp'}/\omega_{pp'}$. When using expression (1.2) for the polarization vector, we have assumed in fact that $r_{pp'} = iv_{pp'}/\omega$. Such a substitution is perfectly justified in the calculation of the absorption, owing to the resonant character of this process. In calculating the dispersion, allowance for this difference can ensure convergence of the integral in expression (3.1) also in the case of an infinitely large band. This remark pertains also to the case of two-photon processes, which are considered in Sec. 4. The results of the corresponding calculations were given in the paper by Keldysh[13].

4. POLARIZATION OF SEMICONDUCTORS IN TWO-PHOTON PROCESSES

If the lifetimes of the electrons in the bands (the recombination times) are shorter than the deceleration times, then we can calculate the polarization by using, as in[9], the following system of equations for the density matrix $\rho_{nm}(\mathbf{p}, \mathbf{p}', t)$:

$$\frac{\partial\rho_{nm}(\mathbf{p},\mathbf{p}',t)}{\partial t} = \frac{i}{\hbar}(\mathscr{E}_m(\mathbf{p}') - \mathscr{E}_n(\mathbf{p}))\rho_{nm} - \frac{1}{\tau_{nm}}(\rho_{nm} - \overset{0}{\rho}_{nm})$$
$$+ \frac{ie}{\hbar}\sum_k \int \left(\overset{(nk)}{r}_{\mathbf{p},\mathbf{p}''}\rho_{km}(\mathbf{p}'',\mathbf{p}',t) - \rho_{nk}(\mathbf{p},\mathbf{p}'',t)\overset{(km)}{r}_{\mathbf{p}''\mathbf{p}}\right)d\mathbf{p}''. \qquad (4.1)$$

Here n, m, and k are the band indices and **p**, **p′**, and **p″** are the corresponding momenta.

The polarization is calculated under the following assumptions.

1. Only direct interband transitions are taken into account. Under this condition, as in[9], we have

$$\overset{(nn)}{r}_{\mathbf{p}\mathbf{p}'} = 0; \quad \overset{(nm)}{r}_{\mathbf{p}\mathbf{p}'} = c\frac{\mathbf{p}}{|\mathbf{p}|}\delta(\mathbf{p}-\mathbf{p}') \equiv r_{\mathbf{p}}\delta(\mathbf{p}-\mathbf{p}'),\ n \neq m. \qquad (4.2)$$

2. We consider the three-band approximation (one valence band and two conduction bands). We designate the valence band by the index 1, and that of the conduction band of the indices 2 and 3. The energies of the forbidden bands are denoted Δ_{12} and Δ_{23}; $\Delta_{23} \neq \Delta_{12}$.

3. The field frequency ω_1 is close to $\Delta_{12}/2\hbar$, i.e.,

$$\mathbf{E} = {}^1\!/_2(\mathbf{E}e^{i\omega t} + \text{c.c.}), \qquad \hbar\omega \approx \Delta_{12}/2. \qquad (4.3)$$

4. We represent the density matrix in the form

$$\rho_{nm}(\mathbf{p},\mathbf{p}',t) = \overset{(0)}{\rho}_{nm}(\mathbf{p},\mathbf{p}',t) + \overset{(1)}{\rho}_{nm}(\mathbf{p},\mathbf{p}',t) + \overset{(2)}{\rho}_{nm}(\mathbf{p},\mathbf{p}',t). \qquad (4.4)$$

Here $\rho^{(0)}_{nm}$ is the dc component and $\rho^{(1)}_{nm}$ and $\rho^{(2)}_{nm}$ are periodic functions with frequencies ω_1 and $2\omega_1$, i.e.,

$$\overset{(1)}{\rho}_{nm}(\mathbf{p},\mathbf{p}',t) = \overset{(1)}{\rho}_{nm}e^{i\omega t} + \text{к.с.}, \quad \overset{(2)}{\rho}_{nm}(\mathbf{p},\mathbf{p}',t) = \overset{(2)}{\rho}_{nm}e^{i2\omega t} + \text{c.c.} \qquad (4.5)$$

5. The lifetime in the lower conduction band (band 2) is much larger than in the upper one. Under this condition, the population of the third band is small compared with the population of the second band, and it can be disregarded in the expression for the polarization vector.

6. We take into account only resonant two-photon processes.

7. The semiconductor is assumed to be isotropic.

Under these assumptions we obtain the following expression for the polarization vector:

$$\mathbf{P}(t) = -\frac{e^4 n}{4}\mathbf{E}^3\int \frac{|r_{13}r_{23}|^2 D(\mathbf{p})}{(\mathscr{E}_1(\mathbf{p}) - \mathscr{E}_3(\mathbf{p}) - \hbar\omega)^2}\cdot$$
$$\cdot\frac{(\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - 2\hbar\omega)\cos\omega t + (\hbar/\tau_2)\sin\omega t}{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - 2\hbar\omega]^2 + \hbar^2/\tau_2^2}d\mathbf{p}. \qquad (4.6)$$

The population difference D(p) is given by the expression

$$D(\mathbf{p}) = \frac{D^0(\mathbf{p})}{1 + R_2^2/\{[\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathbf{p}) - 2\hbar\omega]^2 + \hbar^2/\tau_2^2\}},$$
$$R_2^2 = \frac{e^4|r_{13}r_{23}|^2E^4}{4(\mathscr{E}_1(\mathrm{p}) - \mathscr{E}_3(\mathbf{p}) - \hbar\omega)^2}\frac{\tau_1}{\tau_2}. \qquad (4.7)$$

We note that we have taken into account here only the resonant response of the system to the external field. The quantities τ_1 and τ_2 have the same meaning as before.

From (4.6) and (4.7) we obtain the following expression for the imaginary part of the polarizability due to the interband transitions:

$$\alpha''(\omega) = -\frac{e^4 n E^2}{8}\int \frac{|r_{13}r_{23}|^2 D^0(\mathbf{p})}{[\mathscr{E}_1(\mathbf{p}) - \mathscr{E}_3(\mathbf{p}) - \hbar\omega]^2}\cdot$$
$$\cdot\frac{\hbar/\tau_2}{(\mathscr{E}_2(\mathbf{p}) - \mathscr{E}_1(\mathrm{p}) - 2\hbar\omega)^2 + \hbar^2/\tau_2^2 + R_2^2}d\mathbf{p}. \qquad (4.8)$$

In the case of infinitely narrow bands (three discrete levels), expressions (4.7) and (4.8) are similar to those obtained in the paper by Khronopulo[14].

In order to derive from this the expressions for the absorption coefficients obtained in[3,5], we must neglect saturation, i.e., we must put **E** = 0 ($R_2 = 0$), and then let $\hbar/\tau_2 \to 0$.

Comparing (4.6) with expression (1.15), which describes the polarization of the single-photon and two-photon processes, we can readily see that within the framework of the given model the expressions have the same structure. The saturation parameter in the two-photon processes is an expression proportional to E_1^4 and containing the nonresonant factor $(\mathscr{E}_1(\mathrm{p}) - \mathscr{E}_3(\mathrm{p}) - \hbar\omega)^{-2}$ both in the saturation parameter and in front of the entire expression for the absorption coefficient. It is easy to see, however, that if saturation is neglected ($R_2 = 0$ and $\hbar/\tau_2 \to 0$) then the value of $\mathscr{E}_1(p) - \mathscr{E}_3(p) - \hbar\omega$ is determined from the maximum of the resonance curve. In the integration we shall use the fact that the function $\mathscr{E}_1(p) - \mathscr{E}_3(p) - \hbar\omega$ varies little over the width of the resonance curve also when $R_2 \neq 0$. As a result, it can be taken outside the integral sign and $\mathscr{E}$ replaced by $\mathscr{E}_{max}$. In this approximation, the integration of Eqs. (4.8) and (1.9) is perfectly analogous.

So far we have considered only allowed transition. This means that the matrix element does not depend on the quasimomentum. On the other hand, in the case when the transition from band to band is forbidden, the matrix element is proportional to $\mathbf{p}$. In the case of two-photon processes we deal with two matrix elements $\mathbf{r}_{13}$ and $\mathbf{r}_{23}$, describing transitions from band 1 to band 3 and from band 2 to band 3. In the integration it is necessary to allow for three possibilities: 1) both transitions are allowed, 2) both are forbidden by the selection rules, 3) one is allowed and the other is forbidden.

In this paper it was assumed that all the transitions are allowed. It is easy to generalize to the case of forbidden transitions.

Thus, all the arguments with respect to the absorption coefficient and the dispersion curve, given in Secs. 2 and 3 of the present paper for single-photon processes, can be applied to two-photon processes. The saturation effect denotes as before that at a certain value of the external field $E_1 = E_S$ the coefficient of two-photon absorption becomes equal to zero.

However, whereas in the case of single-photon processes the strong-field criterion was the parameter $(\hbar/\tau_2)^2/(erE/3)^2$, for two-photon processes the role of this criterion is played by the expression

$$\left(\frac{\hbar}{\tau_2}\right)^2 \Big/ \frac{(eE)^4 |r_{13} r_{23}|^2}{\Delta_{13}^2}$$

Just as in Sections 1—3, it is impossible to confine oneself to the single-particle approximation when deceleration is taken into account.

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[1] N. G. Basov, A. Z. Grasyuk, and V. A. Katulin, DAN SSSR **161**, 1306 (1965), Soviet Phys. Doklady **10**, 343 (1965).

[2] N. G. Basov, A. Z. Grasyuk, I. G. Zubarev, and V. A. Katulin, JETP Letters **1**, No. 4, 29 (1965), transl. **1**, 118 (1965).

[3] R. Brounstein and N. Ockman, Phys. Rev. **134A**, 499 (1964).

[4] R. London, Proc. Phys. Soc. **80**, 952 (1963).

[5] N. G. Basov, A. Z. Grasyuk, I. G. Zubarev, V. A. Katulin, and O. N. Krokhin, JETP **50**, 551 (1966), Soviet Phys. JETP **23**, 366 (1966).

[6] O. N. Krokhin, FTT **7**, 2612 (1965), Soviet Phys. Solid State **7**, 2114 (1966).

[7] N. G. Basov and O. N. Krokhin, JETP **46**, 1508 (1964), Soviet Phys. JETP **19**, 1022 (1964).

[8] N. G. Basov and O. N. Krokhin, FTT **5**, 2384 (1963), Soviet Phys. Solid State **5**, 1737 (1964).

[9] Yu. L. Klimontovich and E. V. Pogorelova, JETP **50**, 605 (1966), Soviet Phys. JETP **23**, 402 (1966).

[10] Yu. M. Popov, Quantum Radiophysics, Trudy, Phys. Inst. Acad. Sci. **31**, 3 (1965).

[11] S. V. Vavilov, Deistvie izlucheniĭ na poluprovodniki (Effect of Radiation on Semiconductors), Fizmatgiz, 1963.

[12] R. Smith, Semiconductors (Russ. transl.) IIL, 1962.

[13] L. V. Keldysh, Nonlinear Polarization of Semiconductors, Paper delivered at Second All-union Symposium on Nonlinear Optics, Novosibirsk, June 1966.

[14] Yu. G. Khronopulo, Izv. Vuzov, Radiofizika **7**, 674 (1964).

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