Resonant excitation of carriers in a semiconductor by a high-power light pulse

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An analysis is made of the photoexcitation of carriers in a semiconductor by intense light whose frequency is close to the band gap. It is assumed that the most characteristic relaxation frequency is the frequency of electron–electron collisions. An analysis is made of the evolution of the distribution of nonequilibrium carriers under the influence of a high-power nanosecond laser pulse and the results obtained are compared with the experimental data. The influence of intraband energy relaxation, recombination, and nonmonochromaticity of laser radiation on the evolution of the distribution functions is determined. The results obtained are applicable to the case of interband excitation as well as to the case of photoexcitation from an impurity level.

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Photoexcitation of carriers in a semiconductor by a high-power laser pulse generates a nonequilibrium electron – hole plasma. The nature of the distribution functions of these photocarriers is governed by the properties of the semiconductor: the frequencies of energy relaxation by interaction with acoustic (ν_A) and optical (ν_O) phonons and by collisions of carriers with one another (ν_{ee}), and the recombination frequency ν_R. Light interacting with a semi-conductor alters the carrier density and the inequalities between these frequencies. An electromagnetic wave of sufficiently large amplitude influences also the dynamics of quasiparticles, which is important if the frequency λ of the field-induced transitions is much higher than the relaxation frequencies.[1] All this makes the dependences of the characteristics of an electron – hole plasma on the parameters of an exciting pulse very complex. Elesin[2] solved the transient problem of the distribution of carriers in the field of an electromagnetic wave of any (no matter how large) amplitude by ignoring the frequency ν_{ee}. Levinson and Levinskiĭ[3] found the steady-state distribution function of electrons on condition that $\nu_O, \nu_{ee} \gg \nu_A$.

If the field of a light wave creates carriers of energy insufficient to emit an optical phonon, then $\nu_O = 0$ and $\nu_A \lesssim 10^9$ sec^{-1} (Ref. 3). Under the conditions, beginning from relatively low plasma densities n_c, the process of energy transfer is controlled by collisions between carriers ($n_c \sim 3 \times 10^{14}$ cm^{-3} for GaAs—Ref. 4). Moreover, if the frequency λ satisfies the condition $\lambda \ll \nu_{ee}$, the distribution functions can be regarded as the Fermi functions with nonequilibrium values of the temperature T and chemical potentials of electrons (μ_e) and holes (μ_h). The time dependences of these parameters are found from the equations of balance of the number of particles n and their energy $\langle n\varepsilon \rangle$. The steady-state balance equations were investigated by Krokhin.[5]

We shall consider resonant excitation of carriers by light of frequency ω close to the band gap Δ. We shall assume that $\nu_{ee} \gg \lambda \gg \nu_A, \nu_R$. We shall analyze the evolution of the carrier distribution for times $t \ll \nu_A^{-1}, \nu_R^{-1}$. The influence of relaxation can be ignored for these time intervals t; only the interaction of carriers with the electromagnetic wave field and with one another remain important. If the duration τ of a laser pulse is of the order of 10^{-9} sec, the condition for the adiabatic interaction between the wave field and carriers is satisfied right up to the end of the pulse. The temperature T acquired by electrons and holes at this moment is governed solely by the mass ratio $k = m_e/m_h$ and by the difference $\omega - \Delta$. The values of T found for GaAs in the case when $\omega - \Delta < 15$ meV are in good agreement with the experimental results.[6] We shall analyze qualitatively the phase trajectories (paths) in the (μ_e, T) plane and find the steady-state distributions of carriers in the $\tau \gg \nu_A^{-1}, \nu_R^{-1}$ case. We shall determine the influence of nonmonochromaticity of the laser light on the evolution of the distributions. Our results will apply not only to the case of interband photoexcitation but also to the excitation of electrons from impurity levels.

We shall use a system of units in which $\hbar = 1$.

BALANCE EQUATIONS

We shall consider a two-band semiconductor with effective electron and hole masses m_e and m_h. The kinetic equations for the electron and hole distribution functions [$f_e(\mathbf{p})$ and $f_h(\mathbf{p})$] can be written as follows

$$\begin{aligned} \dot{f}_e(\mathbf{p}) &= S_{eh}\{f_e, f_h\} + S_{ef}\{f_e\} + S_R\{f_e\} \\ &+ 2\pi\lambda_p^2(1 - f_e(\mathbf{p}) - f_h(\mathbf{p}))\,\delta(\varepsilon_e(p) + \varepsilon_h(p) - 2\varepsilon_0), \\ \dot{f}_h(\mathbf{p}) &= S_{he}\{f_h, f_e\} + S_{hf}\{f_h\} + S_R\{f_h\} \\ &+ 2\pi\lambda_p^2(1 - f_e(\mathbf{p}) - f_h(\mathbf{p}))\,\delta(\varepsilon_e(p) + \varepsilon_h(p) - 2\varepsilon_0). \end{aligned} \tag{1}$$

Here, $\varepsilon_e(p) = p^2/2m_e$ and $\varepsilon_h(p) = p^2/2m_h$ are the dispersion laws of the quasiparticles; $\lambda_p = |\mathbf{E}\mathbf{d}_p|$; $\mathbf{E}$ is the electric field in the incident wave; $\mathbf{d}_p$ is the interband matrix element of the dipole moment operator; $2\varepsilon_0 = \omega - \Delta$. Collisions of carriers at a characteristic frequency ν_{ee}, which conserve the number of electrons and holes, are described by the first terms in the system (1). The second terms correspond to the interaction of the quasiparticles with phonons and the terms $S_R\{f_{eh}\}$ describe recombination. The last terms allow for the interaction with a high-frequency field in accordance with perturbation theory. After a time $t \gg \nu_{ee}^{-1}$, the following Fermi distributions are established:

$$f_{eh}(p) = \left[\exp\left(\frac{\varepsilon_{eh}(p) - \mu_{eh}}{T}\right) + 1\right]^{-1}, \tag{2}$$

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whose parameters μ_e, μ_h, and T are governed by the balance equations.[5] These equations follow from Eqs. (1) and (2) and are

$$\dot{n}_e = J(1-f_e(p_0)-f_h(p_0))-R, \tag{3}$$

$$\langle (n\varepsilon)^{\cdot}\rangle = 2\varepsilon_0 J(1-f_e(p_0)-f_h(p_0))-P. \tag{4}$$

Here, n_e is the electron density, $\langle n\varepsilon\rangle$ is the carrier energy,

$$n_e = \frac{1}{(2\pi)^3}\int dp\, f_e(p),$$

$$\langle n\varepsilon\rangle = \frac{1}{(2\pi)^3}\int dp\{\varepsilon_e(p)f_e(p)+\varepsilon_h(p)f_h(p)\},$$

R is the density of electrons recombining per unit time:

$$R = \frac{1}{(2\pi)^3}\int dp\, S_R\{f_e\},$$

P is the energy transferred per unit time to the lattice:

$$P = \frac{1}{(2\pi)^3}\int dp[\varepsilon_e(p)S_{ef}\{f_e\}+\varepsilon_h(p)S_{hf}\{f_h\}],$$

$$p_0 = 2(m\varepsilon_0)^{1/2},\quad J = 2m^{3/2}\bar{\lambda}_p^2\varepsilon_0^{1/2}/\pi,\quad m = m_e m_h/(m_e+m_h),$$

the bar above λ_p^2 denotes averaging over the angle between E and d_p, and m is the reduced mass. (It is assumed that cascade emission of acoustic phonons plays the dominant role in the recombination process; carriers are then captured from the bottoms of the energy bands and the energy balance equation does not contain the recombination terms.) Equations (3) and (4), supplemented by the electrical neutrality condition

$$n_e(\mu_e, T) = n_h(\mu_h, T), \tag{5}$$

represent a complete system for the determination of $\mu_e(t)$, $\mu_h(t)$, and $T(t)$.

If

$$f_e(p_0)+f_h(p_0) = 1, \tag{6}$$

the field ceases to generate electron – hole pairs. This is due to equalization of the electron occupation numbers of the resonant energy levels linked by optical transitions in a strong field. Equations (5) and (6) are specified on the (μ_e, T) plane[1)] by a line and the points on this line will be called the saturation states.

INTEGRAL CURVES WITHOUT ALLOWANCE FOR RELAXATION

We shall now consider the case of a strong field when

$$J \gg R,\quad P/\varepsilon_0. \tag{7}$$

The condition (7) allows us to drop always, with the exception of a small region in the vicinity of the line of saturation states, the relaxation terms from Eqs. (3) and (4). The relationship between μ_e and T is then given by the first integral

$$\langle n\varepsilon\rangle - 2\varepsilon_0 n = \text{const} \tag{8}$$

and by the condition (5). The behavior of the integral curves is easiest to analyze in the $m_e = m_h$ case. Then, the saturation state is reached on the straight line $\mu_e = \varepsilon_0$, and Eq. (8) can be transformed to

$$\frac{3}{2}x^{3/2}F_{3/2}\left(\frac{y}{x}\right) - x^{3/2}F_{1/2}\left(\frac{y}{x}\right) = C. \tag{9}$$

Here, $F_{3/2}(\eta)$ and $F_{1/2}(\eta)$ are the Fermi – Dirac integrals[7]; $x = T/\varepsilon_0$ is the dimensionless temperature; y

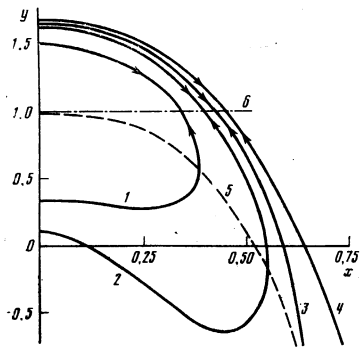

FIG. 1. Intergal curves $y_c(x)$ for different values of C: 1) $C = -0.109$; 2) $C = -0.0162$; 3) $C = 0$; 4) $C = 0.02$. The derivative dx/dy changes its sign on curve 5; the steady-state values of x and y are attained on the line 6.

$= \mu_e/\varepsilon_0$ is the dimensionless chemical potential; the constant C is governed by the initial conditions (the density n_0 and the temperature T_0). Substituting $x = 0$ in Eq. (9), we obtain

$$\frac{\sqrt{\pi}}{3}y^{3/2}\left(\frac{3}{5}y-1\right) = C. \tag{10}$$

If $C > 0$, Eq. (10) has just one solution $y_0 > 5/3$; if $-2\sqrt{\pi}/15 < C \leq 0$, there are two solutions: $0 \leq y_1 < 1$ and $1 < y_2 \leq 5/3$; if $C = -2\sqrt{\pi}/15$, curve (9) contracts to the point $x = 0$, $y = 1$. The integral curve (9) with $C = 0$ consists of two regions: the singular line $x = 0$, $y \leq 0$ and the line $y_0(x)$ which has the asymptote $x = 2/3$, $y \to -\infty$. This line describes a separatrix that divides integral curves $y_c(x)$ of two types: if $C < 0$, both ends of the curve correspond to the temperature $x = 0$ (Fig. 1); if $C > 0$, one end of the integral curve also corresponds to zero temperature, but in the limit $x \to \infty$, the curve $y_c(x)$ approaches asymptotically the line

$$y = x\ln\frac{C}{x^{3/2}(3/2x-1)}.$$

If the thermal energy of equilibrium electrons is greater than ε_0 (i.e., if $T_0 > 2/3\varepsilon_0$), then $C > 0$ and on application of the field the chemical potential μ_e increases, whereas the temperature decreases. If at a moment t the distribution is nondegenerate, then

$$T(t) = {}^2/_3\varepsilon_0 + (T_0 - {}^2/_3\varepsilon_0)/(1+Jt/n_0).$$

Cooling occurs because the energy of electrons transferred to the condition band is less than the average thermal energy, and when electrons become thermalized, the system as a whole experiences cooling. When $T_0 < 2/3\varepsilon_0$, the constant is $C < 0$ and the electron gas becomes heated again, but on increase in the chemical potential μ_e this effect changes to cooling. If $T_0 = 0$, the initial density $n_0 = 0$ and the electron temperature are governed entirely by photocarriers. In this case for a nondegenerate distribution we have $T = 2/3\varepsilon_0$ and the evolution of the system is described by the separatrix $(C = 0)$.

If $m_e \neq m_h$, Eqs. (5) and (8) can be written in the form

$$k^{3/2}F_{1/2}(\eta_e) = F_{1/2}(\eta_h), \tag{11}$$

$${}^3/_2 x^{3/2}[F_{3/2}(\eta_e)+k^{-3/2}F_{3/2}(\eta_h)]-2x^{3/2}F_{1/2}(\eta_e) = C, \tag{12}$$

where $k = m_e/m_h$ is the mass ratio; $\eta_e = \mu_e/T$, $\eta_h = \mu_h/T$,

$x = T/\varepsilon_0$. The integral curves given by the system (11)-(12) are qualitatively no different from the curves plotted for a semiconductor with $m_e = m_h$ (Fig. 1). If $k \ll 1$, Eq. (12) can be simplified in two limiting cases. If $\eta_e \gg 1$, then the ratio is

$$r = F_{3/2}(\eta_h)/k^{3/2}F_{3/2}(\eta_e) \ll 1$$

($r = k$ in the region defined by $\eta_h \gg 1$ and $r \propto \eta_e^{-1}$ if ∞). In the case of nondegenerate distributions ($|\eta_e| \gg 1$, $\eta_h \to -\eta_e < 0$), we have $r = 1$. In both cases, Eq. (12) reduces to an equation of the (9) type. Transforming Eq. (6), we obtain[5]

$$x(\eta_e + \eta_h) = 2. \tag{13}$$

It follows from Eqs. (11) and (13) that the line of saturation states intersects the μ_e axis at the point $\mu = 2\varepsilon_0(1 + m_e/m_h)^{-1}$, and in the limit $x \to \infty$ this line has an asymptote whose slope is governed by the equation

$$k^{3/2}F_{1/2}(\eta_e) = F_{1/2}(-\eta_e).$$

In the excitation of an electron from an impurity level separated by a gap E_i from the bottom of the conduction band the quasimomentum is not conserved and the saturation condition should be written as follows:

$$f_e(E_0) = f_i. \tag{14}$$

Here, f_i and $f_e(E_0)$ are the occupation numbers of electrons at the impurity level and in the band for $\varepsilon_e(p) = E_0$, $E_0 = \omega - E_i$. If we ignore the relaxation processes, the integral curves are given by Eq. (9) (where ε_0 is replaced with E_0). We shall replace the impurity concentration n_i by the chemical potential μ_0:

$$n_i = (2m_e\mu_0)^{3/2}/3\pi^2.$$

If $\mu_0 \gg E_0$, then Eq. (14) and the electrical neutrality condition in the range $\mu_e \ll \mu_0$ readily yield the explicit dependence $T(\mu_e)$:

$$T = (\mu_e - E_0)/\ln\{(\mu_e/\mu_0)^{3/2} - 1\},$$

which determines the line of saturation states.

Let us assume that T_L and μ_L are the coordinates of the point of intersection L between an integral curve a and a line of saturation states b (Fig. 2). In the case of short pulses of duration

$$\tau \ll \tau_1 = \pi n_e(\mu_L, T_L)/2m^{3/2}\lambda_p^2\varepsilon_0^{1/2} \tag{15}$$

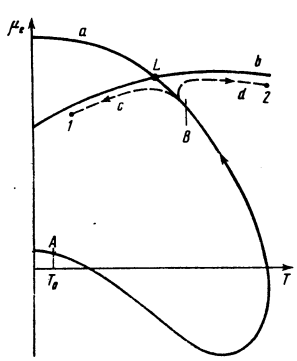

FIG. 2. Schematic form of the trajectory in the case when $\tau \gg \nu_R^{-1}$, ν_A^{-1}: a) integral curve in the adiabatic approximation; b) line of saturation states. The adiabatic (AB) part of the trajectory corresponds to times $t \sim \tau_1$. The times $t \sim \nu_R^{-1}$, ν_A^{-1} correspond to the part of the trajectory c if $\nu_R \ll \nu_A$ and to the part d if $\nu_R \gg \nu_A$. Here, T_0 is the lattice temperature. 1), 2) Steady-state solutions for various relationships between ν_R and ν_A.

the saturation state is not achieved and at the end of the pulse the quantities μ_e and T satisfy the equation $n_e(\mu_e, T) = J\tau$. In the case of intermediate pulse duration τ, we have

$$\nu_R^{-1},\ \nu_A^{-1} \gg \tau \gg \tau_1, \tag{16}$$

where the values of $\mu_e(\tau)$ and $T(\tau)$ lie in the direct vicinity of the point L.

If a laser pulse causes interband transitions, the values of T, μ_e, and μ_h at the point L are given by Eqs. (11)-(13). [It should be noted that when a pulse of $S = 10^6$ W/cm^2 intensity and $\tau = 10^{-10}$ sec duration is incident on GaAs, the inequality (16) is obeyed for all those values of ε_0 that have a lower optical phonon frequency.] At a sufficiently low temperature T_0 the density of photocarriers obeys $n \gg n_0$ (for example, according to Ref. 6, the ratio is $n/n_0 = 10^2$) and we can substitute $C = 0$ in Eq. (12). A numerical solution of the system (11)-(13) for $k = 0.1$ and $C = 0$ gives

$$T = 0.315\varepsilon_0,\quad \mu_e = 2.205\varepsilon_0,\quad \mu_h = -0.205\varepsilon_0. \tag{17}$$

The result (17) allows us to interpret the experimental data[6] for frequencies $\omega < \Delta + 15$ meV without invoking any relaxation mechanism. In the case of GaAs, we have $k = 0.086$-0.14 and a numerical coefficient in the dependence $T = 0.306\varepsilon_0$, deduced from the data of Ref. 6, differs by just 3% from the theoretical value (17). We recall that in our calculations we used just one characteristic of the material, which was the mass ratio m_e/m_h.

In the case of a long pulse, when

$$\tau \gg \nu_A^{-1},\ \nu_R^{-1}, \tag{18}$$

the nature of the solution of the system (3)-(4) depends strongly on the relaxation processes. In the next section we shall assume that the condition (18) is satisfied.

INFLUENCE OF RELAXATION AND OF NONMONOCHROMATICITY OF LASER RADIATION ON THE EVOLUTION OF CARRIER DISTRIBUTIONS

When the inequalities (7) are obeyed, the relaxation processes result in perturbation of the "adiabatic" trajectory of a system except in the direct region of the line of saturation states. If the correction to the unperturbed solutions is calculated, it is easy to show that the width of this region is

$$\Delta\mu_e \sim T\nu n_e(\mu_e, T)/J$$

(ν is the higher of the frequencies ν_A and ν_R). We shall assume that the lattice temperature is $T_0 \ll T_L$. Then, if $\nu_R \ll \nu_A$, the derivative at the point L is $d\{\langle n\varepsilon\rangle - 2\varepsilon_0 n\}/dt < 0$ and the distribution after the adiabatic stage evolves toward lower temperature (curve c in Fig. 2). However, if $\nu_R \gg \nu_A$ and, consequently, $R > P$ at the point L, then the adiabatic stage is followed by the heating of carriers (curve d).

The scattering of phonons fixes not only the temperature of carriers but also their chemical potential. Therefore, the Fermi functions with $T = T_0$ can satisfy simultaneously Eqs. (5) and (6) and cause P to vanish. If $\nu_R = 0$, these functions give the exact solution of the system (3)-(4). Recombination results in a finite lifetime of electrons of energies $\varepsilon < \varepsilon_0$ and makes it possible

to create, by a strong field, pairs of carriers of energy $2\varepsilon_0$. Thus, in each recombination event the system of electrons and holes receives an energy $\varepsilon_0 - \varepsilon > 0$ and its steady-state temperature becomes $T_1 > T_0$.

The influence of light nonmonochromaticity is similar to the effect of recombination. The high-frequency wing of a radiation pulse ($\varepsilon_1 > \varepsilon_0$) creates an electron-hole pair and the low-frequency wing ($\varepsilon_2 < \varepsilon_0$) causes stimulated recombination of this pair. The energy $\varepsilon_1 - \varepsilon_2 > 0$ is then transferred to the gas of carriers causing its heating.

We shall illustrate this by considering a semiconductor with equal electron and hole masses. Let us assume that the profile of the laser radiation line is given by the function $\rho(\varepsilon)$, where $2\varepsilon = \omega - \Delta$, which has a sharp maximum at $\varepsilon = \varepsilon_0$:

$$\int d\varepsilon\, \rho(\varepsilon) = 1; \quad \int d\varepsilon\, \rho(\varepsilon)\varepsilon = \varepsilon_0; \quad \int d\varepsilon\, (\varepsilon - \varepsilon_0)^2 \rho(\varepsilon) = (\Delta\varepsilon)^2 \ll \varepsilon_0^2,$$

and the temperature T_L satisfies the condition

$$\varepsilon_0 \gg T_L \gg (ms^2\varepsilon_0)^{1/3} \tag{19}$$

(s is the velocity of sound). In the case of the polarization scattering mechanism,[8] the energy transferred to the lattice is

$$P(\mu, T) = 2A\mu(T - T_0).$$

(The coefficient A depends on the electron – phonon interaction constant.) If the inequality (19) is obeyed, the number of carriers captured by unit time by the attractive centers tends in the limit of $T \to 0$ to a value $R(\mu, T) = B$, which is independent of μ. If we assume that $(\varepsilon_0 - \mu)/2T \ll 1$, we obtain the system of equations

$$\begin{aligned} \dot{n} &= J(\varepsilon_0 - \mu)/2T - B, \\ \langle (n\varepsilon)^{\cdot} \rangle &= J\varepsilon_0(\varepsilon_0 - \mu)/2T + J(\Delta\varepsilon)^2/T - 2A\mu(T - T_0). \end{aligned} \tag{20}$$

The system (20) differs from the system (3)-(4) by allowance for the dispersion of light $\Delta\varepsilon$, whose influence is significant if $\varepsilon_0 - \mu_0 \lesssim (\Delta\varepsilon)^2/\varepsilon_0$. If we assume that $J \gg B$, we obtain the following steady-state values of T and μ:

$$\begin{aligned} T_1 &= \frac{1}{2}\left(T_0 + \frac{B}{A}\right) + \left[\frac{1}{4}\left(T_0 + \frac{B}{A}\right)^2 + \frac{J(\Delta\varepsilon)^2}{2A\varepsilon_0}\right]^{1/2}, \\ \mu_1 &= \varepsilon_0 - 2T_1 B/J. \end{aligned} \tag{21}$$

The expressions in Eq. (21) are in agreement with the above considerations.

If

$$(\Delta\varepsilon)^2 \sim T_L \varepsilon_0 \nu_R n_e(\mu_L, T_L)/J,$$

the nonmonochromaticity of light and the recombination processes make comparable contributions to the heating of carriers. For example, if we assume that the intensity of a pulse incident on GaAs is $S = 10^6$ W/cm^2 and that $\varepsilon_0 = 10$ meV, $T_L = 10$°K, $\nu_R = 10^9$ sec^{-1}, we obtain $\Delta\varepsilon \sim 1.5 \times 10^{11}$ sec^{-1}. It should be noted that the width of the radiation spectrum emitted by a crystal laser[9] can be greater than or smaller than the above value of $\Delta\varepsilon$.

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[1)] The possibility of investigating the system (3)-(4) with the aid of the (μ_e, T) phase plane was pointed out to the author by I. B. Levinson.

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