Picosecond superradiance in GaAs during interband absorption of intense short light pulses

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(Submitted 26 April 1989; resubmitted 23 October 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 11, 462–465 (10 December 1989)

It is shown theoretically that a recombination superradiance can develop and self-quench over picosecond time scales during interband absorption of intense light pulses in a semiconductor. The self-quenching results from a heating of nonequilibrium carriers and is accompanied by an increase in their density.

During the production of an electron-hole plasma in a direct-gap semiconductor by intense picosecond light pulses, collisions of nonequilibrium carriers with each other and with optical phonons are the primary relaxation processes. If the photon energy $\hbar\Omega$ of the exciting light is only slightly greater than the gap width E_g, these processes can lead to a quasiequilibrium state in the electron-hole plasma even at the leading edge of the pulse.[1-3] This quasiequilibrium state is characterized by a population inversion of the nonequilibrium carriers: $\mu_e - \mu_h > E_g$, where $\mu_e(T)$ and $\mu_h(T)$ are the electron and hole Fermi quasilevels, and T is the plasma temperature.

It is usually assumed that the recombination of electrons and holes does not affect the evolution of the electron-hole plasma during the excitation, since the recombination times are much longer than the pulse length. In the case of an inversion, however, weak recombination emission may be amplified, and it may in turn accelerate the recombination. Furthermore, under the condition $\alpha_m D > 1$ (D is the diameter of the spot of exciting light focused onto the semiconductor, and α_m is the maximum gain in the amplification of the light), the stage is set for the occurrence of recombination superradiance in the plane of the semiconductor layer. In GaAs, for example, in which α_m can have values $\sim 10^3$ cm^{-1}, and the time scale of the amplification is ~ 0.1 ps, we might expect the development of a recombination superradiance at picosecond time scales for $D \gg 10$ μm.

Our purpose here is to show theoretically that a recombination superradiance can occur in GaAs even at picosecond time scales and that this superradiance could, along with the relaxation processes cited above, have a strong effect on the evolution of the electron-hole plasma during the intense excitation pulse. A stimulated picosecond emission has been observed in experiments[4–6] on the excitation of thin layers of semiconductors by intense light pulses.

We will analyze the processes which play out over picosecond time scales in the approximation of a quasiequilibrium distribution function of the nonequilibrium carriers. We assume that the lattice temperature of the semiconductor is low and that the condition $\hbar\Omega - E_g < \hbar\Omega_{ph}$ holds, in which case optical phonons can be ignored (Ω_{ph} is the frequency of an optical phonon). We use a parabolic model of the band structure. The balance equations for the number of particles and for the energy of the electron-hole plasma are

$$\frac{dn}{dt} = \frac{\alpha(\Omega)I}{\hbar\Omega} + c\int \alpha(\omega)\, N_\omega\, \rho(\omega)\,d\omega, \qquad n = n_e = n_h, \tag{1}$$

$$\frac{dW}{dt} = \left(\hbar\Omega - E_g - n\frac{\partial E_g}{\partial n}\right)\frac{\alpha(\Omega)I}{\hbar\Omega} + c\int \alpha(\omega)N_\omega \rho(\omega)(\hbar\omega - E_g)\,d\omega, \tag{2}$$

$$\alpha(\omega) = \alpha_0\sqrt{\hbar\omega - E_g}\,(1 - f_e - f_h),$$

where $f_{e,h}$ and $n_{e,h}$ are the Fermi functions and densities of the electrons and holes under the assumption that the latter have the same temperature[2]; W is the kinetic energy density of the plasma; $E_g = E_g^0 - \gamma n^{1/3}$, where E_g^0 is the width of the band gap of the unexcited semiconductor, and γ is the coefficient of the renormalization of the gap width due to the Coulomb interaction between carriers; and $I(t)$ is the intensity of the pump pulse. Equations (1) and (2) have been written for the case $\alpha(\Omega)d \ll 1$, where d is the thickness of the semiconductor layer, N_ω is the number of photons ($E_g \leqslant \hbar\omega \leqslant \hbar\Omega$) which are propagating along the semiconductor layer in a solid angle $\beta \approx d/D$, $\rho(\omega) = \omega^2/2\pi^2c^3\beta$, and c is the velocity of light in the semiconductor. The equation for N_ω is

$$\frac{d}{dt}N_\omega = -\frac{N_\omega}{\tau} - c\alpha(\omega)N_\omega + c\alpha_0\sqrt{\hbar\omega - E_g}\,f_e f_h, \tag{3}$$

where $\tau \approx D/c$ is the time scale of the decay of a photon, which is determined primarily (in the case $D \sim 10$–$100\ \mu$m) by the time over which a photon escapes from the amplification region. The first terms on the right of Eqs. (1) and (2) describe the influx of carriers and energy due to the excitation pulse; the second terms describe processes of stimulated emission at frequencies $\omega < \Omega$. Spontaneous recombination of electrons and holes is taken into account only in (3), where it determines the rate of generation of the photons which serve as the trigger in the initial stage of the development of recombination superradiance.

Figure 1 shows the results of a numerical solution of Eqs. (1)–(3). In agreement with Refs. 1 and 2, a state of optical saturation, $\mu_e(T_1) - \mu_h(T_1) = \hbar\Omega$, is established even at the front of the excitation pulse.[7] The plasma temperature,

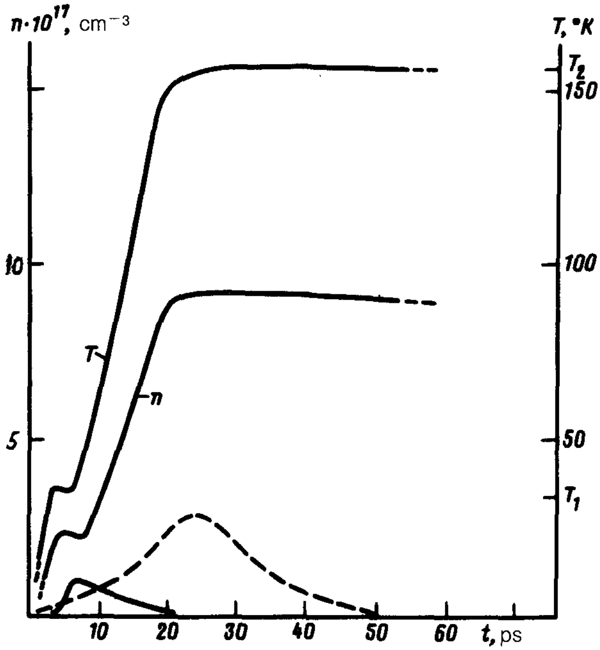

FIG. 1. Lines 1 and 2 show the time evolution of respectively the temperature and density of nonequilibrium carriers in GaAs during the application of an exciting pulse. Shown at the bottom is the behavior of the light intensity in the excitation pulse (the dashed line; the units are arbitrary) and that in the pulse of recombination superradiance (the solid line). The calculations were carried out with the following parameter values: The power density of the excitation pulse was 10^9 W/cm^2; the length of the excitation pulse (at half-maximum) was 25 ps; $\hbar\Omega - E_g^0 = 5$ meV; $D = c\cdot\tau = 20$ μm; $\gamma = 2\times 10^{-8}$ eV·cm; $\alpha_0 = 5.3\times 10^4$ cm^{-1}·eV$^{-1/2}$; $m_e/m_h = 0.1$; and $d = 1$ μm.

$kT_1 \approx 0.15(\hbar\Omega - E_g)$, which was found in Ref. 2, determines the gain. In our case we have $T_1 = 30$ K (with $\hbar\Omega - E_g \approx 17$ meV) and $\alpha_m \approx 2\times 10^3$ cm^{-1}. In the absence of recombination superradiance, the state of optical saturation would persist for times on the order of the electron-hole recombination time.

It can be seen from Fig. 1 that the recombination superradiance appears as a pulse which is shorter than the exciting pulse. The initial stage of development of the recombination superradiance plays out at an essentially constant value of α, so we find $N(\omega) \approx N_{0\omega}\exp(-\alpha ct)$ from (3), where $N_{0\omega}$ is determined by spontaneous emission (in our case we have $N_{0\omega} \sim 1$ and $\tau \gg 1/\alpha c \sim 0.1$ ps). An intense recombination superradiance develops in ~ 1 ps (the value of N_ω increases by several orders of magnitude). The trailing edge of the superradiance pulse is a consequence of a self-quenching of the superradiance, for which the mechanism can be outlined as follows: First, the recombination superradiance causes an escape of carriers, whose density decreases. This decrease disrupts the optical saturation and thus leads to a generation of carriers by the exciting light. Since the electron-hole pairs are created with an energy $\hbar\Omega - E_g$, while the light carries off a smaller energy $\hbar\omega - E_g$, the plasma heats up. As the plasma temperature rises, the gain decreases, so the intensity of the recombination

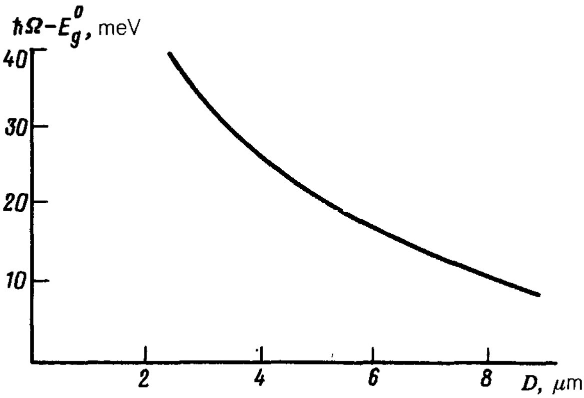

FIG. 2. In the region above the curve, the values of the parameters $\hbar\Omega - E_g^0$ and D are such that a recombination superradiance can occur in GaAs. These calculations were carried out on the basis of (4), with allowance for the renormalization of the width of the band gap.

superradiance decreases, as does the rate at which carriers escape. Very soon, the density of the electron-hole plasma begins to increase again. A theoretical analysis of Eqs. (1)–(3) shows that the semiconductor again reaches a state of optical saturation as a result of the heating, but in this case with a higher density and with a plasma temperature $T_2 > T_1$, which is determined by the condition for complete quenching of the recombination superradiance:

$$\alpha(\omega) \leqslant 1/\tau c; \qquad (E_g \leqslant \hbar\omega \leqslant \hbar\Omega). \tag{4}$$

In the approximation of equal effective masses and under the condition $(\hbar\omega - E_g)/kT_2 \ll 1$, we find from (4) $kT_2 = 1/\sqrt{6}$, $\alpha_0(\hbar\Omega - E_g)^{3/2}D$. Note that with decreasing difference $T_2 - T_1$, the quenching of the superradiance sets in earlier, lowering its intensity. Figure 2 shows the region in the plane of the parameters D and $\hbar\Omega - E_g^0$, in which a recombination superradiance can occur in GaAs.

Conclusions. The following conclusions can be drawn from this theory concerning events which accompany the production of an electron-hole plasma in a direct-gap semiconductor by intense light pulses (in the absence of an interaction with optical phonons).

1. A recombination superradiance arises in the form of a pulse (a picosecond pulse in the case of GaAs), whose length may in fact be shorter than that of the excitation pulse, because of a self-quenching of the superradiance.

2. The mechanism for the self-quenching of the superradiance involves a heating of the electron-hole plasma which results from the difference between the energies of the photoexcited electrons and holes, on the one hand, and those which undergo a stimulated recombination, on the other. In the course of this heating, the carrier density increases, and the semiconductor approaches a state of optical saturation, in which the temperature and density of the electron-hole plasma are determined by quenching conditions (4).

3. The intensity and length of the sueprradiance pulse, as well as the temperature and density of the electron-hole plasma, depend on the diameter of the spot of light focused on the semiconductor in the state of optical saturation.

We wish to thank I. L. Bronevoĭ, S. E. Kumekov, and V. I. Perel' for stimulating discussions and a useful review of the results of this study.

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Translated by Dave Parsons