Mechanism for reversible picosecond brightening of a direct-gap semiconductor during interband absorption of intense light pulses

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A brightening mechanism involving a generation and a recombination of charge carriers, both induced by a light, is proposed. A change in the saturation state during the pulse is caused by intraband absorption, which heats the carriers. The experimental results reported by Bronevoĭ *et al.* {Pis'ma Zh. Eksp. Teor. Fiz. **42**, 322 (1985) [JETP Lett. **42**, 395 (1985)]} are explained.

When a direct-gap semiconductor is illuminated by intense light with a photon energy $\hbar\omega_e$ slightly greater than the gap width E_g, a state of saturation, in which the populations of levels coupled by a direct optical transition are equal, is reached. In this state of saturation, the relation $\mu_e - \mu_h = \hbar\omega_e$ holds[2] (μ_e and μ_h are the Fermi quasilevels of electrons and holes). An unambiguous determination of the saturation state would also require use of the neutrality condition and the energy balance equation.[3,4] The evolution of the state density during the application of an intense light pulse was studied theoretically in Refs. 4 and 5. Bronevoĭ *et al.*[1] carried out an experimental study of the transparency of thin epitaxial films of GaAs during illumination with an exciting light pulse with a width ~ 30 ps at half-maximum and with a maximum energy $E_e^M \sim 100\,\mu$J after penetration through a probing region ~ 0.8 mm in diameter. They measured the absorption of a weak probing light pulse of roughly the same length, of tunable wavelength, which was incident on the sample with an adjustable delay τ_d. Bronevoĭ *et al.*[1] observed an approximately reversible change in the transparency during the pulse and found that the transparency after the pulse was independent of the pulse energy, in qualitative agreement with the conclusions of Ref. 5. This change in transparency, however, turned out to be far greater than would be expected if the only reason for it were a change in the quasiparticle spectrum,[6] as suggested in Ref. 5.

In the present letter we offer an explanation for the results of Ref. 1 under the assumption that the main reason for the reversible "brightening" of the sample is a generation of carriers at the front of the pulse and a photoinduced recombination at the trailing edge of the pulse, both consequences of intraband heating by the exciting light.

We first consider the change in the carrier state density during an intense pulse with $\hbar\omega_e - E_g < kT$ under the assumption of an infinitely rapid energy relaxation, as the carriers interact both with each other and with the lattice. With increasing intensity, the distribution of photoexcited carriers remains a Fermi distribution at all times with the lattice temperature T, and the difference $\mu_e - \mu_h$ increases until saturation is

reached. At this time, the sample becomes transparent to the exciting light; the generation ceases; and the electron and hole distributions remain constant as the intensity is subsequently increased and reduced (if we ignore the changes in the energy spectrum[6]). The quasilevels μ_e and μ_h are determined by the equations

$$\mu_e - \mu_h = \hbar\omega_{e'}, \qquad n = p \tag{1}$$

at the lattice temperature.

We now take into account the finite duration of the interaction with the lattice, τ_ϵ. After saturation is reached, the carrier temperature rises with increasing intensity as a result of intraband absorption. If a state of saturation is to be maintained [i.e., if relation (1) is to be maintained], the carrier concentration should increase. This increase actually results from a small deviation of the populations of the resonant levels from equality, with the lower level having the higher population. When the intensity begins to decrease, the heating decreases, and the carrier temperature and thus the concentration also decrease. The lowering of the concentration occurs as a result of a photoinduced recombination, by virtue of the small amount by which the population of the upper resonant level exceeds that of the lower level. After the pulse, the carriers revert to a saturation state with the lattice temperature.[1)]

Two qualitative conclusions follow from this interpretation. 1) The state of the carriers after the pulse is a saturation state with the lattice temperature. It is determined by conditions (1) and does not depend on the pulse energy. This conclusion agrees completely with the results of Ref. 1. For the experimental conditions of Ref. 1, Eqs. (1) lead to the following values of the parameters after the pulse: $T_e = T = 25$ meV, $n = p = 2\times 10^{18}$ cm^{-3}, $\mu_e = E_c + 60$ meV, and $\mu_h = E_v + 50$ meV (E_c and E_v are the energies of the band edges). 2) Once saturation is reached, the transparency changes in a reversible way, following the changes in the intensity during the pulse, with a time delay on the order of τ_ϵ. Figure 1 compares the cross-correlation function of the exciting and probing pulses (solid line) with the time dependence of the delay τ_d of the increment in the transparency, δD, which vanishes at the end of the excitation: $\delta D = \log(T^{(1)}/T^{(0)}) - \log(T^{(s)}/T^{(0)})$. Here $T^{(0)}$, $T^{(1)}$, and $T^{(s)}$ are the transparencies of the sample in the absence of the exciting pulse, in its presence, and after it. According to the model outlined above, δD stems from intraband heating of carriers.

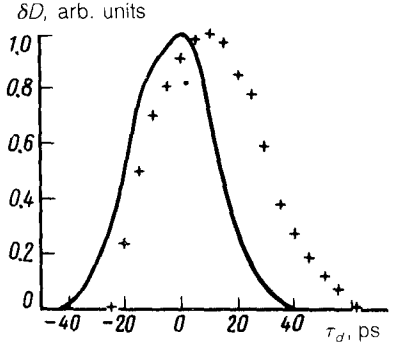

FIG. 1.

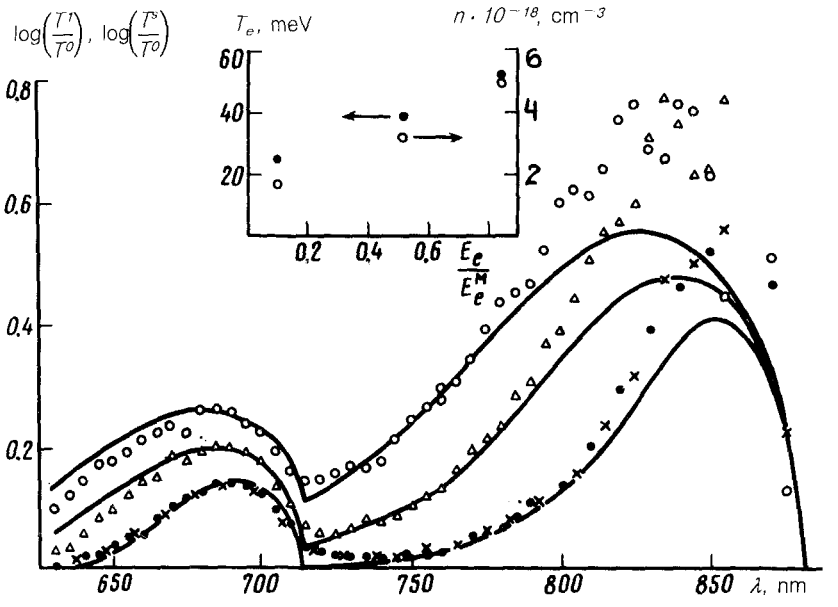

FIG. 2. Spectra of the change in transparency.[1] ○,△—During synchronous probing; ●,×—after the pulse. ○,●—$E_e = 0.85\ E_e^M$; △,×—$E_e = 0.53\ E_e^M$. The solid lines are theoretical.

We see that these results (taken from Ref. 1) confirm the reversibility of the change in transparency. From the results we find the value $\tau_\epsilon \approx 10$ ps.

We have attempted to find a quantitative description of the spectra of changes in the transparency on the basis of the interpretation above. Figure 2 shows experimental data from Ref. 1, along with the results of our calculations. In the calculation of the brightening spectrum after the pulse, we used the values of μ_e and μ_h found above, and we set $T_e = T$. For the curves corresponding to a probing synchronized with the excitation, the temperature T_e serves as an adjustable parameter, while μ_e and μ_h are determined by Eqs. (1). In order to achieve agreement with the experimental spectra, we need to shift the calculated curves 25 meV up the wavelength scale. This shift agrees with the decrease caused in E_g by the Coulomb interaction of photoexcited carriers.[7] The calculation ignores the brightening caused by the filling of states of electrons, heavy holes, and light holes; furthermore, the Sommerfeld factor is ignored. We see a good agreement between the theoretical and experimental results everywhere except near the absorption edge, where screening of the exciton effect makes an additional contribution to the brightening. The points in the inset in Fig. 2 show the values of T_e found from the fit, along with the corresponding concentrations calculated from condition (1). A rough estimate based on the values found here for T_e and the data of Ref. 8 for intraband absorption yields $\tau_\epsilon \sim 10$ ps, in agreement with the data on the time evolution of the brightening (Fig. 1). Such long values of τ_ϵ are apparently a consequence of a heating of phonons[9] or a screening of phonons by photoexcited charge carriers.[10]

Although the renormalization of the quasiparticle spectrum is apparently not being manifested in these effects, the actual ideas involving quasiparticles which were advanced by Galitskiĭ, Goreslavskiĭ, and Elesin[6] have essentially been used in this study. It follows from those ideas that all the intraband relaxation processes, including collisions between charge carriers, lead to a saturation state in the intense field of the light wave.

[1)] If $\hbar\omega - E_g \ll kT$, then essentially the same state after the pulse would result from another possible cause of the rapid concentration decay: recombination superradiance, which is possible under the condition $\mu_e - \mu_h > E_g$.

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