Anomalous emission from gallium arsenide during interband absorption of intense picosecond light pulses

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An anomalous emission from gallium arsenide during interband absorption of intense picosecond light pulses has been observed experimentally. The emission peaks at a photon energy $< E_g^0$. The emission requires the attainment of a threshold. It correlates with the time evolution of the intensity of the exciting light in the picosecond range.

The illumination of a direct-gap semiconductor by an intense picosecond light pulse with a photon energy $\hbar\omega_e$ slightly greater than the band gap of the unperturbed sample, E_g^0, in the experiments of Refs. 1 and 2 was accompanied by an approximately reversible change in the transmission spectrum of the semiconductor during the pulse. The effect becomes more apparent with increasing pulse energy W_e. After the pulse, however, the transmission spectrum $T_1(\lambda)$ of the sample does not depend on W_e. The experimental results of Ref. 1 were explained theoretically in Ref. 3 under the assumption that the reversible increase in transmission of the sample was caused primarily by a generation of carriers at the front of the pulse and a light-induced recombination at the trailing edge of the pulse. After the pulse, the carrier temperature is equal to the lattice temperature, and the saturation condition

$$\mu_e - \mu_h = \hbar\omega_e$$

should hold. Here μ_e and μ_h are the electron and hole Fermi quasilevels. A second reason for the rapid decay of the density (N) of photoexcited carriers to the value which remains after the pulse ($N_1 = N(T_1)$, might be some type of cooperative spontaneous emission (e.g., recombination superradiance was cited in Ref. 3). This effect might occur under the condition $\mu_e - \mu_h \geqslant E_g$, where E_g is the band gap. If the cooperative emission is the basic reason for the rapid decay of N, the following condition should hold after the pulse:

$$\mu_e - \mu_h \approx E_g .$$

In the experiments of Ref. 1, $\hbar\omega_e$ was close to E_g, so it was not possible to draw a conclusion about the involvement of cooperative emission in the mechanism for the rapid decay of N. In some later experiments, we also observed an approximately reversible change in transmission at values of $\hbar\omega_e$ well above E_g. After the excitation was terminated, however, the increase in the transmission of the sample did not depend on $\hbar\omega_e$. It follows that under these conditions the rapid decay of N cannot be described exclusively as a recombination induced by the exciting light[3]; a cooperative emission apparently also arises. In the experiment which we report below, we did indeed observe an emission which arises under conditions of this sort. However, it would be

premature to draw a final conclusion about its nature, because, in particular, we lack a theory for cooperative emission from a semiconductor which would be applicable under these conditions.

The test samples were $Al_{0.5}Ga_{0.5}As$–GaAs–$Al_{0.5}Ga_{0.5}As$ heterostructures, which were described in Ref. 1 and which had respective layer thicknesses of 1.1–2.1–0.6 μm. The $Al_{0.5}Ga_{0.5}As$ layers were transparent to light with $\hbar\omega < 2.2$ eV.

Depending on the purpose of the particular experiment, we used either one or two light pulses, from two simultaneously pumped parametric picosecond sources. The light was focused onto the sample at an angle $\sim 10°$ from its normal; in the case of two pulses, the angle between the beams was $\sim 3°$, and the polarizations were identical. The maximum energy in a single pulse was $W_e^M \sim 100\ \mu$J at a pulse length $T_p \sim 20$ ps and at a focal-spot diameter ~ 0.6 mm. The value of W_e was adjusted by varying the thickness of a neutral absorber with a calibrated attenuation coefficient. An adjustable optical delay τ_d was placed in the path of the second beam. The value $\tau_d = 0$ corresponded to the position of the maximum of the cross-correlation function $G(\tau_d)$ of the two exciting pulses. Part of the emission which arose in the sample during excitation, from a solid angle $\sim 5°$, was focused onto the slit of a monochromator; the exciting rays passed outside the aperture of the focusing system. The energy of the emission from the sample, W_S, was measured from the integrated output signal from a photomultiplier at the exit slit of the monochromator; these measurements were taken as we varied the wavelength λ_S, the energy W_e, and the delay τ_d.

Figure 1 shows spectra of the observed emission from the sample during excitation by light pulses with $\hbar\omega_e \approx 1.49$ eV ($\hbar\omega_e > E_g^0 = 1.423$ eV) for two excitation energies. We see that the emission spectrum is at energies $\hbar\omega_S < E_g^0$ and has a maximum which increases and shifts toward longer wavelengths as W_e is increased.

Measurements of the dependence $W_S = f(W_e)$ at various points in the spectrum

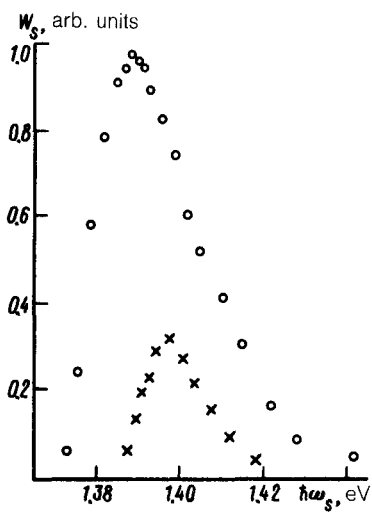

FIG. 1. ×—Emission spectrum of a sample excited by light with $\hbar\omega_e = 1.487$ eV. O—results obtained at an exciting-pulse energy higher by a factor of ten, and otherwise under the same conditions.

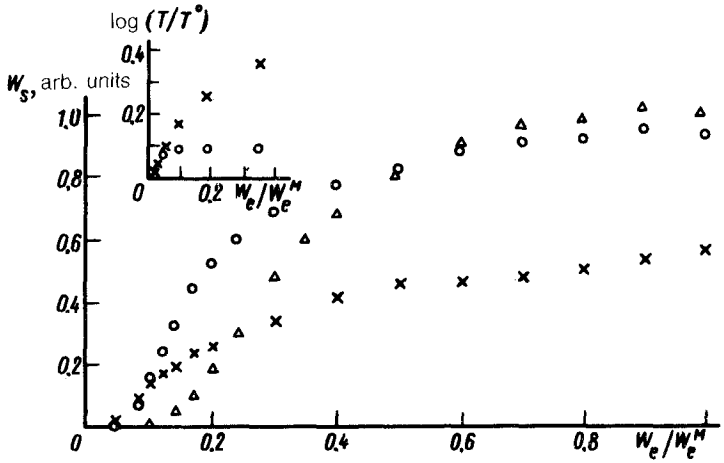

FIG. 2. Energy of the emission from the sample versus the energy of the exciting pulse with $\hbar\omega_e = 1.487$ eV. ×—$\hbar\omega_S = 1.406$ eV; ○—1.395 eV; Δ—1.385 eV. The inset shows the change in the transmission of GaAs for probing light with $\hbar\omega = 1.566$ eV during excitation by a pulse with $\hbar\omega_e = 1.490$ eV. ○—Measured with a delay of 60 ps; ×—5 ps. Here T and T^0 are the transmission levels of the sample in the presence and absence of excitation, respectively.

revealed that it is nonlinear and involves a threshold (Fig. 2): A significant emission appears at a certain $W_e > 0$ and exhibits a tendency toward saturation. This latter tendency may be caused by an increase in the brightening of the sample during excitation.

In a following experiment, the sample was illuminated by two pulses, with approximately equal values of W_e and slightly different values of $\hbar\omega_e$. The delay τ_d between the pulses was varied, so we were able to vary the intensity distribution of the exciting light over time at a fixed total energy. These experiments revealed a correlation dependence between the energy of the emission from the sample, W_S, at λ_S = const, on the one hand, and the delay τ_d, on the other (Fig. 3). This dependence has a minimum at $\tau_d \approx 0$, as follows from a comparison with the function $G(\tau_d)$, shown in the same figure, and it also has two approximately symmetric side maxima.

The emission from the sample apparently occurs even during the excitation pulse, at which time we also observe a reversible increase in transmission (the emission thus arises over a time much shorter than, for example, the recombination time $\tau_R \sim 500$ ps for a weakly excited sample). There are two circumstances that point to this conclusion.

First, at significant levels of W_e the emission from the sample continues to grow as W_e is raised, and the spectrum of this emission undergoes a broadening, including a broadening in the long-wavelength direction (Figs. 1 and 2), while the state of the carriers after the pulse (inferred from the transmission spectrum) and thus the emission spectrum of these carriers have become independent of W_e (see the inset in Fig. 2). The broadening of the spectrum in the long-wavelength direction with increasing

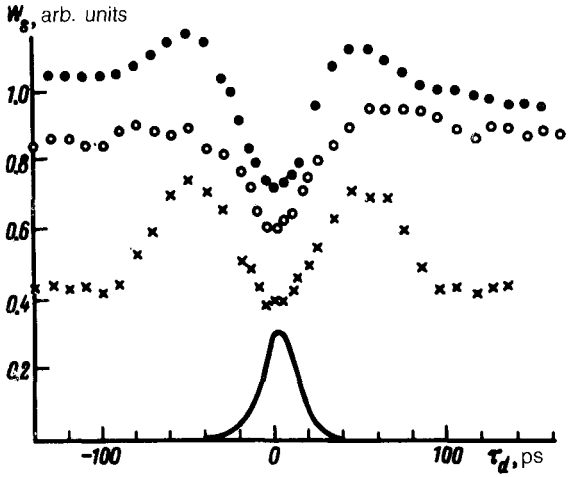

FIG. 3. Energy of the emission with $\hbar\omega_S = 1.395$ eV from a sample excited by two pulses, with $\hbar\omega_e = 1.494$ and 1.483 eV, versus the delay in the exciting pulses. ×—$W_{S_1} = 0.21$, ○—$W_{S_1} = 0.54$; ●—the same, but at $\hbar\omega_S = 1.385$ eV and $W_{S_1} = 0.62$ (W_{S_1} is the energy of the emission from the sample from each separate excitation pulse). Solid line—the function $G(\tau_d)$.

W_e apparently stems from a decrease in E_g during the time interval in which the sample is being excited, because of a Coulomb interaction of carriers[3] and a dynamic Stark effect.[4,5]

Second, under this assumption we can reconcile the dependence $W_S(\tau_d)$ with the dependence $W_S(W_e)$.

The presence of maxima on the $W_S(\tau_d)$ curves may be a consequence of the threshold involved here and of the superlinear behavior $W_S(W_e)$ in the initial stage. The fact that a change in W_S with a change in τ_d becomes observable at $|\tau_d| > T_p$ can be linked with a prolongation of the carrier energy relaxation, which was noted in Ref. 3 for similar experimental conditions. To the best of our knowledge, a correlation dependence of this type for the emission from a semiconductor has not previously been observed.

The threshold involved in the dependence $W_S = f(W_e)$ at $\lambda_S = \text{const}$ is apparently due to the formation of an inversion on this transition.

The distinctive properties of the observed emission which we have outlined here are clearly of interest. Reaching a complete understanding of the nature of the emission, however, will require further research on this topic. A similar emission may possibly cause the picosecond evolution of the gain spectrum which has been observed in multilayer quantum structures[6] and the appearance of an additional emission peak in Ref. 7. The experiments described here lead to the conclusion that the observed emission must be taken into consideration in analyzing effects involving a saturation of the optical absorption of semiconductors.

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