Stimulated emission and the Mott transition in direct-gap semiconductor

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(Submitted 29 January 1990)
Pis'ma Zh. Eksp. Teor. Fiz. **51**, No. 4, 219–222 (25 February 1990)

A change in the delay of a stimulated-light pulse in CdSe (80 K) from 30–40 ps at high excitation levels to 0.6–1 ns at low excitation levels of ultrashort light pulses has been detected. This change is attributable to the change in the recombination mechanism: a transition from an electron-hole plasma to high-density excitons.

In strongly excited semiconductors a screening of Coulomb interaction causes a Mott transition to occur from a weakly ionized exciton gas to a strongly ionized electron-hole plasma (EHP). A decrease in the density of the electron-hole plasma as a result of recombination and diffusion causes an inverse transition from the EHP to excitons. Many attempts to detect the Mott transition from the change in the luminescence spectra of II–VI direct-gap semiconductors were unsuccessful, because the emission spectra of the dense exciton gas and EHP are nearly the same, and because they are strongly distorted due to amplification. The appearance of structural features in the reflection spectra, characteristic of excitons[1] (the kinetics of the luminescence and reflection spectra were measured concurrently), may be unrelated to the Mott transition, since a similar structure may appear in the reflection spectrum during the decay of the electron-hole plasma.[2] A sudden change in the photocurrent, which is determined by the Mott transition from excitons to electron-hole plasma, has not been observed.[3]

In the present letter we report the results of an experimental study of the luminescence of CdSe single-crystal wafers (80 K) 40–50 μm thick, produced as a result of interband excitation by ultrashort pulses from a neodymium laser. The length of these pulses was no greater than 10 ps and the maximum energy was $W = 2.5$ μJ. The laser beam was aimed at a 45° angle with respect to the surface of the samples and focused in a spot 0.1 mm in diameter. The kinetic measurements were carried out using a AGAT-SF high-speed photoelectric recorder (HPR) with a resolution of up to 5 ps. The image of the end-face of the crystal, which scattered the amplified luminescence light, was projected along with the image of the excited region onto the gap of the spectrograph and the HPR. To determine the luminescence rise time and the delay time of the stimulated light pulses, we have simultaneously aimed at the HPR input the pedestal pulses of the pump light which was reflected from the crystal.

At $W = 0.04$–0.8 μJ a broad Q band (690 nm) was the dominant band (Fig. 1) in the light from the excited region. The enhanced-luminescence track appeared at $W > 0.2$ μJ. The induced-luminescence band R' (702 nm) intensified at the long-wavelength wing of the Q band. At maximum pumping the stimulated luminescence (the R band) becomes noticeable even in the light from the exciting region.

When the pumping is done below the threshold at which the stimulated processes

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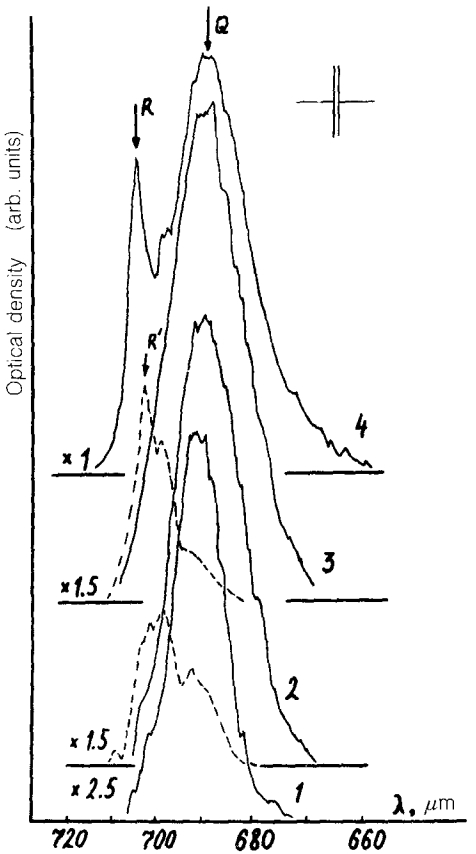

FIG. 1. Densitometer traces of the emission spectra of CdSe (80 K) from the excited volume (solid line) and of the enhanced luminescence light scattered at a crystal end-face (dashed curve) at various ultrashort pulse pumping energies (W). 1—$0.5W_0$; 2—$0.3W_0$; 3—$0.42W_0$; 4—W_0; $W_0 = 0.8$ μJ.

begin to develop, the maximum of the luminescence pulse from the exciting volume (Fig. 2b) is delayed by 0.6 ns relative to the ultrashort pulses. With an increase in W to 0.2 μJ, we recorded a stimulated-light pulse, whose delay time is $\Delta t = 0.9$ ns (Fig. 2c). The vertex of the light pulse from the excited volume in this case exhibits a narrow spike, which probably stems from the induced luminescence (the spike occurred simultaneously with a short pulse of amplified light). At $W > 0.4$ μJ the delay of the light pulses from the exciting region and from the sample's end face decreased radically (Fig. 2d); at a maximum pumping $\Delta t \approx 30$–40 ps (the inset in Fig. 2). The insignificant changes in the luminescence spectra produced by varying the excitation level at first glance allows us to assume that only one recombination mechanism, which is associated with the decay of the electron-hole plasma, develops over the entire range of pump power used experimentally. This conclusion is contradicted by the results of the measurements of the delay of stimulated-light pulse in the pumping of ultrashort pulses. These results differ considerably in the case of strong excitation ($\Delta t_1 = 30$–40 ps) and weak excitation ($\Delta t_2 = 0.6$–0.9 ns), suggesting that at least two recombination mechanisms are being developed: recombination of the electron-hole plasma and of the dense exciton gas. The delay of 30–40 ps is evidently associated with

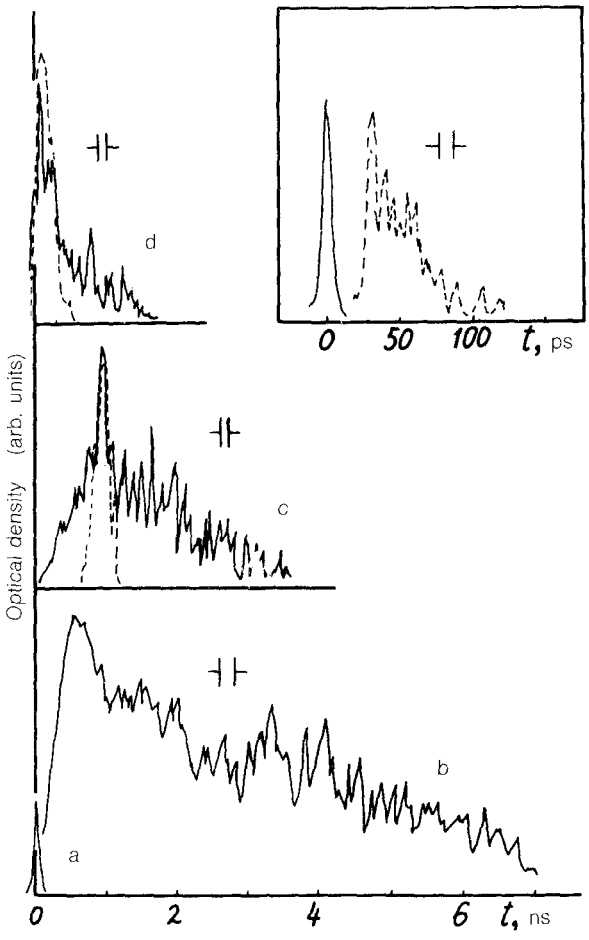

FIG. 2. (a) Densitometer traces of an excitation pulse, of the emission pulses of CdSe (80 K) from the excitation volume (solid line), and of the enhanced luminescence light scattered at the crystal end-face (dashed curve) at various ultrashort pulse pumping energies (W); (b) 0.8 μJ; (c) 0.28 μJ; (d) 0.44 μJ. Inset—0.8 μJ.

the time it takes the EHP to cool down from the initial temperature, which is determined by the excess energy of 0.53 eV of the carriers, to a temperature at which the enhancement of the EHP is greater than its loss. The relatively slow cooling of the EHP in the case of polaron semiconductors may be attributed to the nonuniform filling of the LO phonon modes and to the screening of electron-phonon interaction.[4,5] The rapid diffusion broadening[6] and recombination reduce the density (n_e) of the electron-hole plasma. At $n_e < n_M$ (n_M is the density of the Mott transition from EHR to the exciting gas) the excitons can form. The delay Δt_2 in this case is determined by the time it would take to accumulate an adequate number of excitons for the development of stimulated processes in the exciton system. An increase of the time scale for binding into excitons can be attributed to the screening of the EHP. This is suggested by the shorter luminescence intensity rise time as the exciting level is reduced (Fig. 3). In the absence of screening the cross section for binding of carriers into excitons, σ_0, does not depend on n_e, and the time scale for the binding is $\tau = (\sigma_0 v_t n_e)^{-1}$ (v_t is the

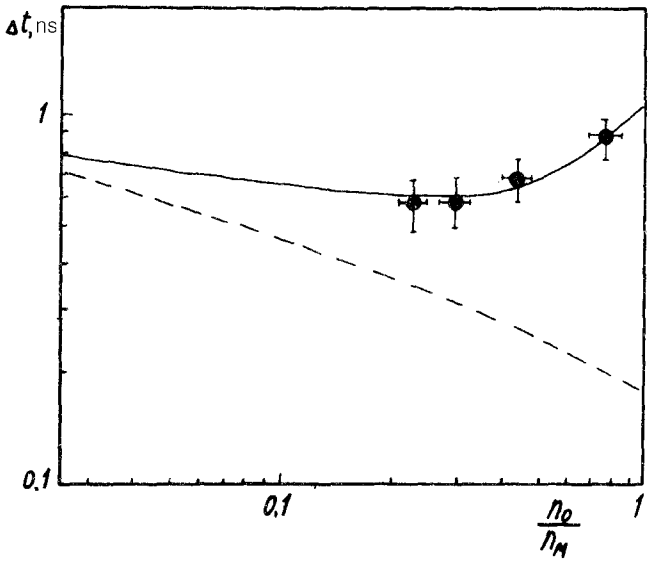

FIG. 3. Delay time Δt of the maximum of the exciton luminescence intensity vs the initial concentration of the photoexcited carriers n_0 ($n_0 \sim W$) ignoring the screening (dashed line) and taking into account the screening (solid line) and the experimental results.

thermal velocity of carriers) increases with decreasing density. The effect of screening on the electron-hole interaction was taken into account by introducing a n_e-dependent dielectric constant: $\varepsilon \sim [1 - n_e/n_M]^{-1}$. Here $\sigma = \sigma_0[1 - n_e/n_M]^3 [\sigma \sim 1/\varepsilon^3$ (Ref. 7)]. Two coupled kinetic equations for the carrier and exciton densities were integrated numerically. The experimental results for the measurement of the luminescence rise time are in agreement with the calculated values for $\sigma_0 = 0.6 \times 10^{-13}$ cm^2 (Fig. 3).

The EHP-(exciton gas) transition corresponds to such a carrier density at which the renormalized edge of the conduction band coincides with the exciton level.[8] In the case of direct recombination the spontaneous and stimulated EHP emission lines should be situated, in terms of energy, near the exciton transition, in contradiction of the experimental results (Q, R, and R' luminescence bands had much longer wavelengths). It can be assumed that in the case of EHP recombination the dominant transitions are not the direct transitions, but rather those involving the emission of LO phonons (plasmon-phonon mixing at $n_e \sim 10^{17}$ cm^{-3} can be ignored). The ratio of the probabilities for the indirect recombination (W^i) and direct recombination (W^d) of an electron in a degenerate EHP is

$$\frac{W^i}{W^d} = \frac{2}{\pi} \frac{e^2}{\hbar\omega_{LO}} \left[\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right](k + k_F)$$

$$\geqslant \frac{2}{\pi} \frac{e^2}{\hbar\omega_{LO}} \left[\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right] k_F \geqslant 0.3,$$

when $n_e \gtrsim 10^{17}$ cm^{-3} (k is the electron wave vector, k_F is the Fermi wave vector, and ε_0 and ε_∞ are the static and the rf dielectric constants). The transitions involving LO phonons become more important in the case of nondegenerate EHP, when the radiation corresponding to direct transitions decreases due to overabsorption in the exciting volume.

We note in conclusion that analogous results on the variation in the delay of the stimulated light were obtained for CdS (80 K) upon excitation by ultrashort pulses of the third-harmonic light from a neodymium laser.

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Translated by S. J. Amoretty