Kinetics of the formation and energy relaxation of excitons in GaAs

Ya. Yu. Aaviksoo, I. Ya. Reĭmand, V. V. Rossin, and V. V. Travnikov
Institute of Physics, Academy of Sciences of the Estonian SSR, Tartu A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 11 March 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **53**, No. 7, 377–381 (10 April 1991)

Experimental study of the kinetics of polariton luminescence of GaAs has shown that the radiation pulse is delayed because of the energy relaxation of electrons and excitons. The energy relaxation was found to be delayed with decreasing concentration of small impurities.

At liquid-helium temperatures the final exciton state is the final stage of the energy relaxation of an excited crystal. We have used the most direct method of determining this energy relaxation by analyzing the kinetics of exciton luminescence produced as a result of a pulsed excitation to different energy states.

The luminescence was excited by a stiril-9 dye laser which was pumped by a mode-locked krypton laser. The length of the excitation pulse was 5 ps, the pulse spacing was 240 ns, and the half-width of the instrumental function of the measuring system was 300 ps. The spectral width of the excitation line, ≈ 0.7 meV, and the spectral resolution, ≈ 0.1 meV, have made it possible to clearly determine the excitation energy and the luminescence energy. We investigated pure epitaxial GaAs layers with various concentrations of residual impurities and various conductivities. The concentration of the shallow donors in the purest Γ13 sample was estimated to be[1] $\approx 10^{12}$ cm^{-3}. The stationary spectrum of this sample is characterized by an intensive polariton luminescence line which is comparable in intensity to the emission lines of bound excitons.[2]

The spectra and the kinetics of exciton luminescence of various samples at a temperature $T = 1.7$ K are shown in Fig. 1. The luminescence pulses were recorded at energies which are indicated by arrows *1* and *2* and which correspond to the radiation from the states of the lower and upper polariton branches. The rise time of the luminescence pulse was found to increase in all of the samples with decreasing energy of the emitting polaritons. The time lag of the maximum of the luminescence pulse, $\Delta t_{\max}$, is plotted in Fig. 2a as a function of radiation energy. This dependence represents the "bottleneck" effect for polaritons, which occurs as a result of the slowing down of the energy relaxation of polaritons near the exciton resonance.[3] The kinetics of relaxation of polaritons was previously reported in Refs. 4–6 in which CdS crystals were used. In contrast with those studies, the bottleneck effect in our case manifested itself in the increase of the rise time of the pulse, and the damping time, equal to $\tau_d \approx 4$–5 ns (sample Γ13), depended only slightly on the luminescence energy. We note that this value of τ_d is higher than the value of the "radiative" lifetime of an exciton obtained in Ref. 7.

A weak dependence of the rise time on the energy of exciton luminescence of

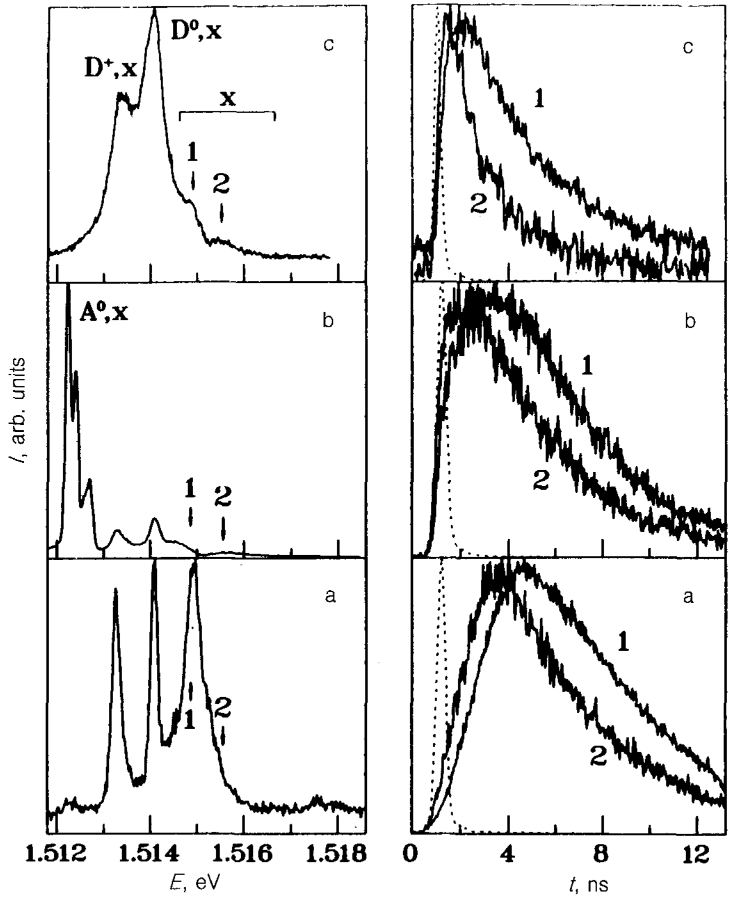

FIG. 1. Spectra (left) and kinetics (right) of the luminescence of various GaAs samples. (a) Γ13($N_D \sim 10^{12}$ cm^{-3}); (b) Yl(*p*-GaAs, $N_A \sim 10^{14}$ cm^{-3}); (c) K88 (*n*—GaAs, $N_D \sim 10^{14}$ cm^{-3}); excitation energy—1.554 eV; ···—laser pulses; curves *1* and *2* correspond to the radiation pulses from the energy states of the lower and upper polariton branches, denoted by arrows *1* and *2* in the spectra.

GaAs was observed in Ref. 8, where the luminescence was linked with the diffusion of polaritons from the interior to the surface. To determine the role of diffusion, we investigated the time-dependent damping of radiation produced as a result of annilation of polaritons from the resonance region, with a simultaneous emission of a *LO* phonon. Since the absorption for such a radiation is very small and the group velocity is large, the spatial distribution of polaritons cannot affect the time-dependent radiation kinetics. The luminescence kinetics of polaritons from the resonance region and from the corresponding points of *LO* phonon repetition was not markedly different. It can thus be concluded that diffusion does not significantly affect the observed delay of resonance polariton luminescence pulses.

The delay of an exciton luminescence pulse as a result of interband excitation was previously observed in GaAs in Ref. 9, but this delay was an order of magnitude

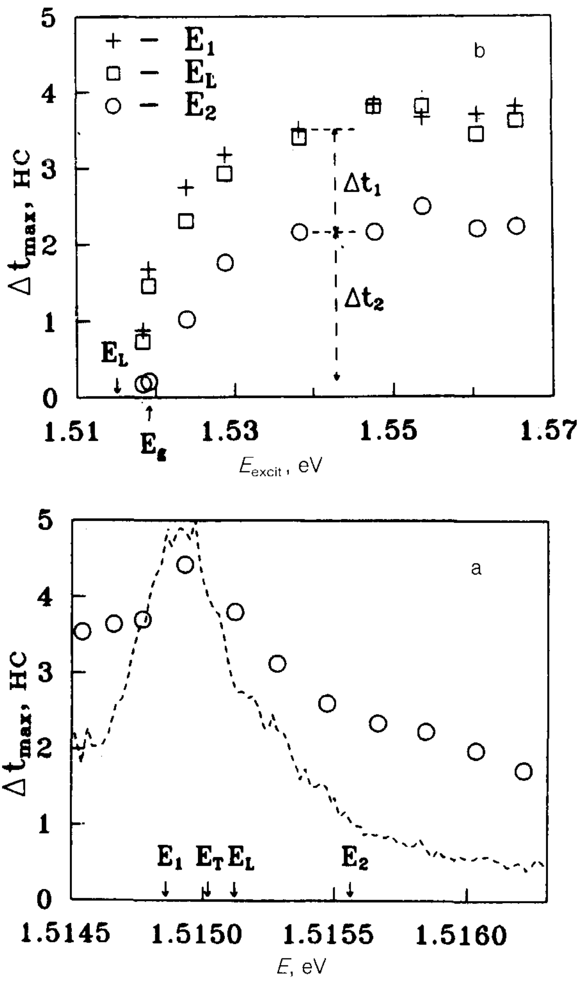

FIG. 2. Delay of the maximum of the radiation pulse versus the energy of the investigated point in the luminescence spectrum [(a) excitation energy—1.554 eV; ···—polariton luminescence spectra] versus the excitation energy [(b) for three points in the luminescence spectrum, shown in Fig. 2a]; E_T and E_L—energies of the bottom of the bands of the tranverse and longitudinal excitons, respectively. Sample Γ13.

smaller than that obtained by us. In GaAs, the main channel for the formation of excitons as a result of excitation of a semiconductor by light whose energy is higher than the width of the band gap is the binding of cold charge carriers into excitons.[10] The delay may therefore be caused by the energy relaxation of the excited charge carriers (such a possibility was noted in Ref. 9) and by their binding into excitons.

The rise time of the polariton luminescence pulse depends essentially on the excitation energy (Fig. 3). We see from Fig. 2b that there are two components of the delay of the luminescence maximum. The first component is virtually independent of the excitation energy and is associated with the energy relaxation of excitons (it is typically $\Delta t_1 \approx 1.5$ ns). The second component, Δt_2, increases (by ≈ 2 ns) for all lumines-

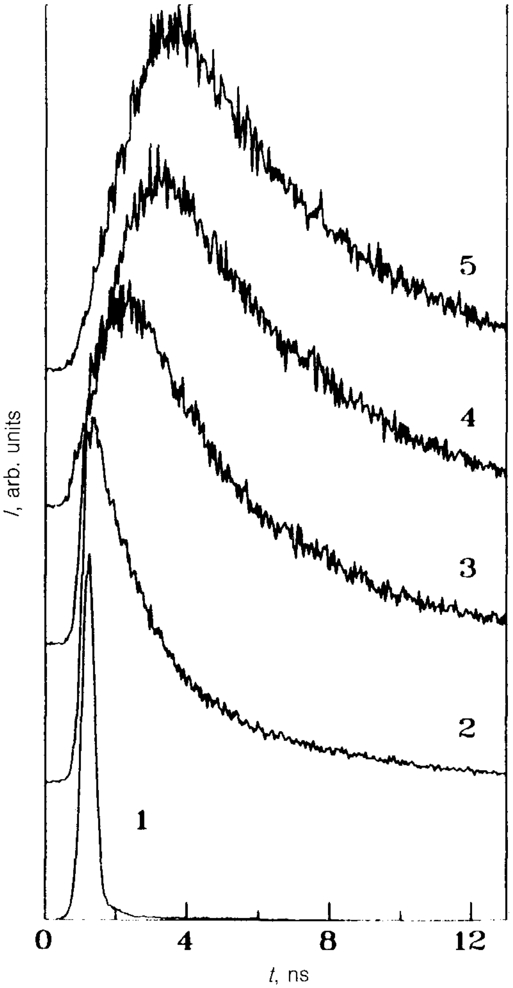

FIG. 3. Kinetics of luminescence from the states of the upper polariton branch (luminescence energy—1.5156 eV) for various excitation energies: *2*—1.520 eV; *3*—1.524 eV; *4*—1.538 eV; *5*—1.554 eV; *1*—laser pulse. Sample Γ13.

cence energies when the excitation energy is increased above the width of the band gap, E_g. This component reflects the energy relaxation of electrons. At the same time, the values of Δt_{max} for polariton luminescence pulse of the upper branch upon excitation to the band edge and to the excited state of an exciton $n = 2$ (the experimental points corresponding to two minimal values of the excitation energy in Fig. 2b) are nearly the same, suggesting that the direct exciton production has virtually no effect on the delay of the polariton luminescence pulse.

As can be seen in Fig. 1, the rise time of the polariton luminescence pulse varies from sample to sample. In an ultra-pure material, the main mechanism for the energy relaxation of electrons, whose energy is lower than that of an optical phonon, is the relaxation on acoustical phonons, which is very slow because of the small effective mass of the electrons in GaAs (according to the estimates in Ref. 11, at an electron temperature $T_e = 10$ K the rate of the energy relaxation of electrons on acoustical phonons in GaAs is $S = 0.1$ meV/ns). An increase in the concentration of small impurities greatly decreases the delay time of the maximum of the polariton luminescence pulse. This suggests that the rate of energy relaxation of electrons increases, for example, because of inelastic scattering by impurities. We note that in an *n*-type sample (Fig. 1c) the rise time of a luminescence pulse is much shorter than that in a *p*-type sample with a comparable concentration of small impurities (Fig. 1b). We find this behavior quite natural since the energy relaxation of electrons is expected to be affected most strongly by the inelastic scattering by small donors.

[1] J. G. Golubev, Yu. V. Zhilyaev, V. I. Ivanov-Omskii *et. al*, Fiz. Teor. Polupro. vodn. **21**, 1771 (1987) [Sov. Phys. Semicond. **21**, 1074 (1987)].

[2] Yu. V. Zhilyaev, G. R. Markaryen, V. V. Possin *et. al.*, Fiz. Tverd. Tela **28**, 2688 (1986) [Sov. Phys. Solid State, **28**, 1506 (1986)].

[3] J. Toyozawa, Suppl. Progr. Theor. Phys. **12**, 111 (1959).

[4] P. Wiesner and U. Heim, Phys. Rev. B **11**, 3071 (1975).

[5] F. Askary and P. Y. Yu, Phys. Rev. B **28**, 6165 (1983).

[6] Ya. Yu. Aaviksoo, R. E. Lippmaa, A. M. Freiberg, and S. F. Savikhin, Fiz. Tverd. Tela **31**, 203 (1989) [Sov. Phys. Solid State, **31**, 462 (1989)].

[7] G. W. 't Hooft, W. A. van der Poel, L. W. Molenkamp, and C. T. Foxon, Phys. Rev. B **35**, 8281 (1987).

[8] T. Steiner, M. L. W. Thewalt, E. S. Koteles, and J. P. Salerno, Phys. Rev. B **34**, 1006 (1986).

[9] C. Weisbuch, Solid State Electr. **21**, 179 (1978).

[10] R. Höger, E. O. Göbel, J. Kuhl *et al.*, J. Phys. C **17**, L905 (1984).

[11] R. Ulbrich, Phys. Rev. B **8**, 5719 (1973).

Translated by S. J. Amoretty