Manifestation of metastable localized hole states in the slow kinetics of the edge luminescence of *n*-GaAs

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A study has been made of the effect of heat pulses on the microsecond-range kinetics of the (free hole)-(neutral donor) luminescence which has been observed in *n*-GaAs at $T_0 = 1.7$ K. The results of this study show that this kinetics is governed by a slow, nonactivated replenishment of the concentration of free holes from localized metastable states with an energy below the top of the GaAs valence band.

The kinetics of the edge luminescence of GaAs at liquid-helium temperatures is characterized by time scales $\sim 10^{-8}$ s, which are set by the fast capture of photoexcited carriers by ionized impurities (Ref. 1, for example). In this letter we report the first observation of a slow component (10^{-6} s) in the decay of the *n*-GaAs photoluminescence spectrum, which is associated with the participation of free holes. The slow component is shown to be caused by a pronounced time delay of the holes in certain localized states with energies below the top of the valence band. The liberation of holes from these states does not involve an activation energy.

The experiments were carried out at $T_0 = 1.7$ K. We studied single-crystal *n*-GaAs films ($n_D - n_A = 10^{14}$–10^{16} cm^{-3}) with a thickness of 2–400 μm grown on Si-GaAs substrates (0.4 mm thick) in the (001) orientation by liquid-phase epitaxy or by gas-phase epitaxy either in a chloride system or by an organometallic compound-hydride method. Figure 1 shows the photoluminescence spectra of several *n*-GaAs samples which we measured with a DFS-24 spectrometer during weak ($P \sim 10^{-2}$ W/cm^2), steady-state excitation with a He–Ne laser ($\lambda = 633$ nm). In general, these

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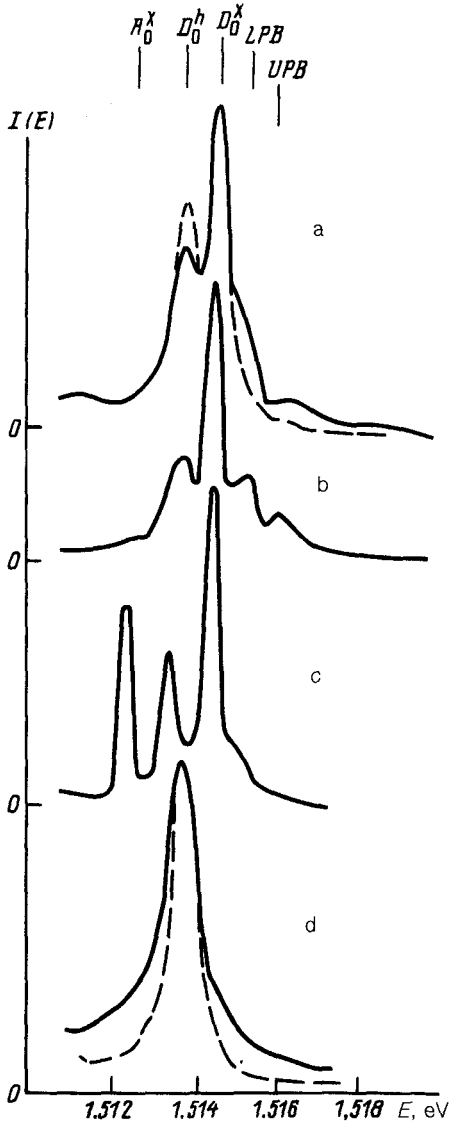

FIG. 1. Steady-state photoluminescence spectra at $T_0 = 1.7$ K of n-GaAs samples with the following values of $n_D - n_A$: a—9×10^{14} cm^{-3}; b—4×10^{14} cm^{-3}; c—3×10^{14} cm^{-3}; d—5×10^{15} cm^{-3}. The dashed lines are spectra recorded after a time delay of 1 μs.

spectra agree with the existing data (see Ref. 2 and the bibliography there). At $n_D - n_A \leqslant 10^{15}$cm^{-3}, the spectrum contains lines (Fig. 1, a and b): lines of free excitons of the upper polariton branch, UPB ($E = 1.5153$ eV), and of the lower branch, LPB ($E = 1.5150$ eV); of an exciton bound at a neutral donor, D_0^x ($E = 1.5141$ eV); and of the transition D_0^h ($E = 1.5133$ eV), which corresponds to the recombination of a free hole h and an electron at a neutral donor. The introduction of acceptors also causes the appearance in the spectrum of the line of an exciton bound at a neutral acceptor, A_0^x ($E = 1.5125$ eV; Fig. 1c). As the impurity concentration is raised to

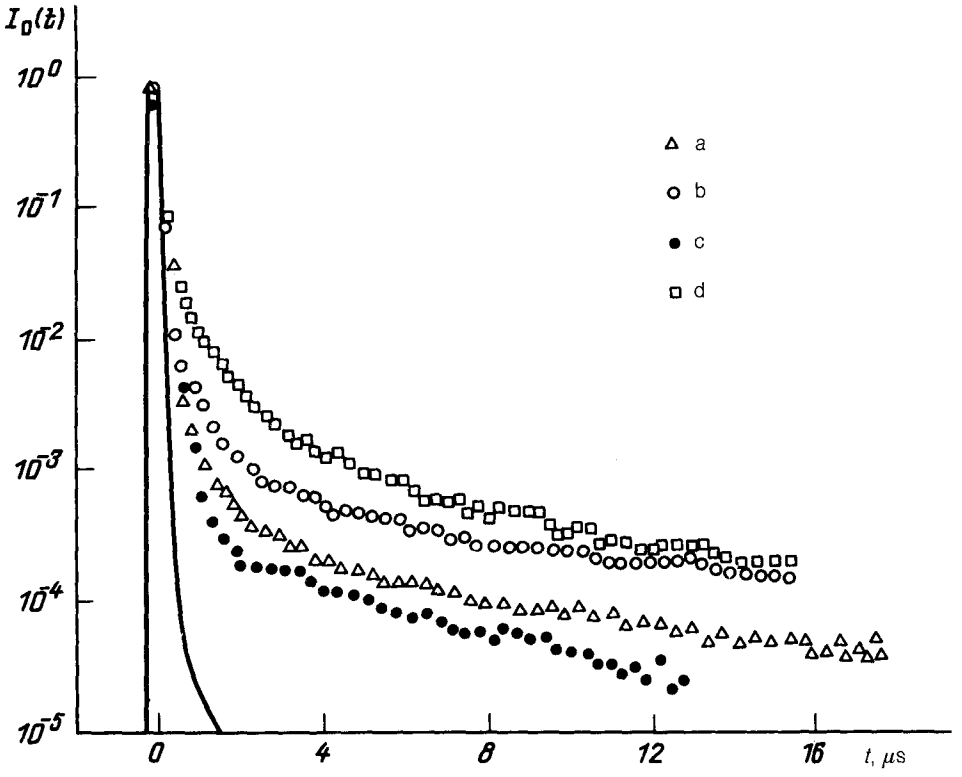

FIG. 2. Kinetics of the photoluminescence, $I_0(t)$, of the D_0^h line. These curves have been normalized. The points represented by different symbols correspond to different spectra in Fig. 1.

$n_D - n_A \sim 10^{16}$ cm^{-3}, the exciton structural features become diffuse, and we see only an inhomogeneously broadened band, which apparently corresponds primarily to the D_0^h transition (Fig. 1d).

Figure 2 shows decay curves $I_0(t)$ of the D_0^h luminescence of these samples. Here the excitation was carried out by pulses (2×10^{-7} s long) of the second harmonic from a Nd:YAG laser ($\lambda = 530$ nm) with pulsed pumping, $P < 10^3$ W/cm^2. The luminescence of all the samples reveals a fast decay, followed by a slow tail, with a relative amplitude which varies from sample to sample. The decay time of this tail, determined from the $I_0(t)$ slope at $t > 5$ μs, is $\tau_1 = 6.5 \pm 0.5$ μs and varies only slightly from sample to sample. The observed time of the fast decay corresponds to the instrumental time, and the actual time of this decay evidently corresponds to the lifetime of photoexcited carriers, τ_0 (in pure GaAs at liquid-helium temperature, we would have[1] $\tau_0 \sim 10^{-8}$ s). We see that the relation $\tau_1 \gg \tau_0$ holds.

The dashed lines in Fig. 1, a and d, show luminescence spectra measured with a time delay $t = 1$ μs in a window $\Delta t = 0.25$ μs. The lines D_0^h and D_0^x persist in the spectrum for a few microseconds after the termination of the optical excitation, but the

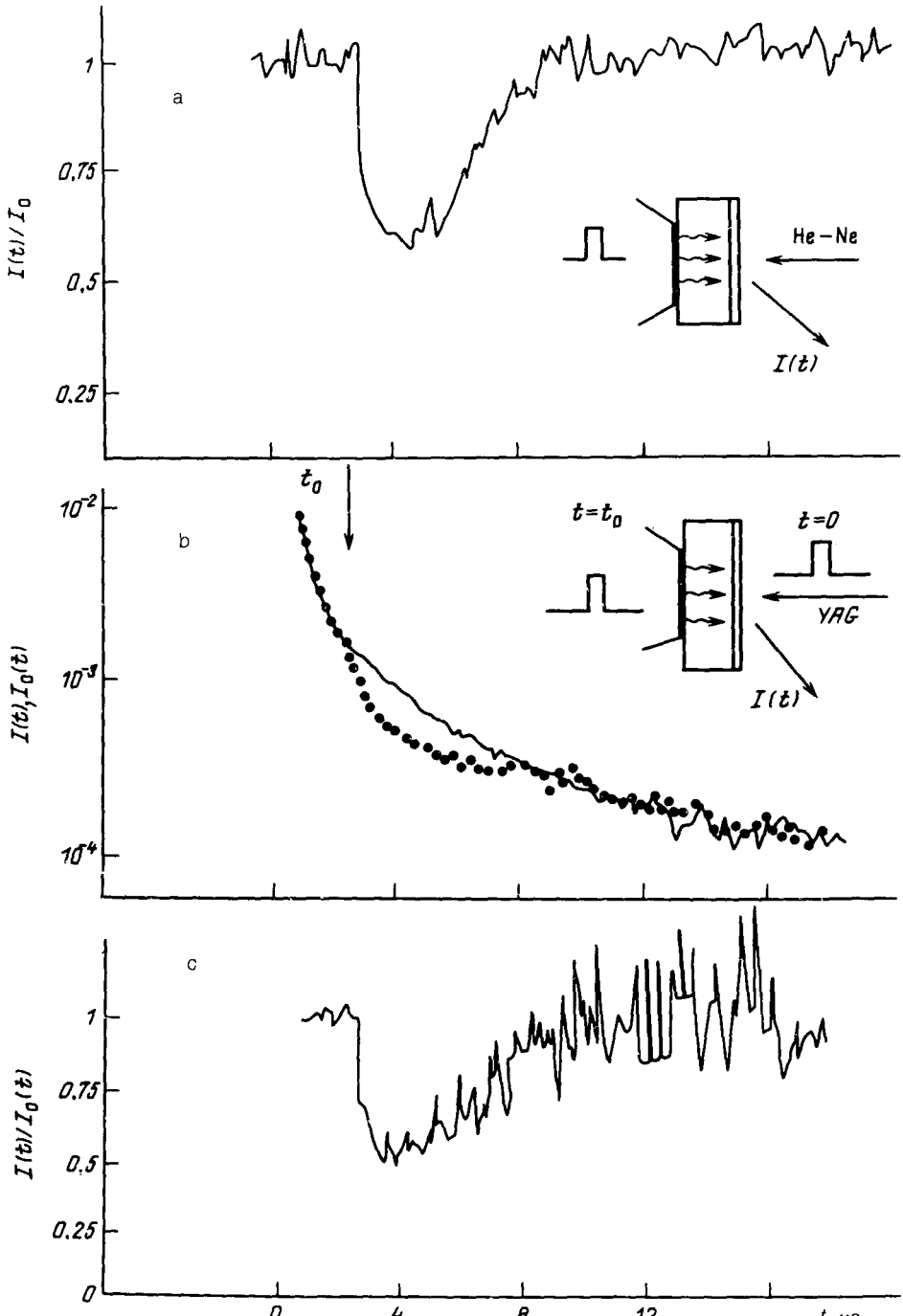

FIG. 3. Effect of heat pulses on the intensity of the D_0^n line. a—The dependence $I(t)/I_0$ during steady-state excitation; b—kinetics of the luminescence decay in the absence [$I_0(t)$; the solid lines] and in the presence [$I(t)$; the points] of heat pulses; c—the time dependence $I(t)/I_0(t)$ during pulsed excitation. The insets in parts a and b show the experimental layout.

emission associated with the recombination of free excitons and with the A_0^x transition is missing (Fig. 1c).

Let us discuss the results. At $T_0 = 1.7$ K n-GaAs always has neutral donors D_0 at the initial time. Accordingly, the existence of a (free hole)-(neutral donor) (D_0^h) luminescence a few microseconds after the end of the excitation is evidence of a slow decrease in the concentration of free holes h in the valence band. Consequently, if the fast decay of the luminescence, D_0^h (Fig. 2 and Ref. 1), reflects the lifetime (τ_0) of free holes in the valence band, then the microsecond tail $I_0(t)$ is due to a slow replenishment of free holes from certain long-lived localized states, where some of the holes are captured.[1)]

To determine the nature of the long-lived localized hole states, it turned out to be important to carry out experiments on the effect of heat pulses on the D_0^h luminescence line. A constantan film 1.5×1 mm^2 in area was deposited on the opposite side of the Si-GaAs substrate. This film was heated with current pulses (10^{-7} s long; the pulsed power dissipation was 50 W). Acoustic phonons were injected into the Si-GaAs substrate. These phonons reached the n-GaAs epitaxial layer by diffusion[4] and raised its temperature to $T \approx 6$ K.

Figure 3a shows the effect of the heat pulses on the D_0^h intensity during steady-state excitation of the luminescence. Here most ($> 90\%$) of the free holes are formed directly—without the involvement of the localized states. During the application of the heat pulses we observed a decrease in the D_0^h intensity (Fig. 3a), in agreement with the temperature quenching of D_0^h, which has been established elsewhere.[2] The shape of the "negative" luminescence pulse, $I(t)/I_0$ (I_0 is the steady-state D_0^h intensity in the absence of the heat pulses), reflects the kinetics of the "temperature" of the phonon pulse which has been transmitted through the Si-GaAs substrate.[4] In some subsequent experiments, involving pulsed optical excitation of the luminescence, the heat pulses affected the time evolution of the luminescence, $I_0(t)$ (Fig. 2). The heat pulses were applied 2 μs after the end of the excitation, at a time at which the fast luminescence decay, $I_0(t)$, had already terminated, and we were left with only the luminescence tail due to the replenishment of free holes from the localized states. In the time evolution of the luminescence, $I(t)$, observed in the presence of the heat pulses (the points in Fig. 3b) we can see a region in which there is a quenching of this luminescence. After the application of the heat pulses, $I(t)$ returns to the previous time evolution $I_0(t)$.

Figure 3c shows the time evolution of the ratio $I(t)/I_0(t)$ found from measurements of the $I(t)$ and $I_0(t)$ curves (Fig. 3b). We see that the curves in Fig. 3, a and c, are essentially identical. Consequently, the heat pulses have identical effects on the steady-state luminescence T_0 during continuous optical generation of free holes (Fig. 3a) and on the luminescence tail $I_0(t)$ under conditions such that there is no optical excitation of holes, and the luminescence results from a replenishment of free holes from localized states. It follows that a more than threefold increase in the temperature (from 1.7 K to 6 K) absolutely does not accelerate the liberation of holes from localized states into the valence band. In other words, the latter process does not require thermal energy and is a spontaneous process. This conclusion rules out the possibility

that the energy level that localizes the hole is in the band gap of GaAs, and it gives us every reason to believe that this level lies below the top of the valence band, i.e., is a metastable level.

The essentially unchanged behavior $I_0(t)$ at $t > 2$ μs (Fig. 2) in the epitaxial samples synthesized by the various techniques, with different degrees of compensation and different random fields due to defects (all these differences are manifested spectroscopically in different inhomogeneous widths of the exciton luminescence lines; Fig. 1), is evidence against a link between the metastable localized states and a localization of holes at fluctuations of the large-scale potential. In principle, one possible cause of localization might be an impurity or intrinsic self-localization of holes at a level below the top of the valence band (metastable self-localized state[5]). In this case, holes are liberated from the localized states by means of a tunneling of the holes through the potential barrier which separates the localized and free states of holes in configuration space.

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[1)] The observation of a slow kinetics in the D_0^x exciton line can be explained in terms of contribution to the emission in this line of processes involving a recombination of free holes liberated from localized states with D^- centers, formed in the optically excited GaAs during the capture of electrons by neutral donors D_0 (Ref. 3). The absence of free excitons and A_0^x from the luminescence spectrum at time $t > 1$ μs is evidence that the existence of the optically excited free electrons in n-GaAs is brief.

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