# Kinetics of exciton trapping by shallow impurity centers

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The binding of excitons to a shallow impurity center in a semiconductor (Si:B) has been studied, and the trapping coefficient has been determined ($8\times10^{-8}$ cm$^3$/s), for the first time. A study has been made of the kinetics of the perturbation caused in the steady-state spatial distribution of excitons in a sample by nonequilibrium acoustic phonons.

The processes by which charge carriers are trapped by impurity centers in semiconductors have now been studied quite thoroughly.[1] At low temperatures, however, most of the carriers are bound in excitons, as we know, and the excitons largely determine the nature of the transport and recombination effects in crystals. In this letter we are reporting direct measurements of the probability for the binding of excitons to a shallow impurity center. The experiments take the following approach. The spatial distribution of free excitons set up by a steady-state optical pump is subjected to a small pulsed perturbation which "deforms" the initial spatial profile. The parameters of this perturbation must be chosen in such a way that they do not cause an ionization of exciton-impurity complexes. An additional number $[\delta n(x,t)]$ of excitons, above the steady-state number $\bar{n}(x)$, appear at a certain point $x$. This additional number undergoes a relaxation to the steady-state value $\bar{n}(x)$ with a time constant $\tau^{-1} = \sigma v_T\ (N_0 - N_{\text{in}})$, where $\sigma$ is the cross section for the trapping of an exciton by an impurity center, $v_T$ is the thermal velocity of the excitons, $N_0$ is the impurity concentration, and $N_{in}$ is the concentration of exciton-impurity complexes, which depends on the optical pump. By arranging the condition $N_{\text{in}} \ll N_0$ and measuring $\tau$, one can determine the coefficient for the trapping of an exciton by an impurity center.

A study was made of Si:B single-crystal samples with concentrations $N_0 \sim 10^{13}$–$10^{14}$ cm$^{-3}$ and typical dimensions $2\times5\times10$ mm and $5\times5\times15$ mm with the orientations $x\|(110)$ and $x\|(111)$ ($x$ is the length of the parallelepiped). Nonequilibrium electron-hole pairs were generated by the beam from a He–Ne or Ar* laser in a spot $d\sim50$ $\mu$m in diameter with a pump density below the threshold for the formation of droplets of an electron-hole liquid. The experiments were carried out at $T = 1.5$ K in superfluid He$^4$. The experimental geometry is shown in the inset in Fig. 1. As the perturbation top create the desired change in the coordinate part of the exciton distribution function we used a flux of nonequilibrium acoustic phonons, which were generated in pulses in an energy band $\Delta(\hbar\Omega_{ph}) \sim 0$–$3$ meV with the help of a constant thermal generator. This generator had dimensions of $0.1\times1$ mm and was placed on a smaller face of the crystal. The nominal power applied to this phonon generator was $P = 25$ W.

  

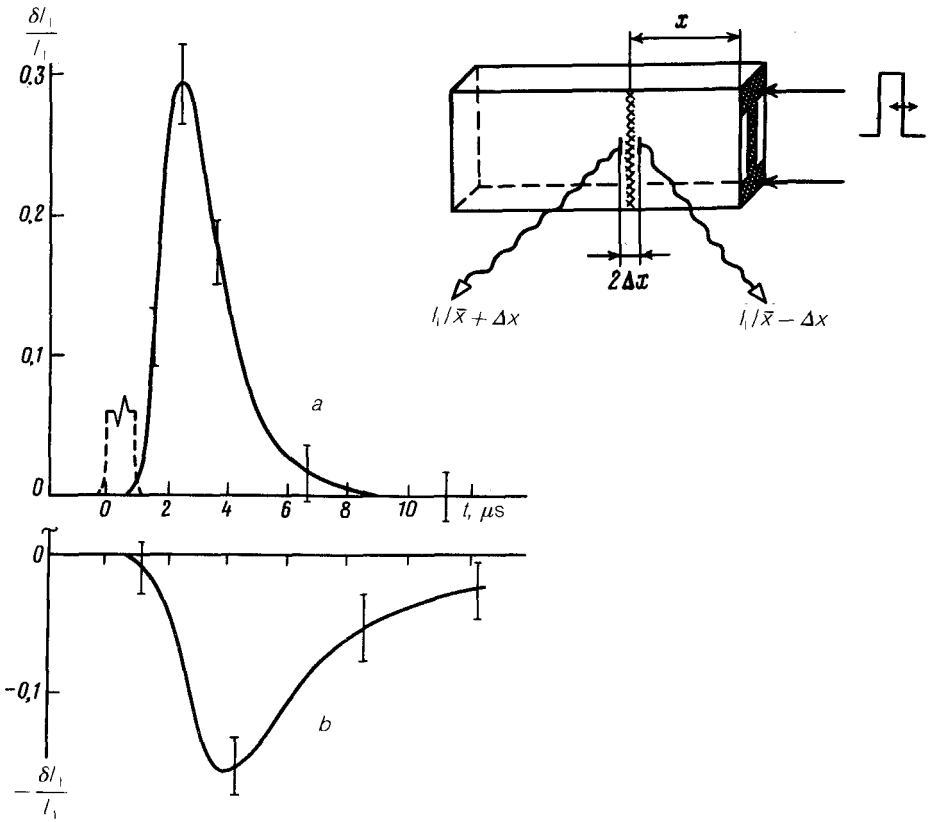


FIG. 1. Temporal kinetics of the exciton luminescence. *a*—In the region $x = \bar{x} + \Delta x$; *b*—in the region $x = \bar{x} - \Delta x$ ($\bar{x} = 5$ mm, $\Delta x = 100$ $\mu$m). The inset shows the experimental geometry [$x \| (110)$]. The dashed line shows the heat pulse.

Nonequilibrium phonons cause a transport of excitons,[2] increasing the concentration of free excitons in the region $\bar{x} + \Delta x$ (Fig. 1; $\bar{x}$ is the coordinate of the center of the region in which there is a steady-state pumping of nonequilibrium electron-hole pairs, reckoned from the face at which the phonon generator is positioned). The coordinate distributions of excitons and of exciton-impurity complexes were measured by scanning the image of a spectrometer slit, which singled out the optical signal at the corresponding wavelength along the propagation direction of the flux of nonequilibrium phonons with a spatial resolution of 50–100 $\mu$m under our conditions. If there is a trapping of excitons, the excess concentration (at the points $\bar{x} + \Delta x$) should relax to the steady-state value over a time $\tau$, as can be shown. In this case, the temporal profile of the response at the point $\bar{x} + \Delta x$ should not change upon a change in the duration of the phonon pulse (Fig. 2).

Indeed, the decay rate of the differential signal in a system of luminescing centers should have been determined by the rate of decay of the thermal pulse. However, a

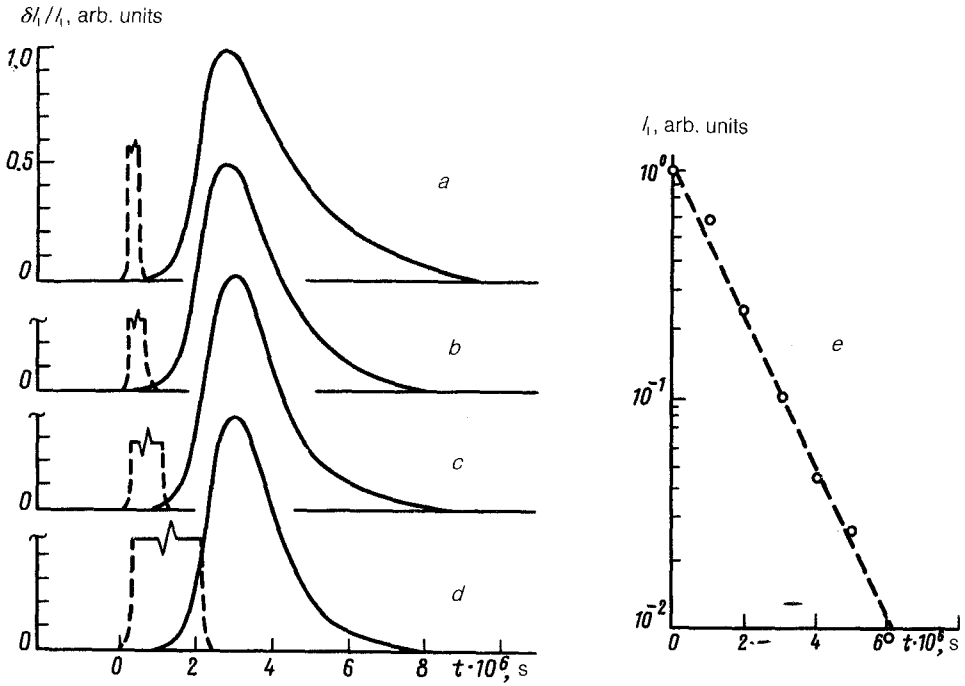


FIG. 2. *a–d*—Shape of the temporal kinetics of the exciton luminescence for various durations of the pulse applied to the phonon generator; *e*—time evolution of the luminescence intensity during the decay. The dashed line show the heat pulse.

system of free excitons is not a closed system; instead, there is a "drainage" to states of exciton-impurity complexes. Under these conditions, an increase in the luminescence intensity resulting from the arrival of excitons entrained by a heat pulse in the detection region reflects a disruption of the dynamic equilibrium of the system consisting of the excitons and the exciton-impurity complexes. This disruption is healed in a time $\tau$. The same process occurs if the duration of the heat pulse exceeds $\tau$. In contrast with the case of a short heat pulse, however, a new quasisteady exciton/(exciton-impurity-complex) ratio is established after some transients. The rate of relaxation of this quasisteady ratio is determined in turn by the kinetics of the decay of the heat pulse (we observed this picture at heat pulse durations greater than 3 $\mu$s).

On the other hand, the signal at the point $\bar{x} - \Delta x$ tracks the duration of the heat pulse, since the binding processes do not determine the shape of the temporal kinetics in this spatial region (Fig. 1). (The scale of the temporal kinetics of the luminescence on a line of exciton-impurity complexes in the region $\bar{x} + \Delta x$ is roughly the same as that for excitons in the region $x = \bar{x} - \Delta x$.)

Let us examine the coordinate distributions of the excitons and the complexes (Fig. 3) as measured 4 $\mu$s after the heat pulse, at a time at which most of the free excitons have been trapped by boron atoms. The primary feature is obvious: a change

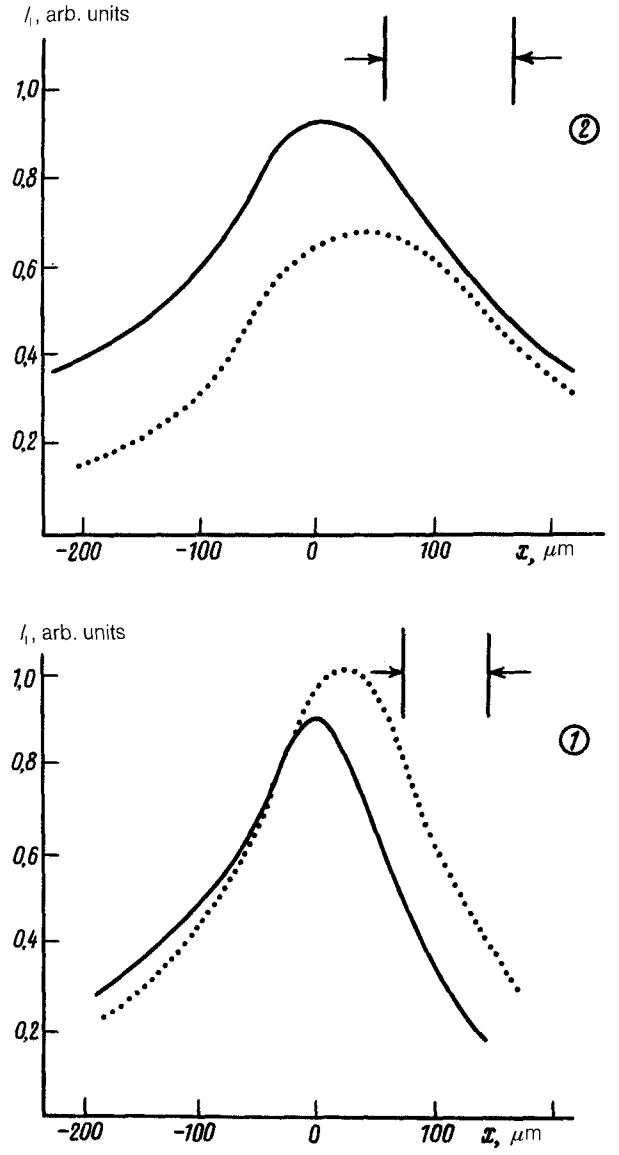


FIG. 3. Spatial profiles of (1) excitons and (2) exciton-impurity complexes along the $z$ axis measured at a time $t = 4$ $\mu$s after the heat pulse. Solid lines—Distributions in the absence of nonequilibrium phonons; dotted lines—distributions in the presence of nonequilibrium phonons.

in the distribution of the complexes, expressed as a shift of the spatial profile into the region $x > \bar{x}$. All the experimental data reported above indicate that the trapping of excitons to a state of an exciton-impurity complex plays a dominant role. This state determines the phonon-induced kinetics of the response of free excitons. From Figs. 1

and 2 we found the value of $\tau$ for this trapping process to be $\tau \simeq (1.3 \pm 0.1) \times 10^{-8}$ s. In this particular sample ($N_0 \simeq 10^{13}$ cm$^{-3}$), this value corresponds to a binding coefficient $\gamma = \sigma v_T \simeq 8 \times 10^{-8}$ cm$^3$/s. Since there is no rigorous theory for excitons, one can derive a theoretical estimate of the binding coefficient $\gamma$ by working from the expression from Ref. 1 for the trapping of a hole by a neutral acceptor, in a process resulting in the formation of an $A^+$ center (which is an analog of the hydrogen ion $H^+$):

$$\gamma = \frac{\pi^3 B^2}{16 l_0} \left(\frac{2\epsilon_0}{m^*}\right)^{1/2} \left(\frac{\hbar}{m^* s}\right)^3 \psi[\text{cm}^3/\text{s}], \tag{1}$$

where

$$\psi = \frac{4}{\pi^2}\left(\arctan\frac{1}{x} - \frac{x}{x^2+1}\right), \quad x = \frac{(2m^* s^2 \epsilon_0)^{1/2}}{\epsilon_0 + \epsilon_k}$$

$\epsilon_k$ is the kinetic energy of the particle being trapped, $s$ is the velocity of sound, $\epsilon_0$ is the binding energy of the complex, $B = 1.1$, and $l_0$ is the energy-loss length.[3]

Substituting the properties of exciton-impurity complexes in Si:B into (1), and assuming $\epsilon_k \to 0$, we find the estimate $\gamma_{\text{theo}} \sim 10^{-7}$ cm$^3$/s, which agrees satisfactorily with the experimental value. The fact that the binding energies of an exciton-impurity complex and an $A^+$ center are not greatly different (3.8 and 2 meV, respectively) demonstrates the validity of using a model based on expression (1) to find an order-of-magnitude estimate of $\gamma$.

The new approach to a study of trapping processes (or radiationless recombination) developed in this study has a wide range of applications and can be used to study exciton trapping not only by shallow impurity centers but also by deep impurity centers. In a subsequent and more detailed paper we will report results on other shallow impurity centers in Si.

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