

Dispersion of lifetimes of localized excitons in the solid solution $\text{CdS}_{1-x}\text{Se}_x$

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(Submitted 30 May 1983)

Pis'ma Zh. Eksp. Teor. Fiz. **38**, No. 1, 22–25 (10 July 1983)

It is established that the lifetimes of localized excitons in the system $\text{CdS}_{1-x}\text{Se}_x$ depend strongly on the localization depth. This dependence reflects the dispersion or radiative times for shallow localized states.

PACS numbers: 71.35. + z, 71.50. + t

Under optical excitation the photoluminescence (PL) spectra of semiconducting solid solutions $\text{CdS}_{1-x}\text{Se}_x$ at helium temperatures contain in the region of intrinsic absorption an intense band situated adjacent to the exciton reflection lines, as well as its LO-phonon sideband. It was shown in Refs. 1–3 that this radiation is due to localized excitons. The excitons are localized in the potential wells of the relief formed by statistical fluctuations of the composition of the solid solution.⁴ In accordance with the general representations of the theory of disordered systems, the localized and delocalized states, which are separated by the mobility threshold, should coexist in the exciton spectrum of semiconducting solid solutions. For $\text{CdS}_{1-x}\text{Se}_x$ solutions, the position of this threshold in the energy spectrum was determined experimentally⁵ from polarization excitation spectra.

The results obtained in Refs. 1–3 and 5 show that the main contribution to radiative recombination in $\text{CdS}_{1-x}\text{Se}_x$ comes from the localized exciton state situated near the mobility threshold. Delocalized states have a short lifetime which is limited by rapid localization in very deep potential wells. Spatial localization of excitons greatly decreases the probability of nonradiative annihilation. This circumstance, as well as the absence of Auger recombination processes for localized excitons, leads to the fact that at the lowest temperatures, their lifetime can be determined by radiative processes. At the same time, the “giant” oscillator strength of shallow localized states⁶ can lead to lifetimes of the order of 10^{-9} s. In addition, a considerable increase should be observed in the lifetime with increasing localization energy ϵ , which along the initial section is described by the law $\tau \sim \epsilon^{3/2}$.⁶

We have performed the first investigation of the dependence of the lifetime of localized excitons on the localization energy ϵ . The lifetime was measured from the damping of the spectrally decomposed PL. A pulsed nitrogen laser was used to excite PL. The maximum excitation density did not exceed $10^{22} \text{ cm}^{-2} \text{ s}^{-1}$ and, at the same time, the form of the PL spectrum was independent of the excitation intensity. The procedure used to perform the measurements with time resolution is described in Ref. 7.

Figure 1 shows the PL spectra of the specimen $\text{CdS}_{0.88}\text{Se}_{0.12}$ recorded at different times. Curve 1 was obtained 5 ns before the maximum of the excitation pulse; curve 2

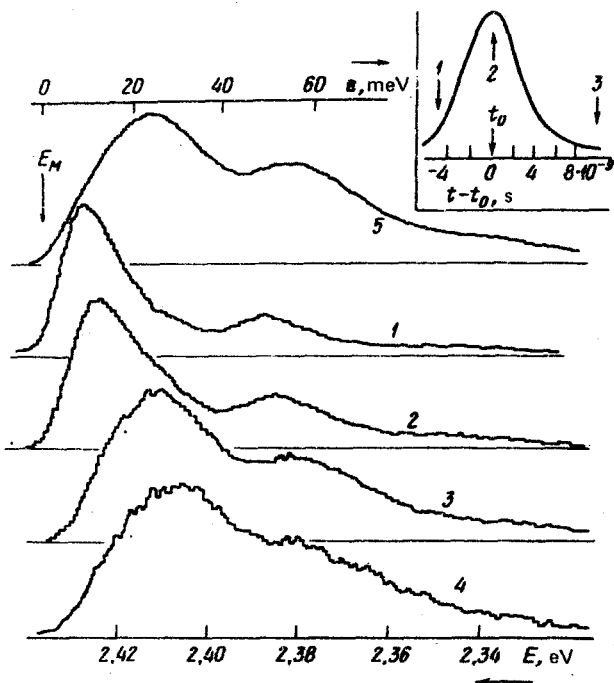


FIG. 1. PL spectra of the solid solution $\text{Cd}_{0.88}\text{Se}_{0.12}$ at different times $(t - t_0)$ with pulsed excitation (curves 1-4) and continuous excitation (curve 5). $T = 2$ K. The insert shows the excitation pulse shape.

was obtained at the pulse maximum; curves 3 and 4 were obtained 10 and 20 ns after the excitation maximum; and, curve 5 is the PL spectrum with continuous excitation. As is evident from the figure, with time the maximum of the shortest wavelength (phononless) emission band is appreciably shifted toward lower energies. Analogous results were also obtained for specimens with other compositions.

The observed shift could reflect migration of energy between separate localized states. However, since this shift occurs in the region of energies much lower than the mobility threshold (the arrow E_M in Fig. 1; the position is determined from the results in Ref. 5), it is more natural to interpret the observed temporal evolution of the emission spectra as a manifestation of the dependence of the radiative lifetime of excitons on their localized depth $\epsilon = E_M - E$.

Investigation of the PL decay curves at a fixed frequency (Fig. 2) showed that states with different localization energies can be described by different decay times τ . The different types of points in Fig. 2 show experimental results for different values of ϵ ; the dashed curve is the experimentally measured pulse shape $I_0(t)$ of the exciting laser; the continuous curves $I(t)$ show the result of convoluting the form of the laser pulse with the exponential function $\exp(-t/\tau)$ according to the equation

$$I(t) = \int_0^t I_0(t') \exp[-(t - t')/\tau] dt'. \quad (1)$$

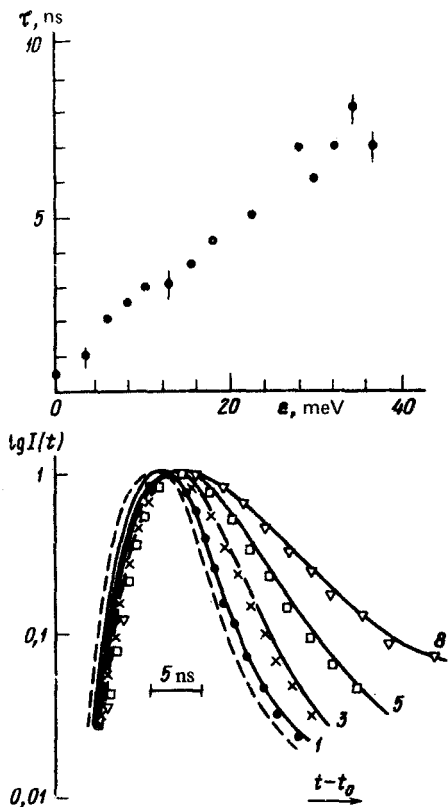


FIG. 2. PL pulse shapes for localized excitons with different localization energies (experimental points). The solid curves show the calculations of $I(t)$ using Eq. (1); the dashed curve is the laser pulse $I_0(t)$. The measured dependence $\tau(\epsilon)$ is shown at the top. The results are for a specimen with $x = 0.12$.

The numbers near each curve indicate the value of τ (in ns) used to obtain the convolution. The upper part of the figure shows the dependence $\tau(\epsilon)$ obtained. It is evident from the figure that the exponential decay that we assumed describes well the form of the decreasing sections of PL pulses. It turned out that within the scope of this assumption, the quantity τ can be determined to within ± 0.5 ns. The difference between the computed curves and experimental points in the region of the leading edge of the pulses indicates that the localized exciton states are not populated instantaneously, but over a time of the order of 1–3 ns.

We assume that the observed lifetimes of localized excitons are determined primarily by radiative recombination. Tunnelling transitions to deeper states are the main nonradiative process for localized excitons at helium temperatures. However, investigations of the form of PL spectra with selective excitation² shows that such transitions can affect the lifetime only for very shallow states in the region $\epsilon < 5$ –10 meV. The values of the lifetimes that we obtained are close to the radiative times of exciton-impurity complexes in GCS crystals, measured by a phasometric method in PL spec-

tra⁸ and estimated from the absorption spectra.⁹ The dependence of the lifetime τ on the localization energy ϵ , on the whole, agrees with the results obtained in Ref. 6 for radiative times of shallow, localized states.

It should be noted that the dependence $\tau(\epsilon)$ that we measured has some systematic uncertainty. As shown in Ref. 2, for nonselective excitation, states with different localization energies contribute to radiation at a fixed frequency, so that each energy in the PL spectrum corresponds to decay times τ averaged over some interval ϵ . A more accurate determination of the dependence $\tau(\epsilon)$ in experiments with selective resonant excitation of PL can give, based on the theory in Ref. 6, information on the relation between the localization energy and the dimensions of the localization region, which is very important for studying the nature of localized exciton states in semiconducting solid solutions.

In conclusion, I thank A. A. Kaplyanskiĭ and É. I. Rashba for discussions.

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Translated by M. E. Alferieff

Edited by S. J. Amoretty