

# Spectral manifestation of the exciton mobility boundary in $\text{CdS}_{1-x}\text{Se}_x$

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The polarization of the luminescence of excitons localized by fluctuations of the component concentrations in the solid solution  $\text{CdS}_{1-x}\text{Se}_x$  is found to depend on the frequency of the exciting light. A study of this dependence has permitted detection of the exciton mobility boundary and determination of its position in the energy spectrum of this solid solution.

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Baranovskii and Éfros<sup>1</sup> have proposed a model for the localization of excitons in the potential wells formed by fluctuations of the composition in semiconductor solid solutions. Studies done in recent years have shown that the main contribution to the radiative recombination of the solid solution  $\text{CdS}_{1-x}\text{Se}_x$  at liquid-helium temperatures is due to localized-exciton states.<sup>2,3</sup> It is well known that the luminescence spectrum of a semiconductor is determined largely by the kinetic properties of the excitons, particularly by their capability to move through the crystal. We therefore hoped that by studying the luminescence spectra we would be able to detect the boundary dividing the localized and delocalized exciton states.

It has been shown<sup>2,4</sup> that during resonant excitation by linearly polarized light in the region of the localized states, the luminescence has a high degree of linear polarization. In this study we have investigated how the degree of polarization of the luminescence of localized excitons depends on the frequency of the exciting light. The samples were placed directly into pumped-off liquid helium and excited by the polarized beam of either an  $\text{Ar}^+$  laser or a coumarin-152 dye laser. We studied the emission of cleavage surfaces or growth faces perpendicular to the optic axis of the hexagonal crystals  $\text{CdS}_{1-x}\text{Se}_x$ . The degree of polarization of the emission was defined as

$$\rho = (I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp}),$$

where  $I_{\parallel}$  and  $I_{\perp}$  are the intensities of the luminescence with the polarization parallel and perpendicular, respectively, to the polarization of the exciting light. Two methods were used to measure the degree of polarization  $\rho$ . One of these methods involved a systematic recording of the spectra of  $I_{\parallel}$  and  $I_{\perp}$  with an OMA-2 optical multichannel analyzer and a subsequent digital processing of the results with the aid of a microprocessor. The second method was based on a simultaneous recording of the spectra of  $I_{\parallel}$  and  $I_{\perp}$  with the aid of a two-channel system operating as a photon counter. In this case as well, the necessary processing of the spectra was done on a microcomputer.

In studying the polarization of the luminescence of the localized excitons, we observed that the degree of polarization of the emission depended on the frequency of the exciting light. The results of the measurement of this dependence for a sample with

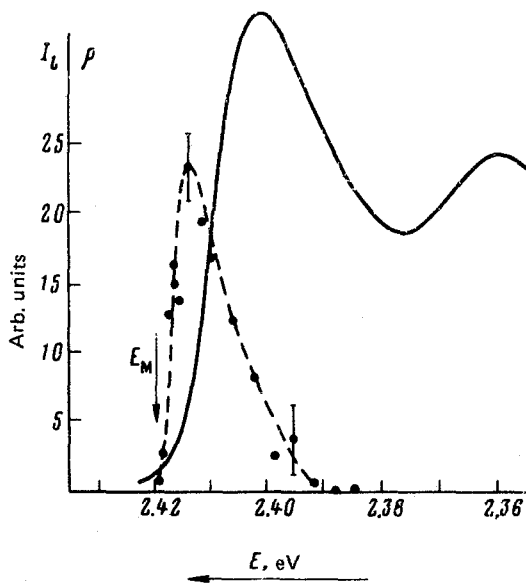


FIG. 1. Degree of polarization  $\rho$  of the localized-exciton emission versus the photon energy  $E$  of the exciting light (points). The solid curve is the luminescence spectrum.

selenium concentration  $x = 0.1$  are shown by the points in Fig. 1. The luminescence was unpolarized during excitation in the fundamental absorption region. A polarization appeared as the frequency of the exciting light was reduced to below a certain energy  $E_M$ , rose rapidly to a maximum in the region of the short-wavelength edge of the emission band, and then decreased to zero as the frequency of the exciting light was decreased further. Similar results were obtained for samples with other concentrations as well.

It has been shown<sup>4</sup> that the polarization of the luminescence is due to the anisotropy and random orientation of the potential wells responsible for the localization of the exciton (the so-called "latent anisotropy"<sup>5</sup>). In this case a polarization will appear in the spectrum if the anisotropy is sufficient to split the  $\Gamma_5$  exciton level and there is no transfer of excitation between different wells.<sup>4</sup>

We attributed the sharp short-wavelength decay of the degree of polarization to the onset of mobility in the system of localized excitons, since the transfer of excitation from the originally excited level to a level in another well should cause a complete depolarization of the emission. The increase in the migration probability as the frequency of the exciting light is increased promotes the growth of the density of exciton states near the bottom of the unperturbed exciton band.<sup>1</sup> This growth should lead first of all to an increase in the overlap of the wave function of the localized excitons and then, at high enough energies, to the formation of delocalized states. The short-wavelength boundary of the polarization excitation spectrum corresponds precisely to those exciton energy states from which the probability of excitation transfer to the deeper wells during the lifetime is essentially 100%. We assume that this boundary can be

regarded as the exciton mobility threshold in the solid solution  $\text{CdS}_{1-x}\text{Se}_x$ .

It is seen in Fig. 1 that the mobility threshold coincides in energy with the beginning of the sharp short-wavelength rise in the intensity of the luminescence produced upon excitation to the fundamental-absorption depth (the solid curve in Fig. 1). From this we can conclude that the main contribution to the radiative recombination in the solid solution  $\text{CdS}_{1-x}\text{Se}_x$  at low temperatures is that of the localized states. Excitons in states with energies higher than the mobility boundary have very short lifetimes because of the rapid localization and contribute virtually nothing to the emission.

The decrease in the degree of polarization in the long-wavelength part of the polarization excitation spectrum is obviously not due to the migration of excitons. It is possibly due to a decrease in the anisotropy of the potential wells as the localization depth increases. At least two explanations for this effect can be offered at the present time.

We suggest that the anisotropy arises as a result of interaction of the wells which are closely situated in space. As the localization energy increases, the decrease in the size of the potential wells and the increase in their separation cause a decrease in the probability of interactions between wells and, consequently, a growth of the contribution of the isolated, nearly isotropic wells.

The second possibility stems from the fact that as the localization depth increases in the deep potential wells of small radius, only the hole is localized.<sup>6</sup> In this case the orbital radius of the electron bound to the hole by the Coulomb interaction will be significantly larger than the dimension of the fluctuation, and the motion of the electron will average out the anisotropy.

In either case one would expect the splitting of the exciton level  $\Gamma_5$  in the anisotropic field to decrease to less than the homogeneous width, causing the polarization of the emission to vanish.

We believe that the relationship between the degree of anisotropy of the localized-exciton states and their localization depth warrants careful theoretical consideration. The paper by Dushmansev and Rashba<sup>7</sup> is of interest in this regard. Those authors showed that in the case of a degenerate valence band the tails on the density of states are due to strongly anisotropic fluctuations. It is not clear, however, to what extent the results of that paper can be applied to hexagonal solid solutions, in which the valence band is split by the crystal field.

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