Effect of pumping intensity on migration of excitations of localized excitons in solid solutions of semiconductors

L. G. Suslina, A. G. Areshkin, V. G. Melekhin, and D. L. Fedorov
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR

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It is found that the luminescence peak of localized excitons in the spectrum of the solid solutions $Zn_xCd_{1-x}S$ is displaced as a function of the pumping intensity. This phenomenon is interpreted as the spectroscopic manifestation of migration of electronic excitations.

Concentration fluctuations in solid solutions of A_2B_6 lead to the formation of potential wells capable of trapping excitons. An inhomogeneous contour of localized exciton states (LES) appears in the spectrum in the form of additional long-wavelength broadening of the exciton absorption line. The short-wavelength part of this line is broadened homogeneously due to the breakdown of momentum conservation for delocalized exciton states.[1–5]

Localized exciton states appear in the emission spectrum in the form of a separate luminescence line (I_L). The I_L line in the spectrum of $Zn_xCd_{1-x}S$ crystals[4] with $0 < x < 0.15$ is situated between the resonant frequency $n = 1$ A of the exciton and the luminescence line of the exciton complex I_2 (exciton bound to a neutral donor) (see Fig. 1). The main characteristics of the I_L line are 1) the Stokes shift relative to the absorption maximum $n = 1$ A and its dependence on the concentration x and 2) the smooth short-wavelength displacement with increasing temperature. These characteristics are the spectroscopic manifestation of migration of excitations of LES.[4]

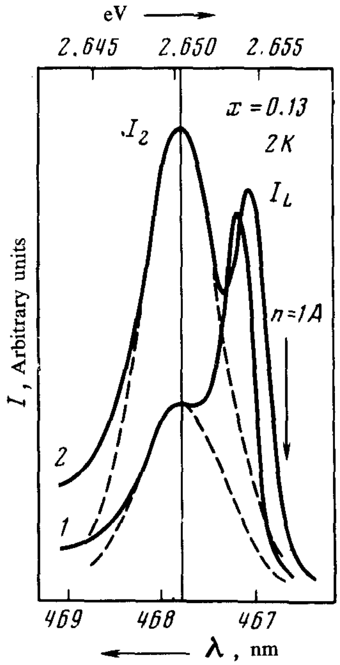

FIG. 1. Luminescence spectra of $Zn_{0.13}Cd_{0.87}S$(T = 2 K) crystals, obtained with different intensities of the exciting light J_{exc}, J_{exc} for spectrum 2 is 2.6 orders of magnitude higher than for spectrum 1. The contours I_L and I_2 were separated, taking advantage of the fact that I_2 has a symmetrical Gaussian contour (dashed curve).

In this paper we present the results of an investigation of a new nonlinear phenomenon in the optics of SSS, arising due to diffusion of excitations of LES: dependence of the luminescence spectrum of LES on the excitation intensity J_{exc}. This displacement of the I_L peak as a function of J_{exc} is related to the dispersion of the radiative lifetimes[6] and density of states[2,5] for the inhomogeneous radiation contour of LES when migration of excitations is taken into account.

The luminescence spectra of $Zn_xCd_{1-x}S$ crystals with concentrations $0<x<0.15$ at $T=2$ K were excited with the help of an He–Cd laser (λ_{exc} = 441.6 nm) with a peak power of 10 mW with the laser beam focused into a spot with a diameter of 0.1–0.15 mm. The maximum pumping intensity (photon flux per 1 cm^2 s^{-1}) amounted to $f=3\times10^{20}$ photons·cm^{-2}·s^{-1}. The intensity of the exciting light was changed by using calibrated neutral filters.

It was observed that the intensity of the exciting light J_{exc} has a significant effect on the luminescence spectrum of $Zn_xCd_{1-x}S$ crystals.[1)] An increase in J_{exc} leads to the following results: 1) shift of the I_L line to the short-wavelength side of the spectrum toward the exciton resonance line $n=1$ A (Fig. 2) and 2) redistribution of the radiation intensity between two lines I_2 and I_L: for small J_{exc}, the I_L line is dominant in the spectrum (which permits analyzing its contour and half-width); for large J_{exc}, the luminescence intensity changes in favor of I_2 (Fig. 1), which indicates the different

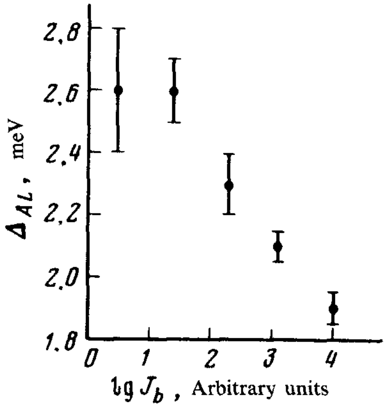

FIG. 2. Dependence of the line shift of LES radiation I_L in the spectrum of crystals $Zn_{0.13}Cd_{0.87}S$ on the exciting light intensity J_{exc}, $T = 2$ K. Δ_{AL} is the distance (in meV) from the exciton resonance, $n = 1$ A.

dependence of the intensity of the I_2 and I_L lines on J_{exc}. The investigations showed that the dependence of the integral intensity I_2 on J_{exc} is linear (or weakly superlinear). However, the dependence of the I_L line on J_{exc} is always weaker than for the I_2 line, it is close to being sublinear. Both experimental results (1 and 2) are the spectroscopic manifestation of transfer of the energy of LES in SSS.

The observed shift in the I_L line as a function of pumping intensity must be viewed as a nonlinear process, which may appear under sufficiently high pumping intensity, when the total number of states N, between which migration occurs, does not differ greatly from the number of states n excited by the light. It is important to take the nonlinearity into account (i.e., terms proportional to the pumping parameter $l = n/N$) when examining a system with inhomogeneous broadening[2)] at high pumping intensities, as well as low temperature in the case of rapid migration.[9]

Estimates show that the experimental situation for excitation of LES with large J_{exc} in the SSS system $Zn_xCd_{1-x}S$ corresponds closely to the nonlinear regime, when the number N of possible LES, determined by their density of states, becomes comparable to the number n of exciton excitations created by laser pumping. The tail of the density of LES is defined as[5]

$$\rho(\epsilon) = 0.1(M/\hbar^2)^{3/2}\frac{|\epsilon|^{3/2}}{E_0} \exp\{-(|\epsilon|/E_0)^{1/2}\}, \quad (1)$$

where $M = m_e + m_h$ is the translational mass of the exciton, and E_0 is the characteristic energy[3)] which describes the drop in the function $\rho(\epsilon)$ in the SSS in the region of negative energies ϵ. Assuming that $M = 1.3m_0$, for SSS with $x = 0.13$, it is possible to estimate $\rho(\epsilon)$ (number of states per meV per unit volume) in the region of the inhomogeneous contour of LES[4,5] with $|\epsilon| = 3$ meV:

$$\rho(\epsilon) \approx 10^{17}\,\text{meV}^{-1}\cdot\text{cm}^{-3} \quad (2)$$

The number N of possible LES in the energy range $\Delta\epsilon = 2$ meV, which correspond to the inhomogeneous contour I_L [4)], is

$$N=\int_{\epsilon}^{\epsilon+\Delta\epsilon}\rho(\epsilon)\,d\epsilon\,;\quad N\cong 2\cdot 10^{17}\ \text{states cm}^{-3}\,. \tag{3}$$

We shall now estimate the density of excitons created with laser excitation with a maximum $f=3\times10^{20}$ photons·cm^{-1}·s^{-1}, using the relation

$$n=f\alpha\tau\,, \tag{4}$$

where τ is the lifetime of the exciton, and α is the coefficient of absorption at the wavelength of the exciting light. Assuming that $\tau\cong10^{-9}$ s,[6] and $\alpha=10^5$ cm^{-1}, we obtain $n=3\times10^{16}$ particles·cm^{-3}. Comparing N and n, it is evident that they differ at maximum J_{exc} by not more than an order of magnitude (i.e., the parameter l is equal to 10^{-1}). This circumstance permits explaining the shift of the I_L peak with the excitation intensity in terms of filling of the tail of the density of LES as J_{exc} is increased. The nonlinear excitation regime is related to the limit on migration for large J_{exc}, since the process of excitation transfer (tunneling) from one potential well to another is inhibited by the presence of a large number of filled wells. The data in Fig. 2 show that for small J_{exc}, the I_L line is not displaced as a function of J_{exc}, indicating a transition to the linear excitation regime (as estimates show, in this case, $l\leqslant10^{-4}$). In the linear excitation regime, the number of vacant wells is large, LES migrate freely, and the shift of the $I_L(\Delta_{AL})$ line is maximum (Fig. 2).

In examining the redistribution of the intensity in the spectrum of radiation of SSS accompanying a change in J_{exc}, it is necessary to include the different nature of the I_2 and I_L lines.[4] Because of the dispersion of the radiative lifetimes of LES[6] over the inhomogeneous contour as a result of the shift in I_L with J_{exc}, the integrated quantum yield of luminescence of I_L changes, and this change affects the dependence of the intensity of I_L on J_{exc}. The theoretical analysis of the magnitude of the shift of the I_L line (and its intensity) as a function of J_{exc} will require an estimate of the parameters and kinetics of migration of excitations of LES in SSS.

The results of these investigations indicate the fundamental possibilities of using the SSS as systems with inhomogeneous spectral broadening, in which a large number of excitations, comparable to the number of wells capable of trapping excitons, can be created. The pumping intensity permits selecting separate LES according to the spectrum by varying the level of excitation and thus can be used as a method for studying the transfer of electronic energy in SSS.

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[1)]In these experiments, the change in the temperature of the specimen can be ignored, since the line I_2 remains undisplaced as J_{exc} is varied.

[2)]A study of the migration of excitations of ions of rare-earth elements in glasses showed[7,8] that the parameter $l=10^{-6}$, i.e., in this case the linear approximation is valid, and the emission spectrum is not displaced with J_{exc}.

[3)]The quantity E_0 was obtained from measurements of exciton reflection spectra.[3] Here $\Delta=14E_0$, where Δ is the half-width of the absorption line.

[4)]This estimate of N is generally approximate, since the dependence (1) is valid for $|\epsilon|\gg E_0$.

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