Effect of one-dimensional disorder on the exciton states in semiconductor solid solutions

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It is shown for the first time both experimentally and theoretically that the observed broadening of the exciton states in semiconductor solid solutions in the existence domain of the sphalerite-wurtzite phase transition can be described in terms of a one-dimensional-disorder model.

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Semiconductor solid solutions (SSS) are classical examples of disordered crystals in which fluctuations of the concentration give rise to a random potential causing smearing of the band edges and the appearance of a long-wavelength tail on the density of localized states.[1–4] The theoretical treatment of the effect of the fluctuational disorder on the exciton states in SSS is based on solution of the three-dimensional problem and is approximate in nature.[2] The one-dimensional problem has an exact solution, and in this case the exciton states are always localized along the direction of the disorder.[4,5] We noted that certain effects observed in real structures [upon disruption of the alternation sequences of close-packed layers in polytypes, in crystals with stacking faults,[6,7] and in SSS upon the mixing of two compounds with different crystals lattices (sphalerite and wurtzite)] can be described in terms of a one-dimensional-disorder model.

In this paper we report a first study, both experimental and theoretical, of the effect of one-dimensional disorder on the exciton states in semiconductor solid solutions, treating the particular case of the $Zn_{1-x}Mg_xS$ system in the existence domain of the sphalerite-wurtzite phase transition.[8]

The one-dimensional disorder in SSS, which is due to the existence of stacking faults, was studied in the $Zn_{1-x}Mg_xS$ system by two methods—birefringence and exciton spectroscopy. The birefringence Δn for crystals with stacking faults is a linear function of the degree of anisotropy α [$\alpha = \Delta n/\Delta n_0$, where Δn_0 is the birefringence in ZnS with the wurtzite structure W (Ref. 7)]: The changes in Δn for the $Zn_{1-x}Mg_xS$ system yield the values of α and thus enable one to track the one-dimensional disorder of these crystals. Studies of the low-temperature (77 and 2 K) reflection spectra revealed the presence of polarized exciton lines. From the polarizations and relative positions of these lines in the spectrum we were able to conclude that crystals with stacking faults exist in the region $0 < x < 0.12$ (Ref. 8). In the phase-transition region we detected a pronounced broadening of the dispersion curves of the reflection—evidence of an effect of one-dimensional disorder on the exciton states. The maximum half-width of the A line, which was observed at $\alpha = 0.5$ ($x = 0.056$ and 0.062; Fig. 1), was an order of magnitude larger than in ZnS-W ($\sim$20 meV).

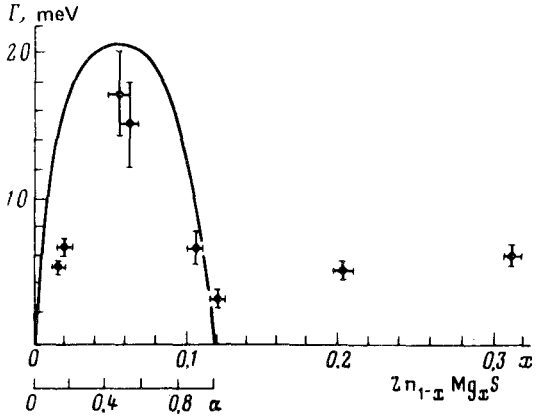

FIG. 1. Concentration dependence of the broadening of the fundamental exciton reflection line in the spectra of the solid solutions $Zn_{1-x}Mg_xS$ at $T = 2$ K. The interval $0 < x < 0.12$ corresponds to crystals containing one-dimensional disorder (having an incomplete sphalerite-wurtzite phase transition), while the interval $0.12 < x < 0.32$ corresponds to crystals with the wurtzite structure.

Two different types of disorder exist in SSS with stacking faults: three-dimensional disorder due to fluctuations of the composition of the solid solution, and one-dimensional disorder due to random sequences of alternation of the close-packed layers. Each disorder is associated with a random potential which alters the position of the edges of the energy bands. The interaction of an exciton with the random potential leads to broadening of the excitonic absorption (reflection) line. In the region of the structural phase transition the governing role is played by the interaction of excitons with the one-dimensional random potential created by local fluctuations of the stacking faults. This potential may be written[4]

$$V = u\frac{\zeta}{N}, \tag{1}$$

where $u = \partial E_A/\partial\alpha$, N is the total number of plane layers, and the fluctuation u is defined in the usual way:

$$\zeta(z) = N_h - \alpha N; \tag{2}$$

here N_h is the local concentration of hexagonal layers, averaged over a dimension larger than $(\alpha N)^{-1}$. We shall assume the absence of correlations in the distribution of the close-packed layers, i.e., we assume that the average value of the correlator is given by

$$<\zeta(z)\zeta(z')> = N\alpha(1-\alpha)\delta(z-z'). \tag{3}$$

With allowance for this condition, the Schrödinger equation describing the motion of the exciton in the one-dimensional random potential

$$\left[-\frac{\hbar^2}{2M}\frac{d^2}{dz^2} + \alpha\frac{\zeta}{N}\right]\Psi = E\Psi \tag{4}$$

can be reduced to dimensionless form with the aid of the substitutions

$$z = \frac{\hbar}{\sqrt{ME_0}} z' ; \qquad \zeta = \left(\frac{\hbar}{N\alpha(1-\alpha)\sqrt{ME_0}} \right)^{1/2} \zeta' , \tag{5}$$

where the characteristic energy E_0 is

$$E_0 = \frac{u^{4/3} \alpha^{2/3} (1-\alpha)^{2/3} M^{1/3}}{\hbar^{2/3} N^{2/3}} . \tag{6}$$

The absorption linewidth is expressed in units of E_0. Solution of the resulting system of dimensionless equations shows[5,4] that the linewidth may be given as

$$\Gamma = 1.42 E_0 . \tag{7}$$

Expression (6) was used to analyze the experimental data on the broadening of the exciton lines in the spectra of the semiconductor solid solutions $Zn_{1-x}Mg_xS$ with stacking faults; it was taken into consideration that the translational mass M of the exciton and the total number of layers N along the optic axis are the same as for ZnS crystals.[4] The parameter $u = \partial E_A / \partial \alpha$ is determined by the gradient of the position E_A of the fundamental exciton line A with changing concentration of the layers in the phase-transition region and can be found from the relation

$$\frac{dE_A}{dx} = \frac{\partial E_A}{\partial x} + \frac{\partial E_A}{\partial \alpha} \frac{d\alpha}{dx} , \tag{8}$$

where $dE_A/dx = 1.21$ eV is determined from the experimental dependence of $E_A(x)$ for SSS of the $Zn_{1-x}Mg_xS$ system containing stacking faults (Fig. 2, straight line 2) and $\partial E_A/\partial x = 0.5 + 0.92x$ is found from the concentration dependence of the position

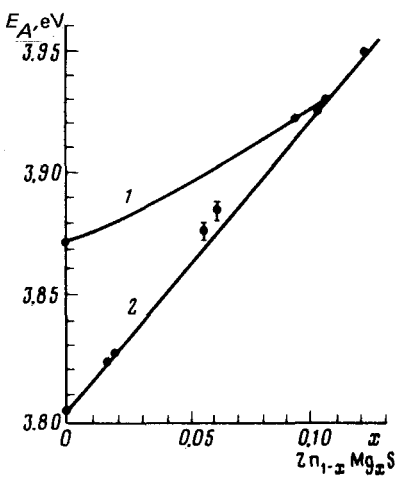

FIG. 2. Concentration dependence of the position of the fundamental exciton line $n = 1$ A in the reflection spectra of the solid solutions with different crystal structures ($T = 2$ K): 1) with the wurtzite structure; 2) with varying degrees of anisotropy. The points are experimental; the lines are: 1) the quadratic dependence (2) of Ref. 8; 2) a linear dependence.

of the line for hexagonal $Zn_{1-x}Mg_xS$ solid solutions (Ref. 8; curve 1 in Fig. 2). According to our experimental data, the phase transition moves at a rate $d\alpha/dx = 8.2$. By substituting these values into (8), we can construct the curve of the broadening Γ of the exciton line that is due to the one-dimensional disorder (the solid curve in Fig. 1); this curve gives a satisfactory description of the experimental data (the points in Fig. 1). For the phase-transition region ($x < 0.12$) the contribution of the three-dimensional disorder is small, since even for $x = 0.12$ the broadening due to this contribution is no more than ~ 5 meV and falls off rapidly with decreasing x (Ref. 2).

In conclusion, it should be stressed that the motion of the exciton as a whole can be localized by fluctuations of the random one-dimensional potential. The difference in the energies of localization at various points in the crystal leads to inhomogeneous broadening of the exciton states. Here there is always a bound-state level in the one-dimensional potential well that is comparable to the well depth,[4] whereas for the three-dimensional fluctuational potential well the corresponding level is high lying.[2] Therefore, the localization energy in the first case can be substantially larger than in the second. Since one-dimensional disorder occurs rather often in the structure of real crystals, it should be manifested quite generally in exciton spectroscopy.

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