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Fluctuation-trapped exciton states in 2D-semiconductor solid solutions

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Abstract. Exciton localization in quantum wells formed by solid solutions has been studied both theoretically and experimentally. The method for calculation of the density of fluctuation states below the edge of two-dimensional exciton band and the spectral density of exciton transitions is developed. The classification of states in respect to its migration properties and contribution to the luminescence processes has been carried out using the continual percolation theory. The shape of emission and absorption bands, as well as the position of mobility edge are calculated. The results of calculations are in good agreement with the optical spectra of ZnCdSe- and GaInAs-based QW structures.

Introduction

Light emission in most of low dimensional heterostructures is governed by localized exciton states. An inhomogeneous character of exciton emission bands with a strong variation of exciton migration properties across the band contour has been first established in GaAs/AlGaAs QW's [1] and has been attributed to exciton localization by potential fluctuations in AlGaAs solid solutions forming the barriers. In this connection a concept of mobility edge for the fluctuation states in the tail of exciton states has been introduced. It is a common point of view that localized exciton states in QW's are due to interface roughness and fluctuation of composition in the case of QW's formed by solid solutions. A specific feature of the localization is an occurrence of fluctuation effects even for solid solutions with a substitution in the cation sublattice unlike the bulk solid solutions, where cation substitution does not lead to a pronounced tailing [2].

1. Theoretical model

In present paper we shortly communicate the results of our studies of the effect of compositional fluctuations on exciton states in solid solution forming the quantum well. As a first step, we have calculated the value and energy dependence of the density of fluctuation states $\rho(\omega)$ and the spectral density of optical transitions $\alpha^0_{1s}(\omega)$ of the ground exciton state. The calculations have shown that the confinement of exciton states in 2D structures strongly enhances the effectiveness of perturbing potential due to compositional fluctuations in comparison with the case of three dimensional localization [3].

For calculation of the emission spectra we have analyzed the properties of fluctuation states at different energies in respect to their mobility followed by energy transfer. Qualitatively, the exciton states be considered either as *(i)* extended over a whole crystal or *(ii)* within a restricted region, with the ability to transfer the energy to the states with lower

energy, or *(iii)* as isolated, for which the probability of energy transfer supposed to be negligible. Both extended and isolated states contribute to absorption processes, whereas the optical emission comes mostly from spatially isolated localized exciton states. Using the Taylor series expansion for the averaged number of clusters in continual classical percolation over the overlapping circles, obtained in Ref. [4], we have calculated quantitatively the relative part of isolated states $P(\omega)$

$$P(\omega) = \left[\exp\{-2\mathcal{P}(\omega)\} + \mathcal{P}(\omega)\exp\{-2.841\,\mathcal{P}(\omega)\} + \ldots\right], \qquad (1)$$

where

$$\mathcal{P}(\omega) = \frac{1}{2}[R_{\rm int}/\overline{r(\omega)}]^2, \qquad \overline{r(\omega)} = \left(\frac{1}{\pi}\frac{1}{\mathcal{N}(\omega)}\right)^{1/2}.$$

The integral density of states $\mathcal{N}(\omega)$ gives the concentration of the potential wells with the localization energy restricted from one side by the ω and from the other side by Lifshitz border [5]. The first term in the right hand side of Eq. (1) gives the relative number of spatially isolated potential wells, while the second term presents number of complexes consisting of pairs of "interacting" wells [4].

As a next step, the emission spectrum has been calculated, as a product of the relative part of isolated states $P(\omega)$, and the emission probability, directly related to the absorption coefficient $\alpha^0_{1s}(\omega)$. Taking into account also the contribution to the emission of the extended (*(i)* and *(ii)*) states we have

$$I^0_{1s}(\omega) \sim \alpha^0_{1s}(\omega)\,\tau_{\rm rad}\,\{P(\omega) + \tau_{\rm rel}/\tau_{\rm rad}[1 - P(\omega)]\}, \qquad (2)$$

where the first term in the brackets on the right hand side $P(\omega)$ presents the fraction of the states with localization energy ω belonging to the isolated clusters and to the ground states of the finite size complexes of clusters (superclusters)*(iii)*. The second term is the part of the states*(ii)* which are mobile partially (higher states of superclusters) or completely (states *(i)* of percolation cluster) and have the possibility for relaxation. As a consequence, the last term is proportional to the ratio of the relaxation lifetime to the lifetime in respect to annihilation $\tau_{\rm rel}/\tau_{\rm rad}$. All other condition being equal, the $\tau_{\rm rel}$ depends on the averaged energy which can be transferred in relaxation processes, therefore, it depends on the width of the inhomogeneous broadening of the exciton band due to fluctuations.

2. Experimental details, results and discussion

We have studied the absorption, photoluminescence (PL) and excitation of luminescence (PLE) spectra of three different sets of MBE-grown samples:
(i) MQW's based on $Ga_{1-c}In_cAs/GaAs$ ($c = 0.03-0.16$) with wellwidth $L_{\rm w} = 6-9$ nm, separated by barriers $L_{\rm b} = 30-90$ nm;
(ii) ZnSe superlattices with 0.5 monolayer insertions of CdSe and 5 nm period;
(iii) single $Zn_{0.92}Cd_{0.08}Se/ZnSe$ QW-structures with different wellwidth $L_{\rm w} = 5-40$ nm.

Absorption and luminescence spectra of some of the (Ga-In)As/GaAs samples with different In concentration have been shown in semilogarithmic plot in the Fig. 1(a-c). The developed model has been applied for the analysis of the experimental spectra, and the results of model fitting are also presented in Fig. 1. The energy dependence of the spectral density for the n = 1s exciton state is shown by the curve 1, whereas the curve 2 shows the calculated luminescence spectrum. Curves 3 and 4 show the contribution into emission of isolated and extended states, respectively. The vertical dotted line corresponds to the

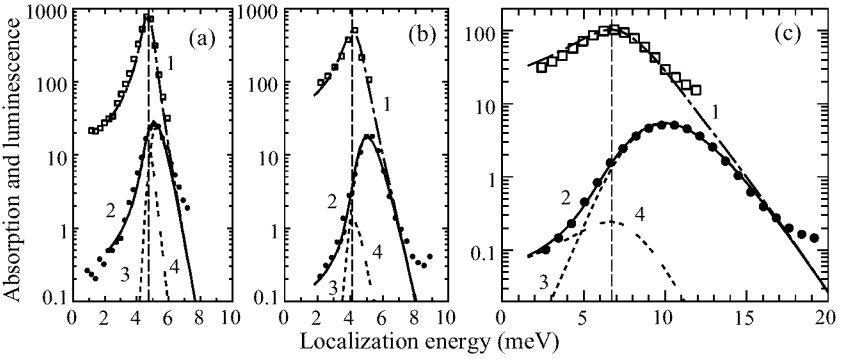

Fig. 1. (a)–(c): Absorption and luminescence spectra (open and solid symbols, respectively) of $Ga_{1-c}In_cAs/GaAs$ QW's for $c = 0.03$, 0.06 and 0.16, respectively. Lines — the results of theoretical calculations (see text). Mobility edge positions (vertical dashed lines) correspond to the photon energies 1.4905 eV, 1.4629 eV and 1.4256 eV for $c = 0.03$, 0.06 and 0.16, respectively.

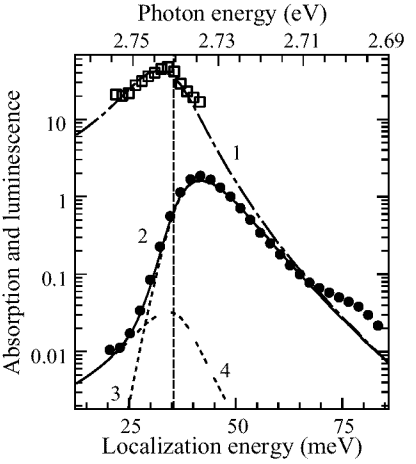

Fig. 2. Comparison of PL and PLE spectra of CdSe/ZnSe submonolayer superlattices (open and solid symbols, respectively) with the results of theoretical calculations (see text).

calculated position of the mobility edge. The energy scale shows the localization energy, measured from the expected position of the n = 1s exciton band bottom, not perturbed by fluctuations.

Experimental PL and PLE spectra of ZnSe superlattices with the submonolayer insertions of CdSe have been presented in Fig. 2. The growth conditions were corresponding to the insertion of submonolayers with 0.5 coverage, however the Cd atoms has been spreading over 4 to 6 lattice periods producing the average concentration of Cd in QW's of order of 10% [6]. The results of comparison are shown in Fig. 2 with the meaning of curves similar to that in Fig. 1.

As it can be seen from the figures, the experimental results can be well described by presented model, assuming the existence of exciton mobility edge near of the main absorption band maximum in accordance with the data presented in [1] and [7]. Similar results have been obtained for the studied set of single ZnCdSe/ZnSe QW's structures.

Summary

We have shown that the shape and Stockes shift of absorption and luminescence spectra of excitons in QW's formed by different solid solutions can be describe within the uniform theoretical approach. It has been shown that due to the two-dimensional character of exciton motion in quantum wells the effect of compositional fluctuation on exciton states is much stronger than in the bulk solid solutions of the same composition.

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