

Relaxation of Localized Excitons in CdSe/ZnSe Heterostructures Containing Quantum Islands of Different Sizes

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We report on localization dynamics of excitons in ensembles of self-organized CdSe islands embedded in ZnSe. The experimental methods employed are temperature dependent, spatially-resolved photoluminescence (μ -PL), spatially-integrated PL (macro-PL), as well as time-resolved PL (TRPL). We see the often observed non-monotonous shift of the PL maximum with temperature which we can explain by a redistribution of the excitons amongst the islands. The measured shift is compared with the exact shift of the bandgap deduced from μ -PL measurements and is found to depend strongly on island size and distribution. These transport processes are recovered in the temporal evolution of the PL. The decay time of the spectrally integrated PL reaches its maximum at exactly the same temperature at which the redshift of the macro-PL turns into a blueshift.

1. Introduction CdSe/ZnSe strained layer heterostructures have been intensively studied during the past years due to high expectations of their potential for optical devices in the visible region of the spectrum. A lot of work about the exact shape of the CdSe islands within these structures, as well as spectroscopy on single islands, has been published [1, 2]. However, in order to evaluate the prospects for macroscopic devices, the interplay between the different islands within these samples will be very important. A related feature is the s-shaped luminescence shift [3], the origin of which is under present investigation of many theoretical and experimental groups (see e.g. [4, 5]). We want to elucidate these problems by a combined study of temperature dependent, spatially and temporally-resolved PL.

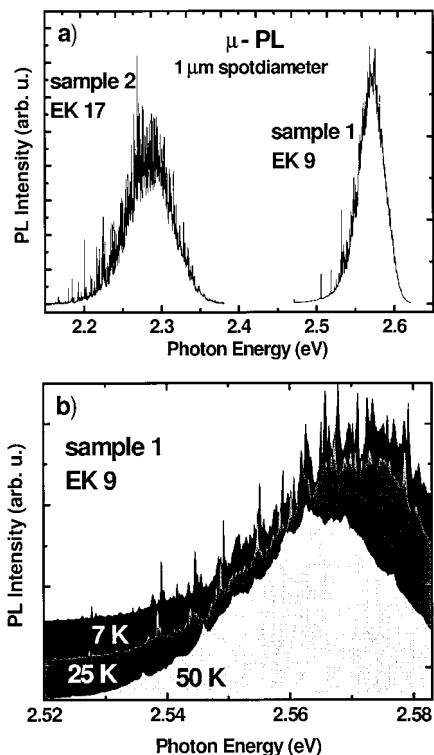
2. Experimental Results In this report, we present results of two CdSe/ZnSe heterostructures grown under similar conditions by molecular beam epitaxy using mixed elemental and compound sources. The only difference during the growth was a longer annealing of sample 2 to initiate Oswald ripening of CdSe islands. The structure of both samples is well known from plane-view Transmission Electron Microscopy (TEM) and, in case of sample 1, side-view high resolution TEM also. Details about the growth and structural properties are given elsewhere [6, 7].

The classification of the islands in CdSe/ZnSe heterostructures is not uniform in the literature. We will use a notation after [8, 9] where the islands are distinguished in three classes namely A, B and C islands. Sample 1 contains islands of class A, i.e. small islands with diameters < 5 nm and small class B islands with a diameter of about 10 nm. The density of A islands is about 10^{11} cm^{-2} and in the range of 10^{10} cm^{-2} for the B

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islands, respectively. In addition to A and B islands, sample 2 contains large class C islands which are bigger than 20 nm having defects in their center. The B islands in sample 2 have a larger distribution in size and Cd content than those of sample 1. The density of the A islands in sample 2 is roughly the same as in sample 1 but the density of B islands is decreased by the forming of C islands.

The μ -PL spectra of sample 1 and 2 are shown in Fig. 1. In case of the μ -PL measurements, the PL of the sample is collected by a microscope objective and focused onto a pinhole. This pinhole defines the spatial resolution of the setup which corresponds in this experiment to a spot 1 μ m in diameter. The pinhole is imaged onto the entrance slit of a 0.75 m focal length double-grating spectrometer which gives us, in combination with the pixel size of the Peltier cooled CCD, a spectral resolution of better than 50 μ eV. As excitation source the 351 nm line of an argon ion laser was used. It is clearly visible that the spectrally broad emission of both samples is superimposed by or in case of sample 2 even composed of sharp emission lines with a FWHM smaller than 100 μ eV. The individual lines in the emission of sample 2 are better resolved because of the larger distribution in size and Cd content causing a larger energy splitting between the lines. The emission energy is much lower than in sample 1 indicating a higher Cd concentration in this sample. The luminescence efficiency of sample 1 is much better because of a smaller number of defects acting as nonradiative centers. A similar reduction of the spectral integrated PL intensity with rising annealing time of the samples is also reported by other groups [10].



μ -PL spectra at different temperatures allow to track the emission of a single island with temperature (see Fig. 1b). This temperature shift directly corresponds to the bandgap shrinkage of the island material with temperature without disturbing influences of carrier redistribution. For further details about this method for a high precision determination of the temperature dependent bandgap shrinkage see [11].

The shift of the macro-PL maximum with regard to the position at 5 K of sample 1 is depicted in Fig. 2 (black boxes). As a comparison the shift of single emission lines (small open circles) and its extrapolation to higher T using Varshni's formula with CdSe parameters (dashed line) are shown. Although the Varshni formula is not adequate for low temperatures

Fig. 1. a) μ -PL spectra from sample 1 (internal name: EK 9) and 2 (EK 17) at $T = 5$ K; b) μ -PL spectra from sample 1 for different temperatures

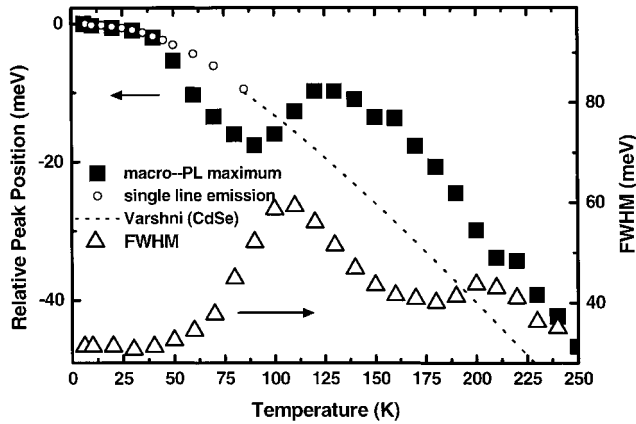


Fig. 2. Peak position of the spatially integrated PL for sample 1 (black boxes) with respect to the value for $T = 5$ K. Shift of the sharp emission lines in μ -PL (small open circles) and extrapolation using Varshni's formula with CdSe parameters (dotted line). Full width at half maximum (FWHM) of the spatially integrated PL (open triangles)

[11], it is still a good and easily comprehensible description for the bandgap shrinkage for higher T . For temperatures < 35 K the macro-PL peak follows the predicted bandgap, but for $35 \text{ K} < T < 90 \text{ K}$ the PL of the island ensemble is strongly redshifted with respect to the bandgap. At 90 K the peak position reaches a minimum and for $90 \text{ K} < T < 130 \text{ K}$ the emission blueshifts over about 10 meV. The redshift for $T > 130 \text{ K}$ follows essentially the expected bandgap shrinkage. The macro-PL peak of sample 2 with stronger localization (not shown here) follows the expected shift of the bandgap for almost 120 K. Only then it shows the mentioned redshift. Because of the high localization energy, the beginning blueshift for $T > 200 \text{ K}$ is already suppressed by nonradiative processes.

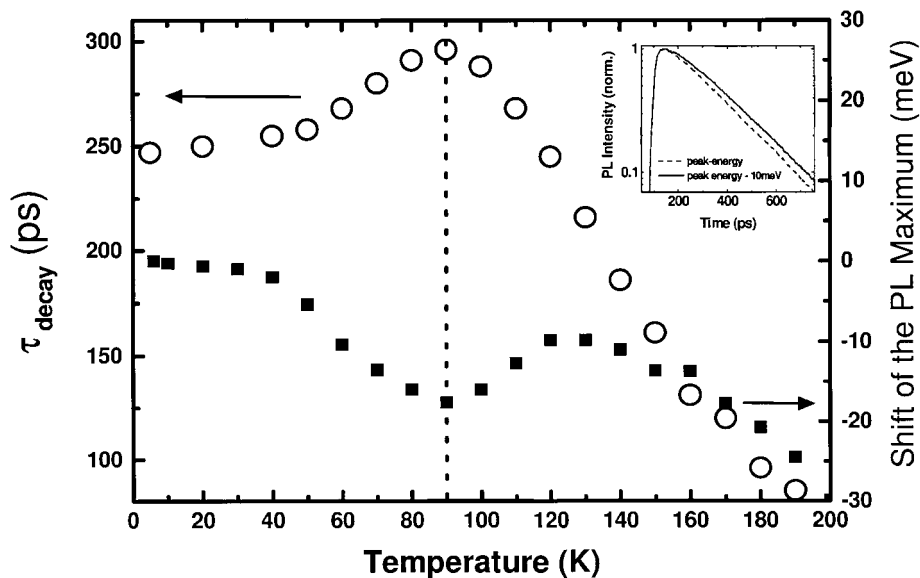


Fig. 3. PL decay time and PL peak position (boxes) vs. temperature for sample 1. To extract the mean decay time of the excitons in the island ensemble we integrated over the whole spectral range of the emission. In the inset the PL decay at the peak energy of the emission (dashed line) and 10 meV below (solid line) for $T = 5 \text{ K}$ is shown

For the same temperature range we performed time-resolved measurements. As excitation source a frequency doubled titanium:sapphire laser at 430 nm with a pulse length of about 150 fs and a repetition rate of 76 MHz was used. The PL was detected temporally and spectrally resolved by the combination of a 0.5 m focal length spectrometer and a synchroscan Streak camera. The overall time resolution of the setup was better than 3 ps.

The PL decay time from sample 1 spectrally integrated over the whole island ensemble is shown in Fig. 3 for different temperatures (open circles). For comparison, the shift of the macro-PL maximum is drawn (small black boxes). For $T < 90$ K the PL decay time increases and shows a maximum at exactly the same temperature as the PL position runs through a minimum. For $T > 90$ K the decay time strongly decreases.

3. Discussion How can it be explained that the minimum of the spectral position of the macro-PL and the maximum of the PL decay time are found at the same temperature? To clarify this point we will focus in the following on the dynamics of sample 1, because of the weaker contribution of nonradiative centers in this sample. For low temperatures < 35 K the emission of the ensemble shifts exactly like the emission of a single CdSe island. In the same temperature range the PL decay time and the FWHM of the macro-PL remain almost constant, which is consistent with the picture of strongly localized carriers. In the temperature range $35 \text{ K} < T < 90 \text{ K}$ where the redshift of the ensemble emission is much steeper than expected from the bandgap shrinkage alone, the PL-decay time increases. This rise is typical for free excitons [12] and also for excitons in quantum dots [13]. However, for free excitons or excitons remaining in one localization site, we would not expect a strong redshift of the PL. This means that the observed increase of the decay time must have other reasons. The most likely reason is a redistribution of the excitons amongst the islands. Excitons localized in islands can reach deeper localization centers in their neighborhood by thermal activation. This explains the strong redshift and the increasing FWHM of the emission. The lengthening of the decay time can then be understood because the decay of deeper localized excitons owing to the reduced coupling to the lightfield is slower than the decay of weakly localized excitons (see inset of Fig. 3). The related transport processes are recovered as a redshift of the emission on a time scale of a few hundreds of picoseconds. For a detailed description of this temporal redshift and saturation processes in these samples see [14].

The dynamics of the system changes drastically at $T = 90$ K. The PL decay time begins to decrease and the macro-PL starts to blueshift. The thermal activation is now high enough for the excitons to escape deeper localization centers and to decay radiatively from delocalized states. This explains both the blueshift of the emission caused by changes in the exciton distribution and the beginning drop of the decay time considering the increasing influence of nonradiative processes. This can be verified from temperature dependent measurements of the external quantum efficiency [15].

At temperatures > 130 K the emission redshifts again. This can easily be understood because most of the excitons have already escaped out of their localization sites and the rising contribution of the bandgap shrinkage governs the scenario. The dramatic decrease of the decay time in this temperature range is most likely due to nonradiative processes which become dominant.

4. Conclusions In summary we have investigated the time-integrated, spatially and temporally-resolved PL for MBE grown CdSe/ZnSe heterostructures containing self-organized CdSe islands of different sizes and Cd content over a temperature range from 5 to 250 K. The non-monotonous shift of the peak energy of the PL can be explained by a thermally activated redistribution of the excitons amongst the islands. This explanation is consistent with the results of time-resolved PL measurements.

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