Time-Resolved Studies and High-Excitation Properties of CdSe/ZnSe Quantum Dots

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Abstract Investigating CdSe/ZnSe quantum dot structures the population processes and the decay mechanisms of different island structures were obtained. For all islands a decay process with a short and a long component was observed at low temperatures, but only for the short component (300ps) a systematic correlation to the quantum dot size was found. According to temperature-dependent investigations and the excitation behavior the longer decay time constant (several ns) is assigned to the influence of population and redistribution mechanisms.

1 Introduction

The CdSe/ZnSe system became a promising candidate for the investigation of the dynamic in quantum dots (QD) in recent years [1]. Because of the large binding energies this system is uniquely suited for investigations of three-dimensionally confined excitons. However, the complex growth kinetics of such ZnSe-based structures produces various types of islands, which differ in size and Cd content [2,3]. While in conventional III-V structures the formation of islands is already established [4], and there, excited states and phonon-assisted processes are responsible for population and recombination of carriers in QDs [5], a different situation is expected in CdSe/ZnSe based QD structures containing small islands of low Cd-content (dubbed type A) and larger islands grown in the Stranski-Krastanow mode (dubbed type B) islands [2,6]. Consequently, competitive processes between the different island types may influence the decay behavior.

In the present paper we will demonstrate that redistribution processes of excitons play an important role in QD structures containing type A and type B islands. Therefore, the excitation properties and the decay behavior of these islands will be discussed.

2 Experimental

Pseudomorphically strained CdSe/ZnSe structures containing single-layer sheets of type A and type B islands embedded in a ZnCdSe wetting layer were studied. The respective island densities (type A: some $10^{11} cm^{-2}$; type B: from $1 \cdot 10^9$ to $5 \cdot 10^{10} cm^{-2}$) can be controlled by the growth process enabling detailed optical and structural investigations. All samples are grown by MBE. More details of the growth procedure and the structural properties are given in [2,7]. For the optical investigations the samples were mounted in a cryostat providing temperatures between 2K and 300K. Time-resolved investigations were performed using a Nd:YAG pumped dye laser with a cavity damper. The luminescence was detected with a multi-channel plate attached to a subtractive 0.35m double-grating monochromator. The laser pulse has a duration of 4ps, while the temporal resolution of the whole system is determined by ¡20ps. The light of a tungsten lamp was dispersed by a 0.3m double-grating monochromator for the photoluminescence excitation (PLE) measurements. The spectral resolution of the setup was better then 1nm.

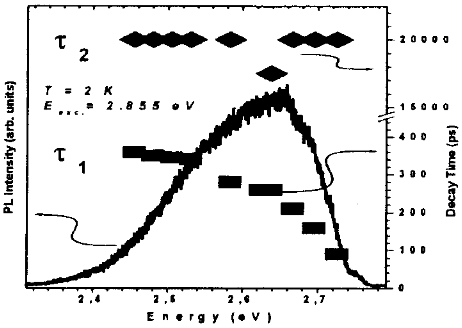

Fig. 1 Photoluminescence and decay constants of a CdSe/ZnSe sample containing type A and type B islands

3 Results and Discussion

To obtain more information about the intricacies of the rise and decay processes, and thus, about the electronic structure of type A and type B islands, PLE and time-resolved spectroscopy with resonant and non-resonant excitation were performed. Fig. 1 shows a PL spectrum of a sample containing both types of islands, type A and type B, and the decay constant as a function of the detection energy . The PL band is originated by the recombination of excitons localized in type A islands while the emission from type B islands contribute to the low energy tail of the PL band as it was revealed by temperature dependent PL investigations [6]. The time decay constants were derived from a convolution procedure. A reasonable fit was achieved with time constants of $\tau_1 \approx 250ps$ and $\tau_2 \approx 10ns$. First, the dominant process with the fast decay constant will be discussed. This constant expresses the radiative decay of excitons local-

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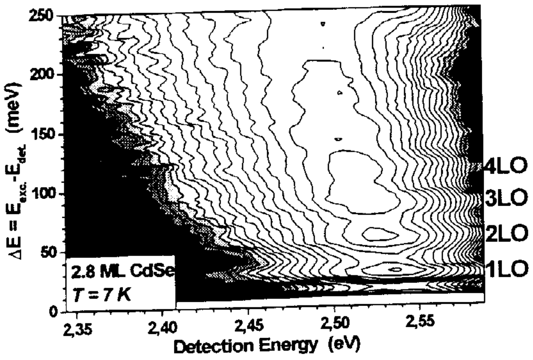

Fig. 2 Color-coded map of the excess energy (DE) as a function of the detection energy for a CdSe/ZnSe sample containing type A and type B islands. Light color indicates high intensity.

ized in QDs. From such recombination it is known that the decay constants are very sensitive to the binding energy and the spatial overlap of the electron- and hole-wavefunction. The detected increase in the decay times of excitons localized in type B islands is attributed to the larger diameter of these islands and the resulting smaller overlap of the carrier wavefunctions. The slow decay component remained unaffected by the island size. The excitation behavior of the islands suggest that the slow components with time constants of several ns are originated by population and redistribution processes. The nature of this slow process is in more detail discussed elsewhere [8] more detailed discussion of this To enlighten the interactions of the carriers between the islands and their excitation mechanisms PLE spectroscopy was performed. Fig. 2 shows a typical color coded map of the excess energy as a function of the detection energy for a sample with type A and type B islands. Areas with light color indicate an efficient excitation channel. As the main result from our investigations we found hot exciton relaxation as the most efficient excitation channel below the band gap energy of the ZnSe barriers. This behavior suggests the relaxation via the wetting layer and a wide distribution of island energy levels. The diagonal line in the left bottom corner of Fig. 2 indicates the first common electronic state of the QDs, the wetting layer. Excitons localized in type A islands overcome this barrier by thermal activation at temperatures above 100K resulting in a larger mobility [8]. This effect was confirmed by temperature-dependent time-resolved PL investigations: evaporation into the wetting layer state caused a pronounced decrease of decay times up to values observed for ZnCdSe quantum wells. Meanwhile the decay times detected at the maximum of the PL band (evtl. fig. 3, wenn noch Platz) are increased accompanied by a redshift being stronger as it would be to expect for ZnSe or CdSe. This can be attributed to the redistribution of excitons via the wetting layer into larger islands (finally, type B islands) with smaller transition energies and deeper potential mirrors to prevent excitons from thermal activated evaporation into the wetting layer [6, 9]. Due to the observed PLE data we assign hopping mechanisms providing the lateral energy transfer between the islands. These processes are enabled by the small localization energy of excitons in type A islands. A significantly influence of excited QD states was not observed.The observed excitation of localized emission with excess energies below the LO phonon energy can be explained by the increased influence of other phonon energies provided by dot structures [10] and by the complex potential landscape in the neighborhood of the islands which is caused by type A islands and roughness of the wetting layer.

4 Conclusion

The radiative decay of the excitons localized in CdSe/ZnSe quantum dots was determined to 250ps to 400ps as a function of the lateral island diameter. Additionally, lateral energy transfer processes contribute to the population of the quantum dots resulting in a longer decay process with a time constant of several ns. According to the size distribution and the high density of the islands hot exciton relaxation was observed as the most efficient excitation channel below the bandgap energy of the ZnSe barriers. The redistribution of excitons in larger islands providing deeper localization sites is attributed to hopping processes via the wetting layer.

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