phys. stat. sol. (b) **229**, No. 1, 463–467 (2002)

Influence of the Quantum Island Distribution on Relaxation of Localized Excitons in CdSe/ZnSe Heterostructures

B. Dal Don[1]), R. Dianoux, S. Wachter, M. Schmidt, E. Kurtz, G. von Freymann, U. Neuberth, C. Klingshirn, M. Wegener, and H. Kalt

Institut für Angewandte Physik, Universität Karlsruhe (TH), Wolfgang-Gaede-Straße 1, D-76131 Karlsruhe, Germany

(Received July 27, 2001; accepted September 30, 2001)

Subject classification: 73.21.La; 78.47.+p; 78.55.Et; 78.67.Hc; S8.12

We report on relaxation of excitons in II–VI heterostructures containing CdSe islands embedded in a ZnSe matrix, and showing different localization properties. We perform spatially resolved photoluminescence (μ-PL), time-resolved PL (TRPL), and time-resolved near-field spectroscopy (n-TRPL) to study the differences between two samples. In the sample showing the weaker localization, we see a correlation between the localized states, which is not obvious in the sample with the stronger localization. We consider as possible explanations for these repeated lines phonon-assisted transport as well as relaxation from excited states into a lower energetic state.

Introduction II–VI semiconductor nanostructures are very promising in the domain of optoelectronic devices working in the visible spectral range and have been intensively studied in the last years. A lot of works deal with the control of the formation of quantum islands. Besides the investigation of optical properties of one single island, it would also be very relevant to understand the interplay between the islands of a sample. In order to discuss this aspect, we investigated two samples containing CdSe quantum islands with different localization properties, and performed space and time resolved photoluminescence spectroscopy to compare them.

Experimental Results In this report, we present the study of two samples containing CdSe quantum islands embedded in a ZnSe matrix. These heterostructures were both grown on a (001) oriented GaAs substrate by molecular beam epitaxy, using a CdS compound source and an elemental Se source. Sample 2 was grown in the same way as sample 1, with an additional annealing period of 300 s by about 600 K, which caused Oswald ripening of the CdSe islands and changed their distribution. High-resolution transmission electron microscopy (HRTEM) gave us information about the structure of the two samples [1]. Sample 1 contains smaller islands of about 5 to 10 nm, at a distance in average of a few nm. Sample 2 contains islands with a larger size distribution and a higher Cd-content, which are more distant from one another. Some of the islands are accompanied by stacking faults, and act as non-radiative recombination centers. As a consequence, the two heterostructures show different localization properties.

A comparison of the (μ-PL) spectra of the two samples is shown in Fig. 1a. The sharp lines caused by the localized states are much better resolved in case of sample 2. The PL emission is lower energetic in case of sample 2, due to the higher Cd-content of the

[1]) Corresponding author; Phone: +49 721 608 3555; Fax: +49 721 608 8480; e-mail: benedicte.daldon@phys.uni-karlsruhe.de

 0370-1972/02/22901-0463 $ 17.50+.50/0

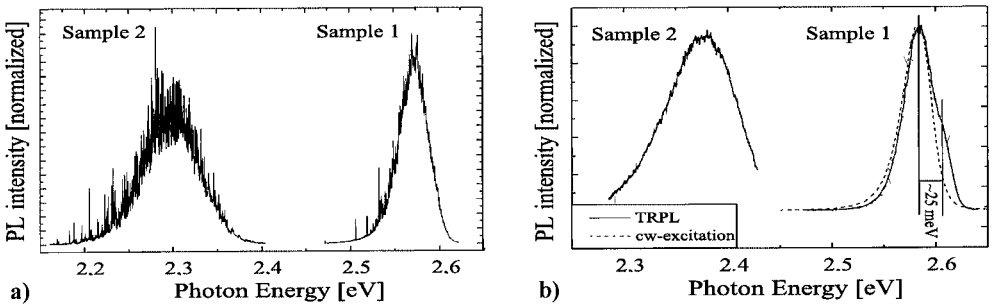

Fig. 1. a) μ-PL of samples 1 and 2, with a spatial resolution of 1 μm. b) TRPL of samples 1 and 2. Dashed line: PL of sample 1 under cw-excitation

islands. This leads to a stronger localization of the excitons in the islands of sample 2 than in sample 1. These features are confirmed by the shift of the PL maximum with temperature [3].

We performed time-resolved PL on the two samples. As an excitation source we used a frequency-doubled Ti:sapphire laser at 430 nm and with a repetition rate of 75 MHz. The PL spectra integrated over the first 100 ps after excitation are pictured in Fig. 1b. We observe that the PL spectrum of sample 1 can be decomposed in two contributions separated by roughly 25 meV, the lower energetic peak corresponding to the PL maximum under continuous-wave excitation conditions. This second PL-line appears under all excitation conditions. For sample 2, the presence of another contribution in the PL is not obvious.

In order to understand the origin of this higher-energetic luminescence line in the PL, we performed time-resolved near-field spectroscopy (n-TRPL). The setup consists of a near-field scanning optical microscope (NSOM) [4] used in internal reflexion mode, coupled with a synchroscan streak camera. The whole system provides a spatial resolution in the order of 200 nm, and a temporal resolution of a few ps [5]. We scan different parts of the sample, in order to extract general trends characteristic of the excitons in the islands and to eliminate individual properties due to the inhomogeneous distribution of the islands. Typical n-TRPL spectra of sample 1 are depicted in Fig. 2.

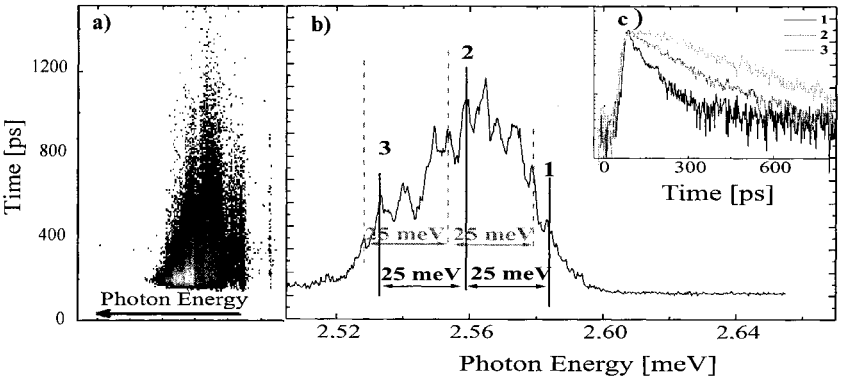

Fig. 2. n-TRPL spectrum of sample 1. a) Streak image; b) n-TRPL with the repeated line separated by 25 meV; c) dynamics of correlated lines

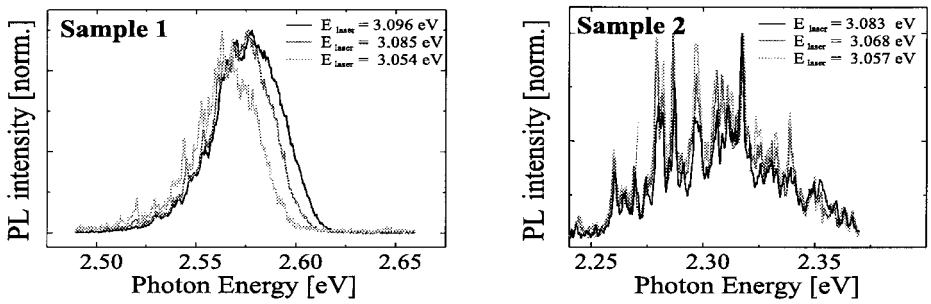

Fig. 3. Influence of the laser energetic position on the PL for both samples

They are composed of sharp lines which, as we can observe on the streak image (Fig. 2a), last a few hundreds of ps.

Figure 2b represents the PL spectrum integrated over the first 100 ps. We can distinguish a few lines above a broad luminescence structure. Most of these lines are repeated at a constant distance of 25 meV, which is consistent with the spatially integrated TRPL measurements. The temporal evolution of correlated lines strongly depends on their energetic position (Fig. 2c). After a steep rising phase of about 50 ps common to all lines, the lower energetic lines show an additional slower rise time or even a plateau-like behaviour, and a longer decay time. In case of sample 2, we did not find any obvious correlation between any of the lines.

The effects of the energetic position of the laser excitation on the excitonic population in both samples are shown in Fig. 3. For these measurements, we tuned the frequency doubled laser excitation from 400 nm to 430 nm, which corresponds to an excitation energy of about 17 LO-phonons above the luminescence of the samples. In case of sample 1, we observe a shift of the whole PL spectrum in the same direction as the shift of the laser excitation. On the lower energy side, we also see that the sharp lines gain or loose strength according to the excitation conditions. For sample 2 the dependence is not as pronounced as for sample 1, nevertheless we see lines being enhanced or disappearing with different laser positions.

Discussion First of all, we notice that our observations under pulsed excitation do not correspond to the continuous wave experiments, where no second contribution to the spatially integrated PL was found. A statistical analysis of space-resolved time-integrated PL of these samples showed a correlation of sharp lines separated by 18 meV [6]. In our time resolved experiments, we are neglecting the longer living processes, which can be observed in the cw experiments. As a consequence, the pulsed and continuous wave experiments point out different properties of the excitons in the sample.

Where does this higher energetic contribution to the PL come from? The energy distance of 25 meV between the lines is very close to the energy of the CdSe-LO-phonon (28 meV in bulk material) and incites us to consider LO-phonon assisted processes as possible relaxation mechanisms. The strong changes in n-TRPL by tuning the laser excitation energy, and that even with an excitation 17 LO-phonons above the luminescence, demonstrates that the coupling to LO-phonons is particularly efficient in sample 1. This is confirmed by the rapid rise time of all the lines, showing that the excitons cre-

ated by the laser pulse are rapidly captured into the islands. The fact, that the sharp lines have a decay time in the order of a few 100 ps permits us to exclude Raman scattering. Moreover, the different dynamics of the correlated sharp lines is in contradiction with the hypothesis of phonon-assisted recombination: in that case, the repeated lines would represent the same state and have the same dynamics. The most probable mechanism involving LO-phonons seems to be phonon-assisted transport between the islands (inter-island relaxation), or within a structurally inhomogeneous island (intra-island relaxation). The fact, that the repeated lines only appear in the sample with the weaker localization strength and the higher island density speaks in favour of this explanation. However, the probability for one exciton to tunnel more than one time is very low.

One other possible explanation is the relaxation of excited states within the islands. This explanation would be consistent with the dynamics of the lines. However, as the lines are repeated several times in our spectra, this hypothesis would imply equidistant excited states, that is to say a parabolic potential for a majority of islands. The energy intervall between the states should also be the same in all the islands, and correspond to the energy of one LO-phonon. This relaxation mechanism should also be observed in sample 2.

Excitonic relaxation in CdSe/ZnSe quantum dot samples has also been discussed by other groups. Using spatially resolved techniques, Robinson et al. [7] observed two different confined states in a quantum dot. Lowisch et al. [8] considered phonon-assisted intra-dot relaxation. On the other hand, inter-dot transfer at elevated temperature has been reported by Seufert et al. [9]. Concerning the low temperatures, Türck et al. [10] also observed a carrier feeding by means of a phonon-assisted tunneling from deep level states in the barrier into the quantum dots. Furthermore, our energy distance of 25 meV between the repeated energetic states is consistent with the observations of Rho et al. [11]: They evaluated the LO-phonon energy to 27.3 meV in case of a CdSe quantum dot and to 23 meV for a 2D-like CdSe island.

Conclusion We have compared two CdSe/ZnSe heterostructures having different localization properties, using time-resolved, spatially resolved, and both space- and time-resolved PL. In the sample showing the weaker localization, and the highest density of islands, we saw correlated excitonic localized states separated by 25 meV. This behaviour was not obvious in the sample with strong localization in the islands. We consider relaxation of an excited state and phonon-assisted transport as possibly involved mechanisms.

Acknowledgement The authors would like to thank the Deutsche Forschungsgemeinschaft for financial support.

References

[1] E. KURTZ, M. SCHMIDT, M. BALDAUF, S. WACHTER, M. GRÜN, D. LITVINOV, S.K. HONG, J.X. SHEN, T. YAO, D. GERTHSEN, H. KALT, and C. KLINGSHIRN, J. Cryst. Growth **214–215**, 712 (2000).
[2] Y.H. CHO, G.H. GAINER, A.J. FISCHER, J.J. SONG, S. KELLER, U.K. MISHRA, and S. P. DENBAARS, Appl. Phys. Lett **40**, 1370 (1998).
[3] S. WACHTER, B. DAL DON, M. SCHMIDT, M. BALDAUF, A. DINGER, E. KURTZ, C. KLINGSHIRN, and H. KALT, phys. stat. sol. (b) **224**, 437 (2001).

[4] G. von Freymann, D. Lüerssen, C. Rabenstein, M. Mikolaiszyk, H. Richter, H. Kalt, Th. Schimmel, M. Wegener, K. Okhama, and D. Hommel, Appl. Phys. Lett. **76**, 203 (2000).
[5] B. Dal Don, R. Dianoux, S. Wachter, E. Kurtz, G. von Freymann, U. Neuberth, C. Klingshirn, and H. Kalt, to be published.
[6] G. von Freymann, E. Kurtz, C. Klingshirn, and M. Wegener, Appl. Phys. Lett. **77**, 394 (2000).
[7] L. M. Robinson, H. Rho, J.C. Kim, H. E. Jackson, L. M. Smith, S. Lee, M. Dobrolowska, and J. K. Furdyna, Phys. Rev. Lett. **83**, 2797 (1999).
[8] M. Lowisch, M. Rabe, F. Kreller, and F. Henneberger, Appl. Phys. Lett. **75**, 2489 (1999).
[9] J. Seufert, M. Rambach, G. Bacher, A. Forchel, M. Keim, S. Ivanov, A. Waag, and G. Landwehr, Phys. Rev. B **62**, 12609 (2000).
[10] V. Türck, S. Rodt, M. Strassburg, U.W. Pohl, and D. Bimberg, phys. stat. sol. (b) **224**, 643 (2001).
[11] H. Rho, L. M. Robinson, L.M. Smith, H.E Jackson, S. Lee, M. Dobrowolska, and J.K. Furdyna, Appl. Phys. Lett. **77**, 1813 (2000).