Superlattices and Microstructures, Vol. 25, No. 1/2, 1999
Article No. spmi.1998.0625
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Exciton relaxation in $Ga_{1-x}In_xAs/GaAs$ self-organized quantum dots

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(Received 26 October 1998)

The exciton dynamics in $Ga_{1-x}In_xAs/GaAs$ self-organized quantum dots grown on GaAs (111)B substrates are studied by the time-resolved photoluminescence (PL). We have found the intra-dot exciton relaxation by the reduction of the linewidth and peak energy and also by the energy-dependent PL rise time in the transient PL spectra. Compared with the energy relaxation in the reference quantum wells, we have confirmed that the exciton relaxation in three-dimensionally confined quantum dots is slower than in the quantum wells.

Key words: transient photoluminescence, self organized quantum dots, hot excitation relaxation.

Three-dimensionally confined quantum dots are of great interest both for device applications and for the research of fundamental physics. Compared with the dry- or wet-etched dots, the self-organized quantum dots are found to have a better interface quality and the non-radiative centers at the interface for the optically injected carriers are greatly reduced. Therefore, a high PL efficiency is obtained in the self-organized quantum dots. Generally, the self-organized quantum dots are grown by MBE in the so called Stranski–Krastanov mode where the strain relaxation between two materials with large lattice mismatch leads to defect free island formation. Recently, Tsai *et al.* [1] found that using the growth characteristic of (111)B GaAs at low substrate temperatures, $Ga_{1-x}In_xAs$ quantum dots with a height of 5 nm and a diameter of 50 nm can be formed. In this case the formation is not caused by strain relaxation. With a suitable selection of the growth temperature, dots are formed with In composition as low as 12%. In this manuscript, we present the time-resolved PL of these dot samples. We show that the three-dimensional confinement of the quantum dots reduces the exciton relaxation to a considerable extent in these $Ga_{1-x}In_xAs$ quantum dots grown in a (111)B direction, which has been discussed previously in other quantum dot systems [2].

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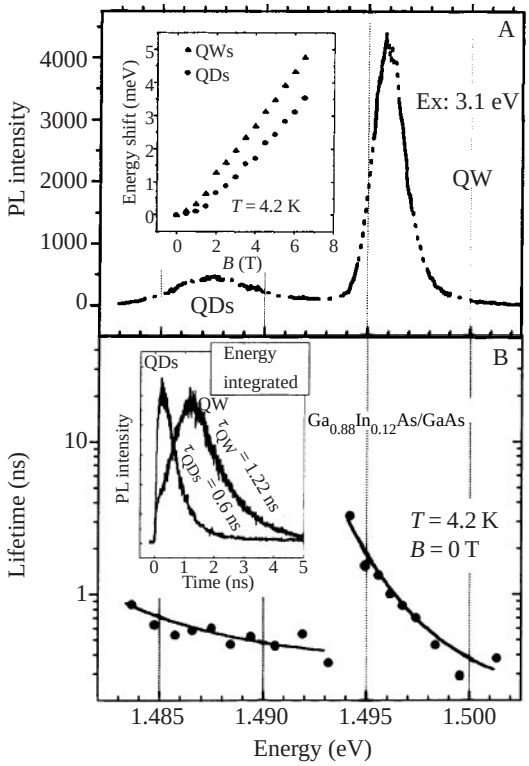

Fig. 1. A, PL spectrum and the diamagnetic shift of the peak energy in magnetic fields. B, The energy-resolved PL intensity decay time and also the time dependence of the energy-integrated PL intensity for the quantum dots and the quantum well.

Two samples with 15% and 12% In will be presented in this study. The sample structure is as follows: on the GaAs (111)B substrate with a 2 degree tilt towards (211), a 300 nm GaAs buffer layer was grown at first. Successively grown were a 1.5 nm $Ga_{1-x}In_xAs$ single quantum well layer and a 1.5 nm (growth time averaged thickness) $Ga_{1-x}In_xAs$ self-organized quantum dot layer separated by a 150 nm GaAs barrier. On top of the quantum dot layer a cap layer of 50 nm GaAs was grown. Optical experiments were performed at liquid helium temperature with magnetic fields up to 7 T. Samples were excited both resonantly and nonresonantly by an amplified mode locked titanium-sapphire laser and optical parametric amplifier (OPA) system from Spectra Physics. The transient PL spectra are taken by a Hamamatsu streak camera C4334 with a time resolution of 5 ps.

The time-integrated PL spectrum of the $Ga_{0.88}In_{0.12}As/GaAs$ sample is shown in Fig. 1A. The spectrum has two peaks corresponding to the emission from the self-organized quantum dot layer at the lower energy side and the reference quantum well at the higher energy side. The linewidth of the quantum well is 1.9 meV while that for the quantum dots is 4 meV showing a very good sample quality. The diamagnetic shift in the Faraday configuration is plotted in the inset of Fig. 1A. The diamagnetic shift of the quantum dots is smaller than that of the quantum well, which is a significant manifestation of the three-dimensional confinement. The diamagnetic shift studied up to 40 T on these samples can be found in Ref. [3]. The PL intensity decay time as a function of the emission energy is plotted in Fig. 1B. For the quantum well, the decay time increases drastically with lower emission energy, which represents clearly the exciton relaxation from the high energy states to the lower ones. The energy dependence of the PL decay time is, however, not so pronounced for quantum dots. Thus, Fig. 1B depicts a difference of the exciton relaxation between the three-dimensionally confined quantum dots and one-dimensionally confined quantum well. The energy-integrated PL decay time

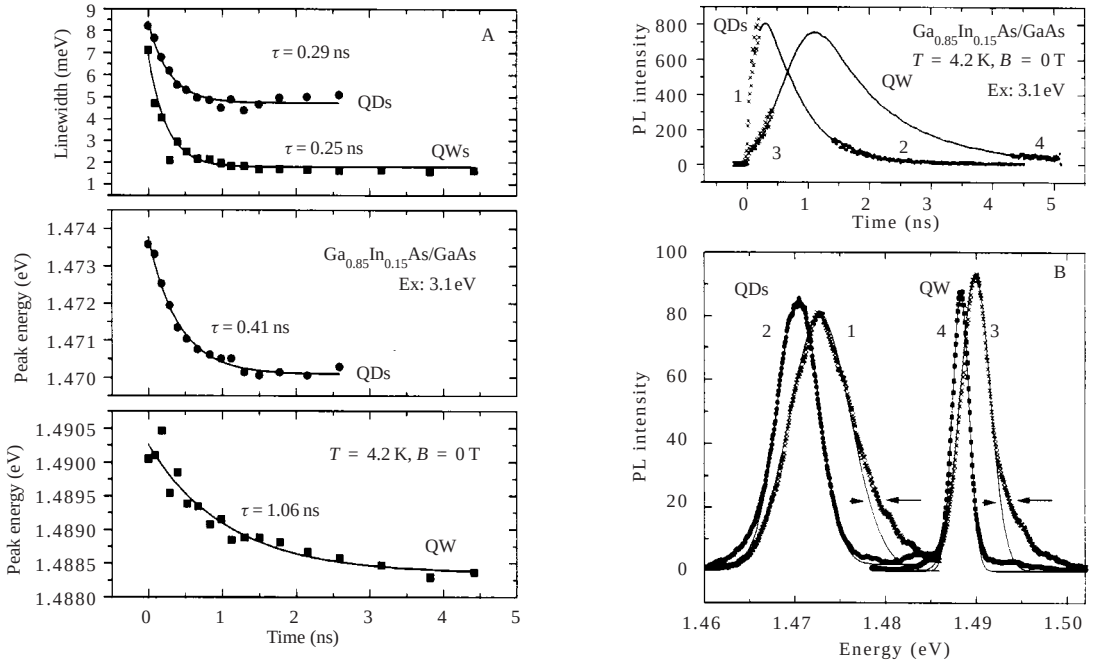

Fig. 2. A, The linewidth and the peak energy of the transient PL spectra versus the time delay with respect to the laser pulse. B, The hot exciton PL spectra 1 and 3 integrated within a short time (300–500 ps) right after the laser excitation and the PL spectra 2 and 4 for relaxed excitons integrated after a certain time delay (1.5–5 ns).

for the quantum dots and the quantum well is shown in the inset of Fig. 1B. The lifetime of the quantum dots is shorter than that of the quantum well, which can be ascribed to a higher number lattice imperfections at the interface of the quantum dots than that of the quantum wells. The long rise time for the quantum well is caused by the carrier migration from the donor and acceptor states in the barrier to the excitonic states in the quantum well driven by the piezo-electric field which will not be described here in detail.

The time dependence of the peak energy and the time dependence of the linewidth in the transient PL spectra for the sample with 15% In, excited by a laser pulse at 3.1 eV, is plotted in Fig. 2A. Similar results have also been obtained on the 12% In samples. The energy decay time of the quantum well is very long and reaches 1.06 ns, while that of the quantum dots is only 0.41 ns. The linewidth of both the quantum well and the quantum dots decreases exponentially with the time. The time constant for the linewidth decreasing in the quantum dots is even longer than that of the quantum well. In order to understand the physical meaning of the decreasing of the transient peak energy and the linewidth, we plot the PL spectra integrated shortly after the laser excitation, noted as spectra 1 and 3 in Fig. 2B and the spectra integrated near the end of the exciton lifetime, noted as spectra 2 and 4 in Fig. 2B. Shortly after the laser excitation, the spectral line shape of both the quantum well and the quantum dots is asymmetrical in energy space. At the high energy side, the transient PL intensity is much higher than the symmetrically fitted Gaussian profile due to the existence of hot excitons after the laser excitation. These hot excitons will relax to low energy states after some time delay. Hence, after a certain time delay, the spectral line shape becomes symmetric and can be well fitted by a Gaussian profile. The linewidth of spectra like 2 and 4 are caused by alloy or well-width fluctuations.

According to the above arguments, we can conclude that the reduction of the linewidth of the transient PL spectra characterizes the relaxation of the hot excitons, or relaxation of excitons located at higher energy

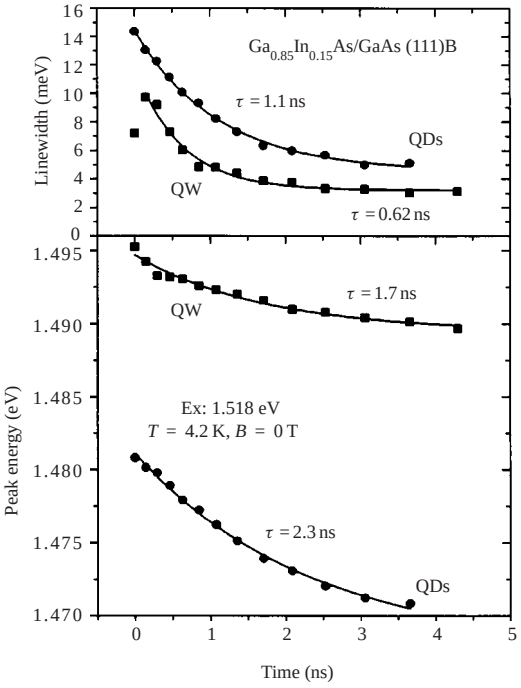

Fig. 3. Linewidth and peak energy of transient PL spectra versus the time delay after laser pulse under excitation below the gap of GaAs at 1.518 eV.

states. Due to the asymmetrical spectral line shape, the interpretation of the peak energy of the emission spectra would be rather complicated. It depends on the initial faster cooling of the photo-excited carriers by optic phonon emission and the following slower cooling by acoustic phonon emission. If the initial cooling of the photo-excited carriers dominates the cooling process, then the transient peak shift would be very slow because most of the carriers are already in the ground states, like the peak shift of the quantum well shown in Fig. 2A. Nevertheless, the linewidth is much more sensitive to the existence of hot excitons and its reduction shown in the upper part of Fig. 2A is more suitable for extracting information about the carrier cooling process. The decay time of the linewidth therefore characterizes the relaxation of excitons in the quantum well as well as in the quantum dots. The experimental results in Fig. 2A show that in quantum dots, the exciton relaxation is slower than that in the quantum well, which corroborates the theoretical predictions [4] as well as the experimental analysis performed in $Cd_{1-x}Zn_xSe/CdSe$ dot systems [2, 5].

The linewidth of the PL spectra increases drastically under excitation below the band gap of GaAs. The temporal variation of the linewidth and the peak energy of the transient PL excited with a laser pulse at 1.518 eV is shown in Fig. 3. The linewidth of the quantum dots directly after the laser pulse increases from about 8.5 meV for 3.1 eV excitation to 15 meV for 1.518 eV excitation, while the linewidths emitted by the relaxed exciton are approximately the same, about 5.5 meV in both cases, compare Fig. 2A and Fig. 3. Similar behavior is also found for the quantum well, except that the saturated linewidth for the quantum well is slightly larger in Fig. 3 than in Fig. 2A. This is caused by a reduced spectral resolution due to a bigger slit width on the spectrometer because of the smaller absorption cross section with excitation below the barrier.

The temporal peak energy shift excited below the barrier band gap is obviously larger than that excited above the barrier band gap. It changes from around 2 meV for the quantum well and 4 meV for the quantum dots under the excitation above the barrier band gap to 5 meV for the quantum well and 10 meV for the quantum

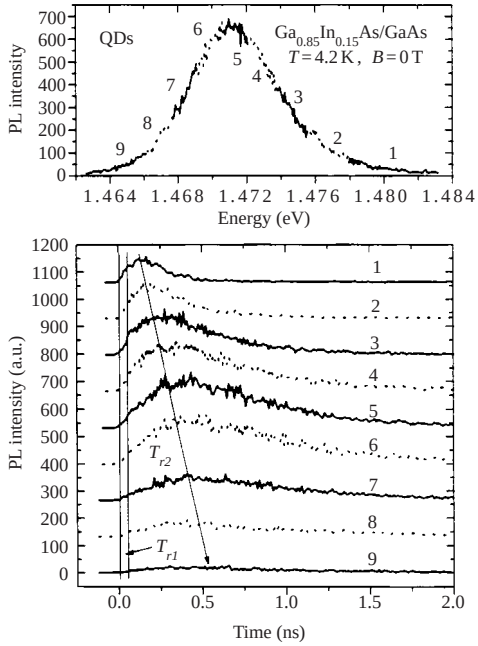

Fig. 4. Spectral dependence of the PL rise time for the quantum dots excited by a laser pulse at 3.1 eV.

dots under the excitation below the barrier band gap. This means that the initial cooling of the photo-excited carriers under resonant excitation is much smaller than under nonresonant excitation. The energy difference between the laser pulse and the excitonic emission in the quantum well, as well as that in the 12% In dots, is less than one LO phonon energy (36 meV) of GaAs. However, the energy difference for the 15% In quantum dots is still larger than one LO phonon energy. Nevertheless, the hot exciton energy distribution has changed drastically also in the 15% In dot layer. This implies that the multi phonon-relaxation is actually the most important energy relaxation process under nonresonant excitation. So, when the energy difference is slightly larger than one LO phonon energy, the excitons obtained are still very hot. Turning to the cooling of the hot excitons represented by the reduction of the linewidth with time delay, we found that under the excitation below the barrier band gap, the decay time for the linewidth reduction is much bigger for the quantum dots than for the quantum well. Hence, the transient PL under resonant excitation demonstrates more clearly that the exciton relaxation for the three-dimensionally confined dots is obviously slower than the exciton relaxation in the one-dimensionally confined quantum wells, as predicted in Ref. [4].

Another direct experimental proof of the exciton relaxation within the quantum dots is the long PL rise time which can be measured accurately under nonresonant excitation. The PL rise time shows a two-step behavior (see Fig. 4). The fast component of the PL rise time might be due to the quick cooling of the photo-excited carriers by emitting optical phonons while the slow component, which can be as long as 0.5 ns at the low energy side, is due to the relaxation of hot excitons from higher energy states to lower ones. This long rise time is, however, not so obviously observed in the energetically integrated PL decay curve (Fig. 1B). Therefore it characterizes the energy relaxation within single quantum dots. The rise time determined here is roughly in agreement with the hot exciton relaxation time determined from the time dependence of the half-width and the transition peak energy in the transient PL spectrum in Fig. 2.

In conclusion, we have compared the exciton relaxation in (111)B $Ga_{1-x}In_xAs/GaAs$ quantum wells and self-organized quantum dots. By analyzing the temporal behavior of the linewidth, the transition peak energy

and the PL rise time in the transient PL spectra excited both above and below the barrier band gap, we found that the exciton relaxation in the three-dimensionally confined quantum dots is much slower than in the quantum well as predicted theoretically before [4].

Acknowledgement—JXS would like to thank the Japan Society for the Promotion of Science under No. P98181 for financial support.

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