phys. stat. sol. (b) **224**, No. 2, 487–491 (2001)

Suppressed Relaxation in InGaAs/GaAs Quantum Dots

H. Born[1]), R. Heitz, A. Hoffmann, F. Guffarth, and D. Bimberg

Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany

(Received July 31, 2000; accepted October 2, 2000)

Subject classification: 73.21.La; 78.47+p; 78.55.Cr; 78.67.Hc; S7.12

Suppressed relaxation in strongly confined InGaAs/GaAs quantum dots (QDs) was investigated by resonantly excited time-resolved photoluminescence spectroscopy. Hot exciton recombination and temperature-dependent relaxation are demonstrated supporting multi-phonon processes to dominate the relaxation between low-lying states at low excitation densities.

Despite a decade of experimental and theoretical work carrier/exciton relaxation processes in QDs still remain a poorly understood issue. Relaxation by inelastic phonon scattering is expected to be suppressed by the discrete energy spectrum with energy gaps normally not matching phonon energies or multiples thereof [1, 2], fueling speculations on alternative relaxation processes, like Auger or Coulomb scattering. However, such relaxation processes generally have similar restrictions as long as the involved carriers remain localized or require the presence of additional carriers only available at higher excitation densities.

Available experimental results on high-quality self-organized InAs/GaAs QDs point, however, to relaxation being much faster than recombination processes: At low excitation densities only ground state photoluminescence (PL) is observed and time-resolved PL (TRPL) reveals relaxation on a time scale of some 10 ps [3, 4]. The temperature dependence of the latter as well as the observation of multi-LO-phonon resonances in PL excitation (PLE) spectra [4] support inelastic phonon scattering to be the dominant relaxation mechanism. The enhanced polar exciton–LO-phonon interaction in such strained QDs [5] might account for the observed short relaxation rates.

The purpose of the paper is to investigate for the first time the relaxation in InGaAs/GaAs QDs by resonantly excited temperature-dependent TRPL. The results are compared to PL and PLE studies demonstrating suppressed relaxation at low temperatures. Hot luminescence from the first excited state transition is identified providing new information on relaxation processes in such strained QDs. The results are discussed in view of the structural properties of the QDs.

The investigated InGaAs/GaAs QDs were grown by metal organic chemical vapor deposition as described in detail in [6, 7]. Cross-sectional transmission electron micrographs reveal a rather flat shape with typically 20 nm lateral size and 3 nm height while the quantum dot density of 3×10^{10} cm^{-2} was estimated from plan view images (not shown here). PL spectra of the investigated sample reveal at 7 K and low excitation densities ground state luminescence at 0.999 eV with a high-energy shoulder separated by $\approx$70 meV (Fig. 1). The high-energy shoulder is attributed to hot-exciton recombination from the first excited state transition due to suppressed relaxation. Indeed, the PLE spec-

[1]) Corresponding author; e-mail: harald@physik.tu-berlin.de

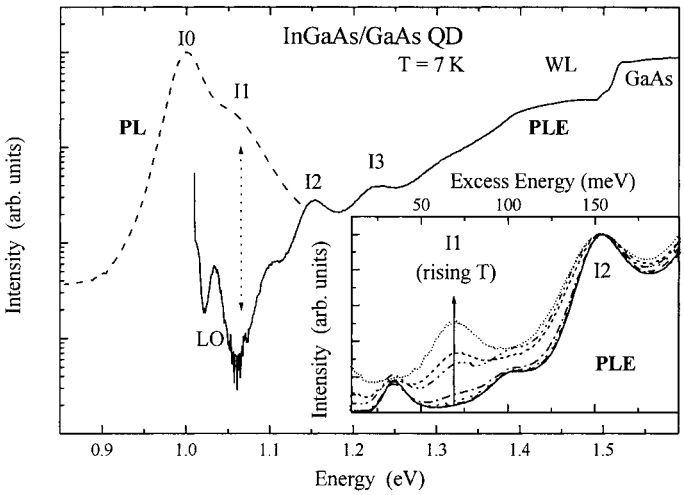

Fig. 1. PL and PLE spectra of InGaAs/GaAs QDs at 7 K. The inset shows PLE spectra for various temperatures recorded at the corresponding ground state transition energies versus the excess energy ($\Delta E = E_{\mathrm{exc}} - E_{\mathrm{det}}$)

trum of the ground state transition shows efficient excitation via the second (I2) and third (I3) excited state transition as well as the wetting layer (WL) and the GaAs barrier but only inefficient excitation via the first excited state transition (I1) leading to a "gap" in the spectrum. However, the excitation via I1 becomes increasingly efficient with increasing temperature as obvious from the inset showing PLE spectra normalized on I2 via the excess energy ($\Delta E = E_{\mathrm{exc}} - E_{\mathrm{det}}$). The normalized excitation efficiency I1/I2 is shown in Fig. 3a as a function of the temperature. The results unambiguously demonstrate slow relaxation between I1 and I0, which accelerates with increasing temperature supporting the notion of inelastic phonon scattering. The identification of I1 as the first excited state transition belonging to the quantum dot ground state I0 was proven by PLE experiments (not shown here). At the detection energies of I0 and I1 we found the same excitation resonances at all temperatures investigated ($T = 7$–300 K) [8].

More detailed information on the dynamical properties is obtained from TRPL measurements detecting the ground state transition (I0) and exciting spectrally sharp ($\sim$1 meV) the first excited state transition (I1). Figure 2 depicts normalized transients for various temperatures showing the rise of the ground state PL to accelerate with increasing temperature. The transients are fitted assuming two exponentials for the rise (τ_{rise}) and decay (τ_{decay}), Fig. 3b. Below 80 K both time constants are virtually identical but become clearly distinguishable at higher temperatures, identifying τ_{decay} and τ_{rise} as lifetimes of the ground (I0) and the first excited (I1) state, respectively. The increasing ground state lifetime is attributed to the thermal population of excited hole states having a lower recombination probability with the ground state electron [9]. More interestingly, the lifetime of the first excited state transition (I1) decreases above $\approx$80 K. In agreement with the PLE results (Fig. 1) the decreasing lifetime is attributed to an increasing relaxation rate with increasing temperature.

Recent TRPL measurements on similar samples exciting resonantly I1 showed the decay of I1 to mirror the rise of I0 at low excitation densities [7], demonstrating single-

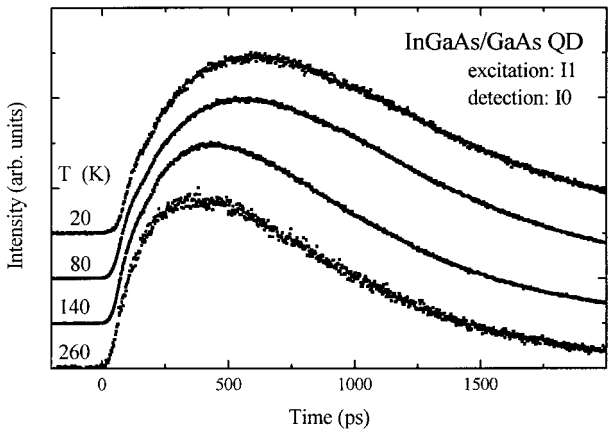

Fig. 2. TRPL of the ground state transition I0 for various temperatures exciting resonantly the first excited state transition I1. The spectra are shifted vertically for clarity.

step relaxation between I1 and I0. Obviously, correlated electron–hole (exciton) relaxation dominates in the investigated self-organized InGaAs/GaAs QDs allowing to describe the PL dynamics in a simple three-level model comprising the vacuum level, I0 and I1, see Fig. 4. The first excited state I1 might decay radiatively (recombination rate w^1_{rad}) to the vacuum level or non-radiatively (relaxation rate w^1_{rel}) to I0. The latter process is assumed to be thermally activated

$$w^1_{\mathrm{rel}}(T) = w_0 \cdot (n_b(T) + b)\,, \qquad n_b(T) = \frac{1}{\exp(\hbar\omega/kT) - 1}\,, \tag{1}$$

with $\hbar\omega$ being the phonon energy and b a constant, bw_0 is the low temperature relaxation rate. The competing radiative recombination and non-radiative relaxation lead to a temperature dependent relaxation yield η,

$$\eta = \frac{w^1_{\mathrm{rel}}}{w^1_{\mathrm{rel}} + w^1_{\mathrm{rad}}} \sim \frac{\mathrm{I1}}{\mathrm{I2}}\,, \tag{2}$$

describing the normalized excitation efficiency (I1/I2, solid line in Fig. 3a). The model reproduces the observed temperature dependence for a phonon energy of 32.5 meV, w_0/w^1_{rad} = 0.08 and b = 0.006. The phonon energy corresponds to the

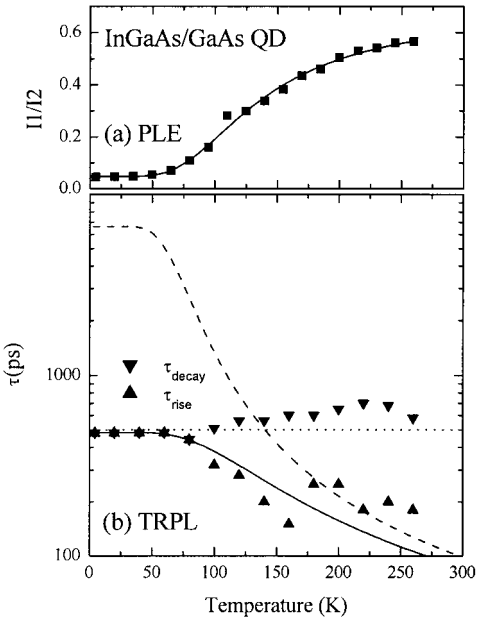

Fig. 3. a) Temperature dependent excitation efficiency I1/I2 of the ground state transition I0 via the first excited state transition I1, and b) the corresponding rise and decay times. Lines present a model as described in the text

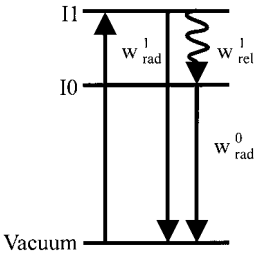

Fig. 4. Schematic three-level model describing the excitation dynamics in the InGaAs/GaAs QDs

QD LO-phonon energy [4, 5]. The model describes as well the lifetime of the first excited state transition (τ_{rise})

$$\frac{1}{\tau_{\text{rise}}} = w^1_{\text{rel}} + w^1_{\text{rad}} \,. \tag{3}$$

The rise time predicted assuming additionally a radiative decay time of 520 ps for the first excited state transition (solid line in Fig. 3b) describes well the experimental data. Obviously the radiative lifetimes of the ground (I0) and first excited (I1) state transitions are very similar as expected from a two-dimensional harmonic oscillator model. The derived temperature-dependent relaxation time τ^1_{rel} is shown as dashed line in Fig. 3b. At He temperatures the relaxation time is estimated to $\approx$7 ns, which decreases to $\approx$100 ps at room temperature. The origin of the finite relaxation probability at low temperatures (described by b) is not clear yet. Note that the relaxation in the present QDs is indeed a multi-phonon process involving at least two LO phonons making a priory assumption on the spontaneous relaxation rate difficult. The observed suppressed relaxation presents, however, an upper limit on the probability of Auger-type relaxation processes. Obviously, such alternative processes are negligible in the present QDs.

We propose the slowed down carrier relaxation observed for the present samples to be closely related to the structural properties of the QDs. The disk-like shape results in rather similar electron and hole wavefunctions and thus in a vanishing polar interaction with LO phonons giving the dominant contribution to the relaxation process. On the contrary, the pyramidal InAs/GaAs QDs [4] lead to significantly different electron and hole wavefunctions which enhance the polar-exciton–LO-phonon interaction [5], accounting for the observed relaxation on a few 10 ps time-scale. Further experiments are necessary to provide detailed information on the effects of the structural properties on the exciton dynamics in self-organized QDs.

In conclusion, our experiments provide detailed insight into the exciton relaxation between the first excited and ground state in large, flat, self-organized QDs. Phonon-related relaxation was experimentally identified by PLE and TRPL to dominate the relaxation between the first excited state and the ground state transition at low carrier densities and low temperatures. A simple model accounting for the temperature-dependent variation of the phonon population explains the observed behavior.

Acknowledgement This work was supported by the Deutsche Forschungsgemeinschaft in the framework of Sonderforschungsbereich 296.

References

[1] H. Benisty, C. M. Sotomayor-Torrès, and C. Weisbuch, Phys. Rev. B **44**, 10945 (1991).
[2] T. Inoshita and H. Sakaki, Phys. Rev. B **46**, 7260 (1992).
[3] B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11532 (1996).
[4] R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kopev, and Zh. I. Alferov, Phys. Rev. B **56**, 10435 (1997).
[5] R. Heitz, I. Mukhametzhanov, O. Stier, A. Madhukar, and D. Bimberg, Phys. Rev. Lett. **83**, 4654 (1999).
[6] F. Heinrichsfdorff, M. Grundmann, O. Stier, A. Krost, and D. Bimberg, J. Cryst. Growth **195**, 540 (1998).
[7] R. Heitz, H. Born, T. Lüttgert, A. Hoffmann, and D. Bimberg, phys. stat. sol. (b) **221**, 65 (2000).
[8] R. Heitz et al., to be published
[9] R. Heitz, I. Mukhametzhanov, A. Madhukar, A. Hoffmann, and D. Bimberg, J. Electron. Mater. **28**, 520 (1999).