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Resonantly Excited Time-Resolved Photoluminescence Study of Self-Organized InGaAs/GaAs Quantum Dots

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Relaxation and capture processes in self-organized InGaAs/GaAs quantum dots (QDs) are studied by time-resolved photoluminescence spectroscopy under resonant excitation. Relaxation between the lowest confined states is shown to be strongly suppressed indicating a pronounced phonon bottleneck effect. In neutral QDs correlated relaxation processes dominate: exciton relaxation between low lying states and Auger-type processes for higher excited states, the latter providing an efficient means to populate the ground state upon non-resonant excitation. The relaxation-limited dynamics is attributed to the disk-like shape of the investigated QDs, suggesting the control of structural properties as possible pathway to device optimization.

The formation of nanoscale coherent islands in highly strained semiconductor epitaxy has been extensively studied as nature's pathway to optically active quantum dots (QDs) [1]. In spite of the limitations in controlling the density, size, and shape such Stranski-Krastanow QDs have been successfully employed in devices [2–5]. Due to the defect-free nature such nm-scale strained QDs are ideal model systems for the study of capture and relaxation processes in strongly confining QDs, which are both of basic physics interest and critical for design and performance of devices.

The discrete density of states was proposed to impose severe constraints on inelastic phonon scattering and, therefore, on corresponding relaxation processes [6, 7], stimulating debate on alternative relaxation mechanisms, like Coulomb scattering [8] or Auger-recombination [9]. The observation of multi-LO-phonon resonances in photoluminescence excitation (PLE) experiments for self-organized InAs/GaAs QDs [10] suggested inelastic phonon scattering as dominant relaxation process. Time-resolved photoluminescence (TRPL) measurements for long wavelength III–V QDs under non-resonant excitation revealed fast rise times of some 10 ps [10–12], but provided only limited information on the details of the capture and relaxation cascade.

In this contribution, we report on photoluminescence (PL), PLE and TRPL measurements on self-organized InGaAs/GaAs QDs using spectrally sharp resonant excitation. We demonstrate strongly inhibited relaxation processes for the investigated disk-like QDs leading to relaxation-limited dynamics and hot exciton luminescence. These observations are in striking contrast to the recombination-limited dynamics observed for pyramid-like InAs/GaAs QDs [10], suggesting a pronounced influence of the shape on the interaction with photons and phonons. The resonantly excited TRPL results provide detailed information on the carrier capture and relaxation processes.

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The investigated InGaAs/GaAs QD samples were grown by metalorganic chemical vapor phase epitaxy on GaAs as described in detail in Ref. [13]. The resulting QDs have a high In concentration and rather disk-like structure [13, 14] with typical average width and height of 20 and 3 nm, respectively. For the PL, PLE, and TRPL measurements the samples were mounted in a continuous-flow He-cryostat or a He-immersion cryostat, providing temperatures between 1.8 and 300 K. Time-integrated spectra were recorded with a tungsten lamp dispersed by a double-grating monochromator as excitation source and a cooled Ge-diode for detection. The TRPL measurements were excited by spectrally narrow (<1 meV) ps-pulses of a Ti:sapphire laser or an optical parametric oscillator system, depending on the wavelength region of interest. The luminescence was dispersed by a subtractive double-grating monochromator and detected with a multi-channel plate photomultiplier with an S1-cathode in photon-counting mode. The system response with a full width at half maximum (FWHM) of ≈20 ps was taken into account in the analysis of the transients using convolution techniques.

The time-integrated optical properties of a typical sample are summarized in Fig. 1. In low-temperature PL the narrow (FWHM = 35 meV) ground state transition peak at 0.9999 eV is accompanied by a weak (≈17%) high-energy shoulder. In the following we will show, that this high-energy shoulder is likely to be the result of hot exciton recombination from the first excited state as a result of suppressed relaxation in the investigated QDs. A first indication of such relaxation-limited dynamics is given by the PLE spectrum detected on the maximum of the PL peak (Fig. 1). The first strong excitation resonance is observed 151 meV above the ground state energy, which is commonly assigned to the second excited state transition (labeled I2) in a two-dimensional harmonic oscillator model [15]. At higher temperatures (200 K) the PL spectrum is shifted by 38 meV towards lower energies and a prominent excitation resonance is observed 71 meV above the ground state transition (dotted spectra). This resonance is completely missing at 7 K. These results point to a strongly suppressed relaxation in the investigated QDs at low temperatures, which prevents population of the ground state exciting resonantly the first excited state transition. Note that this is obviously not an universal property of self-organized InGaAs/GaAs QDs. For example, for approximately pyramidal QDs no excited state PL is observable at low excitation densities and

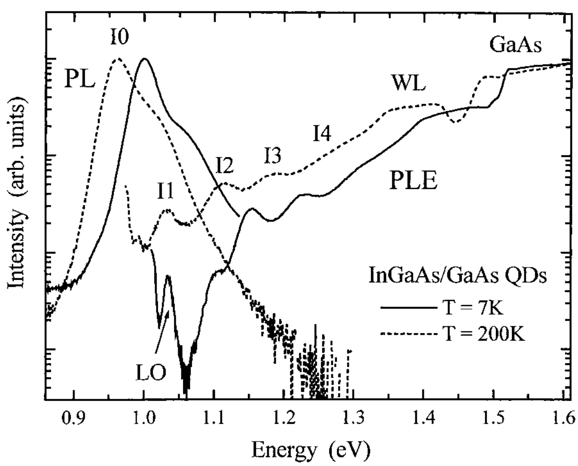

Fig. 1. PL and PLE spectra of a typical InGaAs QD sample. Solid and dotted spectra were recorded at 7 and 200 K, respectively

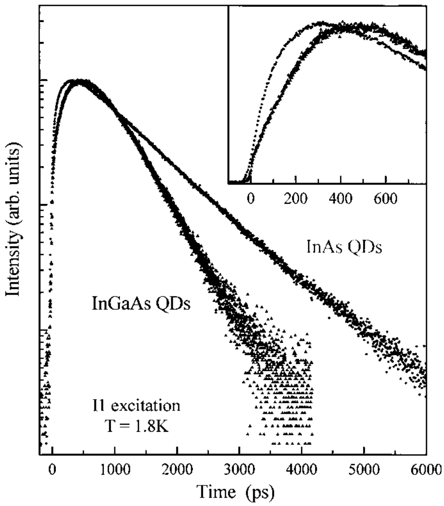

Fig. 2. Transients of the ground state PL (I0) of self-organized InGaAs (block triangles) and InAs (gray dots) QDs excited resonantly in the first excited state transition (I0)

low temperatures [10, 16] and PLE spectra show the relative intensities of the near-resonant excitation resonances to be independent on temperature [17].

Figure 2 provides direct evidence for suppressed relaxation between strongly confined states. The PL transient of the InGaAs QDs (black triangles) recorded for resonant excitation of the first excited state transition (I1) shows a slow rise process leading to a PL maximum only after $\approx$ 500 ps. The transient is fitted assuming one exponential rise and one exponential decay process, yielding almost identical time constants of 370 and 375 ps, respectively. The time constants clearly demonstrate suppressed carrier relaxation between the first excited state and the ground state in the investigated QDs. Note, that the rise time of the ground state PL presents only a lower limit for the actual relaxation time due to the competition with direct radiative recombination of the excited state. The low excitation efficiency (<10%) observed in the low-temperature PLE spectra (Fig. 1) suggests that the observed rise time is indeed close to the excited state lifetime and that the relaxation time is of the order of some ns.

Figure 2 depicts for comparison also a corresponding transient for pyramid-like InAs QDs, which rises much faster and decays slower. A bi-exponential fit yields 150 and 920 ps, respectively. TRPL measurments of such QDs have been reported in detail recently [10, 12, 17]. Indeed, as discussed below the particular shape of the QDs has a tremendous influence on the relaxation and recombination processes, obviously allowing to tune the dynamics in such QDs from a relaxation to a recombination dominated situation. In the latter case hot exciton recombination occurs in the low temperature and low excitation density regime.

Theoretical predictions of inelastic phonon scattering in QDs are so far limited to the case of single carriers in charged QDs with idealized potentials allowing for analytical wavefunctions [6, 7]. However, the situation might be more complex and qualitatively different in neutral QDs following, e.g., resonant optical excitation. In this case, it is a priori not clear if the electron and hole will relax independently, leading to all kinds of intermediate states, or if correlated processes like exciton relaxation or Auger-type processes are more favorable. Indeed the observation of muti-LO-phonon resonances in PLE spectra [10] suggested exciton relaxation to be dominant in resonantly excited neutral QDs. Figure 3 compares PL transients of the ground (I0) and first excited state (I1) transitions for resonant excitation. Figure 3a shows the ground state PL under strictly resonant excitation. Despite the strong stray light superimposing the luminescence signal at short times, an exponential decay with a time constant of $\approx$340 ps is observed for longer delays, which is obviously the radiative lifetime of the ground state.

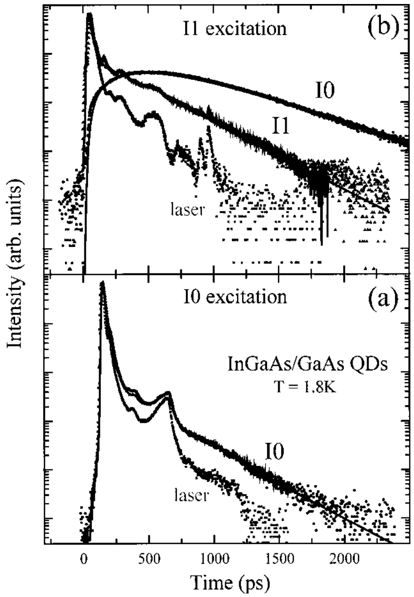

Fig. 3. Resonantly excited PL transients for InGaAs QDs. a) The ground state (I0) PL for strictly resonant excitation. b) The ground state (I0) and first excited sate (I1) PL upon resonant excitation of the first excited state transition. The gray transients represent the system response and solid lines present bi-exponential fits as described in the text

The resonant signal from the ground state transition could be observed in a right-angle geometry due to the inhomogeneity of the QD ensemble and the finite bandwidth ($\approx$1 meV) of the excitation. Exciting the first excited state transition I1 (Fig. 3b) we find again the slow rise of the ground state PL corresponding to a time constant of 370 ps. Simultaneously it is possible to monitor the resonant hot PL of the first excited state transition I1 itself giving a strong signal. The decay of the PL of the first excited state transition mirrors exactly the rise of the ground state PL. This is only possible in case of a single step relaxation process demonstrating that actually correlated electron–hole (exciton) relaxation takes place. The successive relaxation of electrons and holes from their respective excited states to the ground state would lead to an intermediate state with a recombination energy between that of the ground and first excited state transitions. The radiative recombination of such an intermediate state would not be detected in our spectrally resolved measurements, but would cause a mismatch between the decrease of I1 and the rise of I0.

The time-integrated spectra (Fig. 1) suggest that for the investigated samples relaxation in the QDs is indeed not a cascade process but that, e.g., the second excited state can efficiently relax directly to the ground state bypassing the first excited state transition. The low intensity ($\approx$17%) of the first excited state transition I1 in the non-resonantly excited PL spectrum supports the fact that the fast majority of excitons excited in the GaAs barrier bypasses the first excited state in the capture process. Figure 4 compares transients of the ground state transition I0 for excitation of the first and second excited state transition as well as the GaAs barrier. Interestingly, the observed rise time decreases from 370 and 350 ps to 65 ps, respectively. This rather surprising result excludes the possibility of a cascade capture and relaxation for the investigated QDs. Obviously, there exist efficient shunt processes for the higher excited states. We propose Auger-type processes [18] as possible alternative relaxation means based on simple energy considerations. An exciton generated in the first (I1) or second (I2) excited state is unlikely to experience Auger-type relaxation. The respective relaxation energies of 71 and 151 meV are insufficient to excite one carrier into the wetting layer continuum ($\approx$1.38 eV) and the chance to find a suitable localized excited energy level is low. On the contrary, excitons in higher excited states or the WL could experience Auger-type relaxation as recently proposed by Ferraira et al. [18]. The assumption of Auger-type relaxation processes, thus, successfully explains

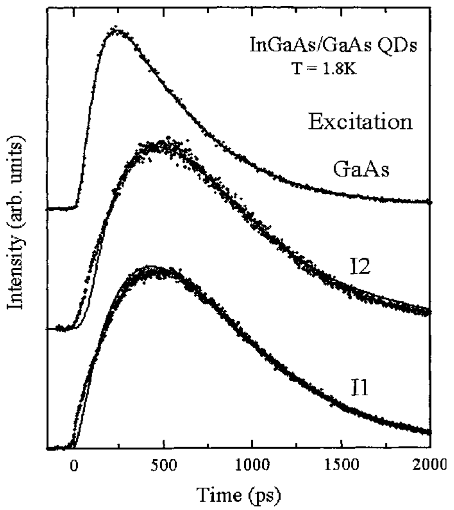

Fig. 4. Transients of the ground state PL (I0) of self-organized InGaAs QDs for resonant excitation of the first (I1) and second (I2) excited state transitions as well as non-resonant excitation of the GaAs barrier. Dots give the experimental results and solid lines represent bi-exponential fits as described in the text

the complex dynamical behavior observed for resonant excitation.

The present time-resolved data are strikingly different to our previous results for pyramid-like InAs/GaAs QDs, which revealed PL rise times of some ten ps and decay times of the order of 1ns both for non-resonant and resonant excitation [10, 12, 17]. These times are significantly shorter and longer, respectively, than for the present disk-like InGaAs QDs, indicating a pronounced effect of the structural properties of the QDs on the relaxation and recombination processes.

In the strong confinement limit the radiative lifetime is determined by the *overlap* integral of the electron and hole wavefunctions [19] whereas the Fröhlich-type coupling of the exciton to LO-phonons, providing the fastest relaxation channel in higher-dimensional systems, is proportional to the local charge density and, thus, the squared absolute value of the Fourier transformed *difference* of the probability densities of the electron and hole wavefunctions [16]. Therefore, both the radiative decay and the relaxation times are expected to depend sensitively on the actual shape of the wavefunctions and therewith on the symmetry of the confining potential. Indeed, the most striking difference between the investigated InGaAs QDs and previously investigated InAs QDs is the different shape. For pyramidal QDs the shear strain induced piezoelectric potential gives rise to a strong asymmetry of the electron and hole wavefunctions as discussed in detail in Ref. [19]. The electron ground state is predominantly s-like in the center whereas the hole ground state is clearly p-type with the highest probability in the (1–10) corners of the pyramid. This asymmetry provides for a comparatively small overlap of the wavefunctions and, thus for a low radiative recombination and a high relaxation probability. The piezoelectric potential is, however, negligible for disk-like QDs leading for both the electron and hole to s-type wavefunctions in the center of the pyramid [20]. The similarity of the electron and hole wavefunctions in the latter case leads to a larger overlap, i.e. a higher radiative recombination probability and a low relaxation probability. These first qualitative insights into the influence of the shape of small strained QDs on the carrier dynamics are in good qualitative agreement with the experimental observations supporting the notion of dominant inelastic phonon scattering for the first excited state.

In conclusion, the present TRPL investigations of InGaAs/GaAs QDs under resonant excitation provide detailed insights into the carrier relaxation processes and into the intricacies of the carrier capture and relaxation cascade in neutral self-organized QDs. A pronounced phonon bottleneck is observed to cause hot exciton recombination. In particular, the results unambiguously demonstrate correlated exciton relaxation for the lowest excited states, which likely proceeds by inelastic phonon scattering, as well as additional Auger-type relaxation processes for higher-excited states, providing for fast population of the ground state after non-resonant excitation. Finally, in such highly strained piezoelectric systems the QD shape strongly influences the carrier dynamics allowing to realize both relaxation-limited and recombination-limited regimes. The possibility to design the carrier dynamics opens up far reaching optimization potential for the use of self-organized QDs in device.

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