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Shape-dependent phonon bottleneck in InGaAs/GaAs quantum dots

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Abstract Suppressed relaxation is demonstrated for strongly confined exciton in InGaAs/GaAs quantum dots indicating a phonon bottleneck. Hot exciton recombination and thermally activated relaxation support correlated electron-hole relaxation by multi-phonon processes.

Despite a decade of experimental and theoretical work carrier/exciton relaxation processes in self-organized quantum dots (QDs) remain controversial. The limited chance to achieve energy conservation in the discrete eigenstate spectrum of QDs has led to predictions of a phonon bottleneck [1,2], preventing the population of the ground state upon non-resonant excitation. Nevertheless, intense ground state luminescence stimulated speculations on alternate relaxation mechanisms like Coulomb scattering, Auger-type processes or defect-mediated relaxation. Additionally, a break down of Fermi's golden rule in the strong coupling regime was proposed to account for fast relaxation [3,4].

Little attention has been paid to exciton relaxation yet, though such processes are usually probed in optical interband experiments. The simultaneous presence of an electron and hole leads to correlated exciton relaxation [5]. For exaple for pyramid-like InAs/GaAs QDs relaxation is 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 10ps [6,7]. The temperature dependence of the latter as well as the observation of multi-LO-phonon resonances in PL excitation (PLE) spectra support inelastic phonon scattering to be the dominant relaxation mechanism. The enhanced polar exciton-LO-phonon interaction in such strained QDs [8] might account for the observed short relaxation rates.

Here we report on temperature-dependent PLE and TRPL experiments on disk-like InGaAs/GaAs QDs. Hot exciton luminescence and slow relaxation from the first excited state indicate a pronounced phonon bottleneck. The intradot exciton relaxation probability is proposed to depend strongly on the QD shape.

The investigated InGaAs/GaAs QDs were grown by metal organic chemical vapor deposition as described in detail in [5,9]. Cross-section transmission electron micrographs reveal disk-like QDs being ~20nm wide and ~3nm high. PL spectra of the investigated sample reveal at 7K and low excitation densities ground state luminescence at 0.999eV with a high-energy shoulder separated by ~69meV (Fig. 1). The high-energy shoulder is attributed to hot-exciton recombination from the first excited state transition being, thus, the finger print of suppressed relaxation. The PLE spectrum demonstrate efficient population of the ground state exciting resonantly the second (I2) and third (I3) excited state transition as well as non-resonantly the wetting layer (WL) and the GaAs barrier. The first excited state (I1) absorption does, however, not populate the ground state causing a "gap" in the PLE spectrum. With increasing temperature I1 appears in the PLE spectra (inset) suggesting thermally activated relaxation. The normalized excitation efficiency I1/I3 is shown in Fig. 3(a) via the temperature.

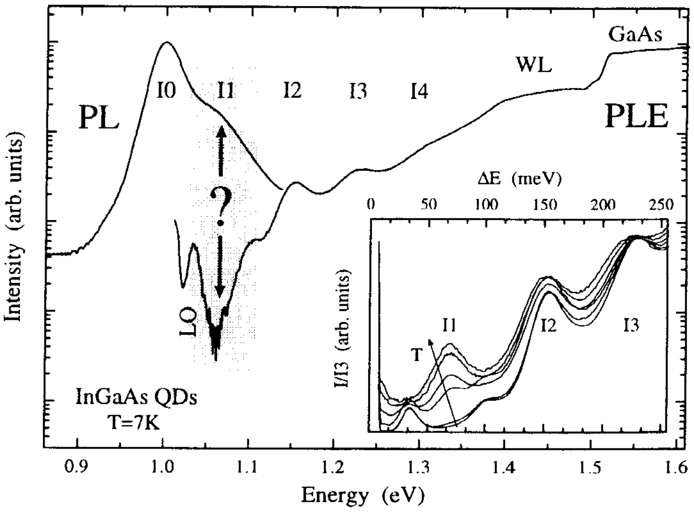

Fig. 1 PL and PLE spectra of InGaAs/GaAs QDs at 7K. The inset shows PLE spectra for various temperatures recorded at respective maximum of the ground state emission I0 via the excess energy ($\Delta E = E_{exc} - E_{det}$).

The exciton dynamics are probed in TRPL measurements detecting I0 and exciting 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 convoluting the system response with exponentials for the rise (τ_{rise}) and decay (τ_{decay}), respectively. The results are shown in Fig. 3(b). Below ~80K both time constants are virtually identical but become distinguishable at higher temperatures, identifying τ_{decay} and τ_{rise} as lifetimes of I0 and I1, respectively. The decay of I0 slows down with increasing temperature due to the thermal population of excited hole states having a lower recombination probability with the electron ground state [10]. Finally, carrier

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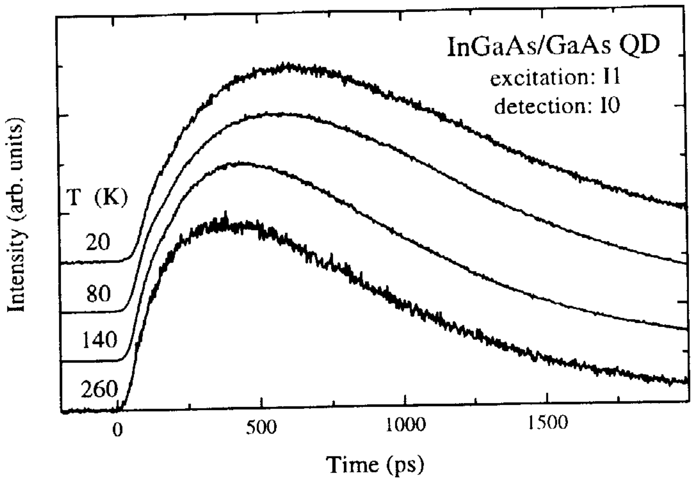

Fig. 2 Transients of emission I0 for various temperatures exciting via the I1 absorption. The spectra are normalized and shifted vertically for clarity.

escape reduces the I0 life time above ~230K. The rise time, i.e. the life time of I1, decreases above ~80K, indicating thermally activated relaxation.

The PLE and TRPL results can be described by the exciton dynamics in a simple three-level model comprising the vacuum level, I0 and I1, inset of Fig. 3(b). The first excited state I1 might decay radiatively (τ_{rad}^{1}) to the vacuum level or relax (τ_{rel}^{1}) to I0. Relaxation is assumed to involve a constant (b) and a thermally activated (E_a) contribution. The competition of recombination and relaxation lead to a temperature-dependent relaxation yield η being proportional to the normalized excitation efficiency I1/I3 in Fig. 3(a). A fit (solid line) yields an activation energy E_a of 32.5meV, corresponding to the QD LO phonon energy [7,8], and b= 0.0037. The parameters describes also the lifetime of I1 (τ_{rise}) with a radiative decay time τ_{rad}^{1} of 515ps (solid line in Fig. 3(b)). Obviously, the radiative lifetimes of I0 (480ps) and I1 are very similar. The derived temperature-dependent relaxation time τ_{rel}^{1} is shown as dashed line in Fig. 3(b). At He temperatures the relaxation time is estimated to ~ 7.8ns, which decreases to ~ 70ps at 300K. The origin of the finite relaxation probability at low temperatures (described by b) is not clear yet. Note that the relaxation is a multi-phonon process involving at least two LO phonons making a priori assumptions on the spontaneous relaxation rate difficult. The observed suppressed relaxation presents, however, an upper limit on the probability of alternative relaxation processes, e.g. Auger-type processes. Obviously, such processes are negligible for the investigated QDs.

We propose the slowed down carrier relaxation to be related to the QD shape. The disk-like shape results in rather similar electron and hole wavefunctions and, thus, in a vanishing polar interaction of the exciton with LO-phonons giving the dominant contribution to inelastic phonon scattering. On the contrary, the pronounced asymmetry of electron and hole wavefunctions in pyramidal InAs/GaAs QDs enhances the polar exciton-LO-phonon interaction [8], accounting for the observed relaxation on a few 10ps time-scale [7].

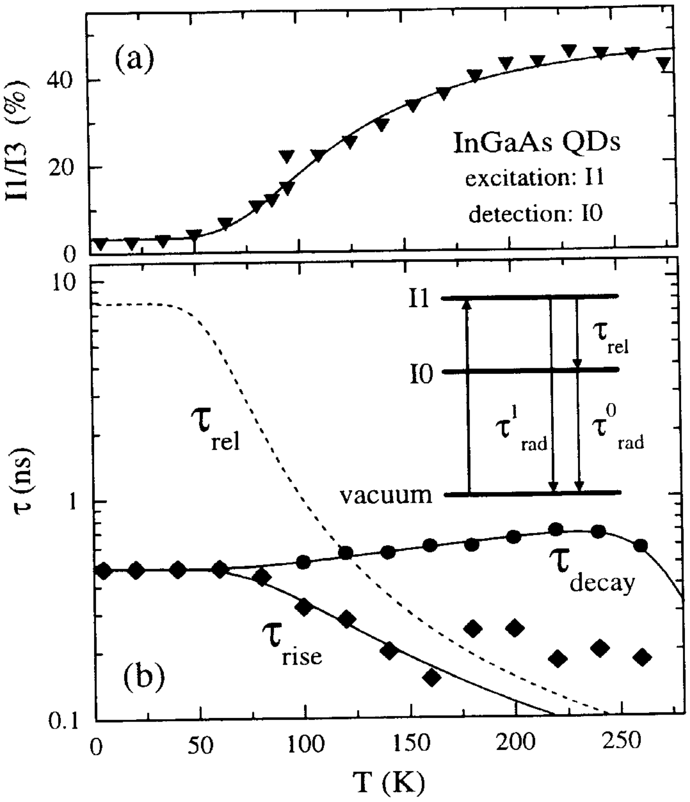

Fig. 3 Temperature-dependent (a) excitation yield (I1/I2) of emission I0 via the I1 absorption and (b) the corresponding rise and decay times. Lines present a fit assuming the three level model shown as inset.

In conclusion, a pronounced phonon bottleneck in disk-like InGaAs QDs is observed due to drastically slowed down exciton relaxation. The low probability of inelastic phonon scattering is attributed to the vanishing Fröhlich interaction in such QDs.

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