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Carrier Capture and Relaxation Processes in InAs/GaAs Quantum Dots

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Excited states and energy relaxation process are studied for self-organized InAs/GaAs quantum dots (QDs). Depending on the sample, excited state transitions or multi-LO-phonon resonances are found in photoluminescence excitation (PLE) spectra, revealing the size-dependent excited state splitting or the carrier relaxation mechanism, respectively. Time-resolved photoluminescence (PL) results indicate sample-dependent non-radiative recombination, leading to a model for the observed PLE behavior, analogous to hot carrier relaxation in higher dimensional systems. Carrier relaxation in the self-organized InAs/GaAs QDs proceeds by multi-LO-phonon scattering on a 40 ps time scale, much shorter than radiative ($>$ 500 ps) and non-radiative ($>$ 100 ps) recombination times, accounting for the absence of a phonon bottleneck effect in PL spectra.

1. Introduction

The spontaneous formation of coherent semiconductor islands in highly strained epitaxy has attracted interest as nature's way to generate nm-scale quantum dots (QDs). The defect-free coherent nature of the islands [1] has enabled the demonstration of QD-based injection lasers [2,3]. However, hot carrier relaxation processes, being crucial for device performance, are not well understood yet [4–13].

The discrete atomic-like energy spectrum of QDs limits inelastic phonon scattering slowing down carrier relaxation [4,5]. Indeed, inter-level scattering times of the order of some 10 ps or longer have been inferred from time-resolved photoluminescence (TRPL) results [2,6,7] or rate equation models [8]. The long

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relaxation-limited lifetime of the excited states makes the QDs sensitive to competing recombination processes, which ultimately might suppress ground state photoluminescence (PL), the so-called phonon bottleneck effect [4]. PL excitation (PLE) spectra of InAs QDs are reported to show excited state transitions [9,10] or phonon resonances attributed to the carrier relaxation dynamics [7,11,12] or resonant enhancement [13].

In this paper we report on PLE and TRPL studies for self-organized InAs/GaAs QDs with sub-state splittings exceeding LO phonon energies in the InGaAs system. Two types of samples distinguished by their respective characteristic PLE and TRPL behavior are compared, yielding insight into the carrier relaxation processes and the relaxation and recombination dynamics.

2. Samples and experimental setup

The InAs/GaAs QD samples were grown by solid source molecular beam epitaxy (MBE) on GaAs(001) substrate. For type A samples multiple 1.74 ML InAs layers were deposited at 500° C and separated as well as capped with GaAs at 400° C using migration enhanced epitaxy as described in Refs. [9,14]. The 20 or 36 ML spacers result in vertical ordering ($> 95\%$) of the InAs islands [14]. For type B samples a single 12 Å InAs layer was deposited and capped with 10 nm GaAs at 480° C as described in Refs. [2,11].

PLE measurements were performed in a continuous-flow He-cryostat using a tungsten lamp dispersed by a 0.27 m double-grating monochromator as tunable light source. TRPL measurements were performed in superfluid He with a tunable Ti-Sapphire laser providing 150 fs pulses and a subtractive double-grating monochromator in combination with an infrared-enhanced streak camera. Time constants down to 5 ps could be resolved taking into account the system response function.

3. Experimental results

Fig.1(a) shows PL (inset) and site-selective PLE spectra for a type A sample with 5 InAs layers and 36 ML spacers. The excess excitation energy $\Delta E = E_{exc} - E_{det}$ (E_i being the excitation and detection energies) of the dominating PLE resonance (FWHM=25 meV) decreases from 96 to 70 meV with decreasing detection energy, reflecting the decreasing excited state splitting with increasing QD size. High density PL spectra (not shown) reveal the first excited state transition 79 meV above the ground state transition in agreement with the PLE results. A comparison of the excited state splitting and its size-dependence with numerical results for pyramidal InAs QDs [10,15] supports the assignment to the

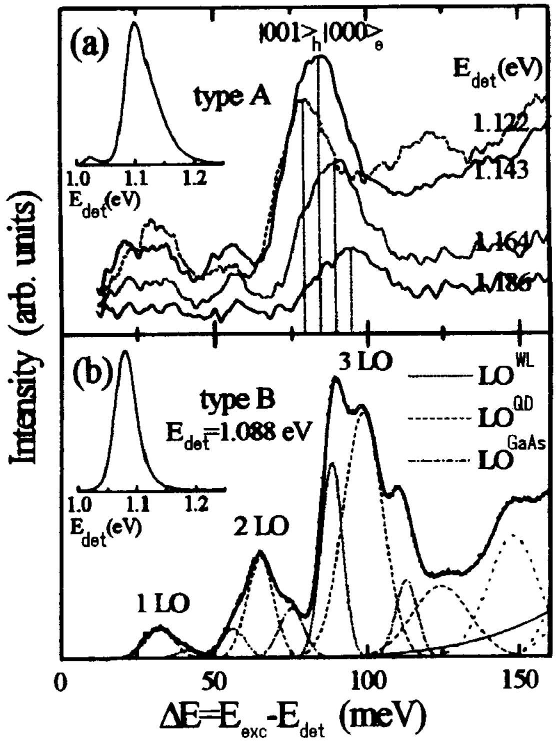

Figure 1. PLE spectra of a 5 layer type A sample with 36 ML spacers (a) and a single layer type B sample (b). The insets show PL spectra for GaAs barrier excitation.

transition between the $|001\rangle_h$ excited hole state and the $|000\rangle_e$ electron ground state (labeling of the electronic states is taken from Ref. [15]). Spectra excited with a tunable Ti-Sapphire laser reveal Raman scattering in the energy region below 40 meV.

PLE spectra of the type B sample reveal richer and better resolved structure as shown in Fig.1(b) together with the corresponding PL spectrum (inset). Starting from the detection energy a series of line groups evolves with the third one centered near 100 meV dominating the spectrum. A threefold substructure is resolved for the 60 (2 LO) and 100 (3 LO) meV line groups. A line shape fit with Gaussians shows that the various excitation resonances with FWHMs between 7 and 14 meV can be explained as multi-phonon replicas with phonon energies of (29.6 ± 0.5), (32.6 ± 0.5) and (37.6 ± 0.5) meV. Multiples but no combinations of the phonon modes are resolved. GaAs Raman scattering contributes to the 1 LO peak [11]. Fig.2 shows a contour plot of the dependence of the PL intensity in the type B sample on the detection wavelength and ΔE.

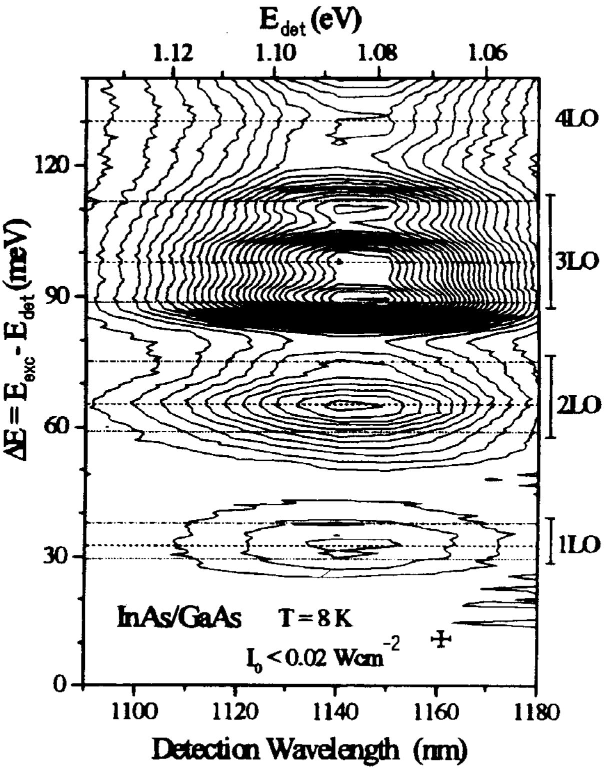

Figure 2. Contour plot of the QD PL intensity as a function of the detection wavelength and the excess excitation energy DE for the type B sample. The horizontal lines represent multiples of WL, QD and barrier LO phonon energies.

The peak energies of the various excitation resonances are independent of the detection energy, although the relative intensities vary slightly. Multiples of the three phonon energies determined in line shape fits are indicated by horizontal lines. Based on the energies the three observed phonon modes are assigned to the InAs wetting layer (WL), the InAs QDs and the strained GaAs barrier near the QDs [7]. The phonon replicas are local probes of the strain in and around the overgrown InAs islands supporting the coherent nature of the optically active QDs. The large FWHM of the PLE resonances (> 7 meV) might result from weakened wavevector selection rules and/or higher order processes involving LA phonons [5].

The different PLE behavior observed for type A and B samples (Fig.1) is determined by the carrier relaxation and recombination dynamics. Fig.3 shows PL transients recorded for excitation densities (I_0), at which the QDs are initially completely saturated. The transients show an almost constant intensity as long as PL from higher energy states is observed. Carrier relaxation from higher energy states keeps the population of lower ones constant. At a given time only PL from the highest populated level will decay with a time constant given by the sum of the recombination rates of all carriers in the QD. Figs.3 (a) and (b) show that the carrier density in the QDs decays much slower in the type A sample causing saturation of the $|001\rangle$ and $|002\rangle$ excited hole states, which is not observed for the type B sample. Multi-exponential fits yield for the type A (B) sample decay times (τ_{decay}) of 480 (95) ps for the first excited $|001\rangle$ state, 180 (42) ps for the second excited $|002\rangle$ state, and 15 (< 5) ps for the WL. A fit of the PL transients for the type A sample using master equations for micro-states [16] yields a radiative lifetime of 1.55 ns for the $|001\rangle$ excited hole state in good agreement with the "forbidden" character of the $|001\rangle_h|001\rangle_e$ transition [15]. The faster decay of excited state PL in the type B sample indicates an extrinsic non-radiative recombination channel with a time constant of about 120 ps for the $|001\rangle$ hole state. Since the PLE results show the optically active InAs islands to be coherent, we propose energy transfer to deep defects in the vicinity of the InAs islands.

In higher dimensional systems multi-LO-phonon resonances in PLE spectra are known to result from hot carrier relaxation when competing non-radiative recombination allows only the most efficient relaxation processes to populate the luminescent state [17]. For QDs having a discrete density of states the inhomogeneous ensemble broadening replaces the spatial dispersion in higher dimensional systems. The non-radiative recombination in the type B sample allows only QDs with rapid carrier relaxation to contribute to the PLE signal. Fig.1(b) demonstrates a resonance condition ($\Delta E = n \cdot \hbar\omega_{LO}$, with n being an integer and $\hbar\omega_{LO}$ being the LO phonon energy) for carrier relaxation in small self-assembled QDs, indicating inelastic multiple-LO phonon scattering to bridge the energy gap between the discrete states. The coupling to various LO modes and the FWHM of the multi-LO-phonon resonances establishes wide energy windows for efficient carrier relaxation. For the type A sample, the lower non-radiative recombination rate allows carrier relaxation also for out-of-resonance QDs. In this case the PLE spectra resemble the density of states and show the excited state transition (Fig.1(a)). In fact, the excited state PLE resonance is strongest for $\Delta E \approx 83$ meV, corresponding to a minimum in the multi-LO-phonon relaxation probability. The results demonstrate the extrinsic nature of the competing recombination channel.

The FWHM (25 meV) of the excited state PLE resonance in the type A sample is attributed to inhomogeneous excited state broadening for a fixed ground

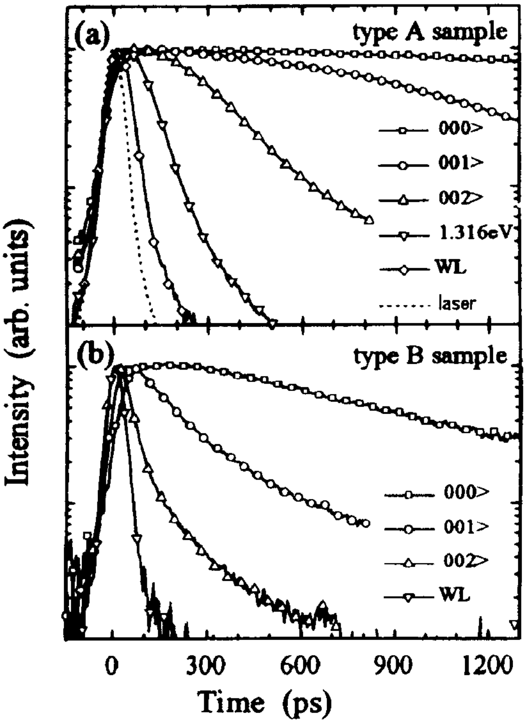

Figure 3. High density transients of the ground and excited QD transition PL peaks for a 5 layer type A sample with 20 ML spacers (a) and a type B sample (b).

state transition energy, resulting from variations of the island shape. Indeed PLE results suggest that in single layer samples the inhomogeneous excited state broadening is even larger and that the vertical self-organization of the islands improves the shape uniformity [10]. The average excited state splitting in the type B sample is also in the 90 meV region accounting for the dominance of the 3 LO resonances in the PLE spectra.

The ground state life time and carrier relaxation rate are determined from low excitation density PL transients for various detection and excitation energies. The transients are generally well fitted by two exponentials (full line in Fig.4(a)), describing the PL rise and decay, respectively. The ground state decay time for type A samples is between 500 and 800 ps and practically independent of the detection energy (i.e. QD size), Fig.4(b), as predicted for the strong confinement regime [18]. The ground state decay time is (1070 ± 100) ps for large QDs in the type B sample and decreases to 500 ps for smaller ones. The non-radiative recombination might influence the ground state transition of the smaller QDs. The present results are in good agreement with previously reported decay times

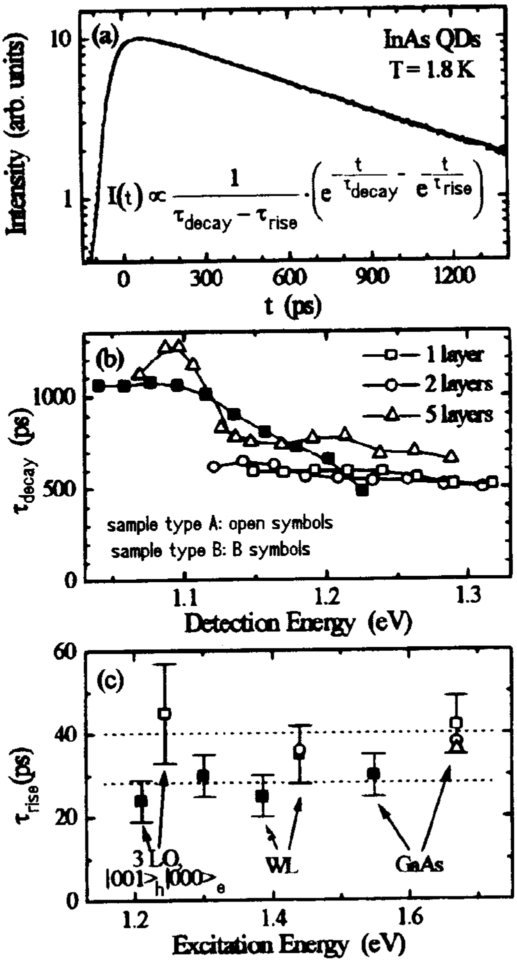

Figure 4. Low excitation density PL transient for a 2 layer type A sample with 36 ML spacer (a), dots represent the experimental results and the full line represents a two-exponential fit. Decay (b) and rise (c) times for type A (open symbols) and B (solid symbols) samples.

[2,19] and suggest that the decay time might depend on the QD shape.

The photoluminescence rise time (τ_{rise}) is independent of the chosen excitation energy/process, being (40±5) and (28±5) ps for type A and B samples, respectively (Fig.3(c)). The same transients are observed exciting non-resonantly the GaAs barrier or the WL or resonantly the $|001\rangle_h|000\rangle_e$ excited state transition, indicating the $|001\rangle$ hole state to be the bottleneck in the capture and relaxation cascade determining τ_{rise}. Taking into account the non-radiative recombination observed for the type B sample the multi-LO-phonon relaxation time of the $|001\rangle_h$ state is (40 ± 5) ps for both sample types. The majority of

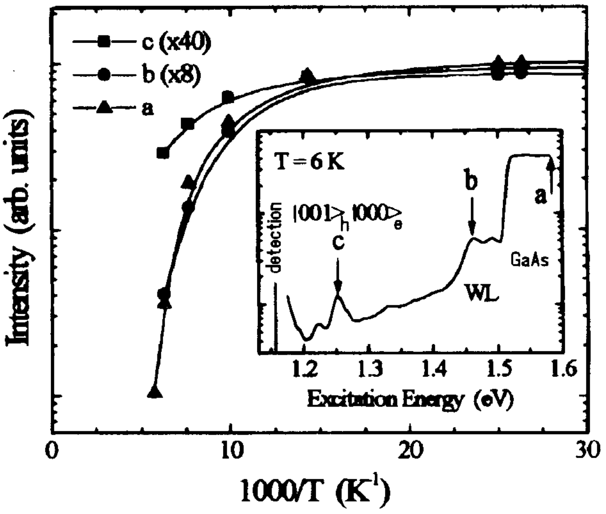

Figure 5. Temperature dependence of the QD PL intensity in the 2 layer sample with 36 ML spacer for excitation in the GaAs barrier (a), in the InAs WL (b) and via the excited $|001\rangle$ hole state (c). The intensities are evaluated from PLE spectra detected at the maximum of the temperature dependent QD PL peak as indicated in the inset.

QDs in the investigated samples is in resonance with 3 LO processes (Figs.1 and 2). For larger QDs with an excited state splitting in resonance with lower-order 2 or 1 LO processes, the inelastic phonon scattering is expected to be more efficient. Fig.5 shows the temperature dependence of the QD PL intensity in a 2 layer type A sample with a 36 ML spacer for different excitation processes. For non-resonant GaAs and WL excitation the intensity decreases above 80 K. For resonant excitation in the $|001\rangle_h|001\rangle_e$ transition, however, the intensity is stable up to 160 K, suggesting thermal quenching of the carrier capture cross section to cause the intensity decrease around 80K observed for non-resonant excitation.

4. Conclusion

The observation of multi-LO-phonon resonances in PLE spectra of small InAs/GaAs QDs is an unambiguous proof for inelastic phonon scattering to be the dominant intra-dot relaxation process. The $|001\rangle$ hole state is found to act as the relaxation bottleneck emphasizing the crucial role of slowed-down carrier relaxation in small QDs. Carrier cooling is about two orders of magnitude slower than in higher dimensional systems but over one order of magnitude faster than radiative recombination explaining the apparent lack of a phonon bottleneck effect in PL experiments. Carrier relaxation is found to limit the PL yield at temperatures above 80 K and might lead to a bottleneck effect in the stimulated emission regime, limiting the high frequency behavior of QD injection lasers and favoring lasing on excited state transitions.

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