Hot carrier relaxation in InAs/GaAs quantum dots

R. Heitz\textsuperscript{a,∗}, I. Mukhametzanov\textsuperscript{b}, H. Born\textsuperscript{a}, M. Grundmann\textsuperscript{a}, A. Hoffmann\textsuperscript{a}, A. Madhukar\textsuperscript{b}, D. Bimberg\textsuperscript{a}

\textsuperscript{a}Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstrasse 36, 10623 Berlin, Germany
\textsuperscript{b}Department of Materials Science and Engineering, University of Southern California, Los Angeles, USA

Abstract

The carrier dynamics in self-organized InAs/GaAs quantum dot (QD) pairs formed in the variable composition amount approach are investigated. Time-resolved photoluminescence measurements as a function of temperature and of the tunnel-barrier thickness demonstrate the influence of slowed-down intradot carrier relaxation in strongly confined systems. In particular, we demonstrate that the carrier-capture efficiency decreases with increasing temperature and find efficient energy transfer between excited states of the QDs. Both effects are attributed to slowed-down carrier-relaxation in the QDs. © 1999 Elsevier Science B.V. All rights reserved.

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Carrier relaxation processes in strongly confined zero-dimensional systems depend on the details of the eigenstate spectrum [1]. The discrete density of states is proposed to impose severe constraints on inelastic phonon scattering slowing down carrier relaxation [2,3]. Alternative non-linear relaxation mechanisms, like Coulomb scattering [4] or Auger recombination [5] are expected to become important with increasing carrier density. The role of such slowed-down carrier relaxation in the optical response of QDs has not yet been investigated in detail.

The spontaneous formation of coherent islands in highly strained InGaAs on GaAs epitaxy provides now routinely high-quality QDs with dimensions in the 10–20 nm region [6]. Such QDs exhibit efficient ground-state photoluminescence (PL), suggesting efficient carrier relaxation in such strongly confined structures. The recent observation of multi-LO-phonon resonances for self-organized InAs/GaAs QDs [1] in samples with fast extrinsic non-radiative recombination unambiguously demonstrated inelastic phonon scattering as dominating relaxation process. Time-resolved PL (TRPL) measurements established relaxation times of some 10 ps [1]. In the absence of competing non-radiative recombination the excited-state transitions are resolved [7]. In this contribution, we demonstrate that for such high-quality samples the slowed-down carrier relaxation again becomes important in case of thermally stimulated carrier escape or energy-transfer processes.

The investigated InAs/GaAs QD samples were grown by molecular beam epitaxy on semi-insulating GaAs(0 0 1) substrates using the variable deposition amount (VDA) approach as described in detail in Refs. [8,9]. Thereby, the optically active layer with either 2.00 or 3.00 ML InAs is grown on...
top of a 1.74 ML seed layer separated by a 36, 45, or 54 ML spacer. The VDA approach enables independent control of the QD density and size and improves the uniformity of the QD ensemble [8]. Furthermore, asymmetric QD pairs (AQDPs) are formed by the small QDs (SQDs) in the seed layer and the larger QDs (LQDs) in the second layer, which are ideally suited to study energy-transfer processes between closely spaced QDs [9]. The QDs are located on top of an ~1 ML thick wetting layer (WL) with a transition energy of ~1.455 eV [9].

The samples were mounted in a continuous-flow He-cryostat or a He-immersion cryostat, providing temperatures between 1.8 and 360 K. PL was excited by an Ar laser, dispersed in a double-grating monochromator, and detected with a cooled Ge-diode. TRPL was excited by 6 ps pulses of a synchronously pumped dye-laser at 670 nm in the GaAs barrier, dispersed by a 0.35 m 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 ~50 ps was taken into account in the analysis of the transients.

The inset of Fig. 1 depicts PL spectra for a 3.00 ML sample with 36 ML spacer. At 6.5 K only a single almost Gaussian peak is observed at 1.028 eV with an FWHM of 25 meV attributed to the LQDs [8] (see also below), which provide ~430 meV exciton localization with respect to the WL. With increasing temperature the QD PL shifts towards lower energies, almost maintaining its shape until above ~200 K a high-energy shoulder indicates thermal population of excited states (I*) [10]. The temperature evolution of the integrated QD PL intensity is given by dots in Fig. 1. The intensity starts to decrease above ~100 K, which accelerates above ~200 K. A satisfactory fit (solid lines) is obtained assuming two thermally activated non-radiative recombination processes to compete with radiative recombination [10]. The derived activation energies of (75 ± 15) and (320 ± 40) meV are close to the excited-state splitting observed in high-density or room-temperature PL experiments (see, e.g., Fig. 1) and the exciton localization energy with respect to the WL, respectively. The latter activation energy is often attributed to thermal stimulation of excitons out of the ground state. However, the exciton binding energy in InAs/GaAs QDs of ~20 meV is considerably smaller than the carrier localization and quantization energies [11]. Since the escape of one carrier already quenches the PL, one would expect to observe single-carrier activation energies. Additionally, a reduction of the carrier capture efficiency with increasing temperature would quench the QD PL intensity for non-resonant excitation [10].

A direct probe for thermally activated carrier escape out of the ground state is the PL decay time, diamonds in Fig. 1. Below ~100 K a constant decay time of (840 ± 50) ps is observed, indicated by the dotted horizontal line. Above ~100 K the lifetime increases up to ~1600 ps before it finally starts to decrease. The radiative recombination probability of a given exciton transition is temperature-independent, accounting for the constant lifetime at low temperatures. With increasing temperature, however, thermal population of excited states becomes important, resulting in an effective lifetime, which depends on the oscillator strength of
the possible excited-state transitions. Due to the smaller substate splitting [11], excited hole states will be populated first and the low probability of recombination between exited hole states and the electron ground state accounts for the increasing lifetime. Finally, above \( \sim 200 \text{ K} \) thermally induced escape reduces the exciton lifetime. Qualitatively similar results for smaller QDs with \( \sim 120 \text{ meV} \) exciton localization have been successfully simulated considering only one localized QD state [12]. In the present case, however, a complete simulation is hampered by the rich spectrum of excited-state transitions in the investigated QDs [10]. A fit of the exciton lifetime, assuming a constant radiative lifetime \( \tau_{\text{rad}} \) of 1600 ps and complete non-radiative recombination of carriers activated into the barrier, yields an activation energy of \((240 \pm 30) \text{ meV}\). This activation energy indicates thermal evaporation of single carriers to the WL to be the driving mechanism behind the decrease of the exciton lifetime. An escape-limited lifetime of 150 ps at room temperature is estimated. The present experimental results show the carrier-capture probability to decrease with increasing temperature. Obviously, carriers are captured into excited QD states and, with increasing temperature, thermally induced escape dominates the slowed-down intradot relaxation.

The constituents of the AQDPs have clearly distinguished ground-state transition energies. Fig. 2 compares PL spectra for 2.00 ML InAs samples with 36, 45, and 54 ML GaAs spacers. For the 36 ML GaAs spacer only a single PL peak at 1.102 eV with an FWHM of 58 meV is observed. The peak energy is \( \sim 100 \text{ meV} \) lower than that of QDs in single-layer samples [13] with 1.74 or 2.00 ML InAs deposition grown at the same conditions, suggesting fast energy-transfer processes to the LQDs in the second layer. With increasing spacer thickness an additional PL peak becomes evident at \( \sim 1.21 \text{ eV} \), resulting in a clear double-peak structure for the 54 ML spacer sample. This peak is attributed to the SQDs in the 1.74 ML seed layer. The increasing PL yield of the SQDs with increasing spacer thickness indicates a decreasing excitation transfer probability in the AQDPs with increasing spacer thickness. Based on PL and PLE results, we have shown recently that the energy-transfer process takes place between localized QD states and made the assumption that the carriers relax to the ground state before interdot transfer takes place [9]. In this case the ground-state lifetime of the SQDs should be strongly decreased yielding further information on the energy-transfer process. The inset of Fig. 2 compares PL transients recorded for the 45 ML sample at 1.102 eV (LQDs) and 1.234 eV (SQDs).
Fig. 3. PL decay (dots) and rise (squares) times derived from two-exponential fits of transients recorded for the 2.00 ML sample with 45 ML spacer.

Carrier diffusion, capture, and relaxation processes determine the rise time. The difference of ~30 ps between the rise times observed for LQDs and SQDs is consistent with the assumption of similar diffusion and capture characteristics but a quenched intradot relaxation time in the SQDs. The TRPL results thus demonstrate strong coupling of the excited states. The competition between interdot energy transfer and intradot carrier relaxation leads to a pronounced decrease in the effective excited-state lifetime which governs the PL rise in the TRPL experiments.

In conclusion, we have presented results of TRPL experiments for asymmetric pairs of self-organized InAs/GaAs QDs as a function of temperature and the intrapair tunnel barrier. The results demonstrate unambiguously the competition between intradot relaxation and competing processes like thermally induced escape and interdot energy transfer. The slowed-down intradot carrier relaxation on a time scale of some 10 ps does not affect the low-temperature PL in high-quality single QDs, but determines the carrier dynamics in the presence of competing processes.

References