Energy relaxation by multiphonon processes in InAs/GaAs quantum dots

R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, and D. Bimberg

Institut für Festkörperphysik, Technische Universität Berlin, 10623 Berlin, Germany

V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov

A. F. Ioffe Physical-Technical Institute, 194021, St. Petersburg, Russia

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Carrier relaxation and recombination in self-organized InAs/GaAs quantum dots (QD's) is investigated by photoluminescence (PL), PL excitation (PLE), and time-resolved PL spectroscopy. We demonstrate inelastic phonon scattering to be the dominant intradot carrier-relaxation mechanism. Multiphonon processes involving up to four LO phonons from either the InAs QD's, the InAs wetting layer, or the GaAs barrier are resolved. The observation of multiphonon resonances in the PLE spectra of the QD's is discussed in analogy to hot exciton relaxation in higher-dimensional semiconductor systems and proposed to be intricately bound to the inhomogeneity of the QD ensemble in conjunction with a competing nonradiative recombination channel observed for the excited hole states. Carrier capture is found to be a cascade process with the initial capture into excited states taking less than a few picoseconds and the multiphonon (involving three LO phonons) relaxation time of the first excited hole state being 40 ps. The $|001\rangle$ hole state presents a relaxation bottleneck that determines the ground-state population time after nonresonant excitation. For the small self-organized InAs/GaAs QD's the intradot carrier relaxation is shown to be faster than radiative (>1 ns) and nonradiative (≈ 100 ps) recombination explaining the absence of a ''phonon bottleneck'' effect in the PL spectra. [S0163-1829(97)09340-5]

I. INTRODUCTION

Carrier relaxation in quasi-0-dimensional semiconductor systems [dubbed quantum dots (QD's)] has been widely discussed in recent years.[1–9] Inelastic phonon scattering in small QD's having large substate splittings as needed for room-temperature optoelectronic applications, is predicted[3,4] to be slow since the discrete spectrum of eigenstates makes energy and momentum conservation difficult. Carrier-relaxation times of the order of radiative and nonradiative recombination times are predicted,[3,5] restricting the population of the ground state upon nonresonant excitation. This effect has been termed ''phonon bottleneck.'' The experimental observation of intense ground-state photoluminescence[10–15] (PL) as well as the demonstration of injection lasing for self-organized QD's (Refs. 16–18) indicate that such a ''phonon bottleneck'' is no intrinsic property of small QD's. Alternative relaxation mechanisms, e.g., Auger recombination[6] and Coulomb scattering of free carriers,[7,8] have been evaluated for their potential to remove the phonon bottleneck. However, a high density of hole states or large free-carrier concentrations would be needed to obtain sufficiently high relaxation rates. At low excitation densities, both conditions are not given for small self-organized QD's,[19] making phonons the most likely candidates to dissipate the energy in carrier relaxation.

A proven method to demonstrate the dominating carrier-relaxation process in higher-dimensional systems is the observation of hot exciton relaxation,[20] revealing LO-phonon resonances in PL excitation (PLE) spectra. Hot exciton relaxation can be observed when the exciton relaxation has to compete with other relaxation or recombination processes. Though for single QD's the classical concept of hot exciton relaxation does not work due to the discrete energy spectrum, we will show that for a QD ensemble an equivalent effect can be observed in PLE, which then gives evidence of the dominating carrier-relaxation process.

The PLE spectra of *single* QD's reveal sharp excitation resonances that are unambiguously attributed to electronic transitions.[21,22] However, comparatively broad excitation resonances 30–130 meV above the detection energy are observed monitoring *large ensembles* of self-organized QD's and controversially discussed either as electronic transitions reflecting the excited-state spectrum[12,13,23] or as multiphonon resonances indicative for carrier-relaxation processes.[10,24,25] Similar (multi)phonon transitions have been observed in near resonance excited PL spectra.[26–28] For InAs/GaAs QD's up to four LO-phonon replica are resolved[10] showing the coupling to LO modes from different regions of the QD structure.[24] The multiphonon resonances have been tentatively explained invoking the inhomogeneity of the QD ensemble and a nonradiative recombination channel for holes in excited states to compete with intradot relaxation.[24] Recently, excited-state PL by itself has been proposed to be sufficiently efficient to compete with intradot relaxation,[25,28] implying the observation of multiphonon resonances to be an intrinsic property. However, PLE results for uncoupled, stacked QD's reveal the variation of the excited-state splitting in dependence of the QD size,[23] indicating excited-state transitions to be resolved in the spectra. The mechanisms determining the appearance of the PLE spectra and therefore their interpretations are not clear yet. Furthermore, hole relaxation[24] and exciton relaxation[28] are controversially discussed, depending on the assignment of the optical transitions to either ''forbidden''[19] or ''allowed''[28] transitions. Both processes cannot be easily distinguished from the en-

ergy spacing of the PLE resonances due to the discrete nature of the involved electronic states.

Here, we present a detailed study of carrier relaxation in self-organized InAs/GaAs QD's combining PLE and time-resolved PL (TRPL) results. This paper is structured as follows: Sec. III A gives detailed PLE results demonstrating the multiphonon nature of the observed excitation resonances, in Sec. III B the multiphonon resonances are shown to be an incoherent phenomenon yielding information on carrier relaxation in the QD's, and in Sec. III C we determine the multiphonon (three LO phonons) relaxation time to 40 ps and show that for the excited hole states in the investigated samples a competing nonradiative recombination channel exists. Finally, in Sec. IV we propose that the observed PLE spectra are intricately bound to the inhomogeneity of the investigated InAs island QD's *and* the nonradiative recombination channel leading to a model similar to hot exciton relaxation in higher-dimensional semiconductors.[20] The nonradiative recombination is associated with defects in the GaAs barrier and enables the characterization of *intrinsic* carrier-relaxation mechanisms in small InAs/GaAs QD's using PLE.

II. SAMPLES AND EXPERIMENTAL SETUP

We report experimental results for two InAs/GaAs QD samples grown by solid-source molecular beam epitaxy on GaAs (100) substrate as described in detail in Refs. 14, 15, and 29. The single InAs layer enclosed by GaAs barriers is sandwiched between two GaAs/$Al_x Ga_{1-x} As$ superlattices grown at 600 °C defining an ''optically active'' region from which carriers can be trapped into the QD's. A GaAs buffer layer is grown on the first superlattice before the temperature is lowered to 480 °C for the growth of 12-Å InAs and a 10-nm-thick GaAs cap. Subsequently, the temperature is increased back to 600 °C for the remaining growth. Samples A and B differ nominally only in the different thickness of the active region being 15 and 116 nm, respectively. TEM results indicate the InAs islands, which sit on top of an InAs wetting layer (WL), to be pyramid-shaped and to vary in size and shape showing side facets close to {101} and {203}.[14,29,30] The QD's have a base length of 12–14 nm, a height of 4–6 nm, and an area density of about 10^{11} cm^{-2}.

For the PL and PLE experiments the samples were mounted in a continuous-flow He cryostat at 6 K. A tungsten lamp dispersed by a 0.27-m double-grating monochromator served as a tunable, low-excitation-density (<0.02 W cm^{-2}) light source and the PL was detected through a 0.5-m single-grating monochromator using a cooled Ge diode. The TRPL measurements were performed in superfluid He with 150-fs pulses at a repetition rate of 76 MHz of a Ti-sapphire laser for excitation and a 0.35-m subtractive double-grating monochromator in combination with a streak camera with an infrared-enhanced photocathode for detection. The full width at half maximum (FWHM) of the system response to the excitation pulses was between 25 and 60 ps depending on the time-range setting. The transients were analyzed taking into account the system response to the excitation pulses enabling the determination of time constants down to 5 ps under optimal conditions.
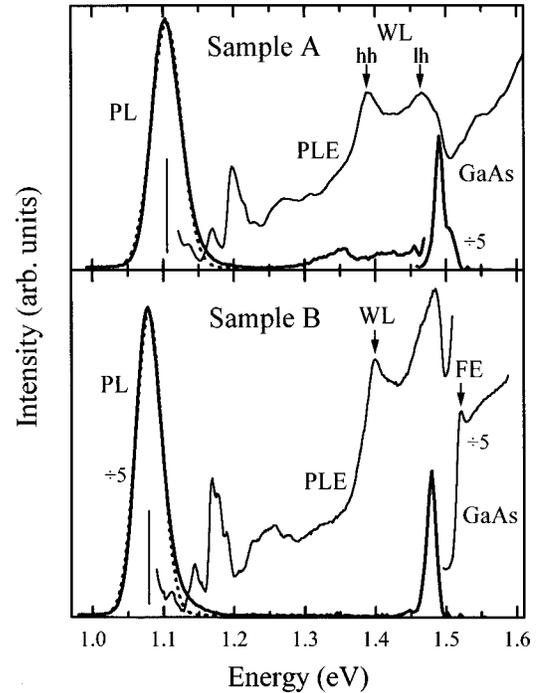
FIG. 1. PL (thick lines) and PLE (thin lines) spectra for samples *A* and *B* taken with $I_0 < 0.02$ W cm^{-2} at $T = 6$ K. The PL is excited at 1.67 eV and the PLE spectra are recorded on the respective PL maximum. The dotted lines represent Gaussian fits of the QD PL. The peak at 1.491 eV is attributed to carbon-related PL in GaAs.

III. EXPERIMENTAL RESULTS

Figure 1 compares low-excitation-density PL (thick lines) and PLE (thin lines) spectra of samples *A* and *B*, with the intensities given on the same scale. Upon excitation above the GaAs band gap the ground-state transition of the InAs QD's leads to PL peaks at 1.105 and 1.079 eV with FWHM's of 42 and 37 meV, respectively, indicating the average InAs island size to be slightly larger in sample *B*. The QD PL peaks are almost of Gaussian shape as indicated by the dotted fits in Fig. 1 and the FWHM is attributed to the inhomogeneity of the QD ensemble with each single QD contributing only a sharp line (<150 μeV).[15] Assuming the carrier-capture cross section and quantum yield to be size independent, the PL peak represents the inhomogeneous QD density, indicating a *size* inhomogeneity of ~5%.[19] The excited-state spectrum of the InAs QD's has been experimentally obtained from high excitation-density PL spectra.[31] Above 50 W cm^{-2} the PL of the QD ground state saturates and two additional peaks appear on the high-energy side shifted by about 85 and 165 meV. The excited-state PL lines are identified as recombination of holes in the $|001\rangle$ and $|002\rangle$ excited states with electrons in the $|000\rangle$ ground state, based on a good agreement with the energy spectrum calculated for strained InAs pyramids.[19] These transitions would be dipole-forbidden in QD's having inversion symmetry but are allowed for the strained InAs pyramids. Figure 2 gives a schematic term scheme of the sample structure showing for the QD's in addition to the $|000\rangle$ ground states the $|001\rangle$ and $|002\rangle$ excited hole states.

A. Multiphonon resonances in the PLE spectra

The excited-state transitions are not resolved in the PLE spectra recorded on the maximum of the QD PL peaks (thin
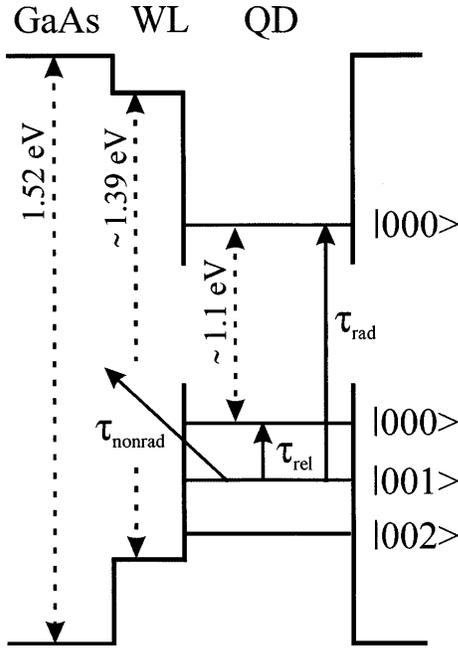
FIG. 2. Schematic term scheme of the InAs QD structures depicting carrier relaxation (τ_{rel}) and radiative (τ_{rad}) as well as nonradiative (τ_{nonrad}) recombination for the excited $|001\rangle$ hole state.

FIG. 3. PLE spectra of sample A in dependence on the detection energy (E_{det}). The spectra are shifted in the y direction for clarity and the crosses mark the respective E_{det}. The inset compares the PL intensity for excitation in the maximum of the 1LO, 2LO, and 3LO resonances and above the GaAs band gap.

lines in Fig. 1). Instead a series of resonances is observed for excitation energies (E_{exc}) below 1.25 eV, which we show below to result from intradot excitation *and* relaxation in the *inhomogeneous* QD *ensemble*. At higher energies the QD PL is excited via carrier capture, revealing absorption by the InAs WL and the GaAs barrier. The GaAs free-exciton is resolved for sample B but not for sample A, for which the narrow width of the active region leads to quantization in the GaAs barrier. The similar PL intensity in both samples for excitation below the GaAs band gap reflects the comparable QD density. Exciting above the GaAs band gap, the PL intensity is about 6 times higher for sample B due to the larger thickness of the active region, providing a higher carrier concentration at the same excitation density. Carriers from the whole active region are equally captured by the QD's. The PLE peaks at 1.388 and 1.465 eV in sample A are attributed to the heavy (hh) and light (lh) -hole absorption of the WL, respectively. Figure 3 shows the dependence of the PLE spectra on the detection energy (E_{det}) for sample A. The spectra are shifted in the y direction for clarity and the respective E_{det} is indicated by the crosses. The energy of the WL resonance is 1.388 eV independent of the detection energy indicating that the QD size and the structural properties of the surrounding WL are not correlated in one sample. The effective thickness of the WL estimated from the transition energy is 1.7 ML.[19] For sample B the heavy-hole absorption appears at a higher energy (1.399 eV) indicating the WL to be thinner in favor of larger QD's.

Exciting below 1.35 eV the PLE spectra assume a characteristic shape as shown in Figs. 1 and 3. Starting from the detection energy, with increasing excitation energy a series of equidistant PLE resonances evolves. The first three resonances are well resolved and increase in intensity, then the fourth appears only weakly and is followed at higher ener-

gies by a weakly modulated broad excitation band. The energy separation of the PLE resonances of about 32 meV lies in the range of typical phonon energies in the InAs/GaAs system. Thus, we refer to the PLE peaks as 1LO, 2LO, etc. resonances as indicated in Fig. 3. The inset of Fig. 3 gives the ground-state PL intensity excited in the maximum of the various LO resonances in dependence of the detection energy. The PL intensity follows the inhomogeneous QD density represented by the QD PL upon GaAs excitation, showing the *relative* intensity of the first three LO resonances to be independent of the QD size and the *absolute* intensity to depend on the QD density. The dominating 3LO resonance falls almost together with the absorption into the $|001\rangle$ excited hole state for pyramids with {101} side facets.[19] We like to note that in our experiments stray light (not shown in the figures) covers the resonantly excited PL of the QD ground state. For QD's having discrete energy levels, absorption and emission match each other within the homogeneous FWHM (<150 μeV).[15] However, calorimetric absorption experiments have shown absorption matching the ground-state PL.[14]
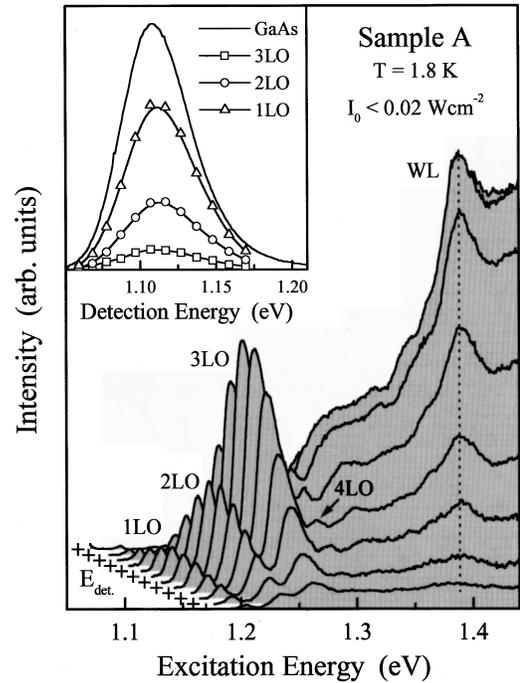
The strongly modulated near-resonant excitation efficiency (Figs. 1 and 3) enables *selective* excitation of QD's with a ground-state transition energy one, two, or three LO-phonon energies below the excitation energy as shown in Fig. 4. In the selectively excited PL spectra, the relative intensity of the LO resonances depends on the excitation energy. With decreasing excitation energy, first the 3LO, then the 2LO, and finally the 1LO replica becomes dominant reflecting the density of QD's with a matching ground-state transition energy. We find no indication of phonon-assisted
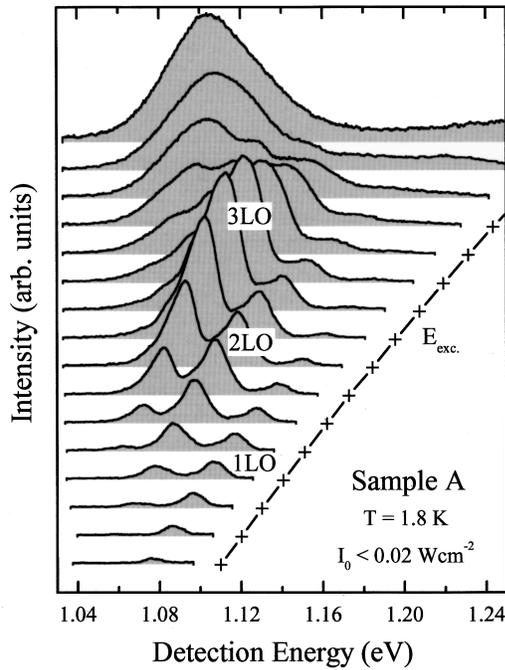
FIG. 4. PL spectra of sample *A* for selective infrared excitation. The spectra are shifted in the *y* direction for clarity and the crosses mark the respective excitation energy (E_{exc}).

FIG. 5. Line-shape fits of the PLE spectra of the QD PL in samples *A* and *B* recorded with $I_0 < 0.02$ W cm^{-2} at $T = 6$ K. The combined experimental resolution of excitation and detection was 3.6 meV.

emission processes consistent with the weak Fröhlich interaction in the covalent III-V semiconductors. For excitation energies above 1.25 eV the inhomogeneous QD peak becomes apparent indicating *nonselective* excitation. This behavior is merely the result of the weak modulation of the PLE spectra above 1.25 eV (Fig. 3) and does not necessitate a nonlocal excitation mechanism as provided by GaAs or WL absorption. Finally, the *sets* of PL and PLE spectra shown in Figs. 3 and 4 both contain the same information, e.g., the PL spectra can be unambiguously generated from the PLE spectra. However, *each* single PL spectrum compares QD's with different ground-state transition energies, whereas in PLE a subset of the QD ensemble defined by the ground-state transition energy is probed. Thus, in the following we prefer the PLE spectra for a more detailed analysis of the phonon modes involved in the multiphonon processes.

Higher resolution PLE spectra for samples *A* and *B* are given in Fig. 5 as a function of the excess excitation energy $\Delta E = E_{exc} - E_{det}$, revealing a threefold substructure for the 2LO and 3LO resonances. In order to determine the energy and FWHM of the various PLE resonances, we performed line-shape fits assuming Gaussian peaks. Full lines in Fig. 5 show the best fit to the PLE spectra (given as thick dots) and the contributing Gaussians are indicated. The large number of Gaussians used is motivated by the rich fine structure resolved for sample *B* and gives also the best fit for sample *A*. The fit of the 2LO and 3LO resonances yields for both samples the same three phonon energies of (29.6 ± 0.5), (32.6 ± 0.5), and (37.6 ± 0.5) meV. The FWHM of the 29.6- and 37.6-meV resonances is ~7 meV, whereas the 32.6-meV resonances are broader (~14 meV for 3LO).[32] For the 1LO resonance only the 32.6-meV mode is resolved. However, PL excited selectively with high density at 1064 nm showed GaAs Raman scattering to contribute to the 1LO peak.[24]
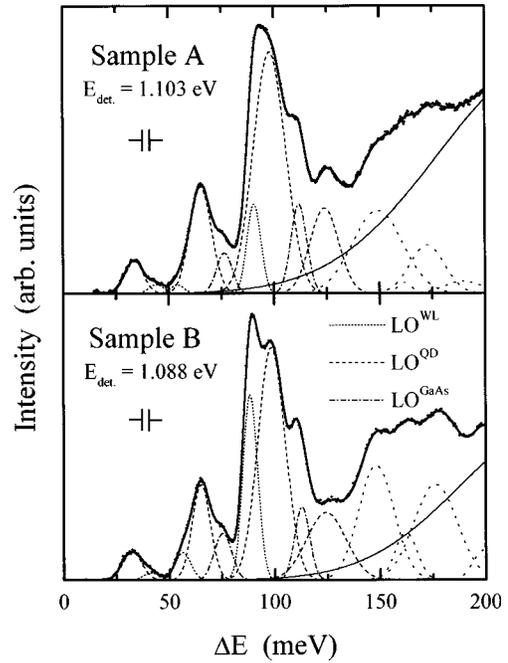
Above 130 meV the resonances become broader and no longer correspond to multiples of the above phonon energies. A fit of a complete series of PLE spectra shows that the peak energies of the various LO resonances and therefore the corresponding phonon energies are independent of the detection energy. This is illustrated in Fig. 6 for sample *B* showing a contour plot of the PL intensity in dependence of the detection wavelength and ΔE. Multiples of the three phonon energies determined in the line-shape fits are indicated by horizontal lines. We like to note that no resonances matching combinations of the different phonon modes are resolved.

The three phonon modes can be attributed to LO phonons in different regions of the strained InAs/GaAs QD structure, taking into account that the LO-phonon energy in a semiconductor nanostructure is altered by strain and confinement.[33] The effects of the biaxial strain and the strong vertical confinement compensate each other for thin InAs quantum wells (QW's).[34] Thus, we identify the 29.6-meV mode, corresponding in energy to the bulk InAs LO phonon, with the LO phonon of the InAs WL. The 32.6-meV mode, which dominates the PLE spectra, is attributed to the LO phonon of the strained InAs QD's, for which phonon confinement can be neglected. A LO phonon energy of 32.1 meV has been estimated from the strain distribution in InAs pyramids with {101} side facets,[19] which is in excellent agreement with the experimental value. The 37.6-meV mode is assigned to the LO phonon in the GaAs barrier. The increased energy compared to bulk GaAs reflects the biaxial strain accommodated by the barrier in the immediate vicinity of the islands. The large FWHM's of the PLE resonances (>7 meV) might be the result of weakened *k*-selection rules, the shape inhomogeneity of the islands, which alters the strain distribution and therewith the phonon energies, or higher-order processes involving LA phonons.[4] Further investigations are needed for a
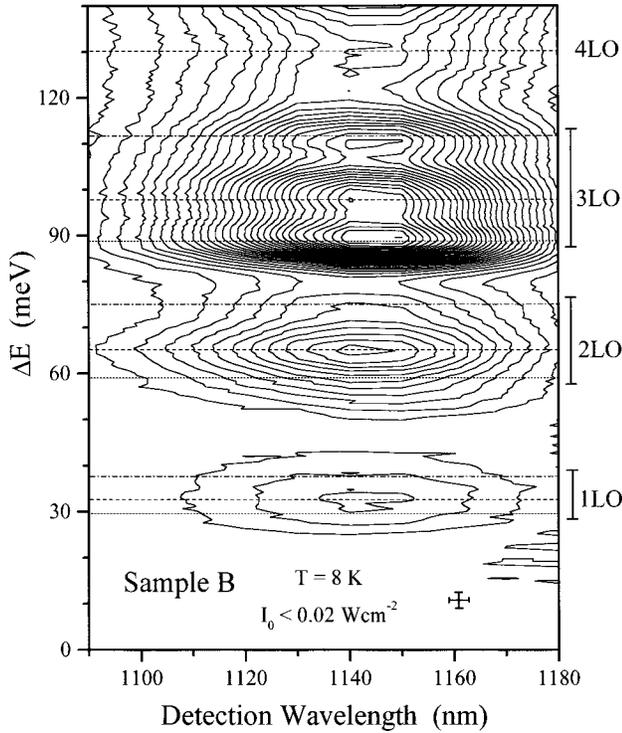
FIG. 6. Contour plot of the QD PL intensity in sample B as a function of the detection wavelength and the excess excitation energy ($\Delta E = E_{exc} - E_{det}$). The horizontal lines represent multiples of the phonon energies determined in the line-shape fits.

FIG. 7. QD PL transients for sample A excited above the GaAs band gap with a density of $100 \, W \, cm^{-2}$, corresponding to $4 \times 10^{10} \, cm^{-2}$ electron-hole pairs per pulse. The inset compares PL spectra for pulsed and cw ($\sim 0.01 \, W \, cm^{-2}$) excitation.

more detailed analysis. Figure 5 shows that compared to the QD phonon replica, the WL and GaAs modes are more prominent for the 3LO resonance than for the 1LO and 2LO ones. The coupling to both modes depends on the extension of the excited-state wave function into the barrier. Thus, the interaction with both modes becomes stronger with increasing ΔE and therefore decreasing carrier localization in the excited state. The phonon replica in the PLE spectra of the QD's are *local* probes of the strain in and around the overgrown InAs islands. The observed phonon energies are consistent with *coherent* InAs islands and can thus be taken as indication for the detected, optically active QD's to be dislocation-free.

B. The incoherent nature of the PLE resonances

Multiphonon resonances in the PLE spectra of bulk semiconductors,[20] QW's,[35] and corrugated superlattices[36] have been controversially discussed in terms of hot exciton relaxation and resonant Raman scattering (RRS). The basic difference between both processes is that hot exciton relaxation is an incoherent process, and RRS is a coherent process. Thus, both processes usually proceed on a different time scale defined by energy relaxation and dephasing processes, respectively. As shown by Toyozawa, Koyani, and Sumi[37] both processes become indistinguishable when both time scales become similar. In order to decide on the nature of the multiphonon resonances observed for the InAs/GaAs QD's we studied TRPL in dependence of the excitation energy.

Typical transients observed for sample A upon excitation above the GaAs band gap with an excitation density of

$100 \, W \, cm^{-2}$ are shown in Fig. 7. The respective detection energies are indicated by the arrows in the inset. Transients detected near the maximum (or on the low-energy side) of the QD peak decay monoexponentially, whereas those detected on the high-energy side show an additional faster component which we attribute to the superposition of excited-state PL of larger QD's and ground-state PL of smaller ones. The PL spectrum for pulsed excitation (full line in the inset) shows a high-energy shoulder not present for low-density cw excitation (broken line). Though the time-averaged excitation density of $100 \, W \, cm^{-2}$ was chosen by avoiding saturation of the QD ground-state PL during the experiments it was sufficient to saturate part of the QD's leading to excited-state PL. At $100 \, W \, cm^{-2}$ each excitation pulse generates about $4 \times 10^{10} \, cm^{-2}$ electron hole pairs in the 15-nm-wide active GaAs barrier, which is close to the QD density ($\sim 10^{11} \, cm^{-2}$).

Figure 8 compares transients of the QD ground-state PL for various excitation energies. Most remarkable, we observe the same time behavior exciting via GaAs or WL absorption, which are completely incoherent excitation processes, and exciting via the 3LO resonance. This observation presents unambiguous justification to treat the multiphonon resonances as an incoherent phenomenon, i.e., as a three-step process: first absorption into excited states, then relaxation, and finally ground-state recombination. Raman scattering might contribute to the one LO resonance as observed in selectively excited PL spectra.[24] Recently, Raymond *et al.*[27] reported for $Al_xIn_{1-x}As/Al_xGa_{1-x}As$ QD's *faster* PL decay exciting 1LO and 2LO phonon energies above the detection. The authors used the fact that the observed decay is still slower than the exciting laser pulse as argument to reject RRS for the observed phonon resonances located on an intense, broad PL background. This background is not present
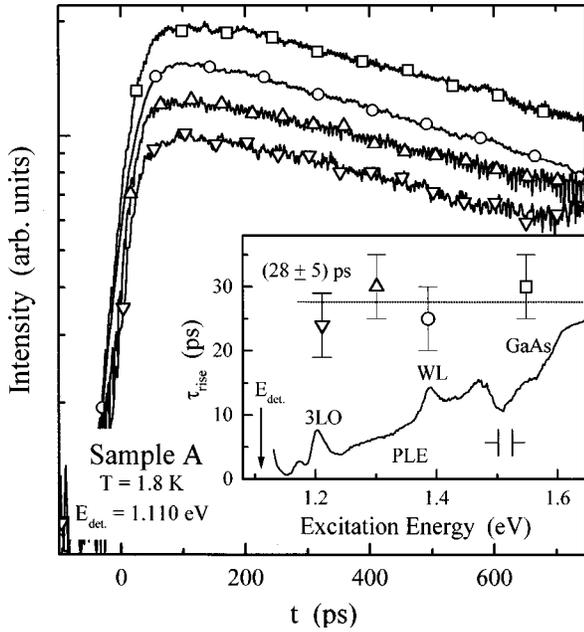
FIG. 8. Transients of the QD PL in sample *A* for different excitation energies. The inset gives the PL rise time in dependence of the excitation energy. The combined spectral resolution of excitation and detection was 20 meV for the time-resolved measurements.

FIG. 9. Energy dependence of the PL lifetimes of the ground state (τ_1) and the first excited state (τ_2) as well as the PL rise time (τ_{rise}) for samples *A* and *B* upon GaAs excitation. The excitation densities were 100 and 1 W cm^{-2}, respectively.

in case of the InAs QD's, Fig. 5. We like to note that the dephasing times of the carriers in the InAs/GaAs QD's are hitherto unknown, but can be expected to be longer than in higher-dimensional systems. The spatial localization of the carriers in the QD's suppresses carrier-carrier scattering, which is the dominant dephasing process in higher-dimensional systems, and the δ-function density of states drastically restricts phonon scattering. Indeed, for bound excitons in bulk semiconductors recombination-limited dephasing times of several hundred ps have been observed.[38,39]

C. Relaxation and recombination dynamics

The PL transients are analyzed taking into account two exponential decay processes for ground (τ_1) and excited (τ_2) -state recombination, and one exponential rise (τ_{rise}) to account for carrier capture and relaxation. Typical results of a least square fit of the experimental data with the convolution of the assumed transient and the system response function are shown as full lines in Fig. 7. Figure 9 compares rise and decay times deduced for samples *A* and *B* upon excitation above the GaAs band gap in dependence of the detection energy. The transients for sample *B*, which have been measured at an excitation density of 1 W cm^{-2} corresponding to an initial carrier density of 3×10^9 cm^{-2} much lower than the QD density ($\sim 10^{11}$ cm^{-2}), decay monoexponentially even on the high-energy side of the QD PL confirming excited-state PL to be not detectable at such low-excitation densities.

The ground-state decay times of 1000 ± 100 ps and 1070 ± 100 ps for samples *A* and *B*, respectively, observed near the center or on the low-energy side of the QD PL peak are in good agreement with previously reported values for In$_x$Ga$_{1-x}$As (Ref. 40) and InAs (Refs. 17 and 41) QD's. With increasing ground-state transition energy and therefore
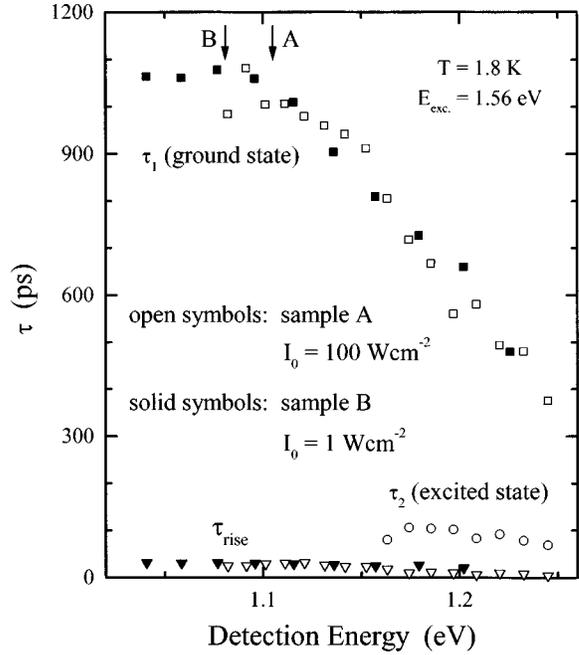
decreasing QD size, the decay time decreases to about 350 ps near 1.25 eV. Lateral phonon-assisted tunneling from small to large InAs QD's has been proposed recently[41] to explain the shorter lifetime of the smaller QD's. Though the QD density in our samples is similar to that of the sample investigated in Ref. 41, we do not observe the reported characteristic deformation of the QD PL peak. Indeed, a ground-state PL lifetime of 340 ps is found for samples having only small QD's[14] in good agreement with the present results. Thus, lateral energy transfer between QD's is unlikely to be responsible for the energy dependence of the ground-state lifetime shown in Fig. 9. The observed decrease of the ground-state lifetime with decreasing QD size could be the result of an increasing oscillator strength. However, for the investigated small InAs/GaAs QD's carrier-quantization energies are much larger than the exciton binding energy.[19] In this strong confinement regime, the oscillator strength is predicted to be size independent.[42] We cannot exclude that nonradiative recombination, demonstrated below for the excited QD states, is also of importance for the ground-state transition of the smaller QD's.

Carrier capture and the subsequent intradot relaxation determine the PL rise after nonresonant excitation. Our TRPL results (Figs. 8 and 9) suggest carrier capture to be a cascade process involving at least the excited $|001\rangle$ hole state as intermediate state. The evaluation of the excitation-energy-dependent transients (Fig. 8), yields a rise time of 28 ± 5 ps, which is within our temporal resolution (~ 5 ps) independent of the excitation process (see inset), indicating that only the last relaxation step, namely, the hole relaxation from the excited $|001\rangle$ state, is resolved. This is supported by the detection energy-dependent data shown in Fig. 9. The ground-state rise time is about 30 ps independent of the QD size, whereas the excited-state PL rises much faster (<10 ps,
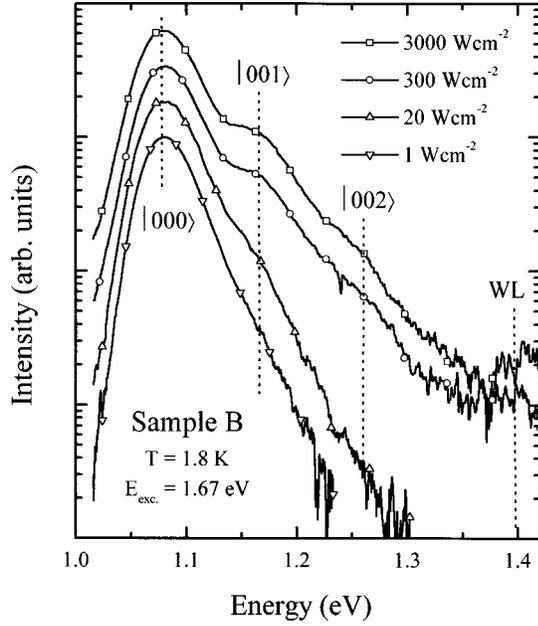
FIG. 10. Excitation-density-dependent PL spectra for sample B and pulsed excitation. The spectra are normalized and shifted in the y direction for clarity. An excitation density of 1 W cm^{-2} corresponds to an initial carrier density of 3×10^9 cm^{-2}.

sample A for $E_{\text{det}} > 1.17$ eV). These observations show that the observed PL rise time of 28 ± 5 ps is the lifetime of the $|001\rangle$ excited hole state and that we are not able to resolve carrier capture in the excited hole states (<5 ps). The initial carrier capture time has been assessed to be on a 1-ps time scale from the PL yield of deep etched mesa structures in dependence of the mesa diameter[43] and from the observation of hot electron resonances in the QD excitation via the GaAs barrier.[23] We find no influence of carrier diffusion in the GaAs barrier, which we attribute to a long-range attractive potential caused by the strain field surrounding the QD's[19] leading to carrier drift.

The lifetime of the $|001\rangle$ excited hole state depends on relaxation (τ_{rel}) to the $|000\rangle$ ground-state, radiative recombination (τ_{rad}) with electrons in the $|000\rangle$ state, and nonradiative recombination (τ_{nonrad}), as indicated in Fig. 2:

$$\tau^{-1} = \tau_{\text{rel}}^{-1} + \tau_{\text{rad}}^{-1} + \tau_{\text{nonrad}}^{-1}. \quad (1)$$

As shown above, at low excitation densities radiative recombination of excited states is suppressed by hole relaxation ($\tau_{\text{rel}} \ll \tau_{\text{rad}}$). The intensity distribution between the ground state and excited-state PL at high-density cw excitation[31] as well as the results of the calculations[19] indicate the oscillator strength of the excited-state transitions to be lower than that of the ground-state transition ($\tau_{\text{rad}} > 1000$ ps).

The nonradiative recombination (τ_{nonrad}) can be assessed in high excitation density TRPL experiments. The saturation of the QD states with two carriers suppresses the intradot relaxation leading to excited-state PL (Refs. 26 and 31) and altering the carrier dynamics. Figure 10 shows for sample B excitation density-dependent PL spectra upon pulsed excitation above the GaAs band gap (the spectra are normalized and shifted in the y direction for clarity). At 1 W cm^{-2} (3×10^9 cm^{-2} electron hole pairs per pulse) only ground-
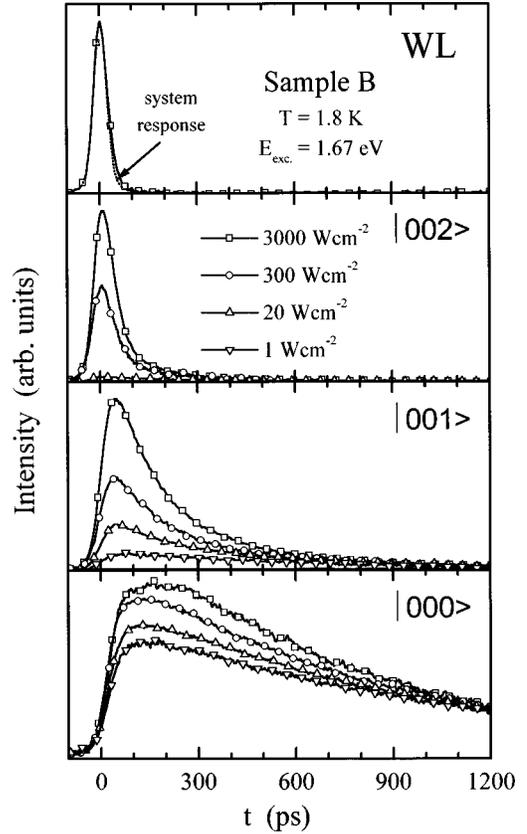
FIG. 11. Transients recorded for sample B at the spectral positions corresponding to PL from the ground state ($|000\rangle$), the excited states ($|001\rangle$, $|002\rangle$), and the WL as indicated in Fig. 10 at various excitation densities. The transients are normalized for the slow decay component.

state PL is evident, and above 300 W cm^{-2} the spectral shape saturates revealing emission from the $|001\rangle$ and $|002\rangle$ excited hole states 82 and 177 meV above the ground-state transition as well as weak signal in the region of the WL, observed at 1.399 eV in PLE (Fig. 1). The excited-state transitions appear much weaker in PL for pulsed excitation than upon cw excitation, compare, e.g., Fig. 3 in Ref. 31, since the decay of the excited-state population favors the ground-state transition. The saturation of the shape of the PL spectrum above 300 W cm^{-2} indicates the QD's to be initially completely filled by each pulse.

The fast intradot relaxation leads to a situation where the holes in each QD behave practically like a hole gas filling the energy states starting from the ground state. Consequently, the PL of the highest populated state will decay following the decrease of the hole density and PL from lower-energy states will be constant as long as the higher ones act as reservoir, supplying holes as soon as one recombines. Lowering the excitation density, the decay is entered at a later stage, corresponding to the lower initial carrier density. This is shown in Fig. 11 comparing for sample B transients recorded with different excitation densities at the spectral positions of the various PL transitions (marked by the dotted lines in Fig. 10). Though the QD's are initially completely filled at the highest excitation density (3000 W cm^{-2}), the transient of the WL follows the system response (dotted curve) to the excitation pulses indicating fast recombination in the WL

($\tau < 5$ ps). With decreasing detection energy the PL decay becomes slower, with time constants of 42 ± 10 and 95 ± 10 ps (100 ps in sample *A*, Fig. 9) for recombination from the $|002\rangle$ and $|001\rangle$ excited hole states, respectively. Finally, the ground-state PL intensity is initially nearly constant and starts to decay only after a delay of 250 ps. Reducing the excitation density, the intensity of the fast decay components attributed to the excited-state PL decreases, but the decay times remain constant. The decay time of the ground-state transition, however, shortens from 1070 to 800 ps increasing the excitation density from 1 to 3000 W cm^2. These results show that in the investigated samples the hole population decays with time constants shorter than 100 ps until only the ground state is populated.

The fast decay of the excited-state PL under saturation conditions implies a nonradiative recombination channel as indicated in Fig. 2, with the observed lifetimes being a measure for the recombination times. Thus, the nonradiative recombination times are of the order of 95 and 42 ps for the $|002\rangle$ and $|001\rangle$ excited hole states and less than 5 ps for the WL. Auger processes, proposed recently to explain the saturation of the integrated PL intensity under high-density cw excitation in similar samples,[17,29] could explain the increasing nonradiative recombination probability with increasing occupation of the QD's. However, the recent observation[23,44] of longer time constants for the decay of excited-state PL in InAs QD's suggests that the nonradiative recombination is not an intrinsic property of the QD's. Our experimental results can be explained assuming the nonradiative recombination to be defect related. 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. The decreasing localization of the holes in the excited states and, in particular, in the WL would then account for the increasing nonradiative recombination probability. Recently, Sercel[9] discussed energy transfer between deep defects in the GaAs barrier and In$_x$Ga$_{1-x}$As QD's and predicted transfer times in the 100-ps region for a QD/deep defect separation of up to 10 nm supporting the interpretation of the nonradiative recombination. The formation of deep defects in the GaAs cap or the 1.7-ML InAs WL might be intricately connected to the strain fields generated by the InAs islands or be caused by the low-growth temperature of the initial GaAs covering layer. For samples having smaller three-dimensional islands and a thinner WL no signs for nonradiative recombination are observed.[17,29]

From the low-excitation-density lifetime and the nonradiative recombination time of the $|001\rangle$ excited hole state, a multiphonon relaxation time of 40 ± 7 ps is determined using Eq. (1). This result is in good agreement with recent results for uncoupled, stacked InAs QD's, offering no nonradiative recombination channel,[23] and for In$_{0.3}$Ga$_{0.7}$As QD's.[45] The hole relaxation from the $|001\rangle$ state needs three LO phonons to span the energy gap to the $|000\rangle$ ground state, and is thus at least a third-order effect. The relaxation time is of the order of that predicted for second-order scattering of LO +LA phonons at low temperatures,[4] indicating that for the small InAs QD's the multiphonon emission probability does not drastically decrease for higher-order processes. The reason might be an increased Fröhlich interaction in the small InAs QD's as proposed for II-VI nanocrystals,[46,47] or the

change in the local charge distribution for the excited states. Recently, Arakawa *et al.*[48] solved the time-dependent Schrödinger equation using the coupled mode equations for electron capture into localized QD states, yielding a weak decrease of the relaxation probability with increasing energy spacing ΔE supporting our results. A detailed discussion of multiphonon relaxation processes between localized QD states should be based on calculations taking into account the actual QD shape (pyramids), the finite-carrier confinement, and proper confined phonon modes, which is beyond the scope of this paper.

IV. DISCUSSION

In the preceding section we have shown that the QD PLE spectra in the investigated samples are dominated by carrier relaxation leading to multiphonon resonances. Such a behavior is well known as hot exciton relaxation in higher-dimensional systems[20,35,36] and is there inseparably bound to the continuous density of states. The present results indicate that a similar effect can be observed for QD's though they have a discrete density of states. In the following, we will show that the observation of ''hot exciton'' relaxation for QD's depends critically on the fact that we probe an inhomogeneous QD ensemble providing an efficient competing relaxation channel, namely, defect-related nonradiative recombination for the excited hole states. Though, the inhomogeneity as well as the nonradiative recombination are undesired properties of the QD ensemble, they result for the investigated samples in a situation where PLE spectra directly demonstrate the dominating carrier relaxation process.

The first excited $|001\rangle$ hole state acts as bottleneck for hole relaxation in the small InAs/GaAs QD's. Comparing with Fig. 2, the quantum yield η for hole relaxation from the $|001\rangle$ to the $|000\rangle$ state, which we assume to be also that of the ground-state PL, is given by the competition between hole relaxation and recombination in the $|001\rangle$ hole state:

$$\eta = \frac{\tau_{rel}^{-1}}{\tau_{rel}^{-1} + \tau_{rad}^{-1} + \tau_{nonrad}^{-1}} = \frac{\tau_{|001\rangle}}{\tau_{rel}} \approx \frac{\tau_{nonrad}}{\tau_{rel} + \tau_{nonrad}}. \quad (2)$$

The lack of excited-state PL upon low-density nonresonant excitation shows that radiative recombination is too slow (>1 ns) to compete with intradot relaxation and, thus, can be neglected in Eq. (2). The nonradiative recombination in the investigated samples, however, is sufficiently fast ($\tau_{nonrad} \sim 100$ ps) to compete with hole relaxation. We like to note that the nonradiative recombination channel has no effect on the intensity ratio of excited- to ground-state PL which is given by the ratio τ_{rel}/τ_{rad} at low excitation densities and the statistics of carrier capture at higher excitation densities.[49]

The quantum yield η for hole relaxation in a QD depends on the relaxation time τ_{rel} for the $|001\rangle$ hole state and thus is expected to be a function of the excited-state splitting.[3,4] For single QD's electronic transitions lead to sharp lines in the PLE spectra, which are not correlated to LO-phonon energies,[21,22] and η modulates only the intensity of the excitation resonances. The situation is the same for an ensemble of QD's of well-defined shape, which have a clear correlation between the ground-state transition energy and the

excited-state splittings. However, the self-organized InAs/GaAs QD's investigated in this paper vary in size *and* shape. Consequently, the excited-state splitting of a subset of QD's with the same ground-state transition energy is inhomogeneously broadened leading to a finite density $N(E_{exc}, E_{det})$ of QD's, which are simultaneously in resonance with excitation *and* detection in a selective PL or PLE experiment. The emission intensity I is then given by

$$I \propto N(E_{exc}, E_{det}) \, \eta(E_{exc} - E_{det}). \qquad (3)$$

The dependence of the quantum yield η on the excess excitation energy $\Delta E = E_{exc} - E_{det}$ determines which QD's contribute to the selective PL or PLE spectra, revealing the dominating relaxation processes if the inhomogeneous broadening of the excited-state splitting is sufficiently large. The PLE spectra of the investigated samples, revealing 2LO and 3LO-phonon replica, indicate that the inhomogeneous broadening of the excited-state spectrum covers this energy range for a given ground-state transition energy and that the dependence of τ_{rel} on ΔE strongly modulates η. For $\eta \sim 1$, i.e., when nonradiative recombination is negligible, Eq. (3) predicts the PLE spectra to reveal the inhomogeneously broadened excited-state splitting, as recently observed for uncoupled, stacked InAs QD's.[23] The FWHM of the PLE resonances indicated the inhomogeneous excited-state broadening to exceed 20 meV.[23]

The observation of multiphonon resonances in the PLE spectra of small InAs/GaAs QD's is thus unambiguous proof for multiphonon emission to be the dominant intradot relaxation process. Every time the excited-state splitting ΔE equals multiples of one of the available LO-phonon energies, the relaxation rate (τ_{rel}^{-1}) has a maximum resulting in a PLE peak. The above discussion shows that the increasing intensity from the 1LO to the 3LO resonance is the result of the inhomogeneity of the excited-state splitting N centered around 85 meV, which acts as envelope function. At higher energies the excitation process involves absorption in the $|002\rangle$ or higher-excited hole states, subject to cascade relaxation. In this case, η is no longer defined by the experimental settings, i.e., E_{exc} and E_{det} but depends on the actual excited-state spectrum, resulting in an effective η_{eff}, averaged over the QD ensemble, in Eq. (3). Thus, we attribute the intensity drop for the 4LO replica as well as the onset of the almost structureless PLE above 1.25 eV, Figs. 1 and 3, to multistep relaxation in the QD's. The situation might be different in an external magnetic field leading to a tunable splitting for the $|001\rangle$ hole state. In this case, the excited-state splitting and therefore also the two-step relaxation processes are well defined and can thus be resolved in the PLE spectra.

The multiphonon relaxation time τ_{rel} is 40 ps for QD's having an excited-state splitting in resonance with the 3LO process and expected to be faster for a lower-order process, i.e., 2LO scattering. For QD's out of resonance τ_{rel} is expected to be much larger corresponding to the lower quantum efficiency observed in PLE. However, the TRPL measurement are dominated by in-resonance QD's, having a high excitation yield, due to the limited energy resolution ($\sim$20 meV) in the near-resonant excited TRPL experiments and the fact that LO phonons with various energies contribute in the relaxation process, allowing efficient carrier relaxation

for a wide range of excited-state splittings. Using the experimental time constants and Eq. (2) we estimate a quantum yield of 70% for the majority of the QD's in the investigated samples, having an excited-state splitting allowing 3LO scattering. Thus, it is not justified to speak of a phonon bottleneck as proposed by Benisty, Sotomayor Torres, and Weisbuch,[3] in particular, since the nonradiative recombination channel is not an intrinsic property of the InAs/GaAs QD's. However, the impact of out-of-resonance QD's is expected to increase in case of samples with higher island uniformity.

The above discussion shows that PLE is a powerful tool for the characterization of an inhomogeneous QD ensemble giving access to the shape nonuniformity of the islands, the carrier relaxation processes, and nonradiative recombination. A detailed knowledge of the energy dependence of η, and therewith of the relaxation and nonradiative recombination probabilities would be a precondition for a detailed modeling of the PLE spectra. However, the relative intensity of the 1LO and 2LO resonances in our experiments are a sensitive measure for the shape inhomogeneity of the islands and a change in the mean excited-state energy would change the intensity distribution among the LO resonances. The relative intensity of the various LO resonances in our PLE experiments (Fig. 3) is independent of the detection energy, indicating the average excited-state energy to be independent of the ground-state transition energy. These results suggest the average island height-to-width ratio to decrease with increasing island size, as recently inferred from high-excitation-density PL spectra of a series of samples.[31]

V. CONCLUSION

We have investigated carrier relaxation and recombination for small self-organized InAs/GaAs QD's. We observe phonon resonances involving up to 4LO phonons in the PLE spectra of the QD ground state and demonstrate with TRPL measurements that these resonances can be discussed as an incoherent phenomenon, i.e., as a multistep process yielding information on excited-state absorption and carrier relaxation in the QD's. The results present unambiguous proof for inelastic phonon scattering to be the dominant carrier relaxation process at low excitation densities.

In particular, the present results show efficient carrier capture after nonresonant excitation in the GaAs barrier and the InAs WL. The initial carrier capture takes place on a ps time scale resulting in the population of the excited $|001\rangle$ hole state, which presents a bottleneck for hole relaxation to the ground state. The multiphonon emission limited lifetime is 40 ± 7 ps for QD's with an excited-state splitting in resonance with a 3LO process. The results show that carrier capture is a cascade process involving at least the excited $|001\rangle$ hole state. We observe ground-state lifetimes of about 1000 ps for InAs QD's with a transition energy around 1.1 eV, which is over an order of magnitude slower than intradot carrier relaxation. The TRPL results for excitation densities sufficient to saturate the hole states in the QD's, reveal a nonradiative recombination channel for holes in excited states with recombination times shorter than 100 ps. The energy dependence of the nonradiative recombination rate suggests energy transfer to deep defects in the vicinity of the InAs islands.

We propose an explanation for the observation of carrier relaxation processes in the PLE spectra of the InAs QD's having a discrete energy spectrum with substate splittings larger than LO-phonon energies, which is in close analogy to hot exciton relaxation in higher-dimensional semi-conductors.[50] The continuous density of states being fundamental to hot exciton relaxation is thereby replaced by the inhomogeneity of the InAs QD ensemble. The observation of multiphonon resonances is the result of the competing non-radiative recombination channel in the investigated samples. QD's with an excited-state splitting in resonance with one of the multi-LO-phonon processes will allow efficient hole relaxation to the ground state instead of nonradiative recombination. From the experimentally derived time constants we estimate a yield of about 70% for hole relaxation in these QD's. For the small InAs QD's a wide energy window for efficient ground-state population results from the coupling to three different phonon modes with energies of 29.6, 32.6, and 37.6 meV, attributed to the InAs WL, the InAs QD's, and the strained GaAs barrier, respectively. The present results show that PLE is a powerful tool to characterize the inhomogeneity and nonradiative recombination in a QD sample.

The $|001\rangle$ hole state acts as a bottleneck for hole relaxation. However, this bottleneck is not observed in PL experiments, since intradot hole relaxation is found to be faster than radiative or nonradiative recombination from the excited states, but might be important for QD-based injection lasers, when stimulated emission accelerates carrier recombination. A hole relaxation time of 40 ps would limit the high-frequency capability and could cause excited states to contribute in the lasing. The present results, however, describe carrier relaxation at low-excitation densities and thus give only a lower limit for the relaxation probability under lasing conditions, where Auger processes and Coulomb scattering might accelerate carrier relaxation.

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