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Carrier dynamics in modulation-doped InAs/GaAs quantum dots

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Photoexcited carrier dynamics was studied in *n* and *p* modulation-doped self-assembled InAs/GaAs quantum dots by means of time-resolved photoluminescence with excitation and detection energies varied through barrier, wetting layer, and quantum dot states. Carrier transfer to the ground state of the dots was found to occur within 5 to 6 and 12 ps for the doped and undoped samples, respectively. The experiments suggest that in all samples the carrier capture into the highest quantum dot levels proceeds by phonon emission. The significant difference in the transfer times is attributed to different relaxation mechanisms for the subsequent process of intradot carrier relaxation. For the doped samples, the presence of built-in carriers in the dots leads to efficient electron-hole scattering, while in the undoped structure scattering by phonons is identified as the main relaxation channel. Additionally, experimental results show decreased carrier lifetimes in the doped structures, which is attributed to nonradiative recombination at doping-induced recombination centers in the vicinity of the quantum dot layers.

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I. INTRODUCTION

Modulation-doped self-assembled quantum dots (QD) have found applications in QD infrared detectors,[1] single electron transistors,[2] QD memory devices,[3] and, recently, QD lasers. In the latter case, *p*-doped lasers were reported to be superior to undoped ones. They show better temperature characteristics[4] and promise improved high-speed properties.[5] The first trait is attributed to countering the effect of a thermal smearing of holes in closely spaced hole levels.[4] The high-speed characteristics of nanostructure lasers, on the other hand, are known to be greatly affected by carrier capture and relaxation. In doped quantum dot structures, the presence of built-in carriers should alter carrier dynamics and lead to enhanced carrier relaxation rates.

Yet, despite the potential for applications, only a few studies of carrier dynamics in modulation-doped QD structures have been reported.[6–10] From resonant photoluminescence experiments, Guasch and co-workers concluded completely different relaxation mechanisms for doped and undoped structures; however, this conclusion was left without a major discussion.[6] Lee *et al.* have measured excited state carrier lifetimes in charged InAs QDs, albeit only with ~100 ps resolution.[7,8] They found that the screening effect caused by the charges in the doped layer and an interaction with ionized impurities lead to a complex dependence of carrier lifetimes on the doping level. Investigations of interlevel electron relaxation in *n*-doped samples revealed a typical relaxation time of ~3 ps.[9] Time-resolved photoluminescence (PL) studies on both *n*- and *p*-doped dots were reported by Gündoğdu *et al.*[10] Carrier relaxation in these structures was found to be extremely fast, especially in the *p*-doped sample, which was attributed to efficient electron-hole scattering. However, the authors have studied only the overall carrier transfer from the barriers to the ground state of the dots, without addressing details of the transfer process. Furthermore, the report does not give any information about carrier lifetimes.

Thus, a more comprehensive picture on carrier dynamics in *n* or *p* modulation-doped QD structures is still missing. In the present paper, we attempt to fill this gap by performing an experimental study in which tunable excitation and the detection of emission in *n*- and *p*-doped, as well as undoped QD structures, is applied, allowing us to separate carrier transport, capture, relaxation, and recombination processes.

II. EXPERIMENT

The samples were prepared using molecular beam epitaxy. Initially, 500 nm of nominally undoped GaAs buffer layers were grown on semi-insulating GaAs substrates. QDs were formed by depositing 3.5 monolayers of InAs at 520 °C. Topographic images obtained by atomic force microscopy (Fig. 1) show lens-shaped dots with typical sizes of 45 nm diameter and 8 nm height, and densities of $3\times10^{10}\ \mathrm{cm}^{-2}$. For optical characterization, the dots were subsequently covered by a 200 nm thick GaAs layer. At a distance of 10 nm below the dot layer, 10 nm thick Si (*n*-type) or Be (*p*-type) modulation-doped layers with nominal dopant concentrations of 5×10^{18} and $1\times10^{19}\ \mathrm{cm}^{-3}$, respectively, were introduced during the growth. To explore the influence of doping level on the carrier dynamics, measurements were repeated on samples in which the doping concentration was by an order of magnitude lower. However, the carrier dynamics in the samples with high and moderate doping were essentially the same, thus, later we present data only for the highly doped samples. For comparison, an undoped reference sample was prepared under the same growth conditions.

Carrier dynamics in the structures were measured by means of time-resolved PL. A tunable mode-locked Ti:sap-

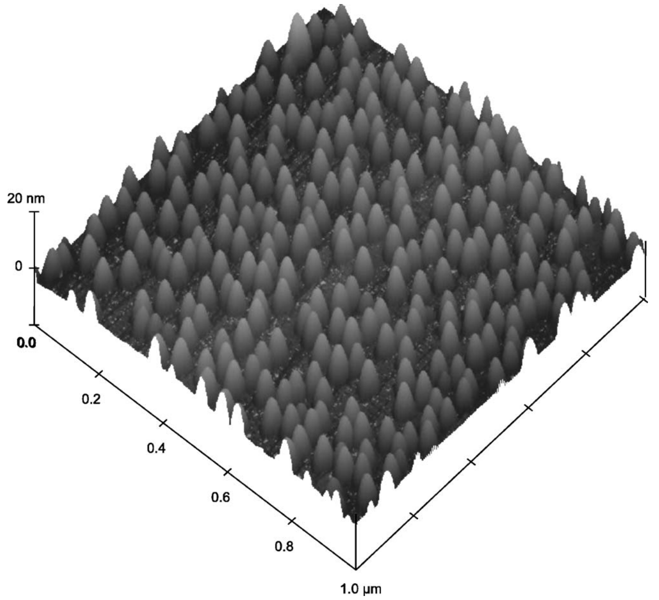

FIG. 1. $1 \times 1\ \mu m^2$ atomic force microscopy topographic image of the undoped reference prior to capping.

phire laser (pulse length 130 fs, repetition rate 76 MHz, wavelength tuning range 700–1040 nm) was used for excitation. Carrier lifetime measurements were performed by detecting the photoluminescence signal with a synchroscan streak camera combined with a 0.25 m spectrometer, providing a temporal resolution of 3 ps. Carrier capture and relaxation was measured using the streak camera and an upconversion setup (temporal resolution 150 fs). The upconverted signal from the $LiIO_3$ nonlinear crystal was detected by a nitrogen-cooled low-noise CCD mounted behind a 0.5 m spectrometer. Most of the experiments were performed at 80 K; certain measurements were also repeated at higher temperatures.

III. RESULTS

A. Photoluminescence spectra

Time-integrated spectra of the undoped sample, measured at 80 K for excitation power densities ranging from 0.23 to 2300 W cm^{-2}, which corresponds to 8×10^9 to 8×10^{13} photoexcited electron-hole pairs per pulse per cm^2, are shown in Fig. 2. The high excitation spectrum for the *p*-doped sample is also included. Below the wetting layer (WL) transition at 1.38 meV, with increased excitation power, the spectra of all samples develop four well-separated peaks that are related to transitions within the quantum dots. A curve fitting procedure with a set of Gaussian peaks yields the full width at half-maximum of 35 meV for the undoped reference sample and 46 meV for the doped structures, suggesting rather uniform dot sizes. The average energy separation between the peaks for the *p*-doped sample is found to be slightly larger compared to the undoped and *n*-doped samples (85 meV vs 77 meV). This indicates somewhat different dot sizes and/or composition, which may be induced by doping-related interdiffusion.[11]

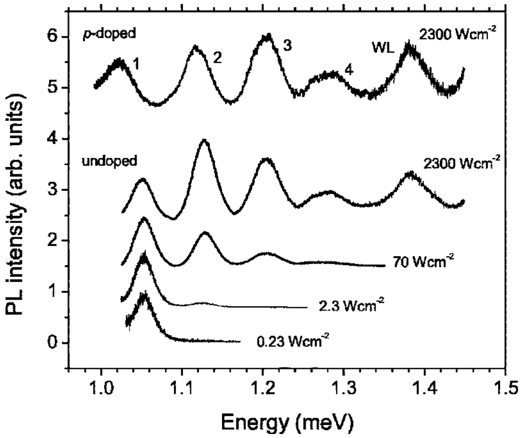

FIG. 2. 80 K time-integrated PL spectra of the *p*-doped and the undoped reference sample obtained at excitation conditions indicated in the graph. Under high excitation, four peaks related to transitions within the quantum dots are observed. The spectra are normalized to the ground state peak emission.

Considering that the PL peaks correspond to transitions between electron and hole single-particle levels with the same quantum number,[12] named *e*1-*e*4 and *h*1-*h*4, the energy difference between the peaks allows an estimation of the energy intervals between the QD levels. To that end, the ratio between the intraband electron and hole level spacings, $\Delta E_c / \Delta E_v$, is needed. Experimentally, interlevel energy spacings for conduction and valence bands in lens-shaped QDs have been studied by several groups. Schmidt *et al.*[13] measured the interlevel splittings in InAs dots using capacitance (*C*-*V*) and photoluminescence spectroscopy and found the electron level spacing $\Delta E_c = 50$ meV and the ratio between electron and hole interlevel energies, $\Delta E_c / \Delta E_v = 2$. Similar results, $\Delta E_c = 49$ meV and $\Delta E_v = 25$ meV, were obtained for InAs QDs from transmission measurements in the far- and near-infrared spectral regions.[14] Interlevel energies in lens-shaped $In_{0.5}Ga_{0.5}As$[15] and InAs[16] QDs have been studied by Chang *et al.* with *C*-*V* and PL spectroscopy. Again, the ratio $\Delta E_c / \Delta E_v \approx 2$ was found. For energetic intervals between the WL and the top QD levels, a close value of 1.6 has been reported.[17] These results encourage us to use the ratio $\Delta E_c / \Delta E_v = 2$ for our dots as well. In that case, the average interlevel spacings are about 54 and 27 meV for the conduction and valence bands, respectively. This is close to two and one InAs LO phonon energies (30 meV) with a broadening of ±3 meV induced by strain, alloy composition variations, and interfaces.[18,19] The sum of the energy difference between the WL states and the highest electron and hole levels, *e*4, *h*4, in the QDs, estimated from the PL spectra, is 109 meV. With the 2:1 ratio for the conduction and valence bands, 73 and 36 meV energy separations are obtained. These numbers match the energies of two and one GaAs LO phonons (36±3 meV).

B. Carrier transport and capture

The dynamics of carrier capture and relaxation are examined from the rising parts of PL transients. PL rise times are

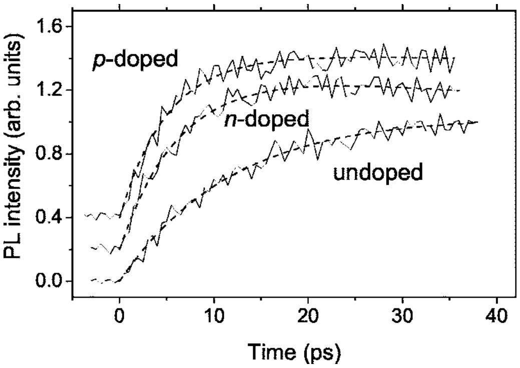

FIG. 3. Typical PL transients for the QD ground state transition ($e1$-$h1$) for different samples at high excitation power. The curves are shifted with respect to each other. Dashed lines indicate curve fits from which the PL rise times are extracted.

obtained by fitting experimental data to rising exponentials. To separate between transport, capture, and relaxation, carriers were photoexcited at 800 nm (in the barriers), 870 nm (WL), and at 960 nm (QD transition $h4$-$e4$). In the case of excitation into the barriers at a high excitation power of 230 W cm^{-2}, the PL dynamics were monitored at the energies of all peaks of the PL spectra. In the low excitation regime of 0.2–2 W cm^{-2} and for excitation into the WL and the QDs, only the ground state PL signal could be detected. The combination of these experiments performed on doped and undoped samples allows distinguishing between effects involved in the carrier transfer process, and determining mechanisms relevant for electron and hole relaxation.

Typical PL transients measured at the QD ground state transition for high excitation power are shown in Fig. 3. Figure 4(a) depicts collected rise times measured at energies of the different peaks of the PL spectra. The PL rise time for the GaAs band gap reflects hot carrier thermalization and relaxation, and is below a picosecond. When monitored at the WL band gap, the PL rise time includes transport in the barriers and capture into the WL. These WL rise times for all the samples are close (~2 ps) and are much shorter than an estimated ambipolar diffusion time over the absorption length (0.8 μm). Modulation doping, due to a partial transfer of majority carriers into the QDs, introduces potential variations and built-in fields in the vicinity of the QDs,[11] which might speed up the transport of minority carriers. However, the same short transport time is observed for all the doped and the undoped samples, indicating that modulation doping does not substantially modify the carrier transport in the barriers. Heitz *et al.*[20] have attributed such an ultrafast transport to a long-range attractive potential caused by the strain field surrounding the QDs. In any case, carrier transport in the barriers is very fast and has a minor influence on the overall process of carrier transfer into the QDs.

The next column of points in Fig. 4(a) corresponds to the PL rise times for the QD transitions $e4$-$h4$. These rise times, 4.9, 5.4, and 6.1 ps for the p-doped, n-doped, and undoped samples, besides the carrier transport, account for capture into the dots. The time for the undoped sample compares reasonably well to 8 ps measured by Sosnowski *et al.* at 10 K for the transition between the barrier and high-energy QD states in an undoped InAs QD structure.[21] To measure the capture time unaffected by transport (but then including relaxation), carrier excitation directly into the WL was performed. In this case, the PL rise time for the ground QD transition is 2, 3, and 6 ps for the p-doped, n-doped, and undoped samples, respectively [Fig. 4(b)]. Comparing PL rise times measured at the WL and $e4$-$h4$ QD transitions for the case of barrier excitation, and the time constants for the WL excitation, we estimate the "pure" capture time to be between 1.5 and 4 ps. Certain variations in the capture time between the samples may be due to different potential profiles in the vicinity of the dots induced by doping and/or strain, and varying alloy composition.

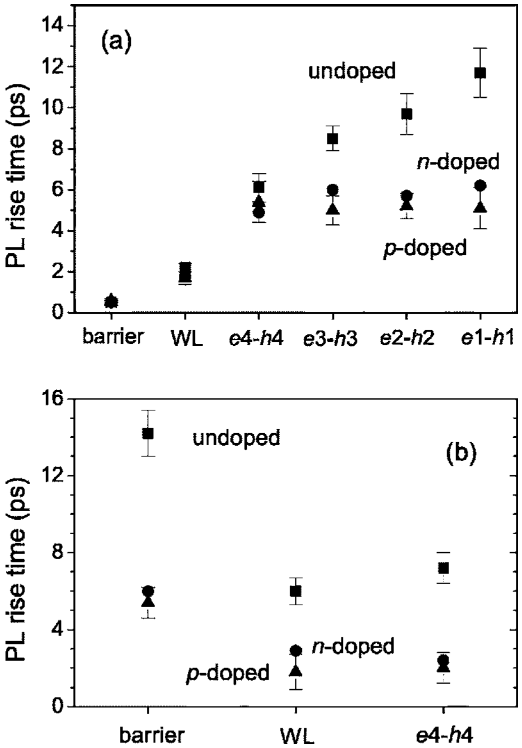

FIG. 4. Photoluminescence rise times of undoped (squares), n-doped (circles), and p-doped (triangles) samples measured at different transition energies after excitation in the barriers at high excitation power (a). Ground state PL rise times for different excitation energies measured in the low excitation regime. (b). The error bars for the n-doped sample, which are similar to those for the other samples, are omitted for viewing convenience.

Carrier capture into QDs may proceed through several mechanisms. According to the calculations, the capture time via emission of a resonant LO phonon at moderate carrier densities is below a picosecond; a two-phonon emission process is several times longer, in the range of 1–10 ps.[22] Experimental investigations have shown that carrier capture and relaxation with multiple phonon (4 LO+1 LA) emission is a fast process too, at 300 K, taking about 10 ps.[23] Capture via carrier-carrier scattering, on the other hand, at low carrier densities, is slow. It strongly depends on the carrier density and reaches the rate of the capture via single LO phonon emission at a density of ~10^{12} cm^{-2}.[24] At carrier densities of the order of 10^{8}–10^{10} cm^{-2}, this capture rate, compared to the phonon-assisted capture, is orders of magnitude lower. In

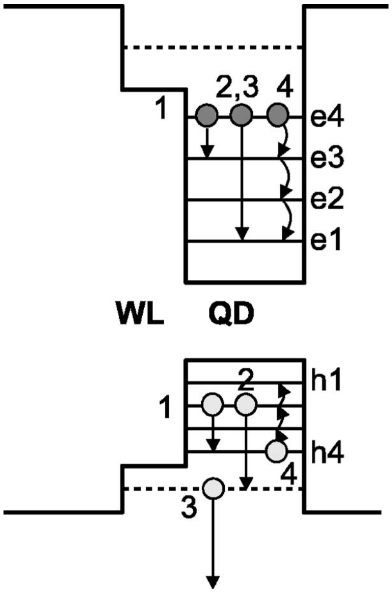

FIG. 5. Schematic flat band energy level diagram of a quantum dot structure, illustrating different carrier relaxation processes. See the text for a further discussion.

our experiments, the PL rise times at the QD ground state energy do not experience a major dependence on the excitation power, increasing from 11.7 to 14.2 ps for the undoped sample and staying at around 5 (6) ps for the *p*- (*n*-) doped samples when the excitation power density is decreased from 230 to 1 W cm^{-2} ($8\times10^{12}-4\times10^{10}$ carriers/cm^2). Such a behavior is characteristic for phonon-assisted capture. Considering peculiarities of the WL and QD level structure discussed previously, the capture takes place with the emission of one (two) GaAs-like LO phonons for the capture of a hole (electron) from the WL. One should note that carriers in the WL, either captured from the barriers or excited directly, are hot, which softens the requirements for the energetic intervals between the WL and QD levels to be strictly resonant with phonon energies.

C. Carrier relaxation

The major difference in the PL rise times for the different samples occurs at lower QD transition energies. For the undoped sample, the rise time increases with a decreasing level number [Fig. 4(a)]. In the doped structures, the rise times are about equal for all QD levels and, furthermore, significantly shorter: We measure ground state PL rise times that are twice as fast in the doped samples compared to the reference. This suggests different relaxation mechanisms in doped and undoped QDs. The viable choices for the fast relaxation are carrier-carrier (Auger) and carrier-LO phonon scattering. The faster relaxation in the doped samples suggests that the carriers built in by doping play an important role.

The carrier-carrier scattering may proceed along different lines. Before the scattering event, the carrier to be upscattered may reside in the dot or in the WL (processes 1, 2, or 3, respectively; Fig. 5). The WL situation has been considered theoretically, and scattering times between 1 and 10 ps for moderate carrier densities ($10^{10}-10^{12}$ cm^{-2}) were obtained.[25] Scattering by holes was found to be less efficient than scattering by electrons because of the larger change of the hole wave vector required for energy conservation.

In the alternative case, both carriers prior to scattering are in the QD. Here, again, we have two different options with the final state of the upscattered carrier being in the dot (situation 1, Fig. 5) or in the barrier/WL (situation 2, Fig. 5). In the first case, the energy conservation requires matching between the interlevel energies in the conduction and valence bands, which, in general, is hardly probable. However, in the dots with $\Delta E_c/\Delta E_v=2$, the energy conservation can be met, especially in the *p*-type sample where the built-in holes reside in more closely spaced levels, which, in addition, are broadened by phonon scattering. On the other hand, scattering of a hole by a built-in electron almost automatically implies WL as the final state because of a wider electron level separation.

Excitation into the WL and directly into the QDs allows distinguishing whether it is carriers in the WL or in the QDs that play the main role in the carrier-carrier scattering. In case of excitation into the WL, the carriers are present in this layer; for excitation at longer wavelengths, they are only in the QDs. Similar PL rise times for both excitation energies [Fig. 4(b)] point to the carriers built in by doping as those responsible for the fast photoexcited carrier relaxation. The small relaxation time difference for low and high excitation power when exiting into the barriers supports this interpretation.

The similar carrier scattering rates into all QD levels in the doped samples seem quite unexpected. A theoretical analysis of Auger carrier capture has shown decreasing scattering rates with increasing exchange energy.[24] Calculations of Auger relaxation have not given such a clear dependence rather the relaxation rate was found to decrease with increased excess energy of the upscattered carrier in the WL.[26] Similar (and very short) scattering times for relaxation into different levels observed in our experiments suggest different transitions for the upscattered carriers with small excess energies in the final state. Examining the level structure, one can see various possibilities for such scattering processes. For instance, for an electron being downscattered from the level *e*4 to the ground state *e*1 with an energy change of ~160 meV, the upscattered hole, initially residing in the ground state, would end up in the barrier with little or no excess energy. For an electron to be downscattered from the level *e*4 to *e*2, the hole will end up in the WL, etc. One should note that this reasoning neglects selection rules, which for dots symmetrical in the growth direction allow interband carrier transitions only between adjacent states of different parity.[25] In real structures, however, the QD symmetry is often reduced, relaxing the selection rules. The complex shape of QDs also prevents a qualitative description of the relaxation processes observed in this study, since both the level structure and the interlevel matrix elements depend critically on the QD shape and composition variations, which are not well known.

Carrier relaxation by phonon emission is also a viable relaxation mechanism, especially for the holes, for which a single phonon scattering is possible. Ultrafast hole relaxation has been observed in undoped samples, which was attributed to a close separation of the hole levels and the availability of various energy-broadened phonons.[19,21] A comparison of the doped and undoped samples allows the exploration of the relevance of this hole relaxation channel in the *n*-doped sample. In case of ultrafast phonon-assisted relaxation, the QDs in the undoped sample would be filled with holes very soon after the capture, and the sample would resemble the *p*-doped structure. This would make the subsequent electron relaxation (and increase of the PL signal) equally fast in the *p*-doped and undoped samples. Our experiments show, however, a considerable difference in the PL rise times, allowing us to rule out subpicosecond hole relaxation in the undoped structure. Thus, we conclude that in the *n*-doped sample as well as in the *p*-doped sample, the intradot carrier relaxation proceeds by scattering with the doping-induced carriers. Besides, the rate of this process does not critically depend on the number of carriers present in the dot. Experiments performed on samples with high and moderated doping provide very close relaxation times, indicating that, as soon there are carriers confined in the QDs prior to the scattering process, the carrier-carrier scattering-assisted relaxation is fast.

Different PL rise times for different QD levels in the undoped sample indicate that carrier relaxation proceeds via a cascade process (Fig. 5, case 4). At low photoexcited carrier densities, the relaxation time, i.e. the PL rise time measured for excitation into the QDs, is equal to 7 ps [Fig. 4(b)]. A similar value, estimated as the difference between the highest-energy and ground level PL rise times, is obtained for the barrier excitation at high densities [Fig. 4(a)]. A weak dependence on the photoexcited carrier density suggests that for the undoped structure the prevailing carrier relaxation mechanism is the emission of LO phonons. Recalling that the hole and electron interlevel energies are close to one and two InAs-like LO phonon energies, the relaxation process, at least at low and medium excitation powers, should be attributed to the emission of one and two LO phonons, respectively. The model of relaxation by phonon emission is supported by the temperature dependence of the relaxation time: The relaxation time at 180 K is 5.5 ps (vs 7 ps at 80 K), which is consistent with the expected increase of the relaxation rate with increased temperature.[23] Our interpretation and value of the relaxation time agrees well with recent experimental results obtained for undoped QD samples. Interband pump-intraband probe experiments[19] evidenced the cascade nature of interlevel relaxation with the interlevel relaxation time of 2.5 ps. A study based on low-temperature time-resolved PL experiments reported an electron relaxation time between the two adjacent levels equal to 1.5 ps.[27] In the case of an electron interlevel energy of 90 meV (3 InAs LO phonons), the infrared absorption studies showed a somewhat longer, but still short, interlevel electron relaxation time of 3 ps.[9] An increase of the relaxation rate in the case of resonance between the interlevel and LO phonon energies has previously been observed in InGaAs QDs.[28] The perfect match between interlevel and LO phonon energies, however, is not critical for a fast carrier relaxation. It has been shown that, because of anharmonic coupling between LO and acoustic phonons, the resonance condition is very relaxed.[28] Even the multiphonon relaxation involving optical and acoustic phonons leads to a fast (~10 ps) carrier relaxation.[23]

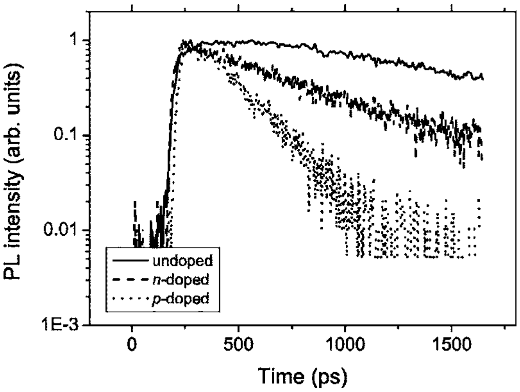

FIG. 6. Normalized photoluminescence transients corresponding to the ground state transition in different samples.

D. Carrier recombination

To evaluate the effect of modulation doping on carrier lifetimes, the evolution of the PL signal on a longer time scale was studied. Figure 6 shows that the QD ground state PL decay times for the doped samples, measured at moderately high excitation intensities (corresponding to $2\times10^{12}\ \mathrm{cm}^{-2}$ electron-hole pairs per pulse), are considerably shorter than the decay time for the reference structure. Ground state PL signals measured at 80 K decay exponentially with time constants of 1.1 ns (undoped), 558 ps (*n*-doped), and 192 ps (*p*-doped sample).[29] These decay times, within the range of used excitation intensities, are essentially independent of the excitation power. Table I lists collected PL decay times for all QD transitions, showing that the excited state lifetimes exhibit the same trends between the samples as the ground state. In general, excited state lifetimes are governed by radiative and nonradiative recombination as well as relaxation to lower-lying states.[30–32] The rapidly decreasing decay time with an increasing QD level number is consistent with our observation of fast interlevel relaxation. At these conditions, i.e., for interlevel scattering rates significantly larger than the recombination rates, excited state lifetimes are determined by the recombination of the low-energy levels alone.[31]

Since the structures are similar except for the doping, the difference in carrier lifetimes should be related to the modulation-doped layer or the doping process. One possible explanation is Auger recombination in the presence of addi-

TABLE I. Photoluminescence decay times at 80 K for different QD transitions.

	Transitions			
Sample	*e*1-*h*1	*e*2-*h*2	*e*3-*h*3	*e*4-*h*4
Undoped	1100 ps	207 ps	59 ps	28 ps
n-doped	558 ps	129 ps	61 ps	27 ps
p-doped	192 ps	64 ps	28 ps	15 ps

tional carriers in the doped dots. However, at high excitation conditions, the QD states are almost completely filled with carriers in all samples, regardless of doping. Auger recombination would therefore have the same effect on carrier lifetimes in doped and undoped samples. Instead, decreased PL decay times should be explained by nonradiative recombination at defects introduced during the QD growth in the presence of the modulation-doped layers.

While the exact nature of the defects is unknown, it is still possible to speculate about their origin. It has been shown that the presence of doping-induced excess carriers can increase the number of certain kinds of defects because of the free carrier influence on interdiffusion occurring during the dot overgrowth.[11] For example, the concentration of interstitials, I_{III}, which can become positively charged, increases in *p*-doped structures, and the concentration of V_{III} vacancies increases in *n*-doped structures. For Si doping and at high doping concentrations, the $\text{Si}_{\text{Ga}}{}^{+}$ donors, occupying Ga sites, can effectively be compensated by $\text{Si}_{\text{As}}{}^{-}$ acceptors or other defects.[33] Taking this into account, we expect the doping-related defect density to be lower in the *n*-doped sample than in the *p*-doped sample. This could result in a smaller number of point defects, acting as nonradiative recombination centers.[34–36] Although no quantitative measure of the defect origin or density can be given, the differences in the doped samples' decay times qualitatively suggest a somewhat lower defect concentration in the *n*-doped sample, in agreement with the model of Ref. 11. Furthermore, the lack of saturation of the nonradiative recombination channel at high excitation (apparent from the carrier lifetimes, which are excitation independent) indicates that the defects, present in the doped samples, act as efficient recombination centers.

IV. CONCLUSIONS

We have presented a time-resolved photoluminescence study of *n* and *p* modulation-doped InAs/GaAs quantum dot structures and compared them to an undoped reference sample. Experiments with excitation and detection energies varying through the barrier, wetting layer, and QD states allowed a detailed examination of carrier capture and relaxation processes. The carrier capture into the highest QD levels was found to proceed via LO phonon scattering in all the structures. The subsequent carrier relaxation in the doped structures is governed by the doping-induced carriers by means of carrier-carrier scattering, while in the undoped sample relaxation progresses in a cascaded manner by the emission of optical phonons. Doping affects the carrier lifetimes as well: Doping and the subsequent interdiffusion leads to an increased amount of nonradiative recombination. As a result, carrier lifetimes are reduced, especially in the *p*-doped sample.

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