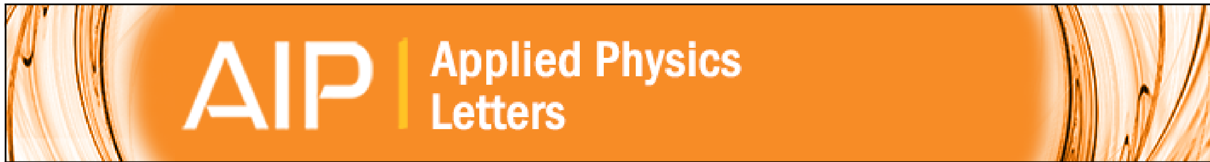

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Ultrafast electron capture into *p*-modulation-doped quantum dots

K. Gündoğdu,[a] K. C. Hall, and Thomas F. Boggess
Department of Physics and Astronomy and Optical Science and Technology Center, The University of Iowa, Iowa City, Iowa 52242

D. G. Deppe and O. B. Shchekin
Microelectronics Research Center, Department of Electrical and Computer Engineering, The University of Texas at Austin, Austin, Texas 78712-1084

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Electron and hole relaxation kinetics are studied in modulation-doped InAs quantum dots using femtosecond time-resolved photoluminescence experiments. We demonstrate that, as a result of doping, carrier relaxation from the barrier layers to the quantum dot ground states is strongly enhanced due to rapid electron–hole scattering involving the built-in carrier population. Results for *p*-doped quantum dots reveal a threefold decrease in the room-temperature electron relaxation time relative to corresponding undoped quantum dots. Our findings are promising for the development of high-speed, GaAs-based quantum dot lasers with modulation speeds in excess of 30 GHz. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1815371]

The discrete energy level structure in semiconductor quantum dots (QDs) offers several advantages over higher-dimensional systems for application to high-performance semiconductor laser technology, including the potential for lower threshold current, reduced thermal sensitivity, and higher modulation speed.[1–3] Nonequilibrium carrier dynamics play a central role in determining the performance limitations of QD lasers,[4] and have therefore been the subject of a considerable research effort in recent years.[5–12] The rate of carrier energy relaxation from the barrier to the QD ground states, which determines a fundamental limit for the laser modulation speed,[4] has been studied extensively using ultrafast optical techniques such as pump probe spectroscopy and time-resolved photoluminescence (TRPL) experiments. The short relaxation times observed by several groups[5,8–10] are promising for high-speed QD laser operation, and have inspired more recent approaches to reduce this relaxation time, including QD electronic structure engineering[7] and the use of tunneling for direct electron injection to the QD ground state.[12]

The incorporation of *p*-doping in the QD laser active region is predicted to provide considerable improvements in the room-temperature modulation speed of QD lasers through an increase in the differential gain.[13–15] This gain enhancement arises from the built-in hole population in the QDs, which reduces the influence of thermal effects on the ground state hole occupation, and is expected to lead to laser modulation speeds in excess of 30 GHz.[14] However, the influence of doping on the carrier capture and relaxation kinetics in these QD heterostructures is not known,[14] despite the importance of these processes for achieving high-frequency QD laser operation.

Here we employ TRPL techniques to study carrier capture and relaxation dynamics in modulation-doped self-assembled InAs QDs. Through a comparison of carrier kinetics in undoped and *p*- or *n*-doped QDs,[16] we demonstrate that electron–hole scattering involving the built-in carriers on the doped QDs leads to a strong enhancement in the carrier relaxation rate from the GaAs barriers to the QD ground states. For *p*-doped QDs, we observe a threefold enhancement in the room-temperature electron relaxation rate relative to the undoped QDs, a result that is promising for the development of low-threshold, high-speed, GaAs-based lasers using semiconductor QDs.

The self-assembled InAs QDs were grown by molecular-beam epitaxy under identical conditions for structures with and without modulation doping. Each sample contains a single layer of QDs in the center of a 30 nm layer of GaAs, cladded on both sides with 340 nm AlGaAs barriers. For all samples, the QDs are formed by deposition of 2.7 monolayers of InAs on 15 nm of undoped GaAs. Atomic force microscopy and transmission electron microscopy measurements indicate that the QDs are 25 nm in diameter and 3 nm in height, with an areal density of 3×10^{10} cm^{-2}. For the undoped QDs, 15 nm of undoped GaAs was deposited over the QDs, and for the modulation-doped structures, 12 nm of undoped GaAs was followed by 3 nm of GaAs doped with Si (*n*-doped QDs) or Be (*p*-doped QDs) at a density of 2×10^{18} cm^{-3}. This doping density corresponds to $\sim$20 carriers per QD. Results of continuous-wave photoluminescence (CWPL) experiments on these QDs are shown in Fig. 1. The excess carriers in the doped QDs cause the ground state optical transition to shift to shorter (longer) wavelengths relative to the undoped QDs for p (n) doping due to Coulomb charging effects.[18–20] The observed energy shifts of several tens of meV indicate substantial carrier accumulation inside the QDs. Two confined optical transitions are observed in the CWPL spectra, which are separated by more than 60 meV for all three QD structures. In the TRPL experiments, 100 fs, 1.42 eV pulses from a Ti:sapphire oscillator are used to excite electron–hole pairs in the GaAs barriers, and the PL from the ground state optical transition in the QDs is time-resolved using sum frequency generation with a second Ti:sapphire pulse in a $KNbO_3$ crystal. The photoexcited carrier density is estimated using the measured fluence and the absorption coefficient of bulk GaAs at 1.42 eV.[21]

The initial transient 77 K PL from the ground state optical transition in the QDs is shown in Fig. 2. The lowest QD

[a]Electronic mail: kenan-gundogdu@uiowa.edu

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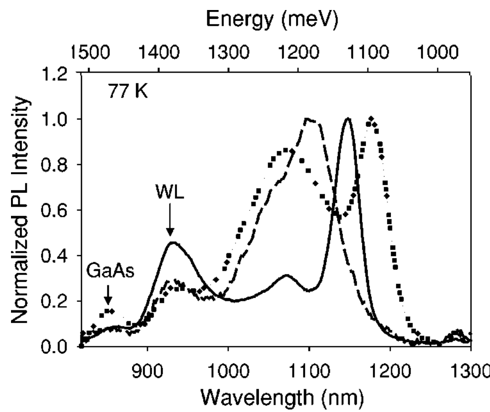

FIG. 1. Results of CWPL experiments on *p*-doped QDs (dashed curve), *n*-doped QDs (dotted curve) and undoped QDs (solid curve). The pump wavelength is 870 nm. The data are normalized to the peak emission from the ground state optical transition for each QD structure (Ref. 17). Peaks associated with emission from the GaAs substrate and InAs wetting layer (WL) are also indicated.

electron and hole energy levels must be occupied to observe ground state PL. As a result, the rise of the TRPL signals in Fig. 2, which is well-characterized by a single exponential in all cases, indicates carrier capture and energy relaxation from the GaAs barriers to the QD ground state. For the modulation-doped QDs, the lowest few electron (*n*-doped QDs) or hole (*p*-doped QDs) levels are occupied prior to optical excitation of electron–hole pairs into the GaAs barriers. Experiments on modulation-doped QDs therefore allow the relaxation dynamics of electrons and holes to be studied separately. In contrast, for the undoped QDs, ground state emission results from the capture and relaxation of optically injected electron–hole pairs. The results in Fig. 2 indicate that carriers relax more rapidly into the doped QDs regardless of the species of carrier involved. The exponential fits in Fig. 2 indicate relaxation times of (450 ± 20) fs (*p*-doped QDs), (1.4 ± 0.2) ps (*n*-doped QDs), and (4.8 ± 0.4) ps (undoped QDs).

The results in Fig. 2 correspond to an optical excitation density of less than one electron–hole pair per QD. In this case, the primary difference between the TRPL experiments on the doped and undoped QDs is the existence of a large built-in electron (hole) population in the *n*-doped (*p*-doped) QDs prior to optical excitation. The results in Fig. 2 therefore

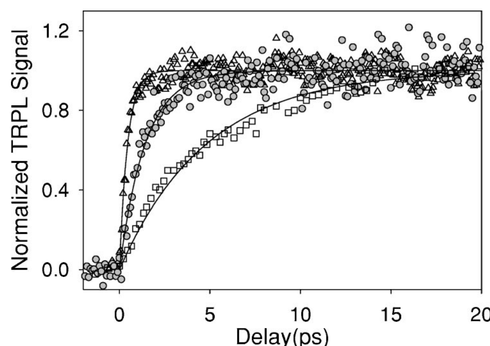

FIG. 2. (a) TRPL results for *p*-doped QDs (triangles), *n*-doped QDs (circles), and undoped QDs (squares) at 77 K with an optical excitation density of 0.6 electron–hole pairs per QD at 1.42 eV. The solid curves indicate fits of the PL rise to a single exponential with time constants of (450 ± 20) fs (*p*-doped QDs), (1.4 ± 0.2) ps (*n*-doped QDs), and (4.8 ± 0.4) ps (undoped QDs).

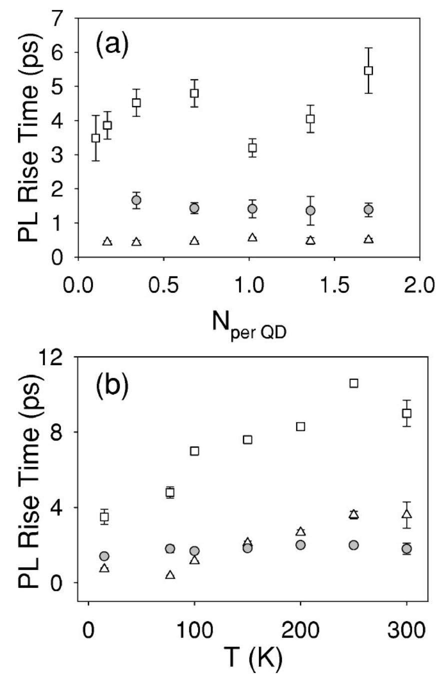

FIG. 3. (a) Time constants extracted from single exponential fits to the rising edge of the TRPL versus the number of optically injected electron–hole pairs per QD ($N_{\text{per QD}}$) at 77 K for *p*-doped QDs (triangles), *n*-doped QDs (circles), and undoped QDs (squares); (b) Temperature dependence of the PL rise time for low excitation density.

suggest that the faster carrier relaxation into the doped QDs is due to a rapid electron–hole scattering process in which an optically injected electron (hole) undergoes energy relaxation through scattering with a built-in hole (electron) in the *p*-doped (*n*-doped) QDs. This assessment is consistent with the observation of no dependence of the measured PL rise times on the optical excitation density for densities well-below the doping density ($\sim$20 carriers per QD), as shown in Fig. 3(a).

Such a carrier-mediated relaxation process was discussed earlier in the context of electron capture into undoped InAs QDs.[5] Sosnowski *et al.* showed that, despite the absence of energy conserving relaxation processes by phonon emission due to the large energy spacing of the QD electron states,[6] rapid electron relaxation to the QD ground state is possible provided a hole exists in the QD with which the electron can scatter to release its excess energy.[5,6] This scattering process is rapid because the hole energy levels are closely spaced and strongly broadened by phonon scattering, thereby providing a wide phase space of available energy-conserving transitions. Since the experiments of Sosnowski *et al.* involved undoped QDs, the single hole participating in the scattering process in each QD was generated by optical excitation and relaxed into the QD more quickly than the electron due to the availability of phonon-mediated relaxation channels. The time for electron capture and energy relaxation extracted from the low-temperature experiments of Sosnowski *et al.* (8 ps) is similar to our findings for the undoped QDs in Fig. 2. *In contrast, for the p-doped QDs, we observe a dramatic reduction in the electron relaxation time, which reaches the subpicosecond regime for temperatures below 100 K*. The rapid electron relaxation we observe in the *p*-doped QDs is attributed to the large built-in hole population, which strongly enhances the rate of electron–hole scattering.

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For the *n*-modulation doped QDs, the lowest electron levels are filled prior to optical excitation, and therefore the rise time of the ground state PL is dictated by the time for relaxation of holes from the GaAs barriers to the ground hole level in the QDs. Since the hole levels are more closely separated in energy than the electron levels [holes: ~10 meV; electrons: ~50 meV (Ref. 22)], holes may undergo relaxation *either* through phonon emission or through scattering with the built-in electron population. In order to determine which of these mechanisms is dominant, TRPL experiments were performed for a range of temperatures [Fig. 3(b)]. If phonon emission were responsible for the hole energy relaxation in our QDs, the rise time of the PL for the *n*-doped QDs would decrease with increasing temperature,[11] in contrast to the results in Fig. 3(b), which indicate no discernible temperature dependence.[23] No temperature dependence is expected for the rate of hole energy relaxation via electron–hole scattering due to the large energy spacing of the electron levels, which implies that the built-in electron distribution in the *n*-doped QDs will not be sensitive to changes in sample temperature, in agreement with the experimental findings in Fig. 3(b). As a result, we attribute the 1.4 ps relaxation time observed for holes in the *n*-doped QDs to electron–hole scattering involving the built-in electrons on the *n*-doped QDs.[24]

The built-in hole distribution in the *p*-doped QDs is sensitive to thermal excitation due to the close spacing of the hole energy levels. As discussed earlier for undoped InAs QDs,[4] this thermal spreading of the hole population across the closely spaced hole levels reduces the electron–hole scattering rate due to increased filling of the excited hole states. This is consistent with our observation of an increase in the electron relaxation time with temperature for both the *p*-doped and undoped QDs [Fig. 3(b)].

Our findings, which indicate that *p*-doping increases the electron relaxation rate into the QDs, have strong implications for current efforts to improve the modulation characteristics of QD lasers. The incorporation of *p*-doping in the active region of a QD laser has already been shown to enhance several other features of laser performance at elevated temperatures, including providing a reduction in the thermal sensitivity,[25] a lower threshold current, and an enhanced differential gain.[15] These improvements arise from the built-in hole population in the QDs, which ensures occupation of the ground state hole levels at high temperatures despite the close spacing of the hole energy levels. The larger room-temperature differential gain in *p*-doped QD lasers should lead to a considerable enhancement in the maximum laser modulation speed;[13,14] however, nonequilibrium carrier dynamics will play a crucial role in achieving these high speeds. As shown in Fig. 3(b), we observe a threefold reduction in the room-temperature electron relaxation time from the GaAs barriers to the QD ground state with *p*-doping. These high electron relaxation rates, which arise from electron–hole scattering with the built-in hole population in the *p*-doped QDs, will support the development of low-threshold, high-speed GaAs-based QD lasers.

In summary, we have applied time-resolved photoluminescence techniques to the study of carrier relaxation kinetics in undoped and modulation-doped self-assembled InAs QDs. The existence of built-in carriers in the doped QDs have allowed us to separately measure the relaxation dynamics of electrons and holes. Our findings indicate that the carrier relaxation rate from the GaAs barriers to the QD ground states is strongly enhanced in doped QDs through rapid electron–hole scattering processes involving the built-in carriers. For *p*-doped QDs, subpicosecond electron relaxation is observed at low temperatures, which increases to a few picoseconds at room temperature, representing a threefold reduction over the corresponding undoped QDs. Our findings are promising for the development of QD lasers with modulation speeds in excess of 30 GHz.

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