Ultrafast carrier capture and relaxation in modulation-doped InAs quantum dots

K. W. Sun, A. Kechiantz, B. C. Lee, and C. P. Lee

Articles you may be interested in

Carrier relaxation dynamics in InAs/GaInAsP/InP(001) quantum dashes emitting near 1.55m
Appl. Phys. Lett. **103**, 083104 (2013); 10.1063/1.4818759

Ultrafast release and capture of carriers in InGaAs/GaAs quantum dots observed by time-resolved terahertz spectroscopy
Appl. Phys. Lett. **94**, 262104 (2009); 10.1063/1.3158958

Energy-dependent carrier relaxation in self-assembled InAs quantum dots
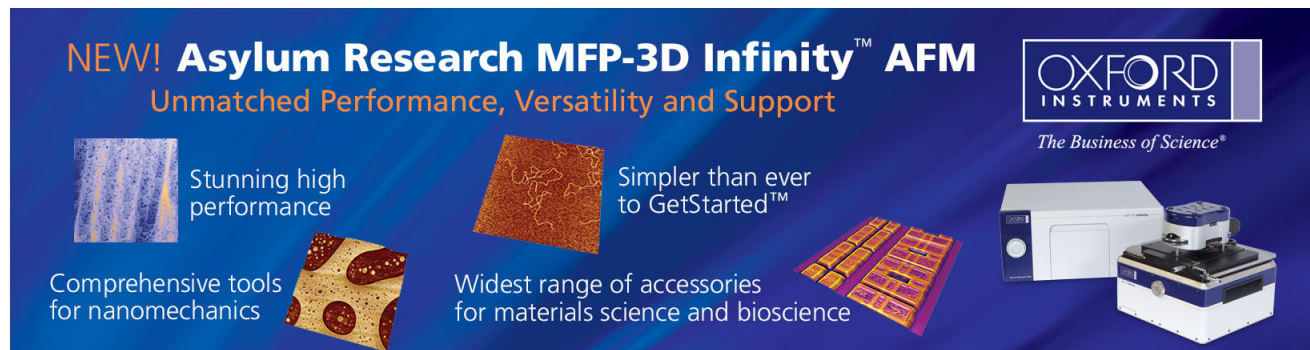J. Appl. Phys. **103**, 124311 (2008); 10.1063/1.2947599

Excited-state dynamics and carrier capture in InGaAs/GaAs quantum dots
Appl. Phys. Lett. **79**, 3320 (2001); 10.1063/1.1418035

Energy level control for self-assembled InAs quantum dots utilizing a thin AlAs layer
Appl. Phys. Lett. **78**, 3247 (2001); 10.1063/1.1373410

Ultrafast carrier capture and relaxation in modulation-doped InAs quantum dots

K. W. Sun[a)] and A. Kechiantz[b)]
Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsin Chu, Taiwan 300, Taiwan

B. C. Lee
Center for Nano Science and Technology, National Chiao Tung University, Hsin Chu, Taiwan 300, Taiwan

C. P. Lee
Department of Electronics Engineering and Institute of Electronics, National Chiao Tung University, Hsin Chu, Taiwan 300, Taiwan

We report investigations on carrier capture and relaxation processes in undoped and modulation-doped InAs/GaAs self-assembled quantum dots (QDs) by using time-resolved spectroscopy technique with a time resolution of ~ 200 fs. We find that carrier capture and relaxation in the ground state of the charged QD are faster compared to the undoped dots even at an excitation level as low as 1×10^{10} cm^{-2}. It is attributed to the triggering of the vibrating polarization field induced by the presence of cold carriers in the doped dots. The rate of an electron been captured by a positively charged QD is also calculated based on our proposed model. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2197309]

The study of carrier relaxation and capture in InAs/GaAs quantum dots (QDs) has attracted much attention[1–4] due to their wide physical interests and their potential device applications. It has been predicated that the discrete atomiclike energy levels may inhibit the efficient carrier relaxation by single phonon emissions.[5,6] The systematically longer PL rise times observed in the higher excited states of InAs/GaAs QDs by Yuan *et al.*[7,8] were interpreted in the framework of sequential state filling, resulting from fast trapping and intradot relaxation. The experimental results of intraband relaxation via polaron decay in InAs QDs were reported over a wide energy range from 40 to ~ 60 meV in Ref. 9. Their measured energy dependent decay time of the transmission change ranged from ~ 65 to ~ 45 ps. Few experiments have examined carrier and spin dynamics in charged QDs. Observation of quantum beats with unusual polarization properties in the PL of InP QDs was reported by Kozin *et al.*[10] In Ref. 11, the spin state of the resident electron in *n*-doped InAs QDs can be manipulated using nonresonant optical excitation. Recently, in the work done by Gündoğdu *et al.*,[12,13] the carrier capture and relaxation to the ground state are much faster in the highly charged dots compared to the neutral dots. The enhancement of carrier capture and relaxation is attributed to the rapid electron-hole scattering involving the built-in carrier population.

In this letter, we investigate and compare the carrier capture and relaxation processes in undoped and lightly doped InAs/GaAs QDs using luminescence upconversion spectroscopy with ~ 200 fs time resolution. We observed ultrafast carrier capture and relaxation in charged QDs' ground states at very low doping concentrations and at low excitation level.

The InAs/GaAs QD samples were grown by using a solid source molecular beam epitaxy (MBE) machine.[14] The *n*-doped samples contain a Si-delta doping layer 2 nm below the QD layer with nominal densities of about 2×10^{10} cm^{-2}. For the *p*-doped samples, a Be-delta doping layer was placed 2 nm below the QD layer with nominal densities of about 2×10^{10} cm^{-2}. Atomic force microscopy (AFM) and transmission electron microscopy (TEM) images of the self-assembled quantum dots reveal a QD density of $\sim 2 \times 10^{10}$ cm^{-2} and an average base width and height for the dots of approximately 20 and 5 nm, respectively. Free carriers from the doped layer accumulated in the low energy states within the InAs QDs. In the lightly *n*- (*p*-) doped samples, most of the dots contain only a single electron (or hole). A QD infrared photodetector structure was also fabricated with the same doping scheme used in these experiments. The intraband absorption measurements and responsivity spectra from the device indicated that the doping in our samples was effective.

The measurements of carrier dynamics were performed by time-resolved photoluminescence.[14] A femtosecond Ti:sapphire laser was operated at 770 nm with full width at half maximum (FWHM) of about 18 meV to excited carriers in the GaAs barrier. According to the focus spot size and the absorption depth at the photoexcitation wavelength, the laser pumping power was adjusted to give injected carrier densities from 1×10^{10} (low) to 5×10^{11} cm^{-2} (high). The room temperature time-integrated PL spectra of the undoped QD sample taken at an excitation wavelength of 770 nm and intensity of 1×10^3 W/cm^2 (corresponding to an injected carrier density of $\sim 10^{11}$ cm^{-2}) are shown in Fig. 1. The spectral lines centered at 872 and 930 nm are attributed to the band gap energies of the GaAs and wetting layer, respectively. Three spectrally well-separated PL lines at longer wavelengths, as shown in Fig. 1(b), are due to electron-hole re-

[a)]Author to whom correspondence should be addressed; electronic mail: kwsun@mail.nctu.edu.tw
[b)]Permanent address: Scientific Research Division, State Engineering University of Armenia, Yerevan, Armenia.
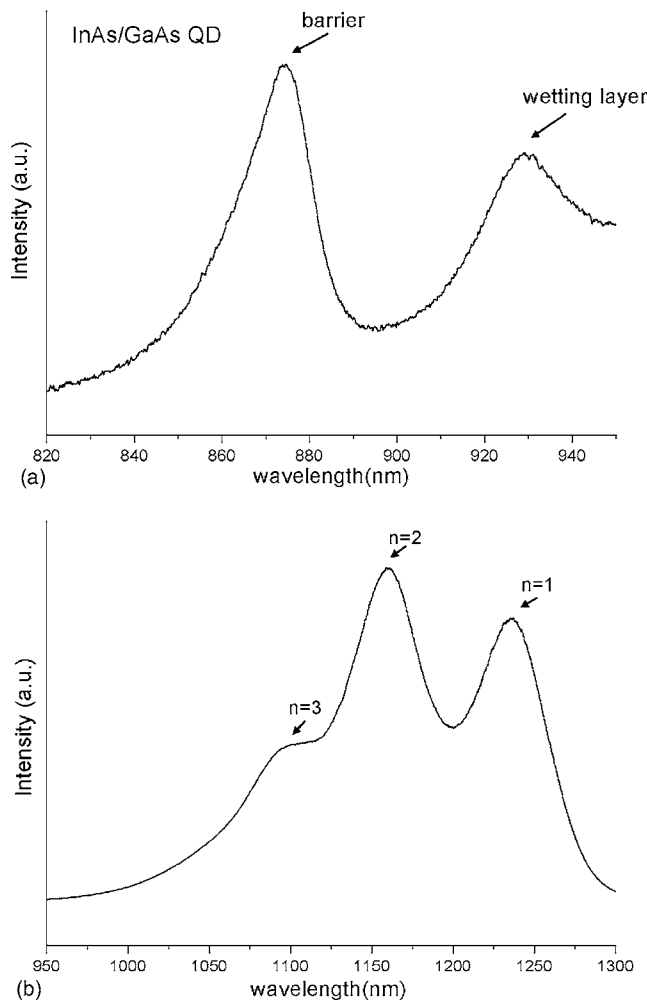
FIG. 1. Room temperature PL spectra of InAs/GaAs self-assembled QDs, displaying GaAs barrier, wetting layer, and excited state radiative recombination in wavelength range from (a) 820 to 950 nm and (b) 950 to 1300 nm. Spectra were excited at 788 nm with a self-mode-locked Ti:sapphire laser.

combination between distinct QD confined states. Only the $n=1$ peak is observed at low excitation intensity (less than 10 W/cm^{-2}) and it is assigned as the QD ground electron to ground hole transition. The center wavelengths of the spectral peak from $n=1$ to $n=3$ are 1240, 1150, and 1080 nm, respectively. In the PL studies on doped QDs, due to the lightly doping, we did not observe significant changes in the spectra line shape or shifting of ground-state optical transition. Carrier capture and relaxation to the QD's ground level were examined as a function of excitation density and temperature by measuring PL rise times at the energy of QD ground state identified in the steady state PL spectra. The time evolution of the PL signal then follows from the analysis of the rate equations,

$$I(t) \propto A[1 - \exp(-t/\tau_r)]\exp(-t/\tau_d), \qquad (1)$$

where τ_r and τ_d are the PL rise and decay time constants.

In Fig. 2 we show the PL transients detected at the ground state of undoped QDs for the first 20 ps. PL spectra measured at three different excitation levels (low, moderate, and high) are displayed in parallel for comparison. At an excitation density as low as 1×10^{10} cm^{-2}, the PL intensity shows a rise time of approximately 5 ps. The PL rise times accelerate as the excitation power increases and reach a
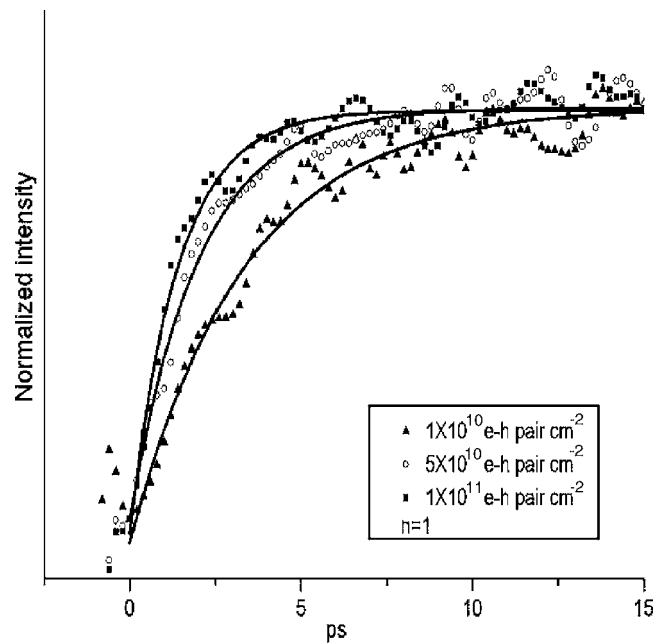

FIG. 2. Time-resolved PL intensity measured at the energy of the undoped QDs' ground states at three different excitation levels and at low temperature. The solid curves indicate fits of the PL rise to a single exponential.

value of less than 1 ps at a photoexcited carrier density $\geqslant 1 \times 10^{11}$ cm^{-2}. It is believed that the carrier density dependence of the ultrafast relaxation is due to Auger-like carrier-carrier scattering.[1,11]

Experiments on modulation-doped QDs allow the relaxation dynamics of electrons and holes to be investigated separately. At low temperature, low excitation densities, the photogenerated carriers do not significantly perturb the well-defined Fermi distribution of doped cold carriers. Therefore, the luminescence dynamics is dominated by the electron (hole) dynamics in *p*- (*n*-) doped QDs. For samples with doping density of 2×10^{10} cm^{-2}, only the lowest electron (*n*-doped QDs) or hole (*p*-doped QDs) level is occupied prior to the optical excitation. The initial PL transient at the ground state in the doped QDs is shown in Fig. 3. The major
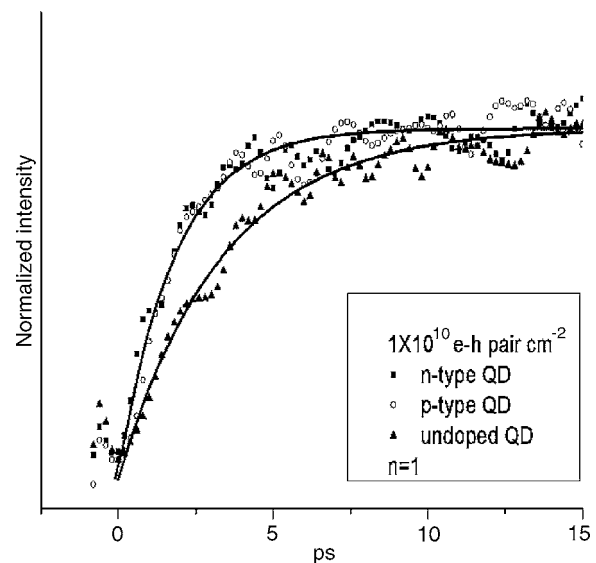

FIG. 3. Results of time-resolved PL experiments at 77 K for *p*-doped (circles), *n*-doped (squares), and undoped QDs (triangles) at an optical excitation level of only one electron-hole pair per dot.
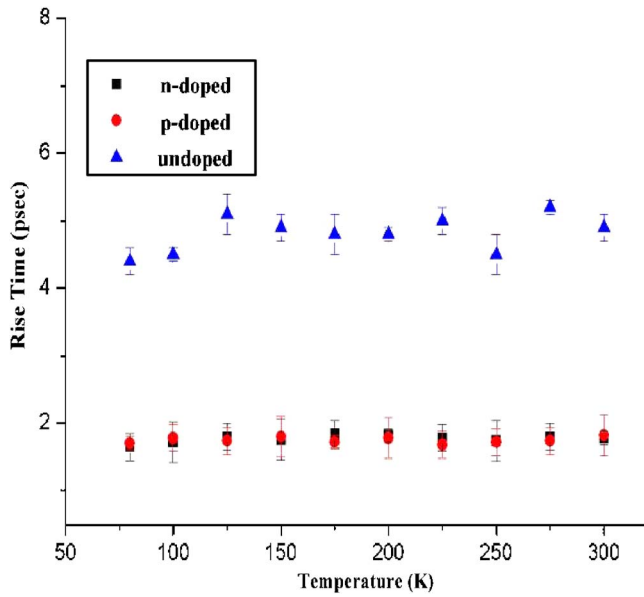
FIG. 4. (Color online) Temperature dependence of the PL rise times extracted from the single exponential fits to the rising edge of time-resolved PL under low excitation level.

difference of PL transients between charged and uncharged QDs is the presence of small built-in electron (hole) population in the *n-* (*p-*) doped QDs prior to optical excitation. However, the fits of the PL transients in Fig. 3 indicate capture times of about 1.7 ps for charged QDs which are more rapid than in undoped QDs, regardless of the species of cold carrier involved. In contrast to the earlier reports in highly charged QDs,[12,13] the total carrier densities in our experiments are only on the order of $\sim 2 \times 10^{10}$ cm^{-2}. The calculated Auger scattering rate[15] was about 2×10^{10} s^{-1} for a plasma density of $\sim 10^{10}$ cm^{-2} and a quantum dot lateral size of 20 nm. At such low excitation intensity, it is unlikely that the Coulomb scattering within the electron-hole plasma is responsible for the ultrafast carrier capture and relaxation observed in our charged QD experiments.[16,17] Another possible mechanism for carrier capture and relaxation under low excitation is through carrier-phonon coupling[18] by which carriers relax sequentially inside the QDs via carrier-phonon interaction. In that case, the capture and relaxation time should be slower and reveal temperature dependence. However, in Fig. 4, we found no discernible temperature dependence on the PL rise time for charged and uncharged QDs. The above results indicate that neither carrier-carrier scattering nor phonon scattering was responsible for the accelerated carrier capture observed in our experiments.

We have developed a theoretical model of carrier capture by taking into account the subtle difference between scattering of carriers into continuous and localized electronic states when the center of the scattering is charged. In response to the localized (doped) charges in the dots, the lattice ions and electrons bonded to those ions were displaced. This displacement produces local strain of crystal lattice and atomic-size electrical dipoles. Due to the cumulative field of those dipoles around the dot, a polarization field must be induced around the QDs. During the scattering, carriers encounter two electric fields of opposite signs: positive (negative) field of a bare hole (electron) confined within the dot and screening field of local polarizations around the dot. When the electric field induced strain around QDs suddenly disap

peared due to the capture of an electron or a hole, local strain of crystal lattice must trigger vibrations of ions and bond electrons around the dot to relax strain and to release energy. By applying time-dependent perturbation calculations on the case of a mobile electron scattered on the polarization field produced by positively charged (*p*-doped) dots, we obtain the carrier capture rate of

$$\tau = \frac{16\pi^2 \varepsilon_o^2 \hbar m^* (\omega_{kc}^2 - \omega_m^2) R_0^{5/2}}{e^4 (1 - 1/\varepsilon_s)^2 n^{1/2} |K(k)|^2}. \tag{2}$$

Derivation of Eq. (2) can be found in Ref. 19. For given parameters in our *p*-doped QD experiments ($E_k = 35$ meV, $\Omega = 2 \times 10^3$ nm^3, $K(k) = 0.41$, $qR_0 = 1.5$, $kR_0 = 2$, $\omega_m \Delta t = 2.13$, and $n = 2 \times 10^{16}$ cm^{-3}), Eq. (2) gives a capture rate of $\tau = \sim 1.7$ ps.

In conclusion, we have investigated carrier capture and relaxation in undoped and doped InAs/GaAs QDs. We observed faster capture and relaxation processes in charged QDs compared to the undoped case at very low excitation densities. Our results suggest that, under low excitation intensity and low doping level, the relaxation of polarization field induced by the confined charge in the quantum dot is the dominant factor for the acceleration of carrier capture and relaxation in charged QDs.

[1]J.-Y. Marzin, J.-M. Gérard, A. Izraël, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994).
[2]G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz, and P. M. Petrov, Appl. Phys. Lett. **64**, 2815 (1994).
[3]R. Ferreira and G. Bastard, Appl. Phys. Lett. **74**, 2818 (1999).
[4]I. Magnusdottir, A. V. Uskov, R. Ferreira, G. Bastard, J. Mørk, and R. Tromborg, Appl. Phys. Lett. **81**, 4318 (2002).
[5]U. Bockelmann and G. Bastard, Phys. Rev. B **42**, 8947 (1990).
[6]H. Benisty, C. M. Sotomayor Torres, and C. Weisbuch, Phys. Rev. B **44**, 10945 (1991).
[7]Z. L. Yuan, E. R. A. D. Foo, J. F. Ryan, D. J. Mowbray, M. S. Skolnick, and M. Hopkinson, Physica B **272**, 12 (1999).
[8]Z. L. Yuan, E. R. A. D. Foo, J. F. Ryan, D. J. Mowbray, M. S. Skolnick, and M. Hopkinson, Phys. Status Solidi B **224**, 409 (2001).
[9]E. A. Zibik, L. R. Wilson, R. P. Green, G. Bastard, R. Ferreira, P. J. Phillips, D. A. Carder, J.-P. R. Wells, J. W. Cockburn, M. S. Skolnick, M. J. Steer, and M. Hopkinson, Phys. Rev. B **70**, 161305(R) (2004).
[10]I. E. Kozin, V. G. Davydov, I. V. Ignative, A. V. Kavokin, K. V. Kavokin, G. Malpuech, Hong-Wen Ren, M. Sugisaki, S. Sugou, and Y. Masumoto, Phys. Rev. B **65**, 241312(R) (2002).
[11]S. Cortez, O. Krebs, S. Laurent, M. Senes, X. Marie, P. Voisin, R. Ferreira, G. Bastard, J.-M. Gerard, and T. Amand, Phys. Rev. Lett. **89**, 207401 (2002).
[12]K. Gündoğdu, K. C. Hall, Thomas F. Boggess, D. G. Deppe, and O. B. Shchekin, Appl. Phys. Lett. **84**, 2793 (2004).
[13]K. Gündoğdu, K. C. Hall, T. F. Boggess, D. G. Deppe, and O. B. Shchekin, Appl. Phys. Lett. **85**, 4570 (2004).
[14]K. W. Sun, J. W. Chen, B. C. Lee, C. P. Lee, and A. M. Kechiantz, Nanotechnology **16**, 1530 (2005).
[15]U. Bockelmann and T. Egeler, Phys. Rev. B **46**, 15574 (1992).
[16]A. V. Uskov, J. Mclnerney, F. Adler, H. Schweizer, and M. H. Pilkuhn, Appl. Phys. Lett. **72**, 58 (1998).
[17]T. R. Nielsen, P. Gartner, and F. Jahnke, Phys. Rev. B **69**, 235314 (2004).
[18]B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11532 (1996).
[19]See EPAPS Document No. E-APPLAB-88-292616 for derivation of Eq. (2). This document can be reached through a direct link in the online article's HTML reference section or via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html).