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E. W. Bogaart,[a] R. Nötzel, Q. Gong, J. E. M. Haverkort, and J. H. Wolter
COBRA Inter-University Research Institute, Eindhoven University of Technology, Department of Applied Physics, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

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The energy and excitation density dependence of the carrier dynamics in self-assembled InAs/InP quantum dots (QDs), emitting in the 1.55 μm wavelength region, is investigated by means of time-resolved pump-probe differential reflection spectroscopy at room temperature. We observe ultrafast carrier capture and subsequential carrier relaxation into the QD ground state within 2.5 ps. The carrier lifetime in the QDs strongly depends on the QD optical transition energy within the QD ensemble as well as the carrier density, and ranges from 560 up to 2600 ps. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1915527]

In the last decade, many studies have been devoted to the investigation of low-dimensional semiconductor structures, due to their importance for applications in high-performance semiconductor devices.[1–4] Hereby, great progress has been made in the growth and the optical properties of self-assembled quantum dots (QDs). Especially, QDs emitting at the 1.55 μm telecommunication wavelength are of great interest. Recently, Gong *et al.*[5,6] have shown that the luminescence wavelength of InAs QDs grown on an InP (100) substrate is reproducibly tuned by the deposition of a thin GaAs[5] or GaP[6] interlayer between the GaInAsP buffer layer and the QD layer. The central wavelength is continuously tuned from 1.6 down to 1.5 μm at room temperature (RT). In these studies,[5,6] the focus has been on the continuous wave optical properties of the QDs, rather than the time characteristics, e.g., the optical response. However, for the application of these structures in optoelectronic devices, accurate knowledge of the optical response is required. The dynamic response, determined by the carrier capture and carrier lifetime, is crucial for the performance, in particular the modulation speed of optoelectronic devices and for their modeling.

In this letter, we report on the experimental study of the carrier dynamics of self-assembled InAs/InP QDs with a thin GaAs interlayer[5] using two-color pump-probe time-resolved differential reflection spectroscopy (TRDR),[7] i.e., time-resolved bleaching, at RT. Using nonresonant excitation, carriers are excited within the barriers surrounding the QDs and diffuse from the barriers towards the QDs where they are captured and relax to the QD ground state. The probe beam monitors the absorption bleaching in the vicinity of the QD ground state transition. This technique allows us to study directly the occupation density of the QD eigenstates in the time domain. We employ TRDR to investigate the carrier capture and carrier recombination dynamics. We will show that the carriers are captured in the InAs/InP QDs within 2.5 ps independent of the QD transition energy.

The self-assembled InAs QDs are grown by chemical beam epitaxy on an InP (100) substrate, misorientated by 2° towards (110). At a temperature of 500 °C, a 200 nm InP buffer layer is grown on the substrate, followed by a 100 nm lattice-matched GaInAsP layer ($\lambda_g = 1.29$ μm). A thin GaAs layer, i.e., 1.9 ML, is deposited before the growth of the InAs QD layer, 3.2 ML InAs, which is capped by 100 nm GaInAsP. The sample has a QD density of approximately $3.1 \cdot 10^{10}$ cm^{-2} as is measured by atomic force microscopy (AFM) of uncovered surface QDs, as shown in Fig. 1(a). For more details concerning the growth procedure, we refer to Ref. 5.

The time-resolved bleaching measurements are performed using two-color pump-probe differential reflection spectroscopy.[7,8] In this configuration, a 76.66 MHz mode-locked Ti:sapphire laser is used as the pump source, and is mechanically chopped with a frequency of 4 kHz. The pump pulses are focused on the sample with a spot size of 55 μm, exciting carriers in the barrier layers. The pump-induced absorption changes within the QDs are probed by 2-ps-long pulses which are generated from an optical parametric oscillator. The probe energy is tuned over the QD optical transition energy within the ensemble. With a graded index lens the probe light is focused on the sample with a spot size of 25 μm. Probe light reflected from the sample is collected by the same lens and is focused on a balanced photodetector and subsequently measured by a lock-in amplifier. Hereby, we are able to achieve differential reflectivity signals, $\Delta R/R_0 = (R - R_0)/R_0$, with a relative sensitivity of 5×10^{-7}. All the measurements are performed at RT.

The photoluminescence (PL) spectrum as well as the differential reflectivity spectrum are presented in Fig. 1(b). The inhomogeneously broadened PL spectrum clearly shows the luminescence of the QDs and of the quaternary layer centered at 0.816 eV, i.e., 1520 nm and 0.960 eV, i.e., 1290 nm, respectively. The spectrum obtained by TRDR is slightly red-shifted with respect to the QD PL and is centered at 0.812 eV, i.e., 1527 nm. We observe that the spectra do not coincide; moreover, the PL spectrum is broadened with respect to the TRDR spectrum, with full width at half maximum (FWHM) of 93 and 41 meV, respectively. However, if we analyze the PL spectrum obtained at 4.8 K, as is depicted in the inset of Fig. 1(b), the spectral shape of the 4.8 K PL spectrum and the RT-TRDR spectrum is similar. The shift as well as the broadening of the RT-PL spectrum is explained

[a]Author to whom correspondence should be addressed; electronic mail: e.w.bogaart@tue.nl

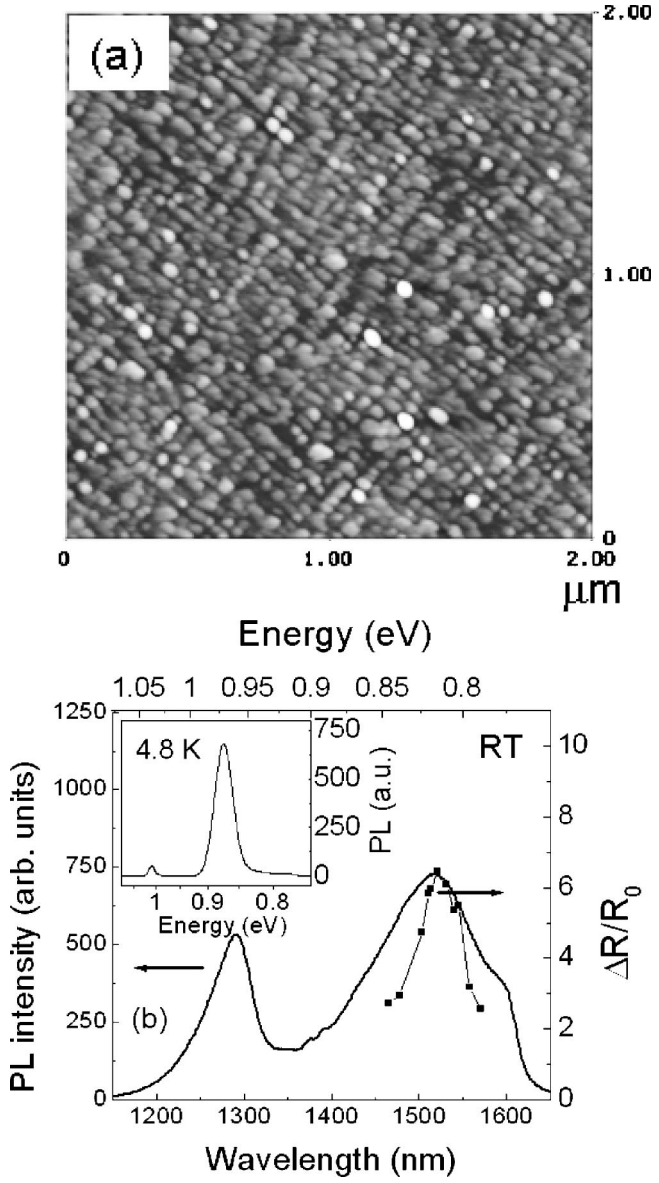

FIG. 1. (a) AFM image of the InAs/InP QD sample. The scan field is 2 $\times 2\ \mu m^2$ and the black-to-white height contrast is 5 nm. (b) Room temperature PL spectrum and the TRDR spectrum of the InAs/InP QDs. The inset depicts the PL spectrum at 4.8 K.

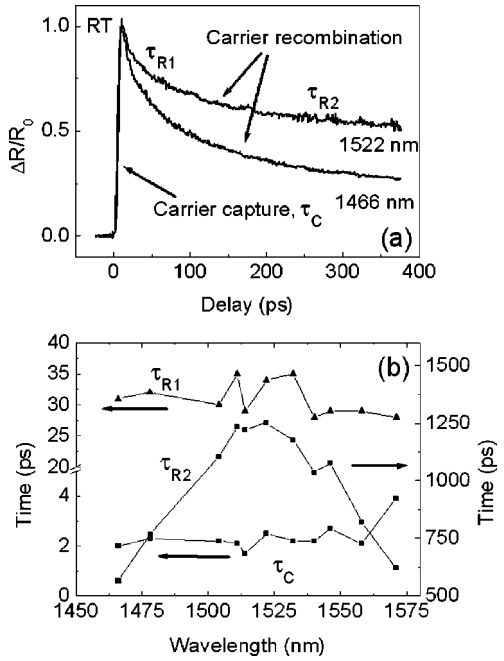

FIG. 2. Transient bleaching signals for different QD transition energies (a), showing the carrier capture and carrier recombination. The amplitudes of the bleaching signals are normalized for clarity. (b) Carrier dynamics vs the QD transition energy.

by thermal carrier activation and redistribution as discussed by Patanè *et al.*[9]

Figure 2(a) depicts the transient bleaching signal from the QD ground state optical transition measured at 1466 and 1522 nm. We observe a steep rise of the bleaching signal followed by a bi-exponential decay. The rise of the signal is characterized by the carrier captured time, τ_C, which includes carrier diffusion towards the QDs, carrier capture, and relaxation in the QDs, whereas the bleaching decay is due to carrier recombination within the QDs, i.e., the carrier lifetime. The bi-exponential decay is due to two different carrier recombination paths, namely nonradiative and radiative carrier recombination, characterized by τ_{R1} and τ_{R2}, as we will show later.

As is illustrated by the curves in Fig. 2(a), the carrier lifetime strongly depends on the QD optical transition energy. To investigate this dependence, we have measured the bleaching dynamics as function of the transition energy. Figure 2(b) depicts the carrier capture time and both carrier recombination times as function of transition energy. We clearly observe that the carrier capture time does not depend on the QD transition energy and has a value of $\tau_C = 2.2 \pm 0.3$ ps. The fast component of the carrier recombination process has a response time of $\tau_{R1} = 31 \pm 3$ ps, which exhibits a small variation around the peak transition energy of the QD ensemble. Contrary to the carrier capture and the fast carrier recombination time, the slower component of the carrier recombination process does have a strong energy dependence and its response time τ_{R2} can be well approximated by a Gaussian function centered at 0.816 eV, i.e., 1519 nm, with a FWHM of 55 meV. A similar behavior has been observed in InAs/GaAs QDs at RT as well as at 5 K, where nonradiative recombination is suppressed.[10] Moreover, the peak energy of this *decay time spectrum* is in accordance with the central energy of the PL spectrum, i.e., 0.816 eV. Hence, we believe that the slow decay time is due to the radiative carrier recombination, and the fast component of the bleaching decay is governed by thermally activated nonradiative recombination. The smaller radiative lifetimes for QDs with higher energies is due to enhanced carrier redistribution, whereas the lifetime for the QDs with longer emission wavelength is reduced due to their larger confinement energy.

To analyze the influence of the carrier density on the carrier capture time as well as on the carrier lifetime within the QDs, we have studied the bleaching dynamics as function of the pump excitation density. The probe beam has a fixed wavelength of 1522 nm, near to the peak emission wavelength. The magnitude of the QD bleaching is depicted in Fig. 3(a) and increases linearly with the excitation density. Note, no saturation of the QD bleaching is observed. The carrier capture time initially increases from 1.6 ps at low density to 2.5 ps with the increase of the excitation density and remains constant with further increase of the excitation power, as is shown in Fig. 3(b). Carrier capture processes are

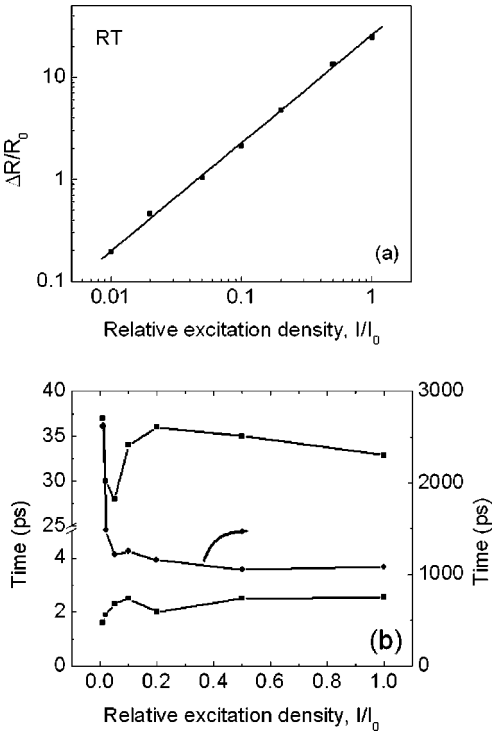

FIG. 3. (a) The QD bleaching magnitude as function of the excitation density, with $I_0 = 10.6 \cdot 10^{13}$ photons/cm^2 per pulse. (b) Carrier capture time and the carrier recombination times vs the pump excitation density. The probe wavelength is set to 1522 nm.

expected to be enhanced due to carrier-carrier scattering at high carrier densities,[11] however, despite the increase of the carrier density, no enhancement of the capture rate is observed. Thus, from both the nonsaturated bleaching and the excitation density independent capture rate, we suggest that during our measurements the average number of electron-hole pairs per QD is less than one, even at high excitation density. In addition, Fig. 3(b) shows that the carrier capture is initially fast, which we assign to the strong structural improvement of the two-dimensional wetting layer upon insertion of the GaAs interlayer and the high QD density, as shown in Ref. 5.

For the nonradiative recombination time a small variation at low excitation density is observed, with a minimum and maximum value of 28 and 37 ps, respectively. On the other hand, the radiative recombination time shows a relatively strong excitation density dependence with a lower limit of 1070 ps. Nonetheless, the long carrier lifetime demonstrates high structural quality of the self-assembled QDs. In addition, we would like to remark that similar results are obtained at lower and higher QD transition energies.

In summary, we have studied the carrier dynamics in self-assembled InAs/InP QDs, using TRDR as function of the QD transition energy as well as the pump-induced carrier density, both at room temperature. The QD structure has a fast optical response, i.e., short carrier capture time, less than 2.5 ps and a carrier lifetime within the QDs of 560–2600 ps, depending on the transition energy and the carrier density. The fast carrier capture makes these QDs suitable for the realization of high-speed QD-laser structures, semiconductor optical amplifiers, and optical switches with large modulation bandwidths operating in the 1.55 μm wavelength range.

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