Carrier relaxation dynamics in InAs/InP quantum dots

P. Miska,[1,2,a)] J. Even,[2] O. Dehaese,[2] and X. Marie[3]
[1]*Laboratoire de Physique des Matériaux, LPM-UMR CNRS 7556, Faculté des Sciences, Université Henry Poincaré–Nancy I, Boulevard des Aiguillettes, BP 239, F-54506 Vandœuvre-lès-Nancy, France*
[2]*Laboratoire FOTON (INSA UMR CNRS 6082), 20 Avenue des Buttes de Coesmes, CS 14315, 35043 Rennes Cedex, France*
[3]*Université de Toulouse, INSA-UPS-CNRS LPCNO, 135 Avenue de Rangueil, F-31077 Toulouse, France*

(Received 6 February 2008; accepted 17 March 2008; published online 13 May 2008)

The electronic properties of InAs/InP(113)B double-cap quantum dots (QDs) emitting around 1.55 μm are investigated. The carrier dynamics in QDs is studied by nonresonant time-resolved photoluminescence experiments. This analysis reveals the electronic structure of the QDs and the transient filling of the confined levels. Under low excitation densities, the spontaneous exciton lifetime is estimated and compared to time-resolved experiments. Under high excitation density, a direct Auger recombination effect is identified. The temperature analysis enables us to distinguish Auger and phonon-assisted relaxation processes. © *2008 American Institute of Physics*.
[DOI: 10.1063/1.2909536]

Carrier relaxation and band filling effects have been widely discussed in recent years due to their key role in the operations of optoelectronic devices such as quantum dot (QD) lasers. Due to the quantum confinement, many electronic states are present inside the QDs. It has been shown that efficient carrier thermalization can occur by multiphonon[1,2] or Coulomb interaction process.[3] The photoluminescence (PL) signal of excited states has also been clearly evidenced under high excitation density.[4–6] Moreover, it has been shown that Auger relaxation is an extremely fast phenomenon[7] and the dominant effect under high excitation density.[8,9]

Despite all these analyses, information about the carrier dynamics in InAs/InP QDs are still lacking. An original growth method has been proposed to obtain InAs/InP(113)B QDs with a tunable emission energy.[10] These structures, named double-cap QDs (DC-QDs), have been both theoretically[11,12] and experimentally analyzed.[13–15] Moreover, laser emission at 1.55 μm has been obtained with this system.[16] In this paper, we present a systematic study of the optical and electronic properties of the DC-QDs by using time-resolved PL (tr-PL) spectroscopy.

We used the DC growth method well described in Ref. 10 to elaborate QDs. The maximum height of the QDs that we analyze here is 2 nm (Ref. 13) and the average QD diameter is about 35 nm. Samples are characterized by tr-PL spectroscopy at low temperature. The experiments are performed at 10 K by using a 790 nm mode-locked Ti:sapphire laser producing 1.2 ps long light pulses with a repetition rate of about 82 MHz. The tr-PL of the ensemble of QDs is then recorded by using either a streak camera or upconverting the luminescence signal in a nonlinear crystal with time resolutions of 20 and 1.2 ps, respectively.

The tr-PL spectra obtained by the upconversion setup for various times after excitation are shown in Fig. 1. The spectra are fitted with three peaks at energy positions corresponding to the excitonic states recorded in our previous studies:[13] the QDs excitonic ground state (QD_0) at 0.94 eV, the QDs first excited excitonic state (QD_1) at 0.99 eV, and the wetting layer (WL) excitonic ground state at around 1.05 eV. The QD and WL levels are visible at short times after excitation (10 ps). For longer times, the carrier population in high energy states diminishes either by light emission or by relaxation to the ground state QD_0. Slight shifts of the peaks can be seen due to the accuracy of the fit process. The shift of the WL emission peak is due to the large emission band of the WL and the stabilization of carriers in the WL high energy levels under high excitation density.

A systematic study was performed with the streak camera setup. Spectra obtained at 10 K with various optical excitation densities for a long time (1650 ps) after excitation are reported in Fig. 2(a). The slight changes between Figs. 1 and 2(a) can be explained by the differences between the two setups.[17] The spectra in Fig. 2(a) are very similar whatever the excitation density is. We note that the carrier relaxation is not completed at $t=1650$ ps as the WL emission peak is still visible. Under low excitation density, this could be explained by the presence of a potential barrier between the QDs and the WL. Under high excitation density, the WL luminescence signal is less intense. This can be explained by an efficient

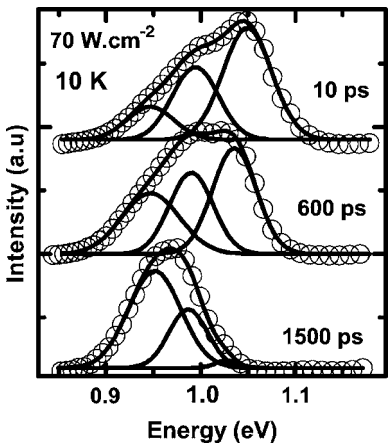

FIG. 1. PL spectra (open circles) recorded at 10 K with the upconversion setup at different times after excitation (10, 600, and 1500 ps) for an excitation density of $I_{exc}=70$ W cm^{-2}. Spectra are fitted with three Gaussian peaks. Results of the fit are reported with straight lines.

a)Electronic mail: patrice.miska@lpm.u-nancy.fr.

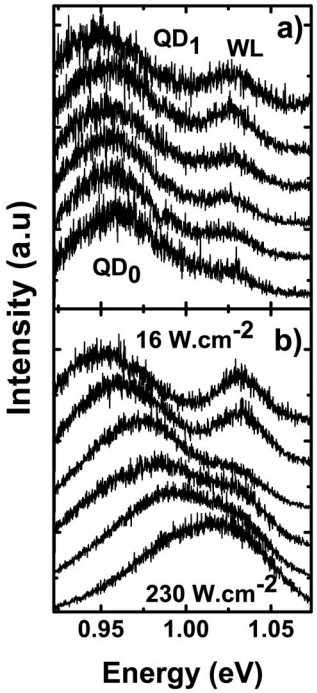

FIG. 2. PL spectra extracted from the streak camera pictures and recorded at 10 K for different excitation densities (I_{exc}=16, 45, 57, 74, 155, and 230 W cm^{-2}). Spectra at (a) long times (1650 ps) and (b) short times (150 ps) after excitation are reported. Spectra are vertically shifted for clarity.

carrier relaxation assisted by the Auger effect. Nevertheless, there may be a competition between this last phenomenon and a saturation of the QD ground state that could slow down the carrier relaxation within the QDs just after excitation. The spectra in Fig. 2(b) recorded at short times after excitation with a low incident optical power (150 ps, 16 W cm^{-2}) are equivalent to the one reported in Fig. 2(a). Indeed, in this case, the luminescence of the QD_0 and the WL states are observed. When the excitation density increases from 45 to 57W cm^{-2}, a luminescence signal appears at about 0.99 eV. It corresponds to the luminescence of the QD's first excited state (QD_1), also identified in Ref. 13. Then, when the excitation density is further increased, we observe a stabilization of the carriers in the WL and the QD_1 states and then a corresponding intense luminescence signal. This behavior reveals the progressive filling of the QD_1 and QD_0 levels. This phenomenon could explain the slight shift to high energies of the QD_0 peak that can be observed in Fig. 2(a) at long times after excitation and for high excitation density. An estimation of the average number of excitons per QD produced by the optical excitation has been calculated with the formula

$$N_0 = \frac{P_{\text{exc}}}{fh\nu} T_{\text{trans}}(1 - e^{-\alpha L}), \tag{1}$$

where P_{exc} is the average excitation density, $h\nu$ is the energy of incident photons, f is the laser repetition rate, α is the absorption coefficient of the InP barrier ($\sim 10^4$ cm^{-1}), L is the InP barrier thickness (30 nm), and T_{trans} is the air/InP interface transmission coefficient. The filling of the QD_1 level observed for an optical density of 45 W cm^{-2} corresponds here to an average exciton number per dot of 3. It shows that the filling of the QD_1 level is mainly associated to

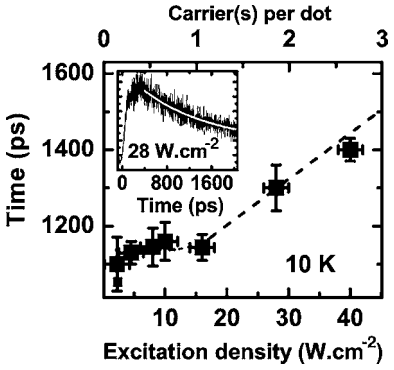

FIG. 3. QD_0 decay time reported as a function of the optical excitation density and the corresponding calculated average number of exciton(s) per QD. The inset shows a typical kinetics from which the decay times are measured (T=10 K).

a saturation of the QD_0 level after formation of the biexciton ground state.

We have also performed an analysis of the QD_0 radiative lifetime. A direct experimental method is to use resonant excitation or pump-probe experiments.[18,19] In the present work, we use nonresonant excitation measurements. The measured QD_0 decay times are reported in Fig. 3 as a function of the excitation density and of the calculated number of excitons per QD. Under low excitation density the decay time is constant, of about $\tau \sim 1150$ ps. Under this regime, the QD_0 level is not saturated (the average population per dot is smaller than 1). We assume then that the exciton recombines with a characteristic time close to the radiative lifetime. This value has been compared to the theory. In the strong confinement regime, the spontaneous emission rate is given by Einstein's coefficient

$$A = \frac{1}{\tau_{\text{spon}}^{\text{QD}}}.$$

This coefficient can be written in considering only a transition between the electron and the heavy hole:[18,20]

$$\frac{1}{\tau_{\text{spon}}^{\text{QD}}} = \frac{n_{\text{op}} e^2 \omega_0 E_p}{3 m_0 h c^3 \varepsilon_0} |\langle F_C | F_V \rangle|^2.$$

The envelope wave function overlapping coefficient $|\langle F_C | F_V \rangle|^2$ is calculated using a model described elsewhere.[14] We find $|\langle F_C | F_V \rangle|^2 = 0.92$ for the fundamental transition. The use of Kane's energy E_p=21.1 eV and the InAs optical index n_{op}=3.9 leads to the 1se-1sh radiative lifetime values of 1200 ps for a QD with a height of 2 nm and a diameter of 35 nm. This calculated value is in very good agreement with the measured one.

A detailed study was conducted by measuring the tr-PL rise times $\tau_r^{QD_0}$ and $\tau_r^{QD_1}$ of QD_0 and QD_1 levels, respectively, as a function of both the excitation density and temperature [Fig. 4(a)]. Under high excitation density, the measured rise times of both levels are independent of temperature. It is probably limited by the time resolution of the streak camera setup. The Auger relaxation is the dominant process in this regime, which yields a very fast carrier relaxation. On the contrary, under weak excitation density, the rise times decrease with temperature. This reveals a strong influence of the phonon relaxation, which is the dominating process in this regime. We have applied here the three level model of Ohnesorge *et al.*[8] by considering our three

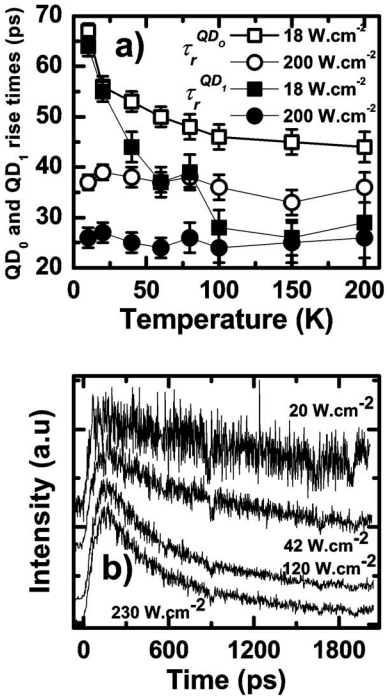

FIG. 4. (a) Evolution of the rise times of the fundamental ($\tau_r^{QD_0}$) (white symbols) and of the first exited ($\tau_r^{QD_1}$) (black symbols) states as a function of the temperature for two excitation regimes: strong (200 W cm^{-2}) (circles) and weak (18 W cm^{-2}) (squares) excitation density. Time evolution spectra recorded at 10 K at the energy of the first excited state (QD$_1$) level for different excitation densities are reported in (b). The kinetics are vertically shifted for clarity.

levels: WL, QD$_0$, and QD$_1$. Then, under weak excitation density, the rise time of the fundamental level can be written as

$$\tau_r^{QD_0} = \tau_r^{QD_1} + \tau_{QD_1 \to QD_0}. \quad (2)$$

This equation corresponds to a cascade relaxation of excitons assisted by phonons emission. According to Ohnesorge *et al.*, under strong excitation regime, the luminescence decay time of the WL ($\tau_{WL \to QD_1} = \tau_r^{QD_1}$) is close to the rise time of the fundamental level $\tau_r^{QD_0}$. Thus, they showed the existence of a direct Auger effect that is responsible for the direct capture from the WL into the dot. The relation $\tau_r^{QD_0} \to \tau_r^{QD_1}$ implies then, according to Eq. (2), $\tau_{QD_1 \to QD_0} \to 0$ under strong excitation regime. Moreover, the decay time of the first excited level QD$_1$ can be written as

$$\frac{1}{\tau_d^{QD_1}} = \frac{1}{\tau_{QD_1 \to QD_0}} + \frac{1}{\tau_{spon}^{QD_1}}.$$

When the condition $\tau_{QD_1 \to QD_0} \to 0$ is realized, one can induce that it yields a decrease of the QD$_1$ luminescence decay time. The QD$_1$ decay times have been measured and are displayed in Fig. 4(b) for different excitation powers. The kinetics clearly show a decrease in QD$_1$ decay time. This behavior evidences then in our three level system the presence of a direct Auger relaxation effect, where carriers directly relax from the WL into the QD$_0$ level.

In conclusion, we analyzed the carrier relaxation processes in InAs/InP(113)B DC-QDs. We evidenced the progressive filling of the identified confined levels QD$_0$ and QD$_1$ with the number of photogenerated carriers. The QD$_1$ level starts filling with an average number of carriers per dot of about 3. We have also measured the radiative lifetime of the QDs fundamental level. Experimental values are in good agreement with the theoretical values. In varying both temperature and optical excitation density, we managed to dissociate the Auger relaxation process from phonon relaxation process in our three level system. The analysis of the QD$_1$ PL decay time with the excitation density has confirmed the presence of a direct Auger effect, responsible for the direct capture of carriers from the WL into the QDs fundamental level.

[1] R. Heitz, M. Grundmann, N. N. Ledentsov, L. Eckey, M. Veit, D. Bimberg, V. M. Ustinov, A. Y. Egorov, A. E. Zhukov, P. S. Kop'ev, and Z. I. Alferov, Appl. Phys. Lett. **68**, 361 (1996).
[2] S. Raymond, S. Fafard, S. Charbonneau, R. Leon, D. Leonard, P. M. Petroff, and J. L. Merz, Phys. Rev. B **52**, 17238 (1995).
[3] Al. L. Efros, V. A. Kharchenko, and M. Rosen, Solid State Commun. **93**, 281 (1995).
[4] M. Grundmann, N. N. Ledentsov, O. Stier, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Z. I. Alferov, Appl. Phys. Lett. **68**, 979 (1996).
[5] H. Lipsanen, M. Sopanen, and J. Ahopelto, Phys. Rev. B **51**, 13868 (1995).
[6] S. Raymond, S. Fafard, P. J. Poole, A. Wojs, P. Hawrylak, S. Charbonneau, D. Leonard, R. Leon, P. M. Petroff, and J. L. Merz, Phys. Rev. B **54**, 11548 (1996).
[7] R. Ferreira and G. Bastard, Appl. Phys. Lett. **74**, 2818 (1999).
[8] B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11532 (1996).
[9] D. Morris, N. Perret, and S. Fafard, Appl. Phys. Lett. **75**, 3593 (1999).
[10] C. Paranthoen, C. Platz, G. Moreau, N. Bertru, O. Dehaese, A. Le Corre, P. Miska, J. Even, H. Folliot, C. Labbé, G. Patriarche, J. C. Simon, and S. Loualiche, J. Cryst. Growth **251**, 230 (2003).
[11] J. Even, S. Loualiche, P. Miska, and C. Platz, J. Phys.: Condens. Matter **15**, 8737 (2003).
[12] C. Cornet, C. Platz, J. Even, P. Miska, C. Labbé, H. Folliot, and S. Loualiche, AIP Conf. Proc. **772**, 787 (2005).
[13] P. Miska, C. Paranthoen, J. Even, O. Dehaese, H. Folliot, N. Bertru, S. Loualiche, M. Sénès, and X. Marie, Semicond. Sci. Technol. **17**, L63 (2002).
[14] P. Miska, J. Even, C. Paranthoen, O. Dehaese, A. Jbeli, M. Senès, and X. Marie, Appl. Phys. Lett. **86**, 111905 (2005).
[15] P. Miska, C. Paranthoen, J. Even, N. Bertru, A. Le Corre, and O. Dehaese, J. Phys.: Condens. Matter **14**, 12301 (2002).
[16] K. Veselinov, F. Grillot, P. Miska, E. Homeyer, P. Caroff, C. Platz, J. Even, X. Marie, O. Dehaese, S. Loualiche, and A. Ramdane, Opt. Quantum Electron. **38**, 369 (2006).
[17] In order to increase the detection sensitivity of the upconversion setup, very large slits were used yielding a rather poor spectral resolution (~10 meV). For the streak camera setup, the spectral resolution is of about 2 meV.
[18] M. Paillard, X. Marie, E. Vanelle, T. Amand, V. K. Kalevich, A. R. Kovsh, A. E. Zhukov, and V. M. Ustinov, Appl. Phys. Lett. **76**, 76 (2000).
[19] C. Cornet, C. Labbé, H. Folliot, P. Caroff, C. Levallois, O. Dehaese, J. Even, A. Le Corre, and S. Loualiche, Appl. Phys. Lett. **88**, 171502 (2006).
[20] There is a mistake in the calculation of $1/\tau_{rad}$ published in Ref. 18. An erratum will be published soon. We use here the right formula.