# Electronic structure and carrier dynamics in InAs/InP double-cap quantum dots

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The carrier dynamics in InAs double-cap quantum dots (DC-QDs) grown on InP(113)B are investigated. The shape of these QDs can be controlled during the growth, yielding an emission wavelength of the system of about 1.55 $\mu$m at room temperature. The DC-QD dynamics is studied by time-resolved photoluminescence experiments at low temperature for various excitation densities. A simplified dynamic model is developed, yielding results consistent with experimental data. This analysis yields the determination of the Auger coefficients and the intradot relaxation time in this system. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3078290]

Reduced dimensions of semiconductor quantum dots (QDs) offer a three-dimensional carrier confinement, yielding discrete atomlike energy spectra. This property is promising for more efficient optoelectronic devices.[1,2] Properties of III–V systems such as InAs/GaAs QDs are now well known.[2,3] Many studies have also been devoted to growth and analyses of optical properties of InAs/InP QDs[4,5] or quantum dashes.[6,7] But information on the carrier dynamics and energy relaxation processes in such InAs/InP QDs are still lacking. Recently, the growth of self-organized InAs QDs on misoriented InP(113)B substrates has been proposed to get QDs with both quantum sizes and a high surface density.[8] The control of the maximum QD height of the sample yields the control of their wavelength emission.[9] These structures are named double-cap quantum dots (DC-QDs) and emit at 1.55 $\mu$m at room temperature.[10] The optical properties of a single DC-QD layer have been analyzed[10–12] and lasing structures were obtained with such nanostructures with low threshold current densities[13,14] In a previous study, we managed to dissociate the two capture (or relaxation) mechanisms described in literature: phonon and Auger assisted relaxations.[15]

In this paper, we report on the analysis of the dynamic properties of InAs DC-QD layers grown on InP(113)B substrate. The samples are obtained by the spontaneous Stranski–Krastanow growth mode after the deposition of 2.1 InAs monolayers (ML) at 480 °C on InP(311)B substrate. The DC growth method[10] is then used to control the QD maximum height. The maximum height of the DC-QDs here is of 2 nm,[10] the average DC-QD diameter of about 35 nm, and the surface density of about $10^{10}$ cm$^{-2}$.[16]

Samples are characterized by tr-PL spectroscopy at low temperature. The experiments were performed at 10 K using a 790 nm Ti-sapphire laser producing 1.2 ps long light pulses with a repetition rate of about 82 MHz. The tr-PL is then recorded using a synchro-scan streak camera with an overall time resolution of ~8 ps.[17]

A simple model described elsewhere[10,11] and including the DC-QD shape, strain, and surface orientation effects is used for the description of the confined electronic states. This model gives the probability densities $|\Psi(r,z)|^2$, the energies, and the oscillator strengths obtained for a polarization $\vec{\varepsilon}$ in the electric dipole approximation by

$$f_{i\to f}^{\vec{\varepsilon}} = \frac{2m_0(E_f - E_i)}{e^2\hbar^2}|\vec{\varepsilon}\,\vec{\mu}_{i\to f}^{\vec{\varepsilon}}|,$$

with $E_f$ and $E_i$ as the energies of the final and initial states, respectively, and $\vec{\mu}_{i\to f}^{\vec{\varepsilon}} = \langle f|-e\vec{r}|i\rangle$ as the dipole of the transition. Experimental points extracted from tr-PL analysis under high excitation density (see Ref. 10) are reported in Fig. 1(a). The experimental points correspond to the energies of the fundamental ($QD_0$) and first excited ($QD_1$) states of the DC-QDs, and of two states of the wetting layer (WL) visible in the tr-PL spectra. The error bars of the confined energy states correspond to the full width at half maximum of the Gaussian fit of the tr-PL spectra. Calculated dipole lengths are reported in Fig. 1(b) as a function of the energy for a truncated lens shaped QD. The model only gives the energies and

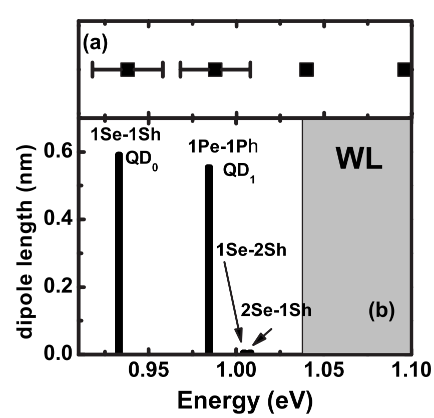


FIG. 1. (a) Experimental and (b) calculated energies and dipole lengths of the two confined states $QD_0$ and $QD_1$ of the DC-QD. The error bars of the experimental data correspond to the full width at half maximum of the Gaussian fit of the tr-PL spectra. Experimental data taken from Ref. 10.

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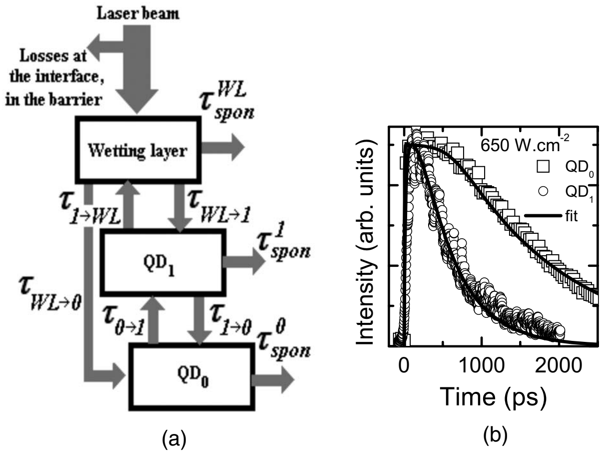


FIG. 2. (a) Sketch of the radiative and nonradiative mechanisms used to described the exciton dynamics and (b) experimental tr-PL curves under high excitation density for the first excited state ($QD_1$, circles) and the fundamental state ($QD_0$, squares). The fit extracted from the rate equation model is shown with continuous line.

oscillator strengths of the confined states. The WL propagative states can not be determined (they are represented with a grey area in the spectrum of Fig. 1(b). A reasonable agreement between theoretical and experimental results is obtained despite the simple model used. It confirms the existence of confined levels in our system.

A complete analysis of the carrier dynamics has been done by combining tr-PL experiments and a dynamic model. The different relaxation paths and the corresponding relaxation times we considered are depicted in Fig. 2(a). Based on the experimental results described above, we choose a three-level model (Fig. 1) to model the DC-QD: the WL level where the carriers are created and two QD confined exciton states $QD_0$ and $QD_1$. We considered the respective spontaneous emission lifetimes of the three levels $\tau_{\rm spon}^{\rm WL}$, $\tau_{\rm spon}^{1}$, and $\tau_{\rm spon}^{0}$. The values of $\tau_{\rm spon}^{\rm WL}$ and $\tau_{\rm spon}^{1}$ taken from Refs. 15 and 18 are 500 and 900 ps, respectively (the fit is not really sensitive to the $\tau_{\rm spon}^{\rm WL}$ value). The value of $\tau_{\rm spon}^{0}$ taken from Ref. 16 is of 1150 ps. For The WL and the first excited state level $QD_1$, we also took into account the capture $\tau_{\rm WL\to 1}$ and intradot relaxation $\tau_{1\to 0}$ times and the reverse processes $\tau_{1\to \rm WL}$ and $\tau_{0\to 1}$ due to the thermal equilibrium of the system. $\tau_{1\to \rm WL}$ and $\tau_{0\to 1}$ are then calculated as in Ref. 19 by applying the detailed balance principle

$$\tau_{0\to 1} = \tau_{1\to 0}\frac{\rho_0}{\rho_1}e^{\left(\frac{E_{\rm QD_1}-E_{\rm QD_0}}{k_BT}\right)}$$

$$\tau_{1\to \rm WL} = \tau_{\rm WL\to 1}\frac{2\rho_1 N_{\rm QD}V_{\rm QD}\pi\hbar^2}{WLm_e^*k_BT}e^{\left(\frac{E_{\rm WL}-E_{\rm QD_1}}{k_BT}\right)},$$

with $\rho_0$, $\rho_1$ and $E_0$, $E_1$ the degeneracy and energies of the confined levels $QD_0$ and $QD_1$, respectively ($\rho_0=1$, $\rho_1=2$). $N_{\rm QD}$ is the QD volume density, $V_{\rm QD}$ is the total QD volume inside a square surface with lateral dimension $W$, and $L$ is the WL thickness. The considered surface corresponds to the laser spot. Capture $\tau_{\rm WL\to 1}$ and intradot relaxation $\tau_{1\to 0}$ times are defined using Auger ($C_{\rm WL\to 1}$ and $C_{1\to 0}$) and phonon ($A_{\rm WL\to 1}$ and $A_{1\to 0}$) coefficients as proposed in Ref. 19,

$$\tau_{\rm WL\to 1} = \frac{1}{A_{\rm WL\to 1}+C_{\rm WL\to 1}N_{\rm WL}}$$

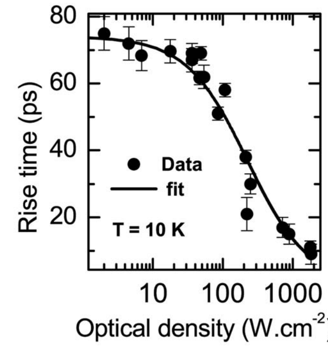


FIG. 3. Experimental rise time $\tau_{\rm WL\to 0}$ ($=\tau_{\rm WL\to 1}$ in our model) of the fundamental state $QD_0$ as a function of the excitation intensity (full circles). The fit extracted from the rate equation model is shown with a continuous line.

$$\tau_{1\to 0} = \frac{1}{A_{1\to 0}+C_{1\to 0}N_{\rm WL}}.$$

We assume here a geminate capture process of the electron-hole pairs. Moreover, we introduced the occupation rates $f_0(t)$, $f_1(t)$, and $f_{\rm WL}$ of the $QD_0$, $QD_1$, and WL levels, respectively. They are defined by

$$f_1(t) = 1 - P_1(t), \qquad f_0(t) = 1 - P_0(t),$$
$$P_1(t) = \frac{N_1(t)V_{\rm 1QD}}{4}, \quad P_0(t) = \frac{N_0(t)V_{\rm 1QD}}{2},$$
$$f_{\rm WL} = 1,$$

with $V_{\rm 1QD}$ as the volume of one QD and $N_0(t)$ and $N_1(t)$ as the populations of the $QD_0$ and $QD_1$ levels, respectively. We consider the WL as an exciton reservoir. Solving this first set of rate equations with those parameters did not give a good fit of the tr-PL curves. We have evidenced in former study[15] the existence of a direct capture process from the WL onto the $QD_0$ level for all the excitation optical density ranges. We introduced then in the model a direct relaxation path between the WL and the $QD_0$ level characterized by a capture time $\tau_{\rm WL\to 0}=\tau_{\rm WL\to 1}$. This model yields the resolution of the following rate equations:

$$\frac{dN_{\rm WL}}{dt} = \frac{N_{\rm WL}}{\tau_0} + \frac{N_1}{\tau_{1\to \rm WL}}\frac{V_{\rm QD}}{V_{\rm WL}}f_{\rm CM} - \frac{N_{\rm WL}}{\tau_{\rm WL\to 1}}f_1 - \frac{N_{\rm WL}}{\tau_{\rm spon}^{\rm WL}} - \frac{N_{\rm WL}}{\tau_{\rm WL\to 0}}\frac{V_{\rm QD}}{V_{\rm WL}}f_0 = 0,$$

$$\frac{dN_1}{dt} = \frac{V_{\rm WL}}{V_{\rm QD}}\frac{N_{\rm WL}}{\tau_{\rm WL\to 1}}f_1 + \frac{N_0}{\tau_{0\to 1}}f_1 - \frac{N_1}{\tau_{1\to \rm WL}}f_{\rm WL} - \frac{N_1}{\tau_{\rm spon}^{1}} - \frac{N_1}{\tau_{1\to 0}}f_0 = 0,$$

$$\frac{dN_0}{dt} = \frac{N_1}{\tau_{1\to 0}}f_0 - \frac{N_0}{\tau_{0\to 1}}f_1 - \frac{N_0}{\tau_{\rm spon}^{0}} + \frac{N_{\rm WL}}{\tau_{\rm WL\to 0}}\frac{V_{\rm QD}}{V_{\rm WL}}f_0 = 0.$$

The number of photogenerated carriers in the WL is given by

$$N_{\rm WL}(t=0) = \frac{P_{\rm exc}}{fh\nu}T^{\rm trans}(1-e^{-\alpha L_z}),$$

with $P_{\rm exc}$ as the average laser excitation power of the tr-PL setup, $h\nu$ as the energy of the incident photons, $f$ as the repetition rate of the laser source, $\alpha$ ($\sim 10^4$ cm$^{-1}$) as the

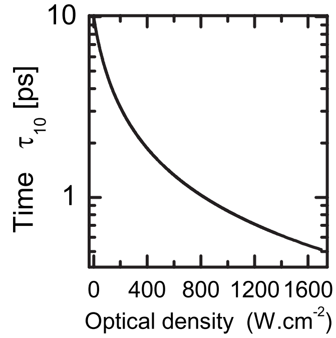


FIG. 4. Intradot relaxation $\tau_{1\rightarrow 0}$ between $QD_0$ and $QD_1$ confined states extracted from the dynamic model and reported as a function of the excitation intensity.

absorption coefficient in the InP barriers, $L_z$ (30 nm) as the thickness of the InP barrier crossed by the photons before reaching the WL, and $T^{trans}=4n_{opt}^{InP}/(1+n_{opt}^{InP})^2$ as the air/InP transmission coefficient evaluated with $n_{opt}^{InP}$ (3.9) the optical index of the InP. $\tau_0$ is the duration of the laser pulse. With this model the fitting parameters are $A_{WL\rightarrow 1}$, $C_{WL\rightarrow 1}$, $A_{1\rightarrow 0}$, and $A_{1\rightarrow 0}$.

The tr-PL curves obtained from these equations give a good fit of both the experimental rise and decay times of the $QD_0$ and $QD_1$ levels in a large excitation intensity range [Fig. 2(b)]. The rise and decay curves at low excitation density have exponential shape. Under high optical excitation, the $QD_1$ rise and decay curves also have exponential shape, but $QD_0$ decay curve shows a plateau. This plateau is due to the saturation of the PL signal corresponding to a complete filling $QD_0$ confined state. Our model gives a good description of this phenomenon.

Important information for the realization of high frequency lasers is the carrier capture time into the QDs. An evaluation of this parameter can be obtained here by evaluating the tr-PL rise time. We have reported in Fig. 3 the experimental and theoretical values of the capture time $\tau_{WL\rightarrow 0}$ ($=\tau_{WL\rightarrow 1}$ in our model) as a function of the excitation intensity. We find a good correspondence between the experimental and calculated values. The exciton rise time decreases from 70 to 8 ps and we can identify two regimes: under $\sim$50 W cm$^{-2}$, a phonon-assisted regime (where the capture time does not depend on the carrier density), and above 50 W cm$^{-2}$, an Auger regime. Then, the values of phonon ($A_{WL\rightarrow 1}$ and $A_{1\rightarrow 0}$) and Auger ($C_{WL\rightarrow 1}$ and $C_{1\rightarrow 0}$) coefficients of our system were extracted from the fit. We found $A_{WL\rightarrow 1}=1.35\times 10^{10}$ s$^{-1}$, $A_{1\rightarrow 0}=1\times 10^{10}$ s$^{-1}$, $C_{WL\rightarrow 1}=5\times 10^{-15}$ m$^3$ s$^{-1}$, and $C_{1\rightarrow 0}=9\times 10^{-14}$ m$^3$ s$^{-1}$. These values are close to the ones found in the literature for similar systems.[19–21]

To complete the study, we extracted from the model the intradot relaxation time $\tau_{1\rightarrow 0}$ between the $QD_0$ and $QD_1$ confined states. This time could be measured directly with resonant excitation tr-PL experiment. This calculated intradot relaxation time is reported in Fig. 4 as a function of the excitation intensity. The values obtained under high excitation are compatible with the data found in the literature.[22,23]

In conclusion, we have analyzed the carrier dynamics in DC InAs/InP QDs emitting around 1.55 $\mu$m. We identified two confined energy levels and gave their energies and oscillator strengths. A dynamic study, coupling tr-PL experiments with a simple rate equation model, has revealed the energy relaxation and recombination processes of the exciton. The values of the phonon and Auger coefficients have been deduced and the intradot relaxation time between the two confined levels has been estimated.


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