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

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ABSTRACT

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 μm at room temperature. The DC-QD dynamics is studied by time-resolved photoluminescence (tr-PL) 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 intra-dot relaxation time in this system.

Self-organized semiconductor quantum dots (QDs) have been intensively studied in the last years. Their reduced dimensions offer a three-dimensional carrier confinement, yielding discrete atom-like energy spectra. This property is promising for more efficient optoelectronic devices [1,2]. The formation process and the optical properties of many III-V systems such as InAs/GaAs QDs are now well known [2,3]. Many studies have also been devoted to growth and analyzes of optical properties of InAs/InP QDs [4,5] or quantum dashes [6,7]. But informations 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]. In the optimized structures, 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 μm at room temperature [10]. The optical properties of a single DC-QDs layer have been analyzed [10,11,12], 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]. Nevertheless, a complete dynamic analysis, necessary to give us useful information for the description of the properties of a InAs/InP laser emitting at 1,55 μm , was still lacking.

In this paper, we report on the analysis of the dynamic properties of InAs DC-QD layers grown on InP(113)B substrate. We used optical experiments together with a dynamic model. The QDs samples are investigated by means of continuous and time-resolved photoluminescence (tr-PL) spectroscopy. We use a static model to describe the energy levels inside the DC-QDs, and a dynamic model based on the resolution of rate equations to fit the tr-PL spectra. The DC-QDs 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 with a gas source molecular beam epitaxy system. The double-cap (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]. Moreover, the WL emission is observed in cw-PL spectra at about 1.05 eV at 10 K [16].

The cw-PL analysis is performed by exciting the samples at 25 K with a 647 nm Kr laser, and the detection is done by a Peltier cooled PbS detector. Samples are also characterized by time-resolved photoluminescence spectroscopy (tr-PL) 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 of the DC-QDs. This model gives the probability densities $|\Psi(r, z)|^2$, the energies and the oscillator strengths, obtained for a polarisation $\vec{\epsilon}$ in the electric dipole approximation by :

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

with E_f and E_i the energies of the final and initial states respectively, and $\vec{\mu}_{i \rightarrow f}^{\vec{\epsilon}} = \langle f | -e\vec{r} | i \rangle$ the dipole of the transition. Calculated dipole lengths are reported in Fig. 1 as a function of the energy for a truncated lens shaped QD. The model only gives the energies and 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 Figure 1a). The theoretical spectrum is compared with experimental points extracted from tr-PL analysis under high excitation

density (see ref. 10). The experimental points correspond to the energies of the fundamental (QD₀) and first excited (QD₁) states of the DC-QDs, and of two states of the 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. A reasonable agreement between theoretical and experimental results is obtained despite the simple model used. Former theoretical and experimental studies revealed the electronic structure of InAs/InP(113)B DC-QDs [10,11,12,16]. For DC-QDs with a maximum height of 2 nm the transition energy between the first excited states (QD₁ transition between 1Pe and 1Ph states) has been identified around 50 meV above the energy of the fundamental transition (QD₀ transition between 1Se and 1Sh states) [10]. This result has been confirmed by a laser emission obtained with such DC-QDs both at the energy of the fundamental transition and at the energy of the transition between the first DC-QD excited states [13].

After the identification of the confined electronic states, 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 QD₀ and QD₁. We considered the respective spontaneous emission lifetimes of the three levels τ_{Spon}^{WL} , τ_{Spon}^1 and τ_{Spon}^0 . The values of τ_{Spon}^{WL} and τ_{Spon}^1 , taken from ref 15 and 18 are of 500 and 900 ps respectively (the fit is not really sensible to the τ_{Spon}^{WL} value) . The value of τ_{Spon}^0 , taken from ref. 16 is of 1150 ps. For The WL and the first excited state level QD₁, we also took into account the capture $\tau_{WL \rightarrow 1}$ and intra-dot relaxation $\tau_{1 \rightarrow 0}$ times and the reverse processes $\tau_{1 \rightarrow WL}$ and $\tau_{0 \rightarrow 1}$ due to the thermal equilibrium of the system. $\tau_{1 \rightarrow WL}$ and $\tau_{0 \rightarrow 1}$ are then calculated as in ref. 19 by applying the detailed balance principle

$$\tau_{0 \rightarrow 1} = \tau_{1 \rightarrow 0} \frac{\rho_0}{\rho_1} e^{\left(\frac{E_1 - E_0}{k_B T}\right)} \quad \tau_{1 \rightarrow WL} = \tau_{WL \rightarrow 1} \frac{2\rho_1 N_{QD} V_{QD} \pi \hbar^2}{W L m_e^* k_B T} e^{\left(\frac{E_{WL} - E_1}{k_B T}\right)}$$

with ρ_0 , ρ_1 and E_0 , E_1 the degeneracy and energies of the confined levels QD₀ and QD₁ respectively ($\rho_0 = 1$, $\rho_1 = 2$). E_0 , E_1 are extracted from Figure 2-a. N_{QD} is the quantum dot volume density, V_{QD} the total QD volume inside a square surface with lateral dimension W , and L the WL thickness. The considered surface corresponds to the laser spot. Capture $\tau_{WL \rightarrow 1}$ and intra-dot relaxation $\tau_{1 \rightarrow 0}$ times are defined using Auger ($C_{WL \rightarrow 1}$ and $C_{1 \rightarrow 0}$) and phonon ($A_{WL \rightarrow 1}$ and $A_{1 \rightarrow 0}$) coefficients as proposed in ref. 19 :

$$\tau_{WL \rightarrow 1} = \frac{1}{A_{WL \rightarrow 1} + C_{WL \rightarrow 1} N_{WL}} \quad \tau_{1 \rightarrow 0} = \frac{1}{A_{1 \rightarrow 0} + C_{1 \rightarrow 0} N_{WL}}$$

We assume here a geminate capture process of the electron-hole pair. Moreover, we introduced the occupation rates $f_0(t)$, $f_1(t)$, f_{WL} of the QD₀, QD₁ and WL level respectively.

They are defined by :

$$\begin{aligned} f_1(t) &= 1 - P_1(t) & f_0(t) &= 1 - P_0(t) & f_{WL} &= 1 \\ P_1(t) &= \frac{N_1(t) V_{1QD}}{4} & P_0(t) &= \frac{N_0(t) V_{1QD}}{2} \end{aligned}$$

with V_{1QD} the volume of one quantum dot, and $N_0(t)$ and $N_1(t)$ the populations of the QD₀ and QD₁ 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 QD₀ level for all the excitation optical density range. We introduced then in the model a direct relaxation path between the WL and the QD₀ level characterized by a capture time:

$$\tau_{WL \rightarrow 0} = \tau_{WL \rightarrow 1}$$

This model yields the resolution of the following rate equations:

$$\begin{aligned}\frac{dN_{WL}}{dt} &= \frac{N_{WL}}{\tau_0} + \frac{N_1}{\tau_{1 \rightarrow WL}} \frac{V_{QD}}{V_{WL}} f_{CM} - \frac{N_{WL}}{\tau_{WL \rightarrow 1}} f_1 - \frac{N_{WL}}{\tau_{spon}^1} - \frac{N_{WL}}{\tau_{WL \rightarrow 0}} \frac{V_{QD}}{V_{WL}} f_0 = 0 \\ \frac{dN_1}{dt} &= \frac{V_{WL}}{V_{QD}} \frac{N_{WL}}{\tau_{WL \rightarrow 1}} f_1 + \frac{N_0}{\tau_{0 \rightarrow 1}} f_1 - \frac{N_1}{\tau_{1 \rightarrow WL}} f_{WL} - \frac{N_1}{\tau_{spon}^1} - \frac{N_1}{\tau_{1 \rightarrow 0}} f_0 = 0 \\ \frac{dN_0}{dt} &= \frac{N_1}{\tau_{1 \rightarrow 0}} f_0 - \frac{N_0}{\tau_{0 \rightarrow 1}} f_1 - \frac{N_0}{\tau_{spon}^0} + \frac{N_{WL}}{\tau_{WL \rightarrow 0}} \frac{V_{QD}}{V_{WL}} f_0 = 0\end{aligned}$$

The number of photogenerated carriers in the WL is given by

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

with P_{exc} the average laser excitation power of the tr-PL setup, $h\nu$ the energy of the incident photons, f the repetition rate of the laser source, α ($\sim 10^4 \text{ cm}^{-1}$) the absorption coefficient in the InP barriers, L_z (30nm) the thickness of the InP

barrier crossed by the photons before reaching the WL, and $T^{trans} = \frac{4n_{opt}^{InP}}{(1 + n_{opt}^{InP})^2}$ the air/InP

transmission coefficient evaluated with n_{opt}^{InP} (3.9) the optical index of the InP. τ_0 is the

duration of the laser pulse. With this model the fitting parameters are $A_{WL \rightarrow 1}$, $C_{WL \rightarrow 1}$, $A_{1 \rightarrow 0}$,

$A_{1 \rightarrow 0}$.

The tr-PL curves obtained from these equations give a good fit of both the experimental rise and the decay times of the QD₀ and QD₁ levels tr-PL curves 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 QD₁ rise and decay curves also have exponential shape, but QD₀ decay curve show a plateau. This plateau is due to the saturation of the PL signal corresponding to a complete filling QD₀ 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 quantum dots. 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 ps to 8 ps, and we can identify two regimes: under approximately 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 $A_{WL \rightarrow 1} = 1.35 \cdot 10^{10} \text{ s}^{-1}$, $A_{1 \rightarrow 0} = 1.10^{10} \text{ s}^{-1}$, $C_{WL \rightarrow 1} = 5 \cdot 10^{-15} \text{ m}^3 \text{ s}^{-1}$ and $C_{1 \rightarrow 0} = 9 \cdot 10^{-14} \text{ m}^3 \text{ s}^{-1}$. These values are close to the ones found in the literature for similar systems [19, 20, 21].

To complete the study, we extracted from the model the intra-dot relaxation time $\tau_{1 \rightarrow 0}$ between the QD₀ and QD₁ confined states. This time could be measured directly with resonant excitation tr-PL experiment. This calculated intra-dot 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 double-cap InAs/InP quantum dots emitting around $1.55 \mu\text{m}$. 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 intra-dot relaxation time between the two confined levels has been estimated.

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Figure Caption

Figure 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.

Figure 2 : a) Sketch of the radiative and non-radiative mechanisms used to describe 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.

Figure 3 : Experimental rise time $\tau_{WL \rightarrow 0}$ ($= \tau_{WL \rightarrow 1}$ 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 continuous line.

Figure 4 : Intra-dot 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.









