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Carrier relaxation dynamics in InAs/InP quantum dots

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ABSTRACT

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 non-resonant time-resolved photoluminescence (tr-PL) experiments. This analysis reveals the QD electronic structure and the transient filling of the confined QD levels. Under low excitation densities, the spontaneous exciton lifetime is estimated and compared to previous time-resolved resonant and non-resonant 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.

Carrier Relaxation and band filling effects have been widely discussed in recent years [1,2,3,4,5,6,7,8,9,10] due to their key role in the operation of optoelectronic devices such as, for example, quantum dot lasers. The detailed understanding of the relaxation processes from a more fundamental point of view is also a field of interest. Due to the quantum confinement, many electronic states are present inside the quantum dot (QD), and the carrier thermalization process has been studied. It has been shown that efficient thermalization can occur by multiphonon [11,12] or Coulomb interaction process [13]. State-filling effect has also been analyzed in many systems. The excited states photoluminescence (PL) signal has been clearly evidenced at high excitation density [14,15,16,17,18,19]. Auger relaxation effects have also been investigated. It has been shown it is the dominant effect under high excitation density [20,21]. Theoretical studies pointed out that Auger transfer is extremely fast for example in InAs/GaAs QDs [22,23].

Despite all these analyses, informations about the carrier dynamics in InAs/InP QDs are still lacking. These QDs are more suitable for telecom applications since they emit in the 1.55 μm wavelength region. An original growth method has been proposed to obtain InAs/InP(113)B quantum dots with a tunable emission energy [24,29]. These structures, named double-cap quantum dots (DC-QDs), have been analyzed both theoretically [25,26] and experimentally [27,28,29], and yield a laser emission at 1.55 μm [29,30]. Confined electronic states have been identified, but informations on the dynamic properties of these levels are scarce. In this paper, we present a systematic study of the optical and electronic properties of the DC-QDs by using tr-PL spectroscopy. We deduce the exciton radiative lifetime of the confined levels and identify the relaxation processes.

The 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 [27] is then used to

control the QD maximum height. The maximum height of the QDs we analyze here is of 2 nm [27], and the average QD diameter is of about 35 nm. The surface density is of about 10^{10} cm^{-2} [28]. Samples are characterized by time-resolved photoluminescence spectroscopy (tr-PL) at low temperature. The experiments are performed at 10 K using a 790 nm mode-locked Ti-Sapphire laser (excitation in the InP barrier) 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 up-converting the luminescence signal in a nonlinear crystal with a time resolution of 20 ps and 1.2 ps respectively [28,34]. The luminescence peak of the chosen 2 nm DC QD excitonic ground state (QD_0) is located at 0.94 eV ($1.32 \mu\text{m}$) at $T=10 \text{ K}$. For this emission energy, the performances of the detectors are better than for DC QD with a maximum height equal to 3 nm emitting at 0.85 eV ($1.55 \mu\text{m}$ at room temperature).

Tr-PL spectra obtained by the up-conversion set-up for various times after excitation are shown on figure 1. The spectra are fitted with three peaks at energy positions corresponding to the excitonic states recorded in our previous studies [27] : the QD excitonic ground state (QD_0) at 0.94 eV, the QD first excited excitonic state (QD_1) at 0.99 eV and the wetting layer excitonic ground state (WL at 1.05 eV). We can notice that at short times ($t \sim 10 \text{ ps}$), the emission of the WL state is the most important one. The QD_0 ground state emission is however also observed, due to the possible direct carrier injection channels from the barrier to the QD_0 state. For longer times, the carrier population in high energy states diminish either by light emission or by relaxation to the ground state QD_0 . A systematic study was performed with the streak camera. Spectra obtained at 10 K with various optical excitation densities for a long time (1650 ps) after excitation are reported in Fig. 2-a. These spectra are very similar whatever the excitation density is. They are equivalent to the one obtained at short times after excitation with a low incident optical power (figure 2-b). In this case indeed (150 ps, 16 W.cm^{-2}), the luminescence of the QD excitonic ground state (QD_0 , 0.94 eV), and the wetting

layer (WL, 1.05 eV) are observed. When the excitation density increases, from 45 to 57 W.cm⁻², a luminescence signal appears at about 0.99 eV. It corresponds to the luminescence of the QD first excited state (QD₁) also identified in ref. 27. Then, when the excitation density is further increased, we observe a stabilization of the carriers in the WL and the QD₁ states, and then an corresponding intense luminescence. This behavior reveals the progressive filling of the QD₁ and the QD₀ levels. This analysis shows that the state filling plays an important role in the QD dynamics and emission properties. It can, for instance, explain the slight shift to high energies of the QD₀ peak that can be observed in Fig. 2-a at long times after excitation for high excitation density. We have performed an estimation of the average number of excitons per QD produced by the optical excitation. It can be estimated with :

$$N_0 = \frac{P_{exc}}{f h \nu} T_{trans} (1 - e^{-\alpha L}) \quad (1)$$

with P_{exc} the average excitation density, $h\nu$ the energy of incident photons, f the laser repetition rate, α the absorption coefficient of the InP barrier ($\sim 10^4$ cm⁻¹), L the InP barrier thickness (30 nm) and T_{trans} the air/InP interface transmission coefficient. The filling of the QD₁ level observed at 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 QD₁ level is associated mainly to a saturation of the QD₀ level after formation of the biexciton ground state.

We have also performed an analysis of the QD₀ level radiative lifetime. Many studies have already been done yet in quantum wires [31] and quantum dots [32,33]. A direct experimental method is to use resonant excitation [34, 35]. In a pump-probe experiment, we have already studied DC-QD but emitting at 0.8eV at room temperature [35]. In the present work, we study the QD₀ radiative lifetime of our DC-QD with non-resonant excitation measurements. In addition to the radiative process, relaxations from higher energy states and saturation of the ground state must be taken into account in these conditions. As the capture

process to the QD₀ state depends on the excitation density, we analyzed the QD₀ decay time under low excitation density. The measured QD₀ decay times are reported in Fig. 3 as a function of the excitation density and of the calculated number of exciton per QD. Under low excitation density, when the average number of carriers per dot is about 1, the decay time is constant, of about $\tau \sim 1150$ ps. Under this regime, the QD₀ level is not saturated (the average population per dot is smaller than 1), and we assume then that the exciton recombines with a characteristic time close to the radiative lifetime. This value has been compared to theory. In the strong confinement regime, the spontaneous emission rate is given by the Einstein's coefficient :

$$A = \frac{1}{\tau_{QD}^{rad}}$$

This coefficient can be written by considering only a transition between the electron and the heavy-hole [34,36]:

$$\frac{1}{\tau_{rad}} = \frac{n_{op} e^2 \omega_0 E_p}{3m_0 h c^3 \epsilon_0} \left| \langle F_C | F_V \rangle \right|^2$$

The envelope wave function overlapping coefficient $\left| \langle F_C | F_V \rangle \right|^2$ is calculated using a model described elsewhere [28]. We find $\left| \langle F_C | F_V \rangle \right|^2 = 0.92$ for the fundamental transition. The use of the 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 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.

The different rise and decay times measured at energies corresponding to the WL, QD₀, and QD₁ levels suggest the presence of cascade relaxation path. Moreover, we observed variations of rise and decay times measured under different temperature and excitation

regime. This could be interpreted as the coexistence of two relaxation processes : Auger and phonon relaxation processes. To check this point, a detailed study has been done by measuring the tr-PL rise times $\tau_r^{QD_0}$ (Fig. 4-a) and $\tau_r^{QD_1}$ (Fig. 4-b) of QD₀ and QD₁ levels respectively as a function of both excitation density and temperature. Under high excitation density, the measured rise times of both levels are independent of temperature ($\tau \sim 30$ ps) ; it is probably limited by the time-resolution of the Streak camera set-up. The Auger relaxation process is dominant in this regime, yielding a very fast relaxation process. 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. This behavior has been observed in InAs/GaAs quantum dots [2]. It has been shown that multiphononic relaxation process is strongly temperature dependent ; different types of phonons are present in this last system, leading to a reinforcement of the effective electron-phonons coupling [37,38] and the possibility of multiphononic relaxation [39]. Theoretical studies have shown that the Huang-Rhys factor, representing the strength of the electron-phonon coupling, increases with the size of the dot. Experimental results also confirm that this factor is enhanced by a factor 5 in InAs/GaAs quantum dots in comparison with bulk InAs [37]. These interpretations could explain the strong influence of phonons in exciton relaxation in our system. In a study of InAs/GaAs QD combining quasi-resonant and non-resonant TRPL experiment, Ohnesorge *et al* [2] have dissociated the relaxation process (phonon and Auger) of carriers. They used a three level model involving the GaAs matrix and two confined states in the InAs QD. This model can be considered as equivalent to ours if we take into account three levels constituted by the WL and the two QD confined states QD₀ and QD₁. They showed that, under weak excitation density, the rise time of the fundamental $\tau_r^{QD_0}$ level can be written :

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

This equation corresponds, according to them, to a cascade relaxation of excitons between the three considered levels assisted by phonon emission. Ohnesorge *et al.* then analyzed the effect of optical excitation density. They evidenced that, under strong excitation regime, luminescence decay time of the barrier $\tau_{Barrier \rightarrow QD_1} = \tau_r^{QD_1}$ is close to $\tau_r^{QD_0}$, the rise time of the fundamental level. Thus, they evidenced a direct Auger effect responsible for the direct capture from the barrier into the dot. The relation $\tau_r^{QD_0} \rightarrow \tau_r^{QD_1}$ implies then, according to equation (2), $\tau_{QD_1 \rightarrow QD_0} \rightarrow 0$ under strong excitation regime. Moreover, the decay time of the first excited level QD₁ can be written :

$$\frac{1}{\tau_d^{QD_1}} = \frac{1}{\tau_{QD_1 \rightarrow QD_0}} + \frac{1}{\tau_{spont}^{QD_1}}$$

Then, if the condition $\tau_{QD_1 \rightarrow QD_0} \rightarrow 0$ is realized, that is to say that a direct efficient Auger effect can occur, one can induce it yields a decrease of the QD₁ photoluminescence level decay time. We can approximate our system as equivalent to the one described in Ref. 2: three levels corresponding to a WL level and two QD confined states. The QD₁ decay times have been measured and the values reported in Fig. 4-c for different excitation powers. The spectra clearly show a reducing of the QD₁ decay time. This behavior, if we rely on the results of Ohnesorge *et al.*, evidence then in our three level system the presence of a direct Auger relaxation effect, where carriers directly relax from the WL into the QD₀ level.

Conclusion

We analyzed the carrier relaxation processes in InAs/InP(113)B double-cap quantum dots. We showed the progressive filling of the identified confined levels QD₀ and QD₁ with the number of photo-generated carriers. The QD₁ level starts filling with an average number of carriers per dot of about 3. We have also measured the QD fundamental level radiative

lifetime which is in agreement with the theoretical value. By 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 QD₁ photoluminescence decay time with the excitation density has confirmed the presence of a direct Auger effect, responsible for the direct capture of carrier from the WL into the QD fundamental level.

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Figure Caption

Figure 1 : PL spectra recorded at 10 K with the up-conversion setup at different times after excitation (10, 600 and 1500 ps) for an excitation density of $I_{\text{exc}} = 70 \text{ W.cm}^{-2}$.

Figure 2 : Tr-PL spectra of QD_1 level extracted from the streak camera data at 10 K for different excitation densities at (a) long times (1650 ps) and (b) short times (150 ps) after excitation. The excitation densities are $I_{\text{exc}} = 16, 45, 57, 74, 155$ and 230 W.cm^{-2}

Figure 3 : QD_0 decay time reported as a function of the optical excitation density and the calculated average number of exciton per QD. The inset shows a typical kinetics from which the decay times are measured. $T=10 \text{ K}$

Figure 4 : Evolution of the rise times a) $\tau_r^{\text{QD}_0}$ of the fundamental (QD_0) and b) $\tau_r^{\text{QD}_1}$ of the first excited state (QD_1) as a function of the temperature. The plotted data correspond to two excitation regimes: strong (200 W.cm^{-2}) and weak excitation density (18 W.cm^{-2}). Dashed lines are a guide for the eye. Time evolution spectra recorded at 10 K and at the energy of the first excited state (QD_1) level for different excitation densities are reported in c).