Thermal activated carrier transfer between InAs quantum dots in very low density samples

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Abstract. In this work we develop a detailed experimental study of the exciton recombination dynamics as a function of temperature on QD-ensembles and single QDs in two low density samples having 16.5 and 25 dots/µm². We corroborate at the single QD level the limitation of the exciton recombination time in the smallest QDs of the distribution by thermionic emission (electron emission in transient conditions). A portion of these emitted carriers is retrapped again in other (larger) QDs, but not very distant from those emitting the carriers, because the process is limited by the diffusion length at the considered temperature.

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1. Introduction

In the last years it was made great efforts to study self assembled InAs Quantum Dots (QDs) by micro-Photoluminescence (μPL) [1]. It is of particular interest the study of single QDs in low density samples, because of their potential use in single photon and entangled photon sources for quantum information processing [2-3]. The limitation of the use of QDs in such applications comes from the temperature effect on carrier-phonon interaction and electron-hole recombination. During the last decade, a great effort has been made studying the temperature evolution of QD emission, obtaining good agreements between experimental data and kinetic exciton recombination models [4]. Thermal escape through the wetting layer (WL) or by phonon assisted tunneling is usually claimed as the most suitable mechanism to describe carrier transfer between QDs both in monomodal and bimodal distributions [5-7].

In the present study we have analyzed this phenomenon in two different samples containing very low density of InAs/GaAs QDs. This is important because we can separate the μPL spectra from different QDs within the illumination area and hence compare more directly microscopic and ensemble exciton recombination dynamics. A detailed experimental study as a function of temperature has been carried out by using ensemble photoluminescence (PL), μPL, time resolved PL (TRPL) and μTRPL techniques. In both samples coexist two QD size distributions: (i) a small size one emitting in the region 1.25-1.35 eV (SQD family) and (ii) a large size one emitting in the region 1.05-1.20 eV (LQD family), as shown in Fig. 1. For these samples we observe a common phenomenology interpreted as follows: QDs belonging to the SQD family have electron confined states close to the WL ones and emit carriers towards such extended states, where they can diffuse and recaptured by other QDs not very far in space (within the diffusion length at the InAs WL at the lattice temperature) with smaller confinement (emitting and lower energies). These QDs emit at the low energy tail of the SQD PL band in the case of the sample with the lowest QD density and at the LQD PL band in the...
second sample where the spatial separation between LQD- and SQD-like dots is short enough (smaller than 200 nm).

2. Sample design and Experimental Set-Up:
The InAs QDs investigated in this paper were grown by MBE on a GaAs semi-insulating substrate. The combination of low growth rate (LGR) and graded coverage allows obtaining very low density samples [8]. In particular, the two samples under study have a density of 16.5 and 25 dots/µm², as estimated by atomic force microscopy images. The PL characterization was performed by using a standard backscattering configuration by holding the sample in a closed cycle cryostat, whereas the µPL setup was based on a confocal microscope inserted in the He exchange gas chamber of an immersion cryostat. In this case, two optical fiber connections to the microscope were used for optical excitation and detection light. Collected light was dispersed by a 0.3 m (0.5 m) focal length double (single) spectrograph and detected with a Si back-illuminated cooled CCD (cooled InGaAs photodiode array) for wavelengths shorter (larger) than 1000 nm. The excitation source was a tunable Ti:sapphire laser, which was operated under either continuous wave or pulsed (<200 fs) modes. Time resolved µPL (µTRPL) measurements were performed by using a Time Correlated Single Photon Counting board to analyze the light dispersed by the same monochromator and detected with a Si APD detector operating in Geiger mode. The temporal response of the system is around 400 ps (full width at half maximum), but time constants above 100 ps can be perfectly deconvoluted in the µPL transients.

3. Results and Discussion:
Figure 1 shows PL spectra of the two studied samples. In these PL spectra we observe two PL bands clearly associated to two main families of sizes, as also detected by AFM, labeled here as Large QD (LQD) and Small QD (SQD), relative to QDs of higher (quasi-pyramidal shape) and smaller (lens shape) sizes. Such a bimodal size distribution has been reported by other authors for samples grown under similar conditions [9]. In Fig. 1 we observe that the PL band associated to the LQD family in the sample of lower QD density (green line) is very week as compared to that of higher QD density (blue line). In fact, the integrated intensity ratio between SQD and LQD PL bands decreases from about a factor of 20 to 0.45 from one to the other sample. From these ratios we would find in average around 0.8 LQD + 16 SQD per µm² in the sample with the lowest QD density (16.5 µm²) and 8 LQD + 17 SQD per µm² in the sample with the highest QD density (25 µm²), approximately. These proportions are close similar to those deduced from AFM images of our samples by measuring height/base dimensions of many QDs [10] and will be important to determine the retrapping of carriers in other QDs different from those where they are escaping (they will be mainly those belonging to the SQD family) towards the WL states in the low temperature range, as discussed below.

Figure 2(a) shows a detail of the SQD-16 (SQD family at the sample with 16.5 QDs/µm²) related PL band together with the PL decay times measured at different PL energies inside this band (blue solid squares), as compared to decay times measured by µTRPL at single QDs of this family emitting at different energies (red solid circles). All TRPL and µTRPL transients exhibit a perfectly exponential decay over more than one decade (approximately the experimentally measured signal to noise ratio). The equivalence between decay times measured under micro (at the neutral exciton µPL line of different QDs) and ensemble (at different detection energies) conditions is indicative of a recombination determined by carriers at the ground state.
The observed decrease of the PL decay time when the detection energy is close to the WL emission (1.428 eV) should be attributed to a non-radiative mechanism that depends on energy and temperature (see experimental data in Fig. 2(b)). This mechanism must be attributed to the thermal promotion of carriers to the WL states. The most widely accepted exciton recombination model postulates a "thermionic emission" of excitons into the WL [4], implying a thermal activation energy for the PL intensity equivalent to the energy difference between the WL emission energy and that considered within the SQD emission band, $E_{WL} - E_{QD}$. We find that this is true in the case of the SQD PL bands for both samples measured under continuous wave excitation, but we cannot explain the evolution with temperature of the PL decay times (as a function of the PL energy), as the ones represented in Figure 2 (b). We can take account of the observed evolution if we calculate $\tau_D$ with a thermal escape term less restrictive:

$$\frac{1}{\tau_D} = \frac{1}{\tau_R} + \left( \frac{1}{\tau_{NR}} \right) + \frac{f_e (E_{WL} - E_{QD})}{e^{\frac{KT}{\tau_e}}}$$

being $\tau_R$ the radiative lifetime of excitons in a SQD, $\tau_e$ the capture (emission) time of an exciton by a SQD from the WL reservoir (for $E_{QD} = E_{WL}$) and $f_e$ a fitting parameter to take account of the observed dependence of $\tau_R$ with energy and temperature in Figure 2(b), and whose physical meaning will be discussed below. We have added a non-radiative recombination term, which may be due to an eventual capture of carriers by dislocations or impurities. Prior to the fitting procedure let mention that we have simulated $\tau_R(E)$ by a linear variation (that measured at the lowest temperatures below 1.36 eV: blue, violet and black curves in Fig 2(b)) to obtain the best fits to the experimental data and a finite constant value for $\tau_{NR}$ in the range from 4 to 20 ns to fit data for $T > 50$ K, in order to present a cleaner fit over most of the temperature range with the same value for $f_e$.

At temperatures above 70 K we observe a decrease of $\tau_D$ with PL energy within the whole emission band (orange, red and black symbols in Fig. 2(b)), whereas it increases at temperatures below 60 K in most of the SQD-16 PL band (see the rest of experimental curves in Fig 2(b)). The increase of $\tau_D$ with PL energy (equal to $\tau_R$ at the lowest energies and temperatures) can be due to the wavefunction delocalization of carriers whose states are very close to the WL electron states, as occur in the case of very shallow confined electrons [11]. This hypothesis would be consistent with a thermal escape of electrons as the possible origin of an activation energy for $\tau_D$ smaller than the thermionic value [12]. In fact, the best fit of the whole experimental data set between 40 and 90 K in Fig. 2(b) to Eq. 1 is achieved by using a fixed value $f_e = 0.35 \pm 0.05$ and a slight variation for $\tau_e$ between 11 to 42 ps, which may be due to the assumption of a capture time independent of the emission (confinement) energy. In principle one would expect a greater contribution to $\tau_e$ from energies near $E_{WL}$ at higher temperatures. That value of $f_e$ would be consistent with the hypothesis about an activation energy equal to the energy difference between confined electron states in QDs within the SQD-16 family to states at the WL. In this way, $f_e$ would then be the fraction of the electron binding energy (measured from the bottom electron WL state) with respect to
the total thermionic energy, \( \Delta E_e/\left(\Delta E_e + \Delta E_h\right) \). This factor can be (as estimated from calculated electronic structures) greater than 0.5 for pyramidal and big lens shape QDs [13], but around 0.4 for a lens shape QD 2.5 nm high [14] and less than 0.3 in a truncated pyramidal QD 1 nm high [15]. Furthermore, we find that the value of \( f_e \) decreases from 0.35 at 40 K to 0.12 at 12 K: let note that QDs at the high energy tail of the SQD-16 band are emitting at only 40 meV below the WL. This reduction in \( f_e \) can be understood by the different electron and hole effective masses, producing a saturation of the electron energy level for very small QDs (the only ones at which electron escape is possible at the lowest temperatures), whereas holes can be still exhibit a variation in their binding energy. One could also assume a change in \( W_e \), but this is not able to fit appropriately the whole energy variation for \( W_D \) at these temperatures, whereas the saturation of \( \tau_e \) below 50 K seems reasonable as an average capture time for the whole SQD band.

The unipolar (electrons) escape mechanism in transient regime is compatible with the ambipolar thermal escape mechanism (thermionic emission) under CW excitation (also observed experimentally for SQD families, as noted above), since the electron escape would be faster than the hole escape, hence determining the decrease of \( \tau_D \) with \( E \), more important at higher temperatures, as reflected by Eq. 1. Once the electron is emitted, the resulting electric field due to the unbalanced charge will favour the emission of the hole, assuring the thermionic emission under steady state conditions.

So far, we have explained a recombination model of excitons confined in QDs of the SQD-16 family, but perfectly applying to the case of the SQD-25 family in the second sample, but smaller the effect of carrier emission toward the WL given the higher energy distance. Decay times measured by \( \mu \)TRPL in many single QDs of this sample are compatible with those measured at SQD-16 family, even if we find a marked dispersion in the measured values. The difference between both samples can be attributed to the greater influence of carrier capture by non-radiative recombination centres (\( W_{\text{NR}} \) below 3 ns), probably dislocations.

In addition to the thermal escape of carriers to the WL (electrons in a first time scale) we also observe in our samples a certain carrier refilling of QDs emitting at lower energies than those whose PL is quenched by thermal escape of carriers. In the case of the sample with the lowest QD density the refilling effect is mainly observed in QDs emitting at the energies below that of the SQD-16 PL peak. In the sample with the higher QD density a portion of the carriers emitted from QDs of the SQD-25 family into the WL are captured by QDs belonging to LQD-25 family. This behaviour is observed as an increase of its corresponding integrated PL intensity in the temperature range for which the PL band related to the SQD-25 family is practically quenched and the loss mechanism of carriers at the WL and QDs of the LQD-25 family are neither limiting nor masking the refilling effect. However, this mechanism appears to be effective in LQDs not too distant from the SQD emitting the carriers toward the WL, as illustrated in Fig 3. This figure represents the \( \mu \)PL (at medium-high excitation power) of a few single QDs belonging to the SQD and LQD families within the illumination area (slightly less than 1 \( \mu \text{m}^2 \)). We clearly observe how the intensity of the LQD related emission increases with increasing temperature (74-94 K range) at the same time that decreases the intensity of the SQD related emission. This is a direct microscopic demonstration of the transfer of carriers from small (emitting at energies close to the WL) to large QDs (emitting at lower energies). The carriers emitted from a SQD to the WL states diffuse within this InAs layer before being trapped in a not too distant QD for which the thermionic emission effect is not important, which are those belonging to the tail of the SQD or LQD related PL bands. It is noteworthy that the carrier diffusion length at the WL would be of the order of...
700 nm, if the mobility (limited by holes) and the electron-hole recombination time in the WL at 70-90 K are the order of $10^4$ cm$^2$/Vs and 75 ps, respectively. Such diffusion length value would be consistent with the observation of a refilling effect in dots of the LQD-25 family, but not in those of the LQD-16. In fact, the average distance between dots from SQD-16 and LQD-16 families is greater than 1 μm, whereas decreases below 200 nm in the case of dots from SQD-25 and LQD-25 families.

4. Conclusion
In summary, samples with very low density of InAs QDs (16.5x10$^8$ and 25x10$^8$ cm$^{-2}$) exhibit a common phenomenology interpreted as follows: very small QDs have electron confined states close to the WL ones and emit carriers towards such extended states, where they can diffuse and recaptured by other QDs not very far in space (within the diffusion length at the InAs WL at the considered temperature) with smaller confinement and hence emitting and lower energies. The recombination dynamics have been proved by combining ensemble and μ-PL/TRPL experiments.

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