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Carrier scattering by Auger mechanism in a single quantum wire

J. Bellessa[1,a], V. Voliotis[1,2,b], T. Guillet[1], D. Roditchev[1], R. Grousson[1], X.L. Wang[3], and M. Ogura[3]

[1] Groupe de Physique des Solides, CNRS, Universités Pierre et Marie Curie et Denis Diderot, 2 place Jussieu, 75251 Paris Cedex 05, France
[2] Université Évry-Val d'Essonne, boulevard François Mitterrand, 91025 Évry Cedex, France
[3] Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba 305, Japan
and
CREST, Japan Science and Technology, 4-1-8 Honcho, Kawagushi 332-0012, Japan

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Abstract. We report on time-resolved microphotoluminescence experiments in a single GaAs/GaAlAs V-shaped quantum wire as a function of optical excitation intensity. At low pump power we observe that excitons are localized in quantum boxes formed by the local potential minima existing along the wire axis. As the pump power is increased, state filling of the lowest lying levels of the boxes appears. When two carriers occupy the first excited level of the box, a very efficient Auger scattering occurs, leading to a transfer of carriers from one box to another neighbouring one. The intradot Auger scattering time has been measured and is of the same order of magnitude as the LA-phonon emission rate.

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1 Introduction

Carrier relaxation and carrier capture in zero-dimensional quantum structures is a remaining question which understanding, apart from the basic scientific interest of the issue, is an important requirement for the improvement of eventual optoelectronic devices. For quantum boxes (QB) due to the discrete spectrum of energy levels, one would expect that at low excitation intensities, emission from excited states should be observed due to the restricted inter-level relaxation rates [1,2]. However several groups reported on rapid carrier relaxation in different QB systems [3–5] at low excitation intensities. Recently we have observed a clear dependence of relaxation times *via* LA-phonons with the inter-level energy spacing [6] and indeed a slowing down of relaxation as the inter-level spacing exceeds a few meV has been evidenced. Nevertheless, the relaxation times remain very fast (< 100 ps) showing that other mechanisms like multi-phonon relaxation [7,8] or Auger-like scattering [9] are very efficient and leading to rapid thermalization of carriers and emission from the lowest lying level.

Furthermore, the finite degeneracy of the QB levels should lead to state filling effects even when a few carriers populate the lowest dot states [10]. This has been clearly observed at high pump power intensities where saturation effects and emission from excited state transitions have been reported [11,12].

In this paper we study time-resolved microphotoluminescence (μPL) of a single GaAs/AlGaAs V-shaped quantum wire (QWR) under high excitation intensities. We have shown previously by μPL experiments at low pump power, that strong localization occurs due to the thickness fluctuations along the axis of the wire [13]. These local potential minima can be viewed as "naturally" formed QBs with a weak confinement potential, leading to the observation of sharp excitonic lines attributed to these bound states. This picture has been also confirmed by other studies in different kinds of V-wires [14,15]. Therefore exciton dynamics are governed by the properties of a QB system at low temperature. The exciton relaxation and recombination processes in the low optical excitation regime have been investigated in [6].

Here we show that when a collection of QBs, specific to our sample, is excited at high excitation intensity, state filling effects appear in the lowest lying levels of the boxes which is a consequence of the Pauli exclusion principle. However, no emission from excited state transitions has been observed but rather a rapid transfer of carriers from one QB to another neighbouring one, through a mechanism that we have attributed to Auger scattering.

[a] *Present address:* Département de Physique des Matériaux, CNRS, Université Claude Bernard Lyon I, 43 boulevard du 11 Novembre 1918, 69622 Villeurbanne Cedex, France
[b] e-mail: voliotis@gps.jussieu.fr

We show that the intradot Auger scattering is a very rapid mechanism of the order of 20 ps.

The paper is organized as follows: in Section 2 we describe the studied sample and give some details on the experimental set-up. Section 3 is devoted to the experimental results. We show first μPL and time-resolved μPL experiments *versus* excitation intensity, excited in the first transition of the QWR what we will call hereafter *non-resonant* experiments. Then a *single* QB is selectively excited and studied by means of what we call *resonant* μPL.

2 Samples and experimental set-up

GaAs/GaAlAs quantum wires (QWRs) are grown on a 4.8 μm pitched V-grooved GaAs substrate by flow rate modulation epitaxy which is a modified metal organic vapour phase epitaxy technique [16]. The Al concentration rate in the barrier is 0.33 for the (001) mesa top face and the thickness of the QWR at the center of the V-groove is about 5 nm. In order to enhance the photoluminescence (PL) signal, the (001) region and part of the (111) side wall AlGaAs barrier layer are removed by wet selective chemical etching [16]. The lateral size of the wire where the first electronic wave function is confined is about 15 nm.

The sample is fixed on the cold finger of a Helium cryostat and cooled at 10 K. The laser beam is focused on the sample by a microscope objective with a large numerical aperture (0.6), and the laser spot diameter is 1 μm. As the different wires on the sample are separated by 4.8 μm, in our experiments only one single wire is excited over 1 μm. Time-resolved experiments have been performed using a mode locked argon laser pumping synchronously a dye laser. The pulse width is 5 ps and the repetition rate is 82 MHz. The time-decay of the μPL signal is analyzed through a double monochromator operating in the subtractive mode and detected by a time-correlated single photon counting system with a time resolution of 40 ps. We systematically use a convolution procedure by the temporal response function of the detection system in order to achieve a time resolution of 20 ps. The spectral resolution in the time-resolved and continuous experiments is 200 and 50 μeV respectively. The number of electron-hole pairs created in the wire can be estimated from the following relation: $N = (\alpha\ell P\tau_0/h\nu)\beta$, where $\alpha\ell$ represents the absorption probability in the material. A typical value which is usually taken in the literature is $\alpha\ell = 0.6\%$ in a GaAs quantum well [17]. P is the excitation power, τ_0 is either the radiative lifetime of excitons in a continuous excitation or the repetition period in a pulsed excitation, and β is the ratio between the illuminated wire surface and the laser spot surface. This geometrical factor is related to the coupling with light and is thus different from one sample to another. From this relation, it appears that when the pump power density is lower than 100 W/cm^2 (continuous excitation), there is always less than one electron-hole pair per QB in average. For pulsed excitation the pump power density must be lower than 25 W/cm^2. This estimation is made assuming that the mean radiative lifetime is about 300 ps, which is consistent with our experimental results and with the fact that about ten QBs are present in 1 μm, which corresponds to the mean number of peaks observed in the μPL spectrum. Here all the presented experiments have been performed on the same sample and the factor β is taken equal to 0.05.

3 Results and discussion

3.1 Non-resonant μPL experiments

μPL spectra with different excitation intensities are given in Figure 1a. All these spectra have been recorded at the same position on the sample and they are reproducible from scan to scan. The excitation energy is 1.689 eV which corresponds to optical absorption in the QWR. Then the carriers can relax in all the QBs present in the section of wire illuminated by the laser spot. Indeed we have observed that there is no carrier diffusion at low temperature in this sample when exciting in the QWR absorption band. The spectrum pumped at 15 W/cm^2 corresponds to an excitation intensity where less than one exciton is present in the structure. The spectrum is formed by a line which is split into several sharp peaks of less than 0.5 meV width. The line is centred around 1.647 eV and the peaks are distributed over about 10 meV. We have shown previously [13] that these peaks can be attributed to lowest lying level emission of localized states due to size inhomogeneities of ± 1 atomic monolayer (ML) in the most confined direction of the wire (5 nm), at the top of the (001) surface. When the excitation power is progressively increased, several interesting features appear on the spectra. We clearly observe that the sharp peaks do not shift in energy, are not broadened (in the limit of our resolution) and their intensity saturates progressively as a background starts to grow up. For an excitation density of 10 kW/cm^2, the sharp peaks have completely disappeared and a broad emission line is observed. If the intensity is further increased up to 100 kW/cm^2 then the high energy side of the PL line grows up. As the excited states are separated from the lowest lying states of the QBs by a mean energy value of 3–4 meV [6], if emission from excited states occurs, it should be clearly observable on the μPL spectra. As the centre of the line is not shifted, we can conclude that emission from the excited states does not appear in this power range. Figure 1b shows the integrated total luminescence intensity as a function of excitation power which has a quasi-linear behaviour over four orders of magnitude of the pump power. The small deviation from an exact linear behaviour is probably due to the uncertainty of the intensity measured at very low excitation power. We have also checked that the μPL temporal decay depends neither on the excitation energy nor on the excitation power [6]. These are good indications that radiative recombination is predominant in this intensity regime.

The origin of the background is qualitatively understood. When the number of electron-hole pairs becomes important, we may attribute it to a multitude of states

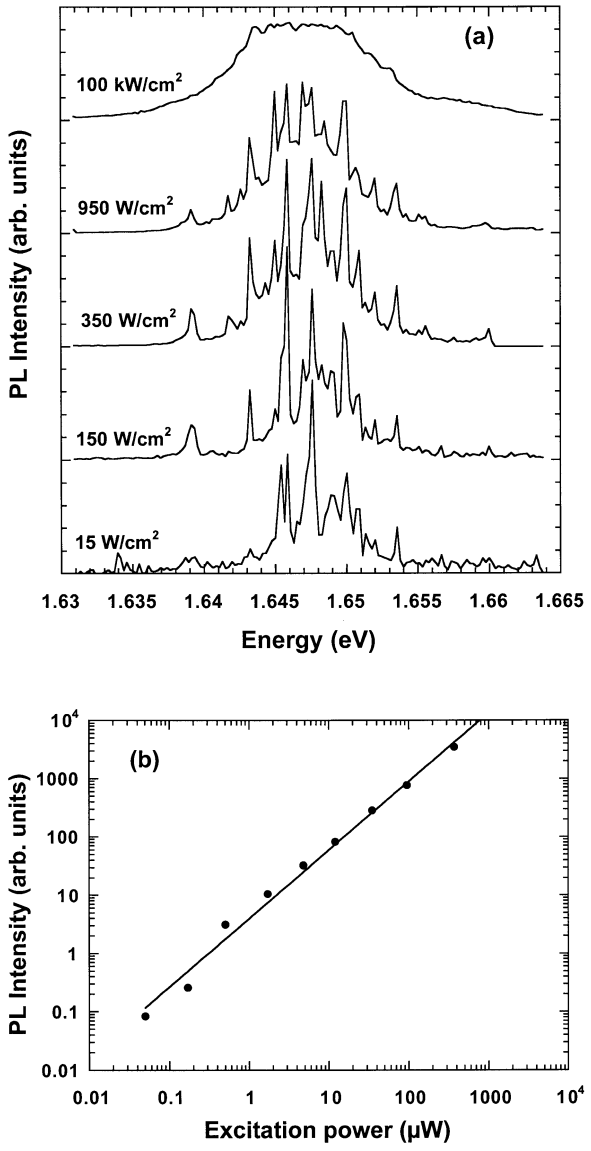

Fig. 1. (a) Some typical non-resonant μPL spectra of a single QWR at different excitation intensities. The excitation energy is 1.689 eV; (b) shows the total integrated luminescence intensity as a function of pump power with the linear fit of the experimental data (the slope is equal to 1.18).

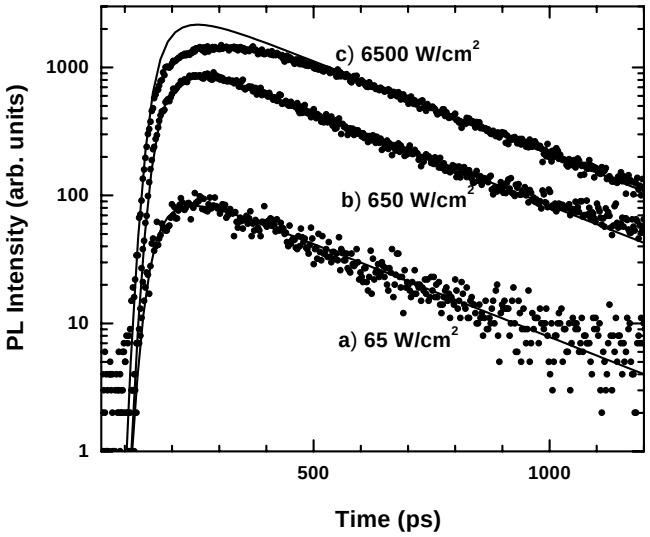

Fig. 2. Non-resonant time-resolved μPL experiments of a given QB (detected at 1.6475 eV) at different pump power densities. The dots correspond to the experimental data and the solid line to the calculated curve obtained by equation (1) convoluted by the temporal response of the detection system.

which become accessible due to coulomb interaction between carriers. Indeed we have experimental evidence of exciton- exciton interaction, like the observation of a biexciton on the low energy side of the μPL line as we increase the pump power. However this observation is not systematic in all the probed samples [18]. We believe that the formation of biexcitons strongly depends on the size of the boxes along the wire, thus on the heterointerface quality. Crottini *et al.* [19] have observed in similar V-grooved wires by near field optical spectroscopy extended exciton states where the formation of biexcitons was clearly evidenced. But this question is beyond the scope of this work and we will not further discuss it.

In similar experiments performed by Vouilloz *et al.* [20] the authors observe a clear blue shift of the μPL lines. It is attributed to the inter-exciton exchange interaction between localized excitons and the 1D gas of weakly interacting excitons present for intermediate excitation intensity. The main difference with our experiments is the thickness of the QWRs which is twice larger in our case (5 nm instead of 2.5 nm). This could explain that the shift of our μPL lines is smaller than our experimental resolution because the exciton-exciton interaction is expected to strongly increase with increasing confinement [21].

Figure 2 depicts typical time-resolved μPL experiments of a given QB taken with different pump power densities. The behaviour of the μPL time-evolution is similar for all the studied QBs. The time-resolved spectra are fitted assuming a simple three levels model by using the following expression:

$$I(t) = A\frac{(\mathrm{e}^{-t/\tau} - \mathrm{e}^{-t/\tau_0})}{(\tau - \tau_0)} \qquad (1)$$

which is then convoluted by the temporal response of the detection system. In this way the rise time corresponds to τ, the relaxation time from the first excited level to the lowest lying state and the decay time to the radiative lifetime τ_0 of the carriers. Spectra 2a and 2b have the same shape, *i.e.* they can be fitted accurately with the same parameters of 45 ps for the relaxation time and 295 ps for the radiative lifetime. The intensities at the maximum of the curves are in the ratio of the excitation power, since the luminescence intensity is linear in this power regime. Spectrum 2c shows two characteristic features: for short delays, a clear saturation of the PL intensity is observed with the characteristic plateau-like shape of the spectrum [12] and after 500 ps from the laser pulse,

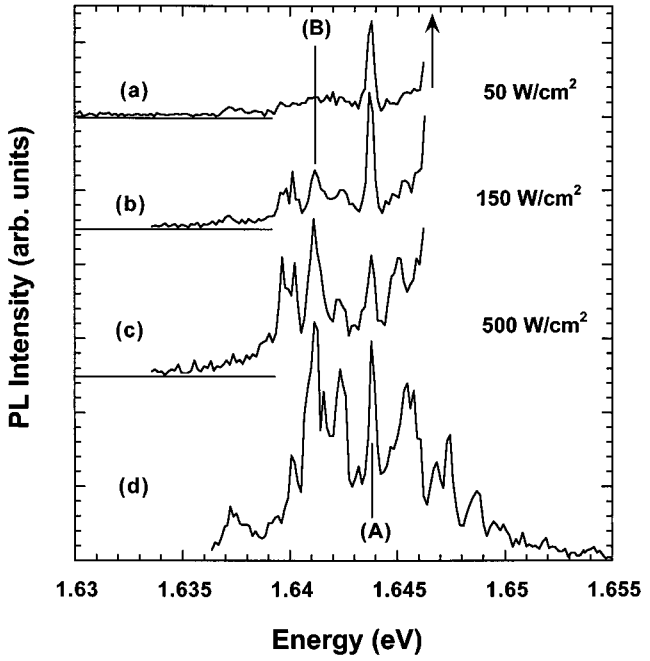

Fig. 3. (a), (b) and (c) are resonant μPL experiments for three different excitation densities; (d) is a non-resonant μPL spectrum of a single QWR recorded at the same wire position as in (a), (b) and (c); The excitation energy is 1.6464 eV and represented by an arrow; (A) and (B) are two μPL peaks corresponding to the lowest level emission of two different in size QBs.

when the number of pairs in the fundamental level has decreased, the decay time has the same exponential behaviour as in 2a and 2b. This saturation is due to state filling of the fundamental level of the QB and explains the discrepancy obtained between the simple model used (Eq. (1)) and the experimental data. State filling has been observed in QBs but always associated to emission from excited states, due to the blocking of relaxation down to the saturated fundamental level. In the present QWRs we have a situation where saturation of the QB fundamental levels occurs without the expected blue shift of the luminescence line associated to the emission from excited states.

3.2 Resonant μPL experiments

In order to have a better understanding of these results, *resonant* μPL experiments have been performed in a single QB. A single QB can be isolated and studied by adding to the spatial resolution a selective excitation. This can be done only if micro-photoluminescence excitation (μPLE) experiments are performed on a single QB in order to determine its excited levels [6]. Figure 3d represents a non-resonant μPL spectrum of a single QWR at low pump power. We have performed μPLE of the QB labelled A on the figure and found that the first excited state lies 2.8 meV above the lowest level [22]. If resonant excitation is performed on this first excited state we observe that only peak A remains on the spectrum showing that this level is the fundamental associated to the excited level of the same box (Fig. 3a). Here the excitation intensity is very low and there is always less than one electron-hole pair present in the box. Therefore resonant μPL performed at low intensity demonstrates that the QBs in our structure are spatially isolated. When the power is increased the intensity of peak A saturates while we see new emission lines emerging (Fig. 3b, c). Their energy positions correspond exactly to those observed in the non-resonant μPL spectrum (Fig. 3d) recorded at the same sample position at low pump power density. Since all the sharp features in the non-resonant spectrum are fundamental emission lines of QBs with different sizes, this clearly indicates that carriers transfer from the selectively excited box to other neighbouring ones as the power is increased. We have also checked that the intensity of each μPL peak is linear with increasing power which means that there is no formation of biexcitons. Emission from excited states does not appear and no blue shift of the resonant μPL is observed. We can also exclude a local heating of the sample because no broadening of the sharp peaks nor a shift towards lower energies occur [23].

We have attributed this transfer to an intradot Auger scattering mechanism. The usual electron-electron Auger scattering is excluded here because the total integrated luminescence is linear as shown in Figure 1b. The importance of Auger effects in QBs has been demonstrated by several authors [3,24–26] showing that this process can be very efficient even when only one electron-hole pair exists in the dot [9]. Since our QBs collection has a weak confinement potential along the wire axis (about 10 meV) the levels are close in energy and this mechanism should be very probable. Here for the resonant experiments we show that this intradot Auger scattering occurs very rapidly as soon as two pairs are created initially in an excited state of a dot. The excess energy between excited and fundamental state is given to one of the carriers which is ejected to a continuum wire state while the other one relaxes to the lowest level of the dot. The ejected carrier is then captured by a neighbouring dot and relaxes down to its ground state. Experimentally, the total intensity of emission remains constant during the process which means that Auger scattering occurs on both electrons and holes, so we still get radiative recombination. This is schematically drawn for one type of carriers in Figure 4.

Time-resolved μPL has been detected on peaks A and B with a resonant excitation on the first excited state of box A. Figure 5 represents the time evolution of the μPL intensity recorded on peak A as a function of optical excitation density. When the excitation power is increased, the shape of the time-resolved spectra is modified. As in the non-resonant case, a saturation of the fundamental level of the box occurs. We also observe that as the power is increased the rise time of the temporal spectra becomes shorter (Fig. 5c). To analyze these results a three level model has been considered in the resonant experiments: the first excited

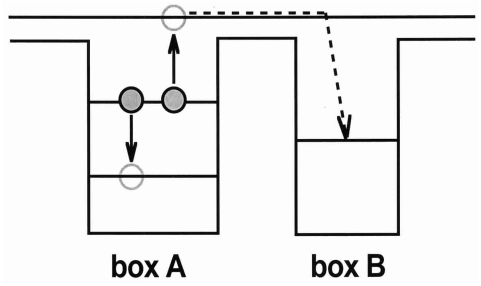

Fig. 4. Schematic representation of the intradot Auger scattering mechanism (solid arrows) for one type of carriers. The initial and final states of the scattering process are represented in full and open circles respectively. After the scattering, one of the carriers relaxes to a neighbouring box, for example box B (dashed arrow).

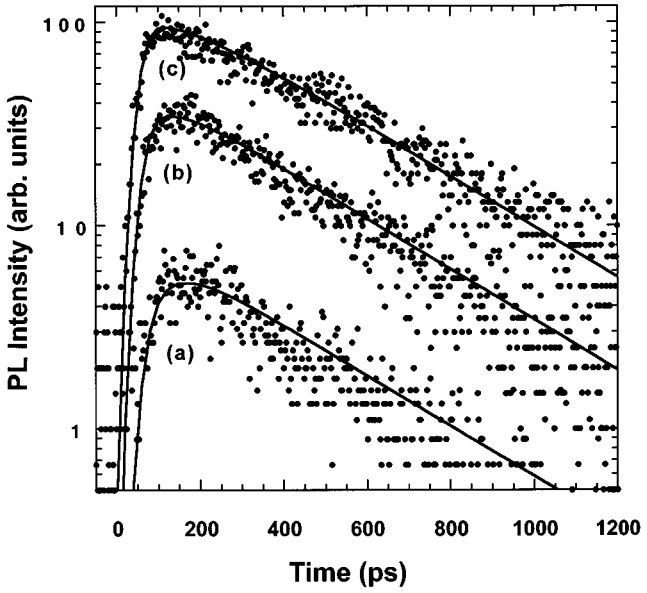

Fig. 5. Resonant time-resolved μPL experiments recorded on peak A at different optical excitation densities: (a) 3 W/cm^2; (b) 30 W/cm^2; (c) 300 W/cm^2. The dots correspond to the experimental data and the solid line to the calculated curve using equations (2–4) convoluted by the temporal response of the detection system.

and the lowest lying levels of a same QB and the $|0\rangle$ ground state. This allows an easier interpretation of the relaxation and saturation processes. We assume that the PL intensity is proportional to the mean number of pairs in the fundamental level of the box. The excited state is occupied by a population $f(t)$ having a Gaussian shape with a full width at half maximum of 5 ps equal to the laser pulse width. The calculated curves are convoluted by the temporal response of the detection system. The state filling effects have been taken into account with a level degeneracy assumed to be equal to 2 because of the QB asymmetry. The Auger relaxation mechanism corresponds to the factor $n_2(t)^2/T$ in the equations below with T being the intradot Auger scattering time. The final state of this scattering consists of one pair in the lowest lying level of QB A while the other one has been ejected in a continuum wire state before being captured by another box. The time evolution of the level populations in QB A can be calculated by resolving the system of differential equations given by the following expressions:

$$\frac{\mathrm{d}n_2(t)}{\mathrm{d}t} = -\frac{n_2(t)}{\tau_2} - \frac{n_2(t)}{\tau}\frac{(2-n_1(t))}{2} - \frac{n_2(t)^2}{T}\frac{(2-n_1(t))}{2} + f(t)\frac{2-n_2(t)}{2} \quad (2)$$

$$\frac{\mathrm{d}n_1(t)}{\mathrm{d}t} = -\frac{n_1(t)}{\tau_1} + \frac{n_2(t)}{\tau}\frac{(2-n_1(t))}{2} + \frac{1}{2}\frac{n_2(t)^2}{T}\frac{(2-n_1(t))}{2} \quad (3)$$

$$f(t) = P\frac{1}{\sqrt{2\pi}\Delta t}\mathrm{e}^{-\frac{1}{2}\left(\frac{t-t_0}{\Delta t}\right)^2}, \quad (4)$$

$n_1(t)$ ($n_2(t)$) are the populations of the lowest lying (first excited) electron-hole pair state in QB A. P is a factor proportional to the excitation power and Δt is the width of the laser pulse. τ_1 and τ_2 are the radiative lifetimes of the lowest lying and first excited states respectively, and τ is the relaxation time through emission of LA-phonons. τ and τ_1 are calculated from the low excitation power spectrum (Fig. 5a) where the non-linear effects (Auger scattering and state filling) are not present. The calculation gives 70 ps for the relaxation time and 350 ps for the radiative lifetime. τ_2 is assumed to be of the same order of magnitude as τ_1 but the value of τ_2 has no influence on the calculation of Auger relaxation. The two other curves represented in Figure 5b, c can be calculated with the same values of τ and τ_1 with the population $f(t)$ of the excited state being in the ratio of the experimental values of the excitation intensity. The value calculated for the factor P is 0.16, 1.6 and 16 for the excitation densities used in the experiments, *i.e.* 3, 30 and 300 W/cm^2 respectively. This power factor would correspond to the number of electron-hole pairs created in a system without saturation effects. This result is also consistent with the simple expression that we discussed in Section 2. The above model describes quite well the temporal behaviour of the μPL of a single QB when the pump power is increased as we can see in Figure 5. In order to better understand the influence of each term (Auger, state filling and linear) appearing in equations (2) and (3) a comparison between them is represented in Figure 6 for the same set of parameters. If we take only into account the state filling we obtain a smooth plateau which does not fit the experimental situation at high optical excitation intensity. The Auger factor is responsible for a modification of the shape of the temporal curve and for the shift towards shorter delays of the curve maximum at high excitation power. Indeed the rise time of the curve in Figure 5c is shorter than in 5a or b, because of the presence of Auger scattering which creates a second channel for relaxation. The intradot Auger scattering time T, is found to be equal to 20 ps. This value

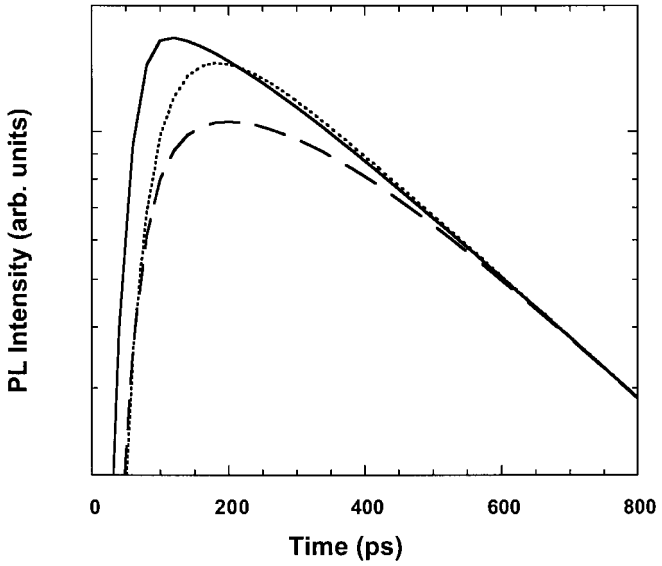

Fig. 6. Calculated temporal evolution of the population occupying the lowest QB level by neglecting all the non-linear effects (dashed line), by including only the level state filling (dotted line) and by including Auger relaxation with state filling (solid line). The model used is described by equations (2) and (3) in the text. The curves have been normalized in order to have the same decay at long delays.

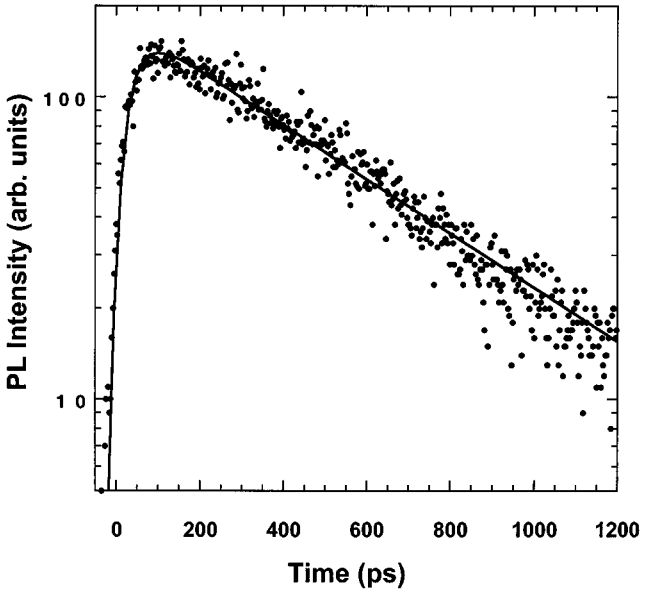

Fig. 7. Resonant time-resolved μPL recorded on peak B at 300 W/cm^2 optical excitation density. The excitation energy is resonant with the excited level of box A. The dots correspond to the experimental data and the solid line to the calculated curve using equation (1) convoluted by the temporal response of the detection system.

is in the limit of our temporal resolution but it remains slightly faster than the relaxation time through emission of LA-phonons. This result shows that Auger scattering is a very efficient process that occurs as soon as two carriers occupy the first excited level of one dot.

Time-resolved μPL experiments *versus* the excitation power have also been performed on peak B with an excitation energy resonant with the first excited level of box A. Emission from box B starts to appear above 100 W/cm^2 excitation density when the lowest level of box A becomes saturated. We have studied QB B in the linear regime before saturation. Then just by using equation (1) we can fit the experimental temporal decay shown in Figure 7. The rise time which corresponds actually to capture and relaxation time in box B is 30 ps and the radiative lifetime is equal to 500 ps. The difference in the radiative lifetimes and relaxation times between box A and B is due to the different size of the boxes [6]. We observe that the rise time is very fast which means that both capture and relaxation in QB B are very fast processes and of the same order of magnitude.

4 Conclusion

Power effects have been investigated in a single QB by studying the time evolution of the μPL intensity. Non-resonant experiments show that state filling of the lowest lying level occurs. By performing resonant experiments in a single box, we observe that as the optical excitation density increases, a very efficient carrier-carrier scattering occurs due to Auger processes. This mechanism allows to empty a saturated QB and fill another neighbouring one through a transfer of carriers (electrons and holes) that keeps in this way the total emission intensity constant. The high efficiency of this scattering mechanism, of the order of 20 ps, explains the lack of emission from excited states which is a less efficient process. It must also be related to the weak potential barrier of our specific QB collection so the excited levels are close to the potential edge leading to a propitious situation for carriers to escape from the boxes.

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