Experimental determination of Auger capture coefficients in self-assembled quantum dots

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(Received 25 October 1999; revised manuscript received 8 March 2000)

Time-resolved studies of the wetting layer photoluminescence are combined with state-filling spectroscopy of the quantum dot and wetting layer emission to obtain carrier transfer rates from the wetting layer to the quantum dot states. With this method, capture rates can be measured for constant carrier concentrations in the wetting layer. The results show that the capture efficiency increases with the carrier concentration in the wetting layer, indicating the important role of Auger processes in the capture dynamics. In the analysis, the concept of capture cross section per unit time is introduced, and this is used to determine the single dot Auger capture coefficient in self-assembled dots. The value obtained can in principle be used as an input to model carrier capture in all self-assembled dot devices with similar dot layers.

Capture rates in self-assembled quantum dots (QD) have been investigated in the past using time-resolved spectroscopy of the QD emission.[1,2,3,4,5,6] In these experiments, a short laser pulse impinges on the sample at $t=0$, and a carrier population is created in the wetting layer (WL) or barrier material. A rise of the time-dependent QD photoluminescence (PL) intensity is then observed as the carrier population in the QD grows, followed by a decay due to the exhaustion of the carrier supply (the barrier population is not replenished between pulses) via radiative and nonradiative recombination. The number of carriers in the system is therefore constantly varying, and the capture times obtained cannot be correlated with a well-defined carrier concentration. It is generally accepted that carrier capture from the WL states to the QD states can be mediated either through multiphonon emission or via carrier-carrier Coulomb scattering (Auger relaxation). The former process is modeled using a capture rate independent of carrier concentration, while the latter one involves a capture time which is to first order linear with the carrier concentration. In this paper, state-filling spectroscopy of QD's is combined with time-resolved PL of the WL to obtain capture rates in QD's as a function of carrier concentration. Thus, the relative importance of multiphonon and Auger processes can be compared quantitatively.

The paper is organized as follows: in the theory section the concept of temporal cross section (TCS) for carrier capture in QD's is introduced, and an expression for this parameter is obtained that depends on measurable quantities. In the results and analysis section, we show how this parameter can be expressed as the sum of an Auger and a phonon contribution to explain the observed results.

I. THEORY

Following the work by Uskov *et al.*[7] the case where carriers relax from the WL states to the QD states is considered. Moreover, for simplicity, correlated electron and hole capture in the dots is assumed,[8] meaning that for dynamic modeling purposes, carriers can be considered to be excitons. Also, the capture rate of carriers is expected to scale with the areal coverage of the QD ensemble,[9] and accordingly we introduce the single dot temporal cross section for carrier capture:

$$\sigma = s\Gamma_c, \tag{1}$$

where s is the surface range over which the QD's can capture carriers efficiently and Γ_c is the capture rate of carriers when they are within that range. With the above definition, the average QD ensemble carrier capture rate is given by $\gamma_c = A_D s\Gamma_c = \sigma A_D$. The rate equation for the QD and WL carrier populations can be written as

$$\frac{dN_{wl}}{dt} = -\gamma^r_{wl}N_{wl} - \sigma A_D N_{wl} + G, \tag{2a}$$

$$\frac{dN_{QD}}{dt} = +\sigma A_D N_{wl} - \gamma^r_{QD}N_{QD}, \tag{2b}$$

where N_{QD} and N_{wl} are the QD and WL carrier densities, respectively, γ^r_{QD} and γ^r_{wl} are the QD and WL carrier recombination rates, A_D is the dot areal density, and G is the carrier generation function. In the steady state, the total carrier loss rate is equal to the carrier generation rate,

$$G = \gamma^r_{QD}N_{QD} + \gamma^r_{wl}N_{wl}. \tag{3}$$

Furthermore, in a PL experiment, the QD and WL luminescence intensities can be expressed as

$$I_{QD} = \gamma^{rad}_{QD}N_{QD}, \tag{4a}$$

$$I_{wl} = \gamma^{rad}_{wl}N_{wl}, \tag{4b}$$

where I_{QD} and I_{wl} are the QD and WL emission intensities, respectively, and γ^{rad}_{QD} and γ^{rad}_{wl} are the QD and WL average radiative recombination rates, respectively. Solving Eqs. (2), (3), and (4) for σ yields

$$\sigma = \frac{1}{A_D}\left(\frac{I_{QD}}{I_{wl}}\frac{\gamma^r_{QD}}{\gamma^{rad}_{QD}}\gamma^{rad}_{wl}\right) \approx \frac{1}{A_D}\left(\frac{I_{QD}}{I_{wl}}\gamma^{rad}_{wl}\right), \qquad (5)$$

where the right-hand-side expression is obtained by neglecting nonradiative recombination in the dots ($\gamma^{rad}_{QD} \approx \gamma^r_{QD}$). This is justified since for typical background concentrations of nonradiative centers of the order of $10^{14}-10^{15}\,\mathrm{cm}^{-3}$, and a QD size of 20 mm at the base, less than one QD in a hundred thousand will contain a nonradiative center. Thus, Eq. (5) shows that the TCS is proportional to the ratio of QD to WL emission intensity which can be readily measured from a PL experiment. The dot areal density can be obtained from structural analysis, such as TEM, AFM, etc., and γ^{rad}_{wl} can be obtained from time-resolved photoluminescence (TRPL) analysis of the wetting layer emission when no QD's are present.

It should be noted that in the above analysis the carrier concentrations are considered invariant in position or time. The laser power will therefore be kept constant throughout the experiment, which will ensure a time invariant carrier concentration. Also, in order to ensure a spatially uniform carrier concentration, a filtering method already described elsewhere is used.[10]

II. EXPERIMENTS AND ANALYSIS

The samples were grown by molecular beam epitaxy and consisted of a single layer of InAs imbedded in GaAs barriers.[10] In the case of the QD sample, 1.9 ML of InAs were deposited, while in the WL sample only 1.8 ML of InAs were deposited, thus keeping the amount of material deposited just below the critical thickness for dot formation. A plan view TEM picture of the QD layer in the 1.9 ML sample was obtained, and the measured dot density is $A_D = 9.5 \pm 2.5\,\mu\mathrm{m}^{-2}$.[11] The uncertainty on the value is rather high due to the poor statistics involved with low QD densities.

Figure 1(a) shows the PL spectra of the WL sample at low temperature. The energy of the PL peak at 1.43 eV matches very well with the emission energy of the WL observed in other samples where QD emission is also present. The PL line in Fig. 1(a) is also narrower than typical QD emission lines, and we therefore attribute this peak to WL emission. The absence of luminescence from QD's indicates that a negligible density of QD's have formed on the surface of the WL, and therefore this sample is suitable to measure γ^{rad}_{wl}.

Figure 1(b) shows the time-correlated trace obtained at 4 K when the sample is excited using 10 ps pulses at 640 nm, with 0.43 fJ per pulse. The detection energy is set at the peak of the WL emission. One can see that a single exponential decay over 1.5 orders of magnitude is obtained. Using a single exponential fit in that region yields a carrier lifetime of 334 ps. Temperature dependent PL measurements show that the integrated emission intensity remains constant until the onset of thermionic emission above the barrier,[12] thus indicating a negligible contribution of nonradiative centers to the recombination process at low temperatures. The radiative recombination rate is therefore set equal to the inverse of the measured lifetime: $\gamma^{rad}_{wl} = 2.99 \times 10^9\,\mathrm{s}^{-1}$.

The ratio of QD to WL emission in the QD sample is obtained as a function of excitation power by varying the intensity of the 514.5 nm line of an Argon laser used as the excitation source. The inset of Fig. 2 presents an example of a spectrum obtained at 5 K for a uniform excitation of 220 W/cm^2. Below 1.4 eV, one observes the emission from five bound states, while the wetting layer emission is observed at 1.43 eV. The PL spectra were fitted using one Gaussian function per observed peak, and the resulting fit is shown as a solid line in the inset. Five Gaussians were used for the dot transitions between 1.0 and 1.4 eV, and summing the corresponding fitted area under the curves yields $I_{QD}(220\,\mathrm{mW})$. For the WL peak at 1.43 eV, only one Gaussian curve was necessary to give a satisfying fit, and the single Gaussian area directly yields $I_{wl}(220\,\mathrm{mW})$. This procedure is repeated at each power density, and Fig. 2 shows the resulting evolution of I_{QD} and I_{wl} with excitation intensity. Using Eq. (5) and the results of Fig. 2, one can now obtain the evolution of

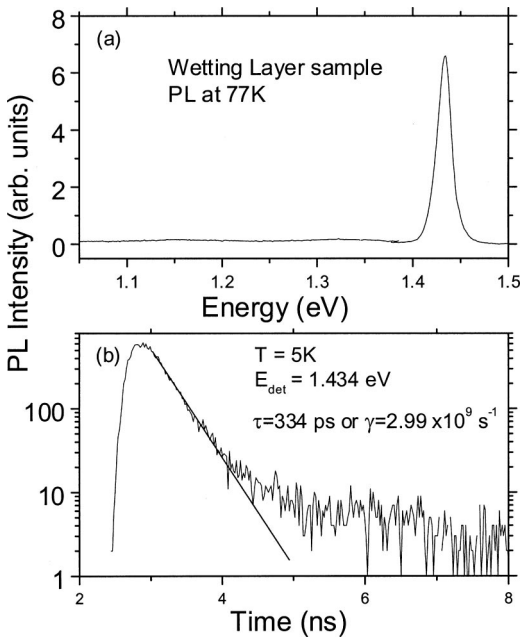

FIG. 1. (a) Photoluminescence of wetting layer sample at 77 K. (b) Time-resolved photoluminescence trace of the peak emission at 5 K.

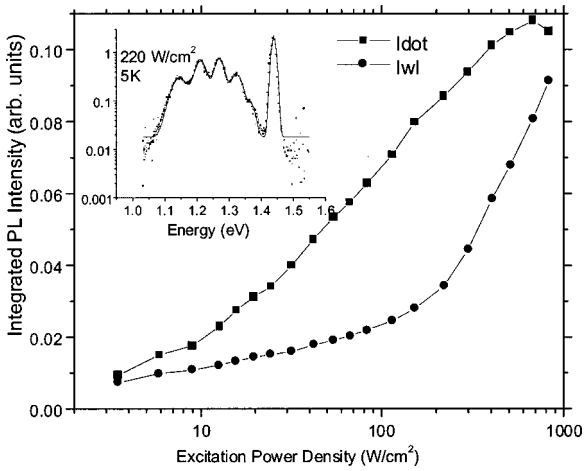

FIG. 2. Evolution of the integrated intensity of QD and WL emission at 5 K. The inset shows an example of one of the spectra obtained with the corresponding multiple Gaussian fit.

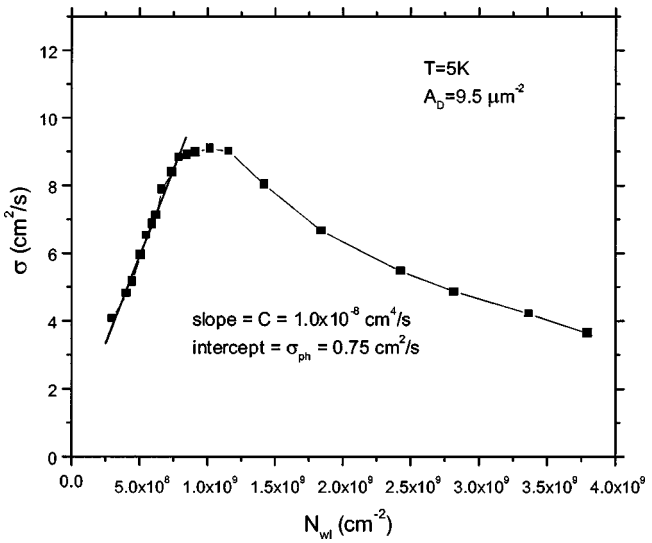

FIG. 3. Evolution of the TCS with wetting layer carrier density at 5 K. The linear fit at low carrier densities is shown along with values of slope and intercept obtained, where the slope corresponds to the Auger capture coefficient.

the TCS as a function of pump power, and the resulting curve is shown in Fig. 3. The values of WL carrier density in Fig. 3 were obtained from the laser intensity using a two step procedure. First, using Eq. (4) and assuming that at high intensities (830 W/cm^2) all QD states are occupied, one obtains that $N_{QD}(830\,\mathrm{W/cm^2}) = 30*9.5\times10^8\,\mathrm{cm^{-2}}$. The WL carrier density at that intensity is then obtained by taking the ratio of Eqs. (4a) and (4b) and isolating for N_{wl}. The ratio of intensities is obtained from Fig. 2, γ_{wl}^{rad} is obtained from Fig. 1(b), and γ_{QD}^{rad} is obtained from a weighted average of the excited states recombination rates as found in a previous paper.[10] Next, all other values can be obtained by considering that the WL carrier density is proportional to the WL emission intensity. In Fig. 3, one can see that for $N_{wl}<8\times10^8\,\mathrm{cm^{-2}}$ the capture rate increases linearly with WL carrier concentration, while a reversal of this effect is observed for $N_{wl}>1.5\times10^9\,\mathrm{cm^{-2}}$. The decrease in the TCS at higher excitation is attributed to the saturation of the occupation probability of the QD states. At high average carrier populations a significant proportion of the QD's have all of their bound states filled, effectively shutting off these capture centers which decreases the average TCS of the QD's. The behavior at low carrier populations is more intriguing. In order to discuss this, the TCS is broken into two contributions, one that describes Auger processes and one that describes capture via phonon emission. In this context, Eq. (2a) is written:

$$\frac{dN_{wl}}{dt} = -\gamma_{wl}^{r}N_{wl} - CN_{wl}^2A_D - \sigma_{ph}N_{wl}A_D + G, \qquad (6)$$

where C is the Auger capture coefficient for an individual dot, and σ_{ph} is the phononic contribution to the total TCS. It should be noted that since the above equation neglects thermionic emission, it is therefore valid only at low temperatures. By comparison of 4 and 6,

$$\sigma = CN_{wl} + \sigma_{ph}. \qquad (7)$$

Thus, for low carrier densities ($N_{wl}<8\times10^8\,\mathrm{cm^{-2}}$), the slope of σ vs N_{wl} gives the Auger capture coefficient and the intercept the phonon contribution to the TCS. For this regime of low carrier densities, one expects carrier properties to be dominated by excitons, thus supporting our assumption of correlated electron-hole capture. From the linear fit in Fig. 3, the following values are obtained at 5 K: $C\approx(1.2\pm0.6)\times10^{-8}\,\mathrm{cm^4/s}$, and $\sigma_{ph}\approx(0.8\pm0.2)\,\mathrm{cm^2/s}$. The uncertainties are obtained by considering lower bound and upper bound values for A_D. Other errors may come from uncertainty on γ_{QD}^{rad}. As discussed above, nonradiative recombination in the QD's or WL does not contribute significantly to the error on the values obtained. From the above values for C and σ_{ph}, the average capture rate for the dot ensemble at carrier densities of $10^9\,\mathrm{cm^{-2}}$ and higher is dominated by Auger relaxation. Indeed, for a carrier density of $10^9\,\mathrm{cm^{-2}}$ the Auger capture rate is $A_DCN_{wl}=1.0\times10^{10}\,\mathrm{s^{-1}}$ compared to $A_D\sigma_{ph}=0.08\times10^{10}\,\mathrm{s^{-1}}$ for the phonon contribution. These rates are slow compared to most of the previously published results, and we believe this is due to the very low dot density present in our sample compared to previous work[1,2,3,4] where dot densities were at least a factor of 5 higher.

It should also be mentioned that the above value obtained for C is a few orders of magnitude higher than the theoretical estimation made by Uskov *et al.*[7] This discrepancy should be the object of further investigations.

In conclusion, we have introduced the concept of temporal cross section for carrier capture in QD's. Considering correlated carrier capture in the dots, a method was outlined to obtain single dot Auger and phonon capture cross sections. In theory these parameters are independent of the QD areal density, and if this is verified, the obtained parameters should prove very useful in QD device design, such as the QD laser. Our results show that even at moderate carrier densities ($10^9\,\mathrm{cm^{-2}}$) the carrier capture rate from WL states to QD states is dominated by Auger relaxation.

We would like to acknowledge Dr. J. P. McCaffrey and Dr. Sylvain Charbonneau for their help in the course of this work. One of us (K.H.) acknowledges NSERC for their support.

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