Dynamics of Excitation Transfer Inside InAs/GaAs Quantum Dot System

K.P. Korona[a,*], A. Babiński[a], S. Raymond[b]
and Z. Wasilewski[b]

[a]Institute of Experimental Physics, Warsaw University

Hoża 69, 00-681 Warsaw, Poland

[b]Institute for Microstructural Sciences, NRC Canada, Ottawa, Canada

We present time-resolved photoluminescence investigations of InAs/GaAs structures containing quantum dots with the ground state at 1.43 eV. State filling effect and a Pauli blocking effect were clearly observed. These effects significantly influenced dynamics of excitation transfer from upper to lower state inside a dot leading to non-exponential dynamics. Numerical model based on nonlinear rate equations was proposed. The model described well the experimental data providing values of: lifetime of the ground state 0.53 ± 0.03 ns, lifetime of excited state (when the ground state is full) 1.1 ± 0.2 ns, and internal relaxation time (when the ground state is empty) 0.07 ± 0.01 ns.

PACS numbers: 78.47.+p, 72.20.Jv, 78.67.Hc, 78.55.Cr

1. Introduction

It has been shown that in InGaAs/GaAs structure excitons can be localized by potential fluctuations in the wetting layer (WL). This is equivalent to the formation of quasi-zero-dimensional semiconductor structures — "natural" quantum dots (QDs) in the WL [1, 2]. Due to the small size of the dots, the density-of--states (DOS) in this system is a series of δ functions representing discrete electron states. Low DOS in QDs is the reason of the state filling effect observed in QDs even when only a few carriers were excited per dot [3].

Emission from individual QD can be investigated using micro--photoluminescence (μPL). Also, time resolved μPL measurements of InP/Ga$_{0.5}$In$_{0.5}$P/GaAs QDs at low excitation density were reported [4]. It showed exponential decays with constant recombination rates. At higher power, numerous

*corresponding author; e-mail: kkorona@fuw.edu.pl

excited states in QDs are filled. Due to Pauli's exclusion principle, their recombination depends on the occupation of lower states. The transfer of excitation is a statistical process, observed well on a large number of QDs. Here we present time-resolved macro-PL investigations of InAs/GaAs structures containing QDs. We clearly observe a state filling effect at the high excitation condition and the Pauli blocking effect that significantly influences dynamics of excitation transfer from upper to lower state inside a dot. Unfortunately, at higher excitation conditions (necessary to observe state filling effect and Pauli blocking effects) number of PL lines increases so the PL emission forms a broad band [1].

2. Experiment

The sample studied in this work was grown using molecular beam epitaxy and it comprised both self-organized InAs/GaAs QDs and natural QDs formed in the InAs WL. The latter QDs were investigated in this work. Number of investigated dots was limited using mesa-patterning. Excitonic emission from the single dots formed in the wetting layer accompanying the InAs/GaAs self-assembled QDs has been confirmed by micro-PL measurements [1].

Time-resolved photoluminescence (TRPL) was measured at 10 K. Laser spot was of the order of 1000 μm^2. Frequency-doubled output pulses (150 fs) of a Ti:sapphire laser had been used for photoexcitation of free electron–hole pairs in the sample. A monochromator and a streak camera were used to obtain the spectral and temporal distribution of the PL.

3. Results and discussion

The low-temperature PL signal (10 K) comprises sharp excitonic lines in the GaAs barrier at 1.515 eV [5], the GaAs free-electron to carbon acceptor transition at 1.495 eV [6]. Photoluminescence from natural InAs/GaAs QDs was observed in the range of 1.43–1.46 eV (see Fig. 1).

At low excitation density, the emission from the natural QDs forms a peak with a width of 13 meV centered at energy of about 1.43 eV (QD1 band). It had recombination rate $r_{QD} = 1.8(1)$ ns^{-1}. In this case, the occupation of the dots was low, so there were single electrons and holes per dots, occupying only the ground states of the dots. Increase in the excitation power density led to saturation of the QD1 band and to the creation of a high-energy PL band at 1.44 eV (QD2 band).

At short time, before 1 ns, the QD2 PL was much stronger than the QD1 PL, and covered it. However, excitons in the QD2 decayed much faster than the QD1, so for time longer than 1 ns QD1 luminescence could be clearly observed (see Fig. 2A).

The both PL bands were broad, FWHM = 14 meV and 10 meV for QD1 and QD2, respectively. Moreover, they were closely spaced (10 meV), so procedure of careful fitting of few theoretical curves was used to separate signals from QD1 and QD2 bands. The QD1 and QD2 bands were fitted with Gaussian peaks (see
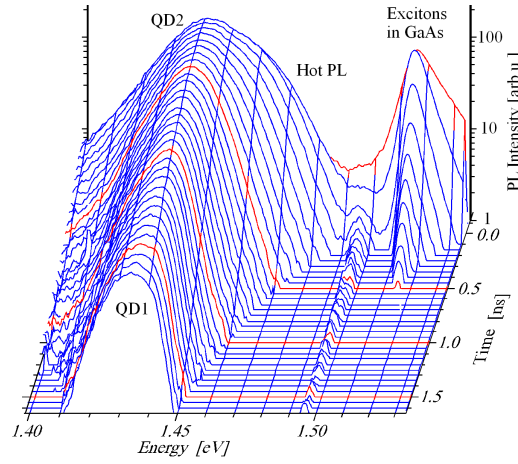
Fig. 1. TRPL spectrum of InAs/GaAs structure with QDs at $T = 11$ K. The QDs photoluminescence gives a band at 1.43–1.46 eV. Its decay rate is from 0.5 ns^{-1} to 2 ns^{-1}, depending on energy.
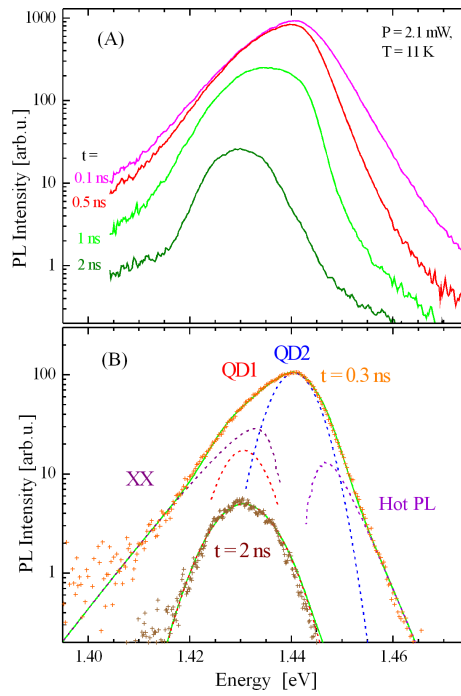

Fig. 2. PL spectra at different times after excitation. (A) As measured, (B) fitted with theoretical curves (crosses — data, lines — fitted curves). It can be seen that PL consists of at least two Gaussian peaks with different time dependencies.

Fig. 2B). Moreover, hot luminescence (probably related to free excitons in the WL) was fitted with theoretical curve taking into account kinetic energy given by thermal distribution (Eq. (2) of Ref. [7]). Similar curve, but symmetrically reflected (Eq. (3) of Ref. [7]) was used to fit low-energy wing of the PL band.

The PL transients obtained by the fitting procedure are presented in Fig. 3A and C. It is visible that the QD2 band decayed significantly faster than the QD1 and its decays were non-exponential. Initially the QD2 decay was slow (decay rate about 1 ns^{-1}). Then, after a few hundreds of picoseconds the decay accelerated to the rate about 20 ns^{-1}. This type of decay is characteristic of an initial ground--state filling effect. When ground states (of density N_0) are filled, one has to take into account the Pauli blocking, and nonlinear terms $dn/dt = \beta_1 n(N_0 - n_0) + \ldots$ become important in the rate equations. When the ground states were occupied, the higher states decay slowly, since $N_0 - n_0$ factor was small. Later, when the ground states became empty, the decay of the higher states accelerated.
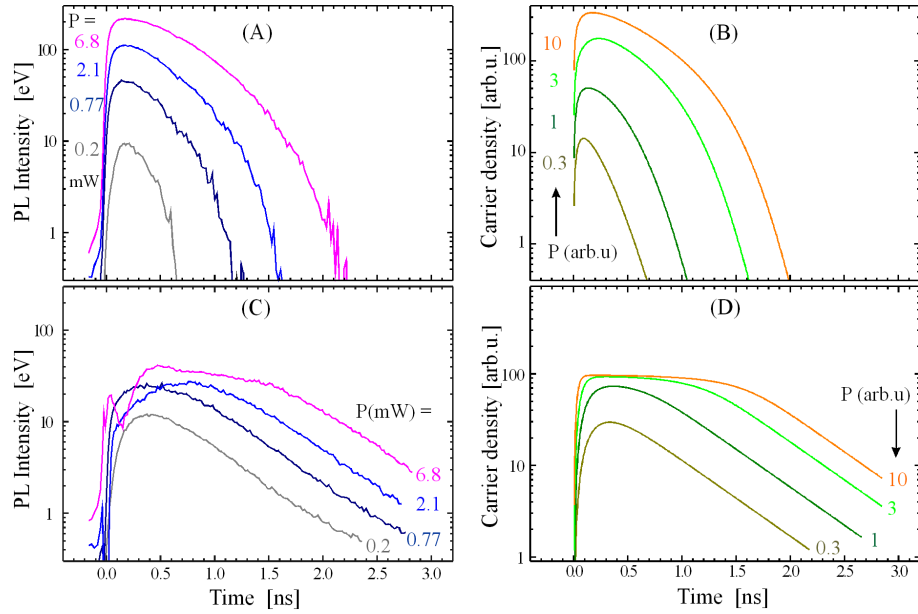

Fig. 3. PL transients of InAs/GaAs QDs. Left (A), (C) — curves obtained from analysis of experimental data, right (B), (D) — calculated curves. Up (A), (B) — excited state QD2, down (C), (D) — the ground state QD1.

In order to describe the observed transients, three groups of states were taken into account: (1) the ground states of quantum dots (QD1), (2) excited states in QDs (QD2) and (3) initial level, that was a source of carriers (probably GaAs). Evolution of carrier concentrations $n_1(t)$, $n_2(t)$, and $n_3(t)$ can be calculated from rate equations

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = -r_{01}n_1 - d_{21}n_1(N_2 - n_2) + d_{12}n_2(N_1 - n_1), \tag{1}$$

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = -r_{02}n_2 - d_{12}n_2(N_1 - n_1) - d_{32}n_2 N_3 + d_{21}n_1(N_2 - n_2)$$

$$+d_{23}n_3(N_2 - n_2), \tag{2}$$

$$\frac{\mathrm{d}n_3}{\mathrm{d}t} = -r_{03}n_3 - d_{23}n_3(N_2 - n_2) + d_{32}n_2 N_3, \tag{3}$$

where N_1, N_2, and N_3 are concentrations of states of groups 1, 2, and 3. Recombination rates r_{0i} are sums of radiative and external non-radiative recombination rates of carriers. Transfer rates d_{ij} describe transfer between states j and i. Number of carriers transferred from state j to state i is proportional to concentration at the initial state $n_j(t)$ and to the concentration of empty final states $N_i - n_i(t)$. The latter factor expresses Pauli's exclusion principle. This factor makes the rate equations nonlinear.

At low excitation conditions, one can assume $N_i \gg n_i(t)$ and approximate $N_i - n_i(t) = N_i$. In this case, analytical solution, $n_i(t) = \sum_k A_{ki} \exp(-r_{ki}t)$, exists. This sum is in fact the discrete Laplace transform of a spectral function A_k. Therefore, an inverse Laplace transform [8] can be used to find decay rates r_{ki}. If excitons flow from group of states 3 to group of states 2, solution for states 3 is mono-exponential (decay rate $r_{13} = r_{03} + d_{23}N_2$) and for states 2 is two-exponential: $n_2(t) = -A_{12}\exp(-r_{13}t) + A_{22}\exp(-r_{22}t)$, where $r_{22} = r_{02} + d_{12}N_1$. The first part of this equation has negative amplitude, so it is increasing function of time, and therefore it represents rising part of the transient. It is interesting to notice that the rising rate is equal to the decay rate of the source state. Therefore, in this case, the inverse Laplace transform (ILT) of the source signal will have a peak at r_{13}, and the ILT of the state signal will have a depression at r_{13} and a peak at r_{22}.

In fact, the ILT of the TRPL spectrum showed a few interesting features. A peak at $E = 1.515$ eV, $r = 6-10$ ns^{-1} was related to recombination of excitons in the GaAs barrier. The QDs photoluminescence had a wide peak at 1.43–1.46 eV, 0.5–2 ns^{-1} and a depression at 1.45 eV, 2–20 ns^{-1} that represented rising of the QDs PL. The similarity of the rising rate of the QDs PL and the decay rate of the barriers PL showed that the WL were mainly fed by excitons from the GaAs barrier.

In the case of low densities of states (N_2 and N_1) in QDs, it was easy to reach high excitation conditions, under which n_i is comparable to N_i. Therefore the best way of measured data analysis was to solve Eqs. (1)–(3) numerically. The calculated curves were plotted in Fig. 3B and D. The obtained numerically curves reproduce very well the experimental data for different excitation densities. In the case of the ground state (QD1) saturation at higher excitation power was observed. After saturation had ended, QD1 decayed with rate 1.9 ± 0.1 ns^{-1} which means that its lifetime was 0.53 ± 0.03 ns. In the case of the excited state,

increase in lifetimes with power was found, both in experiment and in the model. When lower state was full (saturated), the decay of excited state (QD2) was slow (lifetime 1.1 ± 0.2 ns) and after saturation had ended, it accelerated. The internal relaxation rate (QD2 to empty QD1) deduced from fitting of numerical model was r_{int} $(= d_{12}N_1) = 14 \pm 2$ ns^{-1}.

4. Conclusions

TRPL of large number of QDs investigations showed that state filling effect and a Pauli blocking effect were important for QDs dynamics at higher excitations.

Numerical model based on nonlinear rate equations was proposed. The model described well the experimental data showing that when the ground states were fully occupied (saturated), the higher states decayed slowly, mainly by radiative recombination ($r_{02} = 0.9 \pm 0.1$ ns^{-1}). Later, when the ground states became empty (due to recombination), the decay of the higher states accelerated because carriers relaxed non-radiatively to the ground states.

Acknowledgments

References

[1] A. Babiński, S. Raymond, Z. Wasilewski, J. Lapointe, M. Potemski, *Acta Phys. Pol. A* **105**, 547 (2004).

[2] R. Oulton, J.J. Finley, A.I. Tartakovskii, D.J. Mowbray, M.S. Skolnick, M. Hopkinson, A. Vasanelli, R. Ferreira, G. Bastard, *Phys. Rev. B* **68**, 235301 (2003).

[3] S. Grosse, J.H.H. Sandmann, G. von Plessen, J. Feldmann, H. Lipsanen, M. Sopanen, J. Tulkki, J. Ahopelto, *Phys. Rev. B* **55**, 4473 (1997).

[4] V. Zwiller, M.-E. Pistol, D. Hessman, R. Cederstrom, W. Seifert, L. Samuelson, *Phys. Rev. B* **59**, 5021 (1999).

[5] J.A. Rossi, C.M. Wolfe, G.E. Stillman, J.O. Dimmock, *Solid State Commun.* **8**, 2021 (1970).

[6] B.J. Skromme, T.S. Low, T.J. Roth, G.E. Stillmann, J.K. Kennedy, J.K. Abrokwah, *J. Electron. Mater.* **12**, 433 (1983).

[7] K.P. Korona, *Phys. Rev. B* **65**, 235312 (2002).

[8] R.E. Bellman, R.E. Kalba, J. Locket, *Numerical Inversion of the Laplace Transform*, Elsevier, New York 1966.