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Spectroscopy of high-density assemblage of InAs/GaAs quantum dots

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Abstract. High-density arrays of InAs/GaAs quantum dots (QDs) have been studied by means of steady-state and time-resolved photoluminescence (PL) within a wide range of laser power. The ground state tunnelling between neighbouring QDs is suggested to be an important relaxation channel defining the PL spectral shape both at very low excitation density and at the temperature elevation. This channel becomes blocked when the QDs ground states population reaches the saturation at high excitation density.

1. Introduction

High-density self-assembled QDs are of interest for potential optoelectronic applications. However recently produced high-density QDs arrays possess essentially strong scattering both geometrical and relaxation parameters. Under the QDs areal density D_a comparatively high, $D_a \geq 10^{10}$ cm^{-2}, the behaviour of PL response, both steady state and dynamic, differs substantially from that of low-density QDs ensembles [1]. This difference reflects the intrinsic peculiarities of carrier transfer in the system of dense QDs arrays, which are still beyond the clarity.

Here we report a detailed investigation of carrier transfer in dense InAs/GaAs QDs arrays. A range of carrier densities in which inter-dot carrier transfer saturates is identified. This transfer involves transitions from higher lying ground states of small size QDs into lower-lying states of larger QDs.

2. Experimental details

GaAs superlattice structures containing thin layers of InAs were grown on GaAs (001) substrates by gas-source molecular beam epitaxy. Chosen growth conditions allowed to get the dense QDs arrays possessing nevertheless comparatively broad distribution by sizes. The AFM images of a sample with d_{InAs} = 2.46 ML and with substrate temperature T_G = 420 ^{0}C during the InAs growth display a co-existence of, at least, four QDs families

with the average base length of b = 6, 8, 12, and 14 nm and the QDs density ratio of approximately 4:8:4:1, respectively. Transient PL measurements were performed with a self mode-locking Ti:sapphire laser (732 nm, 80 fs, 82 MHz) with excitation density between 10^9 and 10^{13} photons/(pulse×cm^2), allowing predominant excitation of the InAs QDs through the GaAs matrix. The steady-state PL was excited by the 514.5 nm line of Ar^+ laser with excitation densities in the range of 1 mW/cm^2 to 20 W/cm^2.

3. Results and Discussion

The low-temperature (T = 10 K) steady-state PL spectrum of the sample with d_{InAs} = 2.46 ML is depicted in Fig.1, representing a broad spectral band (FWHM ≈ 200 meV). The line-shape analysis shows that the PL signal is well reproduced by a convolution of four Gaussian-shaped peaks. Comparing the size distribution and areal densities derived from the AFM measurement and the results of PL analysis we assume that the peaks at 1.329 eV, 1293 eV, 1.224 eV, and 1.156 eV could be originated from the QDs families with the average bases b = 6, 8, 12, and 14 nm, respectively. The temperature elevation up to

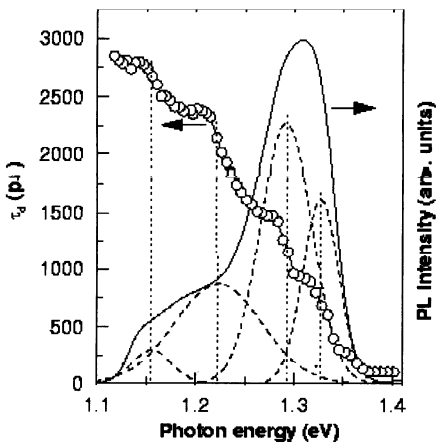

Figure 1. PL spectrum and staircase spectral dependence of $\tau_d(E)$ for InAs/GaAs QD's sample, T = 10 K, excitation density 2×10^{10} photons/(pulse×cm^2). Gaussians are shown by dash lines.

T = 240 K results in the complete decay of high-energy bands, and the low-energy band persists only. Thus the high temperature spectrum becomes effectively shrunken, being totally shifted towards the red side [2]. Time-resolved PL measurements were performed for different detection energies within the broad PL spectrum (Fig.1) and in a wide range of excitation intensities. Up to the excitation densities of 10^{11} photons/(pulse×cm^2), the PL emission at the different detection energies rises within the time resolution of our experiment of 10 ps. It displays a mono-exponential decay characterised by a decay time τ_d. In Fig.1, τ_d -data determined from a least-square fit of the transients for the excitation density 2×10^{10} photons/(pulse×cm^2) are summarised. The pronounced step-like behaviour of τ_d spectral dependence is observed. The τ_d step architecture completely reflects the PL band structure. The parts of the τ_d spectrum quickest slope correspond to the maximums of the PL components. As a function of sample temperature, the steps in τ_d shift by the

same energy as the cw PL spectrum, providing independent evidence for the correlation between transient and steady-state PL features. This correlation is maintained up to the room temperature. The τ_d dependence allows revealing the various QD mode contributions even if these contributions could hardly be resolved spectroscopically in the conventional PL spectrum, thus providing a powerful experimental technique for analysis of very dense arrays of coupled QDs.

The τ_d spectral dependence is consistent with the model of inter-dot carrier transfer that considers the ground state relaxation in a system of coupled QDs. In our model, carriers in the ground state of a QD can relax by radiative recombination giving rise to PL and – in addition - carriers populating the ground states of smaller QDs can be transferred into the levels of larger QDs being even lower in energy. For weak excitation density of the QD ensemble and under the assumption that the carrier relaxation rate from an energy level in a QD is proportional to the number of vacant lower energy levels in adjacent QDs, the rate equation for a particular quantum level E_i can be written as

$$\frac{dn_i}{dt} = -\frac{n_i}{\tau_0^i} - \sum_{j<i} \frac{n_i(N_j - n_j)D_j}{\tau_t^{ij}} + \sum_{i<j} \frac{n_j(N_i - n_i)D_i}{\tau_t^{ji}}. \qquad (1)$$

where τ_0^i is the total ground-state recombination lifetime in the i-th ground state, τ_t^{ij} is the inter-dot carrier transfer time between E_i and E_j states, D_j is the density of the (final) E_j states, N_j is the number of available QD ground states in the j-th dot distribution. If one neglects the non-linear terms in Eqn.(1) it can be solved analytically for the particular D_j distribution. The PL decay time τ_d is determined as $\tau_d = -n_i(t)/(dn_i(t)/dt)$. In fact, the proposed rate equations model of Eqn.(1) holds for a dense QD system independent of the number of modes of its size distribution. The presence of more than one distinct dot size distribution within a large QD ensemble can be taken into account simply by introducing additional Gaussians D_j into Eqn.(1), resulting in several step-like variations of τ_d with emission energy. The calculated transients $n_2(t)$ for the lower energy level of a

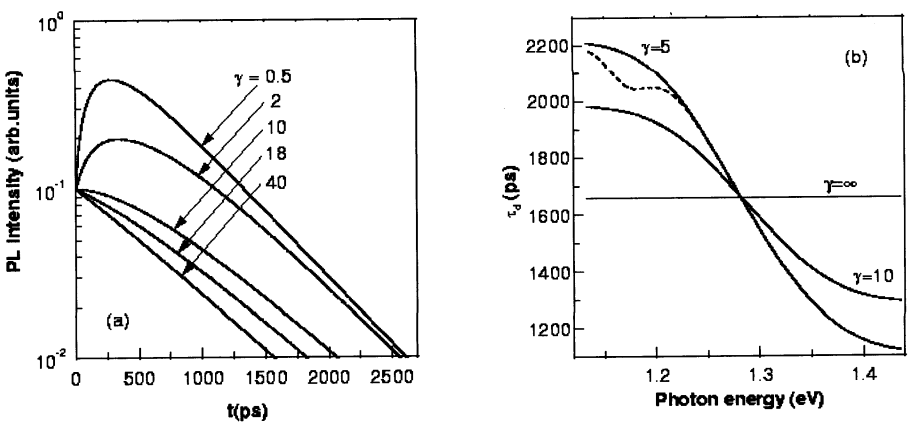

Figure 2. (a) Calculated transients and (b) spectral dependence of $\tau_d(E)$ for different for different ratios $\gamma = \tau/\tau_0$.

simplified QD model system with only two distinct dot sizes are shown in Fig.2a. The parameter $\gamma = \tau_t/\tau_0$ is a measure for the inter-dot coupling and describes the additional population of the lower level 2 by carriers from level 1. Small γ-values indicate strong coupling, i.e. efficient population of level 2. The spectral dependence of τ_d for different γ values is depicted for a single Gaussian (solid curves) and two Gaussians (dashed line) in Fig.2b. The spectral steps are clearly seen.

A realistic description of carrier dynamics in dense QD arrays has to include the case of saturation, i.e. situations in which the number of non-equilibrium carriers n_i reaches the number of available QD ground states N_j in the j-th dot distribution (see Eqn. (1)). The equations of set (1) are nonlinear differential equations, and do not have an analytical solution. Therefore the set was solved numerically with the natural assumption of a single time τ_0 for the recombination lifetime in the i-th ground state and a single time τ_t for the interdot carrier transfer. Applying this rate equation model to the actual experimental situation we can explain the details of QD kinetics. Indeed, the carrier transfer from smaller to larger QDs results in a delayed build-up of population in the latter, a behaviour reflected in the PL kinetics. With increasing excitation flux, saturation of population in such low-energy states occurs, leading to a faster rise of PL. This prediction of the rate equation models is in full agreement with our experimental results. For strong excitation, saturation leads to much faster population increase in the emitting QDs and a subsequent monoexponential decay of PL intensity. Applying the simple model for the low-density case and $\tau_0 = 300$ ps we derive a value of $\gamma = \tau_t/\tau_0 = 1.3$. It is clear that saturation of QD ground states could result in a lineshape variation of the cw PL at the smallest power densities due to saturation of inter-dot carrier transfer channel. Thorough PL study at the extremely low excitation density (less than 1 W/cm^2) displays such modification. It is observed the increase of the low-energy part of PL spectrum with the excitation density that reaches the saturation. We find that this saturation occurs already at 1W/cm^2, because the further power increase does not change the lineshape of this spectral region. The high energy part of the PL spectrum grows with the intensity of excitation, giving hint for the possible contribution of excited states which is expected to develop into pronounced structure under the very high excitation densities. It is also clear that the decrease of τ_d observed experimentally at high pumping levels can arise from many-body effects, which become more important in a dense QD system due to the fact that carriers can readily scatter out of the saturated states. Within the empirical model this depopulation of the lowest energy states could be treated by adding two similar nonlinear terms to the rate equations (1) but with opposite signs and a different, shorter scattering time $\tau_d(P_{exc})$ (which becomes a function of laser power for high excitation). This would lead to an effective decrease in the PL decay time, as experimentally observed in the spectral range of the QD emission regardless of QD size. This has little to do with the recombination times for higher energy states (or of smaller dots) being shorter.

In summary, we reported a study of steady-state and transient PL of dense InAs/GaAs quantum dot arrays. Our data are consistent with the model of carrier transfer from small to large QDs within the ensemble directly influencing the PL kinetics.

References

[1] Mazur Yu I, Tomm J W, Petrov V, Tarasov G G, Kissel H, Walther C, Zhuchenko Z Ya and Masselink W T 2001 Appl. Phys. Lett. 78 3214-6

[2] Tarasov G G, Mazur Yu I, Zhuchenko Z Ya, Maassdorf A, Nickel D, Tomm J W, Kissel H, Walther C and Masselink W T 2000 J. Appl. Phys. 88 7162-70