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Photoluminescence decay time measurements from self-organized InAs/GaAs quantum dots grown on misoriented substrates

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Introduction

Lately a lot of papers were devoted to the investigations of laser heterostructures based on quantum dots (QDs). This interest is based on the fact that QDs provide a zero-dimensional system, with 3D carrier confinement resulting in atomic-like, discrete electronic eigenstates. The δ -like state density preconditions the low threshold current densities of laser diodes [1, 2] and higher values of T_0 [3] compared to existing semiconductor lasers. But the threshold current density of the "classical" QD laser diodes based on the single layer InAs heterostructures is far from predicted values [4, 5]. One of the possible obstacles to achieving the low threshold current density is low internal quantum efficiency in predicted by the theory excitation level range (10–100 A/cm²). The laser diode, which was made during this work, has a threshold current 110 A/cm². The aim of this work is to study the mechanism limiting the quantum efficiency.

1. Experimental details

For our studies we have grown two samples the only one difference between them was that one was a laser diode sample and another one for the photoluminescence (PL) investigations. Samples were grown by the molecular beam epitaxy (MBE) in Stransky–Krastanow growth mode. To increase the density of QD arrays we used InAs QD single layer array grown on GaAs(001) substrate misoriented by 4° to the [010] direction [6]. Using this method in our samples we achieved a relatively high density of QDs ($\approx 10^{11}$ cm⁻²). The interruption time between the end of QD growth and the start of the GaAs layer overgrowth was 10 s. The thickness of the InAs covering was 2.9 monolayer. The InAs QD growth temperature was 470°C and the III/V element flux ratio was 2. The InAs QD array was confined by GaAs barriers (20 nm) which were surrounded by AlAs/GaAs superlattices and by Al_{0.7}Ga_{0.3}As cladding layers and practically repeats the corresponding part of the laser heterostructure reported earlier [5]. Steady-state photoluminescence (PL) spectra were obtained using Ar⁺-ion laser (2.41 eV) and Ti-sapphire tunable laser. The laser beam was focused onto the sample installed in a He cryostat. The PL signal was dispersed by a 2m grating monochromator and detected by a cooled Ge photodiode.

Time-resolved PL was measured at sub-100 fs pulse excitation (repetition rate of 82 MHz) from a Ti-sapphire laser at 1.68 eV. A grating monochromator in combination with a streak camera having an infrared-enhanced photocathode allowed spectral discrimination and detection. The total system response was under 15 ps.

2. Results and discussion

Figure 1 shows the steady-state PL spectra for different excitation power densities equivalent to electron-hole pair density up to $6.7 \times 10^{13} \text{ cm}^{-3}$. These densities are sufficient to cover the range of average carrier densities captured by QDs. All excitation power densities for convenience in comparison with laser diodes excitation power densities are given in current values (A/cm^2).

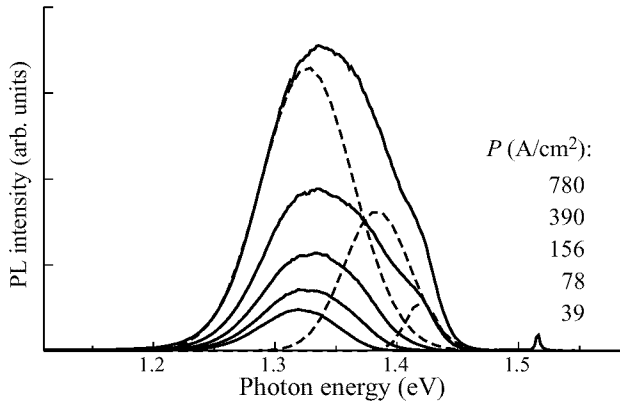


Fig. 1. Steady-state PL spectra at different excitation density of InAs QDs grown on GaAs substrate with 4 degree misorientation. $T = 5 \text{ K}$.

The PL spectra recorded at the low excitation densities (Fig. 1) is described by a single gaussian with peak energy of 1.33 eV. This can be ascribed to the recombination of electron-hole pairs in ground state. As the excitation power density increases, two new gaussians appear in the high energy part of PL spectrum. Their peak energy is 1.38 and 1.42 eV (gaussians shown in Fig. 1 as dashed curves). First one (1.38 eV) is attributed to the recombination of electron-hole pairs in excited state [7] and the second one (1.42 eV) — to the recombination of electron-hole pairs in wetting layer [8].

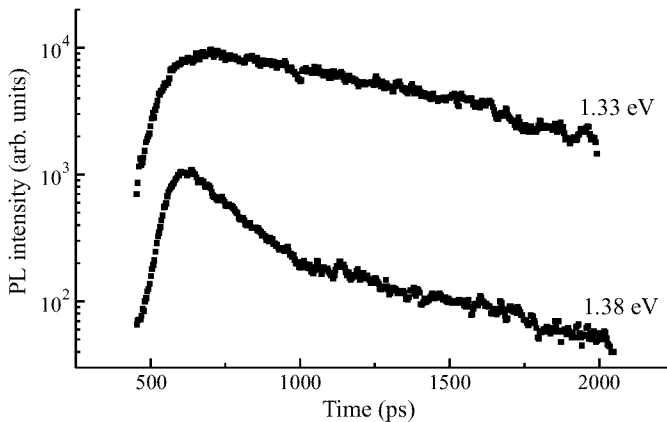


Fig. 2. Time-resolved PL from the ground state (upper curve) and first excited state (lower curve) of InAs QDs at excitation density 7.8 A/cm^2 and $T = 10 \text{ K}$.

Figure 2 shows the PL time decay curves measured for the ground and first excited states of the electron-hole pair (1.33 and 1.38 eV respectively). The PL decay of the ground state is described by one exponent with characteristic time ($t \approx 3.1$ ns). The PL decay curve of the excited state is substantially different from the same of the ground state. Its shape has the biexponential feature: the initial fast component and the slower second component. Their estimated characteristic times are 210 ps and 700 ps respectively. Such kind of the behavior is explained by two processes: fast — free relaxation of the carriers into the unoccupied ground state and slow-radiative recombination from the excited state. An increase of the excitation density (Fig. 3) leads to the fast component disappearance. In accordance with [8] this behavior can be explained by the blocking of relaxation to the fully occupied ground state. As seen from Fig. 3 this effect took place at the excitation density range 20–80 A/cm². The full carrier relaxation scheme is very complex. It should comprise a lot of processes, such as sequential, nonsequential, multiphonon scattering and Auger recombination [9] etc. Taking this fact into account we use more simple model — we are examining only concentration of carriers on the energy levels without considering all above mentioned mechanisms. Later we will show that such approach is a sufficient approximation to explain the addressed problem.

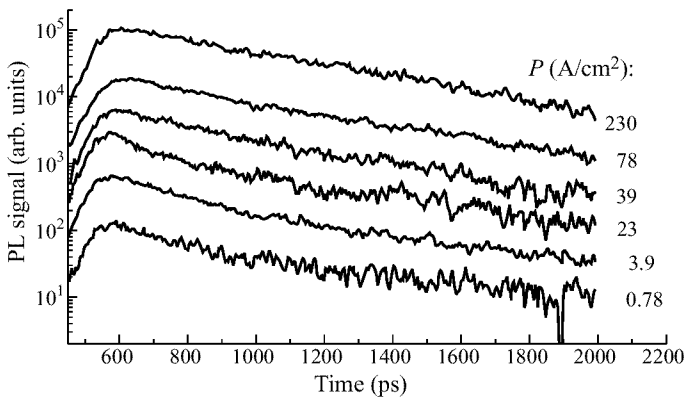


Fig. 3. Time-resolved PL from the excited state of InAs QDs at different excitation density and $T = 10$ K.

Figure 4 shows the ground state, excited state and wetting layer PL intensity divided by the excitation density ($\zeta_{G,S}$, $\zeta_{E,S}$ and $\zeta_{W,L}$ correspondingly) versus excitation density. As seen from the figure, $\zeta_{G,S}$ has three parts — the increase, saturation and decrease. $\zeta_{G,S}$ achieves the maximal value at excitation density ≈ 10 A/cm² and rapidly decreases after 40 A/cm². The value of $\zeta_{E,S}$ starts to increase after 20 A/cm² — directly before the decrease of $\zeta_{G,S}$. Both of these facts clearly demonstrate the filling of the ground state and the blocking of carrier free relaxation from the excited state. In this case the radiative recombination rate on the ground state is determined by the density of QDs. An increase in excitation density leads to the decrease of quantum yield of PL ground state. The estimated excitation density for the ground state filling is 20–30 A/cm². This value of excitation density is very close to the theoretically predicted threshold current of QD laser diodes. In practically developing QD laser need for compensation of inevitably nascent losses can demand such values of current density at which value of quantum yield starts to decrease. It is obvious that in laser diodes everyone have to our results show that the

ground state filling effect should be taken into account in QD laser diodes design.

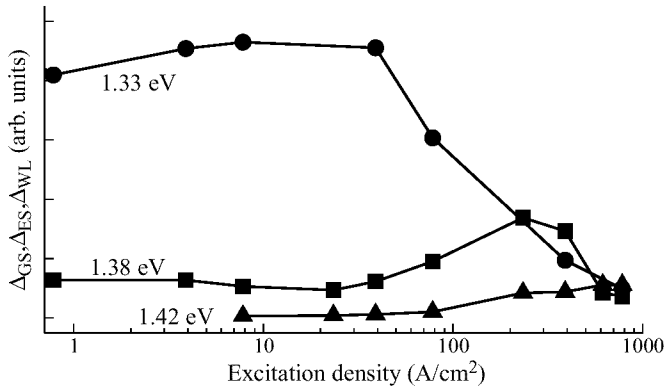


Fig. 4. PL intensity of the ground state, excited state and wetting layer divided by excitation power density versus the excitation density at $T = 10$ K.

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