Time-resolved photoluminescence measurements of InAs self-assembled quantum dots grown on misorientated substrates

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Time-resolved photoluminescence decay measurements have been performed on samples with varying-sized self-assembled InAs/GaAs quantum dot ensembles, formed by substrate misorientation. Ground-state radiative recombination lifetimes from 0.8 to 5.3 ns in the incident power density range of 0.05–3400 W cm^{-2} at a temperature of 77 K have been obtained. It was found that a reduction of the quantum dot size led to a corresponding reduction of the radiative lifetime. The evident biexponential decay was obtained for the ground state emission of the quantum dot array, with the slower second component attributed to a carrier recapturing process. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1637962]

Self-assembled quantum dots (QDs) are currently of much interest due to the predicted advantages caused by δ-like energy state density, which can be used in a wide range of potential applications.[1–4] The radiative lifetime is a critically important QD parameter for the optimization of the heterostructure laser design. Calculations show[5] that the radiative lifetime depends on the QD size. Although some investigations have already been performed on this dependence,[6,7] we report evidence from a set of samples grown at the same time under exactly the same conditions. Typically, the preparation of the InAs QD arrays of different average size requires a change in the growth conditions that can greatly influence material quality and makes the comparison of photoluminescence (PL) characteristics of different QD arrays somewhat less certain. It has been shown previously that the application of misoriented substrates can permit the growth of QD arrays with different average sizes under exactly the same growth conditions.[8] In this work we describe radiative lifetime measurements of QD ensembles with a different QD size determined by substrate misorientation alone.

InAs QD single layer arrays were grown simultaneously using the Stransky–Krastanow method by molecular beam epitaxy (MBE) on an exactly oriented GaAs (001) substrate (sample A) and on substrates intentionally misoriented by 2°, 4°, and 6° to the [010] direction (samples B, C, and D, respectively). To form the regular structure of the terraces on the misoriented substrates the samples were annealed at 650 °C. The average thickness of the InAs layer was 2.9 monolayers. 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) surrounded by 250 nm thick AlAs/GaAs-graded band-gap superlattices and by $Al_{0.7}Ga_{0.3}As$ cladding layers. The structure was completed with a GaAs cap layer.

The size of the QDs and their density were calculated from atomic force microscopy.[8] It was found that in the 0°–4° range of the misorientation angle the lateral size of the QDs decreased from approximately 22 to 12 nm while the QD density increased from 3×10^{10} to 7×10^{10} cm^{-2}. The size and density of QDs in the sample with the 6° misorientation angle (sample D) were found to be in close agreement with the characteristics of the sample with a 4° angle (sample C).

Steady-state PL measurements were performed using an Ar^+ ion laser ($\lambda=514.5$ nm) with an excitation power density of 500 W cm^{-2}. The time-resolved photoluminescence (TRPL) measurements employed the technique of time-correlated single photon counting[9] in conjunction with Si single photon avalanche diodes since such an approach offered TRPL measurements of the required sensitivity and time resolution. TRPL measurements of the QD ground state (GS) emission were performed in a wide incident power density range (0.05–3400 W cm^{-2}) on samples A, B, C, and D using the microscope-based system described previously.[10] The optical excitation was provided by a passively Q-switched picosecond AlGaAs diode laser[11] emitting at a wavelength of 746 nm and focused to a 50 μm diameter spot. Measurements were performed over a 150 ns time window. The detector used was a Perkin–Elmer SPCM module that led to an overall instrumental response time of ~500 ps (full width at half-maximum). Narrow bandpass filters (few nanometers width) were used to spectrally discriminate the luminescence signal.

The PL spectra of all samples presented in Fig. 1 clearly show the blue shift of the PL peaks with decreasing QD size, as the misorientation angle increases. In samples C and D,

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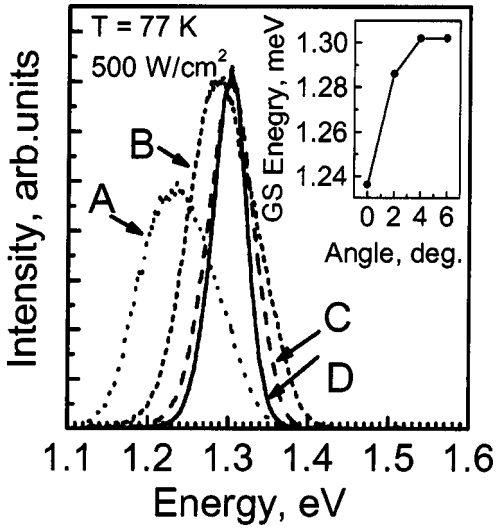

FIG. 1. Steady-state PL spectra at 77 K. The inset shows the position of the GS emission maxima.

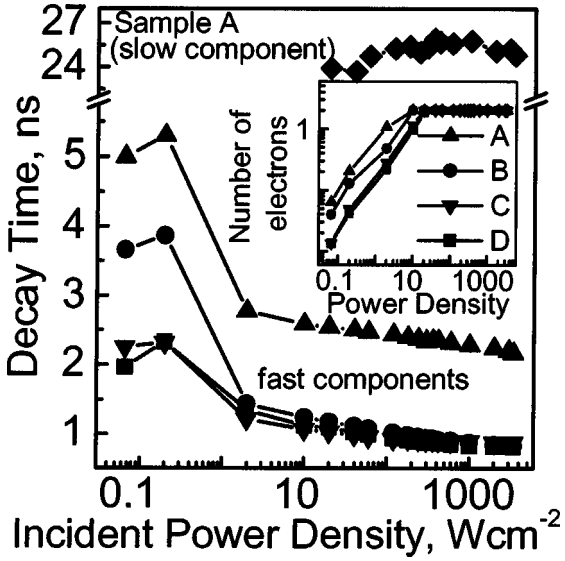

FIG. 3. Decay time versus incident power density at a sample temperature of 77 K. The inset shows average number of electrons per QD versus incident power density.

the average QD size was very similar and the steady-state PL had the same peak wavelength. The PL emission in sample D was spectrally narrower because of the more homogeneous size distribution.[8] The position of the maxima of the GS emission in each sample is presented in the inset in Fig. 1. All TRPL measurements were done at the detection wavelength corresponding to the PL maxima.

Figure 2 shows the results of the TRPL measurement from samples A and C at the excitation power density 180 $\mathrm{W\,cm^{-2}}$ and the temperature of 77 K. The inset shows the TRPL trace from sample B under the same conditions. For sample A the shape of the decay has a biexponential nature with two characteristic times—a fast component (of a few ns time constant) and a significantly slower one (of 10's ns). For the other samples only the faster characteristic time was observed at this temperature. According to additional experiments the internal quantum efficiency of all four samples was found to be approximately 100% in the excitation range 0–100 $\mathrm{W\,cm^{-2}}$ at a temperature of 77 K. Hence, we can attribute the faster decay time to the QD radiative lifetime (τ_{Rad}).[12,13]

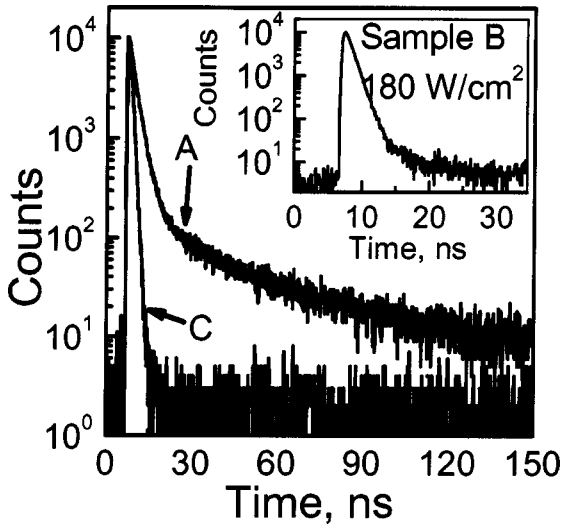

FIG. 2. TRPL traces of samples A and C at 77 K. Inset shows the TRPL trace of sample B under the same conditions.

The radiative lifetime dependence on incident power density for each sample is shown in Fig. 3. These values are consistent with previously reported measurements.[6,7,14] The dependence of the radiative lifetime on incident power density can be described in three parts: (1) τ_{Rad} remains constant at low excitation power densities; (2) τ_{Rad} decreases rapidly at power densities of $\sim 2\ \mathrm{W\,cm^{-2}}$; and (3) τ_{Rad} decreases slightly with increasing power density up to approximately 2 ns (for sample A) and 0.8 ns (for samples B, C, and D). This behavior can be explained by the filling processes of the QD's. The large characteristic time of stage (1) corresponds to an average of one or less electron–hole pairs per QD. The rapid decrease in τ_{Rad} in stage (2) corresponds to a full occupation of the GS, which leads to an increase of the electron–hole wave function overlap as well as to a decrease of the lifetime with the increasing carrier concentration inside the dot. In stage (3), the decrease of the lifetime is caused by the filling of the excited states and adjacent regions [wetting layer (WL) and barriers].

To demonstrate our assumptions we have estimated the average number of electrons in the GS of the QD array (**n**) using the following equation:

$$n = \frac{P \cdot \tau_{\mathrm{Rad}}(P)}{\rho \cdot e}, \tag{1}$$

where P is the excitation power density, ρ is the surface density of QDs and e is the charge of the electron. The results of our estimations for samples A, B, C, and D are shown in the inset of Fig. 3. These results show that, for each sample, the GS is fully occupied in the range of stage (3), where τ_{Rad} is almost constant.

Some previous experimental evidence has shown that the decay time reduces with increasing QD size.[7,15] However, in this work (summarized in Fig. 3), the decay time reduces with decreasing QD size. This can perhaps be explained by the theoretical work of Bimberg *et al.* [16] where it

was shown that the overlap integral could decrease at both extremes of the QD dimensions. For large QD diameters, the piezoelectric effect leads to the electron and hole wave functions expanding in different directions and becoming more elongated, hence reducing the overlap integral. The overlap integral can also decrease at small QD sizes when the confined electron level disappears. This argument means that there should be an optimum size of QD, providing a maximum in the overlap integral and a corresponding minimum in the radiative lifetime. According to the model described in Ref. 16, the piezoelectric effect plays a more significant role in the samples used in these experiments.

Figure 3 also shows the slow decay time for sample A for an incident power density range from 3400 to 10 $W\,cm^{-2}$. Over this range the lifetime was found to be constant at approximately 25 ns. It is important to notice that the second decay constant for the QDs' GS emission of such a long duration has not been observed previously, to the best of our knowledge. We were able to observe and study this decay constant due to the high sensitivity and flexibility of the TRPL setup, which permitted both high signal-to-noise ratios and long measurement windows (~150 ns is shown). The time constant of this decay is several times greater than the characteristic times of basic decay processes, such as radiative[14,17] and nonradiative recombination.[18] In addition, having performed a set of TRPL measurements, we concluded that carrier diffusion had a negligible effect on the decay times measured. Having ruled out these possibilities, the most likely mechanism for such a slow luminescence decay time is a re-capturing process in which the QDs exchange electrons via the WL. The probability of such a process should be greater for sample A because the average QD size in this sample is larger than in the other samples. This will lead to a larger number of excited states in the sample lying closer to the WL, thus improving the coupling to and from the WL. Furthermore, a likely reason why the slow decay was not observed in the misoriented samples is that the carrier exchange between QDs at 77 K is strongly suppressed due to the WL being broken at the terraces formed by the angle of the crystal planes with the substrate surface.

The radiative lifetimes of carriers corresponding to ground state of different size InAs/GaAs quantum dots arrays have been studied by the time-resolved photoluminescence technique at 77 K. The effect of GS filling on the radiative lifetime has been measured as being between 0.8 and 5.3 ns. It was found that the radiative lifetime reduced with a decreasing QD diameter. For the first time, a biexponential decay with a much slower second component—of time constant 25 ns—was observed for the ground state emission of the QD ensemble. This second component was attributed to carrier recapture via the wetting layer.

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