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Observation of the biexponential ground-state decay time behavior in InAs self-assembled quantum dots grown on misoriented substrates

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Biexponential behavior of the time-resolved photoluminescence decay from the ground state has been studied over a temperature range of 77–300 K on samples with varying sized self-assembled InAs/GaAs quantum dot ensembles controlled by substrate misorientation alone. The slower second decay component is considerably longer than the first one, and has been measured to be as long as 300 ns. This slow component is attributed to carrier recapturing and indirect radiative recombination processes. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1938000]

Self-assembled quantum dots (QDs) are currently of much interest due to predicted advantages caused by δ-like energy state density, which can be used in a wide range of potential applications.[1–4] In spite of the great progress in the investigation and fabrication of QD semiconductor devices[4] many different properties of QD arrays remain under investigation.

Typically, the preparation of InAs QD arrays of different average size requires a change in the growth conditions, which can greatly influence material quality and can place some uncertainty in the comparison of photoluminescence (PL) characteristics of different QD arrays. It has been shown previously that application of substrate misorientation to the [010] direction allows the growth of QD arrays with different average size, when grown under exactly the same growth conditions.[5] On such vicinal surfaces there appears to be a net of small terraces, separated by steps on all sides. QD growth on each terrace will occur primarily from the material deposited on the same terrace. By doing so, the density and size dispersion of QDs in the ensemble became controllable via the density and size dispersion of the terraces. In case of substrate misorientation, the wetting layer maintains its surface area but is broken at the terraces borders formed by the angle of the crystal planes with the substrate surface. In a previous letter[6] we performed lifetime measurements on InAs QD arrays with different QD sizes determined by substrate misorientation alone. It was found out that ground state decay time at a temperature of 77 K depended on the QD average size and population of the ground state. In the case of the exactly oriented sample a clear biexponential behavior of the ground state time resolved photoluminescence (TRPL) decays was obtained at 77 K. This slow decay time (SDT) for the exactly oriented sample had a time constant of ~25 ns and was attributed to carrier recapture via the wetting layer (WL) process. In that publication,[6] the SDT of the misoriented samples was not as clearly visible due to detectivity constraints, and was of a much longer duration than the exactly oriented sample. The goal of our present work is to investigate the origins of the SDT by performing TRPL measurements over a wide temperature range on the same samples used in our previous letter.[6]

InAs QD single layer arrays were grown using the Stransky–Krastanow method by molecular beam epitaxy on an exactly oriented GaAs (001) substrate (which we will call sample A) and on substrates intentionally misoriented by 2, 4, and 6 deg to the [010] direction (samples B, C, and D, respectively). All the samples were grown during the same epitaxial process. This method gave a unique possibility to investigate features mainly associated with uniformity and surface density of QDs. The average thickness of the deposited InAs layer was 2.9 monolayers. The InAs QD array was confined by GaAs barriers (20 nm) surrounded by AlAs/GaAs superlattices (250 nm) and by $Al_{0.7}Ga_{0.3}As$ cladding layers (200 nm). The size of the QDs and their densities were estimated from atomic force microscopy images.[5] It was found that in the 0–6 deg range of the misorientation angle the average lateral size of the QDs decreased from approximately 22 to 12 nm and at the same time the QD density increased from 3×10^{10} cm^{-2} to 7×10^{10} cm^{-2}. The increase of the terrace density with the misorientation angle led to the decreasing of the QD mean height from 34 A for the exactly oriented sample (sample A) to 20 A for the 6 deg misorientation (sample D).[5]

To perform TRPL measurements, the technique of time-correlated single photon counting[7] was used in conjunction with Si single photon avalanche diode SPAD detectors. TRPL measurements of the ground state (GS) QD emission were performed under nonresonant pulsed excitation (λ =746 nm) with the same incident power density of 3.4 kW cm^{-2} over a wide temperature range (77–300 K) on samples A, B, C, and D using the microscope-based system described previously.[8] Detailed investigations of SDT temperature dependence have shown that samples B, C, and D, grown on misoriented substrates, also have a biexponential feature, which becomes more evident at higher temperatures. Preliminary PL investigations of the samples[6] showed the blueshift of the PL peaks with the decreasing QD size, as the misorientation angle increases. All TRPL measurements were done at the detection wavelength corresponding to the PL maxima. Figure 1 shows the results of TRPL measurement of sample C at the temperatures of 100 and 200 K. It

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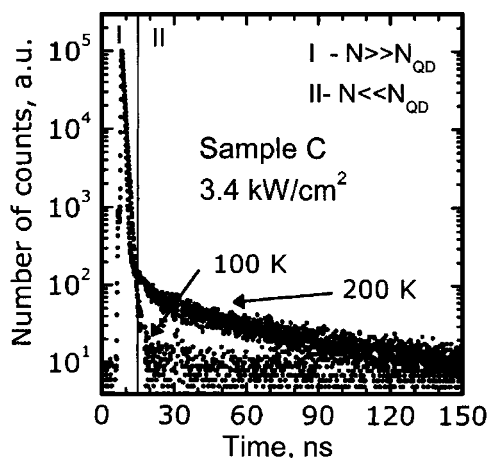

FIG. 1. TRPL decays from sample C at 100 and 200 K.

can be seen that the shape of the TRPL trace at 200 K has a clear biexponential feature with two characteristic times—a fast component and a significantly slower one, as it was observed previously for sample A at 77 K.[6] At lower temperatures the behavior of the TRPL trace is similar but the value of the slow component characteristic time becomes even longer and is closer to the noise floor of the detection process. The value of the fast decay time (FDT) for samples A, B, C, and D in the temperature range 77–300 K was found in the 0.5–2.2 ns range. It was shown previously that at 77 K the FDT represents the radiative lifetime for all samples.[6] With the temperature increasing the FDT slightly decreases. The values and behavior of FDT temperature dependence observed in this present work are consistent with previously published work on InAs/GaAs QDs.[9–11]

Figure 2 shows the SDT for samples A, B, C, and D in the temperature range 77–300 K. It can be seen from the figure that, as the temperature decreases from 300 to 77 K, the SDT increases from 35 to 170 ns for sample B and from 27 to about 300 ns for samples C and D. The value of the SDT for samples B, C, and D appears to remain constant at temperatures lower than 120 K. However, for sample A we do not observe such a behavior—the SDT remains at around 35 ns for the full temperature range. As discussed earlier the value of the SDT in the temperature range 77–100 K appears to be hundreds of nanoseconds for sample B, C, and D.

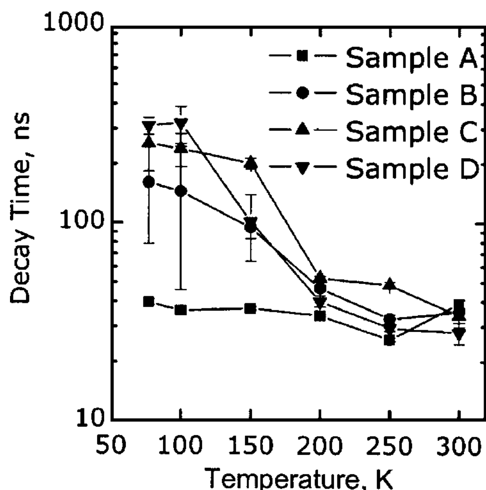

FIG. 2. SDT dependence vs temperature for samples A, B, C, and D.

Those values were estimated with a relatively large absolute error due to the closeness of the TRPL decay to the detection noise floor at longer time scales.

The observed values of the slow decay time for GS QD emission is much greater than the characteristic times of basic decay processes, such as radiative recombination[12,13] and nonradiative recombination.[14] It is unlikely to be related to the changing of the charge state of the QD deep levels due to the difference in temperature dependence of the SDT for samples A, B, C, and D, grown in the same epitaxial process. Separate experiments have shown that those large values of the SDT are not due to the influence of the diffusion process of carriers from weakly pumped areas. More specifically, at 77 K we measured the TRPL of sample A at two very different spot sizes of 50 and 5 μm diameters but with identical incident power densities (2 kW cm^{-2}). We could not identify any difference between the two TRPL traces, illustrating that the diffusion from weaker pumped areas is unlikely to be a major contributing factor in the origin of the slow decay time.[15]

Quantum dots grown on misoriented substrates are more homogeneous and have a higher surface density than those grown on the exactly oriented substrates, thus they should exhibit a higher tunneling rate, which is in contradiction with the obtained results. Therefore differences in quantum dot size homogeniety and density alone cannot be responsible for the larger SDT obtained for the misoriented samples.

In the experiments outlined in Fig. 2, the excitation pulse forms an initial carrier concentration, N, of $\sim 2\times 10^{11}$ cm^{-2}. Such a value is much higher than the concentration of QDs, N_{QD}, both for the exactly oriented and misoriented samples. In this case the QDs are filled uniformly, hence, the capturing time, τ_C, should be considerably less than the radiative recombination time, τ_R.[16] Thus the dominant process corresponding to part I of the decay curve (as denoted in Fig. 1) is direct radiative recombination, which has a characteristic decay time of approximately 1 ns for all samples. During the first few nanoseconds of the decay (corresponding to part I), the luminescence intensity reduces by a factor of 100, consequently the carrier concentration becomes less than 10^{10} cm^{-2}. Hence, the carrier concentration corresponding to part II of the decay curve (Fig. 1) is less than N_{QD}. Relaxation of the electrons and holes occurs independently, hence, the situation when carriers are distributed randomly could take place. Consequently, carriers get localized in different QDs and are spatially separated. In such a circumstance, the measured large values of the decay time are defined by the spatial separated carrier redistribution time with further direct or indirect radiative recombination. Thus at low carrier densities the indirect radiative recombination with a SDT considerably larger than the FDT starts to play a key role. In terms of the spherical QD approximation, the radiative recombination characteristic time (τ) of the electrons and holes localized in the neighboring QDs can be described as follows:[17]

$$1/\tau \cong 1/\tau_{\mathrm{R}} \cdot I_{\mathrm{eh}}, \quad (1)$$

where τ_R is the radiative recombination time of the electron and hole localized in the same QD, and I_{eh} is the overlap integral of the electron and hole wave functions localized in neighboring QDs.

For the misoriented samples the strong increase of the SDT at the low temperatures could be explained by the over-

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all WL potential not existing due to the WL being broken at the terraces formed by the angle of the crystal planes with the substrate surface. In this case electron transport in the plane of the QD array is possible only by thermal escape of the electrons from the QDs to the barriers and eventual recapture into the QDs. A simple analysis[18] shows that the behavior of the SDT temperature dependence in the temperature range 100–250 K correlates to the temperature dependence of the time of the electron thermal escape from the quantum well with a barrier height of 90 meV. This value of the barrier height is consistent with the analysis of the PL spectra from samples B, C, and D. The apparent lack of increase in the SDT at temperatures lower than 100 K could be explained by tunneling processes contributing to the electron redistribution process. The weak temperature dependence of the SDT for sample A is consistent with the presented model, since it was grown on exactly oriented substrate, and hence, an overall WL for the QDs will be present. Superposition of the WL potential with the potential of the QDs decreases the localization of electrons in the QDs and permits enhanced electron tunneling between the QDs. Hence, the SDT for each sample is determined by a sum of two processes with different characteristic times—one is the characteristic time of the tunneling process and the other is the characteristic time of the thermal escape of the carriers from the QDs to the barriers. The time of the tunneling process significantly differs for the exactly oriented sample and for the misoriented samples due to the reasons explained earlier.

In conclusion, the lifetimes of carriers corresponding to the ground state emission of different size InAs/GaAs quantum dot arrays grown in the same fabrication process has been studied by the time-resolved photoluminescence technique over the temperature range 77–300 K. Biexponential behavior of the TRPL trace of the ground state has been observed in both exactly oriented and misoriented samples. The value of the slower recombination lifetime was found to be in the 25–360 ns range. This evident slow component was attributed to electron recapturing and indirect radiative recombination processes. It was shown that the quantum dots remain charged after a relatively long time (hundreds of nanoseconds). This fact could be an important consideration in the design of future semiconductor devices based on quantum dot structures.

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