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# Photoluminescence from low temperature grown InAs/GaAs quantum dots

D. Sreenivasan,[a] J. E. M. Haverkort, T. J. Eijkemans, and R. Nötzel
*Department of Applied Physics, Eindhoven University of Technology, 5600MB Eindhoven, The Netherlands*

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The authors investigated a set of self-assembled InAs/GaAs quantum dots (QDs) formed by molecular beam epitaxy at low temperature (LT, 250 °C) and postgrowth annealing. A QD photoluminescence (PL) peak around 1.01 eV was observed. The PL efficiency quickly quenches between 6 and 40 K due to the tunneling out of the QD into traps within the GaAs barrier. The PL efficiency increases by a factor of 45–280 when exciting below the GaAs band gap, directly into the InAs QD layer. This points towards good optical quality QDs, which are embedded in a LT-GaAs barrier with a high trapping efficiency. © *2007 American Institute of Physics*.
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Ultrafast all-optical switching requires materials with a high bandwidth and thus a fast temporal response of the nonlinear absorption, a high absorption modulation and a low absorption loss when the material is saturated.[1,2] Stransky-Krastanow quantum dots (QDs) have enhanced optical nonlinearity due to their delta function like density of states, which results in a sharp absorption line with high peak absorption. In addition, the presence of a single electron-hole pair within a QD is able to completely bleach the absorption line, while two electron-hole pairs generate optical gain, indicating a large optical nonlinearity, which occurs already at very low pump power. A switching energy as low as 6 fJ in InAs/InP QDs embedded into a Mach-Zehnder switch[3] has been recently observed. Nakamura *et al.* have demonstrated QD absorption saturation at a pulse energy density as low as 13 $\mathrm{fJ}/\mu\mathrm{m}^2$, under resonant excitation.[4]

The time response of the optical nonlinearity is determined by the combination of the radiative and nonradiative decay and by carrier escape out of the QD. It has already been demonstrated that low temperature (LT) grown bulk GaAs as well as InGaAs/InAlAs quantum wells show ultrafast carrier recombination.[1,2,5] LT growth leads to the presence of excess As, which acts as a trapping center. The ultrafast response is thus attributed due to trapping of carriers into arsenic antisite defects.[1,2] In this letter we present a study of LT InAs/GaAs QDs grown using molecular beam epitaxy as an attempt to combine the large QD nonlinearity with an ultrafast response time.

We have investigated a set of InAs/GaAs QDs formed by LT growth and postgrowth annealing.[6,7] All the samples studied here were grown on a (100) GaAs substrate. The sample structure has a 500 nm thick GaAs buffer layer grown at 580 °C. The samples were subsequently cooled down to 250 °C for the growth of a 500 nm LT-GaAs bottom layer, followed by the deposition of 2 ML of InAs, which forms the QDs. The samples R207, R200, R198, R191, R181, and R152 were all grown at 250 °C. The samples further received different annealing treatments, which are detailed in Table I. In Refs. 6 and 7, it has been shown that well-developed InAs dots were formed in these samples after postgrowth annealing above 450 °C, as observed by atomic force microscopy. Finally all samples were capped with a 200 nm LT-GaAs capping layer. In R200 and R207, the structural quality of the LT-GaAs matrix grown on top of the quantum dots was improved[6,7] by inserting a 3 nm GaAs interlayer deposited at 480 °C directly on top of the LT-QDs. In addition, we also investigated sample R228, which consists of high temperature QDs grown at 480 °C in an identical LT-GaAs matrix as in the other samples.

Photoluminescence (PL) spectra were taken using 250 mW excitation power of a Ti:sapphire laser operating at 770 nm, providing a high excitation density of 10 $\mathrm{kW/cm^2}$. The PL was collected and further detected by a monochromator and a cooled InGaAs detector array. The PL spectra of all seven samples at 5 K are shown in Fig. 1.

It is observed that all the samples have a peak around 1.01 eV, which is slightly shifted for each sample, indicating that the peak is from the QDs and not from a deep center in the LT-GaAs. The spectra are also broadened and show a reproducible fine structure commonly observed in micro-PL, where emission from individual QDs can be observed. The fine structure probably reflects the QD size distribution showing statistical fluctuations, indicating that the density of radiative dots is low. Another possible reason for the fine structure is the Coulomb effect between electron-hole pairs within the QD and a nearby defect state.[8–10] The PL peaks are superimposed on a slowly rising background extending down to 0.78 eV, which is the edge of the detector responsivity. In contradiction with the QD photoluminescence, the rising background in the spectra is not quenched at elevated temperature, as shown in Fig. 2. The peak emission energy of the background could not be measured using the experimental setup used, making it difficult to assign the defect state accurately to any particular arsenic antisite defect.

Next the PL efficiency was investigated as a function of the measurement temperature. It is found that all the samples exhibit a very strong temperature dependence of the PL efficiency from the QDs around 1 eV. The PL from the QDs is completely quenched above a temperature of 40 K, which is also observed for the high temperature grown QDs (sample R228). The background PL is not quenched and even slightly increases with temperature. Figure 2 shows the temperature dependence of sample R198. The other samples show similar temperature dependence.

Finally, the behavior of PL efficiency with respect to the pump excitation energy was studied. The samples were ex-

[a]Electronic mail: s.dilna@tue.nl

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TABLE I. Growth details of the samples. All samples have 500 nm thick GaAs buffer layer grown at 580 °C, a lower LT-GaAs barrier layer of 500 nm, and a capping layer of 200 nm LT-GaAs, both grown at 250 °C.

| | | Temperature (°C), time (min[a]) | |
|---|---|---|---|
| Sample | QDs+interlayer | First anneal | Second anneal |
| R152 | 2 ML InAs (250 °C) | No | No |
| R181 | 2 ML InAs (250 °C) | 480 °C, 15 min | No |
| R191 | 2 ML InAs (250 °C) | 480 °C, 10 min | 480 °C, 10 min |
| R198 | 2 ML InAs (250 °C) | 480 °C, 10 min | 600 °C, 10 min |
| R200 | 2 ML InAs (250 °C)+ 3 nm GaAs (480 °C) | No | No |
| R207 | 2 ML InAs (250 °C)+ 3 nm GaAs (480 °C) | 580 °C, 10 min | No |
| R228 | 2 ML InAs (480 °C) | No | No |

[a]Ramping time is not included.

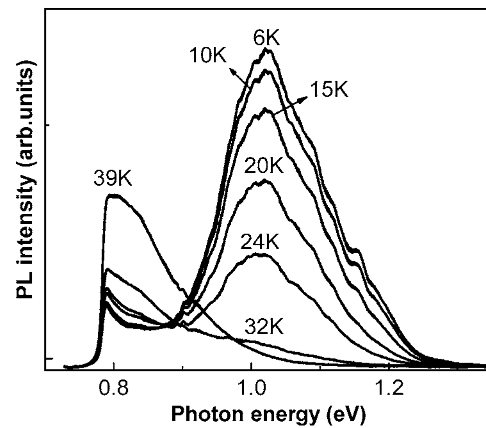

FIG. 2. Temperature dependence of the PL spectrum for sample R198 showing rapid quenching of the PL from the LT-QDs with increasing temperature.

cited in an energy range extending from 1.4 to 1.6 eV, i.e., both below the GaAs band gap directly into the InAs QD layer (<1.518 eV) and also above the GaAs band gap in the GaAs barrier (>1.518 eV). The resulting PL efficiency was recorded for all the samples. The measurements were carried out at liquid helium temperature. Figure 3 shows the PL peak intensity as a function of excitation energy for all the seven samples. A huge increase in PL efficiency is observed when excited below the GaAs band gap directly into the InAs QD layer, which is exactly opposite as observed for conventional QDs. The increase in efficiency varies between a factor of 45 for R198 and 280 for R152. The highest step is observed for R152, which is not annealed and thus has the highest concentration of trapping centers, leading to the highest trapping probability for excitation in the GaAs barrier.

It has already been shown that during LT growth, excess As is incorporated as As antisite point defects ($As_{Ga}$),[1,5,11] which are fast trapping centers for the carriers. These are mid-band-gap point defects, which can act as double donors.[1,5] The concentration of the point defects can be increased by further decreasing the growth temperature.[12] It was estimated by Haiml *et al.*[1] that around 10% of all $As_{Ga}$ centers are ionized $As_{Ga}^{+}$ states due to Ga vacancies, which act as acceptor states. The remaining $As_{Ga}$ are neutral. The electrons are trapped by the ionized $As_{Ga}^{+}$ in the GaAs barrier, which leads to the well known fast response in LT-GaAs. Surprisingly, it is not observed that these trapping centers are also present inside the QDs, since the PL efficiency at 4 K is quite reasonable for excitation below the GaAs band gap. The capture of an electron into the defects in the LT-GaAs can occur along two routes.[13] First is the direct capture of electrons, which are optically excited in the LT-GaAs barrier and which are directly trapped into the trapping centers before they are captured into a QD. A second channel is electron relaxation from the barrier into the QD level, followed by a multiphonon assisted tunneling out of the QD into the trapping center located in its vicinity. Sercel[13] had theoretically shown that the interaction between the electronic wave functions of the QD and the deep trap is likely to provide a tunneling transition.

The rapid fall and the subsequent complete quenching of the PL efficiency with rising temperature can be explained by the tunneling induced trapping of carriers out of the QD into the defects in the LT-GaAs. As temperature increases, the carriers acquire enough energy to tunnel across and get trapped, thus quenching the PL efficiency with rising tem-

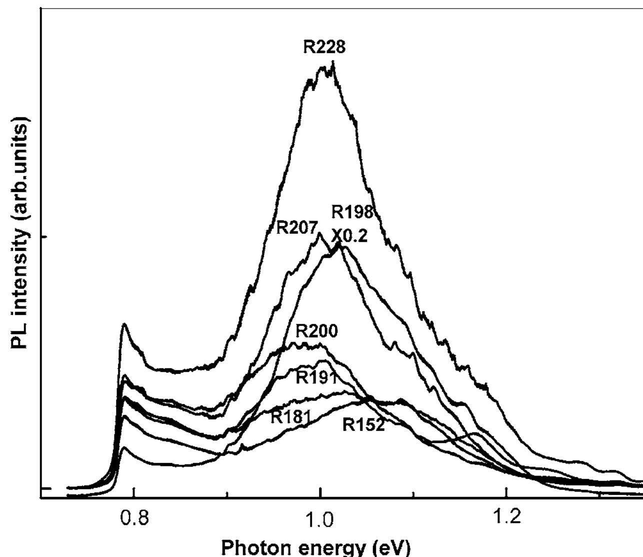

FIG. 1. Photoluminescence from LT grown InAs/GaAs QDs at a temperature of 5 K and a high 10 $kW/cm^2$ excitation density.

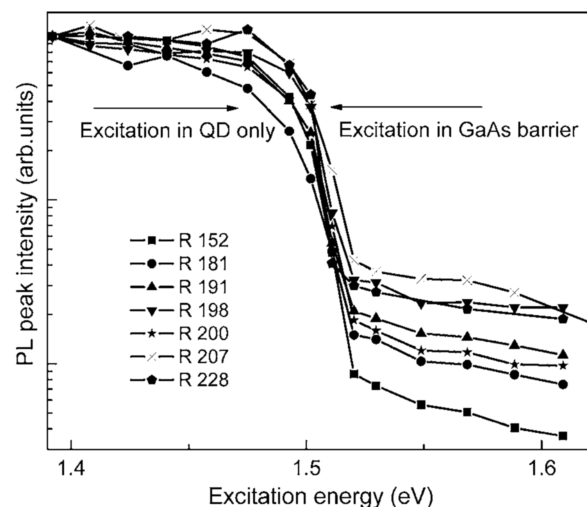

FIG. 3. Excitation energy dependence on PL efficiency showing an unexpected increase of the QD PL efficiency when exciting below the GaAs band gap directly into the QD layer.

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perature. The excitation wavelength dependence confirms our observation that the PL quenching centers are located only in the GaAs barrier and not inside the QDs, which is in agreement with Deep Level Transient Spectroscopy studies reporting point defect in the nearest neighborhood[8–10] of the QDs within the GaAs barrier, but not within the InAs QDs themselves. In conventional high temperature grown QDs, carriers created in the barrier region above the GaAs band gap diffuse towards the QDs in which they relax down towards the QD ground state. For high temperature grown QDs, it is well known that the carrier density excited in the GaAs barrier is much higher than the carrier density for excitation below the GaAs band gap in the QD layer. Thus, excitation into the QD layer is expected to yield a low PL efficiency, since the absorption probability for the excitation beam is very small. Contrary to this observation in normal high temperature grown dots, we find that the PL efficiency is much higher when excited below the GaAs band gap into the QD layer. This is true even for the high temperature QDs embedded in a low temperature grown GaAs barrier. It can be thus concluded that in a LT-GaAs barrier layer, optically excited carriers are efficiently trapped into defects in the barrier before being captured into the QDs. When the carriers are generated in the QD layer, diffusion through the high trapping efficiency LT-GaAs barrier is circumvented, leading to a much higher PL efficiency. Since the PL quenching centers are not present in the wetting layer and QDs, trapping is now only possible when carriers tunnel out of the QD into a trap, which is clearly a less probable trapping mechanism at 4 K. As a result, a higher PL efficiency is observed when the carriers are excited into the QD layer. If the optical absorption in the QD layer itself is considered and the much larger absorption in the LT-GaAs layer is neglected for the moment, we surprisingly still observe that the PL efficiency is considerably higher for excitation in the QD layer below the GaAs band gap than for excitation inside the QD layer above GaAs band gap. When the excitation energy is tuned above the GaAs band gap, optically generated carriers in above-the-barrier unconfined QD levels diffuse towards the LT-GaAs and are subsequently trapped in the LT-GaAs. It can thus be concluded that, even when the carriers are initially within the unconfined levels of QD layer, diffusion followed by trapping is more efficient than relaxation within the InAs QD layer towards the confined QD levels. Sercel[13] had proposed that electrons not only tunnel out of the QD towards a trap but can also tunnel back to the dot from a trap. So an equilibrium density of electrons will be present inside the dots, leading to the observed PL at 5 K. When the temperature increases, the probability for tunneling out of the QD into a trap rapidly increases, leading to the decrease in the PL efficiency. This also explains the high pump excitation density needed to observe the PL spectra.

In conclusion we observe that LT growth gives rise to efficient trapping centers due to excess As in the LT-GaAs barrier but not inside the LT grown QDs. This leads to a reduction in the number of carriers relaxing into the QDs, resulting in a reduction of the QD PL efficiency when excited in the barrier. The strong increase of the PL efficiency when excited directly into the QDs shows that the dots are of good optical quality without the presence of PL quenching traps inside the dots. LT grown InAs/GaAs QDs thus constitute of good optical quality radiative QDs, embedded in an optically "dead" GaAs barrier material, which traps away the carriers before they diffuse into the QDs.

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