Growth and characterization of InAs quantum dots on GaAs (100) emitting at 1.31μm

Vincent Célibert, Bassem Salem, Gérard Guillot, Catherine Bru-Chevallier, Laurent Grenouillet, Philippe Duvaut, Philippe Gilet, Aurélien Million

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V. Celibert [1,2], B. Salem[1], G. Guillot [1], C. Bru-Chevallier [1].
L. Grenouillet [2], P. Duvaut[2], P. Gilet [2], A. Million [2].
[1] LPM, INSA de Lyon, UMR-CNRS-5511, Bat. Blaise Pascal, 7 avenue Jean Capelle
69621 VILLEURBANNE CEDEX, France.
[2] CEA-DRT-LETI/DOPT - CEA/GR - 17 rue des Martyrs
38054 GRENOBLE CEDEX 9 - France

ABSTRACT

Self-organized InAs quantum dots (QDs) were grown in the Stranski-Krastanov regime, by gas-source molecular beam epitaxy (GSMBE), on (100) GaAs substrates. Two important parameters have been optimized in order to grow high quality QDs with a very good reproducibility: InAs growth rate and GaAs cap layer deposition rate. Atomic force microscopy (AFM) analysis shows a unimodal QD distribution and the room temperature photoluminescence (RTPL) spectrum of the optimized sample reveals a 1.3 µm emission with a 19 meV full width at half maximum (FWHM). Photoluminescence (PL) measurements versus excitation power density and photoluminescence excitation (PLE) measurements clearly show multi-component PL emission from transitions associated with fundamental and related excited states of QDs. Furthermore a good growth reproducibility is observed. The results are promising for further work which will lead to laser fabrication.

INTRODUCTION

The interest for quantum dots remains strong in the domain of telecommunications or quantum cryptography for example. Even if recent achievements made possible to approach the 1.53-1.56 µm telecommunication window [1] on GaAs substrates, reserved up to now to InP, it remains very important to control the growth on GaAs of QDs emitting at 1.26-1.31 µm, the second telecommunication window. Some promising results have already been obtained with the demonstration of laser emission around 1.3 µm [2]. Simultaneously ground state and excited state lasing has been recently demonstrated by *Markus et al.* [3]. The realization of optically active QDs emitting in the desired range has no meaning for laser applications if the structures do not provide the benefits of the predicted low-threshold current and high-temperature stability of QD-based lasers. To access these features, the FWHM of the QD PL, currently (30-60 meV) [4], has to be drastically decreased.

In this work, we investigate the influence of a high GaAs cap layer deposition rate and a low InAs growth rate on the elaboration of self-assembled InAs/GaAs QDs. The structural properties of the optimized QDs sample are probed by AFM and the optical properties are studied using PL as a function of the excitation power, as well as PLE spectroscopy.

EXPERIMENT

The samples were grown at 500°C by GSMBE on Si-doped GaAs (100) substrates. After thermal desorption at 585°C of the GaAs native oxide, a 450 nm GaAs buffer layer was grown.

The temperature of the substrate was reduced and precisely adjusted at 500°C with an optical pyrometer. Then a two step growth of InAs quantum dots (QDs) was carried out. In the first step, 1.96 monolayer (ML) InAs deposition was performed just above the critical thickness of the Stranski-Krastanov mode at different growth rates (from 0.04 to 1.4ML/s). The critical threshold between the two-dimensional (2D)/three-dimensional (3D) growth modes was checked by in-situ reflection high electron energy diffraction (RHEED). At this point, the growth was interrupted under arsenic pressure. The second step consists of a 0.32 ML InAs deposition with the same growth conditions as in step 1. After a new growth interruption, a 200 nm GaAs capping layer is performed with different deposition rates. AFM was performed using a Digital D3100 IIIA microscope operating in the contact mode.

PL spectra were recorded in the 8-300K temperature range using the 514.5 nm line of an Ar^+-ion laser as the excitation source. PL signal was detected by a 77K cooled Ge photodetector through a spectrometer, using a conventional lock-in technique. PLE spectra were recorded at 8K using a tungsten lamp, dispersed by a spectrometer as the excitation source.

RESULTS AND DISCUSSION

First, the effects of the GaAs cap layer deposition rate were studied. Different rates were performed from 1.4 to 3.5 ML/s at a constant InAs growth rate (0.1 ML/s). The QD RTPL emission (figure 1a) redshifts from around 1180 to 1265 nm with the increasing GaAs cap layer deposition rate. In the same time, the FWHM is decreasing from 49 to 35 meV.

In order to obtain long wavelength emission, large QD volume and In content are desirable. It has been shown in the literature [5, 6] that a high capping rate "freezes" the QDs and therefore minimizes In segregation and In-Ga intermixing, preserving the QD volume and In concentration during capping, these parameters being important to reach 1.3 µm. The FWHM reduction with the increasing capping growth rate suggests that the QD size homogeneity keeps pretty good because there are no significant changes of the QD dimensions.

However, for a GaAs cap layer deposition rate over 3.1 ML/s, no more PL is observed because the growth rate becomes too high and far away from the optimal GaAs growth conditions. A lot of non-radiative defects will appear in the GaAs cap layer as well as in the QDs, and the emission intensity is drastically reduced.

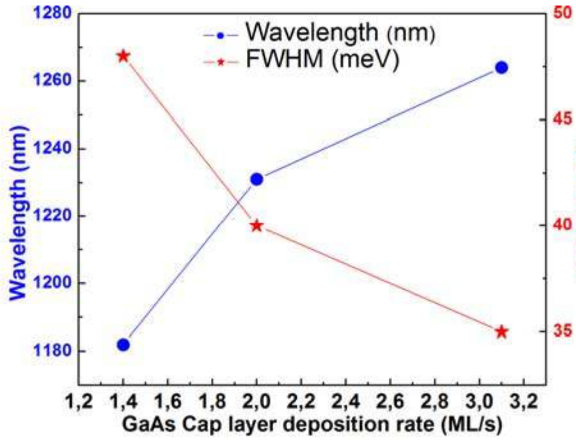


Figure 1a. Wavelength and FWHM of the RTPL fundamental peak as a function of cap layer deposition rate. InAs growth rate is 0.1ML/s.

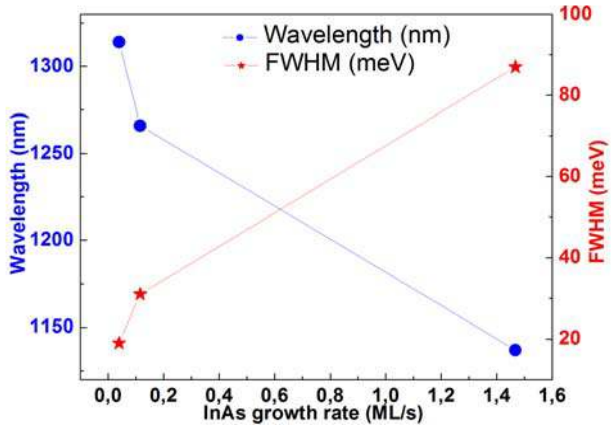


Figure 1b. Wavelength and FWHM of the RTPL fundamental peak as a function of InAs growth rate. GaAs cap layer deposition is 3.1 ML/s.

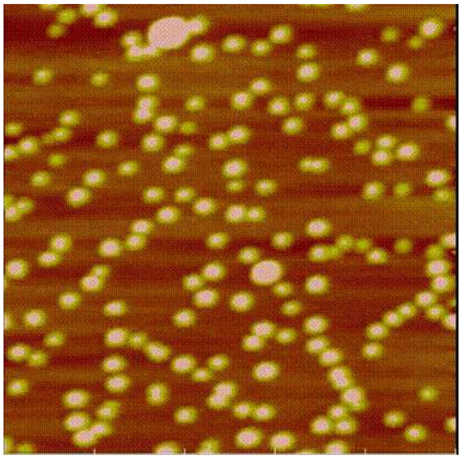
Secondly, keeping constant the GaAs capping rate at 3.1 ML/s, the influence of the InAs growth rates on the emission wavelength and on the FWHM was studied. QDs were grown at rates between 0.04 and 1.4 ML/s. The QD RTPL (figure 1b) redshifts from around 1130 to 1310 nm and the FWHM decreases from 87 to 19 meV.

Lu et al. [7] have shown that a small growth rate provides a longer time for the InAs islands to evolve in size after their nucleation. The InAs islands have more time to grow up during the deposition process. Another reason why the regime of very low growth rate can produce such large QDs is related to the enhanced diffusion of In atoms during deposition, that allows most of the incoming material to be incorporated into the existing islands instead of forming new structures. *Joyce et al.* [8] showed that the island mean height and mean diameter concurrently with the QD size fluctuation become larger by decreasing the growth rate. The increase of the QD dimensions and the decrease in size fluctuations for low growth rate is at the origin of the shift towards 1.3 µm emission and of the reduction in the inhomogeneous FWHM of the PL emission as observed in figure 1b.

A high GaAs cap layer deposition rate and a low InAs growth rate appear therefore to be very important parameters to increase the QD overall dimensions (that will redshift the RTPL).

The optimized sample, which is studied in the following, is grown using a 3.1 ML/s GaAs cap layer deposition rate and a 0.04 ML/s InAs growth rate. Additionally, note that the growth of the optimized sample is highly reproducible: nominally 9 identical samples grown ~14 months apart show very similar PL properties.

Structural characterizations were performed by AFM techniques. Figure 2 presents a typical AFM image of the uncapped version of an optimized sample. The QD density is approximately $2x10^{10}cm^{-2}$. The mean height and base deduced from AFM image are 7 nm and 50x50 nm², respectively. It has to be pointed out that the image shows a unimodal distribution. The two greater spots can be attributed to surface defects or greater QDs formed by coalescence of a small number of QDs.



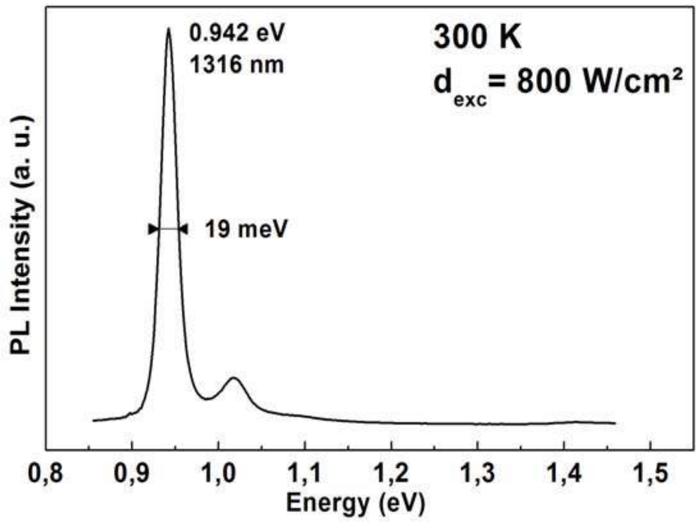


Figure 2. AFM image of the InAs QDs grown on GaAs (100) of the uncapped optimized sample.

Figure 3. RTPL spectrum of the optimized sample.

The RTPL spectrum of the optimized sample is shown in figure 3. The RTPL spectrum, realized in the 0.85-1.45 eV range shows two well-defined peaks. The peak at 0.942 eV (1.316 µm) with a very narrow FWHM (19 meV) is attributed to the ground state of the QD of the optimized sample. The second peak has a PL maximum at 1.02 eV. Its origin will be discussed later. To our knowledge, this QD emission spectrum is at the international state of the art [1, 4, 8, 9].

In order to identify the origin of the optical transition, PL of the optimized sample was carried out at 8K as a function of the excitation power. By increasing the excitation intensity from 0.9 mW/cm² to 4360 W/cm², the spectra change noticeably as shown in figure 4. Only one optical transition is observed at low excitation. This single peak at 1.02 eV is attributed to the ground state of the QDs (at 8K) in agreement with a unimodal distribution as shown by AFM measurements. By increasing the intensity of the excitation power, we observe the arising of two other peaks, in the same time the intensity of the ground state saturates. This behavior suggests excited state signatures for the peaks around 1.1 eV and 1.15 eV.

A small redshift of the fundamental peak is recorded with increasing excitation. This effect is attributed to Coulombian interaction [10] and the energy difference between the fundamental peak and the first excited state is slightly increasing with the increasing excitation because the carriers on a level interact with the carriers on the other levels.

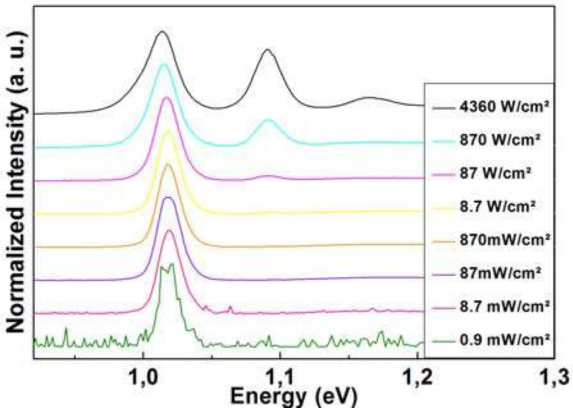


Figure 4. PL spectra as function of excitation density at 8K for the optimized sample.

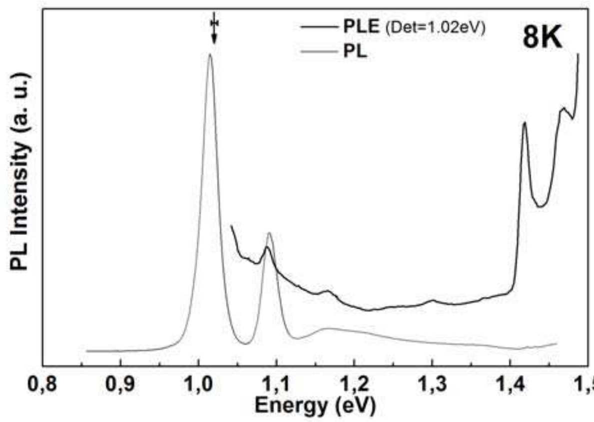


Figure 5. PL and PLE spectra of InAs QDs at 8K for the optimized sample.

The origin of the peaks revealed by the study as a function of power excitation was confirmed by PLE measurements. The excited spectrum of QDs is commonly investigated in high-density PL power excitation spectrum, taking advantage of state filling like shown before. However, such experiments probe the emission of highly populated QDs for which many-particle effects need to be taken into account. PLE spectroscopy is a powerful tool for the investigation of the excited state absorption spectrum of empty QDs. The observation of a PLE signal requires a two-step process. First, an exciton is generated in an excited QD state by absorption and, second, this exciton relaxes to the ground state.

The PL and PLE spectra of the optimized sample at 8K are shown in figure 5. The dotted line represents the PL spectrum and the straight line represents the PLE spectrum at 8K. The arrow at 1.02 eV indicates the energy of the PLE detection. The spectrometer has a 5 meV resolution.

The PL spectrum at 8K and high excitation density clearly shows three emission peaks at 1.02, 1.09 and 1.16 eV. Note that figure 5 demonstrates the match of the resonances of the PLE spectrum with PL emission at 1.09 and 1.16 eV, and thus confirms these peaks are excited states. This result has to be pointed out because PLE spectra reported for self-organized InAs/GaAs QDs were so far mostly dominated by LO-phonon-related resonances [11-13], providing information on the relaxation processes rather than on the excited state spectrum, contrary to our results. The peak at around 1.3 eV is not yet attributed: it could be another excited state or a phonon resonance. The heavy and light hole absorption of the wetting layer leads to excitation resonances at around 1.43 eV and 1.48 eV. These transition energies correspond to a ~1-ML-thick InAs quantum well [14].

CONCLUSION

Therefore through the optimization of the high GaAs cap layer deposition rate and the low InAs growth rate for the QDs formation, we obtained QDs with very good structural and optical properties: RTPL emission at 1.3µm with a very narrow size dispersion (FWHM of 19 meV) comparing to the international state of the art. The density is around 10^{10} cm^{-2}. In addition, via PL versus excitation power and PLE spectroscopy, we have identified excited states of the QDs. Due to a very good growth reproducibility, many samples exhibited very close characteristics: this is promising for the fabrication of laser devices containing QDs in their active layer.

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