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MBE growth optimization and optical spectroscopy of InAs/GaAs quantum dots emitting at 1.3 μm in single and stacked layers

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Self-organized InAs quantum dots (QDs) are grown in the Stranski–Krastanov regime, by molecular beam epitaxy, on (1 0 0) GaAs substrates. In order to grow high-quality QDs emitting at 1.3 μm , an unusual two-step growth procedure is first developed, with a growth interruption during the InAs deposition, just above the critical thickness. Then two important growth parameters are considered. First, the GaAs cap layer deposition rate is optimized, the InAs growth being kept constant. Second, the InAs growth rate is optimized, at optimized GaAs cap layer deposition rate. The optimizations leads to large QDs with a unimodal size distribution and the room temperature photoluminescence (PL) spectrum peaks at 1.3 μm with a 19 meV full-width at half-maximum (FWHM). These optical properties are at the international state of art. Then, three QD layers are stacked with different spacer thickness in order to increase the QD density necessary for laser applications. The best optical properties are obtained for the wider GaAs spacer (45 nm): PL emission around 1.3 μm , narrow FWHM (31 meV), and PL intensity enhanced by a factor of 3. The results are promising for further incorporation of the QD stacks in the active region of a laser.

Keywords: A1. Nanostructures; A2. Quantum dots; A2. Molecular beam epitaxy; B2. Semiconducting III–V Materials; B3. Heterojunction semiconductor devices

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1. Introduction

Low-dimensional carrier confinement nanostructures such as quantum wires and dots are

quite attractive for applications to high-performance electronic and optical devices such as lasers [1,2]. Even if recent achievements made it possible to approach on GaAs substrates the 1.53–1.56 μm telecommunication window [3] reserved up to now to InP, it remains very important to control the growth of QDs on GaAs emitting at 1.26–1.31 μm , the second telecommunication window. Promising results have already been obtained with the demonstration of laser emission around 1.3 μm [4]. But, the realization of optically active QDs emitting in the desired spectral range has no meaning for laser applications if the structures do not provide the benefits of low threshold current and high-temperature stability. To access these features, the full-width at half-maximum (FWHM) of the quantum dot (QD) photoluminescence (PL) needs to be as small as possible (state of the art is less than 20 meV [5,6]) and a high-density of self-assembled QDs is required to enhance the optical gain.

The influence of both GaAs cap layer and InAs deposition rates on the growth of self-assembled InAs/GaAs QDs has already been studied separately in Refs. [7–10] and [3,11,12], respectively. They were never combined before in a single study in order to improve the quality of QDs emitting at 1.3 μm . Several groups have studied the GaAs cap layer rate with a conventional InAs growth rate (generally around 0.1 ML/s), but this was not allowed to reach 1.3 μm emission with good optical characteristics. Other groups have used very low InAs growth rates (around 10^{-3} ML/s) and low GaAs cap layer deposition rates (around 1 ML/s). They succeeded in reaching 1.3 μm , but a long time was needed to grow a single layer, which is compatible neither with high-quality growth nor with mass production. The aim of this study is to optimize both GaAs cap layer deposition rate and InAs QD growth rate, combining both of them in order to reach 1.3 μm emission with very good structural and optical properties. After growth optimization for a single layer, three QD layers are stacked in order to achieve a higher QD density. The influence of GaAs spacer thickness on the optical properties is also studied.

2. Experiment

Single layer samples are grown at 500 °C by gas source molecular beam epitaxy (GSMBE) on Si-doped GaAs (1 0 0) substrates, using the Stranski–Krastanov (S–K) growth mode. After thermal desorption at 585 °C of the GaAs native oxide, a 500 nm GaAs buffer layer is grown. The substrate temperature is reduced and precisely adjusted at 500 °C with an optical pyrometer. Then a two-step growth of InAs QDs is carried out as described below. In the first step, the deposition of a 1.96 InAs monolayer (ML) is performed at different growth rates (from 0.04 to 1.4 ML/s) slightly above the critical thickness of the S–K mode. The critical threshold between the two-dimensional (2D)/three-dimensional (3D) growth modes is checked by in situ reflection high electron energy diffraction (RHEED). At this point, the growth is interrupted under an arsenic beam equivalent pressure of 5×10^{-6} Torr. The second step consists in a 0.32 InAs ML deposition with the same growth conditions as in step 1. After a new growth interruption, a 100 nm GaAs cap layer is deposited at different deposition rates (from 1.4 to 3.1 ML/s).

Stacks of three QD layers are grown by solid source MBE, using the same deposition procedure as described before. It is worth noting that, although the As source is changed from a gas source to a solid source, we checked that neither structural nor optical properties of the QD are altered by this modification. The InAs and GaAs layer deposition rates are fixed following the previous optimization procedure for a single layer growth. Samples with different GaAs spacer layer thickness (ranging between 5 and 45 nm) are grown and their optical properties are evaluated.

Structural properties of uncapped InAs QD are studied by atomic force microscopy (AFM). PL spectra are recorded in the 8–300 K temperature range using the 514.5 nm line of an Ar^{+} -ion laser as the excitation source. PL signal is detected by a 77 K cooled Ge photodetector through a spectrometer, using a conventional lock-in technique.

3. Results and discussion

3.1. Two-step growth procedure

In order to improve the structural and optical properties of InAs QDs, a two-step growth procedure is developed as described in the previous section. Fig. 1 shows typical AFM images and room temperature PL spectra of InAs QDs obtained either following the usual one-step growth procedure (sample 1) or following the two-step growth procedure (sample 2). In both cases, the InAs and GaAs growth rates are fixed at 0.1 ML/s and around 1.4 ML/s, respectively. The QD density (around 10^{10} cm^{-2}) is decreased by a factor of 2 using the two-step growth procedure as compared to the usual one. Both samples exhibit a bi-modal size distribution; however the overall size distribution is reduced in sample 2. The room temperature PL spectrum (RTPL) reveals a large redshift (from $1.18 \mu\text{m}$ in sample 1– $1.26 \mu\text{m}$ in sample 2) and a significant reduction of the FWHM (from 47 to 26 meV) in good agreement with the observed QD size distribution. Actually, the growth interruption which is performed during the InAs deposition immediately at the onset of the 2D–3D transition, is expected to initiate the

seeding of the QDs and to allow for atom migration at the surface, favoring the vanishing of small dots to the benefit of large ones. During the following InAs deposition, the QDs are ripening towards higher overall dimensions: this induces the PL redshift and explains the narrower size distribution of sample 2. Another advantage of such a two-step procedure is an increase in PL intensity as observed in Fig. 1 because a maximum of QDs emit around the same energy. Therefore, the two-step growth procedure is used in the next sessions.

3.2. Growth parameter optimization

In order to obtain long wavelength emission, large QD volume and high In content are required. It has been shown in the literature [8,13] 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. In Fig. 2 are plotted the RTPL spectra recorded in three samples grown with different GaAs deposition rates between 1.4 and 3.1 ML/s, at a constant InAs growth rate (0.1 ML/s). The PL emission is redshifted from 1.18 to $1.265 \mu\text{m}$ with increasing GaAs cap layer

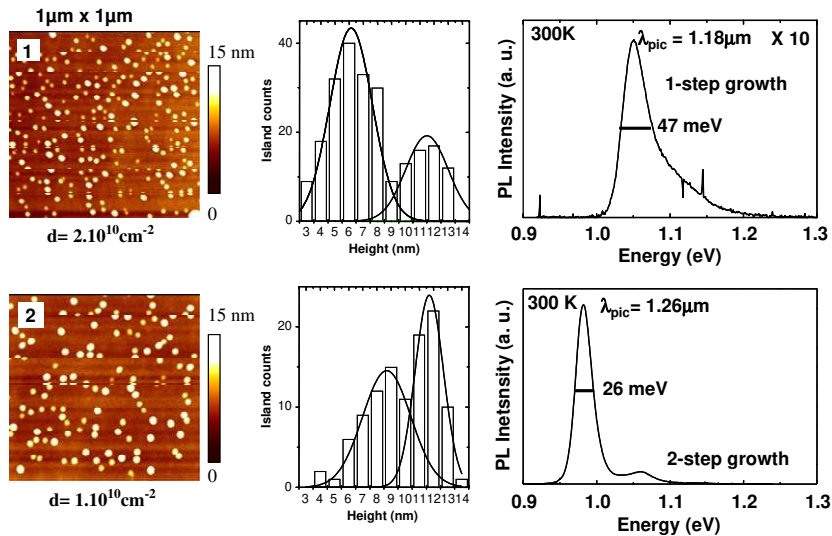


Fig. 1. AFM images, histograms of the QD size distribution and PL spectra for a one-step growth sample (1) and a two-step growth sample (2).

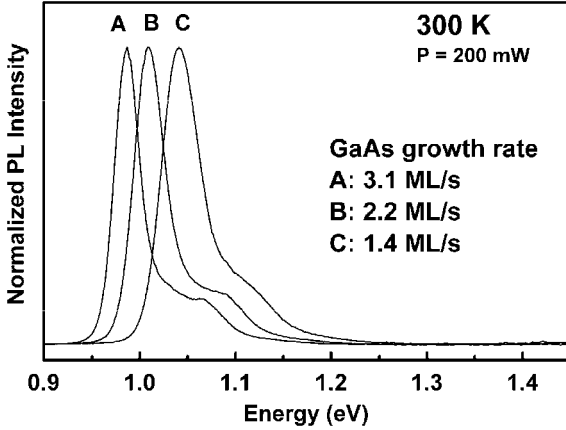


Fig. 2. Normalized PL intensity as a function of energy for different GaAs cap layer deposition rates (from 1.4 to 3.1 ML/s). A high rate “freezes” the QDs and enhances PL emission and FWHM.

deposition rate. At the same time, the FWHM is decreased from 49 to 35 meV. Such optical properties suggest that, as expected, QDs are larger and their size homogeneity is improved by increasing the GaAs cap layer deposition. However, the PL intensity is shown to decrease as the GaAs cap layer deposition rate is increased, and it becomes undetectable over 3.1 ML/s. Indeed in this case, the growth rate becomes too high and far away from the optimal GaAs growth conditions, generating a lot of non-radiative defects which are drastically reducing PL intensity.

Now, keeping constant the GaAs capping rate at 3.1 ML/s, the influence of the InAs growth rate on the optical properties is studied. Indeed, Lu et al. [14] have shown that a small InAs growth rate provides a longer time for the InAs islands to evolve in size because the enhanced diffusion of In atoms allows most of the incoming material to be incorporated into the existing islands instead of forming new structures. Joyce et al. [15] have shown that the QD size increases and the size fluctuations decrease at low InAs deposition rates.

QDs are grown at InAs deposition rates ranging between 0.04 and 1.4 ML/s. The RTPL spectra plotted in Fig. 3 exhibit a redshift from 1.08 up to 1.31 μm and a decrease in the FWHM from 87 down to 19 meV as the InAs deposition rate is

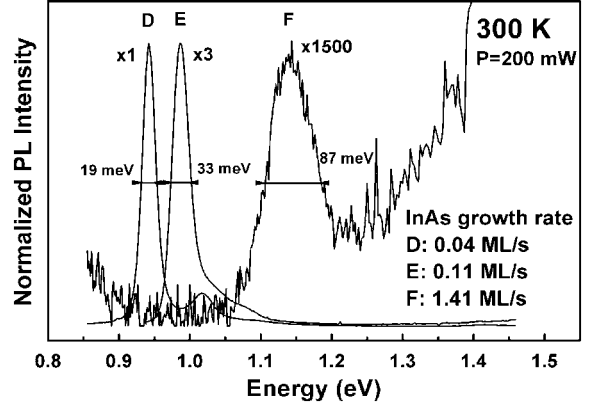


Fig. 3. Normalized PL intensity as a function of energy for different InAs deposition rates (from 0.04 to 1.41 ML/s). A very low rate enhances PL emission and decreases the FWHM.

decreased. This is in good agreement with previous work on this subject. Therefore, besides the fact of introducing a growth interruption during the InAs deposition, it turns out that both a high GaAs cap layer deposition rate together with a low InAs growth rate are still crucial to increase the QD size and homogenize their dimensions in order to get a narrow distribution of QDs emitting at 1.3 μm at 300 K.

In the following, the structural and optical properties of an optimized InAs QDs plane are shown (GaAs deposition rate 3.1 ML/s and InAs deposition rate 0.04 ML/s). Fig. 4 shows a typical AFM image of the uncapped optimized sample. The QD density is approximately $2 \times 10^{10} \text{ cm}^{-2}$. The mean height and base surface deduced from the AFM image are 7 nm and $50 \times 50 \text{ nm}^2$, respectively. It has to be pointed out that the image shows a unimodal distribution. The two larger spots can be attributed to surface defects or larger QDs resulting from the coalescence of a small number of QDs. Note that the AFM measurements show the dimensions of uncapped QDs. The strain induced by a cap layer is expected to reduce the dimensions (and particularly the QD height) [7,9].

The RTPL spectrum of the optimised QDs is plotted in Fig. 5 and exhibits two well-defined peaks. The peak at 0.942 eV (1.316 μm) with a very narrow FWHM (19 meV) is attributed to the

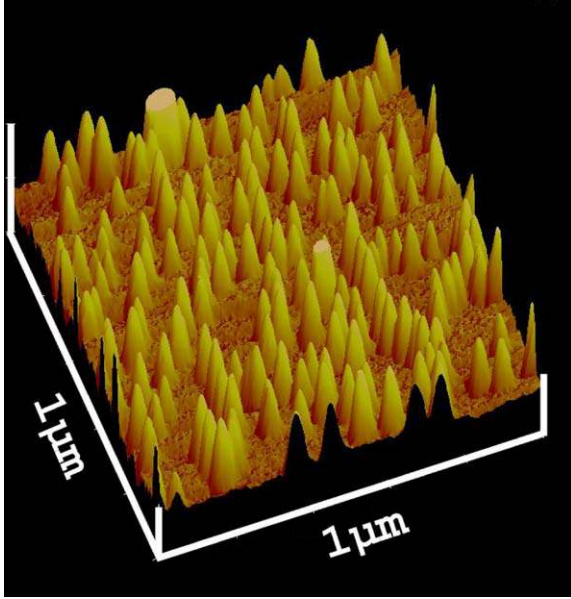


Fig. 4. AFM image ($1\mu\text{m} \times 1\mu\text{m}$) of InAs QDs of the uncapped optimized sample grown on GaAs (100).

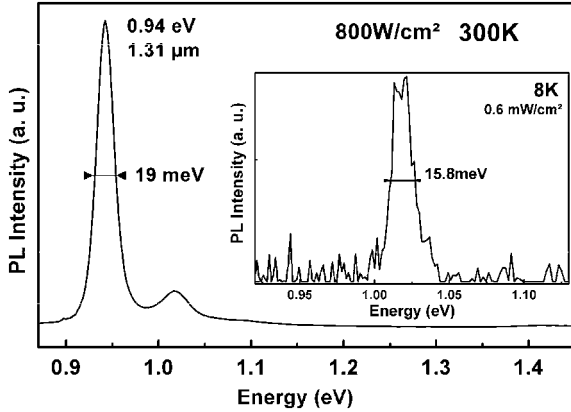


Fig. 5. PL spectrum at 300 K of the QD optimized sample at 800 W/cm^2 . PL spectrum at 8 K (inset) at very low excitation power (0.6 mW/cm^2).

ground state of QDs. The peak at 1.02 eV is attributed to the first excited state following complementary PL excitation spectroscopy and PL study as a function of the excitation power, as previously published [16]. Note that even if the PL spectrum is realized at high excitation density (800 W/cm^2), no PL emission is detected around

1.43 eV which would come from the wetting layer. This is a proof for a good radiative recombination efficiency, as most of the photo-generated carriers fall down in the QDs and mainly recombine in the ground state of the dots. This fact denotes high-quality growth. The inset in Fig. 5 is the PL spectrum of the optimized QDs recorded at 8 K at very low excitation power (0.6 mW/cm^2). Note the very narrow inhomogeneous linewidth of 15.8 meV measured at 8 K. This result is at the state of art compared to recent results obtained by MBE growth [6] and by low-pressure metal organic chemical vapor deposition (MOCVD) [5].

3.3. QD layer stacking

As previously stated, a high density of self-assembled QDs in a small volume is required for laser applications. One way to reach a high QD density is to stack InAs QD layers separated by thin GaAs spacer layers. In this section, the influence of the thickness of the GaAs layer on the QD optical properties is studied.

Four samples containing three QD layers are grown at 500°C with different GaAs spacer layer thickness ($d = 5, 12, 20$ and 45 nm). A single QD layer sample is also grown for comparison. The growth procedure for each QD layer is the same as described in previous sections. The InAs growth rate is set up at an optimized value (0.04 ML/s), whereas the GaAs cap layer deposition rate is fixed at a value (2 ML/s) lower than the optimized one. This deposition rate will lead to an emission wavelength slightly lower than $1.3\mu\text{m}$ at 300 K (Fig. 2). However, it is expected that at higher temperature (i.e. operating temperature of lasers) PL emission will be redshifted over $1.31\mu\text{m}$. In Fig. 6, the RTPL spectra of the four samples are plotted, together with the single layer reference sample. For the largest spacer layer thickness (45 nm), the PL spectrum is very similar to the single layer reference sample: peak energy 0.99 eV (instead of 0.97 eV), FWHM equal to 31 meV (instead of 30 meV) and a threefold increase in PL intensity as expected when stacking three QD planes: in this case, each QD plane is independent. When the spacer layer thickness is decreased down

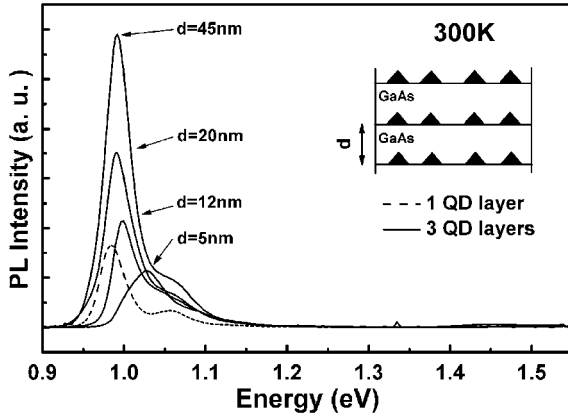


Fig. 6. PL spectra of three QD layers stacks, separated by 5, 12, 20 and 45 nm GaAs spacer layers. Dashed line: single plane reference sample.

to 5 nm, the PL energy blueshifts from 0.99 up to 1.02 eV, while the FWHM increases from 31 to 43 meV and the PL intensity decreases by a factor of 3. As the spacer layer thickness is decreased, both an increase in QD size and a vertical coupling between charge carriers are expected to occur [17,18]. Both of them should result rather in a redshift of the PL spectrum, contrary to our observations. Such a PL blueshift however has already been observed in InAs/GaAs QD stacked planes [19] and is tentatively explained by the effect of the complex strain fields existing in the closely stacked QD. For the smaller spacer thickness (5 nm), PL intensity is drastically reduced and the FWHM is increased. This is attributed to the effect of spacer layer thickness fluctuations that may become more significant at small d and to the deterioration of GaAs intermediate layer quality induced by QD stacking [20].

Therefore, the best choice to get high optical gain for laser application is to use the largest spacer layer thickness (45 nm) in which high quantum efficiency and high homogeneity are achieved.

As done before for the single QD layer, an excitation power dependence PL experiment at 8 K showed clearly the nature of the peaks of each spectrum: the peak at the lowest energy is attributed to the ground state of the QDs and

the peaks at higher energy are excited states. Only one QD population is detected.

4. Conclusion

A two-step growth procedure is proposed for the growth of high-quality InAs QDs on GaAs substrates. It consists in introducing a growth interruption during the QD elaboration. Combined with an optimization of both GaAs cap layer deposition rate, and InAs deposition rate, this procedure allows to grow QDs at the international state of the art: RTPL emission at 1.3 μm with a very narrow size dispersion (FWHM = 19 meV at 300 K and lower than 16 meV at 8 K). The QD density is a few 10^{10} cm^{-2} . In order to increase the QD density as required for laser application, QD stacks are realized with different spacer thickness. The best PL properties are obtained for 45 nm wide GaAs spacers: PL emission at 1.3 μm , narrow FWHM (31 meV), comparing well with the single layer reference sample. The threefold increase in PL intensity is promising for the future introduction of QD stacks in laser active layers.

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