Low Density Metamorphic Quantum Dot structures with emission in the 1.3 - 1.55 µm window

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Abstract. We report on the molecular beam epitaxy growth of InAs/InGaAs metamorphic quantum dot structures for single-photon operation at long wavelengths. Low density of quantum dots has been achieved by depositing sub-critical coverages of InAs, while the redshift of emission was obtained by growing the nanostructures on relaxed InGaAs buffers. By optimizing the design and growth parameters, such as the InAs coverage, the post growth annealing time and the compositions of InGaAs confining layers, we were able to obtain structures with quantum dot densities of the order of the 10^8 cm⁻² and emission in the whole range 1.3 - 1.55 µm at low temperature.

1. Introduction

Future applications in the fields of quantum cryptography and quantum communication will need efficient single photon sources emitting in the telecom windows (1.3 - 1.55 µm). Self-assembled InAs/GaAs quantum dots (QDs) [1-2] have been recognized as ideal nanostructures for such innovative devices [3-4]. However, particular growth procedures are necessary to have a QD surface density low enough to allow study of single photons (in the order of a few dots/µm²) [5-6]. The deposition of an InAs layer with a coverage smaller (sub-critical) than the critical thickness for the two- to three-dimensional (2D–3D) growth mode followed by a growth interruption was demonstrated as being particularly useful for different systems [7-8]. Notwithstanding, low density structures with emission in the telecom windows for structures grown on GaAs have been rarely reported: while 1.3 µm emission at low temperature has been reached [5-6], 1.55 µm emission from low density QDs is still a difficult task. For high density structures, however, this objective has been reached by depositing QDs on metamorphic buffers [9-10]: thanks to the decrease of the QD strain and of band discontinuities the emission wavelength may be consistently redshifted [11]. Very recently, low density QDs on metamorphic buffers have been obtained by stopping the substrate rotation during InAs deposition: single dot emission at 1.55 µm was reported, but only in a narrow region of the sample [12].

Here we report on a new approach to obtain low density InAs QD structures grown on GaAs substrate suitable as single photon sources at 1.55 µm, relying on the deposition of sub-critical InAs coverages on relaxed InGaAs layers. The experimental work was based on the growth by Molecular Beam Epitaxy (MBE) and the characterization by Photoluminescence (PL) and Atomic Force Microscopy (AFM) of different structures with different compositions of relaxed InGaAs lower confining layers (LCLs) and upper confining layers (UCLs). The aim was not limited to the...
achievement of 1.55 µm emission from low density QD structures, but it included also the study of properties of the metamorphic system.

2. Experimental details

QD structures consist of: i) 100 nm of GaAs grown by MBE at 600 °C, ii) an In\textsubscript{0.15}Ga\textsubscript{0.85}As LCL of 500 nm deposited by MBE at 400 °C, iii) 5 nm of GaAs grown by ALMBE at 400 °C to smooth the surface, iv) an InAs layer of coverage \( \theta \) deposited by MBE at 490 °C followed by a Post Growth Annealing (PGA) of time \( \tau \) under As flux at 490 °C and v) an In\textsubscript{0.3}Ga\textsubscript{0.7}As UCL of 20 nm grown by ALMBE at 360 °C. Compositions of LCLs were \( x = 0.15 \) or \( x = 0.30 \), resulting in a mismatch between QDs and LCLs of 6.34 % and 5.19%, respectively: in the InAs/GaAs system the QD-CL mismatch is 7.16 %. These mismatch values were calculated on the basis of the Mareé theory of strain relaxation [13], experimentally confirmed by different techniques [14-15]. AFM measurements were performed on uncapped QD structures to study surface morphology and evaluate the QD density. PL characterization was performed by a 532 nm excitation at 10 K with a Fourier transform spectrometer with 1 meV resolution. Spectra were corrected for the response of the set-up.

3. Results and discussion

As reported in some works on QD growth on metamorphic buffers [16-18], the differences in mismatch, composition and surface morphology affect the evolution from the 2D growth regime to the nucleation of 3D nanosized islands: in particular the value of the critical coverage \( \theta \), for 2D-3D transition depends on the composition \( x \) of the InGaAs buffer layer. Henceforth, it was necessary to measure the value of \( \theta \), to derive the sub-critical coverages to be used for low density structures. \( \theta \) was found to be 1.8 ML and 2.2 ML for \( x = 0.15 \) and \( x = 0.30 \), respectively, by observing the transition from a RHEED streaky diffraction pattern to a spotty one. These values are larger as compared to critical coverage \( \theta = 1.6 \) ML for the deposition of InAs on GaAs.

![AFM images of uncapped QDs grown on In\textsubscript{0.15}Ga\textsubscript{0.85}As LCLs with (a) 1.8 ML InAs coverage and \( \tau = 0 \) s and (b) 1.65 ML InAs sub-critical coverage and PGA time \( \tau = 15 \) s.](image-url)

Figure 1 shows the AFM images of uncapped QDs grown on In\textsubscript{0.15}Ga\textsubscript{0.85}As LCLs with (a) 1.8 ML InAs coverage and \( \tau = 0 \) s and (b) 1.65 ML InAs sub-critical coverage and PGA time \( \tau = 15 \) s. An important feature is evident: the undulation of the surface underlying InAs islands, that is known to affect QD nucleation and positioning [17,19]. As can be seen from Figure 1 the growth of a sub-critical InAs coverage slightly affects the QD density that decreases from 4-5 x 10\textsuperscript{10} cm\textsuperscript{-2} for the \( \theta = 1.8 \) ML sample to 1-2 10\textsuperscript{10} cm\textsuperscript{-2} for the \( \theta = 1.65 \) ML one. Ultra-low QD density lower than 2-3 10\textsuperscript{8} cm\textsuperscript{-2} was achieved by further reducing the sub-critical coverage: AFM micrograph in Figure 2(a) shows the morphology of InAs QDs grown on In\textsubscript{0.15}Ga\textsubscript{0.85}As LCL with \( \theta = 1.5 \) ML and \( \tau = 22 \) s. Similar low densities were also obtained for InAs QDs grown on In\textsubscript{0.3}Ga\textsubscript{0.7}As LCL (Figure 2(b)). In this case the optimized values to obtain densities in the order of few 10\textsuperscript{8} cm\textsuperscript{-2} were \( \theta = 1.8 \) ML and \( \tau = 300 \) s. It is important to note that all the density data were deduced from the statistical analysis of larger scan area AFM images not reported here.
PL emission spectra of structures capped with In$_{0.15}$Ga$_{0.85}$As, shown in Figure 3, can be deconvolved in three components that, on the basis of the AFM studies, can be attributed to different families of QDs. The effects of different PGA times (that will not be discussed here) have been considered, resulting in a change of the density of QDs belonging to different families and in a broadening of their size distribution that, while little affecting emission energies, cause an increase of PL linewidth.

In Figure 3 we report also the PL emission spectra of structures with $x = 0.30$ capped with In$_{0.30}$Ga$_{0.70}$As: clear contributions of different QD families are not evident as in the $x = 0.15$ case, possibly due to increase of dishomogeneities in the dimensions of island families, as seen in AFM images, and the emission wavelength is redshifted to 1.3 $\mu$m. As widely discussed in [11], when the QD-CL mismatch is reduced, the QD strain is decreased, causing a lowering of the QD energy gap. This effect adds up to the energy decrease due to the reduction of QD-CL band discontinuities and results in a redshift of the emission energy. To extend the emission wavelength towards 1.55 $\mu$m, instead of increasing $x$ and incur in worsening of the surface morphology, we opted to increase the UCL composition $y$ beyond the value of 0.30, as it was done in pseudomorphic structures [5-6].

In this way the QD emission was further redshifted, as shown in Figure 3, up to and even beyond 1.55 $\mu$m, at the expenses of a reduction of the emission intensity, in particular when $y = 0.60$ is used. The wavelength redshift may be due to: i) the lowering of UCL-QD band discontinuities, ii) the change in QD size and/or composition due to QD-cap intermixing effects and iii) a reduction of the

Figure 2. AFM images of (a) InAs/In$_{0.15}$Ga$_{0.85}$As QDs with $\theta = 1.5$ ML and $\tau = 22$ s and (b) InAs/In$_{0.30}$Ga$_{0.70}$As QDs with $\theta = 1.8$ ML and $\tau = 300$ s.

Figure 3. PL spectra of metamorphic low density QD structures at 10K for different $x$ values of the In$_x$Ga$_{1-x}$As LCL and $y$ values of the In$_y$Ga$_{1-y}$As UCL.

In this way we can extend the emission wavelength towards 1.55 $\mu$m, at the expenses of a reduction of the emission intensity, in particular when $y = 0.60$ is used. The wavelength redshift may be due to: i) the lowering of UCL-QD band discontinuities, ii) the change in QD size and/or composition due to QD-cap intermixing effects and iii) a reduction of the

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QD strain. It is interesting to note how the width of the PL emission reduces when values of y larger than x are used, an observation that could hint to interaction processes between QDs and UCLs that affect the homogeneity of the QD ensemble. Finally, it can be appreciated how by changing the composition y of the UCLs the emission wavelength can be tuned in the whole 1.3 - 1.6 µm range, while keeping the same QD growth scheme and the same metamorphic LCL parameters.

4. Conclusions
In conclusions, we grew by MBE low density QD structures on GaAs with emission tunable at 1.3 and 1.55 µm, by depositing sub-critical InAs coverages on InₓGa₁₋ₓAs metamorphic LCLs. The relevant parameters for this design have been studied, namely the sub-critical coverage and the PGA time: the peculiarities of the metamorphic system were put in evidence, in particular the different 2D-3D critical coverages and the undulated surface morphology. Values of relevant parameters to obtain QD density in the order of few QDs / µm² were found for x = 0.15 and 0.30. We showed how, by choosing suitable values of x and y, the PL emission wavelength can be tuned in the whole range 1.3 - 1.6 µm.

In particular, for x = 0.30 and y = 0.60 an emission at 1.64 µm was reached, a very high value for nanostructures grown on GaAs. We believe these results demonstrate that the metamorphic approach can be very valuable to obtain nanostructures for single photon sources. Moreover, this design has the potentiality for tuning emission and other single dot properties, by applying the so-called Quantum Dot Strain Engineering approach to low density structures.

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