InGaAs quantum dot structures grown in GaAs barrier by metal organic chemical vapor deposition for high efficient long wavelength emission

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ABSTRACT

In this work we present a method to obtain room temperature ground state emission beyond 1.3 μm from InGaAs QDs, grown by MOCVD, embedded directly into a binary GaAs matrix. The wavelength is tuned from 1.26 μm up to 1.33 μm by varying the V/III ratio during the growth of the GaAs cap layer, without using seeding layer or InGaAs wells. A line-shape narrowing (from 36 meV to 24 meV) and a strong reduction of the temperature dependent quenching of the emission (down to a factor 3 from 10K to 300K) are observed, that represent the best value reported for QD structures emitting at 1.3 μm .

The results are explained in term different morphological evolution and surface reconstruction undergone by the InGaAs islands during the GaAs overgrowth that result in larger QD size and in lower In-Ga intermixing. Indeed, cross sectional TEM images show an increase in the QD size of more than 30% with decreasing the AsH_3 flow.

The overall strain reduction due to the use of the GaAs matrix allows the fabrication of highly efficient staked QD layers. The single and multiple QDs samples show a systematic increase of the emission intensity and similar spectral shape.

Keywords: Quantum dots, 1.3 μm , InGaAs, MOVPE

1. INTRODUCTION

The fabrication of quantum dots (QDs) by selfassembled growth methods has been intensively investigated in the last decade for basic physics and device applications^{1,2}. In recent years, self-organized In(Ga)As/GaAs QDs have been demonstrated as a means of extending the emission wavelength of a GaAs-based system to the important telecommunication wavelengths of 1.3 and 1.55 μm ^{3,4}. The development of a 1.3 μm InAs/GaAs QD laser has progressed rapidly and devices operating at room temperature with high output power, high characteristic temperature and an extremely low threshold current density have been demonstrated by Molecular Beam Epitaxy (MBE)^{5,6,7,8}. However, long-wavelength lasing (beyond 1.3 μm) in QD laser structures grown by metalorganic chemical vapor deposition (MOCVD) has not been reported so far, and few works have shown emission wavelength at 1.3 μm in QDs fabricated by MOCVD^{9, 10, 11}.

Amongst the different approaches used in order to extend the photoluminescence emission of InGaAs QDs beyond the 1.3 μm spectral region, the insertion of the QDs in ternary compounds was demonstrated to be the most effective one, by both MBE and MOCVD growth techniques^{9, 11, 12, 13}. However, the introduction of the InGaAs barrier still presents some problems. Even if the extension of the wavelength range in the In(Ga)As/InGaAs samples could be obtained with a relative small increase of the overall strain of the structure, a GaAs spacer layer with a correct thickness (several tens of nm) must be inserted between the dot layers in order to stack more than one QDs layer, without introducing extended defect in the structures. Due to the strain accumulation in the spacer layer, a continuous decrease

of the QD density is often found in the subsequent layers, together with increased size dispersion^{11, 14}. This increased overall strain of the structures and the progressive reduction of QD density limit the possibility to increase the maximum gain in QD active device, where optical density as high as possible is required.

The epitaxial growth of InGaAs QDs with emission beyond 1.3 μm directly embedded in a GaAs matrix (i.e. with no indium in the barriers) overcomes this limit. This was achieved with a growth method that explores the kinetics of the QD surface reconstruction during the GaAs overgrowth. In our previous work¹⁵ we demonstrated that the properties of the InGaAs/GaAs QD structures are finely tuned by using kinetic effects associated to different growth conditions, obtaining an excellent control of the structural and morphological properties of the QD layer. The optimization of the growth of the InGaAs/GaAs QDs resulted in a high emission efficiency at room temperature (RT), with a maximum wavelength not exceeding 1.26 μm .

By the simple modification of the InGaAs growth conditions, the emission wavelength of QD directly grown on GaAs can be tuned only up to 1.29 μm . Instead, by exploring the kinetics of the QD surface reconstruction during the GaAs overgrowth, we have obtained room temperature long wavelength emission from InGaAs QDs, growth directly into a binary GaAs matrix. The wavelength is tuned from 1.26 μm up to 1.33 μm by varying the V/III ratio during the growth of the GaAs cap layer, without using a seeding layer or InGaAs wells. Strong improvements in terms of the line-shape narrowing and room temperature efficiency are obtained (an increase of about a factor of 2 is found in the RT PL intensity as compared to the sample grown in InGaAs matrix¹⁶). Moreover, the temperature dependent quenching decreases only by a factor of 3 between cryogenic (10K) and room temperature, as opposed to a factor of 9 found for our previous samples and a factor of 7 found for QDs grown in an InGaAs matrix¹⁷. The reduced overall strain due to the lack of In in the barrier, allows the growth of high uniform QD stacked samples. The QDs are nearly perfectly reproduced in the subsequent layers, regardless the number of the layers, provided that the correct temperature is employed for the growth of the GaAs barrier layers. Samples containing up to 6 layer of QD shows efficient emission at 1.3 μm , preserving the same line-shape and the same full width half maximum (FWHM).

2. EXPERIMENTAL

The structures were grown on (100) GaAs substrates by low-pressure metal organic chemical vapor deposition (MOCVD) in an AIXTRON 200 AIX system equipped with a rotating substrate holder, at 20 mbar, with TMGa, TMIIn solution and pure AsH₃ as source materials. Palladium purified H₂ with a flow rate of 7 slm was used as a carrier gas. After the deposition of a 200 nm undoped GaAs buffer layer growth at 750 °C, the QDs were formed by depositing 4 MLs of In_xGa_(1-x)As with a nominal In content of $x = 0.5$ at the growth temperature of 550 °C and growth rate of 1 ML/s. This conditions allows us to obtain single QD layers having excellent structural and morphological properties, with a mean island height and base diameter of around 7 nm and 40 nm, respectively. The island density can be easily controlled from 10^9 cm^{-2} up to around $4 \times 10^{10} \text{ cm}^{-2}$, with a low size dispersion (around 7%), as shown by Fig. 1, where a representative Atomic Force Microscopy (AFM) image of the uncovered samples is shown.

Fig. 1. Representative AFM image of uncovered QDs samples.

The islands are covered by a 30 nm GaAs cap layer. The overgrowth was started after a growth interruption of 90 s under arsine stabilization at the same growth temperature used for the QD layers. During the growth interruption and for the growth of the GaAs cap layer, the AsH₃ partial pressure (P_{AsH_3}) was varied between 1.4×10^{-1} and 5.7×10^{-1} mbar. The QD layer was grown under identical growth condition for all the samples; only the AsH₃ partial pressure was changed for the capping procedures.

The structural properties of the samples were investigated by Cross sectional Transmission Electron Microscopy (TEM) on QD samples overgrowth by a 30 nm thick GaAs capping layer. The Photoluminescence (PL) measurements were performed by using an Ar⁺ laser tuned at 514.4 nm in the temperature range 10K - 300K. The signal was spectrally analysed by a 1 m monochromator and detected by a Ge detector, cooled by liquid nitrogen.

3. RESULTS AND DISCUSSION

3.1 Optical and structural properties

The effect of the different growth conditions used to grow the cap layer on the QD optical properties is clearly visible in Fig. 2a, where the room temperature PL spectra of 4 samples (A, B, C and D) are reported. In all the samples the QD layers were grown under identically growth conditions and they are overgrown with 30 nm of GaAs deposited with AsH₃ partial pressure of 5.7×10^{-1} , 4.3×10^{-1} , 2.8×10^{-1} and 1.4×10^{-1} mbar, respectively.

Fig. 2. (a) Normalized room temperature PL spectra of samples A, B, C and D overgrown with AsH₃ partial pressure of 5.7×10^{-1} , 4.3×10^{-1} , 2.8×10^{-1} and 1.4×10^{-1} mbar, respectively. (b) Room temperature PL spectra with increasing the excitation intensities of sample D

By decreasing the V/III ratio during the growth of the GaAs cap layer, the emission wavelength undergoes a significant red shifts. The ground state peak position starts at 1260 nm (the typical peak position of our QDs grown on GaAs) for the sample grown with the highest P_{AsH_3} (sample A), and increases nearly linearly up to 1330 nm in the sample grown with the lowest P_{AsH_3} (sample D). All spectra show multiple emission peaks which are attributed to the ground level (E_0) and the first (E_1) excited state transition of the QDs, as confirmed by the state filling dynamic obtained with increasing the excitation power (shown in Fig. 2b for sample D).

In Fig. 3 we plot the full width half maximum (FWHM) measured for the spectra of fig. 2a, as a function of the AsH₃ partial pressure used in the capping procedure. The plot clearly shows that with increasing wavelength a systematic reduction of the emission spectral width occurs (from 35 meV for sample A down to 24 meV for sample D), indicating that the highest uniformity in the QD distribution is achieved in samples overgrown with lower AsH₃ partial pressure, despite the QD layer itself was grown under identical growth conditions in all samples.

Fig. 3. Full width half maximum measured for the spectra of Fig. 2a as a function of the AsH₃ partial pressure.

The red shift observed in our spectra can be justified only by an increase in the In content inside the dot and/or an increase of the island size, induced by the capping procedure. Indeed, an increase in the QD size is demonstrated by the cross sectional TEM investigation carried out on the studied samples. Fig. 4a and Fig. 4b show the cross sectional TEM image of the samples A and D, respectively. Even though the precise dimensions could not be determined with high accuracy, because the complex image contrast is affected by strain, composition and sample thickness, an roughly measurement of the island dimensions shows an increase of the dot height from 4.7 nm (sample A) up to around 6.7 nm for the sample overgrown with the lower P_{AsH₃} (sample D) and, correspondently, the dot diameter goes from around 15 nm up to 21 nm, as shown by the plot of Fig. 4c, where the measured dimensions of the islands are reported as a function of the AsH₃ partial pressure employed in the capping procedure.

Fig. 4. (a) Cross sectional Transmission Electron Microscopy image of the InGaAs QDs overgrown with P_{AsH₃} = 1.4 × 10⁻¹ mbar and (b) InGaAs QDs overgrown with P_{AsH₃} = 5.7 × 10⁻¹ mbar. (c) Measured InGaAs island dimensions as a function of the AsH₃ partial pressure employed in the capping procedure.

Beside the wavelength shift and the line-shape narrowing, another important effect obtained by decreasing the V/III ratio in the capping procedure consist in the significant reduction in the temperature dependence of the integrated intensity in the sample overgrown with the lower P_{AsH_3} . The ratio between the low temperature (10 K) and room temperature radiative efficiency ($\eta_{\text{LT}}/\eta_{\text{RT}}$) measured for the spectra of fig. 2a, as a function of the AsH_3 partial pressure used in the capping procedure is plotted in Fig. 5a. As clearly shown in the plot, the quenching of the RT ground state emission goes from factor 9 in sample A, down to 3 in sample D. To our knowledge, this represents the best value reported for QDs emitting at 1.3 μm , and indicate a lower density of nonradiative recombination centres in sample D. The studies of the temperature carrier dynamic carried out in sample D is shows in Fig. 5b, where the PL spectra recorded at different temperatures is reported.

Fig. 5. (a) ratio between the low temperature (10 K) and RT temperature radiative efficiency as a function of the AsH_3 partial pressure. (b) Photoluminescence spectra at various temperatures T . The inset shows the integrated PL intensity versus $1/KT$ obtained from the ground state emission

Together with the ground state and the first excited state transitions, the wetting layer (WL) related emission is observed at around 904 nm and for temperatures lower than 110 K. The temperature dependence of the integrated intensity is shown in the inset of Fig. 5b for the ground state and the WL emissions. With increasing temperature, the integrated PL intensity of the ground state emission increases reaching its maximum intensity at 110 K. Correspondingly, the WL intensity decreases and then vanishes around 110 K. This can be explained in terms of carrier trapping at low temperatures into the WL: the increase of temperature up to 110 K induces thermal activation of carriers trapped in the WL region, which are captured into the dots, resulting in the enhancement of the integrated PL intensity of the QDs states. Above 110 K, carriers from the QDs, are thermally activated, resulting in an intensity PL quenching of only a factor of 3.

3.2 Growth model

Previous studies¹⁸ have shown that significant modifications of the island size, shape and In distribution can be induced in the QD by surface reconstruction during the GaAs overgrowth. Moreover, a relevant blueshift and a narrow FWHM have been found to result from the capping¹¹. This effect is often ascribed to Ga and In strain induced intermixing with the barrier material. The surface reconstruction strongly depends on the group III adatom surface mobility that, in turn, is controlled by the growth conditions, like growth temperature, growth rate and V/III ratio. Due to the complex surface structure resulting from the growth of the InGaAs islands, an important variation of the local GaAs growth rate is expected to occur during the capping procedure. An important contribution for the change in the GaAs local growth rate arises also from variations of the strain field associated to the In distribution inside and around the QDs. When the deposition parameters allow long enough surface diffusion length of the Ga adatoms (as obtained by employing low V/III ratio) the GaAs will grow first on the energetically favorable sites, i.e. on the wetting layer (100 surface and lower local strain¹⁹) and around the edge of the island. Overgrowth on the topmost QD surface, which is the most unfavorable growth site for GaAs (because of the higher local strain), will begin only at a later stage of the growth. Actually, high resolution compositional analysis carried out by electron energy-loss spectroscopy (EELS) and scanning transmission electron microscope (STEM) have shown a substantial local depletion of In from the wetting layer behind and in the vicinity of the QDs (30% lower on average than the In concentrations away from the dot), together with a relevant In enrichment inside the islands²⁰. Moreover, an increase in the atomic spacing from the QD/bulk interface to the QD surface was found in uncovered QD samples¹¹. The delayed GaAs growth on the top of the island prevents the size reduction of the QDs, as found in the overgrowth of the MBE samples¹⁸. On the other hand, in samples overgrown with high V/III ratio, the reduced surface diffusion length of Ga adatoms strongly reduces the local growth rate differences, thus allowing the GaAs growth directly on the top of the QD. The earlier island coverage enhances the In-Ga intermixing reducing both the size and the In mole fraction of the dot, with a consequent increase of the ground state emission energy of the samples.

As a result, two effects should be invoked to explain the behavior found by changing the V/III ratio in the cap layer growth: reduced In-Ga intermixing at the dot-barrier interface and island size variation. The increase of the QD dimensions with reduced V/III ratio is consistent with the cross sectional TEM analysis showed in Fig. 4. A reduced In-Ga intermixing should lead to QDs/barrier sharper interfaces with a consequent deeper confining potential that reduces the carrier thermal escape from the QD electronic states (thus increasing the RT efficiency). This effect is indeed clearly observed in Fig. 5, where a systematic increase of the quenching factor is found with decreased AsH₃ partial pressure. Moreover, reduced In-Ga intermixing was reported to decrease the FWHM of the optical emission^{21, 22}, due to the better compositional uniformity of the dot ensemble.

3.3 Growth of stacked samples

The overall strain reduction due to the use of the GaAs matrix allows the straightforward fabrication of highly efficient vertically stacked QD layers. The studies of the staking properties were carried out on samples containing from 1 to 6 layers of QDs, separated by 30 nm of GaAs barrier. Each sample is terminated with a further layer of uncovered QDs in order to allow the structural characterization of the subsequent layers by AFM measurements. In order to obtain an excellent reproducibility of the QD dimensions and density with increasing the number of the stacked layer layers, the growth of the GaAs barrier must be performed at higher growth temperature with respect the growth of the QDs. Actually, AFM image of the topmost uncovered QD layers (not shown) evidenced a significant reduction of the island density and height, together with an enlarged dot diameter, for growth temperature of the barrier layers lower than 650 °C. By growing the GaAs barrier at 650 °C, a nearly perfect replica of the first QD layer is obtained in the subsequent growth, regardless the number of layers. This effect could probably be ascribed to surface undulation and strain modulation of the spacer layer induced directly by the strain field of the buried dots. At higher growth temperature the GaAs undergoes a better surface reconstruction, with the effect of removing the surface disuniformity, as found in reference 23 and 24 where the same effect was obtained by using annealing step after the growth of the barrier.

Fig. 6 shows the room temperature PL spectra recorded at low excitation intensity for samples containing from 1 up to 6 layers of QDs. The emissions of the vertically stacked QDs samples show the same structures of the corresponding single layer, but with a systematic increase of the ground state intensity with the number of stacked layers. All the spectra retain the same line-shape with about the same FWHM in all the samples. No additional peaks are evidenced at low temperature, demonstrating that each QD layer is well reproduced without changing its size distribution. Together with the increase of the intensity, a systematic wavelength shift is evidenced by the spectra shown in Fig. 6. The ground state emission starts from $\lambda = 1335$ nm from the single layer sample and decreases progressively down to $\lambda = 1311$ nm for the sample containing 6 layers of QDs. Because the FWHM of the samples remained

unchanged with the stacking (demonstrating an uniform size distribution between the layers), the wavelength shift can not be ascribed to the size reduction of QDs in the subsequent layers. More likely, the observed blueshift is a consequence of the higher temperature used for the growth of the GaAs barriers, that increase the energy emission of the QDs by annealing effect.

Fig. 6. Room temperature PL spectra recorded at low excitation intensity of samples containing from 1 up to 6 layers of QDs.

4. CONCLUSIONS

In conclusion, we demonstrated emission beyond 1.3 μm from MOCVD grown InGaAs QDs embedded in GaAs matrix, by exploring the kinetics of the QD surface reconstruction during the GaAs overgrowth. Red-shift of the emission wavelength from 1.26 μm up to 1.35 μm were obtained by reducing the V/III ratio for the growth of the GaAs cap layer. This effect is explained in term of reduced In-Ga intermixing at the dot-barrier interface and island size variation induced by the different surface reconstruction and morphological evolution undergone by the InGaAs islands during the growth of the GaAs cap. Moreover, the reduced In-Ga intermixing is thought to leads to narrow line-shape and, more important, to a relevant reduction (down to a factor of 3) of the quenching factor for the RT ground state emission, that represent, to our knowledge, the lowest value ever reported for QD structures emitting at 1.3 μm .

The overall strain reduction due to the use of the GaAs matrix allows the fabrication of highly efficient stacked QD layers. The single and multiple QDs samples show a systematic increase of the emission intensity and similar spectral shape, retaining about the same FWHM (lower than 24 meV). Only a slightly decreasing in the wavelength emission is found with increasing number of stacked layers, due to the annealing effect undergoes by the samples during the higher temperature growth of the GaAs barrier layers.

5. ACKNOWLEDGEMENTS

This work was supported by the National Project “FIRB Nanoelettronica”. The authors would like to acknowledge the financial support of Agilent Technologies and Mr. G.M. Epifani for his valuable technical assistance

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