

# Optimization of InGaAsN(Sb)/GaAs quantum dots for optical emission at 1.55 $\mu\text{m}$ with low optical degradation

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## A B S T R A C T

Low optical degradation in GaInAsN(Sb)/GaAs quantum dots (QDs) p-i-n structures emitting up to 1.55  $\mu\text{m}$  is presented in this paper. We obtain emission at different energies by means of varying N content from 1 to 4%. The samples show a low photoluminescence (PL) intensity degradation of only 1 order of magnitude when they are compared with pure InGaAs QD structures, even for an emission wavelength as large as 1.55  $\mu\text{m}$ . The optimization studies of these structures for emission at 1.55  $\mu\text{m}$  are reported in this work. High surface density and homogeneity in the QD layers are achieved for 50% In content by rapid decrease in the growth temperature after the formation of the nanostructures. Besides, the effect of N and Sb incorporation in the redshift and PL intensity of the samples is studied by post-growth rapid thermal annealing treatments. As a general conclusion, we observe that the addition of Sb to QD with low N mole fraction is more efficient to reach 1.55  $\mu\text{m}$  and high PL intensity than using high N incorporation in the QD. Also, the growth temperature is determined to be an important parameter to obtain good emission characteristics. Finally, we report room temperature PL emission of InGaAsN(Sb)/GaAs at 1.4  $\mu\text{m}$ .

### Keywords:

A1. Crystal morphology  
A1. Nanostructures  
A3. Molecular beam epitaxy  
A3. Quantum dots  
B2. Semiconducting III-V materials  
B3. Light emitting diodes

## 1. Introduction

Modern telecommunication systems widely use semiconductor diode lasers based on expensive and low yield phosphide compounds emitting at 1.3 and 1.55  $\mu\text{m}$ . Therefore, huge research efforts have been dedicated to develop alternative fabrication techniques to grow epitaxial materials on inexpensive large-area GaAs substrates. In recent years, a special interest has been dedicated to quantum dots structures due to their interesting properties such as the higher carrier localization. In particular, GaInNAs QDs are promising for application in 1.3 and 1.55  $\mu\text{m}$  laser diodes, since introducing low mole fractions of N (< 5%) to GaInAs, predominantly affects the conduction band leading to large conduction band offset.

Actually, GaInNAs QDs have been successfully grown by molecular beam epitaxy (MBE) following the Stranski-Krastanow mechanism. The emission of these samples covers a wide wavelength range from 1.04 to 1.52  $\mu\text{m}$  by varying the N content [1]. However, increasing the N mole fraction results in a degradation of the PL intensity of the GaInNAs QDs by 2–3 orders of magnitude at the longer wavelengths. Other authors [2–4] report GaInNAs QD laser devices operating below 1.2  $\mu\text{m}$ . Consequently, a strong

improvement of the quality of the layers is still necessary to obtain high yield LED and laser devices working at 1.55  $\mu\text{m}$ .

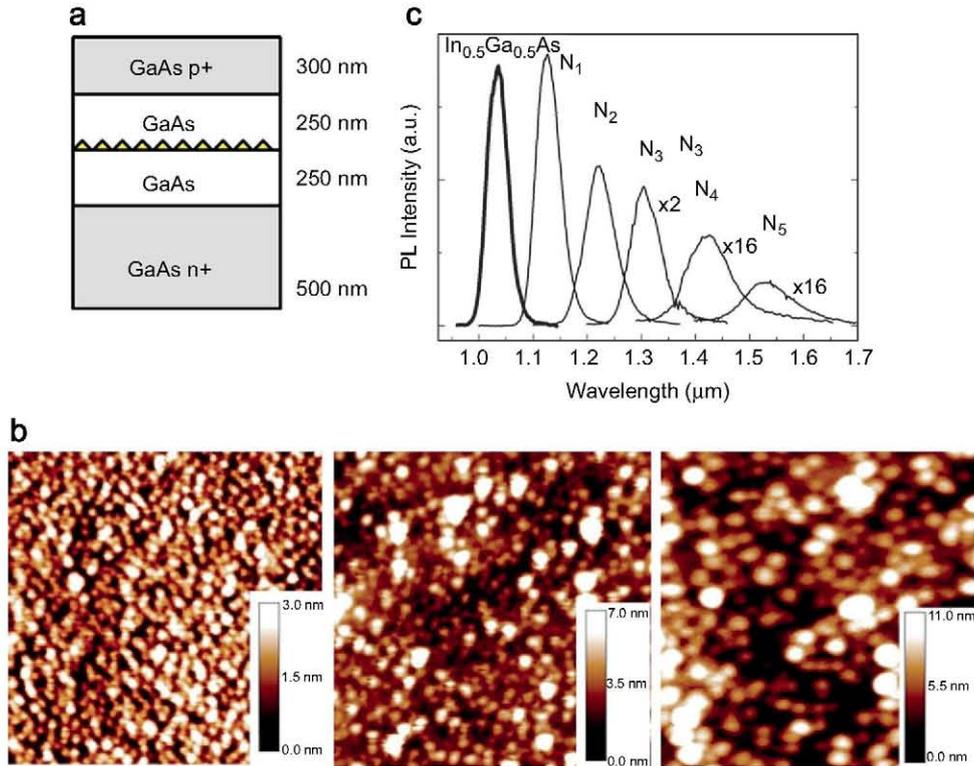
In this work, we directly focus on samples emitting at 1.55  $\mu\text{m}$ . We carry out an optimization of the PL intensity by means of varying different parameters such as: quantum dot growth temperature, nitrogen content and antimony incorporation, as well as post-growth rapid thermal annealing treatments. We observe that the quantum dot growth temperature and nitrogen content are directly related, since the amount of nitrogen incorporated in the QD is higher for lower growth temperature. Besides, the incorporation of Sb extends the emission wavelength towards longer wavelengths. Following the experiments previously performed in (Ga,In)(N,As,Sb) quantum wells [5,11], we achieve room temperature emission in QD p-i-n structures.

Finally, we perform a comparative study of PL characteristics for the different set of growth conditions and established a procedure for future development of emitters working at RT and at 1.55  $\mu\text{m}$ .

## 2. Experiment

The samples are grown on semi-insulating GaAs (1 0 0) substrates in a RIBER 32, solid source molecular beam epitaxy (MBE) system equipped with a RF Oxford Applied Source to supply the atomic nitrogen. The structure of these samples is shown in Fig. 1a. Firstly, we grow a 500 nm Si doped ( $2 \times 10^{18} \text{ cm}^{-3}$ ) GaAs buffer layer on a GaAs substrate at 590  $^{\circ}\text{C}$  and a rate of 1 ML/s. Then an

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**Fig. 1.** (a) Structure of the QD p-i-n samples grown in this work, (b) ( $1 \mu\text{m} \times 1 \mu\text{m}$ ) AFM images of  $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$  QDs (left),  $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}_{0.97}\text{N}_{0.03}$  (center) and  $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}_{0.96}\text{N}_{0.04}$  under Sb flux (right) and their corresponding surface densities are  $7 \times 10^{11}$ ,  $5 \times 10^{10}$  and  $2 \times 10^{10} \text{ cm}^{-2}$ , respectively and (c) low temperature photoluminescence (15 K) of a series of samples with different N concentrations.

intrinsic 250 nm GaAs layer is grown on top. Then, we drop the substrate temperature to  $430 \text{ }^\circ\text{C}$  for the growth of the GaInN(Sb)As QDs. The equivalent amount of deposited material and the growth rate employed are 4ML and 0.15 ML/s, respectively. The N plasma source conditions (nitrogen flux and RF power) are set in each growth depending on the amount of active nitrogen needed. This active N is measured by means of an optical emission detector (OED) tuned to the main optical transition of the N plasma. We use 5 different N concentrations, with OED values increasing from samples N1 to N5, as well as different Sb concentrations in the experiments, which will be described later in detail. Once the QDs are formed, we dropped the temperature  $130 \text{ }^\circ\text{C}$  below the QD growth temperature in order to reduce the possible In segregation helping the QDs to keep their shape and composition. In Fig. 1b we show the AFM images of QDs without and with antimony, which correspond to figures a and b, respectively [12,13]. Further, a 50 nm GaAs capping layer was grown at  $430 \text{ }^\circ\text{C}$  and 1 ML/s. Then, the substrate temperature was raised up to  $590 \text{ }^\circ\text{C}$  and a 250 nm GaAs layer was grown prior to the 300 nm Be doped ( $2 \times 10^{18} \text{ cm}^{-3}$ ) GaAs top contact. Finally, an additional layer of QDs was grown on the surface of the sample using the same conditions employed for the buried ones to carry out structural characterization by means of AFM operating in tapping mode. For the PL measurements, the samples were mounted in a closed-cycle helium cryostat and excited using a 781 nm laser diode. A liquid nitrogen-cooled germanium detector was used to detect the emission using conventional lock-in techniques.

### 3. Results and discussion

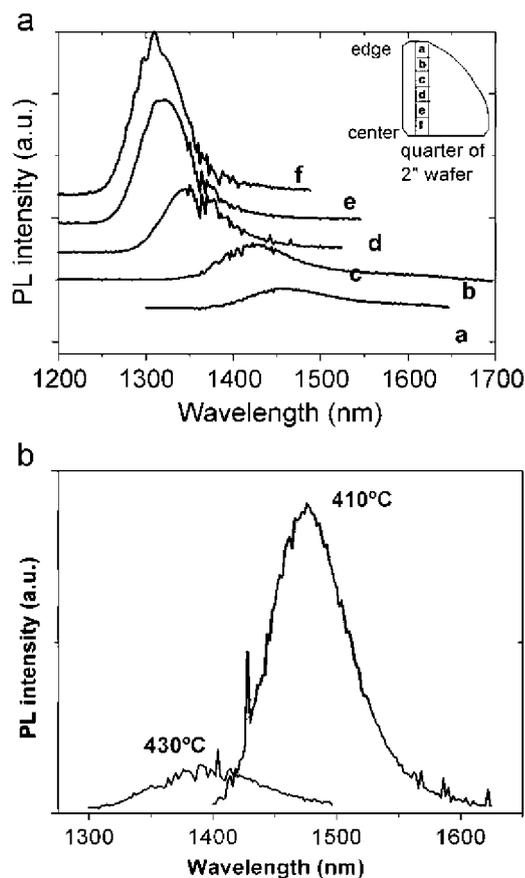
In a previous work [6], we observed how different ratios of Ga/In composition give rise to different structural and optical properties. The configuration of 50% Ga and 50% In ( $\text{Ga}_{0.5}\text{In}_{0.5}\text{NAs}$ ) provides

homogenous QDs layers yielding narrow optical emissions. Since an increment in the nitrogen content results in a subsequent redshift in the wavelength emission, it is possible to tailor the operating characteristics of the device by controlling the nitrogen percentage in the QDs. In fact, by increasing the nitrogen content up to 4%, a  $1.55 \mu\text{m}$  emission is obtained (see Fig. 1c). This N concentration was estimated by previous studies made using QW and RTA studies [6]. Thanks to the growth procedure described in the previous section, it is possible to obtain narrow PL peaks at low temperature with only one order of magnitude of degradation from pure InGaAs QD samples to samples with 4% of N.

With respect to the optimization of the optical emission at  $1.55 \mu\text{m}$ , we show three different experiments. The first one was focused on the influence of growth temperature in the optical properties of the QD layers; the second one studied how the post-growth annealing treatment affects the enhancement of PL emission. Finally, in the last one we analyze the benefits of Sb in the optical quality of GaInNAs QDs

#### 3.1. Influence of QDs growth temperature

All the samples described in this paper were grown in a quarter of a  $2''$  wafer. By studying the PL emission at low temperature from different parts of the sample labeled as N3, we obtained a strong variation in both the PL intensity and the wavelength emission as a function of the different positions in the wafer (see Fig. 2a). Previous studies in the growth of InGaAsN quantum wells [7] showed that there is an unintended temperature gradient in the substrate holder during the growth due to the MBE filament heater inhomogeneity. This temperature difference was estimated to be  $25 \text{ }^\circ\text{C}$  from the center to the edge of the sample observing the desorption temperature of the native oxide by RHEED in several parts of the wafer. Since the N incorporation is strongly dependent



**Fig. 2.** (a) Low temperature photoluminescence spectra taken at different points along the wafer. The temperature gradient during the growth leads to different N incorporations and (b) PL emission of two samples grown at different temperatures. The lower growth temperature improves the N incorporation.

on the growth temperature [8], the measured gradient leads to a different N incorporation along the wafer.

From this study, we conclude that the wavelength emission suffers a clear redshift when the growth temperature decreases. Consequently, it is expected that a lower growth temperature could help to achieve a 1.55  $\mu\text{m}$  emission. However, together with this redshift there is also a significant reduction in the PL intensity. This is normally overcome by rapid thermal annealing treatments after the growth. In our case, Fig. 2b shows the PL intensity of two samples grown under similar conditions except on the growth temperature of the QD layer (one was grown at 430 and the other at 410  $^{\circ}\text{C}$ ). Both samples were annealed at 800  $^{\circ}\text{C}$  for 30 s. By using such growth and annealing conditions, we are able to reach emission at 1.48  $\mu\text{m}$  with a PL intensity enhancement by a factor of 2.

### 3.2. Influence of the annealing temperature

Fig. 1c shows the wavelength redshift due to the different nitrogen content in the QD. It is therefore evident that by controlling the nitrogen mole fraction in the QDs, the wavelength emission could be tailored. In fact, we report clear PL emission at 1.55  $\mu\text{m}$  for sample N5, with an approximate N content of 4%. Nevertheless, as we mentioned in Section 3.1, the increment in wavelength as a consequence of N incorporation, normally leads to a strong reduction in the PL intensity. In year 2000, Sopanen et al. [1] reported a degradation in the PL intensity of GaInNAs QDs by 2–3 orders of magnitude at the longer wavelengths. However, using the growth procedure described in Section 2, we obtained a

degradation in the PL intensity as low as 1 order of magnitude from 1.1 to 1.5  $\mu\text{m}$  wavelength emission.

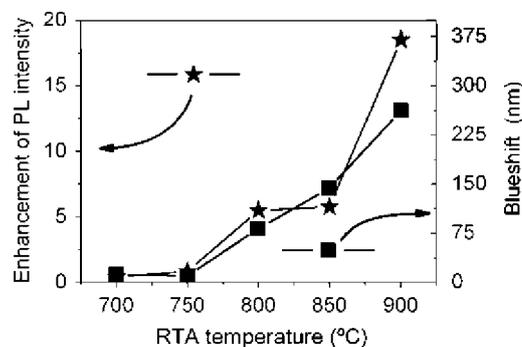
On the other hand, it has been reported that nitrogen incorporation introduces point defects in GaInNAs QW structures leading to the existence of non-radiative recombinations, which can explain the PL intensity degradation. By annealing the samples, the crystal quality is normally improved, resulting in an enhancement of the PL intensity together with a blueshift of the wavelength peak. The origin of this blueshift in QD of GaInNAs has been studied by the authors in Ref. [6]. It is important to optimize the annealing conditions for quantum dots also, in order to obtain the maximum optical intensity with the lowest blueshift.

In our case, due to the strong inhomogeneity found in the samples (as mentioned before), we needed to study relative variations between the RTA and the as-grown samples measuring each piece before and after the RTA treatment. In Fig. 3, we show the relative shift and the enhancement of the PL intensity peak as a function of the RTA temperature (The PL enhancement is calculated as the ratio of both intensities). Although the higher increment of PL intensity is indeed reached at 900  $^{\circ}\text{C}$ , the blueshift for this temperature is also very high (370 nm). However, in the interval between 800 and 850  $^{\circ}\text{C}$ , the PL enhancement is constant and the blueshift can be reduced to 90 nm. The final temperature employed must be taken as a tradeoff between the desired redshift and the efficiency of the PL intensity we need. In our experiments, we used 800  $^{\circ}\text{C}$  since it allows the samples to show emission at 1.55  $\mu\text{m}$ .

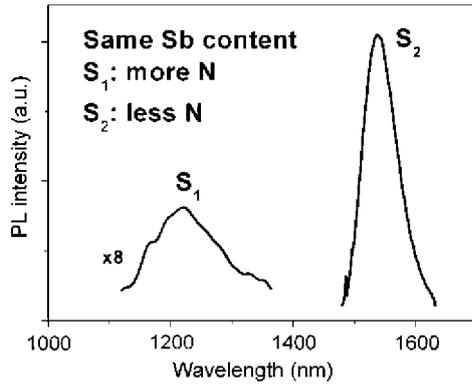
### 3.3. Influence of Sb incorporation

Recent studies concerning InGaAsN quantum wells reveal that the incorporation of Sb to the crystal lattice extends the emission wavelengths toward longer wavelengths [9]. Besides, this element seems to improve the as-grown quality of the dilute nitrides, and actually, (Ga,In)(N,As,Sb) QW were utilized for the successful achievements of room temperature continuous-wave operating lasers emitting at 1.55  $\mu\text{m}$  [10]. Preliminary AFM studies point to the fact that Sb helps to decrease the surface density and to increase the QD size (see Fig. 1c).

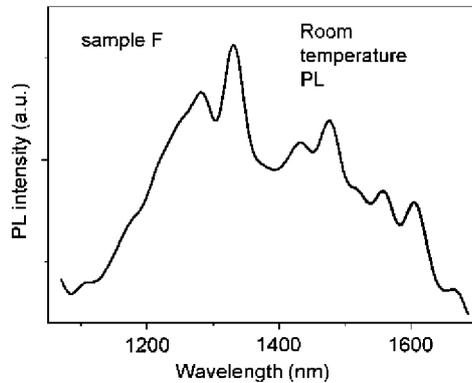
In this experiment, we grew two new samples with the same mole fraction of Sb ( $\sim 5\%$ ), but different nitrogen contents (samples S1 and S2, in Fig. 4, with  $\sim 4$  and  $\sim 2\%$  N, respectively). In this case, we obtained both an improvement in PL intensity and a higher peak wavelength in the sample containing less nitrogen. We can assume that the incorporation of Sb gives rise to a redshift in the wavelength emission improving the PL characteristics at the same time. Therefore, it is possible to reach the wavelength of interest using less mole fractions of N and consequently reducing the



**Fig. 3.** Effect of post-growth rapid thermal annealing on the blueshift and the PL enhancement of the QD samples. Although the PL improves up to 900  $^{\circ}\text{C}$ , the interval between 800 and 850  $^{\circ}\text{C}$  leads to constant enhancement in the PL of 5 together with moderate blueshift. All the points shown in the graph correspond to relative improvements between as-grown and RTA on the same piece of the sample.



**Fig. 4.** PL intensity of two samples with the same Sb content and different N mole fractions. Antimony clearly produces a redshift in the wavelength. The improvement in the PL intensity can be due to the less N mole fraction incorporated in sample S2.



**Fig. 5.** Room temperature PL emission of a QD sample (sample F) with  $\sim 10\%$  Sb and  $\sim 2\%$  N, after a post-growth annealing at  $800\text{ }^\circ\text{C}$ .

damage due to the non-radiative recombinations associated with the point defects. In this figure, a clear peak centered at  $1.55\text{ }\mu\text{m}$  with a linewidth of  $80\text{ nm}$  can be shown.

After these series of experiments, we obtained several samples with good PL emission in  $1.55\text{ }\mu\text{m}$  at  $15\text{ K}$ . However, further studies of the PL with temperature showed that the luminescence quenches at temperatures in between  $90$  and  $120\text{ K}$  in all the cases. Nevertheless, the future development of telecommunication devices needs LED and laser structures with strong emission at room temperature.

Therefore, as a conclusion of this work, we grew a sample gathering all the growth conditions and parameters established in previous experiments. The QD layer of the sample (sample F) was grown at low temperature, with high Sb mole fraction ( $\sim 10\%$ ) and low N content ( $< 2\%$ ). This sample was annealed at  $800\text{ }^\circ\text{C}$  for  $30\text{ s}$ .

The main emission peak (Fig. 5) is observed to be very broad, in between  $1.2$  and  $1.6\text{ }\mu\text{m}$ . The quality is high enough to observe PL even at RT as can be seen in Fig. 5.

Despite the broad peak obtained for this sample, probably due to the size dispersion, the main conditions established in the experiments described, open a new field of interest and reveal a promising way to the future development of cheap light emitting devices in the telecommunication window.

#### 4. Conclusions

In conclusion, we have studied the influence of QDs growth temperature, RTA temperature and Sb incorporation in the emission properties of GaInNAs(Sb) QD samples. We demonstrated significant improvement of this parameter with low growth temperatures of the QD. The post-growth RTA treatment seems to be efficient enough at temperatures between  $800$  and  $850\text{ }^\circ\text{C}$ . Besides, the addition of Sb to the crystal lattice enhances the quality of the QD giving rise to more intense emission at higher wavelengths with less amount of N. Therefore, we obtained a set of growth conditions to obtain practical emission at  $1.55\text{ }\mu\text{m}$  at room temperature and we open a new promising field for the future development of telecommunication devices.

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