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Postgrowth intermixing of strain engineered InAs/GaAs quantum dots

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Abstract

This paper reports on the evolution of InAs/GaAs quantum dots' (QDs) intermixing process depending on the QD position with respect to the InGaAs strain reducing layer. The postgrowth intermixing has been ensured by rapid thermal annealing (RTA) at different temperatures and the processed samples were investigated by photoluminescnece (PL) spectroscopie. For the reference sample containing InAs QDsin pure GaAs matrix demonstrates the highest intermixing degree with an emission energy blueshift up to 300 meV. The reduction of the strain around the QDs for dot-in-well (DWELL) structure has been found to slightly reduces the intermixing degree leading to less emission energy blue shift. However, for the QDs with InGaAs underlying layer, a relative prohibition of the intermixing has been found to occur testifying the existing of thermally stable extented defects.

Keywords: Quantum dots, strain reducing layer, Intermixing.

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Introduction:

Over the past years, self assembled quantum dots (QDs) grown by Stranski-Krastanov (S-K) method have attracted considerable attention for both fundamental physics and applications **[1-3]**. However, this kind of QDs have a lot of limitations, mainly related to the control of their structural parameters, such as QDs shape, material composition and size distribution limit the accurate manipulation of their optical and electronic properties. Meanwhile, the QDs density can be increased and the PL emission energy decreased by introducing InGaAs strain reducing layer (SRL) surrounding the QDs **[4]**. In this case, the QDs present better crystal quality and more effective exciton capture **[5,6]**. Additionally, the QDs properties can be further improved/tuned by means of post growth intermixing **[7-10]**. Such a process can be induced by different techniques such as ion implantation **[11,12]**, laser irradiation **[13,14]**, Impurity Free Vacancy Disordering (IFVD) **[15-17]**, and rapid thermal annealing (RTA) **[10,18]**. The later turns out to be particularly interesting since it is much simpler and faster. Controlled QDs intermixing allow tuning of their emission wavelength which constitute an attractive tool for the manufacturing of multiple-section photonic integrated circuits (PICs) **[19]**. Previous works enrolled in this context have been focused on the investigation of annealed QDs with large inhomogeneous size distribution **[6,20,21]**, doped QDs **[22**,**23]**, multi-stacked QDs **[24**,**25]** and various InAs nanostructures **[10,26**,**27]**. Recently, Keizer et al. **[28]** reported on morphological change of conventional InAs/GaAs QDs layer induced by RTA. The interdiffusion of In/Ga atoms was found to be significantly larger in the QDs where the strain is locally higher. Meanwhile, less attention has been devoted to the investigation of the intermixing effects on the strain engineered QDs. Accordingly the investigation of post growth intermixing of InAs QDs under different strain statues using InGaAs SRL helps understanding the dependency of the atomic interdifusion on the relative position of the QDs with respect to the SRL.

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In this paper, we present a detailed investigation of the optical properties through PL measurement of InAs QDs with different positions within the InGaAs SRL and their behavior upon post growth intermixing by RTA at different annealing temperature.

Experiments:

The investigated samples consist of a single InAs QDs grown by molecular beam epitaxy on Si-doped (001) GaAs substrate**.**. Three different structures were prepared to investigate the influence of the strain on the annealed InAs QDs. Typical InAs QDs embedded in GaAs matrix was grown as reference (QDs sample). Two other samples were grown with different position with respect to the InGaAs layer. This layer is expected to subit strain driven decomposition, modifying the structural and optical properties of the QDs **[24]**. For the QDUL sample, the dots were grown on the top of 10 nm $In_{0.15}Ga_{0.85}As$ SRL, while for the DWELL structure, the InAs QDs were formed on a 2 nm $In_{0.15}Ga_{0.85}As$ and capped with 8 nm thick of $In_{0.15}Ga_{0.85}As$ SRL. Fig. 1 presents schematic representation of this samples. The GaAs capping layer was 60 nm for the refrence QDs sample and 50 nm for DWELL and QDUL samples. The growth rate of InGaAs and GaAs materials are 0.54 Å/s and 2.4 Å/s, respectively. More details concerning InAs QD growth and morphological investigation can be found in ref **[29]**.

RTA was performed on all structures at temperature ranging from 650°C to 800°C for 30s in nitrogen ambient atmosphere. During annealing, the samples were sandwiched between two pieces of semi-insulating GaAs to minimize desorption of As atoms.

The PL measurements were carried out in a set-up using a conventional lock-in technique. The samples were mounted in a closed-cycle temperature-controlled He cryostat and were excited with 514.5 nm line of an Ar+ laser. The PL emission was dispersed through a high resolution Jobin Yvon HRD1 spectrometre and collected using a thermoelectronically cooled InGaAs photodetector.

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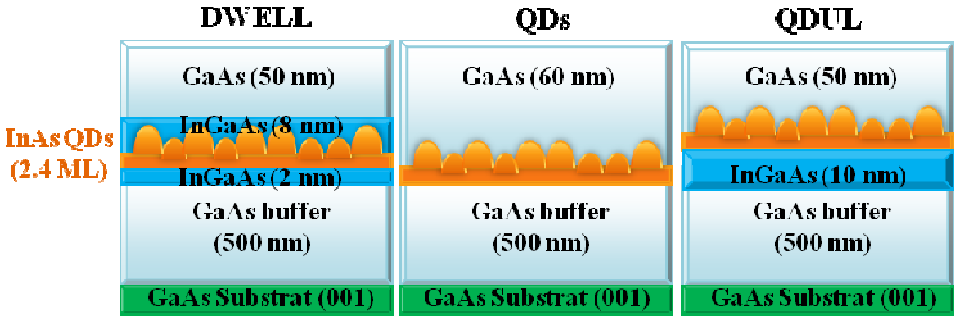


Figure1 schématique representative of the studied structures.

Results and discussion:

The Fig. 2 shows 12K-PL spectra of the as-grown samples recorded at high excitation power. The appearence of multiple peaks in each spectrum arises from the state filling due to 3D carrier confinement. In fact, a clear band filling effect is observed through the decrease of the excitation power density **[29]**, as shown by the inset of the Fig. 2.

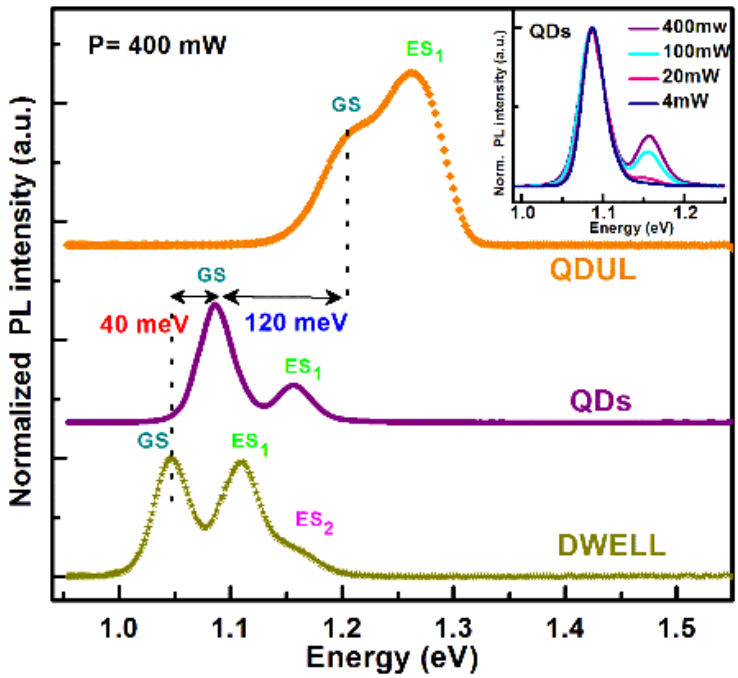


Figure 2 *Normalized 12K-PL spectra of the as grown samples measured at 400 mW excitation power. The inset shows the PL spectra of the QDs structure at different excitation power normalized with respect to the first peak.*

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This suggests that the high energy side peaks comes from the QDs excited states (ES_n) (n=1,2). Moreover, the ground state (GS) emission energy of the DWELL structure is redshifted by approximately 40 meV with respect to that grown in a pure GaAs matrix. This redshift is explained by the redistribution of In and Ga atoms during the deposition of the InGaAs around the InAs QDs leading to a bigger QDs size with larger quantum confinement **[4]**. However, the GS emission energy of the QDUL structure shows a blueshift by approximately 120 meV and broadening of the PL spectra indicating large dot size dispersion [**5,30]**. Morphological investigation of uncapped reference QDs directly formed on GaAs and on InGaAs strain reducing underlaying layer (SRUL) of 2 nm and 10 nm has been previously carried out and reported elsewhere [29]. This study revealed that the density and the size of InAs QDs are strongly dependent on the InGaAs SRUL thickness.

The QDs' density is 2.4×10^{10} cm^{-2} for the QDs grown directly on GaAs. The QDs density increases to 3×10^{10} cm^{-2} for 2 nm thick SRUL and to 4×10^{10} cm^{-2} for 10 nm thick InGaAs SRUL. Additionally, the indium outdiffusion from the SRUL increases the thickness of the nominal InAs material Indeed, when the InAs nominal thickness exceeds a critical value, coalesced/dislocated QDs are formed resulting in a high size dispersion for the QDUL sample. With the formation of coalesced and/or dislocated QDs. The abnormal blueshift transition from the PL spectrum was attributed to the emission of the small QDs..

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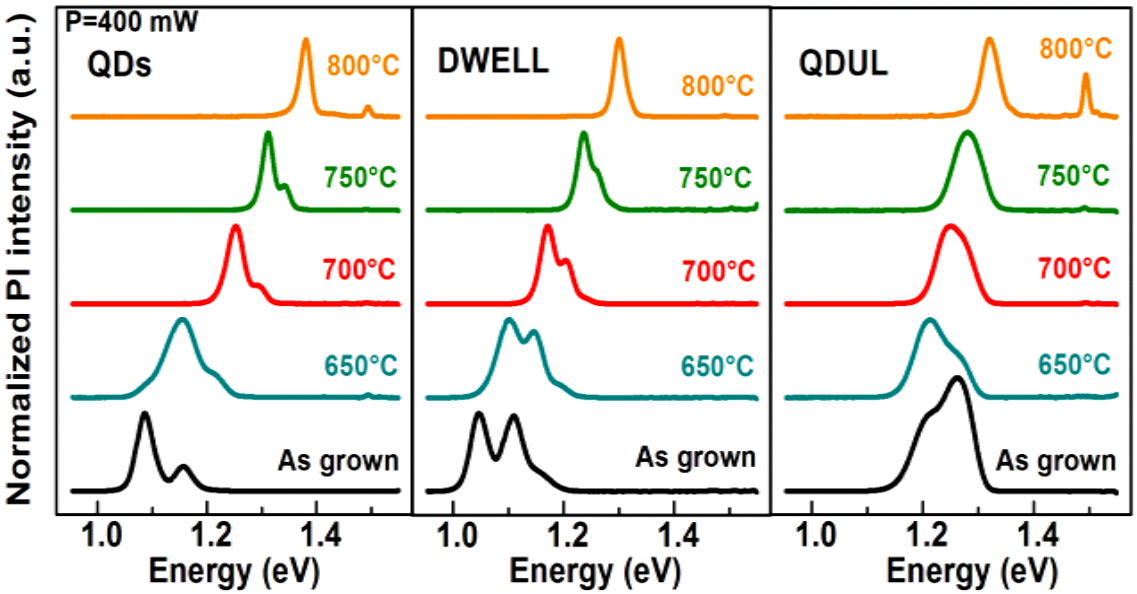


Figure 3 *Normalized 12K PL spectra obtained from the as grown and samples annealed at different temperature of reference QDs, DWELL and QDUL structures.*

The normalized 12 K PL spectra of the as-grown samples as well as samples annealed at different temperatures are depicted in Fig. 3. By Increasing the annealing temperature from 650 to 800 °C, a blueshift of the InAs QDs emission energy and the norrowing of the PL linewidth have been observed. This effect is a direct consequence of the In-Ga atomic interdiffusion across the interfaces between the InAs QDs and the surrounding material . The RTA induced atomic intermixing changes the chemical composition of the QDs by diffusing In atoms from the QDs into the barrier material and Ga atoms in the reverse way, resulting in a change of the carriers' confinement potential and emission energy**[18,25,31]**.

The appearance and improvement of the GaAs PL peak's intensity can be attributed to the annealing induced improvement of its crystalline quality [27]. In order to assess the intermixing effects on the GS emission peak, the variation of the PL FWHM and the integrated PL intensity are plotted as a function of the annealing temperature (Fig. 4).

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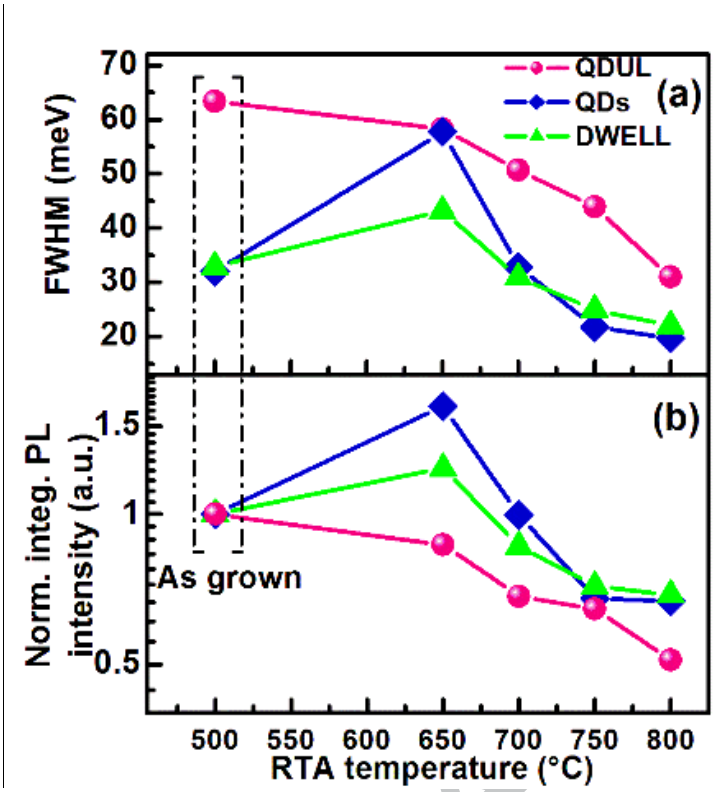


Figure 4 *Evolution of the GS FWHM* ***(a)*** *and Integrated intensity* ***(b)*** *as a function of annealing temperature. The symbols at 500°C designate the PL FWHM and integrated intensity of the as-grown samples.*

The GS emission peak's integrated PL intensity and FWHM of the QDUL sample show a continuous decrease with increasing the intermixing degree. Therefore, the continuous shrinkage suggests an improvement in QDs size uniformity with RTA treatment and the drop of the integrated PL intensity indicates a pronounced enhancement of the nonradiative defects density. These observations are different from those shown by the reference QDs and DWELL samples. Indeed, the integrated PL intensity and FWHM exhibit an increase up to 650°C, as shown in Fig. 4. This behaviors result from non-uniform intermixing process widely shown to occur at low intermixing degree **[17,19,32]**. This inhomogeneous broadening arise from the occurrence of different intermixing degree depending on the dots size and strain distribution from dot to dot. Furthermore, we show that the presence of SRL considerably reduces this phenomena since the integrated PL

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intensity and the FWHM of reference QDs sample are more outstanding than those obtained for DWELL sample at an annealing temperature of 650°C. This trend indicates that this variation depends on the quantity of Ga atoms diffused from the barrier materials into the InAs QDs. For DWELL sample, the presence of the InGaAs SRL around the QDs may reduce the diffusion of Ga atoms towards the QDs due to the presence of In atoms in the surrounding material **[21,25,33]**.

Then, increasing the annealing temperature above 650°C, results in a decrease of the integrated PL intensity and linewidth, suggesting that the intermixing process tends to smooth out the compositional and strain dispersion leading to the observed narrowing of the PL linewidth. Such a phenomena has recently been found to improve the QD's size distribution **[18]**. The narrowest value (20 meV) of FWHM for both reference and DWELL structures appearing at a high annealing temperature (800°C) is lower than that obteined for QDUL structure (around 30 meV). On the other hand, the decrease of the integrated PL intensity may be ascribed to the degradation of the material quality by accumulation and coalescence of defects at high degree of intermixing. The obtained spectrum at low excitation power (not shown here) confirm that the InAs QDs in the samples still survive even at high annealing temperature.

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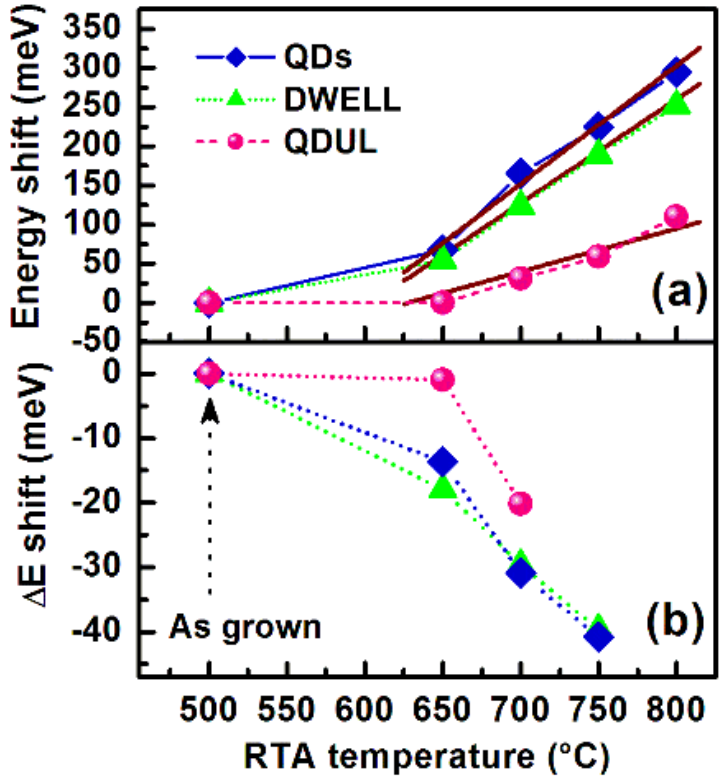


Figure 5 :(a) *the GS PL peak blueshift and* ***(b)*** *the intersublevel spacing energy (ΔE) between GS and first ES, as a function of the annealing temperature.*

Fig. 5 show the relative variation of the GS and the intersublevel spacing energy (ΔE) energy shift as a function of the annealing temperature. The GS emission energy showed a continuous blueshift with increasing annealing temperature, as well as reduction ofΔE . This continuous blueshift is duo to the intermixing of In/Ga atoms , and consequent changes of the QDs composition **[25]**. Meanwhile, ΔE decreases with increasing the intermixing degree as a consequence of the the increased QDs volume **[15,20]**. These observations reveal also that the blueshift is dependent on the surrounding material's composition. The samples can be divided into two groups depending on the total intermixing induced blueshift: reference QDs and DWELL samples with large blueshift reaching a value of 300 meV and 255 meV at 800°C, respectively, and a sample (QDUL) with much lower blueshift of 110 meV, testifying a relative prohibtion of the intermixing process.

As shown by the Fig. 5(a), a linear fit (solid line) to the intermixing induced emission energy blue shift shows a slope of 1.51 meV/°C for the reference sample which is slightly higher than that

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obtained for DWELL sample (~1.32 meV/°C) and much faster than that measured for the QDUL sample (~0.55 meV/°C). The mechanism behind this high sensitivity relay on the density of thermally diffused active vacancies and point defects directly to the single InAs/GaAs interface where the In/Ga intermixing takes place. This effect greatly promotes the Ga/In intermixing across the interface during the RTA. Accordingly, this large blueshift is imputed to the QDs with large size, having large dot barrier interface. Since the intermixing degree is governed by vacancies and point defects density diffused to the QDs, and being directly exposed to the diffused vacancies, the QDs in the reference sample are more sensitive to the RTA. Furthermore, the high strain around the QDs may accentuate the intermixing.

For DWELL sample, having larger QDs, show smaller emission energy blueshift than the reference sample. Indeed, the InGaAs SRL around the QDs compensates the interdiffusion effect by reducing the strain around the QDs and the amount of diffused point defects by recombination at the interface InGaAs/GaAs before reaching the QDs. In this case, the DWELL structure presents less strain at the interface with less Ga in the surrounding material resulting in less intermixing degree **[21,22]**. However, the QDUL sample show less sensitivity to the effect of intermixing, asserted by the small emission energy blueshift. Indeed, as shown by the Fig 3(b), the integrated PL intensity drops with increasing the annealing temperature testifying a pronounced degradation of the QD's structural properties. Morphological investigation of uncapped InAs QDs on InGaAs strain reducing underlaying layer revealed an increased QDs density leading to the coalescence of closely spaced QDs **[29]**. This effect is generally accompanied by a degradation of the QDs' structural properties, through the strain relaxation and generation of threading dislocations prohibiting the intermixing process.

Conclusion

The optical properties of InAs QDs with different positions with respect to the InGaAs SRL has been investigated by PL spectroscopy. The highest intermixing degree has been found to occur

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for InAs QDs in a pure GaAs matrix. The total intermixing induced emission energy shift has been found to be reduced upon insertion of the QDs in InGaAs QW duo to the reduction of the active point defects densities as well as the number Ga atoms surrounding the QDs. However, for the QDUL a total tunable emission energy blueshift, around 100 meV, has been recorded indicating a relative prohibition of the intermixing for this structure mainly duo to the indium segregation and accumulation induced changes of the structural and morphological properties of the QDs.

Our results help understanding the impact of the InAs QDs position with respect to the strain reducing layer on the postgrowth intermixing process.

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Figures Caption:

Figure1:

Schématique representative of the studied structures.

Figure 2:

Normalized 12K-PL spectra of the as grown samples measured at 400 mW excitation power. The inset shows the PL spectra of the QDs structure at different excitation power normalized with respect to the first peak.

Figure 3 :

Normalized 12K PL spectra obtained from the as grown and samples annealed at different temperature of reference QDs, DWELL and QDUL structures.

Figure 4:

Evolution of the GS FWHM **(a)** and Integrated intensity **(b)** as a function of annealing temperature. The symbols at 500°C designate the PL FWHM and integrated intensity of the as-grown samples.

Figure 5 :

(a) the GS PL peak blueshift and **(b)** the intersublevel spacing energy (ΔE) between GS and first ES, as a function of the annealing temperature.

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Highlights of our study are the following:

a. The InAs QDs in a pure GaAs matrix are more sensitive to the RTA temperature allowing the highest emission energy blueshift
b. Reducing of the intermixing degree by using QDs in a Well structure
c. In segregation and accumulation in the InGaAs underlying layer alter the structural and morphological properties of the QDs.
d. The intermixing is found to be partially prohibited for Qds grown on InGaAs strain reducing layer

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