Influence of rapid thermal annealing on Stranski-Krastanov and submonolayer quantum dots

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Abstract

InAs Submonolayer quantum dots (SML-QDs) have emerged as a new type of quantum dots (QDs) capable to overcome most disadvantages of traditional Stranski-Krastanov InAs quantum dots (SK-QDs). These new nanostructures have a higher QD density (that can reach 10^{12} cm^{-2}), allow a flexible control of their height and composition, and have no wetting layer. Although SML-QDs seem extremely promising, their growth is more complex and, since usual characterization techniques like scanning electron microscopy (SEM) and atomic-force microscopic (AFM) are useless, still very little is known about their structural properties [1]. One way to get more information is to perform rapid thermal annealing (RTA), which is a post-growth technique consisting in submitting the sample to a high temperature for a short time. It is known that, when applied to SK-QDs, this technique contributes to lower the density of structural defects and leads to a strong In diffusion out of the QDs, resulting in an increase of the photoluminescence (PL) intensity and in a blueshift of their spectrum [2]. In the present work, we used low-temperature PL to investigate the effects of RTA over two different types of SML-QDs and compared the data with those of conventional SK-QDs. The results suggest that SML-QDs have much less structural defects than SK-QDs and are probably also under much less stress due to their lower average In content.

Introduction

Tuning of the emission or absorption wavelength of InAs/GaAs quantum dots has many applications in devices such as lasers, infrared photodetectors, and solar cells. The main parameters generally used to do that are the size and In composition of the nanostructures. Quantum Mechanics shows that larger QDs have lower energy levels, whereas decreasing their In content increases the energy gap of their material and, consequently, raises their energy levels. The first effect leads to a redshift of the PL spectrum, while the second yields a blueshift. Since the InAs/GaAs system is highly strained, during RTA there is generally a strong diffusion of In atoms out of the QDs that results simultaneously in an increase of their apparent size and a decrease of the overall In content. PL data can provide valuable information about the size, composition and defects in such nanostructures.

Samples

We grew three samples: one with SK-QDs (Sample #1) and two with SML-QDs under a c(4x4) (Sample #2) and (2x4) (Sample #3) surface reconstruction (Fig. 1). *Ex-situ* RTA was performed at 920 °C for 30 s with a rising ramp of 80 °C/s.

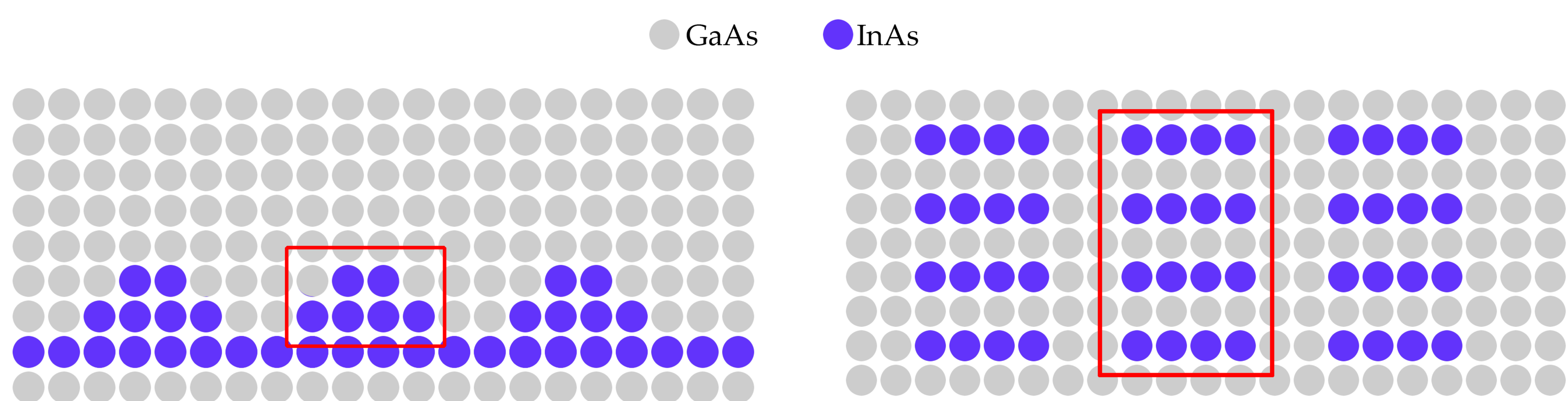

Fig 1: (left) Illustration of a SK-QD (red rectangle). The growth method consists in depositing InAs over GaAs. Above a critical thickness of 1.7 monolayers (MLs), the InAs film relaxes (due to the lattice mismatch between both materials), and 3D InAs islands are nucleated all over the surface, acting as QDs.

(right) Illustration of a SML-QD (red rectangle). The growth method consists in a cycled deposition (6x) of a fraction of a monolayer of InAs (0.5 ML) followed by a few monolayers of GaAs (2.5 MLs). The difference between sample #2 and #3 was the surface reconstruction ((2x4) and c(4x4)) prior to deposition.

Results & Discussion

As can be seen in Fig. 2, after RTA, SK-QDs show a strong change in their PL spectrum that is blueshifted, more intense and narrower than that of the as-grown sample. X-STM measurements of such QDs revealed that there is a large out diffusion of In atoms during RTA, which results in an increase of their size and a consequent reduction of their average In content. The blueshift is due to the lower In content that overcomes the influence of their larger size, while the increase of PL intensity comes from the passivation of a part of the defects introduced during the growth of this highly strained system. The narrowing of the spectrum originates from the fact that quantum effects are weaker in larger structures.

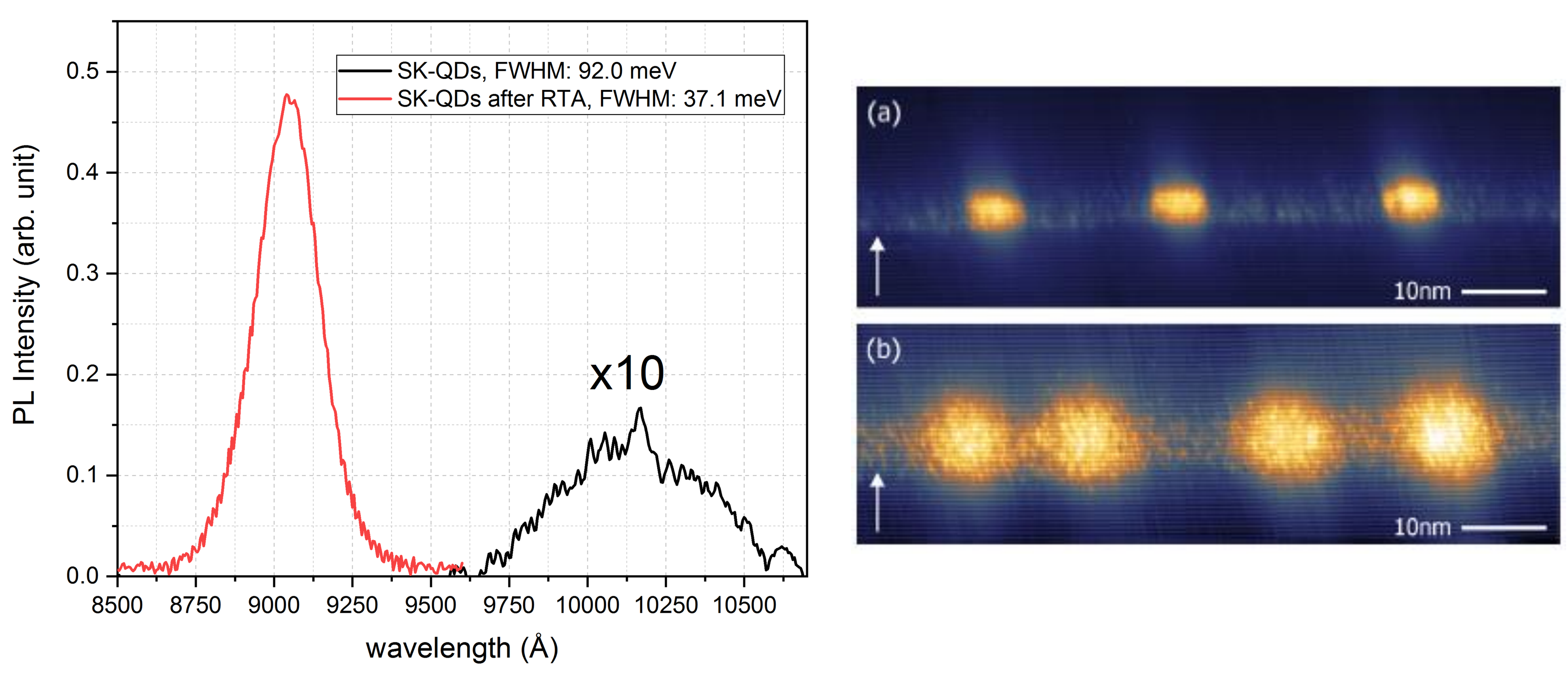

Fig 2: (left) Photoluminescence spectra at 77K of SK-QDs (Sample #1) before (black) and after (red) RTA. (right) Topographic X-STM images [2] of SK-QDs (similar to Sample #1) before (a) and after (b) RTA. The samples were cleaved under ultra-high vacuum and their cross section was analyzed in the (110) plane. The arrows indicate the growth direction [001]. The brightness represents the local corrugations out of the surface due to the strain induced by the In atoms.

Results & Discussion

Figure 3 shows that, in the case of SML-QDs, there is almost no change of intensity, width and emission wavelength after RTA, the latter being naturally much lower than for SK-QDs because of the smaller size and lower In content of SML-QDs that can be observed in Fig. 4. This lower In content also reduces the internal strain of the nanostructures and, consequently, the effects of RTA. The very narrow emission is due to the extended wavefunction of their ground state that overlaps several QDs.

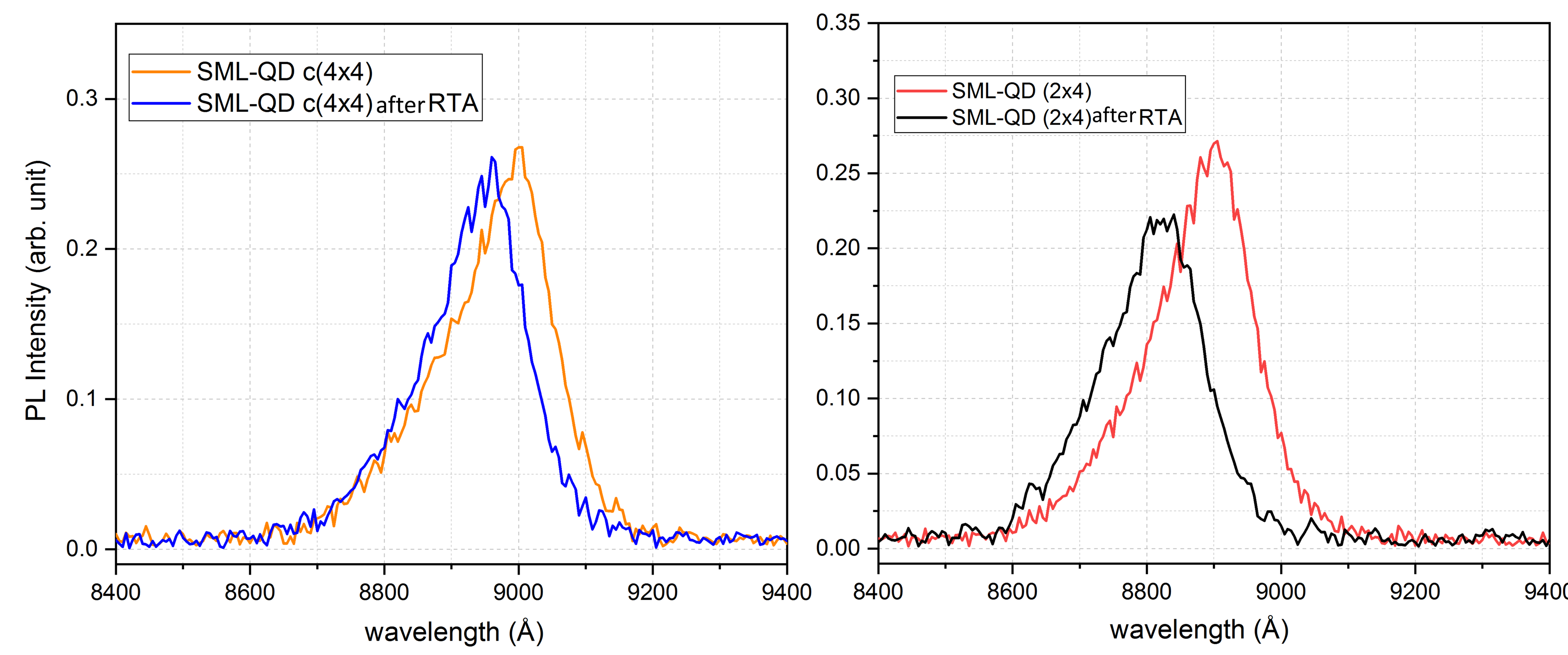

Fig 3: PL spectra at 77K, before and after RTA, of SML-QDs grown in the presence of a c(4x4) (left) and (2x4) (right) reconstruction of the GaAs(001) surface.

Figure 4 shows no evidence of stacks of small 2D InAs islands and, instead, reveals the presence of irregular In-rich clusters dispersed in a dilute InGaAs layer resulting from the strong segregation effect of In atoms. The specific (2×4) growth conditions provide a lower In incorporation, a higher In segregation [1], and a lower density of QDs [3].

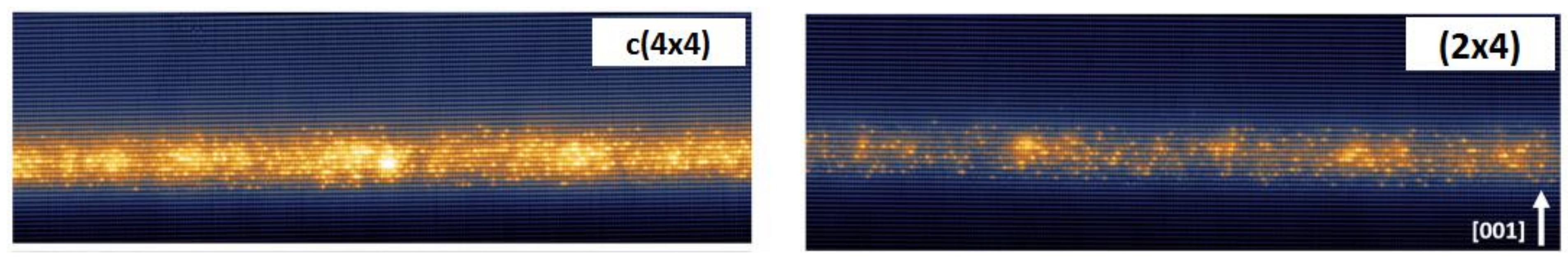

Fig 4: 80×25 nm² topographic X-STM images of SML-QDs (without RTA) grown in the presence of a c(4x4) (left) and (2x4) (right) reconstruction of the GaAs(001) surface [3].

Conclusions

RTA and PL experiments clearly show that strain in SML-QDs is much smaller than in SK-QDs and most probably results from their smaller size and lower In content. This is confirmed by X-STM data which reveal that In segregation inhibits the formation of small 2D InAs islands and their stacking, leading thus to irregular In-rich agglomerates dispersed in a thicker but dilute InGaAs layer. After solving the problem of In incorporation detected for the (2×4) growth conditions, one expects such SML-QDs to overcome those usually grown in c(4×4) conditions.

References & Acknowledgements

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[2] "Atomically resolved study of the morphology change of InAs/GaAs quantum dot layers induced by rapid thermal annealing"; J. G. Keizer, A. B. Henriques, A. D. B. Maia, A. A. Quivy, and P. M. Koenraad; Appl. Phys. Lett. 101, 243113 (2012).

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