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# Control of the lateral growth morphology in GaAs Droplet Epitaxy

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**Abstract.** We present the detailed fabrication method of two different GaAs nanostructures with cylindrical symmetry by the Droplet Epitaxy technique. Concentric Multiple Rings or Coupled Rings/Disks are successfully obtained, exploiting the lateral growth around the Ga droplets, through a fine control of the crystallization dynamics.

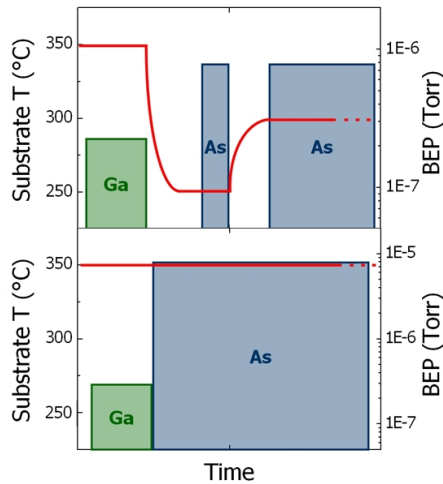
## 1. Introduction

During the past two decades, semiconductor quantum nanostructures (NS) have been extensively investigated because of their potential application in the field of optoelectronics as active elements for lasers [1, 2], light-emitting diodes [3], detectors [4] and quantum information technology, as sources of entangled states [5, 6]. In particular, self-assembled Stranski-Krastanov (S-K) quantum dots (QDs) [7], formed by growing a lattice mismatched structure, as a result of the balance between strain relaxation and surface energy, have widely been investigated. However, these kind of systems mainly show a pyramidal-like shape and therefore the tuning of the NS optoelectronic properties through the careful design of their shape is very difficult.

Amongst the Molecular Beam Epitaxy-based techniques, the Droplet Epitaxy [8, 9] (DE) has shown an unmatched potential for the fabrication of complex III-V semiconductor NS. In DE, unlike the more conventional S-K growth, the III- and V-column elements are alternatively supplied to the substrate surface, for the formation of quantum NS. In the case of GaAs, after the irradiation of Ga molecular beam flux, nanometer-sized droplets with a low size dispersion spontaneously form at the surface. If an As flux is then supplied, new GaAs nanocrystals develop from the original droplets. Depending on the growth conditions, several kind of different morphologies have been obtained ranging from QDs [10], quantum rings [11], concentric multiple rings [12] and coupled ring/disks [13, 14].

We demonstrate the fabrication method of complex GaAs NS with cylindrical symmetry, based on the fine control of the lateral growth around the Ga droplets during the As supply. With a proper choice of the growth conditions, the morphology of the crystal can be tuned from a clear peak-and-valley profile for Concentric Multiple Rings (CMRs) to a flat disk-like structure grown in a layer-by-layer fashion, for Coupled Rings/Disks (CRDs). These structures were characterized by means of Atomic Force Microscopy (AFM) and Reflection High Energy Electron Diffraction (RHEED) analysis. The interplay between the Ga diffusion from the initially formed Ga droplets and the As

adsorption onto the substrate surface in the different experimental conditions is thought to be crucial for the shape engineering of the NS.



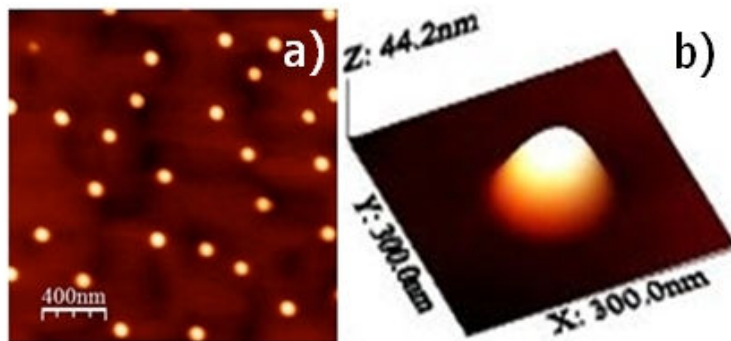
**Figure 1.** Sketch of the experimental procedure for Sample A (upper panel) and B (lower panel). Red lines show the substrate temperature evolution during the growth. Green and blue areas are depicted in correspondence to the gallium and arsenic supply, respectively.

## 2. Experimental

The growth experiments were performed on GaAs (100) wafers in a standard GEN II MBE system equipped with an As valved cell, which provided  $As_4$  molecules. After the native oxide desorption at 580°C, a buffer layer of GaAs and a barrier layer of  $Al_{0.3}Ga_{0.7}As$  were grown at the same temperature, taking care of the atomically flatness of the surface. After that, the substrate temperature was decreased to 350°C, the As flux cut and the background pressure in the chamber reduced to around  $5 \times 10^{-10}$  Torr. The substrate surface was then exposed to a Ga molecular beam equivalent to 10 MLs, with a deposition rate of 0.08 ML/s, to form droplets. Two samples, named A and B were prepared, following different experimental procedures, constantly monitored by means of RHEED. In the case of Sample A, after the Ga droplets formation, the substrate temperature was decreased to 250°C and a short-time (20 sec) As flux of  $8 \times 10^{-7}$  Torr was supplied. Finally the substrate temperature was raised again to 300°C and a second, long-time (20 min) arsenic exposure of the same intensity was used to completely ensure the crystallization. On the contrary, for Sample B, after the Ga deposition at 350°C, the As shutter was immediately opened at the same temperature, providing an  $8 \times 10^{-6}$  Torr arsenic partial pressure. In Fig. 1 the detailed experimental procedure for the fabrication of Sample A (upper panel) and Sample B (lower panel) is shown. After the growth, the samples were taken out from the chamber for the morphological analysis, carried out by Atomic Force Microscopy (AFM).

## 3. Results and discussion

As already described above, the first step for the fabrication of GaAs NS by DE is the formation of Ga droplets at the substrate surface. In our case, 10 MLs of Ga were supplied at 350°C for both Sample A and Sample B. In order to confirm the presence and the morphology of the Ga droplets, another sample was prepared, by stopping the growth just after the Ga deposition. The substrate temperature was rapidly cooled down (40 °C/min) to around 200°C and the sample taken out from the growth chamber. AFM measurements, which confirmed the existence of Ga droplets on the  $Al_{0.3}Ga_{0.7}As$  barrier layer are shown in Fig. 2. The average droplets diameter and height are around 90 nm and 35 nm respectively and the surface density of the Ga droplets is  $8 \times 10^8 \text{ cm}^{-2}$ . Taking into account that 1.7 MLs of Ga are consumed to create a Ga-rich surface reconstruction [15] before the Ga droplets formation, a perfect correspondence is found when comparing the total amount of Ga atoms into the droplets and the initially supplied 10 MLs.



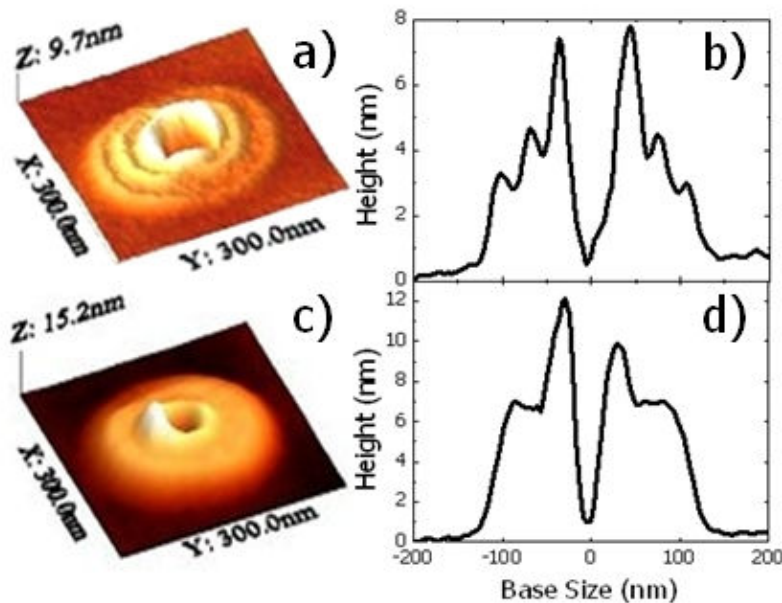
**Figure 2.** Two dimensional 2 micron x 2 micron AFM image of Ga droplets just after the deposition a). Three dimensional magnification of a single droplet b).

In Fig. 3 the AFM images (a, c) as well as the cross sectional height profiles (b, d) for Samples A (upper panels) and B (lower panels) are presented. In the case of Sample A, the final morphology consists of a concentric triple ring with inner, middle, and outer ring diameters of around 80, 140, and 210 nm, respectively and with heights around 8 nm for the inner rings, 4 nm for middle rings, and 3 nm for the outer rings. This triple ring structure present a good rotational symmetry, with a slight elongation of around 11% along the [0-11] direction, which can be caused by the anisotropic surface migration of Ga on the (100) GaAs surface [16]. The shape of Sample B is instead characterized by an inner ring with a diameter and height of around 80 nm and 11 nm respectively, surrounded by a disk with a diameter of around 200 nm and a nearly constant height of around 7 nm. The NS density at the surface is nearly equivalent to the one of the initial Ga droplets, suggesting that every droplet is transformed into the above described GaAs NS at the end of the procedures. The difference in the volume of the final GaAs structures (lower for Sample A) has to be addressed to the different experimental procedures, involving changes in the substrate temperature. During the waiting time, necessary for the substrate temperature stabilization, a partial Ga crystallization, forming a 2D thin layer all over the substrate, is observed even in absence of intentional As supply, leading to a decrease in the effective number of Ga atoms available for the formation of the final nanocrystal.

A nearly perfect correspondence is found when the diameter of the inner ring in both Sample A and B is compared to the original droplets one (80-90 nm in every case). Selective etching of pure Ga showed that, underneath the droplets, a tiny GaAs ring is already formed, just after the Ga deposition [11]. These observations imply that the inner ring formed at the end of the growth is the result of the accretion of the initially formed ring, at the droplets edge. This is the reason why, for both CMRs and CRDs, regardless of the difference in the experimental procedure, the inner ring diameter is nearly constant.

On the contrary, the lateral growth around the inner rings seems to be strongly influenced by the different procedures. Indeed the cross sectional morphology of Sample A is very different from the one of Sample B, since, in the latter, no clear peak-and-valley features are observed in the height profile. As a matter of fact, the development of GaAs NS laterally grown around the Ga reservoir depends on the mobility of Ga atoms which migrate from the droplet and are crystallized, due to the impinging As atoms. However in the first sample, where the arsenization steps are performed at 250°C and 300°C, we observed a peak-and-valley line profile, while in the second sample, where As was supplied at 350°C, a flat disk was grown. Indeed, as already discussed in [12] the first and second arsenisation steps lead to the formation of the middle and outer rings, respectively. This observation suggests that a key role in determining the final shape of the NS might be played by the substrate temperature during the arsenic irradiation to the droplets. The appearance of outer rings, with a well defined peak-and-valley profile can be addressed to the formation of As-rich c(4x4) regions around the droplets, as discovered by RHEED investigations [12]. The establishment of this As-rich regions, which are responsible for the formation of the rings, might be less favorable at  $T \geq 350^\circ\text{C}$ , caused by a higher desorption of  $\text{As}_4$  from the substrate in the high temperature regime [17]. In absence of these zones for the preferential GaAs nucleation, the lateral growth proceeds in a layer-by-layer fashion,

giving rise to the disk-like structure around the original Ga droplet. The dissimilar growth dynamics thus gave rise to the distinct lateral morphologies found in Sample A and Sample B. Therefore the fine control of the interplay between the migration of Ga atoms from the droplets and the As adsorption onto the substrate surface gives the possibility of a shape engineering of the GaAs NS grown by DE. Since electronic properties depend on the geometrical features of the NS, this capability of tuning the shape of the MBE grown GaAs NS is of the outmost relevance in view of the establishment of a future generation of devices.



**Figure 3.** AFM images of a single CMR a) and CRD c) and corresponding cross sectional height profiles taken along the (0-11) direction b), d). For each sample, every droplet is transformed into the described structure, with a size dispersion comparable to the one of the original Ga droplets.

#### 4. Conclusion

In conclusion, we have presented the detailed fabrication method of two different classes of GaAs NS grown by DE: CMRs and CRDs. In both cases, the lateral growth around the initially formed Ga droplets is exploited for the realization of the structure. However, while for the CMRs a clear peak-and-valley cross sectional height profile is obtained, for the CRDs a nearly flat structure is developed. We demonstrated that the morphology of the NS can be finely controlled by a suitable choice of the growth parameters. We believe these innovative GaAs NS to be promising for the future application in optoelectronics and quantum information technology.

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