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The monitoring of 2d-3d transition for InAs/GaAs (001) self-assembled quantum dots by atomic force microscopy

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We present a detailed Atomic Force Microscopy study of InAs/GaAs (001) self-assembled quantum dots, grown by Molecular Beam Epitaxy, during its complete evolution cycle, transition from 2D islands to 3D islands. We created the dots by deposing InAs on a GaAs substrate. After a critical InAs coverage value is reached the dots become self-assembled due to strain. The resulting dots typically have a height of 5.7 nm and an emission at about 900 nm occurs until a second critical coverage point is traversed.

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Keywords: Nanomaterials, Quantum Dots, Self assembling

1. Introduction

In recent years, the surface morphology of the deposited films in heteroepitaxial growth has attracted intense interest because of its importance for basic science and applications in optoelectronic devices. The InAs-GaAs heteroepitaxial system has a lattice mismatch ≅7% and under very specific condition the growth mode follows a version of the Stransky-Krastanov (SK) mechanism, witch results in the formation of coherent (dislocation-free) three-dimensional (3D) islands, after the growth of between one and two monolayers (MLs) in a two dimensional (2D) layer-by-layer pseudomorfic mode [1]-[6]. The 3D islands are usually referred to as self-assembled quantum dots, or more simple QDs. We will focus our attention here to material grown by molecular beam epitaxy (MBE), because almost all of the fundamental results have been obtained using this method. It is generally agreed that the essential driving force for coherent QD formation, after a wetting layer (WL) has been formed, is strain relaxation, whereby the energy gain from the increase in surface area via dot formation more than compensates the increase in interfacial free energy [2].

During the 2D to 3D transition the islands rapidly reach a saturation number density of between 10^9 cm^{-2} and 10^{11} cm^{-2}, depending on growth condition, with a rather narrow size distribution, where each island contains between 10^4 and 5×10^4 atoms, again depending on growth condition, principally deposition rate and substrate temperature.

2. Sample preparation and experimental techniques

In principle, the initial misfit strain is independent of both substrate orientation and surface reconstruction, so there should be no dependence of QD formation on these parameters. But, the experience has demonstrated that the QD form only on the substrate (001)-c (4x4) with strong alloying effect, or wetting layer (WL). We will deal first with the initial 2D WL behavior, before discussing alloying in QD. Afterwards, we concentrated on deposition on (001)-c (4x4) substrates of InAs. In order to obtain InAs quantum dots on GaAs (001) substrate, we have used as growth method a conventional solid source MBE equipped with reflection high energy electron diffraction (RHEED) for *in situ* monitoring of the growth [3]-[6]. In the Fig. 1 and 2 we represent MBE equipment with RHEED and image from RHEED in real time.

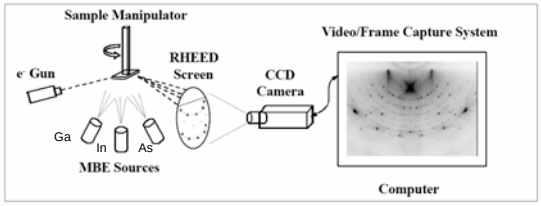

Fig. 1. MBE with RHEED for grown QDs InAs/GaAs.

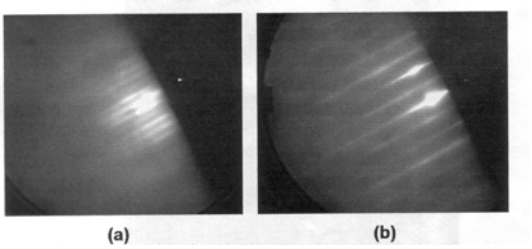

Fig. 2. Diagram indicating the RHEED pattern observed during WL formation of InAs of on a) GaAs (110) and b) on GaAs (1-10).

RHEED analysis gives us information about the 2D-3D transition by changing the diffraction pattern from streaky to spotty. Because the sample used here presents a coverage gradient, RHEED analysis shows several zones where 2D-3D transition was produced (spotty pattern) and others where the film is a 2D one (streaky pattern). The sample has been examined ex-situ by tapping mode Atom Force Microscopy (AFM). We have studied the evolution of the density (ρ) curve as a function of coverage (θ) following the differences between QDs, which are on the mounds and those which are out of them. The influence of the wetting layer stepped structure over the in-plane position of dots has also been investigated.

3. Experimental structure

The experimental structure is shown in Fig. 3 and the flux diagram for preparing this structure is shown in the Fig. 4.

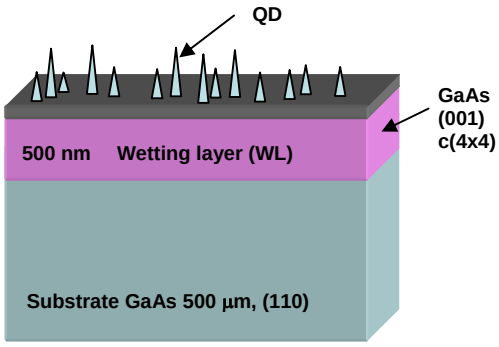

Fig. 3. Structure of QD InAn/GaAs.

In order to grow InAs quantum dots on GaAs (001) substrate we have used a GaAs (001) wafer, its thickness being of 500 μm. This wafer has been bonded using indium to molybdenum block and then put into the load lock and heated for several hours to outgas the sample and block before moving into the preparation chamber. The sample is out gassed again in the preparation chamber at 450^0C before it is moved into the growth chamber. It is necessary to learn that a clean surface is an important feature for epitaxial growth since contaminants from the atmosphere or other sources can easily contaminate a clean GaAs wafer and cause crystal defects or degrade the optical and electrical characteristics of the epitaxial layers. In order to cope with this observation we have used an *epi-ready* GaAs (001) wafer that is pre-cleaned and oxidized in a controlled environment. The RHEED screen will show a haze that is indicative of the amorphous nature of the protective oxide. The substrate temperature is ramped up (~650 ^{o}C) with arsenic over pressure until diffraction pattern (4 x 4) reconstruction appears showing that the oxide has been removed from the surface.

Prior to InAs deposition, a GaAs buffer of approximately 750 nm in thickness is grown on the (001) oriented substrate, in As overflow, at 590 ^{o}C and a rate of 1 μm/h. After 10 minute post-growth annealing, the temperature is lowered to 500 ^{0}C for the InAs deposition. The growth was performed rotating the sample so as to obtain a homogeneous sample. The InAs is evaporated at a rate of 0.029 ML (monolayer)/s. The InAs delivery time is cycled in 5s of evaporation followed by 25 s of growth interruption until the given thickness is reached. The growth rate dependence of the sample position (α) gives the coverage evolution (Φ) along of the one side of the sample (110). In the range of $\alpha = 330^0\text{-}351^0$ this dependence is linearly such as it's enough to measure the growth rate for $\alpha = 340^0$ and $\alpha = 351^0$ from the RHEED oscillations. The sample used in this work has a coverage gradient from 0.9 ML to 2.4 ML and, as a function of this coverage, we can observe on the surface layer the formation of different structures such as: mounds, large and small 2D islands 1 ML height, small quasi-3D islands (quasi-3D QD) of height $\leq$ 2 nm and base size < 20 nm, 3D quantum dots of height 3-4 nm and base size < 40 nm.

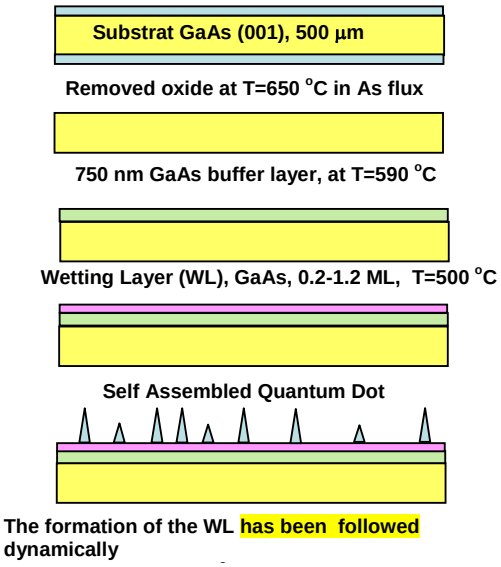

Fig. 4. Diagram Flux for QD.

4. Results and discussions

First, we have analysed the evolution of the material growth for density curve as a function of coverage (from 1.42 ML to 2.2 ML). Under the coverage 1.42 ML we have not observed the formation of the quasi-3D QDs and 3-D QDs. In the Fig. 5 we represent the experimental measurements with AFM after growing layers for different coverage. Regarding the figure 5, we can observe that the nucleation process of quasi-3D QDs and 3D QDs are influenced by the coverage and the nucleation of WL.

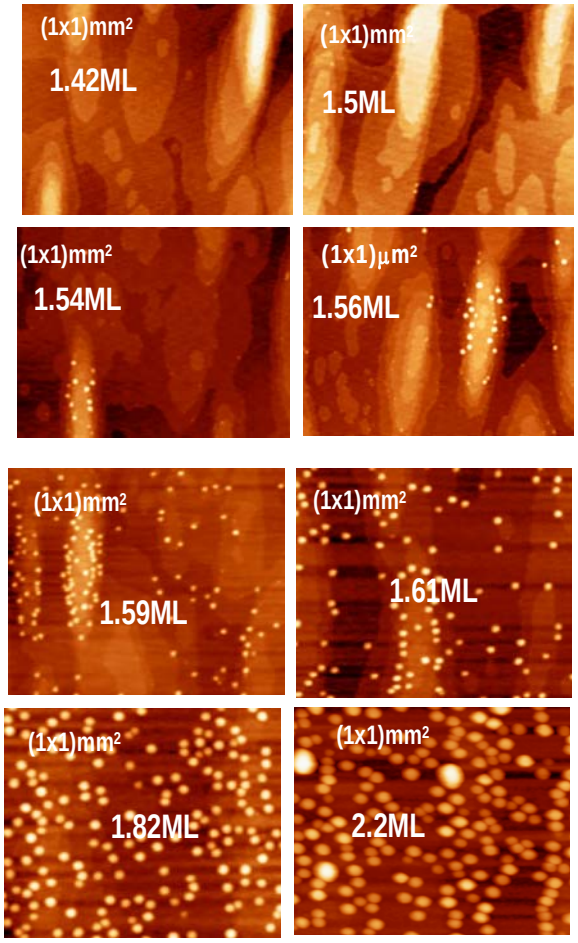

Fig. 5. Images of the AFM for MBE InAs/GaAs structures.

In the early stages of the growth, when the InAs amount deposited on the surface it is not sufficiently to the formation of 3D islands, the growth proceeds bidimensionaly to the formation of the wetting layer (WL). The characteristic structures of these growth stages are the mound and the 2D islands. The origin of mounds is related of the kinetically instability during the growth, while its elongated shape depends of the diffusion anisotropy between [110] and [-110] directions. During the wetting layer formation increases the number of 2D islands as an attempt of strain relaxation because of mismatch-lattice. Approaching the critical thickness, the surface morphology of InAs/GaAs interface becomes quite complex [5]. We can observe different features such as 2D islands (area over 100 nm^2, continuum layer, high 1-2 ML, quasi-3D QDs (diameter 20 nm, high 0.3-0.7 nm, about 1000 atoms) and 3D QDs (diameter 10-20 nm, height more than 2 nm, about 10^4 atoms). In Fig. 6 we represent the AFM image for coverage 1.56 ML where all three elements are evidenced.

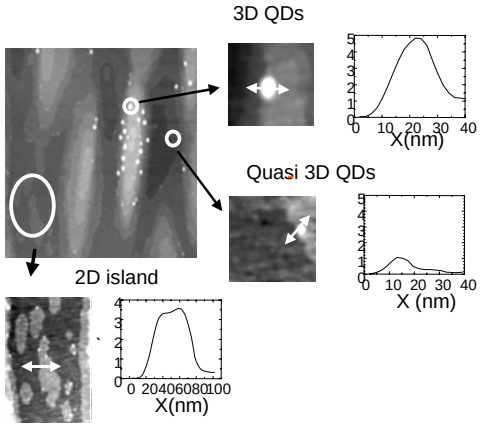

Fig. 6. Image AFM on the InAs/GaAs structure after coverage 1.56 ML.

We have analysed the evolution of the density curve as a function of coverage for quasi-3D QDs and for 3D QDs (Fig. 7 and 8). Regarding the Fig. 7, we can observe that the nucleation process of quasi-3D QDs is not influenced by the presence of the mounds (both curves have the same evolution).

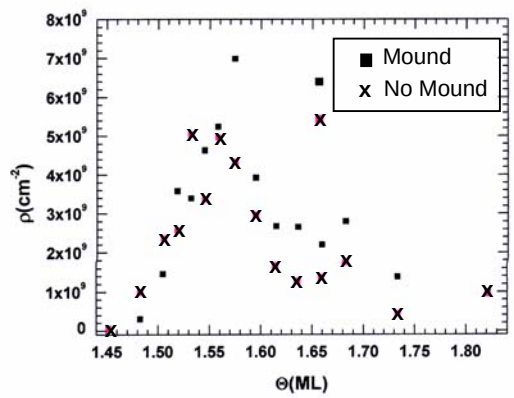

Fig. 7.The evolution of quantum dots density as a function of coverage in the case of quasi-3D dots.

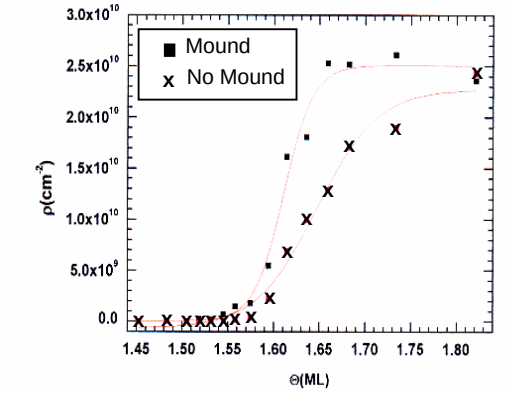

Fig. 8. The evolution of QDs density as a function of coverage.

Further, we have tried to see if the existence of mounds influences the growth process of dots after the nucleation process has taken place. To this aim we have studied the evolution of average volume (V_M) for a single 3D-QD both on the mound and out of the mound (Fig. 9).

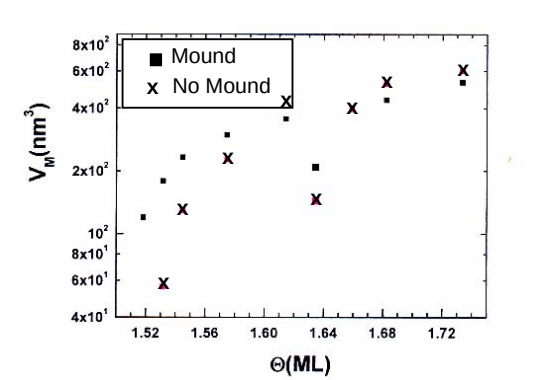

Fig. 9. Evolution of average volume (V_M) for a single 3D-QD.

From this study we can see that the increasing of 3D-QD volume is faster on the mound than out of him. An explanation of this fact can be given if we take into account the relationship between the atoms diffusion process and the mound slope (the higher mound slope, the faster atoms diffusion). The other stage of our research refers to the influence of the wetting layer stepped texture over the in-plane position of the dots. Prior to present the results which have been reported, we have to do a classification of the dots as a function of their position relative to the steps. Thus, we can talk about four kinds of QDs as follows: 3D-QDs which are placed in the upper side of the step (these are named *up* QDs), 3D-QDs which are placed in the proximity of the bottom side of the step (these are named *down* QDs) and 3D-QDs which are placed far from the step (these are named π QDs). The fourth kind of QDs regards the dots which are supported on many steps (it's the case of step bunching structure). These are named bridge dots.

In the Fig. 10 we represent schematic all four type of QDs. From the representation of the density curves for the QDs kinds presented in Fig. 11, we have deduced the following consequences:

-Both on the mound and out of the mound the nucleation of the first 3D-QDs takes place in the proximity of the steps (for instance bridge-dots on the mound and up-dots out of the mound are forming).

-Dots nucleate on the surface starting with 1.57 ML and prevail along the growth process. Their formation is not relied on the presence of extended defects such as steps or step bunching structure so as, from their evolution we could deduce some information about the nucleation process of 3D islands.

-As shown in Fig. 11, the first 3D-QDs form with an anticipation of 1.53 ML (with respect to the π ones). Therefore we can conclude that the presence of the extended defects accelerates the 2D-3D transition.

-In the case of 3D-QDs formed on the mound, the first QDs belong to the bridge category because of the existence of the step bunching structure. We can observe dots which belong to other QDs categories only of ~1.54 ML.

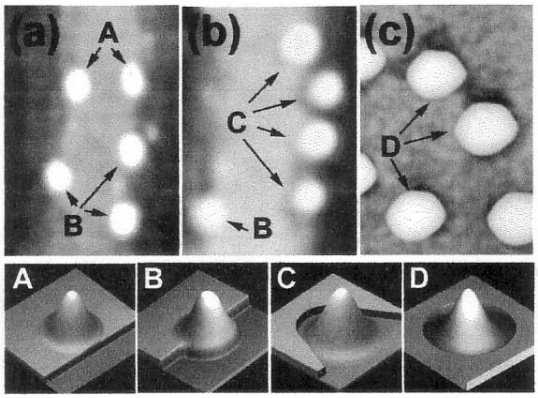

Fig. 10.The types of QDs; A-Qd up; B-QD bridge; C-QD down and D-QD π.

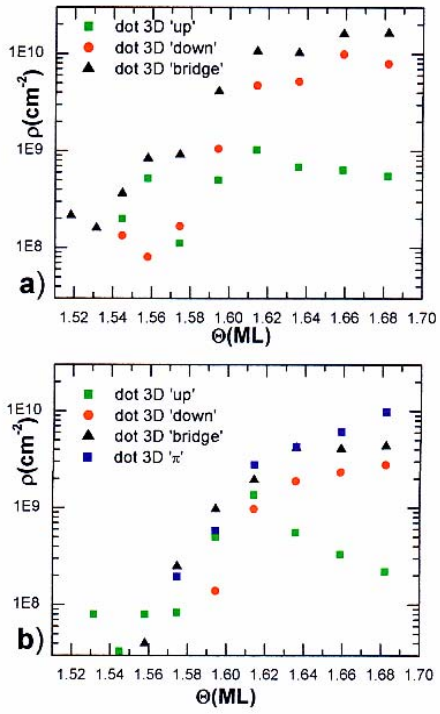

Fig. 11. Density of Quantum Dots for all types as function of grow rate: a) mound and b) out of mound.

In the Fig. 12 we represent the images of InAs/GaAs 3D QD for coverage 1.6 ML.

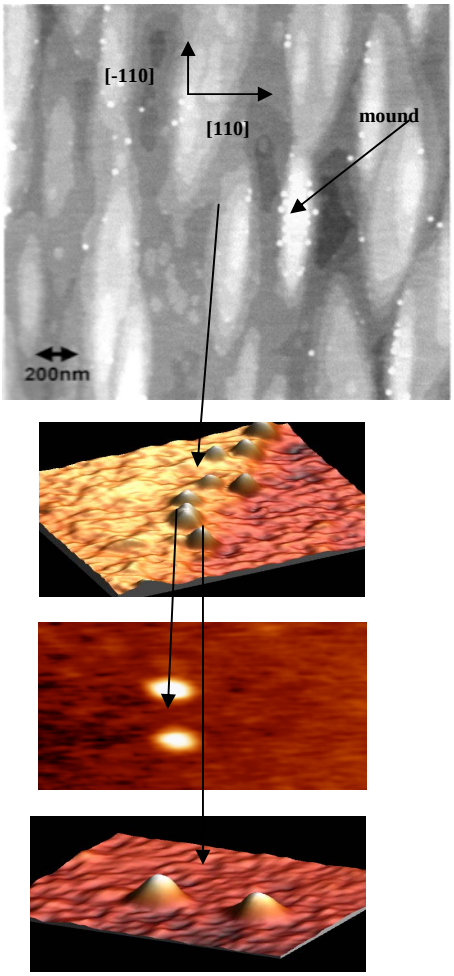

Fig. 12. AFM image (2 x 2) μm^2 of InAs/GaAs for Φ = 1.6 ML (Φ is the coverage).

The quasi-3D QDs start to nucleate between 1.4 and 1.5 ML of InAs, increase in number up to 1.7 ML and vanish above 1.9 ML (see figure 6). The appearance of 3D QDs at 1.5 ML continues with an abrupt increasing of the number density between 1.6 and 1.7 ML. The density 5×10^{10} dot/cm^2 represents the saturation value of the 3D QDs density. Beyond this value do not appear new dots due to the saturation of the possible nucleation sites. We can assume as critical coverage, 1.59 ML (Fig. 13).

From the studying of the two evolution curves of dots density we can observe that the density number value of quasi-3D QDs decreases from 1.1×10^{10} cm^{-2} to 4.3×10^{9} cm^{-2} for a coverage between 1.57 ML and 1.61 ML, while for 3D QDs it increases by a factor of 10 (from 2.1×10^{9} cm^{-2} to 2.3×10^{10} cm^{-2}) so as 3D QDs become the prevailed structures.

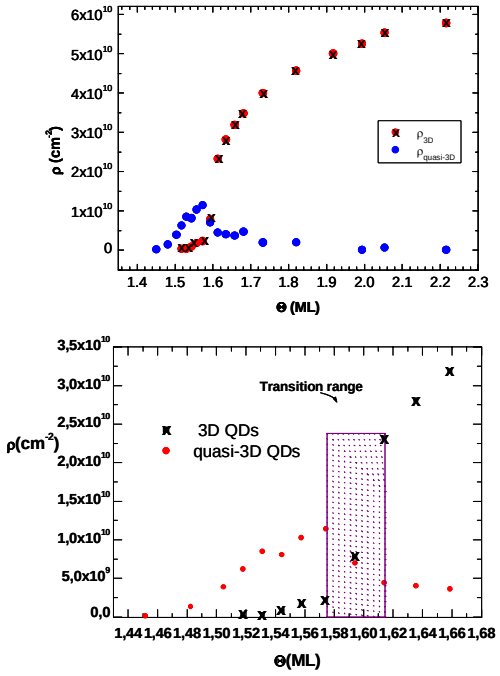

Fig. 13. Quantum dots number density for coverage between 1.45 ML and 2.2 ML.

5. Statistical analysis of quantum dots

In order to perform a statistical analysis regarding the volume and density of the QD, it is necessary to give quantitative criteria which allow us to distinguish between the quasi-3D dots and 3D dots (Fig. 14).

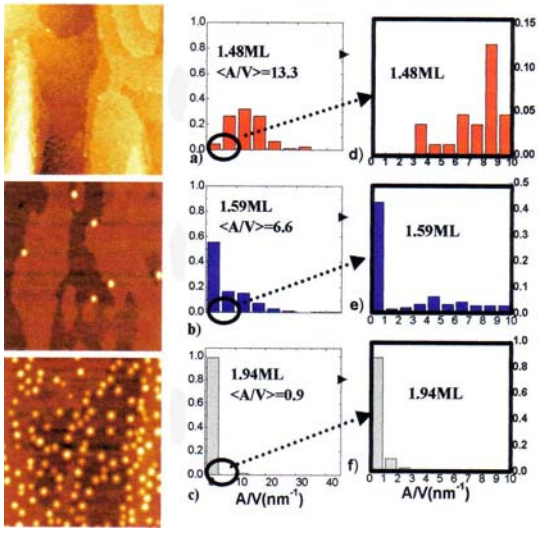

Fig. 14. AFM images, (500 x 500) nm^2 of InAs dots on GaAs (001) at 1.48 ML (a), 1.59 ML (b) and 1.94 ML (c). On the right, the histograms of the A/V ratio are presented.

6. Conclusions

Molecular Beam Epitaxy with RHEED is the only dynamic *in-situ* technique available to observe the 2D to 3D transition directly for InAs/GaAs QDs, but is limited in its sensibility. There is general acceptance that the RHEED pattern changes from streaked rods produced by a 2D surface to a spot pattern formed by transmission diffraction through 3D asperities with the addition of an incremental 0.1 ML (about 5×10^{13} atoms cm^{-2}). We have studied how structures such as mounds and steps can influence the evolution of quantum dots density, which has been considered as a function of the deposited film thickness.

The single-photon source uses semiconductor quantum dots that are 45 nm across and about 10 nm in height.

The dots have been created by depositing InAs on a GaAs substrate. The dots self-assembled because of strain after a critical InAs coverage value is reached.

The resulting dots typically have a height of 5.7 nm and show an emission at about 900 nm until a second critical coverage point is reached. Then the taller dots appear. By controlling the deposition around this second critical coverage, we were able to form a low density of these large dots, emitting radiation close to 1300 nm.

Acknowledgments

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