[10] Wautelet, M., Quenon, P., Jardin, A., *Semic. Sci. Technol.*3, 54-59, 1988.

THE MONITORING OF 2D-3D TRANSITION FOR InAs/GaAs (001) SELF-ASSEMBLED QUANTUM DOTS BY ATOMIC FORCE MICROSCOPY

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Abstract. *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 have performed a statistical analysis regarding quantum dot number density, identifying the existence of two separated distributions (for quasi-3D dots and 3D dots). We have observed 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^{10} cm^{-2} *to* 2.3×10^{10} cm^{-2}*) so as the 3D QDs become the prevailed structures. We can assume as critical coverage, 1.59 ML.*

1. Introduction.

The general importance and desirability for advancement of technology requires high degree of control of composition, size and structure. This is so critical to the area of nanostructures synthesis that self-assembly has been an increasing hallmark of this field. The growth of lattice-mismatch semiconductor systems such as InAs|GaAs has been known to produce structures as 3D islands by Stranski-Krastanow (SK) mode.

Typically, such islands don't form immediately on the substrate. In the first stage of the growth, a pseudomorphic thin layer is created which is elastically distorted. This pseudomorphic layer is named wetting layer (WL) and the islands form, practically, on its top.

The driving force for the self-assembly process during heteroepitaxial growth (the case of GaAs and InAs, the most commonly used pair of compounds) is the misfit between the crystal lattice of the growing layer and that of the substrate. This misfit of lattice constants is

about of 7% for InAs/GaAs and represents an important parameter that together with the growth temperature and growth rate influences the shape and average size of these dots. Quantum dots formed in Stranski-Krastanow growth mode are called self-assembled dots (SAQD). As practical applications of quantum dots we can talk about quantum dot lasers with predicted high performance such as a high characteristic temperature and a low threshold current.

In the following sections we first present the experimental techniques and the sample preparation process. An AFM that operates in tapping mode has been used to analyze a sample with variable coverage (from 1.45 ML to 2.2 ML). For the whole sample have been achieved about 500 images in order to monitor the complete evolution of the surface (from the wetting layer formation to the 3D island ripening). The 2D-3D transition is marked by a change of the RHEED pattern from streaky to spotty. The next step of the paper refers to the presentation of the evolution of the surface morphology, identifying the existence of different structures (2D islands, quasi-3D islands, 3D islands).

2. Experimental techniques and sample preparation.

In order to obtain InAs quantum dots on GaAs (001) substrate, we have used as growth method conventional solid source MBE equipped with reflection high energy electron diffraction (RHEED) for *in situ* monitoring of the growth.

2.1 Experimental techniques.

The MBE system used here has six effusion cells, three of them containing indium, gallium and arsenide. The effusion cells are independently heated until the desired material flux is achieved. Computer controlled shutters are positioned in front of each of the effusion cells to be able to shutter the flux reaching the sample within a fraction of second. The growth rate and film uniformity depend of the sample position relative at the cells.

The technique features of the growth chamber are such as the sample position for which $\alpha = 351^0$ corresponds to a maximum intensity of the beam flux but with minimum uniformity of the grown film while for $\alpha = 330^0$ the growth rate is minimum but we have a maximum film homogeneity. For improved layer uniformity, the sample holder is designed for continual azimuthally rotation of the sample, and is thus commonly termed the CAR. The CAR also has an ion gauge mounted on the side opposite the sample which can read the chamber pressure, or be placed facing the sources to measure beam equivalent pressure (BEP) of the material sources.

The sample used here has been grown without rotation and for a position ($\alpha = 340^0$) situated between the two limits presented previously. In this way we have obtained a coverage gradient along of one of the sample directions ([110]).

The knowledge of the growth rate dependence of the sample position (α) gives us the coverage evolution (Φ) along of the one side of the sample ([110]). In the range of $\alpha = 330^0$-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.

2.2 Sample preparation.

In order to growth 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 know 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 this order 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^{0}C) with arsenic over pressure until diffraction pattern (2 x 4 reconstructions) appears showing that the oxide has been removed on the surface.

Next, prior to InAs deposition, we have grown a GaAs buffer of 750 nm on the GaAs (001) substrate at 590^{0}C and at a rate of 1 μmh^{-1}, J_{As}/J_{In} flux ratio being of 10/1. After 10 min post-growth annealing, the temperature is lowered to 500^{0}C for InAs deposition. The InAs is evaporated at a rate of 0.029 ML (monolayer)/s. The In delivery time is cycled in 5s of evaporation followed by 25s of growth interruption until the given thickness is reached.

Rheed analysis gives us information about 2D-3D transition by changing the diffraction pattern from streaky to spotty. Because the sample used in this work presents a coverage gradient, Rheed analysis show some zones where the 2D-3D transition has taken place (spotty pattern) and others where the film is a 2D one (streaky pattern) (see figure 1).

Fig. 1. Representation of an InAs/GaAs (001) Rheed pattern.

3. Results and discussions.

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 (see figure 2). 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, quasi-3D islands (quasi-3D QD) and 3D islands (3D QD).

Statistical analysis of quantum dots

In order to perform a statistical analysis regarding the quantum dot number density, it is necessary to give quantitative criteria that allow us to distinguish between quasi-3D dots and 3D dots. The ratio A/V (A-area; V-volume) has been used as quantitative criteria in this way: we have considered that quasi- 3D dots are structures for which the ratio A/V is higher than $3nm^{-1}$, while for 3D dots the ratio value is between 0 and $2nm^{-1}$.

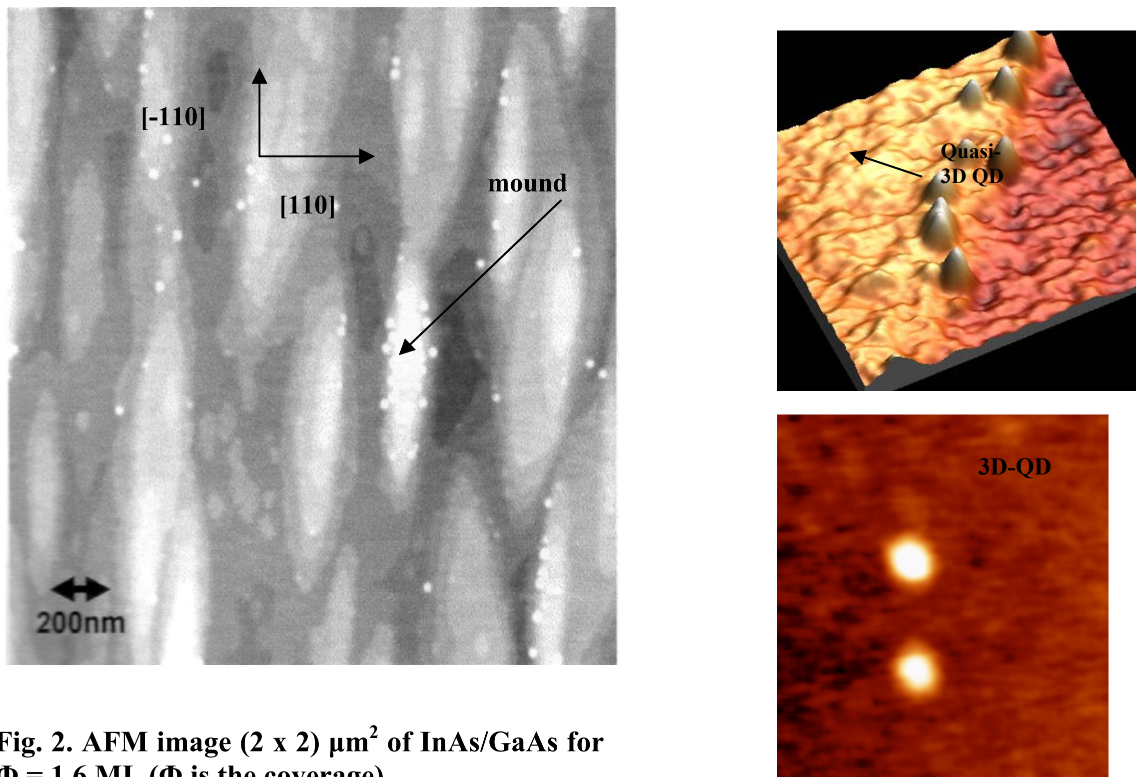
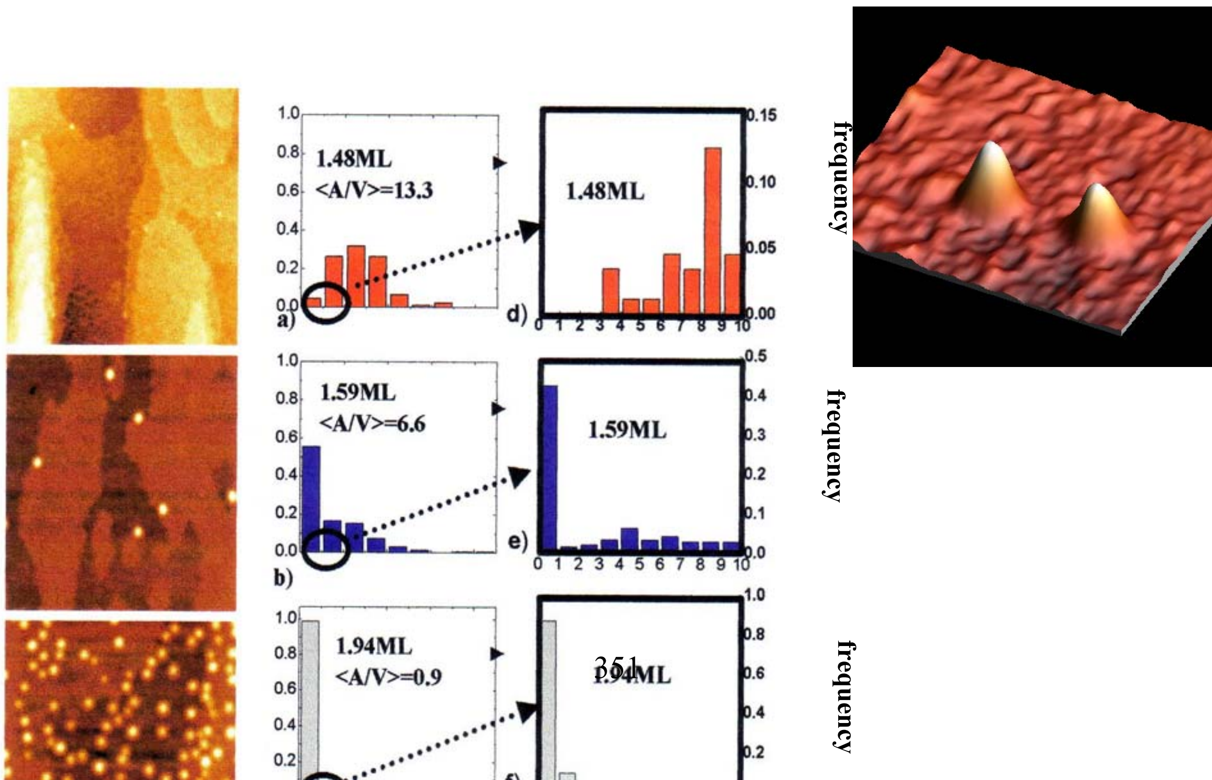


Fig. 2. AFM image (2 x 2) μm^2 of InAs/GaAs for Φ = 1.6 ML (Φ is the coverage).



We identify two clearly separated distributions for quasi-3D dots and 3D dots. 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 4). 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. 5x 10^{10} dot/cm^2 represents the saturation value of the 3D QDs density. Beyond this value won't appear new dots due to the saturation of the possible nucleation sites. 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 x 10^{10} cm^{-2} to 4.3 x 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 x 10^{10} cm^{-2} to 2.3 x 10^{10} cm^{-2}) so as 3D QDs become the prevailed structures. We can assume as critical coverage, 1.59 ML.

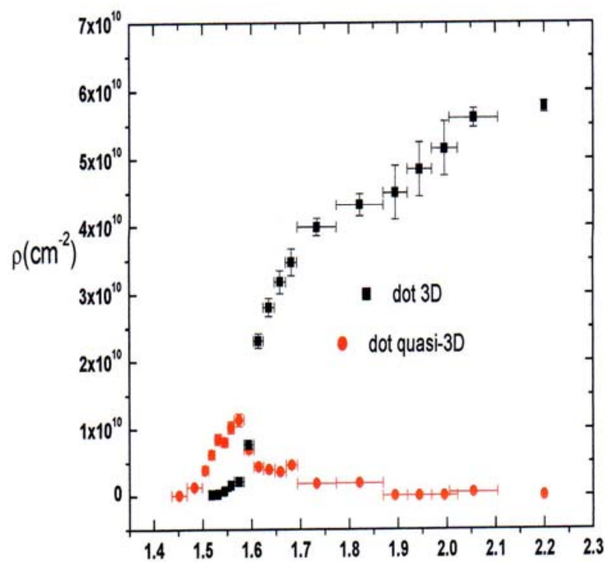


Fig. 4. Quantum dots number density for coverage between 1.45 ML and 2.2 ML.

4. Conclusions

It is worth noting that by growing lattice-mismatched systems it is possible to obtain 3D self-assembled, coherent nanostructures without the use of sophisticated nanolithographic

techniques. Critical thickness of InAs represents an important parameter that depends both on the growth conditions and on the lattice mismatch of the heterostructures [2]. Knowing this parameter it is possible to control the formation of QDs below the critical thickness at any given region by adding a strain layer in that predefined region [1]. In this way will exist an alignment of the QDs that represents an important characteristic for the new optoelectronic devices.

From the monitoring of the 2D-3D transition by atomic force microscopy analysis we have assumed as critical thickness for InAs the value of 1.59 ML.

5. Acknowledgments.

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References

[1] Lee, B. C., Lin, S. D., Lee, C. P., *Appl. Phys. Lett.,* **80** (2002).
[2] Franchi, S., Trevisi, G., Seravalli, L., Frigeri, P., *Progr. in Cryst. Growth,* **47** (2003), 166-195.
[3] Voigtländer, B., Kawamura, M., Paul, N., Cherepanov, V., *J. Phys. Condens. Matter* **16** S1535-S1551, 2004.
[4] Arciprete, F., Balzarotti, A., Fanfoni, M., Motta, N., Pattela, F., Sgarlata, A., *Vacuum Sci. & Tech.* **, 3,** 71-98, 2001.
[5] F. Pattela, A. Sgarlata, F. Arciprete, S. Nufris, P.D. Szkutnik, E. Placidi, M. Fanfoni, N. Motta and A. Balzarotti, *J. Phys. Condens. Matter*, **16,** S1503-S1534, 2004.
[6] Krzyzewski, T. J., Joyce, P. B., Bell, G. R., Jones, T. S., *Phys. Rev.* B **66,** 121307(R) 2002.
[7] Ramachandran, T. R., Heitz, R., Chen, P., Madhukar, A., *Appl. Phys. Lett.* **70,** 640, 1997.
[8] da Silva, M. J., Quivy, A. A., Gonzalez-Borrero, P. P., Marega, E., *J. Cryst. Growth,* **241** 19, 2002.