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Three-dimensional ordering in self-organized (In,Ga)As quantum dot multilayer structures

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Molecular beam epitaxy (MBE) grown $In_{0.5}Ga_{0.5}As$/GaAs multilayer structures with quantum dots chains (QDs), obtained under different growth conditions, were investigated by high-resolution X-ray diffractometry (HRXRD) and AFM. It was determined that self-organized epitaxial growth of $In_{0.5}Ga_{0.5}As$/GaAs can lead to the formation of three-dimensional quantum-dot crystals with triclinic (distorted cubic) unit cell. The mechanisms of QD's ordering in dependence on As flux are analyzed.

1 Introduction An application of self-organized QD systems in the device production requires frequently not only the size and form homogeneity of QDs but also their clear spatial arrangement [1].

However, it is a big problem to obtain such an ideal structure with preliminary given properties. It is known that lithographic techniques allow one to reach the necessary vertical and lateral ordering of QDs [2–4], but they are expensive enough. Moreover, for the patterned surfaces the appearance of growth defects is very probable [5]. On the other hand, the classical Stranski–Krastanow growth mode of InGaAs QDs on GaAs (001) is limited by chaotic character of the islands arrangement [6, 7]. Nevertheless, the interaction between QDs during growth can improve their homogeneity and lateral periodicity.

There is a certain success in solving this problem. For example, the using of vertical stacking of QDs at relatively high growth temperature [6] gave an opportunity to obtain ordered chains of QDs [2, 3]. It is known that ordered QDs successfully grow on high-index substrates [8]. It is also possible to improve optical properties of InAs islands by using As_2 molecular flux instead of As_4 [9, 10]. But these works did not answer the question of QD's ordering.

High-resolution X-ray diffraction (HRXRD) is a non-destructive method for periodical nanostructures characterization. Application of this technique allows to investigate the layers deformation and the ordering degree of QDs. It is known that HRXRD was successfully applied to investigate the spatial ordering of QD's in PbSe/PbEuTe [11].

In this article, we present the study of three-dimentional ordering of InGaAs QDs, embedded in a multilayer GaAs matrix.

We used HRXRD and atomic-force microscopy (AFM) to clarify that self-organized epitaxial growth of $In_{0.5}Ga_{0.5}As$/GaAs leads to the formation of three-dimensional crystals of QDs, which are aligned in triclinic unit cell with vertical stacking sequence. The mechanisms of QD's ordering under different As flux were also analyzed.

2 Experimental procedure $In_{0.5}Ga_{0.5}As$/GaAs structures were grown on semi-insulating GaAs (001) substrates by molecular-beam epitaxy (MBE) in As_2 and As_4 gas flux. After removing of oxidized layer from the substrate surface, 0.3 μm GaAs buffer layer was grown at 580 °C. Then the temperature was reduced to 540 °C for the growth

of 15 period (2.5 nm) $In_{0.5}Ga_{0.5}As$/(60 monolayers (MLs)) GaAs multilayer structure.

The growth rate of GaAs and InAs was 0.24 and 0.27 ML·s^{-1}, respectively. The top InGaAs layer with QDs was uncapped to perform AFM investigation of surface morphology. The samples with In content $x = 0.5$ were grown at the same conditions in As_2 and As_4 gas flux with the ratio $As_2(As_4)/(Ga + In)$ equal to 15:1.

All the measurements were performed by high-resolution X'Pert Pro MRD XL diffractometer equipped with four-fold Ge(220) monochromator and three-fold analyzer of the same type using Cu K_α radiation. Symmetrical 004 and asymmetrical 113, 404, 224 scattering geometries were used.

The simulation on the basis of various models was performed to extract the structural characteristics and the strain field distribution from the observed patterns [12–14].

Surface morphology was analyzed by NanoScope IIIa Dimension 3000™ with tip radius less then 10 nm.

3 Results and discussion Figure 1 presents AFM images indicating an anisotropic character of QDs alignment into the chains oriented along [1$\bar{1}$0] crystallographic direction. But there is a considerable difference in size and lateral alignment of QDs for the samples grown in As_2 and As_4 fluxes. For example, the average height of QDs grown in As_4 flux is $\approx 3 \pm 1$ nm, whereas this value is $\approx 10 \pm 2$ nm for QDs grown in As_2. Moreover the first QDs are of ellipsoidal form with the longer axis along [1–10] direction (insertion in Fig. 1a). The remains of wetting layer between QDs along [110] are observed. QDs grown in As_2 flux (insertion in Fig. 1b) have rounded basis and are well separated. We used topometric Fourier analysis to estimate the average lateral distance between QDs. The square root values of 2D fast-Fourier transform (FFT) module of surface patterns are presented on Figs. 1b–d. The blured FFT maxima of the first order and their positions for QDs grown in As_4 (Fig. 1) indicate that the QDs ordering degree for this samples is worth in compare to those grown in As_2 flux (Fig. 1d). The distance between QDs inside the chains and between them is greater for QDs grown in As_2 flux.

The arrangement of FFT maxima reveals the existence of the statistically averaged rectangle QDs lattice on the surface of the samples oriented along [1$\bar{1}$0] and [110] directions (insertions in Fig. 1a and c). The clear FFT maxima of the first and second order along [110] direction indicate that QDs are better ordered along this direction.

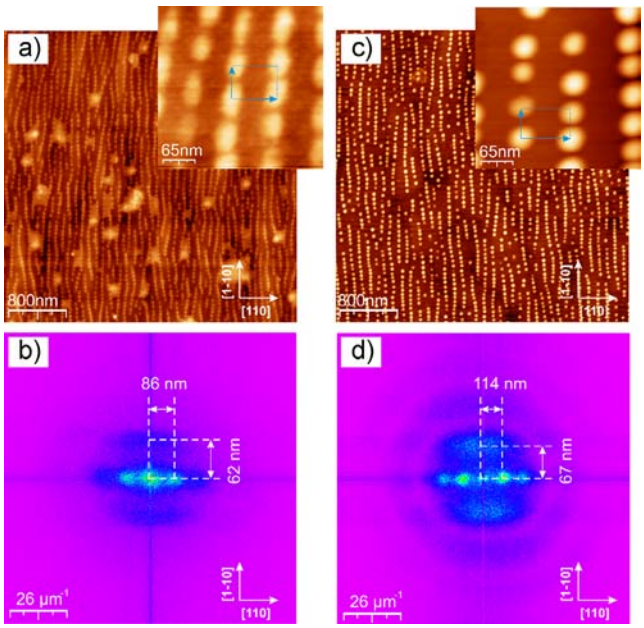

Figure 1 (online color at: www.pss-a.com) AFM images of $In_{0.5}Ga_{0.5}As$/GaAs for different As fluxes (a) – As_4, (c) – As_2. 2D FFT results (c),(d). Averaged 2D unit cells formed by QDs are shown on inserts. The height range equals 18 nm.

The following study of spatial (three-dimensional) ordering of InGaAs QDs was provided by X-ray diffraction, which allows to analyze the averaged X-ray scattering from the whole superlattice volume with statistical ensemble of the ordered QDs.

From the symmetrical 004 reflection curves (RCs) we've obtained the structural parameters of the samples (deformations, vertical period of SL), presented in Table 1. One can see that the deformation level in the structures grown in As_2 is much lower in compare with As_4. But in spite of a lot of experimental data the RC's measurement did not answer the question about the QDs ordering in the samples. That is why we have provided the measurement of reciprocal space maps (RSMs) in symmetrical 004 and asymmetrical 113, 404, 224 scattering geometries, presented on Fig. 2. The existence of lateral satellites for the crystallographic directions [110], [1$\bar{1}$0], [100] confirms the ordering of QDs.

Table 1 Deformation anisotropy in GaAs and InGaAs layers and the parameters of a QD unit cell (* strain values in [110] and [1$\bar{1}$0] crystallographic directions).

flux	deformations $\varepsilon_z \times 10^{-3}$		average vertical period of multilayer structure d (nm)		QDs unit cell parameters					
	GaAs*	InGaAs*	nominal		a (nm)	b (nm)	c (nm)	α (degr)	β (degr)	γ (degr)
As_4	1.71/1.48	19.2/17.5	18.6	18.5	84 ± 3	56 ± 3	18.5 ± 0.2	87 ± 2	85 ± 2	83 ± 2
As_2	1.41/1.16	18.4/17.0	18.6	18.8	100 ± 3	66 ± 3	18.8 ± 0.2	82 ± 2	79 ± 2	87 ± 2

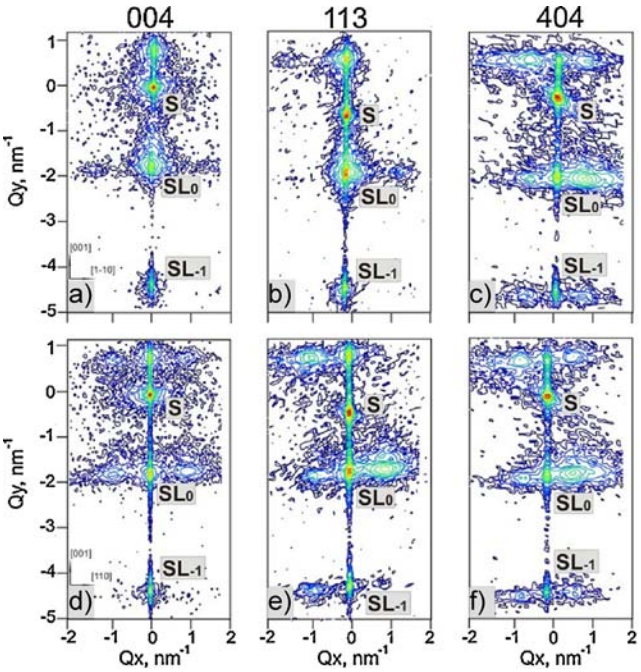

Figure 2 (online color at: www.pss-a.com) RSMs for the sample with As_2 flux 004, 113, 404 reflections; a, b – (110); d, e – $(1\bar{1}0)$; c – (100); f – (010) scattering planes. S denotes substrate peak, SL ± n – SL coherent satellites.

For the detailed investigation of QD's spatial ordering 004 RSMs close to SL_0 (the zeroth order vertical satellite) in various azimuthal directions of the scattering plane were measured (Fig. 3). We have found that the distance between lateral satellites is azimuthally dependent. This is because the diffraction plane differently cuts the QD's system. Moreover for the $\Phi = 45^\circ$ and $\Phi = 135^\circ$ azimuthal directions the SL lateral periods are different. This states that lateral QD's unit

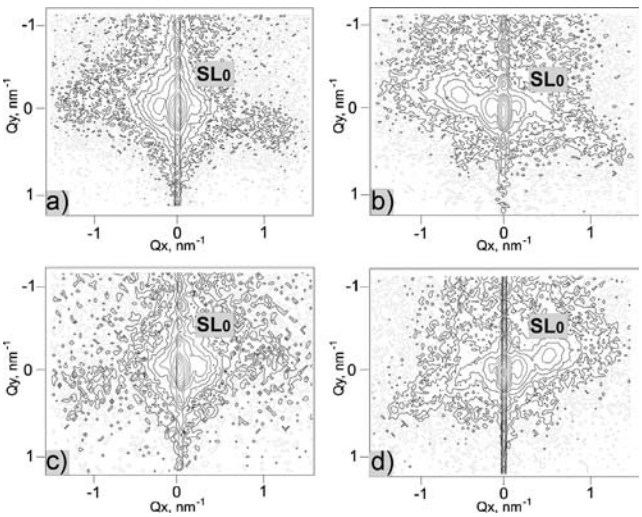

Figure 3 004 RSMs near SL0 for $\Phi = 0^\circ, 45^\circ, 90^\circ, 135^\circ$ azimuthal directions relative to $[1\bar{1}0]$ direction.

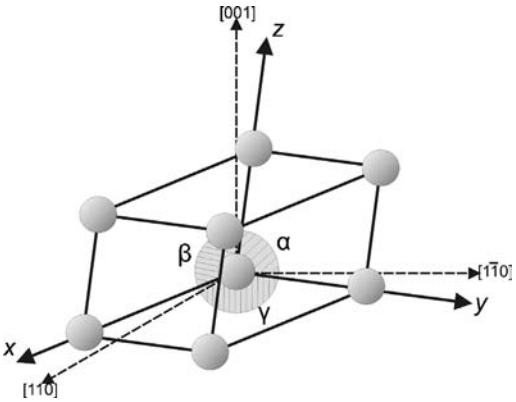

Figure 4 Schematic picture of a three-dimensional QD crystal.

cell is skew-angular and inclined to the $[1\bar{1}0]$ direction with 3–8° angle, which is also proved by AFM (Fig. 1).

From the distances between lateral satellites and their inclination relative to the growth direction we have obtained the SL lateral period and the angles of QD's vertical stacking for different azimuthal directions [15, 16]. On the basis of the obtained data, we succeeded to construct the spatial unit cell of QDs (Fig. 4). In our case, it is primitively triclinic (distorted cubic) lattice with parameters given in Table 1.

An application of As_2 molecular beam instead of As_4 for QD's separation along the chain could be explained by surface diffusion of adatoms. As_2 molecules do not need to break the bond on the GaAs surface to join with Ga [17]. That's why they can move for a longer time on the Ga surface without energy change and find the preferable place to join with Ga before desorbing into vapor phase [18]. Thus in As_2 molecular flux Ga and In diffusion lengths decrease and increase, respectively. This reduces the intermixing of InGaAs along $[1\bar{1}0]$ direction while QDs are formed. As a result, we obtain a better spatial ordering and size and shape homogeneity of QD's which improves their optical properties.

Finally, we want to point to the peculiarity which was observed on 004 RSMs from the samples grown in As_2 flux. Independently on azimuthal direction of diffraction plane there are two systems of lateral maxima near SL_0. One of them, with varying interval, corresponds to the QD's ordering. For another one the interval is kept constant for all the azimuthal directions of diffraction plane (Fig. 5).

We suppose that the appearance of these satellites is given by the presence of correlated system of misfit dislocations [19] with low density $\rho d << 1$, where ρ – dislocations density, d – dislocation layer thickness. When the density increases the lateral maxima of the higher order disappear and the main SL peak broadens.

The appearance of misfit dislocations decreases the stress in the SL system [20]. But in our case, their low amount cannot essentially influence the deformation in compare to the sample grown in As_4.

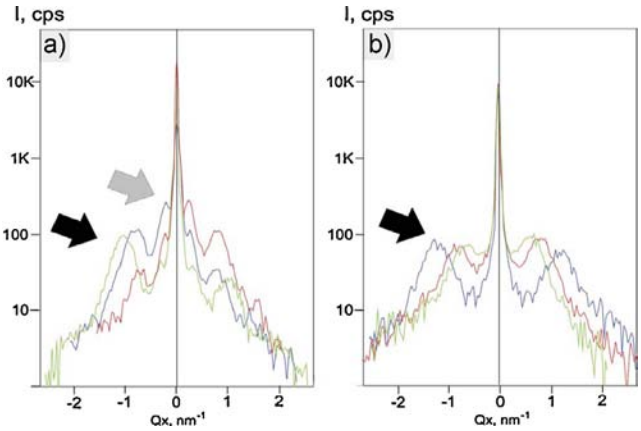

Figure 5 (online color at: www.pss-a.com) ω-scans near the 0th order coherent satellite for various azimuthal directions of diffraction plane. a) As_2 flux, b) As_4 flux. Black arrow – lateral satellites, gray arrow – misfit dislocations satellites.

4 Conclusions We determined that for InGaAs/GaAs(001) material the self-assembled epitaxial growth leads to the formation of three-dimensional crystals of QDs. In these crystals, the QDs are aligned into triclinic unit cell with vertical stacking sequence.

The realization of spatial ordering depends on the correlation of As_2/As_4 flux and In content in wetting layer. For As_2, the spatial triclinic QDs lattice is more inclined relative to the [110] direction and the ordering degree along the chains is higher. These results show that a good vertical and lateral ordering of QDs could be directly achieved on the flat surface by varying the growth conditions.

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References

[1] J. Phillips, J. Appl. Phys. **91**, 4590 (2002).
[2] H. Lee, J. A. Johnson, J. S. Speck, and P. M. Petroff, J. Vac. Sci. Technol. B **18**, 2193 (2000).
[3] Y. Nakamura, O. G. Schmidt, N. Y. Jin-Phillipp, S. Kiravittaya, C. Muller, K. Eberl, H. Grabeldinger, and H. Schwizer, J. Cryst. Growth **242**, 339 (2002).
[4] C. K. Hyon, S. C. Choi, S. H. Song, S. W. Hwang, M. H. Son, D. Ahn, Y. J. Park, and E. K. Kim, Appl. Phys. Lett. **77**, 2607 (2000).
[5] S. Birudavolu, N. Nuntawong, G. Balakrishnan, Y. C. Xin, S. Huang, S. C. Lee, S. R. J. Brueck, C. P. Hains, and D. L. Huffaker, Appl. Phys. Lett. **85**, 2337 (2004).
[6] V. A. Shchukin, N. N. Ledentsov, and D. Bimberg, Epitaxy of Nanostructures (Springer, Berlin, 2004).
[7] D. Leonard, M. Krishnamorthy, C. M. Reaves, S. P. Denbaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[8] P. M. Lytvyn, V. V. Strelchuk, O. F. Kolomys, I. V. Prokopenko, M. Ya. Valakh, Yu. I. Masur, Zu. M. Wang, G. J. Salamo, and M. Hanke, Appl. Phys. Lett. **91**, 173118 (2007).
[9] T. Sugaya, T. Amano, and K. Komory, J. Appl. Phys. **100**, 063107-1 (2006).
[10] T. Sugaya, S. Furue, T. Amano, and K. Komory, J. Cryst. Growth **301-303**, 801 (2007).
[11] G. Springholz, V. Holy, M. Pinczolits, and G. Bauer, Science. **282**, 734 (1998).
[12] O. M. Yefanov and V. P. Kladko, Met. Phys. New Tech. **28**, 227 (2006) (in Russian).
[13] D. K. Bowen and B. K. Tanner, High-Resolution X-ray Diffractometry and Topography (Taylor & Francis, London, 1998).
[14] U. Pietsch, V. Holy, and T. Baumbach, High-Resolution X-ray Scattering from Thin Films to Lateral Nanostructures (Springer, New York, 2004).
[15] V. P. Kladko, M. V. Slobodian, V. V. Strelchuk, O. M. Yefanov, V. F. Machulin, Yu. I. Mazur, Zh. M. Wang, and G. J. Salamo, Phys. Status Solidi A **204**, 2567 (2007).
[16] C. Giannini, L. Tapfer, Y. Zhuang, L. DeCaro, T. Marschnerand, and W. Stolz, Phys. Rev. B **55**, 5276 (1997).
[17] M. H. Son, D. Ahn, Y. J. Park, and E. K. Kim, Appl. Phys. Lett. **77**, 2607 (2000).
[18] A. Ishii, K. Seino, and T. Aisaka, J. Cryst. Growth **236**, 511 (2002).
[19] V. M. Kaganer, R. Kohler, M. Schmidbauer, and R. Opitz, Phys. Rev. B **55**, 1793–1810 (1997).
[20] D. C. Houghton, D. D. Perovic, J.-M. Baribeau, and G. G. Weatherty, J. Appl. Phys. **67**, 1850 (1990).