X-ray study of temperature dependent growth of InAs/AlAs(001) quantum dots.

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We report about the X-ray analysis of non-capped InAs/AlAs(001) quantum dot systems grown at temperatures ranging from 480 °C up to 530 °C. A constant amount of InAs has been deposited resulting in a growth stage where coherently strained dots and plastically relaxed clusters coexist. It is found that with increase of deposition temperature the average size of elastically strained dots increases without changes of their chemical composition and surface density. The observed process is in accordance with the InAs volume decrease stored in plastically relaxed clusters. The results establish the crucial role of strain-induced material intermixing between strained InAs dots and AlAs substrate over the investigated growth temperature range.

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I. INTRODUCTION

Ongoing interest is devoted to the research of semiconductor nanostructures, such as e.g. quantum dots. Due to their small size quantum dots exhibit a zero-dimensional quantum confinement of electrons causing discrete energy states [1]. Control of their size allows the precise tuning of their emission wavelength which is reflected by a great industrial interest on quantum dots as core elements for diverse opto-electronic applications [2, 3].

The molecular beam epitaxy (MBE) of group III-V lattice mismatched semiconductor heterostructures is currently one of the most widely used techniques for quantum dot fabrication [4–6]. This technique exploits the Stranski-Krastanov (SK) growth mode [7]. In SK-mode, the high lattice mismatch between the substrate and deposited material leads to spontaneous reduction of the strain energy changing from the layer-by-layer (wetting layer) growth to the formation of 3D quantum dots [8].

The MBE SK-quantum dot growth has been successfully realized and systematically studied in Ge/Si(001) and InAs/GaAs(001) systems with initial lattice mismatch of 4% and 6.7%, respectively [5, 9, 10]. In these systems the produced quantum dots were shown to possess a narrow size distribution, being elastically strained and dislocations free [11].

For InAs/GaAs(001) dots, it was also found that for moderate In fluxes - ≈0.05 monolayers (ML) per second - the conditions for quantum dot formation require growth temperatures between 470 °C - 530 °C [12].

Strong intermixing between quantum dots and substrate materials was also reported [13, 14]. Intermixing plays an important role in the quantum dot formation. It influences the dot composition and strain state and changes the optical emission of quantum dots. Being a complex function of deposition parameters and mismatch-strain the intermixing in quantum dots is difficult to estimate. A model proposed by Heyn et al. [12] accounts for kinetic exchange processes without consideration of strain and describes well the intermixing in the InAs/GaAs(001) system.

InAs/AlAs(001) is another quantum dot system being currently under investigation. The main advantage of this system is the typical dot size being at least three times smaller than that for the InAs/GaAs(001) system [15, 16]. This leads to more pronounced quantum confinement and accordingly higher emission energies accompanied by the one order of magnitude higher island surface densities which make InAs/AlAs(001) system favorable for opto-electronic applications [17]. In particular, for optical experiments the higher emission energies are attractive because they lay in more sensitive regimes of spectrometers and detectors.

In contrast to InAs/GaAs(001) the growth of InAs on AlAs(001) is less studied and presently not well understood. In comparison to InAs/GaAs(001), InAs/AlAs(001) quantum dots have been shown to exhibit significantly different growth dependence of their morphological properties [18, 19] although it has similar initial lattice mismatch of 6.7% relative to the substrate.

In this paper we present results on InAs/AlAs(001) quantum dots produced at different growth temperatures. We applied grazing incidence X-ray diffraction (GIXRD) to study dot structure and composition. Atomic force microscopy (AFM) was used to provide local information about quantum dot morphology.

II. PREPARATION AND EXPERIMENTAL DETAILS

For our studies we use an amount of 2.7 ML InAs deposited on AlAs(001). At this coverage, in addition to small elastically strained dots some of the deposited material has already formed very large quantum dots containing defects from strain relaxation [20–22]. We assume the large relaxed dots to result from coalescence [23] and thus call them coalesced clusters.

In the coalescence regime the presence of relaxed InAs

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clusters can serve as a reference for the study of coherently strained quantum dots fabricated at different temperatures. This allows us to resolve the temperature dependence of dot material composition which is the main subject of this study.

The AlAs substrate layers are grown on commercial GaAs(001) 2-inch wafers. After the oxide layer was desorbed at 582 °C and a stable GaAs 2x4 surface reconstruction was observed using reflection high energy electron diffraction (RHEED), the temperature was lowered down to 530 °C and a 150 nm thick AlAs layer was deposited. RHEED reflections demonstrated a good surface quality of AlAs(001) layer.

The three InAs/AlAs(001) quantum dot samples were MBE-grown at 480 °C, 500 °C, and 530 °C, correspondingly. Quantum dot formation was in-situ monitored in RHEED. For all samples 2.7 ML of InAs were deposited at a rate of 0.04 ML/s. Considering the temperature and flux dependent desorption rates [24] the time of InAs deposition was adjusted separately for each sample in order to achieve constant InAs surface coverage.

AFM viewgraphs of the samples are shown in Fig. 1. Obviously, all three quantum dot ensembles reached the stage where plastically relaxed InAs clusters with low surface densities are formed. After preparation, the samples were carried to the BW2 wiggler beam-line (HASYLAB, DESY) in a protective environment preventing their long-time exposure to ambient air. Grazing-incidence X-ray diffraction experiments on all samples were carried out at 10 keV synchrotron radiation energy in X-ray compatible chamber filled with He atmosphere.

III. RESULTS AND DISCUSSION

For all samples the line scans in the radial [110] reciprocal space direction through the (220) GaAs Bragg reflection for samples grown at 480 °C, 500 °C, and 530 °C. Label "I" marks the substrate peak position, "II" - marks signal from elastically strained islands, "III" - marks signal from plastically relaxed clusters. "C"-labelled arrows mark dots short-order correlation peak positions.

The intensity on the line scans are normalized by the intensity of the respective value of peak III (horizontal dashed line in Fig. 2). It can be seen that with increase of deposition temperature the relative intensity from dislocated clusters decreases. Also, from comparison of Fig. 1(a-c) it can be seen that the surface density of plastically relaxed clusters decreases with increasing growth temperature. This means, that with increase of deposition temperature coherently strained dots incorporate more material. It should be noticed that for all samples the position of peaks marked by 'C' in Fig. 2 - occurring due to the short-range inter-dot correlation effects between densely-packed strained dots - does not change. This point to the fact that the average surface density of coherently strained dots does not depend on the deposition temperature. Since the amount of deposited indium was fixed and the plastically relaxed clusters contain less material at higher growth temperatures a size increase of the coherently strained dots can be expected. The line scans transversal to the [110] reciprocal space direction through the point \( Q_{[110]} = Q_{[010]} = 1.95 \) GaAs r.l.u. [27] (in reciprocal space this point lies on the intensity profile II stemming from coherently strained dots, see Fig. 2) for all three samples are shown in Fig. 3. The signals in Fig. 3 are shown rescaled to their peak values to enable their comparison. It can be seen that with increase of deposition temperature the full width at half maximum (FWHM) of the diffraction peak decreases. This indicates that the average lateral dots size increases. AFM viewgraphs of strained dots presented in Fig. 4 for samples grown at 480 °C and 530 °C confirm our XRD data.
The intensity distributions along [100]-reciprocal space directions in the vicinity of the (400) and (200) AlAs Bragg reflections were recorded for each sample. The ratio between (400) and (200) intensity profiles for each sample is sensitive to the chemical composition variations (AlAs concentration) along the dot’s vertical axis [26]. For samples grown at 480 °C and 530 °C the corresponding plots are presented in Fig. 5. As in Fig. 2, here the same three main intensity distributions can be distinguished. The quantitative analysis of the (400)/(200) intensity profiles ratio from strained dots (peak 'II') reveals that within only few percent deviation the average dot’s AlAs-content is equal for both samples. These observations directly prove that quantum dots possess equal chemical composition in the whole investigated range of growth temperatures. Furthermore, in the reciprocal space region 'III', where signal from plastically relaxed clusters dominates, the (400) and (200) intensities are quantitatively identical. It reveals clusters composition close to that of pure InAs. In the reciprocal space region 'II' in Fig. 5 where the signal from strained dots dominates the (200) intensity profile is weaker in comparison to (400) intensity distribution. It means that the strained dots are composed not from pure InAs but from the In$_x$Al$_{1-x}$As alloy [26]. From our previous studies the composition of strained dots grown at equal deposition conditions as the 500 °C-sample studied in current paper is close to In$_{0.8}$Al$_{0.2}$As [25]. Taking into account the observed compositional equality of all investigated samples, the determined Al-content can be correlated to the strained dots in all three investigated systems.

### IV. CONCLUSIONS

In this paper the structure and composition characterization by X-ray methods together with local topography analysis of InAs/AlAs(001) quantum dot systems are presented. Samples were grown in the 480 °C - 530 °C temperature range with constant amount of deposited InAs in the coalescence regime, i.e. where coherently strained dots and plastically relaxed clusters coexist. It is found that with increase of deposition temperature the composition of the elastically strained dots remains constant close to In$_{0.8}$Al$_{0.2}$As while the dot dimensions increase. The constant Al-content in strained InAs dots at strongly different deposition temperatures is related to a temperature independent interdiffusion between quantum dots and substrate material. This observation shows that in the InAs/AlAs system the growing strained dots respond to differences in deposition temperature exclusively by changing their size. This is in contrast to the InAs/GaAs(001) quantum dot system, where thermally activated Ga-interdiffusion is assumed to cause the intermixing [12].

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[27] 1 GaAs r.l.u. = \frac{d_{GaAs}^{GaAs}}{d_{GaAs}^{(100)}} = 1.11\AA^{-1}