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Characterisation of droplet-epitaxial GaAs/AlGaAs quantum dot and quantum ring systems using grazing incidence X-ray diffraction

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ABSTRACT

We study with grazing incidence X-ray diffraction GaAs quantum dot (QD) and quantum ring (QR) systems grown via droplet epitaxy. The measurements clearly reveal that QDs and QRs grown on AlGaAs(0 0 1) substrate crystals are single crystalline with the same crystal orientation as the substrate, and are free of elastic strain. The QD and QR systems are found to laterally arrange in chains of closely spaced objects with preferential orientation in ⟨1 1 0⟩ crystallographic directions. We find that for the QD system this ordering is anisotropic, showing intra-chain correlation only in [1 –1 0] direction. In the QR system the intra-chain arrangement is equal in both [1 1 0] and [1 –1 0] directions. The differences in the ordering behaviour of QD and QR chains are believed to originate from the different speeds of As incorporation in the Ga droplets during their crystallization at different As pressures.

1. Introduction

Quantum dots are promising candidates for future devices in optoelectronics and memory applications, as well as for basic research [1]. The growth of self-assembled quantum dots via droplet epitaxy technique is a recent alternative [2–11] to the strain-induced Stranski–Krastanov (SK) QD formation based on lattice mismatch between substrates and epilayers [12,13]. In droplet epitaxy, group III metallic clusters are deposited firstly on the single-crystalline semiconductor surface without supplying group V atoms. In our study, Ga droplets are formed by nominally 3.75 monolayers (ML) gallium deposition. Afterwards, coherent quantum dots are formed by a crystallization step in which arsenic is offered. The high crystalline quality of droplet QDs (absence of deep traps) was shown in PL studies [14,15]. The crystalline structure of QDs grown by droplet epitaxy is assumed to be prescribed by the substrate. Interestingly, the shape of the nanostructures produced with droplet epitaxy can be controlled during crystallization by the arsenic background pressure [16]. Simultaneously, the question arises as to how the mutual surface arrangement of the produced nanostructures changes under the influence of As pressure.

Phase and arrangement analysis of 3D nanostructures using the grazing incidence (GI) X-ray experiments, such as diffraction (GID) and small-angle scattering (GISAXS) with high-intensity synchrotron radiation, have demonstrated their effectiveness as tools for analysis in SK systems [13,17–23]. Recent GID experiments on the binary QD molecules (pairs of closely spaced dots) produced by droplet epitaxy provided the first evidence for the strain-free nature and single-crystalline quality of the nanostructures [17]. The system investigated in Ref. [17] consists of molecule-like double-dot objects produced by crystallization at relatively high temperature, at which a strong material redistribution takes place. In contrast, we

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study QD/QR system crystallized at low temperature, at which a much slower material diffusion is expected. In addition, for our studies a much higher surface density of nanostructures is used, at which different inter-dot correlation effects may be expected. We use GID and GISAXS tools for the analysis of arrangement effects in the GaAs/InGaAs(0 0 1) quantum dot (QD) and quantum ring (QR) systems with high surface density. Atomic force microscopy (AFM) is used for shape and surface density analysis of the produced structures as well as for the visualisation of observed ordering effects.

2. Samples

The samples were grown in a solid-source MBE system equipped with a two-zone valved arsenic cracker source. The group III elements were evaporated from standard Knudsen effusion cells. After thermal desorption of surface oxides at $T = 630\,^{\circ}\mathrm{C}$, a thin GaAs buffer was grown followed by 100 nm $Al_{0.31}Ga_{0.69}As$ at $T = 610\,^{\circ}\mathrm{C}$. The growth rates were $F_{Ga} = 0.43\,\mathrm{ML\,s^{-1}}$ and $F_{Al} = 0.19\,\mathrm{ML\,s^{-1}}$. Afterwards, the substrate temperature T was lowered to 200 °C without arsenic flux. Keeping the arsenic valve closed, 3.75 ML of Ga were deposited, followed by 60 s annealing at the same temperature. Supplying arsenic by the valved cracker at $T = 200\,^{\circ}\mathrm{C}$, the crystallization step was performed. For sample A, we applied a high arsenic pressure $P = 6.6\mathrm{E}{-5}$ Torr measured by an ion gauge. For sample B an arsenic pressure $P = 1.4\mathrm{E}{-5}$ Torr was applied. Before unloading the samples, a further annealing step for $t = 10$ min at $T = 350\,^{\circ}\mathrm{C}$ was performed. Immediately after transfer to the ambient air the samples were inspected with atomic force microscopy. Thereafter, samples were transferred in protective nitrogen atmosphere to the HASYLAB/DESY, Hamburg, where they were investigated in He atmosphere using a grazing-incidence setup (wiggler beam-lines BW2 and BW4 for GID and GISAXS experiments, correspondingly). The experimental conditions for GID and GISAXS investigations were incoming photon energy 10 keV and incident angle 0.2°.

3. Results and discussion

Fig. 1 shows the topographic AFM images of both samples. A narrow lateral size distribution of the nanostructures can be observed. For both samples, surface densities of $2 \times 10^{10}\,\mathrm{cm^{-2}}$ are estimated. On the scale of the AFM viewgraphs there are no obvious differences in the lateral arrangement for both nanostructures. The line-scans over the single island and ring nanostructure are shown in Figs. 1(c) and (d), respectively. In contrast, the morphological difference of both structures is obvious.

We investigate the lateral arrangement in the QD/QR systems using the GID tool. For dot and ring samples the experimental in-plane GID 2D reciprocal space patterns measured in the vicinity of the (2 2 0) GaAs Bragg reflection are shown in Figs. 2(a) and (c), respectively. It can be seen that for both samples the intensity distributions are symmetric relative to the (2 2 0) GaAs Bragg peak position. The diffuse intensity variations are much broader than the natural width of bulk single crystal Bragg reflection full-peak-width at half-maximum (FWHM). FWHM of bulk single crystal is typically close to 0.001 $GaAs^{[1\,1\,0]}$ r.l.u. Due to this fact it can be concluded that the observed diffuse intensity distributions can be associated with elastic strain-free mesoscopic structures having the lateral lattice constant of the substrate crystal.

In the absence of elastic strain the diffuse intensity distribution in reciprocal space is the product of the square value of the Fourier-transformed shape function of the individual dots/rings—an interference function stemming from the relative dot's/ring's arrangement on the surface. Based on the AFM-estimated average lateral dimensions of the investigated 3D nanostructures (≈ 60 nm), for both samples under consideration, the region in reciprocal space where the dot's/ring's Fourier-transformed shape function signal could have an influence extends to about ± 0.006 $GaAs^{[1\,1\,0]}$ r.l.u. from the (2 2 0) GaAs Bragg reflection. The observed intensity variations in the region outside this "shape-function-controlled" area are mainly due to inter-island correlation effects and correspond to the Fourier transform of the 2D position correlation function of the array of nanostructures.

The line-scans made for dot and ring samples in orthogonal $\langle 1\,1\,0 \rangle$ directions through the (2 2 0) GaAs Bragg point are shown in Figs. 2(b) and (d), respectively. For both samples the strong intensity maxima (marked as "K1"), which have equal positions in reciprocal space along $Q_{[1\,1\,0]}$ axis ($\Delta = \pm 0.007 \cdot 1.4$ $GaAs^{[1\,0\,0]}$ r.l.u. from the centre of (2 2 0) GaAs substrate Bragg reflection) can be found. These maxima correspond to the averaged short-range first-neighbour inter-island correlation function $f1$. A corresponding inter-island distance $d_1 = 63$ nm was determined using equation $d = 2\pi/(\Delta^*)$, where $\Delta^* = \Delta^{\times} \cdot 1.11\,\mathrm{A^{-1}}$ is the reciprocal space distance along $Q_{[1\,1\,0]}$ axis in reciprocal angstrom ($1^{GaAs[1\,0\,0]}$ r.l.u. $= 1.11$ $\mathrm{A^{-1}}$). In Figs. 2(b) and (d) one can also observe additional intensity maxima (marked as "K2") in both orthogonal $\langle 1\,1\,0 \rangle$ directions for QR sample and in the [1 −1 0] direction for QD sample. Within the measurement uncertainty the position of the maximum K2 is not periodic with that of maximum K1. In addition, the fast-Fourier-transformed (FFT) patterns from AFM images shown in Figs. 1(a) and (b) revealed centre-symmetric diffuse ring without any evidence of short-range ordering in hexagonal or cubic superlattice, which can be attributed to a highly disordered local dots/rings arrangement [30]. Taking both facts into account the maximum K2 cannot be considered as a second-order correlation maximum of the correlation function for K1. Similar data are reported from the investigation of strain-induced InAs/GaAs quantum rings [29]. There the satellite maxima have been associated to the internal shape of quantum rings. Indeed, with the dimensions of our ring structures determined from AFM micrographs, the shape function of the rings can exhibit a maximum close to the observed K2 position. However, the fact that in our case for the QD samples the same intensity maximum is observed in [1 −1 0] direction points to a different interpretation based on the special "chain-like" lateral arrangement of QD/QR nanostructures. We assume the maximum K2 to be a