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Structure changes caused by quenching of InAs/GaAs(001) quantum dots

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1. Introduction

For decades, the Stranski-Krastanov (SK) growth has been used for self-organization of quantum dot structures. This growth mode is observed in several heteroepitaxial systems with a relatively large lattice mismatch. One of the most intensively investigated SK systems is InAs on GaAs(001), which are expected to be applied for high-performance lasers and highly efficient solar cells. For these applications, the size, number density and size fluctuation of the islands are crucial parameters to be controlled. So far, their control is achieved by optimizing growth parameters on the basis of postgrowth characterization of the islands using scanning probes and electron microscopy. An underlying assumption is that samples quenched quickly enough should keep their initial structures at the growth temperature. In reality, however, it has been suggested that quenching can lead to significant structure changes [1]. While the postgrowth structures of quantum dots (QDs) have been thoroughly investigated by a variety of techniques, less has been known about the QD structures at growth temperatures. Although reflection high energy electron diffraction (RHEED) has been used to observe the evolution of facets [2,3], other structural properties, such as size and strains, have not been fully characterized under in situ conditions. Recently, we have developed an X-ray diffraction technique which allows for real-time monitoring of the molecular-beam epitaxial (MBE) growth of InAs/GaAs(001) QDs [4–6]. Further, we have shown that the structural properties characterized by in situ X-ray diffraction closely correlate with the optical properties measured by photoluminescence spectroscopy [7]. This X-ray technique provides us with a unique opportunity to investigate the QD structure changes which are possibly caused by quenching. In the present work, we compared QD structures before and after quenching by in situ X-ray diffraction to reveal the influences of quenching.

2. Experimental

Experiments were carried out at a synchrotron experimental station, BL11XU of SPring-8, using a surface X-ray diffractometer integrated with an MBE apparatus [8]. The MBE chamber is equipped with X-ray windows made of beryllium along with five evaporation sources and a RHEED system so that in situ X-ray diffraction measurements can be performed during MBE growth.

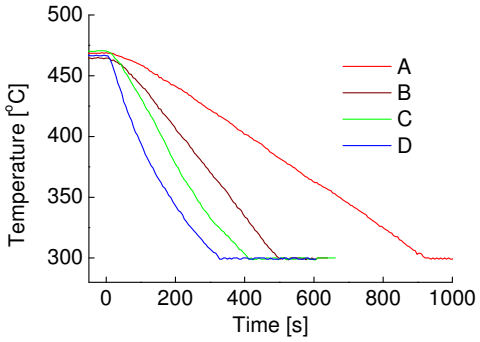

Fig. 1: Substrate temperature measured by a pyrometer as a function of time for samples (A)-(D) after turning down the power of the sample heater. The detection limit of the pyrometer lies at 300°C. The destination temperature is estimated to be 240°C.

X-rays from an undulator source were monochromatized to be 10 keV by a liquid-nitrogen-cooled Si(111) double-crystal system and focused by a pair of bent Pt-coated mirrors. The beam size was set to 0.3 mm × 0.1 mm with Ta-blade slits. The incident X-rays impinging on the sample surface at a glancing angle of 0.2° are diffracted by (220) planes that are perpendicular to the substrate surface and result in 220 diffraction in a glancing angle as well. X-ray diffraction intensity was measured with an X-ray charge coupled device (CCD) camera to construct three-dimensional X-ray intensity mappings near the 220 Bragg reflection.

Substrates were cut from a commercially supplied epi-ready GaAs(001) wafer to 7×5×0.3 mm^3 in size. After thermal evaporation of oxides and the growth of 0.2 μm-thick buffer layer, 2.6 ML InAs was deposited at a rate of 0.01 ML/s in an As pressure of 3-4×10^{-4} Pa. After the deposition of InAs was terminated, the substrate temperature was kept at the growth temperature of 470°C for 3 minutes while X-ray diffraction measurements were being carried out. Subsequently, the substrate temperature was lowered by decreasing the power of the sample

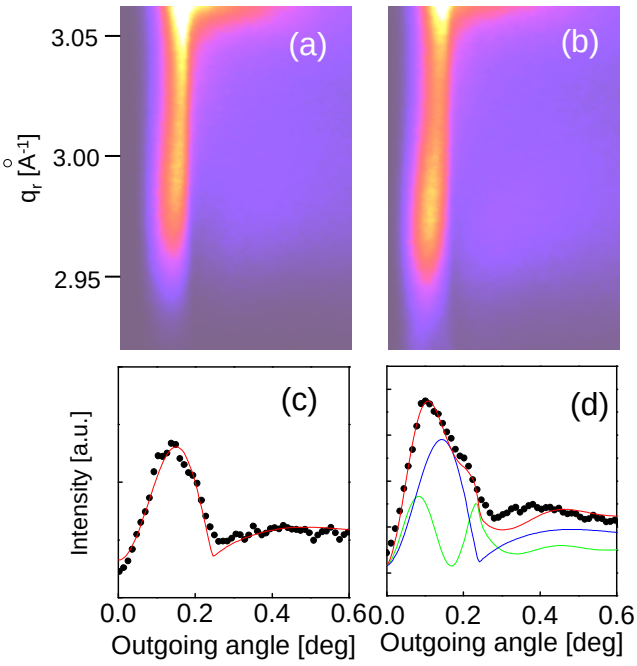

Fig. 2: (a) and (b) X-ray intensity distributions around 220 Bragg reflection from quantum dots in the radial direction (vertical) and in the surface normal direction (horizontal) before (a) and after (b) quenching. (c) and (d) X-ray intensity modulations at $q_r = 2.93$ Å^{-1} as a function of the outgoing angle. Solid lines are simulated curves.

heater at different rates. Figure 1 shows the substrate temperature measured with a pyrometer as a function of time after cooling down the substrate. Since the pyrometer used cannot detect the temperature lower than 300°C, the destination temperature was estimated to be 240°C by extrapolation. This temperature is low enough to prevent further structure changes to occur.

3. Results and discussion

Figures 2 (a) and (b) show X-ray reciprocal space mappings before and after quenching, respectively, for sample D in Fig. 1. The vertical and horizontal axes correspond to the radial direction along [110] and the outgoing angle of the diffracted X-rays, respectively. The 220 Bragg reflection of the GaAs(001) substrate occurs at $q = 3.14$ Å^{-1}, which is out of range in these figures. Thus the diffraction in Figs. 2 are coming from InGaAs islands whose lattice constant is larger than that of GaAs. The diffraction at $q = 2.93$ Å^{-1} is corresponding to fully relaxed InAs.

The intensity modulation depending on the outgoing angle can be explained by multiple diffraction effects occurring in the grazing incidence geometry. Using this modulation, the height of QDs can be determined. The intensity of X-ray diffraction which takes place at a vertical position, z, from the substrate surface is proportional to the intensity of the wave filed at z. When the incident X-rays make an angle of α with the substrate surface, the wave field at z is given by interference of the incident beam E_i and the specularly reflected beam E_r as

$$T(\alpha, z) = |E_i|^2 \left| 1 + \frac{E_r}{E_i} \right| \exp(iq_z z), \qquad (1)$$

where q_z is the surface normal component of the scattering vector. The complex amplitude, E_r/E_i, can be calculated by well-known Fresnel's formula. Figures 2 (c) and (d) are intensity modulation at $q = 2.93$ Å^{-1} of Figs. 2 (a) and (b), respectively. Before quenching, the observed modulations agree well with simulation as shown by the solid line in Fig. 2(c), and the QD height is evaluated as $z = 6.9$ nm. However, to interpret the intensity modulation observed after quenching, a two-height model including 6.9 nm and 15.2 nm high QDs is necessary as shown in Fig. 2(d). This indicates that the quenching resulted in the formation of giant dots that are often observed in AFM images.

Our past study showed that dislocated islands tended to be formed at a substrate temperature of 450°C [6]. Hence, the observed giant dots are likely to be formed when the substrate temperature passed through 450°C. We found no difference in QD structures among four samples A to D. This suggests that the giant dots are formed in such a narrow temperature range that the quenching rates investigated in the present work did not make a difference.

4. Conclusions

By in situ X-ray diffraction, it has been confirmed that quenching results in significant structure changes of InAs/GaAs(001) quantum dots. It is likely that the structural changes take place quickly when the substrate goes through a temperature range in which dislocated islands are preferably formed.

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