Extended Abstracts of the 2007 International Conference on Solid State Devices and Materials, Tsukuba, 2007, pp. 1020-1021

F-7-1 (Invited)

In-Situ X-Ray Diffraction during Semiconductor Nanostructure Growth

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1. Introduction

Molecular beam epitaxy (MBE) growth of nanostrucures is commonly monitored by reflection high energy electron diffraction (RHEED). This technique enabled atomistic-scale control of the thickness of quantum wells. However, as growth of quantum wires and quantum dots are attempted, structural properties other than thickness have become necessary to be monitored. The purpose of our study is to develop a synchrotron-based growth monitor meeting this requirement.

Recently, several X-ray techniques have been developed to investigate a variety of structural properties of quantum structures [1,2]. In the past studies, grown samples were quenched and taken out of the MBE chamber before being subjected to X-ray measurements. However, it has been pointed out that self-organized quantum dots (QDs) undergo significant changes in volume and shape during quenching [3]. In situ measurements are, therefore, essentially important for accurate characterization of semiconductor nanostructures. We have applied the x-ray diffraction technique to the in-situ and real-time monitoring of the MBE growth of InAs/GaAs(001) QDs. The whole growth process including Stranski-Krastanov (SK) growth of InAs, post-growth annealing, encapsulation with GaAs and stacking of multilayer QDs were investigated on the basis of x-ray reciprocal space mapping [6–8]. The highly collimated beam available from a synchrotron light source has enabled high resolution measurement of the strain fields. Moreover, time-resolved measurements at a rate of 9.6 s has become possible with the help of large X-ray intensity.

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 [4]. The MBE chamber is equipped with X-ray windows made of beryllium along with five evaporation sources and a reflection high-energy electron diffraction (RHEED) system so that in situ X-ray diffraction measurements can be performed during MBE growth. X-rays from an undulator source were monochromatized to be 1.24 Å by a Si(111) double-crystal system and focused by a pair of bent Pt-coated mirrors. The beam size used was 0.3 mm × 0.1 mm. As schematically shown in Fig. 1 (a), the incident X-rays, $\boldsymbol{k}_0$, impinged on the sample surface at a glancing angle of 0.2° and generates 220 diffraction, $\boldsymbol{k}_\mathrm{d}$, as well as the specular reflection, $\boldsymbol{k}_\mathrm{r}$. Because the (220) planes are perpendicular to the surface, the diffracted beam makes a glancing angle as well so that the momentum transfer is nearly parallel to the substrate surface.

The diffracted X-rays, $\boldsymbol{k}_\mathrm{d}$ were measured with an X-ray charge coupled device (CCD) camera while the sample was azimuthally rotated by 4°. As a result, X-ray intensity distribution along α and 2θ is recorded in a single CCD image as shown in Fig. 1(b). This data was

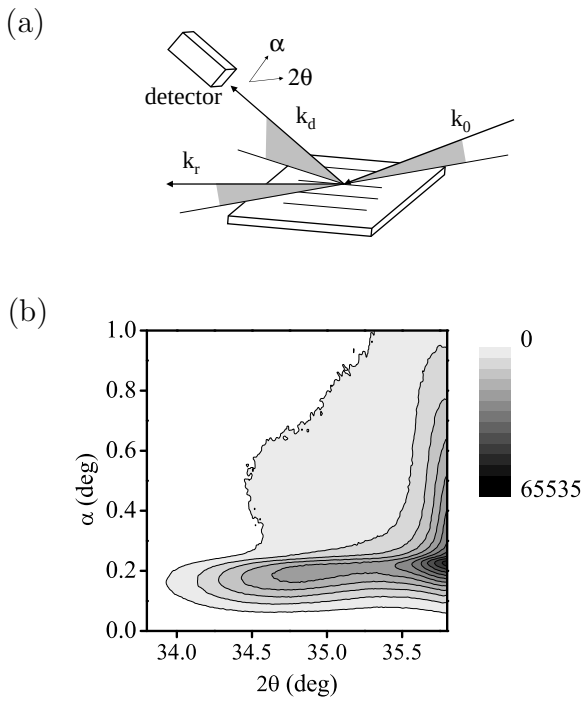

Fig. 1: (a) Experimental setup. When the sample is impinged by the incident beam, $\boldsymbol{k}_0$, 220 diffraction, $\boldsymbol{k}_\mathrm{d}$ and the specular reflection, $\boldsymbol{k}_\mathrm{r}$ are generated. Two-dimensional intensity distribution of the diffracted beam along α and 2θ is measured by an X-ray CCD detector. (b) Typical CCD image obtained from nanoislands grown by depositing 2.5 ML InAs onto GaAs(001) at a substrate temperature of 477°C.

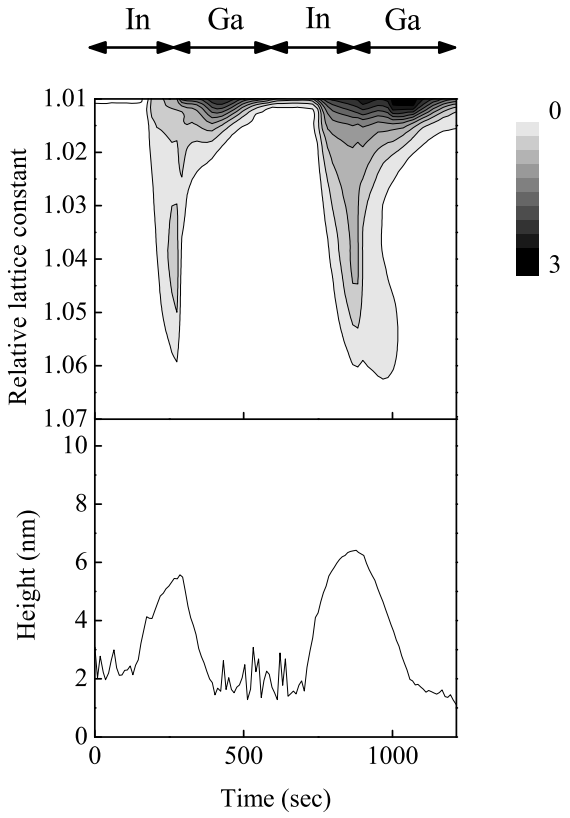

Fig. 2: Evolution of lattice constant distribution and height of nanoislands during two cycles of Stranski-Krastanov growth of InAs and encapsulation with GaAs.

obtained from the sample grown by depositing 2.5 monolayer (ML) InAs at a substrate temperature of 477°C. The intensity distribution along the 2θ direction originates from a lattice constant gradient inside nanoislands. While the lattice constant of nanoislands is close to that of the substrate at the bottom, it is relaxed to be the intrinsic value of InAs near the top. In the out-of-plane direction, on the other hand, intensity modulations are caused by interference of the three beams, $\boldsymbol{k}_0$, $\boldsymbol{k}_\mathrm{d}$ and $\boldsymbol{k}_\mathrm{r}$. From this multiple diffraction effect, the height of nanoislands can be evaluated [5].

3. Results

Figure 2 shows the evolution of the lattice constant distribution and height of SK islands during the growth of a structure made of two QD layers separated by 20 nm thick GaAs. In this figure, the lattice constant calculated from 2θ is expressed in terms of the relative value with respect to the lattice constant of GaAs. At a substrate temperature of 480°C, 3.1 ML InAs and 20 nm thick GaAs capping layers were alternately grown. The back pressure of As was 3×10^{-6} Torr and 7×10^{-6} Torr for GaAs and InAs deposition, respectively. For the first 150 s after beginning InAs deposition, no diffraction coming from relaxed islands was observed, because the amount of InAs was less than the critical thickness for island nucleation. After the critical thickness was reached, islands grew quickly to be as high as 6 nm. As soon as overgrowth of GaAs was started at 604 s, the islands were buried from the bottom, decreasing their height. By the time 20 nm thick GaAs was deposited, diffraction from the relaxed islands almost disappeared. However, the morphology of the sample surface was not fully recovered as indicated by remaining intensity at the relative lattice constant of ~ 1.01. In spite of the identical substrate temperature and As back pressures, the evolution of strain fields and island height shows significant differences between the first and second QD layers. Firstly, the second islands are bigger than the first ones. Secondly, in the course of the encapsulation of the islands in the second layer, an increase in intensity was observed at a relative lattice constant of 1.06, which indicates the onset of misfit dislocations.

Similar X-ray measurements were carried out for several samples under different growth conditions. For each sample, optical quality evaluated by photoluminescence spectra was clearly correlated with structural properties measured by in situ X-ray diffraction. This shows that synchrotron X-ray diffraction is useful to monitor and control the growth of nanostructure devices.

4. Conclusion

We investigated the evolution of strain fields and island height during the whole growth process of InAs/GaAs(001) QDs using synchrotron X-ray diffraction. This technique proved to serve as a monitor of nanostructure growth.

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