Study of InAs/GaAs(001) nanoisland growth process 
by in-situ and real-time X-ray diffraction *

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A monitoring technique for molecular beam epitaxial growth of InAs/GaAs(001) nanoislands is presented. With the help of a combination of synchrotron radiation and a two-dimensional X-ray detector, X-ray diffraction intensity mappings in the reciprocal space have been measured during growth at a rate of 9.6 s per frame. This method provides information on strain distribution and height of Stranski-Krastanov islands under the in situ condition. Because the use of X-rays is not hindered by ambient pressure, this technique is suitable for industry-oriented applications such as organometallic vapor-phase epitaxy as well. [DOI: 10.1380/ejssnt.2006.426]

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I. INTRODUCTION

Semiconductor nanostructures that allow carrier confinement in low dimensions have been attracted much attention because they show fascinating quantum effects which lead to novel device functions. The quantum well structures, in which the quantization is achieved in the growing direction, have already been applied to various devices such as high-performance semiconductor lasers. Molecular beam epitaxy (MBE) growth of quantum well structures can be monitored accurately enough by reflection high energy electron diffraction (RHEED). Thickness of each layer can be controlled in an atomic scale using a phenomenon known as the RHEED oscillation as well as the flatness of the growth front. However, as devices based on quantum wires and quantum dots are attempted, structural properties other than thickness have been necessary to be monitored. A typical semiconductor quantum dot system that has been intensively studied is InAs islands on GaAs(001).[1] Following Stranski-Krastanov (SK) growth mode, nanometer-sized islands of InAs are spontaneously formed on GaAs(001). These SK islands work as quantum dots when they are buried in GaAs capping layers. For laser applications, it is critical to control the three-dimensional shape and the internal strain of the quantum dots, which are inaccessible by RHEED.

Recently, several X-ray techniques have been developed to investigate a variety of structural properties of quantum dots. The dot shape was investigated by grazing-incidence small angle scattering [2–4] and intensity mapping in reciprocal space [5]. The strain distribution inside the dots was studied by thorough X-ray reciprocal space mapping [5–8] and by analysis of X-ray diffraction profiles [9]. The chemical composition in the dots was analyzed by utilizing X-ray anomalous scattering [10–14] and the structure factor of zinc-blende type crystals [5]. These X-ray techniques are promising tools to monitor the growth process of quantum dot structures.

This paper describes an X-ray diffraction technique developed for the in-situ and real-time monitoring of the MBE growth of InAs/GaAs(001) quantum dots. The evolution of the internal strain and height of the dots has been investigated on the basis of X-ray reciprocal space mapping during the growth of InAs dots. Since these structural properties are directly related to the optical properties of the quantum dots, the present X-ray technique is suitable for practical applications.

II. EXPERIMENTAL

The experiments were performed at the synchrotron radiation beamline 11XU at SPring-8, Japan, using a surface X-ray diffractometer directly coupled to an MBE apparatus.[15] The MBE chamber is equipped with X-ray windows made of beryllium along with Ga, In and As evaporation sources. The apparatus is designed such that not only RHEED but X-ray measurements are possible at the same position as the sample is prepared. The sample

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Fig. 1: Schematic of the experimental setup for in situ X-ray reciprocal lattice mapping. With the help of a two-dimensional detector, intensity distribution in the in-plane (2θ) and the out-of-plane (α) directions are measured simultaneously.
was cut to $7 \times 5 \times 0.3 \text{ mm}^3$ from an epi-ready wafer of GaAs(001), mounted on a molybdenum block, and loaded into the MBE chamber. After removal of the oxide layer, and the growth of a 0.2 $\mu$m-thick buffer layer, InAs was deposited at a rate of 0.007 ML/s in an As pressure of $2 \times 10^{-6}$ Torr.

Figure 1 shows a schematic of the setup for the x-ray measurements. X-rays from an in-vacuum undulator were monochromatized to $\lambda = 1.240$ Å by a Si(111) double crystal system and focused to 0.3 mm (horizontal)×0.1 mm (vertical) by a pair of bent Rh-coated mirrors and a Ta-blade slit. The surface normal of the sample was aligned so as to lie in the horizontal plane. The angle of incidence was fixed at 0.200°, which is slightly smaller than the critical angle for total external reflection of X-rays. The X-rays were reflected vertically by the (220) planes perpendicular to the surface. The diffracted X-rays were detected by an X-ray CCD camera placed at a distance of 697.7 mm from the sample. The angular acceptance of the detector was 4.3° in diameter. Using this geometry, the projection of the reciprocal lattice mapping on the (110) plane was recorded in the following way. Each point in the CCD image corresponds to a different scattering angle, $\theta_i$, and outgoing angle, $\alpha$. While the CCD camera was being exposed, the sample was rotated about the [001] axis by 4° at a speed of 1.008°/s. As a result, two dimensional mapping of the integrated intensity in the [110] direction was recorded. The acquisition of one frame took 9.6 s which includes a readout time of $\approx 5$ s. This gives the temporal resolution of the present method.

III. RESULTS

Figures 2 (a) and (b) show X-ray CCD images which were observed when 2.5 ML and 4.9 ML InAs was deposited at a substrate temperature of 480°C, respectively. It takes 9.6 sec to measure a single X-ray CCD image. The horizontal axis represents the outgoing angle measured from the substrate surface. In these pictures, the outgoing angle $\alpha$ is normalized by the critical angle for the total reflection of X-rays, $\alpha_c$. The vertical axis corresponds to the 2θ angle measured in the in-plane direction. From the 2θ value, the in-plane lattice constant of the dots can be calculated. In Figs. 2 (a) and (b), the in-plane lattice constant is expressed in terms of the relative value with respect to the GaAs substrate, $\epsilon = (a - a_{GaAs})/a_{GaAs}$, where $a$ is the in-plane lattice constant of the SK islands. As the growth of InAs proceeds, the overall intensity increases and the intensity distribution extends to larger lattice constants. The whole growth process at 480°C observed by the present method is presented as a movie in Appendix.

The observed intensity distribution, $I(\epsilon, \alpha)$, is interpreted in terms of the product of the lattice constant distribution, $D(\epsilon)$, and the optical effect, $T(\alpha_i, \alpha_f)$, where $\alpha_i$ and $\alpha_f$ are the incoming and outgoing angles measured from the surface, respectively. Because of the reciprocity theorem of optics, the optical effect can be written in the form of $T(\alpha_i, \alpha_f) = t(\alpha_i)t(\alpha_f)$ using an identical function, $t(\alpha)$, for the incoming and outgoing angles. For a flat surface, this function is described by Fresnel’s transmission coefficient [16] whose maximum value is attained for $\alpha = \alpha_c$. However, for the case of SK islands where diffraction occurs at a finite height above the substrate surface, multiple diffraction between the SK islands and the substrate needs to be considered. In this case, the function $t(\alpha)$ is given by a generalized optical function [5] that depends on both the glancing angle, $\alpha$, and the height, $z$. Figure 3 shows the X-ray intensity at fixed in-plane lattice constants as a function of the outgoing angle. For a lattice constant close to the GaAs bulk value, the intensity maximum occurs at an outgoing angle close to $\alpha_c$. For a larger in-plane lattice constant, the peak position shifts to a smaller angle. This peak shift reflects such a strain distribution that the lower part of SK islands is strained to match the substrate while the upper part is more relaxed. Using this optical effect, the height of the SK islands can be evaluated. According to the theory of the generalized optical factor, the height where the diffraction occurs can be calculated from the angle of maximum intensity $\alpha_{max}$ with the following equation [5]:

$$z = \frac{\lambda}{2\pi \alpha_{max}} \cos^{-1} \frac{\alpha_{max}}{\alpha_{c}}$$

(1)

In this study, the maximum of the evaluated height is
FIG. 3: The intensity modulation as a function of the outgoing angle at various in-plane lattice constants.

regarded as the height of SK islands.

The intensity modulation caused by the optical effect complicates the interpretation of the intensity distribution along the $2\theta$ direction. The generalized optical factor is a function of both the incident angle and the height where the diffraction occurs. Thus, even for a fixed incident angle of 0.2°, the generalized optical factor, $t(\alpha_i)$, varies according to the height as shown by a calculation in Fig. 4. Because of this effect, the diffracted intensity is not exactly proportional to the area of the corresponding lattice constant.

Figure 5 shows the temporal evolution of the strain distribution and height of SK islands for the growth at 430 °C. To show the strain distribution, the intensity integrated from $\alpha = 0$ to $3\alpha_c$ is shown as a function of growth time. For the first 240 s, diffraction from relaxed lattices is absent and the height of islands remains zero because the growth proceeds in two dimensional fashion. When the critical thickness is reached, diffraction from relaxed islands starts to grow. The height of islands increases rapidly at the initial stage of island formation and shows a tendency to approach a constant value until $t = 400$ s, which corresponds to the InAs thickness of 2.8 ML. Further growth of InAs cause a kink in the temporal evolution of height. Concomitantly, the strain distribution shifts to a larger lattice constant and finally it shows a bimodal distribution which has another peak at $\epsilon = 0.06$. The change observed at $t = 400$ s indicates the occurrence of dislocation.

At 480°C, the growth of InAs behaves differently as shown in Fig. 6. After the island formation at $t = 240$, the island height grows immediately up to 6 nm and stays almost constant for 100 s. The presence of the plateau in height suggests a mechanism by which the island size is automatically limited[17–19]. At $t = 500$ s, the height of InAs islands shows a kink indicative of the occurrence of dislocations in the same way as the growth at a lower temperature. However, the InAs thickness needed for the occurrence of dislocations is significantly larger than that for the 430°C. The difference of 0.7 ML cannot be due to evaporation of In from the substrate because the difference in the critical thickness for the transformation from 2D to 3D growth shows much smaller temperature dependence. Thus we describe this delay in the occurrence of dislocations to thermally enhanced alloying.

IV. DISCUSSION

The synchrotron X-ray diffraction technique described in this paper has the following two unique properties as a monitor for nanostructure growth. First, it elucidates strain distribution inside nanoislands. To understand and control the growth of InAs on GaAs(001), therefore, in situ observation of internal strain of the nanoislands is essential. So far, strain evolution during layer-by-layer growth of InGaAs/GaAs has been measured by detecting the bending of the substrate that is shaped as a cantilever [20]. In this method, the average strain of the InGaAs layer was estimated from the accumulated stress that was given to the substrate. Contrastingly, X-ray diffraction measure-
ments give the lattice constant of the grown material and thus complementary information is obtained. Moreover, the X-ray method reveals the lattice constant distribution semiquantitatively instead of the average strain over the entire structure. For fully quantitative evaluation of the lattice constant distribution, a correction related to the multiple diffraction effect still needs to be developed. Second, the present X-ray method enables in situ and real-time monitoring of the height of the nanoislands during growth. A number of studies on the shape of SK islands have been performed by atomic force microscope (AFM) and scanning tunneling microscopy (STM) with quenched samples. However, they have rarely been applied to in situ measurements during growth because of difficulty with the implementation of a scanning probe in an MBE chamber. Very recently in situ STM observations of InAs/GaAs have been attempted at an extremely slow growth rate [21]. Our method has achieved a temporal resolution of 9.6 s/sec, which is compatible with practical growth rate for InAs QDs on GaAs(001). A limitation of the present X-ray method is the possibility that it overlooks the size distribution of SK islands. As observed for Ge/Si(001), self-assembled nanoislands may have multiple stable shapes with different aspect ratio [22]. The resultant multimodal size distribution is hard to analyze only from the X-ray diffraction results. Hence ex situ AFM or STM measurements are still necessary to make sure of a uniform size distribution of QDs.

The structural information provided by the present technique is directly related to the optical properties when the QD structure is applied to lasers. The height of nanoislands is an important factor which determines the wavelength and polarization of lights emitted from QD lasers. Thus, monitoring the height of QDs by this method makes it possible to control the wavelength of QD lasers. Further, its sensitivity to dislocations is useful for improvement of productivity. By observing the onset of dislocations, one can grow InAs until dislocations are about to be introduced. As a result, high density QDs can be grown more easily without introducing dislocations that spoil the efficiency of light emission. For this reason, this technique serves as a powerful growth monitor suited for the production of QD lasers. In addition, a penetrating feature of X-rays enables wide application of this method. The experiments presented in this paper were carried out in MBE conditions. However, since the combination of synchrotron X-ray diffraction and organometallic vapor-phase epitaxy (OMVPE) has been established as reported in literature[23], it is not difficult to implement the present technique to OMVPE apparatus.

V. CONCLUSION

In conclusion, we have demonstrated that X-ray reciprocal mapping with synchrotron radiation serves as a unique monitoring technique to study the strain and height evolution during the growth of InAs/GaAs(001) quantum dots by MBE. A temporal resolution of 9.6 s has been achieved by employing an X-ray CCD detector. This technique is suitable for industrial applications.
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