X-ray diffraction study on self-organization of InAs islands on GaAs(001)

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Abstract. The molecular beam epitaxial (MBE) growth and postgrowth annealing of InAs/GaAs(001) quantum dots were investigated by grazing-incidence X-ray diffraction using a diffractometer integrated with an MBE apparatus. The use of synchrotron radiation and a two-dimensional X-ray detector enabled X-ray diffraction measurements on the internal strains and height of dots at a rate of less than 10 s per frame. Mass transport by surface diffusion was found to play a major role in the nucleation of three-dimensional islands of InAs/GaAs(001).

1. Introduction
The understanding of heteroepitaxy is essentially important for the fabrication of semiconductor devices. When thin layers are grown on dissimilar substrates, the morphology of the grown film is known to be one of three distinctive types, which are referred to as the Frank-van der Marwe (FM) type with successive complete two-dimensional (2D) layers, the Volmer-Weber (VW) type with three-dimensional (3D) islands that partially cover the substrate and the Stranski-Krastanov (SK) type with 3D islands on a thin 2D wetting layer. SK growth is typically observed in systems that have similar chemical properties and a relatively large lattice mismatch between grown materials and substrates. A number of technologically important materials, such as Ge on Si(001) and InAs on GaAs(001), follow this growth mode. Many experiments have revealed that the SK islands of Ge/Si(001) and InAs/GaAs(001) are free of dislocations and sufficiently small for quantum effects to be significant. Furthermore, under appropriate growth conditions, a size fluctuation of less than 10% has been achieved. These are desirable characteristics for nanodevice applications.

The transition from the 2D growth of the wetting layer to the 3D growth of islands in the SK mode has posed a problem. The nucleation of 3D islands is much faster than the expectation from the deposition rate of relevant materials. Spencer et al. [1] and Tu and Tersoff [2] have proposed a mechanism in which the morphological instability induced by an increase in the surface composition of deposited materials leads to SK island formation. The mass transport that is necessary in this process is driven by surface diffusion. Experimental evidence for the surface mass transport responsible for the nucleation of islands has been obtained by reflection high-energy electron diffraction (RHEED) [3] and scanning tunneling microscopy (STM) [4] studies.

In this paper, we report on in situ X-ray diffraction measurements that confirm the contribution of surface diffusion to the nucleation of InAs islands on GaAs(001). Recently, we have developed a technique by which the internal strain distribution and height of SK islands can be evaluated during
growth [5-8]. The quantitative analysis enabled by this technique provides detailed insight into the atomistic process of the nucleation of SK islands.

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 [9]. The MBE chamber is equipped with X-ray windows made of beryllium along with five evaporation sources and a RHEED system for in situ X-ray diffraction measurements using MBE growth. X-rays from an in-vacuum undulator source were monochromatized to be 0.124 nm using a Si(111) double-crystal system and focused using a pair of bent Pt-coated mirrors. The beam size used was 0.3 x 0.1 mm².

The sample was cut into 7 x 5 x 0.3 mm³ from an epi-ready wafer of GaAs(001), mounted on a molybdenum block with In, and loaded into the MBE chamber. After the removal of the oxide layer and the growth of a 0.2-µm-thick buffer layer, InAs was deposited at a rate of 0.0088 ML/s and an As pressure of 3x10⁻⁶ Torr. The substrate temperature was kept at 465°C during growth and annealing.

As schematically shown in Fig. 1(a), the incident X-rays, \( k_0 \), impinge on the sample surface at a glancing angle of 0.2° and result in the 220 diffraction, \( k_d \), as well as the specular reflection, \( k_r \). Because (220) planes are perpendicular to the substrate surface, the diffracted beam also makes a glancing angle, \( \alpha \), so that momentum transfer is nearly parallel to the substrate surface.

The diffracted X-rays, \( k_d \), 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 distributions along \( \alpha \) and \( 2\theta \) were recorded in a single CCD image, as shown in Fig. 1(b). This image was obtained from the sample grown by depositing 2.7 ML InAs on GaAs(001). From the intensity distribution along \( 2\theta \), one can evaluate the lattice constant distribution inside SK islands, following the procedure described in Ref. [8]. The continuous intensity distribution along \( 2\theta \) is due to a lattice constant gradient from the bottom to the top of SK islands, which are compressively strained at the bottom and almost relaxed near the top. In the out-of-plane direction, on the other hand, the intensity of diffracted X-rays is strongly modulated so as to attain a maximum near the critical angle for total reflection of X-rays, \( \alpha_c \). For a flat surface, this modulation coincides with Fresnel’s transmission coefficient which has a peak exactly at \( \alpha = \alpha_c \). However, in the case of SK islands, where the diffraction occurs at a finite height from the substrate surface, it has been shown that the angular position for maximum intensity is shifted to \( \alpha < \alpha_c \) [10]. From this peak shift, the height of SK islands can be estimated [8, 10].

![Fig. 1: (a) Schematic of X-ray diffraction measurement setup. Incident X-rays, \( k_0 \), result in the 220 diffraction, \( k_d \), as well as the specular reflection, \( k_r \). The intensity distributions along \( 2\theta \) and \( \alpha \) are measured simultaneously with an X-ray CCD camera. (b) Typical X-ray CCD image. The continuous intensity distribution along \( 2\theta \) comes from a lattice constant gradient inside SK islands. The intensity modulation along \( \alpha \) results from multiple diffraction effects. Using this modulation, one can determine the height at which the diffraction by SK islands occurs.](image-url)
Fig. 2: Evolution of lattice constant distribution and height of Stranski-Krastanov islands during deposition of InAs on GaAs(001) and postgrowth annealing. Upon closing the In shutter at $t = 307$ s (dashed line), the amount of deposited In reached 2.7 ML.

Fig. 3: Evolution of lattice constant distribution and height of Stranski-Krastanov islands during deposition of 1.9 ML InAs on GaAs(001) and postgrowth annealing. Islands still keep growing even after closing the In shutter at $t = 218$ s (dashed line).
3. Results
Figure 2 shows the evolution of the lattice constant distribution and height during the growth of InAs on GaAs(001) and subsequent annealing in an As flux. In this figure, the lattice constant is expressed in terms of its relative value with respect to the lattice constant of GaAs bulk. The deposition of InAs was stopped at $t = 307$ s, which is indicated by a dashed line in the figure. The amount of InAs deposited at this point of time was estimated to be 2.7 ML by assuming a critical thickness of 1.75 ML for the 2D to 3D transition. The first 200 s corresponds to the 2D growth of a wetting layer. During this period of time, no diffraction was observed except the GaAs 220 bulk diffraction because the grown film completely matched the substrate in the in-plane direction. When the critical thickness was reached at $t = 200$ s, a diffraction corresponding to relaxed lattice constants began to emerge and the height of islands started to increase, showing the formation of 3D islands. When the supply of InAs was stopped at $t = 307$ s, the increase in height stopped immediately. Changes in strain and height are subtle during annealing.

A notable behavior of SK islands was observed when a small amount of InAs was deposited. In the experiment shown in Fig. 3, In was supplied for 218 s, which corresponds to the thickness of 1.9 ML. The diffraction from islands and the height increased continuously even after closing of the In shutter. The height of islands reached its maximum at $t = 500$ s and then gradually decreased during annealing. Because the nucleation of islands occurs in the absence of the incident In flux, the materials required for the nucleation process were transported by surface diffusion. Overall intensity was significantly lower than that for the sample with 2.7 ML InAs because the number density of islands was smaller.

4. Discussion and conclusion
By our synchrotron X-ray diffraction technique, the quantitative determination of the strains and height of SK islands during island nucleation and annealing had become possible, in addition to the simple detection of the onset of the island formation. The comparison of the two samples varying in the amount of In indicates the importance of surface mass transport for island nucleation. A major difference between the two samples is the presence or absence of an external In flux during island nucleation. Although the resultant size and number density of SK islands in the 1.9 ML sample are smaller than those of the other sample, the initial increase in the height of SK islands is similarly steep irrespective of the absence of an external In flux. This shows that the time scale of islanding is mainly determined by surface mass transport rather than the deposition rate of the materials. At present, there are two possible factors responsible for surface mass transport under MBE conditions. One factor is the presence of small islands consisting of hundreds of III- and V-group atoms. Before the formation of 3D islands, the appearance of small quasi-3D clusters, which are 2-4 ML high and 20 nm wide, have been observed by STM [11]. Another STM study has revealed that the formation of 2-4-ML-high 3D clusters coexisting with larger 2D islands precedes the 2D-3D transition [12], although it cannot be ruled out that these small clusters were formed during quenching. The other factor is the presence of single atoms or clusters of a few III-group atoms. On the basis of the density and volume distributions of SK islands, STM and AFM studies have shown that the surface mass transport is attributed to the floating In on the InGaAs wetting layer instead of quasi-3D precursors [13].

Another difference between the two samples with 1.9 ML and 2.7 ML In is the structural change during postgrowth annealing. While the height of the islands remains nearly constant for the 2.7 ML sample, that of the islands of the 1.9 ML sample gradually decays as indicated by decreases in diffraction intensity and height. We speculate that the initial size and chemical composition affect the stability of SK islands during annealing. A recent X-ray diffraction study has shown that, in InAs growth on GaAs(001), SK islands become more Ga-rich as growth progresses because Ga atoms in the underlying layers are incorporated into the islands [14], suggesting the compositional difference between the two samples investigated in the present study.

In conclusion, our in situ X-ray diffraction study suggests the contribution of surface diffusion to the nucleation of SK islands. At the initial stage of nucleation, the aggregation of floating In or small
In clusters on the surface results in In-rich islands. As more In is deposited, Ga atoms in wetting layers are incorporated into the islands by surface diffusion, forming alloyed larger islands.

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