Strain Relaxation in GaSb/GaAs(111)A Heteroepitaxy Using Thin InAs Interlayers

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ABSTRACT: We have systematically studied the strain relaxation processes in GaSb heteroepitaxy on GaAs(111)A using thin InAs interlayers. The growth with 1 ML- and 2 ML-InAs leads to formation of an InAsSb-like layer, which induces tensile strain in GaSb films, whereas the GaSb films grown with thicker InAs layers (≥3 ML) are under compressive strain. As the InAs thickness is increased above 5 ML, the insertion of the InAs layer becomes less effective in the strain relaxation, leaving residual strain in GaSb films. This leads to the elastic deformation of the GaSb lattice, giving rise to the increase in the peak width of X-ray rocking curves.

1. INTRODUCTION

GaSb-based semiconductor material systems are of considerable interest for the application to electronic and optoelectronic devices, such as emitters and detectors operating at near-infrared and mid-wavelength regions.1–4 Heteroepitaxy on lattice-mismatched substrates, such as GaAs and Si, is favored for the fabrication of GaSb-based devices, because low-cost wafers with larger diameter are available. However, the extremely large lattice-mismatch (e.g., 7.8% for GaSb on GaAs) prevents the growth of atomically smooth layer-by-layer growth of GaSb films and gives rise to the high density of defects.

Heteroepitaxy with a large lattice mismatch usually follows a Straniski–Krastanov (SK) growth mode: a strained two-dimensional layer is formed under a certain critical thickness, beyond which three-dimensional islands are formed to relieve the strain. Exceptionally, in marked contrast to the conventional SK growth for the (001) orientation,5 InAs grows in a layer-by-layer mode on the GaAs(111)A6,7 and (110)8 substrates: periodic arrays of misfit dislocations are formed at the InAs/GaAs interface to accommodate the large lattice mismatch of 7.1%. It has been also reported that (111)A-oriented InAs films are two-dimensionally grown on the Interminated Si(111) surface,9 despite a larger lattice mismatch of 11.5%. On the other hand, GaSb, having a lattice constant (0.6097 nm) close to that of InAs (0.6055 nm), forms islands on GaAs(111)A10 and Si(111)11 substrates. In addition, the layer-by-layer growth does not occur in the In0.25Ga0.75As/GaAs(111)A system with a smaller lattice mismatch of ≈1.8%.9

We have recently reported that the insertion of a thin interface layer of InAs between GaSb and Si successfully allows smooth layer-by-layer growth of GaSb films.15 The growth with InAs layers is also effective to improve the crystalline quality and surface morphology of the In0.25Ga0.75As film on the GaAs(111)A substrate.12 This paper presents a systematic study on the GaSb heteropitaxy on GaAs(111)A using the InAs interlayer. We found that the strain evolution and growth mode drastically change with the InAs layer thickness. The GaSb films grown with thick (≥3 ML) InAs layers are under compressive strain, as if the growth proceeds on the bulk InAs substrate. On the other hand, Sb atoms are preferentially incorporated in the 1ML and 2 ML-InAs layers forming the InAsSb-like phase, so that tensile strain is accumulated in GaSb films. The specific role of the InAs interlayer in the strain relaxation in GaSb/GaAs(111)A heteroepitaxy, which strongly affects the resultant structural properties of GaSb, will be discussed.

2. EXPERIMENTS

The growth experiments were performed using a multichamber molecular-beam epitaxy system.13 The clean surface of GaAs(111)A was obtained using the procedures described in our previous paper.14 The clean and well-ordered Ga-rich (2×2) reconstruction was confirmed by scanning tunneling microscopy, X-ray photoelectron spectroscopy, and reflection high-energy electron diffraction (RHEED). The InAs films with thickness in the range of 1 ML to 50 ML were grown at 430 °C with an As/In ratio of ~50. After a good ultra-high vacuum condition of 5×10−10 Torr was recovered, the growth...
of GaSb was initiated by simultaneously opening the Ga and Sb shutters at 430 °C with an Sb/Ga flux ratio of ~4. The substrate temperatures were measured using an infrared pyrometer and were calibrated by melting the InSb sample at 525 °C. The growth rates of InAs and GaSb were approximately 0.0167 and 0.085 ML/s (0.3 nm/s), respectively. The growing GaSb surface shows a $\{2\sqrt{3} \times 2\sqrt{3}\}$ RHEED pattern, which is characteristic of the Sb-stabilized reconstruction on the GaSb(111)A surface. The growth processes of GaSb were monitored by RHEED in real time. Structural properties of the samples were characterized by X-ray diffraction (XRD), atomic-force microscopy (AFM), and transmission electron microscopy (TEM). All the AFM images were obtained in the tapping mode. High-resolution XRD measurements were carried out using a monochromatic Cu Kα1 radiation. A channel-cut analyzer crystal was used for X-ray rocking curve (XRC) measurements, because the full width at half-maximum (fwhm) values obtained from the XRC measurements without a slit are overestimated for thin (<100 nm) GaSb films. We note that no significant difference was observed for the GaSb thickness above 100 nm between the XRC results obtained using the crystal analyzer and the open detector.

3. RESULTS AND DISCUSSION

Figure 1a,b shows the lattice constants of 300 nm-thick GaSb films in the directions parallel ($d_{110}$) and normal ($d_{111}$) to the surface, respectively, plotted as a function of the InAs thickness. The values are estimated from the RSMs (Figure S1). Shown by crosses in (b) are the values calculated from $d_{110}$ on the basis of classical elastic theory.

The insertion of 1 ML and 2 ML thick InAs increases (decreases) the $d_{110}$ ($d_{111}$) value: the GaSb film grown with 2 ML-InAs shows a maximum expansion of $d_{110}$ (0.4315 nm). The $d_{110}$ ($d_{111}$) value, then, decreases (increases) with further increasing InAs thickness, and is almost unchanged above 5 ML. These results are closely related with the initial growth processes of GaSb, as we will show below.

Figure 2 shows the variation of $d_{110}$ during the GaSb growth. The $d_{110}$ values were measured from the distance between the 11 and 1̅1̅ reflections in the RHEED patterns. Arrowheads indicate local maxima and minima.

Figure 2. Variation of the in-plane lattice constant ($d_{110}$) of GaSb films grown on InAs (0–10 ML)/GaAs(111)A substrates. The values were measured from the distance between the 11 and 1̅1̅ reflections in the RHEED patterns. Arrowheads indicate local maxima and minima.

When the GaSb was directly grown on the GaAs substrate, the $d_{110}$ value monotonically increases with GaSb thickness and is smaller than the bulk value throughout the growth (Figure 2a). On the other hand, for the GaSb growth with 1 ML-
ML) InAs, the $d_{110}$ value initially increases and has a maximum at 0.6 nm (0.3 nm): the maximum $d_{110}$ values are larger than that of bulk GaSb, and $d_{110}$ values are gradually decreased with further increasing GaSb thickness, as shown in Figure 2b.c. Thus, it appears likely that Sb atoms are preferentially incorporated in InAs layers at the initial stage of the GaSb growth to form an InAsSb-like phase. To confirm this hypothesis, only the Sb$_2$ molecular beam was supplied on the GaAs/(111)A and InAs/GaAs/(111)A surfaces. When the GaAs/(111)A surface was exposed to the Sb beam, no change was observed in the streak position, indicating that the incorporation of Sb in GaAs is negligible. On the other hand, for 2 ML-InAs, a new set of faint streaks with a smaller $d$-spacing appeared in addition to the streaks from the InAs layer. The $d_{110}$ value is estimated to be 0.452 nm, which corresponds to InAs$_{x}$Sb$_{1-x}$ with $x = 0.2$–0.3.

The data for the growth with 3 ML- and 10 ML-InAs (Figure 2d,e) also show the local maxima at 0.5 nm, suggesting the formation of InAsSb-like phases. As the $d_{110}$ values at their local maxima are smaller than that of bulk GaSb, it is likely that the incorporation of Sb in InAs layers is very limited. However, there still remains the question as to why the Sb incorporation is most enhanced on the 2 ML thick InAs film. The $d_{110}$ value, then, decreases and has a local minimum at 0.75 nm (1.2 nm) for the growth on 3 ML-InAs (10 ML-InAs), followed by a monotonic increase. The strain relaxation in GaSb proceeds more rapidly on 3 ML-InAs than on 10 ML-InAs: there are 94 and 76% relaxations in 30 nm-films for 3 ML- and 10 ML-InAs, respectively. We confirmed that the $d_{110}$ data for 5–50 ML-InAs thickness behave similarly as a function of GaSb thickness.

The insertion of InAs layers has a critical effect also on the surface morphology of the growing GaSb film. The GaSb film directly grown on the GaAs substrate is roughened at the very early stage of the growth (0.3–6 nm). On the other hand, for the GaSb films grown with InAs interlayers thicker than 3 ML, planar growth is observed in the whole range of the GaSb film thickness. However, the use of 1 ML and 2 ML-InAs layers has the opposite effect to promote the planar growth: the growth front roughening persists until the GaSb film thickness exceeds 15 nm. As tensile strained growth on (111)A-oriented substrates leads to island formation, the tensile strain in GaSb films grown with 1 ML- and 2 ML-InAs could be responsible for the observed surface roughening.

Figure 3a shows the fwhm values of XRCs of the asymmetric 111 reflection measured from 300 nm thick GaSb films plotted as a function of the InAs thickness. When the InAs thickness is in the range of 1–5 ML, the fwhm value is less than 50 arcsec, whereas the value drastically increases above 5 ML. Shown in Figure 3b are the root-mean-square (rms) roughness of the 300 nm-GaSb films estimated from AFM images (Figure S2). The GaSb films grown with 3–10 ML-InAs layers show smooth surface morphologies with an rms roughness of ~1 nm. On the other hand, for the InAs layer thickness below and above these values, the GaSb surface shows a higher rms roughness. Similar results have been reported for InGaAs/GaAs heteroepitaxy using thin amorphous InGaAs interlayers: the improved crystalline quality of the In$_x$Ga$_{1-x}$As film is achieved using the amorphous In$_x$Ga$_{1-x}$As interlayers with optimized thickness of 2 nm.

It has been generally believed that the broadening of XRC profiles in heteroepitaxial layers is caused by the formation of high densities of threading defects. Thus, one may suppose that high-quality GaSb films with a flat surface are grown on GaAs/(111)A with 3–5 ML thick InAs interlayers. However, our plan-view and cross-sectional TEM observations show that the density of the threading dislocation in the GaSb film grown with 10 ML-InAs is $2 \times 10^9$ cm$^{-2}$, which is significantly lower than the values in GaSb films grown with 3 ML-InAs ($4 \times 10^9$ cm$^{-2}$) and without InAs ($8 \times 10^9$ cm$^{-2}$). These results indicate that narrower XRC widths are not necessarily an indication of lower density of threading defects.
300 nm-GaSb films (Figure 4c,d), whereas the peak profile (c) consists of the two components, the result for 10 ML-InAs (d) is well fitted by a single broad component: the diffuse component associated with misfit dislocations, which is clearly seen in (b), is likely to be hidden behind the main peak with large fwhm in Figure 4d. The fwhm value of peak (d) is 694.3 arcsec, and is more than an order of magnitude larger than that for the sharp main peak in (c) (33.4 arcsec).

The changes in the width and shape of XRCs are closely related with the strain relaxation processes in GaSb, as we will show below. Shown in Figure 5a,b are the evolutions of $d_{110}$ and $d_{111}$ values, respectively. Also plotted in Figure 5b are the $d_{111}$ values calculated from the measured $d_{110}$ values (crosses in Figure 5b) on the basis of classical elastic theory.15 Whereas the $d_{110}$ value of the initial 3 ML-InAs film (0.414 nm) is smaller than that of 10 ML-InAs (0.422 nm), the GaSb lattice on 3 ML-InAs is more expanded laterally than those on 10 ML-InAs. This means that the strain relaxation in GaSb proceeds more rapidly on 3 ML-InAs than on 10 ML-InAs, as mentioned earlier: there are ~96 and ~80% relaxations for 100 nm-GaSb films for 3 ML- and 10 ML-InAs, respectively. As expected from the results in Figure 5a, the $d_{110}$ values of GaSb on 10 ML-InAs are larger than those for 3 ML-InAs (Figure 5b). In addition, the measured $d_{111}$ values are larger than that of bulk GaSb (0.352 nm) and are gradually decreased with increasing film thickness. Here we note that, in Figure 5b, the $d_{111}$ values measured from the 30 nm films (circles) are slightly smaller than the calculated values (crosses). A similar result has been reported for InAs/GaAs(111)A heteroepitaxy: the InAs lattice is under compressive strain in both surface-normal and surface-parallel directions at the initial stage of the growth.7

Figure 5c shows the evolution of the fwhm value of the 111 XRC. For the growth on 10 ML-InAs, the fwhm value (open circles) is less than 50 arcsec below 100 nm, but it is drastically increased above 200 nm. On the other hand, as shown in Figure 5a, the $d_{110}$ value shows nothing particularly remarkable between 100 and 200 nm. As the strain in 100 nm-GaSb on 10 ML-InAs has relaxed by only ~80%, it is plausible that the residual strain accumulated in the GaSb film above 100 nm is hardly relaxed by misfit dislocations alone and that the GaSb lattice begins to be elastically deformed to accommodate the strain: elastic deformation results in the distortion of the crystal unit cell, leading to shift in the position of the diffraction spots, giving rise to the broadening of the XRC width. In marked contrast with 10 ML-InAs, as shown by open circles in Figure 5a, the fwhm value of GaSb grown on 3 ML-InAs remains almost unchanged as the film thickness is increased, and is smaller than 50 arcsec in the whole range of the GaSb thickness. This means that the elastic deformation of the GaSb lattice on 3 ML-InAs hardly occurs, being consistent with the result that the strain in 100 nm-GaSb has plastically relaxed by a large amount of 96% (Figure 5b).

Figure 6 shows the two-dimensional RSMs for the asymmetric 115 reflection of GaSb measured using a one-dimensional detector. The vertical and horizontal axes represent the indices along the [111] and [112] directions, respectively. In good agreement with the XRC results, the 115 reflections from GaSb above 200 nm on 10 ML-InAs are broadened along the direction perpendicular to the [115] azimuth. This means that the broadening of the 111 XRC (Figure 4d) arises from a slight misorientation of the GaSb lattice because of the strain-induced elastic deformation. On the other hand, such a peak broadening is hardly observed for the growth on 3 ML-InAs, further confirming that the elastic deformation is suppressed.

To understand the difference in the strain relaxation processes on 3 ML- and 10 ML-InAs, we studied the structure of the 300 nm GaSb/InAs/GaAs(111)A interface using cross-sectional TEM. Figure 7 shows the variations of the in-plane lattice constant across the interfaces. The data were obtained by carrying out one-dimensional fast Fourier transformation for high-resolution cross-sectional TEM images in the direction parallel to the interface, and by measuring the positions of derived spots, which correspond to the 111 diffraction spots. The $d_{110}$ value for 3 ML-InAs (Figure 7a) appears to change from 0.40 nm (GaAs) to 0.43 nm (GaSb) across the InAs layer. We have reported that, in InGaAs/InAs/GaAs(111)A heteroepitaxy, the thin InAs interlayer is elastically deformed to accommodate the strain in the InGaAs film.12 Thus, similar to the case in the InGaAs/InAs/GaAs(111)A system, it appears likely that the 3 ML-InAs lattice is laterally expanded to be coherent with the GaSb
lattice, promoting the strain relaxation in GaSb. Here, we note that, as mentioned earlier, Sb atoms are preferentially incorporated into the InAs layer to form an InAsSb-like phase. Thus, it could not be ruled out that the possible formation of the GaInAsSb phase and its compositional grading also result in the variation of \( d_{110} \) near the interface.

As shown in Figure 7b, the \( d_{110} \) values for 10 ML-InAs are almost the same as that before the GaSb growth (0.423 nm), indicating that the misfit dislocations are also formed at the GaSb/InAs interface. This means that the 10 ML-InAs layer is too thick to be compliant and is hardly deformed by growing the GaSb film. As both in-plane and surface-normal lattice constants of 300 nm-GaSb hardly depend on the InAs thickness above 5 ML, as shown in Figure 1a, b, it is likely that the thick InAs layer (<5 ML) is less efficient in the strain relief of GaSb.

4. CONCLUSIONS

The effects of the InAs interlayer on the growth mode and strain evolution in GaSb heteroepitaxy on GaAs(111)A have been studied. The GaSb growth with the InAs interlayer proceeds under either compressive or tensile strain depending on the InAs thickness. The structural properties of GaSb are closely related with the growth mode and strain relaxation processes: the GaSb film grown with a thin (3 ML) InAs layer has an XRC width less than 50 arcssec and a high threading dislocation density of \( 4 \times 10^{10} \) cm\(^{-2} \). On the other hand, for thick (10 ML) InAs, the XRC width is increased by more than an order of magnitude, but the defect density is decreased.

**ASSOCIATED CONTENT**

* Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.8b02359.

RSMs of the asymmetric 115 reflection measured from 300 nm thick GaSb films and AFM images of 300 nm thick GaSb films grown on GaAs(111)A using InAs interlayers (PDF)

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**Notes**

The authors declare no competing financial interest.

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