Strain Relaxation in InAs/GaAs(111)*A* Heteroepitaxy

Akihiro Ohtake and Masashi Ozeki
Joint Research Center for Atom Technology (JRCAT), Tsukuba 305-0046, Japan
and Angstrom Technology Partnership (ATP), Tsukuba 305-0046, Japan

Jun Nakamura
RIKEN (The Institute of Physical and Chemical Research), Wako, Saitama 351-0198, Japan
(Received 22 October 1999)

We have studied strain-relaxation processes in InAs heteroepitaxy on GaAs(111)*A* using rocking-curve analysis of reflection high-energy electron diffraction. Strain relaxation in the direction parallel to the surface occurs at ~1.5 bilayers (BL) thickness. On the other hand, the lattice constant in the direction normal to the surface remains almost unchanged below ~3 BL thickness and is estimated to be ~3.3 Å. This value, slightly larger than that of bulk GaAs (3.26 Å), does not quite reach the value predicted by classical elastic theory, 3.64 Å. The present result has been supported by the first-principles total-energy calculations.

PACS numbers: 68.55.–a, 61.14.Hg, 68.35.Bs, 81.15.Hi

In heteroepitaxy, strain and strain relaxation play an important role in determining the structural and electronic properties of overlayers. For a strained, unrelaxed overlayer, lattice constants of the overlayer and a substrate are identical in the plane of the interface up to a certain (critical) thickness. As the strain in the overlayer is relaxed beyond the critical thickness, its in-plane lattice constant changes toward its bulk value. On the other hand, information about the strain relaxation in the direction normal to the surface has so far been lacking. Although it has recently been demonstrated that high-energy x-ray photoelectron diffraction can be used to directly determine strain in the surface-normal direction at a strained, lattice-mismatched heterojunction [1,2], no method has so far been used to observe how the strain in this direction is relaxed as growth proceeds.

This Letter reports the first *in situ* measurements of strain in the surface-normal direction during heteroepitaxy. For this purpose, we have used rocking-curve analysis of reflection high-energy electron diffraction (RHEED) based on dynamical diffraction theory, which has been used to determine atomic structures of crystal surfaces [3–7]. Here, we have applied this technique to *in situ* monitoring of strain-relaxation processes in InAs heteroepitaxy on GaAs(111)*A*.

In the InAs/GaAs system, while coherent three-dimensional growth occurs on the (001) surfaces, InAs grows in a layer-by-layer mode on the (110) [8] and (111)*A* [9] surfaces, despite a large lattice mismatch of ≈7.1%. Thus we can measure RHEED rocking curves from the growing InAs(111)*A* surface in the wide range of film thickness, not only at the initial stage of growth but also after the onset of strain relaxation.

The experiments were performed in a molecular-beam epitaxy system. Clean surfaces of GaAs(111)*A*-(2 × 2) were obtained by growing an undoped homoepitaxial layer on a thermally cleaned GaAs(111)*A* substrate. A detailed description of the apparatus and surface cleaning treatments for the GaAs(111)*A* substrate has been given in our previous papers [6]. The InAs films were grown at 400 °C and an As/Ga flux ratio of ~50. The growth rate of InAs was 0.01 bilayer (BL)/s, which was calibrated by RHEED intensity oscillation measurements on the (001)-oriented InAs substrates.

RHEED rocking curves of the growing InAs films were measured at the azimuthal angle of 7.5° off from the $[2\bar{1}\bar{1}]$ direction with an electron energy of 15 keV. At this azimuth, the lattice constant in the surface-normal direction can be determined from the rocking-curve analysis without considering multiple scattering due to the potential variation parallel to the surface [3]. The glancing angle of the incident electron beam was changed with intervals of 0.015°. While rocking curves were measured in the range of glancing angle between 0.2° and 4.2°, only the 444 Bragg peak at ~3° was used to determine the surface-normal lattice constant of InAs. The reason is that below ~2.5° the shape of the rocking curve significantly changed as growth proceeds, which can be ascribed to the change in the growth-front morphology during InAs growth.

Figure 1(a) shows variations in the in-plane lattice constant, d_{110}, of the growing InAs film, which were measured from the distance between the 11 and $\bar{1}\bar{1}$ reflections in the RHEED patterns. The glancing angle of the incident electron beam was ~2.9°. Below ~1.5 BL, a slight variation in the value of d_{110} was observed: the d_{110} value runs through a maximum and has a minimum at 1 BL. Such a behavior is ascribed to the elastic distortion of InAs lattice during layer-by-layer growth [10]. The d_{110} value drastically increases with film thickness beyond ~1.5 BL. After 100 BL growth, there is a 90.2% relaxation, in good agreement with previous results based on the scanning tunneling microscopy and transmission electron diffraction observations [9].

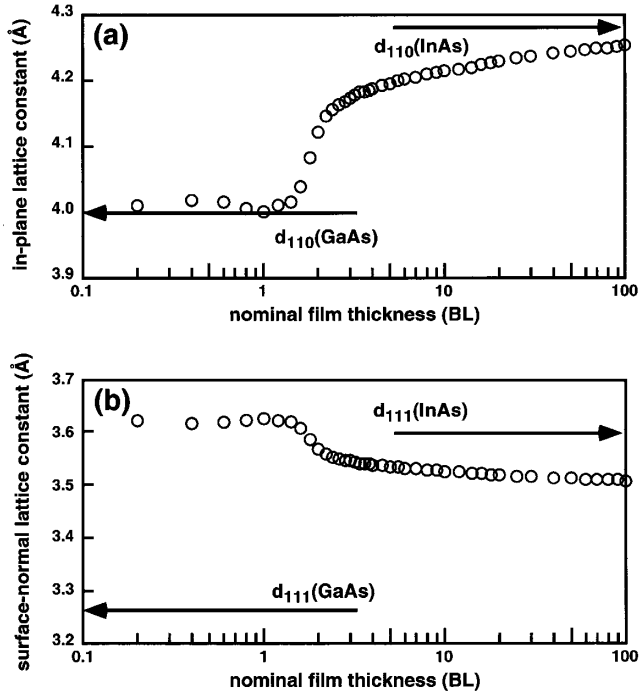

FIG. 1. (a) In-plane lattice constant of the InAs(111)*A* film on the GaAs(111)*A* substrates as a function of InAs film thickness. (b) Surface-normal lattice constant calculated from (a) on the basis of classical elastic theory.

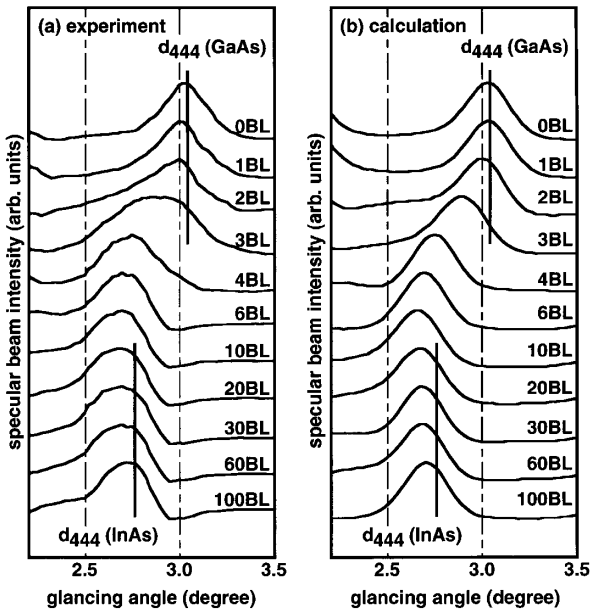

FIG. 2. Measured and calculated RHEED rocking curves from the InAs(111)*A* film growing on the GaAs(111)*A*-(2 × 2) substrates.

According to classical elastic theory, the strain in the surface-normal direction is given by $\varepsilon_{\perp} = (a_{\parallel} - a)/\sigma a$, where a and $a_{\parallel}$ are the in-plane lattice constants of bulk InAs and the growing InAs film, respectively. Poisson's ratio σ of the InAs layer grown on the (111)*A* substrate is $\sigma = (c_{11} + 2c_{12} + 4c_{44})/(2c_{11} + 4c_{12} - 4c_{44})$, where c_{11}, c_{12}, and c_{44} are the elastic coefficients of InAs [11]. Using these equations and the result in Fig. 1(a), we have calculated the lattice constant in the surface-normal direction, d_{111}, as shown in Fig. 1(b). These calculated values are, however, inconsistent with the present RHEED rocking-curve results, as we will show below.

Figure 2(a) shows RHEED rocking curves measured from the growing InAs layers. The peak is shifted to lower glancing angles above 2.0 BL, and reaches a maximum displacement from its initial position in the range of 10–20 BL. On further deposition, the peak position is slightly displaced towards the opposite direction and approaches that of bulk InAs.

In order to convert the position of the peaks in Fig. 2 into the surface-normal lattice constant, we have performed RHEED intensity calculations on the basis of dynamical diffraction theory. RHEED intensities from the growing InAs film with varied film thickness on the GaAs(111)*A* substrate (20-BL thickness) were calculated using the multislice method [12]. We assumed perfect layer-by-layer growth, in which the potential of the incomplete bilayer of the growing surface is proportional to the layer coverage.

Fourier coefficients of the elastic scattering potential were obtained from the atomic scattering factors for free atoms [13]. A correction due to condensation was made to fit the positions of bulk Bragg peaks at large glancing angles. For instance, the resulting mean inner potential of bulk GaAs (InAs) was 13.6 eV (13.9 eV). The imaginary part of the crystal potential for inelastic scattering is assumed to be 12% of its real part. The Debye-Waller parameters were taken to be 2.56 and 0.96 Å^2 for the outermost bilayer and bulk layers, respectively. The calculated rocking curves were convoluted with a Gaussian which has a full width at half maximum of 0.1°, corresponding to the experimental resolution.

In the present RHEED calculation, surface reconstruction of the growing film was taken into account [14]. It is widely accepted that the (111)*A* surfaces of GaAs, InAs, GaSb, and InSb have the vacancy-buckling structure [15–18], in which 0.25 ML of group III atom is missing at the outermost layer, and the remaining three surface atoms recede towards the bulk as shown in Fig. 3. As far as we know, there is practically no experimental information on the surface atomic coordinates for InAs, while the surface bilayers of GaAs and InSb have the interlayer spacings of ~20% of their bulk values [15,18]. Thus, the surface In atoms of the growing InAs film are assumed to be displaced downward by 80% of the bulk interlayer spacing. Although only atomic displacements in the first layer are considered in the present analysis, most features in the rocking curve measured from the GaAs(111)*A*-(2 × 2) surface are well reproduced in the calculated curves.

In the present analysis, the value of d_{111} was systematically varied and a search for the d_{111} value that minimizes the reliability (R) factor defined in Ref. [5] was carried out. Shown in Fig. 2(b) are calculated rocking curves which give optimal agreement with experiment.

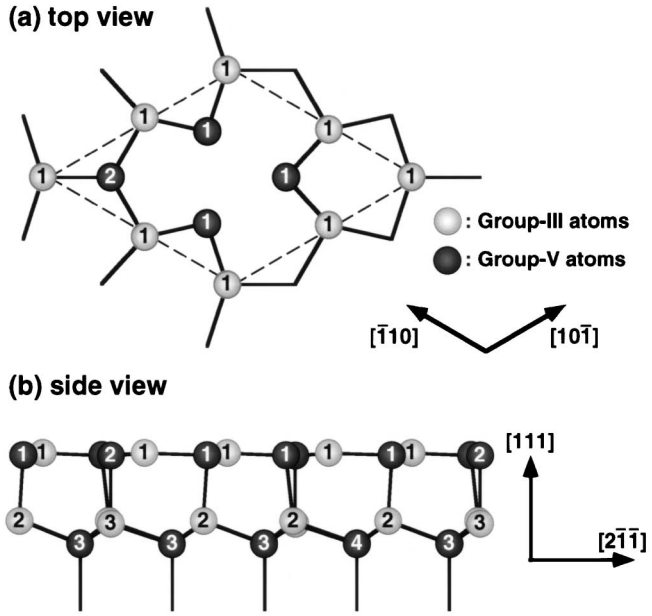

FIG. 3. Vacancy-buckling model for the (111)*A*-(2 × 2) surface of III-V compound semiconductors.

The peak positions of the measured rocking curves [Fig. 2(a)] are well reproduced in the calculated ones, although the shape and width of the peak are slightly different between them.

In Fig. 4(a), the value of d_{111} is plotted as a function of film thickness with errors evaluated from the half-width of the range where the R factor is smaller than $1.1 \times R_{\min}$ [5]. Below ~3 BL, the d_{111} value is somewhat in excess of that of the GaAs substrate (3.26 Å), but is significantly smaller than that predicted by elastic theory (3.64 Å). This is further confirmed by visual inspection of rocking curves: The shape of the rocking curves calculated for larger d_{111} values (e.g., >3.4 Å) is quite different from that of the measured rocking curves in this thickness range. While 1.5 BL is the point when the in-plane lattice constant begins to increase [Fig. 1(a)], the data in Fig. 4(a) exhibit nothing particularly remarkable at this thickness.

Above ~3 BL, the d_{111} value is drastically increased, and the data show a maximum expansion (~135%) between 10 and 20 BL. Through the subsequent growth, the d_{111} value gradually decreases, which is closely related to the progressive increase in the in-plane lattice constant [Fig. 1(a)]. Thus it appears likely that elastic theory can be applied to the InAs films thicker than 20 BL.

The present RHEED analysis has shown that the initially grown InAs lattice is under compressive strain in the surface-normal direction as well as in the surface-parallel direction, contrary to the prediction by elastic theory. In order to address the question as to whether such growth features are energetically favorable, we performed first-principles total-energy calculations [19,20]. A slab geometry is used for the simple calculation, which has the supercell consisting of 2 BL of the GaAs substrate, of 1 BL of the InAs overlayer, and of a vacuum region

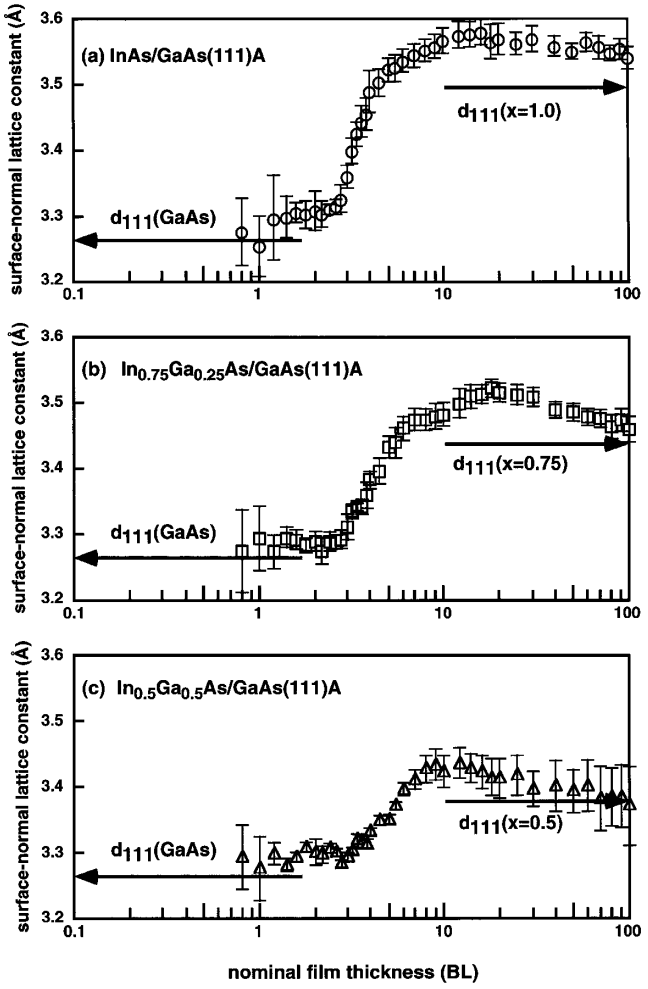

FIG. 4. Surface-normal lattice constant of the InAs (a), $In_{0.75}Ga_{0.25}As$ (b), and $In_{0.5}Ga_{0.5}As$ (c) films on the GaAs(111)*A* substrates as a function of InAs film thickness.

corresponding to 3 BL in thickness. The front side of the slab is composed of the (2 × 2) vacancy buckling model (Fig. 3) [15–18]. We restricted our calculations to the InAs(1 BL)/GaAs(111)*A* model because of the following reasons. The InAs film at 1 BL is fully strained and dislocation free, while the misfit dislocations are formed at the InAs/GaAs(111)*A* interface between 1 and 2 BL, detailed atomic structure of which is not clear. In addition, the unit cell of the model containing dislocations is too large (~60 Å in size [9]) for the first-principles calculation. Nevertheless, the present calculation provides strong support for the experimental result at 1 BL, as we will show below.

Shown in a part of Table I are the surface-normal atomic coordinates for InAs(1 BL)/GaAs(111)*A* after the structure optimization: the surface In atoms are located ~2.68 Å above the outermost Ga layer of the bulk-terminated GaAs(111)*A* substrate. On the other hand, the RHEED result in Fig. 4(a) shows that the growing InAs film has the d_{111} value of ~3.3 Å at 1 BL. If we recall that, in the present RHEED analysis, the surface In atoms

TABLE I. Surface-normal atomic displacements from bulk positions for InAs(1 BL)/GaAs(111)*A* and GaAs(1 BL)/GaAs(111)*A* (in Å). The heights of In(1), Ga(1), As(1), and As(2) atoms are measured from the position of the outermost Ga layer of the bulk-terminated GaAs(111)*A* substrate.

Notations in Fig. 3	InAs(1 BL)/GaAs	GaAs(1 BL)/GaAs
In(1) [Ga(1)]	+2.678	+2.638
As(1)	+2.624	+2.632
As(2)	+2.392	+2.454
Ga(2)	+0.145	+0.141
Ga(3)	−0.086	−0.061
As(3)	−0.012	+0.015
As(4)	+0.113	+0.065

are assumed to be displaced downward by 80% of the bulk interlayer distance (i.e., by 20% of the surface-normal lattice constant), the surface-normal atomic coordinate of In is estimated to be ~2.6 Å, in good agreement with the result in Table I. The present RHEED result is further confirmed by the calculations for GaAs(1 BL)/GaAs: Atoms in the outermost bilayers of InAs and GaAs have the almost identical surface-normal atomic coordinates, as shown in Table I.

We have obtained further interesting results by observing the strain-relaxation processes of $In_xGa_{1-x}As$ on GaAs(111)*A*. On one hand, the in-plane strain relaxation occurs at 2.2 and 3.5 BL for $x = 0.75$ and 0.5, respectively (not shown). On the other hand, as shown in Fig. 4, irrespective of the value of x, the $In_xGa_{1-x}As$ film has the d_{111} value of ~3.3 Å below ~3 BL, beyond which the d_{111} value abruptly increases. This means that the strain relaxation in the surface-normal direction hardly depends on the degree of lattice mismatch, in marked contrast to the in-plane case. This is also evidenced by the fact that all the d_{111} data in Fig. 4 show maxima in the range of 10–20 BL, followed by the gradual decrease in d_{111}.

Another noteworthy finding in Fig. 4 is that the d_{111} value at 100 BL approaches the corresponding bulk value (indicated by arrows in Fig. 4) as the nominal In content is decreased. On the other hand, the in-plane strains have relaxed by 90.2, 96.0, and 104.8% for $x = 1.0$, 0.75, and 0.50, respectively [21]. These results can be explained by considering the surface segregation of In during $In_xGa_{1-x}As$ growth: The segregation efficiency decreases with increasing x value from 0.5 to 1.0 [22]. Since, as mentioned above, the in-plane strain relaxation occurs relating closely to the In content, the surface regions of the $In_xGa_{1-x}As$ film, where the In concentration is enriched, are expected to have the larger in-plane lattice constant than the nominal bulk value. Thus, we can explain why a decrease in the nominal In content facilitates the in-plane strain relaxation. Then, the results for the surface-normal lattice constants, which is insensitive to the In content, can be qualitatively explained by classical elastic theory.

In conclusion, rocking-curve analysis of RHEED has been used to study the strain-relaxation processes in $In_xGa_{1-x}As$/GaAs(111)*A* heteroepitaxy. At the initial stage of the growth, the $In_xGa_{1-x}As$ lattice is under compressive strain in both surface-normal and surface-parallel directions. The in-plane strain relaxation occurs closely relating to the In content in the $In_xGa_{1-x}As$ film, while the strain-relaxation processes in the surface-normal direction scarcely depends on it. The present results provide us new opportunities for a better understanding of strain-relaxation mechanism in heteroepitaxy.

We are indebted to Dr. T. Hanada for use of the RHEED intensity calculation program. We wish to thank Dr. J. Yamauchi and Professor S. Watanabe for use of the first-principles calculation program. This study, partly supported by NEDO, was performed at JRCAT under the research agreement between NAIR and ATP.

[1] S. A. Chambers and V. A. Loebs, Phys. Rev. Lett. **63**, 640 (1989); Phys. Rev. B **42**, 5109 (1990).
[2] R. Gunnella *et al.*, Phys. Rev. B **54**, 8882 (1996).
[3] A. Ichimiya, Surf. Sci. **192**, L893 (1987).
[4] S. Kohmoto and A. Ichimiya, Surf. Sci. **223**, 400 (1989); A. Ichimiya, S. Kohmoto, T. Fujii, and Y. Horio, Appl. Surf. Sci. **41/42**, 82 (1989).
[5] T. Hanada, S. Ino, and H. Daimon, Surf. Sci. **313**, 143 (1994); T. Hanada, H. Daimon, and S. Ino, Phys. Rev. B **51**, 13 320 (1995).
[6] A. Ohtake *et al.*, Phys. Rev. B **59**, 8032 (1999); Appl. Phys. Lett. **74**, 2975 (1999); Phys. Rev. B **60**, 8326 (1999); **60**, 8713 (1999).
[7] J. M. McCoy *et al.*, Surf. Sci. **261**, 29 (1992); Phys. Rev. B **48**, 4721 (1993); Surf. Sci. **418**, 273 (1998).
[8] J. G. Belk *et al.*, Phys. Rev. Lett. **78**, 475 (1997).
[9] H. Yamaguchi *et al.*, Phys. Rev. B **55**, 1337 (1997).
[10] J. Massies and N. Gradjean, Phys. Rev. Lett. **71**, 1411 (1993).
[11] R. M. Martin, Phys. Rev. B **1**, 4005 (1970).
[12] A. Ichimiya, Jpn. J. Appl. Phys. **22**, Suppl. 1, 176 (1983); **24**, 1365 (1985).
[13] P. A. Doyle and P. S. Turner, Acta Crystallogr. Sect. A **24**, 390 (1968).
[14] The atomic displacements parallel to the surface were neglected, because rocking curves obtained at one-beam conditions scarcely depend on them.
[15] S. Y. Tong, G. Xu, and W. N. Mei, Phys. Rev. Lett. **52**, 1693 (1984).
[16] D. J. Chadi, Phys. Rev. Lett. **52**, 1911 (1984).
[17] J. Bohr *et al.*, Phys. Rev. Lett. **65**, 452 (1990).
[18] A. Ohtake and J. Nakamura, Surf. Sci. **396**, 394 (1998).
[19] D. Vanderbilt, Phys. Rev. B **41**, 7892 (1990).
[20] J. Yamauchi *et al.*, Surf. Sci. **341**, L1037 (1995); Phys. Rev. B **54**, 5586 (1996).
[21] In this estimation, the lattice constants of $In_xGa_{1-x}As$ are calculated on the basis of the linear dependence of the lattice constant on the nominal composition (Vegard's law).
[22] J. M. Moisson *et al.*, Phys. Rev. B **40**, 6149 (1989).