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Growth mode and surface morphology of a GaN film deposited along the N-face polar direction on *c*-plane sapphire substrate

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The dependence of polar direction of GaN film on growth conditions has been investigated by changing either the group-V/group-III ratio (V/III ratio) in supplying the source gas or the deposition rate. GaN films were deposited on a nitrided sapphire by two-step metalorganic chemical vapor deposition. The surface morphology changed from flat hexagonal to pyramidal hexagonal facet with the increase of V/III ratio. However, the polar direction of GaN on an optimized buffer layer of 20 nm thickness was N-face $(-c)$ polarity, independent of both the V/III ratio and the deposition rate. The polarity of the GaN epitaxtial layer can be determined by that of an interface (nitrided sapphire, annealed buffer layer or GaN substrate) at the deposition of GaN epitaxial layer. The higher V/III ratio enhanced the nucleation density, and reduced the size of hexagonal facets. The nuclei, forming the favorable hexagonal facets of wurtzite GaN, should grow laterally along the $\{10\bar{1}0\}$ directions to cover a room among the facets until coalescence. After coalescence, $-c$ GaN growth on a flat hexagonal facet results in a pyramidal hexagonal facet. The growth mode for $-c$ GaN has been discussed with respect to surface structure and migration length of adsorbing precursors, in comparison with Ga-face $(+c)$ GaN. *© 2000 American Institute of Physics.* [S0021-8979(00)02114-9]

I. INTRODUCTION

Major progress in growth techniques[1–5] for GaN related materials has led to the success of blue-light emitting devices.[6] The impact of this progress has extended not only to the commercialization of the blue laser diode, but has also contributed to the understanding of heteroepitaxial growth in a highly lattice-mismatched system. Generally, the surface reactions are important for thin film growth.[7] The adsorption, migration and incorporation of precursors are influenced by either the atomic arrangement of a substrate or by a growing surface. Since GaN has a wurtzite crystal structure having polarity along the *c*-axis direction, two surface structures correspond to the polarity of GaN: the (0001) Ga face and the $(000\bar{1})$ N face, which are defined as $+c$ or $-c$, respectively, in the present article. The $+c$ surface is composed of three dangling bonds of nitrogen that point up toward the *c*-plane surface, and the $-c$ surface is comprised of a single dangling bond that points upward. This difference in surface structure indicates the importance of GaN polarity when considering the growth mode of a wurtzite GaN, as well as the development of potential GaN materials for device application.

In contrast to the other III–V materials, devices based on GaN are fabricated on the *c* plane, which is a polar axis of the material. Both the strain of GaN film and the polar direction of GaN growth should determine the direction of the piezoelectric field.[8] This electric field is so crucial to the device performance. For example, the optical characterization of lnGaN quantum wells has been studied intensively with respect to the piezoelectric field.[9–11] The application of GaN materials in electronic devices such as the thin film transistor[12,13] requires consideration of polar direction when designing these devices. Thus, the device application of GaN based materials requires not only investigation of the polarity of GaN film,[14–18] but also an understanding of the mechanism that determines the polar direction of GaN growth.

We have investigated the determination of GaN polarity using coaxial impact collision ion scattering spectroscopy (CAICISS) analysis.[19] The polarity was found to be controllable by the GaN deposition procedures: (1) the nitridation of sapphire substrate resulted in GaN growth with $-c$ polarity, and (2) the polarity should be attributed to that of the buffer layer annealed by heating the substrate up to 1040 °C[19,20] Since the GaN polarity is attributed to that of the annealed buffer, we were able to control the polarity by changing the

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annealing time.[19] In addition to this interface, the kinetic effect during the growth must be taken into account when considering determination of the polar direction.

The purpose of the present study is to investigate $-c$ GaN growth on a nitrided sapphire by changing the growth conditions and the film thickness. The dependence of polar direction on deposition rate and V/III ratio has been investigated. The hexagonal facet surface morphology of $-c$ GaN is not appropriate for fabricating the devices, even though the $-c$ GaN is theoretically required for devices with improved performance.[21] As the first step to improving the quality of $-c$ GaN, further clarification of film growth is necessary. Although the growth of $+c$ GaN has been clarified to a great degree,[22] this knowledge cannot be applied to $-c$ GaN growth directly because the surface structure of $-c$ GaN is different from that of $+c$ GaN. As far as we know, there has been no report of GaN growth in which the polarity is clearly taken into account. The present report considers growth mode of $-c$ GaN film, using CAICISS to determine the polarity of GaN growth. The surface morphology (hexagonal facet size of shape) for $-c$ GaN is discussed with respect to nucleation density. In addition, the growth mode for $-c$ GaN is also discussed based on the migration length of precursors on the surface structures for $-c$ GaN films, compared to the case for $+c$ GaN films.

II. EXPERIMENT

GaN films were grown by atmospheric pressure metal-organic chemical vapor deposition (MOCVD) via a vertical reactor using tri-methyl gallium (TMG) and ammonia as the group III and group V sources, respectively. GaN films were grown on a c-plane sapphire substrate by the two-step growth technique. After the sapphire substrate surface was exposed to H_2 flow for 5 min at 1080 °C (H_2 cleaning), the surface was nitrided in NH_3 gas flow for 5 min at the same temperature. A 20 nm GaN buffer layer, which had been optimized in advance,[20,23] was deposited at 600 °C under a constant V/III ratio of 15 000. The buffer layer was subsequently annealed by ramping the temperature up to 1040 °C under a gaseous mixture of N_2, H_2 and NH_3(2:3:3). GaN epitaxial layer (epi-layer) was deposited on an annealed buffer layer at 1040 °C. Since the crystallinity of GaN films deposited at lower temperatures is poor, in the present study, the temperature for epi-layer deposition was fixed at 1040 °C. For the samples, the V/III ratio in the epitaxial layers was varied over the range from 1000 to 30 000 (TMG1.0–4.2 sccm, $NH_3$250–1500 sccm). When TMG and NH_3 flow rates were changed, the gas flow rates of mixed gases of H_2 and N_2 were also changed in order to maintain the total gas flow rate constant at 2.0 ℓ/min. The film thickness was approximately 1 μm. In our deposition system, the deposition rate is limited by supplying TMG source gas. In order to observe the growth mode on the nitrided sapphire, the film thickness was varied from approximately 50 to 1200 nm for the films that were deposited under the V/III ratios of 5000, 15 000 and 30 000. In order to investigate the kinetic effect on GaN growth, these samples were deposited by varying not only the V/III ratio, but also the deposition rate, from approximately 0.9 to 5.0 μm/h, the entire range of our system. In addition, a GaN homoepitaxial growth was performed on the GaN substrate that was grown on c-plane sapphire by hydrogen vapor phase epitaxy (HVPE) and molecular beam epitaxy (MBE) under the same conditions.

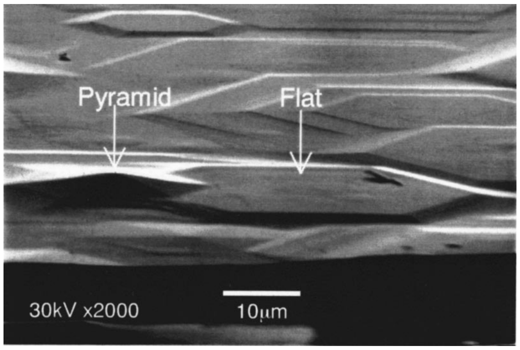

FIG. 1. SEM image of GaN surface deposited under a V/III ratio of 15 000 on nitrided sapphire substrate observed from a 10° incline from the horizontal plane.

Optical microscope, scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used for the observation of the GaN surface morphology. Film thickness was evaluated by observing the cross section via SEM. After a surface was scanned over a wide area ($10\,\mu$m $\times 10\,\mu$m) by AFM, the detailed structure was observed over a small area (200 nm × 200 nm). CAICISS analysis[24,25] was used to determine the polarity of the GaN film. The polarity of the GaN film was determined by comparing these results with previously reported CAICISS results for GaN.[19] The full width at half maximum (FWHM) for rocking curves for on-axis (0002) and off-axis ($10\bar{1}2$) planes was evaluated by four-circle x-ray diffraction as the crystalline quality of GaN films.

III. RESULTS

A. Surface morphology

All surfaces of the 1 μm GaN films that were deposited on the nitrided sapphire substrates showed hexagonal facet morphologies when the V/III ratio during GaN epitaxial growth was varied over the range from 1000 to 30 000. Figure 1 illustrates the surface morphology of GaN film deposited under a V/III ratio of 15 000 as observed by SEM. Two types of hexagonal facets were observed in the GaN surface: smooth facets (flat facet surface) and pointed facets (pyramidal facet surface). The size of hexagonal facets became larger when the V/III ratio was decreased down to 5000, as shown in Fig. 2. Although the surface of GaN deposited under a V/III ratio of 1000 had a hexagonal facet surface, its size was much smaller than that deposited under a V/III ratio of 5000, and its corners were not sharp (forming a distorted dodecagon), presumably due to the excess Ga supply. A V/III ratio of 1000 ratio is considered to be too low for our MOCVD-GaN deposition system. The right axis of Fig. 2

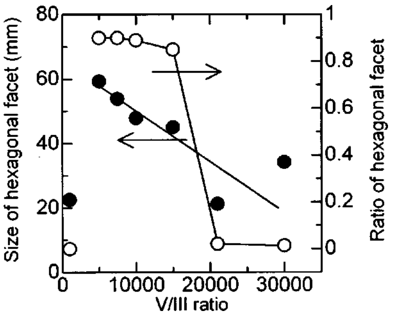

FIG. 2. Dependence of average size of hexagonal facet (left axis) and ratio of flat to pyramidal hexagonal facets (right axis) on V/III ratio from 1000 to 30 000 in the GaN epilayer growth process.

presents the ratio of flat facet in the observed area. This ratio was evaluated based on optical microscope photographs taken at 500× magnification. The number of pyramidal hexagonal facets increased with the increase in the V/III ratio in the growth process.

The SEM image shown in Fig. 1 was observed from a 10° incline from the horizontal plane. The sidewalls of both the flat and the pyramidal hexagonal facets are not steep, but rather are sloped. The angle of these slopes is approximately 7°–8° and is much smaller than that of the facet plane $(10\bar{1}1)(54.74°)$ observed for GaN films deposited by selective growth[26] or lateral epitaxial overgrowth.[27,28] Since no plane in the structure of hexagonal crystalline GaN corresponds to this angle, we hypothesized that the slopes of the hexagonal facets may consist of a step and terrace structure. In order to verify this hypothesis, the GaN surface was also observed by AFM. First, the AFM observation was performed for the GaN film (V/III ratio of 7500) that had a surface that was covered primarily by flat hexagonal facets in the range of up to $10\times10\,\mu\mathrm{m}^2$ [Fig. 3(a)]. After this procedure, the cantilever of AFM was moved down to point B on the slope, and its structure was observed in detail over the small area of $200\times200\,\mathrm{nm}.^2$ As expected, the step and terrace structure was observed in the slope of the hexagonal facets, as shown in Fig. 3(b). The line profile shows a terrace width of approximately 300 Å and a step height of approximately 20 Å. The step should consist of $(10\bar{1}0)$ planes. AFM images of the GaN film (V/III ratio of 30 000) having pyramidal hexagonal facets are shown in Figs. 3(c)–3(e). A hole having a diameter of approximately 300 nm was frequently observed in the center of the pyramidal hexagonal facet, as shown in Fig. 3(e). According to a report by Qian, Rohrer, and Skowronski, the presence of a hole in the center of a facet, called a nano-tube or an open core, can be attributed to spiral growth.[29] Although further confirmation is required, the pyramidal hexagonal facets are believed to be caused by spiral growth because (1) the angle of the slope in the pyramidal hexagonal facet becomes steeper as it approaches the top of the facet, and (2) the terraces that are located on both sides are not symmetric, as shown in Fig. 3(d). Thus, the surface morphology of the GaN epi-layer on a nitrided sapphire was found to vary from flat to pyramidal hexagonal facet as the V/III ratio was varied.

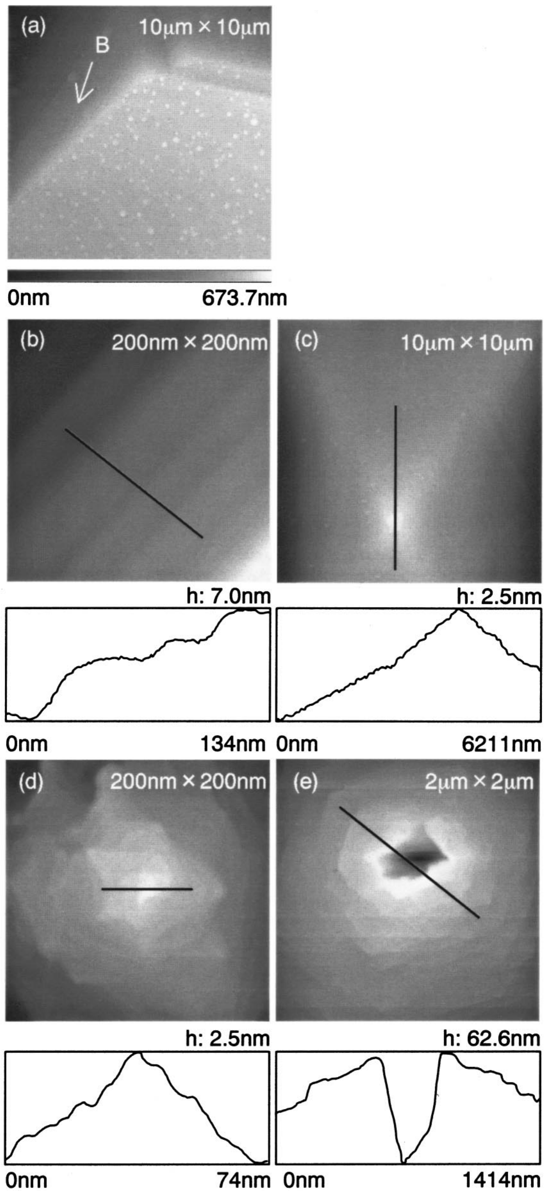

FIG. 3. AFM images of a GaN film surface that is covered by primarily flat hexagonal facet (V/III ratio of 7500): (a) $10\times10\,\mu\mathrm{m}^2$ and (b) $20\times20\,\mathrm{nm}^2$ images observed at point B in (a). GaN film surface covered primarily by pyramidal facet: (c) $10\times10\,\mu\mathrm{m}^2$, (d) $200\times200\,\mathrm{nm}^2$ observed at the top area of (c), and (e) a pyramidal hexagonal facet having a hole in the center of hexagonal facet in the range of $2\times2\,\mu\mathrm{m}^2$. The lines in the images indicate the line profiles shown below each image.

B. Surface morphology and polarity

In order to investigate the correlation between the growth conditions (V/III ratio or deposition rate) and the polar direction, we measured CAICISS for GaN films that were deposited at growth rates of 0.9 μm/h (V/III ratio of 21 000) or 5.0 μm/h (V/III ratio of 1000). The surface of the former GaN film (0.9 μm/h) was covered primarily with pyramidal hexagonal facets, and that of the latter GaN film (5.0 μm/h) is described in Sec. III A. As the CAICISS spectra in Fig. 4 show, both of these GaN films have $-c$ polarity, as revealed by comparison with $-c$ GaN CAICISS spectra. Since the other GaN films shown in Fig. 2 have hexagonal facets, $-c$ polarity is deduced for all the GaN films from our

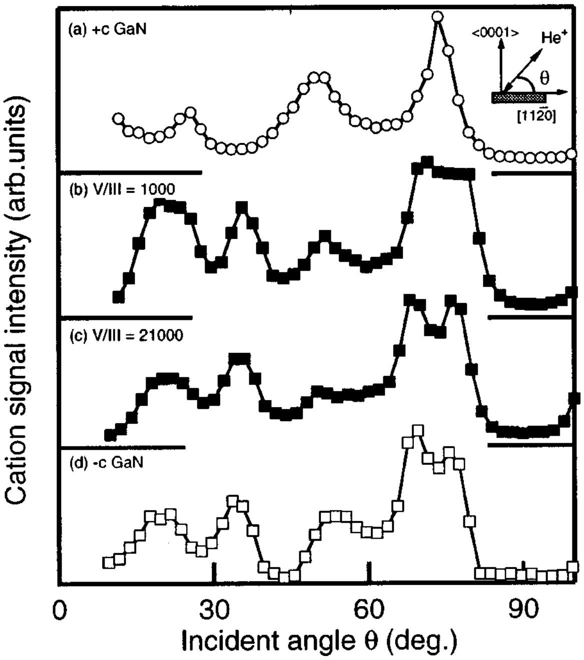

FIG. 4. CAICISS spectra of GaN film on nitrided sapphire substrate deposited under V/III ratios of (b) 1000 and (c) 21 000. The spectra for (a) $+c$ GaN and (d) $-c$ GaN are also included as a reference.

previous experiment.[19] It was confirmed that GaN films deposited on the nitrided sapphire had $-c$ polarity in spite of their surface morphology, and that the kinetic effect (growth rate) had no influence on the polar direction of GaN growth.

Furthermore, in order to confirm the influence of polarity at the interface, the homoepitaxial growth was carried out on GaN substrates grown by HVPE or MBE, for which polarity was decided as $+c$ and $-c$, respectively, by CAICISS. As shown in Fig. 5, which indicates the CAICISS spectra, the GaN polarities on HVPE and MBE GaN are $+c$ and $-c$, respectively. That is the polarity of the GaN grown on both of the GaN substrates was attributed to the polarity of the substrates. Although the growth conditions, such as V/III ratio and deposition rate, can cause the surface morphology to vary, the polarity of an interface (annealed buffer layer or GaN substrate) at an epi-GaN growth can determine polar direction.

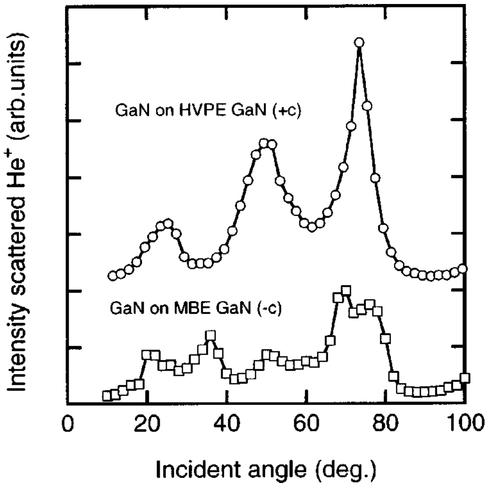

FIG. 5. CAICISS spectra of GaN films on (a) HVPE GaN, and (b) MBE GaN substrate.

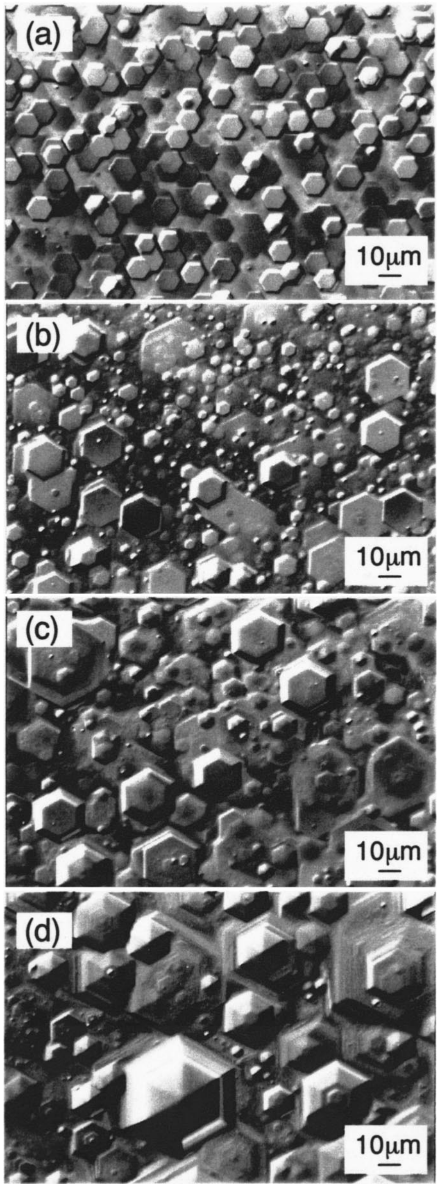

FIG. 6. Microscopic image of GaN surface having a varying thickness under a V/III ratio of 30 000 deposited by controlling growth time on nitrided sapphire substrate: for film thicknesses of (a) 100 nm (5 min deposition), (b) 200 nm (10 min), (c) 400 nm (20 min) and (d) 1200 nm (60 min).

C. Film thickness dependence of GaN growth with $-c$ polarity

From the above-mentioned study in Sec. III B, all GaN films grown on nitrided sapphire have $-c$ polarity. We have investigated growth mode and variation in surface morphology as related to V/III ratio.

GaN films were grown on the nitrided sapphire substrate under a V/III ratio of 5000, 15 000, and 30 000 in GaN epi-layer by varying the film thickness. In this case, the deposition rate effect can be neglected due to the constant TMG flow rate in GaN epi-layer deposition. Figure 6 shows microscopic images of the surface of $-c$ GaN film when varying

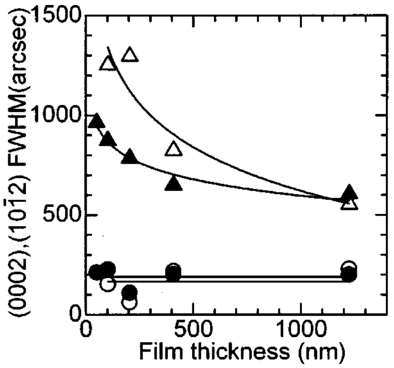

FIG. 7. Variation of rocking curve FWHM value in GaN (0002) (circle) and $(10\bar{1}2)$ (triangle) planes deposited under V/III ratios of 15 000 (closed symbols) and 30 000 (open symbols) on the film thickness measured by four-circle XRD.

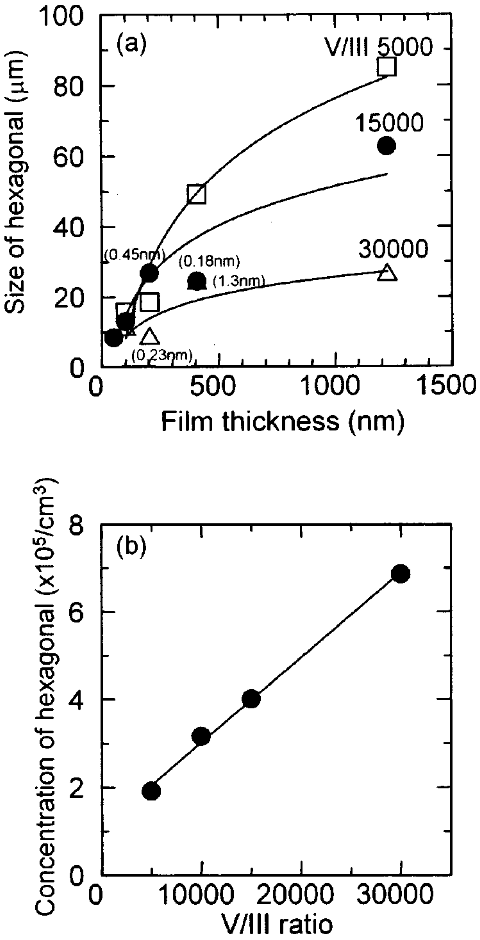

FIG. 8. (a) Dependence of the average size of hexagonal facet on the film thickness for GaN deposited under the various V/III ratios: 5000 (open square), 15 000 (open triangle) and 30 000 (closed circle). (b) Concentration of hexagonal facet on the GaN surface at a film thickness of approximately 100 nm as a function of the V/III ratio.

the deposition time under a V/III ratio of 30 000. The average size of the hexagonal facet is an approximately uniform 10 μm during the initial growth (100 nm), as shown in Fig. 6(a). During the initial growth of $-c$ GaN, a lateral room is observed among the hexagonal facets. The hexagonal facets grow laterally with the increase in film thickness, and nucleation on a flat hexagonal facet at 400 nm is shown in Fig. 6(b). The size distribution of hexagonal facet is enhanced with the increase in deposition time. The surface of this GaN film at a thickness of approximately 1 μm is predominantly covered with pyramidal hexagon [Fig. 6(d)]. Figure 7 shows the relationship between film thickness and the rocking curve FWHM in GaN (0002) and $(10\bar{1}2)$ planes measured by four-circle x-ray diffraction (XRD). Whereas the FWHM value of GaN (0002) is almost constant at 200 arcsec from the initial growth, the FWHM value of GaN $(10\bar{1}2)$ improved with the increase in film thickness. This variation indicates that hexagonal nuclei with high crystallinity along the c axis grow laterally, and that the distribution of in-plane rotation is suppressed with the increase in thickness.

What could determine the size of a hexagonal facet? Figure 8(a) shows the dependence of the average size of hexagonal facet on the film thickness for various V/III ratios. Figure 8(b) shows the density of hexagonal facet of GaN films at a thickness of 100 nm as a function of V/III ratio. Similar to the hexagonal facets shown in Fig. 6(a), the top of all hexagonal facets was flat during the initial growth, despite the V/III ratio. The density of hexagonal facet during initial growth is higher when the V/III ratio increases, indicating that nucleation at initial growth under the high V/III ratio (N rich) occurs easily due to the large amount of nitrogen arriving at the GaN growing surface. The hexagonal facets grow laterally to cover the room until they coalesce with each other. The suppression of in-plane rotation in Fig. 7 is probably due to the coalescence of hexagonal facets. It appears that the lower the nucleation density (lower V/III ratio), the larger the size of hexagonal facet.

As indicated in Fig. 8(a), the surface roughness (R_{ms}) on the flat hexagonal facet of the approximately 400-nm-thick film is much larger (R_{ms}: 1.3 nm) than that at a thickness of less than 200 nm(R_{ms}: 0.23 nm) for a V/III ratio of 30 000. Film having a greater thickness is likely to grow, forming the pyramidal hexagonal facet. For the V/III ratio of 15 000, the surface of the facet at the film thickness of 400 nm is as smooth as R_{ms}: 0.18 nm, which indicates that the nucleation should occur scarcely on the flat hexagonal facet before the coalescence. More deposition is required in order for the coalescence to induce the nucleation on the flat hexagon. According to this scenario, the formation of pyramidal hexagonal facet is expected for the deposition of thicker GaN film, even under the lower V/III ratio. When 2-μm-thick GaN was deposited under a V/III ratio of 5000, the surface of the film was almost covered by pyramidal hexagonal facets.

IV. DISCUSSION

A. Substrate nitridation and GaN polarity

Sapphire substrate is often nitrided as a growth process of GaN deposition. When the sapphire is exposed to NH_3 at high temperature, AlN or AlN_xO_{1-x} was formed.[19,30,31] Our nitridation procedure formed AlN_xO_{1-x} having a thickness of approximately 12 Å, which corresponds to one unit of Al_2O_3 at the surface.[32] The crystallographic orientation between AlN_xO_{1-x} and sapphire was obtained as

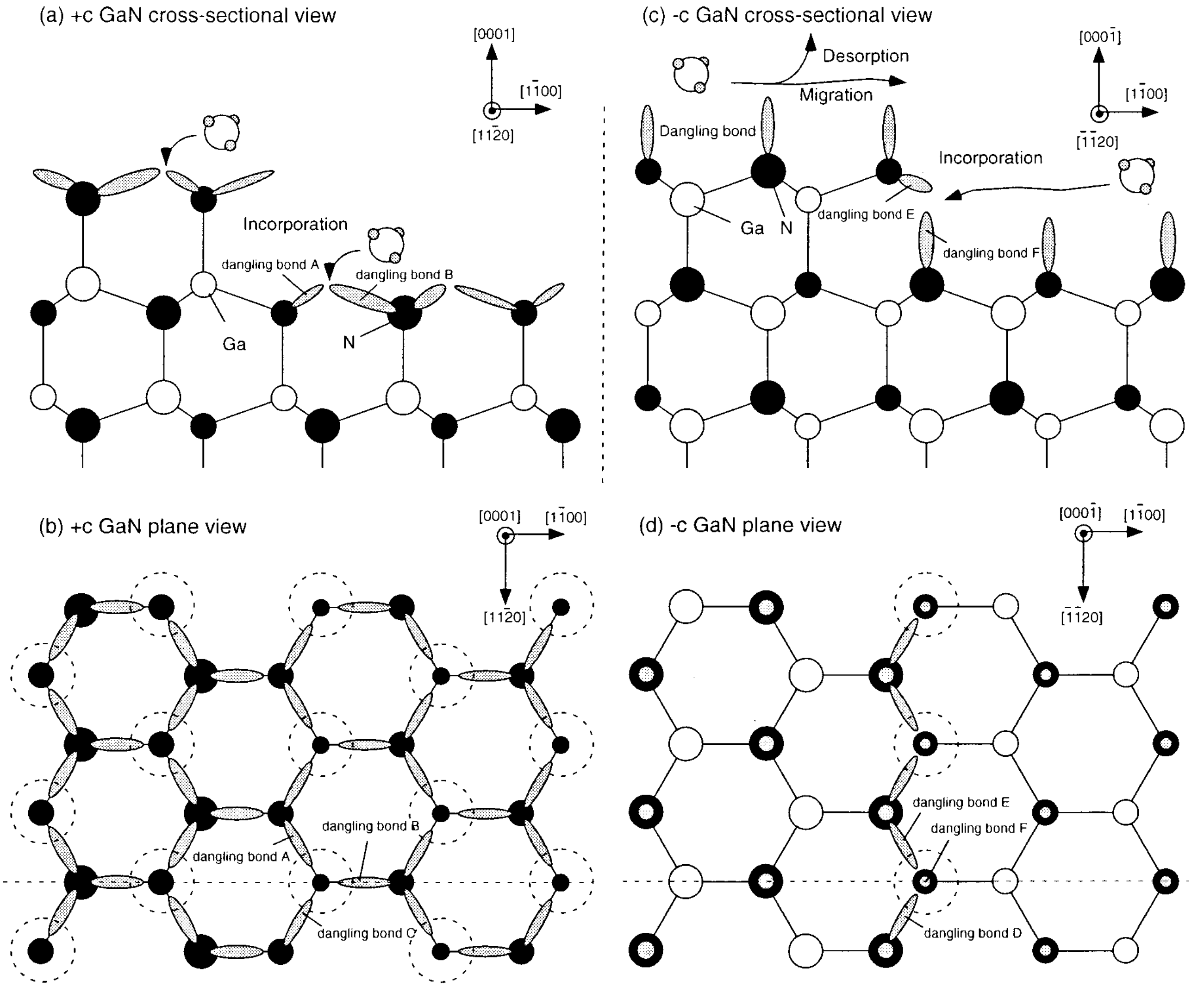

FIG. 9. Atomic arrangement of $+c$ and $-c$ GaN surface structures. (a) and (c) are c-plane views for $+c$ and $-c$ GaN, respectively. (b) and (d) are cross-sectional views along the $[11\bar{2}0]$ direction for $+c$ and $-c$ GaN, respectively.

$[10\bar{1}0]$ AlN_xO_{1-x}//$[11\bar{2}0]$ sapphire. AlN formed by the nitridation has $-c$ polarity.[33] This is considered to be the primary reason why MOCVD-GaN film on a nitrided sapphire has $-c$ polarity.

In GaN film deposition by MBE, a sapphire substrate is typically nitrided by exposure to nitrogen radicals.[34–39] Therefore, it is reasonable that MBE-GaN has $-c$ polarity, as reported by several research groups.[16,40–42] and indicated by the CAICISS analysis for MBE-GaN in the present study. The nitrogen radical is so reactive that nucleation could be greater than in MOCVD. This appears to be similar to the situation of a much higher V/III ratio in the MOCVD process. Based on the obtained results for MOCVD-GaN (Fig. 2), the surface of the MBE-GaN is believed to be composed of tiny hexagonal facets due to a higher nucleation density. Therefore, MBE-GaN is thought to have a rougher surface than MOCVD-GaN, as reported by Angerer *et al.*,[34] and $-c$ polarity, due to the nitridation of sapphire substrate.

B. Growth mode of GaN film having $-c$ polarity

We have confirmed that all GaN films grown on a nitrided sapphire under the conventional MOCVD condition have $-c$ polarity and that the films grow by forming hexagonal facets. The facet growth of $-c$ GaN film seems to be similar to the growth of the GaAs epitaxy on (111)B.[43] Based on the results of GaAs epitaxial growth, during the surface migration process, the absorbed species (Ga) on the growing surface desorb again before adhering to the stable sites. Once the adsorbed Ga atoms are incorporated into a film, As atoms adhere to the adsorbed Ga atoms. The growing surface is covered with As atoms. When the density of the adsorbed species on the surface exceeds a certain value, the adsorbed species can form nuclei as a result of coalescence of the adsorbed species. Once these clusters stick to the substrate surface or growing surface, they will act as growth nuclei and grow larger, gathering the migrating species. Thus, the surface migration and the desorption of adatoms are crucial to the formation of the nuclei.

In order to consider the growth of $-c$ GaN, a schematic diagram of the GaN surface structure is presented in Fig. 9. Figure 9 presents the atomic arrangements of cross section viewed along the $[11\bar{2}0]$ and c plane for $+c$ and $-c$ GaN structures. This is a simple illustration that ignores reconstruction. Taking both the Ga-limited growth and the large

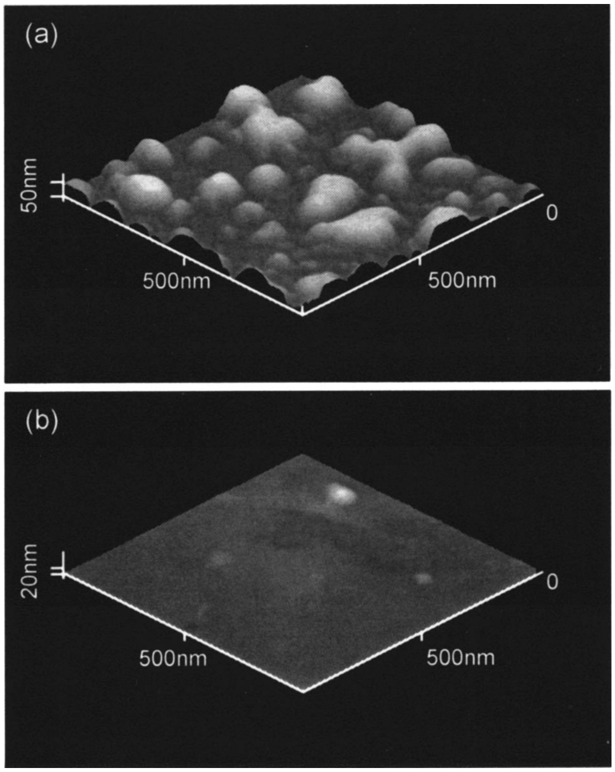

FIG. 10. AFM image (1 μm×1 μm) of 20 nm GaN buffer layers (a) on non-nitrided and (b) on nitrided sapphire substrate after annealing for 10 min by ramping the temperature up to 1040 °C.

amount of nitrogen into account, it can be assumed that the growing surfaces are terminated with nitrogen during the growth. Whereas the surface for $+c$ GaN is covered with nitrogens and their dangling bonds, that for $-c$ GaN is composed of Ga and N atoms in which a single dangling bond points upward.

First, we consider Ga migration on N-terminated surface having $+c$ and $-c$ polarity. Figure 10 shows the surface morphology of the annealed buffer layers on non-nitrided and nitrided sapphire substrate. Although the surfaces of the buffer layer on non-nitrided sapphire $(+c)$ remains rough, that on nitrided sapphire $(-c)$ is quite smooth. These surfaces act as the interface for the precursors of epi-GaN growth. According to a report by Zywiets, Nuegebauer, and Scheffler[44] Ga migration length on $-c$ GaN should be longer than on $+c$ GaN due to the number of nitrogen dangling bond [Figs. 9(a) and 9(c)]. Since Ga adatom has to break three Ga–N bonds during migration, a shorter migration length can be expected for $+c$ GaN than for $-c$ GaN.

Next, we consider the sites at which Ga species are incorporated. Both surface structures have step edges. For $+c$ GaN, three nitrogen dangling bonds point up, as illustrated in Fig. 9(a). The sites indicated by the dotted circles in Fig. 9(b), as represented by the dangling bonds A, B and C, are rather stable for Ga precursors. Whichever site Ga precursors adsorb on the $+c$ surface, they are likely to be incorporated there with less migration due to three dangling bonds of nitrogen on the surface. As a result, rather than lateral growth, growth along the c axis is dominant for $+c$ GaN. In contrast, for the bond configuration at the step edge of $-c$ GaN [Fig. 9(c)], two dangling bonds of nitrogen on the top surface point down along the $[10\bar{1}0]$ and $[01\bar{1}0]$ directions, and one dangling bond of nitrogen in the second layer points up. The step edge composed of dangling bonds D, E and F in Fig. 9(d) can act as sink for the migrating species. Even if the adatom does not have sufficient migration length to arrive at the step edge, it can desorbe back into the gas phase due to a weaker adsorption of Ga atom to the underlying nitrogen for $-c$ GaN growth. Therefore, the Ga adatom on the $-c$ GaN surface may be more likely to adhere to the site of the step edge at which the step-flow mode dominates. Once a Ga adatom is incorporated into a step edge, its surface can be terminated with nitrogen and another step edge can be created continuously. Consequently, $-c$ GaN growth takes place along the $\langle 10\bar{1}0\rangle$ directions where these facets are formed, until the nuclei coalesce with each other.

As shown in Sec. III A, the pyramidal facet should be caused by spiral-like growth on a flat hexagonal surface. However, the hexagonal surface was so smooth that both step-terrace structure and dark spots, corresponding to threading dislocation by a screw component,[45] could not be observed by AFM. Since the step edge barely works as the sink after the coalescence, the growth mode is transferred to spiral-like growth. Although the origin of the different growth mode remains unclear, impurities are regarded as one of the candidates for spiral growth, as reported by Liliental-Weber *et al.*,[46] because the result of our secondary ion mass spectrometry (SIMS) analysis[47] reveals that a greater amount of carbon impurity in GaN having $-c$ polarity was incorporated into the region of pyramidal facet (spiral-like growth). Further research on the origin of spiral-like growth is required.

The following summarizes $-c$ GaN growth based on our experimental results:

(1) Nucleation takes place on a buffer layer having a smooth surface. Since the GaN film growth rate is limited by supplying TMG source gas, the nucleation is more likely to occur under the higher V/III ratio condition (N rich). The larger the V/III ratio, the greater the amount of nucleation that occurs during the initial growth. [Fig. 6(a)]

(2) The hexagonal facets with flat surface, which showed high crystallinity along the c axis, were formed during the initial growth. (Fig. 7). These facets can grow laterally due to the incorporation of Ga adatom at the step edge on the $-c$ GaN growing surface. Since the step-flow growth mode dominates during the initial growth, the hexagonal facets grow laterally until they coalesce [Fig. 8(a)]. The size of the hexagonal facets depends on the nucleation density.

(3) This coalescence predominantly determines the size of the hexagonal facet and improves FWHM of the $(10\bar{1}2)$ plane of the hexagonal facet (Fig. 7). GaN begins to grow even on the flat surface of the hexagonal facet.

(4) After coalescence, the growth along the c axis dominates. A different growth mode (probably spiral growth) occurs and the pyramidal hexagonal facets then increase

under the high V/III ratio. Thus, the surface morphology of GaN having the same thickness can be varied by varying the V/III ratio (Fig. 2).

V. CONCLUSION

We investigated the dependence of polar direction of GaN film on the growth condition of MOCVD. Although the V/III ratio changed the size of the hexagonal facet and/or surface morphology, it had no influence on the polarity. Moreover, the kinetic effect (deposition rate) did not affect the polar direction. MOCVD-GaN on a nitrided sapphire deposited under the conventional growth condition has hexagonal facet and $-c$ polarity.

A step-flow growth mode dominates for $-c$ GaN film during the initial growth. The higher the V/III ratio, the more nucleation occurred. After nucleation on a nitrided sapphire, $-c$ GaN films grow laterally by a step-flow growth mode along the $\langle 10\bar{1}0\rangle$ directions, forming a step and terrace structure. As a result, a hexagonal facet is formed as the favorable structure of $-c$ GaN. Since the hexagonal facets having a flat surface become larger, covering the room among hexagonal facets until they coalesce, the size of the hexagonal facet tends to be smaller as the V/III ratio (higher nucleation density) increases. The different growth mode that occurs after the coalescence can cause pyramidal facet formation. Since the thickness at the coalescence depended on the V/III ratio, the surface morphology, such as flat or pyramidal facets, was observed for $-c$ GaN to have the same thickness when the V/III ratio was varied.

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