Microstructural properties of GaN grown on a Si(110) substrate by gas-source molecular beam epitaxy: Dependence on the ammonia flux

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A R T I C L E I N F O
Article history:
Received 11 September 2014
Received in revised form 29 October 2014
Accepted 15 December 2014
Available online 17 December 2014

Keywords:
Nitrdes
Transmission electron microscopy
Dislocation
Gas-source molecular beam epitaxy
Silicon substrate

A B S T R A C T
The microstructural properties of a GaN thin film grown on a Si(110) substrate under various ammonia (NH₃)-flux conditions were observed to study growth mode and defect evolution. The surface flatness of GaN thin films was improved with the increase of the NH₃ flux while the thickness was decreased by increasing the NH₃ flux. In addition, the crystalline quality of the GaN film grown under the lower NH₃ flux (100 sccm) was better than that of the film under the higher NH₃ flux (400 sccm). The different dislocation behaviors depending on NH₃ fluxes were observed; the low density of dislocations was measured and most of dislocations penetrating the thin film was mixed- and edge-type dislocations when GaN was grown under the low NH₃ flux condition while the high density of dislocation and many mixed- and screw-type dislocations penetrating the film were observed in the GaN film grown under the high NH₃ flux. These phenomena are demonstrated by using a kinetic model related to the role of NH₃.

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1. Introduction
The epitaxial growth of nitride films on a silicon (Si) substrate is one of the most important issues for the research and technology related to the nitride semiconductors because it is possible to achieve many advantages of the substrate, e.g. large size availability, low-cost, and suitability in well-established integration process [1,2].

The Si(100) substrate has four-fold symmetry (square lattice) which is unsuitable for hexagonal symmetry of nitrides. The Si(111) has three-fold symmetry (triangular lattice) which is suitable for nitrides, but it has large lattice matches, e.g. about 16.9% between GaN and Si [3]. The Si(110) is recently considered as one of the most capable candidates for epitaxial growth of nitrides because uniquely biaxial strains stimulate a 2-dimensional growth of nitrides, e.g. the effective lattice mismatch along the [T100] direction of AlN and the [001] direction of Si(110) substrate is approximated to 0.7% and that along the [H12 0] of AlN and the [T10] direction of Si be about 19% [2]. A few research groups have reported that the crystalline quality of epitaxial layers and the device performance realized on Si(110) are similar to those obtained on Si(111) [4,5]. The biaxial property dependent on in-plane directions has a potential to realize more improved high electron mobility transistor. The epitaxial GaN thin film could be obtained by using molecular beam epitaxy (MBE) system that has a nitrogen source either a radio frequency (RF) plasma source or NH₃ gas source. Recently, the NH₃ source is widely used for the growth of nitrides due to its advantages, such as high growth rate and improved crystalline quality [4,6,7]. However, the study on the microstructural properties of nitride semiconductors grown by using ammonia source is insufficient to achieve a high quality thin film. Specifically, the microstructural evolution dependent on the anisotropic lattice mismatch between the nitride and the Si(110) substrate was not studied.

The main purpose of this study was to understand the microstructural evolution depending on the ammonia flux of GaN thin films grown on Si(110) substrates by molecular beam epitaxy method. Insight gained at the microscopic level regarding how GaN layers change their morphology and microstructure depending on the NH₃ flux is essential for the growth of high quality thin films for various applications. Therefore, a detailed study of the structural characteristics of GaN layers was carried out based on transmission electron microscope observation using various techniques.

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http://dx.doi.org/10.1016/j.cap.2014.12.017
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GaN thin films were grown on a Si(110) substrate by using Varian Gen-II MBE system which was able to use the nitrogen RF plasma source or ammonia source to supply nitrogen source. Preferentially, AlN seed layers were deposited under the same condition on the 3-inch Si substrate. The AlN layers were grown at 940°C under a 200 W RF power and a 2 sccm flow of N2. The AlN seed layer has an important role to reduce the cracks of GaN layer by different thermal expansion rate between Si and GaN [1,8]. For the growth of GaN layers, ammonia gas as a nitrogen precursor was injected through a home-made injector and the flux was varied from 100 to 400 sccm, those correspond with NH3 back ground pressure from 2×10⁻⁴ to 8×10⁻⁴ Torr. Gallium (Ga) source was evaporated in the effusion cell, and the Ga beam equivalent pressure was 2.2×10⁻⁶ Torr. The GaN was grown for 1 h at 800°C. The crystalline quality and orientation relationship were analyzed by high-resolution X-ray diffraction (HR-XRD) system. The surface morphology was investigated by using scanning electron microscopy (S-4800, Hitachi). Bright-field (BF) TEM images, selected-area electron diffraction (SAED) patterns, and high-resolution (HR) TEM micrographs were collected using FEI F30 microscopes operating at 300 kV. Specifically, the dislocation propagation in GaN thin films was carefully observed through two-beam analysis.

Fig. 1 shows SEM images of the GaN layers grown on Si(110) substrates. Although the surface morphologies of GaN layers grown under 100, 200, and 300 sccm as the ammonia flux show a mound shape in Fig. 1(a), (b), and (c), the size and the height of an individual mound are steadily decreased with the increase in the ammonia flux. In addition, the emerging and expanding phenomena of flat area at the top of the mounds, was indicated by dotted lines in Fig. 1(b) and (c), were detected through the careful observation. At the end, the surface roughness was dramatically improved when the ammonia flux was reached to 400 sccm. The growth rate of GaN layers was in inverse proportion to the ammonia flux as shown in Fig. 1(e). The surface morphology and the growth rate of films may be related to the growth mode and it must be affected by the ammonia flux. Actually, the RHEED pattern at the final stage of the growth was steadily changed to a streaky pattern from a spotty pattern as the ammonia flux was increased while the RHEED patterns at the initial growth stage showed a spotty pattern always, which means the change of growth mode from 3-dimensional (3D) island growth to 2-dimensional (2D) layer-by-layer growth (not shown in this article). The similar result with this phenomenon is reported in another paper [9]. The reduction of the gallium desorption rate by increasing the NH3 flux was reported by a few research groups [10,11]. However, the opposite phenomenon, an increase of Ga desorption and the
suppression of NH₃ decomposition by excess of hydrogen accumulated in the growth chamber by the decomposition, was also suggested. The reduction of growth rate can be caused by the increased Ga desorption, probably, related to the formation of hydrides, e.g. GaH, GaH₂, and GaH₃ [12]. Since, we have deduced that the growth of GaN layers is severely affected by the NH₃ flux. The nucleation of GaN was accelerated at the initial growth stage because the Ga desorption was suppressed by the high NH₃ flux before the chamber was filled with excess gas when the NH₃ flux was 400 sccm. And then, the Ga desorption was increased by forming hydrides with excess of hydrogen and the NH₃ decomposition was suppressed by the products of NH₃ decomposition, mainly nitrogen and hydrogen. In addition, the etching effect by the nucleation of GaN was accelerated at the initial growth stage for the half-maximum (FWHM) values of symmetric diffraction peak were 1024 arcsec and 2012 arcsec, respectively. The X-ray rocking curve for (1T 02) planes of GaN layers shows the same tendency as the result from (0002) planes (Fig. 2(b)). From the increases of FWHMs, we have deduced that the crystalline quality is degraded with the increase of the NH₃ flux.

From SEM and XRD analyses, we concluded that the crystalline quality was not improved although the surface roughness (or the flatness) was improved with the increase of the NH₃ flux. Since, we have characterized the microstructural properties of GaN layers by using various TEM techniques, such as 2-beam bright-field and high-resolution TEM micrographs, to study the origin of the degradation of crystalline quality. Also, we have tried to find out the clues related to the growth mode of GaN layers.

Fig. 3 shows the selected area electron diffraction patterns obtained from the GaN layer grown under the NH₃ flux of 100 sccm. From the analysis of SAED patterns, the orientation relationship between the wurtzite (WZ) structure of GaN and AlN and the Si(110) substrate was [1120]WZ//[0110]Si and (0002)WZ//(220)Si. The SAED patterns in Fig. 3(a) and (b) taken along the <1100>GaN, <001>Si, and <1100>Si zone axes, respectively, indeed show anisotropic biaxial strain behaviors depending on in-plane directions. In Fig. 3(a), since the (002) interplanar spacing of Si is a little smaller than the (1100) interplanar spacing of GaN, the diffraction spots from the Si are located at the longer distance from the transmitted electron beam along the in-plane direction (in this case, the [002] direction of Si and the [T100] directions of GaN) although it is not easy to differentiate the spots due to the small difference. On the other hand, the diffraction spots for the (220) planes of Si are much closer to the transmitted beam than those for the (1120) planes of GaN in Fig. 3(b) because the (220) interplanar spacing of Si is much larger than the (1100) interplanar spacing of GaN. In principle, the Si(110) substrate may sufficiently cause a tensile strain along the <1120> direction in a GaN thin film because the effective interplanar spacing of a Si(110) substrate is larger than those of nitrides. Our films are, however, not thin enough, i.e. they can be expected to be elastically relaxed, and recover the lattice constants of the bulk form. And, the SAED patterns in Fig. 3(a) and (b) are free from any other diffraction spots originated from tilted and/or twisted grains, which means that the GaN film is well conserving the orientation relationship with the Si substrate and has a single crystalline property. This observation is well consistent with the XRD analysis results.

The bright-field (BF) TEM images in Fig. 4(a)–(c) show cross-sectional views along the [TT20] direction of the GaN thin film grown under the NH₃ flux of 100 sccm. Fig. 4(a), taken under many-beam (MB) diffraction conditions, shows bright-and-dark contrasts in the thin film. These contrasts dissipate within the film in Fig. 4(a), except a few regions near a valley. Although the film in Fig. 4(a)

![Fig. 2. X-ray rocking curves of the GaN thin films grown on Si(110) substrates around (a) symmetric (0002) and (b) asymmetric (1T02) reflections. The FWHM (full-width at half-maximum) values of symmetric diffraction peak were 1024 arcsec and 2012 arcsec for the films grown under 100 and 400 sccm in the NH₃ flux, respectively, and those of asymmetric diffraction peak were 1234 arcsec and 2048 arcsec, respectively.](image1)

![Fig. 3. SAED patterns taken along (a) the [1T0] direction of Si and the [TT20] direction of GaN and (b) the [001] direction of Si and the [T100] direction of GaN as a zone axis in the GaN thin film grown under the NH₃ flux of 100 sccm.](image2)
shows a mounded-surface morphology, the density of penetrating dislocations, indicated by dotted rectangles, is relatively low in the thin film. BF TEM images under two-beam diffraction conditions were obtained to reveal the origin of the bright- and dark-contrasts. Fig. 4(b) corresponds to \( g = [0002] \) of the wurtzite structure of GaN, where the specimen has been tilted slightly from the \([TT20]\) zone axis around a tilting axis perpendicular to the growth surface. Fig. 4(c) was obtained for \( g = [\bar{T}100] \) of the wurtzite structure. Dislocations annihilating inside the film are visible in Fig. 4(b) near the interface between the GaN and the AlN, but not visible in Fig. 4(c). By applying the invisibility criterion, \( g \cdot b = 0 \), where \( g \) is a diffraction vector and \( b \) is a dislocation Burgers vector, it is likely that the dislocations in Fig. 4(b) have a Burgers vector vertical to the interface, most probably \( b = [0001] \) of the wurtzite structure, i.e. they are of pure screw type because the line direction \( \xi \) of a dislocation for \( c \)-oriented GaN is paralleled to the \([0001]\) growth direction. The positions of the dislocations are well consistent with each other in Fig. 4(a) and (b). Since, we could deduce that most of the dislocations annihilating inside the film are of screw type (s). Contreras et al. [13] reported that the screw dislocation could be annihilated by neighboring dislocations with opposite Burgers vectors to make dislocation loops. The threading dislocations reaching to the surface are of mixed type (m) because they are observed in Fig. 4(b) and (c). In addition, the threading dislocation at the valley, indicated by a dotted rectangle, are of mixed type and it may be generated by the coalescence of 3D islands.

The BF TEM images in Fig. 5 show cross-sectional views along the \([TT20]\) direction of GaN thin film grown under 400 sccm in the \( NH_3 \) flux. The types of dislocations were analyzed by using the previous invisibility criterion. Most of bright- and dark-contrasts in Fig. 5(a), are reaching to the surface and the density of contrasts was much higher than that in Fig. 4(a). In addition, unlike in Fig. 4(b), most of dislocations observed in Fig. 5(b), corresponding to \( g = [0002] \) of the wurtzite structure, are reaching to the surface and the density of dislocation is much higher than in Fig. 4(b). On the other hand, dislocations are rarely observed in Fig. 5(c) obtained from the condition of \( g = [\bar{T}100] \) of the wurtzite structure. By combining the threading dislocations observed in the GaN thin film grown under the \( NH_3 \) flux of 400 sccm are of pure screw and mixed types and the number of screw types is more than that of mixed types. And, when compared with Fig. 4, the density of dislocations is much higher in Fig. 5. To obtain an accurate determination of dislocation density, bright field plane view TEM images were taken on tilted \( 10^\circ \) to \( g = [1\bar{1}20] \) with the surface of GaN samples grown under \( NH_3 = 100 \) and 400 sccm. The total dislocation density for GaN sample was of \( 3.7 \times 10^{10}/cm^2 \) at \( NH_3 = 400 \) sccm, respectively.

Fig. 6 shows cross-sectional TEM images taken along the \([001]\) direction of Si and the \([T100]\) direction of GaN to study dislocation behaviors affected by the anisotropic lattice mismatch. In Fig. 6(a) and (d), taken under MB diffraction conditions for the GaN films grown under the \( NH_3 \) fluxes of 100 and 400 sccm, respectively, the density of dislocations in Fig. 6(d) is much higher than that in Fig. 6(a) and which is well consistent with the results observed in Figs. 4 and 5. However, Fig. 6(a) shows clearly more threading dislocations in the GaN thin film than in Fig. 4(a) although Figs. 4(a) and 6(a) were taken from the same sample grown under the \( NH_3 \) flux of 100 sccm. Other BF images in Fig. 6 were obtained under normal two-beam diffraction conditions to reveal the edge, screw, and mixed components of the dislocations. A peculiar characteristic of dislocation behaviors were observed in Fig. 6(b), (c), (e), and (f), which is the clear observation of edge-type threading dislocations in Fig. 6(c) and (f). This dislocation behavior must be related to the anisotropic misfit strain distribution as mentioned above, e.g. the large lattice mismatch between GaN and Si reaching to 16% along the \([11\bar{2}0]\) direction of GaN and the \([T10]\) direction of Si.

These dislocation behaviors may be related to the crystalline qualities of the GaN films. Nevertheless the flat surface of the GaN...
thin film grown under 400 sccm in the NH₃ flux, the large FWHM values of the rocking curves must be originated from the increased density of dislocations, while the good crystalline quality of the GaN thin film grown under the NH₃ flux of 100 sccm is related to the low density of dislocation despite the mounded surface morphology.

Fig. 7(a) and (b) show the representative cross-sectional high-resolution TEM micrographs taken near the surfaces of the GaN thin films grown under the NH₃ fluxes of 100 and 400 sccm, respectively. Various facets were observed on the surface of the GaN film grown under the NH₃ flux of 100 sccm (Fig. 7(a)), while the flat surface was observed at the GaN thin film grown under 400 sccm in the NH₃ flux (Fig. 7(b)). This observation results are well agreed with the increase tendency of flat areas depending on the NH₃ flux in SEM images (Fig. 1). The HR TEM micrographs reveal well-ordered atomic arrangements on the [0001] and [1T00] planes with the interplanar spacing of 0.518 nm and 0.276 nm, which means the complete recovery to the lattice constants of the bulk form of GaN. High index planes of the wurtzite structure observed in the GaN films grown under the low NH₃ fluxes was steadily transformed to the basal plane with the increase of the NH₃ flux.

4. Conclusions

In this study, we have reported the microstructural properties of GaN thin films grown on Si(110) substrates under various NH₃ fluxes. The crystalline quality of the GaN thin film was degraded with the increase of the NH₃ flux while the surface flatness of the films was improved as the NH₃ flux increases. Also, the reduction of growth rate was observed with increasing the NH₃ flux. As increasing of the NH₃ flux, the total dislocation density for GaN thin film was increased from $3.7 \times 10^{9}/\text{cm}^2$ (NH₃ = 100 sccm) and $1.4 \times 10^{10}/\text{cm}^2$ (NH₃ = 400 sccm). High index planes of the wurtzite structure observed in the GaN films grown under the low NH₃ fluxes was steadily transformed to the basal plane with the increase of the NH₃ flux.

Acknowledgments

This work is supported by Korea Research Council of Fundamental Science and Technology through a Basic Research Project managed by the Korea Research Institute of Standards. Work in part supported by “Nano-Material Technology Development Program through the National Research Foundation of Korea (grant number: 2011-0030233).”
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