Structure Analysis of GaN Thin Film with Inversion Domains by High Voltage Atomic Resolution Microscopy

Chihiro Iwamoto\(^1\), Xu-Qiang Shen\(^2\), Hajime Okumura\(^2\), Hirofumi Matsuhashi\(^2\) and Yuichi Ikuhara\(^1\)

\(^1\)Engineering Research Institute, School of Engineering, University of Tokyo, Tokyo 113-8656, Japan
\(^2\)National Institute of Advanced Industrial Science and Technology (AIST) Central 2, Tsukuba 305-8568, Japan

The atomic structures and surface morphologies of three types of GaN films were investigated by high voltage atomic resolution microscopy (HVARM). By HVARM, each atomic column of Ga and N could clearly be resolved and the polarity of the film and inversion domains could be directly determined. The GaN film was grown on a sapphire substrate by molecular beam epitaxy (MBE) after nitridation of the sapphire surface. Inversion domains (IDs) crossed the whole film to the surface and made small pyramids on the surface. The small pyramids had Ga-polarity and the rest had N-polarity. A GaN film with In exposure during film growth had an almost Ga-polarity flat surface. In exposure process reduced the density of inversion domains that have a N-polarity. While a GaN film grown on an AlN buffer layer was unipolar, with a Ga-polarity. HVARM observation revealed that the density of the IDs determine the qualities and the polarity of the film.

(Received January 23, 2002; Accepted April 12, 2002)

Keywords: gallium nitride (GaN), molecular beam epitaxy (MBE), high voltage electron microscopy, high resolution electron microscopy (HREM), inversion domain

1. Introduction

Due to its potential applications in optical and electronic devices, GaN films have been intensively studied.\(^1\)\(^-\)\(^3\) On a sapphire (0001) substrate, a GaN (0001) film grown epitaxially with wurtzite structure. The GaN film has a polar structure along the growth direction, then, Ga or N polar lattice is produced depending on the deposition process.\(^4\)\(^,\)\(^5\) In contrast to a film obtained by metalorganic chemical vapor deposition (MOCVD), the lattice orientation of a film grown by conventional molecular beam epitaxy (MBE) is assumed to be mainly N-polarity. The N-polarity GaN grown by conventional MBE has a poor-quality and much effort has been made to improve the quality of the film.\(^6\)\(^-\)\(^8\) Consequently, it was found that quality of the GaN films with Ga-polarity was dramatically improved compared to those of the GaN films with N-polarity.\(^5\)\(^,\)\(^9\) However, the reason for the quality improvement is not clear and nanostructure analysis is necessary.

Recent techniques of the lattice-polarity characterization, such as coaxial impact collision ion scattering spectra (CAICISS)\(^5\)\(^,\)\(^6\)\(^,\)\(^9\) reflection high-energy electron diffraction (RHEED)\(^10\)\(^,\)\(^11\) convergent beam electron diffraction (CBED)\(^12\)\(^-\)\(^15\) and chemical etching only give an average information about the lattice-polarity in the film. In contrast to these techniques, conventional high-resolution electron microscopy (HREM)\(^16\)\(^-\)\(^18\) was applied to investigate the nanostructure of the GaN film. However, the point-to-point resolution of a conventional electron microscope is usually 0.18–0.2 nm. In order to characterize the GaN, we should observe the sample from \{10\} axis of GaN and the spacial resolution of about 0.113 nm is needed to discriminate between Ga and N, and hence to determine the absolute polarity. Then, in previous experiments, in order to determine the probable atomic structure by conventional HREM, several models were considered for image simulations, and overall agreement between simulation images and experimental high-resolution images using through focus was sought.

In this paper, high voltage atomic resolution microscopy (HVARM) was applied to investigate the GaN film. A point-to-point resolution of the electron microscope is about 0.1 nm. HVARM, therefore, can distinguish Ga from N and determine the absolute polarity together with the nanostructure of the film.\(^19\) Three types of polarity controlled GaN films grown by MBE were investigated and compared.

2. Experimental Procedure

The GaN films were grown on sapphire (0001) substrates by radio-frequency plasma-assisted molecular beam epitaxy (rf-MBE). Three kinds of growth procedure were applied to control the lattice-polarity. One procedure was GaN growth on a nitrided sapphire. The second one was In exposure during GaN film growth. The content of In was just equal to doping level. The third one was the adoption of an AlN high-temperature buffer layer before the GaN growth. The growth temperature of AlN was at 973 K and the growth rate obtained was 0.5 \(\mu\)m/h. In the three kinds of growth procedure, the GaN growth temperature was fixed at 1023 K and the growth rate obtained was 0.6 \(\mu\)m/h. The N- and N\(_2\) flow rate was 5 sccm. The experimental process is described in detail elsewhere.\(^5\) The polarity of the samples was characterized directly by CAICISS in Ref. 5). The samples were sliced perpendicular to the interface, followed by mechanical polishing and dimpling down to 10 \(\mu\)m and ion thinning using an Ar\(^+\) beam under an accelerating voltage of 3 kV. The thinned sample was examined by HVARM (JEOL ARM-1250) at an accelerating voltage of 1250 kV. Spherical and chromatic aberration constants of the objective lens were designed to be \(C_s = 1.4\) mm and \(C_c = 2.4\) mm, respectively. The high-voltage stability was attained to be \(5 \times 10^{-7}\) per 5 seconds.\(^20\) The incident electron beam adjusted to be parallel to the \{10\} axis of GaN. Each atomic column, in the direction, consists of only Ga or N, and the smallest inter-column distance in this orientation is 0.113 nm.
3. Results

3.1 Atomic resolution observation of GaN film

First of all, an imaging condition of GaN was studied by HVARM. Figure 1 is a typical high-resolution image of GaN taken by HVARM. In order to compare to the experimental image, multislice and image simulations were performed using electron microscopy software developed by Ishizuka and Uyeda.\textsuperscript{21} The calculated image at Scherzer defocus and 3.2 nm in thickness and a schematic atomic structure model are inserted in the figure. The insertions suggest that Ga atomic columns are imaged as strong dark spots and N atomic columns are imaged as weak dark spots under these conditions. Experimental image in Fig. 1 is taken under almost the same defocus and thickness conditions as the calculation. In the experiment, focus series around the Scherzer condition were taken, and the thickness of the sample were checked by comparing the experimental image variation and the calculated image variation depending on the amount of the defocus. The experimental image is in good agreement with the calculated image and reveals that the discrimination between Ga and N is possible. Therefore, in this paper, absolute polarity of GaN was determined by the image taken under these conditions.

3.2 Atomic structure of the GaN film grown on a nitrided sapphire

Figure 2 is the bright field image of the GaN film grown on a nitridation sapphire. The polarity of the film assumed to be mainly N-polarity by CAICISS.\textsuperscript{5} In Fig. 2, high density of domains crosses over the GaN film. The surface also contains high density of pyramids that are about 7 nm in height and 7–15 nm in width. An arrow denotes one of the pyramids at the surface. Figure 3 shows a high-resolution image of the GaN surface with pyramids. Arrows denote domain boundaries and “D” denote the domains. The image suggests that the pyramids are formed at the surface of the nanometric domains. Figure 4 is the enlarged image of one of the domain boundaries in Fig. 3. The absolute polarity can be di-

Fig. 1 High-resolution electron micrograph of GaN taken by HVARM, showing that Ga and N can be resolved in the image. Insets are the simulated image obtained at $-35$ nm defocus and 3.2 nm thickness and a schematic of the atomic structure model.

Fig. 2 Low magnification image of the GaN film grown on the sapphire substrate. Many pyramids are formed on the surface. An arrow denotes one of the pyramids.

Fig. 3 High-resolution electron micrograph of the GaN surface with pyramids. Arrows denote domain boundaries and “D” denote the domains. The pyramids are formed at the surface of the nanometric domains.

Fig. 4 High-resolution electron micrograph of the domain boundaries. An arrow denotes a domain boundary and “D” denote the domain. Insets are the atomic structure models of GaN. The domain with pyramid has Ga-polarity and the rest has N-polarity.
rectly determined by the ARHVEM image, i.e., the nanometric domain with a pyramid has Ga-polarity and the rest has N-polarity. Thus the domain with the pyramid can be identified to be the inversion domain (ID) and the arrows in Fig. 3 and Fig. 4 are inversion domain boundaries (IDBs). Comparing with the image simulation calculated in Fig. 1, geometrical models for GaN in matrix and the ID are inserted as the insets in Fig. 4. In Fig. 4, Ga and N are distinguishable in most of areas, but in some regions they are indistinguishable due to the weak contrast of the image. However, the distinguishable images enable us to reconstruct the structure of the six membered ring of GaN and determine the absolute polarity even in the weak contrast area.

### 3.3 Atomic structure of the GaN film with In exposure during film growth

Figure 5 is the bright field image of a GaN film with In exposure during film growth. The polarity of the film assumed to be mainly Ga-polarity by CAICISS. Many domains are observed in the film and originate at the film/substrate interface. In contrast to the previous observations, density of the domains is reduced at the surface of the film. Although some grooves are observed, the pyramids that were formed at the surface of the nanometric domains are not observed and the surface is almost flat. Defects which directions are parallel or oblique to the film/substrate interface are also observed. In the film, two reduction mechanisms of the density reduction of the domains were observed: domains termination by planar defects and gradual reduction of domains. Figure 6 is the magnified image around the planar defects parallel to the film/substrate interface. The defects, which are denoted by arrows, terminate a lot of domains. Figure 7 is the magnified image at the surface of the film. The domains gradually reduce its diameter from the interface to the surface and several domains disappear inside the film. In this paper, we would like to focus on the mechanism of the gradual reduction of the domains. Figure 8 is the high-resolution image of the domain, which terminates near the surface of the film. At the surface, very large groove, which bottom is almost flat, is
formed. Figure 9 is the high-resolution image of the domain, which terminates at about 45 nm from the surface. Although small groove is still observed above the domain, the surface is almost flat. Figure 8 and Fig. 9 suggests that domains, which produce the rough surface by making groove at the surface, gradually buried during growth of the film and the surface become flat. Figure 10 is the high-resolution image of the domain boundary, which is denoted by solid lines. The image suggested that the domain has N-polarity and the rest has Ga-polarity. Thus the domain is ID like previous observation. In contrast, steps are observed on the IDB. The steps, about one Ga–N atomic pair in height, exist perpendicular to the growth direction. Consequently, the steps are considered to cause the gradual reduction of the ID.

3.4 Atomic structure of the GaN film grown on an AlN high-temperature buffer layer

Figure 11 is the bright field image of a GaN film grown on an AlN high-temperature buffer layer. The polarity of the film assumed to be mainly Ga-polarity by CAICISS.5) AlN buffer layer with 200 nm in thickness exists between sapphire and GaN. Around the interface between GaN and AlN or AlN and sapphire, high strain contrast is observed. This configuration is in good agreement with the previous observations.22,23) In the GaN film, only dislocations are observed and domains, which were observed in Fig. 2 or 5, are hardly observed. The surface of the GaN film is flat. Figure 12 is the high-resolution image of the interface between the AlN buffer layer and the substrate. Near the interface between the film and the substrate, many stacking faults and defects were observed. In addition to the wurtzite-type AlN, zincblend-type AlN was also
observed. However, domain boundaries, which are observed in previous observations, are hardly observed. Figure 11 and Fig. 12 suggests that the domain did not form from beginning of the deposition. Figure 13 is a high-resolution image of the film near the surface. The polarity of the film is determined to be Ga-polarity.

4. Discussion

The present results are directly compared to the previous results. The polarity of the poor-quality GaN film grown by conventional MBE was directly characterized by CAICISS and was assumed to be mainly N-polarity.\(^5\) In the present work, HVARM observations of the same sample directly revealed that the poor-quality derived from the high density of Ga-polarity IDs. It was considered that CAICISS gives average information about the lattice-polarity in the film, then, the polarity of nanometric IDs did not significantly contribute to the spectra of the film.

On the contrary, the GaN films grown using two different growth procedures were assumed to be Ga-polarity and had a improved quality.\(^5\) HVARM observations were in good agreement with these results. In addition, different growth modes were revealed. In the case of GaN with In exposure during the growth, GaN film also had N-polarity IDs. However, the density of IDs was reduced near the surface of the film. In contrast, GaN grown on an AlN buffer had no IDs and was unipolar film.

It was considered that the key factor for the improvement in quality was the control of the lattice polarity of the GaN films during the growth.\(^5\) Furthermore, HVARM observations of three types of the films indicated that reduction of the density of the N-polarity IDs improve the film quality. Consequently, the control of the density of the IDs during the growth determines the quality and the polarity of the film.

5. Conclusions

The morphology and the atomic structure of the GaN films, such as surface and ID morphologies and the polarity of the IDs, were directly determined from only one image taken using HVARM. Three types of the GaN films that have different growth procedure were investigated and the polarity and quality of the film were found to be determined by the density of the IDs. HVARM is thus powerful tool for the investigation of the atomic structure of the GaN films.

Acknowledgements

A part of this study was financially supported by Grant-in-Aid for Scientific Research.

REFERENCES