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Interface roughness of double buffer layer of GaN film grown on Si(1 1 1) substrate using GIXR analysis

Y. Yamamoto, N. Yamabe, T. Ohachi *

Department of Electrical Engineering, Doshisha University, 1-3 Miyakodani Tatara, Kyotanabe, Kyoto 610-0321, Japan

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ABSTRACT

A double buffer layer (DBL), interface reaction epitaxy (IRE) AlN/β-Si_3N_4/Si, grown by an IRE of β-Si_3N_4 and AlN films on Si, was fabricated to improve the crystalline quality of successively grown 30 nm GaN on a 30 nm AlN buffer layer using plasma-assisted molecular beam epitaxy (PA-MBE). The DBL was first prepared by surface nitridation of Si and successively prepared by IRE between the deposited Al and N atoms in β-Si_3N_4. Both the AlN buffer layer on the DBL and GaN film on the AlN buffer layer were grown by activity-modulation migration enhanced epitaxy (AM-MEE). Hetero epitaxial grown films of GaN(30 nm)/AlN buffer(30 nm)/DBL/Si(1 1 1) were prepared for analysis using a three layer model of grazing incidence-angle X-ray reflectivity (GIXR), which consisted of three layers of GaN, AlN buffer and Si and of the three interfaces of the GaN surface, GaN/AlN buffer and AlN buffer/DBL/Si. The nitridation temperature dependence of the interface roughness of the DBL was measured to be 0.5 and 0.6 nm, for nitridation temperatures of 780 and 830 °C, respectively. The full width at half maximum (FWHM) of rocking curve GaN(0 0 0 2) measured by X-ray diffraction (XRD) for nitridation temperatures of 780 and 830 °C were 58.2 and 55.2 arcmin, respectively.

1. Introduction

Group III nitride semiconductors are expected to be high-performance devices because of the wide band gap energy. Control of the dislocation and sharp hetero interface are indispensable to develop a high-performance device. Therefore, the measurement of the hetero-interface is very important because the quality of the hetero-interface determines the crystalline quality grown by plasma assisted molecular beam epitaxy (PA-MBE). Grazing incidence-angle X-ray reflectivity (GIXR) has been used to analyze the hetero-interface between GaN or AlN and the substrate [1–3]. Using the GIXR method, various parameters such as film thickness and interface roughness were able to be measured without destructive inspection. For example, Ref. [1] reported that the thicknesses of the intermixed layers at the AlN/Ni hetero-interfaces were measured by the GIXR method. Kobayashi et al. [3] used GIXR analysis to detect interfacial layer formation between GaN and ZnO at temperatures higher than 500 °C.

GaN/AlN/Si structure has been investigated for the III-nitrides grown on Si substrate by many groups because AlN, which had a strong Al–N bond, was easily grown on the Si substrate among the III-nitride materials [4–6]. A double buffer layer (DBL) of interface reaction epitaxy (IRE) AlN/β-Si_3N_4 between the Si substrate and GaN film was investigated for the growth of high quality GaN films with low dislocation [7,8]. The interface reaction between AlN and Si substrate was the key process for the growth of the DBL structure. The reaction between the Al and N atoms in β-Si_3N_4 caused AlN formation on β-Si_3N_4. However, the interface between AlN and β-Si_3N_4 was very difficult to analyze because the formed IRE-AlN is very thin. In addition, the relationship of two interfaces between the GaN/AlN buffer and AlN buffer/DBL/Si has not been investigated until now. The GIXR method can analyze these interface roughnesses simultaneously.

In this study, the interface roughness of the GaN surface, GaN/AlN buffer and AlN buffer/DBL/Si and the thickness of GaN and AlN were analyzed by GIXR using "X'pert Reflectivity" software by PANalytical, in order to grow high crystalline quality GaN films on Si. The surfaces of β-Si_3N_4 and GaN were measured using an atomic force microscope (AFM). The thickness of β-Si_3N_4 was measured by X-ray photoelectron spectroscopy (XPS). The interface growth process was studied by reflection high-energy electron diffraction (RHEED).

2. Experimental

2.1. Cleaning of Si surface

For nitridation of the Si surface, a very flat and clean Si(1 1 1) substrate surface is required. First of all, the Si substrate of 2 in. was rinsed by ultrasonic cleaning after a Si substrate was immersed in a semico-clean solution. Afterward, the Si substrate was oxidized in $HCl:H_2O_2:H_2O = 3:1:1$ at 70 °C for 1.5 min, and cleaned by using

* Corresponding author.
E-mail address: tohachi@mail.doshisha.ac.jp (T. Ohachi).

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running deionized water of 15 MΩ cm for over half an hour. SiO_2 on Si substrate as a sacrificial oxide film was removed by heating rapidly to 1 × 1 surface reconstruction temperature in a growth chamber of MBE and the contamination was not absorbed on the Si surface. It is necessary for the Si surface temperature to reach above that of the phase transition temperature of surface reconstruction 7 × 7 to 1 × 1 of 856 °C [9]. Before the Si temperature reached Si 1 × 1 the bare Si surface appeared by thermal decomposition of SiO_2; small protrusions of SiC were formed and appeared in the AFM image as white dots.

2.2. Growth of DBL, MEE-AlN and MEE-GaN

A DBL is prepared by surface nitridation of Si at first and successively prepared by IRE between the deposited Al and N atoms in β-Si_3N_4. The β-Si_3N_4 buffer layer on Si(1 1 1) was fabricated by adsorbed (ADS) nitrogen atoms at different nitridation substrate temperatures of 780 and 830 °C using IRE. Subsequently, an IRE-AlN template on β-Si_3N_4/Si(1 1 1) was grown by the pre-deposition of Al atoms on β-Si_3N_4. A 30 nm thick AlN buffer layer on the DBL and 30 nm thick GaN on the AlN buffer layer were grown by activity modulation migration enhanced epitaxy (AM-MEE) for PA-MBE [10,11]. AM-MEE uses periodical exposure of nitrogen atoms produced by the high brightness (HB) mode and excited nitrogen molecules $N_2{}^*$ produced by the low brightness (LB) mode. The periodical exposure was performed using an IRFS-501 radio frequency inductively coupled plasma (rf-ICP) cell made by Arios Inc. AM-MEE is an advanced MEE method, which enhanced the migration of Ga or Al atoms by the exposure of the $N_2{}^*$ flux to Ga or Al atoms. The advantage of AM-MEE is that the kinetic energy transfer from $N_2{}^*$ to Ga or Al atoms lowers the growth temperature [10]. The time sequence of AM-MEE is shown in Fig. 1. The HB and LB modes were maintained by changing the input rf power and/or inner pressure of the rf-ICP cell.

Fig. 2 shows a three layer structure model for GIXR analysis. The growth conditions of β-Si_3N_4 at different substrate temperatures are shown in Table 1. T_{sub}, T_{Al} and T_{Ga} are the substrate temperature, K-cell temperature of Al and K-cell temperature of Ga, respectively. Only the nitridation substrate temperatures are different—780 and 830 °C.

Afterward, to optimize the growth condition of GaN, the growth substrate temperature of GaN was altered from 620 to 520 °C. The nitridation temperature was settled at 780 °C. The growth conditions of GaN at different substrate temperatures are shown in Table 2. The time sequence of AM-MEE is the same as in Fig. 1.

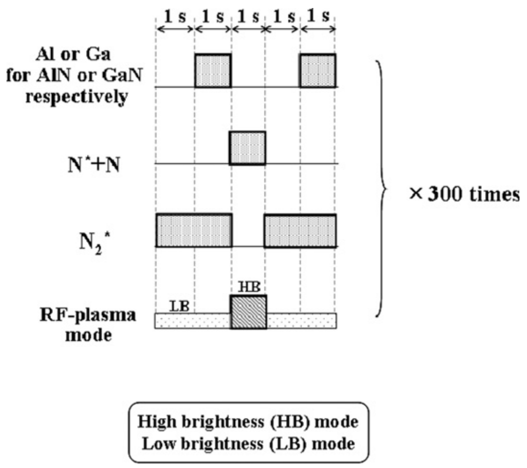

Fig. 1. Time sequences of AM-MEE growth for AlN and GaN.

3. Results and discussion

3.1. Nitridation of Si at different temperatures

The nitridation of Si(1 1 1) for 3 min was performed at 680, 780 and 830 °C. The thickness of β-Si_3N_4 was measured by the ratio of

Table 1
Growth conditions of β-Si_3N_4 at different substrate temperatures.

	T_{sub} = 780 °C		T_{sub} = 830 °C	
Nitridation times (min)		6		β-Si_3N_4
Pressure N_2 (Pa)		75.6		
T_{sub} (°C)	780		830	
Input power (W)		500		
Irradiation Al (s)		8		IRE-AlN
T_{Al} (°C)		1110		
T_{sub} (°C)		830		
T_{Al} (°C)		1165		MEE-AlN
T_{sub} (°C)		830		
Pressure N_2 (Pa)		75.6		
Input power (W)		450		
T_{Ga} (°C)		1040		MEE-GaN
T_{sub} (°C)		620		
Pressure N_2 (Pa)		75.6		
Input power (W)		400		

Table 2
Growth conditions of GaN at different substrate temperatures.

	T_{sub} = 520 °C		T_{sub} = 620 °C	
Nitridation times (min)		6		β-Si_3N_4
Pressure N_2 (Pa)		75.6		
T_{sub} (°C)		780		
Input power (W)		500		
Irradiation Al (s)		8		IRE-AlN
T_{Al} (°C)		1110		
T_{sub} (°C)		830		
T_{Al} (°C)		1165		MEE-AlN
T_{sub} (°C)		830		
Pressure N_2 (Pa)		75.6		
Input power (W)		450		
T_{Ga} (°C)		1040		MEE-GaN
T_{sub} (°C)	520		620	
Pressure N_2 (Pa)		75.6		
Input power (W)		400		

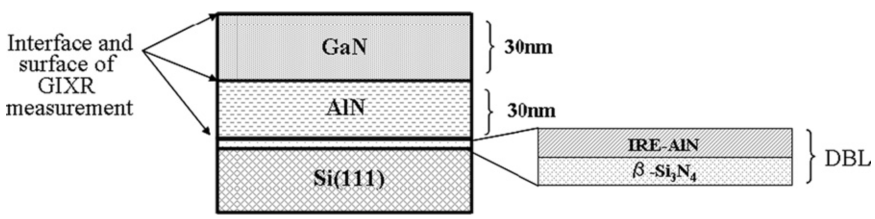

Fig. 2. Three-layer structure of GaN grown on AlN buffer/DBL structure on Si(1 1 1) substrate for GIXR analysis. DBL consisted of IRE-AlN and β-Si_3N_4.

the Si 2p and N 1s (I_{Si2p}/I_{N1s}) signals of XPS measurement from Ref. [12]. The XPS result showed that the thickness of β-Si_3N_4 was 0.35, 0.27 and 0.18 nm for 680, 780 and 830 °C, respectively, as shown in Fig. 3. This thickness difference is caused by the large adsorption of nitrogen atoms to the Si surface at lower temperature.

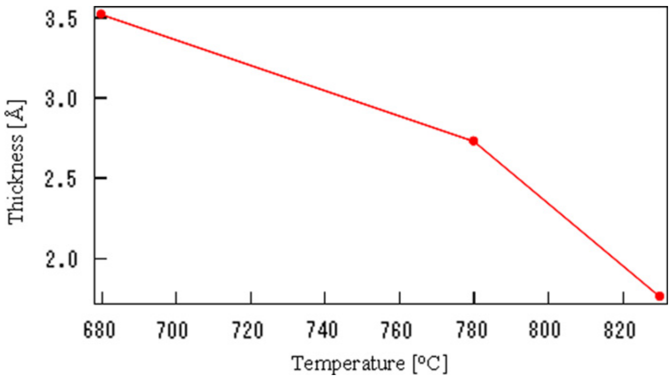

Fig. 3. Thickness of β-Si_3N_4 at different substrate temperatures measured by XPS.

3.2. GIXR analysis of DBL

Fig. 4 shows the results of the GIXR patterns measured three layers structure and the fitting curve with the analyzed parameters simultaneously. The interface of GaN/AlN buffer and AlN buffer/

Table 3
Results of interface roughness measured by GIXR and RMS values of surface measured by AFM at nitridation temperatures of 780 and 830 °C.

Layer description	T_{sub} = 780 °C		T_{sub} = 830 °C	
	Roughness (nm)	RMS (nm)	Roughness (nm)	RMS (nm)
GaN	3	2.9	3.7	3.8
GaN/AlN buffer	0.8		1.1	
AlN buffer/DBL/Si	0.5		0.6	

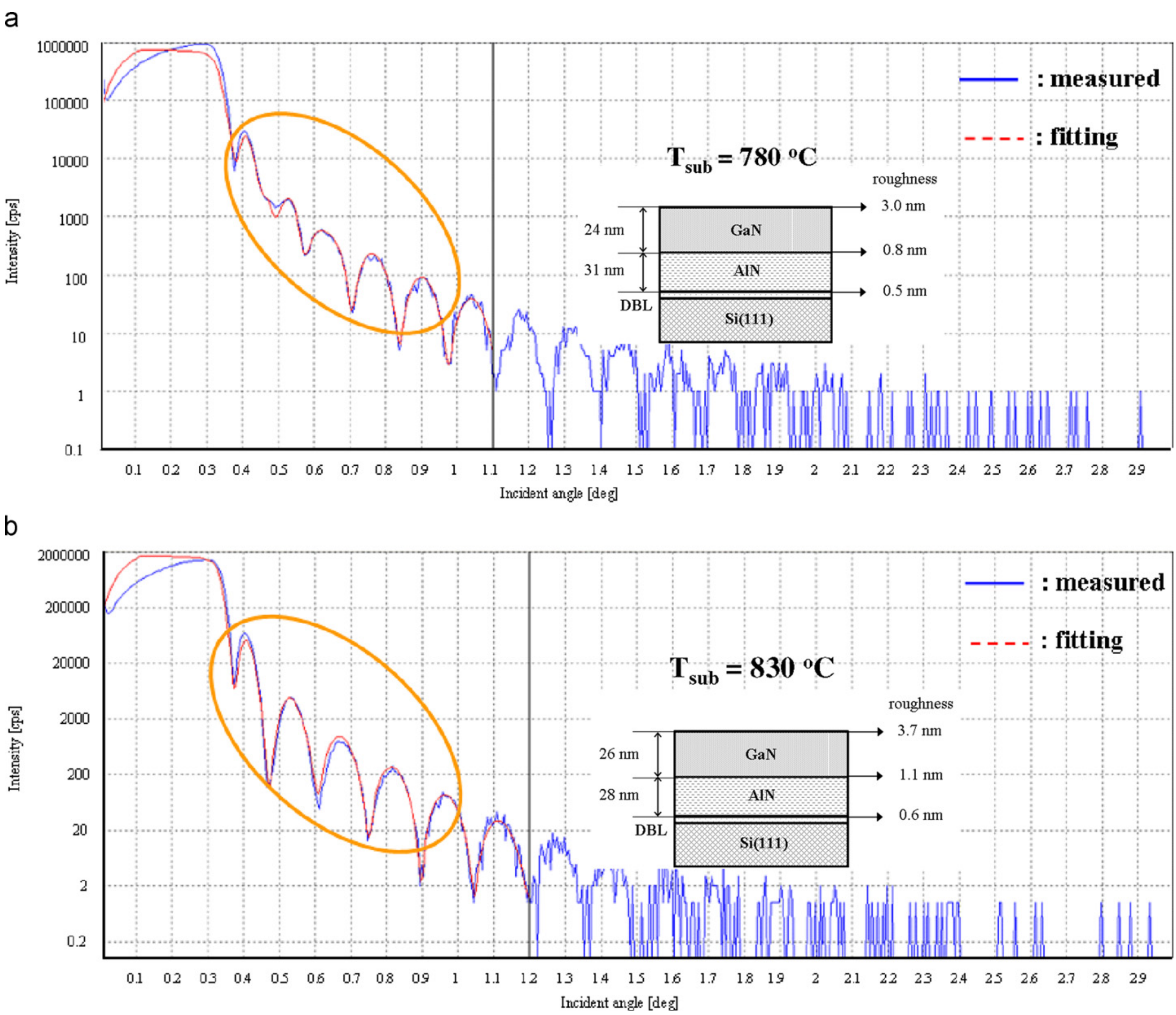

Fig. 4. GIXR data and fitting curve at nitridation temperatures of (a) 780 and (b) 830 °C.

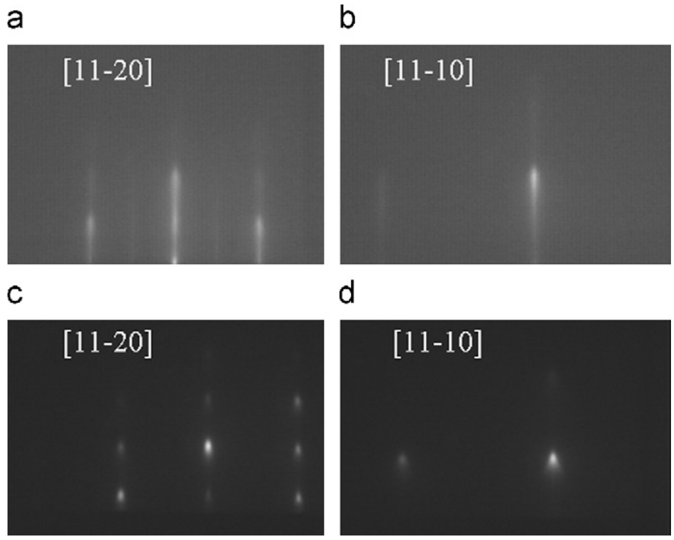

Fig. 5. RHEED patterns taken from [1 1 −2 0] and [1 0 −1 0]: (a and b) 30 nm AlN at growth temperature of 830 °C; (c and d) 30 nm GaN at nitridation temperature of 780 °C.

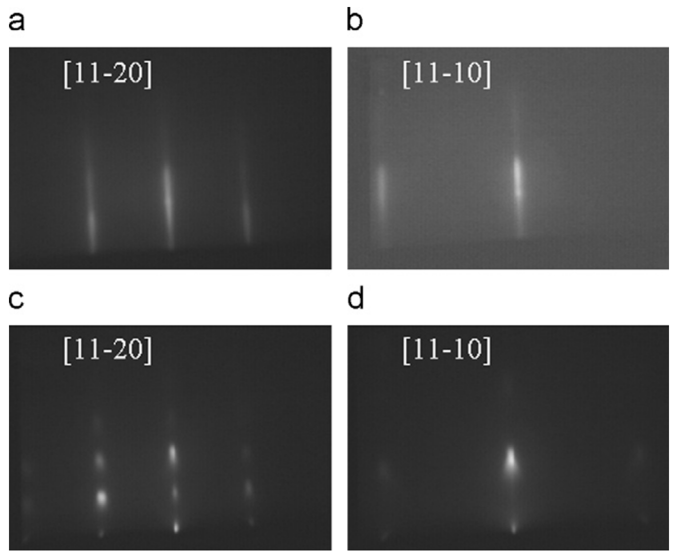

Fig. 6. RHEED patterns taken from [1 1 −2 0] and [1 0 −1 0]: (a and b) 30 nm AlN at growth temperature of 830 °C; (c and d) 30 nm GaN at nitridation temperature of 830 °C.

DBL/Si was able to be measured by GIXR method. Interface roughness of AlN buffer/DBL/Si was 0.5 and 0.6 nm and that of GaN/AlN buffer was 0.8 and 1.1 nm; surface roughness of GaN was 3.0 and 3.7 nm measured by GIXR at nitridation temperatures of 780 and 830 °C, respectively. From these results, both the interfaces of AlN buffer/DBL/Si and GaN/AlN buffer were sharper at the nitridation temperature of 780 °C. RMS values of GaN measured by AFM was 2.9 nm at the nitridation substrate temperature of 780 °C, whereas RMS values of GaN measured by AFM was 3.8 nm at the nitridation substrate temperature of 830 °C. Small RMS of β-Si_3N_4 layer realized the flat surface of DBL and GaN. These results of interface roughness of GaN/AlN buffer and AlN buffer/DBL/Si, surface roughness of GaN film measured by GIXR and surface roughness of β-Si_3N_4 and GaN film measured by AFM were shown in Table 3 and Fig. 4. Within the ellipsoidal circles in Fig. 4, the differences in fitting curves above the critical angle are shown.

ω-FWHM of rocking curve GaN(0 0 0 2) measured by XRD was 58.2 and 55.2 arcmin at 780 and 830 °C, respectively. From these results, the crystalline quality of GaN at the nitridation substrate temperature of 830 °C was better than that of GaN at the nitridation substrate temperature of 780 °C. These results indicated that the thin β-Si_3N_4 layer formed by high temperature nitridation was better for

Table 4
Results of interface roughness measured by GIXR and RMS values of surface measured by AFM at GaN growth temperatures of 520 °C

Layer description	T_{sub}=520 °C	
	Roughness (nm)	RMS (nm)
GaN	1.0	1.2
GaN/AlN buffer	0.8	
AlN buffer/DBL/Si	0.6	

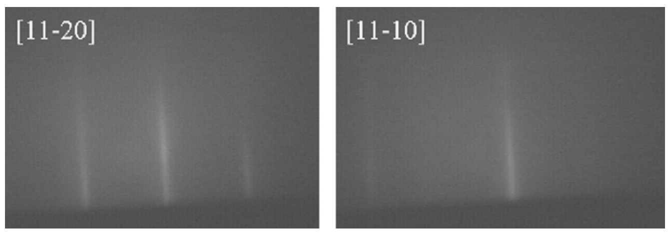

Fig. 8. RHEED patterns taken from [1 1 −2 0] and [1 1 −1 0] for the GaN growth temperature of 520 °C.

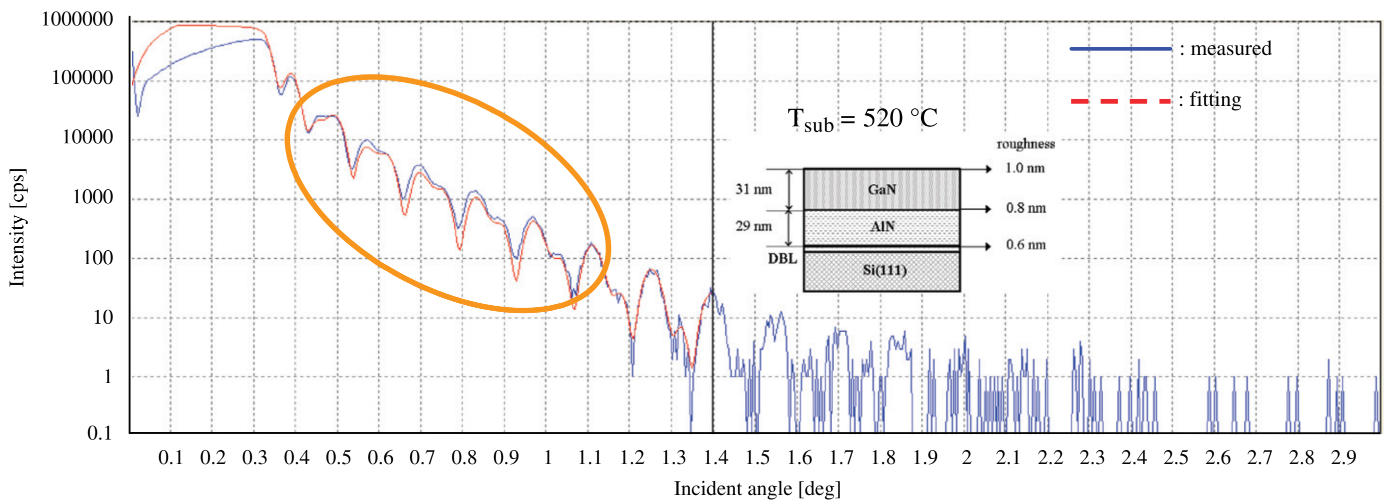

Fig. 7. GIXR data and fitting curve for GaN growth temperature of 520 °C.

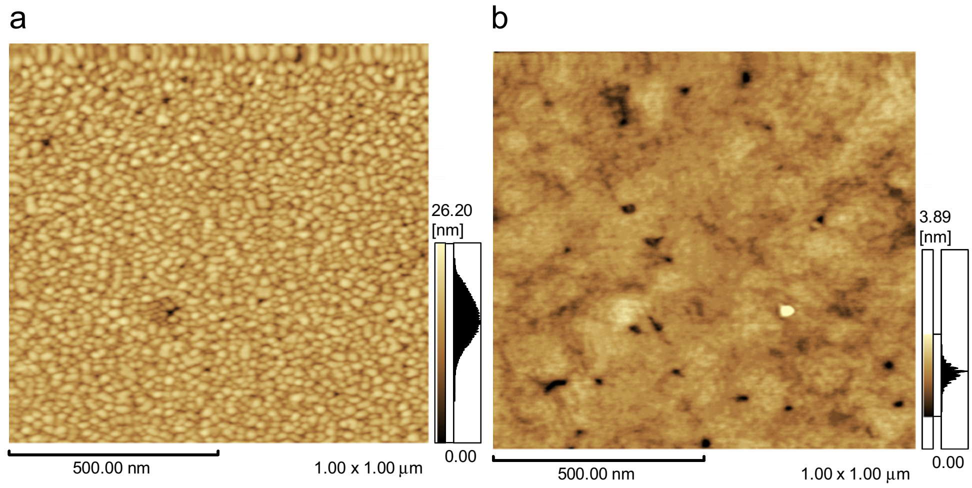

Fig. 9. AFM images of GaN surface at GaN growth temperatures of (a) 620 and (b) 520 °C with nitridation temperature of 780 °C.

GaN quality because the Si or N atom diffused through β-Si_3N_4 and thick β-Si_3N_4 layers, which have long diffusion path distorted crystallinity of β-Si_3N_4. RHEED patterns from GaN and AlN buffer on DBL were shown in Figs. 5 and 6 for 780 and 830 °C, respectively. The RHEED patterns showed a flat surface of AlN due to streak patterns in the AlN buffer as shown in Fig. 5(a and b) and Fig. 6(a and b).

To achieve a smooth surface of GaN layer, the GaN growth condition was optimized by changing the growth temperature. When the growth temperature was altered from 620 to 520 °C, the RMS value of AFM improved from 2.9 to 1.2. Fig. 7 shows the GIXR pattern of GaN growth. The results of interface roughness of GaN/AlN buffer and AlN buffer/DBL/Si, surface roughness of GaN film measured by GIXR and surface roughness of the GaN film measured by AFM were summarized in Table 4. These results showed that the flat surface was achieved. RHEED patterns of 30 nm GaN were streaks as shown in Fig. 8(a and b) and the AlN buffer layer was used as the same layer as shown in Fig. 5(a and b). The surface of GaN was also confirmed by AFM and a smooth morphology of the surface was achieved as shown in Fig. 9. These results showed the improvement of the surface roughness of GaN because Ga evaporation and GaN decomposition by substrate heating was suppressed by low temperature growth of GaN and 2D growth was promoted.

4. Conclusion

The nitridation temperature dependence of the interface roughness of the DBL was measured to be 0.5 and 0.6 nm, at nitridation temperatures of 780 and 830 °C, respectively, using a three layer model of GIXR analysis. The crystalline quality of an epitaxial film of GaN grown on AlN buffer on the DBL was improved by the interface roughness of the DBL. ω-FWHM of rocking curve GaN(0 0 0 2) measured by XRD at nitridation temperatures of 780 and 830 °C were 58.2 and 55.2 arcmin, respectively. GIXR is a helpful tool to characterize the interface of DBL after the growth of AlN or GaN films. Further improvement of DBL changing the nitridation condition is expected to improve the crystalline quality of GaN films on Si.

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