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# Plasma-assisted molecular beam epitaxy growth of crack-free AlN cap layer on GaN-based heterostructures

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We have succeeded in growing crack-free AlN cap layer on GaN with AlN buffer layer on Si (111) substrates by plasma-assisted molecular beam epitaxy. *In situ* reflection high-energy electron diffraction (RHEED) observation was performed to monitor the growth processes. A two-dimensional growth process lead to AlN cap layer of good crystal quality. High-resolution x-ray diffraction (HR-XRD), high-resolution transmission electron microscopy (TEM) and energy dispersive x-ray spectroscopy (EDS) line analysis were used to investigate the structural properties of the films. It was revealed that AlN is single crystalline with low defect.

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*Keywords*: AlN, GaN, Molecular beam epitaxy

## 1. Introduction

AlN has the widest direct band gap of 6.2 eV among the III-nitride compounds, offers considerable potential for applications in deep-ultraviolet (UV), light-emitting diodes (LEDs), laser diodes, and high-temperature electronic devices [1-3]. AlN is useful as a buffer layer for GaN epitaxial growth on Si substrate because of its difficulty to grow GaN film directly on Si due to large mismatches in lattice constant between these materials [4]. Due to its unique property of high thermal conductivity (140-180 vs 1.4 W/mK for $SiO_2$), AlN is expected to be excellent substitutional material for conventional amorphous $SiO_2$ insulation layer in GaN-based devices to improve the device performance and reliability and to reduce self-heating effect [5-7].

Recently, pulsed-laser deposition (PLD), metal-organic vapour phase epitaxy (MOVPE), metal-organic chemical vapour deposition (MOCVD), and molecular beam epitaxy (MBE) have been used to grow high-quality AlN thin films. Compared to PLD, MOVPE and MOCVD, MBE has several advantages. These include significantly low growth temperature (which limits interdiffusion and phase segregation), accurate control film thickness in the order of atomic scale, *in situ* observation using reflection high energy electron diffraction (RHEED) and the use of high purity source materials. However, there are few reports concerning the epitaxial growth of undoped AlN layer on GaN substrates. Moreover, there are serious problems to be overcome for electronic applications because high-quality of AlN has been difficult to grow and cracks are easily generated in AlN grown on GaN [8]. In that case, control of strain in nitrides is important in terms of improvement of crystalline quality, namely, the In-doping technique [9,10], and a combinatory use of $N_2$ and $H_2$ carrier gases [11].

In this work, we report the growth of AlN cap layer on GaN with AlN buffer layer on Si (111) substrates by plasma-assisted molecular beam epitaxy (PAMBE) with reflection high-energy electron diffraction (RHEED) as the *in situ* analysis for growth monitoring. The structural quality of the AlN cap layer was characterized by using high-resolution x-ray diffraction (HR-XRD). The microscopic structure of the films was studied by high-resolution transmission electron microscopy (HRTEM) and the composition of the films was analyzed by using energy dispersive x-ray spectroscopy (EDS) line analysis.

## 2. Experimental

The growth of AlN/GaN with AlN buffer layer is carried out by RF-plasma assisted molecular beam epitaxy (Veeco Gen II MBE system). A standard effusion cell for aluminum and gallium, and RF gas plasma source system to supply activated nitrogen were used. The plasma was operated at typical nitrogen pressure of 1.5 x $10^{-5}$ Torr under discharge power of 300W. Si (111), 3 inch in diameter were used as the growth substrates. The substrate was cleaned by using RCA method prior to loading into MBE system. The Si substrate was outgassed in the load-lock and buffer chambers prior to growth. In the growth chamber, Si substrate was heated at $900^0$C, and a few monolayers of Ga were deposited on the substrate for the purpose of removing the $SiO_2$ by the formation of $GaO_2$. The 7 x 7 silicon surface structure was monitored by reflection high-energy electron diffraction (RHEED) and a clean Si (111) was obtained when RHEED showed a typical Si (111) 7 x 7 surface reconstruction pattern with the presence of prominent Kikuchi lines.

Before the growth of AlN buffer layer, a few monolayers of Al are deposited on silicon to avoid the formation of $Si_xN_y$ layer which is deleterious for the growth of the subsequent epilayers. Then, AlN buffer or wetting layer was first grown by opening both Al (cell temperature at $1120^0$C and flux of 2.2 x $10^{-7}$ Torr) and N cell shutters and starting the N plasma simultaneously for 15 minutes. The role of this layer is to improve the crystalline quality of the latter grown GaN layer and also to electrically insulate the epitaxial film from the conductive substrate. Then, GaN epilayer was grown on top of buffer layer at cell temperature of $1085^0$C and flux of 7.9 x $10^{-7}$ Torr for 1 hour. Subsequently, thin AlN cap layer was grown on top of GaN surface at cell temperature of $1120^0$C and flux of 2.2 x $10^{-7}$ Torr for 10 minutes. The growth rate of our MBE system is nearly 0.48 μm/h and the thickness were obtained by controlling the growth time. High-resolution X-ray diffraction (HR-XRD) was carried out on a PANalytical X'pert Pro MRD with a Cu-Kα1 as a radiation source to evaluate the crystalline quality of the films. Transmission electron microscopy (TEM) and energy dispersive x-ray spectroscopy (EDS) line analysis were used to characterize the microstructure of AlN/GaN films.

## 3. Results and discussion

Fig. 1 shows the evolution of RHEED pattern during the growth process. As shown in Fig. 1 (a), the Si substrate surface showed a clear 7 x 7 surface reconstruction at high temperature. Fig. 1 (b) shows the RHEED pattern becomes streaky and Kikuchi lines disappear when a few monolayers of Ga were deposited on the Si. After closing the Ga shutter, a typical Si (111) 7 x 7 surface reconstruction pattern then appeared with the presence of prominent Kikuchi lines and the RHEED intensity increased, as seen in Fig. 1 (c). It verifies desorption of native oxide and indicates a clean Si (111) surface. Fig. 1 (d) shows the growth of AlN buffer layer. It can be seen that the RHEED pattern displayed a sharp and clear streaky pattern, showing that the AlN buffer layer is in a two-dimensional manner and indicating good surface morphology. This is a good base for the GaN growth. During the growth of GaN, the streaky RHEED pattern in Fig. 1 (e) is sharpened as compared to Fig. 1 (d), suggesting the improvement of the crystalline quality of GaN layer relative to the AlN buffer layer. The absence of spotty RHEED patterns throughout the entire growth indicates that the surface remains flat without any prominent cluster formation. Fig. 1 (f) shows the RHEED pattern for AlN cap layer at the end of the deposition. The bright and sharp streaky lines indicate that the AlN cap layer is single crystalline with a smooth surface. In this way, we monitor and control the growth according to the *in situ* RHEED patterns.

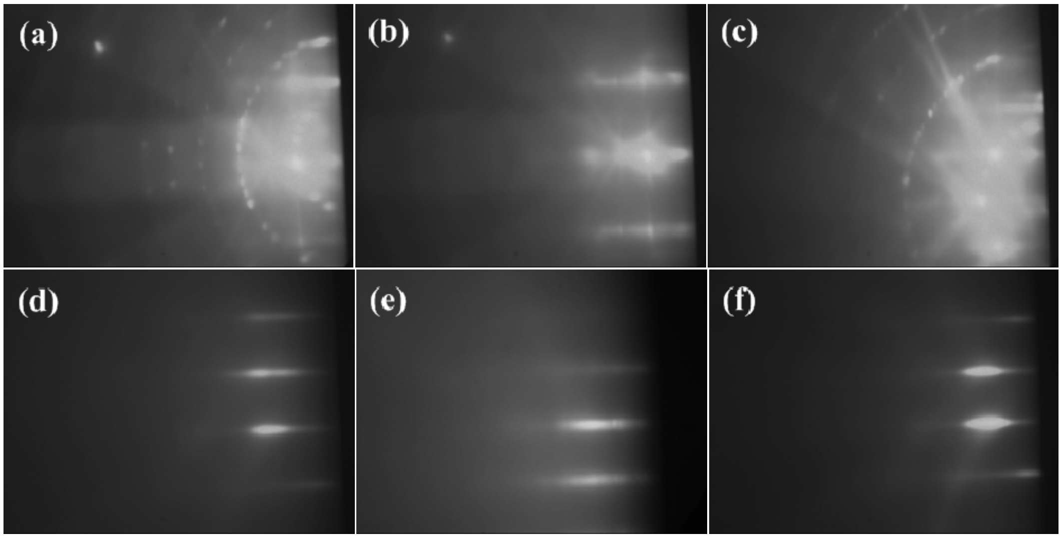
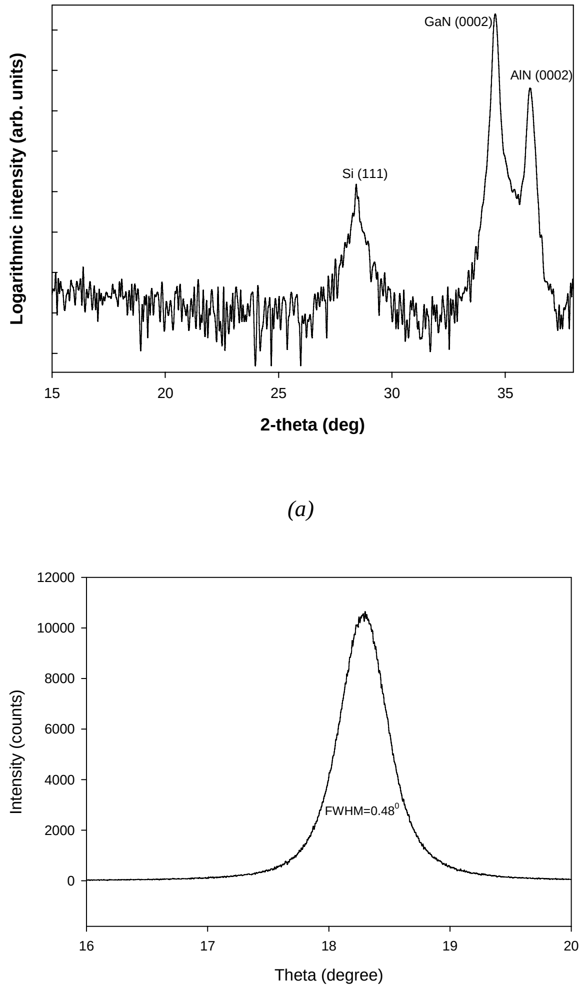


*Fig. 1. RHEED patterns for (a) a clear 7 × 7 surface reconstructions of Si substrate at high temperature; (b) after deposition of a few monolayers of Ga on Si; (c) after closing the Ga shutter; (d) growth of AlN buffer layer; (e) growth of GaN; (f) growth of AlN cap layer.*

In Fig. 2 (a), the XRD spectra with scan range from $15.0^0$ to $38.0^0$ shows the heterostructure of AlN/GaN/AlN were epitaxially grown on Si (111) substrate. Diffraction peaks can be observed at $28.45^0$, $34.56^0$, and $36.10^0$ which correspond to Si (111), GaN (0002) and AlN (0002), respectively. This result suggests that the AlN films are c-axis oriented. Also, it is seen that the sample has wurtzite hexagonal AlN and GaN structure. The XRD spectrum does not reveal any sign of cubic phase of AlN and GaN, so it is confirmed that our samples possessed hexagonal structure. From Fig. 2 (b), the full-width at half-maximum (FWHM) of AlN (0002) peak is $0.48^0$ (29 arcmin) from the rocking curve measurement. This result is similar to or slightly better than other reports [12, 13]. Generally, the result of on-axis XRD curve, like (0002) FWHM, shows the information related to the tilt of the subgrains with respect to the substrate and only the treading dislocations with a screw component make a contribution [14].

Therefore, the narrower (0002) FWHM of AlN contributes to the lower defects of threading dislocations with a screw component.



*(a)*

*(b)*

*Fig. 2 (a). XRD spectra of AlN/GaN/AlN/Si heterostructures; (b) FWHM of AlN (0002) rocking curve.*

In order to explore the defect nature, it is necessary to investigate by TEM observation. Cross-sectional TEM of the deposited film is presented in Fig. 3. It shows that the AlN cap layer grow directly and uniformly on GaN, with slightly distinctive column and threading dislocations. As can be seen in Fig. 2, no crack line can be found throughout the observed area. Hence, this results indicate that the AlN cap layer grown on GaN have good crystalline quality.

To understand the microstructure properties of the interface layers, dark field (DF) images were recorded (Fig. 4). Two kind of threading dislocations are observed: most of them have a nearly vertical line but a few of them are inclined. Also, very few dislocation loops are observed as a consequence of the two-dimensional (2D) layer-by-layer growth mode.

The high-resolution TEM (HRTEM) cross-sectional image in Fig. 5 reveals an atomically sharp and smooth interface between the AlN cap layer and GaN layers and interface between AlN cap layer and GaN is indicated by the white arrow line. Like the heteroepilayers typically grown by MBE and MOCVD, no amorphous phase was observed at the interface. Also the AlN cap layer is a single crystalline and this observation is corroborated by streaky RHEED pattern as well.

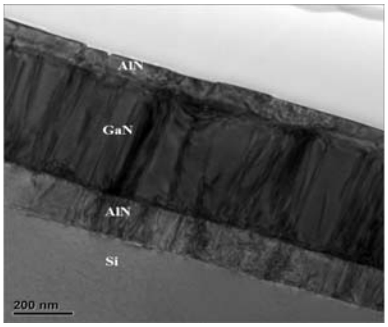


*Fig. 3. Cross-sectional TEM image of AlN/GaN/AlN/Si heterostructures.*

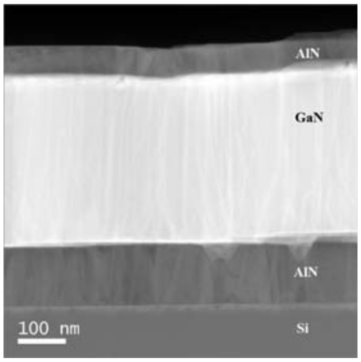


*Fig. 4. Dark field cross-section TEM image of AlN/GaN/AlN/Si heterostructures.*

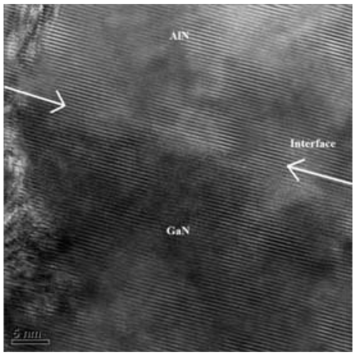


*Fig. 5. Cross-sectional HRTEM image of the AlN/GaN interface.*

In addition, the elemental composition of AlN and GaN were evaluated by the EDS analysis, which was installed on the TEM system and the result is presented in Fig. 6. Therefore, we found that the Al composition of region A and region C are about 62% and 52%, respectively whereas the Ga composition of region B is about 90%. Moreover, the composition of N in region A, region B and region C are about 35%, 10% and 44%, respectively. This result confirmed that all the layers were successfully grown on Si substrates.

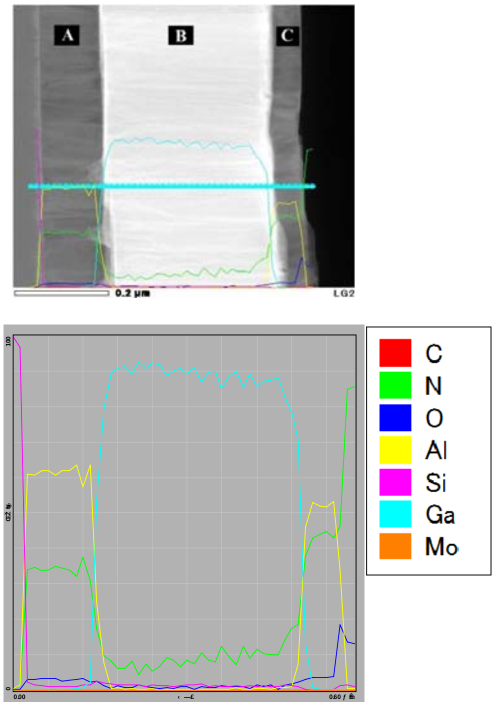


*Fig. 6. EDS line analysis atomic percentage of the AlN/GaN/AlN grown on Si substrate.*

## 4. Conclusions

The characteristics and microstructure of AlN cap layer on GaN heterostructures grown by plasma assisted molecular beam epitaxy on Si (111) substrates have been investigated. Epitaxial growth of AlN cap layer on GaN was obtained. No cracks were observed in AlN as revealed from TEM measurement. AlN is single crystalline with low defect as confirmed by RHEED patterns and HRTEM measurement.

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