



## Embedded AlN/GaN multi-layer for enhanced crystal quality and surface morphology of semi-polar (11-22) GaN on m-plane sapphire



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### ABSTRACT

We demonstrate high quality semi-polar (11-22) gallium nitride thin film grown on m-plane sapphire substrate with the insertion of AlN/GaN multi-layer via MOCVD. The influence of three different number of multi-layers AlN/GaN pairs on the crystal quality and surface morphology of semi-polar (11-22) gallium nitride thin film is investigated. The surface morphology analysis strongly suggests that increasing the number of AlN/GaN pairs from 20 to 60 suppresses the arrowhead-like and undulated features. The increase of AlN/GaN pairs also enhanced the surface quality, with the root mean square roughness improving from 16.24 nm to 6.08 nm. The abruptness of the interface between the AlN/GaN pairs was seen to improve significantly upon reaching the 40th pair where a continuous thin layer was clearly observed for each pair. The crystal quality was also observed to be enhanced at higher number of AlN/GaN pairs, where the on- and off-axis X-ray rocking curve showed significant reduction in the full width at half maximum of at least ~10% and 20%. Finally, x-ray reciprocal space mapping analysis further confirms the enhancement of the crystal quality as the diffuse scattering streak was suppressed, which may indicate a significant reduction of the defect density.

### 1. Introduction

III-nitride light emitting diode (LEDs) have been employed in a wide spectral range from the ultraviolet (UV) to blue and green wavelengths [1–3]. The most popular material for LEDs, gallium nitride (GaN), are typically grown along c-direction. However, the structure of the device along c-direction suffers from polarization effects, which leads to band bending which induces quantum-confined stark effect (QCSE) [4]. This is undesirable as it results in a considerable reduction in the recombination efficiency of GaN-based LEDs [5]. In order to circumvent this issue, several research groups have proposed the GaN growth along the non-polar and semi-polar directions [6–8]. For LEDs with longer wavelengths, growth along the semi-polar direction is preferable as non-polar growth has been shown to exhibit lower indium incorporation, thus tending to produce LEDs with shorter wavelength emission [9,10]. However, it is still challenging to grow non-polar and semi-polar with a very high crystal quality due to the defect densities such as threading dislocation (TD) and basal stacking faults (BSFs) [11–13]. Semi-polar GaN thin film with enhanced crystal quality has been achieved on bulk GaN substrates, but suffers from high production costs and complex production processes [5,14]. Two main alternatives for growth of semi-polar thin films have been employed, namely, the

epitaxial lateral overgrowth (ELOG) technique on patterned substrates and common growth of semi-polar (11-22) epitaxial layer on non-pattern m-plane sapphire. The former technique requires additional steps and regrowth process (*ex-situ*), while the latter suffers rough surface and poor crystal quality resulting from the anisotropic crystallographic mismatch [15–18].

Recent studies, however, have shown that the use of multi-layer (ML), embedded prior to the unintentionally doped (Utd-GaN) epitaxial layer, could enhance the crystal quality for semi-polar epitaxial layers, reducing defect densities such as TDs and BSFs. This includes the use of AlN/GaN and AlN/AlGaIn ML resulting in crystal quality improvement [5,19]. Furthermore, AlN/GaN ML can be employed to investigate the state of the strain in the GaN thin film [20]. Here, we demonstrate the use of AlN/GaN ML to enhance the crystal quality and surface morphology of semi-polar (11-22) epitaxial layers. The compressive stress, the crystal quality and surface morphology will be investigated via room temperature Raman spectroscopy, high resolution X-ray diffraction (HR-XRD), field emission scanning microscopy (FESEM) and atomic force microscopy (AFM).

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## 2. Experimental procedure

The semi-polar (11-22) epitaxial layers were grown on m-plane (10-10)-oriented sapphire substrates via metal organic chemical vapor deposition (MOCVD) (SR-2000, Taiyo Nippon Sanso, Japan). Trimethylaluminum (TMA), Trimethylgallium (TMG) and ammonia ( $\text{NH}_3$ ) were used as the reactant source materials for aluminum (Al), gallium (Ga) and nitrogen (N), respectively. The hydrogen ( $\text{H}_2$ ) gas was utilized as the gas carrier for the whole process along the epitaxial growth. The substrate was firstly baked inside the reactor under  $\text{H}_2$  ambience to remove any contamination on the sapphire substrate surface. After the nitridation process was carried out on the m-plane (10-10) sapphire substrate, an AlN nucleation layer (total thickness  $\sim 80\text{--}100$  nm) was first deposited onto the sapphire substrate. Subsequently, high temperature AlN/GaN ML with 20, 40 and 60 pairs were grown at  $1050^\circ\text{C}$ , followed by a  $4.5\ \mu\text{m}$  thickness of Uid-GaN. The growth condition of semi-polar (11-22) AlN nucleation layer and Uid-GaN grown on top of the ML is reported in [18]. The semi-polar (11-22) GaN epitaxial layers were then grown on the m-plane sapphire substrates with 20, 40 and 60 pairs, denoted as M1, M2 and M3, respectively. The semi-polar (11-22) GaN epitaxial layers were next characterized by SU8200 field emission electron spectroscopy (FESEM), AFM5000II atomic force microscopy (AFM), Rigaku HR-XRD, including  $2\theta$ - $\omega$  scans, X-ray rocking curve (XRC) of on- and off-axis measurement, reciprocal space mapping (RSM) and room temperature Raman spectroscopy.

## 3. Results and discussion

Fig. 1 shows the HR-XRD  $2\theta$ - $\omega$  scans to elucidate the crystal orientation for semi-polar GaN epitaxial layers grown with 20, 40 and 60 pairs of AlN/GaN ML, designated as M1, M2 and M3, respectively. As it can be clearly seen from M3, the diffracted fringes resulted from the

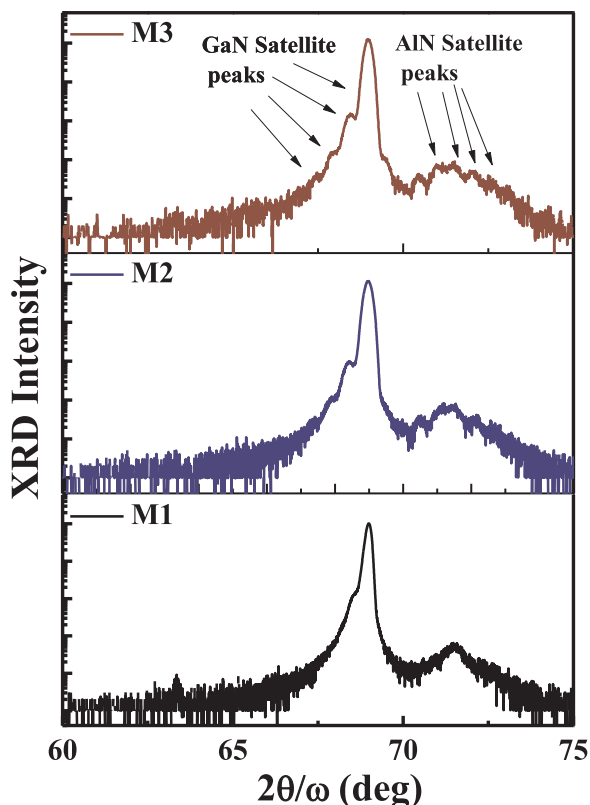


Fig. 1. HR-XRD  $2\theta$ - $\omega$  scan of semi-polar (11-22) GaN grown with 20, 40 and 60 pairs of AlN/GaN ML denoted as M1, M2 and M3, respectively.

Table 1

RMS roughness and peak to valley values of the semi-polar GaN epitaxial layers.

Sample	M1	M2	M3
Number of Pairs	20	40	60
RMS Roughness (nm)	16.24	6.08	6.08
Peak to Valley (nm)	92.10	38.24	40.24

interface and abruptness of semi-polar (11-22) AlN and GaN 60 pairs. However, M2 shows a fewer number of fringes due to less abruptness occurred in the ML. In contrast, the diffracted fringes were almost non-existent as 20 pairs of AlN/GaN ML were used. The diffracted fringes are ascribed to the satellite peaks of semi-polar (11-22) 60 pairs of AlN/GaN ML. Hence, the use of 60 pairs of AlN/GaN ML was crucial in order to attain a structure with an abrupt interface.

FESEM and AFM measurements were employed to characterize the surface morphology of the semi-polar GaN epitaxial layers as shown in Fig. 2(a)-(f). Table 1 lists the classification of as-grown epitaxial thin films with their respective root mean square (RMS) roughness as well as the peak-to-valley.

As seen from the FESEM images of Fig. 2(a)-(c), M1 suffers from undulated features. These features were seen to be progressively reduced as the number of AlN/GaN pairs was increased first to 40 pairs (M2), and then to 60 pairs (M3). Further investigation via AFM over a  $10 \times 10\ \mu\text{m}$  scan in Fig. 2(d) strongly suggests that an arrowhead-like feature was present in M1. Similar to previous reports on semi-polar [21,22], the arrowhead-like features are the result of the anisotropy of the adatom surface diffusion length along the [1-100] and [-1-123]. These features, which might be correlated with high density of BSFs [5], were almost suppressed in M2 and M3 owing to the enhancement of the crystal quality, as depicted in Fig. 2(e) & (f) [1]. In addition, surface enhancement was observed for M2 and M3, which is consistent with the elimination of the arrowhead-like features whereby the RMS roughness was drastically reduced from 16.24 nm to 6.08 nm.

Analysis of the FESEM cross-section of 60 pairs (M3) is presented in Fig. 3(a), (c) & (d). Fig. 3(a) & (b) show the cross-sectional image of semi-polar (11-22) epitaxial layers of M3 and its schematic diagram, respectively. As it can be seen, the  $4.5\ \mu\text{m}$ -thick of semi-polar (11-22) GaN above the ML of AlN/GaN 60 pairs exhibit a 2-dimensional morphology. The cross-sectional image of AlN/GaN 60 pairs displayed in Fig. 3(c) has a non-abrupt structure from the ML base until the 20th pair (highlighted in orange circle) and continued until the 40th pair (highlighted in green circle). However, the structure of AlN/GaN after the 40th pair exhibits an abrupt structure, which is the result of the crystal quality enhancement shown in Fig. 3(d). The improvement of surface quality with the increase of AlN/GaN pairs is thought to be related to a reduction of defects and dislocations resulting from the mismatch between the sapphire substrate and the epitaxial layer [22]. This result is in good agreement with the  $2\theta$ - $\omega$  scan as the structure of 60 pairs of AlN/GaN ML showed the most abrupt interface between the AlN and GaN. When a stack of two materials with different lattice constant are coherently grown, strong strain is produced in between the two materials. The defects and dislocations might be prevented from propagating upwards if they are bent by the strain and a close-loop is generated [23]. This, however, requires that the AlN/GaN ML achieve a threshold crystal quality. Therefore, the interface of the AlN/GaN ML must be abrupt for this condition to be attained.

To investigate the anisotropic properties of the semi-polar (11-22) GaN, on-axis X-ray rocking curve (XRC) was measured by HR-XRD. Fig. 4(a) shows full width at half maximum (FWHM) values of the on-axis (11-22) reflection over  $360^\circ$  for M1, M2 and M3 as a function of the azimuthal angle  $\Phi$  (incident beam parallel to [-1-123] when  $\Phi$  is  $0^\circ$  and parallel to [1-100] when  $\Phi$  is  $90^\circ$ ). As observed in Fig. 4(a), when the incident X-ray beam parallel to [-1-123], the FWHM values of M1, M2 and M3 were  $0.15^\circ$ ,  $0.14^\circ$  and  $0.13^\circ$ , respectively. However, when the

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