

# Investigation on HT-AlN Nucleation Layers and AlGaN Epifilms Inserting LT-AlN Nucleation Layer on C-Plane Sapphire Substrate

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In this study, we have investigated a high-temperature AlN nucleation layer and AlGaN epilayers on c-plane sapphire substrate by low-pressure metal-organic chemical vapor deposition (LP-MOCVD). High resolution X-ray diffraction (HRXRD), atomic force microscopy (AFM), scanning electron microscope (SEM) and Raman scattering measurements have been exploited to study the crystal quality, surface morphology, and residual strain of the HT-AlN nucleation layer. These analyses reveal that the insertion of an LT-AlN nucleation layer can improve the crystal quality, smooth the surface morphology of the HT-AlN nucleation layer and further reduce the threading dislocation density of AlGaN epifilms. The mechanism of inserting an LT-AlN nucleation layer to enhance the optical properties of HT-AlN nucleation layer and AlGaN epifilm are discussed from the viewpoint of driving force of reaction in this paper.

*Keywords* : Metal-organic chemical vapor deposition, Crystal quality, Surface morphology, LT-AlN nucleation layer

*OCIS codes* : (130.0250) Optoelectronics; (160.4670) Optical materials; (160.6000) Semiconductor materials; (240.0310) Thin films

## I. INTRODUCTION

III-nitride compounds have attracted considerable interest in the last decade due to their wide application in solid-state-lighting devices, which are used widely in the blue/green and ultraviolet spectral regions with higher quantum efficiency [1-3]. AlGaN alloys are important for optoelectronic devices such as light-emitting diodes (LEDs), laser diodes (LDs) and photo-detectors in the UV spectral region between the wavelength range from 200 nm to 365 nm, which can be tuned according to Al content in the AlGaN alloy system [4]. Owing to difficulty in fabricating bulk GaN and AlN, nearly all AlGaN films are heteroepitaxially grown on lattice-mismatched substrates, such as sapphire and SiC substrates, which results in threading dislocations, and residual strains occur greatly in AlGaN epifilms. For reducing the threading dislocation density, a thin AlN nucleation layer or GaN buffer layer is utilized to improve the crystal quality [5-7]. D H Wang *et al.* had indicated that the insertion of a low-temperature (LT) AlN nucleation layer effectively improves the thick AlGaN crystal quality, reduces surface roughness and eliminates the threading dislocation density through

inserting a LT-AlN nucleation layer between the high-temperature (HT) AlN nucleation layer and the sapphire [8, 9]. However, few studies have investigated the crystalline quality, surface morphology, and residual strain of HT-AlN nucleation layers. In this paper, we study further the influence on HT-AlN nucleation layers and AlGaN epifilms inserting LT-AlN nucleation layer on c-plane sapphire substrates from the viewpoint of driving force of reaction.

## II. EXPERIMENTAL PROCEDURE

Growth of an HT-AlN nucleation layer was achieved using a cold-wall showerhead low-pressure metal-organic chemical vapor deposition (LP-MOCVD) system. Hydrogen was used as carrier gas, triethylaluminum (TEAl), triethylgallium (TEGa) and ammonia ( $\text{NH}_3$ ) were used as Al, Ga and N sources, respectively. The pressure of the LP-MOCVD growth system was about  $5.2 \times 10^{-3}$  Torr, and the calculated ratio of V/III ( $\text{NH}_3/\text{TEAl}$ ) is 7058. As shown in Fig. 1, a 210-nm-thick HT-AlN nucleation layer was first deposited on (0001) sapphire substrate (2 inch) at  $1100^\circ\text{C}$  denoted as

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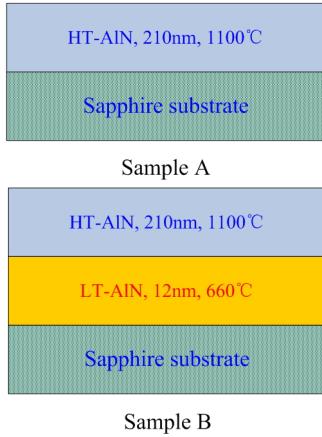


FIG. 1. Cross-section structures of Sample A and Sample B.

Sample A. Sample B has the same structure except that a 12-nm-thick LT-AlN nucleation layer was grown at 660°C between the sapphire substrate (2 inch) and a 210-nm-thick HT-AlN nucleation layer. In order to study and analyze the influence of the LT-AlN nucleation layer on AlGaN epifilms, we have grown an AlGaN with 1200-nm-thickness on Sample A and Sample B at 1030°C. The as-grown samples were characterized by high resolution X-ray diffraction (HRXRD), atomic force microscopy (AFM), scanning electron microscope (SEM) and Raman scattering. For the purpose to reveal and obtain a comprehensive knowledge of the threading dislocation density of AlGaN epifilms grown on the two AlN nucleation layers, we measured X-ray rocking curves (XRCs) for both symmetry and asymmetry diffraction planes by HRXRD. The HRXRD was performed using Bruker D8-discover system equipped with Ge (220) monochromator and channel-cut analyzer, delivering a pure Cu-K $\alpha$  line of wavelength  $\lambda=0.15406$  nm; The atomic force microscopy (AFM) was performed using an Agilent 5500 scanning probe system and the micro-Raman measurements were carried out in backscattering geometry with the Raman spectrometer Jobin Yvon LabRam HR800. An argon laser of 514-nm wavelength was used as an excitation light source and a 50 $\times$  objective was used to focus the incident laser light of a power of 14.2 mW on the sample, the spectrometer was calibrated using single-crystal silicon as a reference.

### III. RESULTS AND DISCUSSION

#### 3.1. High-resolution X-ray Diffraction

XRD-2 $\theta$  scan profiles are shown in Fig. 2(a). The peak located at 36.36° is diffraction from the AlN (002) planes of Sample A and Sample B, respectively; 75.58° and 76.70° are diffraction from the AlN (004) planes of the two samples, respectively. 42.04° is diffraction from the sapphire (006) planes. The intensity of AlN (002) planes for Sample B is much sharper than that of Sample A, indicating

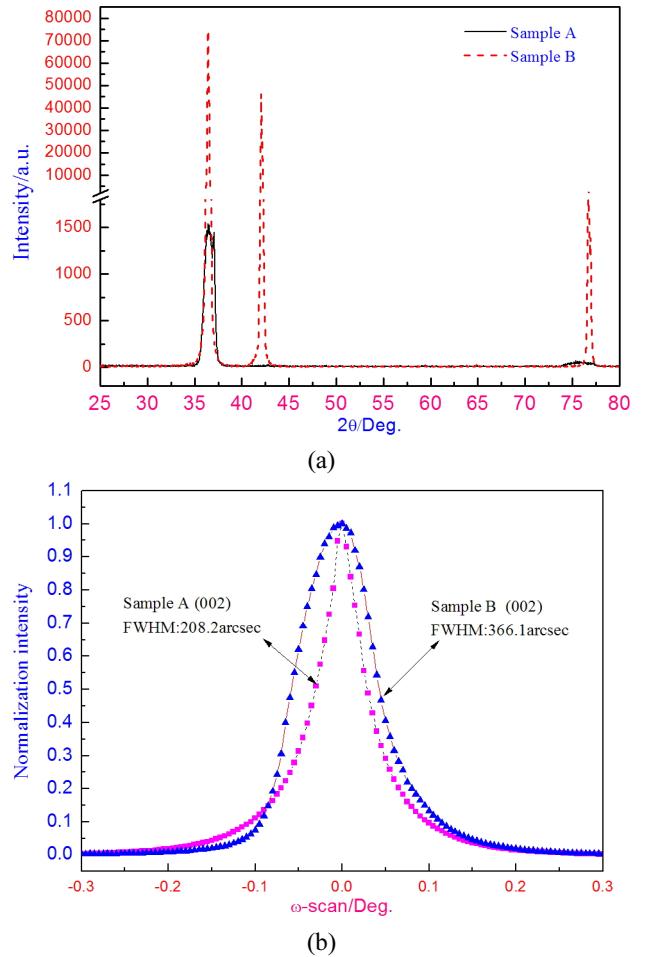


FIG. 2. XRD measurement profiles for Sample A and Sample B.

better crystal quality for the insertion of the LT-AlN nucleation layer between the HT-AlN nucleation layer and sapphire substrates. Furthermore, in order to study the crystalline quality of the two samples, the full-widths at half-maximum (FWHMs) of X-ray rocking curves were also measured as shown in Fig. 2(b). For (002) planes, the FWHMs of Sample A and Sample B are 366.1 and 208.2 arcsec, respectively, while for (102) planes, the respective FWHMs are 703.8 and 698.4 arcsec, which are nearly the same. It is well known that the FWHMs of XRD rocking curve from (002) planes or (102) planes is related to screw or edge dislocation density [10], so we conclude that, by inserting LT-AlN nucleation layer, the screw dislocation density and the total number of threading dislocations of HT-AlN can be reduced.

#### 3.2. AFM, SEM and Surface Morphology

Figure 3 shows the 10×10  $\mu\text{m}^2$  AFM images of the two HT-AlN nucleation layers. We can see the differences in surface morphology between Sample A and Sample B. The micrograph in Fig. 3(a) and (b) indicates an improvement of quality of HT-AlN nucleation layers. These V-shaped pits are similar to those observed in c-plane GaN epifilm.

It was found that the number of V-shaped pit defects on the surface of Sample B is less than on Sample A when a thin film LT-AlN nucleation layer is grown on sapphire substrates. Figs. 3(c) and (d) show the 3D surface morphology of the two HT-AlN nucleation layer samples. It is seen that Sample B shows clearly a lower quantity of dislocations on the surface than Sample A does. In addition, the root mean square (RMS) value is 4.09 nm for Sample A and 0.47 nm for Sample B, which indicates that Sample B has a smoother surface morphology than Sample A does. Furthermore, we have carried out SEM analysis in Figs. 3(e) and (f). As can be shown, there appear irregular convex disks on the surface of Sample A and regular convex disks on the surface of Sample B. From a quantitative point of view, the number of irregular convex disks for Sample A is more than Sample B has. We have marked the size of convex disks, and found the size of the convex disks on Sample A is a little smaller than those on Sample B. Consequently, be that as it may, the higher crystal quality of HT-AlN nucleation layers can be obtained by insertion of the LT-AlN nucleation layer between the sapphire substrate and the HT-AlN nucleation layer.

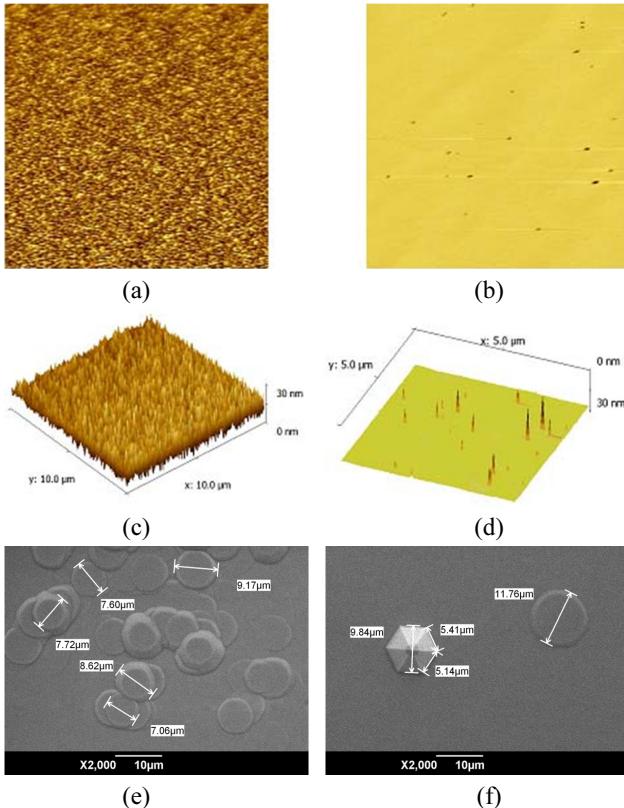


FIG. 3. AFM and SEM images of  $10 \times 10 \mu\text{m}^2$  surface morphologies for Sample A and Sample B. (a) Surface morphology of sample A, (b) Surface morphology of sample B, (c) 3D Surface morphology of sample A (RMS=4.09 nm), (d) 3D Surface morphology of sample B (RMS=0.47 nm), (e) SEM image of sample A, (f) SEM image of sample B.

### 3.3. Raman Scattering

Raman scattering was performed to examine residual strain of the two AlN nucleation layer samples recorded in the z(xx)-z backscattering configuration with 514-nm wavelength at room temperature. As is well known, the lattice and thermal mismatch between the substrate and the epifilm, characteristic for the heteroepitaxial growth of nitrides on a foreign substrate, results in the presence of strain under a linear elasticity biaxial system [11, 12]. Phonon frequency shifts are often employed as a tool for strain assessment in mismatched semiconductor heterostructures. The Raman scattering spectra of Sample A and Sample B are shown in Fig. 4 for a comparison. As can be shown in Fig. 4, the Raman peaks located at  $651.0 \text{ cm}^{-1}$  and  $645.90 \text{ cm}^{-1}$  are  $E_2$  (high) phonon modes for Sample A and Sample B, respectively. It was well known that the red-shift of AlN  $E_2$  (high) phonon mode corresponds to tensile strain in the HT-AlN nucleation layer, which is caused by differences in the thermal expansion coefficient and lattice mismatch between the AlN epilayer and sapphire substrate [5, 9]. The theoretical value of  $E_2$  (high) for AlN is  $657.4 \text{ cm}^{-1}$  of stress-free [13, 14], so there existed red-shift of AlN  $E_2$  (high) mode of  $6.4 \text{ cm}^{-1}$  and  $11.5 \text{ cm}^{-1}$  relative to the theoretical value of  $657.4 \text{ cm}^{-1}$  for Sample A and Sample B, respectively, which implies that Sample A and Sample B are under tensile strain, and the tensile strain existing in Sample B is larger than that in Sample A. As we know, the strain along the growth direction is found to be tensile as elasticity theory predicts in the case of biaxial stress [15-18]. By comparison to the number of V-shaped pits, more tensile strain can be relieved in the condition of more V-shaped pits on the surface of Sample B. Furthermore, it is widely accepted that the larger tensile strain existing in-plane is apt to produce high crystalline quality epifilm. Thus, the conclusion can be drawn that inserting the LT-AlN nucleation layer can improve crystal quality of the HT-AlN nucleation layer.

In order to study further the influence on the crystal

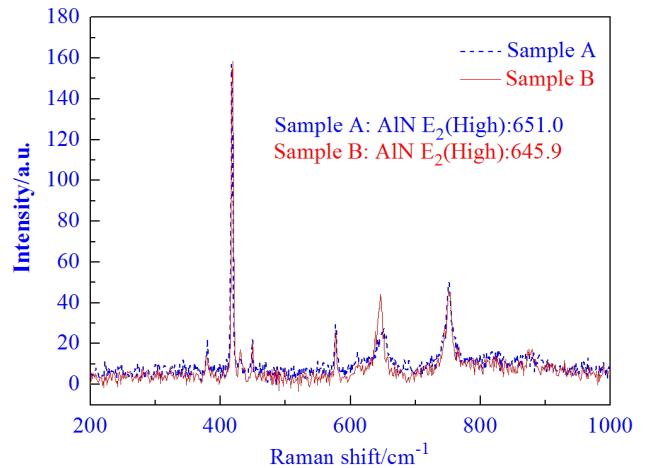


FIG. 4. Raman scattering recorded at room temperature for Sample A and Sample B.

quality for AlGaN epifilms by inserting the LT-AlN nucleation layer, we grew an AlGaN with 1200-nm-thickness on Sample A and Sample B at 1030°C, as denoted as Sample A' and Sample B' respectively, as shown in Fig. 5. In Ref.[9], we have carried out the symmetrical (002) and asymmetrical (102) XRD- $\omega$  scan rocking curve of the two AlGaN epilayers, and the calculated results of threading dislocation densities existed in AlGaN epifilms on Sample A and Sample B are shown as Table 1.

Ref. [19] studied the size of AlN island under different growth temperature modes, as can be seen from Fig. 6, the size of AlN islands grown under low-temperature mode is smaller than the size of those grown under intermediate temperature and high temperature [19], which coincides

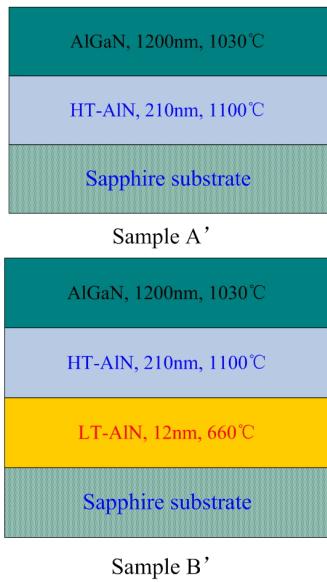


FIG. 5. Cross-section structures of Sample A' and Sample B'.

with AFM images results.

It was well known that the relationship between saturated vapor pressure and radius of grain in a nucleation layer is shown as the Kelvin formula in Eq. (1):

$$RT \ln \frac{P}{P_0} = \frac{2\gamma M}{\rho r} \quad (1)$$

Here,  $\rho$  is the vapor density,  $M$  is molar mass,  $R$  is gas constant,  $\gamma$  is the surface tension,  $T$  is Kelvin temperature,  $P_0$  is the vapor pressure on the plane,  $P$  is the vapor pressure for the radius  $r$  of the grain. We can see that smaller island in the nucleation layer can obtain larger vapor pressure at low temperature, which provides the driving force advantageous for depositing the AlN nucleation. In additional, Ref. [20-22] believed that larger tensile strain in AlN nucleation layers is beneficial to obtain higher crystal quality AlGaN epifilms. So, we have a reason to believe that higher crystal quality AlN nucleation layer eases obtaining higher crystal quality AlGaN epifilms. Therefore, a conclusion is reached that AlGaN epilayer with higher crystal quality and lower threading dislocation density can be obtained using insertion of the LT-AlN nucleation layer between sapphire and the HT-AlN nucleation layer.

#### IV. CONCLUSION

In this paper, we have focused on the influence on AlN nucleation layer and AlGaN layer caused by inserting a LT-AlN nucleation layer between the c-plane sapphire substrate and HT-AlN nucleation layers. HRXRD, AFM, SEM and Raman scattering have been employed to characterize crystal quality, surface morphology and residual strain of

TABLE 1. FWHMs and the calculated threading dislocation densities of AlGaN epifilms on Sample A' and Sample B'

Sample	FWHMs of rocking curve/arcsec		$\rho_s / 10^8 / \text{cm}^2$	$\rho_e / 10^{10} / \text{cm}^2$
	(002)	(102)		
A'	227	1800	1.042	1.754
B'	134	1447	0.18	1.134

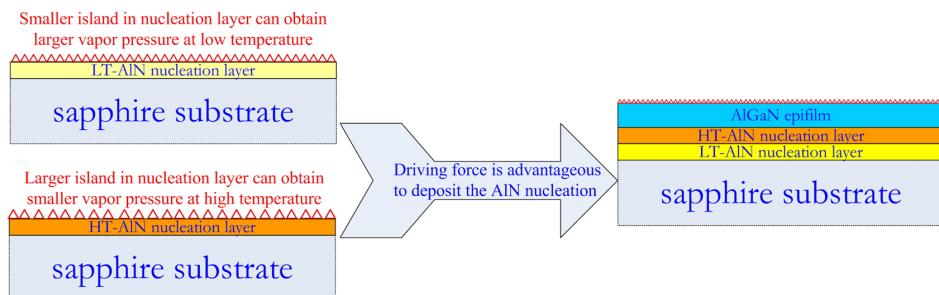


FIG. 6. Mechanism illustration of the morphology evolution for AlN nucleation layer grown at low- temperature and high-temperature.

HT-AlN nucleation layer and AlGaN epilayer. Results indicated that crystal quality of HT-AlN and AlGaN layer can be improved greatly, and the threading dislocation density existing in AlGaN epifilms has been reduced when LT-AlN nucleation layer was inserted, which is consistent with the surface morphology of the HT-AlN nucleation layer and AlGaN epifilm probed by AFM and SEM.

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