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Influence of initial growth stages on AlN epilayers grown by metal organic chemical vapor deposition

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ABSTRACT

AlN layers of thickness of about 2 μm have been grown with AlN nucleation layers (NLs) on (001) sapphire substrates using metal organic chemical vapor deposition. Increasing the AlN-NL deposition temperature from 850 to 1250 °C has been found to have significant effect on the surface morphology and the structural quality of the AlN layers. The surface morphology of the AlN-NLs and the AlN layers has been assessed using atomic force microscopy (AFM). The AFM images of the AlN-NLs reveal the coalescence pattern of NLs. AFM images of the AlN layers and the in-situ reflectance measurement disclose the surface morphology and the growth pattern of the AlN layers, respectively. Smooth surface with macro-steps and terrace features has been achieved for the AlN layer grown on the NL deposited at 950 °C. The structural quality of AlN layers has been studied by high resolution X-ray diffraction and Raman spectroscopy. The screw dislocation density from (002) reflection and the average edge dislocation density from (102), (302) and (100) reflections of the AlN layer on NL deposited at 950 °C are estimated to be $9\times10^{7}\,cm^{-2}$ and $4.4\times10^{9}\,cm^{-2}$, respectively. Lateral correlation length (L) is calculated from the (114) reciprocal space mapping of the AlN layers and correlated with the edge dislocation density of the AlN layers. Raman E_2 (high) phonon mode indicates compressive strain in the AlN layers grown on the NLs deposited at various temperatures. From this work, it has been inferred that the uniform coalescence of the nucleation islands and the complete coverage of AlN-NL determine the surface morphology and the structural quality of the subsequently grown AlN layers.

1. Introduction

Deep ultraviolet (DUV) light emitting diodes (LEDs) and laser diodes (LDs) are revolutionizing the field of optoelectronics with applications in high density optical data storage, biomedicine and water and air purification [1–3]. Aluminum nitride (AlN) and aluminum gallium nitride (AlGaN) are materials with the potential to realize DUV light emission due to their direct wide band gaps [4]. AlN renders minimal lattice mismatch to high Al-content AlGaN layers besides itself offering excellent UV light transparency [5], which makes it a suitable buffer layer for DUV-LED and LD device structures. However, the lack of a native substrate forces the heteroepitaxial growth of AlN on foreign substrates like sapphire, silicon carbide (SiC) and silicon (Si) which is found to result in a high density of threading dislocations (TDs) [6]. Sapphire is the most commonly used substrate for DUV devices due to its UV light transparency. Heteroepitaxial growth of AlN experiences many challenges, which have been addressed by adopting special approaches in the growth conditions: (i) high sticking coefficient of aluminum (Al) that restricts the migration of Al adatoms has been found to be reduced by growing AlN at growth temperatures higher than 1200 °C [7] and by using the pulsed growth or migration-enhanced metal organic chemical vapor deposition (MEMOCVD) method [8], (ii) unwanted parasitic reactions between Al and ammonia (NH_3) have been brought down by allowing high flow of hydrogen (H_2) carrier gas into the reactor by operating at low reactor pressure [9] and (iii) cracking of AlN layers due to thermal expansion mismatch between AlN and sapphire substrate has been minimized by multi-growth mode (i.e., varying the V/III ratio at different stages of AlN growth) [10]. In addition, the pre-treatment of the sapphire substrate has been noted to impact the polarity and surface morphology of AlN [11]. AlN with Al polarity has been found to exhibit a smoother surface while AlN with mixed polarity (Al and N) has a rougher surface morphology [11–13]. On the other hand, precursors flow prior to the

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deposition of the NLs [9], thickness of the NLs, the density and the size of the nucleation islands (NIs) have been found to influence the quality of the AlN layers [14–17]. Nevertheless, studies on the initial stages of AlN growth are still scarce, when compared to GaN.

In this paper, the effect of AlN-NLs deposition temperature and coalescence pattern on the surface morphology and the structural quality of subsequently grown AlN layers are reported. The aim is to grow a thick, specular and crack-free high quality AlN epilayer to utilize it as a template for AlGaN-based DUV-LED device structures.

2. Experimental procedure

AlN layers were grown using an Aixtron 200/4 RF-S metal organic chemical vapor deposition (MOCVD) system. The MOCVD system consists of a horizontal growth chamber with radio frequency (RF) heating. Trimethylaluminum (TMAl) and ammonia (NH_3) were used as the precursors for aluminum and nitrogen, respectively. Hydrogen (H_2) was the carrier gas. All the AlN growths were performed on 2″ *c*-plane sapphire substrates. AlN growth has been classified into two sets. Prior to growth the surface of the substrate was thermally cleaned at 1150 °C for 10 min in H_2 ambient. In the first set, three AlN-NLs have been deposited at 950, 1050 and 1150 °C on the sapphire substrates. These NLs have been recrystallized at 1250 °C for 2 min and unloaded from the reactor for the surface morphological characterization by atomic force microscopy (AFM) to understand the effect of NL deposition temperature.

The second set contains five AlN samples. The growth commences with the deposition of the NL followed by its recrystallization and the growth of a 2 μm thick AlN layer. The thickness of the AlN-NLs was measured from X-ray reflectivity (XRR) and that of the AlN layers calculated from the in-situ reflectance measurement curves and also cross-checked with cross sectional scanning electron microscopy (SEM) images. The only variation among the five AlN samples is the AlN-NL deposition temperature. The NL deposition temperatures were 850 °C, 950 °C, 1050 °C, 1150 °C and 1250 °C for the five AlN samples. The other growth parameters such as recrystallization temperature (T_{rc}) of AlN–NLs (T_{rc}=1250 °C), growth temperature (T_g) of AlN layers (T_g=1300 °C), recrystallization time (2 min), AlN growth time (60 min), molar V/III ratios during AlN-NL deposition (1024) and AlN layer growth (512) were kept identical.

The AlN layers grown on NLs deposited at temperature 850–1250 °C were characterized using AFM, high resolution X-ray diffraction (HR-XRD), and Raman spectroscopy. In addition, in-situ reflectance measurement was also done using a 635 nm laser line during the AlN growth.

3. Results and discussion

3.1. Investigation on nucleation layers

Fig. 1(a), (b) and (c) are the AFM images of the NLs deposited at 950, 1050 and 1150 °C, respectively and recrystallized at 1250 °C. AFM images show that the NLs are composed of three dimensional (3D) nucleation islands (NIs). However, no distinct islands can be seen and all the NIs have coalesced after the recrystallization of the NLs. On the other hand, the dissimilarity in coalescence pattern among the samples is noteworthy.

The surface roughness and thickness of the recrystallized NLs varies with increasing NL deposition temperature, the former found to increase and the latter found to decrease. Root mean square (RMS) roughness values of the NLs deposited at 950, 1050 and 1150 °C are 1.5 nm, 3 nm and 3.9 nm, respectively. The thickness of the NLs deposited at 950, 1050 and 1150 °C has been measured by XRR as 61 nm, 54 nm and 41 nm, respectively. The thickness of the recrystallized NLs appeared to be adequate for the subsequent AlN growth [14]. The variation in the RMS roughness values of the recrystallized NLs could be understood by the difference in the coalescence pattern. The NL deposited at 950 °C shown in Fig. 1(a) exhibits uniformly coalesced islands with the lowest surface roughness, whereas uneven coalescence with rough surface is observed for the NL deposited at 1050 °C (Fig. 1(b)). AlN-NL deposited at 1150 °C exhibits poor surface morphology with high undulations and insufficient coverage on the sapphire substrate as seen in Fig. 1(c). The difference in the coalescence pattern among the three NLs is clearly due to their deposition temperatures. In addition, the size of the NIs appears to play a major role in determining the coalescence pattern. From Fig. 1(a), the size of NIs seems to be similar before recrystallization which in turn can result in uniform coalescence after the recrystallization. The presence of both small and big coalesced island like structures in Fig. 1(b) indicates that there might be some variations in the size of the NIs prior to recrystallization. High roughness with heavy undulation seen in Fig. 1(c) can be attributed to the lack of sufficient NIs coverage on the sapphire surface in some parts and also to the possible considerable difference in the size of NIs. To study the coalescence pattern with respect to the NL deposition temperature on the quality of AlN layer, five samples were grown in the second set as described in Section 2.

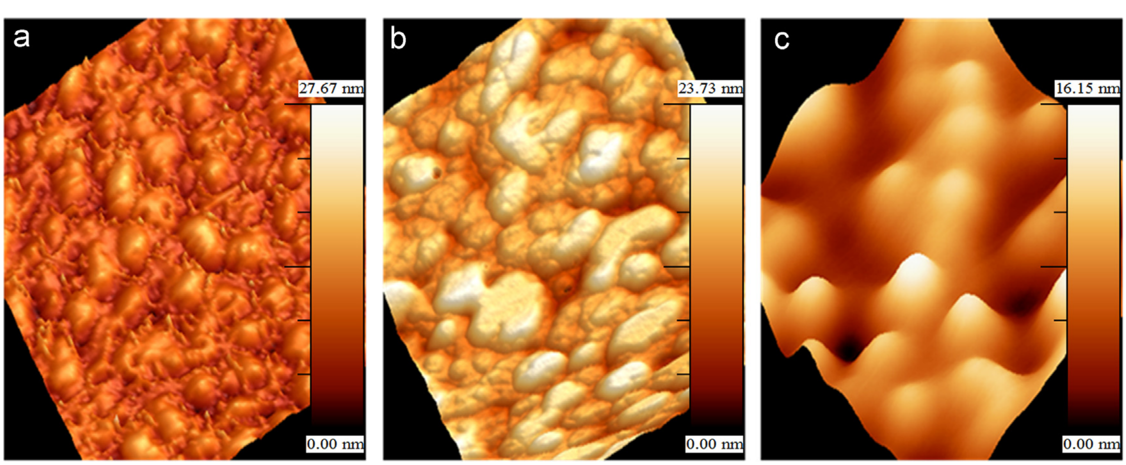

Fig. 1. 3D-AFM images of the recrystallized NLs deposited at (a) 950 °C, (b) 1050 °C and (c) 1150 °C. All scans are 1 μm × 1 μm. Recrystallization temperature was 1250 °C for all.