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Growth Temperature Dependency of High Al Content AlGaN Epilayers on AlN/Al_2O_3 Templates

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Abstract. In this work, MOVPE (Metalorganic Vapor Phase Epitaxy) growth and characterization studies of high Al content AlGaN epilayers are reported. We utilize high resolution X-ray diffraction (HRXRD) and atomic force microscope (AFM) techniques to analyze the crystalline quality and surface morphology of AlGaN epilayers. The role of the growth temperature of AlGaN epilayers on the structural quality and the surface morphology was investigated. Growth and measurement results show that single phase AlGaN epilayers were grown on AlN/Al_2O_3 template. It is concluded that the increasing growth temperature increases the Al content of AlGaN epilayers which enable to control the alloy concentration of AlGaN. Furthermore, the increasing Al content in AlGaN epilayers leads to the smooth surface which indicates that the decreasing number of dislocation density.

Keywords: AlGaN, MOVPE, AlN, Al2O3, X-ray diffraction, Atomic force microscope.

AlN/Al_2O_3 Şablonlar Üzerindeki Yüksek Al İçerikli AlGaN epi-Tabakaların Büyütme Sıcaklığı Bağımlılığı

Özet. Bu çalışmada, yüksek Al içerikli AlGaN epi-tabakaların MOVPE (Metalorganik Buhar Fazı Epitaksi) büyütmesi ve karakterizasyon çalışmaları rapor edilmiştir. AlGaN epi-tabakaların kristal kalitesi ve yüzey morfolojisi analizi için yüksek çözünürlüklü X-ışını kırınımı (HRXRD) ve atomik kuvvet mikroskobu teknikleri kullanılmıştır. Büyütme sıcaklığının AlGaN epi-tabakalarının yapısal kalitesi ve yüzey morfolojisi üzerindeki rolü incelenmiştir. Büyütme ve ölçüm sonuçları AlN/Al_2O_3 şablonu üzerine tek fazda AlGaN epi-tabakaların büyütüldüğünü göstermektedir. Artan büyütme sıcaklığının AlGaN'in Al içeriğini arttırdığı ve bunun AlGaN alaşım konsantrasyonunu kontrol etmeye olanak sağladığı sonucuna varılmıştır. Ayrıca, AlGaN epi-tabakalarındaki Al içeriğinin artışı pürüzsüz yüzeye yol açar ki bu dislokasyon yoğunluğunun azaldığını belirtir.

Anahtar Kelimeler: AlGaN, MOVPE, AlN, Al2O3, X-ışını kırınımı, Atomik kuvvet mikroskobu

1. INTRODUCTION

III-Nitride semiconductor compounds consist of the combination of group III elements such as Ga (gallium), In (indium), Al (aluminum) with N (nitrogen). They can be used widely in electronics [1-3] and optoelectronics [4-6] applications. In recent years new application areas of III-Nitride semiconductors have emerged especially for the UV (ultraviolet) radiation of the electromagnetic spectrum because of their superior advantages such as wide and direct band gap, high thermal stability, high breakdown field etc. [5]. The market forecast for UV-LEDs (light emitting diodes) is increasing from $90 million in the year 2014 to $800 million by the year 2020 due to the increasing demands in epoxy curing, water purification, food or medical sterilization, and many new emerging applications[5]. UV radiation can be divided into three regions, comprising the UV-A region (320–

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400 nm), the UV-B region (280–320 nm), and the UV-C region (100–280 nm) [7]. Due to the eminent optoelectronic properties in the UV region and especially in the deep ultraviolet (DUV) region of the spectrum, AlGaN material has received considerable attention recently. AlGaN-based UV-LEDs have many advantages such as small size, compact structure, low power consumption-low operating voltage, long lifetimes, high efficiency etc. compared to the mercury-based traditional UV light sources. AlGaN is a ternary alloy of direct bandgap of AlN and GaN compounds. Native AlN substrates can be considered the best material for DUV device applications but still AlN substrates are very expensive and they are still limited to very small sizes. There are many studies have been conducted to increase AlGaN and AlN quality on different foreign substrates such as silicon [8-11] but the most used one is sapphire since sapphire's transparency in DUV spectral region, low cost, wide availability in a large diameter, and stability at high temperatures. Generally, AlN buffer layer on sapphire substrate is used as a template for DUV applications because of low lattice mismatch between high Al content AlGaN and AlN. There are many difficulties to grow AlN and high Al content AlGaN alloy on sapphire substrate or on AlN/sapphire templates. There is a large lattice mismatch between AlN and sapphire which leads to a high dislocation density. Also, large thermal expansion coefficient difference between sapphire and AlN causes large strain and there occurs crack during cool down from the growth temperature. Additionally, there is an obstacle to grow AlN or high Al content of AlGaN alloy because of low surface migration mobility of Al atoms and parasitic pre-reactions between TMAl (trimethylaluminum) and NH3 (ammonia) results in dislocation density in AlGaN epilayers.

In this work, epitaxial AlGaN epilayers were grown by MOVPE (metalorganic vapor phase epitaxy) on AlN/sapphire templates with different growth temperatures. Structural quality and surface morphology of these structures were conducted by high resolution X-ray diffraction (HRXRD) and atomic force microscopy (AFM).

2. EXPERIMENTAL

AlGaN epitaxial layers were grown on c-plane sapphire substrates by using an Aixtron 200/4 RF-S low pressure, horizontal-flow MOVPE system with radio frequency (RF) heating. TMAl, trimethylgallium (TMGa) and NH_3 were used as precursors of Al, Ga, and N sources, respectively. Trimethylindium (TMIn) was used as a surfactant to increase the surface mobility of aluminum ad-atoms as studied previously [12-13]. Before start the growth, sapphire substrates were thermally desorbed at 1150 °C for 10 minutes under H_2 ambient to remove surface contamination and adsorbed water. A mixture of H_2 and N_2 was used as the carrier gas for the subsequent growth. A~20 nm thick low temperature AlN nucleation layer then 300 nm thick high temperature PALE (pulsed atomic layer epitaxy) AlN were grown on sapphire substrate. Growth details of this AlN/sapphire template were explained in detail in previous studies [8, 14]. Three (A, B and C) different AlGaN epilayers were grown on optimized AlN/sapphire template at 1020, 1180 and 1220 °C, respectively.

3. RESULTS AND DISCUSSION

The crystalline quality of all AlGaN/AlN/sapphire structures has been investigated by measuring 2θ/ω symmetric (0002) plane reflections via HRXRD (Figure 1). It is seen from the Figure 1 that there are three peaks belong to AlGaN/AlN/sapphire structures for each sample. The sharpest and the highest peak belongs to sapphire substrate. The middle one and the left one belong to AlN template and the AlGaN epilayers, respectively. HRXRD scans and analyses from the Figure 1 show that there are no excess peaks detected which indicates that other phases of AlGaN epilayers were not formed. Additionally, inset of Figure 1 shows that the AlN peak intensity increases with the increase of AlGaN growth temperature. Even the AlN quality is independent of the AlGaN epilayer growth temperature and the template is same for all growths it is thought that the increasing behavior of this peak is because of the deflection of x-ray beam from the AlGaN epilayers and it affected the AlN peak intensity.

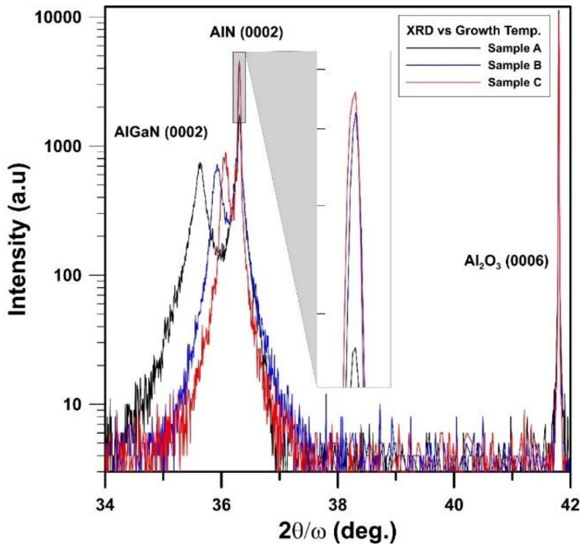

Figure 1. 2θ/ω HRXRD of Sample A (black), Sample B (blue) and Sample C (red). The inset shows the magnified AlN (0002) diffraction peaks of samples.

Figure 2 shows Al content and HRXRD peak FWHM (full width half maximum) of AlGaN epilayers variation with the increase of growth temperature. It is seen from the figure 2 that sample A, B and C have 54.8%, 74.5% and 83.9% Al content in AlGaN epilayers, respectively. It means that Al incorporation was increased linearly by the increase of growth temperature. There may be two factors which increase the Al content in AlGaN epilayers; the first one is the Al incorporation efficiency increases with the increase of growth temperature. The second one is considered to be due to the Ga suppression inability at higher growth temperatures. It can be concluded that it is possible to control the Al content in AlGaN epilayers by controlling the growth temperature. Figure 2 also shows the dependence of AlGaN XRD peak FWHM for (0002) reflection with the increase of growth temperature. It is 746 arcsec for sample A, 650 arcsec for sample B and 496 arcsec for sample C. It seems that the crystal quality of AlGaN epilayers improves as the Al content increases with the increase of growth temperature. It is considered that this is due to a decreasing lattice constant difference between AlGaN and AlN leading to decreasing of misfit dislocations at elevated temperatures because of the increased Al content.

It is also demonstrated in the literature that AlGaN crystal quality deteriorates as the AlN content decreases to approximately 0.40 and then improves as the AlN content decreases further because of the small lattice constant difference between two successive layers. At an AlN content of 0.40 and above, the growth mode changed to the layer by layer mode [two-dimensional (2D) growth mode]. Because the difference in the lattice constants between the AlGaN layer and the AlN underlying layer decreases with increasing AlN mole fraction, the density of misfit dislocations was reduced; thus, the FWHM value decreased as the AlN content was increased from 0.548 to 0.839 [15].

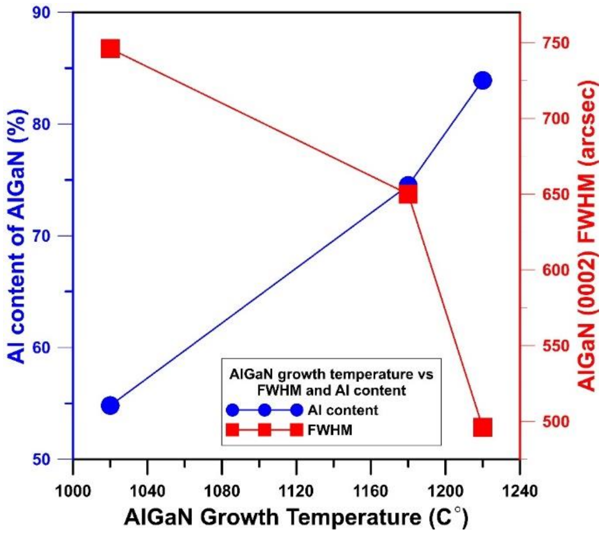

Figure 2. Al content (blue) and HRXRD peak FWHM (red) of AlGaN epilayers versus growth temperature.

In order to analyze the surface morphology of AlGaN epilayers, 5x5 μm^2 and 1x1 μm^2 regions were scanned by AFM at tapping mode. Figure 3 shows 5x5 μm^2 scan area images of AlGaN epilayer surfaces for all samples. Figure 3 demonstrates that lower growth temperature (1020 °C) of AlGaN (Sample A) results with the bumpy surface and for the higher growth temperatures (Sample B (1180 °C) and C (1220 °C) large area surfaces look smoother. The bumpy surface actually occurs because of the spiral type of AlGaN growths [16]. Spiral growth can be grouped into two different types: A and B. Type A occurs around a large dislocation core (e.g., a nanopit). In contrast, the more common type B is composed of two interlacing components rotated by 180◦ [16]. We have both kinds of spiral growth types for the lower growth temperature (sample A). With the decreasing of Al content in AlGaN epilayer, the strain increases, leading to more defects [17]. For the increasing growth temperatures (increased Al content in AlGaN) it seems that AlGaN surfaces don't have spiral growth mode anymore which indicates the decreasing number of density of dislocations.

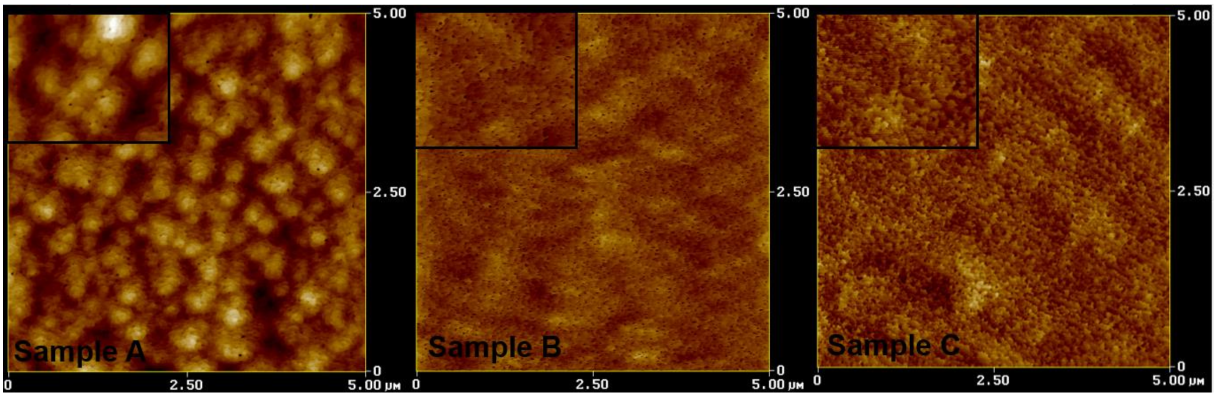

Figure 3. 5x5 μm^2 scan area AFM images of AlGaN epilayer surfaces for sample A, B and C. Upper left images show the magnified images of the region of interest.

When one checks the small area (1x1 μm^2) AFM images of samples it seems that there are many dark spots on the surface. These dark spots cannot be distinguished clearly on the surface of sample A because of the rough surface. These dark spots highlight the non-radiative behavior of the dislocations in AlGaN [18]. Based on their different sizes dark spots can be classified as pure-edge type (small), pure-screw type (large) and mixed-type (middle) threading dislocations [19]. Except for the dark spots atomic steps on the AlGaN surfaces can be clearly seen which indicate the smooth surface morphology. It is also important to note that the average root mean square roughness (rms) of sample surfaces to realize the effect of growth temperature quantitatively.

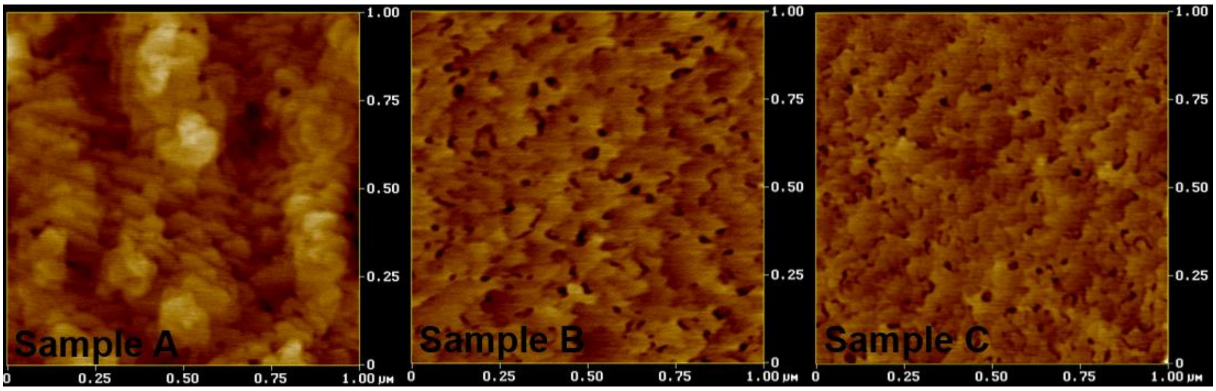

Figure 4. 1x1 μm^2 scan area AFM images of AlGaN epilayer surfaces for sample A, B, and C.

Figure 5 shows the rms values variation of 5x5 μm^2 (red square) and 1x1 μm^2 (blue circle) AFM scan regions for the increasing growth temperature of AlGaN epilayers. The rms values of 5x5 μm^2 and 1x1 μm^2 AFM scan areas for sample A, B and C are 0.61, 0.35, 0.22 nm and 0.30, 0.23, 0.19, respectively. It is clearly seen from the Figure 5 that the growth temperature effects the rms roughness of the AlGaN surfaces so that when the growth temperature increases the surface getting smoother. It is again indicating that the more temperature the less roughness on the surface which demonstrates the reducing of dislocation density.

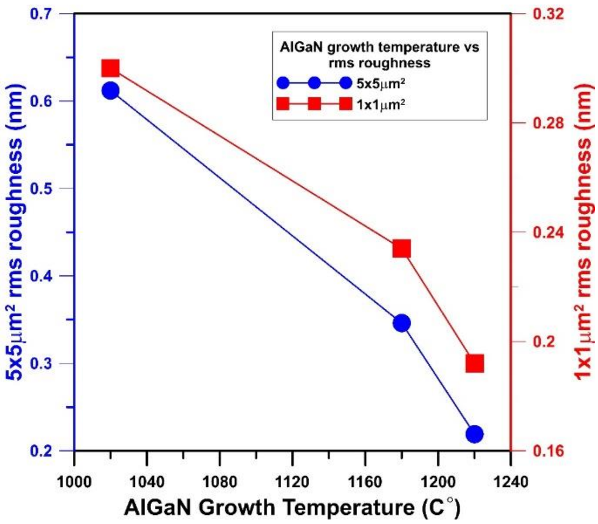

Figure 5. Rms values for 5x5 μm^2 (red square) and 1x1 μm^2 (blue circle) regions versus growth temperature of AlGaN epilayers.

4. CONCLUSIONS

In this paper, the crystalline quality and surface morphology of MOVPE grown AlGaN epilayers has been studied. 2θ/ω HRXRD FWHM of AlGaN (0002) diffraction peaks and surface root mean square roughness variation were investigated by changing the growth temperature. It was shown that the growth temperature change from 1020°C to 1220 °C increased the Al content of AlGaN epilayers from 54.8% to 83.9%. It was also shown that surface root mean square roughness values for 5x5 μm^2 and 1x1 μm^2 scan regions was improved from 0.61 to 0.22nm and from 0.30 nm to 0.19 nm, respectively.

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