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Structural properties of Al-rich AlInN grown on *c*-plane GaN substrate by metal-organic chemical vapor deposition

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Abstract

The attractive prospect for AlInN/GaN-based devices for high electron mobility transistors with advanced structure relies on high-quality AlInN epilayer. In this work, we demonstrate the growth of high-quality Al-rich AlInN films deposited on *c*-plane GaN substrate by metal-organic chemical vapor deposition. X-ray diffraction, scanning electron microscopy, and scanning transmission electron microscopy show that the films lattice-matched with GaN can have a very smooth surface with good crystallinity and uniform distribution of Al and In in AlInN.

Keywords: Metal-organic chemical vapor deposition; Epitaxy; AlInN; GaN

Background

AlInN is a newly developed III-nitride for many promising applications due to its band gap being able to be tuned in a wide range of 0.70 ~ 6.14 eV with high spontaneous polarization [1-3]. AlInN/GaN heterojunction has been suggested as a strong candidate for high-power and high-frequency applications owing to the nearly lattice match ($\text{In} = 0.17$) and stress-free heterostructure in which the piezoelectric polarization charge is eliminated to reduce surface-related current collapse [4-8]. The attractive prospect for AlInN/GaN-based devices for high electron mobility transistors with advanced structure relies on high-quality AlInN epilayer. However, many properties of ternary AlInN alloys have not been well understood because of lack of high-quality films. Growth of AlInN is complicated by thermal control, which usually results in growth with composition inhomogeneities and phase separation [9-11]. Especially, the thermal instability may lead to phase separations and other defects which form on the substrate and may evolve during the growth resulting in rough and poorly uniform surface. The lattice-matched of high-quality AlInN can potentially lead to a reduction in threading dislocation and cracking as well as the elimination of strain-driven piezoelectric polarization field [12].

Various growth techniques have been used for growth of AlInN films, such as metal-organic chemical vapor deposition (MOCVD), radio-frequency molecular beam epitaxy, pulse laser deposition, and magnetron sputtering on *c*-plane sapphire substrates [13-15]. In the past few years, most of AlInN/GaN studies are done on films grown on sapphire substrate which has strong effects on the film properties, and among them, lattice-matched $\text{Al}_{1-x}\text{In}_x\text{N}/\text{GaN}$ with $0.17 \sim x \sim 0.18$ has received intensive attention [16-18]. The nearly lattice-matched AlInN layer grown on GaN/sapphire substrate usually contain some defects like hillocks, dislocations natively present in the GaN layer and the V-defects on GaN surface [19-21]. However, recent studied show that the growth of AlInN on GaN remains a challenge as many issues have to be addressed [22,23]. Hereinafter, we show that a high-quality and smooth $\text{Al}_{1-x}\text{In}_x\text{N}$ film with x less than 0.17 can be epitaxially grown on GaN free-standing substrate with in-plane lattice match by employing MOCVD.

Methods

In this work, we used hydride vapor phase epitaxy (HVPE)-grown GaN wafers as substrates which had a 1.5-in. diameter and 300- μm thickness with a full width at half maximum (FWHM) of the (0002) X-ray rocking curve (XRC) 100 arcsec (dislocation density approximately $1 \times 10^7 \text{ cm}^{-2}$) and a root mean square (RMS) surface roughness of 1.23 nm. A 1.5- μm -thick homoepitaxial GaN

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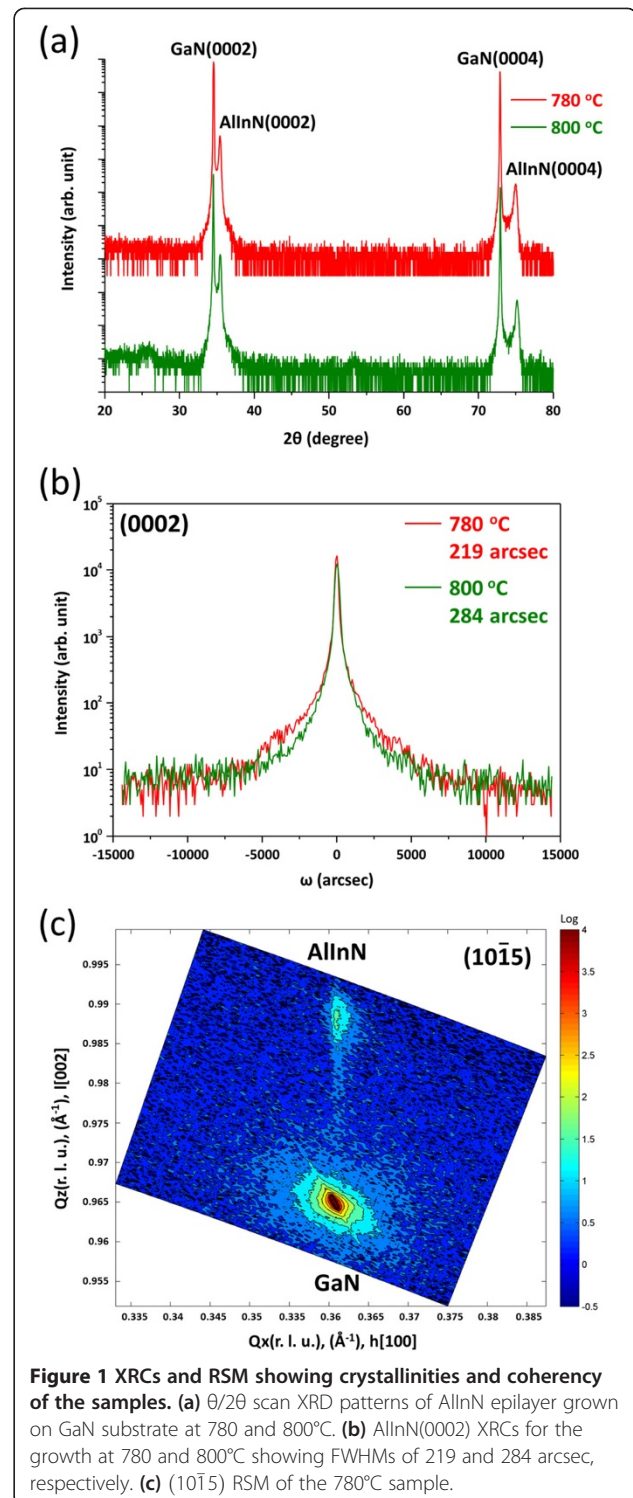
film was firstly grown on GaN substrate at 970°C by MOCVD (Veeco Emcore D-180; Veeco Instruments Inc., Plainview, NY, USA) using TMGa and NH₃ as the precursors for Ga and N to improve GaN crystallinity. After the growth of GaN layer, TMAI and NH₃ were flowed into the chamber at 990°C for 1 min [24], followed by further growth of AlInN epilayer at temperature of 780 and 800°C. A TMAI/TMIn ratio of 1/10, a reactor pressure of 100 Torr, and a NH₃ flow rate of 15,000 sccm were used for all AlInN growth.

The crystallinities of the samples were examined with high-resolution X-ray diffraction (XRD, Bruker D8; Bruker Corp., Billerica, MA, USA), and the surface morphologies were investigated by scanning electron microscopy (SEM, JEOL JSM-6500F; JEOL Ltd., Akishima, Tokyo, Japan) and atomic force microscopy (AFM, Veeco Innova). Structural characterization at atomic scale was performed in a JEOL JEM-ARM200F spherical aberration corrected scanning transmission electron microscope (STEM) in high-angle annular dark field (HAADF) imaging mode, operated at 200 kV. Cross-sectional transmission electron microscopy specimens were prepared in a focused ion beam system (FEI NOVA-200; FEI Company, Hillsboro, OR, USA) using a 30-kV Ga⁺ source.

Results and discussion

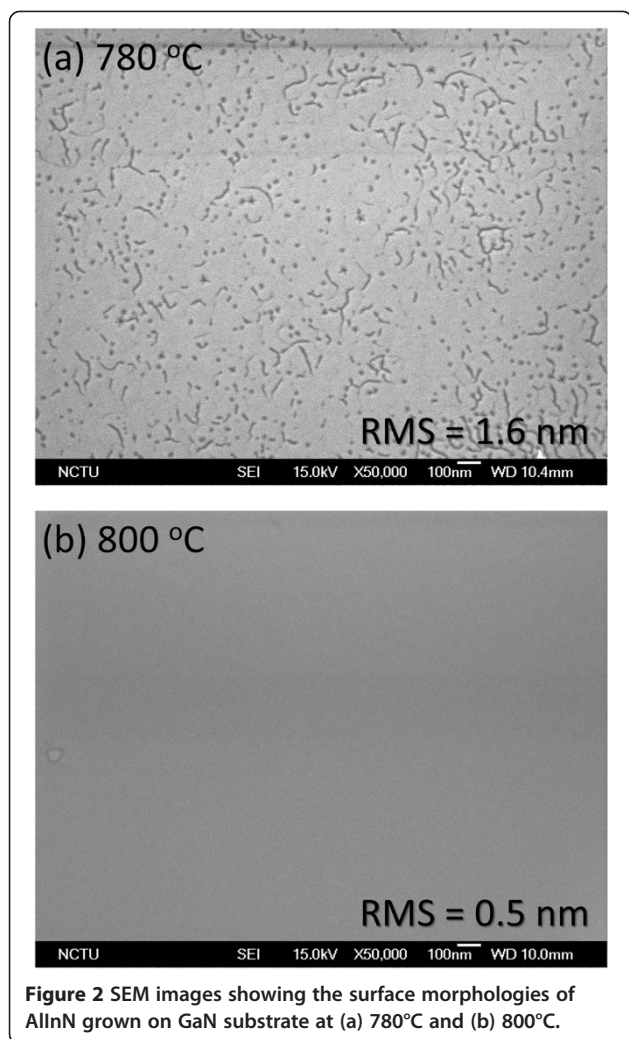
The crystallinities of the samples as examined with XRD can be seen in Figure 1a for the 780 and 800°C-grown AlInN film, in which Al_{1-x}In_xN exhibits only (0002)/(0004) reflections, suggesting that it is of single phase. By using Bragg's law, the measured AlInN (0004) interplanar spacing can give $c = 5.064 \text{ \AA}$ of the 780°C sample. From previous studies by Lorenz et al. [1] and Darakchieva et al. [25], the Al_{1-x}In_xN layer may have a composition with x approximately 0.13 which was determined by Rutherford backscattering spectroscopy and XRD. Also, the film has a better crystallinity with the (0002) XRC full width at half maximum (FWHM) of 219 arcsec as shown in Figure 1b than that of the 800°C-grown AlInN which can be shown x about 0.10. In previous studies of MOCVD growth, it has been shown that the high-temperature growth results in low In composition because of In desorption with temperature [26]. In Figure 1c, asymmetric reciprocal space mapping (RSM) shows that the reciprocal-lattice points of AlInN (10 $\bar{1}$ 5) and GaN (10 $\bar{1}$ 5) are well aligned in Q_x direction, i.e., in-plane lattice match, indicating pseudomorphic growth of AlInN on GaN [27].

The surface morphologies of the AlInN epilayers grown on GaN substrate by MOCVD were investigated with SEM and AFM. The surface of 800°C sample is much smoother than 780°C sample as shown in Figure 2. The RMS surface roughness for AlInN grown at 800°C as determined by AFM from $3 \times 3 \mu\text{m}^2$ scanning area is 0.5 nm, compared with 1.6 nm for the 780°C sample. Similar

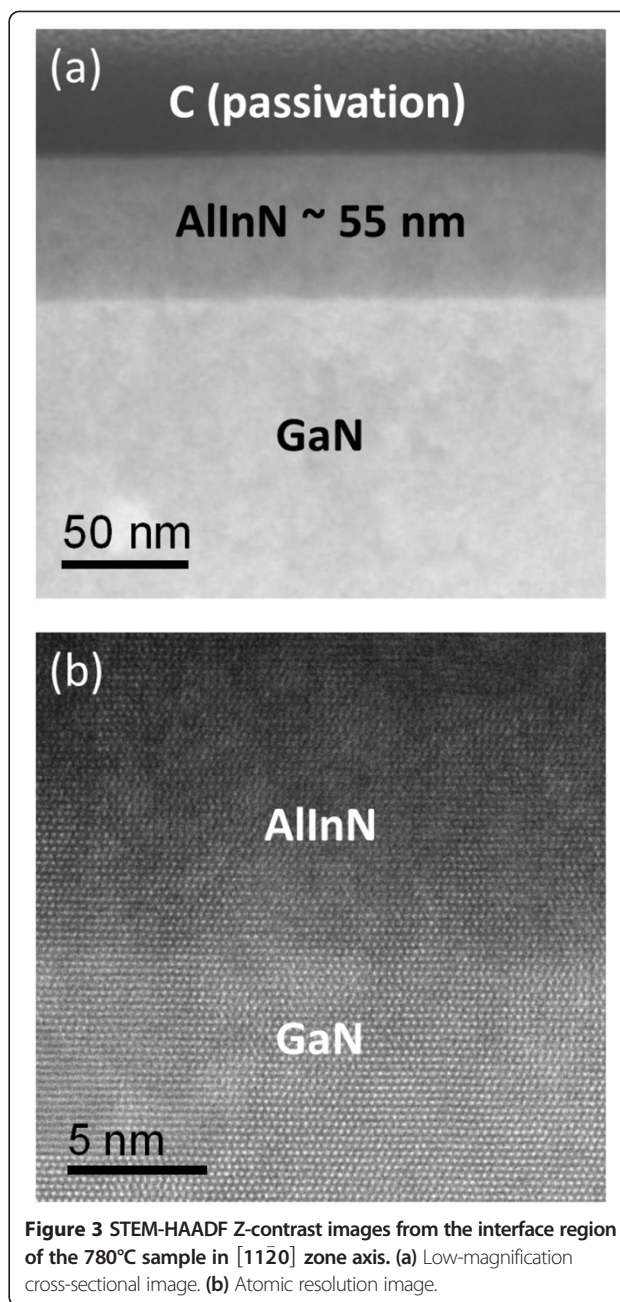


observation has been reported by Ichikawa et al. that the higher growth temperature results in a smoother surface with a decrease of the In composition [28].

As the AlInN film is shown to be lattice-matched with GaN in RSM, the lattice images from high-resolution TEM are difficult to resolve the interface. Therefore, we



used the STEM-HAADF images in Z-contrast for characterization of the interface and examination of the uniformity of the film. The HAADF images were acquired with the inner and outer collection semiangles of 68 and 174.5 mrad, respectively, and with a probe size of 1.5 Å. Figure 3a shows a low-magnification STEM-HAADF image of the 780°C sample in a cross-sectional view along the $[11\bar{2}0]$ zone-axis. As can be seen, the contrast of the GaN substrate is much brighter than that of the AlInN layer, consistent with the Z-contrast interpretation since the average atomic number is greater for GaN than Al-rich AlInN. Also, the image shows that the AlInN/GaN interface is sharp and the AlInN film is about 55 nm thick with a uniform distribution of Al and In in this ternary structure at nanometer scale. An atomic resolution STEM-HAADF image from the interface region is shown in Figure 3b. Each bright dot corresponds to the atomic column Al/InN and Ga in AlInN and GaN with invisible N atoms, and the intensity of Ga is much stronger



than Al/In as expected. From the image contrast, both AlInN and GaN have reasonably uniform distributions around the interfacial region. Furthermore, it can be shown that the Al/In and Ga atomic arrangements are in excellent lattice match at the interface without any misfit dislocations, consistent with the above RSM result.

The above results show that lattice-matched $\text{Al}_{1-x}\text{In}_x\text{N}$ films on GaN with $x < 0.17$ are of high crystallinity with quite smooth surface, which can be comparable with those of $\text{Al}_{1-x}\text{In}_x\text{N}$ films grown on free-standing GaN substrates with x close to 0.17 [18].

Conclusions

In this work, we successfully fabricated high-quality AlInN/GaN heterostructures by MOCVD. XRD, SEM, and STEM results reveal that the film is of good crystallinity and uniformity in composition, smooth surface flatness, and the abrupt heterointerface with lattice-matched AlInN epilayer on free-standing GaN substrate at atomic scale.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

PYL carried out the XRD and STEM works and wrote the manuscript. JYC designed the thin-film deposition process and carried out the SEM work. YSS helped in the RSM measurements. LC conceived of the study and participated in its design and coordination with analysis and interpretation of data and the revision of the manuscript. All authors read and approved the final manuscript.

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