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Studies on dislocation and surface morphology of $Al_xGa_{1-x}N$/GaN heterostructures grown by MOCVD

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ABSTRACT

In this work, $Al_xGa_{1-x}N$ epilayers have been grown on sapphire substrate with GaN template by the metal organic chemical vapor deposition (MOCVD) method. The structural and morphological properties of these samples have been investigated and compared. The growth rate of AlGaN has been found to decrease with increasing Al composition. By increasing Al composition of AlGaN epilayer, tilt and twist angle has been found to increase, indicating higher threading dislocation density. Surface morphology and dislocation density (DD) have been investigated by scanning electron microscopy (SEM) and atomic force microscopy (AFM). Different types of dislocation etch pits have been observed in $Al_xGa_{1-x}N$/GaN epilayers, using orthophosphoric acid (85% H_3PO_4) as defect selective etchant. Three types of etch pits like screw type (α), edge type (β) and mixed type ($\alpha+\beta$) dislocations have been observed. The mechanism of etch pits formation has been explained using Cabrera's thermodynamic model. The etch pit density (EPDs) has been correlated with threading dislocation density (TDs) estimated using HRXRD measurements. Photoluminescence (PL) studies have revealed an increase in intensity of near band edge emission due to etching.

1. Introduction

Group III-nitride (GaN, AlN and InN) semiconductors and its compounds are outstanding electronic materials for the next generation optoelectronics, high frequency and power electronic devices [1–3]. AlGaN ternary system appears attractive for applications in the UV range with a tunable band gap from 3.4 eV for pure GaN to 6.02 eV for pure AlN and are important for optoelectronic devices such as light-emitting diodes (LEDs) and photo-detectors [4]. The major problem in these III-nitrides and its alloys particularly in AlGaN/GaN is the presence of high defect density and biaxial strain due to the heteroepitaxial growth on foreign substrates, which results in lower performance and shortened device lifetime. This results in GaN layers containing threading dislocation densities (TDs) of 10^8–10^{10} cm^{-2} because of a strong mismatch in the lattice constant and the thermal expansion coefficient between the substrate and the buffer layer [5,6]. Identification of these TDs is critical for AlGaN/GaN epilayer growth and device performance analysis [7]. Moustakas [8] have studied that the apparent empirical correlation between non-radiative recombination centers and dislocations may be related to the most active point defects existing at threading dislocations. Zhao et al. [9] have found that the dislocations may act as scattering centers in GaN epilayers. Hussein et al. [10] have reported that AlGaN barrier with larger Al content provides a large conduction band discontinuity and a high Schottky barrier height, where both enhance the performance of devices. Although, the sheet carrier density can be improved by increasing the Al content in the ternary layer, high Al content deteriorates the quality of AlGaN epilayer. Peng et al. [11] have demonstrated that the decrease of two dimensional electron gas (2DEG) mobility is attributed to higher dislocation density in the sample with higher Al composition of AlGaN buffer. Kaun et al. [12] have stated that TDs are shown to influence early degradation in AlGaN/GaN high electron mobility transistors (HEMTs). Ivo et al. have also [13] reported that electrons are bypassing the gate control region via defect clusters in the GaN buffer and the occurrence of severe degradation. To identify and evaluate the TDs of AlGaN/GaN epilayer, numerous evaluation techniques such as high resolution X-ray diffraction (HRXRD), wet etching, cathode luminescence (CL), atomic force microscopy (AFM), high temperature annealing method and transmission electron microscopy (TEM) have been adopted [14,15]. Threading dislocations in AlGaN/GaN, such as screw dislocation, edge dislocation, nanopipes, stacking faults and several other types of dislocations have been found to cause many practical problems in device fabrication. Hence, a systematic investigation of TDs in AlGaN/GaN epilayers is essential to understand the correlation between structure and device performance.

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There are a few reports which have investigated TDs in AlGaN/GaN by wet chemical etching method [16,17].

In the present investigation, focus has been on MOCVD growth and characterization of the AlGaN/GaN with different Al compositions. Structural, morphological and optical properties of these structures have been investigated and compared. HRXRD and AFM have been performed to study the structural properties and crystalline quality, while optical properties have been studied by the room temperature photoluminescence (PL). The dislocation density has been estimated through wet-chemical etching and HRXRD. The etched surface morphology of $Al_xGa_{1-x}N$/GaN has been observed by SEM and AFM. Screw, edge and mixed type dislocations have been categorized by pit shapes and sizes. The TDs have been calculated using HRXRD and compared with etch pit density (EPDs). The relation between types of dislocation, radiative and nonradiative centers have been discussed.

2. Experimental details

$Al_xGa_{1-x}N$/GaN with $0.15 \leqslant x \leqslant 0.51$ epitaxial layers of various thicknesses were grown on a two-inch diameter, 430-μm-thick *c*-plane sapphire substrates using a horizontal MOCVD system (AIXTRON 200/RF-S) with gas foil rotation. Trimethylgallium (TMGa), Trimethylaluminum (TMAl) and Ammonia (NH_3) were used as Ga, Al and N precursors respectively. High purity Hydrogen (H_2) was used as carrier gas. Desorption of native oxide on the substrate was carried out at 1100 °C under the hydrogen ambient to improve the surface quality of sapphire. After desorption, substrate was cooled down to a temperature of 500 °C and 30 nm-thick GaN nucleation layer was deposited. The flow rate of TMGa and NH_3 were maintained at 5 sccm and 1500 sccm respectively with a V/III ratio of 3584. A 1.5 μm-thick undoped GaN layer was grown at 1040 °C, with the growth rate of 1.5 μm/h and V/III ratio of 539. $Al_xGa_{1-x}N$ layers were grown at 1080 °C by changing TMAl flow rate of 35–170 sccm by keeping the growth period constant. The flow rates of TMGa and NH_3 were kept constant as 6 and 1300 sccm respectively for the growth of $Al_xGa_{1-x}N$. In all the $Al_xGa_{1-x}N$ samples, the nucleation and buffer layer growth conditions were kept constant. HRXRD measurements were made by a PANalytical X'pert MRD system using Cu Kα_1 (λ = 1.540553 Å) radiation with double crystal diffraction. The beam divergence and the wavelength spread were reduced in a four-crystal monochromator built from germanium single crystals. X-ray diffraction ω–2θ measurement was used to characterize the Al mole fraction of the $Al_xGa_{1-x}N$ layer. X-ray rocking curves of symmetric (0002) plane and asymmetric (10–12) plane were used to evaluate different types of dislocations.

For the wet etching experiments, $Al_xGa_{1-x}N$ samples were cut into small pieces (2 cm × 2 cm), cleaned with deionized water followed by isopropyl alcohol and etched in commercial 85% H_3PO_4. Samples were immersed in the orthophosphoric acid solution for 5–120 s. Samples were quenched in cool water to halt etching followed by N_2 drying. The etching temperature of 180–230 °C was calibrated with a mercury thermometer. Initially, the morphology of the etched surface and the etch pits were examined by a differential interference contrast optical microscopy (A1 Axio Scope Carl Zeiss). SEM (Carl Zeiss MA15/EVO18) and AFM (Park XE-100) were utilized to observe the etched surface and morphology of the AlGaN epilayers. PL measurements were performed under an excitation power 30 mW using the UV-244 nm line of a continuous wave Ar^+ laser frequency doubling unit. The emissions were detected by an UV-enhanced photomultiplier tube. The chemical reaction involving ortho phosphoric acid and AlGaN in the etching process is given below.

$$AlGaN + H_3PO_4 \rightarrow Ga^{3+} + Al^{3+} + PO_4^{3-} + NH_3 \quad (1)$$

3. Results and discussion

Fig. 1 represents the (0002) HRXRD ω–2θ scan of the $Al_xGa_{1-x}N$/GaN epilayers. The Al composition and thickness has been determined according to Vegard's law by simulation of the X-ray diffraction pattern using the commercial software package PANalytical Epitaxy and Smooth fit. The energy dispersive X-ray spectroscopy (EDX) measurements have been done at least in five regions of each sample and average Al composition has been determined. A variation of about 0.5% has been observed for all the samples. The AlGaN epilayer thickness has been confirmed by cross-sectional SEM images (Fig. 2). The determined composition and the corresponding thickness have been listed in Table 1. The high intensity peak is associated with GaN template, whereas secondary peaks situated at higher angles with lower intensities are due to $Al_xGa_{1-x}N$ epilayer. The peak shift observed is due to the Al composition. The increase of Al composition in the AlGaN epilayer has been noted to lead to the shift of secondary peaks to the higher angle side compared with GaN peak in the (0002) HRXRD ω–2θ scan. This is due to the decrease in lattice parameters due to the incorporation of Al. For the higher Al composition ($\geqslant$0.33) the peak shift is noted to be high compared to lower Al composition AlGaN epilayers.

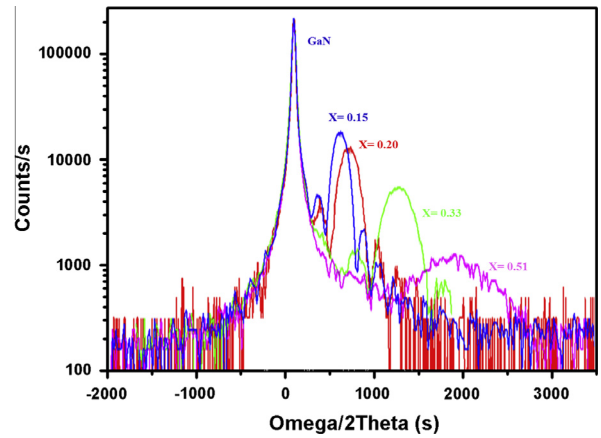

Fig. 1. HRXRD (0002) ω–2θ reflection of $Al_xGa_{1-x}N$/GaN samples.

As the Al composition increases, the XRD peak intensity is found to reduce which is due to the decrease in the thickness of the epilayer as a result of decrease in the growth rate with increase in Al composition and increase in the dislocation density. The decreasing growth rate with increasing Al composition has been attributed to high sticking coefficient of aluminum and the parasitic reaction between TMAl and NH_3 in gas phase. This is because of the fact that the Al–N (2.88 eV) bond strength is higher when compared to Ga–N (2.2 eV) and hence the disruption of AlN is much more difficult than that of GaN. Hence the incorporation of Ga has been reduced due to stronger adsorption of Al and the consequence is found to decrease in the growth rate of AlGaN. The precursor materials TMAl and NH_3 are known to undergo parasitic pre-reactions to form TMAl:NH_3 adduct formation which is noted to decrease the growth rate on increasing the flow rate of TMAl for higher composition.

3.1. Determination of threading dislocation using HRXRD measurement

Fig. 3 shows the Williamson–Hall (W–H) plots based on the broadening of the rocking curve (ω scan) on symmetric (0002, 0004, and 0006) reflections for AlGaN epilayer which is influenced only by the tilt and the coherence length parallel ($L_{||}$) to the substrate surface. $L_{||}$ has been plotted against each reflection and fitted by straight line, where β is the FWHM in angular units, θ is the Bragg reflection angle and λ is the X-ray wavelength. Then the tilt angle α_{tilt} has been obtained from the slope of the linear dependence and the lateral coherence length ($L_{||}$) from the inverse of the *y*-intersection y_0 of the fitted line with the ordinate. Williamson–Hall plot was plotted against each reflection and fitted by a straight line. From the *y*-intersection, y_0 the vertical correlation length $L_\perp$ could be calculated. The twist angles and calculated edge dislocation density have been obtained directly from Φ-scans on asymmetric reflection planes. Table 1 shows the lateral coherence lengths $L_{||}$ and tilt angles of various Al composition. From the table, it can be observed that the tilt angle increases and the corresponding lateral coherence $L_{||}$ lengths decreases with increase in Al composition in the AlGaN epilayer. The vertical coherence lengths $L_\perp$ values have been tabulated in Table 1 and found to be in the range from 0.092 to 0.018 μm. The threading dislocation