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Direct growth of epitaxial In-rich $In_xAl_{1-x}N$ ternary alloys on Si (111) substrate by RF-MOMBE

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1. Introduction

The technological importance of group III nitrides GaN, InN and AlN, particularly for the light-emitting and laser diodes operating in green and blue spectral regions, has stimulated the study of $Al_xGa_{1-x}N$, $In_xGa_{1-x}N$ and $In_xAl_{1-x}N$ alloys. The hexagonal InAlN alloy offers various unique properties which may improve the performance of electronic and optoelectronic devices. Because of InAlN has a direct gap that can be tuned in the range from 6.2eV for AlN to 0.7eV for InN[1]. Especially, In-rich InAlN is a promising material for multi-junction tandem solar cells[2]. For growth of InAlN in large area at low cost, deposition on Si (111) is of great interest. However, it is difficult to grow In-rich InAlN of single phase, and its epitaxy on Si substrate is also a challenge due to large lattice mismatch. Although InAlN-based devices have been produced, the growth mechanism of InAlN on Si substrate is still unclear.

Various methods have been used to fabricate InAlN films, such as radio frequency plasma-assisted molecular beam epitaxy[3], metalorganic chemical vapor deposition[4], pulsed laser deposition[5] and magnetron sputtering[6]. In this work, we focus on high-In-content InAlN/Si(111) heteroepitaxy by radio frequency plasma-assisted metalorganic molecular beam epitaxy (RF-MOMBE). Crystalline and surface morphology are characterized by high resolution x-ray diffraction (XRD), transmission electron microscopy (TEM) and field-emission scanning electron microscopy (SEM).

2. Experimental

In-rich $In_xAl_{1-x}N$ alloys were directly grown on Si (111) substrate by RF-MOMBE. Trimethylindium (TMIn) and trimethylaluminium (TMAl) were used as In and Al sources. TMIn/TMA molar ratio was fixed to 3.3. Active nitrogen radicals were supplied from a RF plasma source (13.56MHz) with 400W and the N_2 flow rate of 1sccm. The Si (111) substrates were cleaned in a wet bench using Radio Corporation of America processes. Also, the substrate was further wet-etched in buffered oxide etch for 30s, and loaded then into the growth chamber for InAlN growth. Prior to InAlN growth, the Si (111) substrate was heated at 900°C for 30 min with base pressure (8×10^{-9}Torr) for surface thermal cleaning and oxide removal. The substrate temperature was then decreased at 460-550°C for growth of InAlN films. During the deposition, the substrate temperature was monitored by a thermocouple (contact with heater backside).

XRD measurements were carried out in a Bruker D8 system using Cu-K_α radiation. The surface morphologies and microstructure of the $In_xAl_{1-x}N$ films were analyzed using a FE-SEM (Hitachi S-4300). The detailed microstructure of the InAlN films and interface were investigated by TEM in cross section (Philips Tecnai 20). High-angle annular dark field (HAADF) images in Scanning transmission electron microscopy (STEM) were obtained from a JEOL 2010F microscope.

3. Results and discussion

Figure 1 plots θ–2θ XRD patterns for InAlN films grown on Si (111) substrate at various substrate temperatures. The Si (111), Si (222), InAlN (0002), and InAlN (0004) reflections are observed in the patterns, suggesting that the InAlN films may consist of single phase of wurtzite structure. Also, it is seen that the (0002) peak appears at higher 2θ angle with increasing the substrate temperature. The Vegard's law has been applied to determine the average In composition of ternary alloy films via measurement of lattice parameters from XRD [7]. The In composition in the deposited $In_xAl_{1-x}N$ films is accordingly determined to be x = 0.8 for 460°C, x = 0.82 for 490°C, x = 0.89 for 520°C, and x= 0.42 for 550 °C. Also only the (0002)/(0004) peaks are observed without other reflections for all $In_xAl_{1-x}N$ films, indicating that these films are preferentially grown along the *c*-axis direction. For growth above 550°C, InAlN exhibits an extremely weak and broad (0002) peak, suggesting that the crystallinity is poor.

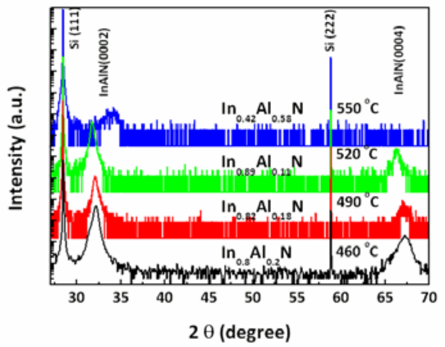

Fig. 1 θ-2θ XRD patterns of InAlN films deposited on Si (111) at different temperature showing varied indium compositions.

The phi-scan of the InAlN $(10\bar{1}1)$ plane is presented in Fig. 2. The diffraction peaks from the $\{10\bar{1}1\}$ plane of InAlN are observed at 60° interval, suggesting that the InAlN film is in epitaxy with Si. Epitaxial relationship can be deduced from the pattern as $(0002)_{InAlN}$ // $(111)_{Si}$ and $[11\bar{2}0]_{InAlN}$ // $[1\bar{1}0]_{Si}$.

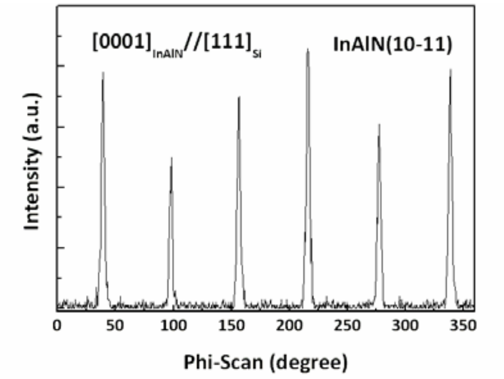

Fig. 2 $(10\bar{1}1)$ phi–scan of $In_{0.8}Al_{0.2}N$ films.

Figure 3 shows cross-sectional SEM images of the InAlN films grown on Si (111). From the images, the growth rate of ~ 0.25 μm/hr is almost unchanged for growth at 460-520°C. However, InAlN grown at 550°C is shown with 40 nm thickness, indicating that the growth rate is much lower probably due to InAlN decomposition similar to InN growth at temperature above 550°C[8]. The surface morphology in the insets of top-view images shows 3D grain feature, suggesting that the films are grown in island growth mode.

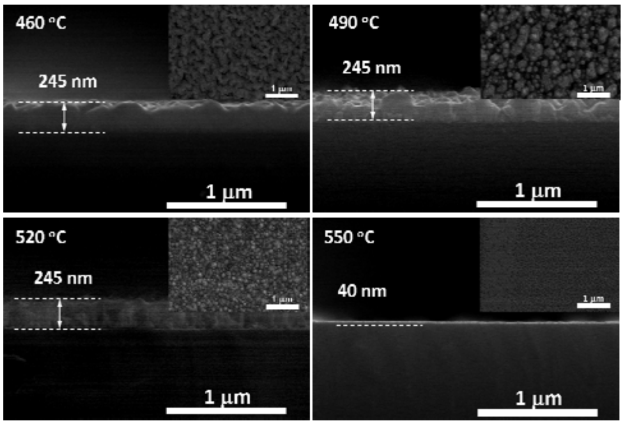

Fig. 3 Cross-sectional SEM images of $In_xAl_{1-x}N$ films grown on Si(111) with different In compositions. The inset shows surface morphology.

Figure 4 shows a bright field TEM image of $In_{0.8}Al_{0.2}N$ film. The surface morphology of the InAlN film exhibits rough shape which might be resulted from 3D growth mode. Selected area diffraction pattern (SADP) indicates that (0001)-oriented hexagonal InAlN has epitaxial relationship with the Si (111) substrate. Also, the STEM-HAADF image of $In_{0.8}Al_{0.2}N$ shows a sharp interface between the InAlN and Si. Furthermore, we can see in the HRTEM image from an InAlN/Si interfacial region that the InAlN layer is in contact with Si without formation of an interlayer between them.

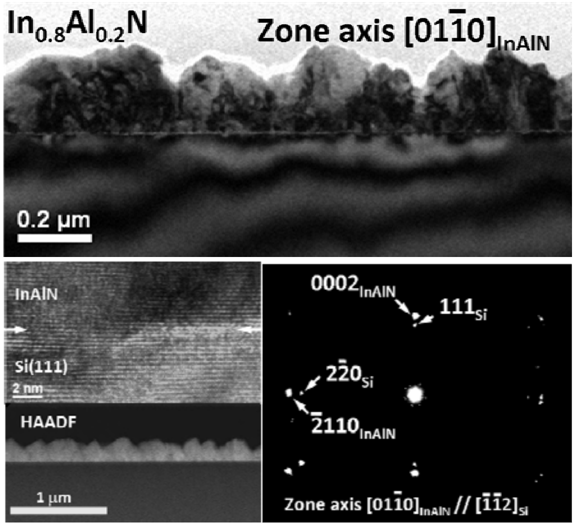

Fig. 4 Cross-sectional TEM image and the corresponding SAD pattern from $In_{0.8}Al_{0.2}N$. Also, STEM-HAADF micrograph of a typical $In_{0.8}Al_{0.2}N$ interface area. HRTEM of InAlN/Si interfacial region.

4. Conclusions

Direct growth of In-rich InAlN film on Si(111) substrate without any buffer layer by RF-MOMBE has been attempted in the temperature range from 460-550°C. All the grown InAlN (0002) films are of single phase and are highly oriented in c-axis. Epitaxial $In_xAl_{1-x}N$ can be grown on Si (111) at about 460°C.

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