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PREFERENTIAL ORIENTATION GROWTH OF AlN THIN FILMS ON Si(111) SUBSTRATES BY PLASMA-ASSISTED MOLECULAR BEAM EPITAXY

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This article reports the use of plasma-assisted molecular beam epitaxy (MBE) to grow AlN on Si(111) substrate at 850°C under UHV conditions for 15, 30, and 45 min. The films were characterized by high-resolution X-ray diffraction (HR-XRD) and micro-Raman spectroscopy. XRD measurement revealed that the AlN was epitaxially grown on Si(111). Micro-Raman result showed that all the allowed Raman modes of AlN and Si were clearly visible. Fourier transform infrared (FTIR) spectroscopy has been used to investigate the A_1 (LO) and E_1 (TO) modes with frequencies (890–899) cm^{-1} and (668–688) cm^{-1}, respectively. The results are in good agreement with reported phonon frequencies of AlN grown on Si(111).

Keywords: AlN; Si; MBE; FTIR.

1. Introduction

Aluminum nitride (AlN) (wurtzite structure type) has specific physical properties like high thermal conductivity (91–190 W/m-K), high electrical resistivity (10^{11}–10^{14} Ωcm), high melting point and large energy gap.[1–6] Its applications as a component of refractory ceramics or buffer layers for GaN epilayers grown on sapphire are widely known. The structural properties of each component of such materials and devices is a factor influencing properties of the product. Therefore, detailed studies of the structural properties may be helpful in design and improving the technologies involving AlN. In the last decade considerable interest arose in the use of thin films of AlN for various applications, from hard coatings and over coatings for magneto-optic media, to thin film transducers and GHz-band surface acoustic wave devices.

AlN has been extensively studied as thin film surface acoustic wave (SAW) material. Thin film SAW devices have an advantage to be integrated with acoustic devices or other electronic devices on the same substrate. Compared to ZnO thin film deposited on the GaAs substrate, AlN thin film deposited on the Si substrate has higher SAW velocity by 2000 m/s in spite of its low piezoelectric constant.

Thin film AlN with good preferred orientation can be grown with chemical vapor deposition (CVD), pulse laser deposition method, or sputtering method. CVD method needs high temperature process at higher than 1000°C and thus leads to high grain

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growth rate, which results in high surface roughness, high lattice mismatch, and increasing stress due to the difference in coefficient of thermal expansion. Sputtering method can be used to deposit AlN with good SAW properties at low temperature.

For this study, we present the growth of AlN on Si(111) substrate by using plasma-assisted MBE (Veeco Gen II) for different durations. The structural and optical properties of grown AlN are then analyzed by a variety of characterization tools. High-resolution X-ray diffractometer is used to assess and determine the crystalline quality. The optical quality of the film is investigated by micro-Raman and Fourier transform infrared (FTIR) spectroscopy.

2. Experimental

AlN was grown on Si(111) substrate using Veeco model Gen II MBE system. MBE system is based on ultra high vacuum (UHV) environment that is typically evacuated to less than 10^{-10} Torr. In our new MBE system, the background pressure of the MBE system was below 5×10^{-11} Torr.

Nitrogen with 7 N purity was channeled to radio frequency (RF) source to generate reactive nitrogen species. The plasma was operated at typical nitrogen pressure of 1.5×10^{-5} Torr under a discharge power of 300 W. The base pressure in the system was below 7×10^{-9} Pa. High purity material sources such as gallium (7 N) and aluminum (6N5) were used in the Knudsen cells. The nitrogen flux through plasma source, causing a nitrogen partial pressure in the MBE chamber, is about 2.0×10^{-3} Pa during growth.

The growth of III-nitrides on three-inch Si(111) substrate starts with the standard cleaning procedure by using RCA method. The substrate was then mounted on wafer holder and loaded into the MBE system. The Si substrate was outgassed in the load-lock and buffer chambers. After outgassing, the Si was transferred to the growth chamber. Prior to the growth of the epilayers, surface treatment of the Si substrate was carried out to remove the SiO_2. Si substrate was heated at 750°C, a few monolayers of Ga were deposited on the substrate for the purpose of removing the SiO_2 by formation of GaO_2. Reflection high energy electron diffraction (RHEED) showed the typical Si(111) 7×7 surface reconstruction pattern with the presence of prominent Kikuchi lines, indicating a clean Si(111) surface.

Before the growth of nitride epilayers, a few monolayers of Al were also deposited on the Si substrate to avoid the formation of Si_xN_y which is deleterious for the growth of the subsequent epilayers. The formation of such amorphous layer has been observed by many groups.[7,8] To grow AlN buffer layer, substrate temperature was heated up to 850°C, both of the Al (cell temperature at 1120°C) and N shutters were opened simultaneously.

In this work, MBE was used to grow AlN on Si(111) substrate for 15, 30, and 45 min. To evaluate the crystalline structure and the quality of the samples, high-resolution PANalytical X'Pert Pro MRD XRD system was used.

Micro-Raman measurement was performed at room temperature using Jobin Yvon HR800UV system. Argon ion (514.5 nm) laser was used as excitation sources for Raman measurement. To focus the laser on the sample surface, microscope objective lenses 100× was employed for Raman measurement. The diameter of the laser spot on the samples was around 20 μm. The emitted light was dispersed by a double-grating monochromator with 0.8 m focal length and equipped with 1800 grove/mm holographic plane grating. Signals were detected by a Peltier cooled charge-coupled device (CCD) array detector. Before the micro-Raman, high quality single crystal silicon sample (with the zone-center-mode at 520.70 cm^{-1}) was used to calibrate the system. The full width at half-maximum (FWHM) of the Si Lorentzian peak width was $\sim 3\,cm^{-1}$. The essential parameters (peak position and full width at half maximum (FWHM)) of the Raman peak were determined by using curve fitting software with Gaussian and Lorentzian model.

3. Results and Discussion

To determine the exact orientation relationship and the content of the sample, high-resolution PANalytical X'Pert Pro MRD XRD system with a Cu-Kα_1 radiation source ($\lambda = 1.5406$ Å) was used. The 2θ-reflection occurs at about 28.4° and 36.1° corresponds to diffraction peaks of (111) Si and (0002) AlN film, respectively; shown in Fig. 1. These results suggest that the AlN films were in wurtzite phase. However, the growth duration has no significant effect on the full width at half maximum (FWHM) of AlN peak. The rocking

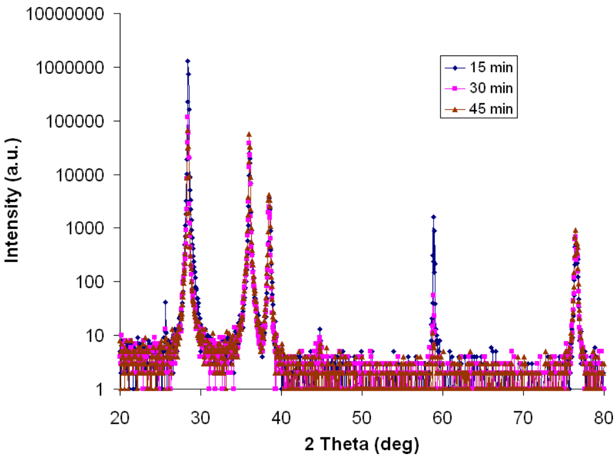

Fig. 1. XRD pattern of AlN thin film deposited on Si(111). Color online.

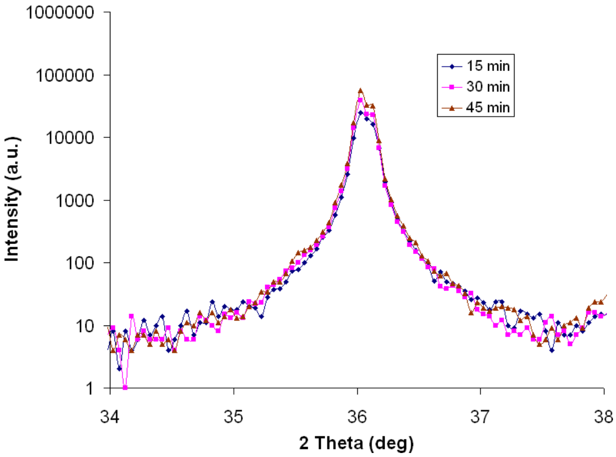

Fig. 2. XRD pattern of AlN thin film. Color online.

curves measured at 2θ-reflections are shown in Fig. 2.

The Raman scattering experiments were carried out in the $z(x, \text{unpolarized})\bar{z}$ scattering configuration. Here, the Porto's notation is used for scattering geometries with z to be parallel to wurtzite c axis, and (x, unpolarized) refers to the polarization of the incident and scattered light. Under this configuration, the allowed zone-center phonon modes of wurtzite GaN will be: E_2 (low), E_2 (high), and A_1 (LO). The E_2 (high) modes in the Raman spectra can be used to estimate the stress because it has been proven to be particularly sensitive to biaxial stress in samples.

Figure 3 shows the corresponding room temperature micro-Raman spectrum of AlN on Si. It is

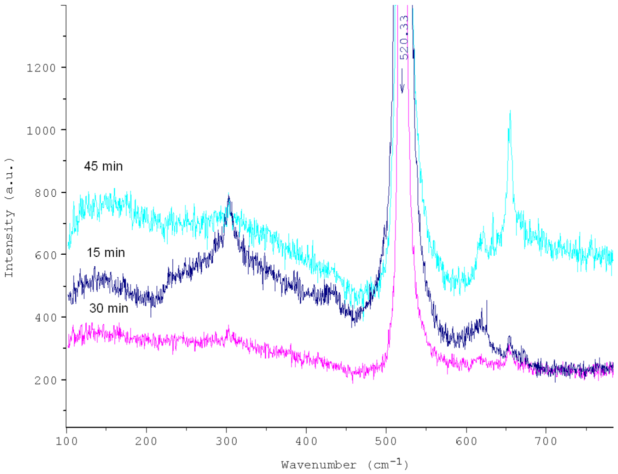

Fig. 3. Room temperature micro-Raman spectra of AlN grown on silicon substrate. Color online.

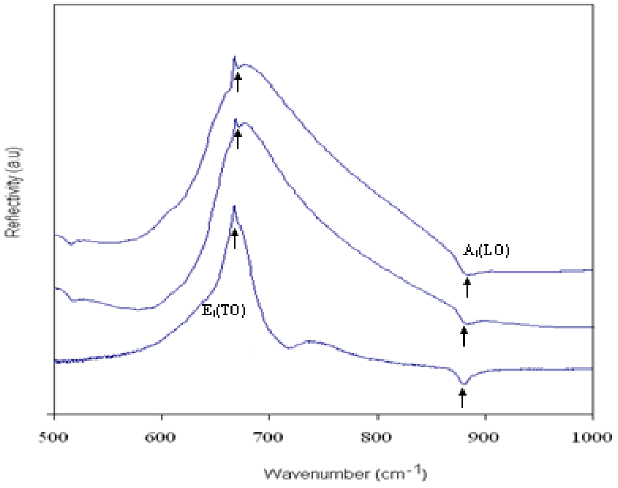

Fig. 4. FTIR phonon energies of AlN films on Si(111) with different thicknesses at (a) 15 min, (b) 30 min, and (c) 45 min.

found that all the allowed Raman phonon modes of AlN, i.e. the E_2 (low), E_2 (high), and A_1 (LO) are clearly visible. The E_2 (high) phonon mode of AlN appears at 654.40 cm^{-1} and deviates from the standard value of 655 cm^{-1} for an unstrained AlN.[10,11] In addition, a peak from the silicon substrate is also seen at 520.33 cm^{-1}, and a band at 300 cm^{-1} due to the acoustic phonons of Si. There are no phonon modes related to the cubic phase AlN to be observed in Raman spectra of our AlN/Si. These provide a further confirmation together with X-ray diffraction (XRD) measurements on the wurtzite structural nature of our MBE grown AlN. FTIR spectroscopy has been used to investigate the A_1 (LO) and E_1 (TO) modes with frequencies (890–899) cm^{-1} and (668–688) cm^{-1}, respectively; shown in Fig. 4. The results are in good agreement with reported phonon frequencies of AlN grown on Si(111).

4. Conclusion

Preferential (0001)-oriented AlN on Si(111) substrates have been performed using plasma-assisted MBE. The structural and optical properties of the thin film have been analyzed by HR-XRD. FTIR spectroscopy has been used to investigate the A_1 (LO) and E_1 (TO) modes with frequencies (890–899) cm^{-1} and (668–688) cm^{-1}, respectively; shown in Fig. 4. The results are in good agreement with reported phonon frequencies of AlN grown on Si(111).

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