PA-MBE growth and characterization of high Si-doped AlGaN on Si (111) substrate

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N-type AlGaN/GaN/AlN heterostructure have been successfully grown by plasma-assisted molecular beam epitaxy (PA-MBE) on Si (111) substrate. The structural, optical and electrical properties of the film have been studied and investigated. X-ray diffraction (XRD) measurement reveals that the AlGaN/GaN/AlN was epitaxy grown on Si substrate with high Al mole fraction equal to 0.257 and FWHM equal to 0.528. PL spectrum exhibited a sharp and intense peak at 363.16 nm with the absence of yellow emission band, indicating good crystal quality. Raman scattering measurement reveals that the optical phonon modes of A_1(Lo) and E_2(H) of the AlGaN show one mode and two modes behavior, respectively. Hall effect measurement shows that the film was highly doped with carrier concentration of 3.93 x $10^{19}cm^{-3}$.

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1. Introduction

Fabrication of the AlGaN/GaN heterostructure is one of the most basic and important issues in the field of group-III nitrides. This heterostructure can be used in many applications, such as in light-emitting diodes, laser diodes, heterojunction bipolar transistors, and UV photodetectors. AlGaN systems are especially suitable for these applications since they cover wavelengths from 200 to 364 nm. However, it is rather difficult to develop high conductivity n-type and p-type AlGaN alloys with high Al fraction, which are necessary for achieving high performance in UV devices [1, 2]. Several groups have used different methods for the Si doping of n-type AlGaN [3]. But, it is still a significant challenge to achieve good optical and electrical properties for n-type AlGaN grown on Si substrate with high mole fraction [4]. On the other hand, high density defects, which can compensate for the dopants, come into being when increasing the Al fraction [5].

In this paper, the structural, optical and electrical properties of Si-doped $Al_xGa_{1-x}N$/GaN/AlN films on Si (1 1 1) substrate grown by plasma-assisted molecular beam epitaxy (PA-MBE) are studied. Reflection high energy electron diffraction (RHEED), scanning electron microscopy (SEM), high-resolution X-ray diffraction (XRD), photoluminescence (PL), Raman spectroscopy and Hall effect measurement are used to investigate reconstruction pattern, surface morphology, Al mole fraction and crystalline quality, energy band gap, optical phonon modes and carrier concentration of the heterostructure, respectively.

2. Experiment

The III- nitrides heterostructure of n-type $Al_xGa_{1-x}N$/GaN/AlN was grown on Si (111) substrate using Veeco model Gen II MBE system. High purity material sources such as gallium (7N) and aluminum (6N5) were used in the Knudsen cells. Nitrogen with 7N 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 300W. The growth of III-nitrides on 3-inch Si (111) substrate has started with the standard cleaning procedure by using RCA method. The substrate was then mounted on the 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. Then Si substrate was heated at 850°C, and 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 buffer or wetting layer, AlN was first grown on the Si substrate. It is well known that this buffer layer plays an important role in determining the crystalline quality of the thin film [6].

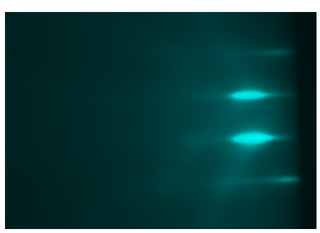
To grow AlN buffer layer, the substrate temperature was heated up to 860°C, both of the Al and N shutters were opened simultaneously for 15 minutes. Subsequently, GaN epilayer was grown on top of the buffer layer for 25 minutes with substrate temperature set at 845°C. Right after the growth of GaN epilayer, the substrate temperature was ramped down to 860°C to prepare for the growth of AlGaN epilayer. To grow Si-doped $Al_xGa_{1-x}N$, the effusion cells of Al, Ga and Si were heated up to 1003, 920 and 1300°C, respectively.

The MBE grown $Al_xGa_{1-x}N$/GaN/AlN heterostructure thin films were characterized by a variety of tools. Scanning electron microscopy (JSM – 6460 LV) model was used to observe the surface morphology of the sample. The crystalline structural and quality as well as the Al mole fraction were investigated by high-resolution PANalytical X'Pert Pro MRD x-ray diffraction (XRD) system with a Cu $K_{\alpha 1}$ radiation source (1.5406 Å). The optical properties of the sample were examined by using Haribc Jobin Yvon HR 800 UV system. For PL measurement, a 325nm He-Cd laser was used as an excitation source, while for Raman scattering experiment a 514.5 nm line of Ar ion laser was used. The incident laser power was 20 mW. The electrical property of the sample was characterized by means of HL5500PC system. The measurement was performed at room temperature with Van der Pauw method.

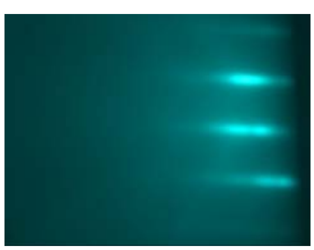
3. Results and discussion

Fig. 1(a–c) shows the RHEED images for the growth process of AlGaN on Si (111) substrate.

(a)



(b)



(c)

Fig. 1. RHEED patterns during the growth of (a) Al pattern (shutter open for 15 min), (b) Ga pattern (shutter open for 25 min and (c) AlGaN patter (shutter open for 20 min).

Fig. 2 shows the SEM image of the AlGaN/GaN/AlN layers grown on Si (1 1 1) substrate.

The AlGaN sample exhibits a smooth and clean surface morphology. However, no resolvable microstructure and no clear grain boundaries can be seen from SEM image. This information is an indication of Frank–Van der Merwe growth mode (layer-bylayer or two-dimensional growth).

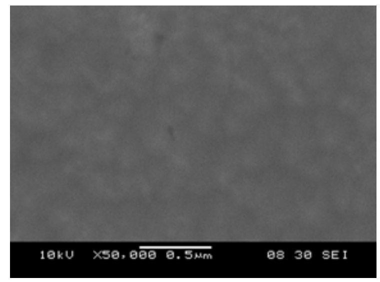


Fig. 2. SEM surface morphology of the AlGaN/GaN/AlN layers grown on Si (1 1 1) substrate.

First, a few monolayers of Al were deposited on Si (111) substrate. Then, AlN buffer layer was grown and a stable pattern which is different from Si (1 1 1) was obtained as shown in Fig. 1 (a). Fig. 1 (b) and (c) shows RHEED pattern changes during GaN and AlGaN growth. Intensity become sharp indicting the improvement of the crystalline quality of AlGaN relative to GaN layer.

Fig. 3 shows the XRD spectrum of AlGaN/GaN/AlN hetrostructure. The XRD result confirmed that the heterostructure of III-nitrides were epitaxial grown on Si (1 1 1). This can be seen from the presence of the peaks at 34.53°, 34.86° and 36.02°, which correspond to GaN(0002), AlGaN (0002) and AlN(0002) diffraction peaks, respectively. From the XRD spectrum, the lattice constant *c* for the GaN and AlN layers as determined by using the Bragg diffraction law is about 5.1893 and 4.982 Å, respectively. These values are in good agreement with the literature values, i.e., the lattice constant *c* for the bulk GaN and AlN, are respectively, 5.186 and 4.978 Å [7]. This leading us to conclude that the layers are fully relaxed.

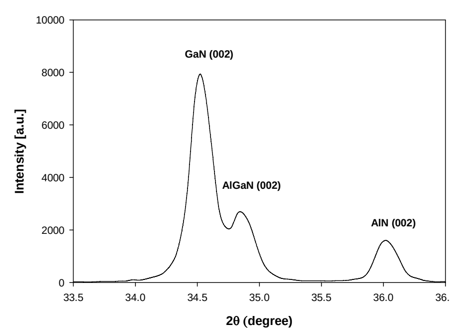


Fig. 3. XRD spectrum of the of n-type $Al_xGa_{1-x}N$sample taken from the (0002) diffraction plane and measured by the 2θ–ω scan mode.

XRD rocking curve (RC) was also carried out to determine the crystalline quality of the epilayers as shown in Fig. 4. From the XRD symmetric RC ω scans of (0002) AlGaN plane and applying the Vegard's law, the Al-mole fraction and FWHM of $Al_xGa_{1-x}N$ sample are found to be 0.257 and 0.528, respectively [8]. The results revealed a high Al-mole fraction with a good crystalline quality of $Al_xGa_{1-x}N$ thin film layer. This is in contrast to the literature [4, 9, 10] which reported that the growth of III-nitrides on Si (1 1 1) produce relatively low crystalline quality when the Al-mole fraction increases.

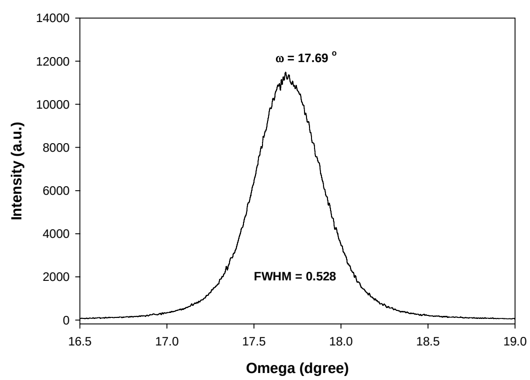


Fig. 4. XRD symmetric RC ω scans of (0002) plane for the $Al_xGa_{1-x}N$ sample.

Fig. 5 shows the PL spectrum of the sample. The PL spectrum is dominated by an intense and sharp peak at 363.16 nm which is attributed to the band edge emission of GaN. The band edge emission of $Al_{0.257}Ga_{0.743}N$ (which is estimated to be 4.119 ev) was not obtained due to the limitation of the excitation source used in this study. No yellow band emission was observed either; this indicates that the thin film is of good optical quality.

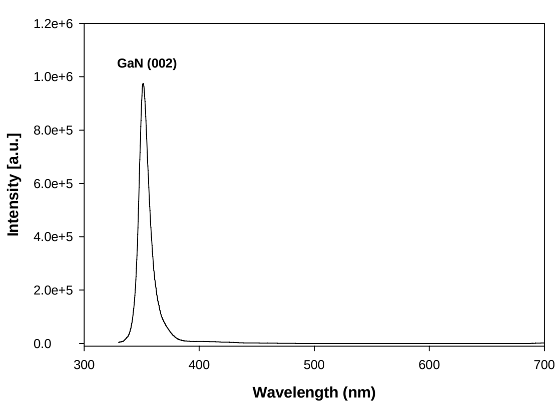


Fig. 5. PL spectrum of the $Al_xGa_{1-x}N$/GaN/AlN grown on Si (111) substrate.

Fig. 6 shows the Raman spectrum of AlGaN/GaN/AlN/Si. The Raman scattering experiments were carried out using $z(x, \mathrm{unpolarized})\bar{z}$ scattering configuration, with z parallel to the c axis. Under this configuration, the allowed zone-center phonon modes that can be detected for wurtzite structure layer will be A_1(LO), E_2(Low), and E_2(High).

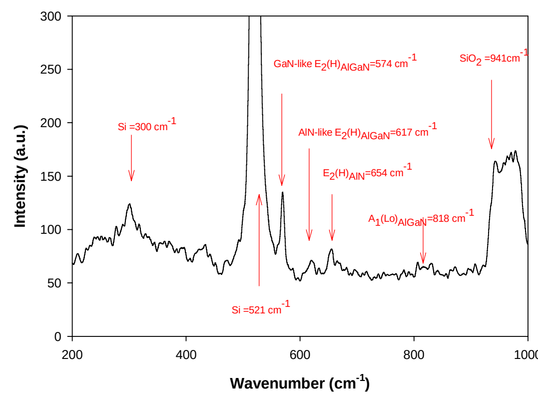


Fig. 6. Room temperature Raman spectrum of n-type $Al_xGa_{1-x}N$/GaN/AlN grown on Si (111) substrate.

From Fig. 6, peaks at 300 and 521 cm^{-1}, correspond to Si modes. Peak at 818 cm^{-1}, correspond to A_1(LO) one mode for AlGaN. While peaks at 574 cm^{-1} and 617 cm^{-1} are correspond to GaN-like $E_2(H)_{AlGaN}$ and AlN-like $E_2(H)_{AlGaN}$ [11] [11]. Result from Hall effect measurement shows that n-type carrier concentration equal to 3.93 x $10^{19} cm^{-3}$ for the AlGaN layer is obtained. This value of carrier concentration is considered higher than the value reported in [12] [12].

4. Conclusions

High Al-mole fraction and high Si-doped n-type AlGaN/GaN heterostructure have been successfully growth by PA-MBE on Si substrate. The structural, optical and electrical properties of the thin film have been analyzed by SEM, HR-XRD, PL, Raman scattering and Hall effect measurement. Good surface morphology and high Al-mole fraction of$Al_xGa_{1-x}N$ epilayer with good crystalline quality has been obtained. In addition, PL measurement has exhibited a sharp and intense band edge emission of GaN with the absence of yellow emission band which is indicative of good crystal quality of the thin film. Raman result revealed that the A_1(LO) and E_2(H) optical phonon modes of AlGaN exhibited one mode and two mode behavior, respictevly. Results of Hall effect measurement revealed that our sample has a high carrier concentration of 3.93×10^{19} cm^{-3}.

Acknowledgements

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