

# Structural properties of high Ga-content $\text{Al}_x\text{Ga}_{1-x}\text{N}$ on Si substrate studied by high resolution X-ray diffraction and photoluminescence

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The high Ga-content AlGa<sub>x</sub>N thin films are grown on GaN/AlN/silicon (111) substrate by plasma assisted molecular beam epitaxy (PA-MBE). The full width at half-maximum (FWHM) of the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  alloys deposited on silicon as determined by XRD symmetric RC  $\omega/2\theta$  scans of (0002) plane at room temperature is  $0.52^\circ$ . The Al mole-fraction of this sample as deduced from Vegard's law and HR-XRD measurement is 0.29. Photoluminescence (PL) spectrums of sample have shown sharp and intense band edge emission of GaN without the existence of yellow emission band, showing good crystal quality of the samples have been successfully grown on Si substrate.

(Received March 31, 2010; accepted May 20, 2010)

*Keywords:* AlGa<sub>x</sub>N/Si, AlGa<sub>x</sub>N/GaN, III-Nitride

## 1. Introduction

AlGa<sub>x</sub>N alloys are extremely important materials with widespread applications for optoelectronic devices because they have a direct wide energy bandgap, which ranges from 6.2 to 3.4 eV. Due to their wide band gap range, the alloys are very attractive materials for applications in ultraviolet (UV) laser diodes (LDs), light emitting diodes (LEDs) and photodetectors [1-3]. The AlN buffer layer grown on Si (111) showed smaller full width at half maximum (FWHM) of AlN (0002) peak, compared to AlN grown on Si (100) [4]. This is because the preferred orientation of AlN films on Si (111) substrate is more easily controlled than those on Si (100) substrates. In the case of GaN layers grown on AlN buffer layer, the crystalline quality is much improved because the lattice mismatch between GaN and AlN is only 2.5%. For these reasons, the Si (111) substrate was used. In this paper, we report on the growth of AlGa<sub>x</sub>N thin films sample on Si(111) substrate, using high-temperature-grown GaN/AlN as a buffer layer by plasma assisted molecular beam epitaxy (PA-MBE). The structural qualities of the sample will be investigated by using high resolution X-ray diffraction (HR-XRD) and photoluminescence (PL) spectroscopy.

## 2. Experimental

The AlGa<sub>x</sub>N(0.225 $\mu\text{m}$ )/GaN(0.081 $\mu\text{m}$ )/AlN(0.031 $\mu\text{m}$ ) was grown on Si (111) substrate using Veeco model Gen II MBE system (See Figure 1). High purity material sources such as gallium (7N), aluminum (6N5) and indium (7N) 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  $1.5 \times 10^{-5}$  Torr under a discharge power of 300W. The Si substrate was firstly heated at 900°C, and a few monolayers of Ga were

deposited on the substrate for the purpose of removing the SiO<sub>2</sub> by formation of GaO<sub>2</sub>. Reflection high energy electron diffraction (RHEED) showed the typical Si (111)  $7 \times 7$  surface reconstruction pattern with the existence of prominent Kikuchi lines, showing a clean Si (111) surface. The AlN buffer layer was also first grown on the Si substrate for 13 minutes with substrate temperature set at 875°C. After that, GaN epilayer was grown on top of the buffer layer for 20 minutes with substrate temperature set at 840°C. To grow AlGa<sub>x</sub>N, the effusion cells of Al, and Ga were heated up to 1005 and 923°C, respectively. During the growth of AlGa<sub>x</sub>N, the substrate temperature was set at 861°C.

In situ reflection high-energy electron diffraction (RHEED) will be used to investigate reconstruction pattern of the sample. The MBE grown sample was characterized by a HR-XRD and PL system. HR-XRD with a Cu-K $\alpha$ 1 radiation source ( $\lambda = 1.5406 \text{ \AA}$ ) was used to assess and determine the crystalline quality of the epilayers. For PL system, a He-Cd laser with emission wavelength of 325 nm is used as the excitation source, in order to study the band gap and quality of these alloys.

AlGa <sub>x</sub> N @ 861 <sup>0</sup> C @ 0.225 $\mu\text{m}$
GaN @ 840 <sup>0</sup> C @ 0.081 $\mu\text{m}$
AlN @ 875 <sup>0</sup> C @ 0.031 $\mu\text{m}$
Si (111)

Fig. 1. Cross-section structure of AlGa<sub>x</sub>N/GaN/AlN/Si substrate.

### 3. Results and discussion

The growth mode of the AlGa<sub>N</sub> layers was studied by RHEED. Fig. 2 shows the schematic of the evolution the AlGa<sub>N</sub> layer with corresponding RHEED. The Si substrate surface showed a clear surface reconstruction at high temperature as shown in Fig. 2(a). Starting from a smooth AlN buffer layer [Fig. 2(b)], the RHEED image remains streaky during initial stage of GaN growth. After the growth of GaN, RHEED displayed a streaky pattern indicative of good surface morphology as revealed in Fig. 2(c). During the growth of GaN (Fig. 2(c)), the streaky RHEED pattern is sharpened, suggesting the improvement of the crystalline quality of GaN relative to the AlN buffer layer. In Fig. 2(d) AlGa<sub>N</sub> growth under low temperature condition follows a Stranski-Krastanow growth mode, leading to AlGa<sub>N</sub> on top of a continuous wetting layer.

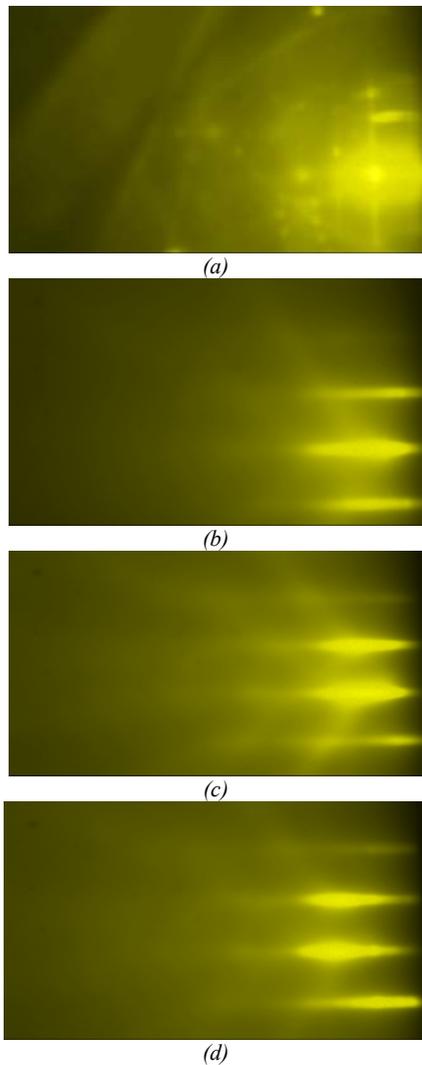


Fig. 2. RHEED pattern for the growth process of AlGa<sub>N</sub> layer in GaN/AlN/Si(111).

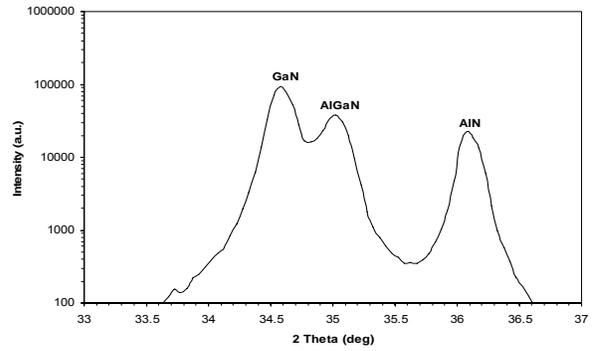


Fig. 3. XRD spectrum of the sample II taken from the (0002) diffraction plane.

Fig. 3 showed that sample has also been successfully grown on silicon substrate and has been confirmed from the presence of the peaks at  $34.575^\circ$ ,  $35.025^\circ$  and  $36.075^\circ$ , which correspond to GaN (0002), AlGa<sub>N</sub> (0002) and AlN (0002), respectively. XRD symmetric RC  $\omega/2\theta$  scans of (0002) plane at room temperature was also conducted to verify the crystalline quality of thin films. The full width at half-maximum (FWHM) of AlGa<sub>N</sub> was  $0.528^\circ$ , which can be seen from Fig. 4.

From the XRD symmetric RC  $\omega/2\theta$  scans of (0002) plane, the lattice parameter  $c$  of the samples can be calculated using the following formula:

$$c = \frac{\lambda l}{2 \sin \theta_{RC}} \quad (1)$$

where  $\lambda$  is the wavelength of the x-ray radiation (1.5406 Å),  $\theta_{RC}$  is the Bragg angle estimated from the peak of the RC, and  $l$  is the Miller indices. In principle, the composition can be calculated through XRD measurements and application of Vegard's law. By assuming the layers are fully relaxed or fully strained, according to Vegard's law, the variation of the lattice constant  $c$  between GaN and AlN is linearly proportional to the Al mole fraction [5]. Based on the obtained  $c$  value, the mole composition of the Al,  $x$ , can be determined by the following formula [6]:

$$x = \frac{(c_{AlGaN} - c_{GaN})}{(c_{AlN} - c_{GaN})} \quad (2)$$

where  $c_{GaN}$ ,  $c_{AlN}$  and  $c_{AlGaN}$  are the lattice constants of GaN, AlN, and AlGa<sub>N</sub>, respectively. From equations (1) and (2), the mole fraction  $x$  of AlGa<sub>N</sub> can be calculated. According to this law and equation (1) and (2), Al mole-fraction was recorded as 0.29 for AlGa<sub>N</sub> thin film. It can be seen that a sample has indicated a better crystalline quality, and then it agreed to [7-9], which revealed that good crystalline quality of III-nitrides materials have been produced when the Al-mole fraction decreases [7-9]. However, the crystalline qualities of the epilayer are not only Al-mole fraction dependent but also growth parameters dependent [10].

The use of silicon (111) substrate for growth of III-nitrides, particularly GaN, always produces relatively low crystal quality; therefore it is difficult to grow high quality GaN-based materials on silicon (111) substrate [11-13]. The large difference in lattice constant, crystal structure and thermal expansion coefficient between the silicon and GaN-based materials are believed to be a several factors which contributed to the poor crystal quality of the GaN-based epilayers [14].

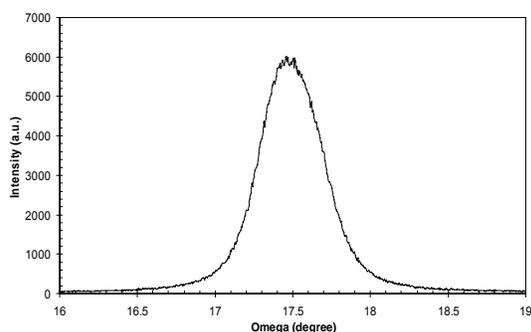


Fig. 4. XRD symmetric RC  $\omega/2\theta$  scans of (0002) plane for sample I and sample II (inset).

Fig. 5 shows the PL spectrum of the sample. The PL spectrums are dominated by intense and sharp peaks at 362.31 nm, which are attributed to the band edge emission of GaN. The band edge emissions for AlGaN were not obtained due to the limitation of the excitation source used in this study. No yellow band emissions were observed either; these indicate that the thin films are of good optical quality.

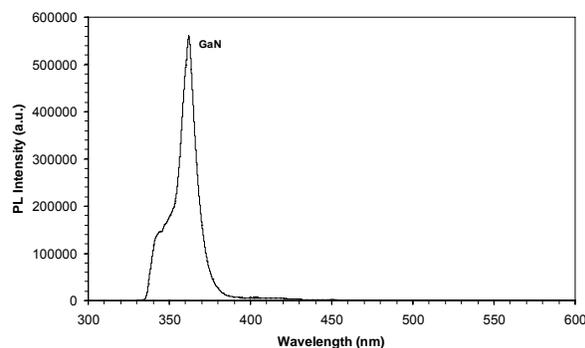


Fig. 4. PL spectra of the AlGaN/GaN/AlN/Si(111).

#### 4. Conclusions

The growth of AlGaN/GaN/AlN samples on Si(111) substrate have been performed successfully using plasma-assisted molecular beam epitaxy. The structural qualities of the thin films have been analyzed by HR-XRD and PL measurements. By using the rocking curve measurement, the full width at half maximum (FWHM) value of the sample is  $0.528^\circ$ . By the application of Vegard's Law, the Al-mole fractions of 0.29 were obtained for the sample.

PL measurements for sample exhibited sharp and intense band edge emission of GaN with no yellow emission band indicating good optical quality of the thin films.

#### Acknowledgments

The support from Universiti Sains Malaysia is also gratefully acknowledged.

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