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HR-XRD and PL studies of $Al_xGa_{1-x}N$ layers grown on Si (111) substrate by plasma assisted MBE

M. Z. Mohd YUSOFF [a,b,*], Z. HASSAN[a], H. Abu HASSAN[a], Y. YUSOF[a], M. J. ABDULLAH[a]
[a]*Nano-Optoelectronics Research and Technology Laboratory, School of Physics, Universiti Sains Malaysia, 11800 Penang, Malaysia*
[b]*Department of Applied Sciences, Universiti Teknologi MARA (UiTM), 13500 Permatang Pauh, Penang, Malaysia*

The aluminum gallium nitride (AlGaN) layers are grown on silicon (Si) (111) substrate by plasma assisted molecular beam epitaxy (PA-MBE) on top of a GaN/AlN buffer in order to reduce the strain of the alloy. Two samples of AlGaN layers were grown on GaN/AlN/Si(111) with different growth conditions. The full width at half-maximum (FWHM) of the two $Al_xGa_{1-x}N$ alloys deposited on silicon as determined by X-ray diffraction (XRD) symmetric rocking curve (RC) ω/2θ scans of (0002) plane at room temperature are 0.62^0 and 0.52^0, respectively. The aluminum (Al) mole-fractions of these samples as deduced from Vegard's law and high resolution X-ray diffraction (HR-XRD) measurement are 0.11 and 0.29, respectively. Photoluminescence (PL) spectrums of both samples have shown sharp and intense band edge emission of gallium nitride (GaN) without the existence of yellow emission band, showing good crystal quality of the samples have been successfully grown on Si substrate.

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1. Introduction

III-nitrides have shown great potential for high power, high-frequency electronic devices [1] and short-wavelength optical devices [2] such as light emitting diodes, lasers, and photodetectors. The epitaxial growth of GaN on Si substrates is of particular interest because one can take advantage of the well-established Si technologies in developing GaN ones. Moreover, the availability of large and high quality Si substrates, as well as its low cost, makes it an attractive alternative for the growth of III-nitride layers [3].

AlGaN 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 [4-6].

Based on the previous reports, by using X-ray diffraction (XRD) measurement, AlN grown on Si (111) showed smaller full width at half maximum (FWHM) of AlN (0002) peak, compared to AlN grown on Si (100) [7]. This is because the preferred orientation of AlN films on Si (111) substrate is more easily controlled than those on Si (100) substrates. The lattices in AlN (0002) and Si (111) are both hexagonal, and thus Si (111) can provide matched template for AlN (0002) plane. The lattice mismatch between these two plane is 19% only. However, the lattice in Si (100) is square, which is unmatched with hexagonal lattice in AlN (0002) plane. The lattice mismatch between AlN (0002) and Si (100) is 42.7%. The larger lattice mismatch between AlN (0002) and Si (100) is a main contribution to the larger strain in the formed films. 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.

X-ray diffraction (XRD) is a non-destructive technique which is widely used in material characterization, particularly to determine structure, crystalline quality and orientation of samples planes. As the name implies, the underlying principle behind the X-ray diffraction technique is diffraction. When X-ray radiation is directed on a crystal, diffraction occurs because its wavelength is the range of interatomic distances (0.1-10 Å).

Photoluminescence (PL) spectroscopy is a non-destructive method of probing the the electronic structure and quality of materials. It is effective for optical evaluation of semiconductors with relatively wide band gap. Usually, a laser with energy above the band gap of a semiconductor is used as the excitation source to promote electronic transitions from a lower energy level to a higher energy level. When an electron in an excited state recombines with a hole in a lower electronic state, there are two process that may occur: a radiative process (luminescence) or a non-radiative process (phonon emission).

In this paper, we report on the growth of two AlGaN thin films samples on Si(111) substrates with different Al-mole fraction, using high-temperature-grown GaN/AlN as a buffer layer by plasma assisted molecular beam epitaxy (PA-MBE). The structural qualities of the samples will be investigated by using high resolution X-ray diffraction (HR-XRD) and photoluminescence (PL) spectroscopy.

2. Experimental methods

Two AlGaN thin films were grown on Si (111) substrate using Veeco model Gen II MBE system. Home grown AlGaN(~0.0001 µm)/GaN(0.240 µm)/AlN(0.090 µm) and AlGaN(0.225 µm)/GaN(0.081 µm)/AlN(0.031 µm) have been assigned as sample I and sample II, respectively (see Fig. 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 1.5×10^{-5} Torr under a discharge power of 300 W. 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_2 by formation of GaO_2. Reflection high energy electron diffraction (RHEED) showed the typical Si (111) 7×7 surface reconstruction pattern with the existence of prominent Kikuchi lines, showing a clean Si (111) surface.

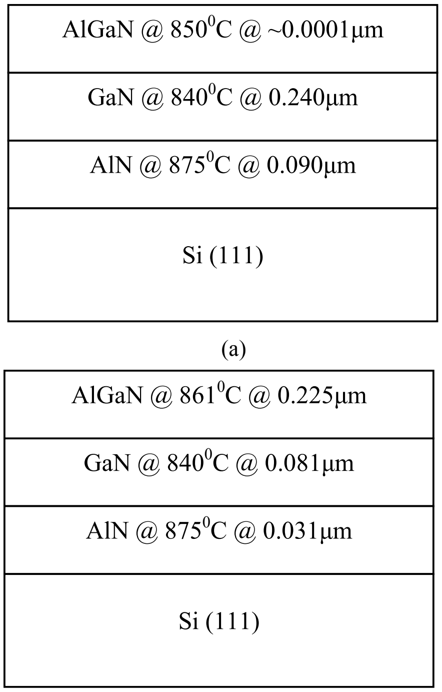

Fig. 1. Cross-section structures of (a) sample I and (b) sample II.

For sample I, the buffer or wetting layer, AlN was first grown on the Si substrate. To grow AlN buffer layer, the substrate temperature was heated up to 875 °C, both of the Al and N shutters were opened simultaneously for 13 minutes. Subsequently, GaN epilayer was grown on top of the buffer layer for 20 minutes with substrate temperature set at 840 °C. Right after the growth of GaN epilayer, the substrate temperature was ramped up to 850 °C to prepare for the growth of AlGaN epilayer. To grow AlGaN, the effusion cells of Al, and Ga were heated up to 1005 and 925°C, respectively (see Fig. 1 (a)).

Meanwhile, for sample II, 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 AlGaN, the effusion cells of Al, and Ga were heated up to 1005 and 923 °C, respectively. During the growth of AlGaN, the substrate temperature was set at 861 °C (see Fig. 1(b)).

The MBE grown sample I and sample II were characterized by a HR-XRD and PL system. HR-XRD with a Cu-Kα1 radiation source (λ= 1.5406 Å) 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.

3. Results and discussions

As shown in Fig. 2, the HR-XRD result validated that the AlGaN heterostructure of sample I was epitaxially grown on silicon substrate. These can be seen from the existence of the peaks at 34.375^0, 34.725^0 and 35.875^0, which match to GaN (0002), AlGaN (0002) and AlN (0002), respectively. Fig. 3 showed that sample II has also been successfully grown on silicon substrate and has been confirmed from the presence of the peaks at 34.575^0, 35.025^0 and 36.075^0, which correspond to GaN (0002), AlGaN (0002) and AlN (0002), respectively. XRD symmetric RC ω/2θ 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 AlGaN for sample I and sample II were 0.624^0 and 0.528^0, respectively, which can be seen from Figs. 2 and 3.

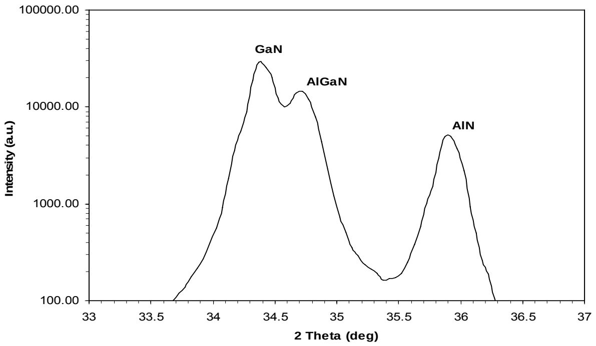

Fig. 2. XRD spectrum of the sample I taken from the (0002) diffraction plane.

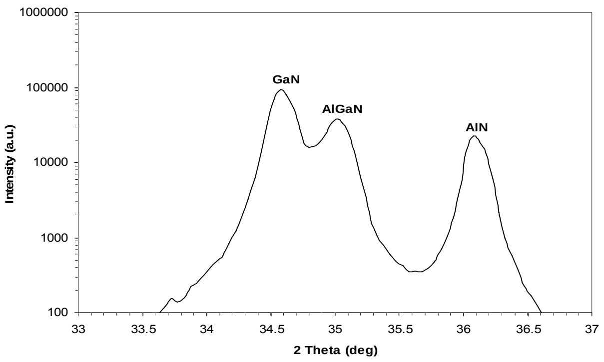

Fig. 3. XRD spectrum of the sample II taken from the (0002) diffraction plane.

From the XRD symmetric RC ω/2θ scans of (0002) plane, the lattice parameter *c* of the samples can be calculated using the following formula:

$$c = \frac{\lambda l}{2 \sin \theta_{RC}} \tag{1}$$

Where λ is the wavelength of the x-ray radiation (1.5406 Å), θ_{RC} is the Bragg angle estimated from the peak of the RC, and *l* is the Miller indices. In principle, the composition can be determined 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 [8]. Based on the obtained *c* value, the mole composition of the Al, *x*, can be determined by the following formula [9]:

$$x = \frac{\left(c_{AlGaN} - c_{GaN}\right)}{\left(c_{AlN} - c_{GaN}\right)} \tag{2}$$

where c_{GaN}, c_{AlN} and c_{AlGaN} are the lattice constants of GaN, AlN, and AlGaN, respectively. From equations (1) and (2), the mole fraction *x* of AlGaN can be calculated. According to this law and equation (1) and (2), Al mole-fractions were recorded as 0.11 and 0.29 for sample I and sample II, respectively. It can be seen that sample II (Al mole-fraction = 0.29) has indicated a better crystalline quality compared to sample I (Al mole-fraction=0.11), however it contradicts to [10-12], which revealed that low crystalline quality of III-nitrides materials have been produced when the Al-mole fraction increases [10-12]. We suspect that these are due to the crystalline qualities of the epilayer are not only Al-mole fraction dependent but also growth parameters dependent [13].

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 [14-16]. The large difference in lattice constant, crystal structure and thermal expansion coefficient between the silicon and GaN-based materials are believed to be several factors which contributed to the poor crystal quality of the GaN-based epilayers [17].

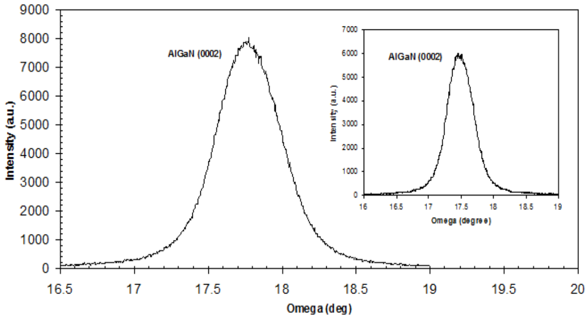

Fig. 4. XRD symmetric RC ω/2θ scans of (0002) plane for sample I and sample II (inset)

Fig. 5 shows the PL spectrum of the sample I and sample II (inset), respectively. The PL spectrums are dominated by intense and sharp peaks at 351.83 nm and 362.31 nm, respectively, 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.

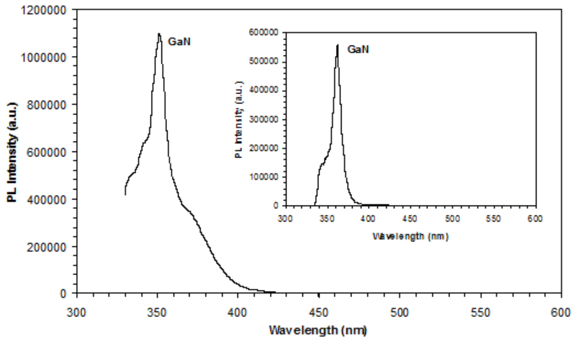

Fig. 5. PL spectra of the sample I and sample II (inset)

4. Conclusions

The growth of two 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 I and sample II are 0.624^0 and 0.528^0, respectively. By the application of Vegard's Law, the Al-mole fractions of 0.11 and 0.29 were obtained for the sample I and sample II, respectively. PL measurements for both samples exhibited sharp and intense band edge emission of GaN with no yellow emission band indicating good optical quality of the thin films.

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*Corresponding author: mzmy83@gmail.com