Paper

227-261 nm AlGaN-based Deep Ultraviolet Light-emitting Diodes Fabricated on High-quality AlN Buffer on Sapphire

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ABSTRACT

We demonstrated AlGaN multi-quantum well (MQW) deep ultraviolet (UV) light-emitting diodes (LEDs) with wavelength in the range of 227-261 nm fabricated on high-quality AlN buffers on sapphire substrates. We achieved crack-free, thick AlN buffer on sapphire with low threading dislocation density (TDD) and atomically flat surface by introducing an ammonia (NH_3) pulse-flow multi-layer (ML) growth method through metal-organic chemical vapor deposition (MOCVD). The edge- and screw-type dislocation densities of AlGaN layer on AlN buffer were reduced to 7.5×10^8 and 3.8×10^7 cm^{-2}, respectively, by using a ML-AlN buffer. We achieved single-peaked high-brightness operations of AlGaN deep-UV LEDs by fabricating them on the ML-AlN buffers on sapphire substrates. The maximum output power and external quantum efficiency (EQE) of the 261 nm and 227.5 nm LEDs were 1.65 mW and 0.23% under room-temperature (RT) CW operation, and 0.15 mW and 0.2%, under RT pulsed operation, respectively.

KEYWORDS: deep-UV LED, AlGaN, AlN, MOCVD, threading dislocation density, external quantum efficiency

1. Introduction

High-brightness deep-ultraviolet (UV) light-emitting diodes (LEDs) or laser diodes (LDs) with emission wavelengths in the range 230-350 nm have a wide range of potential applications, such as in water purification, sterilization, medicine and biochemistry, white light illumination, and light sources for high density optical recording[1]. AlGaN has a direct transition band gap between 3.4 and 6.2 eV and very attractive for realizing high-brightness deep-UV LEDs.

Research into AlGaN-based UV LEDs was initiated by several research groups between 1996-1999[2-4]. Recent advances in high-brightness deep-UV LEDs have been led by the innovations developed by the group at University of South Carolina (USC). They reported 240-280-nm-band high-brightness AlGaN-based LEDs in 2002-2006[5][6]. The shortest wavelength 210 nm AlN LED was reported by NTT in 2006[7], however, the emission efficiency was still quite low.

We reported 230 nm efficient photoluminescence (PL) from AlGaN quantum wells (QWs)[8], and 330 nm-band AlGaN-QW UV LED on SiC in 1999[2]. We have also developed quaternary InAlGaN-based high-efficiency UV LEDs[1][9][10]. Recently, we achieved 231-261 nm AlGaN QW LEDs using high-quality AlN buffer on sapphire[11].

In this report, we demonstrate a high-quality AlN buffer suitable for deep UV LEDs and also demonstrate single-peaked operations of AlGaN-QW LED with emission wavelengths of 227-261 nm. We demonstrate CW milliwatt output power of 253-261 nm LEDs. We also achieved sub-milliwatt output power of 227.5 nm LED under pulsed operation.

2. Experiments and Results

It is required to satisfy several conditions in order to fabricate high-quality AlGaN/AlN templates that are applicable to deep-UV emitters, i.e., low threading dislocation density (TDD), crack free, atomically flat surface and stable Al (+c) polarity. To satisfy all of these conditions, we proposed the ammonia (NH_3) pulse-flow multilayer (ML) AlN growth method. Several methods of fabricating high-quality buffers for the realization of UV-emitting devices have already been reported[5][12][13]. The advantage of our method in comparison with these methods is that all layers are consisting of AlN without using AlGaN layers and therefore they are free from deep-UV absorption[11].

Samples were grown on sapphire (0001) substrates by low-pressure metal-organic chemical vapor deposition (MOCVD). As group III precursors, trimethylaluminum (TMAl) and trimethylgallium (TMGa) were used with H_2 carrier gas.

Figure 1 shows (a) the gas flow sequence used for NH_3 pulse-flow growth and (b) schematic layer structure of the multilayer (ML)-AlN buffer. First, an AlN nucleation layer and a burying AlN layer were deposited, both by NH_3 pulse-flow growth. Low-TDD AlN can be achieved by the coalescence process of AlN nucleation layer. After the growth of the first AlN layer, the surface is still rough because of the low growth rate of the pulse-flow growth mode. Then, after that, we introduced the high-growth-rate continuous-flow mode in order to reduce the surface roughness. By repeating the pulse- and continuous-flow modes, we obtained a crack-free, thick AlN layer with an atomically flat surface. NH_3 pulse-flow growth is effective for obtaining high-quality AlN because of the enhancement of precursor migration. Furthermore, it is effective for obtaining the stable Al (+c) polarity that is necessary for realizing atomically flat surfaces, because of the Al-rich growth condition. We controlled the growth pressure, temperature and V/III ratio to be between 76-200 Torr, 1200-1300 °C and 64-1060, respectively, for the growth of ML-AlN. The growth rates in the pulse- and continuous-flow modes were approximately 0.6 and 6 μm/hour, respectively.

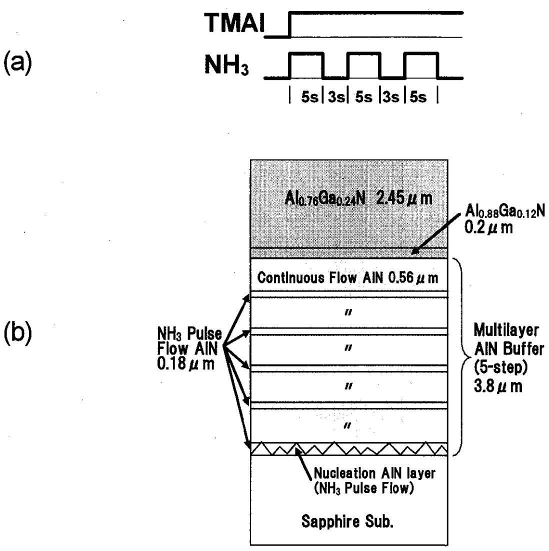

Figure 1 (a) Gas flow sequence used in NH_3 pulse-flow growth
(b) Schematic layer structure of multilayer (ML)-AlN buffer on sapphire

Figure 2 shows a cross-sectional transmission electron microscope (TEM) image of AlGaN/ML-AlN/sapphire template with the layers of 253 nm AlGaN multi (M)QW LED. We can see that the threading dislocations disappear mainly in the first burying process of AlN nucleation layer. The minimum value of the full-width at half maximum (FWHM) of the X-ray (102) ω-scan rocking curve (XRC) of ML-AlN was 371 arcsec. The minimum values of the edge-, screw- and mixed-type dislocation densities of AlGaN layer on ML-AlN were 7.5×10^8, 3.8×10^7 and 2.4×10^8 cm^{-2}, respectively, as observed from the cross-sectional TEM image.

Figure 3 shows an atomic force microscope (AFM) image of the surface of fabricated ML-AlN buffer on sapphire observed for 5μm×5μm square area. We confirmed an atomically flat surface by observation in the step-flow growth mode using an AFM. The typical RMS value obtained from the AFM was 0.16 nm.

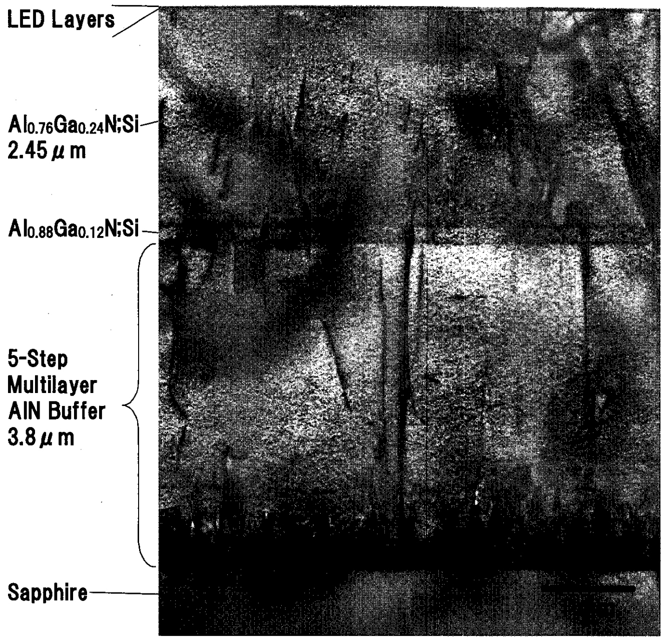

Figure 2 Cross-sectional TEM image of an AlGaN-MQW UV LED with emission wavelength of 253 nm

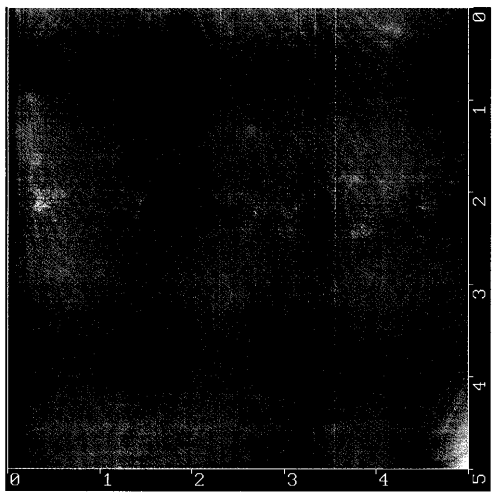

Figure 3 AFM image of the surface of the ML-AlN buffer observed for 5μm×5μm square area

Figures 4 shows a schematic of the sample structure of an AlGaN MQW deep-UV LED with emission wavelength of 227.5 nm fabricated on a ML-AlN buffer and an emission pattern image observed from back side of sapphire substrate. Table 1 shows the designed Al compositions x in $Al_xGa_{1-x}N$ wells, buffer and barrier layers, and electron-blocking layers (EBLs) used for 227.5-261nm AlGaN-MQW LEDs. We grew a 1.7-μm-thick Si-doped AlGaN-buffer-layer, 3-layer MQW consisting of 1.3-nm-thick AlGaN wells and 7-nm-thick AlGaN barriers, 21-nm-thick undoped AlGaN barrier, 15-nm-thick Mg-doped AlGaN EBL, 10-nm-thick Mg-doped AlGaN and an approximately 10-nm-thick Mg-doped GaN contact layer on the ML-AlN.

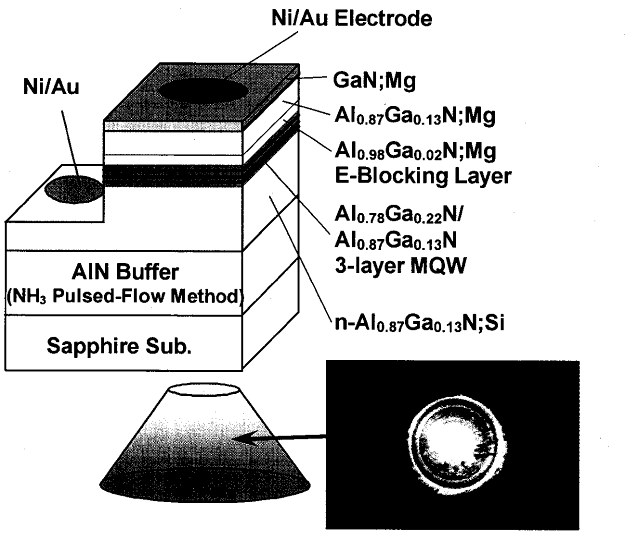

Figure 4 Schematic of the sample structure of an AlGaN MQW deep-UV LED with emission wavelength of 227.5 nm fabricated on a ML-AlN buffer and an emission pattern image observed from back side of sapphire substrate

Table1 Designed values of Al composition x of $Al_xGa_{1-x}N$ wells, buffer and barrier layers, and electron blocking layers (EBLs) used for 227.5-261 nm AlGaN-MQW LEDs

Wavelength	Well	Barrier & Buffer	Electron Blocking Layer
227.5nm	0.79	0.87	0.98
234 nm	0.74	0.84	0.97
248 nm	0.64	0.78	0.96
255 nm	0.60	0.75	0.95
261 nm	0.55	0.72	0.94

We confirmed atomically flat hetero-interfaces of the QWs from the cross-sectional TEM image.

In order to realize high-brightness deep UV-LEDs, it is necessary to achieve high internal quantum efficiency (IQE) for AlGaN-based QWs. We observed remarkable enhancement of deep UV emission from AlGaN-QWs by fabricating them on the high-quality ML-AlN templates. The photoluminescence (PL) intensity of the AlGaN-QW with wavelength between 254-280 nm was increased by approximately 30 times by reducing FWHM of (102) ω-scan XRC of AlGaN buffer on ML-AlN from 1214 to 488 arcsec.

Ni/Au electrodes were used for both *n*-type and *p*-type electrodes. The size of the *p*-type electrode was 300×300 μm. The output power radiated into the back of the LED was measured using a Si photodetector located behind the LED sample, which was calibrated to measure radiant flux from LED sources using an integrated-spheres system. The LEDs were measured under a bare wafer condition.

Figure 5 shows electroluminescence (EL) spectra of the fabricated AlGaN-MQW LEDs with wavelengths of 227.5 nm under pulsed operation and 234, 248, 255 and 261 nm under CW operations, all measured at room temperature (RT), with injection current of 30-50 mA. Pulse width and repetition frequency were 3 μs and 10 KHz, respectively. Single-peaked operations were obtained for every sample.

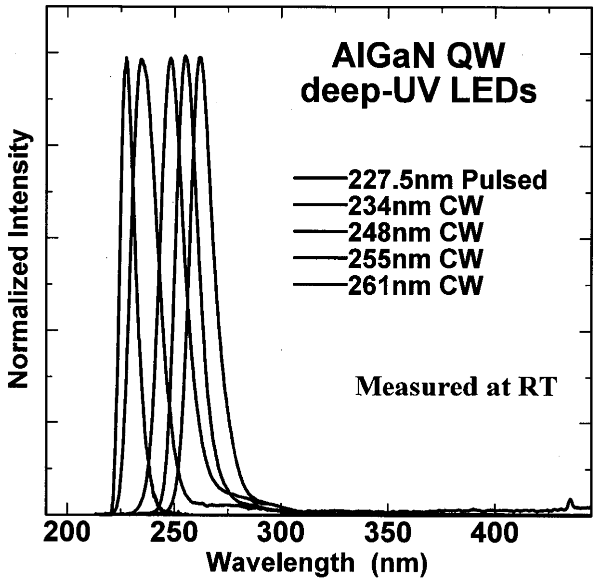

Figure 5 Electroluminescence (EL) spectra of the fabricated AlGaN-MQW LEDs with wavelengths of 227.5 nm under pulsed operation and 234, 248, 255 and 261 nm under CW operations, all measured at room temperature (RT), with injection current of 30-50 mA

Figure 6 shows (a) output power and (b) external quantum efficiency (EQE) η_{ext} for 253 nm and 261 nm AlGaN-MQW LEDs as a function of current measured under RT CW operation. The maximum output power and EQE of the 253 nm and 261 nm LEDs were 1.04 mW and 0.09%, and 1.65 mW and 0.23%, under RT CW operation, respectively. The maximum output power and EQE of the 227.5 nm LED were 0.15 mW and 0.2% under RT pulsed operation, respectively. We confirmed that the EQEs of AlGaN MQW LEDs were much higher than that of AlN LED[8]. The reason for the low EQE for shorter wavelength is considered to be that most of the injection electrons overflowed to the p-side electrode. EQE can be significantly increased if the electron overflow can be suppressed by realizing higher hole concentration in p-type layers.

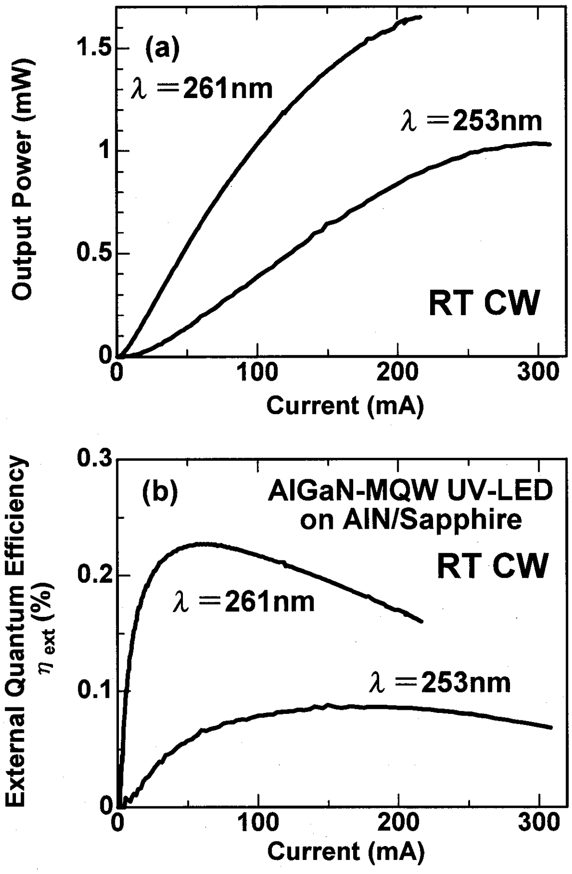

Figure 6 (a) output power and (b) external quantum efficiency (EQE) η_{ext} for 253 nm and 261 nm AlGaN-MQW LEDs as a function of current measured under RT CW operation

3. Conclusions

In conclusions, we demonstrated single-peaked operations of AlGaN-QW deep-UV LEDs on sapphire with peak emission between 227-261 nm. We achieved a high-quality AlGaN/AlN buffer suitable for use in deep-UV emitters, by introducing a ML-AlN formed by the NH_3 pulse-flow method. The maximum output power and EQE of the 261 nm and 227.5 nm LEDs were 1.65 mW and 0.23% under RT CW operation, and 0.15 mW and 0.2%, under RT pulsed operation, respectively.

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