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Investigations on Strained AlGaN/GaN/Sapphire and GaInN Multi-Quantum-Well Surface LEDs Using AlGaN/GaN Bragg Reflectors

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SUMMARY Investigations were carried out on metalorganic-chemical-vapor-deposition (MOCVD)-grown strained AlGaN/GaN/sapphire structures using X-ray diffratometry. While AlGaN with lower AlN molar fraction (< 0.1) is under the in-plane compressive stress, it is under the in-plane tensile stress with high AlN molar fraction (> 0.1). Though tensile stress caused the cracks in AlGaN layer with high AlN molar fraction, we found that the cracks dramatically reduced when the GaN layer quality was not good. Using this technique, we fabricated a GaInN multi-quantum-well (MQW) surface emitting diodes were fabricated on 15 pairs of AlGaN/GaN distributed Bragg reflector (DBR) structures. The reflectivity of 15 pairs of AlGaN/GaN DBR structure has been shown as 75% at 435 nm. Considerably higher output power (1.5 times) has been observed for DBR based GaInN MQW LED when compared with non-DBR based MQW structures.

key words: *light emitting diode (LED), distributed Bragg reflector (DBR), GaN, AlGaN, GaInN*

1. Introduction

GaN-based materials are useful for short-wavelength optical devices because they have a large direct band gap ranging from 1.9–6.2 eV. Metal-Insulator-Semiconductor (MIS) based GaN light emitting diode (LED) was first fabricated and reported by Pankove et al. [1]. This metal-insulator-semiconductor (MIS) structure LED consisted of an unintentionally doped GaN as an n-type and Zn-doped insulating GaN as insulating layer. After realizing p-type GaN by post treatment [2], [3], GaN p-n junction LED [2], [4] and GaN-based double-hetero-structure (DH) LED [5], [6] were fabricated using metalorganic chemical vapor deposition (MOCVD). Particularly, the development of GaInN growth techniques were leads for the growth of high-quality films [7], [8] and quantum-well-structure (QW) [9], [10] that can be used as active layers. Basic understanding of GaN-based light emitters will be very much useful to improve the quality and life-time of GaN-based edge-emitting laser diodes [11].

Recently, GaN-based vertical-cavity surface-emitting laser diodes (VCSELDs) have attracted a great interest for various optical applications [12], [13]. The difference of refractive index between AlN and GaN is not that high as AlAs and GaAs. The distributed Bragg reflector (DBR) consisting of the stacks of AlGaN/GaN layered structures are needed to realize high reflectivity [14]. Fundamental understanding of strained AlGaN on GaN/sapphire structures are essential for the fabrication of AlGaN/GaN DBR. This is because AlGaN grows coherently on GaN to match their in-plane lattice constants [15]. The properties of AlGaN on GaN/sapphire structures are expected to be different from AlGaN on sapphire. Although, properties of strained AlGaN are very interesting for both fundamental understanding and real devices, it is too difficult to suppress the cracks in AlGaN layers which is grown on GaN. Usually the cracks will generate only because of stress during AlGaN growth. Therefore, it is necessary to grow crack-free film to fabricate a DBR with a high reflectivity. Due to the built-in reflection properties of GaN-based DBR, it is useful not only for VCSLEDs but also for various other applications. For example, we believe that DBR can be used as backside mirrors in LED to enhance the external quantum efficiency. A conventional GaN-based LED consists of p-layer, active layer and n-layer on transparent sapphire substrate. In such a configuration, light emitted from the active layer is transmitted towards the top surface of the device and also towards the sapphire substrate. When the emitting light towards the substrate has to be reflected back towards the p-electrode using backside reflector. If we use the backside reflector configuration (that means DBR) for the fabrication of LEDs, the external quantum efficiency will be higher than that of conventional GaN based LEDs.

In this study, the properties of strained AlGaN layer grown on GaN/sapphire structures are reported.

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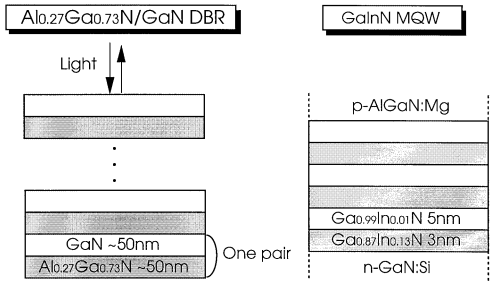

Fig. 1 Schematic diagrams of $Al_{0.27}Ga_{0.73}$/GaN DBR and $Ga_{0.99}In_{0.01}N/Ga_{0.87}In_{0.13}$ MQW structures.

DBR has been realized effectively using AlGaN/GaN strained structures. We have also realized the effectiveness of DBR in the MQW LED structure using AlGaN/GaN strain layer.

2. Experimental

An AlGaN/GaN single-hetero (SH) structure, AlGaN/GaN DBR stacks and GaInN MQW LED structures were grown on sapphire (0001) substrate using conventional horizontal atmospheric pressure MOCVD method. Trimethylgallium (TMG), trimethylindium (TMI), trimethylaluminum (TMA) and ammonia (NH_3) were used as source materials. Monosilane (10 ppm, diluted with hydrogen) and biscyclopentadienyl magnesium (Cp_2Mg) were used as n-type and p-type dopant, respectively. The sapphire substrate was heated at 1100°C for 10 min in a hydrogen atmosphere to clean the surface. A 30-nm-thick GaN layer was deposited as the buffer at 550°C. Then, the substrate was heated up to 1080°C and a GaN layer was grown on the GaN buffer layer. After a purge time, the AlGaN layer was grown on the GaN/sapphire structure. The thickness of GaN and AlGaN was 1.5–2 and 0.1–0.5 μm, respectively. For the realization of DBR, 30 and 15 pairs of quarter-wave $Al_{0.27}Ga_{0.73}N$/GaN reflectors were grown on GaN/sapphire structure. For the application of AlGaN/GaN DBR, GaInN MQW LED structure was grown on 15 pairs of AlGaN/GaN DBR. The GaInN MQW LED structure consisted of 0.5-μm-thick un-doped GaN, 4-μm-thick n-type GaN:Si, 3 consecutive periods of quantum well [consisting of 5-nm-thick $Ga_{0.99}In_{0.01}N$ barrier layer and 3-nm-thick $Ga_{0.87}In_{0.13}N$ well layer], 20-nm-thick p-type $Al_{0.15}Ga_{0.85}N$:Mg and 0.2-μm-thick p-type GaN:Mg. Ti/Al and Ni/Au metal layers were used as n- and p-electrodes (diameter: 400 μm), respectively. Schematic diagram of quarter-wave $Al_{0.27}Ga_{0.73}N$/GaN reflectors and GaInN/GaN MQW structures are shown in Fig. 1.

AlGaN/GaN SH structures were characterized by X-ray diffractometry. The wavelength dispersions of the refractive index for the strained AlGaN on GaN/sapphire were measured by a spectroscopic ellipsometry [16]. The reflectance spectra of DBR were measured by a spectro photometer. Photoluminescence (PL) measurements were carried out with focused He-Cd laser excitation (325 nm, 11 mW) at room temperature. Electroluminescence (EL) spectra from the LED were measured using an optical spectrum analyzer. The output power from the LED was measured using a power meter without the integral sphere.

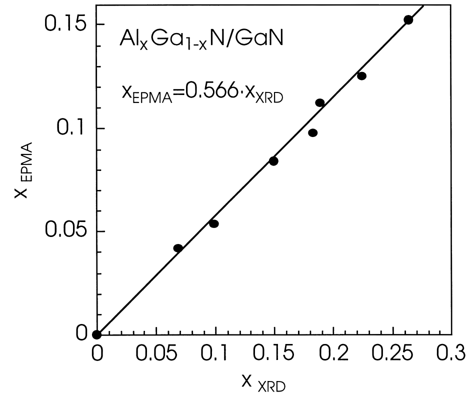

Fig. 2 Comparison of AlN molar fraction x determined by XRD with that determined by EPMA in AlGaN/GaN.

3. Results and Discussion

3.1 Properties of AlGaN/GaN SH Structure

A GaN thin film on the sapphire substrate has a residual stress caused by the difference in the thermal expansion coefficients between GaN and sapphire. The compressive strain is induced in GaN and the in-plane lattice constant is reduced for a thin GaN film on sapphire. For a thin AlGaN on a thick GaN, the in-plane lattice constant of AlGaN is strained to accommodate the lattice mismatch. Therefore, X-ray analysis by Vegard's law involves difficulty to determine the exact molar fraction of AlN in AlGaN grown on GaN/sapphire structures. Simultaneously electron probe micro analyses (EPMA) were also used to confirm the composition of the AlGaN layer directly. Figure 2 shows the AlN molar fraction determined by both EPMA (x_{EPMA}) and X-ray analyses [Vegard's law] ($x_{X\text{-}ray}$). Eventhough x_{EPMA} is not equal to $x_{X\text{-}ray}$, the x_{EPMA} varies linearly with $x_{X\text{-}ray}$. Therefore, the relationship between x_{EPMA} and $x_{X\text{-}ray}$ is expressed as $x_{EPMA} = 0.566 x_{X\text{-}ray}$.

When a thin film was grown on a thick layer with a strain for a wurtzite crystal, lattice constants for a thin film were expressed as follows:

$$c' = c\left[1 - 2\frac{C_{13}}{C_{33}}\left(\frac{a' - a}{a}\right)\right], \tag{1}$$

where a and c are the strain-free lattice constant, a' and c' strained lattice constant and C_{13} and C_{33} are the elastic stiffness constants [17]. By using the X-ray

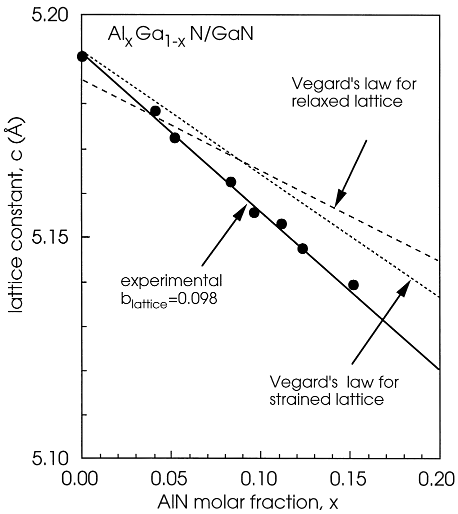

Fig. 3 AlN molar fraction dependence of the lattice constant of AlGaN/GaN/sapphire.

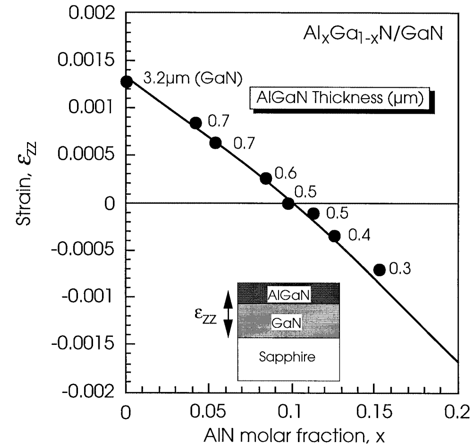

Fig. 4 The strain ε_{zz} in AlGaN grown on GaN/sapphire as a function of AlN molar fraction.

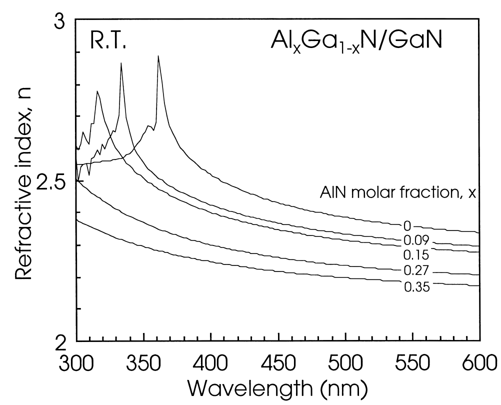

Fig. 5 Wavelength dispersions of refractive index of AlGaN/GaN/sapphire. These refractive indexes were measured by the spectroscopic ellipsometry.

Bond's method, the lattice constant c for GaN grown on sapphire substrate was obtained to be 5.1906Å. Taking a value of 0.598 as $2C_{13}/C_{33}$ [15], the lattice constant a'_{GaN} was 3.1820Å for GaN grown on sapphire substrate.

When AlGaN layer is coherently grown on GaN/sapphire structure, the lattice constant a'_{AlGaN} with any AlN molar fraction is equal to that of GaN on sapphire. Taking this value as a'_{AlGaN}, c'_{AlGaN} can be calculated using Eq. (1). When the lattice constants and the elastic stiffness constants for strain-free AlGaN are assumed to obey the quadratic equations, the compositional dependencies are expressed as follows:

$$c_{AlGaN} = 5.185x + 4.982(1-x) - bx(1-x), \quad (2)$$

$$\begin{cases} C_{13,AlGaN} = C_{13,GaN} + C_{13,AlN}(1-x) - bx(1-x) \\ C_{33,AlGaN} = C_{33,GaN} + C_{33,AlN}(1-x) - bx(1-x)' \end{cases} \quad (3)$$

where C_{13}, AlGaN and C_{33}, AlGaN are the elastic stiffness constants for AlGaN, and b the bowing parameter for lattice constant c. Figure 3 shows AlN molar fraction dependence of the lattice constant c'_{AlGaN}. The bowing parameter b can be obtained by curve fitting using Eqs. (1), (2) and (3) to the experimental data. An excellent fitting showed that b was obtained to be 0.097. The lattice constants of strain-free AlGaN almost varies with AlN molar fraction x linearly, however the curve bows downwards. The along-c-axis strain ε_{zz} in strained AlGaN could be obtained from strained and strain-free lattice constants as shown in Fig. 4. Since strain-free a_{AlGaN} (AlN molar fraction $x < 0.1$) is larger than strained a'_{GaN}, ε_{zz} shows an anomalous behavior as follows. In the range of AlN molar fraction from 0 to 0.1, ε_{zz} is positive value and AlGaN is under the in-plane compressive stress. When the AlN molar fraction is more than 0.1, ε_{zz} is negative value and AlGaN is under the in-plane tensile stress. For the application to the DBR, the high contrast between high (GaN) and low (AlGaN) refractive index layers are needed to realize high reflectivity. Although AlGaN with higher AlN molar fraction has lower refractive index, the stress in such strained AlGaN becomes higher and causes cracks in AlGaN above a thinner thickness. Therefore, trade-off between crack-reduction and low reflactive index, is a serious problem to fabrication nitride-based DBR with high reflectivity.

3.2 Crack-Free DBR Fabrication

The wavelength dispersions of the refractive index for the strained AlGaN layer on GaN/sapphire structures are shown in Fig. 5. The refractive index of the strained AlGaN layer decreases while the mole fraction increases. Simultaneously, the refractive index increases while the wavelength decreases. When considering GaInN as the active layer, the refractive index difference between GaN and $Al_{0.27}Ga_{0.73}N$ were more

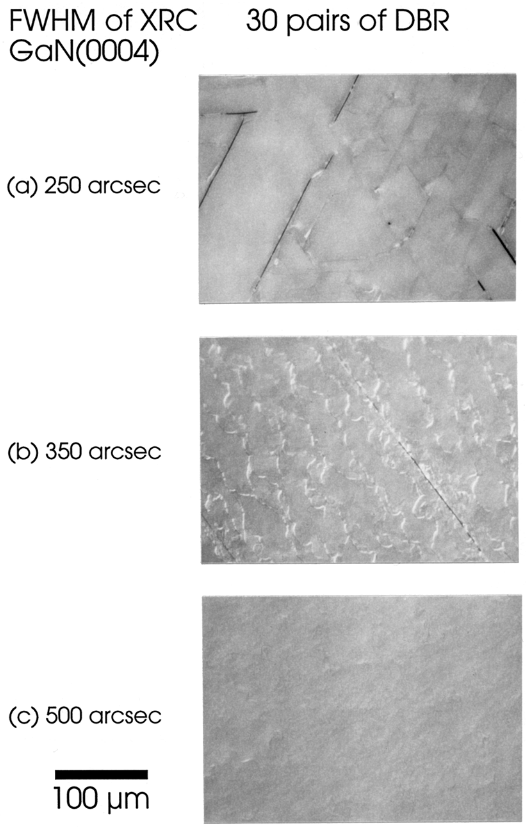

Fig. 6 Surface morphologies of the 30 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR on GaN/sapphire. FWHM of DCXRC for GaN (0004) were (a) 250, (b) 350 and (c) 500 arcsec, respectively.

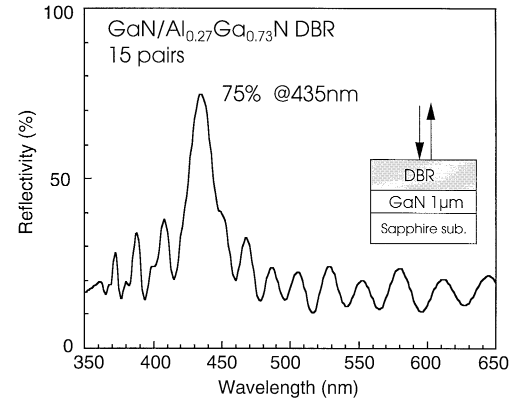

Fig. 7 The reflectance spectrum of the 15 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR on GaN/sapphire structure. The thickness of the GaN buffer layer was 0.1 μm to obtain crack-free DBR.

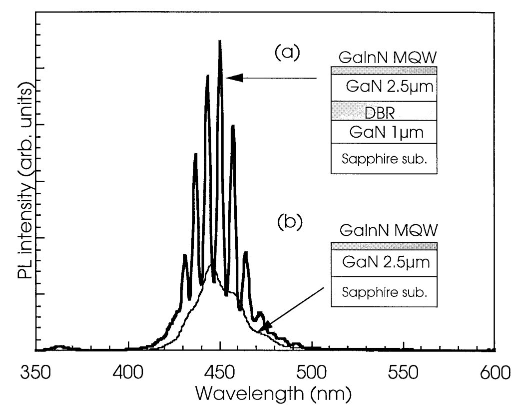

Fig. 8 PL spectra of GaInN MQW (a) with and (b) without $Al_{0.27}Ga_{0.73}N$/GaN DBR at room temperature.

than 0.2 for the wavelength from 370 to 450 nm. The 30 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR are needed to achieve the reflectivity of more than 99% from theoretical calculation. However, the cracks during the growth of thick AlGaN with high AlN molar fraction on GaN/sapphire structures can not be suppressed using conventional growth technique. We found that crack free AlGaN on GaN/sapphire was obtained when the GaN layer quality was not good. This has been confirmed using full width at half maximum (FWHM) of double crystal X-ray rocking curve (XRC) of GaN layer. In the same way, three 30 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR were grown on ~1-μm-thick GaN/sapphire structures with different FWHM of XRC. The FWHM of XRC for (0004) diffraction from the GaN were (a) 250, (b) 350 and (c) 500 arcsesc, respectively. The surface morphologies of these samples are shown in Fig. 6. The cracks of DBR were reduced dramatically while FWHM of XRC of GaN/sapphire was increased. Moreover, the reflectivity of the DBR shown in Fig. 6(c) reaches 94% at 380 nm. It is believed that stresses in AlGaN layer do not concentrate on the crackable directions in the case of broad XRC of GaN/sapphire. In order to increase the FWHM of XRC of GaN/sapphire, two techniques were developed as follows: (1) increasing nitrogen partial pressure during the GaN growth, (2) increasing the thickness of low-temperature GaN buffer layer (~0.1 μm).

3.3 GaInN MQW LED with DBR

Before applying DBR to built-in backside mirrors in LED, we investigated optical properties of GaInN MQW on GaN/sapphire structures with and without DBR. For this experiment, we prepared two GaInN MQW samples that had 3 periods of quantum well consisting of 5-nm-thick $Ga_{0.99}In_{0.01}N$ barrier layer and 3-nm-thick $Ga_{0.87}In_{0.13}N$ well layer: (a) GaInN MQW/GaN/sapphire, (b) GaInN MQW/GaN/(15 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR)/GaN/sapphire. In (b), the above-mentioned crack-reduction technique (2) was used to obtain crack-free DBR. Figure 7 shows reflectance spectrum of 15 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR. The maximum reflectivity of 15 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR was designed to agree with GaInN MQW emission peak at 440 nm. The actual maximum reflectivity was obtained to be 75% at 435 nm. Figure 8 shows the PL spectra of GaInN MQW (a) with and (b) without AlGaN/GaN DBR at room

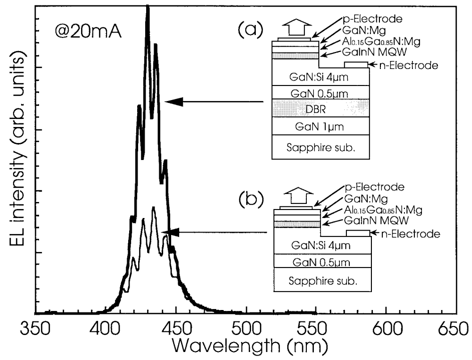

Fig. 9 Emission spectra of the GaInN MQW LED (a) with and (b) without $Al_{0.27}Ga_{0.73}N$/GaN DBR at the forward current of 20 mA.

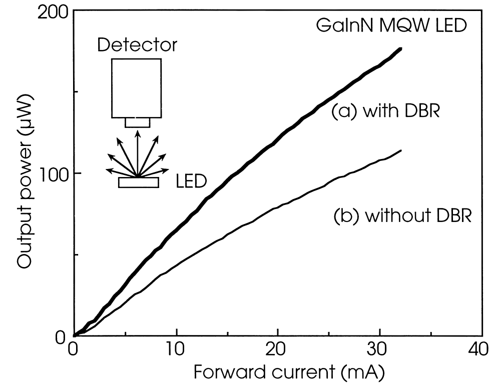

Fig. 10 The light output power of (a) the GaInN MQW LED with DBR compared to (b) that without DBR as a function of the forward current.

temperature. Both samples showed the PL spectra with FWHM of 30 nm. This result indicates that differences between GaInN MQW with and without DBR on the quality are negligible. The PL spectrum of GaInN MQW without $Al_{0.27}Ga_{0.73}N$/GaN DBR showed small fringe that is responsible for the interference reflection between the surface and sapphire substrate. The PL spectrum was modulated strongly and this fringe mode was enhanced to be larger for DBR based structure when it compared with conventional structure. This fringe mode space is assumed to be caused by the vertical cavity formed between the surface and DBR. The fringe mode for DBR based structures are in good agreement with the theoretical calculation. This result indicates that the PL light was reflected by DBR effectively and the vertical cavity was also formed between the surface and DBR.

GaInN MQW LEDs were fabricated on GaN/sapphire with and without 15 pairs of $Al_{0.27}Ga_{0.73}N$/GaN DBR to realize the actual effects of DBR using the above mentioned technique (2). The EL spectra are shown in Fig. 9. Though GaInN MQW on GaN/sapphire with DBR showed obvious modulated PL spectrum, EL from the LED with DBR showed almost the same shape as that of LED without DBR. It is considered that the reflectivity from semitransparent p-electrode/air is inferior to that from GaInN MQW/air. However, a large difference in output power was observed between GaInN MQW LED with and without DBR. The light output powers of both GaInN MQW LED as a function of the forward current is shown in Fig. 10. The output power for the LED with DBR is 1.5 times larger than that of LED without DBR structure. Typical output power of the LED at a forward current of 20 mA was 120 and 79 μW for with and without DBR structure, respectively. The external quantum efficiency at 10 mA is enhanced from 0.16 to 0.23% by use of DBR. Thus the enhancement of optical output power has been realized using built-in DBR.

4. Conclusion

Investigations were carried out on MOCVD-grown strained AlGaN/GaN/sapphire structures using single crystal X-ray diffratometry. While AlGaN with lower AlN molar fraction (< 0.1) is under the in-plane compressive stress, it is under the in-plane tensile stress with high AlN molar fraction (> 0.1). Though tensile stress caused the cracks in AlGaN layer with high AlN molar fraction, we found that the cracks dramatically reduced when the GaN layer quality was not good. Using this technique, GaInN MQW surface emitting diodes were fabricated on 15 pairs of AlGaN/GaN DBR structures. The reflectivity of 15 pairs of AlGaN/GaN DBR structure has been shown as 75% at 435 nm. Considerably higher output power (1.5 times) has been observed for DBR based GaInN MQW LED when it compared with non-DBR based MQW structures.

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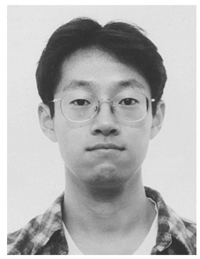

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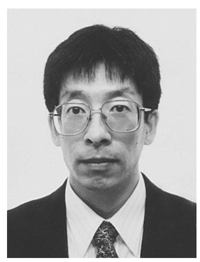

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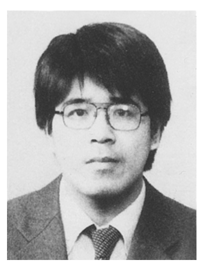

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