

Advantages of Employing the Freestanding GaN Substrates with Low Dislocation Density for White Light-Emitting Diodes

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To improve the luminous efficiency of white light-emitting diodes (LEDs) for general lighting, the InGaN-based-LEDs with thick quantum wells (QWs) were examined on our unique freestanding gallium nitride (GaN) substrates with low-threading dislocation densities (TDDs). With LEDs grown on sapphire, which are commonly used, the crystalline quality of QWs was deteriorated and the luminous efficiency was degraded with the increase in the total thicknesses of InGaN QWs. On the other hand, on the GaN substrates with low TDD, the luminous efficiency at high current density was successfully improved without the deterioration of crystalline quality as the total thicknesses of InGaN QWs increased. Consequently, it was confirmed that the combination of thick InGaN QWs and low-TDD GaN substrates gives rise to highly efficient LEDs required for solid state lighting.

Keywords: LED, droop, GaN, InGaN, GaN substrate, dislocation, defect, QW

1. Introduction

The market for white light-emitting diodes (LEDs^{*1}) using InGaN-based blue LEDs with yellow phosphors is growing rapidly toward solid-state lighting applications. In addition to the various advantages provided by LEDs including the high efficiency and long life time, the high luminous flux is recognized as an increasingly important factor in terms of expanding the application to automotive lighting, indoor lighting, large LCD backlighting, and so on.

Since the luminous flux from one LED chip is only a few lumens, many LED chips are needed to obtain the luminous flux equivalent to a fluorescent lamp. The quantity increase of LED chips costs too much and impedes a spread of LEDs for general lighting. For this reason, the increase of the luminous flux or light output power from one chip by increasing the operating current is strongly demanded.

However, it is difficult to realize the high external quantum efficiency (EQE) at a high current density because InGaN-based blue LEDs suffer from the so-called “efficiency droop,” which is a phenomenon characterized by the saturation of the light output power and the decrease of the internal quantum efficiency with increasing current density, typically above 1-10 A/cm² (1), as shown in Fig. 1. Recently, efforts to clarify the origin of the efficiency droop have been made actively. Several mechanisms, such as the electron leakage⁽²⁾⁻⁽⁴⁾, the carrier delocalization^{(1), (5), (6)}, the defect recombination activated at a high carrier density⁽⁷⁾, and Auger recombination⁽⁸⁾⁻⁽¹²⁾, have been proposed. Approaches to mitigate the efficiency droop have also been studied both experimentally and theoretically, suggesting that the reduction of the injected carrier density per volume in the quantum wells (QWs^{*2}) is effective in the EQE improvement at high current densities^{(10), (11), (13)}. Furthermore, it is reported that the threading dislocation (TD) plays an important role in this approach.

Most of currently commonly-used InGaN-based LEDs

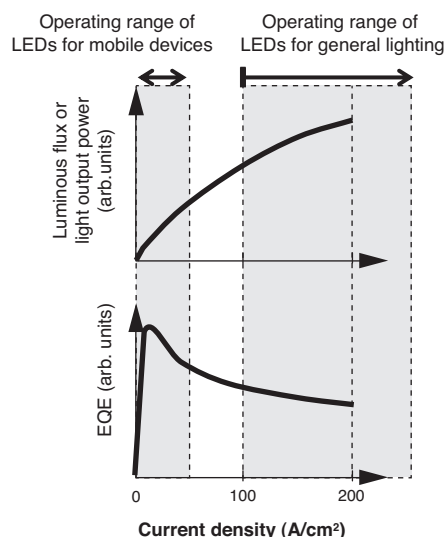


Fig. 1. Light output power and EQE of a typical InGaN-based blue LED as a function of current density

are grown on sapphire, and thus, InGaN active layers include many TDs originated from the lattice mismatch between hetero-epitaxial GaN layers and sapphire exist in InGaN active layers. On the other hand, homo-epitaxial GaN layers grown on GaN substrates contain few TDs. In addition, LEDs on GaN substrates can utilize the characteristics of GaN substrates such as low resistivity and high heat conductivity. We started the first mass production of the 2-inch-size free-standing GaN substrates with low threading dislocation densities (TDDs) in 2003, which made a great contribution to the practical realization of the violet laser diodes for Blu-ray discs⁽¹⁴⁾⁻⁽¹⁶⁾. At a blue wavelength region, moreover, we demonstrated the EQE improvement of LEDs on GaN substrates at high current densities by adopting the thick QWs^{(17), (18)}. Therefore, the

thick QWs with our unique freestanding GaN substrates have a potential to achieve highly efficient white LEDs for solid-state lighting. In this study, we fabricated InGaN-based blue LEDs on low-TDD GaN substrates and sapphire, and investigated the dependence of the EQE on the QW thickness, the QW number, and the TDD at a high current density up to 200 A/cm². We found that the total QW thicknesses have a great impact on the crystalline quality of the blue-emitting InGaN QWs and the EQE at a high current density. Micro-photoluminescence (PL) measurements and scanning transmission electron microscope (STEM^{*3}) analyses revealed that the newly generated structural defects originating from the TDs limit the increase of the total QW thicknesses on sapphire.

2. Experiment

C-plane GaN substrates were produced by hydride vapor phase epitaxy (HVPE^{*4})^{(15), (16)}. The TDDs of GaN substrates and n-GaN layers on c-plane sapphire were typically 1×10^6 cm⁻² and 5×10^8 cm⁻², respectively. Epitaxial growth was simultaneously carried out on both GaN substrates and n-GaN layers on sapphire by organometallic vapor phase epitaxy (OMVPE^{*5}). **Figures 2 (a) and (b)** show the schematic structure of LEDs on (a) GaN substrates and (b) sapphire. A 5- μ m-thick n-type GaN layer was grown on substrates, followed by a 0.05- μ m-thick n-InGaN buffer layer, an InGaN/GaN multiple-QW active region, a 0.02- μ m-thick p-AlGaIn electron-blocking layer, and a 0.06- μ m-thick p-GaN layer. The QW numbers of 3 and 6 were examined, and the QW thicknesses were varied from 3 to 7 nm. The In compositions of the InGaN QWs were adjusted in the range of 13 to 15 % depending on the QW thickness so that the peak wavelengths at 200 A/cm² become 430 to 450 nm. Semi-transparent electrodes of 0.4×0.4 mm² size for p-type ohmic contacts were fabricated by standard photolithography and deposition techniques. Mesa structures were fabricated by reactive-ion etching,

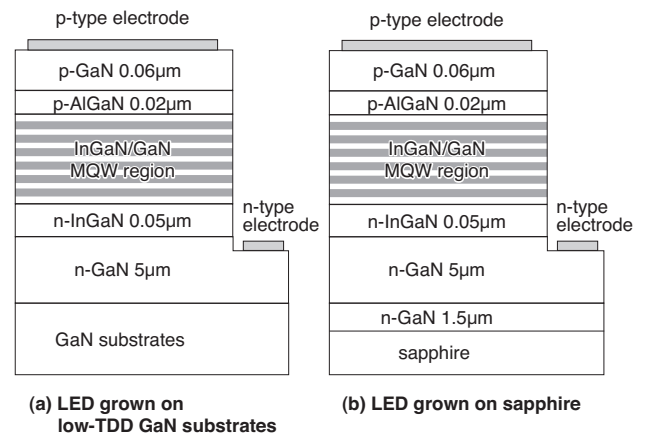


Fig. 2. Schematic structures of LEDs

and n-type ohmic contacts were formed on the exposed n-GaN surface. In order to eliminate the difference of the light extraction efficiency originating in the roughness of the back surfaces, the backsides of all wafers were polished. Electroluminescence (EL) measurements were performed in bare wafer geometry in pulsed mode (10 kHz, duty = 5 %) at room temperature to avoid the heating effect.

3. Results

3-1 Observations of micro-PL images

The crystalline quality of the InGaIn active layers was evaluated by micro-PL measurements. Blue PL from the InGaIn active layers was excited by the 365 nm line of a mercury lamp. In PL images, non-radiative areas at crystalline defects are observable as dark regions. **Figure 3** shows the micro-PL images of LEDs with various QW thicknesses and QW numbers on sapphire and GaN substrates. Whereas the PL image of the 3-nm-thick 6QW LED on sap-

QW number	3QW		6QW		
	3 nm	5 nm	3 nm	5 nm	7 nm
QW thickness	3 nm	5 nm	3 nm	5 nm	7 nm
Total InGaIn QW thickness	9 nm	15 nm	18 nm	30 nm	42 nm
LEDs on sapphire					
LEDs on low-TDD GaN substrates					

Fig. 3. Micro-photoluminescence images of QWs on various substrates, QW numbers, and QW thicknesses

phire was uniform, there were many non-radiative areas in the PL image of the 5-nm-thick 6QW LED on sapphire. On the other hand, the uniform PL images were obtained for both the 3- and the 5-nm-thick 6QW LEDs on GaN substrates. These results were in agreement with our previous cathodoluminescence study^{(17), (18)}. When the QW thickness was further increased to 7 nm on sapphire, the PL intensity was degraded significantly because of the enlargement of the non-radiative areas. On the other hand, only a few non-radiative areas were induced in the 7-nm-thick 6QW LED on a GaN substrate, indicating that the TDs strongly correlate the formation of the non-radiative regions. Thus, the crystalline quality of InGaN active layers can be maintained with increasing QW thickness on freestanding GaN substrates with low TDDs, while the degradation on InGaN active layers are remarkable with increasing QW thickness on sapphire substrates. In the case of the 3QW LEDs with 3- and 5-nm-thick QWs, homogeneous PL images were obtained regardless of the substrates. The effect of the QW number was not addressed in our previous report, where only 6QWs were studied. From these results obtained in this work, in particular the striking difference between the 3-nm-thick 6QWs and the 5-nm-thick 6QWs, it was found that the total QW thicknesses as calculated by a product of QW thicknesses and numbers which are also shown in Fig. 3 have a great impact on the formation of the non-radiative areas on sapphire. It is considered that the amount of strain originated from the lattice mismatch between the thick InGaN QWs and GaN underlying layers were increased with an increase in the total QWs thicknesses, and as a result, many TDs on sapphire enhanced the strain relaxation with the introduction of the crystalline defects.

3-2 EQEs of LEDs at high current densities

We now discuss the EQEs on these LEDs at high current densities. The comparison of the EQE at 200 A/cm² between GaN substrates and sapphire was conducted using the total QW thicknesses, as shown in Fig. 4. It is noted that the EQE values are relatively lower than those of typical LED lamps (around 10%) because of the measurement in bare wafer geometry. As for the LEDs on GaN substrates, the EQE was improved with an increase in the total QW thicknesses up to 30 nm. Furthermore, the EQE did not deteriorate markedly even when the total QW thickness was increased to 42 nm. In contrast, the EQE on sapphire drastically dropped when the total QW thicknesses exceeded 18 nm. Thus, the upper limit of the total QW thicknesses is 18 nm on sapphire substrates in this experiment. This is consistent with the transition of the PL images for the LEDs on sapphire in Fig. 3, indicating that the difference of the EQE between GaN substrates and sapphire at the total QW thicknesses above 18 nm was caused by the generation of non-radiative regions on sapphire.

In order to elucidate the effect of low-TDD GaN substrates, the comparison of the EQEs vs. the current densities on both substrates were shown in Fig. 5. The total QW thicknesses of these LEDs on a GaN substrate and sapphire shown in Fig. 5 were 30 nm and 18 nm, respectively, which were the optimum structures in Fig. 4. The light output powers at 200 A/cm² were obtained 49 mW on sapphire and 57 mW on the GaN substrate, resulting in the 16% improvement of the output power on the GaN substrate. The

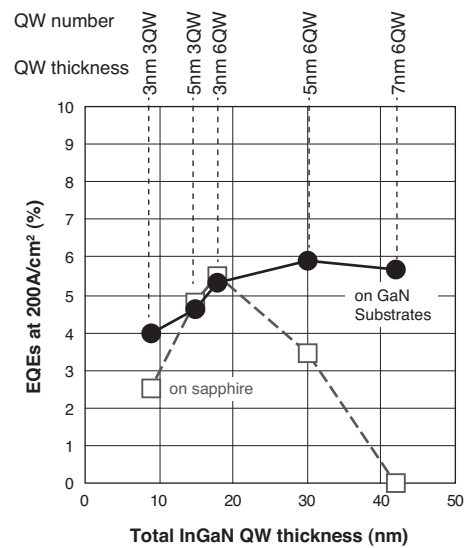


Fig. 4. Dependence of EQEs at 200A/cm² on the total QW thickness

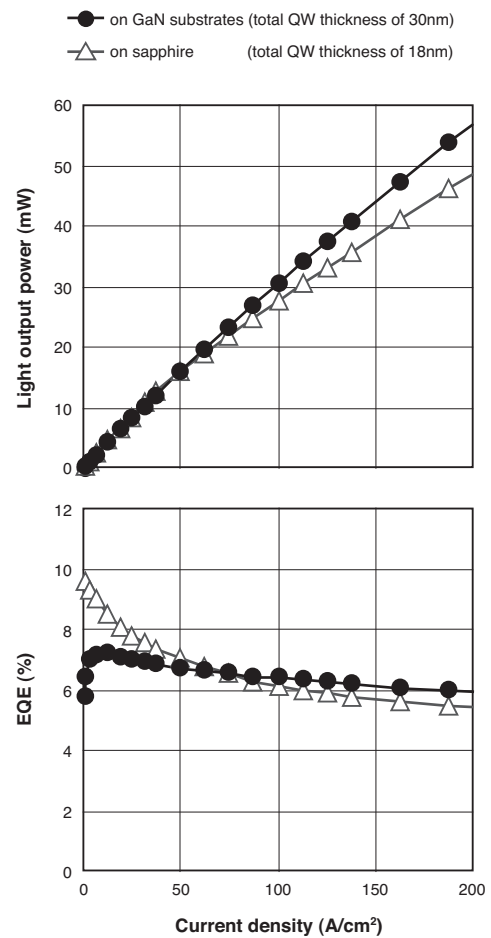


Fig. 5. Light output power and EQE of LEDs on sapphire and GaN substrates. It is noted that EQE values are relatively low because of measurements in bare wafer geometry.

EQE on sapphire was remarkably reduced to 5.5% from the maximum EQE of 9% with the increase of the current density. On the other hand, the EQE on the GaN substrate remained in 6.0% by a slight decrease from the maximum EQE of 7.0%. Consequently, the efficiency droop was considerably improved by the utilization of both low-TDD GaN substrates and thick QWs. While it is difficult to reveal that the origin of efficiency droop is Auger recombination in this study, these results indicate that a combination of the thick QWs and low-TDD GaN substrates successfully give rise to the effective reduction of the injected carrier density per unit volume and improve the EQEs of blue LEDs operated under high current density.

3-3 STEM analyses

As shown in **Fig. 3**, non-radiative regions of the LED with the total QW thickness of 30 nm, which corresponds to the QW thickness of 5 nm and the 6QWs on sapphire, were remarkably increased compared to that on the GaN substrate. In addition, the EQE at 200 A/cm² of the LED on sapphire was remarkably low compared to that on the GaN substrate as shown in **Fig. 4**. In order to clarify the origin of the non-radiative areas on sapphire, STEM analyses were conducted for these LEDs. Samples were fabricated by a micro-sampling method using a focused ion beam (FIB^{*6})⁽¹⁹⁾. An electron beam was accelerated to 200 kV and injected along the <11-20> direction. **Figures 6 (a) and (b)** show the STEM images of the LEDs on the GaN substrate and sapphire, respectively. As shown in **Fig. 6 (a)**, uniform 5-nm-thick 6QW layers with abrupt interfaces were successfully fabricated on the GaN substrate without any QW thickness fluctuations⁽²⁰⁾ or V-shaped pits⁽²¹⁾. On the other hand, many structural defects were observed on sapphire as shown in **Fig. 6 (b)**. These defects consisted of not only TDs which are originated from the interface of the n-GaN and sapphire substrate as indicated by an arrow (1) but also newly generated dislocations from the 4th QW as indicated by an arrow (2) and the regions with non-uniform light-dark contrasts as indicated by arrows (3). It was revealed that these non-uniform regions consist of decomposed InGaN QWs and In segregations by a composition analysis. It is considered that the non-radiative areas in the PL im-

ages corresponded to these structural defects. Since the total QW thicknesses significantly affected the PL morphology, the strain within the QWs caused by the lattice mismatch between the InGaN QWs and the n-GaN underlying layers is considered to play an important role in the defect generation. On the other hand, although the InGaN QWs on GaN substrates were also strained, there were few lattice disorders where the defects start to develop, and thus highly homogenous InGaN QWs were obtained as shown in **Fig. 6 (a)**. These results indicate that the employment of low-TDD GaN substrates is advantageous for growing thick InGaN QWs while suppressing the degradation of the crystalline quality toward high power blue LEDs. Consequently, high output powers and high EQEs at high current density can be realized on low-TDD GaN substrates.

4. Conclusion

InGaN-based blue LEDs were fabricated on c-plane GaN substrates and sapphire, and the dependence of the EQE on the QW thickness, the QW number, and the TDD was investigated at high current densities. The total InGaN QW thickness was found to have a strong correlation with the micro-PL morphology and the EQE. The EQE at 200 A/cm² on GaN substrates was improved with the increase of the QW thickness from 3 nm to 5 nm and the QW number from 3 to 6; the resulting total QW thickness was 30 nm. In addition, the LED on a GaN substrate did not show a marked deterioration of the EQE even when the total QW thickness was further increased up to 42 nm. On the other hand, the EQE of LEDs on sapphire decreased drastically when the total QW thickness exceeded 18 nm, which corresponded to the 3-nm-thick 6QWs, accompanied by the formation of the non-radiative areas in the PL images. The STEM analyses revealed that the structural defects originating from the TDs were responsible for these non-radiative areas. These results indicate that the reduction of the carrier density per volume in the QWs as well as the suppression of the degradation of the QW crystalline qual-

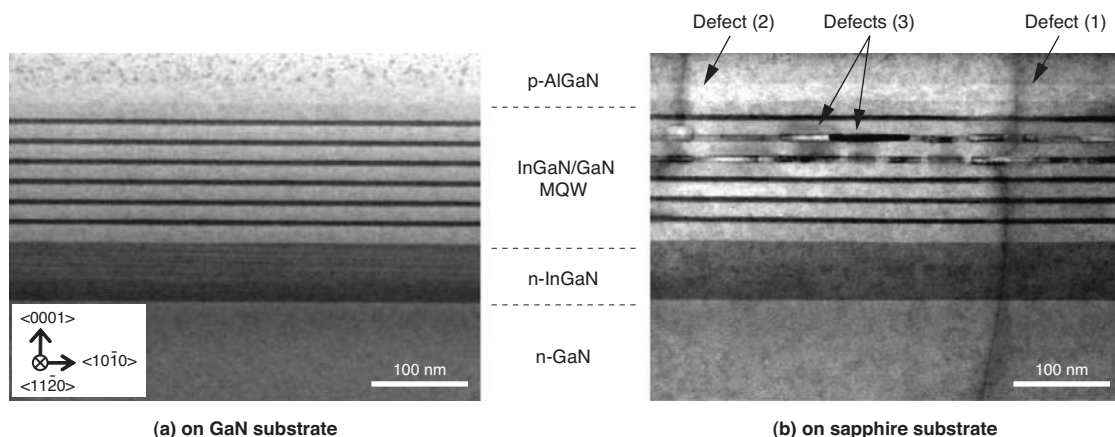


Fig. 6. Cross-sectional STEM images of MQW regions in LEDs on GaN and sapphire substrates

ity is a key factor for the mitigation of the efficiency droop issue. Consequently, the utilization of thick InGaN QWs and low-TDD GaN substrates is advantageous for realizing highly efficient LEDs required for solid state lighting driven at a high current density.

Recently, Sumitomo Electric Industries, Ltd. has developed 6-inch GaN substrates⁽²²⁾ and lower-cost engineered GaN substrates⁽²³⁾. A combination of these GaN substrates and thick QWs enables us to fabricate a lower-cost and high-performance LEDs suited for general lighting.

Technical Terms

- *1 LED (Light Emitting Diode): A Semiconductor device which transforms energy of electrons in to light by current injection.
- *2 QW (Quantum Well): A light emitting layer with a thickness of several nm sandwiched in between barrier layers with wider bandgaps than the light emitting layer. Plural stacking layers of QWs and barriers are called multiple quantum wells (MQWs).
- *3 STEM (Scanning Transmission Electron Microscope): A transmission electron microscope that scans a sample by focusing an electron beam on the sample.
- *4 HVPE (Hydride Vapor Phase Epitaxy): A production method of semiconductors using a hydride gas as a precursor.
- *5 OMVPE (Organometallic Vapor Phase Epitaxy): A production method of semiconductors using an organometallic as a precursor.
- *6 FIB (Focused Ion Beam): A sampling method for fabricating an STEM specimen by irradiating ion beams, such as Ga⁺, to cut out the micro region.

References

- (1) T. Mukai, M. Yamada, and S. Nakamura, "Characteristics of InGaN-based UV/ blue/ green/ amber/ red light-emitting diodes," *Jpn. J. Appl. Phys.* 38, pp. 3976-3981, 1999.
- (2) I. A. Pope, P. M. Smowton, P. Blood, J. D. Thomson, M. J. Kappers, and C. J. Humphreys, "Carrier leakage in InGaN quantum well light-emitting diodes emitting at 480 nm," *Appl. Phys. Lett.* 82, pp. 2755-2757, 2003.
- (3) J. Xu, M. F. Schubert, A. N. Noemaun, D. Zhu, J. K. Kim, E. F. Schubert, M. H. Kim, H. J. Chung, S. Yoon, C. Sone, and Y. Park, "Reduction in efficiency droop, forward voltage, ideality factor, and wavelength shift in polarization-matched GaInN/GaN multi-quantum-well light-emitting diodes," *Appl. Phys. Lett.* 94, pp. 011113, 2009.
- (4) X. Ni, Q. Fan, R. Shimada, Ü. Özgür, and H. Morkoç, "Reduction of efficiency droop in InGaN light emitting diodes by coupled quantum wells," *Appl. Phys. Lett.* 93, pp. 171113, 2008.
- (5) X. A. Cao, Y. Yang, and H. Guo, "On the origin of efficiency roll-off in InGaN-based light-emitting diodes," *J. Appl. Phys.* 104, pp. 093108, 2008.
- (6) B. Monemar and B. E. Sernelius, "Defect related issues in the 'current roll-off' in InGaN based light emitting diodes," *Appl. Phys. Lett.* 91, pp. 181103, 2007.

- (7) J. Hader, J. V. Moloney, and S. W. Koch, "Density-activated defect recombination as a possible explanation for the efficiency droop in GaN-based diodes," *Appl. Phys. Lett.* 96, pp. 221106, 2010.
- (8) Y. C. Shen, G. O. Mueller, S. Watanabe, N. F. Gardner, A. Munkholm, and M. R. Krames, "Auger recombination in InGaN measured by photoluminescence," *Appl. Phys. Lett.* 91, pp.141101, 2007.
- (9) K. T. Delaney, P. Rinke, and C. G. Van de walle, "Auger recombination rates in nitrides from first principles," *Appl. Phys. Lett.* 94, pp. 191109, 2009.
- (10) A. Laubsch, M. Sabathil, J. Baur, M. Peter, and B. Hahn, "High power and high efficiency InGaN based light emitters," *IEEE Trans. Electron Devices* 57, pp. 79-87, 2009.
- (11) N. F. Gardner, G. O. Müller, Y. C. Shen, G. Chen, S. Watanabe, W. Götz, and M. R. Krames, "Blue emitting InGaN GaN double heterostructure light emitting diodes reaching maximum quantum efficiency above 200A/cm²," *Appl. Phys. Lett.* 91 pp. 243506, 2007.
- (12) A. David and M. J. Grundmann, "Droop in InGaN light-emitting diodes: A differential carrier lifetime analysis," *Appl. Phys. Lett.* 96, pp. 103504, 2010.
- (13) M. Maier, K. Köhler, M. Kunzer, W. Pletschen, and J. Wagner, "Reduced nonthermal rollover of wide-well GaInN light-emitting diodes," *Appl. Phys. Lett.* 94, pp. 041103, 2009.
- (14) K. Motoki, T. Okahisa, N. Matsumoto, M. Matsushima, H. Kimura, H. Kasai, K. Takemoto, K. Uematsu, T. Hirano, M. Nakayama, S. Nakahata, M. Ueno, D. Hara, Y. Kumagai, A. Koukitu, and H. Seki, "Preparation of large freestanding GaN Substrates by hydride vapor phase epitaxy using GaAs as a Starting Substrate," *Jpn. J. Appl. Phys.* 40, pp. L140-L143, 2001.
- (15) K. Motoki, "Development of gallium nitride substrates," *SEI Technical Review*, No. 70, pp. 28-35, 2010.
- (16) M. Takeya, T. Hashizu, and M. Ikeda, "Degradation of GaN-based High-power lasers and recent advancements," *Proc. SPIE* 5738, pp. 63-71, 2005.
- (17) K. Akita, T. Kyono, Y. Yoshizumi, H. Kitabayashi, and K. Katayama, "Improvements of external quantum efficiency of InGaN-based blue light-emitting diodes at high current density using GaN substrates," *J. Appl. Phys.* 101, pp. 033104, 2007.
- (18) K. Akita, T. Kyono, Y. Yoshizumi, H. Kitabayashi, and K. Katayama, "High-efficiency InGaN blue light-emitting diodes on low-threading-dislocation-density GaN substrates," *SEI Technical Review*, No. 65, pp. 35-40, 2007.
- (19) A. Yamaguchi, "Material characterization of semiconductor devices," *SEI Technical Review*, No. 70, pp. 17-27, 2007.
- (20) N. K. van der Laak, R. A. Oliver, M. J. Kappers, and C. J. Humphreys, "Characterization of InGaN quantum wells with gross fluctuations in width", *Appl. Phys. Lett.* 91, pp. 121911, 2007.
- (21) A. Hangleiter, F. Hitzel, C. Netzel, D. Fuhrmann, U. Rossow, G. Ade, and P. Hinze, "Suppression of nonradiative recombination by V-shaped pits in GaInN/GaN quantum wells produces a large increase in the light emission efficiency", *Phys. Rev. Lett.* 95, pp. 127402, 2005.
- (22) A press release of Sumitomo Electric, "Sumitomo Electric announces the world's first 6-inch GaN substrates for white LED applications," November 16, 2010.
- (23) A press release of Sumitomo Electric, "Sumitomo Electric and Soitec announce collaboration on development of engineered GaN substrates," December 1, 2010.



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