AIP Advances
CELEBRATING 5 YEARS

Influence of laser lift-off on optical and structural properties of InGaN/GaN vertical blue light emitting diodes

M. H. Doan, S. Kim, J. J. Lee, H. Lim, F. Rotermund, and Kihong Kim

Citation: AIP Advances **2**, 022122 (2012); doi: 10.1063/1.4717493
View online: http://dx.doi.org/10.1063/1.4717493
View Table of Contents: http://scitation.aip.org/content/aip/journal/adva/2/2?ver=pdfcov
Published by the AIP Publishing

AIP ADVANCES **2**, 022122 (2012)

Influence of laser lift-off on optical and structural properties of InGaN/GaN vertical blue light emitting diodes

M. H. Doan,[1] S. Kim,[1] J. J. Lee,[1,a] H. Lim,[1] F. Rotermund,[2] and Kihong Kim[2]
[1]*Department of Electrical and Computer Engineering, Ajou University, Suwon 443-749, Korea*
[2]*Department of Physics, Ajou University, Suwon 443-749, Korea*

(Received 19 October 2011; accepted 19 April 2012; published online 4 May 2012)

The influences of the laser lift-off (LLO) process on the InGaN/GaN blue light emitting diode (LED) structures, grown on sapphire substrates by low-pressure metalorganic chemical vapor deposition, have been comprehensively investigated. The vertical LED structures on Cu carriers are fabricated using electroplating, LLO, and inductively coupled plasma etching processes sequentially. A detailed study is performed on the variation of defect concentration and optical properties, before and after the LLO process, employing high-resolution transmission electron microscopy (HRTEM), scanning electron microscopy (SEM) observations, cathodoluminescence (CL), photoluminescence (PL), and high-resolution X-ray diffraction (HRXRD) measurements. The SEM observations on the distribution of dislocations after the LLO show well that even the GaN layer near to the multiple quantum wells (MQWs) is damaged. The CL measurements reveal that the peak energy of the InGaN/GaN MQW emission exhibits a blue-shift after the LLO process in addition to a reduced intensity. These behaviors are attributed to a diffusion of indium through the defects created by the LLO and creation of non-radiative recombination centers. The observed phenomena thus suggest that the MQWs, the active region of the InGaN/GaN light emitting diodes, may be damaged by the LLO process when thickness of the GaN layer below the MQW is made to be 5 μm, a conventional thickness. The CL images on the boundary between the KrF irradiated and non-irradiated regions suggest that the propagation of the KrF laser beam and an accompanied recombination enhanced defect reaction, rather than the propagation of a thermal shock wave, are the main origin of the damage effects of the LLO process on the InGaN/GaN MQWs and the n-GaN layer as well. *Copyright 2012 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License*. [http://dx.doi.org/10.1063/1.4717493]

I. INTRODUCTION

High brightness InGaN/GaN light emitting diodes (LEDs) have attracted great attentions due to their promising applications for flat panel display, signage, and general lighting.[1] However, LEDs for general solid-state lighting still have several issues related to the improvement of the LED efficiency and output power. For example, the InGaN/GaN LEDs usually grown on substrates such as sapphire (Al_2O_3) and SiC suffer from the problems associated with the poor electrical and thermal conductivities of the substrates. Solving such problems is essential to achieve the high operating current LEDs. The electrical and optical characteristics of the InGaN/GaN LEDs grown on Si substrate, which has reasonably good thermal and electrical conductivities, are not yet comparable with those of the LEDs on sapphire substrate due to the large lattice and thermal expansion coefficient mismatches between the GaN and Si.[2,3]

---

[a] Corresponding author; electronic email: jaejin@ajou.ac.kr

Recently, many works have been reported on the vertical InGaN/GaN LED structure for which the sapphire substrate is removed using a laser lift-off (LLO) technique and carrier materials such as Cu or Si are employed on top of the LEDs for the LLO process.[4–8] The resulting vertical LEDs show an improved high current operation capability than conventional LEDs.[4] However, a high power KrF UV laser which is usually employed for the LLO process is known to affect the material properties of the InGaN/GaN LED structures.[5–8] Chen *et al.* have reported that, in addition to the superficial damage caused by KrF laser absorption, the shock waves can generate a cluster of half loops above the LLO interface.[5] The screw dislocations generated by the LLO process are believed to lead to an increase in the leakage current in the vertical LEDs.[6] Even though many studies have been reported on the damage effect of the LLO process on the GaN layers,[5–9] few works have been reported on the effects of the LLO process on the InGaN/GaN multiple quantum well (MQW) layers of the GaN-based LEDs. And most of the reported optical studies have been performed by using photoluminescence (PL) characteristics.

In this paper, we report the influence of the LLO process on the optical and structural properties of the InGaN/GaN blue LEDs studied by cathodoluminescence (CL), high-resolution X-ray diffraction (HRXRD) measurement, and electron microscopy observations. It is observed that the LLO process induces a quantum well (QW) intermixing as well as a dislocation generation in the n-GaN layer, when the thickness of the GaN layer below the QW active layer is chosen as a conventional value of about 5 μm. Moreover, from a detailed study on the depth-dependence of the dislocation generation and the position-dependence of the CL images, it is concluded that the aforementioned damage effects of the LLO process, i.e., the generation of dislocation loops in the bottom n-GaN layer and the QW intermixing, are due to the recombination enhanced defected reactions (REDRs) and the large energy difference between the photo-generated carriers and their relevant band edges rather than the generation and propagation of shock waves at/from the surface.

II. EXPERIMENTAL PROCEDURE

The growth process employed for the samples in this study is reported elsewhere.[10] The structure is composed of GaN-based double heterostructures, which included, from the bottom, an undoped GaN buffer layer of 1 μm on 30-nm nucleation layer, a Si-doped n-GaN layer of 4 μm, six $In_{0.3}Ga_{0.7}N$/GaN MQWs, and an Mg-doped p-GaN top cladding layer of 250 nm. The thickness of the bottom n-GaN layer is taken to be 5 μm, since this thickness is the most commonly adopted value. Moreover, we employed flat surface sapphire substrates, instead of patterned surface ones, to avoid the diffraction of the KrF laser beam in the LLO process, even though the later one is known to give better device characteristics. After cleaning, a Ti layer is deposited on top of the p-GaN layer by an electron beam evaporator. Then, a 100-μm thick Cu film is deposited using an electroplating technique. To remove the sapphire substrate by the LLO, a 248-nm KrF excimer laser with a pulse width of 25 ns and laser fluence of 544 mJ/cm^2 is irradiated to the sapphire substrates. Then, the nucleation and undoped GaN buffer layers are removed by inductively coupled plasma (ICP) etching using a gas mixture of Cl_2 and BCl_3. Certain parts of the n- or p-GaN are also further etched out by the ICP etching when necessary for comparison purpose.

Structural properties of the samples are examined by a HRTEM, field emission scanning electron microscopy (FE-SEM), and HRXRD. Optical properties are studied by the CL measurements. The CL spectra and CL images are obtained at room temperature using a 10-keV electron beam with a Gatan Mono CL3 system equipped with a high-sensitivity photomultiplier tube (PMT) attached to an SEM (S-4300SE). The room-temperature PL measurements are also performed with a 266-nm laser to compare the PL spectra with the CL spectra.

III. RESULTS AND DISCUSSION

As the dislocation density of the GaN layer near the substrate is well known to be increased after the LLO of the sapphire substrate,[5,9] we have considered the dislocation formation deep in the bottom n-GaN layer. Figure 1 shows the SEM images of the n-GaN surfaces at 2 μm from the bottom (a) when the p-GaN, InGaN/GaN MQW layers, and n-GaN of 2 μm are subsequently etched

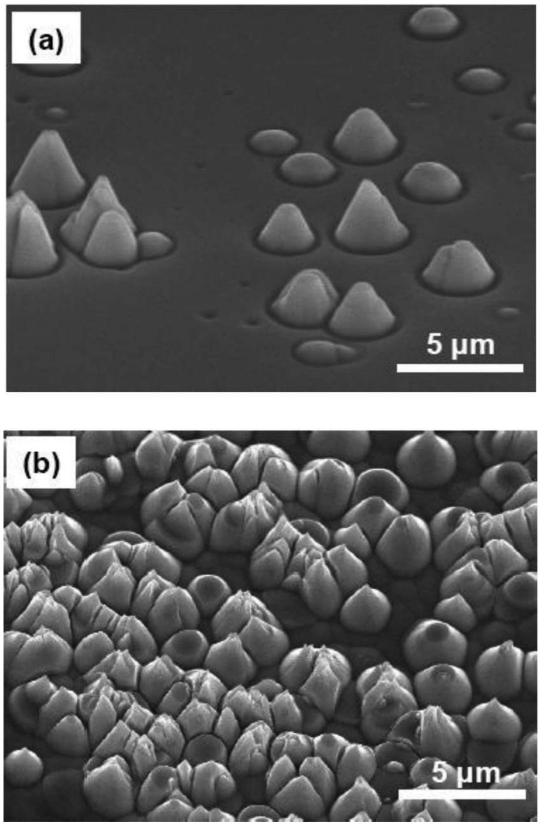

FIG. 1. SEM images of the n-GaN surfaces at 2 μm up from the bottom (a) when the p-GaN, InGaN/GaN MQW layers, and n-GaN of 2 μm (out of 4 μm) are etched out from the top to the bottom without removing the sapphire and (b) when the 4-μm thick bottom n-GaN layer is etched out by 2 μm after the LLO.

out from top to the bottom without removing the sapphire substrate and (b) when the bottom n-GaN layer is etched out by a thickness amount of 2 μm after the LLO. We observe that, after the LLO, the cone-shaped defects with an average size of about 2 μm are formed across the whole surface of the n-GaN layer. This increase of the cone shape patterns at the ICP-etched n-GaN surface, after the LLO process, is believed to be keenly correlated with the increase of dislocation density in the bottom GaN layer as discussed by Huang *et al.*.[9]

The influence of the LLO process on the optical properties of the vertical LEDs is investigated by CL measurements (Fig. 2). For the CL measurements on the vertical LEDs, the bottom n-GaN layer is thinned down to about 0.5 μm by an ICP etching in order for the 10-keV electron beam to fully penetrate into the InGaN/GaN MQWs from the bottom side; a 10-keV electron beam penetrates the GaN layer by about 0.7 μm with a quite broad energy deposition profile.[11,12] Figure 2 and its insets show the CL spectra and the CL images, respectively, the latter being obtained with the monochromatic wavelength corresponding to the MQW blue peaks before and after the LLO ($\lambda = 445$ and 440 nm, respectively). The CL peaks around 365 and 445 nm are coming from the GaN layers and MQWs, respectively. After the LLO, the CL intensity from the MQW is reduced by a factor of four with a slight blue-shift of its peak energy, and the so-called yellow luminescence around 2.2 eV is increased. The room-temperature PL spectra also showed a similar trend of intensity variation, before and after the LLO, with a 5-fold reduction in PL intensity of the MQW emission (not shown here). The CL images in the inset of Fig. 2 reveal again that defects are generated in the bottom n-GaN layer during the LLO process. We thus believe that the increase (decrease) of the yellow line (MQW emission) intensity after the LLO is keenly related to the increased defect concentration at the bottom layer.[13–15] However, the blue-shift of the peak energy from the MQWs may not be explained by such a defect generation.

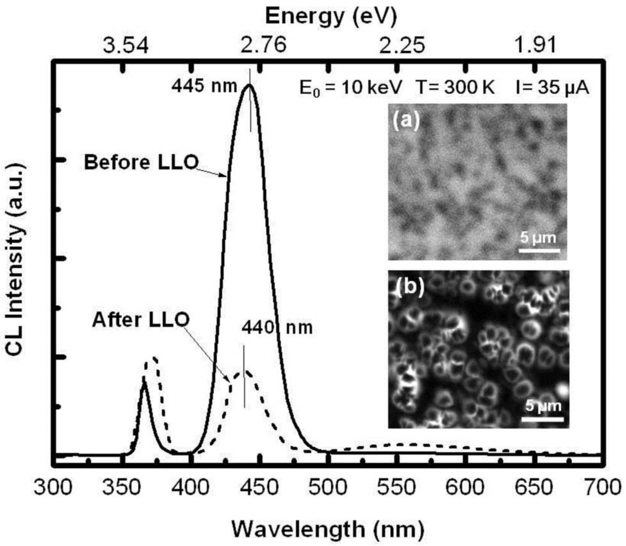

FIG. 2. CL spectra of the LEDs before and after the LLO. The insets show the CL images obtained with the monochromatic wavelength corresponding to the blue peaks of CL spectra (a) before and (b) after the LLO. The CL spectra and images are taken at the p-GaN side for the lateral LED and at the n-GaN side for the vertical LED after the thickness of the n-GaN layer was thinned down to 500 nm.

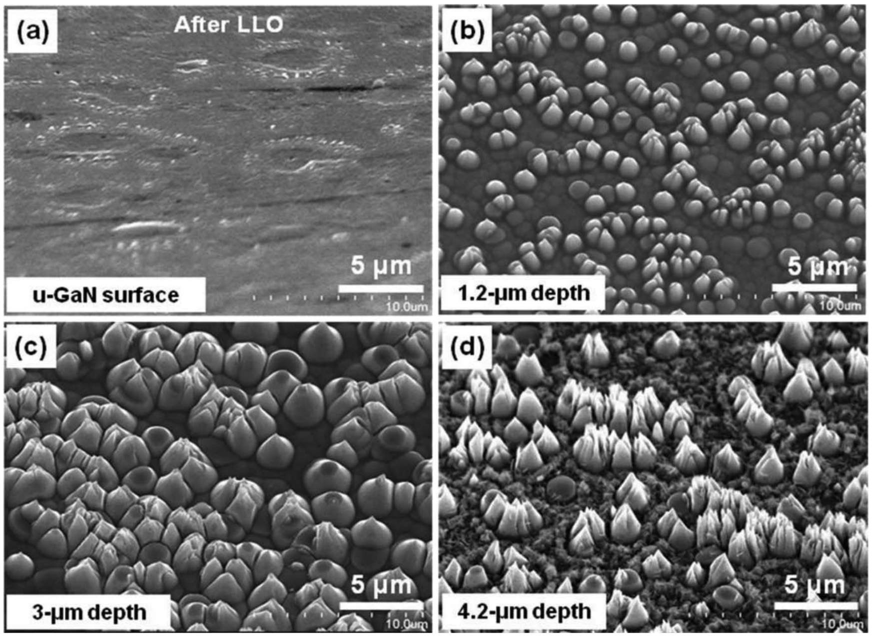

FIG. 3. SEM images of the GaN surfaces after the LLO process and ICP etching with various etching time for the positions of (a) u-GaN surface, (b) 1.2 μm, (c) 3 μm, and (d) 4.2 μm deep from the GaN/Al_2O_3 interface.

To understand how the propagation of KrF photons into the GaN layer affects the creation of dislocations, we have studied the variation of the cone size and density as a function of the depth from interface with sapphire substrate. As we see in Fig. 3, the cones are rather small with a high density just near the surface, and then undergo coalescence process, and finally the density of the cones decrease, while their size is still larger than the one near to the surface as the number of photons arriving at this region decreases. Figure 4 shows a typical tendency of such a behavior. The

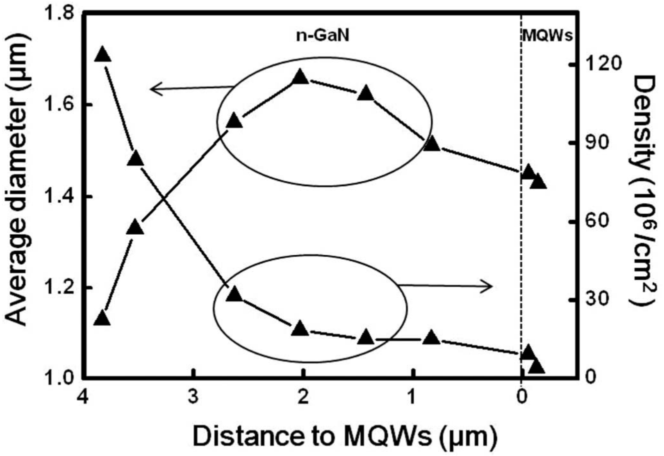

FIG. 4. Dependence of average size and density of the cones formed after the LLO and ICP etching processes. Note that size of the cones near to the MQWs is still larger than that near to the GaN/Al_2O_3 interface.

exact size of the defects varies from sample to sample. However, the trends for diameter and density of these defects remain for various samples.

The influence of such defect generation is studied more closely by performing the cross-sectional CL measurements on the points shown in the inset of Fig. 5(a), with large enough spatial intervals to avoid any overlaps of CL emitting regions. Taking into account the fact that CL intensity strongly depends on the defect concentration and thus material inhomogeneity, we have repeated CL measurements three times at slightly different lateral positions for a fixed vertical position, and then the averaged values are taken. Figure 5(a) shows the CL intensities at these points before and after the KrF laser irradiation to the sapphire substrate and Fig. 5(b) shows their differences. These figures show well that, after the KrF laser beam irradiation, the CL intensities of the spectral regions coming from the GaN and the MQWs decrease and the CL intensity of the yellow luminescence from the bottom n-GaN layer (MQW) increases appreciably (slightly). Therefore, it is clear that the LLO process induces some defects even in the MQW region as well as the bottom n-GaN layer when the thickness of the GaN layer below the MQW region is about 5 μm, as commonly adopted.

Figures 6(a) and 6(b) show the HRTEM images of the MQWs in the LEDs for which sapphire is non-irradiated and irradiated by the KrF laser, respectively. The MQWs are intermixed when sapphire is irradiated by the KrF laser with a laser fluence of 544 mJ/cm^2. The QW intermixing is also verified by the disappearance (existence) of sharp 2nd- and 3rd-order peaks in the HRXRD patterns when the sapphire substrates is irradiated (non-irradiated) by the KrF laser. Therefore, the blue-shift of the CL peak energy from the MQW (Fig. 2) is due to the intermixing of the InGaN/GaN MQWs, and the decrease of the MQW CL intensity after the KrF irradiation in Fig. 2 and Fig. 5 should be attributed to the creation of non-radiative recombination centers[15] in the course of the QW intermixing in addition to the formation of the dislocations in the bottom GaN layer. One would expect to observe a disordered structure of the InGaN/GaN MQW region in TEM images of the sample with intermixed QWs. However, we consider that the QW intermixing in our case is not complete and periodicity of the InGaN/GaN MQWs is not destroyed completely. We still observed InGaN layers in HRTEM images after the LLO, as shown in Fig. 6(b), but the InGaN/GaN interfaces are not sharp compared to those in the sample before the LLO [Fig. 6(a)]. The blue-shift in luminescence peak of the MQWs after the LLO may be caused by a reduction of In composition in the InGaN layers due to a diffusion of In through the LLO-generated defects.[16]

We believe that the laser fluence of the KrF laser employed in our LLO process (544 mJ/cm^2) is not so excessive, since this laser fluence is just slightly above the threshold,[4] and the sapphire substrate could not be removed when a KrF laser fluence of 485 mJ/cm^2 was used instead of 544 mJ/cm^2. Therefore, the results up to now mean that the structural defects may be created rather deep in the bottom layer even with a KrF laser fluence slightly exceeding the threshold fluence. We believe that

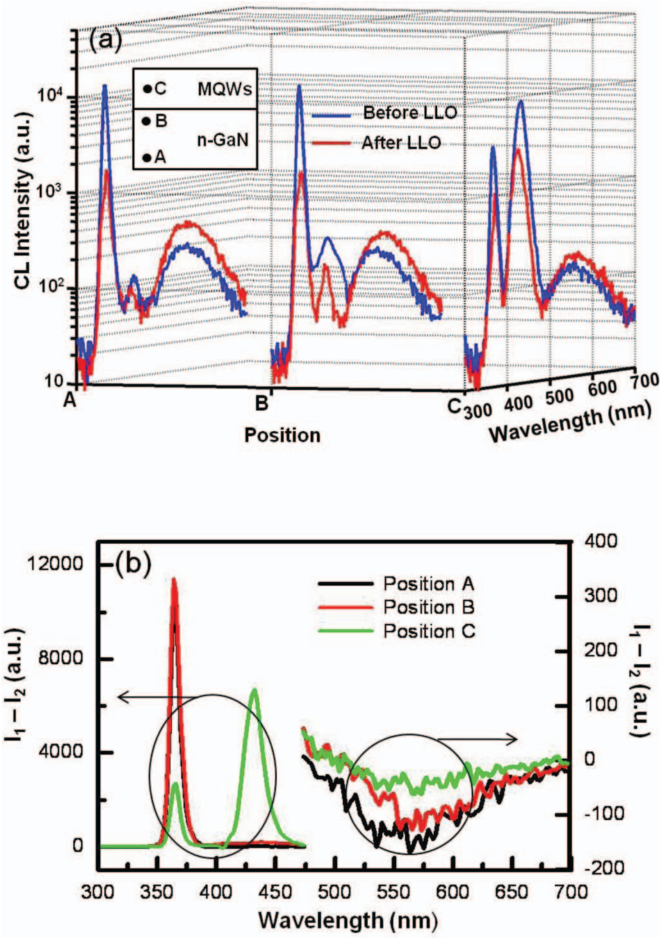

FIG. 5. (a) Cross-sectional CL spectra at the different positions of the LED structure before and after the LLO, and (b) the differences of the CL intensities at these positions. I_1 and I_2 are the CL intensities before and after the LLO, respectively. After the LLO, the CL intensities of the GaN and MQW emissions decrease while the yellow emission intensity increases.

the most probable energy source for the athermal solid state reactions needed in the defect formation and the QW intermixing could come from the photons and shock waves induced by the LLO process. The intermixing of the InGaN/GaN MQWs becomes appreciable when thermal annealing is performed around 1400°C for about 15 min,[17] which is physically irrelevant to our case. However, the electron-hole (EH) pairs generated by the 5 eV KrF photons may excite vibrational states in their recombination process needed for the defect reactions leading to the so-called recombination enhanced defect reaction (REDR).[18,19] Our KrF laser irradiation condition corresponds to about $7 \times 10^{17}/\mathrm{cm}^2$ photons impinging on the sapphire surface. About 25% of these photons are measured to be reflected and absorbed by the sapphire from transmission and absorption experiments. Counting the absorption of the 5 eV KrF photons by 5 μm thick GaN layer,[20] the number of photons arriving at the MQWs is calculated to be about $1.5 \times 10^{16}/\mathrm{cm}^2$. The EH pairs generated by these photons will reduce the effective activation energy of the migration through the REDR while they recombine.[18] The kinetic energy of electrons (holes) generated by the 5 eV KrF photons will also induce localized lattice vibrations while they move to the conduction (valence) band edge of the GaN and InGaN. Moreover, atomic vibrations caused by the propagation of the shock wave may also enhance the defect reactions. All these excitations might induce the observed athermal defect generation and QW intermixing.

To clarify which mechanism, between the generation of the EH pairs by the propagated KrF photons or the propagation of the thermal shock wave, is the main origin of atomic movements

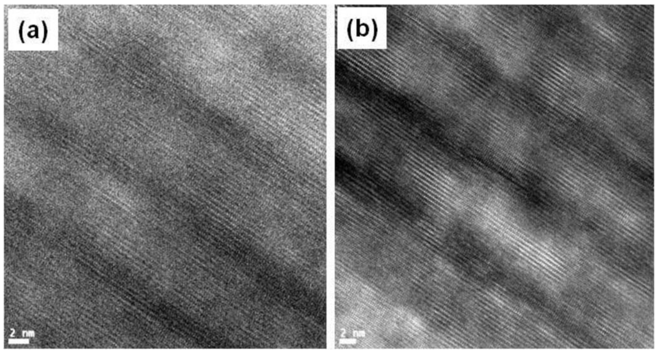

FIG. 6. HRTEM images of the MQW regions in the LEDs (a) before and (b) after the LLO process. The dark lines correspond to the InGaN quantum wells. The QW intermixing is observed after the LLO as in Fig. 6(b). The scale bars are 2 nm.

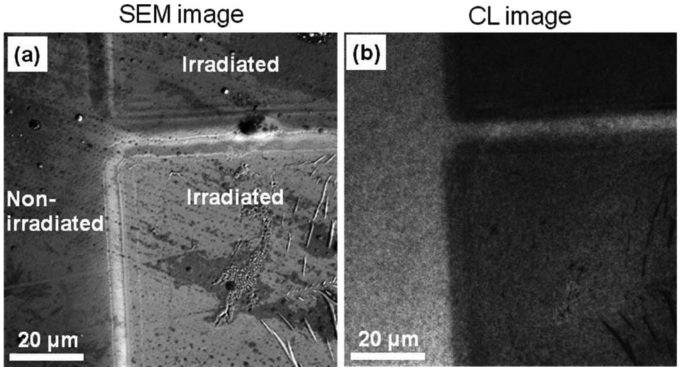

FIG. 7. (a) SEM and (b) corresponding monochromatic CL images taken at the p-GaN side of the LED after its sapphire substrate is irradiated by the KrF laser. The space between two laser steps, that is about 5 μm, is observed as a bright line in Fig. 7(b). Some scars and patterns observed in the SEM image are due to the contamination in the sample preparation.

inducing the dislocation generation and MQW intermixing, we have studied the position dependence of CL and SEM images assuming that the generated shock wave will propagate rather radially (Huygens' principle) while the KrF laser beam will propagate quite plainly. Figure 7 shows a typical monochromatic CL and SEM images taken at the surface opposite to the sapphire substrate after the KrF laser irradiation. Some scars and patterns are due to contamination during the sample preparation. It is observed from the monochromatic CL image of Fig. 7(b) that the width of non-irradiated region between the two KrF irradiated regions, the distance which is determined by the condition of the employed LLO process, remained nearly constant with a value of 5 μm, while the boundary between two regions was rather blurred. The panchromatic CL image showed similar tendency (not shown here), even through the boundary is observed to be more blurred. This fact manifests that the atomic movements needed for the observed dislocation generation and MQW intermixing are mainly due to the combined effects of REDR and the phonon generation as the generated electrons and holes move to their relevant band edges to recombine, while many people have argued that the generation and propagation of shock wave at/from the GaN/Al_2O_3 interface would be the main origin of the increase of the dislocations in the bottom GaN layer after the LLO process.[5–7] This reasoning can also explain how the QW intermixing may be so apparent even with a rather small number of the KrF photons arriving at the MQWs (1.5×10^{16}/cm^2). The difference between the KrF photon energy and the bandgap energy of GaN ($In_{0.3}Ga_{0.7}N$) is 1.6 (2.2) eV. This energy should be dissipated by the heat, generating the thermal phonons whose maximum energy in wurtzite GaN is about 30 meV.[21] Assuming the phonon energy, $\hbar\omega$, is comparable to the thermal energy, k_BT, at room temperature based on the high temperature approximation for phonon statistics, we can expect that nearly 10^2 order of thermal phonons would be generated for one KrF photon absorption. Additional influence of such atomic vibrations to the REDR mechanism would induce enough number of atomic movements needed for the QW intermixing.

IV. CONCLUSION

The influences of the LLO process on the optical and structural properties of the InGaN/GaN vertical blue LEDs are comprehensively investigated. We have shown for the first time that the damage effects of the LLO process can reach even to the active MQW region when the thickness of the GaN layer below the MQWs is conventionally taken to be 5 μm. It is also argued that the REDR process and the transition of electrons and holes from their photo-excited states to their relevant band edge states, rather than the propagation of the thermal shock wave from the GaN/Al_2O_3 interface, would mainly cause the damage effects of the LLO process.

ACKNOWLEDGMENTS

This work is supported by the National Research Foundation Grants (Mid-career Researcher Program: 2009-0083803 and KRF-2007-331-D00250). This work is also supported by the Ajou University research fellowship of 2009.

[1] J. K. Kim and E. F. Schubert, Opt. Exp. **16**, 21835 (2008).
[2] C. A. Tran, A. Osinski, R. F. Karlicek, Jr., and I. Berishev, Appl. Phys. Lett. **75**, 1494 (1999).
[3] F. Schulze, A. Dadgar, J. Blasing, A. Diez, and A. Krost, Appl. Phys. Lett. **88**, 121114 (2006).
[4] C. F. Chu, F. I. Lai, J. T. Chu, C. C. Yu, C. F. Lin, H. C. Kuo, and S. C. Wang, J. Appl. Phys. **95**, 3916 (2004).
[5] W. H. Chen, X. N. Kang, X. D. Hu, R. Lee, Y. J. Wang, T. J. Yu, Z. J. Yang, G. Y. Zhang, L. Shan, K. X. Liu, X. D. Shan, L. P. You, and D. P. Yu, Appl. Phys. Lett. **91**, 121114 (2007).
[6] Y. S. Wu, J. H. Cheng, W. C. Peng, and H. Ouyang, Appl. Phys. Lett. **90**, 251110 (2007).
[7] H. P. Ho, K. C. Lo, G. G. Siu, C. Surya, K. F. Li, and K. W. Cheah, Mater. Chem. and Phys. **81**, 99 (2003).
[8] A. Elgawadi and J. Krasinski, J. Appl. Phys. **103**, 033519 (2008).
[9] H. W. Huang, C. H. Lin, C. C. Yu, K. Y. Lee, B. D. Lee, H. C. Kuo, S. Y. Kuo, K. M. Leung, and S. C. Wang, Mater. Sci. Eng. B **151**, 205 (2008).
[10] M. H. Doan, H. Lim, J. J. Lee, D. H. Nguyen, F. Rotermund, and S. I. Mho, J. Korean Phys. Soc. **57**, 1295 (2010).
[11] X. Li and J. J. Coleman, Appl. Phys. Lett. **70**, 438 (1997).
[12] S. Pereira, M. R. Correia, E. Pereira, K. P. O'Donnell, C. T.- Cowan, F. Sweeney, and E. Alves, Phys. Rev. B **64**, 205311 (2001).
[13] J. Neugebaner and C. G. Van de Walle, Appl. Phys. Lett. **69**, 503 (1996).
[14] M. A. Reshchikow and H. Morkoc, J. Appl. Phys. **97**, 061301 (2005).
[15] M. Albrecht, J. L. Weyher, B. Lucznik, I. Grzegory, and S. Porowski, Appl. Phys. Lett. **92**, 231909 (2008).
[16] N. Duxbury, U. Bangert, P. Dawson, E. J. Thrush, W. Van der Stricht, K. Jacobs, and I. Moerman, Appl. Phys. Lett. **76**, 1600 (2000).
[17] M. D. McCluskey, L. T. Romano, B. S. Krusor, N. M. Johnson, T. Suski, and J. Jun, Appl. Phys. Lett. **73**, 1281 (1998).
[18] L. C. Kimering, Solid-State Electron. **21**, 1391 (1978).
[19] M. G. Kim, S. D. Kwon, C. H. Kim, J. B. Lee, B.-D. Choe, and H. Lim, Appl. Phys. Lett. **63**, 1366 (1993).
[20] O. Ambacher, W. Rieger, P. Ansmann, H. Angerer, T. D. Moustakas, and M. Stutzman, Solid State Commun. **97**, 365 (1996).
[21] T. Ruf, J. Serrano, M. Cardona, P. Pavone, M. Pabst, M. Krisch, D' Astuto, T. Suski, I. Grzegory, and M. Leszczynski, Phys. Rev. Lett. **86**, 906 (1993).