Materials Science in Semiconductor Processing 116 (2020) 105158

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: http://www.elsevier.com/locate/mssp

Performance-enhanced vertical LED using laser irradiation treatment to control wafer-level n-GaN protrusion arrays

Zhong-Liang Zhou [a,1], Cong Wang [a,1], Luqman Ali [a], Keun-Woo Lee [c], Zhao Yao [b,*], Ho-Kun Sung [c,**]

[a] School of Electronics and Information Engineering, Harbin Institute of Technology, Harbin, 150001, China
[b] School of Electronic Information, Qingdao University, Qingdao, 266071, China
[c] Korea Advanced Nano Fab Center (KANC), Suwon, 443270, Republic of Korea

ARTICLE INFO

Keywords:
Controllable
Surface texturing
Laser irradiation
Vertical LED

ABSTRACT

A controllable, mask-free, and wafer-level surface texturing method is proposed, which is applied to n-GaN protrusion array using laser irradiation treatment targeting to achieve high-performance vertical light-emitting diodes. The size and density of the n-GaN protrusions could be modified by controlling the energy density and pulse number of laser irradiation. Measurement results of current-voltage and light output power (LOP) reveal that the textured surface structure increases the LOP up to 47.89% at an injection current of 350 mA without affecting the current spread, such an enhancement is attributed to a higher probability that light could escape from the textured n-GaN surface.

1. Introduction

Group–III–nitrite-based light-emitting diodes (LEDs) are promising candidates for solid-state devices in lighting, panel displays, traffic signals, and many other applications [1,2]. Vertical thin-film gallium nitride (GaN) LEDs have recently been reported to improve the efficiency of high-power LEDs over conventional LED structures because of its improved current injection, excellent heat dissipation, reliability in high-power and value-added products [3–5]. Although the epitaxial layer is commonly grown on the sapphire substrate, the difference in the thermal expansion coefficients of the sapphire substrate and the epitaxial film significantly affects the LED performance, as well as the threading dislocations occurring on GaN epitaxial films. Furthermore, the inferior performance of the sapphire substrate in terms of conducting the heat generated in the GaN-based vertical LEDs (VLEDs) hinders high-power LED performance [6]. To resolve these issues, different methods are being pursued to improve the output power by enhancing the light-extraction efficiency (LEE) through random scattering from a roughened surface, these methods include nano-imprint lithography [7], photonic crystal structures [8], nanostructure arrays [9], patterned sapphire substrates [10,11], surface texturing by laser irradiation [12,13], and wet chemical etching [14]. In addition, although implementing a distinctive laser lift-off (LLO) procedure has been suggested to resulting in high output power for high-performance LEDs, this technique has not yet been fully investigated [4]. Hence, to overcome the aforementioned limitations and enhance the LEE along with the luminescence power, this paper proposes an alternative method of modifying a portion of the n-GaN epitaxial film with optimized laser irradiation conditions in order to generate an appropriate morphological structure that satisfies the optimum LEE condition.

Efficient GaN LED designs require excellent LEE to enhance the LED performance through total internal reflection (TIR) at the interface between n-type GaN (n-GaN) and air. However, because of the noticeable difference in the refractive index (n) of n-GaN ($n = 2.45$), air ($n = 1$), and TIR, the LEE is limited to few percentage points [15,16]. One of the most effective methods to improve the LEE is to roughen the surface of n-GaN by increasing the random scattering with KrF laser irradiation and KOH wet chemical etching [13,17]. In this study, a unique technique was introduced, in which the carrier metals are deposited on top of the VLEDs by wafer-level eutectic bonding after peeling the epitaxial film from the sapphire substrate with LLO technologies applied to a thin layer of undoped GaN (u-GaN). The patterned u-GaN thin-film on top of the

* Corresponding author.
** Corresponding author.
E-mail addresses: yzh17@qdu.edu.cn (Z. Yao), hokun.sung@kanc.re.kr (H.-K. Sung).
[1] Zhong-Liang Zhou and Cong Wang contributed equally to this work.

https://doi.org/10.1016/j.mssp.2020.105158
Received 9 November 2019; Received in revised form 24 March 2020; Accepted 21 April 2020
Available online 30 April 2020
1369-8001/

n-GaN layer has been effectively demonstrated to increase the output power by improving the LEE with surface texturing generated by KrF laser irradiation [18]. Although the laser irradiation technique has been developed over several years, many investigations of LLO and wet etching of the GaN layer have focused on improving the LEE, only a few research studies have focused on the effects of both the size and density of micro-patterned n-GaN structures on the LEE and the light output power (LOP) of the VLED. Among them, our group also implement a preliminary study [19], the main creativity of [19] is to decide whether and when have to introduce the LLO procedure in LED fabrication, meanwhile energy density of LLO can also be evaluated simply. This proposed paper, using the previous optimized fabrication processes mentioned in Ref. [19], analyzes energy density and pulse number of LLO in-depth and obtains better measurement results. Compared with the conventional LED, LOP is improved of 47.89% through twice equivalent calculation, whereas [19] is only improved around 24.8%.

In this study, various process conditions of peeling the n-GaN from the epitaxial film by laser irradiation are implemented and discussed. Focused ion beam (FIB) cross-sectional images of the fabricated LED chips illustrate the proposed structure and scanning electron microscope (SEM) images of power density for different laser irradiation intensities are presented, along with atomic force microscopy (AFM) images of the surface roughness generated by the n-GaN protrusion arrays. In addition, the influence of the LLO process on the efficiency of the proposed VLEDs is verified by analyzing output power in relation to the varied size and density of micro-scale GaN protrusion arrays, and electroluminescence (EL) spectrum measurements further verify the optimized LEE induced by the formation of micro-scale protrusion arrays on the n-GaN layer.

2. Experimental details

Fig. 1 shows the fabrication process of the proposed GaN VLED (size of 1×1 mm^2) with a micro-scale protrusion array using laser irradiation treatment. Step (a): The epi-layer of the VLED was grown on a 2-inch (0001) sapphire substrate by metal-organic chemical vapor deposition. The epi-layer consisted of a 300 Å GaN buffer layer, a 3.5-μm-thick u-GaN layer, a 3.8-μm-thick n-GaN layer, ten undoped periods of GaN/InGaN (30/100 nm) multiple quantum wells, and a 200-nm-thick p-type GaN (p-GaN) layer. Step (b): The high reflective Ni/Ag/Ni/Au (1/20/100/200 nm) metal system of as high as 95% reflectivity at 460 nm was then deposited by e-beam evaporation with an appropriate deposition rates of 1 Å/s, 2 Å/s, 3 Å/s, and 3 Å/s, respectively and annealed at 600 °C for 1 min to achieve a p-type ohmic contact metal and highly reflective mirror via rapid temperature annealing process. Step (c): Ni/[(Ti/Pt) $\times$ 6 times]/Ti/Au (200/[(100/30) $\times$ 6 times]/50/500 nm) barrier metals and the AgIn/Ag (2600/800 nm) bonding metal were deposited by e-beam evaporation with deposition rates of 3 Å/s and 5 Å/s for the barrier and bonding metal, respectively. Step (d): The sapphire substrate was then reversed, lapped, and polished to reduce its thickness following by the Ag–In wafer bonding process using a chemical mechanical polishing (CMP) technique (DS-Precision, DS Precision Industrial Co. Ltd.). Several parameters of the CMP process were fixed as follows: the slurry flow rate and the distance between the center of the table and the center of the head were 80 mL/min and 10 cm, respectively. Step (e): Subsequently, the VLED on the sapphire substrate was bonded to a graphite substrate as receptor at a bonding temperature of 320 °C and bonding pressure of 160 kgf for 360 s via wafer level bonding process [20]. Step (f): LLO (SLC-100, QMC) of the sapphire substrate using a 248-nm KrF excimer laser with a power of 700 mW, pulse wavelength of 248 nm, pulse width of 25 ns, and laser irradiation spot size of 4 mm $\times$ 4 mm was followed by Step (e) to separate the VLED with the sapphire substrate. Step (g): The metallic Ga residuals produced by the LLO process were removed through wet cleaning using diluted hydrochloric acid (HCl), and the remaining u-GaN layer was etched to 1.1 μm thick by inductively coupled plasma (ICP) etching using Cl_2 and BCl_3

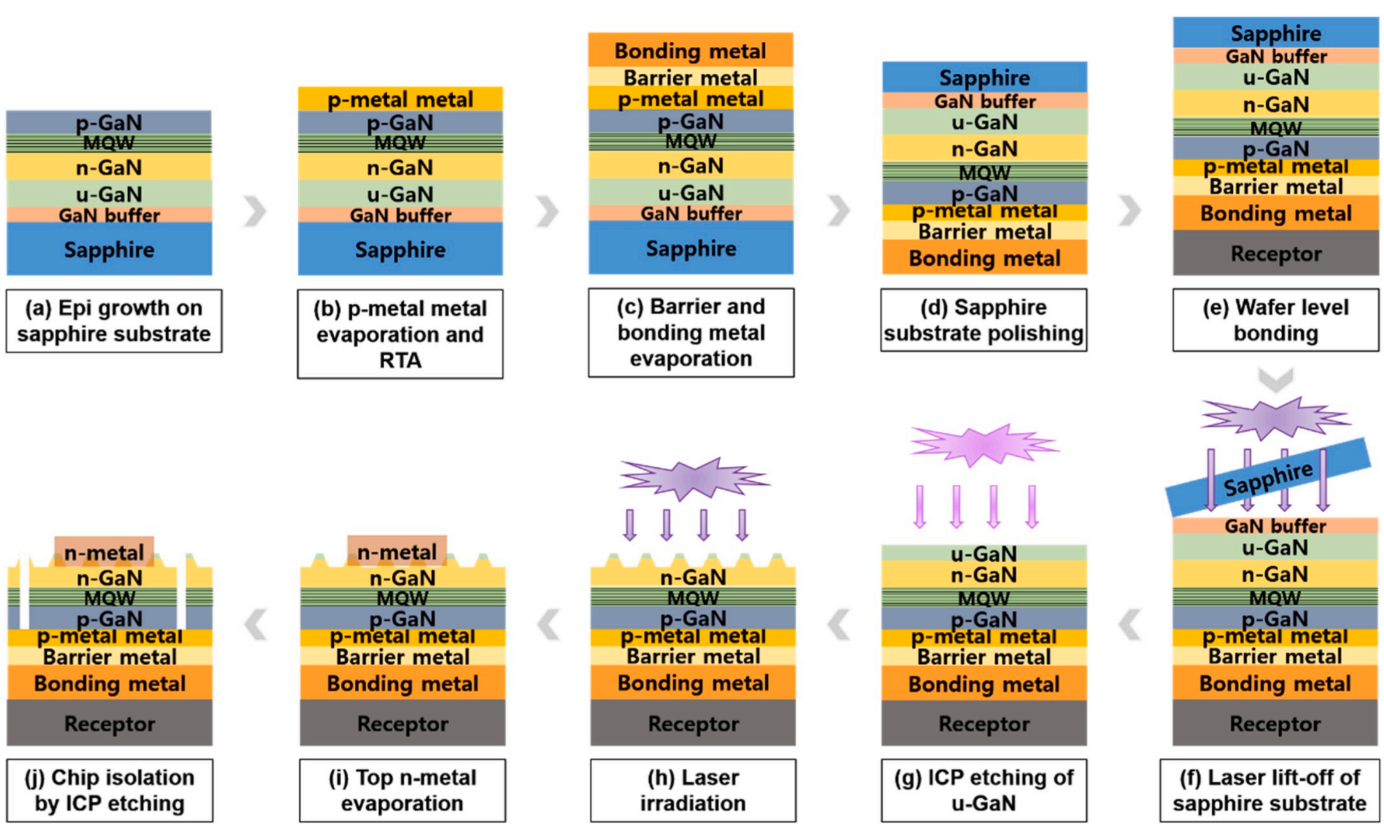

Fig. 1. Fabrication process for the proposed VLED with controllable n-GaN protrusion array produced by laser irradiation treatment.

gases with an ICP power of 900 W. Step (h): Then, the micro-scale protrusion array was formed by laser irradiation at energy densities of 550, 600, 650, and 700 mJ/cm^2, with the pulse number of 40, 60, 80, 100, 120, 140, respectively. Step (i): The top Cr/Au (10/500 nm) metal was deposited as n-type contact metal by e-beam evaporation with deposition rates of 2 Å/s and 3 Å/s following by the standard negative photoresist lithography process. Step (j): Finally, the top n-GaN layer was patterned by positive photoresist and etched by ICP reactive ion etching to isolate the chip under a Cl_2/BCl_3 plasma atmosphere. The VLEDs treated by laser irradiation under the energy densities of 550, 600, 650, and 700 mJ/cm^2 with an optimal pulse number of 60, were labelled as VLED-I, VLED-II, VLED-III, and VLED-IV, respectively. As a comparison, the VLED without laser irradiation treatment was also prepared as a reference data and labelled as VLED-Ref. Each controllable n-GaN protrusion array generated by laser irradiation was inspected by FIB (Quanta 3D FEG, FEI), SEM (S-4800, Hitachi), AFM (XE015, PSIA), and room temperature EL spectra (Robot, MaxMile Technol). The LOP of the fabricated VLEDs on 2-inch wafers were measured by a LED test prober.

3. Results and discussions

Fig. 2(a) shows the 3D schematic structure of the proposed VLED on a graphite substrate. A consistent pattern of circular truncated cone-like n-GaN protrusions were obtained on the surface of the top n-GaN layer, which was expected to enhance the LEE of the designed VLED by roughening the top surface and thus reducing the TIR [21,22]. The FIB data in Fig. 2(b) shows the layers of the fabricated VLED with the n-GaN protrusion array formed by laser irradiation at an energy density of 550 mJ/cm^2 and pulse number of 60. The combined thickness of the u-GaN and n-GaN layers was 7.3 μm. Then, after laser irradiation and wet cleaning, the minimum thickness of the remaining n-GaN layer was 2.99 μm, which was very close to the targeted thickness of 3.2 μm. The FIB data implied that a 0.81-μm thickness of n-GaN was etched, and at least a 0.66-μm thickness of u-GaN remained, because the cross-section of the protrusion labelled in Fig. 2(b) may not show the topmost point of the u-GaN layer. The data also indicated that all of the u-GaN layer and parts of the n-GaN layer were transformed into the protrusion array, and a thin u-GaN layer could protect the bottom n-GaN layer from further etching to avoid higher resistance [9]. The inset in Fig. 2(b) enlarges the Ti/Pt barrier layers, which are alternated six times to protect the above active layers from diffusion during the wafer bonding process. Fig. 2(c) compares the light exaction modes between the flat surface (dashed line) and patterned surface (solid line). For the flat surface, from the conventional VLEDs, most of the light emitted from the MQW layer will be trapped inside the GaN layer because of the TIR [23]. However, the n-GaN protrusion array enables light extraction from the patterned surface with angles even larger than the critical one, increasing the light escaping probability and enhancing the LEE. Therefore, the effect of the size and density of the fabricated protrusions on the performance of GaN-based VLED was further investigated. Fig. 2(d) shows an optical image of the fabricated VLED chips on a 2-inch wafer and an enlarged microscope image of a VLED chip with a size of 1 × 1 mm^2.

Fig. 3(b-1)-(b-6) and (c-1)-(c-6) show 2D and 3D AFM images of the surface morphologies of n-GaN protrusion arrays patterned by various pulse number at energy density of 600 mJ/cm^2 and 650 mJ/cm^2 in laser irradiation process, respectively. Circular truncated cone-like n-GaN protrusions were obtained on the surface of the top n-GaN layer after KrF laser irradiation, which broke the GaN bonds near threading dislocations to form the protrusion array. If suitable energy density was obtained, the number of GaN broken bonds and the pulse number would be positively correlated. As clearly seen in Fig. 3(b-1)-(b-6) and (c-1)-(c-6), which revealed the AFM results under the energy densities of 600 and 650 mJ/cm^2 and pulse number from 40 to 140 with the steps of 20. The root mean square (RMS) values increased initially, then decreased afterwards and finally increased again. Laser irradiation of 60 pulses broke more GaN bonds and generated larger and denser protrusions than others meanwhile the RMS value reached to be maximized. From 3D morphologies demonstrated in Fig. 3(b-3) and (c-3), with the top diameter decreasing when the number of laser irradiation pulse exceeded 60, protrusions became larger and bottom diameter increased rapidly. When the pulse number reached above 80, redundant pulse not

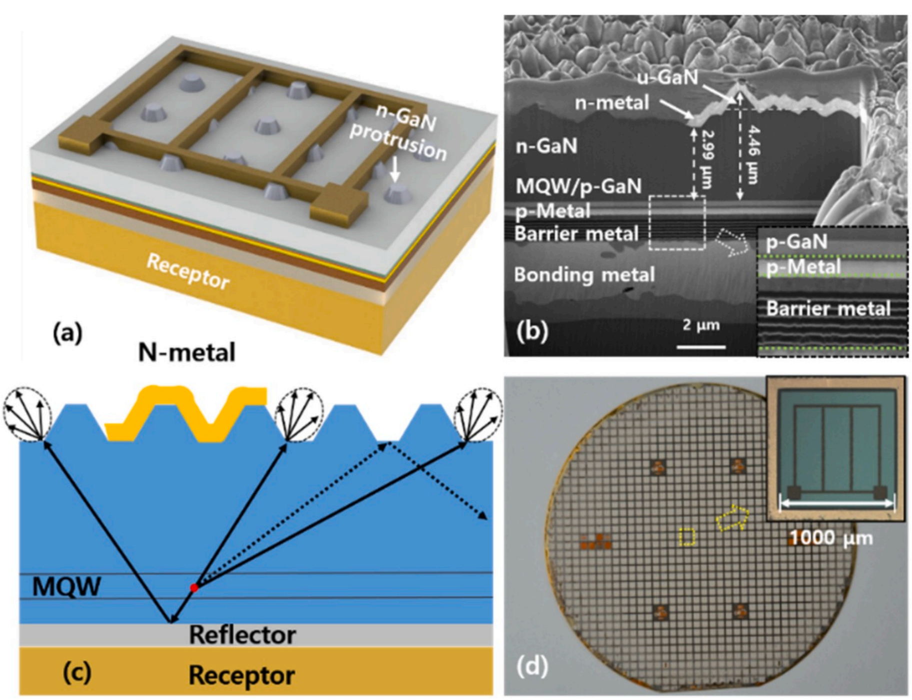

Fig. 2. The proposed VLED structure (a) 3D schematic of the proposed VLED with a chip size of 1 × 1 mm^2 on a graphite substrate, (b) cross-sectional FIB image of the proposed VLED; the inset details the p-metal and barrier metal region, (c) schematic of light extraction from traditional flat surface (dashed line) and optimized surface with protrusion array (solid line), and (d) optical image of the fabricated VLED on a 2-inch graphite substrate; the inset shows the top electrode structure.

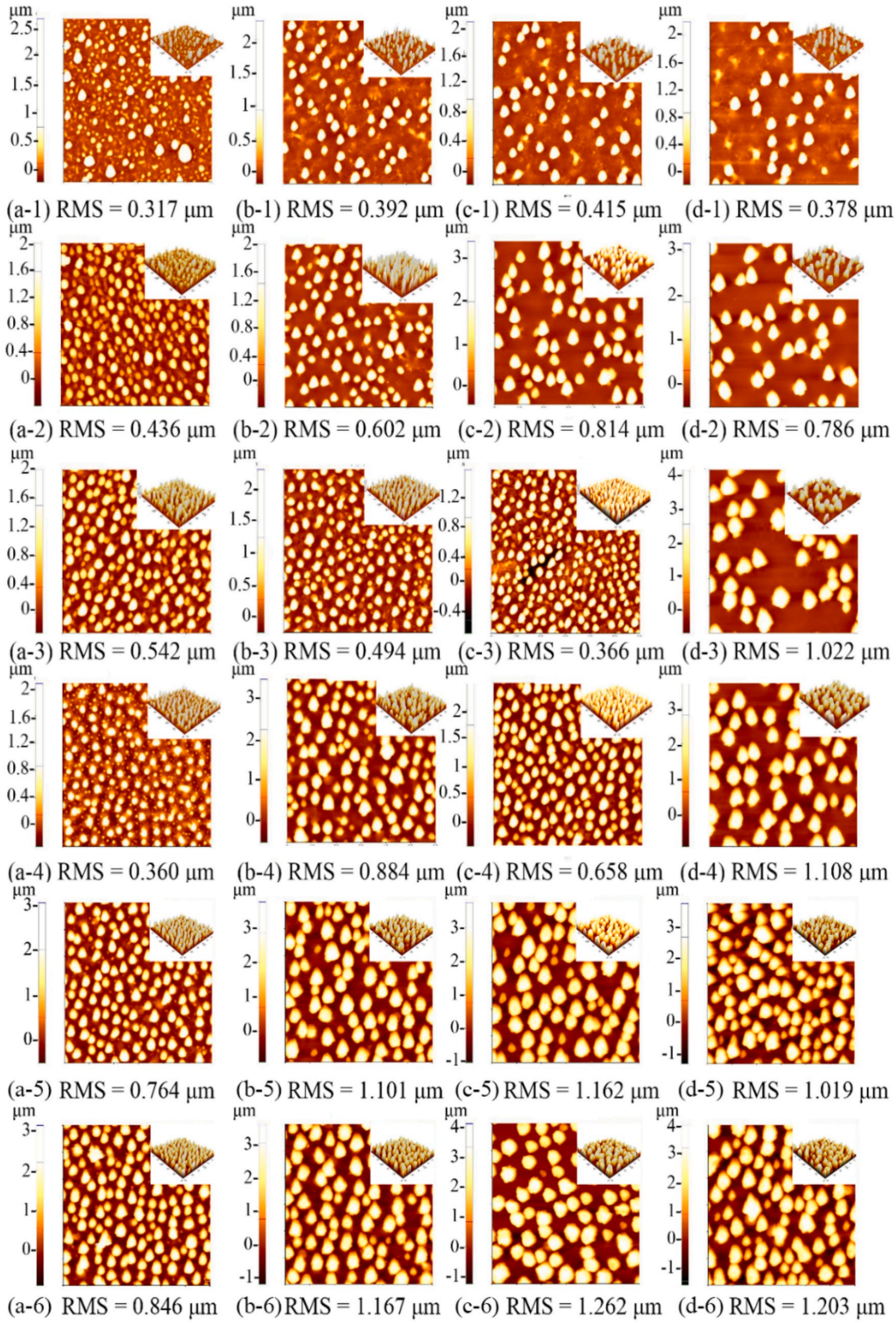

Fig. 3. Surface morphologies of n-GaN protrusion arrays fabricated by varying energy densities and pulse number of laser irradiation. (a)–(d) AFM images showing 2D surface morphologies for irradiation with energy densities of 550 mJ/cm^2, 600 mJ/cm^2, 650 mJ/cm^2, and 700 mJ/cm^2, respectively. (1)–(6) AFM images of 2D view for different pulse number of 40, 60, 80, 100, 120, and 140 for laser irradiation, respectively. Inserts are corresponding to theirs 3D views and the RMS values are attached to each figure with the size of AFM images is 60 μm × 60 μm.

only broken GaN bonds but also greatly etch the lower n-GaN epitaxial layer with GaN surface appeared micro-crack and had chance to cause lift-off damage, causing RMS increased unusually as displayed in Fig. 3 (b-4)-(b-6) and (c-4)-(c-6), respectively. Comparing to energy density of 600 and 650 mJ/cm^2, the RMS values of the samples treated under the energy density of 550 mJ/cm^2 also followed the same trend. But its first RMS peak emerged with the pulse number of 80 due to the low energy density, which employed the late generation of this phenomenon as shown in Fig. 3(a-1)-(a-6). However, when the energy density was in a higher level of 700 mJ/cm^2 as displayed in Fig. 3(d-1)-(d-6), the GaN protrusions continuously became big but sparse and altitude of protrusions grew lower with the bottom diameter increasing, which will cause the performance of VLEDs degradation. Meanwhile, the height indication bar also shown that high intensity of laser energy density etched the n-GaN epitaxial layer, causing the lighting reflection performance degradation [23,24], all of the related RMS data are summarized in Table 1.

Moreover, if the relative height difference between the protrusion of n-GaN and the depression of the epitaxial layer is larger than 4 μm, then the lighting reflection performance would get degradation [24,25]. The experimental results indicated that the relative height difference mainly depended on the variation of epitaxial layer depth. It had been found that the depth of the epitaxial layer depression varied more drastically compared to the height of protrusions. Therefore, the best pulse number was 60 and the main influence factor of performance was energy density, which was demonstrated by comparison via images and data.

Fig. 4 (a)-(d) show the SEM images of the surface morphologies of n-GaN protrusion arrays patterned by various energy densities with the same pulse number of 60 during the laser irradiation process. Well-aligned circular truncated cone-like n-GaN protrusions were obtained on the surface of the top n-GaN layer after KrF laser irradiation, which broke the Ga–N bonds near threading dislocations to form this protrusion array [13,26]. To further clarify the surface evolution under suitable pulse number of laser irradiation, 2D and 3D AFM images of the surface morphologies of n-GaN protrusion arrays under various laser irradiation energy densities were measured, as shown in Fig. 4 (a-1)-(d-1) and Fig. 4(a-2)-(d-2), respectively. In terms of the roughness value indicated by the RMS value, the highest roughness was achieved under an energy density of 650 mJ/cm^2, reducing when the energy density increased to 700 mJ/cm^2. More details are displayed compared to the above, as shown in Fig. 4(a-1), at the lower energy density of 550 mJ/cm^2, lots of small GaN protrusions were generated by the laser irradiation. With energy density rising, the number of broken Ga–N bonds also increased, which produced larger and less dense protrusions, as shown in Fig. 4(b-1), 4(c-1), and 4(d-1). As the energy density reached to 700 mJ/cm^2, as mentioned before, the bottom diameter of the protrusion continuously increased, whereas the top diameter decreased, the detailed description was displayed in Fig. 5(a). This result implied that the higher energy density may enhance the breaking of Ga–N bonds not only in the as-deposited n-GaN layer but also in the as-fabricated n-GaN protrusion, therefore, the height of the GaN protrusion was reduced at this stage as shown in Fig. 5(c). Both the height and density decreased at the energy density of 700 mJ/cm^2, and consequently, the roughness also decreased as shown in Fig. 4(d-2) when comparing with Fig. 4(a-2), Fig. 4(b-2), and Fig. 4(c-2).

Table 1
Measurement parameters of the generated n-GaN protrusions under different laser irradiation conditions.

Energy	Pulse	Height (μm)			
		Min	Max	Rpv (Max-Min)	Rq (RMS)
550	40	−0.240	2.583	2.823	0.317
	60	−0.376	2.107	2.483	0.436
	80	−0.282	2.369	2.652	0.542
	100	−0.311	2.070	2.382	0.360
	120	−0.509	3.061	3.57	0.764
	140	**−0.903**	**3.137**	**4.040**	**0.846**
600	40	−0.183	2.282	2.465	0.392
	60	−0.315	2.646	2.961	0.602
	80	−0.239	2.303	2.541	0.494
	100	**−0.606**	**3.540**	**4.146**	**0.884**
	120	**−0.900**	**3.766**	**4.666**	**1.101**
	140	**−1.176**	**3.638**	**4.814**	**1.167**
650	40	−0.218	2.032	2.250	0.415
	60	−0.352	3.394	3.746	0.814
	80	−0.691	1.593	2.284	0.366
	100	−0.426	2.487	2.913	0.658
	120	**−1.018**	**3.768**	**4.786**	**1.162**
	140	**−1.416**	**4.146**	**5.562**	**1.262**
700	40	−0.175	2.201	2.376	0.378
	60	−0.504	3.187	3.691	0.786
	80	**−0.645**	**4.157**	**4.802**	**1.022**
	100	**−0.913**	**3.703**	**4.617**	**1.108**
	120	**−1.311**	**3.742**	**5.053**	**1.019**
	140	**−1.620**	**4.043**	**5.663**	**1.203**

The data in Fig. 5 was based on laser irradiation under various energy densities with the same pulse number of 60, theirs AFM and SEM images were shown in Fig. 4. At the lower energy density of 550 mJ/cm^2, many small and tight protrusions generated and the size of protrusions grown with the energy density increasing. The well-aligned circular truncated cone-like protrusion became bigger and loosely tight when the energy density at 650 mJ/cm^2, which causing the better VLED performance due to the fact that the RMS reached maximum value. While the energy density reaching at 700 mJ/cm^2, the size of protrusions became thick and laser irradiation etched the epitaxial layer, causing performance reduction. As shown in Fig. 5(a), the average diameter range of the n-GaN protrusions significantly increased from 2.18-3.37 μm to 2.41–4.46 μm when the energy density of the laser irradiation changed from 550 to 700 mJ/cm^2 with the pulse number of 60. Meanwhile, the density of protrusion arrays was gradually decreased from 3.20×10^4 mm^{-2} to 9.60×10^3 mm^{-2}, as shown in Fig. 5(b). These data further indicate that the average size and density of the n-GaN protrusion array can be controlled by the energy density of the laser irradiation treatment. Fig. 5(c) and (d) provide detailed information on the n-GaN protrusion arrays from AFM analysis. Based on the calculated results, as the energy density increased from 550 to 700 mJ/cm^2 in increments of 50 mJ/cm^2, the average height of the fabricated protrusion array initially grew to 2.48, 2.96, and 3.74 μm, and then reduced to 3.69 μm. The surface roughness also shows a similar evolution compared with the height because the RMS roughness represents the sum of the squares of each height's difference from the mean value. Furthermore, the rates of growth in the average size, height, and roughness increase as the energy densities enhance. This phenomenon may result from the increase in the etching rate with enhancing energy density [23], which implies that controlling the energy density of the laser irradiation treatment can significantly modify the surface condition of the n-GaN layer. Therefore, the light extraction probability is considered to have a close relationship with surface roughness [26].

Although the density of the protrusions decreased, their average size and height increased when the energy density increased from 550 to 650 mJ/cm^2. Therefore, the final surface roughness of the textured GaN layer was enhanced at 650 mJ/cm^2, resulting in higher light extraction probability, which could improve the output power obviously. Furthermore, four VLEDs with different surface morphologies were investigated to evaluate the effect of the controllable n-GaN protrusion array on LOP in this study.

To investigate the effect of the textured surface formed by the n-GaN protrusion arrays on VLEDs, room temperature EL spectra of five types of VLED were also measured at an injection current of 350 mA, as shown in Fig. 6(a). The peak positions of the proposed VLED-I, –II, –III, -IV, and -Ref were measured to be 462.19 nm, 461.51 nm, 459.48 nm, 460.16 nm, and 462.37 nm, respectively. The slight shifting of the peak wavelength might be due to the measurement tolerance and fabrication processes. However, the enhanced EL intensity of VLED-III was ascribed to the increased surface roughness resulting from the variation of the n-

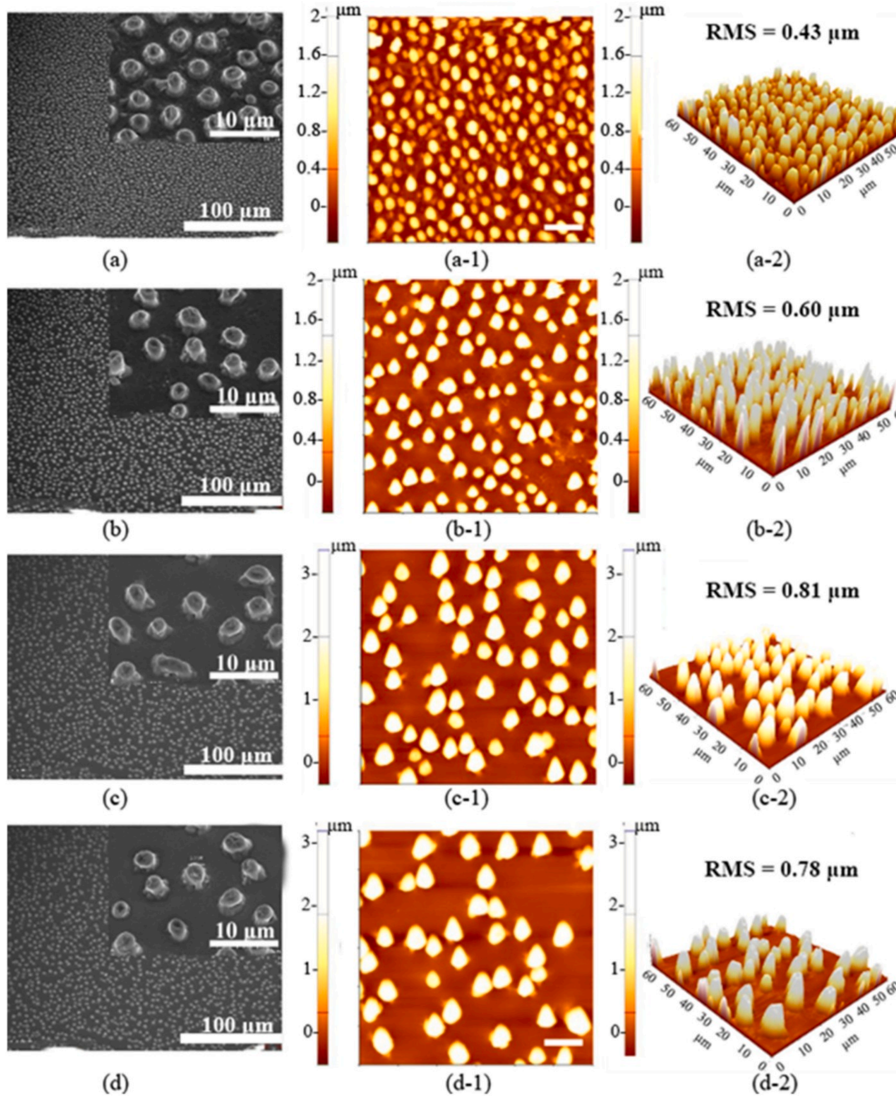

Fig. 4. Surface morphologies of n-GaN protrusion arrays fabricated by different energy densities of 550 mJ/cm^2, 600 mJ/cm^2, 650 mJ/cm^2, and 700 mJ/cm^2 and the same pulse number of 60 for laser irradiation, respectively. (a)–(d) SEM images for front view, (a-1)-(d-1) AFM images of 2D view, and (a-2)-(d-2) AFM images of 3D view. The size of AFM images is 60 μm × 60 μm.

GaN protrusion array, which agreed well with the aforementioned surface evolution. Therefore, the textured n-GaN protrusion array was proposed to enhance light extraction from the interface between n-GaN and air, and higher surface roughness was proposed to increase the output power. The measured current-voltage (I–V) and LOP characteristics of the VLEDs with and without laser irradiation treatment are shown in Fig. 6(b) and (c), respectively. All four types of VLED show similar I–V behaviors. The average forward voltage at an injection current of 350 mA was found to be 3.41 V, implying that the textured surface formed by laser irradiation has only a slight effect on the current spreading of the n-GaN film. The slight difference in forwarding voltage may be caused by the various heights of n-GaN protrusions [27].

Futhermore, the LOP exhibits a noticeable trend with various laser irradiation energy densities and pulse number. The average LOP of VLEDs with various n-GaN protrusions at a 350-mA injection current was measured to be 281.33, 303.81, 359.03, 313.05 mW, and 243.44 mW, corresponding to VLED-I, –II, –III, and -IV, and –Ref, respectively. Compared to the VLED without laser irradiation treatment, VLED-I, VLED-II, –III, and -IV showed enhanced LOP of 15.56%, 24.80%, 47.89%, and 28.59%, respectively. The highest improvement in LOP for VLED-III may be attributed to the roughened n-GaN surface, resulting in higher light escaping probability from the textured n-GaN surface. In order to adjust both the energy density and pulse number as the key parameters during the laser irradiation, an optimized energy density of 650 mJ/cm^2 and pulse number of 60 can be obtained for the specific VLED fabrication under a lower injection current.

In order to get maximum LOP, plenty of preparatory work [19] has been analyzed to decide whether and when introduce the LLO procedure in LED fabrication. Beside this, the improvement of [19] just consider two conditions (500 mJ/cm^2 and 600 mJ/cm^2) of energy density is insufficient. This proposed paper use the same optimized fabrication processes as comparison to ensure the improvement. Furthermore, through controlling two significant parameters (energy density and pulse number) of LLO as improvement variable, the performance of VLED is further analyzed in-depth with a better measurement result. The improvement of LOP in VLED-III is 47.89%, while [19] only improved 24.8%, both compared to VLED-Ref.

For this proposed work, the target is to exploring the LOP enhancement based on energy density and pulse number during the laser irradiation process and extra processes and higher injection current are not included in order to be cost-effective and save energy, thusly, making this work a potential candidate in the VLED market and the future

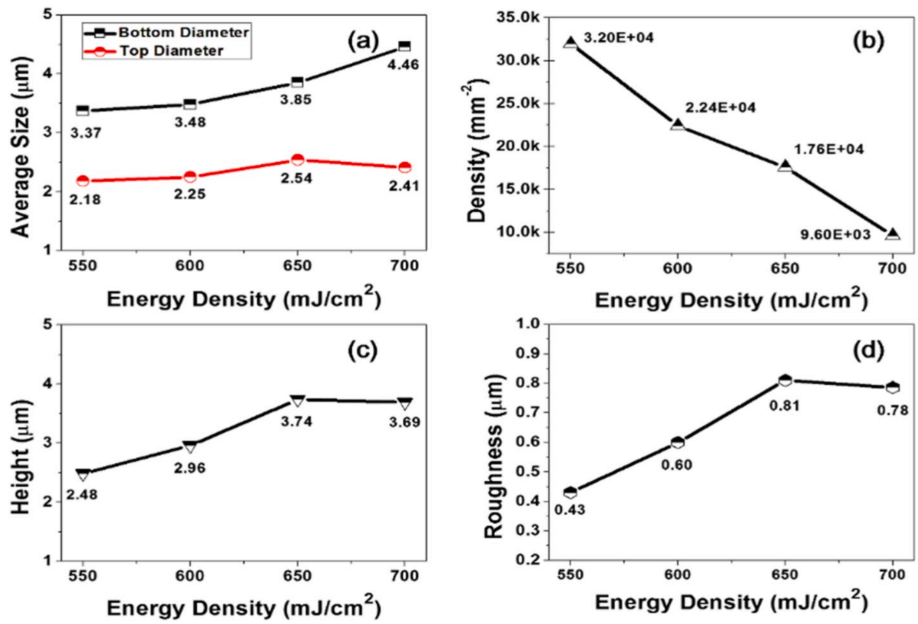

Fig. 5. Summary plots of the protrusion dimensions after laser irradiation under various energy densities. (a) Average top and bottom diameter of the circular truncated cone-like protrusion, (b) density of the protrusion array calculated from the SEM results, (c) height, and (d) surface roughness calculated by the AFM results.

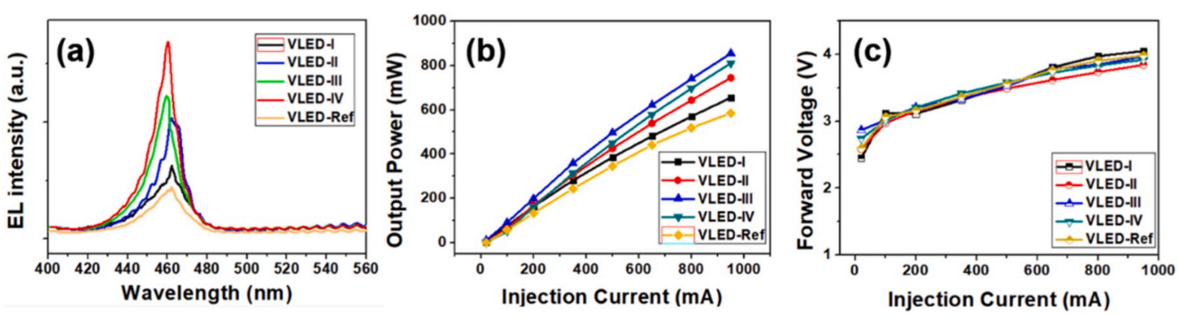

Fig. 6. Measurement result of the fabricated VLEDS. (a) Room temperature EL spectra at an injection current of 350 mA, (b) current-voltage curves, and (c) LOP characteristics of the fabricated VLEDs with different laser irradiation energy densities (VLED-I, VLED-II. VLED–III, and VLED–IV, and VLED-Ref).

micro-LED application.

4. Conclusions

In conclusion, a controllable, mask-free, and wafer-level surface roughness approach using laser irradiation for high-performance GaN-based blue VLEDs with the peak wavelengths of 459.48 nm was demonstrated. Through applying different laser irradiation energy densities and pulse number, various circular truncated cone-like n-GaN protrusion arrays were achieved, which were comprehensively evaluated through FIB, SEM, AFM and EL analysis. Four types of VLED with controllable surface conditions and the optimal pulse number of 60 were prepared by laser irradiation under different energy densities and compared with the unmodified VLEDs. Analysis results showed that the VLED with the well-modified surface treated at a laser irradiation energy density of 650 mJ/cm^2 exhibited the highest LOP, which was attributed to an enhanced LEE. Compared to the unmodified VLEDs, the improvement in the LOP was measured to be 47.89% at an injection current of 350 mA. Such a mask-free and controllable n-GaN protrusion array can be practical for VLEDs expecting to save costs in the mass production of high-performance LEDs.

Declaration of competing interest

The authors declare that they have no competing interests.

CRediT authorship contribution statement

Zhong-Liang Zhou: Methodology, Writing - original draft. **Cong Wang:** Methodology, Writing - original draft. **Luqman Ali:** Data curation. **Keun-Woo Lee:** Methodology. **Zhao Yao:** Supervision, Writing - review & editing. **Ho-Kun Sung:** Supervision, Methodology.

Acknowledgement

This work was supported by the General Financial Grant from the China Postdoctoral Science Foundation (2017M611367), Heilongjiang Postdoctoral Fund (LBH-Z17056), Fundamental Research Fund for the Central Universities, and sponsored by Zhejiang Lab (2019MC0AB03). This work was also supported by the National Natural Science Foundation of China under Grant No. of 61904092, the Shandong Provincial Natural Science Foundation of China under Grant No. of ZR2018BF027, and a Project of Shandong Province Higher Educational Science and Technology Program under Grant No. of J18KA382.

References

[1] J.H. Choi, A. Zoulkarneev, S.I. Kim, C.W. Baik, M.H. Yang, S.S. Park, Nearly single-crystalline GaN light-emitting diodes on amorphous glass substrates, Nat. Photon. 5 (12) (2011) 763–769.

[2] B.U. Ye, B.J. Kim, Y.H. Song, J.H. Son, H.K. Yu, M.H. Kim, J.L. Lee, J.M. Baik, Enhancing light emission of nanostructured vertical light-emitting diodes by minimizing total internal reflection,, Adv. Funct. Mater. 22 (3) (2012) 632–639.

[3] H.X. Yin, C.R. Zhu, Y. Shen, H.F. Yang, Z. Liu, C.Z. Gu, B.L. Liu, X.G. Xu, Enhanced light extraction in n-GaN-based light-emitting diodes with three-dimensional semi-spherical structure, Appl. Phys. Lett. 104 (2014), 061113.

[4] H.K. Sung, C. Wang, N.Y. Kim, Electrical and optical properties of laser irradiation treated vertical light-emitting diodes, Electron. Lett. 52 (12) (2016) 1041–1043.

[5] S. Kimura, H. Yoshida, K. Uesugi, T. Ito, A. Okada, S. Nunoue, Performance enhancement of blue light-emitting diodes with InGaN/GaN multi-quantum wells grown on Si substrates by inserting thin AlGaN interlayers, Appl. Phys. Expr. 120 (2016) 113104.

[6] V. Goyal, A.V. Sumant, D. Teweldebrhan, A.A. Balandin, Direct low-temperature integration of nanocrystalline diamond with GaN substrates for improved thermal management of high-power electronics, Adv. Funct. Mater. 22 (7) (2012) 1525–1530.

[7] H.H. Park, X. Zhang, Y. Cho, D.W. Kim, J.D. Kim, K.W. Lee, J. Choi, H.K. Lee, S. H. Jung, E.J. Her, C.H. Kim, A.Y. Moon, C.S. Shin, H.B. Shin, H.K. Sung, K.H. Park, H.H. Park, H.J. Kim, H.K. Kang, Wafer-scale surface roughening for enhanced light extraction of high power AlGaInP-based light-emitting diodes, Optic Express 22 (2014). S3:A723.

[8] T.B. Wei, Z.Q. Huo, Y.H. Zhang, H.Y. Zheng, Y. Chen, J.K. Yang, Q. Hu, R.F. Duan, J.X. Wang, Y.P. Zeng, J.M. Li, Reference efficiency enhancement of homoepitaxial InGaN/GaN light-emitting diodes on freestanding GaN substrate with double embedded SiO_2 photonic crystals, Optic Express 22 (S4) (2014) A1093.

[9] Y.C. Leem, N.Y. Kim, W.T. Lim, S.T. Kim, S.J. Park, Enhanced optical output power of InGaN/GaN vertical light-emitting diodes by ZnO nanorods on plasma-treated N-face GaN, Nanoscale. 6 (17) (2014) 10187.

[10] S.J. Tu, J.K. Sheu, M.L. Lee, C.C. Yang, K.H. Chang, Y.H. Yeh, F.W. Huang, W. C. Lai, Enhanced output power of GaN-based LEDs with embedded AlGaN pyramidal shells, Optic Express 19 (13) (2011) 12719–12726.

[11] C.F. Shen, S.J. Chang, W.S. Chen, T.K. Ko, C.T. Kuo, S.C. Shei, Nitride-based high-power flip-chip LED with double-side patterned sapphire substrate, IEEE Photon. Technol. Lett. 19 (10) (2007) 780–782.

[12] W.C. Lee, K.M. Uang, T.M. Chen, D.M. Kuo, P.R. Wang, P.H. Wang, S.J. Wang, “Enhanced light output of vertical-structured GaN-based light-emitting diode with surface roughening using KrF laser and ZnO nanorods” Jpn, J. Appl. Phys. 49 (2010), 04DG12.

[13] W.C. Lee, S.J. Wang, K.M. Uang, T.M. Chen, D.M. Kuo, P.R. Wang, P.H. Wang, Enhanced light output of GaN-based vertical-structured light-emitting diodes with two-step surface roughening using KrF laser and chemical wet etching, IEEE Photon. Technol. Lett. 22 (17) (2010) 1318–1320.

[14] C.E. Lee, Y.C. Lee, H.C. Kuo, T.C. Lu, S.C. Wang, Further enhancement of nitride-based near-ultraviolet vertical-injection light-emitting diodes by adopting a roughened mesh-surface, IEEE Photon. Technol. Lett. 20 (10) (2015) 803–805.

[15] J.K. Kim, A.N. Noemaun, F.W. Mont, D. Meyaard, E.F. Schubert, D.J. Poxson, H. S. Kim, C.S. Sone, Y.J. Park, “Elimination of total internal reflection in GaInN light-emitting diodes by graded-refractive-index micropillars, ” Appl, Phys. Lett. 93 (2008) 221111.

[16] J.H. Son, J.U. Kim, Y.H. Song, B.J. Kim, C.J. Ryu, J.L. Lee, Design rule of nanostructures in light emitting diodes for complete elimination of total internal reflection, Adv. Mater. 24 (17) (2012) 2259–2262.

[17] T.H. Lin, S.J. Wang, Y.C. Tu, C.H. Hung, T.H. Yu, Improving the performance of power GaN-based thin-film flip-chip LEDs through a twofold roughened surface, Mater. Sci. Semicond. Process. 45 (2016) 69–75.

[18] R.H. Horng, X. Zheng, C.Y. Hsieh, D.S. Wuu, Light extraction enhancement of InGaN light-emitting diode by roughening both undoped micropillar-structure GaN and p-GaN as well as employing an omnidirectional reflector, Appl. Phys. Lett. 93 (2008), 021125.

[19] Y. Li, Q. Wu, F.Y. Meng, H.K. Sung, Z. Yao, R.Y. Sun, C. Wang, Enhanced performance of large-area vertical light-emitting diodes treated by laser irradiation, Micro & Nano Lett. 12 (6) (2017) 369–372.

[20] Z. Yao, H.K. Sung, N.Y. Kim, Investigation of n-ohmic contact of vertical GaN-based light-emitting diodes on graphite substrate with Ag-In bonding, Mater. Sci. Semicond. Process. 59 (2017) 5–9.

[21] T. Fujii, Y. Gao, R. Sharma, E.L. Hu, S.P. DenBaars, S. Nakamura, Increase in the extraction efficiency of GaN-based light-emitting diodes via surface roughening, Appl. Phys. Lett. 84 (6) (2004) 855–857.

[22] C.C. Kao, H.C. Kuo, K.F. Yeh, J.T. Chu, W.L. Peng, H.W. Huang, T.C. Lu, S.C. Wang, Light-output enhancement of nano-roughened GaN laser lift-off light-emitting diodes formed by ICP dry etching, IEEE Photon. Technol. Lett. 19 (11) (2007) 849–851.

[23] C.F. Chu, F.I. Lai, J.T. Chu, C.C. Yu, C.F. Lin, H.C. Kuo, S.C. Wang, Study of GaN light-emitting diodes fabricated by laser lift-off technique, J. Appl. Phys. 95 (8) (2004) 3916.

[24] M. Sui, P. Pandey, M.Y. Li, Q.Z. Zhang, J.H. Lee, Au-assisted fabrication of nano-holes on c-plane sapphire via thermal treatment guided by Au nanoparticles as catalysts, Appl. Surf. Sci. 393 (2017) 23–29.

[25] P. Pandey, M. Sui, M.Y. Li, Q.Z. Zhang, S. Kunwar, J. Wu, Z.M. Wang, G.J. Salamo, J.H. Lee, “Nanoparticles to nanoholes: fabrication of porous GaN with precisely controlled dimension via the enhanced GaN decomposition by Au nanoparticles, ” Cryst, Growth Des. 16 (2016) 3334–3344.

[26] Y.J. Lee, H.C. Kuo, T.C. Lu, S.C. Wang, High light-extraction GaN-based vertical LEDs with double diffuse surfaces, IEEE J. Quant. Electron. 42 (12) (2006) 1196–1201.

[27] H.M. An, J.I. Sim, K.S. Shin, Y.M. Sung, T.G. Kim, Increased light extraction from vertical GaN light-emitting diodes with ordered cone-shaped deep-pillar nanostructures, IEEE J. Quant. Electron. 48 (7) (2012) 891–896.