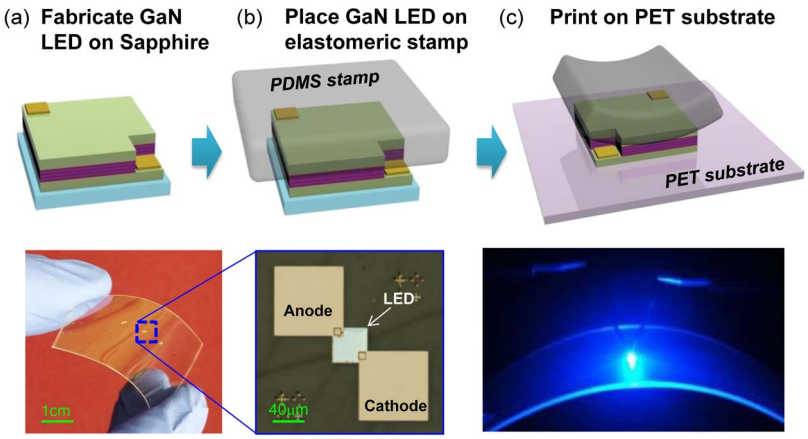
IEEE Photonics Journal
An IEEE Photonics Society Publication

Open Access

A Simplified Method of Making Flexible Blue LEDs on a Plastic Substrate

Volume 7, Number 2, April 2015

Jung-Hun Seo, Member, IEEE
Jing Li
Jaeseong Lee
Shaoqin Gong
Jingyu Lin
Hongxing Jiang
Zhenqiang Ma, Senior Member, IEEE



DOI: 10.1109/JPHOT.2015.2412459
1943-0655 © 2015 IEEE

IEEE photonics SOCIETY

IEEE

A Simplified Method of Making Flexible Blue LEDs on a Plastic Substrate

Jung-Hun Seo,[1] *Member, IEEE*, Jing Li,[2] Jaeseong Lee,[1] Shaoqin Gong,[3] Jingyu Lin,[2] Hongxing Jiang,[2] and Zhenqiang Ma,[1] *Senior Member, IEEE*

[1]Department of Electrical and Computer Engineering, University of Wisconsin–Madison, Madison, WI 53706 USA
[2]Department of Electrical and Computer Engineering, Texas Tech University, Lubbock, TX 79409 USA
[3]Department of Biomedical Engineering, Wisconsin Institute for Discovery, and Materials Science Program, University of Wisconsin–Madison, Madison, WI 53706 USA

DOI: 10.1109/JPHOT.2015.2412459

Manuscript received February 11, 2015; revised March 4, 2015; accepted March 6, 2015. Date of publication March 11, 2015; date of current version March 25, 2015. The work was supported by the Air Force Office of Scientific Research PECASE under Grant FA9550-09-1-0482. The program manager is Dr. G. Pomrenke. Corresponding author: Z. Ma (e-mail: mazq@engr.wisc.edu).

Abstract: A much-simplified method of making flexible GaN blue light-emitting diode (LED) array on a plastic substrate was demonstrated. A sticky elastomeric stamp was first brought into contact with prefabricated GaN LED array on a sapphire substrate. Laser liftoff was applied by shining laser light through the sapphire substrate. The released LED array sitting on the stamp was transferred to a polyethylene terephthalate substrate that was coated with an adhesive layer to finish the fabrication process. Careful investigation of the built-in stress in the GaN LED layer using Raman spectroscopy revealed that the maximum stress that allows for intact GaN LED layer release and transfer was 0.7 GPa. The method drastically simplifies the cumbersome conventional GaN layer transferring method while preserving the original layout of the GaN LED array. Due to its simple and practical characteristics, the method is expected to greatly facilitate the development of versatile transferrable GaN LED applications on various substrates at a much-reduced cost.

Index Terms: GaN LEDs, laser liftoff, direct transfer printing.

1. Introduction

Gallium nitride (GaN)-based light-emitting diodes (LEDs) have been widely used for high-performance solid-state lighting systems due to their high internal and external quantum efficiencies, low power consumption, and long-term stability [1]–[3]. Since 1999, after the first successful separation of a GaN layer from a sapphire substrate using ultraviolet (UV) laser scanning, [4] the process, referred to as laser lift-off (LLO), has been used extensively to transfer GaN-based LED structures from sapphire substrates to Si or metal substrates for their enhanced high-current operation capabilities [5], [6]. Recently, the LLO process has been used to transfer GaN-based LED structures onto flexible substrates so that they can be used for versatile flexible applications such as deformable displays and advanced biomedical devices [7], [15]. Flexible electronics, including flexible displays, are often implemented on large-area substrates. Considering the limited diameter size of single-crystal source substrates, inexpensive process and thus simplified processing procedures are highly desirable. To further reduce the cost of large-area

applications, inexpensive substrate such as polyethylene terephthalate (PET) is considered as the most desirable one among all others for its mechanical strength and large area [8].

However, the current LLO process requires a series of complicated steps to transfer-print GaN LED structure onto a flexible substrate, including bonding the structure to a hard substrate before LLO, selective removal of the bonding layer after LLO, transferring the flexible substrate using an elastomeric stamp, and another round of transfer-printing if a flip transfer is necessary [see Fig. S1(a)] [9], [16]. These complicated processing steps lead to a long process time and often cause a low transfer yield. Therefore, there is a strong need to develop a simplified method that can produce a higher yield transfer while preserving the original layout of the GaN LED array. A simpler transfer-printing method could also allow for the development of a more cost-effective mass production process for flexible devices with complicated layouts. The most recent progress on simplifying the fabrication of flexible LED was reported by Won-Sik *et al.* [10]. However, the realization of the processing procedures relies on the use of polyimide substrate that has much higher-temperature tolerance and is much more expensive than PET. In addition, the demonstrated process cannot be applied to a substrate that is greater than the sapphire substrate, limiting the use of this process for large-area applications. Furthermore, considering the required LED fabrication temperature after finishing their laser lift-off, PET substrates (glass transition temperature: 170 °C), even with the same diameter size as the sapphire substrate cannot be used.

To substantially simplify the fabrication process of flexible GaN LEDs and make the process easily applicable to any types of flexible substrates of any size, we first explore the dependence of LLO-released GaN film quality on the GaN thickness. We performed the LLO process on 1 cm^2 diced GaN epi substrates with various thicknesses grown on a double-side polished sapphire substrate while attaching the GaN layer with a poly-dimethylsiloxane (PDMS) stamp.

2. Experiment

Fig. 1(a) shows the GaN LED layer structure. The InGaN/GaN MQWs LED structures used in this study were all grown by metal–organic chemical vapor deposition (MOCVD). With the exception of the 1.3 μm thick LED, prior to the growth of the LED structure, an undoped GaN epi-template (1 μm) was first deposited at $\sim$530 °C on a 2-in double-side polished c-plane sapphire substrate to serve as a dislocation filter. This was then followed by the growth of a highly conductive Si-doped n-GaN $(n = 3 \times 10^{18}\ cm^{-3})$ cladding layer at 1100 °C, the 8-period $In_{0.2}Ga_{0.8}N$/GaN multiple quantum wells (MQWs) active region, and then a Mg-doped p-GaN layer with a thick of about 300 nm $(p \approx 1 \times 10^{18}\ cm^{-3})$ at 1040 °C. The thickness of the n-GaN layer was varied from around 0.8 to 3.3 μm. The 1.3 μm thick LED structure was directly deposited on a sapphire substrate with a 30 nm thick undoped GaN epi-template and the thickness of the n-GaN layer was about 1 μm. Each sample was diced into 1 cm^2 size prior to the test. The composition of the $In_{0.2}Ga_{0.8}N$/GaN MQWs active layers were kept the same for all of the samples except the thicknesses of the Si doped n-GaN (GaN:Si) layer, which was used to vary the total layer thickness of the GaN layer structure from 1.3 to 4.6 μm. It is noted that the thinner is the total GaN layer thickness, the lower is the epi-growth cost. The LLO process under the same conditions was carried out to separate the GaN layer from the sapphire substrate. Fig. 1(b) shows the average size (i.e., the length and width) of the transferred GaN layer pieces as a function of the GaN layer thickness. As shown in Fig. 1(b) and (c), when the thickness of the GaN layer was thicker than 4 μm, the GaN layer became intact (i.e., undamaged) after the entire LLO and transfer processes. It should be noted that the determination of fracture-free GaN layer thickness is solely related to the built-in stress and not to the LLO process conditions, as described below. Instead, our LLO process conditions, such as laser power, duration, pitch, were first optimized following the conventional LLO method such that a 2.1 μm thick GaN LED layer was able to be separated from sapphire substrate. It is also noted from Fig. 1(c) that the cracked GaN layer (when thickness less than 4 μm) showed a directional orientation (i.e., the horizontal direction) such that the fractured GaN layer pieces were longer along the laser scanning direction.

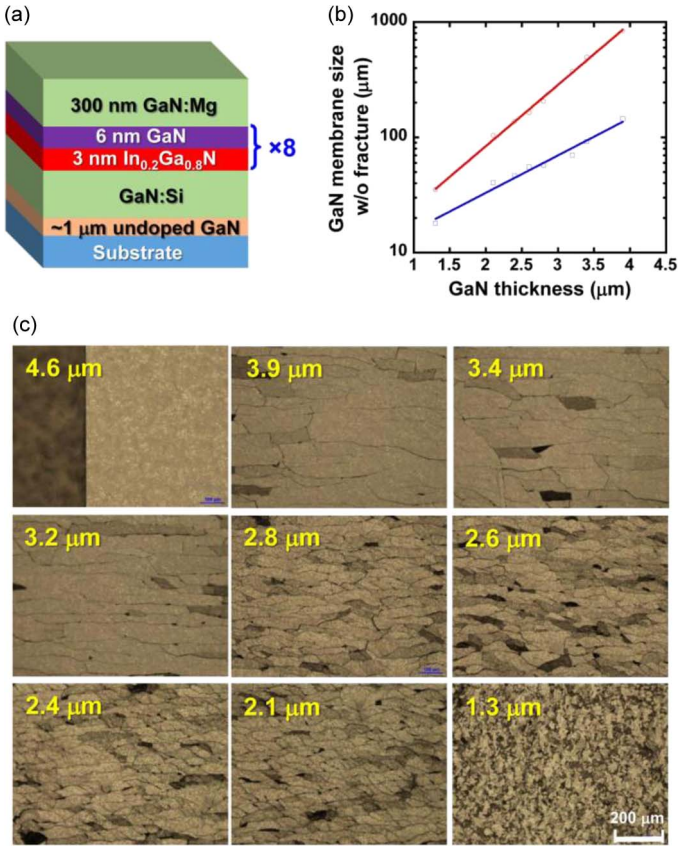


Fig. 1. (a) Schematic of the GaN LED structure grown on a c-plane sapphire substrate. (b) Sizes of transfer-printed GaN layers without fracture as a function of the thickness of the GaN layers after the laser-lift off (LLO). (Red: width; Blue: length). (c) Microscopic images of the GaN LED layers with different thicknesses on a PDMS stamp after the LLO process. The scale bar for all images is the same as the one shown in the last image.

3. Results and Discussion

It is always desirable to use a thin GaN LED layer. However, the GaN LED layer grown on foreign substrates needs to be at least a few microns thick in order to achieve a good GaN crystal quality. For this reason, it is inevitable to have a built-in stress in a GaN LED structure. Raman spectroscopy studies were employed to understand the relation between the fracture pattern of the GaN layer and its built-in stress [11]. Raman spectra were taken both from the GaN layer structure on a sapphire substrate and from the released GaN layers that transferred to a PET substrate using a Horiba LabRAM ARAMIS Raman confocal microscope with an 18.5 mW of He-Ne (532 nm) green laser. It should be noted that the PDMS stamp bonds firmly to the LED samples upon employing desired mixing ratio between PDMS solution and its curing agent. The elastic and soft nature and the ultra surface smoothness of PDMS further enable ultra bonding uniformity between PDMS and LED samples. No extra mechanical bonding procedure or special tool is needed to realize such high fidelity bonding. The strong bonding over the entire contact area sustains during LLO process. The PDMS stickiness is also sustained for multiple times of the LED sample attachment. Fig. 2(a) shows the typical Raman spectra of a 4.3 μm thick GaN layer on a sapphire substrate and the released GaN layer on a PET substrate, along with a spectrum taken from a remaining sapphire substrate (after release of its GaN layer) for comparison. The E_2 phonon mode of the GaN layer was located at 570.7 cm^{-1} for both GaN layers. Fig. 2(b) shows that the Raman peaks of the GaN layers on the sapphire substrate decreased from 575.6 cm^{-1} to 573.7 cm^{-1} when the GaN layer thickness decreased from 4.6 μm

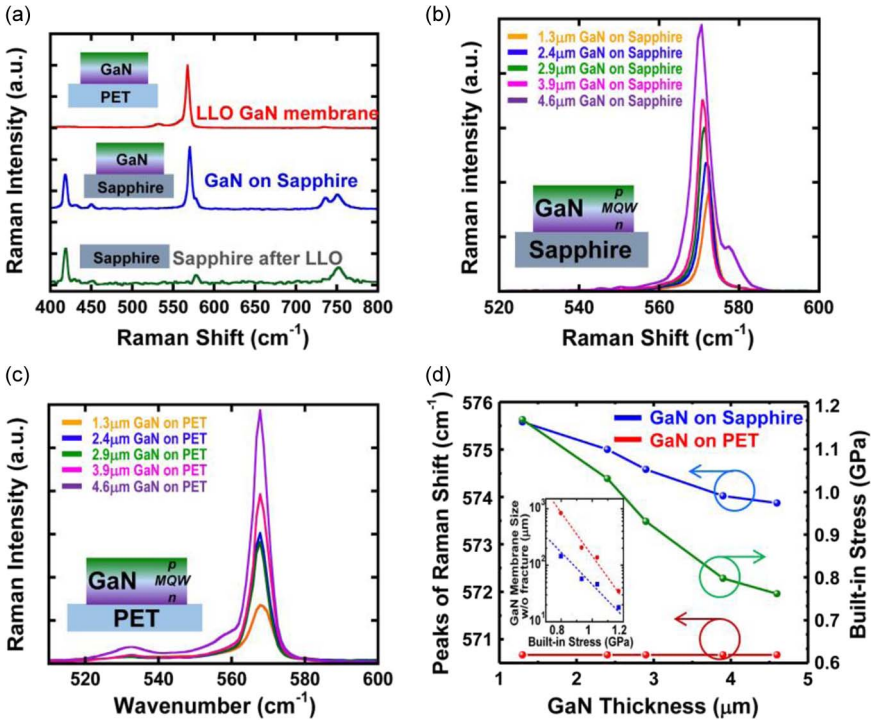


Fig. 2. (a) Raman spectra of the GaN LED structure on a sapphire substrate (blue), sapphire substrate after laser lift-off (green), and the LLO GaN layer on a PET substrate (red). (b) Comparison of Raman spectra of the GaN LED layers with various thicknesses grown on a sapphire substrate. (c) Comparison of Raman spectra of the LLO GaN LED layers with various thicknesses after transfer-printed on a PET substrate. (d) Raman peak and the calculated built-in stress as a function of the GaN layer thickness. Blue: Raman peak of the GaN layer grown on a sapphire substrate (i.e., before LLO); Red: Raman peak of the GaN layer transferred on a PET substrate after LLO. Green: Calculated built-in stress measured from the GaN layer grown on a sapphire substrate. The lines are used for guiding the view of the data points. The inset plots the dependence of the released GaN membrane size (Red: width; Blue: length.) versus built-in stress.

to 1.3 μm. The right shift of the Raman peak indicates that the GaN layer on the sapphire substrate possessed a smaller compressive stress as the GaN layer became thicker. In contrast, the Raman peak positions remained at the same position (i.e., 570.7 cm^{-1}) for the released GaN layer after being transfer to the PET substrate, regardless of the thickness [see Fig. 2(c)]. This value is the same as the typical value for unstressed GaN. The complete relaxation of built-in stress of the GaN on PET is considered valuable for practical flexible and bendable applications. Fig. 2(d) plots the comparison of the Raman shifts between the GaN layer on a sapphire substrate and that on a PET substrate with varying GaN layer thicknesses. In the figure, the calculated built-in stress for the sapphire case using the equation proposed by Kisielowski *et al.* [11] was also plotted. The measured Raman spectra show that the GaN E_2 phonon peaks were right-shifted by 4.9 cm^{-1} and 3 cm^{-1} for the 1.3 μm and 4.6 μm thick GaN layers and they correspond to a compressive stress (σ_{GaN}) of 1.17 GPa and 0.71 GPa, respectively, using typical parameters of hexagonal GaN [11]. We speculate that stress smaller than 0.71 GPa can be absorbed by the soft and elastic stamp during the LLO process, so that the separated GaN layer can remain intact. The inset of Fig. 2(d) shows the dependence of released GaN membrane size versus the built-in stress (for the sapphire case), verifying that the built-in stress, instead of LLO process conditions, is responsible for the GaN layer fracturing. Combining the built-in stress analyses with the visual observation of the LLO GaN layer, we believe that the GaN layer can remain intact during PDMS supported LLO process when the built-in stress is less than 0.7 GPa. Further optimization of PDMS composition and curing conditions may allow some more tolerance of built-in stress in GaN layer (thus thinner GaN) for obtaining fracture-free GaN layer using LLO process.

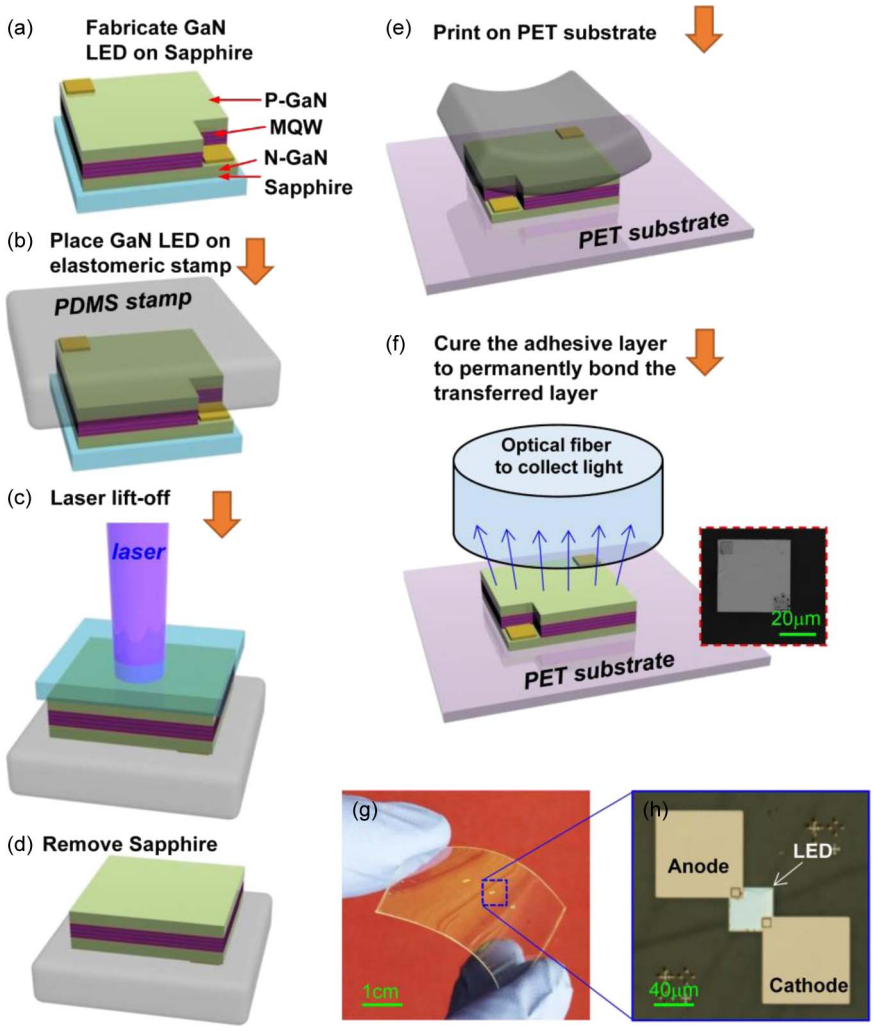


Fig. 3. Schematic of the fabrication process for GaN LEDs on a PET substrate using a laser lift-off (LLO) and transfer-printing method. (a) Fabrication of GaN LEDs on a sapphire substrate. (b) Place fabricated GaN LEDs on a PDMS elastomeric stamp. (c) Laser lift-off from the sapphire side. (d) Gentle removal of the sapphire substrate. (e) Transfer-print GaN LED layer onto the PET substrate. (f) Curing of the adhesive layer to permanently bond the transfer-printed GaN LED layer. An optical fiber is placed on top of the GaN layer for light collection. (g) Image of fabricated LED array on a bent PET substrate. (h) Zoomed-in image of an individual LED on PET.

The above study allowed us to demonstrate a much simplified method to make GaN LEDs on any flexible substrate of any size. Here we employed PET as the demonstration substrate. Fig. 3 shows the fabrication processes and Fig. S1 provides a comparison between our method and the conventional LLO method [16]. A 1 cm^2 diced 4.6 μm thick GaN epi substrate was used to fabricate LEDs. To obtain the highest LED performance, the entire LED fabrication procedures, including thermal anneals, were carried out on sapphire substrate [see Fig. 3(a)]. The p-GaN and MQW layers with an area of 2500 μm^2 and a depth of 800 nm was etched using inductively coupled plasma reactive ion etching (ICP-RIE, PlasmaTherm 770 ICP) to expose the n-GaN layer. After finishing the etching of the n-GaN layer, the GaN LED active area was defined by photolithography and further etched down to the sapphire layer to isolate individual GaN islands. An electron beam-evaporated Ni/Au (30/150 nm) layer stack was used for p-GaN and a Ti/Al/Ni/Au (30/100/20/150 nm) layer stack was used for n-GaN to provide respective ohmic contacts. The sample was then annealed at 500 °C for 1 min using rapid thermal anneal

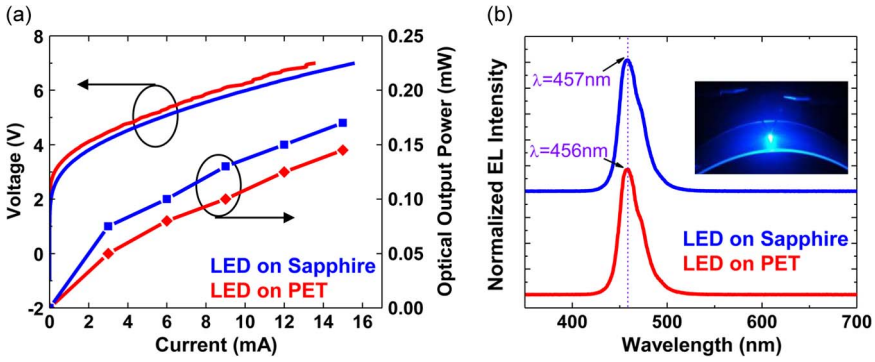


Fig. 4. (a) Light output power-current-voltage (L-I-V) characteristics and (b) electroluminescent light spectra of the GaN LEDs on sapphire substrate (before laser lift-off) and PET substrates (after transfer-printing). The insert in Fig. 4(b) shows the light emission images on a PET substrate.

(RTA) to enhance the ohmic contact property. Thereafter, the sample was flipped and the GaN layer was made in tight contact with a PDMS stamp [see Fig. 3(b)]. Note that the PDMS base to curing agent ratio was carefully adjusted to 1 : 20 (by weight) in order to obtain the proper stickiness from the PDMS stamp for the desired tight contact. The LLO process was performed by shining laser light (diode-pumped solid-state laser: $\lambda = 335$ nm; beam spot: 150 μm; line scan speed: 50 mm/sec) through the sapphire substrate [see Fig. 3(c)]. After separating the GaN LED layer (now sitting on the PDMS stamp) from the sapphire substrate, the sample was dipped into a 1 : 10 diluted HCl solution immediately to remove any Ga residues on the separated GaN layer [see Fig. 3(d)]. The separated LED layer was then transferred to a PET substrate (of any size) that was coated with an adhesive layer (SU-8, Microchem, sticker than PDMS) with nearly 100% of yield [see Fig. 3(e)] [16], [17]. Subsequently, a flood exposure was performed to cure the adhesive layer in order to permanently bond the transfer-printed GaN LED layers to the PET substrate [see Fig. 3(f)]. A 1.8 μm thick SU-8 was spun on the GaN LED printed PET substrate and patterned for interconnection. The sample was then dipped into acetone before the SU-8 was completely cured to make a smoother topological profile. Finally, the interconnected metal lines were patterned and deposited.

The electrical and optical characteristics of GaN LEDs were measured using a 4155B Agilent semiconductor parameter analyzer and a spectrometer (USB 2000, Ocean Optics), respectively. The light output from the LEDs was measured using an optical fiber that was placed on top of the GaN layer (note: GaN was coated with SU-8) [see Fig. 3(f)]. No melting of degradation of PET substrate was observed during LED measurements. Fig. 4(a) shows the comparison of the I-V characteristics under wafer probing between an unpackaged GaN MQW LEDs on a sapphire substrate (unreleased) and that on a PET substrate produced using the above described method. The GaN LEDs on both types of substrates showed a similar turn-on voltage of 3.3–3.5 V. The LED output powers before and after the LLO process were very close to each other and both increased linearly as the injection current increased to 15 mA. The slightly lower optical power from the LED on PET is considered due to SU-8 coating. Fig. 4(b) shows the measured EL spectra with a 10 mA current for the LEDs on both types of substrates. The EL spectral peak was located at 457 and 455 nm for the LEDs on the sapphire and on the PET substrate, respectively. The full width at half maximum (FWHM) of the EL spectral peaks of the two LEDs were 27 and 30 nm, respectively. The slight difference in the EL spectral peaks from the GaN LEDs on both types of substrates before and after LLO process is partially caused by the relaxed compressive stress and the band filling effects in the GaN LED layer, as described previously [12]–[14]. PET has poorer thermal conductivity than sapphire. A slight increase in LED junction temperature in comparison to a sapphire substrate is thus expected. However, the effects of junction temperature increase on the LED characteristics are unknown at the moment, and further study may be needed.

4. Conclusion

In summary, a much simplified method of fabricating GaN LEDs on any flexible substrate of any size has been established using a stickiness-controlled PDMS elastomeric stamp as the LLO-temporary holder and post-LLO transfer medium. Careful investigation of the built-in stress present in the GaN LED layer on a sapphire substrate revealed the maximum built-in stress and the minimum GaN layer thickness without fracture that are required for the simplified method. The practical application of the demonstrated method is expected to lead to reduction of the production cycle time and cost. The method may also be potentially adapted to a wide range of other transferrable devices.

References

[1] S. D. Lester, F. A. Ponce, M. G. Craford, and D. A. Steigerwald, "High dislocation densities in high efficiency GaN-based light-emitting diodes," *Appl. Phys. Lett.*, vol. 66, no. 10, pp. 1249–1251, Mar. 1995.
[2] D.-H. Kim *et al.*, "Enhanced light extraction from GaN-based light-emitting diodes with holographically generated two-dimensional photonic crystal patterns," *Appl. Phys. Lett.*, vol. 87, no. 20, Nov. 2005, Art. ID. 203508.
[3] L. Stafford, L. F. Voss, S. J. Pearton, H. T. Wang, and F. Ren, "Improved long-term thermal stability of InGaN/GaN multiple quantum well light-emitting diodes using TiB2-and Ir-based p-Ohmic contacts," *Appl. Phys. Lett.*, vol. 90, no. 24, Jun. 2007, Art. ID. 242103.
[4] W. S. Wong *et al.*, "Fabrication of thin-film InGaN light-emitting diode membranes by laser lift-off," *Appl. Phys. Lett.*, vol. 75, no. 10, pp. 1360–1362, Sep. 1999.
[5] C.-F. Chu *et al.*, "Study of GaN light-emitting diodes fabricated by laser lift-off technique," *J. Appl. Phys.*, vol. 95, no. 8, pp. 3916–3922, Apr. 2004.
[6] W.-Y. Lin *et al.*, "High-power GaN-mirror-Cu light-emitting diodes for vertical current injection using laser liftoff and electroplating techniques," *IEEE Photon. Technol. Lett.*, vol. 17, no. 9, pp. 1809–1811, Sep. 2005.
[7] T.-I. Kim *et al.*, "Injectable, cellular-scale optoelectronics with applications for wireless optogenetics," *Science*, vol. 340, no. 6129, pp. 211–216, Apr. 2013.
[8] R. H. Reuss *et al.*, "Macroelectronics: Perspectives on technology and applications," *Proc. IEEE*, vol. 93, no. 7, pp. 1239–1256, Jul. 2005.
[9] T.-I. Kim *et al.*, "High-efficiency, microscale GaN light-emitting diodes and their thermal properties on unusual substrates," *Small*, vol. 8, no. 11, pp. 1643–1649, Jun. 2012.
[10] W.-S. Choi, H. J. Park, S.-H. Park, and T. Jeong, "Flexible InGaN LEDs on a polyimide substrate fabricated using a simple direct-transfer method," *IEEE Photon. Technol. Lett.*, vol. 26, no. 21, pp. 2115–2117, Nov. 2014.
[11] C. Kisielowski *et al.*, "Strain-related phenomena in GaN thin films," *Phys. Rev. B, Condens. Matter*, vol. 54, no. 24, pp. 17 745–17 753, 1996.
[12] N. Khan and J. Li, "Effects of compressive strain on optical properties of InxGa1-xN/GaN quantum wells," *Appl. Phys. Lett.*, vol. 89, no. 15, Oct. 2006, Art. ID. 151916.
[13] B.-J. Pong *et al.*, "Abnormal blue shift of InGaN micro-size light emitting diodes," *Solid State Electron.*, vol. 50, no. 9/10, pp. 1588–1594, Sep./Oct. 2006.
[14] Z. Gong *et al.*, "Size-dependent light output, spectral shift, and self-heating of 400 nm InGaN light-emitting diodes," *J. Appl. Phys.*, vol. 107, no. 1, Jan. 2010, Art. ID. 013103.
[15] S. Y. Lee *et al.*, "Water-resistant flexible GaN LED on a liquid crystal polymer substrate for implantable biomedical applications," *Nano Energy*, vol. 1, no. 1, pp. 145–151, Jan. 2012.
[16] W. Zhou *et al.*, "Progress in 2D photonic crystal Fano resonance photonics," *Progr. Quantum Electron.*, vol. 38, no. 1, pp. 1–74, Jan. 2014.
[17] Y. Yang *et al.*, "Arrays of silicon micro/nanostructures formed in suspended configurations for deterministic assembly using flat and roller-type stamps," *Small*, vol. 7, no. 4, pp. 484–491, Feb. 2011.