Highly Efficient and Bright LEDs Overgrown on GaN Nanopillar Substrates

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***Abstract*—We presented a study of high-performance GaN-based light emitting diodes (LEDs) using a GaN nanopillars (NPs) structure grown on sapphire substrate by integrating RF-plasma molecular beam epitaxy (MBE) and metal–organic chemical vapor deposition (MOCVD). Nanoscale air voids were clearly observed at the interface between GaN NPs and the overgrown GaN layer by cross-sectional scanning electron microscopy. It can increase the light-extraction efficiency due to additional light scattering. The transmission electron microscopy images suggest the air voids between GaN NPs introduced during nanoscale epitaxial lateral overgrowth of GaN can suppress the threading dislocation density. Moreover, Raman spectrum demonstrated that the strain of the GaN layer grown on GaN NPs was effectively eliminated, resulting in the reduction of quantum-confined Stark effect in InGaN/GaN quantum wells. Consequently, the LEDs fabricated on the GaN NPs template exhibit smaller electroluminescent peak wavelength blue shift and great enhancement of the light output (70% at 20 mA) compared with the conventional LEDs.**

***Index Terms*—Light emitting diodes (LEDs), metal–organic chemical vapor deposition (MOCVD), molecular beam epitaxy (MBE), quantum-confined Stark effect (QCSE).**

I. INTRODUCTION

GAN-BASED optoelectronic devices can be used in a wide range of applications due to its wide band gap coverage (from ultraviolet to infrared), sustainability of high electrical field, and high temperature operation. In the last two decades, we saw a strong demand of GaN-based lasers or LEDs, which will eventually change our daily life. However, lack of a suitable, inexpensive substrate restrains the improvement of GaN-based devices. Even though many semiconductor companies produce and sell pure GaN substrates today, their prices are high and not very accessible to ordinary applications. Typically, GaN-based epitaxial layers were grown on sapphire substrate by heteroepitaxial technique, such as metal–organic chemical vapor deposition (MOCVD) [1], [2]. Due to the large lattice mismatch and thermal expansion coefficient misfit between GaN and sapphire, the subsequent-grown GaN epitaxial layers usually contained high threading dislocation densities (TD densities) (around $10^8 - 10^{10}$ cm^{-2}) [3]. To improve the crystalline quality of GaN-based epitaxial layers on sapphire substrate, various growth techniques have been proposed, such as epitaxial lateral overgrowth (ELO) [4], [5], cantilever epitaxy (CE) [6], defect selective passivation [7], microscale SiN$_x$ or SiO$_x$ patterned mask [8]–[10], anisotropically etched GaN-sapphire interface [11], plastic relaxation through buried AlGaN cracks [12], and patterned sapphire substrate (PSS) [13]–[15].

EVEN with these techniques, it is still difficult to reduce TD density to a level $\sim 10^7$ cm^{-2} unless certain complicated or expensive method such as double ELOG [16] or epitaxy on GaN substrate [17] is used. Recently, nanoscale epitaxial lateral overgrowth (NELOG) was found to be a promising method. During the NELOG process, coalescence overgrowth of nanostructures not only improves crystal quality [18], but also produces a scattering effect on the emitted photons, leading to higher light-extraction efficiency (LEE) [19]. The nanostructures were generally fabricated by top-down methods [20]–[22], such as etching process, in which the dry etching procedure normally generates defect states on the column surfaces, causing reduction of internal quantum efficiency (IQE). In this paper, we report a NELOG of high-quality GaN layer on bottom-up nanostructure [self-assembled GaN nanopillars (NPs)] grown by molecular beam epitaxy (MBE) [23]. Detailed analyses of the grown InGaN/GaN film will be demonstrated, and electrooptical properties of LEDs based on such GaN NPs template will also be discussed.

II. EXPERIMENTS

The epitaxial structure for GaN-based LED on sapphire with GaN NP was prepared as follows. First, the self-assembled GaN NP structure was grown on sapphire substrate by a RF-plasma MBE system (ULVAC MBE), and the related processes

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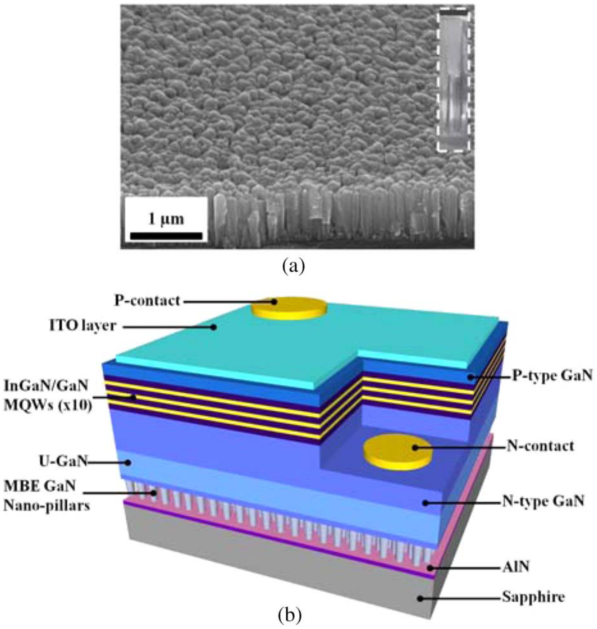


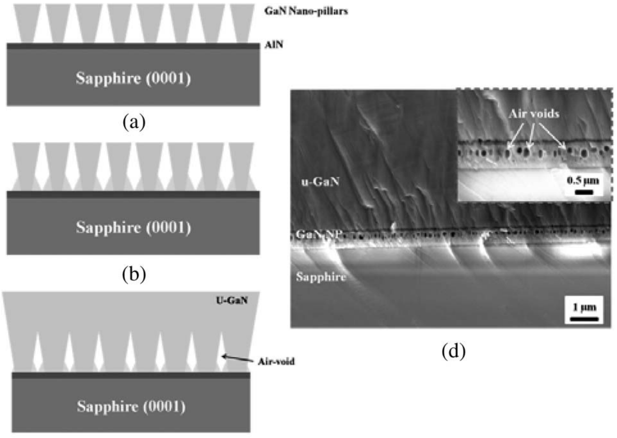
(a)

(b)

Fig. 1. (a) Cross-sectional SEM image of GaN NPs template. The inset shows the funnel-like GaN NP. (b) Schematic of GaN-based LED structures on GaN NPs template.

have been reported in our previous study [24]. Fig. 1(a) shows scanning electron microscope (SEM) image of the grown GaN NPs. It can clearly be seen that the GaN NP is in funnel-like form shown on the inset Fig. 1, which might be beneficial for the following regrowth of GaN-based LED structure. In addition, the density, the diameter, and the height are estimated to be around 1.15×10^{10} cm^{-2}, 50 nm and 0.8 μm, respectively. Next, we deposited a GaN-based LED structure on this NP template by a low-pressure MOCVD (Veeco D75) system, denoted as NP-LEDs. In the mean time, the same GaN-based LED structure was also grown on sapphire without GaN NP for comparison, denoted as conventional LEDs (i.e., C-LEDs). During the growth, trimethylgallium (TMGa), trimethylindium (TMIn), and ammonia (NH_3) were used as gallium, indium, and nitrogen sources, respectively. Silane (SiH_4) and biscyclopentadienyl magnesium (CP_2 Mg) were used as the *n*-dopant and *p*-dopant source. The epitaxial structure of the GaN-based LED overgrowth on NP is depicted in Fig. 1(b), consisting of 30-nm GaN nucleation layer (GaN NL), 1-μm *un*-doped GaN (*u*-GaN), 3-μm *n*-doped GaN (*n*-GaN), 10-pairs InGaN/GaN multiquantum wells (MQWs), and 0.2-μm *p*-doped GaN (*p*-GaN) cap layer.

The surface morphology of the overgrown GaN NPs sample was measured by atomic force microscopy (AFM). The room temperature Raman scattering was used to analyze the residuals strain of GaN epitaxial layers and GaN-based LEDs. The distribution and behaviors of TDs in epitaxial layers were then resolved by transmission electron microscopy (TEM). The electrooptical properties of GaN-based LEDs were studied by electroluminescence (EL) measurements. Finally, after normal clean-room processes are finished, we compared the current–voltage (*I*–*V*) and optical output (*L*–*I*) of two types of LEDs on a conventional probe station and an integration-sphere setup.



(a)

(b)

(c)

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Fig. 2. (a)–(c) Procedure of the air-voids formation between a GaN NPs and *u*-GaN epitaxial layer; (d) Cross-sectional SEM image. The inset shows air voids.

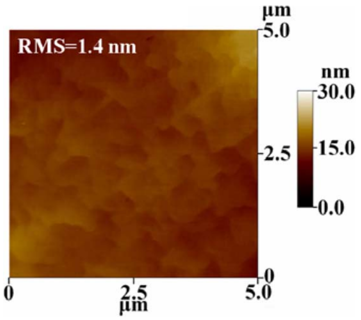


Fig. 3. Surface morphology of overgrown GaN NPs template scanned by AFM.

III. RESULTS AND DISCUSSION

It is first of our great interest to find out what happened to these NPs after regrowth. Fig. 2 shows the proposed steps of the air-voids formation during the entire material growth procedure. First, funnel-like shaped GaN NPs were formed on a sapphire substrate by MBE at substrate temperature of 740 °C shown in Fig. 2(a). As the NP grows upward, there is also lateral growth on the sidewall of individual pillar. Such lateral growth eventually narrows the gap between columns and forms holes with 0.2–0.25 μm in size, which is shown in Fig. 2(b). Next, we transfer the template to a MOCVD system to finish the growth. The regrowth temperature of GaN film is about 1050 °C. Under this high temperature, recrystallization of GaN is very possible and final coalescence *u*-GaN NPs template was performed and air voids were encapsulated, as shown in Fig. 2(c) and (d). From the SEM pictures in Fig. 2(d), we can estimate the average diameter of these air voids is about 100 nm. These embedded air voids shall be able to increase the LEE due to extra light scattering from these air bubbles [25].

The quality of the film can first be evaluated by its surface roughness. After the *u*-GaN layer was deposited, without growth of remaining LED layers, the surface morphology was measured by AFM, as shown in Fig. 3. The root mean square (rms) value of the surface roughness is about 1.4 nm, indicating high surface

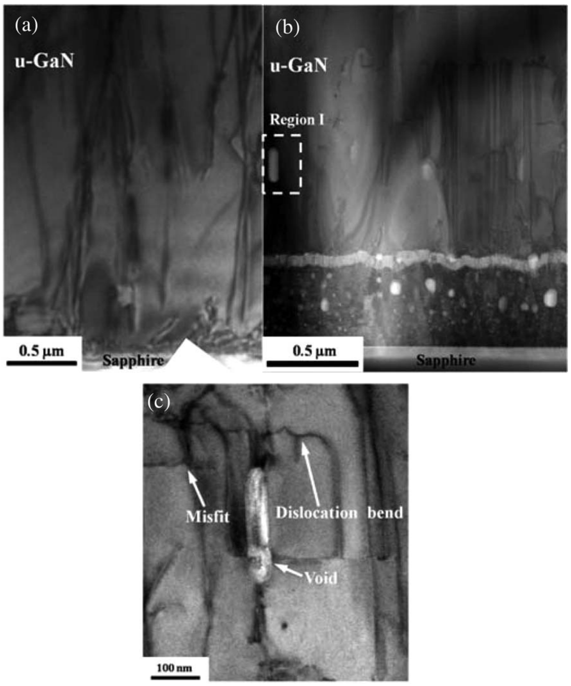


Fig. 4. TEM image of (a) C-LEDs, (b) NP-LEDs, and (c) high-resolution TEM image of region I in (b). The diffraction condition is $g = 0002$.

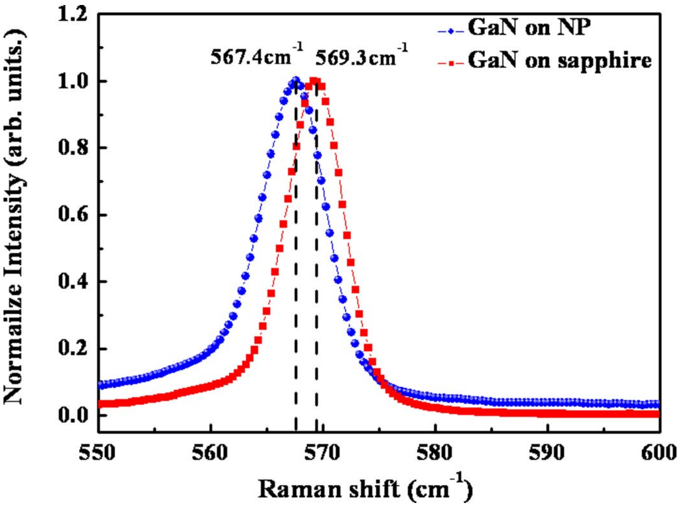


Fig. 5. Raman spectrum for GaN epilayer overgrown on GaN NPs template and sapphire.

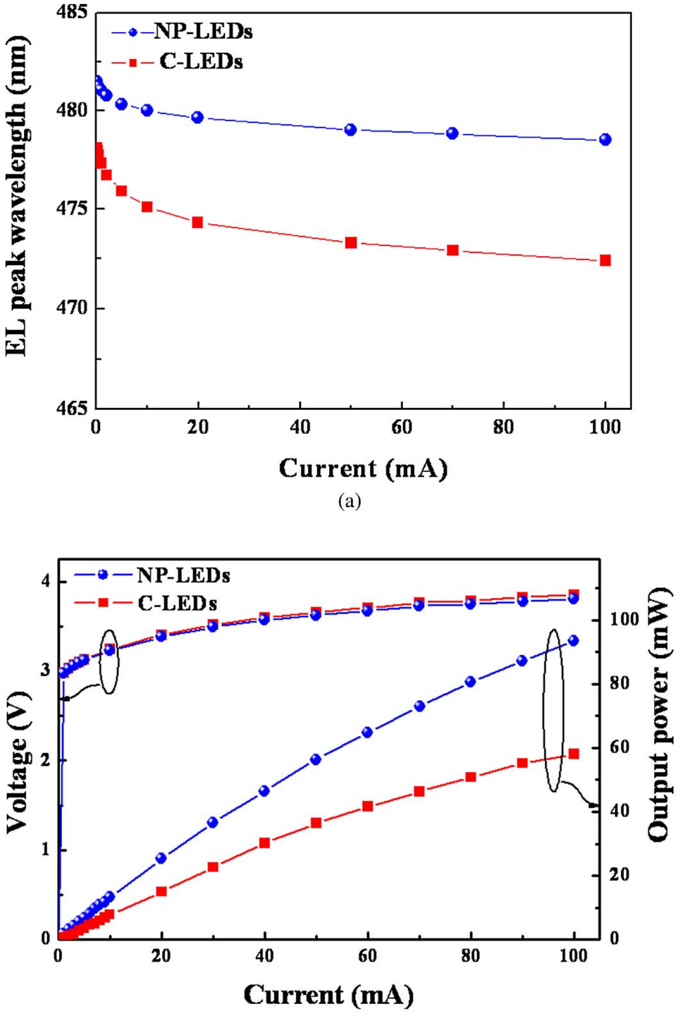
quality and excellent coalescence overgrown on GaN NPs template. To analyze the detailed epitaxial layer quality, we used TEM to compare the cross section between two types of devices (NP-LEDs and C-LEDs). As we can see from Fig. 4(a), in the case of the GaN epitaxial layer grown on sapphire without GaN NPs, numbers of TD propagate vertically from the interface of GaN and sapphire, all the way to the top device layers. As a result, the TDs density in conventional GaN layer can be as high as 10^9 cm^{-2}. Whereas, for the GaN epitaxial layer grown on sapphire with GaN NP [(see Fig. 4(b)], it can be clearly found that the crystallography is drastically different from that of conventional ones. Fewer TDs are observable within the range in view. The dislocation density on the top of n-GaN, MQWs is calculated to be around 7×10^7 cm^{-2}. The reduction of TDs density can be attributed to the misfit (mainly perpendicular to the c-axis) and dislocation bending occurred just above the voids, as shown in the inset of Fig. 4(b). Such behaviors are similar to those occurred in the NELOG method on a SiO$_2$ nanorod-array-patterned sapphire substrate [26].

In addition to material defect density, another important feature to watch is the internal stress of the epitaxial film since the nanosized holes of template can potentially alleviate the built-in stress due to lattice mismatch. To analyze the residual strain in the GaN films, Raman backscattering measurements were performed at room temperature. Fig. 5 shows the Raman spectrum for GaN epitaxial layer grown on sapphire with and without GaN NPs. The Raman shift peaks of E$_2$ (high) mode for GaN epitaxial layer grown on sapphire with and without GaN NPs are located at around 567.4 and 569.3 cm^{-1}, respectively. The in-plane compressive stress σ for GaN epitaxial layer is estimated to decrease from 1.24 to 0.4 GPa with presence of GaN NP templates, by using the following equation [27]:

$$\Delta\omega = \omega_{E_2} - \omega_0 = C\sigma \tag{1}$$

where $\Delta\omega$ is the Raman shift peak difference between the strained GaN epitaxial layer ω_{E2} and the unstrained GaN epitaxial layer ω_0 (566.5 cm^{-1}), and C is the biaxial strain coefficient, which is 2.25 cm^{-1}/GPa. Since the film on NP template bears less strain, consequently we can expect that the GaN-based LED grown on such template have weaker quantum-confined Stark effect (QCSE) [28]. LED devices with a chip size of 350 × 350 mm^2 were then fabricated from the completed epitaxial structures grown on sapphire with and without GaN NPs. Fig. 6(a) shows EL emission peak wavelength as a function of injection current for NP-LEDs and C-LEDs. The emission peak wavelength of NP-LED is slightly red shifted (about 3.4 nm) from that of C-LED, and this is reasonable since lateral strain relaxation favors higher indium incorporation [29]–[31]. More importantly, as we increase the injection current, the emission peak wavelength of NP-LEDs exhibits smaller blue shift (around 2.9 nm) compared with that of C-LEDs (around 5.6 nm). This result indicates that the QCSE does become weaker due to the strain relaxation in epitaxial layer overgrown on GaN NPs template, as we expected. Fig. 6(b) displays the typical power—current–voltage (*L*–*I*–*V*) characteristics of NP-LEDs and C-LEDs. With an injection current of 20 mA, the forward voltages are 3.38 and 3.40 V, and the output powers are 25.3 and 14.8 mW, for NP-LEDs and C-LEDs, respectively. The light enhancement of *L*–*I*–*V* characteristics can be attributed to the following factors: First, the TD density reduction of epitaxial layers. This reduction leads to much fewer nonradiative recombination events in the NP devices and increases the photon-generation efficiency. Second, more lights can be extracted from the LED because of the light-scattering effect from the embedded nanoscale air voids.

In order to confirm the efficiency improvement of our NP-LED, the PL IQE measurement was performed. A general approach to evaluate the IQE of LEDs is to compare the PL-integrated intensity between low and room temperatures [32].



(a)

(b)

Fig. 6. (a) EL peak wavelength as a function of injection current of two fabricated LEDs. (b) *L–I–V* characteristics of the two fabricated LEDs.

Fig. 7 shows the measured IQE as a function of excitation power at 15 and 300 K for NP-LEDs and C-LEDs. The efficiency is defined as the collected photon numbers divided by the injected photon numbers and normalized to the maximum efficiency at low temperature [33]. At 20 mW of excitation power, it can be found that the IQE increase from 58% (C-LEDs) to 72% (NP-LEDs), which corresponds to 1.24 times enhancement of efficiency. At this excitation level, we could calculate the corresponding generated carrier density to be 2×10^{17} cm^{-3}, approximately same level of 20 mA at room temperature in our device. Thus, part of the efficiency improvement of GaN-NP-based LED can be linked directly to the improvement of IQE due to better crystal quality.

On the other hand, we still need to quantify how much improvement of *L–I–V* is coming from the better light-extraction scheme due to air voids. A 2-D finite difference time domain (FDTD) simulation was applied to calculate the LEE of the LEDs using the FullWAVE program [34]. The calculated electric-field distribution with air-void period of 0.25 μm is shown in Fig. 8(a), where an array of air-filled rectangular holes

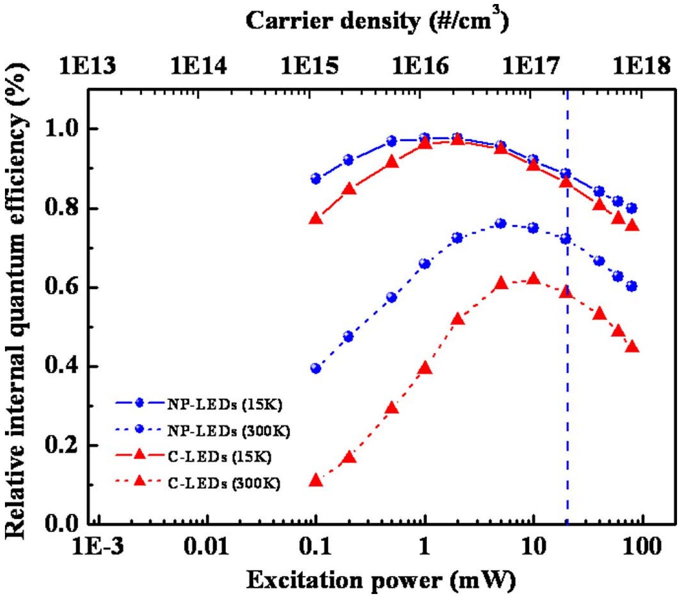
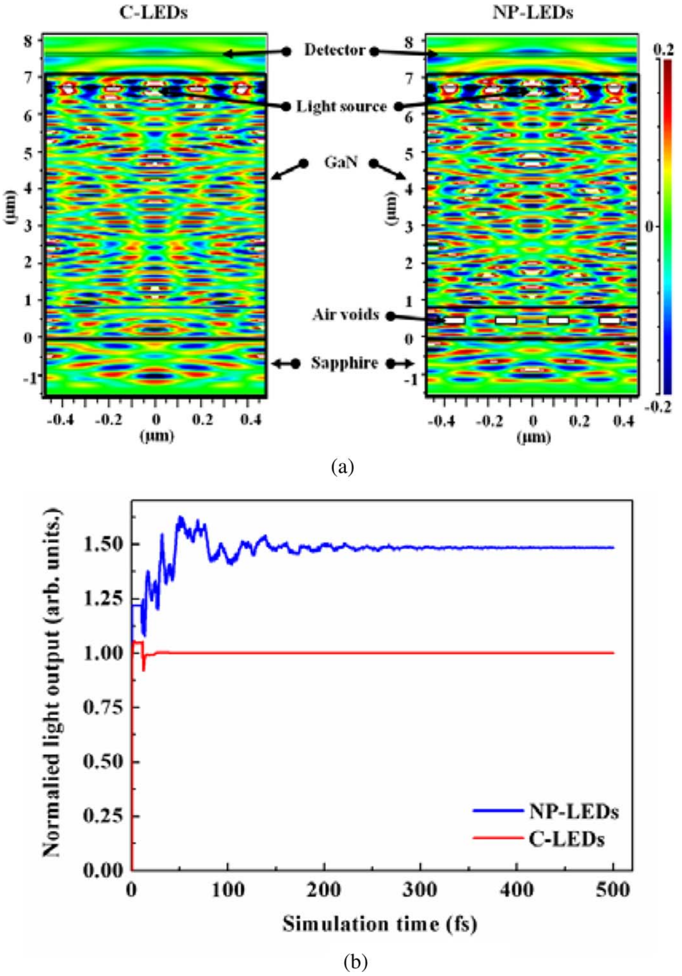


Fig. 7. Relative IQE as a function of excitation power for C-LEDs and NP-LEDs.



(a)

(b)

Fig. 8. (a) 2-D FDTD of the calculated electric-field distribution of NP-LEDs and C-LEDs. (b) Normalized light output power as functions of the simulation time for C-LEDs and NP-LEDs.

represents air voids in our devices. The size of each rectangular hole is 0.2 μm × 0.1 μm. We set single dipole illumination sources placed at 0.5 μm below the top of surface structures and the detector around the simulated device [35]. As it can be seen in the figure, the electric-field intensity of NP-LEDs is higher than C-LEDs at the monitor. It indicates that the photons emitted from the MQWs escape out into the air easier in NP-LEDs than in C-LEDs. The corresponding normalized light output as functions of the simulation time are calculated and plotted in Fig. 8(b), and the enhancement of extra light scattering is defined as the ratio of steady-state light output of NP-LEDs to that of C-LEDs. From the simulated results, light output of NP-LEDs is around 1.48 times higher than that of the C-LEDs. Combining with previous PL IQE measurement, we can see a total enhancement of 82% (48% from LEE and 24% from IQE) when we compare the result to conventional LED structure. The actual increase in power output of LED, which is 70%, is lower than prediction. This is possibly due to randomness of the air-void formation, which makes our FDTD analysis overestimating the light-scattering effect.

IV. CONCLUSION

In summary, high-quality GaN-based LED structure was successfully fabricated on GaN NPs template by using MOCVD and MBE. It was found that the residual stress was reduced from 1.24 to 0.4 GPa in GaN epitaxial layer by inserting the GaN NPs. Consequently, the NP-LEDs exhibit smaller EL peak wavelength blue shift and great enhancement of the light output 70% at 20 mA compared to the C-LEDs. From SEM measurement, a layer of air voids was formed at the interface of the GaN NP and subsequent GaN layer. In addition, TEM revealed obvious dislocations-bending behavior above the voids, resulting in low dislocation density in 10^7 cm^{-2} range. So our nanostructure shows two folds of the improvement: one in IQE enhancement from better crystal quality and the other in light extraction due to air-void layer. With low-temperature PL measurement and 2-D FDTD simulation, we can estimate the enhancement brought by each factor (IQE and light extraction) should be 24% and 48%, respectively, and this is close to what we observed in *L–I–V* measurement (70% at 20 mA).

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