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Bridging “green gap” of LEDs: Giant light output enhancement and directional control of LEDs via embedded nano-void photonic crystals†

Yu-Lin Tsai,^{a,b} Che-Yu Liu,^a Chirenjeevi Krishnan,^c Da-Wei Lin,^a You-Chen Chu,^a Tzu-Pei Chen,^a Tien-Lin Shen,^a Tsung-Sheng Kao,^a Martin D. B. Charlton,^c Peichen Yu,^a Chien-Chung Lin,^{*d} Hao-Chung Kuo,^{*a} and Jr-Hau He,^{*b}

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Green LEDs do not show the same level of performance as their blue and red cousins, greatly hindering the solid-state lighting development, which is so-called “green gap”. In this work, nano-void photonic crystals (NVPCs) were fabricated to embed within the GaN/InGaN green LEDs by using epitaxial lateral overgrowth (ELO) and nano-sphere lithography techniques. The NVPCs act as an efficient scattering back-reflector to outcouple the guided and downward photons, which not only boosting light extraction efficiency of LEDs with an enhancement of 78% but also collimating the view angle of LEDs from 131.5° to 114.0°. This could be because the highly scattering nature of NVPCs which reduce the interference giving rise to Fabry-Perot resonance. Moreover, due to the threading dislocation suppression and strain relief by the NVPCs, the internal quantum efficiency was increased by 25% and droop behavior was reduced from 37.4% to 25.9%. The enhancement of light output power can be achieved as high as 151% at a driving current of 350 mA. Giant light output enhancement and directional control via NVPCs points the way towards a promising avenue of solid-state lighting.

1. Introduction

Light-emitting diodes (LEDs) have drawn much attention due to the promising huge market for solid-state lighting.^{1,2} However, green LEDs occupied on a part of the spectrum critical for color perception are much less efficient than red and blue LEDs due to low light extraction efficiency (LEE) and internal quantum efficiency (IQE).^{3,4} This efficiency drop of the green LEDs, as known as the “green gap”, limits the development of LEDs.^{5,6} Moreover, many advanced LED applications such as vehicle head lamps and LED projectors are obstructed by a broad emission pattern of conventional LEDs. Therefore, it is important to develop high-efficiency green LEDs with directional radiation patterns for next-generation lighting sources.

Low LEE and broad emission pattern can be attributed to the downward scattering and multiple internal reflection within LED

chip caused by high refractive index (*n*) contrast between air (n=1) and GaN (n=2.43).^{7–9} Various methods have been utilized to improve the LEE, such as the use of surface texturing, dielectric sub-wavelength structure and graded-refractive-index coatings.^{10–16} However, neither the extraction of downward photons nor broad radiation patterns can be solved by these methods. After undergoing multiple internal reflection, reabsorption probability of photons by active layer would be also increased and thus result in reduction of IQE.¹⁷ Moreover, the threading dislocations and in-plane strain in active layer can degrade the IQE.¹⁸ To develop high-efficiency directional LEDs, it is imperative to find an effective way to achieve the concurrent improvement in IQE, LEE, and radiation patterns.

Photonic crystals (PCs) have been demonstrated as a promising approach for enhancing the light extraction and controlling the radiation pattern of LEDs.^{19,20} However, the use of omnidirectional PC bandgaps to extract all of the guided photons is difficult due to the relatively low refractive index of GaN and the thickness of typical epilayers.²¹ Therefore, the utilization of scattering or diffracting properties of PCs to outcouple the guided photons has been demonstrated as an alternative approach to enhance LEE.^{22,23} Several methods have been utilized to fabricate the PCs, such as nano-imprint lithography,²⁴ electron beam lithography²⁵ and laser interference lithography.²⁶ However, these methods are still limited by the high demand of facilities or the difficulties in large-area fabrication. Self-assembly nano-sphere lithography is considered as an alternative way to fabricate the PCs due to the low deman-

^a Department of Photonics & Institute of Electro-Optical Engineering, National Chiao Tung University, Hsinchu 300, Taiwan, ROC
E-mail: hckuo@faculty.nctu.edu.tw

^b Computer, Electrical and Mathematical Sciences and Engineering (CEMSE) Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia
E-mail: jrhou.he@kaust.edu.sa

^c School of Electronics and Computer Science, University of Southampton, Southampton SO17 1BJ, United Kingdom

^d Institute of Photonic System, National Chiao Tung University, Tainan 711, Taiwan, ROC
E-mail: chienchunglin@faculty.nctu.edu.tw

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of facilities, low cost and the capability to reach large-area fabrication.²⁷

In this study, embedded nano-void photonic crystals (NVPCs) via the epitaxial lateral overgrowth (ELO) and nano-sphere lithography techniques act as an efficient scattering back-reflector in a GaN/InGaN green LEDs to outcouple the guided and downward photons. As a result, the LEE was boosted with an enhancement of 78% and the view angle of LEDs was collimated from 131.5° to 114.0° . Moreover, the IQE was increased by 25% and efficiency droop was reduced from 37.4% to 25.9% due to the suppression of threading dislocations (TDs) and residual strain by employing NVPCs^{28,29}. At a driving current of 350 mA, the light output power (LOP) enhancement was increased by 151%, as compared to the reference LED. The enhanced light extraction was also verified by the simulation based on finite difference time domain (FDTD) method. The presented concept paves the way for filling the “green gap” for realizing high-efficiency solid-state lighting.

2. Experimental

The process flow of NVPCs in u-GaN is schematically described in Fig. 1a. First, close-packed polystyrene (PS) nano-spheres at a diameter of 550 nm were homogeneously coated on a 4- μm -thick u-GaN layer on a c-plane sapphire substrate (also shown in Fig. S1a in the ESI†). The size of the coated PS nano-spheres was trimmed to 500 nm by the oxygen plasma treatment. Then, nickel was evaporated onto the wafer and filled in the gaps between the nano-spheres. The nano-spheres were then removed afterwards through acetone immersion, creating a nickel hard

mask with honeycomb-like structures (also shown in Fig. S1b in the ESI†). By using inductively coupled plasma reactive ion etching (ICP-RIE) technique, nano-crown arrays were generated. At the end, u-GaN was deposited onto the nano-crown arrays in a metal organic chemical vapor deposition (MOCVD) system, forming a monolayer of NVPCs embedded in u-GaN. It should be noted that, most of the reported method to fabricate the embedded air-void photonic crystal are conducted through the selective epitaxial lateral overgrowth (ELO) using passivation masks made of SiO_2 or metal to keep the air-void during GaN overgrowth^{28,30,31}. In this work, we propose an alternative way to generate embedded homogeneous nano-void PC LEDs without any passivation mask. This passivation-free fabrication process is based on different growth rate of different facet. Such a fabrication process was repeated to form triple-layer NVPCs. Note that the uniform u-GaN growth on four-layer NVPCs becomes difficult due to its highly porous nature.

After the growth of triple-layer NVPCs, the GaN/InGaN multiple quantum well (MQWs) green LEDs were epitaxially grown on top of NVPCs. From the bottom to the top, the LED structures comprise a 20-period of $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}/\text{GaN}$ (3 nm/3 nm) superlattice and a 5-period of $\text{In}_{0.25}\text{Ga}_{0.75}\text{N}/\text{GaN}$ (3 nm/15 nm) undoped MQWs sandwiched by a 2- μm -thick Si-doped n-GaN layer and a 200-nm-thick Mg-doped p-GaN layer. A 110-nm-thick indium-tin-oxide (ITO) conducting layer was deposited onto p-GaN, then the LED was defined into a $1 \times 1 \text{ mm}^2$ mesa by an ICP-RIE system. Finally, Cr/Ti/Au (50/50/190 nm) was deposited as electrodes. Fig. 1b and 1c show the schematic diagram and the finished quarter-sized wafer of the triple-layer NVPC LEDs (PCLEDs), respectively.

Fig. 1 (a) Process flow of embedded triple-layer NVPCs in u-GaN. (i) Distribute polystyrene (PS) nanospheres homogeneously on a u-GaN layer; (ii) Fill nickel in the gaps between the spheres via an e-beam evaporator and then remove the PS spheres to form a nickel hard mask; (iii) Generate nano-crown nanostructures through the ICP-RIE process; (iv) Deposit GaN onto the patterned u-GaN layer, forming embedded NVPCs in a u-GaN. (b) Schematic of GaN/InGaN MQW green triple-layer NVPC LEDs. (c) The picture of the finished quarter-sized wafer of the triple-layer NVPC LED.

3. Results and discussion

Fig. 2a shows the SEM image of the nano-crown arrays of u-GaN after ICP-RIE process. The height and the diameter of nano-crown are around 1 μm and 500 nm, respectively. For forming a monolayer of NVPCs embedded in u-GaN, u-GaN was deposited on nano-crown arrays by using MOCVD. The approximate height and width of single nano-voids are 500 nm and 300 nm, respectively, and the distance between each NVPC layers is ~ 400 nm. Fig. 2b indicates the SEM image of a finished sample with triple-layer NVPCs after employing the above-mentioned fabrication processes for 3 times. In addition, scanning transmission electron microscopy (STEM) was employed for nanostructure characterization. To investigate the distribution of TDs, the STEM image of u-GaN with triple-layer NVPCs is shown in Fig. 2c. One should note that one of challenges in the development of highly efficient GaN-based green LEDs is how to increase IQE. The lattice mismatch between GaN and sapphire, causing a large number of TDs from nucleation layer to MQWs serving as non-radiative recombination centers and leakage current pathway, leading to the decrease in the IQE.^{32,33} As shown in Fig. 2c, TDs generated

from the nucleation layer stretch vertically into the u-GaN epilayer. The TDs are suppressed by the introduction of triple-layer NVPCs via two mechanisms: 1) the TDs are bended and thus stopped due to the ELO mode of NVPCs; and 2) the TDs are blocked by the NVPCs. Therefore, by enhancing ELO mode of NVPCs, the area without TDs in GaN can be greatly enlarged. The insets in Fig. 2c show the high-magnification cross-sectional image for STEM analysis. The energy-dispersive X-ray spectroscopy (EDX) mapping was employed to study the elemental distribution of GaN with triple-layer NVPCs. One can see that as shown in Fig. 2d–2f, both nitrogen (green) and gallium (purple) elements are distributed uniformly in GaN excepts for the nano-void region, indicating the NVPCs have been successfully fabricated while maintaining the uniformity of the GaN growth. Note that to keep the comparison valid, we deliberately kept the total thickness of the u-GaN layer constant among all samples. With the same thickness of u-GaN, the overall stress in the epitaxial structure and the optical path of the photons in the devices are about the same level and the impact of NVPCs can be clearly observed.

Fig. 2 (a) Cross-section and 45°-tilted SEM image of the nano-crown arrays. (b) SEM image with the false-colored of embedded triple-layer NVPCs in u-GaN layer. (c) STEM image of u-GaN epilayer with triple-layer NVPCs. (d) STEM image of a nano-void and (e)-(f) the corresponding elemental mapping of N and Ga elements.

The LOP-current-voltage (L-I-V) characteristics of reference LED and PCLED are presented in Fig. 3a. The forward voltage under 20 mA injection current of each device shows the same value of 2.95 volts and the series resistance (R_s) of the PCLED is slightly lower than that of the reference LED, which can be attributed to the suppression of TDs in MQWs. The lower R_s represents the better current injection, leading to the higher efficiency of LEDs. With the introduction of triple-layer NVPCs, the LOP is enhanced by 151% at an injection current of 350 mA, which can be attributed to the strong guided photon extraction and the improved IQE. The mechanisms of enhanced

LOP will be discussed with experimental and simulation results in the following paragraphs. The L-I-V characteristics of LEDs with different numbers of NVPC layers are also shown in Fig. S2 in the ESI†. The insets in Fig. 3a are the top-view optical images of the reference LED and PCLED operating at 25 mA. The emission wavelengths are 519 nm and 528 nm for reference LED and PCLED, respectively. The redshift in emission wavelength is observed in PCLED and is attributed to the reduced strain brought by NVPCs, increasing the indium incorporation when the MQWs are grown³⁴, which can be estimated by Raman spectroscopy through the shift of the E₂

high phonon peaks. As comparing to the E₂ high phonon peak of strain-free GaN (566.5 cm⁻¹), 3.7 and 2.75 cm⁻¹ of the Raman peak shift correspond to 1.45 and 1.07 GPa of residual stress for the reference LED and PCLED, respectively (see Fig. S3 in the ESI†).

External quantum efficiency (EQE) is an important characteristic for revealing the LED performance. The EQE of the LEDs with and without NVPCs at different injection currents are presented in Fig. 3b. The EQE of reference LED and PCLED at an injected current of 350 mA are 5.56% and 14.47%, respectively, demonstrating a 160.2% enhancement by NVPCs. Efficiency droop, another important parameter for the solid-state lighting³⁵ can be defined as

$$\text{Efficiency droop} = \frac{EQE_{\text{peak}} - EQE_{350 \text{ mA}}}{EQE_{\text{peak}}} \quad (1)$$

where the EQE_{peak} is the highest EQE achieved and EQE_{350 mA} is the EQE at an injection current of 350 mA. From the normalized EQE shown in Fig. 3b, the efficiency droop is reduced from 37.4% of reference LED to 25.9% of the PCLED, which is attributed to the suppressed electron overflow related to the reduction of strain-induced polarization field in MQWs^{36,37}. The improved droop behavior demonstrates that the PCLED can sustain high EQE at a high current level. The EQE and normalized EQE of LEDs with different numbers of NVPC layers are also shown in Fig. S4a-S4b in the ESI†. As we mentioned earlier, the enhanced performance of PCLED can be attributed to several mechanisms. Therefore, it is very important to de-couple all of them for revealing the impact related to NVPCs. To gain insight into the light extraction brought by NVPCs, the reflectivity was measured without MQWs. All the samples were measured with 80-nm-thick SiNx as anti-reflective layers which can minimize the Fresnel reflection at GaN/air interface. The reflectance spectra of u-GaN with and without NVPCs are presented in Fig. 3c. Comparing to the reference sample, the reflection can be enhanced by NVPCs and thus the photon harvesting on the front side becomes more effective. At the wavelength of 520 nm, approximately 80% reflection enhancement is observed, for example. Due to the large refractive index difference between air voids (n=1) and GaN (n=2.43), the NVPCs act as an efficient scattering back-reflector to outcouple guided photons and downward photons. However, the reflectivity is lower than our expectation probably because of incoherent reflection³⁸. Detailed description and calculation of NVPC layers-dependent incoherent reflection in u-GaN are shown in Fig. S5a-S5c in the ESI†. By comparing between calculated and experimental results, incoherent reflection caused by NVPCs does play a role in light extraction efficiency.

To qualify the LEE of PCLED, IQE was conducted. The relationship between IQE and LEE is as shown in formula (2).

$$\eta_{\text{LEE}} = \eta_{\text{EQE}} / \eta_{\text{IQE}} \quad (2)$$

The PL quantum efficiency is used to estimate IQE and can be calculated by formula (3).³⁹

$$\eta_{\text{PL}} = C \frac{I_{\text{Col}} / E_{\text{Col}}}{I_{\text{Inj}} / E_{\text{Inj}}} \quad \text{View Article Online at: DOI: 10.1039/C5NR05555E}$$

where I_{Col} and I_{Inj} are collected light power and injected laser power, respectively. E_{Col} and E_{Inj} are collected photon energy and injected photon energy, respectively. C is a constant affected by carrier injection and light extraction. By normalizing the PL efficiencies obtained at low temperature (LT, 15K in our case) and room temperature (RT, 300K in our case), the relative PL efficiency can be obtained. Then, the IQE can be acquired by assuming that the relative PL efficiency at LT is 100% regardless of excitation carrier density due to the suppression of non-radiative recombination. Under this assumption, the IQE at room temperature can be acquired.

Fig. 3d shows the IQE of the reference LED and PCLED at the temperature of 300 K (RT) and 15 K (LT) using a He-Cu laser (λ = 325 nm) with an excitation laser power variable from 0.5 mW to 100 mW. The IQE of LED with different numbers of NVPC layers are also shown in Fig. S6 in the ESI†. As shown in Fig. 3d, the IQE has high degree of relevance with injected carrier density. Under low excitation power, the IQE of both reference LED and PCLED rises with PL excitation power, due to the increasing level of quenching to Quantum Confinement Stark Effect (QCSE)⁴⁰ and suppressed nonradiative recombination⁴¹ by carriers, and then subsequently the IQE of both devices starts to drop as the PL excitation exceeds to certain point, due to the Auger recombination⁴² and carrier overflow⁴⁰. The IQE at 300K with the excitation power of 10 mW are 28.61% and 35.74% for reference LED and PCLED respectively. The improvement of IQE is attributed to the suppressed TDs and residual stress in epilayers. From formula (2), an unencapsulated LEE of 54.56% in PCLED can be achieved, while the unencapsulated LEE of reference LED is 30.65%. The high LEE of PCLED can be attributed to the remarkable extraction of guided and downward photons by triple-layer NVPCs. To analyze the light output enhancement related to NVPCs, reflectivity enhancement (R), IQE enhancement at P_{incident}=10 mW (IQE@10mW), LOP enhancement at I_{injection}=40 mA (LOP@40mA) and calculated LOP enhancement (LOP_{cal.}) are shown together for comparison, as shown in Fig. 3e. The calculated LOP enhancement can be acquired through formula (4). The definition of enhancement and enhancement ratio are as shown in formula (5) and (6), respectively.

$$LOP_{\text{cal. enhancement ratio}} = R_{\text{enhancement ratio}} \times IQE_{\text{enhancement ratio}} \quad (4)$$

$$\text{Enhancement ratio} = \frac{\text{Value}_{\text{PCLED}}}{\text{Value}_{\text{Reference}}} \quad (5)$$

$$\text{Enhancement} = \frac{\text{Value}_{\text{PCLED}} - \text{Value}_{\text{Reference}}}{\text{Value}_{\text{Reference}}} \times 100\% \quad (6)$$

From the previous studies, the incident laser power can be transferred in to carrier density^{28,43}. Therefore, we choose the characteristic under electrical injection with 40 mA and excitation power of 10 mW is due to the approximately same injection carrier density. Obviously, the reflectivity enhancement is greatly larger than IQE enhancement which means the most of LOP enhancement is brought by efficient

guided photon extraction. As shown in Fig. 3e, the enhancement of calculated LOP is 125.69% shows a good agreement with measured LOP (122.51%), indicating the accuracy of our assessment on the model. The relationship between the enhancement of IQE, measured LOP, calculated LOP and reflectivity as the function of the NVPC layers are also shown in Fig. S7 in the ESI†. More experiments and simulations for optimizing the NVPCs for maximizing LOP are under investigation and will be published elsewhere.

To further unveil the light extraction behavior of NVPCs, a simulation based on FDTD was applied. The optical structures of the triple-layer NVPCs and LEDs for simulation model are determined based on the practical structures shown in experimental section. The computational domains are $1.8\ \mu\text{m}$ in the x-direction and $10\ \mu\text{m}$ in the z-direction. The wavelength of point dipole excitation source is set to be at 520 nm. The electric-field distribution inside the PCLED and reference LED are revealed in Fig. 4a. In the region of triple-layer NVPCs, the strong field intensity can be observed between nano-voids which is attributed to constructive wave interferences, indicating the highly scattering nature of triple-layer NVPCs. Due to large refractive index contrast between GaN ($n=2.43$) and nano-void ($n=1$) and additional interaction between NVPC layers, the guided photons can be deflected into various directions and thus much easier to hit the escape cone and leave the semiconductor, resulting in stronger electric field outside the LEDs. As shown in Fig. 4b, the steady state light output for the PCLED is 86% greater than that for the reference LED, which agrees well with the reflectivity enhancement shown in Fig. 3c. The effects of size and density of the nano-voids on light extraction enhancement based on FDTD simulation are also available in Fig. S7 in the ESI†. In addition to the giant LOP enhancement, the highly scattering nature of NVPCs is expected to modify the emission pattern of LEDs. Fig. 4c shows the measurement setup for angular far-field emission pattern and mapping in the angular range of $0\sim 85^\circ$, with 0° as the surface normal to the LEDs. Fig. 4d shows the normalized electroluminescent angular far-field emission pattern of reference LED and PCLED at 25 mA injection current, the view angle of LEDs is collimated from 131.5° of reference LED to 114.0° of PCLED, indicating the light control capability of NVPCs. The larger view angle can be attributed to side emission⁴⁴. For reference LED, a large number of photons emitted from the MQWs, instead of being extracted directly from the top surface, suffer from multiple internal reflection within the LEDs. After suffering multiple internal reflection, the photons were extracted towards the periphery of LEDs and emitted laterally, resulting in a larger view angle. In contrast, the NVPCs couple most of photons emitted from the MQWs to

the escape cone and then reduce optical path before guided photons are extracted. Consequently, the photon extraction along normal direction is enhanced, resulting in small emission angle in PCLED. The normalized angular far-field emission patterns of LEDs with different number of NVPC layers are also shown in Fig. S8 in the ESI†.

The LEE enhancement of PCLED could be due to two mechanisms: 1) Increased scattering at the NVPC layer. 2) The NVPC layer provides an intermediate reflective boundary, defining the lower boundary of the FP cavity. To further clarify the light extraction enhancement mechanisms of PCLED, the normalized electroluminescent angular far-field emission mappings of LEDs were conducted. Fig. 4e shows the normalized electroluminescent angular far-field emission mappings of reference LED and PCLED at 25 mA injection current. The reference LED shows a number of additional fringes, which can be attributed to the Fabry-Perot (FP) resonance between the top of sapphire substrate and the top of GaN³¹, indicating the photons are guided within reference LED. In contrast, the FP resonance-induced multiple-fringe pattern of PCLED is deteriorated. The normalized electroluminescent angular far-field emission mappings of LEDs with different number of NVPC layers are also conducted and shown in Fig. S9 in the ESI†. The normalized angular far field emission mappings for LEDs with single-layer NVPC exhibit similar angular and spectral FP characteristics to the reference device (see Fig. S9 in the ESI†), although intensity (modulation depth) of the FP fringes becomes slightly reduced. As the number of NVPC layer increase, FP fringes become much reduced in intensity however the number and positions remain pretty much the same. Finally for PCLED with triple NVPC layer, the FP fringe pattern is not visible at all. Scattering effects (mechanism 1) would manifest themselves as change in modulation depth of the FP fringes as seen in Fig. S9a- S9d, whereas change in effective FP cavity size (mechanism 2) would manifest itself as a change in frequency spacing and angular position of the FP fringes.

Overall these results suggest that the NVPC acts as a buried scattering layer within the epitaxy layers of the LED, scrambling the otherwise weakly confined FP modes supported by the GaN epitaxial layers (mechanism 1 described earlier). We can therefore infer that increased outcoupling is mainly due to diffuse scattering of weakly confined higher order modes to radiative modes rather than reduction of total number of modes.

It is possible that both mechanisms occur simultaneously. Thus light extraction is increased as the effective cavity size is reduced and due to the diffuse reflection from the NVPC layer, FP modes (or other confined modes) are not supported by the epitaxy layers of the LED.

Fig. 3 The characteristics of PCLED and reference LED : (a) Light output power-current-voltage characteristic at various current densities, the inset are the real chip at a driving current of 25 mA. (b) EQE and normalized EQE at various current densities. (c) Reflectance spectra of GaN template with and without triple-layer NVPCs with 80nm-thick SiN_x as anti-reflective layer. (d) The relative PL efficiency at various excitation power at 15 K and 300 K. (e) The relationship between the reflectivity enhancement, the IQE enhancement, the LOP enhancement and the calculated LOP enhancement of PCLED.

Fig. 4 (a) Calculated and normalized field distribution in reference LED and PCLED. (b) Calculated light output as a function of simulation time. (c) Measurement setup for electroluminescent angular far-field emission mapping. (d) Normalized electroluminescent far-field emission pattern of reference LED and PCLED. (e) Normalized electroluminescent far-field emission mapping of (i) reference LED and (ii) PCLED.

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4. Conclusions

In this work, NVPCs were fabricated to embed within the GaN/InGaN LEDs by combining ELO and nano-sphere lithography techniques. The NVPCs act as an efficient scattering back-reflector to greatly boost the LEE by a factor of 78% and collimate the view angle of LEDs from 131.5° to 114.0° . This could be because the highly scattering nature of NVPCs which reduce the interference giving rise to Fabry-Perot resonance. Moreover, with the help of NVPCs, the ELO has been achieved and led to the improved IQE and droop behavior due to the threading dislocation suppression and strain relief. Consequently, the LOP can be enhanced by 151% at a driving current of 350 mA, as comparing to reference LED. The concept presented in this study paves the way to the high-efficiency directional LEDs for next-generation solid-state lighting.

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