Supporting Information

Bridging "green gap" of LEDs:

Giant light output enhancement and directional control of LEDs via embedded nano-void photonic crystals

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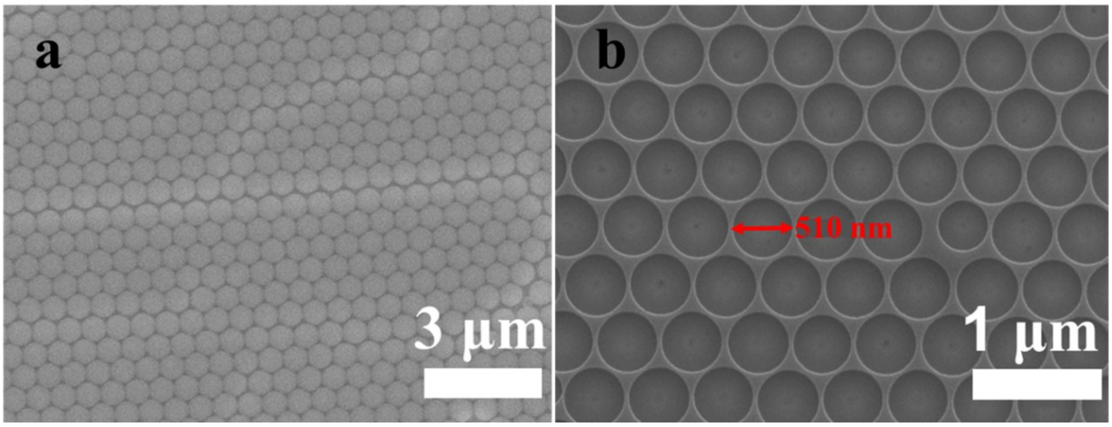

Fig. S1. The SEM image of (a) close-packed monolayer of polystyrene (PS) nano-sphere on GaN template, and (b) a nickel hard mask with honeycomb-like structures.

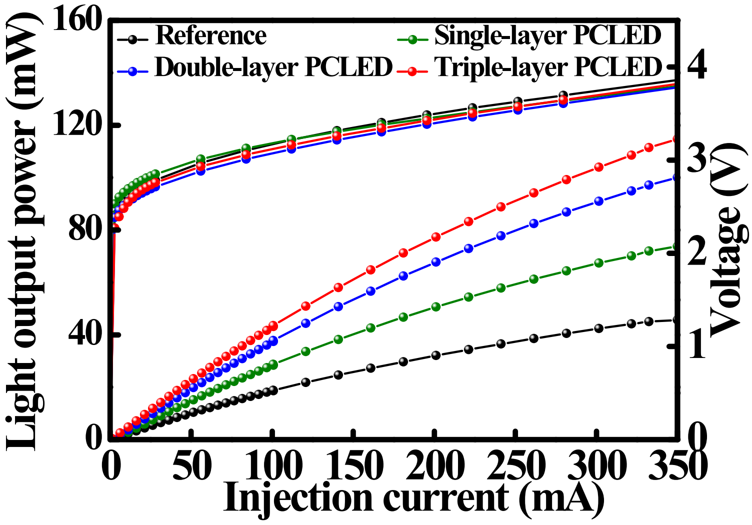

Fig. S2. Current-voltage characteristic and light output power of LEDs with different numbers of NVPC layers at various injected current.

The Raman spectra and residual stress of LEDs with different NVPC layers

The residual strain in GaN epitaxial layers can be estimated by the shift of the E_2 (high) mode. In Fig. S3, the E_2 (high) phonon peaks of Raman scattering were observed at 570.2, 569.88, 569.57 and 569.25 cm^{-1} for reference LED, LED with single-, double-, and triple-layer NVPC layers, respectively. As the Raman peaks shift toward a lower value, the strain is relieved in GaN epitaxial layers. The relation between the stress and the Raman peak shift of E2 (high) mode can be correlated by: [S1]

$$\Delta\omega_{E2} = \omega_{E2} - \omega_0 = C\sigma \tag{S1}$$

where the ω_{E2} is the measured E_2 (high) phonon frequencies, ω_0 is the phonon frequencies of the strain free GaN layer (566.5 cm^{-1}), C is biaxial strain coefficient and σ is in-plane stress, respectively. The Raman peak shift was 3.7, 3.38, 3.07 and 2.75 cm^{-1} for the reference LED and LED with NAVs layer from 1 to 3 layers and the corresponding residual stress are 1.45, 1.32, 1.2 and 1.07 GPa. This result indicates the existence of NVPCs can affect the residual stress in GaN thin film.

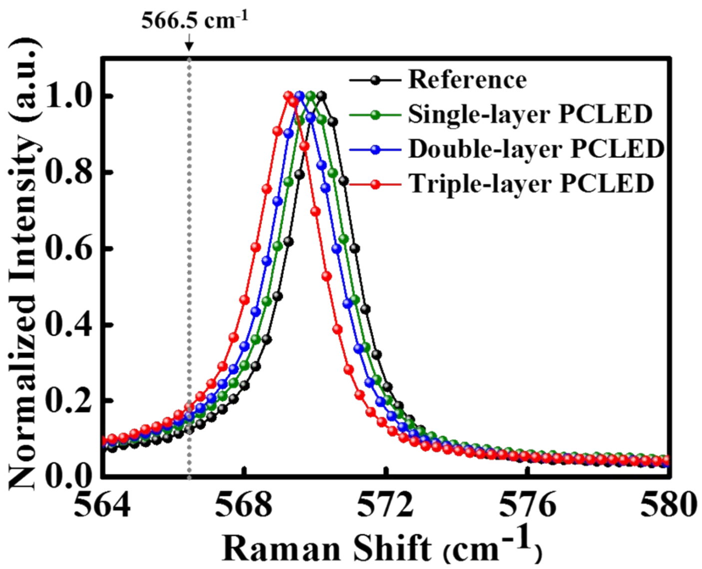

Fig. S3. Raman spectra of GaN/InGaN MQW LEDs with different numbers NVPC layers.

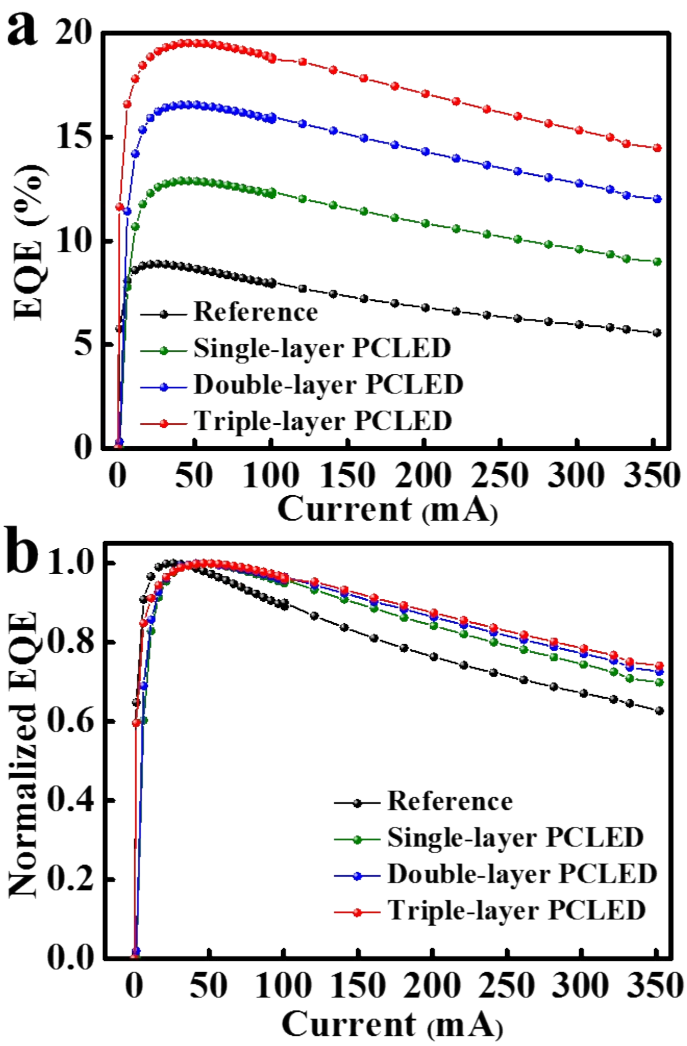

Fig. S4. (a) External quantum efficiency (b) normalized external quantum efficiency of LEDs with different numbers of NVPC layers at various current densities.

The calculation of incoherent reflection

One of the characteristics of NVPCs is the strongly scattered photons during operation. When an optical structure scatters the photons in various directions, the reflection or transmission will not be able to preserve phase information as the general EM wave case, only the magnitude of the field can be added up, which is usually called incoherent reflection or transmission.[S2] Fig. S5a shows multiple reflection between interfaces in triple-layer NVPCs. The software, TFcalc, was used to calculate the reflectance at first interface (air and 80-nm-thick SiN_x) which was denoted as R_1, and T_1=1-R_1. The reflectance of interface 2, 3 and 4 (different layer of NVPCs and GaN) is denoted as R_2, R_3 and R_4, respectively, and T_2=1-R_2, T_3=1-R_3 and T_4=1-R_4. In this calculation, the absorption of GaN and high order reflectivity are neglected. The reflectance of the LEDs with single-, double-, and triple-layer NVPCs can be calculated by the following formulas:

$$R_{reference} = R_1 \quad \text{(S2)}$$

$$R_{Single-layer\ nano-void\ PC} = R_1 + {T_1}^2 R_2 \quad \text{(S3)}$$

$$R_{Double-layer\ nano-void\ PC} = R_1 + {T_1}^2 R_2 + {T_1}^2 {T_2}^2 R_3 \quad \text{(S4)}$$

$$R_{Triple-layer\ nano-void\ PC} = R_1 + {T_1}^2 R_2 + {T_1}^2 {T_2}^2 R_3 + {T_1}^2 {T_2}^2 {T_3}^2 R_4 \quad \text{(S5)}$$

Fig. S5b shows the measured reflectivity of all samples. One can see is the deterioration of Fabry-Perot effect, which is attributed to the disruption of coherence of light. To fit the measured reflectivity, R_2, R_3 and R_4 are set with the same value of 1.9%. Fig. S5c shows the calculated reflectivity of all samples under incoherent reflective scheme, which shows a good agreement with the measured results shown in Figure S5b.

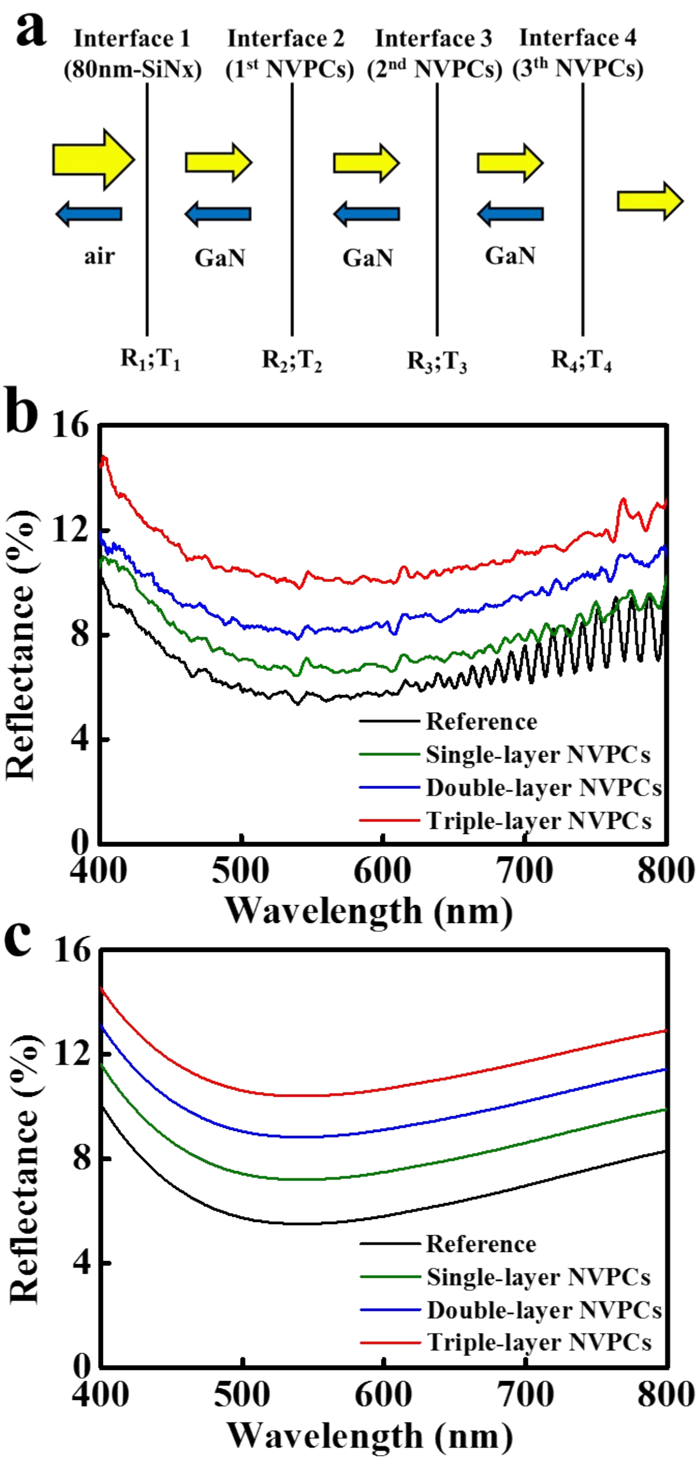

Fig. S5. (a) The illustration of reflectivity of multiple interface in triple-layer NVPC LEDs. (b) Measured and (c) calculated reflectance spectra of GaN template with different layer numbers of NVPCs with 80-nm-thick SiN_X as an anti-reflective layer.

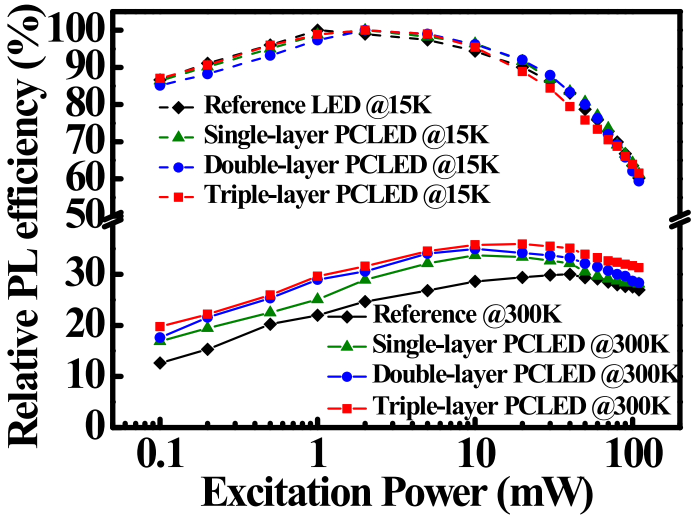

Fig. S6. The relative PL efficiency of the LEDs with different numbers of NVPC layers at various excitation power at 15 K and 300 K.

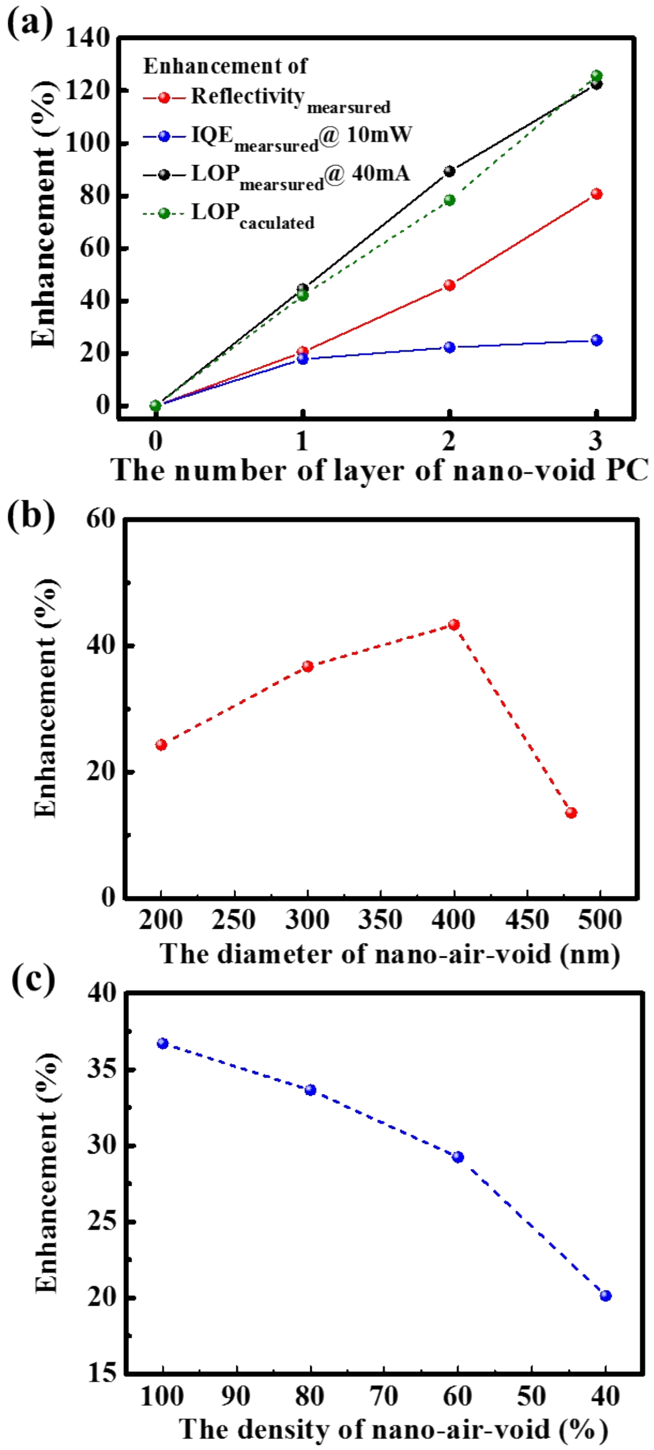

Fig. S7. (a) The relationship between the enhancement of IQE, measured LOP, calculated LOP and reflectivity as the function of the NVPC layers (experimental data). The light extraction enhancement as a function of nano-air-void (b) diameter (c) density (simulation data).

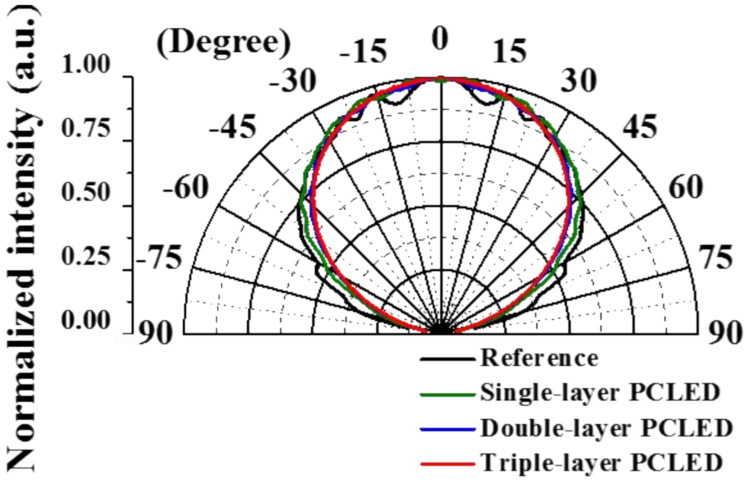

Fig. S8. Normalized far field emission pattern of LEDs with different numbers of NVPC layers.

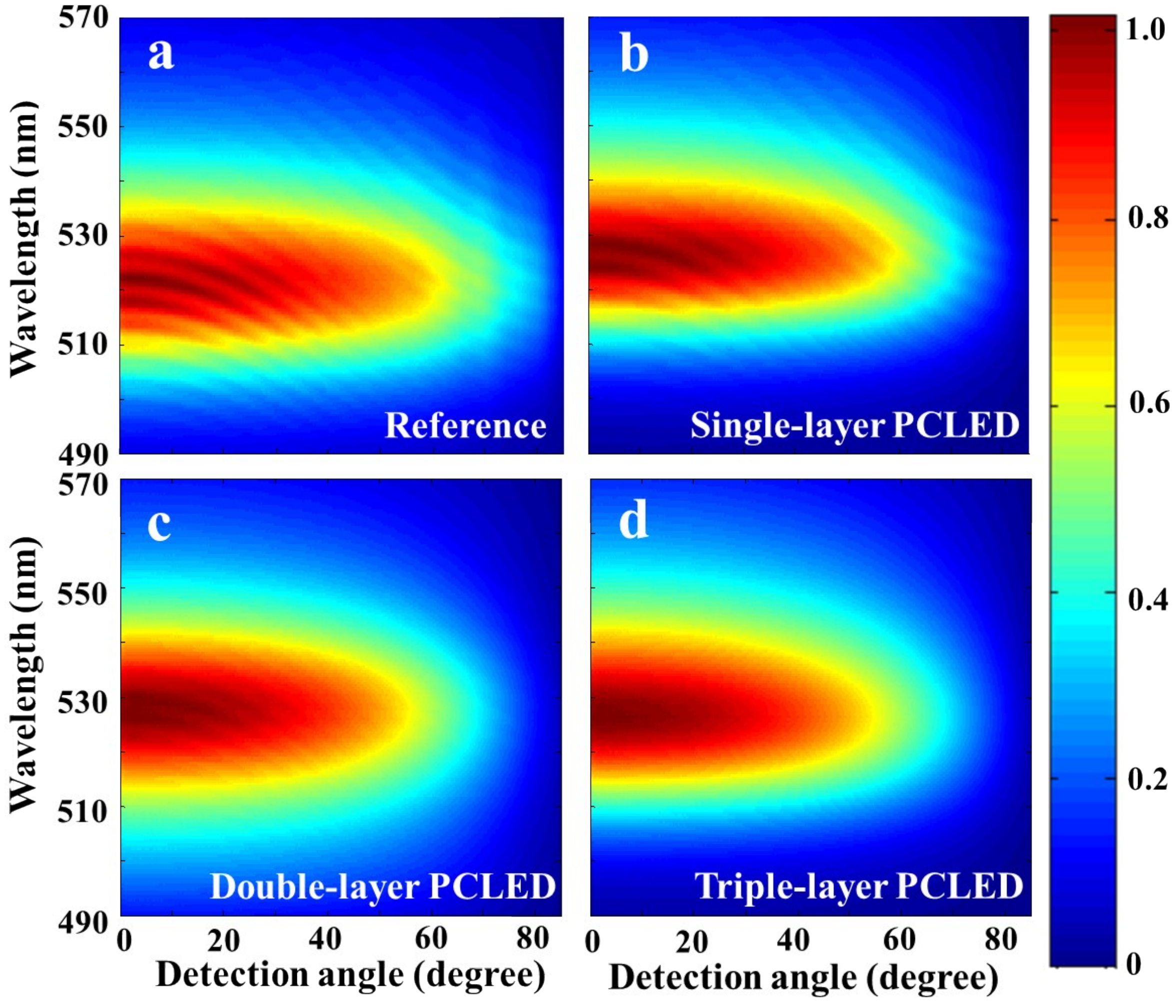

Fig. S9. Normalized angular far field emission mappings LEDs (a) without NVPCs; with (b) single-, (c) double- and (d) triple-layer of NVPCs.

Reference

(S1) Puech, P.; Demangeot, F.; Frandon, J.; Pinquier, C.; Kuball, M.; Domnich, V.; Gogotsi, Y. *J. Appl. Phys.* **2004**, 96, 2853.

(S2) Harbecke, B. *Appl. Phys. B* **1986**, 39, 165-170.