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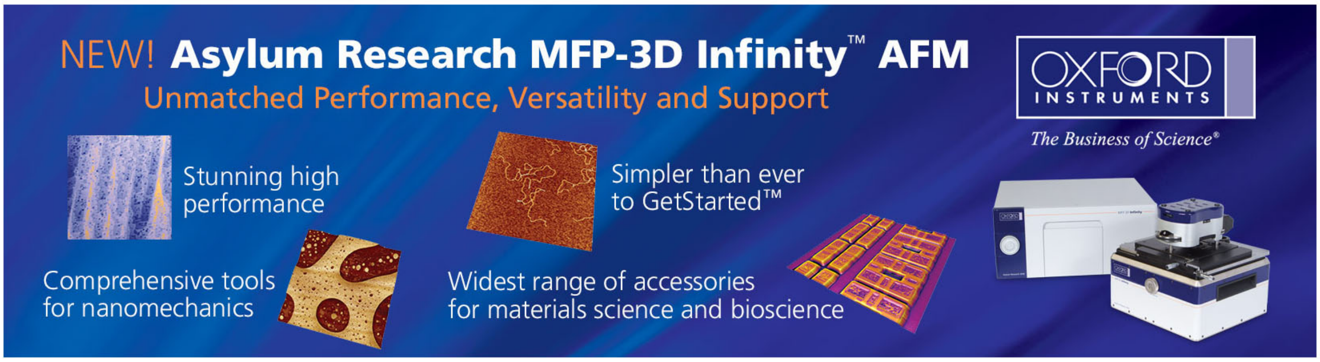

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Efficiency and droop improvement in green InGaN/GaN light-emitting diodes on GaN nanorods template with SiO_2 nanomasks

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This study presents the green InGaN/GaN multiple quantum wells light-emitting diodes (LEDs) grown on a GaN nanorods template with SiO_2 nanomasks by metal–organic chemical vapor deposition. By nanoscale epitaxial lateral overgrowth, microscale air voids were formed between nanorods and the threading dislocations were efficiently suppressed. The electroluminescence measurement reveals that the LEDs on nanorods template with SiO_2 nanomasks suffer less quantum-confined Stark effect and exhibit higher light output power and lower efficiency droop at a high injection current as compared with conventional LEDs. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4768950]

In recent decades, group-III nitride material has been regarded as one of the most promising materials for developing full-color light-emitting diodes (LEDs) because it has a wide range of direct bandgaps (0.7–6.2 eV). The high efficiency blue InGaN-based LEDs have been combined with red and yellow phosphors to provide next generation white-light sources. However, the internal quantum efficiency (IQE) of InGaN-based LED becomes low as the emission energy decrease to green or red emission light. The inefficiency of green and red InGaN-based LEDs is caused by the severe quantum-confined Stark effect (QCSE) induced by high indium composition in quantum wells and strong internal piezoelectric field. An alternative quaternary material InGaAlP has been used to develop high quantum efficiency red LEDs for years,[1] while the green LEDs are still left without a suitable solution. The lag of the green LED efficiency limits the development of LED in both solid-state lighting (SSL) and display applications. As a result, solving this so-called "efficiency green gap" and developing the equally efficient red, green, blue (RGB) LEDs are the most significant issues.

The external quantum efficiency (EQE) of an LED can be expressed as the product of current injection efficiency, internal quantum efficiency, and light extraction efficiency (LEE). Our previous studies have demonstrated that by using nanoscale patterned sapphire substrate (NPSS) technique, the IQE and LEE of a blue LED can be greatly improved.[2,3] In addition, we also employed the technologies of nanopyramid structure[4] and m-plane bulk substrate[5] to fabricate semipolar and nonpolar blue-green LEDs. Owing to their high carrier density dependent wavelength stability and low efficiency droop, they show a promising potential in developing high indium content LEDs. In order to develop high efficiency green LEDs, Li *et al.* also reported an improvement in both IQE and LEE for a 525 nm green LED on the nanopatterned sapphire substrate.[6] In this work, a nanoscale patterned substrate, comprised high aspect ratio GaN nanorods (NRs) and SiO_2 nanomasks on the top of NRs, was applied to develop high performance green LEDs. The growth mechanism, the enhancement of LEE, and the properties of band diagram were also discussed in detail.

The GaN nanorods template was prepared in the following procedures:[7] (1) deposition of a 2 μm thick undoped GaN on c-plane sapphire by metal–organic chemical vapor deposition (MOCVD); (2) deposition of a 200 nm SiO_2 layer by plasma-enhanced chemical vapor deposition (PECVD); (3) evaporation of a 10 nm thick Ni layer followed by rapid thermal annealing (RTA) with a flowing nitrogen gas at 850 °C for 1 min to form self-assembled Ni clusters with approximately 200 nm in diameter; (4) dry etching with the Ni clusters served as etch masks for forming NRs by reactive ion etching (RIE) and inductive coupled plasma (ICP); (5) removal the residual Ni masks by dipping the sample into nitric acid solution (HNO_3) at 100 °C for 5 min. Figure 1(a) shows the cross-sectional scanning electron microscopy (SEM) image of the GaN NRs with SiO_2 nanomasks. The height of the GaN NRs is about 2 μm while the diameters of them are in a range of 200–300 nm. The green InGaN/GaN multiple quantum wells (MQWs) LED structure was grown on this GaN NRs template by a low pressure MOCVD (Veeco D75) system. For comparison, a sample with the same green LED structure was also grown on c-plane sapphire. The epitaxial structure consists of a 3 μm n-GaN, six periods of 3 nm InGaN QWs and 12 nm GaN barriers, a 20 nm p-$Al_{0.1}Ga_{0.9}N$ electron blocking layer (EBL), and a 0.2 μm p-GaN. Typical n-(Si) and p-(Mg) type dopants were used. Subsequently, the LED chips were fabricated by regular chip process with $300 \times 300\,\mu m^2$ diode mesas, indium-tin-oxide (ITO) current spreading layer, and Ni/Au contact pads.

To investigate the mechanism of nanoscale epitaxial lateral overgrowth (NELOG) in detail, the transmission electron microscopy (TEM) was employed. Figure 1(b) shows

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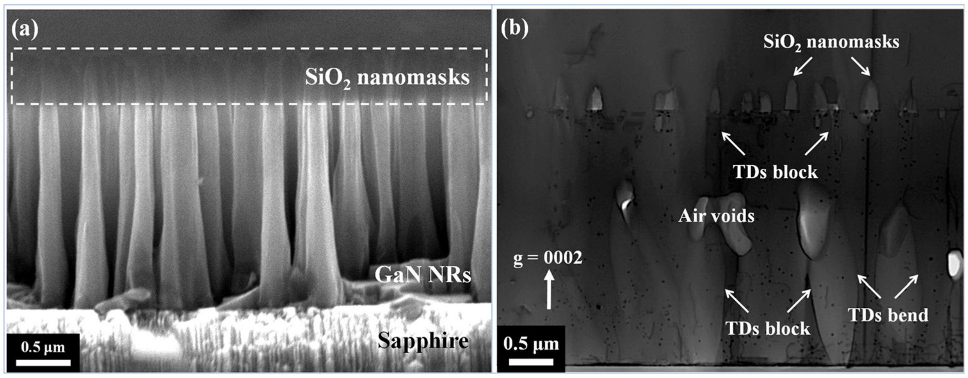

FIG. 1. Cross-sectional (a) SEM image of GaN NRs template and (b) TEM image of the GaN epilayer overgrown on GaN NRs template.

the cross-sectional TEM image of GaN epilayer overgrown on the GaN NRs template with SiO_2 nanomasks. Owing to each NR has a SiO_2 nanomask on the top of it, the GaN epilayer can only grow on the sidewall of NRs. Our previous study have showed that this growth process is a combination of GaN epilayer grown on the M-plane (10-10) sidewall of NR and inclined R-plane facets (1-102) which are close to the top of the NRs.[8] By laterally growing GaN epilayer on these two kinds of facets, the spacing between NRs become closer and finally coalesces. However, because of the rapid lateral growth and the high aspect ratio of NRs template, microscale air voids were formed between NRs, as shown in Figure 1(b). In addition, one can observe that there are many threading dislocations (TDs) below the SiO_2 nanomasks layer, while the TDs above this layer become much less than below. There are two reasons for the reduction of TDs in GaN epilayer above the SiO_2 nanomasks layer. First, the formation of air voids can act as microscale mask to prevent the TDs from going through to the upper epilayer. In addition, the SiO_2 nanomasks have the similar function as the air voids, which also block parts of TDs. Second, due to the strong lateral growth by using NELOG, the TDs tend to follow the growth direction. This characteristic gives the TDs a chance to bend to the air voids and the SiO_2 nanomasks. The cross-sectional TEM image indicates that many TDs are ended with these two types of masks, which results in a high crystalline quality GaN epilayer.

One of the problems leading to the inefficiency of green LED is the QCSE induced by high indium composition in quantum wells and strong internal piezoelectric field. To keep the wavelength of green LED, it is hard to decrease the indium composition for improving the QCSE. Under this circumstance, it is much important to decrease the residual strain in the epitaxial layer. The Raman scattering spectrum was performed to measure the strain of the GaN epilayer on both GaN NRs template and sapphire substrate. The E_2 (high) peaks of the Raman scattering spectrum of the GaN epilayers on NRs template and sapphire substrate are at 568.5 and 570.5 cm^{-1}, respectively (not shown here). From the E_2 (high) peaks, the in-plane compressive strain can be estimated to be 0.88 GPa and 1.77 GPa by using the following equation:[9]

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

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. The Raman scattering spectrum indicates that the GaN epitaxial layer on GaN NRs template exhibited lower compressive strain than GaN epilayer on sapphire. With the strain relaxation characteristic of GaN epilayer grown on GaN NRs template, one can expect that the QCSE in the green MQWs grown on this template can be suppressed, which causes an increase in wave function overlap between holes and electrons and consequently enhance the IQE of the MQWs.

In order to clarify the influence of QCSE on the LED devices, the electroluminescence (EL) system was employed. Figures 2(a) and 2(b) show the EL spectra under different injection current for the LED devices with the dimensions of 300 × 300 μm^2 on GaN NRs template and sapphire substrate, respectively. The insets of them show the corresponding EL emission peak wavelength as a function of injection current. The emission peak wavelength of NR-LED is slightly redshifted from C-LED since the effect of strain relaxation may

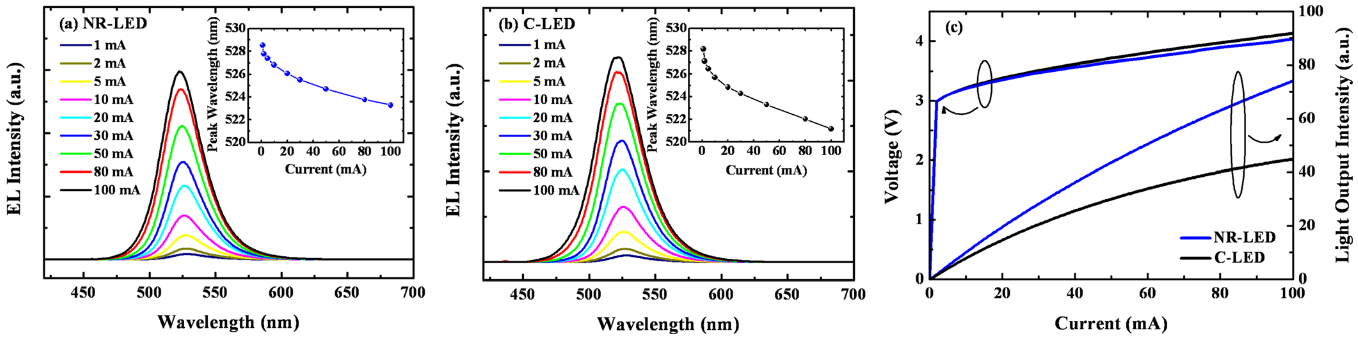

FIG. 2. The EL emission peak wavelength as a function of injection current for (a) NR-LED and (b) C-LED. (c) Forward voltage and light output power as a function of injection current of NR-LED and C-LED.

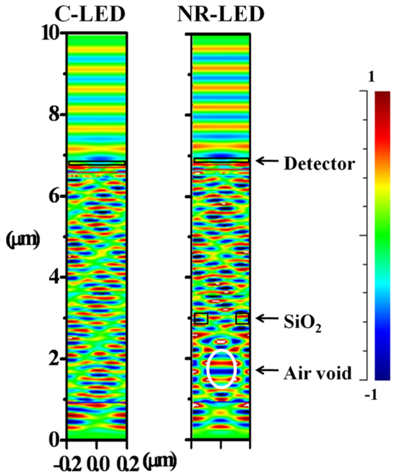

FIG. 3. 3D-FDTD calculated electric-field distribution of NR-LED and C-LED.

slightly increase the indium incorporation rate.[10,11] In addition, as the injected current increases, the emission peak wavelength of NR-LED exhibits a blue-shift by 5.25 nm, which is less than 7.03 nm for C-LED. This result reveals that the QCSE is weaker for NR-LED because parts of residual strain had been released through the NELOG. Figure 2(c) shows the power-current-voltage (L-I-V) curves of NR-LED and C-LED. The forward voltages (V_f) at an injected current of 20 mA for NR-LED and C-LED were 3.35 and 3.38 V, respectively, which reveal the electrical characteristics of these two devices are similar. On the other hand, the light output powers of NR-LED were 33.3% and 65.5% higher than that of C-LED at 20 mA and 100 mA current injection.

The enhancement of light output power for NR-LED can be attributed to several reasons. First, the TDs were greatly reduced by the microscale air voids and the SiO$_2$ nanomasks, which can effectively suppress the formation of nonradiative centers. Second, the weaker QCSE for the MQWs on NRs template enhances the recombination of electron-hole pairs. Third, the LEE for a blue LED with the embedded air voids can be greatly enhanced.[7] To quantify the light extraction efficiency for NR-LED and C-LED, a 3D finite difference time domain (FDTD) simulation was applied by using FULLWAVE$^{\text{TM}}$ program.[12] The simulated model of NR-LED includes the 5×5 random NRs with the height of 2 μm and 7.56 μm^2 for each unit cell. In addition, the microscale air voids and SiO$_2$ nanomasks were extracted from the SEM image in Figure 1(b). A detector was placed on the p-side of the simulated LED structure to collect the light output intensity emitted from MQWs. Figure 3 shows the calculated scattered light-wave distribution for both cases. The higher calculated electric-field distribution at the detector for NR-LED than C-LED implies that the NR-LED with better light extraction efficiency, which results in higher light intensity. The overall enhancement of LEE acquired from the steady-state light intensity for NR-LED was 1.37 times higher than that of C-LED. However, the enhancement of LEE might be a little overestimated due to the imperfection of air void shapes and sizes and the random distribution of them in the real GaN epilayer. This is why the light output power of NR-LED was only 1.33 times higher than that of C-LED at 20 mA.

Moreover, the normalized efficiency of NR-LED and C-LED as a function of injection current is shown in Figure 4(a). The efficiency droop, which is defined by η_{peak}-$\eta_{100\text{ mA}}$/η_{peak}, for NR-LED (25.9%) is smaller than C-LED (56.1%). To explore the origin of efficiency droop for NR-LED and C-LED, the APSYS simulation software was used to analyze the band structure of the green MQWs for both cases.[13] The simulated parameters, including a Shockley–Read–Hall recombination lifetime and an Auger recombination coefficient were set as 5 ns and 2×10^{-30} cm^6/s, respectively. The simulated results of efficiency droop for both NR-LED and C-LED, which are also shown in Figure 4(a), agreed with the experimental data. Moreover, the corresponding energy band diagram of NR-LED and C-LED at a forward current density 100 mA is shown in Figure 4(b). It is reasonable that the band diagram of C-LED is severely bent due to the high piezoelectric field. The severe upward conduction band edge from n-GaN side to MQWs and the triangular MQWs greatly suppressed the radiative recombination of electron-hole pairs. In comparison, the band diagram of NR-LED is more uniform due to less QCSE. As a result, the NR-LED exhibits lower efficiency droop than the conventional one.

In conclusion, we have demonstrated the green InGaN/GaN MQWs LEDs grown on a GaN NRs template with SiO$_2$ nanomasks. The improvement of EQE and efficiency droop can be attributed to high IQE and LEE for green LEDs on NRs template. The threading dislocations and the residual strain are effectively suppressed by using NELOG. The embedded microscale air voids and SiO$_2$ nanomasks not only improve the crystalline quality but also greatly enhance the LEE of the green LEDs. The corresponding simulated results are agreed well with experimental results.

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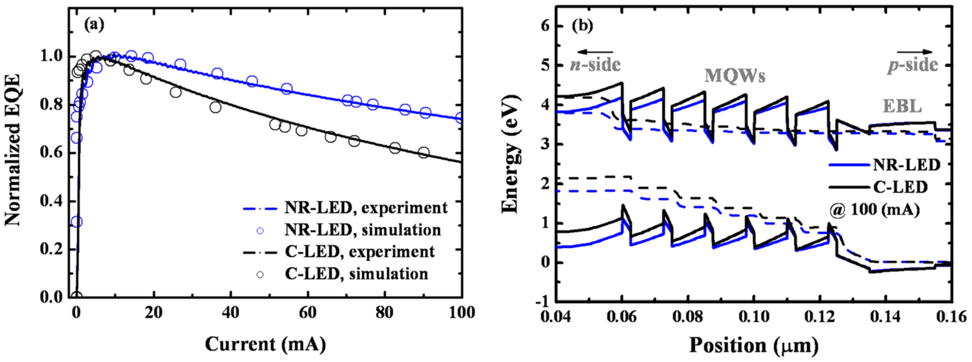

FIG. 4. (a) Experimental and simulated EQE as a function of injection current for NR-LED and C-LED. (b) The calculated band diagram of NR-LED and C-LED under a 100 mA forward bias operation.

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