Defect-reduced green GalnN/GaN light-emitting diode on nanopatterned sapphire

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Green GaInN/GaN quantum well light-emitting diode (LED) wafers were grown on nanopatterned *c*-plane sapphire substrate by metal-organic vapor phase epitaxy. Without roughening the chip surface, such LEDs show triple the light output over structures on planar sapphire. By quantitative analysis the enhancement was attributed to both, enhanced generation efficiency and extraction. The spectral interference and emission patterns reveal a 58% enhanced light extraction while photoluminescence reveals a doubling of the internal quantum efficiency. The latter was attributed to a 44% lower threading dislocation density as observed in transmission electron microscopy. The partial light output power measured from the sapphire side of the unencapsulated nanopatterned substrate LED die reaches 5.2 mW at 525 nm at 100 mA compared to 1.8 mW in the reference LED. © 2011 American Institute of Physics. [doi:10.1063/1.3579255]

All-color group-III nitride light-emitting diodes (LEDs) are the likely solution for energy efficient and high color quality lighting.^{1,2} The external quantum efficiency (EQE) is the product of current injection efficiency (INJ), internal quantum efficiency (IQE), and light extraction efficiency (LEE). In thin film LEDs, LEE can be controlled by chip surface shaping, while IQE is strongly affected by crystal defects in the quantum well (QW) layers, particularly for the longer wavelength green and yellow LEDs. 3,4 The patterning of the growth surface of a substrate on the micrometer range, has shown to improve light output power (LOP) in blue LEDs and was attributed to enhanced LEE (Refs. 5 and 6) while in near-UV LEDs, LOP enhancement was also attributed to IQE improvements. For substrate patterning on the nanometer scale, a 33% LOP improvement in 455 nm blue LEDs was attributed to an enhanced LEE.⁸ Chiu et al.⁹ report an equal enhancement of both in 450 nm blue LEDs using SiO₂ nanorod covered sapphire as a substrate. Here we study 525 nm green LED wafers on nanopatterned sapphire substrate and quantify the contributions of IQE and LEE in the overall performance enhancement.

The basal plane growth surface of sapphire wafers was patterned using nano-imprint lithography in four steps: ¹⁰ (1) replication of a nanopattern by pressing a patterned silicon transfer die against a resin-covered sapphire wafer; (2) removal of backlog resin by reactive ion etching (RIE) in O₂ ambient; (3) sapphire etching to a depth of 125 nm by RIE under BCl₃ ambient, and (4) removal of resin by RIE under O₂ ambient. The final pattern is a hexagonal array of cylindrical holes (diameter 250 nm, period 450 nm). Green GaInN/GaN QW LED structures were grown by vertical flow metal-organic vapor phase epitaxy on the so prepared substrates. For comparison, a conventional reference LED was simultaneously grown. Ga-face growth was performed

TEM micrographs of the unpatterned structure [Fig. 1(a)] show the typical threading dislocations (TDs) emerging from the sapphire/GaN interface and to propagate along the growth direction. In the patterned substrate LED [Fig. 1(b)], a much lower number of TDs is found to originate from the

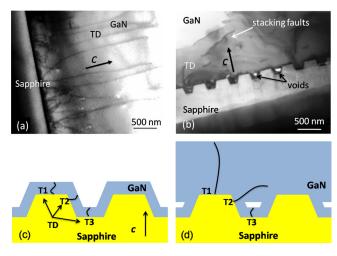


FIG. 1. (Color online) Cross-section TEM of the epitaxial GaN/sapphire interface on (a) planar and (b) nanopatterned sapphire. Schematics showing early (c) and late (d) phases of the TD (T1, T2, T3) formation on the nanopatterned substrate.

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along the polar c-axis of GaN: (1) a low-temperature deposited GaN buffer layer; (2) 6 μ m n-GaN; (3) eight periods of 3 nm Ga_{1-x}In_xN QWs and 20 nm GaN barriers; (4) a 10 nm p-Al_{0.18}Ga_{0.82}N electron blocking layer, and (5) 0.2 μ m p-GaN. Typical dopants of Si and Mg were used. The Ga_{1-x}In_xN alloy composition of the QWs (0.10<x<0.20) was determined by an x-ray diffraction and transmission electron microscopy (TEM) analysis at 200 keV.¹¹ The far field emission pattern of fabricated dies was measured by a photodetector across a half sphere around the LED.

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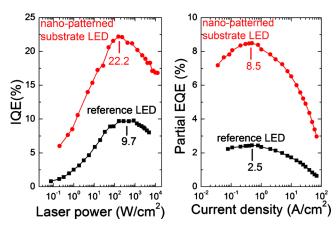


FIG. 2. (Color online) (a) IQE as a function of excitation power density, (b) EQE as a function of current density for the nanopatterned substrate (circles) and the reference (squares) LED.

same interface. A few dislocations with strong inclination and stacking faults are seen to annihilate without progressing to the active region. Within the substrate's patterned holes, voids can be seen indicating overgrowth in lateral direction. These areas are free from further TDs and no defect of growth front coalescence is seen. We propose the following scheme for the early [Fig. 1(c)] and late [Fig. 1(d)] phases of the TD formation: TDs initiated at the bottom of the etched substrate (T3) are mostly stopped by open voids. TDs originating from the inclined facets change direction to propagate within the growth plane (T2). Only TDs originating in the un-etched portions of the substrate (T1) propagate toward the active region. The TD density, estimated from plane view TEM, is 3.6×10^8 cm⁻² on the patterned and 6.4 $\times 10^8$ cm⁻² on the planar substrates. The GaN (102) x-ray diffraction shows a linewidth of 427 arcsec on the patterned and 524 arcsec for the reference. Both together prove that nanopatterning of the substrate can improve the quality of epitaxial GaN.

For an upper limit of IQE, the photoluminescence (PL) efficiency as a function of excitation power density was measured at room temperature (RT) and at 7 K. IQE at RT was determined by scaling to the point of highest PL efficiency at low temperature [Fig. 2(a)]. By choosing an excitation wavelength of 408 nm, within the bandgap of GaN, photocarriers are generated directly within the QWs. We therefore, assume INJ as unity.

IQE varies strongly with excitation power density. The patterned LED shows a maximum of 22% at $155~\text{W/cm}^2$ while the reference only reaches 9.7% at $278~\text{W/cm}^2$. Apparently, the patterned substrate LED outperforms the unpatterned by a factor of 2.0 to 2.2.

Electroluminescence (EL) of the fabricated LED dies (without encapsulation) of dimensions $350\times350~\mu\text{m}^2$ was measured from the substrate side. Both LEDs show a single green emission line. LOP of the patterned substrate LED reaches 2.3 mW (5.2 mW) at 537 nm (523 nm) with a line width (full width half maximum) of 42 nm compared to 0.88 mW (1.8 mW) at 541 nm (527 nm) with a line width of 46 nm in the reference at 33 A/cm² or 30 mA (110 A/cm² or 100 mA). EQE versus current density [Fig. 2(b)] shows a maximum of 8.5% at 0.5 A/cm² for the patterned LED and 2.5% at 0.8 A/cm² for the reference. This amounts to a large

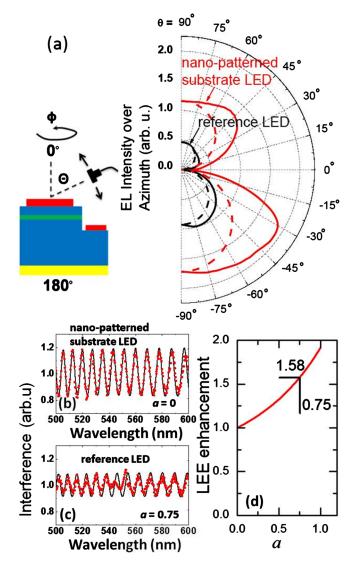


FIG. 3. (Color online) (a) Measured (solid) EL and Lambertian emission patterns (dashed) of nanopatterned and reference LED. Inset shows the geometry. Measured (dots) and simulated (line) interference fringes for the nanopatterned substrate (b) and reference (c) LEDs; (d) simulated LEE enhancement as function of scattering coefficient *a*.

The far-field emission pattern was analyzed by angleresolved EL measurement in upper and lower quarter spheres. Figure 3(a) shows the azimuth ϕ -integrated LOP at 5 mA versus elevation θ . LOP is found higher through the substrate side than for the partially blocking p-contact side. For planar surface emitters a Lambertian distribution pattern is expected with maxima at normal directions [Fig. 3(a), dashed lines]. The reference LED mostly follows this model while the nanopatterned substrate LED strongly exceeds the pattern near θ =45° and θ =-40° emission.

The simulation of photonic crystal LEDs finds that emission in the $|\theta|$ =20°-45° range should originate in optical modes of the GaN layer modes that find extraction beyond the regular escape cone by help of a photonic crystal surface pattern. We may assume that the nanopatterned sapphire/GaN interface serves the same purpose in our case. The wider distribution in the lower half sphere is likely due to the intermediary refractive index of the sapphire layer leading to a wider deflection of the enhancement maximum.

2.5% at 0.8 A/cm² for the reference. This amounts to a large To quantify the role of substrate patterning on LEE, the

 $(\theta=-90^\circ)$ EL spectra were analyzed. A relative interference spectrum was deduced by dividing the raw spectrum by a $\Delta\lambda=10$ nm wavelength-averaged emission spectrum [Fig. 3(c), dots]. The mean interference amplitude of the nanopatterned substrate LED was found to be 50% of that in the reference.

This result can be simulated by multiple internal reflection modes under normal incidence. The subwavelength nanopatterned interface is described by a variable light power scattering factor a, $0 \le a \le 1$, with a = 0 for the reference. The experimental ratio of inference amplitude leads to a = 0.75 in the patterned LED data. This implies that 3/4 of the internally incident light is scattered by the nanopattern and may fall into the extraction cone, while the remaining 1/4 forms the interference pattern.

Without scattering, the fraction of light to fall into the escape cone of $2\varphi_c$ is $\eta_0 = [1 - \cos(2\varphi_c)]/2$. Including scattering, the same increases to $\eta_s = 1/[1 - a(1 - \eta_0)]$. From this the LEE enhancement factor is calculated as a function of the scattering coefficient a [Fig. 3(d)]. For a = 0.75, LEE=1.58 was interpolated.

In conclusion, green GaInN-based LEDs on nanopatterned sapphire substrate were quantitatively compared to reference LEDs on planar substrate. The GaN epilayer on the patterned substrate shows a 44% lower TD density. This is found to enhance IQE by a factor of 2.24, i.e., reduce non-radiative recombination from 90% to 78%, as determined in temperature and excitation density dependent PL. From an analysis of interference fringes we derive an LEE enhancement factor of 1.58. Both effects combined can explain a measured 3.4-fold enhancement of EQE and LOP in the green LED on nanopatterned sapphire substrate as prepared simultaneously in our standard processes. Such substrate therefore has the potential to both, enhance light generation efficiency and light extraction without any surface pattern-

ing. This could potentially be also a benefit over most micropatterned substrate technologies.

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