MOCVD growth of GaN nanopyramid and nanopillar LED with emission in green to orange color

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ABSTRACT

We report the fabrication and demonstration of electrically driven green, yellow-green, and amber color nanopyramid LEDs. The quantum wells were grown on nanopyramid facets, which have low polarization field and allow high In incorporation.

Keywords: GaN LED, nanopyramid, MOCVD epitaxy

1. INTRODUCTION

The III-nitride based semiconductor light emitting diode (LED) has gained great research interests because of its promising potential in high efficient lighting applications. The bandgap of InGaN semiconductor can be varied from UV to near IR. In principle, such devices can cover UV to visible wavelength and enable smart lighting applications. InGaN/GaN multiple quantum wells (MQWs) in blue emission have been extensively studied. Conventional InGaN/GaN multiple quantum wells (MQWs) are often grown on c-plane GaN surface. The emission wavelength is typically in the blue region, where the efficiency is optimal. To move to longer wavelength, high In incorporation is required. However, the internal quantum efficiency (IQE) drops significantly with increasing In content, in particular for emission at green and longer wavelength, because of the strain induced internal electric field (IEF) [1]. The electric field traverses through MQWs and results in significant spatial charge separation and therefore lower internal quantum efficiency. The IEF strongly depends on the crystallographic orientation. It can be greatly reduced by growing MQWs on semipolar or nonpolar planes. Substrates in these crystal orientations are however not readily available.

The semipolar and nonpolar crystal surfaces can be found in naturally formed crystalline structure by 3-dimensional epitaxial growth. Nano size hexagonal pyramids can be grown by selective area growth from the opening holes of a SiO\textsubscript{2} masked c-plane GaN substrate, which is more readily available. The pyramid facets are typically \{10-11\} or \{11-22\} semipolar planes. In addition to the intrinsic low polarization field of these crystal planes, the small footprint of nano structure onto the substrate reduces the strain build up, resulting in lower defect density and piezoelectric field. Photoluminescent (PL) studies of MQWs grown on these nano scale facets have demonstrated significant reduction in the polarization field and the increase of internal quantum efficiency [2-5]. There have been strong interests in using these semipolar pyramid facets for high In content LED applications. The emission wavelength as far as 600 nm has been reported. The experiments were however mostly carried out by photo excitation. Reports on electrical performance are very limited. [5] A thick layer of p-GaN was often grown on top of the nanopyramids to planarize the surface for electrical contacts. The thick p-GaN layer at the same time also compromised the electrical performance due to its high resistivity. The nanopyramid structure even though shows promising advantages in optical emission properties. The 3D nanogeometry often poses challenges for making electrical injection, which is an important issue to be studied from practical application point of view.

Here, we report the fabrication and performance of electrically driven nanopyramid LEDs. Emission wavelengths varying from blue, green, to amber colors by tuning In content concentration were demonstrated. The schematic of fabrication steps are shown in Fig. 1(a)-(d). SiO\textsubscript{2} nano disks of 250 \textmu m in diameter were first patterned on a n-GaN substrate. The SiO\textsubscript{2} disks were used as etching masks in inductively coupled plasma reactive ion etching (RIE). The SiO\textsubscript{2} disks were subsequently removed by a buffer oxide etch, leaving arrays of GaN nanopillars (Fig. 1(a)). Spin-on
glass was spun on the substrate to planarize the surface. After curing the spin-on glass at 400 °C for 60 min., the nanopillar side walls were covered by spin-on glass with air voids among them. The substrate was then etched by RIE to expose the top portion of nanopillars (Fig. 1(b)). The substrate was put back into MOCVD for GaN epitaxial regrowth. GaN pyramids were selectively grown on the tops of nanopillars (Fig. 1(c)). Ten pairs of InGaN/GaN MQWs were grown on the pyramid facets, followed by a 20-nm electron blocking layer of Mg-doped p-type AlGaN and a 200-nm Mg-doped p-type GaN layer. Figure 1(d) shows the plane view of the fabricated LED. The surface was rough due to the nanopillar structure. To test the wavelength tuning capability, three different In flow rates during MOCVD growth were used to fabricate three LED chips, targeting green, yellow-green, and amber emission. The fabricated LEDs are labeled as G-, Y-, and A-LED, respectively. The scanning electron microscopy (SEM) images of a typical sample at the corresponding intermediate fabrication steps are shown in Fig. 2 (a)-(d). Figure 2(a) shows the fabricated nanopillars after the nano imprint patterned etching. Figure 2(b) is the image of the nanopillars showing the spin-on-glass coated side walls and the exposed top surfaces. After epitaxial regrowth, pyramids were formed on tops of nanopillars as shown in Fig. 2(c). The air voids among nanopillars were remained due to the spin-on glass, which prevented the growth of GaN on the pillar side walls. The SEM plane view of the final fabricated LED is shown in Fig. 2(d). The light gray color regions are the n- and p-metal contacts. The rough surface is due to the nanopillar array structure.

The scanning tunneling electron microscopy (STEM) image of a typical fabricated nanopillar sample is shown in Fig. 3(a). It shows that MQWs are grown on the inclined pyramid facets. There are air voids among nanopillars due to the oxide passivation of nanopillar sidewalls. The pyramid semipolar plane is identified as \{10-11\} from the inclined 62° angle. The QW and barrier are clearly visible at the region away from pyramid apex and valley locations (Fig. 3(b)). The QW and barrier widths are 2 nm and 8 nm, respectively. The color at the pyramid coalescent valley is darker. This is an
indication of higher In content or coalescent defects. The QW-barrier boundary is not as clear at the upper region of V shape coalescent valley (Fig. 3(c)). Similar phenomenon is also observed at the pyramid apex region (Fig. 3(d)). The QW-barrier contrast is however better at the lower region of V shape coalescent valleys (Fig. 3(e)). The degraded well-barrier contrast is likely due the build up strain in QWs as layer number increases.

The emission property was investigated by the spectrally resolved cathodoluminescent (CL) measurement. The G-LED was used in the measurement to illustrate the general CL properties of the fabricated nanopyramid LEDs. A plane view scanning electron microscope (SEM) image was first taken, as shown in Fig. 4(a). The spectrally resolved CL images were then scanned at 500, 540, and 560 nm, as shown in Fig. 4(b)-(d). The emission pattern basically follows the pyramid height contour, as can be seen by comparing the bright contours in Fig. 4(b)-(d) to the pyramid shape in SEM image Fig. 4(a). The contours move toward the tips of nanopyramids as the observed wavelength is tuned to longer wavelength. It indicates that the MQW emission redshifts from the bottom to top region of nanopyramids. This may be due to the increase of In incorporation as the region moves up the nanopyramid facet. The In concentration in In$_x$Ga$_{1-x}$N quantum well was analyzed by energy dispersive x-ray spectrometer. It shows an increase of In from 15% to 30% when the sampled region moves from the valley to apex region, in consistent with the CL analysis.

These three substrates were fabricated into 300 µm x 300 µm LED chips using standard LED fabrication steps. Figure 5(a)-(c) shows the optical microscope (OM) images of these LEDs under electrical injection along with light-current-
voltage (L-I-V) curves. The emission is fairly uniform across entire LED surface, indicating well spread current despite the corrugated pyramidal surface. The dark pats are the p- and n-metal contacts. The I-V curves show reasonable diode turn on behavior around 3 volts. The turn-on threshold becomes less distinct as the sample changes from G-LED to A-LED. Above turn-on, the voltage increases significantly up to 6 volts at 200 mA. This is higher than the normal c-plane LED driving voltage, indicating a higher serial resistance. It is attributed to the imperfection in the current spreading layer on the corrugated surface, which requires further optimization. The L-I curves show typical increasing output power as current increases. There is however a slow turn on of light, which becomes worse in particular for A-LED. The light does not turn on until current reaches 40 mA, implying a significant current leakage. Since this problem becomes worse for the longer wavelength sample, it indicates that the increase of In content affects not only the optical emission but also the electrical injection efficiency. The leakage paths are likely at the pyramid coalescent boundaries, which become more defective with increasing In concentration.

The EL spectra in Fig. 5(d)-(f) show the peak emission wavelength shifts as current increases. The blue shift is a result of the screening of polarization field by the injected carriers and gradual turn on of MWQs on the different portion of nanopyramid facets. The blue shifts for G-, O-, and A-LED are about 13, 25, and 50 nm, respectively. The increasing
blue shift is because the strain induced polarization field in MQWs becomes larger as In concentration increases. These blue shift values are nevertheless still relatively small compared with c-plane MQWs, which would have much larger blue shifts. The extended PL tail at the longer wavelength side is due to the increasing In content in MQWs toward the nanopyramid apex as described in the previous CL measurement.

Figure 6 shows the turn on evolution of EL spectrum as the injection current increases. Data taken from green nanoparticle LED is shown here to illustrate the common behavior. At turn on, the emission starts at much longer wavelength, 600-650 nm in this case. As the current gradually increases, this emission blue shifts while the intensity only increase slightly. At the same time, another emission around 510 nm emerges. As the current increases continuously, this emission blue shifts and its intensity quickly takes over the initial longer wavelength emission peak. We model the total spectrum composed of two emission peaks and do a curve fitting calculation. The fitted two emission peaks at various injection currents are shown in Fig. 6 with dotted lines. The OM images of the evolution of emission color are also shown. We remark that such a first turn on of a longer wavelength followed by the turn on of a much stronger emission at shorter wavelength is due to the inhomogeneous In distribution in the MQW on nanopyramid facets. At the initial current injection, the MQW at the apex region is turned on first due to the lower energy bandgap. As current increases, the current fills up this region and over flows from apex to the MQW at the facet region, which emits shorter wavelength. As current increases continuously, MQW on the full facet region are all turn on. The shorter wavelength emission becomes the dominant emission peak because the MQW area is the largest. We contribute this large initial blueshift of overall emission spectrum to the spatial inhomogeneous In distribution in nanopyramid MQW. The smaller blueshift at the later stage is due to the screening of carriers of the polarization field in MQWs.

Figure 6 The turn on evolution of EL spectrum as the injection current increases. The initial much longer wavelength emission is due the turn on of the MQW at the nanopyramid apex region. The later emission at shorter wavelength is due the current over flown to the MQW at the nanopyramid facets.

In summary, we have fabricated green, yellow-green, and amber color nanoparticle LEDs, where MQWs are grown on the semipolar facets of nanopyramids. Spatially resolved CL analysis shows the redshift of emission wavelength as location moves from the bottom to apex region of nanopyramids. The electrical injection of these nanoparticle LEDs were demonstrated. The emission is fairly uniform across the entire LED chip area, despite the corrugated nanoparticle surface. The serial resistance and current leakage due to fabrication and growth defects still need to be further optimized. The electrical injection demonstration nevertheless shows the promising potential of using nanoparticle design for high In concentration LED applications.
REFERENCES