Mechanisms of enhanced light emission in GaN-based light-emitting diodes by V-shaped micropits

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Abstract: We have investigated the light emission mechanisms of V-shaped, hexagonal-micropit-arranged, InGaN/GaN light-emitting diodes (HMA-LEDs). By near-field scanning optical microscopy and the confocal scanning electroluminescence microscopy, we found that enhanced light output of HMA-LED was significantly contributed by not only improved light-extraction efficiency but also locally improved crystalline quality near the micropit. Etch pit density and cross-sectional transmission electron microscope image indicate that threading dislocation (TD) bending lead to decrease the TD density near the micropit. Furthermore, partially linearly polarized light from the inclined facets of the HMA-LEDs was observed.

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References and links


1. Introduction

GaN-based lighting technology is currently driving a lifestyle revolution in modern society because of the strong demands for energy savings and simple packaging requirements [1,2]. In particular, InGaN/GaN quantum wells have high internal quantum efficiency, even though GaN crystals contain high dislocation densities (~109 cm−2) induced by the large lattice mismatch between GaN and sapphire. However, the light output power of GaN-based LEDs is limited by several factors [3,4]. One of the main factors that suppress the light output power is the low light-extraction efficiency, which is caused by photons being trapped inside the chip because of total internal reflection at the LED surface. This photon trapping arises from the large difference between the refractive indices of GaN (n = 2.5) and air (n = 1) [5–7]. According to Snell’s law, the critical angle(θc = sin−1(nAir/nGaN)) for these structures is approximately 23.6°. To surmount this small critical angle, arrangements of V-shaped micropits, which require the use of a regrowth method, can be applied to the top of the LED [8–11]. Consequently, the light output of an LED with V-shaped micropits is increased


compared to that of a conventional LED. It is believed that this enhanced light output is predominantly caused by the improved light-extraction efficiency resulting from the enlarged photon escape cone of the LED surface [12,13]. However, the V-shaped micropits formed during the growth process can also affect the internal quantum efficiency which is significantly influenced by threading dislocation (TD) density [14–16]. Therefore, precise interpretation of the light emission mechanisms with decisive evidence is essential for further advances of high brightness LEDs. For precise interpretation, investigation of light emission mechanism of LED with micropit in terms of not only epilayer LED structure but also fully fabricated LED chip is necessary. In particular, spatially resolved optical characterization methods, such as confocal scanning electroluminescence microscopy (CSEM) or near-field scanning optical microscopy (NSOM), are appropriate for clarifying the optical characteristics of microstructures [17–23].

In this study, we report not only a reliable physical mechanism for the enhancement of the light output but also the phenomenon of quasi-polarized light emission from LEDs with V-shaped micropits by using those methods including the CSEM, the NSOM, the PL/Raman/EL spectroscopy.

2. Experimental

For the NSOM measurements, the sample was excited with a He-Cd laser (325 nm) and an Al-coated fused silica fiber (optical aperture = 100 nm) was used as the probe. Shear-force feedback was used to maintain the gap between the tip and sample, which allowed the sample topography to be recorded [24]. Light emitted from the sample was collected using an objective lens beneath the sample (illumination mode). The collected light was detected by a CCD detector and passed through a monochromator to obtain spectroscopic information. Raman scattering measurements were performed using a micro-Raman system with a He-Ne laser (633 nm). The etched pits on the sample’s surface were examined with field emission scanning electron microscopy (FE-SEM) at an acceleration voltage of 15 kV (S4300SE, Hitachi). To observe the threading dislocation (TD) bending phenomenon, TEM was used (JEOL 3010, operated at 200 kV). For the CSEM measurements, a static current of 0.2 mA was applied to the sample with a Keithley 2400s SourceMeter during scanning. A high numerical aperture (N.A. = 0.9) objective was used as a light collector for CSEM [25–27].

The microscale pattern of the sample was formed with a conventional SiO2 patterning process: the 100-nm-thick SiO2 hexagonal-shaped structures, with a diameter of 3 μm and a 6 μm gap between the structures, were deposited on a sapphire substrate using plasma-enhanced chemical vapor deposition, as illustrated in Fig. 1(a). After the micro-patterning process, the GaN-based LED structure was grown via metal-organic chemical vapor deposition. The LED structure consisted of an Si-doped, n-type, GaN layer as the electron transport layer, five pairs of InGaN/GaN multi-quantum wells (MQWs) as the active layer for light emission at 490 nm, and a Mg-doped, p-type, GaN layer as the hole transport layer. Trimethylgallium, trimethylindium, and ammonia were used as the precursors for Ga, In, and N, respectively. H2 was used as the carrier gas, except for the InGaN MQWs and GaN barrier layers, for which N2 was used. Note that GaN crystal was not directly grown on SiO2 pattern due to different crystal structure and lattice constant. A conventional LED fabrication process was then used on the epitaxially grown GaN layers. Finally, we obtained a hexagonal micropit arranged LED (HMA-LED), as illustrated in Fig. 1(b). Figure 1(c) depicts a plane-view SEM image of the fabricated HMA-LED. As can be seen, more than 200 micropits were periodically arranged on a single LED chip over an area of 315 × 315 μm. The inset presents an image of the HMA-LED under a forward bias of 0.1 mA. As described in section 1, the micropits increase the critical angle for the escape of photons from the LED, and therefore, the intensity observed from the micropits is much stronger than that from the flat surface.
3. Results and discussion

In previous studies, the improved external quantum efficiency of HMA-LEDs has been explained in terms of the improved light-extraction efficiency, which is achieved through the increased range of photon escape angles resulting from the micropits [25,28]. To determine the emission mechanisms of HMA-LEDs accurately with respect to crystalline quality, NSOM was employed in this study. NSOM is an effective tool for estimating the optical properties related to topographical characteristics because it allows both spatio-spectrally resolved PL and topographical information to be obtained simultaneously. The NSOM images were measured under atmospheric conditions at room temperature using optical fiber nanoprobe with 100nm aperture size. In our experiment, spatial resolution is somewhat larger than the aperture size owing to the photoexcited carrier diffusion which is usually occurred in illumination mode.

![Fig. 1](image1)

Fig. 1. Schematics of the (a) fabricated SiO2/sapphire template and (b) final structure of the HMA-LED. Over 200 micropits were created on the LED surface. (c) Plane-view SEM image of the HMA-LED. The inset shows the HMA-LED under a forward bias of 0.1 mA. The emission intensity near the micropits was higher than that in the flat regions.

![Fig. 2](image2)

Fig. 2. (a) Topographic image acquired near a micropit on the HMA-LED surface. (b) NSOM-PL image of the same region shown in (a). (c) Photoluminescence (PL) intensity (red dotted line) and topographic profiles (black solid line) along the white dotted lines in (a) and (b). (d) Local PL spectra measured at three different points: a flat region far from the micropit (A), near the micropit (B), and an inclined facet of the micropit (C).
Figures 2(a) and 2(b) present the surface topography and panchromatic PL mapping images of the HMA-LED epitaxial structure, respectively. The scanning area was $10 \times 10 \mu m$. The hexagonal micropit has six inclined facets, each of which is a semi-polar plane with respect to the c-plane GaN surface shown in Fig. 2(a). The depth of the investigated hexagonal micropit was approximately 6 $\mu m$. As shown in Fig. 2(b), there is intense PL emission near the micropit, whereas there is very weak PL emission inside and far from the micropit. The PL intensity of the inclined facets is higher than that at the center of the micropit because weak emission occurs from MQWs on the inclined facets, whereas PL spectrum for MQWs was not detected at the center of the micropit because of the SiO$_2$ mask.

In particular, the PL intensity of the region far from the micropit was lower than that of the region near the micropit. This could be explained by higher crystalline quality near the micropit than that far from the micropit; this higher crystalline quality can be interpreted as a lower dislocation density [29]. Figure 2(c) shows the PL intensity and topographic profiles measured along the white dotted lines in Figs. 2(a) and 2(b). These line profiles directly demonstrate the relationship between the topography and distribution of PL intensity. The PL intensity near the micropit is clearly higher than that of other regions. The strong PL intensity near the micropit dramatically decreases with increasing distance from the micropit. This is attributed to less threading dislocation (TD) density nearby micropit by bending of TDs toward micropit during lateral crystal growth as previously reported [14,30,31]. For the spectroscopic analysis, PL spectra were measured at three different positions, denoted by the letters A–C in Fig. 2(b). These positions were selected as representatives of the three different regions: A is a flat region far from the micropit, B is a flat region near the micropit, and C is an inclined facet of the micropit. The corresponding PL spectra of these positions are displayed in Fig. 2(d). Both the PL spectra measured at points A and B have a strong main peak at 486 nm and a weak peak at 360 nm.

![Fig. 3. Micro-Raman spectra measured at points A and B. The inset is a plane-view SEM image of the HMA-LED.](image)

The peaks at 486 nm and 360 nm are associated with the InGaN/GaN MQWs grown on the flat c-plane surface and the near-band-edge (NBE) transition in GaN, respectively [32,33]. However, the PL intensity of the 486 nm peak is stronger at point B than that at point A. This implies that fewer nonradiative recombination centers exist near the micropit, where the number of nonradiative recombination centers is strongly related to the crystalline quality [34–36]. At point C, a relatively strong NBE peak at 360 nm and weak peak at 420 nm are present, compared to those at points A and B. The peak at around 420 nm originates from
MQWs grown on the inclined facets. Because the thickness of the MQWs on the inclined facets is thinner than that of the MQWs on the flat $c$-plane surface, the wavelength of the emitted light is shorter for the MQWs on the inclined facets. Additionally, the low PL intensity revealed at inclined facets could be affected by non-efficient excitation of the inclined facets via NSOM probe.

As revealed in the NSOM-PL results, there is an inhomogeneous distribution of nonradiative recombination centers which are closely related to the crystalline quality [37,38]. Micro-Raman measurements were carried out to analyze the crystalline quality near to and far from the micropit. Figure 3 presents the micro-Raman spectra of the HMA-LED measured with a backscattering geometry at room temperature. The inset is a plane-view SEM image of the positions measured on the surface. For both positions, the $E_2$ (high) phonon modes were observed at 569.2 cm$^{-1}$. The full width at half maximum (FWHM) of the $E_2$ (high) phonon mode provides an indication of the crystalline quality at the measured position. The FWHMs of the $E_2$ (high) peaks measured at points A and B are 3.32 cm$^{-1}$ and 3.05 cm$^{-1}$, respectively. The lower FWHM value of the spectrum taken at point B implies an improved crystalline quality compared to position A, possibly because of a reduced TD density. This is a significant evidence of the locally improved crystalline quality of the HMA-LED.

To achieve a more reliable verification of the practical EL behavior near the micropit, HMA-LED was fabricated as explained in experimental method section. CSEM measurements, with a high spatial resolution of ~200 nm, were conducted for spatially resolved EL spectroscopy of fully fabricated HMA-LED [25]. Figure 4(a) presents a two-dimensional EL mapping image of a micropit, which was acquired using CSEM with a scan size of $10 \times 10 \mu$m along the x and y axes.

The scan shows a bright ring consisting of six yellow kernels near the micropit, with the EL intensity near the micropit relatively brighter than that in the $c$-plane flat region. Note that the reason for different light emission distribution of NSOM-PL and CSEM images is attributed to carrier density which was induced by different excitation area. For the spectroscopic analysis, EL spectra were measured at three different points: a flat surface far
from the micropit (A), near the micropit (B), and an inclined facet (C), as indicated in Fig. 4(a). Figure 4(b) shows the EL spectra measured at these points. Each peak of the EL spectra has been experimentally identified, with the peaks at 470 nm and 420 nm originating from MQWs on the flat surface and inclined facets, respectively [25]. In the EL spectrum acquired at point A, only the peak at 470 nm is evident, and it exhibits a weaker intensity compared to the same peak in the other EL spectra.

The relatively weak intensity is attributed to the small photon escape angle of the conventional c-plane surface [39,40]. In the EL spectrum acquired at point B, the main peak at 470 nm has a shoulder peak at ~420 nm. The EL spectrum measured at point C has a doublet at 420 nm and 470 nm. It should be noted that the EL intensity of the peak at 470 nm in the point B spectrum is stronger than that in the point C spectrum, although the EL intensity of the peak at 420 nm is much stronger in the point C spectrum. This indicates that the enhanced light output is the result of not only the increased light-extraction efficiency, but also the improved crystalline quality near the micropit. Moreover, it is brighter than that in the c-plane flat regions, shown in the point A of Fig. 4(a). This phenomenon is also evident in the cross-sectional CSEM image shown in Fig. 4(c); this image was obtained by scanning the x and z axes. The V-shaped bright region is consistent with the inclined facets inside the micropit. The higher EL intensity near the micropit is also apparent in this image. The EL intensity of the inclined facet region is the strongest of the scanned regions shown in Figs. 4(a) and 4(c) because the inclined facets of the micropit act as a path for photons to escape from the LED and additional photons are generated by the MQWs. In order to confirm the significance of the locally improved crystalline quality in the practical LED, we also performed injection-current-dependent EL measurements.

Figure 4(d) presents the integrated EL intensities over the full wavelength range and at 470 nm and 420 nm as a function of the injection current. The behavior of the intensity at 470 nm is relatively similar to that of the full wavelength intensity as the injection current increases. The saturation of the integrated EL intensity shown in Fig. 4(d) can be explained by the efficiency being reduced because of electron overflow and Auger recombination [41,42]. Since occupied area of MQWs on c-plane region is larger than that of MQWs on inclined facet, higher EL intensity of the MQWs on c-plane region could be expected over the whole injection current range. However, we observed unpredictable phenomena that the integrated EL intensity at 420 nm decreases as the injection current increases. This implies that total light output power of the HMA-LED was much more influenced by MQWs on c-plane than our expectation which is almost proportional to occupied area. Because locally improved crystalline quality affects EL intensity of 470 nm peak, we can expect that locally improved
crystalline quality considerably contributes to the total light output power of HMA-LED in high injection current range.

Typically, crystal defects in GaN, such as dislocations, are well known to act as nonradiative recombination centers [43,44]. Therefore, the enhanced PL intensity near the micropit could be induced by fewer crystal defects. To verify less dislocation density near the micropit, we employed the etch pit density (EPD) method; the EPD can be used to evaluate dislocations and is widely used on III-V compound semiconductors [45,46]. The etching process was performed in a mixed H₂SO₄ and H₃PO₄ solution (with a 3:1 ratio) at 275 °C for 3 min. The EPD of the sample surface was then determined using SEM, as shown in Fig. 5(a). To distinguish the TD distribution easily, the regions far from and near to the micropit are denoted by “I” and “II”, respectively. The white dotted circles indicate the etched pits in the SEM image. In Fig. 5(a), the density of etched pits in region II is lower than that of region I. To obtain further insight into the origin of the light emission and inhomogeneous intensity, the cross-sectional structure of the sample was examined with TEM. Figure 5(b) presents a cross-sectional TEM image of the HMA-LED structure. The thin TEM specimen was prepared with standard mechanical polishing and ion-milling techniques to achieve electron transparency [47]. Figure 5(b) clearly demonstrates that extended TDs (indicated by the white arrows) have propagated towards the inclined facet region, with the TDs originating from the GaN/sapphire interface. From the TEM image, we confirmed that the low dislocation density around the micropit structure is attributed to TD bending. Thus, the crystalline quality was improved near the micropit compared to the quality relatively far from the micropit. Consequently, PL intensity near the micropit shows higher than that far from micropit.

To further investigate the emission mechanism of HMA-LED, we performed polarization-angle-dependent confocal EL mapping using CSEM, because HMA-LED have multiple different crystal planes. Figure 6(a) shows a panchromatic CSEM image of a single micropit acquired without a polarizer. Six symmetrical bright ellipses, which represent the light emitted from the inclined facets, can be seen in the image. Figures 6(b)-6(d) present CSEM images acquired with a polarizer at (b) 0, (c) 60, and (d) 120 degree. Quasi-polarization-dependent emission is clearly exhibited by the micropit.

Fig. 6. Polarization-angle-dependent CSEM images of a single micropit. (a) A CSEM image acquired without a polarizer. (b-d) CSEM images acquired with a polarizer at (b) 0, (c) 60, and (d) 120 degree. Quasi-polarization-dependent emission is clearly exhibited by the micropit.
obtained using a polarizer at angles of 0, 60, and 120, respectively. As shown in these images, the pair of bright ellipses move in a clockwise direction as the polarizer rotates in the same direction. However, the light emission near the micropit is not affected by the angle of the polarizer, which indicates that the partially linearly polarized light is emitted from the inclined facets of the HMA-LED. Although, it has been theoretically and experimentally studied that the crystal plane affects the polarization ratio of light emitted from InGaN/GaN MQWs, direct visualization is still not reported [48–50]. These polarization-angle-dependent CSEM results are important as they provide direct evidence of the emission of partially linearly polarized light from semi-polar planes in HMA-LED.

4. Conclusions

We have investigated the mechanisms of enhanced light emission in GaN-based light-emitting diodes by V-shaped micropits. Through spatially resolved optical characterization, we observed the distribution of radiative recombination centers near a micropit in a HMA-LED. From NSOM, micro Raman and CSEM results, we observed that enhancement of the light output in the HMA-LED was significantly contributed by not only increased light-extraction efficiency but also improved crystalline quality near the micropit. Furthermore, quasi-polarized light emitted from inclined facets on the HMA-LED surface was observed in polarization-angle-dependent CSEM images. We believe that our results will be beneficial in understanding the emission mechanisms of microstructure appended LED structures for energy conservation and high power applications.

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