InGaN Light emitting diodes with a bandpass-filter-like GaN:Si nanoporous structures

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Abstract

InGaN-based light-emitting diode (LED) structures with GaN:Si nanoporous and air-gap structures were fabricated through a wet lateral etching (LE) process. Light output power of the LE-LED structure was enhanced 58% compared to a non-treated LED structure that had a high light extraction process on the GaN:Si nanoporous structure. Light transmittance of the nanoporous GaN:Si structure was analyzed using a photoluminescence emission light from the LED epitaxial layers acted as a reference light. The light transmittance ratios of the LE-LED were measured at 2.56 times for the blue-light region and at 0.43 times for the yellow-light region, respectively, compared to the non-treated LED structure at the lateral 35° detected angle. Optical property of the GaN:Si nanoporous structure is similar like a bandpass-filter with a 460nm center wavelength and 70nm band-width that enhances the light extraction efficiency in InGaN LEDs.

Introduction

Gallium nitride (GaN) materials have attracted considerable interest in the development of optoelectronic devices such as laser diodes and white light-emitting diodes (LEDs).[1] InGaN-based LEDs have been intensively investigated for various applications like backlights in liquid crystal displays and as solid-state lighting sources. Bright blue LEDs require an increase in their internal and external quantum efficiencies. The lower external quantum efficiency of the InGaN LED is due to a larger refractive index difference between the GaN layer and the surrounding air ($\Delta n \sim 1.5$). Bottom patterned Al_2O_3 substrates[2], top p-type GaN:Mg rough surface processes[3], selective etched nanorods in periodic microholes[4], grade-refractive-index amorphous titanium oxide films with porous structures[5], pattern-nanoporous p-type GaN:Mg surfaces[6], conical air prism arrays as an embedded reflectors[7], overcut sideholes formed by wet etching,[8] anisotropically etched GaN-sapphire interfaces[9], inclined GaN undercut structure[10], platinum nanoparticles[11], self-organized nanoscale patterning of p-type GaN[12], TiO microsphere arrays [13], and Li–Al layered-double-hydroxide platelet structures [14] have all been used to increase light-extraction efficiency in InGaN-based LEDs on Al_2O_3 substrates. Soh et al.[15] reported on the high optical performance of amber emitting quantum dots incorporated into an InGaN LED structure regrown on an UV-enhanced electrochemically etched nanoporous GaN structure. Lee et al.[16] reported the effect of gallium oxide hydroxide (GaOOH) nanorod arrays on the light extraction of InGaN LEDs. Cheah et al.[17] reported on the surface phonon polariton properties characteristic of honeycomb nanoporous GaN thin films.

In this paper, InGaN LED structure consisted of a non-etched GaN layer, a nanoporous GaN layer, and an air-gap structure fabricated through a wet-etching process on n-type GaN:Si layers with different Si-doping concentrations. Various selective wet-etching processes occurred on the n-type GaN:Si layers when varying the Si-doping concentrations. A high lateral etching rate was observed on the Si-heavy doped GaN layer with a newly-formed air-gap structure that increased the light reflectance at GaN/air-gap interface. The light extraction efficiency of the LE-LED structure was increased through the formation of nanoporous and air-gap structures close to the laser-scribing line regions. The light transmittance properties at the lateral direction of the nanoporous GaN structure were measured using the self-emission PL spectra as a reference light source. A bandpass-filter-like property between 420 to 500nm was observed in the nanoporous GaN structure that is suitable for the efficiency of high light extraction in blue LED structures. The porous formation mechanisms of the Si-doped GaN layers have already been studied. Here, the optical properties and the selective wet-etching processes of the designed InGaN LED structures are analyzed and discussed in detail.

Experiments

The InGaN LED structures were grown on a 2 in. optical-grade c-face (0001) sapphire substrate through metal-organic chemical vapor deposition (MOCVD) system. Trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia (NH3) were used as gallium (Ga), indium, and nitrogen sources material, respectively. Silan (SiH_4) and biscyclopentadienyl magnesium (CP_2Mg) were used as the n-type doping and p-type doping source, respectively. The LED epitaxial layer consisted of a 30nm-thick GaN buffer layer grown at 550°C, a 1.4μm-thick unintentionally doped GaN (u-GaN) layer (1150°C), a 0.1μm-thick Si-heavy-doped n-GaN layer (1150°C, $2\times10^{19}cm^{-3}$), a 1.2μm-thick u-GaN layer (1150°C), a 3.0μm-thick n-GaN layer (1150°C, $5\times10^{18}cm^{-3}$), nine pairs of In0.2GaN/In0.01GaN (3nm/13nm) multiple-quantum wells (MQWs, 830°C), a 0.3μm-thick p-GaN layer (950°C, $6\times10^{17}cm^{-3}$). The mesa regions of the LED structures were defined as a 1.7μm-depth through the ICP dry etching process. Subsequently, a 250nm-thick ITO film was deposited on the mesa region as a transparent conductive layer (TCL), and the Cr/Au (50nm/2500nm) metal layers were deposited as the n-type and the p-type metal contacts. Then, the LED chips were separated through the laser scribe processes by using a 355nm laser along one direction to form the channels for the lateral wet etching process. The size of LED chip was $240\times180\mu m^2$. The samples were immersed in a 0.5M oxalic acid solution with an external dc bias fixed at 15V for 20min and illuminated with a 400W Hg lamp. The schematic diagram of the lateral wet etched LED structures with a nanoporous n-GaN:Si layer and an air-gap structure were shown in Fig. 1, that was defined as the lateral-etched LED (LE-LED). The LED structure without the lateral wet etching process was defined as a standard LED (ST-LED). The geometric morphologies of the LE-LED structures were observed by optical microscopy (OM) and a field-emission scanning electron microscope (FE-SEM, JEOL 6700F). The light-intensity profiles that went across the whole LED chip were measured by a beam profiler. The light output power and the far-field radiation patterns were measured on non-encapsulated LEDs in chip form. The optical properties of the LED samples were measured through the micro-photoluminescence (μ-PL) measurement by using a 325nm HeCd laser as the excited source. The PL and the electroluminescence (EL) spectra were characterized by a monochromators (JOBIN YVON iHR550) with a TE-cooled charge coupled device (CCD) detector.

Results and discussion

SEM micrographs of the LE-LEDs are shown in Fig. 2. After the lateral wet-etching process, the nanoporous structure is observed at the n-type GaN:Si layer as shown in Fig. 2(a). The lateral-etched widths are measured at 14μm for the nanoporous structure and at 28μm for the air-gap structure, respectively, from the laser scribing (LS) line to the mesa edge region. The lateral-etched air-gap structure, with an 84μm/h etching rate, is observed below the nanoporous region caused by the high etching rate on the 0.1μm-thick Si-heavy-doped GaN:Si layer. The mesa region of the InGaN LED is defined by the plasma dry etching process. A dry-etched n-type GaN is observed around the mesa region. After the wet etching process, the 1.7μm-thick nanoporous GaN layer, the 1.2μm-thick undoped GaN layer, and the 0.1μm-thick air-gap structure are all observed in the dry-etched GaN region close to the laser-scribing line as shown in Fig. 2(b). A branch-like air-void structure is observed in the nanoporous GaN:Si layer.[18] A tilted branch-like air-void structure is observed close to the sidewall of the laser scribing region indicating that the lateral etching direction occurred from the LS line. The porosity in the GaN:Si layer can be controlled through Si-doping concentration and an external bias voltage in the oxalic acid solution. In this study, the pore diameter of the n-GaN:Si layer ($5\times10^{18}cm^{-3}$) was about 40nm under a 15V bias voltage that is similar to previously published literature.[18]

In Figs. 2(c) and 2(d), the light-intensity profiles of the LED structures were analyzed by a beam profiler at a 20mA operating current. In Fig. 2(c), the light emission intensity of the ST-LED is uniformly distributed on the mesa region. After the lateral wet etching process, a high light emission intensity of the LE-LED is observed around the mesa region that has a high light extraction process occurring on the nanoporous and the air-gap regions as shown in Fig. 2(d). In the LE-LED structure, the light emission intensity of the dry-etched GaN:Si surface, treated as a nanoporous structure close to the laser scribing line, was slightly increased. In Fig. 3(a), the μ-PL spectra of the LE-LED structures were measured at room temperature with the measured positions labeled in Fig. 1. The dominated PL peak wavelength of the InGaN active layer was measured at 468.6nm in the non-treated mesa region (position 1), at 467.1nm in the mesa region with the air-gap structure (position 2), and at 465.5nm in the mesa region with the air-gap/nanoporous structures (position 3). When the laser spot focused on the dry-etched GaN:Si surface (position 4), the strong band emission spectra of the nanoporous GaN structure was observed at 367.2nm. The PL peak wavelengths of the InGaN active layer have a slightly blue-shift phenomenon from the 468.6nm (non-treated region) to the 465.5nm (treated region) perhaps caused by the partially compressed strain release in the InGaN layer by forming the strain-release nanoporous/air-gap structures below the InGaN active layer. In Fig. 3(a), the interference signal of the PL spectrum observed at the InGaN active layer with a bottom air-gap structure (position 2) indicating a smooth etched-surface on the air-gap structure after etching and removal of the heavy Si-doped GaN:Si sacrificial layer.

In Fig. 3(b), the light output powers and the peak emission wavelengths of both LED structures are measured at room temperature. The light output power of the LE-LED structure has a 58%

enhancement compared to that of the ST-LED at a 20mA operating current. The high light output power of the LE-LED is caused by increasing the light extraction efficiency that occurs at the bottom of the air-gap/nanoporous structures around the mesa region. By increasing the injection current, the peak wavelengths of the EL spectra are blueshifted from 470.0nm (1mA) to 458.6nm (30mA) for the ST-LED structure and from 469.8nm (1mA) to 460.1 nm (30mA) for the LE-LED structure. The peak wavelength blue-shift phenomenon of the LE-LED (9.7nm) is smaller than the ST-LED (11.4nm) indicating that the compressed strain induced piezoelectric field in the InGaN active layer is slightly reduced in the LE-LED. The low external quantum efficiency of the InGaN LED is due to a larger refractive index difference between the GaN layer and the surrounding air that induces the EL emission light to be trapped in the LED chip. When the EL emission light emitted from the central LED chip is propagated to the low refractive index nanoporous structure around the LED chip, the emission light can be extracted out of the LED chip.

In Fig. 4, the PL line-scan profile of the LE-LED is measured from the laser scribing line, the lateral wet-etched region, to the non-treated mesa region across the LED chip. The PL emission intensities and the emission wavelengths (at about 468nm) of the InGaN active layer on the non-treated mesa region are almost the same. When compared to the central non-treated mesa region, a higher PL intensity and a shorter emission wavelength are observed at the lateral-etched region close to the laser scribing line. A high light extraction efficiency of the LE-LED is observed at the lateral wet-etched region. A PL wavelength blue-shift phenomenon is observed at the InGaN active layer with the bottom nanoporous structure at the lateral wet-etched region. A lattice constant of an InGaN well layer is larger than the GaN barrier layer in the InGaN/GaN MQW active structure. A large compressed strain induced piezoelectric field in the InGaN well layer caused a band-tilted effect and reduced the electron-hole recombination efficiency. The internal quantum efficiency of the treated LED chip can be slightly increased at the localized treated region by partial releasing the compress strain in the InGaN active layer. The external quantum efficiency of the LE-LED structure has been improved through the formation of the nanoporous and the air-gap structures in InGaN-based LED structures.

To analyze the light extraction property of the nanoporous structure, the PL spectra were put through angle-resolved photoluminescence measurements with a 325nm excitation laser focused and excited from the normal direction for LED chips with a 10μm-diameter spot size as shown in Fig. 5(a). From the front-side laser illumination, the PL emission peaks of both LED structures are observed at 470nm and 560nm for the blue InGaN active layer and the yellow defect level, respectively, as shown in Figs. 5(b) and 5(c). The PL spectra, were measured from 0o (lateral direction) to 35o. All angles beyond 35o are blocked by the laser focus lens as shown in Fig. 5(a). The PL emission spectra of both LED structures were analyzed at the lateral direction where the emission light had penetrated through the nanoporous structure in the LE-LED structure. In the ST-LED, the broadband emission spectra consisted of a 470nm peak for InGaN active layer and a 560nm peak for the defect level as shown in Fig. 5(b). The interference signals of the PL spectra were observed in the ST-LED structure indicating smooth surfaces at the top of the GaN:Mg/air interface and at the bottom of the GaN/sapphire interface. The Fabry–Pérot (FP) interference line-patterns spectra were observed by varying the detected angles as shown on the left-side of Fig. 5(b). The laser excited PL spectra acted as a broadband emission light source in order to analyze the optical properties of the GaN nanoporous structure at the lateral direction. From the PL detected angles, 0o to 35o, the PL emission spectra of the LE-LED were measured through the GaN nanoporous structure where the emission light from the InGaN active layer had been partially reflected at the top of the GaN/air interface and at the bottom of the GaN/sapphire interface. In the LE-LED structure, the interference signal of the InGaN active layer (at 470nm) vanished as shown in Fig. 5(c). By dividing the PL spectra of the LE-LED to the ST-LED at each detected angle, the spectra of the enhanced ratios are shown in Fig. 5(d) with the spectra and the distribution patterns of the enhanced ratios. The enhanced ratios of the InGaN peak (470nm) and the defect level emission peak (560nm) are calculated as shown in Fig.

5(d). The interference signals in the enhanced ratio spectra are observed as caused by the FP interference spectra in the ST-LED structure.

By forming a nanoporous structure around the mesa region, the enhanced ratios of the InGaN peaks increased from 1.5 to 2.5 by varying the detected angles from 0^{o} to 35^{o} as shown in Fig. 5(e) The enhanced ratio of the defect level decreased to a value of 0.5. When the laser spot focused at the central mesa region (non-treated region) with a 10μm-diameter spot size, the intensity of the PL emission light was almost the same in both LED structures. In Fig. 5(d), the higher enhanced ratios of the PL spectra were observed at the wavelength range from 420 nm to 500nm indicating that the light extraction efficiency of the LE-LED structure increased through the GaN nanoporous structure. The dimension of the GaN:Si nanoporous structure was about 40nm through the lateral wet etching process on the Si-doped GaN:Si layer. Through an angle-resolved photoluminescence measurement, a higher enhanced ratio was observed from the wavelength range of 420nm to 500nm, and the PL intensity ratio at 520nm to 700nm wavelength range was suppressed in the LE-LED structure. In the ST-LED structure, part of the emission light from the InGaN active layer was trapped due to the total reflection at GaN/air and GaN/sapphire interfaces. The light transmittance of the GaN:Si nanoporous structure was measured by using the self-emission broadband light of the PL emission spectra. High light transmittance at the wavelength range between 420nm to 500nm was observed at the GaN:Si nanoporous structure which is similar to the optical properties of a bandpass filter. The center wavelength and the band-width of the GaN nanoporous filter were measured at 460nm and 70nm, respectively, at the 35^{o} detected angle. Nanoporous GaN:Si structures have a bandpass-filter-like property that is suitable for high light extraction efficiency in blue LED structures.

Conclusion

GaN:Si nanoporous and air-gap structures were fabricated through a lateral wet-etching process on an InGaN LED structure. The porosity of the Si-doped GaN:Si layer can be controlled by varying

Si-doping concentrations in the n-type GaN layers. After the wet etching process, the n-GaN:Si layer (5×10^{18}cm^{-3}) and the Si-heavy doped GaN:Si layer (2×10^{19}cm^{-3}) were etched as a nanoporous structure and an air-gap structure, respectively. A high lateral wet-etching rate was observed in the Si-heavy doped GaN layer where the porosity of the GaN layer increased and was etched to form the air-gap structure. High light transmittance at the wavelengths ranging from 420nm to 500nm was measured at the lateral direction and it is similar to the optical properties of a band-pass filter. InGaN LEDs, with nanoporous GaN:Si structures, have a high light extraction that can be used for high efficiency nitride-based LED applications.

Acknowledgments

The authors gratefully acknowledge the financial support for this research from the National Science Council of Taiwan under grant Nos. NSC 101-2221-E-005-073, NSC 102-2622-E-005-012-CC2, and 102-2221-E-005-067.

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FIGURE CAPTIONS

Fig. 1. Schematic of the LE-LED fabrication steps consisted of the laser scribing process and the lateral wet etching process. The laser excited positions were labeled for the μ-PL measurement.

Fig. 2 The SEM micrographs of the LE-LED with (a) an lateral etched nanoporous GaN:Si region and (b) an nanoporous GaN:Si structure. The light-intensity profiles of the (c) ST-LED and (d) the LE-LED structures were measured.

Fig. 3 The μ-PL spectra of the LE-LED structures were measured that the positions were labeled in the schematic of the LE-LED. (b) The light output power and the EL peak emission wavelength of both LED structures were measured.

Fig. 4 The PL line-scan profile of the LE-LED was measured from the laser scribing line, the lateral wet etched region, and the non-treated mesa region corresponding to the inserted OM images of the LED chip.

Fig. 5 (a) Schematic of the setup for the angle-resolved PL measurements. The PL spectra of (b) ST-LED and (c) LE-LED were measured by varying the detected angle from 0o to 36o. (d) The enhanced-ratio spectra of the dividing the PL spectra of the LE-LED to the ST-LED were shown. (e) The enhanced ratio of the blue peak (InGaN/GaN MQW active layers) and the yellow peak (defect level) were calculated by varying the detected angle.

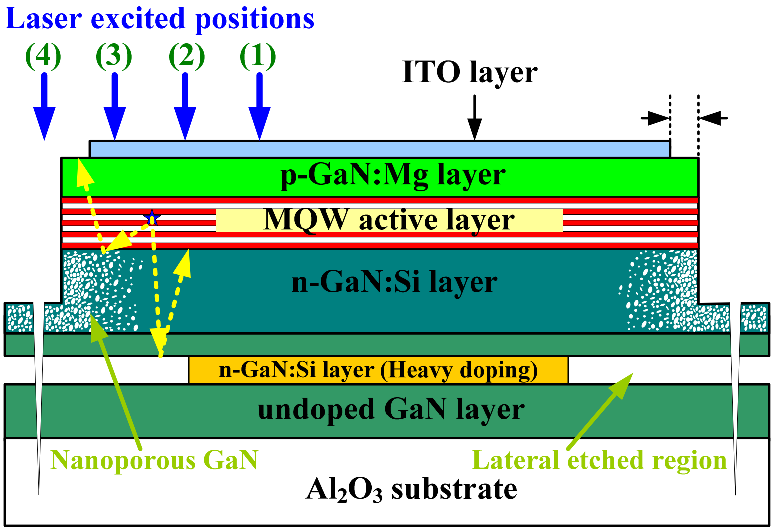

Laser excited positions
(4)
(3)
(2)
(1)
ITO layer
p-GaN:Mg layer
MQW active layer
n-GaN:Si layer
n-GaN:Si layer (Heavy doping)
undoped GaN layer
Nanoporous GaN
Lateral etched region
Al2O3 substrate

Figure 1

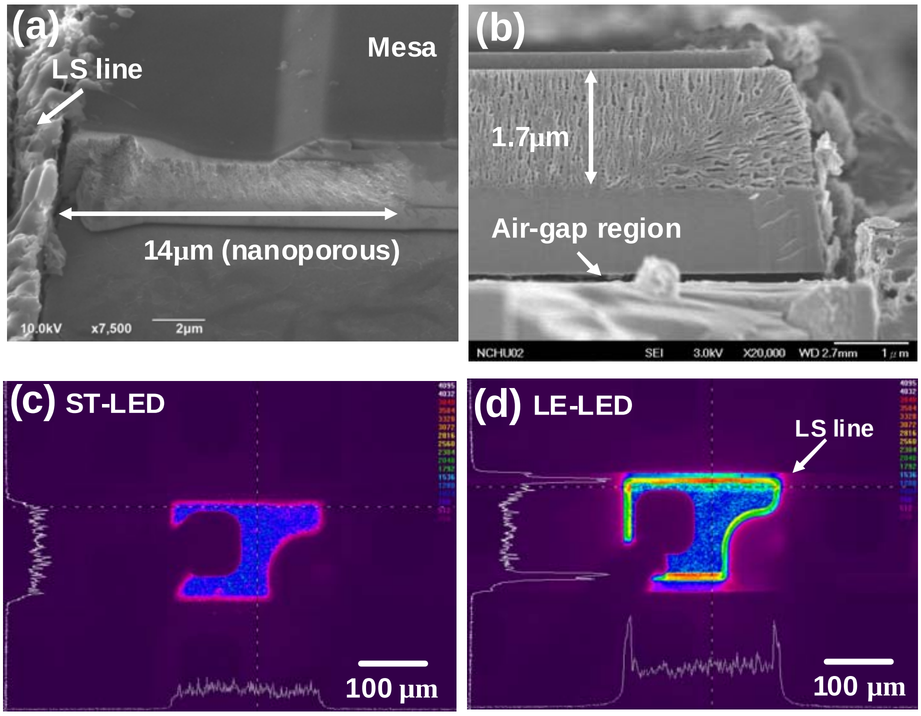

(a)
Mesa
LS line
14µm (nanoporous)
10.0kV x7,500 2µm
(b)
1.7µm
Air-gap region
NCHU02 SEI 3.0kV X20,000 WD 2.7mm 1 µm
(c) ST-LED
100 µm
(d) LE-LED
LS line
100 µm

Figure 2

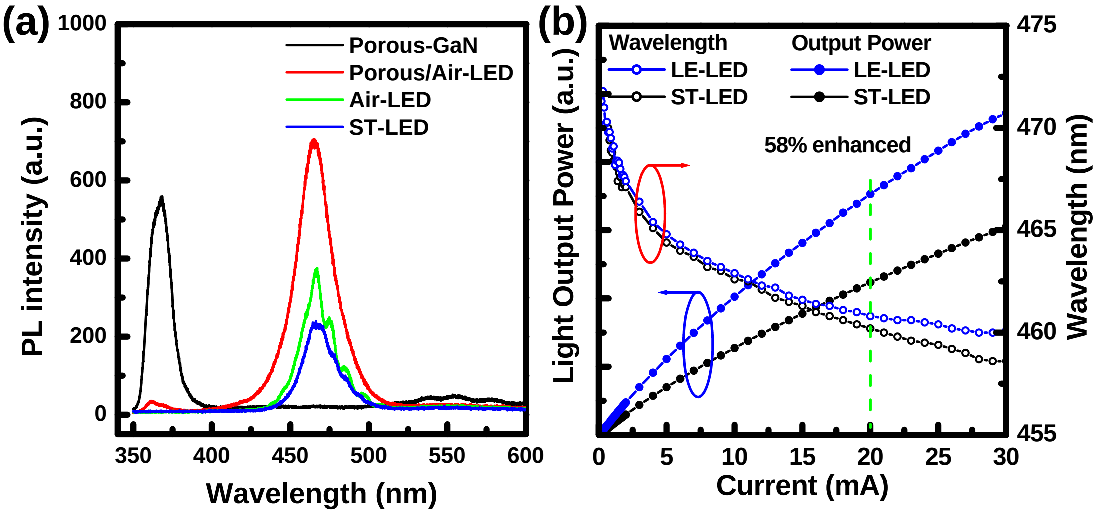
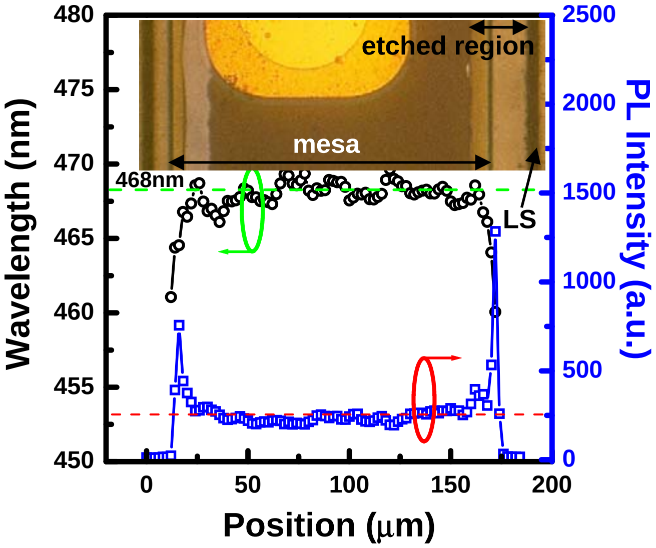

(a)
PL intensity (a.u.)
0
200
400
600
800
1000
350
400
450
500
550
600
Wavelength (nm)
Porous-GaN
Porous/Air-LED
Air-LED
ST-LED
(b)
Light Output Power (a.u.)
Wavelength (nm)
455
460
465
470
475
0
5
10
15
20
25
30
Current (mA)
Wavelength
LE-LED
ST-LED
Output Power
LE-LED
ST-LED
58% enhanced

Figure 3



480
475
470
465
460
455
450
Wavelength (nm)
2500
2000
1500
1000
500
0
PL Intensity (a.u.)
0
50
100
150
200
Position (μm)
etched region
mesa
468nm
LS

Figure 4

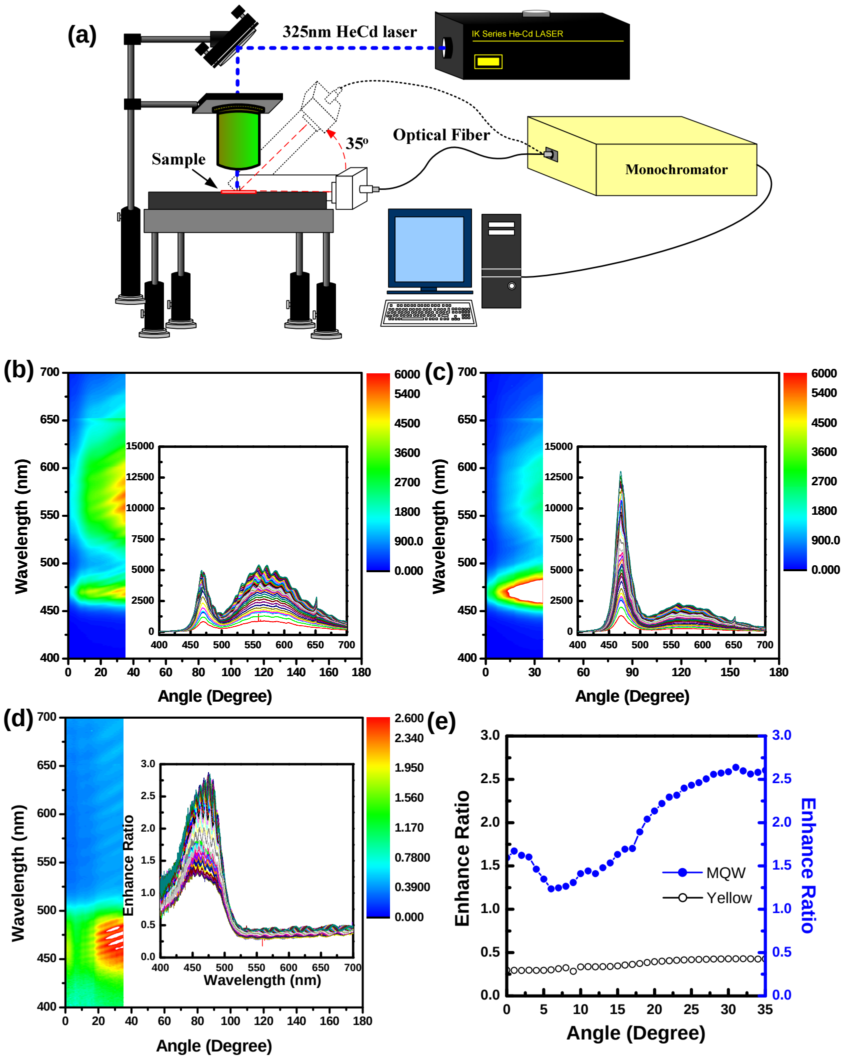

(a)
325nm HeCd laser
IK Series He-Cd LASER
Optical Fiber
35°
Sample
Monochromator
(b)
Wavelength (nm)
Angle (Degree)
(c)
Wavelength (nm)
Angle (Degree)
(d)
Wavelength (nm)
Enhance Ratio
Wavelength (nm)
Angle (Degree)
(e)
Enhance Ratio
Enhance Ratio
MQW
Yellow
Angle (Degree)

Figure 5