Enhanced optical output power by the silver localized surface plasmon coupling through side facets of micro-hole patterned InGaN/GaN light-emitting diodes

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Abstract: Light extraction efficiency of GaN-based light emitting diodes were significantly enhanced using silver nanostructures incorporated in periodic micro-hole patterned multi quantum wells (MQWs). Our results show an enhancement of 60% in the wall-plug efficiency at an injection current of 100 mA when Ag nano-particles were deposited on side facet of MQWs passivated with SiO2. This improvement can be attributed to an increase in the spontaneous emission rate through resonance coupling between localized surface plasmons in Ag nano-particles and the excitons in MQWs.

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1. Introduction

High power III-nitride light-emitting diodes (LEDs) are attractive to solid state luminaries due to their high electro-optical conversion efficiency, simple dimming control system, small size and long lifetime [1]. To date, endeavoring research have been dedicated to improving the opto-electrical performance of LEDs through the use of more efficient radiative recombination and light extraction structures. The internal quantum efficiency (IQE) improvement is nearly saturated due to various attempts to develop high quality III-nitride epitaxial growth techniques and effective energy bandgap engineering of LED structures. Also, the external quantum efficiency (EQE) factors enabling high optical output power have been improved in various light extraction structures and low thermal resistance with heat sink system of LEDs. Many interesting approaches have been attempted to accomplish the highest quantum efficiency of LEDs, such as modified multi quantum wells (MQWs) [2], reduced polarization field effects and electron blocking layer for high IQE [3], thin light-emitting layers with surface texturing and a nanostructure embedded emitting surface for enhancing the light extraction efficiency (LEE) [4,5]. In recent years, many investigations exploit the wave nature of light, such as photonic crystals to enhance light extraction [6,7], and output coupling through surface plasmons excited at metal nanostructures [8–10].

In particular, the most notable phenomenon occurring with the metal nano-structures on insulators is electromagnetic resonances due to collective charge oscillations of the metal’s free electrons, termed localized surface plasmons (LSPs) and surface plasmon polaritons (SPPs). These plasmonic features have attracted interest for the enhancement of quantum efficiency of LEDs. It has been reported that LSPs coupled with InGaN MQWs of nitride based LEDs enhance both the photoluminescence (PL) intensity and the spontaneous emission rate through the efficient and rapid resonance with excitonic oscillation energy in MQWs. In addition, many previous works presented the Purcell effect [10], and dispersion
diagram of LSPs for cases related to the relationship between metal nano-films or nanoparticles such as Ag, Au, Al, Pt, Pd, Cu, and Ru and various dielectric materials such as SiO$_2$ [11], GaN [12], and AlGaN [13]. In general applications of LEDs, the plasmon energy of Ag (~3 eV), Au (~2.3 eV), and Al (~5 eV) thin film forms of metal/GaN interface is suitable for surface plasmon coupling to blue, green, and ultraviolet emission associated with MQWs, respectively [8]. Also, various metal nano-particles has attracted much interest with enhancement of light output power of LEDs [14]. The plasmon coupled blue and green LEDs have been successfully demonstrated using Ag or Au metal thin film [15], nano patterned nano-particles with dielectric gratings [16], nano-hole arrays [17], and embedded metal nanoparticles in an n- or p-GaN cladding layer [18,19]. However, the major disadvantage of LSPs coupled LEDs is the resonance coupling enhancement whereby the self-sustainability of surface plasmon is strongly dependent on the penetration depth of the LSPs into MQWs. The penetration depth was estimated to be a few tens of nanometers for GaN-based LEDs [8,19]. Accordingly, the thickness of the p-GaN layer should be less than the penetration depth of an LSP evanescent field, but such thickness is too thin for the operation of a p-n junction of LED.

In this work, to further enhance the extraction efficiency of GaN-based blue LEDs, micro-holes ($\mu$-holes) are fabricated in LEDs which passes through MQWs. Moreover, Ag nanoparticles are deposited on the side facet of MQWs passivated by SiO$_2$ to enhance the light output power by the LSPs coupling effects with InGaN/GaN MQWs.

2. Experiment

Fig. 1. Schematic diagram and Nomarski microscope images of Ag surface plasmon embedded periodic $\mu$-holes LEDs. The $\mu$-holes passed through MQWs and Ag nanoparticles are deposited on the nearest side facet of MQWs with SiO$_2$ passivation. The motive principles are the promotion of coupling between edge-emitting excitons and Ag LSPs. In addition, the Ag nanowires were deposited on ITO contact layer to create a connecting network over the $\mu$-holes.

The epitaxial structure of the original LED wafers grown on a c-oriented sapphire substrate by metalorganic chemical vapor deposition (MOCVD) is composed of a low temperature GaN nucleation layer, a 2 $\mu$m thick Si-doped n-type GaN epilayer grown at 1100 °C, five pairs of InGaN/GaN MQWs with 3 nm thick well and 10 nm thick barrier grown at 800 °C, and a 200 nm thick Mg-doped p-type GaN epilayer. During the MOCVD growth, trimethylgallium, trimethylindium, and NH$_3$ were used as precursors for Ga, In, and N, respectively. Hydrogen was used as the carrier gas, except for the growth of InGaN/GaN MQWs, where nitrogen gas was used. The LED wafers were then thermally annealed at 850 °C for 3 min in nitrogen in a rapid thermal-annealing furnace to activate Mg acceptors. The In composition in InGaN/GaN MQWs region was optimized for blue emission. The high power
LED chips with 500 μm × 500 μm effective emission top area were fabricated as shown in schematic diagram in Fig. 1. Firstly, the LED designed for mesa structure was dry-etched using a standard photolithography process and inductively coupled plasma (ICP) etched with Cl₂ and BCl₃ reaction gases to obtain the n-type GaN contact layer. A 200 nm thick indium thin oxide (ITO) transparent conducting layer was deposited on p-GaN emitting layers for the Ohmic contact and current spreading using the dielectric sputtering system from an amorphous ITO target source. Secondly, the photoresist patterns which consist of 10 μm diameter open circles with a hexagonal periodicity were formed on top of ITO layer, except for p-type electrode region. The ITO films were wet-etched using a diluted HCl solution. Subsequently, the μ-holes were pierced through p-GaN and MQWs to the n-GaN layer using chlorine-based ICP dry etching process.

![Figure 2](image)

**Fig. 2.** Top view scanning electron microscope (SEM) images of (a) Ag nano-wires and (b) nano-particles. (c) SEM images of periodic μ-holes patterned LEDs and Ag nano-particle deposited μ-holes area. (d) High magnification SEM images of the side facet of μ-holes with Ag nano-particles and Ag nano-wire bridge networks.

The μ-holes were formed as a bowl shape due to the undercut effects during the wet and dry etching process. Then, the μ-holes formed templates were loaded on plasma-enhanced chemical vapor deposition (PECVD) chamber and approximatively ~50 nm thick SiO₂ layer was deposited on inner-side facets of μ-holes for the electrical isolation and passivation of surface imperfections. To realize LSP-enhanced LEDs, the thickness of SiO₂ layer is critical for the coupling of excitons of emitter and LSPs of metal nanoparticles. The penetration depth of the LSPs field into the SiO₂ passivation layer is given by \( Z = \frac{\lambda}{2\pi(\varepsilon'_{\text{SiO}_2} - \varepsilon'_{\text{metal}})} \), where \( \varepsilon'_{\text{SiO}_2} \) and \( \varepsilon'_{\text{metal}} \) are the real part of the dielectric constant of the SiO₂ and metal. However, in real-complex system such as various multilayer structures and various sizes and shapes of metal nano-particles, the penetration depth profile of LSPs field distribution is complicated. Accordingly, we refer to the previous works done by Jang et.al, which give the analysis of the LSP resonance of Ag/SiO₂ nano-particles by the full three dimensional finite-difference time-domain simulations [20].

Finally, the Ag nano-wires and Ag nano-particles, shown the scanning electron microscope (SEM) images in Figs. 2(a) and 2(b), were sprinkled on μ-holes patterned emission area. Figures 2(c) and 2(d) show the top view SEM images of periodic μ-holes array.
patterned LEDs with Ag nanoparticles and nano-wires. Ag nano-particles were randomly deposited on the inner bowl surface covered by SiO$_2$ dielectric insulating layer on the side facet of LED; it is expected that some of Ag nano-particles are attached to the near distance to InGaN quantum wells. Additionally, the Ag nano-wires were deposited on ITO contact layer and formed a bridge network over the $\mu$-holes.

3. Results and discussions

Figures 3(a) and 3(b) show the excitation power dependent photoluminescence (PDPL) spectra of $\mu$-hole array structures with [Fig. 3(b)] and without [Fig. 3(a)] sprayed Ag nano-particles, respectively. The PDPL spectra were measured from the top side of samples using He–Cd laser ($\lambda = 325$ nm) with an excitation laser power ranging from 4.5 mW to 45 mW for a spot diameter of about 30 $\mu$m which covers 2 or 3 $\mu$-holes.

![Excitation power dependent PL spectra of $\mu$-holes patterned LED (a) without and (b) with Ag LSPs. The abnormal peaks in $\sim$450 nm were caused by grating change of PL instrument. (c) Plots of integrated PL intensity and (d) dominant maximum PL peak energy depending on excitation power.](image)

Fig. 3. Excitation power dependent PL spectra of $\mu$-holes patterned LED (a) without and (b) with Ag LSPs. The abnormal peaks in $\sim$450 nm were caused by grating change of PL instrument. (c) Plots of integrated PL intensity and (d) dominant maximum PL peak energy depending on excitation power.

It is noted that the PL spectra of Ag LSPs coupled LED show the distinguishable peak shift towards long wavelengths by 28.8 meV as shown in Fig. 3(b). The manifested PL peak energy is associated with the LSP energy of Ag nano-particles which is slightly lower than the exciton energy of bare MQWs. When the LSP mode is coupled to excitons in MQWs, the PL from Ag LSPs coupled exciton in $\mu$-hole LED is enhanced resulting in the peak shift towards low energy. The integrated photoluminescence (PL) intensity illustrated in Fig. 3(c) shows that the Ag nano-particles lead to the low threshold and a strong increase in PL intensity with excitation power. Also, the PL intensity of $\mu$-holes MQWs with Ag nanoparticles does not show the saturation even at a high injection level. Possible factors influencing the emission improvement of Ag nano-particles incorporated MQWs are the generation of LSPs at SiO$_2$/Ag interface and coupling effect with excitons in MQWs. The LSPs’ strong localized electrical fields increase the quasi-radiative exciton recombination and the spontaneous emission efficiency. In particular, the emission enhancement mechanisms involved in LSPs show that the exciton energy is non-radiatively coupled to LSPs before radiative scattering through the surface SPPs into far-field [8,10]. This radiative scattering path through the coupling with LSPs occurs much faster than other non-radiative phonon
scattering due to the closer extinction energy and strong localized fields of LSPs. Figure 3(d) illustrates the dominant maximum PL peak energy as a function of excitation power. The PL peak energies of bare μ-holes LED without Ag nano-particles shifted around ~18 meV toward higher energy when the excitation power increase from 4.5 to 45 meV, while the dominant peak energy of Ag LSPs coupled μ-holes LED does not. The blue shift in the PL peak position with increasing excitation power was often observed in compositionally inhomogeneous InGaN/GaN MQW where the high injection carriers may cause the band-filling effect and the screening of the piezoelectric field [21–23]. However, in the case of Ag LSPs coupling modes with MQWs, the peak energy does not change with excitation power, showing a characteristics of a coupled mode.

The local emission distribution of the fabricated LED has been investigated by confocal laser scanning microscope (CLSM). Figure 4 shows the CLSM images of μ-holes LEDs excited by the semiconductor diode laser (λ = 405 nm) and utilized a band filter to collect the light from 450 nm to 480 nm wavelength. As shown in Fig. 4(a), the μ-hole patterned LED structure exhibits the light output power enhancement effect due to the light escape cone from side walls. Even though μ-hole effects on EQE of LEDs were not focused in this work, the light output power of μ-holes LEDs is enhanced by 15% at an injection current of 80 mA compared with conventional planar LEDs as reported in previous work [24]. However, spatial light emission distribution of μ-holes LEDs with Ag-LSPs shows the brighter power intensity on active regions and darker at μ-holes region in confront to μ-holes LEDs without Ag-LSPs as shown CLSM images in Fig. 4(b). This discrepancy of luminescence spatial distribution was possibly attributed to the Ag LSPs induced the EQE due to the evanescent nature of the LSPs field parallel to the exciton oscillation field of TE polarization, which was confined in narrow quantum well. In fact, for the case of compressive-strained InGaN/GaN MQW based LEDs, the polarization resolves emission power ratio \( P = (I_{TE} - I_{TM})/(I_{TE} + I_{TM}) \) shows high in edge emission as compared to top-surface emission [25]. Consequently, the generated photons effectively extract to the free space to enhance LEE. Also, the far-field emission directions can change the escaping light angles according to the SPPs directions through the adjacent Ag nano-particles on the side walls.

We successfully demonstrated the Ag LSPs enhanced blue emission LEDs which contains in μ-holes array structure obtained through a viable fabrication process. The Ag plasmon enhanced μ-holes LEDs were successfully operated with electrical injection current up to 200 mA. The electrical operation characteristics and the electroluminescent (EL) output power of the μ-holes LEDs with and without Ag LSPs are measured on-chip probing by a current-voltage (I-V) tracer and optoelectronic photodiode. The EL output powers and operation voltages of LEDs both with and without Ag LSPs as a function of injection current are shown in Fig. 5(a). The light output power, in particular, showed strong enhancement in the Ag nano-particles incorporated LEDs. EL performance at an injection current of 150 mA of the
Ag LSPs coupled LEDs was about 60% higher than that of LED without Ag nano-particles. Also, the degradation begins at higher injection current levels in LEDs with Ag LSPs as can be seen in Fig. 5(a). These EL enhancements can be explained by the polarization matched TE mode extraction through side edge facet and faster radiative resonance coupling effect between the strong localized electric field in Ag LSPs and excitons in MQWs, similar to the previously discussed PL enhancement mechanism. The forward operation voltage with and without Ag LSPs coupling was 4.1 V and 4.4 V, respectively, at an injection current of 150 mA. The I-V curve slope mostly depends on the parasitic serial resistance ($R_s$) of LED. In fact, the honeycomb shaped structures formed by $\mu$-holes patterning, results in the reduction of the surface current spreading. However, adding Ag nano-wires played a role of the connecting network over $\mu$-holes to provide better conduction electrons path and to thus improve the current spreading. It is expected that the slope of I-V curve increases due to decreasing $R_s$ of LED.

The wall-plug efficiency (WPE), $\eta_{WPE}$, which is the ratio of the optical output power, $P_{optical}$, to the input electrical power, i.e., $\eta_{WPE} = P_{optical}/(IV)$, can be viewed as an overall figure of merit of LEDs. Figure 5(b) shows the WPE of LEDs with and without Ag LSPs. The optical output power was measured roughly using on-chip probing system. The $\eta_{WPE}$ increases with injection current up to ~60 mA and then drop rapidly with further increase in the injection current. Even though the maximum EL intensity value is obtained at ~150 mA, we evaluated the typical WPE droop value at 100 mA before showing the dominant thermal-burning decay at high current. WPE droop for Ag LSPs is 7.2%, approximately half of the value, 15.4% for LED without Ag LSPs estimated at 100 mA. The origin of efficiency droop phenomenon has been debated including a crystal imperfections in active region such as threading dislocations, Auger scattering, electron overflow at high injection level, and QCSE; these carrier recombination processes lead to the increase of the non-radiative recombination rate due to much faster decay time than radiative carrier recombination. However, Ag LSPs incorporated LEDs reduce the efficiency droop through the fastest recombination path with LSPs coupling. Therefore, the LSPs efficiency enhancement factor ($F_{WPE}$) is dependent on the probability of photon extraction rate through Ag LSPs coupling with excitons in MQWs. It can be estimated from wall-plug efficiency ratio of LEDs with and without Ag LSPs, $F_{WPE}=(\eta_{with}/\eta_{without})$. Despite the reduction in the light output power, the $F_{WPE}$ value increase at a high injection current as shown in Fig. 5(b). This increasing $F_{WPE}$ values suggest that carrier density in active layer reduces quickly and prevents the band filling and electron overflows.
4. Conclusion

In conclusion, we demonstrated and characterized the Ag LSPs enhanced blue emission LEDs with μ-holes array structures based on InGaN/GaN MQWs. The LSPs of Ag on side facet of MQWs exhibited a strong WPE enhancement due to the evanescent nature of the LSPs field parallel to the exciton oscillation field. This improvement can be attributed to an increase in the spontaneous emission rate through resonance coupling between localized surface plasmons in Ag nanoparticles and the excitons in MQWs. Our results showed the feasibility of the high quantum efficiency of GaN related LEDs using Ag plasmon-assisted emission enhancement.

Acknowledgments

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