Experimental observation of electroluminescence enhancement on green LEDs mediated by surface plasmons

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Abstract: We experimentally demonstrate the 1.5-fold enhancement of the electroluminescence (EL) of surface-plasmon (SP)-mediated green LEDs. On the p-clad surface of InGaN/GaN multi-quantum well LEDs, a 2-dimensional, second-order grating structure is textured and coated with an Ag electrode. With this setup, a larger EL enhancement factor is obtained at a higher injected current, which suggests that SP-LEDs can be a possible solution to efficiency droop, which is one of the main problems in developing high-power LEDs. Details regarding the implementation of our device are discussed.

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References and links

- M. R. Krames, O. B. Shchekin, R. Mueller-Mach, G. O. Mueller, L. Zhou, G. Harbers, and M. G. Craford, "Status and future of high-power light-emitting diodes for solid-state lighting," J. Disp. Tech. 3(2), 160–175 (2007).
- T. Fujii, Y. Gao, R. Sharma, E. L. Hu, S. P. Denbaars, and S. Nakamura, "Increase in the extraction efficiency of GaN-based light-emitting diodes via surface roughening," Appl. Phys. Lett. 84(6), 855–857 (2004).
- I. Schnitzer, E. Yablonovitch, C. Caneau, T. J. Gmitter, and A. Scherer, "30% external quantum efficiency from surface textured, thin-film light-emitting diodes," Appl. Phys. Lett. 63(16), 2174–2176 (1993).
- T. Baba, R. Watanabe, K. Asano, F. Koyama, and K. Iga, "Theoretical and experimental estimations of photon recycling effect in light emitting devices with a metal mirror," Jpn. J. Appl. Phys. 35(Part 1, No. 1A), 97–100 (1996).
- M. Boroditsky, T. F. Krauss, R. Coccioli, R. Vrijen, R. Bhat, and E. Yablonovitch, "Light extraction from optically pumped light-emitting diode by thin-slab photonic crystals," Appl. Phys. Lett. 75(8), 1036–1038 (1999).
- S. P. DenBaars, D. Feezell, K. Kelchner, S. Pimputkar, C.-C. Pan, C.-C. Yen, S. Tanaka, Y. Zhao, N. Pfaff, R. Farrell, M. Iza, S. Keller, U. Mishra, J. S. Speck, and S. Nakamura, "Development of gallium-nitride-based light-emitting diodes (LEDs) and laser diodes for energy-efficient lighting and displays," Acta Mater. 61(3), 945–951 (2013).
- A. Kock, E. Gornik, M. Hauser, and W. Beinstingl, "Strongly directional emission from AlGaAs/GaAs light-emitting diodes," Appl. Phys. Lett. 57(22), 2327–2329 (1990).
- I. Gontijo, M. Boroditsky, E. Yablonovitch, S. Keller, U. K. Mishra, and S. P. DenBaars, "Coupling of InGaN quantum-well photoluminescence to silver surface plasmons," Phys. Rev. B 60(16), 11564–11567 (1999).
- K. Okamoto, I. Niki, A. Shvartser, Y. Narukawa, T. Mukai, and A. Scherer, "Surface-plasmon-enhanced light emitters based on InGaN quantum wells," Nat. Mater. 3(9), 601–605 (2004).
 H.-W. Huang, C. C. Kao, J. T. Chu, H. C. Kuo, S. C. Wang, and C. C. Yu, "Improvement of InGaN–GaN light-
- H.-W. Huang, C. C. Kao, J. T. Chu, H. C. Kuo, S. C. Wang, and C. C. Yu, "Improvement of InGaN–GaN lightemitting diode performance with a nano-roughened p-GaN surface," IEEE Photon. Technol. Lett. 17(5), 983– 985 (2005).
- J. Yoon, S. H. Song, and J.-H. Kim, "Extraction efficiency of highly confined surface plasmon-polaritons to farfield radiation: an upper limit," Opt. Express 16(2), 1269–1279 (2008).
- M.-K. Kwon, J.-Y. Kim, B.-H. Kim, I.-K. Park, C.-Y. Cho, C.-C. Byeon, and S.-J. Park, "Surface-plasmonenhanced light-emitting diodes," Adv. Mater. 20(7), 1253–1257 (2008).
- H. Zhang, J. Zhu, Z. Zhu, Y. Jin, Q. Li, and G. Jin, "Surface-plasmon-enhanced GaN-LED based on a multilayered M-shaped nano-grating," Opt. Express 21(11), 13492–13501 (2013).
 J. Henson, E. Dimakis, J. DiMaria, R. Li, S. Minissale, L. Dal Negro, T. D. Moustakas, and R. Paiella,
- J. Henson, E. Dimakis, J. DiMaria, R. Li, S. Minissale, L. Dal Negro, T. D. Moustakas, and R. Paiella, "Enhanced near-green light emission from InGaN quantum wells by use of tunable plasmonic resonances in silver nanoparticle arrays," Opt. Express 18(20), 21322–21329 (2010).

 K. G. Lee, K. Y. Choi, and S. H. Song, "Optimal design of green InGaN/GaN LEDs mediated by surfaceplasmon gratings," submitted.

1. Introduction

Due to their small size, low power consumption, and environmental friendliness, lightemitting diodes (LEDs) have become one of the most widely used light sources [1]. The luminescent efficiencies of the currently available LEDs, however, still need to be improved further. The performance of LEDs in the green spectral range is particularly limited in the visible spectral range, in contrast to the rapid improvement in blue LEDs during the last few decades. In discussing the total luminescent efficiency of green LEDs, the following two major factors need to be considered. The first is the chip extraction efficiency (EE). Most materials for LEDs have high refractive indices, assigning wide total internal reflection angle ranges on device-air interfaces. Therefore, most of the photons generated inside the system cannot escape out to be lost as heat. As an example, the EE of a bare LED chip is limited to a few percent [2]. To circumvent this problem, various strategies, including the implementation of surface texturing [2,3], photon recycling [4], and photonic crystals [5], have been demonstrated. The second factor is the internal quantum efficiency (IQE), which is defined as the conversion ratio of electron-hole pairs into photons. The IQE of the currently available blue LEDs has been improved up to \sim 80%; however, it is still in the range of \sim 30% for green LEDs, which seriously degrades the performance [6].

Since the first report regarding surface-plasmon (SP) mediated LEDs [7], there have been many theoretical and experimental studies for realizing SP-LEDs with sufficiently high efficiency for practical usage [8–14]. An additional decay channel of the electron-hole recombination into the SPs supported by the metallic structure near the active layer can enhance the IQE. The EE may also be improved due to the modified radiation pattern given by the scattering characteristics of the SPs in the metallic structure. In 2008, the electroluminescence (EL) of blue LEDs mediated by SPs was firstly demonstrated [12]; however, there are still many challenges in realizing *practically* effective SP-LEDs, especially in the green spectral region. Because the IQE of green LEDs is just ~30%, a large enhancement factor can be expected. In addition, the propagation length of the SPs is longer in green wavelengths than in blue, so more directional emissions can be achieved, resulting in a large EE value [15].

InGaN/GaN LEDs mediated by SPs have been intensively studied for generating efficient LEDs in short spectral ranges (blue to green), owing to the coincidentally well-matched band-gap energy ($\hbar\omega_{BG}$) of the blue InGaN quantum well (QW) of the ~460 nm emission wavelength and the SP energy ($\hbar\omega_{SP}$) at the GaN-Ag interface ~3.0 eV, which corresponds to a 410 nm wavelength [9]. Moreover, Ag is a preferred metal for flip-chip type LEDs due to its excellent reflectance in the visible spectrum and good ohmic contact to p-GaN.

However, there are several physical and technical difficulties in the experimental demonstration of efficient SP-LEDs. First of all, surface plasmon is a very lossy wave. Unless the plasmon-to-photon out-coupling efficiency is substantially larger than the original internal quantum efficiency, the efficient energy transfer to the surface plasmon only ends up decreasing the internal quantum efficiency of the device. A high Purcell factor can be achieved near the cutoff frequency of the surface plasmon due to the large photonic density of states, but it comes at the cost of large propagation loss and increasing fabrication difficulties. In such a device, no grating can be effective enough to scatter the surface plasmon into radiation mode before it decays out. Secondly, the active layer must be located within reach of the surface plasmon mode field, which is tightly localized along the semiconductor-metal interface and extends only up to a few tens of nanometers into the GaN layer, depending on the frequency. Therefore, a significant compromise in the epitaxial layer structure becomes inevitable. Usually, InGaN/GaN multi-QW LEDs grown using a state-of-the-art metal-organic-chemical-vapor-deposition (MOCVD) technique suffer from large leakage current as

G. Sun, J. B. Khurgin, and R. A. Soref, "Practicable enhancement of spontaneous emission using surface plasmons," Appl. Phys. Lett. 90(11), 111107 (2007).

long as the p-clad layer is thinner than 30 nm. One may suggest placing the GaN-Ag interface on the n-side of the LED, but there has been no successful attempt to invert the order of the epitaxial growth of GaN-based LEDs. Backside-polished thin film LEDs render access to the n-type surface, but then again, no precision polishing will guarantee the exact thickness of the remaining n-clad layer. Thirdly, direct patterning of the p-GaN surface using any dry etching method will damage the p-GaN layer and/or the underlying active layer, resulting in a significant increase of either the forward bias voltage or the leakage current or both. It is known that dry etching of p-GaN induces a damage layer in which nitrogen vacancies are created. Those nitrogen vacancies act as donors to convert the p-type layer into n-type, resulting in complete destruction of the p-n junction structure of the LED.

In this work, we have carefully considered those issues in our system design and fabrication, and experimentally demonstrate for the first time the EL enhancement of green SP-LEDs. We observed a 1.5-fold enhancement of the EL from a green InGaN/GaN multi-QW LED for which the p-clad surface was periodically textured and coated with a two-dimensional Ag pillar array. The increased electron-hole recombination rates were verified using time-resolved photo-luminescence (TRPL) measurements. A larger improvement in the wall-plug efficiency (WPE) at a higher injected current is observed, which suggests that the SP-LEDs can be a solution to the problem of efficiency droop, the phenomenon of decreasing LED efficiency at higher working powers.



Fig. 1. A schematic of flip-chip reflection type InGaN QW LEDs mediated by SPs.

2. System design and fabrication

Figure 1 shows the schematic of a flip-chip reflection type InGaN QW LED mediated by SPs formed in a 2-dimensional silver pillar-shaped grating structure. The inset shows the structure parameters of period Λ , pillar diameter D, pillar height h, and separation gap s. The separation gap s between the top of the silver pillar and the multi-QWs is maintained at 30 nm throughout all of the calculations and experiments in this study. By using MOCVD, epitaxial layers of n-GaN, InGaN multi-OWs, and p-GaN (110 nm thick) were sequentially deposited on a sapphire substrate. A 2-dimensional pillar grating structure was made by evaporating a silver layer on a pre-patterned p-GaN layer by using holographic lithography (HeCd laser, $\lambda_{\text{center}} = 325 \text{ nm}, 50 \text{ mW}$). Other size parameters of the grating structure were optimized by a 3-dimensional finite-difference time-domain (FDTD) calculation to maximize the performance of the SP-LEDs. From the calculations, the SP-mode wavelengths at the frequencies of vacuum wavelengths 465 nm and 527 nm are calculated as 72 nm and 125 nm, respectively. Fabricating gratings with such short periods using holographic lithography is technically challenging, so we have considered only the 2nd order grating with $\Lambda = 250$ nm for the green wavelength. At $\lambda = 527$ nm, the propagation length of the SPs is calculated as 212 nm, which is shorter than the 2nd order grating period (250 nm). In this case, the

relaxation of the electron-hole pair in the QWs couples efficiently only to the nearest Ag pillar, and any couplings to farther pillars become negligible. In this situation, the resonance conditions of the SPs are predominantly determined by the individual pillar geometry rather than by the periodicity of the gratings.

Figure 2(a) shows the dependency of the Purcell factor on *D* of the silver pillars with $\Lambda = 250$ nm and h = 80 nm at $\lambda = 527$ nm. The separation gap *s* is 30 nm. In this calculation, an oscillating dipole is simulated on its radiating power, which is normalized to the reference case: an Ag layer is deposited on an unpatterned p-GaN layer with 110 nm thickness. The Purcell factor shows oscillatory behavior as the pillar diameter changes, and the maximum was found at D = 170 nm. The reflectance of the grating is also considered for optimizing *h* as shown in Fig. 2(b). For the 1st order grating (black curve), the reflectance has a peak value of 25% at h = 40 nm before decaying when h > 40 nm. For the 2nd order grating (red curve), on the other hand, the reflectance is 13% which is almost constant for the range of 40 nm < h < 100 nm. A larger Purcell factor is expected for a smaller *s*; however, due to the problem of the leakage current shortly mentioned above, we could not decrease the separation *s* below 30 nm in the experiment. Therefore, finally we have chosen the structure parameters of h = 80 nm and s = 30 nm with the 110 nm thickness of the p-GaN before Ag patterning.



Fig. 2. (a) Purcell enhancement factors as a function of the diameter of the Ag pillar. (b) Reflectance of the Ag-pillar-grating as a function of the grating depth.

The quality of the samples can be checked using tunneling electron microscope (TEM) and scanning electron microscope (SEM) images. Figure 3(a) shows a side view of the MQW (5 layers and 110 nm thick in total), and Fig. 3(b) is an SEM image of a grating structure etched on the p-GaN layer before the deposition of the Ag layer. The grating dimensions are $\Lambda = 250$ nm, D = 170 nm, and h = 80 nm. The hexagonal shapes of the pillars are due to the crystal structure of GaN. Excellent adhesion with good ohmic contact at the p-GaN/Ag interface can be achieved as shown in Fig. 3(c).



Fig. 3. (a) A TEM image of InGaN MQW (side view). SEM images of grating structure etched on p-GaN layer (b), and a side view of the Ag layer evaporated on p-GaN layer (c).

3. Measurements and results

We begin our analysis by checking the working spectral ranges of the SP-LEDs from photoluminescence (PL) measurements. As shown in Fig. 4(a), the emission peak is 514.2 nm and the full-width-half-maximum is 32.6 nm, demonstrating the working wavelengths in green. The PL intensity of the SP-LED was compared to the reference case, for which an Ag layer deposited on an unpatterned p-GaN layer. For the reference, the coupling of the QW into the SPs is expected to be negligible because the p-GaN is suitably thick (110 nm). At the center wavelength ($\lambda = 514$ nm) region, the PL intensity enhancement factor was about 1.2. This PL intensity enhancement may be caused by SP-QW coupling between the metallic structure and the active QW layers. To confirm this, we measured the PL decay times at low and room temperatures by using time resolved photo-luminescence (TRPL) measurements.



Fig. 4. (a) PL intensity spectra. (b) PL decay lifetimes measured at low (17 K) and room (300 K) temperatures. Data of the SP-LED and the reference are shown as red and black opencircles, respectively, and the solid lines are the fitting.

In Fig. 4(b), the fitting curves (solid lines) show double exponential decays. The fast decay comes from exciton recombination and the slow decay from localized carrier recombination as described in ref. 12. The fast decay lifetimes are measured as 6.25 ns (SP-LED) and 8.33 ns (reference) at 17 K, and as 3.23 ns (SP-LED) and 3.45 ns (reference) at room temperature. The total decay rates, the inverses of the decay lifetime, are enhanced 1.33 times at low temperature (17 K), and 1.07 times at room temperature. The enhancement of the total decay rate can be written as: $\gamma_{enhancement} = (\gamma_r + \gamma_{nr} + \gamma_{SP})/(\gamma_r + \gamma_{nr})$, where γ_r , γ_{nr} , and γ_{SP} are the radiative decay rate, non-radiative decay rate, and the coupling rate of electronhole recombination into SPs in the Ag grating, respectively. Because the non-radiative decay rate is smaller at lower temperatures [12]. Without knowing the relative strength between γ_r and γ_{nr} , it is not possible to derive the Purcell factor from these measurements [11]; however, from the overall increase in the total decay rate we may attribute to the contributions of SPs.

In the next step, we characterized the EL properties of the SP-LED. In Fig. 5(a), the open red circles denote the luminous flux from an SP-LED as a function of the injected current, and the open black rectangles indicate the reference. Our measurements were performed without a heat sink, so the injected current value was limited to 0.1 A. The luminous flux of the SP-LED is increased by 25% at 0.1 A compared to the reference. In addition to the EL enhancement, the SP-LED shows an improvement in the current-voltage (I-V) characteristics. The improvement factor was measured as 13% for the entire range of the injected current, which might be caused by the enhancement of the luminous flux.



Fig. 5. (a) The EL of the SP-LED as a function of the injected current in the range of $0.01 \sim 0.1$ A. 25% of the luminous enhancement is obtained at 0.1 A. (b) The current-voltage characteristic of the SP-LED. Almost constant improvement of 13% was observed in the entire current range.

The luminous efficiency of LEDs tends to decrease as the injected current increases. This 'efficiency droop' is caused by the damaged IQE of LEDs at high operating power, and is one serious problem in the development of high power LEDs [14]. Because mediating SPs can provide larger improvements in the performance of LEDs with a lower initial IQE [16], SP-LEDs are anticipated to be a possible solution for alleviating the efficiency droop. To verify this point, we measured the WPE of the SP-LED, which is defined as the ratio of the radiating power to the applied electric power. The WPE of the SP-LED is higher than that of the reference in the entire current range, as shown in Fig. 6(a). The WPE of the reference dropped from 2.87% at 0.01 A to 1.9% at 0.1 A, while the WPE of the SP-LED dropped from 4.12% at 0.01 A to 2.88% at 0.1 A. Here, we define a parameter of the WPE enhancement-factor: $\Delta \eta_{wall-plug} = (\eta_{SP-LED} - \eta_{reference}) / \eta_{reference}$, where η_{SP-LED} and $\eta_{reference}$ are the WPEs of the SP-LED and the reference, respectively. If the enhancement of the SP-LED is caused by other mechanisms than the SP-QW coupling such as by the increased optical scattering by the grating structure, then the enhancement factor should be a constant for all current values. As shown in Fig. 6(b), the consistent increase in $\Delta \eta_{wall-plug}$ from 44.2% at 0.01 A up to 51.2% at 0.1 A reveals that the SP-coupling would greatly alleviate the efficiency droop problem.



Fig. 6. (a) The WPE of the SP-LED and the reference as a function of the injected current. (b) The WPE enhancement factor $\Delta \eta_{power}$ is consistently increasing with the injected current, up to 51.2% at 0.1 A.

4. Conclusions

We measured experimentally the EL enhancement of SP-mediated green GaN LEDs and found evidence of SP-QW coupling in our data. In considering the technical issues involved in fabrication, we chose a reflection type InGaN/GaN LED with a metallic (Ag) pillar array on the p-contact. The carefully chosen system parameters allowed a 20% enhancement of the PL, and 25% enhancement of the EL. The increased electron-hole recombination rate due to the SPs was verified by TRPL. The enhancement of the WPE increased with the injected current, and reached up to 51.2% at the highest current value of 0.1 A, which proves the capability of SP-LEDs as a solution for high-power green LEDs. Here we should note that the measured enhancement factors of the green SP-LED in this work are much smaller than those predicted in our previous theoretical study [15] due to a large compromise in the system design for an experimental demonstration. However, we believe that SP-LEDs possess enough potential to be a solution in realizing high-power green LEDs if provided with sufficiently high fabrication skills.

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