Light-extraction enhancement of red AlGaInP light-emitting diodes with antireflective subwavelength structures

Y. M. Song\(^1\), E. S. Choi\(^1\), J. S. Yu\(^2\), and Y. T. Lee\(^1,\ast\)

\(^1\)Department of Information and Communications, Gwangju Institute of Science and Technology, 1 Oryong-dong, Buk-gu, Gwangju 500-712, Republic of Korea

\(^2\)Department of Electronic Engineering, Kyung Hee University, 1 Seocho-eup, Giheung-gu, Yongin-si, Gyeonggi-do 446-701, Republic of Korea

\(\ast\)ytlee@gist.ac.kr

Abstract: We demonstrate the enhancement of light extraction in 633 nm AlGaInP light-emitting diodes (LEDs) with antireflective subwavelength structures (SWS). From the contour plots by the rigorous coupled wave analysis method, it is found that the reduction of the internal reflection strongly depends on the period of SWS. The Ag nanoparticles formed by thermal dewetting were used as an etch mask for dry etch process to fabricate antireflective SWS on the LED surface. The tapered pillars on the GaP were fabricated, on average, with distances below 200 nm, satisfying the required antireflection condition at the emission wavelength. The improvement in light output power by \(~26.4\%\) was achieved for the fabricated AlGaInP LEDs with SWS compared to the conventional LEDs due to a strongly reduced Fresnel internal reflection at the GaP/air interface. The improved directionality in the far-field pattern was also obtained due to the directional light extraction enhancement.

©2009 Optical Society of America

OCIS codes: (050.6624) Subwavelength structures; (220.4241) Nanostructure fabrication; (230.3670) Light-emitting diodes.

References and links

1. P. Lalanne, and G. M. Morris, “Antireflection behavior of silicon subwavelength periodic structures for visible light,” Nanotechnology 8(2), 53–56 (1997).
2. K. Kintaka, J. Nishii, A. Mizutani, H. Kikuta, and H. Nakano, “Antireflection microstructures fabricated upon fluorine-doped SiO\(_2\) films,” Opt. Lett. 26(21), 1642–1644 (2001).
3. Y. Kanamori, M. Ishimori, and K. Hane, “High efficient light-emitting diodes with antireflection subwavelength gratings,” IEEE Photon. Technol. Lett. 14(8), 1064–1066 (2002).
4. M. Ishimori, Y. Kanamori, M. Sasaki, and K. Hane, “Subwavelength antireflection gratings for light emitting diodes and photodiodes fabricated by fast atom beam etching,” Jpn. J. Appl. Phys. 41(1 Part 1, No. 6B), 4346–4349 (2002).
5. Z. Yu, H. Gao, W. Wu, H. Ge, and S. Y. Chou, “Fabrication of large area subwavelength antireflection structures on Si using trilayer resist nanoimprint lithography and liftoff,” J. Vac. Sci. Technol. B 21(6), 2874–2877 (2003).
6. Y. M. Song, S. Y. Bae, J. S. Yu, and Y. T. Lee, “Closely packed and aspect-ratio-controlled antireflection subwavelength gratings on GaAs using a lenslike shape transfer,” Opt. Lett. 34(11), 1702–1704 (2009).
7. Y.-F. Huang, S. Chattopadhyay, Y.-J. Jen, C.-Y. Peng, T.-A. Liu, Y.-K. Hsu, C.-L. Pan, H.-C. Lo, C. H. Hsu, Y. H. Chang, C.-S. Lee, K.-H. Chen, and L.-C. Chen, “Improved broadband and quasi-omnidirectional anti-reflection properties with biomimetic silicon nanostructures,” Nat. Nanotechnol. 2(12), 770–774 (2007).
8. P. Yu, C.-H. Chang, C.-H. Chiu, C.-S. Yang, J.-C. Yu, H.-C. Kuo, S.-H. Hsu, and Y.-C. Chang, “Efficiency enhancement of GaAs photovoltaics employing antireflective indium tin oxide nanocolumns,” Adv. Mater. 21(16), 1618–1621 (2009).
9. T. Lohmüller, M. Helgert, M. Sundermann, R. Brunner, and J. P. Spatz, “Biomimetic interfaces for high-performance optics in the deep-UV light range,” Nano Lett. 8(5), 1429–1433 (2008).
10. Y. Kojima, and T. Kato, “Nanoparticle formation in Au thin films by electron-beam-induced dewetting,” Nanotechnology 19(25), 255605 (2008).
11. J.-M. Lee, and B.-I. Kim, “Thermal dewetting of Pt thin film: Etch-masks for the fabrication of semiconductor nanostructures,” Mater. Sci. Eng. A 449–451, 769–773 (2007).
12. S. Wang, X. Z. Yu, and H. T. Fan, “Simple lithographic approach for subwavelength structure antireflection,” Appl. Phys. Lett. 91(6), 061105 (2007).
1. Introduction

Light-emitting diodes (LEDs) have been widely used in various applications, such as traffic signals, indoor/outdoor full-color displays and backlight units for liquid crystal display, as energy-saving and environment-friendly light sources. Many practical applications require high performance LEDs. However, the light extraction efficiency of LEDs is limited strongly by the internal reflection loss due to the high refractive index contrast between the semiconductor material of LEDs and the air. For AlGaNp LEDs, ~30% of Fresnel internal reflection occurs at the interface between the GaP and the air, which significantly degrades the device performance. Recently, subwavelength structures (SWS) with a tapered profile have been a promising candidate for high-efficiency optical devices, including solar cells, LEDs, photodetectors and transparent glasses, due to their excellent antireflection properties [1–9].

On the view of the effective medium theory, the SWS with tapered features can be considered as a homogeneous medium with a graded refractive index determined by the fill factor. Thus the Fresnel reflection loss in LED structures can be reduced significantly by introducing SWS on the top surface of the LED. While many studies have been reported on the reflectivity characterizations of SWS, there are only few studies on SWS integrated LEDs [3,4].

For the fabrication of SWS, the masks were formed usually by e-beam, nanoimprint, or laser holographic lithography [1–6]. Thermal dewetting process of thin metal films provides subwavelength scale etch mask patterns without lithography process, which enables cost-effective fabrication [10–12]. Moreover, the average size of nanoparticles and their separation can be controlled by film thickness at a given annealing temperature [10]. In this letter, we fabricated the AlGaNp LEDs with antireflective SWS formed by the dry etch process of thermally dewetted Ag nanoparticles, thus improving the light extraction of the device. Light output characteristics including far-field patterns were measured and its theoretical explanation by the reflectivity calculation was also shown.

2. Fabrication of the SWS integrated AlGaNp LEDs

Figure 1 shows the schematic illustration for the fabrication procedure of the SWS integrated AlGaNp LEDs using thermally dewetted Ag nano masks. Epitaxial layers are composed of 8-um thick p-GaP window layer, 20 pairs of GaInP/AlGaNp multiple quantum wells surrounded by AlGaNp cladding layers, 16 pairs of AlAs/AlGaAs distributed Bragg reflectors on n-type GaAs substrate. For comparison, we fabricated 400 μm × 400 μm conventional AlGaNp LEDs. For p- and n- ohmic contacts, AuBe/Au (50 nm/500 nm) and Ni/Au/Ge/Ni/Au (20 nm/100 nm/50 nm/30 nm/500 nm) metals were deposited on the p-GaP layer and the backside of the wafer, respectively, by using an e-beam evaporator. Each device was etched down to the n-GaAs substrate by using an inductively coupled plasma (ICP) for device isolation.

For the fabrication of SWS integrated AlGaNp LEDs, 50 nm thick SiO₂ layer was deposited on the fabricated LED surface by using a plasma enhanced chemical vapor deposition. Then, Ag thin film with a thickness of 10 nm was deposited. After that, thermal dewetting process was carried out at 500 °C for 1 min under a nitrogen atmosphere by using a rapid thermal annealing. The annealing temperature was determined to reduce the ohmic resistance as well as to form the separated Ag nanoparticles by self-assembled agglomeration. The SiO₂ films using the Ag nano mask were patterned by a reactive ion etching (RIE) in CF₄/O₂ gas mixture to expose the GaP layer. To form the SWS with a tapered profile, the
underlying GaP layer was etched by ICP at an optimum condition, i.e., SiCl\textsubscript{4}/Ar (7.5 sccm/2.5 sccm) with rf power of 150 W for 9 min. The residual Ag/SiO\textsubscript{2} masks were removed by CF\textsubscript{4}/O\textsubscript{2} RIE. The scanning electron microscopy (SEM) measurements were performed to observe the thermally dewetted Ag nanoparticles and the fabricated LED structures with SWS. All characterizations were carried out in a wafer level at room temperature. The light output power from the top of the LEDs was measured by a large-size Si photodiode of 1 × 1 cm\textsuperscript{2} in close proximity to the device. The light emission intensity was also measured as function of emission angle.

Even though the SWS fabricated using Ag nanoparticles do not have periodic structure, it is noteworthy to analyze the reflectance by the variation of the period of SWS to determine the optimal distance of each pillar of SWS. The theoretical calculations of reflectance were implemented by using the rigorous coupled-wave analysis (RCWA) method, which is proposed by Moharam [13,14]. The RCWA method is generally used in SWS simulations because of fast computation time and readable result [3,6]. The model was constructed to GaP tapered pillars with a hexagonal structure on GaP substrate. Figure 2(a) shows the contour plot of the variation of reflectance caused by the external reflection from the air to the GaP as a function of pillar period and wavelength for a pillar height of 400 nm. As expected, the low reflectance band broadens and shifts towards a higher wavelength region as the period of the array increases [15]. The SWS with a period of ~200-500 nm shows the reflectance below 5% in the wavelength range of λ ~500-1000 nm. In the case of optical device applications such as solar cells and photodetectors, it is evident that the period of 200-500 nm, which can be easily implemented by a laser holographic lithography, is enough to reduce the reflection loss over a wide spectral range. In contrast, for LED structures, the internal reflection from the semiconductor material to the air should be considered.

Figure 2(b) shows the contour plot of the variation of reflectance caused by the internal reflection from the GaP to the air as a function of pillar period and wavelength for a pillar height of 400 nm. At the incident wavelength of 633 nm, the SWS with the period of > 250 nm exhibit very high reflectance (> 30%) due to the higher order diffraction. When the light is
incident on the grating structure with a period $\Lambda$, the angles of the reflected diffraction waves $\theta_{r,m}$ in the m-th diffraction order are given by the following grating equation [16]:

$$\sin \theta_{r,m} = \frac{m\lambda}{\Lambda n} + \sin \theta_i$$

(1)

where $n$ is the refractive index of incident medium, $\theta_i$ is the incident angle, and $\lambda$ is the incident wavelength. If the period of gratings becomes much smaller than the optical wavelength, we find that only zeroth order diffraction is allowed to reflect and all the others are evanescent. In the case of internal reflection, the refractive index of incident medium is much higher than that of air. Hence, the grating period should be much smaller than in the case of external reflection. The distance between the pillars of SWS should be smaller than ~200 nm to reduce the Fresnel reflection loss as shown in Fig. 2(b).

![Fig. 2. Contour plot of the variation of reflectance caused by (a) the external reflection from the air to the GaP and (b) the internal reflection from the GaP to the air as a function of pillar period and wavelength for a pillar height of 400 nm.]

3. Results and discussion

The average diameter and distance between the nearest Ag nanoparticles can be controlled by the Ag film thickness and the annealing temperature. Figure 3 shows the SEM images of the Ag films on SiO$_2$ (a) as-deposited with the thickness of 10 nm and annealed at 500 °C for 1 min under a nitrogen atmosphere with thicknesses of (b) 5 nm, (c) 10 nm, and (d) 20 nm. In order to clarify the characteristics of Ag nanoparticles, we estimated the average diameter and
positional correlation using a commercial image processor (ImageJ 1.42q, NIH). The estimated average diameters of Ag nanoparticles with film thicknesses of 5, 10 and 20 nm were $36.20 \pm 30.43$, $79.74 \pm 68.08$, and $179.19 \pm 130.20$ nm, respectively. The average distances of the nearest particles were 53.17, 116.8, and 194.5 nm, respectively, which are deduced by the pair correlation function [10,17]. Hence, 10 nm thickness of Ag film, which is enough to make SWS smaller than 200 nm, was chosen for the fabrication of SWS integrated LEDs.

![SEM images of the Ag films on SiO2](image)

Fig. 3. SEM images of the Ag films on SiO2 (a) as-deposited with the thickness of 10 nm and annealed at 500 °C for 1 min under a nitrogen atmosphere with thicknesses of (b) 5 nm, (c) 10 nm, and (d) 20 nm.

Figure 4(a) shows the SEM image of the fabricated AlGaInP LED integrated with SWS. The magnified image shows the SWS on GaP and contact metal surface at an oblique angle. Due to the etch process over the whole surface, AuBe/Au p-contact metal was also textured. Figure 4(b) shows the light-current-voltage (L-I-V) curves of the conventional LED and the LED with SWS at room temperature. The light output power was improved by 26.4% for the LED with SWS compared to the conventional LEDs at a bias current of 100 mA. It is clear that the improvement of the light extraction is attributed to the strongly reduced internal reflection at the GaP/air interface. For the conventional LED, the turn-on voltage was ~1.77 V and the differential series resistance was ~4.75 $\Omega$. The electrical characteristics were not degraded seriously after the SWS process though the forward voltage of the LED with SWS was slightly increased with increasing current due to the leakage current caused by the roughened surface. As shown in the inset of Fig. 4(b), the peak wavelength of the conventional LED were centered at 633 nm with 13.17 nm full width at half maximum under the bias current of 100 mA and it did not change by the introduction of SWS. The LED with SWS exhibited higher light intensity than the conventional LED at the emission wavelength.
The beam patterns of LEDs were measured to further elucidate the optical characteristics underlying the enhancement of the light extraction. A silicon photodiode is mounted on a rotating arm driven by a motorized actuator, which is controlled via a stepping motor. The detector is placed at a distance of ~15 cm from the LEDs and can be rotated by 180 degree with an angular resolution of 0.15 degree. Figure 5(a) shows the measured far-field patterns of the conventional LED and the LED with SWS. The emission intensity of the LED with SWS is enhanced along all directions. Particularly, the highest intensity enhancement was obtained along the normal direction and it decreased gradually as the emission angle increased, indicating an improved photon extraction from the top surface of the LED. This directional light with significantly increased intensity can be reasonably explained by the reflectance calculation at different incident angles of light. Figure 5(b) shows the calculated internal reflectance of the GaP with and without SWS as a function of incident angle at a wavelength of 633 nm. The GaP with SWS exhibited much lower reflectance than the flat GaP surface for both TE and TM polarizations at 0 degree of incident angle (i.e., normal incidence). However, as the incident angle was increased, the reflectance difference between the GaP with SWS and the flat GaP was diminished. Thus, this leads to the angle dependent light enhancement of the LED with SWS.
Fig. 5. (a) Measured far-field patterns of the conventional LED and the LED with SWS, and (b) Calculated internal reflectance of the GaP with and without SWS at a wavelength of 633 nm.

4. Conclusion

In conclusion, we fabricated the 633 nm AlGaInP LEDs with antireflective SWS by the dry etch process using Ag nanoparticles, which are formed by the thermal dewetting process, together with the RCWA simulation. It is found that the average distance between the pillars of SWS is critical to reduce the Fresnel reflection from the semiconductor material to the air. The fabricated LED with SWS exhibited an enhancement of 26.4% in light output power due to the reduced Fresnel internal reflection with improved directional light emission characteristics. From these results, we believe that the LED with SWS is cost-effective and suitable for high-efficiency light source and directional display applications.

Acknowledgement

This work was supported by the IT R&D program of MKE/IITA [2007-F-045-03].