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Disordered antireflective nanostructures on GaN-based light-emitting diodes using Ag nanoparticles for improved light extraction efficiency

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In this study, we demonstrate GaN light-emitting diodes (LEDs) with antireflective subwavelength structures (SWS) for enhanced light extraction efficiency. To eliminate the internal Fresnel reflection, SWS were fabricated on an indium-tin-oxide (ITO) surface using an overall dry etch process of Ag nanoparticles. The average size of the Ag nanoparticles was carefully chosen by theoretical calculation of the reflective diffraction efficiency using a rigorous coupled-wave analysis (RCWA) method. Improvement in light output power of ~30.2% was achieved for the fabricated ITO SWS LEDs compared to conventional LEDs, with no significant increase in the forward voltage. © *2010 American Institute of Physics*. [doi:10.1063/1.3488001]

As energy-saving and environment-friendly light sources, GaN-based light-emitting diodes (LEDs) have attracted considerable attention for a variety of applications, such as traffic signals, indoor/outdoor displays, and backlight units for liquid crystal displays.[1,2] However, the external quantum efficiency of GaN-based LEDs is low, because the refractive index of the nitride epitaxial layer differs greatly from that of air. For GaN LEDs, when the light generated from the active layer propagates in the normal direction of the LED surface, internal Fresnel reflection of ~20% occurs at the interface between the GaN and the air. As the incident angle increases from normal to the critical angle (~23°), the Fresnel reflection reaches 100%, which significantly degrades the device performance. To eliminate this Fresnel reflection, the use of graded refractive index layers has recently been reported as a potential method.[3,4] By using graded index layers, the internal Fresnel reflection can be significantly reduced within the escape cone.

Similarly, antireflective subwavelength structures (SWS) with an ordered or disordered tapered feature can be considered as a homogeneous medium with a graded refractive index, which is determined by the packing density.[5–9] Having excellent antireflection properties, antireflective SWS have been extensively characterized. However, there have been few reports on SWS integrated LEDs.[5,6] Even though e-beam or laser interference lithography is commonly used to form SWS, these methods are complex and difficult for mass production. Very recently, several researchers applied thermally dewetted metal nanoparticles to fabricate nanostructures.[10,11] Compared with lithography processes, the use of metal nanoparticles enables simple and low-cost fabrication. In this study, we fabricated GaN LEDs with antireflective SWS formed by an etch process of Ag nanoparticles for improving the light extraction of the device. Detailed process steps and the properties of the fabricated LEDs are also shown.

A schematic diagram of the fabrication procedure for disordered antireflective nanostructures on GaN-based LEDs is presented in Fig. 1. Blue InGaN/GaN LED structures emitting at $\lambda \sim 475$ nm were grown on a *c*-plane sapphire (Al_2O_3) substrate using a metal-organic chemical vapor deposition (MOCVD) system. As shown in Fig. 1(a), the epitaxial structures consisted of a 1.3 μm undoped GaN buffer layer, a 5 μm Si-doped n-type GaN layer, a 1.3 μm highly Si-doped n-type GaN layer, five periods of InGaN/GaN multiple quantum wells, and a 0.1 μm Mg-doped p-type GaN layer. First, conventional GaN-based LEDs with a planar ITO contact layer were fabricated by a standard process. A 300 nm SiO_2 was deposited on an epilayer by plasma enhanced chemical vapor deposition (PECVD) as an etch mask and the film was patterned by photolithography. After mesa etching by an inductively coupled plasma (ICP)

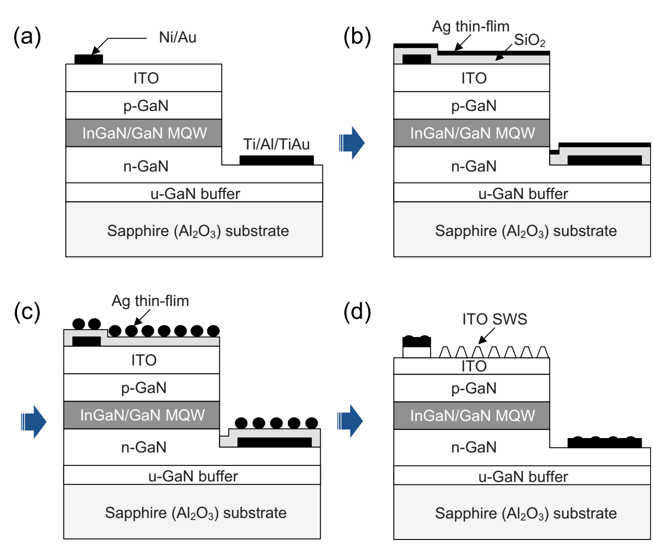

FIG. 1. (Color online) Schematic illustrations of fabrication procedures for disordered antireflective nanostructures on GaN-based light-emitting diodes.

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etcher, a 400-nm-thick ITO layer was deposited onto the top InGaN layer and was annealed at 500 °C in air ambient for 1 min. Ni/Au (30 nm/300 nm) and Ti/Al/Ti/Au (30 nm/80 nm/20 nm/300 nm) metals were then deposited by using an e-beam evaporator for p- and n-metal contacts, respectively. Each device, with dimensions of $450 \times 450\ \mu m^2$, was etched down to the undoped GaN by an ICP etcher for device isolation.

For fabrication of SWS integrated GaN LEDs, a 50-nm-thick SiO_2 layer was deposited on the fabricated LED surface by PECVD, and then Ag thin film with a thickness of 15 nm was deposited using an e-beam evaporator. The insertion of a SiO_2 layer serves as both a durable etch mask and a buffer layer to form Ag nanoparticles. A thermal dewetting process was carried out at 500 °C for 1 min under a nitrogen atmosphere via rapid thermal annealing. The dewetting temperature and time was carefully chosen to reduce the contact resistance as well as to form separated Ag nanoparticles by self-assembled agglomeration. The SiO_2 layer was patterned by an overall dry etch process in a CF_4/O_2 gas mixture using Ag nanoscale masks. In order to create ITO SWS with a tapered profile, the underlying ITO layer was etched by an ICP etcher at optimum conditions, i.e., $CH_4/H_2/Ar$ (20/40/20 SCCM) with rf power of 100 W for 5 min. The residual Ag/SiO_2 masks were removed by CF_4/O_2 reactive ion etching. The thermally dewetted Ag nanoparticles and the fabricated LED devices were observed by using a field-emission scanning electron microscope (FE-SEM) with an operating voltage of 10 kV. The final processed LED devices were mounted on a lead frame and all characterizations were carried out using a conventional integration sphere.

Porous thin films formed by oblique-angle deposition or an electrochemical etching process can be simply considered as an effective index medium, because the gaps between each pore (<50 nm) are much smaller than the optical wavelength. In SWS fabricated by Ag nanoparticles, however, the average size and diameter should be carefully chosen to eliminate higher order diffraction in a grating structure. Hence, it is necessary to calculate the reflectance by the variation in the period of the SWS to determine the optimal distance of the SWS. The theoretical calculation of reflectance was performed by using a rigorous coupled-wave analysis method.[12] The model used in this calculation was constructed for an ITO SWS with a tapered profile on a GaN substrate. The material dispersion of GaN and ITO was considered to obtain the exact results at each wavelength.[13,14] The height of the tapered profile and the thickness of ITO were fixed to 150 nm and 250 nm, respectively. We also assumed that the light comes from GaN to air at normal incidence to take account into the reflectance from the interface between GaN and ITO as well as that from the ITO/air interface. In this calculation, the reflectance from the interface between GaN and ITO was considered. Figure 2 shows the contour plot of the reflectance variation caused by the internal reflection as a function of SWS period and incident wavelength. At an incident wavelength of 475 nm, the SWS with a period of >300 nm exhibit very high reflectance (>20%) due to higher order diffraction. When the light is incident on the grating structure with a period of Λ at the normal direction, the angle of the reflected diffraction waves $\theta_{r,m}$ in the m-th diffraction order is given by the grating equation, i.e., $\sin \theta_{r,m} = m\lambda/\Lambda n$, where λ is the incident

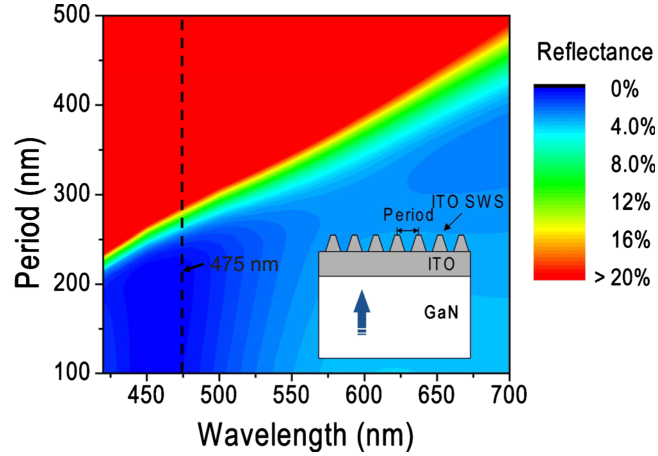

FIG. 2. (Color online) Contour map of reflectance variations of ITO SWS on GaN substrate as a function of period and wavelength.

wavelength and n is the refractive index of the incident medium.[15] If the grating period becomes much smaller than the optical wavelength, it is found that only zeroth order diffraction is allowed to reflect and all the higher orders are evanescent. Hence, the distance between the tapered pillars of the SWS should be smaller than ∼220 nm in order to reduce the Fresnel reflection loss at the blue emitting wavelength, as depicted in Fig. 2.

The average size of Ag nanoparticles can be controlled by the Ag film thickness. Figure 3 shows SEM images of the Ag films on $SiO_2/ITO/GaN/Sapphire$ (a) as-deposited with a thickness of 30 nm and annealed at 500 °C for 1 min under

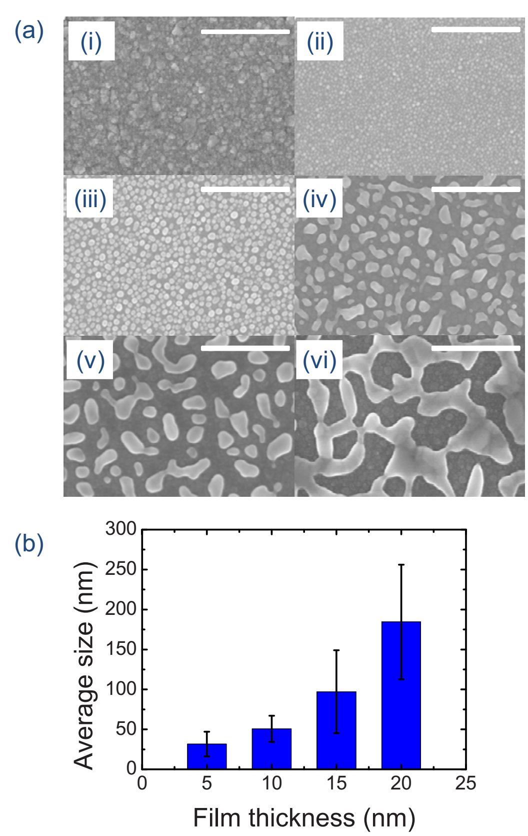

FIG. 3. (Color online) (a) SEM images of Ag thin films on $SiO_2/ITO/GaN/sapphire$ annealed (i) as deposited (30 nm) and annealed at 500 °C for 1 min with thickness of (ii) 5 nm, (iii) 10 nm, (iv) 15 nm, (v) 20 nm, and (vi) 30 nm, respectively. Scale bar corresponds to 1 μm. (b) Average diameter of Ag nanoparticles with different deposited film thickness.

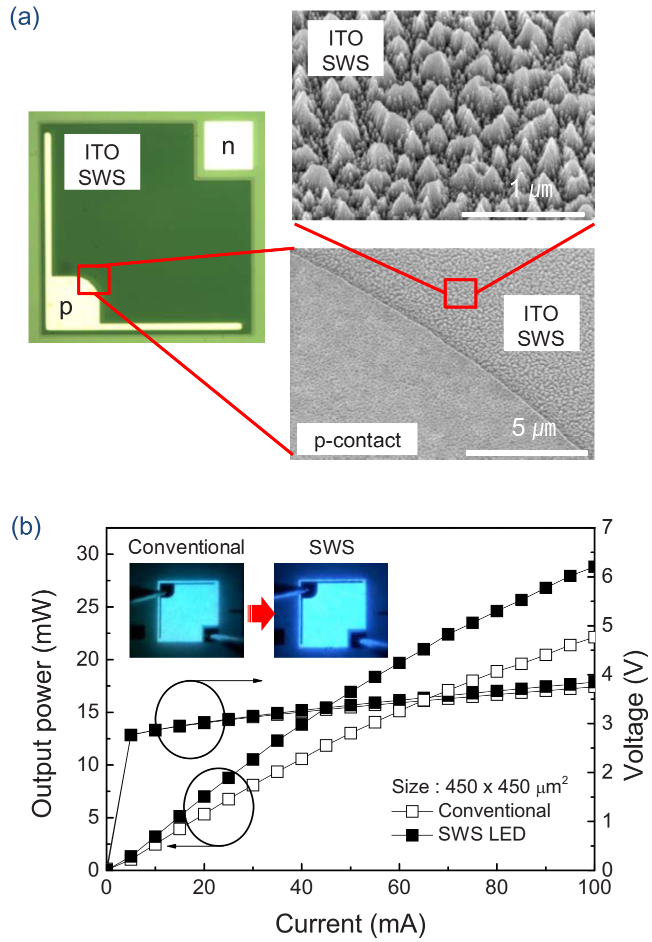

FIG. 4. (Color online) (a) Top view of microscope image of the fabricated SWS integrated GaN light-emitting diode (left) and tilted angle view of SEM images of the fabricated ITO SWS. (b) Light-output power of conventional and SWS integrated top emitting InGaN LEDs, as a function of injection current. Inset shows light illumination images of conventional and SWS integrated InGaN LEDs at a injection current of 5 mA.

a nitrogen atmosphere with different Ag film thicknesses. Because the surface energy of Ag film is much higher than that of SiO_2 film, the former is unstable on SiO_2, resulting in dewetting of the film with the annealing process. To clarity the effect of Ag film thickness, the average diameter of the Ag nanoparticles was estimated using a commercial image processor (ImageJ 1.42q, NIH). As shown in Fig. 3(b), the average diameter is increased as the deposited film thickness increases; this is in agreement with the findings of a previous study in which an e-beam induced dewetting process was used.[10] Thick Ag films (>30 nm) lead to large and bicontinuous shapes, which are not adequate for subwavelength mask patterns. On the other hand, very thin films below 10 nm cannot act as a etch mask due to a mask erosion effect arising from the high density of energetic ions in the plasma. Under these considerations, a 15 nm thick Ag film, which is sufficient to make SWS smaller than 220 nm, was chosen for the fabrication of SWS integrated GaN LEDs.

Figure 4(a) shows a top view microscope image (left) of the fabricated SWS integrated GaN LEDs with FE-SEM images (right) of the SWS on the ITO and the p-metal electrode. As depicted in Fig. 4(a), the fabricated ITO SWS has a thicknesses of 150–250 nm with tapered features, which can reduce the Fresnel reflection efficiently while sustaining the electrical characteristics. Due to the overall dry etch process, the Ni/Au p-contact metal was also textured slightly. Figure 4(b) shows the light-current-voltage (L-I-V) curves of the conventional and SWS integrated LEDs at room temperature. The forward voltage and differential series resistance of SWS LEDs were ~3.025 V and ~13.9 Ω, respectively. The slightly larger forward voltage from the SWS LEDs is caused by the roughened surface; it would not, however, result in significant degradation of the electrical properties of the LEDs. The light output power was improved by 30.2 % for the LED with ITO SWS compared to the conventional LEDs at a bias current of 100 mA. The improvement of the light extraction is attributed to the elimination of Fresnel internal reflection as expected from the theoretical calculation shown in Fig. 2.

In conclusion, we fabricated GaN LEDs with antireflective SWS, which act as a graded effective refractive medium, by overall dry etching of Ag nanoparticles. From the theoretical calculation of the reflective diffraction efficiency of the LED surface with SWS, we found that a SWS period of below 220 nm is needed to reduce the internal Fresnel reflection. The fabricated LED with SWS exhibited 30.2% enhanced light output power relative to conventional LEDs without degradation of electrical properties. The results indicate that the SWS integrated LEDs have considerable potential for future low-cost and high-efficiency GaN LED applications.

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