Enhanced optical power and low forward voltage of GaN-based light-emitting diodes with Ga-doped ZnO transparent conducting layer

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Ga-doped ZnO (ZnO:Ga) films were grown by metalorganic chemical vapor deposition as transparent conducting layers for GaN light-emitting diodes (LEDs). The forward voltage of LEDs with ZnO:Ga was 3.3 V at 20 mA. The low forward voltage was attributed to the removal of a resistive ZnGa2O4 phase, decreased resistivity of ZnO:Ga films, and increased hole concentration in p-GaN by thermal annealing process. The light output power of LEDs with ZnO:Ga was increased by 25% at 20 mA compared to that of LEDs with Sn-doped indium oxide due to the enhanced transmittance and the increased hole concentration in p-GaN. © 2010 American Institute of Physics.

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The efficiency of GaN-based light-emitting diodes (LEDs) has been greatly improved and LEDs have found many applications in automobiles, full-color displays, traffic signals, and solid-state lighting. To use GaN LEDs in high efficiency applications, further improvement of the external quantum efficiency is indispensable. One of the issues for realizing highly efficient GaN LEDs is the improvement of light extraction efficiency due to a photon loss through a total internal reflection of light in LEDs. Extensive investigation has been carried out to improve light extraction efficiency by introducing surface roughening, photonic crystals, distributed Bragg reflectors (DBRs) or omnidirectional reflectors (ODRs), patterned substrate, and chip shaping. In addition, other limitation on the low light extraction efficiency is poor transparency (~70%) of Ni/Au on p-GaN. To overcome the shortcomings of the Ni/Au transparent conducting layer (TCL), highly transparent conducting oxide (TCO) films such as impurity-doped SnO2, In2O3, and CdO binary compounds were investigated. Tin-doped indium oxide (In2O3/Sn, ITO) is currently the most popular TCO due to its low resistivity (~10-4 Ω cm) and high transparency in the visible region. However, indium is a rare and expensive element and thermal stability of ITO is poor for an application to transparent electrodes in LEDs. Therefore, an alternative TCO material is needed for high efficiency LEDs. Recently, ZnO-based TCOs have received considerable attention as an alternative substance for p-GaN ohmic contact layer because it is abundant in nature, non-toxic material, and stable in hydrogen plasma process and high temperature process. In addition, ZnO-based TCOs are highly transparent due to a wide band gap (3.37 eV) and they have electrical properties similar to ITO. However, GaN LEDs with ZnO-based TCO have shown high forward voltages or poor-ohmic contact properties with rectifying characteristics.

In this work, we report on the properties of GaN LEDs with Ga-doped ZnO (ZnO:Ga) grown by metalorganic chemical vapor deposition (MOCVD). The light output power of GaN LEDs with ZnO:Ga was increased by 25% at 20 mA, compared to that of GaN LEDs with ITO. Furthermore, the forward voltage of GaN LEDs with ZnO:Ga at 20 mA was as low as 3.3 V.

The InGaN/GaN MQW LED wafers were grown on c-plane sapphire substrates by MOCVD. The LED structures consisted of a Si-doped n-GaN layer, an InGaN/GaN MQW active layer with an emission wavelength of 450 nm and a Mg-doped p-GaN layer. ZnO:Ga were grown on p-GaN at a working pressure of 50 Torr and a growth temperature of 500 °C by MOCVD. Diethylzinc (DEZn), oxygen (O2), and triethylgallium (TEG) were used as the Zn, oxygen, and n-type Ga doping source, respectively. The flow rates of DEZn, O2, and TEG were 20.1 μmol/min, 0.33 mol/min, and 17 mmol/min, respectively. The thickness of the ZnO:Ga layer was 280 nm. To fabricate top-emitting LEDs with a size of 300×300 μm2, ZnO:Ga layers were partially etched in a diluted HCl solution, and then LEDs structures were etched until the n-GaN layer was exposed by using an inductively coupled plasma with CH4/Cl2/H2/Ar gas sources. To compare electrical and optical properties, GaN LEDs with TCLs of ZnO:Ga, ITO (230 nm), and Ni (5 nm)/Au (5 nm) were prepared. The LEDs with ZnO:Ga were annealed at 700 °C in a nitrogen ambience for 1 min and the LEDs with ITO and Ni/Au TCLs were also annealed at 500 °C in an air ambience for 1 min. A Cr/Au (30/80 nm) layer was deposited as p- and n-electrodes.

In order to use ZnO:Ga films as TCLs in GaN LEDs, the resistivity of ZnO:Ga films was measured on the samples annealed up to 800 °C and it was found that the performance of GaN LEDs with ZnO:Ga films annealed at 700 °C was superior than LEDs annealed at different temperatures. The resistivity, Hall mobility, and electron concentration of ZnO:Ga films grown on u-GaN/sapphire was 5×10-4 Ω cm, 212 cm2/V s, and 5.4×1019 cm-3, respectively after thermal annealing at 700 °C for 1 min in N2 ambient gas. Figure 1 shows the transmittance of Ni/Au (5/5 nm), ITO (230 nm), and ZnO:Ga films (280 nm) deposited on sapphire substrates. As shown in Fig. 1, the transmittance...
of ZnO:Ga film annealed at 700 °C is 95% at a wavelength of 450 nm, and those of ITO and Ni/Au are 89% and 75%, respectively. It was reported that the transmittance of ZnO films can be enhanced by a higher c-axis orientation and an increased stoichiometry of the films due to decreased scattering center of light. As shown in Figs. 3(a)–3(c), XRD 2θ peaks show that ZnO:Ga films grown on p-GaN by MOCVD are oriented along the c-axis due to the wurtzite structure of ZnO:Ga. Furthermore, an atomic ratio of Zn and O was obtained by measuring the XPS depth profile of ZnO:Ga films after thermal annealing at 700 °C. The atomic ratio of Zn and O was calculated to be 1.03, indicating that ZnO:Ga films grown by MOCVD have a good stoichiometry. These results indicate that the high transparency of ZnO:Ga films shown in Fig. 1 is due to the improved stoichiometry and high c-axis-orientation of ZnO:Ga films.

The ZnO:Ga films were deposited on p-GaN as a TCL of GaN LEDs. Figure 2 shows the I-V characteristics of GaN LEDs with TCLs of Ni/Au, ITO, ZnO:Ga before and after thermal annealing. The forward voltage measured at 20 mA was 3.6, 3.6, and 4.0 V for GaN LEDs with Ni/Au, ITO, and ZnO:Ga, respectively. However, the forward voltage at 20 mA of GaN LEDs with ZnO:Ga was remarkably decreased from 4.0 to 3.3 V after thermal annealing at 700 °C. Kuo et al. reported that Al-doped ZnO TCL deposited on n+-InGaN/GaN short-period superlattice by e-beam evaporation showed a low forward voltage of 3.3 V. It is believed that the low forward voltage is attributed to low resistive ohmic contact by a high carrier concentration in n+-InGaN/GaN short-period superlattice contact layer. The low forward voltage in this study is attributed to a low resistivity of ZnO:Ga films, which was decreased from 3.2 to 5 × 10−4 Ω cm by thermal annealing at 700 °C. The carrier concentration and mobility were increased from 1 × 10^{19} cm^{-3} and 0.13 cm²/V s to 5 × 10^{19} cm^{-3} and 212 cm²/V s, respectively after thermal annealing. Figure 3(b) shows the XRD 2θ peaks at 52°, 56°, and 64°, which are due to the ZnGa₂O₄ phase formed in the as-grown ZnO:Ga films. However, these XRD 2θ peaks mostly disappeared after thermal annealing at 700 °C, as shown in Fig. 3(c). The ZnGa₂O₄ phase is believed to be formed at the interface between ZnO and GaN and it has a high resistivity of ~10 Ω cm at room temperature. The low forward voltage of GaN LEDs with ZnO:Ga after thermal annealing can be
attributed to a removal of the resistive ZnGa$_2$O$_4$ phase at the interface between ZnO:Ga and $p$-GaN by thermal annealing. Figure 3(d) shows that the Ga $2p_{3/2}$ XPS core level peak of the $p$-GaN after removal of ZnO:Ga films is shifted to the valence band edge from 1115.8 to 1115.3 eV. This indicates an increased hole concentration near the $p$-GaN surface region due to the outdiffused Ga atoms and indiffused Zn atoms for substitution of Ga sites. Furthermore, it was confirmed that the contact resistance obtained by circular transmission line model (CTLM) was greatly decreased from 9.12 $\times$ 10$^{-3}$ to 1.39 $\times$ 10$^{-4}$ $\Omega$ cm$^2$ after thermal annealing of ZnO:Ga TCL on p-GaN. These results show that the decrease in forward voltage of GaN LEDs with ZnO:Ga after thermal annealing is mainly due to the removal of the resistive ZnGa$_2$O$_4$, the decreased resistivity of ZnO:Ga films, and the increased hole concentration in p-GaN at the interface between ZnO:Ga and p-GaN after thermal annealing.

The light output powers of GaN LEDs with ZnO:Ga, ITO, and Ni/Au were measured as shown in Fig. 4. Light output power of GaN LEDs with ZnO:Ga was increased by 25 and 75% compared to those of GaN LEDs with ITO and Ni/Au, respectively. The increased light output power of GaN LEDs with ZnO:Ga can be attributed to the increased hole concentration at the interface between ZnO:Ga and p-GaN. As shown in Figs. 2 and 4, GaN LEDs with ZnO:Ga showed a lower forward voltage and a higher light output power compared to those of LEDs with ITO and Ni/Au. These results show that the power efficiency of LEDs with ZnO:Ga, which is defined by the ratio of optical output power to electrical input power, is much increased. At 20 mA of injection current, the power efficiency of GaN LEDs with ZnO:Ga was increased by 35 and 91% compared to those of GaN LEDs with ITO and Ni/Au, respectively. This study shows that ZnO:Ga TCO with low resistivity and high transmittance can replace the ITO layer that is widely used as a TCL in high efficiency GaN LEDs.

In summary, the low forward voltage of 3.3 V was observed at 20 mA for GaN LEDs with an annealed ZnO:Ga. This was attributed to the removal of the ZnGa$_2$O$_4$, the decreased resistivity of ZnO:Ga films, and the increased hole concentration in p-GaN. The light output power of GaN LEDs with ZnO:Ga was also increased by 25% compared to that of GaN LEDs with ITO. The increase in light optical power was attributed to the enhanced optical transmittance and an increased hole concentration in p-GaN at the interface between p-GaN and ZnO:Ga after thermal annealing at 700 °C. The power efficiency of LEDs with ZnO:Ga was increased by 35% due to the low forward voltage and the enhanced light output power compared to GaN LEDs with ITO.

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FIG. 3. (Color online) Light output power as a function of injection current for GaN LEDs with ITO, Ni/Au, and ZnO:Ga after thermal annealing.

FIG. 4. (Color online) Light output power as a function of injection current for GaN LEDs with ITO, Ni/Au, and ZnO:Ga after thermal annealing.