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Sputter deposition of Sn-doped ZnO/Ag/Sn-doped ZnO transparent contact layer for GaN LED applications

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ABSTRACT

Sn-doped ZnO/Ag/Sn-doped ZnO (ZAZ) multilayers prepared using sputtering process was employed in GaN-based blue light-emitting diodes(LEDs) as transparent contact layers (TCLs). The ZAZ layer had better optical and electrical properties without any thermal annealing. The ZAZ TCLs improved the current spreading on p-GaN layer and increased the light output power in the large area devices. The efficiency droop was also quite small in the case of adopting a ZAZ TCL in GaN-based LEDs. These results are promising for the development of a ZAZ TCL using sputtering process for GaN LED applications.

1. Introduction

Although there are new applications using GaN [1,2], GaN-based light-emitting diodes (LEDs) are still important because they save energy and offer high efficiency and a reliable lifetime [3,4]. Nowadays, the applications of GaN LEDs has widened to traffic signals, automotive lighting, communications, and illumination. However, further improvements in their light extraction efficiency and brightness are necessary for large-area devices. The light extraction efficiency is generally limited by poor current spreading [5] and the total internal reflection [6]. These two problems have become more dominant as the device area has increased because of electrical resistance and the lateral waveguiding effect. To overcome these problems, intensive research has been carried out and several possible solutions have been suggested, including the use of a hybrid transparent contact layer (TCL) [7,8], surface texturing of the TCL [9,10], and new LED designs [11,12]. Until now, indium tin oxide (ITO) has been widely used as a TCL for GaN LEDs. However, an alternative TCL with optical and electrical performances similar to or better than those of ITO materials in a large-area GaN LED is needed because indium is a scarce metal and its price has increased to by more than 10 times since the 1960s [13,14]. Recently, metal oxide-metal-metal oxide multilayer electrodes have emerged as promising TCLs for organic LEDs, organic solar cells, and flexible touch panels [15–18] due to the low resistivity and the high optical transmittance. The insertion of metal layers between thin oxide films ensures the long-term electrical stabilities of the metal layers by preventing metal oxidization [17].

In this work, we introduce Sn-doped ZnO/Ag/Sn-doped ZnO (ZAZ) multistructure as a new TCL, which is fabricated by a sputtering process for GaN blue LEDs. It has better electrical and optical properties than ITO without thermal annealing. Al-doped ZnO is popular material for a TCL, but Sn doping in ZnO improves its properties in electrical conductivity and optical transmittance, compared to Al doping [19]. Therefore, we adopt Sn-doped ZnO instead of Al-doped ZnO for ZAZ multistructure. Transparent electrode materials are commonly deposited by sputtering due to its effectiveness, but electron-beam evaporation is used to deposit TCL onto the p-GaN top layer because the sputtering process degrades the electrical property of p-GaN due to the high energy ions in the process, resulting in the failure of ohmic contact between TCL and p-GaN. We employed buried contact islands using ITO in the ZAZ multilayer and thus eliminated the problem of an ohmic contact failure. This structure shows better device performances even in a large area device than a conventional ITO TCL.

2. Experimental section

To apply as TCLs on fabricated GaN LEDs, a rectangular mesa was defined by dry etching to a thickness of 0.8 μm to expose the n-layer using an inductively-coupled plasma reactive ion etching system on which a Ti/Al/Ni/Au (30/70/30/70 nm) layer was

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deposited as an n-electrode by an e-beam evaporator. Rapid thermal annealing was performed at 550 °C for 1 min in N_2 ambient to form an n-type ohmic contact. To form ZAZ with ITO buried contact islands on the p-layer (for ZAZ-LEDs), 5 μm-size ITO islands (thickness = 100 nm), deposited by an e-beam evaporator with thermal annealing at 550 °C for 1 min in air ambient, were uniformly distributed by using a photolithographic process and then the ZAZ multilayer was formed by sputtering of an Sn-doped ZnO target (99.9% purity) and a pure Ag target (99.99% purity) at room temperature and a pressure of 5 mTorr. The Sn-doped ZnO layer and Ag were deposited with the thickness of about 42 nm and 10 nm, using an RF power of 200 W and DC power of 10 W, respectively. The ZAZ multilayer was patterned as a TCL on p-GaN by wet chemical etching using an oxide etchant ($HCl:HNO_3 = 7:3$) at room temperature. For a comparative study, reference GaN LEDs were fabricated with 300 nm ITO film as a TCL deposited by an e-beam evaporator and thermally annealed at 550 °C for 1 min in air ambient (for reference ITO-LEDs).

To form probing pad on the n- and p-electrodes, Ti/Au (20/10 nm) was deposited by e-beam evaporator. To implement our study, commercially available LED wafers were used; these were grown on c-plane sapphire substrates by a metalorganic chemical vapour deposition. The structure of the LEDs comprised 2.0 μm of undoped GaN, 3.5 μm of n-GaN, 5-period GaN/InGaN multiple quantum well active regions with 450 nm-emission, a 0.024 μm p-AlGaN electron blocking layer, and a 0.14 μm p-GaN layer. To investigate the electrical characteristics of the ZAZ multilayer with ITO buried contact islands to p-GaN, a transmission line model (TLM) method with circular geometry with an inner radius of 100 μm and a gap spacing of 5 μm was used to evaluate the contact resistance and ohmic behavior.

3. Results and discussion

Fig. 1(a) shows the optical specular transmittance spectra of the ZAZ layers with and without ITO buried contact islands; for comparison, the transmittance spectrum of a reference ITO film of 300 nm are also shown. All samples were fabricated with similar sheet resistances of 12–14 Ω/sq. Two spectra related to the ZAZ layers show significant drops at wavelengths below 400 nm due to the band-to-band absorption and the transmittance difference greater than 2% over 475 nm. In this study, the interesting wavelength is 450 nm at which a commercial GaN blue LED wafer shows an electroluminescence (EL) peak. The EL spectrum of the fabricated LEDs in Fig. 1(a) was acquired at 20 mA. Therefore, it is considered that the ZAZ layers with and without ITO buried contact islands have a very similar optical transmittance property around 450 nm, which is better than that of the reference ITO film.

Fig. 1(b and c) show schematic cross-sectional diagrams of the ZAZ-LED and the reference ITO-LED. The probing pad is a Ti/Au electrode and the length (L) and width (W) of the mesa were 200 and 500 μm, respectively. Fig. 1(d and e) show ITO buried contact islands on the p-GaN surface. Fig. 1(f) shows the current-voltage (I-V) characteristics of the ZAZ layer with and without ITO buried contact islands on the p-GaN, measured by the TLM method [20]. A nonlinear I-V characteristic is exhibited by the ZAZ layers without ITO buried contact islands and the ohmic property is clearly observed in the ZAZ layers with ITO buried contact islands. The specific contact resistance was estimated to be about 7.1E − 3 Ω-cm^2 in the ZAZ layer with ITO buried contact islands, which is two order of magnitude lower than that of the ZAZ layer without ITO buried contact islands (5.5E − 1 Ω-cm^2). ITO buried contact islands cover only about 2.5% of the whole TCL surface area, but enable the sputter-deposited ZAZ layer to function as a TCL, protecting the ohmic contact to p-GaN from sputtering damage. Fig. 2(a) shows the I–V curves of the ZAZ-LEDs and reference ITO-LEDs with various lateral mesa lengths (L). The series resistance of the ZAZ-LED is higher than that of the ITO-LED due to the small contact area in the ZAZ-LED, but the light output power versus injection-current is much higher in the ZAZ-LED than in the ITO-LED as shown in Fig. 2(b).

In particular, the increase in the light output power is more significant in a large ZAZ-LED compared to the reference ITO-LED, which is attributed to the current spreading by the ZAZ layer among the ITO contact islands. As expected from Fig. 1(f), the contact resistance of the ZAZ layer on p-GaN is much higher than that of ITO contact islands; therefore, the ZAZ layer can act as a

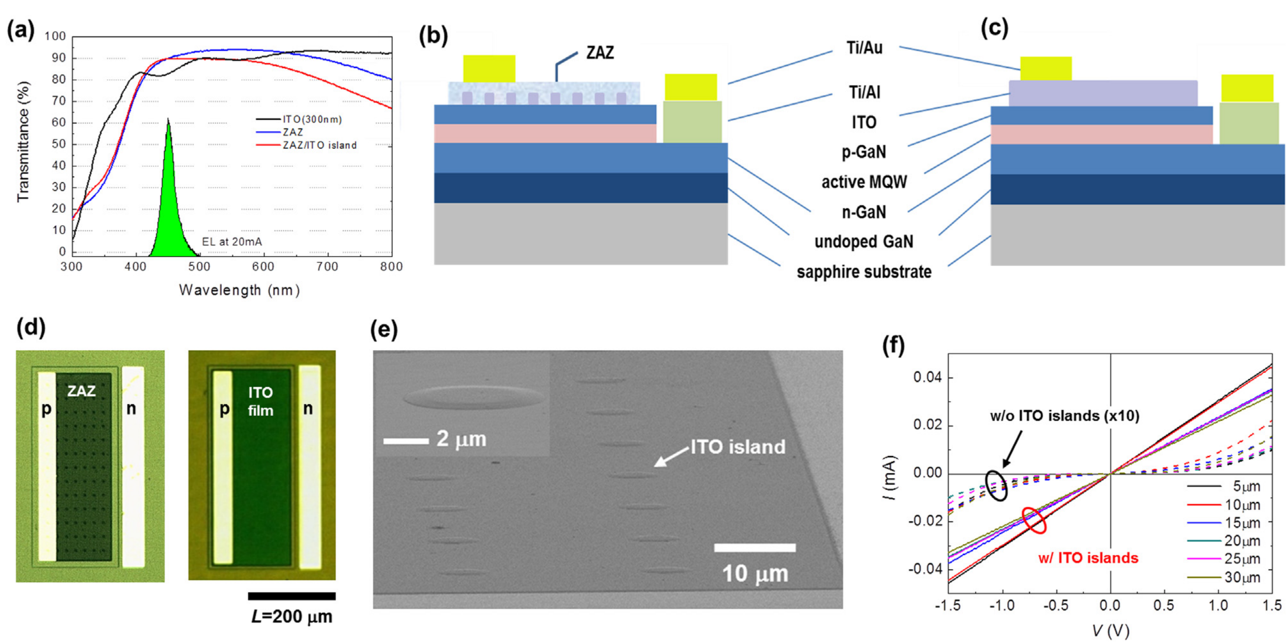

Fig. 1. (a) Optical specular transmittance spectra of various TCL materials. Schematic cross-sectional diagrams of (b) ZAZ-LED and (c) ITO-LED. (d) Optical microscopic top views of ZAZ- and ITO-LEDs. (e) Scanning electron microscopic image of the top surface of the p-GaN side in ZAZ-LED. (f) I-V curves of the ZAZ layer with and without ITO buried contact islands on the p-GaN.

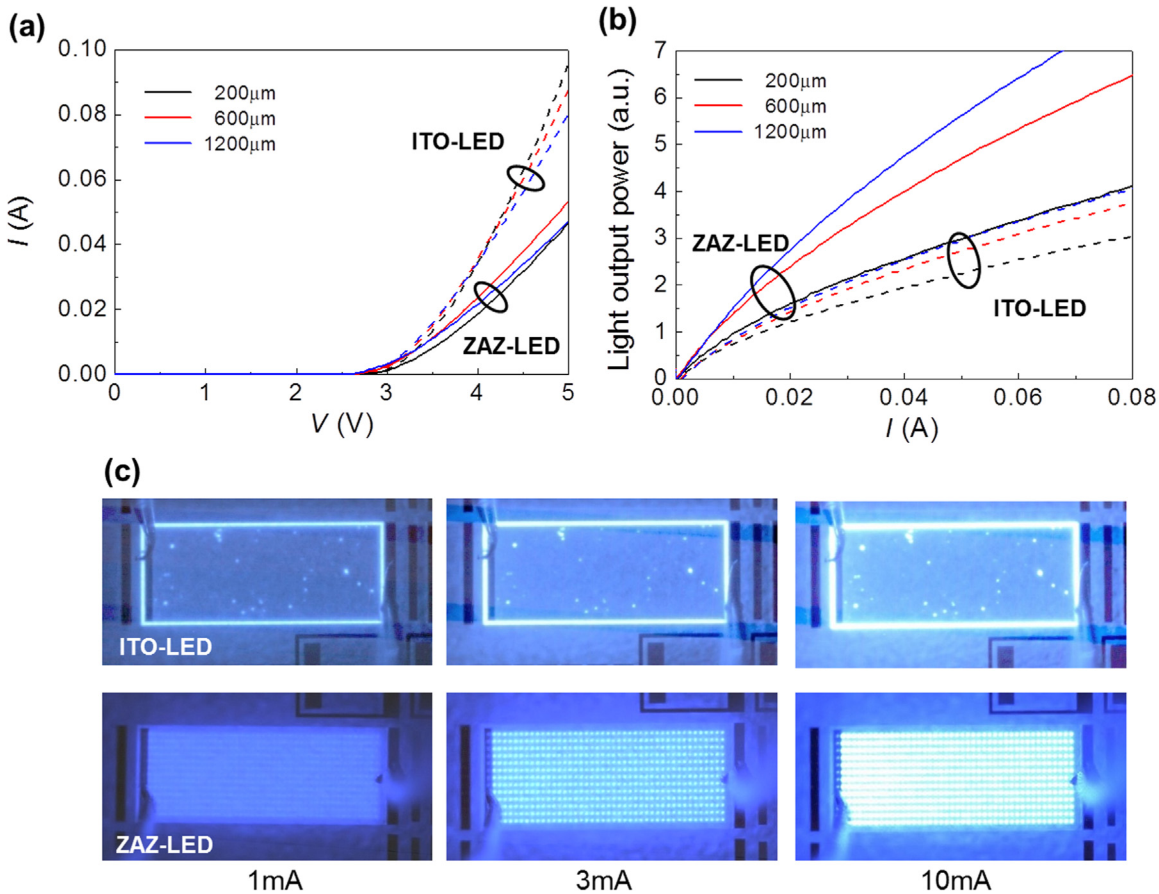

Fig. 2. (a) I–V curves of the ZAZ-LEDs and ITO-LEDs with various lateral mesa lengths. (b) Optical output power versus injection current for ZAZ-LEDs and reference LEDs with various lateral mesa lengths. (c) Optical images of LED top surfaces with various injection currents ranging from 1 mA to 10 mA; the lateral mesa length in all LEDs is $L=1200$ μm.

current spreader among contact islands on p-GaN. ZAZ-LEDs show the optical output powers over 25% higher than those of the reference ITO-LEDs with increasing L. The EL images also show the notable brightness of a ZAZ-LED compared to a reference ITO-LED at various injection currents as shown in Fig. 2(c).

To investigate the current spreading property in ZAZ-LEDs, we fabricated devices with various L ranging from 100 to 1200 μm and measured the light output power during operation at an injection current of 100 mA (Fig. 3). To obtain the current spreading length (L_s), defined as the length over which the current density drops to the 1/e value at the mesa edge, the current densities (J) of both LEDs were plotted as a function of the forward voltage with various L, as shown in Fig. 3(a) where the area is the total TCL area, which is the same in both types of LEDs. Using the experimental J–L characteristics plotted as a function of the bias voltage, the L_s was estimated by theoretical fitting of the experimental J–L data [21]. Fig. 3(b) shows the L_s values obtained as a function of the bias voltage. It is evident that the L_s values of the ZAZ-LEDs are longer than those of the reference ITO-LEDs, which is consistent with the EL images as shown in Fig. 2(c). Therefore, the enhanced optical output power of ZAZ-LEDs may be due to the improved current spreading. However, the L_s values are likely to be underestimated in an LED with a long L. This is attributed to the lack of an exact theoretical model to describe our LED samples. The model is only valid when the vertical p-layer resistances, including the p-contact and the p-GaN, are small enough to be negligible [21], whereas that of our samples is not small but has a significant bias voltage dependence. The better electrical and optical performance observed for ZAZ-LEDs and their longer L_s values suggest that the ZAZ layer can be used as a TCL in large-area devices. The suitability of ZAZ electrodes for large-area devices is further confirmed by their area dependence of light output power as shown in Fig. 3(c). At a constant current injection of 100 mA, the optical output power increased with increasing device area (or L), which is essentially due to the reduced efficiency droop [22]. Interestingly, the light output powers of the ZAZ-LEDs increased rapidly up to a level of $L \leq 400$ μm and changed insignificantly at $L \geq 400$ μm, whereas those of the reference ITO-LEDs slowly increased with increasing L up to 1200 μm. The efficiency droop, the loss of external quantum efficiency at high injection current, was also found to be quite small in the ZAZ-LEDs as shown in Fig. 3(d). For example, the efficiency droop values measured from the maximum peak intensity to those measured at 80 mA were 47.0% and 60.8% for ZAZ-LEDs and reference ITO-LEDs, respectively. Several studies [23–28] have reported various origins of efficiency droop, such as crystallographic defects [23], Auger recombination [24], electron leakage [25], current crowding [26], and heating effects [27]. Among these origins, the effect of defects can be excluded because the same LED wafers were used in this study. The two origins of Auger recombination and electron leakage can also be excluded because the efficiency droop of ZAZ-LEDs was greatly alleviated despite their much higher current density are expected due to the reduced contact area. The heating effect also cannot be the origin of the reduced efficiency droop in this study because ZAZ-LEDs have a higher contact resistance than reference ITO-LEDs. Therefore, a reasonable explanation for the reduced efficiency droop is the enhanced current spreading, which would be responsible for the improved optical output power.

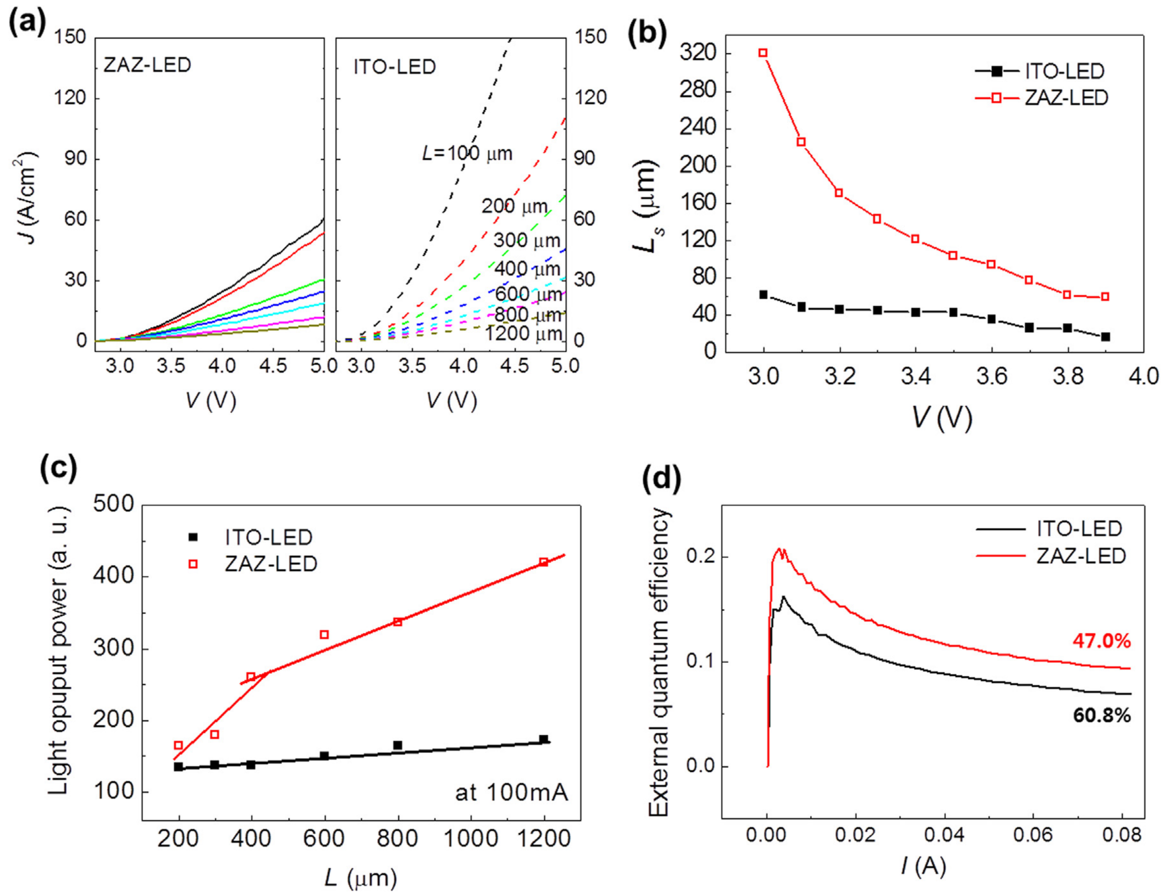

Fig. 3. (a) J–V curves of ZAZ-LEDs and reference ITO-LEDs various lateral mesa lengths (L). (b) L_s values obtained for both LED types as a function of the bias voltage. (c) Area dependence of the optical output power of both LED types. (d) External quantum efficiency versus current for both LED types.

4. Conclusion

In summary, we demonstrated sputter-deposited ZAZ layer as a TCL on GaN blue LEDs. The evaporated ITO buried contact islands on the p-GaN top surface in the ZAZ layer were employed in order to overwhelm the problem of an ohmic contact failure due to the sputtering process. It was found that LEDs using ZAZ layer as a TCL showed a higher light output power due to the good current spreading property of the ZAZ layer than that of the reference ITO-LEDs even in large devices.

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