Light extraction efficiency enhancement of GaN-based light emitting diodes by a ZnO current spreading layer*

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Abstract: Gallium nitride (GaN) based light emitting diodes (LEDs) with a thick and high quality ZnO film as a current spreading layer grown by metal-source vapor phase epitaxy (MVPE) are fabricated successfully. Compared with GaN-based LEDs employing a Ni/Au or an indium tin oxide transparent current spreading layer, these LEDs show an enhancement of the external quantum efficiency of 93% and 35% at a forward current of 20 mA, respectively. The full width at half maximum of the ZnO (002) ω-scan rocking curve is 93 arcsec, which corresponds to a high crystal quality of the ZnO film. Optical microscopy and atomic force microscopy are used to observe the surface morphology of the ZnO film, and many regular hexagonal features are found. A spectrophotometer is used to study the different absorption properties between the ZnO film and the indium tin oxide film of the GaN LED. The mechanisms of the extraction quantum efficiency increase and the series resistance change of the GaN-based LEDs with ZnO transparent current spreading layers are analyzed.

Key words: GaN; LED; ZnO; extraction efficiency; transparent electrode

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1. Introduction

GaN-based blue light emitting diodes (LEDs) are attractive for lighting and display applications due to their high brightness and high power capability. Generally, increasing the internal quantum efficiency (IQE) is an important approach to improve the external quantum efficiency (EQE). But there is an obvious gap, called light extraction efficiency (LEE), between the IQEs of GaN-based blue LEDs and their EQEs. The reason for this difference is the difficulty for the light to escape from the high refractive index semiconductor. For the refractive indexes of GaN (2.5 at 460 nm), the escape angle for internal light is only ~23°, as imposed by Snell's law. This narrow escape cone for spontaneous emission covers a solid angle of $\sim (1/4n^2)4\pi$. Only 4% of the internally generated light can escape[1, 2]. To further improve the performance of GaN-based LEDs, much effort has made to improve the light extraction efficiency: chip shaping[3, 4], surface roughness[5], patterned sapphire substrate[6], transparent electrode[7], high reflection film[8], photonic crystal[9], etc. So, a material that has a high refractive index, a low absorption, and is easy to make is required[10].

ZnO is a promising material for optoelectronic devices, as it has a wide band gap (3.37 eV), a high refractive index (~2.1 at 460 nm), a large excitonic binding energy (60 meV), and a high optical gain (300 cm^{-1})[11]. It provides a high transparency in the blue region. GaN and ZnO have the same crystal structure, very similar lattice constants, and little difference in their in-plane linear thermal expansion coefficients. ZnO grown on a GaN layer has a higher crystallinity than that grown on a sapphire substrate[12]. Also, ZnO can be selectively etched in various acids, allowing many structures to be created[13]. The growth of ZnO has been investigated by various growth methods, such as chemical vapor deposition (CVD), metal-organic chemical vapor deposition (MOCVD), and liquid-phase-deposition (LPD). These growth processes are complicated or expensive. Some of them use a high temperature, or have a very slow growth process. Compared with these methods, the metal-source vapor phase epitaxy (MVPE) method[14] has many advantages, such as an acceptable temperature, a high growth rate (>120 μm/h), a good crystal quality, a good thickness uniformity, and low cost. There are some groups that use ZnO transparent layers for GaN-based LEDs and achieve EQEs of about 20%[15].

In this study, an MVPE method is employed to grow a high crystalline ZnO thick film on a GaN-based blue LED structure at about 750 °C. A great enhancement of the EQE with a ZnO current spreading layer was achieved and measured. The mechanism of the EQE increase is analyzed.

2. Experiment

The GaN-based LED in this study was grown on a sapphire substrate by MOCVD. It consists of a GaN buffer layer and a 2 μm Si-doped layer, 5-period InGaN–GaN quantum wells (QWs) as the active region for the blue LED (460 nm), followed by 200 nm magnesium-doped GaN layers. The wafer was annealed in a N_2 atmosphere at 750 °C for 30 min to acti-

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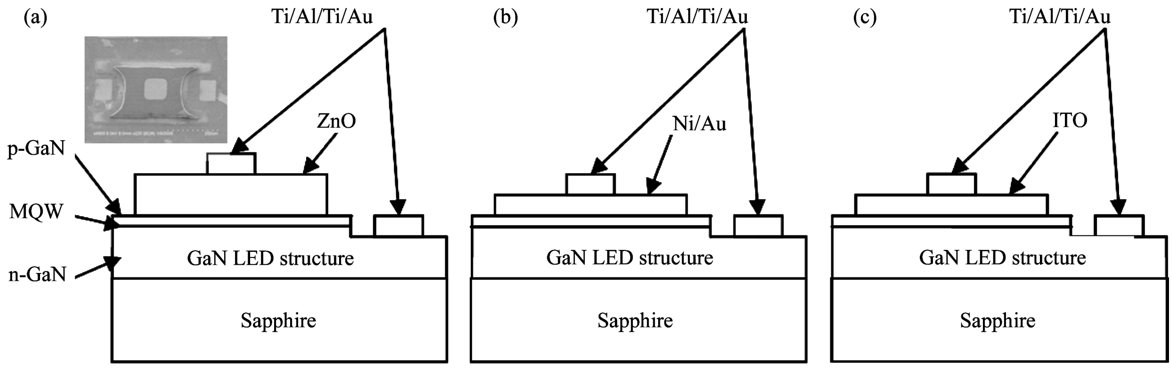

Fig. 1. Schematic structure of the GaN-based LEDs with (a) ZnO (8 μm), (b) Ni/Au (5 nm/5 nm), and (c) ITO (200 nm) as the transparent electrode.

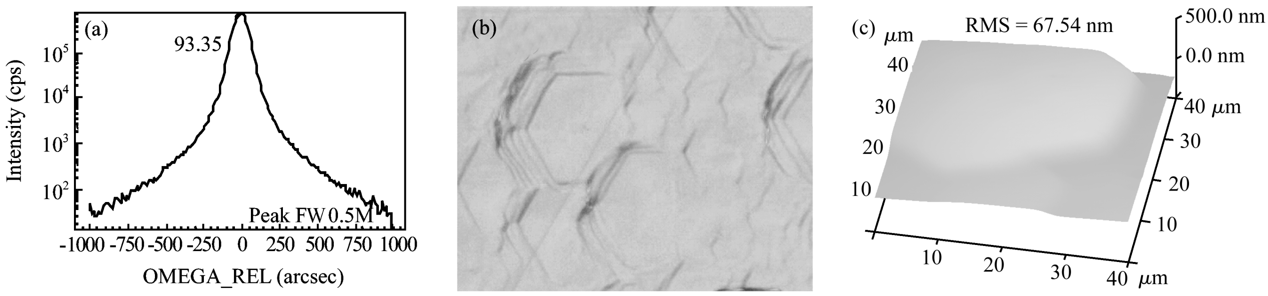

Fig. 2. Characteristics of the ZnO layers: (a) Rocking curve of ZnO (002); (b) Microscopic image of the ZnO surface; (c) AFM views of the ZnO surface.

vate the Mg in the p-GaN layer.

The sample was separated into three parts, A, B, and C, to fabricate LED chips. The same set of photo-masks was used, and the chip area was 500 × 300 μm^2. The schematic structures are shown in Figs. 1(a), 1(b), and 1(c).

For sample A, using N_2 as a carrier gas and Zn and H_2O vapor as the Zn and O sources, a ZnO film was grown on the top of the GaN-based LED structure. The substrate temperature and growth time was kept at 750 °C and 10 min. The thickness of the ZnO film is 8 μm. The chemical reaction in the process is as follows[12]:

$$Zn + H_2O \leftrightarrow ZnO + H_2.$$

ZnO patterning was achieved by etching the undesired ZnO film with a dilute HCl solution at a temperature of about 50 °C, with a 600-nm SiO_2 mask, which was deposited by plasma enhanced chemical vapor deposition (PECVD). Then an inductively coupled plasma (ICP) process was performed to explore the n-GaN layer. The Ti/Al/Ti/Au film was sequentially electron beam evaporated on the ZnO and the n-GaN as an electrode.

For sample B, a standard Ni/Au semi-transparent electrode process was used as follows: the n-type mesa was defined by an ICP, and then a Ni/Au (5 nm/5 nm) layer was evaporated onto the p-GaN and annealed in O_2 to serve as the p-contact. A Ti/Al/Ti/Au stack was deposited onto the exposed n-GaN layer to serve as the electrode. The schematic structure is shown in Fig. 1(b).

For sample C, a common ITO transparent electrode process was used as follows: the n-type mesa was defined by an ICP process. An ITO film (about 200 nm) was evaporated by an e-beam, and the ITO pattern was defined by etching the undesired part with HCl : HNO_3 : H_2O (4 : 1 : 1). After annealing, a Ti/Al/Ti/Au stack was evaporated by an e-beam as an electrode. The final structure is shown in Fig. 1(c).

The morphology and the cross section of the ZnO film were studied by an Olympus MX51 microscope, an AFM (Dimention-3100), and an SEM (Hitachi S-4800). The sheet resistance was tested by a four-probe method. The transmission spectrum was measured by a Cary 5E spectrophotometer. The ZnO (002) ω-scan rocking curve was measured by a Bede D1 HRXRD. After the chip processes were finished, the three samples were polished, scribed, and packaged. The I–V characteristic, the EL spectrum, and the luminous flux of 20 LEDs were tested by an Everfin-PMS50.

3. Results and discussion

The ω-scan rocking curve is shown in Fig. 2(a). From the figure we can get the full width at half maximum (FWHW), which is only 93 arcsec, corresponding to a high crystalline quality[12]. The surface feature is shown in Figs. 2(b) and 2(c). In the microscope image there is no flat and smooth surface of large scale. The images show many regular hexagonal features, which may come from the high growth speed of about 1 μm/min during the MVPE growth process[14]. The AFM image shows that the roughness is 67.54 nm, but the surface on top of the mesa is just several nanometers. So, the light from the active layer will have more chance to go through this surface morphology into the open[15].

The ZnO film provides a higher transparency in the blue

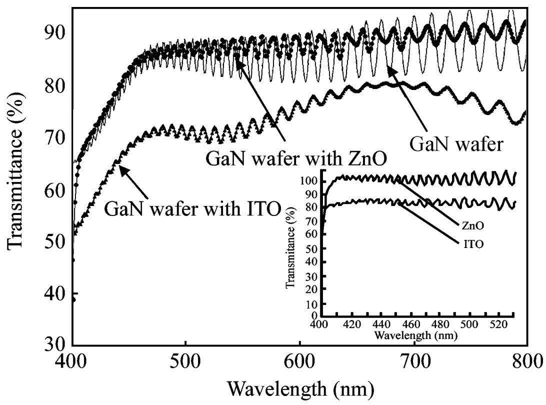

Fig. 3. Transmittance spectra of ZnO (~8 μm) and ITO (~200 nm) on GaN.

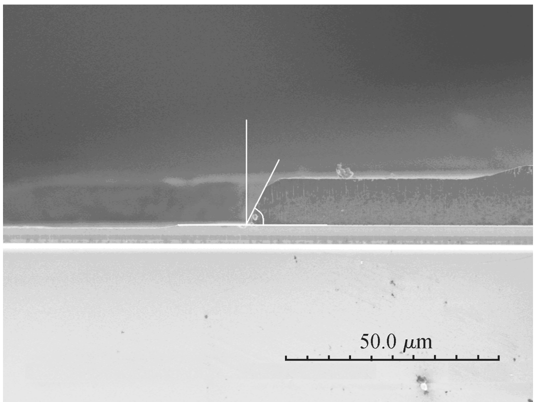

Fig. 4. SEM picture of the cross section of an etched ZnO film.

wavelength region than the ITO film. Figure 3 shows the transmission spectrum of the GaN substrate without any layer, with a ZnO film, and with an ITO film. It is obvious that ZnO has a much lower absorption loss than ITO. At 460 nm, the transmittance of the GaN substrate with the ITO is only about 70% (the transmittance of ITO film at 460 nm is about 85%, shown in the small figure), whereas the transmittance of the GaN substrate with the ZnO can reach ~85%, i.e., a value very similar to the transmittance of the GaN substrate itself (the transmittance of ZnO film at 460 nm is above 95%). These results suggest that ZnO has a very low absorption coefficient and it is indeed optically suitable to serve as the upper transparent current spreading layer of blue LEDs.

Figure 4 shows the cross section of an etched ZnO film. It can be seen that the ZnO etch process using a HCl solution forms a declivity sidewall in the ZnO film. The angle between the sidewall and the surface is about 60°. It is considered that the exposed ZnO sidewall plane is the $(10\bar{1}\bar{1})$ plane of ZnO. This structure of the sidewall will partly change the light orientation in the LED plate and also improve the LEE[15].

The room-temperature electroluminescence (EL) spectra as a function of the wavelength for samples A, B, and C under a forward current of 20 mA are shown in Fig. 5. The EL measurement was done at room temperature using a dc current. The EL intensity of the GaN-based LED with ZnO film clearly increases. At the same time, the peak wavelength of the LED with a ZnO electrode does not move. This indicates that the ZnO growth temperature in the MVPE caused no serious defects in the LED active layer.

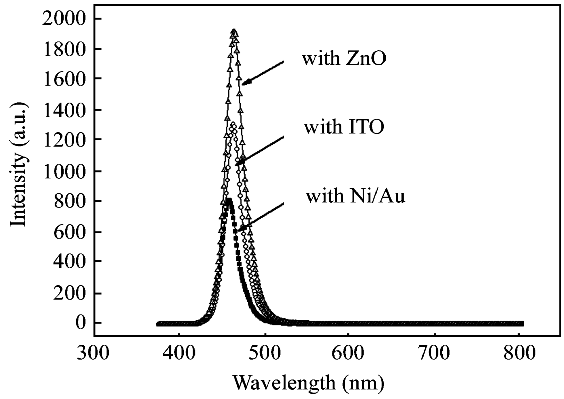

Fig. 5. EL spectra of LEDs with ZnO, ITO, and Ni/Au current spreading layers at 20 mA.

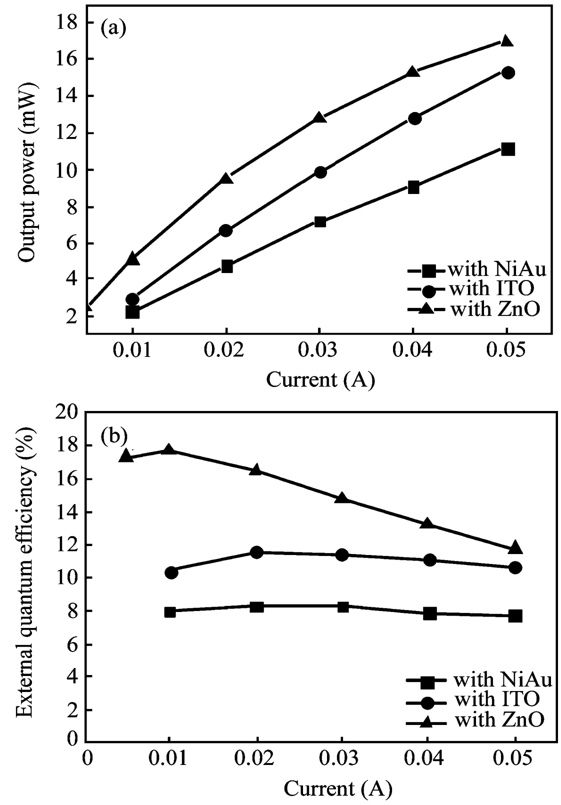

Fig. 6. (a) Output power and (b) external quantum efficiency of GaN-based LEDs with Ni/Au, ITO, and ZnO current spreading layers at forward currents from 5 to 50 mA.

The optical output power of samples A, B, and C as a function of forward current is shown in Fig. 6(a). The measurements were done in an integrated sphere under dc current injection conditions. The highest EQE is about 18% at 10 mA. At an injection current of 20 mA, the output power of the sample A shows a 93% and 35% improvement compared with samples B and C. Each sample gives an increasing light output power from 5 or 10 to 50 mA. The output power of the samples B and C at 5 mA is lower than the resolution of the test equipment. Figure 6(b) displays the EQE of samples A, B, and C as a function of the forward current. The EQE of the LED with a ZnO layer obviously reduces when the injection current increases over 10 mA. Compared to the Ni/Au and ITO

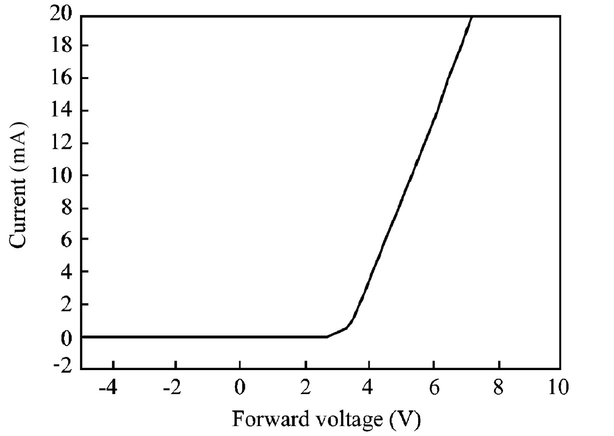

Fig. 7. *I–V* characteristics of the LED with a ZnO transparent layer.

samples, the higher series resistance of the LEDs with a ZnO current spreading layer generates more Joule heat; so, these LEDs work at a higher temperature, and their IQE and EQE are clearly reduced[16].

The current versus voltage relationship of sample A is shown in Fig. 7. No significant leakage current is visible at –5 V. Compared with a common ITO electrode GaN-based LED, a much higher series resistance of approximately 200 Ω is observed. This is attributed to the high resistance of the non-doped ZnO film and/or the ZnO/p-GaN contact resistance. The typical electrical resistance of our MVPE undoped ZnO film is about 0.5 Ω·cm. It implies that the serial resistance of the LED with a ZnO electrode (Sample A) is due to the junction contact resistance. A heavy doped interlayer may reduce it[17].

4. Conclusions

In summary, high quality ZnO layers are successfully grown on GaN-based light emitting diodes by MVPE. The light emitting diodes are fabricated with these ZnO films as current spreading layers. The external quantum efficiency of the LEDs with a ZnO transparent current spreading layer at 20 mA is increased by 35% and 93% compared to the LEDs using indium tin oxide or Ni/Au layers, respectively. The full width at half maximum of the ZnO (002) ω-scan rocking curve is 93 arcsec, which corresponds to a high crystal quality. An AFM is used to observe the surface morphology of the ZnO film, and many regular hexagonal features are found. The absorption properties between ZnO and indium tin oxide films are studied by a spectrophotometer. An acceptable temperature, a high growth rate, and a low cost of the MVPE method are very important to fabricate GaN-based LEDs with ZnO transparent current spreading layers. The reasons for the light extraction efficiency increase and the change of the series resistance of the light emitting diodes with ZnO layers are analyzed. In future studies, the problem of the high series resistance of the ZnO film will be investigated.

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