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Rapid thermal annealing effects on vacuum evaporated ITO for InGaN/GaN blue LEDs*

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Abstract: 8 mil × 10 mil InGaN/GaN blue LEDs with indium tin oxide (ITO) emitting at 460 nm were fabricated. A vacuum evaporation technique was adopted to deposit ITO on P-GaN with thickness of 240 nm. The electrical and optical properties of ITO films on P-GaN wafers, as well as rapid thermal annealing (RTA) effects at different temperatures (100 to 550 ℃) were analyzed and compared. It was found that resistivity of 450 ℃ RTA was as low as 1.19×10^{-4} $\Omega\cdot$cm, along with a high transparency of 94.17% at 460 nm. AES analysis indicated the variation of oxygen content after 450 ℃ annealing, and ITO contact resistance showed a minimized value of 3.9×10^{-3} $\Omega\cdot$cm^2. With 20 mA current injection, it was found that forward voltage and output power were 3.14 V and 12.57 mW. Furthermore, maximum luminous flux of 0.49 lm of ITO RTA at 550 ℃ was measured, which is the consequence of a higher transparency.

Key words: rapid thermal annealing; light emitting diodes; ITO; InGaN/GaN

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1. Introduction

GaN-based blue light-emitting diodes (LEDs) have recently attracted much attention for their versatile applications. As is well known, a transparent Ohmic contact to P-GaN is essential in fabricating high-brightness GaN-based LEDs. However, poor ohmic contact at the metal/P-GaN interface is a problem that leads to LEDs with limited performances[1−3]. To this effect, indium tin oxide (ITO) is used as a contact to P-GaN because of its unique characteristics such as effective current spreading, good conductivity and high optical transparency in the blue wavelength range[4, 5]. However, the poor Ohmic nature between the ITO and high resistive p-GaN contact layers still remain a considerable problem. Margalith *et al.*[6] presented the ITO contact with p-GaN, but it had a forward voltage that was too high for LEDs. Recently, many approaches have been taken to try to enhance this Ohmic property, for example, applying an InGaN/GaN super lattice structure[7,8] or an InGaN capping layer on p-GaN[9], or thermally diffusing Au and then removing the thin layer[10]. However, these methods suffer from photon loss at the blue spectra. In this paper, rapid thermal annealing at different temperatures (100 to 550 ℃) was used to obtain high quality ITO films, and the effects of annealing on the properties of the films were investigated. The vacuum evaporation technique is one of the most commonly deposition methods used to deposit ITO films on P-GaN[11]. The square transmission line model (s-TLM) was adopted to measure the specific contact resistance of the ITO/p-GaN contact and the distribution of In, Sn, O, Ga in the interface are characterized by Auger electron spectroscopy (AES). Furthermore, nitride-based LEDs were fabricated by using ITO as the upper contact layers. The current–voltage (I–V), output power and intensity–current (L–I) characteristic of the fabricated LEDs will be reported.

2. Experiment

InGaN–GaN LED wafers with a peak wavelength at around 460 nm were grown on sapphire (Al_2O_3) substrates by metal organic vapor phase epitaxy. Details of the growth procedures and epitaxial structure can be found elsewhere[12−15]. The fabrication process began by first cleaning the GaN epiwafer with acetone, alcohol and then deionized water. Next, ITO films were deposited onto P-GaN by evaporation under a vacuum press of 6×10^{-4} Pa, followed by 350 ℃ in O_2. The deposition rate was 24 nm/min, and the ITO thickness was chosen to be 240 nm. The deposited ITO films were then rapid thermal annealed at various temperatures for 30 min in a N_2 atmosphere. In order to measure the transparency of the ITO, the films were also deposited onto glass substrates under the same conditions. The transparency spectra of the deposited films were measured by using a U-4100 spectrophotometer. Van der Pauw resistivity measurements were used to determine the contact resistance of ITO on P-GaN by using a four feet probe, and carrier concentration and hall mobility were also obtained.

Nitride-based blue LEDs were fabricated. The n-GaN was exposed by an inductively coupled plasma (ICP) etcher using Cl_2/Ar as the etching gases. Ni/Au was subsequently evaporated on to P-GaN to form a P-type electrode, while the n-type

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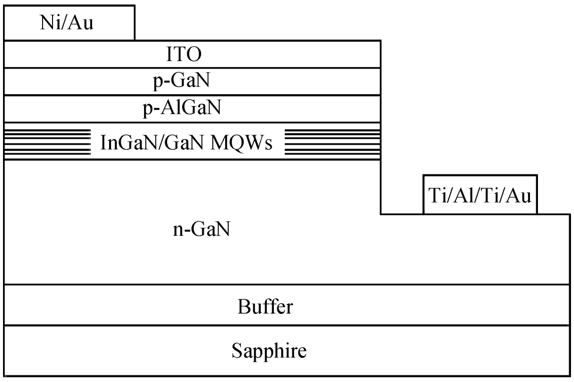

Fig. 1. Schematic diagram of the epitaxial layers and LED structures.

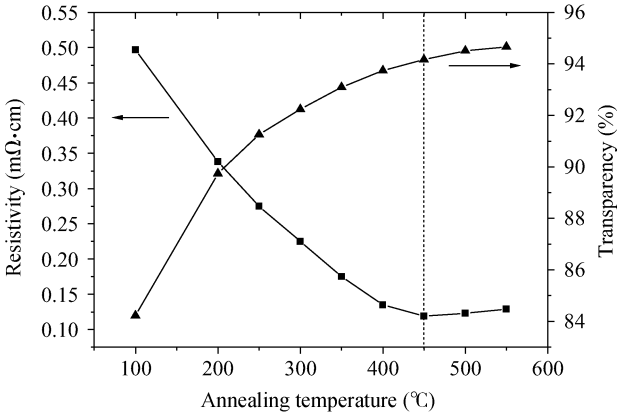

Fig. 2. Optical transparency (λ = 460 nm) and resistivity of ITO for different annealing temperatures.

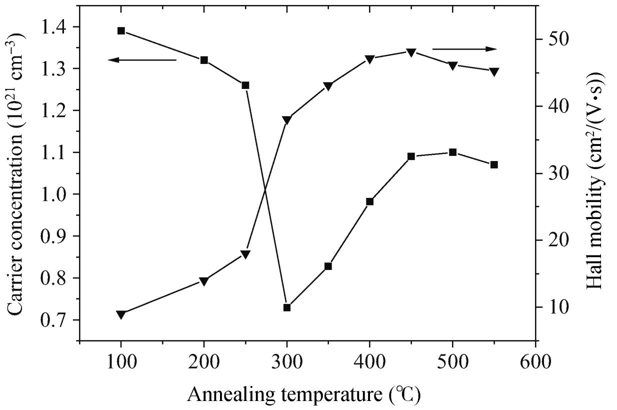

Fig. 3. Carrier concentration (n) and Hall mobility (μ) of ITO for different annealing temperatures.

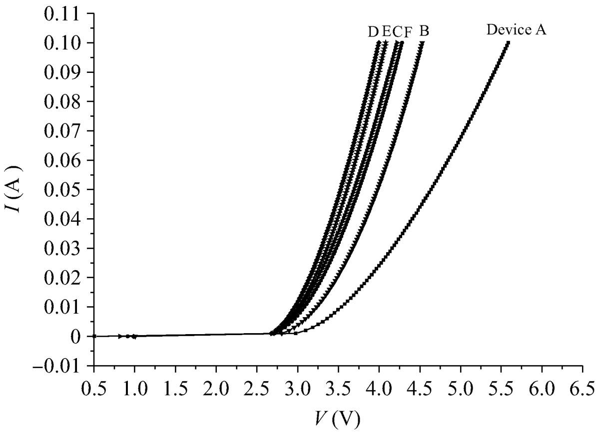

Fig. 4. I–V characteristics of ITO contacts annealed at various temperatures on P-GaN.

electrode was Ti/Al/Ti/Au. Figure 1 schematically depicts the epitaxial layers and LED structures fabricated on sapphire substrates. The sapphire substrates were polished down to 90 μm and then scribed and broken to 8 mil × 10 mil InGaN/GaN LED chips. After bonding to the tube, these packaged chips were measured by using a 4200 Semiconductor Characterization System under a nominal current of 20 mA and I–V curve was fitted. Luminous flux and output power were then measured using LED-spec.

3. Results and discussion

Figure 2 shows the variation of transparency of ITO films in the visible region ($\lambda = 460$ nm) and resistivity as a function of annealing temperature. The transparency was normalized with respect to the glass substrate. It is observed that the transparency increases as annealing temperature increased from 100 to 550 ℃. The transparency of ITO films is as high as about 94.66% at 550 ℃. Such a result can be attributed to the fact that the stress of ITO films is released after RTA[16]. Furthermore, the high transparency under high temperature suggests improved structural crystallinity and better oxidation[17]. The effect of annealing on the resistivity of ITO films is also shown in the graph. The resistivity was initially decreased from 4.97×10^{-4} to 1.19×10^{-4} Ω·cm with annealing temperature up to 450 ℃. Low resistivity values of 1.19×10^{-4} Ω·cm were achieved after annealing at 450 ℃ and the transparency is 94.17% which is only 0.49% smaller than that of 550 ℃. The improvement of the conductivity of the ITO films was the result of induced dislocations and compound formation[18]. However, the resistivity increases to 1.29×10^{-4} Ω·cm at higher temperature (550 ℃). This phenomenon means that the properties of ITO degraded when the annealing temperature was too high. We deduce that more oxygen accumulated at high temperature, and an oxygen-rich interfacial layer was formed. From the two curves in Fig. 1, it is found that the RTA of ITO films at 450 ℃ would be a relatively appropriate temperature.

Figure 3 shows the variation of carrier concentration (n) and Hall mobility (μ) with annealing temperature. The carrier concentration, which initially decreases from 1.39×10^{21} to 7.29×10^{20} cm^{-3}, later increases when the annealing temperature exceeds 300 ℃. The appreciable change may be due to ionized tin and the role of the compensation mechanism. As Figure 3 shows, Hall mobility increases with annealing temperature and then remains almost unchanged. It also confirmed that the crystallinity is improved after annealing because of the reduced grain boundary and scattering[19].

Figure 4 shows forward-biased I–V curves of fabricated LEDs with Ni/Au and ITO contact annealed at five different temperatures. The six LEDs were denoted as device A with Ni/Au contact and B, C, D, E, F of ITO RTA at 350, 400, 450, 500, 550 ℃, respectively. It can be seen that the LED for-

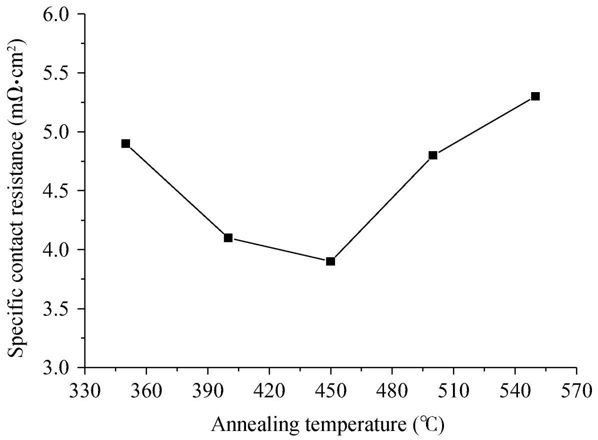

Fig. 5. Specific contact resistance of the p-pad/ITO/p-GaN interface as a function of temperature.

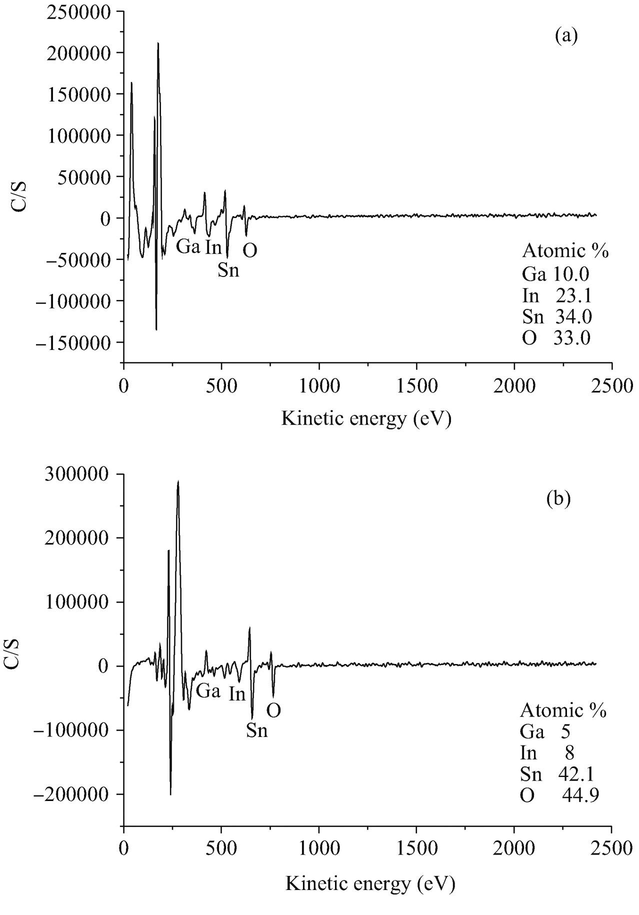

Fig. 6. AES spectra of ITO/GaN interface (a) before and (b) after 450 ℃ annealing.

ward voltage measured at 20 mA current injection was 3.88, 3.48, 3.25, 3.14, 3.21, 3.31 V for devices A, B, C, D, E, F. The 0.74 V reduction in V_F for the device D than device A could be explained by improved ITO contact to P-GaN. The lower forward voltage observed from device D should be attributed to the lowest resistivity and higher carrier concentration as confirmed above. As a result, device D with ITO annealed at 450 ℃ retains excellent ohmic and current spreading characteristics.

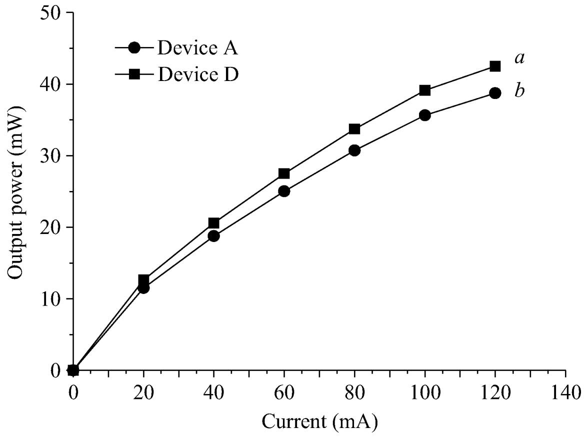

Fig. 7. Output power as a function of current of GaN LEDs with (*a*) ITO annealing at 450 ℃ and (*b*) Ni/Au contact.

Table 1. Comparative data for GaN-based LEDs with Ni/Au and ITO contacts to P-GaN.

LED with ITO	Flux @ 20 mA (lm)	Transparency @ $\lambda = 460$ nm (%)
Before RTA	0.2841	82.13
RTA at 350 ℃	0.3387	93.09
RTA at 400 ℃	0.3961	93.74
RTA at 450 ℃	0.4523	94.17
RTA at 500 ℃	0.4734	94.51
RTA at 550 ℃	0.4859	94.66

The specific contact resistances of p-pad/ITO/p-GaN interface were obtained by using the square transmission line model (s-TLM). As shown in Fig. 5, it was found that the specific contact resistances decreased from 4.9×10^{-3} Ω·cm^2 at 350 ℃ to 3.9×10^{-3} Ω·cm^2 at 450 ℃, then increased to 5.3 $\times$ 10^{-3} Ω·cm^2 at 550 ℃. Such a result suggests that carrier transport was dominated by thermionic emission for the sample. In other words, measured specific contact resistances were strongly dependent on the annealing temperature. Comparing Fig. 3 with Fig. 5, it is found that variation tendency of carrier concentration is in accordance with the specific contact resistance. This seems to suggest that carrier transport determined the variation of the contact resistance. Auger electron spectroscopy (AES) measurements were taken to analyze the distribution of In, Sn, O, Ga in the interface before and after 450 ℃ annealing, as shown in Fig. 6. We observe that the diffusion of In, Sn and O into GaN and the diffusion of Ga into ITO after the annealing. It is supposed that, during the contact annealing, gallium atoms diffuse out from the GaN substrate to the ITO contact and create gallium vacancies, and indium atoms diffuse from the ITO contact to gallium vacancy sites in addition to the diffusion of Sn and O to GaN, therefore, forming a mixed interfacial layer composed of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}$, $\mathrm{In}_x\mathrm{Ga}_y\mathrm{O}_z$, etc., at the ITO/GaN interface[20]. Furthermore, the oxygen content is found to be higher when the sample is annealed at 450 ℃. Thus, the phenomenon that electrical properties degrade can be explained by the AES analysis.

Figure 7 shows the intensity–current (L–I) characteristics of devices A and D. It can be seen that device D of ITO RTA at 450 ℃ achieved higher output power. At 20 mA, output powers of 11.53 and 12.57 mW for devices A and D are obtained. It also can be seen that the output power of the two devices increase with increasing current injection. Table 1 shows the luminous flux for GaN-based LEDs with ITO and Ni–Au contacts, respectively. The luminous flux of ITO contact RTA at 550 ℃ increase of 71% is higher than that of Ni/Au contact, which should be ascribed to higher transparency.

4. Conclusion

InGaN/GaN blue LEDs emitting at 460 nm with Ni/Au and ITO RTA at different temperatures were fabricated. The effects of annealing on the electrical and optical properties have been investigated. It was found that 450 ℃ RTA ITO (240 nm) formed good ohmic contact on P-GaN with a minimum value of resistivity of 1.19×10^{-4} Ω·cm, which should be due to high carrier concentration and mobility. At 20 mA current injection, it was found that the forward voltage and output power were 3.14 V and 12.57 mW for LED with 450 ℃ RTA ITO. The increase in optical transparency with temperature should contribute to the highest luminous flux at 550 ℃.

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