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Effect of graphene/ZnO hybrid transparent electrode on characteristics of GaN light-emitting diodes*

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In order to reduce the Schottky barrier height and sheet resistance between graphene (Gr) and the p-GaN layers in GaN-based light-emitting diodes (LEDs), conductive transparent thin films with large work function are required to be inserted between Gr and p-GaN layers. In the present work, three kinds of transparent conductive oxide (TCO) zinc oxide (ZnO) films, Al-, Ga-, and In-doped ZnO (AZO, GZO, and IZO), are introduced as a bridge layer between Gr and p-GaN, respectively. The influence of different combinations of Gr/ZnO hybrid transparent conducting layers (TCLs) on the optical and thermal characteristics of the GaN-LED was investigated by the finite element method through COMSOL software. It is found that both the TCL transmittance and the surface temperature of the LED chip reduce with the increase in Gr and ZnO thickness. In order to get the transmittance of the Gr/ZnO hybrid TCL higher than 80%, the appropriate combination of Gr/ZnO compound electrode should be a single layer of Gr with ZnO no thicker than 400 nm (1L Gr/400-nm ZnO), 2L Gr/300-nm ZnO, 3L Gr/200-nm ZnO, or 4L Gr/100-nm ZnO. The LEDs with hybrid TCLs consisting of 1L Gr/300-nm AZO, 2L Gr/300-nm GZO, and 2L Gr/300-nm IZO have good performance, among which the one with 1L Gr/300-nm GZO has the best thermal property. Typically, the temperature of LEDs with 1L Gr/300-nm GZO hybrid TCLs will drop by about 7 K compared with that of the LEDs with a TCL without ZnO film.

Keywords: finite element methods, light-emitting diodes, graphene, ZnO

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1. Introduction

As a new generation of solid-state light sources, the GaN-based light-emitting diode (GaN-LED) has been extensively studied for its low energy consumption, long life, and environmental friendliness. Currently, the GaN-LED is widely used in information display panels, general lighting, and background light sources.[1] The transparent conducting layer (TCL) in LEDs facilitates current spreading and uniform light emission. Indium tin oxide (ITO) is currently the most widely used TCL material on the market.[2] However, due to the high cost of indium (In), the instability in an acid-based environment, and the high temperature of ITO preparation,[3,4] it is urgent to look for an alternative TCL material with high performance and a reasonable price. Two-dimensional (2D) graphene (Gr) possesses excellent photoelectric properties,[5] extraordinary thermal conductivity, and splendid conductivity,[6,7] and therefore it has aroused widespread attention in recent years. As a TCL film in the GaN-LED, Gr can increase current spreading and transmission characteristics, greatly reduce the current crowding phenomenon, and improve the uniformity of light emission.[8,9] However, owing to the large difference of work function between Gr (4.4 eV) and p-GaN (7.5 eV) layers,[10] the direct contact between Gr and p-GaN will result in a large Schottky barrier height and a high sheet resistance. Some studies have shown that the introduction of metal or metal oxide as an intermediate layer between Gr and p-GaN can effectively reduce the overall current transfer resistance.[11–13] Since the metal oxide film has a higher light transmittance than the metal in the visible light range, the transparent conductive oxide (TCO) metal oxide films are more often used as an intermediate layer.[14] In our previous work, ITO and NiO_x were tried, and it was found that the thermal and electrical

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performance of LEDs can be improved by the optimization of the combination of NiO_x/Gr and ITO/Gr. Due to low resistivity, high transmittance, nontoxicity, good stability in hydrogen plasma processes, and resource availability,[15,16] zinc oxide (ZnO) is usually regarded as an alternative transparent electrode to ITO. Therefore, in the present work, we intend to introduce ZnO as a bridge layer between Gr and p-GaN, and investigate the effect of Gr/ZnO compound films on LED performance. Generally, the undoped ZnO film has a low carrier concentration and a high resistivity, and thus In, aluminum (Al), and gallium (Ga) are often applied to dope ZnO to improve the conductivity.[17–19] Therefore, three kinds of ZnO, i.e. Al-,Ga-, and In-doped ZnO (AZO, GZO, and IZO, respectively), are observed in this paper. Furthermore, the influence of the thickness of the ZnO and Gr layers on the performance of the LED and the optimal combination of Gr and ZnO are also studied.

In our previous work, the thermal and electrical property of NiO_x/Gr-LED,[20] ITO/Gr-LED,[21] and metal/Gr-LED[5] were calculated by COMSOL simulation software. The calculated forward voltages of NiO_x/Gr-LED and ITO/Gr-LED were compared with those measured by Zhang *et al.*[12] and Xu *et al.*[22]. It is found that the relative errors between our calculated and experiment results are 6.50%[20] and 4.94%, respectively,[21] indicating that our simulation method is accurate and reliable. Therefore, in our present work, we adopt this method to study the influence of three kinds of ZnO/Gr hybrid structures (Gr/AZO, Gr/GZO, and Gr/IZO) on LED performance. To meet the requirement of transparency, the thickness range of possible combinations of Gr and ZnO was firstly calculated by the RF module, and then the temperature distribution of the LED chips with various Gr/ZnO composite TCLs was obtained by the Joule heat module. Finally, the optimal combination of TCLs was obtained by analyzing the thermal and optical properties of the LEDs.

2. Simulation

The semiconductor module, RF module, and Joule heat module in COMSOL Multiphysics software were adopted in this work.

2.1. Model structure

Numerical simulations for GaN-LED have been proposed to overcome many technical challenges, including the thermal management and the current crowding phenomenon.[5,23] Using the three-dimensional (3D) finite element method in the simulation software COMSOL, we can establish the geometric model of the GaN-LED chip. Figure 1 shows a schematic diagram of a typical GaN-LED chip whose size is 300 μm × 300 μm.[24,25] The physical parameters of various materials are listed in Table 1.

Table 1. Physical parameters of various materials.

Material	Sapphire	u-GaN	n-GaN	MQWs	p-GaN	GZO	AZO	IZO	Gr	p/n-pad
Thickness/μm	100	1.5	2	0.07	0.1	0.1	0.1	0.1	0.00038	0.3
$K/W{\cdot}m^{-1}{\cdot}k^{-1}$	38	130	160	130	130	26.7	30.1	26.7	5000	385
$\sigma/S^{-1}{\cdot}m^{-1}$	–	–	1×10^4	1×10^3	35	2×10^5	1.4×10^5	7.8×10^4	3.2×10^7	4.5×10^6

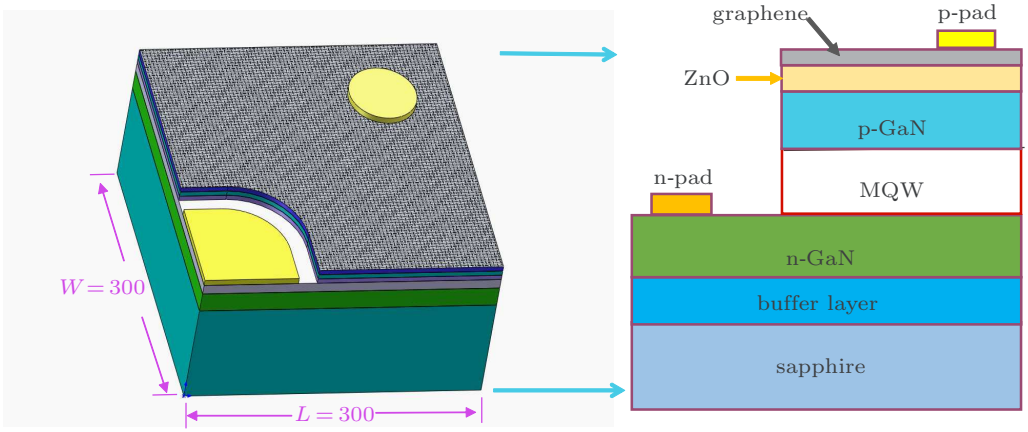


Fig. 1. (color online) 2D/3D structure diagram of the GaN-LED chip with Gr/ZnO as the TCL.

2.2. Simulation methods

The steady-state method is applied to simulate the Joule heat generation, the TCL transmittance, and the operation voltage in GaN LEDs. The relationship between Joule heat Q,[26] current density J, and electrical field E is

$$Q = J \times E, \tag{1}$$

where E is the gradient of the electric potential $\varphi, E = -\nabla\varphi$. φ is determined by the external current density J and the elec-

trical conductivity σ under the static conditions,[27] that is,

$$\nabla(\sigma \cdot \nabla\varphi - J) = 0. \tag{2}$$

Each element in the active layer has an equivalent conductivity as proposed by

$$\sigma = \left(l_{\mathrm{e}}/V_{\mathrm{j}}\right) J_{\mathrm{e}}, \tag{3}$$

where l_{e} is the elemental thickness of the mesh, V_{j} is the voltage drop between the active layer, and J_{e} is the elemental current density. J_{e} and V_{j} of each element satisfy the Shockley equation,[28,29] which describes the J–V characteristics of the LED,

$$J_{\mathrm{e}} = J_0 \left[\exp(eV_{\mathrm{j}}/nKT) - 1\right], \tag{4}$$

where J_0 is the saturation current density, e is the elementary charge (1.6×10^{-19} C), K is the Boltzmann constant (1.38×10^{-23} J/K), T is the absolute temperature, and n is the ideality factor, which depends on the material quality and device structure. Nevertheless, J_0 is also affected by the temperature of the chip. In this work, the saturation current and n are set to be 4.72×10^{-22} A and 2.5, respectively.[30] The relationship between the distribution of the temperature field T and time t is

$$\rho c \frac{\partial T}{\partial t} = \nabla \cdot k_{\mathrm{m}} \nabla(T) + Q, \tag{5}$$

$$k_{\mathrm{m}} \nabla T = h\left(T_{\mathrm{a}} - T\right), \quad T_{\mathrm{bottom}} = T_{\mathrm{a}} = 300\ \mathrm{K}, \tag{6}$$

where Q is the heat source density; T is the temperature; k_{m} is the thermal conductivity of materials; T_{a} and T_{bottom} are the ambient temperature and the sapphire bottom temperature,[21] respectively, both of which are set to be 300 K; and h is the convection heat transfer coefficient, which is equal to 20 W/(m^{2}K).[14] Therefore, from equations (1)–(6), T can be solved by combining the temperature-dependent models of material parameters and the boundary conditions. The current density uniformity is characterized by the standard deviation of the multiple quantum well (MQW) surface current density (σ_{j}), which is expressed by the number n of mesh nodes, the current density J_i of the ith mesh node, and the average current density $\bar{J}$ as

$$\sigma_{\mathrm{j}} = \sqrt{\frac{1}{n-1}\sum_{i=1}^{n}(J_i - \bar{J})^2}, \tag{7}$$

where

$$\bar{J} = \frac{1}{N}\sum_{i=1}^{n} J_i. \tag{8}$$

The transmissions of different TCLs are calculated by COMSOL in an RF frequency. Usually a 550 nm wavelength is used as a reference for comparison of light transmittance among different TCO films whose transmittance should be more than 80% in the range of visible light wavelengths (380–780 nm);[31–33] therefore, the transmittance of various TCLs at 550 nm were calculated in the present work. Meanwhile, the light reflection and absorption depend on the refractive index. Thus, in order to get precise calculated results, the actual refractive index of various doped ZnO should be chosen. Typically, the refractive indices of Gr, IZO, AZO, and GZO are taken as $n = 2.24 + 0.07\mathrm{i}$,[34] $1.97 + 0.01\mathrm{i}$,[17] $1.91 + 0.05\mathrm{i}$,[18] and $1.87 + 0.01\mathrm{i}$,[19] respectively.

The transmission of TCLs is equal to the ratio of the square of the electric field strength. The formulas are

$$\nabla \times \mu_{\mathrm{r}}^{-1}(\nabla \times E) - k_0^2 \varepsilon_{\mathrm{r}} E = 0, \quad \mu_{\mathrm{r}} = 1,$$
$$\varepsilon_{\mathrm{r}} = (n - \mathrm{i}k)^2, \quad \mathrm{Tran} = \frac{\mathrm{Port}_2\left(\mathrm{abs}\left(E^2\right)\right)}{\mathrm{Port}_1\left(\mathrm{abs}\left(E^2\right)\right)}, \tag{9}$$

where μ_{r} is the relative permeability, ε_{r} is the relative conductivity, n and k are the real and imaginary parts of the material's refractive index, respectively. The maximum temperature ($T_{\max}$) and the current density uniformity are applied as the optimal indices of the thermal–electrical performance of GaN-LEDs, and the goal function is expressed as[35]

$$\begin{aligned}\mathrm{cost} = {} & \alpha \cdot K_{\mathrm{T}}\left[T_{\max,l} - \min\left(T_{\max}\right)\right] \\ & + (1-\alpha)\cdot K_{\mathrm{j}}\left[\sigma_{\mathrm{j,m}} - \min\left(\sigma_{\mathrm{j}}\right)\right],\end{aligned} \tag{10}$$

where the K_{T} and K_{j} are

$$K_{\mathrm{T}} = \frac{1}{\max(T_{\max}) - \min(T_{\max})},$$
$$K_{\mathrm{j}} = \frac{1}{\max\left(\sigma_{\mathrm{j}}\right) - \min\left(\sigma_{\mathrm{j}}\right)}, \tag{11}$$

where $\alpha \in [0,1]$, l , and m represent the data points of simulation, and $T_{\max,l}$ and $\sigma_{\mathrm{j,m}}$ are the lth and mth data, respectively. In order to significantly improve the GaN-LED property, $T_{\max}$, σ_{j}, and the cost should be as low as possible.

Each layer of the LED chip is adopted as a homogeneous material.[36,37] The thermal and electrical parameters of various materials are listed in Table 1. It is reported that the resistivity of Gr changes with its number of layers,[38] and therefore the conductivity of Gr with different layers was not taken to be a constant, but adjusted correspondingly in our simulation. The effects of the number of Gr layers on the optical and thermal properties of LEDs with Gr/ZnO composite TCLs are discussed in detail in Section 3 of this paper.

The greatest concern for the approach is if it is precise and accurate enough to predict the thermal and electrical performance of the GaN-LED. In our previous work, the precision and accuracy of the simulation model were validated by comparing the calculated and experimental forward voltages of NiO_x/Gr-LED and ITO/Gr-LED.[12,20–22] However, to the best of our knowledge, there are few reports about the well-fabricated ZnO/Gr-LED and its forward voltages, and thus the comparison of forward voltages is not currently available.

Therefore, in the present work the transmittance was chosen as an alternative comparison reference. The measured transmittances of 200 nm AZO, 280 nm GZO, 440 nm IZO, and 1L–3L Gr at the wavelength of 550 nm have been measured and reported by Park *et al.*,[17] Lin *et al.*,[18] Lameche *et al.*,[19] and Li *et al.*,[34] respectively. Thus, the calculated and experimental transmittances of these six kinds of TCLs were compared, and the results are given in Fig. 2. It shows that the relative error is in the range of 0.3% and 1.6%, indicating that our calculation model has high accuracy and reliability.

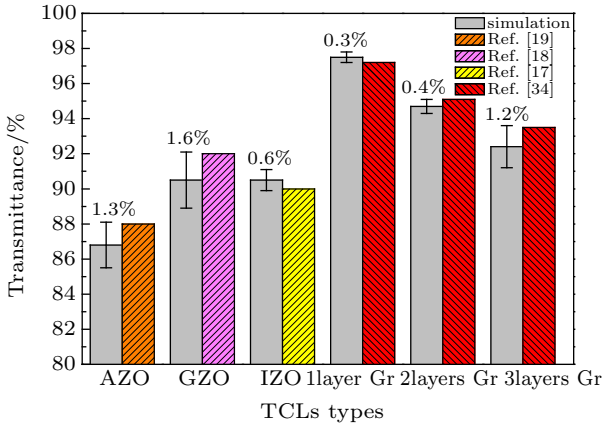


Fig. 2. (color online) The precision and accuracy of the light transmittance between the simulation results obtained from the model built and those from Refs. [17]–[19] and [34] in which TCLs are 200 nm AZO, 280 nm GZO, 440 nm IZO, 1L Gr, 2L Gr, and 3L Gr, respectively.

3. Results and discussion

3.1. Optical properties

Excellent light transmittance is essential for TCLs of LEDs. In order to effectively transmit visible light, the transmittance of the TCLs is generally required to be higher than 80% in the wavelength range of visible light (380–780 nm), which means that ZnO and Gr should not be too thick. To get the possible thickness range of Gr and ZnO, the transmittance of composite films of ZnO and Gr with different thickness combinations was calculated. The calculated transmittances of the hybrid films of single-layer Gr with different thicknesses of AZO, GZO, and IZO are shown in Fig. 3(a). The result indicates that the transmittance of the composite film decreases gradually with the increase in the thickness of AZO, GZO, and IZO. When the ZnO thickness is 400 nm, the light transmittances of the composite films (Gr/AZO, Gr/GZO, and Gr/IZO) are 80.2%, 80.6%, and 81.5%, respectively. However, when the ZnO thickness increases to 500 nm, the transmittances of these three composite films respectively decreases to 79.1%, 78.7%, and 79.9%, which means that this kind of combination does not satisfy the constraint of light transmission. Therefore, the ZnO films should not be thicker than 400 nm in the composite film.

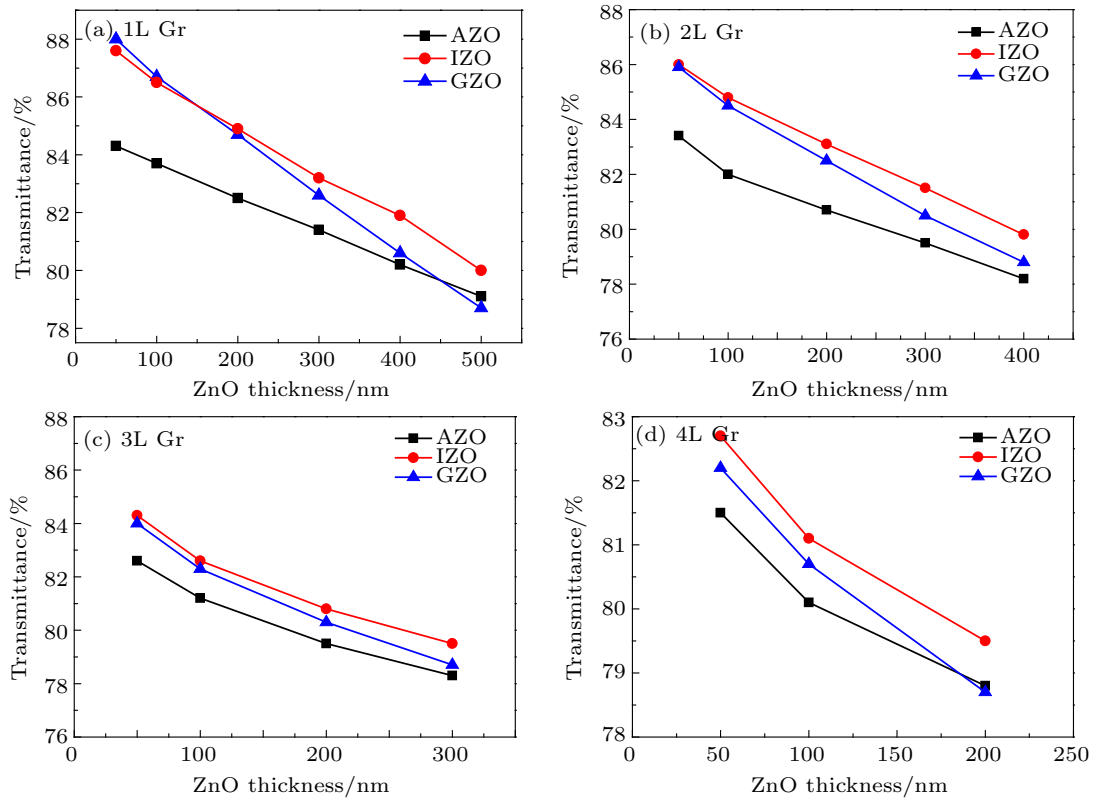


Fig. 3. (color online) The simulated light transmittances of composite films with different layers of Gr and different thicknesses of AZO, GZO, and IZO: (a) 1L Gr/ZnO, (b) 2L Gr/ZnO, (c) 3L Gr/ZnO, and (d) 4L Gr/ZnO.

The transmittances of composite films with different combinations of multilayer (two–four layers) Gr, and ZnO thinner than 400 nm, were also calculated. The results are given in Figs. 3(b)–3(d). They display that the transmittances of composite films are, respectively, 79.1%, 81.5%, and 80.5% when the Gr is two layers and the thicknesses of AZO, GZO, and IZO are 300 nm; however, the transmittances of composite films reduces to 77.5%, 79.8%, and 78.5% for 400 nm AZO, GZO, and IZO, respectively, which means that at in case of 2L Gr, AZO should be less than 300 nm, but GZO and IZO should be less than 400 nm. Similarly, the thickest ZnO films corresponding to 3 L or 4L Gr can be obtained. In summary, to meet the requirement of transparency, the maximum ZnO thickness matched with different Gr layers in the composite Gr/ZnO TCL were determined as 1L Gr:400 nm ZnO, 2L Gr:300 nm ZnO, 3L Gr:200 nm, and 4L Gr:100 nm ZnO.

It is worth noting that the interference effect of the ZnO film on the emission performance of the GaN-LED can be ignored. Based on the simple light path of the LED, GaN→ ZnO→ Gr→ air, the optical path difference between the direct outgoing light and the light reflected by the upper and lower surfaces of the ZnO is $\Delta X = 2 \cdot n \cdot d$, where d and n are, respectively, the thickness and the refractive index of ZnO ($n_{\mathrm{ZnO}} = 1.9$, $n_{\mathrm{GaN}} = 2.4$, and $n_{\mathrm{Gr}} = 2.6$). It is well known that when interference happens, bright or dark stripes will occur for the case of $\Delta X = K\lambda$ or $\Delta X = (2K+1)\lambda/2$, $K = 1, 2, 3, \ldots$. The ZnO thicknesses corresponding to the interference bright and dark stripes at different wavelengths can be calculated from this equation. Typically, the bright and dark stripes will occur, respectively, at 118.4 nm and 57.3 nm ZnO thickness for the 450 nm blue LED; meanwhile, the transmission light of the LED will be interference-enhanced and -weakened at ZnO thicknesses of 144.7 nm and 72.4 nm, respectively, for the 550 nm green LED. Owing to the strong absorption of ZnO below 367 nm (3.37 eV band gap), the Gr/ZnO composite transparent electrode proposed in this paper is mainly aimed at the blue or green GaN-based LED, and thus no strong bright or dark stripes caused by the interference will occur when the ZnO thicknesses are 100 nm, 200 nm, and 300 nm. This indicates that the effect of film interference on the LED emission performance is insignificant and negligible.

3.2. Thermal properties

In order to investigate the role of the TCL in promoting current spreading, alleviating the current crowding effect, and reducing the LED junction temperature, we simulated the overall temperature distribution and the maximum temperature of the LED chip at a current injection of 20 mA. The lower T_{max} is important for the better performance of the LED device. Therefore, we can use T_{max} to characterize the current spreading and heat dissipation of the TCL. The dependences of T_{max} of Gr/ZnO-LEDs on the number of Gr layers and the ZnO thickness at a current injection of 20 mA are revealed in Fig. 4 and Fig. 5, respectively.

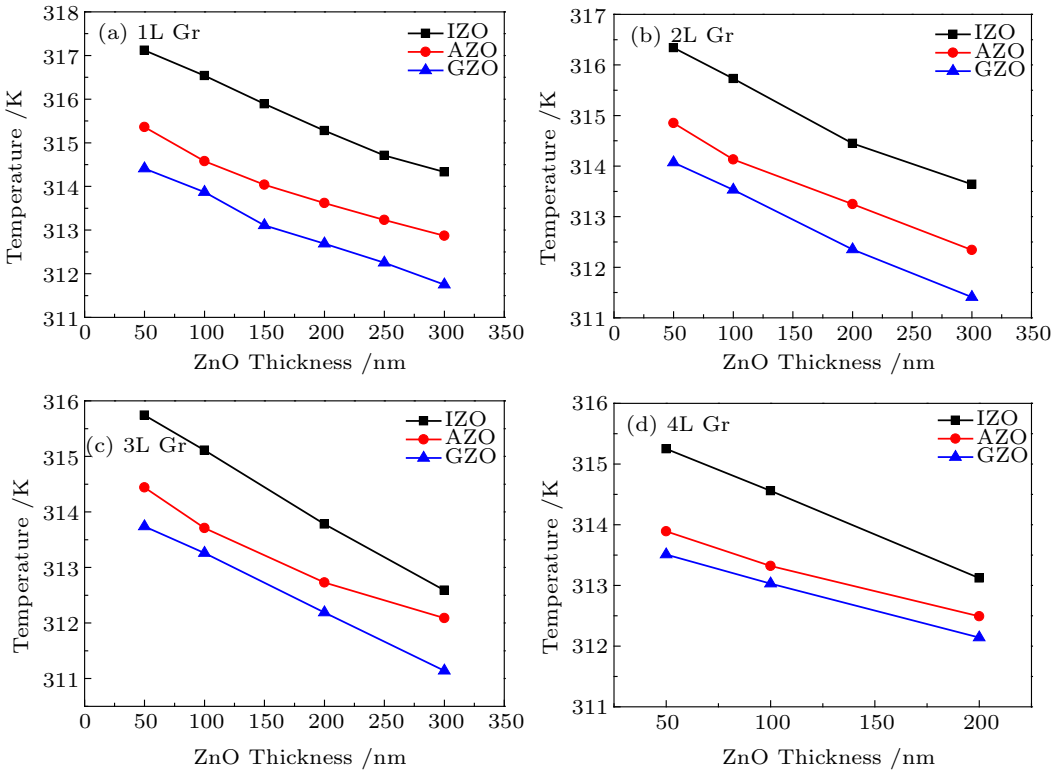


Fig. 4. (color online) The dependence of T_{max} of Gr/ZnO-LEDs on the ZnO thickness at a current injection of 20 mA. (a) 1L Gr/ZnO, (b) 2L Gr/ZnO, (c) 3L Gr/ZnO, and (d) 4L Gr/ZnO.

As shown in Fig. 4, T_{max} of the 1 L–4L Gr/ZnO-LEDs chips gradually decreases with the increase in ZnO thickness when the number of Gr layers is fixed in the range of 1 L–4 L. When the thickness of AZO, GZO, and IZO are the same, the LED with Gr/GZO has the lowest T_{max}, but the one with Gr/IZO has the highest T_{max}, indicating that Gr/GZO has better heat dissipation. The dependence of T_{max} on the number of Gr layers of Gr/ZnO-LEDs when the ZnO thickness is fixed at 100 nm or 200 nm is depicted in Fig. 5. It can be easily found that T_{max} of the chip decreases with the increase in Gr layers or ZnO thickness, and the LED chip with a transparent electrode of four Gr layers has the lowest T_{max} among the cases with the same ZnO thickness. Meanwhile, it is obvious that the drop in T_{max} with the number of Gr layer is more significant at a thin Gr layer than that at a thick Gr layer, which means that T_{max} reduces nonlinearly with the number of Gr layers.

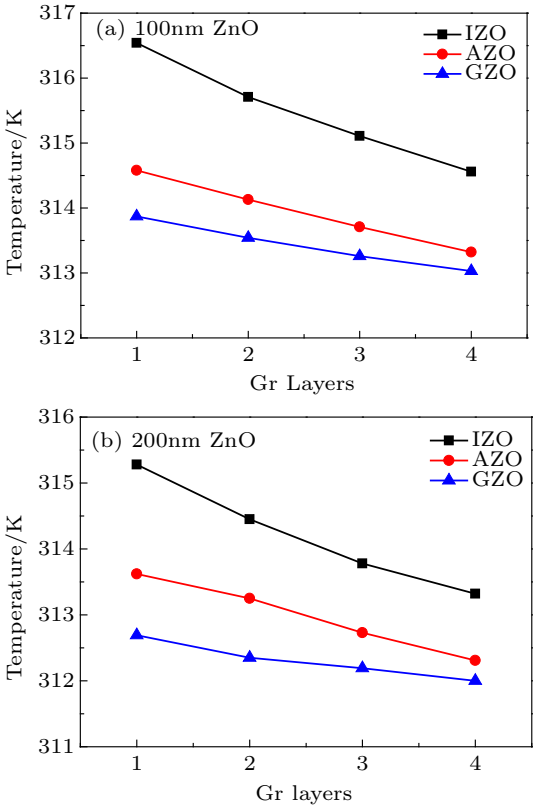


Fig. 5. (color online) The dependence of T_{max} of Gr/ZnO-LEDs on the number of Gr layers at a current injection of 20 mA. (a) Gr/100-nm ZnO and (b) Gr/200-nm ZnO.

The above results can be explained as follows: (i) By inserting a thin ZnO buffer layer, the ohmic contact between Gr and the p-GaN layer is greatly improved. (ii) With the increase in Gr layers and ZnO thickness, the current density decreases in the horizontal direction and the current density uniformity increases, which eliminates the current crowding phenomenon, and thus T_{max} decreases with the number of Gr layers or ZnO thickness. (iii) Nirmalraj *et al.*[38] reported that the resistivity of Gr increases nonlinearly upon increasing the number of Gr layers. Since the thickness of Gr increases at the same time, the sheet resistance of Gr (the ratio of the resistivity to thickness) may reduce with the increase in the number of Gr layers, which will result in a decrease in the total resistance of the composite electrode, the Joule heat, and finally the T_{max}. However, the overall trend of T_{max} decreasing with the increasing number of Gr layers is not linear. Specifically, when the number of layers of Gr is small, for example, increased from one to two, T_{max} decreases more obviously. When the number of layers is larger, for example from three to four layers, the downtrend of T_{max} slows down.

With the increase in ZnO thickness or Gr layers, the thermal performance of LEDs gets better, but the optical properties of the TCL become worse. Therefore, there is an appropriate or optimal combination of Gr and ZnO thickness to trade off the thermal and optical performance of LEDs with Gr/ZnO TCL. Some typical calculation results are listed in Table 2. It can be seen that the LEDs with the TCL (1L Gr/300-nm AZO, 1L Gr/300-nm GZO, and 2L Gr/300-nm IZO) hybrid electrode films have better performance in the corresponding combination of Gr and the same element-doped ZnO films. T_{max} and the transmittance of the LED chip (1L Gr/300-nm AZO, 1L Gr/300-nm GZO, and 2L Gr/300-nm IZO) are 312.87 K and 81.4%, 311.75 K and 82.6%, and 313.64 K and 81.5%, respectively. Furthermore, 1L Gr/300-nm GZO has the best thermal and optical property among the three combinations of TCLs.

Table 2. The transmittance and T_{max} of Gr/ZnO-LED.

Combination	Gr layers/L	ZnO thickness/nm	T_{max}/K	Transmittance/%
Gr	1	0	320.97	97.18
	2	0	320.21	95.15
	3	0	319.55	93.54
	4	0	319.13	91.29
Gr/AZO	1	300	312.87	81.40
	2	200	313.25	80.74
	3	200	313.71	81.23
	4	100	313.32	80.08
Gr/GZO	1	300	311.75	82.61
	2	300	311.41	80.54
	3	200	312.19	80.28
	4	100	313.03	80.66
Gr/IZO	1	300	314.33	83.21
	2	300	313.64	81.49
	3	200	313.78	80.84
	4	100	314.56	81.12

The distributions of surface temperature and active layer current density of the LEDs fabricated with 4L Gr (LED A), 1L Gr/300-nm AZO (LED B), 1L Gr/300-nm GZO (LED C), and 2L Gr/300-nm IZO (LED D) are shown in Fig. 6. T_{max} values of LED A, LED B, LED C, and LED D are 319.13 K, 312.87 K, 311.75 K, and 313.64 K, respectively, indicating that the LEDs with the 300-nm GZO/1L Gr hybrid TCLs could

drop about 7 K compared with the LED with only Gr as the TCL. T_{max} occurs in the p-electrode of the LED chip, which may be attributed to the fact that the lateral current converges and crowds at the ground p-pad.[2] The active layer surface current densities of LEDs B, C, and D distribute more homogeneously than those of LED A.

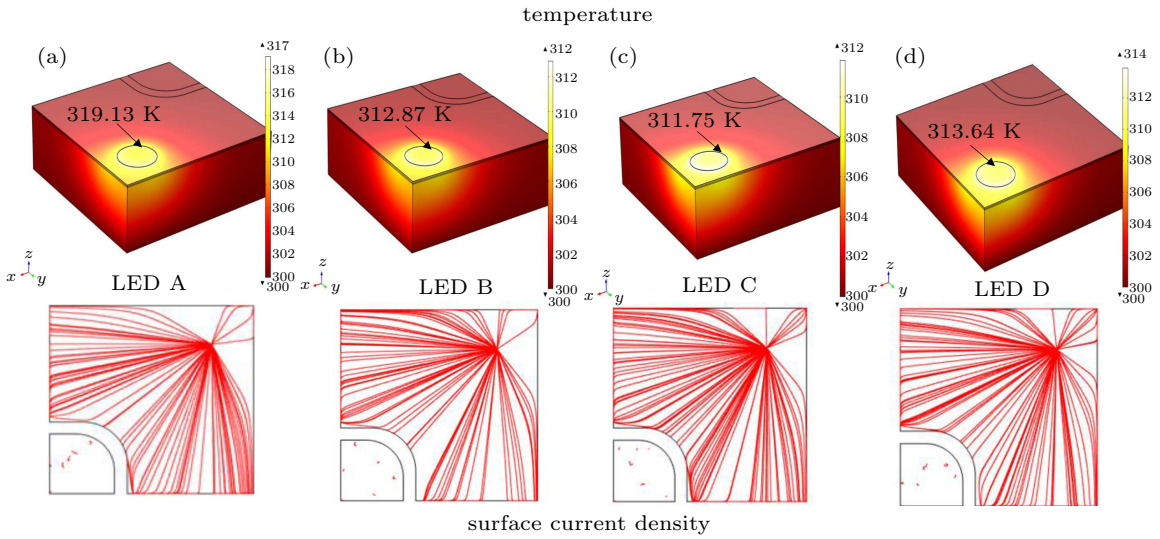


Fig. 6. (color online) The distributions of temperature and active layer surface current density of four kinds of LED chips with various TCLs. (a) 4L Gr, (b) 1L Gr/300-nmAZO, (c) 1L Gr/300-nmGZO, and (d) 2L Gr/300-nm IZO.

4. Conclusion

The optical and thermal properties of different combinations of Gr and doped ZnO (GZO, AZO, and IZO) as the hybrid electrode and current spreading layers on GaN-LED chips are investigated. It is found that the LEDs with hybrid TCLs consisting of 1L Gr/300-nm AZO, 1L Gr/300-nm GZO, and 2L Gr/300-nmIZO have better performance in the corresponding combination of Gr and the same element-doped ZnO films. However, 1L Gr/300-nm GZO has the best thermal and optical property among the above three combinations of TCLs, in which case the T_{max} of LEDs drops by about 7 K compared with the LED with only Gr as the TCL.

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