Simulation on effect of metal/graphene hybrid transparent electrode on characteristics of GaN light emitting diodes*

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In order to decrease the Schottky barrier height and sheet resistance between graphene (Gr) and the p-GaN layers in GaN-based light-emitting diodes (LEDs), some transparent thin films with good conductivity and large work function are essential to insert into Gr and p-GaN layers. In this work, the ultra-thin films of four metals (silver (Ag), golden (Au), nickel (Ni), platinum (Pt)) are explored to introduce as a bridge layer into Gr and p-GaN, respectively. The effect of a different combination of Gr/metal transparent conductive layers (TCLs) on the electrical, optical, and thermal characteristics of LED was investigated by the finite element methods. It is found that both the TCLs transmittance and the surface temperature of the LED chip reduces with the increase of the metal thickness, and the transmittance decreases to about 80% with the metal thickness increasing to 2 nm. The surface temperature distribution, operation voltage, and optical output power of the LED chips with different metal/Gr combination were calculated and analyzed. Based on the electrical, optical, and thermal performance of LEDs, it is found that 1.5-nm Ag or Ni or Pt, but 1-nm Au combined with 3 layered (L) Gr is the optimal Gr/metal hybrid transparent and current spreading electrode for ultra-violet (UV) or near-UV LEDs.

Keywords: finite element methods, graphene, temperature distribution, transmittance, light-emitting diodes

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1. Introduction

Owing to their numerous advantages, such as high brightness, low energy consumption and long lifetime,[1,2] the GaN-LED have attracted extensive attention in the past few decades and been widely used in various fields including general lighting, traffic lights, automobile headlights, and backlight units for liquid crystal displays.[3,4] Usually the transparent conductive tin-doped indium oxide (ITO) is employed as the transparent electrode and current spreading layer for the contact on p-type GaN of LEDs. Although the device performance with ITO-based transparent p-type contacts has been improved, the exclusive uses of these oxides have faced serious problems due to the high cost, dwindling supply of indium, chemical instability in the presence of acids or alkali, and high processing temperature.[5,6] Furthermore, ITO shows extraordinary high absorption in the UV region, which makes it difficult to practically use as a transparent and current spreading electrode in UV LEDs. Hence, a transparent alternative electrode with the optical transmittance and electrical performances near to or better than those of the ITO layer is necessary.[7] Gr, which is formed of covalently bonded carbon atoms with a hexagonal lattice structure, is considered as an ideal ultra-thin two-dimensional candidate to replace conventional ITO electrodes because of its excellent optical and electrical properties. So a few investigations on the application of Gr as the transparent conducting electrodes in GaN-based LEDs have been reported recently.[8–10] However, due to the large difference of work function between the Gr (4.4 eV) and p-GaN layers (7.5 eV), a direct contact of Gr on the p-GaN layers will lead to a high sheet resistance and a large Schottky barrier height.[11–13] Therefore, forming a good Ohmic contact on the p-GaN layer is a key way to improve the performance of GaN-based LEDs with the Gr transparent electrodes layer.[14] To resolve these problems, ITO and NiO_x as insertion lay-

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ers between Gr and P-GaN have been investigated and some reasonable results have been achieved.[3,14] But there are still some problems to be solved, such as high work temperature of LED and unmatched conductivity between the electrode and p-GaN layers.[1,15] The metals (Ag, Au, Ni, Pt) possess good conductivity, high thermal conductivity, and large work function (Au/5.2 eV, Pt/5.7 eV, Ni/5.2 eV, Ag/4.4 eV, respectively) which are nearer to that of the p-GaN layer (7.5 eV). The insertion of an ultrathin metal material between the Gr layer and the p-GaN layer is expected to reduce the Schottky barrier, the contact resistance and the surface temperature if the metal is thin enough to make no significant optical loss.[12,16] Some work has been reported. For example, Ag nanowire,[7] Ni thin film,[12] Au nanocluster[17] were explored to insert between p-GaN and Gr to enhance the light output power in the UV LEDs, and Au-doped Gr was attempted by Seo *et al.*[13] and Kim *et al.*[16] These works indicate that the hybrid metal/Gr structure can be used as a transparent and current spreading electrode in UV or near-UV LEDs to improve the external quantum efficiency (EQE) and reduce the current crowding effect (CCE).[18] However, the influence of the thickness of the metal and Gr layers on the performance of LED and the optimal combination of Gr and different metal are still unknown. In the present work, the three-dimensional finite element simulation method is applied to analyze the effect of four kinds of metal/Gr hybrid structure (Ag/Gr, Au/Gr, Ni/Gr, and Pt/Gr) as the transparent electrode on the electrical, optical and thermal properties of GaN LEDs, and finally the optimal combination of Gr and metals (Ag, Au, Ni, Pt) is suggested.

2. Simulation

In this work, semiconductor module, RF module, and Joule heating module in COMSOL Multiphysics software were used.

2.1. Simulation structure

Numerical simulations for GaN LEDs have been proposed to overcome the technical challenges, including the thermal management and the current crowding phenomenon. We used the three-dimensional finite element method with the simulation software COMSOL to build the geometry model of the GaN-LED chip. Figure 1 is the schematic diagram of a typical GaN-LED chip whose size is 350 μm×350 μm.[7,13,17] The physical parameters of various materials are listed in Table 1. Here, three layers of Gr are about 1-nm thick.

Table 1. Physical parameters of various materials. Notes: sapphire and buffer layers are defined as heat transfer in a solid in the simulation procedure, thus their electrical conductivity is not needed. K: thermal conductivity, σ: electrical conductivity.

Material	Sapphire	u-GaN	n-GaN	MQWs	p-GaN	Ag	Au	Ni	Pt	Gr	p/n-pad
Thickness/μm	100	1.5	3	0.1	0.15	0.001	0.001	0.001	0.001	0.001	0.2
K/(W/m·K)	38	130	160	130	130	429	317	90	71.6	5000	317
σ/(S/m)	–	–	10^4	10^3	35	6.3×10^7	4.5×10^6	1.4×10^7	8.9×10^6	3.2×10^7	4.5×10^6

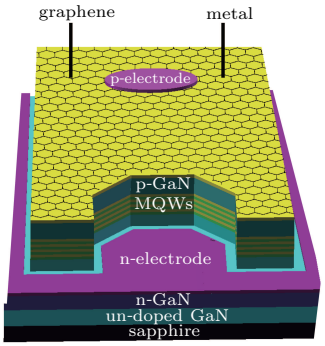


Fig. 1. (color online) Schematic diagram of the GaN-LED chip with the transparent and current spreading electrode.

2.2. Simulation methods

The steady-state method was used to simulate the Joule heat generation, the TCLs transmittance, and the operation voltage in GaN LEDs. The relationship between Joule heat Q,[19] current density J, and electrical field E is

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

where $\boldsymbol{E}$ is the gradient of the electric potential φ; $E = -\nabla\varphi$; φ is determined by an external current density $\boldsymbol{J}$ and the electrical conductivity σ under the static conditions,[20]

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

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

$$\sigma = (l_{\mathrm{e}}/V_j)J_{\mathrm{e}}, \tag{3}$$

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. The J_{e} and V_j of each element satisfy the Shockley equation,[21,22] which describes the J–V characteristics of the LED,

$$J_{\mathrm{e}} = J_0\left[\exp\left(\frac{eV_j}{nkT}\right) - 1\right], \tag{4}$$

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 were set to be 4.72×10^{-22} A and 2.5,[15] respectively. The

relationship between the distribution of the temperature field T and time t is

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

$$k_m \nabla T = h(T_a - T), \quad T_{bottom} = T_a = 300\ \mathrm{K}, \tag{6}$$

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,[23] respectively, both of which are set to be 300 K; h is the convection heat transfer coefficient, which is equal to 20 W/(m^{2}K).[23] Therefore, from Eqs. (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 i-th mesh node and the average current density $\bar{J}$,

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

where

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

The transmissions of different TCLs are calculated by the COMSOL in an RF frequency domain at 550 nm, and the refractive indices of Gr, Au, Ag, Ni, and Pt are taken from the literature, $n = 2.24 + 0.07\mathrm{i}$,[24] $1.7 + 0.54\mathrm{i}$,[25] $1.4 + 0.96\mathrm{i}$,[26] $2.95 + 0.68\mathrm{i}$,[27] $1.65 + 0.42\mathrm{i}$,[28] respectively. The transmission of TCLs is equal to the ratio of the square of the electric field strength. The formulas are

$$\nabla \times \mu_r^{-1}(\nabla \times E) - K_0^2 \varepsilon_r E = 0, \quad \mu_r = 1,$$
$$\varepsilon_r = (n - \mathrm{i}k)^2, \quad \mathrm{Tran} = \mathrm{Port}_2(\mathrm{abs}(E^2))/\mathrm{Port}_1(\mathrm{abs}(E^2)). \tag{9}$$

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 Eq. (10).[29] In order to improve the GaN-LED property significantly, T_{max}, σ_j and the cost should be as low as possible,

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

where the K_T and K_j are

$$K_T = \frac{1}{\max(T_{max}) - \min(T_{max})}, \tag{11}$$

$$K_j = \frac{1}{\max(\sigma_j) - min(\sigma_j)}, \tag{12}$$

where $\alpha \in [0, 1]$; l and m represent the data points of simulation; T_{max} and σ_j are the data of l-th and m-th, respectively.

Each layer of the GaN-LED chip is adopted as a homogeneous material.[30,31] The most concern for the approach is whether it is precise and accurate enough to predict the thermal and electrical performance of GaN-LED or not.[32] Based on the approximation between the simulation result and the experimental result of Seo *et al.*,[13] the original thermal and electrical parameters of different materials, which are derived from the literature, are adjusted considering the precision and accuracy of the Joule heat model. The final thermal and electrical parameters are listed in Table 1. In our previous work, the thermal and electrical property of NiO_x/Gr-LED and ITO/Gr-LED were calculated by using this method, and it is found that the relative errors of the simulated voltage for NiO_x/Gr-LED and ITO/Gr-LED are 6.5% and 4.94%, respectively,[23,33] indicating that our numerical method is reliable. So in the present work, we adopt this method to study the influence of four kinds of metal/Gr hybrid structure (Ag/Gr, Au/Gr, Ni/Gr, and Pt/Gr) on the LED performance.

3. Simulation results and discussion

3.1. Electrical properties

The I–V characteristics, optical output powers and EQE of GaN-LEDs with Ag/Gr, Au/Gr, Ni/Gr, Pt/Gr, and Gr as TCLs were calculated through the semiconductor module. Figure 2(a) shows the I–V characteristics of GaN-LEDs with various current spreading layers. It is obvious that the operation voltage of all metal/Gr-LEDs is lower than that of Gr-LEDs, and the Ag/Gr-LED holds the largest operation voltage among the four metal/Gr-LEDs. The reason is that the work function of metal is higher than that of Gr and more close to p-GaN, so metal/Gr-LEDs have lower operation voltage, indicating that a thin metal film deposited with Gr can reduce the sheet resistance of Gr and the operation voltage. Due to the lowest work function of Ag among the four metals, so the operation voltage of Ag/Gr-LED is higher than that of the other three metal/Gr-LEDs.

Typically, the forward voltages at 20-mA current injection were 4.78 V, 3.91 V, 3.84 V, 3.92 V, and 6.03 V for GaN-LED with Ag/Gr, Au/Gr, Ni/Gr, Pt/Gr, and Gr, respectively. Compared with the experimental forward voltages of Au/Gr-LED (4.25 V) and Gr-LED (7.2 V) reported by Seo *et al.*,[17] the relative errors of the simulated voltage for Au/Gr-LED and Gr-LED are 8.0% and 16.25%, respectively. The errors may result from two factors: First, the boundary variables of metal and Gr in the simulation module were different from the experimental parameters;[17] second, an ideal Schottky contact was considered in the simulation model, but actually the contact between the electrode and GaN layer is not an ideal Schottky one. By the way, the Gr layer is usually regarded as smooth in the simulation work, but in fact it is maybe wrinkled on the p-GaN, while the metals (Ag, Au, Ni, and Pt) with a certain thickness are considered to be pyknotic and continuous in both simulation and practice, thus the relative error of Gr-LED operation voltage is larger than that of Au/Gr-LED.

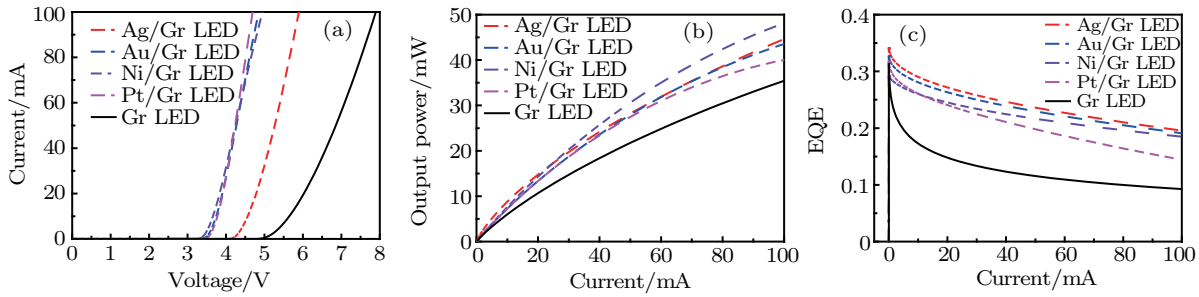


Fig. 2. (color online) (a) *I*–*V* characteristics and (b) optical output powers versus injection current and (c) EQE versus current curves of GaN-LEDs with Ag/Gr, Au/Gr, Ni/Gr, Pt/Gr and Gr as TCLs.

Figure 2(b) reveals the optical output power versus injection current curves of the LEDs. The light output power was increased with the increase of current. The variation tendency of light output power is similar to the experiment result of Liu *et al.*[34] typically, the light output powers are 45.97, 43.56, 48.20, 43.21, and 39.93 mW at 100 mA for the LEDs fabricated with Ag/Gr, Au/Gr, Ni/Gr, Pt/Gr, and Gr as TCLs, respectively. It can be seen that the metal/Gr LED achieves a higher light output power. The metal could reduce the operation voltage and the sheet resistance, thus reduce the temperature of the LED chip and increase the luminous flux of the LED chip.

From Fig. 2(c), it is found that the EQEs of the five LEDs significantly decrease with the increase of the injection current. The efficiency droops at 100 mA are 53.3%, 52.4%, 54.4%, 51.7%, and 70.8% for LEDs with Ag/Gr, Au/Gr, Ni/Gr, Pt/Gr, and Gr as TCLs, respectively, indicating that the efficiency droop of the metal/Gr-LED is lower than that of the Gr-LED, which can be assigned to the metals inserted as a bridge layer between Gr and p-GaN providing good electric conductivity and thermal conductivity. Several studies have been reported that the efficiency droop is maybe caused by various origins, such as from crystallographic defects (point defects[35] and threading dislocations[36]), electron leakage (caused by limited electron blocking layers,[37] lower hole-injection efficiency,[38] incomplete capture carrier by quantum wells[39]), Auger recombination,[40] heating effects,[41] and current crowding.[42] Wang *et al.* has carefully studied the correlation of EQE between current diffusion by a modified ABC model, and found that the poor current diffusion will decrease the internal quantum efficiency (IQE), light extraction efficiency (LEE), and EQE.[18] In the present work, the Auger recombination, optical transition, and trap-assisted recombination were taken into account in our simulation, but because the semiconductor module we built is one-dimensional, the lateral current diffusion was not considered.

3.2. Optical properties

A high optical transmittance is indispensable when the top electrode film is used as the TCLs in LEDs. The light efficiency of LEDs strongly relies on the transmittance of TCLs. As displayed in Fig. 3(a), the light transmittance of the metal film decreases with the increase of metal thickness. The average transmittance at the wavelength of 550 nm shows a linear dependence on metal thickness (nm) with a negative slope of ($\sim -10\%$/nm), which is consistent with the result of Lee *et al.*[12]

Therefore, metal thickness is an important influential factor to cause optical loss. Figure 3(b) demonstrates the layer-number dependence of the transmittance of stacking Gr at $\lambda = 550$ nm. It can be seen that the transmittance of Gr is more than 90% for one to three layers, and remains 88% for four layers. The transmittance error is within 1.61% compared with the experimental result.[43] Thus, our simulation result is proper and reliable.

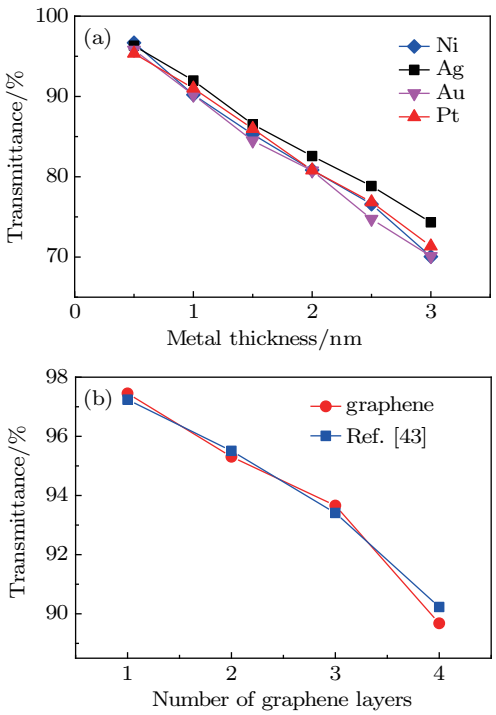


Fig. 3. (color online) (a) Optical transmittance characteristics of four metals at 550-nm wavelengths (b) optical transmittance characteristics of Gr layer at 550-nm wavelengths.

Optical transmittance of graphene with different layers and the metal (Ag, Au, Ni, Pt) with different thicknesses has been studied. The transmittance of visible light monotonously

declines with the increase of the metal/Gr layers. However, the transmittances of metal (0.5 nm)/2L Gr and metal (2 nm)/3L Gr are still up to ∼ 91% and ∼ 80%, respectively, which are similar to those of the experiment results.[7,12,17] So metal/Gr was deemed to be potentially used in the LED chip as window electrodes.

3.3. Thermal properties

The lower T_{max} is important to get the better performance of the device.[44] The variation of the maximum temperature with 1L-Gr and the metal thickness was depicted in Fig. 4(a). The T_{max} declines with metal thickness, which is attributed to the fact that the conductivity of the continuous and pyknotic metal film increase with the increase of the thickness. As found in Fig. 4(b), the standard deviation of current density decline progressively with the increase of the metal thickness. The decrease of current density is owing to the lower contact resistance of metal/p-type GaN. Metal has a relatively large work function and a lower Schottky barrier contact with p-GaN.

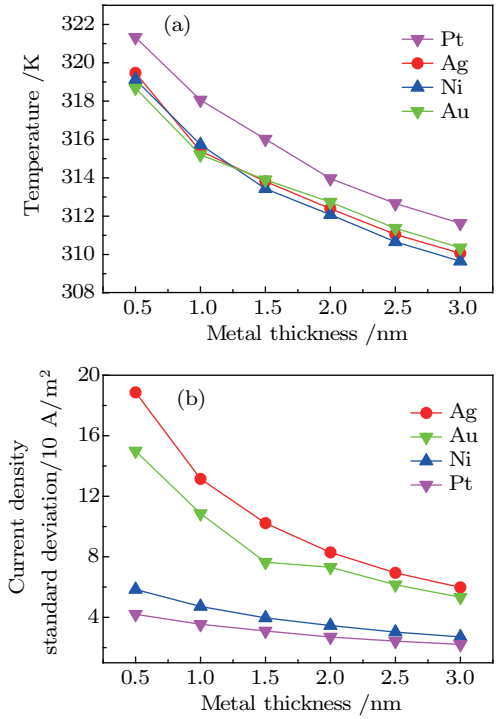


Fig. 4. (color online) (a) Maximum temperature and (b) standard deviation of current density with 1 layer Gr and different thickness of metals.

The effects of TCLs with the diverse combinations of Gr (1∼4 layers) and metal (0.5 nm∼ 3 nm) on the maximum temperature of the LED chip were calculated, and the results were shown in Fig. 5. The T_{max} decreases with the increase of the thickness of metal and the number of Gr layers. The 4 L of Gr-LED chip has the lowest service temperature, which is attributed to the trade-off from two aspects: the decreased current density in the horizontal direction and the increased electrical conductivity of Gr with the increase of Gr layers. In addition, since the conductive metal film provides a bridge between Gr and p-GaN, the T_{max} of metal/Gr-LED is lower than that of Gr-LED. The LEDs with the TCLs (1.5-nm Ag/3L Gr, 1-nm Au/3L Gr, 1.5-nm Ni/3L Gr, and 1.5-nm Pt/3L Gr) hybrid electrode films hold the better performance according to the goal function combined with the electrical properties, optical properties, and thermal properties of the TCLs. The T_{max} and σ_j of LED chip (1.5-nm Ag/3L Gr, 1-nm Au/3L Gr, 1.5-nm Ni/3L Gr, and 1.5-nm Pt/3L Gr) is 311.32 K, 7.9×10^{10} A/m^2; 311.58 K, 6.82×10^{10} A/m^2; 311.56 K, 2.83×10^{10} A/m^2, and 312.85 K, 1.93×10^{10} A/m^2, respectively. The transmittance of TCLs (1.5-nm Ag/3L Gr, 1-nm Au/3L Gr, 1.5-nm Ni/3L Gr, and 1.5-nm Pt/3L Gr) is 81.18%, 85.77%, 89.75%, 81.35%, respectively. The result (transmittance, T_{max} and σ_j) of optimal composite LED chip is shown in Table 2.

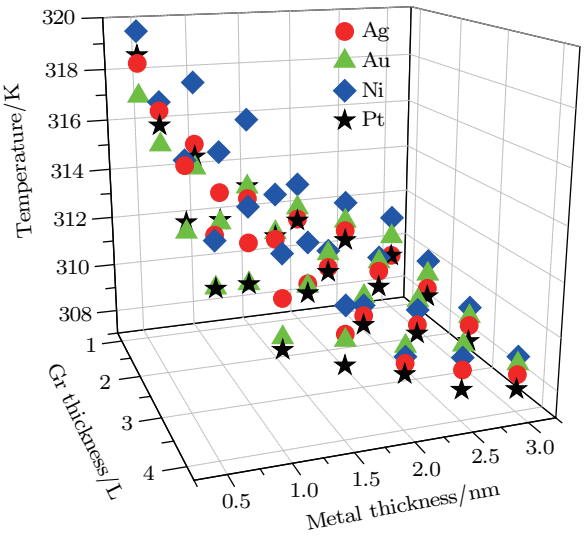


Fig. 5. (color online) The T_{max} distribution of LED with metal film and Gr layer at the injection current 20 mA.

Table 2. The transmittance, T_{max} and σ_j for metal/Gr optimization ratio.

Metal/Gr	Transmittance/%	T_{max}/K	$\sigma_j/10^{10}$ A/m^2
1.5-nm Ag/3LGr	81.18	311.32	7.9
1-nm Au/3L Gr	85.77	311.58	6.82
1-nm Ni/3L Gr	89.75	311.56	2.83
1.5-nm Pt/3L Gr	81.35	312.85	1.93

The distribution of the surfaces temperature and MQW current density of LEDs are shown in Fig. 6. The T_{max} of four metal/Gr-LED chips occurs in the regions near the p-electrode. The fact is that the lateral current converges and crowds at the p-electrode. The current crowding of metal/Gr-LEDs chip is lower than that of the Gr-LED or metal-LED chip, and the current density of metal/Gr-LEDs decreases about 22.70%, 37.14%, 40.04%, 34.84% compared with the TCLs of metals (Ag, Au, Ni, Pt), respectively. This is attributed to the decreased contact resistance, which leads to more current passing from p-GaN to MQWs and n-GaN.[46] As a result, the light emission of GaN-based LEDs with the metal (Ag, Au, Ni, Pt) /Gr hybrid electrode is enhanced and the vertical current also increases in MQW film.

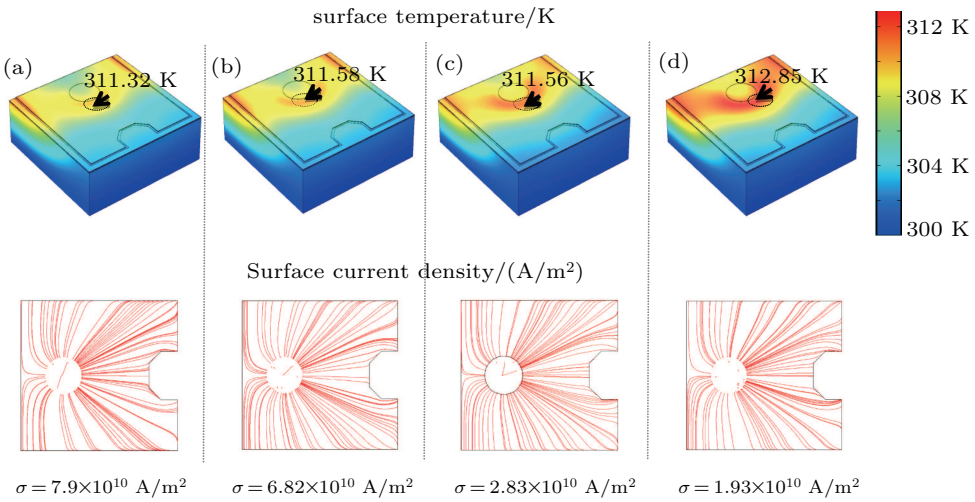


Fig. 6. (color online) The surface temperature and MQW surface current distribution of LED, (a) Ag/Gr as TCLs, (b) Au/Gr as TCLs, (c) Ni/Gr as TCLs, (d) Pt/Gr as TCLs.

4. Conclusion

In summary, the effect of different combinations of metal (Ag, Au, Ni, Pt) and Gr as the hybrid electrode and current spreading layers on the optical, thermal, and electrical performance of GaN-LEDs have been investigated. It shows that the metals inserted between Gr and p-GaN layer as a bridge layer can reduce the sheet resistance and contact resistance through a lower Schottky barrier, which improves the external quantum efficiency and optical output power and decreases the operation voltage. It is found that 1.5-nm Ag/3L Gr, 1-nm Au/3 L Gr, 1.5-nm Ni/3L Gr and 1.5-nm Pt/3L Gr as TCLs are the optimal combinations. Typically, when metal (Ag, Au, Ni, Pt)/3-layers Gr film hybrid TCLs are added, the temperature of LEDs could drop by about 12 K compared with that of only Gr as TCLs.

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