Design of near-unity transmittance dielectric/Ag/ITO electrodes for GaN-based light-emitting diodes

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Abstract

We designed a near-unity transmittance dielectric/Ag/ITO electrode for high-efficiency GaN-based light-emitting diodes by using the scattering matrix method. The transmittance of an ultrathin metal layer, sandwiched between a dielectric layer and an ITO layer, was investigated as a function of the thickness and the optical constant of each constituent layer. Three different metals (Ag, Au, and Al) were examined as the metal layer. The analytical simulation indicated that the transmittance of a dielectric/metal/ITO multilayer film is maximized with an approximately 10-nm-thick Ag layer. Additionally, the transmittance also tends to increase as the refractive index of the upper dielectric layer increases. By tailoring the thickness of the dielectric layer and the ITO layer, the dielectric/Ag/ITO structure yielded a transmittance of 0.97, which surpasses the maximum transmittance (0.91) of a single ITO film. Furthermore, this extraordinary transmittance was present for other visible wavelengths of light, including violet and green colors. A complex phasor diagram model confirmed that the transmittance of the dielectric/metal/ITO multilayer film is influenced by the interference of reflected partial waves. These numerical findings underpin a rational design principle for metal-based multilayer films that are utilized as transparent electrodes for the development of efficient light-emitting diodes and solar cell devices.

1. Introduction

Dielectric/metal/dielectric multilayer films are considered an efficient and facile transparent electrode scheme for the development of a variety of optoelectronic applications, including solar cell devices [1,2], light-emitting diodes [3,4], and general color-selective light absorbers [5–7], due to their superior optical (transmission > 90% [8]) and electrical properties (sheet resistance < 10 Ω/sq [8]). In addition, a dielectric/metal/dielectric multilayer film that is composed of relatively thin (<a few tens of nm) metal and dielectric layers is suitable for use in flexible optoelectronic devices [9,10]. Conversely, a thick (over ~100 nm) transparent conducting oxide precludes inclusion within flexible devices due to its brittleness [8]. For upper and/or lower dielectrics in a multilayer film, various oxide compounds, such as ITO [11–13], ZnO [12,14], Al-doped ZnO [15], and In-doped ZnO [16], have been examined thus far. For example, Mohamed et al. reported ZnO/Ag/ZnO electrodes with a sheet resistance of <6.3 Ω/sq and a maximum transmittance of 0.93 at λ = 550 nm [14]. Girtan et al. studied ITO/Ag/ITO electrodes placed on a low refractive index substrate (e.g., glass and PET) that yielded a resistivity of 3 × 10–5 Ω cm and a transmittance of up to 97% at λ = 550 nm [11]. Although metal-based multilayer electrodes have been shown to exhibit decent optical, electrical and mechanical performances, the design principles related to their transmission properties are not yet completely understood.

In this paper, we studied the optical properties of dielectric/Ag/ITO multilayer films placed on a semi-infinite GaN layer where light is generated in the GaN medium. Using the scattering matrix method [17,18], the transmittance of dielectric/Ag/ITO multilayer films was investigated as a function of the thickness and the optical constant of each constituent layer. A complex phasor diagram model was introduced to illustrate the trends in transmittance [7]. By applying rational design principles, the optimum Ag-based multilayer film that yielded the greatest transmittance was determined. These numerical findings help to provide a generic route for designing efficient transparent electrodes for the development of high-efficiency light-emitting diodes.
2. Simulation model

Fig. 1(A) shows the schematic of a dielectric/metal/ITO multilayer film placed on a GaN (refractive index n = 2.5) layer. A normal plane wave of \( \lambda = 450 \text{ nm} \) was generated in the GaN medium. Using the scattering matrix method, the transmittance was calculated with respect to the thickness of the dielectric layer (d) and the thickness of the metal layer (t). For simplicity, the thickness of the ITO layer was set to be equal to that of the dielectric layer. The complex scattering matrix can be induced by applying electric and magnetic boundary conditions as in [18]:

\[
M_j = \begin{bmatrix} \cos \delta_j & i P_j \sin \delta_j \\ i P_j \sin \delta_j & \cos \delta_j \end{bmatrix},
\]

(1)

where

\[
P_j = (n_j + ik_j) \cos \theta_{j, \text{incident}}.
\]

(2)

\[
\delta_j = \frac{2 \pi P_j d_j}{\lambda}
\]

(3)

Here, \( \theta_{j, \text{incident}} \) is the incident angle upon a j-th optical medium and \( \delta_j \) is the phase shift experienced by the optical medium with a thickness of \( d_j \). For a dielectric/metal/ITO multilayer film, the scattering matrix is expressed as:

\[
M = M_{\text{ITO}} M_{\text{metal}} M_{\text{dielectric}} = \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix}.
\]

(4)

Finally, the transmittance (T) is given by:

\[
T = \frac{n_{\text{air}}}{n_{\text{GaN}}} \left( \frac{1}{(m_{11} + m_{22})^2 + (m_{12} + m_{21})^2} \right)^2.
\]

(5)

For this calculation, we chose a Ag material as the middle metal layer and set its optical constants at \( \lambda = 450 \text{ nm} \) as \((n, k) = (0.04, 2.65)\) [19]. For the ITO layer, the optical constants \((n, k)\) were \((2.06, 0.014)\) [20]. The refractive index of the upper dielectric layer was 2.0, which corresponds to that of \( \text{Si}_3\text{N}_4 \). The calculated transmittance reveals three key aspects related to the optical properties of a dielectric/metal/ITO multilayer film [Fig. 1(B)]. First, the transmittance is sinusoidally modulated with d due to the interference between reflected waves. Second, the overall transmittance is degraded with increasing t. However, it is noteworthy that the maximum transmittance of the multilayer film within a range of \( t = 4–10 \text{ nm} \) is kept nearly constant despite different Ag thicknesses. The maximum transmittance was nearly constant at the optimum Ag thicknesses because for dielectric/metal/dielectric multilayer films, their partial reflected waves can be interfered destructively at the conditions, which will be discussed in Fig. 2. Increasing Ag thickness up to 10 nm yields the maximum transmittance and enables the fabrication of a dielectric/Ag/ITO electrode [21,22]. For example, Dhar et al. reported that the deposited Ag film was changed from islands to a continuous film at a critical thickness \( t_c \) of 9.5 nm [22]. Hence, the optical and electrical properties of the Ag film were stabilized when its thickness was larger than \( t_c \). More importantly, the maximum transmittance of 0.97 surpasses that of a single ITO film (0.91) [black line, Fig. 1(B)]. Third, the maximum transmittance peaks of the multilayer film occur at the same thicknesses \((d \sim 30 \text{ and } 140 \text{ nm})\), which are marginally affected by t. The transmittance has local maxima at every destructive condition for the partial reflected waves. The modulation period \( \Delta d \) of 110 nm is determined by the quarter wavelength antireflecting coating condition of dielectric materials [17]. Note that the maximum transmittance decreases with increasing d because of the material absorption of the ITO layer.

Light-emitting diodes generate omnidirectional light from their multiple quantum wells. Therefore, one must investigate the angle-dependent optical properties of the transparent electrode [23]. The angular transmittance with transverse-electric (TE) polarized light for a multilayer film \((t = 12 \text{ nm})\) with was compared with a single ITO film [Fig. 1(C)]. For normally incident light, the maximum transmittance of the multilayer film with \( t = 12 \text{ nm} \) was nearly the same as that of a single ITO film [Fig. 1(B)]. Interestingly, the transmittance of the multilayer film remains almost constant within a critical angle \((\sim 24^\circ)\) as compared with the ITO film. The difference in the transmittance between the two films is more significant at a steeper incident angle. The broadband angular transmittance from the dielectric/ITO/Ag stems from the multiple interferences between reflected waves that occur at each of the four different interfaces: GaN/ITO, ITO/Ag, Ag/dielectric, and dielectric/air [24]. Taken together, a dielectric/metal/ITO multilayer film with a substantial \((t > 8 \text{ nm})\) Ag layer provides near-unity transmittance and an ultralow sheet resistance, which makes it a strong candidate for next-generation transparent electrodes.

3. Theory

To better understand the transmission behavior of a dielectric/metal/ITO multilayer film, we represented phasors for the reflected partial waves in the complex plane, accounting for Fresnel reflections and transmission coefficients [17]. Fig. 2(A) represents the four lowest orders of optical paths: \(l, m = (0, 0), (2, 0), (4, 0), \) and \((2, 2)\), where \(l (m)\) is the number of lights passing through the ITO.
and minimum (D) transmittance conditions in Fig. 1(B). The thickness of the Ag layer was 12 nm.

For simpler analysis, group (4, 0) and other higher order groups were not included in the phasor diagram because their phasors were not accounted for because of their large extinction coefficient [19]. For each group, a representative phasor (dashed arrow) was constructed by adding the partial phasors (solid arrows) that correspond to each different optical path [Fig. 2(B)]. For example, for the (2, 0) group, the representative phasor was the vector sum of the partial phasors assigned to optical paths b, c, and d.

We examined two different cases in the complex phasor diagram: (t, d) = (12, 30) and (12, 100) nm, which yielded the maximum [Fig. 2(C)] and minimum [Fig. 2(D)] transmittance in Fig. 1(B), respectively. All of the reflected partial waves in the same group undergo the same phase shift, as caused by the same thicknesses of the ITO and dielectric layers; thus, the corresponding partial phasors and their representative phasor are rotated equally in the complex plane. For example, the rotation angle ($\phi_{lm}$) for a group ($l, m$) is expressed by:

$$\phi_{lm} = l \times \frac{2\pi n_{ITO} d_{ITO}}{\lambda} + m \times \frac{2\pi n_{dielectric} d_{dielectric}}{\lambda}$$

For simpler analysis, group (4, 0) and other higher order groups were not included in the phasor diagram because their phasors were negligibly small. For the maximum transmittance case [Fig. 2(C)], each phasor is added destructively such that the total phasor spirals the origin in the complex plane [7]. Conversely, for the minimum transmittance case [Fig. 2(D)], each phasor is added in phase such that the total phasor significantly deviates from the origin [7]. Hence, one can conclude that the modulated transmittance that results from changes in the dielectric thickness is a direct consequence of the interference between reflected partial waves.

4. Results and discussion

The transmittance of a dielectric/metal/ITO multilayer film is dictated by the destructive or constructive interference of reflected light. This can be modulated by changing either the optical constants or thickness of each layer. We first investigated the transmittance of a dielectric/Ag/ITO multilayer film as a function of $d$ for a variety of dielectric refractive indices ($n$) [Fig. 3(A)]. For these simulations, the wavelength of incident light was 450 nm. First, the thickness of the Ag layer was set to be 8 nm because the maximum transmittance occurred at the condition as shown in Fig. 1(B) [left, Fig. 3(A)]. The calculated results clearly show that the maximum transmittance occurs at a smaller dielectric thickness with increasing $n$ (e.g., for $t = 12$ nm, $d = 54$ nm with $n = 1.5, d = 32$ nm with $n = 2.0$, and $d = 22$ nm with $n = 2.4$) because an optical path length is proportional to the refractive index of the dielectric material. Also, the modulation period in transmittance decreases with increasing the refractive index of the dielectric material. For an upper dielectric layer with $n = 2.4$, the transmittance reaches a minimum at $d = 50 - 60$ nm, which meets the first constructive interference condition for the partial reflected waves. These findings can be understood as a general trend of interference. More importantly, transmission increases gradually until $n$ is equal to approximately 2.0. An upper dielectric layer with a high refractive index produces a large reflection coefficient at the interface between the dielectric and air, which is beneficial to the complete destructive interference condition for reflected light. The effect of high refractive index becomes more apparent for the multilayer films with $t = 12$ nm; the maximum transmittance increases slightly more even when $n$ is larger than 2.0 [right, Fig. 3(A)]. Dielectric materials with $n \geq 2.0$ (e.g., Si3N4, TiO2, or ZrO2) can be readily prepared by conventional deposition techniques. Furthermore, the optical absorption of the dielectric material can be ignored in these types of thin layers ($d = 25$ nm), which is another advantage of Ag-based multilayer films.

Next, we examined three different metals with distinct optical constants ($n$, $k$): Au (1.38, 1.92) [19], Ag (0.04, 2.65) [19], and Al (0.62, 5.30) [20] at $\lambda = 450$ nm [Fig. 3(B)]. For all of the metals, the
thickness was fixed at 8 nm. The calculated results show that the multilayer film with a Ag layer provides the greatest transmission as compared with the other metal layers. The large extinction coefficient of Al gives rise to significant optical absorption as light passes the metal layer. Alternatively, for Au, the real part of the optical constants (i.e., the refractive index) is relatively large, which diminishes the reflection coefficients for the ITO/Au and Au/dielectric interfaces. A metal suitable for use in a high-transmittance multilayer film must have a small refractive index ($n$) and a moderate extinction coefficient ($k$). The optical constants of Ag meet these criteria. Notably, the transmittance peaks appear at the same $d$ for the three different metals. The reason for this is that the refractive index of metals is typically small compared to that of dielectric materials, which marginally rotates phasors in the complex plane. Using the same explanation, the maximum transmittance is observed at the same $d$ for different Ag thicknesses, as shown in Fig. 1(B).

GaN-based light-emitting diodes emit violet, blue, and green colors because their band gaps can be tuned by modulating the In [25] or Al [26] composition. To examine the performance of a dielectric/Ag/ITO multilayer film for other wavelengths, we conducted the same simulations for $\lambda = 400$ nm [left, Fig. 4], 550 nm [middle, Fig. 4], and 280 nm [right, Fig. 4]. The optical constants ($n$, $k$) of Ag were (0.05, 2.10) at $\lambda = 400$ nm, (0.55, 3.60) at $\lambda = 550$ nm, and (1.41, 1.29) at $\lambda = 280$ nm [19]. The absolute transmittance at the different wavelengths depends on the details of the optical constants; however, the overall trend with respect to the dielectric thickness is the same. In general, for the visible spectrum, the transmittance of a dielectric/Ag/ITO multilayer film tends to decrease at longer wavelengths because of the increased extinction coefficient ($k$). Such high-transmittance of the dielectric/Ag/ITO structure over visible spectrum is consistent with previous works [3,8]. For example, Lee et al. demonstrated that the transmittance of ITO (15 nm)/Ag (7 nm)/ITO (15 nm) multilayer films is superior to that of a single ITO film (60 nm) for $\lambda = 350$–600 nm [3]. The maximum transmittance of the ITO/Ag/ITO structure was ~90% at $\lambda = 400$ nm, ~92% at $\lambda = 450$ nm, and ~89% at $\lambda = 550$ nm, which could be more augmented by adjusting the thickness of the ITO layer. In addition, we examined the dielectric/Ag/ITO multilayer films for a deep ultraviolet wavelength ($\lambda = 280$ nm) because the wavelength region is of great interest due to the development of water purifiers and sterizers [27]. The transmittance result showed that the dielectric/Ag/ITO structure was superior to a single ITO film only when $t$ was smaller than 8 nm [right, Fig. 4]. The refractive index ($n$) of Ag increases abruptly at $\lambda < 350$ nm (e.g., $n = 1.41$ at $\lambda = 280$ nm [20]), which diminishes the reflection coefficients at the interfaces of multilayer films, as discussed in the example of Au [Fig. 3(B)]. Therefore, the optical effect of the dielectric/Ag/ITO multilayer films is less significant at deep ultraviolet wavelengths.

We have investigated dielectric/Ag/ITO multilayer films where the thickness of the dielectric layer ($d_1$) is the same as that of the ITO layer ($d_2$). However, from Eq. (6), the rotation angle will have an additional degree of freedom if $d_1$ is dissimilar from $d_2$, as shown in Fig. 5(A). Therefore, we calculated the transmittance while varying $d_1$ and $d_2$ simultaneously; this is depicted as a surface plot exhibiting the transmittance [left, Fig. 5(B)]. For these simulations, the thickness of the Ag layer was fixed at 12 nm where the maximum transmittance of the multilayer film was nearly the same as that of a single ITO film [Fig. 1(B)]. The refractive index of the dielectric layer was 2.0. The surface plot shows that the transmittance is modulated with changes to both $d_1$ and $d_2$; however, the modulation is less significant when $d_2$ is changed. As discussed in Fig. 2, the
The transmittance of asymmetric dielectric/Ag/ITO multilayer films. (A) Schematic of an asymmetric dielectric/Ag/ITO multilayer film placed on a GaN medium. In this structure, the thickness of the dielectric layer can differ from that of the ITO layer. (B) Left: Surface plot representing the transmittance of dielectric/Ag/ITO multilayer films with respect to the thicknesses of the dielectric and ITO layers. Right: transmittance of dielectric/Ag/ITO multilayer films as a function of the ITO thickness along the dashed horizontal line ($d_1 = 34$ nm) of the surface plot.


References


5. Summary and conclusions

We studied the transmission features of dielectric/metal/ITO multilayer films placed on a GaN medium using the scattering matrix method. Simulation results indicated that the multilayer film consisting of a thick (~10 nm) Ag layer exhibits a transmittance of 0.94, which is larger than the maximum transmittance of a single ITO film (~0.91). The transmittance of the multilayer film can be further enhanced (up to 0.97) by individually tailoring the thicknesses of the dielectric and ITO layers. Such extraordinary transmission from dielectric/Ag/ITO multilayer films was not limited to the blue wavelength of light, but also exhibited a similar effect for violet and green wavelengths. The modulation of transmittance with respect to the structural parameters was completely investigated using a phasor diagram model; the maximum and minimum transmittances were a direct consequence of the destructive and constructive interference of reflected light. Designing dielectric/metal/ITO multilayer films for light-emitting diode and solar cell applications represents a unique strategy for realizing more efficient transparent electrodes with improved optical and electrical performances.

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