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Color-saturated and angle-stable blue top-emitting organic light-emitting diodes based on semitransparent bilayer cathode: Theory and experiment

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Based on a modified electromagnetic theory, a bilayer metal cathode consisting of an electron injection layer and a silver (Ag) layer is designed to improve the color chromaticity in blue top-emitting organic light-emitting diodes (TEOLEDs). The effects of the complex refractive index of the electron injection material on the reflectivity and transmittivity of the bilayer cathode are investigated in detail, and then samarium (Sm) is selected as the electron injection material due to its proper refractive index of ~1.22 + 1.12i and work function of ~2.7 eV. Then, the emission peak wavelength, the full width at half maximum, and the Commission International de L'Eclairage coordinates of the blue TEOLEDs with different Sm/Ag bilayer cathodes are calculated and discussed. According to the theoretical results, a blue TEOLED with the optimized bilayer cathode of Sm (15 nm)/Ag (5 nm) is fabricated. The measurement results indicate that the blue TEOLED possesses an excellent chromaticity which is even better than that of a bottom-emitting organic light-emitting diode. Besides, the excellent angle stability is observed in the blue TEOLED even with a large viewing angle change of 0–75°.

1. Introduction

Top-emitting organic light-emitting diodes (TEOLEDs) can be fabricated on an opaque substrate and emit light through the semitransparent top electrode. Display panels based on TEOLEDs own a high aperture ratio, a bright luminance, a low operating voltage, and a long lifetime compared to those based on bottom-emitting organic light-emitting diodes (BEOLEDs) since all circuits can be placed at the bottom of the substrate and high-quality active matrix organic light-emitting diodes (OLEDs) can be easily fabricated on the substrate, such as silicon [1,2].

As is well known, a TEOLED consists of a reflective anode, several organic functional layers and a semitransparent cathode, which is usually an optical microcavity. Due to the strong multi-beam interference in the microcavity, the emission intensity at the resonance wavelength which satisfies the resonance condition (1) will be greatly enhanced [3,4].

$$-|\phi_1| - |\phi_2| + \frac{4\pi L}{\lambda} = m2\pi \qquad (1)$$

In this expression, L is the optical length of the cavity, λ is the wavelength, ϕ_1 and ϕ_2 are the phase change on reflectance of mirrors, and m is the resonance mode number. If the resonance wavelength is consistent with the peak wavelength of an emission material, the spectrum of the TEOLED would be obviously narrowed, which is beneficial to obtain a high-saturated color for some monochromatic OLEDs. In red or green TEOLEDs, the total thickness of organic layers around 100 nm not only realizes the resonance at the emission peak, but also provides the efficient

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carriers' injection and transmission, and then high-efficiency and saturated-color devices can be got [5–7]. While in blue TEOLEDs with a usually-used total organic layer thickness of ~100 nm, the resonance wavelength (~550 nm) mismatches with the emission wavelength (<470 nm) of the blue emission material and results in the redshift of the emission peak.

According to Eq. (1), two ways are usually used to the design of the structure of the blue TEOLEDs: (i) modulating the total thickness of layers between the two reflective electrodes, i.e. the cavity length, and (ii) varying the reflectance of the top semitransparent cathode. Some researchers modulate the cavity length by adjusting the indium tin oxide (ITO) thickness of silver/ITO (Ag/ITO) bilayer anode to obtain highly saturated blue emission in TEOLEDs [8,9]. Although this method is beneficial to the chromaticity and the efficiency of the blue TEOLEDs, the preparation technique is actually complicated. So the design of the semitransparent top electrode plays a key role in the optimization of the blue TEOLEDs.

Ag is usually utilized as a cathode material for its good conductivity and stability. But due to its high work function, electrons are difficult to inject from the Ag cathode to an organic electron transport layer. Some approaches are used to improve the electron injection of Ag [6,10,11], in which a bilayer cathode based on Ag and a low work function metal has been proven to be an effective way due to its high injection efficiency and not complicated operation technique. However, the high reflection and absorption from both two metal layers influences the light output performances such as brightness and color. A capping layer is an effective measure to decrease the reflectance of the top cathode and improve the chromaticity of the blue TEOLEDs [12–15]. But the optical properties of the semitransparent cathode have been less focused on during the research work in the past two decades.

In this paper, the reflectance of the cathodes, the spectra, and the Commission International de L'Eclairage (CIE) coordinates of the blue TEOLEDs are simulated with a classic electromagnetic model modified by us. Then, based on the optimized bilayer cathode the blue TEOLEDs with an improved color purity and angle stability are designed and fabricated. Finally the optical and electric characteristics of the devices are discussed.

2. Experiment

The TEOLEDs investigated in this paper have structures of glass substrate/Ag (70 nm)/MoOx (3 nm)/4,4′,4″-tris(3-methylphenylphenylamino) triphenylamine (m-MTDATA, 25 nm)/N,N′-di(naphthalene-1-yl)-N,N′-diphenyl-benzidine (NPB, 10 nm)/4,4′-bis(carbazol-9-yl)-biphenyl (CBP, 30 nm): iridium (III) bis[(4,6-difluorophenyl)-pyridinato-N,C2′]picolinate (FIrpic, 7 wt.%)/4,7-diphenyl-1,10-phenanthroline (Bphen, 40 nm)/electron injection layer (EIL, *y* nm)/Ag (*x* nm). Here, m-MTDATA, NPB, CBP, FIrpic and Bphen are utilized as a hole inject layer, a hole transport layer, a blue host layer, a blue phosphorescent guest, and an electron transport layer, respectively. The thick Ag layer of 70 nm is utilized as the anode, modified with MoOx (3 nm) to improve the hole injection efficiency. The EIL (*y* nm) and a thin Ag (*x* nm) layer are used as the semitransparent bilayer cathode.

The blue TEOLEDs were grown on the glass substrates. Before the device fabrication, the substrates were cleaned sequentially with acetone and ethanol by using an ultrasonic bath, rinsed then with deionized water, and finally dried in an oven. All depositions were finished in a high vacuum about 10^{-6} Torr with a rate of 0.1–0.3 nm/s. Current density–voltage–luminance characteristics of the devices were measured with the recombination of a Keithley 2400 source measurement unit with a Photo-Research-655 Spectroscan Colorimeter in room-temperature air. The photoluminescence (PL) spectrum of the blue phosphor FIrpic was measured with a RF-5301PC spectrofluorimeter.

3. Model and design

In the classic electromagnetic theory, the radiation emission from recombining excitons is modeled with oscillating electric dipoles. An arbitrarily oriented dipole embedded in a layered medium can be decomposed into a parallel component ($d_{\parallel}$) and a vertical one ($d_{\perp}$) respect to the interfaces of the layered system. The electric field emitted by an electric dipole can be represented as a superposition of s-(TE) and p-(TM) polarized plane waves. Considering that $d_{\perp}$ has no contributions to s-polarized wave, the classical emission patterns of an arbitrarily oriented dipole can be decomposed into contributes of three components (p_s, p_p, p_v), where p_s and p_p are s- and p-polarized waves of the parallel dipole components, and p_v is the vertical dipole component [16].

The emission patterns of such components in an unbounded medium with a refractive index n_e are of forms

$$p_s^0 = \frac{3}{8\pi}, \quad p_p^0 = \frac{3}{8\pi}\cos\alpha_e, \quad \text{and} \quad p_v^0 = \frac{3}{8\pi}\sin\alpha_e \tag{2}$$

where α_e is the emission angle in medium [16]. For the dipoles located in a multilayer device, the emission patterns will be modulated by the interference on the interfaces. Because the s-polarized and p-polarized waves emitted from parallel and perpendicular dipoles experience different reflection on an interface, the modulation on the three components are different. Considering the wide-angle and multi-beam interferences in a microcavity [1], the emission patterns from the three dipole components embedded in a multilayer device can be expressed as

$$p_s(\lambda,\alpha) = \frac{3}{8\pi}\frac{\left|1 + r_B^s(\lambda,\alpha)\exp(i4\pi n_e z_e\cos\alpha_e/\lambda)\right|^2}{\left|1 - r_T^s(\lambda,\alpha)r_B^s(\lambda,\alpha)\exp(i4\pi n_e d_e\cos\alpha_e/\lambda)\right|^2}\left|t_T^s(\lambda,\alpha)\right|^2, \tag{3}$$

$$p_p(\lambda,\alpha) = \frac{3}{8\pi}\cos^2(\alpha_e)\frac{\left|1 - r_B^p(\lambda,\alpha)\exp(i4\pi n_e z_e\cos\alpha_e/\lambda)\right|^2}{\left|1 - r_T^p(\lambda,\alpha)r_B^p(\lambda,\alpha)\exp(i4\pi n_e d_e\cos\alpha_e/\lambda)\right|^2}\left|t_T^p(\lambda,\alpha)\right|^2, \tag{4}$$

$$\text{and} \quad p_v(\lambda,\alpha) = \frac{3}{8\pi}\sin^2(\alpha_e)\frac{\left|1 + r_B^p(\lambda,\alpha)\exp(i4\pi n_e z_e\cos\alpha_e/\lambda)\right|^2}{\left|1 - r_T^p(\lambda,\alpha)r_B^p(\lambda,\alpha)\exp(i4\pi n_e d_e\cos\alpha_e/\lambda)\right|^2}\left|t_T^p(\lambda,\alpha)\right|^2. \tag{5}$$

They are the functions of the wavelength λ and the propagation direction α in air. Here, ${r_{T/B}}^{s/p}$ and ${t_{T/B}}^{s/p}$ represent the reflection and transmission coefficients. Superscript s and p denote the s- and p-polarized waves.