Alleviate microcavity effects in top-emitting white organic light-emitting diodes for achieving broadband and high color rendition emission spectra

Shuming Chen, Hoi-Sing Kwok

Center for Display Research, Department of Electronic and Computer Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong

A R T I C L E  I N F O

Article history:
Received 25 July 2011
Received in revised form 21 August 2011
Accepted 22 August 2011
Available online 9 September 2011

Keywords:
Top-emitting
White organic light-emitting diodes
Microcavity
Multiple emission peaks
High color rendering index
Microdisplay

A B S T R A C T

It is challenging to obtain broadband emission spectra in top-emitting white organic light-emitting diodes (WOLEDs) due to the well known microcavity effects. In this work, we demonstrate that the microcavity effects can be greatly alleviated by employing a low reflection tri-cathode-layer Yb (5 nm)/Au (15 nm)/MoO3 (35 nm). Top-emitting WOLEDs with evenly separated red, green and blue emission peaks have been achieved. The white emission spectra show weak angle dependence as well. At a luminance of 1000 cd/m², the top-emitting WOLEDs show an efficiency of 23 cd/A, 10.5 lm/W, a 1931 Commission International de L’Eclairage coordinate of (0.36, 0.42) and a high color rendering index of 85. The top-emitting WOLEDs have been successfully applied in the Si based microdisplay.

© 2011 Elsevier B.V. All rights reserved.

1. Introduction

White organic light-emitting diodes (WOLEDs) have received intensive study recently due to their potential applications in full color display and solid-state lighting [1,2]. In high resolution full color display such as the Si based microdisplay, the resolution typically is higher than SVGA+ and the sub-pixel size is smaller than 5 × 15 μm [3]; it is thus unpractical to realize full color microdisplay by laterally depositing the red (R), green (G) and blue (B) emission layers using fine metal shadow mask due to the difficulty of manufacturing, maintaining and aligning the shadow mask with tiny pixel pitch [4]. Therefore, it is more desirable to adopt the unpatterned WOLEDs combined with patterned RGB color filters to generate the three primary colors [5–7]. For microdisplay applications, the WOLEDs are required to be integrated on opaque Si based complementary metal-oxide semiconductor (CMOS) backplane. Thus one has to develop the top-emitting WOLEDs with a reflective bottom electrode and a semitransparent top electrode to force the light emitting from the top surface. The top-emitting WOLEDs should possess broadband emission spectra with three evenly separated R, G and B peaks so that the output white light can be separated into three primary colors after passing through the color filters. However, it is challenging to obtain the top-emitting WOLEDs with broadband spectra due to the well known microcavity effects which shape and narrow the emission spectra [5,6,8]. To alleviate the microcavity effects presented between two reflective electrodes, one has to reduce the reflection of either electrode. Many approaches have been proposed to achieve a low reflection electrode, for example, adopting a semitransparent Ca/Ag or LiF/Al/Ag cathode capped with an index matching layer [7,9–13], employing a transparent indium-tin-oxide top electrode [14], or using a low reflection anode material like Mo [15] or Cu [16]. Although top-emitting WOLEDs have been successfully demonstrated by employing the above-mentioned low reflection electrodes, the spectra or
efficiency remain unsatisfied for high color gamut full color display or high color rendition illumination applications.

Au, one of the metals with low absorption, low reflection and high transmission, would be potentially an ideal cathode material for top-emitting WOLEDs [17]. However, few works have been reported on using Au as a low reflection and semitransparent cathode for top-emitting WOLEDs [18], mainly due to the high work function (~5.1 eV) of Au, rendering it unsuitable as a cathode material for injecting electrons. In this work, we demonstrate that with a 5 nm Yb (Ytterbium) modification layer, Au can be an excellent cathode for injecting electrons. The Au based tri-cathode-layer Yb (5 nm)/Au (15 nm)/MoO$_3$ (35 nm) exhibits an average reflection of 9.8% over the visible wavelength, significantly lower than 24% for the Ag based tri-cathode-layer Yb (5 nm)/Ag (15 nm)/MoO$_3$ (30 nm). By employing the low reflection Au based tri-cathode-layer, the microcavity effects have been weakened largely. Thus, high efficiency and high color rendition top-emitting WOLEDs with evenly separated R, G and B emission peaks have been achieved. At a high luminance of 1000 cd/m$^2$, the top-emitting WOLEDs exhibit an efficiency of 23 cd/A, 10.5 lm/W, a 1931 Commission International de l’Eclaireage (CIE) coordinate of (0.36, 0.42) and a high color rendering index (CRI) of 85. The top-emitting WOLEDs demonstrated here would be an ideal candidate for high resolution full color active-matrix display or indoor lighting applications.

2. Experimental

Bis[3,5-difluoro-2-(2-pyridyl)phenyl]-(2-carboxypyridyl) iridium(III), FlrPic, and bis[2-benzothiophen-2-yl-pyridine] (acetylacetonate)iridium(III). Ir(btp)$_2$(acac) with main emission peak at 475 and 612 nm were selected as sky blue and saturated red emitter, respectively. In order to generate the white spectra with three evenly separated R, G and B emission peaks, a new yellowish-green emitter bis[5-(trifluoromethyl)-2-p-tolylpyridine] (acetylacetonate)iridium(III) 1 with emission peak at 544 nm was specially designed and employed in this study [1]. The top-emitting WOLEDs were fabricated in a multi-source vacuum chamber at a base pressure of around 5 × 10$^{-7}$ Torr with structure glass/Al (100 nm)/MoO$_3$ (8 nm)/NPB (20, 30, 40 nm)/6% Ir(btp)$_2$(acac):CDBP (10 nm)/4% FlrPic:CDBP (5 nm)/TAZ (40 nm)/Yb 5 nm/Au (15 nm)/MoO$_3$ (35, 30 nm), (NPB: N,N’-bis[naphthalen-1-yl]-N,N’-bis-phenyl)-benzidine; CDBP: 4,4’-bis[carbazol-9-yl]-2,2’-dimethylbiphenyl; TAZ: 3-(4-biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole), where Al, MoO$_3$, and NPB function as reflective anode, hole-injection and hole-transporting layer, respectively, CDBP works as host, TAZ serves as exciton-blocking and electron-transporting layer, Yb/Au (or Ag)/MoO$_3$ acts as semitransparent tri-cathode-layer. The energy level and molecular structure of the emitters are shown in Fig. 1. The transmission and reflection spectra of the tri-cathode-layer were characterized by UV–Vis spectrophotometer. The current density–voltage–luminance characteristics of the devices were measured by the HP4145B semiconductor parameter analyzer and a calibrated UDT PIN-25D silicon photodiode. The electroluminescent (EL) spectra were obtained with the PR650 spectrophotometer.

3. Results and discussion

To alleviate the microcavity effects of the top-emitting WOLEDs, important parameters which affect the interference are analyzed first. The top-emitting OLEDs with the emission layers sandwiched between two reflective metal electrodes can be treated as a Fabry–Perot cavity optically [8,9,13,19–22]. Light emitting from the top semitransparent electrode and light reflected by the bottom reflective electrode interfere with each other. The emission intensity at $\lambda$ is enhanced when the cavity effective length $L_{eff}$ satisfies the below equation [8,20]:

$$L_{eff} = \frac{(\phi_m + \phi_{bm})\lambda}{4\pi} + \sum n_i d_i = m\frac{\lambda}{2} \text{ (m : integer)},$$

(1)

where $\phi_m$ and $\phi_{bm}$ are the phase shift of the light reflected by the top and bottom electrode, respectively; $n_i$ and $d_i$ are the refractive index and thickness of the organic layers, respectively. The cavity behaves like an optical filter which modulates the free space emission with the modulation spectra given by [8,13,19–21]:

$$I_{ca}(\lambda) \propto \frac{1 + R_1 + 2\sqrt{R_1} \cos \left(\frac{4\pi L_{eff}}{\lambda} + \phi_m + \phi_{bm}\right)}{1 + R_1R_2 - 2\sqrt{R_1R_2} \cos \left(\frac{4\pi L_{eff}}{\lambda} + \phi_{bm} + \phi_m\right)} T_2,$$

(2)

where $L_1$ is the optical distance between the exciton main recombination zone and the bottom reflective electrode; $L$ is optical length of the organic layers; $R_1$ and $R_2$ are the reflectance of the bottom and top electrode, respectively; $T_2$ is the transmittance of the top electrode. The full width at half maximum (FWHM) of the $I_{ca}(\lambda)$ can be estimated by [8,9,13]:

$$\text{FWHM} = \frac{\lambda^2}{2L_{eff}} \times \frac{1 - \sqrt{R_1R_2}}{\pi \sqrt{R_1R_2}}.$$  

From Eq. (3), one can see that larger reflectance of either electrode results in smaller FWHM, implying a stronger interference presented in the cavity. In order to obtain...
broadband emission spectra, one has to maximize the FWHM by reducing the reflection of either electrode. It is more desirable to reduce the reflection of the top electrode, since reducing the reflection of the bottom electrode, though effective, are at a cost of decreasing the light coupling efficiency.

Among the metal materials, thin film of Ag and Au are two mostly selected semitransparent top electrode materials due to their intrinsically low absorption and high transmission. Fig. 2 compares the measured reflection and transmission spectra of the Ag and Au based semitransparent top cathodes. It can be seen that the Au based top cathodes exhibit a substantially lower reflection over the visible wavelength than that of the Ag. Also, at yellow to red color, the transmittance of the Au based top cathodes is higher than that of the Ag based top cathodes. For example, the average reflectance (transmittance) of Yb (5 nm)/Au (15 nm) is 34% (52%), significantly lower (higher) than 54% (42%) for Yb (5 nm)/Ag (15 nm). By capping the semitransparent top cathodes with an index matching layer with optimized thickness, the reflectance (transmittance) of the Au and Ag based top cathodes are further reduced (enhanced). For example, the average reflectance (transmittance) has been remarkably reduced (enhanced) from 34% (52%) for the Au top cathodes without index matching layer to 9.8% (15 nm)/MoO3 (35 nm) exhibits a rather flat modulation intensity from 440 to 600 nm, implying a good possibility of obtaining broadband white emission spectra with equal R, G and B intensity in such cavity.

To verify this assertion, top-emitting WOLEDs with different top cathodes were fabricated. Fig. 4 shows the EL spectra of the devices with different top cathodes. For devices with Yb (5 nm)/Ag (15 nm) cathode, due to strong interference at \( \lambda = 525 \text{ nm} \) as predicted by the simulated cavity modulation spectra, the green emission is enhanced significantly while the blue and red emission are suppressed, resulting in unbalanced white spectra as shown in Fig. 4a inset. For the devices with Ag and Au based tri-cathode-layer, the cavity length was tuned deliberately...
by changing the thickness of NPB to further compare the interference effects of the cavity with different top cathodes. As shown in Fig. 4a, for devices with Ag based tri-cathode-layer, as NPB thickness increases, the blue emission decreases rapidly, which may be attributable to the interference effects or shifting of the recombination zone. Also, the spectra exhibit slightly red-shift as the cavity length increases, a typical phenomenon in interference cavity. However, for the devices with Au based tri-cathode-layer, the white spectra only exhibit a slight change as the increased of cavity length, which may be mainly caused by the shifting of the recombination zone. Noted that, both devices have the same emission and injection layers, thus the exciton recombination zone should be identical. The severe change and red shifting white spectra as the increased of cavity length of the devices with Ag based tri-cathode layer are therefore most likely caused by interference effects; hence to obtain balanced white emission, one should carefully tune the thickness of the cavity length. While the relatively stable white spectra as the increased of cavity length of the devices with Au based tri-cathode-layer implies a much weakened interference effect; hence broadband white spectra with evenly separated R, G, B peaks and equal intensity are much easier to obtain, as shown in Fig. 4b. At NPB = 30 nm, the white spectra exhibit a CIE color of (0.36, 0.42) and a high CRI of 85, rendering such top-emitting WOLEDs to be ideal candidate for display or lighting applications. We have successfully applied such top-emitting WOLEDs in the Si based micro-display with SVGA+ resolution, as shown in Fig. 4b inset. It is noted that the emission color of the demonstrated top-emitting WOLEDs is not pure white and the CIE coordinate is far from (0.33, 0.33). To remedy this deficiency, deep fluorescent blue emitter like 4,4′-bis(9-ethyl-3-carbazovinylene)-1,1′-biphenyl (BcZVBi) may be employed to replace the sky blue emitter FlrPic [1].

The viewing characteristics of the white spectra are further examined. As shown in Fig. 5a, for the devices with Ag based tri-cathode-layer, the red emission decreases while the blue emission increases as the increased of viewing angle, mainly due to the reduction of cavity length viewed from an oblique angle. While the red emission remains unchanged regardless of the change of viewing angle for the devices with Au tri-cathode-layer as shown in Fig. 5b, again confirms a neglectable interference effect. However, the blue emission decreases as the increased of viewing angle, which is attributable to the strong absorption of the Au at the blue color region. At oblique angle, light see a thicker Au layer than that of their cousins at normal angle, thus leading to lower blue emission intensity at oblique angle. Fig. 5c and d show the EL spectra under different driving voltages. At high voltage, the exciton recombination zone shifts towards the green/red emission layer, thus one observes a decreasing blue emission and an increased red emission. The CIE coordinates changing from (0.34, 0.40) at 6 V to (0.36, 0.42) at 10 V for the devices with Au based tri-cathode layer.

Fig. 6 compares the key characteristics of the top-emitting WOLEDs with the Ag and the Au tri-cathode-layer. Both devices show similar current density–voltage–luminance and efficiency–luminance characteristics due to their identical structures; for example, both devices turn on at 3.8 V for 1 cd/m² and at ~7 V for 1000 cd/m², indicating that with the modification of Yb electron injection layer, Au works excellently as a cathode [17,23] despite its high work function. The devices with the Au based tri-cathode-layer exhibit a slightly higher efficiency than that of the devices with the Ag based tri-cathode-layer, which may be mainly due to the higher transmission of the Au based tri-cathode-layer than that of the Ag based tri-cathode layer [17]. For example, at a typical application luminance level of 1000 cd/m², the devices with the Au tri-cathode-layer demonstrate an efficiency of 23 cd/A and 10.5 lm/W, higher than 21 cd/A and 9.1 lm/W for the devices with the Ag based tri-cathode-layer. Further optimization, for example, electrical doping the charge transporting layer [24], employing new emitters/host with high carrier mobility and efficiency, applying out-coupling techniques [25,26], can be employed to boost the device performance.

4. Summary

In conclusion, the interference effects of the microcavity can be greatly alleviated by reducing the reflection of the
top cathode. Au, modified by a 5 nm Yb electron injection layer, can not only be an excellent cathode for injecting electrons, but also exhibits lower reflection and higher transmission than that of the mostly used Ag top cathode. By employing the low reflection Yb (5 nm)/Au (15 nm)/MoO3 (35 nm) cathode, top-emitting WOLEDs with evenly separated R, G and B emission peaks have been demonstrated. At a luminance of 1000 cd/m², the top-emitting WOLEDs exhibit a CIE coordinate of (0.36, 0.42), a high CRI of 85 and an efficiency of 23 cd/A, 10.5 lm/W. As well, the spectra of the top-emitting WOLEDs demonstrate a very weak angle dependence due to the negligible interference effects. The top-emitting WOLEDs demonstrated here, which have been successfully applied in the Si based microdisplay, would be an ideal candidate for high resolution active-matrix display or indoor lighting applications.

Acknowledgments

The authors thank Prof. Wai-Yeung Wong’s group for providing the green-yellowish emitter 1, and Mr. Pengfei Sun for driving the microdisplay panel. This work was supported by the Hong Kong Government Research Grants Council Grant Number 614410 and AOE/P0308PG2.

References


