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Viewing-angle independence of white emission from microcavity top-emitting organic light-emitting devices with periodically and gradually changed cavity length

Yue-Feng Liu [a], Jing Feng [a,*], Da Yin [a], Hai-Feng Cui [a], Xu-Lin Zhang [a], Yan-Gang Bi [a], Dan-Dan Zhang [c], Lei-Shi Liu [d], Ai-Wu Li [a], Jun-Feng Song [a], Qi-Dai Chen [a], Hong-Bo Sun [a,b,*]

[a] *State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, People's Republic of China*

[b] *College of Physics, Jilin University, 119 Jiefang Road, Changchun 130023, People's Republic of China*

[c] *Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Institute of Functional Nano & Soft Materials, Soochow University, Suzhou 215123, People's Republic of China*

[d] *Suzhou Goodark Electronics Company Ltd., Suzhou 215123, People's Republic of China*

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ABSTRACT

We demonstrate a broad-bandwidth and viewing-angle independent white emission from top-emitting organic light-emitting devices (TOLEDs) by integrating a microstructured cavity with a periodically and gradually changed cavity length. The results indicate that the microstructured cavity can resolve the viewing angle-dependence effect persisted in the conventional planar TOLEDs. The viewing-angle independence in both broad-band emission spectra and Commision Internationale de L'Eclairage coordinates are obtained. Moreover, the microstructured white TOLEDs show comparable luminance and current efficiency to that of the planar TOLEDs.

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1. Introduction

White organic light-emitting devices (WOLEDs) have been attracting great attention due to their potential applications in low power consumption solid-state lighting sources and full-color flat-panel displays [1–4]. Compared to bottom-emitting WOLEDs, white top-emitting OLEDs (WTOLEDs) are considered more attractive in both lighting and display applications [5,6]. As lighting sources, WTOLEDs based on metallic anode instead of indium tin oxide possess low cost manufacturing potential. As displays, WTOLEDs integrated with a low-temperature poly-silicon thin film transistor active matrix backplane have been recognized as one of the best combination to achieve high-quality display image due to the fact that more complicated drive circuit is allowed underneath each of pixel without affecting its aperture ratio (AR) [7]. In addition, it is also well established that pixels with high AR invariably lead to prolonged operational stability owing to less current density required to drive each pixel in order to achieve a desired luminance [6]. Unfortunately, strong microcavity effect persisted in the TOLEDs results in narrowed emission bandwidth with strong angle-dependent emission spectra [8,9], which is an obstacle to achieve WTOLEDs. Thus, an angle-independent broadband emission is needed.

* Corresponding authors. Address: State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, People's Republic of China. Tel./fax: +86 431 85168281 (H.-B. Sun).

E-mail addresses: jingfeng@jlu.edu.cn (J. Feng), hbsun@jlu.edu.cn (H.-B. Sun).

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The TOLED is generally constructed from two metallic electrodes, one high-reflectivity anode and one semitransparent cathode, with organic layers sandwiched between them. The electrodes parallel to each other and form a Fabry–Perot resonator, which results in the strongly resonant microcavity effect [8,10]. Employing a low-reflectivity anode to alleviate the undesirable microcavity effect on the viewing characteristics has been proposed and demonstrated [7,11–14]. Introducing a refractive-index-matching layer on top of the semitransparent cathode has also been demonstrated with optimized viewing characteristics by decreasing the reflectance of the cathode [6,9,14–16]. However, all such demonstrations are unable to eliminate the microcavity effect but only minimize the microcavity effect by careful design of the metallic electrodes or its capping layer [6,7,11,12,15]. Despite its negative effect on the viewing characteristics, the microcavity resonance has a positive effect on the emission efficiency through enhancing the spontaneous emission intensity [17]. However, most of the above methods were taking effect through alleviating the microcavity effect, which would lead to a less spontaneous emission. Recently, a microstructured cavity with a periodically and gradually changed cavity length has been demonstrated in order to resolve the angular-dependence effect workable for monochromatic TOLEDs [18]. The resonant wavelength covering the whole visible range is possible in the microstructured cavity.

In this letter, the microstructured cavity has been applied into WTOLEDs to realize angle-independence broad-band emission. In the microstructured WTOLEDs, the shift of the peak emission wavelength with the viewing angle is successfully suppressed. Moreover, a desired broad emission spectrum and stable Commision Internationale de L'Eclairage (CIE) coordinates at various viewing angle are both obtained.

2. Experimental details

2.1. Fabrication of microstructure on substrate

A holographic lithography technique combined with filling process of the groove by spin coating of a polymer film has been employed to fabricate the WTOLEDs with periodic microstructure, which provides a simple approach with high controllability and reproducibility. Si substrates precoated with a 1600 nm-thick SiO_2 layer was cleaned using acetone, alcohol, and deionized water. Then photoresist (NOA63, Norland) diluted in acetone at a concentration of 25 mg/ml was spin coated on the substrate at 7000 rpm speed for 20 s. The lithography experiments were performed by using an all-solid-state, diode-pumped, continuous-wave, single-frequency 266 nm deep-ultraviolet laser. The sample was exposed for 900 ms by two laser beams which were split from the UV laser with beam size of ~6 mm in diameter. At last, they were developed in acetone for 30 s. The microstructure recorded on the photoresist film with different period and depth can be obtained by adjusting the writing angle and the exposure time. The morphologies of the microstructure were characterized by an atomic force microscopy (AFM, iCON, Veeko) in the tapping mode.

2.2. Fabrication and characterization of WTOLEDs

The fabrication progress of WTOLEDs with the microstructured cavity was shown in Fig. 1. Prepared Si substrates coated with corrugated photoresist film were immediately loaded into a thermal evaporation chamber. The Ag anode was grown at a rate of 1 Å s^{-1} at a base pressure of 5×10^{-4} Pa and the thickness is 80 nm. Then Ag film was exposed in ultraviolet-ozone for 70 s to obtain a thin Ag_2O as the anodic modification to improve the hole injection. Poly(*N*-vinyl carbazole) (PVK): 4,4′,4″-tris(3-methylphenylphe-nylamino)triphenylamine (m-MTDATA) (5 mg/ml) with doping ratio of 1:1 was then spun onto the Ag anode at 6000 rpm speed for 20 s for filling and smoothing the groove. PVK was a conducting polymer widely used in OLEDs as a hole-transport material, and the m-MTDATA doped into it for enhancing the hole injection and transport. The solvent is chloroform. Then the sample was baked in vacuum for 40 min at 60 °C to evaporate the organic solvent. After that, the sample was loaded into the thermal evaporation chamber again. The organic layers, LiF, thin Al and Ag cathode were deposited sequentially. The AFM images of surface morphology for the photoresist, Ag anode and PVK: m-MTDATA layers are shown in Fig. 2a–c, respectively. The period of microstructures is 2 μm, and the groove depth of the three layers is 86.77, 86.03 and 30.92 nm, respectively. Therefore, a variation range of ~55 nm for the gradually changed cavity length is obtained after deposition of the Ag cathode. The white emission of the WTOLEDs is realized by using two primary

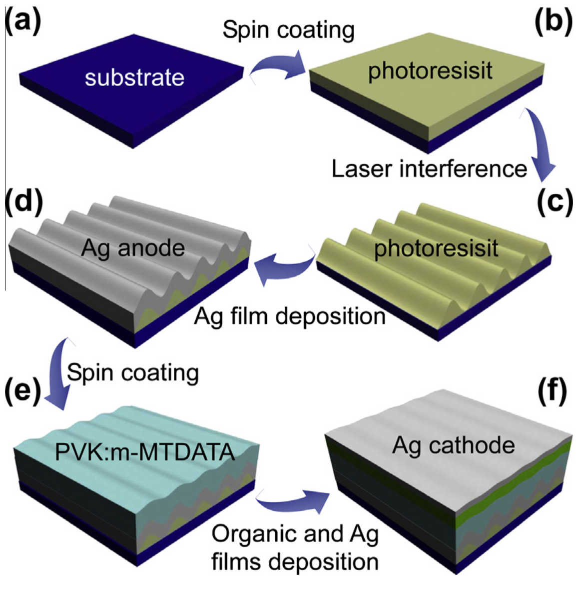

Fig. 1. Schematic fabrication process of a microstructured WTOLED. (a) Cleaning the glass substrate, (b) spin coating photoresist, (c) introducing periodic microstructure by two-beam laser interference lithography, (d) depositing Ag anode, (e) spin coating PVK: m-MTDATA and (f) depositing organic and cathode layers.

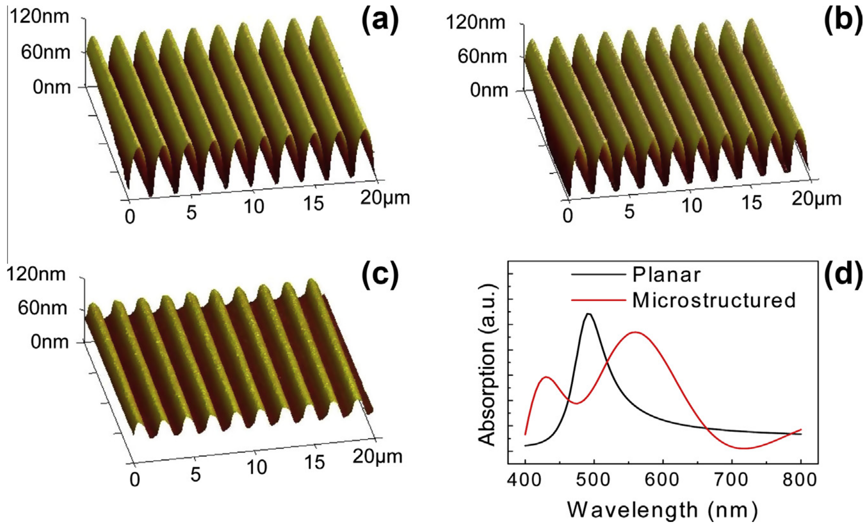

Fig. 2. Surface morphologies of photoresist layer (a), Ag anode (b), PVK: m-MTDATA layer (c) and (d) simulated absorption spectra of the planar and microstructured devices.

colors of blue and orange emission generated from emitting layer. An orange-emitting phosphorescent bis-(7,8-benzoquinolinato)iridium(III) (*N,N*′-diisopropyl-benzamidine) ($(bzq)_2Ir(dipba)$) [19] doped into a suitable complement deep-blue-emitting fluorescent complex bis(2-(2-hydroxyphenyl)-pyridine)beryllium) ($Bepp_2$) is used as the emitting layer. The structure of the WTOLEDs is Ag (80 nm)/PVK: m-MTDATA/*N,N*′-diphenyl-*N,N*′-bis(1-naphthyl)-(1,1′-biphenyl)-4,4′-diamine (NPB, 6 nm)/ $Bepp_2$:$(bzq)_2Ir(dipba)$ (3.5% wt, 20 nm)/$Bepp_2$ (25 nm)/LiF (1 nm)/Al (1 nm)/Ag (20 nm). The active area of the device is 2×2 mm^2. The electroluminescent (EL) spectra at different observation angle were measured by Fiber Optic Spectrometer, and a slit was used to limit the angular acceptance to $\sim 1°$. The characteristics of the devices were measured by Keithley 2400 programmable voltage–current source and Photo Research PR-655 spectrophotometer. All of the measurements were conducted in air at room temperature.

3. Results and discussion

Considering the microcavity effect of the TOLEDs, the phase shift of the light wave after one cycle in the microcavity is given by

$$-\Phi_b - \Phi_t + \frac{2\pi}{\lambda}\sum_i 2n_i d_i = 2m\pi \tag{1}$$

where λ is resonant wavelength, Φ_b and Φ_t are the phase changes upon reflection at the bottom and the top contact, respectively, d_i and n_i is the thicknesses and refractive indices of all organic layers within the cavity [20]. The resonant wavelength depends on $\sum n_i d_i$, namely the optical length of the cavity. The microstructure integrated into the cavity can provide the gradually changed cavity length. A wide resonant spectrum covering the whole visible range is expectable, if the variation range of the cavity lengths is sufficient. In order to verify if the variation range of 55 nm of the gradually changed cavity length is sufficient to WTOLEDs, absorption spectra of WTOLEDs with and without microstructures were calculated by scattering matrix approach. For the microstructures considered here, the structure of TOLED is divided into individual layers along the deposition direction of the device, each layer is either a diffraction grating or a uniform dielectric slab, and all grating slabs have the same periodicity [21]. The overall scattering of the TOLEDs is determined by firstly evaluating a matrix of scattering parameters for each individual layer through solving Maxwell's equation, and then forming a scattering matrix for the entire structure by the relevant boundary conditions. Given an incoming wave from the top side, the reflection waves have been simulated. Absorption spectra are obtained from complementary relation between reflection and absorption [22]. The simulated absorption spectra are shown in Fig. 2d. The absorption spectrum of the microstructured device is obviously broadened compared to that of the planar one, and almost covers the whole visible range. There are two peaks in the absorption spectrum of the microstructured devices. They are corresponding to resonance from the cavity formed at the crest and trough of the periodic corrugation, respectively, where stronger resonance could be formed compared to that of the side portion between the crest and trough. These two peaks correspond to the blue and orange emission region, respectively, and are suitable for two-color-based white emission. Unlike the groove depth, the period of the microstructure is not a very critical parameter for obtaining a desired bandwidth of the emission spectra [18].

The angular dependent EL spectra of the planar and microstructured devices are measured and shown in Fig. 3. The spectra of the planar device show obvious angular dependence. A 40 nm blueshift can be observed at blue emission region of spectra, when the viewing angle is increased from 0° to 60°. Moreover, the blue emission

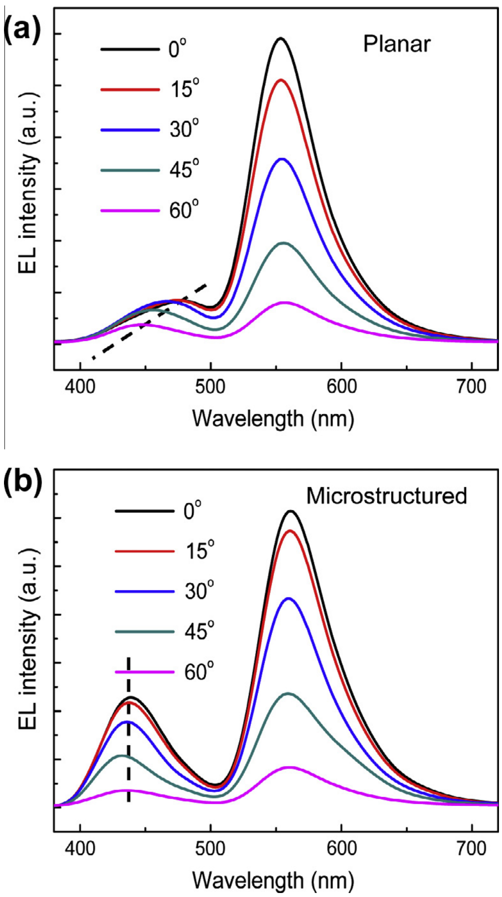

Fig. 3. EL spectra of the planar (a) and microstructured (b) WTOLEDs at viewing angles of 0°, 15°, 30°, 45°, 60° off the surface normal.

was restrained obviously at the forward direction and its intensity increased compared to the orange emission with the increasing of the observation angle. The resonant wavelength for the planar device is around 480 nm as can be seen from Fig. 2d, which is close to the blue region of the white TOLEDs. Therefore, the angular dependence occurs at the blue region. With the increasing of the observation angle, the resonant wavelength move to short wavelength and overlap with blue emission, which results in the increased intensity of the blue emission at larger observation angle compared to that of the orange emission. While for the microstructured WTOLEDs, the usual variation of the peak wavelength with viewing angles associated with microcavity effects has been eliminated. In addition, the blue emission was extracted successfully. The Commision Internationale de L'Eclairage (CIE) coordinates of both planar and microstructured WTOLEDs are shown in Fig. 4. The results indicate that the CIE coordinates of planar devices obviously deviate from the white range and have a large shift from (0.37, 0.52) to (0.34, 0.40) when the viewing angle is changed from 0° to 60°. While in the case of the microstructured devices, the variation of the CIE coordinates are within ±0.01, which are from (0.37, 0.42) to (0.36, 0.41) when the viewing angle changed from 0° to 60°. The photograph of the operating

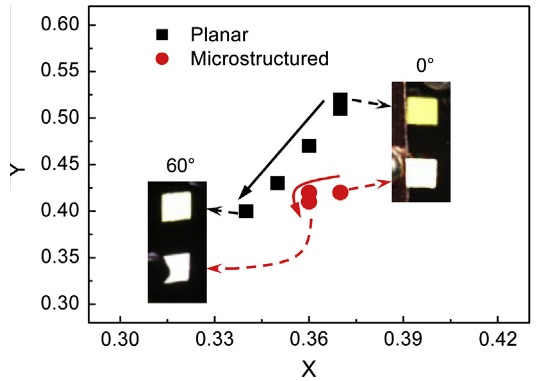

Fig. 4. CIE coordinates of the planar and microstructured WTOLEDs at different viewing angles. Inset: photograph of the operating WTOLEDs with and without the microstructure at observation angles of 0° and 60°.

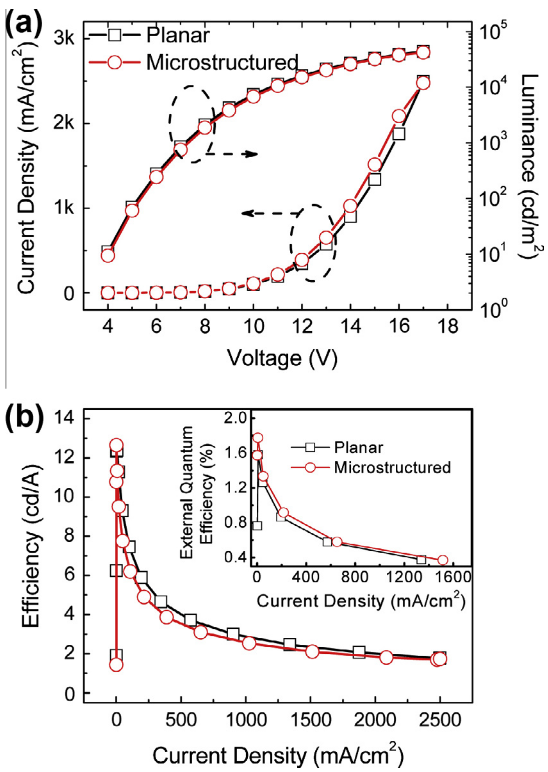

Fig. 5. (a) Current density–voltage–luminance and (b) efficiency-current density characteristics of the planar and microstructured WTOLEDs. Inset in (b): external quantum efficiency-current density characteristics of the planar and microstructured WTOLEDs.

WTOLEDs with and without the microstructure at observation angles of 0° and 60° are shown in the inset in Fig. 4. We can obviously observe a color shift in planar devices from yellowish to white. It is disadvantageous in practical application of the WTOLEDs. In contrast, no color variation can be observed by the naked eyes from the microstructured WTOLEDs, and a consistent white emission can be observed at different viewing angle. Such results suggest that viewing angle-independent broadband white emission can be realized by employing the microstructured cavity.

The EL performances of the planar and microstructured WTOLEDs are comparable. Current density–voltage–luminance and efficiency-current density characteristics of the planar and microstructured WTOLEDs are shown in Fig. 5. The maximum luminance and current efficiency of the microstructured devices at forward direction are 42150 cd/m^2 at 17 V and 12.65 cd/A at 6 V, respectively, while it is 44155 cd/m^2 at 17 V and 12.39 cd/A at 7 V, respectively, for the planar TOLEDs. Moreover, external quantum efficiency (EQE) has been calculated based on the measured EL spectra at different viewing angles [23,24]. The result is shown in the inset of Fig. 5b. The EQE of the microstructured WTOLEDs is higher than that of the planar WTOLEDs, which is attributed to the slower decrease of the emission intensity with the increasing viewing angle for the microstuctured devices as shown in Fig. 3. The above data indicates that the employing of the microstructured cavity with the gradually changed cavity length in the WTOLEDs not only eliminates the microcavity effect but also retains high EL performances.

4. Conclusions

In conclusion, WTOLEDs with angle-independent broadband emission have been demonstrated by integrating a periodic microstructure into the device structure to construct a cavity with gradually changed cavity length. In the microstructured WTOLEDs, the shift of the peak emission wavelength with the viewing angle is successfully suppressed. Moreover, a desired broad emissive spectrum and stable CIE coordinates are both obtained. The variation of the CIE coordinates is within ±0.01 when the viewing angle changed from 0° to 60°. The results indicate a WTOLED can be fabricated by the simple and effective method. It is essentially important for the applications of WTOLEDs in both display and solid-state lighting.

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