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White *p-i-n* organic light-emitting devices with high power efficiency and stable color

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Highly efficient *p-i-n* two-component white organic light-emitting devices have been fabricated with a thin dual emission layer system comprised of one codeposited emitting layer with blue and yellow dyes and one blue emitting layer, which gives rise to a balance white emission. The *p-i-n* white device achieved an electroluminescence efficiency of 10 cd/A and a power efficiency of 9.3 lm/W at 1000 cd/m^2 and a low voltage of 3.4 V with a Commission Internationale de l'Eclairage coordinates of (0.32, 0.43). The electroluminescent color of this *p-i-n* white organic light-emitting diode device has been shown to be immune to drive current density variations. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2784971]

Organic light-emitting diodes (OLEDs) are of considerable interest in recent years for flat panel display applications, particularly in white OLEDs (WOLEDs) which have attracted a lot of commercialization interests due to their demonstrated applications in the fabrication of full color displays with a color filter,[1] or as backlight for liquid crystal displays as well as in solid-state lightings.[2,3] WOLEDs coupled with color filter for full color displays can circumvent the problematic issues of high resolution shadow mask and achieve higher effective aperture ratio of pixels. However, with this approach, the color filters will invariably lower the brightness of the full color display. In order to obtain low power consumption as well as bright and sharp display image, the development of highly efficient and stable WOLEDs has become one of the major endeavors in the OLED community.[4]

White-light emission which requires the mixing of two complementary colors or three primary colors have been widely reported.[5–7] Careful control of the location of exciton recombination zone (RZ) and the energy transfer between the host and dopant molecules have been shown to be critical in obtaining a balanced white emission of high efficiency.[8,9] WOLEDs based on phosphorescent emitter systems[10,11] have the highest reported efficiencies which can achieve up to 100% internal quantum efficiency. However, most of these highly efficient phosphorescent OLEDs decrease rapidly with increasing drive current, and the operating reliability still requires further research and improvement.[10]

By introducing *p-i-n* structure to an OLED device, the operating voltage can be considerably reduced for both fluorescent[12] and phosphorescent[13] systems. The highly conductive *p*- and *n*-doped layers could enhance the charge injection from the contacts and reduce the Ohmic losses in these layers.[12] To further reduce the drive voltage in *p-i-n* OLEDs, the thickness of low conductive layer based on organic materials should be as thin as possible. However, it is difficult for WOLEDs with multiemission layer to achieve a stable white color due to the shift of RZ in thin organic layer, which often leads to undesirable $CIE_{x,y}$ color change with respect to drive current.

In this study, we demonstrate a dual emission layer (DEML) system for *p-i-n* WOLEDs in which the first emission layer is the codopant emitting layer with 2-methyl-9,10-*di*(2-naphthyl)anthracene (MADN):[14]5% *N*,*N*′-*bis*(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB):3% *p-bis*(*p*-*N*,*N*-diphenyl-aminostyryl)benzene (DSA-Ph):[14]0.2% rubrene (Rb) and the second one is a blue emitting layer of MADN:5% NPB:3% DSA-Ph. Three types of *p-i-n* WOLED devices have been fabricated, as depicted in Fig. 1. The total thickness of DEML in these *p-i-n* devices is only 15 nm. Device I is the *p-i-n* WOLED without codeposited NPB in the DEML system while device II is the *p-i-n* WOLED with the DEML system. The pure NPB layer of device II was replaced with 1,1-*bis*[*N*,*N*-di(*p*-tolyl)aminophenyl]cyclohexane[15] (TPAC) as electron blocker in device III. In our *p-i-n* architecture, 50% v/v tungsten oxide (WO_3) doped NPB and 2% cesium carbonate (Cs_2CO_3) doped 4,7-diphenyl-1,10-phenanthroline were used as the *p*-doped transport layer (*p*-HTL) and *n*-doped transport layer (*n*-ETL), respectively. In the DEML system, MADN, NPB, DSA-Ph, and Rb were used as host material, assistant dopant, blue, and yellow fluorescent dopants, respectively. For comparison, we also fabricated a conventional WOLED device with structure of ITO/CF_x/NPB (50 nm)/NPB:1.2% Rb (20 nm)/MADN:3% DSA-Ph (40 nm)/*tris*(8-quinolinolato)aluminum (Alq_3) (10 nm)/LiF (1 nm)/Al (150 nm), in which CF_x, NPB, and Alq_3 were

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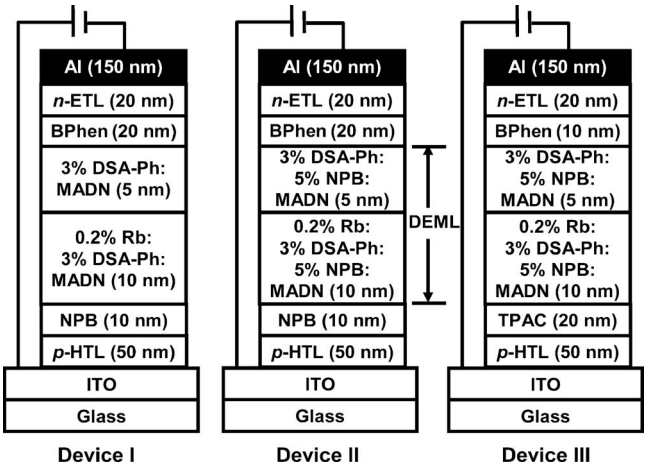

FIG. 1. Schematic device architecture of *p-i-n* WOLEDs.

used as the hole injection material,[16] hole, and electron transport materials, respectively.

It is known that Rb with lowest unoccupied molecular orbital/highest occupied molecular orbital of 3.2/5.4 eV can be a carrier trap for electrons, especially at low electric field,[17] which will cause the problematic white emission color change with various drive currents in thin emission layer of *p-i-n* two-component WOLEDs. In order to alleviate the unstable color issue associated with the carrier-trapping property of Rb, we have purposely coevaporated Rb with low doping concentration of 0.2% and 3% DSA-Ph in MADN, which would cause the yellow emission generated by the energy-transfer process from blue to yellow emitter. The energy-transfer process can be demonstrated by the solid-state emission spectra depicted in the inset of Fig. 2, the thin film composed of MADN:3% DSA-Ph:0.2% Rb emits intense yellow emission and relatively weak blue-greenish emission. It is evident that the emission of MADN around 430 nm essentially quenched and there is an effective energy-transfer characteristic from DSA-Ph to Rb, which is primarily due to the favorable spectral overlap between the emission peak of DSA-Ph and the absorption peak of Rb at 495 nm.[18]

Therefore, we designed and developed a DEML system by inserting a blue emitting layer of MADN:3% DSA-Ph

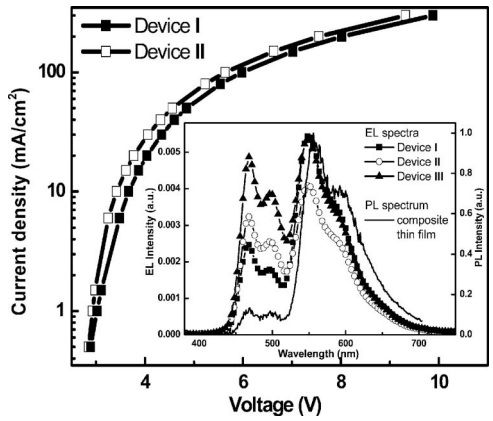

FIG. 2. Current density–voltage (*J-V*) characteristics of devices I and II. Inset: solid PL spectrum of composite thin film and EL spectra of devices I, II, and III at 20 mA/cm^2.

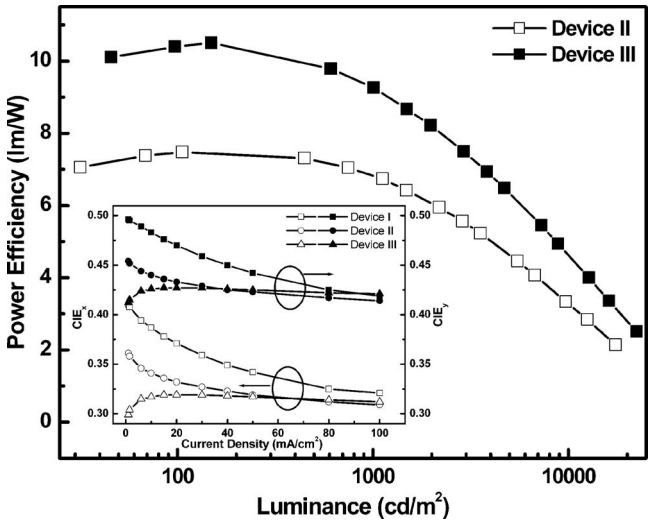

FIG. 3. Power efficiency vs luminance characteristics of devices II and III. Inset: $CIE_{x,y}$ coordinates vs current density characteristics of devices I, II, and III.

after the coevaporated EML into the structure of device I to enhance the blue emission intensity. From the inset of Fig. 2, the electroluminescence (EL) spectrum of device I indeed shows the enhancement of the blue emission intensity with respect to the solid PL of coevaporated thin film. However, the yellow emission intensity is still much higher than the blue emission, leading to undesirable white emission with $CIE_{x,y}$ coordinates of (0.37, 0.47). Furthermore, a significant EL color shift of device I is observed with respect to various drive currents as the $CIE_{x,y}$ coordinates are shifted from (0.410, 0.496) at 1 mA/cm^2 to (0.321, 0.419) at 100 mA/cm^2 with $\Delta CIE_{x,y} = \pm(0.089, 0.077)$, as shown in the inset of Fig. 3, in which the yellow emission intensity decreases with the increasing current density. We inferred the unstable EL color is due to the RZ shifts toward the blue emitting layer under high current stress.

Therefore, we codeposited 5% NPB in the DEML system of device II as the assistant dopant with the purpose of shifting the RZ toward the blue emitting layer to balance the blue and yellow emission intensities under low current density. Figure 2 reveals that device II also has a better *J-V* characteristic than device I which indicates that NPB molecules play an important role in enhancing the hole transport in the DEML system. In addition, the EL spectrum of device II depicted in the inset of Fig. 2 shows a more balanced white emission with $CIE_{x,y}$ of (0.33, 0.43) at 20 mA/cm^2 and the relative intensity of blue light has been increased as compared with device I. This phenomenon which exhibits the codeposited NPB molecules can indeed shift the RZ to the blue emitting layer and improve white $CIE_{x,y}$ coordinates. Moreover, the EL color shift with respect to varying drive currents has also been improved to $\Delta CIE_{x,y} = \pm(0.05, 0.04)$ from 1 to 100 mA/cm^2, as shown in the inset of Fig. 3. However, the reduced relative intensity of yellow emission leads to a lower EL efficiency of 7.4 cd/A at 20 mA/cm^2.

To further enhance the efficiency and improve the color stability of these *p-i-n* white devices, we turn to refine the exciton confinement in device III, in which the NPB layer

TABLE I. EL performances of white devices driven at 20 mA/cm^2.

Device	Voltage (V)	Yield (cd/A)	Efficiency (lm/W)	$CIE_{x,y}$
Conventional	6.4	9.0	4.4	(0.32, 0.41)
I	4.4	9.1	7.1	(0.37, 0.47)
II	3.6	7.4	6.4	(0.32, 0.43)
III	3.8	9.0	8.2	(0.32, 0.43)

was replaced by TPAC with a high LUMO energy level (2.0 eV) and high hole mobility (10^{-4}–10^{-3} cm^2/V s),[15] which can be an effective electron-blocking as well as hole transport material. Indeed, both blue and yellow intensities of device III have been enhanced, as shown in the inset of Fig. 2, and its emission achieved a white $CIE_{x,y}$ of (0.32, 0.43). Furthermore, the EL performance can be boosted to 9.9 cd/A and 8.2 lm/W at 20 mA/cm^2. It is also observed that the white emissive color becomes more stable with respect to drive current density as the EL color shift is only of $\Delta CIE_{x,y} = \pm(0.013, 0.009)$ from 1 to 100 mA/cm^2, as shown in the inset of Fig. 3.

Detailed EL performances of these devices measured at 20 mA/cm^2 are summarized in Table I. All *p-i-n* white devices show a much lower drive voltage and a dramatic gain in power efficiency as compared with those of conventional white device. Figure 3 shows the power efficiency versus luminance characteristics of devices II and III. Device III can achieve 10 cd/A and 9.3 lm/W at 1000 cd/m^2 which are considerably better than those of device II with 7.3 cd/A and 6.8 lm/W. It is noteworthy that both *L-V* and *J-V* curves of device III are steeper than those of conventional white device, as depicted in Fig. 4. The threshold voltage of device III is around 2.9 V. When driven at 6 V, device III can reach 10 000 cd/m^2 which is nearly eight times brighter than the conventional device. These results prove that the efficient exciton confinement is one of the most important factors in controlling the RZ shift under various drive currents and it is also indispensable for the development of a highly efficient and color stable *p-i-n* WOLED.

In summary, we have demonstrated highly efficient *p-i-n* WOLEDs with a dual emission layer (DEML) system. The codeposited NPB molecules in the DEML were used as the assistant dopant (NPB) to shift the RZ towards the blue emitting layer and balance the blue and yellow emission intensities. In addition, the refined exciton confinement of *p-i-n* WOLEDs has been shown to significantly improve the EL performance, giving rise to 10 cd/A and 9.3 lm/W at 1000 cd/m^2 and a stable white color under various drive conditions.

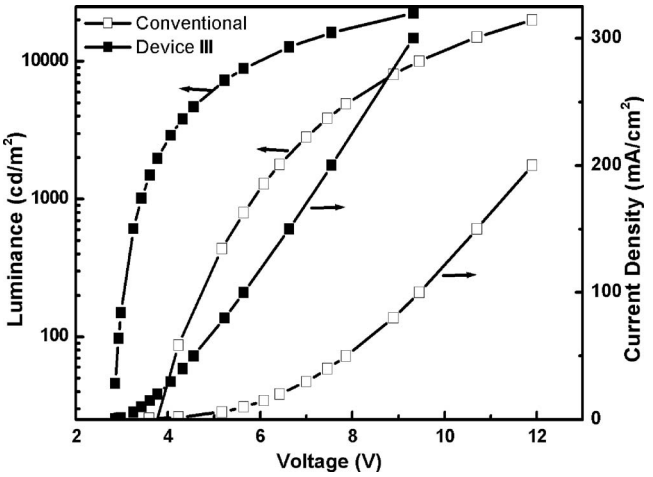

FIG. 4. Luminance–current density–voltage (*L-J-V*) characteristics of conventional white device and device III.

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