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Highly efficient white fluorescence/phosphorescence hybrid organic light emitting devices based on an efficient hole-transporting blue emitter

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ABSTRACT

4,4'-di-(1-pyrenyl)-4''-[2-(9,9'-dimethylfluorene)]triphenylamine was proved to be a hole-transporting blue emitter with a hole-mobility of 6.40×10^{-5} cm^2 V^{-1} s^{-1}. A blue organic light emitting device using the compound as a fluorescent dopant shows a maximum luminance efficiency of 5.8 cd A^{-1} (5.2 lm W^{-1}, 4.5%) with Commission Internationale de L'Eclairage coordinates of x = 0.16, y = 0.18. Fluorescence/phosphorescence hybrid white device employed the compound and $Ir(2\text{-phq})_3$ respectively as blue and orange emitter has been fabricated, which shows a warm white emission with a maximum efficiency of 16.8 cd A^{-1} (16.1 lm W^{-1}). Moreover, high efficiency three-color fluorescence/phosphorescence hybrid white organic light emitting device using the compound as blue fluorescent emitter combining with phosphorescent yellowish-green emitter ($Ir(ppy)_2bop$) and red emitter ($Ir(piq)_3$) has been achieved. The white device shows a maximum efficiency of 42.5 cd A^{-1} (38.2 lm W^{-1}, 15.5%), with a Color-Rendering Index of 76, which is among the best results for previously reported fluorescence/phosphorescence white devices.

1. Introduction

White organic light-emitting diodes (WOLEDs) have attracted considerable interest for applications in large flat panel displays and in solid-state lighting [1–5]. In the early days, various approaches have been devised to realize WOLEDs, such as all-fluorescence WOLEDs and all-phosphorescence WOLEDs [6–8]. WOLEDs based on all-fluorescence emitters typically have a lower efficiency, because fluorescence emission is theoretically capped to an internal quantum efficiency of about 25% due to singlet-triplet statistics [9]. While, fully phosphorescent WOLEDs have obtained the highest reported efficiencies for capturing both singlet and triplet excitions to achieve 100% internal quantum efficiency [10–14]. However, one bottleneck for further enhancing the efficiency of fully phosphorescent WOLEDs is the performance of blue phosphorescent emitters, which lags far behind those of green and red phosphorescent emitters [15]. Although many blue phosphorescent emitters have been reported, the lifetimes of the WOLED based on the common widely used blue phosphorescent emitters, iridium(III) bis[(4,6-difluorophenyl)-pyridinato-N,$C^{2'}$] picolinate (FIrPic), are still very poor [16]. Moreover, the color purity of the WOLEDs based on FIrPic is always compromised due to its blue-green emission with two peaks centered at 472 and 500 nm, respectively [17,18]. Thus, the fluorescence/phosphorescence (F/P) hybrid WOLEDs, which use efficient blue fluorescent emitters to replace phosphorescent emitters, have been proposed [19–22].

Till now, there are several strategies to obtain highly efficient F/P hybrid WOLEDs. One strategy is the spatial separation of singlet and triplet excitons by using an interlayer, which is introduced between fluorophor and phosphor, such as reported by Sun et al., [19] which is one of the widely-used approaches to fabricate F/P hybrid WOLEDs. However, the complicated device structure implies additional complexity and cost for the mass production of WOLEDs. Another method is to use a blue fluorescent emitter with high triplet energy level, in which the blue fluorescent emitter not only acts as a blue-emitter but also functions as the host for the

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phosphor, so that the high triplet energy of the blue emitter is indispensable to sensitize phosphors. For example, as reported by Hung et al., [23] using a bipolar material which has a deep blue fluorescence and serves as the host for the yellow-green phosphor, simultaneously, the device achieves an efficiency of 15.5 cd A^{-1}. However, it is known that there are many difficulties associated with obtaining blue emitters with both high PL quantum efficiency and high triplet energy level, this type of material is still very rare to date. To see the original construction of F/P white OLEDs, which has a very simple device structure of blue fluorescent/red and green or orange phosphorescent emitter but shows low efficiency. For instance, Li et al. demonstrated a hybrid WOLED employing anthracene as the fluorescent emitter combined with $Ir(ppy)_3$ and $Ir(piq)_2(acac)$ as phosphor to obtain an efficiency of 6.4 cd A^{-1} at 100 cd m^{-2} [24]. The low efficiency may be ascribed to the location of excitons recombination zone, which principally confined in the blue fluorescent emitter. To improve the performance of the original F/P WOLEDs using this simple structure, it is essential to obtain blue emitters, which possess not only high efficiency pure-blue emission but also excellent hole-transporting property.

In this work, we demonstrate an excellent fluorescence/phosphorescence white OLED with simple device structure. 4,4'-di-(1-pyrenyl)-4″-[2-(9,9'-dimethylfluorene)]triphenylamine is chose as fluorescent emitter for its high hole mobility and good blue emission. The hole mobility of 4,4'-di-(1-pyrenyl)-4″-[2-(9,9'-dimethylfluorene)]triphenylamine is calculated to be 6.40×10^{-5} cm^2 V^{-1} s^{-1}. Then a two-color F/P hybrid white OLED using DPFA and $Ir(2\text{-phq})_3$ has been fabricated and gives a warm white emission with a maximum efficiency of 16.8 cd A^{-1} (16.1 lm W^{-1}), a CIE coordinates of (0.47, 0.40) and a correlated color temperature (CCT) of 2430 K. To further improve the performance of the white OLED, three-color F/P hybrid white OLED, using blue fluorescent emitter (DPFA), phosphorescent yellowish-green emitter ($Ir(ppy)_2bop$) and red emitter ($Ir(piq)_3$) has been achieved. The white OLED shows a maximum efficiency of 42.5 cd A^{-1} (38.2 lm W^{-1}, 15.5%) with a CIE coordinates of (0.38, 0.45) and CRI of 76. Performance of the white OLED is among the best reported and the work also demonstrate a strategy to fabricated highly efficient white F/P hybrid white OLEDs with simple device structure.

2. Experimental

Indium tin oxide (ITO) coated glasses with a sheet resistance of 15 Ohm per square was used as substrates. Before device fabrication, the ITO glass substrates were pretreatment carefully by washing with isopropyl alcohol and deionized water, dried in an oven at 120 °C over 1 h and then treated with ultraviolet-ozone for 25 min before loaded in a vacuum deposition chamber with a base vacuum better than 10^{-6} Torr. Organic layers were successively deposited on the ITO glass substrates with a rate of 1–2 Å/s. Hole/electron-injecting layer of MoO_3/LiF and cathode of Al were respectively deposited with the rates of 0.1 and 10 Å/s.

The electroluminescence (EL) spectra, CIE coordinates, current density-voltage-luminance (J-V-L) characteristics and correlated color temperature (CCT) were all measured with a computer-controlled Keithley 2400 source meter and a Spectrascan PR655 photometer under ambient atmosphere.

3. Results and discussion

In this paper, a series of device based on a blue emitter DPFA have been fabricated and investigated [25]. The hole-mobility of DPFA was obtained by using its hole-only device. Meanwhile, DPFA acted as fluorescent dopant and give a pure-blue emission with optimized device structures. Moreover, high efficiency fluorescence/phosphorescence (F/P) hybrid white OLED has been achieved by using the good hole-transporting and excellent blue-emitting property of DPFA.

Hole-only device was fabricated with the structure of ITO/MoO_3 (10 nm)/DPFA (80 nm)/MoO_3 (10 nm)/Al. In the device, most of electrons can be restrained due to the large energy gap between the LUMO of MoO_3 (2.3 eV) and the work function of Al cathode (4.3 eV). Holes can be injected from the anode to the organic layer, thus the hole-mobility of DPFA can be obtained. For comparison, NPB based hole-only device was fabricated with the same structure of ITO/MoO_3 (10 nm)/NPB (80 nm)/MoO_3 (10 nm)/Al. Fig. 1 (insert) depicts the current density versus voltage (*J-V*) characteristics of the two hole-only devices. The remarkable higher hole-current density in the DPFA based device is clearly obvious to demonstrate that DPFA is capable of good hole-transporting property. To further investigate the hole-mobility, SCLC method is used for the hole-only device [26], which is described as:

$$J = \frac{9}{8}\varepsilon_r\varepsilon_0\mu\frac{V^2}{L^3} \tag{1}$$

and

$$\mu = \mu_0 \exp\left(\beta\sqrt{\frac{V}{L}}\right) \tag{2}$$

Where ε_r is relative permittivity ($\approx$3.00), ε_0 is vacuum permittivity ($\approx 8.85 \times 10^{-14}$ F cm^{-1}), μ is the carrier mobility determined by SCLC measurements, L is the cathode–anode distance (100 nm for the device), μ_0 is zero-field mobility and β is a coefficient that is proportional to the Poole-Frenkel factor [27]. The value of μ_0 and β of DPFA were calculated to be 1.81×10^{-5} cm^2 V^{-1} s^{-1} and 1.33×10^{-3} $(V\ cm^{-1})^{-1/2}$ by linear fitting and the fitting data is listed in Fig. 1. By using the obtained values, the field-dependent holes mobility can be obtained by eq (2). Fig. 1 shows the plot of hole-mobility against electric field ($E^{1/2}$) character. The hole-mobility of DPFA was obtained with 6.40×10^{-5} cm^2 $V^{-1}s^{-1}$ at high electric field of 0.9 MV cm^{-1}, which is little higher than that of NPB (2.6×10^{-5} cm^2 $V^{-1}s^{-1}$ at the same electric field).

High efficiency blue-emitting OLED (device B1) with a configuration of ITO/MoO_3 (0.5 nm)/TAPC (50 nm)/DPFA (30 nm)/TPBi (30 nm)/LiF/Al has been developed. Fig. 2 shows the EL spectra of device B1 and B2 at 1000 cd m^{-2} and the molecule structure of DPFA (insert). The device shows a pure-blue emission with a peak centered at 464 nm along with the CIE coordinates of CIEx = 0.14 and CIEy = 0.18. Fig. 3 shows the current efficiency, external

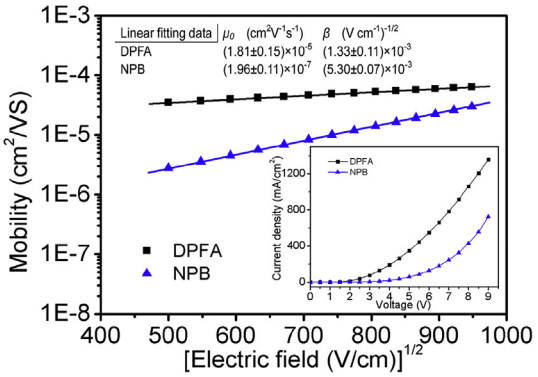

Fig. 1. Dependence of hole mobility on the electric field, the insert curve and data are J-V characteristics of hole-only devices and linear fitting data, respectively.