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Balanced white organic light-emitting diode with non-doped ultra-thin emissive layers based on exciton management

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ABSTRACT

The non-doped ultra-thin emissive layer (EML) is a promising novel structure for fabricating organic light-emitting diodes (OLEDs). We attempt to perform exciton management in order to obtain a highly efficient and balanced light-emitting phosphorescent red–green–blue (RGB) white OLED (WOLED). These studies include optimization of the locations of the three RGB EMLs based on investigations concerning the spatial distribution of excitons, along with the energy transfer mechanisms between different materials. More importantly, by introducing a 1,1-bis((di-4-tolylamino)phenyl) cyclohexane (TAPC) interlayer, which acts as a hole-trapping layer, a redistribution of excitons is realized, giving rise to a more balanced white light emission. This simple method is an efficient way to adjust the exciton distribution, and could therefore be used in future OLED devices.

1. Introduction

Organic light-emitting diodes (OLEDs) have attracted significant attention recently as a result of their favorable properties, such as impressive color rendering, wide viewing angles, and high compatibility with flexible substrates [1,2]. The major focus of OLED research is now on white organic light-emitting diodes (WOLEDs) as such devices can be used for future full-color displays and large-area lighting sources [3,4]. In general, white emission in OLEDs can be realized by blending complementary colors (such as blue and orange) or primary colors (such as red, green, and blue) emitted from single or multi-emissive layers [5,6]. This light-emissive layer is usually a host–guest system that facilitates energy transfer from the wide-bandgap host to the dopants [7]. A precise doping concentration, although difficult to guarantee, is critical to overall device performance.

Indeed, simplifying the emissive layer (EML) structure does help to improve the manufacturing yield in WOLED production. It has been shown that by introducing an ultra-thin layer of pure orange phosphorescent dyes between the hole-transporting layer and the electron-transporting layer, an OLED with higher efficiency than that of the host–guest doped orange device can be realized [8]. Zhao et al. explored this novel concept and found that the non-doped, EML-based OLED structure might possibly be widely applied to many phosphorescent dopants [9]. Further, high-efficiency mono-color OLEDs that incorporate ultra-thin, non-doped EMLs have been demonstrated [10]. Such OLEDs have many merits, such easy optimization and host independence [9]. Unfortunately, only a few systematic investigations have been conducted into utilization of this promising structure in WOLEDs.

Taking both efficiency and balanced light emission into consideration, three RGB ultra-thin EMLs should be carefully located in the WOLED structure. In this paper, we investigate the uneven spatial distribution of excitons in our single-layer WOLED, along with the mechanism of energy transfer that leads to interaction between different EMLs. In addition, we optimize the location of non-doped RGB EMLs. Further, by introducing a thin interlayer of 1,1-bis((di-4-tolylamino)phenyl) cyclohexane (TAPC), which acts as a hole-trapping layer, exciton redistribution is realized, which facilitates the acquisition of a more balanced light emission. This proves to be a simple but efficient way to manage the exciton distribution, and should easily lend itself to use in future research.

2. Experimental details

All organic materials were electroluminescence grade and were used as received. PEDOT:PSS (Clevios AI4083) was used to form a hole-injecting layer (HIL). TAPC, 1,3-bis (9-carbazolyl) benzene (mCP), and 1,3,5-tris(2-N-phenylbenzimidazolyl) benzene (TPBi) served as an electron/exciton blocking layer, a phosphorescent host material, and a hole/exciton blocking layer, respectively. Note that TAPC plays a critical role in realizing exciton redistribution. mCP facilitates charge transport and is responsible for energy

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transfer to the emissive dopants. The basic device configuration is ITO/PEDOT:PSS/TAPC/mCP/EML/TPBi/LiF/Al. The phosphorescent blue-emitting dopant used in this study was bis(3,5-difluoro-2-(2-pyridyl) phenyl-(2-carboxypyridyl) iridium III (FIrpic). Tris(2-phenylpyridine) iridium(III) ($Ir(ppy)_3$) was used as a green phosphorescent dopant. The red dopant was a commercial iridium-based red dye.

The pre-patterned ITO substrates (Kintec Company, Hong Kong) were cleaned with deionized water and sonicated successively in acetone and isopropanol 15 min each. Samples were further treated with 20 W oxygen plasma (PE100 from Plasma Etch, Inc.) for 2 min prior to PEDOT:PSS deposition. PEDOT:PSS was filtered through a 0.45 μm PVDF membrane syringe. A 25 nm PEDOT:PSS layer was then formed by spin-coating the solution at 9000 rpm for 60 s. The TAPC, EML, TPBi, LiF, and top Al layers were thermally deposited at an operating pressure of $<5 \times 10^{-6}$ Torr. The deposition rate was monitored using quartz crystal microbalances. Current–voltage (IV) and luminescence–voltage (LV) characteristics were measured via a computer-controlled Keithley 2400 SMU and a BM-7A luminance colorimeter. All measurements were carried out in ambient atmosphere and at room temperature.

3. Results and discussion

Energy diagrams for each material used in this study are shown in Fig. 1. Under an applied electric field, holes were injected from the anode (ITO) while electrons were injected from the cathode (LiF/Al). The corresponding hole (or electron) injection barrier was determined by the energy level difference between the work function of the electrode and the highest occupied molecular orbital (HOMO) of TATC (5.5 eV [11]) or the lowest unoccupied molecular orbital (LUMO) of TPBi (2.9 eV [12]). Charge carriers migrated through mCP and formed excitons, which eventually recombined in the phosphorescent dyes, thereby generating light emission. Energy transfer occurred between mCP and the phosphorescent dye.

In order to determine the optimal location of the three RGB, ultra-thin, non-doped EMLs in this single-layer structure, we first fabricated a series of individual RGB devices to explore the spatial distribution of excitons. A 0.2 nm EML was used as a probe layer and, by varying the location of this probe layer (represented as "x" in Fig. 2(a)), the approximate exciton distribution inside the mCP layer was obtained. As proved in previous research, molecules of the interlayer are not in the form of a neat layer but partially penetrate into the host material when the interlayer is as thin as 0.5 nm [10]. Therefore, we assumed that the influence of this extremely thin probe layer on the overall distribution of excitons is insignificant. The maximum external quantum efficiencies (EQEs) associated with various probing locations are shown in Fig. 2(b). The maximum EQE peak for all three individual RGB devices appeared around $x = 16$ nm, indicating that the exciton recombination zone is primarily located at the mCP/TPBi interface.

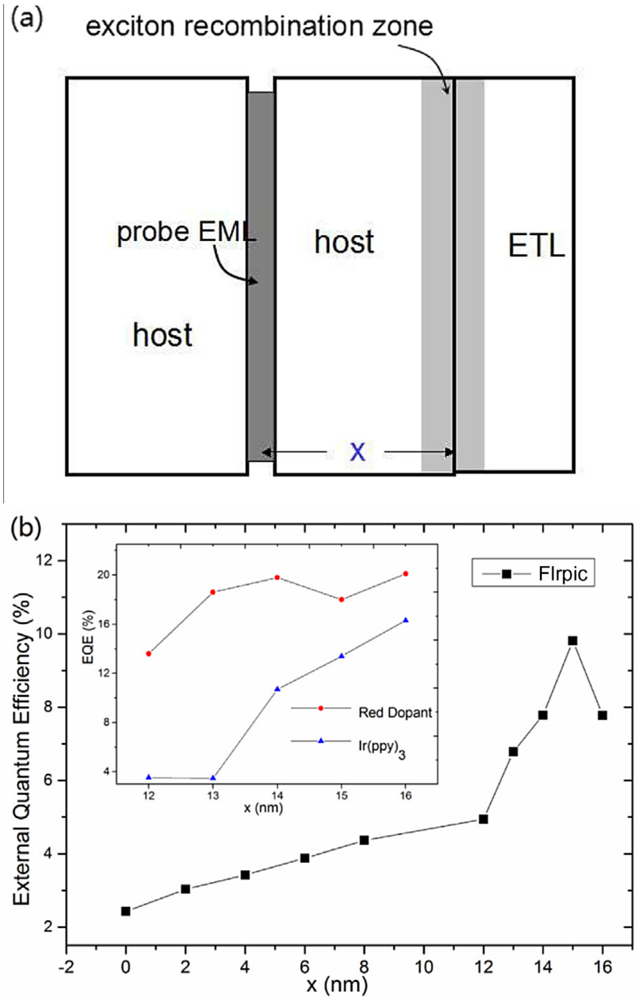

Fig. 2. (a) Schematic diagram of individual device structures with a probe emissive layer (EML) (red, green, or blue). (b) Maximum external quantum efficiency (EQE) as a function of the recombination location (x) of the ultra-thin non-doped EML with respect to the TAPC/mCP interface. The device structure is ITO/PEDOT:PSS (25 nm)/TAPC (20 nm)/mCP (x)/FIrpic or red dopant or $Ir(ppy)_3$ (0.1 nm)/mCP ($16 - x$ nm)/TPBi (40 nm)/LiF (1 nm)/Al (100 nm). ETL: electron-transporting layer. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

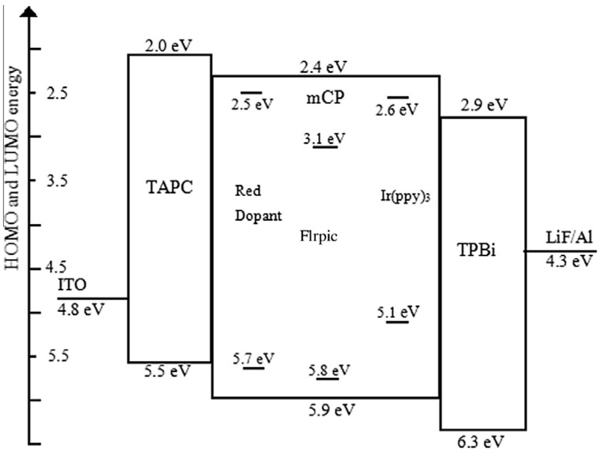

Fig. 1. Energy diagrams of the different materials used.

Exciton recombination experienced a considerable reduction once the probe layer's location was moved away from the mCP/TPBi interface. This reduction can be attributed to the fact that the hole mobility of mCP (i.e., 1.2×10^{-4} cm^2/Vs) is much larger than its electron mobility (i.e., 3.4×10^{-5} cm^2/Vs) [13]. In the case of blue-emitting devices, the recombination rate decays to 1/2 of the original (8–9% EQE) when the location of the probe layer is moved 4 nm away from the mCP/TPBi interface, indicating that triplet–triplet annihilation (TTA) is rather significant in this FIrpic-doped mCP EML as a result of high exciton density. Similar decay in the recombination rate is observed in green-emitting devices. In the case of red-emitting devices, this kind of decay is less serious. This may be due to less energy being needed to stimulate red emission. The number of excitons in $13 \leqslant x \leqslant 15$ nm