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High efficiency phosphorescent white organic light-emitting diodes with an ultra-thin red and green co-doped layer and dual blue emitting layers

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ABSTRACT

Phosphorescent white organic light emitting diodes (WOLEDs) with a multi-layer emissive structure comprising two separate blue layers and an ultra-thin red and green co-doped layer sandwiched in between have been studied. With proper host and dopant compositions and optimized layer thicknesses, high-performance WOLEDs having a power efficiency over 40 lm/W at 1000 cd/m^2 with a low efficiency roll-off have been produced. Through a systematic investigation of the exciton confinement and various pathways for energy transfer among the hosts and dopants, we have found that both the ultra-thin co-doped layer and two blue emitting layers play a vital role in achieving high device efficiency and controllable white emission.

1. Introduction

WOLEDs are currently being utilized for both display and lighting applications. Ever since their first demonstration, the research focus has been on improving the WOLED efficiency, brightness, and lifetime. To produce high efficiency WOLEDs, phosphorescent emitters are indispensable, as they provide a pathway of achieving emission with a nearly 100% internal quantum efficiency. Significant enhancement in efficiency has also been realized in various device layer architectures, including a single-layer emitter with multiple color dopants [1,2], a multiple-layer emitter consisting of two or more adjoining emitting layers [3–7], and hybrid WOLEDs [8–10]. To obtain multi-fold improvements in both lifetime and brightness, tandem structures are often implemented in WOLEDs at the expense of layer complexity [11,12].

To date, most research interest in WOLEDs is focused on multiple-layer emitter structures because they provide better control of the recombination and emission processes enabling a higher efficiency and superior whiteness. From the perspective of fabrication, it is much easier to adopt an emitter structure in a WOLED consisting of two broadband emitting layers producing complementary blue-green and orange-red color layers. In contrast, WOLEDs with three primary colors tend to produce white with better color rendering index [13–15]. Introduction of an extra layer to accommodate three emitters, however, makes it challenging to manage interlayer charge transport and energy transfer between the various hosts and dopants. Those processes not only control the emission efficiency and the color balance, but also affect color stability at various driving voltages [7,16–21].

In this paper, we describe a WOLED with a triple-layer emitter structure consisting of an ultra-thin co-doped red and green layer sandwiched in between two blue emitting layers. By tailoring the doping concentration and layer thicknesses, we can control the exciton energy transfer amongst the hosts and dopants. Our device structure produces WOLEDs with an extremely high EQE (over 20%) and a power efficiency of 40 lm/W at 1000 cd/m^2 and 3.7 V. At the same time, the color variation is minimal over a wide range of emission intensities.

2. Materials and methods

All devices were fabricated by thermal evaporation at a base

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pressure of 10^{-6} torr (without breaking vacuum) on patterned indium-tin-oxide (ITO) (110 nm thickness, 15 Ω/sq sheet resistance) on glass substrates. Prior to film deposition, the substrates were cleaned in ultrasonic deionized water and organic baths sequentially, followed by an O_2 plasma treatment.

To reduce the operating voltage, 1,4,5,8,9,11-hexaazatriphenylene-hexanitrile (HATCN) was deposited on top of pre-cleaned ITO substrates as the hole injection layer. 1,1-bis((di-4-tolylamino)phenyl)cyclohexane (TAPC) was chosen as the hole transporting material. The electron transporting material 1,3,5-Tri(*m*-pyridin-3-ylphenyl)benzene (TmPyPB) was doped with cesium carbonate (Cs_2CO_3) to help increase electron injection efficiency from the cathode. Hole transporting material 4,4,4-tris(N-carbazolyl)triphenyl-amine (TCTA) and bipolar transporting materials 2,6-bis(3-(carbazol-9-yl)phenyl)-pyridine (DCzPPy) were chosen as the two host materials for blue emitter iridium(III) bis(4,6-difluorophenyl-pyridinato-N,C^2) picolinate (FIrpic). In between the two blue emitting layers, red emitter bis(2-phenylquinoline)-(acetylacetonate)-iridium(III) ($Ir(2\text{-}phq)_2(acac)$) and green emitter fac-tris(2-phenyl-pyridinato)-iridium(III) ($Ir(ppy)_3$) were doped into an ultra-thin TCTA layer. The deposition rate of each organic layer was monitored by quartz crystal sensors via a side aperture on the boats. Selected because of their relatively high triplet energy levels, TAPC (2.9 eV) and TmPyPB (2.78 eV) serve to confine the triplet excitons generated in FIrpic (2.62 eV), $Ir(ppy)_3$ (2.4 eV) and $Ir(2\text{-}phq)_2(acac)$ (2.0 eV) within the emitting layers. These triplet energy levels are indicated (in parenthesis) in the energy level diagram as shown in Fig. 1(a) along with the LUMO and HOMO energy levels (labelled on top and bottom of rectangles) for the sequence of layers from TAPC to TmPyPB. For clarity, the corresponding WOLED configuration including the layer thicknesses and dopant concentrations is shown in Fig. 1(b).

The current density-voltage (J-V) data of the devices were measured by a Keithley 2400. A SpectroScan PR650 was employed to measure the electroluminescent (EL) data and Commission Internationale de l'Eclairage (CIE) color coordinate. UV-Vis absorption spectra were obtained using a Perkin Elmer Lambda 900 spectrophotometer. Photoluminescence (PL) spectra were recorded on a Hitachi F-4600 fluorescence spectrophotometer.

3. Results and discussion

The WOLED structure for this study is as follows: ITO (110 nm)/HATCN (3 nm)/TAPC (37 nm)/TCTA: FIrpic (4 nm)/red-green co-doped layer (0.5 nm)/DCzPPy:FIrpic (4 nm)/TmPyPB (10 nm)/TmPyPB:Cs_2CO_3 (30 nm)/Al (100 nm). The thickness of each individual layer was optimized to achieve the highest external quantum yield possible without necessarily increasing the drive voltage.

The injected holes enter the emitting layers first through the blue (TCTA:FIrpic) layer and then the red-green (TCTA:$Ir(2\text{-}phq)_2(acac)$:$Ir(ppy)_3$) layer. Since both of these layers use TCTA, a hole-transport material, as the host, the majority of holes are expected to traverse these two layers. The injected electrons enter the emitting layers through the blue (DCzPPy:FIrpic) layer, where DCzPPy, a bipolar-transport material, is the host. As shown in Fig. 1(a), the energy offsets between TCTA and DCzPPy are substantial (0.35 eV for HOMO and 0.16 eV for LUMO), providing a suitable interface to localize electron-hole recombination. Due to this specific arrangement for the emitting layers, the long-lived triplet excitons formed as a result of these recombination events can effectively diffuse in the TCTA and DCzPPy hosts and are subsequently redistributed between the blue, green and red dopants commensurate with their concentrations in these hosts and their distance from the TCTA/DCzPPy interface.

We fabricated four devices, B_1/B_2, B_1/R&G/B_2, B_1/R/B_2, and B_1/G/B_2, having different composite emitter layers. B_1/B_2 is a blue device with two different blue emitting layers as the composite emitter comprised of 15% FIrpic doped TCTA (B_1) and 20% FIrpic doped DCzPPy (B_2). B_1/R/B_2, B_1/G/B_2 and B_1/R&G/B_2, are devices with three emitting layers as the composite emitter where an ultra-thin red, green or red and green (co-doped) emitting layer is inserted between the blue emitting layers B_1 and B_2, respectively. The red doping concentration is fixed at 2% and the green doping concentration is fixed at 6% for these three devices. The thickness of this interlayer is only 0.5 nm and the dopant concentrations were adjusted to produce a balanced white emission with high efficiency.

Fig. 2 shows the plot of external quantum efficiency (EQE) versus current density for the four devices. Table 1 summarizes the performance data at 5 mA/cm^2. It can be seen that all four devices exhibit high EQE ranging from 17.5% for the blue device B_1/B_2 to 20.3% for predominately green device $B_1/G/B_2$. The drive voltages for these devices are also very similar, approximately 3.8 ± 0.1 V (at 5 mA/cm^2). The power efficiency (PE, lumens per watt) varies substantially due to a large variation in emission colors from these devices, ranging from 31 lm/W for B_1/B_2 to 52 lm/W for $B_1/G/B_2$. The B_1/R&G/B_2 device provides a warm white emission with color co-ordinates of (0.458, 0.448) that shift only slightly over a luminance range of 400–4000 cd/m^2. In contrast, device B_1/R/B_2 shows

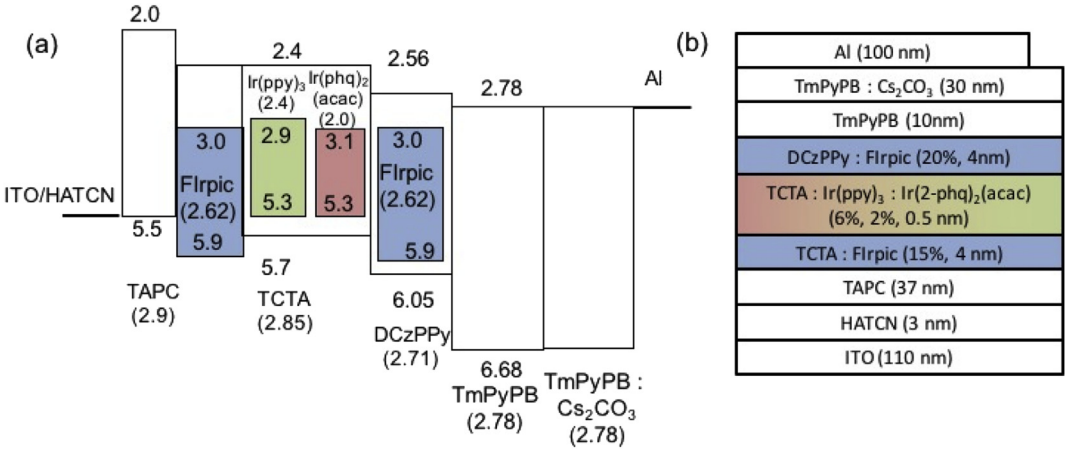

Fig. 1. Energy level diagram and device architecture of a WOLED with an ultra-thin red, green co-doped emitting layer (LUMO and HOMO energy levels are labelled on top and bottom of rectangles, triplet energy levels are indicated in parenthesis). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)