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Efficient non-doped monochrome and white phosphorescent organic light-emitting diodes based on ultrathin emissive layers

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ABSTRACT

Efficient red, orange, green and blue monochrome phosphorescent organic light-emitting diodes (OLEDs) with simplified structure were fabricated based on ultrathin emissive layers. The maximum efficiencies of red, orange, green and blue OLEDs are 19.3 cd/A (17.3 lm/W), 45.7 cd/A (43.2 lm/W), 46.3 cd/A (41.6 lm/W) and 11.9 cd/A (9.2 lm/W). Moreover, efficient and color stable white OLEDs based on two complementary colors of orange/blue, three colors of red/orange/blue, and four colors of red/orange/green/blue were demonstrated. The two colors, three colors and four colors white OLEDs have maximum efficiencies of 30.9 cd/A (27.7 lm/W), 30.3 cd/A (27.2 lm/W) and 28.9 cd/A (26.0 lm/W), respectively. And we also discussed the emission mechanism of the designed monochrome and white devices.

1. Introduction

White organic light-emitting diodes (OLEDs) exhibit great potential application in the solid-state lighting and full-color display due to their light weight, flexibility, high resolution, wide viewing angle [1–6]. Lots of device concepts have been proposed to improve the performance of white OLEDs, such as doping multiple dyes into one single emission layer (EML) [7–10], using multiple EMLs in which each layer emits different color light [11–14], using excimer or exciplex emission [15,16], using tandem or stacked structure [17]. So far, nearly all state of the art monochrome and white phosphorescent OLEDs were fabricated by the method of coevaporation [18–20]. As we all know, the doping method is beneficial to decreasing the severe concentration quenching effect, leading to high efficiency and low efficiency roll-off. But it also puts a few obstacles in the commercialization, such as accurately controlling the doping concentration by adjusting relative deposition rates of the host and guest, searching for the appropriate bipolar host materials with high energy to efficiently confine the excitons [21,22], which aggravate the complication in the manufacture process and increase the cost of the devices fabrication [23].

Since the non-doped white OLEDs were firstly demonstrated by Tsuji et al. [24], they have attracted increasing interest [25–31]. There are considerable advantages to be gained by using non-doped method to fabricate white OLEDs, such as the save of the materials, the simplified fabrication process, the better repeatability and the lower production cost, which make non-doped method more suitable for the industrial application for solid-state lighting and display. Liu et al. fabricated non-doped phosphorescent OLEDs based on double quantum-well structures with maximum power efficiency of 30.5 lm/W [28]. Chen et al. reported non-doped white OLEDs using ultrathin bluish-green and red dyes with maximum current efficiency of 7.4 cd/A [29]. Recently, Zhao et al. demonstrated efficient ultrathin nondoped blue/orange, blue/green/red, and blue/green/orange/red white OLEDs with maximum current efficiencies of 41.3 cd/A, 34.6 cd/A and 21.1 cd/A, respectively [31]. Moreover, the emission mechanism of the OLEDs using ultrathin nondoped emissive layers was still not clear. Generally speaking, to obtain efficient white non-doped OLEDs, the carrier injection layer, the transport layer and the spacer or interlayer between different color dyes are always necessary. But the more organic function materials used, the higher the cost. So it's very meaningful and necessary to develop white OLEDs with simplified device structure.

In this paper, we have performed a systematic study on how to realize efficient and stable two colors, three colors and four colors non-doped white OLEDs based on ultrathin emission layers. And all designed orange/blue, red/orange/blue and red/orange/green/blue white OLEDs exhibit very stable spectra and high efficiencies. Moreover, the working mechanism of the OLEDs was also discussed and we concluded that the position of the recombination region located at the interface of hole transport layer (HTL) and

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the electron transport layer (ETL). The stable energy transfer was the determinate factor for achieving such excellent spectra and high efficiency.

2. Experimental

Fig. 1 shows the chemical structures of red, orange, green and blue phosphorescent dyes, the detailed energy level diagram of the materials, and the structures of the two colors, three colors, and four colors white OLEDs. All the organic materials used in our work were purchased from Luminescence Technology Corporation.

Indium tin oxide (ITO) with sheet resistance of 20 Ω per square, covered by 2 nm MoO_3 was used as anode. ITO substrates were cleaned in an ultrasonic bath with acetone, ethanol and deionized water in sequence, and then treated in O_2 plasma. 1,3-Bis (carbazol-9-yl)benzene (MCP) and 1,3-Bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPhB) served as the HTL and ETL, respectively. In order to further simplify the device structure, MCP was also used as the spacer between EMLs to adjust the relative emission intensity. Bis(2-methyldibenzo[f,h]quinoxaline)(acetylacetonate)-iridium(III) [$Ir(MDQ)_2(acac)$], Bis(4-phenylthieno[3,2-c]pyridinato-N,C2′) acetylacetonate iridium(III) (PO-01), Tris(2-phenylpyridine)iridium(III) [$Ir(ppy)_3$], and Bis(3,5-difluoro-2-(2-pyridyl) phenyl-(2-carboxypyridyl)iridium(III) (FIrPic) were used as red, orange, green and blue phosphorescent dyes, respectively. All OLEDs were fabricated in a single chamber tool with the substrate temperature of ~30 °C by high vacuum thermal evaporation under the pressure of $\sim 3 \times 10^{-4}$ Pa. In this work, we used in situ quartz-crystal monitor to monitor the thickness of the vacuum depositions. Typical deposition rates range from 0.001 to 10 Å/s. The deposition rate could be controlled by temperature variation of the source material oven. The HTL and ETL were grown at the deposition rate of ~1 Å/s, while the phosphorescent ultrathin EMLs were deposited at the rate of ~0.01 Å/s. 100 nm Al cathode was defined through a shadow mask with an active area of 3 mm × 3 mm. The electroluminescence (EL) characteristics were measured using a programmable Keithley 2400 source measure unit and PR655 spectroscan spectrometer. All the devices were measured without encapsulation under ambient conditions.

3. Results and discussion

To demonstrate efficient OLEDs with ultrathin layer, it is very important to place the ultrathin EML at the main exciton recombination region to efficiently utilize the electrically generated excitons. We firstly fabricated four devices as shown in Fig. 2(a) to investigate the position of the recombination region. Fig. 2(b)–(d) shows the normalized EL spectra at the voltage of 5 V, the current efficiency–voltage, and luminance–voltage characteristics of devices A–D. As can be seen that devices A–C all show a primary orange emission from PO-01. The peak current efficiencies of devices A–C are 5.3 cd/A, 18.3 cd/A and 37.4 cd/A, indicating that the majority of excitons are generated at the narrow interface of MCP/BmPyPhB.

The efficient orange emission of PO-01 in device B may be attributed to two possible channels: (i) the generated excitons at the narrow region diffuse through BmPyPhB or MCP and migrate into the ultrathin EML where they transfer to PO-01. (ii) From the energy level diagram in Fig. 1(a), we can see that PO-01 acts as deep traps for both holes and electrons. So the charge carriers

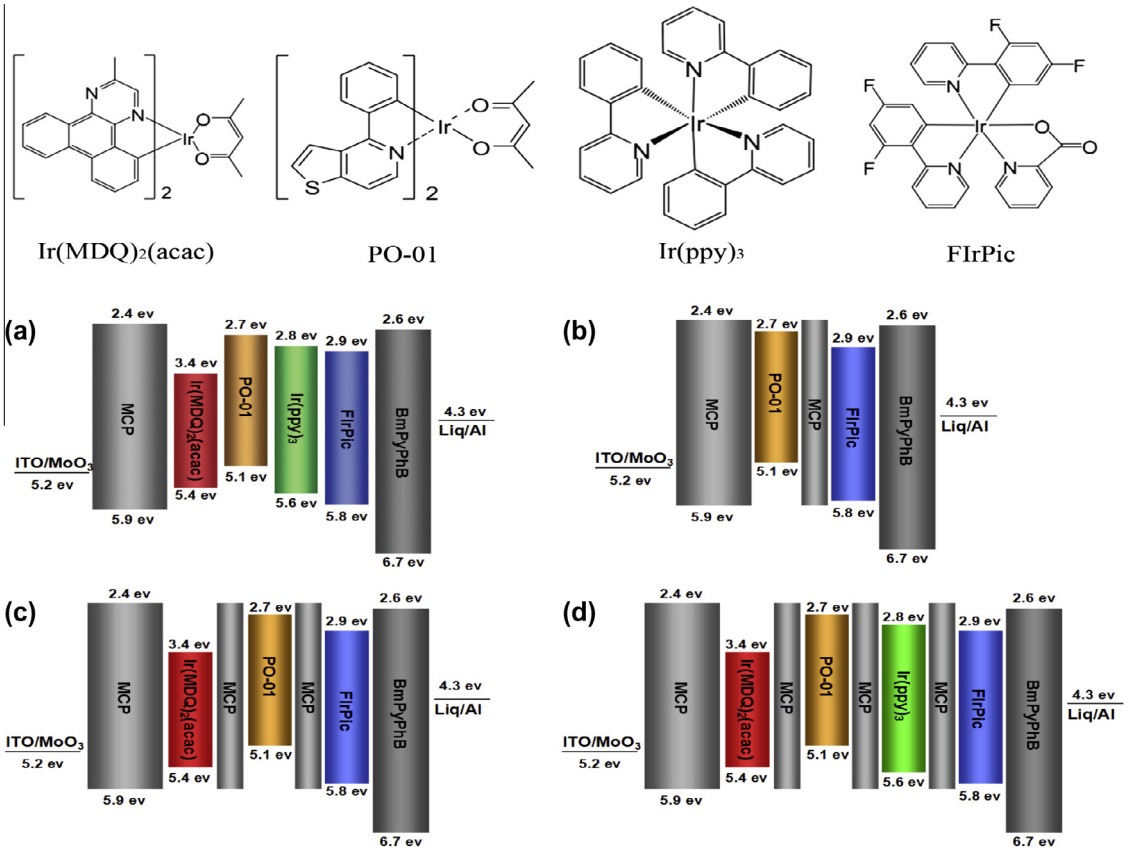

Fig. 1. The chemical structures of phosphorescent dyes, (a) the detailed energy level diagram of the materials and the structures of (b) two colors, (c) three colors, and (d) four colors white OLEDs. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)