



## Full Length Article

# Manipulation of recombination zone by utilizing the donor of electroplex as a spacer

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## ABSTRACT

The emission of electroplex is varied with applied voltages, leading to poor color stability of devices. In this work, the recombination region was investigated in organic light-emitting diodes (OLEDs) based on the electroplex between N,N'-diphenyl-N,N'-bis(1-naphthyl-phenyl)-1,1'-biphenyl-4,4'-diamine (NPB) and 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,2,4-oxadiazole (PBD). Color stability was manipulated by introducing the donor of electroplex as a spacer, which plays an important role of recombination zone for electroplex-based OLEDs. The 5-nm-spacer-device brought out a tolerable chromaticity coordinate shift of less than 0.01 for both x- and y-value. White light emission with CIE coordinates of (0.37,0.28) and (0.33,0.26) at 12 V was achieved when the thickness of spacer was 7 and 10 nm, respectively. The 7-nm-spacer-device exhibited better color stability than the 10-nm-spacer-device. This proves that adjusting spacer thickness is a simple and efficient method to control recombination region and can be useful for fulfilling color-stable electroplex-based white emission.

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## 1. Introduction

In organic light-emitting diodes (OLEDs), the light is generated by the decay of monomer excitons under an electric field. Nevertheless, the emission of other species, such as excimers and exciplexes [1–5], can be also observed because of intermolecular interactions of organic materials. An excimer, originally short for excited dimer, is formed between the same molecules. An exciplex, formed between different kinds of molecules, is originally short for excited complex. The emission of excimers and exciplexes can be detectable in both photoluminescence (PL) and electroluminescence (EL). Unlike of the above two species, the emission of another excited state, electroplex, is observed under high electric field but not under photo-excitation [6–11], which is the reason that the term “electro” was used. Meanwhile, this excited state species shows some characteristics of exciplex since they are both formed between molecules from different kinds. Therefore, a compound of “electro” combined with “plex” was explored to define this phenomenon.

Electroplex was discovered in the 1990s [6,7]. As one of charge-transfer complexes (CTCs), electroplex is also an association of two or more molecules, or of different parts of one large molecule, in which a fraction of electrons is transferred between the molecular

entities. In OLEDs, electroplex can be usually formed at an interface of electron-transfer and hole-transfer materials. The source molecule from which the electron is transferred is called donor (D) and the receiving species is called acceptor (A). The emission of electroplex originates from direct optical transition between the lowest unoccupied molecular orbital (LUMO) of the acceptor and the highest occupied molecular orbital (HOMO) of the donor [7]. Consequently, the resulting emission is red-shifted compared to that from either of the constituents and is considerably broadened, which can be utilized to tune emission color and achieve a white emission [11–14]. To date, white OLEDs (WOLEDs) based on bimolecular excited species (excimer/exciplex/electroplex) are considered to be a simple and cheap technique [12–16]. The utilization of these excited state emissions is vitally important for achieving high-performance WOLEDs. However, this technique suffers the drawback of color shift with different applied voltages, which is also present in most WOLEDs [16–18].

To improve the color stability of WOLEDs, a thin spacer layer is usually introduced between different emissive regions, especially for hybrid WOLEDs incorporating fluorophore with phosphor [19]. Spacers can control energy transfer between different emitters and facilitate balancing the charges within the devices, resulting in fewer color-shifts of OLEDs [20,21]. Unfortunately, few investigations were conducted into the effect of a spacer on the performance of electroplex-based OLEDs so far.

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In this paper, OLEDs were fabricated based on the electroplex between *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl-phenyl)-1,1'-biphenyl-4,4'-diamine (NPB) and 2-(4-biphenyl)-5-(4-*tert*-butylphenyl)-1,2,4-oxadiazole (PBD). The fundamental device structure is ITO/NPB/DCJT/ NPB/PBD/Al. NPB close to ITO (indium tin oxide) anode, labeled by NPB<sub>1</sub>, acts as hole-transport layer (HTL). DCJT, 4-(dicyanomethylene)-2-*tert*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran, serves as a red emitter. NPB between DCJT and PBD functions as the donor of the electroplex and a blue emitter as well as a spacer between the red emitter and the electroplex, labeled by NPB<sub>2</sub>. PBD plays roles of electron-transport layer (ETL) and the acceptor of the electroplex. The recombination zone and color stability were investigated by varying the thickness of the spacer. The experimental results demonstrated that the introduction of a spacer layer is an efficient approach to control carrier distribution, leading to manipulating the exciton recombination zone of electroplex-based OLEDs.

## 2. Experiments

The detailed device structures in this work are ITO/NPB<sub>1</sub> (40-*x* nm)/DCJT(0.3 nm)/NPB<sub>2</sub>(*x* nm)/PBD(50 nm)/Al, depicted in Fig. 1. The values of *x*, namely the thickness of spacer (NPB<sub>2</sub>), are 3, 5, 7 and 10, respectively. The corresponding devices are labeled as devices I–IV.

Prior to the device fabrication, the patterned ITO-coated glass substrates were conventionally cleaned by ultrasonic bath in detergent, deionized water, acetone and alcohol, finally treated by UV-ozone for 10 min. The cleaned ITO substrates were loaded into a vacuum chamber, and thermally deposited NPB, DCJT and PBD in turn under a pressure of  $2\text{--}5 \times 10^{-4}$  Pa. All materials, with a purity of over 99%, are commercially available and used without further purification. The thicknesses and rates are monitored *in situ* by a 6 MHz quartz crystal oscillator during deposition. Then, the substrates were transferred into another chamber without breaking vacuum to thermally evaporate Al cathode from a tungsten boat under a pressure of  $3 \times 10^{-3}$  Pa. The overlap of ITO and Al (3 mm × 3 mm) is defined as active area of samples.

The EL spectra were measured by a charge-coupled device (CCD) spectrometer with a Keithley model 2410 source under air atmosphere at room temperature. The PL spectra of NPB, PBD and NPB/PBD films on quartz glass substrate were recorded by Fluorolog-3 spectrometer.

## 3. Results and discussion

EL spectra (normalized to the longest-wavelength peak) of devices I–IV under an applied voltage of 10 V are displayed in Fig. 2(a). All EL spectra are composed of multi-emission bands and cover the entire visible spectrum. Obviously, the relative contribution of short-wavelength emission (mainly belonging to NPB emission) increases with the thickness of NPB<sub>2</sub> spacer. This is easily understood since more excitons can be recombined in NPB<sub>2</sub> spacer for the devices with thicker spacer. From CIE coordinates of Fig. 2(b), it can be seen that the color of devices gradually shifts from orange to white as the thickness of NPB<sub>2</sub> spacer increases from 3 to 10 nm, i.e. from device I to IV.

In order to confirm the origins of each emission band, EL spectra are decomposed into three curves by Gaussian fitting, presented in Fig. 3. The peaks of three fitting curves are located at  $445 \pm 5$ ,  $487 \pm 7$  and  $615 \pm 10$  nm, originating from the emission of NPB<sub>2</sub> (peak1) [8], electroplex (NPB<sub>2</sub><sup>+</sup>PBD<sup>-</sup>) (peak2) [8,9] and DCJT (peak3) [22–24], respectively.

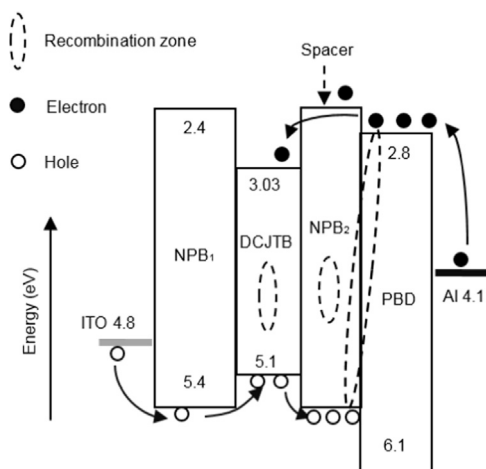


Fig. 1. Schematic diagram of device structure and energy levels in this work.

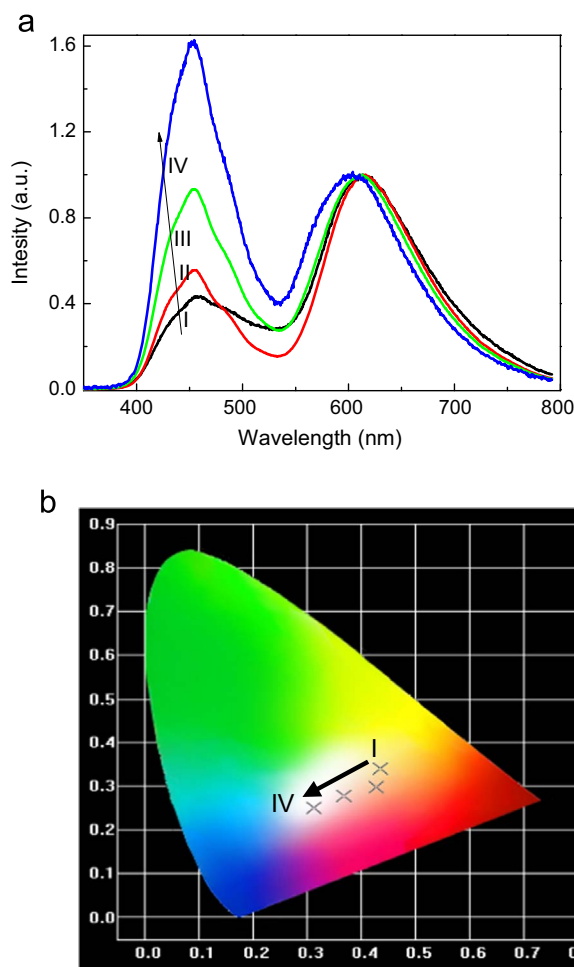


Fig. 2. (a) Normalized EL spectra and (b) CIE coordinates of devices I–IV under 10 V. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

According to the energy level diagram shown in Fig. 1, holes injected from ITO anode can be partly confined in DCJT due to its high HOMO level (i.e. closer to vacuum level) relative to that of NPB [25]. Some of holes transfer into NPB<sub>2</sub> spacer and accumulate at NPB<sub>2</sub>/PBD interface due to hole blocking effect of PBD induced by its low-lying HOMO level [26]. On the other hand, some of electrons injected from Al cathode accumulate at NPB<sub>2</sub>/PBD interface due to electron blocking effect of NPB induced by its high

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