



# Study of organic light-emitting diodes with exciplex and non-exciplex forming interfaces consisting of an ultrathin rubrene layer



Xu Wang, Run Wang, Dianli Zhou, Junsheng Yu\*

State Key Laboratory of Electronic Thin Films and Integrated Devices, School of Optoelectronic Information, University of Electronic Science and Technology of China (UESTC), Chengdu 610054, PR China

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## ABSTRACT

Fluorescent organic light-emitting diodes (OLEDs) with exciplex and non-exciplex forming interfaces, where an ultrathin 5,6,11,12-tetraphenyl-naphthalene (rubrene) emissive layer was sandwiched, were fabricated. The performances of 4,4',4''-tris(*N*-carbazolyl)-triphenylamine (TCTA)/1,3,5-tris(*N*-phenylbenzimidazole-2-yl)benzene (TPBi) exciplex-type device and 4,4'-di(9*H*-carbazol-9-yl)biphenyl (CBP)/TPBi no-exciplex-type device were optimized in terms of the thickness of rubrene ultrathin layer. The results showed that the exciplex-type device yielded high device efficiency, attributing to the utilization of triplet excitons as the triplets of the exciplex can be up-converted into singlets via reverse intersystem crossing. The light emission process in exciplex-type device was dominated by energy transfer, while the non-exciplex type device was dominated by charge trapping emission mechanism, which was verified by the single carrier devices.

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## 1. Introduction

Organic light-emitting diodes (OLEDs) are considered as the potential next generation display and lighting source, and a large volume of works have been carried out to develop high performance OLEDs. Especially, fluorescent OLEDs have been extensively studied, due to their advantages over phosphorescent OLEDs in terms of low cost, long device lifetime and short exciton lifetime [1]. Generally, the electroluminescent (EL) performance of fluorescent OLEDs is very sensitive to the doping ratio of fluorescent emitters. Meanwhile, fluorescent emitters have strong concentration quenching effect caused by intermolecular interactions [2]. Therefore, most fluorescent emitters can neither be fabricated into non-doped thick emissive layer (EML) nor be heavily doped into a host material. The optimal doping concentration for fluorescent emitters is often less than 5 wt.% [3], while it is a challenge to accurately control low doping ratios so as to obtain high repeatability in large scale industrial production. As for the fabrication of EML, compared with conventional host-guest doping method, the employment of ultrathin EML can solve the above problems [4,5].

Recently, exciplex-type hosts have attracted lots of attentions. Generally, exciplex emission provides broad emission spectrum

and is not good for color purity. Some works also use exciplex to generate very stable white light emission in OLEDs [6]. The exciplex is well known as a charge-transfer (CT) state formed between electron-donors and electron-acceptors upon photo- and electrical excitation [7]. The exciplex has a small singlet-triplet ( $S_1-T_1$ ) energy gap ( $\Delta E_{ST}$ ), which is low enough to boost thermally activated delayed fluorescence, in order to break the 25% theoretical internal quantum efficiency limit of fluorescent OLEDs [8,9]. Some breakthroughs have already been made by using exciplex forming host structure or using energy transfer from interfacial exciplex. By doping a fluorescent emitter into an exciplex forming host of 4,4',4''-tris(*N*-3-methylphenyl-*N*-phenylamino)-triphenylamine (m-MTDATA) and 4,7-diphenyl-1,10-phenanthroline (Bphen), a high performance fluorescent OLED was reported by Zhou et al. [10]. Recently, Kim et al. also reported highly efficient fluorescent OLEDs by doping a fluorescent emitter into a exciplex forming mixed host of 4,4',4''-tris(*N*-carbazolyl)-triphenylamine (TCTA) and 4,6-bis[3,5-(dipyrid-4-yl) phenyl]-2-methylpyrimidine [11]. Above all, most works contain host-host mixing and host-guest doping methods. In order to avoid the above methods to simplify manufacturing process, a doping-free configuration needs to be developed.

Herein, to simplify fabrication procedures and to gain more insight into the emission mechanisms, we constructed OLEDs with exciplex and non-exciplex forming interfaces. TCTA and 1,3,5-tris(*N*-phenylbenzimidazole-2-yl) benzene (TPBi) were used to form the exciplex interface and an ultrathin fluorescent EML,

\* Corresponding author.

E-mail address: [jsyu@uestc.edu.cn](mailto:jsyu@uestc.edu.cn) (J. Yu).

5,6,11,12-tetraphenyl-naphthalene (rubrene), was inserted in between this exciplex forming interface for emission. Moreover, 4,4'-di(9*H*-carbazol-9-yl) biphenyl (CBP) and TPBi were used to construct a non-exciplex forming interface for comparison. The emission mechanisms of both types of OLEDs were studied in detail. It was demonstrated that energy transfer mechanism was the dominant emission mechanism in exciplex-type device, while the main emission mechanism in non-exciplex-type device was charge trapping.

## 2. Experimental

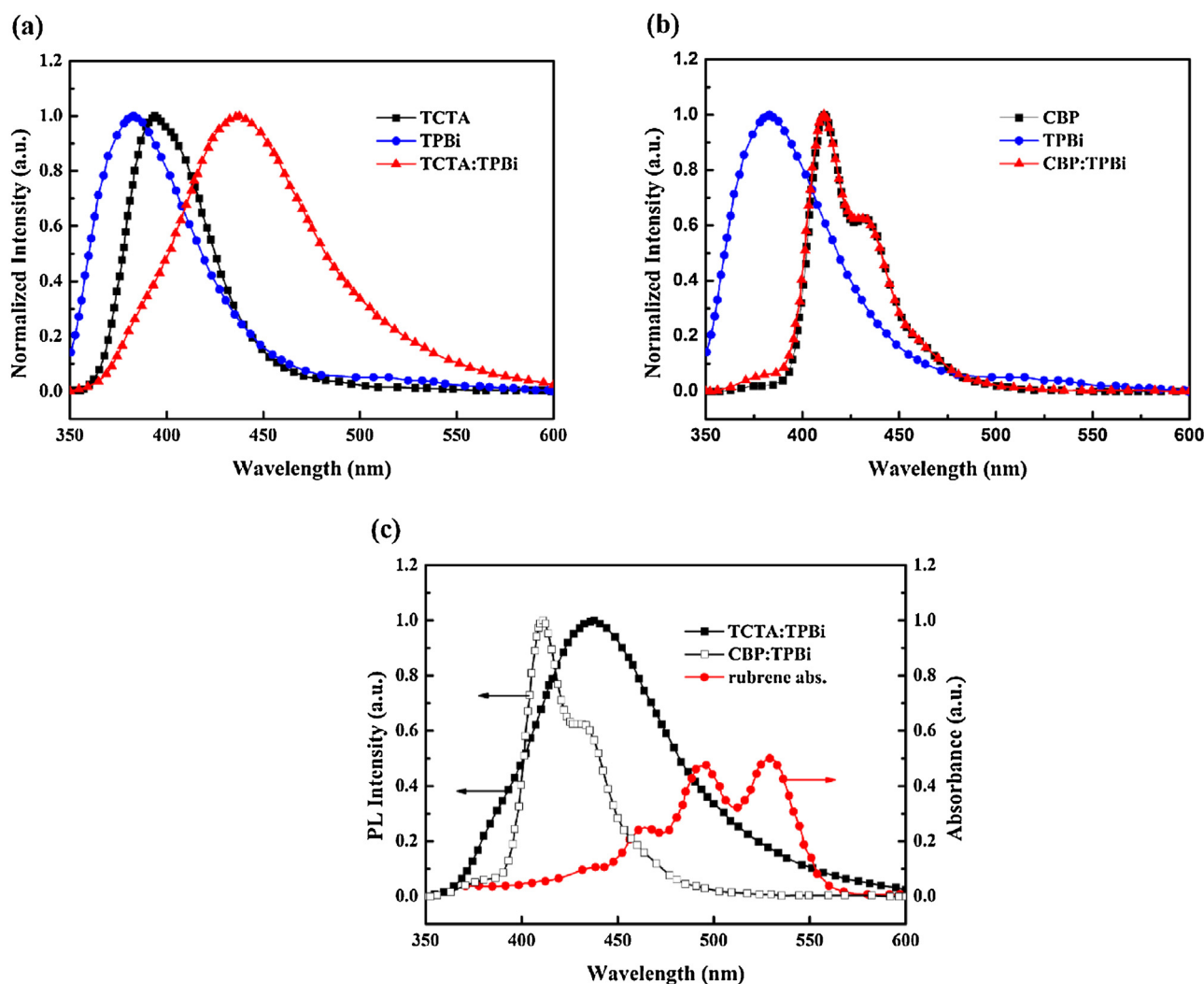
Indium tin oxide (ITO) coated glass substrates with a sheet resistance of  $15 \Omega/\text{sq}$  were cleaned in detergent water, acetone, deionized water and isopropyl alcohol by using an ultrasonic bath for 30 min, respectively. Afterwards, the substrates were dried with nitrogen gas flow and then, they were treated with  $\text{O}_2$  plasma in a vacuum chamber under a pressure of 25 Pa for 5 min, to clean the surface and enhance hole injection ability of ITO. Organic and metallic layers were subsequently deposited in separate chambers under a pressure of  $3 \times 10^{-4}$  and  $3 \times 10^{-3}$  Pa, respectively, without breaking the vacuum. Deposition rates and thin film thicknesses were monitored in situ using a quartz crystal oscillator. Luminance-current density-voltage (*L*-*J*-*V*) curves were measured using

Keithley 4200 source meter and an ST-86LA luminance meter. Electroluminescence (EL) spectra and the Commission Internationale de l'Eclairage (CIE) coordinates of the devices were recorded with an OPT-2000 spectrophotometer. The photoluminescent (PL) spectra of the organic films were recorded with the OPT-2000 spectrophotometer at an excitation wavelength of 365 nm. The absorption spectra of the organic films were measured with a SHIMATZU UV-1700 UV-vis spectrophotometer. All measurements were performed in air under ambient condition without encapsulation.

## 3. Results and discussion

### 3.1. EL characteristics

The normalized PL spectra of TCTA, TPBi and TCTA:TPBi films are depicted in Fig. 1a. The molar ratio of TCTA to TPBi in the mixed film is 1:1. The TCTA:TPBi film shows a broad and featureless PL spectrum that is bathochromic-shifted from the intrinsic TCTA and TPBi emission spectra. Meanwhile, the energy of the TCTA:TPBi emission peak is  $\sim 2.9$  eV, which is similar to the energy difference between the highest occupied molecular orbital (HOMO) level of TCTA and the lowest unoccupied molecular orbital (LUMO) level of TPBi, proving that TCTA and TPBi molecules form an exciplex upon



**Fig. 1.** (a) PL spectra of TCTA, TPBi and TCTA:TPBi thin films. (b) PL spectra CBP, TPBi and CBP:TPBi thin films. (c) Normalized UV-vis absorption spectra of rubrene, along with the PL of TCTA:TPBi and CBP:TPBi.

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