

Essential electro-optical differences of exciplex type OLEDs based on a starburst carbazole derivative prepared by layer-by-layer and codeposition processes



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ABSTRACT

The star-shaped compound tri(9-hexylcarbazol-3-yl) amine (THCA) and 4,7-diphenyl-1,10-phenanthroline (Bphen) were used for the preparation of bilayer and bulk exciplex based organic light emitting diodes (OLEDs) in which THCA and Bphen were selected as donor and acceptor components, respectively. The dependencies of the electroluminescence and impedance characteristics of the exciplex type OLEDs depending on the technologies of the formation of the active layers were studied. The correlations between the appearance of negative differential resistance in current–voltage characteristics and negative capacitance observed at low frequencies in impedance spectra of OLEDs with the resonant tunneling of electrons into THCA through Bphen/THCA barrier and generation–recombination processes in THCA were established.

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1. Introduction

The considerable attention to white organic light-emitting diodes (WOLED) is recently paid mainly due to the perspectives of their application in solid lighting systems [1]. Such devices must have a high efficiency and a low cost of production [2]. Highly-efficient WOLEDs, are commonly obtained from phosphorescent organometallic complexes, that can be used as guest components in the guest–host systems in a complex multilayer structures [3]. The main advantage of such phosphorescent materials is the involvement of singlet as well as triplet excited states (excitons) in the process of radiative recombination [4]. The considerable drawbacks of such approach are high costs of rare metal materials and rather low time stability, especially for blue light emitting materials [5].

One of the most simple, cheap and technologically conceptual approach for the development of WOLEDs is based on mixing of the molecular exciton emission (usually the blue one) and the exciplex emission of bimolecular excited species (usually the orange one) [6,7]. The implemented cross-interaction of electrons from the

LUMO level of a donor molecular material and holes from the HOMO of an acceptor molecular material occurs with the formation of the exciplex [7]. In such systems HOMO and LUMO energy levels are separated by a relatively long distance, which leads to the very small difference between singlet (S_1) and triplet (T_1) energy levels (ΔE_{ST}), which can cause the reverse intersystem crossing (RISC) from T_1 to S_1 resulting in thermally activated delayed fluorescence (TADF) [8–11]. It should be noted, that the exciplex-type OLEDs are characterized by the high values of the external quantum efficiency [12]. The necessary conditions for the formation of exciplexes are availability of high energy barriers for the charge carriers in the organic–organic interface [7], as well as star-shape molecular conformation providing effective overlap of electron clouds of donor and acceptor molecules [13].

In this work, we identified exciplex emission at the organic interface between starburst carbazole derivative tri(9-hexylcarbazol-3-yl) amine (THCA) [7] and 4,7-diphenyl-1,10-phenanthroline (Bphen) [12] (Fig. 1). We studied the effect of the technologies used for the preparation of the active layers such as step-by-step deposition or codeposition on electro-optical parameters of the exciplex type OLED. The choice of the active materials used for the fabrication of exciplex type OLED was predetermined by the following considerations: THCA and Bphen are capable of exciplex formation with the different organic and organometallic

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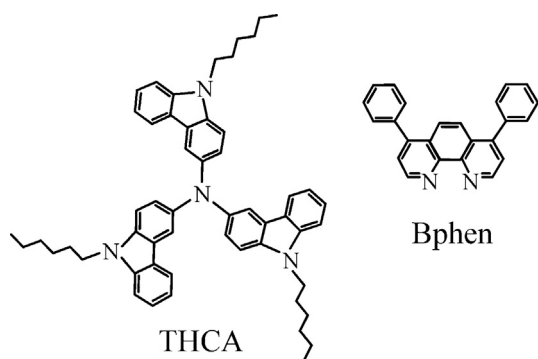


Fig. 1. Chemical formulas of THCA and Bphen.

compounds [7,12–14], with the availability of the thermally activated delayed fluorescence.

2. Experimental details

OLEDs with the structure indium tin-oxide(ITO)/CuI(8 nm)/THCA(30 nm)/Bphen(30 nm)/Ca(50 nm)/Al(200 nm) (device A) and indium tin-oxide(ITO)/CuI(8 nm)/THCA:Bphen(60 nm)/Ca(50 nm)/Al(200 nm) (device B) were fabricated by step-by-step deposition or codeposition of different organic layers and metal electrodes onto pre-cleaned ITO-coated glass substrate under a vacuum of 10^{-5} Torr. Energy-band diagrams of the fabricated devices are shown in Fig. 2. CuI was used for the preparation of hole-transporting layer [15], THCA and Bphen (Fig. 1), were applied as donor and acceptor materials respectively. Since Ca is highly reactive and corrodes quickly in the ambient atmosphere, Ca layer topped with 200 nm aluminum (Al) layer was used as the cathode. The active area of the obtained devices was $3 \times 6 \text{ mm}^2$.

The density current–voltage and luminance–voltage characteristics were measured using a semiconductor parameter analyzer (HP 4145A) in air without passivation immediately after the formation of the device. The brightness measurements were done using a calibrated photodiode [16]. The electroluminescence spectra were recorded with an Ocean Optics USB2000 spectrometer.

The single layers of the THCA and Bphen were prepared by thermo vacuum deposition whereas the layer of the molecular mixture THCA: Bphen was prepared by casting onto clean quartz substrate from the THF solution of the mixture consisting of 50% of THCA and 50% of Bphen.

Photoluminescence spectra and photoluminescence decay curves were recorded with the Edinburgh Instruments FLS980 spectrometer at room temperature using a low repetition rate μF920H Xenon Flash lamp as the excitation source.

Impedance spectroscopy was used for the investigation of bulk and interface parameters of the fabricated devices. The impedance

measurements were done in the frequency range of $10/10^6$ Hz at the constant bias voltages of 0, 1.0, 2.0, 3.0 V, using instrument “AUTO LAB” with FRA-2 and GPES software. The frequency dependencies of the complex resistivity Z were analyzed by graphic-analytical method using ZView 2.3 (Scribner Associates) software. The approximation inaccuracy did not exceed 6%.

3. Results and discussion

The photoluminescence (PL) spectrum of the spin coated film of the molecular mixture of THCA and Bphen is characterized by three emission bands (Fig. 3).

The PL spectrum of the film of the molecular mixture has a peak at the wavelength of 567 nm and two shoulders at 470 nm and 640 nm. The short-wavelength emission band observed at 470 nm is characteristic of the pure THCA singlet emission (Fig. 3, the curve 2) [7,14].

It is obvious, that PL spectrum of the spin coated film of the molecular mixture of THCA and Bphen is not a simple superposition of the fluorescence spectra of the individual components (Fig. 3, the curves 1 and 2). We can consider the existence of the bimolecular excited species, the photoluminescence spectra of which are characterized by the red-shifted broad bands (567 nm and 640 nm) [12]. Since of the interface between the molecules of THCA and Bphen is extensive we suggest that it leads to the expansion of the long-wavelength region and to the appearance of a shoulder in the 640 nm region [13].

To get the better understanding of the mechanism of luminescence at the long-wavelength region the decay curve of PL of the spin coated film of the molecular mixture of THCA and Bphen at 567 nm was recorded (Fig. 4). For the adequate representation of the decay the triple exponentials function was required. Two components with the lifetimes of 15 ns and 60 ns are

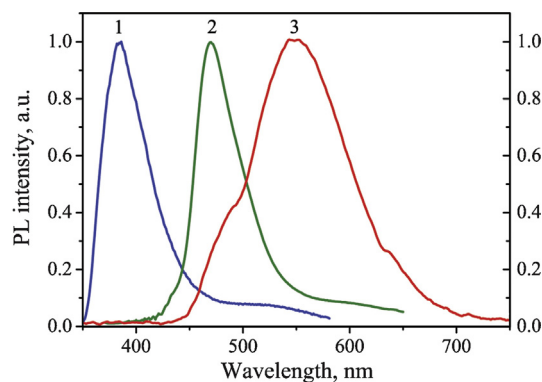


Fig. 3. Normalized PL spectra of the films of Bphen (1), THCA (2), and of the spin coated film of the molecular mixture of THCA (50%) and Bphen (50%) (3).

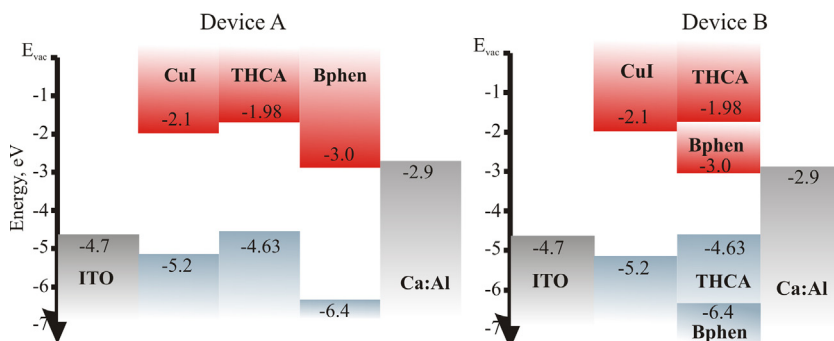


Fig. 2. Energy-band diagrams of the fabricated devices [7,17].

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