

The Study on the Relationship between Dynamic Balance Energy Distribution and Spectral Stability with Voltage Change in White Organic Light Emitting Diode

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Abstract

The stable spectrum can be obtained when the voltage changes, which is a necessary condition for the white organic light emitting diode (WOLED) device to be widely used in the field of solid-state lighting. However, with the increase of voltage, the movement of the recombination zone (RZ) is inevitable because the perfect bipolar host material is difficult to obtain, which will redistribute the energy in the light emitting layer (EML) and affect the stability of the spectrum. We fabricate a series of ternary hybrid WOLEDs with a simple structure by inserting ultra-thin PO-T2T into the blue exciplex (TCTA:TPBi) to form the green interface exciplex. Without considering the movement of RZ, device B2 realizes the dynamic balance energy distribution in EML and stable spectrum by controlling two processes of the Dexter energy transfer and exciton capture. By modifying the doping ratio of the host material, we also find that the broadened RZ is helpful to further improve the spectral stability of the device. When the voltage changes from 3 V to 7 V, the change range of color coordinates is only (0.026, 0.025).

Keywords

WOLED, Spectral Stability, Exciplex, Energy Transfer, Recombination Zone

1. Introduction

White organic light emitting device (WOLED) has great application potential in the field of full-color flat panel display and solid-state lighting due to its advantages of self-luminous, low power consumption, bright color and fast response [1]-[7]. In daily lighting applications, WOLED requires not only high efficiency

and color rendering index (CRI), but also high color and spectral stability [8] [9] [10]. It is generally believed that the change of color and spectrum results from the movement of the recombination zone (RZ) with the change of voltage in WOLED devices, which changes the energy distribution between different color sub-emitting layers (sub-EMLs) at different voltages. However, because the carrier transmission in EML is not balanced, and the mobility of carriers under different voltages is also different, it is difficult to completely avoid the movement of RZ. Commission Internationale de L'Eclairage (CIE) coordinates and correlated color temperature are two very important parameters to characterize the color and spectral stability of the device [11] [12].

In order to achieve more stable spectra, methods such as stabilizing RZ and increasing the compensation layer are used respectively. The bipolar host material in EML is helpful to expand RZ [13] [14] [15]. For example, Chen *et al.* demonstrated high-efficiency and superior color-stability white phosphorescent WOLED based on double blue mixed-host emission layers (EMLs) with different mixed ratios [13]. Zhao *et al.* have proved that the bipolar mixed TCTA:Bepp₂ is a charge carrier switch by regulating the distribution of charge carriers and then the exciton recombination zone, which plays an important role in improving efficiency and stabilizing the spectrum [14]. To alleviate the aggregation of excitons and carriers, some researchers begin to insert interlayer with bipolar charge carrier transport properties or heterojunction in EML [16] [17]. Although a wider and more dispersed RZ is conducive to achieving a more stable spectrum, real bipolar materials are difficult to obtain [13] [14] [15] [16] [17]. In addition, the spectral stability can also be achieved by adding a compensation layer to compensate for the decline of some light intensity caused by the movement of RZ [18] [19]. Tang *et al.* fabricated a series of white phosphorescent OLED devices with multiple emitting layers with multiple dopant (MEML-MD) structure, and the variation of CIE coordinate of device with the voltage increasing from 4 V to 7 V is only (0.006, 0.004) [18]. However, the structure of the device obtained in this way is complex, which increases the difficulty of manufacturing.

In this paper, we present a simple design of multiple EMLs in the hybrid-WOELD without the bipolar host material and complex compensation layer. The main structure of EML of all devices is composed of a blue exciplex (TCTA:TPBi). The material of interlayer layer (PO-T2T) in EML can form a green interface exciplex (PO-T2T/TCTA) with TCTA around it. The red EML between electron transport layer (ETL) and blue EML (TCTA:TPBi, B-EML1). The basic structure of the device is as follows: ITO (100 nm)/HAT-CN (10 nm)/TAPC (35 nm)/TCTA (5 nm)/B-EML2 (8 - x nm)/Interlayer (1 nm)/B-EML1 (x nm)/R-EML (2 nm)/TmPyPB (40 nm)/Liq (2 nm)/Al (100 nm). The main materials in B-EML1, B-EML2 and R-EML layers are TCTA_y:TPBi_{1-y}, TCTA_z:TPBi_{1-z} and TCTA:TPBi:1.5%Ir(piq)₂(acac), respectively. We investigate the influence of the position of the green exciplex sub-EML and the position of RZ on the spectral stability of the device. Under the condition that the RZ moves with the voltage, we realize the dynamic balance energy distribution in EML and obtain the

WOLED device with stable spectrum by regulating two processes of energy transmission and direct energy capture.

2. Experimental

All devices are manufactured on glass substrates with indium tin oxide (ITO), the surface resistance of the substrate surface is $15 \Omega/\text{sq}$. Before deposition, the glass substrate needs to be ultrasonically cleaned. During the cleaning process, it is necessary to place them in deionized water, isopropyl alcohol and alcohol respectively for ultrasonic cleaning. Each process needs to last at least 10 min. After cleaning, the substrate is dried for 30 min and cooled for 30 min. The thermal evaporation method is adopted in this experiment, and the vacuum value in the evaporation environment is lower than 5×10^{-4} Pa. The evaporation rate and deposition thickness of all materials is measured by a calibrated quartz crystal oscillator. The deposition rate of all organic materials is $0.05 \text{ \AA/s} - 2 \text{ \AA/s}$. The evaporation rate of cathode material Al is $3 \text{ \AA/s}$. The luminous area of device is 9 mm^2 . The luminous flux and luminous efficacy include other electro-optical data and spectral data are measured and recorded with a computer-controlled Keithley 2400 power meter and PR655 spectrometer.

The bis(1-phenylisoquinoline) (acetylacetonate)iridium(III) ($\text{Ir}(\text{piq})_2(\text{acac})$) is used as red phosphorescent dye [20]. 1,4,5,8,9,11-Hexaazatriphenylene-hexacarbonitrile (HAT-CN) and 8-hydro-xyquinolinolato-lithium (LiQ) are used as the hole injection layer (HIL) and the electron injection layer (EIL) [21] [22]. 4,4',4''-Tris(N-carbazolyl)tri-phenylamine (TCTA), 1,1'-bis[4-(di-p-tolyl-amino)-phenyl] cyclohexane (TAPC) and 1,3,5-tri[(3-pyridyl)-phen-3-yl] benzene (TmPyPB) are selected as the first hole transport layer (HTL1), the second hole transport layer (HTL2) and the electron transport layer (ETL), respectively [23] [24] [25]. (1,3,5-Triazine-2,4,6-triyl)tris(benzene-3,1-diyl)tris(diphenyl-phosphine oxide) (PO-T2T) are used as interlayer material [26] [27]. The schematic diagram of device structure and partial material structure are shown in **Figure 1**.

3. Results and Discussion

Firstly, we introduce the green interface exciplex (PO-T2T/TCTA), which has a spectral peak of 535 nm, to convert the device from binary WOLED to ternary WOLED, and investigate the influence of its appearance on the electro-optical characteristics and spectral characteristics of the device. In devices A1, A2 and A3, TCTA:TPBi, PO-T2T:TPBi and PO-T2T with a thickness of 1 nm are inserted into the blue exciplex (TCTA:TPBi) EML as interlayer. The electro-optical characteristic curve and data of devices A1-A3 are shown in **Figure 2** and **Table 1**. On the one hand, because the electron mobility of PO-T2T ($4.4 \times 10^{-3} \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$) is greater than that of TPBi ($3.3 \times 10^{-5} \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$) [28] [29], the current density in EML increases with the increase of PO-T2T in interlayer as shown in **Figure 2(a)**. On the other hand, it can be seen clearly in **Figure 2(b)** that increasing the content of PO-T2T also enhances the interface effect, which disperses and balances

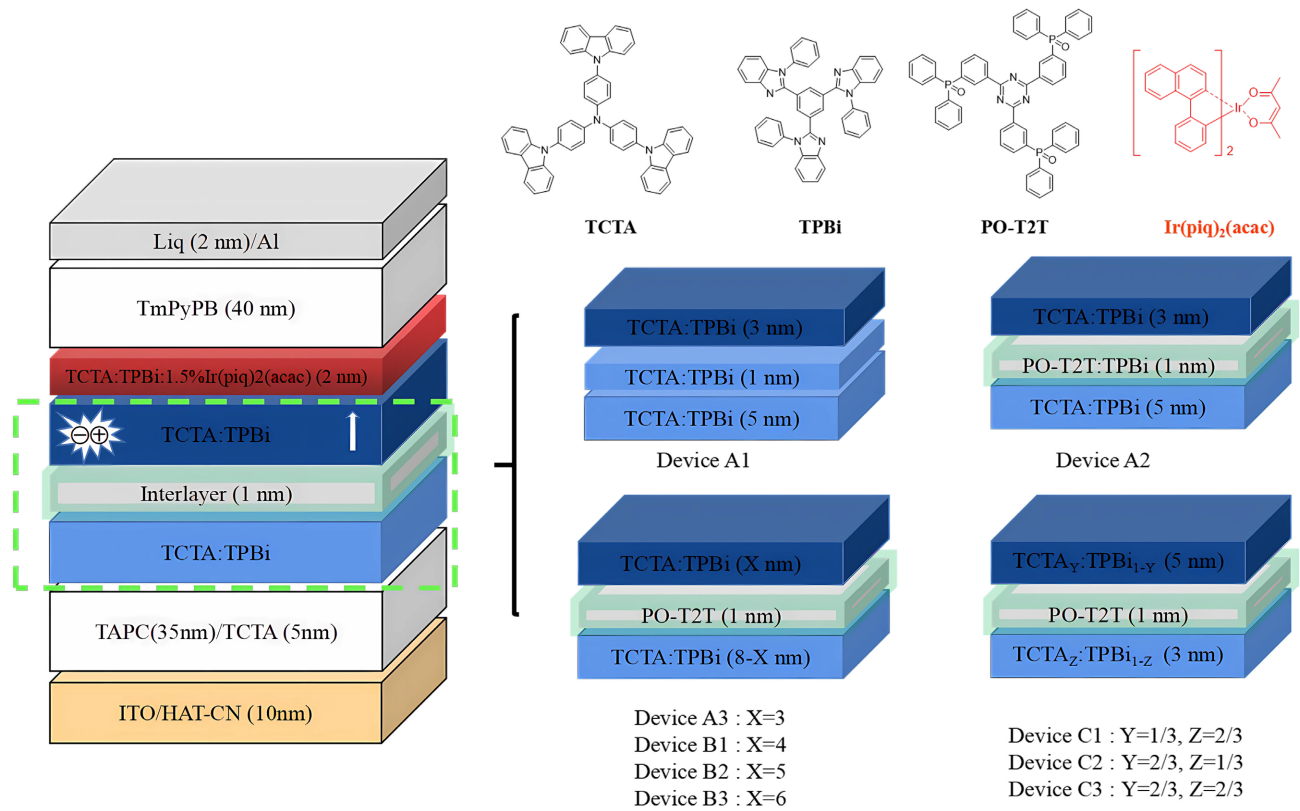


Figure 1. Schematic diagram of the device structure and molecular structure of the emitting layer material.

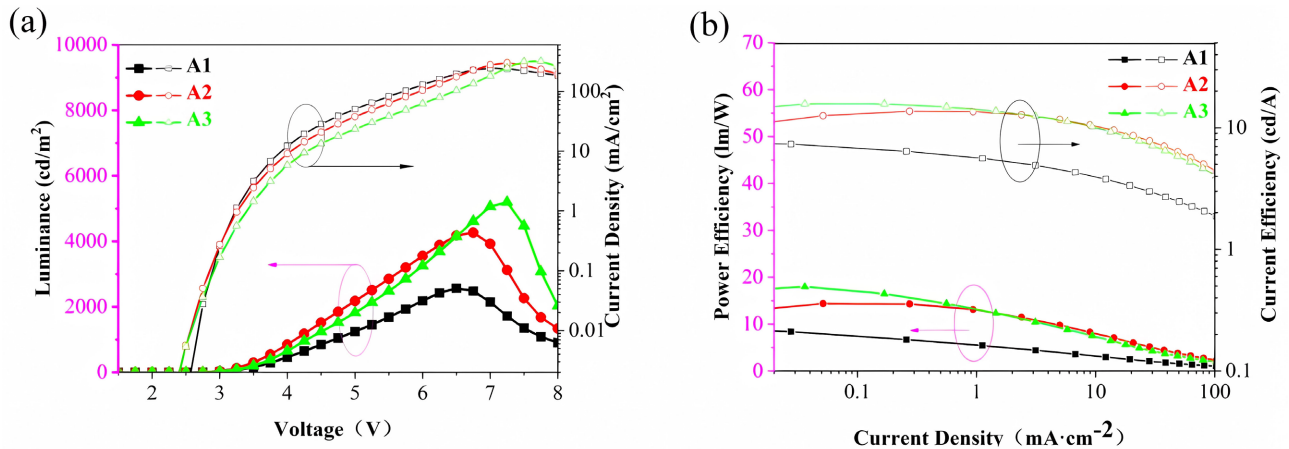


Figure 2. (a) Luminance-voltage-current density curves of devices A1-A3; (b) Power efficiency-current density-current efficiency curves of devices A1-A3.

the carriers in EML, thus improving the efficiency of the device. Therefore, due to the maximum efficiency and current density, device A3 achieves the maximum luminance of 5194 cd/m^2 .

Figure 3 exhibits the normalized electroluminescence (EL) spectra of devices A1-A3 under different voltages. Because there is no green interface exciplex in EML, the spectra of device A1 is only composed of red and blue light, which belongs to binary WOLED. As the voltage increases, the hole mobility of TCTA

Table 1. EL characteristics of devices tested with different structures.

Device	CD _{max}	L _{max}	PE _{max}	CE _{max}	CCT/K		CIE	
	mA/cm ²	cd/m ²	lm/W	cd/A	3 V	7 V	3 V	7 V
A1	242.84	2566	8.38	7.33	0	1848	(0.616, 0.300)	(0.442, 0.282)
A2	300.12	4260	14.29	13.65	3689	2346	(0.438, 0.513)	(0.492, 0.416)
A3	319.80	5194	17.98	15.74	3774	2350	(0.435, 0.525)	(0.480, 0.401)
B1	420.65	5595	17.23	15.09	3423	2461	(0.454, 0.506)	(0.472, 0.400)
B2	395.27	5500	14.65	12.83	2682	2590	(0.492, 0.466)	(0.464, 0.403)
B3	390.94	5527	10.95	9.58	1767	2488	(0.548, 0.401)	(0.468, 0.398)
C1	220.92	4056	10.79	9.45	0	1698	(0.616, 0.368)	(0.546, 0.386)
C2	256.59	4931	12.73	11.14	2199	2322	(0.523, 0.441)	(0.496, 0.416)
C3	215.92	4209	9.74	8.52	0	1628	(0.659, 0.330)	(0.548, 0.376)

CD_{max} is the maximum current density. L_{max} is the maximum luminance. PE_{max} is the maximum power efficiency. CE_{max} is the maximum current efficiency.

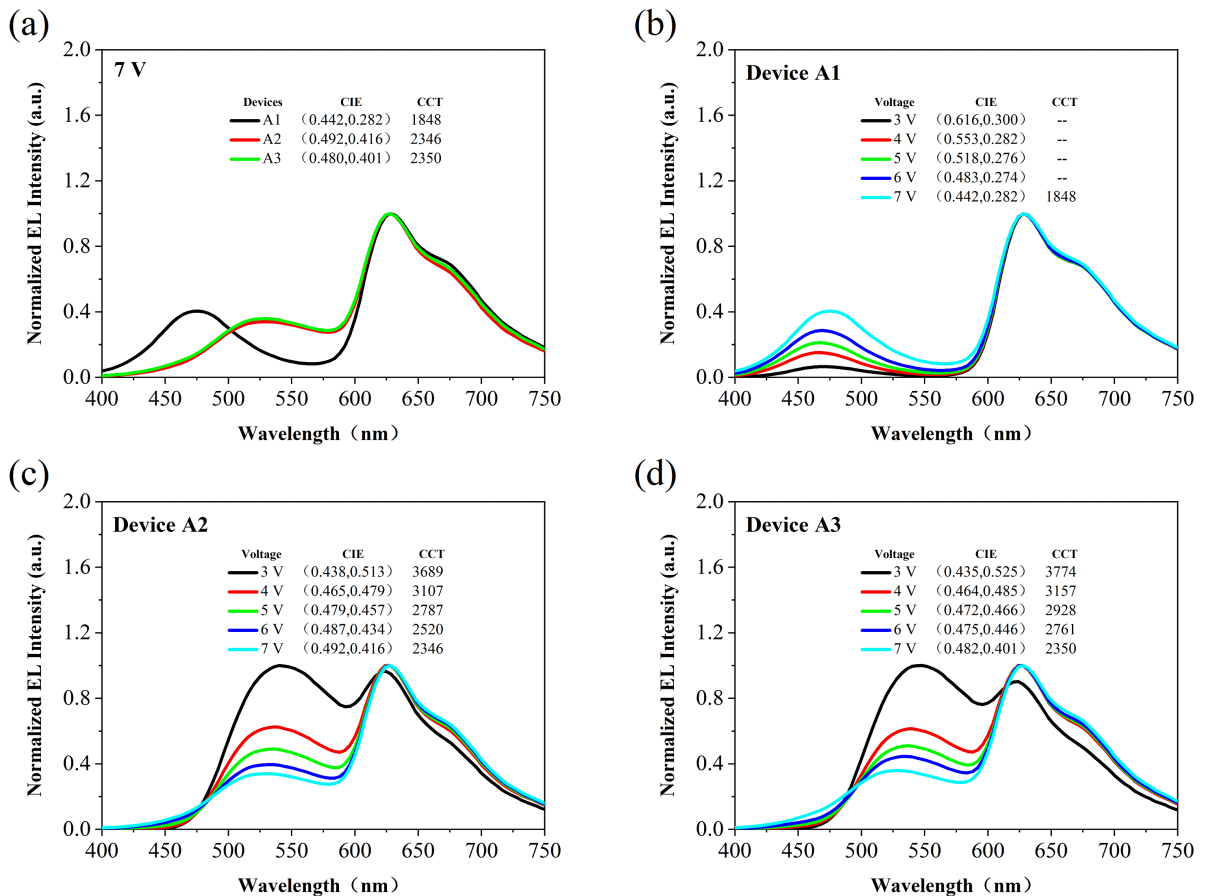


Figure 3. (a) The normalized EL spectra of devices A1-A3 at 7 V; (b)-(d) The normalized EL spectra of devices A1-A3 at different driving voltages.

will increase faster than that of TPBi in EML, which causes RZ to shift to the side of ETL [30]. At low voltage, Dexter energy in EML of device A1 will transfer

from the blue EML to the red EML. With the increase of voltage, the excess energy will be directly captured by the blue EML after the red spectral intensity reaches saturation, thus improving the blue spectral intensity at high voltage. The spectral and color coordinates of device A1 are unstable. As shown in **Figure 3(c)** and **Figure 3(d)**, the spectral stability and CCT of devices A2 and A3 are similar at the same voltage. Although the content of PO-T2T in EML of device A3 is twice that of A2, the distance from the position of green interface exciplex to RZ is almost the same. **Figure 4(a)** describes the schematic diagram of energy transmission path in EML of devices A2 and A3. At low voltage, the blue exciplex near RZ obtains energy and transfers energy to the red and green EML on both sides by means of Dexter energy transfer, so the spectral intensity of green and red is much higher than that of blue. With the increase in voltage, the RZ moves away from the green EML and towards the red EML, so the energy transmission from the blue EML to the green EML is weakened, resulting in the decrease in the green spectral intensity. Because the singlet energy level of the blue EML is higher than that of the green EML, the existence of B-EML1 limits the energy transfer from the green EML to the red EML.

Secondly, we plan to increase the distance between the green EML and RZ and reduce the energy transmission between them, so as to reduce the green spectral intensity at low voltage and obtain a more stable spectrum. By changing the position of the green EML in the overall EML, a series of WOLEDs with a similar position of RZ are prepared on the basis of device A3. The thickness of layer B-EML1 (x nm) is 3, 4, 5 and 6 nm, corresponding to devices A3, B1, B2 and B3 respectively. Because the thickness of the PO-T2T layer is only 1 nm, its position change has little effect on the position of RZ and the electro-optical characteristics of these four devices. The electro-optical characteristic curve and data of devices B1-B3 are shown in **Figure 5** and **Table 1**.

Figure 6(a) shows the normalized EL spectra of devices A3 and B1-B3 at the voltage of 7 V. **Figures 6(b)-(d)** shows the normalized EL spectra of devices B1-B3

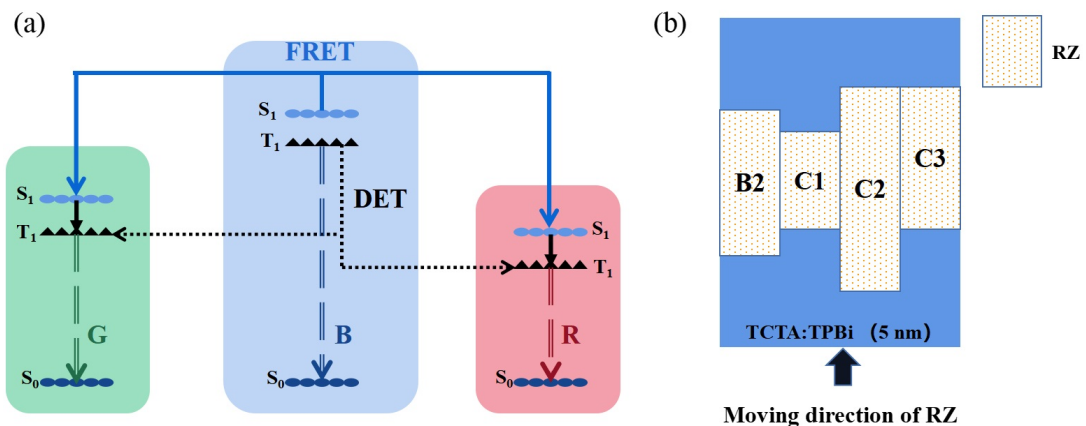


Figure 4. (a) The schematic mechanism diagrams of exciton energy transfer path for all devices in this study; (b) The main RZ of different devices. S₁, T₁, S₀, ISC and RISC represent the singlet state level, triplet state level, ground state level, intersystem crossing and reverse intersystem crossing, respectively.

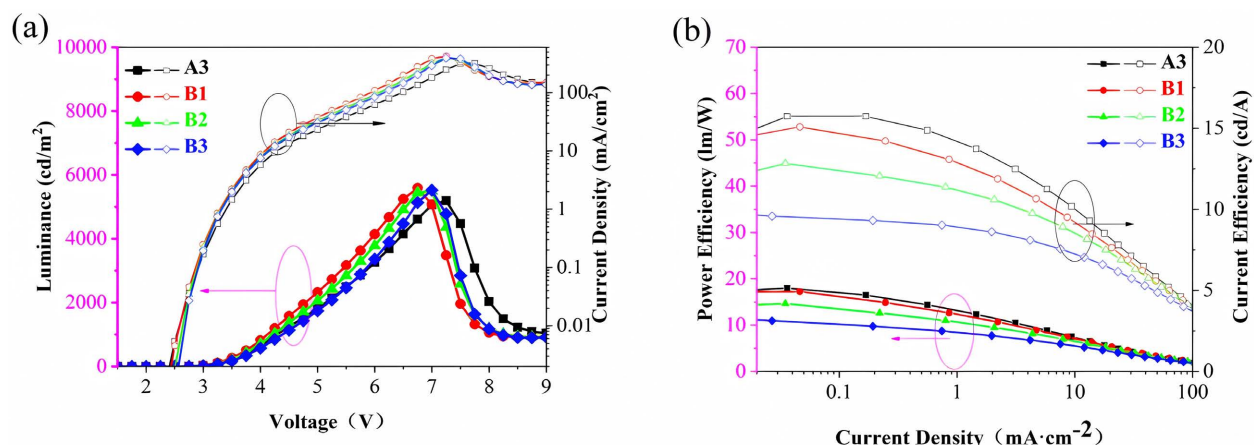


Figure 5. (a) Luminance-voltage-current density curves of devices A3 and B1-B3; (b) Power efficiency-current density-current efficiency curves of devices A3 and B1-B3.

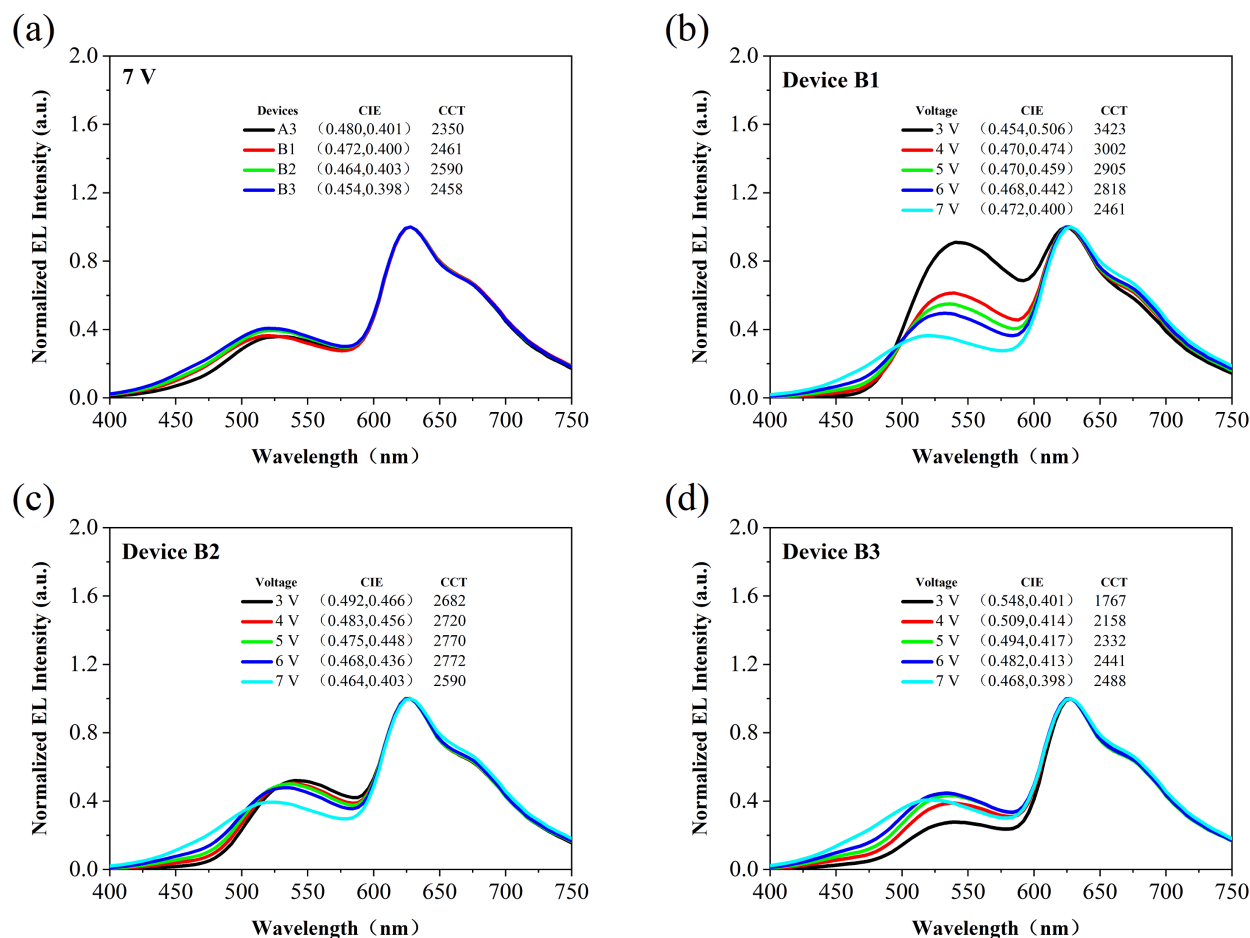


Figure 6. (a) The normalized EL spectra of devices A3 and B1-B3 at 7 V; (b)-(d) The normalized EL spectra of devices B1-B3 at different driving voltages.

at different voltage from 3 V to 7 V. At high voltage of 7 V, the green emission intensities of the four devices are similar, which corresponds to the maximum value of energy that can be directly captured by the green interface exciplex. As

the thickness of B-EML1 (x nm) increases from 3 nm to 5 nm, the green light emitting layer also gradually moves away from RZ. With the increase of voltage, RZ will move towards ETL, and the distance between the green light emitting layer and RZ will further increase, which will inhibit the transfer of Dexter energy from the blue light emitting layer to the green light emitting layer. At a low voltage of 3 V, device A3 and device B3 get the highest and lowest green spectral intensity, respectively. The change trend of green spectral intensity of devices A3 and B3 with voltage is opposite, mainly due to the decrease of Dexter energy transmission and the increase of exciton directly captured by green light emitting layer. In device B2, The spectrum is relatively stable because the reduced Dexter energy and the increased direct capture energy in the green light emitting layer are in a dynamic balance during the change from 3 V to 7 V as shown in **Figure 6(c)**. Therefore, without focusing on the movement of RZ and the addition of the compensation layer, we can achieve a dynamic balance energy distribution and stable spectrum by simply adjusting the position of sub-EMLs.

Finally, we fine-tune the position and range of RZ based on device B2 in order to further stabilize the spectrum while the position of the green EML remains unchanged. We prepare devices C1-C3 on the basis of device B2. The device structure diagram and the RZ location are shown in **Figure 1** and **Figure 4(b)**. The electro-optic characteristics of device C1-C3 are slightly worse than that of device B2 due to the change of the composite region as depicted in **Figure 7**. As shown in **Figure 8**, the spectrum of device C2 becomes more stable as the voltage increases from 3 V to 7 V, which is due to the expansion effect of RZ in device C2, the change of its color in CIE-1931 color coordinate diagram is shown in **Figure 9**. At low voltage, the green spectral intensities of device C1 and C3 is further decreased due to the narrowing of RZ and the shift of RZ to ETL away from the green light-emitting layer, respectively. The changing trend of the green spectral intensity with voltage is similar to that of device B3, which is also consistent with the analysis of device B3.

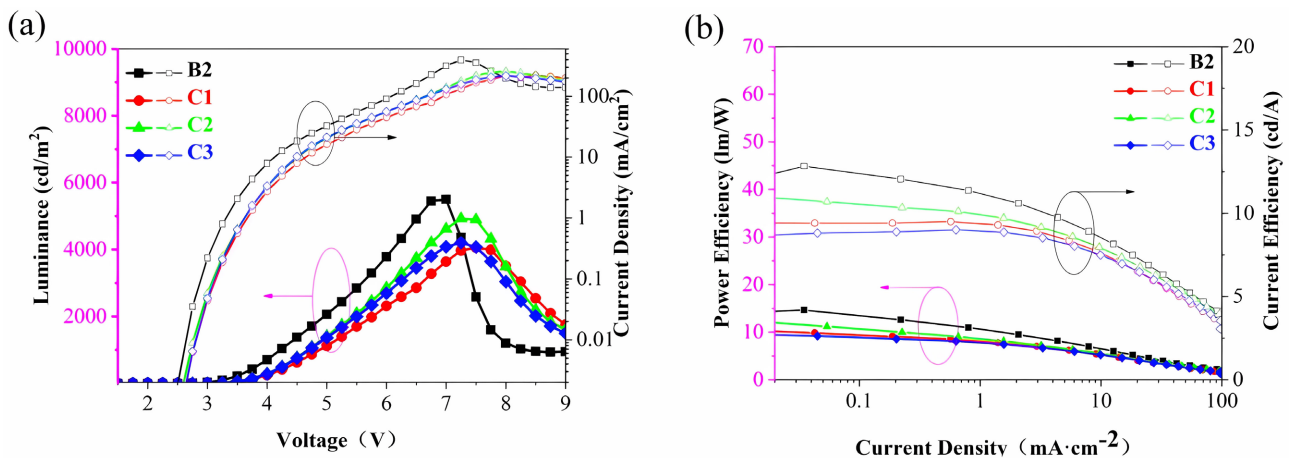


Figure 7. (a) Luminance-voltage-current density curves of devices B2 and C1-C3; (b) Power efficiency-current density-current efficiency curves of devices B2 and C1-C3.

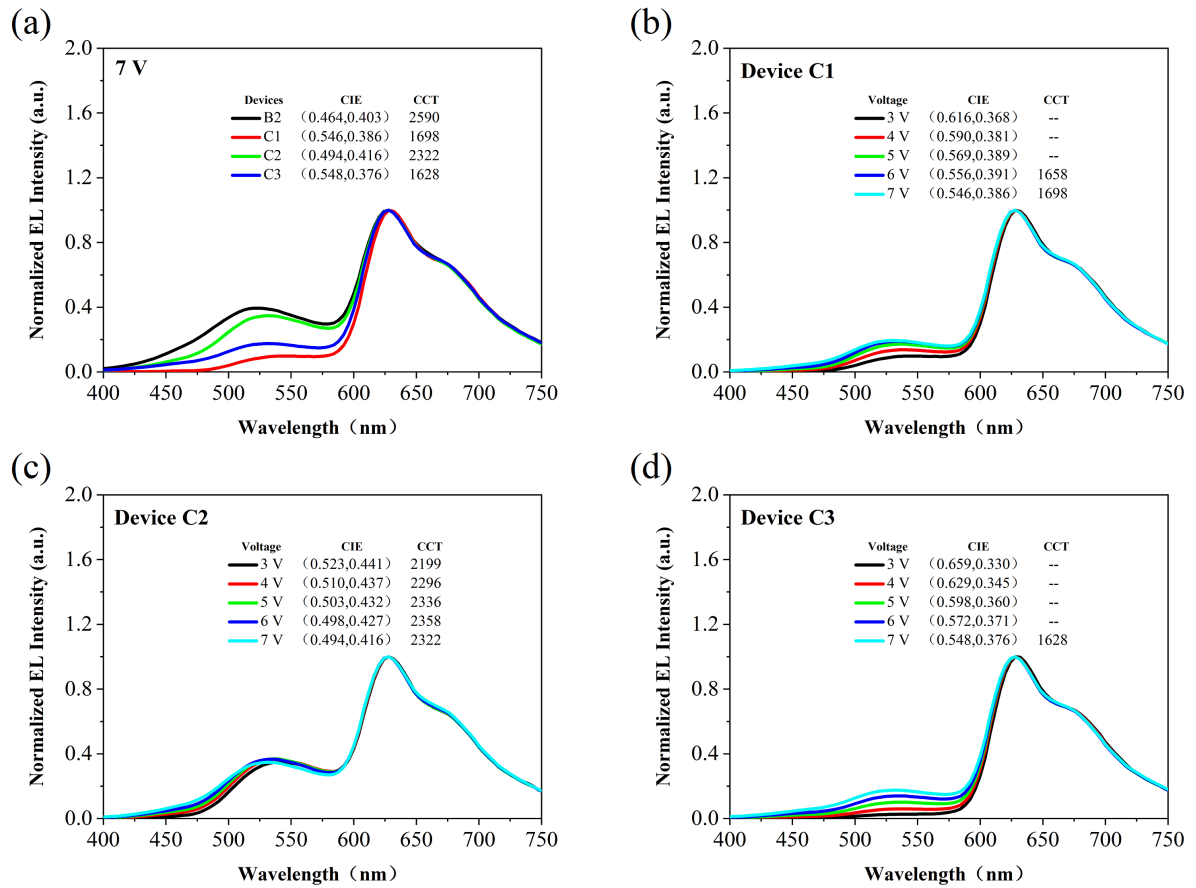
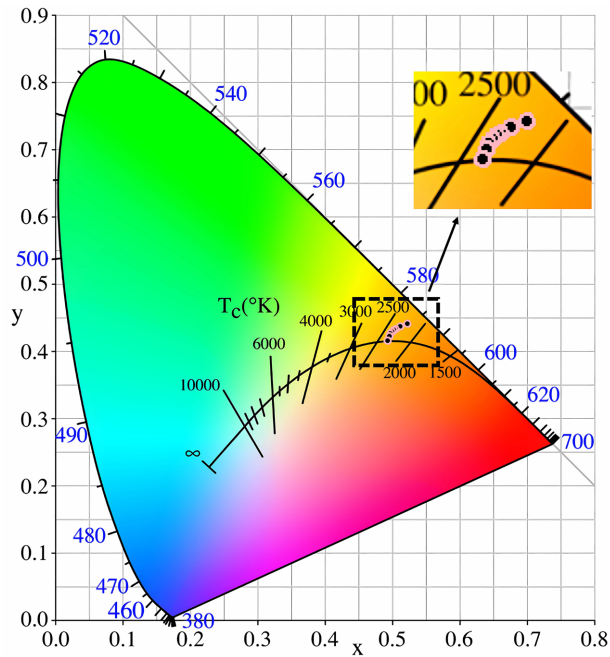


Figure 8. (a) The normalized EL spectra of devices B2 and C1-C3 at 7 V; (b)-(d) The normalized EL spectra of devices C1-C3 at different driving voltages.



4. Conclusion

We have achieved the dynamic balance energy distribution in the ternary WOLED device with a simple structure, thus achieving a stable spectrum. On the one hand, the Dexter energy transfer will weaken with the increase in distance between the EML and RZ. On the other hand, the number of excitons in EML will increase with the increase in voltage, which will also increase the number of excitons directly captured by the luminescent material itself and improve the luminous intensity. The above two reasons will lead to the redistribution of energy in EML with the change of voltage, resulting in spectral instability. By moving the green interface exciplex sub-EML and increasing the distance between it and RZ, we achieve the dynamic balance between Dexter energy transmission and exciton direct capture in the green EML of device B2 as the voltage changes from 3 V to 7 V, which also leads to the relatively stable spectrum of the whole device. When the position of the green EML remains unchanged, the position and range of the RZ can be affected by changing the doping ratio of the main material of EML. The experimental results show that the farther distance from the green EML and the wider RZ contribute to the more stable spectrum. When the voltage changes from 3 V to 7 V, the CCT of device C2 changes from 2199 K to 2322 K, and the color coordinate changes in the range of (0.026, 0.025).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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