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Highly efficient and spectra stable warm white organic light-emitting diodes by the application of exciplex as the excitons adjustment layer

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ABSTRACT

We aim to simplify the device structure due to the low concentration precision and repeatability of white organic light-emitting diodes (WOLEDs) based on exciplex host with phosphorescent dopant. Two layers of exciplex used here play the role of blue emitter and excitons adjustment layer, respectively, to manage the excitons. Green and red phosphorescent ultrathin layer are inserted into exciplex layer to form white light-emitting. The maximum efficiency values of current efficiency, power efficiency and EQE reach to 38.8 cd/A, 34.9 lm/W and 15.3%, and remain 29.7 cd/A, 20.4 lm/W and 11.6% at 1000 cd/m^2, respectively. Besides, the change of Commission Internationale de l'Eclairage coordinates is only (0.024, 0.013) with the enhanced driving voltages from 4 V to 6 V. Such a high performance of warm WOLEDs with simple structure demonstrates the reasonable excitons management and efficient energy transfer from exciplex and excitons adjustment layer to ultrathin phosphorescent emitters.

1. Introduction

The applications of exciplex in the organic light-emitting diodes (OLEDs) have got more and more attention these years as a result of its huge advantages of excitons utilization [1–3]. Differing from the thermally activated delayed fluorescent (TADF) materials, which combine donor segment and acceptor segment into monomer molecule, while, exciplex can separate donor and acceptor segment into two molecules. And the corresponding highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy level are also decided by donor and acceptor materials, respectively. Also, the small energy level split (ΔE_{ST}) between singlet and triplet energy level can be achieved by reasonable selection of donor and acceptor materials, which is more simple than TADF materials [4,5]. And smaller the ΔE_{ST} is, the more efficient reverse intersystem crossing (RISC) will be, which the triplet excitons can up-convert from triplet energy level to singlet energy level [6,7]. Finally, the 100% internal quantum efficiency (IQE) could be realized based on TADF materials or exciplex in theory. In the past few years, many exciplexes with high efficiency and photoluminescence quantum yield had been reported, which also indicated the huge potential application of exciplex [8–10].

The application of exciplex host on white OLEDs (WOLEDs) also received more attention due to the highly efficient energy transfer from exciplex host to fluorescent or phosphorescent dopant. However, the WOLEDs with exciplex host adopted the host-guest doped system generally, which the emitting layer (EML) consist of three kinds of materials at least and lead to the fabrication complexity, low concentration precision and repeatability [11–14]. To simplify the fabrication process of WOLEDs based on exciplex host, ultrathin EML is an impactful method without affecting the devices performance. There are also some reports of WOLEDs based on ultrathin EML, but more ultrathin layers are added on the interface of EML and charge transport layer [15–18]. So efficiency roll-off and poor color stability are problems because triplet excitons will gather at the interface and result in serious singlet – triplet annihilation (STA) and triplet – triplet annihilation (TTA) under high current density [19,20]. Moreover, these problems are more

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clearer than the WOLEDs based fluorescent emitters when the guest emitters belong to phosphorescent materials [21]. So the reasonable management of excitons is important to reduce the negative influence of ultrathin EML [22]. Besides, exciplex layer is conducted as emitter or host commonly, but employing exciplex layer as the excitons adjustment layer (EAL) is absent up to now. Therefore, combine the exciplex with ultrathin EML is an effective method to simplify device fabrication process and enhance excitons utilization, further the application of exciplex layer as EAL should also be explored.

In this manuscript, the exciplex of *m*-bis(*N*-carbazolyl)-benzene (mCP): 1,3,5-triazine-2,4,6-triyl) tris(benzene-3,1-diyl) tris(diphenylphosphine oxide) (PO -T2T) with ultrathin phosphorescent EML of tris (2-phenylpyridinato)iridium(III) ($Ir(ppy)_3$) and iridium(III) bis(2-phenylquinoline) acetylacetonate ($Ir(pq)_2acac$) are combined for white emission. mCP: PO-T2T is an highly efficient blue exciplex, which plays the role of blue emitter, meantime the EAL is also acted by the same exciplex of mCP: PO-T2T to manage the excitons. Red and green phosphorescent ultrathin layers of $Ir(pq)_2acac$ and $Ir(ppy)_3$ are inserted into the exciplex layer with different position. This simple device structure achieves high efficiency warm white light-emitting due to the reasonable management of singlet and triplet excitons. The best warm WOLEDs achieve the maximum efficiency values of 38.8 cd/A, 34.9 lm/W and 15.3% for current efficiency, power efficiency and EQE, respectively. Meantime, the change of Commission Internationale de l'Eclairage (CIE) coordinates is only (0.024, 0.013) with the enhanced operation voltages from 4 V to 6 V.

2. Experimental section

All the OLEDs were fabricated on Indium tin oxide (ITO) coated glass substrates at a sheet resistance of 10 Ω per square. The ITO surface was cleaned with acetone, deionized water, acetone sequential, dried in an oven at 50 °Cfor 4 h and then treated by ultraviolet-ozone for 15 min before loading into a deposition chamber ($\sim 4 \times 10^{-4}$ Pa) for subsequent deposition. All layers were grown in succession by thermal evaporation without breaking the vacuum. The organic layers were deposited at a rate of 1.0 Å/s. The inorganic layers of MoO_3 and LiF at the deposition rate of 0.1 Å/s. Al cathode was deposited in the end at a rate of 5.0 Å/s with a shadow mask, which defined the device area of $3 \times 3\,mm^2$. The photoluminescence (PL) spectra of the films were tested utilizing FluoroMax-4 fluorescence spectrometer (HORIBA Jobin Yvon). The UV–Vis absorption spectrum was recorded with Hitachi U-3900 scanning spectrophotometer. Electroluminescence (EL) spectra were measured through PR-655 spectra scan spectrometer with computer controlled. The current-voltage-luminance characteristics were measured by using Keithley 2400 power supply combined with BM-7A luminance colorimeter. EQE was calculated from EL spectra data and the current density-voltage-luminance curve. All the organic materials were procured commercially without further purification. All the measurements were carried out at room temperature under ambient conditions without any protective coatings.

3. Results and discussion

Fig. 1 shows the molecular structure of mCP, PO-T2T, $Ir(ppy)_3$ and $Ir(pq)_2acac$, the PL spectra of mCP, PO-T2T, mCP:PO-T2T mixed films (1:1) and the absorption spectra of phosphorescent emitters. mCP and PO-T2T are chosen as the electron donor and acceptor materials to form the exciplex and phosphorescent emitters of $Ir(pq)_2acac$ and $Ir(ppy)_3$ are selected as the red and green materials for WOLEDs. The observed emission peak of 468 nm is from mCP:PO-T2T mixed film, that is different from the emission of donor of mCP (365 nm) and acceptor of PO-T2T (381 nm), indicates the exciplex formation. Besides, the peak value of 2.64 eV is similar to the energy gap between the HOMO of mCP (6.1 eV) and LUMO of PO-T2T (3.5eV), which also demonstrates the formation of exciplex between mCP and PO-T2T. The exciplex of mCP:

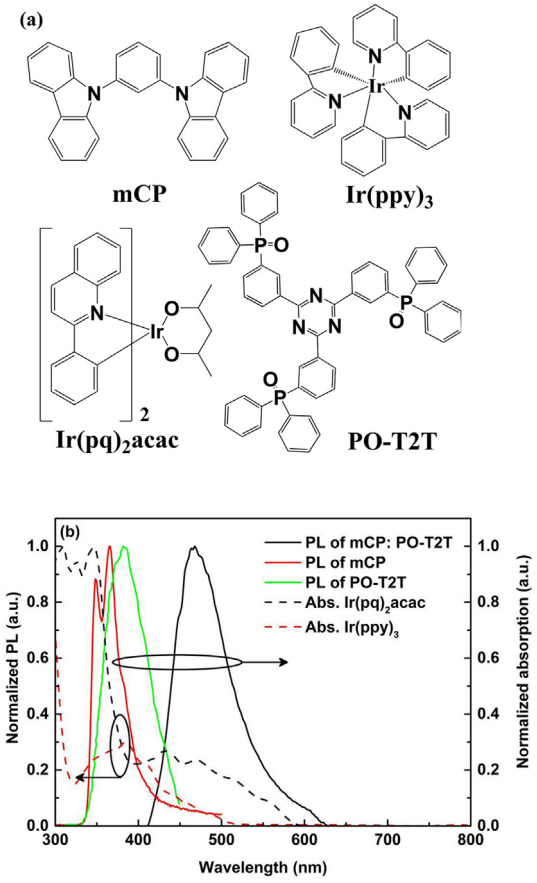

Fig. 1. (a) The molecular structure of mCP, PO-T2T, $Ir(pq)_2acac$ and $Ir(ppy)_3$. (b) The optical characteristics of mCP, PO-T2T, mCP: PO-T2T mixed film and the absorption spectra of $Ir(pq)_2acac$ and $Ir(ppy)_3$.

PO-T2T also had been reported for high efficiency blue emission and its host application for energy transfer was also studied [23]. The absorption spectra of green and red phosphorescent emitter exist large overlap with the PL spectrum of mCP: PO-T2T exciplex, which indicate the a high energy transfer efficiency from exciplex to $Ir(ppy)_3$ and $Ir(pq)_2acac$ (Fig. 1b) [24].

First, we designed the simple device structures as follows: ITO/MoO_3 (3 nm)/mCP (20 nm)/mCP: PO-T2T (1:1) (8 nm)/$Ir(ppy)_3$ (0.01 nm)/mCP: PO-T2T (1:1) (x nm)/$Ir(pq)_2acac$ (0.05 nm)/DPEPO (3 nm)/$Bepp_2$ (40 nm)/LiF (1 nm)/Al. The x = 0, 1, 2 and 3, which are denoted as Device A, B, C and D, respectively. The energy level diagram of the device structure is showed in Fig. 2. In this simple device structures, MoO_3 as the hole injection layer (HIL); mCP as the hole transport layer (HTL); mCP: PO-T2T (8 nm) as the exciplex formation layer with blue emitter; $Ir(ppy)_3$ and $Ir(pq)_2acac$ as the green and red

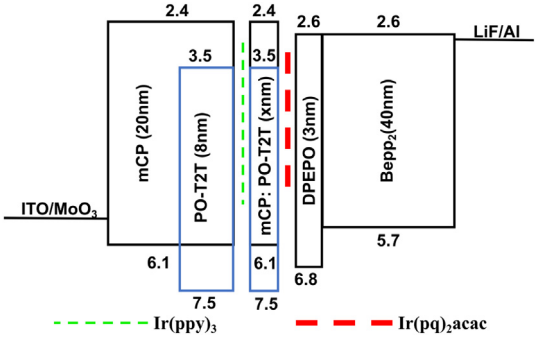

Fig. 2. The device structure of Device A-D, x is 0, 1, 2 and 3.

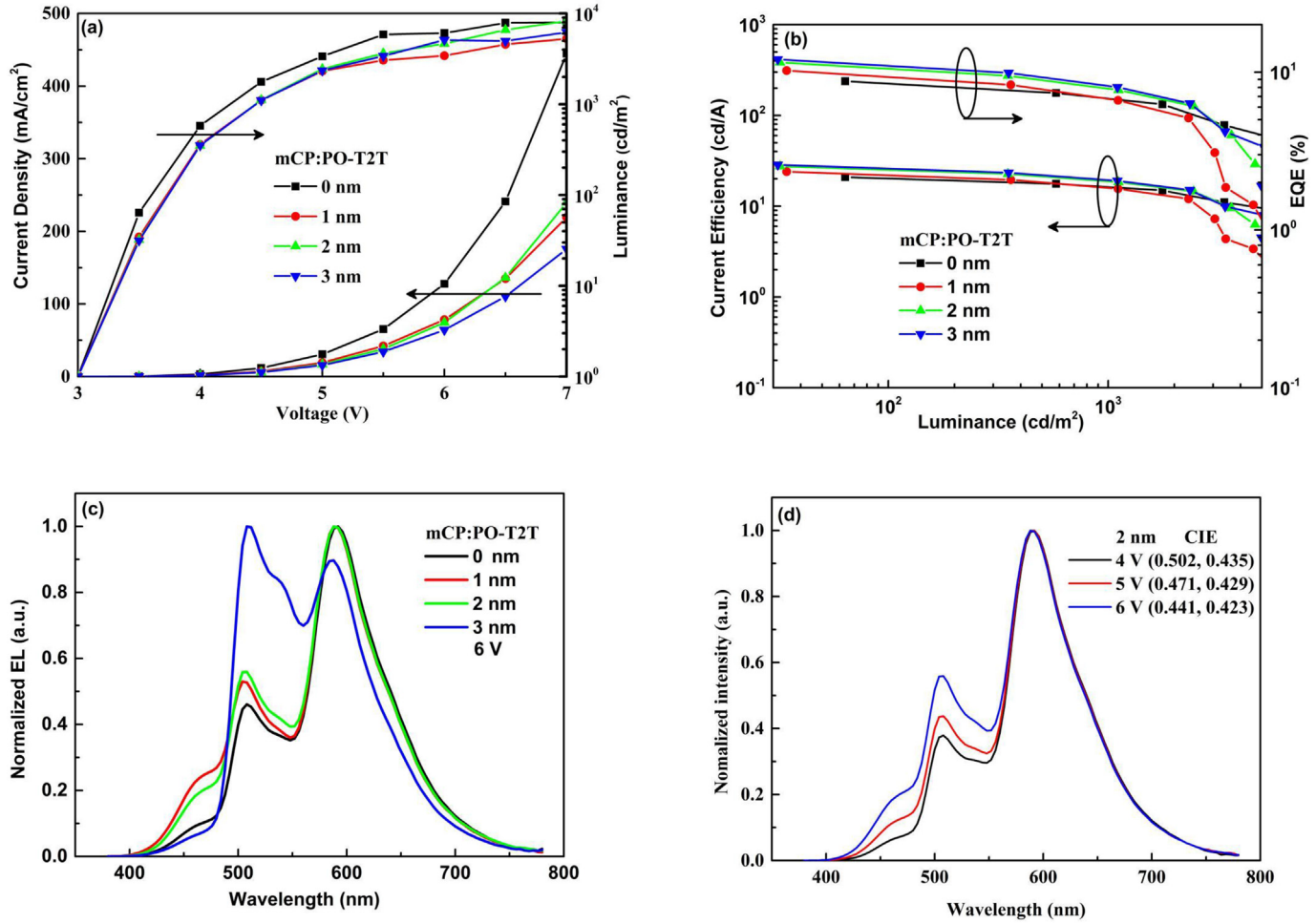

Fig. 3. The EL performances of Device A-D. (a) Current density-voltage-luminance curves. (b) Current efficiency-luminance-EQE curves. (c) EL spectra at 6 V of Device A-D. (d) The EL spectra of Device C from 4 V to 6 V.

phosphorescent ultrathin layer with the thickness as low as 0.01 nm and 0.05 nm, respectively; DPEPO as the exciton block layer due to its high triplet energy level of 2.98 eV, which could confine the excitons in EML [25–27]. The HOMO of the DPEPO can also confine the holes in EML to form exciton efficiently; $Bepp_2$ as the electron transport layer (ETL); The same LUMO energy level of DPEPO and $Bepp_2$ (2.6 eV) and the small energy barrier of 0.2 eV between the DPEPO and exciplex layer ensure the efficient transport and injection of electrons. Here, the mCP: PO-T2T (x nm) is the EAL to control the excitons distribution and transfer.

The EL performances of current density, luminance, current efficiency, EQE, EL spectra of Device A-D are showed in Fig. 3 and all the devices earn the warm white light-emitting. As we can see from Fig. 3a of current density-voltage-luminance curves, all the WOLEDs achieve a low turn-on voltage at 3 V due to the barrier-free charge transport. And the current density decreases as the insertion of EAL, which may come from the charge trapping by phosphorescent emitter. The shallow HOMO and deep LUMO energy level of green and red emitter could trap the holes and electrons, which lead to the decrease of current density. Meantime, the maximum EL efficiencies of the devices with EAL have an obvious improvement compared to Device A of reference device. As shown in Fig. 3b, the maximum EQEs of Device B-D are 10.2%–12.1%, which is higher than reference Device A of 8.8%. However, Device A shows lower efficiency roll-off than the other devices, which may stem from the efficient energy transfer from green emitter to red emitter in Device A, and the decreased green triplet excitons concentration contribute to reduce the efficiency roll-off. Besides, the EL spectra of Device A-D also have a distinction under the same operation voltage of 6 V, which shown in Fig. 3c. The differences of the efficiency and EL spectra could be attributed to the management function to excitons because of the existence of EAL.

From the device structure, we know that the EML is composed of exciplex, which is the mixture of donor and acceptor molecular. So the exciplex EML conduct well charge bipolar transport ability and a broad excitons recombination zone [28]. The emissions of ultrathin phosphorescent emitters are mainly come from the energy transfer of 8 nm blue exciplex layer. In detail, the green emission is achieved by the energy transfer from high energy blue exciplex, and the red emission is from the energy transfer of green emitter and blue exciplex due to the lower triplet energy level. However, the green emission intensity of Device D has a large enhancement compared to other devices at the voltage of 6 V, which shown in Fig. 3c. We consider the obvious higher green emission intensity in Device D derive from the suppressive triplet energy transfer from green to red phosphorescent emitter because of the thick exciplex EAL of 3 nm. But the thickness of 3 nm is still too thin to play the role of main excitons recombination zone and blue emitter, so EAL in this structure are more act as the function of separating the green and red excitons to prevent the energy transfer between them. Further, this is also the reason of low spectra stability with increased voltages [29,30], which are displayed in Fig. 3d. So in one word, the efficiency and spectra stability are low and need to be improved with farther device structure optimization.

Next, we make a simple change based on the structure mentioned above, which shown in Fig. 4, to enhance the utilization of exciplex

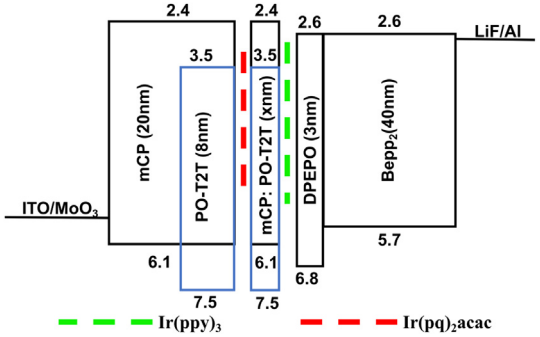

Fig. 4. The device structure of Device E-G, x is 3, 6 and 9.

layer and further manage the excitons, which is changing the position of green and red phosphorescent emitter and adding the thickness of EAL. So the WOLEDs are fabricated as follows: ITO/MoO_3 (3 nm)/mCP (20 nm)/mCP: PO-T2T (1:1) (8 nm)/Ir(pq)$_2$acac (0.05 nm)/mCP: PO-T2T (1:1) (x nm)/Ir(ppy)$_3$ (0.05 nm)/DPEPO (3 nm)/Bepp$_2$ (40 nm)/LiF (1 nm)/Al. The x = 3, 6 and 9, which are denoted as Device E, F and G, respectively. The EL performances are showed in Fig. 5. The current density decreases further with the increased EAL thickness, but the turn-on voltage still keeps at 3 V, which demonstrates the advantage of exciplex EML. The EL efficiency also have great improvement with maximum current efficiencies and EQEs of 26.7 cd/A and 11.2%, 38.8 cd/A and 15.3%, 45.7 cd/A and 15.7%, respectively, from Device E to G. We consider the improved efficiency with increased EAL thickness could be ascribed to two points: one is the increased thickness expands the exciton recombination zone, which will enhance the utilization of excitons. The other is that the EAL plays the role of excitons management, and the thick EAL is benefit to the manage excitons and improve the energy transfer efficiency. The EL spectra of Device E-G under the same voltage are showed in Fig. 5c. We can see that with the thickness increases of EAL from 3 to 9 nm, the green emission intensity has an obvious enhancement, which suggests the new function of EAL that is the excitons recombination zone and energy transfer host, especially for Device G with 9 nm thickness EAL. That means the injected electrons and holes will also recombine at the EAL due to the thick mCP: PO-T2T layer, so the green emission are mainly from the energy transfer of EAL but not from the front 8 nm exciplex emitter. In addition to this, the white light-emitting spectra stability under different voltages also gets better than Device A-D. For example, the spectra of Device F, showed in Fig. 5d, exhibit high color stability with ΔCIE coordinate of (0.024, 0.013) from 4 to 6 V. The improvement of spectra stability could be ascribed to the direct energy transfer from exciplex layer and EAL to green and red phosphorescent emitter because the energy transfer between green and red emitter may lead to the instable white light-emitting spectra, which have been confirmed through the structure above. And the thick EAL in Device F plays the roles of the excitons recombination zone and energy transfer host, which realize a stable exciton recombination and efficient energy transfer, further stabilize the EL spectra. Overall, Device F with 6 nm EAL conducts the best EL performance with maximum current efficiency, power efficiency and EQE of 38.8 cd/A, 34.9 lm/W and 15.3%, respectively, and high color stability white light-emitting spectra are also achieved, simultaneously. The power efficiency curves are showed in Fig. 6, and the EL performances of all the WOLEDs in this paper are listed in Table 1.

Finally, we describe the function of EAL and energy transfer process in these two kinds of device structures in Fig. 7. As we can see, the EAL consist of mCP: PO-T2T, which is the same as blue exciplex layer. The exciton recombination could distribute on both exciplex layer and EAL. The main difference between these two cases is the function of EAL. In Device B-D, the main function of thin EAL is to weaken the energy transfer between green and red emitter. So the green emission comes

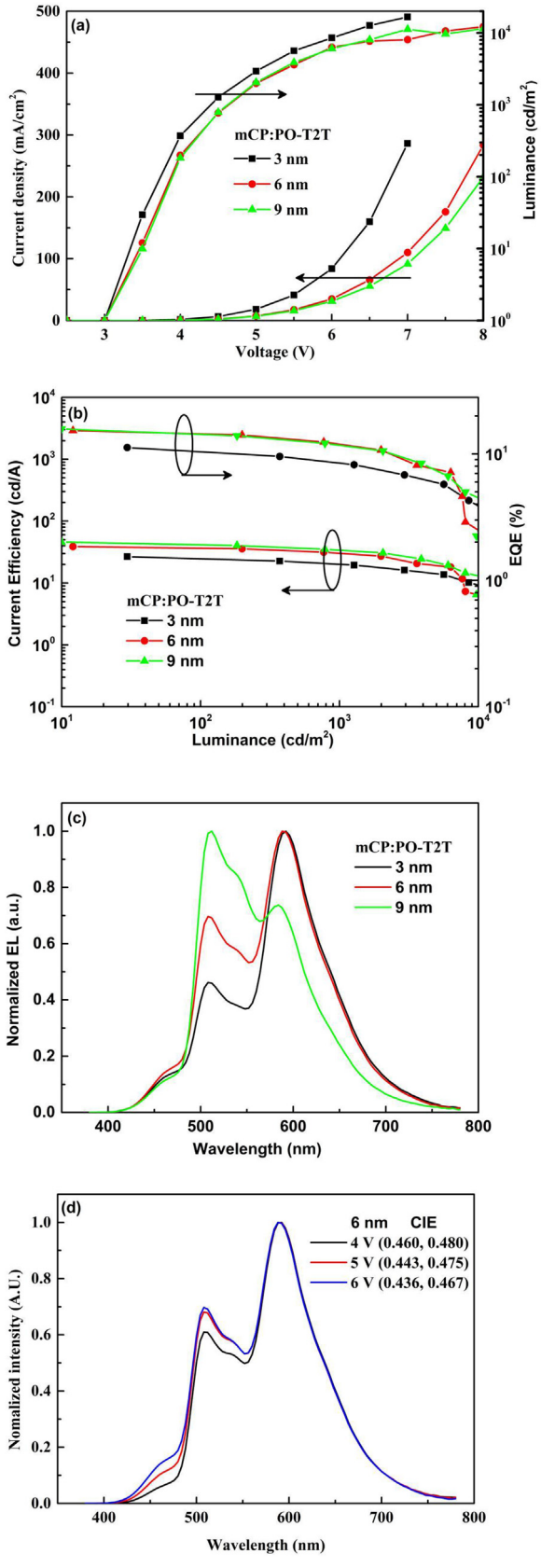

Fig. 5. The EL performances of Device E-G. (a) Current density-voltage-luminance curves. (b) Current efficiency-luminance-EQE curves. (c) EL spectra at 6 V of Device E-G. (d) The EL spectra of Device F from 4 V to 6 V.

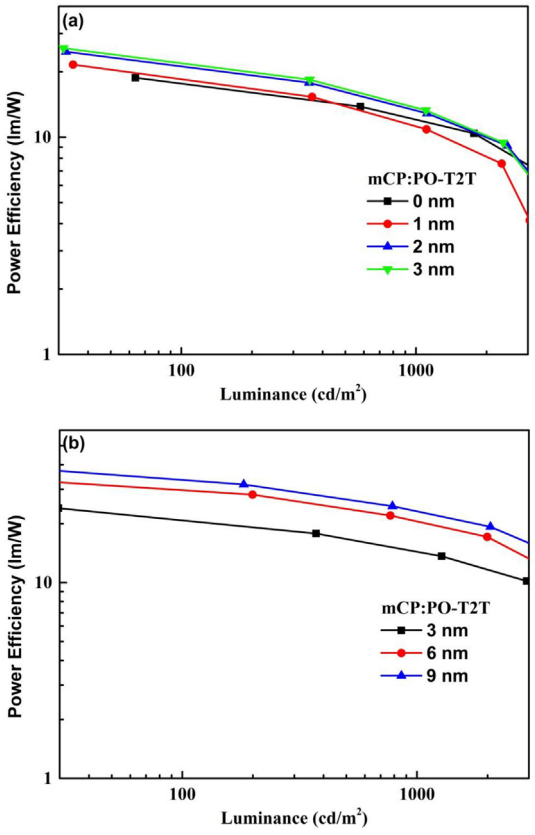

Fig. 6. The power efficiency-luminance curves. (a) Device A-D. (b) Device E-G.

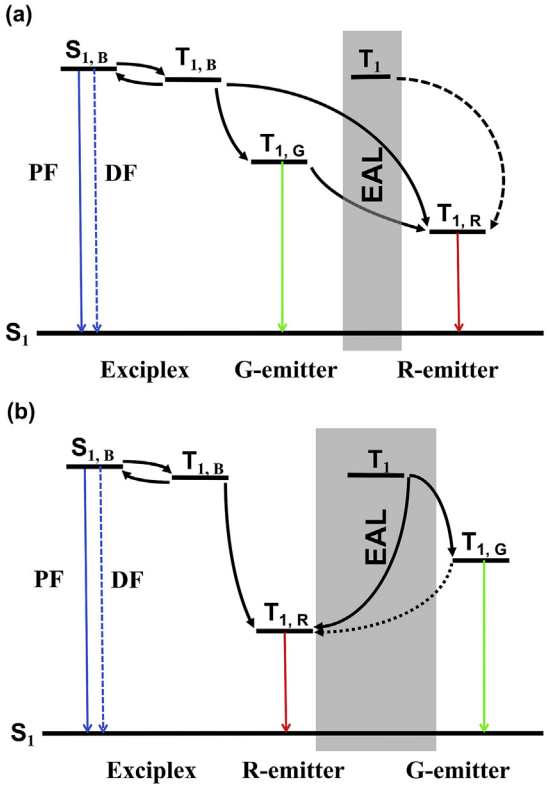

Fig. 7. The schematic diagram of energy transfer process in this paper. (a) Device A-D. (b) Device E-G. The black full line means the efficient energy transfer process and the black dashed line means the invalid energy transfer process. PF: prompt fluorescence; DF: delayed fluorescence.

Table 1
A summary EL performances of all the OLEDs in this paper.

Devices	V_{on} [V]	$CE_{Max.}/PE_{Max.}/EQE_{Max.}$[a] [cd/A/lm/W/%]	$CE_{1000}/PE_{1000}/EQE_{1000}$[b] [cd/A/lm/W/%]	CIE coordinates [6 V] <
A	3.0	20.9/18.8/8.8	16.2/15.2/7.0	(0.475,0.448)
B	3.0	24.1/21.6/10.2	15.8/11.0/6.8	(0.440,0.423)
C	3.0	27.6/24.8/11.5	18.6/14.2/7.7	(0.445,0.437)
D	3.0	28.6/25.7/12.1	19.1/13.4/8.0	(0.451,0.432)
E	3.0	26.7/23.9/11.2	20.4/14.4/8.4	(0.443,0.443)
F	3.0	38.8/34.9/15.3	29.7/20.4/11.6	(0.436,0.467)
G	3.0	45.7/41.0/15.7	34.2/24.1/12.1	(0.374,0.521)

[a] Maximum current efficiency (CE), power efficiency (PE) and EQE.
[b] CE, PE and EQE at 1000 cd/m².

from the energy transfer of 8 nm blue exciplex layer, while the red emission from the energy transfer of 8 nm blue exciplex layer and green emitter (Fig. 7a). However, the thick EAL in Device E-G act as the excitons recombination zone and energy transfer host, simultaneously. The green emission derives from the energy transfer of EAL, and red emission from the energy transfer of 8 nm blue exciplex layer and EAL (Fig. 7b). Thereby, the reasonable excitons distribution and efficient energy transfer due to the application of EAL result in the high efficiency and high spectra stability WOLEDs.

4. Conclusion

In conclusion, we adopt an alternate method to fabricate WOLEDs by combining the exciplex as blue emitter and EAL with phosphorescent ultrathin layer as the green and red emitter. The best performance WOLEDs achieve high efficiency with maximum current efficiency, power efficiency and EQE of 38.8 cd/A, 34.9 lm/W and 15.3% respectively. The change of CIE coordinates is as low as (0.024, 0.013) from 4 V to 6 V. We also discuss the energy transfer process and the function of EAL by comparing two kinds of device structure. We believe that the application of exciplex as EAL and blue emitter can manage the excitons distribution efficiently and it can also be applied to other device structures.

Acknowledgements

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