

Achieving Highly Efficient Simple-Emission Layer Fluorescence/Phosphorescence Hybrid White Organic Light-Emitting Devices via Effective Confinement of Triplets

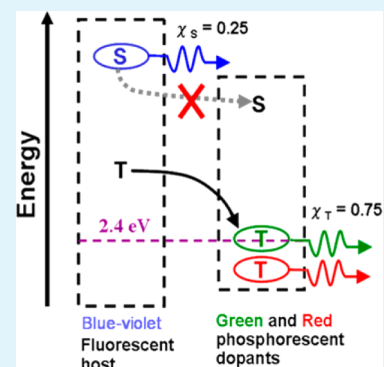
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Supporting Information

ABSTRACT: Achieving high efficiencies in simple device configurations is a long-standing and meaningful target for organic light-emitting devices (OLEDs). Herein, by utilizing an efficient blue-violet fluorophor (CzS1) that has a high triplet energy of 2.62 eV, the significance of effective confinement of the green triplets in fluorescence/phosphorescence hybrid white devices (F/P-WOLEDs) that have highly simplified emission layers (EMLs) containing only RGB emitters was demonstrated. The non-p-i-n warm-white device exhibited excellent performance with a maximum forward power efficiency high up to 42.1 lm W⁻¹, and maintaining at 26.3 lm W⁻¹ at a practical luminance of 1000 cd m⁻².



KEYWORDS: white OLED, blue fluorophor, high triplet energy, simplified structure, high performance

INTRODUCTION

White organic light-emitting devices (WOLEDs) have emerged as a commercially competitive solid-state lighting source, with unique advantages such as lightweight, large-area emission, and potential fabrication on flexible/transparent substrates. For realizing high efficiencies, all singlet and triplet excited states should be harvested for light generation. With this consideration, two approaches have been used for generating white light. The first one is based on fully phosphorescence devices (P-WOLEDs), converting both singlets and triplets into phosphorescent photons via transferring them to heavy metal-based phosphors of two complementary (e.g., blue/orange) or three primary (blue/green/red, RGB) colors. Although several researches on P-WOLEDs have successfully demonstrated a high internal quantum efficiency (IQE, defined as the electron-to-photon conversion efficiency) close to 100%,^{1–3} it still remains a tough challenge to find a sufficiently stable deep-blue phosphor. Accordingly, for practicality concern, another approach was used through incorporating a blue fluorophor instead of the less stable blue phosphor.⁴ The resulting combination of blue fluorescence and other color phosphorescence (green/yellow/orange/red) thus gave rise to so-called hybrid devices (F/P-WOLEDs).^{4–24} Theoretically, the IQE in these hybrid devices can also approach 100%

because of the individually tailored channels for singlets to undergo a fluorescent decay on the blue fluorophor and triplets to diffuse toward those phosphors for phosphorescent emission. Additionally, these resonant energy transfer ways may allow F/P-WOLEDs to achieve higher power efficiency (PE) than P-WOLEDs, which inherently suffer from the exchange energy loss from the host's singlets to the phosphorescent guest's triplets.⁴ Forrest and Thompson et al. first established the concept of F/P-WOLEDs with their devices demonstrating maximum total external quantum efficiency (EQE) of 18.7% and PE of 37.6 lm W⁻¹, respectively.⁴ Toward achieving higher efficiencies, Leo et al. proposed to use a novel deep-blue fluorophor, *N,N'*-di-1-naphthalenyl-*N,N'*-diphenyl-(1,1':4',1'':4'':1'''-quaterphenyl)-4,4'''-diamine(4P-NPD).^{6,9} The relatively high triplet energy (E_T) of 4P-NPD (2.3 eV) is beneficial for suppressing the nonradiative loss of the triplets formed on 4P-NPD by transferring them to the lower E_T (~2.0 eV) orange phosphors nearby. Using 1,3,5-tri(1-phenyl-1H-benzo[d]imidazol-2-yl)-benzene (TPBI) as an additional host for the green phosphor,

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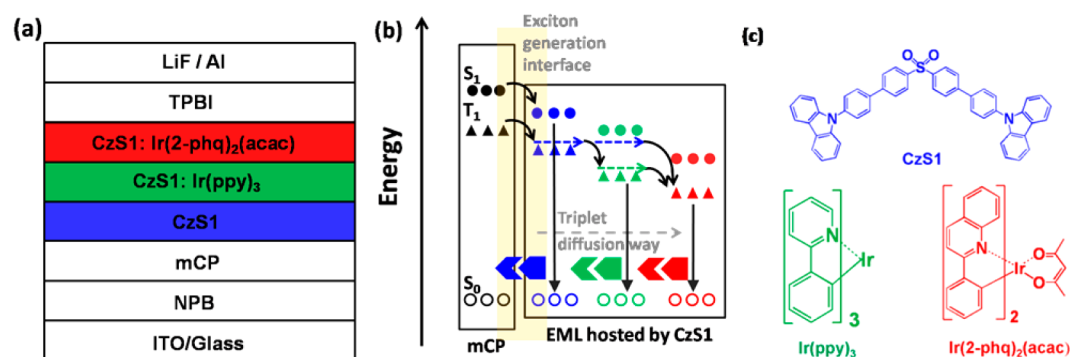


Figure 1. (a) Device configuration, (b) proposed energy transfer and RGB emission mechanism, and (c) RGB emitters of the F/P-WOLED.

a series of 4P-NPD-based RGB F/P-WOLEDs were fabricated. Upon careful design of the emitting layer (EML), the total EQE/PE of these p-i-n-structured devices were constantly enhanced from 16.1% /37.5 lm W^{-1} (separate 4P-NPD and orange layers) to 24.1%/49.3 lm W^{-1} (directly doping the orange phosphor into 4P-NPD at a very low doping concentration) at a luminance of 1000 cd m^{-2} . This shows that introducing a high- E_T blue fluorophor, in other words, a blue fluorescent host, will significantly improve the performance of F/P-WOLEDs provided that the material simultaneously offers good performs as a fluorescent emitter and as a phosphorescent host. Recent related researches have been primarily devoted to exploring F/P-WOLEDs with simplified EMLs, which contain only those necessary emitters for white light emission, including a blue fluorescent host and one or two phosphors while excluding any additional interlayer materials and nonradiative hosts.^{11–18} For examples, Wong's group realized a maximum EQE/PE of 7% /12.8 lm W^{-1} by constructing a blue/yellow F/P-WOLED;¹³ and Jabbour' group fabricated a RGB F/P-WOLED which exhibited EQE of 11% and PE of 14.8 lm W^{-1} at 500 cd m^{-2} , respectively.¹⁴ Very recently, our group has reported a high-performance blue/orange F/P-WOLED based on a sky-blue host, 2,8-di[4-(diphenylamino)phenyl]dibenzothiophene-*S,S*-dioxide (DADBT).¹⁵ The resulting warm white device shows a maximum forward EQE/PE of 15.6% /39.5 lm W^{-1} . Using two other blue hosts 4-(4,6-diphenoxy-1,3,5-triazin-2-yl)-*N,N*-diphenylaniline (POTA)¹⁷ and di[4-(4-diphenylaminophenyl)phenyl]sulfone (DAPSF)¹⁸ with higher E_T for even hosting the green phosphor (>2.4 eV), we further demonstrated efficient three-color-based F/P-WOLEDs, which showed competitive forward EQE/PE reaching a maximum value of 14.7% /35.6 lm W^{-1} . However, performances of these three-color-based F/P-WOLEDs with simplified EMLs are still below their blue/orange-based counterparts. Considering that the human-eye is most sensitive to green light and that green devices have the highest electroluminescence (EL) efficacies among monochrome devices,²⁵ better utilization of green phosphorescence should be beneficial for enhancing the performance of F/P-WOLEDs. Actually, both POTA and DAPSF have triplet energies (2.44 and 2.44 eV) very close to that (2.41 eV) of the green phosphor, *fac*-tris(2-phenylpyridine) iridium ($\text{Ir}(\text{ppy})_3$).²⁶ These comparable energy levels may result in an inefficient confinement of the green triplets, or an endothermic process of energy back-transfer from $\text{Ir}(\text{ppy})_3$ to the fluorescent host, leading to some loss of the green triplets. For instance, the $\text{Ir}(\text{ppy})_3$ -based green device using POTA as the host only obtained a moderately high PE of 52.3 lm W^{-1} , whereas in the

case of DAPSF, the blue fluorescence of the host can be observed and became more obvious at a high luminance. These results suggested that hosts with an even higher E_T (e.g., > 2.5 eV) would be needed for further efficiency enhancement.

Meanwhile, a relatively small energy splitting between the lowest singlet excited state and the lowest triplet excited state (ΔE_{ST}) is also reasonably required for the fluorescent host, in order to maintain its intrinsic fluorescence within the blue color gamut. Because ΔE_{ST} is proportional to the overlap between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), aromatic compounds with spatially separated electron-donor (D) and acceptor (A) are much preferred as these materials typically feature a small ΔE_{ST} .^{11,27} However, when the ΔE_{ST} was remarkably decreased, triplets formed on the fluorophor by electrical excitation will possibly up-convert to singlets through reverse intersystem crossing (ISC) by thermal activation, finally leading to thermally activated delayed fluorescence (TADF).²⁷ Although this reverse ISC process enable harvesting triplet energy without using any phosphorescent material, it goes against the working mechanism in F/P-WOLEDs where the triplets are extracted from the singlets and then harvested by phosphors after diffusing a distance. Zhang et al. recently reported several blue TADF materials having a ΔE_{ST} range of 0.54–0.32 eV, and clearly revealed that the proportion of the delayed fluorescence in solution decreased with an increase in the ΔE_{ST} and almost disappeared in the case of 0.54 eV.²⁸ Accordingly, to minimize the possibility of triplet up-conversion, higher (e.g., above 0.54 eV) but still relatively small ΔE_{ST} may be considered for the fluorescent host in F/P-WOLEDs, and its singlet energy therefore should be shifted to above 3.0 eV, basically corresponding to deep-blue or blue-violet fluorescence.

Herein, basing on such an ideal blue-violet fluorescent host di(4-[4(carbazol-9-yl)phenyl]phenyl)sulfone (CzS1) with a high E_T of 2.62 eV and a moderately small ΔE_{ST} of 0.62 eV, the performance of non-p-i-n F/P-WOLED were successfully enhanced to a higher level with forward EQE and PE reaching 16.4% and 42.1 lm W^{-1} , and maintaining at 15.1% and 26.3 lm W^{-1} at the illumination relevant luminance of 1000 cd m^{-2} , respectively. CzS1 has previously been identified to have a D- π -A molecular structure with an impressive film-state prompt fluorescent quantum yield of 0.98.²⁹ In this work, via exploring the novel function of CzS1 as a phosphorescent host, effective confinement of the green triplets by CzS1 was demonstrated by realizing high EQE/PE up to 19.7% /67 lm W^{-1} in a $\text{Ir}(\text{ppy})_3$ -based green device. Figure 1 shows the device configuration, proposed energy transfer and RGB emission mechanism, and

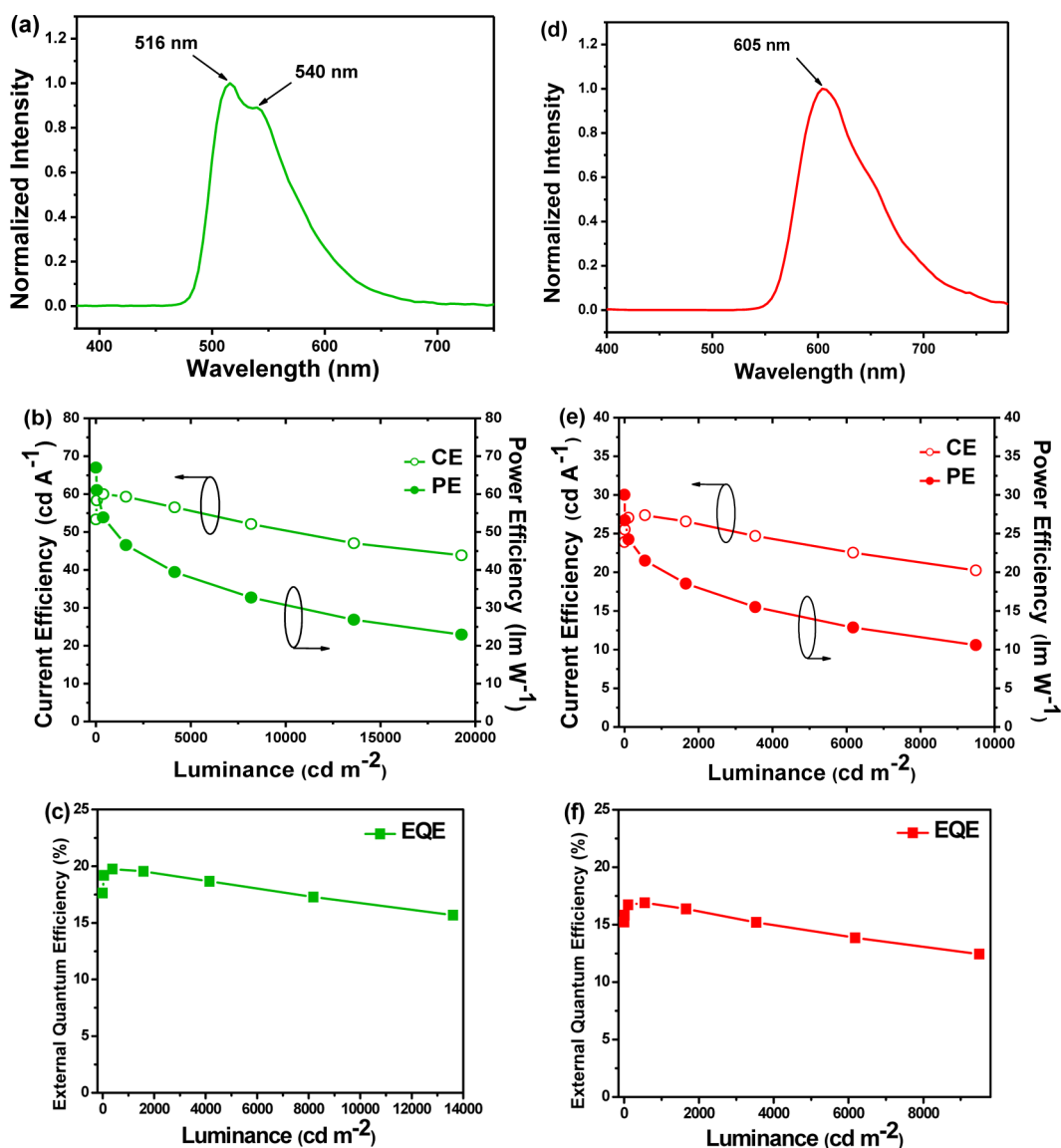


Figure 2. EL spectra, CE–L–PE plots, and EQE–L plots of (a–c) the green device and (d–f) the red device.

the RGB emitters in the F/P-WOLED. Nonradiative loss of triplets formed on the host CzS1 is supposed to be avoided in a rational triplet arrangement that 1,3-bis(*N*-carbazolyl)benzene (mCP) with higher E_T of 2.9 eV easily blocks these high-energy triplets and naturally diffuse them to green phosphors, and then to red phosphors in a step-by-step energy-decreasing order.

RESULTS AND DISCUSSION

The function of CzS1 as a host for green and red phosphorescence was first examined by fabricating two devices with a similar configuration: ITO/NPB (30 nm)/mCP (10 nm)/EML (30 nm)/TPBI (30 nm)/LiF (1.5 nm)/Al. The EML consists of either CzS1 with 4 wt %-doped Ir(ppy)₃ or CzS1 with 4 wt %-doped bis(2-phenylquinoline)-(acetylacetonate)iridium (Ir(2-phq)₂(acac)). Indium tin oxide (ITO) and LiF/Al are the anode and the cathode, respectively; 4,4'-bis[*N*-(1-naphthyl)-*N*-phenylamino]biphenyl (NPB) and TPBI are employed as the hole-transporting layer (HTL) and the electron-transporting layer (ETL), respectively; whereas, mCP is the electron- and exciton-blocking layer (EBL). Figure 2 presents the EL spectra and efficiencies of the two

monochrome devices. Both of the intrinsic green emission from Ir(ppy)₃ and red emission from Ir(2-phq)₂(acac) are cleanly revealed in the EL spectra, without any additional light component from the host or the adjacent functional layers. The Ir(2-phq)₂(acac)-based red device revealed a CIE coordinate of (0.62, 0.37) and achieved maximum current efficiency (CE) and PE of 27.4 cd A⁻¹ and 30 lm W⁻¹, respectively. To the best of our knowledge, these efficiencies currently are the best result for Ir(2-phq)₂(acac)-based monochrome devices with a single-layer EML (see the Supporting Information, Table S1). More importantly, with a CIE coordinate of (0.32, 0.62), the green device hosted by CzS1 also exhibited a high EQE of 19.7% at 380 cd m⁻² and an excellent maximum PE of 67 lm W⁻¹, which is much higher than the result of 52.3 lm W⁻¹ achieved with POTa mentioned above. These high efficiencies along with the clean green phosphorescence should imply an effective confinement of the Ir(ppy)₃ triplets, which can be attributed to the high E_T (2.62 eV) of CzS1. The transient photoluminescence (PL) process of 4 wt % Ir(ppy)₃-doped CzS1 film also reveals a first-order exponential decay at 512 nm with a lifetime of 1.36 μs (see the Supporting Information, Figure S1).

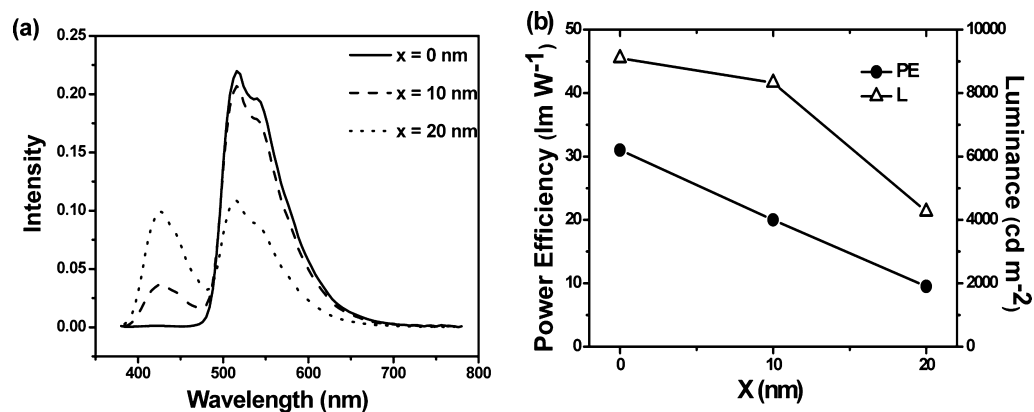


Figure 3. (a) EL spectra and (b) PE- x -L plots of the three probing devices with 10 nm-thick Ir(ppy)₃-doped layer at different distances (x) from the mCP/CzS1 interface.

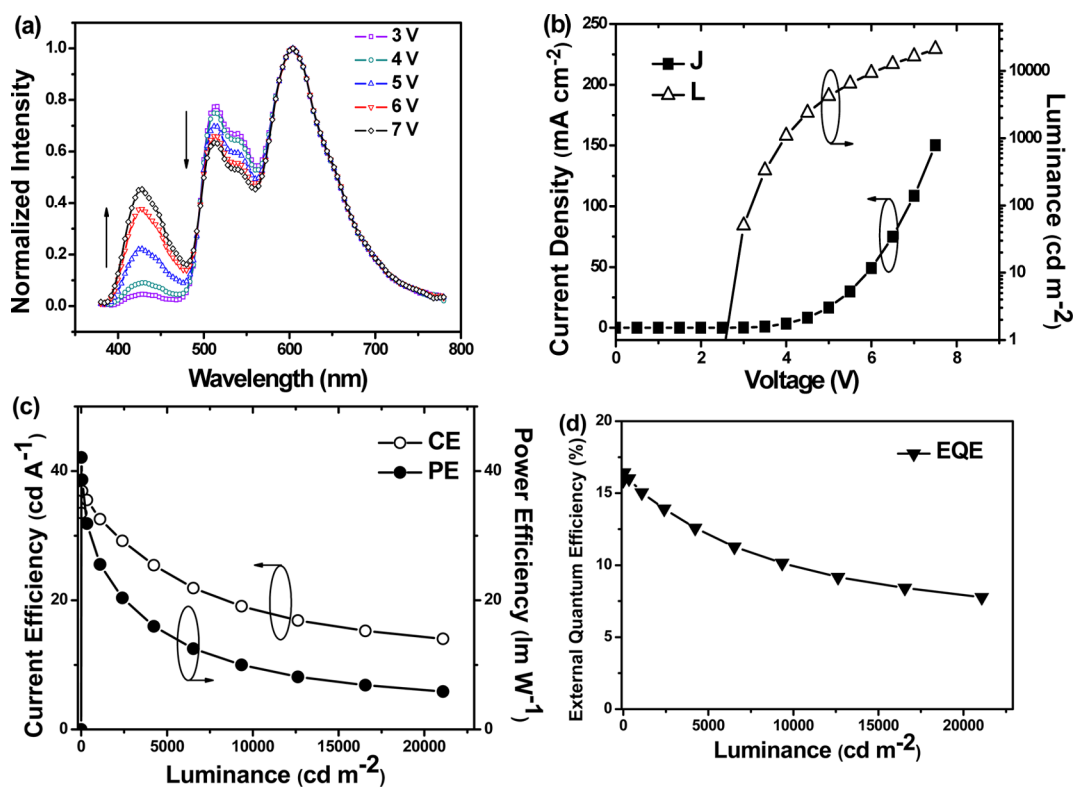


Figure 4. (a) EL spectra at different voltages, (b) J - V - L , (c) CE - L - PE plots, and (d) EQE - L characteristic of the warm-white F/P-WOLED.

Therefore, the blue-violet fluorophor CzS1 has been demonstrated to be an excellent host for both green and red phosphorescence, and this is an important premise toward constructing high-efficiency and simple-EML RGB F/P-WOLEDs.

As the key point to obtain hybrid white emission is to separate triplets from singlets, the location of the carrier recombination zone within the EML thus should be determined before constructing the F/P-WOLED, and a simple partially doping method was used here. Basically, it tends to be at one or two interfaces of the EML because of the charge-hopping energy barriers established by those adjacent transporting layers. With other parts unaltered from the monochrome devices, three probing devices were fabricated with an EML structure of CzS1 (x nm)/CzS1:4 wt % Ir(ppy)₃ (10 nm)/CzS1 (20 - x nm), x was varied between 0, 10, 20

nm. EL spectra, PE, and luminance values as a function of x of these devices at a driving voltage of 5 V are shown in Figure 3. As the doping region was adjacent to the mCP/EML interface ($x = 0$ nm), the EL spectra almost solely consisted of the green emission from Ir(ppy)₃ (Figure 3a), indicative of nearly complete conversion of both singlets and triplets into green phosphorescent emission. This device also exhibited the highest luminance and PE among the three probing devices (Figure 3b). On the other hand, as the doped layer successively moved away from the mCP/EML interface toward the TPBI/EML interface (x was varied from 0 to 10 and then to 20 nm), distinct blue fluorescence of CzS1 peaked at 428 nm was observed. However, the green emission gradually weakened along with apparently lowered device efficiency and luminance. These results primarily suggest that the electron-hole recombination mainly occurs close to the mCP/EML interface.

Given that the recombination zone is free of phosphors, singlets there may have a fast fluorescent decay for their short lifetimes (\sim ns), and the left triplets possessing much longer lifetimes (\sim μ s–s) should rationally diffuse toward the TPBI/EML interface because of the exciton-blocking function of mCP. Thus, RGB white light can be theoretically available once sequentially doping the green and red phosphors along the triplet diffusion route at a balanced color control.

After adjusting the doping concentration and the individual thickness of the R/G/B emitting layers, an optimized F/P-WOLED was obtained with a EML structure of CzS1 (10 nm)/CzS1:2 wt % Ir(ppy)₃ (4 nm)/CzS1:8 wt % Ir(2-phq)₂(acac) (16 nm) and the remaining parts are unchanged from those non-p-i-n-structured monochrome devices (Figure 1a). Figure 1b illustrated the proposed energy transfer and RGB emission mechanism. Near the mCP/EML interface, singlets on CzS1, either directly formed or transferred from mCP, typically are not able to migrate beyond 4 nm,⁴ but undergo rapid blue fluorescent emission. On the other hand, the recombined triplets with much longer lifetimes (longer migration distance \sim 100 nm)⁴ can naturally diffuse along a step-by-step energy-decreasing order: mCP (2.9 eV), CzS1 (2.62 eV), Ir(ppy)₃ (2.41 eV), Ir(2-phq)₂(acac) (2.0 eV). Ir(ppy)₃ harnesses the triplets from CzS1 for green phosphorescent emission while Ir(2-phq)₂(acac) can harvest both the triplets from CzS1 and Ir(ppy)₃, to give red phosphorescent emission. The device results were achieved in the forward direction without the use of any out-coupling enhancement techniques. Figure 4a shows normalized EL spectra of the device at different driving voltages. At higher voltages, the blue component was gradually enhanced along with a slightly reduced green emission. Correspondingly, the CIE coordinates show a blue shift from (0.46, 0.49) to (0.42, 0.40) when the luminance increases from 50 to 10,000 cd m⁻². This spectral change should be strongly correlated with the variation of inherent exciton distribution and activity profile. The electrically generated exciton concentration is high under high driving voltages, especially for the triplets which have a nature of slow decay. T-T annihilation thus becomes obvious, leading to a decrease of relative ratio of active triplet to singlet within the recombination zone. And still, upon high exciton generation, more triplets could enter the red emissive layer or saturate the green phosphors, leading to relative intensity increase of the red emission as compared to the green one. The warm-white device can be turned on (1 cd m⁻² is achieved) at a low voltage of 2.5 V and only required a low driving voltage of 3.9 V at 1000 cd m⁻² (Figure 4b). The efficacies are presented in Figure 4c, d. This F/P-WOLED has maximum forward EQE and PE of 16.4% and 42.1 lm W⁻¹, respectively, and these efficacies remain to be 15.1% and 26.3 lm W⁻¹ at 1000 cd m⁻². The efficiency roll-off could also primarily be attributed to the aforementioned T-T annihilation. However, the PE at this practical luminance is enhanced by about 33% from the PEs of 19.7 lm W⁻¹ and 19 lm W⁻¹ respectively achieved by those previous best two-complementary-color based and RGB-color-based F/P-WOLEDs with highly simplified EMLs (Table 1).^{15,17} This improvement should rationally be attributed to the effective utilization of these excitons, especially the effective confinement of the green triplets by the high- E_T CzS1. Additionally, it should be noticed that in characterizing WOLEDs for white lighting, it is reasonable to also account for light that is waveguided in the glass substrate and other constituting layers, because the resultant photons exiting from

Table 1. Summary of EQE and PE Values of Non-p-i-n-structured F/P-WOLEDs

ref	EQE _{max} (%)	forward PE (lm W ⁻¹)		total ^a PE (lm W ⁻¹)	
		max	1000 cd m ⁻²	max	1000 cd m ⁻²
this work ^b	16.4	42.1	26.3	71.6	44.7
5	6.12				
7		33.2			
8	7.0	11.1			
10	13.1	28.7	12.8		
12 ^b	5.2	4.7			
13 ^b	7	12.8			
14 ^b	12.2				
15 ^b	15.6	39.5	19.7	67.2	33.5
17 ^b	14.8	35.6	19	60.5	32.3
18 ^b	11.9	28.3	15.8	48.2	26.8
19			30.1		
20	16.3	18.5			
21		15.6	14.1		
22		22.5	9.4		

^aReferential total efficiency according to Forrest's research with a result of 1.7 times enhancement as compared to the forward measurement. ^bWith highly simplified EMLs that contain only those necessary emitters for white light emission.

the sides of the device could be redirected in an efficient lamp fixture into the space being illuminated. According to Forrest's research, this total light collection could result in an increase of 1.7 times in PE as compared to the forward measurement.^{4,30} Hence, the total PE in this work may have a potential to exceed 70 lm W⁻¹ and maintain above 40 lm W⁻¹ at 1000 cd m⁻². Table 1 lists the EL efficacy parameters (EQE and PE) of this work as well as other non-p-i-n F/P-WOLEDs reported in literatures. It reveals that our current device efficiencies are among the best results of non-p-i-n-structured F/P-WOLEDs. Comparing with those traditional F/P-WOLEDs, the WOLED here also has a distinct advantage in saving time and reducing cost for the device fabrication process because of the use of the non-p-i-n configuration and the highly simplified EML structure, which consists only of the three RGB emitters.

CONCLUSION

In summary, a blue-violet fluorescent host CzS1 with a high E_T of 2.62 eV and a moderately small ΔE_{ST} of 0.62 eV was used to effectively confine the green triplets in the RGB F/P-WOLED. The Ir(ppy)₃-based green phosphorescent device achieved an inspiring EQE of 19.7% and a high PE of 67 lm W⁻¹. Following the location determination of the hole–electron recombination zone, a highly efficient warm-white F/P-WOLED was fabricated basing on the dual roles of CzS1 as a blue fluorophor as well as a host for the green and red phosphorescence. The WOLED exhibited a maximum forward EQE/PE high up to 16.4% /42.1 lm W⁻¹, and maintained at 15.1% /26.3 lm W⁻¹ at 1000 cd m⁻², the best results for EML-simplified non-p-i-n F/P-WOLEDs. Therefore, it was proven here that effective confinement of green triplets within the simplified EML of RGB F/P-WOLEDs is one indispensable key point to achieve high performance.

ASSOCIATED CONTENT

Supporting Information

Details of OLED fabrication and measurement, transient PL decay profile, Table S1 of the efficiency summary of Ir(2-

phq)₂(acac)-based single-layer-EML red phosphorescent OLEDs. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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