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Author(s)	Luo, Dongxiang; Xiao, Ye; Hao, Mingming; Zhao, Yu; Yang, Yibin; Gao, Yuan; Liu, Baiquan
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Doping-free white organic light-emitting diodes without blue molecular emitter: An unexplored approach to achieve high performance via exciplex emission

Dongxiang Luo,¹ Ye Xiao,¹ Mingming Hao,¹ Yu Zhao,¹ Yibin Yang,¹ Yuan Gao,³ and Baiquan Liu^{2,3,a)}

¹School of Materials and Energy, Guangdong University of Technology, Guangzhou 510006, China

²Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, China

³LUMINOUS! Center of Excellence for Semiconductor Lighting and Displays, School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, 639798 Singapore

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Doping-free white organic light-emitting diodes (DF-WOLEDs) are promising for the low-cost commercialization because of their simplified device structures. However, DF-WOLEDs reported thus far in the literature are based on the use of blue single molecular emitters, whose processing can represent a crucial point in device manufacture. Herein, DF-WOLEDs without the blue single molecular emitter have been demonstrated by managing a blue exciplex system. For the single-molecular-emitter (orange or yellow emitter) DF-WOLEDs, (i) a color rendering index (CRI) of 81 at 1000 cd/m² can be obtained, which is one of the highest for the single-molecular-emitter WOLEDs, or (ii) a high efficiency of 35.4 lm/W can be yielded. For the dual-molecular-emitter (yellow/red emitters) DF-WOLED, a high CRI of 85 and low correlated color temperature of 2376 K at 1000 cd/m² have been simultaneously achieved, which has not been reported by previous DF-WOLEDs. Such presented findings may unlock an alternative avenue to the simplified but high-performance WOLEDs. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4975480>]

Generally, the doping technique (i.e., the p-doping and n-doping charge transport layers, doping emitting layers (EMLs)) is necessary to achieve high-performance organic light-emitting diodes (OLEDs), particularly for white OLEDs (WOLEDs).^{1–5} However, this technique may (i) result in the complex control system of the codeposition rate and precise dopant concentration (i.e., 0.1% or even lower) in fabrication processes for EMLs,^{6,7} (ii) need to carefully select hosts for different-color guests, (iii) consider the effect of additional functional layers for both the hosts and guests in EMLs, and (iv) enhance the cost for using hosts in EMLs.^{8–10} To loosen the bottlenecks, the doping-free technology is conducive, since it can simplify the device engineering, shorten the fabrication procedure, avert the host, and lower the cost.^{11–17} Recently, both monochromatic and white doping-free OLEDs have been actively studied. For example, Adachi *et al.* established doping-free blue and green OLEDs via thermally activated delayed fluorescent (TADF) emitters, achieving ~100% internal quantum efficiency.¹¹ For doping-free WOLEDs (DF-WOLEDs), Omary *et al.* built an all-phosphorescent WOLED via a metal-organic Pt(II)-pyridylazolate phosphor, achieving a maximum external quantum efficiency (EQE) of ~14%.¹² We constructed hybrid WOLEDs via ultrathin EMLs, yielding a series of high-performance doping-free devices.¹³ Hybrid WOLEDs refer to the devices adopting blue fluorescent emissions and green/red or complementary phosphorescent emissions.^{18–20} Based on these facts, it can be concluded that the doping-free technology is an effective scheme to develop

OLEDs. Among DF-WOLEDs, it is noted that blue single molecular emitters are necessary. However, the manipulation of blue single molecular emitters is not an easy thing, particularly for hybrid WOLEDs. For instance, Forrest *et al.* built a hybrid WOLED requiring interlayers between blue fluorophors and green/red phosphors to prohibit mutual energy transfer and exciton quenching.¹⁸ Leo *et al.* reported the triplet-harvesting concept to fabricate hybrid WOLEDs, in which blue fluorophors required high triplet energies.¹⁹ Lee *et al.* fabricated a single-EML hybrid WOLED, which required a fluorescent material functioning as both the host of the orange guest and the blue emitter.⁷ Duan *et al.* established a hybrid WOLED, in which blue TADF emitters are required to avoid the external heavy-atom effect.²⁰ In fact, regardless of doping or doping-free hybrid WOLEDs, one of the most significant device engineering is the management of blue single molecular emitters.^{5–7,13,18–20}

On the other hand, the exciplex emission, which is generated with charge-transfer characters between electron-donating and electron-accepting molecules, is becoming a hot topic recently.^{21–23} By codoping electron-donating and electron-accepting materials in one layer or depositing the two kinds of materials sequentially, the exciplex emission can be formed. Particularly, the effect of codeposition rate is avoided for the doping-free exciplex system, which can be more easily controlled in fabrication processes. For example, Su *et al.* organized fluorescent organic planar pn heterojunction devices with TADF properties, obtaining a doping-free yellow exciplex emission with an EQE of 12.02%.²⁴ Also, the red and green emissions have been realized in their work. However, it is seen that the doping-free blue exciplex

^{a)}E-mail: bqliu1012@gmail.com

emission is rarely reported, which may not be beneficial to the applications of full-color displays and solid-state lightings. Besides, if doping-free blue exciplex system can be used to design DF-WOLEDs, DF-WOLEDs without the blue single molecular emitter may be developed, although no such work has been documented.

Herein, we have developed high-performance DF-WOLEDs without the blue single molecular emitter by managing the doping-free blue exciplex emission. Due to the combination of blue fluorescent emission and complementary phosphorescent emission, this kind of device is also classified into hybrid WOLEDs.^{18–20} Therefore, the difficulty of manipulation of blue single molecular emitters in hybrid WOLEDs may be alleviated by dint of this approach. As displayed in Figure 1, the configuration of single-molecular-emitter WOLEDs is indium tin oxide (ITO)/1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN) (100 nm)/4, 4', 4''-tris[3-methylphenyl(phenyl)amino]triphenylamine (m-MTDATA) (20 nm)/(fbi)₂Ir(acac) (0.6 nm)/m-MTDATA (X nm)/3-(4-Biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) (10 nm)/1,3,5-tri(*m*-pyrid-3-yl-phenyl)benzene (TmPyPB) (35 nm)/LiF (1 nm)/Al (160 nm), where X = 0, 3, 6, and 9 for devices W11, W12, W13, and W14, respectively. Here, indium tin oxide (ITO) is the anode, 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN) is the hole injection layer (HIL), 20 nm 4, 4', 4''-tris[3-methylphenyl(phenyl)amino]triphenylamine (m-MTDATA) is the hole transport layer (HTL), bis(2-(9,9-diethyl-9H-fluoren-2-yl)-1-phenyl-1Hbenzimidazole-N,C³)iridium (acetylacetonate) (fbi)₂ Ir(acac) is the orange phosphorescent emitter, m-MTDATA and 3-(4-Biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) is the blue exciplex system, 1,3,5-tri(*m*-pyrid-3-yl-phenyl)benzene (TmPyPB) is the electron transport layer (ETL), LiF is the electron injection layer, and Al is the cathode. The structure of device W2 (Figure 1) is similar to that of W12 except for the 0.6 nm yellow emitter

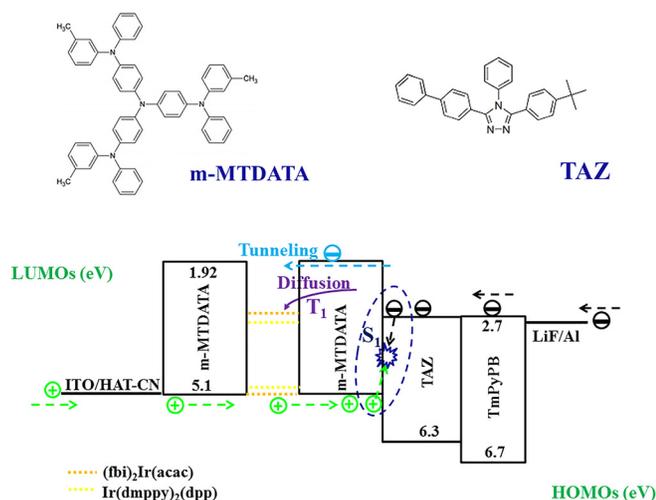


FIG. 1. Top: Molecular structures of m-MTDATA and TAZ. Bottom: Schematic diagrams of the emission mechanisms of DF-WOLEDs without the blue single molecular emitter. Holes and electrons can be effectively injected and transported to the m-MTDATA/TAZ interface, forming exciplexes. When the m-MTDATA layer is thin, electrons can pass through it via the tunneling effect and then meet holes injected from the anode to form excitons directly on the phosphors while the triplets of exciplexes can be harvested by phosphors via the diffusion process.

bis(2-phenyl-4,5-dimethylpyridinato)[2-(biphenyl-3-yl)pyridinato] iridium(III) (Ir(dmppy)₂(dpp)). The detailed fabrication and measurement of devices followed well-established processes and as reported elsewhere.¹³

To achieve DF-WOLEDs without the blue single molecular emitter, some strategies have been employed. First, a doping-free charge confining structure has been utilized. HAT-CN is selected as the HIL due to its strong hole injection ability, and it is demonstrated that the thickness of HAT-CN shows almost no effect on the electrical characteristic of devices.^{13,25,26} Here, we have used a thick HAT-CN (100 nm), which can better planarize the ITO surface compared with the thin HAT-CN, improving the device performance.¹³ m-MTDATA has been used as the HTL, which can (i) reduce the number of evaporation sources, since m-MTDATA also functions as the donor in the exciplex system; (ii) confine electrons since m-MTDATA has a high LUMO (lowest unoccupied molecular orbital) of 1.92 eV.²⁷ Although TAZ is an n-type material, its electron mobility is low (10^{-5} cm²/V s).⁵ Hence, TmPyPB is selected as the ETL due to its high electron mobility (10^{-3} cm²/V s) and the same LUMO with TAZ (2.7 eV), which can effectively improve the electron injection.²⁸ Then, to compensate the blue emission, doping-free phosphorescent EMLs have been utilized, since they can harvest both singlet and triplet excitons. Significantly, phosphorescent EMLs can harvest the triplets unconsumed in the exciton generation zone via the diffusion process, further enhancing the efficiency. Finally, to realize this kind of WOLED, a doping-free blue exciplex system is explored.

Taking the above factors into account, we first demonstrate that the blue exciplex emission can be generated by using m-MTDATA and TAZ. Figure 2 depicts the photoluminescent (PL) spectra of m-MTDATA, TAZ, and m-MTDATA:TAZ (1:1) mixed films together with the electroluminescent (EL) spectrum of the m-MTDATA/TAZ device (device B1, the structure is ITO/HAT-CN (100 nm)/m-MTDATA (20 nm)/TAZ (10 nm)/TmPyPB (35 nm)/LiF (1 nm)/Al (160 nm)). Compared with PL spectra of m-MTDATA and TAZ, m-MTDATA:TAZ has a new red-shifted broad emission with a

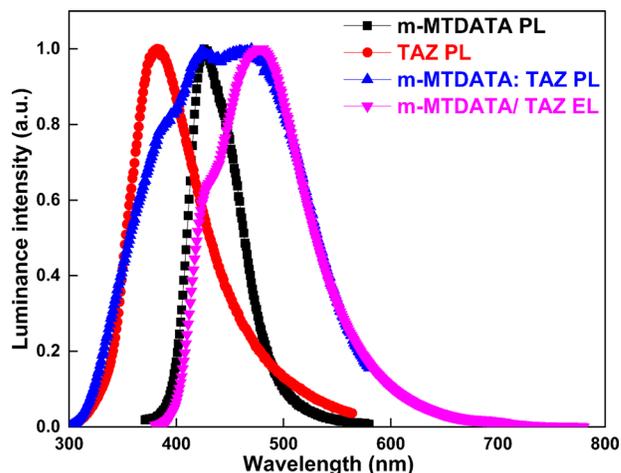


FIG. 2. Photophysical properties of various samples. Normalized PL spectra of m-MTDATA, TAZ, and m-MTDATA:TAZ films, together with normalized EL spectrum of B1.

peak at 470 nm, accompanied by a PL shoulder from m-MTDATA. The energy of the new emission (2.64 eV) is close to the energy gap between the HOMO of m-MTDATA and LUMO of TAZ, indicating that m-MTDATA and TAZ form the exciplex excited state.^{21–24} Besides, since m-MTDATA and TAZ are well-known p-type and n-type materials, respectively, no EL emission has been previously reported for them, which can further indicate that the EL emission originates from the exciplex between m-MTDATA and TAZ. Significantly, the full width at half maximum of B1 is 110 nm, which is among the widest blue exciplex systems.

On this ground, we explored the possibility to avail the blue emission of such exciplex system to design DF-WOLED without the blue single molecular emitter. To demonstrate the possibility of this kind of WOLED, we have first combined the blue exciplex system with an orange phosphorescent emitter (fbi)₂Ir(acac) to achieve the white emission.²⁹ The difference of W11, W12, W13, and W14 is the different thickness of electron-donating layer m-MTDATA. As shown in the inset of Figure 3(a), W11, W12, W13, and W14 exhibit almost no blue emission but strong orange emission, warm-white emission, pure-white emission, and strong blue emission but weak orange emission, respectively. These

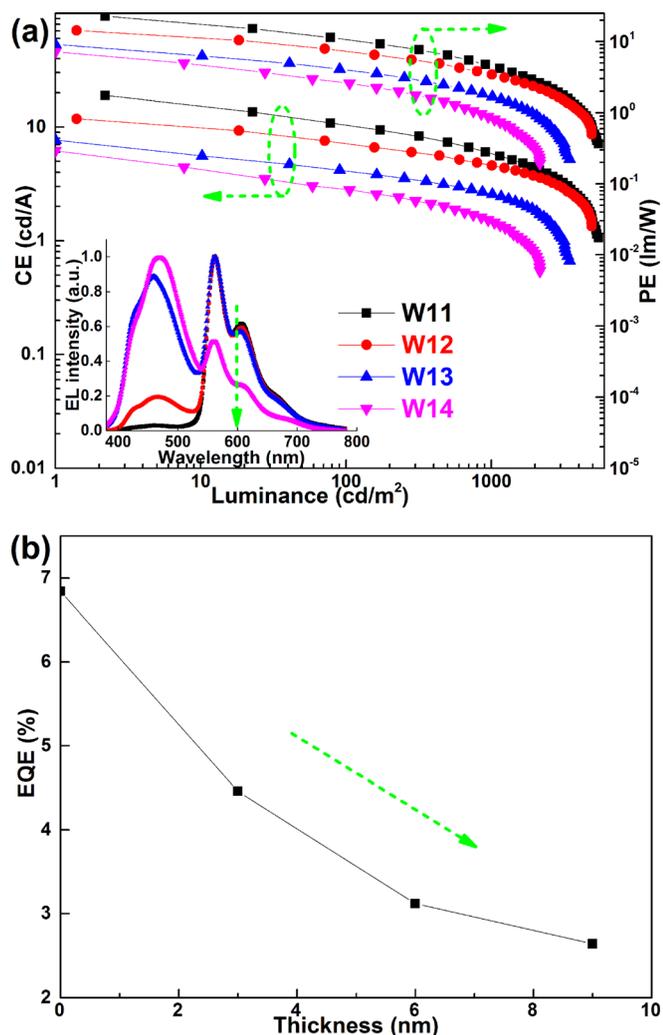


FIG. 3. (a) Forward-viewing current efficiency (CE) and power efficiency (PE) as a function of luminance. Inset: EL spectra at 1000 cd/m². (b) EQE as a function of the m-MTDATA thickness (at a luminance of 1–10 cd/m²).

phenomena can be explained as follows. For these structures, due to the existed large energy barriers together with the fact that m-MTDATA is a p-type material while TAZ is an n-type material, charges will be accumulated at the m-MTDATA/TAZ interface, forming exciplexes, as shown in Figure 1. As a result, both singlet and triplet excitons are generated with a ratio of 1:3 in the narrow region of this interface.²⁹ Singlets can be easily harvested to furnish the blue exciplex emission, while triplets are decayed non-radiatively in device B1 considering the low EQE (0.5%). In WOLEDs, however, triplets can be harvested by the phosphors via the diffusion process.¹⁹ For W11, it has no m-MTDATA layer between (fbi)₂Ir(acac) and TAZ, indicating that (fbi)₂Ir(acac) is located in the exciton generation zone, which is formed by the HTL m-MTDATA and TAZ. As a result, most of the singlet and triplet excitons are harvested by (fbi)₂Ir(acac), leading to the strong orange emission. However, since the thickness of (fbi)₂Ir(acac) is ultrathin, singlets cannot be totally consumed by it, leading to the fact that the poor blue exciplex emission can be generated from the HTL m-MTDATA and TAZ. One may expect that the blue emission can be totally eliminated by increasing the thickness of (fbi)₂Ir(acac); however, due to the aggregation-caused quenching,³⁰ the device efficiency is decreased by thicker (fbi)₂Ir(acac) (the EML thickness has been optimized in the lab). Hence, it is reasonable that W11 exhibits almost no blue emission but strong orange emission. Considering the similar structure of W11 and B1 except for the addition of (fbi)₂Ir(acac), (fbi)₂Ir(acac) can harvest most of the singlet and triplet excitons since the EQE of W11 (6.8%) is 13.7 times higher than that of B1 (0.5%), as shown in Table I. With the increasing thickness of electron-donating layer m-MTDATA, (i) electrons will be difficult to pass through this layer via the tunneling effect because the tunneling probability is decreased when increasing the thickness of materials,^{31,32} and electrons will be difficult to pass through this layer since m-MTDATA is a p-type material, leading to the fact that less excitons can be formed directly on (fbi)₂Ir(acac) and (ii) triplets generated in the exciton generation zone will be difficult to diffuse to the (fbi)₂Ir(acac) layer via the triplets diffusion process, resulting in less triplets being harvested by (fbi)₂Ir(acac).¹⁹ Although the triplets diffusion is as long as ~100 nm because of the intrinsically long lifetime of triplets,¹⁸ the steady-state exciton density will be exponentially decreased with the increasing distance from the exciton generation zone, assuming lateral homogeneity of the organic layer and isotropic materials properties. On the other hand, since the triplet energy (T₁) of m-MTDATA (2.6 eV) is higher than that of exciplexes, m-MTDATA can prevent the diffusion of excitons.²⁵ And the critical thickness of the exciton-blocking layer with high T₁ can be ~7 nm,³³ indicating that few triplets can be harvested via the diffusion process in W14. As a result, the orange intensity is decreased with the increasing thickness of m-MTDATA by the above two factors (tunneling and diffusion). These analyses can be further confirmed by their efficiencies. Since less excitons can be harvested by (fbi)₂Ir(acac), the efficiencies are decreased with the increasing m-MTDATA thickness, as shown in Figure 3(b). However, since the singlets diffusion is very small, they can be harvested to produce the blue exciplex

TABLE I. Summary of performances of fabricated devices.

Device	V_{on}^a (V)	$EQE_{max/1000}^b$ (%)	$CE_{max/1000}^c$ (cd/A)	$PE_{max/1000}^d$ (lm/W)	CIE ^e	CRI ^f	CCT ^g (K)
B1	2.55	0.5/0.4	0.86/0.71	0.92/0.36	(0.1834, 0.2462)
W11	2.55	6.84/2.1	18.9/5.9	22.9/4.6	(0.4957, 0.4759)	41	2733
W12	2.6	4.46/1.74	11.8/4.6	14.3/3.4	(0.4313, 0.4219)	53	3239
W13	2.65	3.12/1.04	7.7/2.6	9.1/1.7	(0.3075, 0.3052)	81	7063
W14	2.7	2.64/0.63	6.2/1.5	7.2/0.8	(0.2387, 0.2631)

^aThe turn-on voltage (1 cd/m²).

^bMaximum EQE and EQE at 1000 cd/m².

^cMaximum CE and CE at 1000 cd/m².

^dMaximum PE and PE at 1000 cd/m².

^eCommission International de l'Éclairage (CIE) coordinates at 1000 cd/m².

^fCRI at 1000 cd/m².

^gCCT at 1000 cd/m².

emission in W12 and W13 with the suitable thickness of m-MTDATA, leading to the balanced emissions. Here, excitons formed on m-MTDATA and TAZ are not considered, since they are much less compared with that of the exciplex system. In terms of the practical use, the CRI is an indication of how well the light source will render colors of objects it illuminates, which must be considered when designing WOLEDs for lighting.^{34–36} For a source to be human eye-friendly, a WOLED with a CRI above 80 is required.² Remarkably, W13 exhibits a CRI of 81 at 1000 cd/m², which is one of the highest for single-molecular-emitter WOLEDs.

Although W12 and W13 possess white emissions, their efficiencies are not high (Figure 3(a) and Table I). To further enhance the efficiency of DF-WOLEDs without blue single molecular emitter, a yellow phosphor Ir(dmppy)₂(dpp) has been used to replace (fbi)₂Ir(acac) (device W2).³⁷ As shown in Figure 4, W2 exhibits a maximum forward-viewing PE of 20.8 lm/W, which is ~1.5 times higher than that of W12. As illumination sources are generally characterized by their total emitted power and the total efficiency is larger by a factor of 1.7 than the forward-viewing efficiency,^{18–20} the maximum total PE of W2 is 35.4 lm/W, which is close to that of fluorescent lamps (40–70 lm/W) and even higher than that of some DF-WOLEDs with all-phosphorescent emitters,¹⁵

indicating the advance of this kind of device. The maximum total EQE and CE are 9.9% and 30.6 cd/A, respectively. These facts also indicate that the performance of this kind of WOLED can be further improved, if more efficient blue exciplex systems or efficient complementary emitters can be obtained.

Finally, it is noted that CRI and CCT are two significant parameters to measure the quality of lightings. In general, CRI should be ≥ 80 for indoor lightings,² while it is reported that lightings with low CCTs are more beneficial to human's health.^{20,38–40} To demonstrate that WOLEDs with the blue single molecular emitter-free and doping-free architecture can satisfy both CRI and CCT requirements, the W3 device has been fabricated including two thin layers of red and yellow iridium emitters and the m-MTDATA/TAZ exciplex system. The structure is ITO/HAT-CN (100 nm)/m-MTDATA (20 nm)/Ir(piq)₃ (0.3 nm)/m-MTDATA (3 nm)/Ir(dmppy)₂(dpp) (0.6 nm)/m-MTDATA (3 nm)/TAZ (10 nm)/TmPyPB (35 nm)/LiF (1 nm)/Al (160 nm), where tris(1-phenylisoquinolinolato-C²,N) iridium(III) (Ir(piq)₃) is the red emitter¹³ and 3 nm m-MTDATA is used as an emission-regulating layer between the red and yellow EMLs to guarantee the high performance. Without this emission-regulating layer, only red-emission-dominating spectra can be obtained (as shown in the inset of Figure 4). To date, no DF-WOLED simultaneously achieving high CRI (≥ 80) and low CCT (≤ 2500 K) has been reported. As shown in Figure 5, W3 exhibits a CRI of 85 and CCT of 2376 K, indicating that W3 can satisfy the demand of physiologically friendly indoor lightings. The maximum total of CE and PE are 20.9 cd/A and 23.5 lm/W, respectively. These efficiencies of W3 are lower than those of W2, which may be attributed to the additional use of m-MTDATA as the emission-regulating layer between yellow and red EMLs. However, the maximum total EQE is 11.3%, higher than that of W2, since the EQE is decided by the spectra besides the current density and luminance. Lower CE and PE but higher EQE of three-color WOLEDs than those of two-color WOLEDs have also been reported in other works.¹⁴

In summary, high-performance DF-WOLEDs without the blue single molecular emitter have been developed by managing a doping-free blue exciplex system. The single-molecular-emitter (orange or yellow emitter) DF-WOLEDs can exhibit: (i) one of the highest CRI for single-molecular-

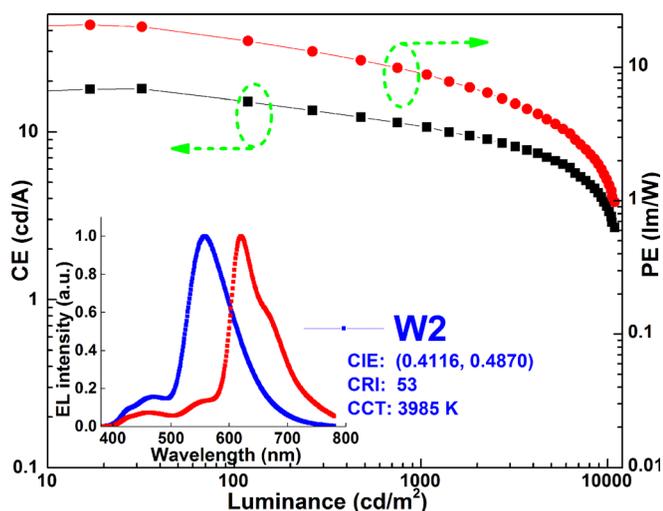


FIG. 4. Forward-viewing CE and PE as a function of luminance of W2. Inset: the spectra of W2 (blue line) and W3 lacking the m-MTDATA layer between the red and yellow EMLs (red line) at 1000 cd/m².

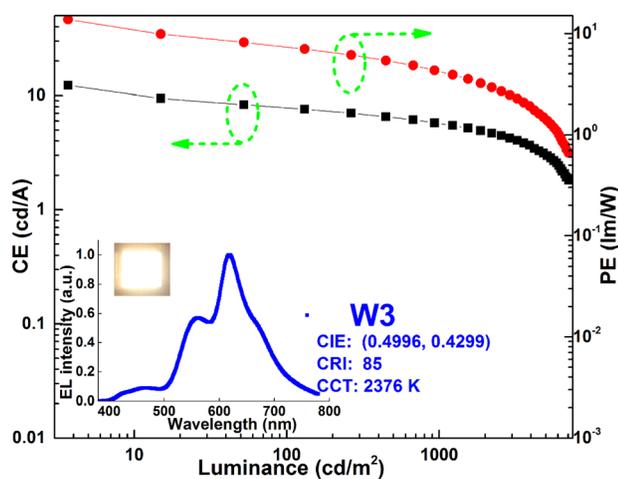


FIG. 5. Forward-viewing efficiencies of W3. Inset: the EL spectrum at 1000 cd/m^2 and the photograph of W3.

emitter WOLEDs (81) or (ii) a high efficiency of 35.4 lm/W , which is close to that of the fluorescent lamps. The dual-molecular-emitter (yellow/red emitters) DF-WOLEDs can simultaneously achieve high CRI (≥ 80) and low CCT (≤ 2500 K), which cannot be realized by previous DF-WOLEDs. The findings may unlock an alternative avenue to the simplified but high-performance WOLEDs, which is promising for the next-generation lightings and displays.

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