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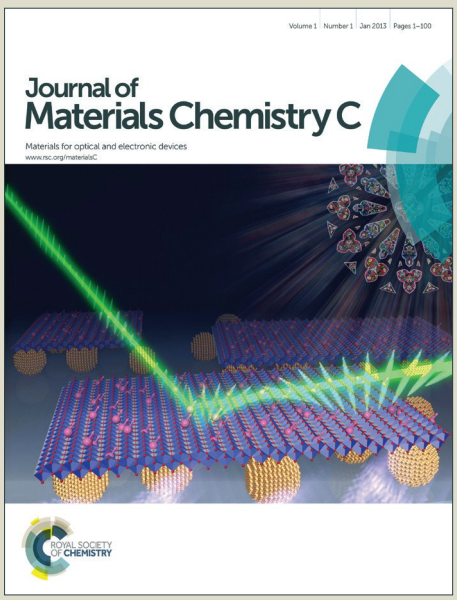

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A host-guest system comprising high guest concentration to achieve simplified and high-performance hybrid white organic light-emitting diodes

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Abstract: Single-emitting-layer (single-EML) hybrid white organic light-emitting diodes (WOLEDs) have attracted a great deal of attention due to their simplified structures. However, the guest concentration is usually too low, which is quite difficult to control and reproduce in the coevaporation process. Herein, for the first time, N,N'-di(naphthalene-1-yl)-N,N'-diphenyl-benzidine has been used as both the host and blue emitter in single-EML WOLEDs. By dint of this multifunctional material, the concentration is as high as 1.5%. This device exhibits a maximum total efficiency of 65.3 lm/W, indicating a significant step towards the real commercialization. Besides, low voltages (i.e., the turn-on voltage is 2.4 V and 3.45 V at 1000 cd/m^2) and a color rendering index (CRI) of 77 are obtained for this two-color WOLED. Unlike the working mechanisms in previous single-EML hybrid WOLEDs with low guest concentrations, devices comprising high concentrations exhibit more sophisticated engineering, in which the device smartly allow the utilization of both the

fluorescence from the host itself, and the complementary phosphorescence from the guest by incomplete Förster energy transfer, Dexter energy transfer as well as direct exciton formation on the guest. Moreover, we have incorporated this unique host-guest system into a dual-EML hybrid WOLED. A maximum efficiency of 17.2 lm/W and 10.2 lm/W at 1000 cd/m^2 (3.85 V) with an ultrahigh CRI of 93 are achieved, providing a new opportunity to accomplish the simplified structure/ low voltage/ high efficiency/ ultrahigh CRI trade-off.

Keywords: white, organic light-emitting diodes, hybrid, single-emitting-layer, color rendering index

1, Introduction

White organic light-emitting diodes (WOLEDs) have been the object of intense research owing to their extraordinary characteristics, such as high efficiency, fast switching, light weight, long lifetime and flexibility.[1-3] The field has evolved to the extent that commercial applications for cell-phones, televisions and indoor lighting are available nowadays. Generally, three types of WOLEDs are reported according to the employed emissive materials, including all-phosphorescent WOLEDs, all-fluorescent WOLEDs and hybrid WOLEDs which are based on hybrid (fluorescent (F) and phosphorescent (P)) emitters schemes.[4] To boost the efficiency, the utilization of P emitters is desirable since they can allow for a conversion of up to 100% of injected charges into emitted photons (both singlet and triplet excitons are harvested), resulting in a theoretical internal quantum efficiency of unity.[5-7] Unfortunately, there is no ideal blue P emitter in terms of lifetime and color-stability

until now, restricting the development of all-phosphorescent WOLEDs.[8]

To address the above issues, the utilization of hybrid WOLEDs, which usually combine stable blue F emitters with efficient green-red/orange P emitters, is considered to be an alternative way due to their merits, like high efficiency, stable color, high color rendering index (CRI) and long lifetime.[4] To date, two approaches have been used to create hybrid WOLEDs according to the triplet energy (T_1) of blue F emitters. On the one hand, hybrid WOLEDs can be constitutive of blue fluorophors with low triplet energies, which are lower than those of complementary phosphors. A critical feature of preparing this kind of devices is utilizing an interlayer (IL), locating between the blue F emitter and the complementary P emitter, which can i) prevent Förster energy transfer from F blue emitters to red-green/orange P emitters, ii) eliminate the Dexter energy transfer from red-green/orange P emitters to F blue emitters, iii) tune the emission colors.[4,9-12] For this kind of WOLEDs, Sun et al. developed the first device by employing the combination of stacked F-IL-P-P-IL-F emitters to manage both singlet and triplet excitons, obtaining a maximum forward-viewing power efficiency (PE) of 22.1 lm/W.[4] Wong et al. reported a two-element hybrid WOLED with a P-IL-F structure by utilizing a novel heteroleptic orange-emitting Ir-complex, obtaining an efficiency of 13.5 lm/W.[9] Ma et al. set up a hybrid WOLED with a P-P-IL-F structure by using a bipolar IL as the charge carrier switch, obtaining an efficiency of 32.0 lm/W.[10] Previously, we realized a P-IL-F concept by investigating and optimizing of each organic layer, achieving a forward-viewing efficiency of 58.4 lm/W.[11] We also constructed an efficient hybrid

WOLED by studying the effect of n-type IL, achieving a long lifetime of $>10^8$ h at 100 cd/m^2 ($>3\times10^5$ h at 1000 cd/m^2).[12]

On the other hand, hybrid WOLEDs can be composed of blue fluorophors with high triplet energies, which are higher than those of complementary phosphors. As a result, an advantage of this kind of WOLEDs is no using IL, which can simplify the structures.[13-15] Leo et al. demonstrated the triplet harvesting strategy in a triple-emitting-layer (triple-EML) hybrid WOLED by using the blue emitter *N*,*N*′-di-1-naphthalenyl-*N*,*N*′-diphenyl-[1,1′:4′,1″:4″,1‴-quaterphenyl]-4,4‴-diamine (4P-NPD) with a high T_1 of 2.3 eV, rendering it possible to harvest triplets by letting them diffuse to an orange P Ir-complex with a T_1 of 2.0 eV.[13] However, these devices are not simplified enough due to the use of multi-EML structures.[13-15] Therefore, single-EML hybrid WOLEDs have been the recent research interest to address this issue, since they only need one EML comprised of a high T_1 blue host doped with different color guests, offering many advantages, such as simpler structures, shorter fabrication processes, less structural heterogeneities inherent in the multiple-color band architectures, lower voltages, lower cost and so forth. As a matter of fact, some endeavors about single-EML hybrid WOLEDs have been made. For example, Hung et al. fabricated a deep-blue electrofluorescence/yellow-green electrophosphorescence single-EML hybrid WOLED with a guest concentration of 0.1%, yielding a PE of 12.8 lm/W.[16] Ma et al. set up a single-EML WOLED with a concentration of 0.03%, yielding a total PE of 80 lm/W.[17] Zhang and Lee et al. realized a single-EML hybrid WOLED with a concentration of 0.1% by using a bipolar F material as the host of

orange guest and the sky-blue emitter, yielding a total PE of 67.2 lm/W.[18] They also realized a single-EML WOLED with a concentration of 0.2% via a blue fluorophor with small singlet-triplet splitting, yielding a total PE of 57.3 lm/W.[19] Based on these facts, it can be concluded that the use of single-EML structure is a promising manner to develop hybrid WOLEDs.

Although the efficiencies of these representative WOLEDs are respectable, it is easily seen that the guest concentrations are too low, [16-19] which is quite difficult to control and reproduce in the coevaporation process, limiting their real commercialization.[20,21] Therefore, single-EML hybrid WOLEDs with high guest concentrations (＞1%) are desperately needed. However, despite a large number of blue hosts have been used to realize single-EML hybrid WOLEDs, only two materials 4P-NPD and bis[2-(2-hydroxyphenyl)-pyridine] beryllium ($Bepp_2$) can fulfill the requirements of high guest concentrations to date, as far as we know. [22-25] As a result, the highest efficiency for the single-EML hybrid WOLEDs with high concentrations is only 48.8 lm/W, which was reported by Wang et al. with a concentration of 2%,[23] indicating that an effort is urgently needed to further enhance the efficiency. Besides, whether the working mechanisms of single-EML hybrid WOLEDs with high guest concentrations are same as those of devices with low concentrations, which is still unclear. Moreover, how to fully take advantage of the host-guest system obtained from the single-EML WOLEDs has never been documented.

In this paper, we, for the first time, have demonstrated that N,N’-di(naphthalene-1-yl)-N,N’-diphenyl-benzidine (NPB) can be used as both the

host and blue emitter in single-EML hybrid WOLEDs. By dint of this multifunctional material, the guest concentration can be as high as 1.5%. The single-EML WOLED can exhibit a maximum total PE of 65.3 lm/W, which is the highest efficiency among single-EML hybrid WOLEDs with high guest concentrations (＞1%), to the best of our knowledge. Besides, the device exhibits low operational voltages. A low turn-on voltage of 2.4 V is obtained, which is the lowest value among hybrid WOLEDs. At 100 cd/m^2 and 1000 cd/m^2, the voltage is 2.75 V and 3.45 V, respectively. Moreover, a peak color rendering index (CRI) of 77 is obtained for this two-color WOLED. Unlike the working mechanisms in previous single-EML hybrid WOLEDs with low guest concentrations, devices comprising high concentrations exhibit more sophisticated engineering, in which the device smartly allow the utilization of both the fluorescence from the NPB host itself (deep-blue emission), and the complementary phosphorescence from the triplet Ir(dmppy)$_2$(dpp) guest (orange emission) by incomplete Förster energy transfer, Dexter energy transfer as well as direct exciton formation on this guest. To further exploit this unique host-guest system, we have incorporated it into a dual-EML hybrid WOLED. For this WOLED, a maximum efficiency of 17.2 lm/W and 10.2 lm/W at 1000 cd/m^2 (3.85 V) with an ultrahigh CRI of 93 are achieved, which provides a new opportunity to accomplish the simplified structure/ low voltage/ high efficiency/ ultrahigh CRI trade-off.

2, Experimental

As depicted in figure 1, the configuration of the single-EML hybrid WOLED (W11) is ITO/HAT-CN (100 nm)/NPB (20 nm)/ NPB: Ir(dmppy)$_2$(dpp) (35 nm,

1.5%)/ $Bepp_2$ (35 nm)/LiF (1 nm)/Al (200 nm), where ITO is indium tin oxide, HAT-CN is 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile [as a hole injection layer (HIL)], $Ir(dmppy)_2(dpp)$ is Bis(2-phenyl-4,5-dimethylpyridinato)[2-(biphenyl-3-yl)pyridinato] iridium(III) (as an orange emitter), $Bepp_2$ is an electron transport layer (ETL). All material layers were thermally deposited without breaking the vacuum at a base pressure of 2×10^{-7} Torr. In the deposition of the doping layers, deposition rates of both host and guest were controlled by their correspondingly independent quartz crystal oscillators. The devices were encapsulated immediately after preparation under a nitrogen atmosphere using epoxy glue and glass lids. The electroluminescent (EL) spectra, Commission International de I'Eclairage (CIE) color coordinates and CRI of packaged devices were obtained by a Konica Minolta CS2000 spectra system. The emission area of the devices is 3×3 mm^2 as defined by the overlapping area of the anode and cathode. The luminance (L)-current density (J)-voltage (V) characteristics were recorded simultaneously, using a computer-controlled source meter (Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. All the measurements were carried out at room temperature under ambient conditions.

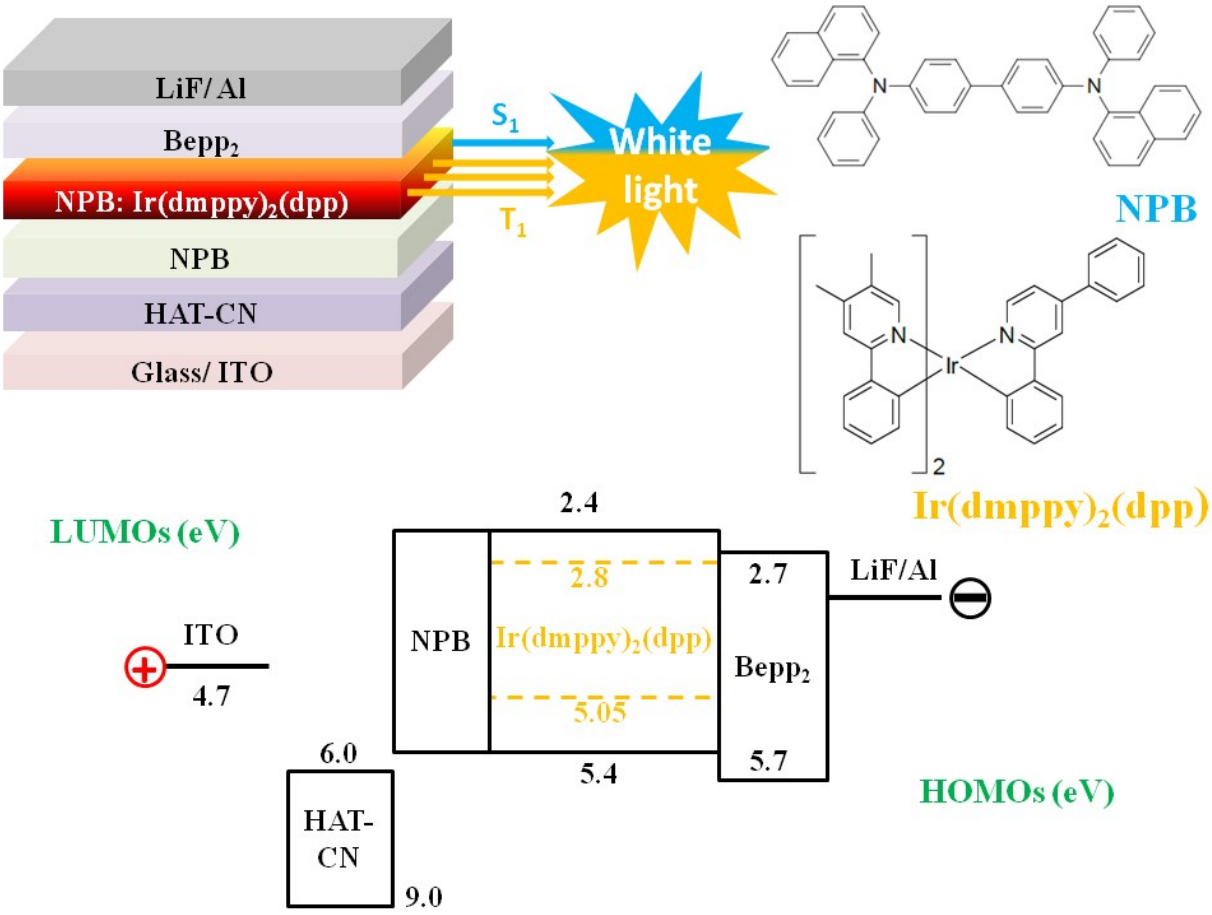


Fig. 1. Top: The schematic layer structure of processed WOLEDs and the chemical structure of emissive materials. Bottom: Proposed energy-level diagram of the WOLED (W11).

3. Results and discussions

3.1 Single-EML hybrid WOLEDs

3.1.1 Devices design strategies

To achieve high-performance WOLEDs, several design strategies are employed. First, the greatest motivation of single-EML hybrid WOLEDs is the introduction of an appropriate material functioning as both the blue F emitter and the host of complementary color phosphor. If the material has a low T_1, triplet excitons can transfer from the phosphor to fluorophor, resulting in the non-radiative decay. That is, triplets in the phosphor can be quenched by the fluorophor, leading to no white

emission. Therefore, the introduction of blue F emitters with relatively high triplet energies is a key feature of designing this kind of WOLEDs, in which triplets in the blue fluorophor can be harvested by the complementary color phosphor. So far, it is still a big challenge to obtain molecular systems fulfilling these requirements.[26] To alleviate this difficulty, herein, we, for the first time, have demonstrated that NPB can be used as both the blue F emitter and the host of orange P emitter, because: i) The T_1 of NPB is 2.3 eV,[27] high enough to satisfy the demand of the orange emitter $Ir(dmppy)_2(dpp)$ (T_1＜2.3 eV),[28] which can prevent reverse energy transfer from guests to the host and confine triplets in the whole EML, effectively consuming the electrically generated triplets contributing to emission.[3] ii) NPB can exhibit a deep blue color with a peak emission of ~440 nm.[29] Therefore, white emission can be expected by simultaneous host/guest emission in single-EML WOLEDs.

Next, NPB is used as the hole transport layer (HTL), which can i) give an effective hole transport due to its high hole mobility (4×10^{-4} cm^2/V s).[27] ii) confine the triplets in the EML owing to the high T_1, ensuring the high efficiency.[13] iii) eliminate the heterojunction between the HTL and EML, enhancing the device performance (i.e., higher efficiency, lower voltage and longer lifetime).[3] iv) simplify the device structure, since only three organic layers are used in W1 to generate the white emission, which is even more simplified than the previous single-EML hybrid WOLEDs.[18,19] It is noteworthy that 4P-NPD may also satisfy these demands and the guest concentration in 4P-NPD based single-EML WOLED can be high (＞1%),[22,24,25] however, the recorded efficiency is only 10.5 lm/W,[22] further indicating the advantage of the

exploitation of NPB in our device.

Finally, $Bepp_2$ is used as an excellent ETL, which can i) give an effective electron injection due to the high electron mobility (10^{-4} cm^2/V s),[10] and thus an improved carrier balance. ii) effectively confine triplets in the EML due to the high T_1 of 2.6 eV, [10]. iii) block holes transport due to the deep highest occupied molecular orbital (HOMO) of 5.7 eV.[10] Therefore, a carrier- and exciton-confining structure is formed via the combination of NPB and $Bepp_2$, enhancing the performance.[11]

Taking the above factors into account, we have developed a high-performance single-EML hybrid WOLED with a guest concentration as high as 1.5%.

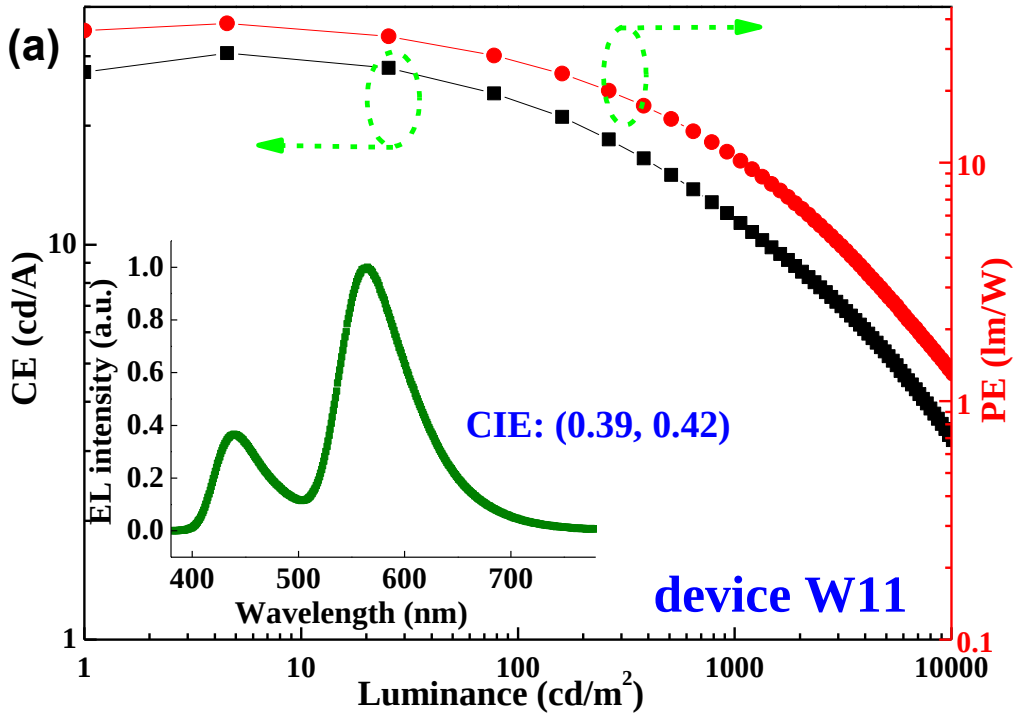
3.1.2 Performance of the single-EML hybrid WOLED

The forward-viewing CE as well as PE of W11 in dependence of the luminance is shown in figure 2a. The maximum forward-viewing CE and PE of the device are 30.5 cd/A and 38.4 lm/W at 5 cd/m^2, respectively. As illumination sources are typically characterized by their total emitted power,[4,17-19] the maximum total PE of W11 is 65.3 lm/W, representing the highest efficiency among single-EML hybrid WOLEDs with high guest concentrations (＞1%), indicating a significant step towards the real commercialization. In fact, the efficiency is also comparable to these best single-EML WOLEDs,[17-19] however, our device shows more competitive edge since it can effectively solve the problem of requiring a very low guest amount (i.e., 0.03%, 0.1% and 0.2%) to attain white light. At 100 cd/m^2, the efficiency is 45.9 lm/W. At 1000 cd/m^2, the efficiency is 18.2 lm/W with the CIE coordinates of (0.39, 0.42), as shown in figure 2a inset. It is noted that the efficiency roll-off is somewhat serious and the

color is not stable enough at high luminances (see the supplementary figure S1), which are common phenomena in single-EML hybrid WOLEDs, even for those using bipolar hosts.[18,19] The phenomena in W11 may be explained as follows. Owing to the existed energy barriers between NPB and $Bepp_2$ together with the fact that NPB is a hole-type material and $Bepp_2$ is an electron-type material, the main exciton generation zone is located at the NPB/ $Bepp_2$ interface, which is not wide enough.[13] As a result, the triplet-triplet annihilation (TTA) and triplet-polaron quenching (TPQ) easily occur, which are detrimental to the efficiency roll-off and color-stability.[30] Besides, the incomplete triplet utilization is also responsible for the phenomena.[19] Moreover, since both holes and electrons can be trapped by $Ir(dmppy)_2(dpp)$ (will be demonstrated later), a strong orange emission is observed at low luminances.[3] However, the trapped charges can be saturated with the increasing luminance/ driving voltage, indicating that more untrapped holes can be injected into the HOMO of NPB molecules and more untrapped electrons can be transported along the lowest unoccupied molecular orbital (LUMO) of NPB molecules, resulting in more excitons being formed on NPB molecules to give off more blue emission relative to the orange emission at high luminances. Thus, W1 exhibits a blue-shifted color with the increasing luminance.

Remarkably, W11 exhibits rather low voltages, as shown in figure 2b. For example, the turn-on voltage is only 2.4 V (for a luminance of 1 cd/m^2), which is the lowest value among hybrid WOLEDs. At 100 cd/m^2 and 1000 cd/m^2, the voltage is 2.75 V and 3.45 V, respectively. Since it is still a challenge for WOLEDs to achieve low driving voltages for practical use (e.g., $<$3 V for onset and $<$4 V at 100 cd/m^2

for portable display), it is obviously noted that our device can effectively alleviate this difficulty.[31] Apparently, it is important to note that much higher efficiency and lower voltage can be expected if a p-i-n structure were used.[22] Moreover, a CRI of 77 can be obtained at 10000 cd/m^2 (see the supplementary figure S2), which is one of the highest values among two-color WOLEDs.[21,32]



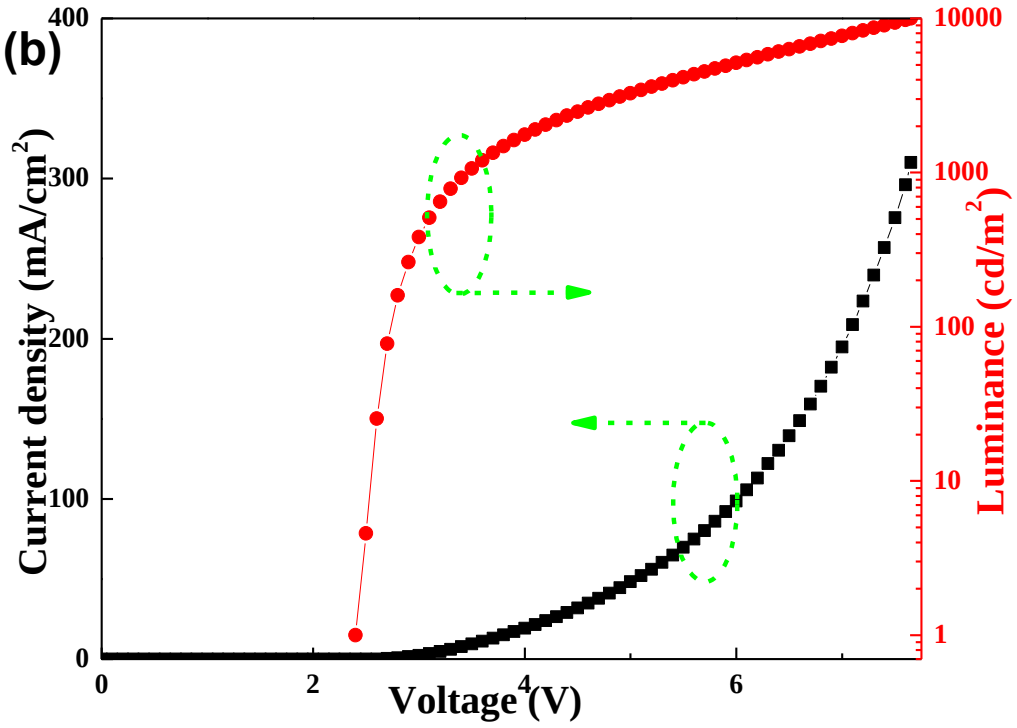


Fig. 2. a) Forward-viewing current and power efficiencies as a function of luminance for W11. Inset: EL spectrum at a luminance of 1000 cd/m^2. b) J-V-L curves of W11.

3.1.3 Working mechanisms of the single-EML hybrid WOLED

To gain an insight of the high performance of W11, it is necessary to unveil the working mechanism of this unique host-guest system. In OLEDs, host-guest energy transfer and direct charge trapping on the guest are two primary mechanisms of exciton formation and emission.[3,33] For host-guest systems with low concentrations, excitons would be mainly located at the blue fluorophor molecules for their preponderant amount.[18] As shown in figure 3a, since diffusion lengths of singlets (R_S) and triplets (R_T) are about 3 nm and 100 nm,[4] respectively, Zhang and Lee et al. proposed the concept that some fluorophor molecules find no P guest molecule within their vicinity of 3 nm, while P molecules exist within 100 nm, leading to the fact that singlets on the fluorophors remain for the blue emission and triplets transfer their

energy to the guests for P emission.[18] Therefore, the Förster energy transfer between fluorophors and phosphors is considered to be prohibited in this kind of device,[18] as shown in figure 3b.

However, the above concept cannot be simply applied to our device, since the concentration of W11 is as high as 1.5%, which is 15 times larger than Zhang and Lee's device.[18] In the case of W11, a portion of fluorophor molecules inevitably meet P guest molecule within their vicinity of 3 nm due to the high concentration, resulting in the fact that the Förster energy transfer from fluorophors to phosphors is possible, as shown in figure 3c. Such energy transfer is not wasted, because the orange emission will be enhanced via the following intersystem crossing (ISC) process, although it can decrease the blue emission. Besides, $Ir(dmppy)_2(dpp)$ can harvest the triplet excitons generated in the NPB host via the Dexter energy transfer, giving off the orange emission. On the other hand, the fluorophor molecules which cannot meet P guest molecule within their vicinity of 3 nm would guarantee the singlet excitons on the fluorophors for the blue emission.

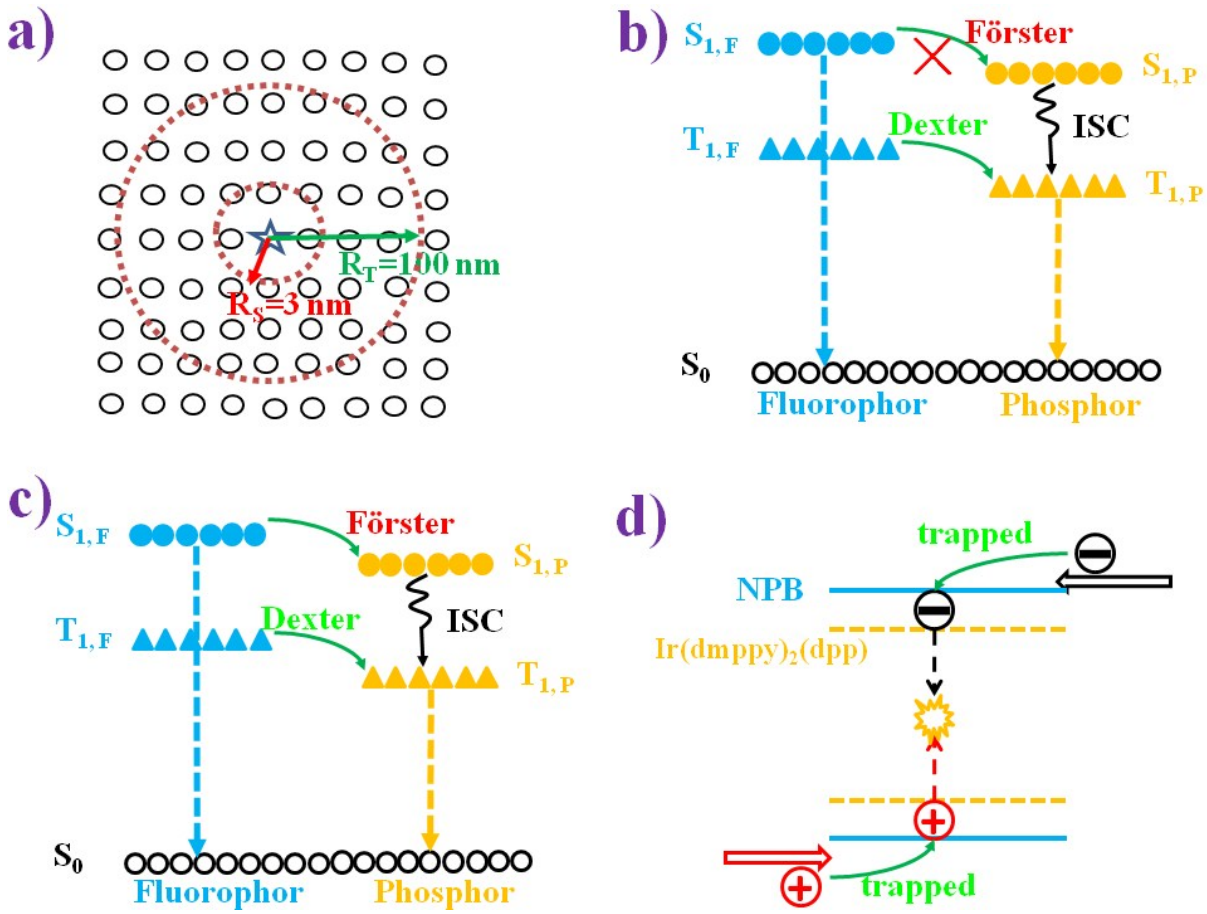


Fig. 3. Schematic diagrams of the working mechanisms of the single-EML hybrid WOLEDs. a) The diffusion lengths of singlets and triplets. b) Excitons decay in single-EML hybrid WOLEDs with low concentrations, S_1 represents the singlet energy. c) Excitons decay in W11 with high concentrations. d) The direct charge trapping process in W11.

In addition to the energy transfer from the NPB host to the Ir(dmppy)$_2$(dpp) guest to furnish orange emission, direct charge trapping on the guest is possible to occur in W11, although this emission mechanism is not considered in the host-guest system with low concentration.[18] This is because Ir-complex guests usually have a strong hole/electron-capture ability when high concentrations are used.[31] From figure 1, it is seen that the HOMO of Ir(dmppy)$_2$(dpp) (5.05 eV)[28] is higher than that of NPB (5.4 eV),[27] while the LUMO of Ir(dmppy)$_2$(dpp) (2.8 eV) [28] is lower than that of NPB (2.4

eV),[27] indicating that both holes and electrons can be nicely trapped by $Ir(dmppy)_2(dpp)$ in this host-guest system.[3] In other words, direct exciton formation following charge trapping contributes to $Ir(dmppy)_2(dpp)$ emission, as shown in figure 3d. Since direct exciton formation on the guest phosphor can eliminate the need to electrically excite the host, while allowing for efficient carrier collection, exciton formation, and recombination at the guest molecular sites, it is effective to enhance the efficiency.[11] Besides, since charges can be trapped by $Ir(dmppy)_2(dpp)$, both the hole leakage into the ETL and electron leakage into the HTL are decreased, further enhancing the efficiency.[3] Therefore, it is reasonable that W11 with a high efficiency is achieved.

To further prove the above analysis, we have checked the J-V characteristics at various concentrations of $Ir(dmppy)_2(dpp)$, since the two primary emission mechanisms (i.e., energy transfer and direct charge trapping) can be distinguished from the dependence of drive voltage on guest concentration.[3] The configuration of single-EML devices is ITO/HAT-CN (100 nm)/NPB (20 nm)/ NPB: $Ir(dmppy)_2(dpp)$ (35 nm, 0.5%, 1.5%, 5% or 10%)/ $Bepp_2$ (35 nm)/LiF (1 nm)/Al (200 nm), where 0.5% denotes device W12, 5% denotes device W13 and 10% denotes device W14. As shown in figure 4, the voltage is shifted towards higher value as the concentration of $Ir(dmppy)_2(dpp)$ increases, indicating that the direct charge trapping on $Ir(dmppy)_2(dpp)$ molecules is also the mechanism for exciton formation and emission in W11.[34]

In a word, W11 can smartly allow the utilization of both the fluorescence from

the NPB host itself (deep-blue emission), and the complementary phosphorescence from the triplet Ir(dmppy)$_2$(dpp) guest (orange emission) by incomplete Förster energy transfer, Dexter energy transfer as well as direct exciton formation on this guest, leading to the warm-white emission.

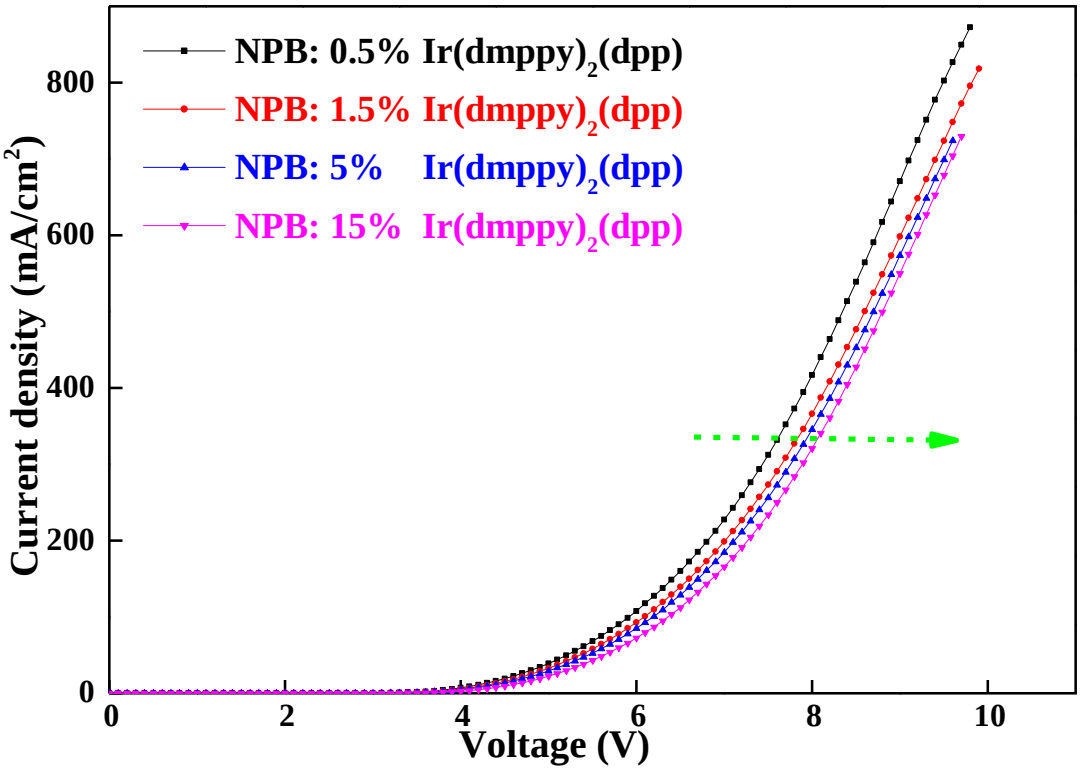
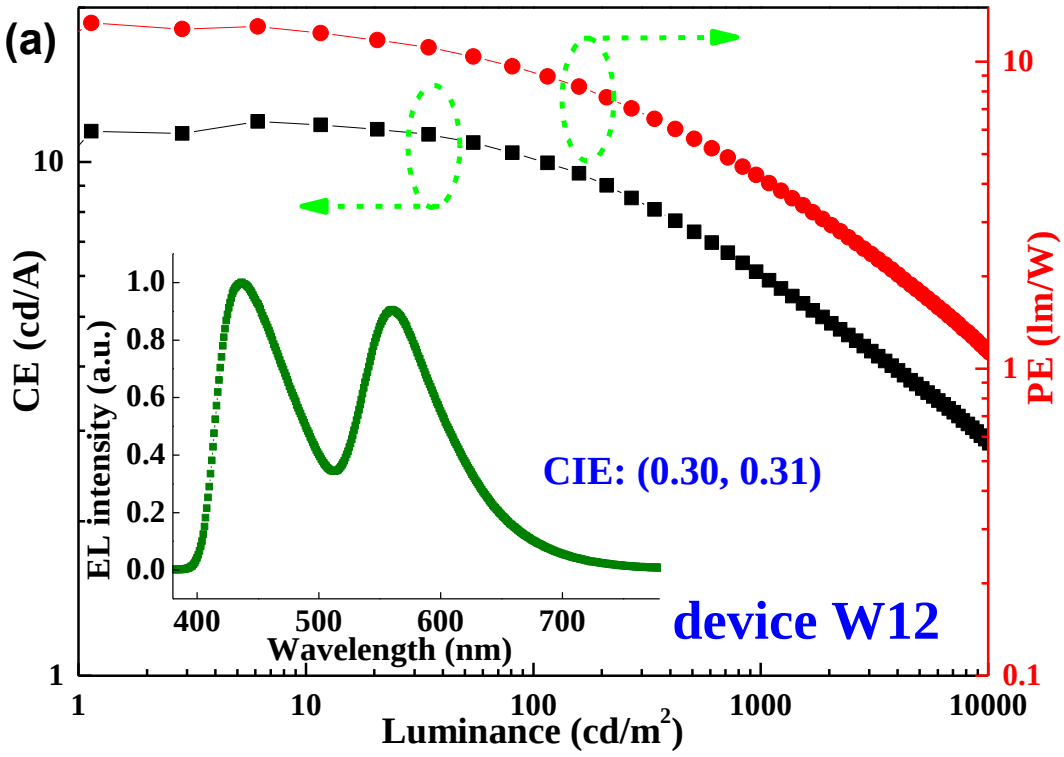


Fig. 4. J-V characteristic of the single-EML devices.

To further understand this efficient host-guest system, we have measured the performance of W12 and W13. As shown in figure 5, low forward-viewing efficiencies of 12.0 cd/A (13.4 lm/W) and high efficiencies of 49.3 cd/A (55.3 lm/W) are obtained in W12 and W13, respectively. When the concentration is as low as 0.5%, the generated triplets cannot be fully harvested by Ir(dmppy)$_2$(dpp), resulting in the low efficiencies in W12. This is also the reason why W12 exhibits a relatively balanced blue and orange emission, as shown in figure 5a inset. On the other hand, as the concentration is very high (5%), most of the singlet and triplet excitons are used to

produce orange emission since both S_1 and T_1 of NPB are much higher than those of $Ir(dmppy)_2(dpp)$, leading to the high efficiencies in W13. This is also the reason why W13 exhibits almost no blue emission but strong orange emission, as shown in figure 5b inset. Besides, as shown in figure S1, the color variations of W12, W11and W13 are (0.10, 0.13), (0.06, 0.08) and (0.00, 0.01), respectively, indicating that devices with higher concentrations exhibit more stable colors. This is because more charges are trapped with the guest concentrations increasing, which is beneficial to the color-stability, further demonstrating the direct charge trapping emission mechanism in this host-guest system.[3,21]



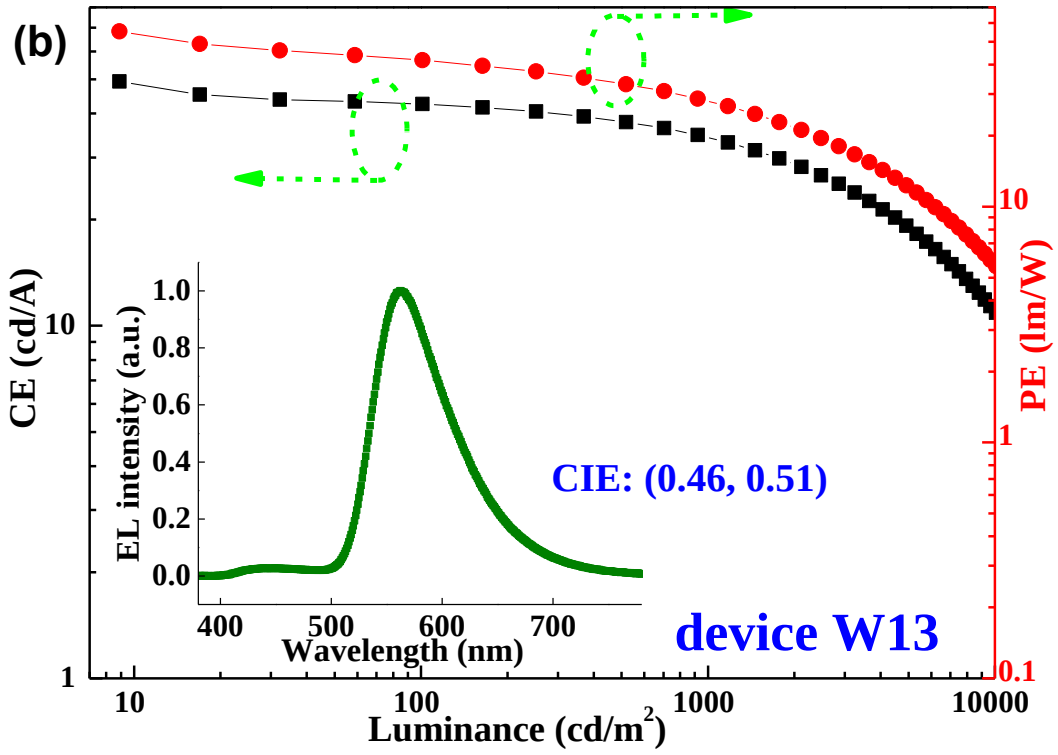


Fig. 5. a) Forward-viewing efficiencies of W12. Inset: EL spectrum at 1000 cd/m^2. b) Forward-viewing efficiencies of W13. Inset: EL spectrum at 1000 cd/m^2.

3.2 The dual-EML hybrid WOLEDs with ultrahigh CRI

It is well-known that host-guest systems are significant to the EMLs, regardless of the monochromatic OLEDs or WOLEDs. Therefore, galvanized by the excellent performance of W11, particularly for the high CRI (77), and to further take advantage of this unique host-guest system, we incorporate it into a dual-EML hybrid WOLED structure (W2). The configuration of W2 is ITO/HAT-CN (100 nm)/NPB (20 nm)/ NPB: Ir(dmppy)$_2$(dpp) (20 nm, 1.5%)/ Bepp$_2$: Ir(ppy)$_3$: Ir(piq)$_3$ (15 nm,1: 6%: 1.3%)/ Bepp$_2$ (35 nm)/LiF (1 nm)/Al (200 nm), where tris(2-phenylpyridinne)iridium(III) [Ir(ppy)$_3$] and tris(1-phenylisoquinolinolato-C^2,N) iridium(III) [Ir(piq)$_3$] were codoped into Bepp$_2$ as the green and red emitter, respectively. The detailed chemical structures of the two emitters can be seen in the supplementary figure S3. Since the emission

peak of NPB, $Ir(ppy)_3$, $Ir(dmppy)_2(dpp)$ and $Ir(piq)_3$ are around 440 nm, 510 nm,[22] 560 nm and 620 nm,[35] respectively, exhibiting broad emission spectra covering about most of the visible spectrum, an ultrahigh CRI (＞90) can be expected.

As displayed in figure 6a, the maximum total efficiency of W2 is 17.2 lm/W at 26 cd/m^2, which decreases to 16.3 lm/W and 10.2 lm/W at 100 cd/m^2 and 1000 cd/m^2, respectively. Remarkably, an ultrahigh CRI of 93 is obtained at 1000 cd/m^2, as shown in figure 6a inset. A CIE coordinates of (0.51, 0.44) and a low color-correlated temperature of 2360 K are also obtained at 1000 cd/m^2, which can effectively satisfy the demand of physiologically-friendly indoor illumination.[36] Besides, W2 exhibits low voltages. The turn-on voltage is only 2.55 V, 3.05 V for 100 cd/m^2 and 3.85 V for 1000 cd/m^2, as shown in figure 6b.

Previously, a few efforts have been taken to attempt WOLEDs with ultrahigh CRIs (＞90), such as using a single phosphorescence dopant of heteroleptic Pt-complex with an excimer/aggregate emission,[25] studying the effect of the active layer thickness,[37] combining yellow/ green fluorescence emission located between the monomer and excimer emission,[38] fabricating a five-spectrum device with double white EMLs and incorporating a hole-modulation layer to regulate the intensities of emitters,[39] and so forth. However, these devices usually exhibit: i) complicated structures, since more than two EMLs, additional ILs or excimer emissions are needed. ii) high operational voltages, since virtually no WOLED with ultrahigh CRI exhibits a turn-on voltage ＜3 V and ＜4 V at 1000 cd/m^2. iii) low efficiencies, since the efficiency at 100 cd/m^2 is usually below 10 lm/W, as shown in table 1. Herein, we

demonstrate a simplified but effective WOLED to overcome these problems, providing a new opportunity to accomplish the simplified structure/ low voltage/ high efficiency/ ultrahigh CRI trade-off.

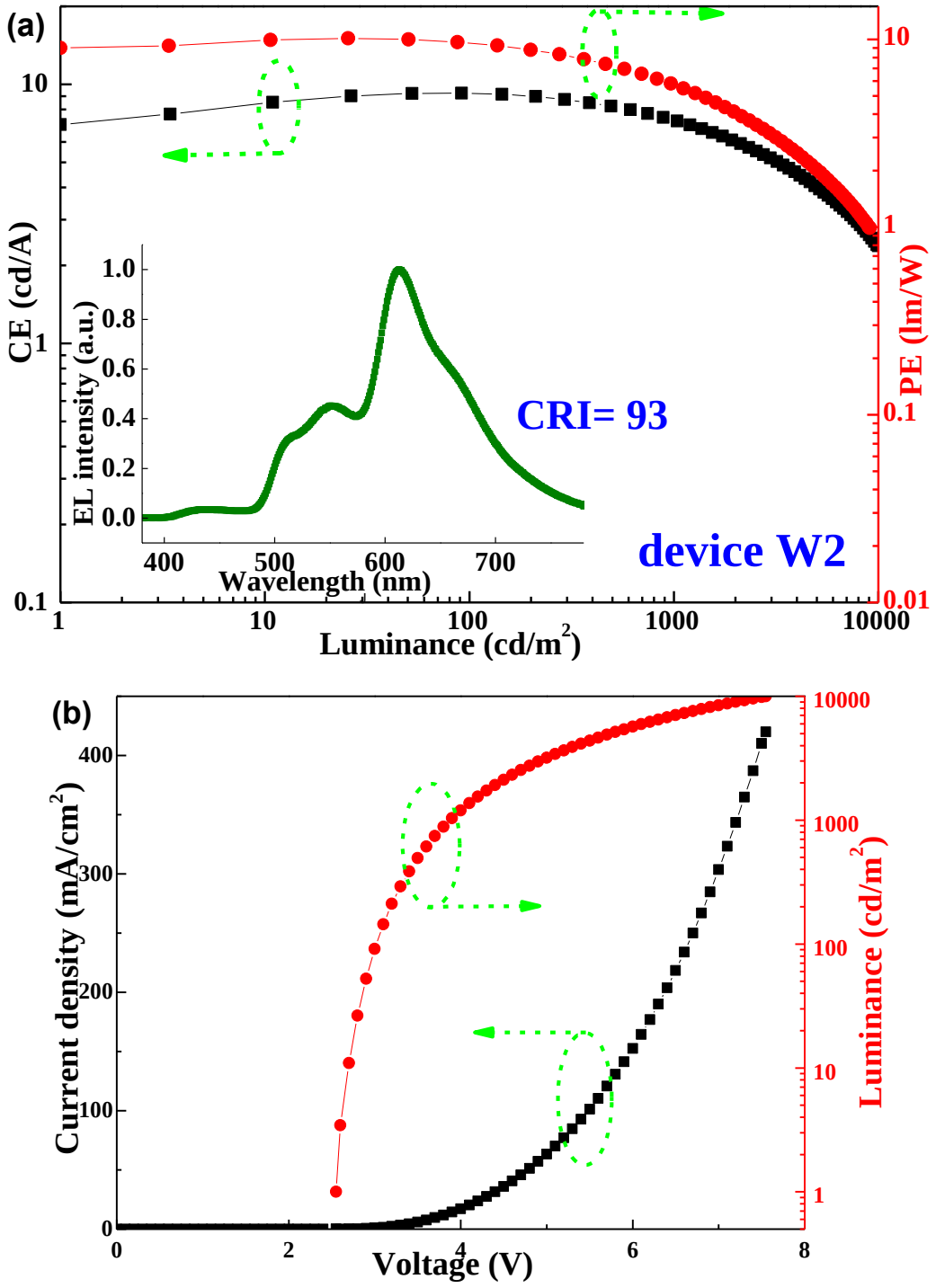


Fig. 6. a) Forward-viewing efficiencies as a function of luminance for W2. Inset: EL spectrum at 1000 cd/m^2. b) J-V-L curves of W2.

Table 1

Summary of representative performances of WOLEDs with ultrahigh CRIs.

Device	$V_{on}/_{100}/_{1000}$ [a] (V)	$PE_{max/100/1000}$ [b] (lm/W)	CRI [c]	CIE [d]
Ref. [25] (2 EMLs & excimer/aggregate emission)	4/-/-	6.8/-/-	91	(0.33, 0.31)
Ref. [37] (5 EMLs & 2 ILs)	3.5/~4.2/~5	15.2/-/-	97	(0.51, 0.42)
Ref. [38] (2 EMLs, 1 IL & excimer)	-	10.6/-/-	94	(0.42, 0.43)
Ref. [39] (2 EMLs & 1IL)	-	-/8.3/5.2	98	(0.34, 0.36)
Ref. [40] (4 EMLs & 1 IL)	3.1/4.05/4.6	~8.2/-/8.19	98	(0.45, 0.40)
Ref. [41] (3 EMLs & I IL)	-	-/5.5/-	91.2	(0.32, 0.34)
Ref. [42] (3 EMLs)	-/~4.5/~6.2	-	93	(0.34, 0.34)
Ref. [43] (3 EMLs)	-/~5/8.06	3.16/-/3.08	91	(0.33, 0.33)
Ref. [44] (solution processed)	4/-/-	0.25/-/-	92.2	(0.28, 0.30)
Ref. [45] (solution processed)	3/~3.3/~4.1	8.9/-/-	91	(0.36, 0.37)
This work (2 EMLs)	2.55/3.05/3.85	17.2/16.3/10.2	93	(0.51, 0.44)

[a] The turn-on voltage, voltage at 100 cd/m^2 and voltage at 1000 cd/m^2. [b] Maximum PE, PE at 100 cd/m^2 and PE at 1000 cd/m^2. [c] Maximum CRI. [d] CIE coordinates at the maximum CRI.

Finally, it is deserved to note the efficiency of W2 is lower than that of W11, despite W2 exhibits a higher efficiency compared with WOLEDs having ultrahigh CRIs, which can be explained as follows. The exciton generation zone of W2 is located at the middle of EML (i.e., the NPB/ $Bepp_2$ interface) due to the existed

energy barriers together with the fact that NPB is hole-type material and $Bepp_2$ is electron-type material, resulting in both singlet and triplet excitons being formed at this interface with a ratio of 1:3.[3] Since the T_1 of NPB is 2.3 eV,[27] which is lower than that of $Bepp_2$ (2.6 eV) and $Ir(ppy)_3$ (2.4 eV),[22] triplets with high triplet energies can be easily quenched by NPB, leading to the energy loss.[3] As a result, a low efficiency is obtained. To confirm this effect, we have inserted 3.5 nm 4,4',4''-tri(9-carbazoyl) triphenylamine (TCTA): $Bepp_2$ as the IL to eliminate the Dexter energy transfer between the two EMLs, since the Dexter energy transfer is significant within a range of 1-2 nm.[22] As expected, the efficiencies of devices with ILs are dramatically enhanced (see the supplementary figure S4). Besides, despite $Bepp_2$ can be an efficient blue emitter,[17] it should be pointed out that there is no emission originating from $Bepp_2$ in W2, which is attributed to the fact that 6% $Ir(ppy)_3$ can effectively harvested all generated excitons in this host-guest system [$Bepp_2$-$Ir(ppy)_3$)] (see the supplementary figure S5). In other words, the ultrahigh CRI of 93 is obtained from four emitters instead of five emitters (i.e., NPB, $Ir(ppy)_3$, $Ir(dmppy)_2(dpp)$ and $Ir(piq)_3$). Moreover, since the T_1 of red emitter $Ir(piq)_3$ is only 2.0 eV, which is the lowest among these materials (i.e., $Bepp_2$, NPB, $Ir(ppy)_3$, $Ir(dmppy)_2(dpp)$ and $Ir(piq)_3$), the triplet excitons can be easily harvested by $Ir(piq)_3$.[3] This is the reason why W2 exhibits a strong red emission, although the concentration of $Ir(piq)_3$ is the lowest.

4. Conclusion

In summary, we have developed a very efficient host-guest system, in which

NPB is the first time demonstrated to be used as both the host and blue emitter in single-EML WOLEDs. By virtue of this versatile material, the guest concentration is as high as 1.5%, effectively avoiding the difficulty in the control and reproduction process. Remarkably, the single-EML WOLED can exhibit a maximum efficiency of 65.3 lm/W, which is much higher than previous single-EML hybrid WOLEDs with high guest concentrations (＞1%), indicating a significant step towards the real commercialization. Besides, the device exhibits low voltages. The turn-on voltage, voltage and voltage for 1000 cd/m^2 are 2.4 V and 3.45 V, respectively. Moreover, a CRI of 77 is obtained for this two-color device. The working mechanism of the device comprising high concentrations are discussed, in which the device smartly allow the utilization of both the fluorescence from the NPB host itself, and the complementary phosphorescence from the triplet Ir(dmppy)$_2$(dpp) guest by incomplete Förster energy transfer, Dexter energy transfer as well as direct exciton formation on this guest. Furthermore, we have incorporated this unique host-guest system into a dual-EML WOLED. A maximum efficiency of 17.2 lm/W and 10.2 lm/W at 1000 cd/m^2 (3.85 V) with an ultrahigh CRI of 93 are achieved, providing a new opportunity to accomplish the simplified structure/ low voltage/ high efficiency/ ultrahigh CRI trade-off.

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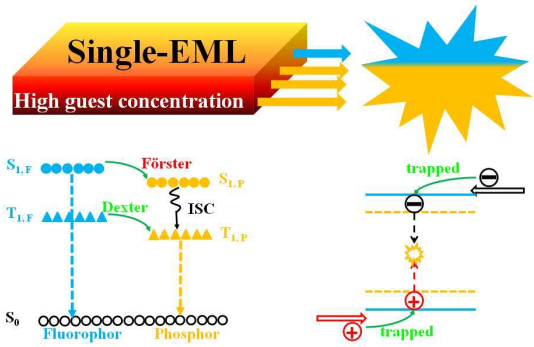

A host-guest system comprising high guest concentration (1.5%) is used to develop the high-performance single-EML hybrid WOLED and dual-EML WOLED.