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Abstract: High performing tetra-chromatic hybrid white organic light-emitting diode (WOLED), with a fluorescent blue emitting layer (EML) of Bis[2-(2-hydroxyphenyl)-pyridine]beryllium (Bepp$_2$) sandwiched between a pair of phosphors-doped hole transporting layer (HTL) and electron transporting layer (ETL), was developed. This was achieved by controlling the location of the green phosphor doped in HTL and yellow phosphor doped in ETL at ~1 nm away from the HTL/EML and EML/ETL interfaces, and incorporating an ultrathin red phosphorescence layer (< 0.1 nm) in the center of Bepp$_2$ EML. The resulting hybrid WOLED realizes good warm white emission, showing stable electroluminescence spectra with a maximum color rendering index (CRI) of 94 and a low correlated color temperature of 2440−2468 K over a wide voltage range from 5−9 V. Meanwhile, this WOLED also achieves high device efficiency, the maximum current efficiency, power efficiency, and external quantum efficiency up to 34.15 cd/A, 29.51 lm/W, and 17.71 %, respectively. Such high performance is realized through the precise manipulation and effective exploitation of singlet and triplet excitons via novel device design. Moreover, the proposed WOLED also removes the additional interlayers between the fluorescent and phosphorescent emitting regions that are commonly employed in the conventional hybrid WOLEDs, inducing a simplified device structure with reduced heterojunction interfaces, which is very beneficial for promoting the commercial development of WOLEDs.

1. Introduction

White organic light-emitting diodes (WOLEDs), exhibiting the unique features such as fabrication flexibility, ultra-thin format, light-weight, and high luminous efficiency,

etc, have attracted considerable attentions because of their potential applications in solid-state lighting and flat panel displays fields.[1-3] The hybrid WOLEDs, combining emissions from short-wavelength blue fluorophore and long-wavelength complementary color phosphors, take the advantages of the excellent stability of blue fluorescent emitter and the high efficiency of long-wavelength phosphorescent emitters,[4-10] which are considered as a feasible method for realizing high efficiency and long-life WOLEDs. Generally, two types of typical device structures, single-emissive layer (single-EML) structure[5,11-14] and multi-emissive layers (multi-EMLs) structure [1,15-17], are employed to develop hybrid WOLEDs.

For single-EML hybrid WOLEDs, the devices are typically fabricated by simultaneous doping of blue fluorophore and complementary color phosphors in a common host.[11] The hybrid WOLEDs are also fabricated by doping long-wavelength phosphors in a blue fluorophore host.[5,12-14] To realize ideal white emission, the concentrations of different fluorescent and phosphorescent emitters need to be precisely controlled.[18] This is because a slight change in the dopant concentration would cause a pronounced variation in the electroluminescence (EL) spectra due to the undesired energy transfer processes occurred amongst the different doped emitters,[4-5,11,17] that is unfavorable for the efficient operation of WOLEDs. Compared with the single-EML hybrid WOLEDs, the multi-EMLs hybrid WOLEDs allow flexible manipulation of each EML,[7-9,19] and the incorporation of an interlayer between the phosphorescent and fluorescent dopants layers separates singlet and triplet excitons in their respective emitting zones,[4,7-11,20] allowing a completely electricity generated excitons harvest for efficient light emission.

Thus, the multi-EMLs strategy seems to be a reliable method for attaining high

performance hybrid WOLEDs.[7,21] However, the use of the interlayer introduces additional heterojunction interfaces, leading to the adverse effect on the operation and performance of WOLEDs. These include i) inefficient carriers injection/transport and accumulation of space charges at these heterojunction interfaces,[15] and ii) an inevitable increase in the possibility of exciplex formation, which impairs the device efficiencies of hybrid WOLEDs.[4,19] Based on the above causes, although a lot of research work based on multi-EMLs hybrid WOLEDs has been carried out by some groups, the external quantum efficiencies (EQEs) for these hybrid WOLEDs with additional interlayers are typically below the theoretically maximum EQE of 20%.[1,4,15,19,21-25] In addition, the insertion of extra interlayers is also unfavorable for applications in commercial products at a low cost.[19] Thus, if the singlet and triplet excitons in multi-EMLs hybrid WOLEDs without additional interlayers can be manipulated and utilized well for light emission, it would not only boost device efficiency, but also induce a simplified device structure.

Recently, multi-EMLs hybrid WOLEDs without using such interlayer, e.g., doping a blue fluorescent emitter in mixed hosts comprising of electron- and hole-transporting materials, were proposed, demonstrating an EQE over the range from 16.5−19.0% under forward-viewing direction [19]. However, triplet energy in the blue fluorophore layers cannot be fully utilized due to the excitons back-transferred from the green phosphor to the blue fluorophore, caused by the low triplet energy level of the blue fluorophore. Lee and co-workers employed blue exciplex-forming co-host as a fluorescent EML, combined with a yellow phosphorescent EML, developing a series of interlayer-free multi-EMLs hybrid WOLEDs.[26] Although the high EQE of 12.7−19.3% for these hybrid WOLEDs in forward-viewing direction is obtained, the devices have a relatively low color rendering index (CRI) of 67−77,

which are obviously lower than the threshold value (80) for WOLEDs used in lighting. In addition, as illumination sources, the EL spectra of WOLEDs are required to be stable at a wide voltage range. So, the color stability is as important as other performance parameters such as brightness, device efficiency, and CRI.[27-31] Although many groups have reported the high device efficiencies for hybrid WOLEDs, the problems, including low CRI and poor color stability, still exist.[2,5-6,32] Specially, for a multi-EMLs hybrid WOLED without additional interlayers, realizing excellent color stability, high CRI, and high efficiency simultaneously, remains an open challenge.

In this work, a fluorescent Bis[2-(2-hydroxyphenyl)-pyridine]beryllium (Bepp$_2$) with a high triplet energy level (2.60 eV)[33] was used as blue EML in the hole transporting layer (HTL)/EML/electron transporting layer (ETL) type hybrid WOLEDs. 4,4′,4″-tris(N-carbazolyl) triphenylamine (TCTA) HTL, with an energy bandgap of 3.40 eV and a triplet energy level of 2.80 eV,[34] and 1,3,5-tris(phenyl-2 -benzimidazolyl)benzene (TPBi) ETL, having an energy bandgap of 4.00 eV and a triplet energy level of 2.70 eV,[35] were used. By doping green and yellow phosphorescent emitters in HTL and EML, respectively, and incorporating an ultrathin red phosphorescent layer in the middle of Bepp$_2$ EML, a high performance tetra-chromatic multi-EMLs hybrid WOLED without additional interlayer was demonstrated. The hybrid WOLED realizes good warm white emission, showing stable EL spectra with a maximum CRI of 94 and a low correlated color temperature (CCT) of 2440−2468 K under a broad voltage range from 5−9 V. In addition, this white device also achieves the maximum current efficiency (CE), power efficiency (PE), and EQE of 34.15 cd/A, 29.51 lm/W, and 17.71 %, respectively.

2. Experimental

Materials information

In this work, Bepp₂, exhibiting a high triplet energy level (2.60 eV) and a high fluorescent quantum yield (0.80), was employed as an efficient blue fluorophor. Three different phosphorescent dyes tris(2-phenylpyridine)iridium(III)) (Ir(ppy)₃) (green), bis(2-(3-trifluoromethyl-4-fluorophenyl)-4-methylquinolyl)(acetylacetonate)iridium(III) (Ir(ffpmq)₂(acac)) (yellow), bis(1-phenylisoquinoline)(acetylacetonate)iridium(III) (Ir(piq)₂(acac)) (red), were used in conjunction with Bepp₂ for producing white light emission. Apart from Ir(ffpmq)₂(acac) which was synthesized in house, other active organic functional materials used in the hybrid WOLEDs were purchased through commercial sources and used without further purification. The chemical structures of all organic materials used in the hybrid WOLEDs are displayed in **Fig.1**.
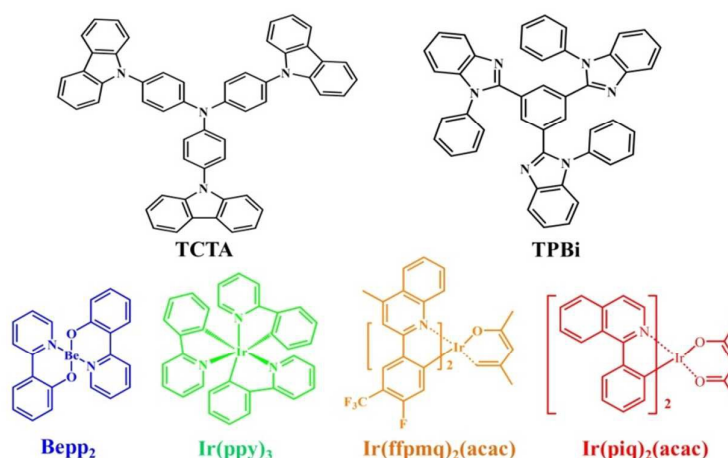

Fig. 1 The chemical structures of the functional organic materials used in the hybrid WOLEDs.

Device fabrication and characterization

The pre-patterned indium tin oxide (ITO)/glass substrates, with a sheet resistance of 15 Ω/square, were used for fabrication of WOLEDs. They were cleaned sequentially by detergent, acetone, isopropyl alcohol and deionized water for 20 minutes each before use. The surface of the ITO/glass substrates were exposed to the UV ozone for

10 minutes to remove the residual chemicals left on the ITO surface during the wet

cleaning processes. The UV ozone treatment also helps to improve the surface work

function of the ITO anode facilitating the charge injection. They were then loaded in a

evaporation chamber, with a base pressure of 5×10^{-4} Pa, for device fabrication. The

thicknesses of the functional organic layers and the electrode and the corresponding

deposition rates were controlled and monitored by the calibrated crystal quartz

sensors. The organic interlayers (e.g., HTL, ETL, et al) were prepared using a

constant deposition of about 1 Å/s, while the organic emissive layers were formed

using a lower rate of 0.02 Å/s. LiF cathode interlayer and the Al top contact were

deposited at 0.1 Å/s and 3−6 Å/s, respectively. The resulting WOLEDs have an active

emissive area of 3 mm ×3 mm, defined by the overlap between the front ITO anode

and the rear Al cathode.

A computer controlled PR-655 Spectra scan spectrometer was used to measure

the EL spectra, Commission Internationale de L'Eclairage (CIE) coordinates, CCT,

and CRI of the WOLEDs. The current density−voltage−luminance ($J–V–L$)

characteristics were measured using a computer-controlled Keithley 2400 source

integrated with a BM-70A luminance meter. Luminous efficiency and power

efficiency of the WOLEDs were calculated from the J-V-L characteristics. EQE of the

WOLEDs was calculated using the J-V-L characteristics and spectra data. The

WOLEDs were not encapsulated and were characterized immediately after the

fabrication.

3. Results and discussion

Detection of carrier recombination zone

In a previous work, we demonstrated the Bepp$_2$ based OLED[25], ITO/ MoO$_3$ (3 nm)/

TCTA (40 nm)/ Bepp$_2$ (10 nm)/ TPBi (50 nm)/ LiF (1 nm)/ Al (200 nm) (named

device B, as shown in Figure 2), exhibited good blue emission with a peak at 440 nm

and achieved a high maximum luminance of 5565 cd/m^2. Herein, it is employed as a

basic blue device to develop hybrid WOLEDs. To obtain ideal EL spectra and high

device efficiency, the singlet and triplet excitons need be well manipulated and

managed, where it is the prerequisite to know the precise distribution of carrier

recombination zone in devices. So, a 0.5 nm-thick 4-(dicyanomethylene)-2-tert-butyl

-6-(1,1,7,7-tetramethyljulolidin-4-yl-vinyl)-4H-pyran (DCJTB) layer was introduced

at the TCTA/Bepp$_2$ interface, the centre of Bepp$_2$ EML, and the Bepp$_2$/TPBi interface

of the device B to fabricate a series of probe devices, corresponding to devices D1,

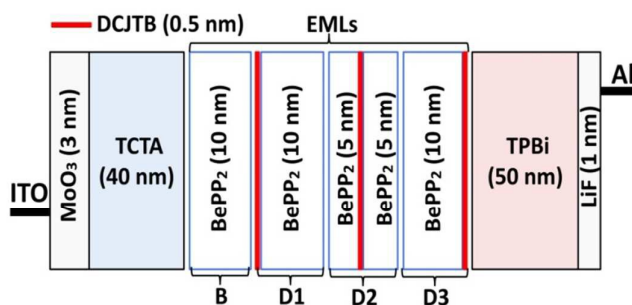D2, and D3, respectively, and the device structures are also shown in **Fig. 2**.

Fig. 2 The device structure diagram of the Bepp$_2$ based blue device B and probe
devices D1, D1, and D3.

    Fig. 3 shows the normalized EL spectra of the devices B, D1, D2, and D3 at the

voltages of 4 V, 5 V, 6 V, and 7 V. Compared with the reference device B, apart from

an intrinsic emission of Bepp$_2$ at 440 nm, an obvious emission peak of about 604 nm

in the EL spectra of the devices D1, D2, and D3 is also observed at the voltages of

4−7 V, which is typically derived from the emission of DCJTB. And as the

incorporation position of ultra-thin DCJTB layer keeps away from the TCTA/Bepp$_2$

interface corresponding to the devices D1, D2, to D3, with the increase of the voltage

from 4 V to 7 V, although there is an increasing trend for blue peak in the EL spectra for the devices D1, D2 and D3, the intensity ratios of the 604-nm red-light to the 440-nm blue-light in EL spectra for the device D1, D2 and D3 at the voltages of 4 V, 5 V, 6 V, and 7 V all gradually decrease, indicating that the electrons and holes mainly combine at TCTA/EML interface and some of electrons and holes also combine at EML/ETL interface and whole EML. This is attributed to i) the high energy barriers of 0.30 eV and 0.60 eV at TCTA/$Bepp_2$ and $Bepp_2$/TPBi interfaces (see **Fig. 4(a)**) and good electron transport capability for $Bepp_2$ (the mobility is ~10^{-4} cm^2/Vs)[36]. The knowledge of carrier combination zone will provide the scientific basis for the design of hybrid WOLEDs structure and the manipulation control of singlet and triplet excitons later.
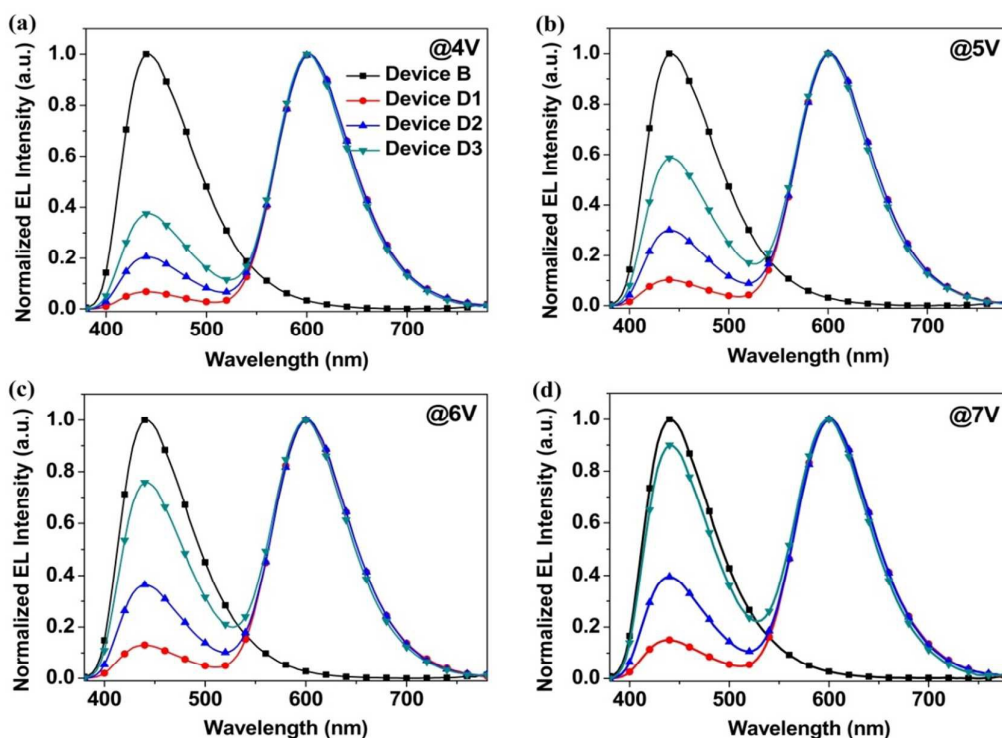

Fig. 3 The normalized EL spectra of the devices B, D1, D2, and D3 at the voltages of 4 V, 5 V, 6 V, and 7 V.

Di-chromatic hybrid WOLEDs

At above blue devices, taking into account the higher triplet energy levels of TPBi (2.70 eV) and TCTA (2.80 eV) than Bepp$_2$ (2.60 eV),[33-35] although the formed triplet excitons in the carrier recombination zone can be well limited in EML, the radiative decay for these triplet excitons is a forbidden transition, leading to a 75 % electricity generated excitons loss. The prerequisite for developing high efficient hybrid WOLEDs is to design artful device structure to manipulate and utilize these triplet excitons for generating light. Our motive is to let these triplet excitons limited in EML escape and migrate to TCTA and TPBi layers at two sides of EML, where the triplet excitons can be captured by lower triplet level phosphorescent emitters doped in TCTA and TPBi for light emission. So, a series of di-chromatic white devices series-W0 were prepared by doping yellow phosphor (Ir(ffpmq)$_2$(acac)) in HTL and ETL away from TCTA/Bepp$_2$ and Bepp$_2$/TPBi interfaces for different distance. The structures for devices series-W0 are as follows: ITO/ MoO$_3$(3 nm)/ TCTA(35-x nm)/ TCTA: 6 wt% Ir(ffpmq)$_2$(acac)(5 nm)/ TCTA(x nm)/ Bepp$_2$(10 nm)/ TPBi(x nm)/ TPBi: 6 wt% Ir(ffpmq)$_2$(acac)(5nm)/ TPBi(45-x nm)/ LiF(1 nm)/ Al(200 nm), here, x is 0, 1, 2, and 3, corresponding to the devices W0-1, W0-2, W0-3, and W0-4, respectively.

Fig. 4(a) displays the schematic diagram of the emission mechanism of white devices W0-1, W0-2, W0-3, and W0-4. In these white devices, the x nm-thick TCTA and TPBi layers are employed to manipulate the triplet excitons limited in Bepp$_2$ EML, for boosting device efficiency and obtaining white emission. This is because a 0−3 nm-thick TCTA or TPBi layer should not form a complete film,[35,37] where many unoccupied sites exist in film.[38] Although the triplet energy levels of TCTA (2.80 eV) and TPBi (2.70 eV) are higher than that of Bepp$_2$ (2.60 eV), the triplet excitons generated in Bepp$_2$ EML should also penetrate a x(0-3) nm-thick TCTA and TPBi

layers via unoccupied sites due to a long exciton diffusion distance (> 10 nm)[4,7,29,39]
and further transfer energy to Ir(ffpmq)$_2$(acac) by Dexter energy transfer for yellow
emission, inducing a high utilization rate of excitons. And combined with blue
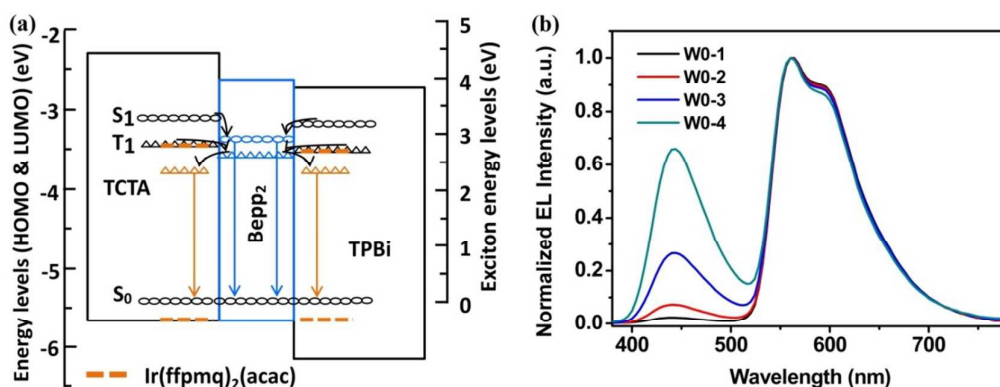emission from Bepp$_2$, it is expected that white emission can be obtained.

Fig. 4 (a) The schematic diagram of the emission mechanism of di-chromatic
WOLEDs W0-1, W0-2, W0-3, and W0-4; (b) The normalized EL spectra of four
white devices W0-1, W0-2, W0-3, and W0-4 at 6 V.

Fig. 4(b) depicts the normalized EL spectra of four white devices W0-1, W0-2,
W0-3, and W0-4 at 6 V. Clearly, the EL spectra for devices series-W0 all show two
emission peaks at blue and yellow wavelength, and a dominant peak is located at 560
nm, which is stemmed from Ir(ffpmq)$_2$(acac). The intensity ratio of the 560-nm
yellow-light to the 440-nm blue-light in EL spectra gradually decrease when the
distance x increases from 0 nm to 3 nm, indicating that the triplet excitons energy
transfer from Bepp$_2$ to Ir(ffpmq)$_2$(acac) can be well manipulated by only tuning the
value of x. These results effectively demonstrate the viability of above our motive.
However, the di-chromatic WOLEDs series-W0 obviously show poor color quality.
Considering a small percentage of blue intensity in EL spectrum for warm white, the
value for x is identified as 1 for developing high color quality hybrid WOLEDs later.

Tri-chromatic hybrid WOLED

To improve color quality of WOLEDs, the tri-chromatic WOLED W1 was fabricated

with the structure of ITO/ MoO_3(3 nm)/ TCTA (34 nm)/ TCTA: 8 wt% $Ir(piq)_2(acac)$

(5 nm)/ TCTA (1 nm)/ $Bepp_2$ (10 nm)/ TPBi(1 nm)/ TPBi: 6 wt% $Ir(ffpmq)_2(acac)$ (5

nm)/ TPBi (44 nm)/ LiF(1 nm)/ Al (200 nm). The distinction to WOLED W0-2,

replacing yellow emitter $Ir(ffpmq)_2(acac)$, the red emitter of $Ir(piq)_2(acac)$ was doped

in the HTL of TCTA in WOLED W1. From **Fig. 5(a)**, it can be seen that owing to i) a

lowest triplet energy level for $Ir(piq)_2(acac)$ and ii) a thin TCTA layer of 1 nm

between EML and $Ir(piq)_2(acac)$ doped TCTA layer as the case in WOLED W0-2, the

triplet excitons at TCTA/$Bepp_2$ interface should also penetrate easily 1 nm-thick

TCTA layer and be captured by $Ir(piq)_2(acac)$ for red light emission.

Fig. 5(b) depicts the EL spectra of WOLED W1 at different driving voltages, and

the corresponding luminance, CIE coordinates, CCT, and CRI parameters were also

listed at different driving voltages. Compared with WOLED W0-2, the EL spectra of

WOLED W1 obviously contain three emission peaks, and a dominant peak is located

at 612 nm corresponding to the emission of $Ir(piq)_2(acac)$. With the appearance of red

peak in EL spectra, device W1 exhibits improved white color quality with a maximum

CRI of 73. However, CRI is still not up to the threshold value of 80 for WOLEDs

used in lighting[7,25,35,40]. And from **Fig. 5(c)** and Figure 5(d), the device W1 also shows

relatively low device efficiency, maximum CE, PE, and EQE reaching 28.57 cd/A,

26.77 lm/W, and 12.11 %, respectively. Low device efficiencies may be ascribed to

the fact that a portion of triplet excitons are still limited in $Bepp_2$ EML, which cannot

be utilized for generating light, leading to an incomplete excitons harvest. This can be

verified from the EL spectra for WOLED W1 under different voltages. From **Fig.5 (b)**,

clearly, as the voltage increases from 5 V to 9 V, the normalized EL spectra of

WOLED W1 show an obviously increase in blue emission intensity, indicating that

the generated triplet excitons in $Bepp_2$ EML cannot be completely used to generate
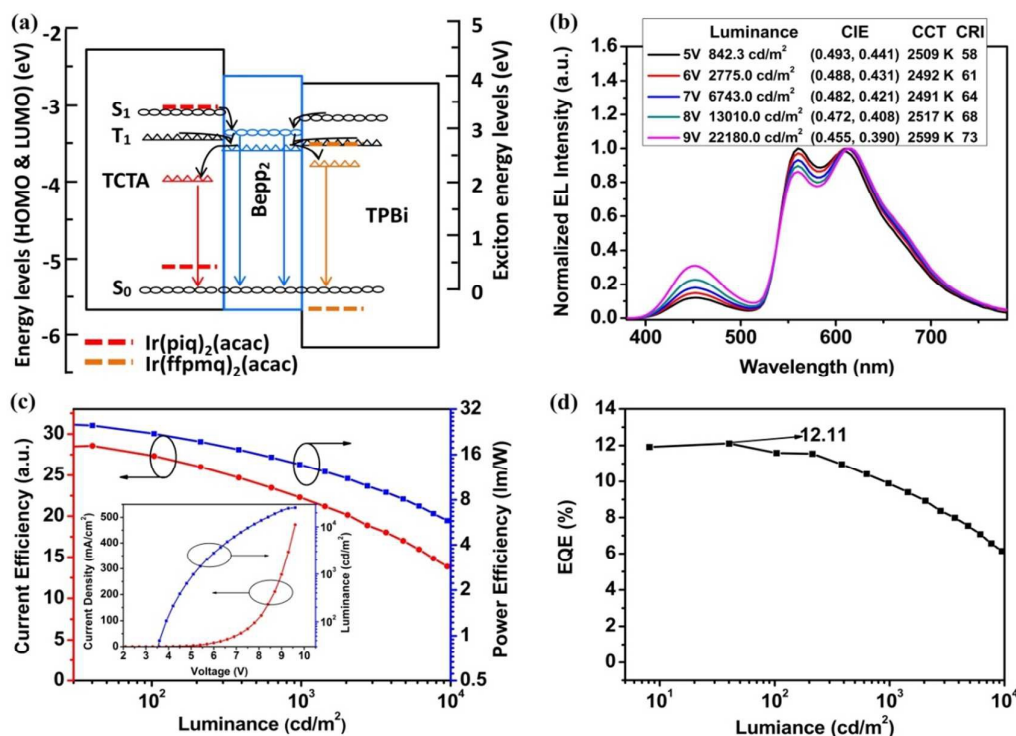
light under a higher voltage.

Fig. 5 (a) The schematic diagram of the emission mechanism of WOLED W1; (b) The

EL spectra, luminance, CIE coordinates, CCT, and CRI of WOLED W1 at different

voltages; (c) The CE-L-PE curves of WOLED W1, Inset: The J-V-L curves of

WOLED W1; (d) The EQE-L curves of WOLED W1.

Tetra-chromatic hybrid WOLEDs

In order to enhance device efficiency, the prerequisite is still to utilize triplet excitons

still limited in $Bepp_2$ EML for light emission. Here, we incorporated an ultrathin green

phosphorescence layer (0.02 nm) in the center of $Bepp_2$ EML in WOLED W1,

fabricated WOLED W2 with the structure of ITO/ MoO_3(3 nm)/ TCTA (34 nm)/ TCTA: 8 wt% $Ir(piq)_2(acac)$ (5 nm)/ TCTA (1 nm)/ $Bepp_2$ (5 nm)/ $Ir(ppy)_3$(0.02 nm) /$Bepp_2$ (5 nm)/ TPBi(1 nm)/ TPBi: 6 wt% $Ir(ffpmq)_2(acac)$ (5 nm)/ TPBi (44 nm)/ LiF(1 nm)/ Al (200 nm). From **Fig. 6(a)**, the generated singlet excitons in $Bepp_2$ EML can immediately recombine in $Bepp_2$ for blue emission, and some of the triplet excitons formed in $Bepp_2$ EML can penetrate 1 nm-thick TCTA and TPBi layers to transfer energy to $Ir(piq)_2(acac)$ and $Ir(ffpmq)_2(acac)$ for red and yellow emission, respectively. The remaining triplet excitons limited in $Bepp_2$ EML should migrate to 0.02 nm-thick $Ir(ppy)_3$ layer located at the center of $Bepp_2$ EML for green emission due to i) a lower triplet level for $Ir(ppy)_3$ (2.40 eV) than for $Bepp_2$ (2.60 eV) and ii) a long diffusion distance (> 10 nm) for triplet excitons.[4,7,29,39] So, in theory, the complete electricity generated excitons in WOLED W2 can be harvested for generating light, indicating that high device efficiency should be obtained.

From **Fig. 6(b)**, the EL spectra of white device W2 visibly contain a green emission peak at 512 nm from $Ir(ppy)_3$, which demonstrates our viewpoint that the triplet excitons generated in $Bepp_2$ EML indeed cannot completely migrate to TCTA and TPBi layers at both sides of $Bepp_2$ EML, and the remaining triplet excitons are still limited in $Bepp_2$ EML in WOLED W1, which can be exactly harvested by $Ir(ppy)_3$ for green emission. These are further demonstrated from **Fig. 6(c)** and **Fig. 6(d)**, where the device efficiencies for WOLED W2 are effectively improved, the maximum CE, PE, and EQE reaching 44.6 cd/A, 40.20 lm/W, and 16.83 %, respectively. Here, the EQE for WOLED W2 is higher (by 1.39 time) than that of WOLED W1. However,

the EQE for WOLED W2 is still below a theoretical value of above 20 % for OLEDs.

The reason may be that a 0.02 nm-thick Ir(ppy)$_3$ layer is too thin to completely

harvest remaining triplet excitons in Bepp$_2$ EML. Besides, WOLED W2 also exhibits

relatively poor color quality with a low CRI of 68, owing to a low red intensity in EL

spectra. If a thicker Ir(ppy)$_3$ layer (> 0.02 nm) is inserted in the center of Bepp$_2$ EML,

which will be helpful for further improving device efficiency. But a thicker Ir(ppy)$_3$

layer will also lead to a worse color quality because of an further increased green
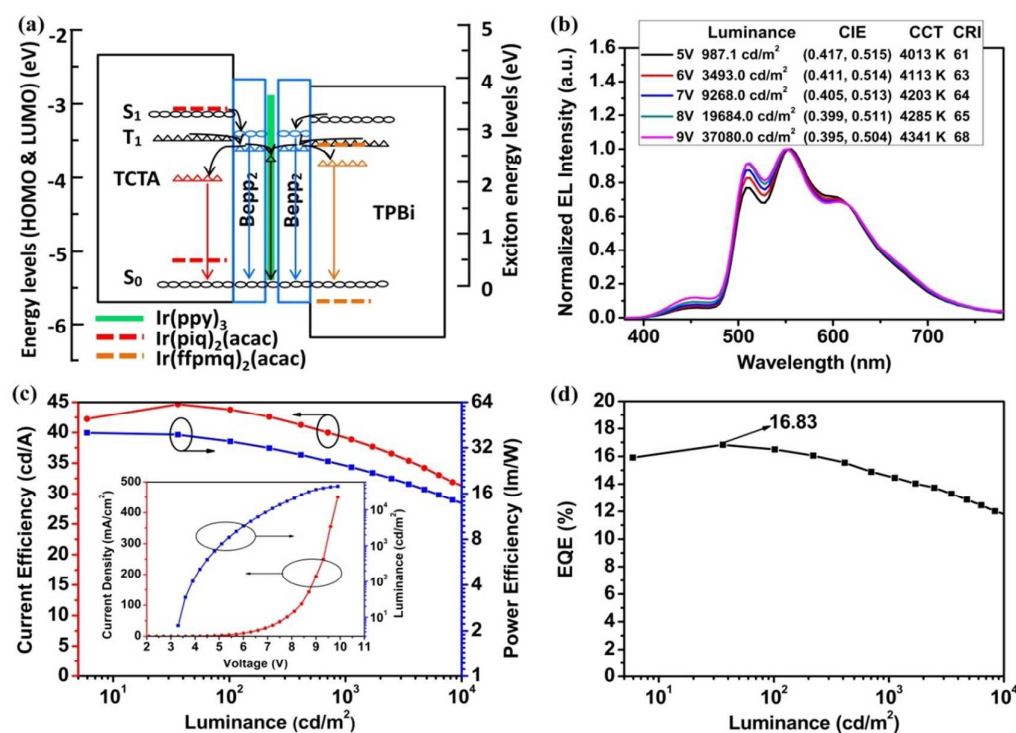
intensity in EL spectra.

Fig. 6 (a) The schematic diagram of the emission mechanism of WOLED W2; (b) The

EL spectra, luminance, CIE coordinates, CCT, and CRI of WOLED W2 at different

voltages; (c) The CE-L-PE curves of WOLED W2, Inset: The J-V-L curves of

WOLED W2; (d) The EQE-L curves of WOLED W2.

Considering a good tradeoff between device efficiency and color quality, to further

boost device efficiency and color quality of WOLEDs simultaneously, an ultrathin green phosphorescence layer (0.02 nm) is replaced with an ultrathin red phosphorescence layer (0.05 nm), that was incorporated in the center of $Bepp_2$ EML, and green phosphorescent $Ir(ppy)_3$ was doped in TCTA 1 nm away from TCTA/$Bepp_2$ interface, to fabricate WOLED W3 with the structure of ITO/ MoO_3(3 nm)/ TCTA (35 nm)/ TCTA: 6 wt% $Ir(ppy)_3$ (4 nm)/ TCTA (1 nm)/ $Bepp_2$ (5 nm)/ $Ir(piq)_2(acac)$ (0.05 nm) /$Bepp_2$ (5 nm)/ TPBi(1 nm)/ TPBi: 6 wt% $Ir(ffpmq)_2(acac)$ (5 nm)/ TPBi (44 nm)/ LiF(1 nm)/ Al (200 nm). Here, a ultrathin red phosphorescence layer (0.05 nm) is thicker than a ultrathin green phosphorescence layer (0.02 nm) in WOLED W2, which is employed to completely harvest remaining triplet excitons limited in $Bepp_2$ EML for red emission to enhance device efficiency. And the adequate red emission intensity is also beneficial for improving the color quality of white device at the same time.

From **Fig. 7(b)**, surprisingly, as we expect that WOLED W3 realizes ideal warm white emission. The EL spectra cover most wavelengths of the visible region from 380 nm to 780 nm with four main emission peaks at 452 nm, 512 nm, 560 nm, and 612 nm corresponding to the emissions of $Bepp_2$, $Ir(ppy)_3$, $Ir(ffpmq)_2(acac)$, and $Ir(piq)_2(acac)$, respectively. And the EL spectra for WOLED W3 are similar to the spectra of candle light, showing a step increasing blue, green, yellow, red emission intensity. All of these indicate the singlet and triplet excitons in this WOLED are well manipulated and managed for the emission of individual color. A dominant red light emission peak in EL spectra also effectively boosts CRI[25], reaching a maximum CRI of 94. Here, to dope green $Ir(ppy)_3$ in TPBi layer and yellow $Ir(ffpmq)_2(acac)$ in

TCTA layer, in theory, the device can still achieve the high efficiency because the triplet energy levels of TPBi and TCTA are higher than that of Ir(ppy)$_3$ and Ir(ffpmq)$_2$(acac) dopants, which ensure the triplet excitons in TCTA and TPBi layers, escaped from Bepp$_2$ EML, can be capture by Ir(ffpmq)$_2$(acac) and Ir(ppy)$_3$ for light emission. However, such doping change of green and yellow emitters will lead to the variation in EL spectra, which is not what we expected.
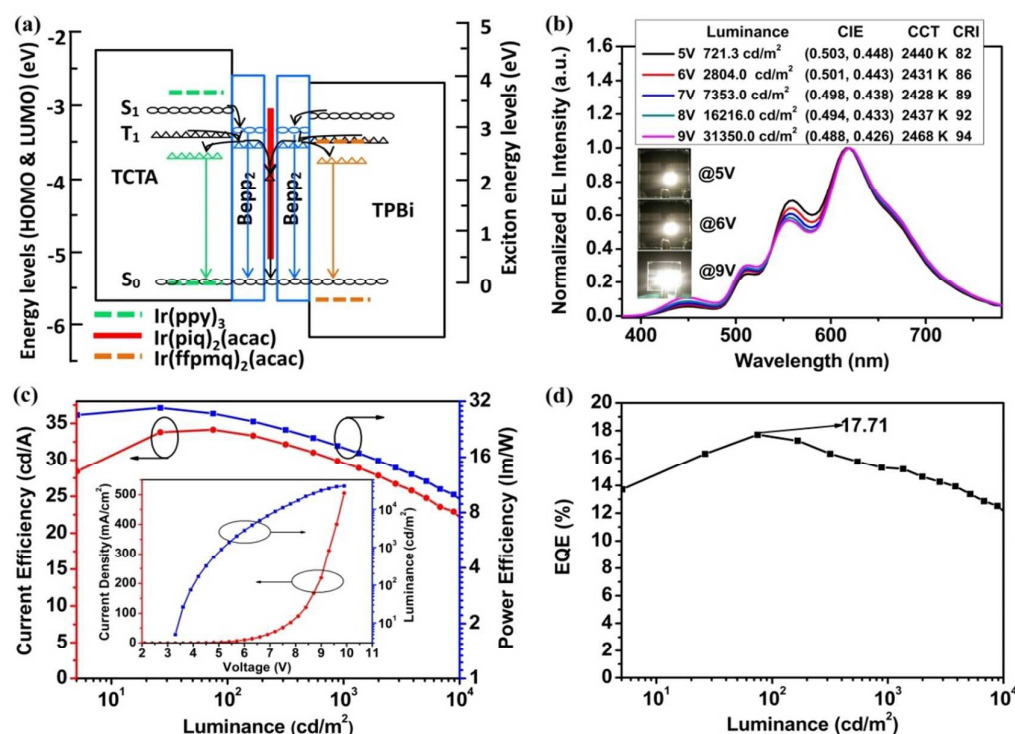

Fig. 7 (a) The schematic diagram of the emission mechanism of WOLED W3; (b) The normalized EL spectra, luminance, CIE coordinates, CCT, and CRI of WOLED W3 at different voltages; (c) The CE-L-PE curves of WOLED W3, Inset: The J-V-L curve of WOLED W3; (d) The EQE-L curves of WOLED W3.

From **Fig. 7(c)** and **Fig. 7(d)**, WOLED W3 also shows a low turn-on voltage of 3.3 V and reaches a maximum luminance of 40970 cd/m^2. And with increased red intensity in EL spectra, the EQE of WOLED W3 is also further improved, reaching

17.71 %, compared with WOLED W2 (16.83%). However, the EQE (17.71%) is still slightly lower than the theoretical maximum value of 20%, which may be ascribed to the energy loss due to the transfer of triplet excitons from $Bepp_2$ to phosphors (red, green, yellow). The maximum CE and PE reach 34.15 cd/A and 29.51 lm/W, respectively. Some EL performance parameters for WOLEDs W1, W2, and W3 are summarized in Table 1. The improved device efficiency should be attributed to an almost complete harvest of remaining triplet excitons in $Bepp_2$ EML by 0.05 nm-thick $Ir(piq)_2(acac)$ layer. In addition, the WOLED W3 also exhibits the excellent color stability, showing a slight EL spectra change over the voltage range of 5−9 V (corresponding to a luminance from 721.3 cd/m^2 to 31350 cd/m^2). Specially, with the voltage increases from 5 V to 8 V, the intensity of blue emission (512 nm) in the normalized EL spectra remains almost the constant, which further confirms that a 0.05 nm-thick $Ir(piq)_2(acac)$ layer enables to harvest completely the remaining triplet excitons in $Bepp_2$ EML under a higher voltage.

Table 1. The performance summary of different WOLEDs.

Devices	V_{on} [V][a]	Maximum			CIE(x,y) @7 V	CCT @5 V−9 V	CRI @5/6/7/8/9 V
		EQE [%]	CE [cd/A]	PE [lm/W]			
W1	3.3	12.11	28.57	26.77	(0.482,0.421)	2509−2599	58/61/64/68/73
W2	3.3	16.83	44.69	40.20	(0.405,0.513)	4013−4341	61/63/64/65/68
W3	3.3	17.71	34.15	29.51	(0.498,0.438)	2440−2468	82/86/89/92/94

[a] V_{on} : Voltage at a luminance of > 1 cd/m^2.

It is worth mentioning, in this work, that the proposed hybrid WOLED avoids an additional interlayer between the fluorescent and phosphorescent zones, inducing a simplified device structure with reduced heterojunction interfaces, which is very

meaningful for promoting the commercial development of WOLEDs. And the proposed hybrid WOLED also realizes a good manipulation and exploitation of singlet and triplet excitons for an almost 100% excitons harvest, achieving a high device performance. To our knowledge, this is one of the best results to date for a hybrid WOLED exhibiting superior efficiency (17.71 %), high CRI (94) and good color stability simultaneously[5,7,10,15,19,40].

4. Conclusions

In this work, a high performance tetra-chromatic hybrid WOLED without additional interlayers was demonstrated. The hybrid WOLED realizes good warm white emission with a maximum CRI of 94, low CCT of 2440−2468 K and stable EL spectra over a wide voltage range of 5−9 V. Moreover, the device also achieves the maximum CE, PE, and EQE of 34.15 cd/A, 29.51 lm/W, and 17.71 %, respectively, measured without any light out-coupling features, rendering the best performing warm WOLED. This new WOLED concept, attained through efficient excitons manipulation and harvest, opens up a new avenue for high performance hybrid WOLEDs.

Conflicts of interest

There are no conflicts to declare.

Acknowledgments

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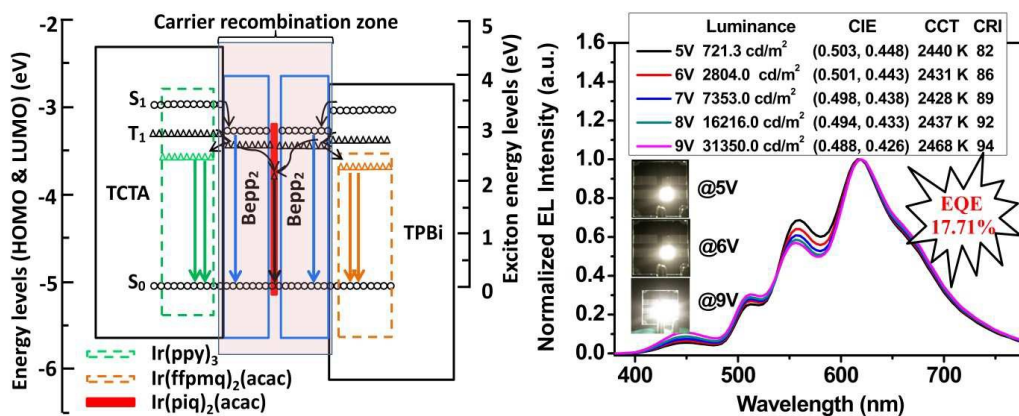
The tetra-chromatic hybrid WOLED without additional interlayers between the fluorescent and phosphorescent emitting regions exhibits good warm white emission, showing stable EL spectra with a maximum CRI of 94 and a low CCT of 2440−2468 K over a wide voltage range from 5−9 V, and also reveals a high EQE of 17.71 %.

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