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# Solution-Processed White Organic Light-Emitting Diodes with bi-Component Emitting Layer Based on Symmetry Blue Spiro-Sulfone Derivative

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## Abstract

We report a white organic light-emitting diode (WOLED) of simple construction with a bi-component emitting layer, in which a novel, dual-role deep blue TADF emitter as both the non-doped blue emitter and efficient host of an orange TADF emitter of TXO-TPA. The deep blue molecule (SDPS-4PhCz, CIE<sub>y</sub> ~ 0.04) was designed using spiro-diphenylsulfone (SDPS) as electron-accepting group and four 9-phenyl-9H-carbazole (N-PhCz) as the electron-donating groups. SDPS-4PhCz showed delayed fluorescence with short delayed lifetime of 1.4  $\mu$ s via fast reverse intersystem crossing (RISC) from "hot exciton" triplet excited state. The solution-processed, non-doped deep blue inverted OLED device exhibited a maximum EQE of 4.92% with low efficiency roll-off. The two-color WOLED with extremely simple device architecture showed an EQE above 17%, CIE (0.39, 0.44) and high CRI of 64 by spin-coating method.

## 1. Introduction

White organic light-emitting diodes (WOLEDs) have attracted significant attention in both

scientific and industrial communities due to their great applications in full color displays and solid-state lighting [1-4]. To realize high efficiency and high color rendering index (CRI) of WOLEDs, much progress on all-phosphorescence and fluorescence-phosphorescence hybrid WOLEDs has been widely made [5-8]. However, the low efficiency of fluorescence and low stability of phosphorescence devices spur the exploration of new alternatives. Since Adachi and co-workers firstly reported WOLEDs based on thermally activated delayed fluorescence (TADF) emitters [9], TADF emitters have been widely used for WOLEDs with high efficiency and high CRI due to their theoretical 100% internal quantum efficiency (IQE) and broad full width at half maximum (FWHM) [10, 11]. For example, Ma and co-workers reported a TADF-based hybrid WOLED with superior efficiency/CRI/color stability and low efficiency roll-off [12]. Su's group reported WOLEDs based on all-fluorescence to give high external quantum efficiency (EQE), satisfied Commission International de l'Eclairage (CIE) coordinates, CRI and color stability [13]. However, high efficiency with low roll-off and high CRI can only be achieved for complicated WOLED device structure. Very recently, our group reported a high efficient bi-component WOLED by using a new blue TADF of o, o'-NPh2 and a yellow TADF of TXO-PhCz4, an EQE of 12.5% and a high CRI of 71.0 were obtained due to the broad emission spectra of both blue and yellow TADF emitter [14].  $o_1 o_2 - NPh_2$  serves as not only blue energy source but also the efficient host of yellow TADF emitter to produce properly balanced color chromaticity in the simple WOLEDs. Thus, preparing WOLEDs with higher efficiency and simple structure was desirable [15-17]. Although several blue TADF emitters have been reported since 2012, blue-emitting materials have been the short board that hampers the performance of WOLEDs due to the problem of efficiency and stability [18-20]. Especially the lack of deep blue TADF materials may make it difficult to fully cover a wide range of the visible-light spectral region (380-780 nm) in the simple-structure WOLEDs, resulting in low CRI of the WOLEDs [21]. All these inspire us to develop novel deep blue emitting materials with high efficiency for the WOLED with simple structure.

A limited  $\pi$ -conjugation length and high rigidity geometry is required to achieve a deep blue emitter because of its wide band gap. In recent years, spiro molecular system with its unique advantages has attracted much attention in the field of organic photoelectric materials [22, 23]. The spiro structure can relieve the self-aggregation of molecules, thereby reducing exciton annihilation, which is expected to solve the problem of efficiency roll-off in non-doped devices [24]. Our group reported an effective strategy of constructing emitter MTXSFCz with a thioxanthone (TX) acceptor and

PhCz donor linked by a saturated sp<sup>3</sup> hybridized carbon of spirofluorene, which served as host of highly efficient PHOLEDs with low EQE roll-off [25]. In addition, the spiro materials possess several advantages, such as improving the thermal [26] and morphological stabilities [27] increasing the solubility [28] and charge-transport properties [29]. Lee and his co-workers reported a series of tert-butylated spirofluorene derivatives, all devices based on them showed efficient blue emission with good device performances [30]. In 2012, Adachi and co-workers developed a spiro derivative using cyano as acceptor and diphenylamine/acridine as donors showing TADF property, which yet exhibited green emission color [31, 32]. Herein, we report a spiro-diphenylsulfone based TADF material that emits in deep blue region with CIE coordinates of (0.15, 0.04). Notably, solution-processed non-doped single blue inverted electroluminescent (EL) device and highly efficient single-EML WOLED device doped with orange TADF emitter have been demonstrated.

#### 2. Experimental details

#### 2.1 Materials and instruments

Unless otherwise noted, all chemicals and reagents were commercially available and were used without further purification. Proton nuclear magnetic resonance (<sup>1</sup>H NMR) spectra were recorded on a Bruker Advance 400M spectrometer using TMS as internal standard at room temperature. Mass spectra were recorded using a BIFLEXIII mass spectrometer. Ultraviolet-visible (UV-vis) absorption spectra and photoluminescence (PL) spectra were measured by using Hitachi U-3900 spectrophotometer and F-4600 fluorescence spectrometer. The transient PL decay characteristics were measured via Laser Flash Photolysis Spectrometer (LP980-KS, Edinburgh Instruments). The photoluminescence quantum yields (PLQYs) were measured by Hamamatsu absolute PLQY spectrometer C11347. Differential scanning calorimetry (DSC) was carried out using a TA Instruments DSC 2910 thermal analyzer under a nitrogen atmosphere at a heating rate of 10 °C min<sup>-1</sup>. Thermogravimetric analyses (TGA) were performed on a TA Instruments TGA 2050 thermal analyzer. Ultraviolet photoelectron spectroscopy (UPS) was performed in a Kratos Axis Ultra Dld system using a He-I gas discharge lamp source with the excitation energy of 21.22 eV and the energy resolution of 150 meV.

#### 2.2 Theoretical Calculations

All the calculations were performed with the Gaussian 09 program package at the

B3LYP/6-31G<sup>\*\*</sup> level. The molecular geometries of ground states for SDPS-4PhCz were optimized by density functional theory (DFT). Based on the optimized geometries, time-dependent DFT (TDDFT) was then used to calculate the lowest singlet ( $S_1$ ) and triplet ( $T_1$ ) states.

#### 2.3 Devices Fabrication and Characterization

Before device fabrication, the ZnO precursor was synthesized according to the method that was described in previous report [33]. Accordingly, zinc acetate dehydrate (Zn(CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O) and ethanolamine (NH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH, 99.0%) (0.45 mol L<sup>-1</sup>) were stirring vigorously for 12 h in air. Afterwards, we filtered the solution with a polytetrafluoroethylene (PTFE) filter (0.2 µm) to remove precursor aggregates. The ITO glass substrates were sequentially cleaned by standard procedure and dried at 75 °C (15 min) in an oven. 10 min of oxygen plasma treat was conducted before spin-coating in order to remove any organic residues and to polarize the ITO surface for better ZnO precursor adhesion. The ZnO precursor was spin-coated onto the pre-patterned ITO substrates at a spin speed of 7000 rpm and annealed at 150 °C for 1 h in ambient condition. After that, PEIE:  $Rb_2CO_3$  (m/m = 1:1) blend solution was spin-coated on the ZnO layers at a spin speed of 7000 rpm and the substrates were rinsed with DMF, leaving ultrathin layers of PEIE: Rb<sub>2</sub>CO<sub>3</sub> on top of the ZnO films. After annealing at 100 °C for 10 min in a glove box, the solution of SDPS-4PhCz in chlorobenzene was spin-coated on the PEIE: RbCO<sub>3</sub> layer at a spin speed of 3000 rpm. For the WOLED, different concentrations of TXO-TPA were added into SDPS-4PhCz and dispersed in toluene. The binary constituent solution was spin-coated onto the PEDOT:PSS-PFI (m/m = 3/1) as same as above steps and annealed at 90  $\Box$  for 15 min. Then the sample was transferred into a vacuum deposition system, organics and the corresponding electrodes were deposited under a pressure below  $1 \times 10^{-4}$  Pa. The current-voltage characteristics were measured with a computer-controlled Keithley 2400 Source Meter and CS-200 under ambient atmosphere. The transient EL decay characteristics were measured using a pulse generator (Tektronix AFG3011C) and a spectrometer (Princeton Instruments, Acton Spectrapro SP-2300).

#### **3. Results and discussion**

#### 3.1 Synthesis and thermal Properties

Based on the design principles and the excellent characteristics of spiro structure, we selected spiro-diphenylsulfone (SDPS) as electron-accepting group and four 9-phenyl-9H-carbazole as the

donors which are connected to the para-position of phenylsulfone. SDPS-4PhCz is synthesized in six-steps with overall yield of 18% (Scheme 1 and Supporting Information). Addition of thioxanthone to (2-Iodophenyl) (phenyl) sulfane with n-BuLi in THF at -78 °C resulted in the formation of spiro-diphenylsulfide. Subsequently, spiro-diphenylsulfide underwent selective bromine and oxidation to produce bromo-spiro-diphenylsulfide. Finally, bromo-spiro-diphenylsulfide was allowed to react with 9-phenyl-9H-carbazol-3-yl boronic acid under palladium-catalysed Suzuki-Miyaura cross coupling conditions to obtain the target compound SDPS-4PhCz, which was characterized by <sup>1</sup>H, <sup>13</sup>C-NMR, high-resolution mass spectrometry and elemental analysis. In addition, due to the introduction of N-PhCz, SDPS-4PhCz showed good solubility in solvents as toluene and chlorobenzene which is favorable for device fabrication. The thermal properties of SDPS-4PhCz were investigated by TGA and DSC. Owing to the spiro and rigid skeleton of the emitter, SDPS-4PhCz has a high decomposition temperature (T<sub>d</sub>, 5% weight loss) of 574 °C and a glass transition temperature (T<sub>g</sub>) of 252 °C (Fig. S1). The HOMO energy level of SDPS-4PhCz was estimated to be 5.74 eV by UPS measurements, and the LUMO energy level was calculated to be 2.49 eV according to the band gap of 3.25 eV (Fig. S2). The thermal stability and frontier molecular orbital energy levels of SDPS-4PhCz are summarized in Table 1.



Scheme 1 Synthetic route of SDPS-4PhCz

<sup>3.2</sup> Photophysical properties

Photophysical properties of SDPS-4PhCz were analyzed using UV-vis and PL spectrometer. The absorption and PL peaks of SDPS-4PhCz in 2-methyltetrahydrofuran (2-Me THF) and in neat thin film and its PLQY are summarized in Table 1. SDPS-4PhCz exhibits absorption maximum at 337 nm and unstructured deep blue emission band at 413 nm in 2-Me THF. The absorption spectra were slightly influenced by the polarities of solvents, while remarkable positive solvatochromism property for emission spectra of SDPS-4PhCz was observed (Fig. S3), which was typical for those of reported intramolecular charge transfer (ICT) compounds [34-36]. The SDPS-4PhCz exhibited slightly red-shift absorption and PL spectra at 353 nm and 429 nm, respectively, as well as PLQY of 50% under nitrogen atmosphere in pure film (Fig. 1a).

The fluorescence and phosphorescence emission of SDPS-4PhCz were examined at 77 K in 2-Me THF. As shown in Fig. 1b, SDPS-4PhCz exhibited unstructured fluorescence band at 390 nm, which is assigned to the fluorescence from  $S_1$  state. The band at 466 nm in the phosphorescence spectrum is assigned to the phosphorescence from  $T_1$  state. Accordingly, the energy levels of  $S_1$  and  $T_1$  were achieved to be 3.18 eV and 2.66 eV, and a large  $\Delta E_{ST}$  of 0.52 eV was qualitatively achieved. Noticeably, after magnification, it is visible for the phosphorescence spectrum at 388 nm, which was consistent with the fluorescence emission at 77 K, suggesting the delayed fluorescence properties.



Figure 1 (a) UV-vis absorption and PL spectra of SDPS-4PhCz in 2-Me THF and thin film. (b) Fluorescence and phosphorescence spectra of SDPS-4PhCz in 2-Me THF at 77 K. ( $\lambda_{ex} = 350$  nm)

Compound	$\lambda_{abs}[nm]^{a)}$	$\lambda_{PL}[nm]^{a)}$	$\phi_{PL}[\%]^{a)}$	$T_d^{b}/T_g^{c}$	HOMO <sup>d)</sup> [eV]	LUMO <sup>d)</sup> [eV]	$\tau_p^{e)}$ (ns)	$\tau_d^{e)}$ (µs)
SDPA-4PhCz	337/353	413/429	83/50	574/252	-5.74	-2.49	100	1.45

Table 1 Optical, electrochemical and thermal data of SDPS-4PhCz.

<sup>a)</sup> Measured in 2-Me THF  $(1 \times 10^{-5} \text{ M})$  solutions and neat thin film at room temperature;

<sup>b)</sup> T<sub>d</sub>: decomposition temperature (5% weight loss);

<sup>c)</sup> T<sub>s</sub>: glass transition temperature;

- <sup>d)</sup> HOMO is calculated from the UPS spectra of SDPS-4PhCz thin film, LUMO is calculated from the onset of the absorption edge of the thin film;
- $^{\rm e)}$  PL lifetime of prompt and delayed decay component in deoxygenated 2-Me THF (10  $\mu$ M) solution at room temperature.

To confirm the delayed fluorescence of SDPS-4PhCz, the transient photophysical properties were investigated. The transient PL decay curves of SDPS-4PhCz in oxygen-free 2-Me THF revealed obvious two-order exponential decay, with a fast 100 ns relaxation and a second delayed 1.45  $\mu$ s relaxation (Fig. 2a). The microsecond-scale delayed component of SDPS-4PhCz was much shorter than those of traditional blue TADF materials, which helped to solve the problem of efficiency roll-off of the device [37]. As shown in Fig. 2b, the good overlap between the PL spectra of prompt component and delayed component indicated that both the prompt and delayed emissions stem from the radiative decay of the singlet excitons. Similar to other conventional TADF emitters, the emission intensity and delayed lifetime of SDPS-4PhCz were sensitive significantly toward oxygen in 2-Me THF [38]. The oxygen quenching results are summarized in Table S1 and Table S2. The delayed lifetime of SDPS-4PhCz was suppressed by the presence of oxygen and the PLQYs significantly enhanced with nitrogen bubbling (Fig. 2c and 2d). Consequently, it can be expected that the triplet states were involved in the PL process and the delayed fluorescence of SDPS-4PhCz arise from the emission harvesting from the triplet via reverse intersystem crossing (RISC). Additionally, similar temperature-dependence of the delayed lifetime was observed for the classical TADF emitters. At lower temperature (77 K), the delayed lifetime of SDPS-4PhCz dramatically decreased to 0.61 µs when compared with that at room temperature, implying RISC process of SDPS-4PhCz from lower triplet to singlet state. Transient experiment results showed that photophysical properties of SDPS-4PhCz were significantly different with CzS2 reported by Zhang and co-workers [39]. The RISC rate constant was calculated to be  $5.6 \times 10^5$  s<sup>-1</sup>, which was smaller than those of the conventional blue TADF emitters (Table S3). The small  $k_{TADF}$  indicated a large energy gap between the singlet and triplet ( $\Delta E_{ST}$ ) for the RISC.



Figure 2 (a) PL transient spectra of SDPS-4PhCz in 2-Me THF with saturated nitrogen at 300 K and 77 K. (b) Prompt and delayed fluorescence spectra of SDPS-4PhCz in 2-Me THF at 300 K. (c) PL transient spectra of SDPS-4PhCz in 2-Me THF under oxygen and nitrogen atmospheres, respectively. Concentration of SDPS-4PhCz is 10  $\mu$ M. (d) Fluorescence spectra of SDPS-4PhCz in 2-Me THF with saturated nitrogen and oxygen at 300 K. (excitation and detection wavelength were 355 nm and 404 nm)

#### 3.2 Theoretical calculations

To gain further insight into the excited state properties of SDPS-4PhCz, the landscape for singlet and triplet excited states and the highest occupied and lowest unoccupied natural transition orbitals (NTO) of the S<sub>1</sub> and T<sub>n</sub> (n = 1~3) states were performed. The NTO particle wave function of the S<sub>1</sub> state is mainly localized on the SDPS unit and the adjacent linked phenyl of PhCz, while the NTO hole wave function is localized on the PhCz unit and the adjacent linked phenyl of the SDPS (Fig. 3a). The substantial overlap between the NTO hole and particle leads to the weak CT characteristics in S<sub>1</sub> state in nature, corresponding to the large exchange energy, *K* [40]. However, for the T<sub>1</sub> state, NTO hole and particle wave functions are mainly confined the PhCz moiety and adjacent phenyl ring on SDPS moiety, suggesting the locally excited (LE) characteristics in T<sub>1</sub>. Large  $\Delta E_{ST}$  (S<sub>1</sub>-T<sub>1</sub>) of 0.57 eV was achieved (Fig. 3b), which coincides with the results obtained from steady PL at 77 K and exceeds the upper limit

value for  $\Delta E_{ST}$  of TADF emitters (0.3 eV). For the NTO of higher excited states,  $\Delta E_{ST}$  (S<sub>1</sub>-T<sub>2</sub>) of 0.44 eV and  $\Delta E_{ST}$  (S<sub>1</sub>-T<sub>3</sub>) of 0.16 eV were obtained due to the significant LE characteristics of the T<sub>2</sub> and T<sub>3</sub> state. The spin orbital coupling (SOC) matrix element values were calculated to be 0.05751 cm<sup>-1</sup> between S<sub>1</sub> and T<sub>1</sub>, 0.17920 cm<sup>-1</sup> between S<sub>1</sub> and T<sub>2</sub> for SDPS-4PhCz, combined with the larger  $\Delta E_{ST}$ (S<sub>1</sub>-T<sub>1</sub>) and  $\Delta E_{ST}$  (S<sub>1</sub>-T<sub>2</sub>), the small SOC will inhibit the germination of RISC process between S<sub>1</sub> and T<sub>1</sub> or S<sub>1</sub> and T<sub>2</sub>. Therefore, we speculated that the RISC process occurs from higher energy T<sub>3</sub> to S<sub>1</sub> because of the small  $\Delta E_{ST}$  of 0.16 eV and stronger SOC matrix element of 0.12759 cm<sup>-1</sup> between S<sub>1</sub> and T<sub>3</sub>. Thus, SDPS-4PhCz exhibited fast RISC via high-lying triplet excited state, and "hot exciton" TADF mechanism is expected in our systems [41-43]. Therefore, the calculations further demonstrate that the energy levels of the singlet and triplet excited states in SDPS-4PhCz changes significantly as compared with CzS2, ultimately resulting in different photophysical properties.



Figure 3 (a) Natural transition orbitals (hole: upper, particle: lower) for the  $S_1$ ,  $T_1$ ,  $T_2$  and  $T_3$  excitations for SDPS-4PhCz (the weight for the transition is given on the right of the arrow). (b) The energy diagram of the singlet and triplet excited states of SDPS-4PhCz.

#### 3.3 Electroluminescent Performance

To study the electroluminescence properties of the blue emitter, solution-processed devices were fabricated using inverted device structure of ITO/ZnO (30 nm)/PEIE:  $Rb_2CO_3$  (5 nm)/EML (80 nm)/TCTA (30 nm)/MoO\_3 (10 nm)/Al (100 nm) (Fig. S4). In the devices, ZnO and MoO\_3 served as the electron-injection and hole-injection layers, respectively, and PEIE:  $Rb_2CO_3$  and TCTA served as electron-transporting and hole-transporting layers, respectively. The EL spectrum of device was blue-shifted to 417 nm, compared to the PL spectrum of the solid thin film. The device exhibited excellent color saturation with CIE coordinates (0.15, 0.04) and showed a turn on voltage (V<sub>T</sub>) of 3.0 V, a current efficiency (CE) of 1.15 cd/A, a power efficiency (PE) of 1.12 lm/W, and the external quantum

efficiency (EQE) is determined to be 4.92% (Fig. 4), which represent the state-of-the-art device performance for "hot exciton" deep blue TADF OLEDs (Table S4). The exciton utilization of the OLED is estimated according to the following equation:

$$EQE = \eta_{rec} \times \eta_{PL} \times \eta_s \times \eta_{out}$$
(1)

Where the PL efficiency of thin film  $\eta_{PL}$  was determined to be 50%, the charge balance factor  $\eta_{rec}$ and optical out-coupling  $\eta_{out}$  are assumed to be 1.0 and 0.2~0.3, respectively. The fraction of singlet exciton  $\eta_s$  is estimated to be 49.4~32.8%, overcoming the 25% theoretical limit imposed by spin statistics. These results confirmed that the triplet excitons of SDPS-4PhCz were effectively utilized. According to the energy-gap law [44], the small energy gap between T<sub>2</sub> and T<sub>3</sub> may significantly increase the internal conversion rate. As a result, serious internal conversion from T<sub>3</sub> can be expected, leading to the low triplet excitons utilization. More fascinatingly, the deep blue device exhibited low efficiency roll-off compared with most of the traditional blue TADF OLEDs, giving EQE value up to 2.08% at the luminescence of 1000 cd/m<sup>2</sup> (Table S5). The low efficiency roll-off was ascribed to the relatively short lifetime of the delayed component of SDPS-4PhCz [45-47]. Furthermore, the transient EL was measured to confirm the TADF property of the emitter in the device. The delayed EL component lasted for several microseconds and the EL spectra with different delay times well matched (Fig. 5). Thus, the efficient RISC facilitated the delayed fluorescence under electric excitation and afforded a high EQE.



Figure 4 The performances of blue OLED: (a) current efficiency and power efficiency versus luminance curves; (b) current density and luminance versus voltage (J-V-L) curves; (c) EQE versus luminance curves; (d) EL spectra of blue device.



Figure 5 Transient EL decay under electrical excitation at 300 K (inset: EL spectra were resolved into a prompt component (black) and delayed component (red)).

We further applied the deep blue emitters for the fabrication of simple structure WOLEDs. TXO-TPA was chosen as the orange TADF emitter due to its appropriate CIE coordination [36]. As shown in Fig.6a, the emission band of SDPS-4PhCz and the absorption band of TXO-TPA showed significant overlap which is favorable for the Förster resonance energy transfer from SDPS-4PhCz host to TXO-TPA emitter. Increasing the dopant concentration from 1 wt% to 10 wt% decreased the emission intensity of SDPS-4PhCz, while retained the intensity of TXO-TPA in blend films (Fig.6b).

The red-shifted emission of TXO-TPA is not only attributed to the interaction between the host and dopant but also between the guests. As a result, a pure white spectrum was achieved by tuning the concentration of TXO-TPA.



**Figure 6** (a) The absorption spectra of TXO-TPA and PL spectra of SDPS-4PhCz in pure film; (b) The PL spectra of SDPS-4PhCz: TXO-TPA film with different doping concentrations; (c) The CIE coordinates alignment of SDPS-4PhCz and TXO-TPA used in the WOLED.

Based on the above results, we fabricated a simple white EL device via solution method with a structure of ITO/PEDOT:PSS-PFI (30 nm)/x wt% TXO-TPA:SDPS-4PhCz (60 nm)/TMPYPB (65 nm)/LiF (1 nm)/Al (100 nm), where PEDOT:PSS-PFI served as the hole-injection layer, TMPYPB served as electron-transporting layer, and TXO-TPA worked as orange emitter (Fig. S5). All devices start to emit at 3.1–3.3 V, and display intense white light ( $L_{max} = 3000$ –8000 cd/m<sup>2</sup>). The device with 0.2 wt% TXO-TPA afforded an EQE of 10.4% and a CIE coordinates of (0.35, 0.40), very close to the pure white emission. More significantly, high CRI value of 71 of WOLEDs was achieved. When TXO-TPA concentration was 1.0 wt%, the optimized performance of WOLEDs was achieved with EQE, CE and PE of 17.6%, 45.6 cd/A, 40.9 lm/W, respectively (Fig. 7 and Table 2). The CIE coordinates was calculated to be (0.39, 0.44). These results are significantly higher than previously reported single-EML WOLED device by solution process (Table 3).



Figure 7 The performances of WOLED: (a) current density and luminance versus voltage (J-V-L) curves; (b) current efficiency

and power efficiency versus	luminance curves;	(c) EQE versus	luminance curves	(d) EL spectra	of WOLED
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Concentration	$V_{on}$	CE	PE	Luminacaanaa	EQE	CIE	CRI
	(V)	(cd/A)	(lm/W)	Luminescence	(%)	$@1000cd/m^2$	@1000cd/m <sup>2/</sup> /max
0.2 wt%	3.1	24.06	22.90	3000	10.40	(0.35,0.40)	63/72
0.5 wt%	3.3	33.73	30.28	4000	13.89	(0.39,0.42)	61/65
1.0 wt%	3.2	45.56	40.90	8000	17.57	(0.39,0.44)	58/64

Table 2 The EL parameters of the WOLEDs with different concentrations of TXO-TPA.

EML	CE	PE	EOE(0/)	CIE	CDI	Ref	
(blue/orange)	$(cd A^{-1})$	(lm W <sup>-1</sup> )	EQE (%)	CIE	CRI		
DMAC-TRA/Ir(dpm)P2	28.80	18.10	4.71	(0.38, 0.44)	67	48	
5CzOXD/4CzPNPh	21.60	26.10	7.20	(0.35, 0.44)	62	15	
B-G2:Ir(Fipy-CF <sub>3</sub> ) <sub>3</sub>	59.40	58.8	19.60	(0.44,0.45)	43	49	
G2/ Ir(bt) <sub>2</sub> (acac)	17.69	7.88	10.10	(0.32, 0.33)	-	50	
G0/CN-DPASDB	17.00	15.60	6.45	(0.32, 0.37)	64	3	
tBuCN-FIrpic-Mcp/	12 50	15 70	20.60	(0.25, 0.25)	70	51	
m-CF <sub>3</sub> DPQ) <sub>2</sub> Ir(pic)	45.50	15.70	20.00	(0.35, 0.35)	/8	51	
PTPATPPO/Ir(2-phq)2acac	10.50	7.20	6.10	(0.40,0.34)	-	52	
interface exciplex/	21.10	20.71	10.02	(0.32.0.33)	85	16	
ТХО-ТРА	21.10	20.71	10.02	(0.32,0.33)	85	10	
SDPS-4PhCz/	45.60	40.90	17.60	(0.39, 0.44)	64	This work	
1.0 wt% TXO-TPA	45.00	40.20	17:00		04	THIS WORK	
SDPS-4PhCz/	24.10	22.00	10.40	(0.25.0.40)	71	This worl-	
0.2 wt% TXO-TPA	24.10	22.90	10.40	(0.55,0.40)	/1	THIS WOLK	

Table 3 The representative solution-processed two-color WOLEDs.

#### 4. Conclusions

In summary, the broad and continuous emission covering the full visible lighting spectral region was required in high efficiency and high CRI WOLEDs. A new deep blue spiro-diphenylsulfone was reported, non-doped single inverted EL device fabricated with SDPS-4PhCz showed EQE of nearly 5% with a low efficiency roll-off. Steady and transient photophysical characterization combined with quantum-chemical calculations confirm that fast RISC process can occur via a higher excited triplet state  $T_3$ , the "hot exciton" mechanism is responsible for the short delayed lifetimes. The best WOLED device was fabricated by using SDPS-4PhCz as deep blue emitter and simultaneously as the host of an orange TADF emitter TXO-TPA by solution process, which realized intense white light ( $L_{max} = 8000$  cd/m<sup>2</sup>), a maximum EQE of 17.6%, a maximum current efficiency of 45.56 cd/A and a maximum power efficiency of 40.90 lm/W.

Keywords: White organic light-emitting diodes, Thermally activated delayed fluorescence, Deep blue, Hot exciton, Solution-processed

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#### **Conflict of interest**

The authors declare that they have no conflict of interest.

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## Highlights

- A new deep blue emitter via a spiro concept was designed.
- The non-doped deep blue OLED showed a low efficiency roll-off.
- The fast RISC process in SDPS-4PhCz coincided with "hot exciton" mechanism.
- The two-color WOLED showed an EQE above 17% by spin-coating method.

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#### **Author contributions**

Ying Wang supervised the project.

Ruifang Wang carried out the experiments, analyzed the data and wrote the manuscript.

Yanwei Liu helped for the devices fabrication.

Taiping Hu and Yuanping Yi helped for the theoretical calculations.

Pengfei Wang, Xiaofang Wei, Jianjun Liu, Zhiyi Li and Xiaoxiao Hu helped for the theoretical analysis of photophysical properties.