Electronic Supplementary Information for

Highly Efficient White Light-Emitting Diodes with a bi-Component Emitting Layer based on a Blue and Yellow Thermally Activated Delayed Fluorescence Emitters

Yongzhen Chen,[a,b] Shengyong Li,[c] Taiping Hu,[b] Xiaofang Wei,[a,b] Zhiyi Li,[a,b] Yanwei Liu,[b] Jianjun Liu,[a,b] Ruifang Wang, [a,b] Yuanping Yi,[d] Cuihua Zhao,[c] Ying Wang,*[a,b] Pengfei Wang[a,b]*

[a] Key Laboratory of Photochemical Conversion and Optoelectronic Materials, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing, 100190, P. R. China

[b] University of Chinese Academy of Sciences, Beijing, 100049, P. R. China

[c] School of Chemistry and Chemical Engineering, Key Laboratory of Special Functional Aggregated materials, Ministry of Education, Shandong University, Jinan, 250100, P. R. China

[d] Beijing National Laboratory for Molecular Sciences, Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing, 100190, China

*Email - wangy@mail.ipc.ac.cn

KEYWORDS: white light-emitting diodes (WOLEDs), simple structure, full fluorescence, thermally activated delayed fluorescence

EXPERIMENTAL SECTION

Materials. *o,o′*-NPh_2 and 4-pheyl-4′-carbazole-9-H-thioxanthen-9-one-10,10-dioxide (TXO-PhCz4) was synthesized according to the reported procedure.[S1,S2] All of other materials used in the devices were purchased and the purities of them were higher than 99%. The hole injection material of poly(styrene sulfonic acid)-doped poly(3,4-ethylenedioxythiphene) (PEDOT:PSS, Baytron PVP A4083) was purchased from Heraeus Holding GmbH. Dipyrazino[2,3-f:2′,3′-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HATCN), 1,1-bis[4-[N,N′-di(p-tolyl)amino]phenyl]cyclohexane (TAPC), 1,3-bis(carbazol-9-yl)benzene (mCP), 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) and 2,2′,2″-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) were purchased from Jilin Optical and Electronic Materials Co. Ltd. Besides, bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO) and N,N′-bis(naphthalen-1-yl)-N,N′-bis(phenyl)-2,2′-dimethylbenzidine (α-NPD) were purchased from Luminescence Technology Corp.

Device Fabrication. Organic light-emitting devices were fabricated on glass substrates precoated with 150 nm ITO. The substrates should be cleaned with detergent, ultra-sonicated in water, acetone and ethyl alcohol firstly, and subsequently dried at 75 ℃ (15 min) in an oven. Afterwards the substrates were exposed to oxygen plasma (10 min) in order to remove organic residues and improve the work function of ITO. The PEDOT:PSS aqueous solution was spin-coated onto the precleaned substrates and then annealed at 120 ℃ for 30 min under nitrogen environment. Then the substrates were transferred to a thermal evaporation chamber with a pressure lower than 4×10^{-4} Pa for organic semiconductor layers and metal cathode deposition.

PL Characterization and Device Measurement. The ultraviolet-visible (UV-vis) spectra and fluorescence spectra were obtained with Hitachi U-3900 and F-4600 spectrophotometers, respectively. The phosphorescence spectra were measured in 2-MeTHF glass matrix at 77 K using a Hitachi F-4600 fluorescence spectrometer. The absolute fluorescence quantum yields of the solid films were measured with an integrating sphere. The transient PL decay characteristics were measured using an

Edinburgh Instruments FLS920 spectrometer. The electroluminescence (EL) spectra were measured by a Spectra scan PR655 photometer. The current−voltage−brightness characteristics were measured by using a computer-controlled Keithley source measurement unit (Keithley 2400) with a Konica Minolta CS-200 luminance-meter under dark and ambient atmosphere. External quantum efficiencies (EQE, %), current efficiencies (cd/A) and power efficiencies (lm/W) were calculated from the electrical and optical properties. For this calculation we assumed Lambertian light distribution.

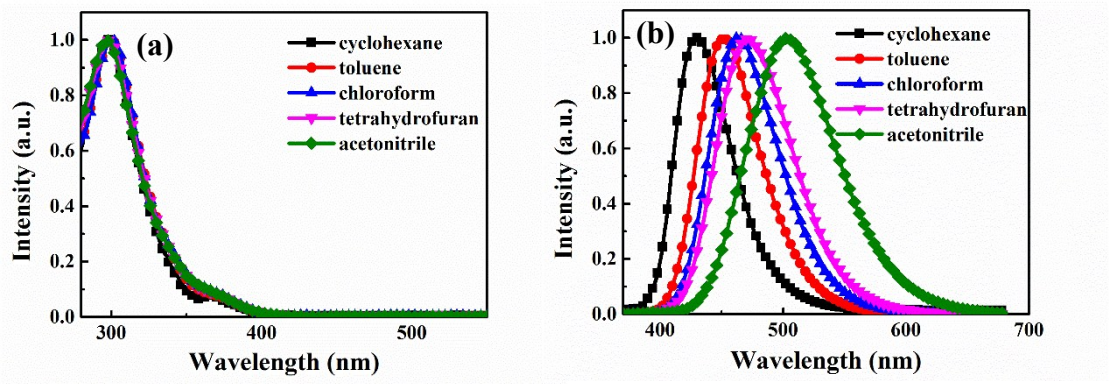
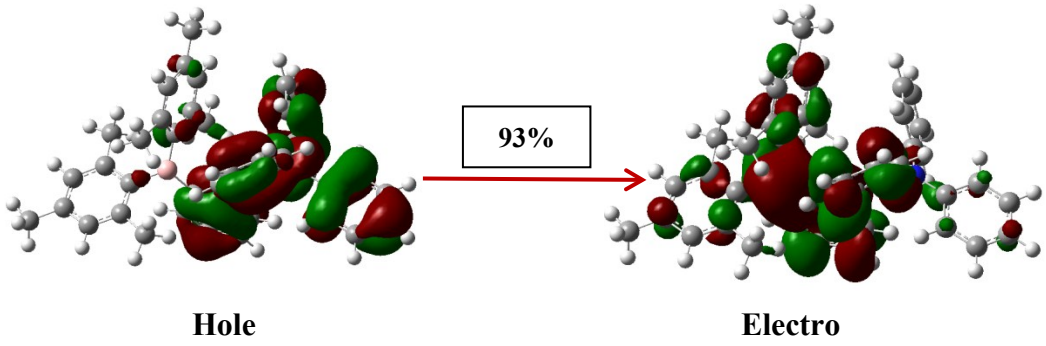


Figure S1. Normalized absorption (a) and PL spectra (b) of *o,o′*-NPh_2 in different solvents.



Natural transition orbital

Figure S2. Highest occupied NTO and lowest unoccupied NTO according to the results of time-dependent DFT for the S_1 state of *o,o′*-NPh_2 using the optimized structure of the S_0 state.

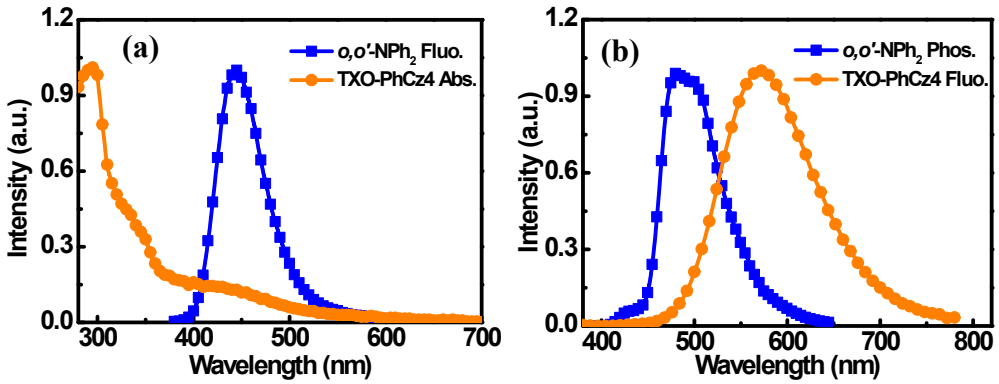


Figure S3. (a) The overlap between absorption spectrum of TXO-PhCz4 and PL spectrum of *o,o′*-NPh_2. (b) The phosphorescence spectrum of *o,o′*-NPh_2 and fluorescence spectrum of TXO-PhCz4.

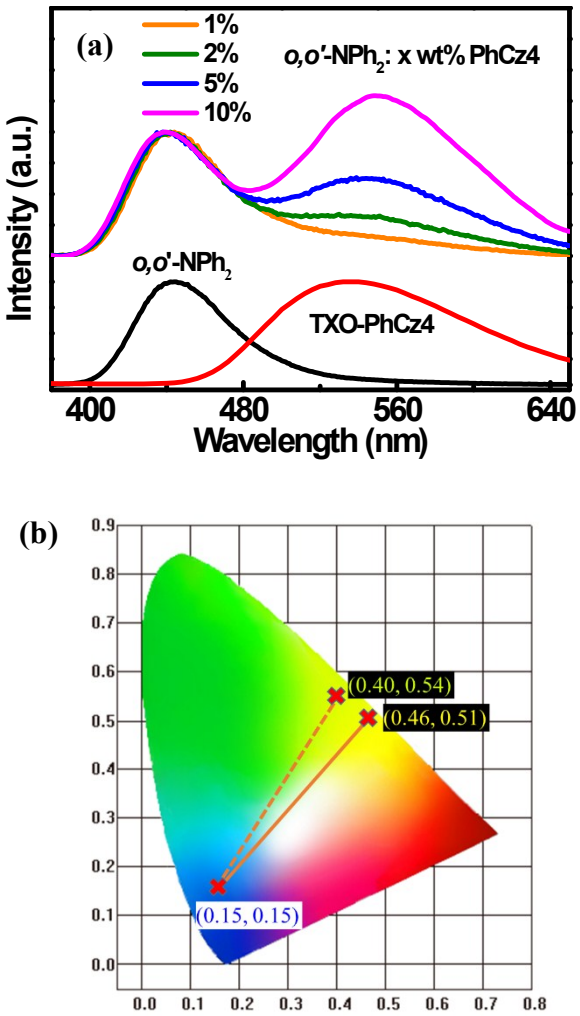


Figure S4. (a) The PL spectra of *o,o'*-NPh_2:TXO-PhCz4 film with different doping concentration. (b) The CIE coordinates variation of TXO-PhCz4 doped in PMMA (dashed) and *o,o'*-NPh_2 (solid).

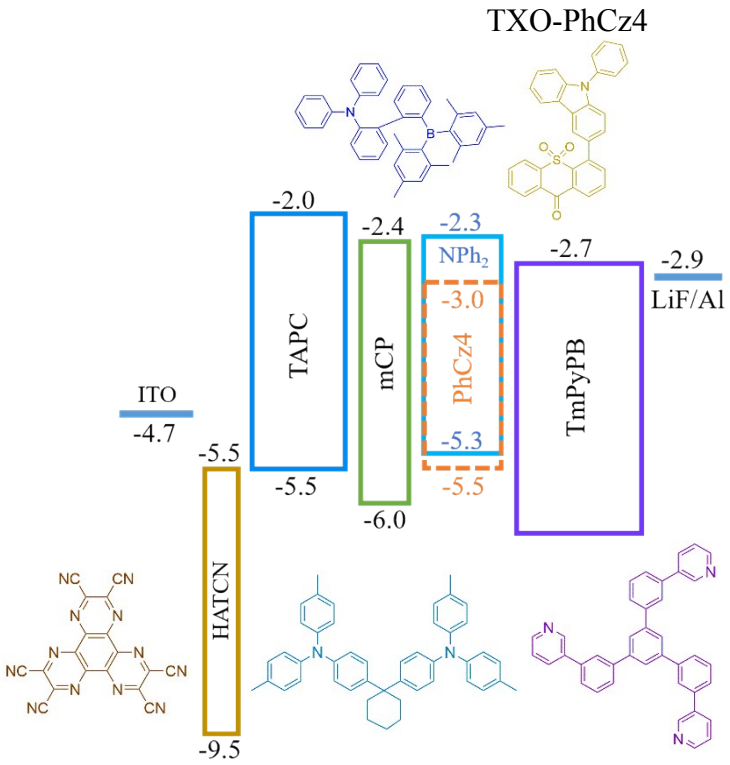


Figure S5. Molecular structures and energy levels of WOLEDs based on bi-component TADF EML with simple structure.

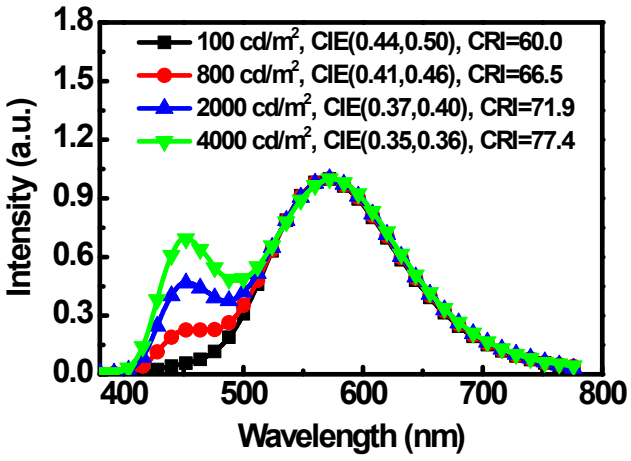


Figure S6. EL spectra of WOLED without the TSPO1 buffer layer at different luminance.

Table S1. Photophysical data extracted from the transient PL spectra of DPEPO: 10 wt% *o,o'*-NPh_2 at room temperature.

$\emptyset_{total}$ (%)	$\emptyset_p$ (%)	$\emptyset_d$ (%)	τ_p (ns)	τ_d (μs)	k_p ($\times 10^7$ s^{-1}) [a]	k_d ($\times 10^3$ s^{-1}) [a]	k_{RISC} ($\times 10^3$ s^{-1}) [a]
34.5	33.7	0.8	9.17	1.57	3.7	5.0	1.7

[a]Calculated according to the reported method.[S3]

Table S2. Summary of the performance of WOLEDs based on TADF emitters and on bi-component EML.

Type	EML	V_{on} (V)	EQE_{max} (%)	CE_{max} (cd/A)	PE_{max} (lm/W)	CIE(x, y)	CRI	Ref.
bi-Component Emitting Layer system	*o,o'*-NPh_2:1 wt% TXO-PhCz4	3.5	12.5	30.2	27.1	(0.38,0.40)	71	This work
	4P-NPD:2 wt% $Ir(dhfpy)_2(acac)$	–	5.5	–	10.5	(0.45, 0.43)	38	S4
	CPhBzIm:0.1 wt% $(pbi)_2Ir(acac)$	2.5	7.0	15.5	12.8	(0.31, 0.33)	–	S5
	DADBT:0.1 wt% $Ir(2\text{-}phq)_3$	2.4	26.6	53.5	67.2	(0.41, 0.43)	–	S6
	4P-NPD:8 wt% FPtOPhND	4.0	7.4	13.0	6.7	(0.36, 0.28)	81	S7
	$Bepp_2$:0.1 wt% $(fbi)_2Ir(acac)$	–	–	4.8	5.0	(0.27, 0.25)	88	S8
	$Bepp_2$:1.0 wt% $(fbi)_2Ir(acac)$	–	–	37.6	44.9	(0.47, 0.48)	39	S8
	TPA-SO2:0.5 wt% PO-01	2.5	13.2	39.6	43.0	(0.42, 0.46)	–	S9
	BBPI:1.0 wt% $Ir(2\text{-}phq)_3$	2.6	23.8	56.1	62.9	(0.49, 0.41)	–	S10
	BBPI:0.4 wt% PO-01	2.6	12.9	36.8	38.6	(0.44, 0.48)	–	S11
	DMAC-DPS:0.6 wt% Rubrene	2.5	7.4	20.2	15.9	(0.36, 0.44)	–	S12
	1,8-D:17 wt% T4AC	5.5	–	3.91	2.05	(0.30, 0.43)	56	S13
	5CzOXD:4CzPNPh	2.4	7.2	21.6	26.1	(0.35, 0.44)	62	S14
	DMAC-DPS:0.2 wt% TBRb	2.8	14.0	45.1	48.0	(0.37, 0.48)	–	S15
TADF-based Emitting Layer system	DPEPO:DMAC-DPS:0.2 wt% TBRb	2.8	14.6	44.4	51.6	(0.34, 0.47)	–	S15
	CzAcSF:TBPe:0.2 wt% TBRb	< 3	15.2	–	–	(0.23, 0.30)	–	S16
	DPEPO:50 wt% DMAC-DPS:0.03 wt% TBRb	< 3	17.6	39.3	41.0	(0.23, 0.31)	44.7	S17
	DPEPO:50 wt% DMAC-DPS:0.05 wt% TBRb	< 3	15.5	38.4	39.3	(0.28, 0.35)	58.6	S17
	CDBP:PO-T2T:7.5 wt% 2CzPN:0.6 wt% AnbCz	2.3	19.0	50.1	63.0	(0.34, 0.44)	63	S18
	mCP:20 wt% 2CzPN: 0.2 wt% PO-01	–	19.6	–	50.2	(0.42, 0.48)	53	S19
	PPF:6wt% CC2BP/mCPB:18 wt% Px2BBP	5.0	6.7	16.4	–	(0.32, 0.39)	–	S20
	mCBP: 10 wt% 4CzPN/mCBP: 6 wt% 4CzPN:2	3.6	17.6	38.6	30.3	(0.26, 0.38)	–	S21

	wt% 4CzTPN-Ph/PPT: 10 wt% 3CzTRZ							
	mCP:1 wt% DBP:15 wt% TPPA/mCP/DMAC-DPS	3.2	12.3	–	18.9	(0.23, 0.30)	–	S22
	mCP:5wt% TXO-TPA/mCP: 2 wt% 4P-NPB	4.0	4.4	10.9	8.5	(0.37, 0.42)	–	S23
	DMAC-DPS:0.5 wt % DBP/DMAC-DPS:1.0 wt% DBP:0.5 wt% TTPA/DMAC-DPS:0.2 wt% TTPA	2.8	18.2	40.9	44.6	(0.32, 0.39)	82	S24
	CBP:7 wt% NI-1-PhTPA/CBP/CBP:3 wt% PXZDSO2/CBP:5 wt% PXZDSO2:0.3 wt% DBP/CBP:3 wt% PXZDSO2/CBP/CBP:7 wt% NI-1-PhTPA	3.4	19.2	51.4	47.5	(0.35, 0.45)	69	S25
	CBP:6 wt% $Ir(pq)_2acac$/4P-NPD/CBP:4 wt% 4CzIPN	–	11.2	–	26.2	(0.41, 0.46)	81.5	S26

Reference

(S1) H. Pan, G.-L. Fu, Y.-H. Zhao and C.-H. Zhao, *Org. Lett.*, 2011, **13**, 4830-4833.

(S2) X. Wei, Y. Chen, R. Duan, J. Liu, R. Wang, Y. Liu, Z. Li, Y. Yi, Y.-T. Yukiko, P. Wang and Y. Wang, J. Mater. Chem. C, 2017, DOI: 10.1039/C7TC04025C.

(S3) Q. Zhang, B. Li, S. Huang, H. Nomura, H. Tanaka, C. Adachi, *Nat. Photon.,* 2014, **8**, 326.

(S4) G. Schwartz, S. Reineke, T. C. Rosenow, K. Walzer and K. Leo, *Adv. Funct. Mater.*, 2009, **19**, 1319-1333.

(S5) W.-Y. Hung, L.-C. Chi, W.-J. Chen, Y.-M. Chen, S.-H. Chou and K.-T. Wong, *J. Mater. Chem.*, 2010, **20**, 10113.

(S6) J. Ye, C. J. Zheng, X. M. Ou, X. H. Zhang, M. K. Fung and C. S. Lee, *Adv. Mater.*, 2012, **24**, 3410-3414.

(S7) A. Poloek, C.-W. Lin, C.-T. Chen and C.-T. Chen, *J. Mater. Chem. C*, 2014, **2**, 10343-10356.

(S8) B. Liu, M. Xu, L. Wang, J. Zou, H. Tao, Y. Su, D. Gao, H. Ning, L. Lan and J. Peng, *Org. Electron.*, 2014, **15**, 2616-2623.

(S9) Y. Li, Z. Wang, X. Li, G. Xie, D. Chen, Y.-F. Wang, C.-C. Lo, A. Lien, J. Peng, Y. Cao and S.-J. Su, *Chem. Mater.*, 2015, **27**, 1100-1109.

(S10) X. Ouyang, X. L. Li, L. Ai, D. Mi, Z. Ge and S. J. Su, *ACS Appl. Mater.*

Interfaces, 2015, **7**, 7869-7877.

(S11) X. L. Li, X. Ouyang, D. Chen, X. Cai, M. Liu, Z. Ge, Y. Cao and S. J. Su, *Nanotechnology*, 2016, **27**, 124001.

(S12) B. Zhao, T. Zhang, W. Li, Z. Su, B. Chu, X. Yan, F. Jin, Y. Gao and H. Wu, *Org. Electron.*, 2015, **23**, 208-212.

(S13) J. Ye, C. Liu, C. Ou, M. Cai, S. Chen, Q. Wei, W. Li, Y. Qian, L. Xie, B. Mi, Z. Gao and W. Huang, *Adv. Opt. Mater.*, 2014, **2**, 938-944.

(S14) D. Zhang, X. Cao, Q. Wu, M. Zhang, N. Sun, X. Zhang and Y. Tao, *J. Mater. Chem. C*, 2018, DOI: 10.1039/c7tc04969b.

(S15) Z. Wu, L. Yu, X. Zhou, Q. Guo, J. Luo, X. Qiao, D. Yang, J. Chen, C. Yang and D. Ma, *Adv. Opt. Mater.*, 2016, **4**, 1067-1074.

(S16) W. Song, I. Lee and J. Y. Lee, *Adv. Mater.*, 2015, **27**, 4358-4363.

(S17) W. Song, I. H. Lee, S.-H. Hwang and J. Y. Lee, *Org. Electron.*, 2015, **23**, 138-143.

(S18) W. Liu, C. J. Zheng, K. Wang, M. Zhang, D. Y. Chen, S. L. Tao, F. Li, Y. P. Dong, C. S. Lee, X. M. Ou and X. H. Zhang, *ACS Appl. Mater. Interfaces*, 2016, **8**, 32984-32991.

(S19) D. Zhang, L. Duan, Y. Zhang, M. Cai, D. Zhang and Y. Qiu, *Light: Sci. Appl.*, 2015, **4**, e232.

(S20) S. Y. Lee, T. Yasuda, Y. S. Yang, Q. Zhang and C. Adachi, *Angew. Chem. Int. Ed. Engl.*, 2014, **53**, 6402-6406.

(S21) J.-i. Nishide, H. Nakanotani, Y. Hiraga and C. Adachi, *Appl. Phys. Lett.*, 2014, **104**, 233304.

(S22) T. Higuchi, H. Nakanotani and C. Adachi, *Adv. Mater.*, 2015, **27**, 2019-2023.

(S23) L. Meng, H. Wang, X. Wei, X. Lv, Y. Wang and P. Wang, *RSC Adv.*, 2015, **5**, 59137-59141.

(S24) Z. Wu, Q. Wang, L. Yu, J. Chen, X. Qiao, T. Ahamad, S. M. Alshehri, C. Yang and D. Ma, *ACS Appl. Mater. Interfaces*, 2016, DOI: 10.1021/acsami.6b10087.

(S25) X. L. Li, G. Xie, M. Liu, D. Chen, X. Cai, J. Peng, Y. Cao and S. J. Su, *Adv. Mater.*, 2016, **28**, 4614-4619.

(S26) S. E. Lee, H. W. Lee, H. J. Baek, T. J. Yun, G. J. Yun, W. Y. Kim and Y. K. Kim, *J. Photon. Energy*, 2016, **6**, 042507.