Supporting Information for

Organic Upconversion Display with an over 100% Photon-to-photon Upconversion Efficiency and a Simple Pixelless Device Structure

Qiaogang Song^{a,b,c}, Tong Lin^{b,c}, Zisheng Su^{a,b*}, Bei Chu^b, Huishan Yang^{a*}, Wenlian Li^b,

Chun-Sing Leed*

^a College of Physics and Information Engineering, Key Laboratory of Information

Functional Material for Fujian Higher Education, Quanzhou Normal University,

Quanzhou 362000, P. R. China

^b State Key Laboratory of Luminescence and Applications, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, P.R. China

^c University of Chinese Academy of Sciences, Beijing 100049, P. R. China

^d Center of Super-Diamond and Advanced Films (COSDAF) and Department of Chemistry, City University of Hong Kong, Hong Kong SAR, P. R. China

*E-mail: <u>suzs@ciomp.ac.cn</u> (Zisheng Su); <u>yanghuishan1697@163.com</u> (HuishanYang); apcslee@cityu.edu.hk (Chun-Sing Lee)

1. Calculation of the responsivity and detection external quantum efficiency of the devices The responsivity (*R*) of the devices is calculated with the formula:

$$R = \frac{J_{ph}}{P_{in}} = \frac{J_{total} - J_{dark}}{P_{in}} \tag{1}$$

where the J_{ph} is the photocurrent density of the NIR photodetector under illumination, J_{total} is the total current density measured under illumination, J_{dark} is the dark current, and P_{in} is the incident power density of the NIR laser. Then the detection external quantum efficiency (EQE_{det}) can be expressed as:

$$EQE_{det} = \frac{hc}{\lambda} \times R = \frac{1240}{\lambda} \times R$$
(2)

where *h* is the Planck constant, *c* is the speed of light, and λ is the wavelength of the incident NIR laser. The J_{total} of the NIR photodetector ITO/PbPc (60 nm)/C₆₀ (65 nm)/Al (100 nm) under incident of an 808 nm NIR laser with a power density of 0.052 mW/cm² and the J_{dark} at - 10 V are 17.79 and 11.19 mA/cm², respectively. Thus *R* of the NIR photodetector is calculated to be 127 A/W. Correspondingly, the EQE_{det} is 1.95×10⁴ %.

2. Calculation of the emission external quantum efficiency of the devices

The emission external quantum efficiency (EQE $_{em}$) is determined by the ratio between

the numbers of emitted photons (n_p) and injected electrons (n_e) , which can be

calculated with:

$$EQE_{em} = \frac{n_p}{n_e}$$

The n_e can be obtained from the measured current (I_c) of the devices:

$$n_e = \frac{I_c}{q_0}$$

where q_0 the charge of elementary carrier. The n_p can be expressed as:

$$n_{p} = \int n(\lambda) d\lambda = \alpha \int I(\lambda) d\lambda$$
(5)

where $n(\lambda)$ is the number of emitted photons at a single wavelength λ , $I(\lambda)$ is the relative emission intensity at a single wavelength λ which can be obtained from the EL

spectrum of the device, and α is a factor between $n(\lambda)$ and $I(\lambda)$. The relationship of

photon flux (*F*) and irradiation flux $P(\lambda)$ is:

$$F = \int K_m \phi(\lambda) P(\lambda) d\lambda$$

(6)

where K_m is photo-power equivalent and equates 683 Im/W, $\phi(\lambda)$ is visible function. Meanwhile, the relationship between *F* and luminance (*L*) is:

$$L = \frac{dF}{S\cos\theta d\Omega}$$

(7)

where *S* is emission area of the device, θ is the angle between the measurement direction and vertical direction of emission area, and Ω is the space angle. In our devices, θ is 0 and Ω equates π due to the bottom emission of the devices. Then the relationship between *F* and *L* is:

 $F = LS\pi$

(8)

In addition:

$$P(\lambda) = h \upsilon n(\lambda) = \frac{hc}{\lambda} n(\lambda)$$

(9)

where v is photon frequency. From expressions (5), (6), (8), and (9) we obtain:

$$\int K_m \phi(\lambda) \frac{hc}{\lambda} n(\lambda) d\lambda = \int K_m \phi(\lambda) \frac{hc}{\lambda} \alpha I(\lambda) d\lambda = \pi SL$$

(10)

Then the factor α can be expressed as:

$$\alpha = \frac{\pi SL}{K_m hc} \frac{1}{\int \frac{\phi(\lambda)I(\lambda)}{\lambda} d\lambda}$$

(11)

and the emitted photon number is:

$$n_{p} = \frac{\pi SL}{K_{m}hc} \frac{\int I(\lambda)d\lambda}{\int \frac{\phi(\lambda)I(\lambda)}{\lambda}d\lambda}$$

(12)

Then the $\mathsf{EQE}_{\mathsf{em}}$ can be calculated with the formula:

$$EQE_{em} = \frac{LS}{I_c} \frac{\pi q_0}{K_m hc} \frac{\int I(\lambda) d\lambda}{\int \frac{\phi(\lambda)I(\lambda)}{\lambda} d\lambda}$$

(13)

Thus we can obtain the EQE_{em} from the measured current I_c , luminance L, device area

S, and EL spectrum (determines $I(\lambda)$).

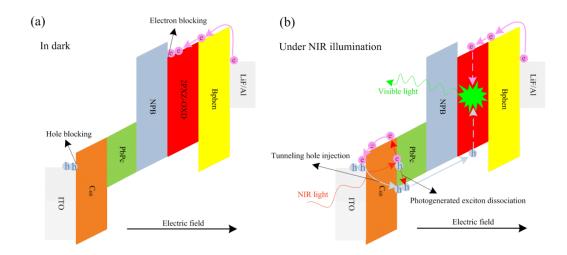


Figure S1. Schematic operation principles of the upconverter in dark and under NIR illumination.

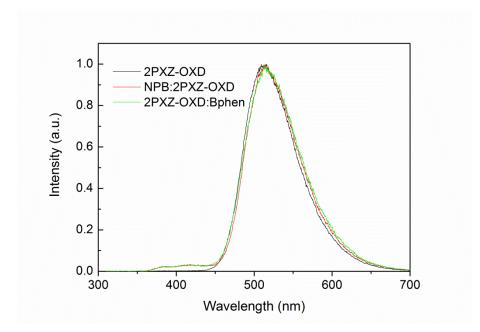


Figure S2. Normalized PL spectra from 30 nm films of 2PXZ-OXD, NPB:2PXZ-OXD (1:1), and 2PXZ-OXD:Bphen (1:1). The similar PL spectra confirm that 2PXZ-OXD forms exciplex with neither NPB nor Bphen.

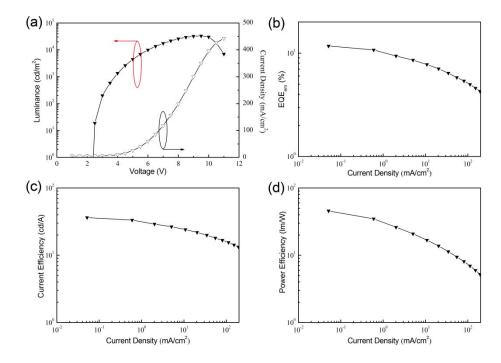


Figure S3. (a) Luminance-current density-voltage, (b) EQE-current density, (c) current efficiency-current density, and (d) power efficiency-current density characteristics of the non-doped TADF-OLED with a structure of ITO/NPB (30 nm)/2PXZ-OXD (30 nm)/Bphen (30 nm)/LiF (1 nm)/Al (100 nm).

Table S1. Comparison of the turn-on voltage (V_T), luminance (L), and emission external quantum efficiency (EQE_{em}) of the OLED in this work to the other reported state-of-the-art non-doped TADF-OLEDs.

Device	V _T	L	EQE_{em}	Ref.	
	[V]	[cd/m ²]	[%]		
ITO/NPB/2PXZ-OXD/Bphen/LiF/AI	2.4	199@3 V	11.7	This work	
ITO/MoO ₃ /mCP/DMAC-BP/TPBi/LiF/AI	2.6	100@3 V	18.9	[1]	
ITO/PEDOT:PSS/TAPC/mCP/DMAC-TRZ/DPPs/3TPYMB/LiF/AI	3.0	100@~4.5V	20	[2]	
ITO/TAPC/DBT-BZ-DMAC/TmPyPB/LiF/AI	2.7	100@~3.4V	14.2	[3]	
ITO/TAPC/DBT-BZ-PXZ/TmPyPB/LiF/AI	2.7	100@~3.6V	9.7	[4]	

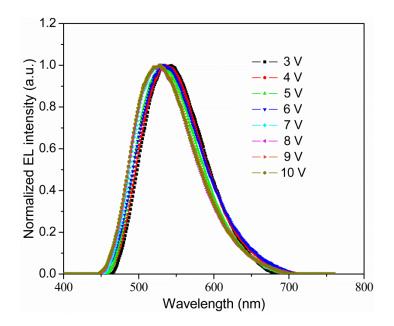


Figure S4. Normalized EL spectra of the TADF-OLED ITO/NPB (30 nm)/2PXZ-OXD (30

nm)/Bphen (30 nm)/LiF (1 nm)/Al (100 nm) at different applied voltages.

Table S2. Turn-on voltage (V_7), luminance (L), emission external quantum efficiency

(EQE_{em}), detection external quantum efficiency (EQE_{det}), photon to photon conversion

efficiency (η_{photon}), and power to power conversion efficiency (η_{power}) of the NIR-

upconverter under illumination of an 808 nm laser with different intensity.

Illumination											
intonoitu	V_{T}	La	Lb	EQE_{em}^{a}	$EQE_{em}{}^{b}$	EQE_{det}^{a}	EQE_{det}^{b}	$\eta_{ ext{photon}}{}^{ ext{a}}$	$\eta_{ ext{photon}}$ b	$\eta_{power}{}^{a}$	η_{power} b
intensity [mW/cm ²]	[V]	[cd/m ²]	[cd/m ²]	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[%]

0	7.5	-	116	-	2.5	-	-	-	-	-	-
0.052	3.1	0.9	271	5.9	1.7	14.4	14733	0.8	256	1.3	398
0.10	2.5	1.8	282	5.5	1.8	16.3	7436	0.9	138	1.4	214
0.33	2.4	5.0	288	5.6	1.9	12.5	2306	0.7	43	1.1	66.3
1.09	2.3	13.1	319	6.7	1.9	8.7	740	0.6	14.4	0.9	22.3
2.13	2.2	39.5	546	8.6	2.8	10.6	440	0.9	12.6	1.4	19.5
3.09	2.2	93.7	991	9.4	9.4	15.8	467	1.5	15.7	2.3	24.4
4.91	2.0	196	1903	8.3	4.4	23.7	432	2.0	19.0	3.0	29.5
9.06	2.0	210	2893	7.8	5.2	14.6	279	1.2	15.6	1.8	24.3
18.68	2.0	294	4321	5.9	4.1	13.1	276	0.8	11.3	1.2	17.6
33.75	2.0	307	4705	5.6	3.8	7.9	179	0.4	6.8	0.7	10.6
52.46	2.0	329	5870	5.8	3.9	5.3	142	0.3	5.5	0.5	8.5

^{a)} at voltage of 3 V; ^{b)} at voltage of 15 V

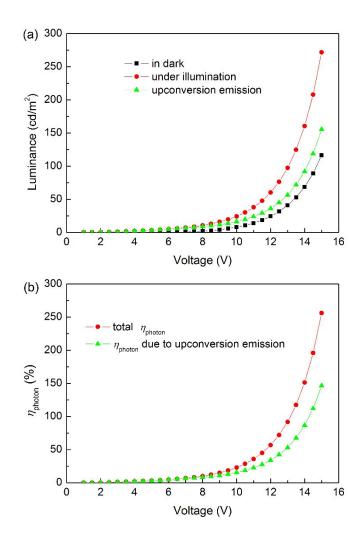


Figure S5. (a) Luminance of the the NIR-upconverter in dark (\blacksquare), under illumination of NIR laser with an intensity of 0.052 mW/cm² (\bullet), and the upconversion emission (\blacktriangle obtained by \bullet - \blacksquare); (b) Total photon to photon conversion efficiency (η_{photon}) (\bullet) and η_{photon} due to the upconversion (\blacktriangle) of the NIR-upconverter.

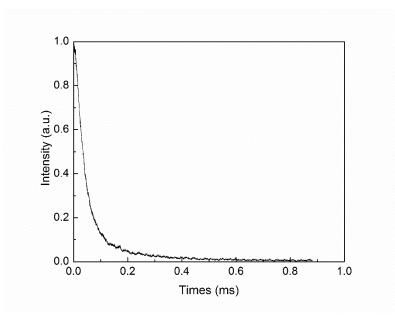


Figure S6. Response speed of the upconversion emission of the device at 3 V to the

excitation of an 808 laser.

References:

 Zhang, Q.; Tsang, D.; Kuwabara, H.; Hatae, Y.; Li, B.; Takahashi, T.; Lee, S. Y.; Yasuda, T.; Adachi, C. Nearly 100% internal quantum efficiency in undoped electroluminescent devices employing pure organic emitters. *Adv. Mater.* 2015, *27*, 2096-2100.

- (2) Tsai, W-L.; Huang, M.-H.; Lee, W.-K.; Hsu, Y.-J.; Pan, K.-C.; Huang, Y.-H.; Ting, H.-C.; Sarma, M.; Ho, Y.-Y.; Hu, H.-C.; Chen, C.-C.; Lee, M.-T.; Wong, K.-T.; Wu, C.-C. A versatile thermally activated delayed fluorescence emitter for both highly efficient doped and non-doped organic light emitting devices. *Chem. Commun.* **2015**, *51*, 13662-13665.
- (3) Guo, J.; Li, X.-L.; Nie, H.; Luo, W.; Gan, S.; Hu, S.; Hu, R.; Qin, A.; Zhao, Z.; Su, S.-J.; Tang, B. Z. Achieving high-performance nondoped OLEDs with extremely small efficiency roll-off by combining aggregation-induced emission and thermally activated delayed fluorescence. *Adv. Funct. Mater.* 2017, *27*, 1606458.
- Guo, J; Li, X.-L.; Nie, H.; Luo, W.; Hu, R.; Qin, A.; Zhao, Z.; Su, S.-J.; Tang, B. Z.
 Robust luminescent materials with prominent aggregation-induced emission and thermally activated delayed fluorescence for high-performance organic light-emitting diodes. *Chem. Mater.* 2017, *29*, 3623-3631.