## **Supporting Information**

# Asymmetric Strategy Based on 5H-Benzofuro[3,2c]carbazole Enable Efficient Narrowband Multi-Resonance Thermally Activated Delayed Fluorescence Emitters

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#### **1. General Information**

All regents and solvents used for the synthesis of the compound were purchased from commercial source and used without further purification. Flash column chromatography was performed on glass column of silica gel (300 mesh). Highresolution mass spectra were recorded using Bruker ultraflextreme MALDI-TOF/TOF mass spectrometer. Photoluminescence spectra and Phosphorescence spectra (The time-gated window for measuring the low-temperature phosphorescence is around 25 ms.) were measured on a Hitachi F-4600 spectrometer. Absorption spectra were studied using a UV-vis spectrophotometer (TU-1901, PERSEE). The time-resolved transient photoluminescence decay characteristics were measured using an Edinburgh Instruments FLS1000 spectrometer. The absolute fluorescence quantum yields of the solid films were measured with an integrating sphere. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were measured with a Q600 Simultaneous TG-DSC instrument. The temperature of 5% weight loss was defined as the decomposition temperature ( $T_d$ ) at a heating rate of 10 °C min<sup>-1</sup> from 30 °C to 700 °C under nitrogen. The DSC curves were obtained at a heating rate of 10 °C min<sup>-1</sup> from 30 °C to 250 °C under nitrogen, and no obvious glass transition temperature was observed of BFCz-B-Cz abd BFCz-B-O.

#### 2. Electrochemical Characterization

The electrochemical measurement was made using a CHI660E voltammetric analyzer, including cyclic voltammetry (CV) and differential pulse voltammetry (DPV). The oxidation potential was determined by CV or DPV using 0.1 M tetrabutylammonium hexafluorophosphate (TBAPF<sub>6</sub>) in degassed CH<sub>2</sub>Cl<sub>2</sub>. Ferrocene was added as a calibrant after each set of measurements, and all potentials reported were quoted regarding the ferrocene-ferrocenium (Fc/Fc+) couple E(Fc/Fc<sup>+</sup>) at a scan rate of 100 mV/s. ITO, Ag/AgCl and Pt mesh were used as working electrode, reference electrode and counter electrode, respectively. DPV was conducted with an increment potential of 0.004 V and a pulse amplitude, width, and period of 50 mV, 0.05, and 0.5 s, respectively. The oxidation E<sub>O</sub> are evaluated from the first DPV and CV peak positions. HOMO levels were conducted from  $E_{HOMO} = -[4.8 + E_O - E(Fc/Fc^+)] eV$  and  $E_{LUMO} = E_{HOMO} + E_{opt}$ , therefore,  $E_{opt}$  was obtained from the absorption edges of normalized absorption spectra.

### 3. OLED Fabrication and Characterization

Before device fabrication, the ITO glass substrates were sequentially cleaned with detergents, de-ionized water, dried at 75 °C (15 min) in an oven, and treated with oxygen plasma for 10 min in order to remove organic residues and improve the work function of ITO. After that, the clean substrates were transferred into a vacuum deposition system with a pressure below  $1 \times 10^{-4}$  Pa for organic and metal deposition. The devices were fabricated by evaporating organic materials onto the substrate while LiF at a rate of 0.1 Å s<sup>-1</sup> and Al metal through a rate of 10 Å s<sup>-1</sup>. EL luminescence spectra were measured with a Spectra scan PR655 photometer and the current-voltage-brightness characteristics were measured with a computer-controlled Keithley 2400 Source Meter and CS-200 under ambient atmosphere.

#### 4. Transient photophysical measurement

The transient photoluminescence decay characteristics were measured using an Edinburgh Instruments FLS1000 spectrometer. The transient decay curves were fitted with the multi-exponential equation:

$$I(t) = \sum_{i} A_{i} exp(-\frac{t}{\tau_{i}})$$
(S1)

Where  $A_i$  is the pre-exponential factor,  $\tau_i$  is the decay time, and I(t) is the emission intensity.

The other parameters are calculated with following equation:

$\boldsymbol{\Phi}_{\mathrm{prompt}} = \boldsymbol{\Phi}_{\mathrm{PL}} R_{\mathrm{prompt}}$	(S2)
$\Phi_{\text{delayed}} = \Phi_{\text{PL}} R_{\text{delayed}}$	(\$3)
$k_{\mathrm{F}} = \boldsymbol{\varPhi}_{\mathrm{prompt}} / \tau_{\mathrm{prompt}}$	(S4)
$\Phi_{\rm PL} = k_{\rm F}/(k_{\rm F}+k_{\rm IC})$	(85)
$\Phi_{\text{prompt}} = k_{\text{F}}/(k_{\text{F}} + k_{\text{IC}} + k_{\text{ISC}})$	(S6)
$\Phi_{\rm IC} = k_{\rm IC}/(k_{\rm F} + k_{\rm IC} + k_{\rm ISC})$	(S7)
$\Phi_{\rm ISC} = k_{\rm ISC}/(k_{\rm F} + k_{\rm IC} + k_{\rm ISC}) = 1 - \Phi_{\rm prompt} - \Phi_{\rm IC}$	(S8)
${m \Phi}_{ m RISC}={m \Phi}_{ m delayed}/{m \Phi}_{ m ISC}$	(S9)
$k_{\rm RISC} = (k_{\rm p} k_{\rm d} \Phi_{\rm delayed}) / (k_{\rm ISC} \Phi_{\rm prompt})$	(S10)
$k_{\rm p} = 1/\tau_{\rm prompt}; \ k_{\rm d} = 1/\tau_{\rm delayed}$	(S11)

The  $k_{\text{RISC}}$  in TADF systems can be described using the Marcus theory expression as follows:

$$k_{RISC} = \frac{2\pi}{\hbar} |\langle S|\hat{H}_{SOC}|T\rangle|^2 \frac{1}{\sqrt{4\pi\lambda_{RISC}k_BT}} exp^{(i)} \left(\frac{-\Delta E_{ST}}{k_BT}\right)$$
(S12)

Where  $\langle S|\hat{H}_{SOC}|T\rangle$  denotes the spin-orbit coupling (SOC) matrix element,  $\lambda_{RISC}$  is the reorganization energy for RISC,  $k_{\rm B}$  is the Boltzmann constant, T is the absolute temperature, and  $\Delta E_{\rm ST}$  is the activation energy of the RISC.

#### 5. Quantum Chemical Calculations

The ground state structures were optimized using the Gaussian 09 program package. The electron density distribution of frontier molecular orbital (FMO) were visualized with Gaussview 5.0. Structure optimizations were carried out using density functional theory (DFT) calculations. The ground state structures were optimized with B3LYP/def2-svp level. Natural transition orbital (NTO) analysis for the S<sub>1</sub> and T<sub>m</sub> states are analyzed at the B3LYP/def2-svp level and visualized with Multiwfn  $3.8^{[1]}$  and VMD  $1.9.3^{[2]}$ . The energy levels for S<sub>1</sub> and T<sub>1</sub> were calculated at SCS-CC2/cc-pVDZ level, using the MRCC program <sup>[3,4]</sup>. The spin-orbital coupling (SOC) was also calculated by the TD-DFT method at the B3LYP/def2-svp level using ORCA 5.0 software package.

#### 6. Synthesis

All reagents and chemicals were purchased from commercial sources and used without further purification. Compounds 1 and 2 were prepared according to the literature 5 and 6. Compound tCzBN was prepared according to the literature 7. The final products were firstly purified by column chromatography, and then further refined by temperature-gradient vacuum sublimation.



Scheme S1. Synthetic routes of BFCz-B-Cz and BFCz-B-O.

Synthesis of 2: A solution of BFCz (2.2 g, 8.8 mmol), cesium carbonate (5.7 g, 17.6 mmol) and 1 (4.0 g, 8.8 mmol) in dry DMF (80 mL) was stirred for 8 h at 130 °C. After cooling to room temperature, the reaction mixture was extracted with dichloromethane and the solvent was removed under reduced pressure. The crude product was purified by column chromatography on silica gel (petroleum ether/dichloromethane = 4:1, v/v) to give 2 as a white solid (yield = 5.7 g, 95%). <sup>1</sup>H NMR (400 MHz, Chloroform-d)  $\delta$  = 8.65 (d, J = 7.7 Hz, 1H), 8.22 (s, 2H), 8.08 – 8.02 (m, 2H), 7.78 (t, J = 9.0 Hz, 4H), 7.58 (q, J = 6.3, 5.2 Hz, 3H), 7.50 (q, J = 7.0 Hz, 2H), 7.43 (t, J = 7.3 Hz, 1H), 7.32 (d, J = 8.1 Hz, 1H), 7.21 (q, J = 8.3 Hz, 3H), 1.53 (s, 18H). <sup>13</sup>C NMR (151 MHz, CDCl<sub>3</sub>)  $\delta = 156.40, 151.29, 143.31, 141.30, 140.50, 139.97, 139.25, 139.10, 131.51, 131.08,$ 129.58, 126.12, 126.00, 125.51, 125.12, 123.92, 123.53, 123.00, 121.18, 121.07, 119.84, 118.34, 117.04, 116.59, 111.79, 109.99, 109.50, 108.91, 105.65, 34.89, 32.15. Synthesis of BFCz-B-Cz: To a solution of 2 (1 g, 1.4 mmol) in m-Xylene (10 ml) was slowly added n-BuLi (2.5 M in hexane, 1.7 mL, 4.3 mmol) at 0 °C. After heating for 4 h at 60 °C, BBr<sub>3</sub> (1.1 g, 4.3 mmol) was slowly added at -30 °C, and then the mixture was stirred at room temperature for 2 h. After addition of DIPEA (1 mL) at 0 °C, the reaction mixture was further stirred at 150 °C for 12 h. After cooling to room temperature, the reaction mixture was carefully quenched by addition of water. The reaction mixture was extracted with dichloromethane and the solvent was removed under reduced pressure. The crude product was purified by column chromatography on silica gel (petroleum ether/dichloromethane = 4:1, v/v). The product was then purified by recrystallization from toluene/methanol to afford BFCz-B-Cz as a bright yellow solid (yield = 0.4 g, 43%). This compound was further purified by temperature-gradient sublimation under vacuum. <sup>1</sup>H NMR (400 MHz, Chloroform-*d*)  $\delta$  = 9.23 (s, 1H), 8.99 (s, 1H), 8.50 (d, J = 7.5 Hz, 1H), 8.39 (s, 1H), 8.37 (d, J = 8.4 Hz, 1H), 8.28 - 8.21 (m, 2H), 8.17 (dd, J = 21.3, 7.9 Hz, 2H), 8.05 (d, J = 7.5 Hz, 1H), 7.81 (t, J = 8.3 Hz, 1H), 7.66 (dd, J = 25.3, 7.2 Hz, 2H), 7.57 – 7.43 (m, 4H), 1.72 (s, 9H), 1.54 (s, 9H). MS (MALDI-TOF): m/z calcd 618.2842 [M]<sup>+</sup>: found 618.2865.

Synthesis of 4: Compound 4 was synthesized according to the same procedure described above for the synthesis of 2, using BFCz (2.2 g, 8.8 mmol), cesium carbonate (5.7 g, 17.6 mmol) and 3 (2.8 g, 8.8 mmol), yielding 4 as a white solid (yield = 4.6 g, 94%).<sup>1</sup>H NMR (400 MHz, Chloroform-*d*)  $\delta$  = 8.59 (d, *J* = 7.6 Hz, 1H), 7.97 (dd, *J* =

11.2, 8.0 Hz, 2H), 7.75 (d, J = 8.0 Hz, 1H), 7.51 – 7.36 (m, 7H), 7.30 (d, J = 7.7 Hz, 1H), 7.20 (d, J = 7.9 Hz, 1H), 7.09 (dd, J = 20.3, 8.5 Hz, 4H), 1.36 (s, 9H). <sup>13</sup>C NMR (151 MHz, CDCl<sub>3</sub>)  $\delta = 156.61$ , 156.33, 153.80, 151.24, 147.30, 141.29, 140.45, 138.54, 128.94, 125.87, 125.35, 125.25, 125.15, 122.90, 122.82, 121.01, 120.81, 119.75, 119.25, 118.64, 118.07, 116.78, 116.12, 111.72, 110.18, 108.73, 105.86, 34.49, 31.55.

Synthesis of BFCz-B-O: Compound BFCz-B-O was synthesized according to the same procedure described above for the synthesis of BFCz-B-Cz, using 4 (0.8 g, 1.4 mmol), n-BuLi (2.5 M in hexane, 1.7 mL, 4.3 mmol), BBr<sub>3</sub> (1.1 g, 4.3 mmol), and DIPEA (1 mL), yielding BFCz-B-O as a bright yellow solid (yield = 0.2 g, 31%).<sup>1</sup>H NMR (400 MHz, Chloroform-*d*)  $\delta$  = 8.86 (s, 1H), 8.62 (s, 1H), 8.29 (d, *J* = 7.3 Hz, 1H), 8.10 (d, *J* = 8.3 Hz, 1H), 7.92 – 7.84 (m, 2H), 7.75 – 7.65 (m, 2H), 7.63 (d, *J* = 8.2 Hz, 1H), 7.51 – 7.33 (m, 5H), 7.12 (d, *J* = 8.1 Hz, 1H), 1.57 (s, 9H). MS (MALDI-TOF): m/z calcd 489.1900 [M]<sup>+</sup>: found 489.1939.

#### 7. Supplementary Figures



Figure S1 Cyclic voltammogram (CV) and differential pulse voltammetry (DPV) of

BFCz-B-Cz and BFCz-B-O in degassed dichloromethane (DCM).



**Figure S2** Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) curves of BFCz-B-Cz and BFCz-B-O under nitrogen at a heating rate of 10 °C

 $\min^{-1}$ .



**Figure S3** The optimized configuration of the intermediate with HOMO distributions (in the gas phase at the B3LYP/ def2-svp level).

In order to suggest possible reaction pathways, HOMO distributions of intermediates were calculated. The BFCz-B-Cz and BFCz-B-O show more electron gathered on the 1 position than 2 position, indicating relatively higher reaction activity of 1 position for electrophilic substitution reaction of B atom [8,9]. Therefore,

asymmetric BFCz-B-Cz and BFCz-B-O with B substitute at the 1 position were obtained from the cascade lithiation-borylation-annulation reactions.



Figure S4 [a] PL spectra of BFCz-B-O in 3 wt% and 5 wt% doping film of PhCzBCz.

[b] PL spectra of BFCz-B-Cz in 3 wt% and 5 wt% doping film of PhCzBCz.



Figure S5 Transient photoluminescence decay curves (scale of ns) in 5 wt% doping films of PhCzBCz under vacuum.



**Figure S6** Transient photoluminescence decay curves in 5 wt% doping films of BFCz-B-Cz and BFCz-B-O in PhCzBCz under air condition; the inset shows the corresponding fractional contributions of delayed component.



**Figure S7** Devices of BFCz-B-Cz in PhCzBCz with different doping ratio: [a] Current density-voltage-Luminance characteristics of the devices; [b] Current efficiency- luminance-power efficiency curves for the devices; [c] External quantum efficiency versus luminance curves for the devices; [d] Electroluminescence spectra of the devices.



Figure S8 Devices of BFCz-B-O in PhCzBCz with different doping ratio: [a] Current density-voltage-Luminance characteristics of the devices; [b] Current efficiency-luminance-power efficiency curves for the devices; [c] External quantum efficiency versus luminance curves for the devices; [d] Electroluminescence spectra of the devices.



**Figure S9** Devices of BFCz-B-O in different hosts with 5 wt% doping ratio: [a] Current density-voltage-Luminance characteristics of the devices; [b] Current efficiency- luminance-power efficiency curves for the devices; [c] External quantum efficiency versus luminance curves for the devices; [d] Electroluminescence spectra of the devices.



Figure S10 NTO analysis, energy level alignment, and SOC matrix elements of tCzBN.



Figure S11 a) UV-visible absorption, fluorescence spectra and phosphorescence spectra of tCzBN in toluene (1×10<sup>-5</sup> M) b) Transient photoluminescence decay curves (tCzBN) in 5 wt% doping films of PhCzBCz under vacuum.



**Figure S12** a) EL spectra of OLED based on tCzBN at 5 wt% doping films of PhCzBCz; b) Comparison of OLED performances between tCzBN, BFCz-B-Cz and BFCz-B-O.



Figure S13 The EL spectrum of (a) BFCz-B-Cz, (b) BFCz-B-O in PhCzBCz with 5 wt% doping concentration under different applied voltages.



Figure S14 <sup>1</sup>H NMR spectrum of 2.



Figure S16 <sup>1</sup>H NMR spectrum of BFCz-B-Cz.



Figure S17 MALTI-TOF spectrum of BFCz-B-Cz.



Figure S18 <sup>1</sup>H NMR spectrum of 4.













Figure S21 MALTI-TOF spectrum of BFCz-B-O.

Emitter	НОМО	LUMO <sup>c</sup>	$\lambda_{abs}{}^d$	$\lambda_{PL}^{e}$	Stoke Shift <sup>f</sup>	$E_{\rm S1}/E_{\rm T1}{}^{\rm g}$	$\Delta E_{ST}^{h}$	FWHM <sup>i</sup>
	[eV]	[eV]	[nm]	[nm]	[nm]	[eV]	[eV]	[nm]
tCzBN			467	485	18	2.57/2.45	0.12	24
BFCz-B-Cz	-5.39 <sup>a</sup> /-5.33 <sup>b</sup>	-2.85/-2.79	472	488	16	2.60/2.52	0.08	24
BFCz-B-O	-5.60 <sup>a</sup> /-5.53 <sup>b</sup>	-2.81/-2.74	431	449	18	2.84/2.75	0.09	25

Table S1 Electrochemical data and photophysical data of emitters.

[a] HOMOs from CV via  $E_{\text{HOMO}} = -[4.8 + E_{\text{O}}-\text{E}(\text{Fc/Fc}^+)]$ . [b] HOMOs from DPV via  $E_{\text{HOMO}} = -[4.8 + E_{\text{O}}-\text{E}(\text{Fc/Fc}^+)]$ . [c] LUMOs from  $E_{\text{LUMO}} = E_{\text{HOMO}} + E_{\text{opt}}$  ( $E_{\text{opt}}$  was obtained from the absorption edges of normalized absorption spectra). [d] Absorption peak wavelength in toluene solution at a concentration of  $1 \times 10^{-5}$  M. [e] PL peak wavelength in toluene solution at a concentration of  $1 \times 10^{-5}$  M. [e] PL peak wavelength in toluene solution at a concentration of  $1 \times 10^{-5}$  M. [g] Obtained from the onset of the PL spectrum and Ph spectrum at 77 K in toluene solution at a concentration of  $1 \times 10^{-5}$  M. [h]  $\Delta E_{\text{ST}} = E_{\text{S1}} - E_{\text{T1}}$ . [i] Full-width at half-maximum.

Emitter	PLQY <sub>air</sub> <sup>a</sup> /PLQY <sub>vacuum</sub> <sup>b</sup>	$\tau_p{}^c/\tau_d{}^d$	$k_{ m F}{}^{ m e}$	$k_{ m ISC}{}^{ m f}$	$k_{\rm RISC}{}^{\rm g}$
	(%)	$[ns/\mu s]$	$[10^7 s^{-1}]$	$[10^{7}s^{-1}]$	[10 <sup>5</sup> s <sup>-1</sup> ]
tCzBN	61/75	9.5/82.6	1.1	8.5	0.8
BFCz-B-Cz	81/92	8.7/20.6	1.7	9.5	3.0
BFCz-B-O	69/80	10.1/32.9	1.5	7.5	1.6

 Table S2 Summary of kinetics parameters of emitters in 5 wt%-doped films with PhCzBCz host

[a] Photoluminescence quantum yields under air. [b] Photoluminescence quantum yields under vacuum. [c] Prompt fluorescent lifetimes. [d] Delayed fluorescent lifetimes. [e] The radiative decay rate constant of  $S_1$ . [f] The intersystem crossing rate constant. [g]  $k_{RISC}$  is the reverse intersystem crossing rate constant.

	1 4	8			
Method	Energy Level	tCzBN	BFCz-B-Cz	BFCz-B-O	
	[eV]				
B3LYP/ def2-svp <sup>a</sup>	$S_1$	2.85	2.73	2.94	
	$T_1$	2.44	2.42	2.59	
	$\Delta E_{ m ST}$	0.41	0.31	0.35	
SCS-CC2/cc-pVDZ <sup>b</sup>	$S_1$	2.95	2.91	3.11	
	$T_1$	2.84	2.85	3.03	
	$\Delta E_{ m ST}$	0.11	0.06	0.08	

Table S3 Computed energy levels using different methods

[a] Excited state energy levels calculated in the gas phase at the B3LYP/ def2-svp level. [b] Excited state energy levels calculated in the gas phase at the SCS-CC2/cc-pVDZ level.

concentration.							
Device	rice V <sub>on</sub> <sup>a</sup>		EQE <sup>c</sup>	$\lambda_{EL}{}^d$	FWHM <sup>e</sup>		
	(V)	$(cd m^{-2})$	(%)	(nm)	(nm)		
BFCz-B-Cz 3wt%	3.2	7349	19.6	492	38		
BFCz-B-Cz 5wt%	3.1	8910	26.0	492	42		
BFCz-B-Cz 10wt%	3.1	9430	23.5	492	45		
BFCz-B-O 3wt%	3.6	1710	9.1	456	44		
BFCz-B-O 5wt%	3.7	1985	11.0	456	45		
BFCz-B-O 10wt%	3.5	2570	9.6	456	54		

 Table S4. Electroluminescence data of BFCz-B-Cz and BFCz-B-O at different doping

 concentration

[a] Turn-on voltage at 1 cd/m<sup>2</sup>. [b] Maximum luminance. [c] Maximum external quantum efficiency. [d] Peak wavelength of the EL spectrum. [e] Full-width at half-maximum.

 Device	V <sub>on</sub> <sup>a</sup>	L <sub>max</sub> <sup>b</sup>	EQE <sup>d</sup>	$\lambda_{EL}^{e}$	FWHM <sup>f</sup>
	(V)	$(cd m^{-2})$	(%)	(nm)	(nm)
BFCz-B-O in mCBP	3.3	2860	16.5	456	50
BFCz-B-O in PhCzBCz	3.7	1985	11.0	456	45
BFCz-B-O in 26DCzPPy	3.9	2640	16.3	456	40

Table S5. Electroluminescence data of BFCz-B-O in different host at 5 wt% doping concentration.

[a] Turn-on voltage at 1 cd/m<sup>2</sup>. [b] Maximum luminance. [c] Maximum external quantum efficiency. [d] Peak wavelength of the EL spectrum. [e] Full-width at half-maximum.

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