Supporting Information

High-Performance Deep-Blue Electroluminescence from Multi-resonance TADF Emitters with Spirofluorene-Fused Double Boron Framework

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1. Experimental Section

1.1. Materials and Methods

All raw materials and anhydrous solvents were commercially available and used without further purification. The reactions were carried out under the protection of the high-purity argon atmosphere. All reactions were heated by metal sand bath (WATTCAS, LAB-500, https://www.wattcas.com). NMR measurements were conducted on Bruker Advance 400 (or 500, 600) spectrometer. Tetramethylsilane was used as the internal standard and $CDCl_3$ or DMSO-d_6 as the solvent. The high resolution mass spectrometry (HR-MS) for the compounds was conducted on a Thermo Scientific LTQ Orbitrap XL with ESI ion source.

1.2. Synthesis

***Synthesis of 3,5-Dibromo-N,N-diphenylaniline* (1):** 3,5-Dibromoaniline (5.00 g, 27.90 mmol), CuI (1.00 g, 5.58 mmol), 1,10-Phenanthroline (1.00 g, 5.58 mmol), KOH (12.52 g, 223.2 mmol) were dissolved in 10 mL Iodobenzene at room temperature. After blowing argon for 15 minutes, the reaction was stirred at 110 ºC for 12 h. After cooling to room temperature, toluene was removed by rotary evaporation. Subsequently, the resultant crude product underwent purification through the employment of silica gel column chromatography (eluent: petroleum ether/dichloromethane = 6:1, *v/v*), giving the intermediate **1** as a white solid (7.62 g, yield 68.2%).^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.30 (t, J=7.9, 4H), 7.19 (t, J=1.6, 1H), 7.14 – 7.07 (m, 7H), 7.05 (d, J=1.6, 2H).

***Synthesis of N1,N3-dimesityl-N5,N5-diphenylbenzene-1,3,5-triamine* (2):** Intermediate **1** (4.00 g, 9.93 mmol), 2,4,6-Trimethylaniline (3.36 g, 24.82 mmol), $Pd(OAc)_2$ (0.22 g, 0.99 mmol), *t*-$Bu_3P{\cdot}HBF_4$ (0.78 g, 2.58 mmol) and *t*-BuONa (2.56 g, 29.79 mmol) were dissolved in dry toluene (40 mL) at room temperature, after blowing argon for 15 minutes, the reaction was stirred at 110 ºC for 18 h. After cooling to room temperature, toluene was removed by rotary evaporation. Subsequently, the resultant crude product underwent purification through the employment of silica gel column chromatography (eluent: petroleum ether/dichloromethane = 7:1, *v/v*), giving the intermediate **2** as a white solid (3.14 g, yield 62%). 2:^{1}H NMR (500 MHz, DMSO) δ = 7.19 (t, J=7.9, 1H), 6.90 (d, J=11.4, 2H), 6.79 (s, 1H), 5.45 (s, 0H), 5.39 (s, 0H), 2.16 (s, 1H), 2.05 (s, 3H).

***Synthesis of N1,N3-di(9,9'-spirobi[fluoren]-3-yl)-N1,N3-dimesityl-N5,N5-diphenylbenzene-1,3,5-triamine* (3):** To a mixture of intermediate **2** (2.00 g, 3.90 mmol),3-bromo-9,9'-spirobi[fluorene] (3.40 g, 8.59

mmol), $Pd(OAc)_2$ (0.08 g, 0.39 mmol), *t*-$Bu_3P \cdot HBF_4$ (0.29 g, 1.02 mmol) and *t*-BuONa (1.13 g, 11.72 mmol) in 30 mL dry toluene,was added under argon atmosphere, then the reaction was stirred at 110 ºC for 12 h. After cooling to room temperature, toluene was removed by rotary evaporation. Subsequently, the resultant crude product underwent purification through the employment of silica gel column chromatography (eluent: petroleum ether/dichloromethane = 1:1, *v/v*), giving the intermediate **3** as a white solid (2.60 g, yield 58%). 3: ^{1}H NMR (500 MHz, DMSO) δ = 7.97 (d, J=7.6, 1H), 7.87 (d, J=7.7, 0H), 7.60 (d, J=1.7, 1H), 7.42 – 7.31 (m, 2H), 7.16 – 7.08 (m, 3H), 7.01 (d, J=7.7, 1H), 6.85 (t, J=7.3, 1H), 6.81 (s, 1H), 6.55 – 6.48 (m, 2H), 6.36 (dt, J=21.5, 5.1, 1H), 6.07 (s, 0H), 6.03 (d, J=1.7, 0H), 2.08 (s, 2H), 1.95 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ = 149.52, 149.15, 148.66, 147.61, 147.46, 145.95, 142.28, 141.72, 141.60, 140.35, 139.99, 137.09, 136.36, 129.68, 128.90, 127.71, 127.62, 127.50, 127.31, 124.08, 124.02, 123.80, 122.28, 120.02, 119.86, 111.16, 108.49, 106.14, 65.41, 20.87, 18.56.

***Synthesis of DB-SF1*:** In an argon atmosphere, a solution of intermediate 3 (1.30 mmol) in 15 mL of highly dry 1,2-dichlorobenzene was prepared. BBr_3 (4.5 mL, 47.16 mmol) was then added to the solution at -78 ºC. After stirring at room temperature for 1 hour, the reaction mixture was heated to 220 ºC and stirred for 48 hours. Upon cooling the mixture to room temperature, *N*,*N*-diisopropylethylamine (10.39 mL, 62.88 mmol) was added at 0 ºC to quench the excessive BBr_3 and generated hydrogen bromide, followed by gradual addition of methanol. Subsequently, the 1,2-dichlorobenzene was removed by rotary evaporation. The resulting crude product was subjected to purification using silica gel column chromatography (eluent: petroleum ether/dichloromethane = 12:1, *v/v*), yielding the desired products **DB-SF1** (0.60 g, 40% yield) as a yellow solid. **DB-SF1:**^{1}H NMR (500 MHz, $CDCl_3$) δ = 8.20 (s, 1H), 8.16 (d, *J*=8.2, 1H), 8.03 (d, *J*=9.0, 1H), 7.95 (d, *J*=8.4, 1H), 7.87 (d, *J*=8.1, 1H), 7.65 (d, *J*=8.2, 1H), 7.46 (t, *J*=8.0, 1H), 7.31 (dd, *J*=18.4, 9.5, 3H), 7.24 – 7.14 (m, 3H), 7.09 (t, *J*=7.5, 1H), 7.06 – 6.98 (m, 3H), 6.94 (d, *J*=8.0, 1H), 6.70 (dd, *J*=22.5, 8.1, 2H), 5.30 (s, 0H), 2.52 (s, 3H), 1.76 (d, *J*=16.8, 6H). ^{13}C NMR (126 MHz, toluene-d_8) δ = 150.84, 149.86, 149.63, 148.57, 147.36, 146.71, 146.58, 145.66, 141.92, 141.84, 141.69, 141.11, 137.80, 137.30, 137.11, 136.84, 134.09, 130.52, 130.04, 129.52, 128.81, 128.69, 128.50, 128.31, 127.93, 127.88, 127.78, 127.59, 127.52, 127.40, 125.04, 124.95, 124.76, 124.56, 124.40, 124.32, 124.18, 123.66, 123.20, 120.37, 120.05, 119.99, 106.19, 89.75, 65.98, 21.08, 20.51, 20.36, 20.20, 20.05, 19.90, 19.75, 19.59, 17.13, 16.89.

***Synthesis of N1,N3-di(9,9'-spirobi[fluoren]-2-yl)-N1,N3-dimesityl-N5,N5-diphenylbenzene-1,3,5-triamine* (4):** To a mixture of intermediate **2** (2.00 g, 3.90 mmol),2-bromo-9,9'-spirobi[fluorene] (3.40 g, 8.59 mmol), $Pd(OAc)_2$ (0.08 g, 0.39 mmol), *t*-$Bu_3P{\cdot}HBF_4$ (0.29 g, 1.02 mmol) and *t*-BuONa (1.13 g, 11.72 mmol)in 30 mL dry toluene was added under argon atmosphere, then the reaction was stirred at 110 ºC for 12 h. After cooling to room temperature, toluene was removed by rotary evaporation. Subsequently, the resultant crude product underwent purification through the employment of silica gel column chromatography (eluent: petroleum ether/dichloromethane = 3:1, *v/v*), giving the intermediate **4** as a white solid (0.99 g, yield 90%). **4:**^{1}H NMR (500 MHz, DMSO) δ = 7.92 (d, *J*=7.6, 1H), 7.81 (d, *J*=7.6, 1H), 7.75 (d, *J*=8.4, 1H), 7.35 – 7.26 (m, 2H), 7.05 (t, *J*=7.8, 1H), 7.01 – 6.94 (m, 2H), 6.86 (t, *J*=7.4, 1H), 6.69 (s, 1H), 6.62 (d, *J*=7.8, 1H), 6.48 (t, *J*=5.9, 1H), 6.40 (d, *J*=7.6, 0H), 6.18 (d, *J*=1.8, 0H), 6.04 (s, 0H), 5.54 (d, *J*=1.7, 0H), 2.14 (s, 2H), 1.49 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ = 149.30, 149.13, 148.72, 148.19, 146.86, 146.75, 145.70, 141.66, 141.50, 140.04, 136.74, 135.98, 134.27, 129.41, 128.59, 127.87, 127.47, 126.28, 123.79, 123.55, 123.53, 121.97, 120.42, 119.76, 118.94, 118.78, 114.27, 109.57, 106.39, 65.85, 20.89, 18.17.

***Synthesis of DB-SF2*:** In an argon atmosphere, a solution of intermediate 4 (1.30 mmol) in 15 mL of highly dry 1,2-dichlorobenzene was prepared. BBr_3 (4.5 mL, 47.16 mmol) was then added to the solution at -78 ºC. After stirring at room temperature for 1 hour, the reaction mixture was heated to 220 ºC and stirred for 48 hours. Upon cooling the mixture to room temperature, *N*,*N*-diisopropylethylamine (10.39 mL, 62.88 mmol) was added at 0 ºC to quench the excessive BBr_3 and generated hydrogen bromide, followed by gradual addition of methanol. Subsequently, the 1,2-dichlorobenzene was removed by rotary evaporation. The resulting crude product was subjected to purification using silica gel column chromatography (eluent: petroleum ether/dichloromethane = 12:1, *v/v*), yielding the desired products **DB-SF2** (0.60 g, 40% yield) as a yellow solid. **DB-SF2:**^{1}H NMR (400 MHz, $CDCl_3$) δ = 9.31 (s, 1H), 8.99 (d, *J*=7.8, 1H), 8.38 (d, *J*=7.9, 1H), 8.05 (d, *J*=7.6, 1H), 7.71 (dd, *J*=7.5, 3.8, 2H), 7.60 (dd, *J*=12.1, 3.8, 2H), 7.39 (t, *J*=7.4, 1H), 7.30 (d, *J*=3.0, 1H), 7.09 – 6.97 (m, 3H), 6.80 (d, *J*=7.5, 1H), 6.64 (dd, *J*=9.7, 8.0, 2H), 6.57 (s, 1H), 6.48 (s, 1H), 6.09 (s, 1H), 4.95 (s, 0H), 2.18 (s, 3H), 1.41 (s, 3H), 1.24 (s, 3H). Due to the poor solubility of this compound, we did not obtain its carbon spectrum.

1.3. Single Crystal X-ray Crystallography

The single crystal of **DB-SF1** and **DB-SF2** was prepared from a methanol/chlorobenzene solution by evaporation method. X-ray single crystal data was collected on a Bruker D8 Venture diffractometer using CuKα radiation (λ = 1.54178) source. The selected crystal was kept at 158.0 K during data collection. Using Olex2[1], the structure was solved with the ShelXT[2] structure solution program using Intrinsic Phasing and refined with the ShelXL[3] refinement package using Least Squares minimization. Selected crystal data are listed in **Table S1-S2**. All crystallographic information in CIF format have been deposited at the Cambridge Crystallographic Data Center (CCDC) under deposition number 2369010 and 2369013 for **DB-SF1** and **DB-SF2** via www.ccdc.cam.ac.uk/data_request/cif, or by emailing data_request@ccdc.cam.ac.uk, or by contacting the Cambridge Crystallographic Data Center, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033.

1.4. Quantum Chemical Calculations

All of the calculations were carried out with Gaussian 16 program package. Density functional theory (DFT) calculations on the geometrical and electronic properties of the ground-state were performed based on B3LYP density functional method with basis set 6-31G(d,p). The density functional dispersion correction was conducted by Grimme's D3 version with Becke-Johnson damping function. Time-dependent DFT (TD-DFT) calculations were also carried out by this method. Vertical excitation calculations were based on optimized S_0 geometries, and geometry optimizations for excited S_1 and T_1 states were carried out at the same level of theory. The analysis of hole and electron pair distributions was conducted using the Multiwfn code.[4,5] Spin-Component Scaling second-order approximate Coupled-Cluster (SCS-CC2) calculations were performed using the MRCC program with the cc-pVDZ basis-set.[6,7]

The spin-orbit coupling (SOC) matrix elements between the S_1 and T_1 states for the emitters were calculated with PySOC using the T_1 geometries, by considering that the three T_1^m substrates (m = 1, 0, -1) are degenerate, *i.e.* $\langle S_1 | \hat{H}_{soc} | T_1 \rangle = \sqrt{\sum_{m=0,\pm1} \langle S_1 | \hat{H}_{so} | T_1{}^m \rangle^2}$, where the $\hat{H}_{soc}$ represents the interaction of the SOC.[8] All SOCs were obtained at the TD-DFT level of theory using the B3LYP functional and the 6-31G(d,p) basis set.

The Huang-Rhys factors (HRF) and reorganization energy for $S_1 \rightarrow S_0$ and $S_0 \rightarrow S_1$ transition were conducted with the DUSHIN module in MOMAP (Molecular Materials Property Prediction Package).

1.5. Thermal and Electrochemical Characterization

Thermogravimetric analysis (TGA) was undertaken using TGA-Q50 Instrument (TA Instruments, America) at a heating rate of 10 °C/min from 50 °C to 800 °C under nitrogen flushing. The thermal decomposition temperatures (T_d) were determined by the recorded temperature at 5% weight loss. Cyclic voltammetry (CV) measurements were carried out on a CHI600 electrochemical analyzer (Chenhua, China) at room temperature and a scan speed of 50 mV s^{-1}, with a conventional three-electrode system consisting of a glassy carbon working electrode, a platinum wire auxiliary electrode, and an Ag/AgCl standard electrode using as the reference electrode. The supporting electrolyte was 0.1 M tetrabutylammonium hexafluorophosphate (Bu_4NPF_6) in anhydrous dichloromethane solution, and ferrocene was added as a calibrant in the whole measurement. The HOMO energy levels of the compounds were calculated according to the formula: E_{HOMO} (eV) = - [4.8 + ($E_{1/2(ox/red)}$ - $E_{1/2(Fc+/Fc)}$)] eV. The LUMO energy levels of the compounds were then deduced from the HOMO levels and the UV-Vis absorption onsets of the longer wavelength.

1.6. Photophysical Characterization

The UV-vis absorption spectra were obtained on a Shimadzu UV-2600 spectrophotometer (Shimadzu, Japan) at room temperature with a concentration of 1×10^{-5}M. Phosphorescence spectra were measured on a Hitachi F-7100 fluorescence spectrophotometer at 77 K. The transient photoluminescence (PL) decay curves were obtained by FluoTime 300 (PicoQuant GmbH) with a Picosecond Pulsed UV-LASTER (LASTER375) as the excitation source. The solid-state PL quantum efficiencies (Φ_{PL}s) were measured on a Hamamatsu UV-NIR absolute PL quantum yield spectrometer (C13534, Hamamatsu Photonics) equipped with an integrating sphere. The integrating sphere was purged with dry argon to maintain an inert atmosphere and all the samples were excited at 320 nm. Nanosecond time-resolved transient absorption spectra and decay kinetics were measured on LFP instrument (LP 980, Edinburgh Instruments LTD). The pump laser beam and the probe beam crossed perpendicularly through the liquid sample in a quartz cuvette (10 mm × 10 mm). A dynamic decay curve was recorded with a digital phosphor oscilloscope (TDS 3012C, Tektronix Inc.).

1.7. Analysis of Rate Constants

The estimation of rate constants pertaining to radiative decay (k_r) and nonradiative decay (k_{nr}) transitioning from S_1 to S_0, as well as the rate constants associated with intersystem crossing (k_{ISC}) and reverse intersystem crossing (k_{RISC}), can be accomplished by employing the subsequent equations.[9-11]

$$k_r = \Phi_p k_p + \Phi_d k_d \approx \Phi_p k_p \quad \text{Eq.(1)}$$

$$k_{nr} = \frac{1 - \Phi_{PL}}{\Phi_{PL}} k_r \quad \text{Eq.(2)}$$

$$k_{ISC} = k_p - k_r - k_{nr} \quad \text{Eq.(3)}$$

$$k_{RISC} = (k_p k_d \Phi_d)/(k_{ISC} \Phi_p) \quad \text{Eq.(4)}$$

Where k_p and k_d represent the decay rate constants for prompt and delayed fluorescence, respectively, which are in reciprocal relationship with the decay time constants (τ_p and τ_d) experimentally determined from transient PL characteristics. Φ_p and Φ_d indicate prompt and delayed fluorescence components and can be distinguished from the total Φ_{PL} by comparing the integrated intensities of prompt (r_p) and delayed components (r_d) in the transient PL spectra. r_p and r_d were determined using τ_p and τ_d and fitting parameter (A_p, A_d) as follows.

$$I(t) = A_p e^{-\frac{1}{\tau_p}} + A_d e^{-\frac{1}{\tau_d}} \quad \text{Eq.(5)}$$

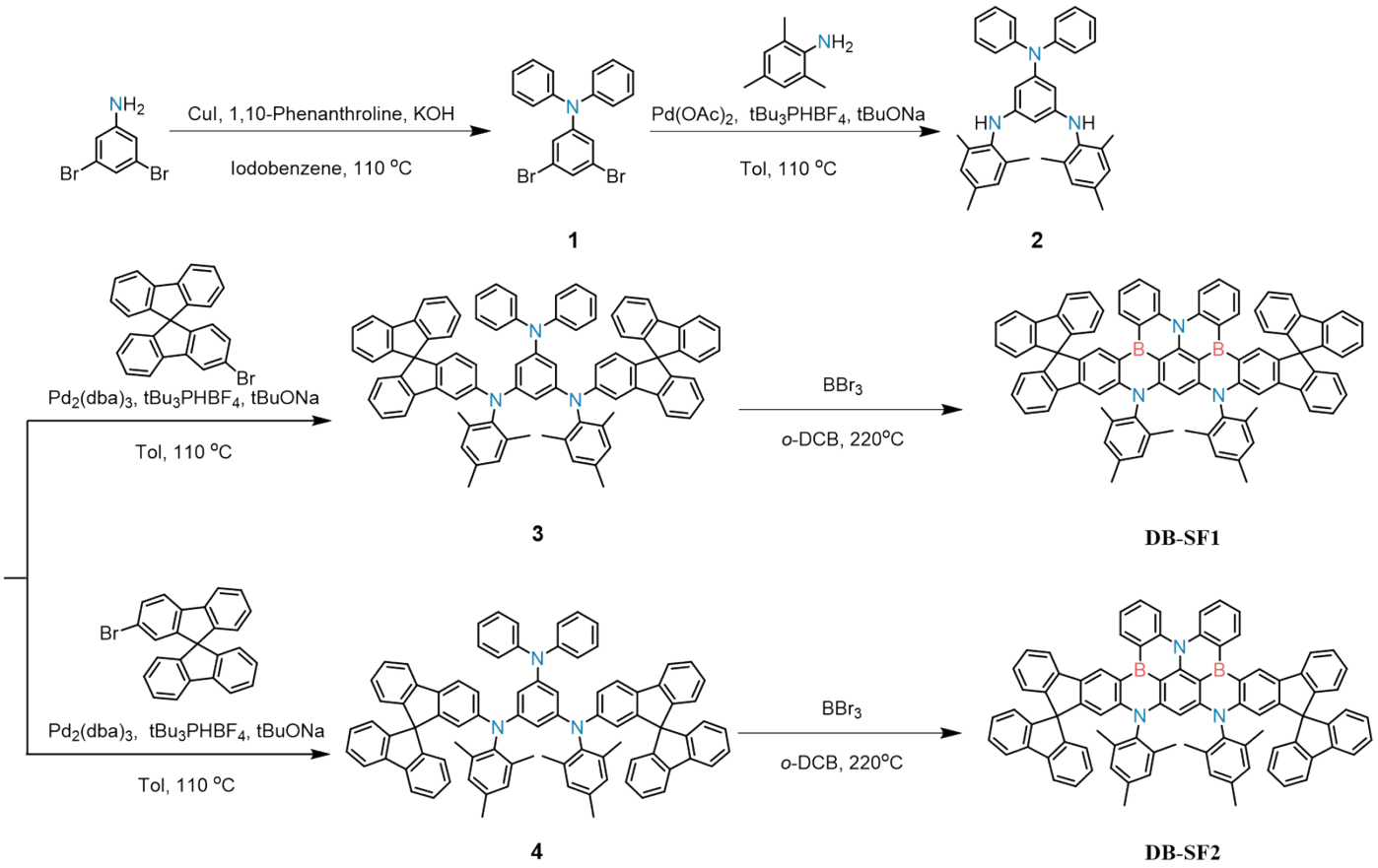
$$r_p = A_p \tau_p / (A_p \tau_p + A_d \tau_d) \quad \text{Eq.(6)}$$

$$r_d = A_d \tau_d / (A_p \tau_p + A_d \tau_d) \quad \text{Eq.(7)}$$

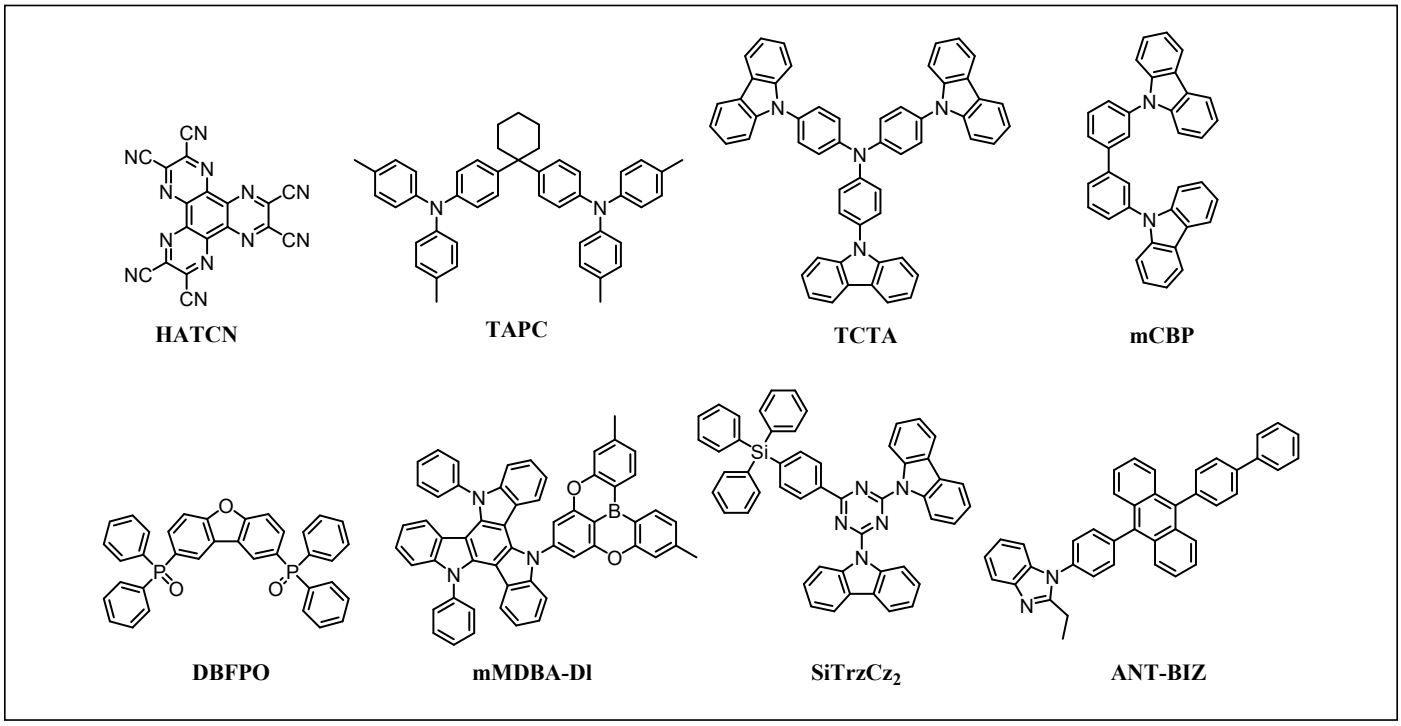
1.8. Device Fabrication and Measurement

The ITO coated glass substrates with a sheet resistance of 15 Ω square^{-1} were consecutively ultrasonicated with acetone/ethanol and dried with nitrogen gas flow, followed by 20 min ultraviolet light-ozone (UVO) treatment in a UV-ozone surface processor (PL16 series, Sen Lights Corporation). Then the sample was transferred to the deposition system. Both 8-hydroxyquinolinolato-lithium (Liq) as electron injection layer and aluminum (Al) as cathode layer were deposited by thermal evaporation at 5×10^{-5} Pa. The organic layers were deposited at the rates of 0.2-3 Å/s. After the organic film deposition, Liq and Al layer were deposited with rates of 0.1 and 3 Å/s, respectively. The emitting area of the device is about 0.09 cm^2. The current density-voltage-luminance (*J-V-L*), EQE-*L* curves and electroluminescence spectra were measured using a Keithley 2400 source meter and an absolute EQE measurement system (C9920-12, Hamamatsu Photonics, Japan). The device operational lifetime (LT_{50}) was evaluated by measuring the luminance over time at constant current densities (FS-MP64, Suzhou FSTAR Scientific Instrument Co. Ltd., China).

2. Schemes, Figures and Tables



Scheme S1. Synthetic route of **DB-SF1**and **DB-SF2**.



Scheme S2. Chemical structures of materials utilized for device fabrication.

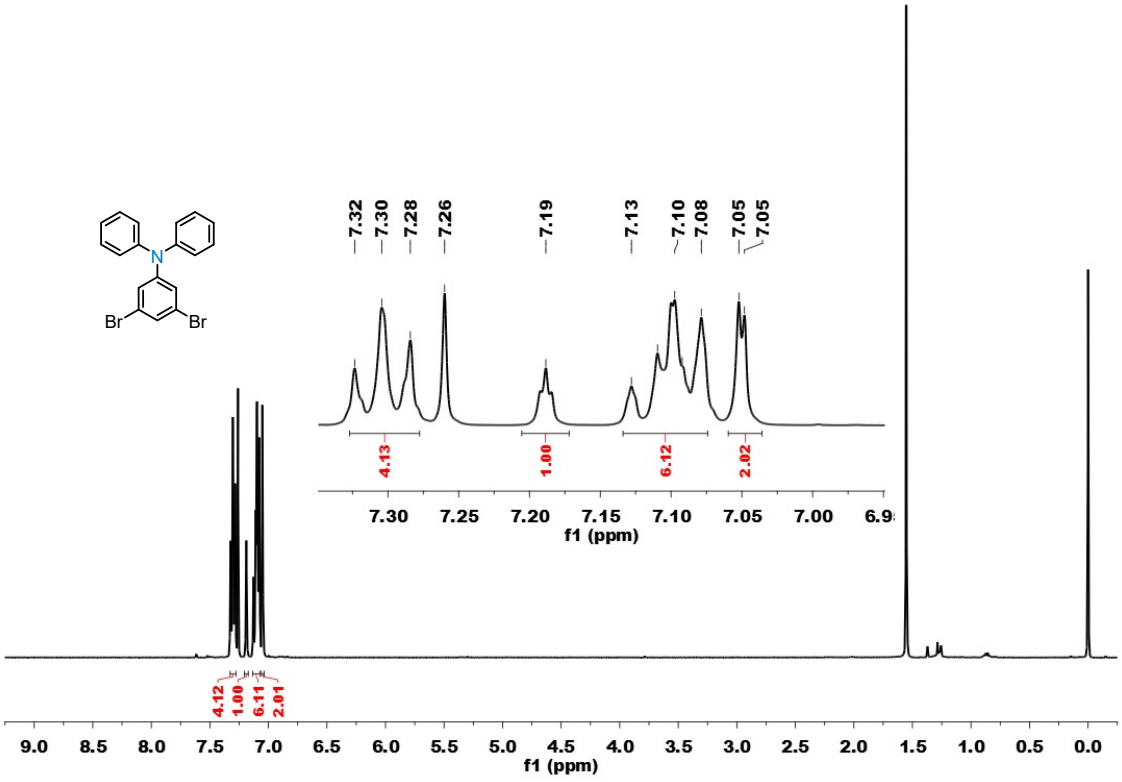


Figure S1. ^{1}H NMR spectrum of **1** in $CDCl_3$ (400 MHz, 25 ^{o}C).

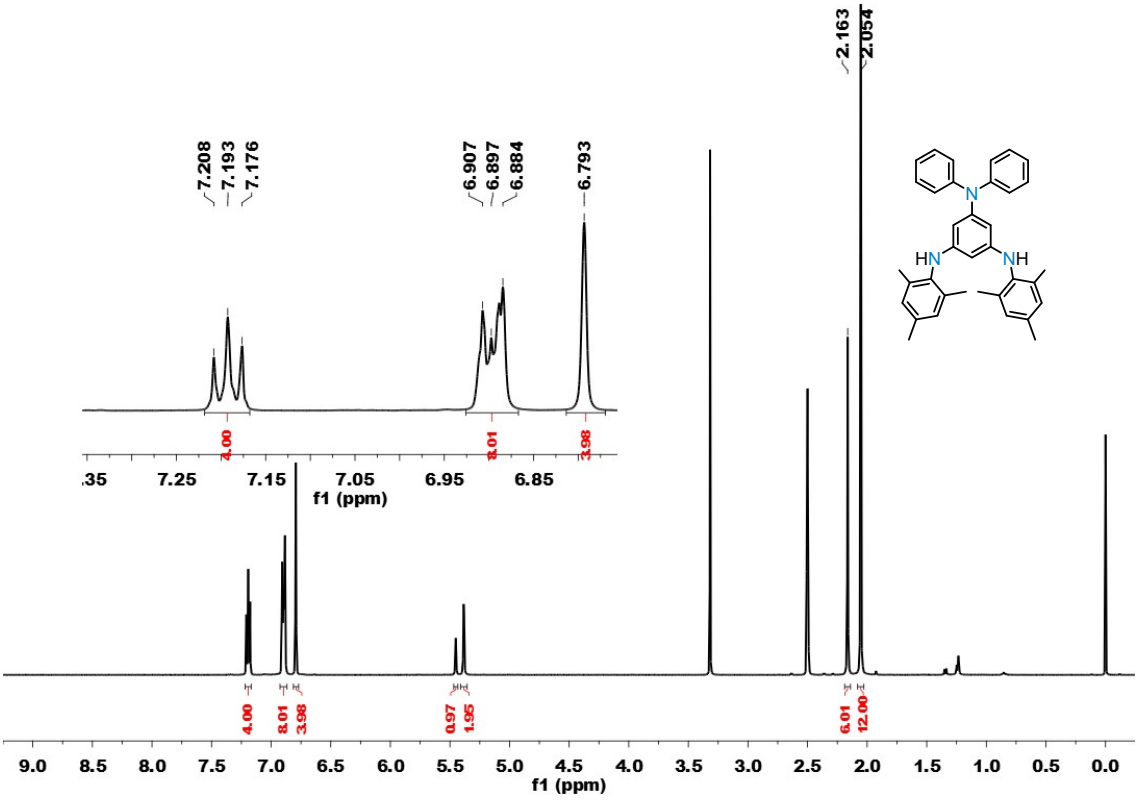


Figure S2. ^{1}H NMR spectrum of **2** in DMSO-d_6 (500 MHz, 25 °C).

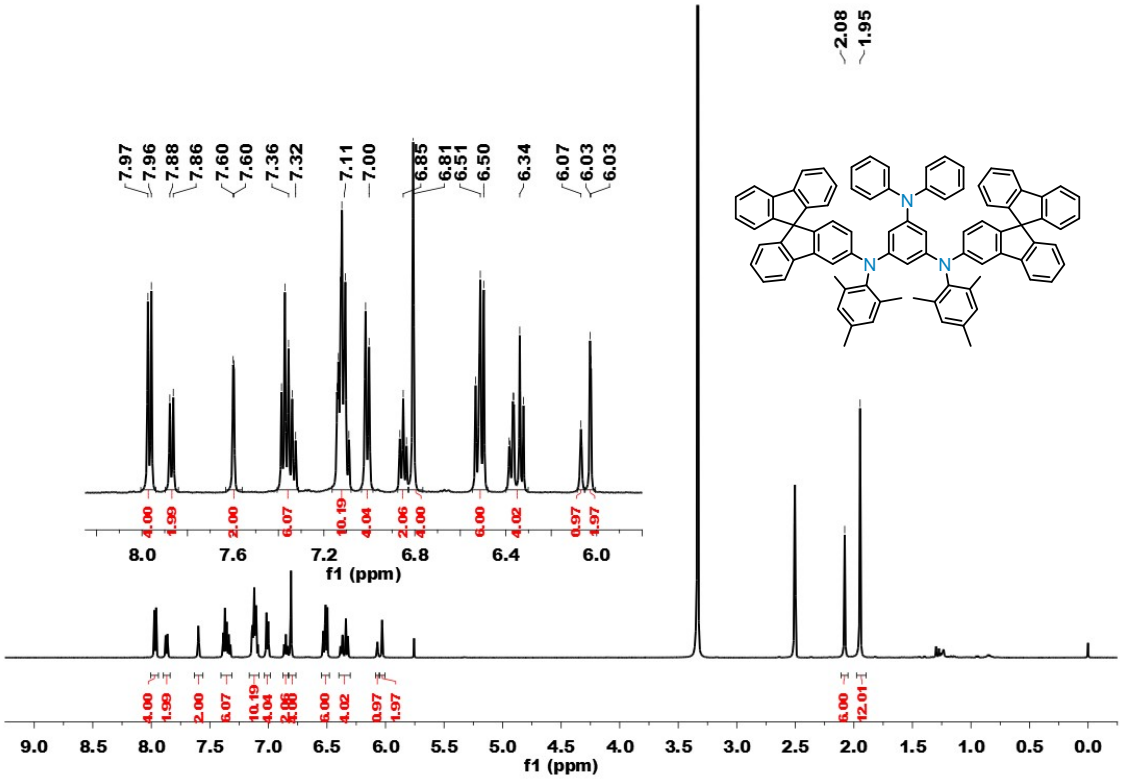


Figure S3. ^{1}H NMR spectrum of **3** in DMSO-d_6 (500 MHz, 25 °C).

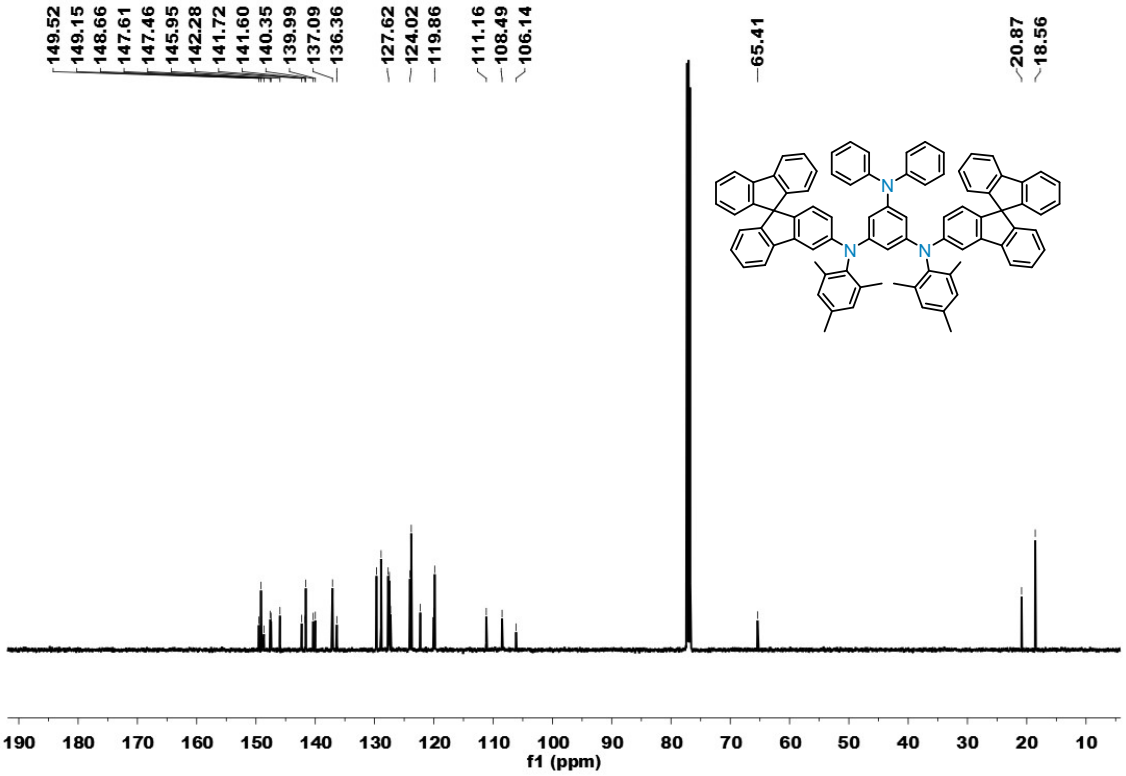


Figure S4. ^{13}C NMR spectrum of **3** in $CDCl_3$ (126 MHz, 25 °C).

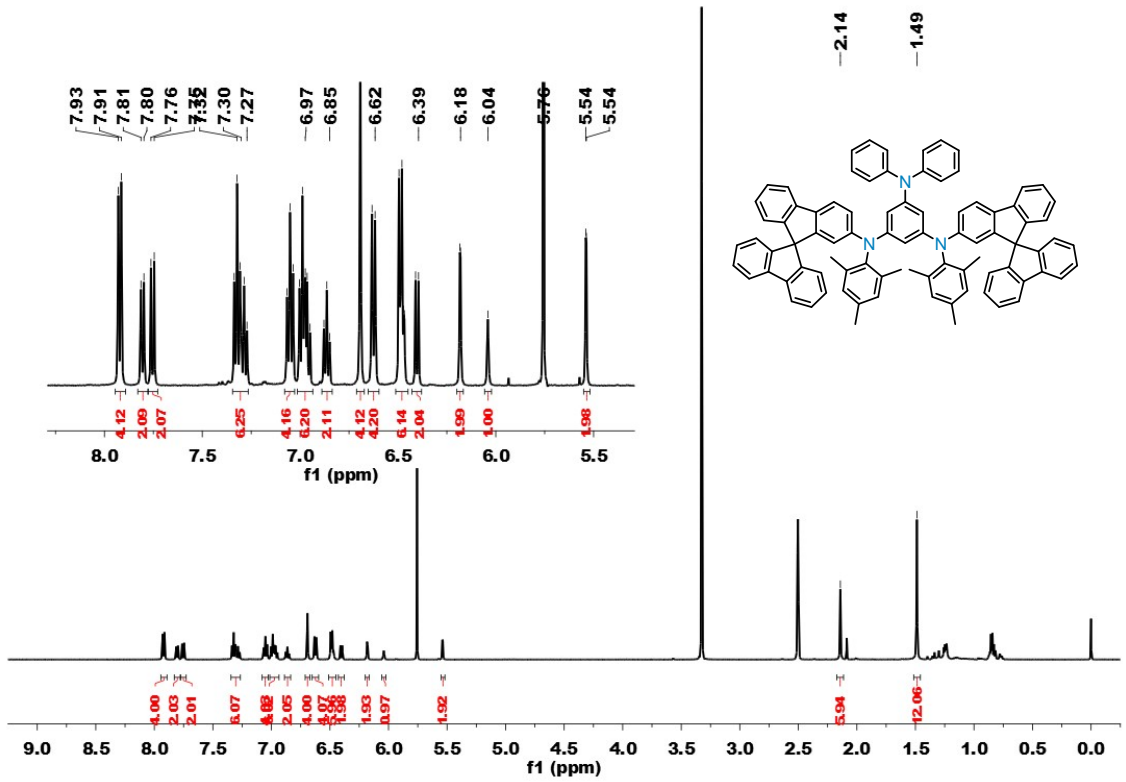


Figure S5.[1]H NMR spectrum of **4** in DMSO-d_6 (500 MHz, 25 °C).

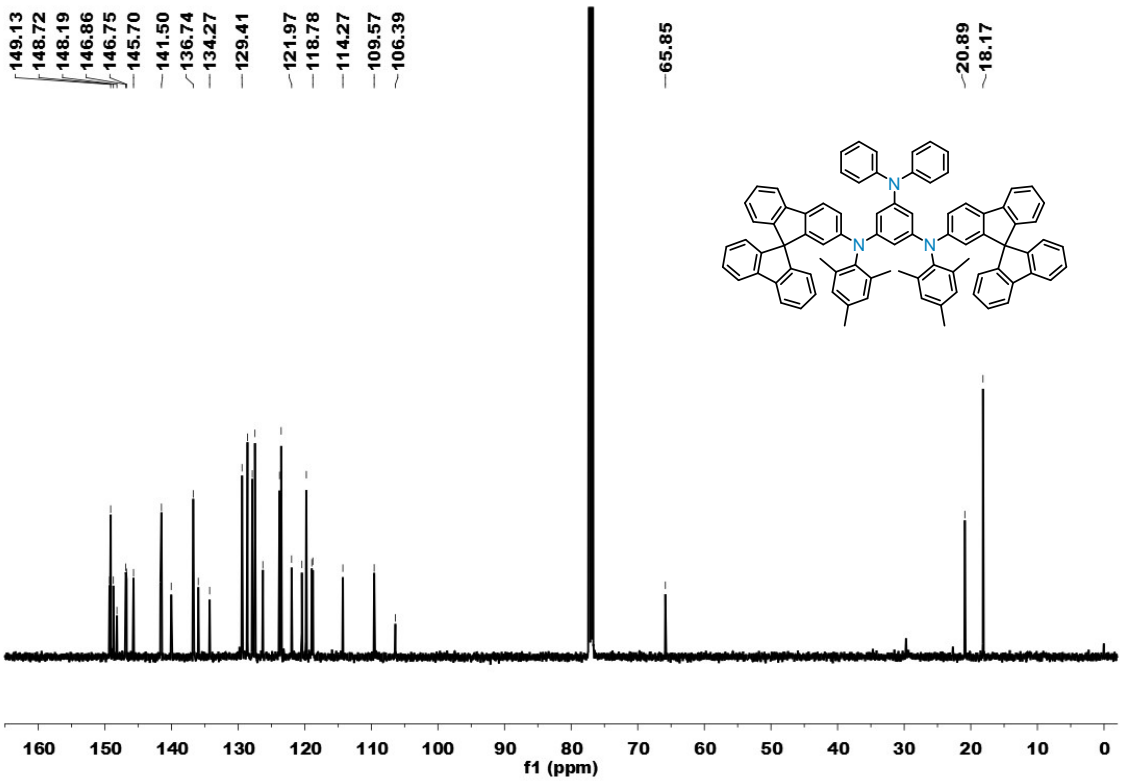


Figure S6. [13]C NMR spectrum of **4** in $CDCl_3$ (126 MHz, 25 °C).

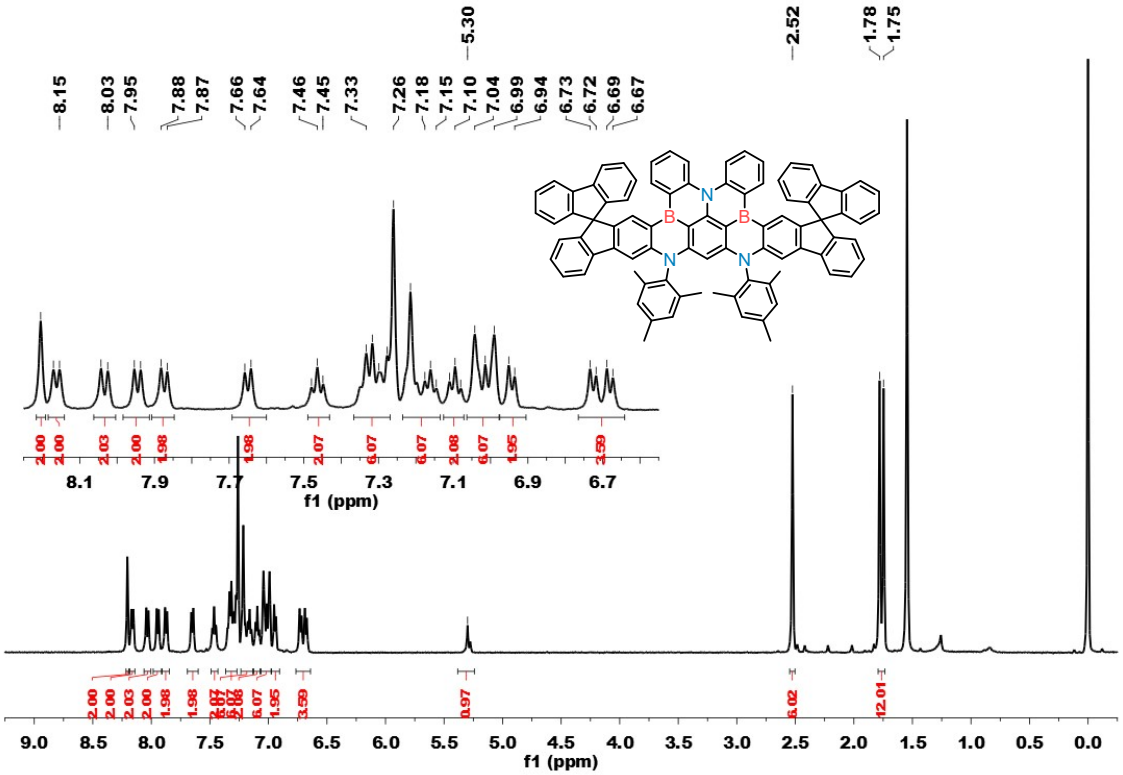


Figure S7. ^{1}H NMR spectrum of **DB-SF1** in $CDCl_3$ (500 MHz, 25 °C).

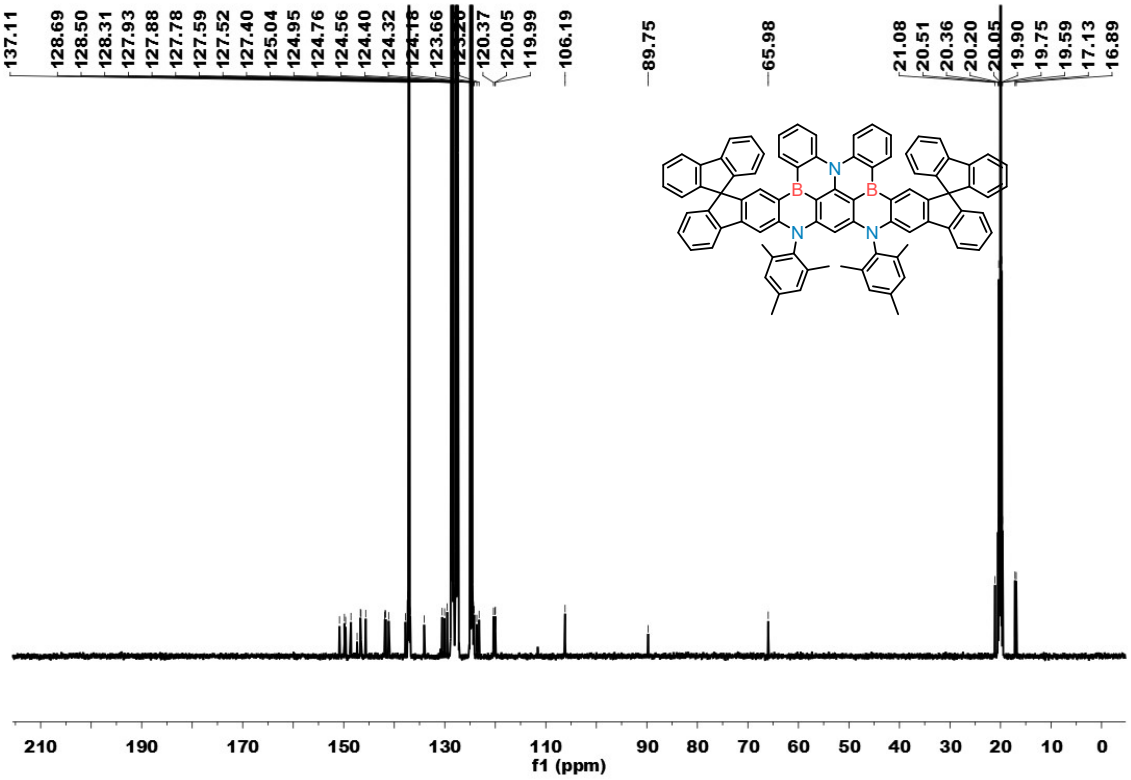


Figure S8. ^{13}C NMR spectrum of **DB-SF1** in toluene-d_8 (500 MHz, 25 °C).

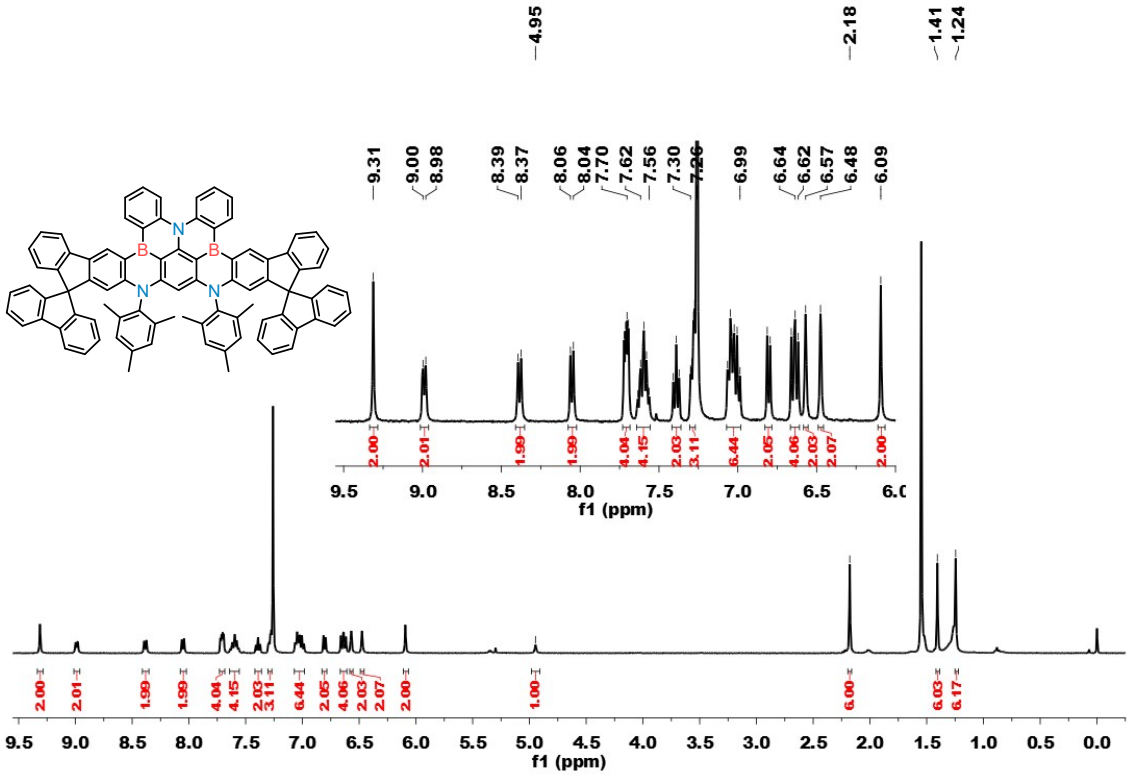


Figure S9. ^{1}H NMR spectrum of **DB-SF2** in $CDCl_3$ (500 MHz, 25 °C).

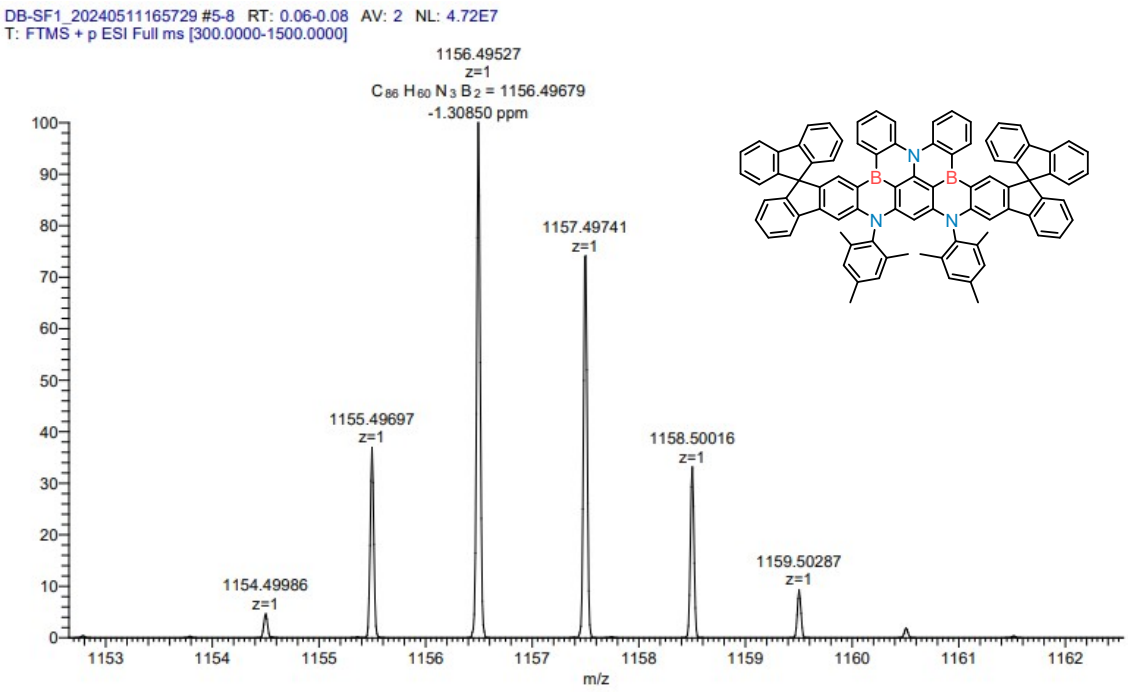


Figure S10. HR-MS spectrum of **DB-SF1**.

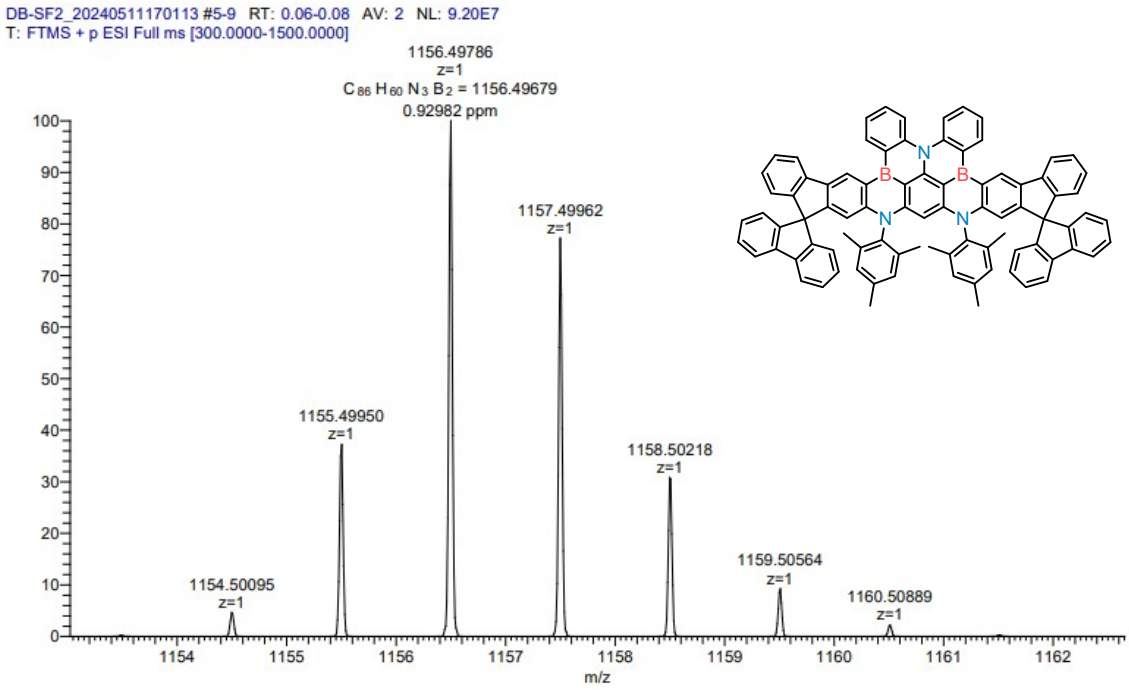


Figure S11. HR-MS spectrum of **DB-SF2**.

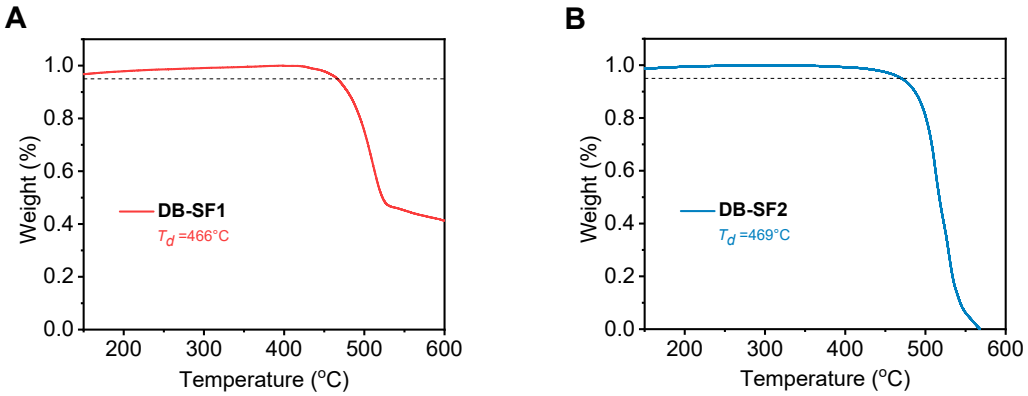


Figure S12. Thermal gravimetric analysis (TGA) curves at a heating rate of 10 °C min^{-1}.

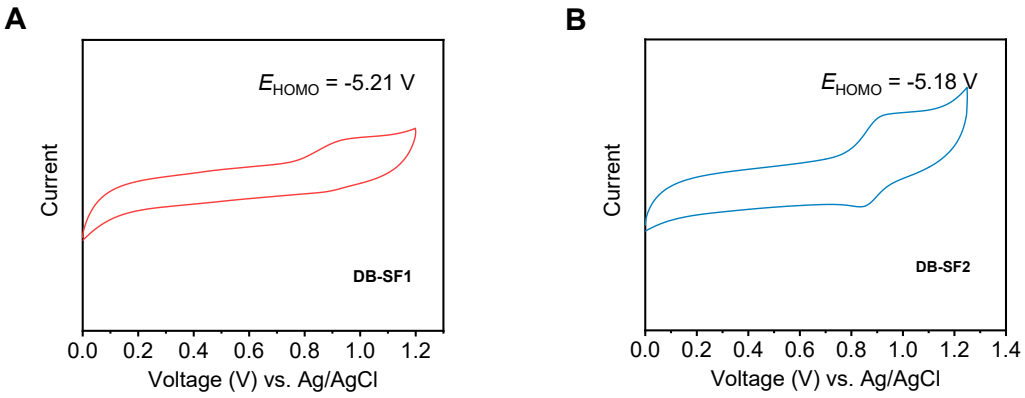


Figure S13. The oxidation curves obtained from the cyclic voltammetry (CV) measurement for the emitters.

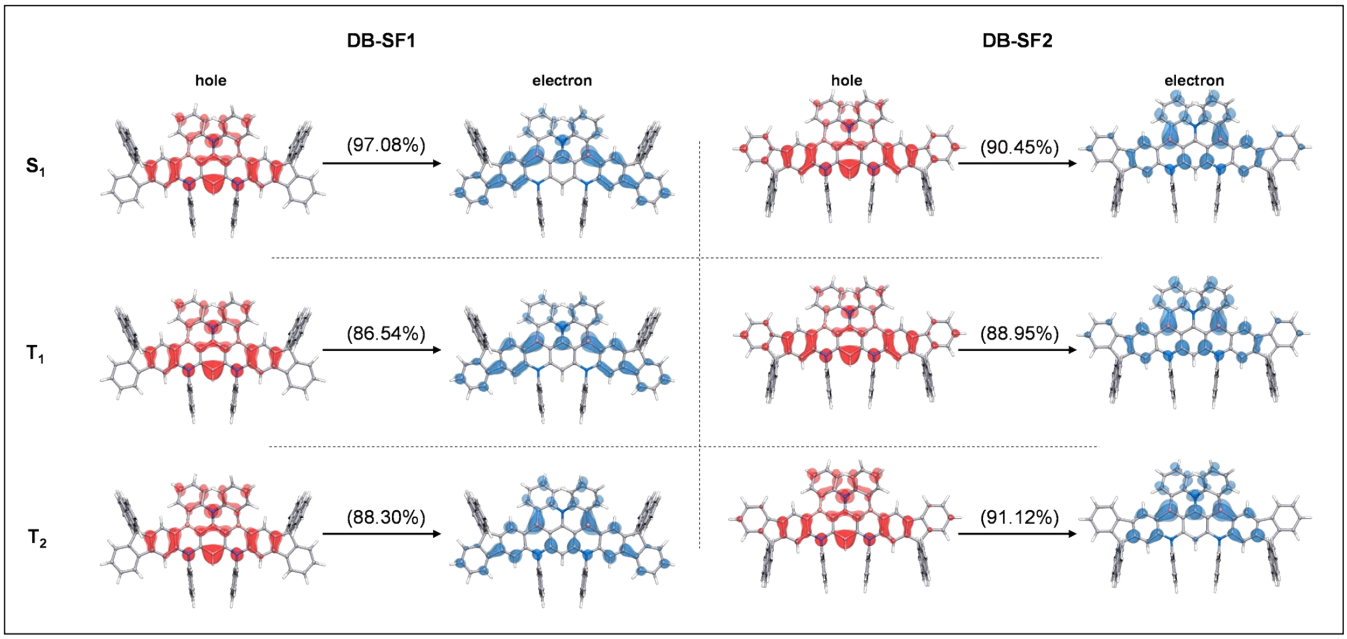


Figure S14. Natural transition orbital (NTO) distributions of the emitters with an iso-value of 0.025.

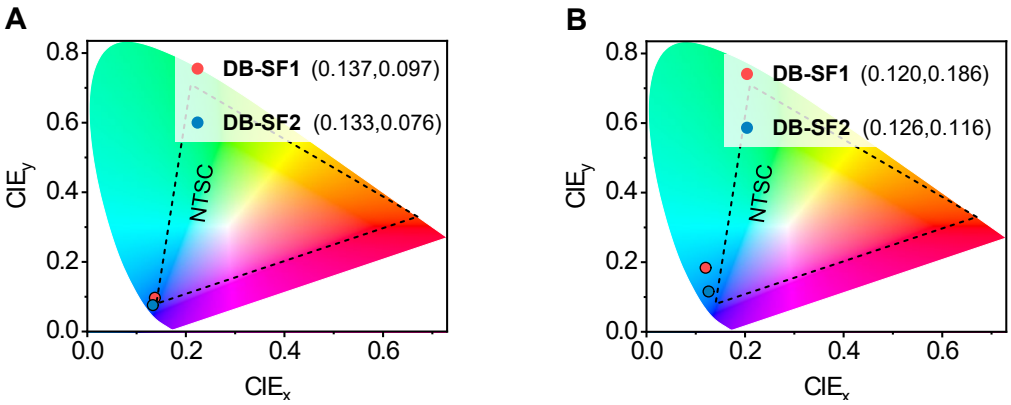


Figure S15. CIE color coordinates of the emitters in (A) 1×10^{-5} M toluene and (B) PMMA thin film (2 wt%).

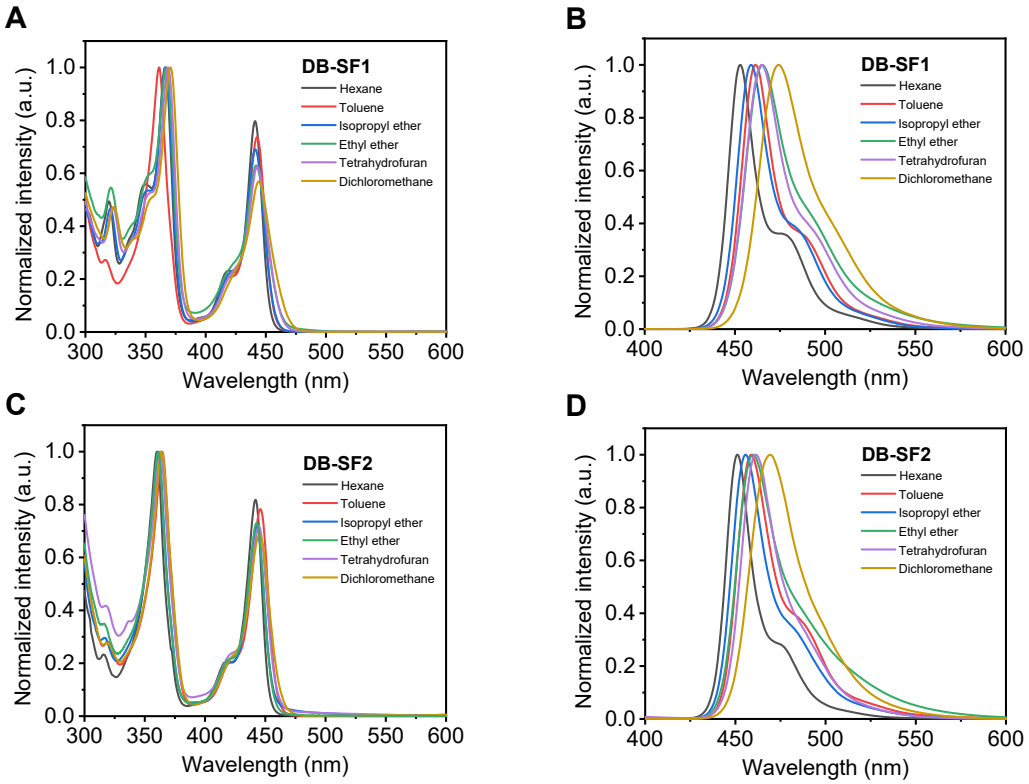


Figure S16. (A, C) Absorption and (B, D) photoluminescence (PL) spectra of **DB-SF1** and **DB-SF2** in different solvents at fixed concentration of 1×10^{-5} M.

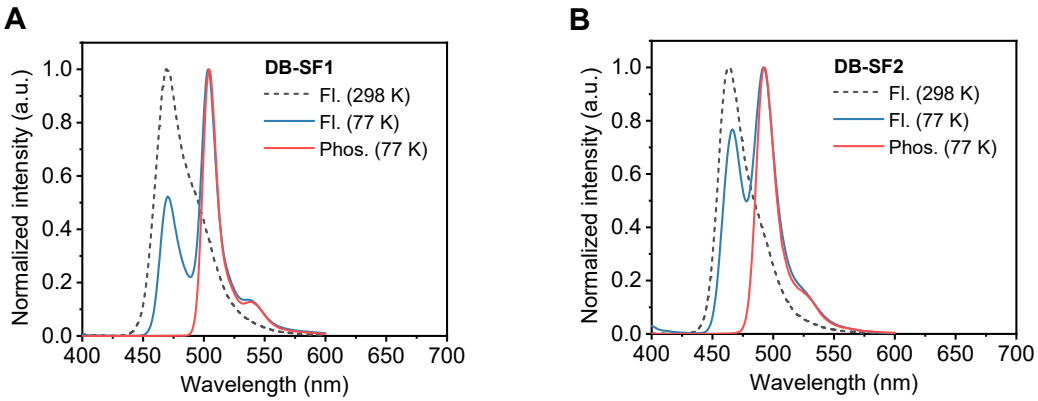


Figure S17. Normalized fluorescence (Fl.) at 298 K and 77 K, as well as phosphorescence spectra (Phos.) recorded at 77 K of the emitters in 2 wt% polymethyl methacrylate (PMMA) doped films. At 298 K, the fluorescence maxima and FWHM for **DB-SF1** and **DB-SF2** were observed at 469/35 nm and 464/31 nm, respectively. The S_1 and T_1 energy levels were determined from the peak maxima of fluorescence and phosphorescence at 77 K, yielding values of 2.64/2.46 eV for **DB-SF1** and 2.66/2.52 eV for **DB-SF2**,

corresponding to ΔE_{ST} of 0.18 eV and 0.14 eV, respectively.

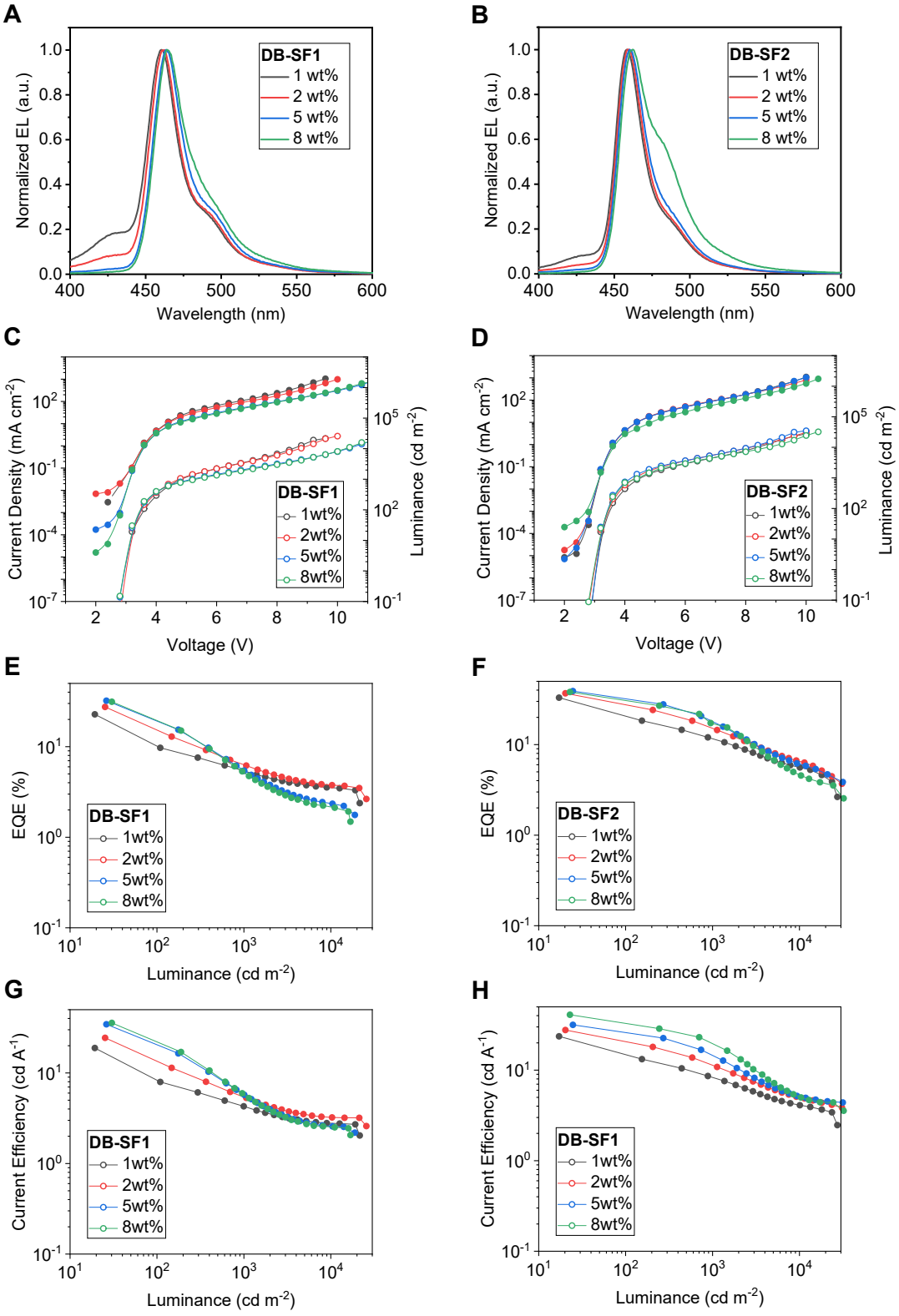


Figure S18. (A, B) EL spectra, (C, D) current density and luminance *vs* voltage (*J*-*V*-*L*) curves, (E, F) external quantum efficiency *vs* luminance (EQE-*L*) characteristics, (G, H) current efficiency *vs* luminance (CE-*L*) characteristics of the non-sensitized devices based on **DB-SF1** and **DB-SF2** at different doping ratios.

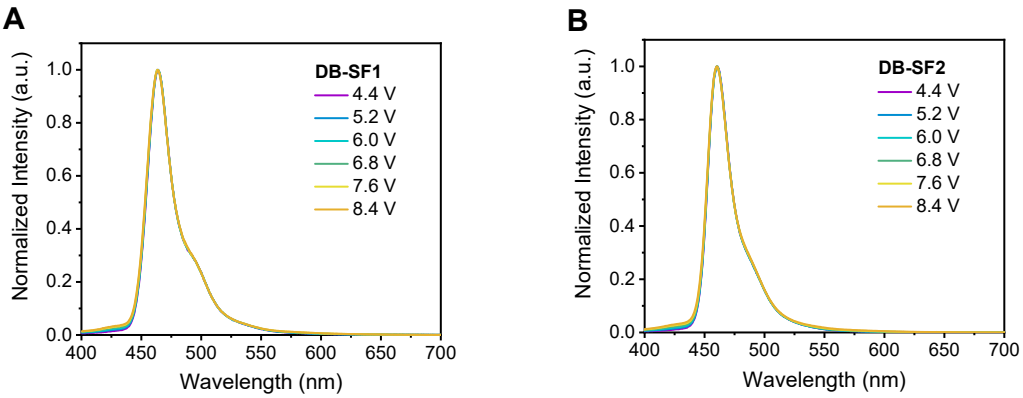


Figure S19. Electroluminescence (EL) of the non-sensitized devices with 5 wt% doping ratio based on (A) **DB-SF1**, (B) **DB-SF2** at different driving voltages.

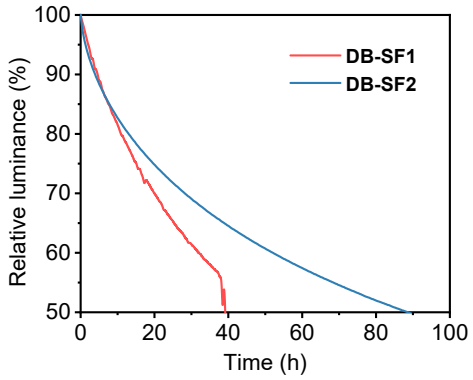


Figure S20. Operational lifetime measurement of the non-sensitized devices with 5 wt% doping ratio based on **DB-SF1** and **DB-SF2** at specified initial luminance of 100 cd m^{-2}.

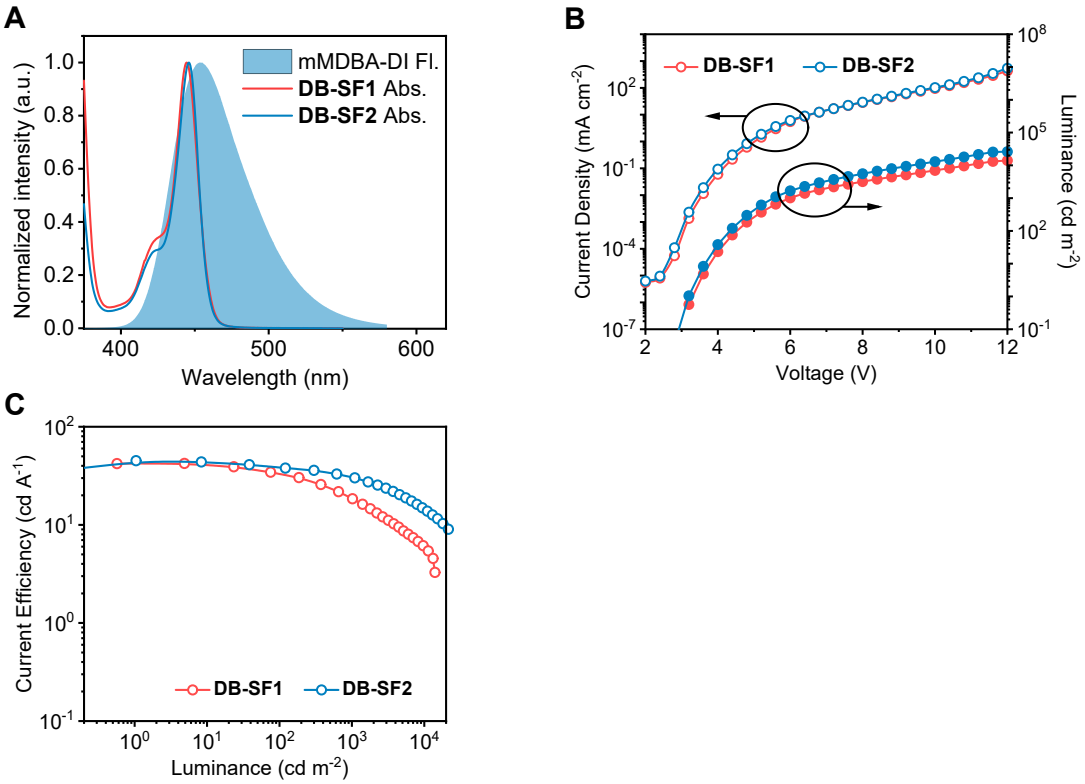


Figure S21. (A) Overlay of the sensitizer's PL spectrum with the absorption spectra of **DB-SF1** and **DB-SF2**. (B) *J*-*V*-*L*, and (C) CE-*L* characteristics of **DB-SF1** and **DB-SF2** based hyperfluorescence devices.

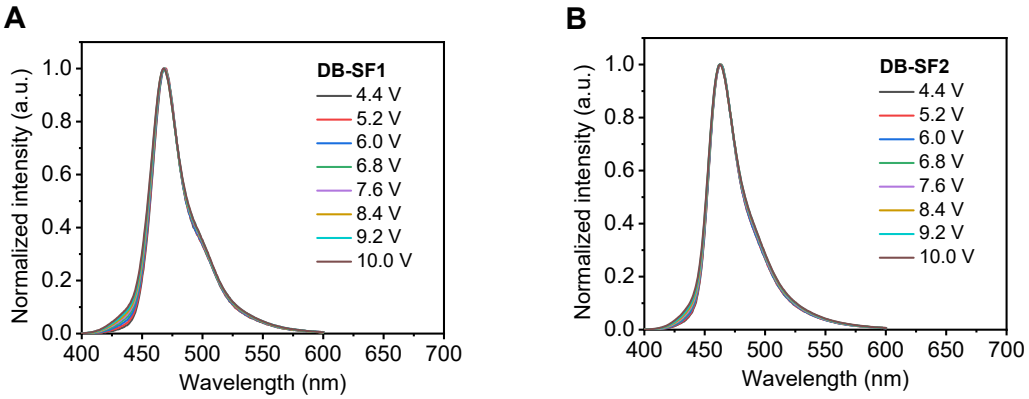


Figure S22. EL spectra of the hyperfluorescence devices based on (A) **DB-SF1**, (B) **DB-SF2** at different driving voltages.

Table S1. Crystal data and structure refinement for racemic **DB-SF1**.

Empirical formula	$C_{86}H_{59}B_2N_3$
Formula weight	1155.98
Temperature/K	170.00
Crystal system	triclinic
Space group	P-1
a/Å	13.8977(6)
b/Å	16.6183(7)
c/Å	20.3259(8)
α/°	113.4050(10)
β/°	95.369(2)
γ/°	91.274(2)
Volume/Å^3	4280.2(3)
Z	2
ρ_{calc}g/cm^3	0.897
μ/mm^{-1}	0.249
F(000)	1212.0
Crystal size/mm^3	0.09 × 0.07 × 0.05
Radiation	GaKα (λ = 1.34139)
2Θ range for data collection/°	4.148 to 121.496
Index ranges	$-18 \leq h \leq 18$, $-21 \leq k \leq 21$, $-26 \leq l \leq 26$
Reflections collected	93546
Independent reflections	19549 [R_{int} = 0.0610, R_{sigma} = 0.0545]
Data/restraints/parameters	19549/0/826
Goodness-of-fit on F^2	1.040
Final R indexes [I>=2σ (I)]	R_1 = 0.0861, wR_2 = 0.2770
Final R indexes [all data]	R_1 = 0.1026, wR_2 = 0.2896
Largest diff. peak/hole / e Å^{-3}	0.44/-0.39

Table S2. Crystal data and structure refinement for racemic **DB-SF2**.

Empirical formula	$C_{86}H_{59}B_2N_3$
Formula weight	1155.98
Temperature/K	170.00
Crystal system	triclinic
Space group	P-1
a/Å	14.1931(11)
b/Å	16.6449(12)
c/Å	17.7614(14)
α/°	71.081(3)
β/°	69.022(4)
γ/°	78.650(4)
Volume/Å^3	3690.7(5)
Z	2
ρ_{calc}g/cm^3	1.040
μ/mm^{-1}	0.289
F(000)	1212.0
Crystal size/mm^3	0.16 × 0.02 × 0.02
Radiation	GaKα (λ = 1.34139)
2Θ range for data collection/°	4.824 to 108.362
Index ranges	$-17 \leq h \leq 17$, $-18 \leq k \leq 20$, $-21 \leq l \leq 21$
Reflections collected	47417
Independent reflections	13490 [R_{int} = 0.1290, R_{sigma} = 0.1657]
Data/restraints/parameters	13490/0/826
Goodness-of-fit on F^2	1.067
Final R indexes [I>=2σ (I)]	R_1 = 0.1112, wR_2 = 0.3059
Final R indexes [all data]	R_1 = 0.1824, wR_2 = 0.3547
Largest diff. peak/hole / e Å^{-3}	0.58/-0.40

Table S3. Summary of TD-DFT data for **DB-SF1** and **DB-SF2**.

Compound	Transition	Wavelength (nm)	Energy (eV)	Oscillator Strength	Coefficient of Orbital
DB-SF1	S_0-S_1	428.29	2.8949	0.0710	HOMO→LUMO (0.9707)
	S_0-T_1	495.28	2.5033	0.0000	HOMO-3→LUMO+1 (0.0228) HOMO→LUMO (0.8653)
	S_0-T_2	475.36	2.6082	0.0000	HOMO-1→LUMO (0.0318) HOMO→LUMO+1 (0.8829)
DB-SF2	S_0-S_1	419.27	2.9572	0.6759	HOMO-1→LUMO+1 (0.0662) HOMO→LUMO (0.9045)
	S_0-T_1	477.13	2.5985	0.0000	HOMO-1→LUMO+1 (0.0321) HOMO→LUMO (0.8895)
	S_0-T_2	475.44	2.6078	0.0000	HOMO-2→LUMO+1 (0.0278) HOMO→LUMO+1 (0.8553) HOMO→LUMO+5 (0.0289)

Table S4. Cartesian coordinates of **DB-SF1** at the optimized S_0 geometry.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.203317	1.130044	-0.093115
2	6	0	-0.000008	1.835296	-0.000231
3	6	0	-1.203323	1.130037	0.092762
4	6	0	-1.231491	-0.288003	0.125253
5	6	0	-0.000003	-0.970713	-0.000163
6	6	0	1.231485	-0.288002	-0.125553
7	5	0	2.532020	-1.025915	-0.289476
8	6	0	3.807066	-0.177167	-0.157536
9	6	0	3.668815	1.243792	-0.147744
10	7	0	2.399261	1.841474	-0.174059
11	7	0	-0.000003	-2.370844	-0.000188
12	6	0	1.072400	-3.079375	-0.615795

13	6	0	2.364613	-2.501129	-0.713171
14	6	0	5.109372	-0.712475	-0.041133
15	6	0	6.214945	0.106424	0.017620
16	6	0	6.061699	1.507401	-0.035075
17	6	0	4.802689	2.082788	-0.108338
18	6	0	0.810053	-4.326842	-1.211957
19	6	0	1.823155	-5.030576	-1.845279
20	6	0	3.114432	-4.497672	-1.924219
21	6	0	3.360392	-3.246272	-1.380991
22	6	0	2.014956	5.187537	-1.731694
23	6	0	1.908735	6.020624	-0.612309
24	6	0	2.008691	5.448802	0.657819
25	6	0	2.200883	4.074164	0.831458
26	6	0	2.294697	3.272742	-0.312243
27	6	0	2.203443	3.811240	-1.604231
28	6	0	-1.072395	-3.079406	0.615419
29	6	0	-2.364599	-2.501140	0.712898
30	6	0	-3.360354	-3.246314	1.380709
31	6	0	-3.114400	-4.497774	1.923814
32	6	0	-1.823146	-5.030703	1.844754
33	6	0	-0.810055	-4.326935	1.211446
34	5	0	-2.532019	-1.025913	0.289247
35	6	0	-3.807070	-0.177156	0.157381
36	6	0	-3.668815	1.243804	0.147580
37	7	0	-2.399256	1.841475	0.173800
38	6	0	-5.109392	-0.712456	0.041081
39	6	0	-6.214965	0.106447	-0.017571
40	6	0	-6.061711	1.507425	0.035123
41	6	0	-4.802691	2.082802	0.108291
42	6	0	-2.008855	5.448847	-0.657968
43	6	0	-1.908550	6.020590	0.612172
44	6	0	-2.014440	5.187433	1.731533
45	6	0	-2.203004	3.811147	1.604036
46	6	0	-2.294644	3.272736	0.312042
47	6	0	-2.201103	4.074222	-0.831638
48	6	0	-7.393062	2.123087	-0.017416
49	6	0	-8.357855	1.100503	-0.102819
50	6	0	-7.689417	-0.273487	-0.120368
51	6	0	-8.153598	-1.202680	0.999140
52	6	0	-8.673234	-2.398022	0.467877
53	6	0	-8.595491	-2.331835	-0.998091

54	6	0	-8.025047	-1.097665	-1.361893
55	6	0	7.689387	-0.273515	0.120560
56	6	0	8.357829	1.100472	0.103083
57	6	0	7.393048	2.123061	0.017597
58	6	0	8.153675	-1.202712	-0.998900
59	6	0	8.673240	-2.398060	-0.467584
60	6	0	8.595346	-2.331873	0.998375
61	6	0	8.024881	-1.097697	1.362119
62	6	0	8.974328	-3.247447	1.980343
63	6	0	8.776882	-2.914317	3.322028
64	6	0	8.209335	-1.686017	3.677823
65	6	0	7.828362	-0.767082	2.694628
66	6	0	8.096482	-0.996006	-2.369259
67	6	0	8.569059	-2.000265	-3.220553
68	6	0	9.087728	-3.189903	-2.697818
69	6	0	9.143780	-3.398816	-1.318140
70	6	0	9.710672	1.401488	0.162699
71	6	0	10.102067	2.744031	0.136687
72	6	0	9.147120	3.763657	0.052422
73	6	0	7.785424	3.461958	-0.007819
74	6	0	-7.785431	3.461986	0.008037
75	6	0	-9.147131	3.763691	-0.052076
76	6	0	-10.102090	2.744067	-0.136254
77	6	0	-9.710702	1.401524	-0.162309
78	6	0	-8.974587	-3.247403	-1.980020
79	6	0	-8.777273	-2.914275	-3.321724
80	6	0	-8.209745	-1.685984	-3.677578
81	6	0	-7.828658	-0.767055	-2.694422
82	6	0	-8.096258	-0.995977	2.369494
83	6	0	-8.568757	-2.000232	3.220835
84	6	0	-9.087495	-3.189864	2.698153
85	6	0	-9.143696	-3.398772	1.318482
86	1	0	-0.000019	2.911375	-0.000295
87	1	0	5.234955	-1.788034	0.015994
88	1	0	4.686805	3.159316	-0.123772
89	1	0	-0.193874	-4.731045	-1.198126
90	1	0	1.595635	-5.987018	-2.306478
91	1	0	3.903622	-5.041204	-2.434080
92	1	0	4.340720	-2.802413	-1.510427
93	1	0	1.939263	5.618309	-2.727039
94	1	0	1.926217	6.082526	1.536853

95	1	0	-4.340661	-2.802440	1.510246
96	1	0	-3.903584	-5.041328	2.433659
97	1	0	-1.595619	-5.987191	2.305851
98	1	0	0.193858	-4.731170	1.197536
99	1	0	-5.234986	-1.788012	-0.016044
100	1	0	-4.686801	3.159329	0.123741
101	1	0	-1.926583	6.082622	-1.536986
102	1	0	-1.938441	5.618140	2.726883
103	1	0	9.414124	-4.202686	1.709227
104	1	0	9.065940	-3.616565	4.098014
105	1	0	8.062359	-1.445116	4.726133
106	1	0	7.385612	0.186721	2.965177
107	1	0	7.689778	-0.072228	-2.769531
108	1	0	8.532061	-1.856342	-4.295969
109	1	0	9.449362	-3.959695	-3.372778
110	1	0	9.544898	-4.325075	-0.917463
111	1	0	10.448355	0.607556	0.228633
112	1	0	11.156602	2.998049	0.182361
113	1	0	9.469612	4.800245	0.033320
114	1	0	7.046540	4.255129	-0.074308
115	1	0	-7.046538	4.255155	0.074457
116	1	0	-9.469618	4.800281	-0.032943
117	1	0	-11.156629	2.998090	-0.181830
118	1	0	-10.448394	0.607595	-0.228179
119	1	0	-9.414369	-4.202634	-1.708859
120	1	0	-9.066421	-3.616519	-4.097682
121	1	0	-8.062872	-1.445086	-4.725903
122	1	0	-7.385923	0.186743	-2.965016
123	1	0	-7.689499	-0.072206	2.769723
124	1	0	-8.531641	-1.856312	4.296247
125	1	0	-9.449066	-3.959651	3.373150
126	1	0	-9.544869	-4.325026	0.917844
127	1	0	2.274049	3.647738	1.810083
128	1	0	1.754244	7.073168	-0.727131
129	1	0	2.276374	3.183849	-2.467922
130	1	0	-1.754044	7.073129	0.727017
131	1	0	-2.274511	3.647853	-1.810270
132	1	0	-2.275706	3.183703	2.467708

Table S5. Cartesian coordinates of **DB-SF1** at the optimized S_1 geometry.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.203317	1.130044	-0.093115
2	6	0	-0.000008	1.835296	-0.000231
3	6	0	-1.203323	1.130037	0.092762
4	6	0	-1.231491	-0.288003	0.125253
5	6	0	-0.000003	-0.970713	-0.000163
6	6	0	1.231485	-0.288002	-0.125553
7	5	0	2.532020	-1.025915	-0.289476
8	6	0	3.807066	-0.177167	-0.157536
9	6	0	3.668815	1.243792	-0.147744
10	7	0	2.399261	1.841474	-0.174059
11	7	0	-0.000003	-2.370844	-0.000188
12	6	0	1.072400	-3.079375	-0.615795
13	6	0	2.364613	-2.501129	-0.713171
14	6	0	5.109372	-0.712475	-0.041133
15	6	0	6.214945	0.106424	0.017620
16	6	0	6.061699	1.507401	-0.035075
17	6	0	4.802689	2.082788	-0.108338
18	6	0	0.810053	-4.326842	-1.211957
19	6	0	1.823155	-5.030576	-1.845279
20	6	0	3.114432	-4.497672	-1.924219
21	6	0	3.360392	-3.246272	-1.380991
22	6	0	2.014956	5.187537	-1.731694
23	6	0	1.908735	6.020624	-0.612309
24	6	0	2.008691	5.448802	0.657819
25	6	0	2.200883	4.074164	0.831458
26	6	0	2.294697	3.272742	-0.312243
27	6	0	2.203443	3.811240	-1.604231
28	6	0	-1.072395	-3.079406	0.615419
29	6	0	-2.364599	-2.501140	0.712898
30	6	0	-3.360354	-3.246314	1.380709
31	6	0	-3.114400	-4.497774	1.923814
32	6	0	-1.823146	-5.030703	1.844754
33	6	0	-0.810055	-4.326935	1.211446
34	5	0	-2.532019	-1.025913	0.289247
35	6	0	-3.807070	-0.177156	0.157381

36	6	0	-3.668815	1.243804	0.147580
37	7	0	-2.399256	1.841475	0.173800
38	6	0	-5.109392	-0.712456	0.041081
39	6	0	-6.214965	0.106447	-0.017571
40	6	0	-6.061711	1.507425	0.035123
41	6	0	-4.802691	2.082802	0.108291
42	6	0	-2.008855	5.448847	-0.657968
43	6	0	-1.908550	6.020590	0.612172
44	6	0	-2.014440	5.187433	1.731533
45	6	0	-2.203004	3.811147	1.604036
46	6	0	-2.294644	3.272736	0.312042
47	6	0	-2.201103	4.074222	-0.831638
48	6	0	-7.393062	2.123087	-0.017416
49	6	0	-8.357855	1.100503	-0.102819
50	6	0	-7.689417	-0.273487	-0.120368
51	6	0	-8.153598	-1.202680	0.999140
52	6	0	-8.673234	-2.398022	0.467877
53	6	0	-8.595491	-2.331835	-0.998091
54	6	0	-8.025047	-1.097665	-1.361893
55	6	0	7.689387	-0.273515	0.120560
56	6	0	8.357829	1.100472	0.103083
57	6	0	7.393048	2.123061	0.017597
58	6	0	8.153675	-1.202712	-0.998900
59	6	0	8.673240	-2.398060	-0.467584
60	6	0	8.595346	-2.331873	0.998375
61	6	0	8.024881	-1.097697	1.362119
62	6	0	8.974328	-3.247447	1.980343
63	6	0	8.776882	-2.914317	3.322028
64	6	0	8.209335	-1.686017	3.677823
65	6	0	7.828362	-0.767082	2.694628
66	6	0	8.096482	-0.996006	-2.369259
67	6	0	8.569059	-2.000265	-3.220553
68	6	0	9.087728	-3.189903	-2.697818
69	6	0	9.143780	-3.398816	-1.318140
70	6	0	9.710672	1.401488	0.162699
71	6	0	10.102067	2.744031	0.136687
72	6	0	9.147120	3.763657	0.052422
73	6	0	7.785424	3.461958	-0.007819
74	6	0	-7.785431	3.461986	0.008037
75	6	0	-9.147131	3.763691	-0.052076
76	6	0	-10.102090	2.744067	-0.136254

77	6	0	-9.710702	1.401524	-0.162309
78	6	0	-8.974587	-3.247403	-1.980020
79	6	0	-8.777273	-2.914275	-3.321724
80	6	0	-8.209745	-1.685984	-3.677578
81	6	0	-7.828658	-0.767055	-2.694422
82	6	0	-8.096258	-0.995977	2.369494
83	6	0	-8.568757	-2.000232	3.220835
84	6	0	-9.087495	-3.189864	2.698153
85	6	0	-9.143696	-3.398772	1.318482
86	1	0	-0.000019	2.911375	-0.000295
87	1	0	5.234955	-1.788034	0.015994
88	1	0	4.686805	3.159316	-0.123772
89	1	0	-0.193874	-4.731045	-1.198126
90	1	0	1.595635	-5.987018	-2.306478
91	1	0	3.903622	-5.041204	-2.434080
92	1	0	4.340720	-2.802413	-1.510427
93	1	0	1.939263	5.618309	-2.727039
94	1	0	1.926217	6.082526	1.536853
95	1	0	-4.340661	-2.802440	1.510246
96	1	0	-3.903584	-5.041328	2.433659
97	1	0	-1.595619	-5.987191	2.305851
98	1	0	0.193858	-4.731170	1.197536
99	1	0	-5.234986	-1.788012	-0.016044
100	1	0	-4.686801	3.159329	0.123741
101	1	0	-1.926583	6.082622	-1.536986
102	1	0	-1.938441	5.618140	2.726883
103	1	0	9.414124	-4.202686	1.709227
104	1	0	9.065940	-3.616565	4.098014
105	1	0	8.062359	-1.445116	4.726133
106	1	0	7.385612	0.186721	2.965177
107	1	0	7.689778	-0.072228	-2.769531
108	1	0	8.532061	-1.856342	-4.295969
109	1	0	9.449362	-3.959695	-3.372778
110	1	0	9.544898	-4.325075	-0.917463
111	1	0	10.448355	0.607556	0.228633
112	1	0	11.156602	2.998049	0.182361
113	1	0	9.469612	4.800245	0.033320
114	1	0	7.046540	4.255129	-0.074308
115	1	0	-7.046538	4.255155	0.074457
116	1	0	-9.469618	4.800281	-0.032943
117	1	0	-11.156629	2.998090	-0.181830

118	1	0	-10.448394	0.607595	-0.228179
119	1	0	-9.414369	-4.202634	-1.708859
120	1	0	-9.066421	-3.616519	-4.097682
121	1	0	-8.062872	-1.445086	-4.725903
122	1	0	-7.385923	0.186743	-2.965016
123	1	0	-7.689499	-0.072206	2.769723
124	1	0	-8.531641	-1.856312	4.296247
125	1	0	-9.449066	-3.959651	3.373150
126	1	0	-9.544869	-4.325026	0.917844
127	1	0	2.274049	3.647738	1.810083
128	1	0	1.754244	7.073168	-0.727131
129	1	0	2.276374	3.183849	-2.467922
130	1	0	-1.754044	7.073129	0.727017
131	1	0	-2.274511	3.647853	-1.810270
132	1	0	-2.275706	3.183703	2.467708

Table S6. Cartesian coordinates of **DB-SF2** at the optimized S_0 geometry.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	1.201486	0.365425	0.105746
2	6	0	-0.000023	-0.340975	0.000133
3	6	0	-1.201518	0.365439	-0.105611
4	6	0	-0.000004	2.466389	0.000019
5	6	0	1.229445	1.783903	0.147012
6	5	0	2.527513	2.522379	0.331045
7	6	0	3.807493	1.677042	0.204902
8	6	0	3.663223	0.256863	0.173984
9	7	0	2.397550	-0.343765	0.183353
10	7	0	0.000001	3.867017	0.000005
11	6	0	1.060633	4.575303	0.634431
12	6	0	2.350732	3.996976	0.753806
13	6	0	5.108752	2.211521	0.085546
14	6	0	6.220140	1.388545	-0.015741
15	6	0	6.043181	-0.010216	0.018008
16	6	0	4.796839	-0.583285	0.114601
17	6	0	0.788046	5.822597	1.226475
18	6	0	1.789549	6.526000	1.878213
19	6	0	3.079116	5.992757	1.979892

20	6	0	3.333998	4.741876	1.439438
21	6	0	1.968103	-3.820207	1.418417
22	6	0	1.974915	-4.553488	0.228269
23	6	0	2.150088	-3.869326	-0.977724
24	6	0	2.316977	-2.483028	-1.017761
25	6	0	2.304986	-1.783514	0.196410
26	6	0	2.132209	-2.433627	1.425222
27	6	0	-1.060613	4.575283	-0.634451
28	6	0	-2.350748	3.996995	-0.753738
29	6	0	-3.334006	4.741907	-1.439354
30	6	0	-3.079092	5.992745	-1.979906
31	6	0	-1.789494	6.525922	-1.878353
32	6	0	-0.787993	5.822507	-1.226615
33	5	0	-2.527534	2.522398	-0.330962
34	6	0	-3.807516	1.677066	-0.204833
35	6	0	-3.663246	0.256878	-0.174001
36	7	0	-2.397580	-0.343752	-0.183281
37	6	0	-5.108779	2.211533	-0.085489
38	6	0	-6.220175	1.388555	0.015702
39	6	0	-6.043215	-0.010202	-0.018147
40	6	0	-4.796875	-0.583267	-0.114760
41	6	0	-2.150563	-3.869390	0.977692
42	6	0	-1.975030	-4.553488	-0.228279
43	6	0	-1.967790	-3.820134	-1.418384
44	6	0	-2.131855	-2.433554	-1.425166
45	6	0	-2.305012	-1.783500	-0.196368
46	6	0	-2.317412	-2.483083	1.017757
47	6	0	-1.229466	1.783910	-0.146959
48	6	0	7.651869	1.671567	-0.173440
49	6	0	8.349407	0.448350	-0.239445
50	6	0	7.383476	-0.728333	-0.122424
51	6	0	-7.383504	-0.728333	0.122310
52	6	0	-8.349440	0.448344	0.239349
53	6	0	-7.651902	1.671567	0.173409
54	6	0	7.395279	-1.685597	-1.311905
55	6	0	7.621857	-3.006357	-0.881790
56	6	0	7.800965	-2.999555	0.576779
57	6	0	7.677391	-1.675237	1.037948
58	6	0	-7.395191	-1.685528	1.311857
59	6	0	-7.621689	-3.006328	0.881826
60	6	0	-7.800905	-2.999615	-0.576730

61	6	0	-7.677452	-1.675310	-1.037977
62	6	0	8.051394	-4.035865	1.476442
63	6	0	8.177055	-3.732021	2.833697
64	6	0	8.054826	-2.413716	3.285994
65	6	0	7.802392	-1.373309	2.385837
66	6	0	7.181541	-1.396330	-2.651625
67	6	0	7.197822	-2.446390	-3.575637
68	6	0	7.423349	-3.761414	-3.154107
69	6	0	7.637102	-4.052233	-1.805091
70	6	0	8.346554	2.878555	-0.260731
71	6	0	9.734595	2.847254	-0.412883
72	6	0	10.421363	1.630341	-0.477151
73	6	0	9.727486	0.418375	-0.389973
74	6	0	-9.727523	0.418361	0.389842
75	6	0	-10.421404	1.630322	0.477055
76	6	0	-9.734637	2.847239	0.412855
77	6	0	-8.346592	2.878548	0.260733
78	6	0	-7.181373	-1.396172	2.651546
79	6	0	-7.197497	-2.446186	3.575613
80	6	0	-7.422945	-3.761252	3.154168
81	6	0	-7.636776	-4.052157	1.805184
82	6	0	-8.051380	-4.035980	-1.476312
83	6	0	-8.177213	-3.732212	-2.833569
84	6	0	-8.055111	-2.413921	-3.285944
85	6	0	-7.802632	-1.373457	-2.385866
86	1	0	-0.000041	-1.417322	0.000196
87	1	0	5.227529	3.288892	0.043691
88	1	0	4.689797	-1.660529	0.114061
89	1	0	-0.215426	6.226957	1.194997
90	1	0	1.554042	7.482096	2.336004
91	1	0	3.858340	6.535998	2.505262
92	1	0	4.310901	4.295144	1.584999
93	1	0	1.825768	-4.337480	2.363717
94	1	0	2.150995	-4.425147	-1.911655
95	1	0	-4.310943	4.295227	-1.584842
96	1	0	-3.858329	6.535986	-2.505258
97	1	0	-1.553948	7.481962	-2.336241
98	1	0	0.215507	6.226811	-1.195221
99	1	0	-5.227561	3.288900	-0.043550
100	1	0	-4.689840	-1.660511	-0.114294
101	1	0	-2.151784	-4.425256	1.911597

102	1	0	-1.825193	-4.337368	-2.363666
103	1	0	8.147899	-5.060753	1.130431
104	1	0	8.372831	-4.527533	3.546272
105	1	0	8.156875	-2.196801	4.344823
106	1	0	7.705712	-0.348516	2.731278
107	1	0	7.004760	-0.374099	-2.972194
108	1	0	7.034360	-2.239587	-4.628779
109	1	0	7.432020	-4.564693	-3.884597
110	1	0	7.809543	-5.074960	-1.483036
111	1	0	7.818869	3.826456	-0.211762
112	1	0	10.287591	3.779182	-0.482191
113	1	0	11.500466	1.626392	-0.595442
114	1	0	10.255038	-0.529502	-0.438923
115	1	0	-10.255077	-0.529518	0.438732
116	1	0	-11.500510	1.626366	0.595319
117	1	0	-10.287637	3.779163	0.482187
118	1	0	-7.818909	3.826453	0.211808
119	1	0	-7.004648	-0.373910	2.972048
120	1	0	-7.033977	-2.239315	4.628733
121	1	0	-7.431494	-4.564494	3.884701
122	1	0	-7.809152	-5.074916	1.483193
123	1	0	-8.147792	-5.060857	-1.130240
124	1	0	-8.373029	-4.527769	-3.546082
125	1	0	-8.157299	-2.197063	-4.344771
126	1	0	-7.706051	-0.348676	-2.731371
127	1	0	-2.450685	-1.970739	1.947618
128	1	0	-2.125505	-1.883188	-2.342748
129	1	0	-1.848864	-5.615955	-0.240411
130	1	0	2.126173	-1.883310	2.342835
131	1	0	2.449979	-1.970636	-1.947635
132	1	0	1.848701	-5.615949	0.240379

Table S7. Cartesian coordinates of **DB-SF2** at the optimized S_1 geometry.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-1.203014	0.339270	-0.092563
2	6	0	0.000000	-0.367339	-0.000002
3	6	0	1.203014	0.339269	0.092560

4	6	0	0.000000	2.439581	-0.000001
5	6	0	-1.230316	1.757657	-0.137959
6	5	0	-2.527342	2.498809	-0.320698
7	6	0	-3.809942	1.659913	-0.183095
8	6	0	-3.674429	0.239809	-0.136989
9	7	0	-2.406991	-0.363898	-0.155658
10	7	0	0.000000	3.840534	-0.000001
11	6	0	-1.059505	4.548534	-0.635459
12	6	0	-2.349534	3.970447	-0.753776
13	6	0	-5.107476	2.204356	-0.072635
14	6	0	-6.225021	1.390605	0.027266
15	6	0	-6.058370	-0.009948	0.013527
16	6	0	-4.813956	-0.592228	-0.062701
17	6	0	-0.785701	5.794653	-1.229418
18	6	0	-1.785957	6.496308	-1.884835
19	6	0	-3.075013	5.962153	-1.988296
20	6	0	-3.331203	4.713064	-1.444307
21	6	0	-2.083530	-3.875112	-1.354070
22	6	0	-2.032699	-4.567944	-0.143775
23	6	0	-2.137788	-3.874845	1.063362
24	6	0	-2.283867	-2.488651	1.062137
25	6	0	-2.324168	-1.800567	-0.150875
26	6	0	-2.229565	-2.488236	-1.359810
27	6	0	1.059506	4.548534	0.635458
28	6	0	2.349534	3.970446	0.753774
29	6	0	3.331203	4.713064	1.444304
30	6	0	3.075014	5.962152	1.988294
31	6	0	1.785958	6.496307	1.884834
32	6	0	0.785701	5.794653	1.229418
33	5	0	2.527342	2.498809	0.320697
34	6	0	3.809942	1.659913	0.183094
35	6	0	3.674429	0.239809	0.136987
36	7	0	2.406991	-0.363898	0.155655
37	6	0	5.107475	2.204355	0.072634
38	6	0	6.225020	1.390605	-0.027266
39	6	0	6.058370	-0.009948	-0.013527
40	6	0	4.813956	-0.592228	0.062700
41	6	0	2.137790	-3.874844	-1.063370
42	6	0	2.032697	-4.567944	0.143765
43	6	0	2.083524	-3.875113	1.354061
44	6	0	2.229560	-2.488237	1.359803

45	6	0	2.324168	-1.800567	0.150870
46	6	0	2.283870	-2.488650	-1.062142
47	6	0	1.230315	1.757657	0.137957
48	6	0	-7.656452	1.688815	0.153208
49	6	0	-8.366890	0.473296	0.210579
50	6	0	-7.409929	-0.713989	0.122328
51	6	0	7.409929	-0.713989	-0.122327
52	6	0	8.366890	0.473296	-0.210578
53	6	0	7.656452	1.688815	-0.153208
54	6	0	-7.486400	-1.671796	1.308506
55	6	0	-7.741983	-2.984507	0.869396
56	6	0	-7.866621	-2.972680	-0.594694
57	6	0	-7.685976	-1.652956	-1.049673
58	6	0	7.486401	-1.671798	-1.308503
59	6	0	7.741984	-2.984509	-0.869391
60	6	0	7.866621	-2.972679	0.594700
61	6	0	7.685975	-1.652954	1.049677
62	6	0	-8.117346	-4.000765	-1.503751
63	6	0	-8.185479	-3.693211	-2.864253
64	6	0	-8.006252	-2.379313	-3.310483
65	6	0	-7.753670	-1.347277	-2.400815
66	6	0	-7.323193	-1.388166	2.656344
67	6	0	-7.418025	-2.435274	3.578838
68	6	0	-7.670791	-3.742280	3.148148
69	6	0	-7.834736	-4.027745	1.791008
70	6	0	-8.340889	2.903176	0.216943
71	6	0	-9.732205	2.886275	0.337569
72	6	0	-10.432046	1.676331	0.393675
73	6	0	-9.748295	0.457267	0.329840
74	6	0	9.748296	0.457267	-0.329838
75	6	0	10.432046	1.676331	-0.393673
76	6	0	9.732205	2.886274	-0.337567
77	6	0	8.340889	2.903176	-0.216942
78	6	0	7.323197	-1.388171	-2.656342
79	6	0	7.418030	-2.435280	-3.578834
80	6	0	7.670796	-3.742285	-3.148141
81	6	0	7.834740	-4.027748	-1.791000
82	6	0	8.117344	-4.000762	1.503760
83	6	0	8.185476	-3.693206	2.864261
84	6	0	8.006248	-2.379307	3.310488
85	6	0	7.753667	-1.347273	2.400818

86	1	0	0.000000	-1.442839	-0.000004
87	1	0	-5.219050	3.282754	-0.043064
88	1	0	-4.719215	-1.670029	-0.046508
89	1	0	0.217792	6.198927	-1.196820
90	1	0	-1.549798	7.451376	-2.344440
91	1	0	-3.853078	6.503460	-2.517365
92	1	0	-4.307853	4.266315	-1.590935
93	1	0	-2.005216	-4.412490	-2.293748
94	1	0	-2.101588	-4.412000	2.005656
95	1	0	4.307854	4.266314	1.590932
96	1	0	3.853079	6.503460	2.517362
97	1	0	1.549800	7.451376	2.344438
98	1	0	-0.217792	6.198927	1.196819
99	1	0	5.219050	3.282754	0.043064
100	1	0	4.719214	-1.670030	0.046508
101	1	0	2.101593	-4.411997	-2.005665
102	1	0	2.005207	-4.412493	2.293738
103	1	0	-8.258053	-5.022100	-1.162389
104	1	0	-8.380836	-4.482223	-3.584157
105	1	0	-8.064119	-2.159247	-4.372005
106	1	0	-7.613030	-0.325987	-2.741468
107	1	0	-7.125827	-0.372216	2.984617
108	1	0	-7.294929	-2.232388	4.638235
109	1	0	-7.741303	-4.543289	3.877816
110	1	0	-8.031140	-5.043874	1.461840
111	1	0	-7.802868	3.845562	0.174241
112	1	0	-10.277542	3.823875	0.388616
113	1	0	-11.513544	1.683491	0.487468
114	1	0	-10.285903	-0.485219	0.372968
115	1	0	10.285903	-0.485219	-0.372966
116	1	0	11.513544	1.683491	-0.487466
117	1	0	10.277542	3.823874	-0.388615
118	1	0	7.802867	3.845562	-0.174241
119	1	0	7.125830	-0.372222	-2.984616
120	1	0	7.294934	-2.232396	-4.638231
121	1	0	7.741309	-4.543295	-3.877807
122	1	0	8.031142	-5.043877	-1.461830
123	1	0	8.258053	-5.022097	1.162399
124	1	0	8.380832	-4.482217	3.584166
125	1	0	8.064114	-2.159239	4.372010
126	1	0	7.613026	-0.325982	2.741470

127	1	0	2.355611	-1.929516	-1.988742
128	1	0	2.259246	-1.928962	2.288664
129	1	0	1.913388	-5.646765	0.140951
130	1	0	-2.259253	-1.928959	-2.288669
131	1	0	-2.355605	-1.929518	1.988736
132	1	0	-1.913391	-5.646765	-0.140963

Table S8. Vertical excitation energies for the singlet and triplet excited states estimated using the SCS-CC2 /cc-pVDZ method.

Compound	Transition	Wavelength (nm)	Energy (eV)
DB-SF1	S_0-S_1	393.76	3.1491
	S_0-T_1	414.27	2.9932
	S_0-T_2	395.93	3.1319
DB-SF2	S_0-S_1	394.57	3.1427
	S_0-T_1	406.34	3.0516
	S_0-T_2	398.39	3.1125

Table S9. Emission profiles of the emitters in different solvents and PMMA host.

Solvent	DB-SF1		DB-SF2	
	λ_{em} (nm)	FWHM (nm/meV)	λ_{em} (nm)	FWHM (nm/meV)
Hexane	453	18/108	451	17/104
Toluene	461	22/130	457	21/125
Isopropyl ether	459	22/129	455	22/129
Ethyl ether	465	31/177	459	29/171
Tetrahydrofuran	465	27/153	461	25/148
Dichloromethane	474	34/186	469	33/184
PMMA	469	35/201	464	31/174

Table S10. Summary of the non-sensitized device data.

Device (emitter)	Doping ratio	V_{on}[a] (V)	λ_{EL}[b] (nm)	FWHM[c] (nm)	L_{max}[d] (cd m^{-2})	CE_{max}[e] (cd A^{-1})	$EQE_{max/1000}$[f] (%)	CIE[g] (x, y)
DB-SF1	1%	2.8	461	22	21520	18.9	22.8/5.4	(0.14,0.09)
	2%	2.8	462	23	25663	24.5	27.5/6.2	(0.14,0.10)
	5%	2.8	464	24	18911	34.5	32.2/6.1	(0.13,0.12)
	8%	2.8	464	28	16817	35.7	31.4/5.2	(0.13,0.14)
DB-SF2	1%	2.8	459	21	27260	23.6	33.0/11.7	(0.14,0.08)
	2%	2.8	459	22	30861	27.7	36.9/15.4	(0.14,0.08)
	5%	2.8	460	22	34851	31.6	39.0/20.7	(0.13,0.09)
	8%	2.8	462	35	32236	40.9	38.2/17.2	(0.13,0.13)

[a)] Turn-on voltage recorded at the luminance of 1 cd m^{-2}; [b)] maximum EL wavelength; [c)] FWHM of EL spectra; [d)] maximum brightness; [e)] maximum current efficiency; [f)] efficiency at maximum and 1000 cd m^{-2}; [g)] EL color coordinates in the CIE 1931 chromaticity diagram recorded at 6 V.

Table S11. Summary of reported binary deep-blue TADF OLED with CIE_y < 0.12.

Emitter	Host	λ_{EL} (nm)	FWHM (nm)	$EQE_{max/100/1000}$ (%)	CIE (*x*, *y*)	Ref.
(*R*/*S*)-**DOBN**	2,6-DczPPy	459	38	23.9/12.5/8.6	0.14, 0.10	*Adv. Mater.* **2022**, 34, 2204253.
(*R*/*S*)-**DOBNT**	2,6-DczPPy	464	35	25.6/13.9/8.6	0.13, 0.12	*Adv. Mater.* **2022**, 34, 2204253.
QA-1	PPCz	455	49	17.1/-/-	0.14, 0.12	*Angew. Chem. Int. Ed.* **2021**, *60,* 7643-7648.
SPXZPO	mCP	448	-	6.3/4.5/1.8	0.16, 0.12	*Chem. Mater.* **2016**, 28, 5667.
m-*v*-DABNA	DBFPO	471	18	36.2/-/-	0.12, 0.12	*Chem. Eng. J.* **2022**, 432, 134381.
4F-*v*-DABNA	DBFPO	464	18	35.8/-/-	0.13, 0.08	*Chem. Eng. J.* **2022**, 432, 134381.
4F-m-*v*-DABNA	DBFPO	461	18	33.7/-/-	0.13, 0.06	*Chem. Eng. J.* **2022**, 432, 134381.
DTPPI	CBP	452	-	6.3/-/4.5	0.15, 0.12	*Adv. Funct. Mater.***2022**，2112969
CTPPI	CBP	396	-	7.9/-/3.9	0.16, 0.04	*Adv. Funct. Mater.***2022**，2112969
CBPI	CBP	428	-	5.4/-/2.6	0.15, 0.06	*Adv. Funct. Mater.***2022**，2112969
***v*-DABNA**	DOBNA-Oar	469	18	34.4/32.8/26.0	0.12, 0.11	*Nat. Photonics.* **2019**, 13, 678.
DABNA-NP-TB	DOBNA-Tol	457	33	19.5/17.5/12.0	0.14, 0.11	*Angew. Chem. Int. Ed.* **2021**, 60, 2882-2886.
CzPh-CNNPI	CBP	436	72	9.0/- /3.1	0.155, 0.102	*Adv. Funct. Mater.* **2020**, 30, 2002323.
***N*-DABNA-O-Me**	DOBNA-Tol	465	23	29.5/23.8/22.1	0.13, 0.10	*Angew. Chem. Int. Ed.* **2021**, 60, 17910 – 17914.
4	DPEPO	455	-	16.0/12.3/-	0.141, 0.099	*Adv. Opt. Mater.* **2018**, 6, 1800385.
5	DPEPO	449	-	9.3/4.1/-	0.151, 0.058	*Adv. Opt. Mater.* **2018**, 6, 1800385.

Cz-TRZ3	DPEPO	450	-	19.2/-/-	0.148, 0.098	*Angew. Chem. Int. Ed.* **2017**, 56, 1571-1575.
Cz-TRZ4	DPEPO	450	-	18.3/-/-	0.150, 0.097	*Angew. Chem. Int. Ed.* **2017**, 56, 1571-1575.
TDBA-SAF	DPEPO	456	55	28.2/24.2/17.6	0.142, 0.090	*Adv. Mater.* **2020**, 32, 2004083.
DABNA-1	mCBP	459	28	13.5/6.4/-	0.13, 0.09	*Adv. Mater.* **2016**, 28, 2777-2781.
IczCz	DPEPO	416	49	2.3/-/-	0.17, 0.04	*ACS Appl. Mater. Interfaces* **2017**, 9, 37864-37872.
IczAc	DPEPO	454	56	13.7/-/-	0.15, 0.09	*ACS Appl. Mater. Interfaces* **2017**, 9, 37864-37872.
TDBA-PAS	DPEPO	435	50	22.35/17.99/-	0.155, 0.042	*Adv.Mater.* **2022**, 34, 2200537
TDBA-DPAC	DPEPO	450	60	24.61/11.03/2.42	0.148, 0.085	*Adv.Mater.* **2022**, 34, 2200537
Ban-(3,5)-CF3	CBP	435	66	5.0/- /-	0.156, 0.083	*J. Mater. Chem. C* **2013**, 1, 8117- 8127.
Icz-DPS	DPEPO	435	-	11.6/-/10.9	0.15, 0.08	*ACS Appl. Mater. Interfaces* **2021**, 13, 57713-57724.
pDTCz-DPS	PPT	428	-	2.7/2.6/ -	0.15, 0.08	*J. Mater. Chem. C* **2019**, 7, 6664-6671.
pDTCz-DPS	DPEPO	425	-	4.6/3.2/ -	0.15, 0.08	*J. Mater. Chem. C* **2019**, 7, 6664-6671.
DTC-mBPSB	DPEPO	440	-	5.5/-/-	0.15, 0.08	*Chem. Commun*. **2015**, 51, 16353.
CNICCz	DPEPO	449	56	12.4/6.4/-	0.15, 0.08	*J. Mater. Chem. C* **2018**, 6, 5012-5017.
PXB-mIC	PPBI	450	-	12.5/-/5.2	0.15, 0.08	*ACS Appl. Mater. Interfaces* **2019**, 11, 14909-14916.
PPINCN-Cz	CBP	430	-	9.9/-/-	0.15, 0.08	*J. Mater. Chem. C* **2021**, 9, 15683- 15697.

BOBO-Z	mCBP	445	18	13.6/9.8/3.3	0.15, 0.04	*Adv. Mater.* **2022**, 34, 2107951.
BOBS-Z	mCBP	456	23	26.9/24.0/15.0	0.14, 0.06	*Adv. Mater.* **2022**, 34, 2107951.
BSBS-Z	mCBP	463	22	26.8/24.0/15.9	0.13, 0.08	*Adv. Mater.* **2022**, 34, 2107951.
PAB	mCP	456	31	14.7/-/＜2	0.145, 0.076	*Org. Electron.* **2021**, 97, 106275.
2tPAB	mCP	456	27	16.8/-/＜2	0.145, 0.076	*Org. Electron.* **2021**, 97, 106275.
3tPAB	mCP	460	26	19.3/-/＜2	0.141, 0.076	*Org. Electron.* **2021**, 97, 106275.
DIDOBNA-N	TSPO1	429	70	15.2/3.8/-	0.152, 0.073	*Adv. Mater.***2023**, 35, 2300997
MesB-DIDOBNA-N	TSPO1	405	31	13.6/-/-	0.165, 0.055	*Adv. Mater.***2023**, 35, 2300997
tBDCz-DPS	DPEPO	423	-	9.9/-/-	0.15, 0.07	*J. Am. Chem. Soc.* **2012**, 134, 14706-14709.
DCzBN1	DPEPO	418	65	2.5/0.1/-	0.151, 0.047	*Adv. Funct. Mater.* **2018**, 28, 1706023.
DCzBN2	DPEPO	436	66	7.7/3.3/-	0.153, 0.069	*Adv. Funct. Mater.* **2018**, 28, 1706023.
DCzBN3	DPEPO	428	65	10.3/5.4/-	0.156, 0.063	*Adv. Funct. Mater.* **2018**, 28, 1706023.
NOBNacene	TSPO1	412	41	11.2/-/-	0.176, 0.068	*Angew. Chem. Int. Ed.***2023**,62, e202215522
CZ2CO	mCP	445	23	13/9.9/3.9	0.154, 0.061	*Angew. Chem. Int. Ed.* **2023**, 62, e202215226
CZCO	mCP	432	35	15.6/6/5.9	0.154, 0.047	*Angew. Chem. Int. Ed.* **2023**, 62, e202215226

PyINA	PIMNA	432	-	5.1/5.1/4.7	0.156, 0.060	*ACS Appl. Mater. Interfaces* **2016**,8,28771-28779.
TDBA-Ac	PPBI	448	48	21.5/-/9.7	0.15, 0.06	*Nat. Photonics* **2019**, 13, 540-546.
BD1	CBP	424	44	8.9/4.3/3.7	0.16, 0.05	*Adv. Funct.Mater.***2014**, 24, 2064–2071
BD2	CBP	430	47	9.5/5.2/4.0	0.15, 0.06	*Adv. Funct.Mater.***2014**, 24, 2064–2071
BD3	CBP	432	45	12.0/5.3/4.2	0.15, 0.06	*Adv. Funct.Mater.***2014**, 24, 2064–2071
TB-tCz	mCP	412	44	15.9/-/-	0.17, 0.06	*Adv. Funct. Mater.* **2021**, 31, 2102588
TB-tPCz	mCP	420	44	14.1/-/-	0.17, 0.05	*Adv. Funct. Mater.* **2021**, 31, 2102588
TPIBCz	CBP	432	-	5.5/-/-	0.156, 0.047	*Chem. Sci.* **2017**, 8, 3599-3608.
TPINCz	CBP	440	-	7.0/-/6.6	0.153, 0.059	*Chem. Sci.* **2017**, 8, 3599-3608.
TPIBNCz	CBP	428	-	6.0/-/-	0.157, 0.051	*Chem. Sci.* **2017**, 8, 3599-3608.
B-O-dpa	DPEPO	443	32	16.3/2.2/-	0.15, 0.05	*ACS Appl Mater Interfaces* **2021**, 13, 45798.
DSiTPI	CzSi	396	47	7.4/5.0/-	0.16, 0.05	*Adv. Opt. Mater.* **2021**, 9, 2100965.
CSiTPI	CzSi	382	46	5.2/4.4/-	0.16, 0.05	*Adv. Opt. Mater.* **2021**, 9, 2100965.
3a	CBP	422	54	3.0/-/-	0.15, 0.046	*J. Mater. Chem. C* **2021**, 9, 260-269.
3b	CBP	422	58	3.0/-/-	0.15, 0.049	*J. Mater. Chem. C* **2021**, 9, 260-269.
3c	CBP	423	59	3.0/-/-	0.15, 0.043	*J. Mater. Chem. C* **2021**, 9, 260-269.
DMACN-B	DPEPO	444	44	10.0/-/-	0.151, 0.045	*Adv. Funct. Mater.* **2021**, 31, 2009488
BIC-mCz	DBFPO	431	38	7.0/-/-	0.16, 0.04	*Angew. Chem. Int. Ed.* **2022**, 61, e202206916.
mDBIC	DBFPO	433	34	5.7/-/-	0.16, 0.04	*Angew. Chem. Int. Ed.* **2022**, 61,

						e202206916.
POPCN-2CP	mCP	404	51	7.9/7.7/6.2	0.159, 0.031	*Angew. Chem. Int. Ed.* **2022**, 61, e202116810.
POPCN-2CP	DBFPO	404	50	8.2/7.9/5.7	0.161, 0.034	*Angew. Chem. Int. Ed.* **2022**, 61, e202116810.
5	mCPPO1	412	-	3.1/-/-	0.16, 0.024	*J. Mater. Chem. C,* **2014**, 2, 9083–9086
TFPy2	mCP	414	51	3.8/-/-	0.15, 0.03	*Adv. Opt. Mater.* **2020**, 8, 2001074.
TFPy3	mCP	406	50	4.8/-/-	0.16, 0.02	*Adv. Opt. Mater.* **2020**, 8, 2001074.
TFPy4	mCP	404	54	5.1/-/-	0.16, 0.02	*Adv. Opt. Mater.* **2020**, 8, 2001074.
DB	DOBNA-OAr	443	26	23.4/17.4/9.0	0.154, 0.048	*Adv. Mater.***2023**, 2308314
DB-O	DOBNA-OAr	445	24	27.5/21.3/11.4	0.150, 0.041	*Adv. Mater.***2023**, 2308314
DB-S	DOBNA-OAr	447	24	29.3/23.6/12.2	0.148, 0.047	*Adv. Mater.***2023**, 2308314
DB-SF1	SiTrzCz2	465	24	32.2/5.4	0.13,0.12	This work
DB-SF2	SiTrzCz2	460	22	39.0/18.5	0.13,0.09	This work

3. References

1 V. Dolomanov, L. J. Bourhis, R. J. Gildea, J. A. Howard, H. Puschmann, *J. Appl. Cryst.* **2009**, *42*, 339-341.

2 G. M. Sheldrick, *Acta Cryst.* **2015**, *A71*, 3-8.

3 G. M. Sheldrick, *Acta Cryst.* **2015**, *C71*, 3-8.

4 T. Lu, F. Chen, *J. Comput. Chem.* **2012**, *33*, 580-592.

5 Z. Liu, T. Lu, Q. Chen, *Carbon* **2020**, *165*, 461-467.

6 M. Kállay, P. R. Nagy, D. Mester, Z. Rolik, G. Samu, J. Csontos, J. Csóka, P. B. Szabó, L. Gyevi-Nagy, B. Hégely, I. Ladjánszki, L. Szegedy, B. Ladóczki, K. Petrov, M. Farkas, P. D. Mezei, Á. Ganyecz, *J. Chem. Phys.* **2020**, *152*, 074107.

7 M. Kállay, P. R. Nagy, D. Mester, L. Gyevi-Nagy, J. Csóka, P. B. Szabó, Z. Rolik, G. Samu, J. Csontos, B. Hégely, Á. Ganyecz, I. Ladjánszki, L. Szegedy, B. Ladóczki, K. Petrov, M. Farkas, P. D. Mezei, R. A. Horváth, MRCC, a quantum chemical program suite. See www.mrcc.hu.

8 X. Gao, S. Bai, D. Fazzi, T. Niehaus, M. Barbatti, W. Thiel, *J. Chem. Theory Comput.* **2017**, *13*, 515-524.

9 Q. Zhang, H. Kuwabara, W. J. Potscavage, S. Huang, Y. Hatae, T. Shibata, C. Adachi, *J. Am. Chem. Soc.* **2014**, *136*, 18070-18081.

10 Q. Zhang, B. Li, S. Huang, H. Nomura, H. Tanaka, C. Adachi, *Nat. Photonics* **2014**, *8*, 326-332.

11 T.-L. Wu, M.-J. Huang, C.-C. Lin, P.-Y. Huang, T.-Y. Chou, R.-W. Chen-Cheng, H.-W. Lin, R.-S. Liu, C.-H. Cheng, *Nat. Photonics* **2018**, *12*, 235-240.