Supporting Information

Chiral BINOL-Bridged Imidazole Dimer Possessing Sub-Millisecond Fast Photochromism

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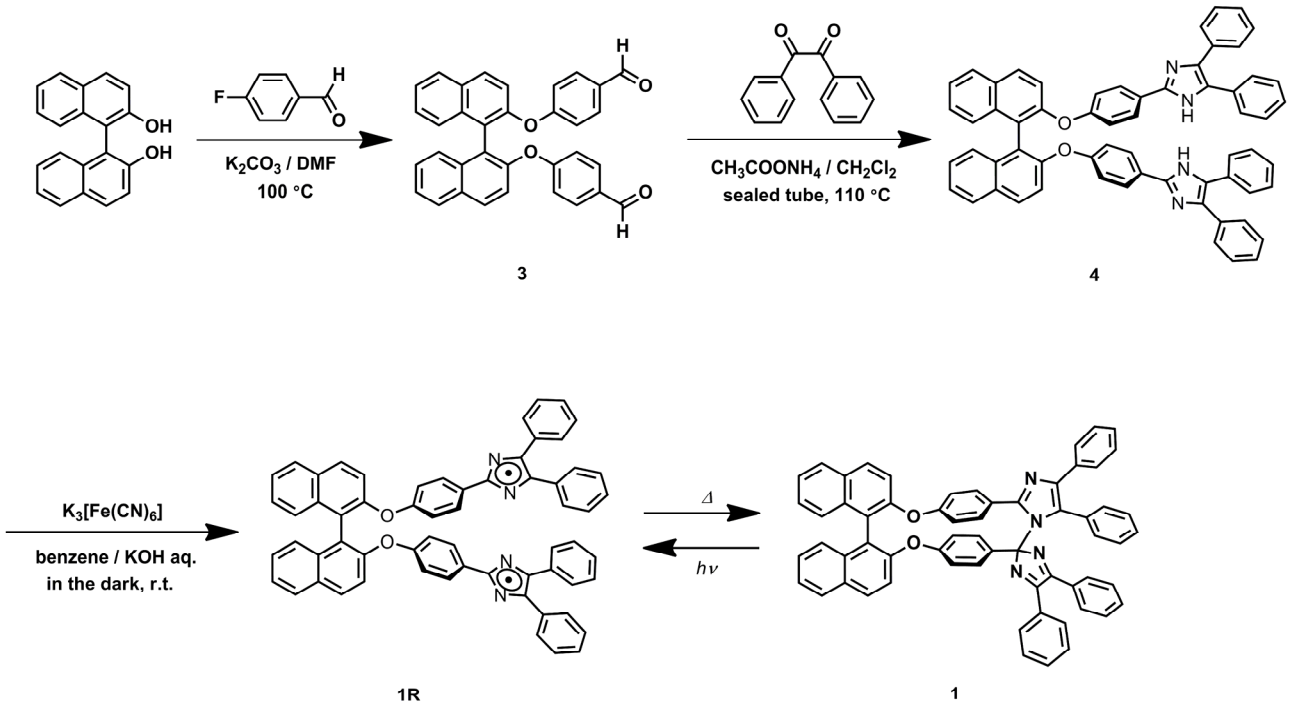
1. Synthesis

All reactions were monitored by thin-layer chromatography carried out on 0.2 mm E. Merck silica gel plates (60F-254). Column chromatography was performed on the silica gel (Silica Gel 60N (spherical, neutral), 40-50 μm, Kanto Chemical Co., Inc.). ^{1}H-NMR spectra and ^{13}C-NMR spectra were recorded on a Bruker AVANCE III 400 NanoBay. DMSO-*d*$_6$, $CDCl_3$ and CD_2Cl_2 were used as deuterated solvent. MASS spectra (ESI-TOF-MS) were measured by using a Bruker micrOTOF II-AGA1. All reagents were purchased from Tokyo Chemical Industry Co., Ltd., Wako Pure Chemical Industries, Ltd., and Aldrich Chemical Company, Inc. and were used without further purification. All reaction solvents were distilled on the appropriate drying reagents prior to use.

1.1 Synthesis of 1

Scheme S1. Synthetic procedure of **1**.



Compound 3

1,1'-Bi-2-naphthol (2.00 g, 6.99 mmol), 4-fluorobenzaldehyde (1.80 mL, 17.1 mmol) and potassium carbonate (2.42 g, 17.5 mmol) were stirred at 100 °C in DMF (50 mL). After 42 h, the reaction mixture was cooled to room temperature and the target compound was extracted with CH_2Cl_2. The organic phase was washed with water, dried, and evaporated. The residue was purified by column chromatography over silica gel with $CHCl_3$ as eluent to give a light yellow solid (1.28 g, 2.58 mmol, yield; 37 %). ^{1}H NMR (400 MHz, DMSO-d_6) : δ 9.79 (s, 2H), 8.13 (d, J = 9.0 Hz, 2H), 8.04 (d, J = 8.2 Hz, 2H), 7.69 (d, J = 8.6 Hz, 4H), 7.50 (t, J = 7.6, 9.0 Hz, 2H), 7.43-7.32 (m, 4H), 7.17 (d, J = 8.2 Hz, 2H), 6.92 (d, J = 8.6 Hz, 4H). HR-MS (ESI+) calculated for $C_{34}H_{23}O_4$ $[M+H]^+$ 495.1596, found 495.1614.

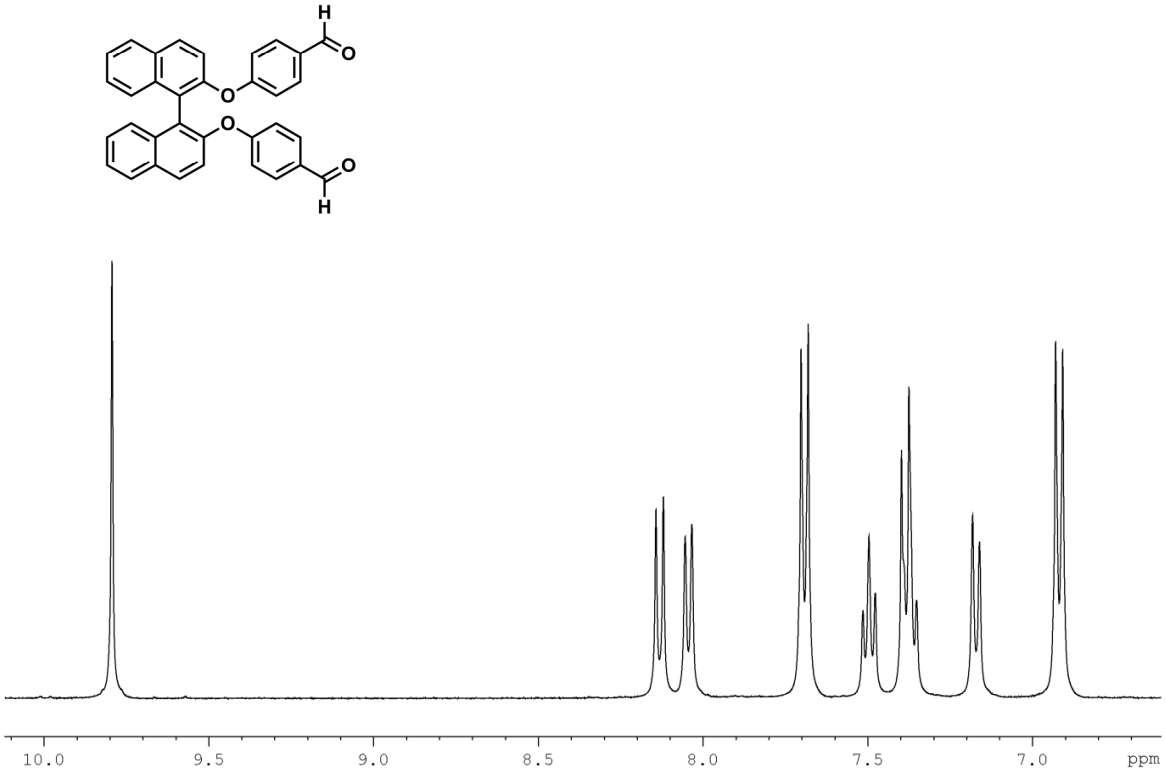


Figure S1. ^{1}H NMR spectrum of **3** in DMSO-d_6.

Compound 4

Compound **3** (150 mg, 0.303 mmol), benzil (220 mg, 1.05 mmol), and ammonium acetate (350mg, 4.54 mmol) were stirred at 110 °C in CH_2Cl_2 (1.0 mL) in a sealed tube. After 14 h, the reaction mixture was cooled to room temperature and washed with water, followed by ethanol. The precipitate was filtered and washed with hexane to give a white powder (194 mg, 0.222 mmol, yield; 73 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 12.50 (s, 2H), 8.11 (d, J = 8.7 Hz, 2H), 8.04 (d, J = 7.8, 2H), 7.94 (d, J = 7.8 Hz, 4H), 7.53 – 7.17 (m, 28H), 6.93 (d, J = 7.8 Hz, 4H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: 156.91, 151.59, 145.01, 136.88, 135.15, 133.48, 131.04, 130.21, 128.47, 128.25, 128.02, 127.86, 127.54, 126.97, 126.66, 126.34, 125.35, 125.06, 124.98, 121.65, 119.35, 118.15; HR-MS (ESI+) calculated for $C_{62}H_{43}N_4O_2$ $[M+H]^+$ 875.3381, found 875.3297.

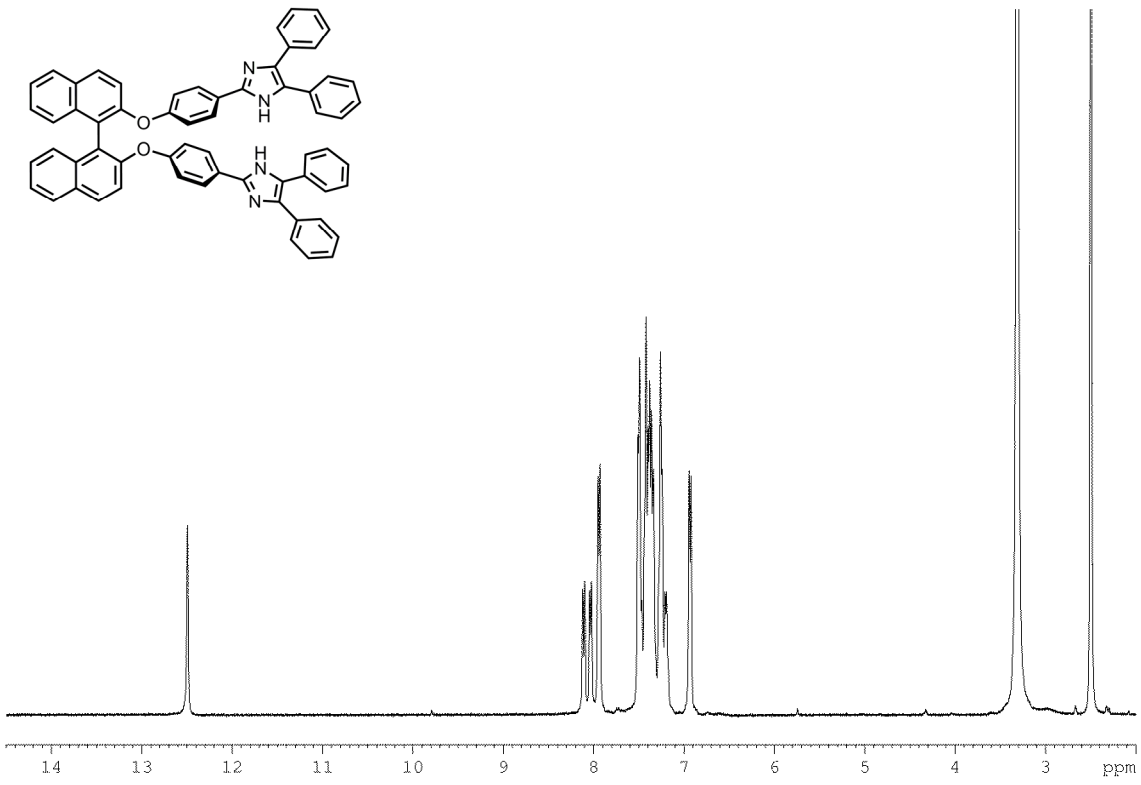


Figure S2. ^{1}H NMR spectrum of **4** in DMSO-*d*$_6$.

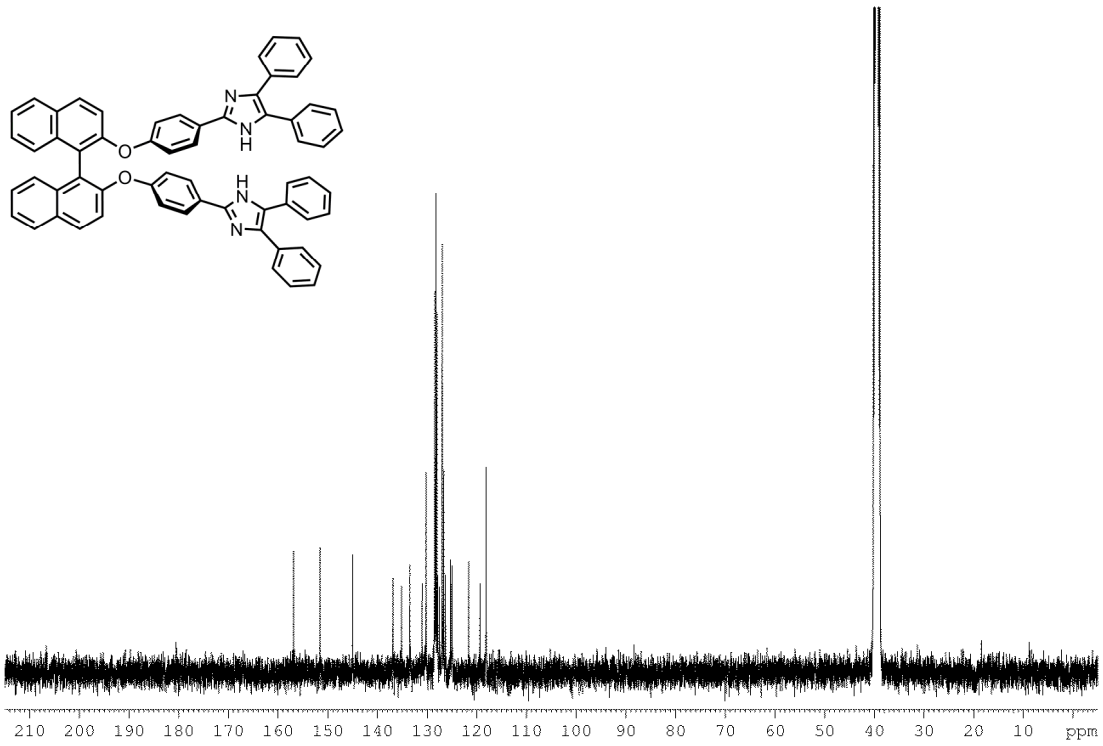


Figure S3. ^{13}C NMR spectrum of **4** in DMSO-*d*$_6$.

Compound 1

All manipulations were carried out with the exclusion of light. Under nitrogen, a solution of potassium ferricyanide (11.8 g, 35.8 mmol) and potassium hydroxide (4.05 g, 72.2 mmol) in water (200 mL) was added to a solution of compound **4** (629 mg, 0.718 mmol) in benzene (200 mL). The reaction mixture was vigorously stirred at room temperature for 2 h. The organic layer was washed with water, dried, and evaporated. The residue was purified by column chromatography over silica gel with AcOEt/hexane = 1/3 as eluent and followed by recrystallization from $CHCl_3$/hexane to give a slightly green crystal of **3** (129 mg, 0.148 mmol, yield; 21 %). ^{1}H NMR (400 MHz, CD_2Cl_2) δ: 8.13 (d, J = 8.9 Hz, 1H), 8.06-7.95 (m, 3H), 7.66-7.02 (m, 32H), 6.93 (d, J = 8.8 Hz, 1H), 6.75 (d, J = 8.2 Hz, 1H), 6.61 (d, J = 8.7 Hz, 1H), 6.41 (d, J = 8.4 Hz, 1H), 6.06 (d, J = 8.5 Hz, 1H), 5.94 (d, J = 8.8 Hz, 1H); ^{13}C NMR (100 MHz, CD_2Cl_2) δ: 166.82, 166.34, 160.09, 158.96, 150.23, 149.92, 148.39, 138.14, 135.26, 135.23, 135.07, 134.75, 133.50, 133.39, 132.37, 132.34, 132.12, 131.89, 131.85, 131.36, 131.32, 131.27, 131.16, 130.77, 130.38, 130.24, 129.90, 129.83, 129.74, 128.92, 128.82, 128.70, 128.61, 128.46, 128.42, 128.27, 128.19, 128.04, 127.45, 127.33, 127.27, 126.53, 126.20, 126.13, 124.93, 124.34, 123.51, 122.47, 116.59, 115.81, 113.98, 113.17; HR-MS (ESI+) calculated for $C_{62}H_{41}N_4O_2$ $[M+H]^+$ 873.3224, found 873.3113.

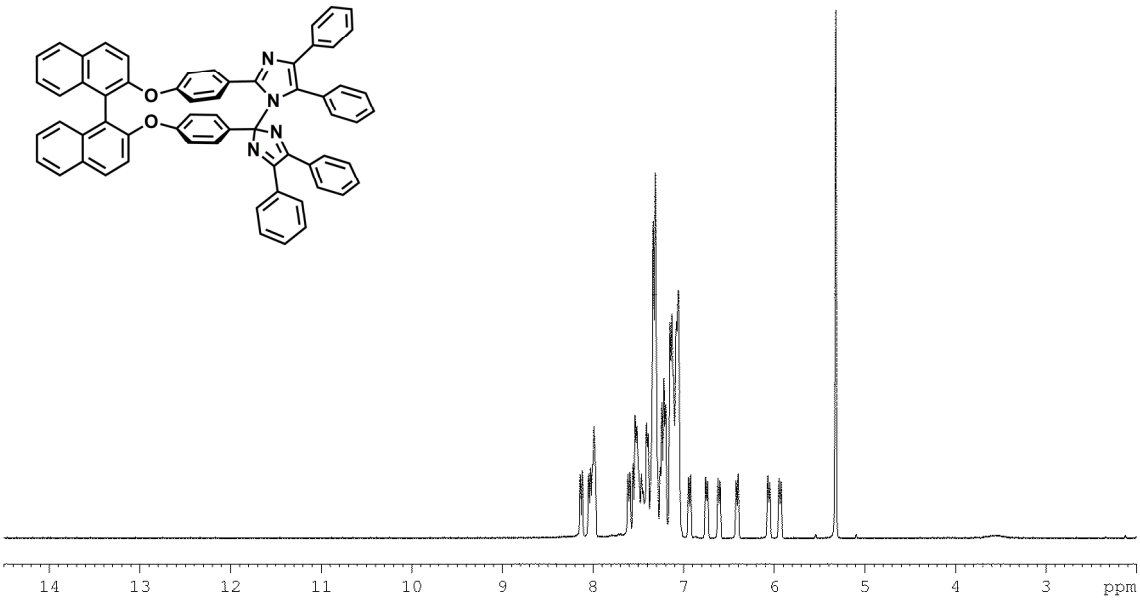


Figure S4. ^{1}H NMR spectrum of **1** in CD_2Cl_2.

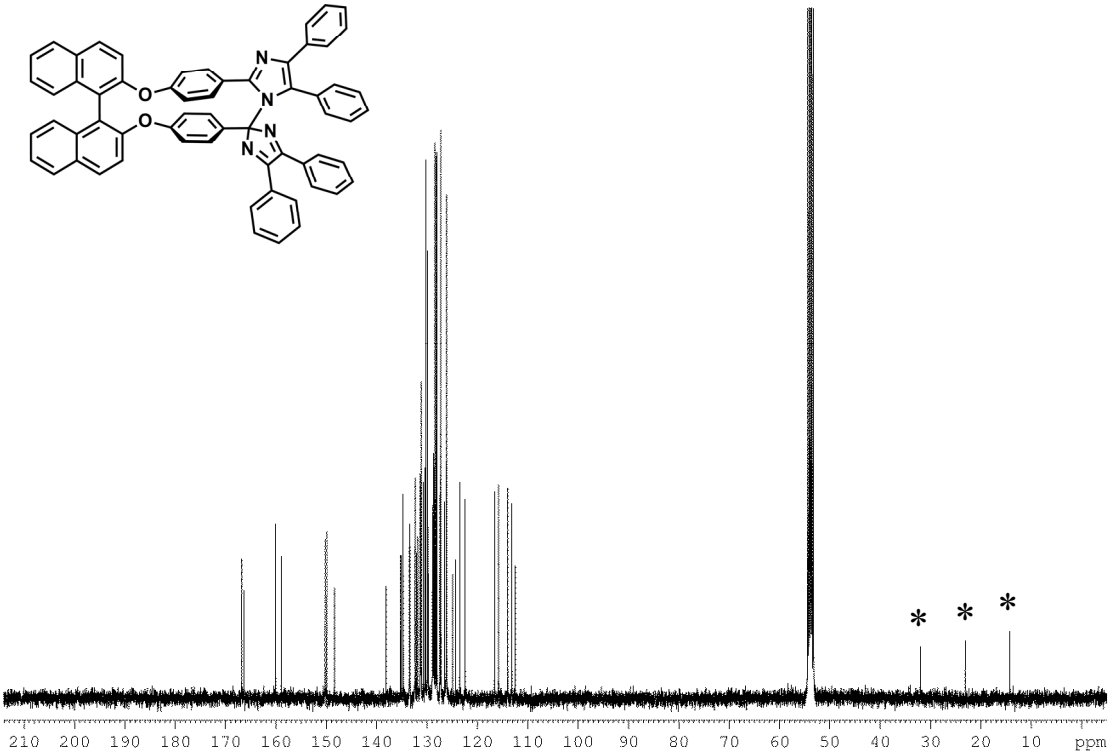
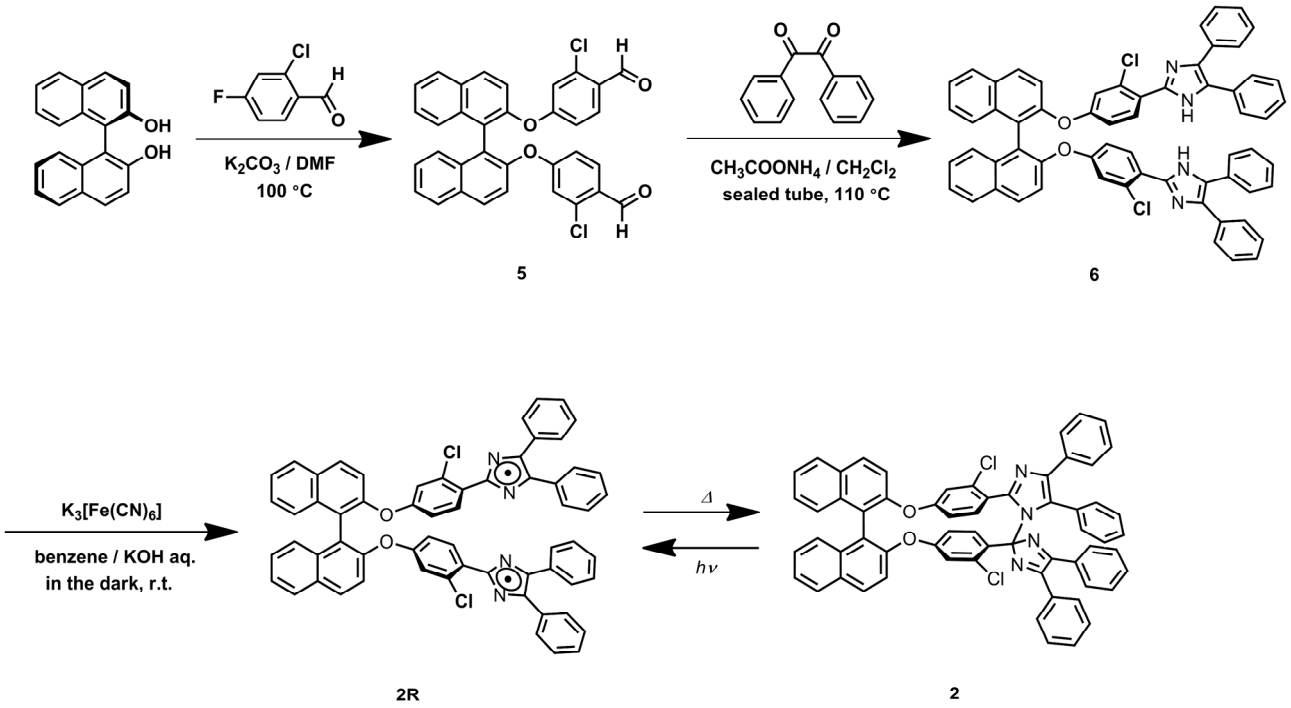


Figure S5. ^{13}C NMR spectrum of **1** in CD_2Cl_2 (*Solvent Peaks).

1.2 Synthesis of 2 (racemate)

Scheme S2. Synthetic procedure of **2**.



Compound 5

(*R*)-1,1'-Bi-2-naphthol (2.00 g, 6.99 mmol), 2-chloro-4-fluorobenzaldehyde (2.66 g, 2.85 mmol) and potassium carbonate (2.51 g, 5.85 mmol) were stirred at 100 °C in DMF (50 mL). After 15 h, the reaction mixture was cooled to room temperature and the target compound was extracted with CH_2Cl_2. The organic phase was washed with water. The organic phase was dried over anhydrous sodium sulfate, filtered, and evaporated to give a yellow solid. The yellow solid was purified by column chromatography over silica gel with CH_2Cl_2 as eluent to give a white amorphous solid (2.86 g, yield; 72 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.19 (d, J = 9.2 Hz, 2H), 8.09 (d, J = 8.6 Hz, 2H), 7.61 (d, J = 8.6 Hz, 2H), 7.54 (dd, J = 7.3, 7.9 Hz, 2H), 7.48 (d, J = 9.2 Hz, 2H), 7.41 (dd, J = 8.6, 9.2 Hz, 2H), 7.12 (d, J = 8.6 Hz, 2H), 6.83 (dd, J = 9.2, 2.4 Hz, 2H), 6.77 (d, J = 2.4 Hz, 2H); HR-MS (ESI+) calculated for $C_{34}H_{21}Cl_2O_4$ $[M+H]^+$ 563.0811, found 563.0819.

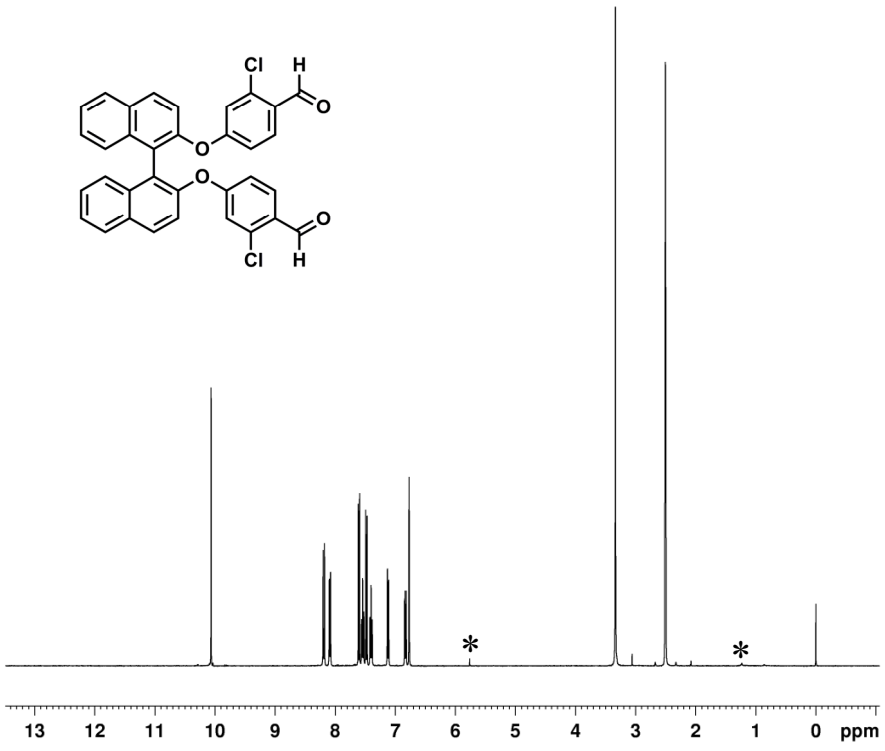


Figure S6. ^{1}H NMR spectrum of **5** in DMSO-d_6 (*Solvent Peaks).

Compound 6

Compound **5** (287 mg, 0.510 mmol), benzil (324 mg, 1.54 mmol) and ammonium acetate (599 mg, 7.77 mmol) were stirred at 110 °C in CH_2Cl_2 (1.7 mL) in a sealed tube. After 2 days, the reaction mixture was cooled to room temperature and washed with water, followed by ethanol. The precipitate was filtered and washed with hexane to give a white powder (374 mg, 0.267 mmol, yield; 77.8 %).^{1}H NMR (400 MHz, DMSO-d_6) δ 12.5 (s, 2H), 8.18 (d, J = 9.2 Hz, 2H), 8.08 (d, J = 7.9 Hz, 2H), 7.69 (d, J = 8.6 Hz, 2H), 7.54–7.19 (m, 28H), 7.05 (d, J = 2.4 Hz, 2H), 6.98 (dd, J = 9.2, 2.4 Hz, 2H); HR-MS (ESI+) calculated for $C_{62}H_{41}Cl_2N_4O_2$ $[M+H]^+$ 943.2601, found 943.2634.

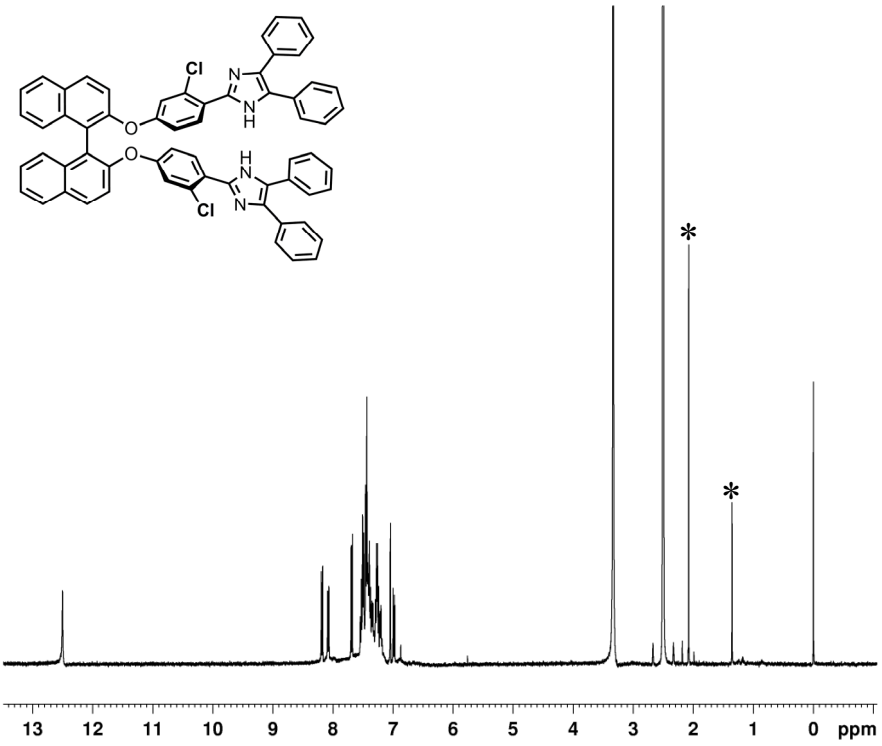


Figure S7. ^{1}H NMR spectrum of **6** in DMSO-d_6 (*Solvent Peaks).

Compound 2

All manipulations were carried out with the exclusion of light. Under nitrogen, a solution of potassium ferricyanide (5.91 g, 18.0 mmol) and potassium hydroxide (2.05 g, 36.6 mmol) in water (125 mL) was added to a solution of compound **6** (344 mg, 0.363 mmol) in benzene (125 mL). The reaction mixture was vigorously stirred at room temperature for 3 h. The organic layer was washed with water, dried, and evaporated. The residue was purified by column chromatography over silica gel with AcOEt/hexane = 1/3 as eluent to give a yellow solid (209 mg, yield; 61 %). The obtained compound was the racemate as was shown in Figure S27 because the refluxes at >100 °C racemize the (*R*)-1,1'-bi-2-naphthol moiety. ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.30 (d, J = 9.2 Hz, 1H), 8.18 (m, 3H), 7.63–6.85 (m, 32H), 6.09 (dd, J = 9.2, 2.4 Hz, 2H), 5.97 (dd, J = 9.2, 2.4 Hz, 2H); HR-MS (ESI+) calculated for $C_{62}H_{39}Cl_2N_4O_2$ $[M+H]^+$ 941.2445, found 941.2478.

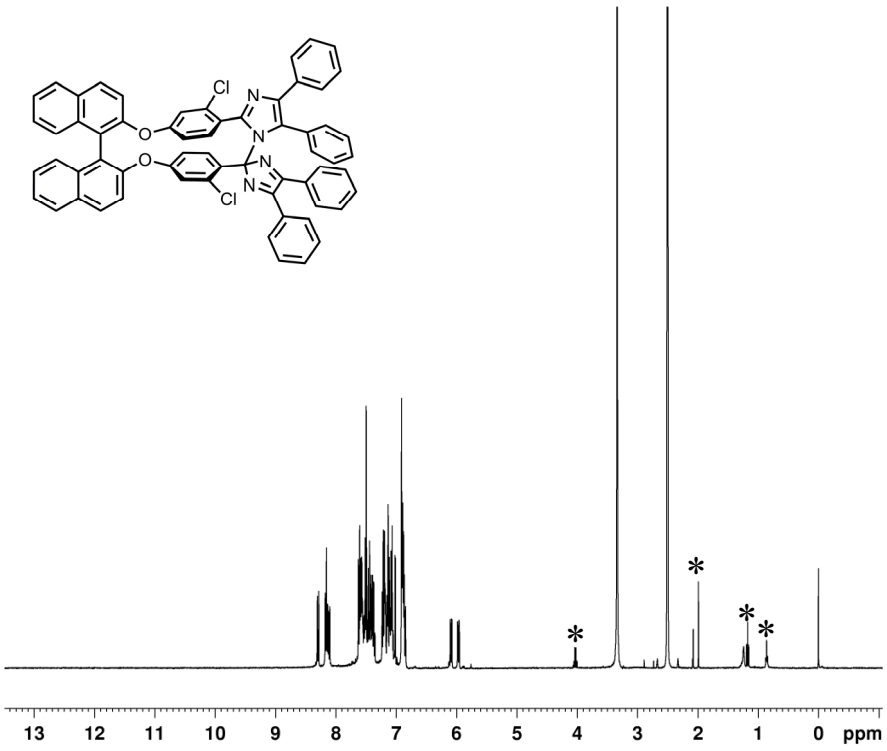
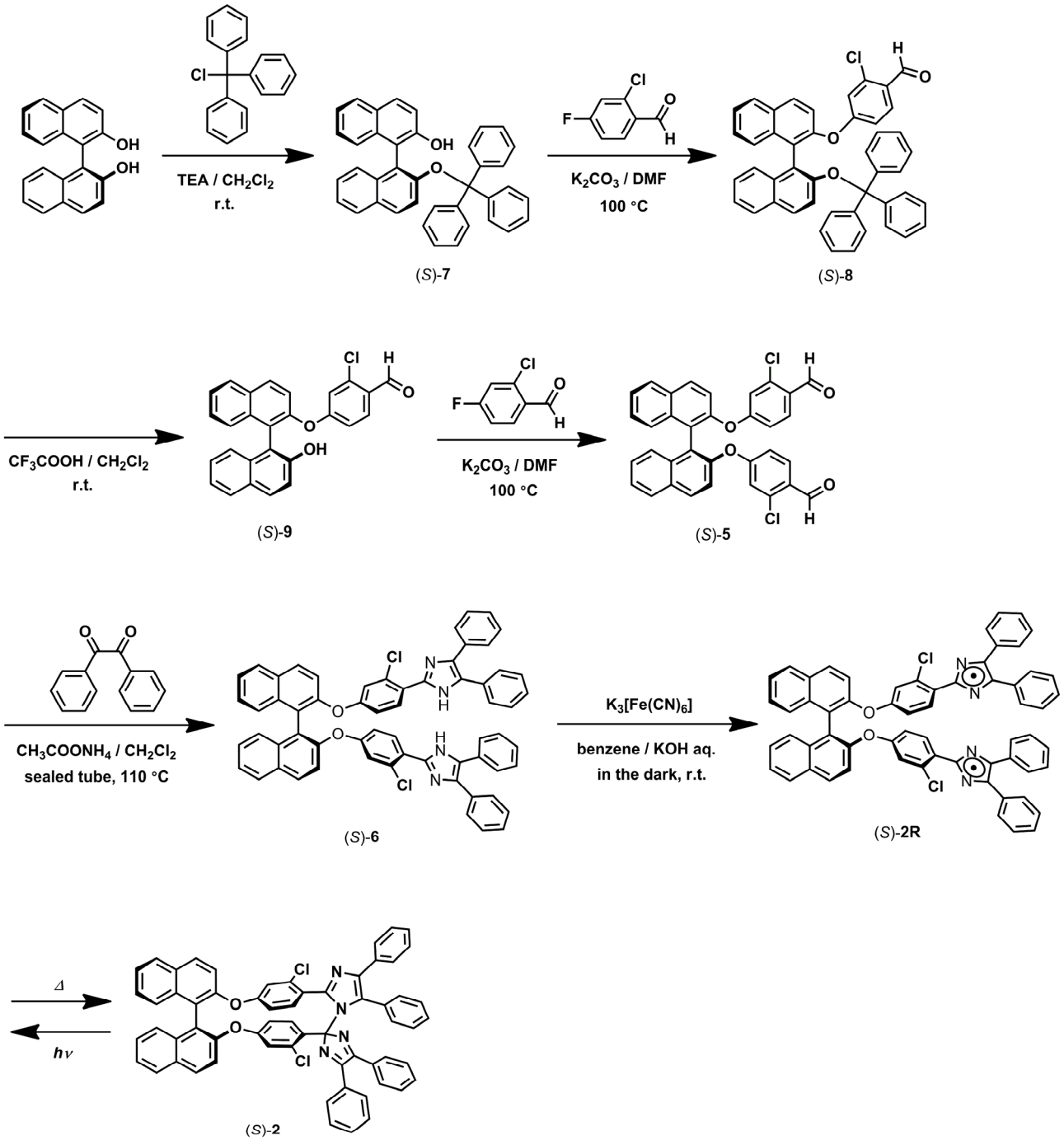


Figure S8. ^{1}H NMR spectrum of **2** in DMSO-d_6 (*Solvent Peaks).

1.3 Synthesis of (*S*)-2

Scheme S3. Synthetic procedure of (*S*)-**2** (the synthesis of (*R*)-**2** was the identical to that of (*S*)-**2** except that (*R*)-1,1'-bi-2-naphthol was used as the starting material) .



Compound (*S*)-7

Trityl chloride (3.89 g 13.9 mmol) in CH_2Cl_2 (45 mL) were added to (*S*)-1,1-bi-2-naphthol (4.00 g, 13.9 mmol), triethylamine (2.32 mL, 16.7 mmol) and CH_2Cl_2 (45 mL) at 0 °C. The mixture was allowed to warm to room temperature and was stirred for 2 h. The reaction mixture was washed with water. The organic phase was dried over anhydrous sodium sulfate, and the solution was filtered. The evaporation of the solvent afforded a yellow solid and the recrystallization from CH_2Cl_2/hexane gives a white crystal of compound (*S*)-**7** (5.08 g, yield; 68.7 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 9.56 (s, 1H), 7.94 (d, J = 8.9 Hz, 1H), 7.89 (dd, J = 6.3, 2.4 Hz, 1H), 7.74 (d, J = 8.0 Hz, 1H), 7.49 (d, J = 9.2 Hz, 1H), 7.43 (d, J = 8.9 Hz, 1H), 7.29–7.13 (m, 19H), 7.06 (d, J = 8.3 Hz, 1H), 6.86 (dd, J = 6.3, 2.4 Hz, 1H), 6.73 (d, J = 8.9 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 152.99, 151.16, 146.90, 143.99, 134.07, 133.82, 131.49, 129.64, 129.30, 129.23, 128.77, 128.39, 128.07, 127.99, 127.98, 127.73, 127.54, 127.31, 127.12, 126.97, 126.34, 125.32, 125.08, 124.40, 124.26, 124.09, 118.64, 117.82, 117.51, 115.89, 90.11; HR-MS (ESI−) calculated for $C_{39}H_{27}O_2$ $[M-H]^-$ 527.2011, found 527.2013.

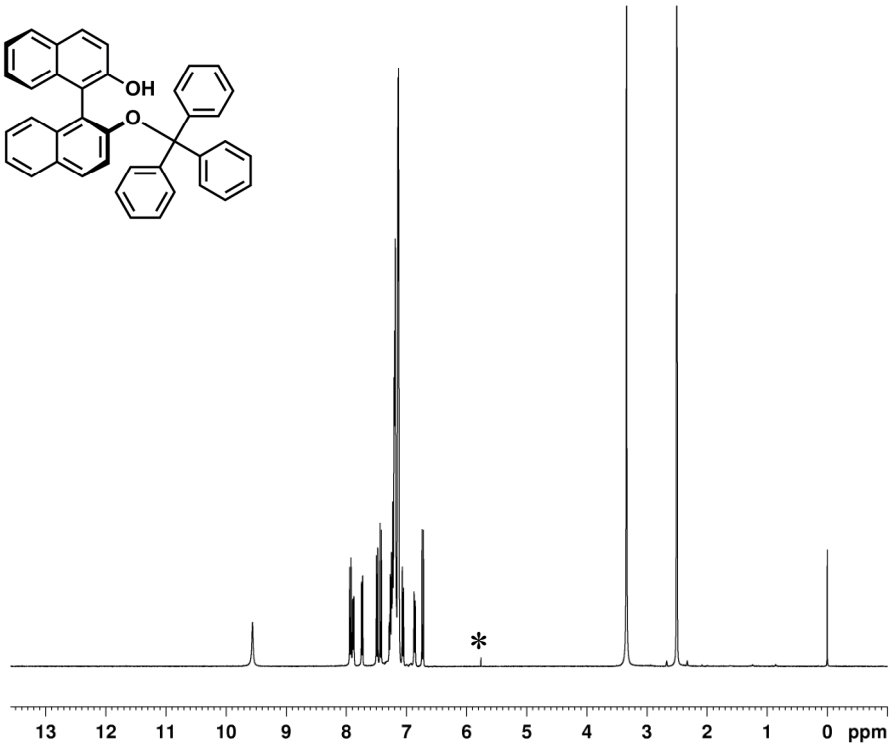


Figure S9. ^{1}H NMR spectrum of (*S*)-**7** in DMSO-*d*$_6$ (*Solvent Peak).

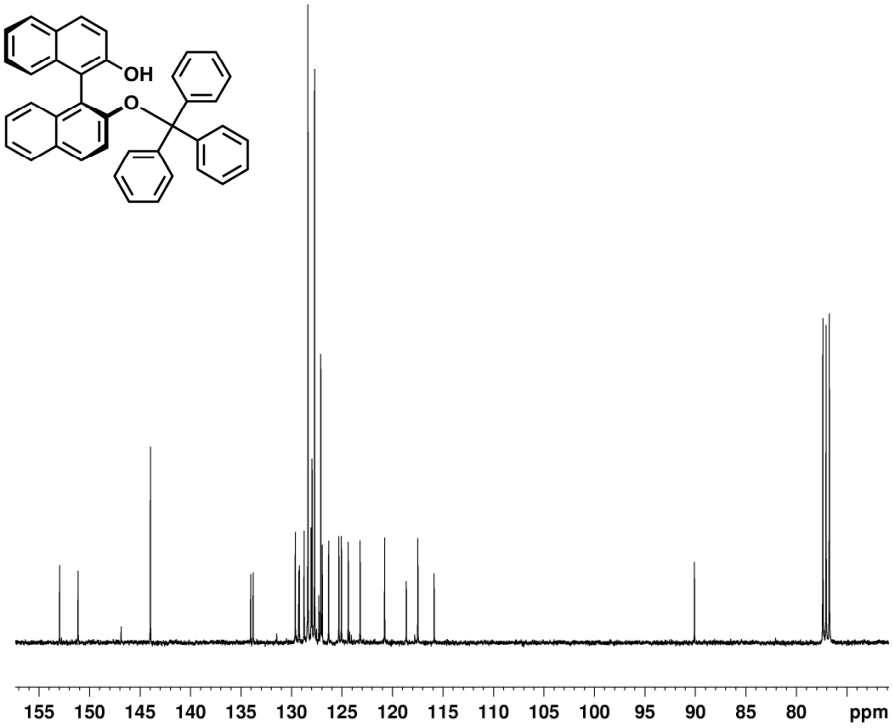


Figure S10. ^{13}C NMR spectrum of (*S*)-**7** in $CDCl_3$.

Compound (*S*)-8

Compound (*S*)-**7** (2.00 g, 3.78 mmol), 2-chloro-4-fluorobenzaldehyde (780 mg, 4.92 mmol) and potassium carbonate (1.36 g, 9.84 mmol) were stirred at 100 °C in DMF (50 mL). After 23 h, the reaction mixture was cooled to room temperature and the target compound was extracted with AcOEt. The organic phase was washed with water. The organic phase was dried over anhydrous sodium sulfate, filtered, and evaporated to give a yellow oil solid. The yellow oil solid was purified by column chromatography over silica gel with CH_2Cl_2/hexane as eluent to give a white amorphous solid (1.54 g, yield; 61.1 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.08 (s, 1H), 8.28 (d, J = 9.0 Hz, 1H), 8.15 (d, J = 8.1 Hz, 1H), 7.70 (dd, J = 7.0, 1.4 Hz, 1H), 7.65–7.42 (m, 5H), 7.34–7.08 (m, 23H), 6.94 (d, J = 2.3 Hz, 1H), 6.89 (dd, J = 8.6, 2.2 Hz, 1H), 6.71 (d, J = 9.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 188.47, 163.06, 151.93, 150.12, 144.07, 139.28, 134.29, 133.56, 131.58, 130.74, 129.79, 128.42, 128.10, 127.96, 127.88, 127.81, 126.80, 126.77, 126.51, 126.16, 126.05, 125.68, 124.93, 123.61, 120.29, 119.70, 119.32, 118.65, 116.11, 89.64, 53.46; HR-MS (ESI+) calculated for $C_{46}H_{31}ClNaO_3$ $[M+Na]^+$ 689.1859, found 689.1841.

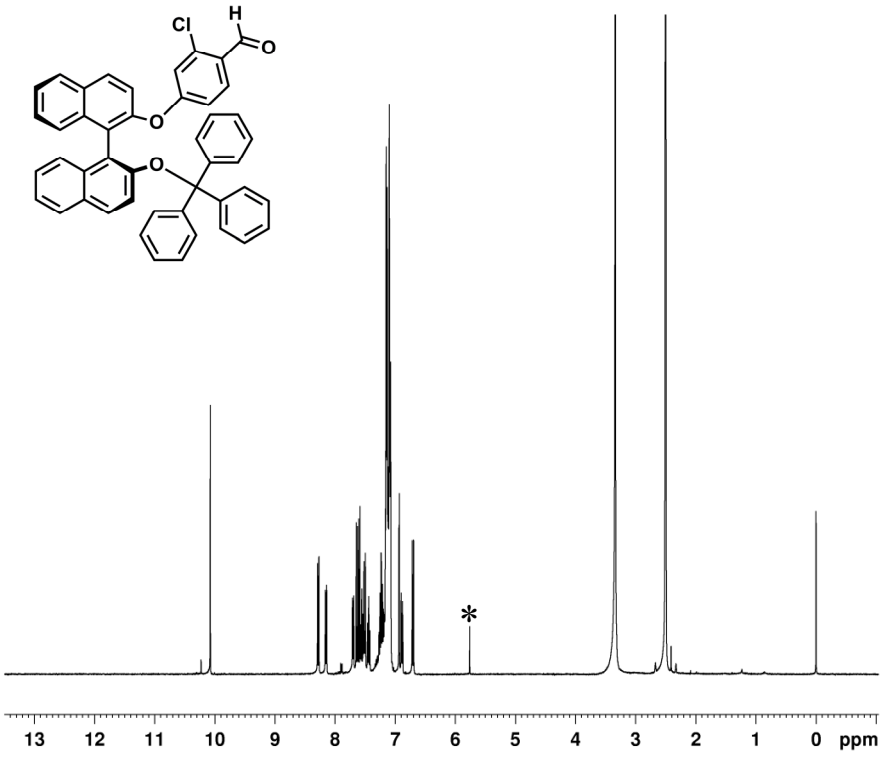


Figure S11. ^{1}H NMR spectrum of (*S*)-**8** in DMSO-d_6 (*Solvent Peak).

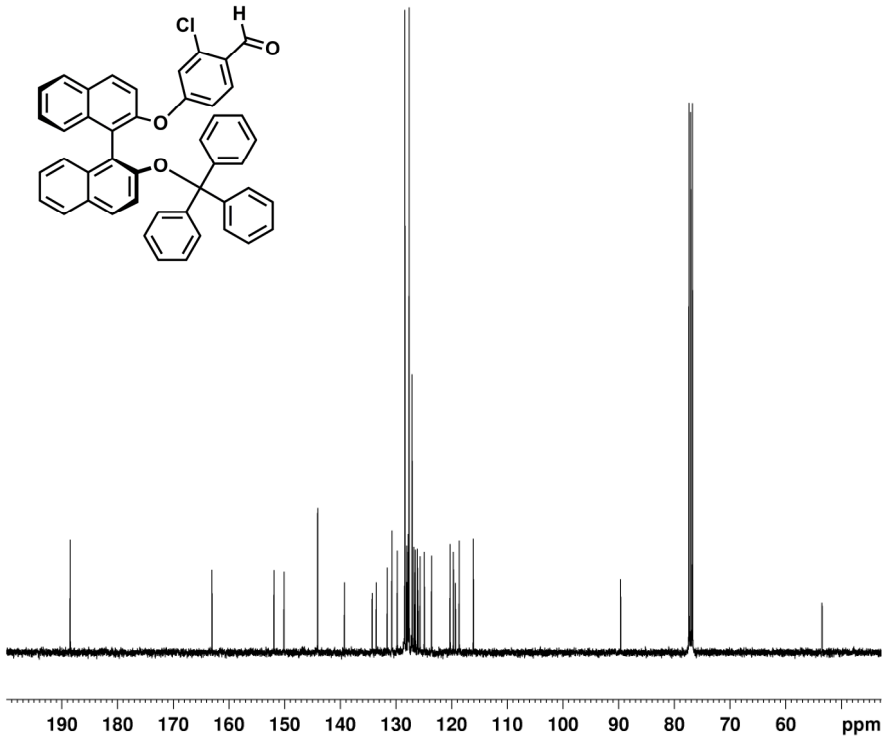


Figure S12. ^{13}C NMR spectrum of (*S*)-**8** in $CDCl_3$.

Compound (*S*)-9

TFA (9.70 mL) was added to compound (*S*)-**8** (1.08 g, 1.62 mmol) in CH_2Cl_2 (200 mL) at 0 °C. The mixture was allowed to warm to room temperature and was stirred for 2 h, quenched with $NaHCO_3$ aqueous. The organic phase was washed with water, dried over anhydrous sodium sulfate, filtered, and evaporated to give a brown oil solid. The brown oil solid was purified by column chromatography over silica gel with CH_2Cl_2/hexane as eluent to give a white amorphous solid (544 mg, yield; 79.2 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.13 (s, 1H), 9.61 (s, 1H), 8.17 (d, J = 8.9 Hz, 1H), 8.09 (d, J = 8.2 Hz, 1H), 7.84–7.82 (m, 2H), 7.69 (d, J = 8.7 Hz, 1H), 7.55–7.52 (m, 1H), 7.47 (d, J = 8.9 Hz, 1H), 7.41–7.37 (m, 1H), 7.32–7.19 (m, 3H), 7.15 (d, J = 8.4 Hz, 1H), 6.97–6.91 (m, 3H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 188.24, 162.70, 153.04, 150.01, 137.58, 133.60, 133.54, 131.24, 131.18, 130.21, 129.60, 128.27, 127.98, 127.73, 127.49, 126.88, 126.60, 125.55, 125.04, 123.87, 122.53, 120.67, 118.29, 118.11, 116.54, 112.98; HR-MS (ESI−) calculated for $C_{27}H_{16}ClO_3$ $[M–H]^-$ 423.0788, found 423.0790.

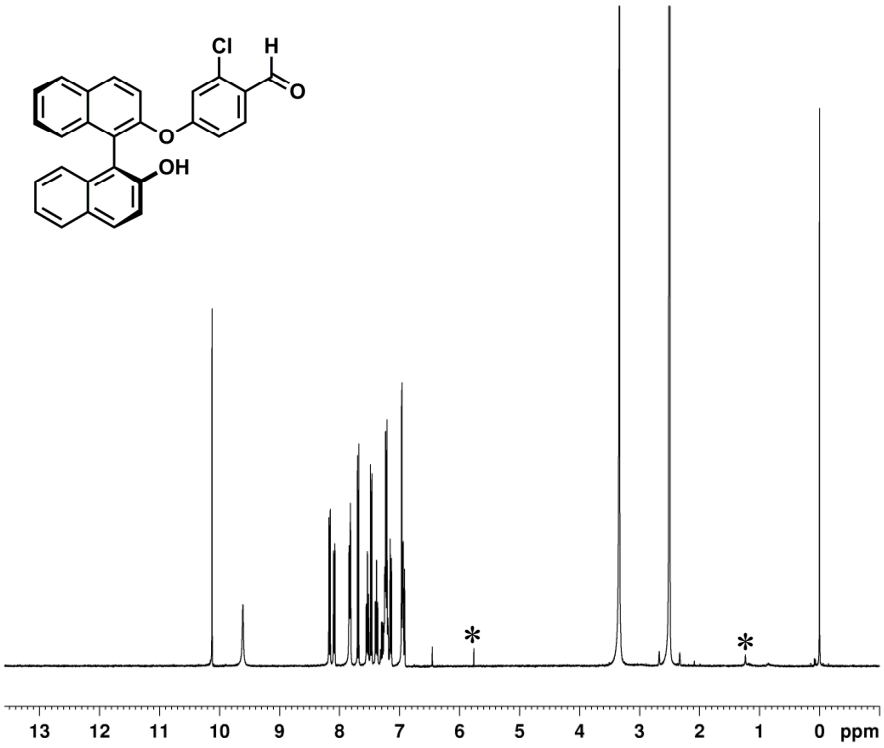


Figure S13. ^{1}H NMR spectrum of (*S*)-**9** in DMSO-*d*$_6$ (*Solvent Peaks).

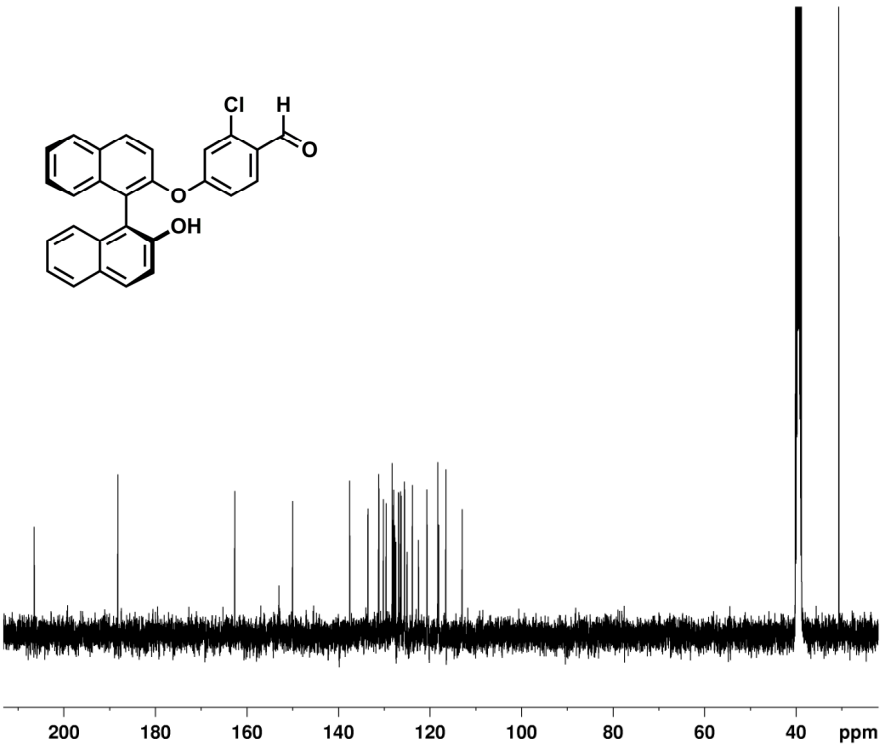


Figure S14. ^{13}C NMR spectrum of (*S*)-**9** in DMSO-d$_6$.

Compound (*S*)-5

Compound (*S*)-**9** (807 mg, 1.90 mmol), 2-chloro-4-fluorobenzaldehyde (456 mg, 2.85 mmol) and potassium carbonate (808 mg, 5.85 mmol) were stirred at 100 °C in DMF (50 mL). After 18 h, the reaction mixture was cooled to room temperature and the target compound was extracted with AcOEt. The organic phase was washed with water. The organic phase was dried over anhydrous sodium sulfate, filtered, and evaporated to give a yellow solid. The yellow solid was purified by column chromatography over silica gel with CH_2Cl_2/hexane as eluent to give a white amorphous solid (855 mg, yield; 79.9 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.19 (d, J = 9.2 Hz, 2H), 8.09 (d, J = 8.6 Hz, 2H), 7.61 (d, J = 8.6 Hz, 2H), 7.54 (dd, J = 7.3, 7.9 Hz, 2H), 7.48 (d, J = 9.2 Hz, 2H), 7.41 (dd, J = 8.6, 9.2 Hz, 2H), 7.12 (d, J = 8.6 Hz, 2H), 6.83 (dd, J = 9.2, 2.4 Hz, 2H), 6.77 (d, J = 2.4 Hz, 2H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 188.23, 162.70, 153.03, 150.01, 147.73, 137.58, 133.60, 133.54, 131.24, 131.18, 130.21, 129.59, 128.27, 127.97, 127.71, 127.48, 126.88, 126.60, 126.47, 126.32, 125.59, 125.55, 125.04, 123.87, 122.53, 120.67, 118.29, 118.10, 116.54, 112.98; HR-MS (ESI+) calculated for $C_{34}H_{21}Cl_2O_4$ $[M+H]^+$ 563.0811, found 563.0796.

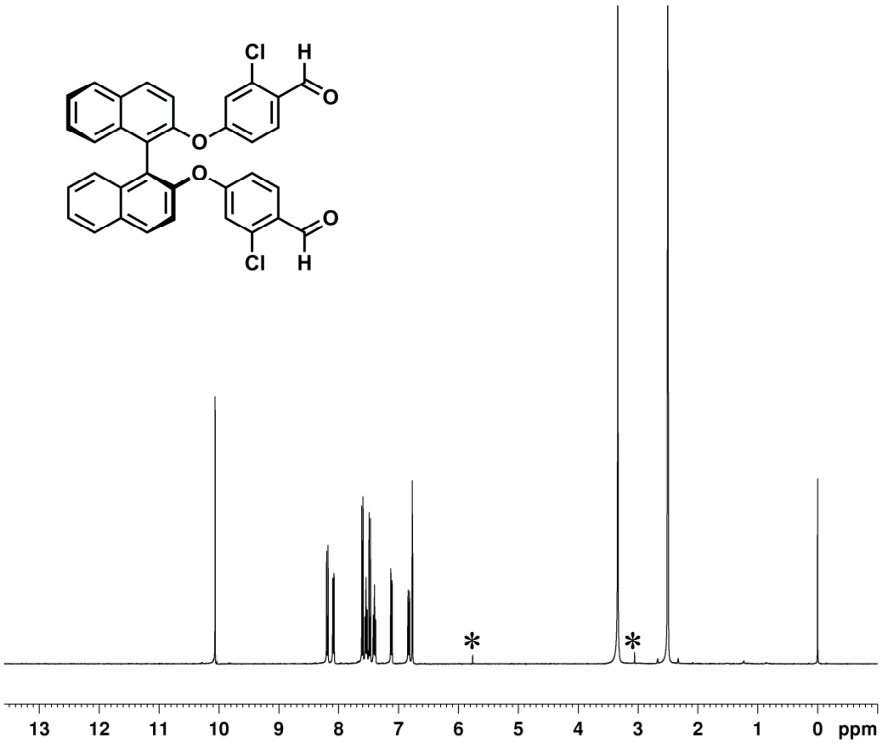


Figure S15. ^{1}H NMR spectrum of (*S*)-**5** in DMSO-*d*$_6$ (*Solvent Peaks).

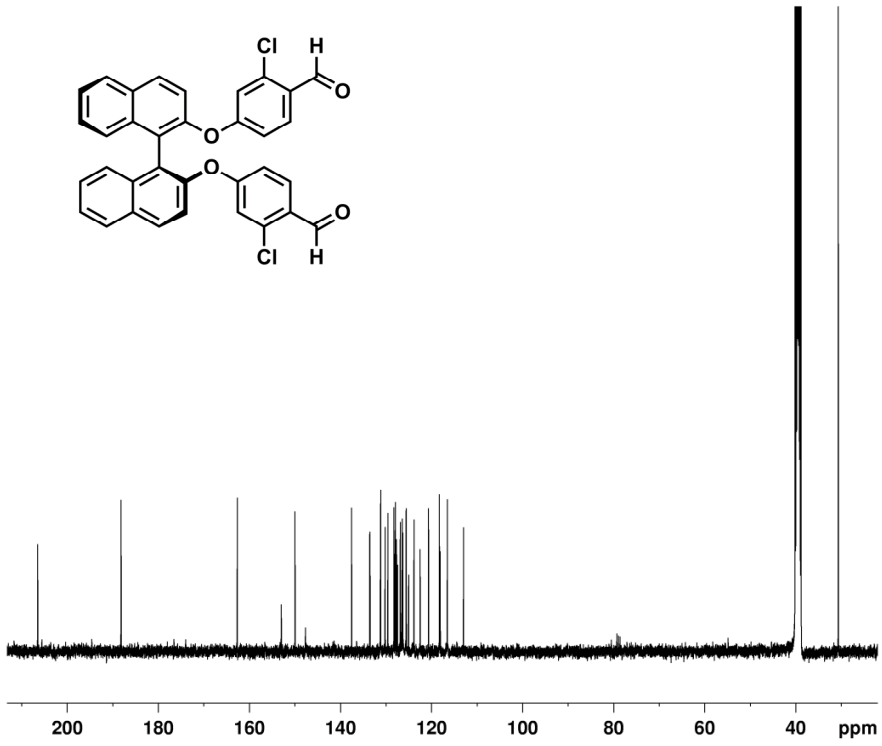


Figure S16. ^{13}C NMR spectrum of (*S*)-**5** in DMSO-*d*$_6$.

Compound (*S*)-6

Compound (*S*)-**5** (287 mg, 0.510 mmol), benzil (324 mg, 1.54 mmol) and ammonium acetate (599 mg, 7.77 mmol) were stirred at 110 °C in CH_2Cl_2 (1.7 mL) in a sealed tube. After 2 days, the reaction mixture was cooled to room temperature and washed with water, followed by ethanol. The precipitate was filtered and washed with hexane to give a white powder (374 mg, 0.267 mmol, yield; 77.8 %).^{1}H NMR (400 MHz, DMSO-d_6) δ 12.5 (s, 2H), 8.18 (d, J = 9.2 Hz, 2H), 8.08 (d, J = 7.9 Hz, 2H), 7.69 (d, J = 8.6 Hz, 2H), 7.54–7.19 (m, 28H), 7.05 (d, J = 2.4 Hz, 2H), 6.98 (dd, J = 9.2, 2.4 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.03, 150.80, 142.92, 137.56, 134.56, 134.12, 131.62, 130.96, 130.76, 130.37, 129.97, 128.90, 128.28, 128.24, 127.81, 127.72, 127.55, 127.21, 127.05, 126.95, 125.79, 125.42, 122.73, 122.28, 119.69, 118.83, 117.41; HR-MS (ESI+) calculated for $C_{62}H_{41}Cl_2N_4O_2$ $[M+H]^+$ 943.2601, found 943.2604.

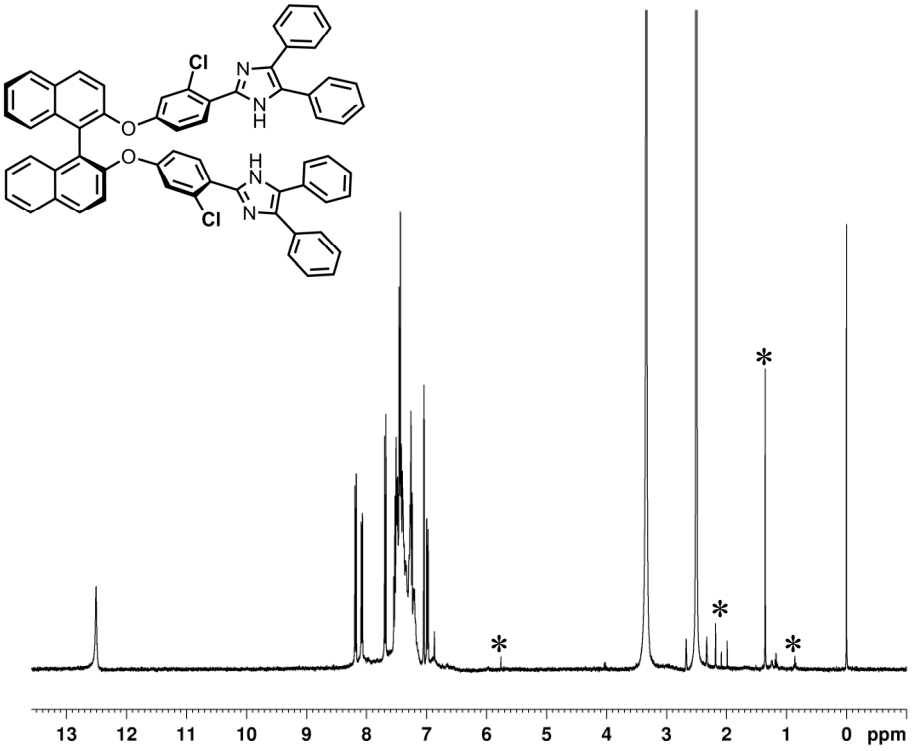


Figure S17. ^{1}H NMR spectrum of (*S*)-**6** in DMSO-*d*$_6$ (*Solvent Peaks).

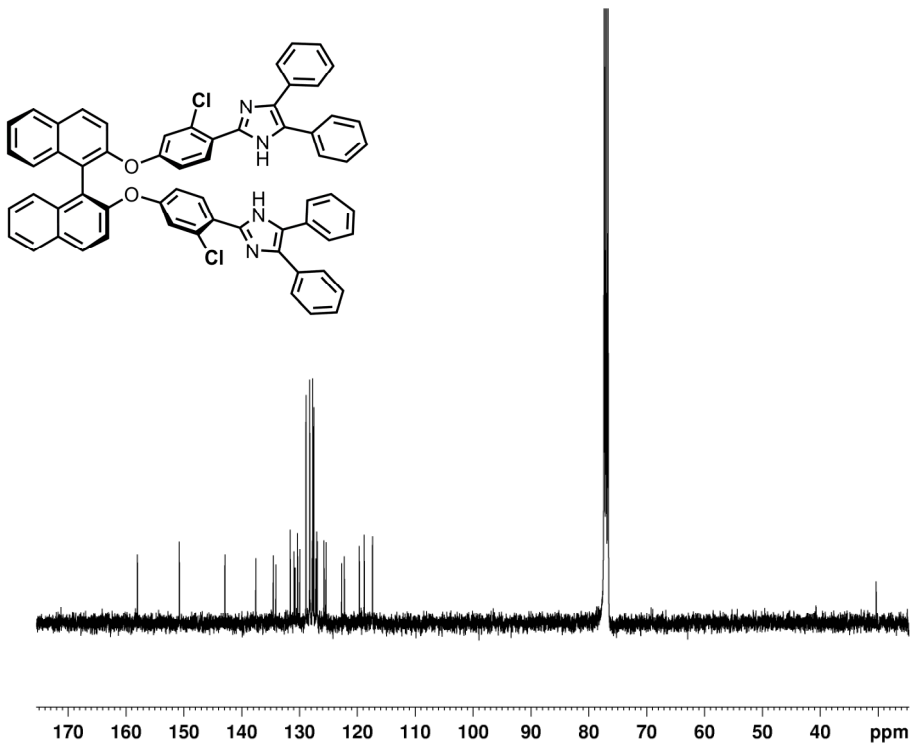


Figure S18. ^{13}C NMR spectrum of (*S*)-**6** in $CDCl_3$.

Compound (*S*)-2

All manipulations were carried out with the exclusion of light. Under nitrogen, a solution of potassium ferricyanide (3.78 g, 11.5 mmol) and potassium hydroxide (1.36 g, 24.2 mmol) in water (60 mL) was added to a solution of compound (*S*)-**6** (202 mg, 0.214 mmol) in benzene (60 mL). The reaction mixture was vigorously stirred at room temperature for 3 h. The organic layer was washed with water, dried, and evaporated. The residue was purified by column chromatography over silica gel with AcOEt/hexane = 1/3 as eluent to give a yellow solid (61.0 mg, yield; 30 %). ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.30 (d, J = 9.2 Hz, 1H), 8.18 (m, 3H), 7.63–6.85 (m, 32H), 6.09 (dd, J = 9.2, 2.4 Hz, 2H), 5.97 (dd, J = 9.2, 2.4 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.35, 165.01, 160.36, 160.15, 149.42, 149.42, 149.05, 144.95, 137.44, 136.34, 135.61, 134.87, 134.80, 134.28, 133.20, 133.05, 132.96, 131.78, 131.62, 131.51, 131.38, 131.03, 130.84, 130.64, 130.51, 130.27, 130.06, 129.94, 129.35, 128.97, 128.48, 128.31, 128.20, 128.16, 128.07, 127.71, 127.31, 127.20, 127.10, 126.88, 126.28, 126.03, 125.99, 125.85, 125.81, 123.82, 122.48, 121.84, 120.21, 116.90, 112.52, 112.19, 111.08; HR-MS (ESI+) calculated for $C_{62}H_{39}Cl_2N_4O_2$ $[M+H]^+$ 941.2450, found 941.2445.

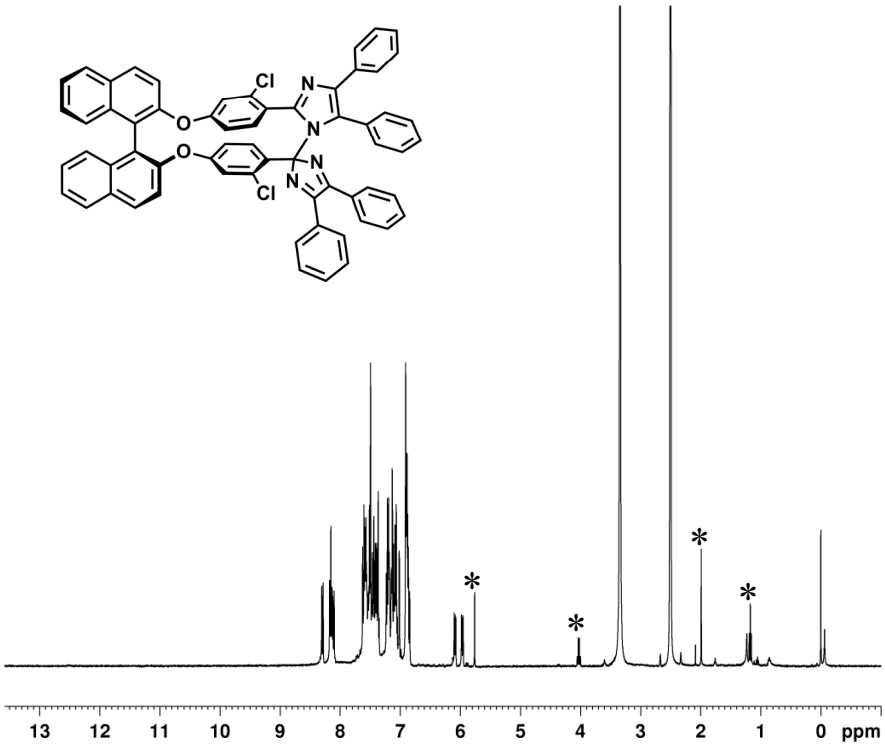


Figure S19. ^{1}H NMR spectrum of (*S*)-**2** in DMSO-d_6 (*Solvent Peaks).

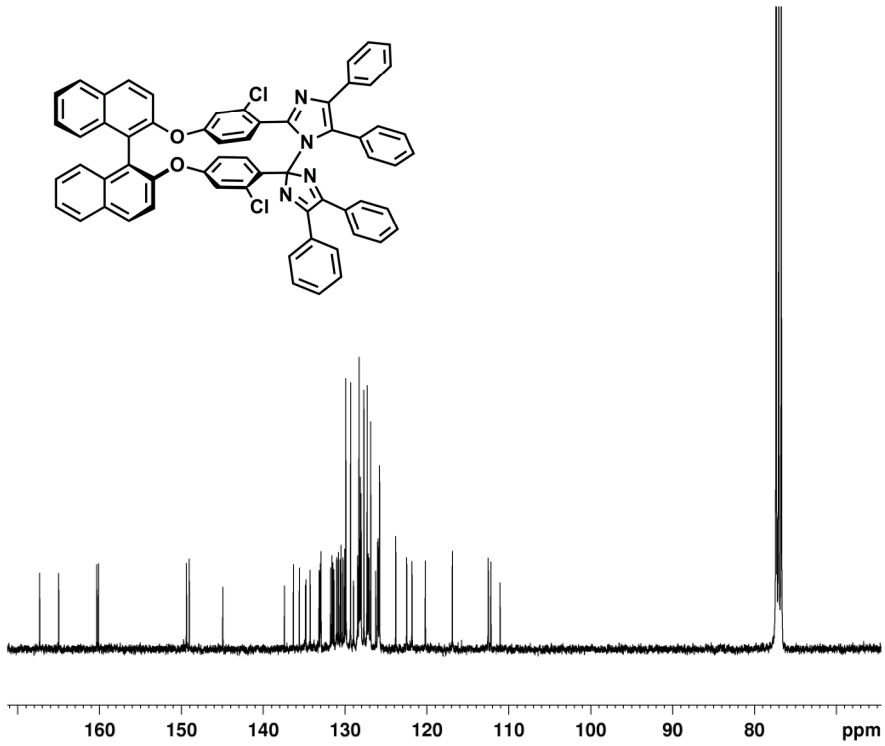


Figure S20. ^{13}C NMR spectrum of (*S*)-**2** in $CDCl_3$.

2. HR-ESI-TOF-MS-spectra

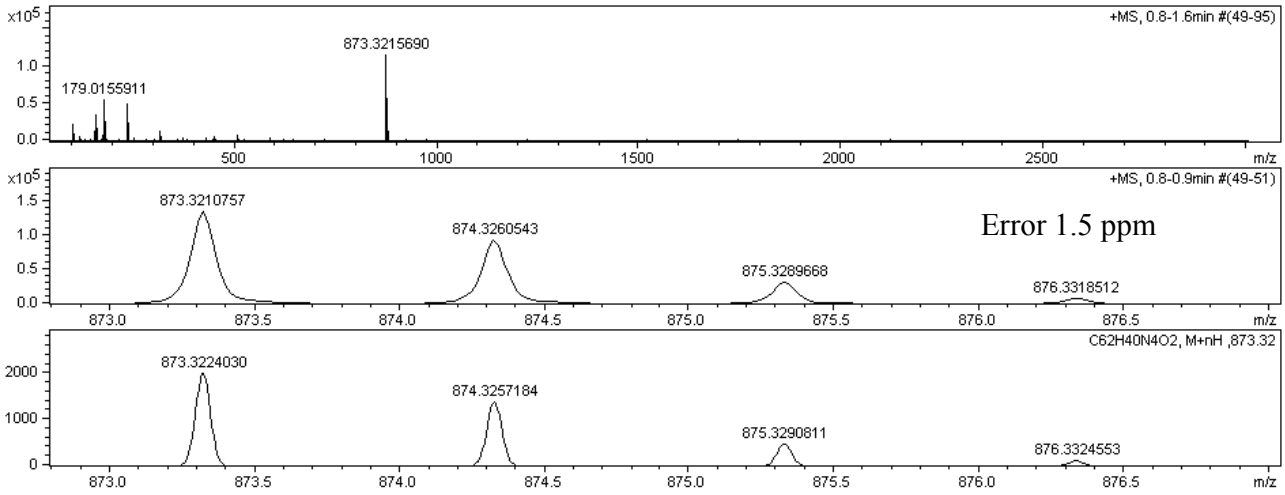


Figure S21. HR-ESI-TOF-MS of **1**.

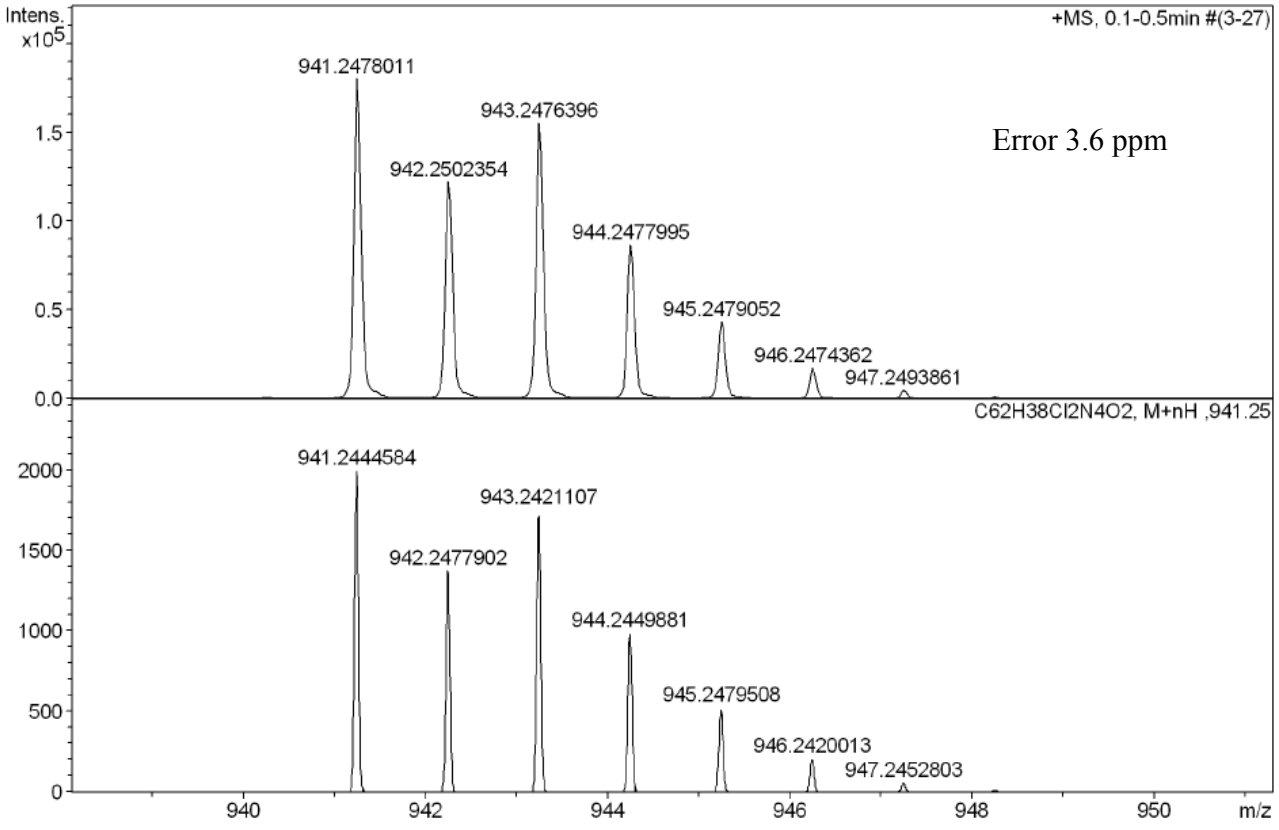


Figure S22. HR-ESI-TOF-MS of **2**.

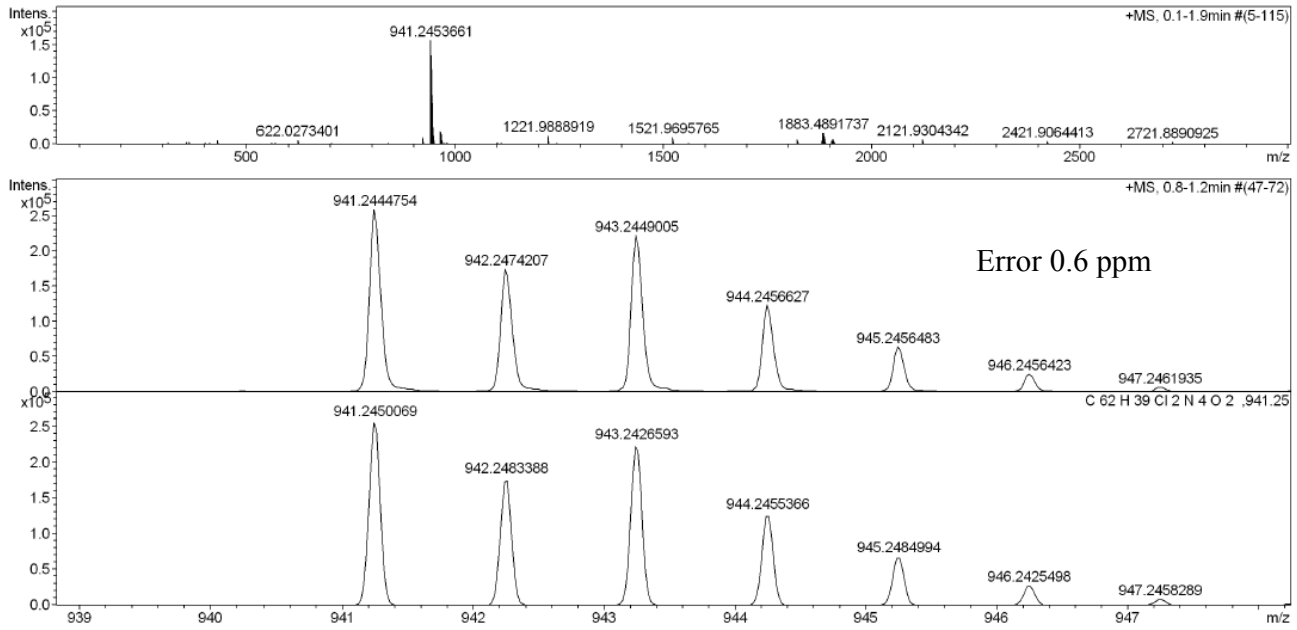


Figure S23. HR-ESI-TOF-MS of (*S*)-**2**.

3. HPLC analyses for determination of purity

HPLC analyses using THF/CH_3CN = 1/15 as an eluent was performed with system composed of a Mightysil RP18 (4.6 mm × 25 cm) column and JASCO PU-2080 Plus pump equipped with a UV-2075 Plus UV/VIS detector. The mobile phase was THF/CH_3CN = 1/15 with a flow rate of 1.0 mL/min, inject volume; 2.0 μL.

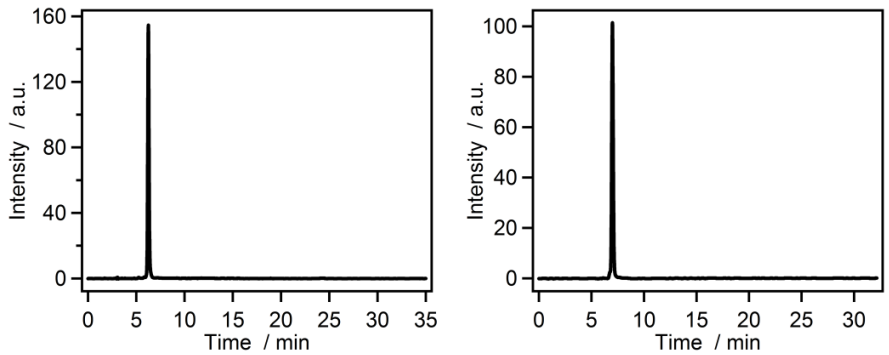


Figure S24. HPLC chromatogram of the racemate of **1** (left) and **2** (right), detection wavelength; 254 nm, purity 99%, respectively.

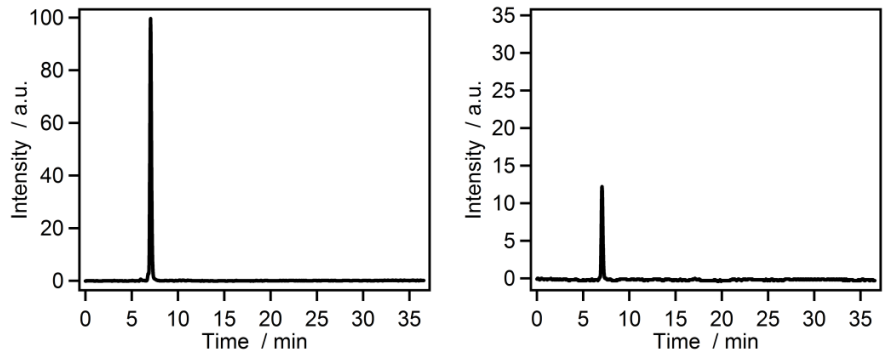


Figure S25. HPLC chromatograms of (*R*)-**2**, detection wavelength; 254 (left) and 340 nm (right), purity 99%.

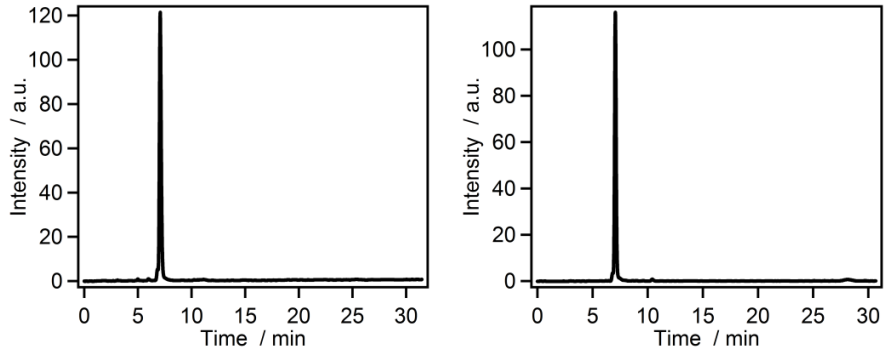


Figure S26. HPLC chromatograms of (*S*)-**2**, detection wavelength; 254 (left) and 340 nm (right), purity 99%.

4. Chiral HPLC analyses for determination of enantiomeric excesses (ee)

HPLC analyses using CH_2Cl_2/hexane = 1/1 as an eluent was performed with a system composed of a DAICEL CHIRALPAK IC column (4.6 mm × 25 cm) and JASCO PU-2080 Plus pump equipped with a UV-2075 Plus UV/VIS detector.

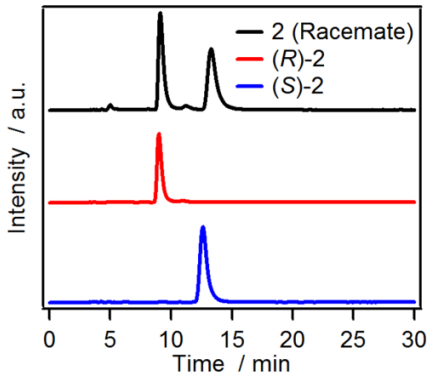


Figure S27. HPLC charts of **2**, (*R*)-**2** and (*S*)-**2**.

5. X-ray crystallographic analysis

The diffraction data of the single crystal of **1** are collected on the Bruker APEX II CCD area detector (MoK_α, λ = 0.71073 nm). During the data collection, the lead glass doors of the diffractometer were covered to exclude the room light. The data refinement was carried out by the Bruker APEX II software package with SHELXT program.[S1)] All non-hydrogen atoms were anisotropically refined.

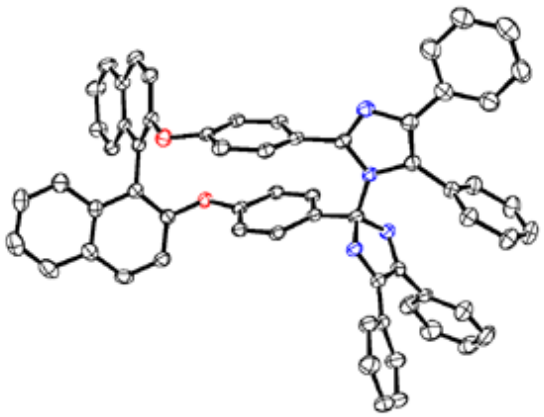


Figure S28. ORTEP representation of the molecular structure of **1** with thermal ellipsoid at 50% probability. The hydrogen atoms and solvent molecules are omitted. Oxygen and nitrogen atoms are highlighted in red and blue, respectively.

Table S1. X-ray crystallographic data of **1**.

Identification code	1
Empirical formula	C62 H40 N4 O2
Formula weight	872.98
Temperature	90 K
Wavelength	0.71073 Å
Crystal system	Triclinic
Space group	P-1
Unit cell dimensions	a = 10.8574(15) Å α= 90.116(2)°.
	b = 14.598(2) Å β= 105.115(2)°.
	c = 15.654(2) Å γ = 110.614(2)°.
Volume	2229.8(5) Å3
Z	2
Density (calculated)	1.300 Mg/m3
Absorption coefficient	0.079 mm-1
F(000)	912
Crystal size	0.40 x 0.35 x 0.01 mm3
Theta range for data collection	1.50 to 26.40°.
Index ranges	-13<=h<=11, -18<=k<=12, -16<=l<=19
Reflections collected	23502
Independent reflections	8830 [R(int) = 0.0137]
Completeness to theta = 26.40°	96.4 %
Absorption correction	Empirical
Refinement method	Full-matrix least-squares on F2
Data / restraints / parameters	8830 / 0 / 613
Goodness-of-fit on F2	Goodness-of-fit on F21.023
Final R indices [I>2sigma(I)]	R1 = 0.0565, wR2 = 0.1105
R indices (all data)	R indices (all data) R1 = 0.0791, wR2 = 0.1263
Largest diff. peak and hole	0.519 and -0.267 e.Å-3

6. Laser flash photolysis

The laser flash photolysis experiments were performed with a Unisoku TSP-1000 time-resolved spectrophotometer. A 10 Hz Q-switch Nd:YAG laser (Continuum Minilite II) with the third harmonic at 355 nm (ca. 4 mJ per 5 ns pulse) was employed for the excitation light. A halogen lamp (OSRAM HLX64623) was used for a probe light and the transmitted light through the sample was guided into the monochrometer and a detector (Unisoku MD200 and Hamamatsu R2949 photomultiplier tube, respectively) with an optical fiber scope.

7. Eyring plots

To obtain the activation parameters of the thermal back reactions of **1R** and **2R**, we conducted the temperature dependence of the decay profiles of the thermal back reactions of **1R** and **2R**. The rate constants for the thermal back reaction for **1R** and **2R** are tabulated in **Table S2** and **S3**, respectively. These temperature dependences were analyzed by Eyring plots and these plots are shown in **Figure S28** for **1R** and **Figure S29** for **2R**, respectively.

Table S2. First-order rate constants for the thermal back-reaction of **1** in degassed benzene (2.1×10^{-5} M) at different temperature.

T / ℃	k / s^{-1}
5	4.8
10	6.8
15	9.5
20	13.2
25	18.2
30	24.7
35	33.2
40	44.3

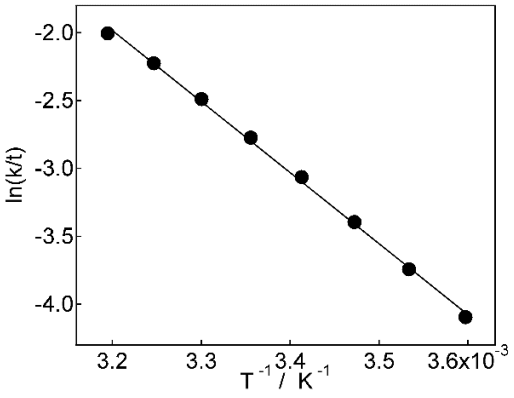


Figure S29. The Eyring plot for the thermal back-reaction of colored species of **1** in degassed benzene (2.1×10^{-5} M).

Table S3. First-order rate constants for the thermal back-reaction of **2** in degassed benzene (2.3×10^{-5} M) at different temperature.

T / °C	k / s^{-1}
5	3.49
10	4.19
15	4.98
20	5.86
25	6.68
30	7.84
35	9.11
40	10.2

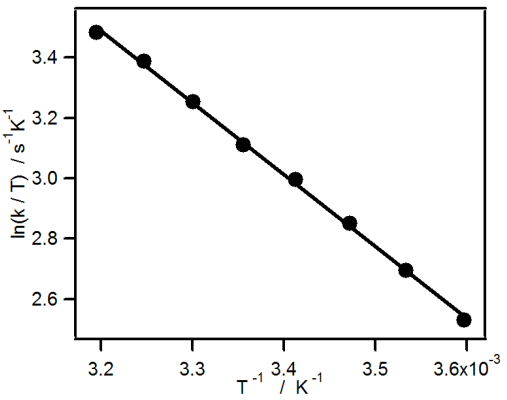


Figure S30. The Eyring plot for the thermal back-reaction of colored species of **2** in degassed benzene (2.3×10^{-5} M).

8. CD spectroscopy

Circular dichroism (CD) spectra were recorded on a JASCO J-820 spectropolarimeter. The CD spectra of (*R*)-**2** and (*S*)-**2** in acetonitrile (3.3×10^{-5} M) were measured with a 10-mm quartz cell at room temperature. Photoracemization processes of (*R*)-**2** and (*S*)-**2** were analyzed using a Q-switch Nd:YAG laser (Continuum Minilite II) with the third harmonic at 355 nm (ca. 4 mJ per 5 ns pulse) as the excitation beam.

9. UV-vis absorption spectroscopy and investigation for fatigue resistance

UV-vis absorption spectra were recorded on a Shimadzu UV-3150 spectrometer. The UV-vis absorption spectra of **1** (2.1×10^{-5} M) and **2** (2.3×10^{-5} M) in benzene were measured with a 10-mm quartz cell at 25 °C. The fatigue resistances of **1** and **2** were measured using a Q-switch Nd:YAG laser (Continuum Minilite II) with the third harmonic at 355 nm (ca. 4 mJ per 5 ns pulse) as the excitation beam.

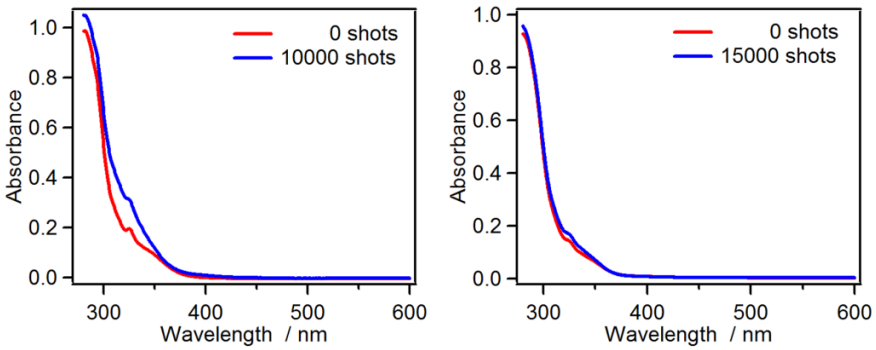


Figure S31. UV-vis absorption spectra of **1** (2.1×10^{-5} M) (left) and **2** (2.3×10^{-5} M) (right) in benzene before and after laser shots at 25 °C.

10. DFT calculations

All calculations were carried out using the Gaussian 09 program (Revision D.01)[S2]. The molecular structures were fully optimized at the M062X/6-31G(d) level of the theory for **2** and the UM062X/6-31G(d) level of the theory for **2R.** The analytical second derivatives were computed using the vibrational analysis to confirm each stationary point to be a minimum. The TDDFT calculations were performed at the MPW1PW91/6-31+G(d) level of the theory for **2** and UMPW1PW91/6-31+G(d) level of the theory for **2R** for the optimized structures.

Table S4. Standard orientation of the optimized geometry for **2**.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	17	0	0.628470	0.813532	3.508147
2	17	0	0.881086	0.940766	-2.989685
3	8	0	-3.989950	0.901268	1.510275
4	8	0	-3.666619	-0.634175	-1.651224
5	7	0	2.213950	1.079457	-0.008867
6	7	0	2.089257	-1.074957	0.976874
7	7	0	2.684124	-0.891175	-1.304792
8	7	0	2.184394	3.030688	1.068875
9	6	0	-5.525890	-0.815527	-0.171582
10	6	0	-4.544712	-1.415149	-0.923920
11	6	0	-6.458611	-1.655593	0.524947
12	6	0	-6.345858	-3.070627	0.425199

13	6	0	-7.270476	-3.894527	1.115345
14	1	0	-7.169807	-4.973301	1.027351
15	6	0	-5.313092	-3.630124	-0.373911
16	1	0	-5.239201	-4.711031	-0.457411
17	6	0	-7.503153	-1.115397	1.321327
18	1	0	-7.596744	-0.037684	1.408352
19	6	0	-4.432608	-2.823519	-1.035804
20	1	0	-3.641311	-3.231147	-1.657501
21	6	0	-8.269716	-3.344184	1.875847
22	1	0	-8.973391	-3.982536	2.400777
23	6	0	-8.382989	-1.938854	1.978298
24	1	0	-9.173671	-1.507214	2.584159
25	6	0	-5.699955	0.666242	-0.131175
26	6	0	-6.953086	2.677590	-0.834884
27	6	0	-4.954503	1.467457	0.700646
28	6	0	-6.156917	3.451262	0.051992
29	1	0	-6.340310	4.519586	0.127730
30	6	0	-5.181531	2.863073	0.804090
31	1	0	-4.565797	3.438631	1.488322
32	6	0	-6.729795	1.275238	-0.924613
33	6	0	-7.967981	3.274632	-1.624269
34	1	0	-8.125026	4.347049	-1.541920
35	6	0	-8.733880	2.518104	-2.473344
36	1	0	-9.508507	2.983955	-3.074279
37	6	0	-7.539590	0.518114	-1.812874
38	1	0	-7.375977	-0.551659	-1.894431
39	6	0	-8.512981	1.125109	-2.566508
40	1	0	-9.119947	0.529410	-3.241253
41	6	0	-1.817698	1.029226	2.401804
42	1	0	-2.189415	0.547039	3.298585
43	6	0	-2.699193	1.349173	1.372425
44	6	0	-0.508519	-1.167728	0.138862
45	1	0	-0.144769	-1.545581	1.087627
46	6	0	-0.459600	1.274831	2.229684

47	6	0	-1.856884	-1.287186	-0.165052
48	1	0	-2.534681	-1.744669	0.546621
49	6	0	0.386997	-0.498970	-0.695367
50	6	0	-2.248449	2.011371	0.231813
51	1	0	-2.934845	2.270607	-0.566435
52	6	0	-0.889894	2.247652	0.092214
53	1	0	-0.517436	2.710406	-0.817650
54	6	0	-0.114286	0.008910	-1.901642
55	6	0	-2.334998	-0.721108	-1.343082
56	6	0	-1.456552	-0.110878	-2.235989
57	1	0	-1.841364	0.327361	-3.149733
58	6	0	3.441854	2.826584	0.551908
59	6	0	1.480393	1.981393	0.746143
60	6	0	2.952513	-1.973247	0.687741
61	6	0	3.500368	1.614788	-0.101181
62	6	0	3.337129	-1.851895	-0.771031
63	6	0	1.839939	-0.326238	-0.258998
64	6	0	4.177719	-4.806693	3.626098
65	1	0	4.480022	-5.525642	4.381570
66	6	0	3.414584	-2.957794	1.684659
67	6	0	3.528594	-2.564131	3.021921
68	1	0	3.305143	-1.534314	3.284056
69	6	0	3.917374	-3.485379	3.986628
70	1	0	4.013861	-3.173995	5.021929
71	6	0	4.047366	-5.206467	2.298504
72	1	0	4.239159	-6.237393	2.018221
73	6	0	3.671355	-4.285081	1.326720
74	1	0	3.568973	-4.596898	0.291386
75	6	0	4.373751	-2.575739	-1.528271
76	6	0	4.239783	-2.685870	-2.917295
77	1	0	3.338363	-2.302626	-3.385716
78	6	0	5.253408	-3.260349	-3.672473
79	1	0	5.141779	-3.349201	-4.748690
80	6	0	6.559434	-3.592717	-1.670007

81	1	0	7.470264	-3.929593	-1.184722
82	6	0	6.415789	-3.713786	-3.049201
83	1	0	7.210655	-4.158864	-3.640323
84	6	0	5.541259	-3.028601	-0.907700
85	1	0	5.670858	-2.905101	0.162696
86	6	0	4.694578	0.979481	-0.705971
87	6	0	4.832359	0.818293	-2.089119
88	1	0	4.007256	1.096618	-2.737328
89	6	0	5.750131	0.607097	0.130997
90	1	0	5.646398	0.748293	1.203401
91	6	0	6.000287	0.282592	-2.620345
92	1	0	6.091244	0.148421	-3.694112
93	6	0	7.051904	-0.081868	-1.780080
94	1	0	7.962449	-0.500150	-2.198423
95	6	0	6.925456	0.083580	-0.404411
96	1	0	7.740313	-0.197437	0.256352
97	6	0	4.489511	3.848844	0.730327
98	6	0	5.572324	3.979237	-0.148001
99	1	0	5.658266	3.320723	-1.006037
100	6	0	4.381960	4.741819	1.803476
101	1	0	3.529175	4.652397	2.468866
102	6	0	6.532891	4.963979	0.057998
103	1	0	7.364015	5.052586	-0.635449
104	6	0	6.425894	5.838868	1.135572
105	1	0	7.176618	6.607604	1.292459
106	6	0	5.342929	5.726189	2.004604
107	1	0	5.245184	6.409538	2.843164
108	6	0	0.036934	1.837483	1.051461

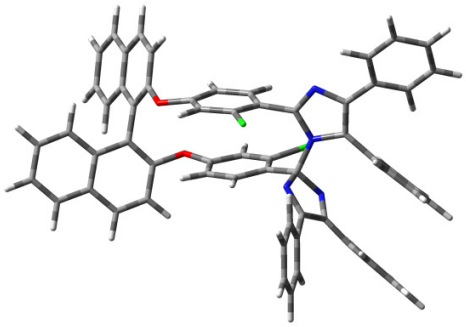


Figure S32. Optimized structure of **2**.

SCF Done: E(RM062X) = -3674.20681880 A.U.
Zero-point correction = 0.840173 (Hartree/Particle)
Thermal correction to Energy = 0.892764
Thermal correction to Enthalpy = 0.893709
Thermal correction to Gibbs Free Energy = 0.750066
Sum of electronic and zero-point Energies = -3673.366645
Sum of electronic and thermal Energies = -3673.314054
Sum of electronic and thermal Enthalpies = -3673.313110
Sum of electronic and thermal Free Energies = -3673.456753
Low frequencies --- -8.3944 -4.4372 -0.0014 0.0002 0.0013 3.7886
Low frequencies --- 10.5397 16.1376 16.6243

The Result for the TDDFT calculation for **2**

Excitation energies and oscillator strengths:

Excited State 1: Singlet-A 2.5889 eV 478.91 nm f=0.0048 <S**2>=0.000
244 -> 245 0.70233
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-KS) = -3674.69470866
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: Singlet-A 3.3013 eV 375.56 nm f=0.0468 <S**2>=0.000
242 -> 245 0.14252

243 -> 245 0.67263

Excited State 3: Singlet-A 3.5666 eV 347.63 nm f=0.0044 <S**2>=0.000
235 -> 245 -0.12277
239 -> 245 0.51249
240 -> 245 -0.42995

Excited State 4: Singlet-A 3.7223 eV 333.09 nm f=0.0240 <S**2>=0.000
237 -> 245 -0.14892
242 -> 245 0.66210
243 -> 245 -0.13513

Excited State 5: Singlet-A 3.8042 eV 325.92 nm f=0.0151 <S**2>=0.000
225 -> 245 0.11349
226 -> 245 0.12050
232 -> 245 -0.11062
235 -> 245 -0.15999
236 -> 245 -0.35050
238 -> 245 -0.20708
239 -> 245 0.26061
240 -> 245 0.40069

Excited State 6: Singlet-A 3.8350 eV 323.29 nm f=0.0125 <S**2>=0.000
244 -> 246 0.66336
244 -> 248 -0.20970

Excited State 7: Singlet-A 3.8541 eV 321.69 nm f=0.0004 <S**2>=0.000
241 -> 245 0.68634
243 -> 245 0.11487

Excited State 8: Singlet-A 3.8806 eV 319.50 nm f=0.0051 <S**2>=0.000
244 -> 246 0.16945
244 -> 247 -0.29360
244 -> 248 0.60508

Excited State 9: Singlet-A 3.8832 eV 319.29 nm f=0.0213 <S**2>=0.000

235 -> 245 0.12062

236 -> 245 -0.14188

237 -> 245 0.12076

238 -> 245 0.57423

239 -> 245 0.18518

240 -> 245 0.21616

Excited State 10: Singlet-A 3.9538 eV 313.58 nm f=0.0006 <S**2>=0.000

244 -> 246 0.12454

244 -> 247 0.62414

244 -> 248 0.27435

Excited State 11: Singlet-A 3.9781 eV 311.67 nm f=0.0210 <S**2>=0.000

225 -> 245 -0.11472

226 -> 245 -0.14065

230 -> 245 0.12515

232 -> 245 0.16552

235 -> 245 0.37227

237 -> 245 -0.34277

238 -> 245 -0.18746

239 -> 245 0.25014

240 -> 245 0.12987

Excited State 12: Singlet-A 4.0425 eV 306.70 nm f=0.0225 <S**2>=0.000

235 -> 245 -0.16573

236 -> 245 0.52101

237 -> 245 0.26014

239 -> 245 0.22873

240 -> 245 0.25035

Excited State 13: Singlet-A 4.1264 eV 300.46 nm f=0.1130 <S**2>=0.000

237 -> 245 0.17576

242 -> 246 -0.13356

243 -> 246 0.60353

Excited State 14: Singlet-A 4.1308 eV 300.15 nm f=0.0094 <S**2>=0.000

229 -> 245 0.14237

230 -> 245 0.13298

233 -> 245 0.13891

235 -> 245 0.18596

236 -> 245 -0.20204

237 -> 245 0.44354

238 -> 245 -0.21610

242 -> 245 0.11484

243 -> 246 -0.24144

Excited State 15: Singlet-A 4.1599 eV 298.04 nm f=0.0081 <S**2>=0.000

244 -> 249 0.66673

244 -> 250 -0.12382

Excited State 16: Singlet-A 4.1931 eV 295.69 nm f=0.0199 <S**2>=0.000

241 -> 246 0.11137

241 -> 247 0.15026

242 -> 246 -0.15761

242 -> 247 -0.10474

243 -> 247 0.59634

Excited State 17: Singlet-A 4.2155 eV 294.11 nm f=0.0540 <S**2>=0.000

242 -> 246 0.63773

243 -> 246 0.13329

243 -> 247 0.13092

Excited State 18: Singlet-A 4.2279 eV 293.25 nm f=0.0069 <S**2>=0.000

225 -> 245 0.20477

230 -> 245 -0.22563

231 -> 245 -0.27296

232 -> 245 -0.30516

233 -> 245 0.14562

235 -> 245 0.38704

236 -> 245 0.12607

Excited State 19: Singlet-A 4.2689 eV 290.43 nm f=0.0100 <S**2>=0.000

231 -> 245 0.16721

233 -> 245 0.47534

235 -> 245 -0.16804

244 -> 250 -0.16922

244 -> 252 0.28618

244 -> 254 -0.19229

Excited State 20: Singlet-A 4.2923 eV 288.85 nm f=0.0167 <S**2>=0.000

233 -> 245 0.17011

241 -> 246 -0.36241

241 -> 247 0.21078

242 -> 247 0.46284

244 -> 250 0.10439

244 -> 252 -0.15783

Excited State 21: Singlet-A 4.2954 eV 288.64 nm f=0.1051 <S**2>=0.000

233 -> 245 -0.35871

241 -> 246 -0.15392

241 -> 247 0.10232

242 -> 247 0.19912

244 -> 250 -0.23783

244 -> 252 0.37849

244 -> 254 -0.19980

Excited State 22: Singlet-A 4.3711 eV 283.65 nm f=0.1093 <S**2>=0.000

230 -> 245 -0.21504

231 -> 245 -0.36046

232 -> 245 0.51649

233 -> 245 0.10456

Excited State 23: Singlet-A 4.4003 eV 281.76 nm f=0.0093 <S**2>=0.000
234 -> 246 0.12017
241 -> 246 0.49154
241 -> 247 0.11281
242 -> 247 0.30454
242 -> 250 -0.10596
243 -> 246 -0.10690
243 -> 249 0.18431

Excited State 24: Singlet-A 4.4082 eV 281.26 nm f=0.0593 <S**2>=0.000
244 -> 250 -0.31449
244 -> 251 -0.32839
244 -> 253 -0.20452
244 -> 254 0.45273

Excited State 25: Singlet-A 4.4402 eV 279.23 nm f=0.0285 <S**2>=0.000
241 -> 247 0.10037
244 -> 250 -0.17301
244 -> 251 0.60233
244 -> 252 0.11248
244 -> 253 -0.10421
244 -> 254 0.22175

Excited State 26: Singlet-A 4.4530 eV 278.43 nm f=0.0548 <S**2>=0.000
241 -> 247 0.57477
242 -> 247 -0.25317
243 -> 247 -0.19792

Excited State 27: Singlet-A 4.4841 eV 276.50 nm f=0.0285 <S**2>=0.000
229 -> 245 -0.31584
230 -> 245 0.29732
231 -> 245 -0.15674

234 -> 245 0.32120

241 -> 246 0.14392

243 -> 249 -0.28451

Excited State 28: Singlet-A 4.5133 eV 274.71 nm f=0.0183 <S**2>=0.000

231 -> 245 -0.10182

234 -> 245 0.39057

237 -> 246 0.11037

238 -> 246 -0.10819

241 -> 247 0.13122

241 -> 250 0.12652

242 -> 249 0.14104

243 -> 249 0.39083

Excited State 29: Singlet-A 4.5335 eV 273.49 nm f=0.0086 <S**2>=0.000

229 -> 245 0.10074

234 -> 245 -0.15883

234 -> 247 -0.16648

237 -> 246 0.22083

238 -> 246 -0.20912

241 -> 249 0.12574

241 -> 250 0.19958

242 -> 249 0.10848

242 -> 253 -0.15149

243 -> 247 0.16228

243 -> 248 -0.20719

243 -> 249 -0.15781

243 -> 250 0.20609

Excited State 30: Singlet-A 4.5380 eV 273.21 nm f=0.0108 <S**2>=0.000

225 -> 245 0.16495

226 -> 245 0.14985

228 -> 245 0.34256

229 -> 245 -0.11803

230 -> 245 0.35940

232 -> 245 0.13717

234 -> 245 -0.18873

243 -> 249 0.27113

Excited State 31: Singlet-A 4.5454 eV 272.77 nm f=0.0080 <S**2>=0.000

244 -> 249 0.14513

244 -> 250 0.42590

244 -> 252 0.39834

244 -> 254 0.22265

244 -> 256 -0.14996

Excited State 32: Singlet-A 4.5582 eV 272.00 nm f=0.0092 <S**2>=0.000

228 -> 245 0.33747

229 -> 245 0.37594

231 -> 245 0.11219

234 -> 245 0.19728

234 -> 246 -0.10353

237 -> 247 0.11409

242 -> 247 0.10464

242 -> 250 0.15813

243 -> 249 -0.12728

Excited State 33: Singlet-A 4.5652 eV 271.59 nm f=0.0360 <S**2>=0.000

228 -> 245 -0.15411

229 -> 245 -0.16184

234 -> 245 -0.22600

234 -> 246 -0.21776

237 -> 247 0.19334

238 -> 247 -0.16875

241 -> 249 0.12012

241 -> 253 -0.13384

242 -> 247 0.15328

242 -> 250 0.28947

243 -> 246 0.11144

Excited State 34: Singlet-A 4.5831 eV 270.53 nm f=0.0186 <S**2>=0.000
227 -> 245 0.11540
242 -> 248 0.11452
243 -> 248 0.59511

Excited State 35: Singlet-A 4.5923 eV 269.98 nm f=0.0205 <S**2>=0.000
222 -> 245 0.16669
225 -> 245 0.13020
226 -> 245 0.23518
227 -> 245 0.39397
228 -> 245 -0.18830
231 -> 245 0.12620
233 -> 245 -0.11781
234 -> 245 0.23883
235 -> 245 0.15303
243 -> 248 -0.14956

Excited State 36: Singlet-A 4.6078 eV 269.07 nm f=0.0094 <S**2>=0.000
244 -> 255 0.16305
244 -> 256 0.39635
244 -> 258 0.44585
244 -> 260 -0.16994

Excited State 37: Singlet-A 4.6679 eV 265.61 nm f=0.0052 <S**2>=0.000
225 -> 245 0.17555
226 -> 245 0.10028
227 -> 245 -0.30212
229 -> 245 -0.30053
230 -> 245 -0.23427
231 -> 245 0.37005
232 -> 245 0.16880
237 -> 245 0.11459

Excited State 38: Singlet-A 4.7331 eV 261.95 nm f=0.0113 <S**2>=0.000

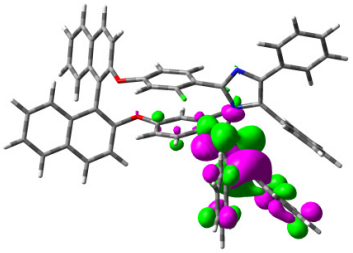
222 -> 245 0.14079
225 -> 245 -0.30046
226 -> 245 -0.11572
227 -> 245 0.17278
228 -> 245 0.32077
229 -> 245 -0.21429
230 -> 245 -0.18313
233 -> 245 -0.12377
244 -> 253 0.17922
244 -> 257 -0.16477

Excited State 39: Singlet-A 4.7387 eV 261.64 nm f=0.1070 <S**2>=0.000

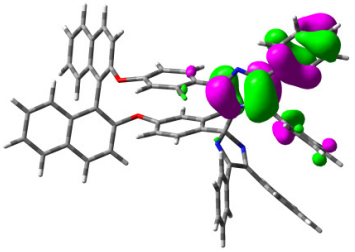
225 -> 245 0.13657
228 -> 245 -0.14846
229 -> 245 0.10462
244 -> 253 0.39082
244 -> 254 0.15747
244 -> 256 0.16443
244 -> 257 -0.38951
244 -> 258 -0.11781

Excited State 40: Singlet-A 4.7959 eV 258.52 nm f=0.0706 <S**2>=0.000

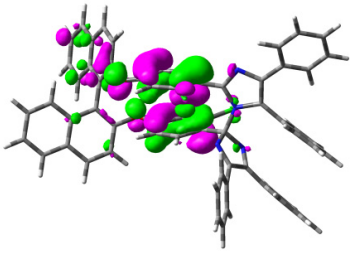
238 -> 246 0.14386
239 -> 248 -0.25307
240 -> 248 0.21679
243 -> 250 0.25467
243 -> 252 -0.16128
244 -> 255 0.36907
244 -> 256 -0.19873



MO245



MO244



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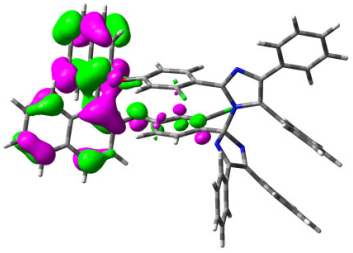
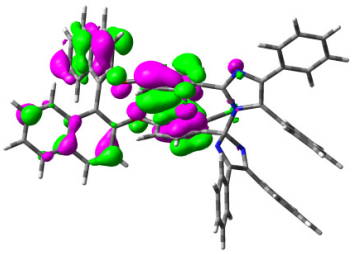


MO242



MO237

Figure S33. The relevant molecular orbitals of **2** calculated at the MPW1PW91/6-31+G(d)//M062X/6-31G(d) level.

Table S5. Standard orientation of the optimized geometry for **2R**.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms) X	Y	Z
1	6	0	-8.743167	3.193483	1.895170
2	6	0	-8.755948	2.689803	0.573852
3	6	0	-7.840986	1.748362	0.174556
4	6	0	-6.860182	1.260755	1.079212
5	6	0	-6.847352	1.770993	2.407875
6	6	0	-7.807474	2.742130	2.789660
7	6	0	-5.886582	0.285882	0.688180
8	6	0	-4.962302	-0.128770	1.617107
9	6	0	-4.952589	0.359802	2.945872
10	6	0	-5.874601	1.293390	3.326429
11	6	0	-5.886129	-0.285917	-0.688452
12	6	0	-6.859568	-1.260673	-1.080154
13	6	0	-6.845901	-1.770941	-2.408786
14	6	0	-5.872500	-1.293431	-3.326683
15	6	0	-4.950632	-0.359954	-2.945508
16	6	0	-4.961119	0.128603	-1.616736
17	6	0	-7.841030	-1.748172	-0.176145
18	6	0	-8.755801	-2.689549	-0.576023
19	6	0	-8.742169	-3.193278	-1.897320
20	6	0	-7.805839	-2.742018	-2.791190
21	8	0	-4.074935	-1.122685	1.258621
22	8	0	-4.073796	1.122307	-1.257599
23	6	0	-2.734458	-0.928190	1.422205
24	6	0	-2.733315	0.928014	-1.421540
25	6	0	-1.952084	-2.075147	1.306873
26	6	0	-0.570154	-1.981771	1.341258
27	6	0	0.071654	-0.735610	1.511781
28	6	0	-0.757558	0.397830	1.634500
29	6	0	-2.137689	0.322126	1.601153
30	6	0	-1.951039	2.075063	-1.306607
31	6	0	-0.569087	1.981792	-1.341151

32	6	0	0.072796	0.735634	-1.511295
33	6	0	-0.756357	-0.397898	-1.633708
34	6	0	-2.136477	-0.322292	-1.600296
35	6	0	1.505456	-0.524422	1.550151
36	6	0	1.506582	0.524416	-1.549535
37	7	0	2.034320	-0.733048	-1.428788
38	6	0	3.338685	-0.558740	-1.565339
39	6	0	3.585592	0.890605	-1.769676
40	7	0	2.415052	1.508612	-1.767315
41	7	0	2.413808	-1.508779	1.768392
42	6	0	3.584296	-0.890794	1.770450
43	6	0	3.337716	0.558460	1.565692
44	7	0	2.033319	0.732911	1.429110
45	6	0	4.258945	1.696758	1.591857
46	6	0	4.807126	-1.683175	1.907494
47	6	0	4.808194	1.683490	-1.906647
48	6	0	4.259425	-1.697479	-1.591525
49	6	0	3.913628	-2.863264	-0.890088
50	6	0	4.765436	-3.958903	-0.894217
51	6	0	5.960588	-3.918196	-1.611645
52	6	0	6.291464	-2.780291	-2.344328
53	6	0	5.445231	-1.677277	-2.339837
54	6	0	6.038276	1.280018	-1.370695
55	6	0	7.138661	2.127777	-1.409294
56	6	0	7.032849	3.384893	-1.999621
57	6	0	5.816407	3.792559	-2.547736
58	6	0	4.710191	2.957048	-2.491636
59	6	0	4.711090	-2.955174	2.495954
60	6	0	5.818044	-3.789820	2.551582
61	6	0	7.032947	-3.382718	1.999644
62	6	0	7.136614	-2.127135	1.405608
63	6	0	6.035501	-1.280345	1.367371
64	6	0	3.912913	2.863388	0.891920
65	6	0	4.765309	3.958555	0.896046

66	6	0	5.961319	3.916581	1.611969
67	6	0	6.292457	2.777846	2.343219
68	6	0	5.445655	1.675288	2.338733
69	1	0	-9.473629	3.937750	2.196506
70	1	0	-9.496138	3.054614	-0.131493
71	1	0	-7.854147	1.369650	-0.842616
72	1	0	-7.784285	3.121925	3.807814
73	1	0	-4.211053	-0.023858	3.639649
74	1	0	-5.880039	1.675811	4.343443
75	1	0	-5.877294	-1.675825	-4.343711
76	1	0	-4.208628	0.023600	-3.638840
77	1	0	-7.854844	-1.369430	0.841007
78	1	0	-9.496490	-3.054278	0.128841
79	1	0	-9.472473	-3.937503	-2.199131
80	1	0	-7.781985	-3.121855	-3.809313
81	1	0	-2.437187	-3.031424	1.149885
82	1	0	-0.267546	1.360303	1.741870
83	1	0	-2.742324	1.220467	1.661378
84	1	0	-2.436197	3.031349	-1.149850
85	1	0	-0.266278	-1.360365	-1.740830
86	1	0	-2.741059	-1.220689	-1.660281
87	1	0	2.989665	-2.872970	-0.318249
88	1	0	4.502691	-4.843816	-0.322871
89	1	0	6.627378	-4.775356	-1.606788
90	1	0	7.205736	-2.754417	-2.929304
91	1	0	5.693426	-0.803155	-2.933844
92	1	0	6.126153	0.302327	-0.913782
93	1	0	8.078279	1.807521	-0.969485
94	1	0	7.894367	4.045101	-2.031442
95	1	0	5.728944	4.770984	-3.010548
96	1	0	3.748785	3.272963	-2.883116
97	1	0	3.750766	-3.270696	2.890431
98	1	0	5.732335	-4.767150	3.017030
99	1	0	7.894929	-4.042337	2.031153

100	1	0	8.074849	-1.807659	0.962278
101	1	0	6.120767	-0.304411	0.906014
102	1	0	2.988293	2.874095	0.321167
103	1	0	4.502354	4.844159	0.325865
104	1	0	6.628551	4.773397	1.607019
105	1	0	7.207446	2.750906	2.927026
106	1	0	5.694195	0.800527	2.931604
107	17	0	0.292817	3.470176	-1.086718
108	17	0	0.291906	-3.469887	1.086000

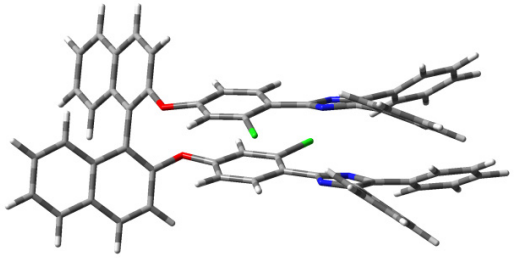


Figure S34. Optimized structure of **2R**.

SCF Done: E(UM062X) = -3674.18880187 A.U.

S**2 before annihilation 0.8370, after 0.1535

Zero-point correction = 0.838224 (Hartree/Particle)

Thermal correction to Energy = 0.891260

Thermal correction to Enthalpy = 0.892204

Thermal correction to Gibbs Free Energy = 0.750214

Sum of electronic and zero-point Energies = -3673.350578

Sum of electronic and thermal Energies = -3673.297542

Sum of electronic and thermal Enthalpies = -3673.296598

Sum of electronic and thermal Free Energies = -3673.438588

Low frequencies --- -3.3728 -0.0020 0.0007 0.0018 6.1894 7.1811

Low frequencies --- 13.6164 18.0737 25.5366

The Result for the TDDFT calculation for **2R**

Excitation energies and oscillator strengths:

Excited State 1: 1.732-A 1.0269 eV 1207.42 nm f=0.0024 <S**2>=0.500

244A -> 245A 0.70505

244B -> 245B 0.70424

This state for optimization and/or second-order correction.

Total Energy, E(TD-HF/TD-KS) = -3674.73338606

Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: 0.983-A 1.4051 eV 882.41 nm f=0.0534 <S**2>=-0.009

244A -> 245A -0.70133

244B -> 245B 0.70213

Excited State 3: 2.596-A 1.8925 eV 655.14 nm f=0.0000 <S**2>=1.434

239A -> 245A 0.22041

240A -> 245A 0.62387

239B -> 245B 0.22133

240B -> 245B 0.62187

Excited State 4: 2.642-A 1.9760 eV 627.46 nm f=0.0051 <S**2>=1.495

241A -> 245A 0.32752

242A -> 245A -0.45441

243A -> 245A 0.37723

241B -> 245B -0.32711

242B -> 245B 0.45496

243B -> 245B -0.37517

Excited State 5: 2.217-A 2.0193 eV 613.99 nm f=0.0447 <S**2>=0.978

234A -> 245A 0.11661

238A -> 245A -0.11651

239A -> 245A -0.35280

240A -> 245A -0.52430

234B -> 245B -0.11681

238B -> 245B 0.10273

239B -> 245B 0.35807

240B -> 245B 0.52625

Excited State 6: 2.128-A 2.1720 eV 570.82 nm f=0.2472 <S**2>=0.883

231A -> 245A	0.11548
232A -> 245A	-0.22509
233A -> 245A	0.12310
234A -> 245A	0.16893
236A -> 245A	-0.10775
237A -> 245A	0.16252
238A -> 245A	0.17953
241A -> 245A	-0.24453
242A -> 245A	0.34304
243A -> 245A	-0.32478
231B -> 245B	0.11751
232B -> 245B	-0.22254
233B -> 245B	0.12187
234B -> 245B	0.16886
236B -> 245B	-0.10499
237B -> 245B	0.16535
238B -> 245B	0.17589
241B -> 245B	-0.24543
242B -> 245B	0.34502
243B -> 245B	-0.32448

Excited State 7: 2.279-A 2.3010 eV 538.82 nm f=0.0004 <S**2>=1.048

228A -> 245A	-0.16440
229A -> 245A	0.10852
231A -> 245A	-0.17570
232A -> 245A	0.41800
233A -> 245A	-0.21821
234A -> 245A	-0.21958
235A -> 245A	0.11595
236A -> 245A	0.18744
237A -> 245A	-0.25095
238A -> 245A	-0.11061

243A -> 245A -0.13175
228B -> 245B 0.16071
229B -> 245B -0.10872
231B -> 245B 0.17832
232B -> 245B -0.41162
233B -> 245B 0.21449
234B -> 245B 0.21820
235B -> 245B -0.11203
236B -> 245B -0.18089
237B -> 245B 0.25311
238B -> 245B 0.10552
243B -> 245B 0.12402

Excited State 8: 2.420-A 2.3417 eV 529.46 nm f=0.1591 <S**2>=1.214
228A -> 245A 0.16369
231A -> 245A 0.10694
232A -> 245A -0.28802
233A -> 245A 0.17274
234A -> 245A 0.17469
236A -> 245A -0.20756
237A -> 245A 0.15187
240A -> 245A 0.12100
243A -> 245A 0.40972
228B -> 245B 0.16527
231B -> 245B 0.11305
232B -> 245B -0.29394
233B -> 245B 0.17559
234B -> 245B 0.17947
236B -> 245B -0.20795
237B -> 245B 0.16024
240B -> 245B 0.12262
243B -> 245B 0.41023

Excited State 9: 2.558-A 2.3859 eV 519.65 nm f=0.0082 <S**2>=1.386

234A -> 245A -0.10652
239A -> 245A 0.21324
241A -> 245A -0.17763
242A -> 245A 0.45519
243A -> 245A 0.37767
243A -> 246A 0.10169
234B -> 245B -0.10489
239B -> 245B 0.20080
241B -> 245B -0.17286
242B -> 245B 0.44408
243B -> 245B 0.36367
243B -> 246B 0.10126

Excited State 10: 2.075-A 2.4020 eV 516.18 nm f=0.0014 <S**2>=0.826
239A -> 245A -0.33735
240A -> 245A 0.19453
241A -> 245A 0.14473
242A -> 245A -0.29291
243A -> 245A -0.44686
239B -> 245B 0.34344
240B -> 245B -0.19480
241B -> 245B -0.15049
242B -> 245B 0.30622
243B -> 245B 0.46147

Excited State 11: 2.136-A 2.4679 eV 502.38 nm f=0.0190 <S**2>=0.891
238A -> 245A -0.18134
239A -> 245A -0.38678
240A -> 245A 0.38798
242A -> 245A 0.20797
243A -> 245A 0.31065
238B -> 245B 0.16779
239B -> 245B 0.39259
240B -> 245B -0.38848

242B -> 245B -0.20821
243B -> 245B -0.31084

Excited State 12: 2.079-A 2.5253 eV 490.96 nm f=0.0603 <S**2>=0.830
233A -> 245A 0.10379
236A -> 245A 0.10212
238A -> 245A 0.13338
239A -> 245A 0.55199
240A -> 245A -0.24334
242A -> 245A -0.23526
233B -> 245B 0.10427
236B -> 245B 0.10032
238B -> 245B 0.11409
239B -> 245B 0.55748
240B -> 245B -0.24429
242B -> 245B -0.23620

Excited State 13: 2.969-A 2.5445 eV 487.27 nm f=0.0008 <S**2>=1.954
241A -> 245A 0.46096
241A -> 246A 0.22293
242A -> 245A 0.31833
242A -> 246A 0.14996
242A -> 247A -0.11762
243A -> 247A -0.22040
241B -> 245B -0.45855
241B -> 246B -0.22292
242B -> 245B -0.31629
242B -> 246B -0.14992
242B -> 247B 0.11677
243B -> 247B 0.22086

Excited State 14: 2.616-A 2.5645 eV 483.46 nm f=0.0052 <S**2>=1.462
235A -> 245A -0.13478
238A -> 245A -0.31090

239A -> 245A 0.11927
241A -> 245A 0.34410
241A -> 247A 0.18593
242A -> 245A 0.26836
242A -> 246A -0.12116
242A -> 247A 0.11099
243A -> 246A -0.21723
235B -> 245B -0.13462
238B -> 245B -0.32026
239B -> 245B 0.10954
241B -> 245B 0.34402
241B -> 247B 0.18592
242B -> 245B 0.26746
242B -> 246B -0.12014
242B -> 247B 0.11128
243B -> 246B -0.21776

Excited State 15: 2.560-A 2.5914 eV 478.45 nm f=0.0023 <S**2>=1.389
232A -> 245A -0.11809
235A -> 245A 0.11205
237A -> 245A -0.54763
238A -> 245A 0.28704
232B -> 245B 0.12036
235B -> 245B -0.14189
236B -> 245B 0.11723
237B -> 245B 0.56902
238B -> 245B -0.33958

Excited State 16: 3.030-A 2.5935 eV 478.07 nm f=0.0053 <S**2>=2.046
235A -> 245A 0.21457
236A -> 245A -0.27170
237A -> 245A -0.14143
238A -> 245A 0.31346
241A -> 247A 0.24285

242A -> 246A -0.16040
242A -> 247A 0.14377
243A -> 245A 0.15904
243A -> 246A -0.28955
235B -> 245B 0.19229
236B -> 245B -0.25864
238B -> 245B 0.26734
241B -> 247B 0.24267
242B -> 246B -0.16049
242B -> 247B 0.14493
243B -> 245B 0.15101
243B -> 246B -0.28945

Excited State 17: 2.146-A 2.6183 eV 473.53 nm f=0.0001 <S**2>=0.901
235A -> 245A 0.15751
237A -> 245A -0.32983
238A -> 245A 0.27034
241A -> 245A 0.43846
242A -> 245A 0.16462
243A -> 246A 0.12014
235B -> 245B 0.15104
237B -> 245B -0.31352
238B -> 245B 0.26306
241B -> 245B 0.43816
242B -> 245B 0.16469
243B -> 246B 0.12012

Excited State 18: 2.337-A 2.6583 eV 466.40 nm f=0.0132 <S**2>=1.116
231A -> 245A -0.12404
232A -> 245A 0.17551
234A -> 245A -0.18093
236A -> 245A -0.33440
237A -> 245A 0.39886
238A -> 245A 0.13125

241A -> 245A 0.26746
231B -> 245B -0.12564
232B -> 245B 0.17553
234B -> 245B -0.17997
236B -> 245B -0.33441
237B -> 245B 0.40035
238B -> 245B 0.12956
241B -> 245B 0.25562

Excited State 19: 3.140-A 2.6609 eV 465.96 nm f=0.0010 <S**2>=2.214
236A -> 245A -0.11061
241A -> 245A -0.32699
241A -> 246A 0.28414
242A -> 245A -0.18361
242A -> 246A 0.18807
242A -> 247A -0.16129
243A -> 247A -0.29824
241B -> 245B 0.33792
241B -> 246B -0.28424
242B -> 245B 0.18504
242B -> 246B -0.18629
242B -> 247B 0.15849
243B -> 245B -0.10127
243B -> 247B 0.29969

Excited State 20: 2.141-A 2.7080 eV 457.85 nm f=0.0022 <S**2>=0.896
230A -> 245A 0.13351
231A -> 245A -0.17720
232A -> 245A 0.15189
234A -> 245A -0.16016
236A -> 245A -0.43372
237A -> 245A 0.20443
238A -> 245A 0.30760
239A -> 245A -0.11555

241A -> 245A 0.13943
230B -> 245B 0.13563
231B -> 245B 0.17577
232B -> 245B -0.15333
234B -> 245B 0.16017
236B -> 245B 0.43547
237B -> 245B -0.20921
238B -> 245B -0.31155
239B -> 245B 0.10322
241B -> 245B -0.14066

Excited State 21: 2.391-A 2.7351 eV 453.30 nm f=0.0017 <S**2>=1.180
230A -> 245A -0.12367
231A -> 245A -0.11965
233A -> 245A 0.16454
235A -> 245A 0.46345
236A -> 245A 0.33158
237A -> 245A 0.36045
239A -> 245A -0.12413
230B -> 245B 0.10919
231B -> 245B -0.11239
233B -> 245B 0.15128
235B -> 245B 0.38822
236B -> 245B 0.28794
237B -> 245B 0.32993
239B -> 245B -0.10335

Excited State 22: 2.058-A 2.7504 eV 450.78 nm f=0.0002 <S**2>=0.809
232A -> 245A 0.15929
235A -> 245A -0.51191
236A -> 245A -0.24268
237A -> 245A -0.15447
238A -> 245A -0.11428
239A -> 245A 0.10707

229B -> 245B 0.10123
232B -> 245B -0.17514
235B -> 245B 0.58130
236B -> 245B 0.28447
237B -> 245B 0.19961
238B -> 245B 0.12397
239B -> 245B -0.11783

Excited State 23: 2.520-A 2.7576 eV 449.61 nm f=0.0047 <S**2>=1.337
228A -> 245A 0.11671
232A -> 245A -0.29266
233A -> 245A -0.19978
234A -> 245A -0.13311
235A -> 245A 0.34453
236A -> 245A -0.21551
238A -> 245A -0.28220
243A -> 245A -0.16824
228B -> 245B 0.11532
232B -> 245B -0.28989
233B -> 245B -0.20023
234B -> 245B -0.13546
235B -> 245B 0.33751
236B -> 245B -0.22193
238B -> 245B -0.28420
243B -> 245B -0.16913

Excited State 24: 2.265-A 2.7836 eV 445.41 nm f=0.0011 <S**2>=1.033
227A -> 245A -0.13926
232A -> 245A 0.34339
233A -> 245A 0.24256
234A -> 245A 0.24874
235A -> 245A -0.16548
236A -> 245A 0.18996
238A -> 245A 0.30481

243A -> 245A 0.10264
227B -> 245B 0.13896
230B -> 245B -0.10271
232B -> 245B -0.34222
233B -> 245B -0.24316
234B -> 245B -0.25031
235B -> 245B 0.15926
236B -> 245B -0.19524
238B -> 245B -0.30736
243B -> 245B -0.10342

Excited State 25: 2.471-A 2.8877 eV 429.35 nm f=0.0012 <S**2>=1.276
226A -> 245A -0.12941
229A -> 245A 0.13212
230A -> 245A -0.12811
231A -> 245A -0.34770
232A -> 245A 0.14724
233A -> 245A 0.36933
234A -> 245A 0.17879
236A -> 245A -0.21567
237A -> 245A -0.10970
238A -> 245A -0.15350
226B -> 245B -0.12879
229B -> 245B 0.13074
230B -> 245B 0.12543
231B -> 245B -0.35527
232B -> 245B 0.14272
233B -> 245B 0.37355
234B -> 245B 0.17677
236B -> 245B -0.21458
237B -> 245B -0.11034
238B -> 245B -0.15410

Excited State 26: 2.510-A 2.9147 eV 425.38 nm f=0.0025 <S**2>=1.325

227A -> 245A 0.29624
229A -> 245A 0.26108
231A -> 245A -0.29894
233A -> 245A 0.37794
234A -> 245A -0.13741
235A -> 245A -0.17013
227B -> 245B -0.29645
229B -> 245B -0.25615
231B -> 245B 0.29736
233B -> 245B -0.37045
234B -> 245B 0.13862
235B -> 245B 0.17365

Excited State 27: 2.183-A 2.9445 eV 421.07 nm f=0.0049 <S**2>=0.942
225A -> 245A 0.18045
226A -> 245A -0.20958
227A -> 245A -0.22526
229A -> 245A 0.15300
230A -> 245A -0.12028
231A -> 245A -0.19095
233A -> 245A 0.14726
234A -> 245A 0.25235
235A -> 245A 0.14537
236A -> 245A -0.26492
238A -> 245A -0.23904
239A -> 245A 0.16013
225B -> 245B -0.17724
226B -> 245B 0.20825
227B -> 245B 0.22253
229B -> 245B -0.15077
230B -> 245B -0.11967
231B -> 245B 0.19847
233B -> 245B -0.15040
234B -> 245B -0.25119

235B -> 245B -0.14139
236B -> 245B 0.26736
238B -> 245B 0.24499
239B -> 245B -0.14867

Excited State 28: 2.370-A 2.9677 eV 417.78 nm f=0.0040 <S**2>=1.154
225A -> 245A -0.23132
226A -> 245A 0.45724
227A -> 245A 0.12864
228A -> 245A 0.19492
230A -> 245A 0.13481
232A -> 245A 0.25475
233A -> 245A 0.18317
239A -> 245A -0.12111
225B -> 245B -0.23800
226B -> 245B 0.47384
227B -> 245B 0.13309
228B -> 245B 0.20010
230B -> 245B -0.13838
232B -> 245B 0.26444
233B -> 245B 0.19278
239B -> 245B -0.12569

Excited State 29: 2.171-A 2.9891 eV 414.78 nm f=0.0010 <S**2>=0.929
225A -> 245A -0.24746
226A -> 245A 0.36235
228A -> 245A 0.22397
229A -> 245A -0.13770
232A -> 245A 0.27758
233A -> 245A 0.22724
235A -> 245A 0.18670
238A -> 245A -0.17255
225B -> 245B 0.23572
226B -> 245B -0.34670

228B -> 245B -0.21539
229B -> 245B 0.13800
232B -> 245B -0.27337
233B -> 245B -0.21994
234B -> 245B -0.10203
235B -> 245B -0.18721
238B -> 245B 0.17071

Excited State 30: 2.394-A 3.0190 eV 410.68 nm f=0.0021 <S**2>=1.183
225A -> 245A 0.11842
226A -> 245A -0.18308
227A -> 245A -0.12672
228A -> 245A 0.16013
230A -> 245A 0.20417
231A -> 245A 0.33252
232A -> 245A 0.30795
234A -> 245A 0.19133
235A -> 245A 0.24011
237A -> 245A 0.10657
225B -> 245B 0.12329
226B -> 245B -0.19222
227B -> 245B -0.12665
228B -> 245B 0.15802
230B -> 245B -0.20197
231B -> 245B 0.32988
232B -> 245B 0.30948
234B -> 245B 0.19115
235B -> 245B 0.24283
237B -> 245B 0.10350
238B -> 245B -0.10365

Excited State 31: 2.431-A 3.0865 eV 401.70 nm f=0.0005 <S**2>=1.228
225A -> 245A 0.16963
226A -> 245A -0.19567

229A -> 245A -0.10539
230A -> 245A 0.58915
236A -> 245A 0.17374
225B -> 245B -0.16794
226B -> 245B 0.19702
229B -> 245B 0.10204
230B -> 245B 0.59066
236B -> 245B -0.17615

Excited State 32: 2.210-A 3.1132 eV 398.26 nm f=0.0005 <S**2>=0.971
225A -> 245A 0.12892
226A -> 245A -0.20119
227A -> 245A 0.10065
228A -> 245A 0.16132
230A -> 245A 0.44906
231A -> 245A -0.11935
233A -> 245A 0.15288
234A -> 245A -0.28231
236A -> 245A 0.15281
225B -> 245B 0.12845
226B -> 245B -0.20279
227B -> 245B 0.10691
228B -> 245B 0.16151
230B -> 245B -0.44425
231B -> 245B -0.10977
233B -> 245B 0.15606
234B -> 245B -0.28457
236B -> 245B 0.15154

Excited State 33: 2.467-A 3.1355 eV 395.41 nm f=0.0011 <S**2>=1.271
225A -> 245A 0.21886
226A -> 245A -0.23978
227A -> 245A 0.23972
228A -> 245A 0.14561

230A -> 245A -0.16684
231A -> 245A 0.35698
232A -> 245A 0.16643
233A -> 245A 0.10676
234A -> 245A -0.21779
235A -> 245A 0.15003
225B -> 245B -0.21421
226B -> 245B 0.23648
227B -> 245B -0.24013
228B -> 245B -0.14142
230B -> 245B -0.17639
231B -> 245B -0.35208
232B -> 245B -0.16893
233B -> 245B -0.10668
234B -> 245B 0.21309
235B -> 245B -0.15057

Excited State 34: 2.151-A 3.1837 eV 389.43 nm f=0.0001 <S**2>=0.907
226A -> 245A -0.11310
227A -> 245A 0.20159
228A -> 245A 0.16802
230A -> 245A -0.35801
231A -> 245A 0.29316
233A -> 245A 0.11342
234A -> 245A -0.26159
244A -> 248A 0.15301
226B -> 245B -0.11372
227B -> 245B 0.20500
228B -> 245B 0.16844
230B -> 245B 0.36510
231B -> 245B 0.28635
233B -> 245B 0.11637
234B -> 245B -0.26144
244B -> 248B 0.15297

Excited State 35: 2.810-A 3.2121 eV 386.00 nm f=0.0008 <S**2>=1.724

224A -> 245A	-0.11010
230A -> 245A	0.11717
244A -> 248A	0.52096
224B -> 245B	-0.11034
230B -> 245B	-0.11621
244B -> 248B	0.52082

Excited State 36: 2.651-A 3.2324 eV 383.56 nm f=0.0024 <S**2>=1.507

227A -> 245A	-0.16763
229A -> 245A	-0.25333
230A -> 245A	-0.10972
231A -> 245A	-0.13222
233A -> 245A	0.14373
234A -> 245A	-0.17488
238A -> 245A	0.10003
244A -> 246A	-0.16495
244A -> 249A	-0.33986
244A -> 252A	0.14927
227B -> 245B	0.16781
229B -> 245B	0.24147
230B -> 245B	-0.10135
231B -> 245B	0.12975
233B -> 245B	-0.14134
234B -> 245B	0.17115
244B -> 246B	0.16682
244B -> 249B	0.34282
244B -> 252B	-0.15016

Excited State 37: 2.362-A 3.2459 eV 381.97 nm f=0.0010 <S**2>=1.145

227A -> 245A	-0.12655
228A -> 245A	0.29215
229A -> 245A	0.49251

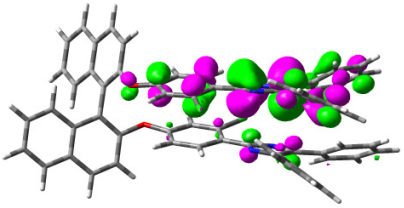
230A -> 245A 0.11106
231A -> 245A 0.21009
233A -> 245A -0.13317
234A -> 245A -0.10421
244A -> 249A -0.14563
227B -> 245B 0.12238
228B -> 245B -0.29399
229B -> 245B -0.46134
230B -> 245B 0.10263
231B -> 245B -0.20505
233B -> 245B 0.12676
234B -> 245B 0.10857
244B -> 249B 0.14521

Excited State 38: 2.092-A 3.2644 eV 379.81 nm f=0.0179 <S**2>=0.844
226A -> 245A 0.12550
228A -> 245A 0.13251
229A -> 245A 0.59783
233A -> 245A -0.13192
226B -> 245B 0.12954
228B -> 245B 0.15773
229B -> 245B 0.62315
233B -> 245B -0.14160

Excited State 39: 2.647-A 3.2915 eV 376.68 nm f=0.1839 <S**2>=1.501
227A -> 245A 0.28445
228A -> 245A -0.20203
229A -> 245A -0.10903
230A -> 245A 0.16378
234A -> 245A 0.18613
239A -> 245A 0.10686
242A -> 249A -0.11173
244A -> 246A -0.18770
244A -> 249A -0.21231

244A -> 252A 0.10529
227B -> 245B 0.28458
228B -> 245B -0.20734
229B -> 245B -0.10737
230B -> 245B -0.16454
234B -> 245B 0.18739
239B -> 245B 0.10318
242B -> 249B -0.11195
244B -> 246B -0.18623
244B -> 249B -0.21014
244B -> 252B 0.10397

Excited State 40: 2.742-A 3.2934 eV 376.46 nm f=0.0354 <S**2>=1.630
220A -> 245A -0.12387
224A -> 245A -0.18847
225A -> 245A -0.10504
227A -> 245A -0.12225
228A -> 245A -0.10870
231A -> 245A 0.12928
237A -> 255A 0.10254
242A -> 248A 0.10988
244A -> 248A -0.36582
244A -> 250A -0.21359
220B -> 245B 0.12326
224B -> 245B 0.18694
225B -> 245B 0.10370
227B -> 245B 0.12387
228B -> 245B 0.10317
231B -> 245B -0.12981
237B -> 255B -0.10182
242B -> 248B -0.10989
244B -> 248B 0.36597
244B -> 250B 0.21352



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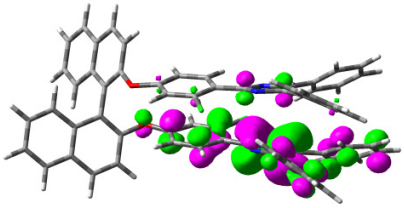

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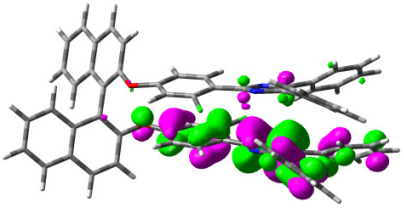

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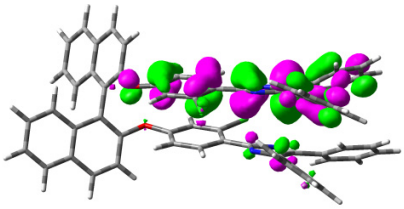

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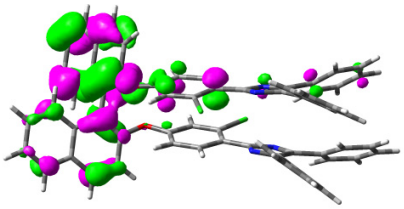

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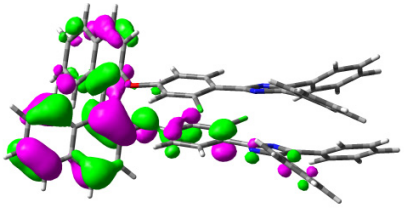

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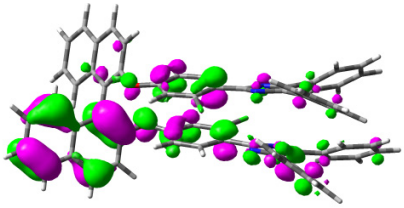

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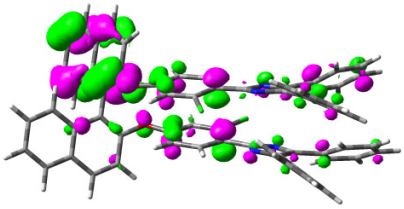

MO242β

Figure S35. The relevant molecular orbitals of **2R** calculated at the UMPW1PW91/6-31+G(d)//UM062X/6-31G(d) level.

11. References

S1. (1) Sheldrick GM. *SHELXS-97* and *SHELXL-97* 1997; University of Gottingen, Germany. (2) Sheldrick GM. *SADABS* 1996; University of Gottingen, Germany.

S2. Gaussian 09, Revision D.01, Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; Scuseria, G. E.; Robb, M. A.; Cheeseman, J. R.; Scalmani, G.; Barone, V.; Mennucci, B.; Petersson, G. A.; Nakatsuji, H.; Caricato, M.; Li, X.; Hratchian, H. P.; Izmaylov, A. F.; Bloino, J.; Zheng, G.; Sonnenberg, J. L.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Vreven, T.; Montgomery, J. A., Jr.; Peralta, J. E.; Ogliaro, F.; Bearpark, M.; Heyd, J. J.; Brothers, E.; Kudin, K. N.; Staroverov, V. N.; Kobayashi, R.; Normand, J.; Raghavachari, K.; Rendell, A.; Burant, J. C.; Iyengar, S. S.; Tomasi, J.; Cossi, M.; Rega, N.; Millam, N. J.; Klene, M.; Knox, J. E.; Cross, J. B.; Bakken, V.; Adamo, C.; Jaramillo, J.; Gomperts, R.; Stratmann, R. E.; Yazyev, O.; Austin, A. J.; Cammi, R.; Pomelli, C.; Ochterski, J. W.; Martin, R. L.; Morokuma, K.; Zakrzewski, V. G.; Voth, G. A.; Salvador, P.; Dannenberg, J. J.; Dapprich, S.; Daniels, A. D.; Farkas, Ö.; Foresman, J. B.; Ortiz, J. V.; Cioslowski, J.; Fox, D. J. Gaussian, Inc., Wallingford CT, 2009.