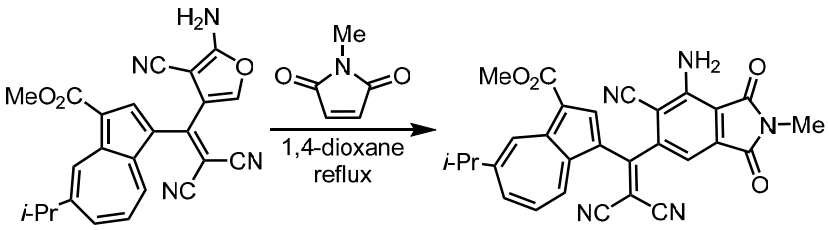
Electronic Supplementary Information (ESI)

Synthesis of Phthalimides Cross-Conjugated with Azulene Ring via Diels-Alder Reaction of 2-Aminofurans with Maleimides, and Their Structural, Optical and Electrochemical Properties

Taku Shoji,* Nanami Iida, Akari Yamazaki, Yukino Ariga, Akira Ohta, Ryuta Sekiguchi, Tatsuki Nagahata, Takuya Nagasawa, Shunji Ito

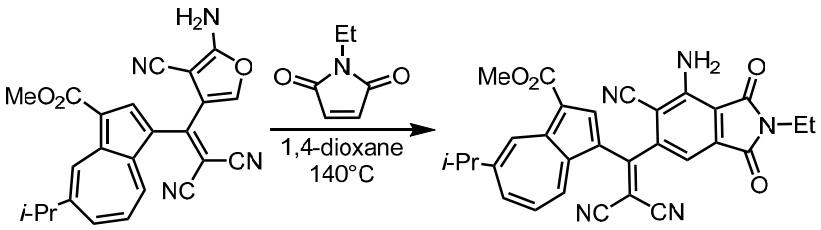
➢ *Contents*

Compound 2a



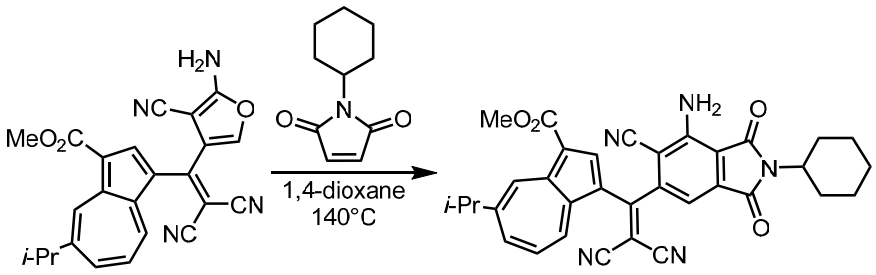
A solution of **1** (100 mg, 0.243 mmol) in *N*-methylmaleimide (55 mg, 0.495 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 23 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with $CHCl_3$ as an eluent to afford **2a** (49 mg, 0.0973 mmol, 42%) as red solid. M.p. 235−237 °C; IR (AT-IR): ν_{max} = 3448 (w), 3347 (w), 2970 (w), 2211 (m), 1771 (m), 1701 (s), 1627 (m), 1601 (w), 1527 (w), 1511 (m), 1483 (s), 1464 (s), 1439 (s), 1414 (s), 1385 (m), 1361 (s), 1337 (m), 1286 (w), 1245 (m), 1210 (s), 1176 (m), 1127 (m), 1091 (w), 1062 (m), 985 (m), 956 (w), 931 (w), 903 (m), 879 (m), 843 (w), 817 (m), 809 (m), 782 (m), 773 (w), 749 (m), 735 (w), 712 (w), 688 (w), 666 (w), 659 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 250 (4.62), 272 sh (4.47), 300 (4.42), 338 (4.16), 392 sh (4.07), 429 sh (4.22), 458 (4.24), 511 sh (3.92) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 299 (4.42), 337 sh (4.12), 453 (4.25), 498 sh (4.19), 532 sh (3.97) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.99 (d, 1H, J = 2.0 Hz, H_8), 8.29 (d, 1H, J = 9.7 Hz, H_4), 8.17 (s, 1H, H_2), 8.09 (d, 1H, J = 10.9 Hz, H_6), 7.86 (t, 1H, J = 10.0 Hz, H_7), 7.39 (s, 1H, Bz), 6.03 (s, 2H, NH_2), 3.93 (s, 3H, CO_2Me), 3.34 (t, 1H, J = 6.9 Hz, *i*-Pr), 3.21 (s, 3H, Me), 1.47 (d, 6H, J = 6.9 Hz, *i*-Pr) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.4, 166.4, 164.7, 162.7, 156.1, 148.3, 147.3, 146.0, 143.4, 142.2, 142.0, 140.6, 137.3, 136.6, 131.7, 120.6, 118.6, 114.4, 114.2, 113.5, 113.1, 112.9, 102.1, 80.3, 51.7, 39.6, 24.6, 24.3 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{29}H_{21}N_5O_4^+$ $[M]^+$ 503.1588, found: 503.1618, $C_{29}H_{21}N_5O_4 + Ag^+$ $[M + Ag]^+$ 610.0639, found: 610.0666.

Compound 2b



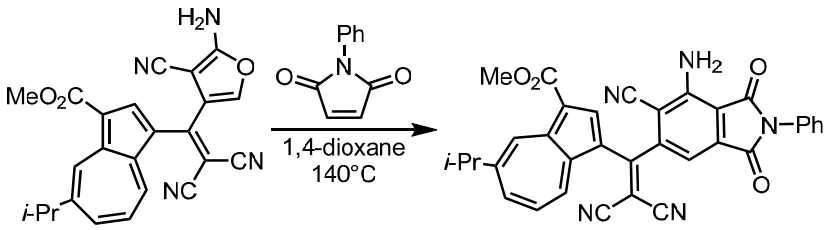
A solution of **1** (803 mg, 1.96 mmol) in *N*-ethylmaleimide (494 mg, 3.95 mmol) in 1,4-dioxane (15 mL) was stirred at 140 °C for 12 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with $CHCl_3$/AcOEt (5 : 1) as an eluent to afford **2b** (691 mg, 1.37 mmol, 70%) as red solid. M.p 235−237 °C; IR (AT-IR): ν_{max} = 3482 (w), 3361 (w), 2980 (w), 2218 (w), 1765 (w), 1703 (s), 1630 (m), 1607 (w), 1531 (w), 1496 (s), 1464 (m), 1442 (s), 1417 (m), 1369 (m), 1339 (m), 1299 (w), 1283 (w), 1246 (m), 1211 (m), 1178 (m), 1134 (w), 1113 (w), 1085 (w), 1067 (m), 1054 (w), 1025 (w), 1001 (w), 970 (w), 931 (w), 906 (w), 878 (w), 821 (m), 801 (w), 778 (w), 758 (w), 745 (w), 712 (w), 685 (w), 664 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 249 (4.66), 273 sh (4.51), 299 (4.46), 337 (4.20), 390 sh (4.10), 428 sh (4.26), 456 (4.29), 513 sh (3.94) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 259 sh (4.58), 300 (4.46), 342 sh (4.16), 454 (4.29), 494 sh (4.23), 530 sh (4.02) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.99 (d, 1H, J = 1.7 Hz, H_8), 8.30 (d, 1H, J = 10.0 Hz, H_4), 8.17-8.08 (m, 2H, $H_{2,6}$), 7.86 (t, 1H, J = 10.2 Hz, H_7), 7.38 (s, 1H, Bz), 6.02 (s, 2H, NH_2), 3.93 (s, 3H, CO_2Me), 3.76 (q, 2H, J = 7.2 Hz, Et), 3.34 (sept, 1H, J = 6.9 Hz, *i*-Pr), 1.47 (d, 6H, J = 6.9 Hz, *i*-Pr), 1.32 (t, 3H, J = 7.2 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.0, 166.1, 164.5, 162.6, 155.7, 148.0, 147.3, 145.7, 143.3, 142.1, 141.8, 140.2, 137.1, 136.4, 131.5, 120.5, 118.3, 114.2, 114.1, 113.5, 113.0, 112.5, 101.6, 80.0, 51.4, 39.2, 33.2, 24.3, 13.7 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{30}H_{23}N_5O_4^+$ $[M]^+$ 517.1745, found: 517.1741, $C_{30}H_{23}N_5O_4$ + Ag^+ $[M + Ag]^+$ 624.0795, found: 624.0768.

Compound 2c



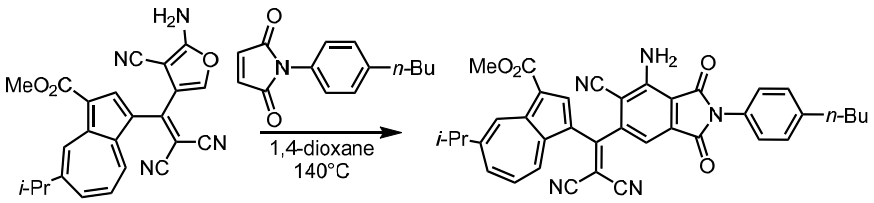
A solution of **1** (21 mg, 0.0511 mmol) in *N*-cyclohexylmaleimide (19 mg, 0.106 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 17.5 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with $CHCl_3$/AcOEt (4 : 1) as an eluent to afford **2c** (21 mg, 0.0367 mmol, 72%) as red solid. M.p. 234−236 °C; IR (AT-IR): ν_{max} = 3452 (w), 3354 (w), 2931 (w), 2853 (w), 2221 (w), 1764 (w), 1698 (s), 1634 (s), 1602 (w), 1550 (w), 1506 (s), 1466 (m), 1442 (s), 1420 (m), 1397 (m), 1374 (s), 1329 (s), 1284 (w), 1247 (m), 1221 (s), 1176 (m), 1136 (w), 1111 (w), 1088 (w), 1058 (w), 1010 (w), 953 (w), 895 (m), 804 (w), 774 (w), 756 (m), 733 (w), 714 (w), 697 (w), 673 (w), 664 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 250 (4.60), 275 sh (4.44), 298 (4.41), 337 (4.15), 390 sh (4.04), 427 sh (4.21), 456 (4.25), 513 sh (3.89) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 264 (4.49), 300 (4.41), 340 (4.10), 455 (4.24), 499 sh (4.16), 538 sh (3.83) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.98 (d, 1H, J = 1.7 Hz, H_8), 8.31 (d, 1H, J = 10.0 Hz, H_4), 8.17 (s, 1H, H_2), 8.09 (d, 1H, J = 100 Hz, H_6), 7.86 (t, 1H, J = 10.0 Hz, H_5), 7.35 (s, 1H, Bz), 6.03 (s, 2H, NH_2), 4.12-4.07 (m, 1H, *c*-Hex), 3.93 (s, 3H, CO_2Me), 3.34 (sept, 1H, J = 6.9 Hz, *i*-Pr), 2.22-2.13 (m, 2H, *c*-Hex), 1.91-1.88 (m, 2H, *c*-Hex), 1.78-1.71 (m, 3H, *c*-Hex), 1.47 (d, 6H, J = 6.9 Hz, *i*-Pr), 1.42-1.25 (m, 3H, *c*-Hex) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.6, 166.3, 164.7, 162.9, 156.0, 148.1, 147.4, 146.0, 143.5, 142.2, 141.9, 140.6, 137.3, 136.4, 131.7, 120.7, 118.6, 114.3, 114.2, 113.6, 113.2, 112.7, 101.8, 80.3, 51.65, 51.61, 39.5, 30.0, 26.0, 25.1, 24.6 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{34}H_{29}N_5O_4^+$ $[M]^+$ 571.2214; found: 571.2216, $C_{34}H_{29}N_5O_4$ + Ag^+ $[M + Ag]^+$ 678.1265; found: 678.1249.

Compound 2d



A solution of **1** (102 mg, 0.248 mmol) in *N*-phenylmaleimide (86 mg, 0.496 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 18 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with $CHCl_3$/AcOEt (4 : 1) as an eluent to afford **2d** (48 mg, 0.0848 mmol, 34%) as red solid. M.p. 190−191 °C; IR (AT-IR): ν_{max} = 3462 (w), 3361 (w), 2962 (w), 2357 (w), 2220 (w), 1770 (w), 1712 (s), 1632 (s), 1599 (m), 1502 (s), 1466 (m), 1442 (s), 1419 (s), 1385 (s), 1343 (m), 1287 (w), 1219 (s), 1178 (m), 1121 (m), 1095 (m), 1065 (w), 1047 (w), 1003 (w), 963 (w), 878 (m), 810 (w), 778 (m), 758 (m), 735 (m), 713 (w), 690 (m), 677 (w), 660 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 248 (4.66), 298 (4.45), 340 (4.14), 394 sh (4.09), 429 sh (4.22), 457 sh (4.23), 514 sh (3.90) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 265 (4.54), 300 (4.46), 341 (4.11), 453 (4.25), 496 sh (4.18), 536 sh (3.91) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.99 (d, 1H, J = 2.0 Hz, H_8), 8.33 (d, 1H, J = 10.0 Hz, H_4), 8.21 (s, 1H, H_2), 8.10 (d, 1H, J = 10.9 Hz, H_6), 7.87 (t, 1H, J = 10.2 Hz, H_7), 7.55-7.52 (m, 3H, Ph), 7.47 (s, 1H, Bz), 7.44 (d, 2H, J = 8.3 Hz, , Ph), 6.15 (s, 2H, NH_2), 3.93 (s, 3H, CO_2Me), 3.33 (sept, 1H, J = 6.9 Hz, *i*-Pr), 1.47 (d, 6H, J = 6.9 Hz, *i*-Pr) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 167.4, 165.3, 164.7, 162.6, 156.2, 148.8, 147.8, 146.1, 143.4, 142.2, 142.0, 140.7, 137.3, 136.0, 131.8, 130.9, 129.4, 128.7, 126.4, 120.6, 118.7, 114.2, 113.8, 113.6, 113.2, 113.1, 102.3, 80.3, 51.7, 39.6, 24.6 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{34}H_{23}N_5O_4^+$ $[M]^+$ 565.1745; found: 565.1726, $C_{34}H_{23}N_5O_4 + Ag^+$ $[M + Ag]^+$ 672.0795, found: 672.0795.

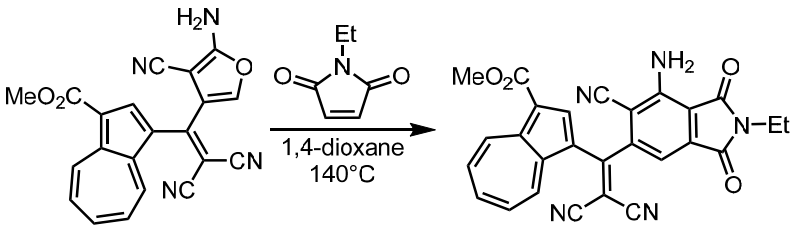
Compound 2e



A solution of **1** (16 mg, 0.0389 mmol) in *N*-*n*-butylmaleimide (18 mg, 0.0785 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 18 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with hexane/AcOEt (4 : 1) as an eluent to afford **2e** (14 mg, 0.0225 mmol, 58%) as red solid. M.p. 192 − 193 °C; IR (AT-IR): ν_{max} = 3468 (w), 3357 (w), 2962 (w), 2218 (m), 1772 (w), 1708 (s), 1626 (m), 1604 (m), 1515 (m), 1486 (m), 1464 (m), 1443 (s), 1419 (s), 1394 (s), 1370 (s), 1337 (m), 1310 (w), 1247 (m),1226 (s), 1210 (m), 1174 (m), 1138 (m), 1123 (m), 1087 (m), 1065 (m), 1046 (m), 1003 (w), 967 (w), 930 (w), 900 (m), 887 (m), 868 (w), 841 (w), 805 (m), 794 (m), 777 (m), 768 (m), 748 (m), 734 (m), 715 (w), 695 (w), 664 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 249 (4.71), 271 sh (4.58), 301 (4.52), 342 sh (4.28), 385 sh (4.12), 438 (4.36), 512 sh (3.80) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 264 (4.61), 300 (4.52), 343 sh (4.16), 400 sh (4.12), 454 (4.32), 498 sh (4.24), 535 sh (3.99) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 10.00 (d, 1H, J = 1.7 Hz, H_8), 8.33 (d, 1H, J = 10.0 Hz, H_4), 8.19 (s, 1H, H_2), 8.11 (d, 1H, J = 10.9 Hz, H_6), 7.89 (t, 1H, J = 10.2 Hz, H_7), 7.48 (s, 1H, Bz), 7.31-7.35 (m, 4H, p-*n*-BuPh), 6.14 (s, 2H, NH_2), 3.93 (s, 3H, CO_2Me), 3.34 (sept, 1H, J = 6.9 Hz, *i*-Pr), 2.68 (t, 2H, J = 7.7 Hz, *n*-Bu), 1.67-1.61 (m, 2H, *n*-Bu), 1.47 (d, 6H, J = 6.9 Hz, *i*-Pr), 1.39 (m, 2H, *n*-Bu), 0.95 (t, 3H, J = 7.3 Hz, *n*-Bu) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 167.5, 165.4, 164.7, 162.6, 156.2, 148.6, 147.7, 146.0, 143.8, 143.4, 142.2, 142.1, 140.6, 137.3, 136.1, 131.8, 129.4, 128.2, 126.2, 120.6, 118.6, 114.2, 113.9, 113.6, 113.1, 102.2, 80.1, 51.7, 39.5, 35.5, 33.5, 24.6, 22.4, 14.1 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{38}H_{31}N_5O_4^+$ $[M]^+$ 621.2371; found: 621.2373, $C_{38}H_{31}N_5O_4$ + Ag^+ $[M + Ag]^+$ 728.1421; found:

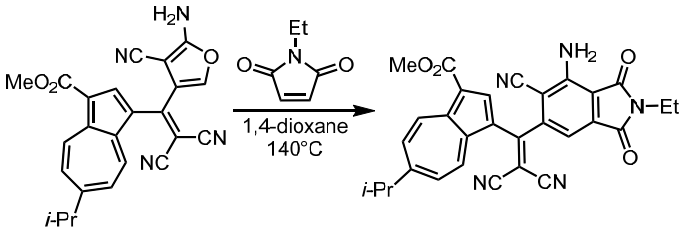
728.1430.

Compound 4a



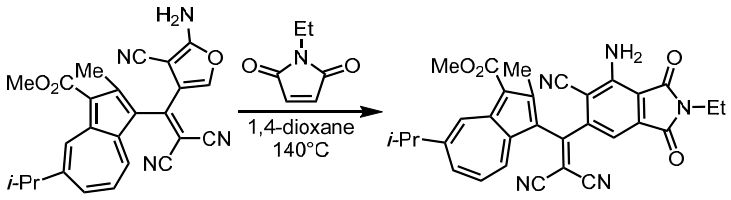
A solution of **3a** (154 mg, 0.418 mmol) in *N*-ethylmaleimide (105 mg, 0.839 mmol) in 1,4-dioxane (10 mL) was stirred at 140 °C for 16.5 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with $CHCl_3$/AcOEt (4 : 1) as an eluent to afford **4a** (109 mg, 0.229 mmol, 55%) as red solid. M.p. 186−189 °C; IR (AT-IR): ν_{max} = 3566 (w), 3446 (w), 3354 (w), 2951 (w), 2223 (w), 1763 (w), 1697 (s), 1629 (s), 1602 (w), 1536 (w), 1507 (m), 1444 (m), 1422 (m), 1401 (s), 1372 (m), 1336 (m), 1248 (w), 1209 (s), 1183 (m), 1158 (w), 1065 (m), 1042 (w), 1020 (m), 907 (m), 875 (w), 814 (w), 780 (m), 755 (m), 708 (w), 689 (w), 676 (w), 668 (w), 657 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 248 (4.66), 298 (4.41), 333 (4.14), 438 (4.22), 485 sh (3.91) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 300 (4.41), 335 (4.11), 447 (4.24), 495 sh (3.99) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.91 (d, 1H, J = 10.0 Hz, H_8), 8.43 (d, 1H, J = 9.7 Hz, H_4), 8.20 (s, 1H, H_2), 8.16 (t, 1H, J = 9.7 Hz, H_6), 7.97 (t, 1H, J = 9.9 Hz, H_7), 7.91 (t, 1H, J = 9.7 Hz, H_5), 7.39 (s, 1H, Bz), 6.04 (s, 2H, NH_2), 3.93 (s, 3H, CO_2Me), 3.76 (q, 2H, J = 7.2 Hz, Et), 1.31 (t, 3H, J = 7.2 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.2, 166.1, 164.5, 163.1, 148.0, 147.3, 145.8, 143.2, 142.8, 142.1, 140.9, 138.8, 136.6, 133.3, 131.8, 121.6, 119.4, 114.5, 113.9, 113.3, 113.1, 112.7, 101.8, 81.7, 51.8, 33.5, 14.0 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{27}H_{17}N_5O_4^+$ $[M]^+$ 475.1275; found: 475.1268, $C_{27}H_{17}N_5O_4 + Ag^+$ $[M + Ag]^+$ 582.0326; found: 582.0351.

Compound 4b



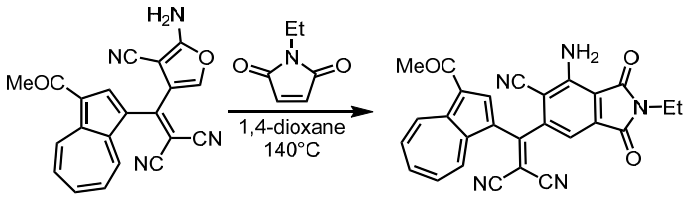
A solution of **3b** (298 mg, 0.726 mmol) in *N*-ethylmaleimide (185 mg, 1.47 mmol) in 1,4-dioxane (15 mL) was stirred at 140 °C for 18 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with toluene/AcOEt (4 : 1) as an eluent to afford **4b** (311 mg, 0.600 mmol, 83%) as red solid. M.p. 179−181 °C; IR (AT-IR): ν_{max} = 3491 (w), 3365 (w), 2960 (w), 2223 (w), 1763 (w), 1706 (s), 1628 (m), 1601 (w), 1585 (w), 1540 (w), 1506 (m), 1445 (m), 1424 (m), 1401 (m), 1374 (m), 1332 (m), 1242 (w), 1209 (s), 1134 (w), 1082 (w), 1065 (w), 1053 (m), 1035 (w), 1019 (w), 903 (w), 855 (w), 815 (w), 790 (w), 775 (w), 757 (w), 734 (w), 703 (w), 692 (w), 682 (w), 664 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 249 (4.57), 275 sh (4.41), 306 (4.51), 338 (4.17), 440 (4.23), 496 sh (3.95) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 308 (4.52), 342 sh (4.13), 450 (4.23), 486 sh (4.16), 514 sh (4.02) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.78 (d, 1H, J = 10.6 Hz, H_8), 8.32 (d, 1H, J = 10.3 Hz, H_4), 8.13 (s, 1H, H_2), 7.87 (dd, 1H, J = 10.6, 1.4 Hz, H_7), 7.80 (dd, 1H, J = 10.6, 1.7 Hz, H_5), 7.36 (s, 1H, Bz), 6.08 (s, 2H, NH_2), 3.90 (s, 3H, CO_2Me), 3.73 (q, 2H, J = 7.2 Hz, Et), 3.25 (sept, 1H, J = 6.9 Hz, *i*-Pr), 1.41 (d, 6H, J = 6.9 Hz, *i*-Pr), 1.29 (t, 3H, J = 7.2 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.2, 166.6, 166.1, 164.5, 162.9, 148.1, 147.3, 144.6, 142.1, 140.9, 140.3, 138.4, 136.5, 132.7, 131.3, 121.3, 119.1, 114.2, 114.1, 113.5, 113.1, 112.6, 101.7, 80.4, 51.6, 40.0, 33.4, 24.2, 13.9 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{30}H_{23}N_5O_4^+$ $[M]^+$ 517.1745, found: 517.1760, $C_{30}H_{23}N_5O_4 + Ag^+$ $[M + Ag]^+$ 624.0795, found: 624.0791.

Compound 4c



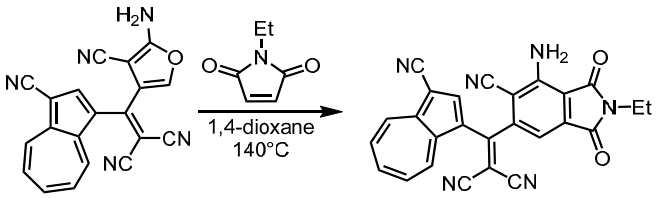
A solution of **3c** (308 mg, 0.725 mmol) in *N*-ethylmaleimide (183 mg, 1.46 mmol) in 1,4-dioxane (10 mL) was stirred at 140 °C for 16.5 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with hexane/AcOEt (4 : 1) as an eluent to afford **4c** (366 mg, 0.688 mmol, 95%) as red solid. M.p. 224 − 226 °C; IR (AT-IR): ν_{max} = 3455 (w), 3336 (w), 2959 (w), 2230 (w), 1762 (w), 1698 (s), 1684 (s), 1627 (m), 1598 (w), 1535 (w), 1493 (m), 1435 (s), 1422 (s), 1402 (m), 1381 (m), 1337 (m), 1283 (w), 1248 (w), 1226 (m), 1200 (m), 1144 (w), 1086 (m), 1065 (w), 1047 (w), 1030 (w), 994 (w), 973 (w), 955 (w), 903 (w), 876 (w), 851 (w), 809 (w), 779 (w), 760 (m), 741 (w), 727 (w), 708 (w), 688 (w), 670 (w), 660 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 252 (4.60), 275 sh (4.50), 306 (4.55), 383 (4.00), 431 (4.05), 489 (3.97), 537 sh (3.73) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 306 (4.55), 368 sh (4.02), 446 (4.04), 501 (3.99), 535 sh (3.88) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 9.79 (s, 1H, H_8), 8.02 (d, 1H, J = 10.0 Hz, H_4), 7.91 (d, 1H, J = 10.3 Hz, H_6), 7.67 (t, 1H, J = 10.0 Hz, H_7), 7.38 (s, 1H, Bz), 6.03 (s, 2H, NH_2), 3.98 (s, 3H, CO_2Me), 3.73 (q, 2H, J = 7.2 Hz, Et), 3.27 (sept, 1H, J = 6.9 Hz, *i*-Pr), 2.58 (s, 3H), 1.43 (d, 6H, J = 6.9 Hz, *i*-Pr), 1.29 (t, 3H, J = 7.2 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.1, 166.1, 165.9, 163.6, 154.9, 154.5, 147.8, 147.4, 145.0, 141.9, 139.9, 138.7, 136.2, 134.3, 131.1, 122.2, 117.7, 114.3, 113.7, 113.4, 113.2, 113.0, 101.9, 85.3, 51.5, 39.6, 33.5, 24.6, 17.5, 13.9 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{31}H_{25}N_5O_4^+$ $[M]^+$ 531.1901, found: 531.1901, $C_{31}H_{25}N_5O_4 + Ag^+$ $[M + Ag]^+$ 638.0952, found: 638.0960.

Compound 4d



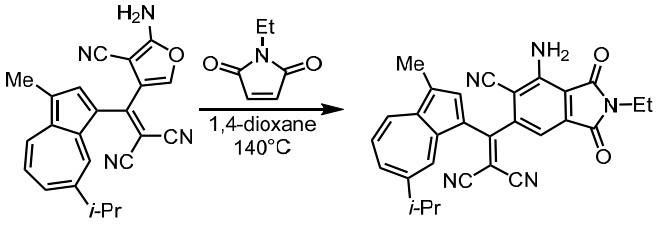
A solution of **3d** (129 mg, 0.366 mmol) in *N*-ethylmaleimide (92 mg, 0.735 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 15.5 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with hexane/AcOEt (4 : 1) as an eluent to afford **4d** (90 mg, 0.194 mmol, 54%) as red solid. M.p. 186−189 °C; IR (AT-IR): ν_{max} = 3455 (w), 3336 (w), 2959 (w), 2230 (w), 1762 (w), 1698 (s), 1684 (s), 1627 (m), 1598 (w), 1535 (w), 1493 (m), 1435 (s), 1422 (s), 1402 (m), 1381 (m), 1337 (m), 1283 (w), 1248 (w), 1226 (m), 1200 (m), 1144 (w), 1086 (m), 1065 (w), 1047 (w), 1030 (w), 994 (w), 973 (w), 955 (w), 903 (w), 876 (w), 851 (w), 809 (w), 779 (w), 760 (m), 741 (w), 727 (w), 708 (w), 688 (w), 670 (w), 660 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 249 (4.51), 297 (4.30), 340 (3.99), 392 sh (3.93), 431 sh (4.06), 452 (4.08), 509 sh (3.78) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 258 (4.50), 310 (4.38), 348 sh (4.10), 382 (4.05), 446 (4.26) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 10.08 (d, 1H, J = 10.0 Hz, H_8), 8.27-8.31 (m, 2H, $H_{2,4}$), 8.10 (t, 1H, J = 9.6 Hz, H_6), 7.94 (t, 1H, J = 9.9 Hz, H_7), 7.82 (t, 1H, J = 9.7 Hz, H_4), 7.39 (s, 1H, Bz), 6.10 (s, 2H, NH_2), 3.73 (q, 2H, J = 7.1 Hz, Et), 2.64 (s, 3H, COMe), 1.29 (t, 3H, J = 7.0 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 195.2, 168.1, 166.1, 162.9, 147.7, 147.4, 145.2, 143.1, 142.7, 142.6, 138.6, 136.7, 134.5, 132.2, 126.7, 121.3, 114.6, 114.0, 113.2, 113.1, 112.6, 101.7, 81.9, 33.5, 29.3, 13.9 ppm, one signal is overlapped with other signal; HRMS (MALDI-TOF, positive): calcd for $C_{27}H_{17}N_5O_3$ + H^+ $[M + H]^+$ 460.1404, found: 460.1398, $C_{27}H_{17}N_5O_3$ + Ag^+ $[M + Ag]^+$ 566.0377, found: 566.0363.

Compound 4e



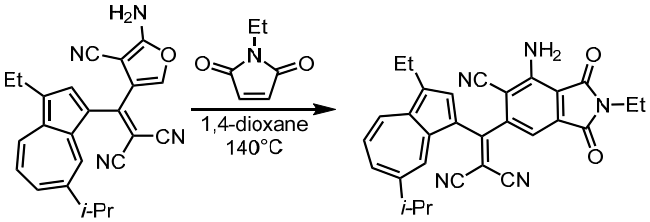
A solution of **3e** (128 mg, 0.381 mmol) in *N*-ethylmaleimide (96 mg, 0.767 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 16.5 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with hexane/AcOEt (4 : 1) as an eluent to afford **4e** (89 mg, 0.201 mmol, 53%) as red solid. M.p. 268−269 °C; IR (AT-IR): ν_{max} = 3425 (w), 3332 (w), 3257 (w), 3222 (w), 3093 (w), 2979 (w), 2221 (w), 2211 (m), 1764 (w), 1709 (s), 1641 (s), 1601 (w), 1578 (w), 1530 (w), 1506 (m), 1467 (w), 1453 (m), 1432 (m), 1399 (m), 1362 (m), 1328 (s), 1305 (w), 1238 (w), 1206 (w), 1173 (m), 1098 (w), 1065 (m), 1017 (w), 978 (w), 906 (m), 891 (w), 850 (w), 819 (w), 807 (w), 781 (w), 758 (s), 728 (w), 701 (w), 691 (w), 682 (w), 666 (w), 659 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 246 (4.63), 293 (4.44), 326 (4.20), 347 sh (4.07), 435 (4.31), 518 sh (3.32) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 292 (4.45), 327 (4.15), 445 (4.31) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 8.90 (d, 1H, J = 9.7 Hz, H_8), 8.47 (d, 1H, J = 10.0 Hz, H_4), 8.25 (t, 1H, J = 9.9 Hz, H_6), 8.02-7.96 (m, 3H, $H_{2,5,7}$), 7.39 (s, 1H, Bz), 6.06 (s, 2H, NH_2), 3.76 (q, 2H, J = 7.2 Hz, Et), 1.31 (t, 3H, J = 7.2 Hz, Et) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.1, 165.9, 162.3, 147.4, 147.3, 147.2, 143.7, 142.6, 139.8, 139.7, 139.5, 136.8, 132.6, 132.4, 122.3, 115.1, 114.8, 113.5, 113.0, 112.8, 112.5, 101.5, 101.1, 83.5, 33.6, 14.0 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{26}H_{14}N_6O_2$ + H^+ $[M + H]^+$ 443.1251, found: 443.1224, $C_{26}H_{14}N_6O_2$ + Ag^+ $[M + Ag]^+$ 549.0224, found: 549.0206.

Compound 4f



A solution of **3f** (58 mg, 0.158 mmol) in *N*-ethylmaleimide (39 mg, 0.311 mmol) in 1,4-dioxane (5 mL) was stirred at 140 °C for 17 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with toluene/AcOEt (5 : 1) as an eluent to afford **4f** (36 mg, 0.0760 mmol, 48%) as purple solid. m.p. 270−271 °C; IR (AT-IR): ν_{max} = 3440 (w), 3327 (w), 3224 (w), 2967 (w), 2934 (w), 2214 (m), 1765 (w), 1706 (s), 1642 (s), 1602 (w), 1509 (w), 1490 (s), 1465 (m), 1432 (s), 1409 (s), 1380 (s), 1355 (m), 1328 (m), 1303 (m), 1294 (m), 1268 (w), 1247 (w), 1205 (m), 1172 (w), 1140 (w), 1097 (w), 1070 (m), 1035 (w), 954 (w), 930 (w), 906 (m), 871 (m), 806 (w), 794 (w), 779 (w), 757 (m), 739 (w), 712 (w), 685 (w), 673 (w), 661 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 247 (4.63), 300 (4.27), 343 (4.05), 345 (4.05), 417 (4.13), 507 (4.17), 557 sh (3.97) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 262 (4.48), 291 (4.42), 340 sh (4.13), 378 sh (4.03), 441 (4.03), 529 (4.03), 560 sh (4.02), 600 sh (3.87) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 8.32 (d, 1H, J = 9.2 Hz, H_8), 8.05 (d, 1H, J = 1.4 Hz, H_4), 7.84 (d, 1H, J = 10.3 Hz, H_6), 7.74 (s, 1H, H_2), 7.59 (t, 1H, J = 10.0 Hz, H_7), 6.02 (s, 2H, NH_2), 3.75 (q, 2H, J = 7.2 Hz, Et), 3.10 (t, 1H, J = 6.9 Hz, *i*-Pr), 2.56 (s, 3H, Me), 1.30 (t, 3H, J = 7.2 Hz, Et), 1.26 (d, 6H, J = 6.9 Hz, *i*-Pr) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.3, 166.2, 160.6, 151.7, 149.5, 147.2, 146.0, 141.2, 1140.34, 140.25, 136.9, 136.3, 135.0, 129.5, 129.4, 121.2, 115.5, 114.8, 113.8, 113.2, 102.5, 75.2, 39.3, 33.4, 24.8, 14.0, 12.7 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{29}H_{23}N_5O_2$ + H^+ $[M + H]^+$ 473.1846, found: 473.1848, $C_{29}H_{23}N_5O_2$ + Ag^+ $[M + Ag]^+$ 580.0897, found: 580.0914.

Compound 4g



A solution of **3g** (298 mg, 0.783 mmol) in *N*-ethylmaleimide (197 mg, 1.57 mmol) in 1,4-dioxane (10 mL) was stirred at 140 °C for 16 h under an Ar atmosphere. After the reaction, solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography with toluene/AcOEt (5 : 1) as an eluent to afford **4g** (200 mg, 0.410 mmol, 52%) as purple solid. M.p. 244 °C; IR (AT-IR): ν_{max} = 3422 (w), 3326 (w), 3218 (w), 2962 (w), 2214 (m), 1765 (w), 1707 (s), 1634 (s), 1603 (w), 1543 (w), 1482 (m), 1459 (m), 1429 (s), 1405 (s), 1362 (s), 1336 (m), 1309 (w), 1253 (w), 1200 (w), 1092 (w), 1066 (m), 1022 (w), 926 (w), 904 (w), 878 (w), 850 (w), 800 (w), 757 (m), 739 (w), 712 (w), 688 (w), 673 (w), 665 (w), 657 (w) cm^{-1}; UV/Vis (CH_2Cl_2): λ_{max} = 246 (4.63), 298 (4.27), 341 (4.05), 345 (4.05), 418 (4.12), 507 (4.15), 574 sh (3.86) nm; UV/Vis (10% CF_3CO_2H/CH_2Cl_2): λ_{max} = 292 (4.48), 346 (4.15), 374 sh (4.09), 440 sh (3.89), 497 (3.98), 561 sh (3.81), 598 sh (3.66) nm; ^{1}H NMR (500 MHz, $CDCl_3$): δ_H = 8.36 (d, 1H, J = 9.7 Hz, H_8), 8.06 (d, 1H, J = 1.4 Hz, H_4), 7.84 (d, 1H, J = 10.3 Hz, H_6), 7.77 (s, 1H, H_2), 7.58 (t, 1H, J = 9.9 Hz, H_7), 7.36 (s, 1H, Bz), 6.03 (s, 2H, NH_2), 3.75 (q, 2H, J = 7.2 Hz, Et), 3.10 (sept, 1H, J = 6.9 Hz, *i*-Pr), 2.98 (q, 2H, J = 7.5 Hz, Et), 1.35 (t, 3H, J = 7.5 Hz, Et), 1.30 (t, 2H, J = 7.2 Hz, Et), 1.26 (d, 5H, J = 6.9 Hz, *i*-Pr) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ_C = 168.3, 166.2, 160.7, 151.7, 149.5, 147.2, 145.1, 141.3, 140.4, 138.6, 136.9, 136.3, 136.0, 134.7, 129.4, 121.3, 115.5, 114.8, 113.8, 113.1, 102.5, 75.3, 39.3, 33.4, 24.7, 20.2, 14.7, 14.0 ppm; HRMS (MALDI-TOF, positive): calcd for $C_{30}H_{25}N_5O_2^+$ $[M]^+$ 487.2003, found: 487.2004, $C_{30}H_{25}N_5O_2$ + Ag^+ $[M + Ag]^+$ 594.1054, found: 594.1081.

1. Copies of ^{1}H NMR, ^{13}C NMR, COSY and HRMS of reported compounds (Figures S1−S48).

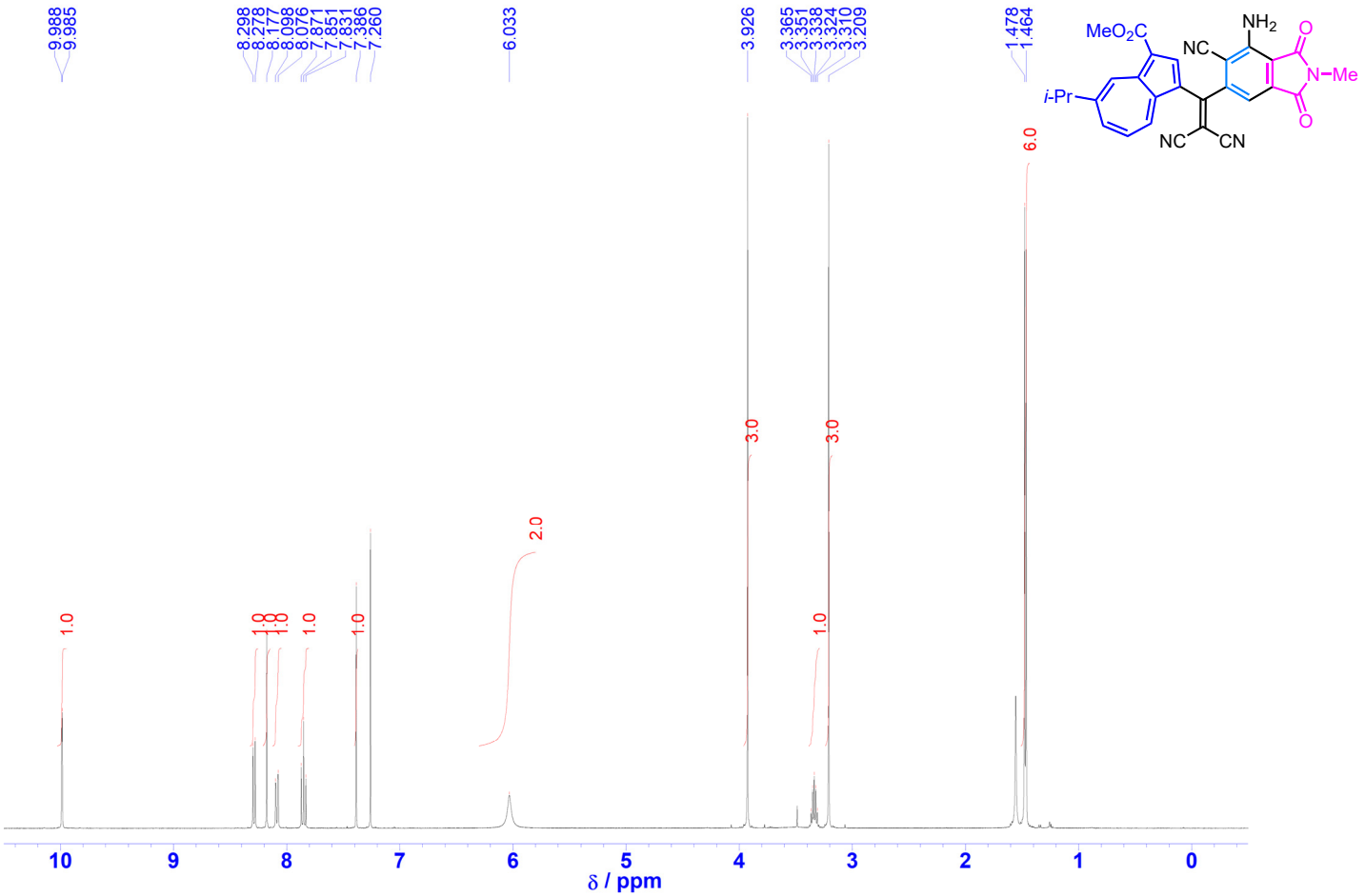


Figure S1. ^{1}H NMR spectrum of **2a** in $CDCl_3$ (500 MHz).

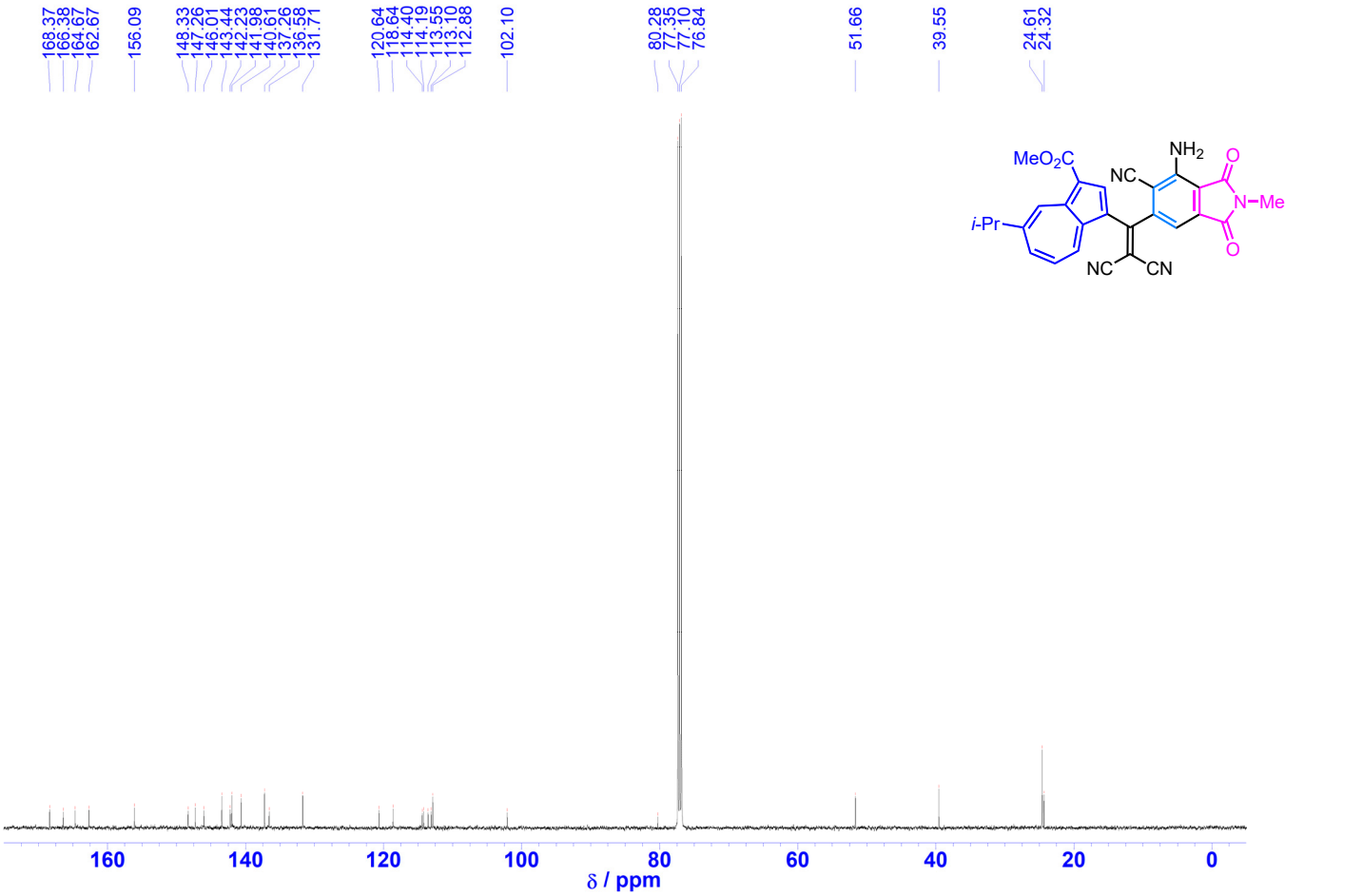


Figure S2. ^{13}C NMR spectrum of **2a** in $CDCl_3$ (125 MHz).

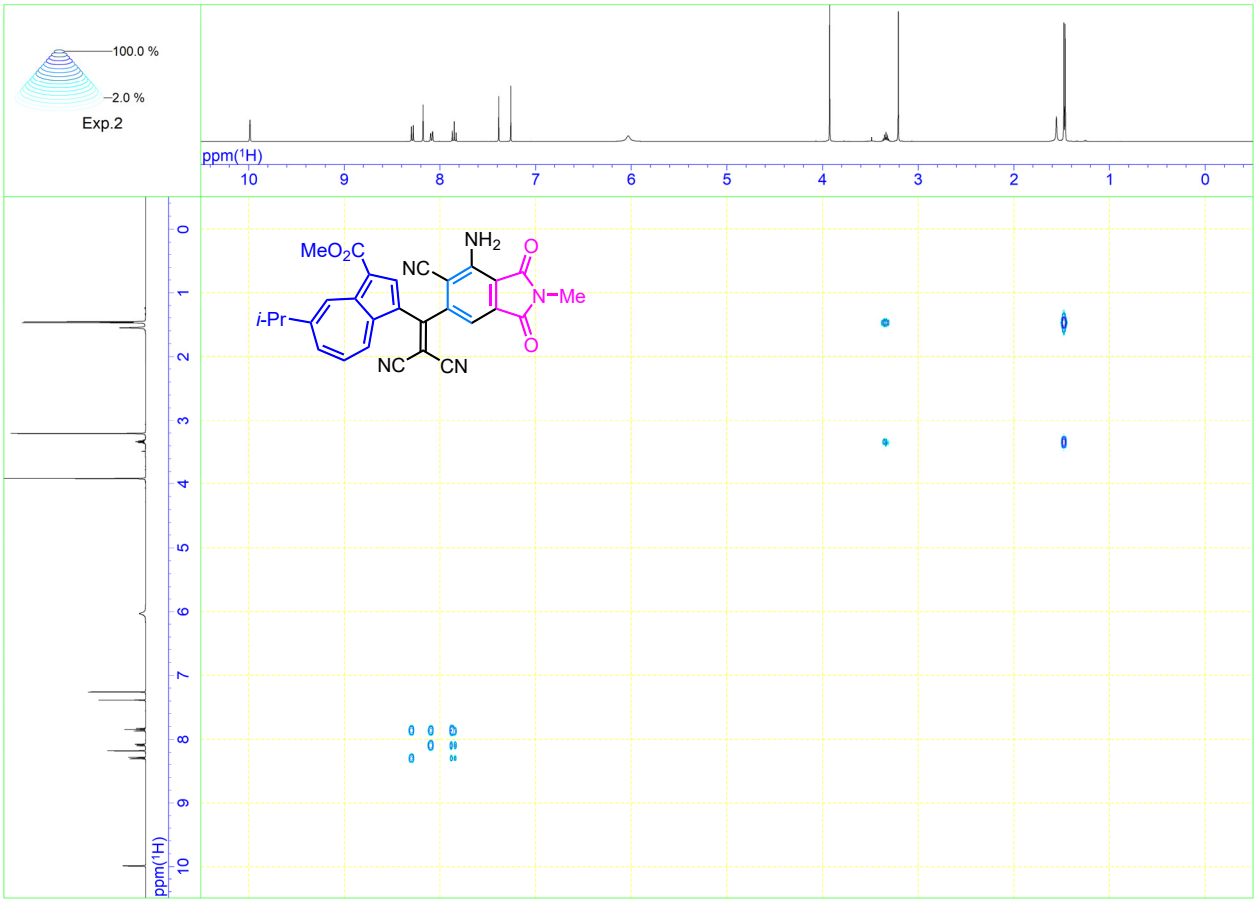


Figure S3. COSY spectrum of **2a** in $CDCl_3$ (500 MHz).

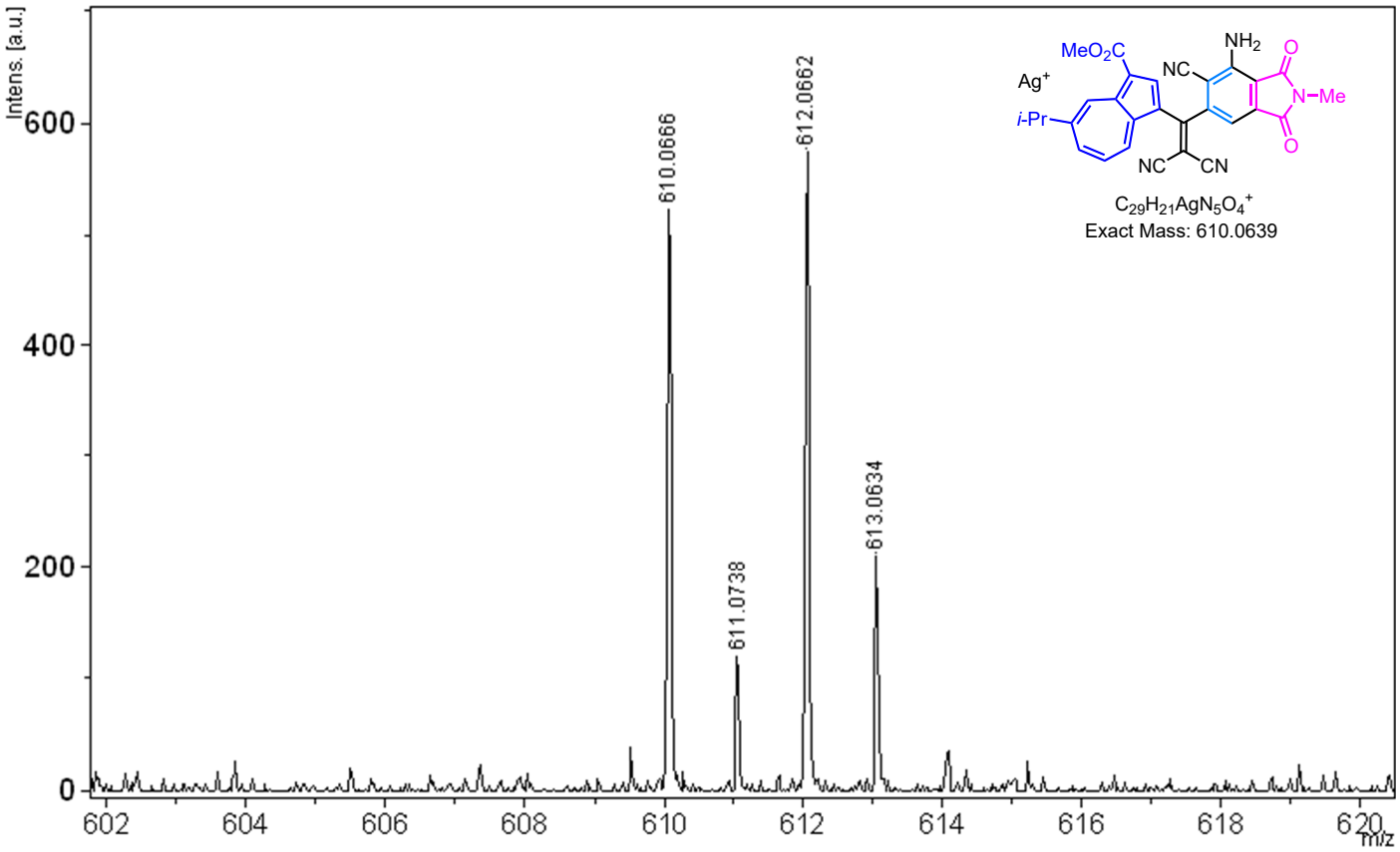


Figure S4. HRMS (MALDI-TOF, positive) of **2a**.

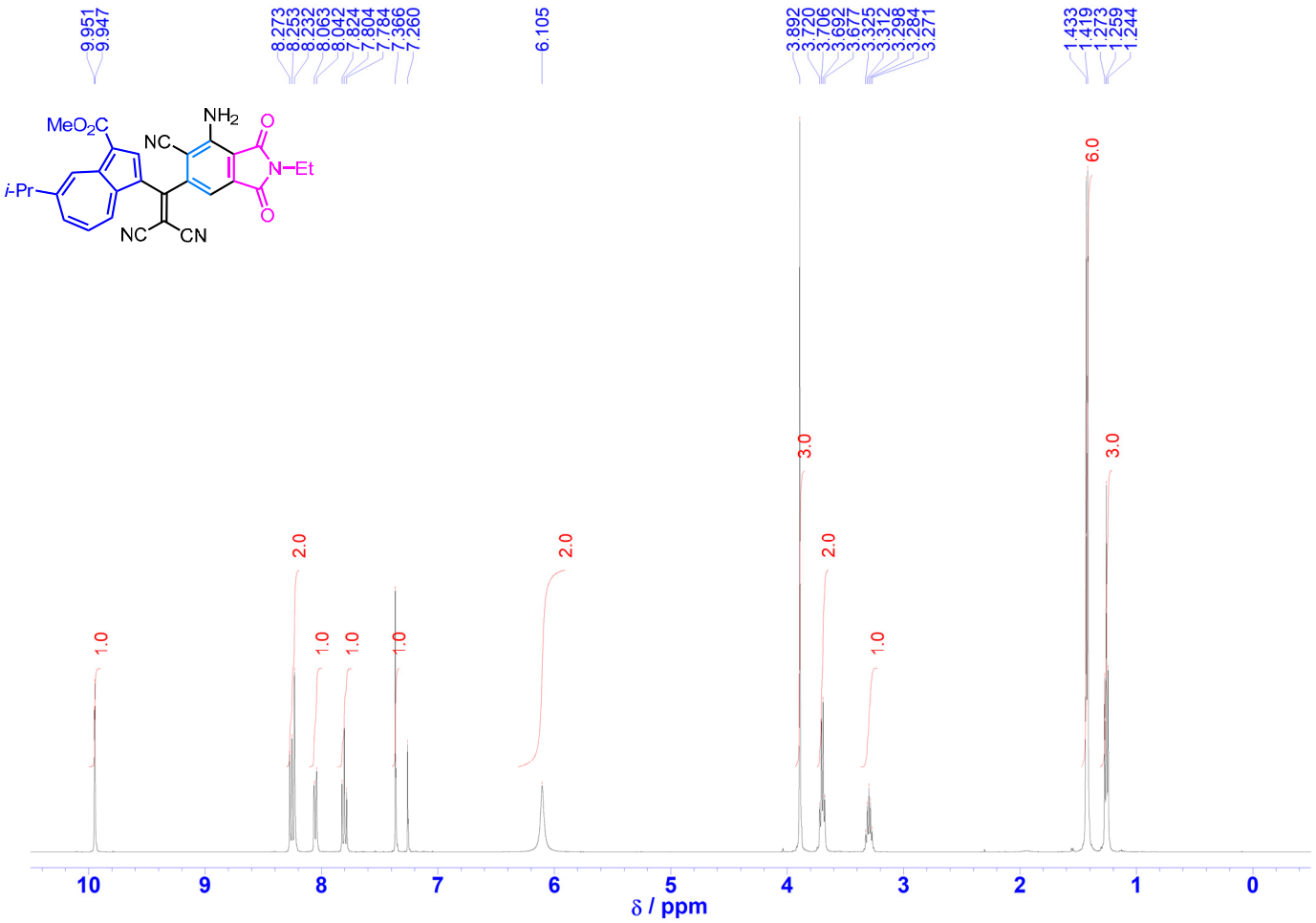


Figure S5. ^{1}H NMR spectrum of **2b** in $CDCl_3$ (500 MHz).

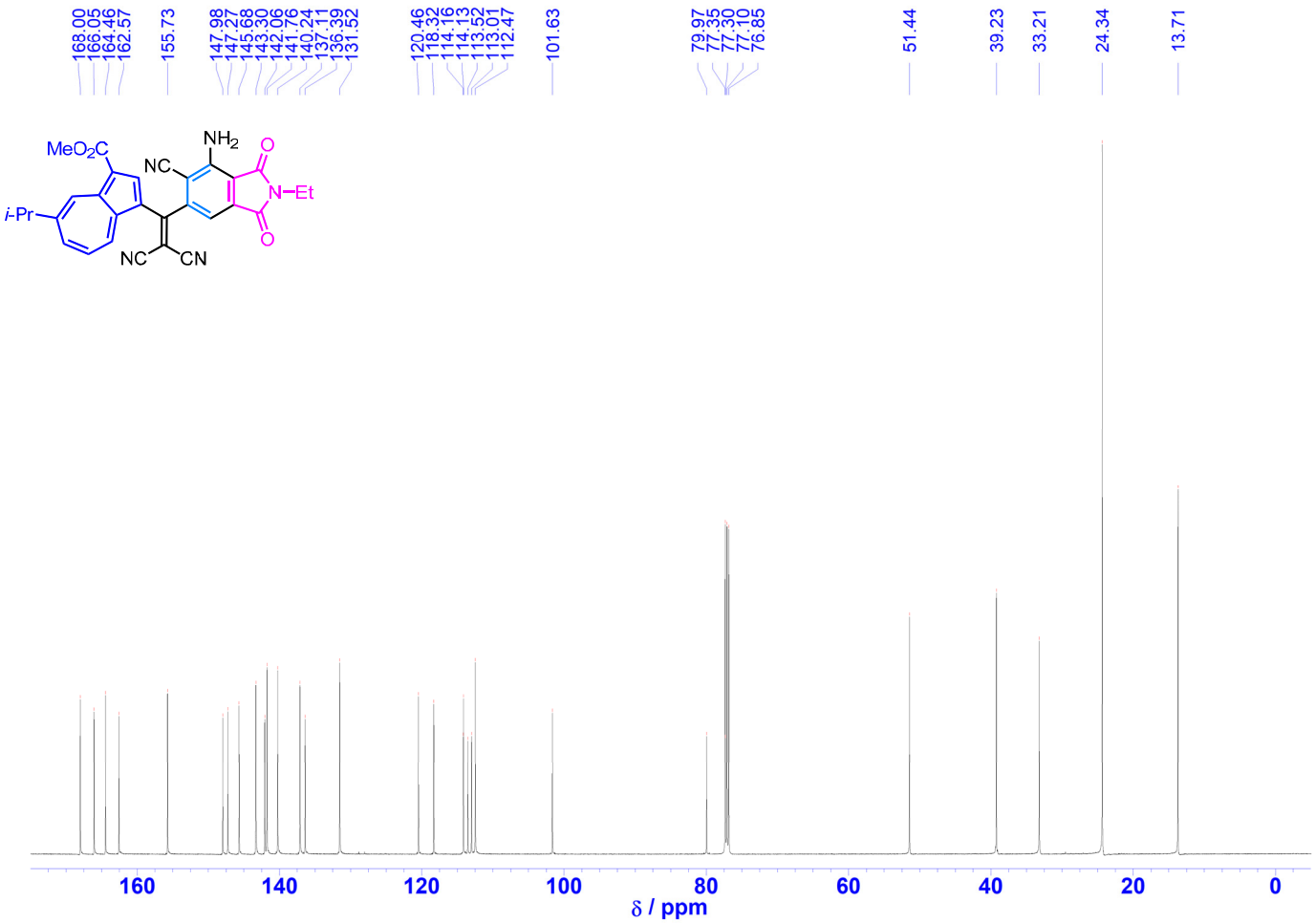


Figure S6. ^{13}C NMR spectrum of **2b** in $CDCl_3$ (125 MHz).

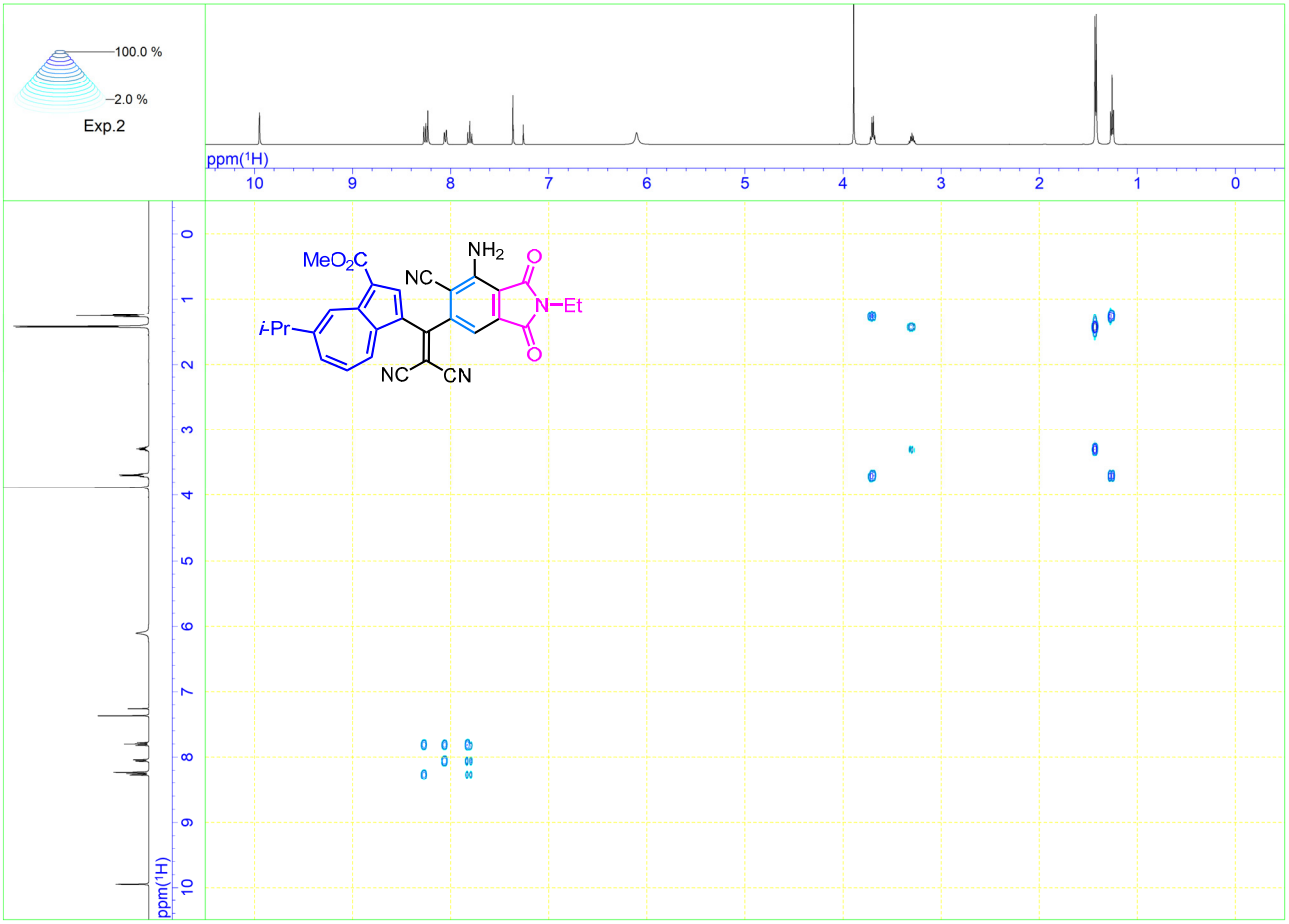


Figure S7. COSY spectrum of **2b** in $CDCl_3$ (500 MHz).

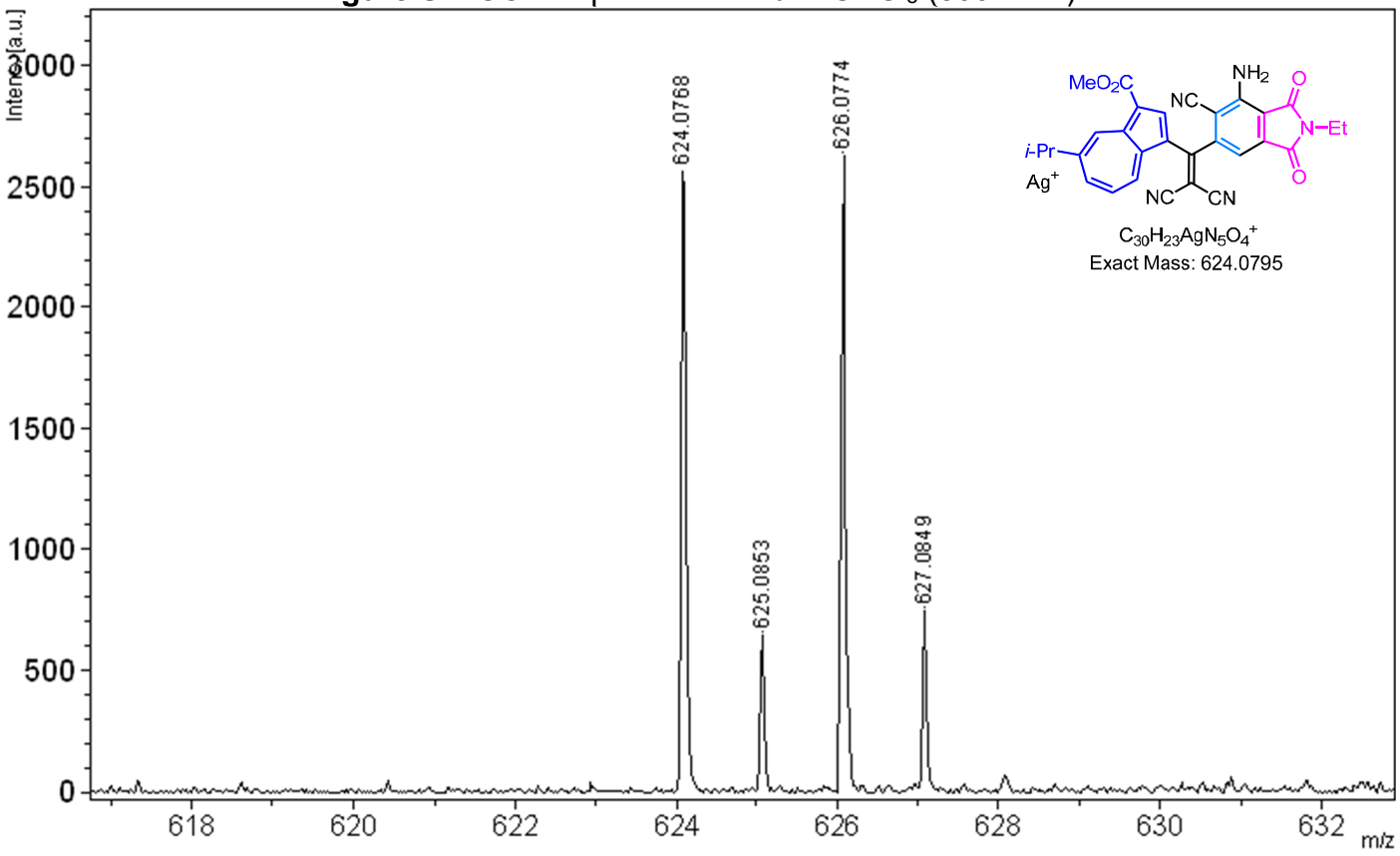


Figure S8. HRMS (MALDI-TOF, positive) of **2b**.

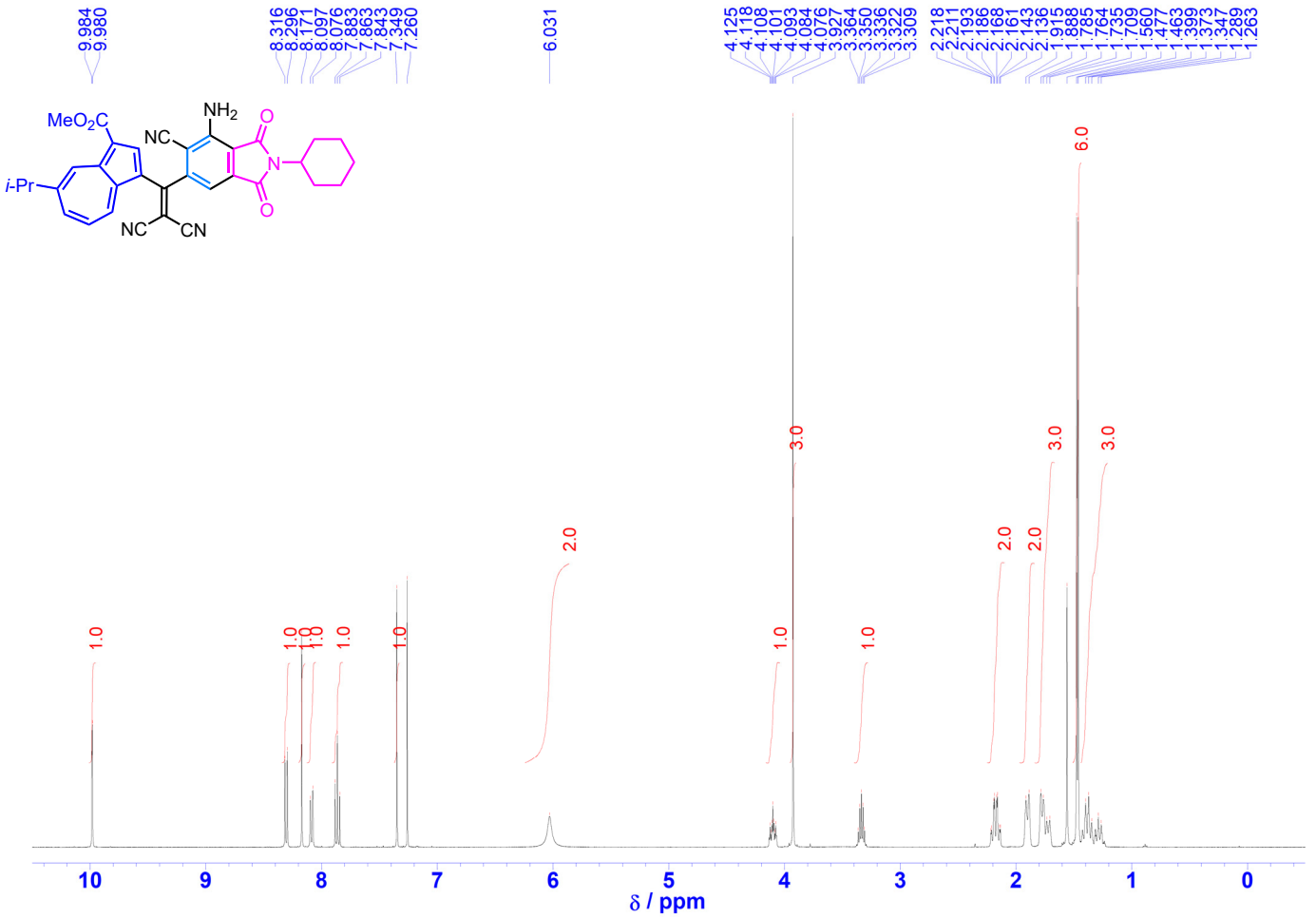


Figure S9. ^{1}H NMR spectrum of **2c** in $CDCl_3$ (500 MHz).

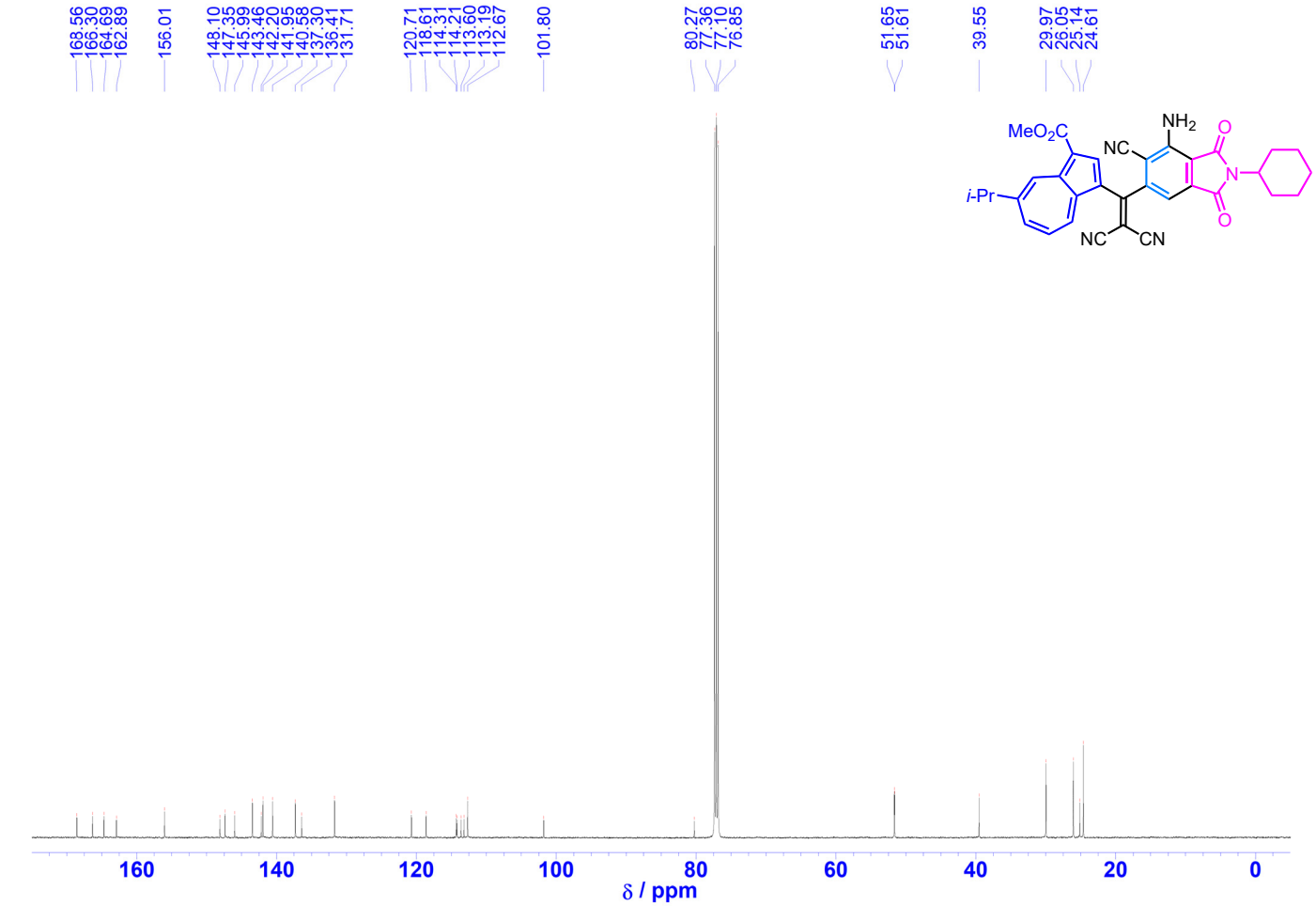


Figure S10. ^{13}C NMR spectrum of **2c** in $CDCl_3$ (125 MHz).

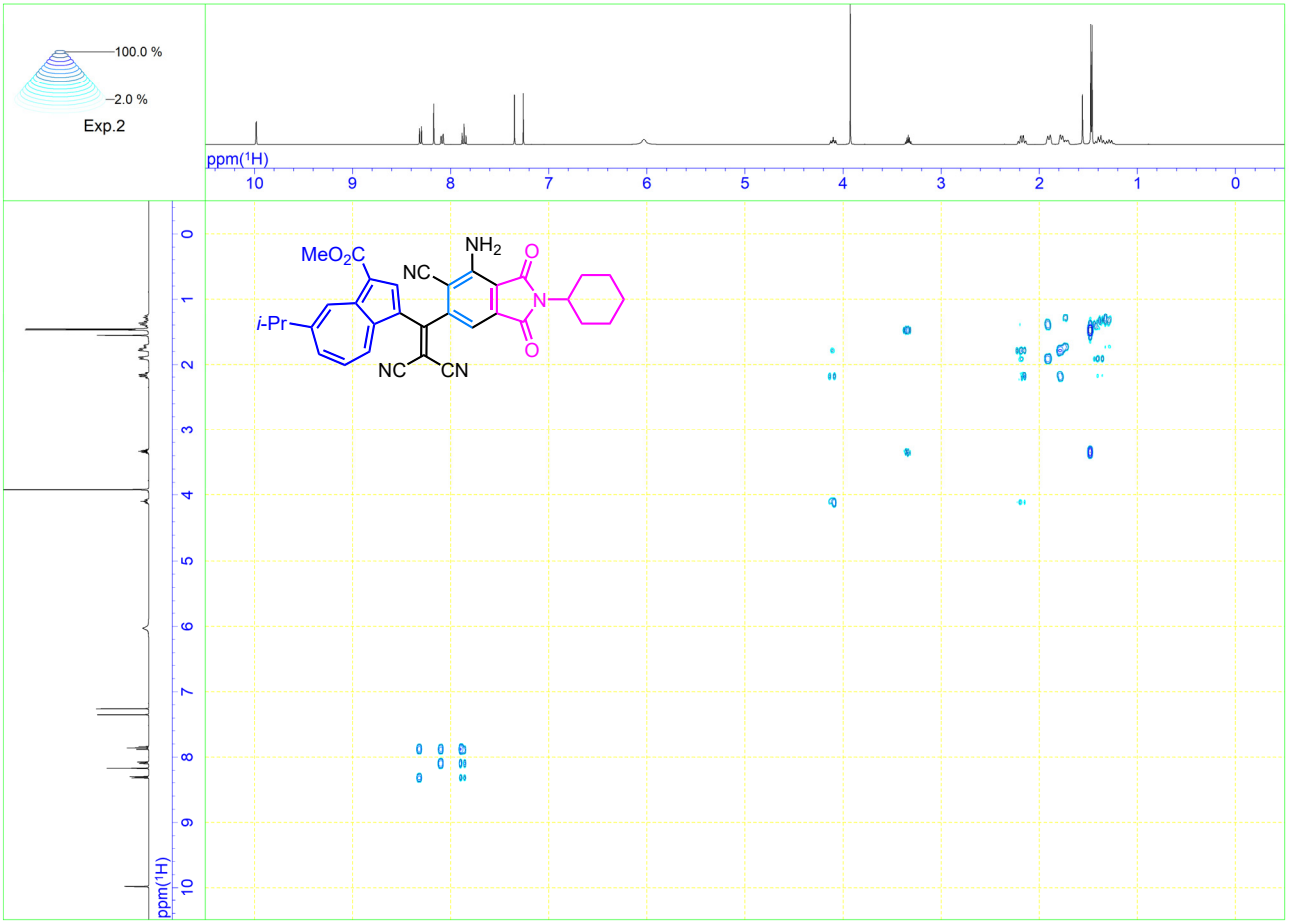


Figure S11. COSY spectrum of **2c** in $CDCl_3$ (500 MHz).

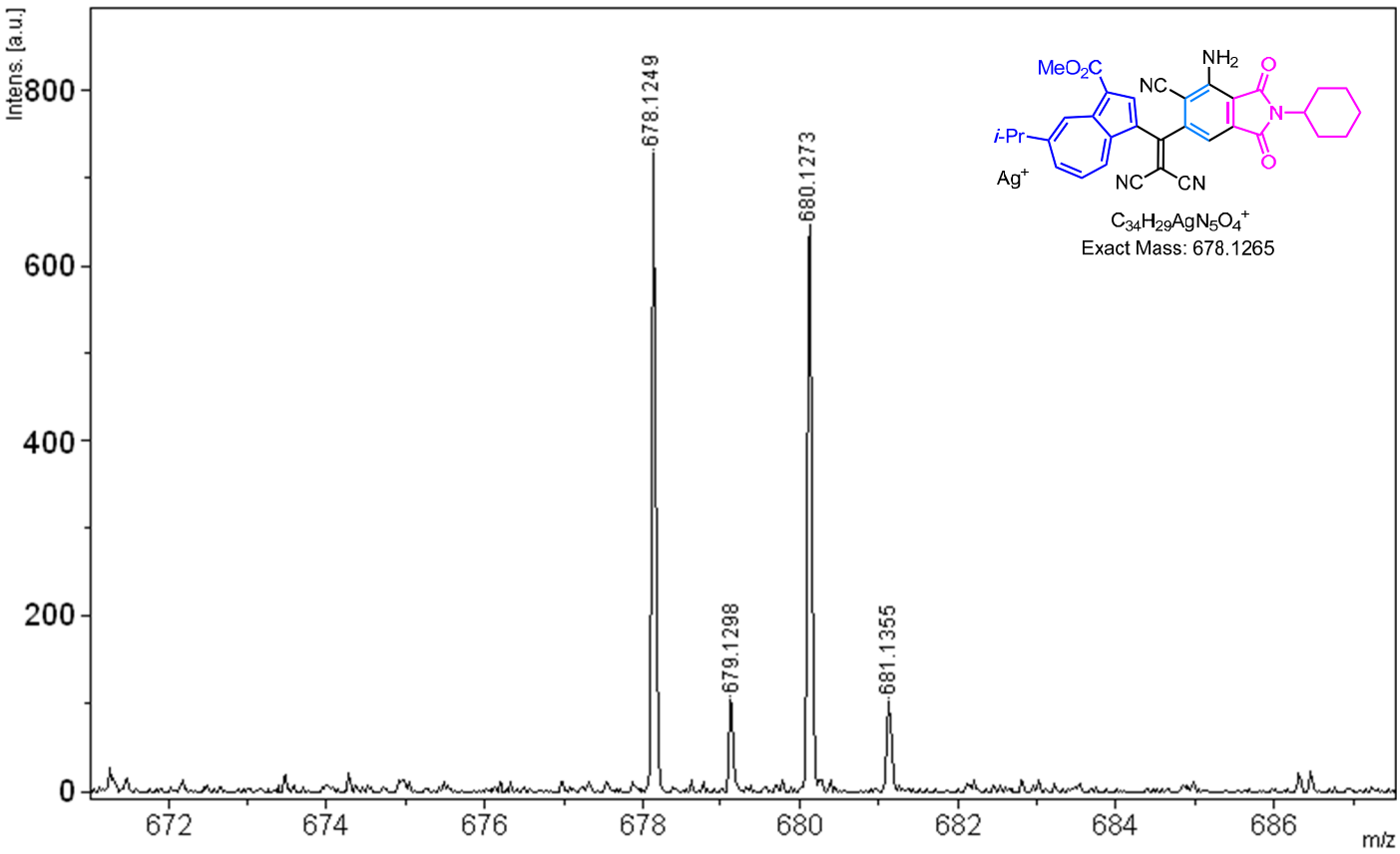


Figure S12. HRMS (MALDI-TOF, positive) of **2c**.

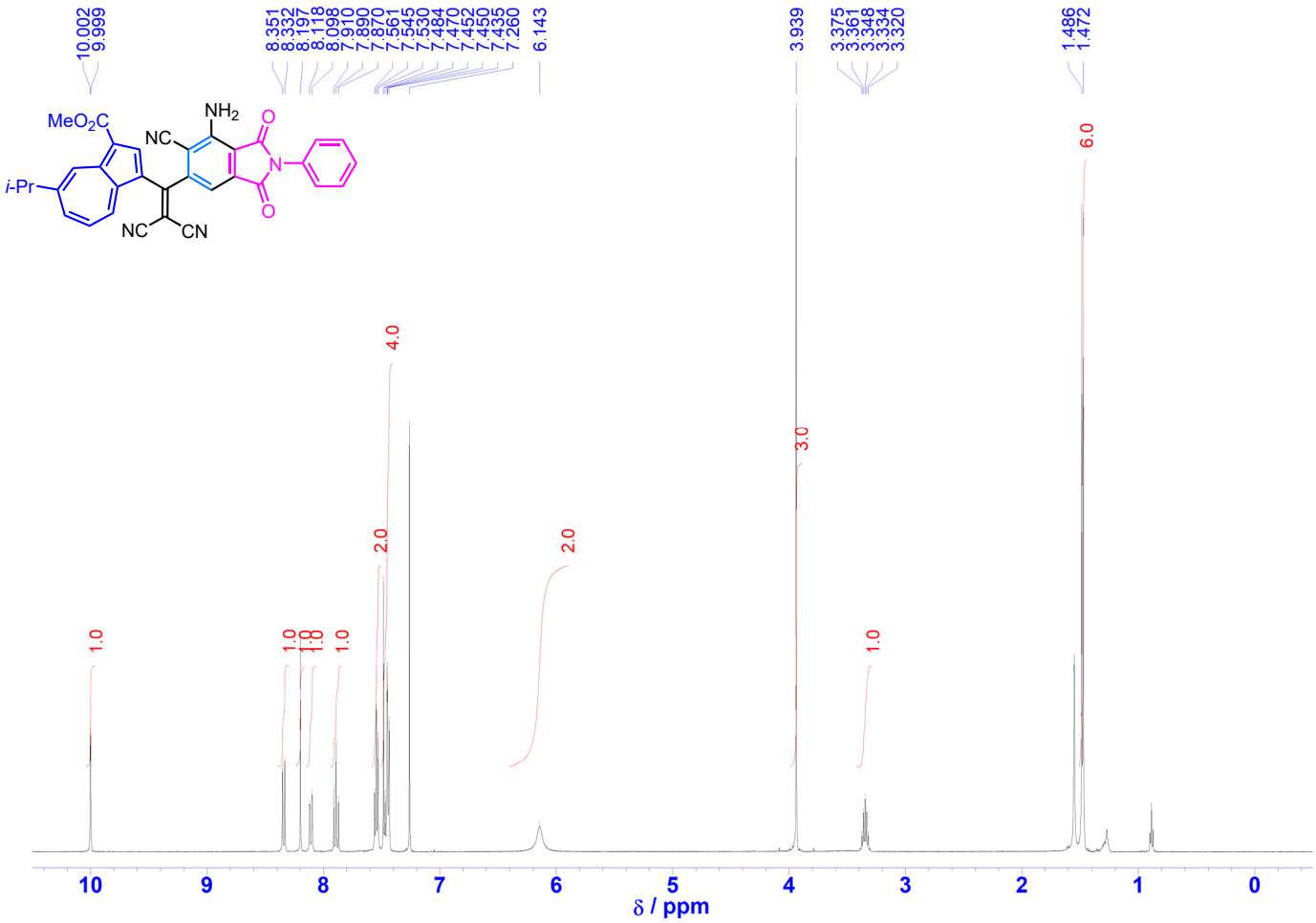


Figure S13. ^{1}H NMR spectrum of **2d** in $CDCl_3$ (500 MHz).

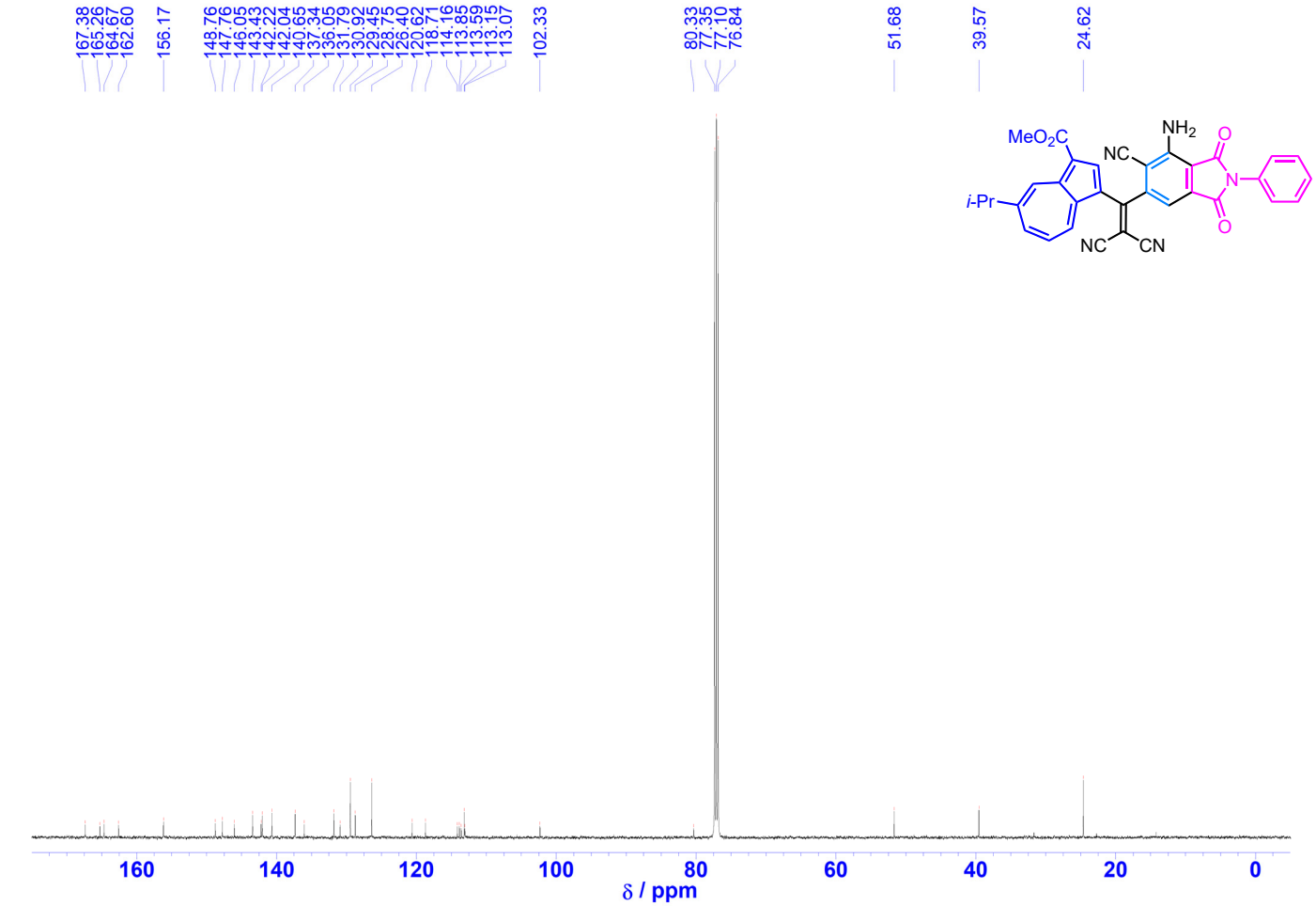


Figure S14. ^{13}C NMR spectrum of **2d** in $CDCl_3$ (125 MHz).

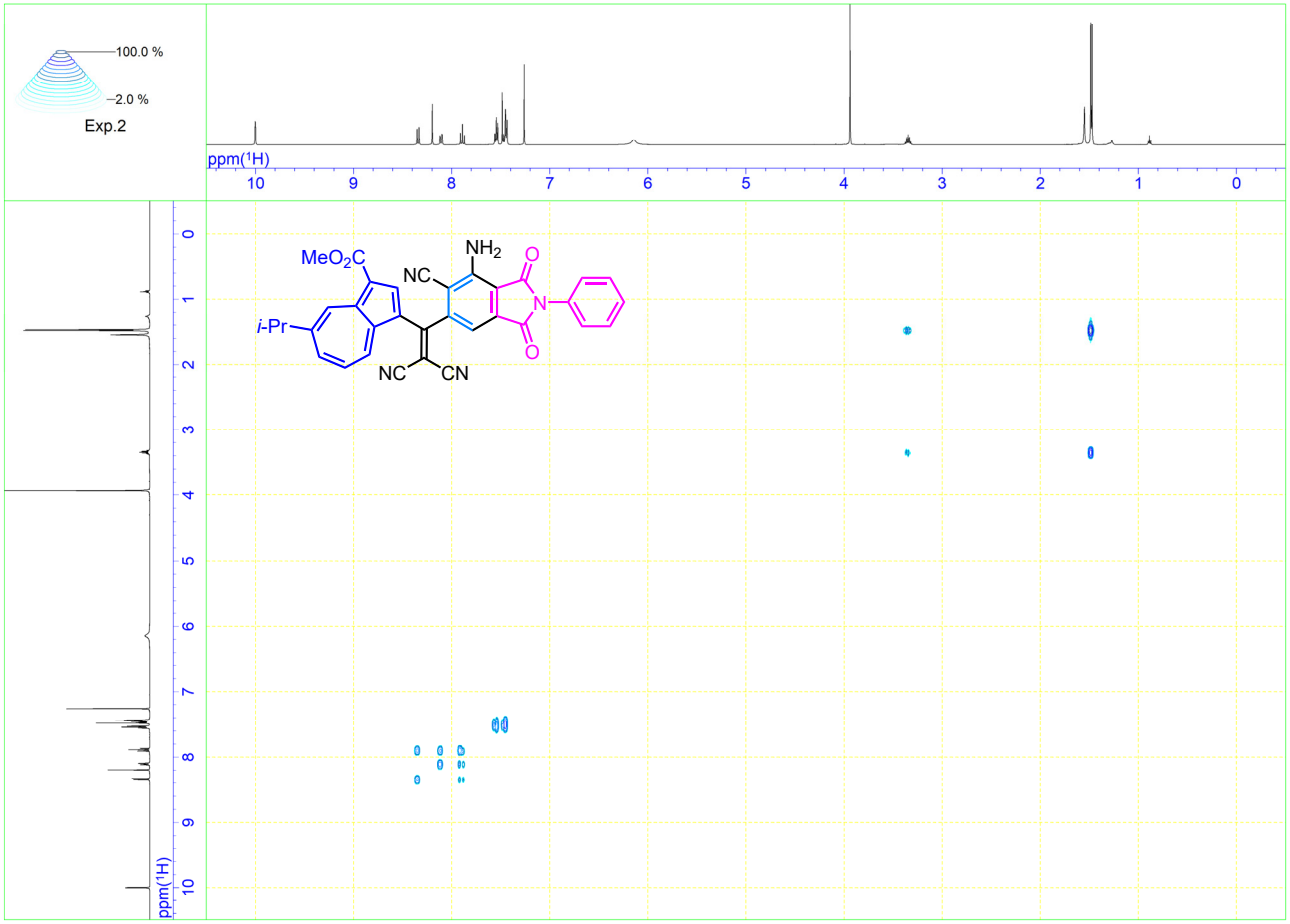


Figure S15. COSY spectrum of **2d** in $CDCl_3$ (500 MHz).

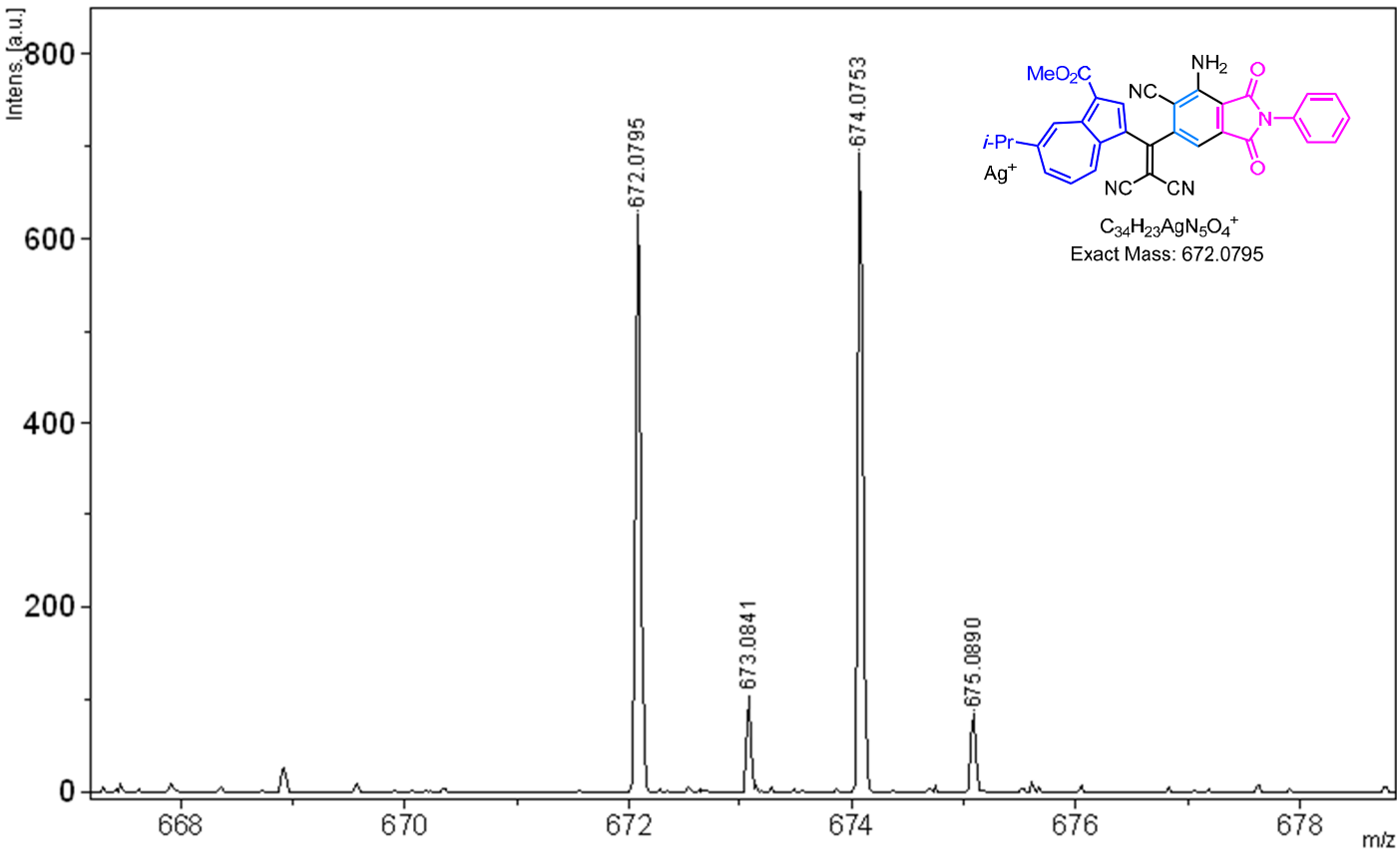


Figure S16. HRMS (MALDI-TOF, positive) of **2d**.

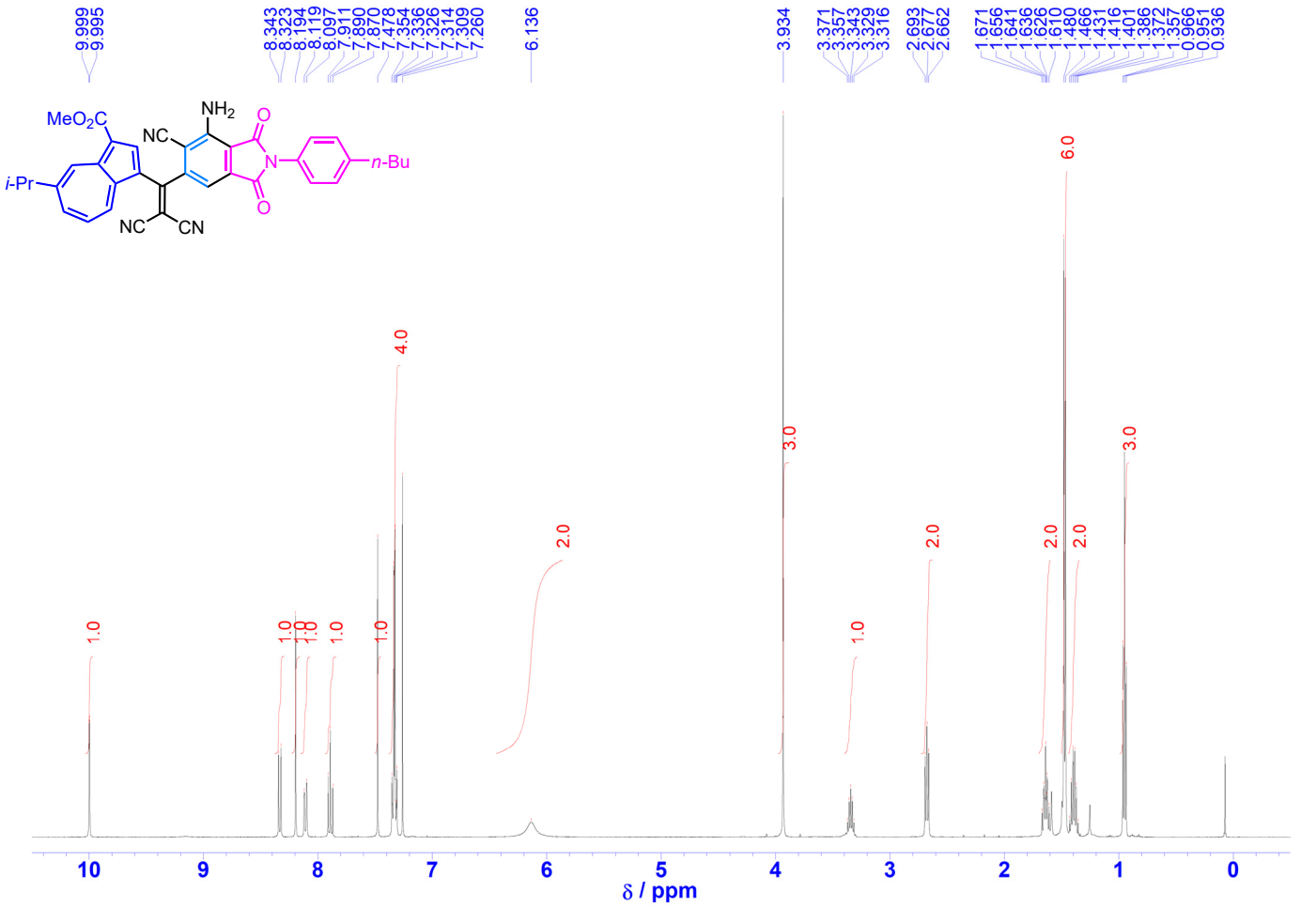


Figure S17. ^{1}H NMR spectrum of **2e** in $CDCl_3$ (500 MHz).

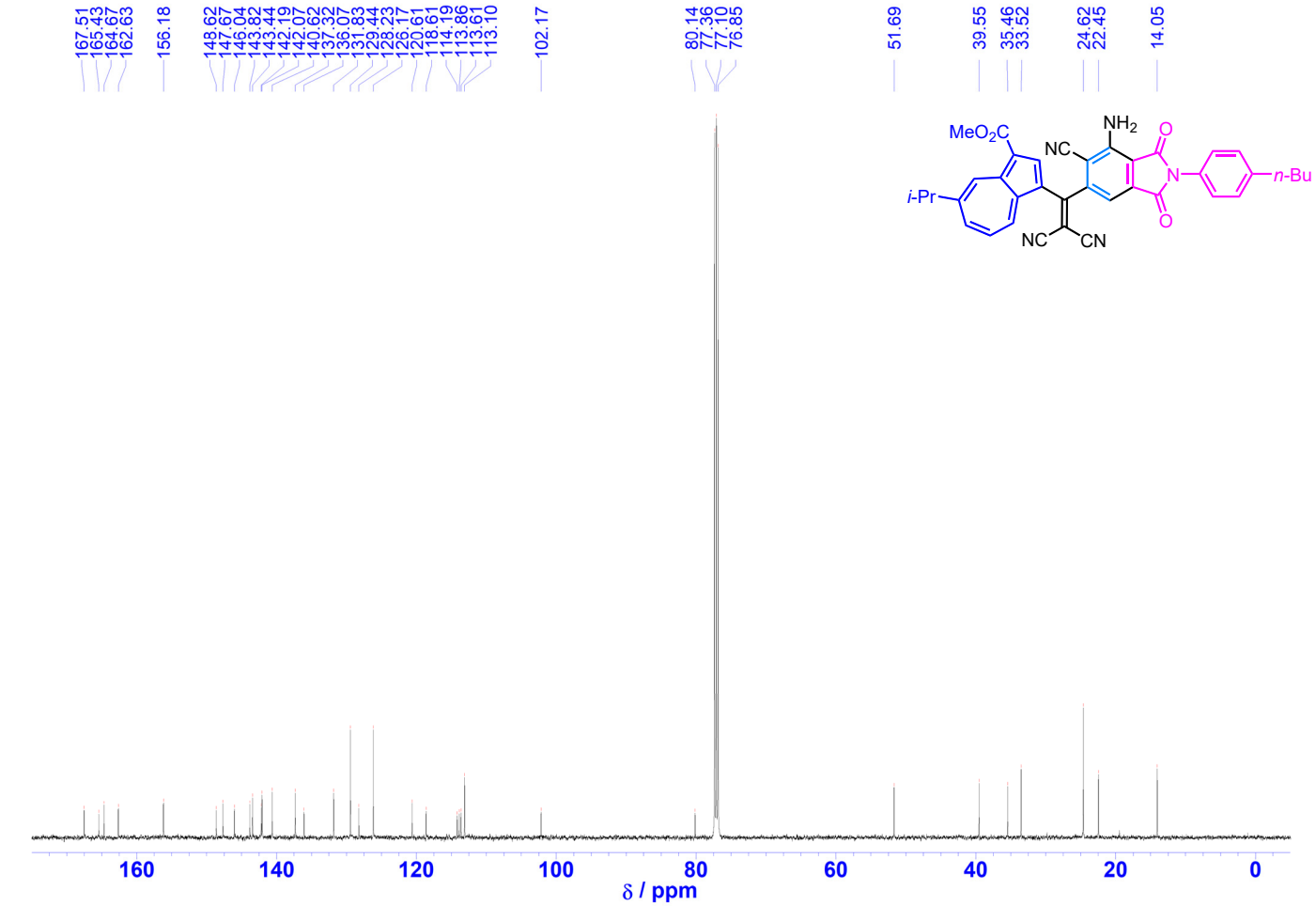


Figure S18. ^{13}C NMR spectrum of **2e** in $CDCl_3$ (125 MHz).

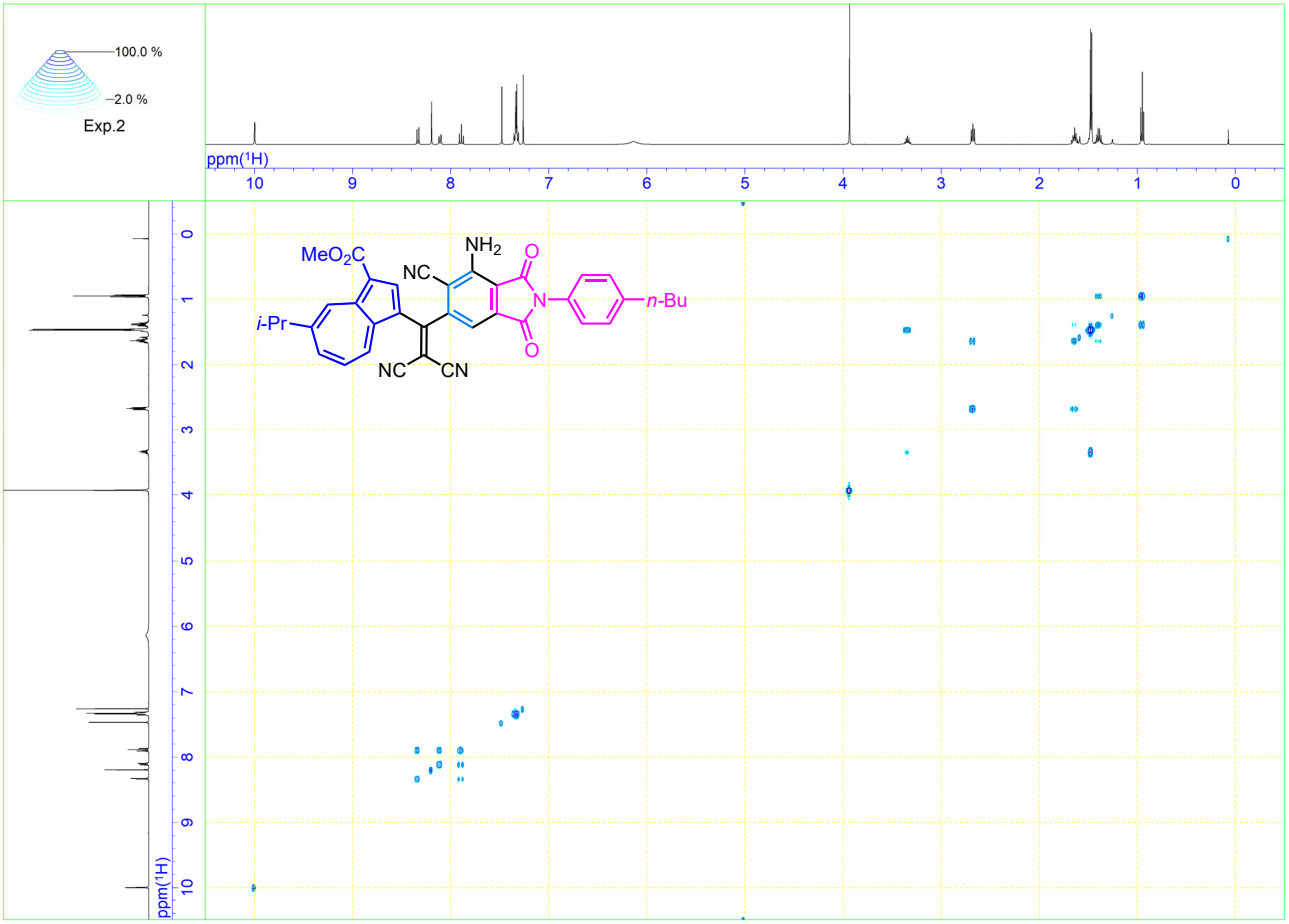


Figure S19. COSY spectrum of **2e** in $CDCl_3$ (500 MHz).

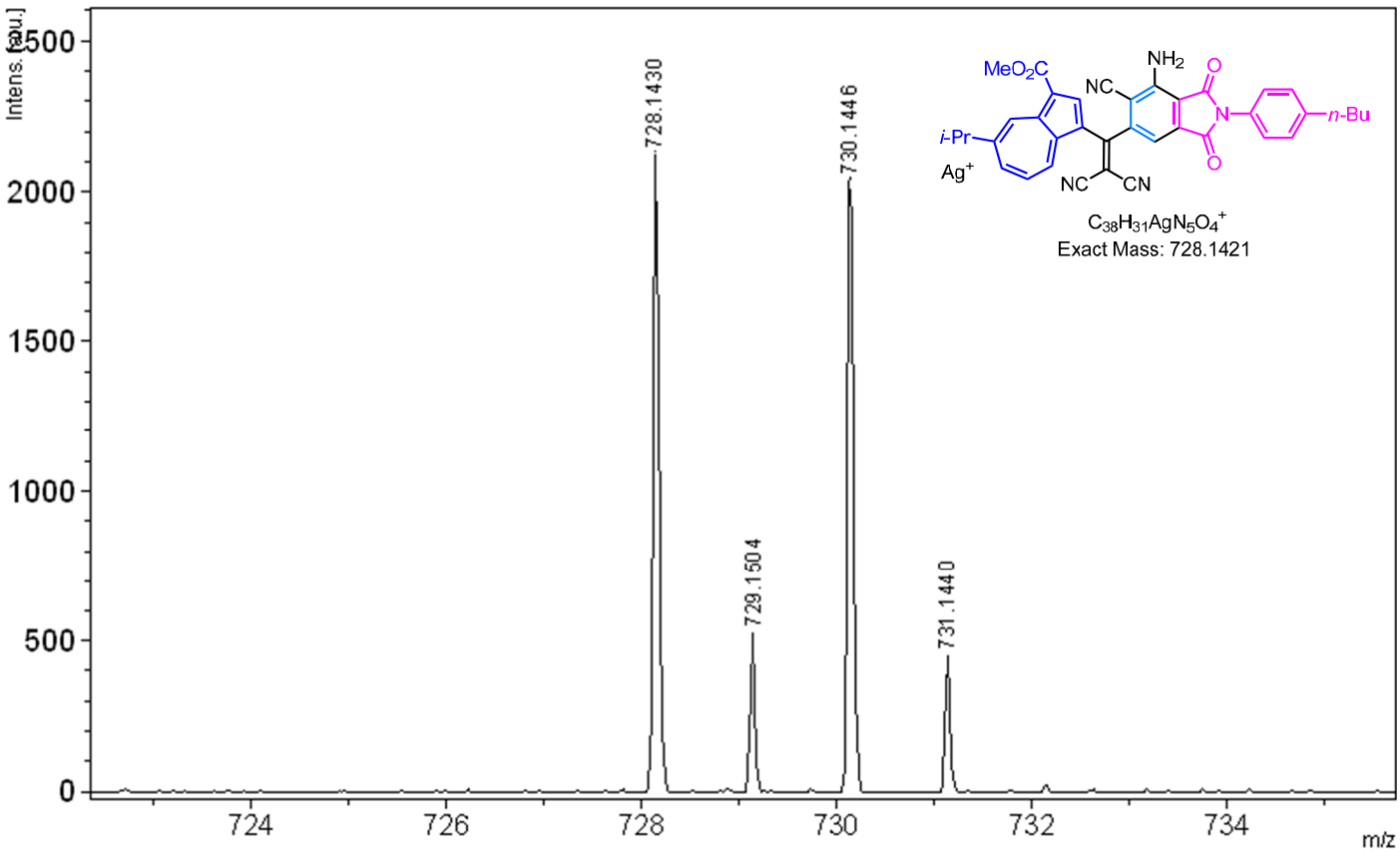


Figure S20. HRMS (MALDI-TOF, positive) of **2e**.

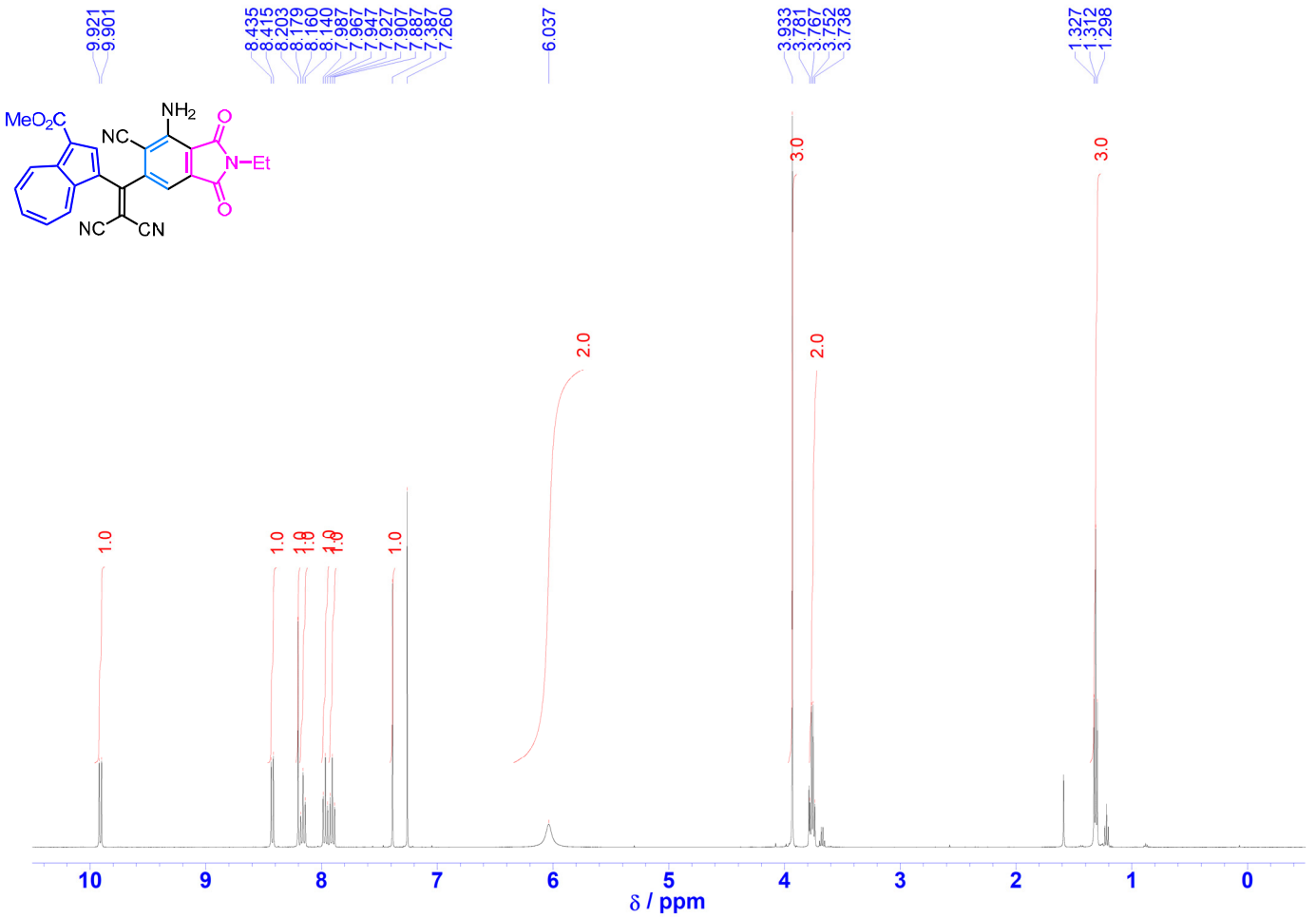


Figure S21. ^{1}H NMR spectrum of **4a** in $CDCl_3$ (500 MHz).

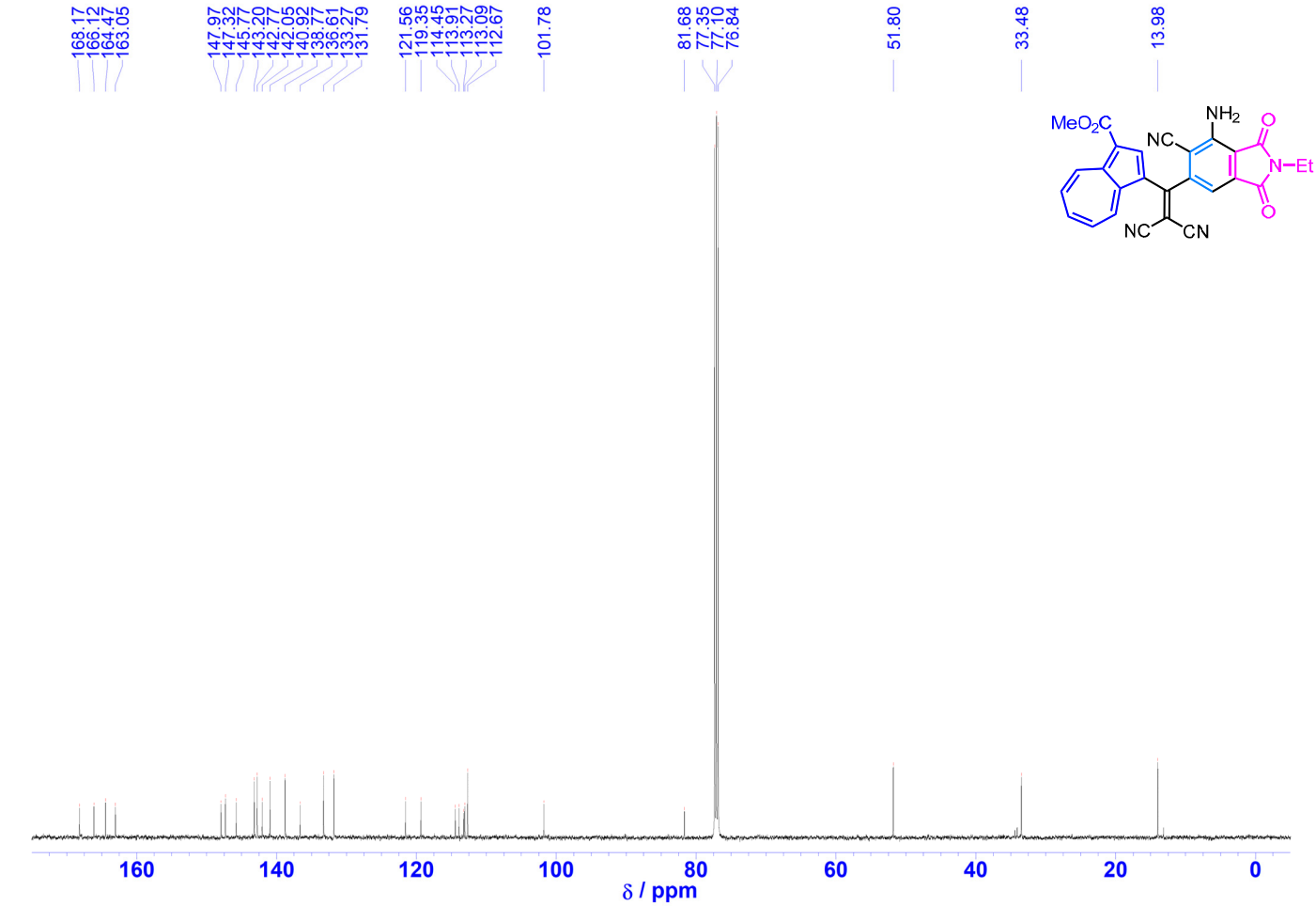


Figure S22. ^{13}C NMR spectrum of **4a** in $CDCl_3$ (125 MHz).

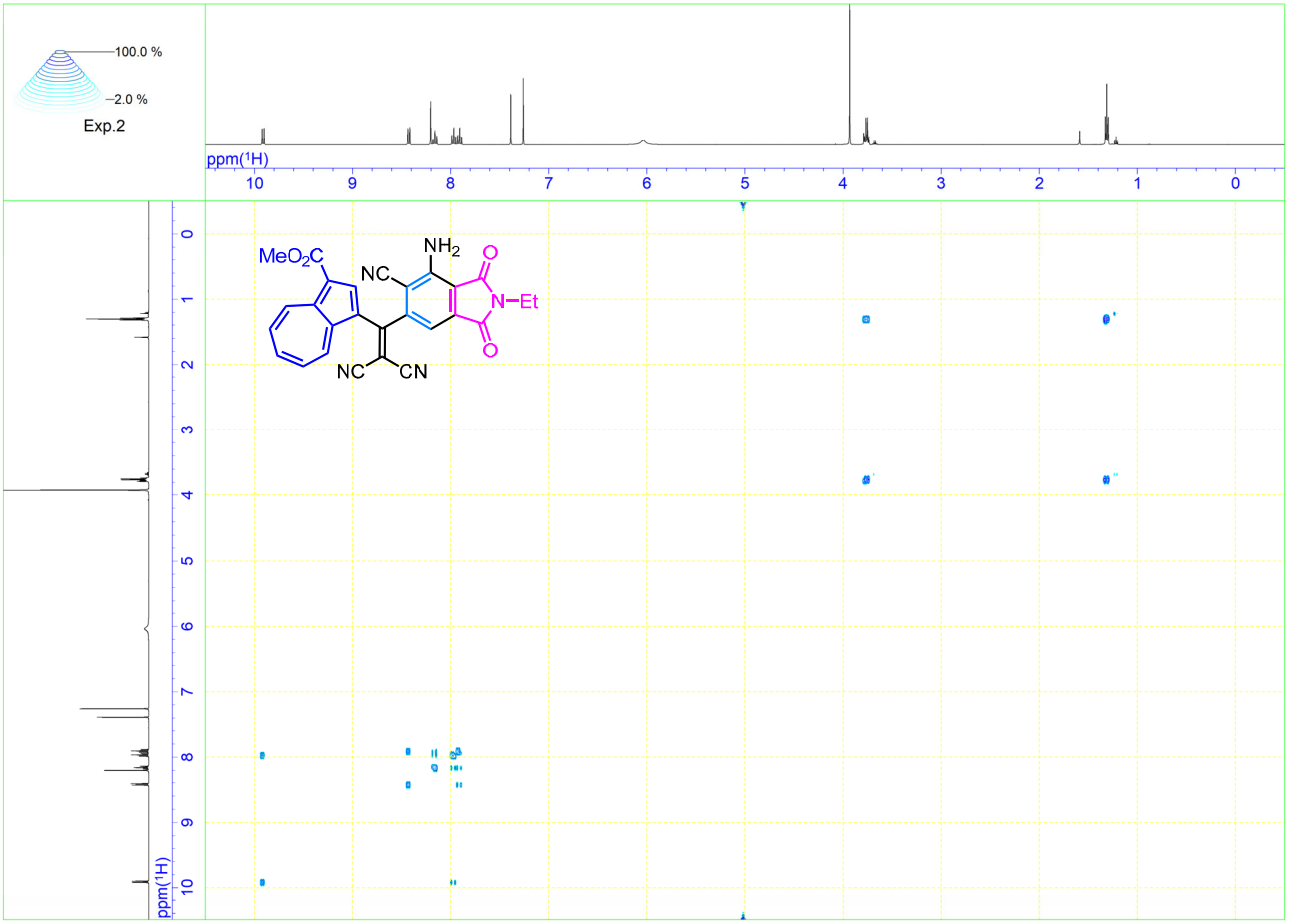


Figure S23. COSY spectrum of **4a** in $CDCl_3$ (500 MHz).

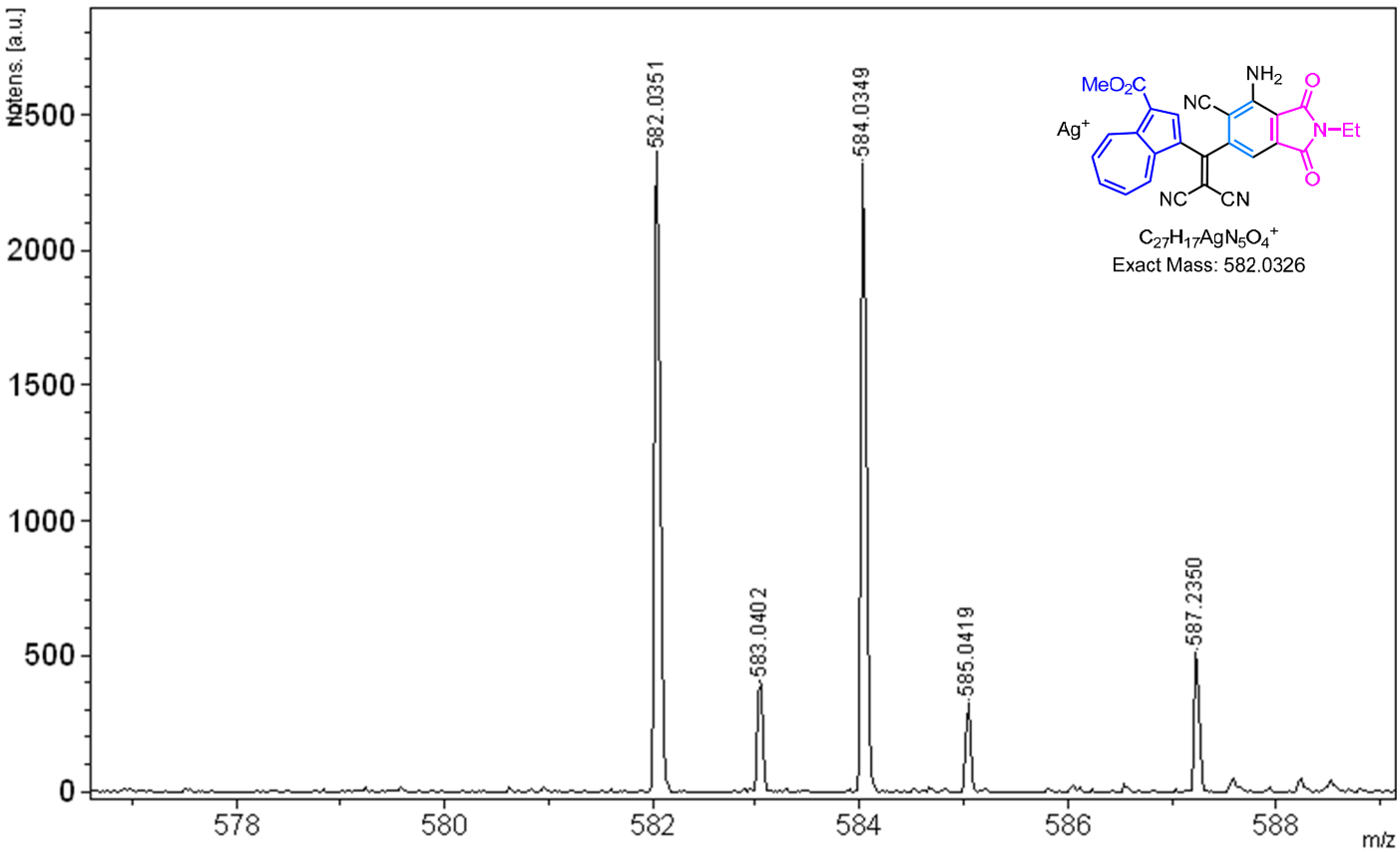


Figure S24. HRMS (MALDI-TOF, positive) of **4a**.

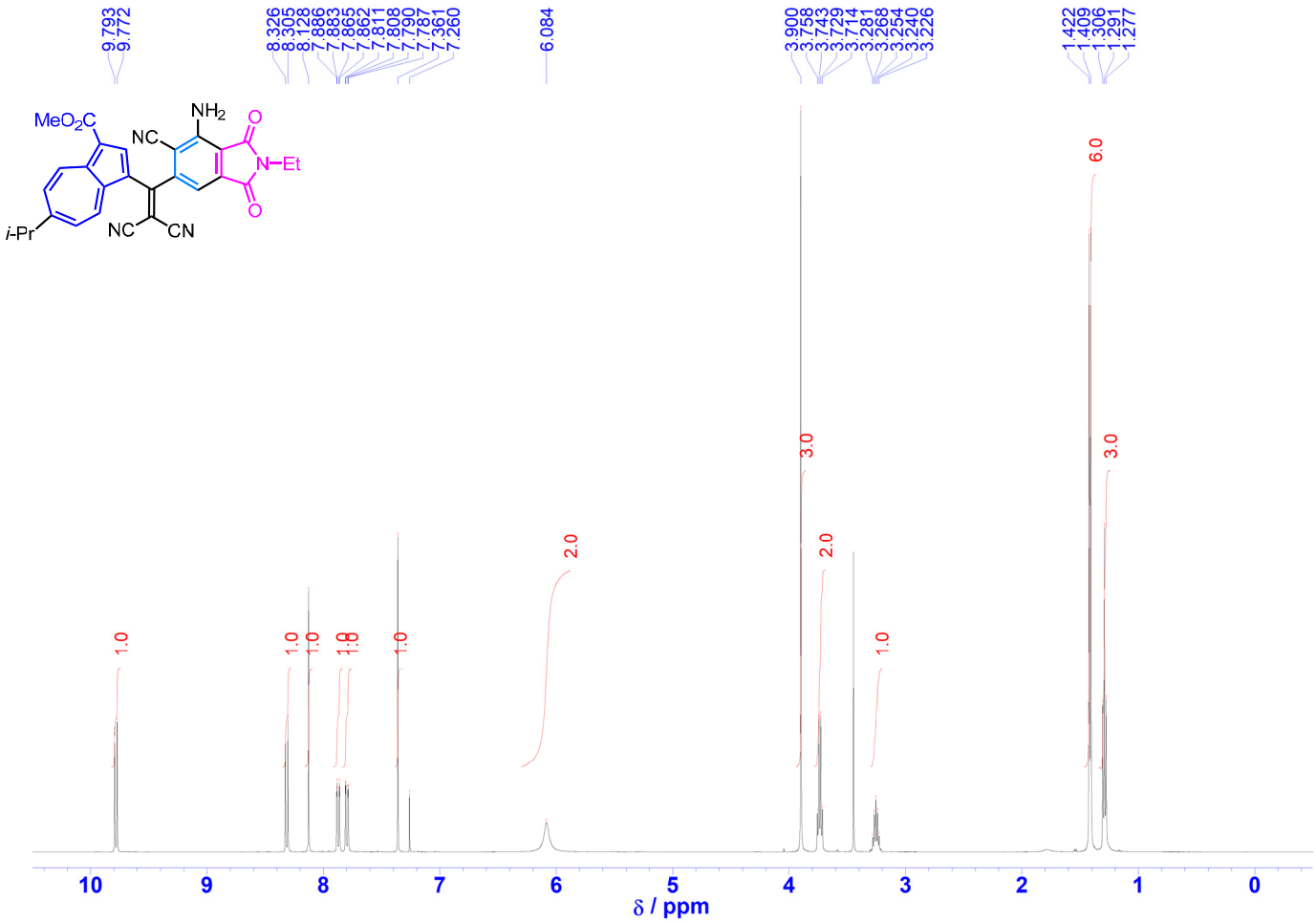


Figure S25. ^{1}H NMR spectrum of **4b** in $CDCl_3$ (500 MHz).

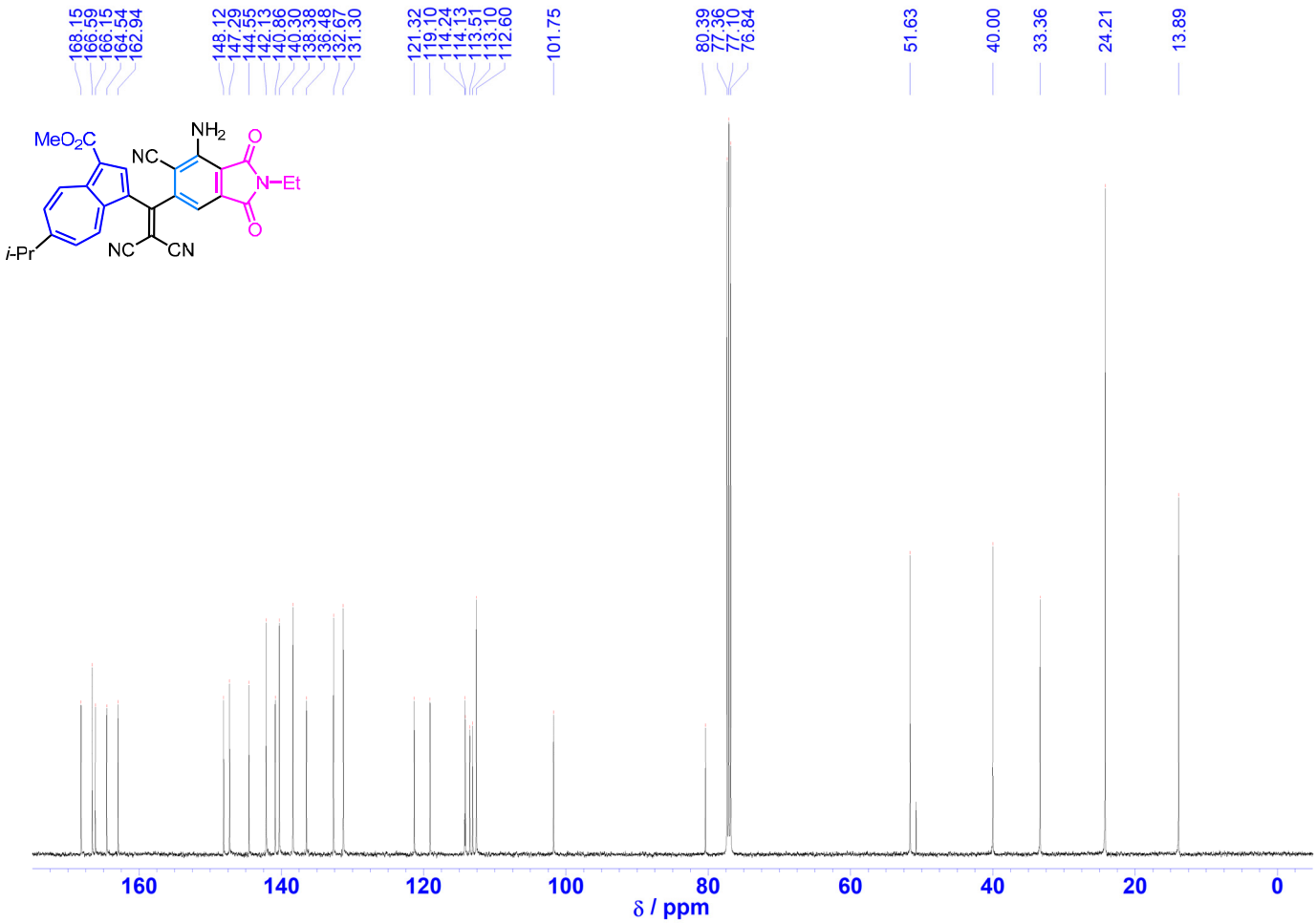


Figure S26. ^{13}C NMR spectrum of **4b** in $CDCl_3$ (125 MHz).

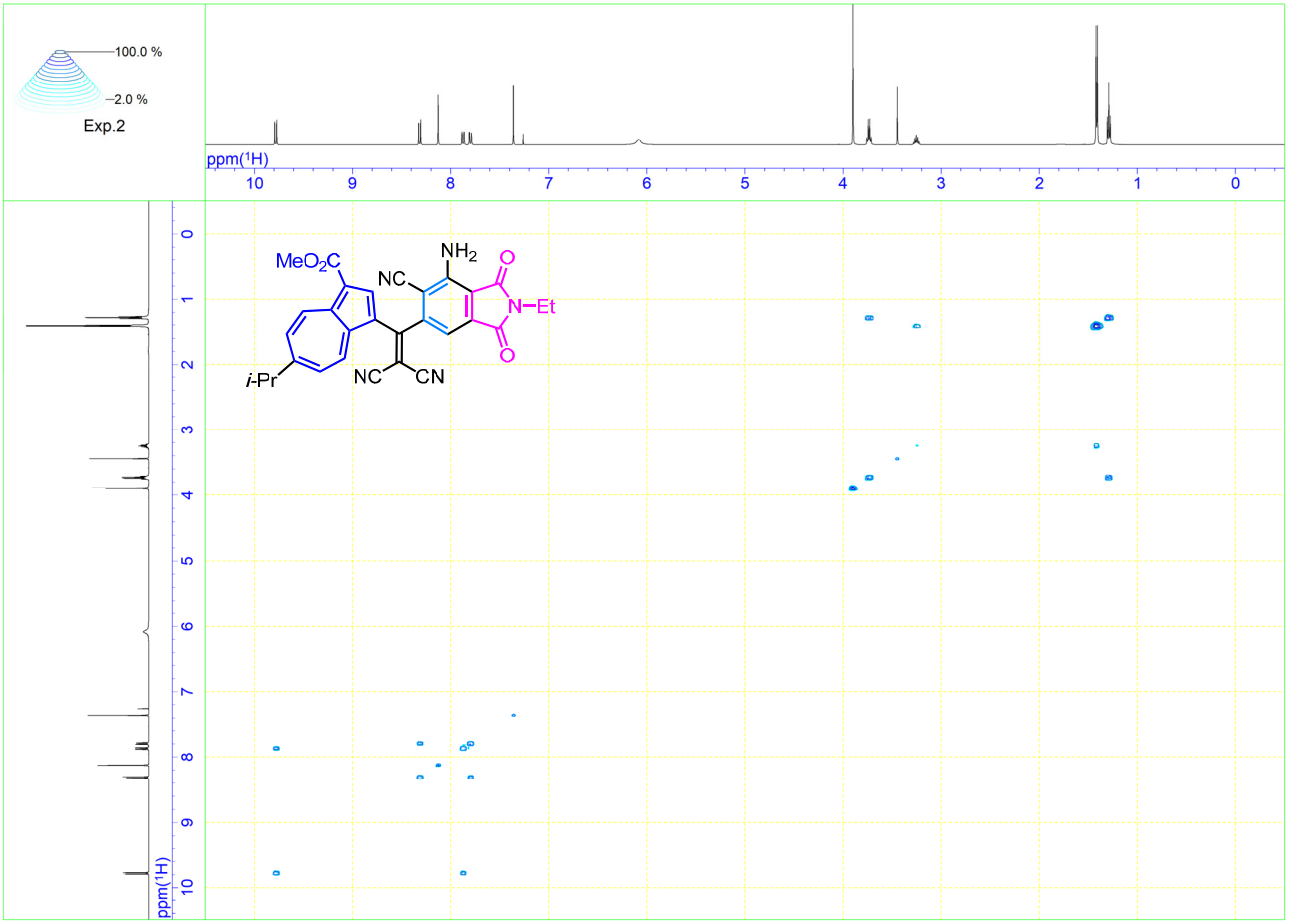


Figure S27. COSY spectrum of **4b** in $CDCl_3$ (500 MHz).

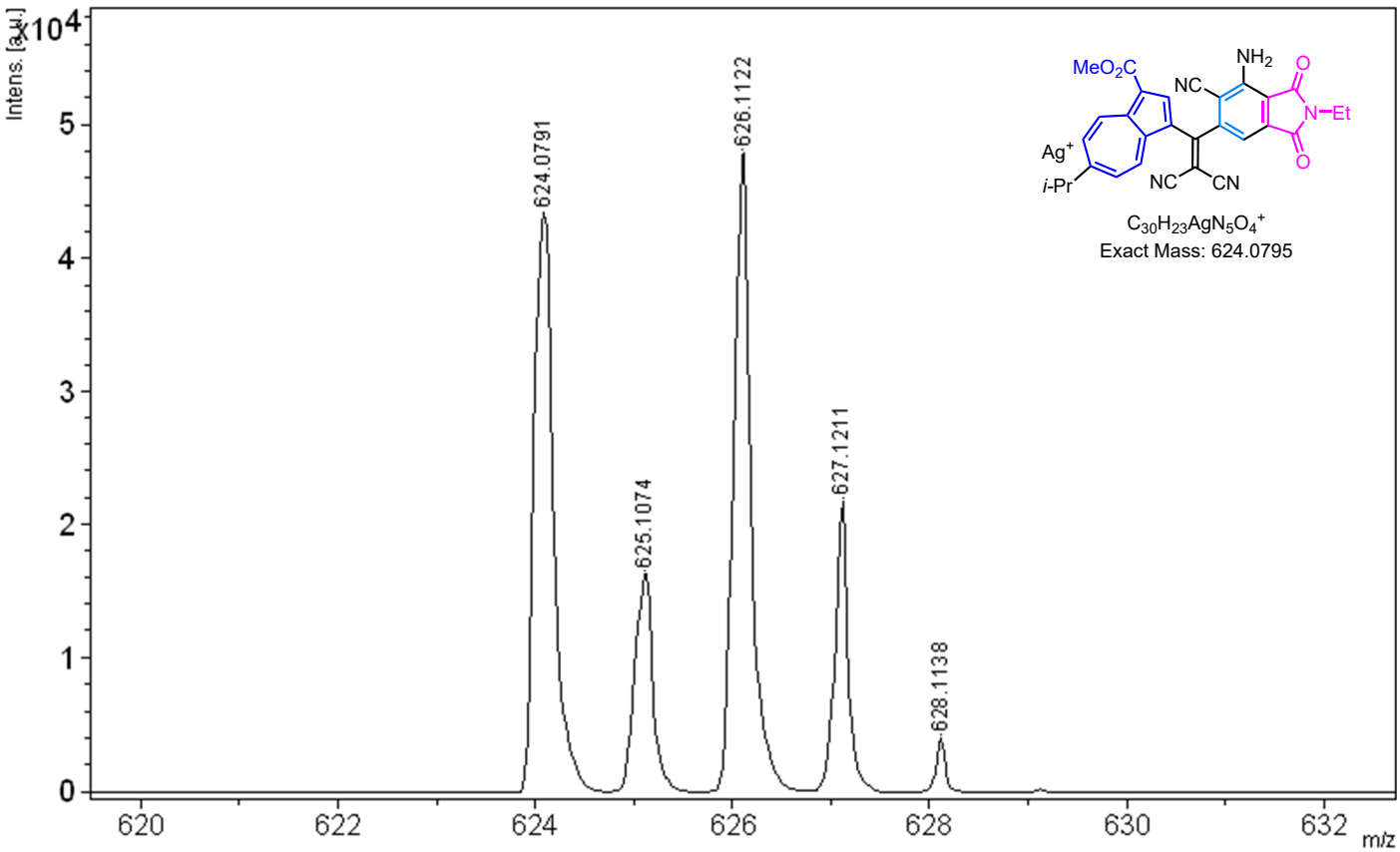


Figure S28. HRMS (MALDI-TOF, positive) of **4b**.

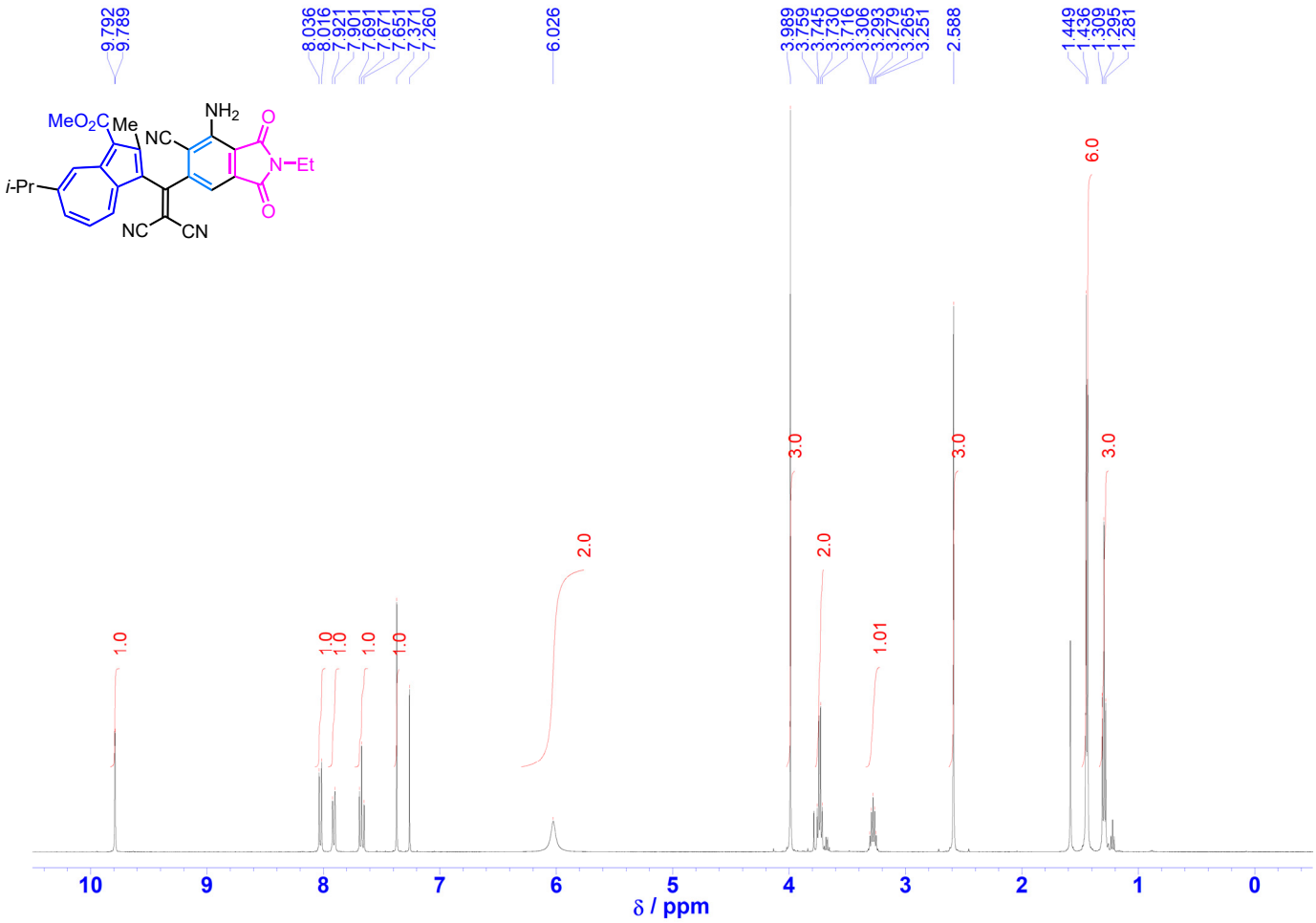


Figure S29. ^{1}H NMR spectrum of **4c** in $CDCl_3$ (500 MHz).

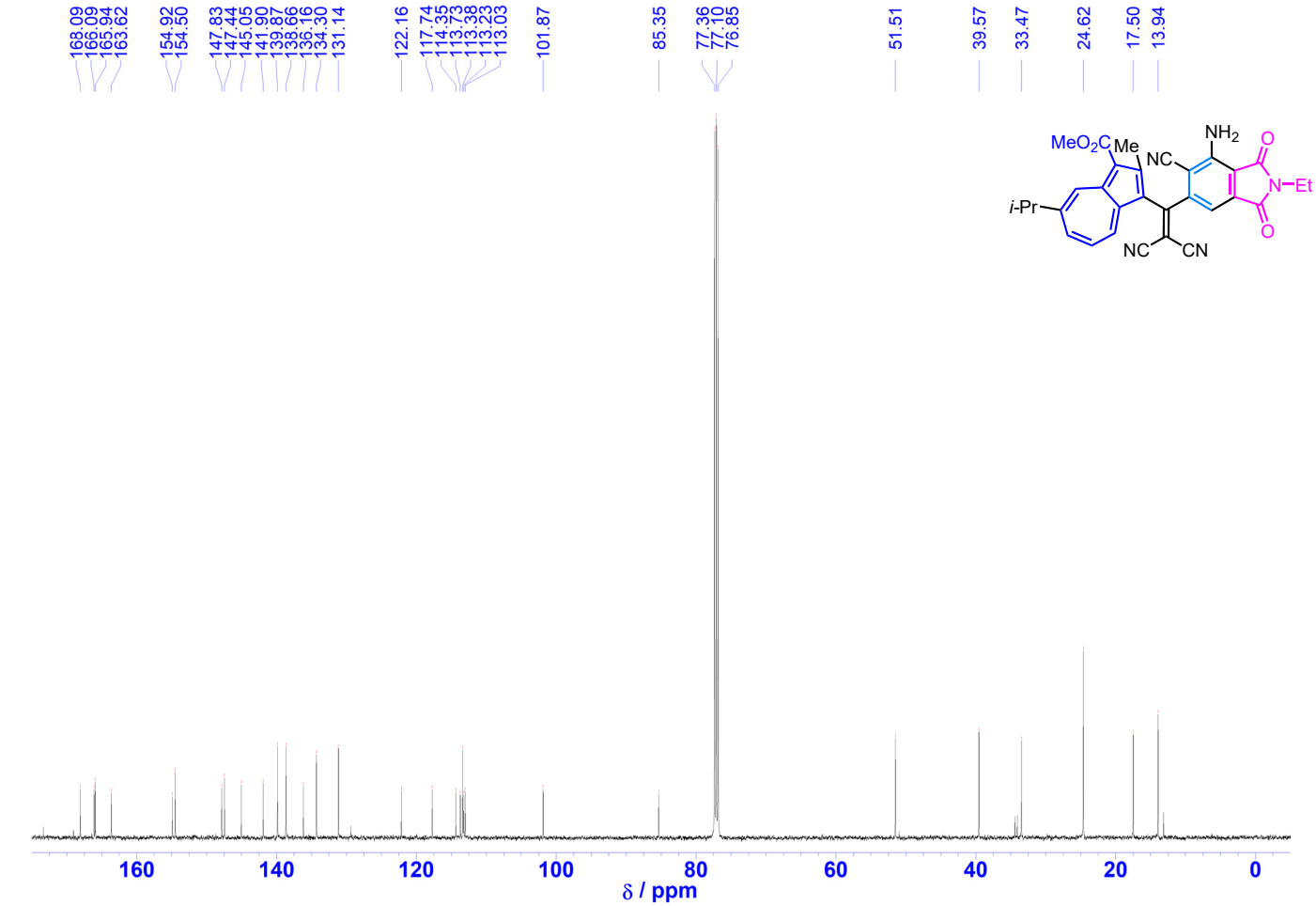


Figure S30. ^{13}C NMR spectrum of **4c** in $CDCl_3$ (125 MHz).

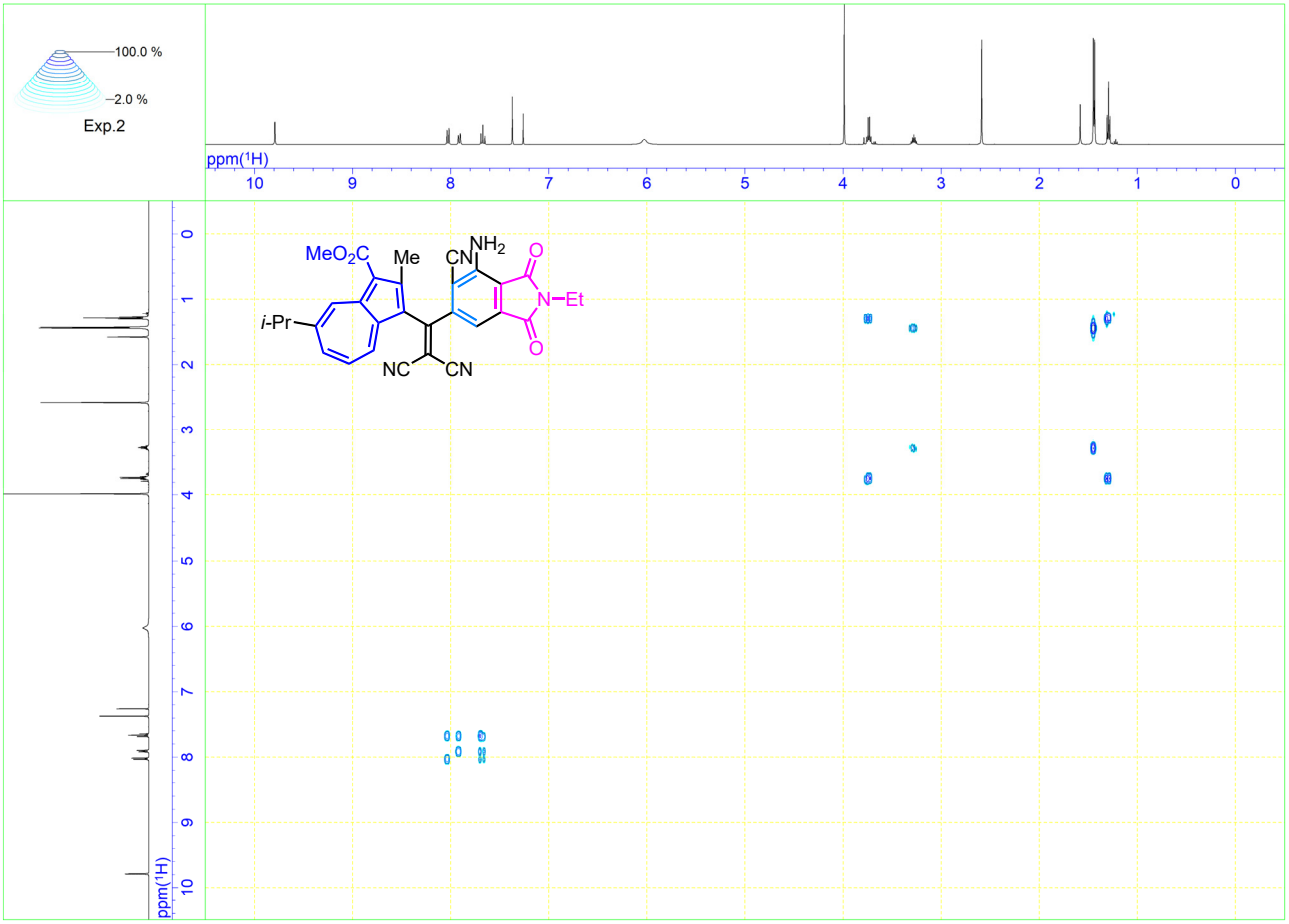


Figure S31. COSY spectrum of **4c** in $CDCl_3$ (500 MHz).

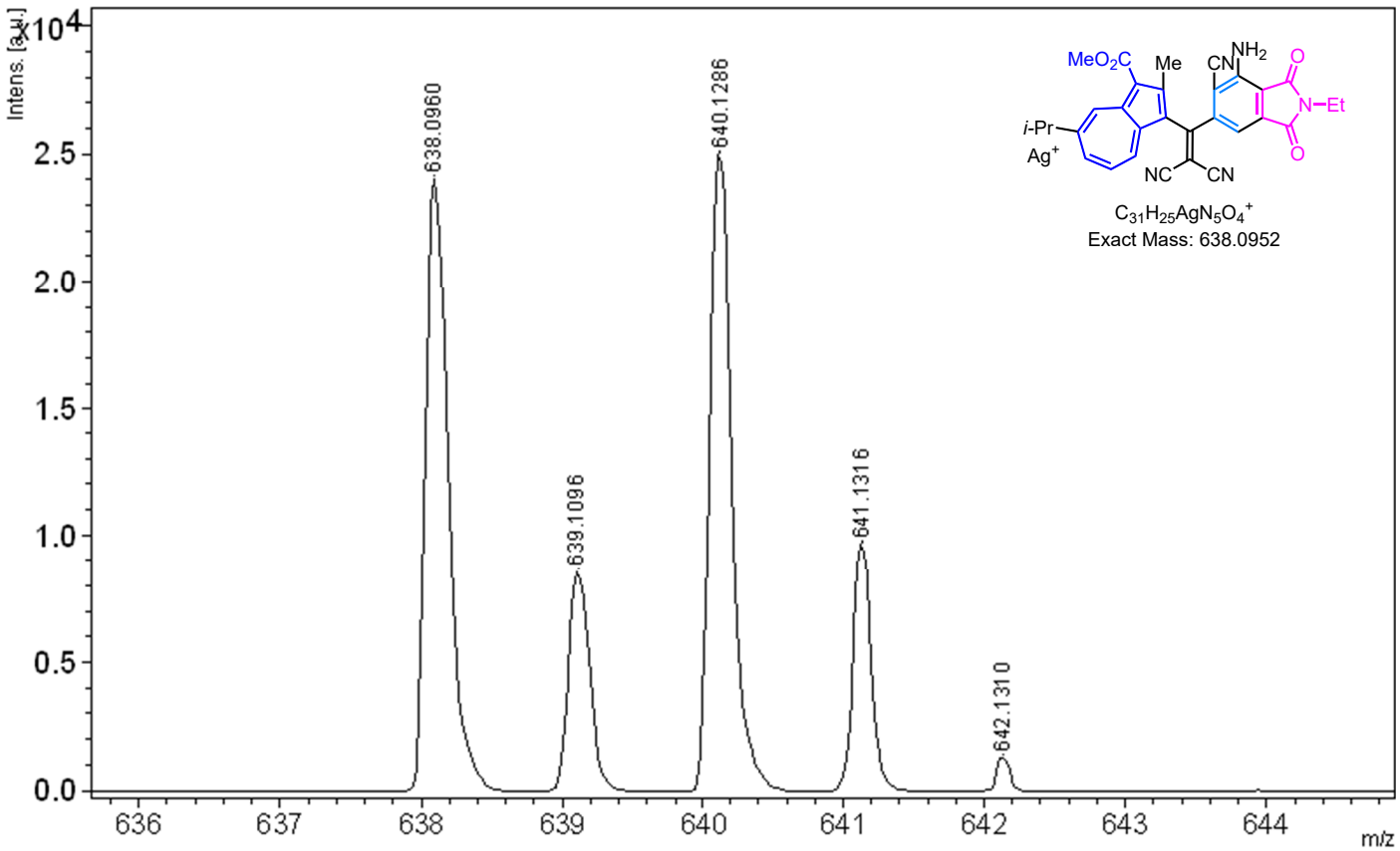


Figure S32. HRMS (MALDI-TOF, positive) of **4c**.

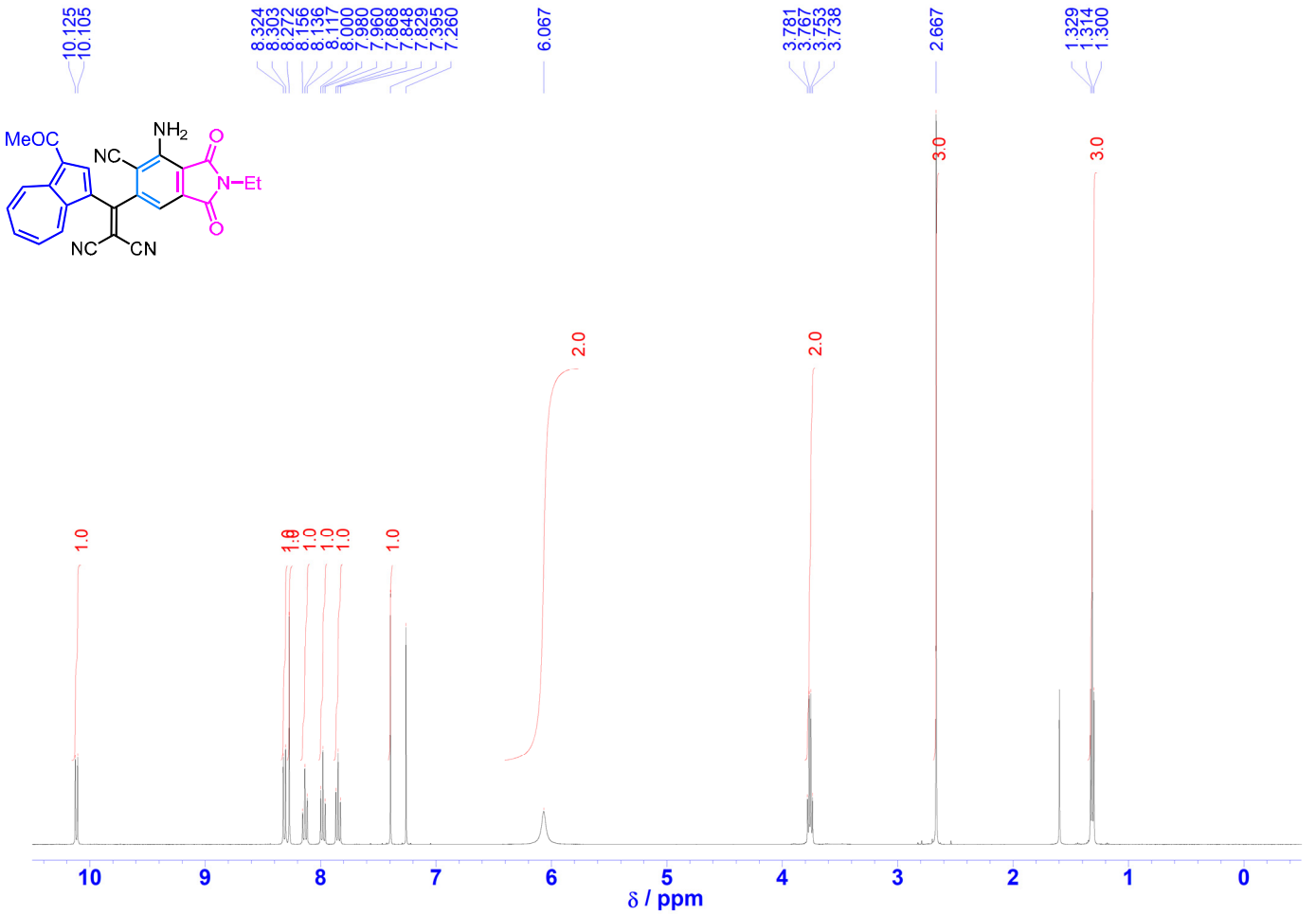


Figure S33. ^{1}H NMR spectrum of **4d** in $CDCl_3$ (500 MHz).

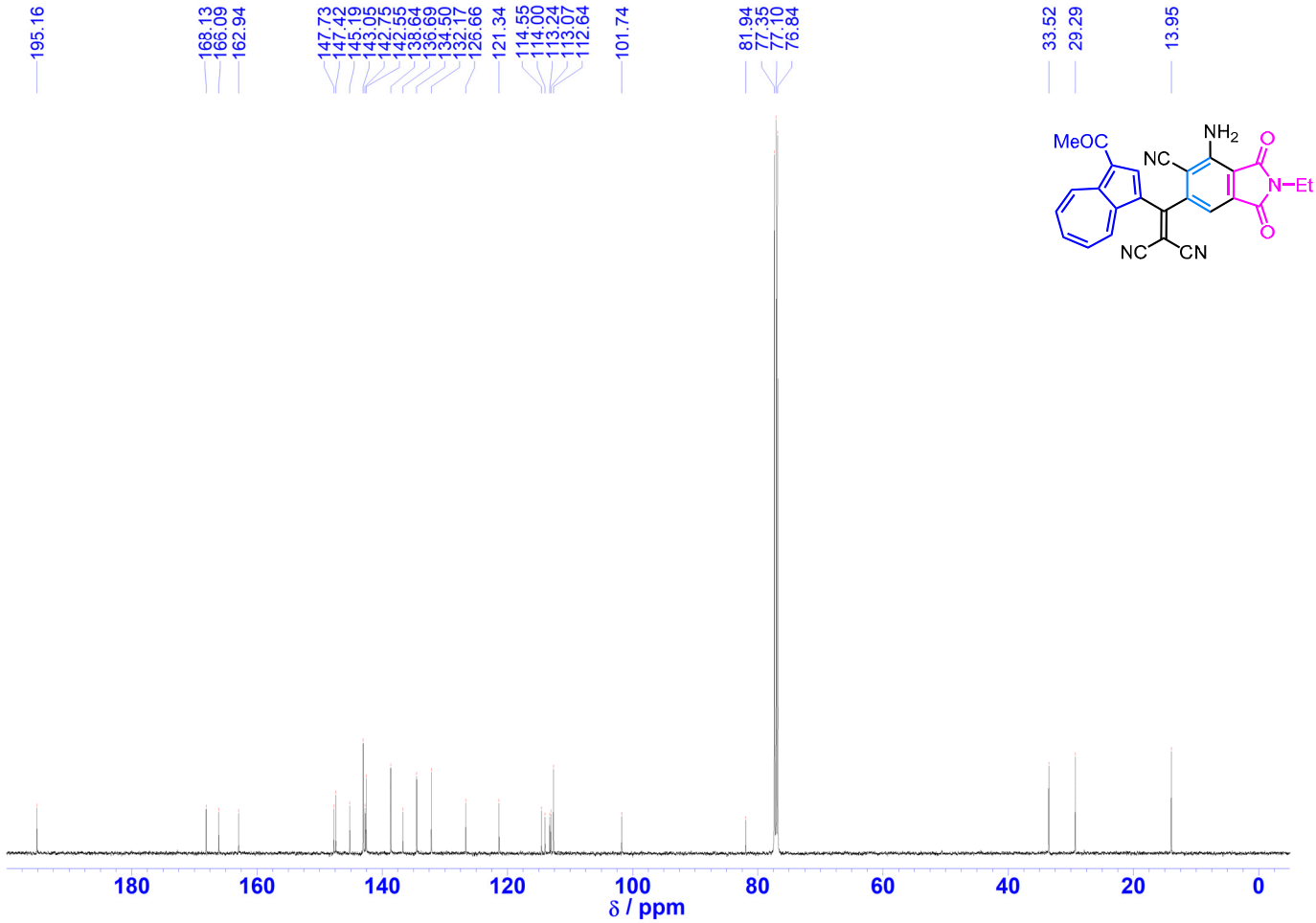


Figure S34. ^{13}C NMR spectrum of **4d** in $CDCl_3$ (125 MHz).

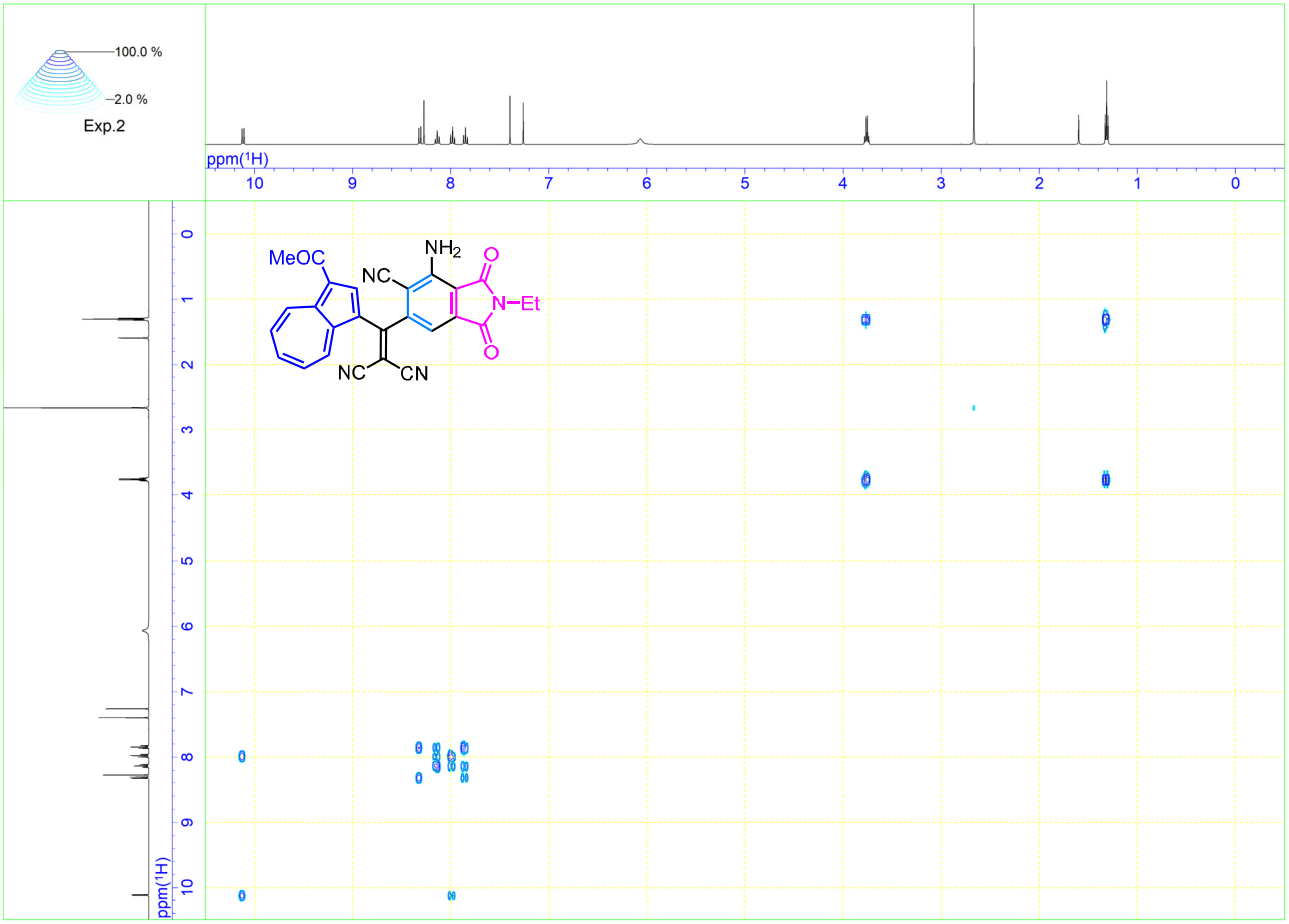


Figure S35. COSY spectrum of **4d** in $CDCl_3$ (500 MHz).

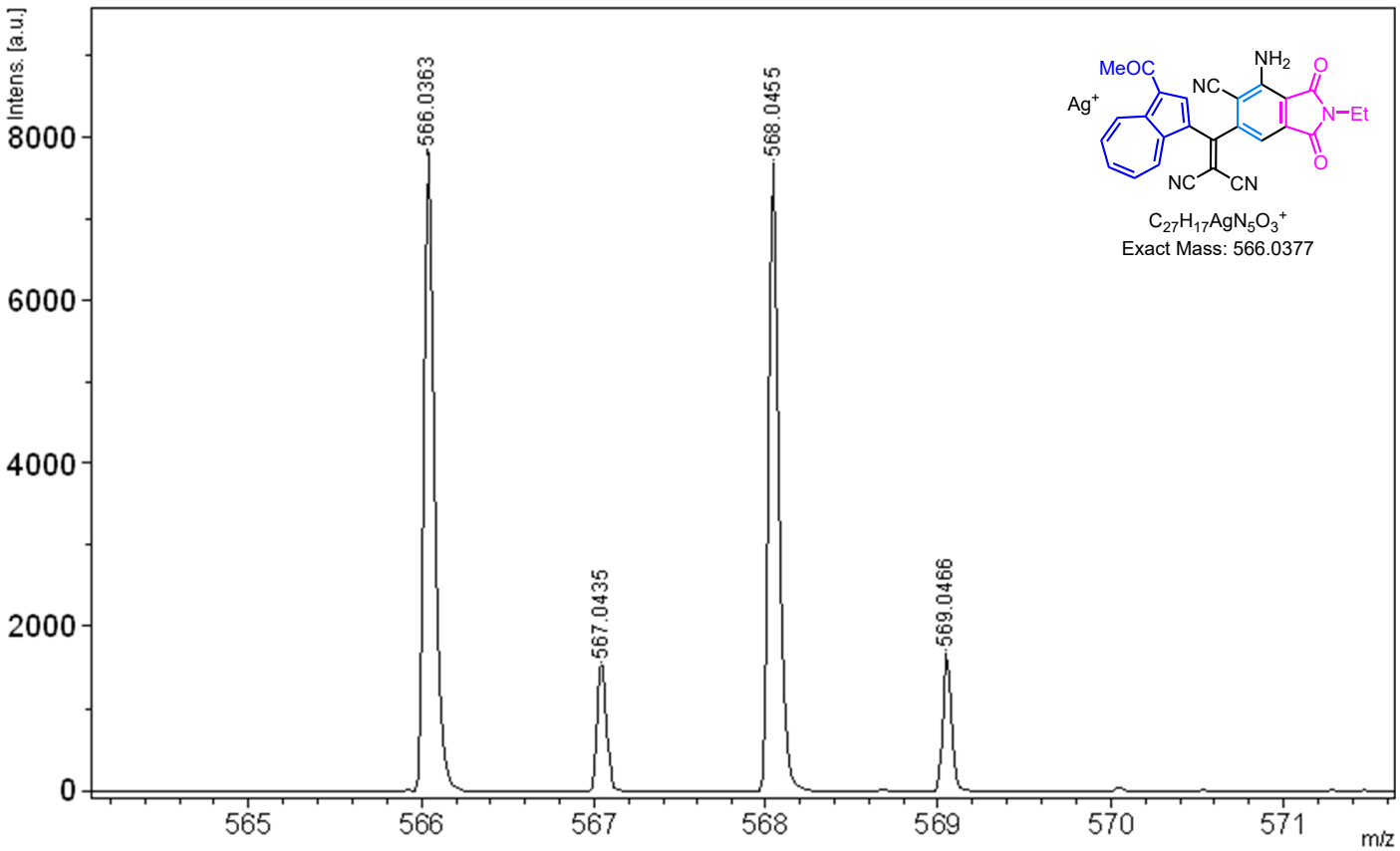


Figure S36. HRMS (MALDI-TOF, positive) of **4d**.

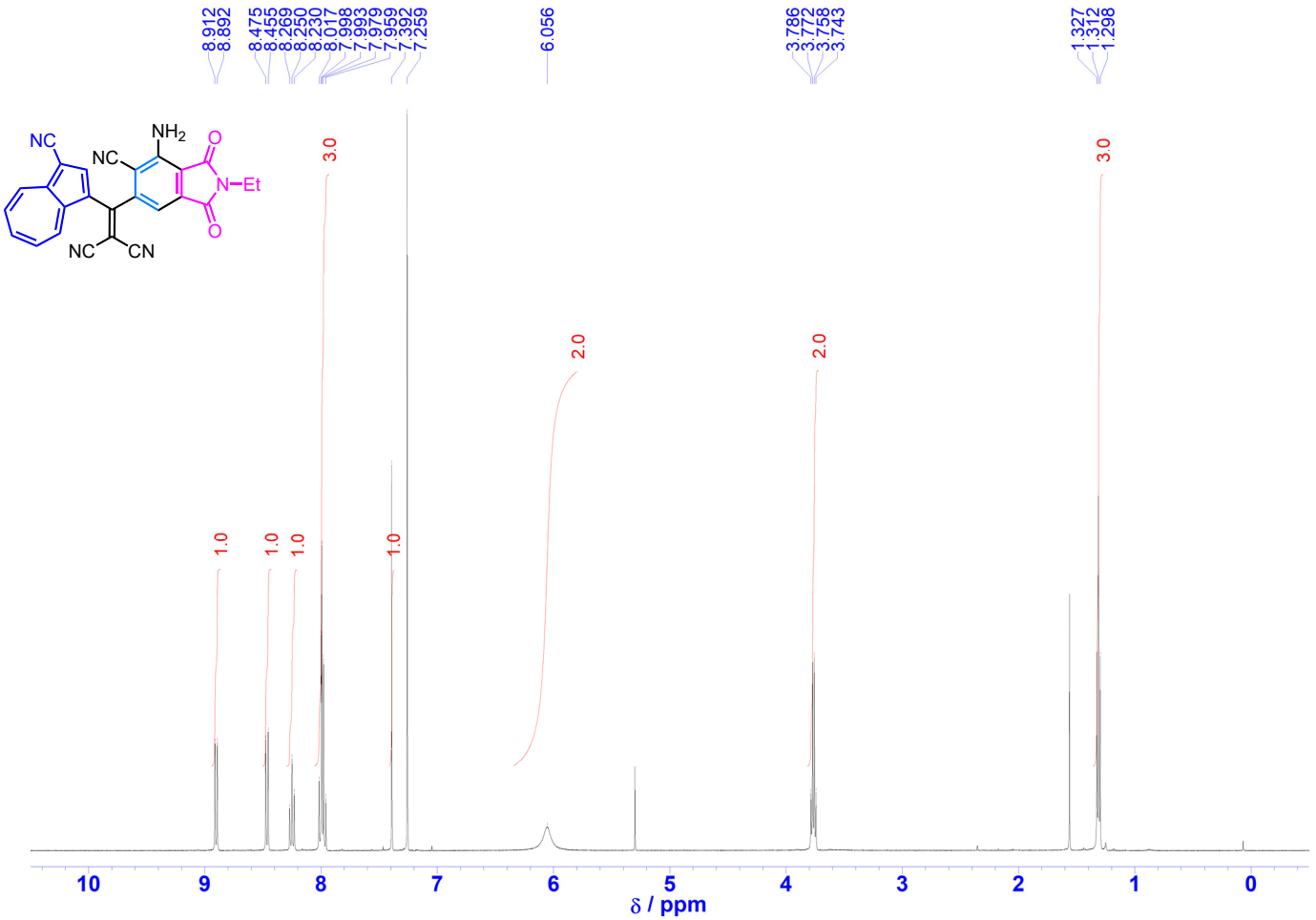


Figure S37. ^{1}H NMR spectrum of **4e** in $CDCl_3$ (500 MHz).

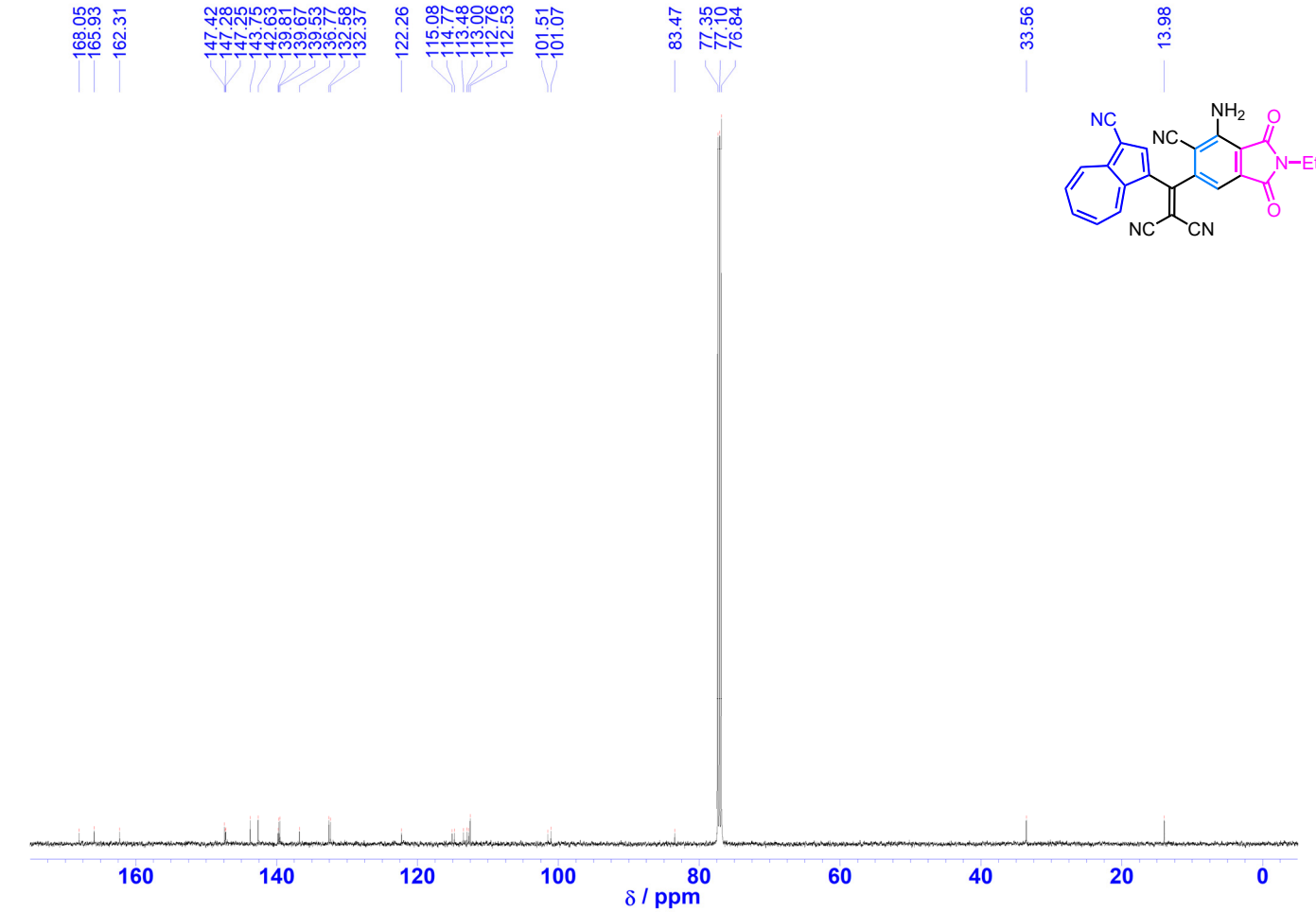


Figure S38. ^{13}C NMR spectrum of **4e** in $CDCl_3$ (125 MHz).

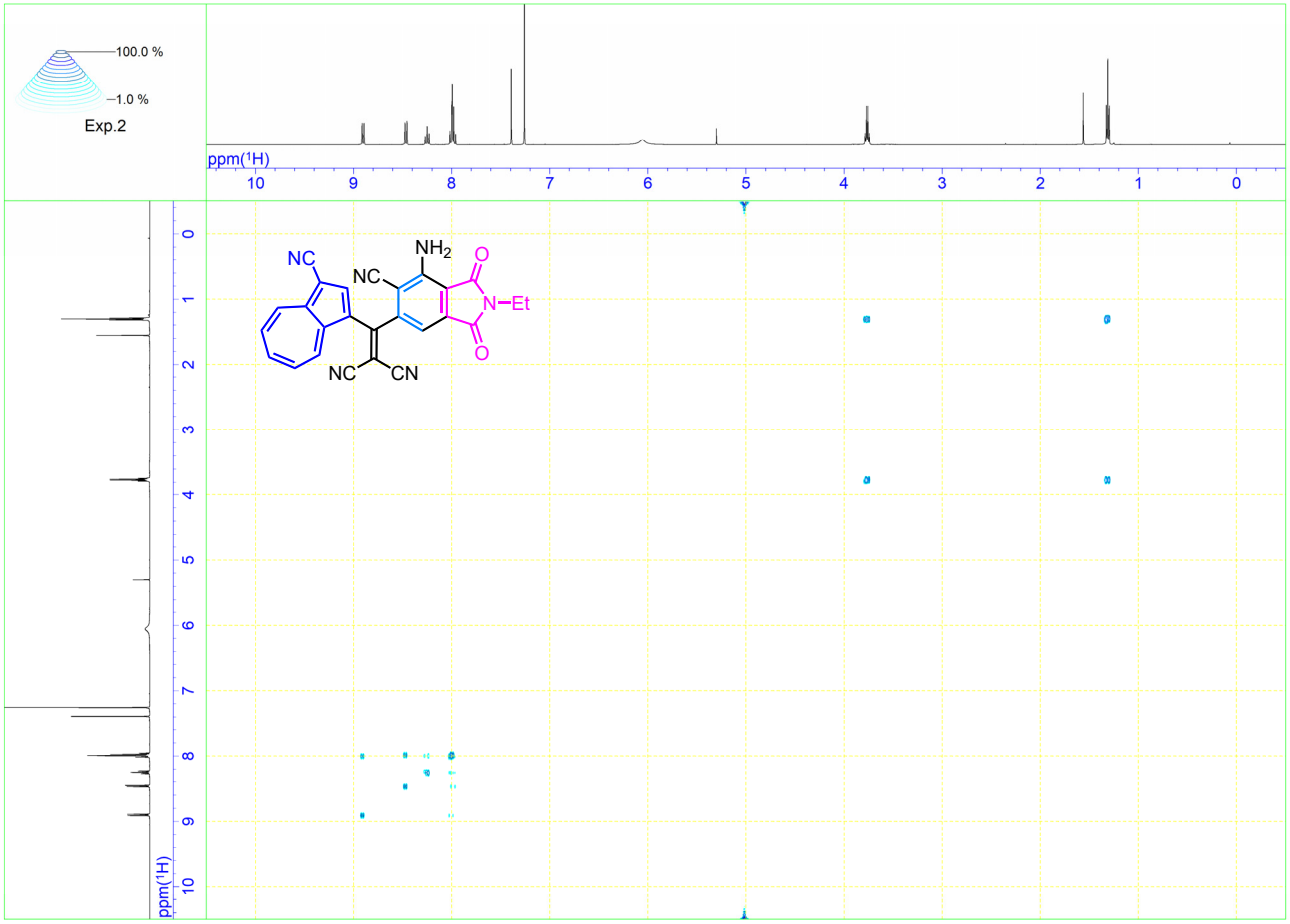


Figure S39. COSY spectrum of **4e** in $CDCl_3$ (500 MHz).

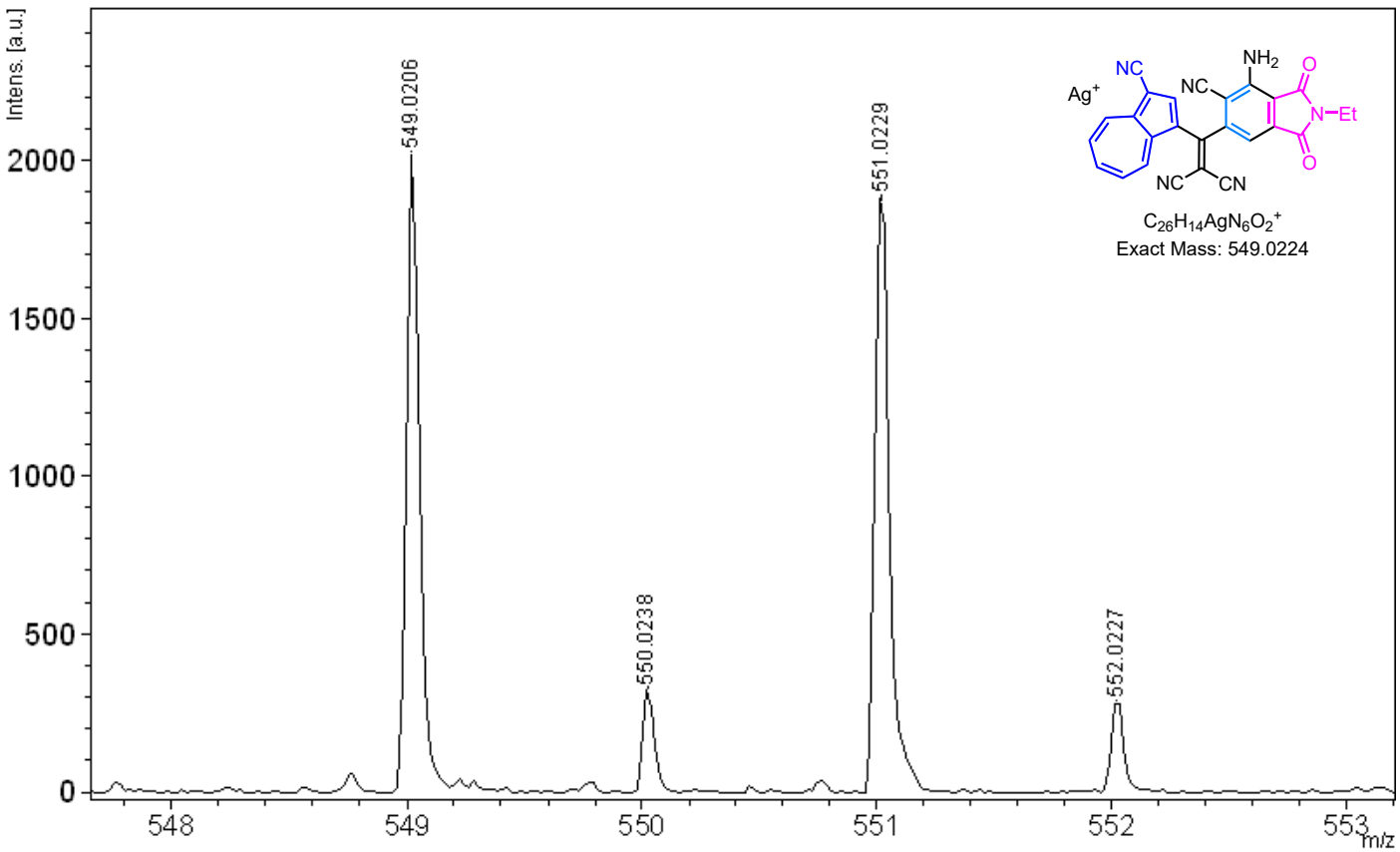


Figure S40. HRMS (MALDI-TOF, positive) of **4e**.

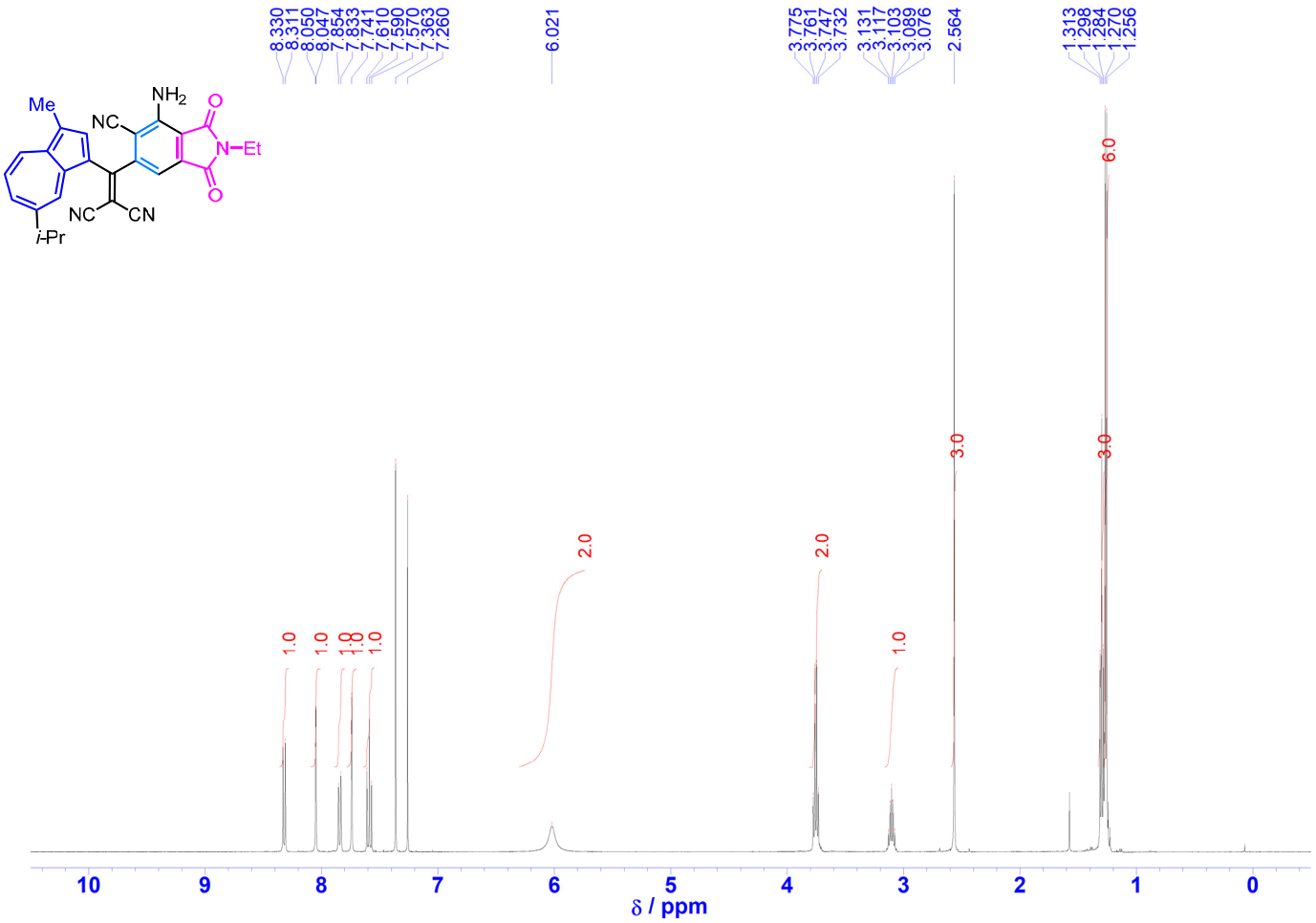


Figure S41. ^{1}H NMR spectrum of **4f** in $CDCl_3$ (500 MHz).

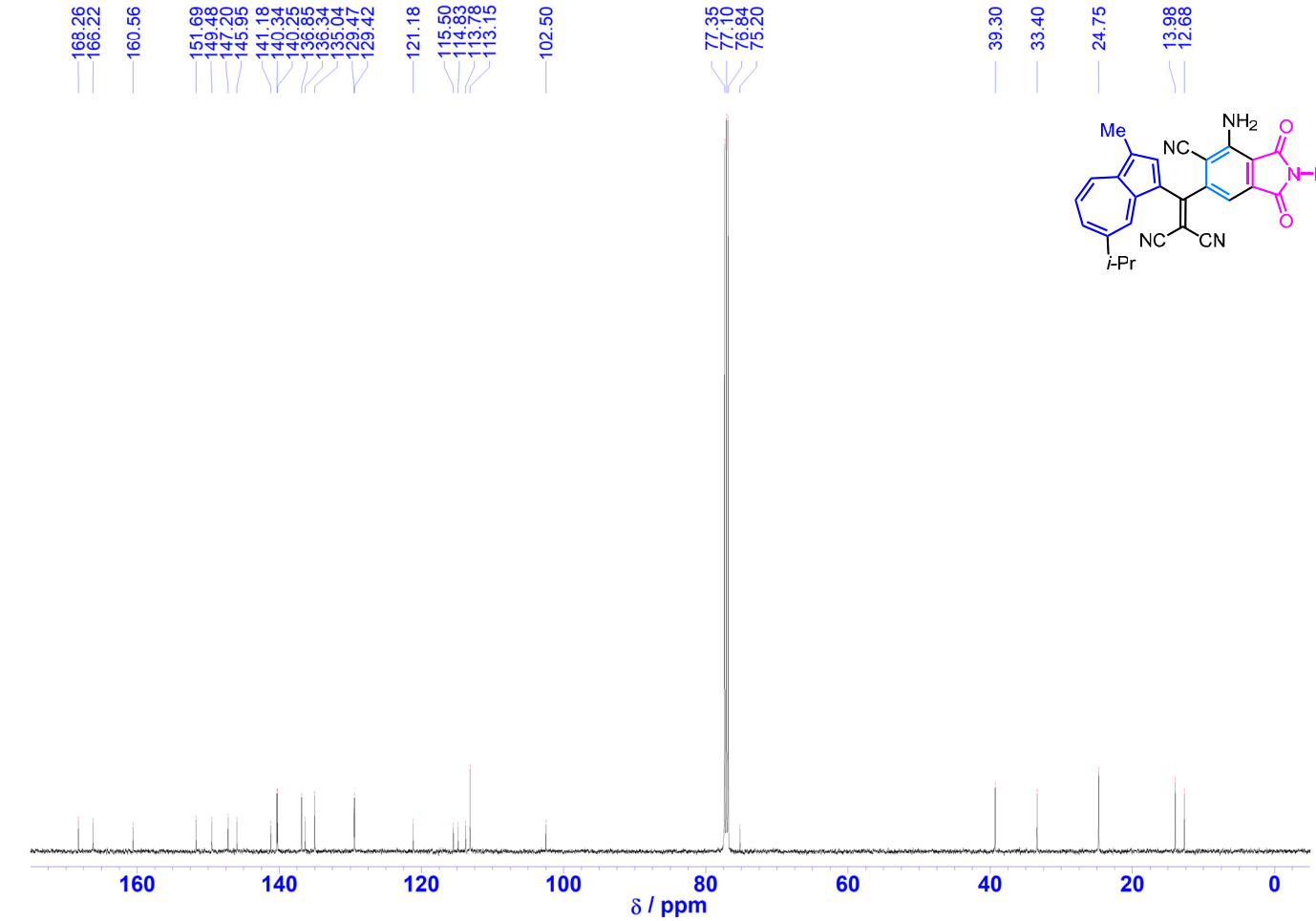


Figure S42. ^{13}C NMR spectrum of **4f** in $CDCl_3$ (125 MHz).

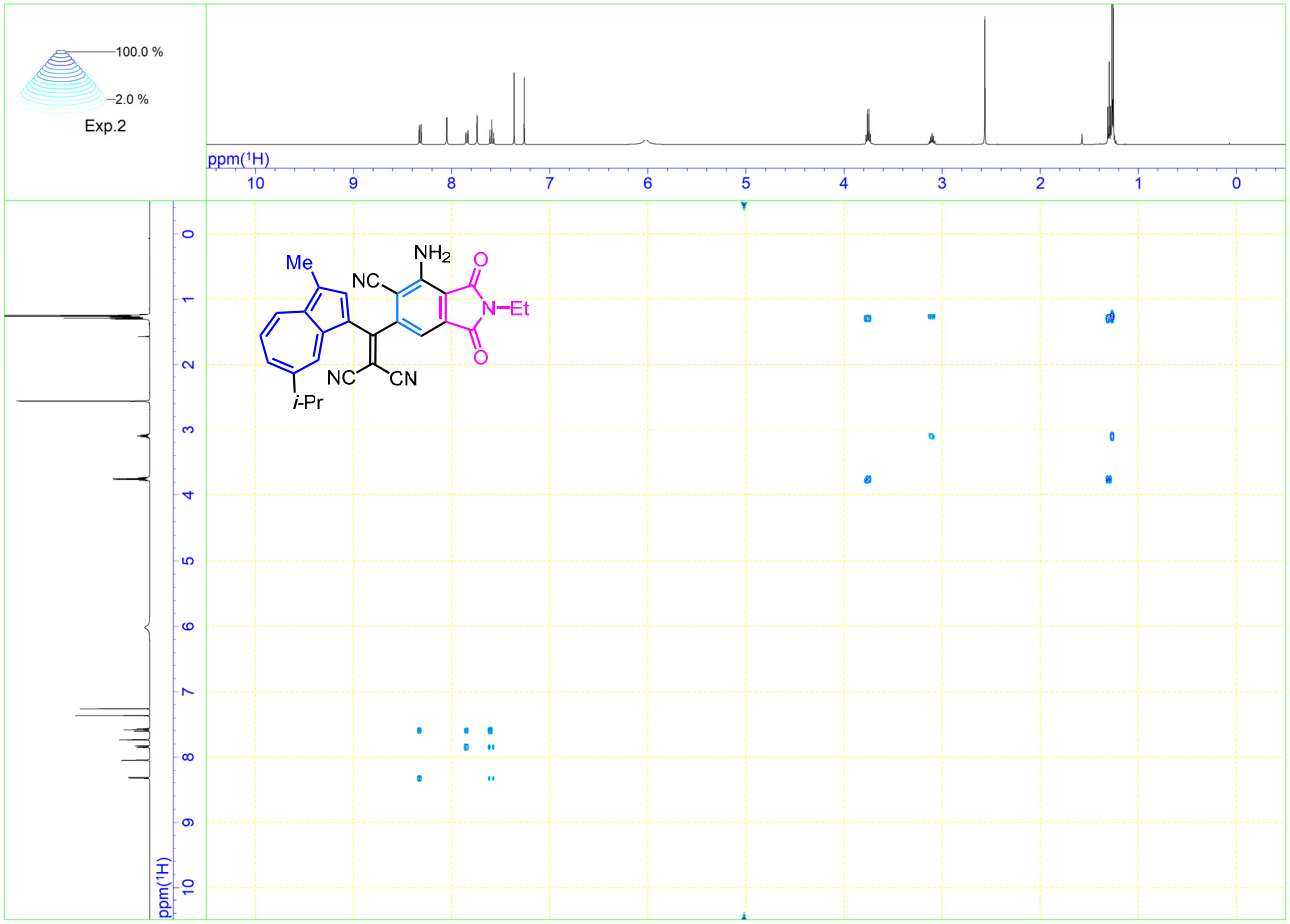


Figure S43. COSY spectrum of **4f** in $CDCl_3$ (500 MHz).

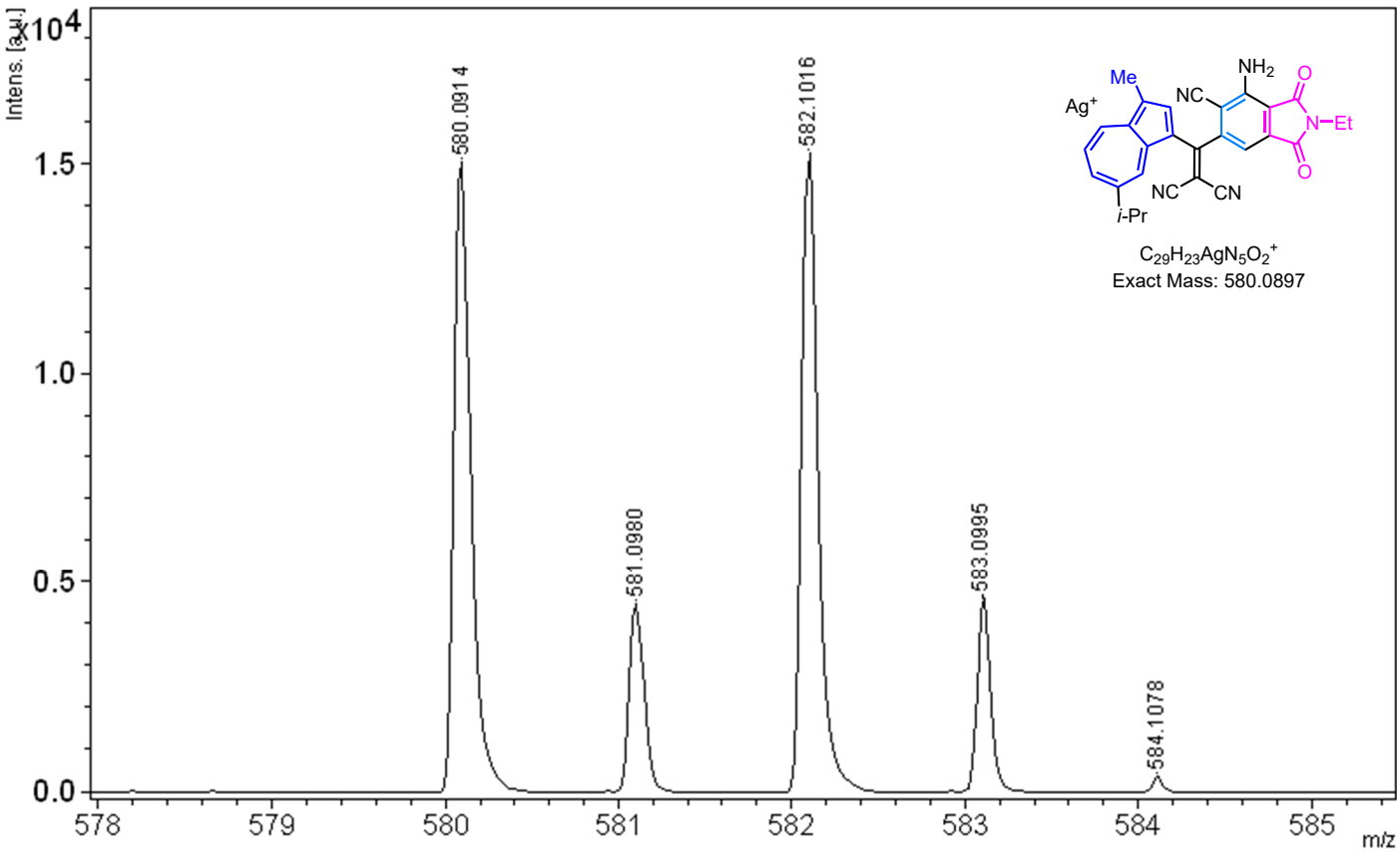


Figure S44. HRMS (MALDI-TOF, positive) of **4f**.

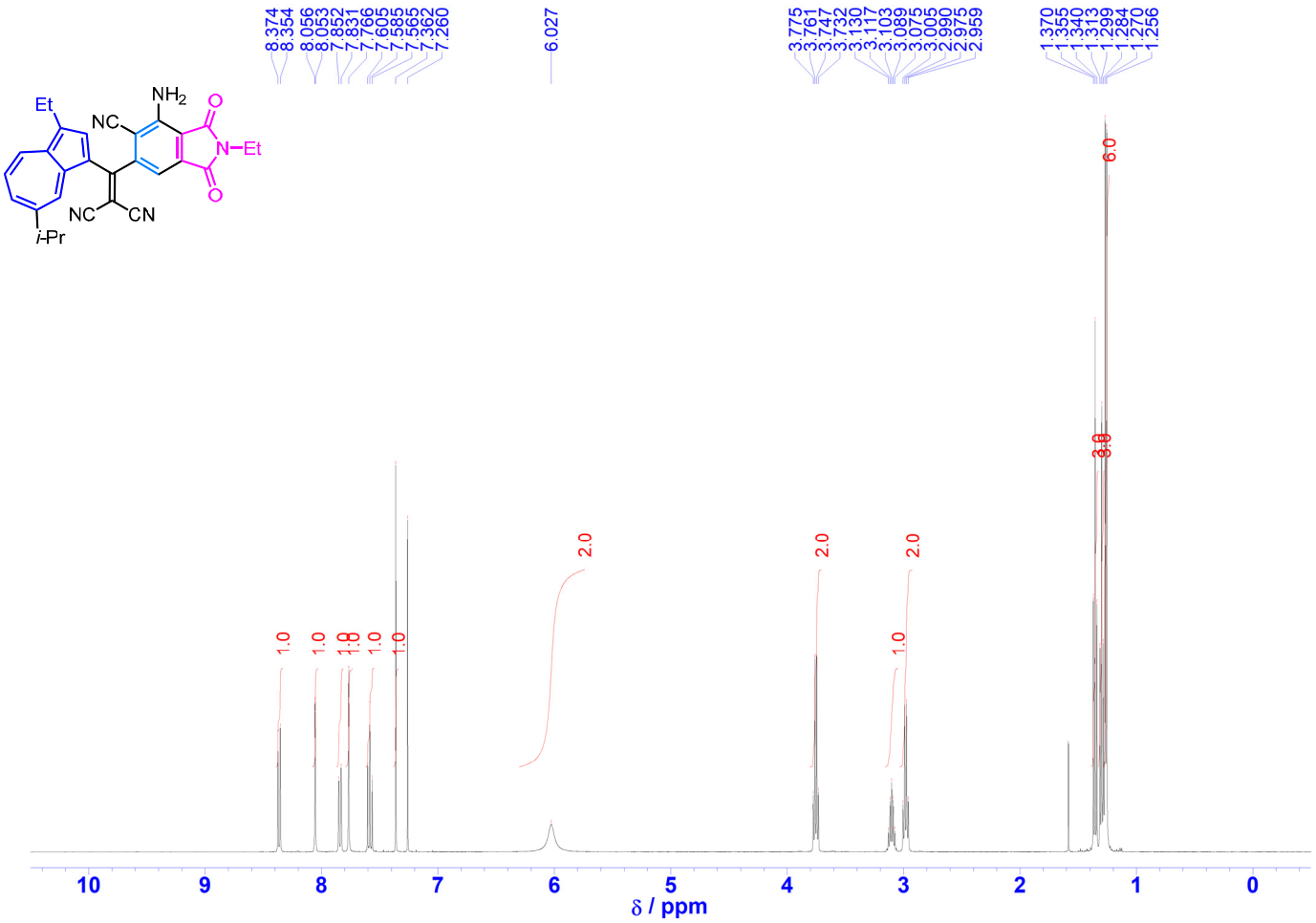


Figure S45. ^{1}H NMR spectrum of **4f** in $CDCl_3$ (500 MHz).

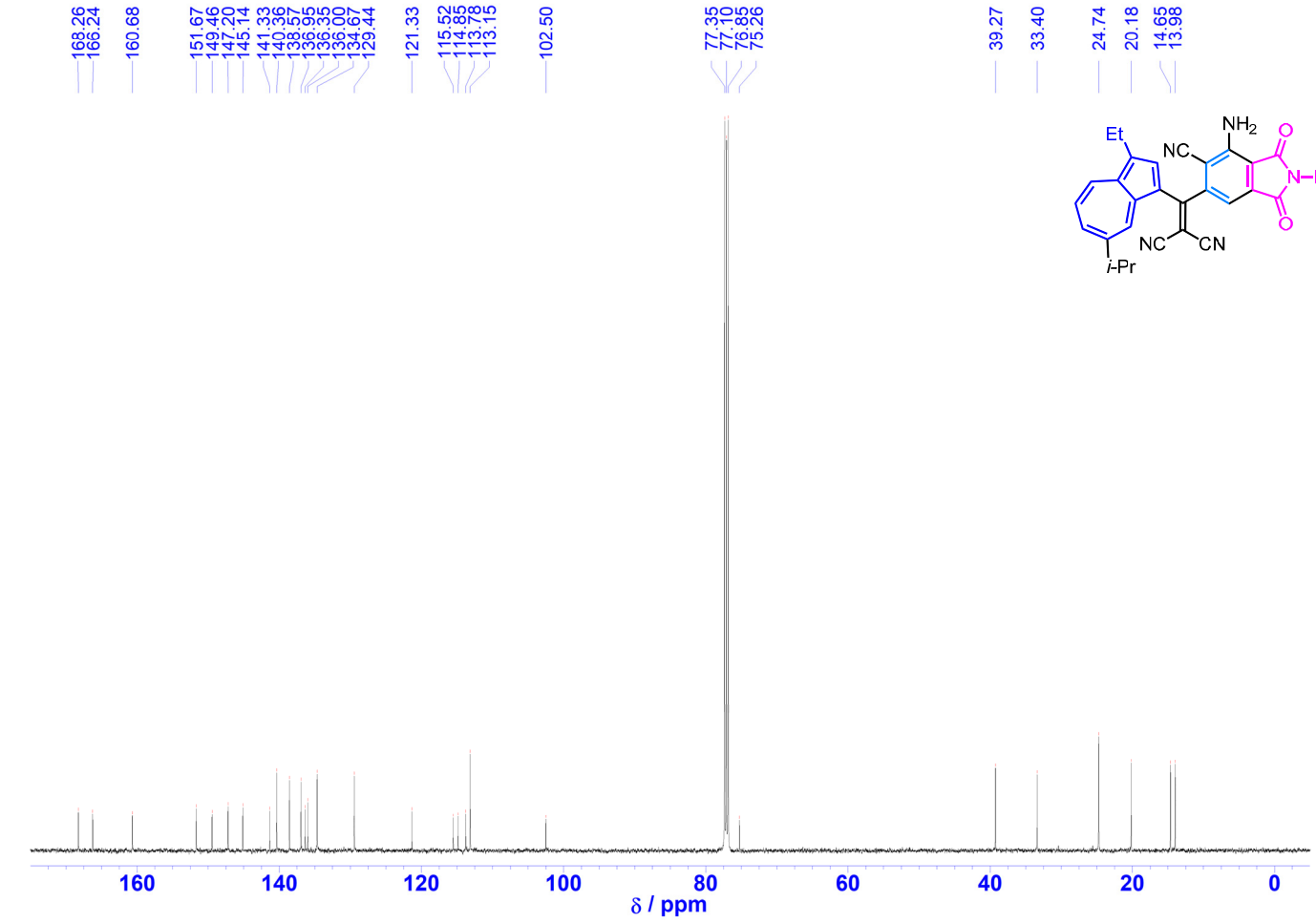


Figure S46. ^{13}C NMR spectrum of **4f** in $CDCl_3$ (125 MHz).

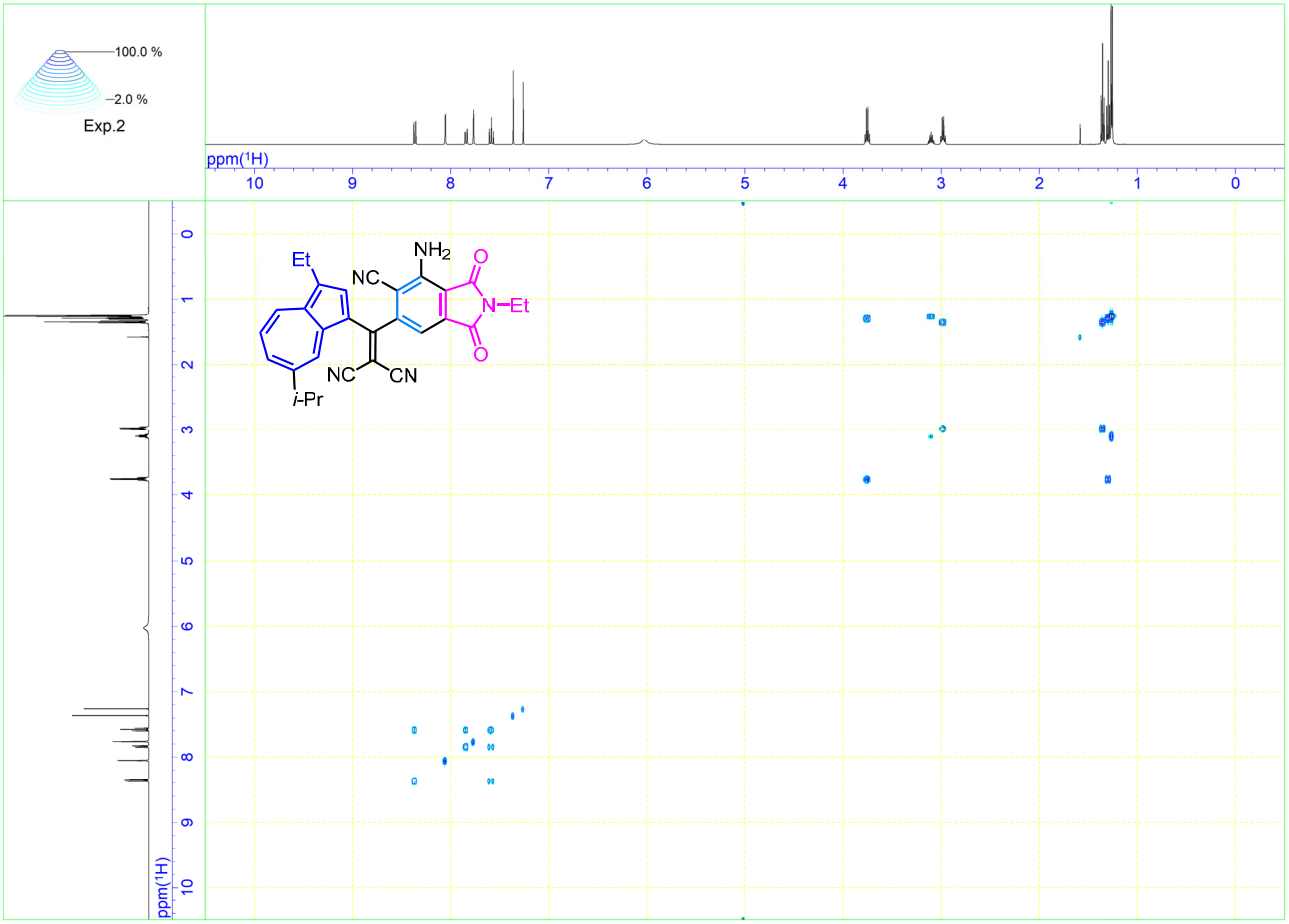


Figure S47. COSY spectrum of **4f** in $CDCl_3$ (500 MHz).

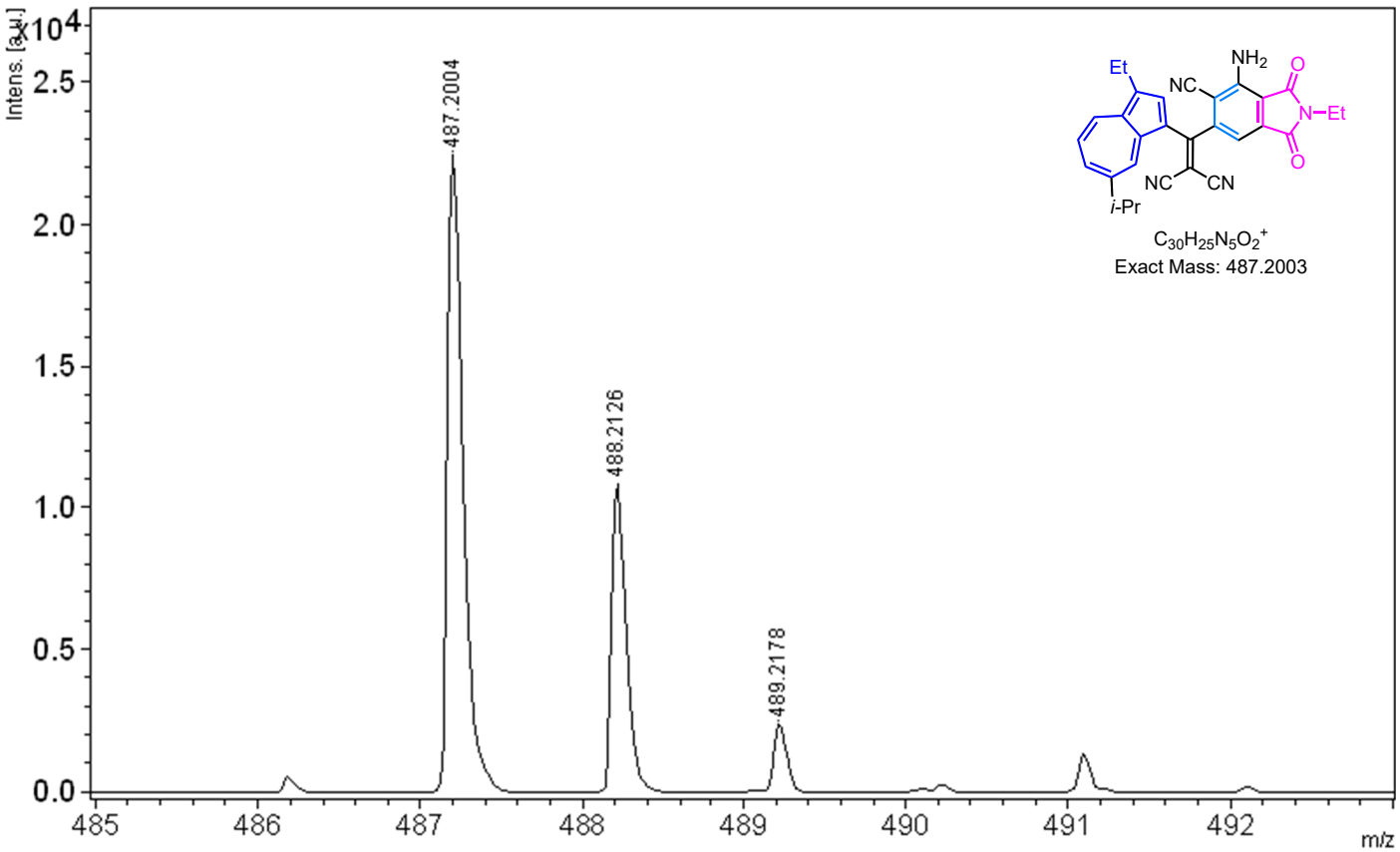


Figure S48. HRMS (MALDI-TOF, positive) of **4f**.

2. UV/Vis and fluorescent spectra of 2a−2e and 4a−4g (Figures S49−S86).

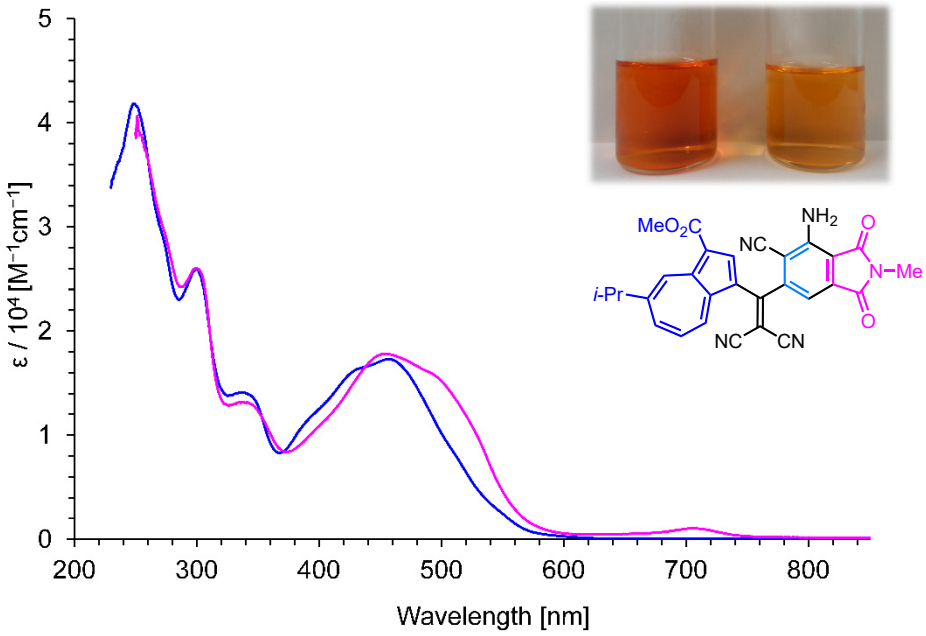


Figure S49. UV/Vis spectra of **2a** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

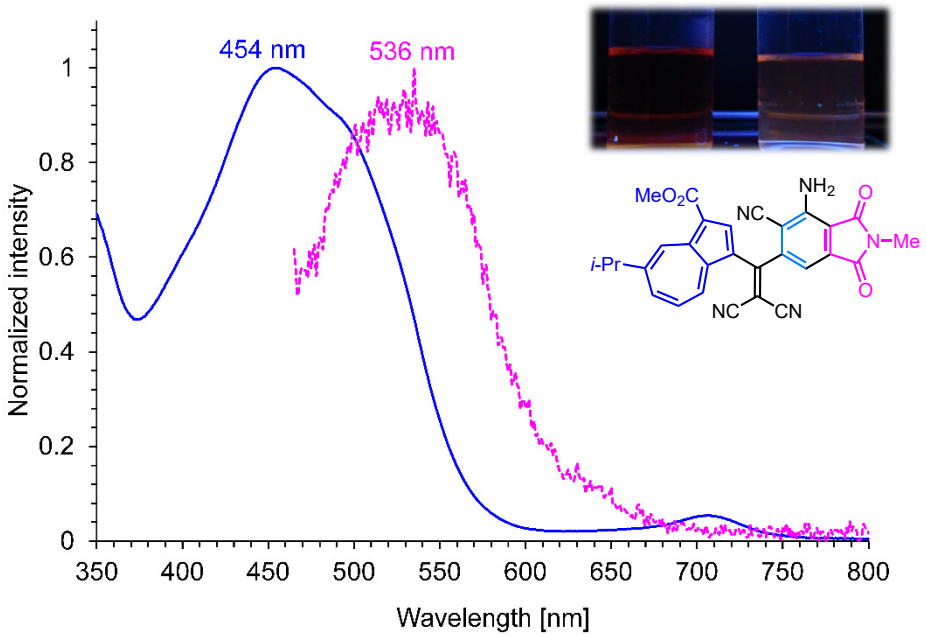


Figure S50. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **2a** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

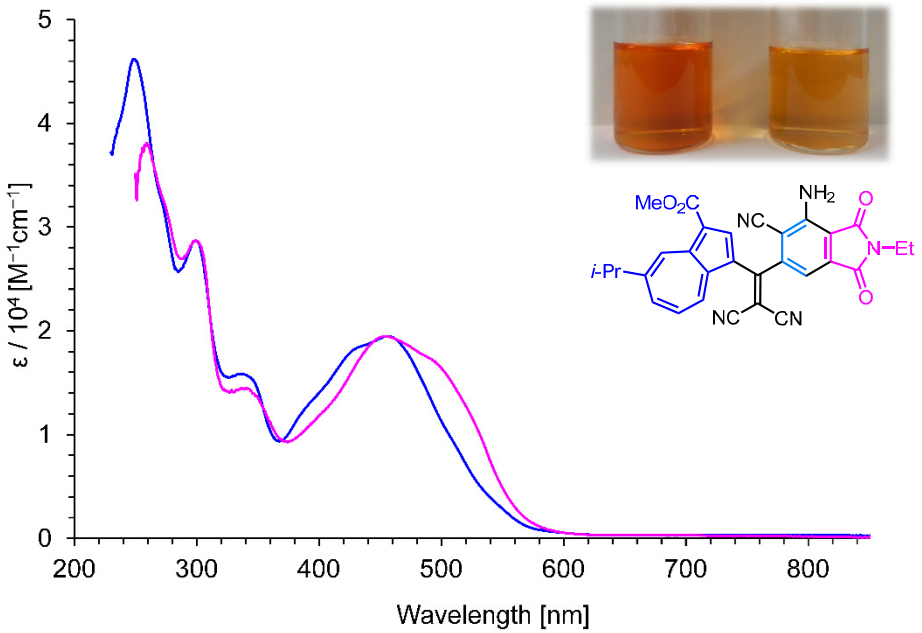


Figure S51. UV/Vis spectra of **2b** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

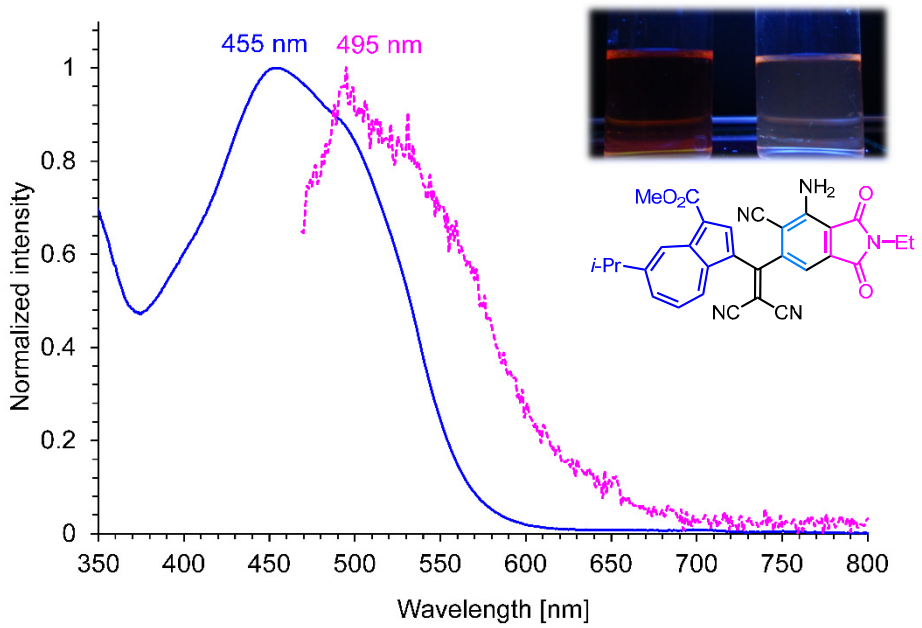


Figure S52. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **2b** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

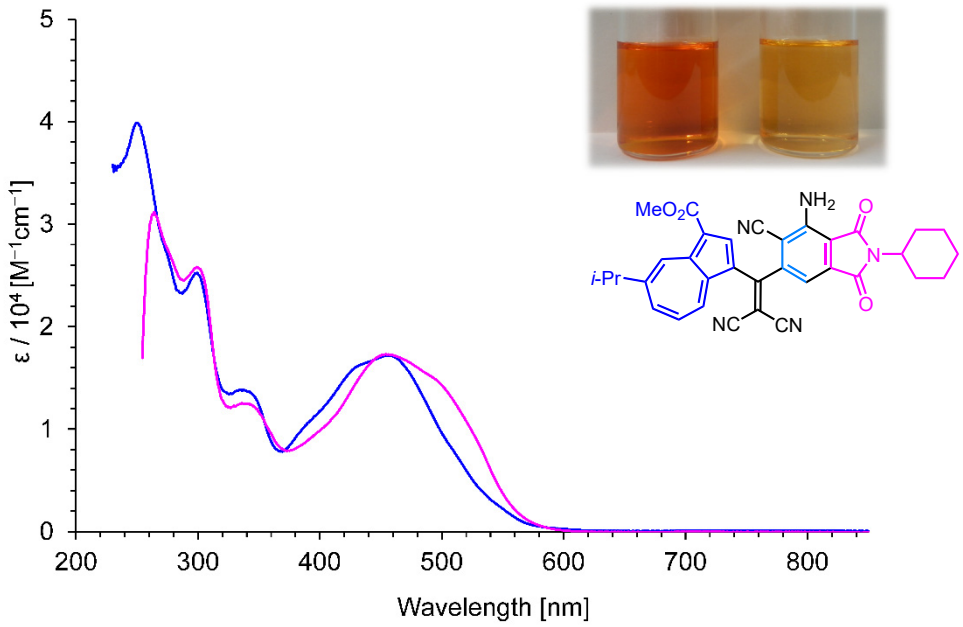


Figure S53. UV/Vis spectra of **2c** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

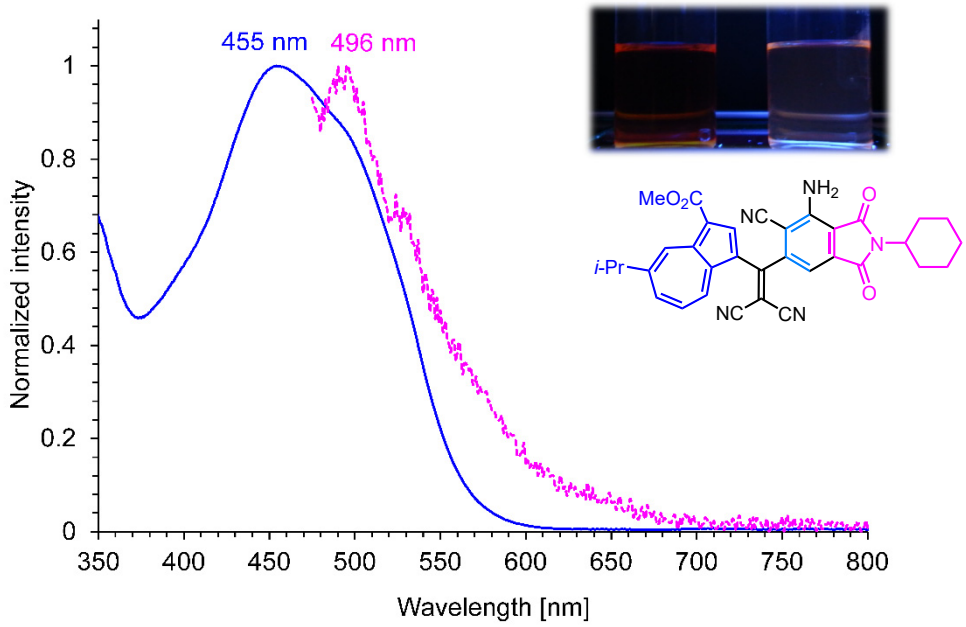


Figure S54. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **2c** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

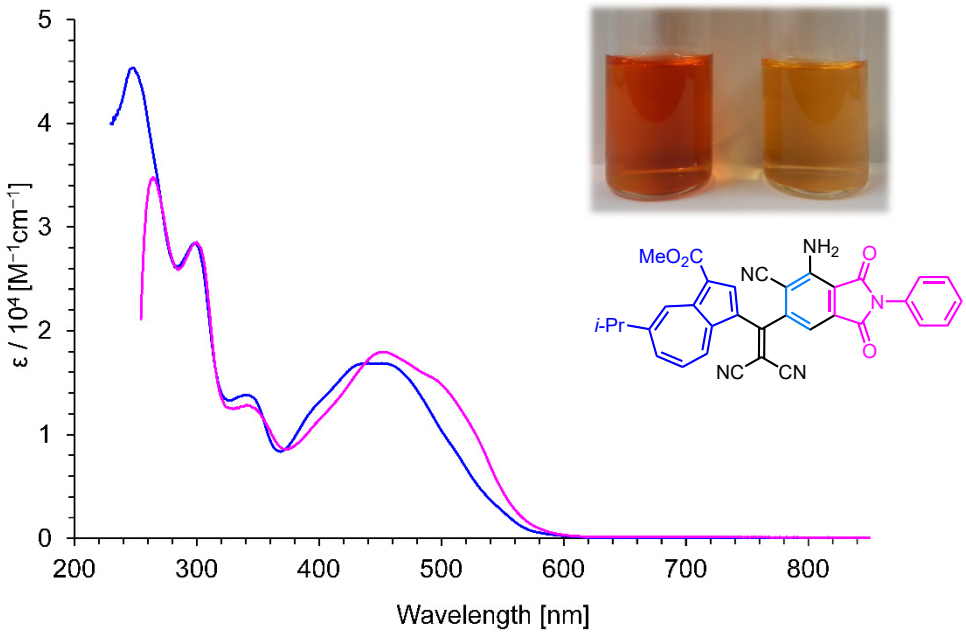


Figure S55. UV/Vis spectra of **2d** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

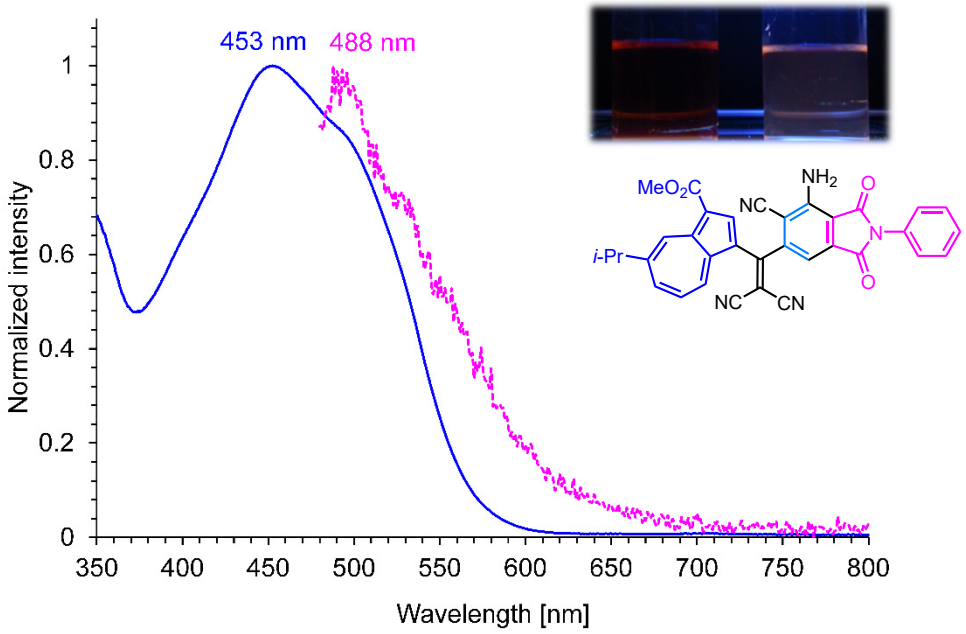


Figure S56. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **2d** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

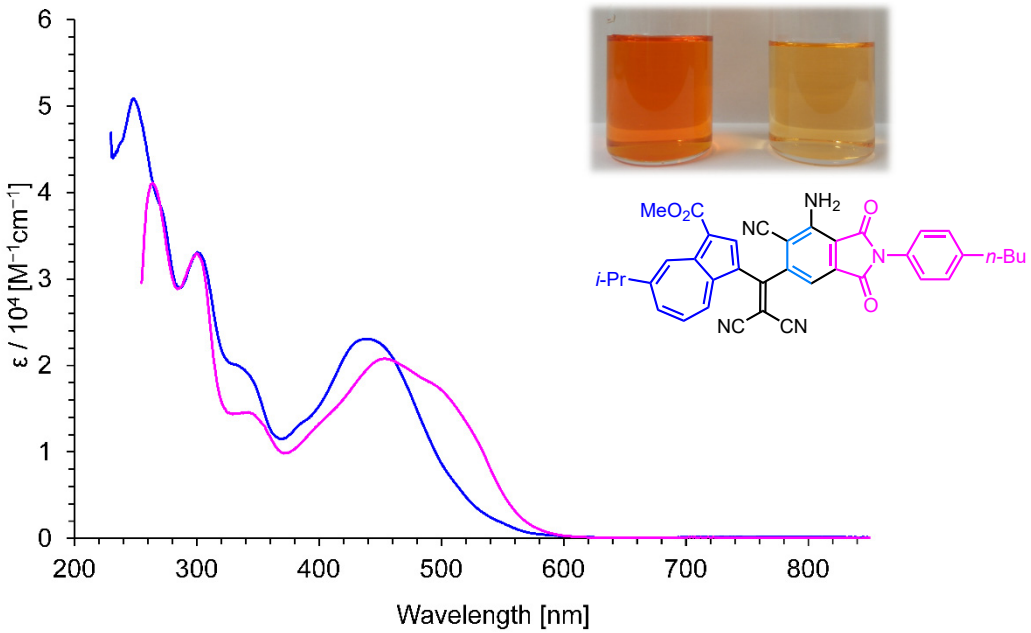


Figure S57. UV/Vis spectra of **2e** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

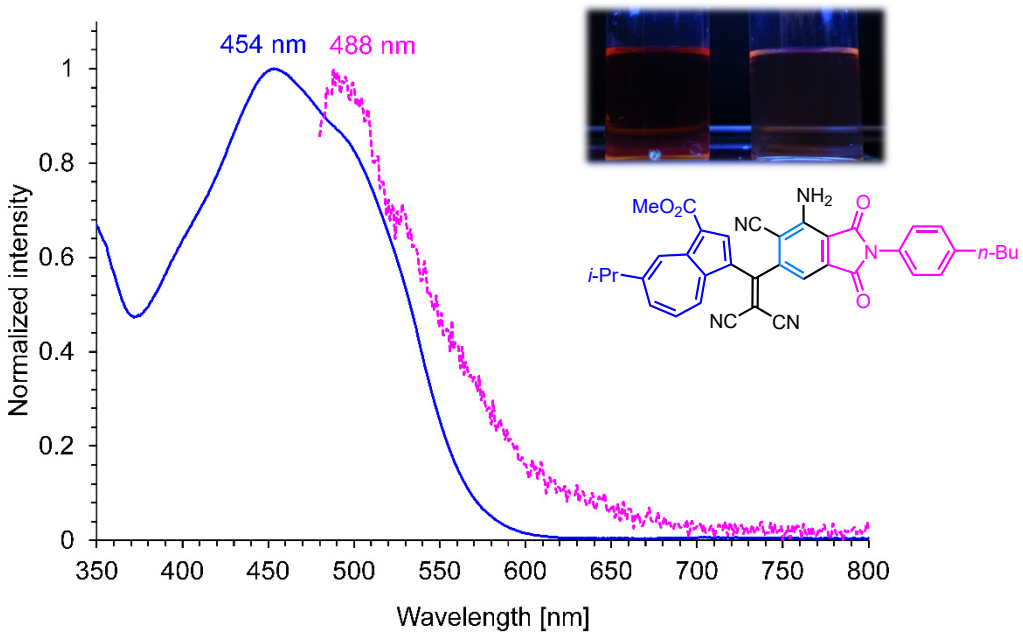


Figure S58. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **2e** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

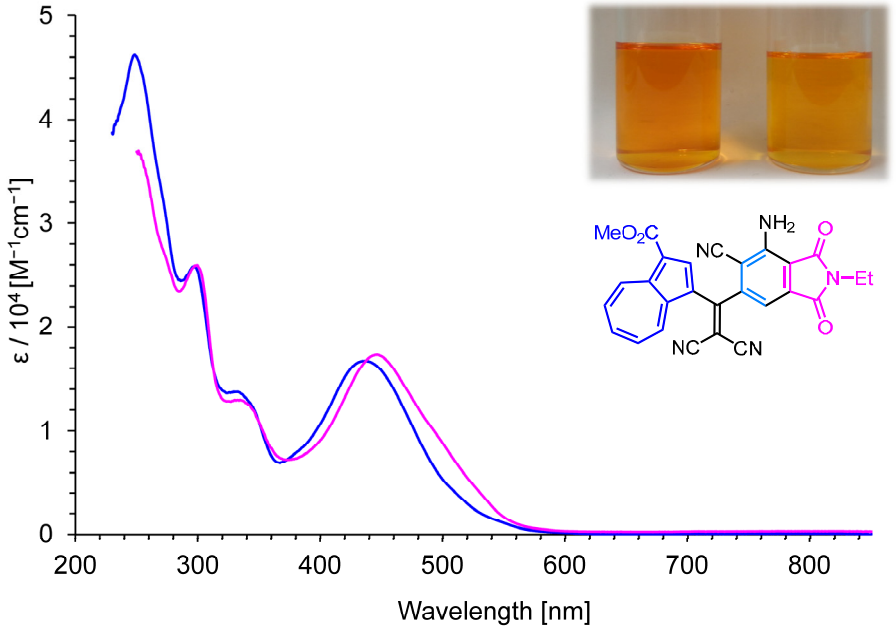


Figure S59. UV/Vis spectra of **4a** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

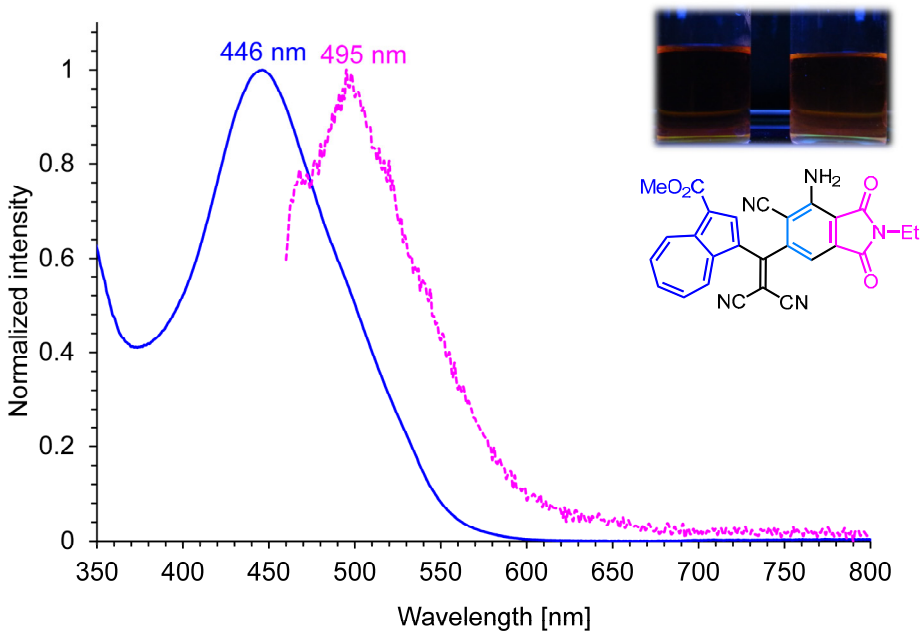


Figure S60. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4a** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

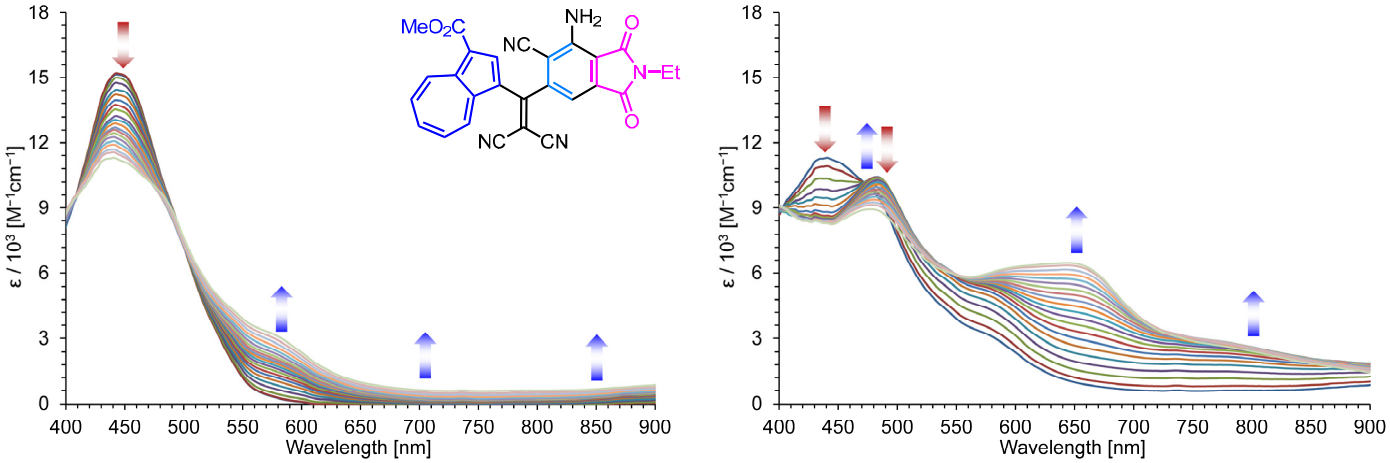


Figure S61. Continuous change in the visible spectrum of **4a**: constant-voltage electrochemical reduction at −1.20 V (left) and further reduction at −1.60 V (right) in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

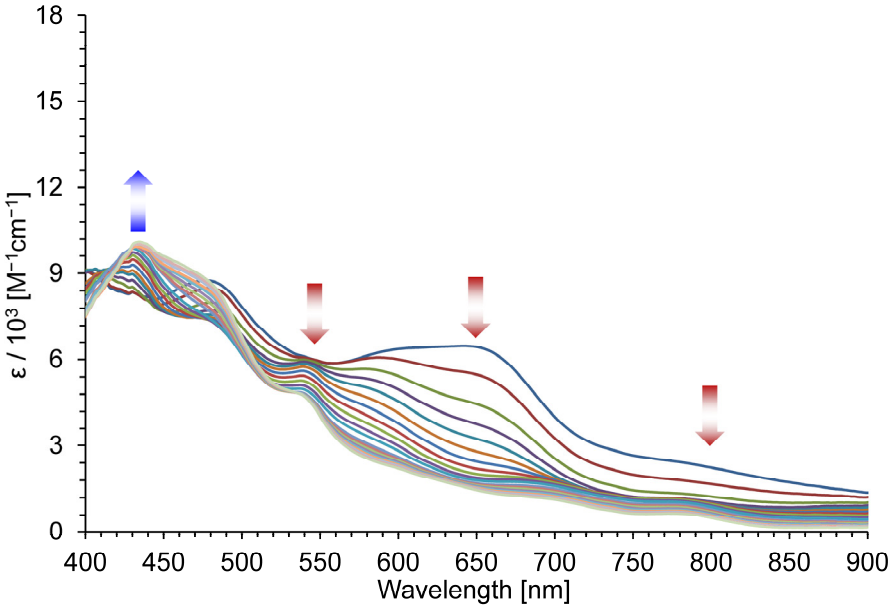


Figure S62. Continuous change in the visible spectrum of **4a**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

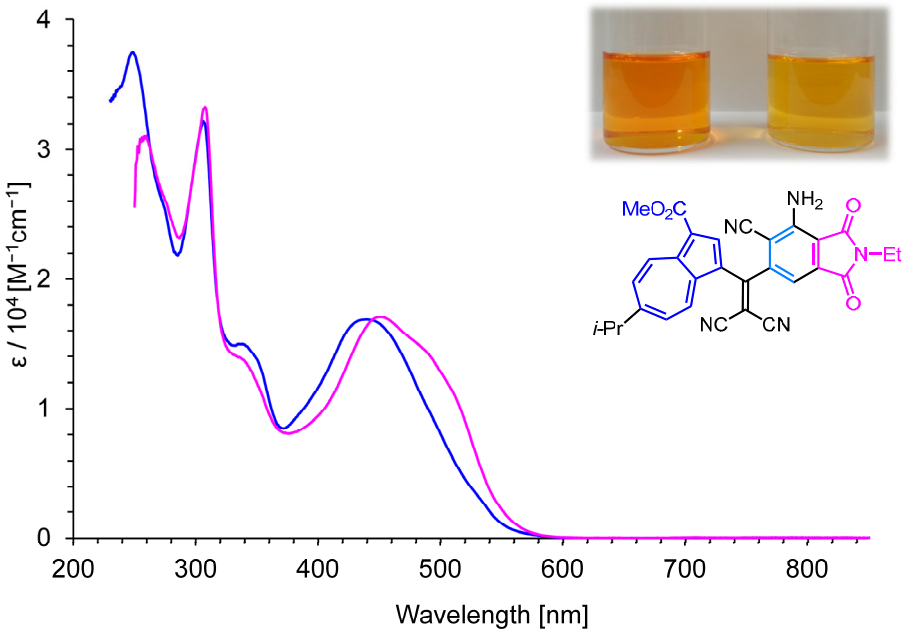


Figure S63. UV/Vis spectra of **4b** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

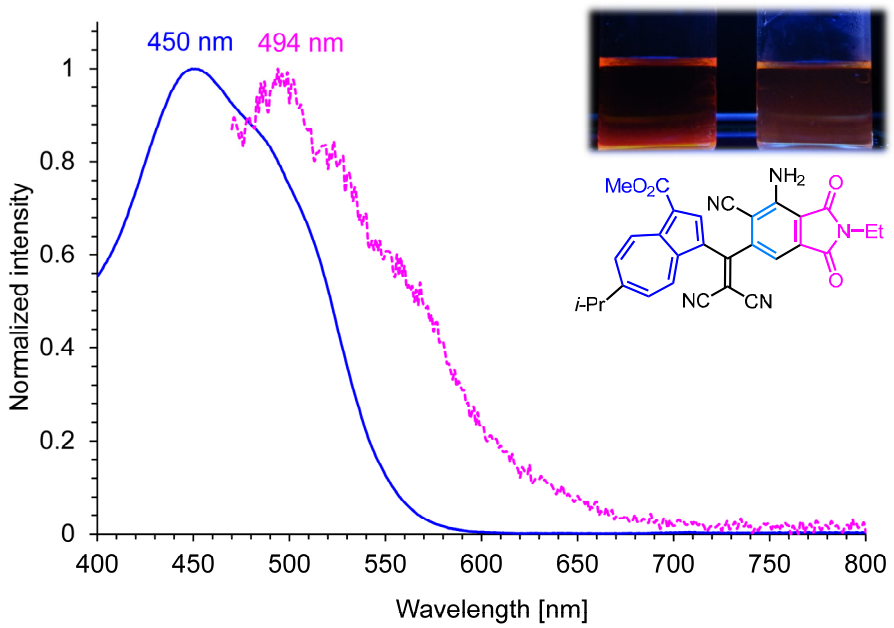


Figure S64. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4b** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

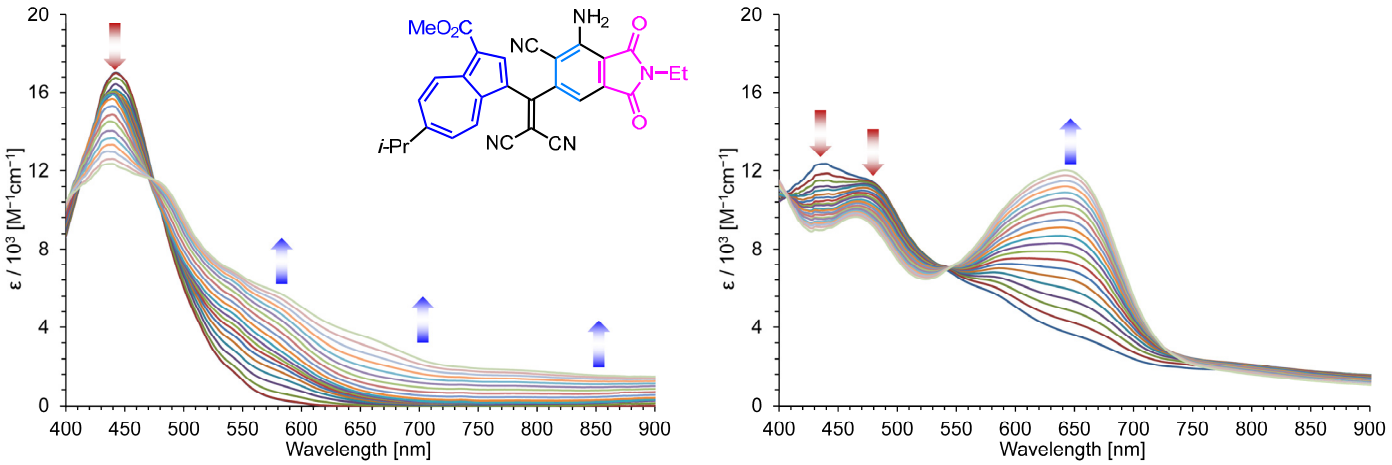


Figure S65. Continuous change in the visible spectrum of **4b**: constant-voltage electrochemical reduction at −1.15 V (left) and further reduction at −1.60 V (right) in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

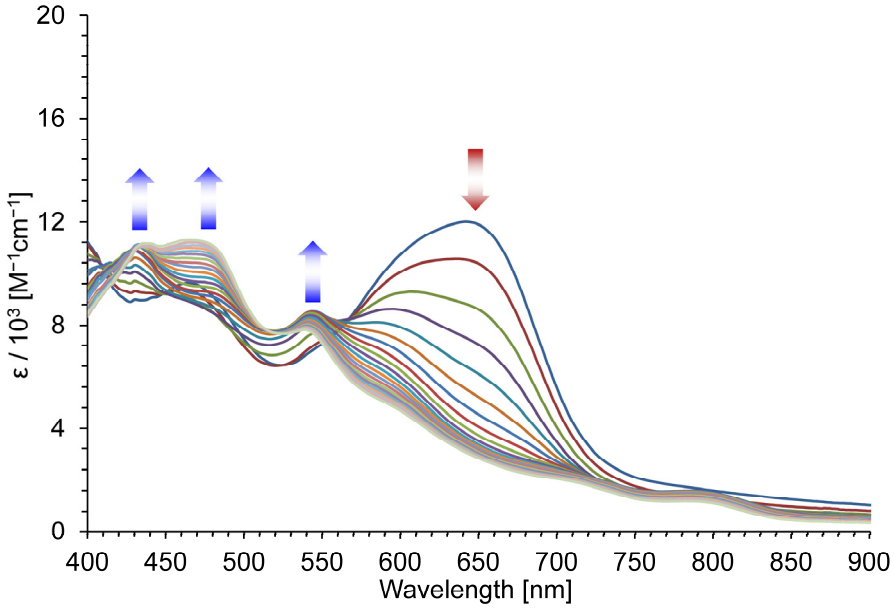


Figure S66. Continuous change in the visible spectrum of **4b**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

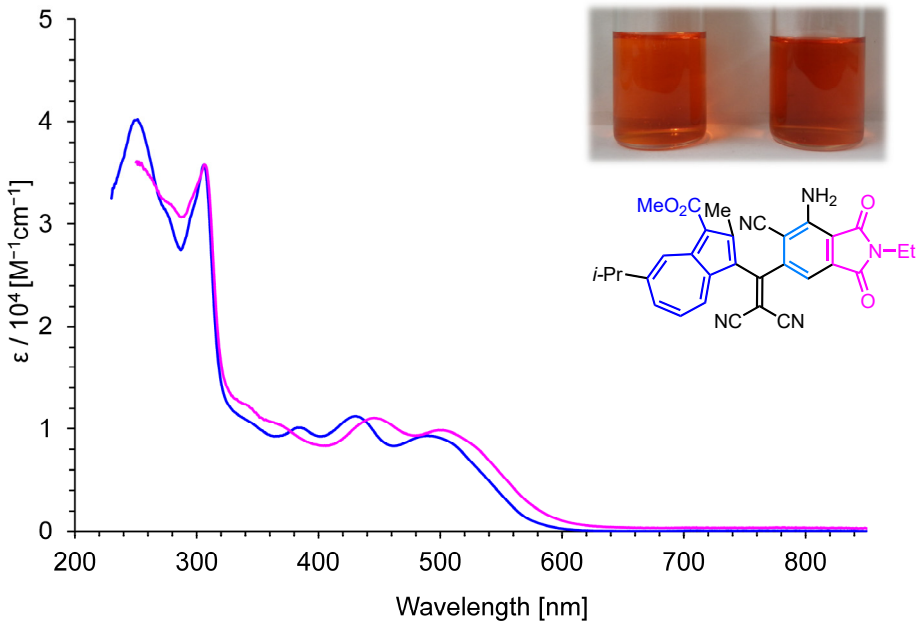


Figure S67. UV/Vis spectra of **4c** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

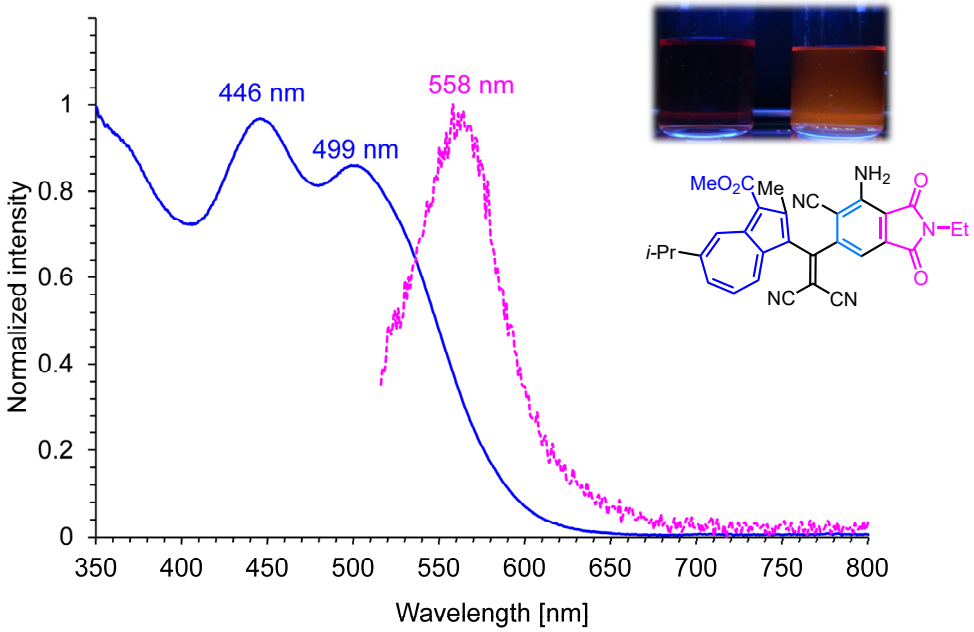


Figure S68. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4c** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

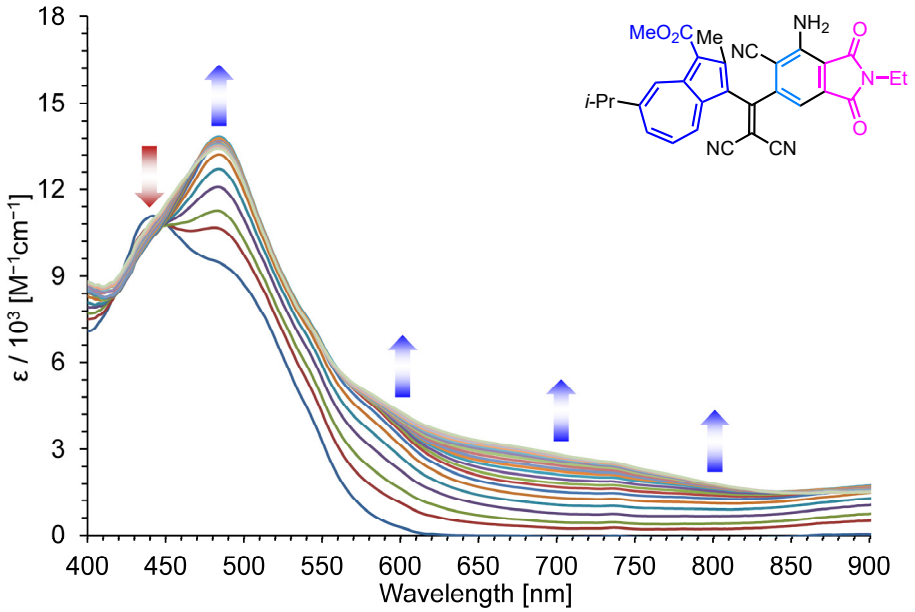


Figure S69. Continuous change in the visible spectrum of **4c**: constant-voltage electrochemical reduction at −1.60 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

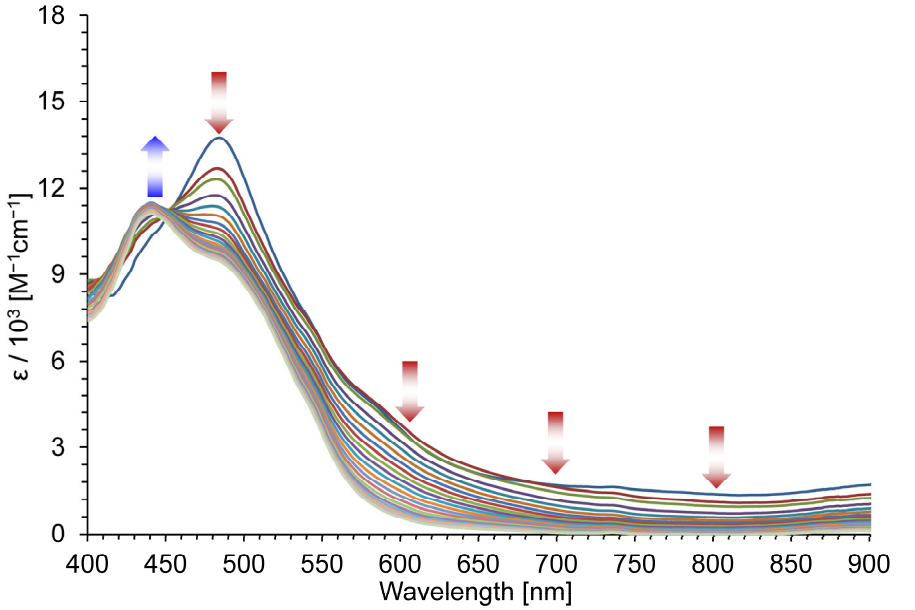


Figure S70. Continuous change in the visible spectrum of **4c**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

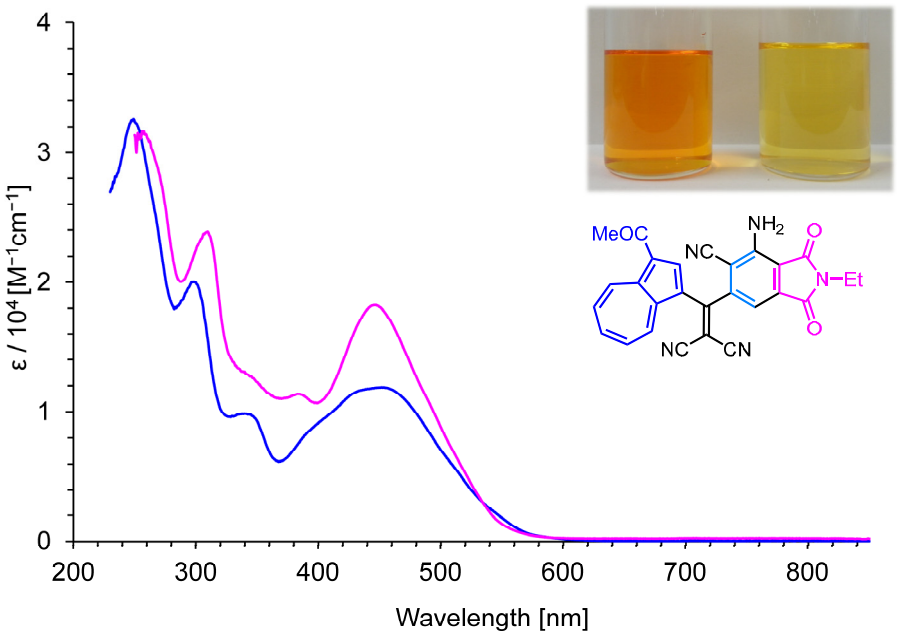


Figure S71. UV/Vis spectra of **4d** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

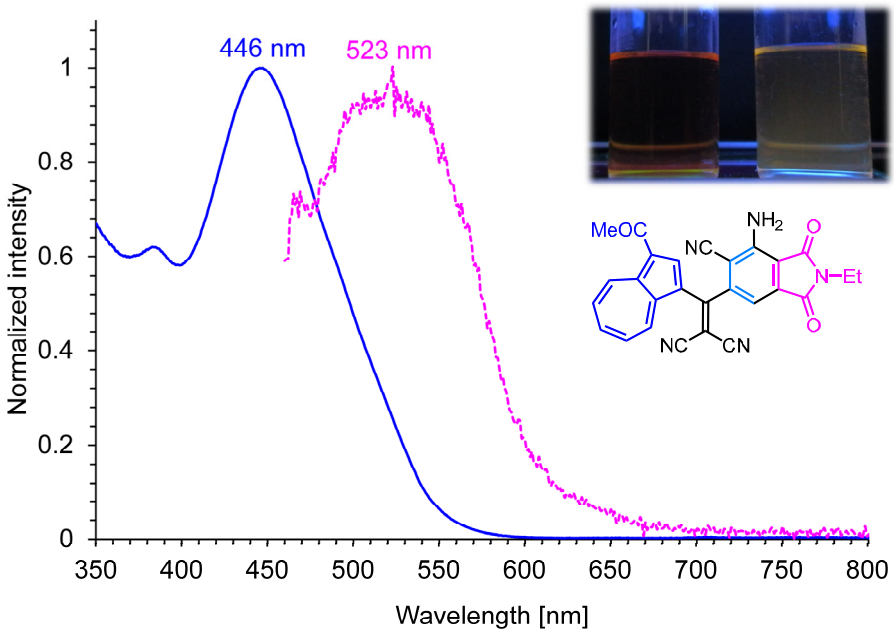


Figure S72. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4d** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

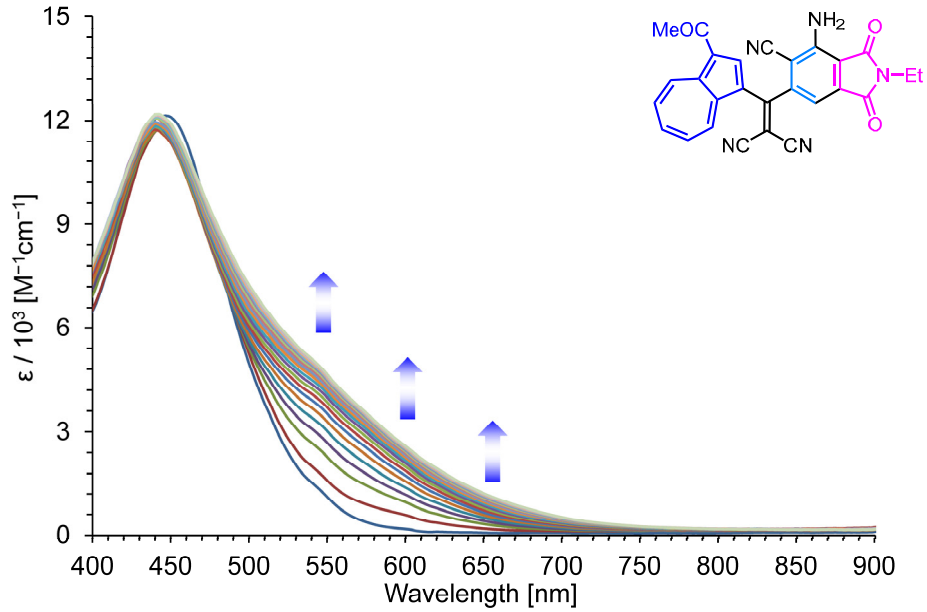


Figure S73. Continuous change in the visible spectrum of **4d**: constant-voltage electrochemical reduction at −2.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

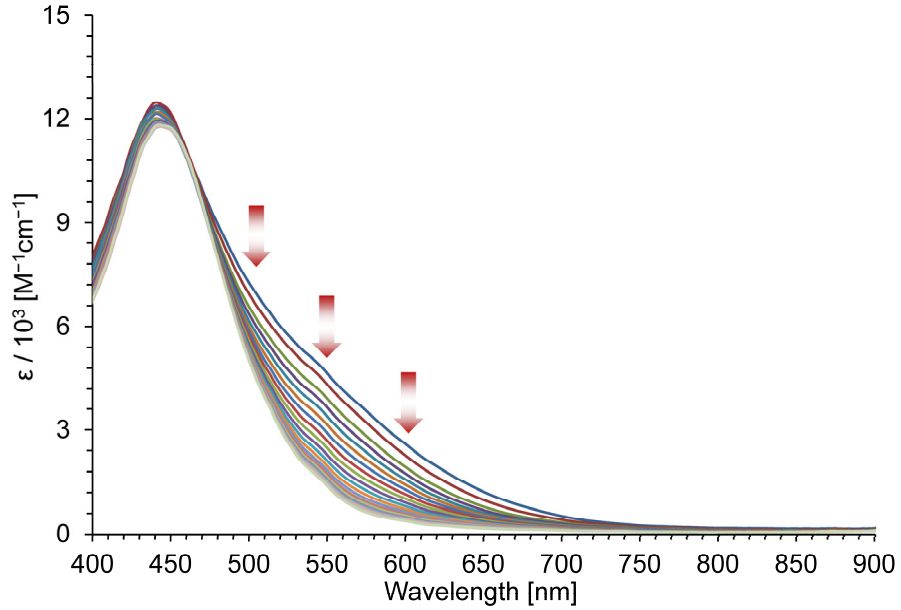


Figure S74. Continuous change in the visible spectrum of **4d**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

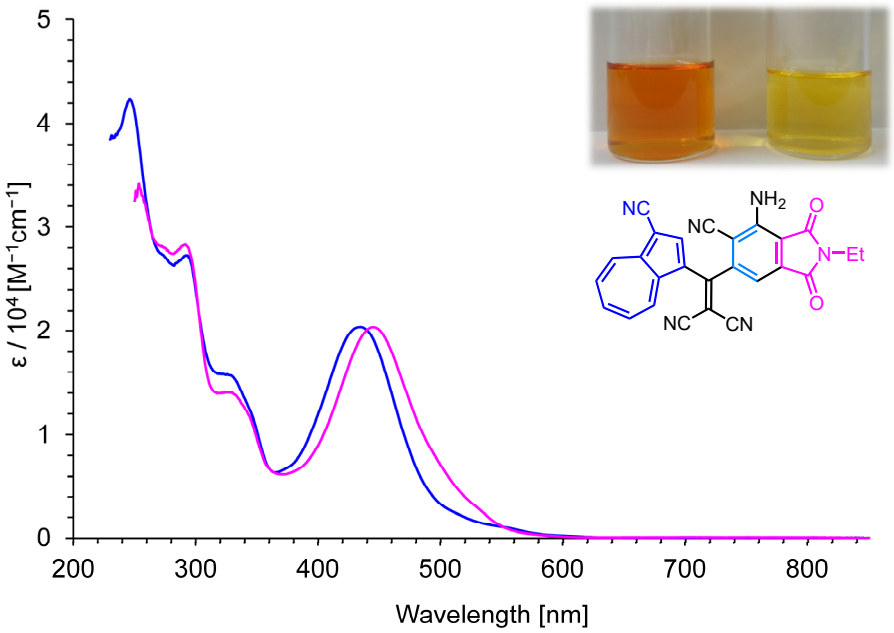


Figure S75. UV/Vis spectra of **4e** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

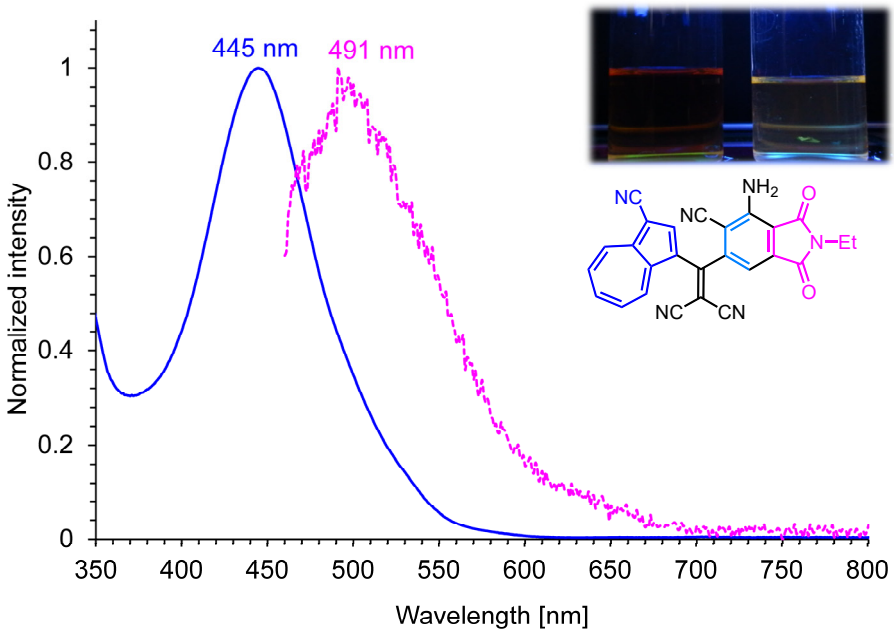


Figure S76. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4e** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

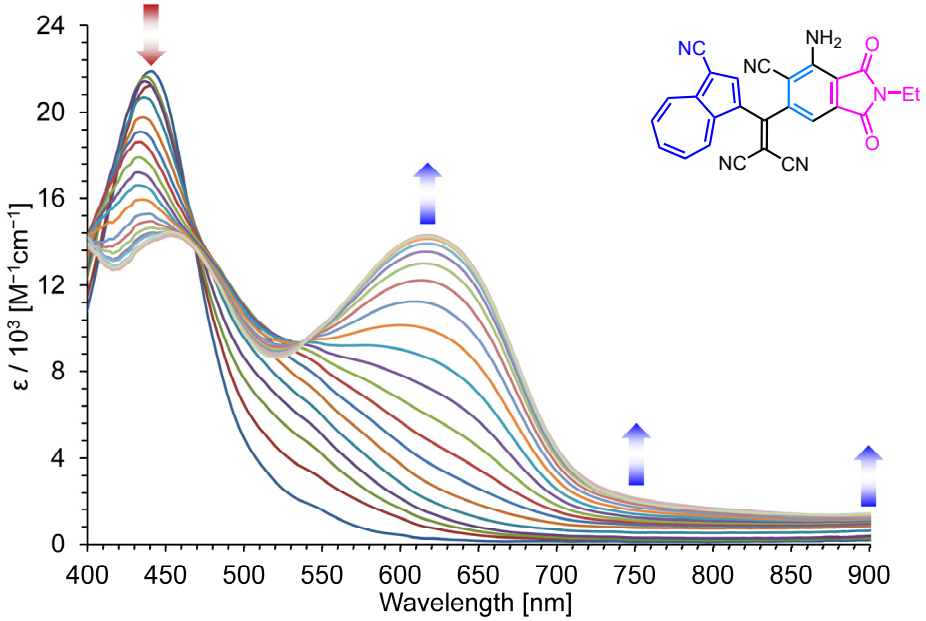


Figure S77. Continuous change in the visible spectrum of **4e**: constant-voltage electrochemical reduction at −2.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

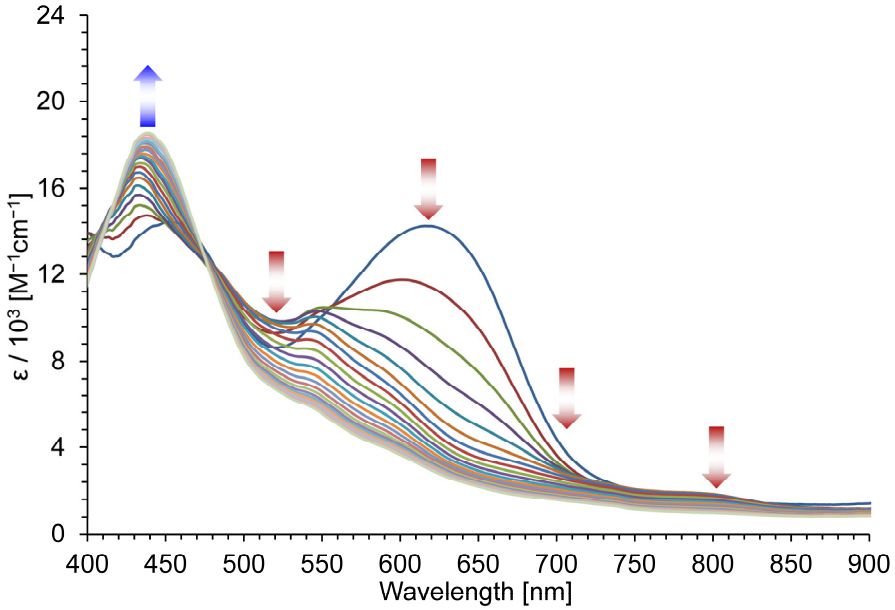


Figure S78. Continuous change in the visible spectrum of **4e**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

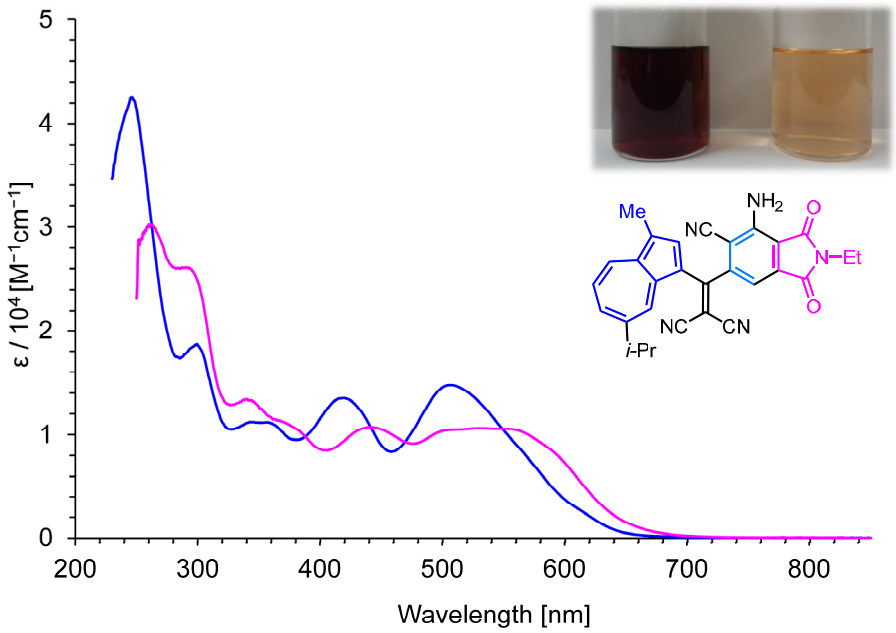


Figure S79. UV/Vis spectra of **4f** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

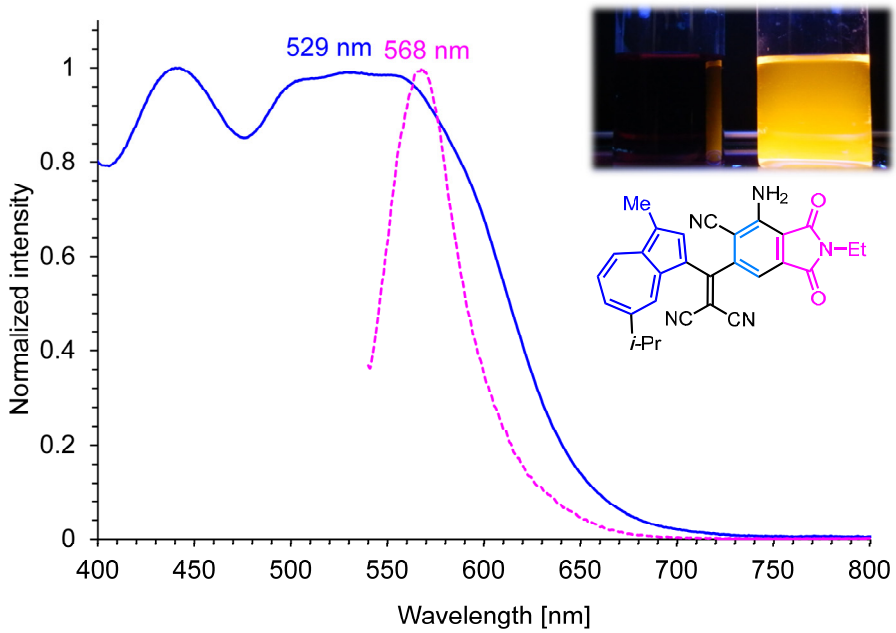


Figure S80. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4f** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

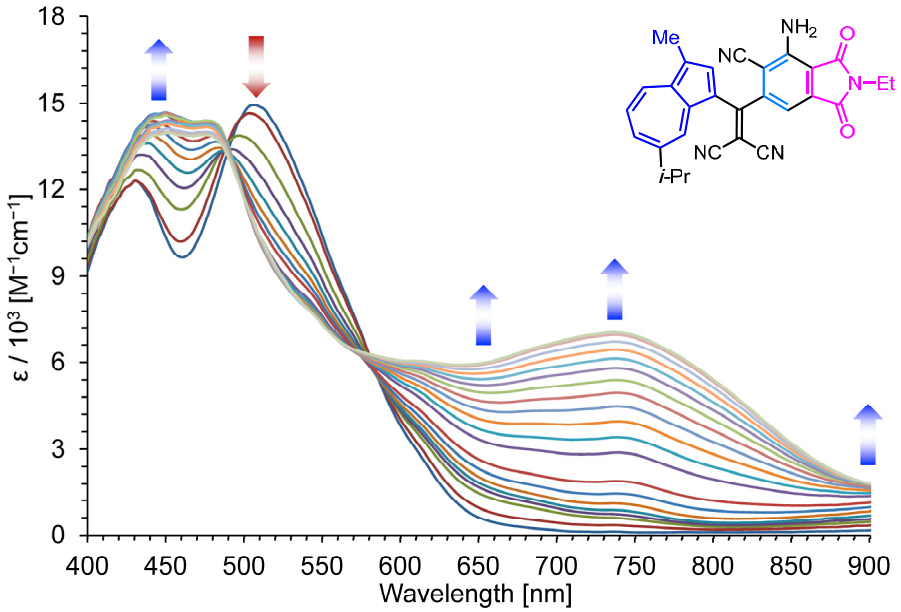


Figure S81. Continuous change in the visible spectrum of **4f**: constant-voltage electrochemical reduction at −2.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

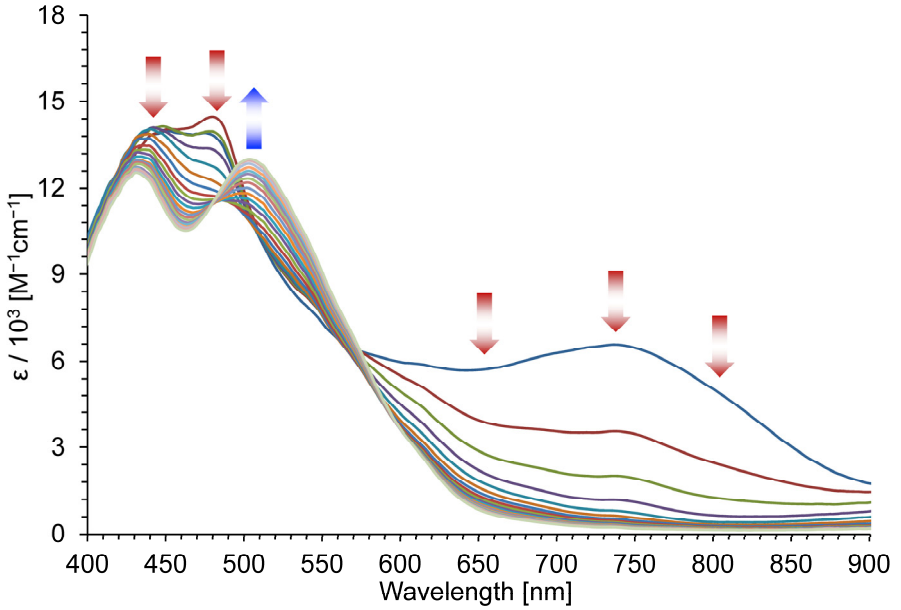


Figure S82. Continuous change in the visible spectrum of **4f**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

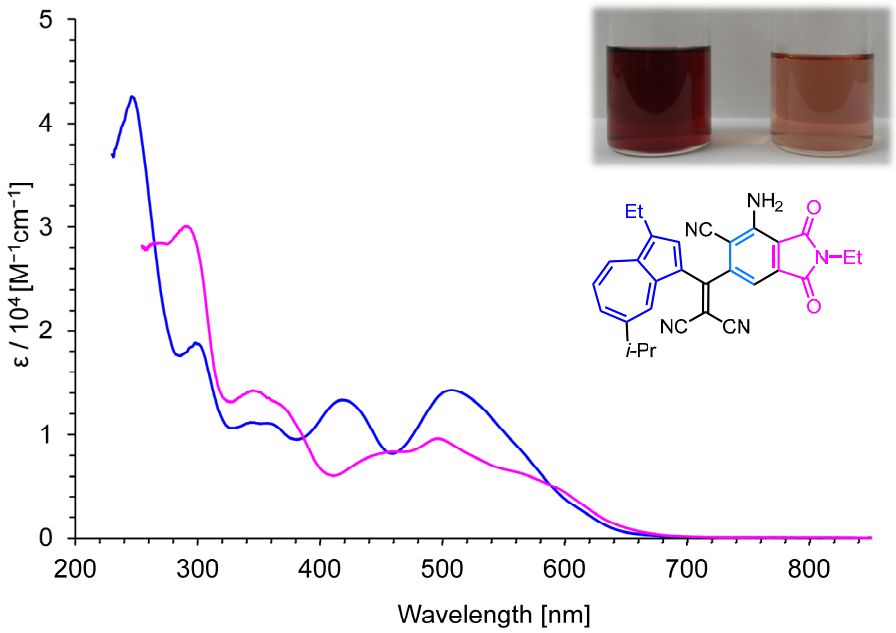


Figure S83. UV/Vis spectra of **4g** in CH_2Cl_2 (blue line) and in 30% CF_3CO_2H/ CH_2Cl_2 (pink line); photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right).

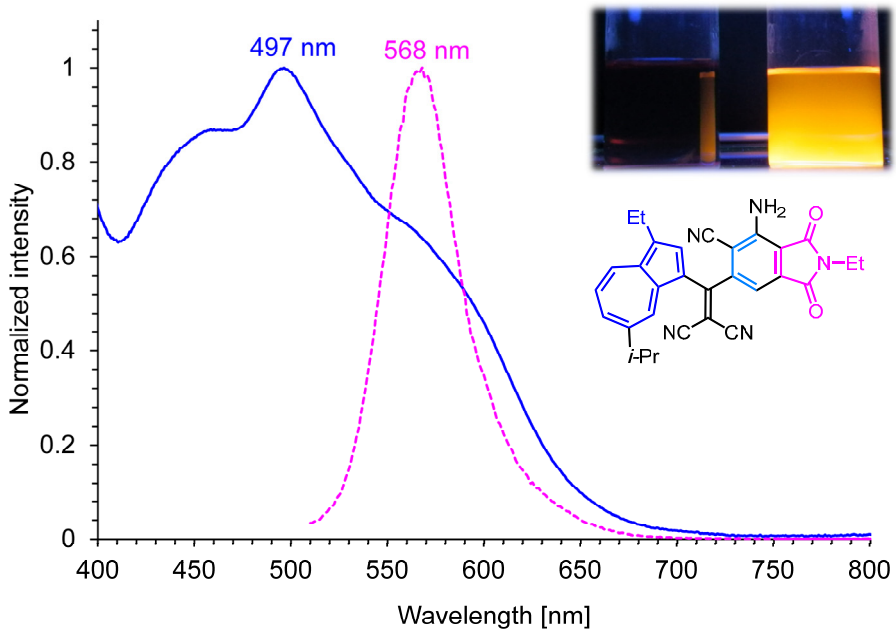


Figure S84. UV/Vis spectrum (blue line) and fluorescent spectrum (pink dotted-line) of **4g** in 30% CF_3CO_2H/ CH_2Cl_2; photo: in CH_2Cl_2 (left) and in 30% CF_3CO_2H/ CH_2Cl_2 (right) under the UV-light irradiation at λ_{ex} = 365 nm.

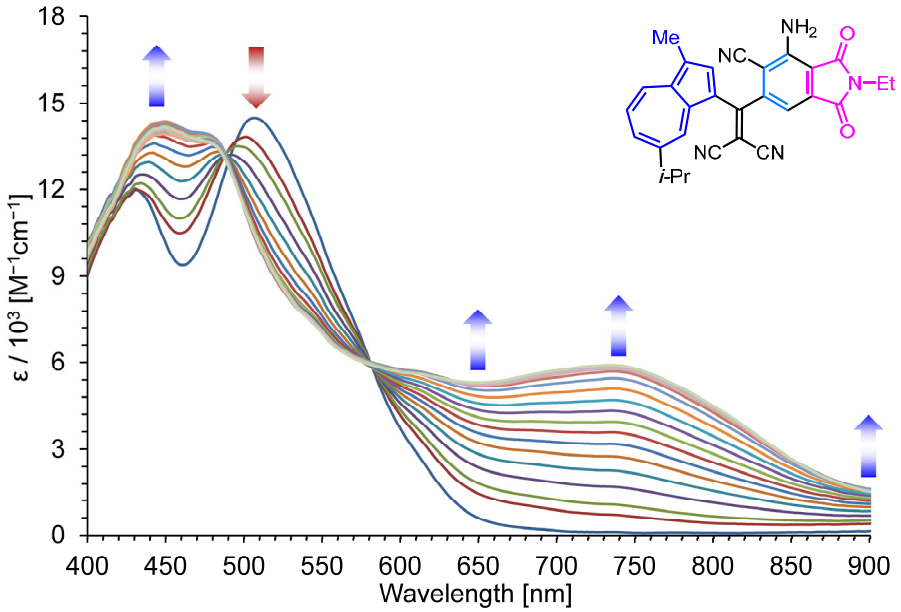


Figure S85. Continuous change in the visible spectrum of **4g**: constant-voltage electrochemical reduction at −2.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

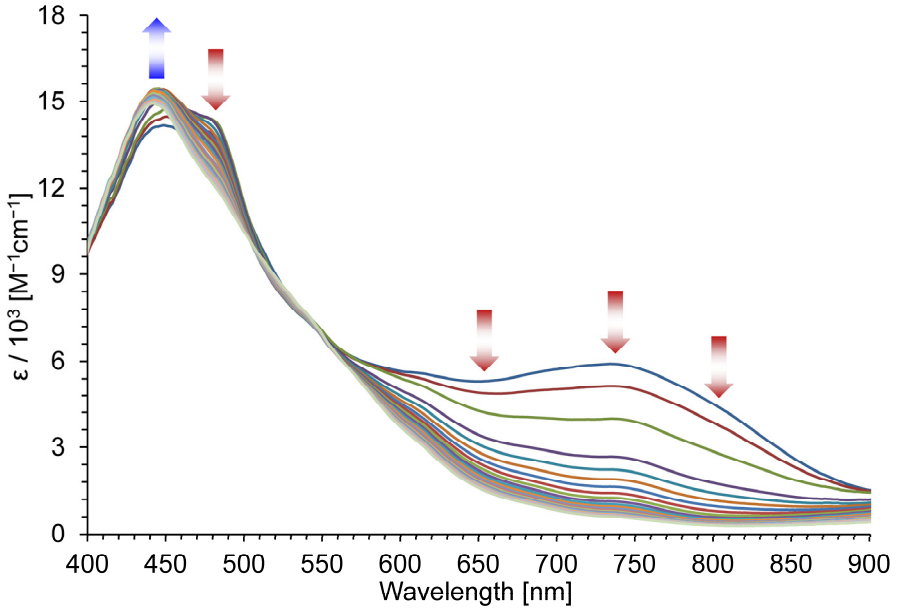


Figure S86. Continuous change in the visible spectrum of **4g**: constant-voltage electrochemical oxidation of the reduced species at ±0.00 V in benzonitrile containing Et_4NClO_4 (0.1 M) at 20 sec intervals.

3. Frontier Kohn–Sham orbitals of compounds 2a−2e and 4a−4g (Figures S87−S98).

HOMO (−6.25 eV) LUMO (−3.37eV)

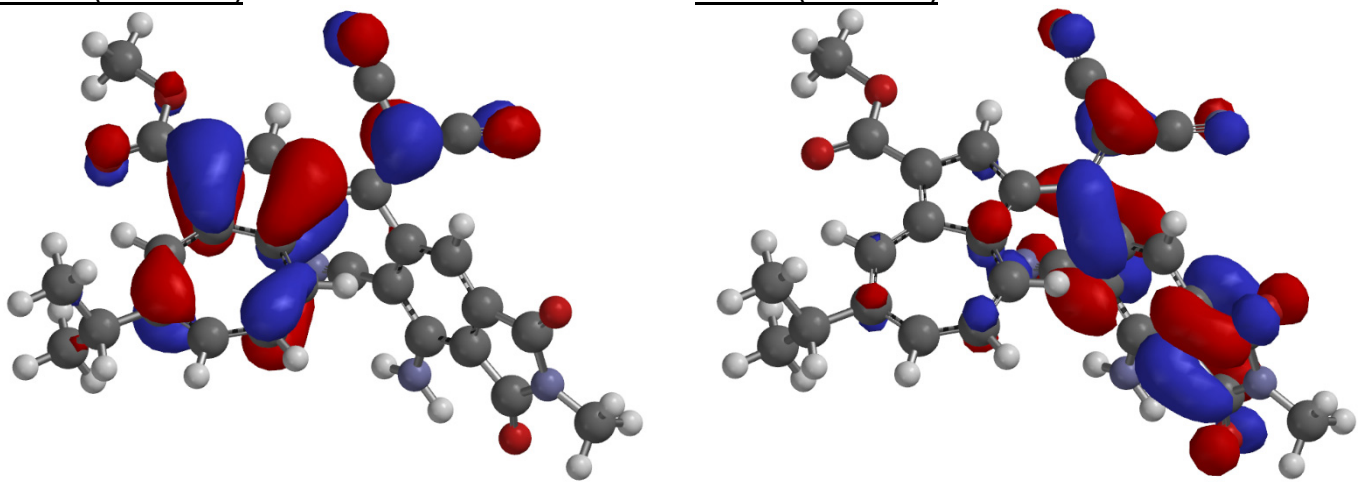


Figure S87. Frontier Kohn–Sham orbitals of TCBD **2a** at the B3LYP/6-311G* level.

HOMO (−6.24 eV) LUMO (−3.36 eV)

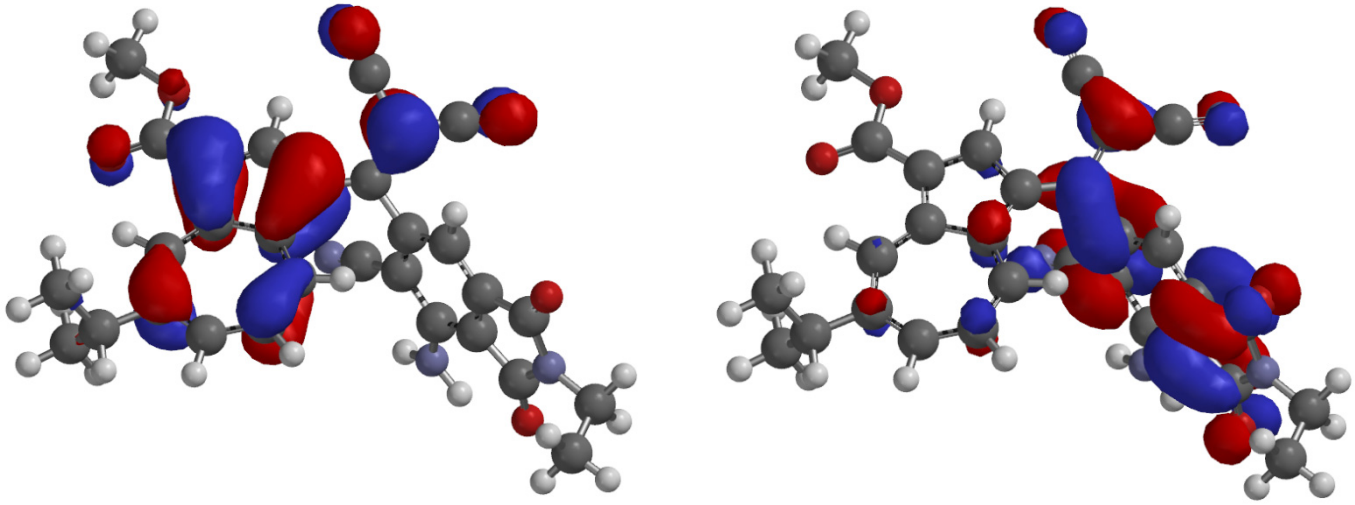


Figure S88. Frontier Kohn–Sham orbitals of TCBD **2b** at the B3LYP/6-311G* level.

HOMO (−6.22 eV) LUMO (−3.33 eV)

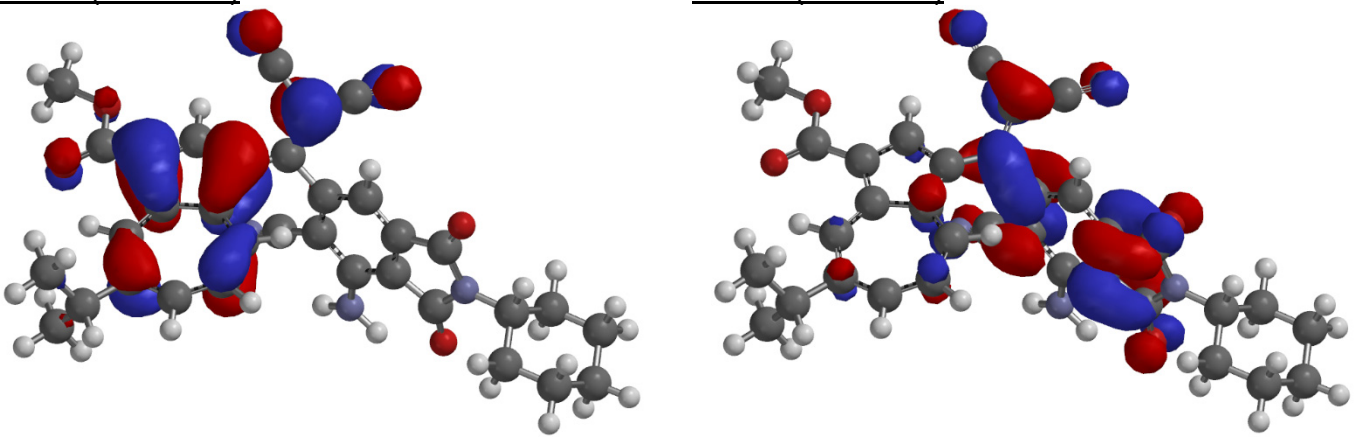
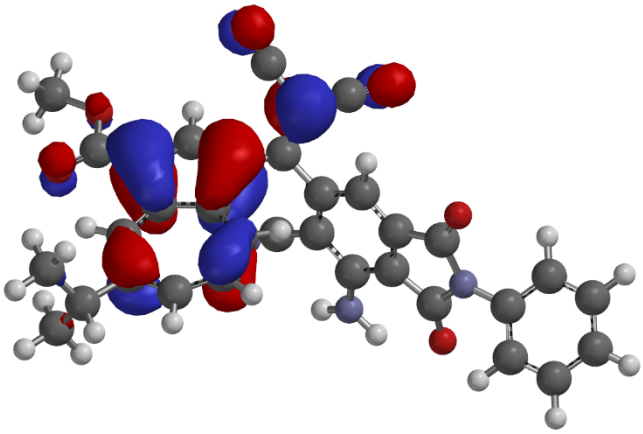


Figure S89. Frontier Kohn–Sham orbitals of TCBD **2c** at the B3LYP/6-311G* level.

HOMO (−6.26 eV)



LUMO (−3.40 eV)

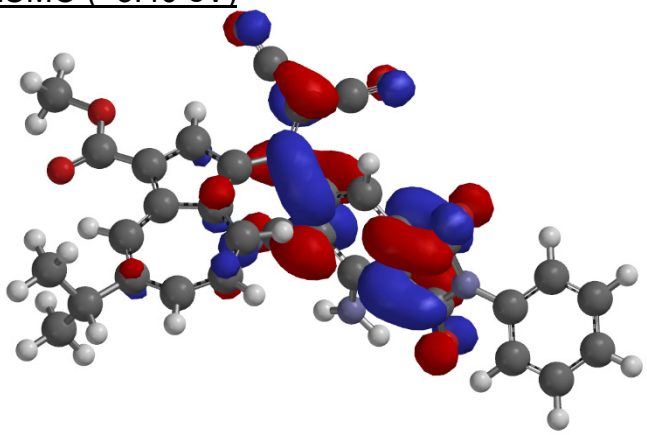
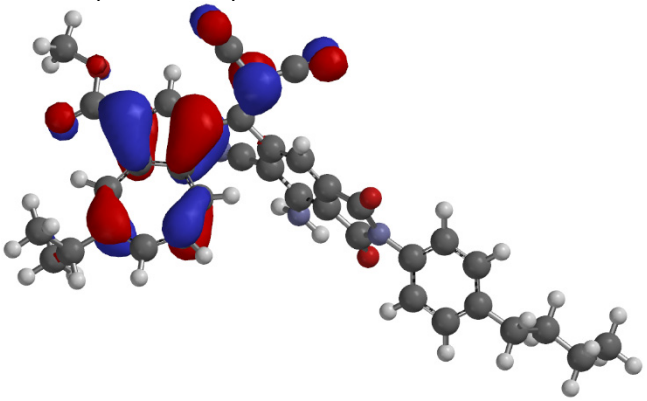


Figure S90. Frontier Kohn–Sham orbitals of TCBD **2d** at the B3LYP/6-311G* level.

HOMO (−6.24 eV)



LUMO (−3.36 eV)

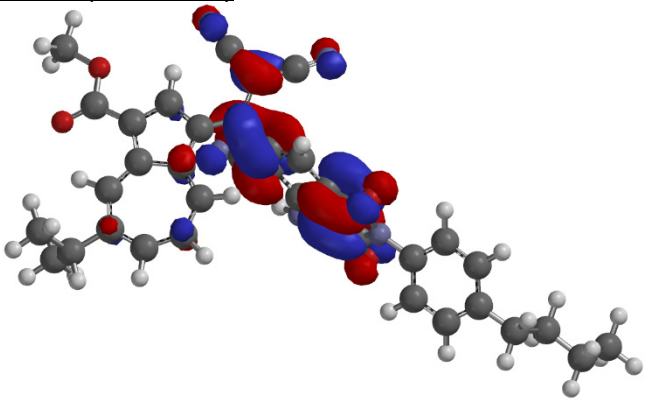
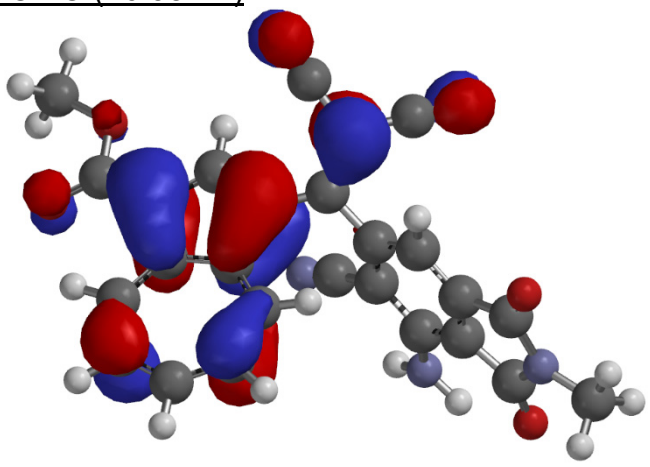


Figure S91. Frontier Kohn–Sham orbitals of TCBD **2e** at the B3LYP/6-311G* level.

HOMO (−6.38 eV)



LUMO (−3.43 eV)

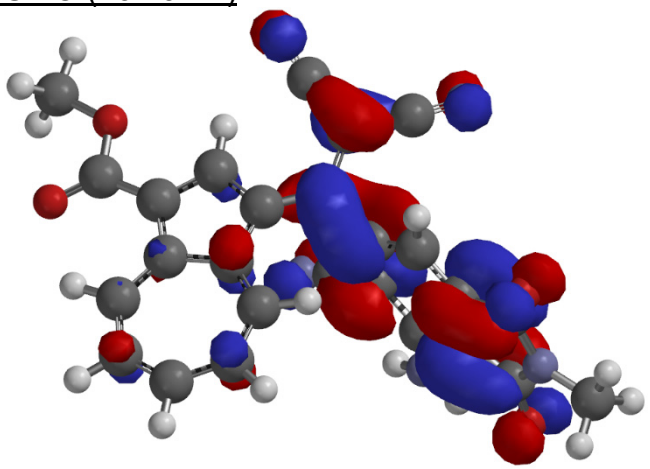
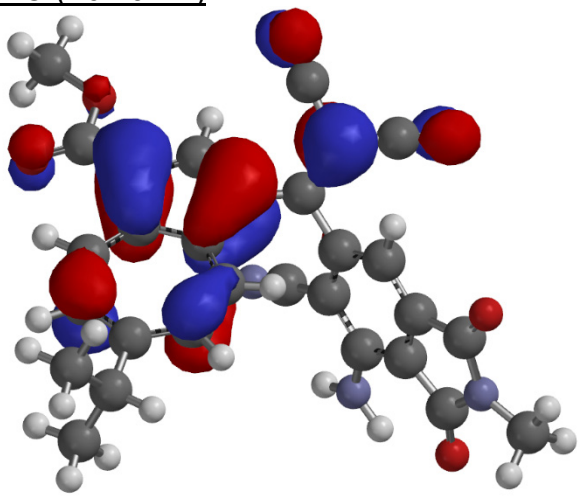


Figure S92. Frontier Kohn–Sham orbitals of TCBD **4a** at the B3LYP/6-311G* level.

HOMO (−6.29 eV)



LUMO (−3.37 eV)

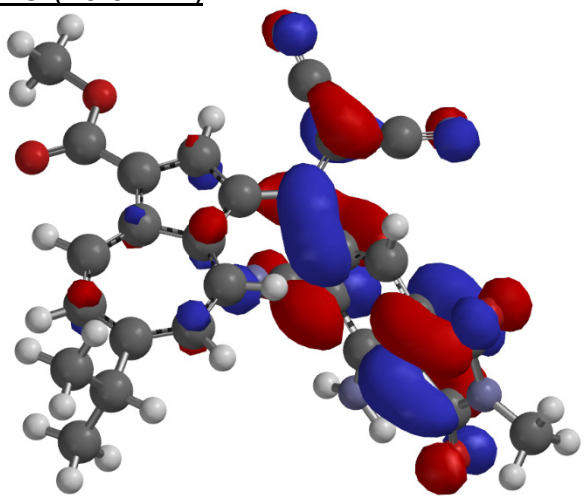
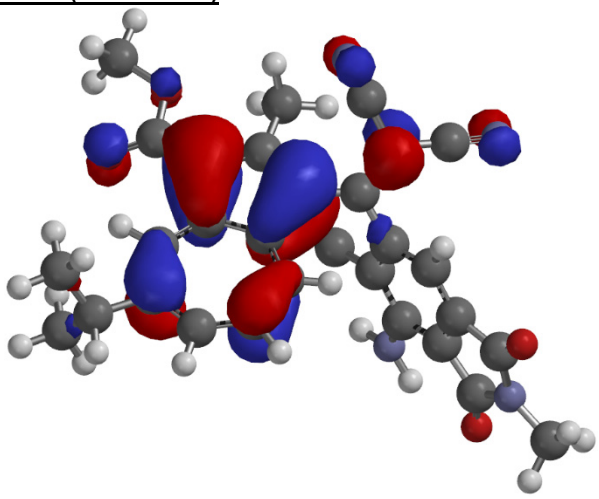


Figure S93. Frontier Kohn–Sham orbitals of TCBD **4b** at the B3LYP/6-311G* level.

HOMO (−6.19 eV)



LUMO (−3.43 eV)

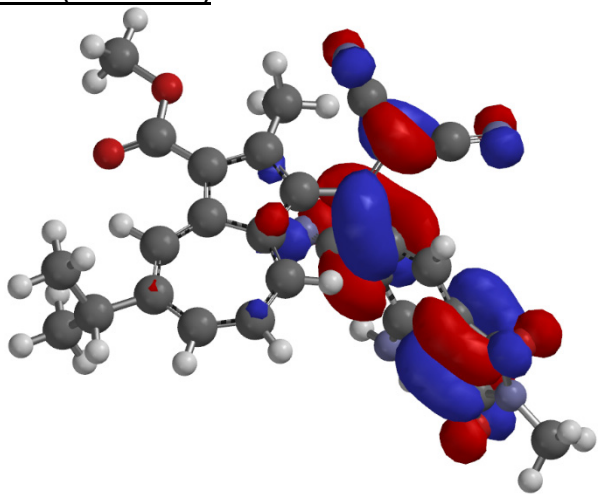
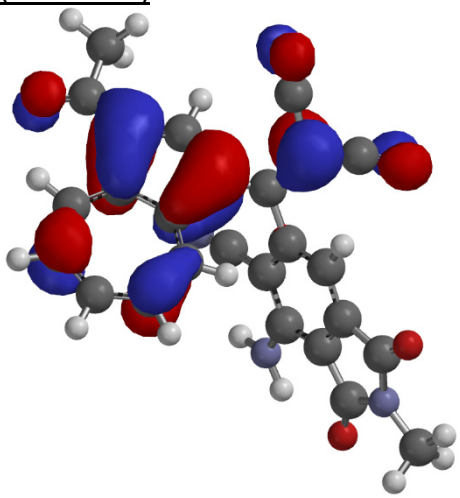


Figure S94. Frontier Kohn–Sham orbitals of TCBD **4c** at the B3LYP/6-311G* level.

HOMO (−6.43 eV)



LUMO (−3.49 eV)

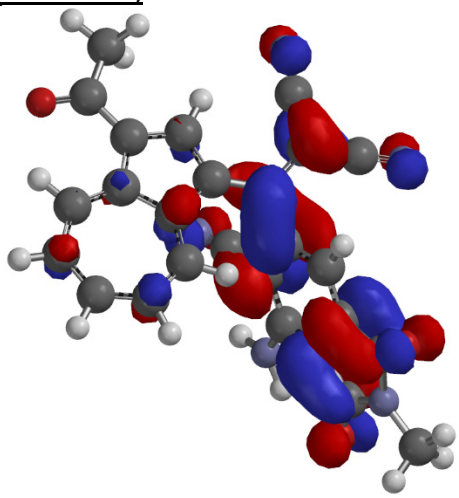
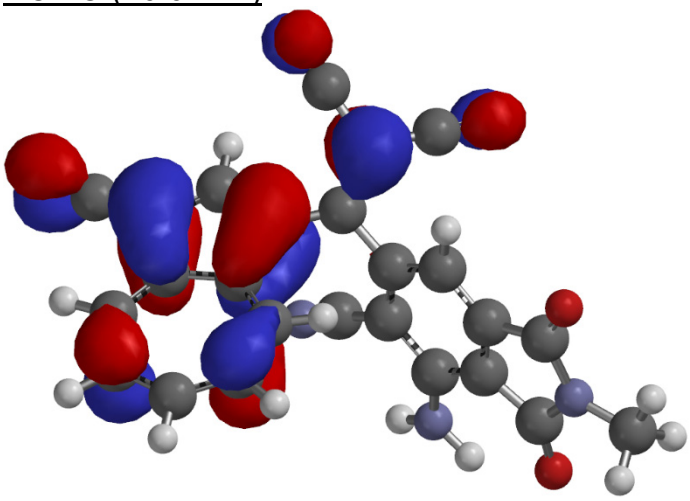


Figure S95. Frontier Kohn–Sham orbitals of TCBD **4d** at the B3LYP/6-311G* level.

HOMO (−6.61 eV)



LUMO (−3.63 eV)

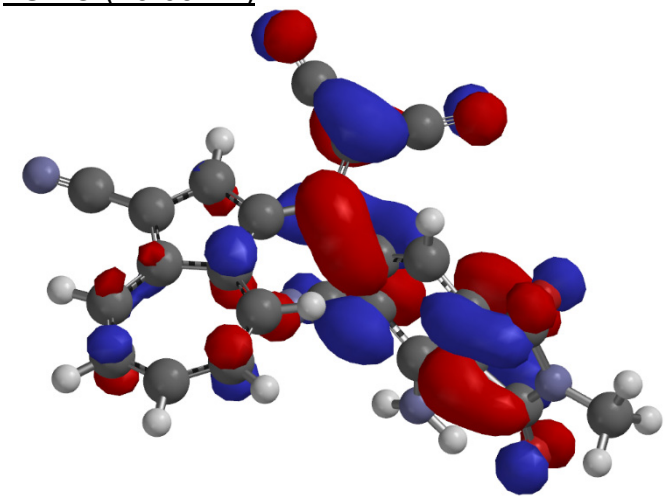
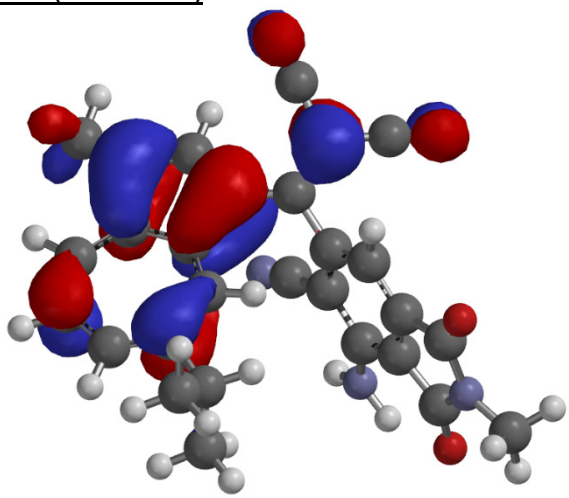


Figure S96. Frontier Kohn–Sham orbitals of TCBD **4e** at the B3LYP/6-311G* level.

HOMO (−5.93 eV)



LUMO (−3.29 eV)

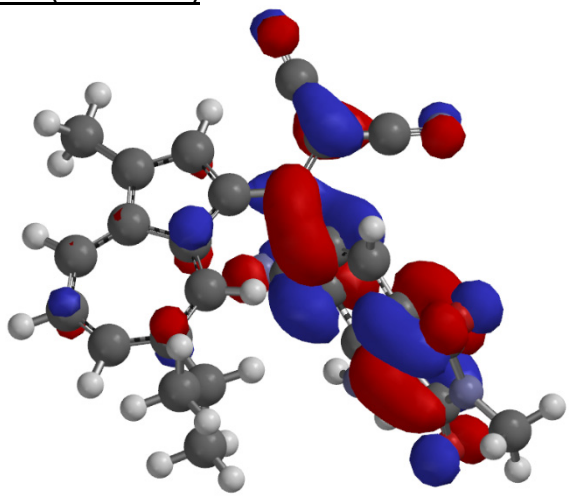
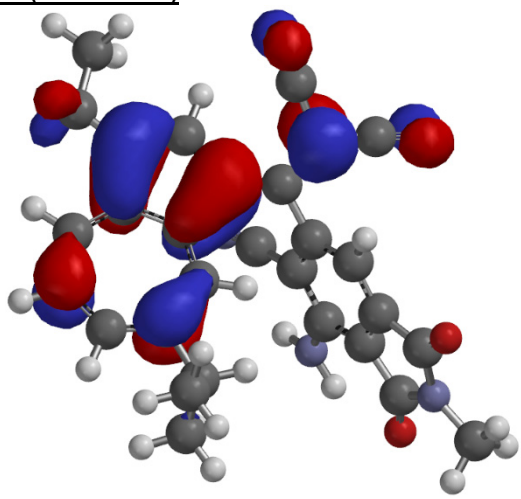


Figure S97. Frontier Kohn–Sham orbitals of TCBD **4f** at the B3LYP/6-311G* level.

HOMO (−5.91 eV)



LUMO (−3.28 eV)

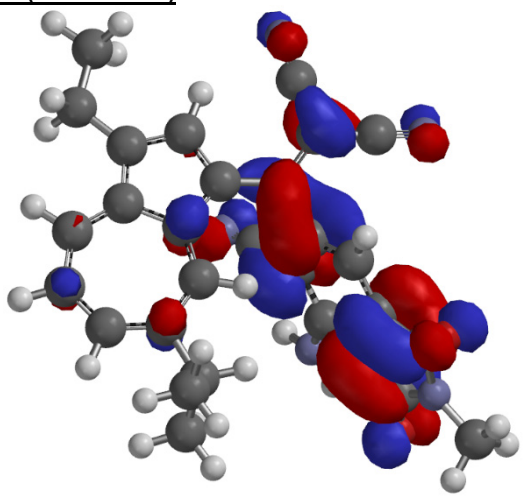


Figure S98. Frontier Kohn–Sham orbitals of TCBD **4g** at the B3LYP/6-311G* level.

Table S1. Cartesian coordinates of **2a**; absolute energy is − 1692.74938 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	5.453963	0.622032	0.529487	31	H	H12C	6.56834	3.022627	0.399094
2	O	O2	4.553036	2.667219	0.80248	32	C	C13	-0.45414	1.710203	-0.42974
3	O	O3	-5.53535	-1.94822	1.860487	33	C	C14	-0.68465	3.014294	-0.81664
4	O	O4	-5.29138	-0.26952	-2.40811	34	C	C15	0.32275	3.856244	-1.37734
5	N	N1	1.104346	4.565462	-1.84902	35	C	C16	-1.96391	3.635422	-0.67666
6	N	N2	-2.98215	4.17162	-0.56583	36	C	C17	-1.63976	0.895009	-0.02166
7	N	N3	0.062843	0.71635	3.121675	37	C	C18	-1.70777	0.356503	1.282307
8	N	N4	-2.92772	-0.93971	2.957065	38	C	C19	-2.82972	-0.41999	1.715467
9	H	H4NA	-3.76476	-1.4438	3.20844	39	C	C20	-3.83141	-0.62034	0.756358
10	H	H4NB	-2.23677	-0.74453	3.662803	40	C	C21	-3.74855	-0.09133	-0.52147
11	N	N5	-5.7453	-1.24528	-0.34692	41	C	C22	-2.67691	0.673955	-0.94152
12	C	C1	3.128431	0.971229	0.079649	42	H	H22	-2.63339	1.083242	-1.94272
13	C	C2	2.036566	1.83021	0.020828	43	C	C23	-0.68935	0.588438	2.252237
14	H	H2	2.081178	2.876968	0.279112	44	C	C24	-5.10496	-1.35659	0.890068
15	C	C3	0.868862	1.12732	-0.35605	45	C	C25	-4.98626	-0.50189	-1.26551
16	C	C4	1.247231	-0.23877	-0.57144	46	C	C26	-7.04476	-1.82358	-0.64336
17	C	C5	0.440393	-1.26849	-1.04879	47	H	H26A	-7.23977	-1.67173	-1.70332
18	H	H5	-0.58792	-1.00846	-1.27379	48	H	H26B	-7.82563	-1.3373	-0.05568
19	C	C6	0.771043	-2.59971	-1.28676	49	H	H8	-7.04468	-2.88991	-0.41426
20	H	H6	-0.04286	-3.21481	-1.66084	50	C	C27	4.377934	-3.79032	-0.587
21	C	C7	1.983706	-3.26754	-1.11549	51	H	H3	3.992282	-4.76071	-0.91826
22	H	H7	1.954853	-4.32569	-1.36473	52	C	C28	5.545213	-3.41615	-1.51831
23	C	C8	3.224063	-2.79053	-0.6832	53	H	H1	6.317345	-4.19011	-1.48884
24	C	C9	3.507912	-1.46166	-0.33642	54	H	H10	6.01344	-2.47469	-1.22033
25	H	H9	4.533138	-1.25245	-0.05316	55	H	H11	5.213611	-3.31284	-2.55466
26	C	C10	2.689654	-0.33828	-0.28657	56	C	C29	4.857775	-3.95681	0.866316
27	C	C11	4.48836	1.358513	0.481721	57	H	H4	5.631007	-4.72788	0.926125
28	C	C12	5.84349	3.153168	1.203429	58	H	H12	4.038881	-4.25139	1.527858
29	H	H12A	6.193379	2.622267	2.089456	59	H	H13	5.285837	-3.0299	1.257341
30	H	H12B	5.698832	4.20865	1.420102						

Table S2. Cartesian coordinates of **2b**; absolute energy is −1732.07420 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	5.711274	0.325755	0.449505	32	C	C13	-0.12604	1.822162	-0.38764
2	O	O2	4.978708	2.444906	0.653253	33	C	C14	-0.27057	3.130722	-0.80176
3	O	O3	-5.43203	-1.39099	2.053265	34	C	C15	0.781213	3.886538	-1.40189
4	O	O4	-5.1015	0.115383	-2.27432	35	C	C16	-1.50097	3.841895	-0.65337
5	N	N1	1.601144	4.525641	-1.90755	36	C	C17	-1.3629	1.106422	0.055497
6	N	N2	-2.47841	4.447795	-0.5345	37	C	C18	-1.45948	0.619826	1.377595
7	N	N3	0.348769	0.91912	3.191115	38	C	C19	-2.62876	-0.06328	1.84176
8	N	N4	-2.75394	-0.53102	3.10156	39	C	C20	-3.6473	-0.22903	0.893522
9	H	H4NA	-3.62559	-0.95805	3.376933	40	C	C21	-3.53669	0.250029	-0.40179
10	H	H4NB	-2.04845	-0.3544	3.797804	41	C	C22	-2.41899	0.927354	-0.8517
11	N	N5	-5.60789	-0.75797	-0.18099	42	H	H22	-2.35438	1.299989	-1.86606
12	C	C1	3.406598	0.834679	0.048277	43	C	C23	-0.41946	0.814184	2.332718
13	C	C2	2.379823	1.770479	-0.00913	44	C	C24	-4.96829	-0.87045	1.057195
14	H	H2	2.512338	2.819269	0.207455	45	C	C25	-4.80414	-0.10111	-1.12604
15	C	C3	1.151268	1.144663	-0.3244	46	C	C26	-6.94632	-1.2726	-0.46274
16	C	C4	1.421167	-0.25384	-0.49704	47	H	H26A	-7.37072	-0.63344	-1.23809
17	C	C5	0.52334	-1.23827	-0.89969	48	H	H26B	-7.53495	-1.14708	0.447639
18	H	H5	-0.49142	-0.91154	-1.0947	49	C	C27	4.281557	-4.0276	-0.49715
19	C	C6	0.744958	-2.59892	-1.09264	50	H	H3	3.793507	-4.98547	-0.70832
20	H	H6	-0.12678	-3.16617	-1.40726	51	C	C28	5.368183	-3.82221	-1.56774
21	C	C7	1.911846	-3.34753	-0.93994	52	H	H1	6.085856	-4.64727	-1.54558
22	H	H7	1.794975	-4.40953	-1.14329	53	H	H10	5.927437	-2.89667	-1.40471
23	C	C8	3.200828	-2.94815	-0.57575	54	H	H11	4.937566	-3.7799	-2.57159
24	C	C9	3.593931	-1.63232	-0.29195	55	C	C29	4.894166	-4.1273	0.910926
25	H	H9	4.641953	-1.48803	-0.05464	56	H	H4	5.602653	-4.9589	0.957783
26	C	C10	2.860926	-0.45049	-0.25507	57	H	H12	4.126195	-4.2979	1.669737
27	C	C11	4.803955	1.13257	0.394758	58	H	H13	5.43698	-3.21872	1.18371
28	C	C12	6.314318	2.845481	0.996211	59	C	C30	-6.93911	-2.73471	-0.90584
29	H	H12A	6.648612	2.330777	1.897737	60	H	H8	-6.36384	-2.86358	-1.82551
30	H	H12B	6.258701	3.918047	1.164798	61	H	H14	-7.96105	-3.07192	-1.09789
31	H	H12C	7.002479	2.620412	0.180529	62	H	H15	-6.5128	-3.3767	-0.13183

Table S3. Cartesian coordinates of **2c**; absolute energy is −1888.15669 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	6.640951	-0.0168	0.486864	37	C	C18	-0.48669	0.864827	1.255824
2	O	O2	6.060601	2.157722	0.545872	38	C	C19	-1.71113	0.315012	1.752643
3	O	O3	-4.61149	-0.7198	2.045705	39	C	C20	-2.74266	0.190594	0.81171
4	O	O4	-4.1652	0.511082	-2.36644	40	C	C21	-2.58984	0.582363	-0.50676
5	N	N1	2.907238	4.265369	-2.26911	41	C	C22	-1.41505	1.125551	-0.99229
6	N	N2	-1.17265	4.643514	-0.96955	42	H	H22	-1.31956	1.431851	-2.0261
7	N	N3	1.333672	1.117517	3.064074	43	C	C23	0.563363	1.026766	2.20593
8	N	N4	-1.87691	-0.06833	3.036134	44	C	C24	-4.12112	-0.30561	1.011147
9	H	H4NA	-2.78362	-0.4008	3.328669	45	C	C25	-3.88989	0.323101	-1.20769
10	H	H4NB	-1.15902	0.084882	3.725169	46	C	C27	4.908843	-4.30763	-0.20918
11	N	N5	-4.75745	-0.20008	-0.2299	47	H	H3	4.366301	-5.23655	-0.41753
12	C	C1	4.384944	0.629193	0.012396	48	C	C28	6.072495	-4.22073	-1.21207
13	C	C2	3.431263	1.631446	-0.12716	49	H	H1	6.722779	-5.09348	-1.10845
14	H	H2	3.635458	2.679654	0.028143	50	H	H10	6.686993	-3.33198	-1.04491
15	C	C3	2.166483	1.07781	-0.43235	51	H	H11	5.711527	-4.18963	-2.24326
16	C	C4	2.334862	-0.34423	-0.51266	52	C	C29	5.426539	-4.39461	1.237772
17	C	C5	1.375067	-1.28385	-0.87887	53	H	H4	6.091904	-5.25523	1.349595
18	H	H5	0.391609	-0.89773	-1.12139	54	H	H12	4.606266	-4.50847	1.951138
19	C	C6	1.50134	-2.66623	-0.98604	55	H	H13	5.993023	-3.50323	1.519416
20	H	H6	0.597218	-3.18727	-1.28897	56	C	C26	-6.16148	-0.53852	-0.51802
21	C	C7	2.606885	-3.48595	-0.76004	57	C	C30	-7.90554	-2.35987	-0.67013
22	H	H7	2.418365	-4.547	-0.90597	58	C	C31	-8.58666	0.004507	-0.06305
23	C	C8	3.912954	-3.1595	-0.38526	59	C	C32	-8.88172	-1.48791	0.130301
24	C	C9	4.395608	-1.86086	-0.16787	60	C	C33	-7.13085	0.343551	0.285873
25	H	H9	5.446159	-1.77816	0.087989	61	C	C34	-6.4454	-2.03584	-0.32234
26	C	C10	3.751451	-0.62935	-0.22351	62	H	H8	-8.0706	-2.20146	-1.74394
27	C	C11	5.794862	0.847267	0.366079	63	H	H14	-8.78586	0.284252	-1.10551
28	C	C12	7.416763	2.482925	0.887291	64	H	H15	-8.79926	-1.73861	1.195578
29	H	H12A	7.696796	2.012731	1.830648	65	H	H16	-6.95872	0.189428	1.355235
30	H	H12B	7.439158	3.566167	0.976941	66	H	H17	-6.24416	-2.3107	0.717524
31	H	H12C	8.099202	2.148277	0.105182	67	H	H18	-6.27354	-0.29847	-1.57953
32	C	C13	0.945734	1.843994	-0.57008	68	H	H19	-8.10055	-3.42062	-0.48282
33	C	C14	0.914007	3.122594	-1.08835	69	H	H20	-9.26171	0.609377	0.551117
34	C	C15	2.031887	3.737899	-1.72872	70	H	H21	-9.91317	-1.71063	-0.16123
35	C	C16	-0.25229	3.945409	-1.01841	71	H	H23	-6.922	1.397387	0.073103
36	C	C17	-0.34719	1.264339	-0.09242	72	H	H24	-5.76401	-2.62088	-0.94853

Table S4. Cartesian coordinates of **2d**; absolute energy is −1884.52144 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	6.476809	0.012091	0.346186	34	C	C15	1.782003	3.793501	-1.65058
2	O	O2	5.897445	2.185382	0.447539	35	C	C16	-0.48248	3.970815	-0.8678
3	O	O3	-4.67657	-0.79858	2.286043	36	C	C17	-0.52814	1.285207	0.030703
4	O	O4	-4.36111	0.436658	-2.14934	37	C	C18	-0.62341	0.892308	1.383873
5	N	N1	2.63775	4.334094	-2.20898	38	C	C19	-1.82438	0.326542	1.91897
6	N	N2	-1.41061	4.654579	-0.78006	39	C	C20	-2.87759	0.173267	1.005548
7	N	N3	1.247719	1.177881	3.134673	40	C	C21	-2.7685	0.562633	-0.31767
8	N	N4	-1.94719	-0.04737	3.209167	41	C	C22	-1.61882	1.126672	-0.83783
9	H	H4NA	-2.83579	-0.4006	3.53113	42	H	H22	-1.55767	1.42832	-1.87561
10	H	H4NB	-1.2074	0.11255	3.873145	43	C	C23	0.452533	1.073166	2.301168
11	N	N5	-4.90344	-0.29473	0.006879	44	C	C24	-4.227	-0.36952	1.244011
12	C	C1	4.205086	0.660693	-0.04225	45	C	C25	-4.06823	0.259688	-0.99599
13	C	C2	3.245791	1.663	-0.13542	46	C	C27	4.728556	-4.27235	-0.34586
14	H	H2	3.454407	2.70974	0.023744	47	H	H3	4.180935	-5.19933	-0.54936
15	C	C3	1.971232	1.110699	-0.40025	48	C	C28	5.857182	-4.16771	-1.38677
16	C	C4	2.138701	-0.31049	-0.50349	49	H	H1	6.509435	-5.0429	-1.32348
17	C	C5	1.168362	-1.24788	-0.84692	50	H	H10	6.478318	-3.28287	-1.22357
18	H	H5	0.175587	-0.86211	-1.04771	51	H	H11	5.460581	-4.11632	-2.40399
19	C	C6	1.29338	-2.62819	-0.97884	52	C	C29	5.294978	-4.37869	1.081379
20	H	H6	0.379938	-3.14694	-1.25669	53	H	H4	5.963128	-5.24079	1.160046
21	C	C7	2.407889	-3.44819	-0.80486	54	H	H12	4.499081	-4.50174	1.82047
22	H	H7	2.216605	-4.50742	-0.95998	55	H	H13	5.870898	-3.49085	1.355211
23	C	C8	3.725769	-3.12391	-0.4713	56	C	C26	-6.24916	-0.72456	-0.21142
24	C	C9	4.213143	-1.82719	-0.25343	57	C	C30	-8.86844	-1.5659	-0.65093
25	H	H9	5.271968	-1.74565	-0.03372	58	C	C31	-7.12122	0.076668	-0.95065
26	C	C10	3.565355	-0.5962	-0.26994	59	C	C32	-6.68429	-1.94432	0.310618
27	C	C11	5.626642	0.876694	0.263797	60	C	C33	-7.99548	-2.35636	0.092997
28	C	C12	7.264409	2.508683	0.745508	61	C	C34	-8.4263	-0.35218	-1.17217
29	H	H12A	7.577971	2.027277	1.672566	62	H	H8	-6.77867	1.01866	-1.35828
30	H	H12B	7.288524	3.590776	0.847856	63	H	H14	-6.0082	-2.5572	0.892688
31	H	H12C	7.919504	2.184984	-0.06415	64	H	H15	-8.33173	-3.30224	0.504518
32	C	C13	0.745325	1.875828	-0.4858	65	H	H16	-9.09965	0.270537	-1.75154
33	C	C14	0.68892	3.160862	-0.98505	66	H	H17	-9.88828	-1.89358	-0.82294

Table S5. Cartesian coordinates of **2e**; absolute energy is −2041.81134 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	7.641176	-0.40941	0.357444	40	C	C21	-1.52196	0.870048	-0.27591
2	O	O2	7.226832	1.798925	0.504449	41	C	C22	-0.3395	1.337457	-0.81942
3	O	O3	-3.48915	-0.2915	2.379319	42	H	H22	-0.26765	1.612813	-1.86381
4	O	O4	-3.14013	0.805147	-2.08948	43	C	C23	1.760703	1.186442	2.297804
5	N	N1	4.169404	4.179225	-2.25547	44	C	C24	-3.02372	0.078066	1.321146
6	N	N2	0.159642	4.85137	-0.84882	45	C	C25	-2.84753	0.637685	-0.93445
7	N	N3	2.571594	1.245072	3.120623	46	C	C27	5.574726	-4.52998	-0.4512
8	N	N4	-0.7084	0.279228	3.255854	47	H	H3	4.96607	-5.40757	-0.69584
9	H	H4NA	-1.61606	-0.00627	3.591411	48	C	C28	6.724615	-4.46838	-1.47238
10	H	H4NB	0.050429	0.389822	3.908202	49	H	H1	7.313217	-5.38881	-1.43141
11	N	N5	-3.70633	0.164316	0.088913	50	H	H10	7.402936	-3.63545	-1.26957
12	C	C1	5.430173	0.419389	-0.03827	51	H	H11	6.348813	-4.35378	-2.49235
13	C	C2	4.554045	1.495889	-0.13059	52	C	C29	6.110948	-4.72829	0.978198
14	H	H2	4.841799	2.521779	0.040496	53	H	H4	6.720004	-5.63497	1.031406
15	C	C3	3.242877	1.04842	-0.41076	54	H	H12	5.297118	-4.82698	1.70136
16	C	C4	3.296964	-0.37965	-0.52513	55	H	H13	6.739142	-3.89111	1.294163
17	C	C5	2.254973	-1.23262	-0.87907	56	C	C26	-5.07848	-0.18854	-0.10067
18	H	H5	1.295715	-0.76583	-1.07192	57	C	C30	-7.77904	-0.88458	-0.46228
19	C	C6	2.270912	-2.61626	-1.03208	58	C	C31	-5.9121	0.622241	-0.87165
20	H	H6	1.319061	-3.05752	-1.31491	59	C	C32	-5.58692	-1.34905	0.484359
21	C	C7	3.319346	-3.52328	-0.87886	60	C	C33	-6.92484	-1.68105	0.306615
22	H	H7	3.046929	-4.56112	-1.05569	61	C	C34	-7.2444	0.265808	-1.05135
23	C	C8	4.658997	-3.30781	-0.54407	62	H	H8	-5.5195	1.514793	-1.34139
24	C	C9	5.244263	-2.05791	-0.29493	63	H	H14	-4.94544	-1.9815	1.084355
25	H	H9	6.305001	-2.06583	-0.07134	64	H	H15	-7.30897	-2.58433	0.771799
26	C	C10	4.695642	-0.78005	-0.28798	65	H	H16	-7.88006	0.900652	-1.66184
27	C	C11	6.860177	0.519009	0.286462	66	C	C35	-9.24186	-1.23459	-0.61012
28	C	C12	8.609685	2.011209	0.82745	67	H	H17	-9.36299	-2.3237	-0.57875
29	H	H12A	8.871759	1.488065	1.747845	68	H	H18	-9.59436	-0.91413	-1.59537
30	H	H12B	8.713753	3.085965	0.954281	69	C	C36	-10.1155	-0.60237	0.491712
31	H	H12C	9.250761	1.656077	0.0199	70	H	H19	-9.69572	-0.87424	1.466673
32	C	C13	2.081618	1.909514	-0.50187	71	H	H21	-10.041	0.49001	0.42604
33	C	C14	2.129517	3.186501	-1.01949	72	C	C37	-11.5924	-1.02102	0.4476
34	C	C15	3.271931	3.717087	-1.69218	73	H	H20	-11.6615	-2.11364	0.524834
35	C	C16	1.027947	4.091298	-0.91851	74	H	H24	-12.0884	-0.62637	1.341819
36	C	C17	0.770167	1.430395	0.034452	75	C	C38	-12.3524	-0.54558	-0.79425
37	C	C18	0.662624	1.072444	1.39634	76	H	H23	-11.9637	-0.99324	-1.71289
38	C	C19	-0.57155	0.613449	1.956479	77	H	H25	-12.2881	0.541934	-0.90465

39	C	C20	-1.6432	0.519641	1.056958	78	H	H26	-13.4124	-0.80652	-0.73202

Table S6. Cartesian coordinates of **4a**; absolute energy is −1574.78186 au.

		Atom	X	Y	Z		Atom	X	Y	Z
1	O	O1	5.882361	-1.13115	0.280474	26	C	C10	2.962926	-1.37105	-0.50531
2	O	O2	5.472705	1.032957	0.75047	27	C	C11	5.108801	-0.19739	0.334323
3	O	O3	-5.39735	-1.32711	1.784235	28	C	C12	6.849118	1.186149	1.130449
4	O	O4	-4.82227	0.65211	-2.32163	29	H	H12A	7.086651	0.539228	1.975944
5	N	N1	2.48704	3.872106	-1.55221	30	H	H12B	6.956723	2.232232	1.406202
6	N	N2	-1.56082	4.288073	-0.15163	31	H	H12C	7.505958	0.939153	0.295768
7	N	N3	0.665079	-0.06935	3.115354	32	C	C13	0.347388	1.323358	-0.32123
8	N	N4	-2.6238	-1.01565	2.893613	33	C	C14	0.412418	2.675295	-0.58437
9	H	H4NA	-3.55075	-1.34026	3.124674	34	C	C15	1.574453	3.318047	-1.10926
10	H	H4NB	-1.90143	-1.04475	3.594277	35	C	C16	-0.69126	3.551232	-0.34485
11	N	N5	-5.46337	-0.39825	-0.34799	36	C	C17	-0.98618	0.759921	0.053673
12	C	C1	3.685141	-0.23749	-0.03608	37	C	C18	-1.16138	0.131777	1.30661
13	C	C2	2.808225	0.842295	0.03731	38	C	C19	-2.42304	-0.41776	1.700746
14	H	H2	3.091603	1.82173	0.390954	39	C	C20	-3.45175	-0.30678	0.755404
15	C	C3	1.507765	0.45483	-0.35202	40	C	C21	-3.26508	0.310123	-0.47062
16	C	C4	1.568385	-0.9329	-0.71323	41	C	C22	-2.05524	0.859637	-0.85098
17	C	C5	0.53137	-1.69831	-1.24141	42	H	H22	-1.93325	1.341386	-1.81284
18	H	H5	-0.41372	-1.18527	-1.37828	43	C	C23	-0.10769	0.04657	2.26251
19	C	C6	0.523244	-3.03804	-1.62759	44	C	C24	-4.8546	-0.75885	0.857366
20	H	H6	-0.42641	-3.40415	-2.0071	45	C	C25	-4.56728	0.24826	-1.21507
21	C	C7	1.558724	-3.96957	-1.59482	46	C	C26	-6.85604	-0.66341	-0.66716
22	H	H7	1.302377	-4.96705	-1.9423	47	H	H26A	-7.08373	-0.1528	-1.60108
23	C	C8	2.878799	-3.79337	-1.18634	48	H	H26B	-7.50081	-0.28833	0.128226
24	C	C9	3.502356	-2.6396	-0.71511	49	H	H1	3.514931	-4.67107	-1.2553
25	H	H9	4.556025	-2.72432	-0.47339	50	H	H8	-7.02896	-1.73497	-0.78651

Table S7. Cartesian coordinates of **4b**; absolute energy is −1692.74953 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	5.74323	0.303627	0.651945	31	C	C13	-0.02622	1.816015	-0.55181
2	O	O2	5.051894	2.447197	0.635968	32	C	C14	-0.12728	3.068449	-1.12071
3	O	O3	-5.4618	-1.04156	2.047434	33	C	C15	0.963837	3.7299	-1.76127
4	O	O4	-4.9657	-0.02232	-2.40433	34	C	C16	-1.347	3.812966	-1.1084
5	N	N1	1.816697	4.291012	-2.30348	35	C	C17	-1.29105	1.175749	-0.07347
6	N	N2	-2.31474	4.445485	-1.10704	36	C	C18	-1.44044	0.845771	1.291374
7	N	N3	0.302738	1.344472	3.12453	37	C	C19	-2.63443	0.232736	1.788866
8	N	N4	-2.80925	-0.08559	3.088685	38	C	C20	-3.62253	-0.03	0.83055
9	H	H4NA	-3.69439	-0.47232	3.380094	39	C	C21	-3.46224	0.29928	-0.50572
10	H	H4NB	-2.1252	0.161884	3.784808	40	C	C22	-2.32003	0.907849	-0.99046
11	N	N5	-5.55155	-0.65754	-0.24511	41	H	H22	-2.21724	1.161625	-2.03788
12	C	C1	3.466871	0.816258	0.12508	42	C	C23	-0.43289	1.142202	2.255097
13	C	C2	2.458775	1.761928	-0.06031	43	C	C24	-4.95631	-0.63652	1.01918
14	H	H2	2.602831	2.824221	0.064076	44	C	C25	-4.70981	-0.11493	-1.23015
15	C	C3	1.235185	1.128883	-0.36522	45	C	C26	-6.88364	-1.17501	-0.50678
16	C	C4	1.48825	-0.28613	-0.39654	46	H	H26A	-7.08847	-1.03557	-1.56658
17	C	C5	0.589148	-1.28859	-0.74538	47	H	H26B	-7.62397	-0.63561	0.085723
18	H	H5	-0.40893	-0.95588	-1.00696	48	H	H1	3.913533	-3.74575	-0.04066
19	C	C6	0.764304	-2.66991	-0.82177	49	H	H8	-6.93868	-2.23616	-0.25789
20	H	H6	-0.12693	-3.21465	-1.12195	50	C	C27	1.72576	-4.9746	-0.73591
21	C	C7	1.890197	-3.46316	-0.57337	51	H	H4	0.695303	-5.15019	-1.06123
22	C	C8	3.152407	-2.9856	-0.19124	52	C	C28	1.912448	-5.71584	0.600547
23	C	C9	3.6021	-1.68558	0.011106	53	H	H7	2.934932	-5.62596	0.976595
24	H	H9	4.64761	-1.57929	0.279331	54	H	H10	1.702334	-6.78122	0.472723
25	C	C10	2.909571	-0.47676	-0.08087	55	H	H11	1.239008	-5.32959	1.369709
26	C	C11	4.858202	1.119447	0.492585	56	C	C29	2.650239	-5.54299	-1.82685
27	C	C12	6.383464	2.85216	0.98867	57	H	H3	3.705143	-5.45245	-1.55496
28	H	H12A	6.670394	2.426597	1.951079	58	H	H12	2.506834	-5.02954	-2.78078
29	H	H12B	6.349306	3.937411	1.044224	59	H	H13	2.442182	-6.60513	-1.98199
30	H	H12C	7.095904	2.528812	0.228792						

Table S8. Cartesian coordinates of **4c**; absolute energy is −1732.07212 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	5.252624	0.430733	0.909356	32	C	C14	-0.71668	2.639027	-1.5427
2	O	O2	4.52283	2.550592	0.903409	33	C	C15	0.343189	3.230134	-2.29646
3	O	O3	-5.5674	-1.645	2.234874	34	C	C16	-1.98459	3.286226	-1.67986
4	O	O4	-5.31379	-0.89434	-2.29019	35	C	C17	-1.71399	0.833039	-0.18366
5	N	N1	1.175688	3.716653	-2.93326	36	C	C18	-1.78765	0.580828	1.205562
6	N	N2	-2.98942	3.843347	-1.80479	37	C	C19	-2.90027	-0.10628	1.78783
7	N	N3	-0.06398	1.364669	2.954602	38	C	C20	-3.88851	-0.52657	0.887616
8	N	N4	-3.00426	-0.34745	3.111197	39	C	C21	-3.80392	-0.27537	-0.472
9	H	H4NA	-3.82709	-0.81663	3.458303	40	C	C22	-2.74446	0.40687	-1.03862
10	H	H4NB	-2.30865	-0.02075	3.761442	41	H	H22	-2.70119	0.588787	-2.10492
11	N	N5	-5.76591	-1.43335	-0.07328	42	C	C23	-0.78869	1.033579	2.115726
12	C	C1	3.009439	0.886389	0.209311	43	C	C24	-5.14073	-1.25884	1.164609
13	C	C2	1.913056	1.761207	0.033146	44	C	C25	-5.01777	-0.87302	-1.12202
14	C	C3	0.8073	1.010802	-0.4572	45	C	C26	-7.03856	-2.11092	-0.2523
15	C	C4	1.206556	-0.35218	-0.58738	46	H	H26A	-7.25389	-2.13057	-1.3189
16	C	C5	0.434211	-1.38998	-1.10271	47	H	H26B	-7.83326	-1.57831	0.272806
17	H	H5	-0.57156	-1.1288	-1.41361	48	H	H8	-6.98413	-3.13042	0.13244
18	C	C6	0.770973	-2.72829	-1.28115	49	C	C27	4.318698	-3.88938	-0.31146
19	H	H6	-0.0163	-3.35039	-1.69829	50	H	H3	3.95954	-4.86309	-0.66249
20	C	C7	1.967673	-3.38822	-1.00955	51	C	C28	5.545644	-3.51822	-1.1637
21	H	H7	1.962464	-4.45039	-1.24218	52	H	H1	6.31076	-4.29581	-1.08644
22	C	C8	3.170089	-2.89443	-0.49546	53	H	H10	5.997523	-2.57986	-0.83265
23	C	C9	3.427107	-1.56277	-0.13998	54	H	H11	5.282043	-3.40953	-2.21885
24	H	H9	4.425266	-1.35783	0.226047	55	C	C29	4.700313	-4.04635	1.171192
25	C	C10	2.612898	-0.43229	-0.16908	56	H	H4	5.481029	-4.80381	1.284802
26	C	C11	4.353861	1.224594	0.702481	57	H	H12	3.842749	-4.35512	1.774857
27	C	C12	5.819161	2.947957	1.376669	58	H	H13	5.083201	-3.11136	1.588464
28	H	H12A	6.035706	2.481958	2.338381	59	C	C30	1.862579	3.206927	0.413193
29	H	H12B	5.769508	4.029755	1.476603	60	H	H2	2.269219	3.350429	1.413907
30	H	H12C	6.591468	2.663544	0.66117	61	H	H14	0.844408	3.592542	0.397156
31	C	C13	-0.52225	1.528518	-0.75447	62	H	H15	2.466719	3.8115	-0.26731

Table S9. Cartesian coordinates of **4d**; absolute energy is −1499.52896 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O1	6.116366	0.922642	-0.41708	26	C	C11	5.349074	-0.02265	-0.50964
2	O	O3	-5.17428	1.49252	-1.64946	27	C	C13	0.539194	-1.37931	0.236938
3	O	O4	-4.54599	-0.57712	2.403622	28	C	C14	0.568716	-2.73991	0.462977
4	N	N1	2.651027	-4.02002	1.299817	29	C	C15	1.730797	-3.43188	0.920701
5	N	N2	-1.47195	-4.27543	0.06496	30	C	C16	-0.57093	-3.57349	0.243213
6	N	N3	0.80729	0.051101	-3.17554	31	C	C17	-0.78639	-0.76526	-0.08382
7	N	N4	-2.44556	1.112828	-2.84455	32	C	C18	-0.9766	-0.10952	-1.32047
8	H	H4NA	-3.36764	1.471683	-3.04228	33	C	C19	-2.22971	0.488463	-1.66818
9	H	H4NB	-1.74415	1.13033	-3.56666	34	C	C20	-3.23333	0.395054	-0.69414
10	N	N5	-5.20979	0.527719	0.467439	35	C	C21	-3.03198	-0.24971	0.514901
11	C	C1	3.929991	0.070122	-0.10227	36	C	C22	-1.83086	-0.84626	0.850222
12	C	C2	3.003922	-0.96966	-0.18002	37	H	H22	-1.69811	-1.34991	1.799415
13	H	H2	3.219751	-1.95454	-0.56668	38	C	C23	0.051858	-0.04681	-2.30509
14	C	C3	1.726986	-0.54927	0.252698	39	C	C24	-4.62389	0.891586	-0.74815
15	C	C4	1.847717	0.825237	0.645902	40	C	C25	-4.31038	-0.16124	1.297338
16	C	C5	0.847519	1.613805	1.213707	41	C	C26	-6.58608	0.824657	0.827803
17	H	H5	-0.11215	1.130643	1.356923	42	H	H26A	-6.79531	0.324658	1.771582
18	C	C6	0.893688	2.941973	1.631032	43	H	H26B	-7.26378	0.45857	0.055846
19	H	H6	-0.03468	3.332086	2.038408	44	H	H1	3.934769	4.475101	1.237486
20	C	C7	1.961929	3.837535	1.599542	45	H	H8	-6.73188	1.900387	0.944016
21	H	H7	1.748368	4.834698	1.975588	46	C	C12	5.834058	-1.35337	-1.05753
22	C	C8	3.265123	3.62334	1.160866	47	H	H3	6.897155	-1.27286	-1.27785
23	C	C9	3.837996	2.458944	0.648856	48	H	H4	5.29886	-1.61675	-1.97482
24	H	H9	4.888684	2.510584	0.387823	49	H	H10	5.674066	-2.16419	-0.34123
25	C	C10	3.250921	1.216266	0.417532						

Table S10. Cartesian coordinates of **4e**; absolute energy is −1439.10713 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O3	-4.90968	-1.61936	1.538076	24	C	C13	0.775118	1.429923	-0.14667
2	O	O4	-4.254	0.608767	-2.42574	25	C	C14	0.778505	2.791878	-0.35043
3	N	N1	2.84354	4.137114	-1.12599	26	C	C15	1.934113	3.518039	-0.77177
4	N	N2	-1.30552	4.275972	0.019988	27	C	C16	-0.38558	3.596463	-0.14678
5	N	N3	1.017329	-0.14385	3.215066	28	C	C17	-0.53949	0.773228	0.129285
6	N	N4	-2.20656	-1.23906	2.789363	29	C	C18	-0.73612	0.065722	1.336501
7	H	H4NA	-3.12572	-1.61946	2.958425	30	C	C19	-1.98337	-0.56755	1.641001
8	H	H4NB	-1.51873	-1.26724	3.524159	31	C	C20	-2.97312	-0.45627	0.654326
9	N	N5	-4.93002	-0.5763	-0.54165	32	C	C21	-2.76443	0.235787	-0.52641
10	C	C1	4.174439	0.037209	0.257451	33	C	C22	-1.56972	0.866867	-0.8194
11	C	C2	3.236957	1.065746	0.34663	34	H	H22	-1.43274	1.407701	-1.74733
12	H	H2	3.437833	2.034936	0.778591	35	C	C23	0.27629	-0.01877	2.335703
13	C	C3	1.988051	0.630014	-0.14975	36	C	C24	-4.35648	-0.97663	0.668092
14	C	C4	2.148891	-0.72455	-0.58997	37	C	C25	-4.02938	0.156245	-1.33176
15	C	C5	1.195594	-1.52061	-1.22006	38	C	C26	-6.29754	-0.87739	-0.93163
16	H	H5	0.226037	-1.06341	-1.38161	39	H	H26A	-6.49155	-0.36831	-1.87376
17	C	C6	1.303865	-2.83287	-1.68167	40	H	H26B	-6.99295	-0.52359	-0.16966
18	C	C7	2.397222	-3.69673	-1.64306	41	H	H1	4.376084	-4.29876	-1.23222
19	H	H7	2.226	-4.6836	-2.0646	42	H	H8	-6.43359	-1.9526	-1.06145
20	C	C8	3.680288	-3.46949	-1.14854	43	H	H20	0.404699	-3.23745	-2.13715
21	C	C9	4.190396	-2.30851	-0.57273	44	C	C11	5.521872	0.085989	0.689222
22	H	H9	5.235101	-2.34689	-0.27302	45	N	N6	6.626693	0.091934	1.031115
23	C	C10	3.549442	-1.09942	-0.32557						

Table S11. Cartesian coordinates of **4f**; absolute energy is −1504.13247 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O3	-4.82499	-0.61407	1.871172	29	C	C18	-0.65251	0.968551	1.278006
2	O	O4	-4.01665	0.158059	-2.58375	30	C	C19	-1.91178	0.456613	1.72672
3	N	N1	2.990181	4.055577	-2.2808	31	C	C20	-2.859	0.217575	0.722427
4	N	N2	-1.1808	4.48891	-1.32531	32	C	C21	-2.59951	0.475288	-0.6147
5	N	N3	1.007728	1.420232	3.198126	33	C	C22	-1.3925	0.98468	-1.05293
6	N	N4	-2.17936	0.200116	3.025192	34	H	H22	-1.21129	1.184131	-2.10143
7	H	H4NA	-3.10714	-0.10574	3.277187	35	C	C23	0.315927	1.235853	2.289508
8	H	H4NB	-1.52664	0.446328	3.751265	36	C	C24	-4.23902	-0.29055	0.856562
9	N	N5	-4.76021	-0.33055	-0.44123	37	C	C25	-3.82901	0.108841	-1.39397
10	C	C1	4.331821	0.678725	0.426796	38	C	C26	-6.10566	-0.76735	-0.77155
11	C	C2	3.384937	1.634615	0.105216	39	H	H26A	-6.66259	0.041828	-1.2467
12	H	H2	3.563839	2.700201	0.140084	40	H	H26B	-6.59759	-1.05704	0.15509
13	C	C3	2.128761	1.0364	-0.20639	41	H	H1	4.520387	-3.89952	0.695461
14	C	C4	2.304916	-0.38062	-0.09947	42	H	H8	-6.07306	-1.61858	-1.4535
15	C	C5	1.368463	-1.3628	-0.40773	43	C	C11	5.744182	0.942367	0.854721
16	H	H5	0.413371	-0.98744	-0.75808	44	H	H3	6.470905	0.476288	0.180875
17	C	C6	1.442542	-2.76224	-0.3461	45	H	H4	5.943275	0.560763	1.861589
18	C	C7	2.558836	-3.50886	0.051881	46	H	H10	5.9515	2.013535	0.863812
19	H	H7	2.419565	-4.58661	0.04808	47	C	C12	0.180519	-3.53988	-0.73558
20	C	C8	3.829674	-3.09954	0.443768	48	H	H11	-0.55571	-2.80344	-1.07394
21	C	C9	4.344771	-1.80755	0.544186	49	C	C27	0.4221	-4.50625	-1.90856
22	H	H9	5.385494	-1.74069	0.851447	50	H	H6	1.108829	-5.31343	-1.6384
23	C	C10	3.712617	-0.59486	0.307508	51	H	H13	0.841701	-3.98684	-2.77363
24	C	C13	0.9255	1.770009	-0.51222	52	H	H14	-0.51845	-4.97012	-2.2181
25	C	C14	0.923794	2.999902	-1.14753	53	C	C28	-0.43045	-4.2736	0.471952
26	C	C15	2.079422	3.569215	-1.75991	54	H	H12	0.246958	-5.03823	0.86314
27	C	C16	-0.25058	3.806905	-1.24041	55	H	H15	-1.3591	-4.77435	0.184472
28	C	C17	-0.40242	1.227515	-0.08725	56	H	H16	-0.65813	-3.58315	1.287876

Table S12. Cartesian coordinates of **4g**; absolute energy is −1543.45314 au.

		Atom	X	Y	Z			Atom	X	Y	Z
1	O	O3	-5.02319	-0.20019	1.948242	31	C	C20	-3.01053	0.44123	0.755084
2	O	O4	-4.21243	0.387039	-2.53502	32	C	C21	-2.752	0.641209	-0.59208
3	N	N1	3.118599	3.656911	-2.42323	33	C	C22	-1.5131	1.034351	-1.06106
4	N	N2	-0.96952	4.51475	-1.37286	34	H	H22	-1.33115	1.189225	-2.11694
5	N	N3	0.989628	1.372659	3.1381	35	C	C23	0.266464	1.223474	2.247663
6	N	N4	-2.30047	0.431874	3.048206	36	C	C24	-4.42459	0.049387	0.920149
7	H	H4NA	-3.24633	0.209287	3.319784	37	C	C25	-4.01781	0.354133	-1.34589
8	H	H4NB	-1.61822	0.638903	3.759118	38	C	C26	-6.34732	-0.33289	-0.65828
9	N	N5	-4.96766	0.016556	-0.36723	39	H	H26A	-6.50185	-0.20194	-1.72776
10	C	C1	4.182318	0.199781	0.290243	40	H	H26B	-7.02685	0.315509	-0.1038
11	C	C2	3.325001	1.239252	-0.02212	41	H	H1	3.908399	-4.36887	0.665054
12	H	H2	3.603458	2.282346	-0.01937	42	H	H8	-6.54858	-1.37032	-0.38419
13	C	C3	2.004636	0.764749	-0.28246	43	C	C11	5.638506	0.304809	0.657833
14	C	C4	2.038488	-0.65964	-0.14561	44	H	H3	6.23398	-0.26828	-0.065
15	C	C5	0.996494	-1.54792	-0.39646	45	H	H4	5.80418	-0.18548	1.625827
16	H	H5	0.071939	-1.0857	-0.72235	46	C	C12	6.180398	1.734374	0.726609
17	C	C6	0.927192	-2.94484	-0.30048	47	H	H10	7.239659	1.727858	0.994231
18	C	C7	1.974357	-3.79471	0.079643	48	H	H11	5.654952	2.326825	1.480345
19	H	H7	1.726451	-4.85241	0.111929	49	H	H12	6.086539	2.251021	-0.2323
20	C	C8	3.293194	-3.50858	0.416054	50	C	C27	-0.41784	-3.59512	-0.64119
21	C	C9	3.941618	-2.27404	0.465232	51	H	H13	-1.10513	-2.78472	-0.90536
22	H	H9	4.994397	-2.31258	0.73332	52	C	C28	-1.02771	-4.3303	0.565242
23	C	C10	3.430247	-1.00736	0.220708	53	H	H14	-0.42294	-5.19069	0.865804
24	C	C13	0.876847	1.613002	-0.57468	54	H	H15	-2.02508	-4.70437	0.317953
25	C	C14	0.983791	2.829078	-1.22882	55	H	H16	-1.12023	-3.66955	1.430693
26	C	C15	2.175306	3.272657	-1.87532	56	C	C29	-0.32228	-4.51635	-1.86977
27	C	C16	-0.10761	3.746491	-1.30508	57	H	H6	0.327157	-5.3772	-1.68725
28	C	C17	-0.48996	1.212538	-0.11615	58	H	H17	0.071182	-3.98188	-2.73789
29	C	C18	-0.7393	1.014614	1.259022	59	H	H18	-1.31082	-4.90334	-2.13276
30	C	C19	-2.03101	0.627286	1.739531						

4. ORTEP Drawing of 2b (Figures S99).

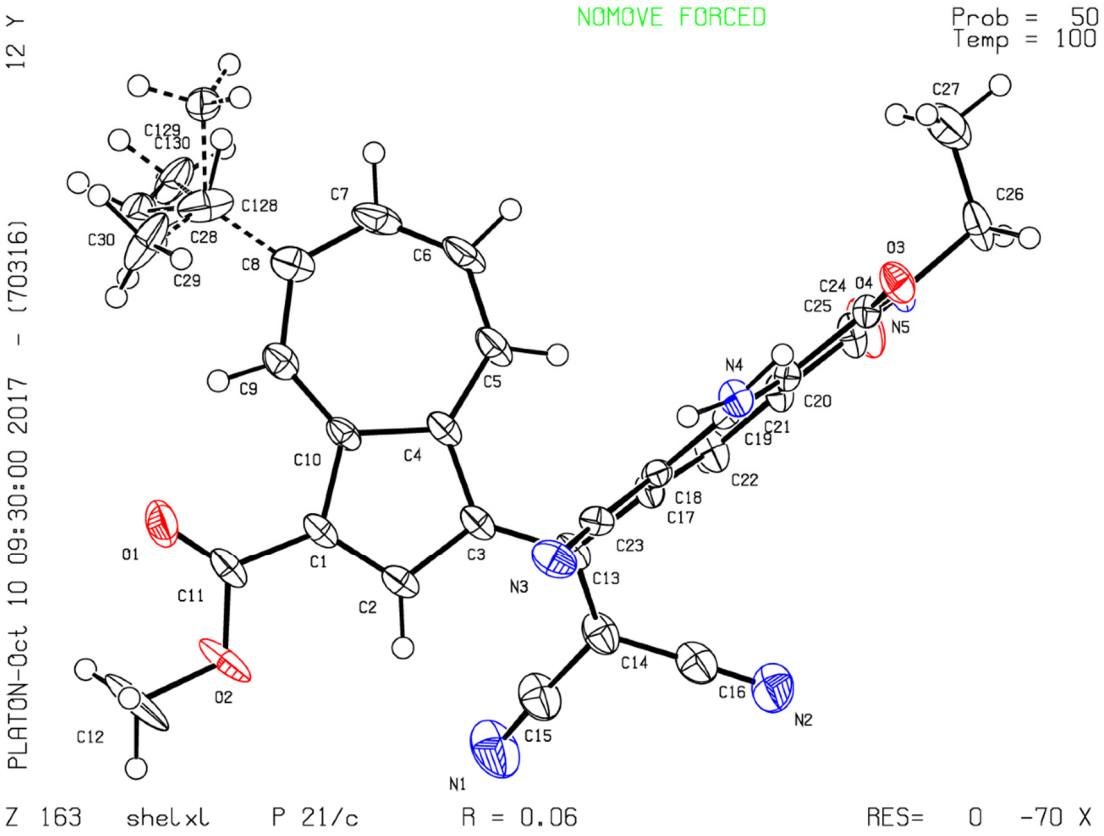


Figure S99. ORTEP diagrams of **2b (CCDC 1579069)**; ellipsoids are drawn at the 50% probability level; recrystallized from CH_2Cl_2/MeOH; monoclinic, $P2_{1/c}$ (#14), a = 15.1789(4) Å, b = 11.1406(3) Å, c = 15.2829(5) Å, β = 96.945(3)°, V = 2565.41(13) Å^3, Z = 4, D_{calc} = 1.340 g/cm^3, μ(MoKa) = 0.915 cm^{-1}, R_1 (I>2.00σ) = 0.0572, R (All reflections) = 0.0776, $_wR^2$ (All reflections) = 0.1321.