Supporting Information for

Synthesis of Core-Modified Third-Generation Light-Driven Molecular Motors

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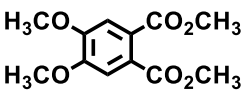
Table of contents

List of all synthesized compounds

dimethyl 4,5-dimethoxyphthalate (11)



11

(white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.15 (s, 2H), 3.90 (s, 6H), 3.84 (s, 6H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 167.89, 150.75, 125.19, 111.42, 56.23, 52.60.

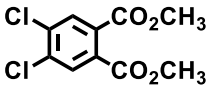
FT-IR (dry powder) (cm^{-1}): 3018 (*C-H*), 2955 (*C-H*), 1709 (*C=O*).

HR-MS (*m/z*): $[M+Na]^+$ calcd for $C_{12}H_{14}O_6Na$ 276.0683; found 276.0680 (0.8 ppm).

M_p 86.2-87.1 ºC.

Compound **12** was previously characterized.[S1]

dimethyl 4,5-dichlorophthalate (13)



13

(white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.78 (s, 2H), 3.88 (s, 6H).

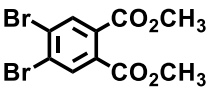
^{13}C-NMR {^{1}H}($CDCl_3$, 150 MHz, δ): 166.04, 135.82, 131.44, 130.99, 53.08.

FT-IR (dry powder) (cm^{-1}): 3096 (*C-H*), 3037 (*C-H*), 2956 (*C-H*), 1720 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{10}H_9Cl_2O_4$ 262.9872; found 262.9873 (0.2 ppm).

M_p 44.5-46 ºC.

dimethyl 4,5-dibromophthalate (14)



14

(white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.96 (s, 2H), 3.91 (s, 6H).

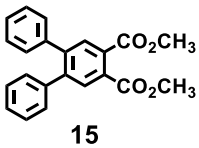
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 166.13, 134.08, 132.03, 128.38, 53.17.

FT-IR (dry powder) (cm^{-1}): 2953 (*C-H*), 2923 (*C-H*), 2852 (*C-H*), 1730 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{10}H_9Br_2O_4$ 352.8842; found 352.8841 (0.2 ppm).

M_p 72.6-74.3 ºC.

dimethyl [1,1':2',1''-terphenyl]-4',5'-dicarboxylate (15)



15

Synthesized with general method G.

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.80 (s, 2H), 7.24-7.23 (m, 6H), 7.15-7.13 (m, 4H), 3.94 (s, 6H).

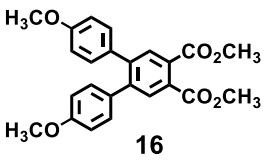
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 167.98, 143.57, 139.71, 131.39, 130.89, 129.76, 128.28, 127.55, 52.84.

FT-IR (dry powder) (cm^{-1}): 2955 (*C-H*), 1721 (*C=O*).

HR-MS (*m/z*): $[M+Na]^+$ calcd for $C_{22}H_{18}O_4Na$ 369.1097; found 369.1094 (0.9 ppm).

M_p 108.8-110.1 ºC.

dimethyl 4,4''-dimethoxy-[1,1':2',1''-terphenyl]-4',5'-dicarboxylate (16)



16

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.74 (s, 2H), 7.07-7.06 (AA'BB' system, 4H), 6.79-6.77 (AA'BB' system, 4H), 3.93 (s, 6H), 3.78 (s, 6H).

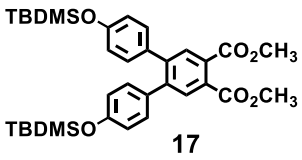
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 168.10, 159.08, 143.04, 132.23, 131.32, 130.89, 130.47, 113.81, 55.34, 52.77.

FT-IR (dry powder) (cm^{-1}): 2955 (*C-H*), 2841 (*C-H*), 1722 (*C=O*).

HR-MS (*m/z*): $[M+Na]^+$ calcd for $C_{24}H_{22}O_6Na$ 429.1309; found 429.1303 (1.4 ppm).

M_p 113.6-115.1 ºC.

dimethyl 4,4''-bis((tert-butyldimethylsilyl)oxy)-[1,1':2',1''-terphenyl]-4',5'-dicarboxylate (17)



17

(transparent oil).

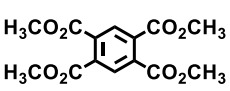
^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.75 (s, 2H), 6.99-6.98 (AA'BB' system, 4H), 6.71-6.69 (AA'BB' system, 4H), 3.93 (s, 6H), 0.97 (s, 18H), 0.18 (s, 12H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 168.16, 155.29, 143.23, 132.89, 131.18, 130.91, 130.45, 120.85, 119.99, 116.04, 52.78, 25.85, 25.81, 18.39, -4.28.

FT-IR (dry liquid) (cm^{-1}): 2954 (*C-H*), 2930 (*C-H*), 2858 (*C-H*), 1728 (*C=O*), 1244 (*Si-O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for HR-MS $C_{34}H_{47}O_6Si_2$ 607.2906; found 607.2887 (3.1 ppm).

tetramethyl benzene-1,2,4,5-tetracarboxylate (18)



18

(white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.07 (s, 2H), 3.94 (s, 12H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 166.47, 134.38, 129.79, 53.22.

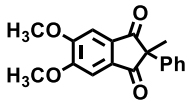
FT-IR (dry powder) (cm^{-1}): 2956 (*C-H*), 1720 (*C=O*).

HR-MS (*m/z*): $[M+Na]^+$ calcd for $C_{14}H_{14}O_8Na$ 333.0581; found 333.0579 (0.6 ppm).

M_p 137.1-138.9 ºC.

Compound **19** was previously reported[S2] and characterized (synthesized in 75% yield).

5,6-dimethoxy-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (20)



20

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.32 (s, 2H), 7.28-7.15 (m, 5H), 3.96 (s, 6H), 1.63 (s, 3H).

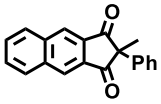
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 201.02, 156.41, 138.42, 136.47, 129.42, 128.82, 128.80, 128.77, 127.57, 126.75, 103.95, 57.63, 56.84, 19.97.

FT-IR (dry powder) (cm^{-1}): 3012 (*C-H*), 2979 (*C-H*), 2959 (*C-H*), 1688 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{18}H_{18}O_4$ 297.1121; found 297.1120 (0.5 ppm).

M_p 168.2-170.7 ºC.

2-methyl-2-phenyl-1H-cyclopenta[b]naphthalene-1,3(2H)-dione (21)



21

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.58 (s, 2H), 8.12-8.11 (m, 2H), 7.73-7.71 (m, 2H), 7.39-7.38 (m, 2H), 7.31-7.23 (m, 3H), 1.78 (s, 3H).

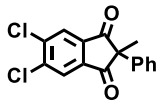
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 202.30, 138.22, 136.87, 136.32, 130.71, 129.84, 128.95, 127.70, 126.88, 125.20, 59.65, 20.26.

FT-IR (dry powder) (cm^{-1}): 2979 (*C-H*), 2934 (*C-H*), 2867 (*C-H*), 1688 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{20}H_{16}O_2$ 287.1067; found 287.1065 (0.5 ppm).

M_p 113.1-115.6 ºC.

5,6-dichloro-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (22)



22

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.07 (s, 2H), 7.28-7.21 (m, 5H), 1.66 (s, 3H).

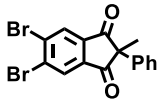
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 199.73, 141.64, 140.07, 137.17, 129.12, 128.07, 126.67, 125.74, 58.46, 20.27.

FT-IR (dry powder) (cm^{-1}): 2929 (*C-H*), 1709 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{16}H_{12}Cl_2O_2$ 305.0131; found 305.0129 (0.4 ppm).

M_p 104.9-106.8 ºC.

5,6-dibromo-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (23)



23

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.25 (s, 2H), 7.27-7.21 (m, 5H), 1.66 (s, 3H).

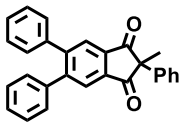
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 199.81, 140.52, 137.14, 134.46, 129.13, 129.12, 128.96, 128.08, 126.68, 58.44, 20.25.

FT-IR (dry powder) (cm^{-1}): 3072 (*C-H*), 2934 (*C-H*), 1707 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{16}H_{12}Br_2O_2$ 392.9120; found 392.9189 (0.4 ppm).

M_p 145.6-147.1 ºC.

2-methyl-2,5,6-triphenyl-1H-indene-1,3(2H)-dione (24)



24

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.07 (s, 2H), 7.41-7.39 (m, 2H), 7.34-7.31 (m, 2H), 7.27-7.23 (m, 7H), 7.16-7.14 (m, 4H), 1.76 (s, 3H).

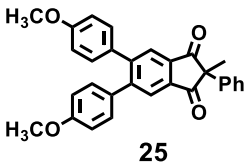
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 201.83, 149.19, 140.28, 139.66, 138.07, 129.71, 128.99, 128.44, 128.06, 127.78, 126.88, 125.89, 58.58, 20.26.

FT-IR (dry powder) (cm^{-1}): 3060 (*C-H*), 1707 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{22}H_{22}O_2$ 389.1536; found 389.1534 (0.4 ppm).

M_p 195.3-197.2 ºC.

5,6-bis(4-methoxyphenyl)-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (25)



25

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.02 (s, 2H), 7.40-7.39 (m, 2H), 7.34-7.31 (m, 2H), 7.27-7.25 (m, 1H), 7.10-7.09 (AA'BB' system, 4H), 6.82-6.80 (AA'BB' system, 4H), 3.80 (s, 6H), 1.75 (s, 3H).

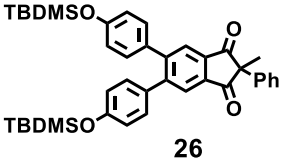
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 201.94, 159.51, 148.74, 139.99, 138.21, 132.17, 130.96, 128.96, 127.72, 126.88, 125.72, 114.00, 58.54, 55.39, 20.19.

FT-IR (dry powder) (cm^{-1}): 3059 (*C-H*), 2961 (*C-H*), 1701 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{30}H_{25}O_4$ 449.1747; found 449.1738 (2.0 ppm).

M_p 196.2-198.7 ºC.

5,6-bis(4-((tert-butyldimethylsilyl)oxy)phenyl)-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (26)



26

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.02 (s, 2H), 7.40-7.39 (m, 2H), 7.34-7.31 (m, 2H), 7.27-7.25 (m, 1H), 7.02-7.01 (AA'BB' system, 4H), 6.74-6.73 (AA'BB' system, 4H), 1.75 (s, 3H), 0.98 (s, 18H), 0.19 (s, 12H).

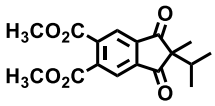
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 201.95, 155.80, 148.91, 140.01, 138.23, 132.85, 130.97, 129.65, 128.96, 128.87, 127.72, 126.89, 125.60, 120.18, 58.55, 25.81, 20.20, 18.41, -4.26.

FT-IR (dry powder) (cm^{-1}): 2931 (*C-H*), 2858 (*C-H*), 1703 (*C=O*), 1266 (*Si-O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{40}H_{49}O_4Si_2$ 649.3164; found 649.3147 (2.6 ppm).

M_p 134.2-136.5 ºC.

dimethyl 2-isopropyl-2-methyl-1,3-dioxo-2,3-dihydro-1H-indene-5,6-dicarboxylate (27)



27

(yellow solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.25 (s, 2H), 3.96 (s, 6H), 2.16 (h, *J* = 6 Hz, 1H), 1.27 (s, 3H), 0.91 (d, *J* = 6 Hz, 6H).

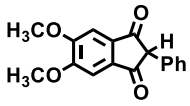
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 203.53, 166.45, 142.70, 138.64, 123.94, 57.52, 53.40, 34.67, 18.12, 17.37.

FT-IR (dry powder) (cm^{-1}): 2957 (*C-H*), 2876 (*C-H*), 1729 (*C=O*), 1710 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{17}H_{19}O_6$ 319.1176; found 319.1175 (0.4 ppm).

M_p 92.2-94.6 ºC.

5,6-dimethoxy-2-phenyl-1H-indene-1,3(2H)-dione (29)



29

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 7.40 (s, 2H), 7.34-7.28 (m, 3H), 7.18-7.16 (m, 2H), 4.20 (s, 1H), 4.04 (s, 6H).

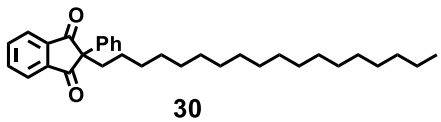
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 197.45, 156.35, 137.98, 133.94, 129.08, 128.79, 127.86, 103.81, 59.59, 56.90.

FT-IR (dry powder) (cm^{-1}): 3006 (*C-H*), 2946 (*C-H*), 1686 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{17}H_{15}O_4$ 283.0965; found 283.0961 (1.3 ppm).

M_p 101.2-103.5 ºC.

2-octadecyl-2-phenyl-1H-indene-1,3(2H)-dione (30)



30

(off-white solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, δ): 8.03-8.02 (m, 2H), 7.86-7.85 (m, 2H), 7.42-7.40 (m, 2H), 7.30-7.28 (m, 2H), 7.24-7.22 (m, 1H), 2.25 (t, *J* = 9 Hz, 2H), 1.30-1.13 (m, 35H), 0.88 (t, *J* = 6 Hz, 3H).

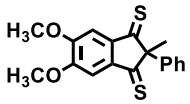
^{13}C-NMR {^{1}H} ($CDCl_3$, 150 MHz, δ): 202.17, 142.25, 137.41, 136.00, 128.89, 127.70, 126.97, 123.67, 62.48, 36.51, 32.07, 30.13, 29.84, 29.80, 29.76, 29.74, 29.69, 29.62, 29.51, 29.30, 25.38, 22.84, 14.26.

FT-IR (dry powder) (cm^{-1}): 2949 (*C-H*), 2916 (*C-H*), 2850 (*C-H*), 1705 (*C=O*).

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{33}H_{47}O_2$ 475.3571; found 475.3557 (2.9 ppm).

M_p 63.2-64.3 ºC.

5,6-dimethoxy-2-methyl-2-phenyl-1H-indene-1,3(2H)-dithione (32)

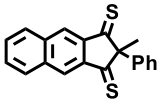


32

(green solid).

^{1}H-NMR ($CDCl_3$, 300 MHz, δ): 7.40 (s, 2H), 7.21-7.17 (m, 5H), 4.09 (s, 6H), 1.92 (s, 3H).

2-methyl-2-phenyl-1H-cyclopenta[b]naphthalene-1,3(2H)-dithione (33)

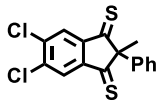


33

(green solid).

^{1}H-NMR ($CDCl_3$, 300 MHz, δ): 8.63 (s, 2H), 8.15-8.12 (m, 2H), 7.72-7.69 (m, 2H), 7.22-7.12 (m, 5H), 2.00 (s, 3H).

5,6-dichloro-2-methyl-2-phenyl-1H-indene-1,3(2H)-dithione (34)

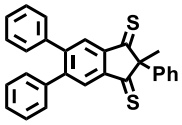


34

(green solid).

^{1}H-NMR ($CDCl_3$, 400 MHz, δ): 8.15 (s, 2H), 7.24-7.15 (m, 5H), 1.91 (s, 3H).

2-methyl-2,5,6-triphenyl-1H-indene-1,3(2H)-dithione (35)

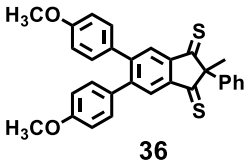


35

(green solid).

^{1}H-NMR ($CDCl_3$, 400 MHz, δ): 8.14 (s, 2H), 7.31-7.29 (m, 10H), 7.25-7.22 (m, 5H), 2.01 (s, 3H).

5,6-bis(4-methoxyphenyl)-2-methyl-2-phenyl-1H-indene-1,3(2H)-dithione (36)

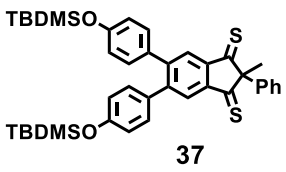


36

(green solid).

^{1}H-NMR ($CDCl_3$, 400 MHz, δ): 8.05 (s, 2H), 7.28-7.21 (m, 5H), 7.16-7.14 (AA'BB'system, 4H), 6.83-6.80 (AA'BB'system, 4H), 3.81 (s, 6H), 1.96 (s, 3H).

5,6-bis(4-((tert-butyldimethylsilyl)oxy)phenyl)-2-methyl-2-phenyl-1H-indene-1,3(2H)-dione (37)

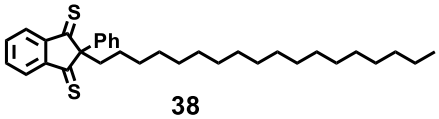


37

(green solid).

^{1}H-NMR ($CDCl_3$, 400 MHz, δ): 8.06 (s, 2H), 7.25-7.20 (m, 5H), 7.08-7.06 (AA'BB' system, 4H), 6.75-6.73 (AA'BB' system, 4H), 1.97 (s, 3H), 0.98 (s, 18H), 0.19 (s, 12H).

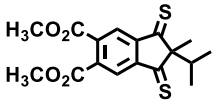
2-octadecyl-2-phenyl-1H-indene-1,3(2H)-dithione (38)



38

Synthesized with general method D. The compound was obtained in a ternary mixture (860 mg) composed by 15% desired product **38**, 46% starting indanedione **30**, and 39% product of single conversion. This material was used in the next step without further purification, adjusting the amount of 9-diazo9H-fluorenone necessary in the B-K olefination.

dimethyl 2-isopropyl-2-methyl-1,3-dioxo-2,3-dihydro-1H-indene-5,6-dicarboxylate (27)



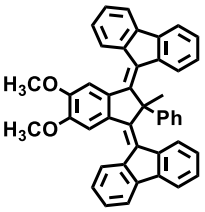
39

(green solid).

^{1}H-NMR ($CDCl_3$, 400 MHz, δ): 8.27 (s, 2H), 3.96 (m, 6H), 2.41 (h, *J* = 7 Hz, 1H), 1.52 (s, 3H), 0.86 (d, *J* = 7 Hz, 6H).

Reference motor **1** was previously reported[S2] and characterized.

(2)



2

(deep red solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ) (signals of the main isomer): 8.41 (d, *J* = 6 Hz, 2H), 7.80 (d, *J* = 6 Hz, 2H), 7.72 (d, *J* = 6 Hz, 2H), 7.68 (d, *J* = 6 Hz, 2H), 7.65 (s, 2H), 7.30 (t, *J* = 6Hz, 3H), 7.27 (t, *J* = 6Hz, 3H), 7.18 (t, *J* = 6Hz, 3H), 7.13 (t, *J* = 6Hz, 3H), 3.86 (s, 6H), 2.44 (s, 3H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 160.69, 157.25, 154.35, 152.20, 150.17, 149.70, 142.21, 140.95, 140.39, 140.12, 139.58, 139.54, 138.86, 137.61, 137.38, 137.32, 132.95, 130.44, 128.69, 128.60, 127.97, 127.53, 127.28, 127.17, 127.04, 126.83, 126.73, 126.67, 126.58, 126.20, 126.05, 125.77, 123.61, 119.92, 119.60, 119.53, 119.39, 110.67, 105.38, 71.39, 68.75, 56.91, 56.70, 56.57, 23.38, 19.44.

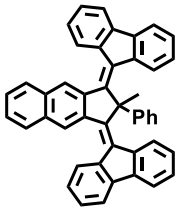
HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{44}H_{34}O_2$ 593.2475; found 593.2461 (2.3 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 247 (149200), 471 (69400).

M_p 260-262 ºC.

Single crystals for XRD were obtained from slow diffusion of hexane (antisolvent) into a saturated solution of 1,2-dichloroethane (solvent).

(3)



3

(orange solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ): 8.72 (s, 2H), 8.64 (d, *J* = 6 Hz, 2H), 7.84-7.82 (m, 4H), 7.80-7.78 (m, 2H), 7.71 (d, *J* = 6 Hz, 2H), 7.68-7.67 (m, 2H), 7.57-7.55 (m, 4H), 7.73 (t, *J* = 6 Hz, 3H), 7.29-7.26 (m, 3H), 7.15-7.12 (m, 5H), 2.48 (s, 3H).

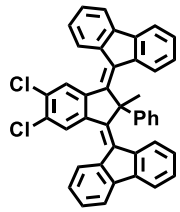
^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 159.46, 156.58, 143.98, 141.81, 141.62, 140.65, 140.18, 140.07, 140.00, 139.57, 139.44, 137.52, 135.59, 134.33, 134.01, 133.55, 133.50, 131.96, 130.87, 129.54, 128.97, 128.86, 128.79, 128.61, 127.96, 127.78, 127.67, 127.51, 127.31, 127.22, 126.54, 126.43, 126.25, 125.21, 125.05, 123.07, 122.87, 119.63, 119.50, 119.41, 118.82, 70.01, 67.63, 20.11, 19.67.

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{46}H_{31}$ 583.2420; found 583.2410 (1.8 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 244 (239352), 400 (94064).

M_p > 300 ºC.

(4)



4

(orange solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ) (signals of the main isomer): 8.35 (d, *J* = 6 Hz, 2H), 8.32 (s, 2H), 7.73-7.68 (m, 5H), 7.63 (d, *J* = 6 Hz, 2H), 7.34 (t, *J* = Hz, 3H), 7.26 (t, *J* = 6 Hz, 3H), 7.21 (t, *J* = 6 Hz, 3H), 7.10 (t, *J* = 6 Hz, 3H), 2.42 (s, 3H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 157.14, 154.41, 154.16, 147.21, 144.65, 140.84, 140.39, 140.26, 140.18, 139.83, 139.65, 138.46, 138.41, 137.13, 135.31, 135.01, 134.41, 133.97, 133.10, 132.71, 130.74, 130.41, 129.64, 128.76, 128.33, 128.16, 128.07, 127.88, 127.74, 127.46, 126.61, 126.44, 125.42, 125.32, 124.40, 123.51, 123.27, 122.81, 120.35, 120.25, 119.80, 119.65, 119.54, 119.05, 70.71, 68.49, 19.70, 18.91.

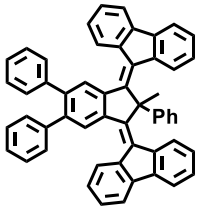
HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{42}H_{27}Cl_2$ 601.1484; found 601.1481 (0.5 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 241 (156766), 441 (59518).

M_p 269-271 ºC.

Single crystals for XRD were obtained from slow diffusion of hexane (antisolvent) into a saturated solution of 1,2-dichloroethane (solvent).

(5)



5

(red solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ) (signals of the main isomer): 8.63 (d, *J* = 6 Hz, 2H), 8.30 (s, 2H), 7.80 (d, *J* = 6 Hz, 3H), 7.69-7.64 (m, 6H), 7.31-7.11 (m, 20H), 2.50 (s, 3H).

^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 159.78, 156.61, 147.01, 144.58, 141.53, 141.29, 140.53, 140.25, 140.07, 140.00, 139.97, 139.79, 139.58, 139.15, 137.51, 135.63, 133.19, 132.73, 131.90, 131.40, 129.81, 129.76, 129.31, 128.88, 128.68, 128.50, 128.29, 127.99, 127.74, 127.64, 127.57, 127.51, 127.26, 127.05, 126.90, 126.65, 126.35, 126.26, 126.14, 125.10, 123.67, 123.34, 119.57, 119.40, 118.91, 70.73, 68.57, 19.88, 19.42.

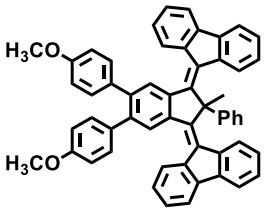
HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{54}H_{37}$ 685.2890; found 685.2881 (1.3 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 246 (209192), 391 (63390), 449 (86394).

M_p > 300 ºC.

Single crystals for XRD were obtained from slow diffusion of hexane (antisolvent) into a saturated solution of 1,2-dichloroethane (solvent).

(6)



6

(red solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ) (signals of the main isomer): 8.62 (d, *J* = 6 Hz, 2H), 8.25 (s, 2H), 7.79 (d, *J* = 6 Hz, 2H), 7.68 (d, *J* = 6 Hz, 2H), 7.64 (d, *J* = 6 Hz, 2H), 7.29 (t, *J* = Hz, 3H), 7.25 (t, *J* = 6 Hz, 3H), 7.18 (t, *J* = 6 Hz, 3H), 7.15 (d, *J* = 6 Hz, 4H), 7.12 (t, *J* = 6 Hz, 3H), 6.80 (d, *J* = 6 Hz, 4H), 3.82 (s, 6H), 2.49 (s, 3H).

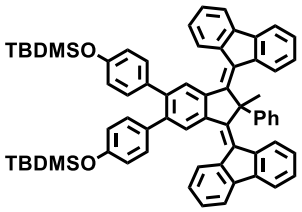
^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 160.05, 158.10, 158.00, 156.86, 146.64, 144.25, 141.39, 141.05, 140.82, 140.60, 139.90, 139.72, 139.51, 139.21, 137.53, 135.65, 132.82, 132.75, 132.57, 132.38, 131.70, 130.97, 130.90, 130.54, 128.66, 127.62, 127.40, 127.21, 126.68, 126.30, 126.22, 125.06, 123.67, 123.33, 119.54, 119.37, 113.18, 70.64, 68.49, 55.29, 19.85, 19.47.

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{56}H_{41}O_2$ 745.3101; found 745.3088 (1.7 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 244 (206000), 395 (69400), 450 (84200).

M_p 268-270 ºC.

(7)



7

(red solid).

^{1}H-NMR ($Cl_2DCCDCl_2$, 500 MHz, 90 ºC, δ) (signals of the main isomer): 8.62 (d, *J* = 6 Hz, 2H), 8.26 (s, 2H), 7.79 (d, *J* = 6 Hz, 2H), 7.68 (d, *J* = 6 Hz, 2H), 7.65 (d, *J* = 6 Hz, 2H), 7.30 (t, *J* = Hz, 3H), 7.25 (t, *J* = 6 Hz, 3H), 7.18 (t, *J* = 6 Hz, 3H), 7.12 (t, *J* = 6 Hz, 3H), 7.09 (d, *J* = 6 Hz, 4H), , 6.72 (d, *J* = 6 Hz, 4H), 2.49 (s, 3H), 1.03 (s, 18 H), 0.23 (s, 12H).

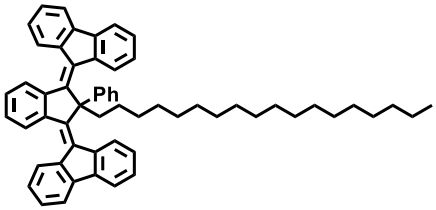
^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 160.11, 156.93, 154.44, 154.31, 146.64, 144.21, 141.33, 141.09, 140.65, 139.93, 139.87, 139.72, 139.47, 139.25, 137.55, 135.65, 133.48, 133.32, 132.79, 132.35, 131.54, 130.99, 130.91, 128.66, 127.56, 127.39, 127.21, 126.65, 126.22, 123.36, 119.71, 119.53, 119.36, 70.72, 68.55, 29.89, 25.60, 19.85, 19.56, 18.24, -4.41.

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{66}H_{65}O_2Si_2$ 945.4518; found 945.4505 (1.4 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 246 (178420), 396 (59400), 451 (72400).

M_p 248-251.6 ºC.

(8)



8

(orange solid).

^{1}H-NMR ($CDCl_3$, 500 MHz, -45 ºC, δ): 8.45 (d, *J* = 8.0 Hz, 2H), 8.24-8.22 (m, 2H), 7.75 (d, *J* = 8.0 Hz, 2H), 7.71-7.67 (m, 4H), 7.39-7.25 (m, 11H), 7.15 (t, *J* = 8 Hz, 2H), 7.19 (t, *J* = 8 Hz, 2H), 3.04 (m, 2H), 1.23 (m, 20H), 1.01 (m, 2H), 0.87 (m, 5H), 0.74 (m, 2H), 0.67 (m, 2H), 0.56 (m, 2H), 0.44 (m, 2H), 0.34 (m, 2H).

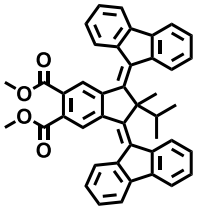
^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 155.08, 148.67, 141.04, 140.01, 139.78, 138.83, 137.38, 133.29, 130.46, 129.14, 128.49, 127.53, 127.45, 127.36, 126.79, 126.09, 126.00, 123.55, 119.38, 119.30, 72.06, 32.06, 30.72, 29.95, 29.93, 29.88, 29.85, 29.65, 29.62, 29.54, 29.02, 28.92, 27.87, 24.74, 22.91, 14.51.

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{59}H_{63}$ 771.4924; found 771.4913 (1.5 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 241 (105922), 453 (40224).

M_p 194.2-196.6 ºC.

(9)



9

(orange solid).

^{1}H-NMR ($CDCl_3$, 600 MHz, 25 ºC, δ): 8.44 (s, 2H), 8.20 (d, *J* = 6 Hz, 2H), 8.01 (d, *J* = 6 Hz, 2H), 7.77 (d, *J* = 6 Hz, 2H), 7.73 (d, *J* = 6 Hz, 2H), 7.39 (t, *J* = 6 Hz, 2H), 7.34-7.30 (m, 6H), 7.13 (t, *J* = 6 Hz, 2H), 3.89 (s, 6H), 3.01 (h, *J* = 6 Hz, 1H), 2.39 (s, 3H), 1.07 (d, *J* = 6 Hz, 6H). [HMPA present in the sample due to a strong interaction with the compound; the doublet at 2.65 in the ^{1}H-NMR spectrum belongs to HMPA]

^{13}C-NMR {^{1}H} ($CDCl_3$, 125 MHz, -45 ºC, δ): 167.84, 155.82, 150.23, 145.21, 140.95, 139.72, 139.60, 138.08, 136.10, 134.32, 131.15, 129.09, 128.94, 128.30, 128.17, 127.82, 127.35, 126.72, 126.36, 125.18, 123.53, 120.05, 119.74, 119.63, 75.05, 69.98, 53.35, 39.88, 28.84, 24.17.

HR-MS (*m/z*): $[M+H]^+$ calcd for $C_{43}H_{35}O_4$ 615.2530; found 615.2517 (2.0 ppm).

UV-Vis (CH_2Cl_2) λ_{max}, nm (ε): 238 (196366), 374 (49228), 440 (65830).

M_p > 300 ºC.

^{1}H and ^{13}C NMR spectra

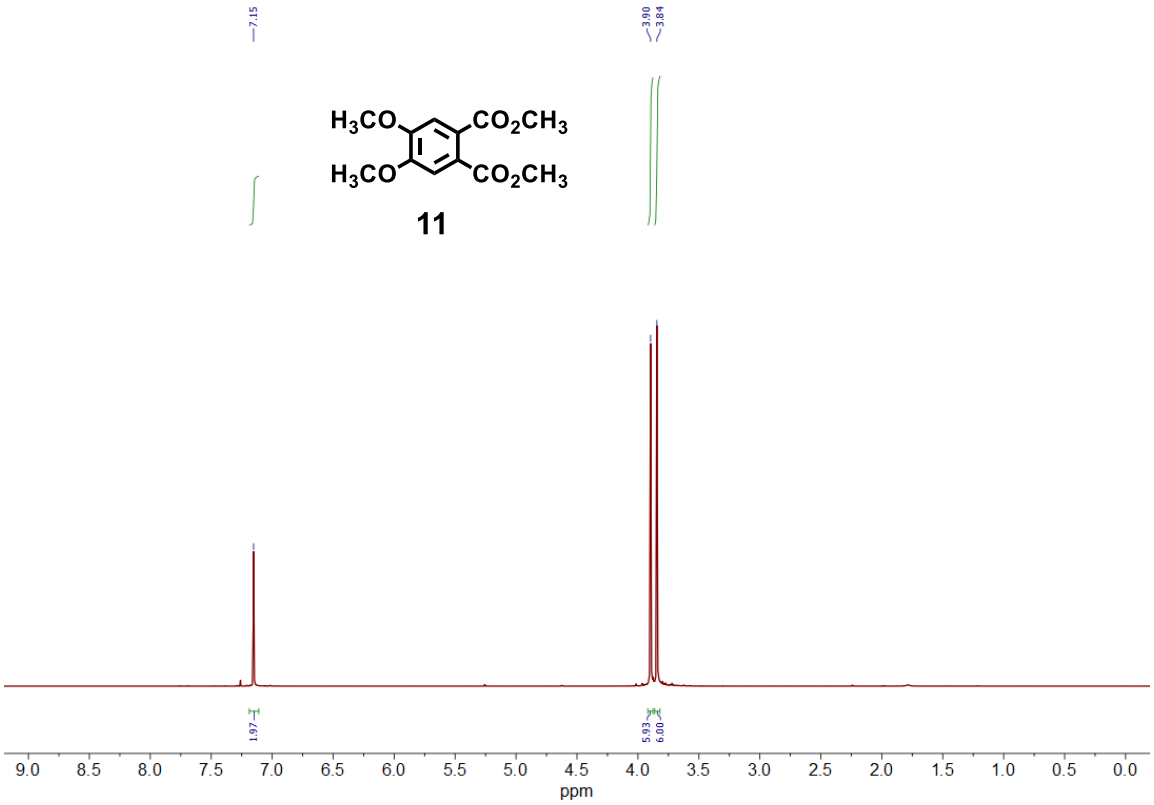


Figure S1. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 11.

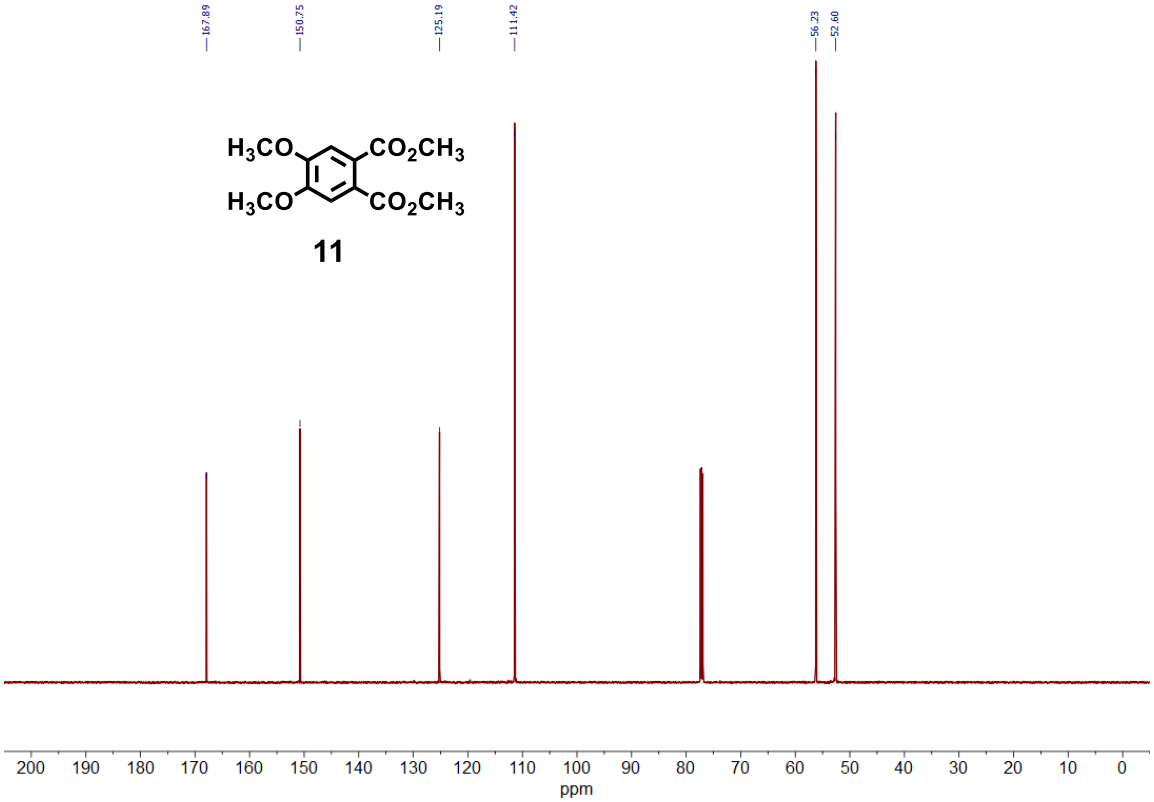


Figure S2. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 11.

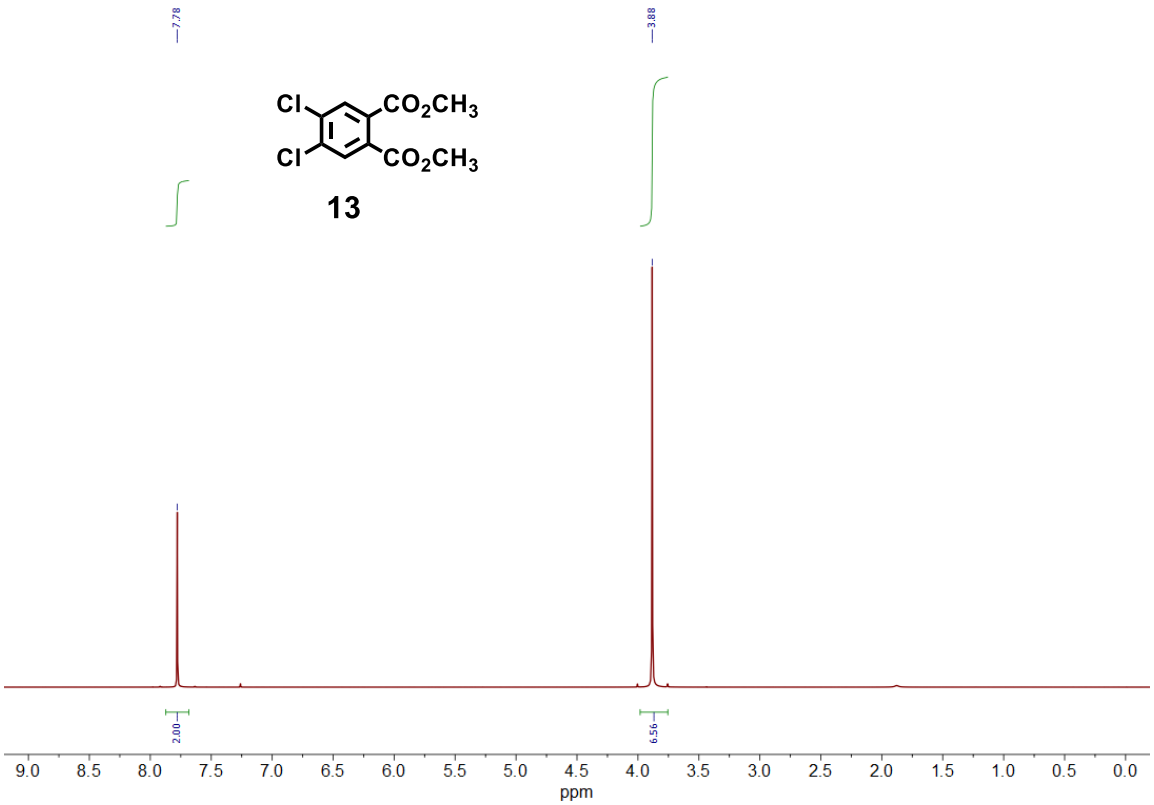


Figure S3. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 13.

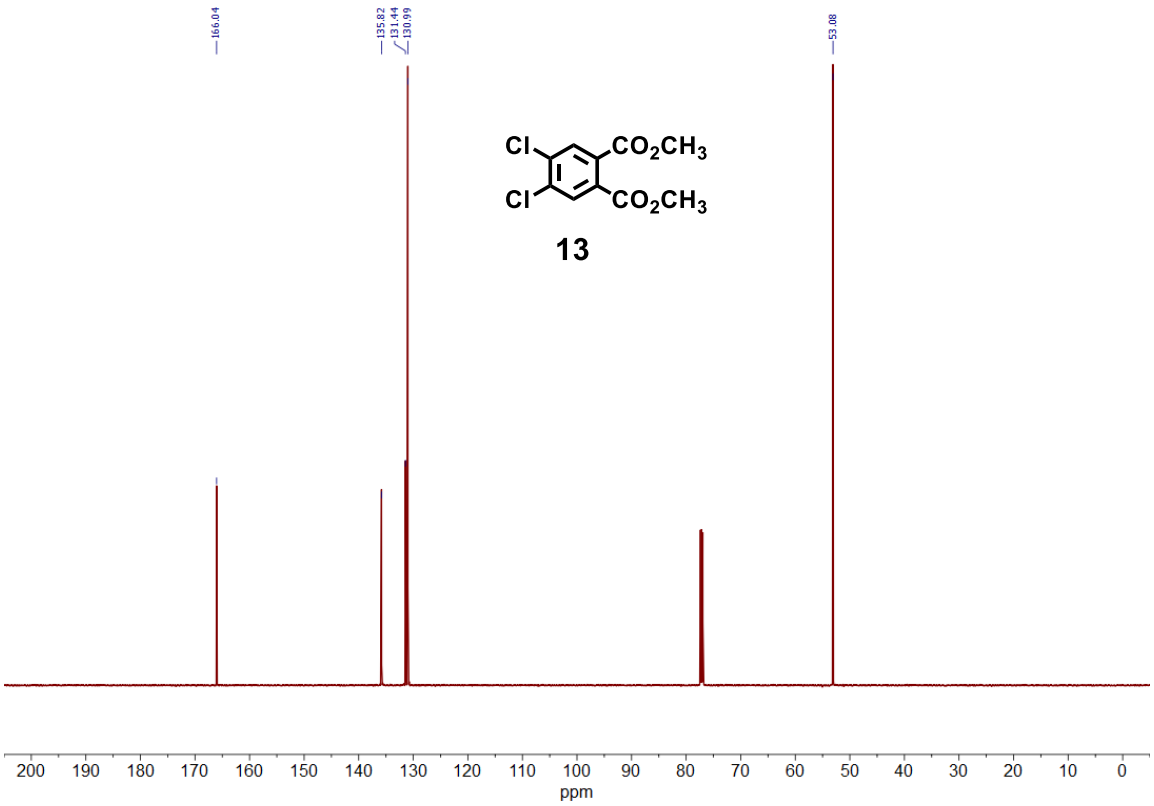


Figure S4. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 13.

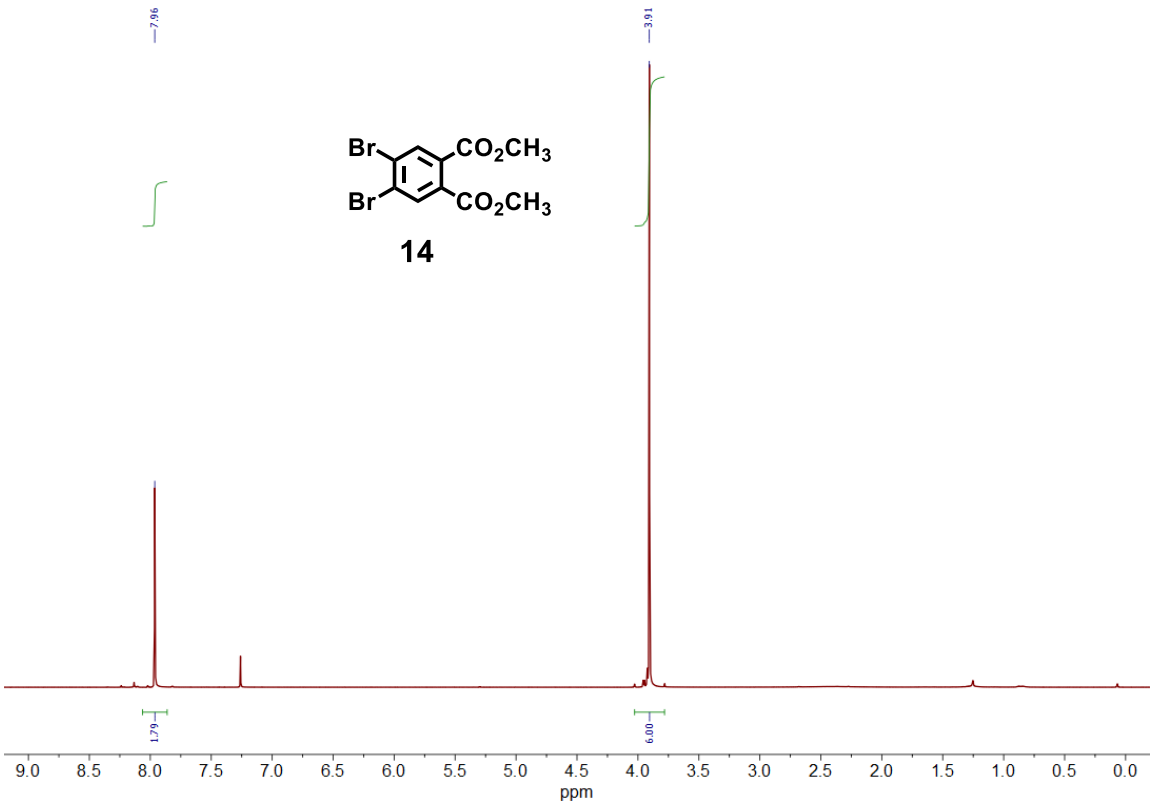


Figure S5. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 14.

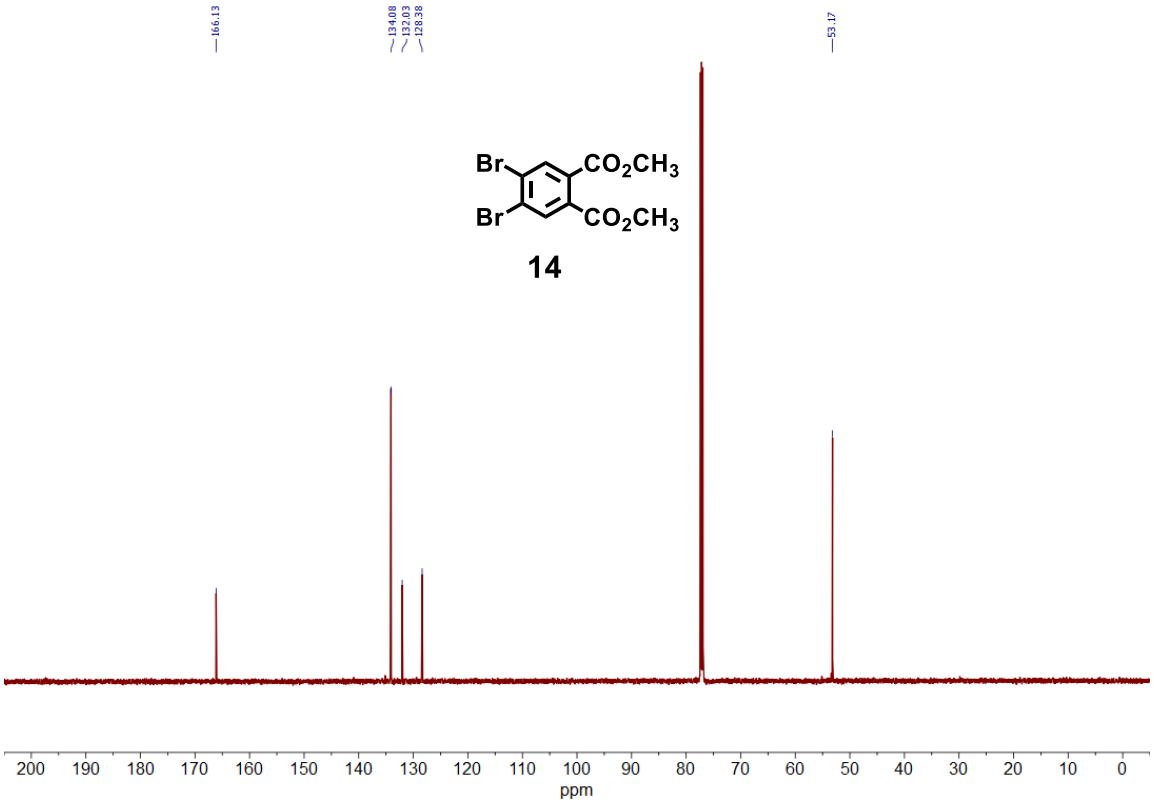


Figure S6. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 14.

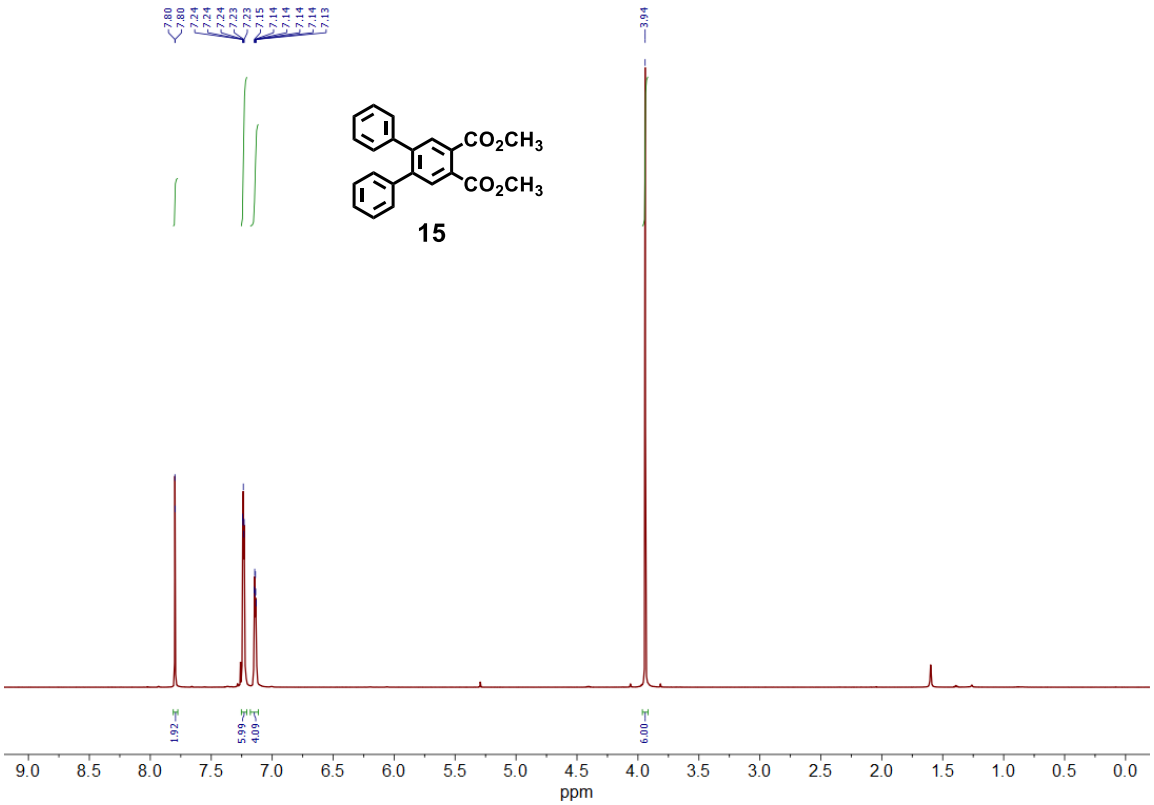


Figure S7. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 15.

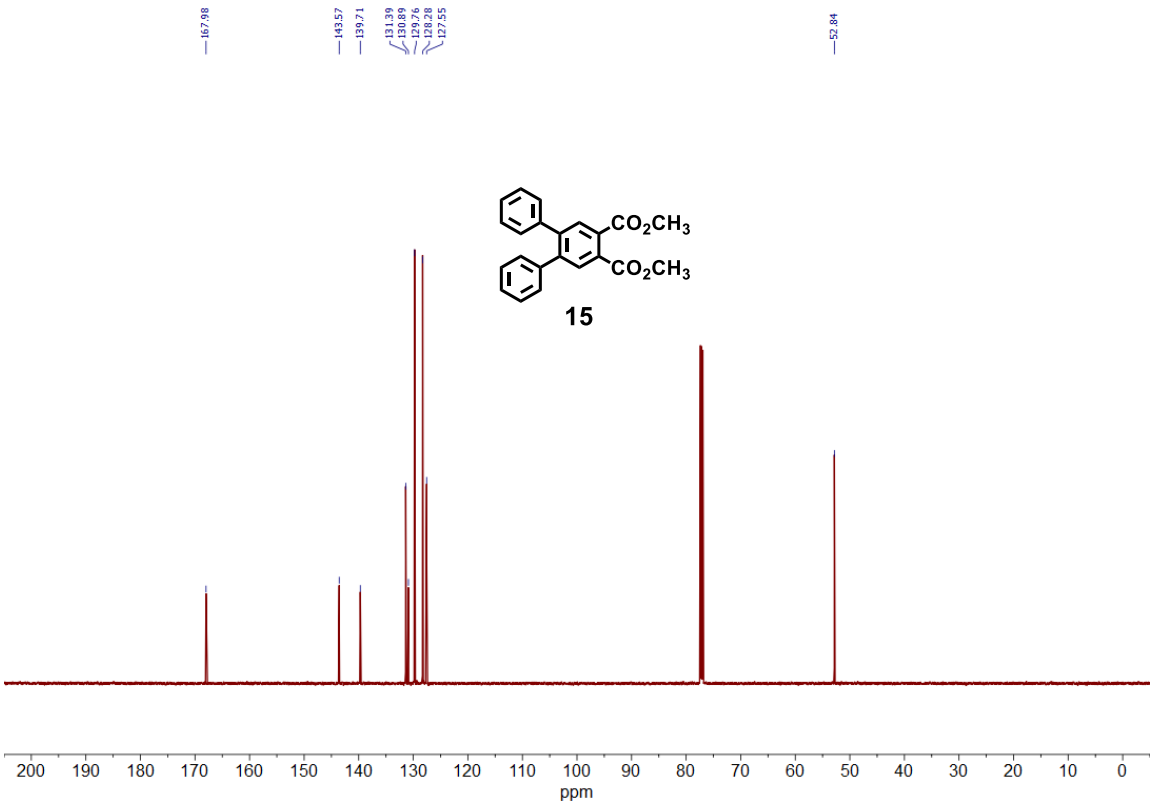


Figure S8. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 15.

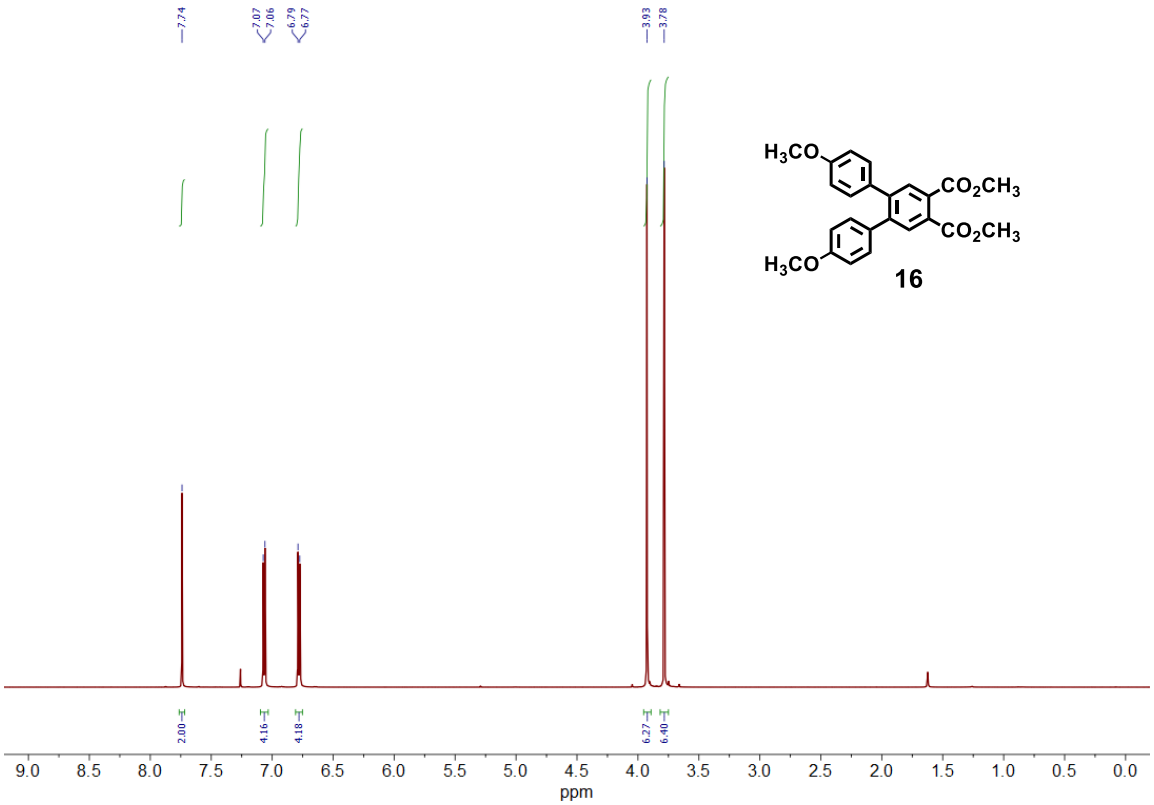


Figure S9. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 16.

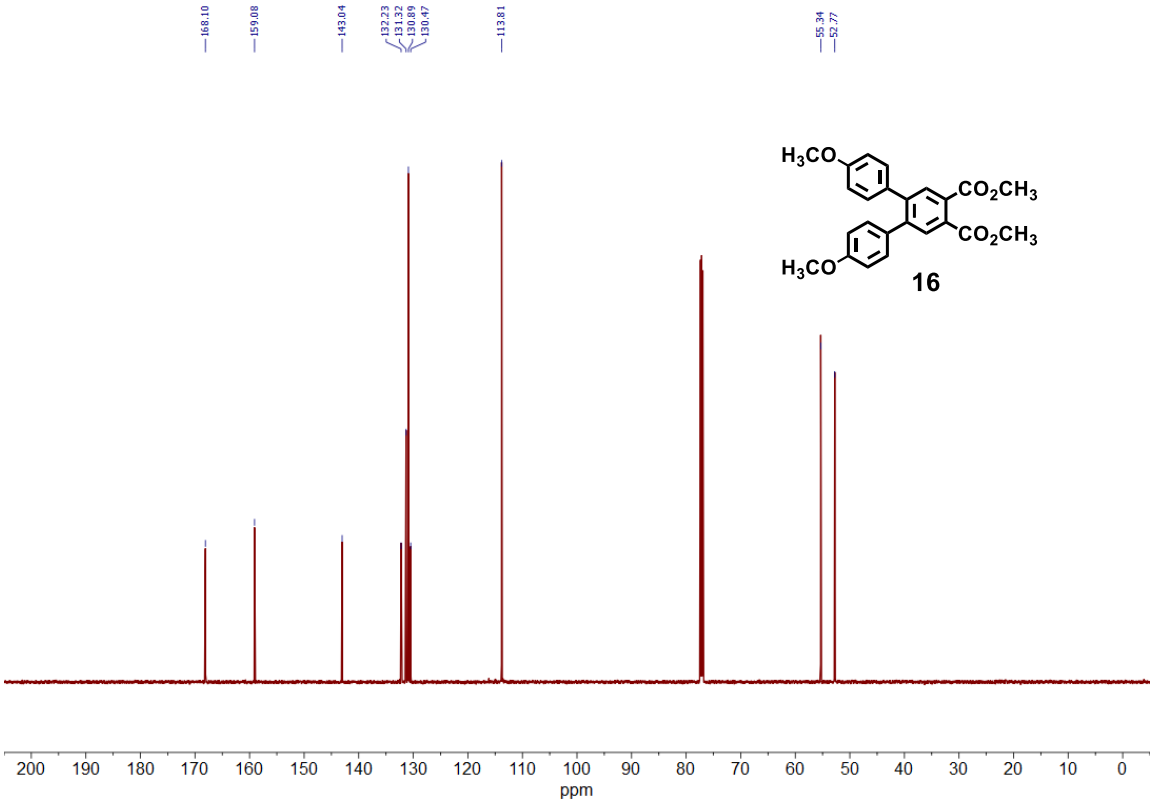


Figure S10. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 16.

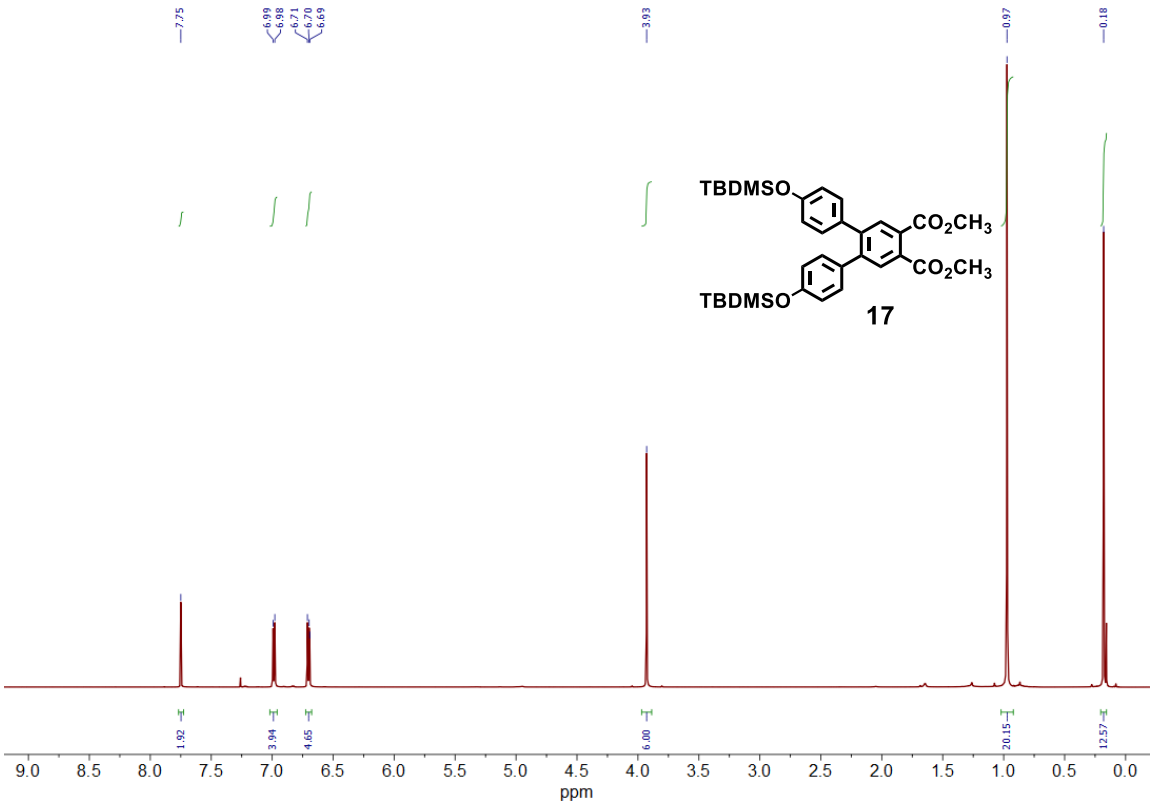


Figure S11. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 17.

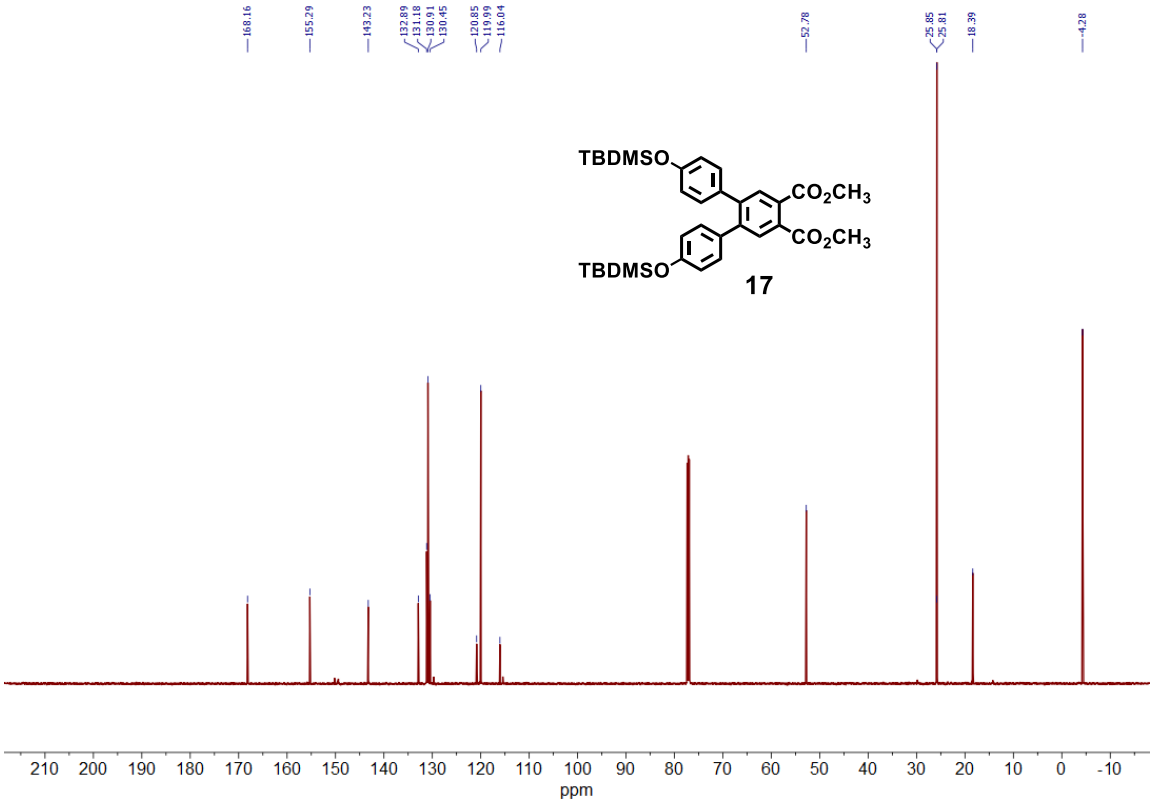


Figure S12. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 17.

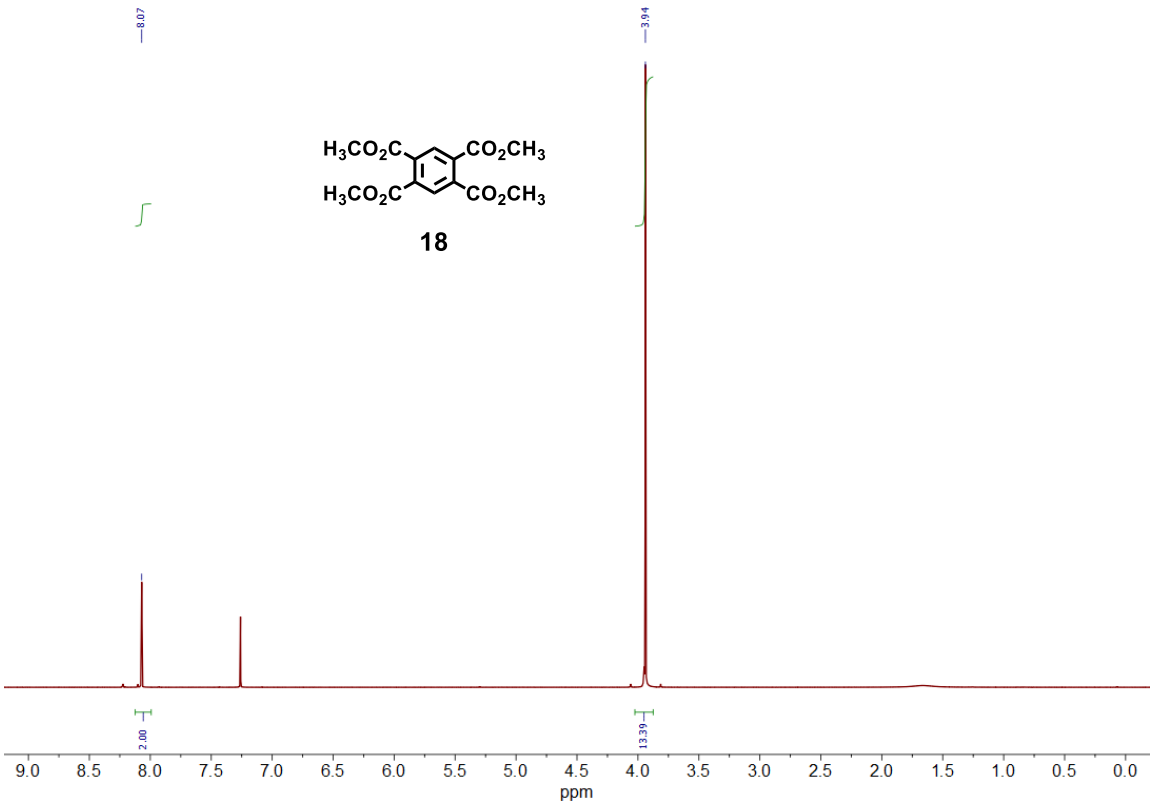


Figure S13. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 18.

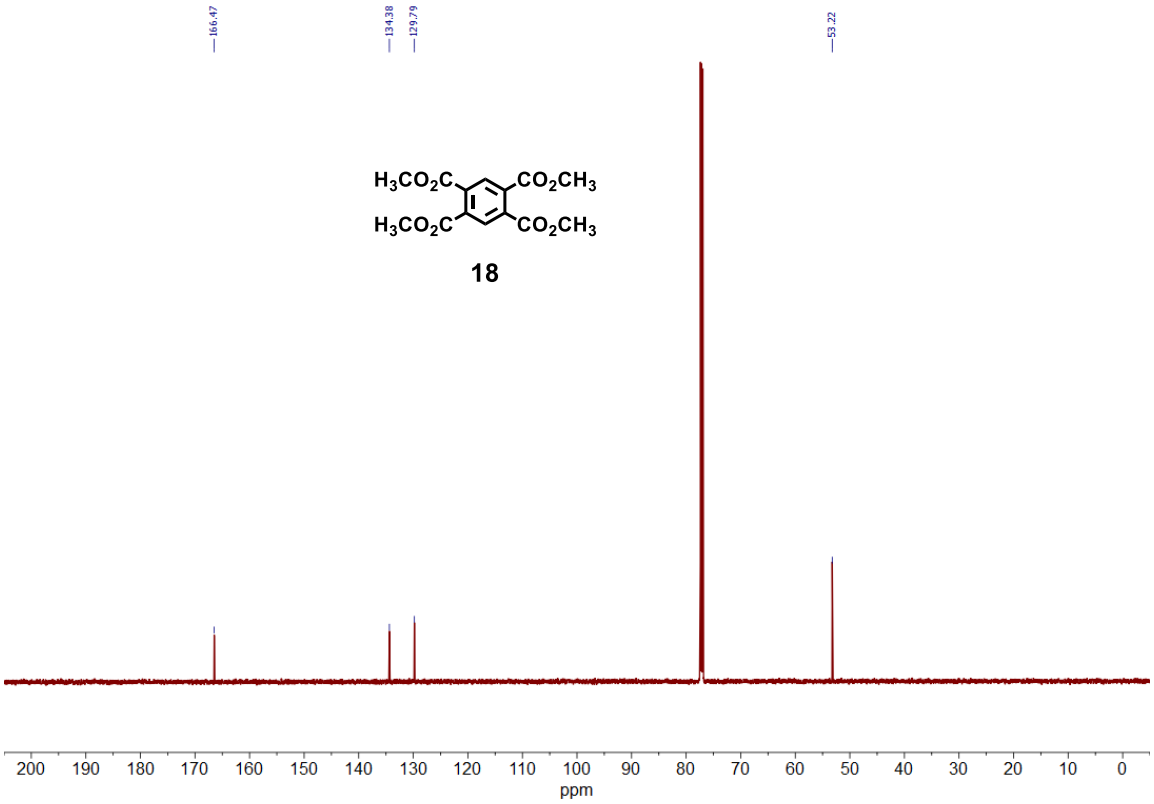


Figure S14. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 18.

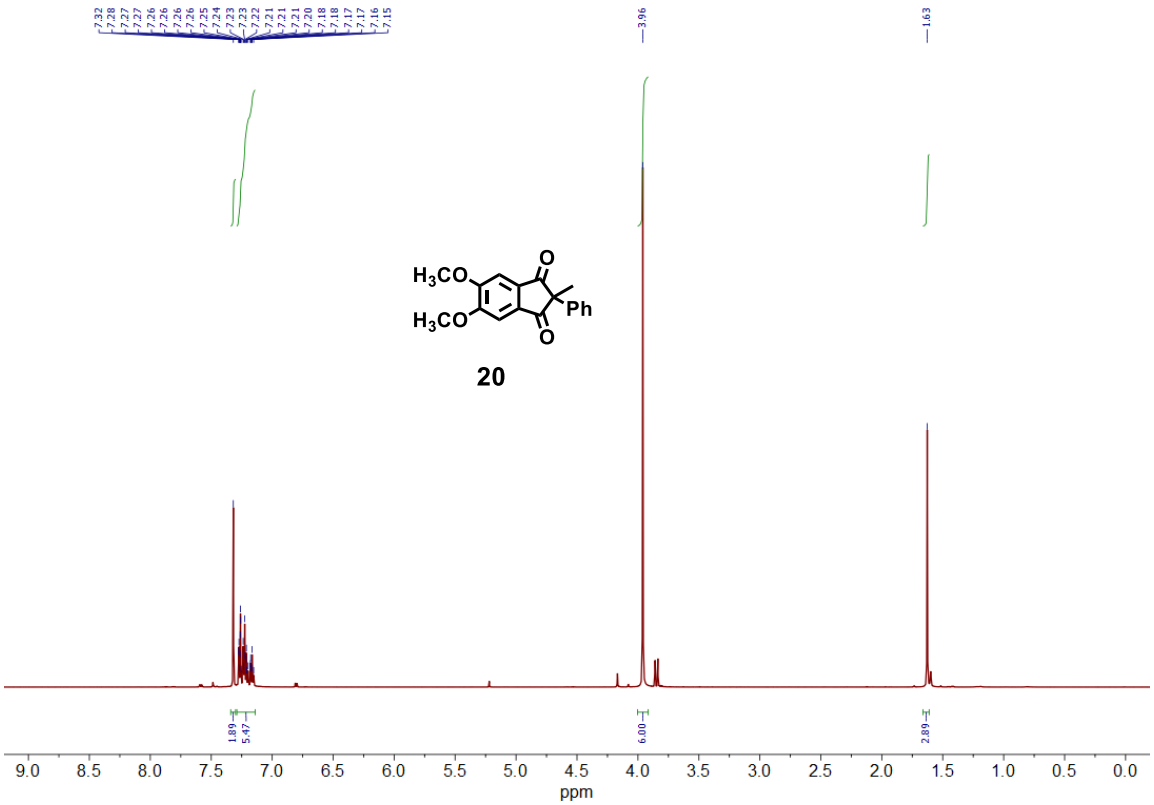


Figure S15. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 20.

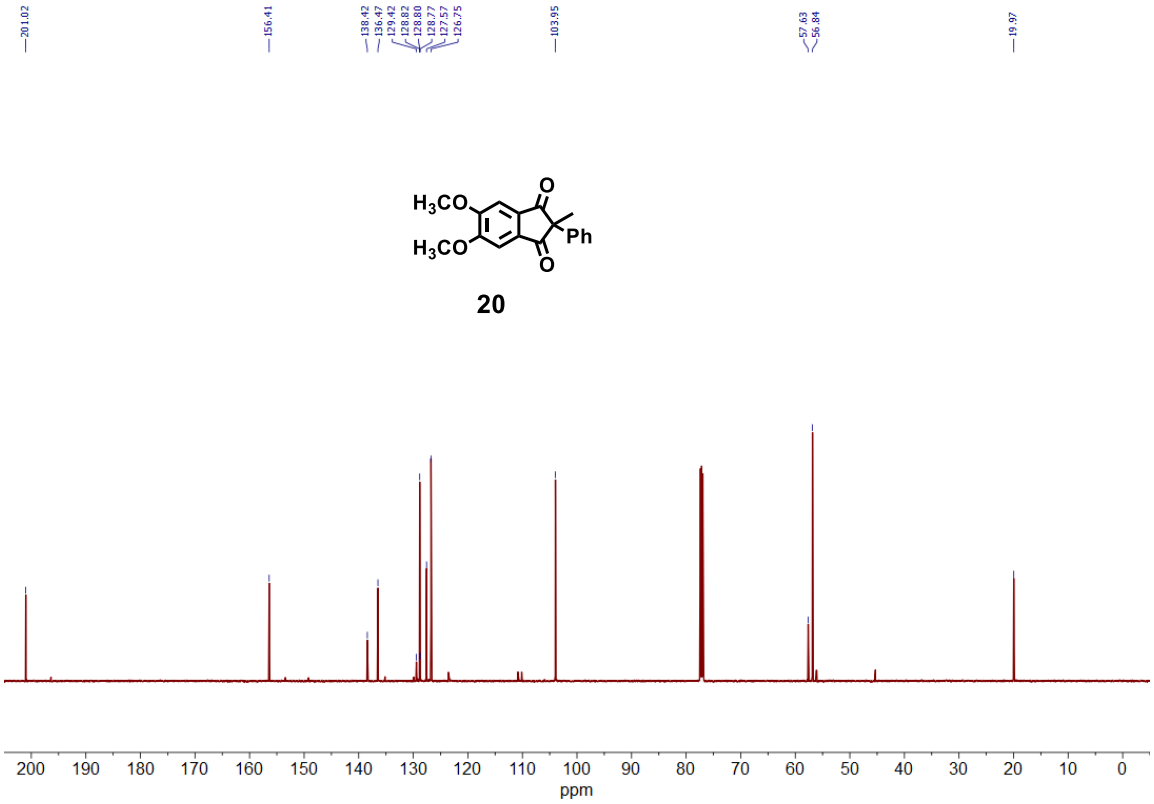


Figure S16. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 20.

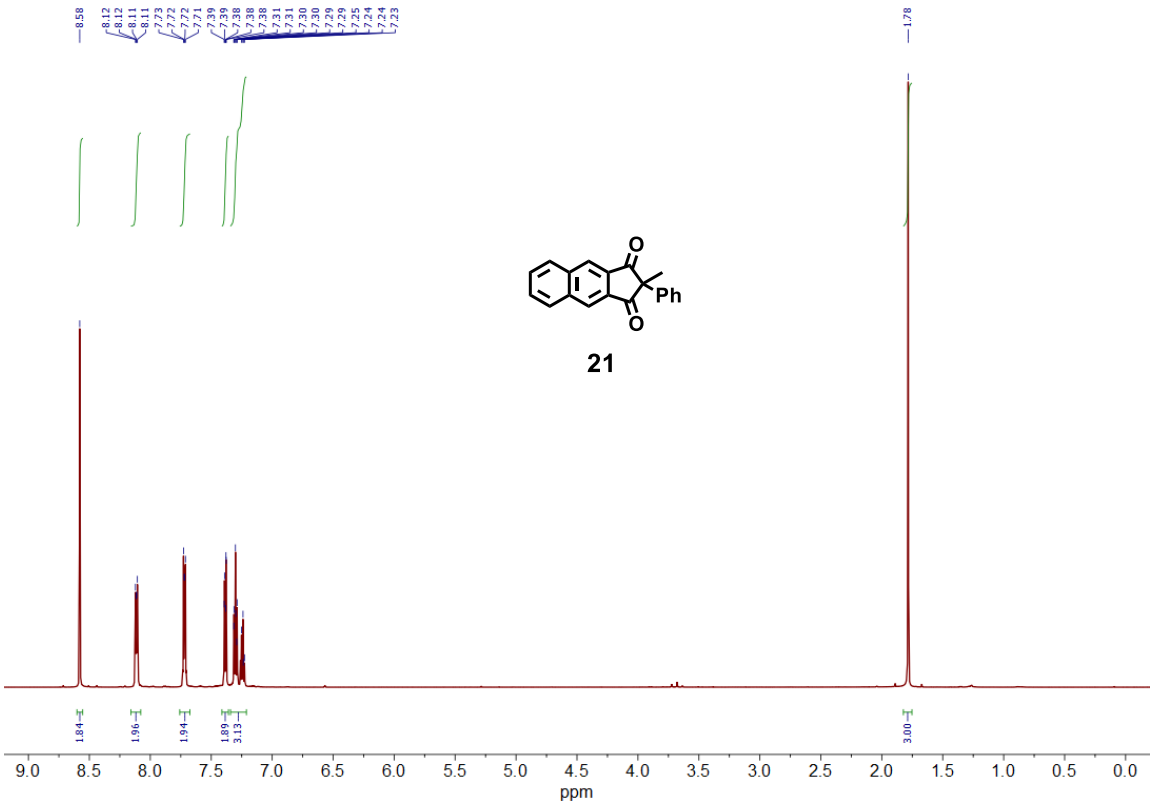


Figure S17. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 21.

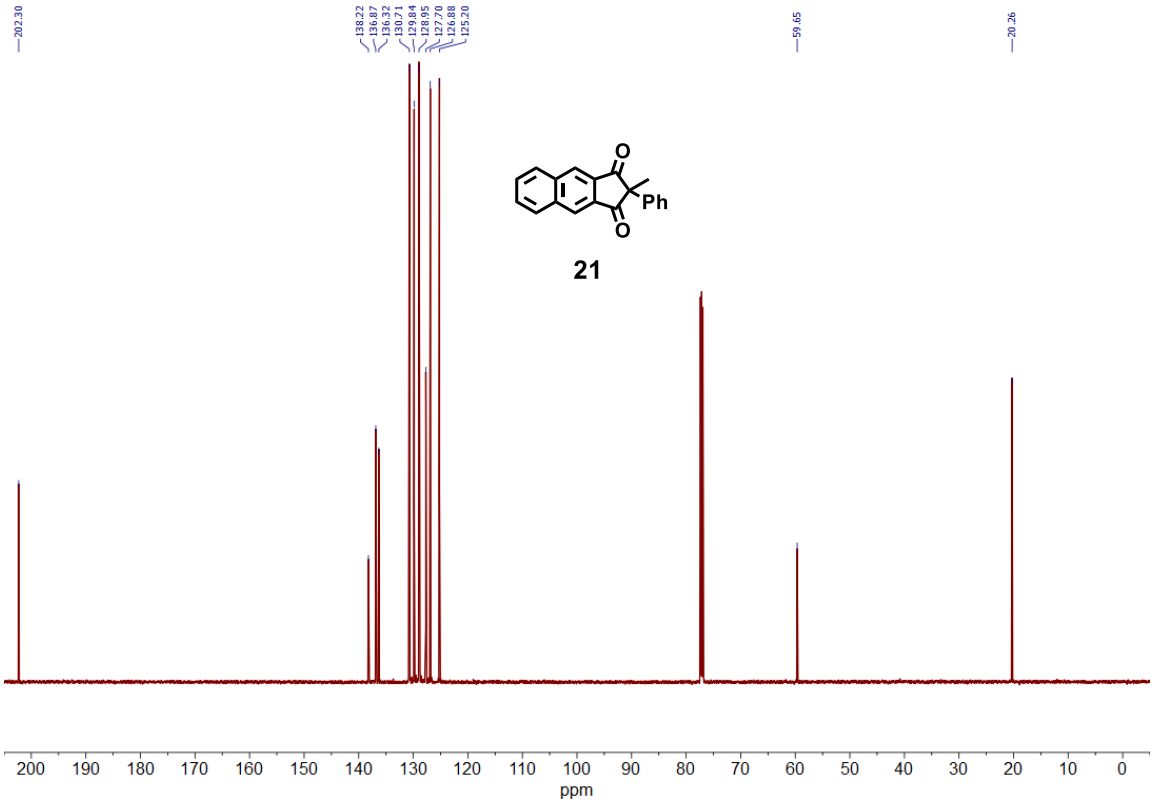


Figure S18. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 21.

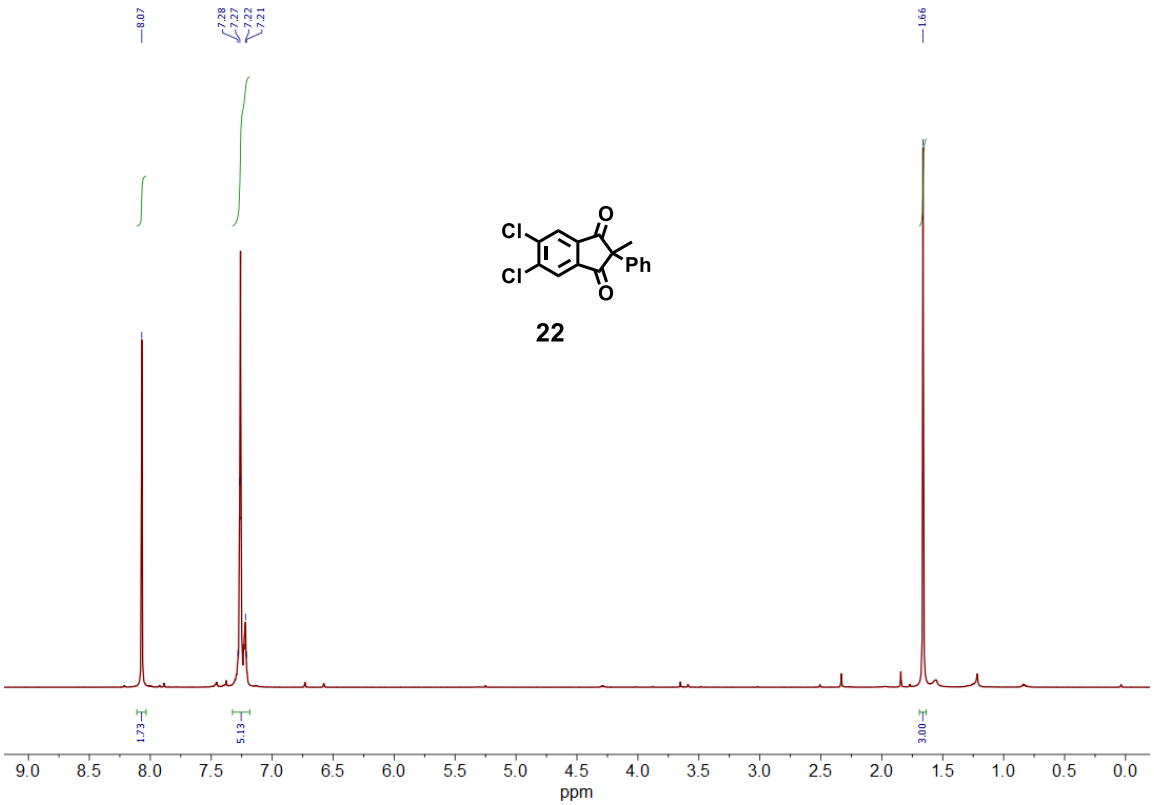


Figure S19. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 22.

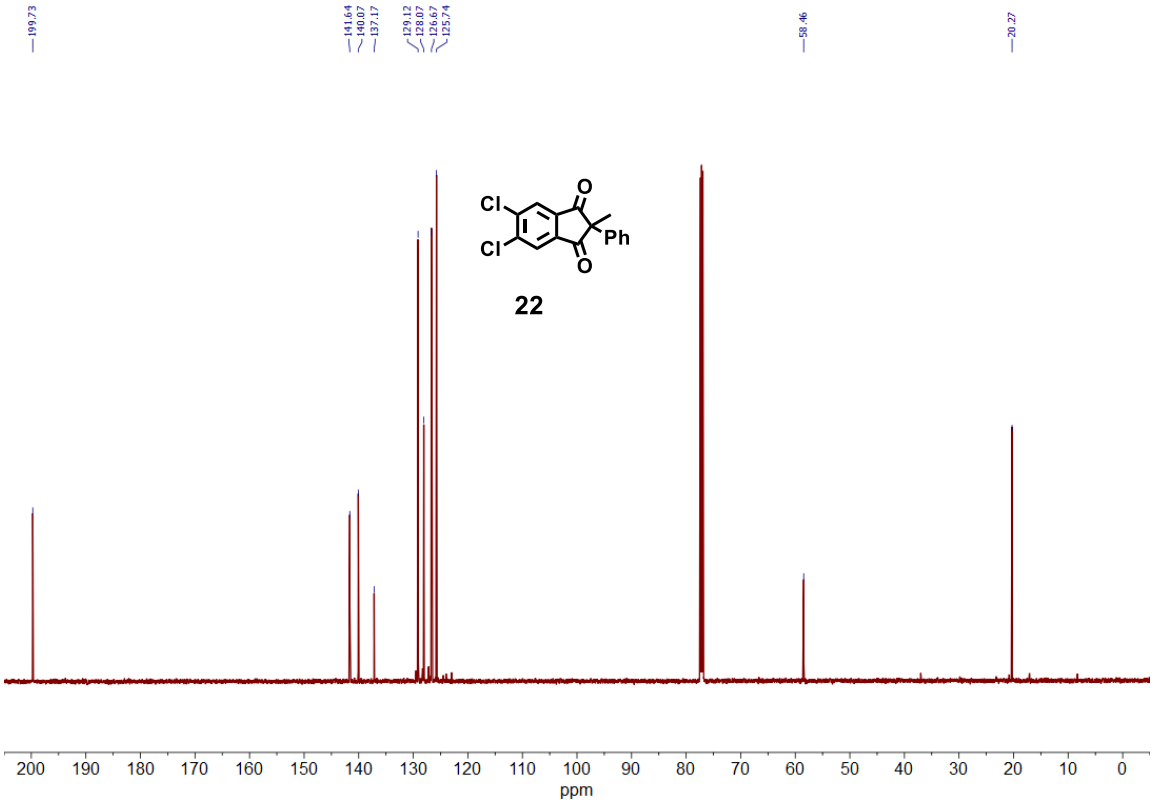


Figure S20. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 22.

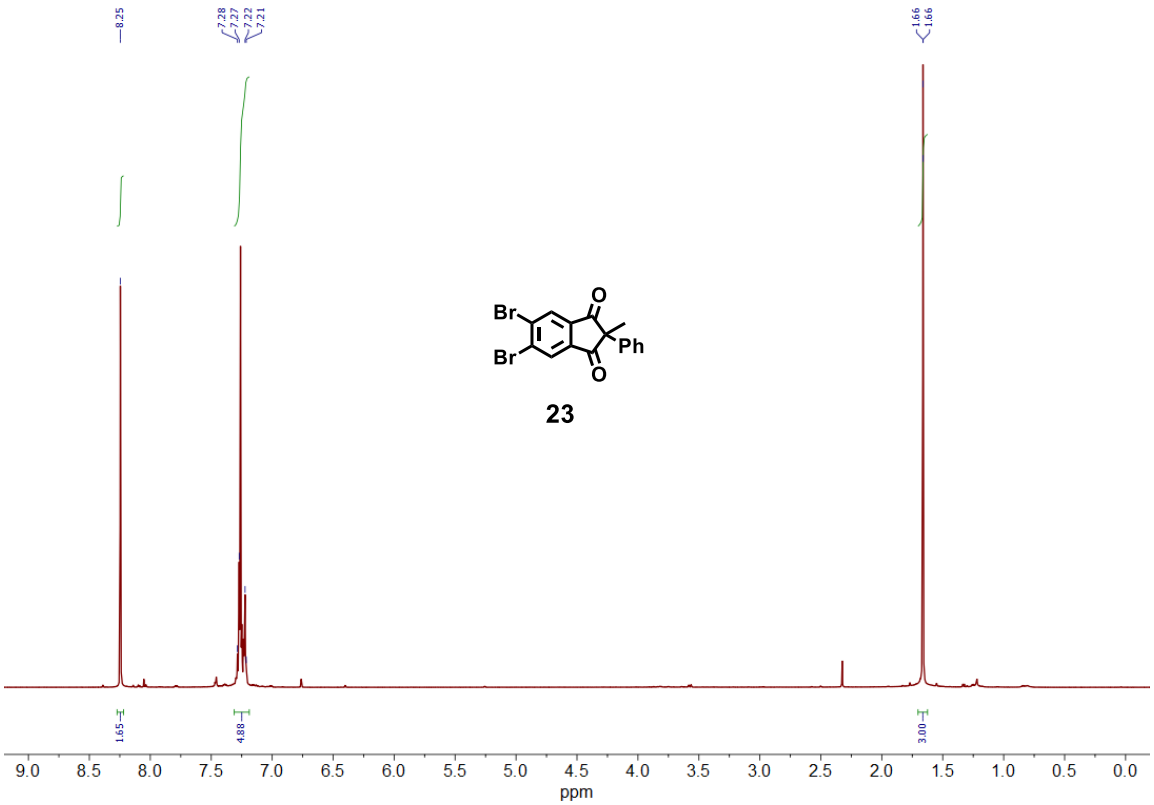


Figure S21. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 23.

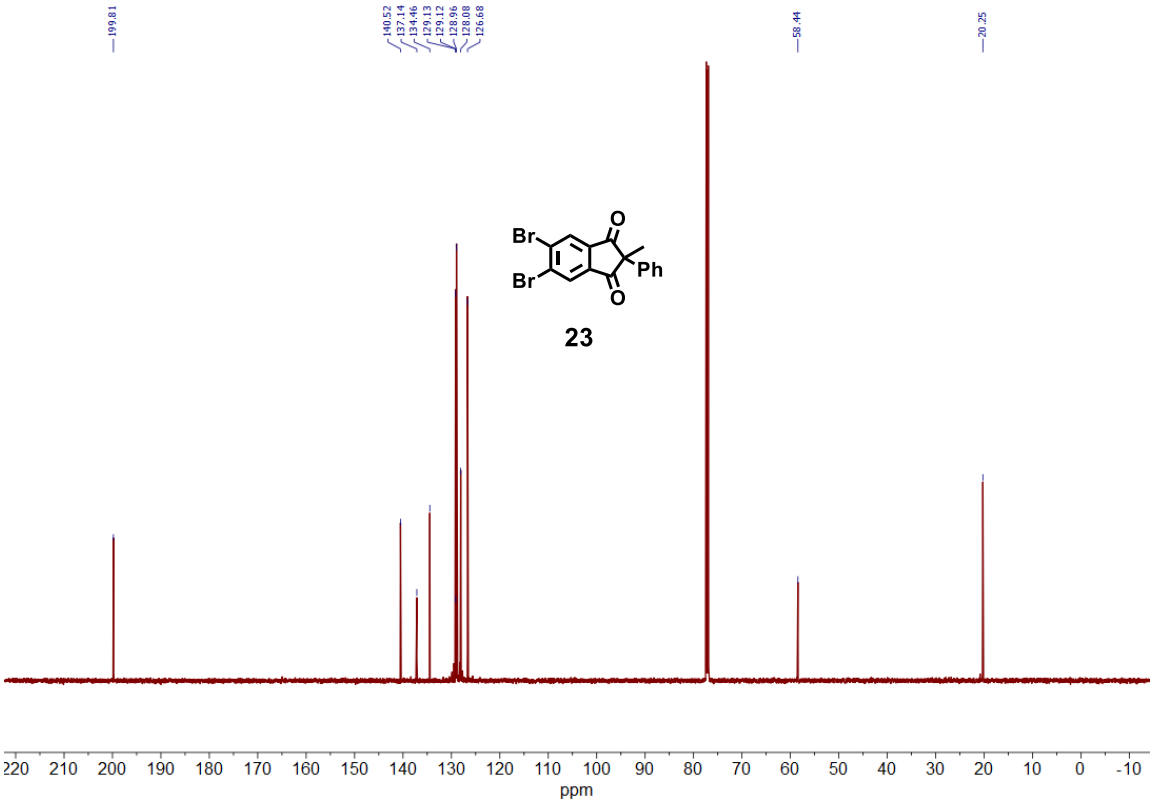


Figure S22. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 23.

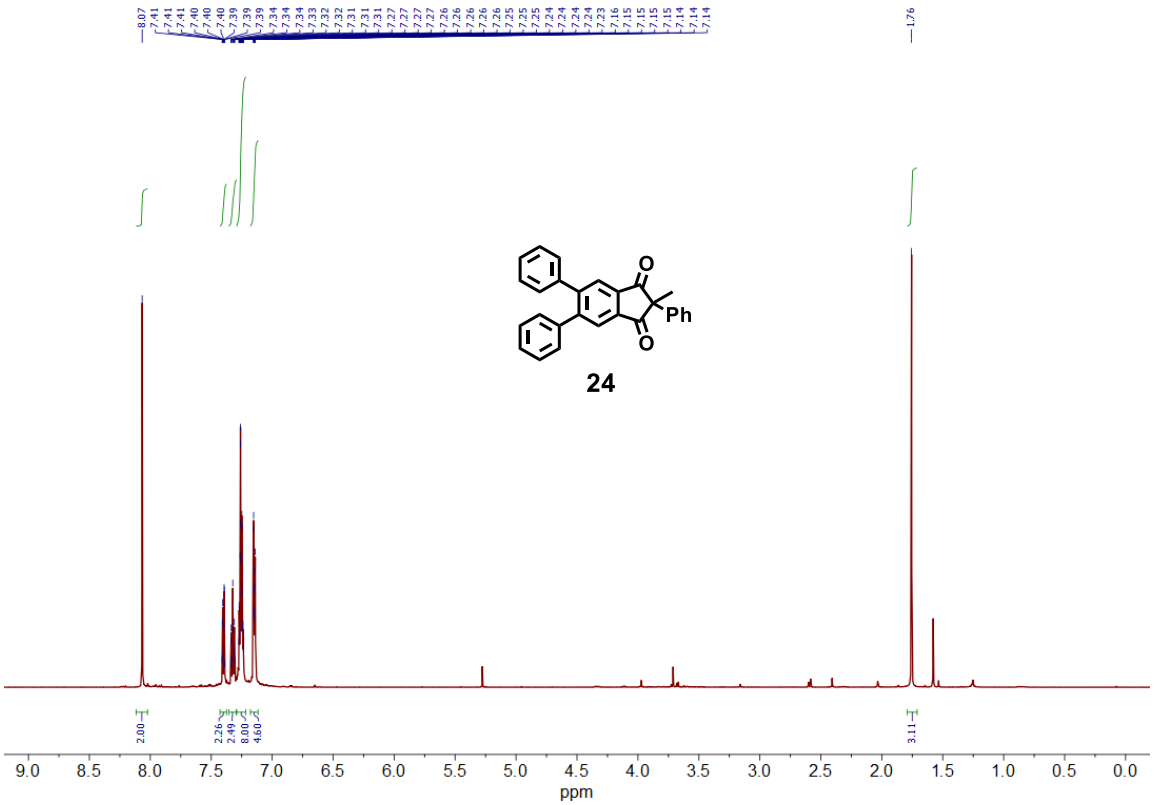


Figure S23. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 24.

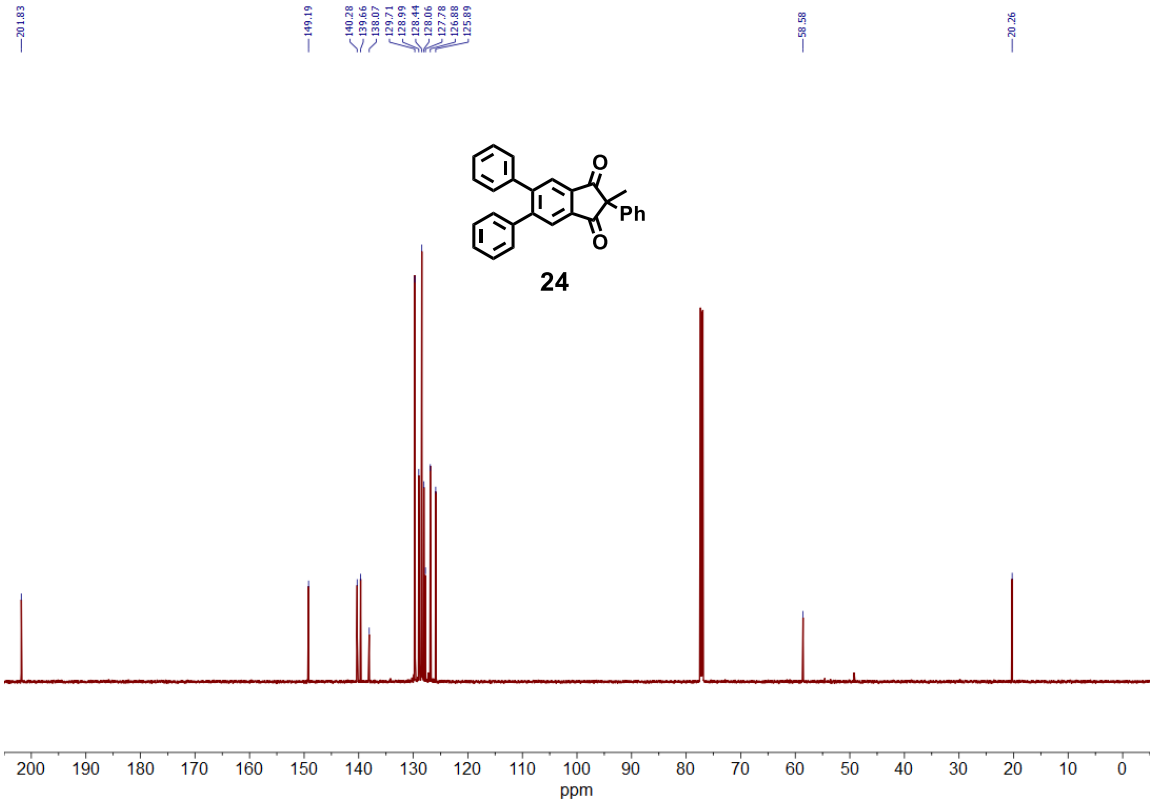


Figure S24. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 24.

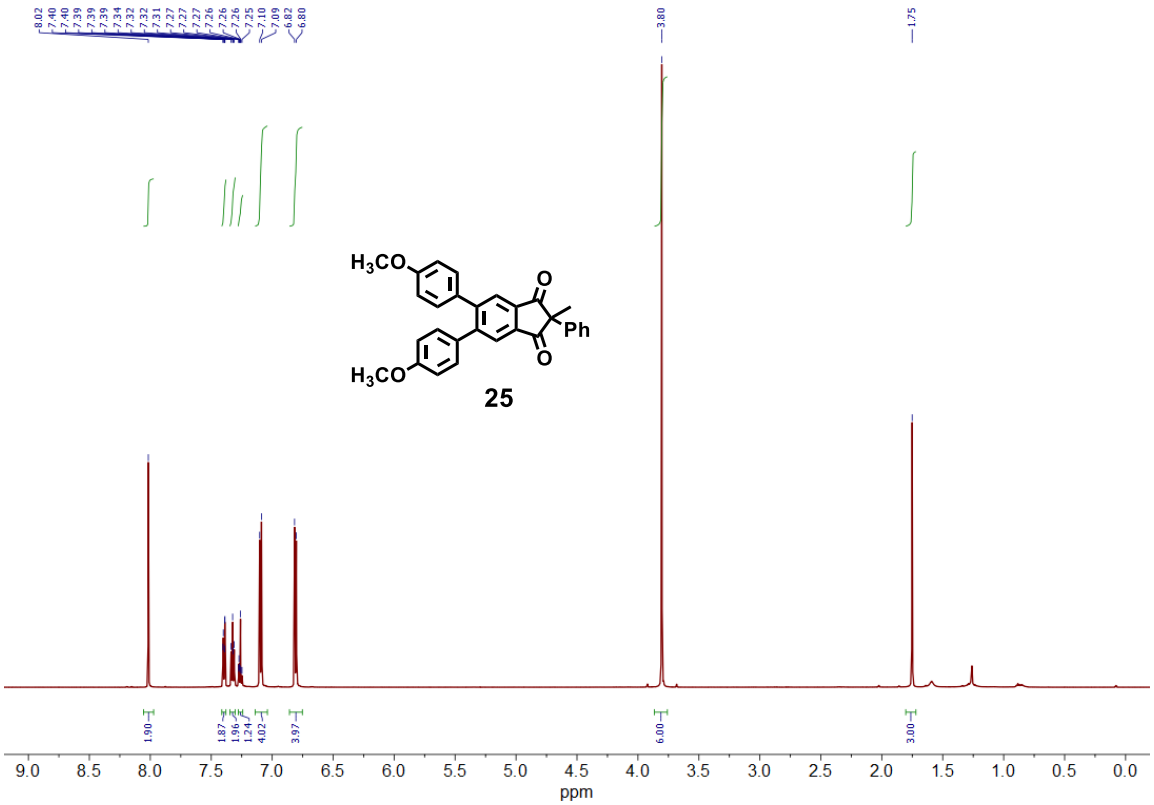


Figure S25. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 25.

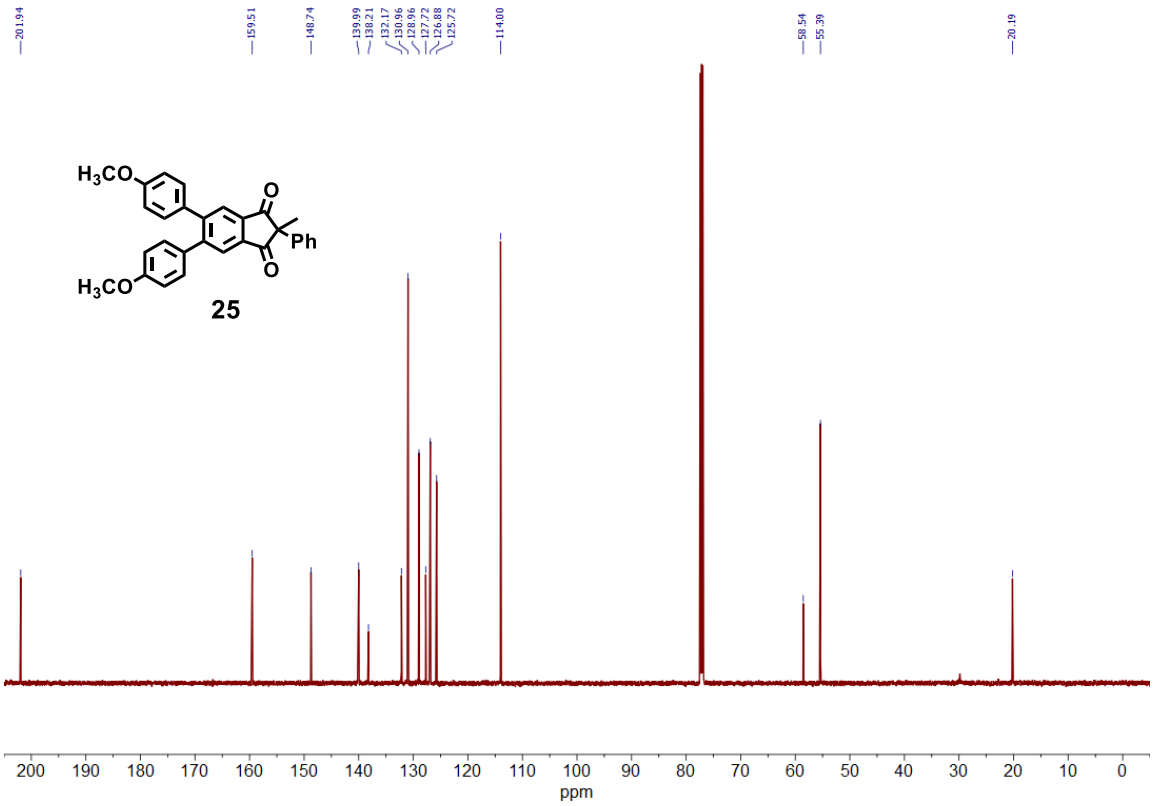


Figure S26. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 25.

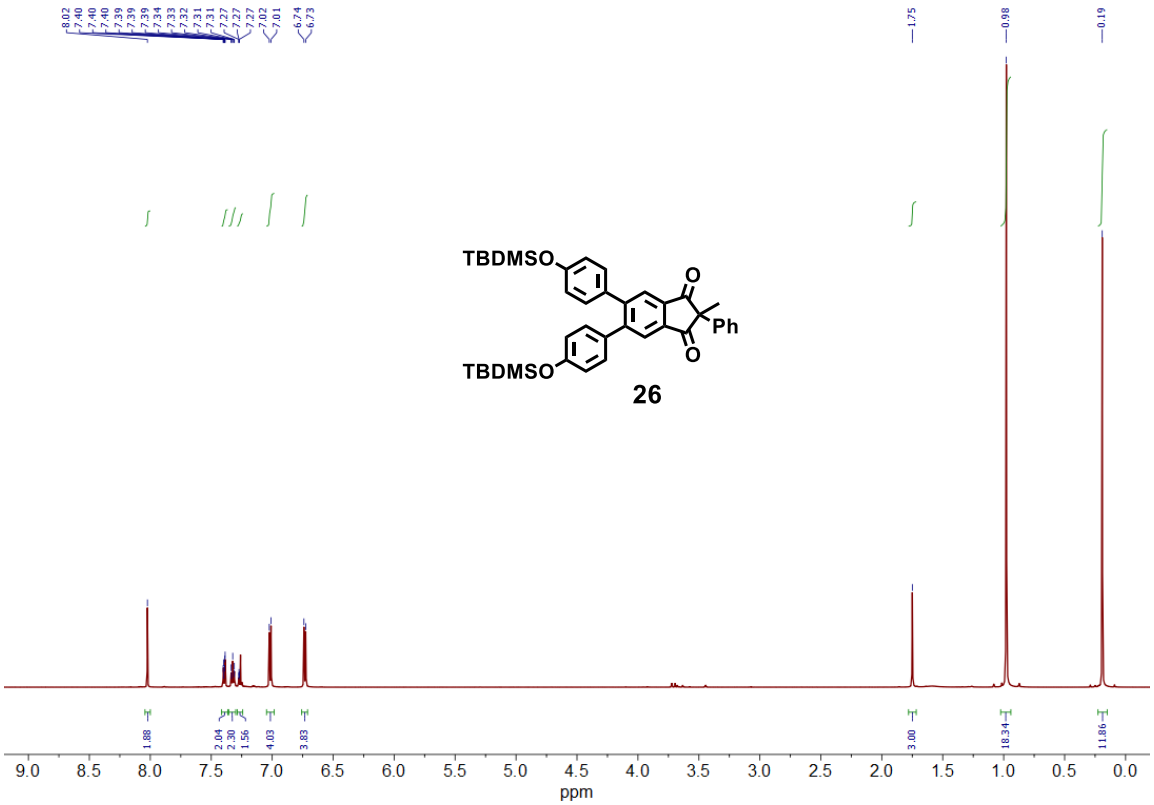


Figure S27. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 26.

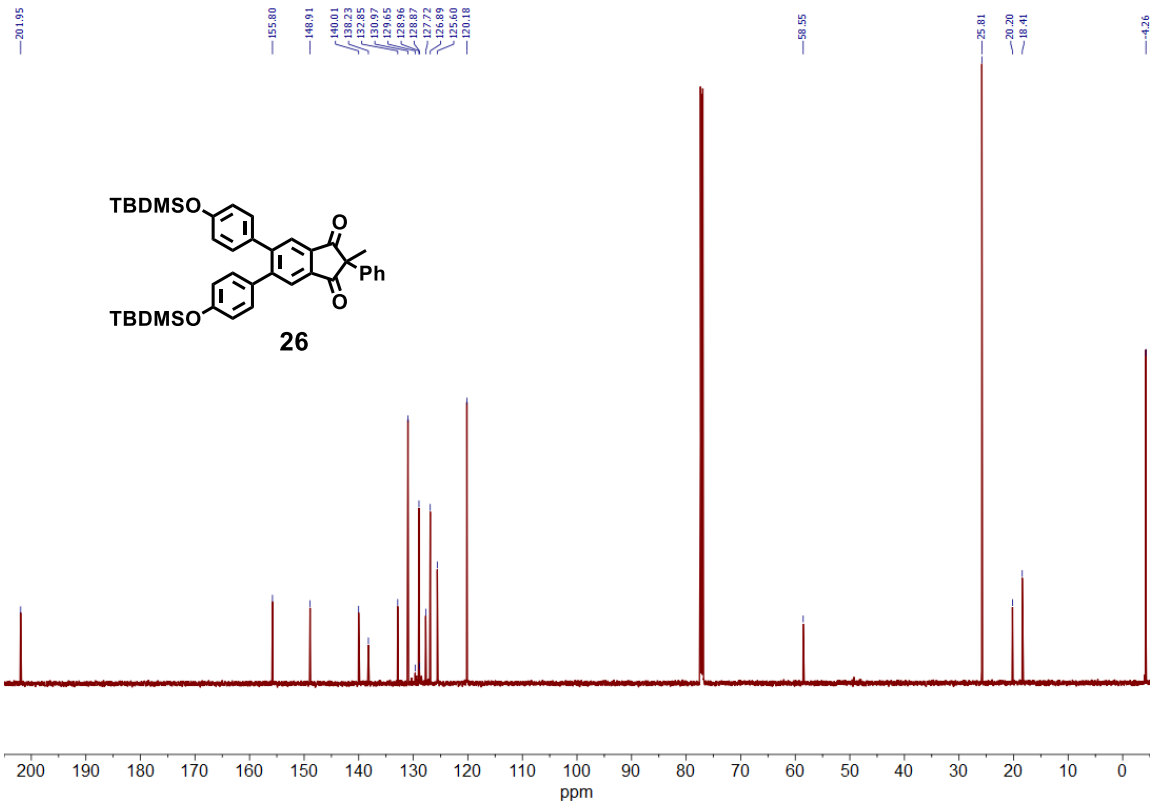


Figure S28. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 26.

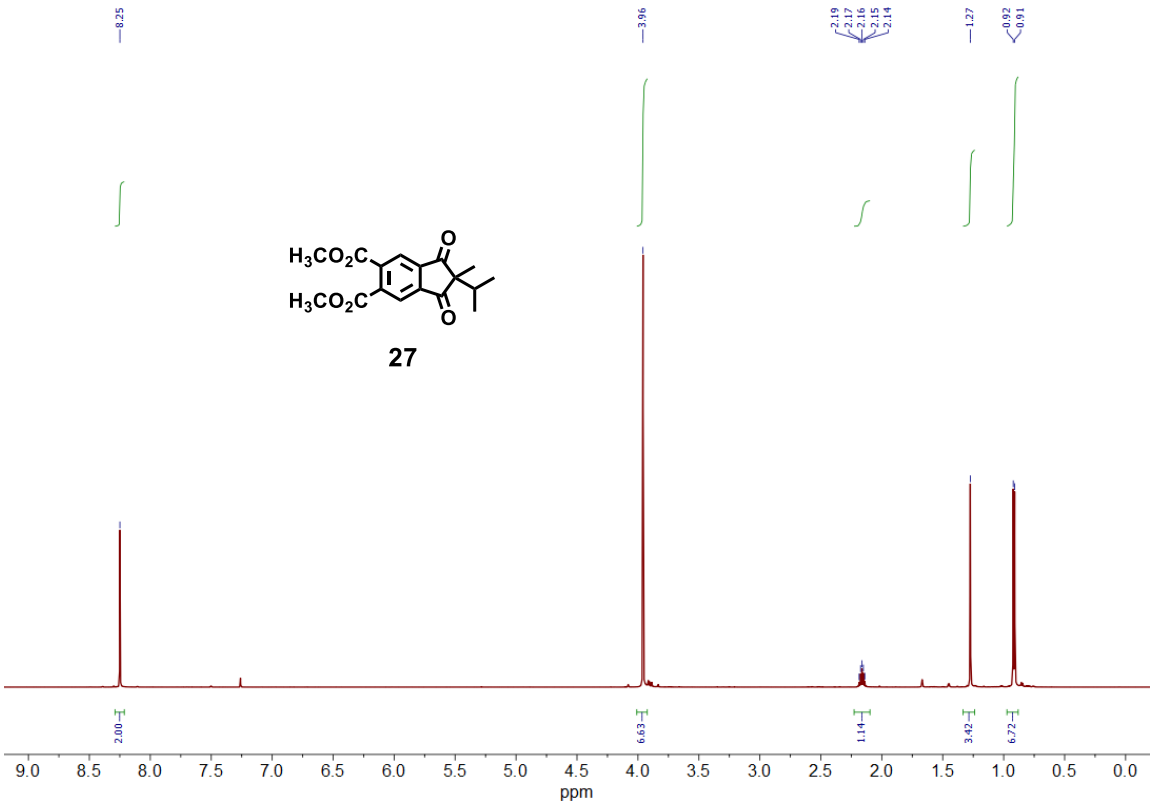


Figure S29. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 27.

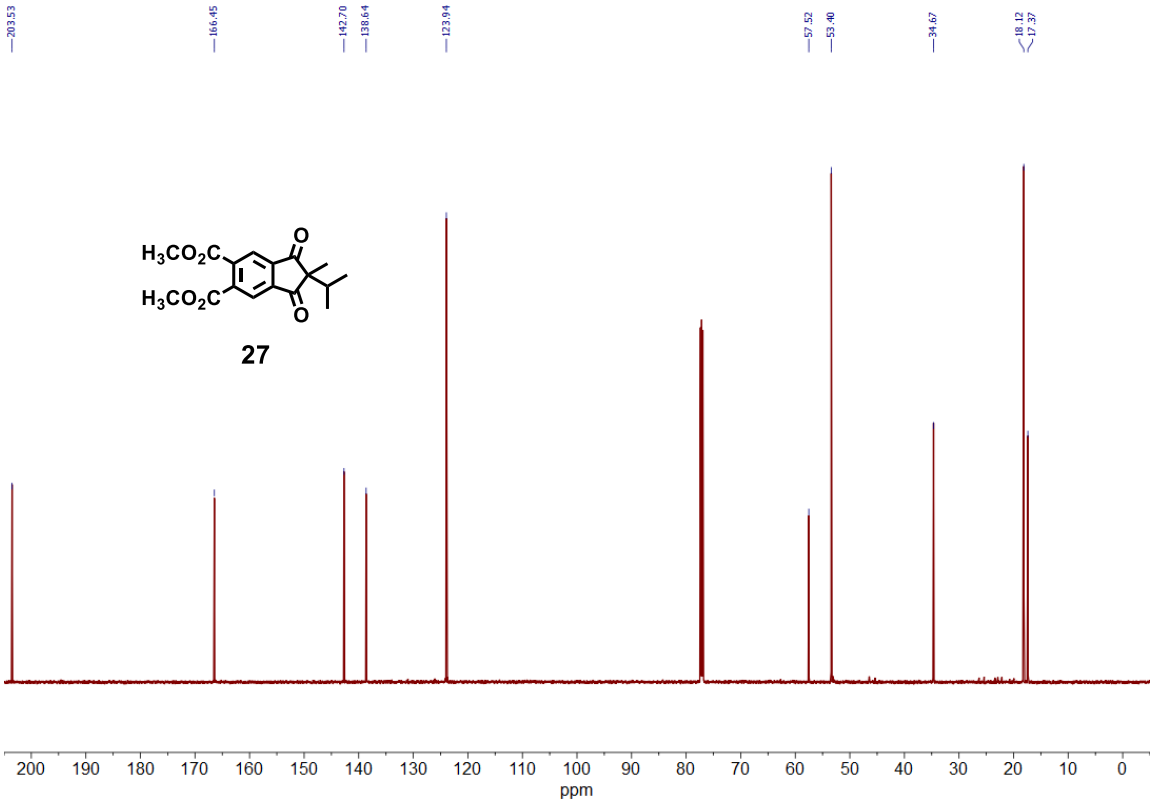


Figure S30. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 27.

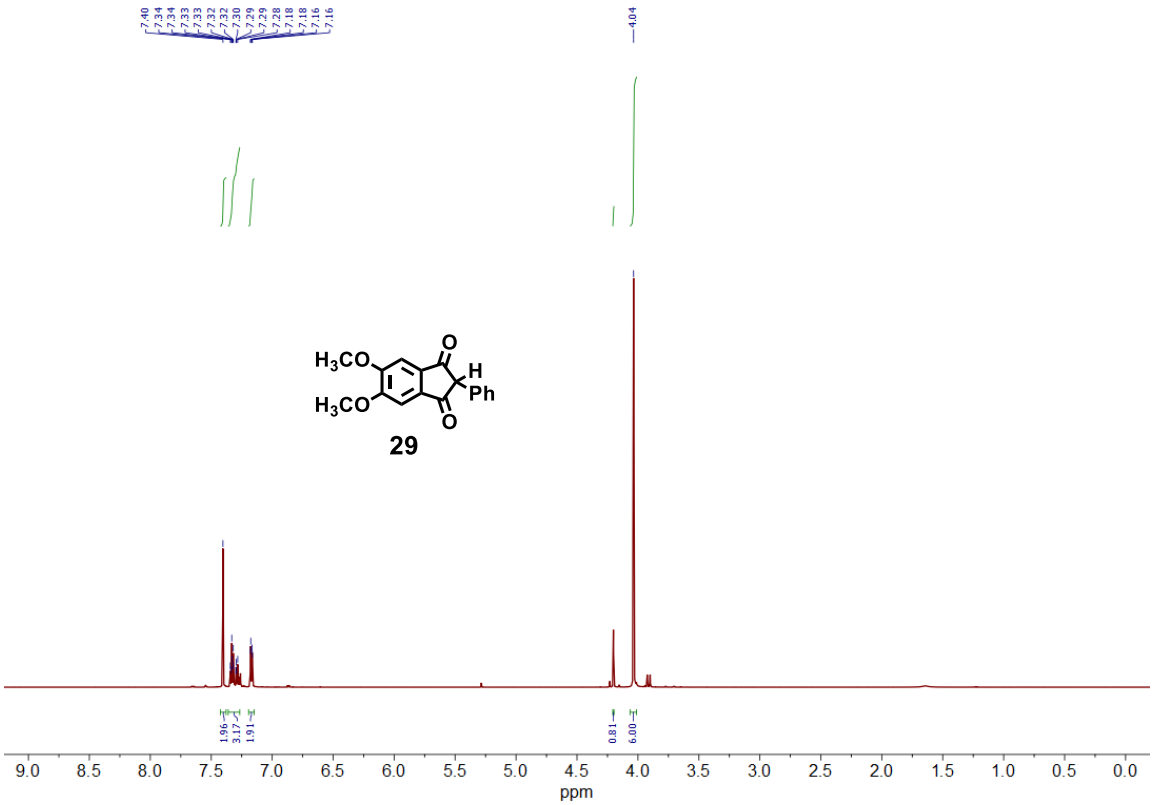


Figure S31. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 29.

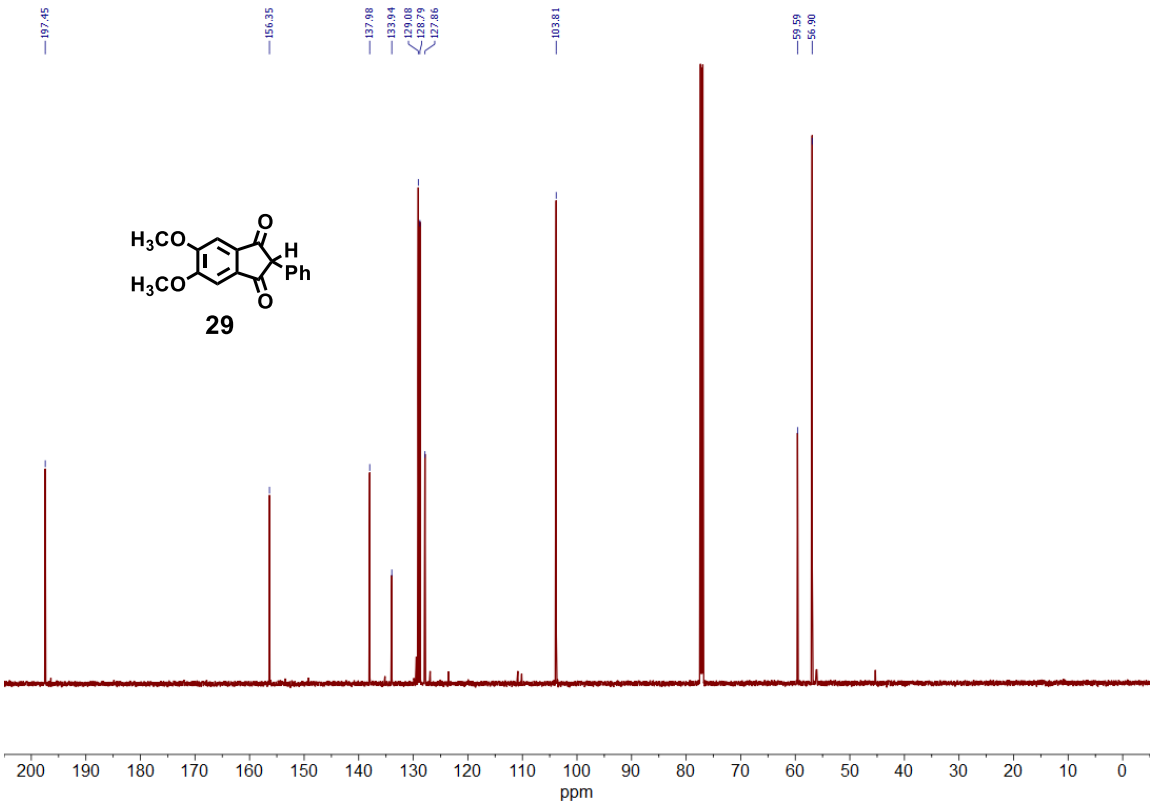


Figure S32. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 29.

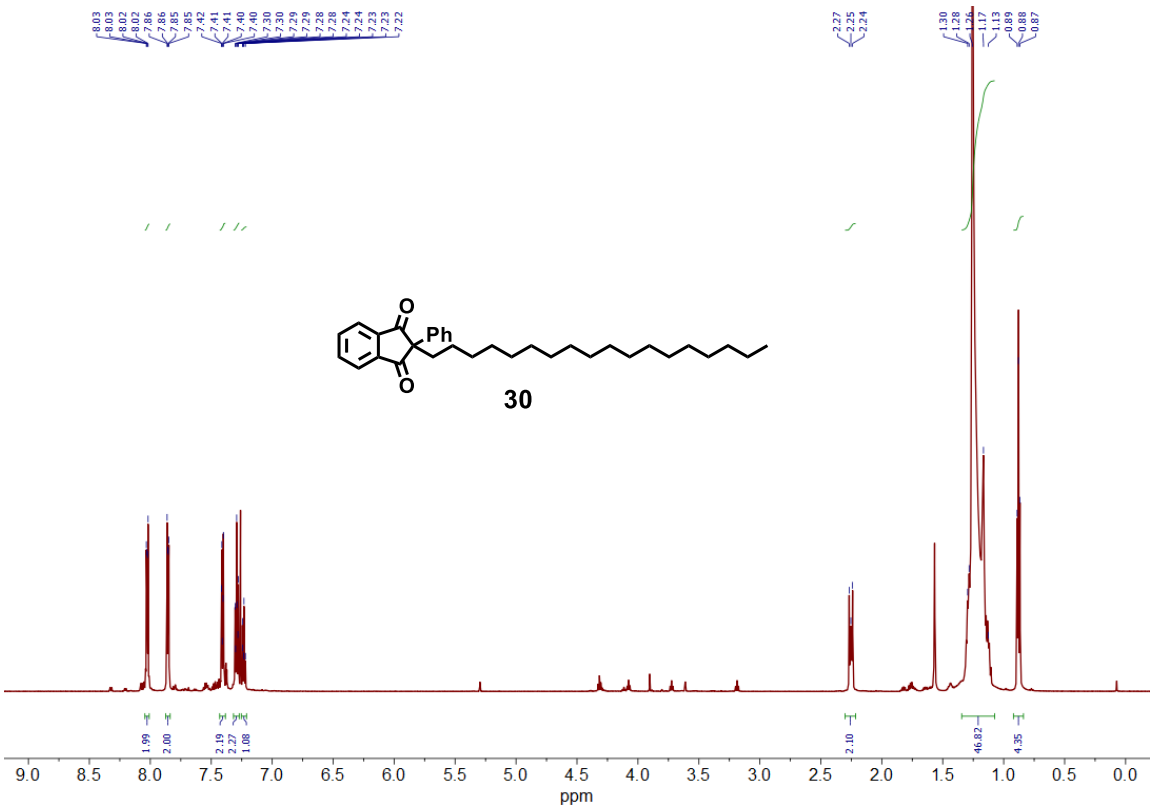


Figure S33. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 30.

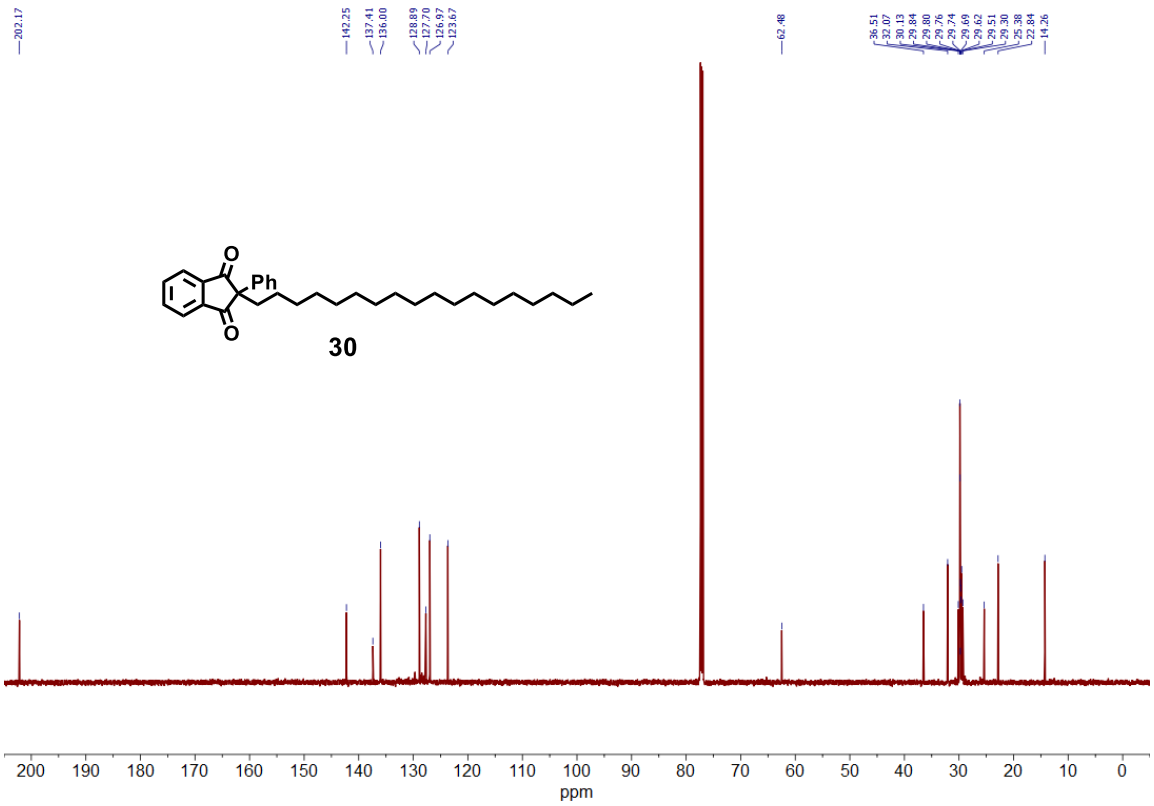


Figure S34. ^{13}C NMR spectrum (150 MHz, $CDCl_3$, 25 °C) of 30.

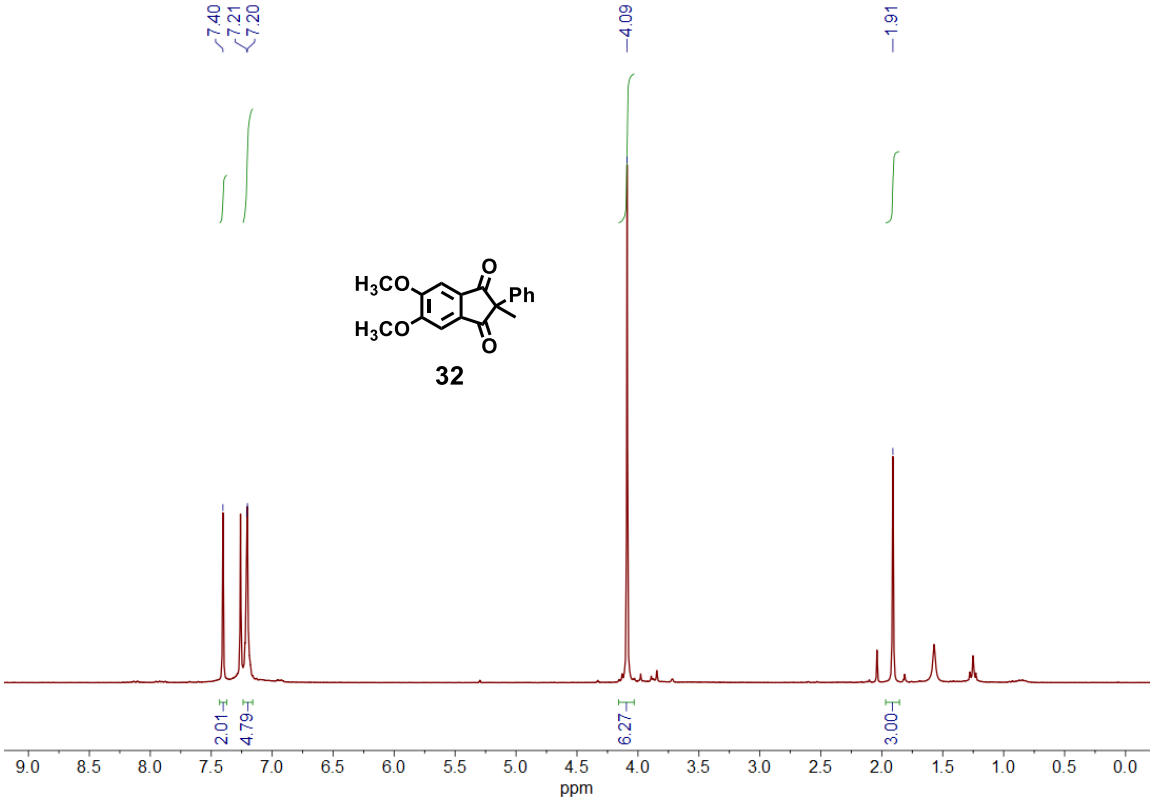


Figure S35. ^{1}H NMR spectrum (300 MHz, $CDCl_3$, 25 °C) of 32.

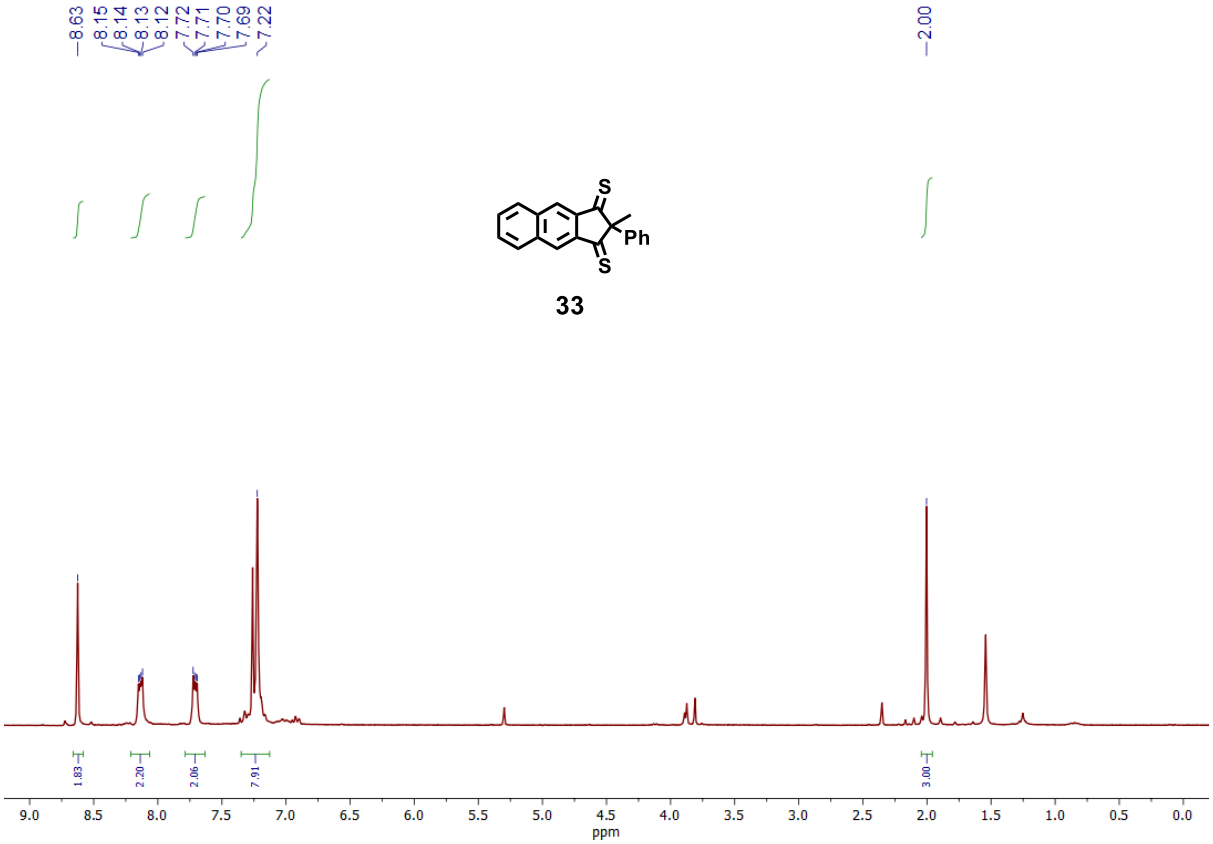


Figure S36. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 33.

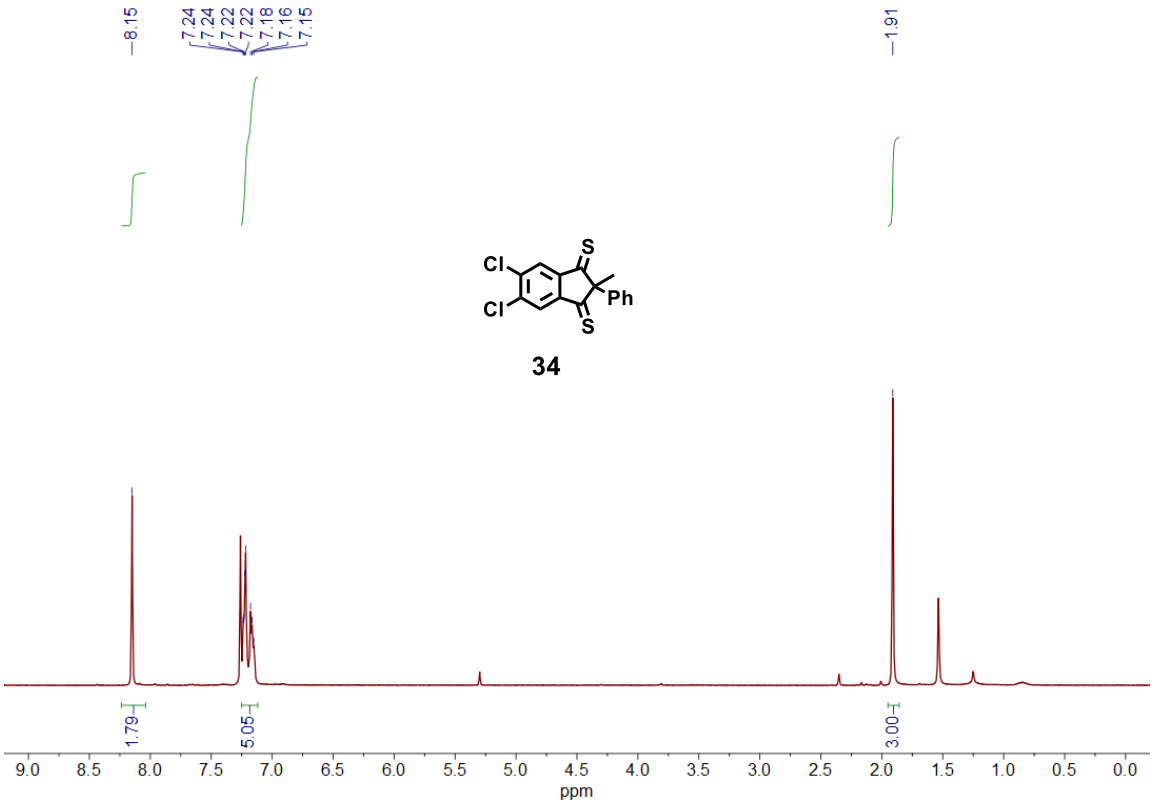


Figure S37. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 34.

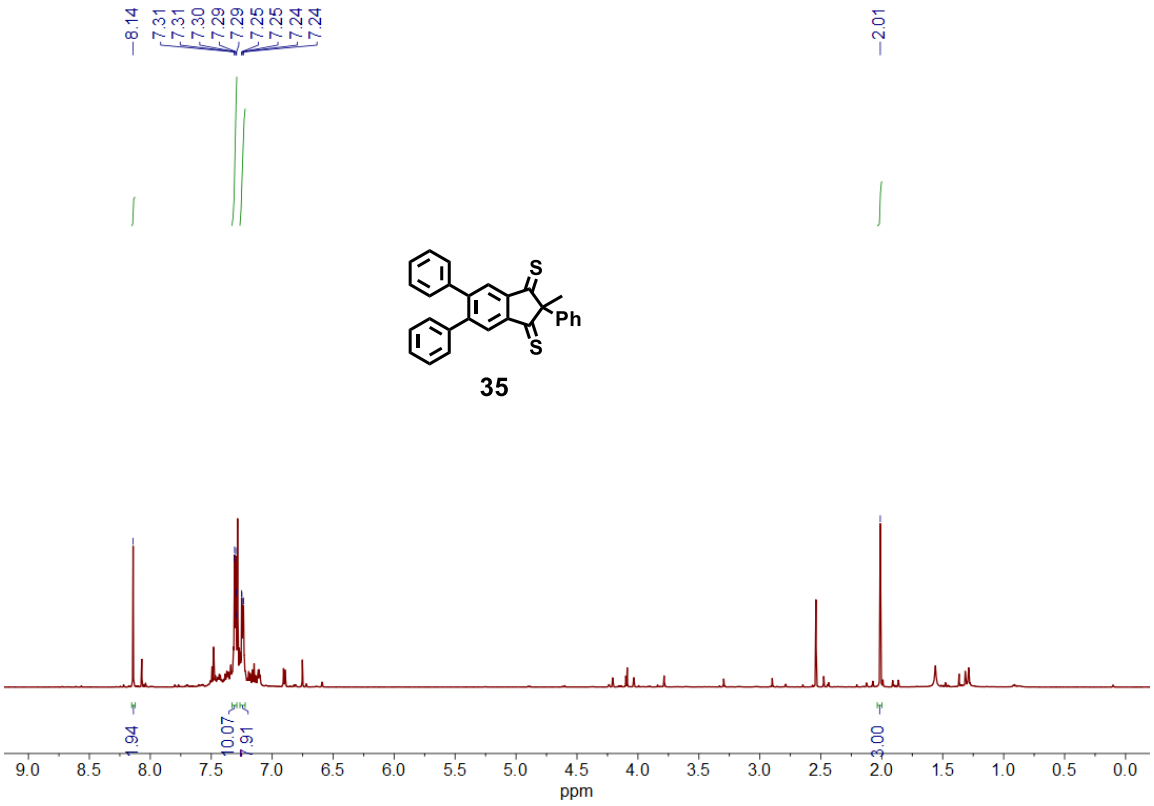


Figure S38. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 35 (impure compound).

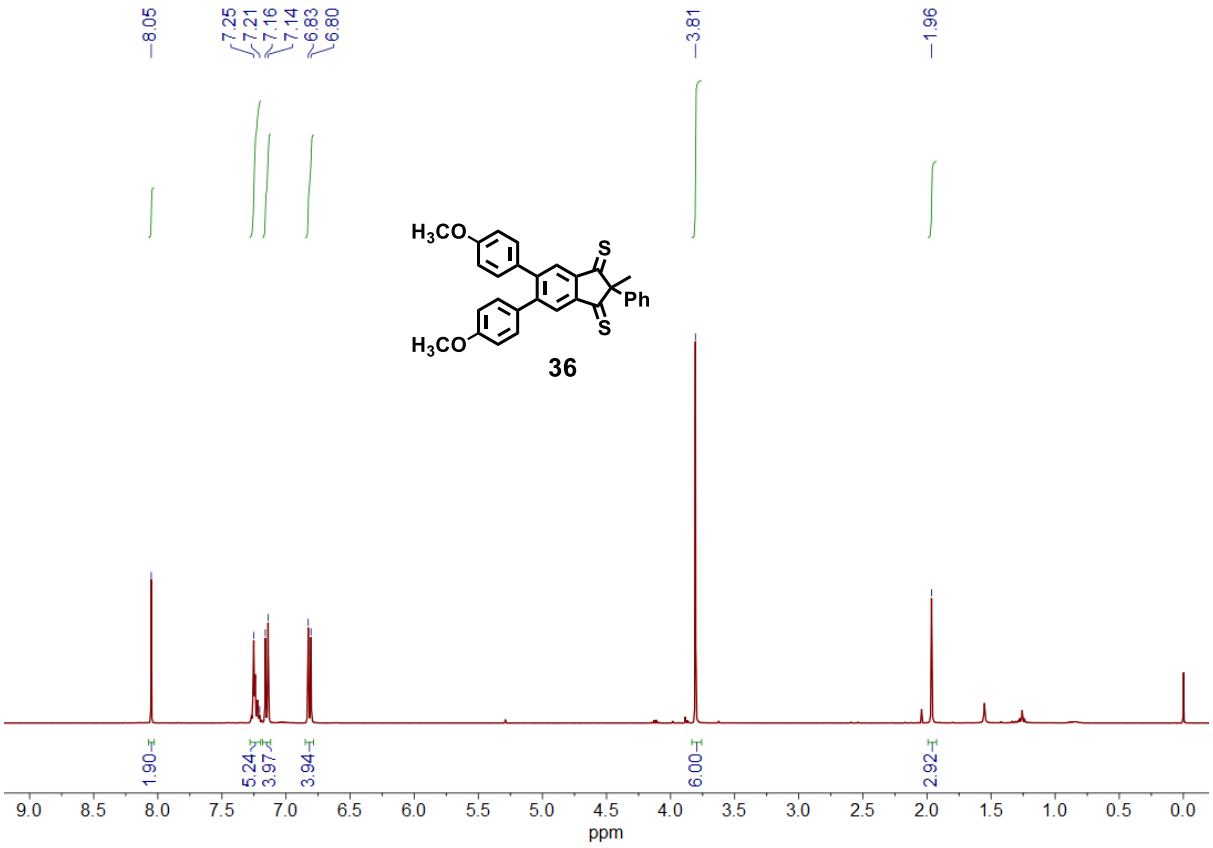


Figure S39. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 36.

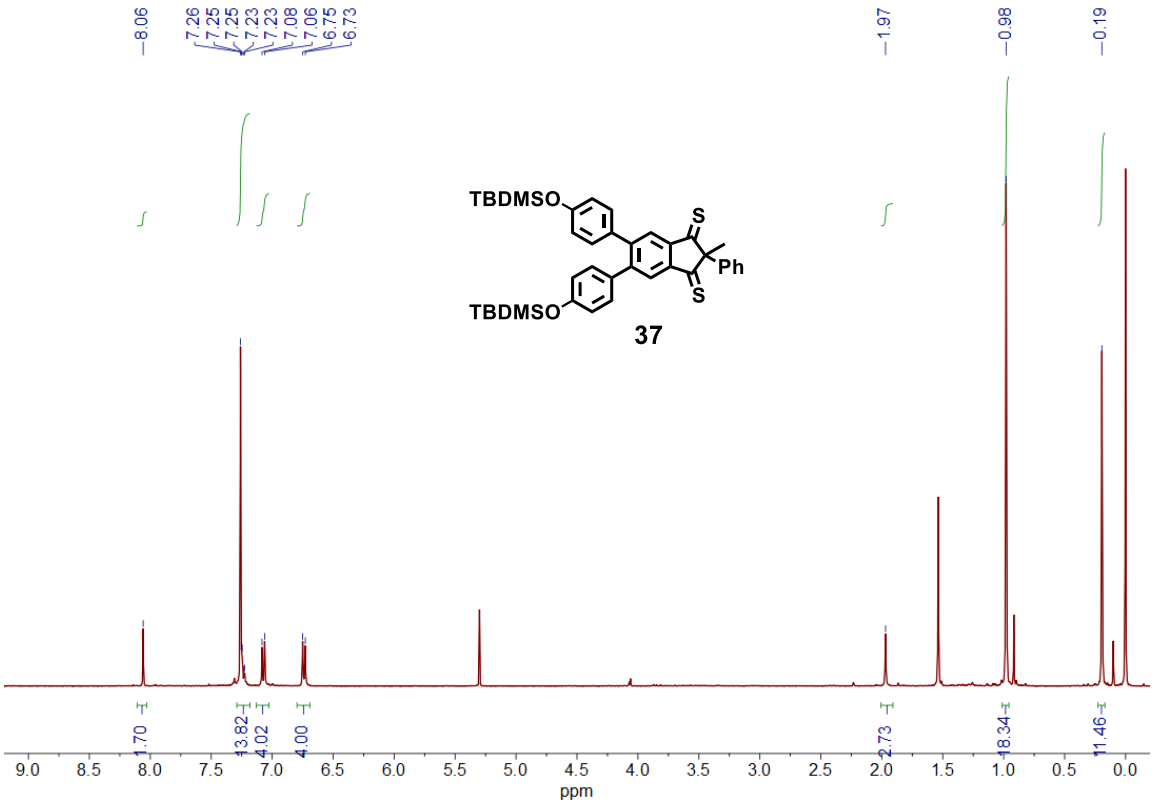


Figure S40. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 37.

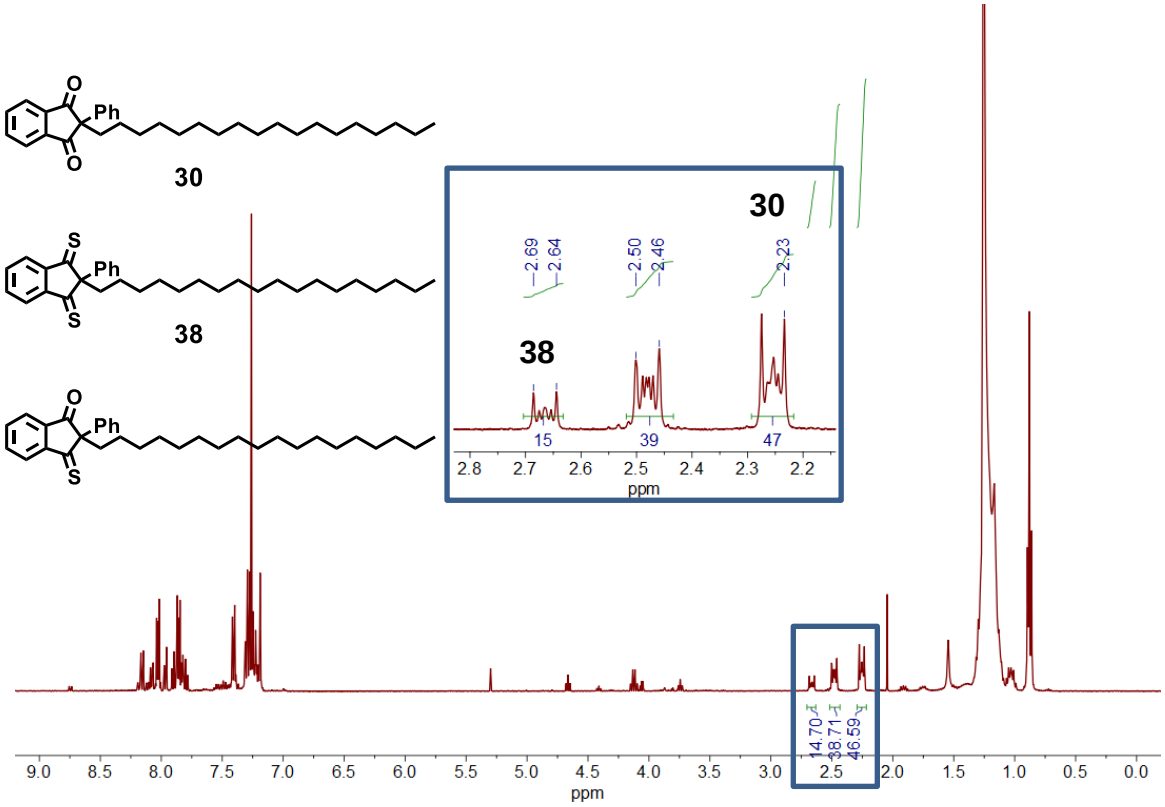


Figure S41. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of the 15:39:47 ternary mixture of 38, 30 and singly converted product, respectively.

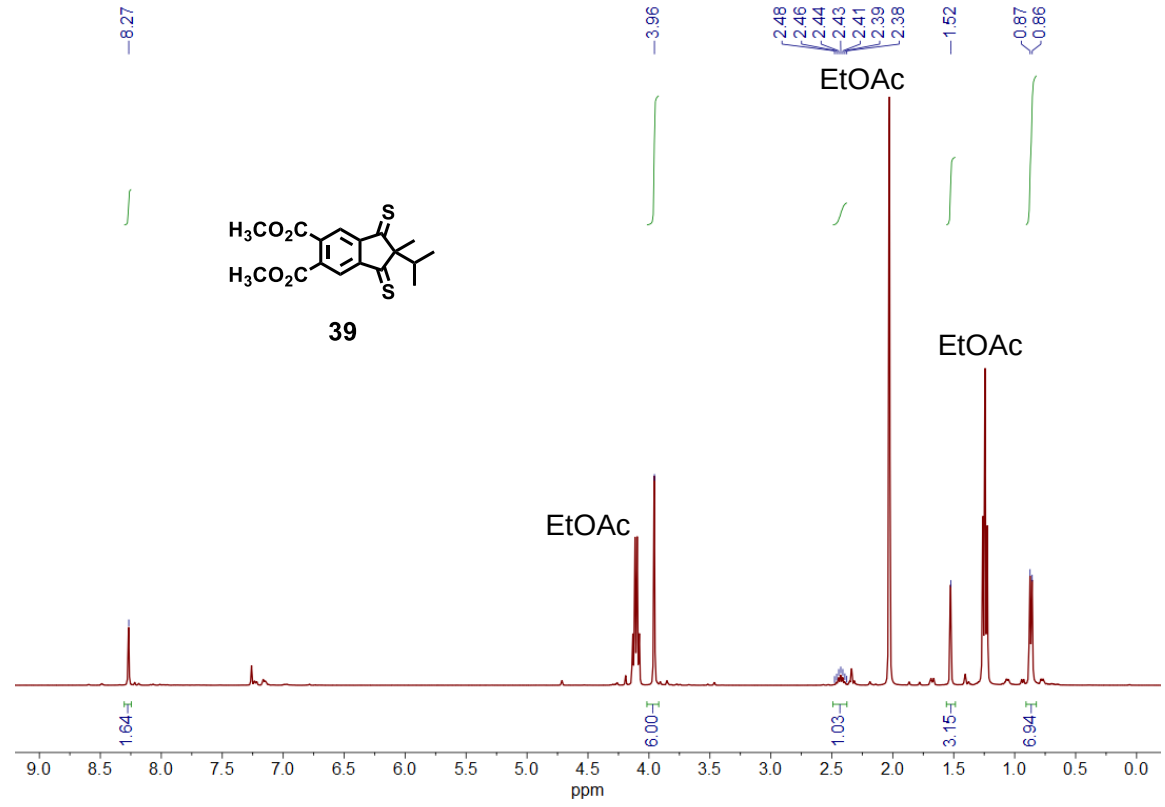


Figure S42. ^{1}H NMR spectrum (400 MHz, $CDCl_3$, 25 °C) of 39.

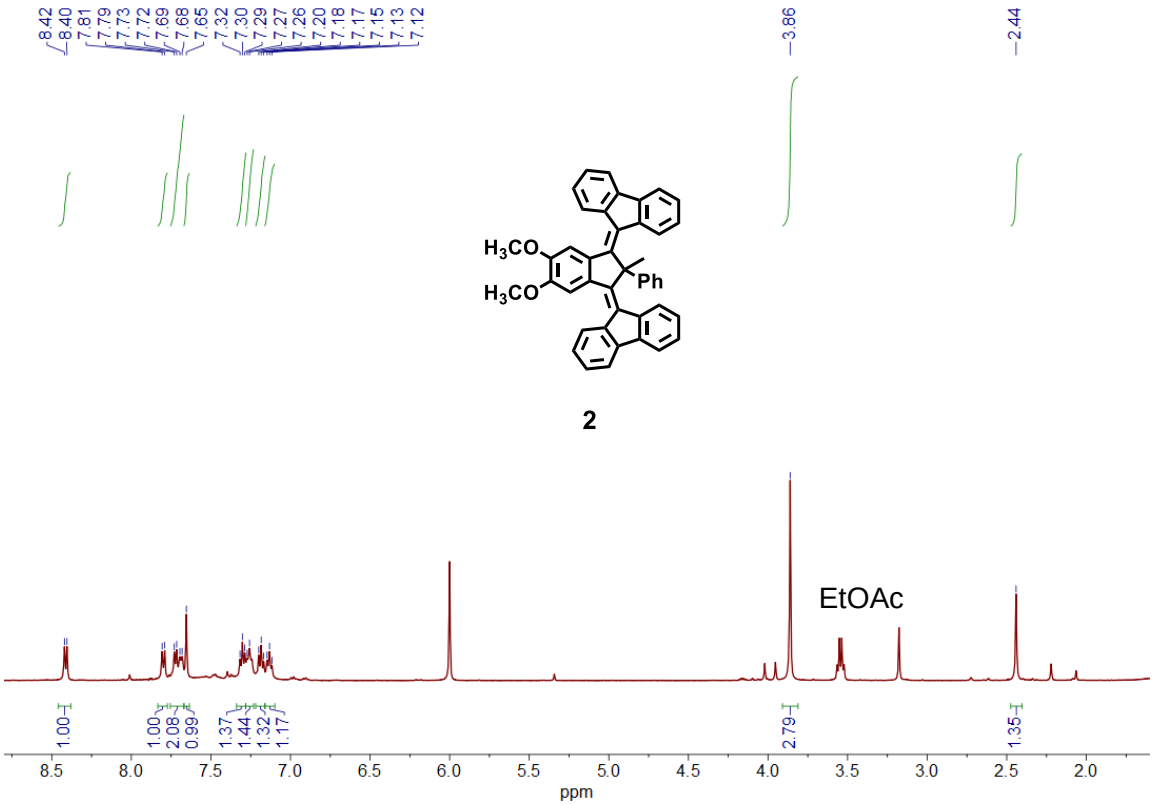


Figure S43. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 2.

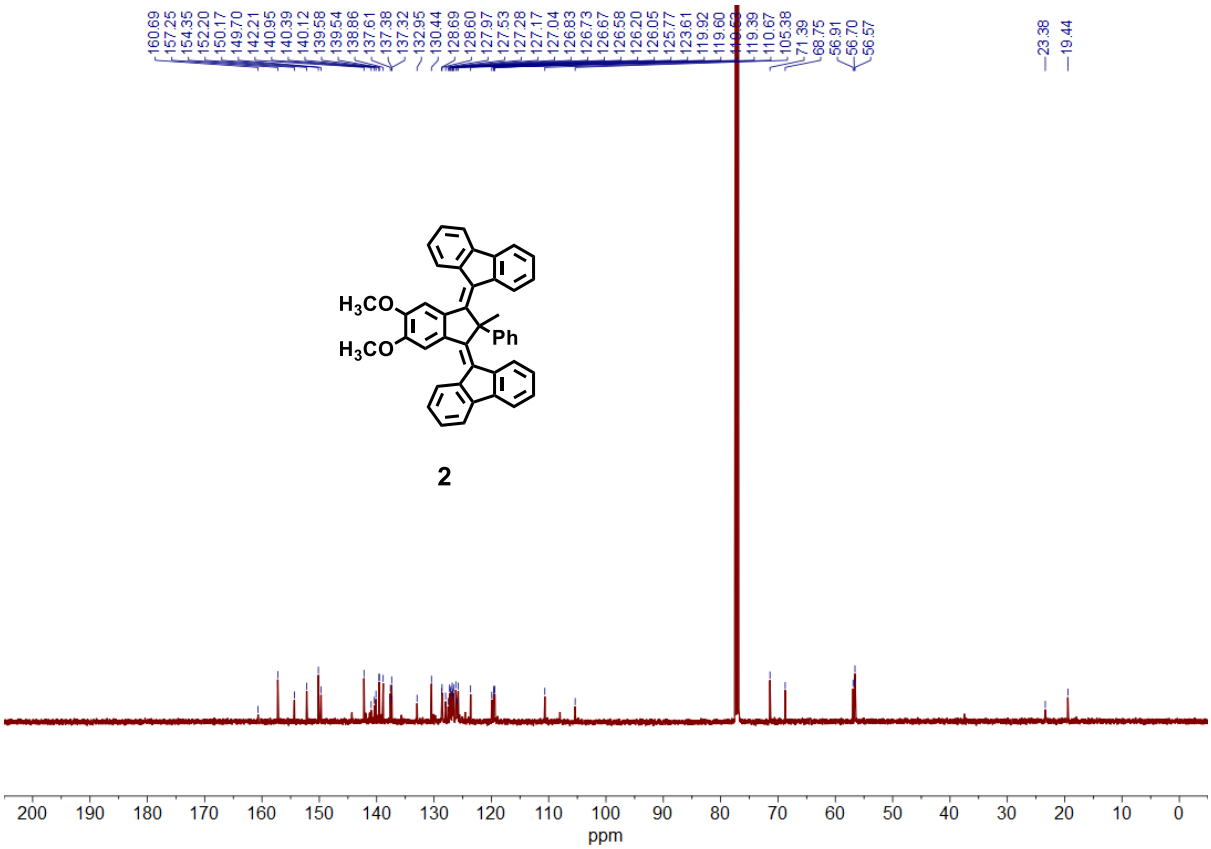


Figure S44. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 2.

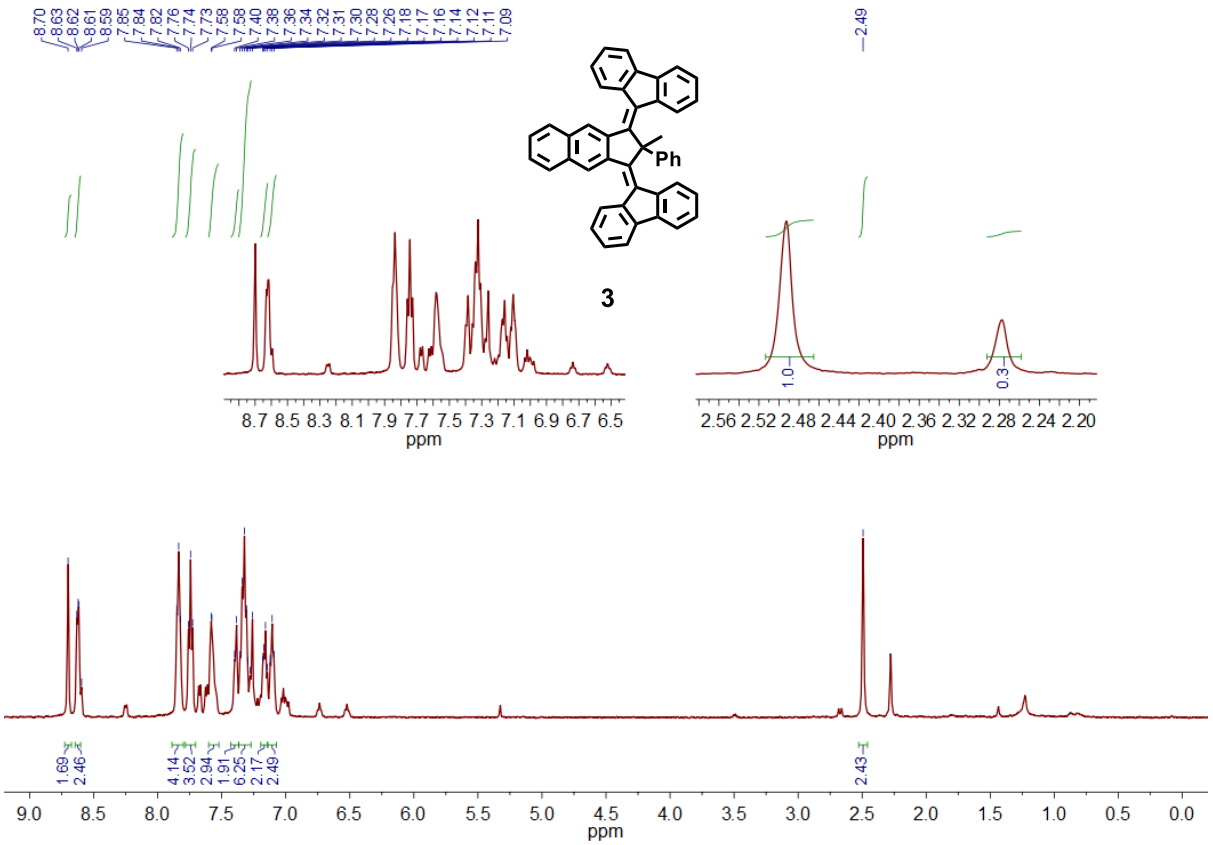


Figure S45. ^{1}H NMR spectrum (500 MHz, $CDCl_3$, -45 °C) of 3.

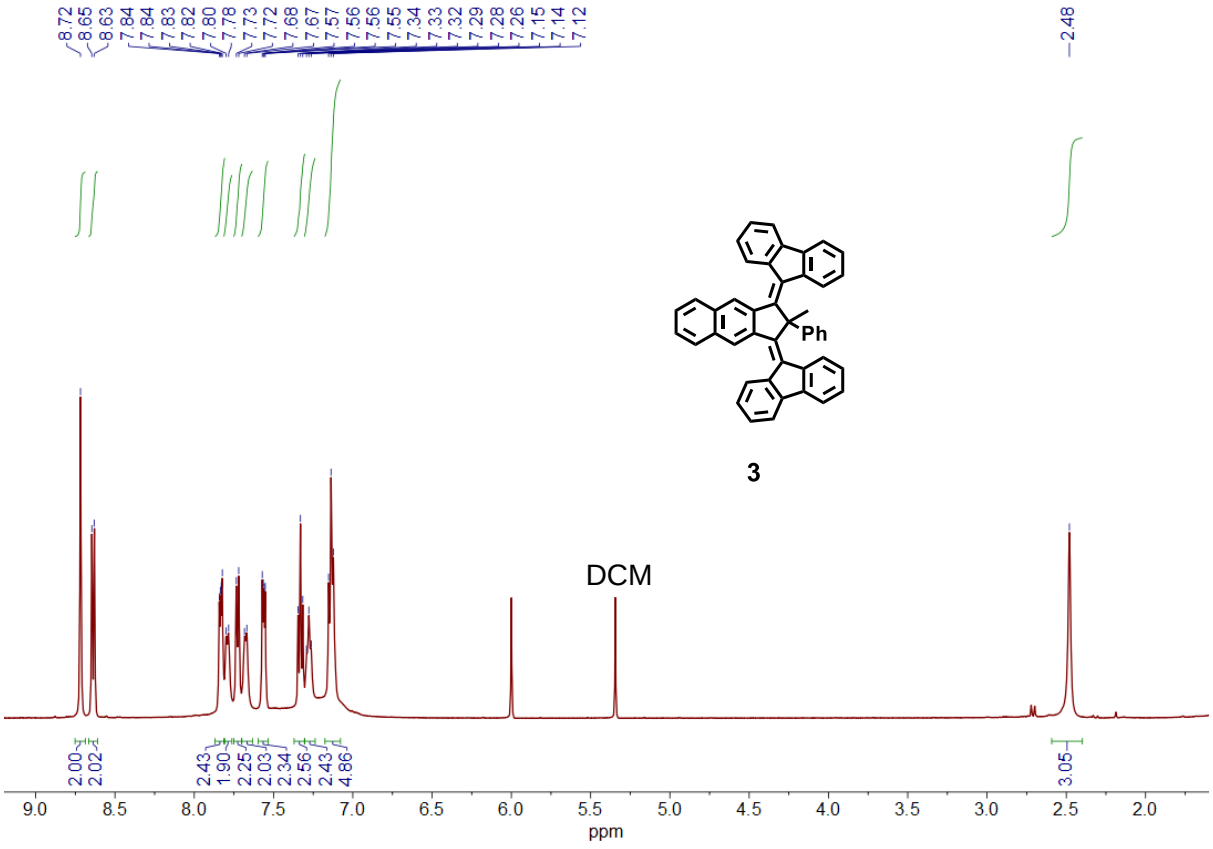


Figure S46. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 3.

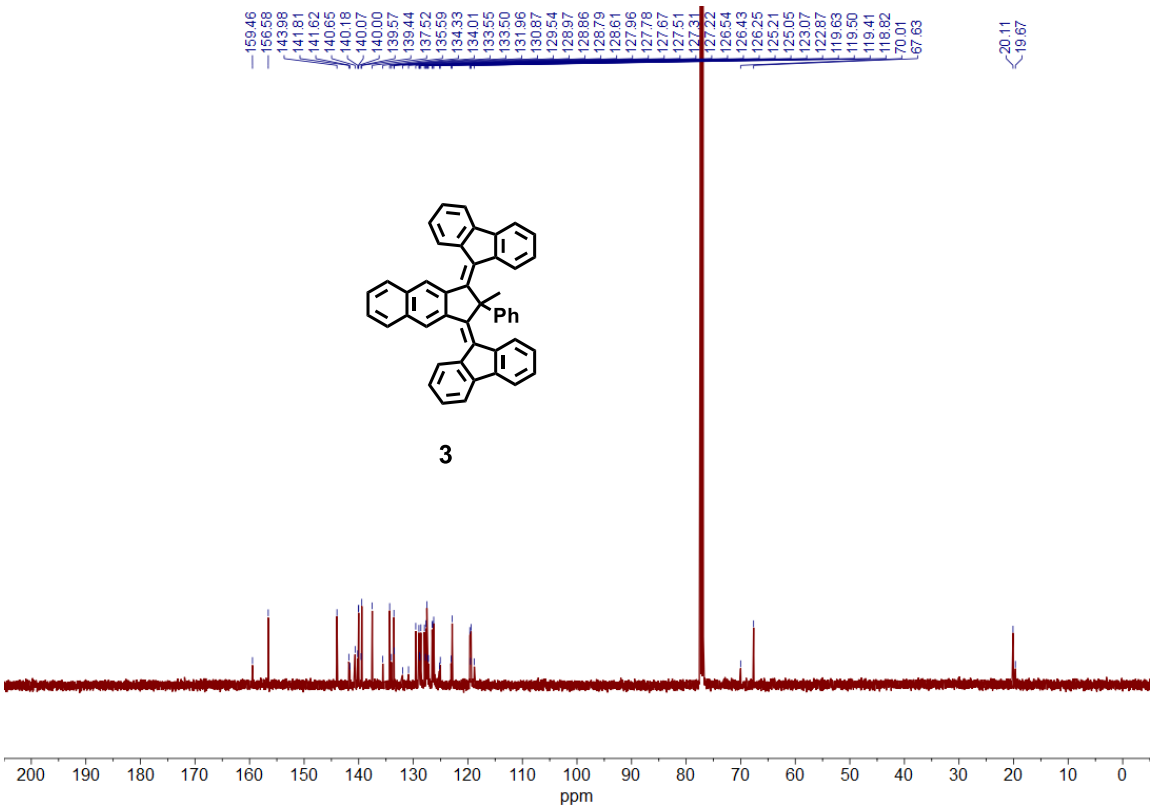


Figure S47. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 3.

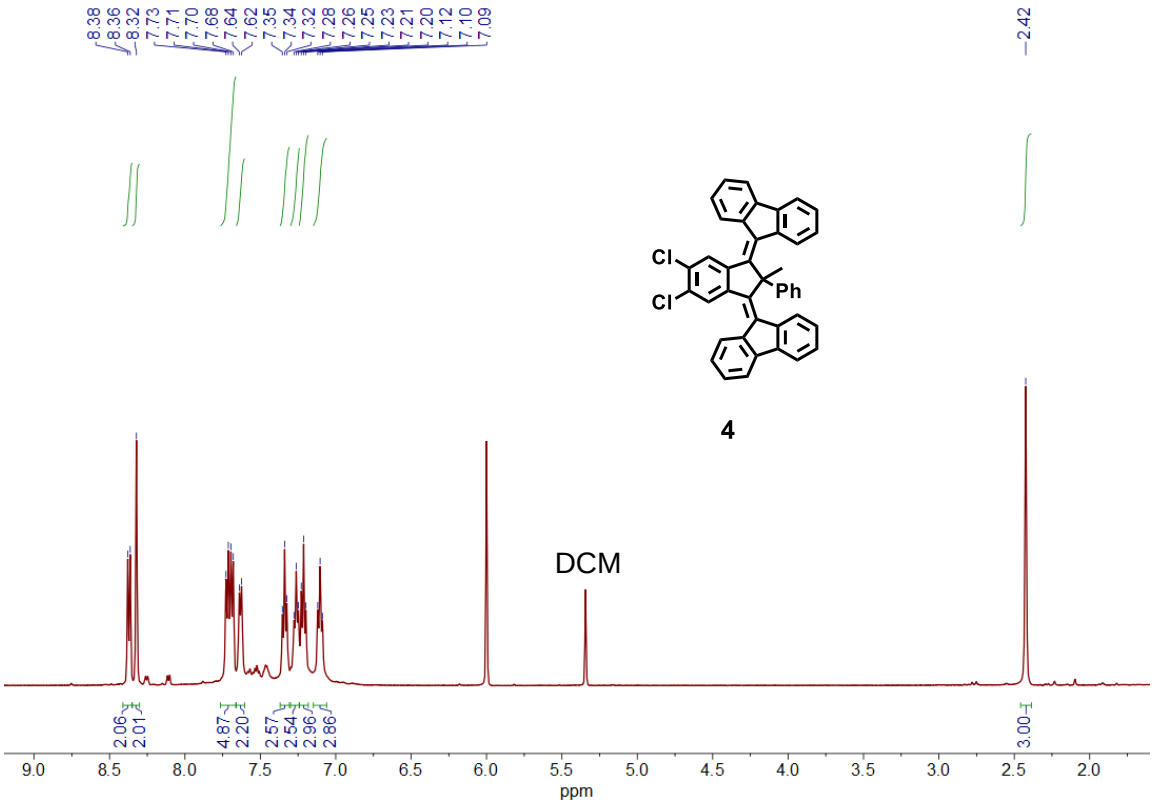


Figure S48. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 4.

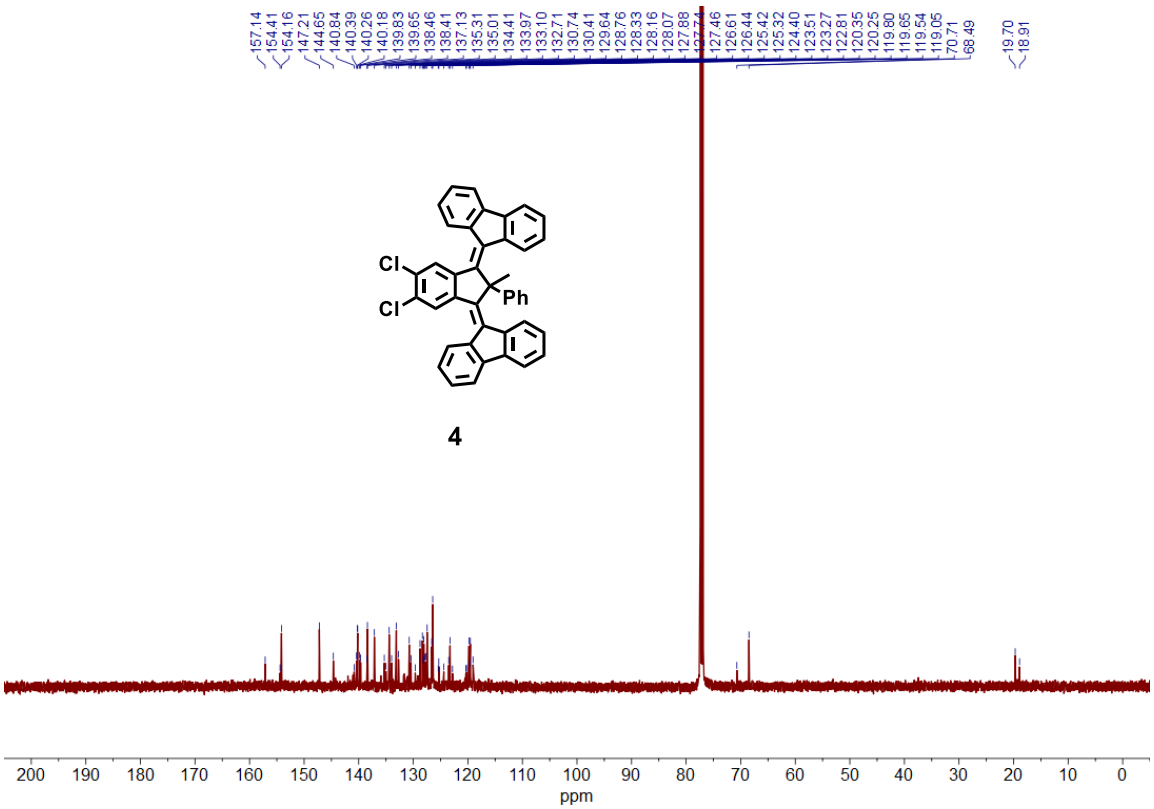


Figure S49. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 4.

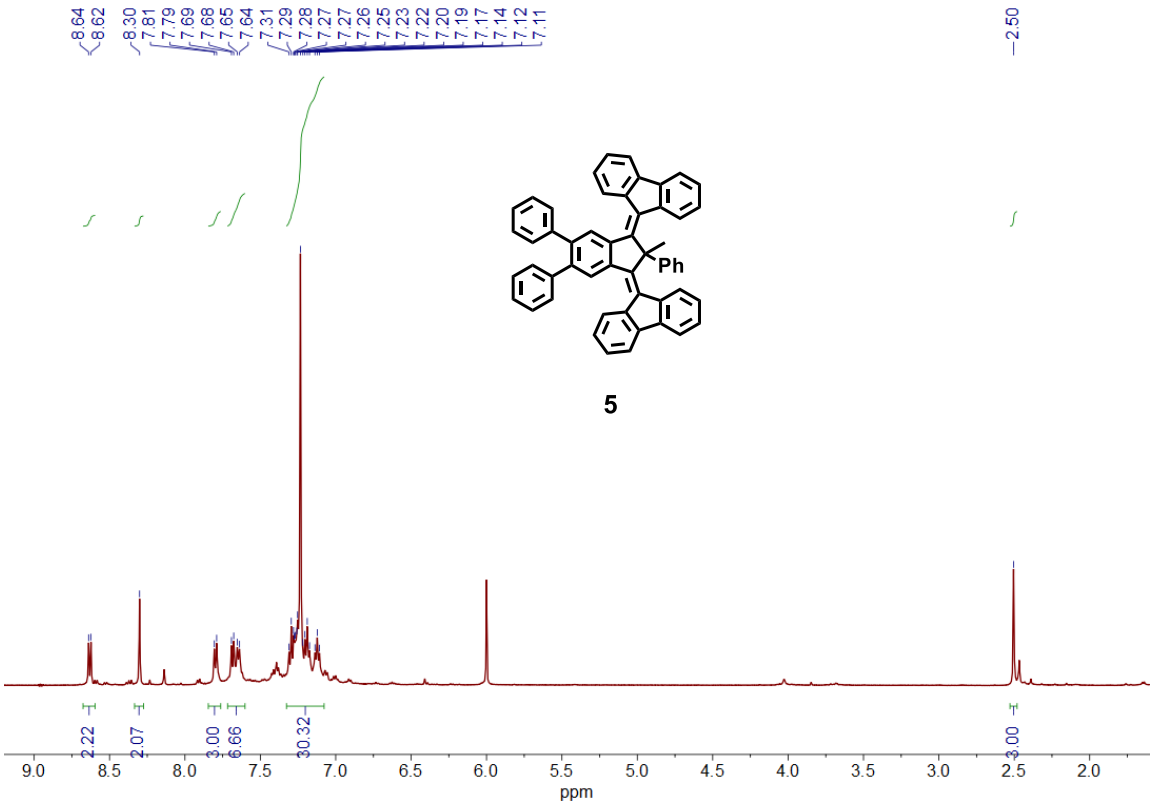


Figure S50. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 5.

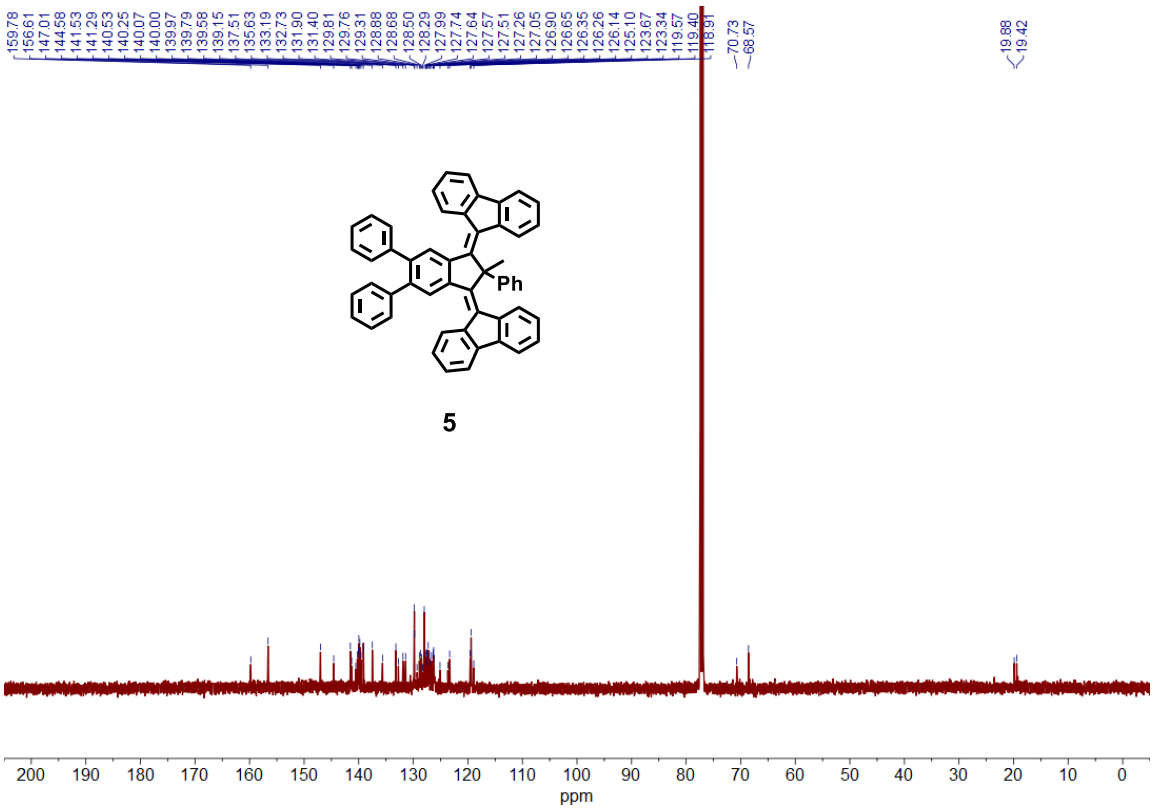


Figure S51. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 5.

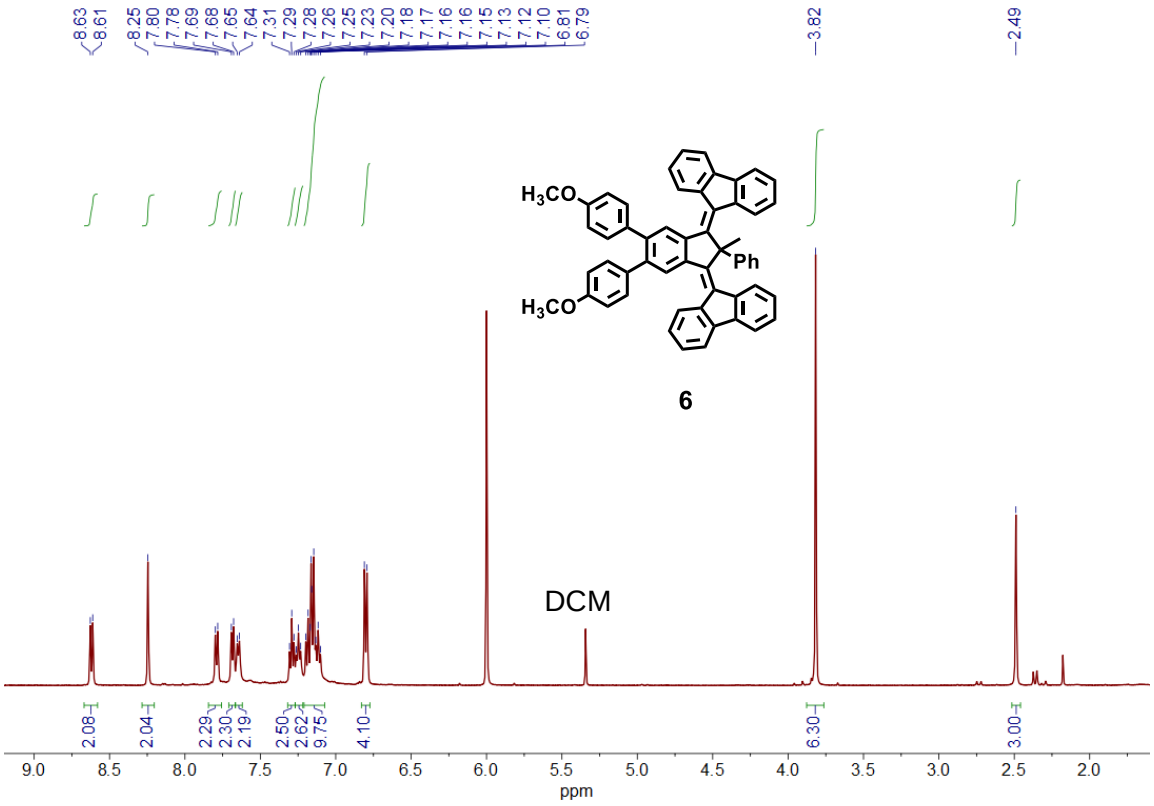


Figure S52. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 6.

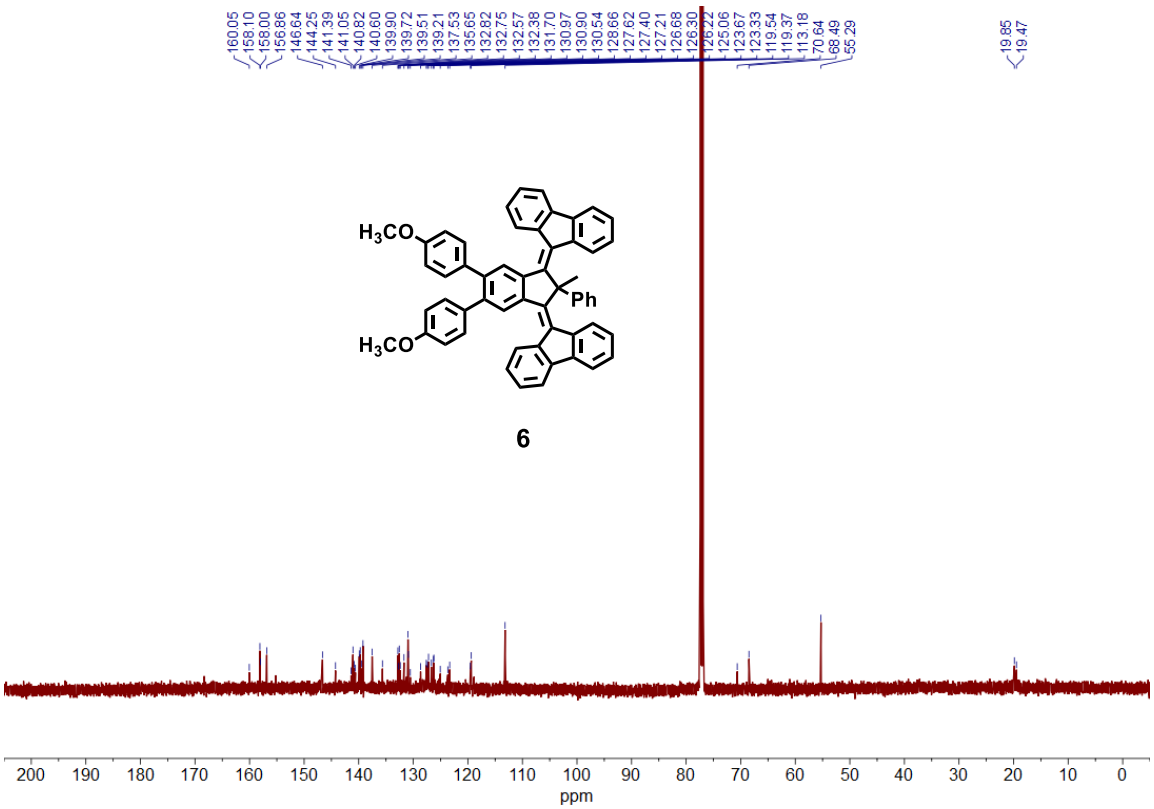


Figure S53. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 6.

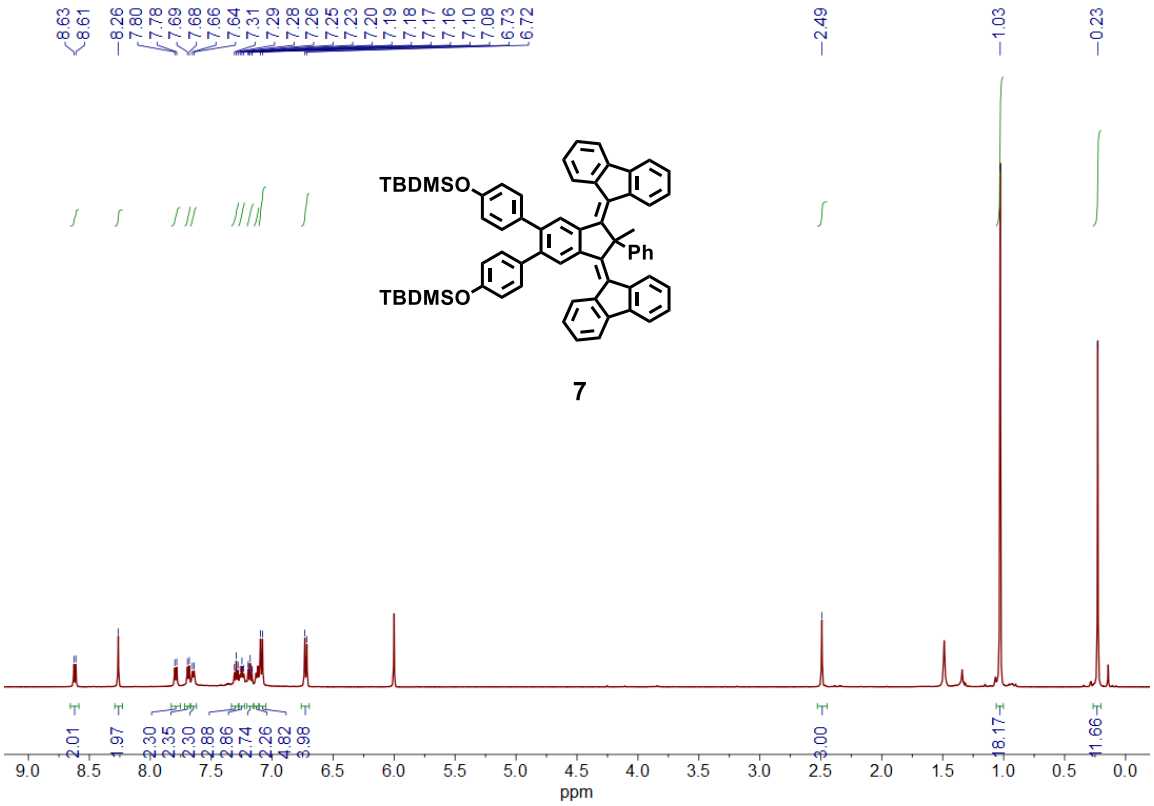


Figure S54. ^{1}H NMR spectrum (500 MHz, $Cl_2DCCDCl_2$, 90 °C) of 7.

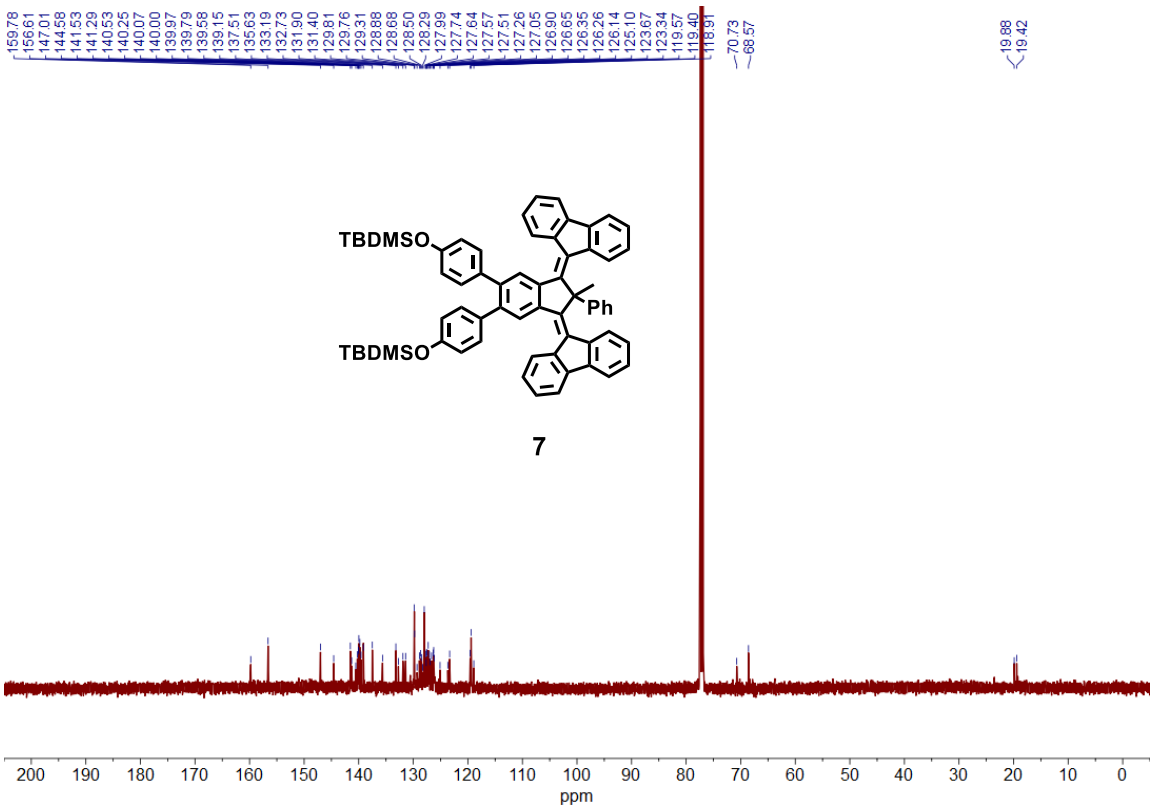


Figure S55. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 7.

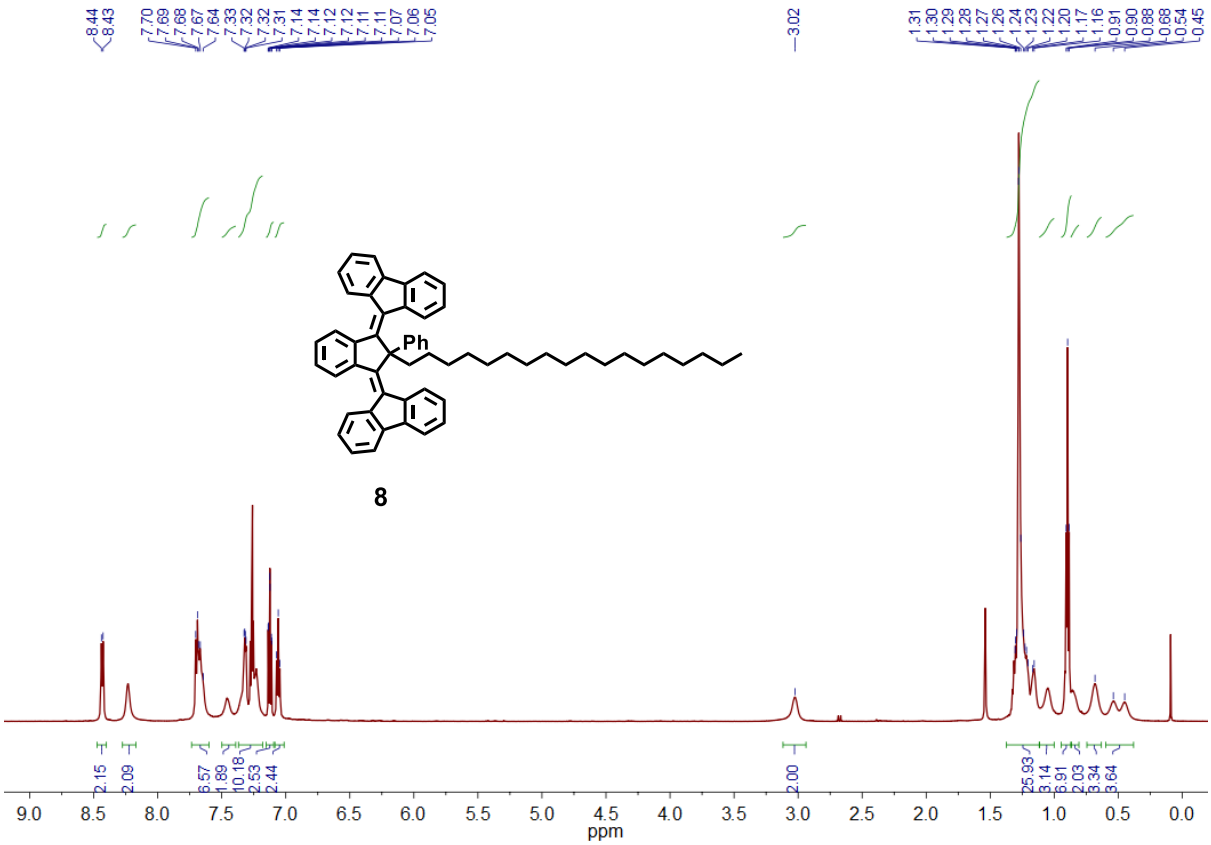


Figure S56. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 8.

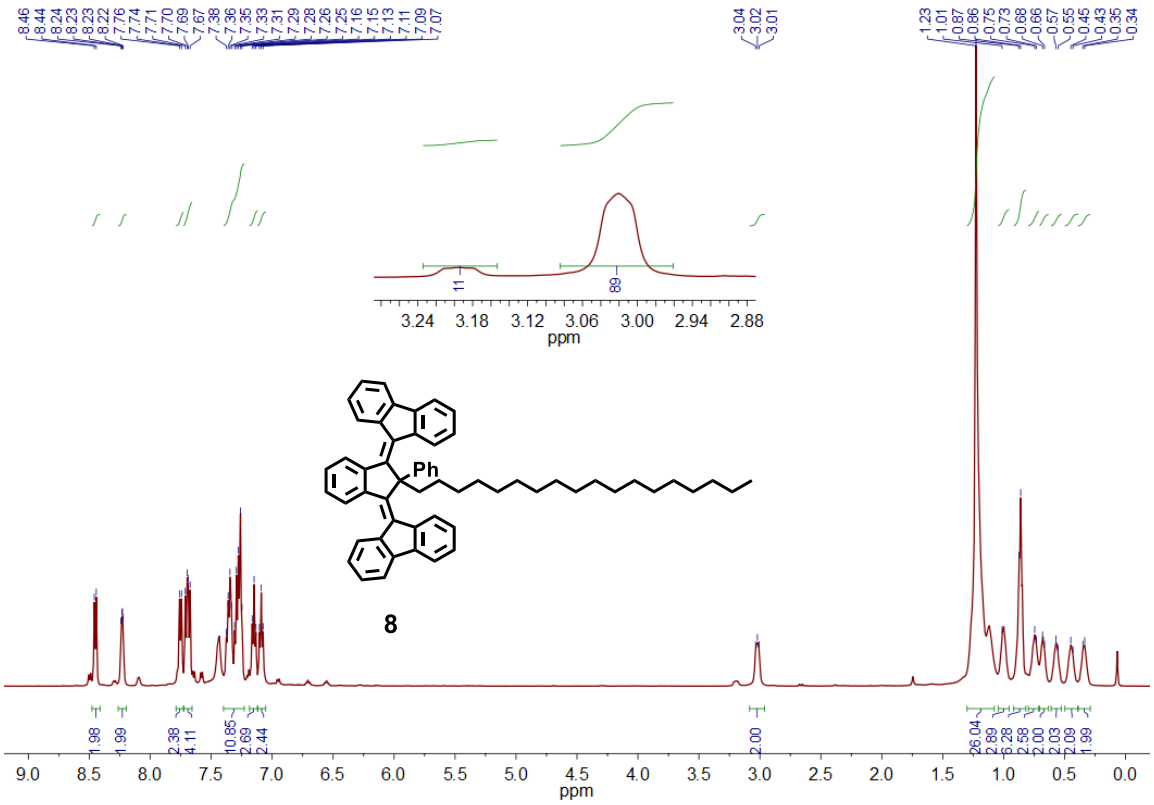


Figure S57. ^{1}H NMR spectrum (500 MHz, $CDCl_3$, -45 ºC) of 8.

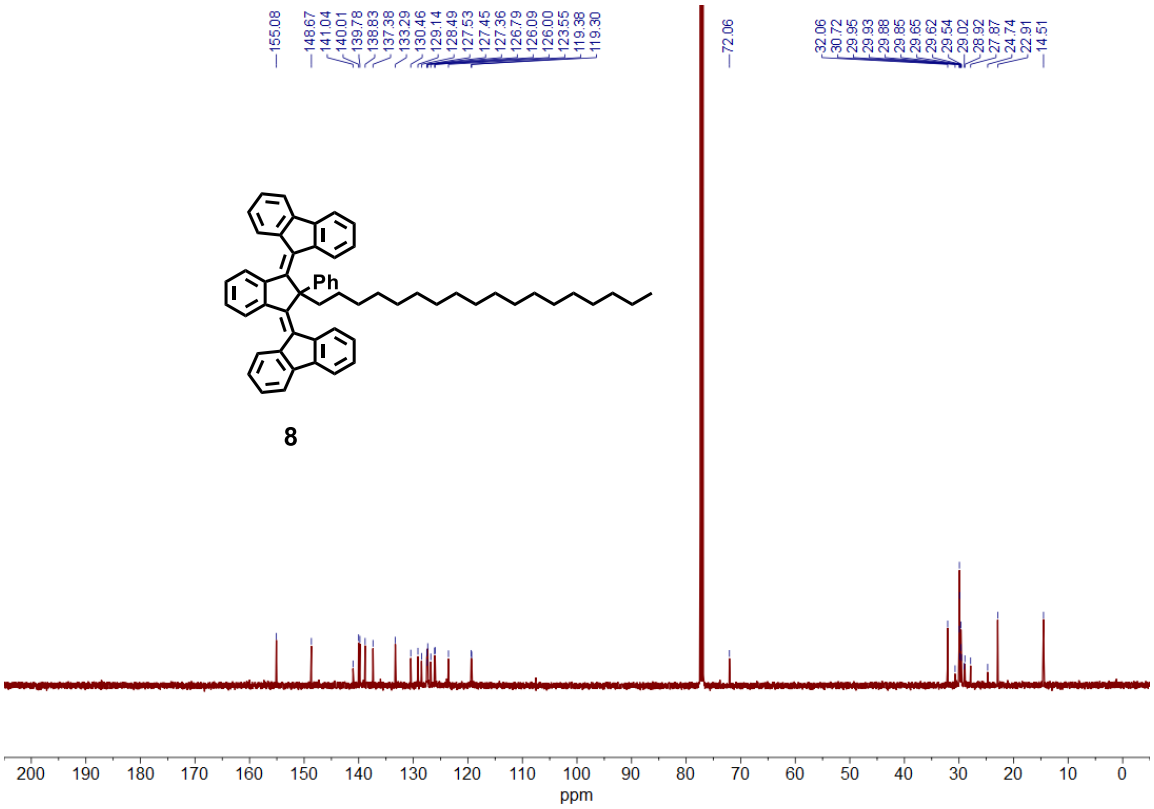


Figure S58. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 ºC) of 8.

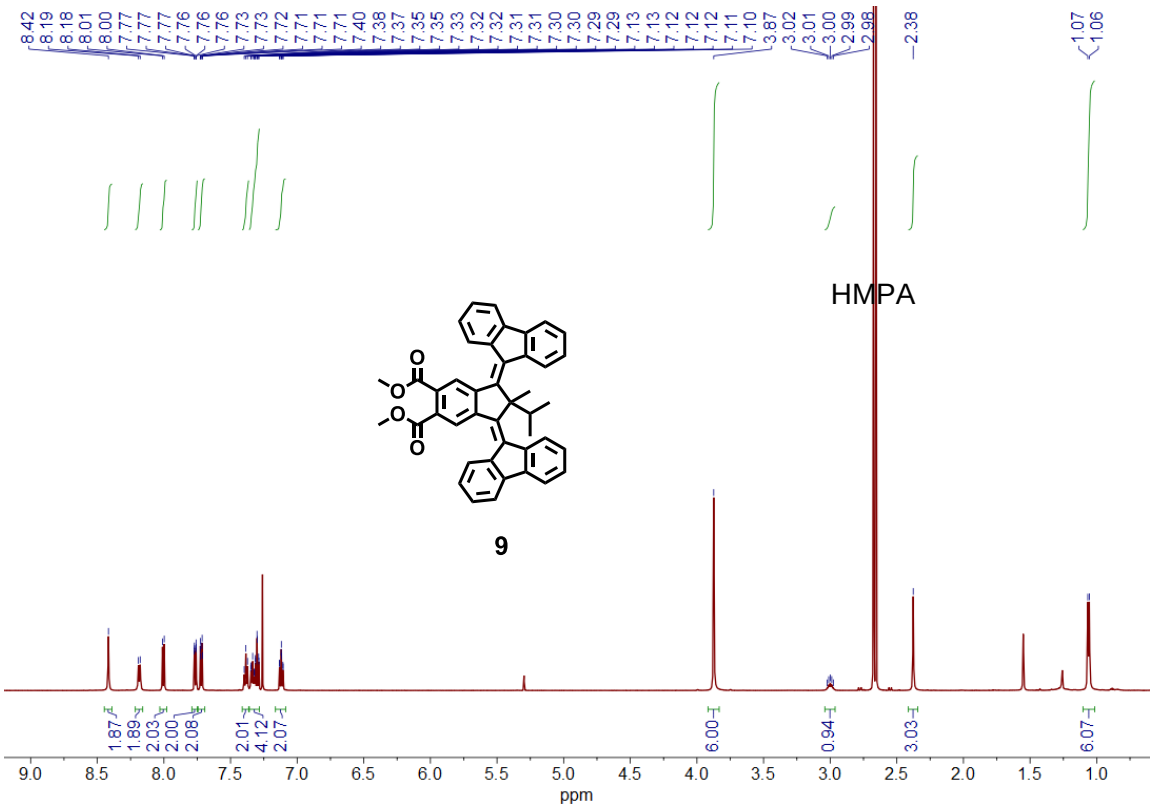


Figure S59. ^{1}H NMR spectrum (600 MHz, $CDCl_3$, 25 °C) of 9.

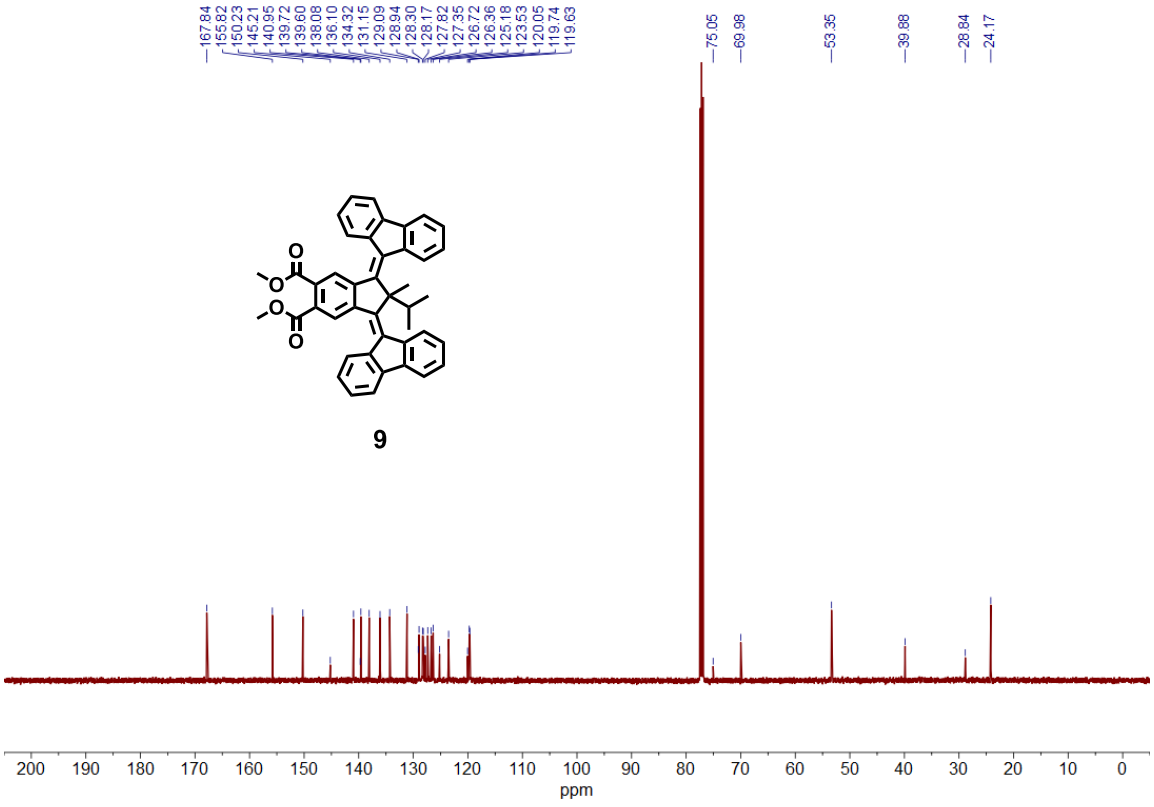


Figure S60. ^{13}C NMR spectrum (125 MHz, $CDCl_3$, -45 °C) of 9.

Isomeric ratio and degree of unidirectionality of motors 2-8

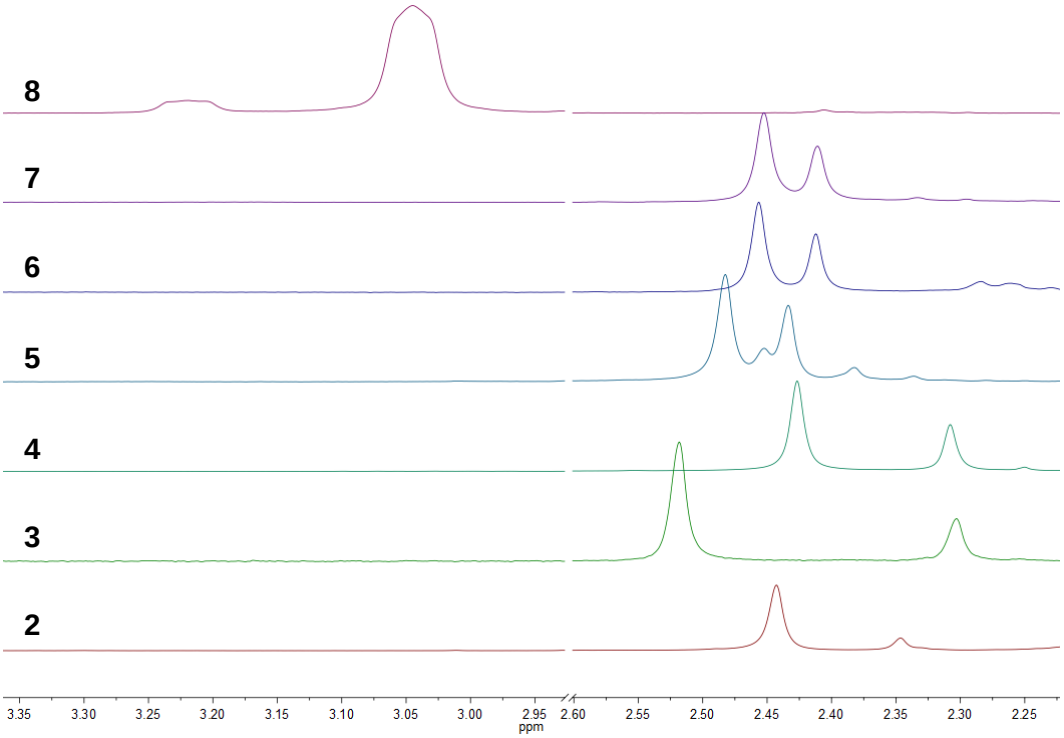


Figure S61. ^{1}H NMR spectra (500 MHz, $CDCl_3$, -45 ºC) of motors 8, 7, 6, 5, 4, 3 and 2 (from top to bottom). Integration of the signals allowed determining the ratio between isomers and degree of unidirectional rotary motion for these motors at the given temperature (see Table S1).

Table S1. Determination of the ratio between *r*- and *s*-isomer of motors 2-8, and their consequent degree of unidirectionality, in $CDCl_3$ at -45 ºC. Integral values and chemical shifts were obtained from analysis of the ^{1}H NMR spectra shown in Figure S61.

Motor	*r*-isomer chemical shift (ppm)	*r*-isomer percentage	*s*-isomer chemical shift (ppm)	*s*-isomer percentage	Degree of unidirectionality
2	3.22	17%	3.04	83%	66%
3	2.30	25%	2.52	75%	50%
4	2.31	34%	2.43	66%	32%
5	2.43	42%	2.48	58%	16%
6	2.41	38%	2.46	62%	24%
7	2.41	38%	2.45	62%	24%
8	3.22	11%	3.04	89%	78%

UV-vis spectra of motors 1-9

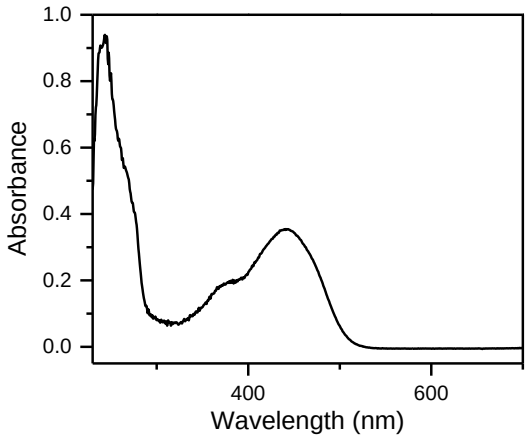


Figure S62. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 1.

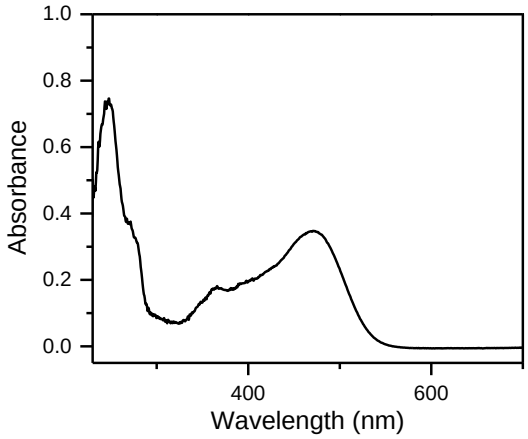


Figure S63. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 2.

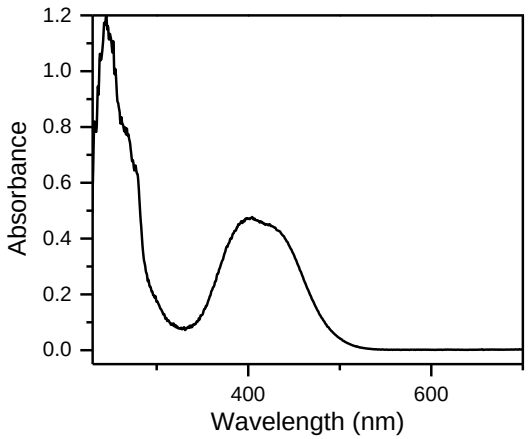


Figure S64. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 3.

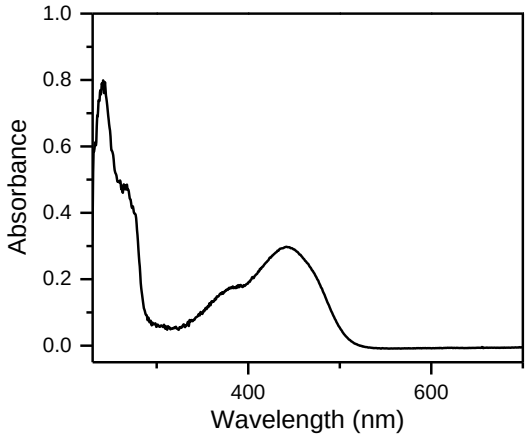


Figure S65. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 ºC) of 4.

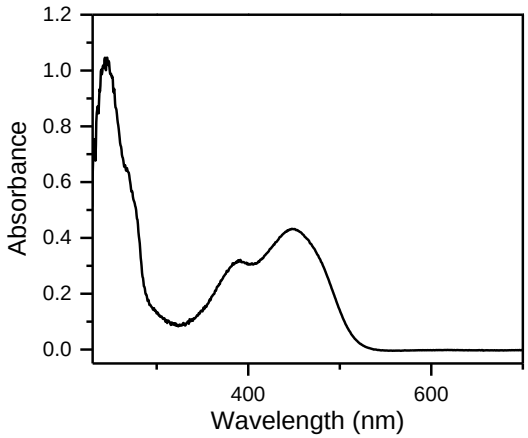


Figure S66. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 ºC) of 5.

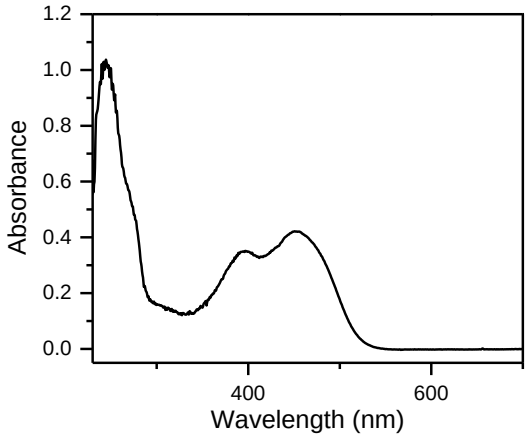


Figure S67. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 ºC) of 6.

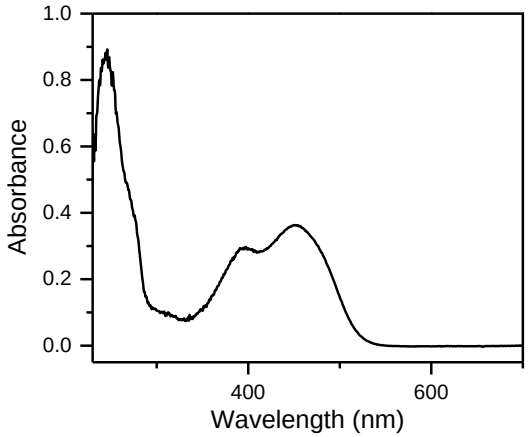


Figure S68. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 7.

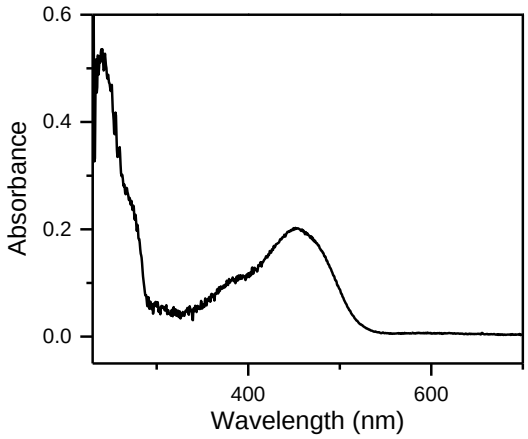


Figure S69. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 8.

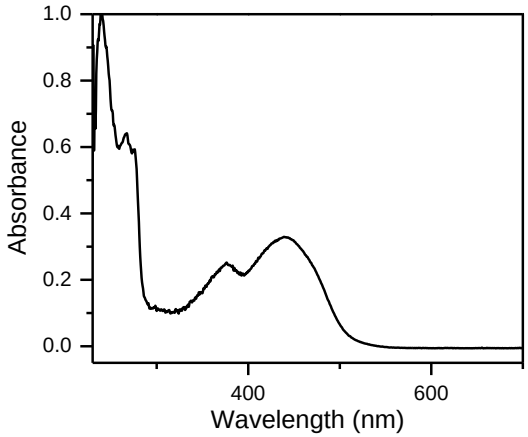


Figure S70. UV-vis spectrum (CH_2Cl_2, 5×10^{-6} M, 25 °C) of 9.

IR spectra

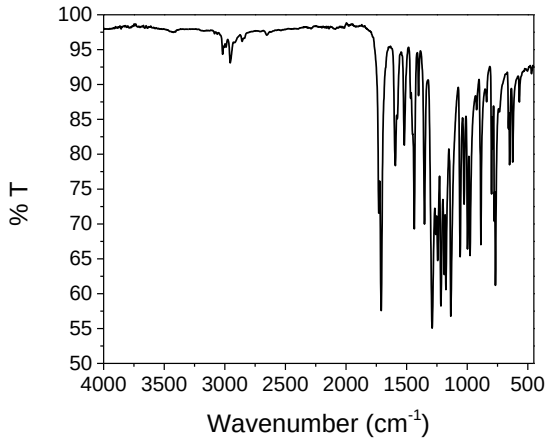


Figure S71. FT-IR spectrum of 11.

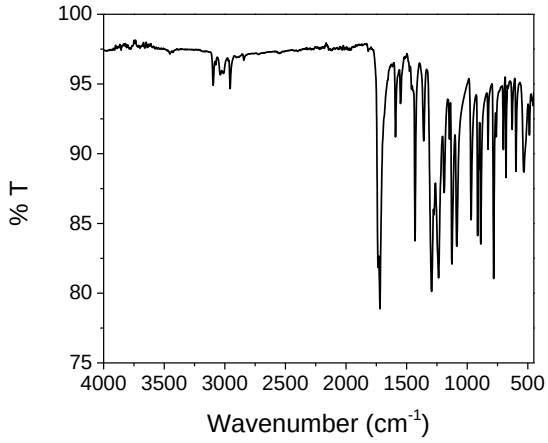


Figure S72. FT-IR spectrum of 13.

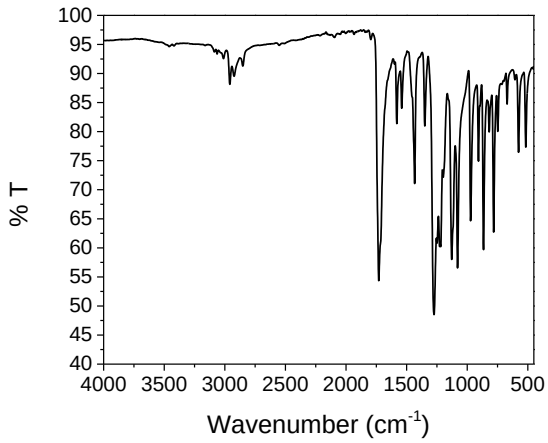


Figure S73. FT-IR spectrum of 14.

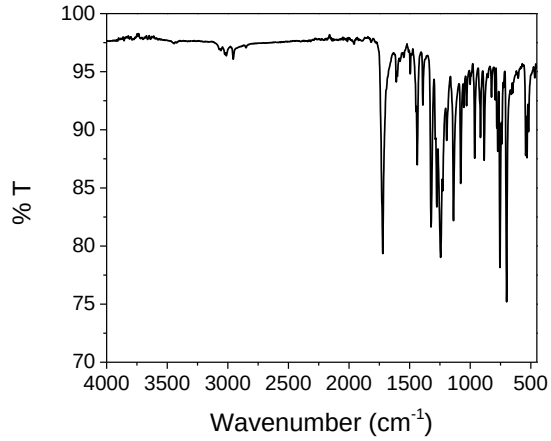


Figure S74. FT-IR spectrum of 15.

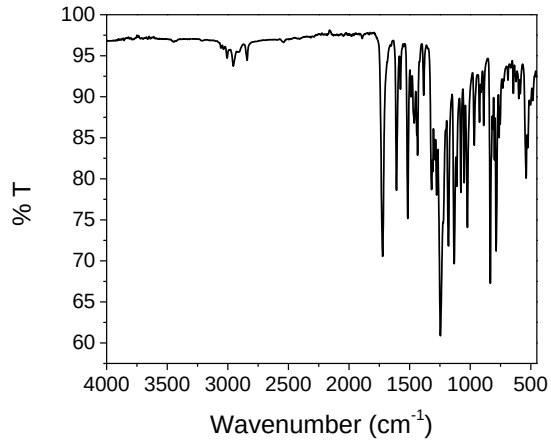


Figure S75. FT-IR spectrum of 16.

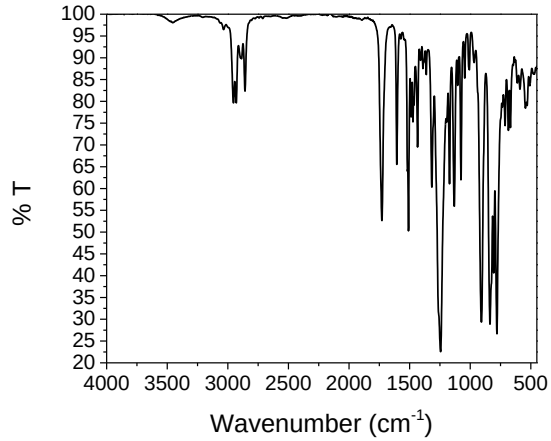


Figure S76. FT-IR spectrum of 17.

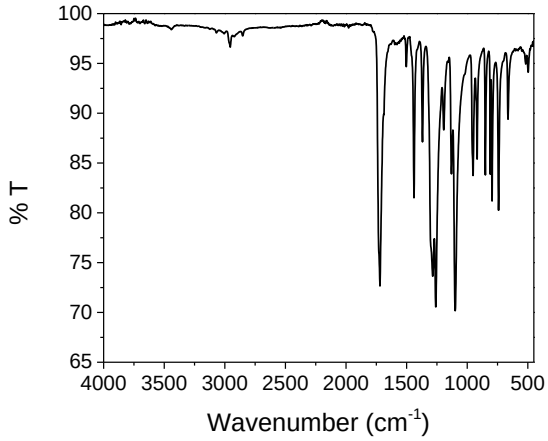


Figure S77. FT-IR spectrum of 18.

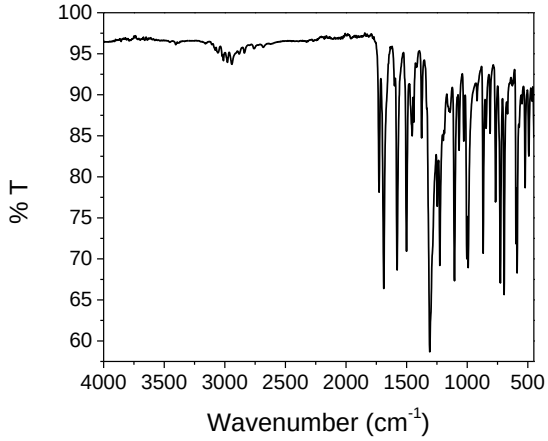


Figure S78. FT-IR spectrum of 20.

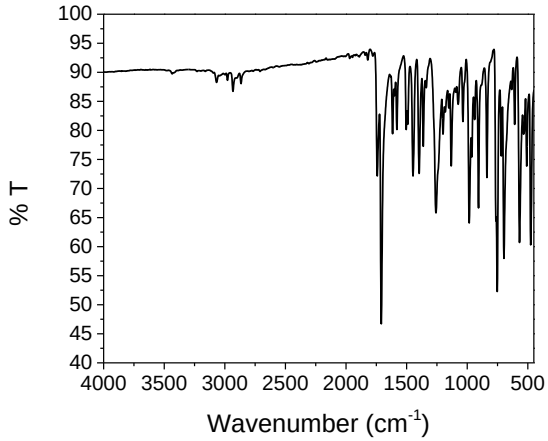


Figure S79. FT-IR spectrum of 21.

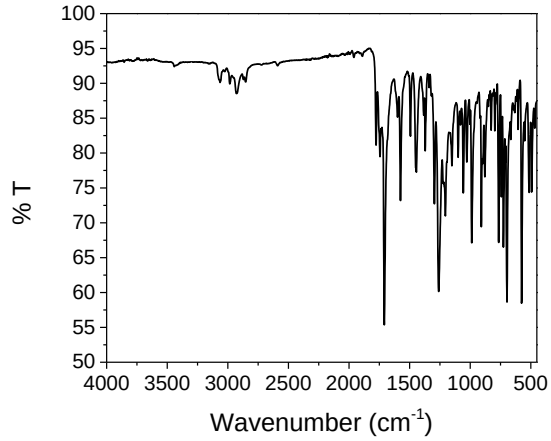


Figure S80. FT-IR spectrum of 22.

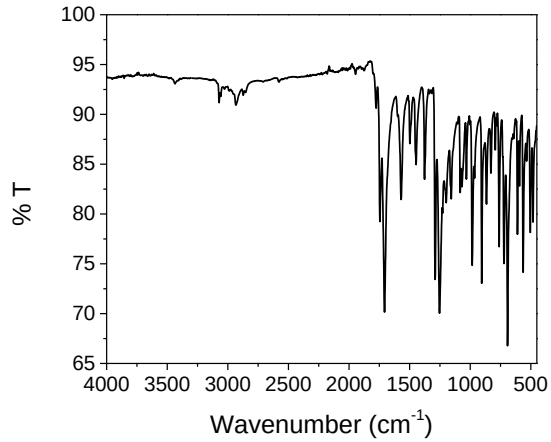


Figure S81. FT-IR spectrum of 23.

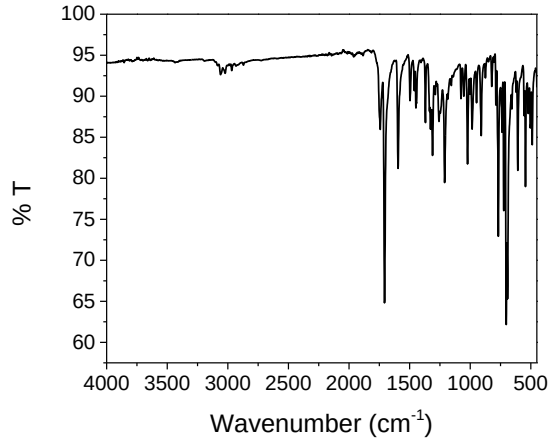


Figure S82. FT-IR spectrum of 24.

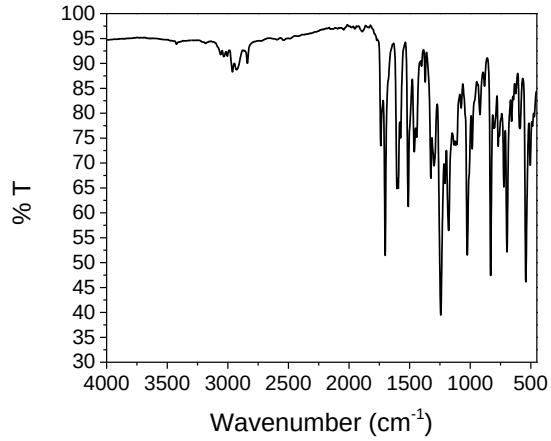


Figure S83. FT-IR spectrum of 25.

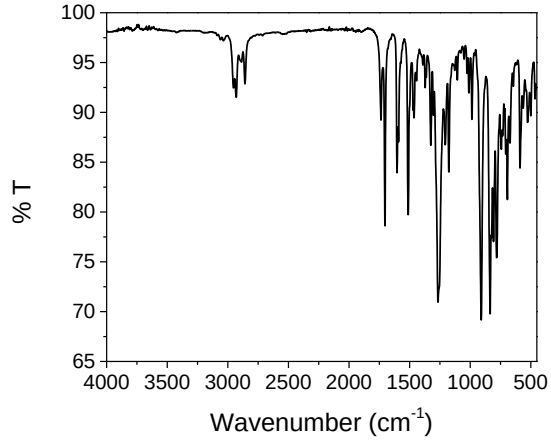


Figure S84. FT-IR spectrum of 26.

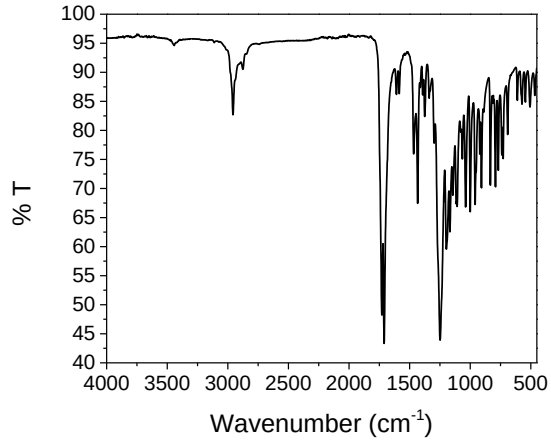


Figure S85. FT-IR spectrum of 27.

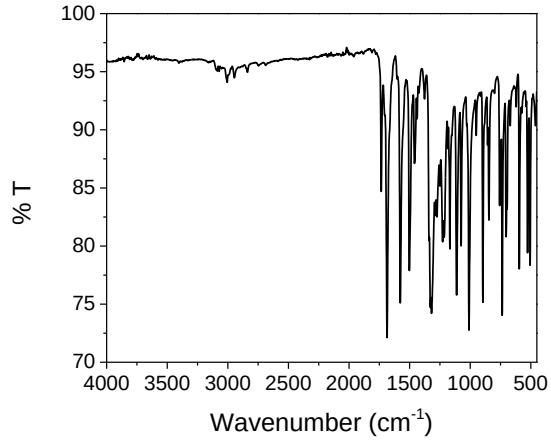


Figure S86. FT-IR spectrum of 29.

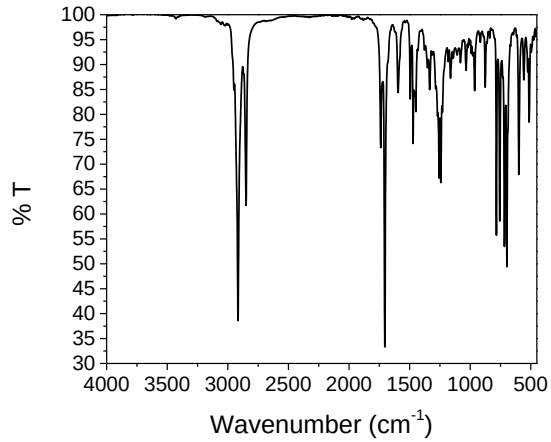


Figure S87. FT-IR spectrum of 30.

Crystal structures

Single-Crystal X-Ray Structures

Single-crystals were obtained by slow diffusion of hexane into saturated solutions of **2**, **4** and **5** in DCE. Figure S87 shows displacement ellipsoid plots of the refined structures.

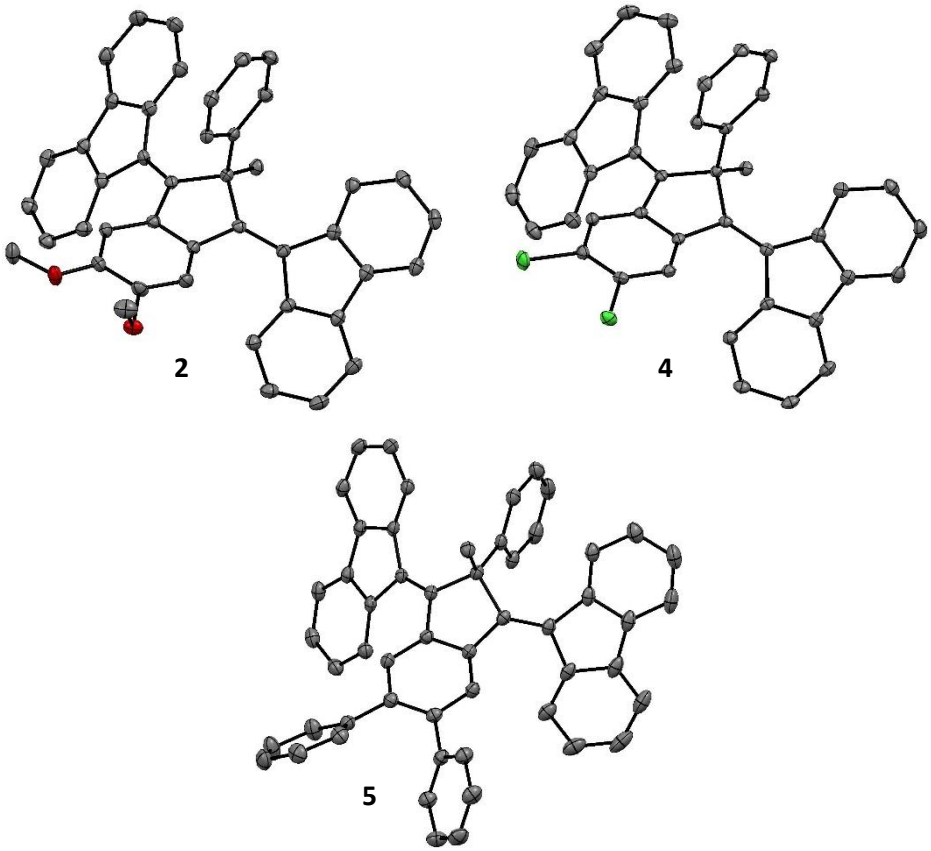


Figure S88. Single-crystal X-ray structures of compounds **2**, **4** and **5**. Displacement ellipsoid plot drawn at 50% probability; hydrogens and solvent molecules are omitted for clarity.

Table S2 shows a summary of critical parameters of single-crystal X-ray structures of compounds **2**, **4** and **5**.

Table S2. Summary of critical parameters of single-crystal X-ray structures of compounds **2**, **4** and **5**.

Nr.	**2**	**4**	**5**
Name	9,9'-(5,6-Dimethoxy-2-methyl-2-phenyl-1*H*-indene-1,3(2*H*)-diylidene)bis(9*H*-fluorene)	9,9'-(5,6-Dichloro-2-methyl-2-phenyl-1*H*-indene-1,3(2*H*)-diylidene)bis(9*H*-fluorene)	9,9'-(2-Methyl-2,5,6-triphenyl-1*H*-indene-1,3(2*H*)-diylidene)bis(9*H*-fluorene)
Formula	$C_{44}H_{32}O_2$	$C_{42}H_{26}Cl_2$	$C_{54}H_{36}$, $C_2H_4Cl_2$
Molecular Weight	592.69	601.53	783.78
Crystal System	monoclinic	monoclinic	monoclinic
T [K]	100(2)	100(2)	100(2)
Space Group	P 21/c	P 21/n	P 21/n
a [Å]	14.1264(3)	15.4794(4)	13.4014(3)
b [Å]	11.2302(5)	11.3565(3)	16.7991(4)
c [Å]	20.1932(5)	17.3278(4)	17.6974(4)
α [°]	90	90	90
β [°]	108.1510(10)	100.2820(10)	94.0590(10)
γ [°]	90	90	90
V [$Å^3$]	3044.08(13)	2997.17(13)	3974.25(16)
Z	4	4	4
D_{calc} [$g \cdot cm^{-3}$]	1.293	1.333	1.310
F(0 0 0)	1248	1248	1640
h_{min}, h_{max}	–17, 17	–19, 19	–16, 16
k_{min}, k_{max}	–12, 14	–14, 14	–20, 20
l_{min}, l_{max}	–25, 25	–21, 21	–21, 22
μ [mm^{-1}]	0.602	2.172	1.765
Crystal Size [mm]	0.20 x 0.20 x 0.10	0.20 x 0.20 x 0.15	0.24 x 0.20 x 0.18
Colour, Shape	clear_pale_red plate	clear_pale_red block	clear_pale_orange block
R_{int}	0.0320	0.0537	0.0598
θ_{min}, θ_{max} [°]	3.292, 79.075	3.529, 77.368	3.632, 74.493
Total Reflections (before merge)	31122	61550	72237
Data (I>3 x sigma(I)) [Reflections,Restraints,Parameters]	6492, 438, 0	6345, 398, 0	8113, 561, 5
S (=GooF)	1.027	1.007	1.053
Min. Residual Density [e/$Å^3$]	–0.239	–0.310	–0.595
Max. Residual Density [e/Å3]	0.353	0.638	0.821
Threshold Expression	I>2sigma(I)	I>2sigma(I)	I>2sigma(I)
R_1	0.0479	0.0437	0.0578
wR_2	0.1059	0.0938	0.1296

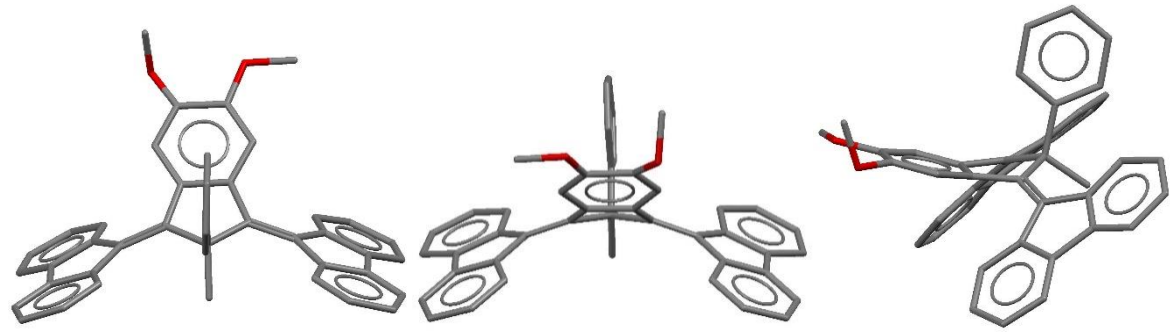


Figure S89. Front (left), back (middle) and side view of single-crystal X-ray structure of **2**. Hydrogens are omitted for clarity.

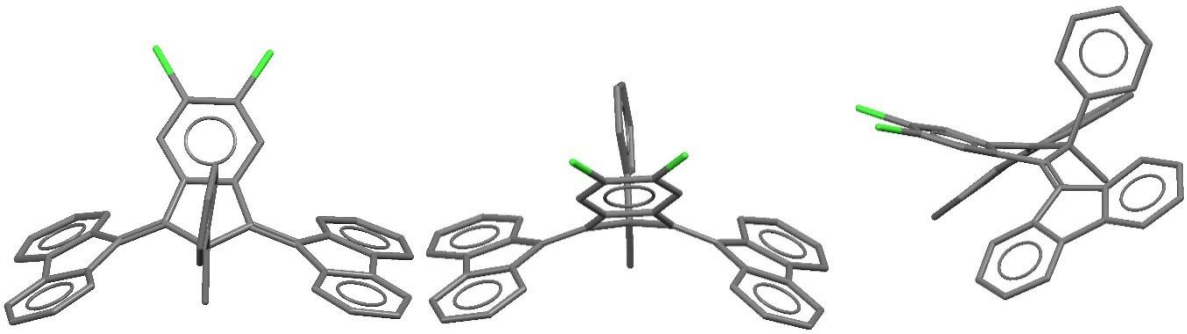


Figure S90. Front (left), back (middle) and side view of single-crystal X-ray structure of **4**. Hydrogens are omitted for clarity.

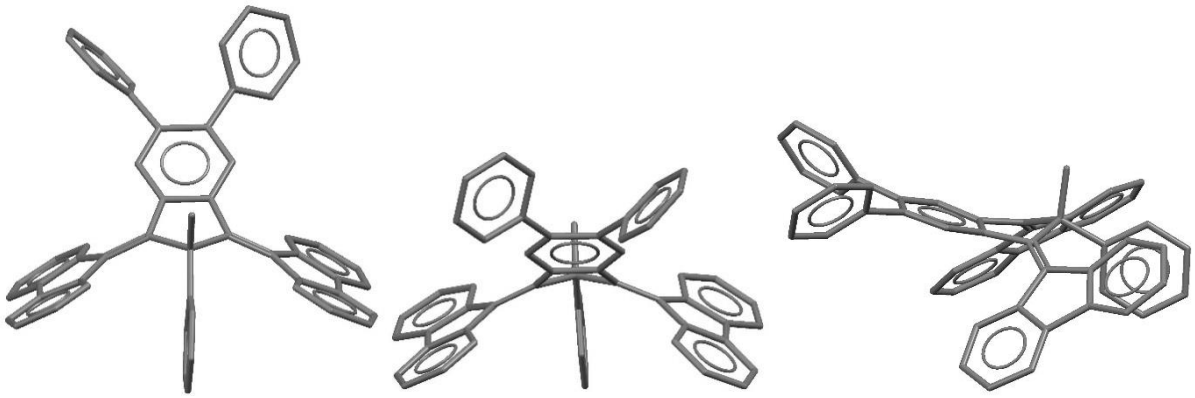


Figure S91. Front (left), back (middle) and side view of single-crystal X-ray structure of **5**. Hydrogens are omitted for clarity.

TD-^{1}H NMR experiments with motor 6

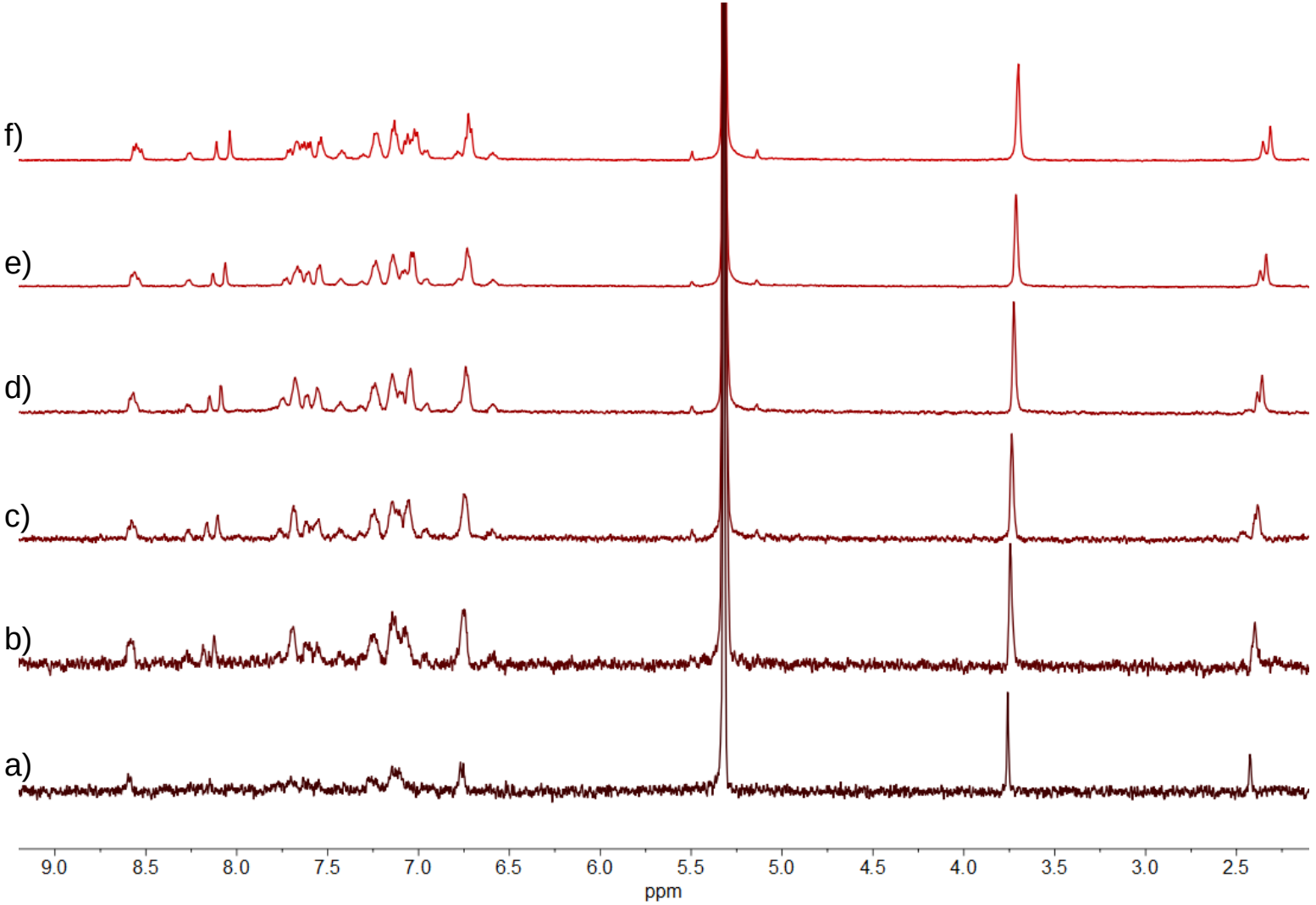


Figure S92. ^{1}H-NMR (CD_2Cl_2, 500 MHz) spectra of motor 6 at: a) 25 ºC; b) 0 ºC; c) -20 ºC; d) -40 ºC; e) -60 ºC; f) -80 ºC.

^{1}H NMR *in situ* irradiation experiments at -80 ºC with motor 6

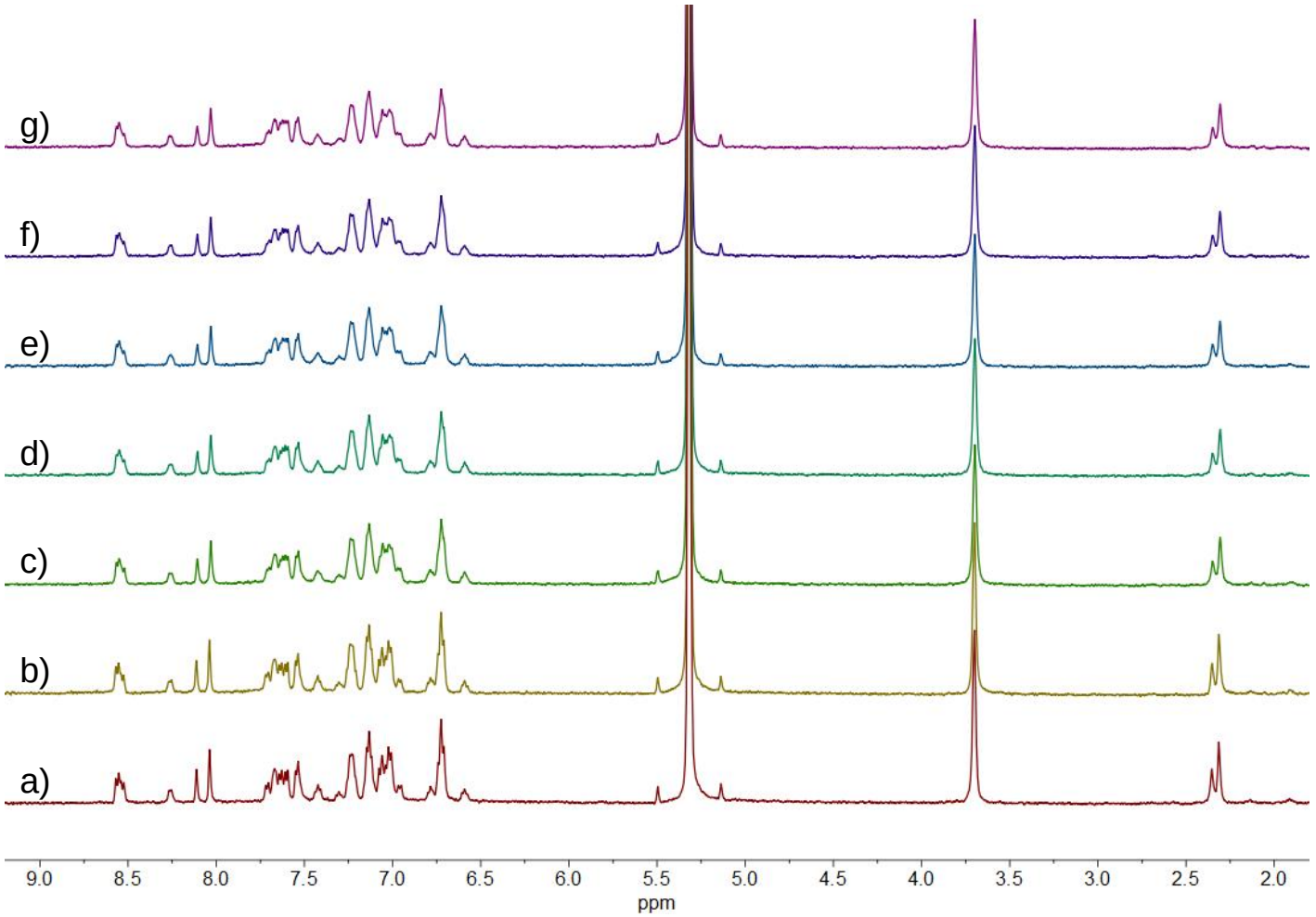


Figure S93. ***In situ*** **^{1}H-NMR (CD_2Cl_2, 500 MHz, -80ºC) irradiation experiment on motor 6. Traces: a) 6 at -80 ºC; b) while irradiating with 365 nm light; c) 1 minute after irradiation with 365 nm light; d) while irradiating with 395 nm light; e) 1 minute after irradiation with 395 nm light; f) while irradiating with 455 nm light; g) 1 minute after irradiation with 455 nm light.**

References

S1 Buckle, D. R.; Morgan, N. J.; Ross, J. W.; Smith, H.; Spicer, B. A. Antiallergic Activity of 2-Nitroindan-l,3-Diones. *J. Med. Chem.* **1973**, *16* (12), 1334–1339.

S2 Kistemaker, J. C. M.; Štacko, P.; Roke, D.; Wolters, A. T.; Heideman, G. H.; Chang, M. C.; Van Der Meulen, P.; Visser, J.; Otten, E.; Feringa, B. L. Third-Generation Light-Driven Symmetric Molecular Motors. *J. Am. Chem. Soc.* **2017**, *139* (28), 9650–9661.