Electronic Supplementary Information

Solvent-free *N*-iodosuccinimide-promoted synthesis of spiroimidazolines from alkenes and amidines under ball-milling conditions

Hui Xu,[a] Kuan Chen,[a] Hong-Wei Liu[a] and Guan-Wu Wang*[a,b]

[a] *CAS Key Laboratory of Soft Matter Chemistry, iChEM (Collaborative Innovation Center of Chemistry for Energy Materials), Hefei National Laboratory for Physical Sciences at Microscale, and Department of Chemistry, University of Science and Technology of China, Hefei, Anhui 230026, P. R. China.*
[b] *State Key Laboratory of Applied Organic Chemistry, Lanzhou University, Lanzhou, Gansu 730000, P. R. China*

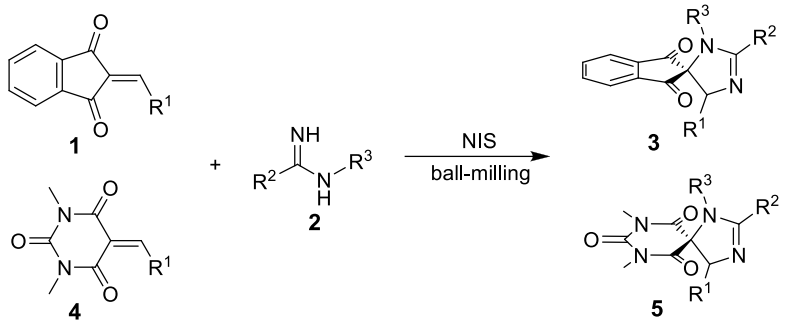
E-mail: gwang@ustc.edu.cn

Table of contents

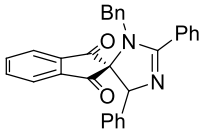
1. General information

All reagents were obtained from commercial sources and used without further purification. NMR spectra were recorded on a 400 MHz NMR spectrometer (400 MHz for ^{1}H NMR; 101 MHz for ^{13}C NMR). ^{1}H NMR chemical shifts were determined relative to internal TMS at δ 0.0 ppm. ^{13}C NMR chemical shifts were determined relative to $CDCl_3$ at δ 77.16 ppm. Data for ^{1}H NMR and ^{13}C NMR are reported as follows: chemical shift (δ, ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet, sept = septet). High-resolution mass spectra (HRMS) were measured with ESI-TOF in a positive mode. Ball-milling reactions were performed in a MM400 mixer mill (Retsch GmbH, Haan, Germany).

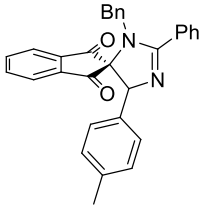
2. Synthesis and characterization of 3 and 5



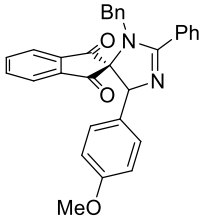
A mixture of alkenes (0.2 mmol), amidines (0.3 mmol) and NIS (0.24 mmol) together with a stainless ball (10 mm in diameter) were introduced into a stainless steel jar (10 mL). The reaction vessel along with another identical empty vessel was closed and fixed on the vibration arms of a Retsch MM400 mixer mill, and was vibrated vigorously at a rate of 1800 rounds per minute (30 Hz) at room temperature for 20 min. After completion of the reaction, the reaction vessel was washed with acetone three times (3 × 6 mL), and the combined solution was evaporated to remove the solvent in vacuo. The residue was separated by flash column chromatography on silica gel with acetone/petroleum ether as the eluent to afford spiroimidazolines.



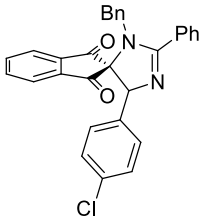
3-Benzyl-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3aa). General procedure was followed to afford **3aa** as a yellow solid (79.4 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87–7.81 (m, 2H), 7.73 (d, J = 7.5 Hz, 1H), 7.67 (td, J = 7.4, 1.0 Hz, 1H), 7.61 (td, J = 7.4, 1.2 Hz, 1H), 7.53–7.45 (m, 3H), 7.40 (d, J = 7.5 Hz, 1H), 7.13–6.94 (m, 10H), 5.60 (s, 1H), 4.59 (d, J = 14.8 Hz, 1H), 4.11 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.4, 196.0, 167.4, 141.1, 140.9, 136.9, 136.1, 135.54, 135.49, 130.6, 130.4, 129.4 (2C), 128.8 (2C), 128.7 (2C), 128.1 (2C), 127.95 (4C), 127.92, 127.8, 123.0, 122.9, 80.3, 79.8, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{23}N_2O_2$ $[M + H]^+$ 443.1760, found 443.1761.



3-Benzyl-2-phenyl-5-*p*-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ba). General procedure was followed to afford **3ba** as a yellow solid (74.0 mg, 81% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87–7.80 (m, 2H), 7.73 (d, J = 7.2 Hz, 1H), 7.67 (td, J = 7.3, 1.0 Hz, 1H), 7.62 (td, J = 7.3, 1.0 Hz, 1H), 7.52–7.45 (m, 3H), 7.43 (d, J = 7.4 Hz, 1H), 7.06–6.95 (m, 5H), 6.88 (d, J = 8.1 Hz, 2H), 6.84 (d, J = 8.1 Hz, 2H), 5.57 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.11 (d, J = 14.8 Hz, 1H), 2.20 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.5, 196.1, 167.2, 141.1, 141.0, 137.4, 136.0, 135.53, 135.48, 133.9, 130.6, 130.4, 129.4 (2C), 128.9 (2C), 128.71 (2C), 128.65 (2C), 128.1 (2C), 127.9, 127.8 (2C), 123.0, 122.9, 80.2, 79.6, 50.6, 21.2; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1926.

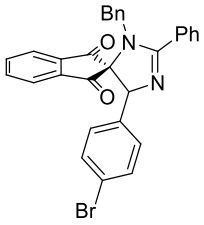


3-Benzyl-5-(4-methoxyphenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ca). General procedure was followed to afford **3ca** as a yellow solid (71.0 mg, 75% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86–7.80 (m, 2H), 7.74–7.71 (m, 1H), 7.67 (td, J = 7.3, 1.2 Hz, 1H), 7.62 (td, J = 7.3, 1.4 Hz, 1H), 7.52–7.46 (m, 3H), 7.46–7.42 (m, 1H), 7.06–6.96 (m, 5H), 6.88 (d, J = 8.7 Hz, 2H), 6.61 (d, J = 8.7 Hz, 2H), 5.55 (s, 1H), 4.59 (d, J = 14.8 Hz, 1H), 4.12 (d, J = 14.8 Hz, 1H), 3.69 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.5, 196.1, 167.1, 159.1, 141.1, 140.9, 136.1, 135.52, 135.50, 130.6, 130.4, 129.4 (2C), 129.14 (2C), 129.05, 128.8 (2C), 128.7 (2C), 128.1 (2C), 127.9, 123.0, 122.9, 113.3 (2C), 80.4, 79.5, 55.2, 50.6; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_3$ $[M + H]^+$ 473.1865, found 473.1878.

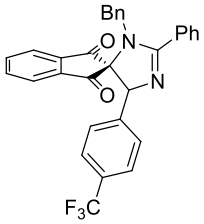


3-Benzyl-5-(4-chlorophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3da). General procedure was followed to afford **3da** as a yellow solid (79.9 mg, 84% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86–7.79 (m, 2H), 7.74–7.71 (m, 1H), 7.69 (td, J = 7.0, 1.3 Hz, 1H), 7.65 (td, J = 7.0, 1.6 Hz, 1H), 7.53–7.44 (m, 4H), 7.06 (d, J = 8.4 Hz, 2H), 7.04–6.95 (m, 5H), 6.91 (d, J = 8.4 Hz, 2H), 5.56 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.11 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.2, 195.9, 167.6, 141.1, 140.8, 136.3, 135.8, 135.6, 135.2, 133.7, 130.5,

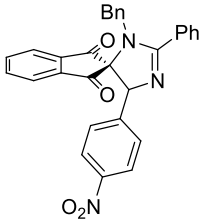
130.4, 129.5 (2C), 129.3 (2C), 128.8 (4C), 128.2 (2C), 128.1 (2C), 128.0, 123.1, 123.0, 79.8, 78.9, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{35}ClN_2O_2$ $[M + H]^+$ 477.1370, found 477.1369.



3-Benzyl-5-(4-bromophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ea). General procedure was followed to afford **3ea** as a yellow solid (89.8 mg, 86% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86–7.79 (m, 2H), 7.74–7.71 (m, 1H), 7.69 (td, J = 6.9, 1.2 Hz, 1H), 7.66 (td, J = 6.9, 1.8 Hz, 1H), 7.53–7.45 (m, 4H), 7.21 (d, J = 8.4 Hz, 2H), 7.03–6.95 (m, 5H), 6.86 (d, J = 8.4 Hz, 2H), 5.55 (s, 1H), 4.57 (d, J = 14.8 Hz, 1H), 4.11 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.2, 195.9, 167.6, 141.1, 140.8, 136.3, 136.1, 135.8, 135.2, 131.1 (2C), 130.5, 130.3, 129.6 (2C), 129.5 (2C), 128.8 (4C), 128.1 (2C), 128.0, 123.08, 123.06, 122.0, 79.7, 78.9, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{79}BrN_2O_2$ $[M + H]^+$ 521.0865, found 521.0861.

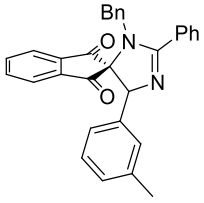


3-Benzyl-2-phenyl-5-(4-(trifluoromethyl)phenyl)-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3fa). General procedure was followed to afford **3fa** as a yellow solid (87.6 mg, 86% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87–7.81 (m, 2H), 7.74 (d, J = 7.2 Hz, 1H), 7.70 (td, J = 7.2, 1.0 Hz, 1H), 7.65 (td, J = 7.2, 1.4 Hz, 1H), 7.55–7.47 (m, 3H), 7.43 (d, J = 7.4 Hz, 1H), 7.35 (d, J = 8.1 Hz, 2H), 7.11 (d, J = 8.1 Hz, 2H), 7.04–6.95 (m, 5H), 5.64 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.12 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.1, 195.8, 167.9, 141.2, 141.1, 140.7, 136.4, 135.9, 135.1, 130.6, 130.3, 130.0 (q, J = 32.3 Hz), 129.5 (2C), 128.83 (2C), 128.79 (2C), 128.3 (2C), 128.13 (2C), 128.07, 124.9 (q, J = 3.6 Hz, 2C), 124.1 (q, J = 272.2 Hz), 123.1, 123.0, 79.6, 78.8, 50.6; ^{19}F NMR (376 MHz, $CDCl_3$) δ -62.53 (s, 3F); HRMS (ESI-TOF) calcd for $C_{31}H_{22}F_3N_2O_2$ $[M + H]^+$ 511.1633, found 511.1635.

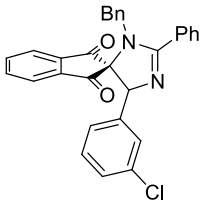


3-Benzyl-5-(4-nitrophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ga). General procedure was followed to afford **3ga** as a yellow solid (83.7 mg, 86% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 (d, J = 8.7 Hz, 2H), 7.88–7.81 (m, 2H), 7.77–7.73 (m, 1H), 7.73

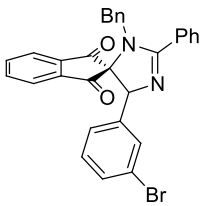
(td, J = 7.5, 1.0 Hz, 1H), 7.67 (td, J = 7.0, 1.9 Hz, 1H), 7.57–7.49 (m, 3H), 7.44 (d, J = 7.4 Hz, 1H), 7.18 (d, J = 8.7 Hz, 2H), 7.03–6.94 (m, 5H), 5.68 (s, 1H), 4.58 (d, J = 14.7 Hz, 1H), 4.13 (d, J = 14.7 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.8, 195.7, 168.1, 147.5, 144.7, 141.0, 140.6, 136.6, 136.1, 134.8, 130.8, 130.0, 129.5 (2C), 128.90 (2C), 128.85 (2C), 128.76 (2C), 128.17, 128.15 (2C), 123.3, 123.2 (2C), 123.1, 79.1, 78.3, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}N_3O_4$ $[M + H]^+$ 488.1610, found 488.1612.



3-Benzyl-2-phenyl-5-*m*-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ha). General procedure was followed to afford **3ha** as a yellow solid (76.6 mg, 84% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88–7.81 (m, 2H), 7.74 (d, J = 7.5 Hz, 1H), 7.67 (td, J = 7.4, 1.0 Hz, 1H), 7.62 (td, J = 7.4, 1.2 Hz, 1H), 7.53–7.45 (m, 3H), 7.41 (d, J = 7.4 Hz, 1H), 7.07–6.96 (m, 5H), 6.94 (t, J = 7.4 Hz, 1H), 6.89 (d, J = 7.6 Hz, 1H), 6.78 (s, 1H), 6.70 (d, J = 7.3 Hz, 1H), 5.57 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.12 (d, J = 14.8 Hz, 1H), 2.12 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.5, 196.0, 167.2, 141.2, 141.0, 137.5, 136.8, 136.0, 135.5, 135.4, 130.6, 130.4, 129.4 (2C), 128.9 (2C), 128.7 (2C), 128.6, 128.5, 128.1 (2C), 127.9, 127.7, 124.9, 123.0, 122.8, 80.4, 79.8, 50.6, 21.3; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1919.

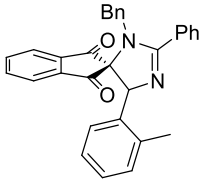


3-Benzyl-5-(3-chlorophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ia). General procedure was followed to afford **3ia** as a yellow solid (85.4 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87–7.81 (m, 2H), 7.76–7.73 (m, 1H), 7.70 (td, J = 7.1, 1.2 Hz, 1H), 7.66 (td, J = 7.1, 1.5 Hz, 1H), 7.54–7.48 (m, 3H), 7.48–7.44 (m, 1H), 7.09 (ddd, J = 7.9, 1.9, 1.1 Hz, 1H), 7.04–6.95 (m, 7H), 6.82 (d, J = 7.6 Hz, 1H), 5.56 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.12 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.1, 195.7, 167.7, 141.0, 140.8, 139.1, 136.3, 135.8, 135.2, 134.1, 130.6, 130.3, 129.5 (2C), 129.2, 128.82 (2C), 128.80 (2C), 128.2, 128.12 (2C), 128.08, 128.02, 126.0, 123.1, 123.0, 79.8, 78.8, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{35}ClN_2O_2$ $[M + H]^+$ 477.1370, found 477.1377.

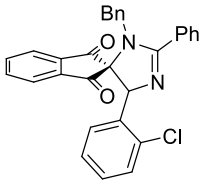


3-Benzyl-5-(3-bromophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ja). General procedure was followed to afford **3ja** as a yellow solid (97.3 mg, 93% yield); ^{1}H

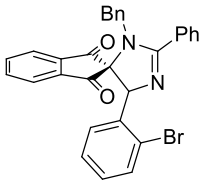
NMR (400 MHz, $CDCl_3$) δ 7.87–7.80 (m, 2H), 7.75 (d, J = 7.0 Hz, 1H), 7.70 (td, J = 7.1, 0.9 Hz, 1H), 7.66 (td, J = 7.1, 1.3 Hz, 1H), 7.54–7.48 (m, 3H), 7.46 (d, J = 7.1 Hz, 1H), 7.24 (d, J = 7.8 Hz, 1H), 7.10 (s, 1H), 7.05–6.97 (m, 5H), 6.95 (t, J = 7.8 Hz, 1H), 6.88 (d, J = 7.7 Hz, 1H), 5.54 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.12 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.1, 195.7, 167.7, 141.0, 140.8, 139.4, 136.3, 135.8, 135.2, 131.05, 130.99, 130.6, 130.3, 129.5 (3C), 128.83 (2C), 128.80 (2C), 128.1 (2C), 128.0, 126.5, 123.2, 123.0, 122.2, 79.8, 78.8, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{79}BrN_2O_2$ $[M + H]^+$ 521.0865, found 521.0867.



3-Benzyl-2-phenyl-5-o-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ka). General procedure was followed to afford **3ka** as a yellow solid (79.5 mg, 87% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88–7.81 (m, 2H), 7.73–7.68 (m, 1H), 7.65 (td, J = 7.2, 1.2 Hz, 1H), 7.60 (td, J = 7.2, 1.3 Hz, 1H), 7.52–7.46 (m, 3H), 7.45 (dd, J = 7.7, 0.8 Hz, 1H), 7.43–7.38 (m, 1H), 7.19 (t, J = 7.5 Hz, 1H), 7.07–6.95 (m, 6H), 6.74 (d, J = 7.5 Hz, 1H), 5.86 (s, 1H), 4.59 (d, J = 14.9 Hz, 1H), 4.15 (d, J = 14.9 Hz, 1H), 1.67 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.7, 195.9, 167.0, 141.3, 140.2, 136.2, 135.5 (2C), 135.0, 134.9, 130.6, 130.4, 130.2, 129.7, 129.4 (2C), 128.8 (2C), 128.7 (2C), 128.1 (2C), 127.9, 127.7, 126.1, 122.8, 122.6, 79.8, 76.0, 50.5, 19.2; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1914.

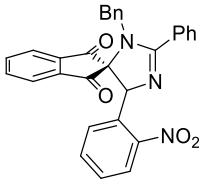


3-Benzyl-5-(2-chlorophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3la). General procedure was followed to afford **3la** as a yellow solid (85.2 mg, 89% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88–7.81 (m, 2H), 7.72 (d, J = 7.4 Hz, 1H), 7.66 (td, J = 7.4, 1.1 Hz, 1H), 7.64–7.58 (m, 2H), 7.53–7.46 (m, 3H), 7.41 (d, J = 7.5 Hz, 1H), 7.30 (td, J = 7.6, 0.9 Hz, 1H), 7.09 (td, J = 7.6, 1.6 Hz, 1H), 7.04–6.93 (m, 6H), 6.07 (s, 1H), 4.54 (d, J = 15.0 Hz, 1H), 4.16 (d, J = 15.0 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.1, 196.0, 167.5, 142.1, 140.1, 135.8, 135.7, 135.24, 135.23, 132.8, 131.6, 130.5, 130.4, 129.4 (2C), 129.1, 128.84 (2C), 128.78 (2C), 128.5, 128.0 (2C), 127.9, 127.0, 122.9, 122.6, 78.3, 75.1, 50.4; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{35}ClN_2O_2$ $[M + H]^+$ 477.1370, found 477.1376.

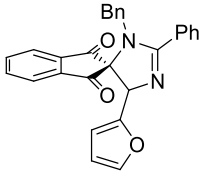


3-Benzyl-5-(2-bromophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione

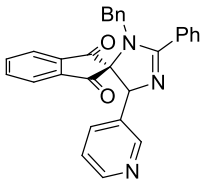
(3ma). General procedure was followed to afford **3ma** as a yellow solid (93.4 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88–7.81 (m, 2H), 7.74 (d, J = 7.4 Hz, 1H), 7.66 (td, J = 7.4, 1.1 Hz, 1H), 7.61 (td, J = 7.4, 1.2 Hz, 1H), 7.59 (dd, J = 7.8, 1.7 Hz, 1H), 7.53–7.46 (m, 3H), 7.41 (d, J = 7.4 Hz, 1H), 7.35 (td, J = 7.6, 1.0 Hz, 1H), 7.16 (dd, J = 8.0, 1.1 Hz, 1H), 7.05–6.93 (m, 6H), 6.07 (s, 1H), 4.54 (d, J = 15.0 Hz, 1H), 4.18 (d, J = 15.0 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.1, 195.9, 167.5, 142.3, 140.2, 136.8, 135.8, 135.7, 135.2, 132.0, 131.9, 130.5, 130.4, 129.44, 129.38 (2C), 128.84 (2C), 128.78 (2C), 128.0 (2C), 127.9, 127.5, 123.4, 123.0, 122.7, 78.2, 77.3, 50.4; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{79}BrN_2O_2$ $[M + H]^+$ 521.0865, found 521.0869.



3-Benzyl-5-(2-nitrophenyl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3na). General procedure was followed to afford **3na** as a yellow solid (88.9 mg, 91% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.89 (dd, J = 8.0, 1.3 Hz, 1H), 7.88–7.83 (m, 2H), 7.82–7.75 (m, 2H), 7.72 (td, J = 7.7, 1.2 Hz, 1H), 7.67 (td, J = 7.5, 0.9 Hz, 1H), 7.56 (td, J = 7.5, 1.0 Hz, 1H), 7.54–7.47 (m, 3H), 7.43–7.34 (m, 2H), 6.98–6.89 (m, 5H), 6.15 (s, 1H), 4.46 (d, J = 15.3 Hz, 1H), 4.25 (d, J = 15.3 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.6, 196.0, 167.6, 147.1, 142.0, 140.5, 135.9, 135.5, 135.0, 134.1, 133.6, 132.7, 130.8, 130.1, 129.2 (2C), 128.9 (3C), 128.8 (2C), 128.1 (2C), 128.0, 124.2, 123.1, 122.7, 76.3, 73.4, 50.3; HRMS (ESI-TOF) calcd for $C_{30}H_{22}N_3O_4$ $[M + H]^+$ 488.1610, found 488.1619.

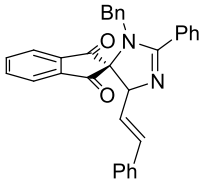


3-Benzyl-5-(furan-2-yl)-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3oa). General procedure was followed to afford **3oa** as a yellow solid (67.6 mg, 78% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.83–7.77 (m, 2H), 7.77–7.68 (m, 3H), 7.66–7.60 (m, 1H), 7.52–7.44 (m, 3H), 7.05–6.94 (m, 6H), 6.30 (d, J = 3.2 Hz, 1H), 6.22 (dd, J = 3.2, 1.8 Hz, 1H), 5.61 (s, 1H), 4.53 (d, J = 14.9 Hz, 1H), 4.15 (d, J = 14.9 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.8, 195.4, 167.7, 151.1, 142.1, 141.0, 140.9, 136.1, 135.6, 135.2, 130.5, 130.3, 129.4 (2C), 128.9 (2C), 128.7 (2C), 128.1 (2C), 128.0, 123.3, 122.8, 110.6, 108.9, 77.9, 73.4, 50.4; HRMS (ESI-TOF) calcd for $C_{28}H_{21}N_2O_3$ $[M + H]^+$ 433.1552, found 433.1558.

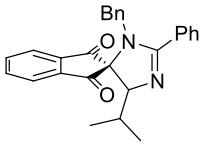


3-Benzyl-2-phenyl-5-(pyridin-3-yl)-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3pa).

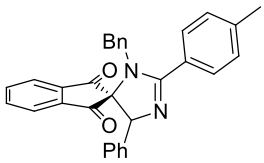
General procedure was followed to afford **3pa** as a yellow oil (79.2 mg, 89% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.36 (d, J = 4.4 Hz, 1H), 8.00 (s, 1H), 7.87–7.80 (m, 2H), 7.74 (d, J = 7.4 Hz, 1H), 7.70 (t, J = 7.3 Hz, 1H), 7.64 (t, J = 7.2 Hz, 1H), 7.59 (d, J = 7.8 Hz, 1H), 7.55–7.47 (m, 3H), 7.43 (d, J = 7.4 Hz, 1H), 7.16 (dd, J = 7.7, 4.9 Hz, 1H), 7.05–6.93 (m, 5H), 5.60 (s, 1H), 4.58 (d, J = 14.8 Hz, 1H), 4.14 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.8, 195.9, 168.0, 149.3, 148.8, 141.0, 140.6, 136.5, 136.14, 136.05, 135.0, 132.9, 130.6, 130.2, 129.5 (2C), 128.83 (2C), 128.76 (2C), 128.12 (2C), 128.08, 123.3, 123.2, 123.0, 79.4, 76.8, 50.6; HRMS (ESI-TOF) calcd for $C_{29}H_{22}N_3O_2$ $[M + H]^+$ 444.1712, found 444.1714.



(*E*)-3-Benzyl-2-phenyl-5-styryl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3qa). General procedure by extending the reaction time to 30 min was followed to afford **3qa** as a yellow solid (68.5 mg, 73% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.83–7.76 (m, 2H), 7.76–7.65 (m, 4H), 7.51–7.43 (m, 3H), 7.24–7.12 (m, 5H), 7.08–6.95 (m, 5H), 6.32 (d, J = 15.8 Hz, 1H), 6.11 (dd, J = 15.8, 8.6 Hz, 1H), 5.06 (d, J = 8.6 Hz, 1H), 4.51 (d, J = 14.8 Hz, 1H), 4.23 (d, J = 14.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.0, 196.3, 166.7, 141.02, 140.99, 136.3, 136.2, 135.8, 135.3, 133.9, 130.42, 130.38, 129.5 (2C), 128.8 (2C), 128.7 (2C), 128.4 (2C), 128.1 (2C), 128.0, 127.9, 126.7 (2C), 125.8, 123.2 (2C), 78.7, 77.9, 50.4; HRMS (ESI-TOF) calcd for $C_{32}H_{25}N_2O_2$ $[M + H]^+$ 469.1916, found 469.1916.

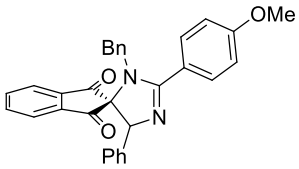


3-Benzyl-5-isopropyl-2-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ra). General procedure was followed to afford **3ra** as a yellow solid (74.2 mg, 91% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.84 (d, J = 7.5 Hz, 1H), 7.76 (td, J = 7.3, 0.9 Hz, 1H), 7.73–7.66 (m, 3H), 7.64 (d, J = 7.5 Hz, 1H), 7.46–7.39 (m, 3H), 7.01–6.89 (m, 5H), 4.42 (d, J = 15.2 Hz, 1H), 4.21 (d, J = 8.5 Hz, 1H), 4.00 (d, J = 15.2 Hz, 1H), 2.01–1.88 (m, 1H), 1.16 (d, J = 6.6 Hz, 3H), 0.52 (d, J = 6.6 Hz, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 200.2, 197.5, 164.7, 141.5, 140.6, 136.0, 135.9, 135.6, 130.6, 130.1, 129.1 (2C), 128.7 (2C), 128.6 (2C), 127.9 (2C), 127.7, 123.3, 122.6, 83.0, 76.8, 50.2, 30.6, 21.3, 20.3; HRMS (ESI-TOF) calcd for $C_{27}H_{25}N_2O_2$ $[M + H]^+$ 409.1916, found 409.1922.

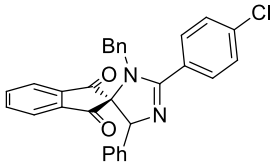


3-Benzyl-5-phenyl-2-*p*-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ab). General procedure was followed to afford **3ab** as a yellow solid (76.2 mg, 83% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (d, J = 8.0 Hz, 2H), 7.72 (d, J = 7.3 Hz, 1H), 7.66 (t, J = 7.3 Hz, 1H),

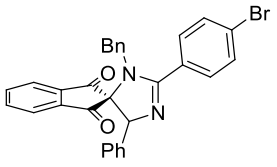
7.60 (t, J = 7.4 Hz, 1H), 7.39 (d, J = 7.5 Hz, 1H), 7.29 (d, J = 8.0 Hz, 2H), 7.12–6.93 (m, 10H), 5.58 (s, 1H), 4.61 (d, J = 14.9 Hz, 1H), 4.12 (d, J = 14.9 Hz, 1H), 2.42 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.5, 196.1, 167.4, 141.1, 140.9, 140.5, 137.0, 136.1, 135.6, 135.5, 129.41 (2C), 129.38 (2C), 128.8 (2C), 128.1 (2C), 128.0 (2C), 127.92 (2C), 127.87, 127.81, 127.6, 123.0, 122.8, 80.2, 79.8, 50.6, 21.6; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1915.



3-Benzyl-2-(4-methoxyphenyl)-5-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ac). General procedure was followed to afford **3ac** as a yellow solid (70.9 mg, 75% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.79 (d, J = 8.8 Hz, 2H), 7.74 (d, J = 7.5 Hz, 1H), 7.67 (td, J = 7.4, 1.1 Hz, 1H), 7.61 (td, J = 7.4, 1.2 Hz, 1H), 7.39 (d, J = 7.5 Hz, 1H), 7.12–7.03 (m, 5H), 7.03–6.93 (m, 7H), 5.58 (s, 1H), 4.63 (d, J = 15.0 Hz, 1H), 4.15 (d, J = 15.0 Hz, 1H), 3.86 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.5, 196.2, 167.2, 161.3, 141.1, 141.0, 137.1, 136.1, 135.7, 135.5, 130.4 (2C), 129.3 (2C), 128.1 (2C), 127.99 (2C), 127.95 (2C), 127.88, 127.85, 123.0, 122.9, 122.7, 114.1 (2C), 80.3, 79.7, 55.5, 50.7; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_3$ $[M + H]^+$ 473.1865, found 473.1865.

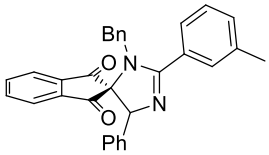


3-Benzyl-2-(4-chlorophenyl)-5-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ad). General procedure was followed to afford **3ad** as a yellow solid (85.9 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (d, J = 8.5 Hz, 2H), 7.74 (d, J = 7.8 Hz, 1H), 7.68 (td, J = 7.4, 1.1 Hz, 1H), 7.62 (td, J = 7.4, 1.3 Hz, 1H), 7.46 (d, J = 8.5 Hz, 2H), 7.40 (d, J = 7.4 Hz, 1H), 7.13–6.98 (m, 8H), 6.97–6.91 (m, 2H), 5.58 (s, 1H), 4.55 (d, J = 14.9 Hz, 1H), 4.10 (d, J = 14.9 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.2, 195.9, 166.4, 141.1, 140.9, 136.6, 136.5, 136.2, 135.6, 135.3, 130.2 (2C), 129.3 (2C), 129.03 (2C), 128.99, 128.2 (2C), 128.00 (3C), 127.95, 127.91 (2C), 123.1, 122.9, 80.5, 79.7, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{35}ClN_2O_2$ $[M + H]^+$ 477.1370, found 477.1371.

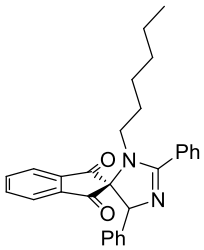


3-Benzyl-2-(4-bromophenyl)-5-phenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ae). General procedure was followed to afford **3ae** as a yellow solid (91.7 mg, 88% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (d, J = 7.4 Hz, 1H), 7.70 (d, J = 8.5 Hz, 2H), 7.68 (td, J = 7.5, 1.1 Hz, 1H), 7.619 (d, J = 8.5 Hz, 2H), 7.616 (td, J = 7.5, 1.3 Hz, 1H), 7.40 (d, J = 7.5 Hz, 1H), 7.13–6.97 (m, 8H), 6.97–6.91 (m, 2H), 5.58 (s, 1H), 4.54 (d, J = 14.9 Hz, 1H), 4.10 (d, J = 14.9 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.2, 195.9, 166.5, 141.1, 140.9, 136.6, 136.2, 135.6, 135.3, 132.0 (2C), 130.4 (2C), 129.5, 129.3 (2C), 128.2 (2C), 128.01, 127.99 (2C), 127.95, 127.90 (2C), 124.8,

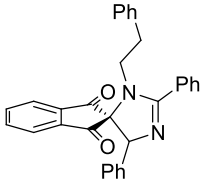
123.0, 122.9, 80.4, 79.7, 50.6; HRMS (ESI-TOF) calcd for $C_{30}H_{22}{}^{79}BrN_2O_2$ $[M + H]^+$ 521.0865, found 521.0863.



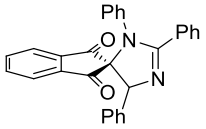
3-Benzyl-5-phenyl-2-m-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3af). General procedure was followed to afford **3af** as a yellow solid (75.3 mg, 82% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (d, J = 7.4 Hz, 1H), 7.69–7.64 (m, 2H), 7.64–7.57 (m, 2H), 7.40 (d, J = 7.7 Hz, 1H), 7.36 (t, J = 7.6 Hz, 1H), 7.30 (d, J = 7.5 Hz, 1H), 7.12–6.93 (m, 10H), 5.59 (s, 1H), 4.59 (d, J = 14.8 Hz, 1H), 4.11 (d, J = 14.8 Hz, 1H), 2.42 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.4, 196.0, 167.6, 141.1, 140.9, 138.6, 136.9, 136.1, 135.6, 135.5, 131.1, 130.4, 129.6, 129.4 (2C), 128.5, 128.1 (2C), 127.93 (2C), 127.91 (2C), 127.87, 127.81, 125.7, 123.0, 122.8, 80.3, 79.7, 50.6, 21.5; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1913.



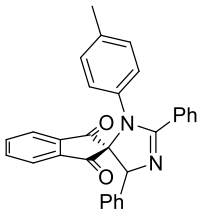
3-Hexyl-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ag). General procedure was followed to afford **3ag** as a yellow oil (69.8 mg, 80% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (d, J = 7.6 Hz, 1H), 7.85 (td, J = 7.5, 0.7 Hz, 1H), 7.79–7.73 (m, 2H), 7.74 (t, J = 7.4 Hz, 1H), 7.54–7.47 (m, 4H), 7.13–7.04 (m, 3H), 6.99–6.92 (m, 2H), 5.56 (s, 1H), 3.29 (ddd, J = 14.5, 9.4, 6.3 Hz, 1H), 2.96 (ddd, J = 14.5, 9.8, 4.8 Hz, 1H), 1.31–0.97 (m, 8H), 0.75 (t, J = 7.2 Hz, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.9, 196.3, 167.7, 141.1, 140.9, 136.9, 136.5, 136.0, 130.8, 130.2, 128.7 (2C), 128.5 (2C), 128.0 (2C), 127.9 (2C), 127.8, 123.5, 123.2, 81.5, 80.3, 46.1, 31.1, 30.2, 26.3, 22.4, 13.9; HRMS (ESI-TOF) calcd for $C_{29}H_{29}N_2O_2$ $[M + H]^+$ 437.2229, found 437.2230.



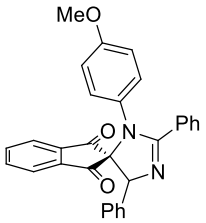
3-Phenethyl-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ah). General procedure was followed to afford **3ah** as a yellow solid (78.5 mg, 86% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (d, J = 7.6 Hz, 1H), 7.85 (td, J = 7.5, 0.9 Hz, 1H), 7.74 (td, J = 7.5, 0.9 Hz, 1H), 7.67–7.61 (m, 2H), 7.51 (d, J = 7.7 Hz, 1H), 7.49–7.42 (m, 3H), 7.16–7.05 (m, 6H), 6.98–6.93 (m, 2H), 6.93–6.88 (m, 2H), 5.57 (s, 1H), 3.53 (ddd, J = 14.8, 9.9, 6.5 Hz, 1H), 3.21 (ddd, J = 14.8, 9.7, 5.7 Hz, 1H), 2.63 (ddd, J = 13.6, 9.7, 6.5 Hz, 1H), 2.53 (ddd, J = 13.6, 9.9, 5.7 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.8, 196.5, 167.6, 141.3, 141.0, 138.1, 136.8, 136.6, 136.0, 130.6, 130.3, 128.8 (2C), 128.7 (2C), 128.6 (2C), 128.5 (2C), 128.1 (2C), 128.0 (3C), 126.5, 123.6, 123.3, 82.1, 80.5, 47.7, 36.8; HRMS (ESI-TOF) calcd for $C_{31}H_{25}N_2O_2$ $[M + H]^+$ 457.1916, found 457.1920.



2,3,5-Triphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ai). General procedure was followed to afford **3ai** as a yellow solid (76.9 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01 (d, J = 7.6 Hz, 1H), 7.80 (t, J = 7.5 Hz, 1H), 7.69 (t, J = 7.5 Hz, 1H), 7.62 (d, J = 8.0 Hz, 2H), 7.46 (d, J = 7.6 Hz, 1H), 7.37 (t, J = 7.4 Hz, 1H), 7.29 (t, J = 7.2 Hz, 2H), 7.17–6.97 (m, 8H), 6.95 (d, J = 7.8 Hz, 2H), 5.77 (s, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.0, 195.7, 164.5, 141.8, 141.2, 139.6, 136.6 (2C), 136.0, 130.4, 129.9, 129.4 (2C), 129.0 (2C), 128.2 (4C), 128.1 (3C), 127.3 (2C), 126.5, 123.7, 123.4, 83.5, 80.1; HRMS (ESI-TOF) calcd for $C_{29}H_{21}N_2O_2$ $[M + H]^+$ 429.1603, found 429.1601.

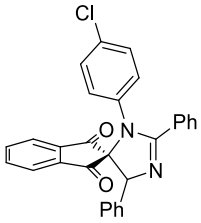


2,5-Diphenyl-3-*p*-tolyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3aj). General procedure was followed to afford **3aj** as a yellow solid (81.4 mg, 92% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00 (d, J = 7.7 Hz, 1H), 7.77 (td, J = 7.5, 0.9 Hz, 1H), 7.66 (td, J = 7.5, 0.9 Hz, 1H), 7.65–7.60 (m, 2H), 7.44 (d, J = 7.6 Hz, 1H), 7.36 (t, J = 7.4 Hz, 1H), 7.27 (t, J = 7.4 Hz, 2H), 7.16–7.08 (m, 3H), 7.06–7.00 (m, 2H), 6.86 (d, J = 8.8 Hz, 2H), 6.84 (d, J = 8.8 Hz, 2H), 5.76 (s, 1H), 2.14 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.2, 195.8, 164.6, 141.7, 141.2, 137.0, 136.7, 136.5, 136.3, 135.9, 130.2, 129.9, 129.6 (2C), 129.3 (2C), 128.11 (2C), 128.08 (2C), 128.02 (2C), 127.99, 127.3 (2C), 123.6, 123.3, 83.4, 79.9, 21.0; HRMS (ESI-TOF) calcd for $C_{30}H_{23}N_2O_2$ $[M + H]^+$ 443.1760, found 443.1755.

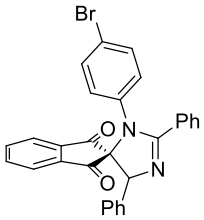


3-(4-Methoxyphenyl)-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3ak). General procedure was followed to afford **3ak** as a yellow solid (82.5 mg, 90% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00 (d, J = 7.7 Hz, 1H), 7.78 (td, J = 7.5, 0.9 Hz, 1H), 7.66 (td, J = 7.5, 1.0 Hz, 1H), 7.64–7.59 (m, 2H), 7.44 (d, J = 7.6 Hz, 1H), 7.35 (t, J = 7.4 Hz, 1H), 7.27 (t, J = 7.4 Hz, 2H), 7.14–7.08 (m, 3H), 7.06–7.01 (m, 2H), 6.98 (d, J = 9.0 Hz, 2H), 6.58 (d, J = 9.0 Hz, 2H), 5.75 (s, 1H), 3.63 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.3, 195.9, 164.8, 158.1, 141.8, 141.3, 136.8, 136.5, 135.9, 132.4, 130.2, 129.8, 129.5 (2C), 129.3 (2C), 128.10 (2C), 128.08 (2C), 128.05 (2C), 127.99, 123.6, 123.3, 114.0 (2C), 83.7, 79.7, 55.3; HRMS (ESI-TOF) calcd for $C_{30}H_{23}N_2O_3$ [M +

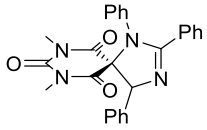
H]$^+$ 459.1709, found 459.1711.



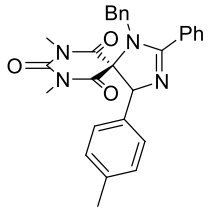
3-(4-Chlorophenyl)-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3al). General procedure was followed to afford **3al** as a yellow solid (82.4 mg, 89% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02 (d, J = 7.7 Hz, 1H), 7.81 (td, J = 7.5, 0.9 Hz, 1H), 7.70 (td, J = 7.5, 0.9 Hz, 1H), 7.64–7.59 (m, 2H), 7.45 (d, J = 7.6 Hz, 1H), 7.40 (t, J = 7.4 Hz, 1H), 7.31 (t, J = 7.4 Hz, 2H), 7.17–7.08 (m, 3H), 7.02 (d, J = 8.8 Hz, 2H), 7.03–6.98 (m, 2H), 6.90 (d, J = 8.8 Hz, 2H), 5.75 (s, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.7, 195.5, 164.2, 141.7, 141.1, 138.3, 136.7, 136.3, 136.1, 132.1, 130.6, 129.5, 129.3 (2C), 129.1 (2C), 128.6 (2C), 128.3 (2C), 128.2, 128.1 (4C), 123.7, 123.5, 83.5, 80.1; HRMS (ESI-TOF) calcd for $C_{29}H_{20}{}^{35}ClN_2O_2$ [M + H]$^+$ 463.1213, found 463.1221.



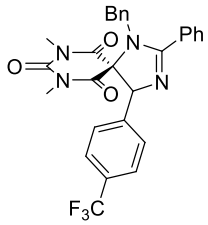
3-(4-Bromophenyl)-2,5-diphenyl-3,5-dihydrospiro[imidazole-4,2'-indene]-1',3'-dione (3am). General procedure was followed to afford **3am** as a yellow solid (95.3 mg, 94% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02 (d, J = 7.7 Hz, 1H), 7.81 (td, J = 7.5, 0.8 Hz, 1H), 7.70 (td, J = 7.5, 0.9 Hz, 1H), 7.64–7.59 (m, 2H), 7.46 (d, J = 7.6 Hz, 1H), 7.40 (t, J = 7.4 Hz, 1H), 7.32 (t, J = 7.5 Hz, 2H), 7.17 (d, J = 8.7 Hz, 2H), 7.15–7.08 (m, 3H), 7.03–6.98 (m, 2H), 6.82 (d, J = 8.7 Hz, 2H), 5.75 (s, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.6, 195.5, 164.1, 141.7, 141.1, 138.8, 136.7, 136.3, 136.1, 132.1 (2C), 130.6, 129.5, 129.3 (2C), 128.8 (2C), 128.4 (2C), 128.2, 128.1 (4C), 123.7, 123.5, 120.1, 83.4, 80.1; HRMS (ESI-TOF) calcd for $C_{29}H_{20}{}^{79}BrN_2O_2$ [M + H]$^+$ 507.0708, found 507.0700.



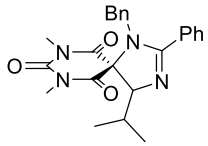
1-Benzyl-7,9-dimethyl-2,4-diphenyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5aa). General procedure was followed to afford **5aa** as a white solid (63.7 mg, 70% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.80–7.73 (m, 2H), 7.51–7.45 (m, 3H), 7.32–7.27 (m, 3H), 7.23–7.12 (m, 7H), 5.61 (s, 1H), 4.55 (d, J = 13.8 Hz, 1H), 4.18 (d, J = 13.8 Hz, 1H), 3.12 (s, 3H), 2.53 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.7, 167.5, 166.0, 149.8, 136.3, 134.5, 130.4, 130.3 (3C), 128.8, 128.7 (4C), 128.4 (2C), 128.32, 128.27 (2C), 127.5 (2C), 83.1, 80.0, 50.8, 29.1, 28.0; HRMS (ESI-TOF) calcd for $C_{27}H_{25}N_4O_3$ [M + H]$^+$ 453.1927, found 453.1930.



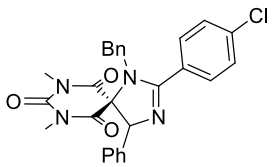
1-Benzyl-7,9-dimethyl-2-phenyl-4-p-tolyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5ba). General procedure was followed to afford **5ba** as a white solid (62.5 mg, 67% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.79–7.72 (m, 2H), 7.50–7.43 (m, 3H), 7.23–7.17 (m, 3H), 7.17–7.12 (m, 2H), 7.09 (d, J = 8.1 Hz, 2H), 7.04 (d, J = 8.1 Hz, 2H), 5.57 (s, 1H), 4.54 (d, J = 13.9 Hz, 1H), 4.18 (d, J = 13.9 Hz, 1H), 3.12 (s, 3H), 2.55 (s, 3H), 2.30 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.7, 167.4, 166.0, 149.9, 138.6, 134.6, 133.2, 130.5, 130.3 (3C), 129.0 (2C), 128.7 (2C), 128.6 (2C), 128.2 (3C), 127.4 (2C), 83.1, 80.1, 50.8, 29.0, 28.0, 21.3; HRMS (ESI-TOF) calcd for $C_{28}H_{27}N_4O_3$ $[M + H]^+$ 467.2083, found 467.2086.



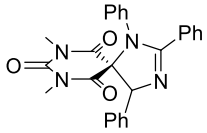
1-Benzyl-7,9-dimethyl-2-phenyl-4-(4-(trifluoromethyl)phenyl)-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5ca). General procedure was followed to afford **5ca** as a white solid (73.7 mg, 71% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81–7.75 (m, 2H), 7.56 (d, J = 8.1 Hz, 2H), 7.53–7.47 (m, 3H), 7.31 (d, J = 8.1 Hz, 2H), 7.25–7.19 (m, 3H), 7.16–7.09 (m, 2H), 5.67 (s, 1H), 4.55 (d, J = 13.7 Hz, 1H), 4.20 (d, J = 13.7 Hz, 1H), 3.11 (s, 3H), 2.54 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.5, 167.9, 165.7, 149.6, 140.6, 134.0, 131.0 (q, J = 32.6 Hz), 130.6, 130.4 (2C), 130.1, 128.8 (2C), 128.7 (2C), 128.6, 128.4 (2C), 128.1 (2C), 125.3 (q, J = 3.6 Hz, 2C), 123.9 (q, J = 272.4 Hz), 82.3, 79.0, 50.8, 29.2, 27.9; ^{19}F NMR (376 MHz, $CDCl_3$) δ -62.66 (s, 3F); HRMS (ESI-TOF) calcd for $C_{28}H_{24}F_3N_4O_3$ $[M + H]^+$ 521.1801, found 521.1800.



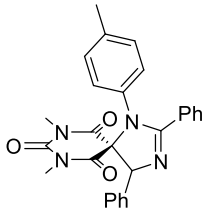
1-Benzyl-4-isopropyl-7,9-dimethyl-2-phenyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5da). General procedure was followed to afford **5da** as a white solid (67.0 mg, 80% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.69–7.63 (m, 2H), 7.47–7.40 (m, 3H), 7.29–7.20 (m, 3H), 7.14–7.09 (m, 2H), 4.42 (d, J = 14.3 Hz, 1H), 4.29 (d, J = 9.7 Hz, 1H), 3.98 (d, J = 14.3 Hz, 1H), 3.22 (s, 3H), 2.94 (s, 3H), 1.99–1.86 (m, 1H), 1.18 (d, J = 6.5 Hz, 3H), 0.71 (d, J = 6.5 Hz, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 170.9, 167.0, 164.2, 150.0, 134.5, 130.6, 130.1, 129.7 (2C), 128.7 (2C), 128.6 (2C), 128.53, 128.46 (2C), 87.8, 74.9, 50.8, 30.5, 29.0, 28.3, 21.2, 20.4; HRMS (ESI-TOF) calcd for $C_{24}H_{27}N_4O_3$ $[M + H]^+$ 419.2083, found 419.2090.



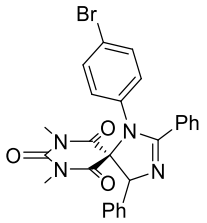
1-Benzyl-2-(4-chlorophenyl)-7,9-dimethyl-4-phenyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5ad). General procedure was followed to afford **5ad** as a white solid (69.1 mg, 71% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 8.4 Hz, 2H), 7.44 (d, J = 8.4 Hz, 2H), 7.31–7.27 (m, 3H), 7.23–7.19 (m, 3H), 7.18–7.12 (m, 4H), 5.59 (s, 1H), 4.51 (d, J = 13.9 Hz, 1H), 4.16 (d, J = 13.9 Hz, 1H), 3.14 (s, 3H), 2.53 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.5, 166.6, 165.9, 149.7, 136.5, 136.0, 134.4, 130.2 (2C), 130.1 (2C), 129.0 (3C), 128.8, 128.5 (2C), 128.4, 128.3 (2C), 127.5 (2C), 83.0, 80.3, 50.9, 29.1, 28.0; HRMS (ESI-TOF) calcd for $C_{27}H_{24}{}^{35}ClN_4O_3$ $[M + H]^+$ 487.1537, found 487.1530.



7,9-Dimethyl-1,2,4-triphenyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5ai). General procedure was followed to afford **5ai** as a white solid (77.6 mg, 89% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.60–7.55 (m, 2H), 7.40–7.33 (m, 4H), 7.31–7.24 (m, 4H), 7.15 (t, J = 7.3 Hz, 2H), 7.09 (t, J = 7.2 Hz, 1H), 7.04–6.99 (m, 2H), 5.78 (s, 1H), 3.41 (s, 3H), 2.54 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.0, 165.8, 164.7, 150.1, 139.2, 136.0, 130.5, 129.7, 129.3 (2C), 129.2, 128.9 (2C), 128.6 (2C), 128.2 (2C), 127.8 (2C), 126.9 (2C), 126.5, 83.7, 83.0, 29.6, 28.3; HRMS (ESI-TOF) calcd for $C_{26}H_{23}N_4O_3$ $[M + H]^+$ 439.1770, found 439.1770.

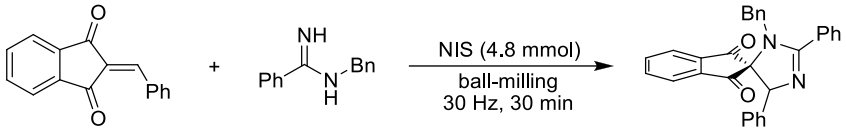


7,9-Dimethyl-2,4-diphenyl-1-*p*-tolyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5aj). General procedure was followed to afford **5aj** as a white solid (79.8 mg, 88% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.60–7.55 (m, 2H), 7.39–7.32 (m, 4H), 7.31–7.24 (m, 4H), 6.94 (s, 4H), 5.76 (s, 1H), 3.40 (s, 3H), 2.52 (s, 3H), 2.23 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 169.2, 165.8, 164.9, 150.1, 136.6, 136.5, 136.1, 130.4, 129.8, 129.5 (2C), 129.3 (2C), 129.2, 128.6 (2C), 128.1 (2C), 127.8 (2C), 127.1 (2C), 83.7, 83.2, 29.5, 28.3, 21.1; HRMS (ESI-TOF) calcd for $C_{27}H_{25}N_4O_3$ $[M + H]^+$ 453.1927, found 453.1933.



1-(4-Bromophenyl)-7,9-dimethyl-2,4-diphenyl-1,3,7,9-tetraazaspiro[4.5]dec-2-ene-6,8,10-trione (5am). General procedure was followed to afford **5am** as a white solid (95.3 mg, 92% yield); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59–7.54 (m, 2H), 7.39 (t, J = 7.4 Hz, 1H), 7.37–7.33 (m, 3H), 7.31 (t, J = 7.5 Hz, 2H), 7.28–7.22 (m, 4H), 6.92 (d, J = 8.8 Hz, 2H), 5.75 (s, 1H), 3.41 (s, 3H), 2.52 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 168.8, 165.6, 164.4, 149.9, 138.3, 135.7, 132.0 (2C), 130.7, 129.3 (2C), 129.2 (2C), 128.7 (2C), 128.6 (2C), 128.3 (2C), 127.7 (2C), 120.1, 83.8, 83.0, 29.6, 28.3; HRMS (ESI-TOF) calcd for $C_{26}H_{22}{}^{79}BrN_4O_3$ $[M + H]^+$ 517.0875, found 517.0875.

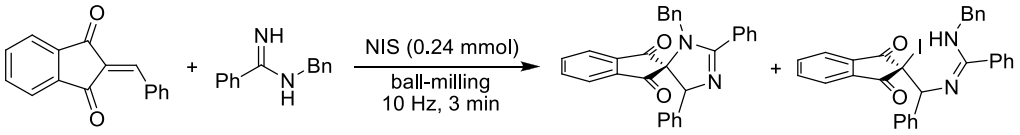
3. Typical procedure for the gram-scale synthesis of 3aa



1a (4 mmol) **2a** (6 mmol) **3aa** (1.51 g, 85%)

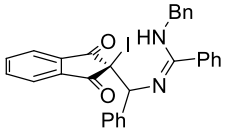
A mixture of **1a** (468 mg, 2 mmol), **2a** (631 mg, 3 mmol) and NIS (540 mg, 2.4 mmol) together with a stainless ball (12 mm in diameter) were introduced into a stainless steel jar (10 mL). The same mixture was also introduced into another parallel jar. The two reaction vessels were closed and fixed on the vibration arms of a Retsch MM400 mixer mill, and were vibrated vigorously at a rate of 1800 rounds per minute (30 Hz) at room temperature for 30 min. After completion of the reaction, the reaction vessels were washed with acetone three times (3 × 6 mL), and the combined solution was evaporated to remove the solvent in vacuo. The residue was separated by flash column chromatography on silica gel with acetone/petroleum ether (1:6) as the eluent to afford **3aa** as a yellow solid (1.51 g, 85%).

4. Synthesis and characterization of 6aa



1a (0.2 mmol) **2a** (0.3 mmol) **3aa**, 58% **6aa**, 28%

A mixture of **1a** (0.2 mmol), **2a** (0.3 mmol) and NIS (0.24 mmol) together with a stainless ball (10 mm in diameter) were introduced into a stainless steel jar (10 mL). The reaction vessel along with another identical empty vessel were closed and fixed on the vibration arms of a Retsch MM400 mixer mill, and were vibrated at a rate of 600 rounds per minute (10 Hz) at room temperature for 3 min. After completion of the reaction, the reaction vessel was washed with acetone three times (3 × 6 mL), and the combined solution was evaporated to remove the solvent in vacuo. The residue was separated by flash column chromatography on silica gel with acetone/petroleum ether (1:6) as the eluent to afford **6aa** as a yellow solid (31.8 mg, 28%), along with **3aa** (51.0 mg, 58%) and recovered **1a** (4.6 mg, 10%).



***N*-Benzyl-*N′*-((2-iodo-1,3-dioxo-2,3-dihydro-1*H*-inden-2-yl)(phenyl)methyl)benzimidamide (6aa)**. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00 (d, J = 7.6 Hz, 1H), 7.84–7.80 (m, 2H), 7.78 (td, J = 7.5, 1.3 Hz, 1H), 7.71 (td, J = 7.3, 1.0 Hz, 1H), 7.67 (d, J = 7.4 Hz, 1H), 7.51–7.45 (m, 3H), 7.33–7.21 (m, 6H), 7.17–7.12 (m, 2H), 7.02 (d, J = 6.6 Hz, 2H), 5.11 (s, 1H), 4.88 (d, J = 15.8 Hz, 1H), 4.05 (d, J = 15.8 Hz, 1H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 199.7, 196.4, 170.9, 142.5, 141.5, 136.2, 135.8, 135.3, 134.3, 130.8, 130.3, 128.9 (2C), 128.85 (2C), 128.80 (2C), 128.6 (2C), 128.4, 128.05 (2C), 128.02 (2C), 127.9, 123.9, 123.8, 81.3, 69.4, 49.5; HRMS (ESI-TOF) calcd for $C_{30}H_{23}N_2O_2$ [M – I]$^+$ 443.1754, found 443.1753.

5. NMR spectra of 3, 5 and 6aa

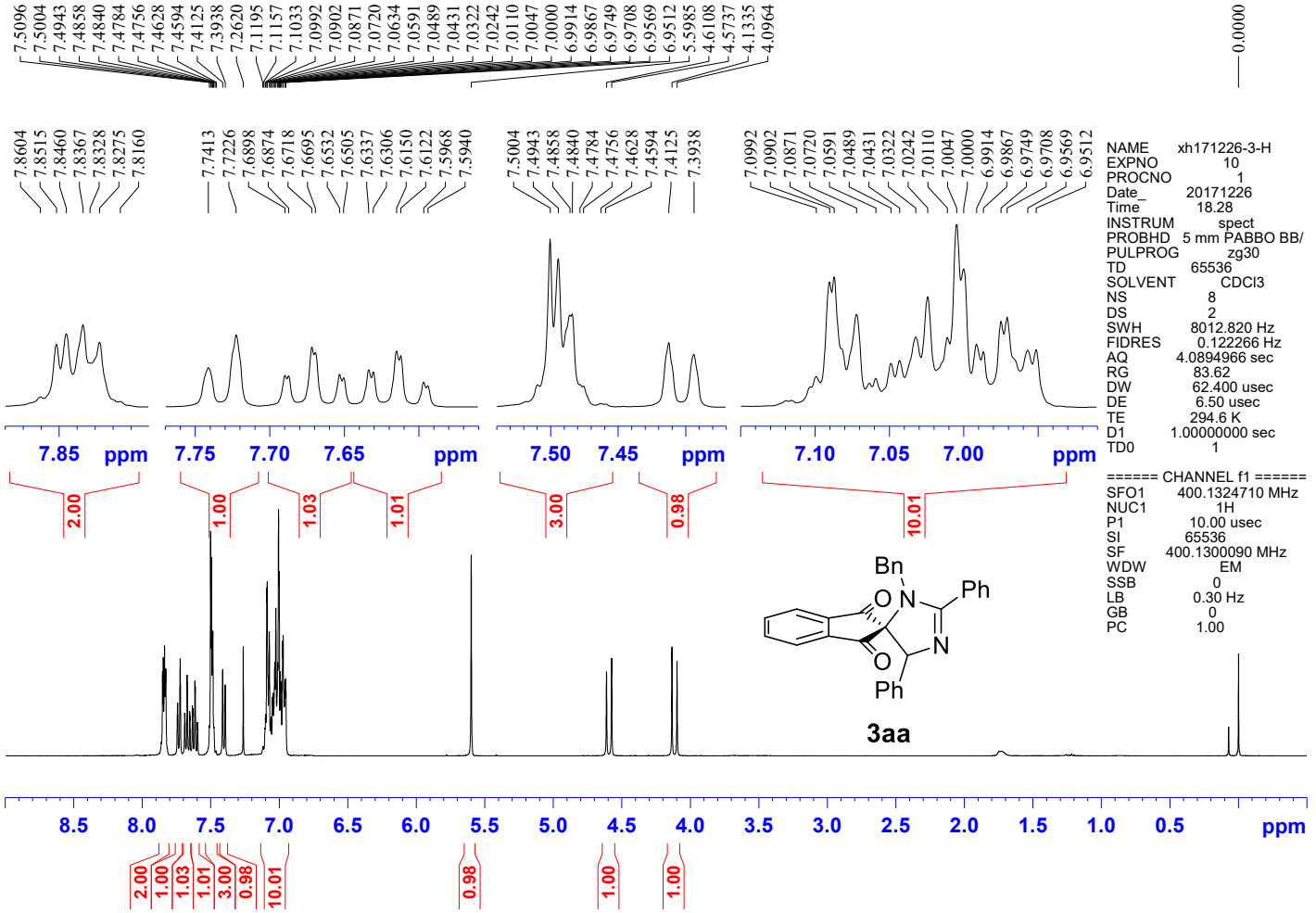


Figure S1. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3aa

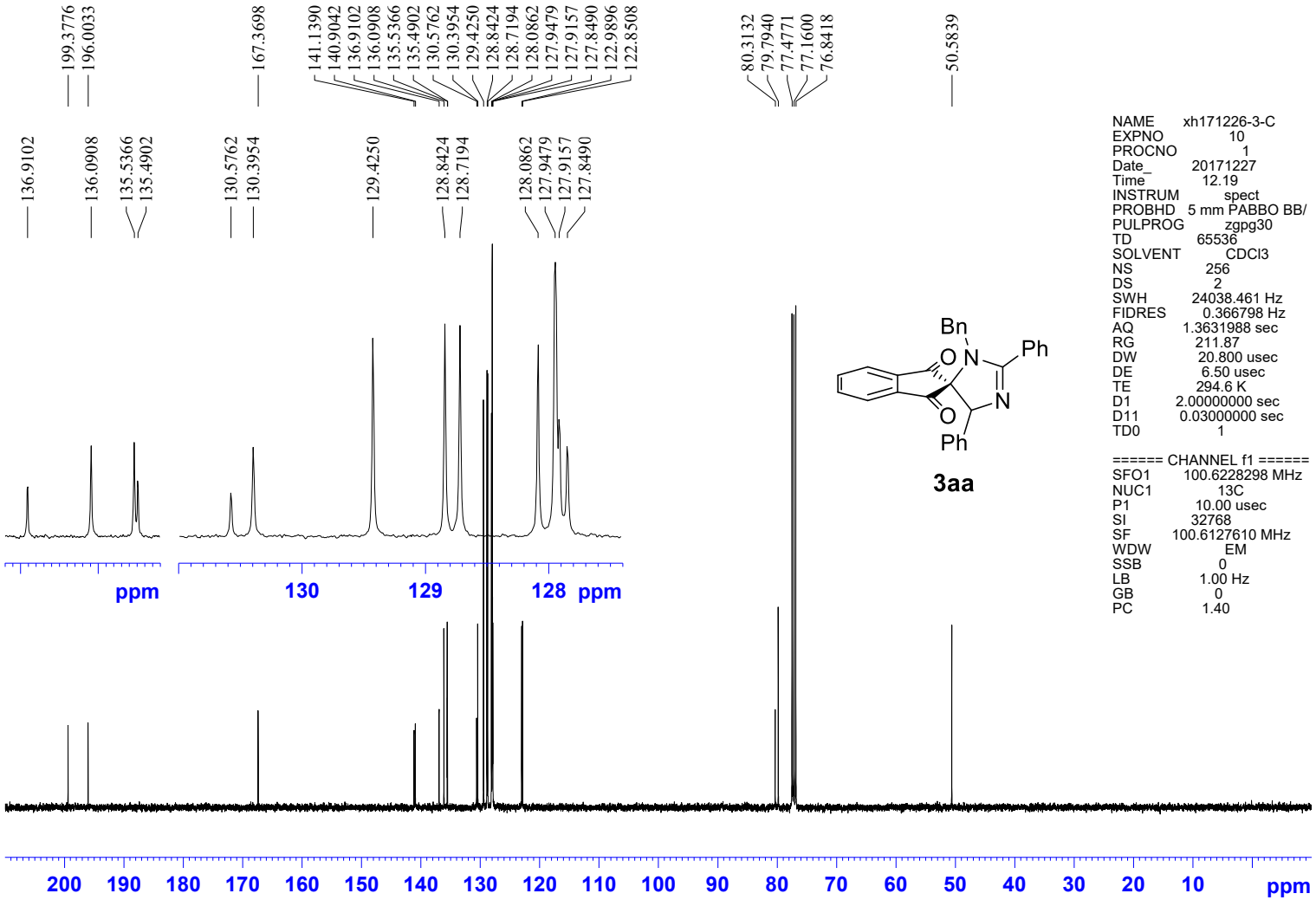


Figure S2. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3aa

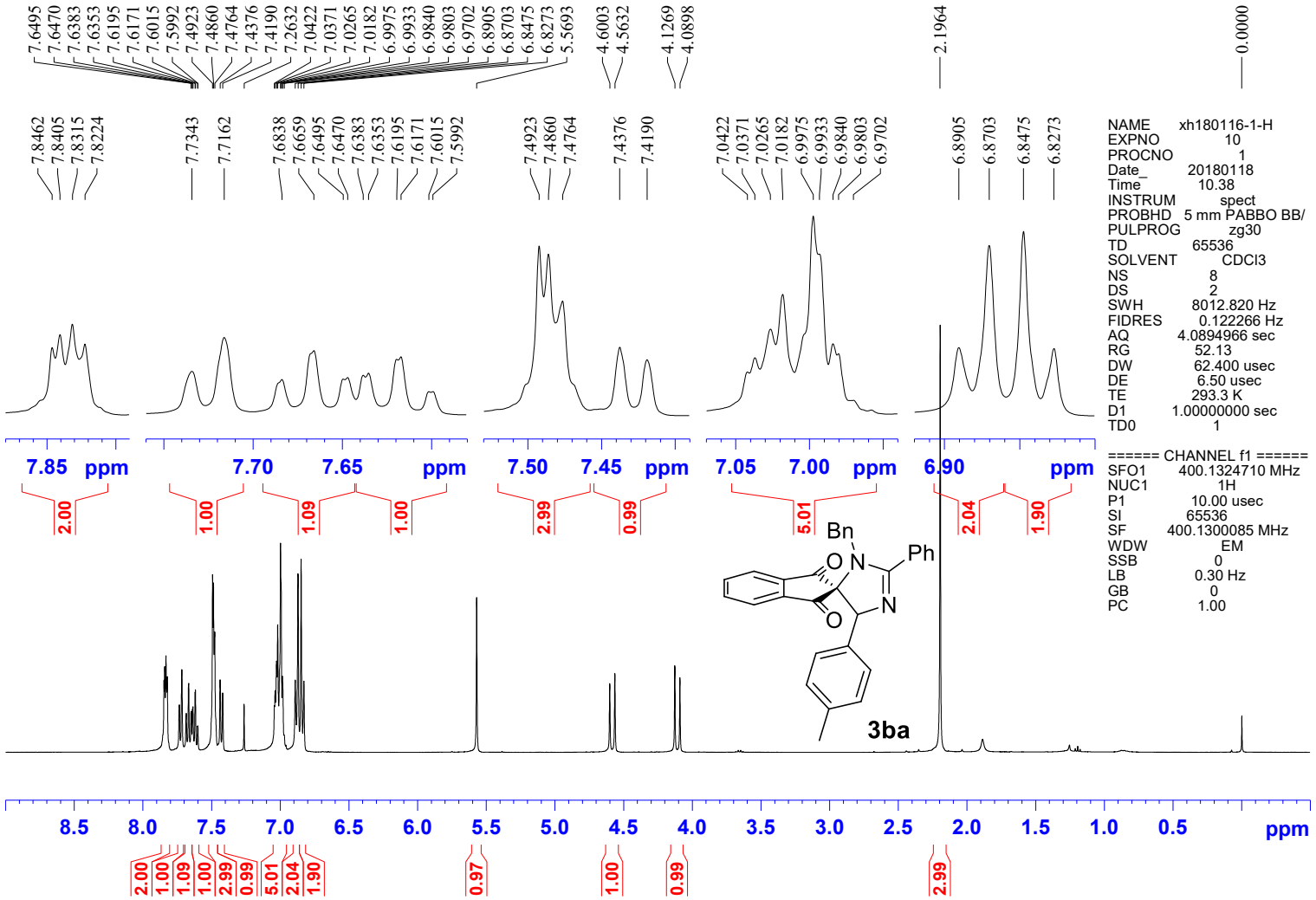


Figure S3. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ba

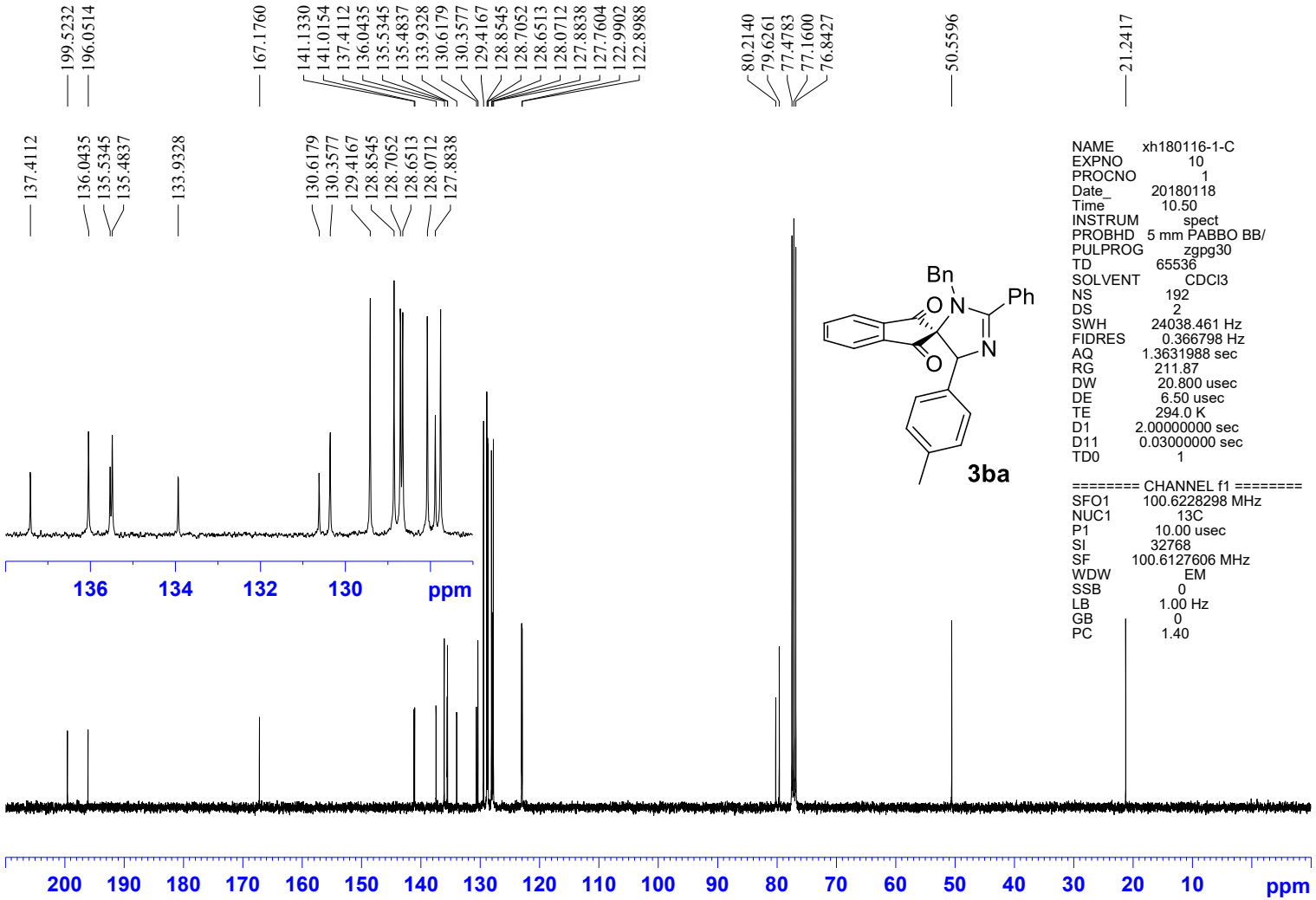


Figure S4. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ba

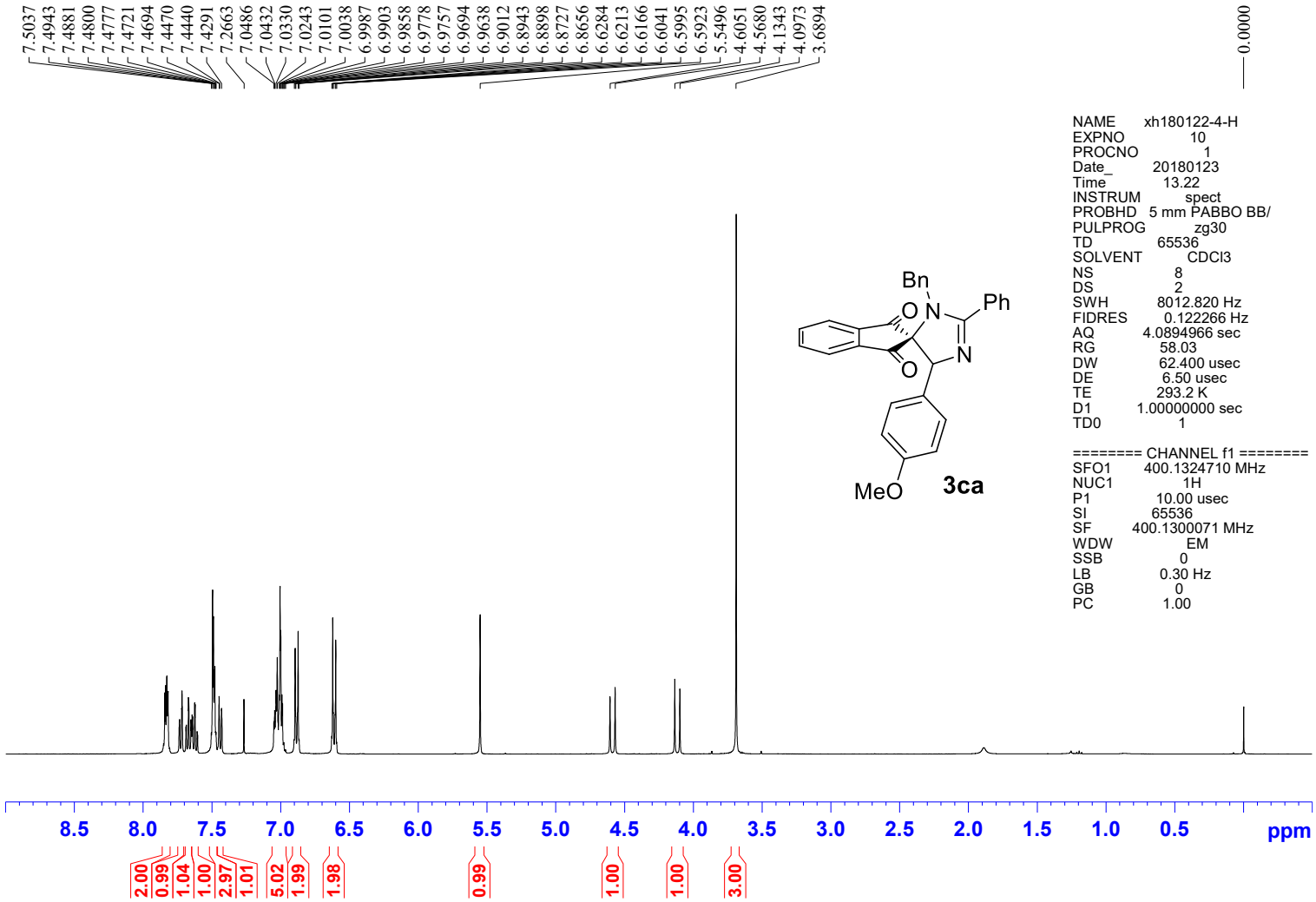


Figure S5. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ca

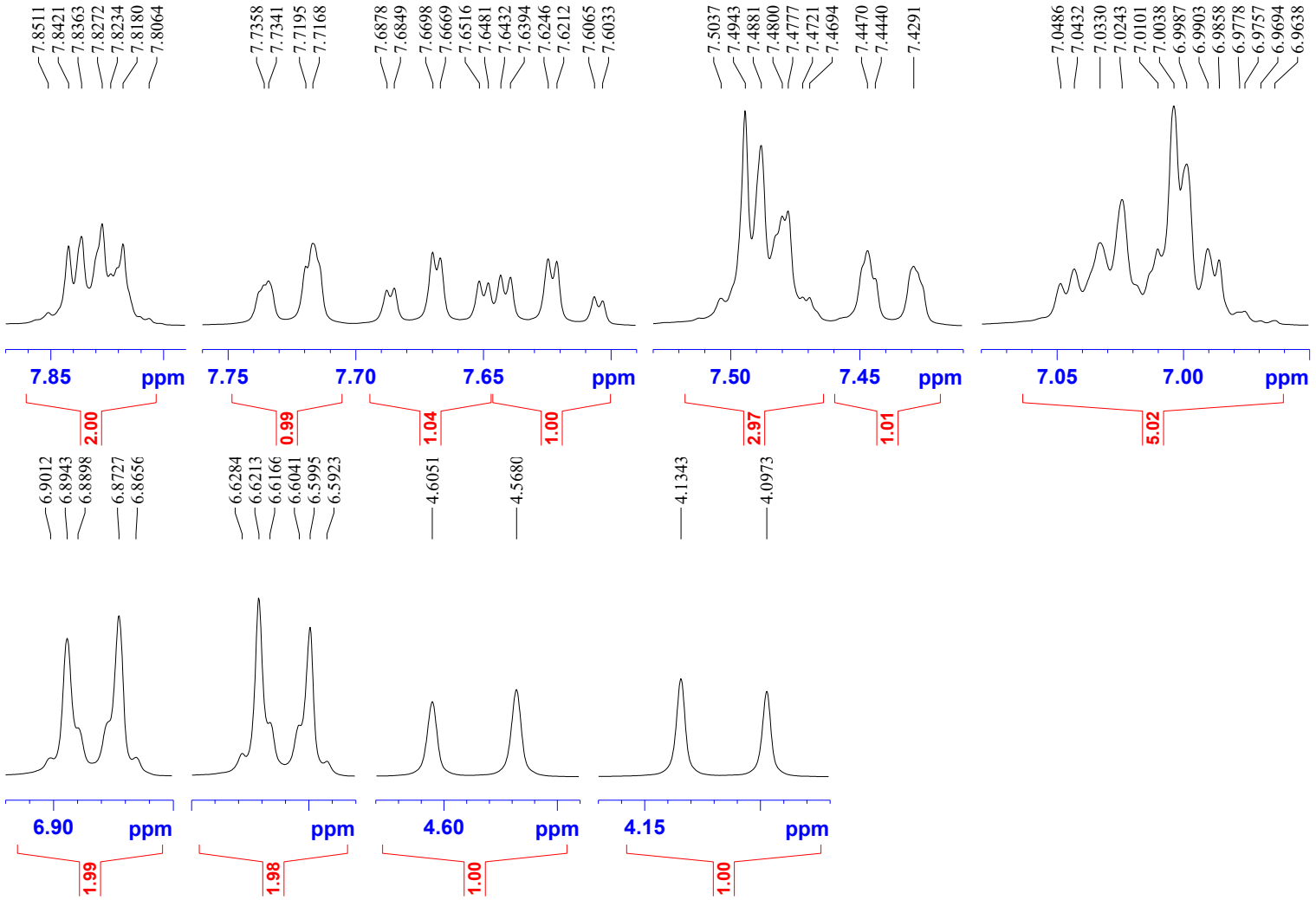


Figure S6. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ca

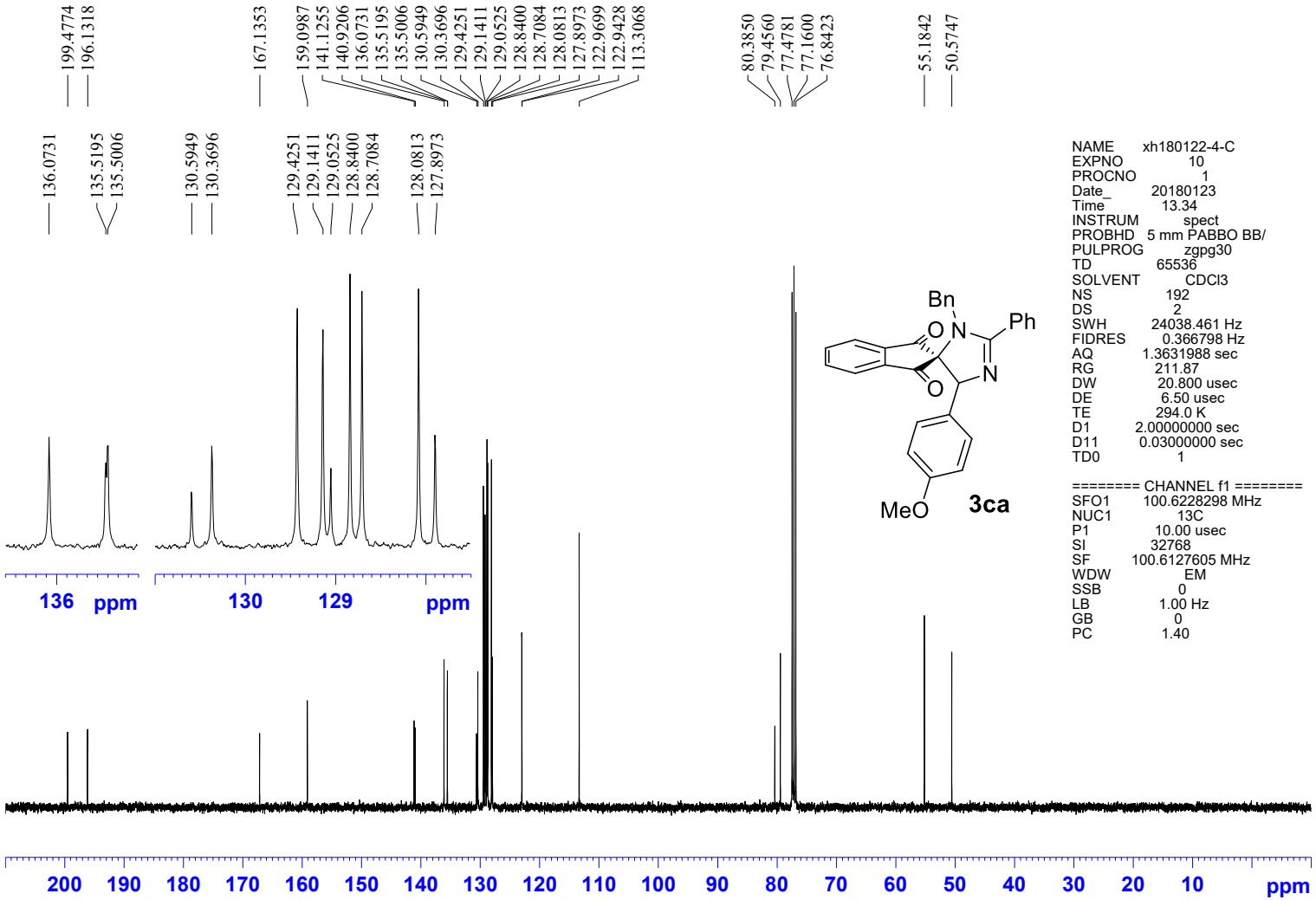


Figure S7. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ca

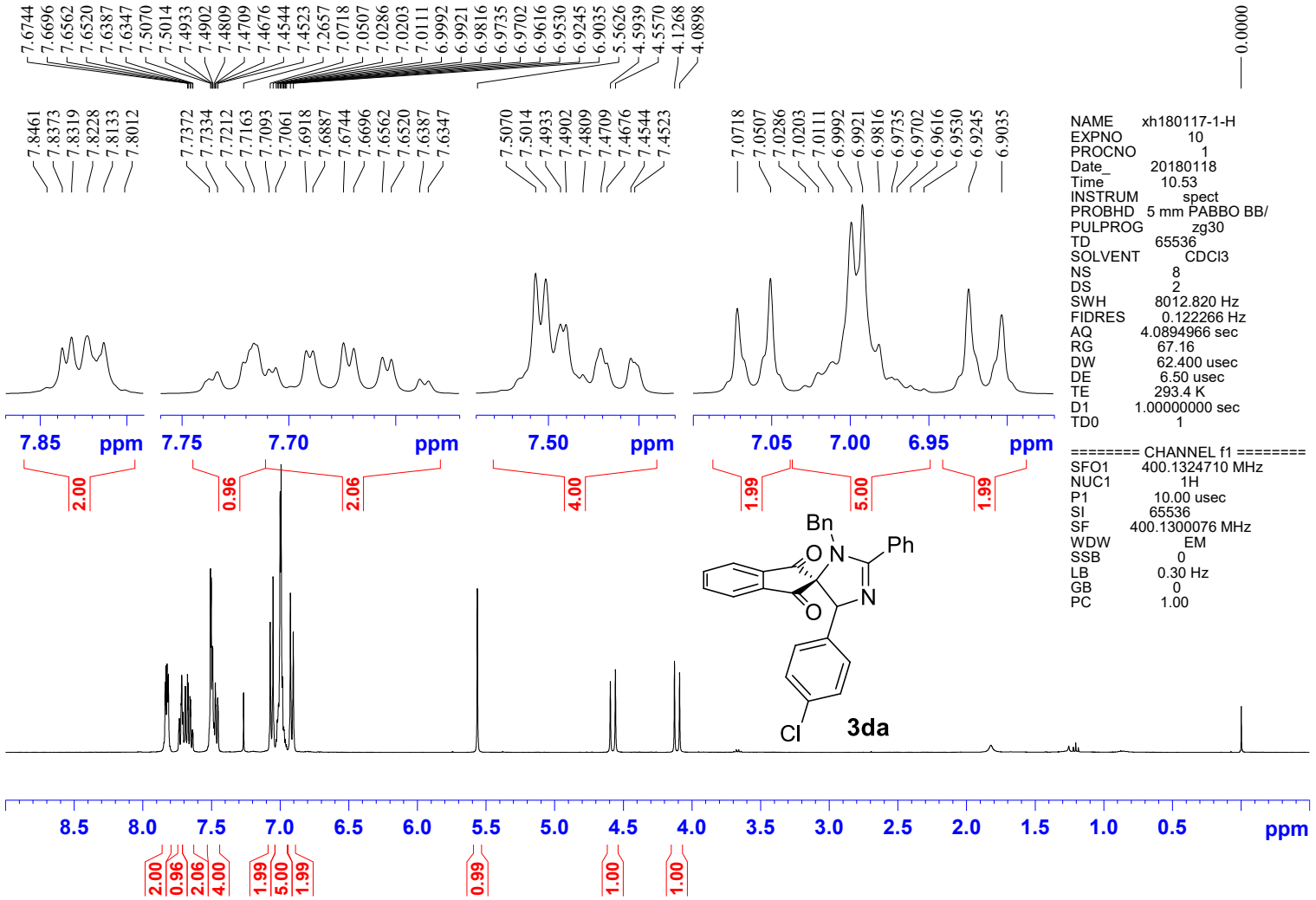


Figure S8. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3da

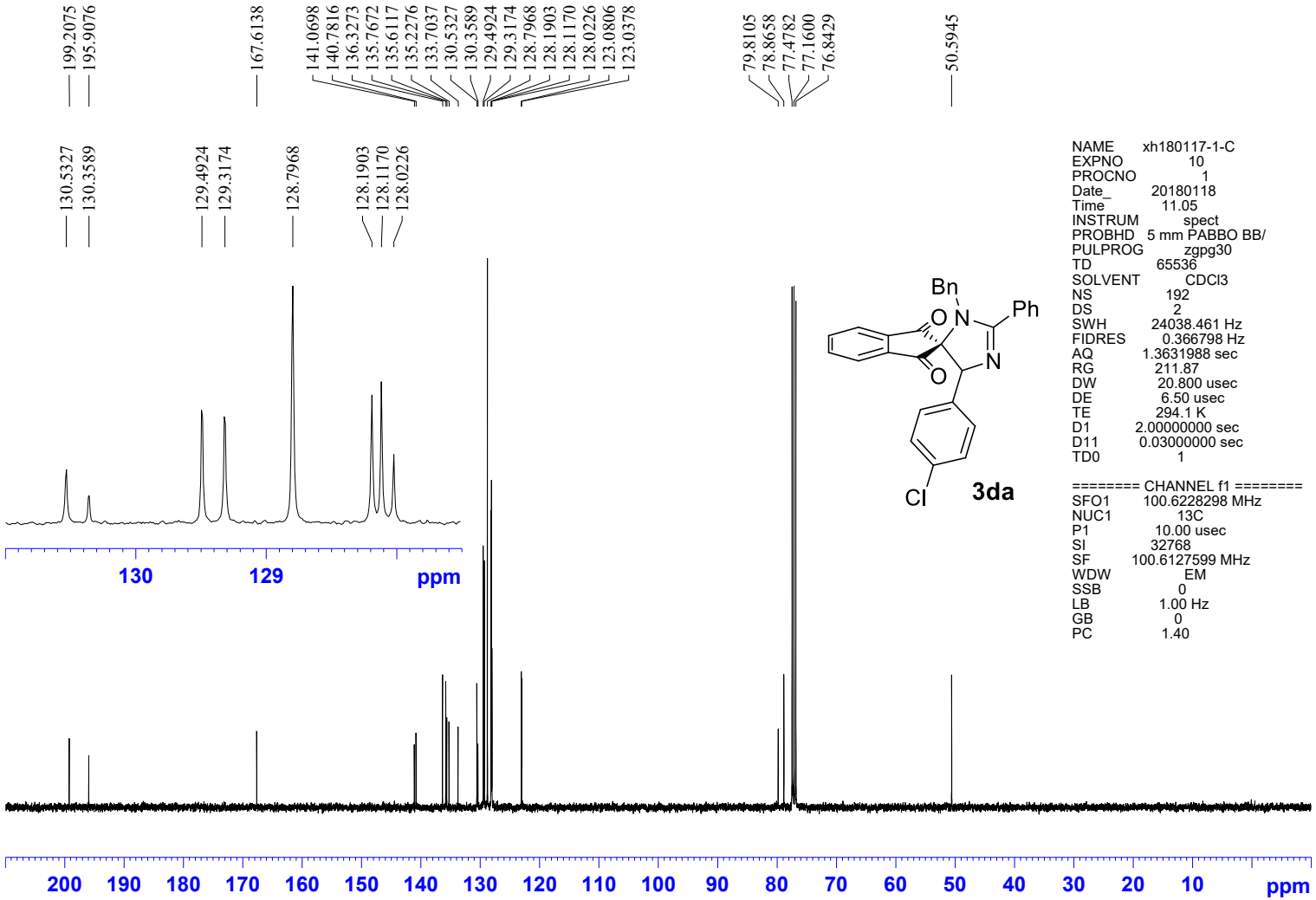


Figure S9. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3da

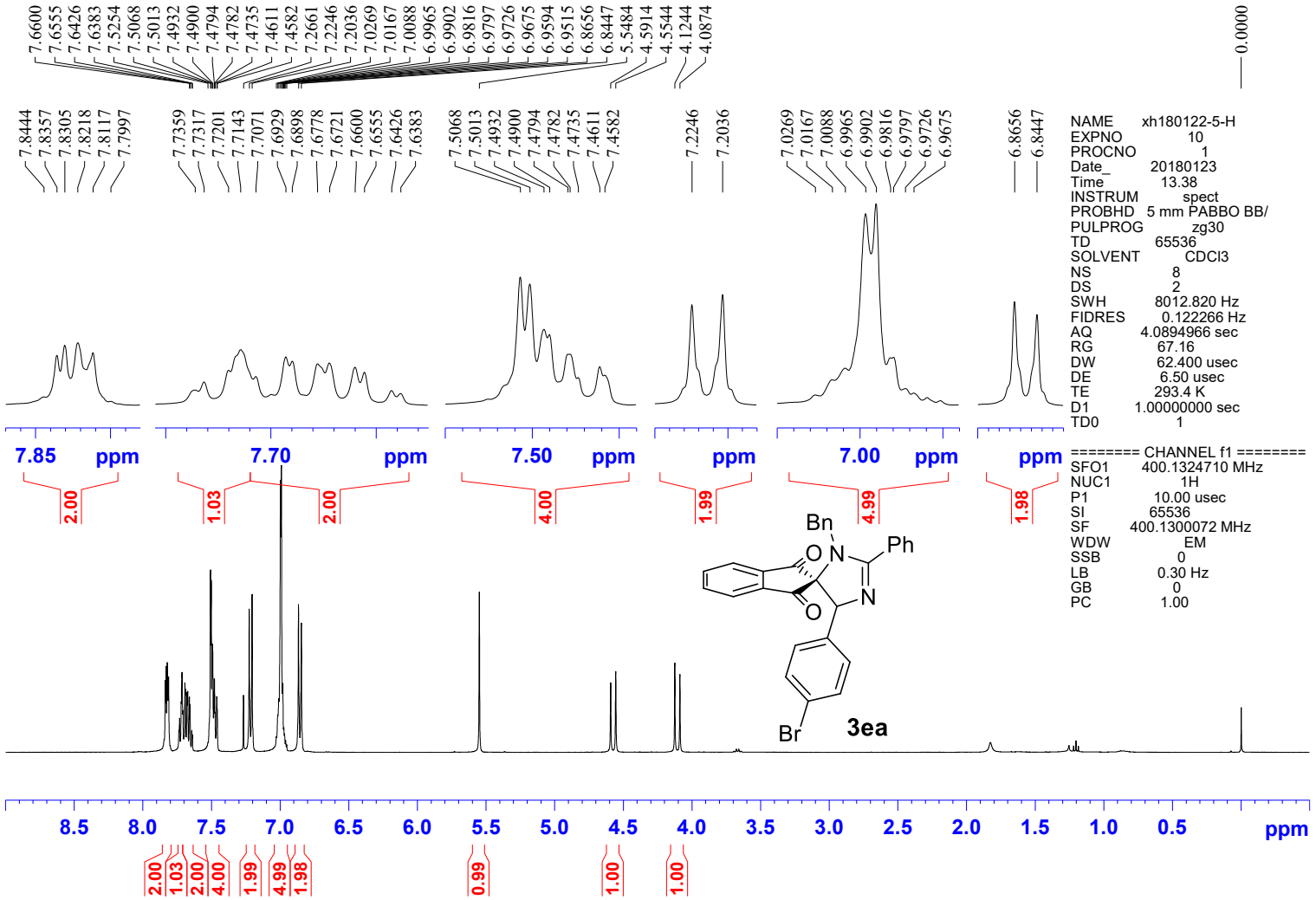


Figure S10. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ea

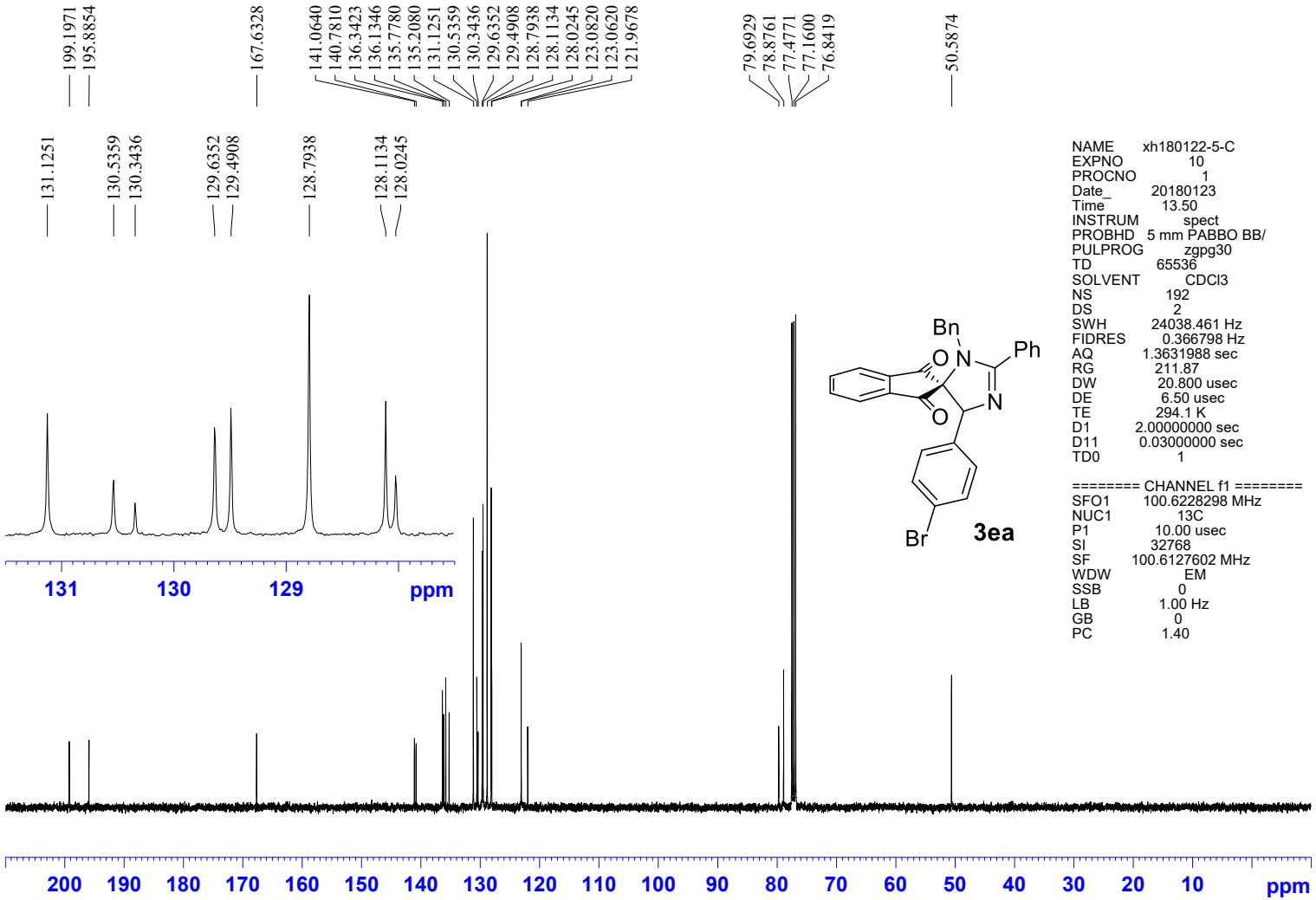


Figure S11. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ea

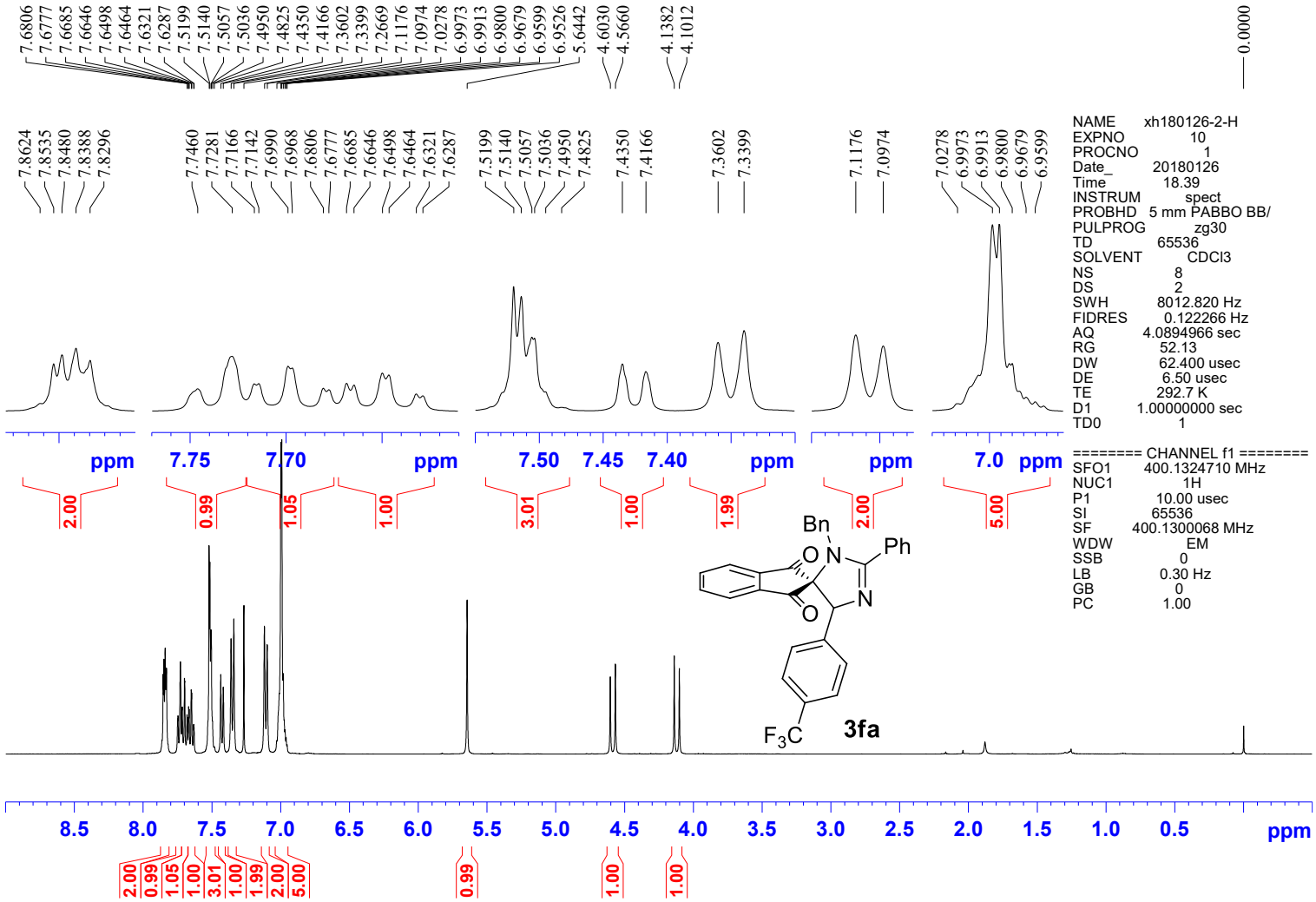


Figure S12. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3fa

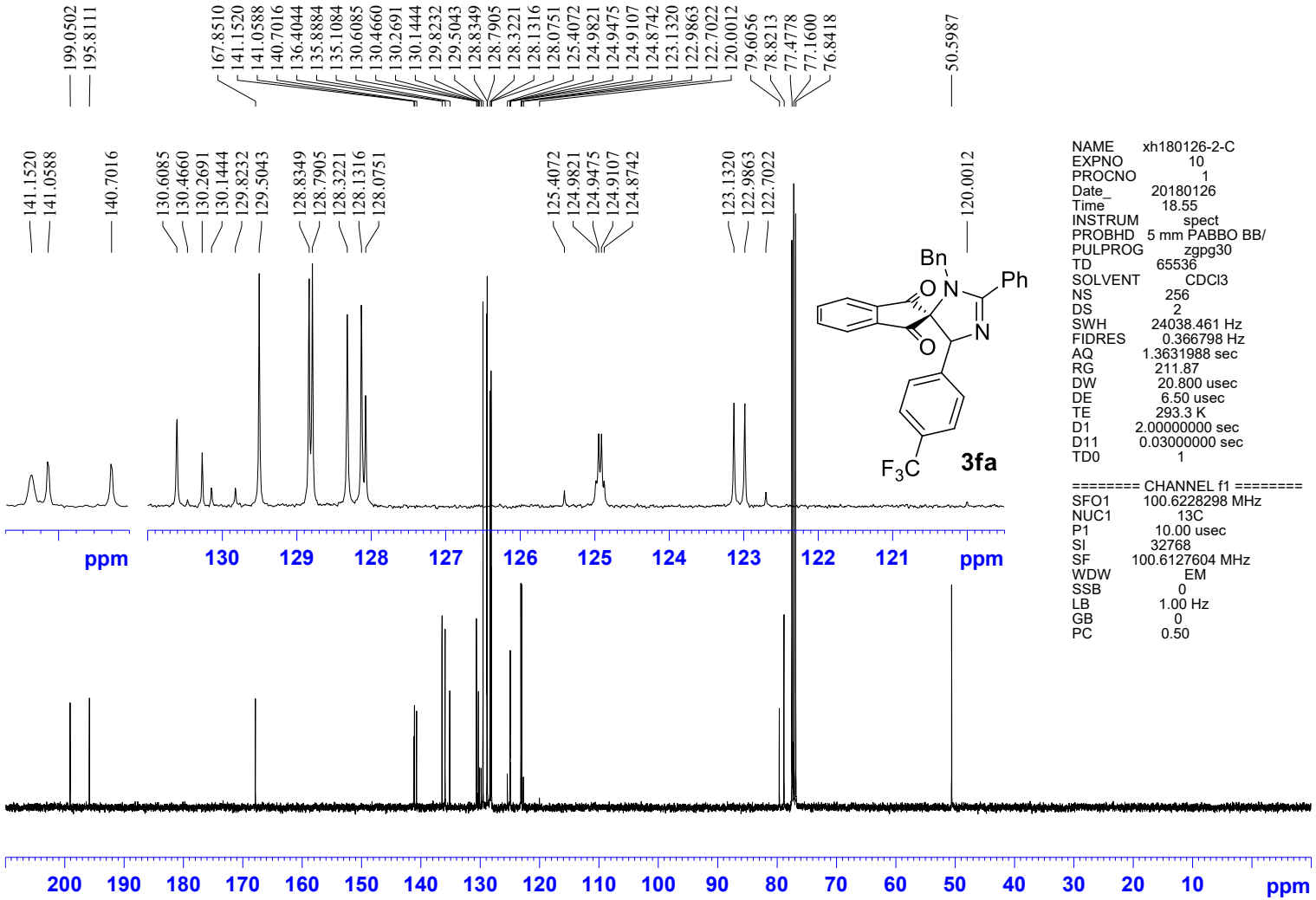


Figure S13. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3fa

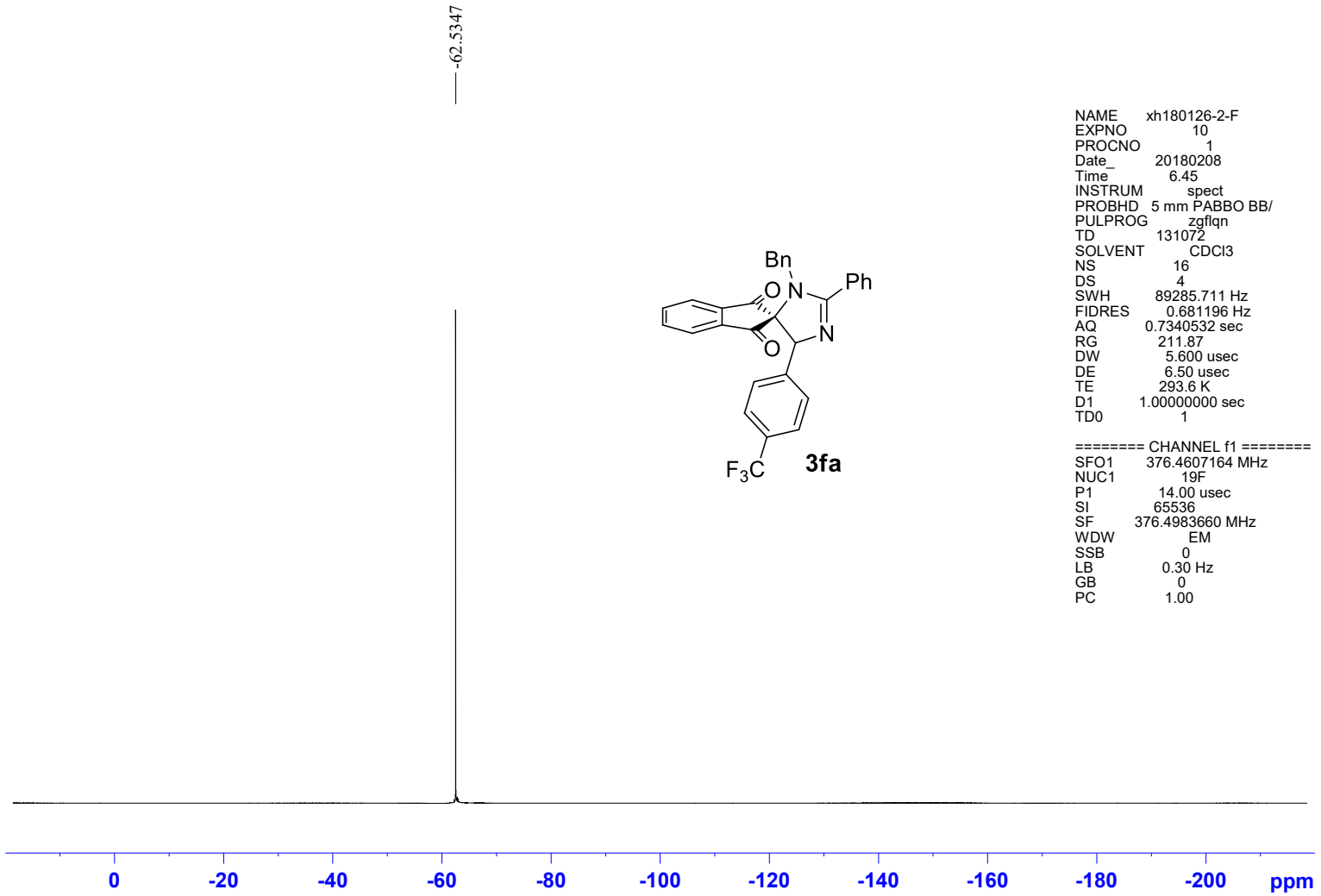


Figure S14. ^{19}F NMR (376 MHz, $CDCl_3$) of compound 3fa

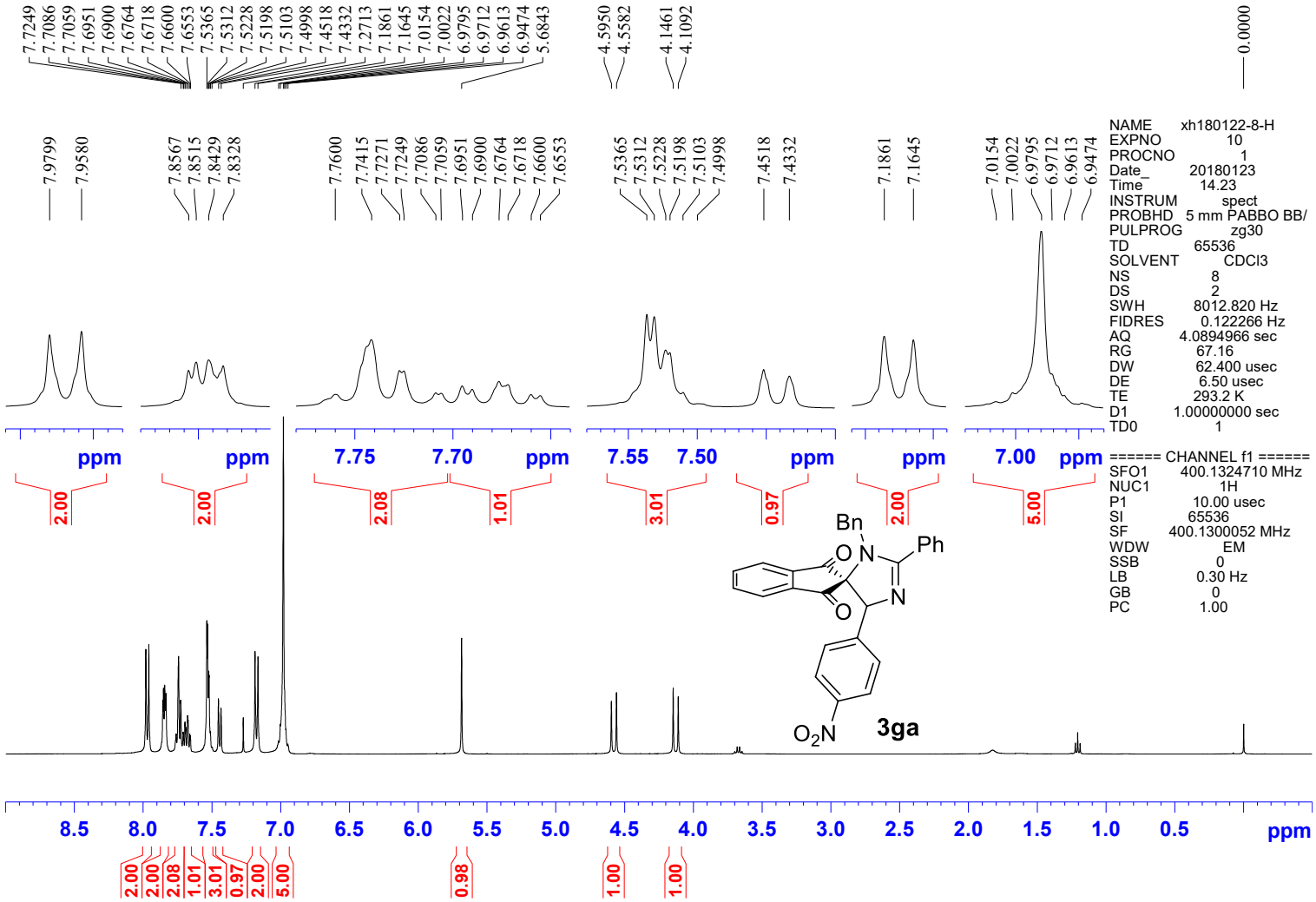


Figure S15. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ga

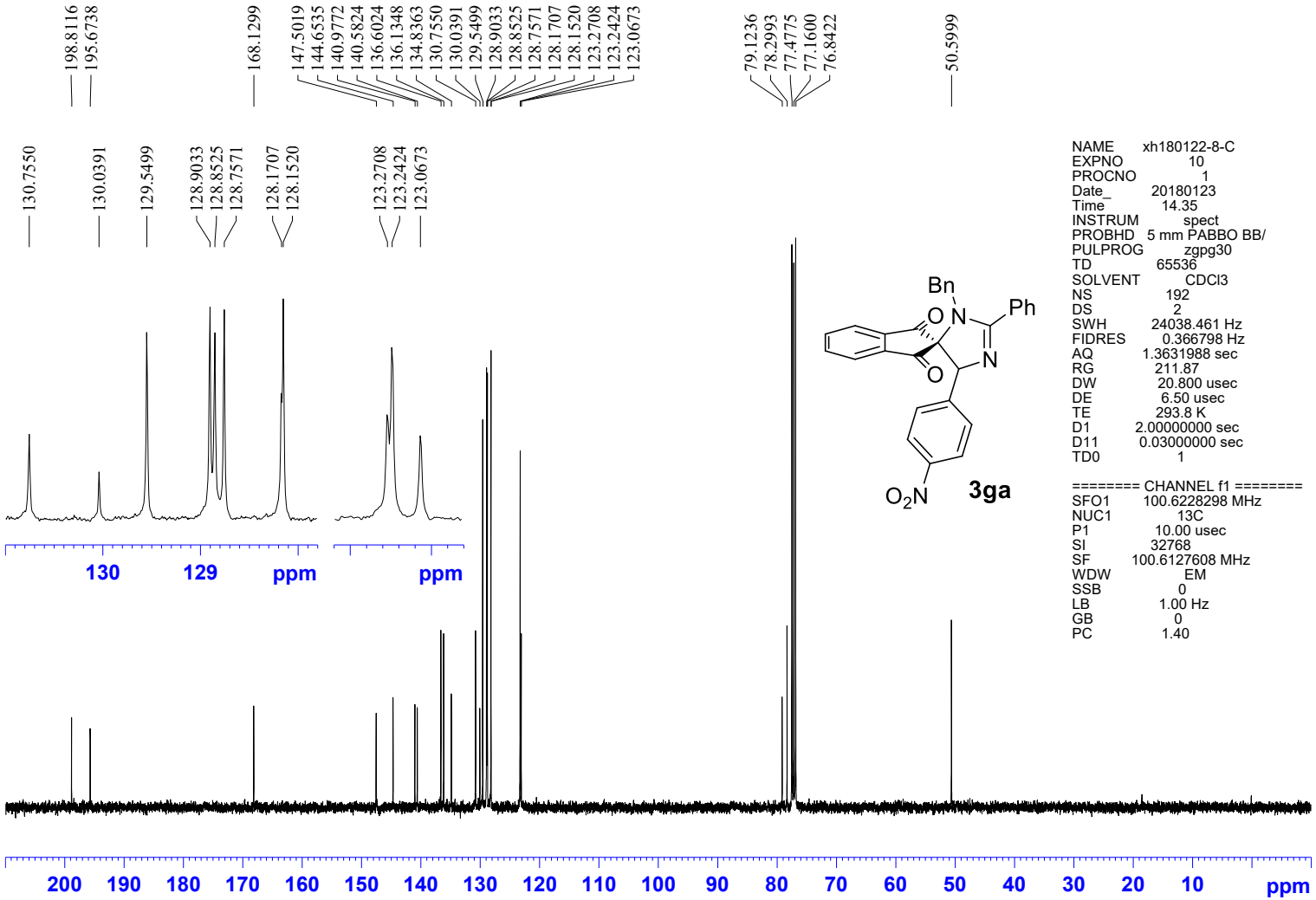


Figure S16. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ga

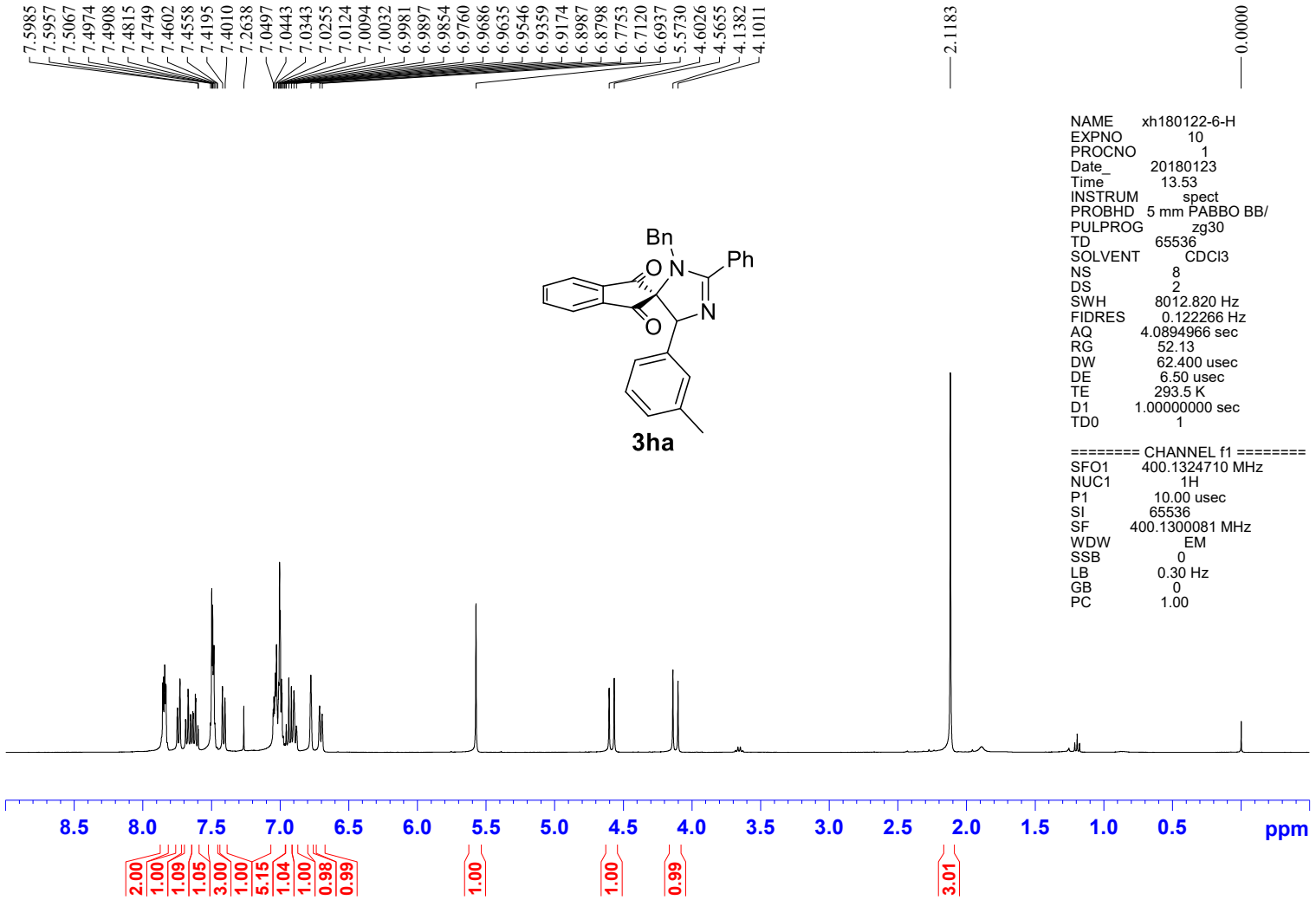


Figure S17. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ha

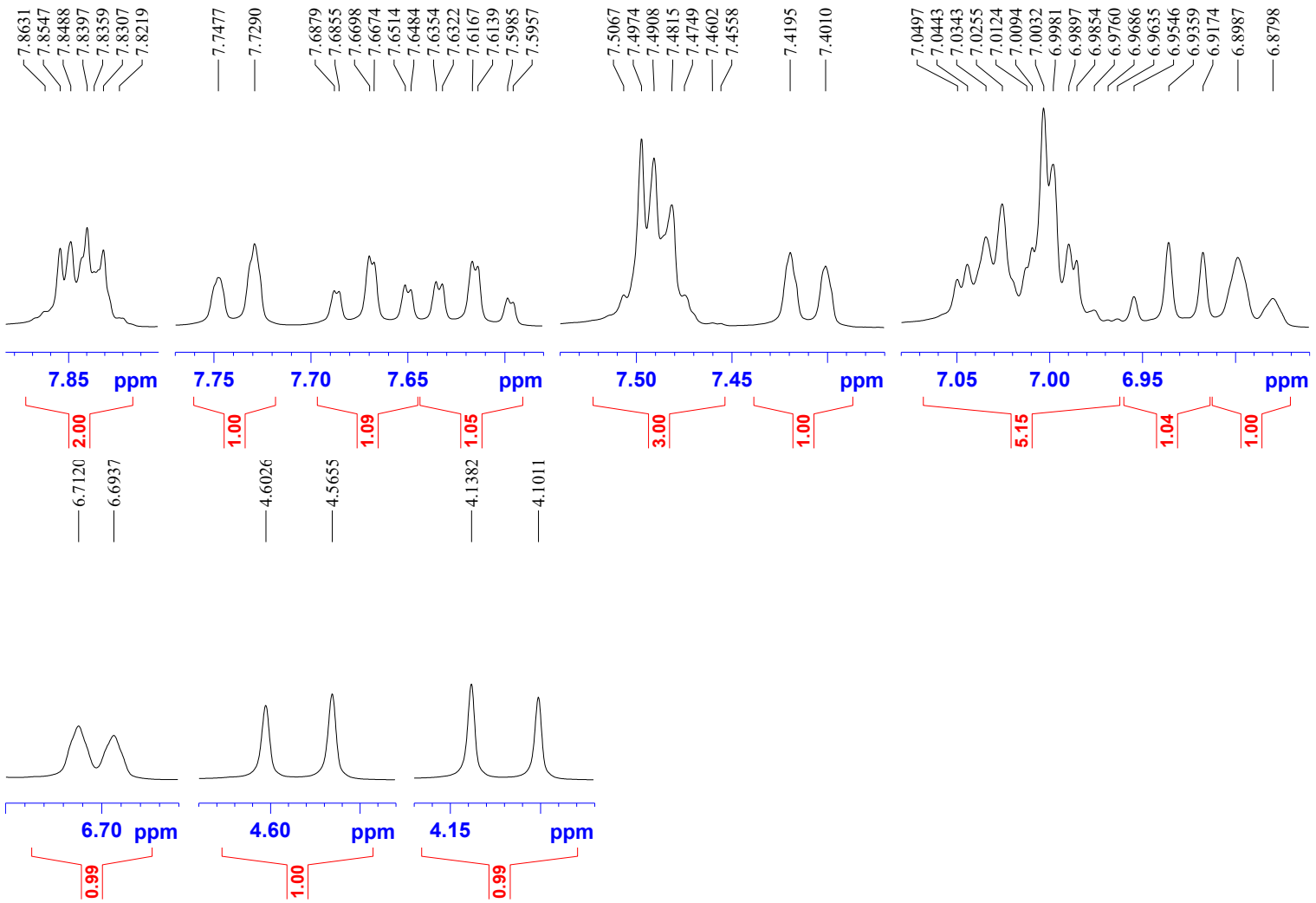


Figure S18. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ha

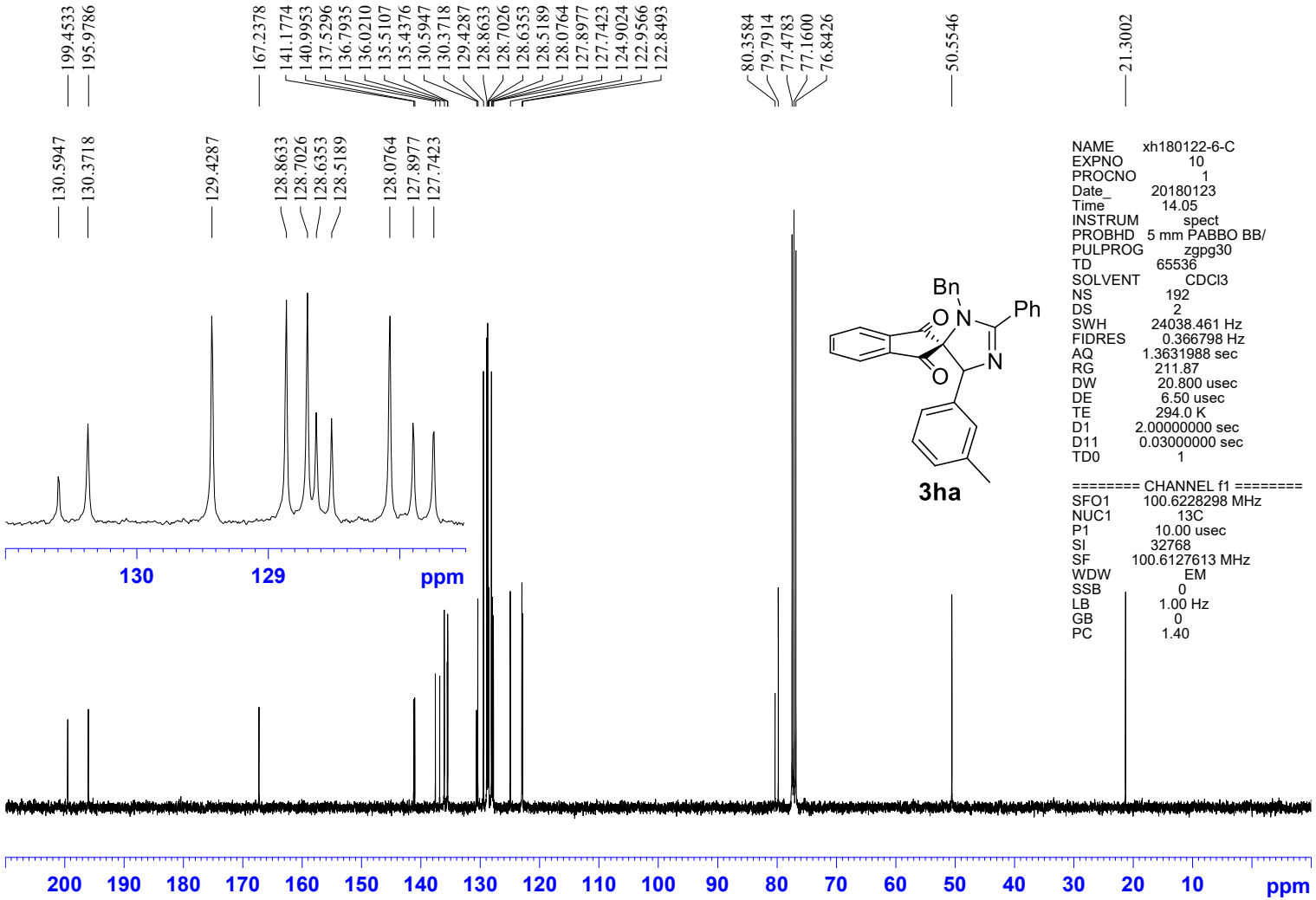


Figure S19. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ha

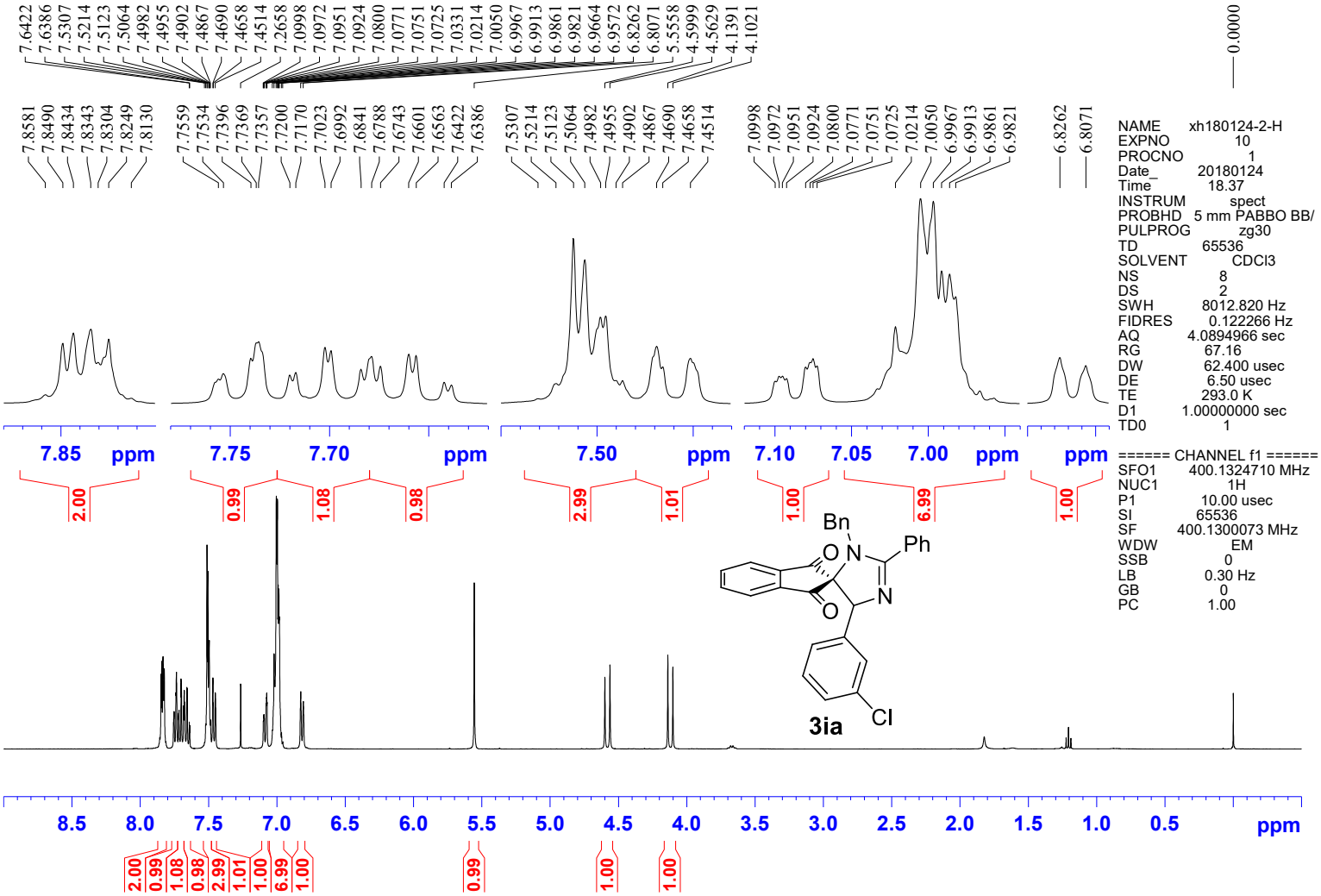


Figure S20. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ia

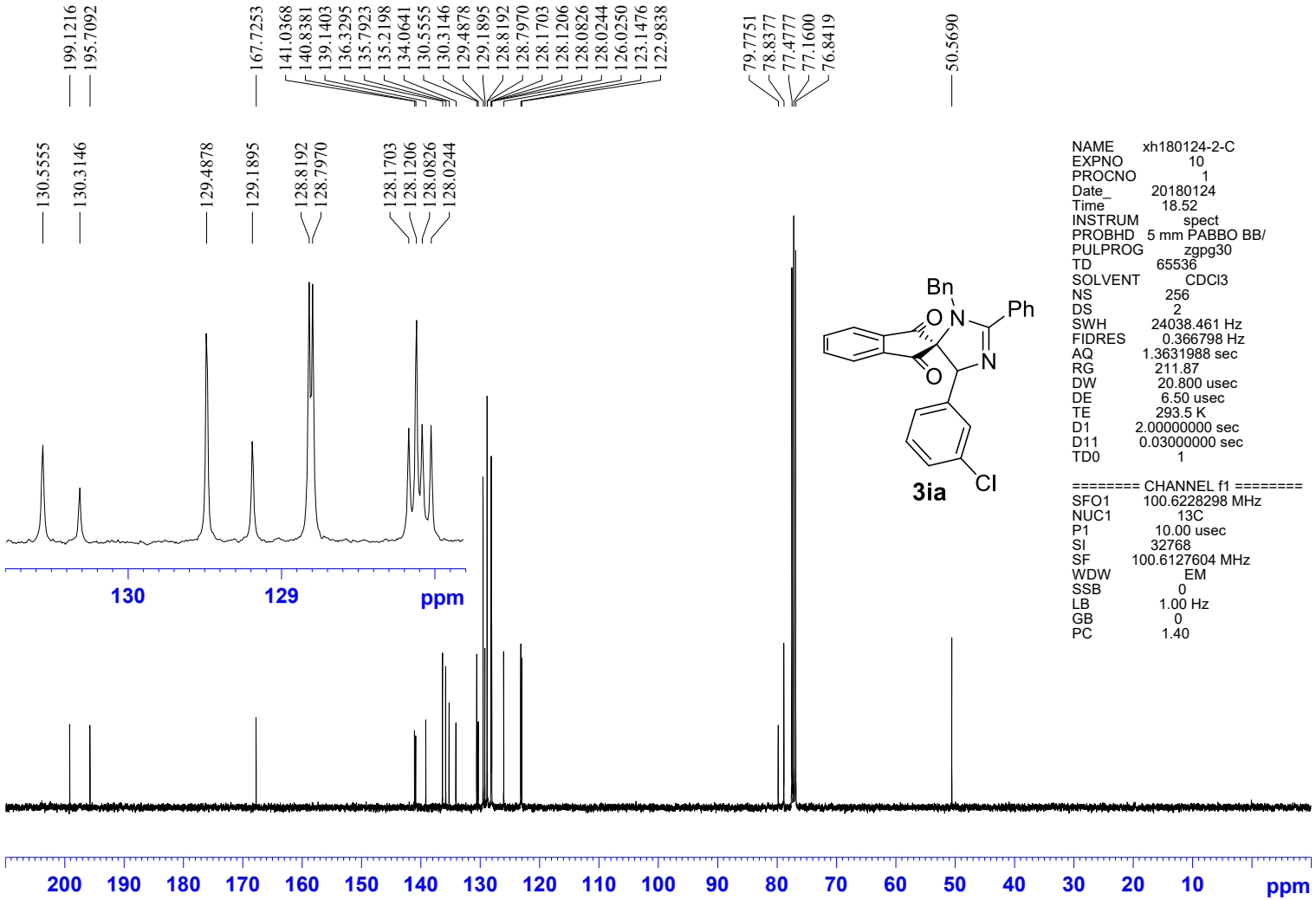


Figure S21. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ia

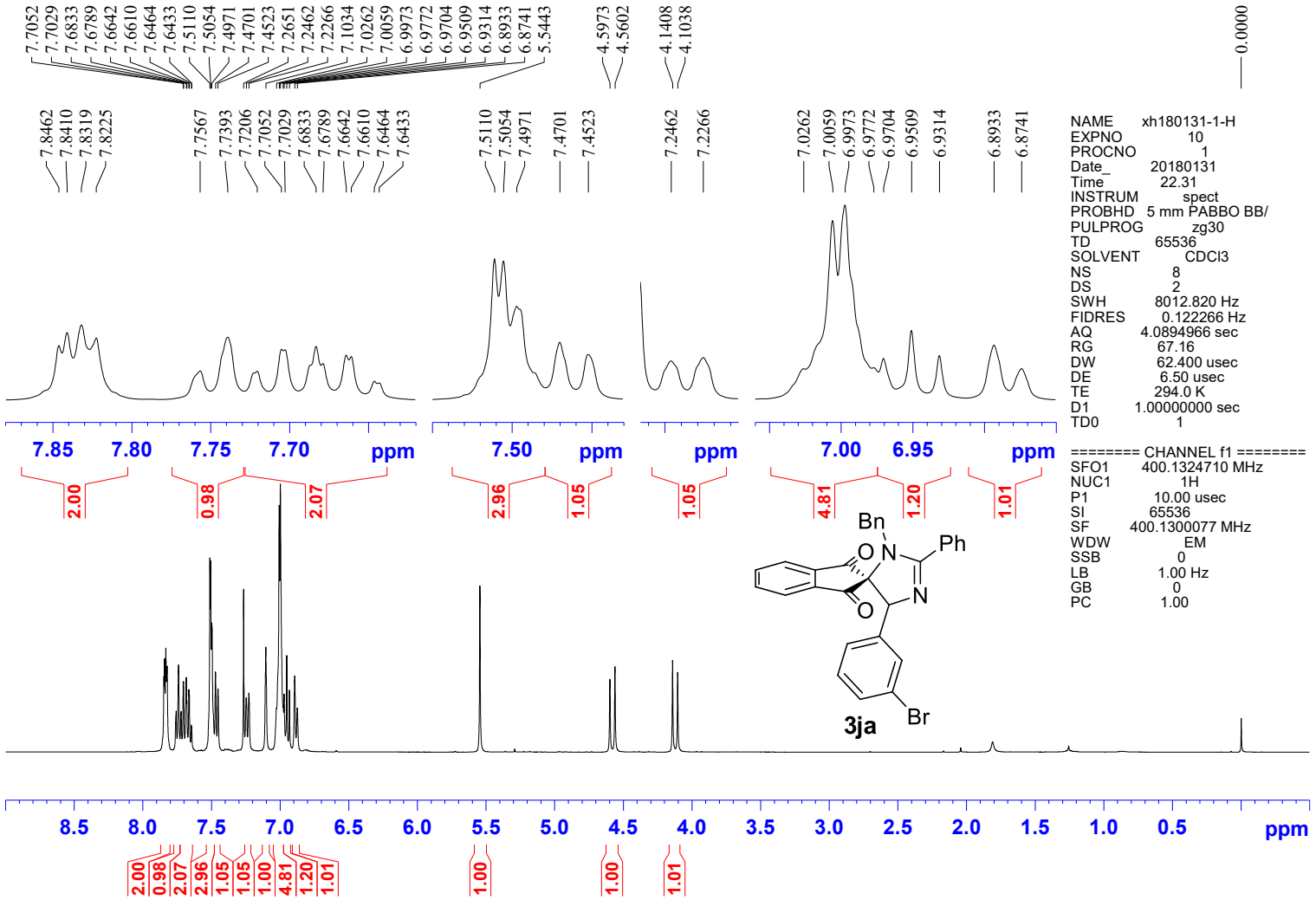


Figure S22. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ja

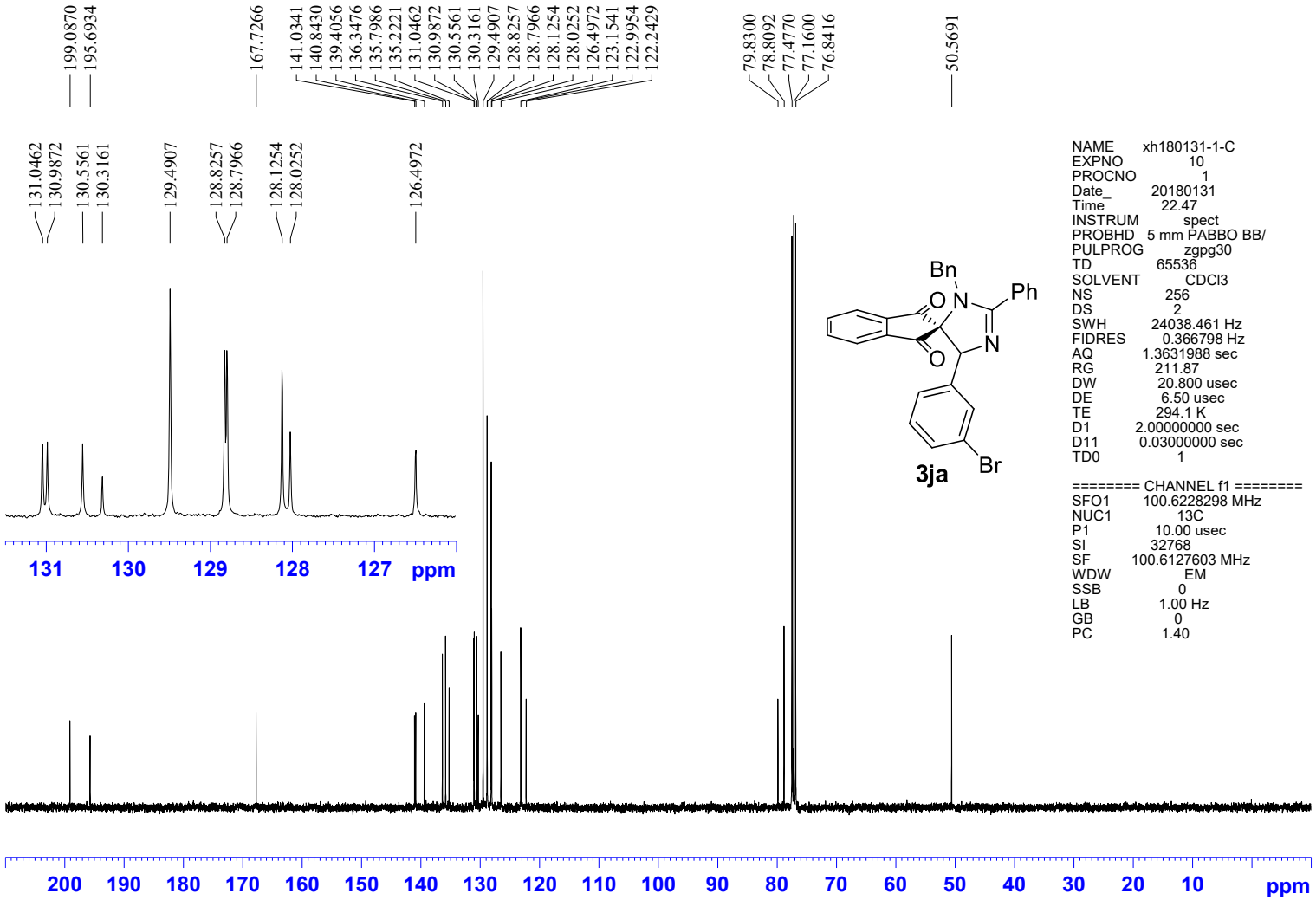


Figure S23. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ja

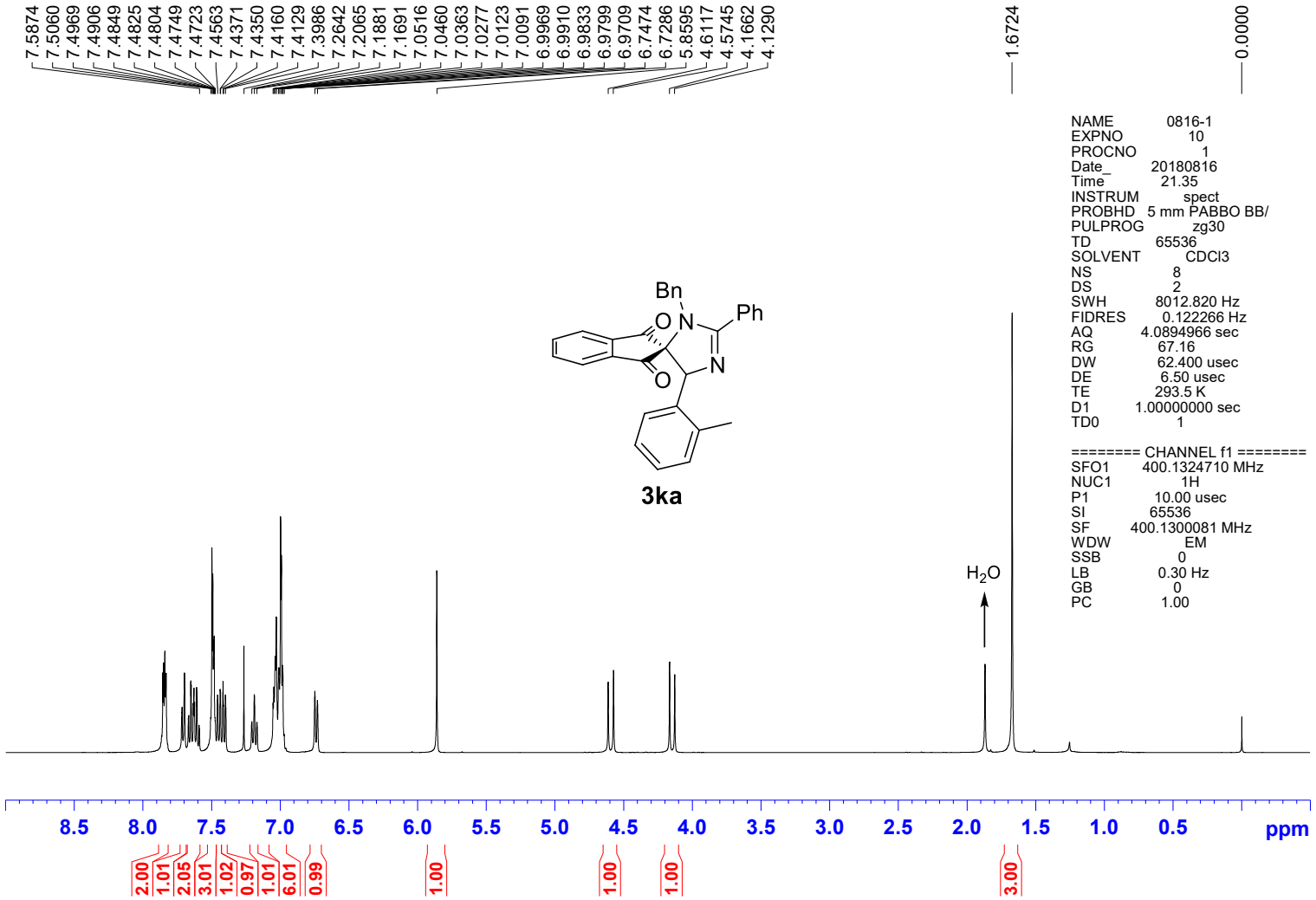


Figure S24. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ka

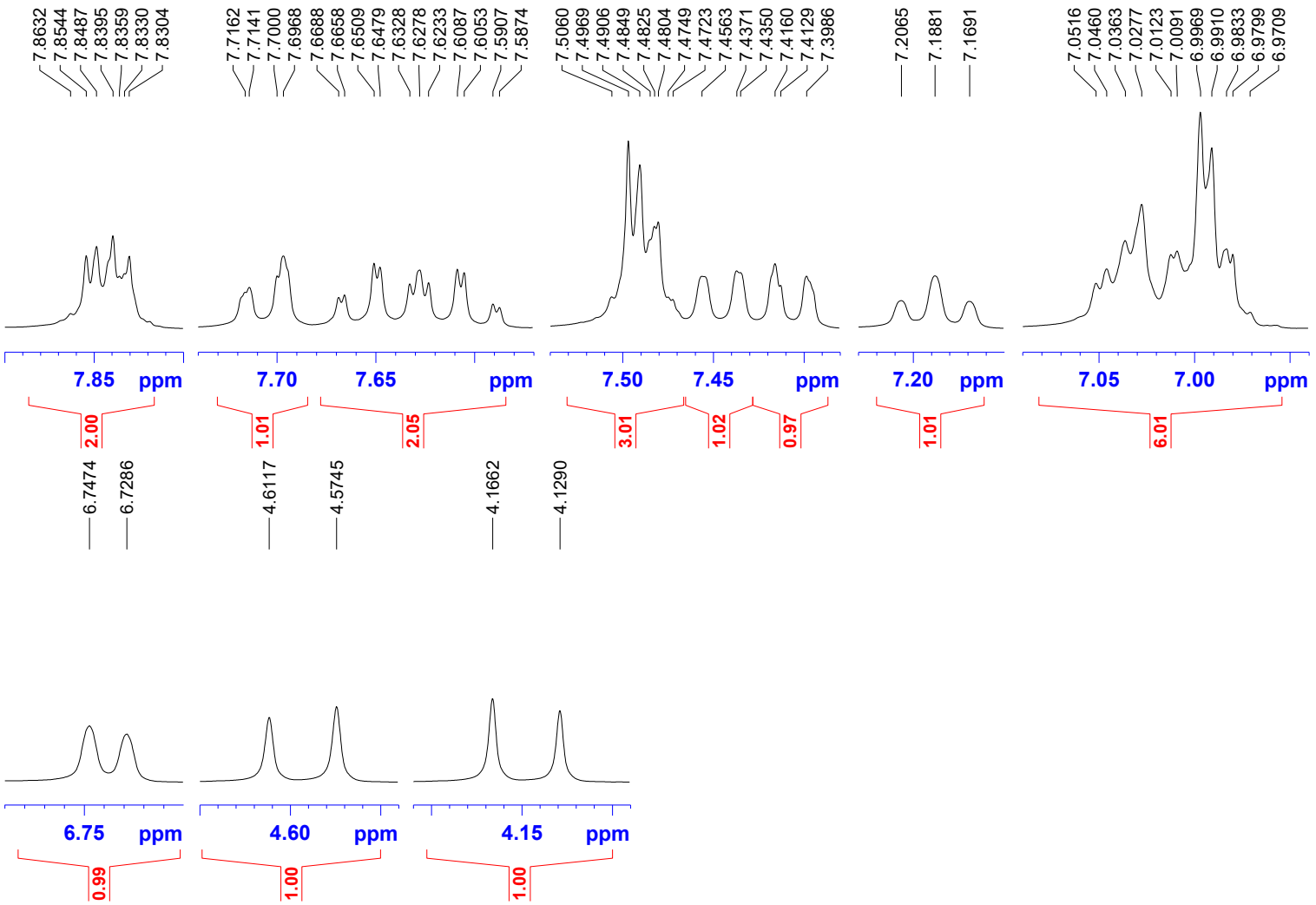


Figure S25. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ka

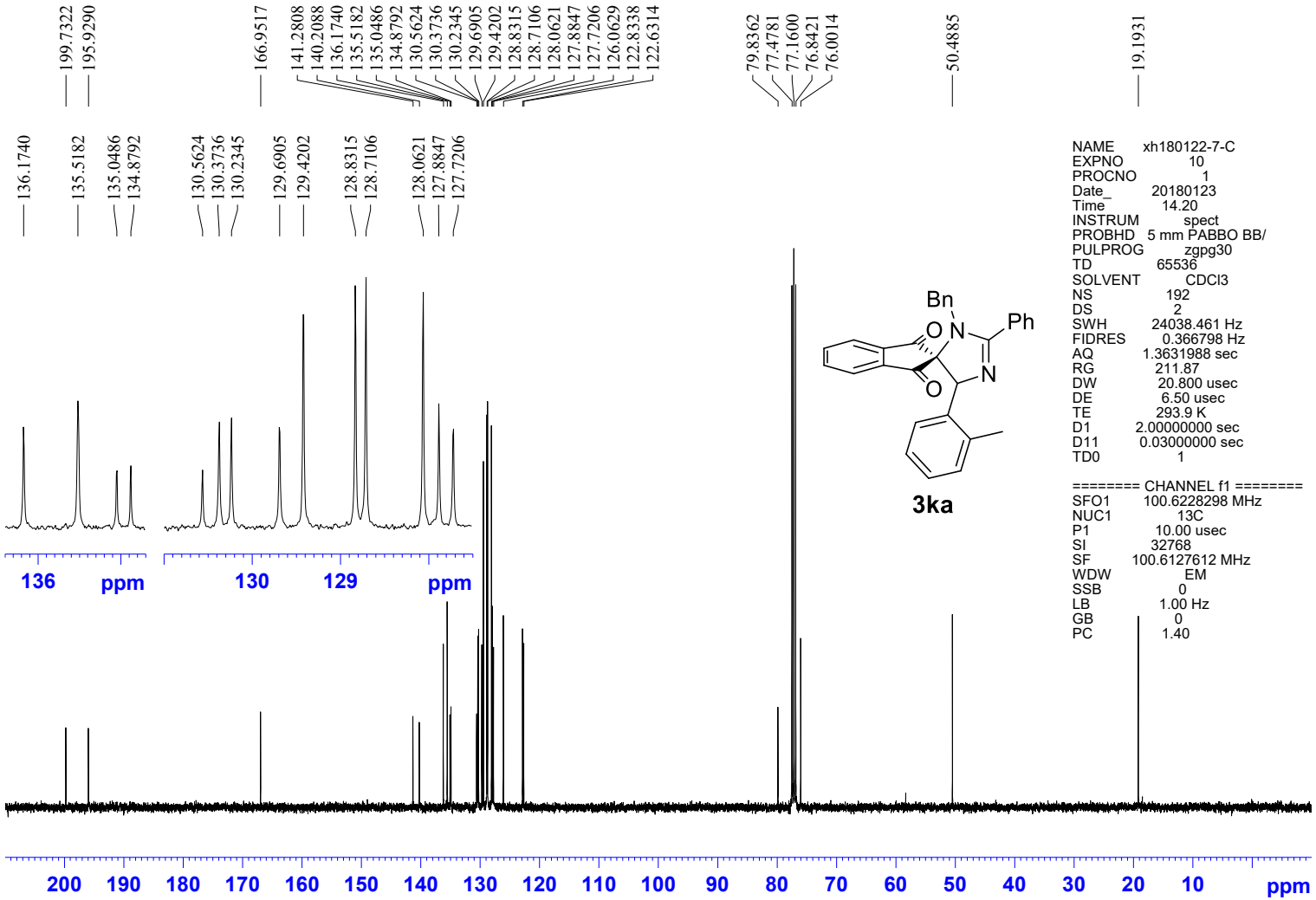


Figure S26. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ka

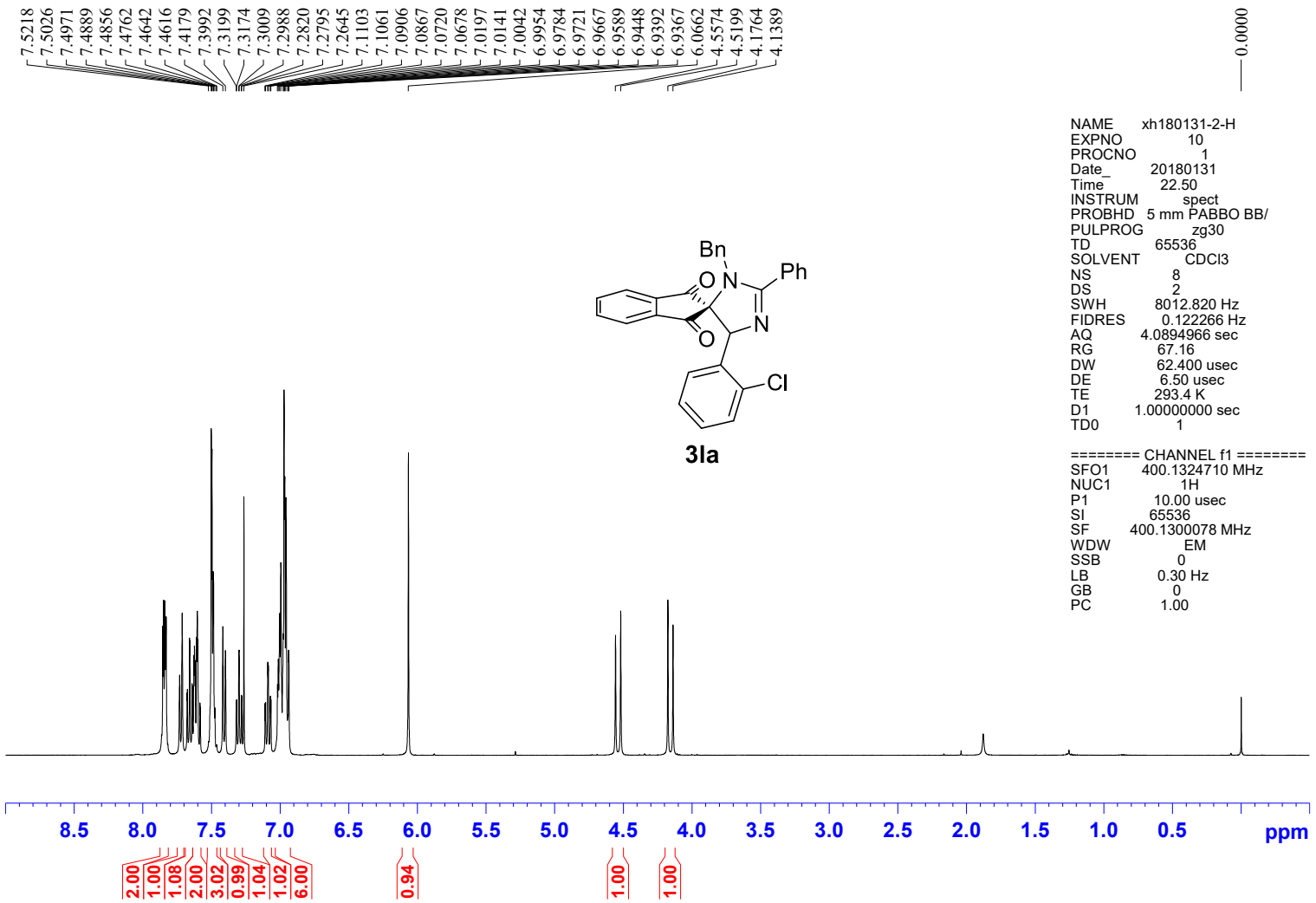


Figure S27. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3la

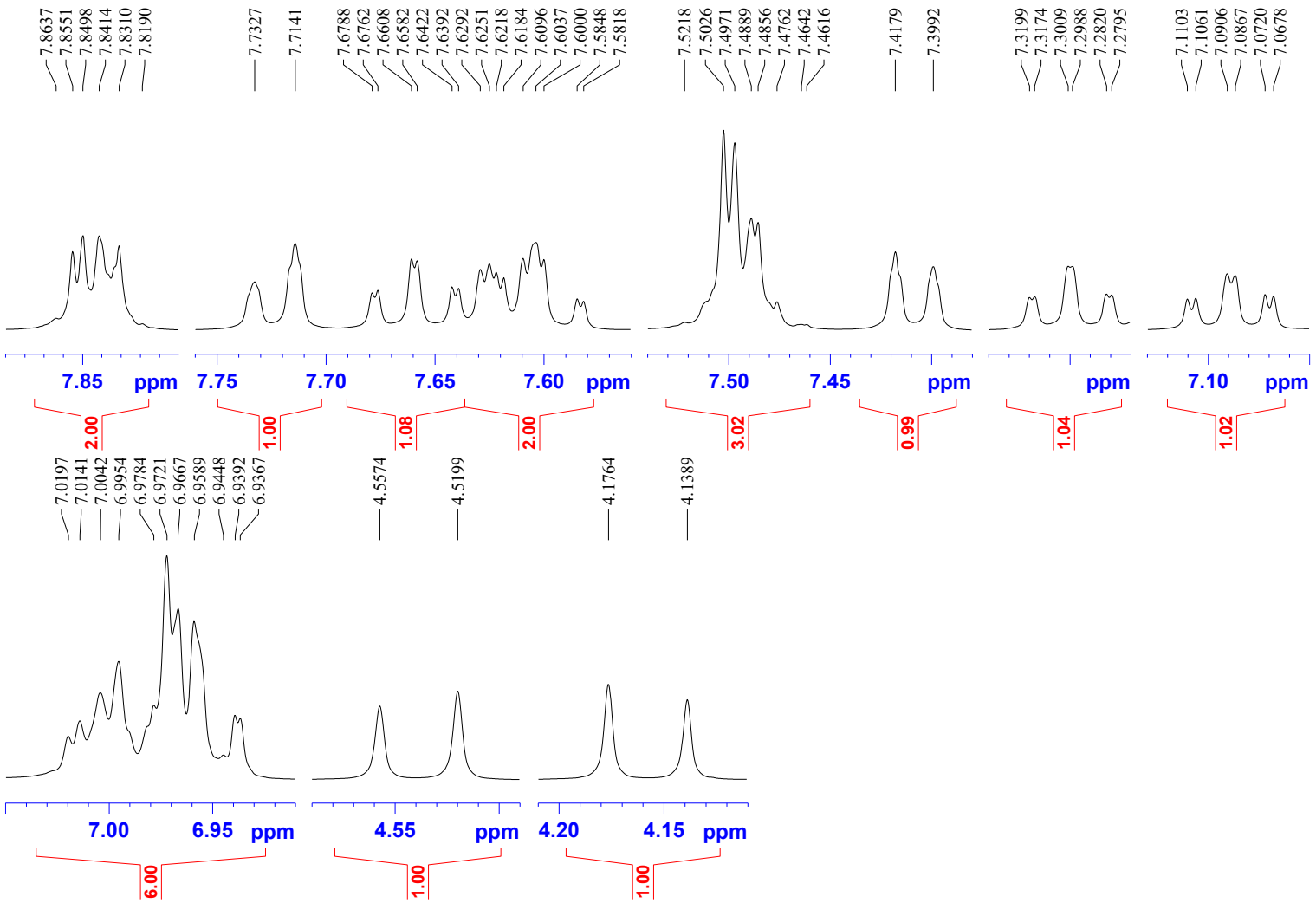


Figure S28. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3la

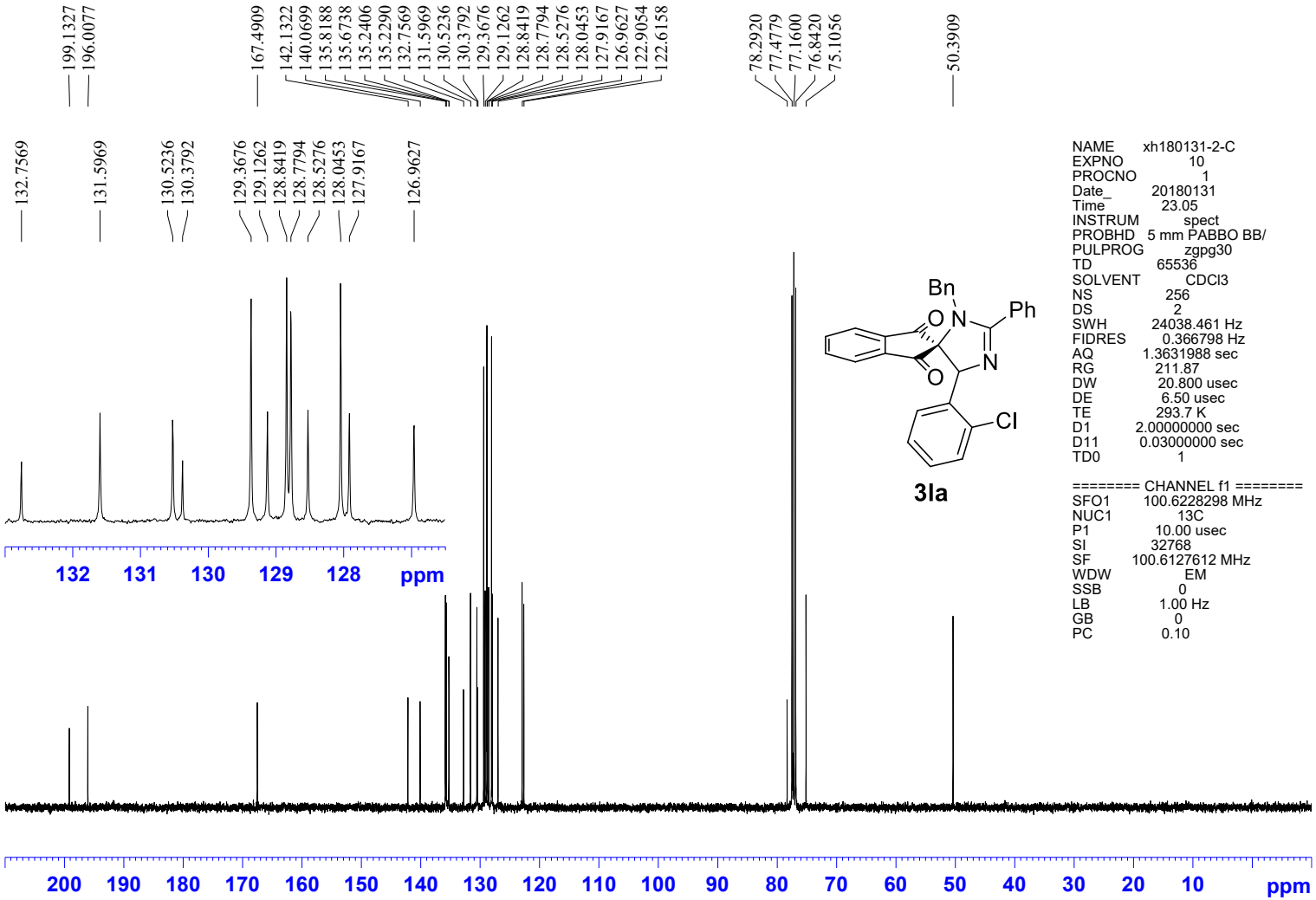


Figure S29. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3la

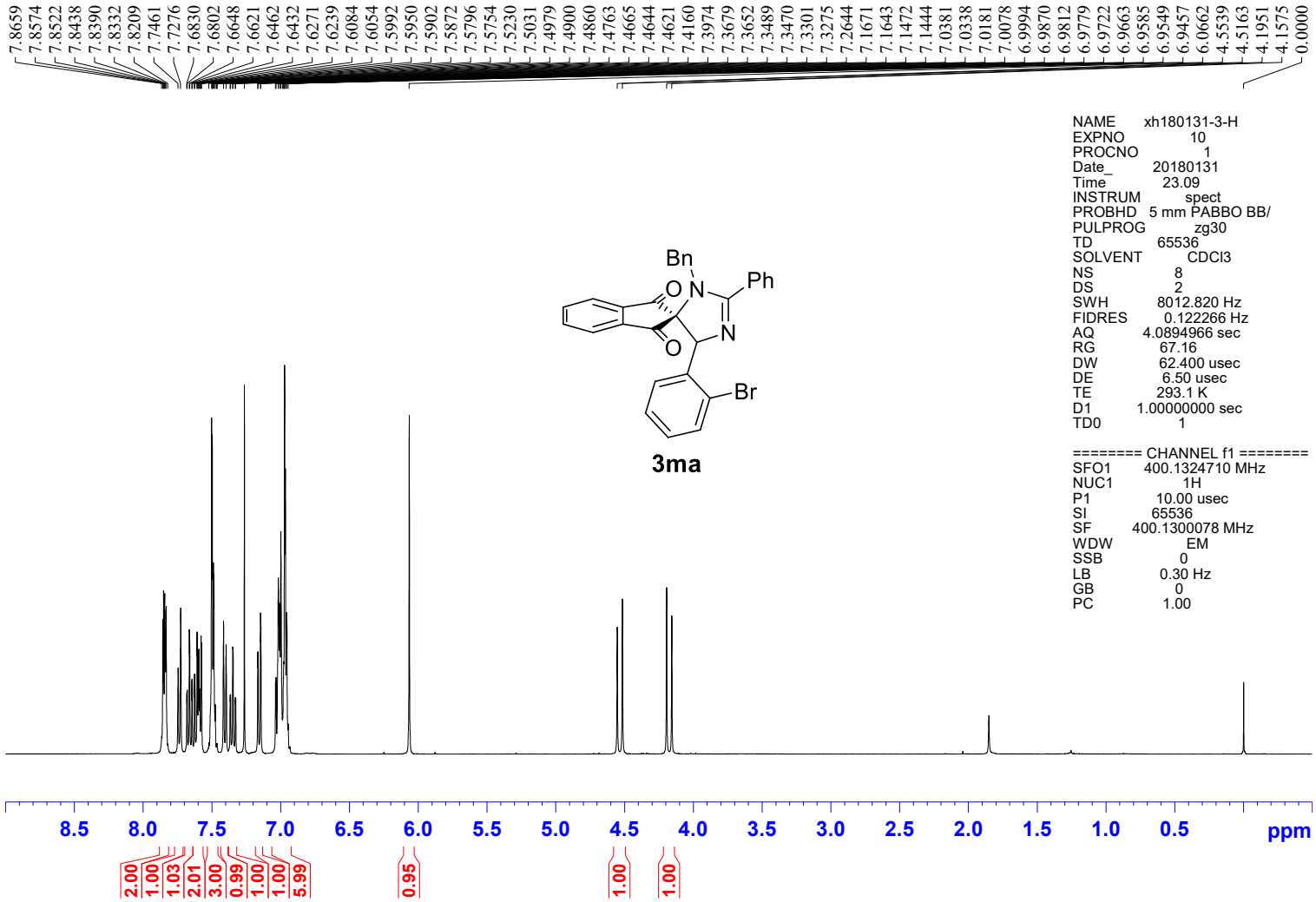


Figure S30. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ma

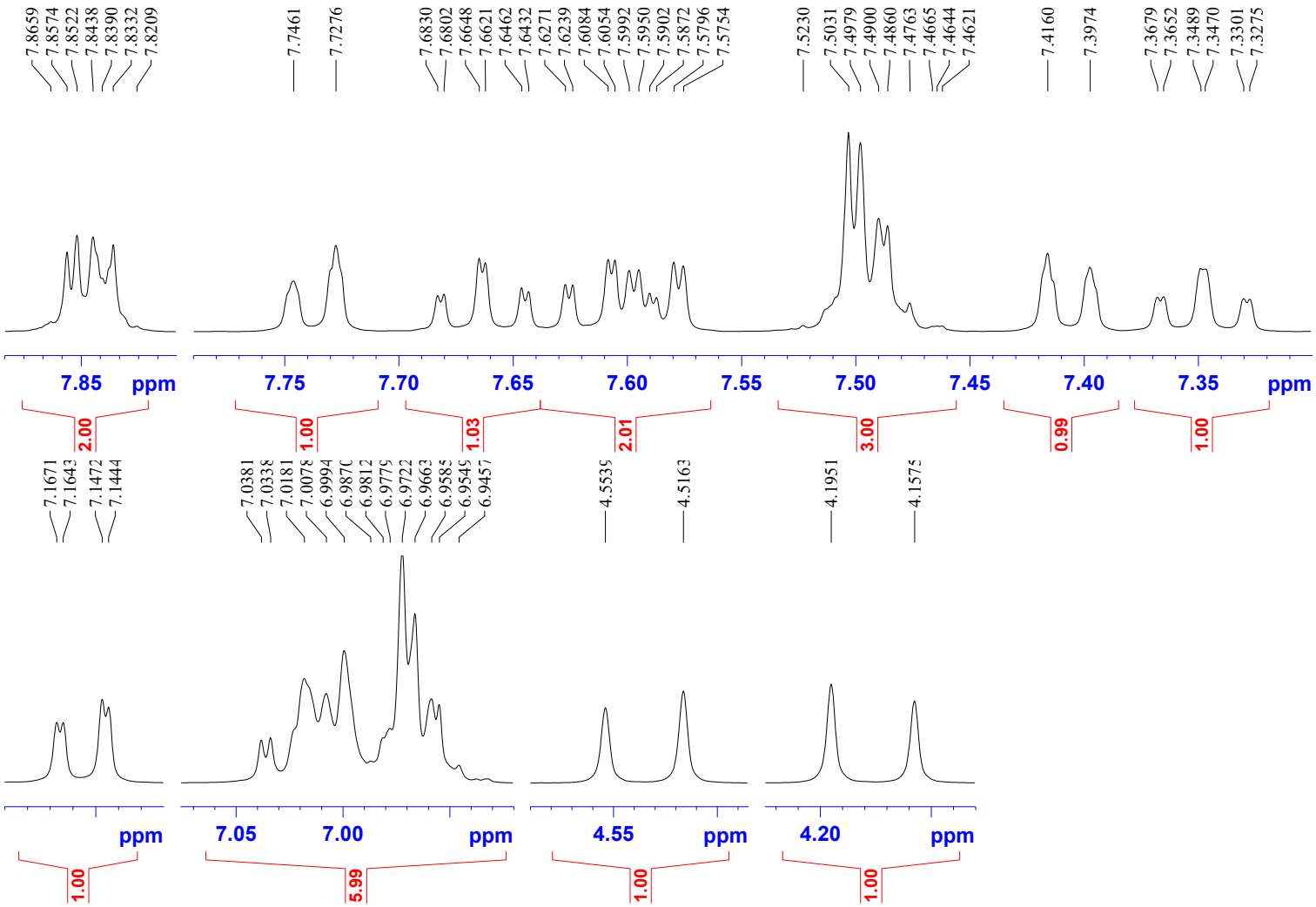


Figure S31. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ma

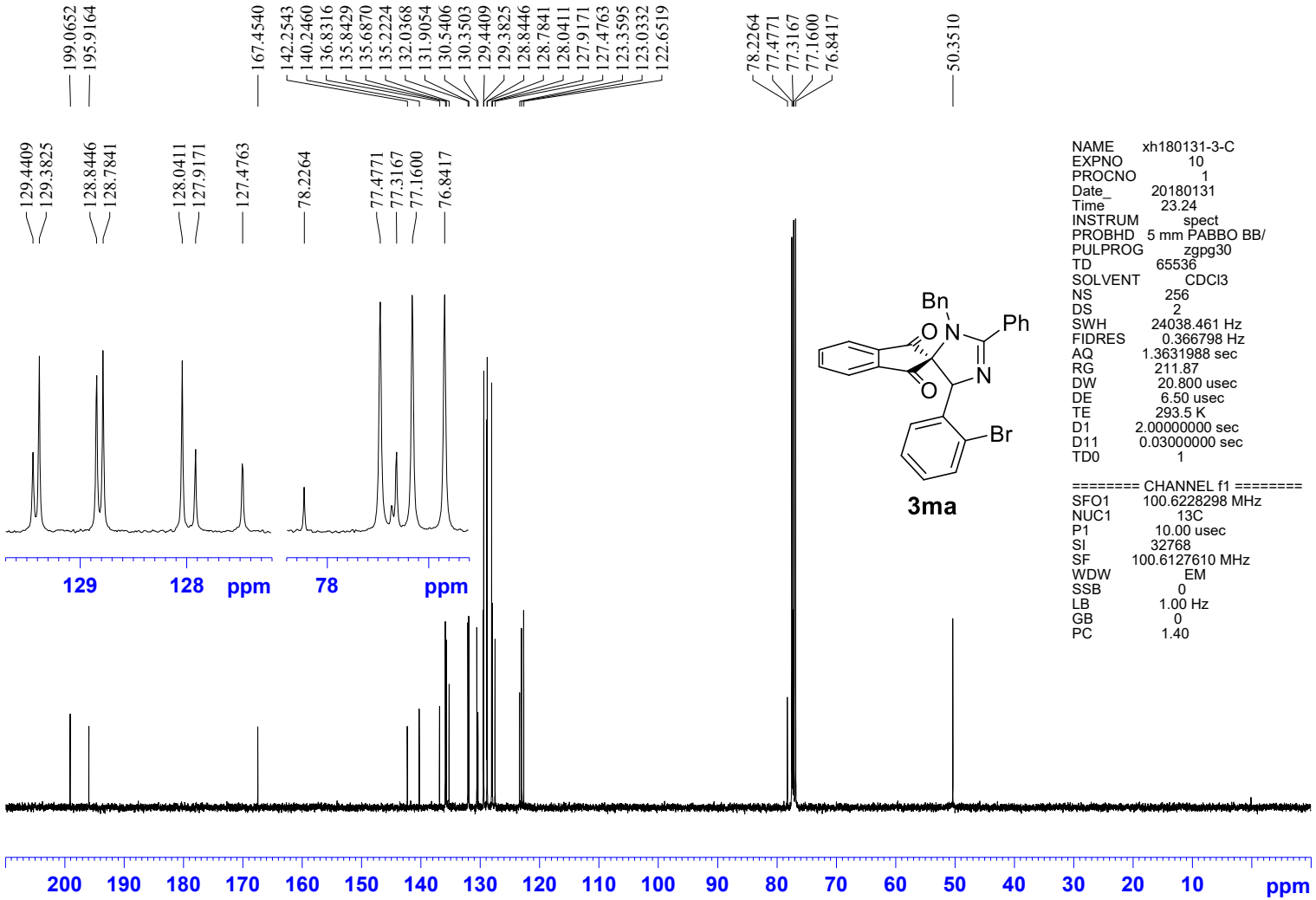


Figure S32. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ma

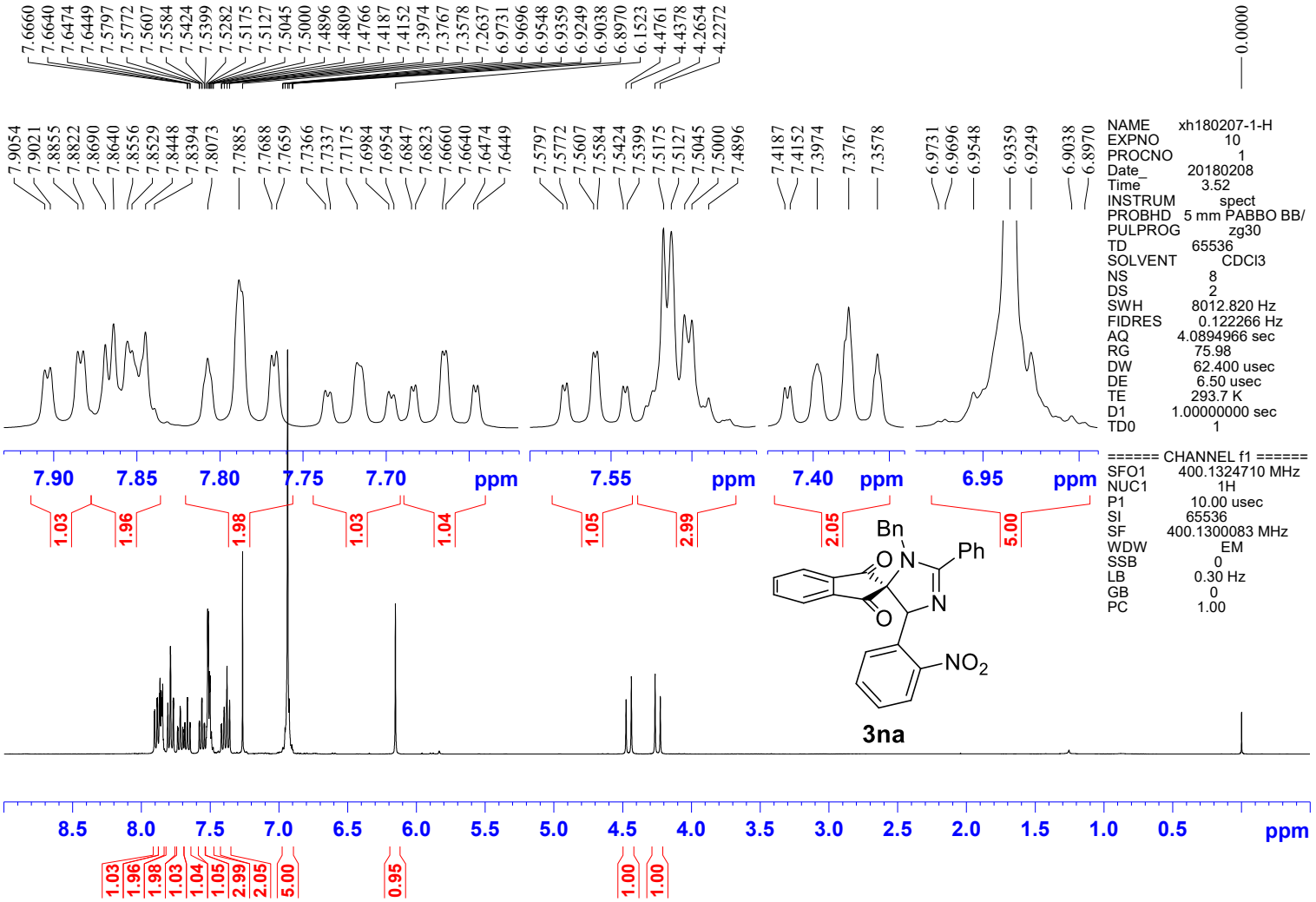


Figure S33. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3na

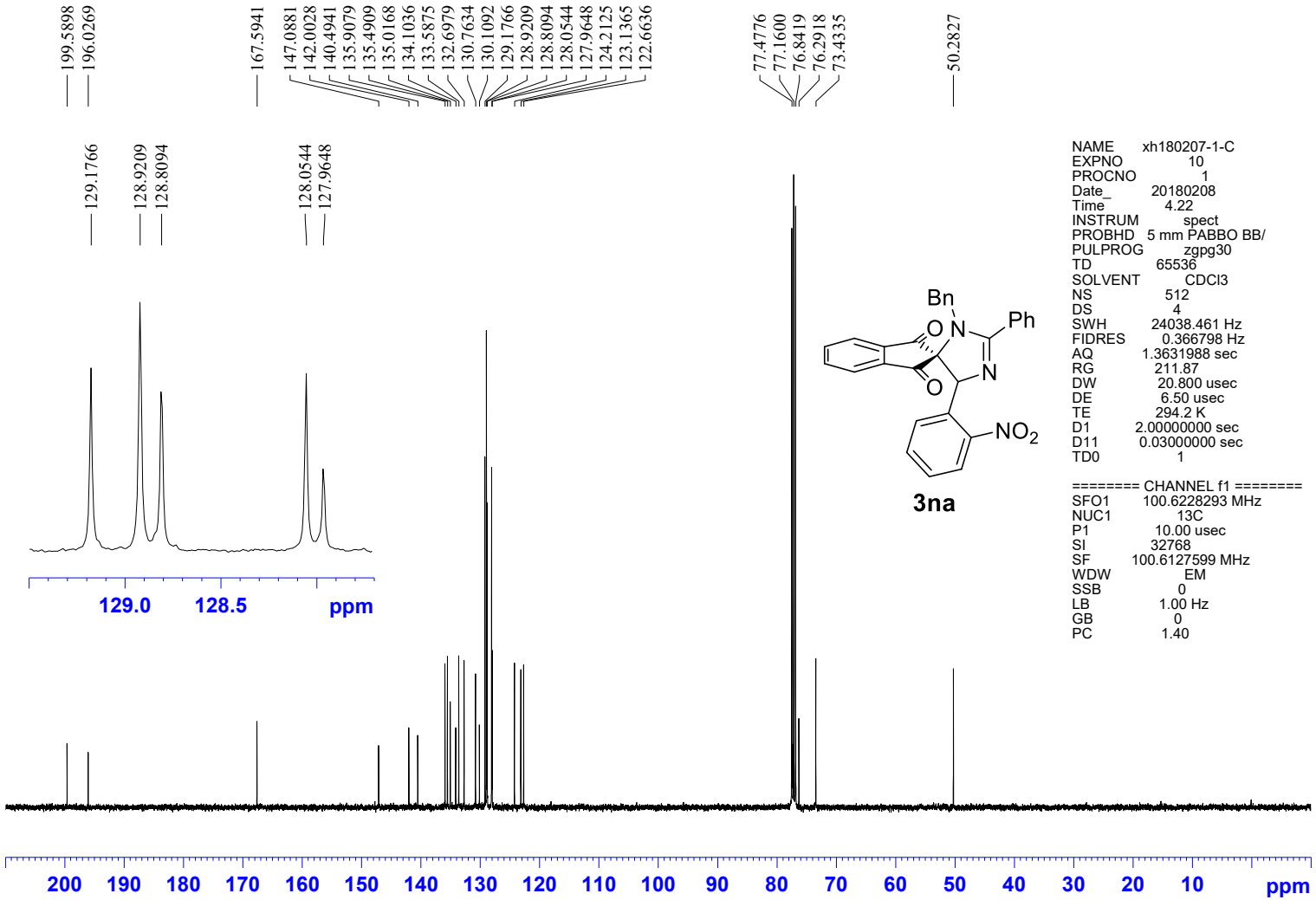


Figure S34. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3na

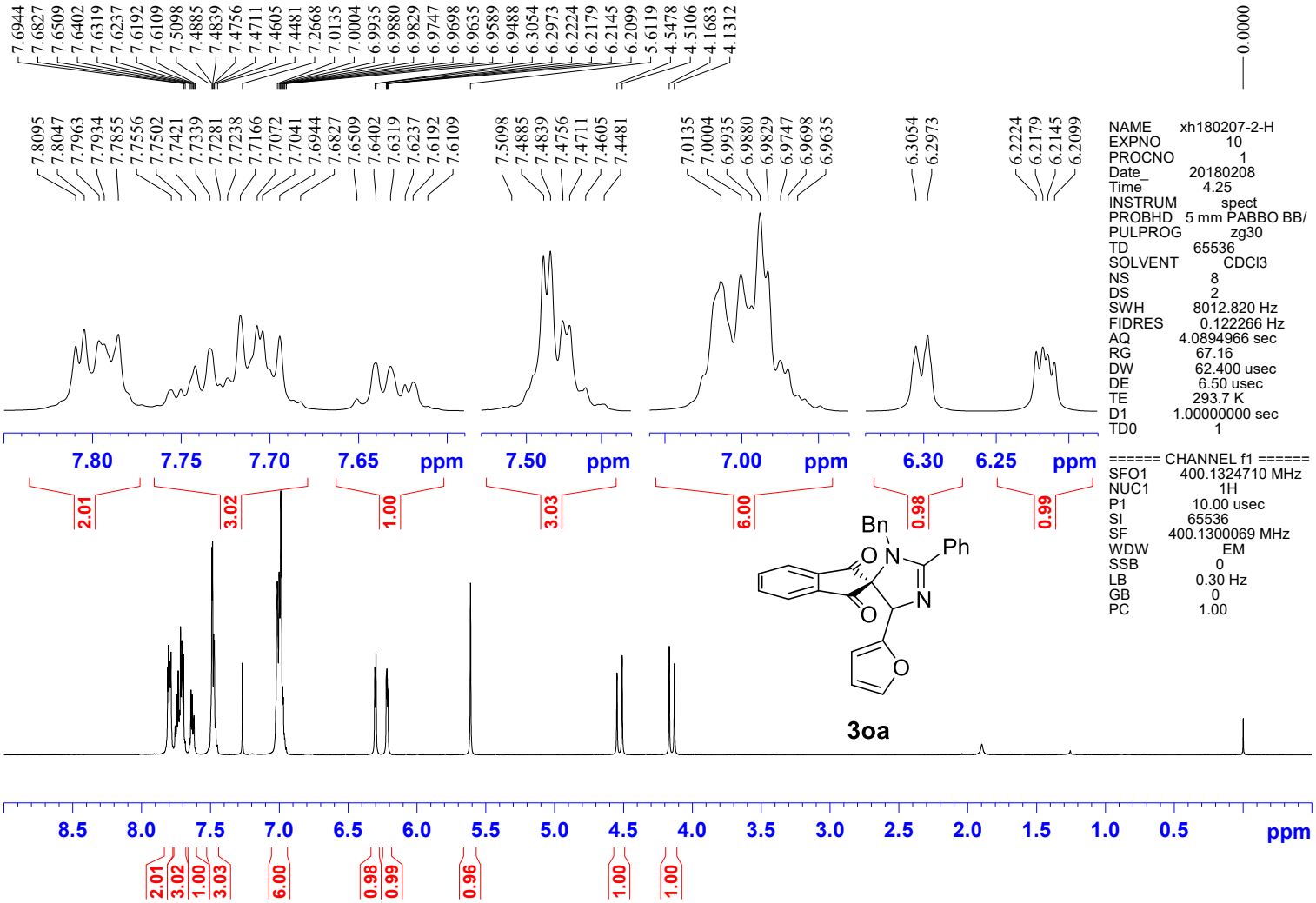


Figure S35. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3oa

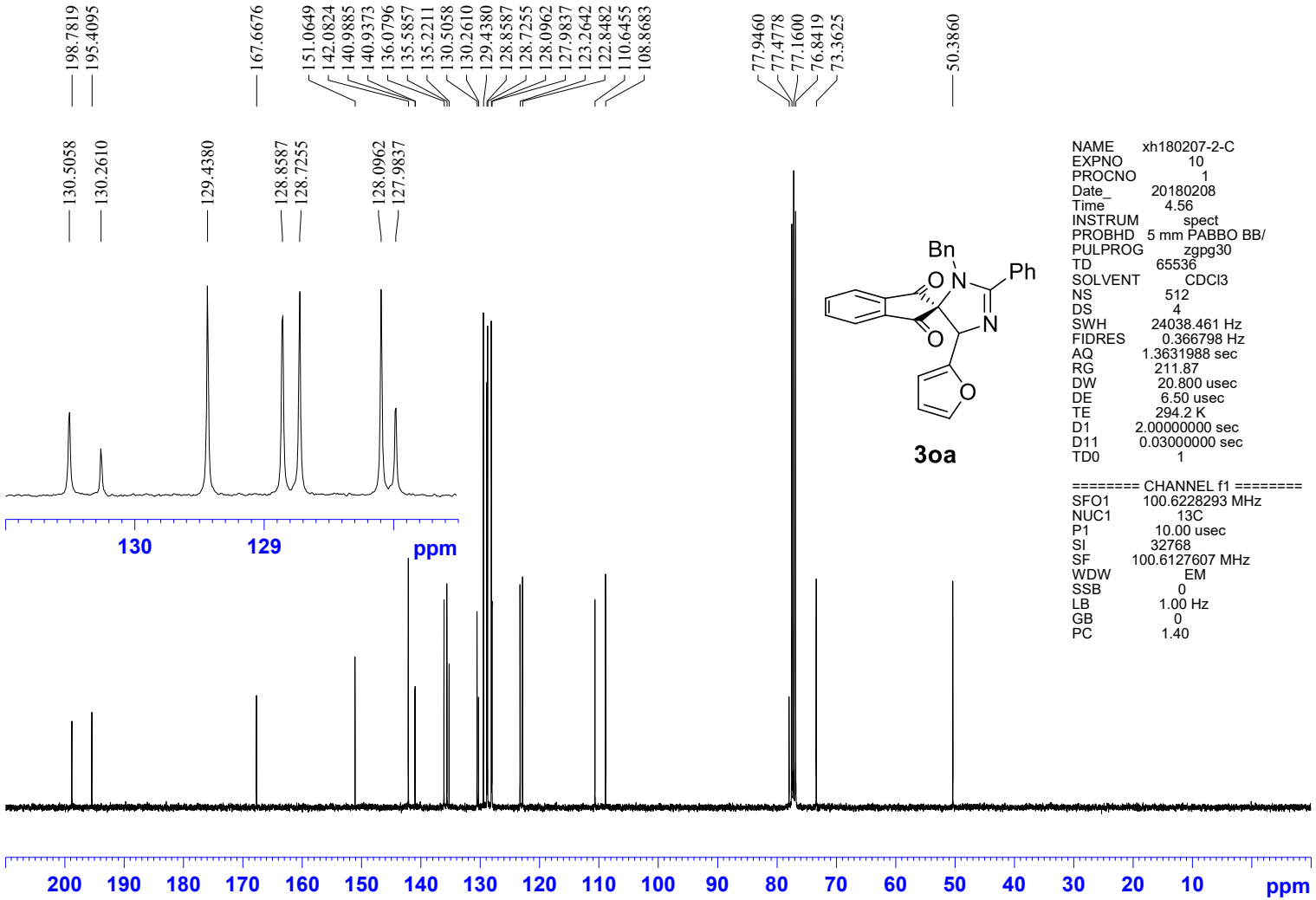


Figure S36. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3oa

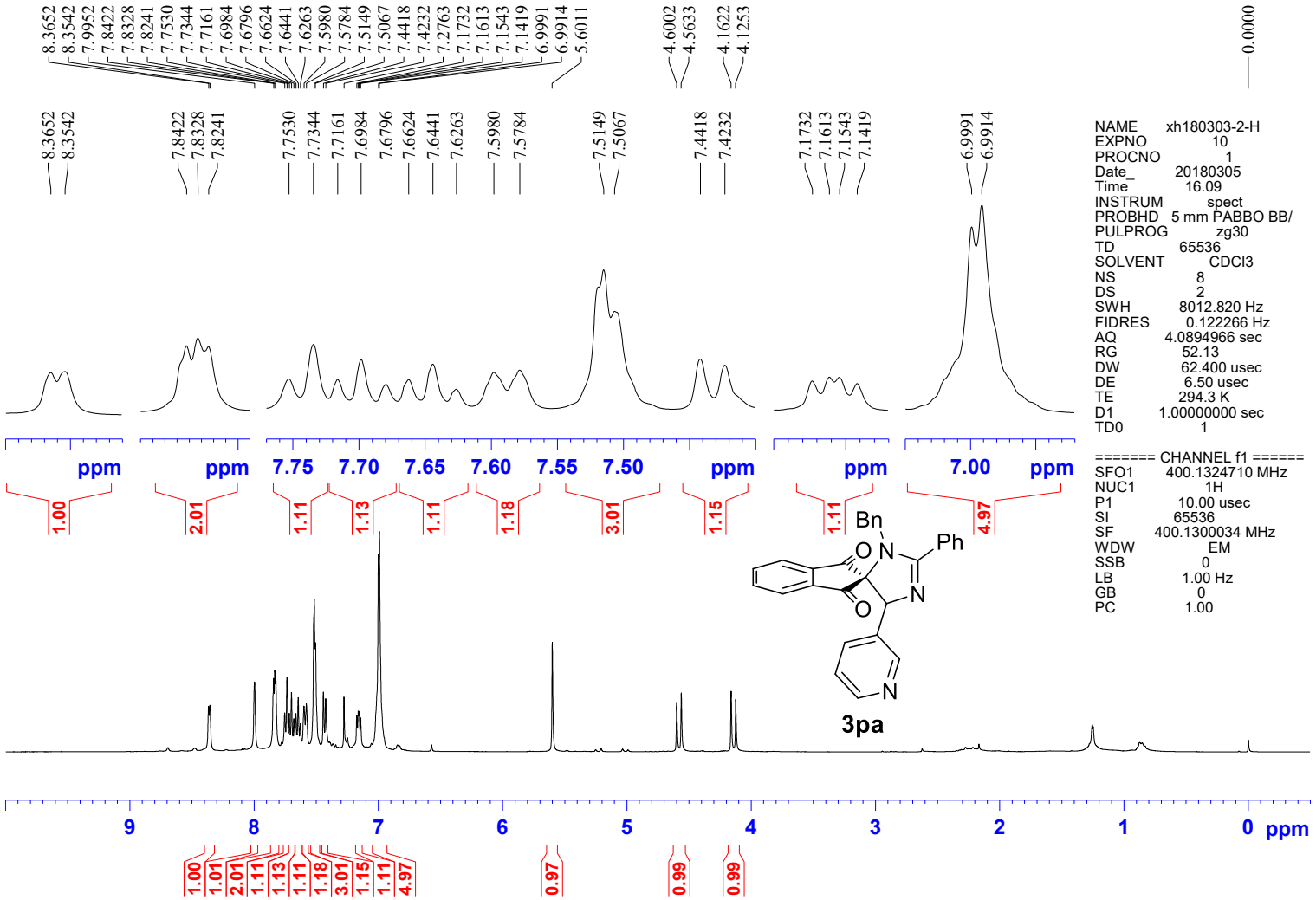


Figure S37. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3pa

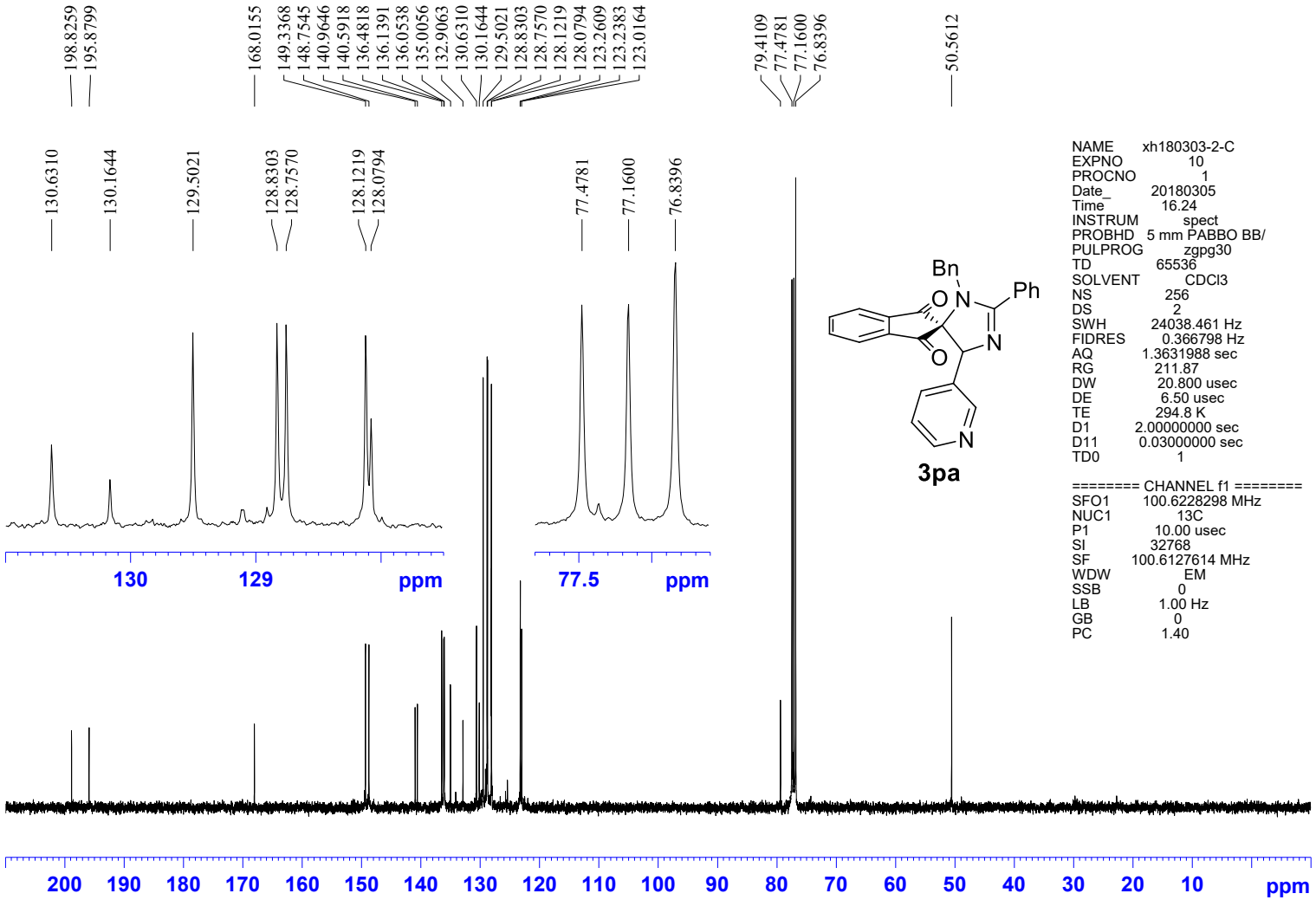


Figure S38. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3pa

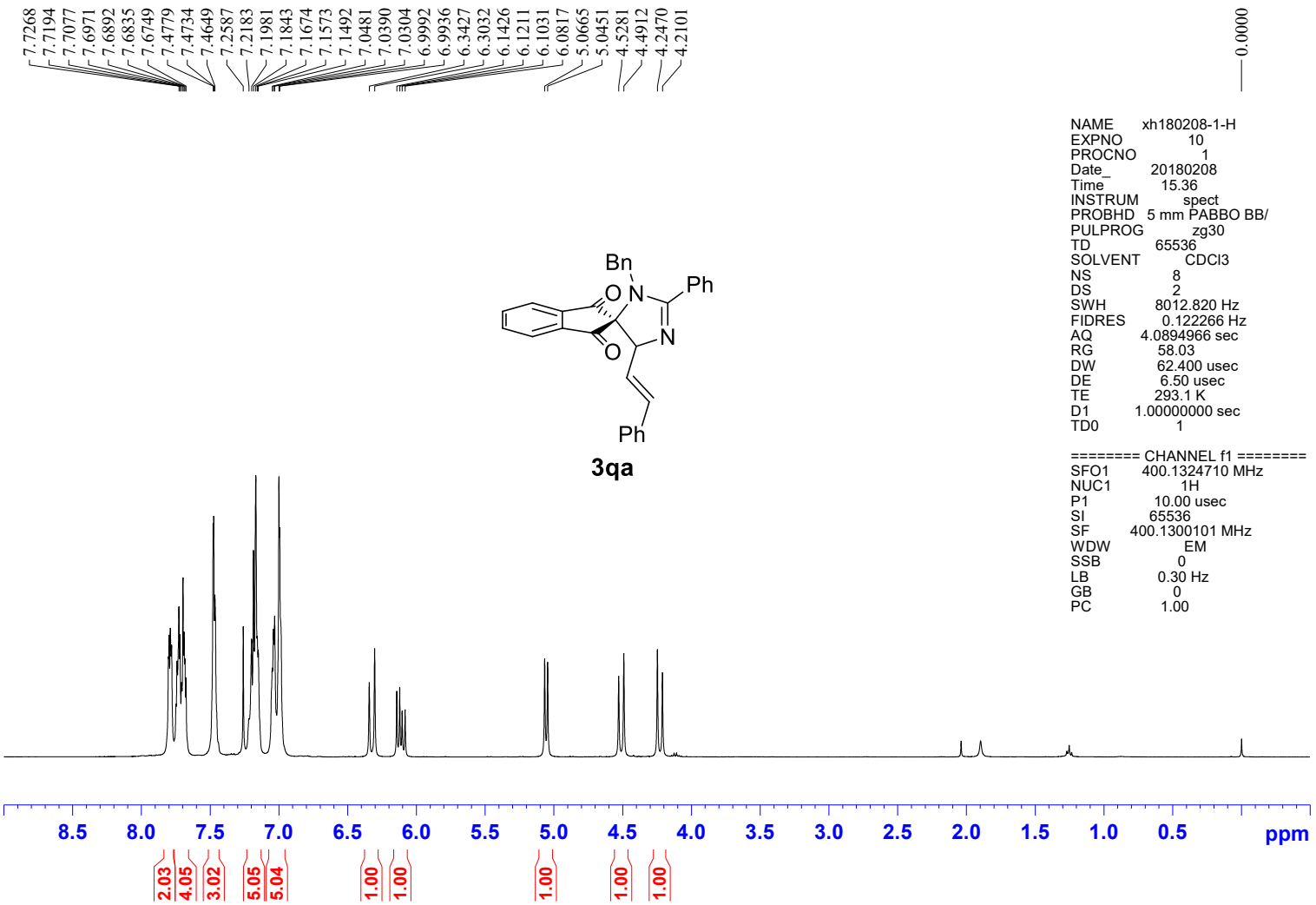


Figure S39. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3qa

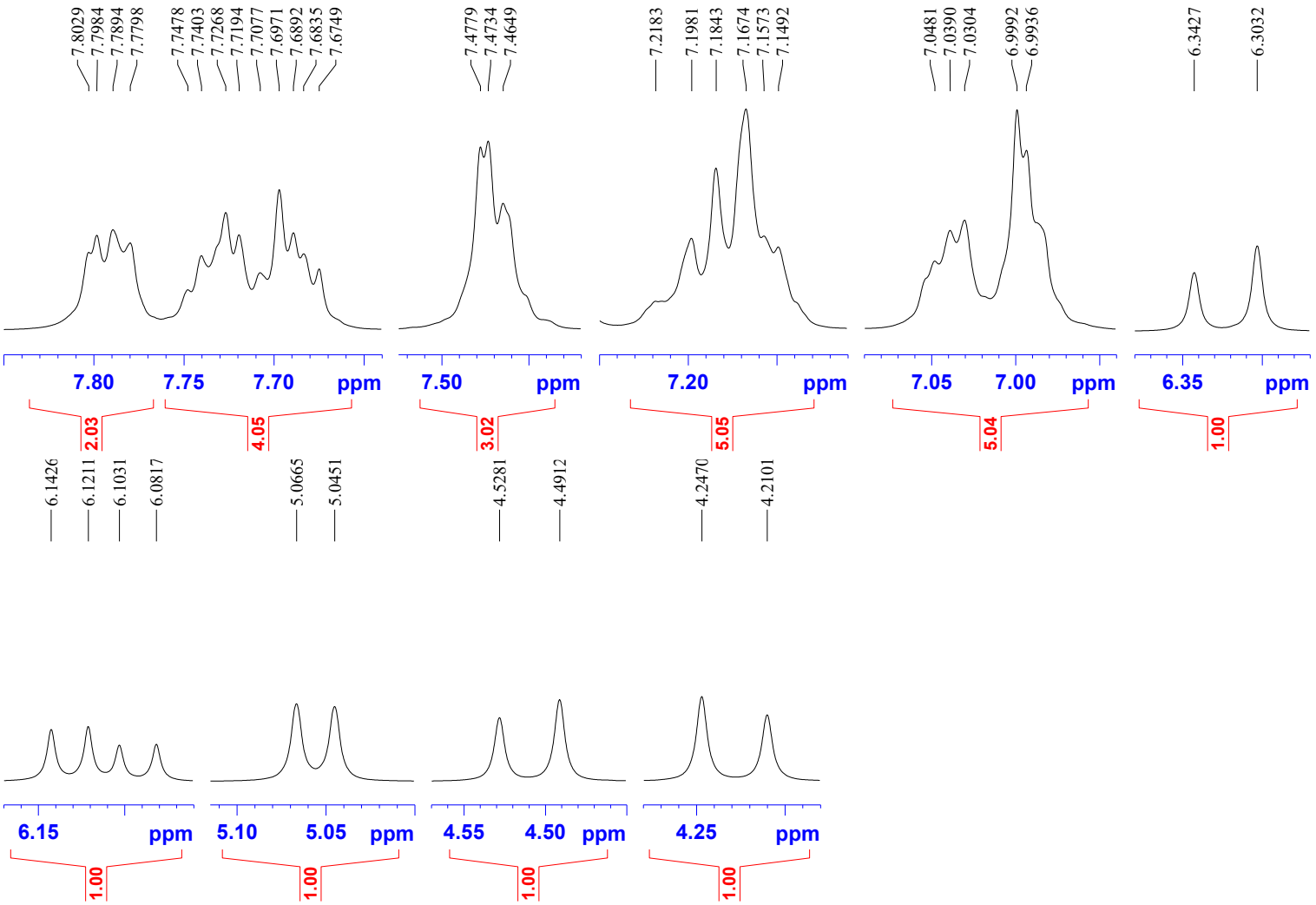


Figure S40. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3qa

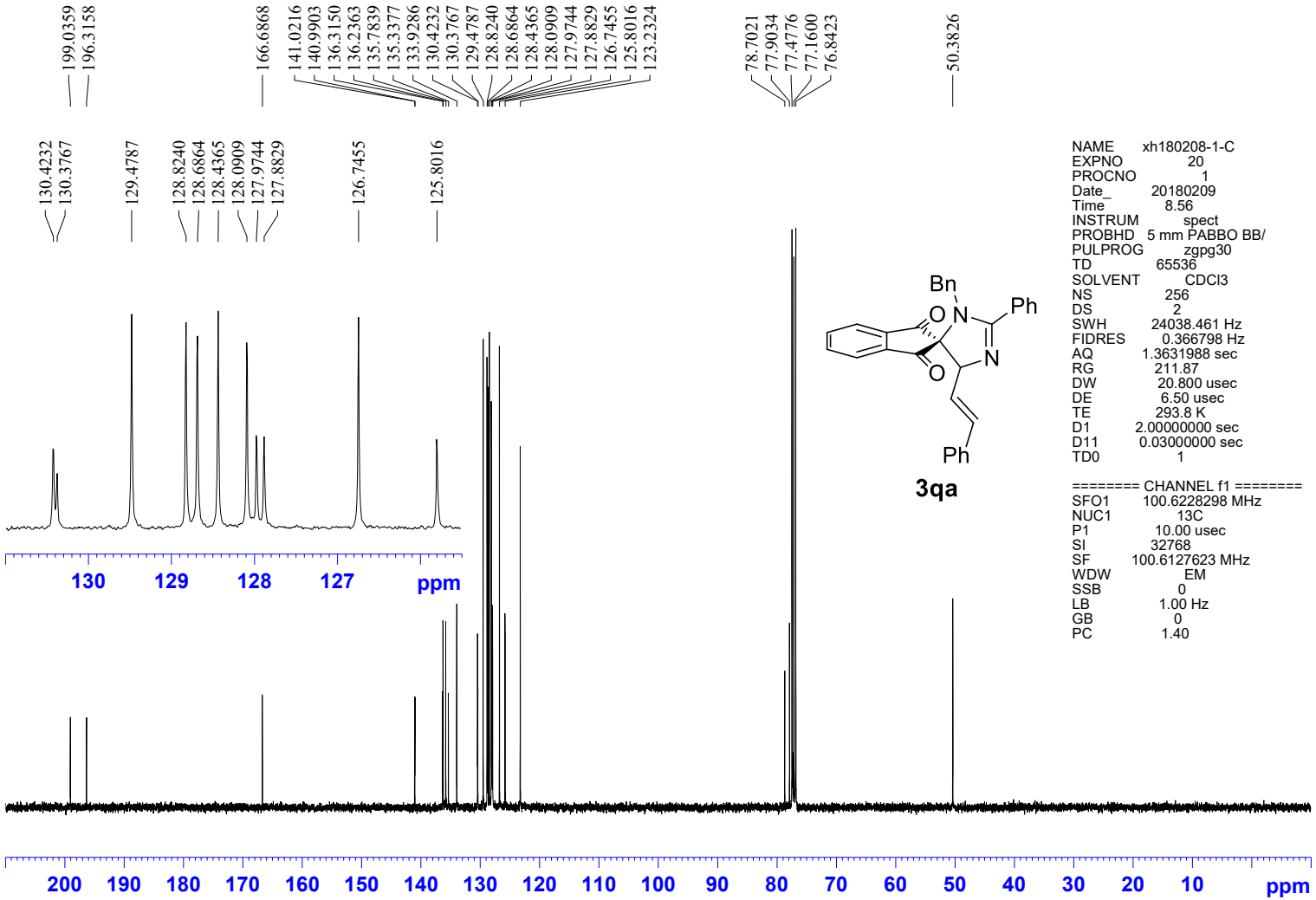


Figure S41. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3qa

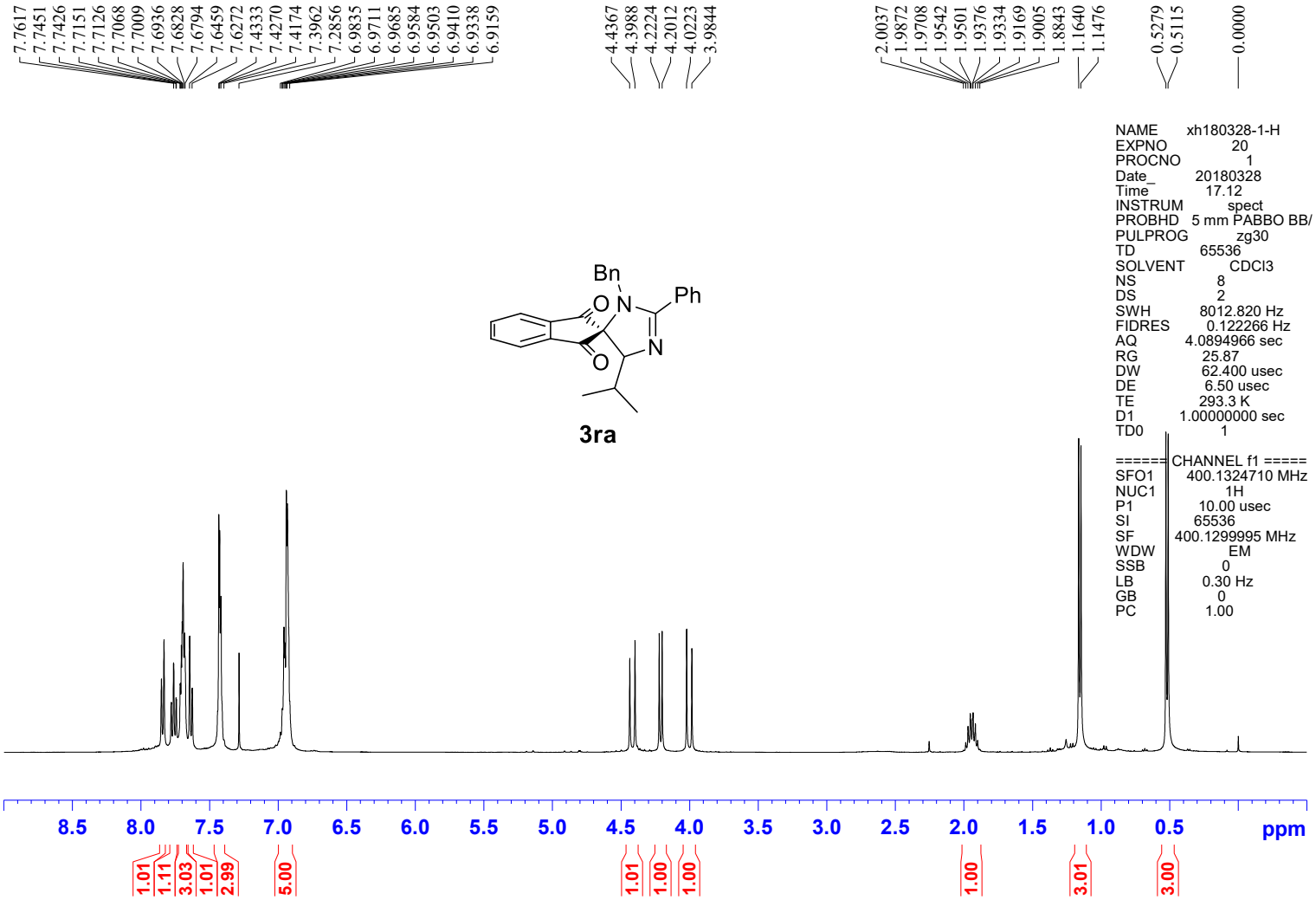


Figure S42. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ra

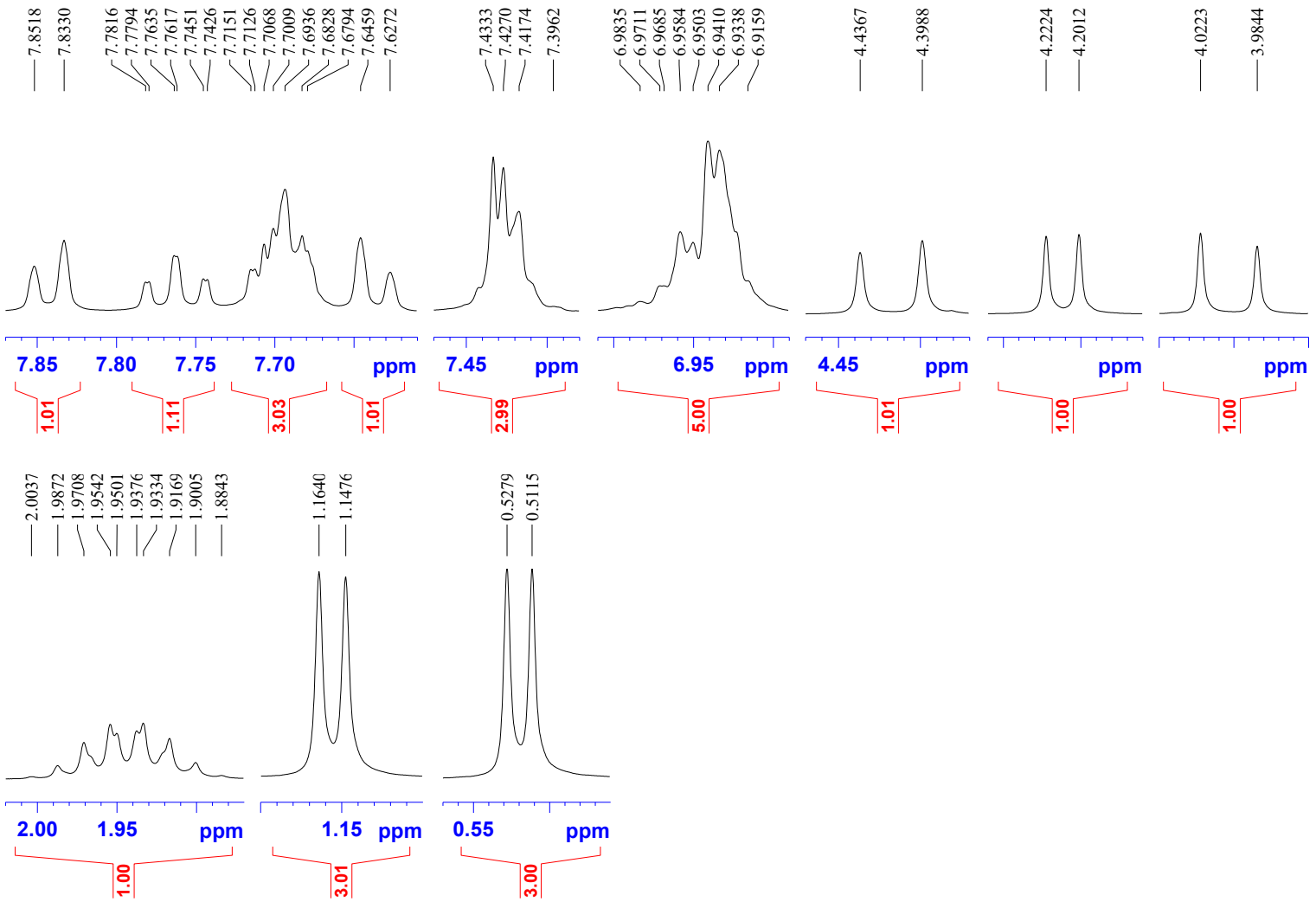


Figure S43. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ra

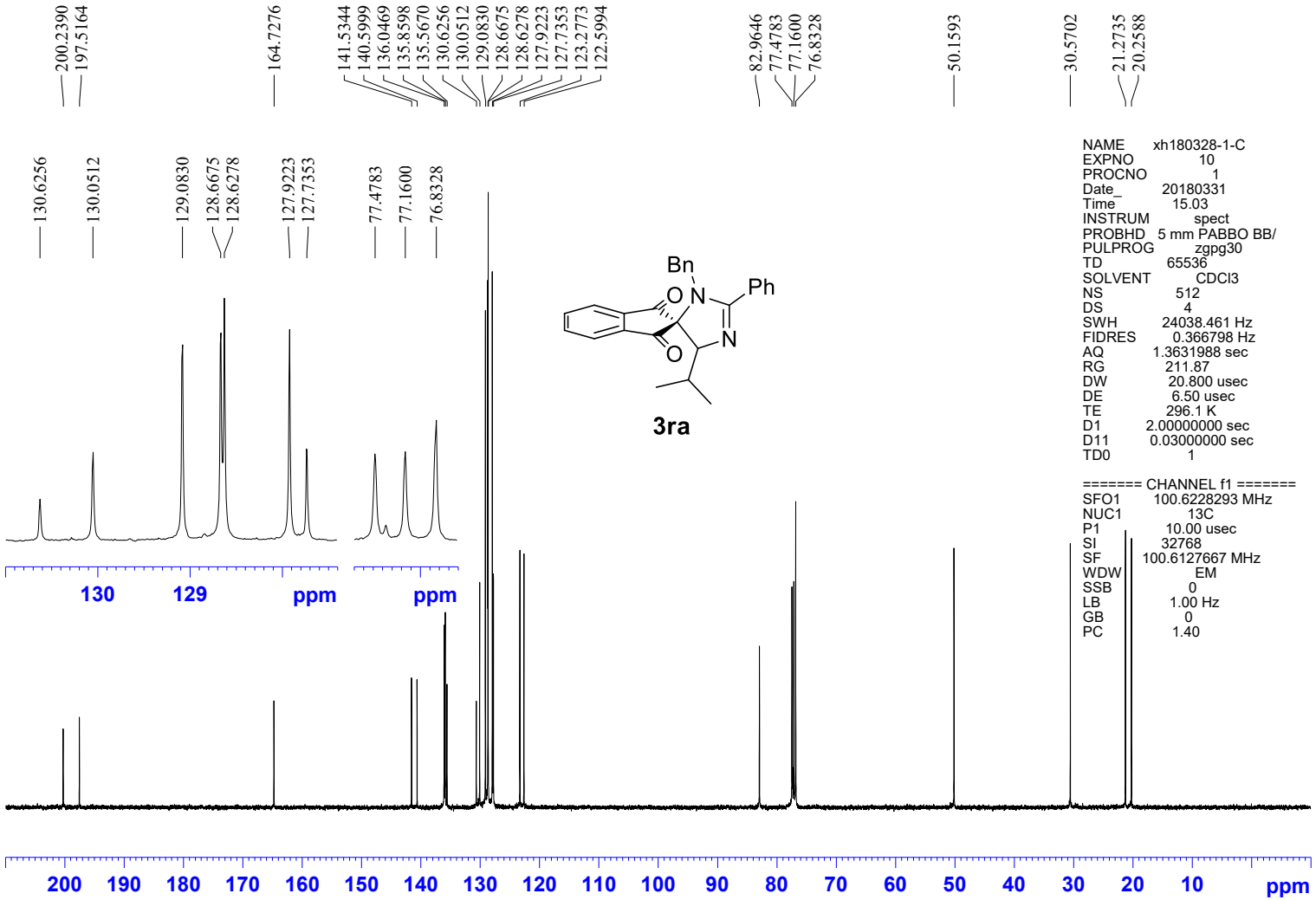


Figure S44. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ra

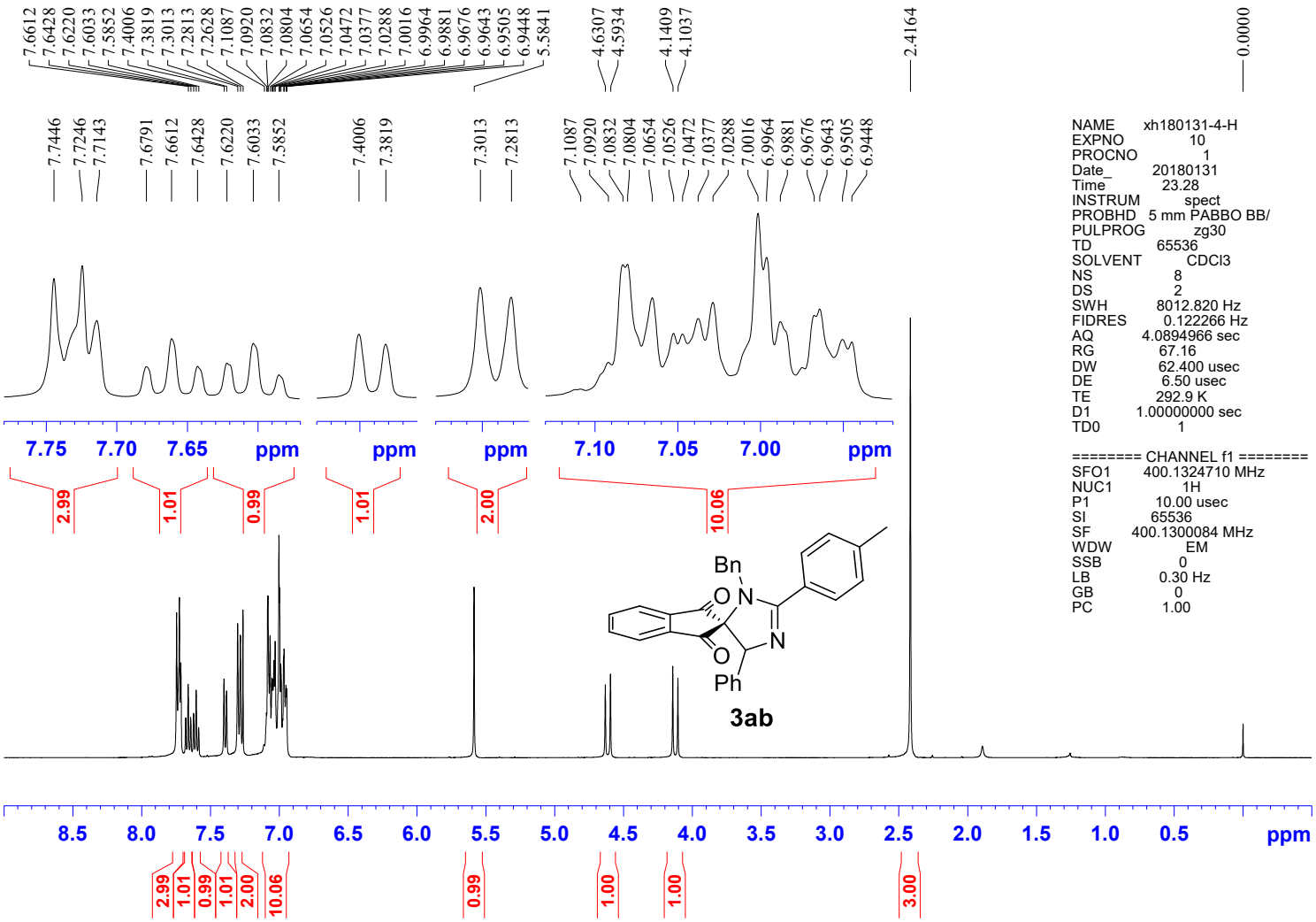


Figure S45. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ab

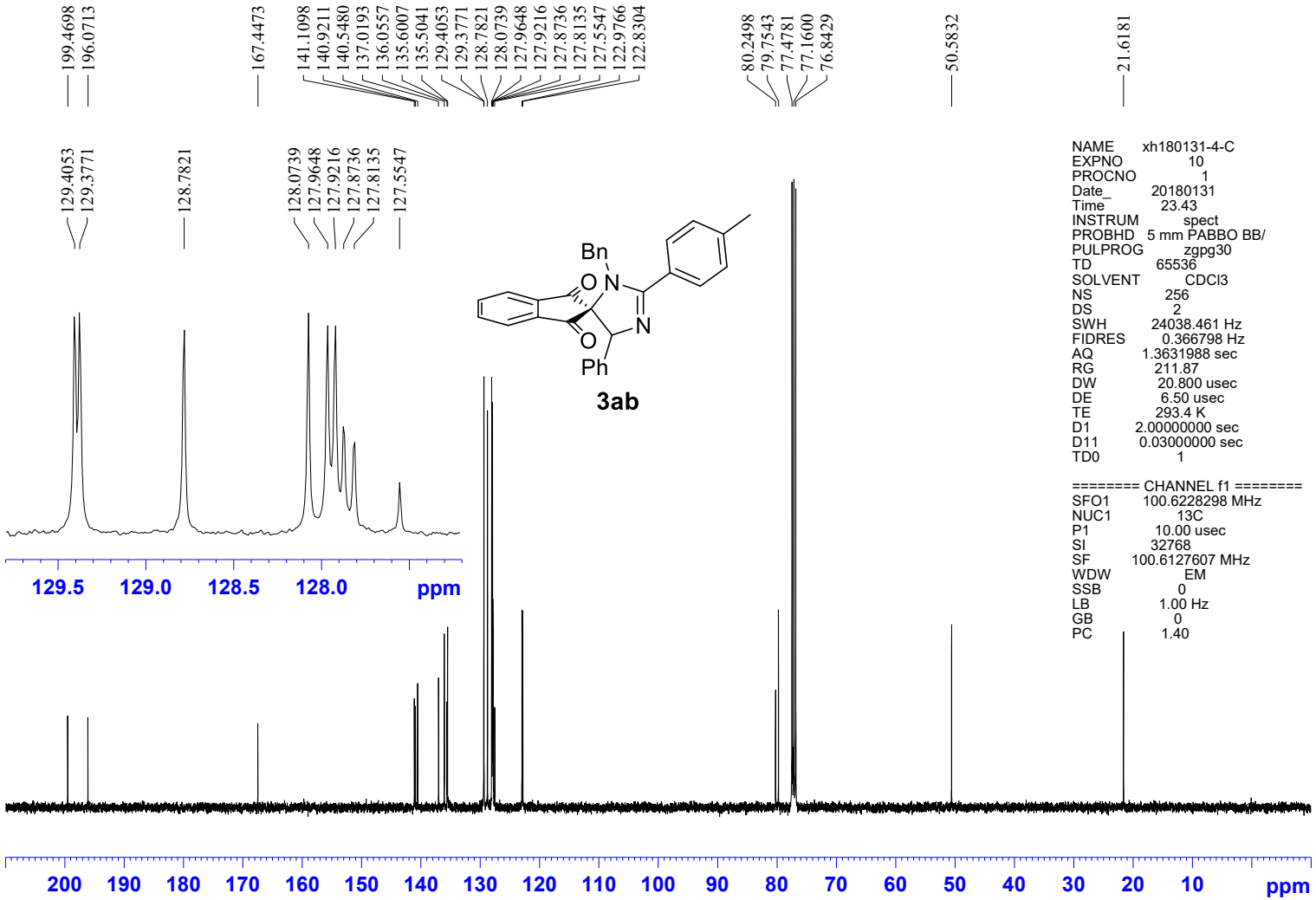


Figure S46. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ab

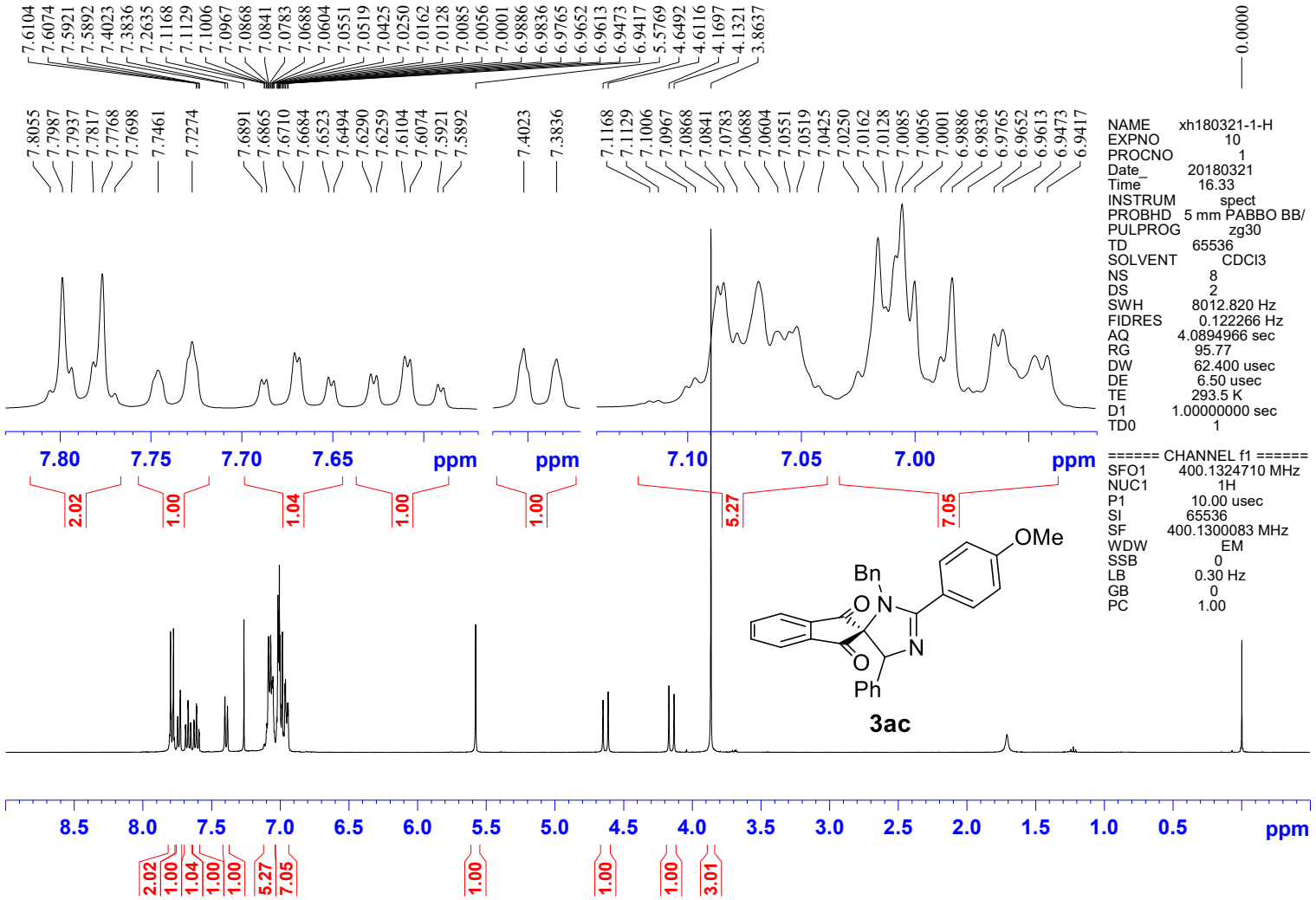


Figure S47. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ac

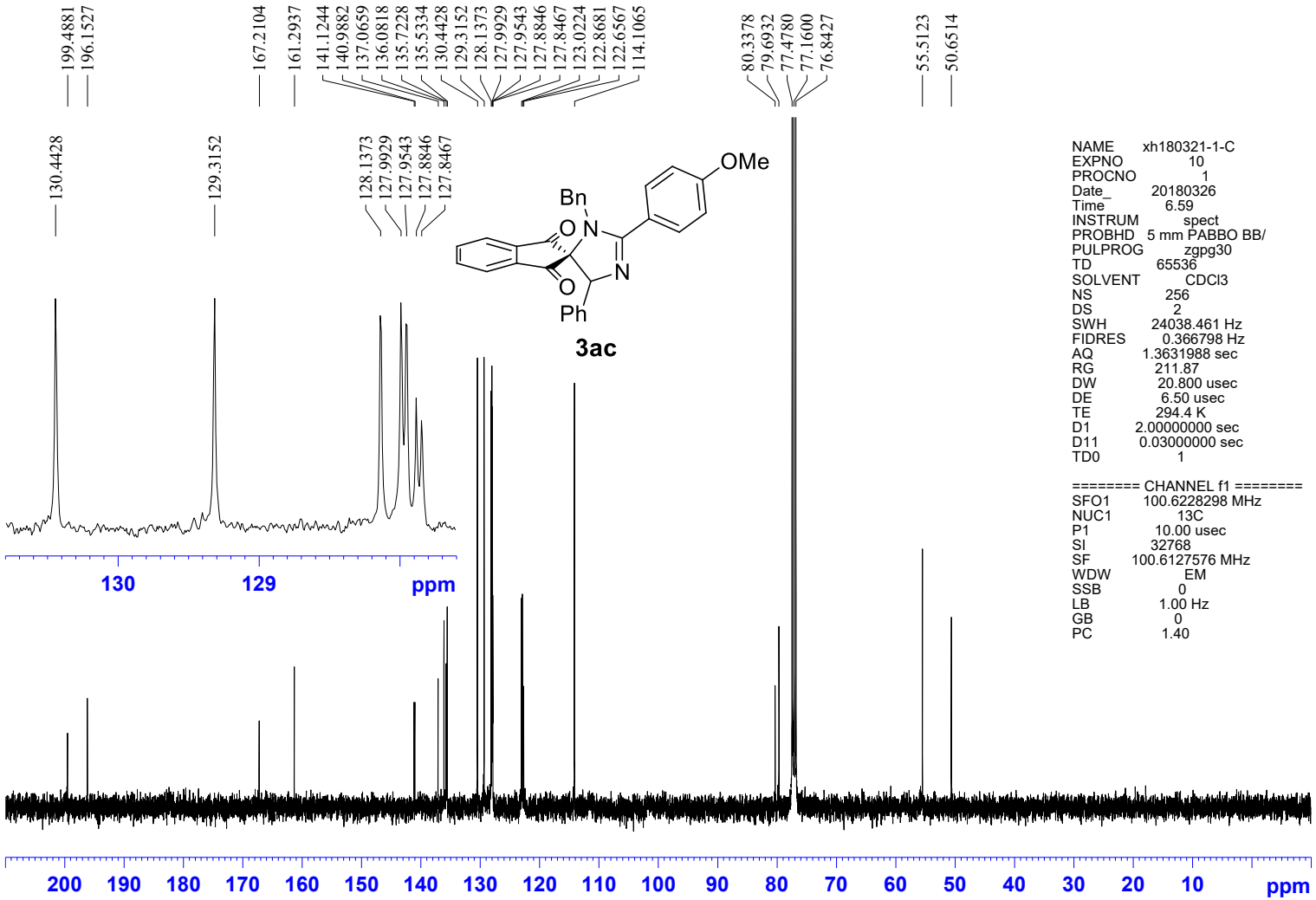


Figure S48. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ac

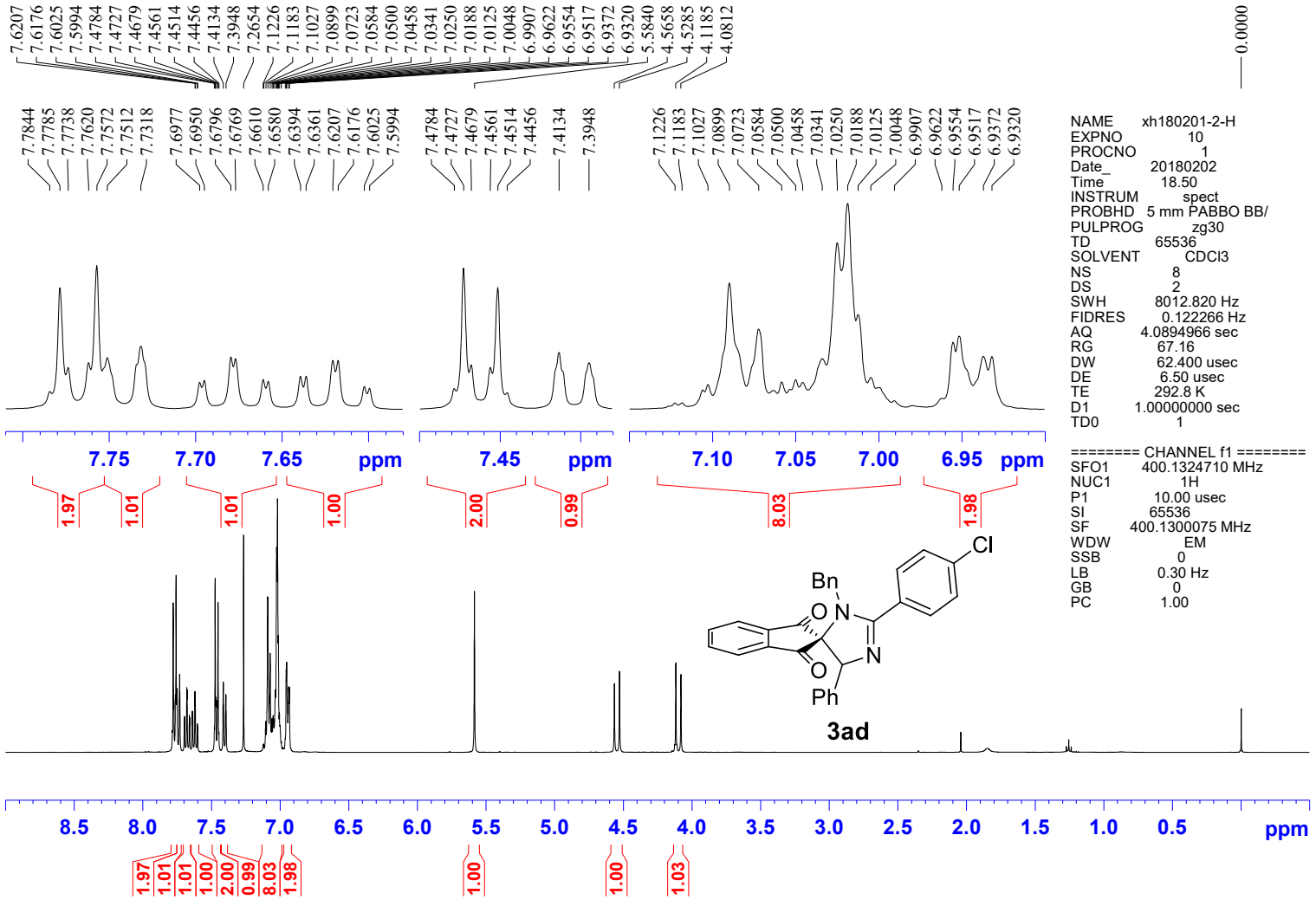


Figure S49. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ad

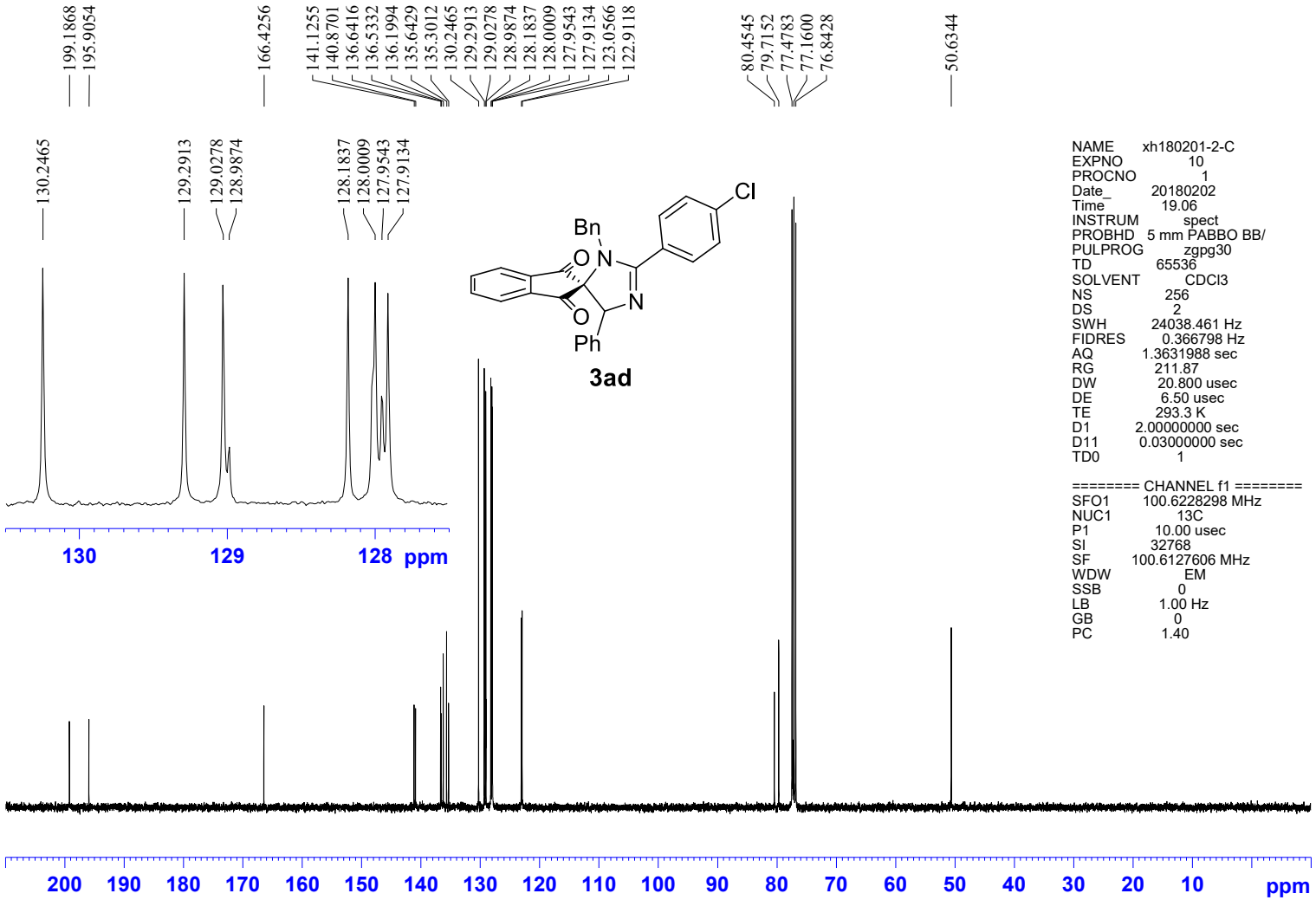


Figure S50. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ad

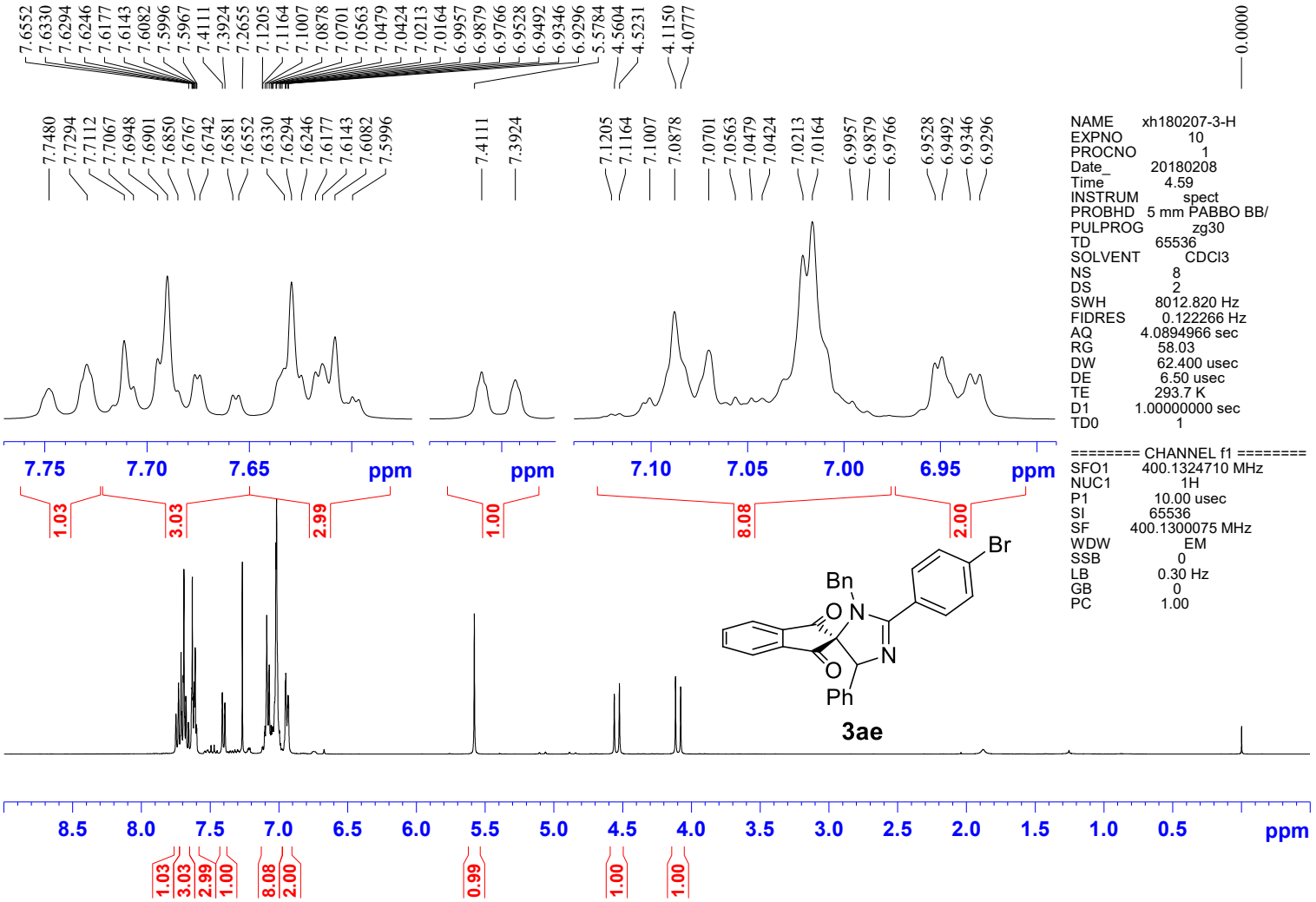


Figure S51. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ae

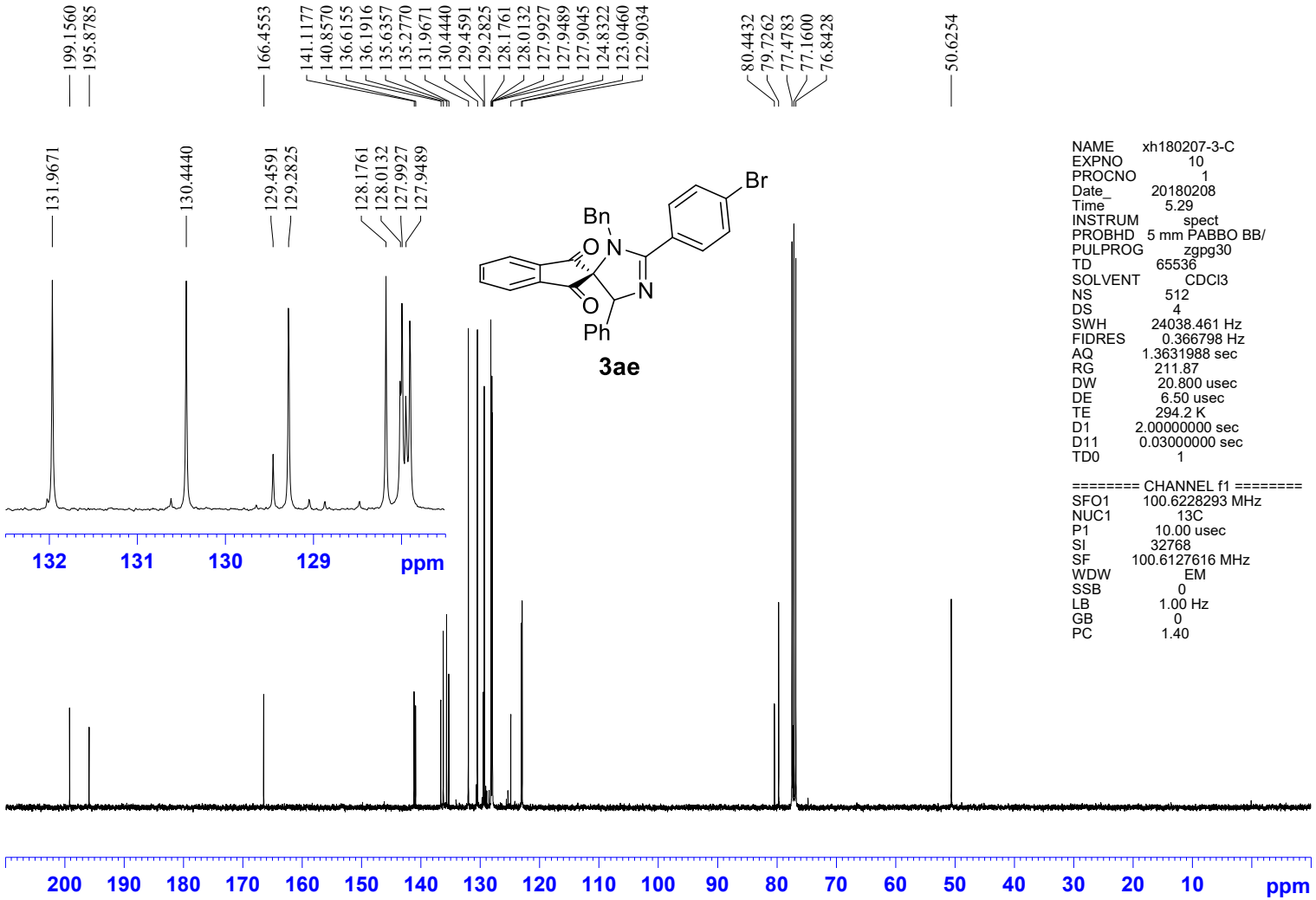


Figure S52. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ae

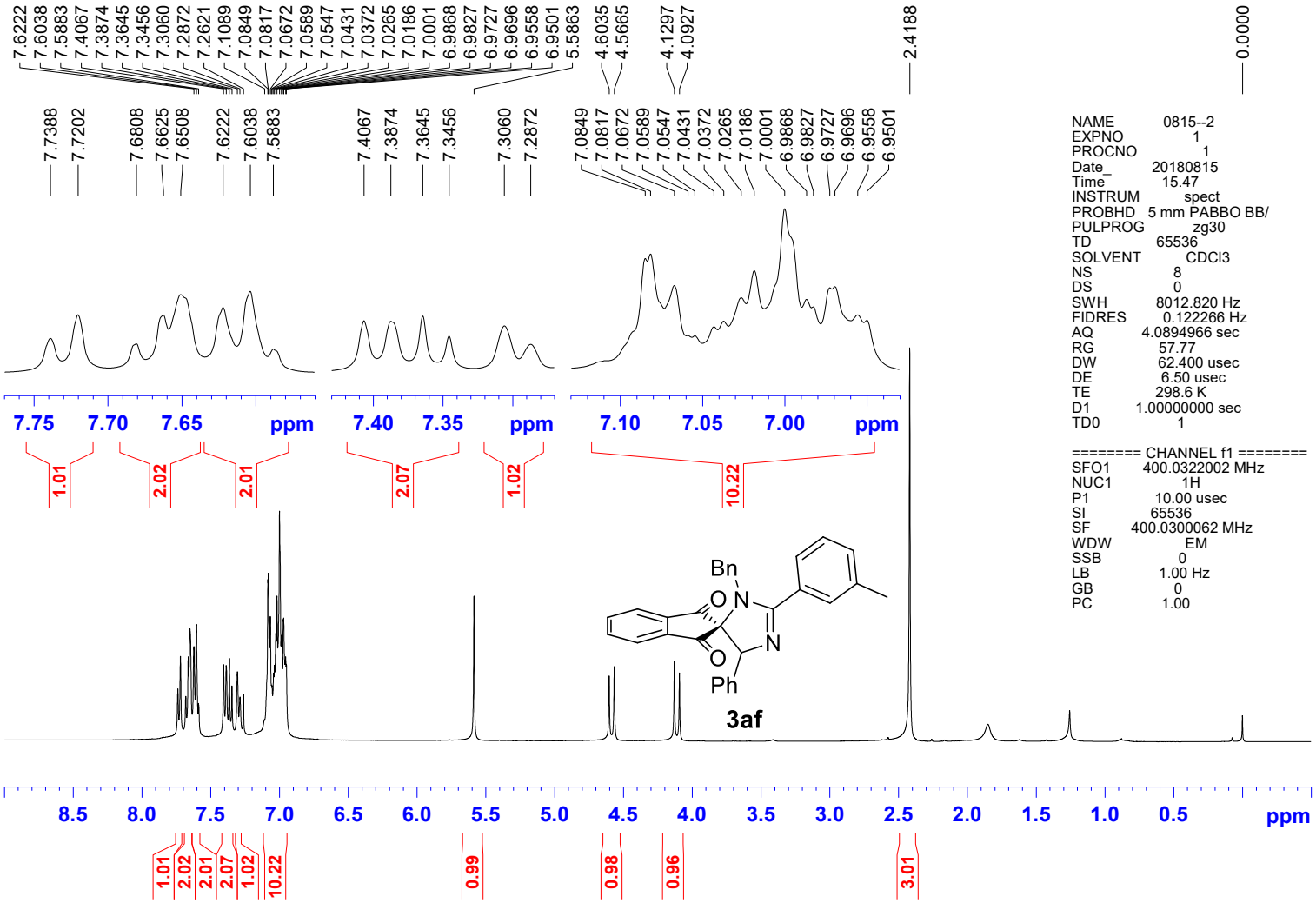


Figure S53. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3af

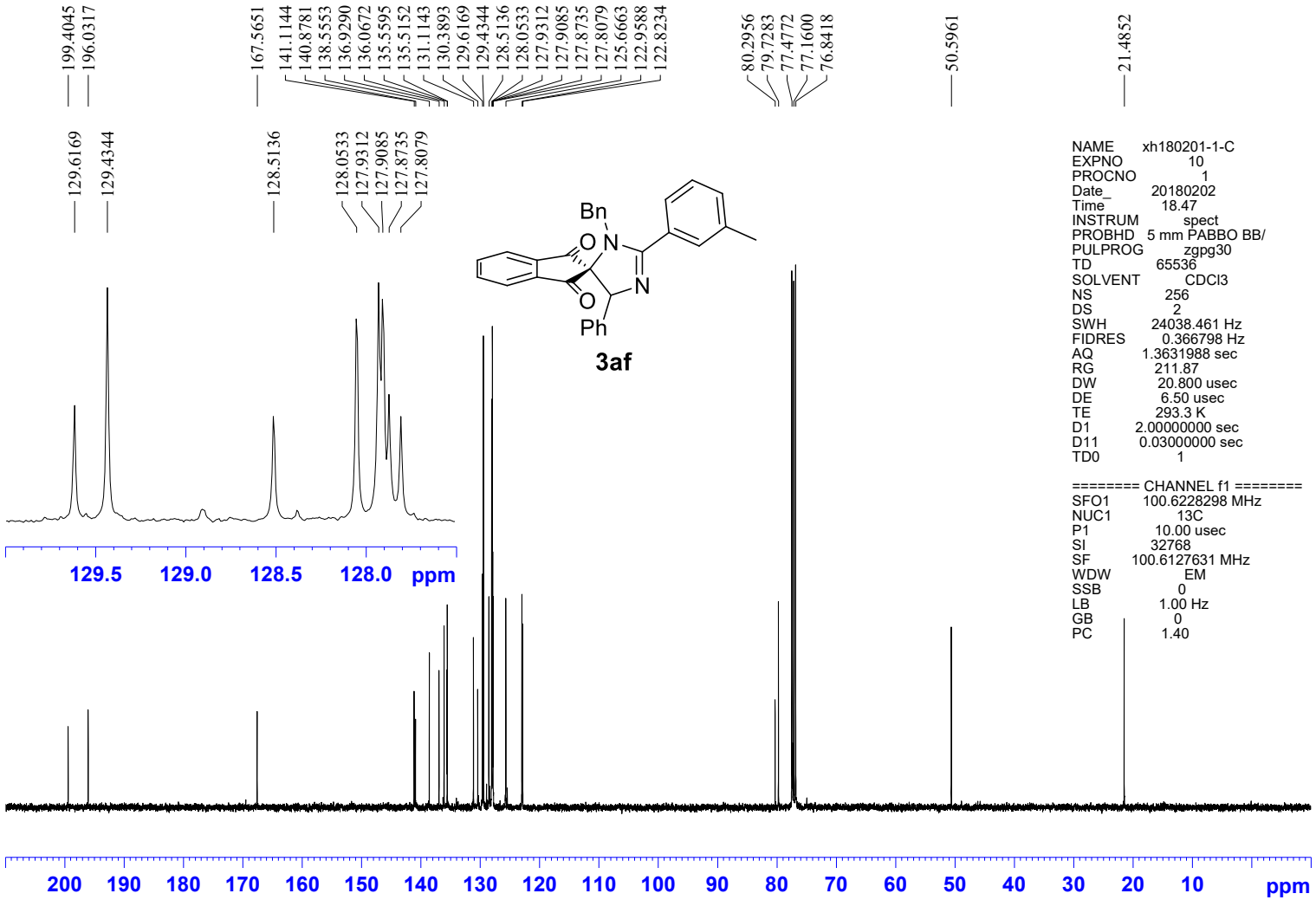


Figure S54. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3af

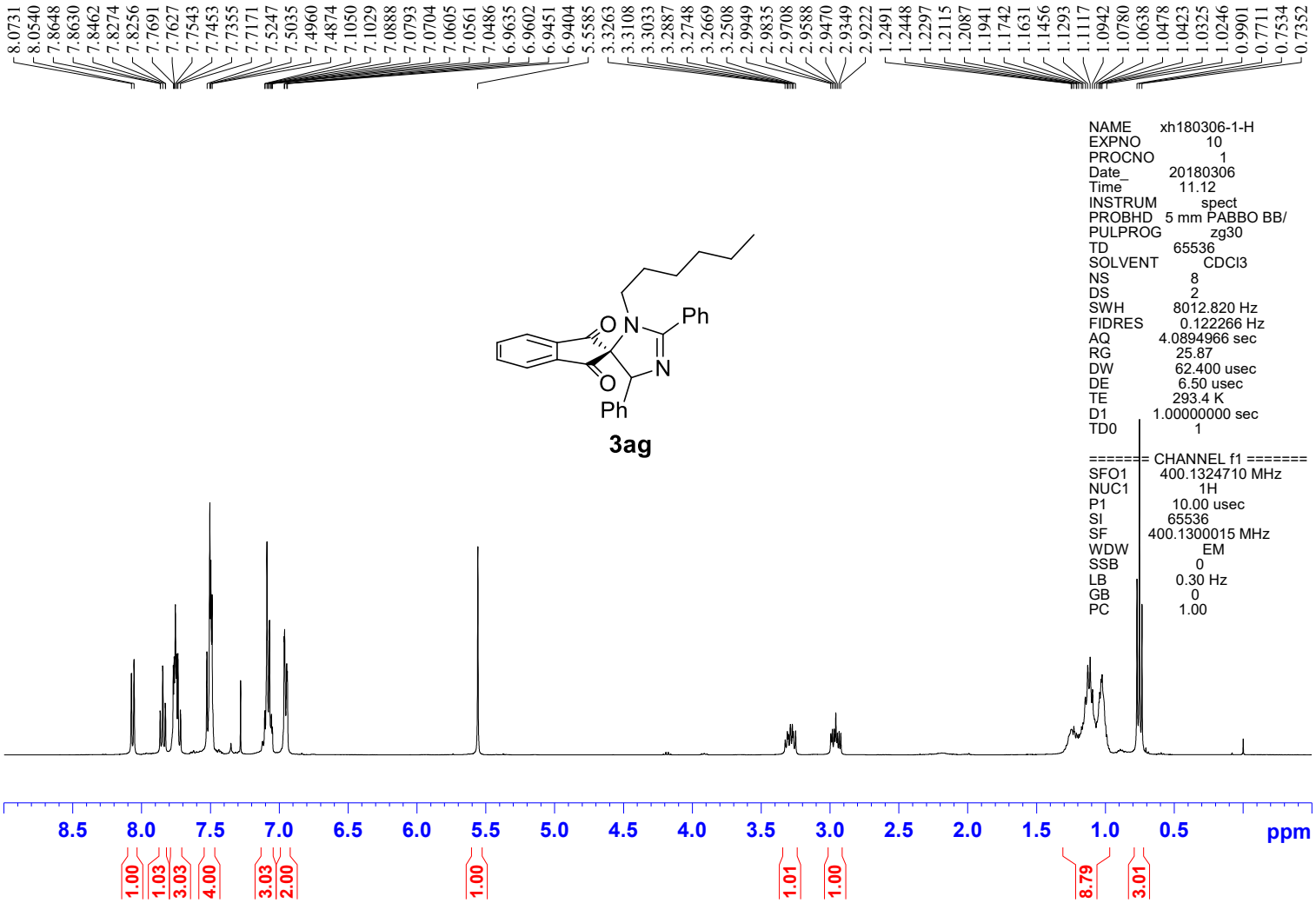


Figure S55. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ag

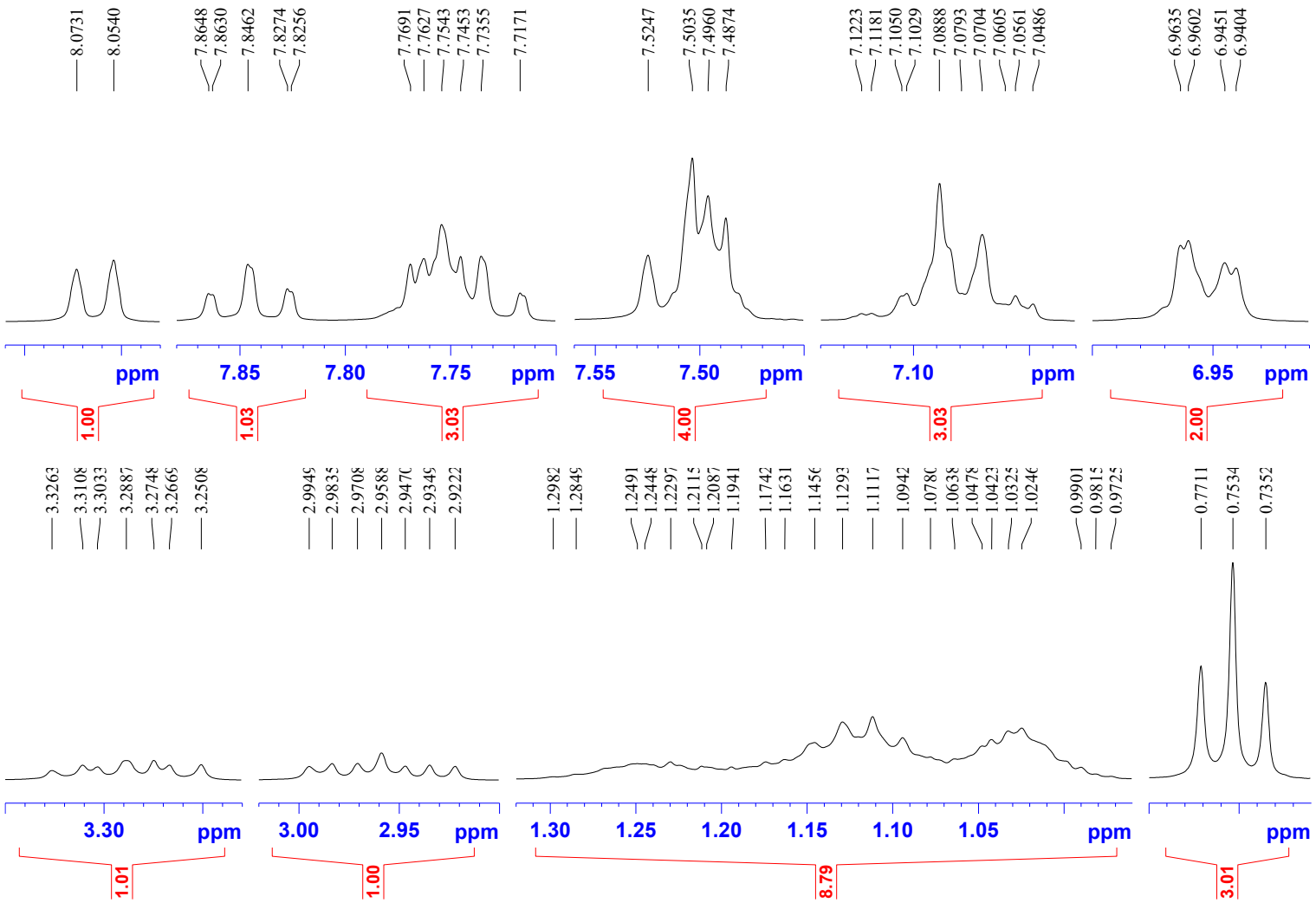


Figure S56. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ag

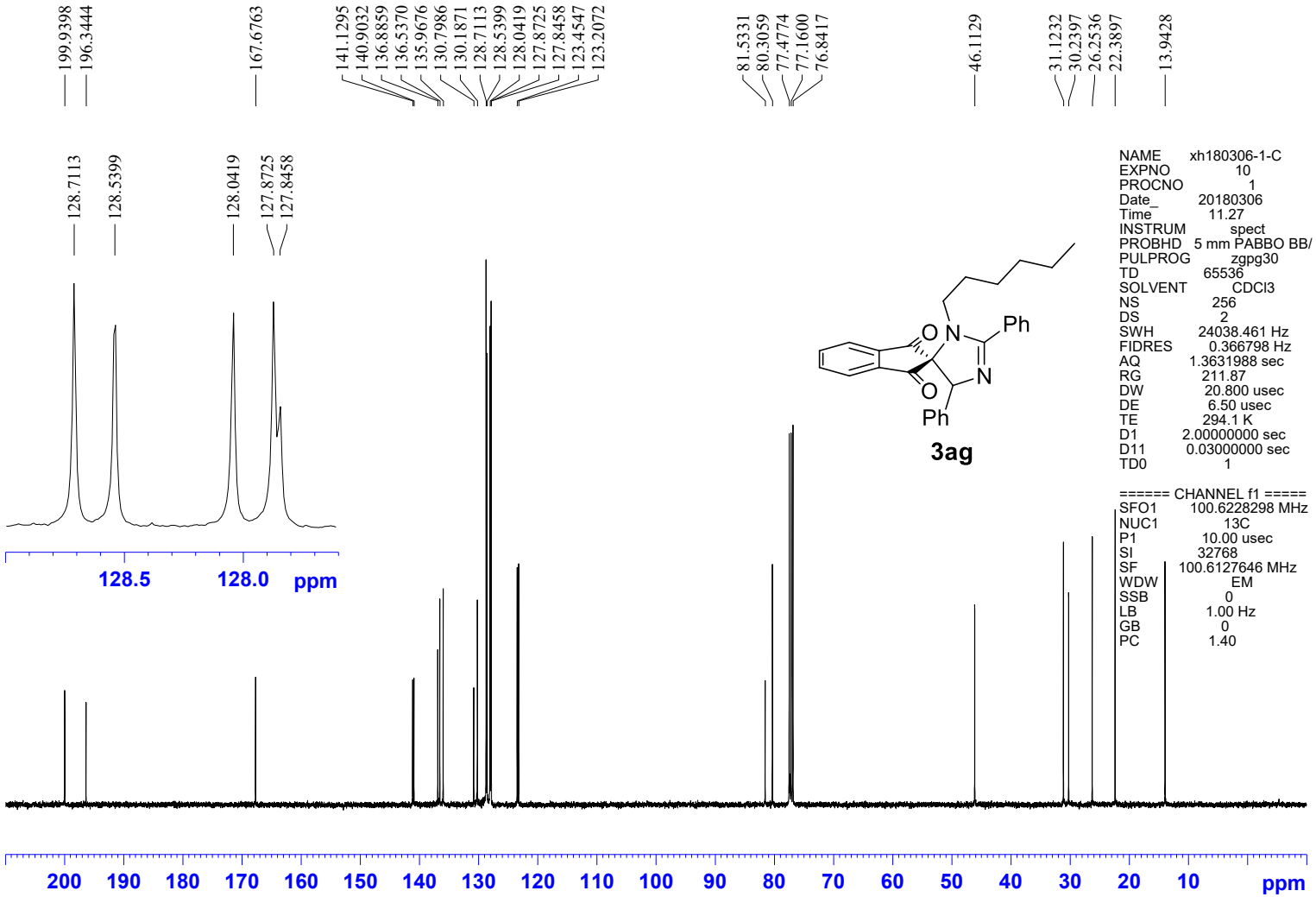


Figure S57. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ag

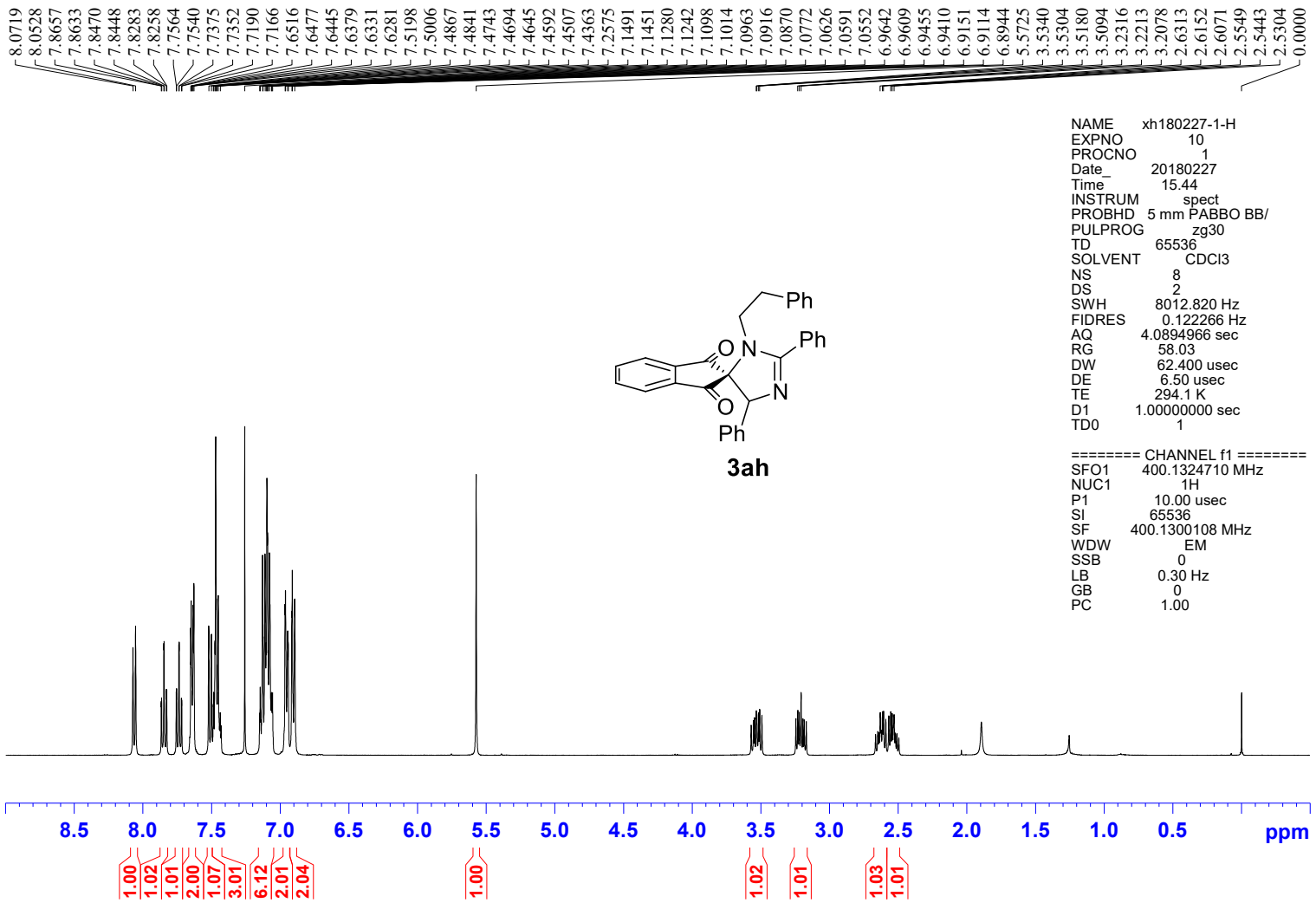


Figure S58. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ah

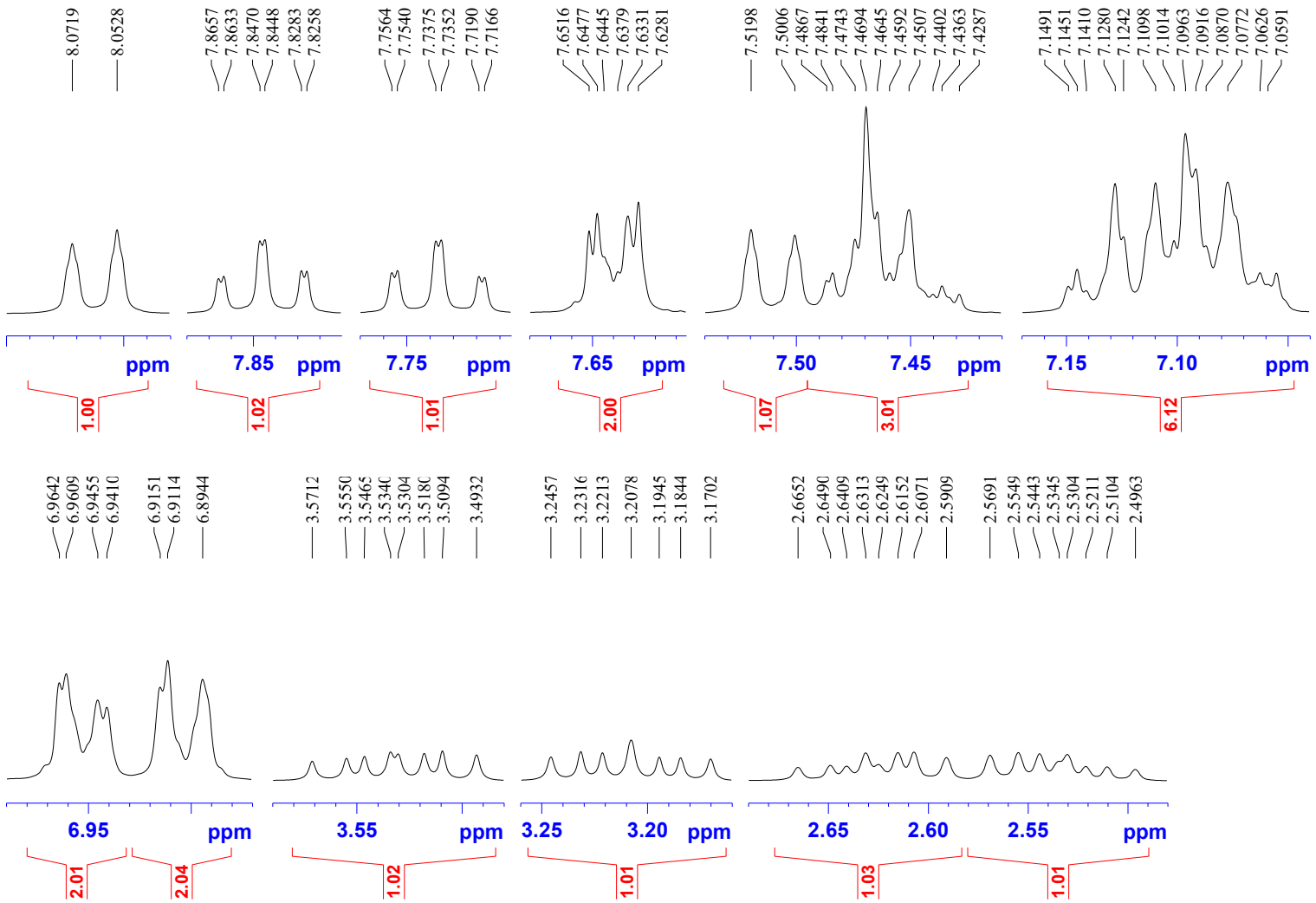


Figure S59. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ah

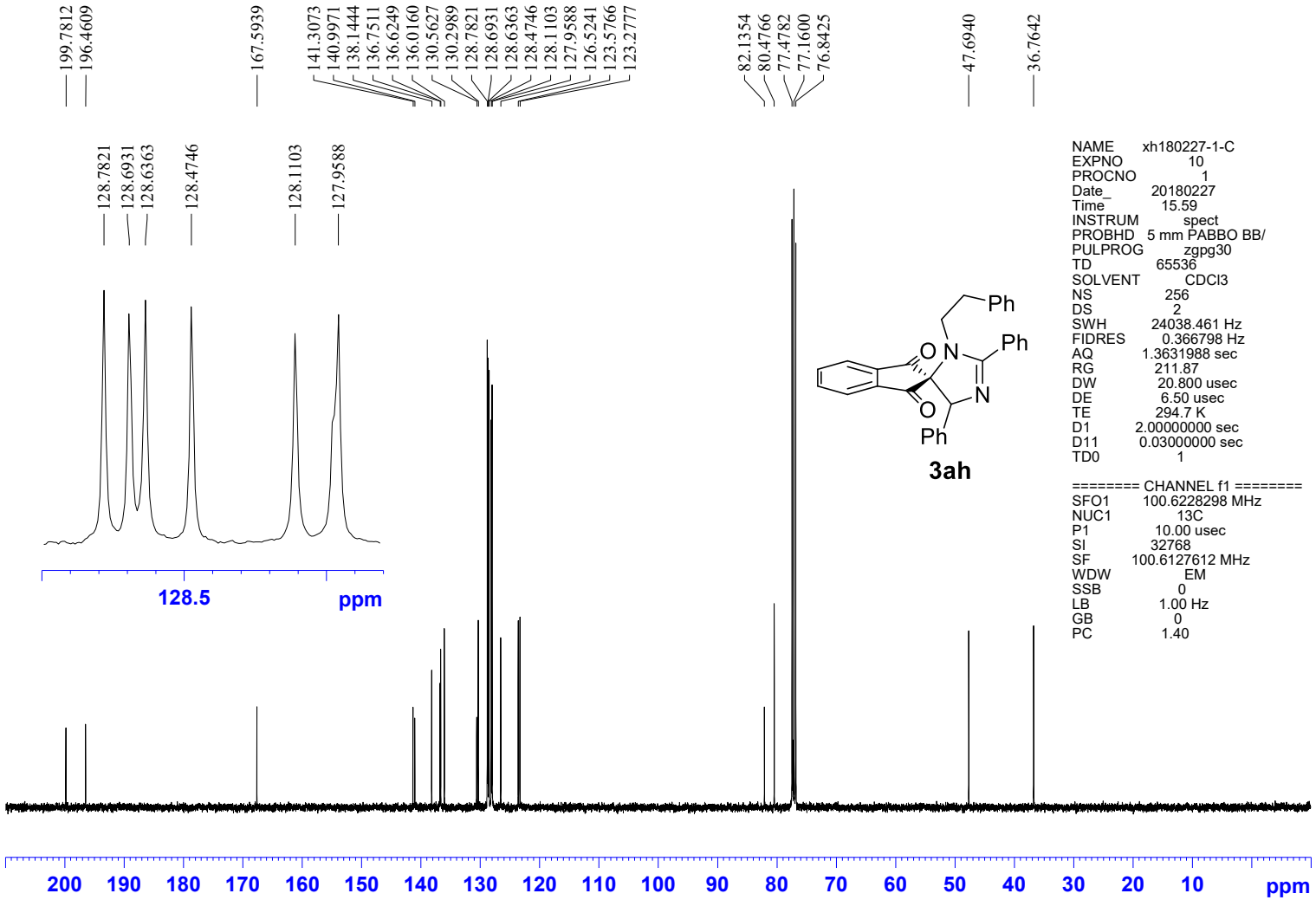


Figure S60. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ah

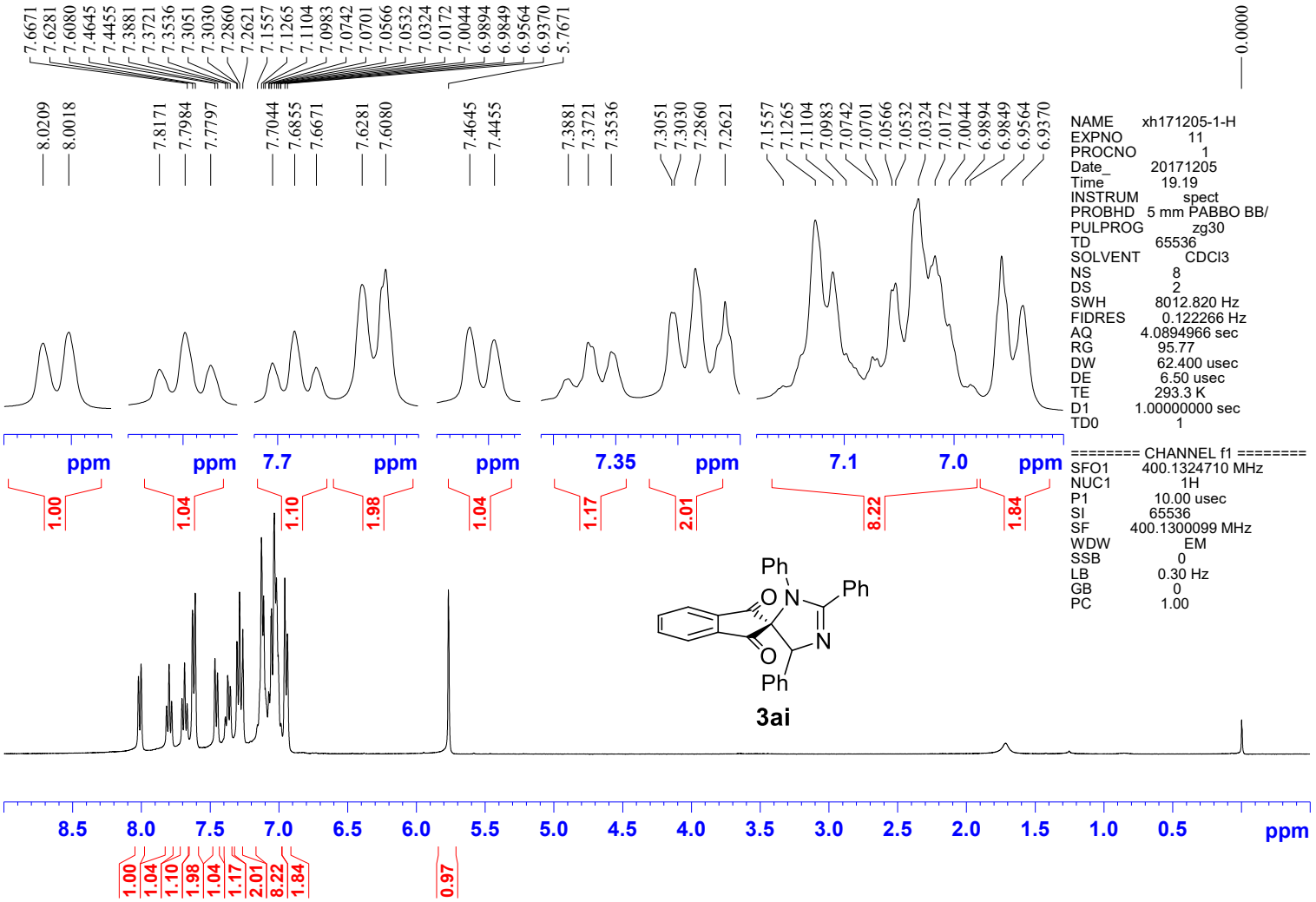


Figure S61. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ai

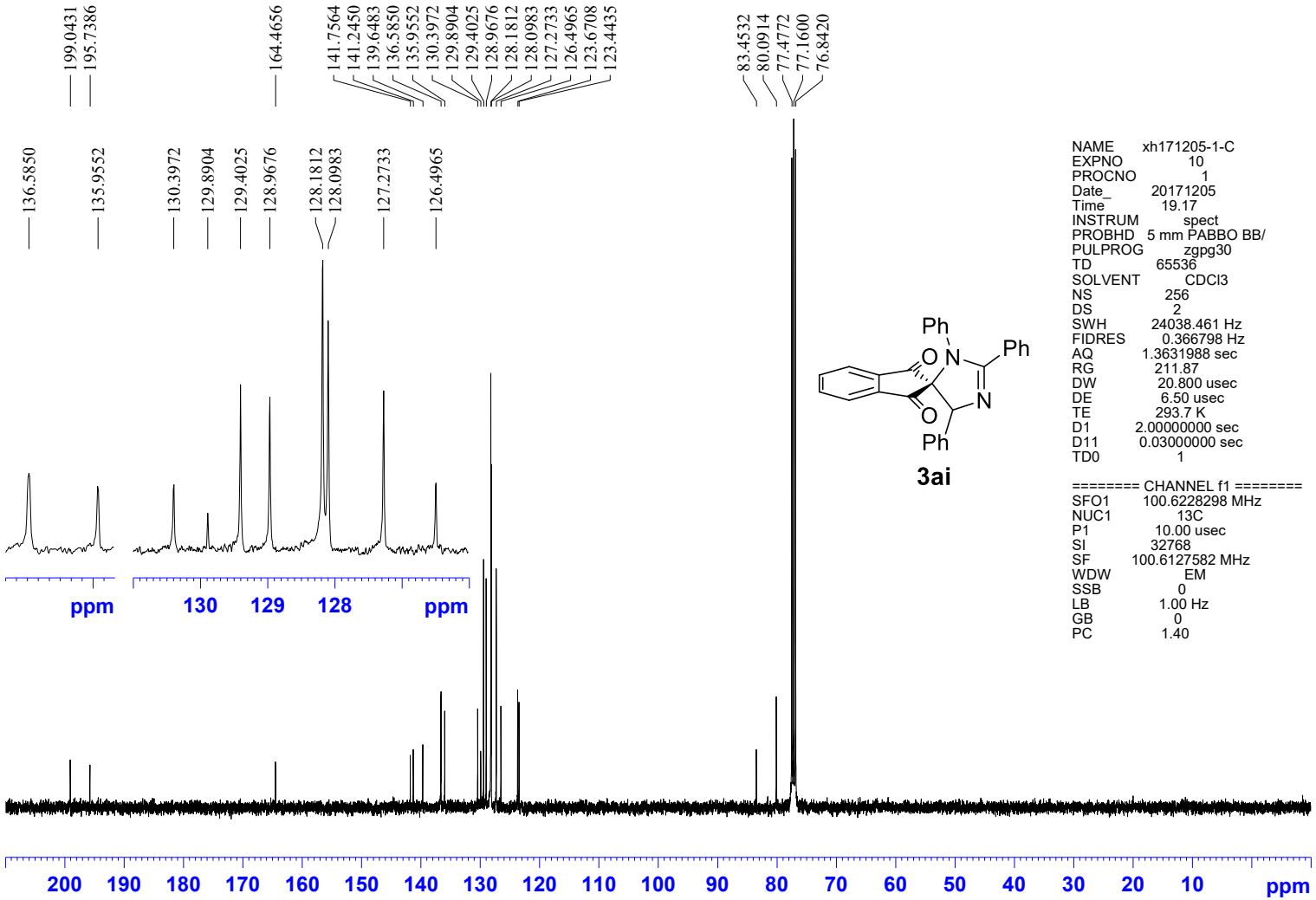


Figure S62. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ai

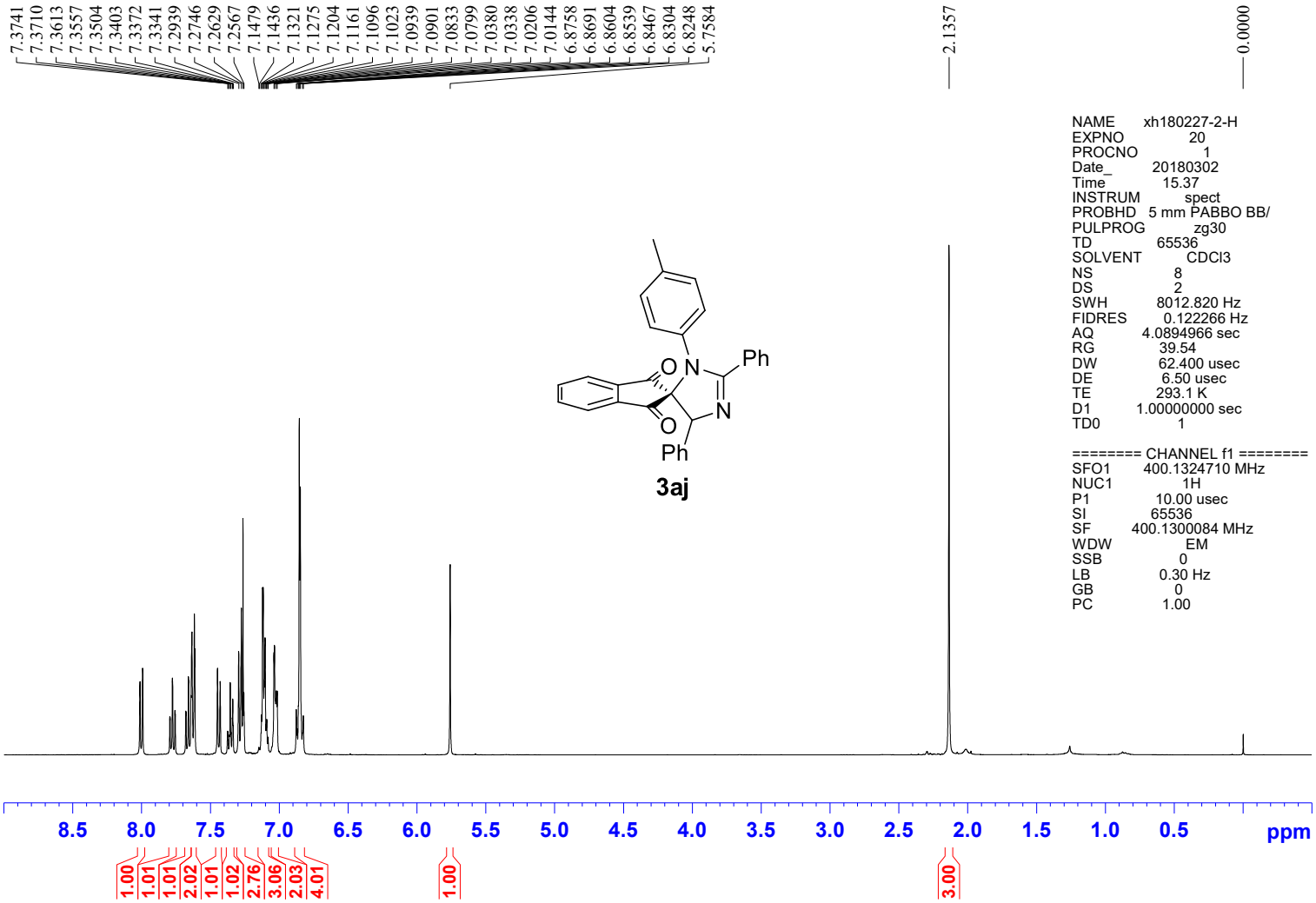


Figure S63. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3aj

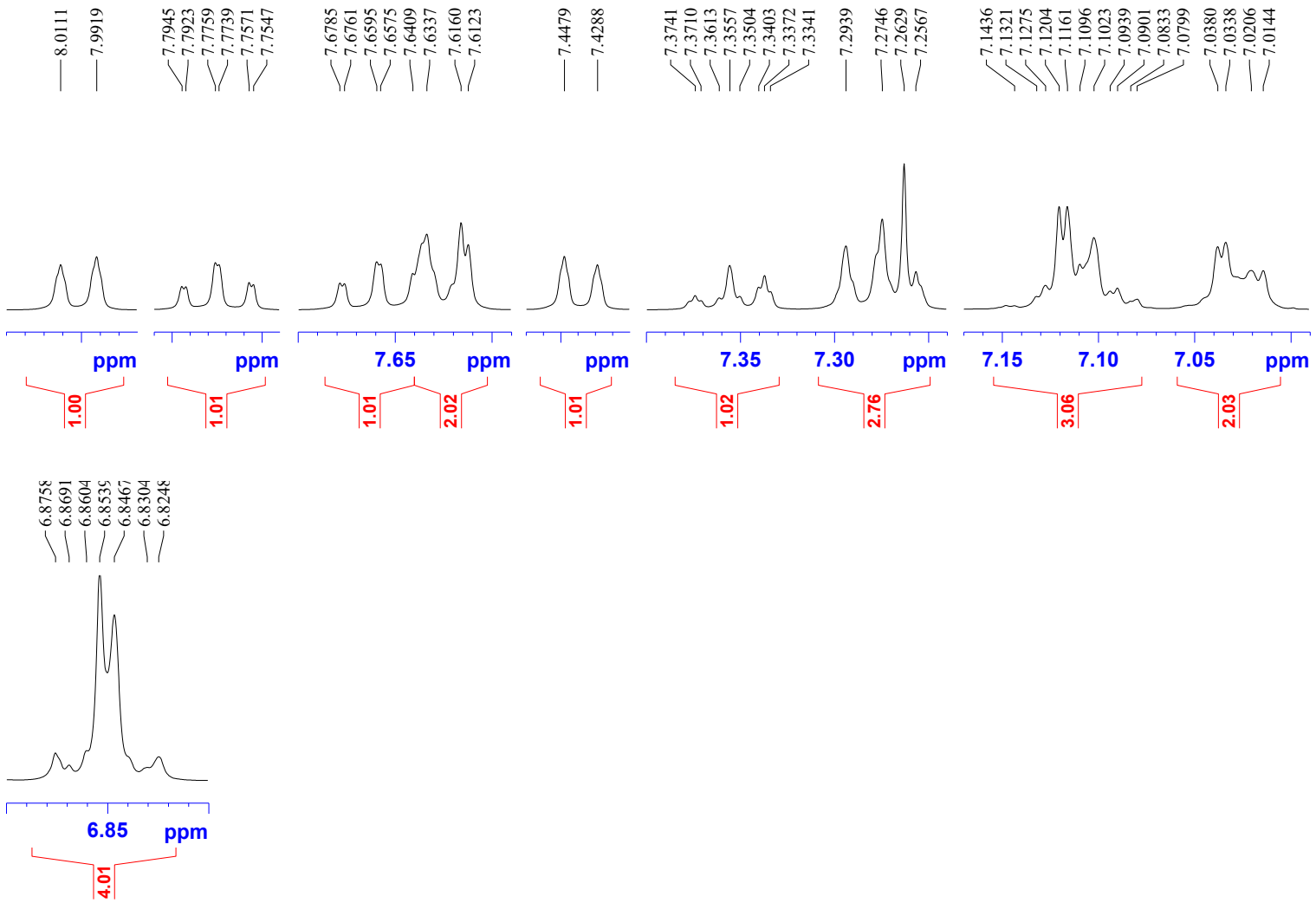


Figure S64. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3aj

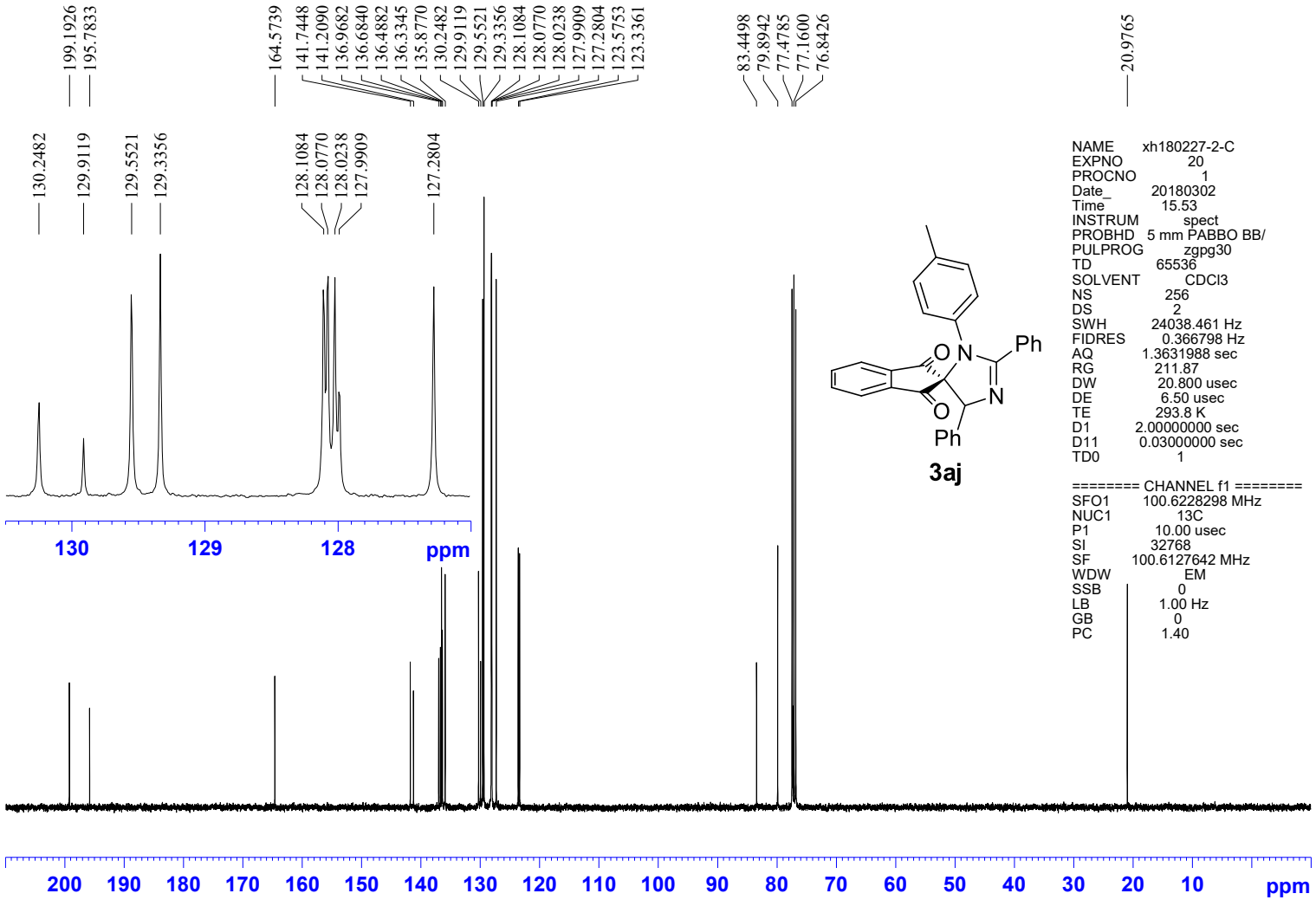


Figure S65. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3aj

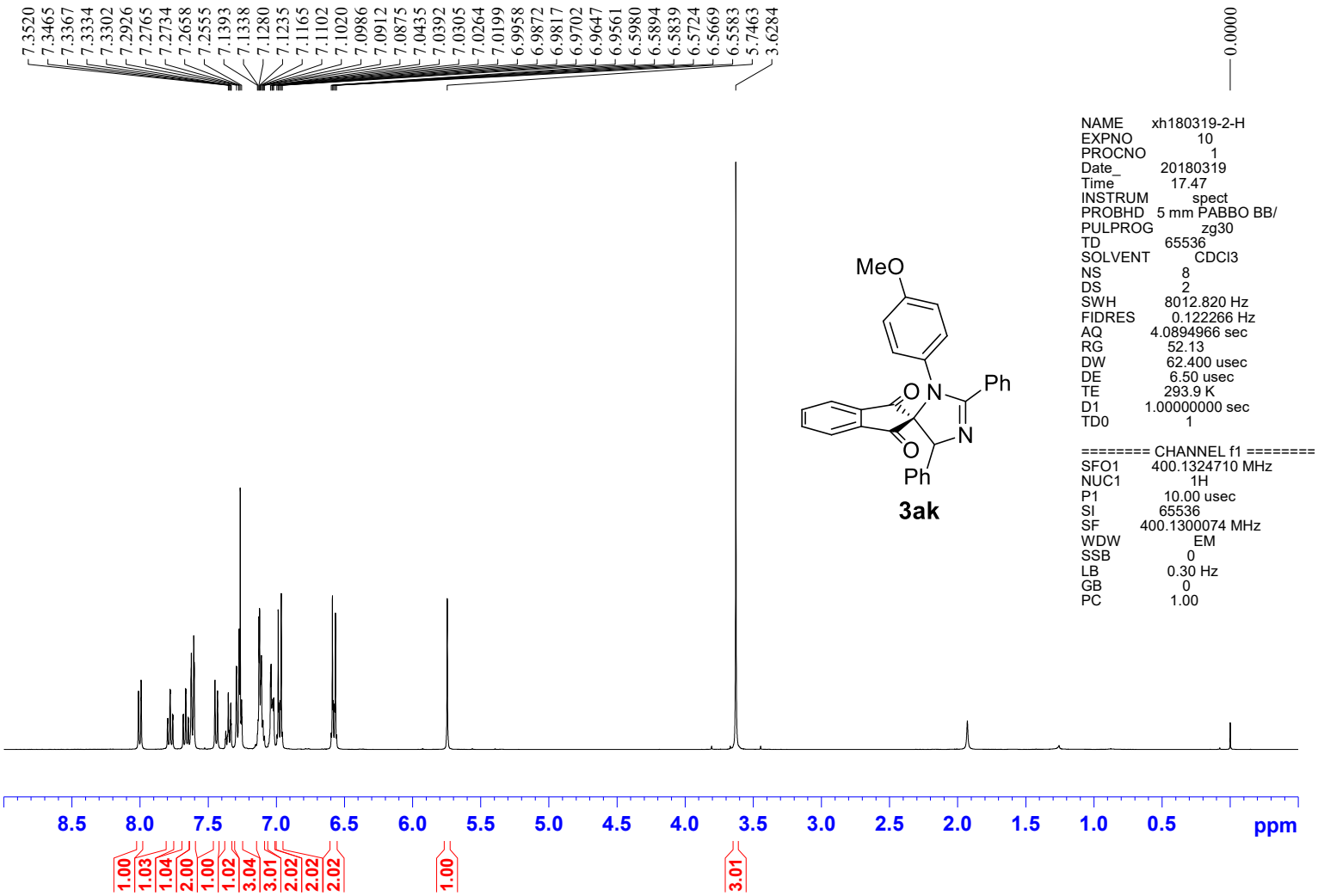


Figure S66. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ak

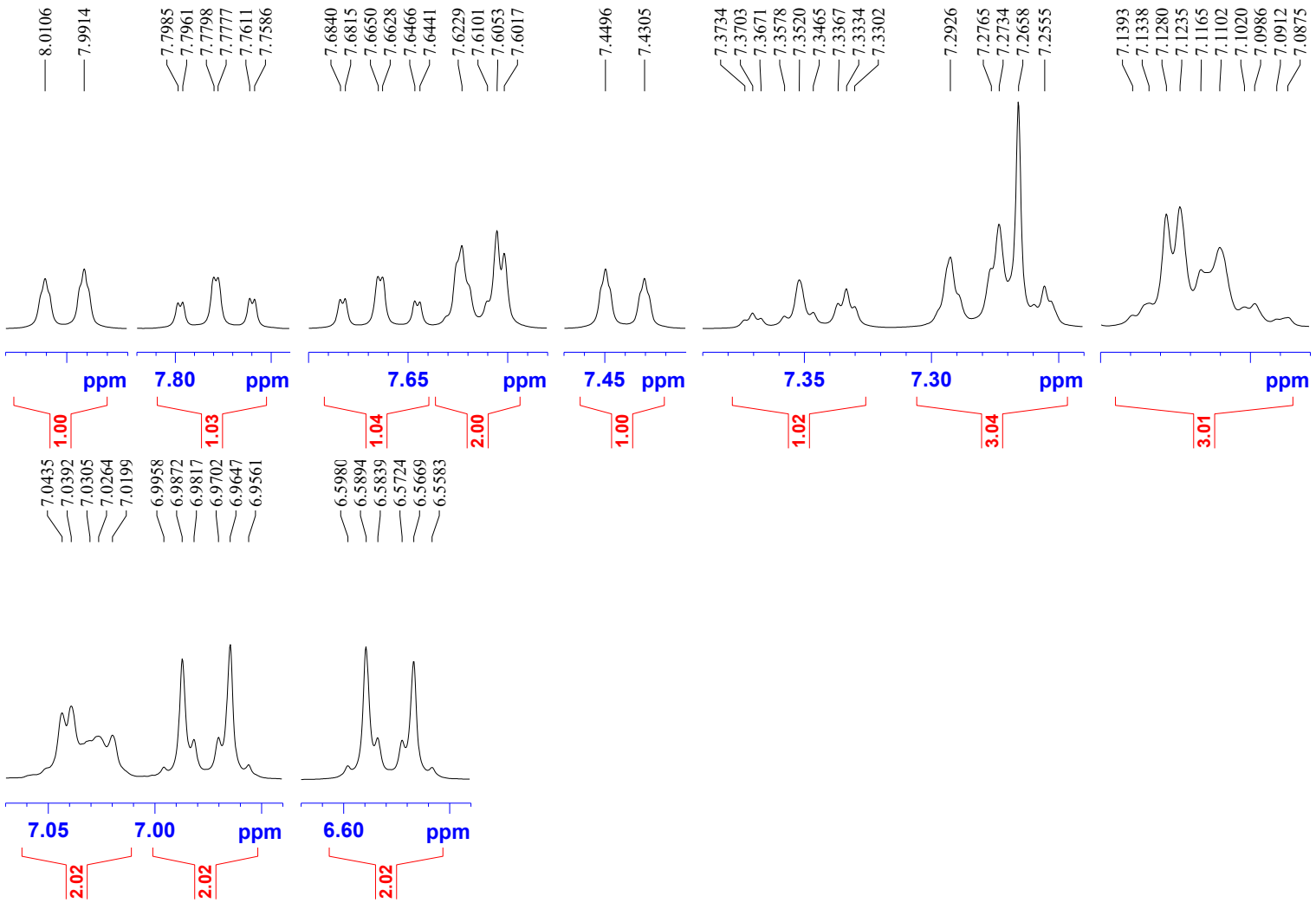


Figure S67. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3ak

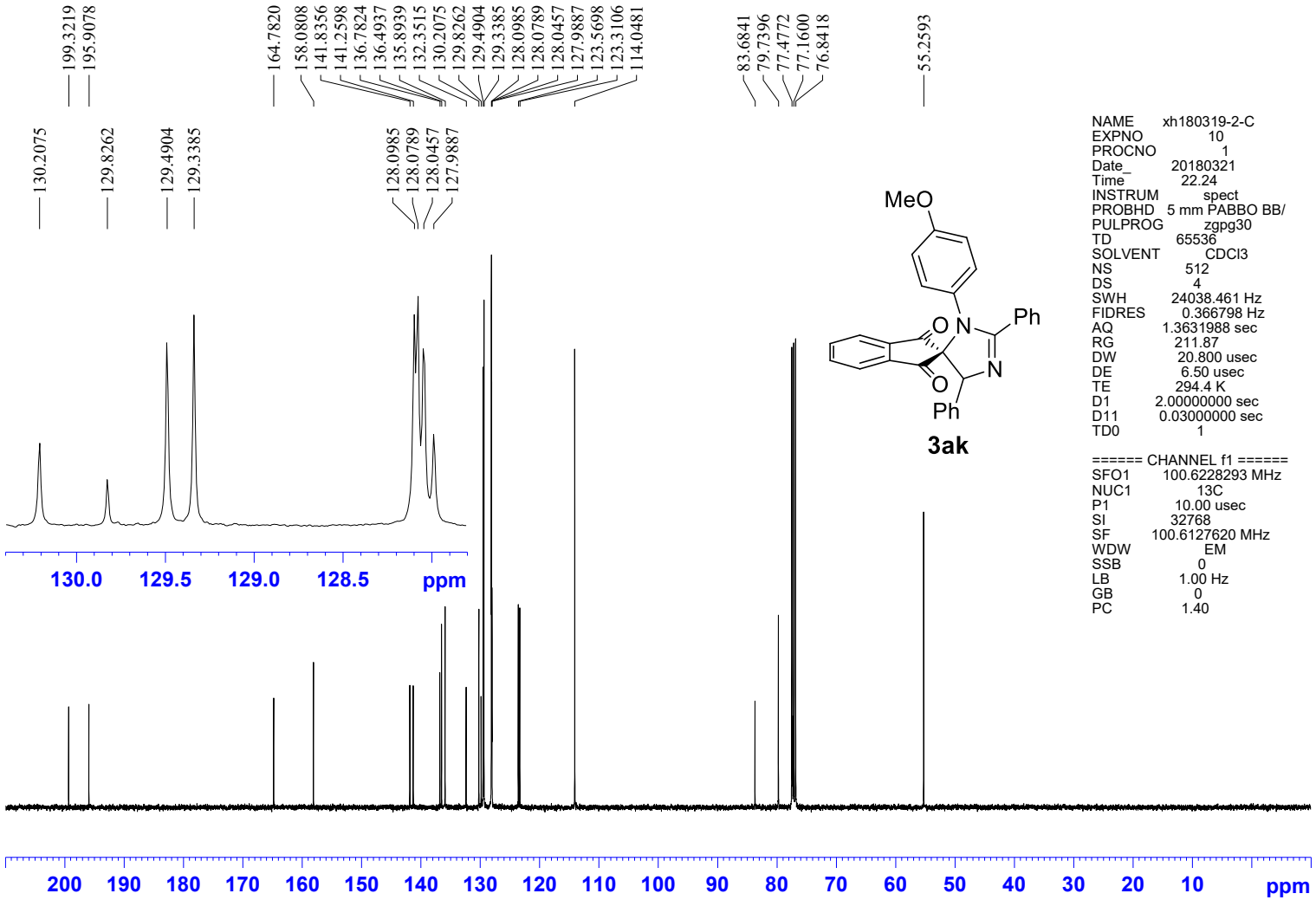


Figure S68. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3ak

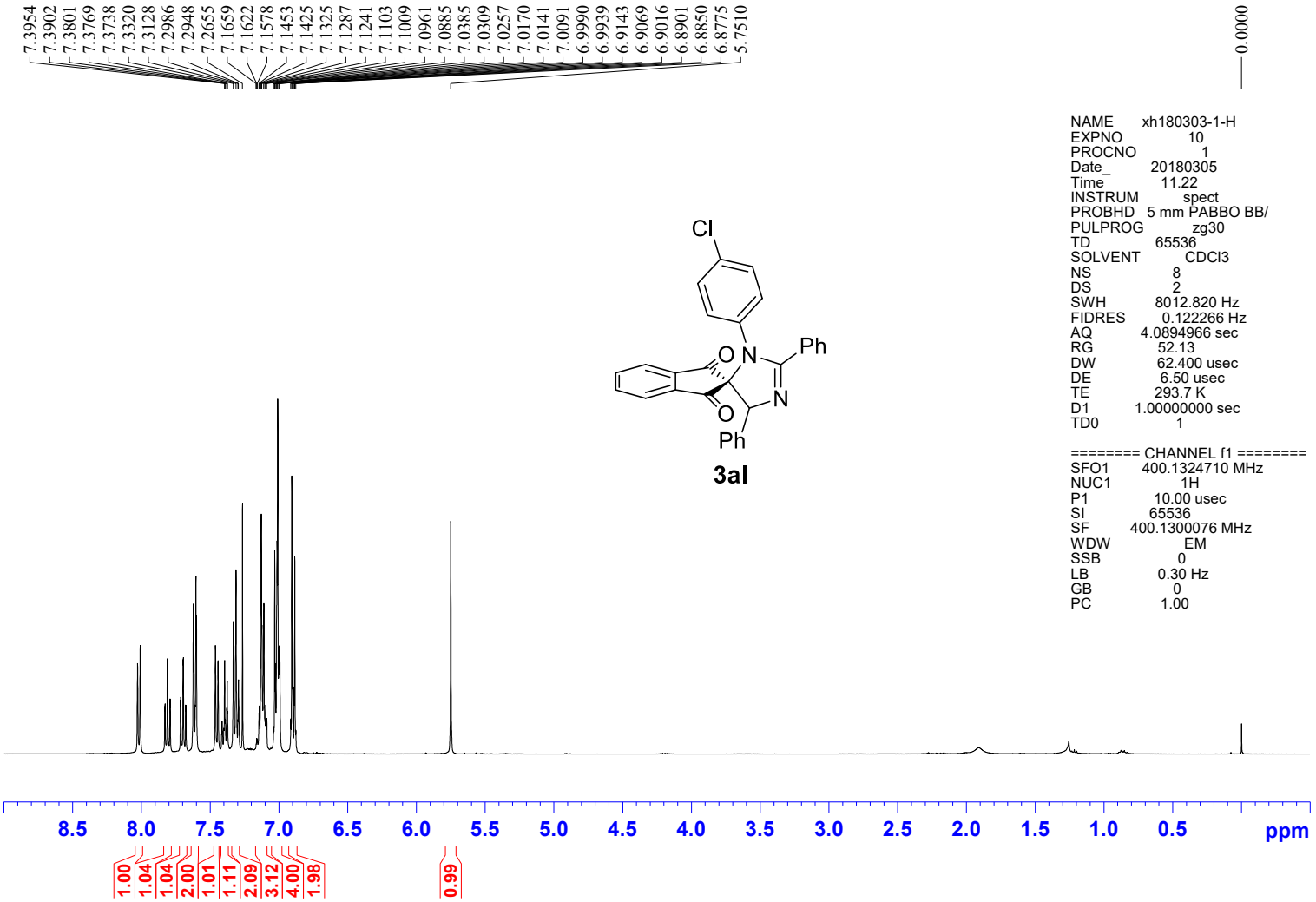


Figure S69. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3al

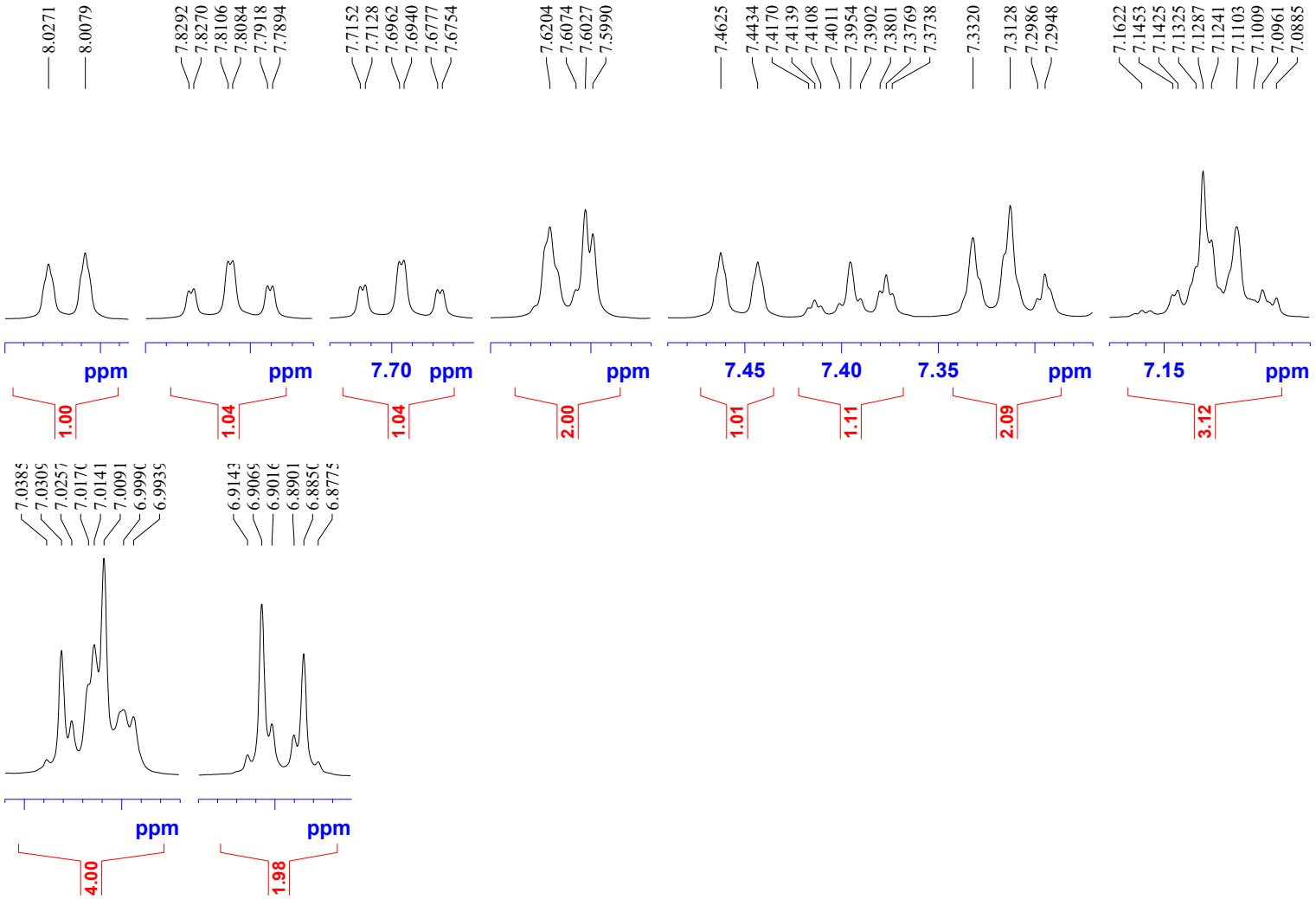


Figure S70. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3al

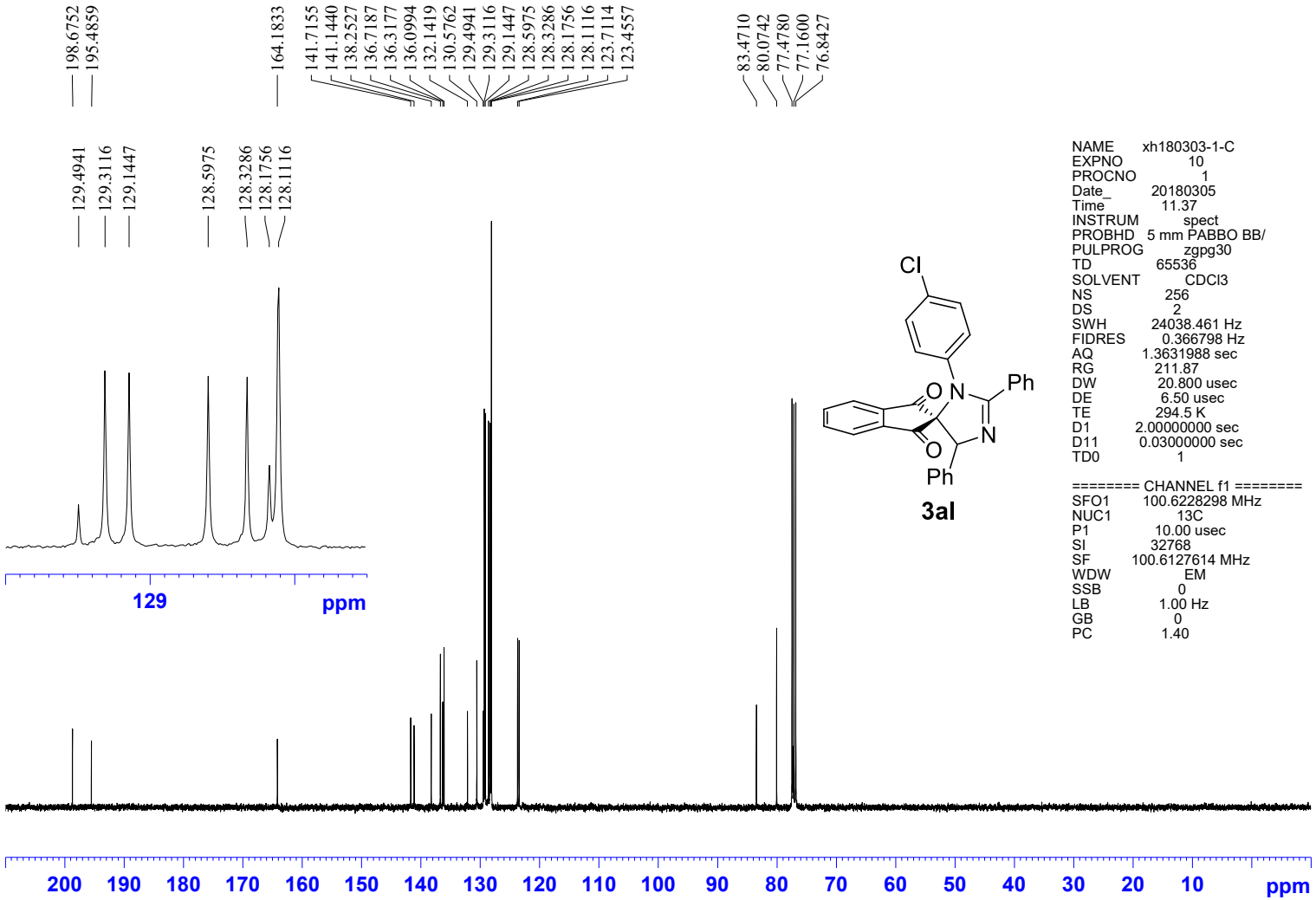


Figure S71. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3al

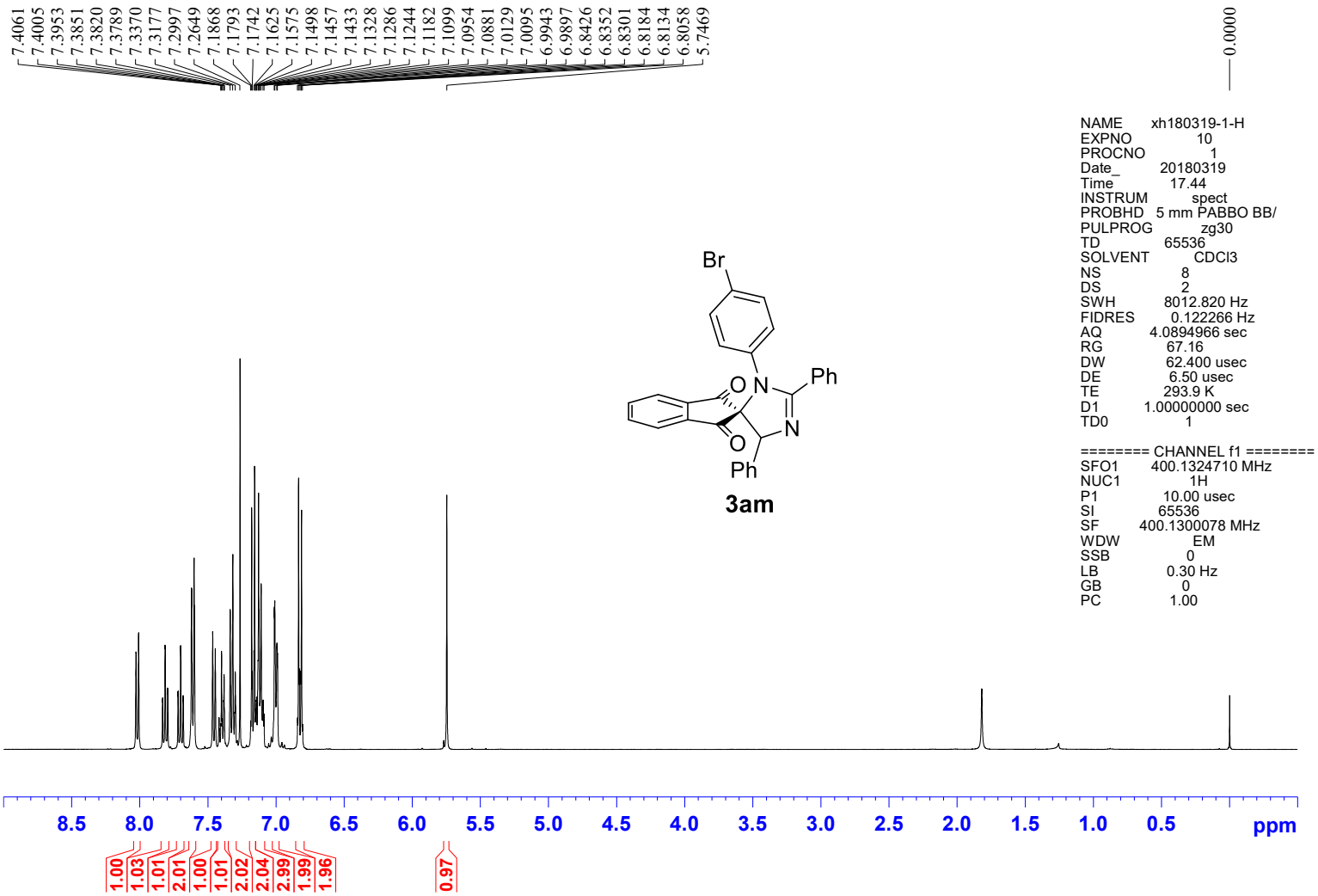


Figure S72. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3am

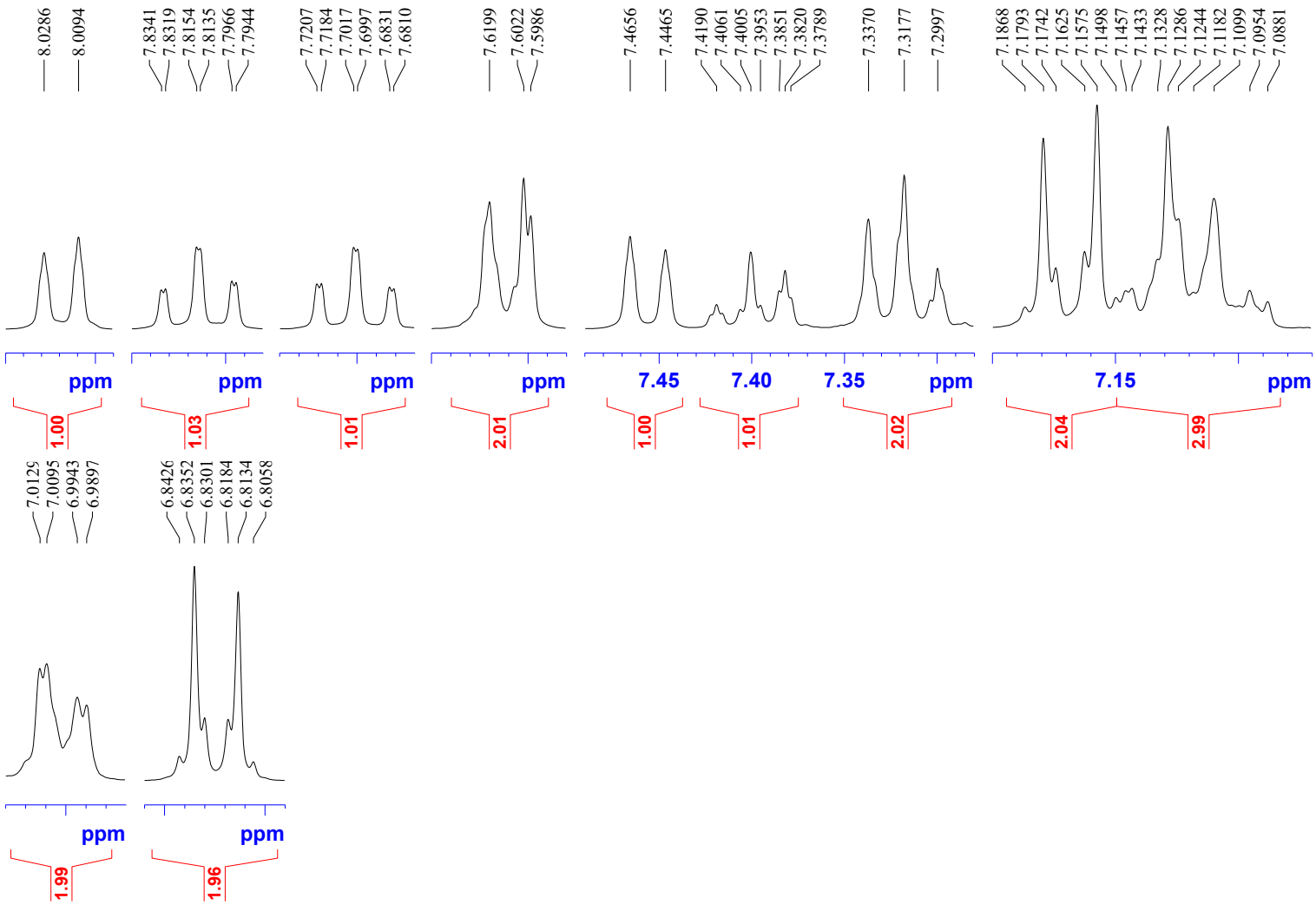


Figure S73. Expanded ^{1}H NMR (400 MHz, $CDCl_3$) of compound 3am

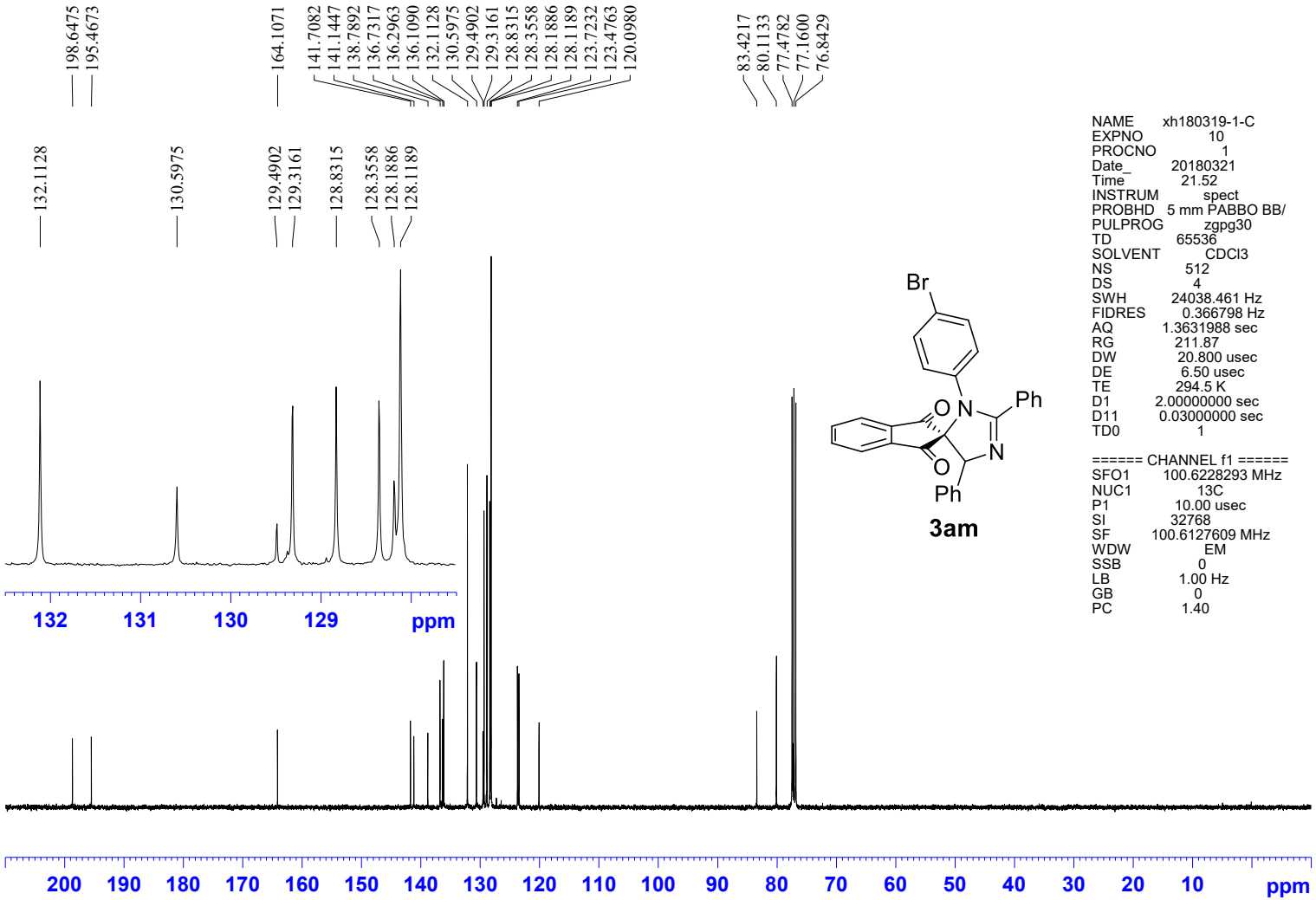


Figure S74. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 3am

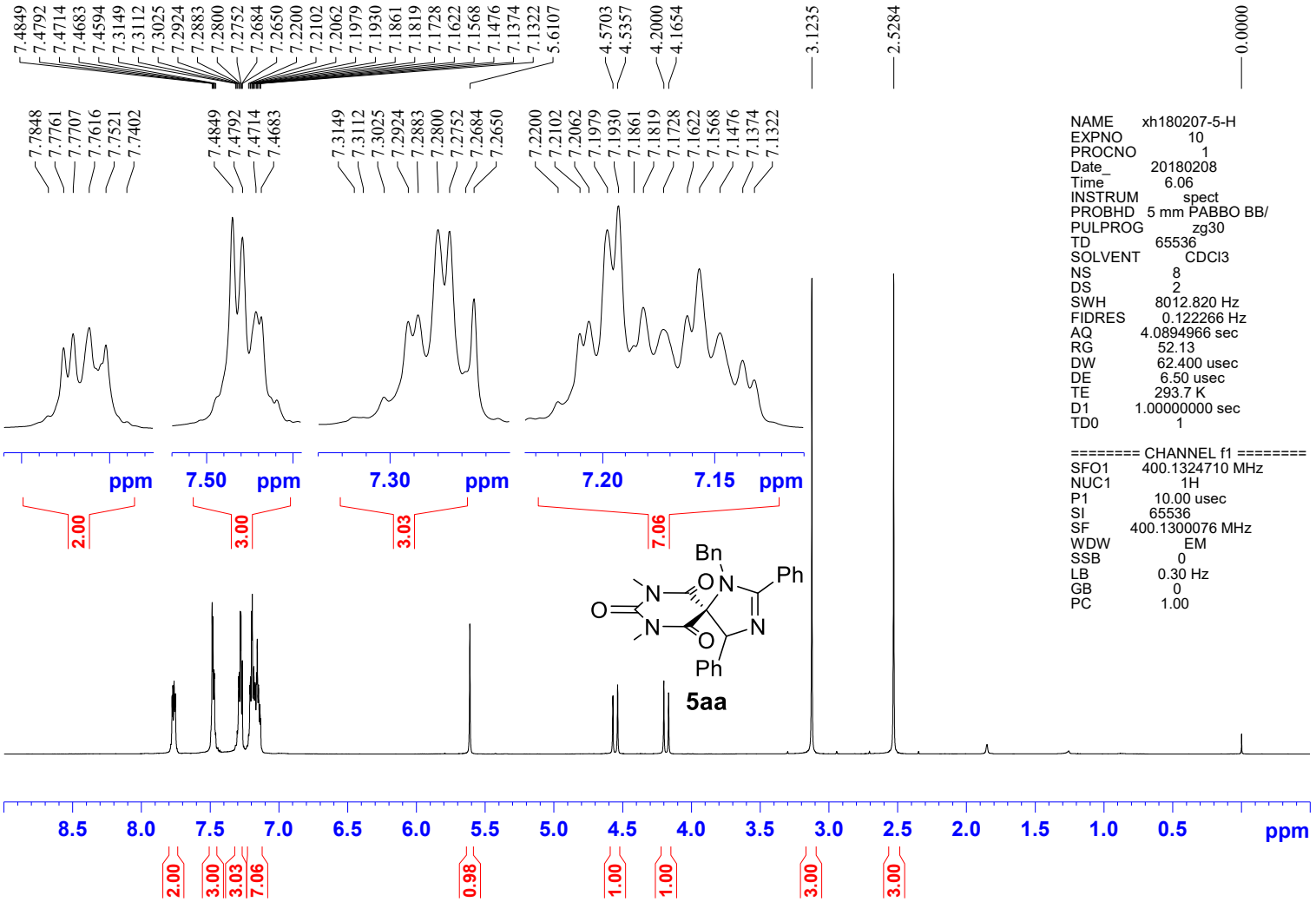


Figure S75. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5aa

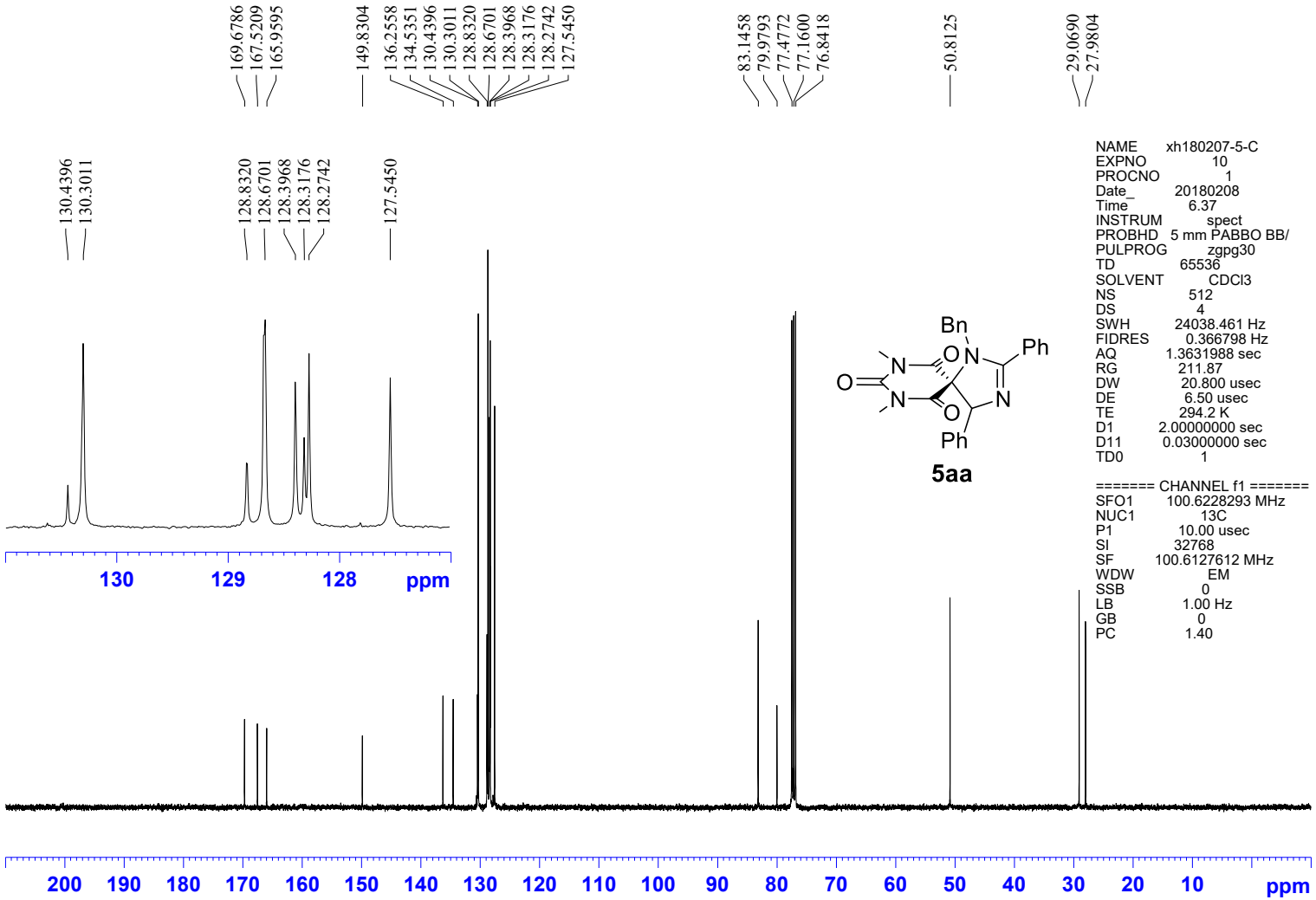


Figure S76. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5aa

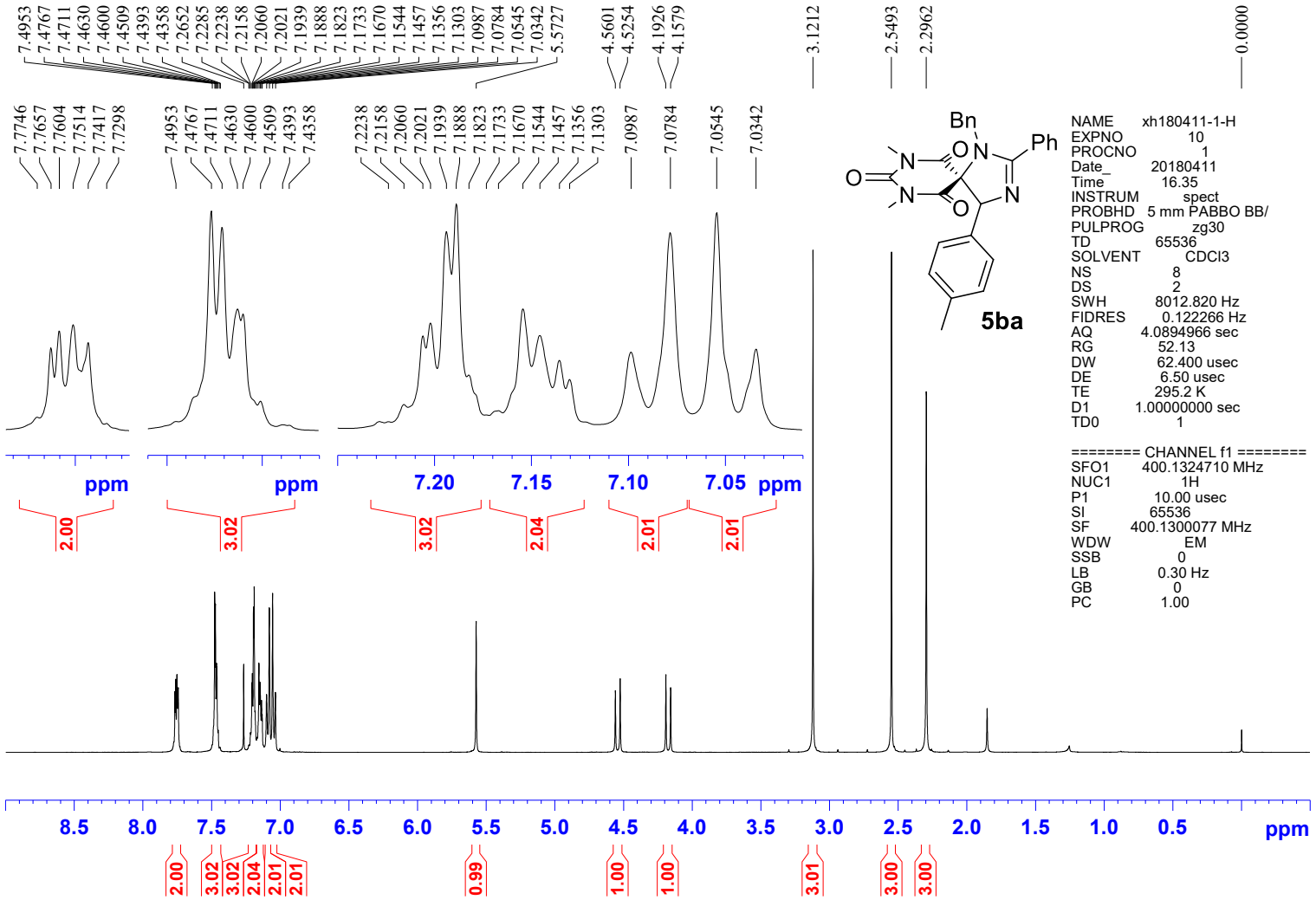


Figure S77. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5ba

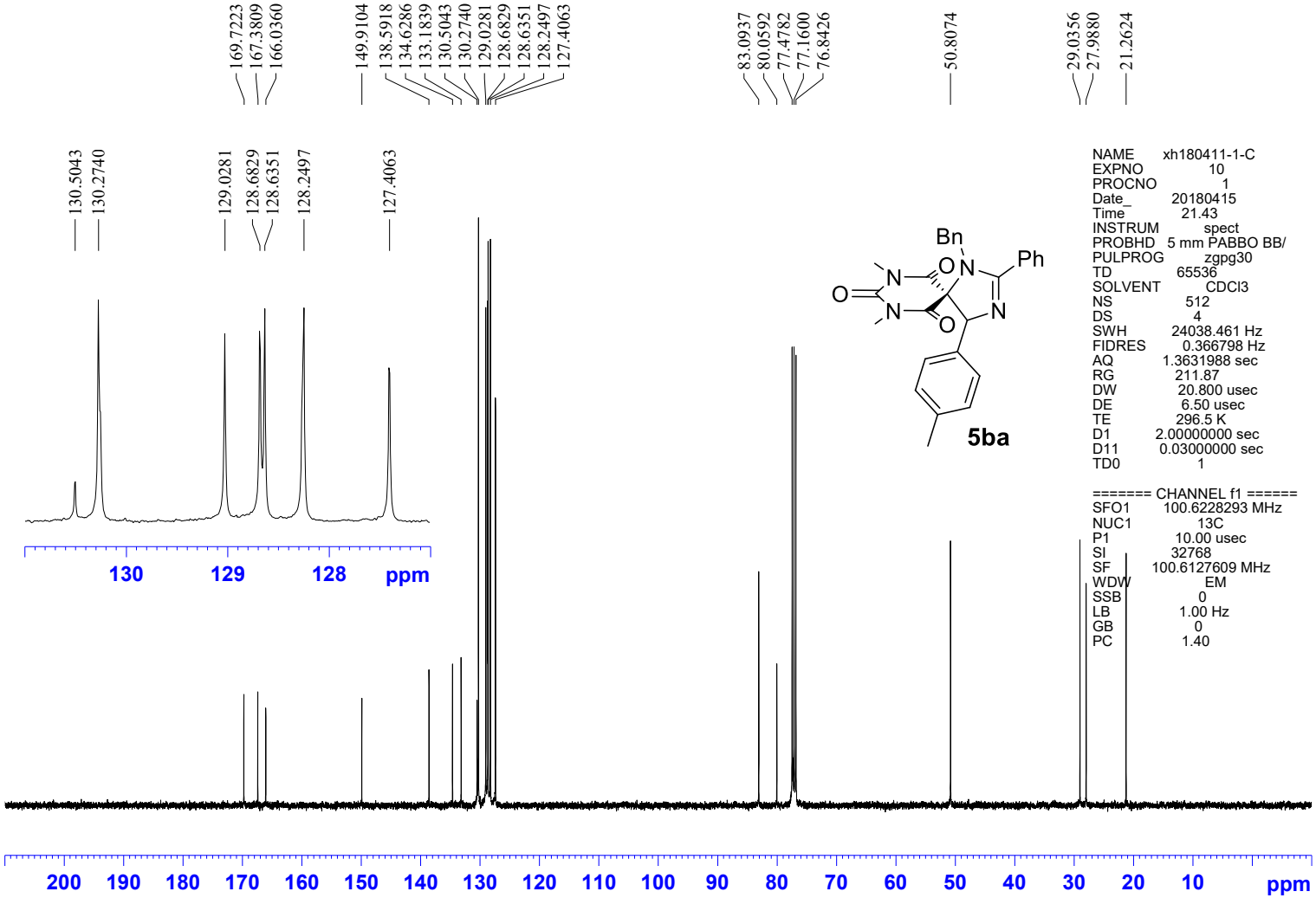


Figure S78. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5ba

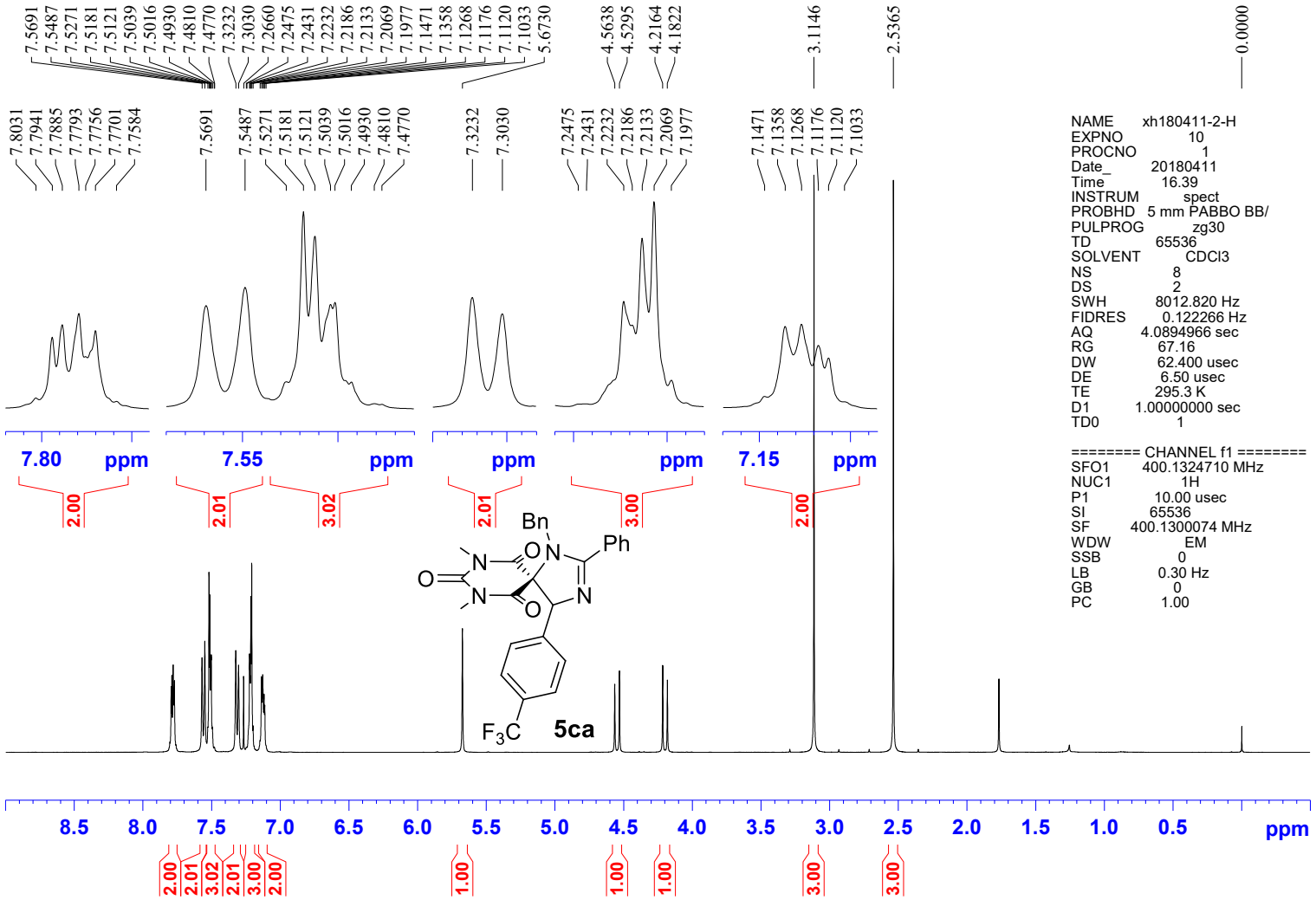


Figure S79. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5ca

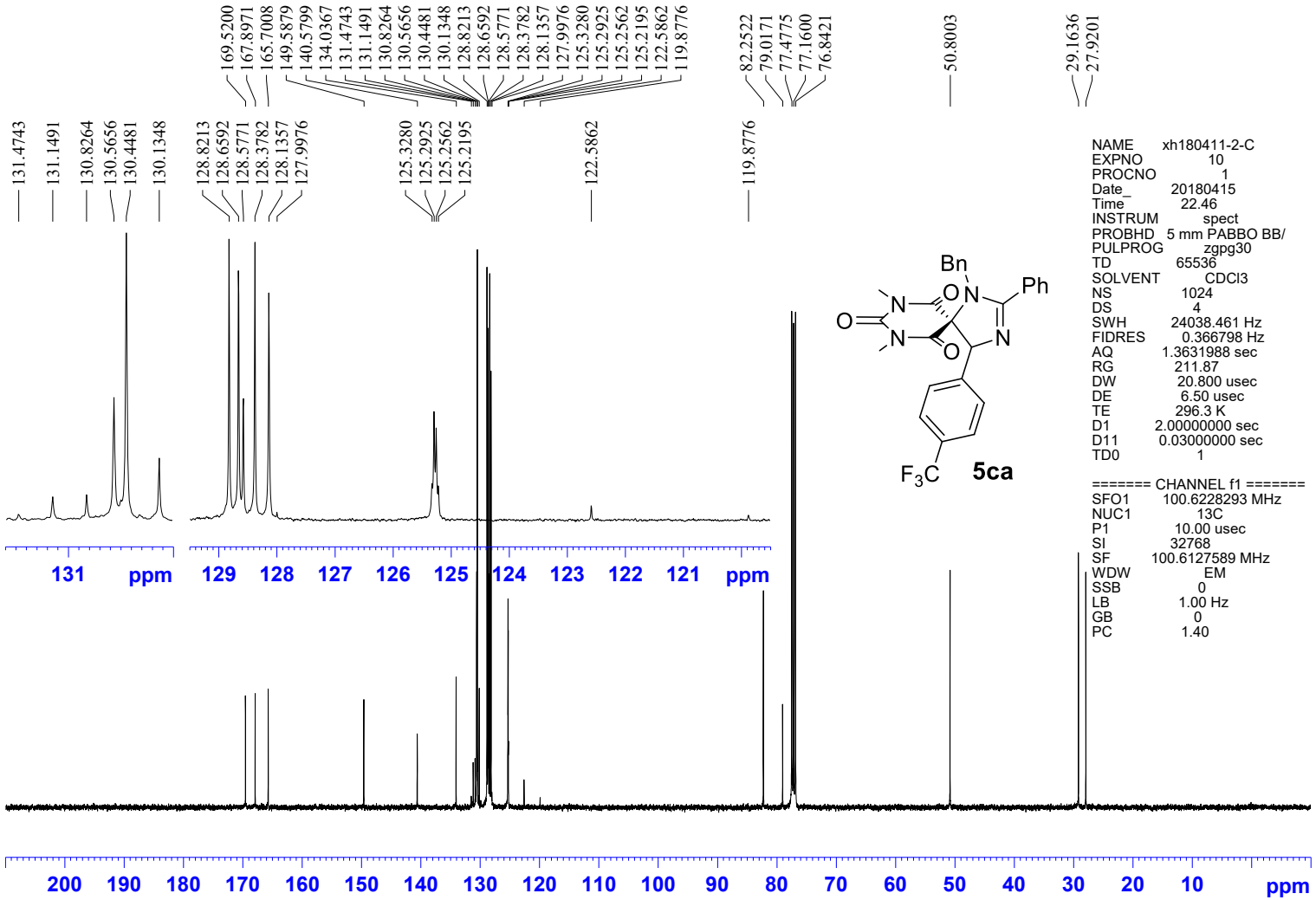


Figure S80. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5ca

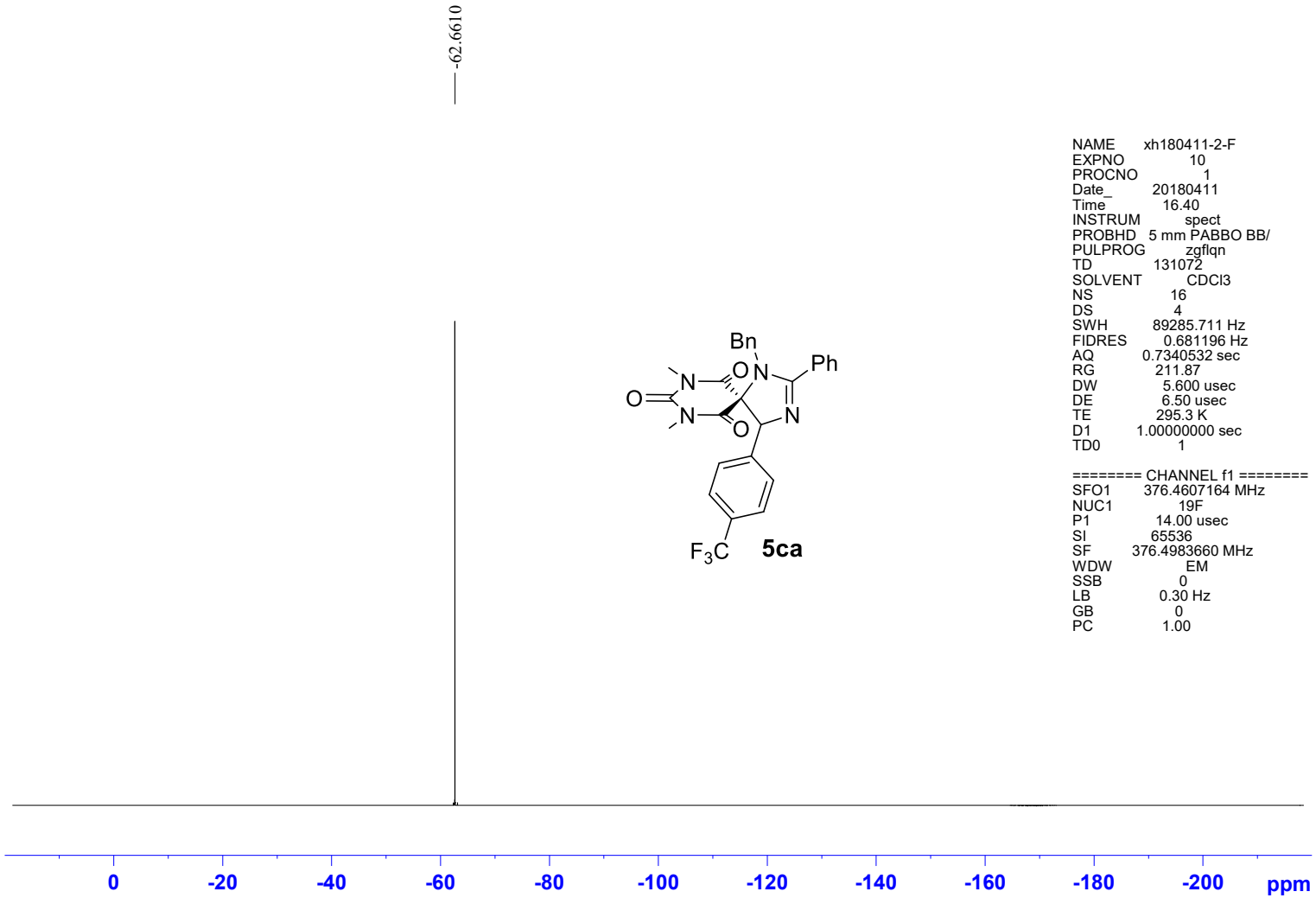


Figure S81. ^{19}F NMR (376 MHz, $CDCl_3$) of compound 5ca

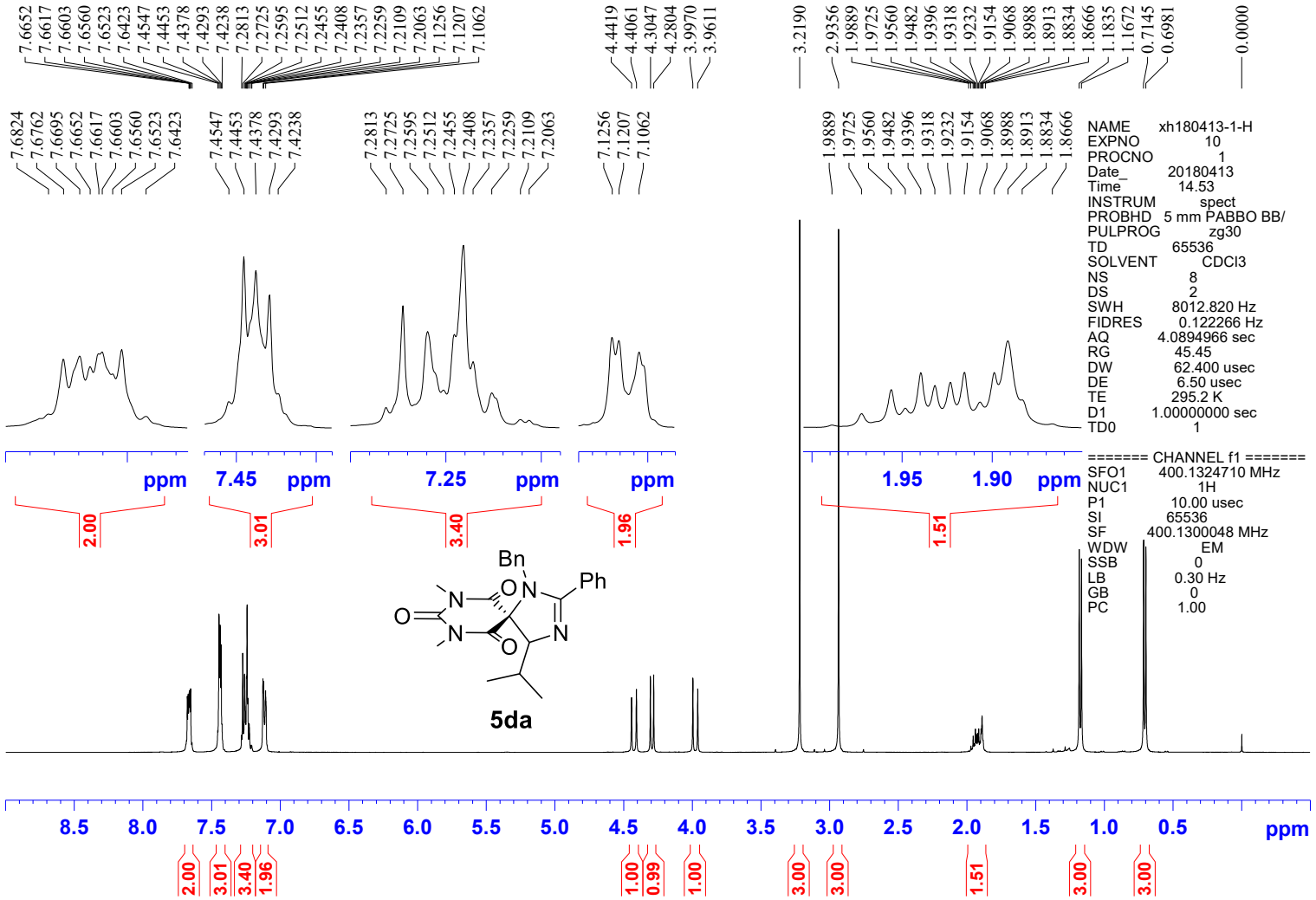


Figure S82. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5da

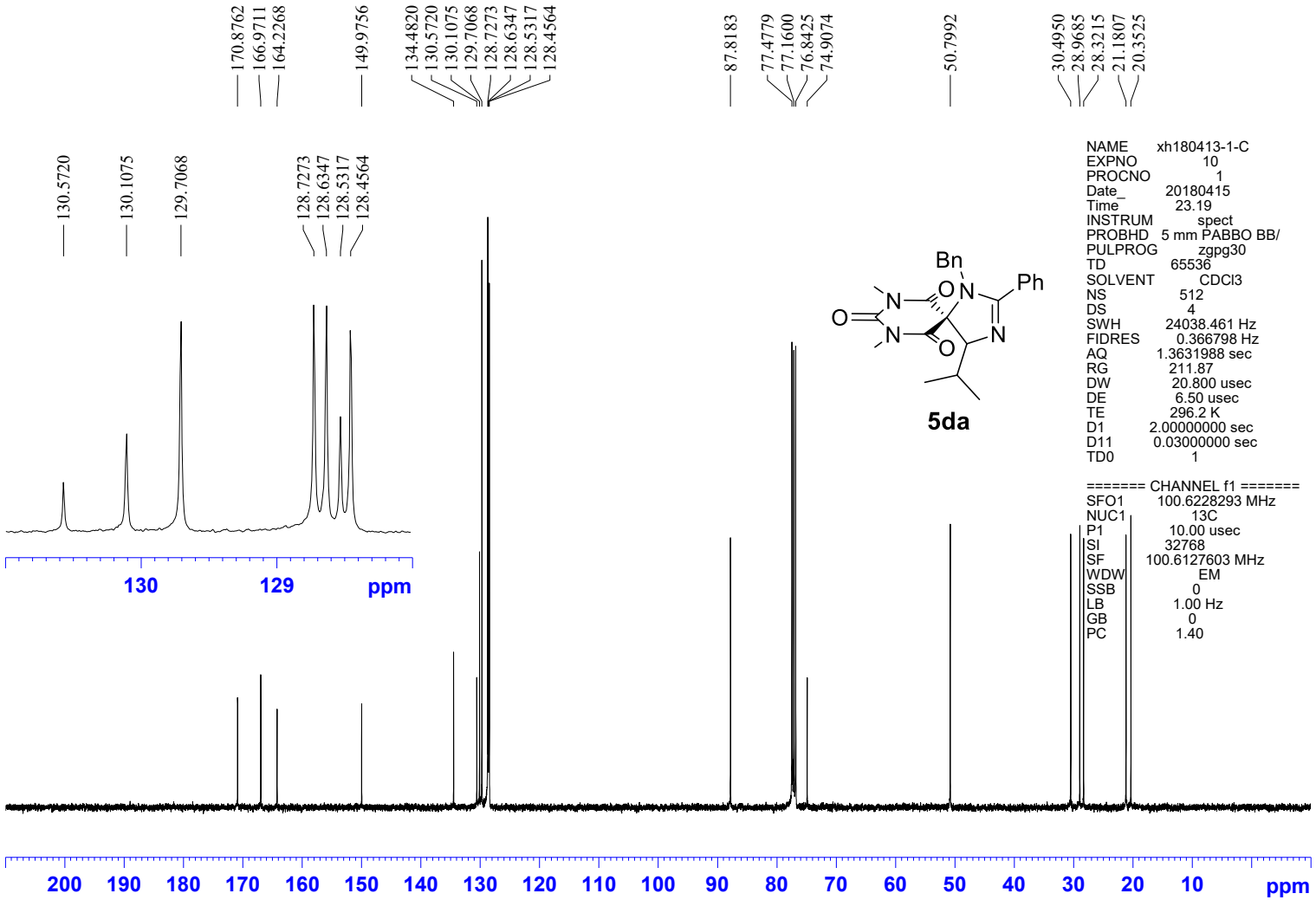


Figure S83. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5da

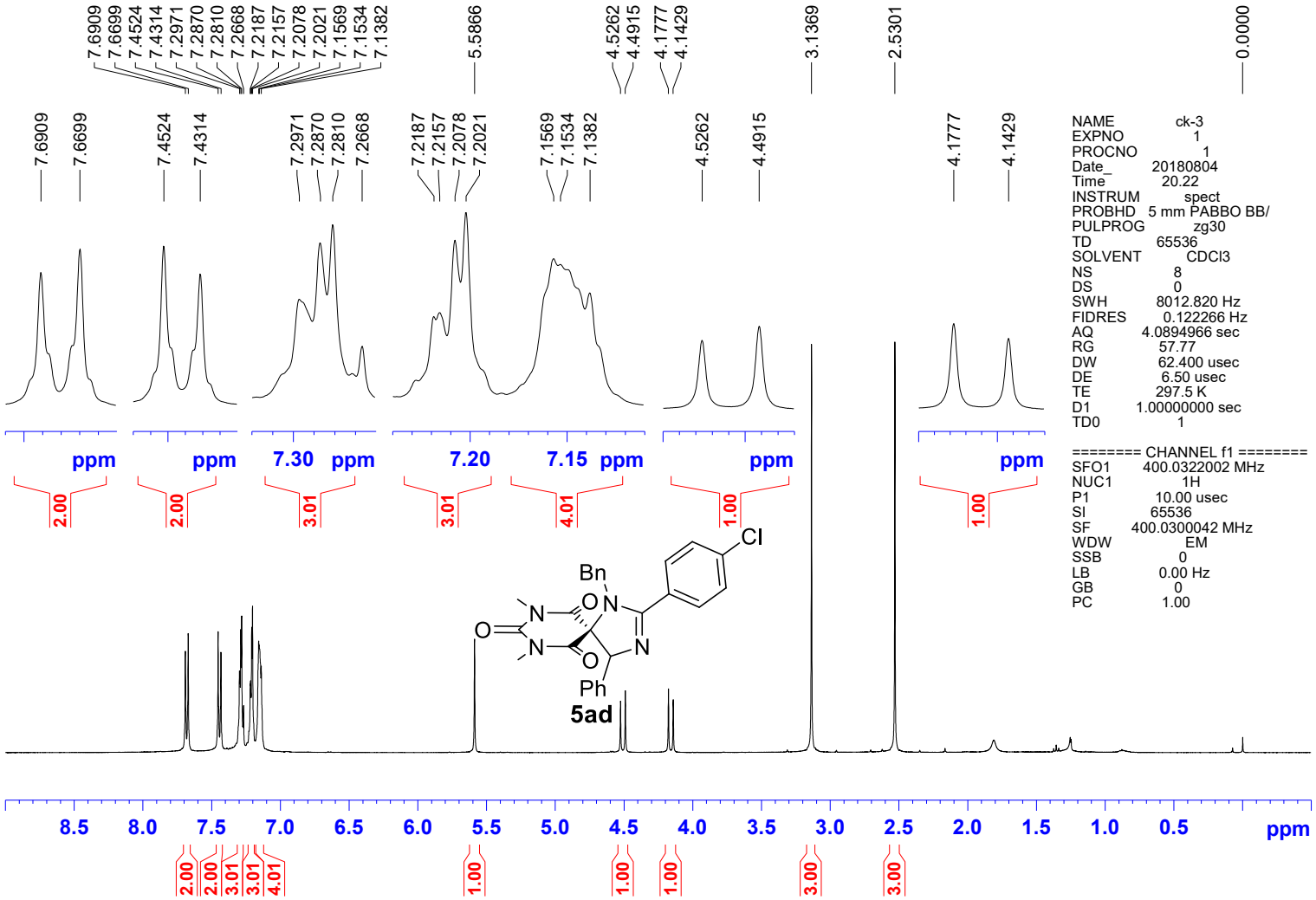


Figure S84. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5ad

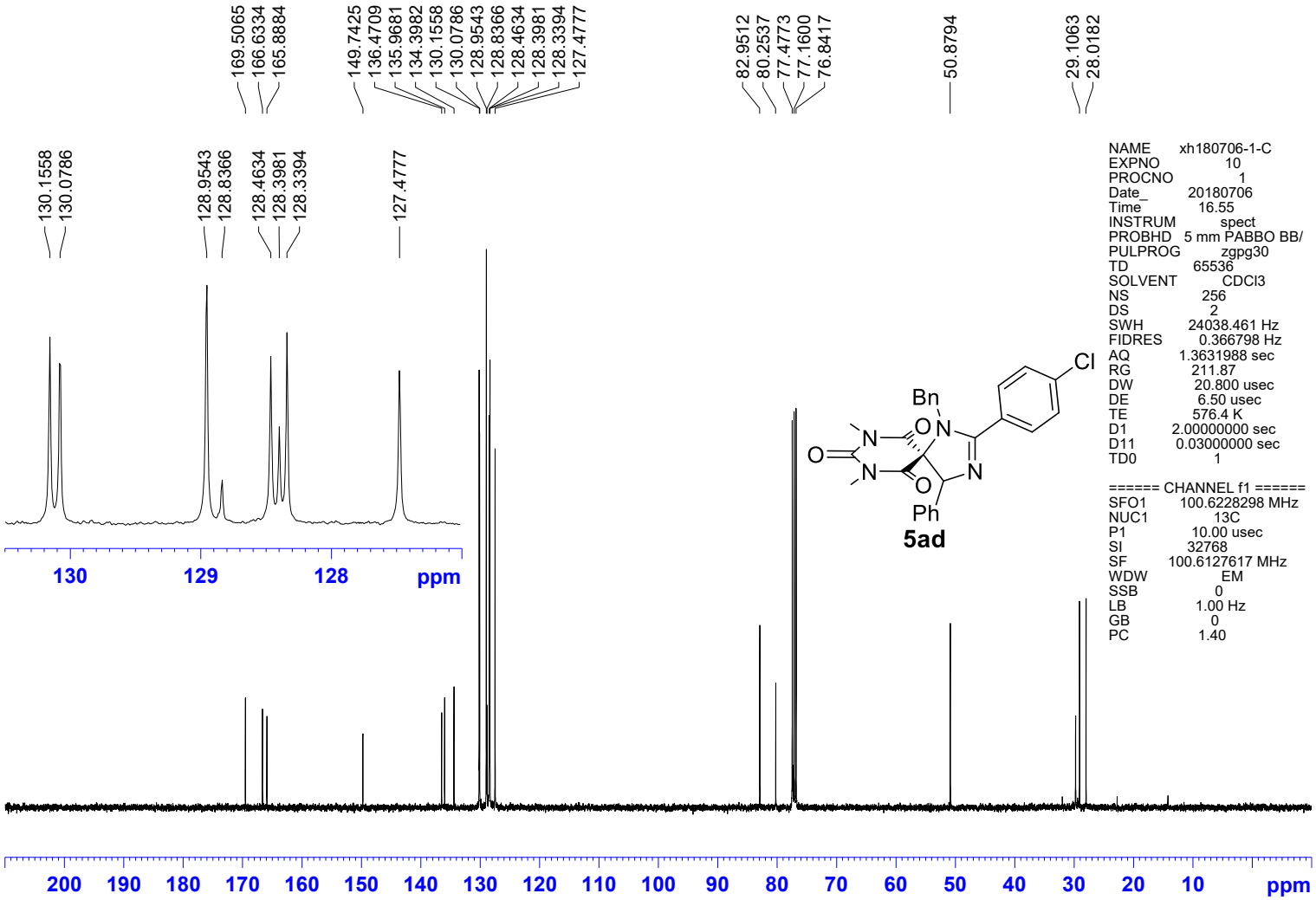


Figure S85. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5ad

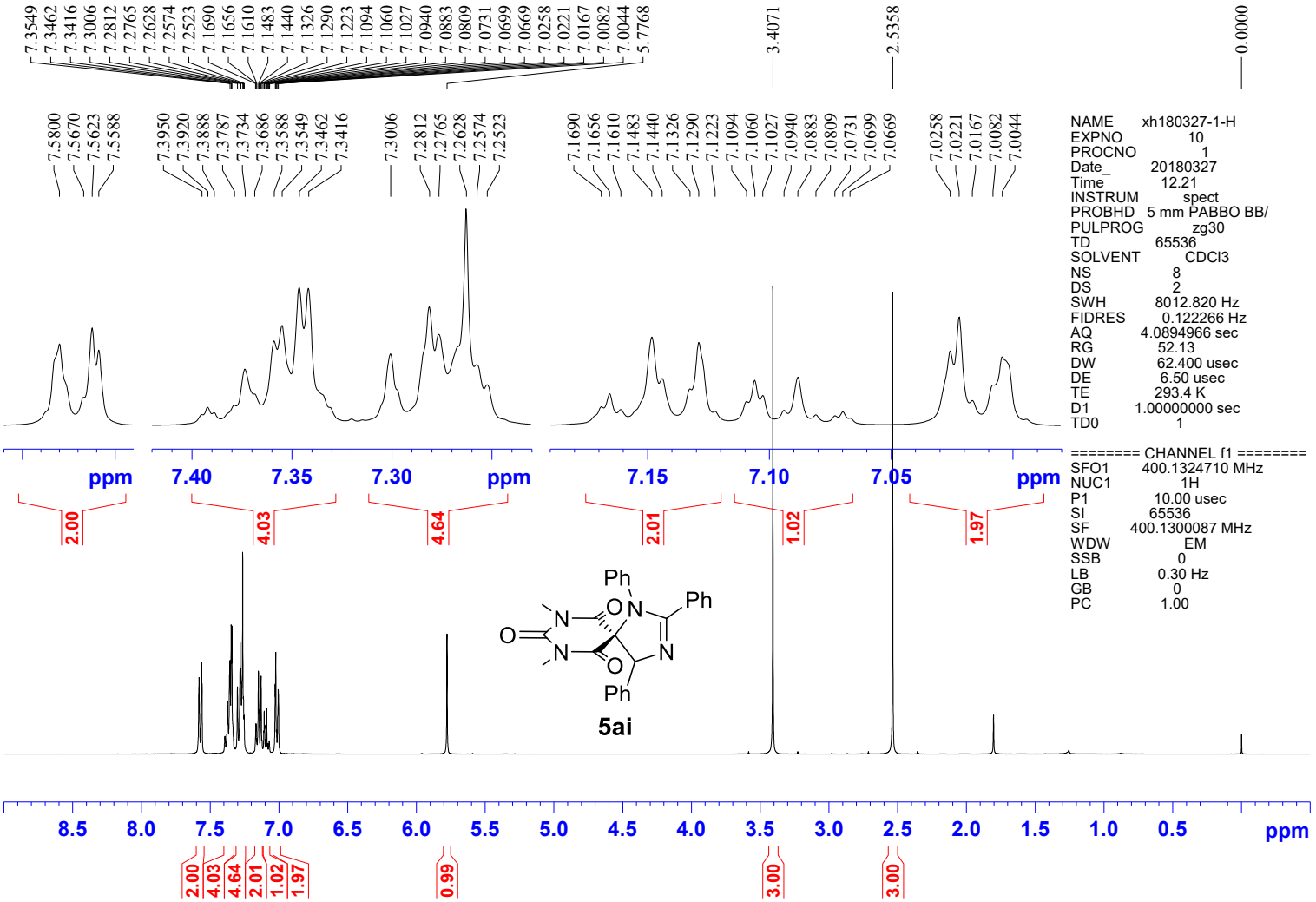


Figure S86. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5ai

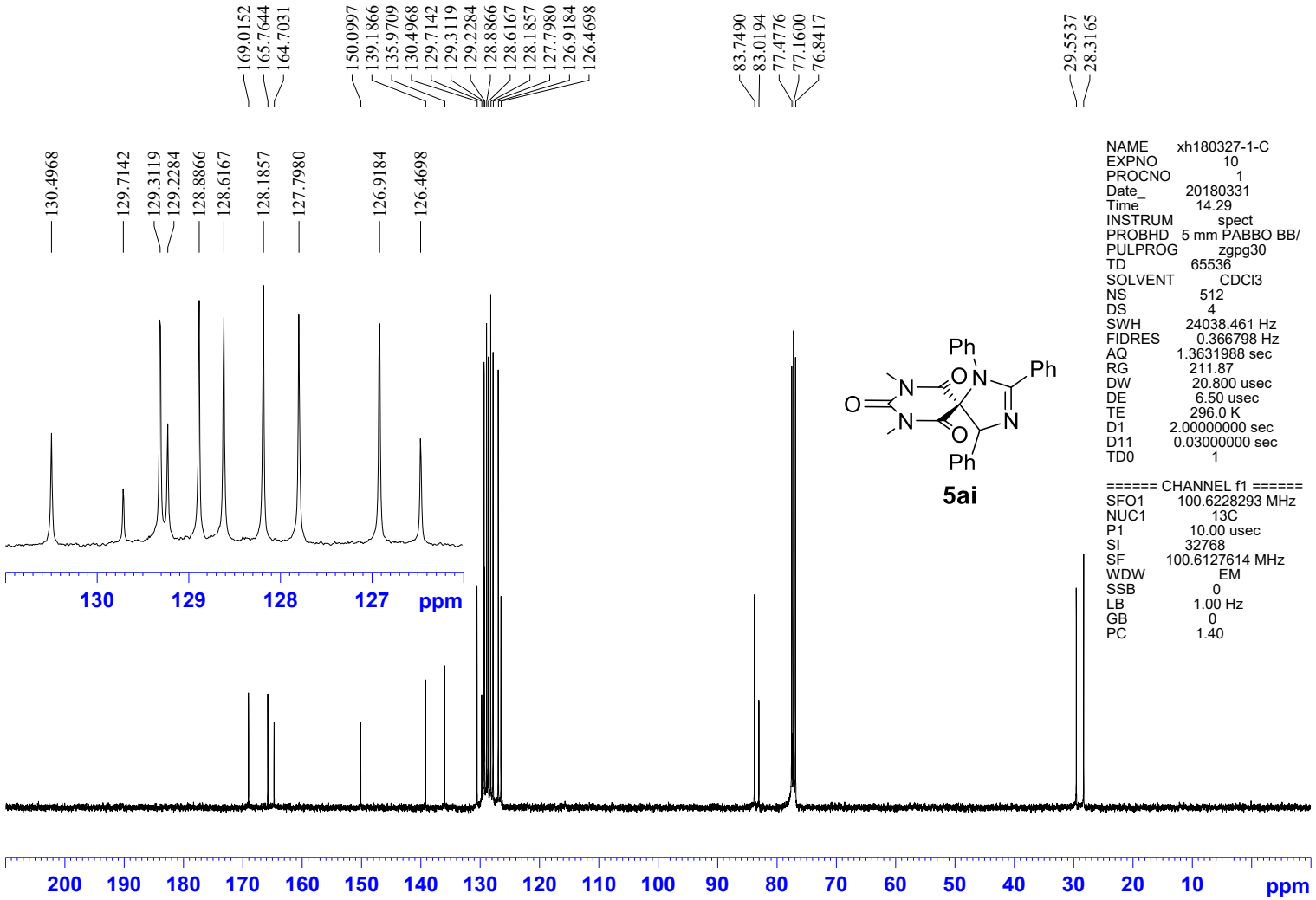


Figure S87. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5ai

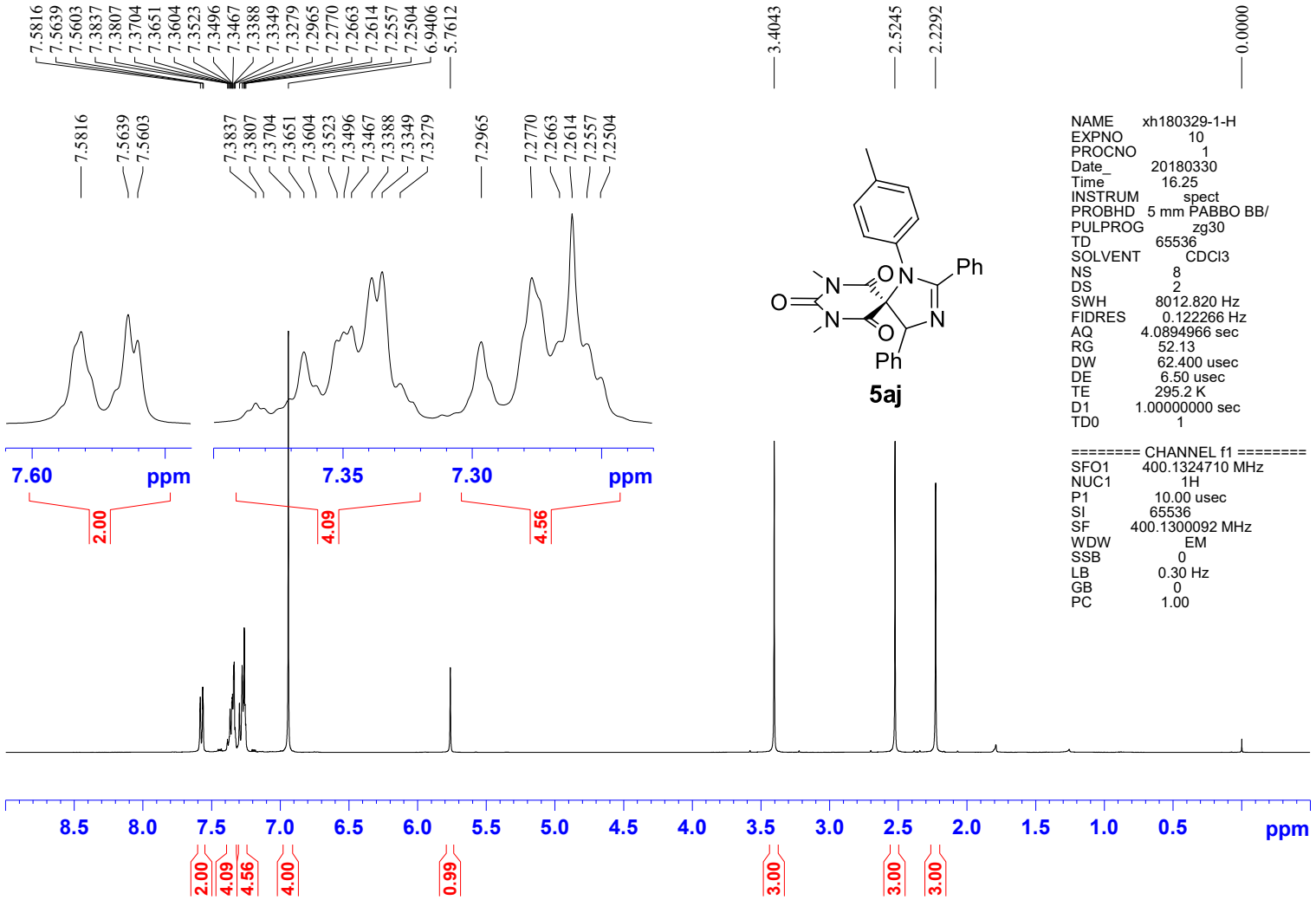


Figure S88. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5aj

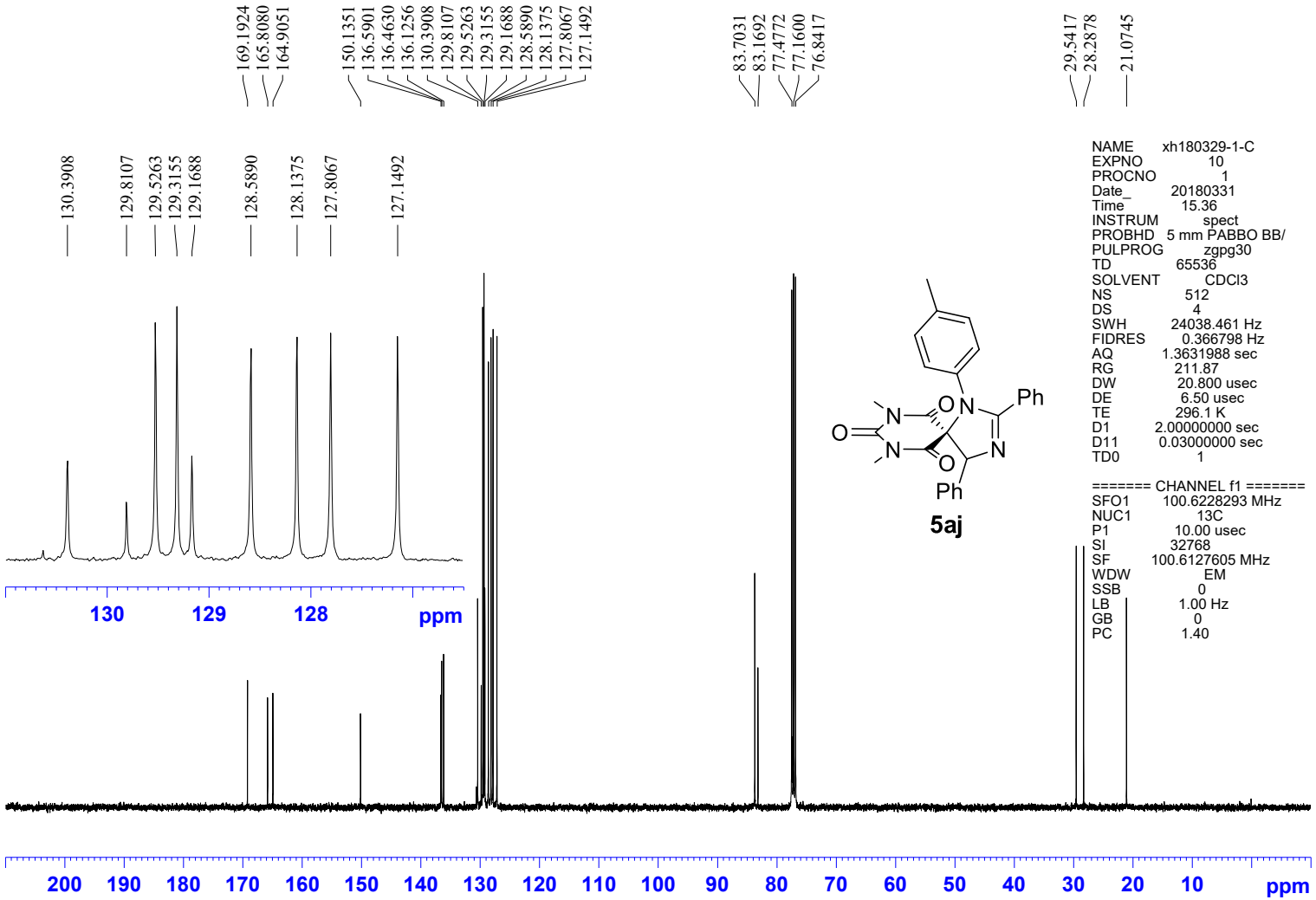


Figure S89. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5aj

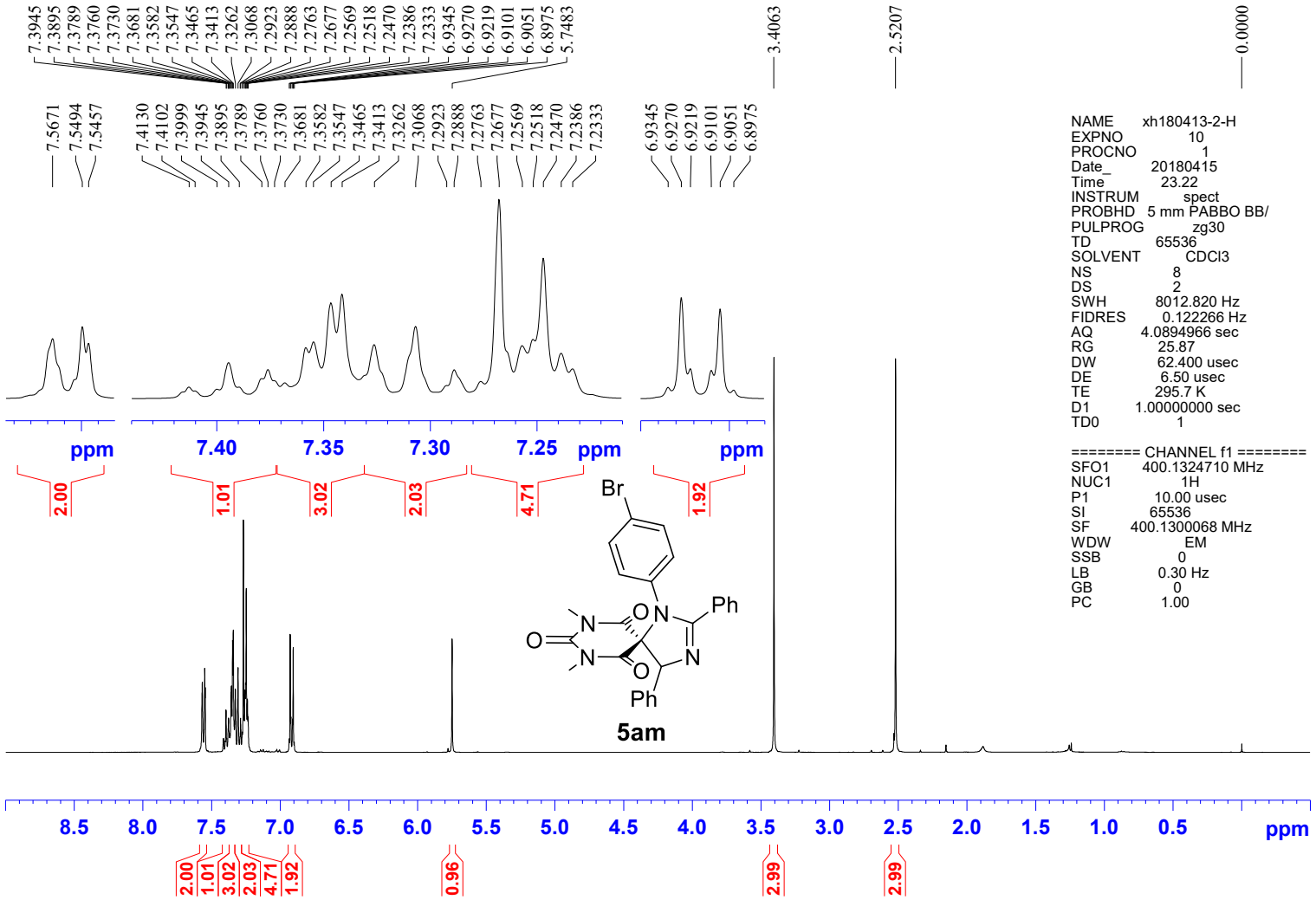


Figure S90. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 5am

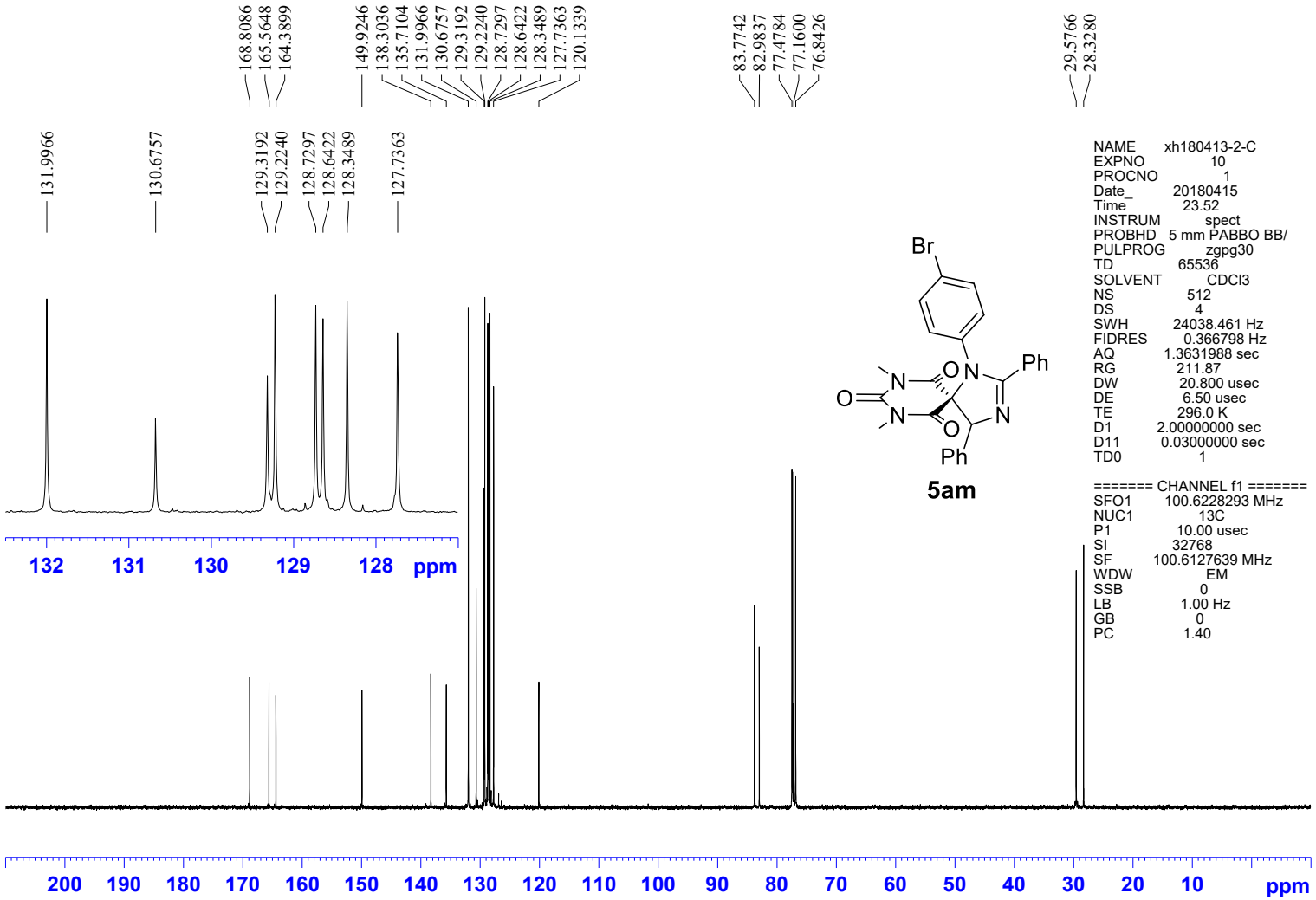


Figure S91. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 5am

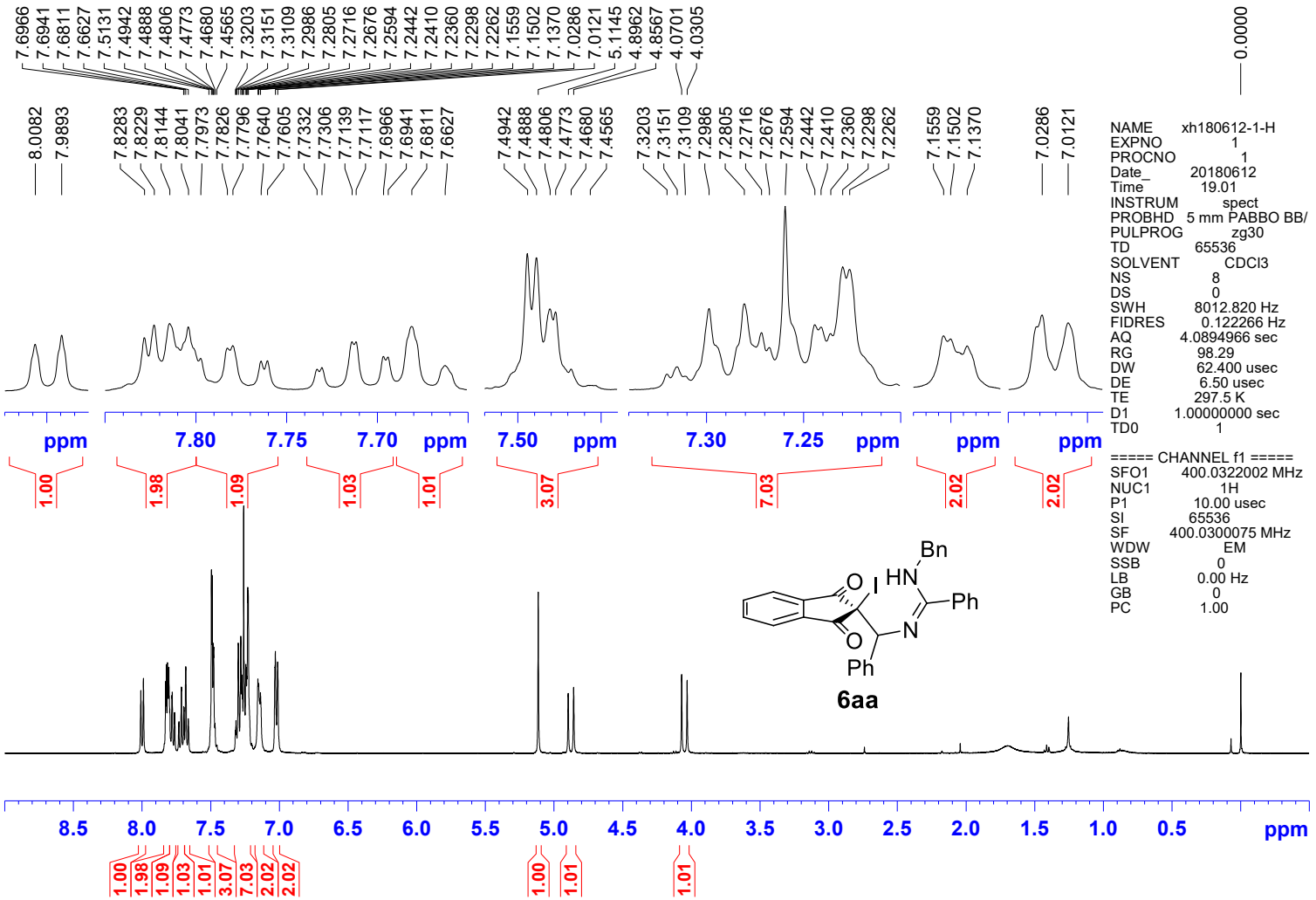


Figure S92. ^{1}H NMR (400 MHz, $CDCl_3$) of compound 6aa

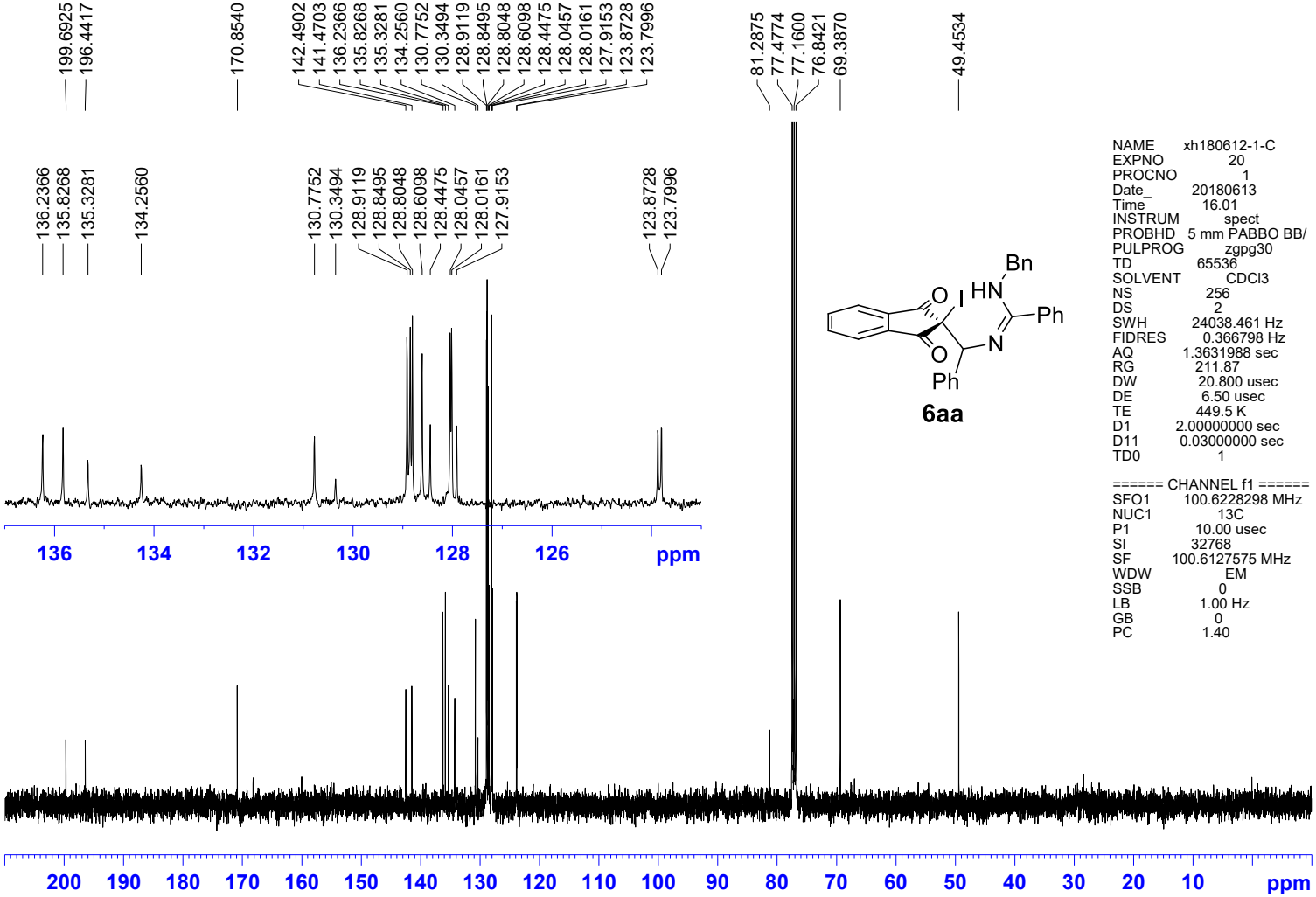


Figure S93. ^{13}C NMR (101 MHz, $CDCl_3$) of compound 6aa

6. Single-crystal X-ray crystallography of 3la

Single crystals of **3la** were obtained by slow evaporation from a mixture of acetone/*n*-hexane at 5 °C. Single-crystal X-ray diffraction data were collected on a diffractometer (Gemini S Ultra, Agilent Technologies) equipped with a CCD area detector using graphite-monochromated CuKα radiation (λ = 1.54184 Å) in the scan range 9.132 < 2θ < 142.318°. The structure was solved with direct methods using SHELXS-97 and refined with full-matrix least-squares refinement using the SHELXL-97 program within OLEX2. Crystallographic data have been deposited in the Cambridge Crystallographic Data Centre as deposition number CCDC 1829368.

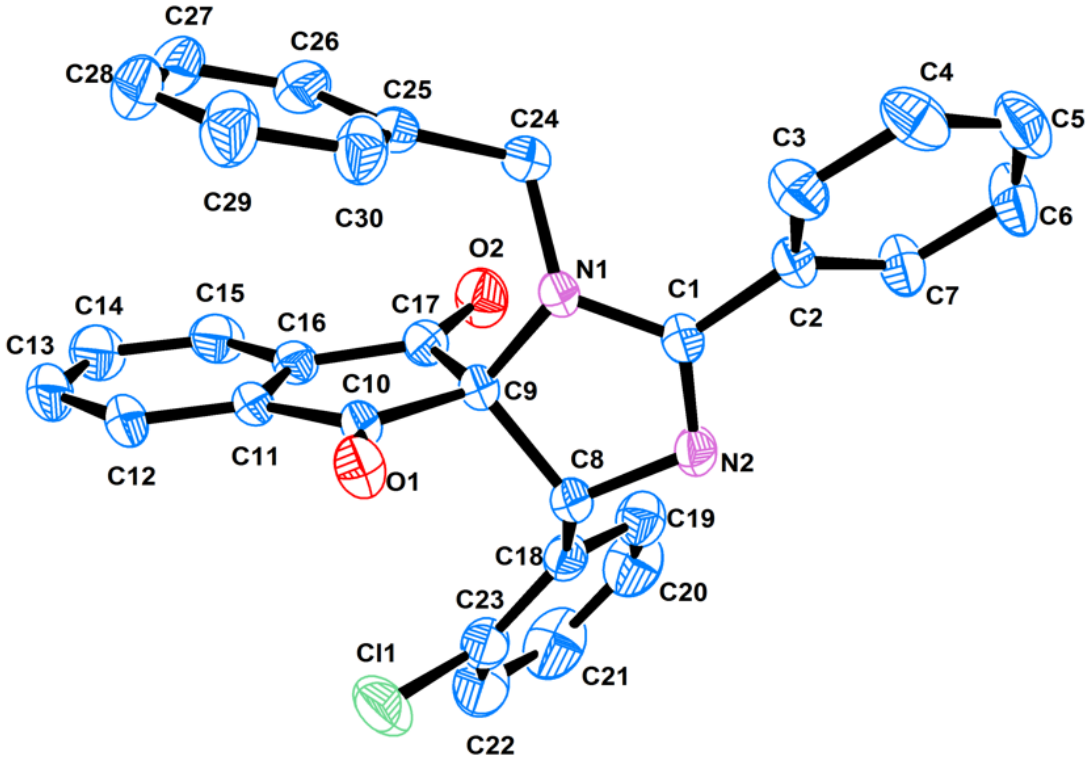


Figure S94. ORTEP Diagrams of 3la with 30% thermal ellipsoids

Table S1 Crystal data and structure refinement for 3la.

Identification code	1829368
Empirical formula	$C_{30}H_{21}ClN_2O_2$
Formula weight	476.94
Temperature/K	291(2)
Crystal system	triclinic
Space group	P-1
a/Å	8.5511(3)
b/Å	10.4286(3)
c/Å	14.3935(4)
α/°	88.875(2)
β/°	87.601(2)
γ/°	68.209(3)
Volume/Å^3	1190.78(7)
Z	2
ρ_{calc}g/cm^3	1.330
μ/mm^{-1}	1.664
F(000)	496.0
Crystal size/mm^3	0.310 × 0.250 × 0.240
Radiation	CuKα (λ = 1.54184)
2Θ range for data collection/°	9.132 to 142.318
Index ranges	$-10 \le h \le 9$, $-12 \le k \le 11$, $-17 \le l \le 17$
Reflections collected	7629
Independent reflections	4450 [R_{int} = 0.0172, R_{sigma} = 0.0180]
Data/restraints/parameters	4450/0/316
Goodness-of-fit on F^2	1.040
Final R indexes [I>=2σ (I)]	R_1 = 0.0464, wR_2 = 0.1270
Final R indexes [all data]	R_1 = 0.0501, wR_2 = 0.1306
Largest diff. peak/hole / e Å^{-3}	0.16/-0.46