Stereoselective Synthesis of Alkynyl Cyclopropanes by Silver-catalyzed Alkynylcyclopropanation of Alkenes with Alkynyl *N*-nosylhydrazones

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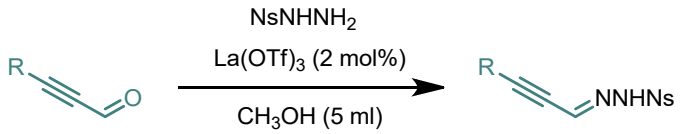
Contents

I. General information

All reagents were purchased from commercial sources and used without purification unless otherwise mentioned. The products were purified by column chromatography over silica gel (300-400). NMR spectra were recorded on a Brüker Advance 600 (^{1}**H**: 600 MHz, ^{13}**C**: 150 MHz) and Brüker Advance 500 (^{1}**H**: 500 MHz, ^{13}**C**: 125 MHz, ^{19}**F**: 471 MHz) at ambient temperature. Data were reported as chemical shifts in ppm relative to TMS (0.00 ppm) for ^{1}H and $CDCl_3$ (77.0 ppm) for ^{13}**C**. The following abbreviations were used to explain the multiplicities: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, br = broad. Mass spectra were recorded on BRUKER AutoflexIII Smartbeam MS-spectrometer. High-resolution mass spectra (HRMS) were recorded on Bruck microTof by using ESI method.

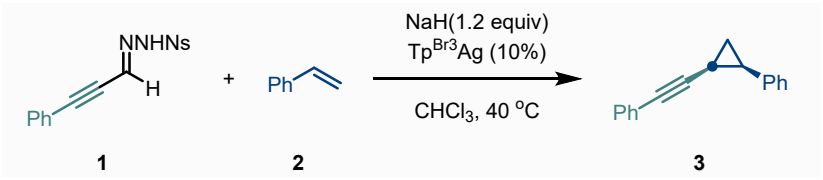
II. General procedures for the synthesis of alkynyl *N*-nosylhydrazones.



According to our previous reported method,[1] to a rapidly stirred suspension of $NsNHNH_2$ (2.0 mmol, 1.0 equiv) and $La(OTf)_3$ (2.0 mol %) in CH_3OH (2.0 mL), phenylpropiolaldehyde (2.2 mmol, 1.1 equiv) was added and stired at 0 °C for 10-20 min (monitored by TLC) and the crude products are obtained as solid precipitates. Then the products were purified by flash chromatography on silica gel. The yields were around 50% in general.

[1]. Y. Yang, Z. Liu, A. Porta, G. Zanoni, X. Bi, *Chem. Eur. J.* **2017**, *23*, 9009-9013.

III. General procedures for synthesis of alkynyl cyclopropanes (with 3a as an example):

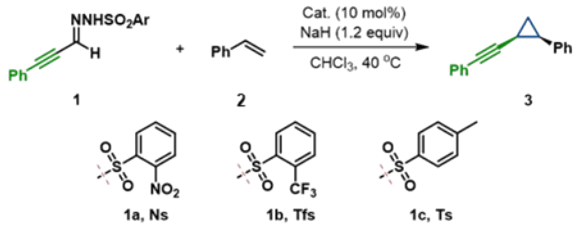


General procedure (with 3a as an example): In the glove box, a sealed tube was charged with alkynyl *N*-nosylhydrazones **1a** (0.3 mmol), NaH (14.0 mg, 60 wt.% dispersion in mineral oil, 1.2 equiv), **2a** (0.6 mmol, 2.0 equiv), $Tp^{Br3}Ag$ (5 mol%) and dry $CHCl_3$ (5.0 mL). The resulting mixture was stirred at 60 °C for 24 h. When the reaction was completed, the reaction was allowed to cool to room temperature, and filtered through a short pad of silica gel with EtOAc as an eluent. After removal of the solvent under vacuum, the residue was purified by flash chromatography on silica gel (using PE and EA as eluent) to obtain the final product **3a**.

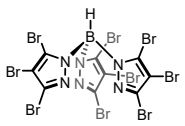
IV. Optimization of the Reaction Conditions

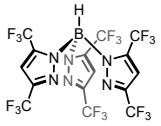
First, alkynyl *N*-nosylhydrazone **1a** was studied as a potential alkynylcarbene precursor in order to evaluate the suitability of the catalysts in the reaction of **1a** with styrene (2) and NaH at 40°C in $CHCl_3$ (Table 1). Pleasingly, owing to the use of the more electrophilic $Tp^{Br3}Ag$ (10 mol%) and $Tp^{(CF3)2}Ag$ (10 mol%) as a catalyst, the desired cyclopropane product was obtained in 93% and 84% yields, respectively,

with excellent diastereoisomeric ratios (>20:1) (See SI, Table 1, entry 1-2). The reaction could also proceed using a simple silver salt, such as AgOAc, and afforded the cyclopropanation product **3** in a 46% yield and in a diastereoisomeric ratio (d.r.) of 5:1 (entry 3). Interestingly, $Rh_2(Oct)_4$ afforded the cyclopropanation product in a 92% yield, with a poor diastereoselectivity (4.8:1 d.r.) (entry 4).[13] Furthermore, the other most commonly used transition metal catalysts for carbene chemistry were investigated. These catalysts, such as $Cu(OAc)_2$, $Pd(OAc)_2$, and Fe(TPP)Cl,[18] were not suitable for this cyclopropanation reaction and resulted in lower yields or were observed to be ineffective (entries 5-7). Interestingly, the choice of diazo surrogate had a marked effect on the efficiency of the reaction, with lower yields obtained using *N*-triftosylhydrazone **1b** or *N*-tosylhydrazone **1c** (entries 8 and 9).



Entry	[M] Cat.	1	Yield	d.r. of 3
1	$Tp^{Br3}Ag$ (10 mol%)	**1a**	93%	>20:1
2	$Tp^{(CF3)2}Ag$ (10 mol%)	**1a**	84%	>20:1
3	AgOAc (10 mol%)	**1a**	46%	5:1
4	$Rh_2(Oct)_4$ (2 mol%)	**1a**	92%	4.8:1
5	$Cu(OAc)_2$ (10 mol%)	**1a**	48%	5:1
6	Fe(TPP)Cl (2 mol%)	**1a**	32%	9:1
7	$Pd(OAc)_2$ (5 mol%)	**1a**	trace	--
8	$Tp^{Br3}Ag$ (10 mol%)	**1b**	73%	20:1
9	$Tp^{Br3}Ag$ (10 mol%)	**1c**	20%	20:1



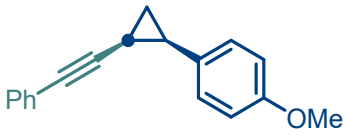


[a] Reaction conditions: **1** (0.3 mmol), **2** (0.6 mmol), NaH (1.2 equiv), catalyst in $CHCl_3$ (5 mL) at 40 °C for 24 h. [b] Isolated yield. TPP = tetramethoxyphenylporphyrin.

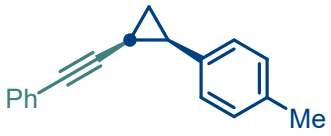
V. Characterization data of prepared compounds.

(3) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.35-7.28 (m, 4H), 7.25-7.20 (m, 1H), 7.19-7.15 (m, 3H), 7.13-7.11 (m, 2H), 2.38 (dd, *J* = 15.0, 8.4 Hz, 1H), 2.01-1.93 (m, 1H), 1.46-1.39 (m, 1H), 1.31-1.24 (m, 1H). **^{13}C NMR**

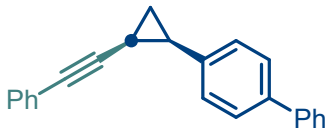
(150 MHz, $CDCl_3$) δ 138.0, 131.4, 128.3, 128.0, 127.7, 127.3, 126.2, 123.8, 89.7, 80.2, 24.0, 15.0, 10.2. **HRMS** (ESI) m/z calcd. for $C_{17}H_{13}^-$ $[M-H]^-$: 217.1023; Found: 217.1018.



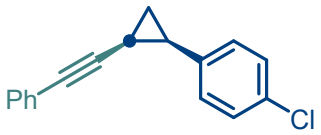
(4) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.26-7.23 (m, 2H), 7.21-7.17 (m, 3H), 7.17-7.13 (m, 2H), 6.89-6.85 (m, 2H), 3.80 (s, 3H), 2.35 (dd, *J* = 15.0, 8.5 Hz, 1H), 1.97-1.89 (m, 1H), 1.45-1.38 (m, 1H), 1.23-1.18 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 158.2, 131.4, 130.1, 129.4, 128.0, 127.3, 123.9, 113.3, 90.0, 80.1, 55.3, 23.3, 15.0, 9.8. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}O^+$ $[M+H]^+$: 249.1274; Found: 249.1270.



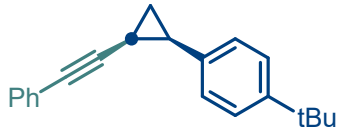
(5) Colorless oil, **H NMR** (500 MHz, $CDCl_3$) δ 7.25-7.13 (m, 5H), 7.17-7.14 (m, 2H), 7.12 (d, *J* = 8.0 Hz, 2H), 2.34 (s, 3H), 2.39-2.30 (m, 1H), 1.99-1.91 (m, 1H), 1.46-1.40 (m, 1H), 1.27-1.21 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 135.8, 134.9, 131.4, 128.5, 128.2, 128.0, 127.4, 123.9, 90.0, 80.2, 23.7, 21.1, 15.1, 10.1. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}^+$ $[M+H]^+$: 233.1325; Found: 233.1322.



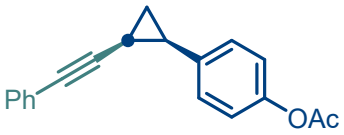
(6) Colorless oil, **H NMR** (500 MHz, $CDCl_3$) δ 7.60 (d, *J* = 8.0 Hz, 2H), 7.56 (d, *J* = 8.0 Hz, 2H), 7.44 (t, *J* = 7.5 Hz, 2H), 7.39 (d, *J* = 8.0 Hz, 2H), 7.33 (t, *J* = 7.5 Hz, 1H), 7.21-7.13 (m, 5H), 2.44 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.06-1.99 (m, 1H), 1.52-1.46 (m, 1H), 1.33 (dd, *J* = 12.0, 6.0 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 141.1, 139.1, 137.3, 131.4, 128.7, 128.7, 128.1, 127.4, 127.0, 126.5, 123.7, 89.7, 80.4, 23.7, 15.3, 10.4. **HRMS** (ESI) m/z calcd. for $C_{23}H_{19}^+$ $[M+H]^+$: 295.1481; Found: 295.1480.



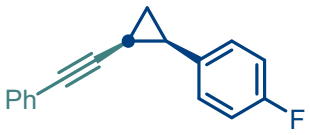
(7) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.28 (d, *J* = 8.5 Hz, 2H), 7.23 (d, *J* = 8.5 Hz, 2H), 7.21-7.19 (m, 3H), 7.17-7.12 (m, 2H), 2.34 (dd, *J* = 15.0, 8.0 Hz, 1H), 2.02-1.95 (m, 1H), 1.48-1.42 (m, 1H), 1.23 (dd, *J* = 12.0, 6.0 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 136.6, 132.0, 131.4, 129.7, 128.1, 127.8, 127.5, 123.5, 89.2, 80.5, 23.4, 15.2, 10.3. **HRMS** (ESI) m/z calcd. for $C_{17}H_{14}Cl^+$ $[M+H]^+$: 253.0779; Found: 253.0774.



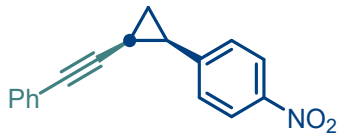
(8) Colorless oil, **H NMR** (500 MHz, $CDCl_3$) δ 7.38-7.32 (m, 2H), 7.28-7.23 (m, 2H), 7.21-7.12 (m, 3H), 7.11-7.05 (m, 2H), 2.39 (dd, *J* = 15.0, 8.0 Hz, 1H), 1.98-1.80 (m, 1H), 1.46-1.39 (m, 1H), 1.33 (s, 9H), 1.27 (dd, *J* = 12.0, 5.5 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 149.1, 135.0, 131.4, 128.1, 128.0, 127.3, 124.7, 123.9, 90.2, 80.0, 34.4, 31.4, 23.6, 14.9, 9.9. **HRMS** (ESI) m/z calcd. for $C_{21}H_{23}^+$ $[M+H]^+$: 275.1794; Found: 275.1791.



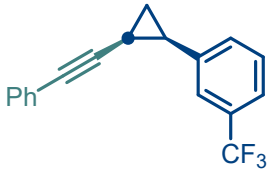
(9) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.23-7.17 (m, 4H), 7.17-7.13 (m, 3H), 7.11 (d, *J* = 7.5 Hz, 1H), 7.04 (d, *J* = 7.5 Hz, 1H), 2.34 (s, 3), 2.40-2.31 (m, 3H), 2.01-1.93 (m, 1H), 1.47-1.40 (m, 1H), 1.30-1.24 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 137.9, 137.2, 131.4, 129.1, 128.0, 127.6, 127.3, 127.0, 125.3, 123.8, 89.8, 80.2, 23.9, 21.5, 15.1, 10.2. **HRMS** (ESI) m/z calcd. for $C_{19}H_{17}O_2^+$ $[M+H]^+$: 277.1223; Found: 277.1220.



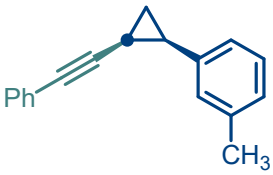
(10) Colorless oil, **H NMR** (500 MHz, $CDCl_3$) δ 7.31-7.25 (m, 2H), 7.24-7.16 (m, 3H), 7.18-7.10 (m, 2H), 7.05-6.95 (m, 2H), 2.36 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.01-1.90 (m, 1H), 1.48-1.41 (m, 1H), 1.25-1.19 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 161.6 (d, *J* = 242.5 Hz), 133.7 (d, *J* = 2.5 Hz), 131.3, 129.8 (d, *J* = 7.5 Hz), 128.1, 127.5, 123.6, 114.5 (d, *J* = 21.3 Hz), 89.5, 80.3, 23.2, 15.1, 10.0. **HRMS** (ESI) m/z calcd. for $C_{17}H_{14}F^+$ $[M+H]^+$: 237.1074; Found: 237.1072.



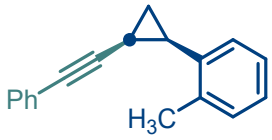
(11) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 8.18 (d, *J* = 8.5 Hz, 2H), 7.44 (d, *J* = 8.5 Hz, 2H), 7.25-7.17 (m, 3H), 7.18-7.12 (m, 2H), 2.47 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.12-2.11 (m, 1H), 1.61-1.56 (m, 1H), 1.40-1.35 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 146.5, 146.3, 131.3, 128.8, 128.2, 127.9, 123.1, 122.9, 88.1, 81.6, 24.0, 16.2, 11.6. **HRMS** (ESI) m/z calcd. for $C_{17}H_{12}NO_2^-$ $[M-H]^-$: 262.0874; Found: 262.0876.



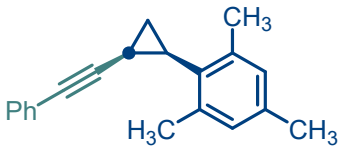
(12) Colorless oil, **H NMR** (500 MHz, $CDCl_3$) δ 7.58 (s, 1H), 7.52-7.47 (m, 2H), 7.42 (t, *J* = 8.0 Hz, 1H), 7.24-7.16 (m, 3H), 7.17-7.10 (m, 2H), 2.44 (dd, *J* = 15.5, 8.0 Hz, 1H), 2.10-2.02 (m, 1H), 1.54-1.48 (m, 1H), 1.35-1.28 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 139.2, 131.8, 131.3, 130.1 (d, *J* = 32.5 Hz), 128.14, 128.07, 127.6, 124.9 (d, *J* = 3.8 Hz), 124.3 (d, *J* = 270.0 Hz), 123.4, 123.1 (d, *J* = 3.8 Hz), 88.7, 80.9, 23.7, 15.4, 10.5. **HRMS** (ESI) m/z calcd. for $C_{18}H_{14}F_3^+$ $[M+H]^+$: 287.1042; Found: 287.1039.



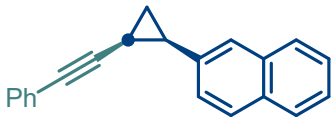
(13) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.23-7.17 (m, 4H), 7.17-7.13 (m, 3H), 7.11 (d, *J* = 7.5 Hz, 1H), 7.04 (d, *J* = 7.5 Hz, 1H), 2.40-2.32 (m, 4H), 2.0-1.92 (m, 1H), 1.47-1.40 (m, 1H), 1.29-1.24 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 137.9, 137.2, 131.4, 129.1, 128.0, 127.6, 127.3, 127.0, 125.3, 123.8, 89.8, 80.2, 23.9, 21.5, 15.1, 10.2. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}^+$ $[M+H]^+$: 233.1325; Found: 233.1321.



(14) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.23-7.15 (m, 4H), 7.15-7.09 (m, 3H), 6.98-6.94 (m, 2H), 2.44 (s, 3H), 2.34 (dd, *J* = 15.0, 8.0 Hz, 1H), 2.03-1.97 (m, 1H), 1.45-1.39 (m, 1H), 1.36-1.30 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.8, 136.4, 131.4, 129.5, 128.1, 127.9, 127.2, 126.5, 125.3, 123.8, 89.9, 78.9, 22.5, 19.7, 13.9, 8.6. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}^+$ $[M+H]^+$: 233.1325; Found: 233.1322.



(15) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.19-7.12 (m, 3H), 7.07-7.02 (m, 2H), 6.86-6.83 (m, 2H), 2.43 (s, 6H), 2.26 (s, 3H), 2.17-2.08 (m, 1H), 2.03-1.95 (m, 1H), 1.62-1.56 (m, 1H), 1.17-1.08 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 139.0, 135.8, 131.7, 131.5, 128.7, 127.9, 127.2, 124.0, 91.0, 78.4, 20.9, 20.8, 20.7, 17.6, 8.8. **HRMS** (ESI) m/z calcd. for $C_{20}H_{21}^+$ $[M+H]^+$: 261.1638; Found: 261.1634.



(16) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.86-7.73 (m, 4H), 7.51-7.38 (m, 3H), 7.18-7.08 (m, 3H), 7.08-7.02 (m, 2H), 2.55 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.11-2.02 (m, 1H), 1.57-1.50 (m, 1H), 1.46-1.39 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 135.7, 133.2, 132.3, 131.3, 128.0, 127.6, 127.5, 127.3, 127.1, 127.0, 126.7, 125.8, 125.2, 123.6, 89.7, 80.4, 24.2, 15.4, 10.5. **HRMS** (ESI) m/z calcd. for $C_{21}H_{17}^{+}$ $[M+H]^{+}$: 269.1325; Found: 269.1323.

(17) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.25-7.17 (m, 5H), 7.17-7.14 (m, 1H), 6.99-6.94 (m, 2H), 2.54 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.10-1.92 (m, 1H), 1.57-1.46 (m, 1H), 1.30-1.17 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 142.2, 131.5, 128.1, 127.5, 126.4, 125.3, 123.7, 123.4, 89.4, 80.1, 19.2, 17.1, 10.8. **HRMS** (ESI) m/z calcd. for $C_{15}H_{13}S^{+}$ $[M+H]^{+}$: 225.0733; Found: 225.0732.

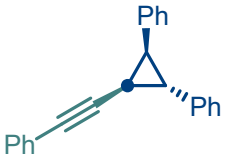
(18) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.43-7.38 (m, 4H), 7.26-7.22 (m, 6H), 2.01-1.87 (m, 2H), 1.41-1.36 (m, 1H), 1.20-1.15 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 131.8, 128.1, 127.7, 123.6, 89.0, 79.5, 17.7, 9.9. **HRMS** (ESI) m/z calcd. for $C_{19}H_{15}^{+}$ $[M+H]^{+}$: 243.1168; Found: 243.1165.

(19) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.38-7.34 (m, 2H), 7.29-7.22 (m, 3H), 2.12-2.03 (m, 1H), 1.85-1.78 (m, 1H), 1.78-1.71 (m, 2H), 1.69-1.62 (m, 1H), 1.56-1.52 (m, 1H), 1.30-1.07 (m, 6H), 1.01-0.95 (m, 1H), 0.89-0.79 (m, 1H), 0.54-0.47 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 131.6, 128.2, 127.3, 124.3, 91.4, 77.3, 39.6, 32.8, 32.7, 26.6, 26.33, 26.29, 25.2, 14.1, 5.4. **HRMS** (ESI) m/z calcd. for $C_{17}H_{21}^{+}$ $[M+H]^{+}$: 225.1638; Found: 225.1635.

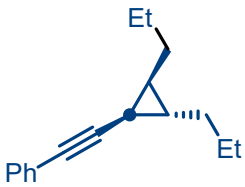
(20) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.45-7.41 (m, 2H), 7.38-7.31 (m, 2H), 7.26-7.22 (m, 1H), 7.17-7.12 (m, 3H), 7.04-6.98 (m, 2H), 1.75-1.71 (m, 1H), 1.48 (s, 3H), 1.40-1.38 (m, 1H), 1.24-1.21 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 142.4, 131.2, 129.1, 128.0, 127.2, 126.4, 123.9, 91.0, 80.4, 29.6, 27.2, 22.5, 15.8. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}^{+}$ $[M+H]^{+}$: 233.1325; Found: 233.1324.

(21) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.49-7.45 (m, 2H), 7.35-7.21 (m, 7H), 7.21-7.13 (m, 4H), 7.10-7.04 (m, 2H), 2.36 (dd, *J* = 9.0, 6.0 Hz, 1H), 1.79 (dd, *J* = 6.0, 5.0 Hz, 1H), 1.72 (dd, *J* = 9.0, 4.5 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 144.8, 141.0, 131.3, 130.3, 128.4, 128.03, 128.01, 127.99, 127.4, 126.7, 126.5, 123.7, 90.2, 81.7, 38.2, 23.5, 16.6. **HRMS** (ESI) m/z calcd. for $C_{23}H_{19}^{+}$ $[M+H]^{+}$: 295.1481; Found: 295.1482.

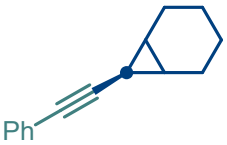
(22) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.39-7.34 (m, 2H), 7.28-7.21 (m, 3H), 1.70-1.53 (m, 7H), 1.50-1.42 (m, 1H), 1.41-1.35 (m, 1H), 1.29 (dd, *J* = 8.5, 5.0 Hz, 1H), 1.26-1.19 (m, 1H), 0.81 (dd, *J* = 8.5, 4.5 Hz, 1H), 0.63 (t, *J* = 5.0 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 131.5, 128.1, 127.2, 124.3, 91.5, 77.8, 36.2, 32.1, 27.1, 26.0, 25.5, 25.3, 22.4, 13.4. **HRMS** (ESI) m/z calcd. for $C_{16}H_{19}^{+}$ $[M+H]^{+}$: 211.1481; Found: 211.1480.



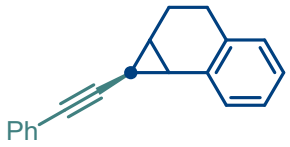
(23) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.25-7.20 (m, 5H), 7.20-7.16 (m, 6H), 7.15-7.09 (m, 4H), 2.82 (d, *J* = 8.5 Hz, 2H), 2.64 (t, *J* = 4.5 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 135.0, 131.3, 130.7, 128.1, 127.6, 127.4, 126.2, 123.9, 87.1, 85.9, 28.6, 15.2. **HRMS** (ESI) m/z calcd. for $C_{23}H_{19}^{+}$ $[M+H]^{+}$: 295.1481; Found: 295.1478.



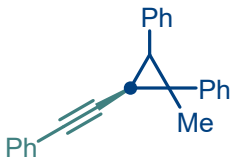
(24) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.39-7.34 (m, 2H), 7.28-7.20 (m, 3H), 1.58-1.37 (m, 7H), 1.29-1.23 (m, 1H), 1.22-1.14 (m, 1H), 1.01-0.9 (m, 6H), 0.87-0.78 (m, 2H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 131.5, 128.1, 127.2, 124.4, 91.2, 77.9, 35.7, 32.1, 29.3, 26.2, 22.4, 22.2, 14.1, 13.9, 13.0. **HRMS** (ESI) m/z calcd. for $C_{17}H_{23}^{+}$ $[M+H]^{+}$: 227.3710; Found: 227.3712.



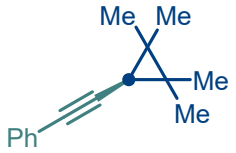
(25) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.43-7.39 (m, 2H), 7.30-7.23 (m, 3H), 1.98-1.90 (m, 2H), 1.78-1.72 (m, 2H), 1.59 (t, *J* = 8.4 Hz, 1H), 1.49-1.42 (m, 2H), 1.32-1.21 (m, 4H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 131.5, 128.2, 127.3, 124.3, 89.9, 82.6, 21.8, 20.1, 15.5, 11.1. **HRMS** (ESI) m/z calcd. for $C_{15}H_{17}^+$ $[M+H]^+$: 197.1325; Found: 197.1324.



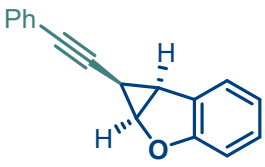
(26) Colorless oil, **^{1}H NMR** (600 MHz, $CDCl_3$) δ 7.33-7.30 (m, 1H), 7.20-7.12 (m, 5H), 7.08 (d, *J* = 7.8 Hz, 1H), 7.06-7.01 (m, 2H), 3.08-2.98 (m, 1H), 2.76-2.67 (m, 1H), 2.39 (t, *J* = 8.4 Hz, 1H), 2.22-2.15 (m, 1H), 2.13-2.06 (m, 1H), 2.04 (t, *J* = 8.4 Hz, 1H), 1.87-1.80 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 136.5, 133.7, 131.2, 130.0, 128.2, 128.0, 127.3, 125.9, 125.8, 123.9, 88.8, 82.1, 27.2, 21.9, 19.8, 19.1, 15.4. **HRMS** (ESI) m/z calcd. for $C_{19}H_{17}^+$ $[M+H]^+$: 245.1325; Found: 245.1324.



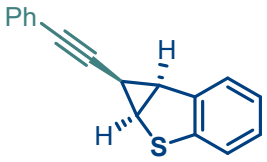
(27) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.59-7.55 (m, 2H), 7.49-7.45 (m, 2H), 7.39-7.34 (m, 4H), 7.34-7.30 (m, 2H), 7.30-7.24 (m, 5H), 2.76 (d, *J* = 9.0 Hz, 1H), 2.39 (d, *J* = 9.0 Hz, 1H), 1.39 (s, 3H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 146.9, 136.0, 131.4, 130.6, 128.6, 128.2, 128.0, 127.60, 127.55, 126.52, 126.48, 124.0, 88.0, 84.0, 33.6, 32.9, 20.4, 19.6. **HRMS** (ESI) m/z calcd. for $C_{24}H_{21}^+$ $[M+H]^+$: 309.1638; Found: 309.16379.



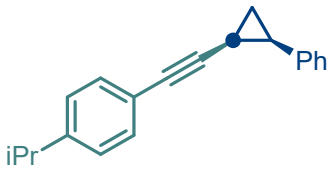
(28) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.41-7.36 (m, 2H), 7.28-7.22 (m, 3H), 1.17 (s, 12H), 1.07 (s, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 131.5, 128.1, 127.2, 124.5, 90.0, 80.8, 26.5, 25.5, 22.5, 18.5. **HRMS** (ESI) m/z calcd. for $C_{15}H_{19}^+$ $[M+H]^+$: 199.1481; Found: 199.1479.



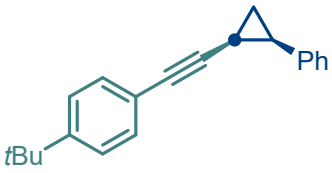
(29) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.40 (dd, *J* = 7.5, 1.5 Hz, 1H), 7.20 (td, *J* = 8.0, 1.5 Hz, 1H), 7.18-7.09 (m, 3H), 6.95 (td, *J* = 7.5, 1.0 Hz, 1H), 6.93-6.89 (m, 3H), 4.96 (t, *J* = 5.5 Hz, 1H), 3.17 (dd, *J* = 8.5, 5.0 Hz, 1H), 1.89 (dd, *J* = 8.5, 5.5 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 160.4, 131.3, 127.91, 127.88, 127.4, 126.4, 125.0, 123.2, 120.9, 109.4, 83.3, 80.6, 64.6, 28.1, 8.2. **HRMS** (ESI) m/z calcd. for $C_{17}H_{13}O^+$ $[M+H]^+$: 233.0966; Found: 233.0964.



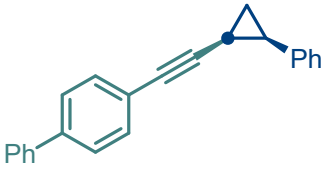
(30) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.39 (d, *J* = 8.0 Hz, 1H), 7.23-7.18 (m, 2H), 7.17-7.11 (m, 3H), 7.10-7.06 (m, 1H), 6.99-6.94 (m, 2H), 3.48 (t, *J* = 7.0 Hz, 1H), 3.43 (t, *J* = 8.0 Hz, 1H), 2.15 (dd, *J* = 8.0, 7.0 Hz, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 143.0, 136.6, 131.5, 127.9, 127.6, 127.5, 126.0, 124.1, 123.3, 121.2, 84.4, 81.9, 35.3, 31.4, 9.2. **HRMS** (ESI) m/z calcd. for $C_{17}H_{13}S^+$ $[M+H]^+$: 249.0732; Found: 249.0730.



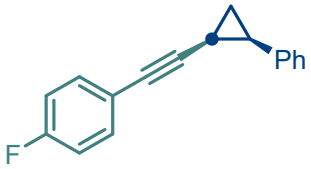
(31) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.34-7.29 (m, 4H), 7.25-7.20 (m, 1H), 7.09-7.03 (m, 4H), 2.88-2.76 (m, 1H), 2.44-2.33 (m, 1H), 2.02-1.93 (m, 1H), 1.48-1.39 (m, 1H), 1.29-1.24 (m, 1H), 1.19 (s, 3H), 1.18 (s, 3H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 148.3, 138.1, 131.4, 128.3, 127.7, 126.19, 126.16, 121.1, 88.8, 80.3, 33.9, 23.9, 23.8, 15.1, 10.2. **HRMS** (ESI) m/z calcd. for $C_{20}H_{21}^+$ $[M+H]^+$: 261.1638; Found: 261.1637.



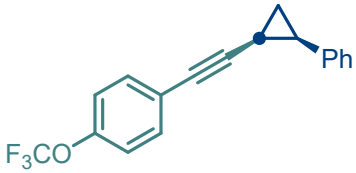
(32) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.33-7.30 (m, 4H), 7.25-7.19 (m, 3H), 7.09-7.05 (m, 2H), 2.42-2.35 (m, 1H), 2.01-1.93 (m, 1H), 1.47-1.40 (m, 1H), 1.28-1.25 (m, 1H), 1.26 (s, 9H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 150.5, 138.1, 131.1, 128.3, 127.7, 126.2, 125.0, 120.7, 88.9, 80.2, 34.6, 31.1, 23.9, 15.1, 10.3. **HRMS** (ESI) m/z calcd. for $C_{21}H_{23}^+$ $[M+H]^+$: 275.1794; Found: 275.1791.



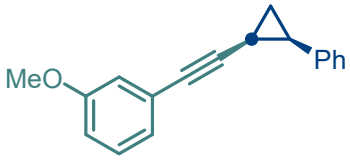
(33) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.52 (d, *J* = 7.5 Hz, 2H), 7.44-7.38 (m, 4H), 7.36-7.28 (m, 5H), 7.27-7.22 (m, 1H), 7.20 (d, *J* = 8.0 Hz, 2H), 2.41 (dd, *J* = 15.0, 8.5 Hz, 1H), 2.05-1.95 (m, 1H), 1.50-1.42 (m, 1H), 1.34-1.28 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 140.5, 140.1, 138.0, 131.8, 128.8, 128.3, 127.8, 127.4, 126.9, 126.7, 126.3, 122.7, 90.5, 80.1, 24.1, 15.1, 10.3. **HRMS** (ESI) m/z calcd. for $C_{23}H_{19}^+$ $[M+H]^+$: 295.1481; Found: 295.1483.



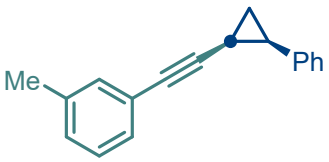
(34) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.35-7.28 (m, 4H), 7.27-7.21 (m, 1H), 7.11-7.06 (m, 2H), 6.90-6.84 (m, 2H), 2.44-2.36 (m, 1H), 1.99-1.93 (m, 1H), 1.48-1.41 (m, 1H), 1.32-1.26 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 161.9 (d, J = 246.3 Hz), 137.9, 133.1 (d, J = 8.0 Hz), 128.3, 127.8, 126.3, 119.8, 115.2 (d, J = 21.3 Hz), 89.4, 79.1, 53.4, 23.9, 14.9, 10.1. **HRMS** (ESI) m/z calcd. for $C_{17}H_{14}F^+$ $[M+H]^+$: 237.1074; Found: 237.1074.



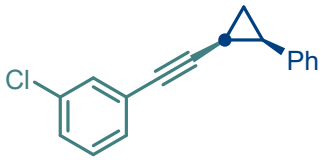
(35) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.35-7.28 (m, 4H), 7.27-7.22 (m, 1H), 7.14-7.10 (m, 2H), 7.05-7.00 (m, 2H), 2.47-2.39 (m, 1H), 2.01-1.93 (m, 1H), 1.49-1.42 (m, 1H), 1.33-1.27 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 148.2, 137.8, 132.8, 128.4, 127.8, 126.4, 122.6, 120.6, 120.3 (d, J = 256.3 Hz), 90.8, 78.8, 24.0, 14.9, 10.0. **^{19}F NMR** (564 MHz, $CDCl_3$) δ -57.9. **HRMS** (ESI) m/z calcd. for $C_{18}H_{14}F_3O^+$ $[M+H]^+$: 303.0991; Found: 303.0988.



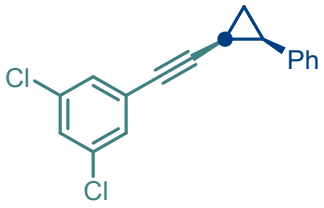
(36) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.34-7.31 (m, 4H), 7.25-7.21 (m, 1H), 7.11-7.07 (m, 1H), 6.77-6.72 (m, 2H), 6.64-6.62 (m, 1H), 3.72 (s, 3H), 2.46-2.38 (m, 1H), 2.01-1.95 (m, 1H), 1.49-1.42 (m, 1H), 1.32-128 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 158.1, 137.0, 128.1, 127.4, 126.7, 125.3, 123.8, 122.9, 115.2, 113.1, 88.7, 79.1, 54.1, 23.0, 14.0, 9.1. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}O^+$ $[M+H]^+$: 249.1274; Found: 249.1272.



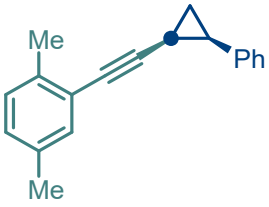
(37) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.35-7.28 (m, 4H), 7.25-7.21 (m, 1H), 7.07 (t, J = 7.5 Hz, 1H), 7.00 (d, J = 8.0 Hz, 1H), 6.97-6.92 (m, 2H), 2.43-2.35 (m, 1H), 2.24 (s, 3H), 2.01-1.94 (m, 1H), 1.47-1.41 (m, 1H), 1.30-1.24 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.0, 137.6, 132.0, 128.4, 128.32, 128.26, 127.9, 127.7, 126.2, 123.6, 89.3, 80.3, 24.0, 21.1, 15.1, 10.2. **HRMS** (ESI) m/z calcd. for $C_{18}H_{17}^+$ $[M+H]^+$: 233.1325; Found: 233.1324.



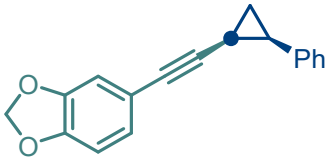
(38) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.36-7.28 (m, 4H), 7.27-7.23 (m, 1H), 7.18-7.14 (m, 1H), 7.13-7.07 (m, 2H), 7.01-6.97 (m, 1H), 2.47-2.38 (m, 1H), 2.01-1.93 (m, 1H), 1.49-1.43 (m, 1H), 1.33-1.27 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 137.8, 133.8, 131.3, 129.5, 129.2, 128.3, 127.8, 127.6, 126.4, 125.5, 91.3, 78.8, 24.1, 14.9, 10.1. **HRMS** (ESI) m/z calcd. for $C_{17}H_{14}Cl^+$ $[M+H]^+$: 253.0779; Found: 253.0778.



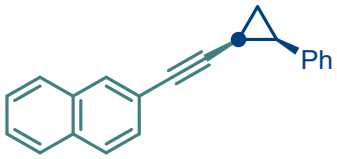
(39) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.37-7.31 (m, 2H), 7.31-7.24 (m, 3H), 7.17 (t, J = 2.0 Hz, 1H), 6.95 (d, J = 2.0 Hz, 2H), 2.50-2.41 (m, 1H), 1.99-1.91 (m, 1H), 1.49-1.44 (m, 1H), 1.34-1.29 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 137.6, 134.5, 129.6, 128.3, 127.9, 127.7, 126.60, 126.55, 93.0, 77.7, 24.2, 14.9, 10.0. **HRMS** (ESI) m/z calcd. for $C_{17}H_{13}Cl_2^+$ $[M+H]^+$: 287.0389; Found: 287.0388.



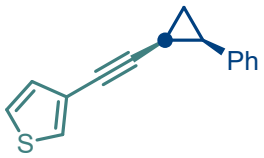
(40) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.34-7.26 (m, 4H), 7.22-7.17 (m, 1H), 7.04 (d, J = 8.0 Hz, 1H), 6.87 (s, 1H), 6.82 (d, J = 7.5 Hz, 1H), 2.40 (dd, J = 15.0, 8.0 Hz, 1H), 2.24 (s, 3H), 2.05-2.01 (m, 1H), 2.00 (s, 3H), 1.47-1.40 (m, 1H), 1.31-1.26 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 139.8, 138.1, 137.2, 131.8, 129.9, 128.5, 127.8, 126.2, 126.0, 120.5, 92.5, 79.0, 24.0, 21.3, 20.2, 14.8, 10.4. **HRMS** (ESI) m/z calcd. for $C_{19}H_{19}^+$ $[M+H]^+$: 247.1481; Found: 247.1480.



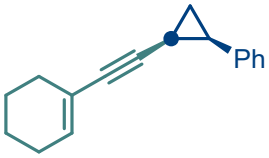
(41) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.36-7.28 (m, 4H), 7.25-7.21 (m, 1H), 6.68-6.61 (m, 2H), 6.57 (d, J = 1.0 Hz, 1H), 5.90 (s, 2H), 2.42-2.35 (m, 1H), 1.98-1.92 (m, 1H), 1.46-1.39 (m, 1H), 1.29-1.24 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 147.13, 147.08, 138.0, 128.3, 127.7, 126.3, 125.7, 117.1, 111.5, 108.2, 101.0, 87.9, 79.9, 23.9, 15.0, 10.1. **HRMS** (ESI) m/z calcd. for $C_{18}H_{15}O_2^+$ $[M+H]^+$: 263.1072; Found: 263.1071.



(42) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.74 (d, *J* = 8.0 Hz, 1H), 7.69 (d, *J* = 8.5 Hz, 1H), 7.54 (d, *J* = 8.5 Hz, 1H), 7.43-7.39 (m, 4H), 7.36 (t, *J* = 7.0 Hz, 2H), 7.33-7.27 (m, 3H), 2.56-2.45 (m, 1H), 2.18-2.09 (m, 1H), 1.54-1.48 (m, 1H), 1.46-1.40 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 138.0, 133.3, 133.0, 130.0, 128.8, 128.1, 127.9, 127.7, 126.5, 126.4, 126.3, 126.0, 125.1, 121.5, 94.7, 78.0, 24.2, 14.5, 10.4. **HRMS** (ESI) m/z calcd. for $C_{21}H_{17}^{+}$ $[M+H]^{+}$: 269.1324; Found: 269.1323.

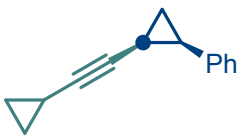


(43) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.35-7.27 (m, 4H), 7.25-7.20 (m, 1H), 7.09 (d, *J* = 5.0 Hz, 1H), 6.91 (d, *J* = 3.5 Hz, 1H), 6.87-6.83 (m, 1H), 2.47-2.31 (m, 1H), 2.08-1.94 (m, 1H), 1.53-1.41 (m, 1H), 1.33-1.28 (m, 1H). **^{13}C NMR** (150 MHz, $CDCl_3$) δ 137.8, 131.0, 128.2, 127.8, 126.6, 126.3, 125.9, 124.0, 93.8, 73.3, 24.2, 15.2, 10.4. **HRMS** (ESI) m/z calcd. for $C_{15}H_{13}S^{+}$ $[M+H]^{+}$: 225.0733; Found: 225.0731.

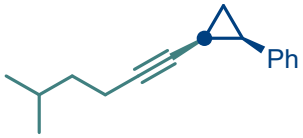


(44) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.31-7.24 (m, 4H), 7.20 (t, *J* = 7.0 Hz, 1H), 5.85-5.73 (m, 1H), 2.30 (dd, *J* = 15.5, 8.5 Hz, 1H), 2.02-1.96 (m, 2H), 1.92-1.83 (m, 3H), 1.56-1.46 (m, 4H), 1.42-1.33 (m, 1H), 1.20-1.12 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.2, 133.1, 128.3, 127.6, 126.1, 120.8, 86.6, 82.1, 29.1, 25.5, 23.8, 22.3, 21.5, 15.1, 10.2. **HRMS** (ESI) m/z calcd. for $C_{17}H_{19}^{+}$ $[M+H]^{+}$: 223.1481; Found: 223.1481.

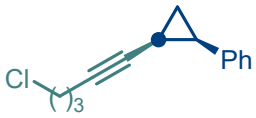
(45) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.29-7.19 (m, 7H), 7.19-7.16 (m, 1H), 7.05 (d, *J* = 7.0 Hz, 2H), 2.62-2.56 (m, 2H), 2.33-2.25 (m, 2H), 2.25-2.19 (m 1H), 1.78-1.71 (m, 1H), 1.33-1.28 (m, 1H), 1.09-1.04 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 140.9, 138.3, 128.4, 128.3, 128.2, 127.6, 126.1, 79.9, 79.7, 35.3, 23.2, 20.9, 14.6, 9.8. **HRMS** (ESI) m/z calcd. for $C_{19}H_{19}^{+}$ $[M+H]^{+}$: 247.1481; Found: 247.1473.



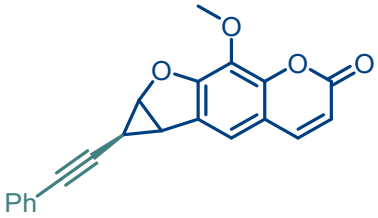
(46) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.31-7.26 (m, 2H), 7.24-7.18 (m, 3H), 2.21 (dd, *J* = 15.0, 8.5 Hz, 1H), 1.76-1.68 (m, 1H), 132-1.26 (m, 1H), 1.13-0.99 (m, 2H), 0.60-0.52 (m, 2H), 0.40-0.30 (m, 2H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.4, 128.3, 127.6, 126.0, 83.5, 74.6, 23.3, 14.8, 9.7, 8.0, 7.8, **HRMS** (ESI) m/z calcd. for $C_{14}H_{15}^{+}$ $[M+H]^{+}$: 183.1174; Found: 183.1170.



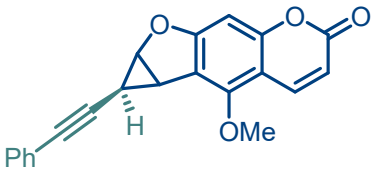
(47) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.30-7.23 (m, 4H), 7.22-7.19 (m, 1H), 2.29-2.15 (m, 1H), 2.03-2.00 (m, 2H), 1.79-1.71 (m, 1H), 1.46-1.36 (m, 1H), 1.32-1.26 (m, 1H), 1.20-1.13 (m, 2H), 1.11-1.07 (m, 1H), 0.77 (d, *J* = 2.0 Hz, 3H), 0.75 (d, *J* = 2.0 Hz, 3H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.4, 128.3, 127.6, 126.0, 80.4, 79.0, 37.7, 26.7, 23.1, 22.1, 16.6, 14.5, 9.8. **HRMS** (ESI) m/z calcd. for $C_{16}H_{21}^{+}$ $[M+H]^{+}$: 213.1643; Found: 213.1640.



(48) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.32-7.18 (m, 5H), 3.37 (t, *J* = 6.5 Hz, 2H), 2.89-2.21 (m, 1H), 2.08-2.01 (m, 2H), 1.78-1.70 (m, 1H), 1.57-1.50 (m, 2H), 1.42-1.35 (m, 2H), 1.32-1.26 (m, 1H), 1.14-1.07 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 138.3, 128.4, 127.7, 126.1, 80.0, 79.2, 44.7, 31.0, 25.8, 23.1, 17.9, 14.2, 9.6. **HRMS** (ESI) m/z calcd. for $C_{15}H_{18}Cl^{+}$ $[M+H]^{+}$: 233.1092; Found: 233.1090.

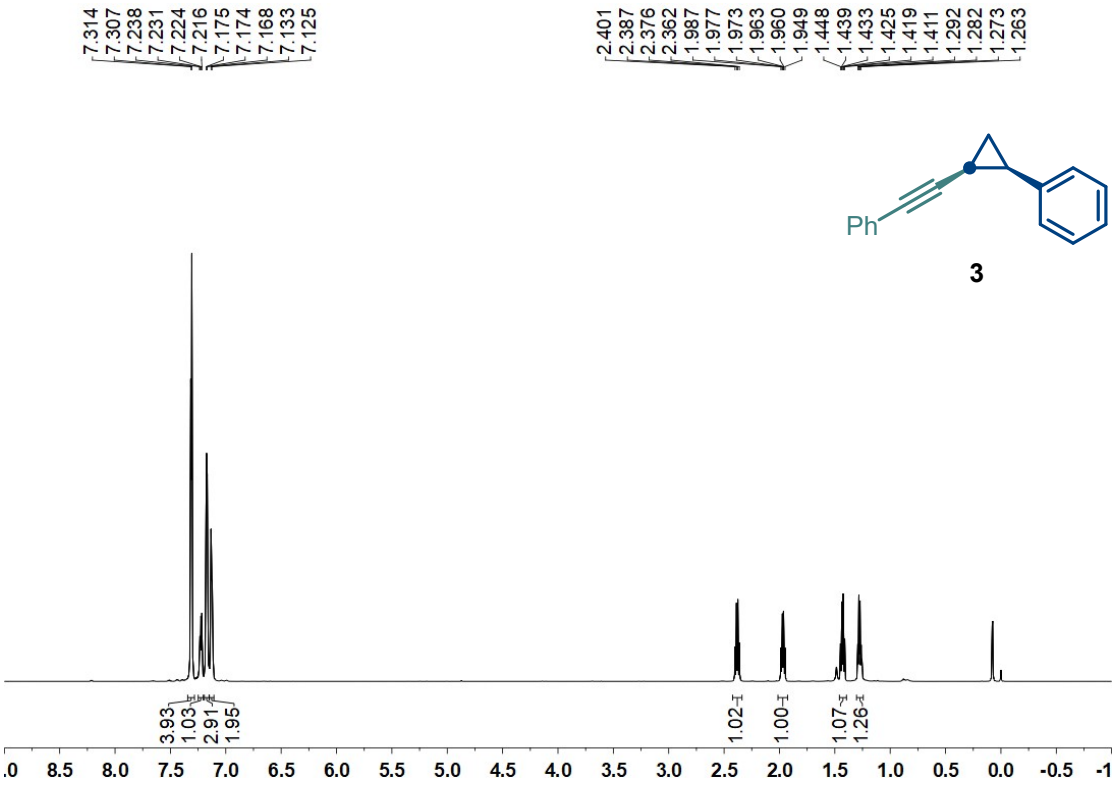


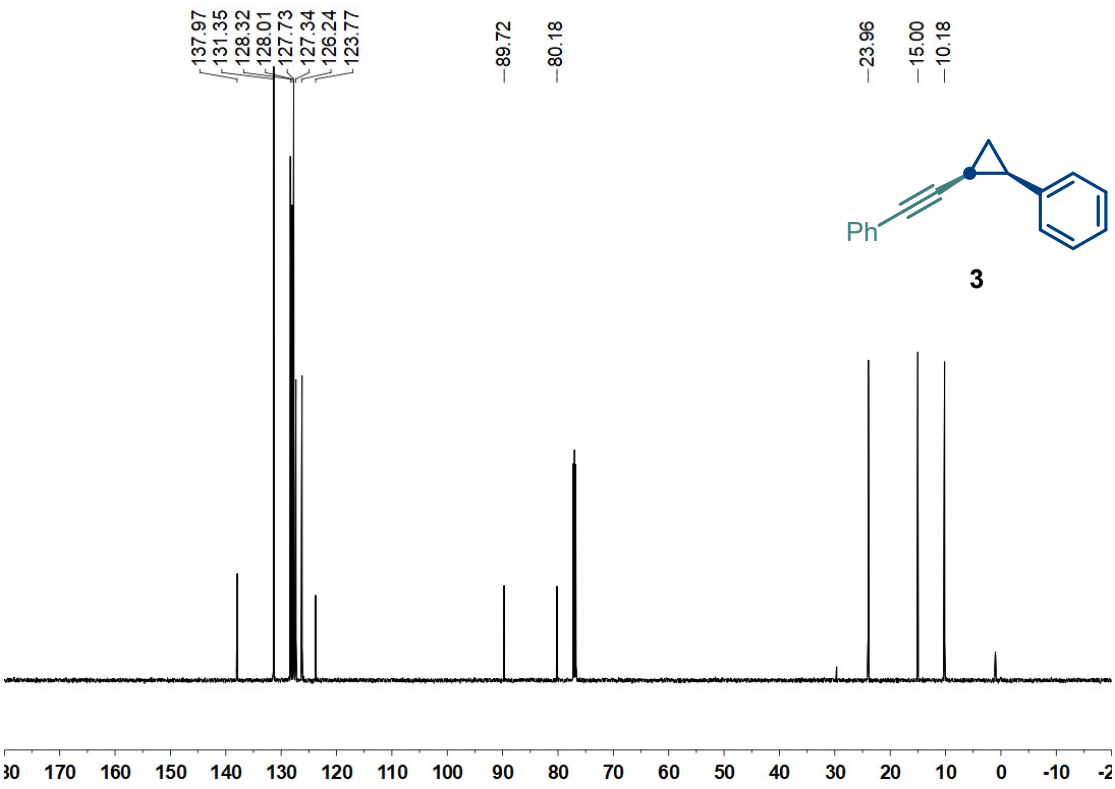
(50) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.99 (d, *J* = 10.0 Hz, 1H), 7.23-7.12 (m, 3H), 7.01-6.95 (m, 2H), 6.59 (s, 1H), 6.19 (d, *J* = 10.0 Hz, 1H), 5.06 (t, *J* = 5.5 Hz, 1H), 4.17 (s, 3H), 3.39 (dd, *J* = 8.5, 5.5 Hz, 1H), 2.14-2.00 (m, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 165.5, 161.4, 156.0, 153.8, 139.0, 131.3, 128.2, 128.0, 122.6, 111.3, 110.5, 106.1, 92.9, 81.9, 66.4, 60.4, 26.4, 9.4. **HRMS** (ESI) m/z calcd. for $C_{21}H_{15}O_4^{+}$ $[M+H]^{+}$: 331.0965; Found: 331.0962.

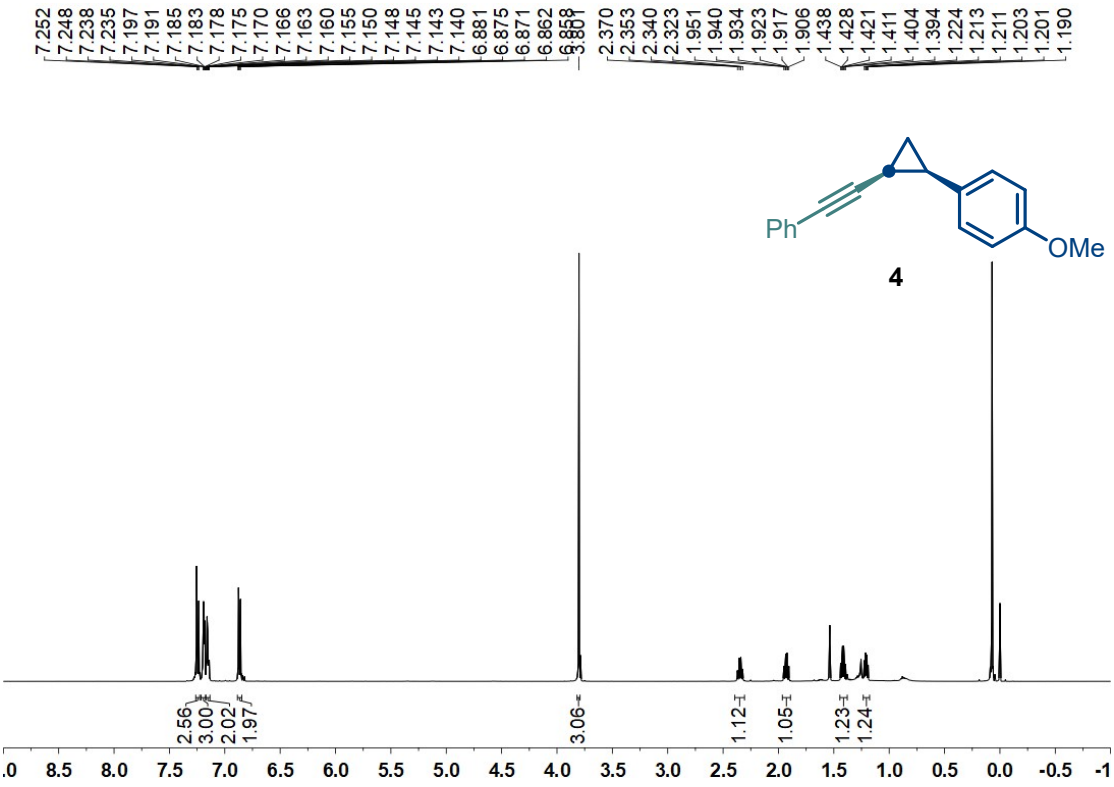


(52) Colorless oil, **^{1}H NMR** (500 MHz, $CDCl_3$) δ 7.64 (d, *J* = 9.5 Hz, 1H), 7.22–7.13 (m, 4H), 6.97 (dd, *J* = 8.0, 1.0 Hz, 2H), 6.28 (d, *J* = 9.5 Hz, 1H), 5.14 (t, *J* = 5.0 Hz, 1H), 4.00 (s, 3H), 3.20 (dd, *J* = 8.5, 5.0 Hz, 1H), 2.06 (dd, *J* = 8.5, 5.5 Hz, 1H). **^{13}C NMR** (125 MHz, $CDCl_3$) δ 160.7, 154.1, 147.1, 143.9, 131.6, 131.3, 128.2, 128.0, 125.2, 122.6, 117.4, 114.0, 113.2, 82.3, 81.5, 66.4, 60.9, 27.5, 9.2. **HRMS** (ESI) m/z calcd. for $C_{21}H_{15}O_4^+$ $[M+H]^+$: 331.0965; Found: 331.0962.

VI. Copies of NMR spectra



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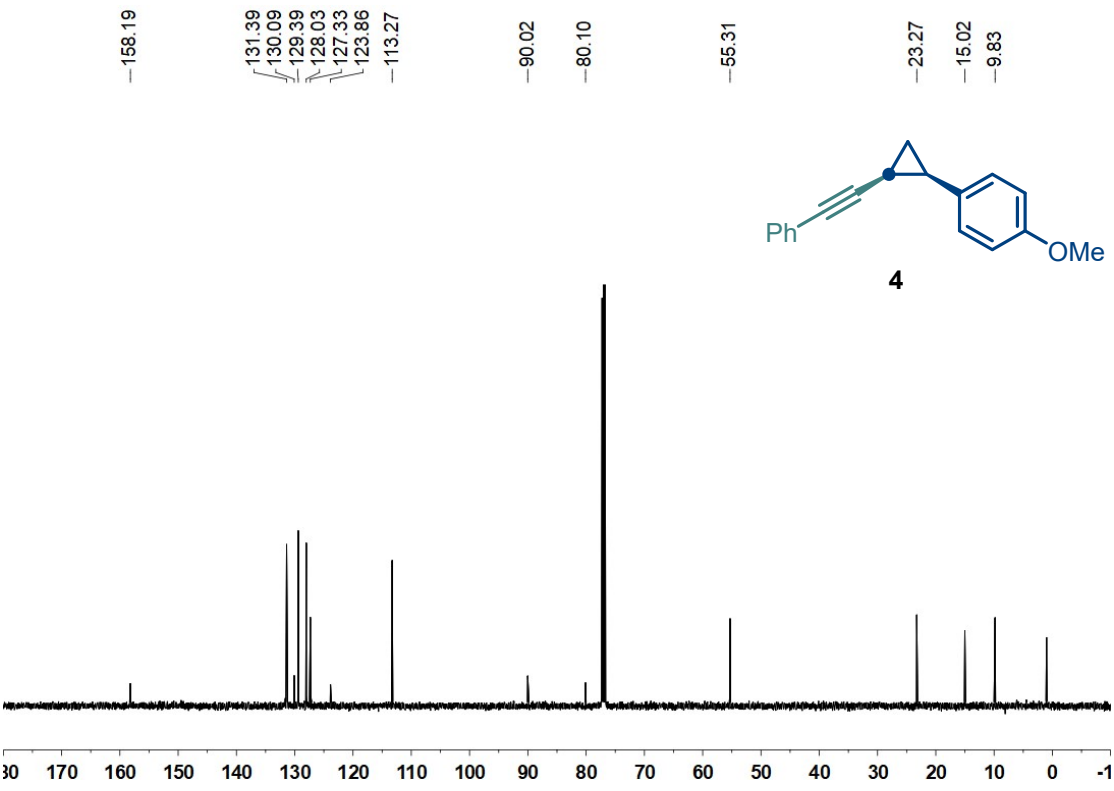
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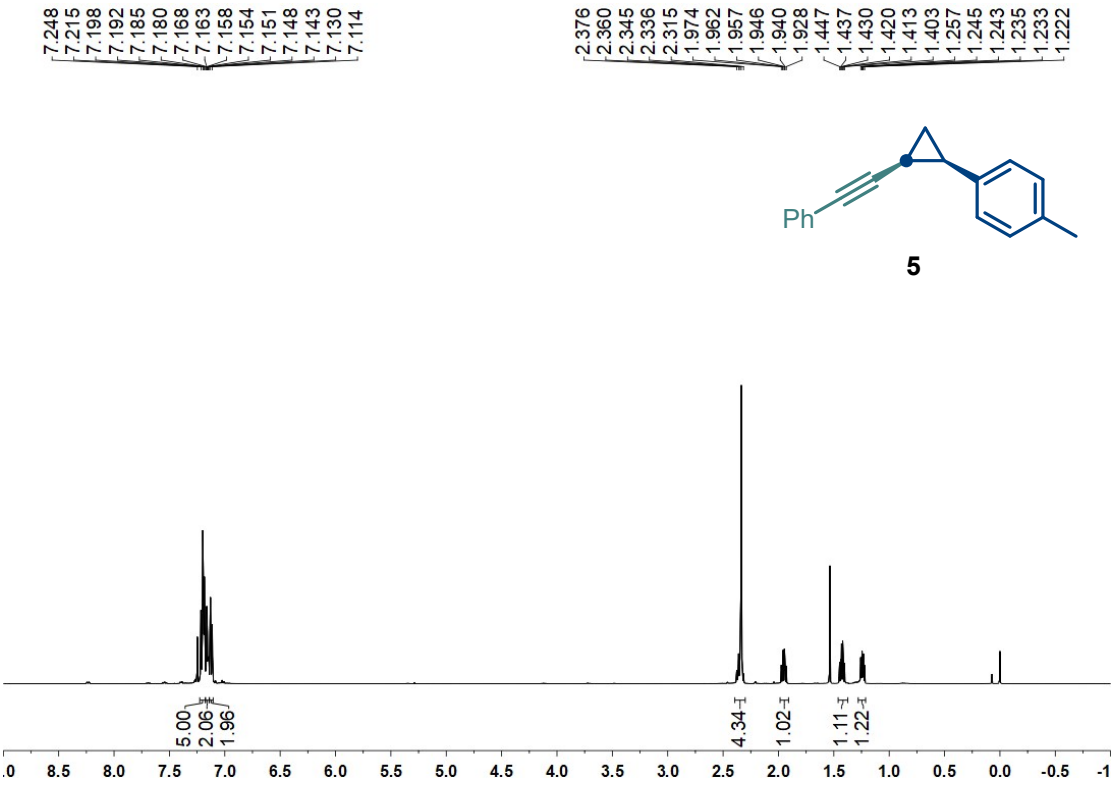
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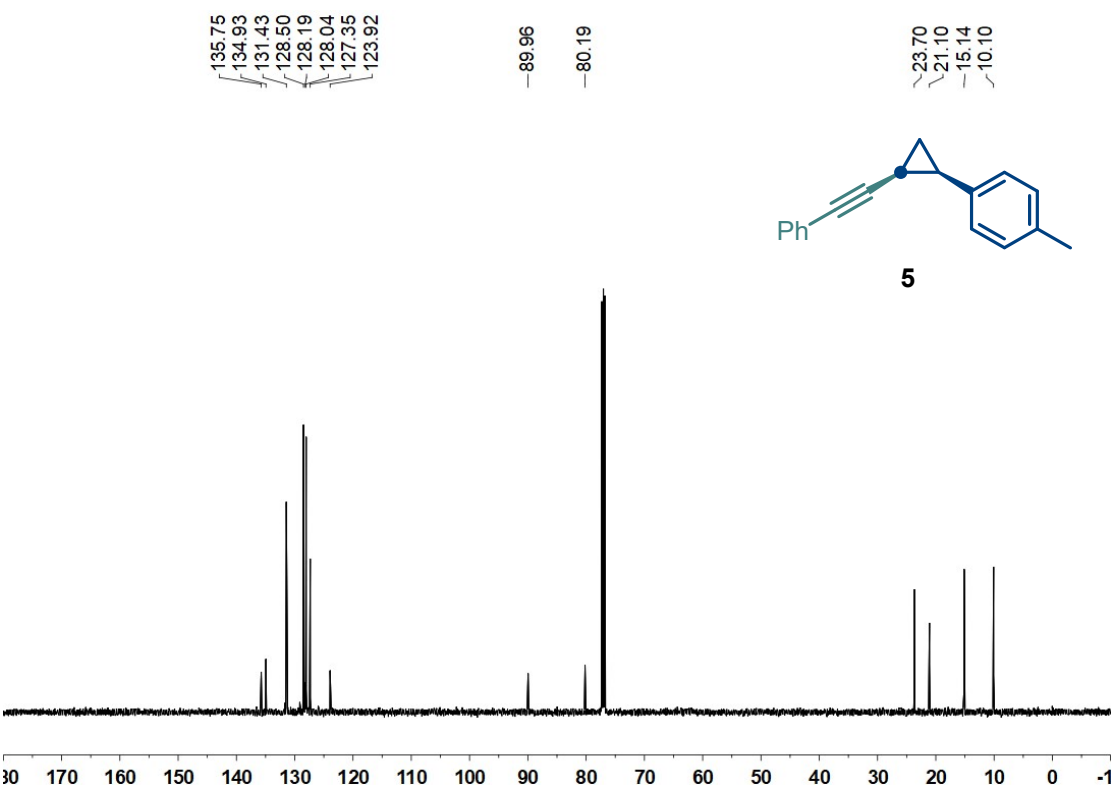
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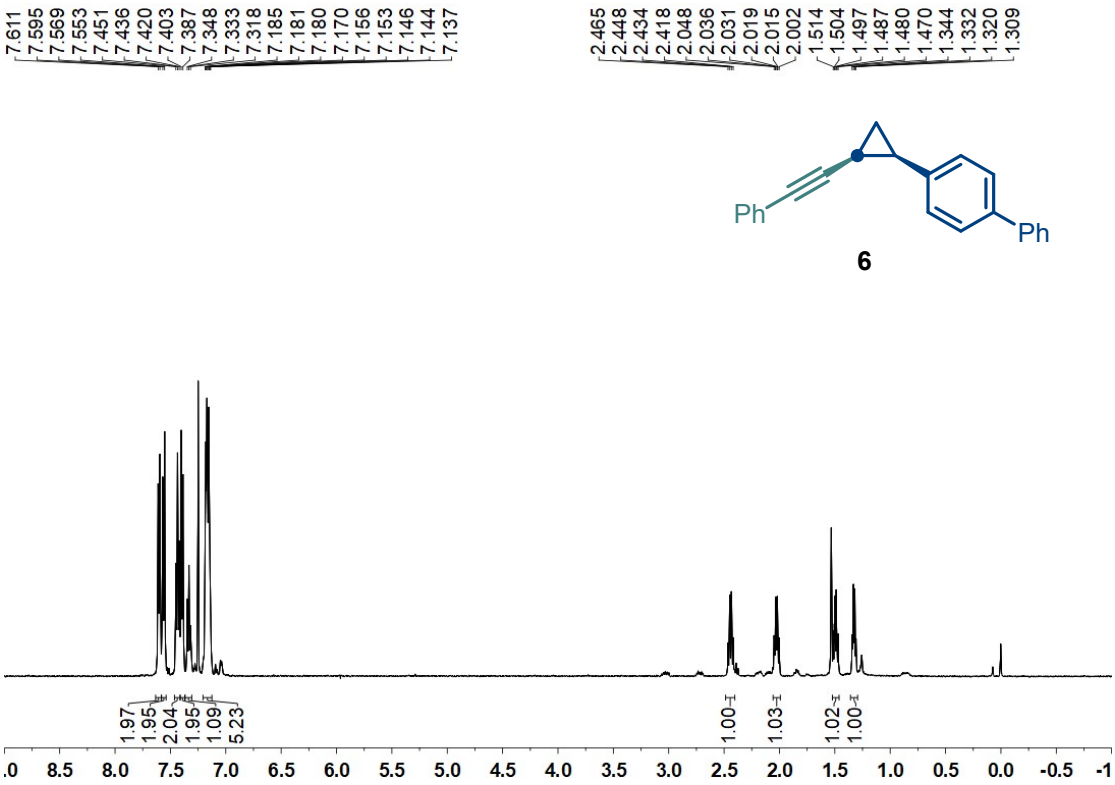
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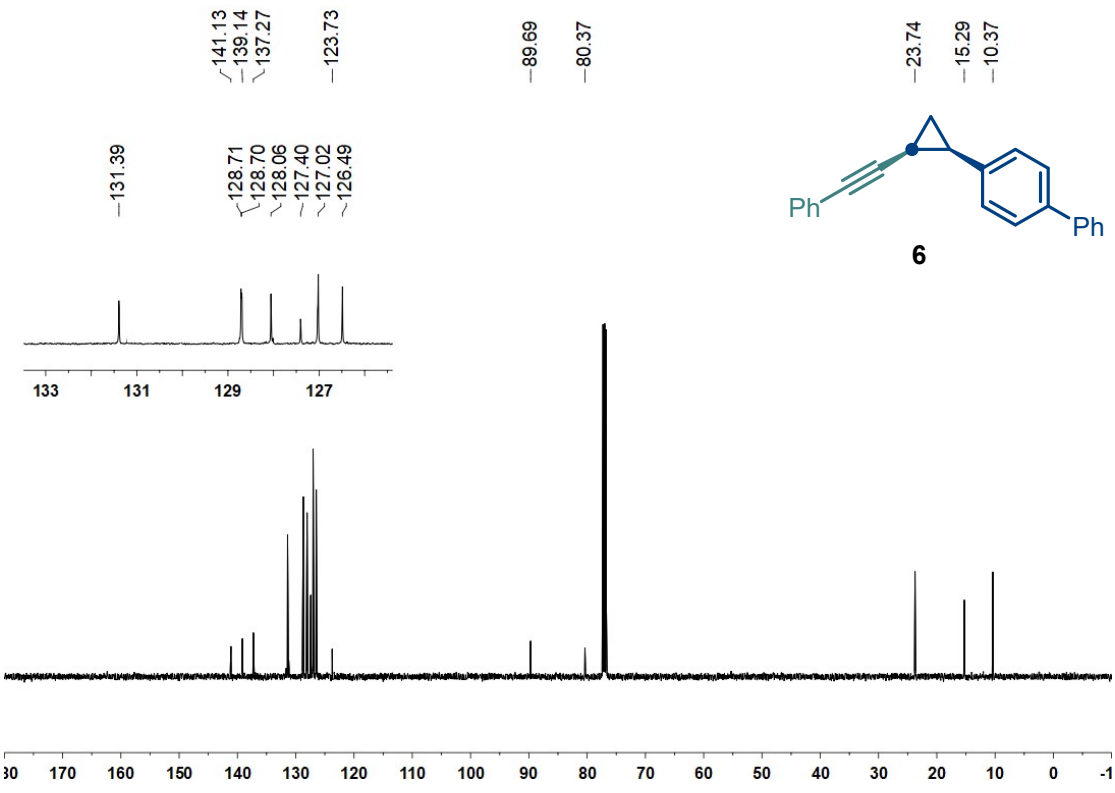
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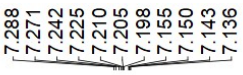
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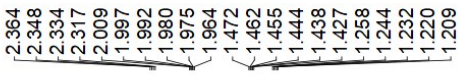
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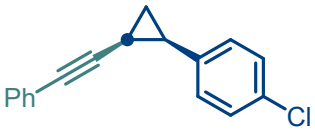
Ph
Cl

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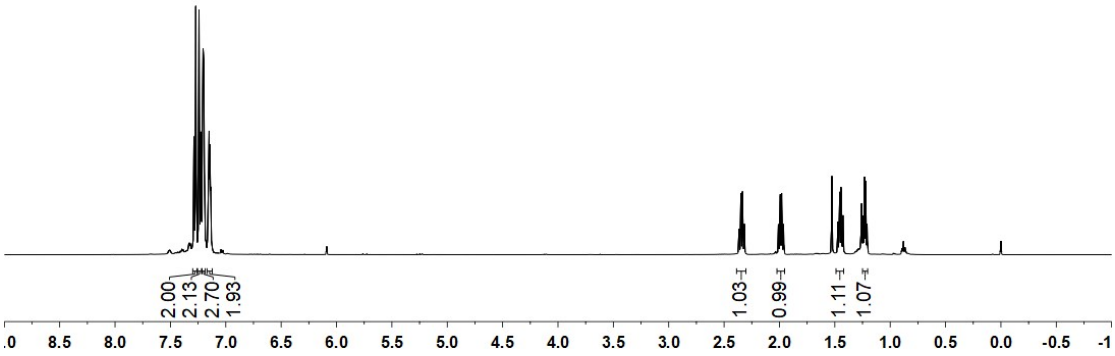
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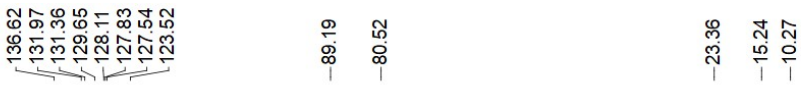
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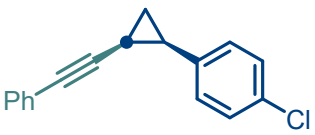
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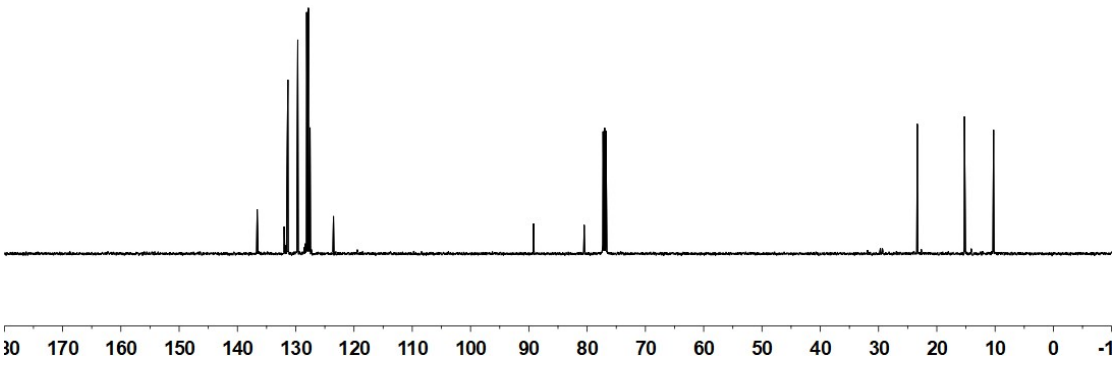
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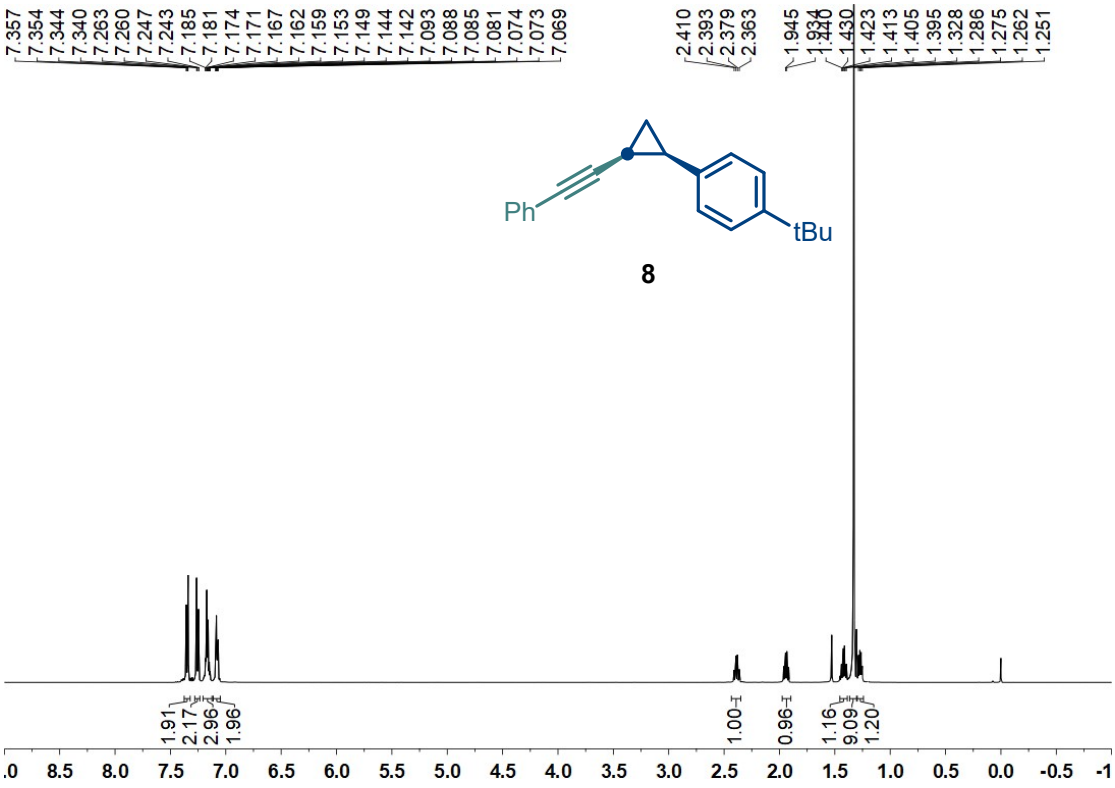
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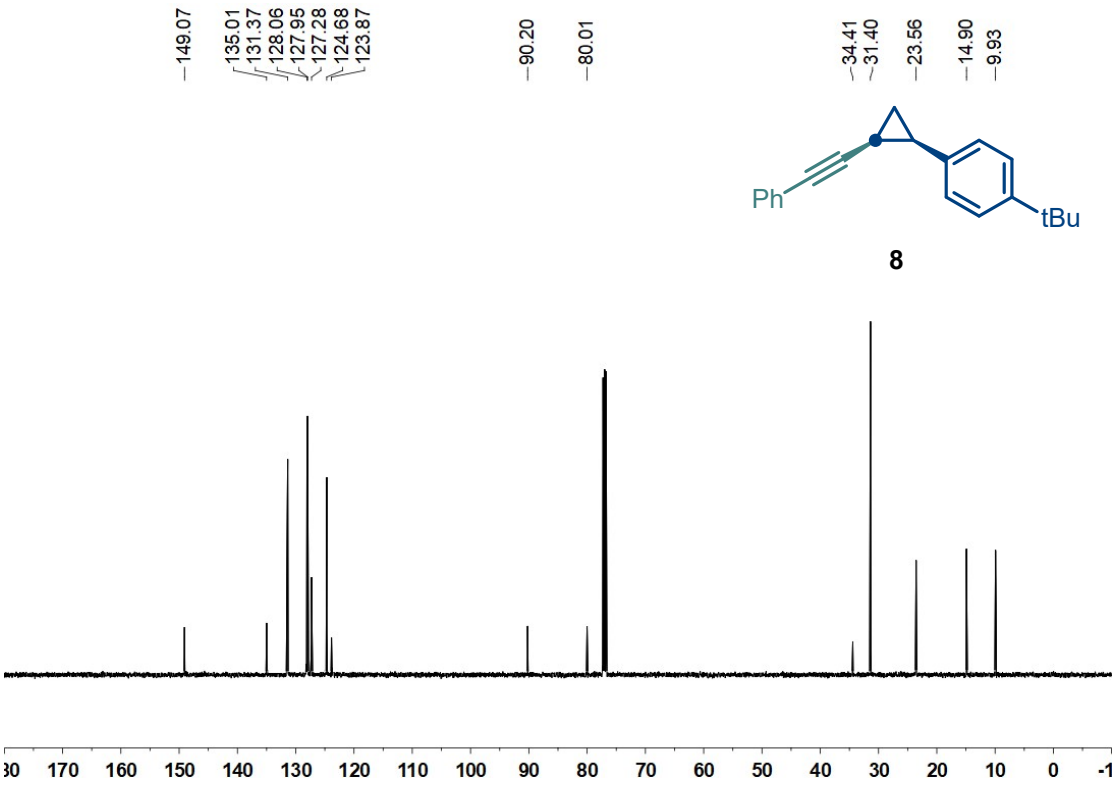
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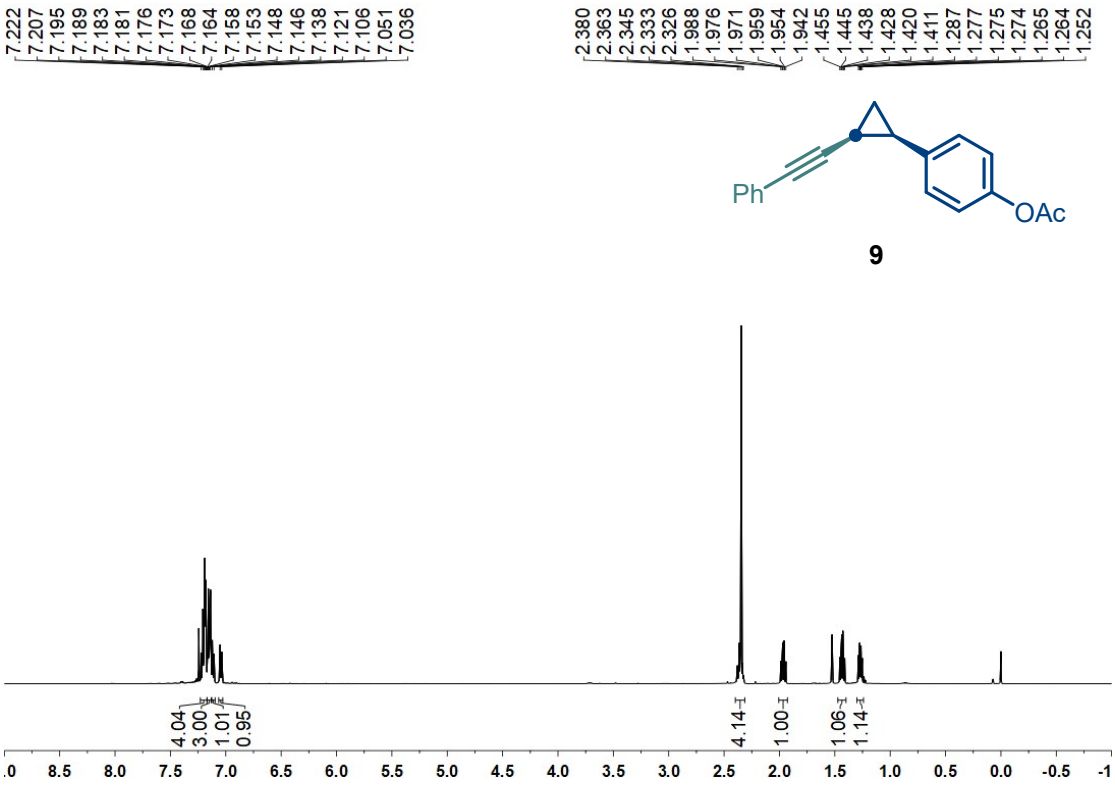
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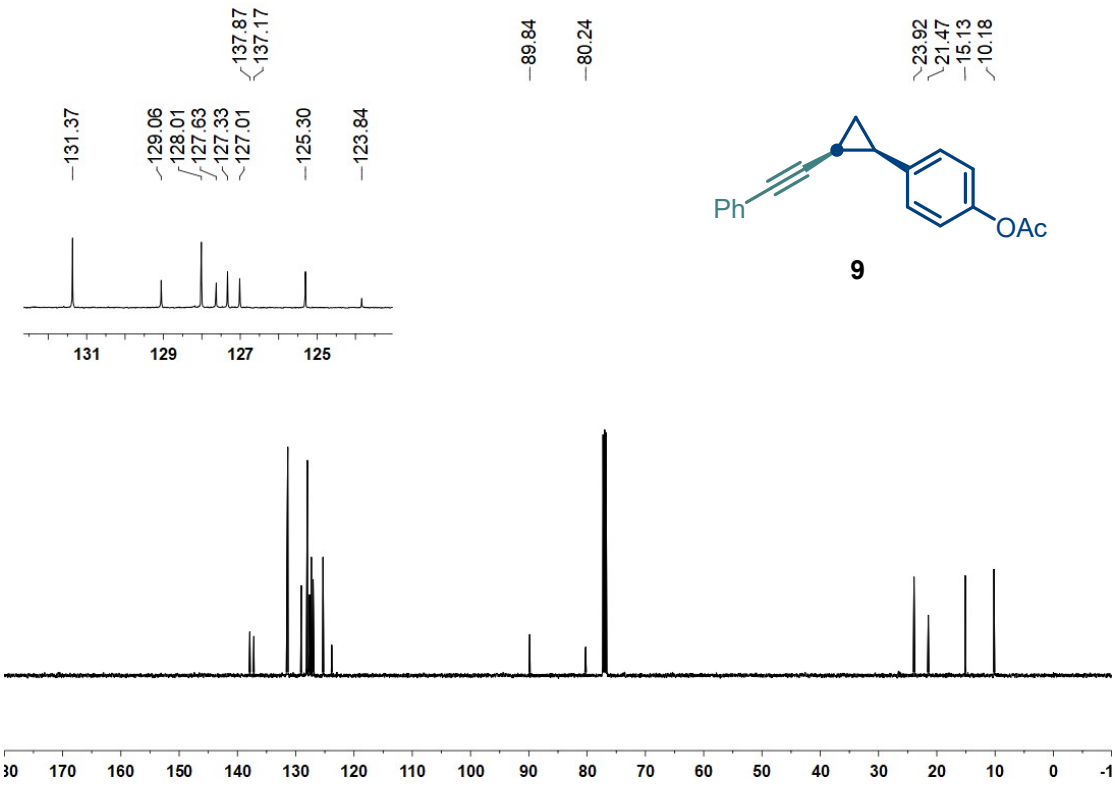
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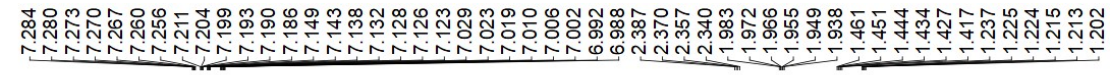
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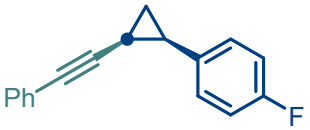
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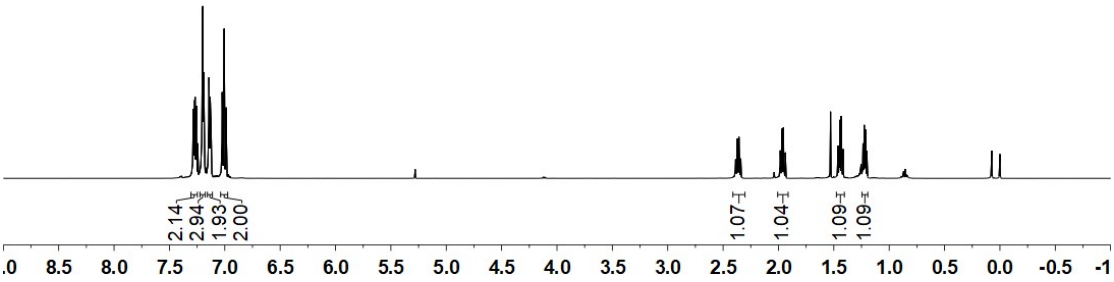
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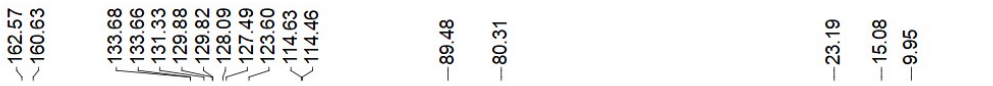
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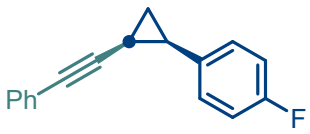
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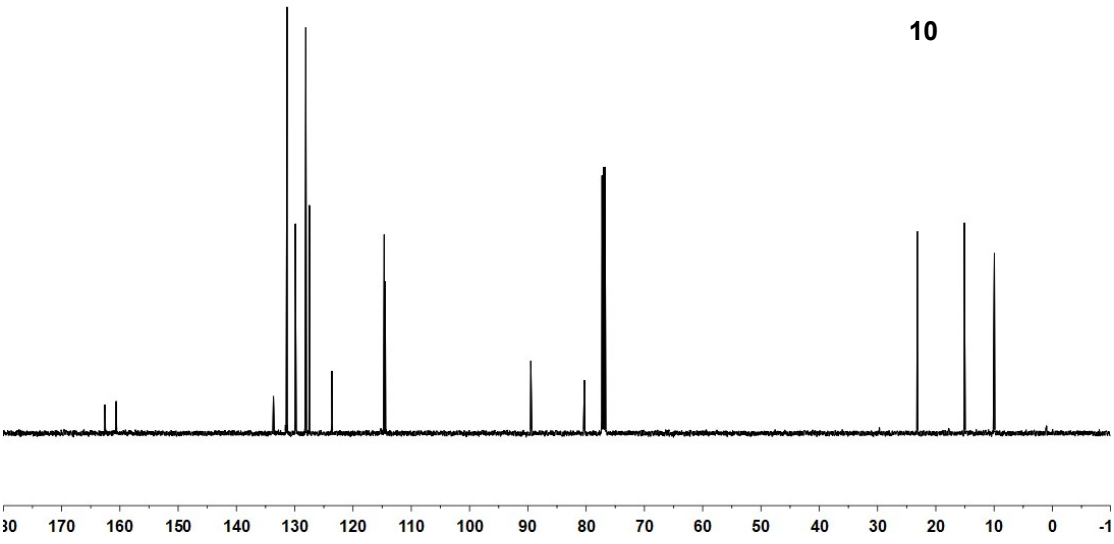
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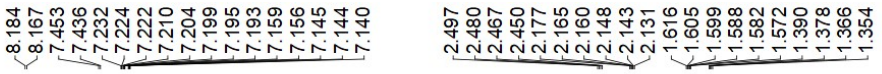
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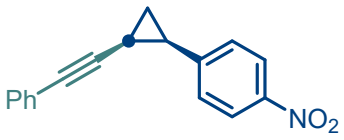
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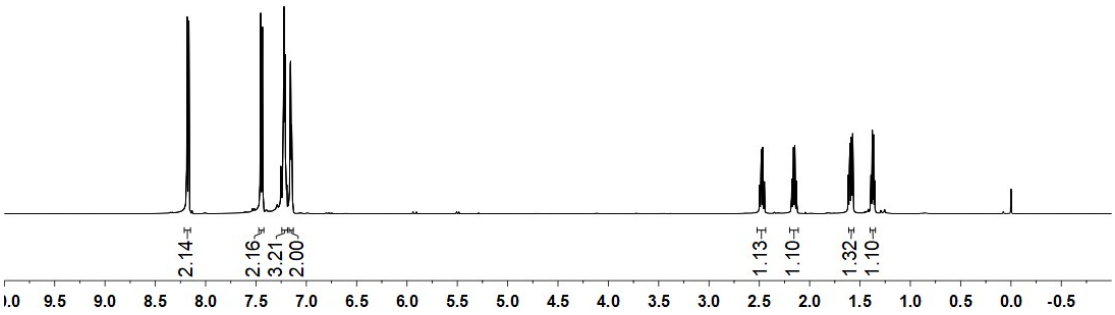
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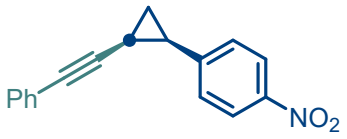
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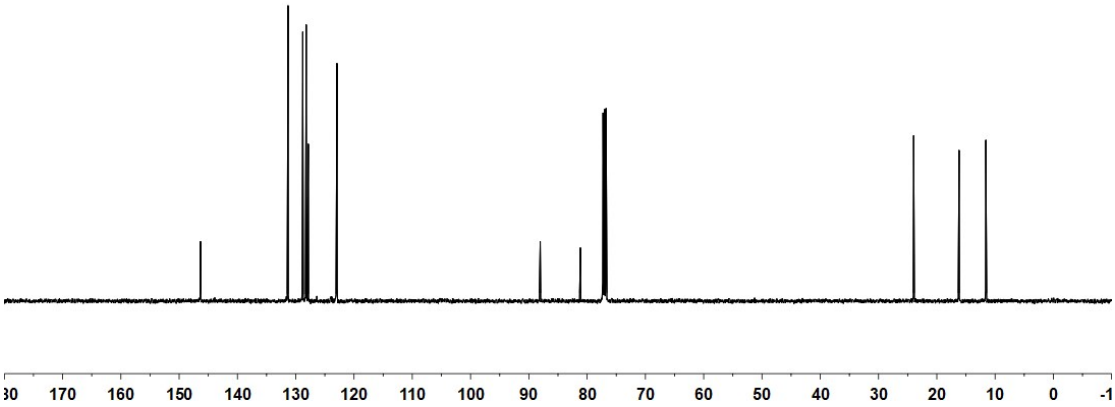
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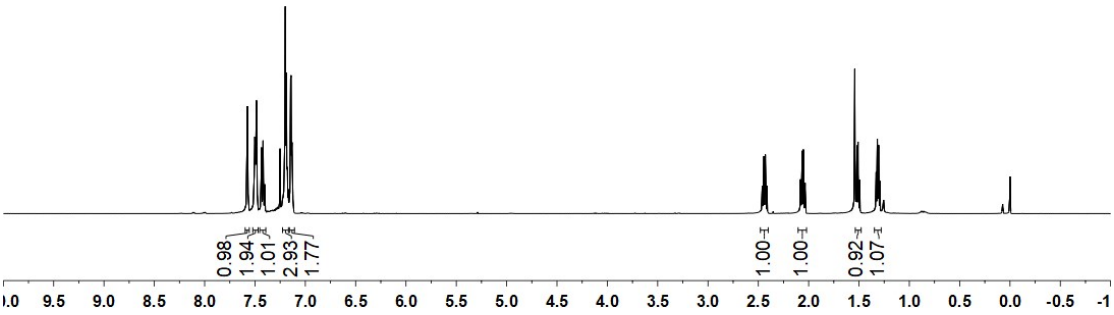
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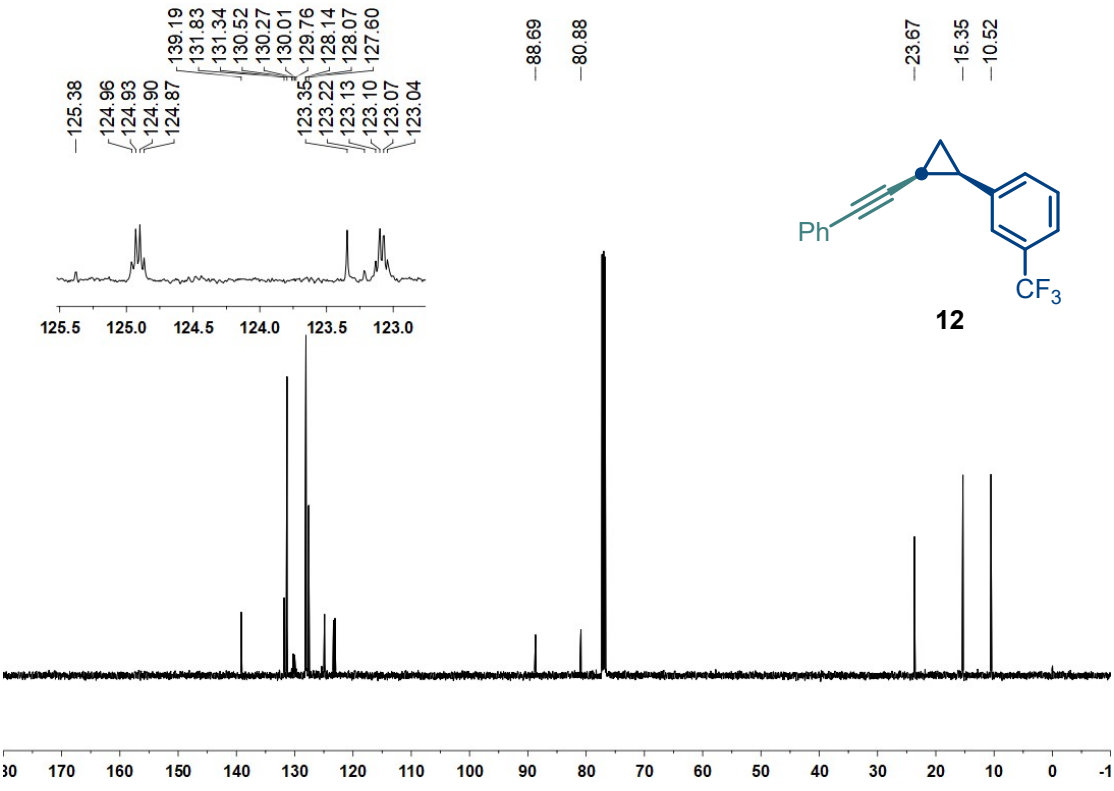
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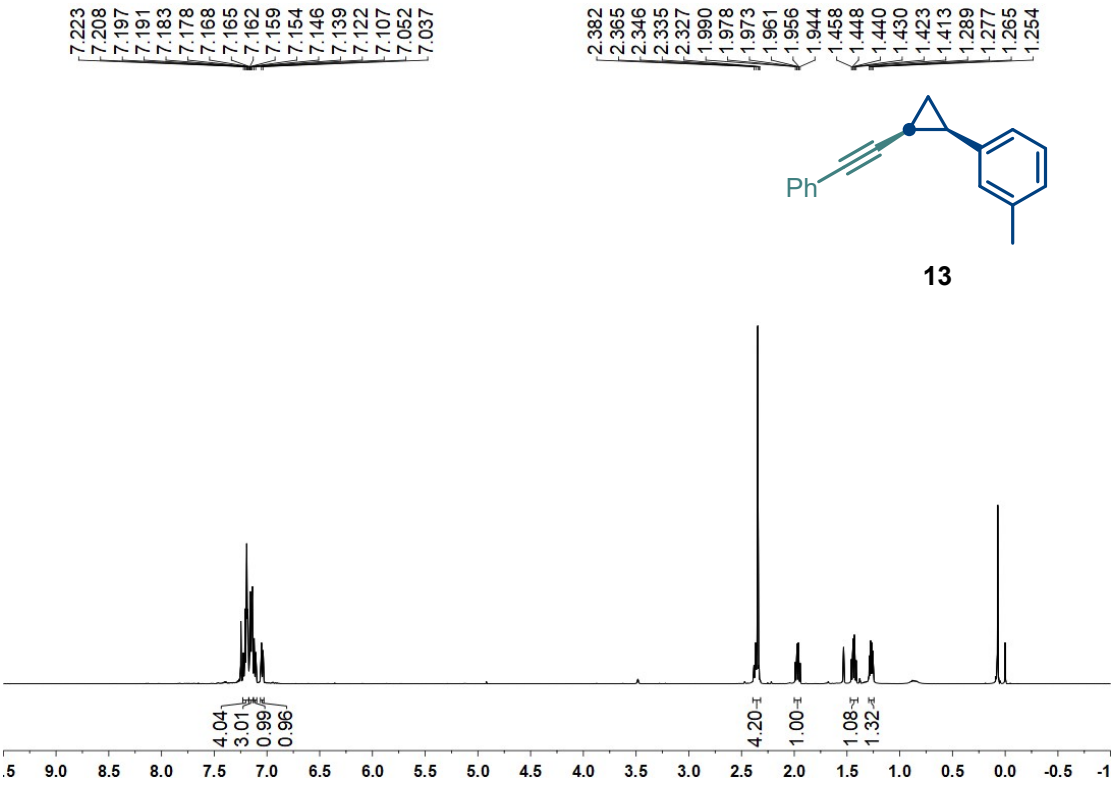
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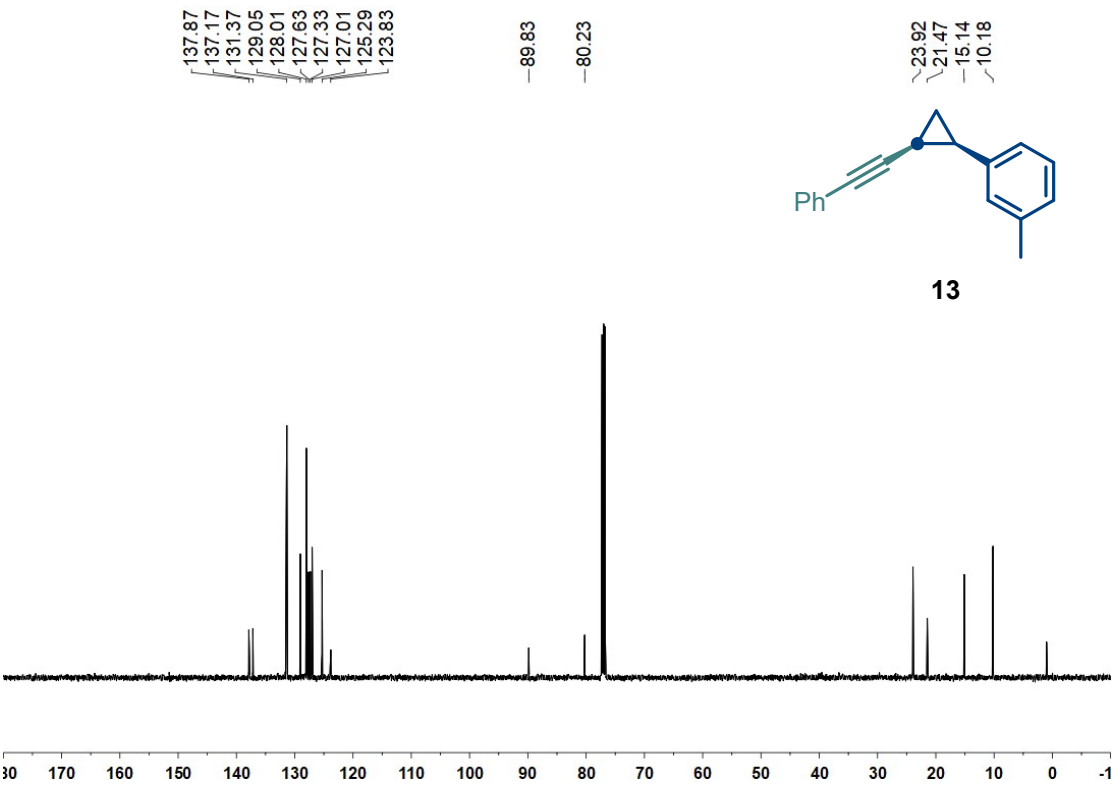
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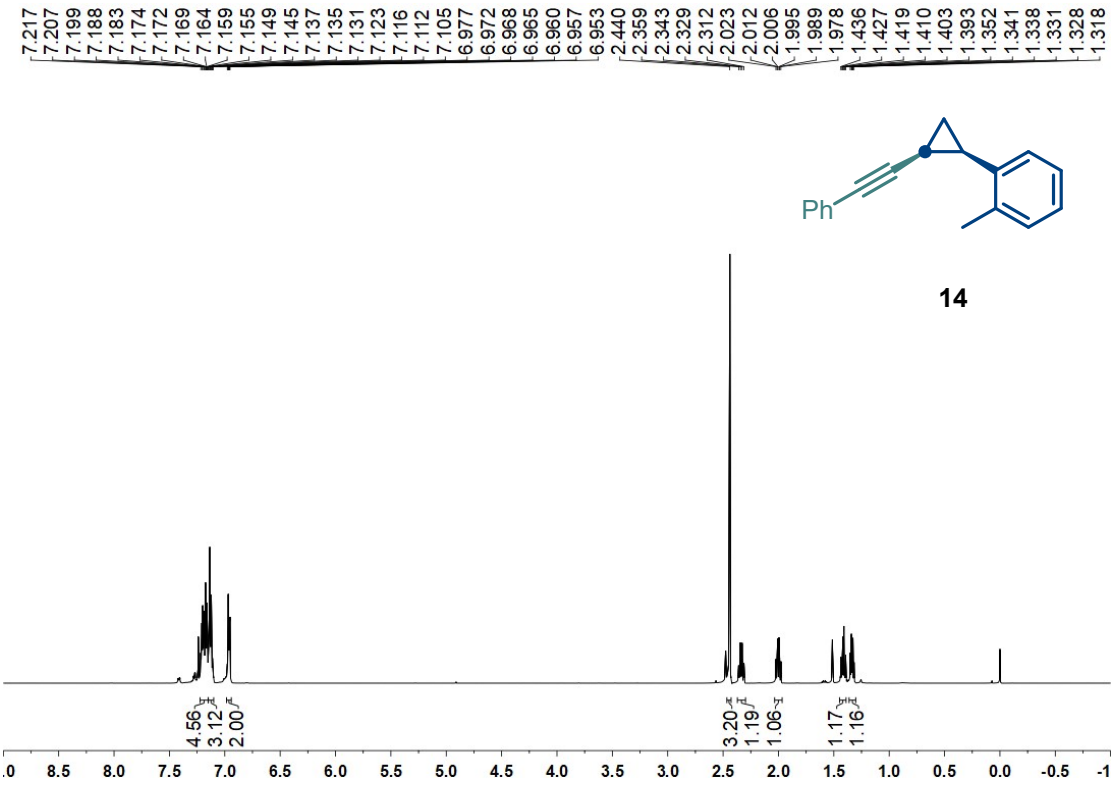
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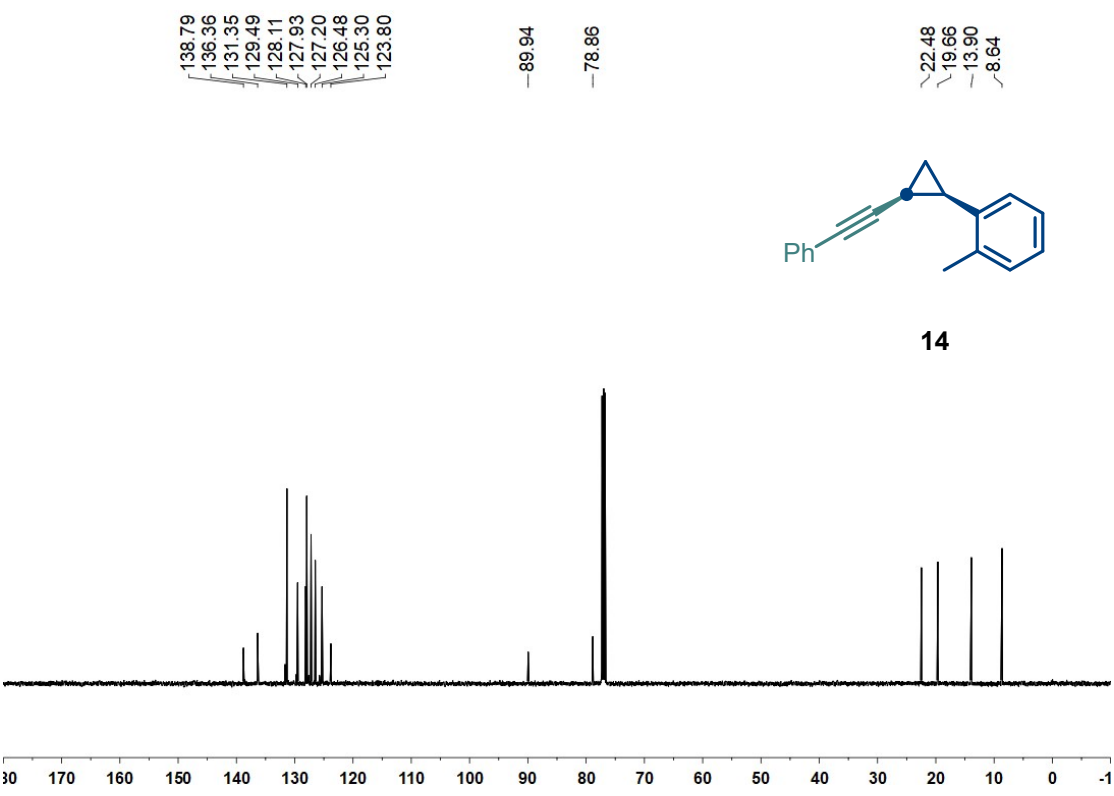
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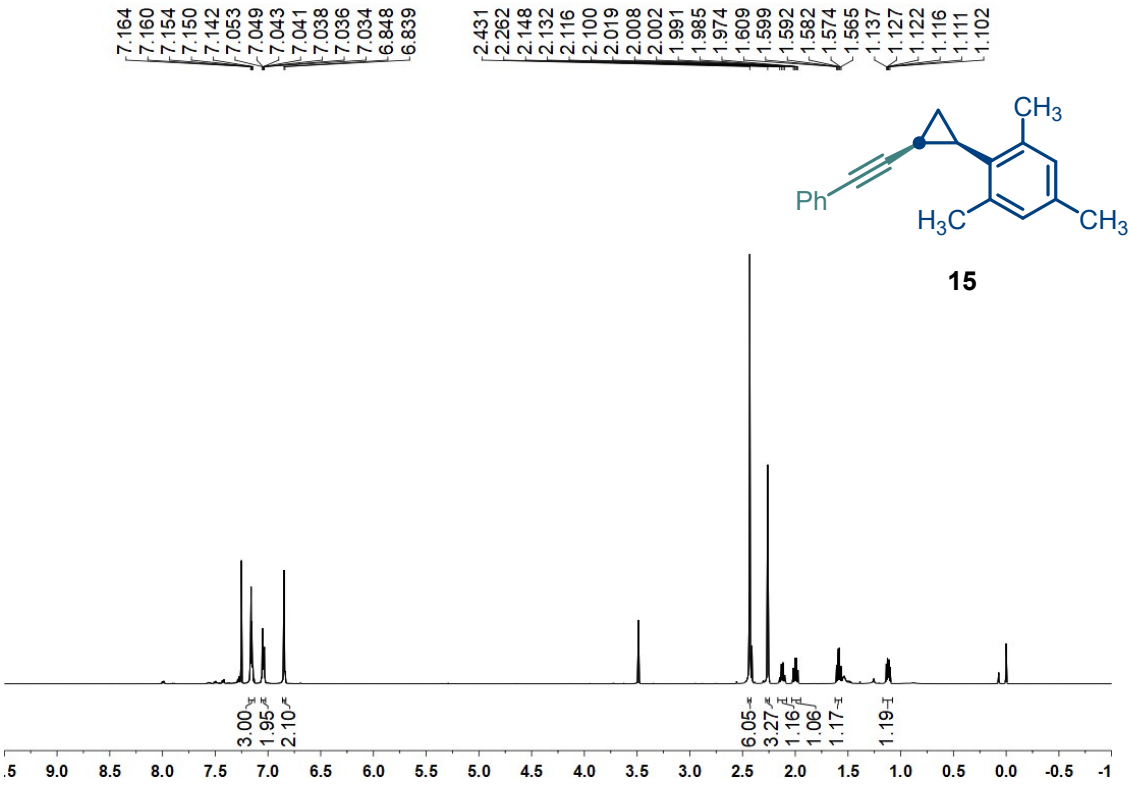
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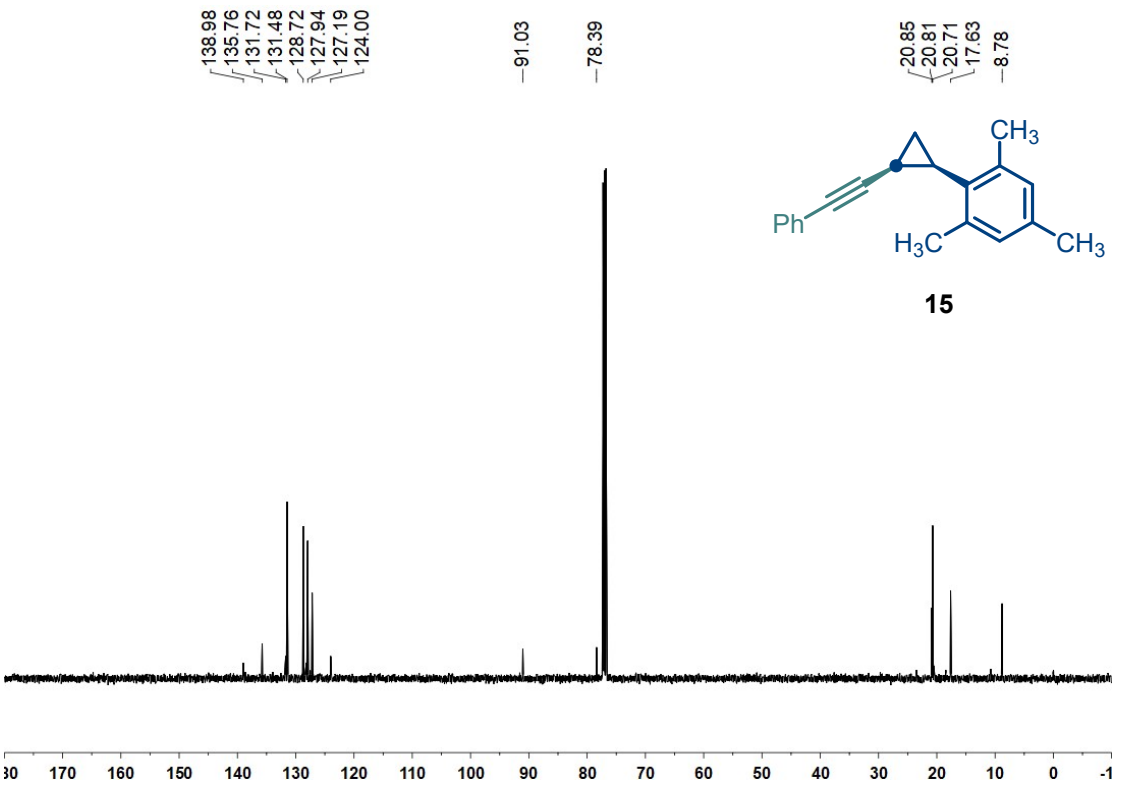
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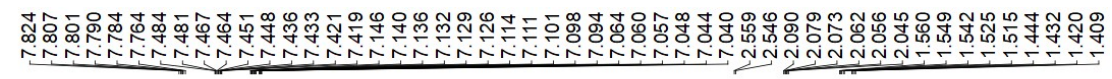
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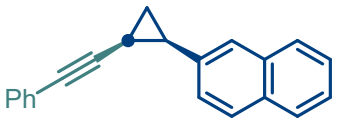
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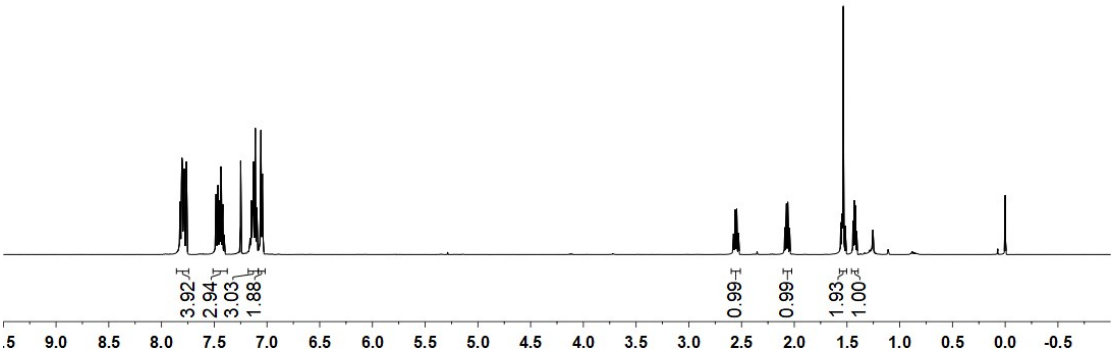
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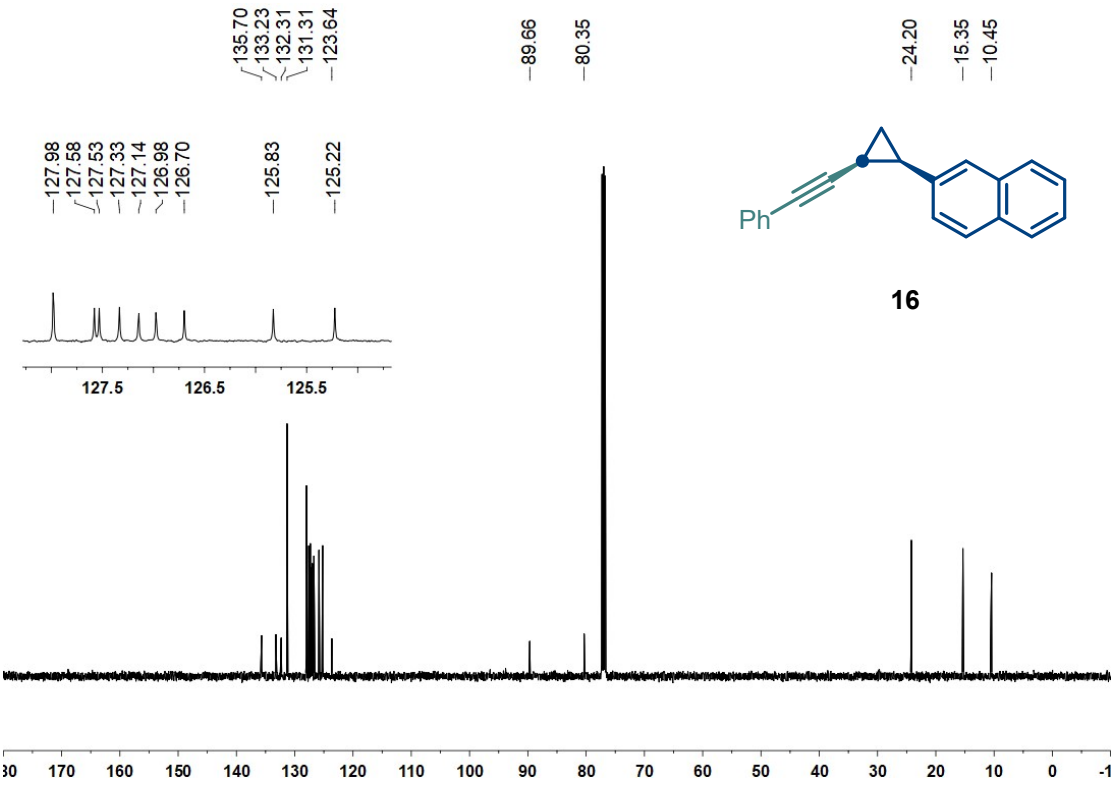
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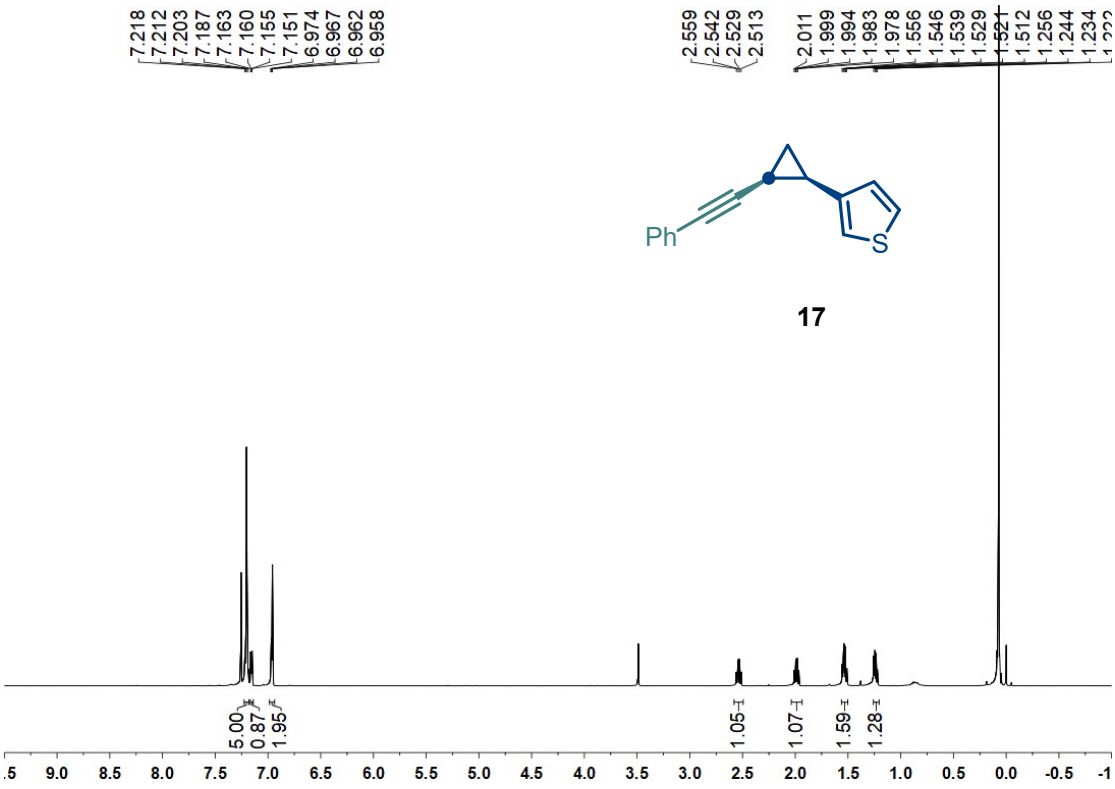
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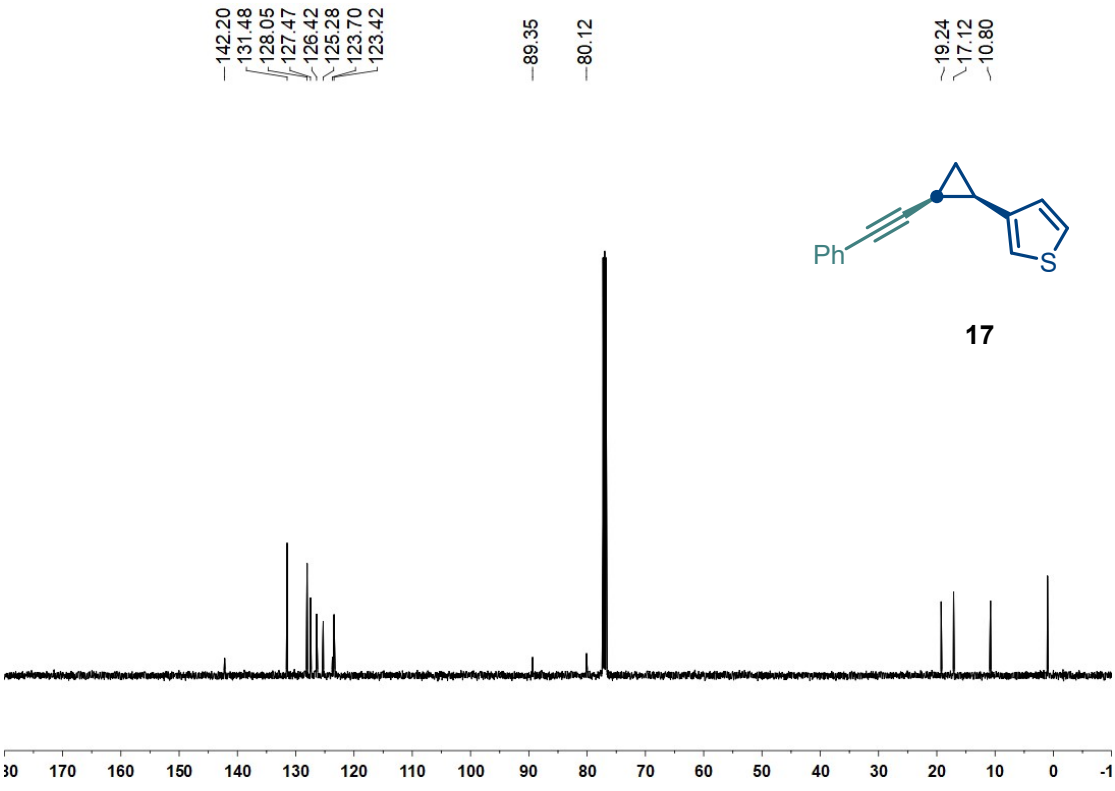
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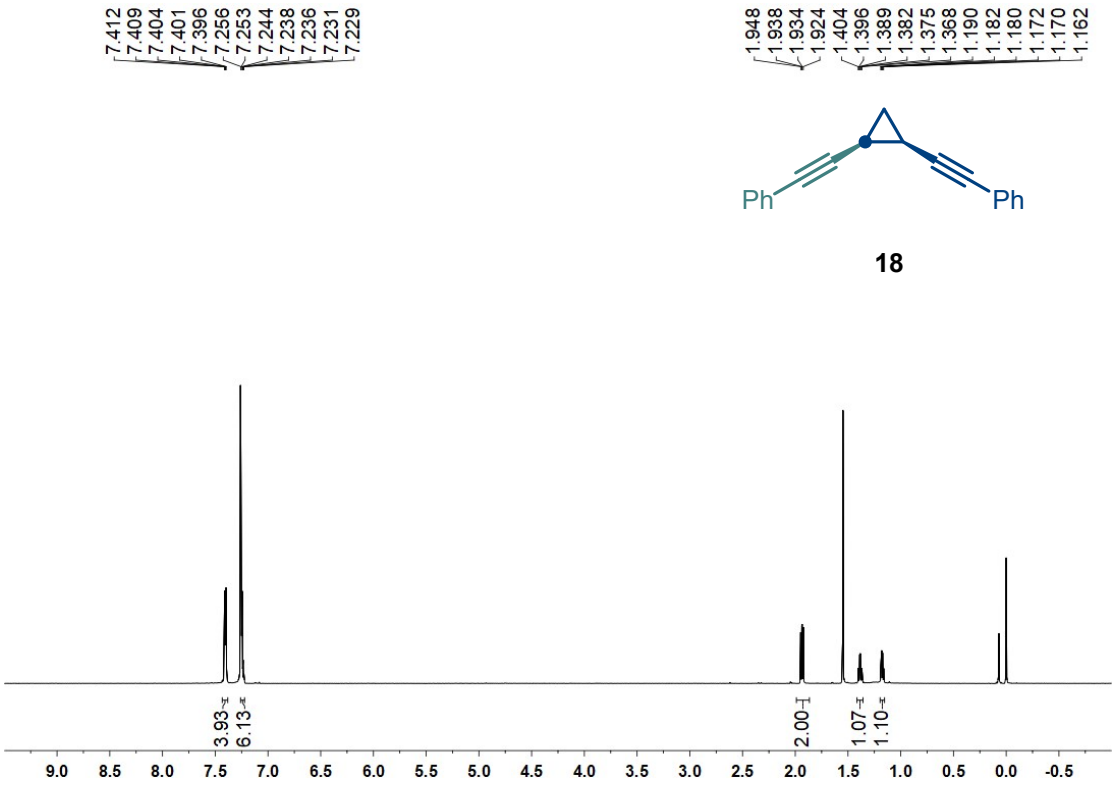
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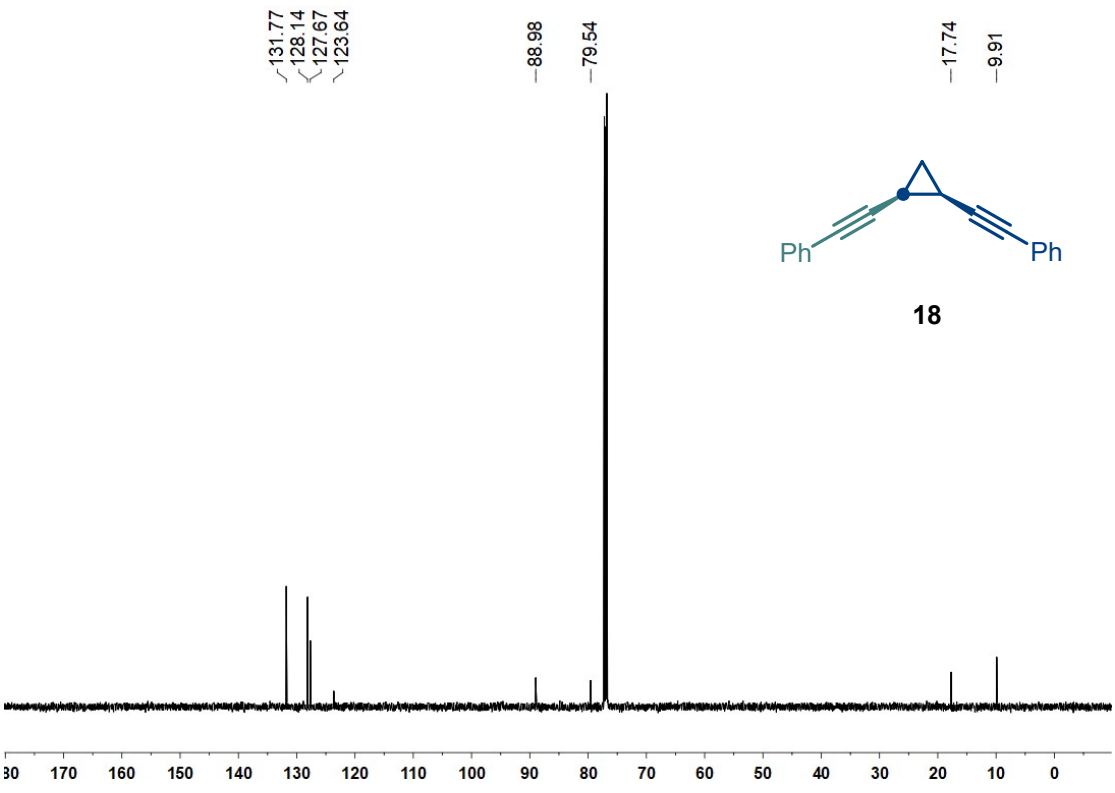
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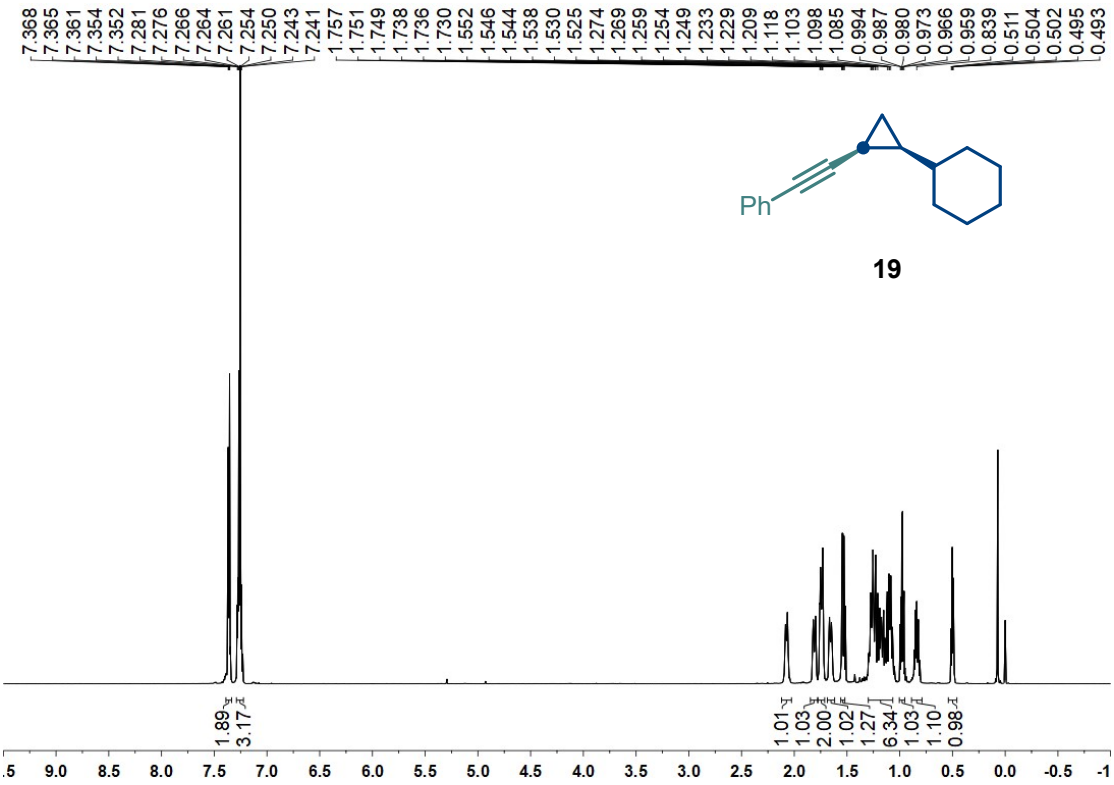
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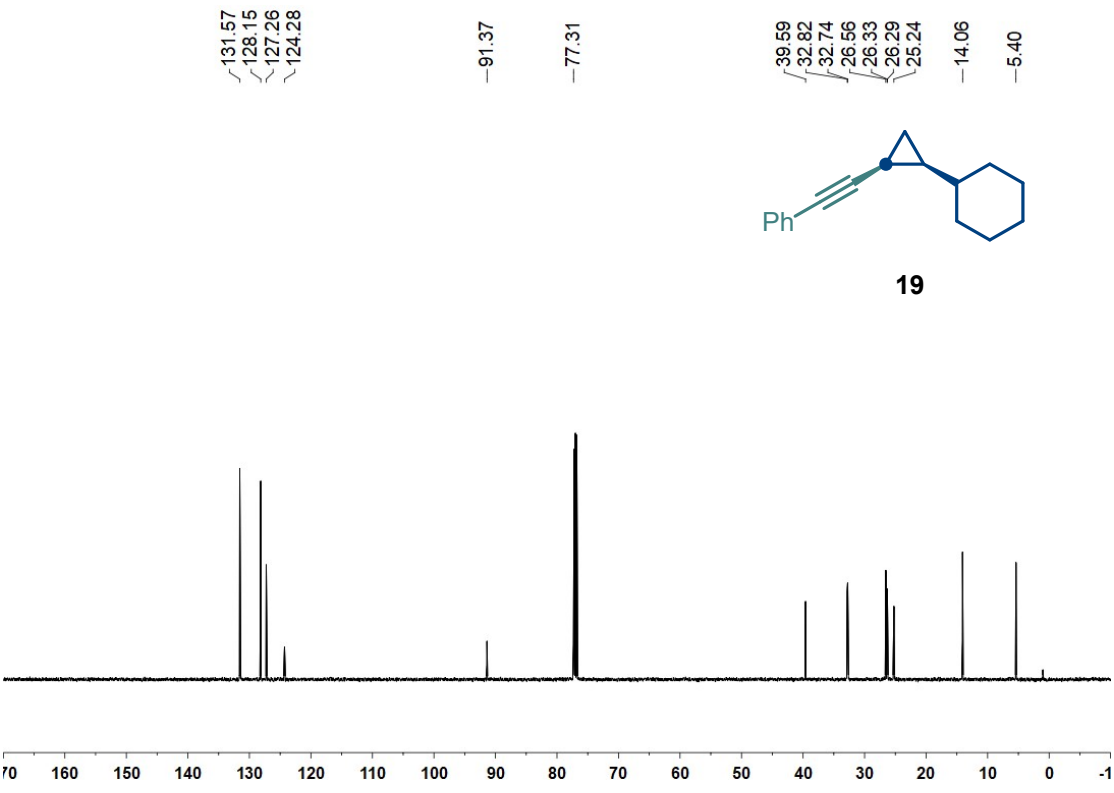
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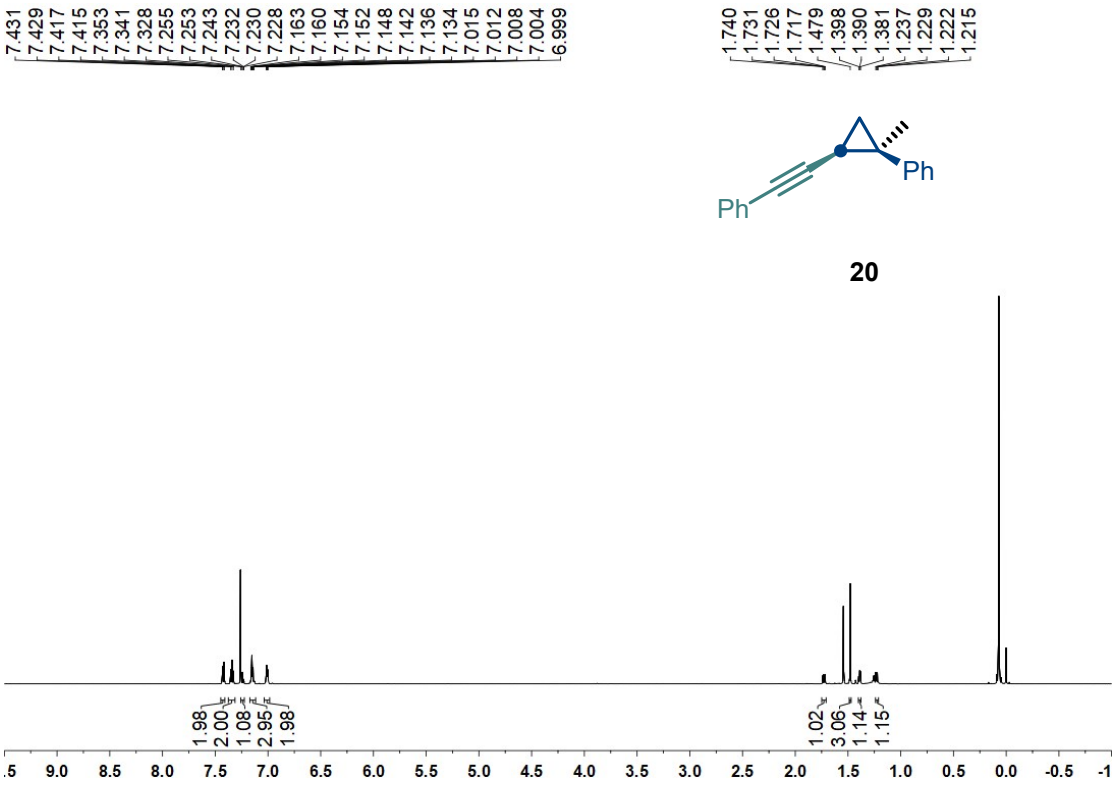
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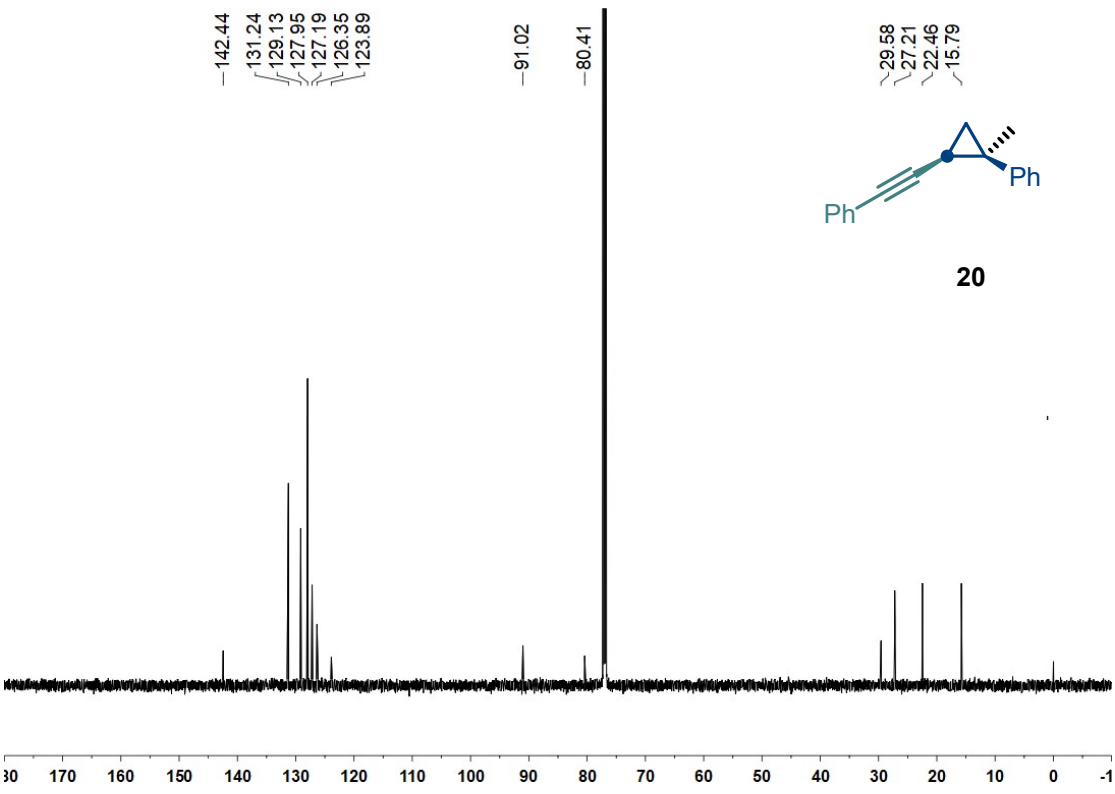
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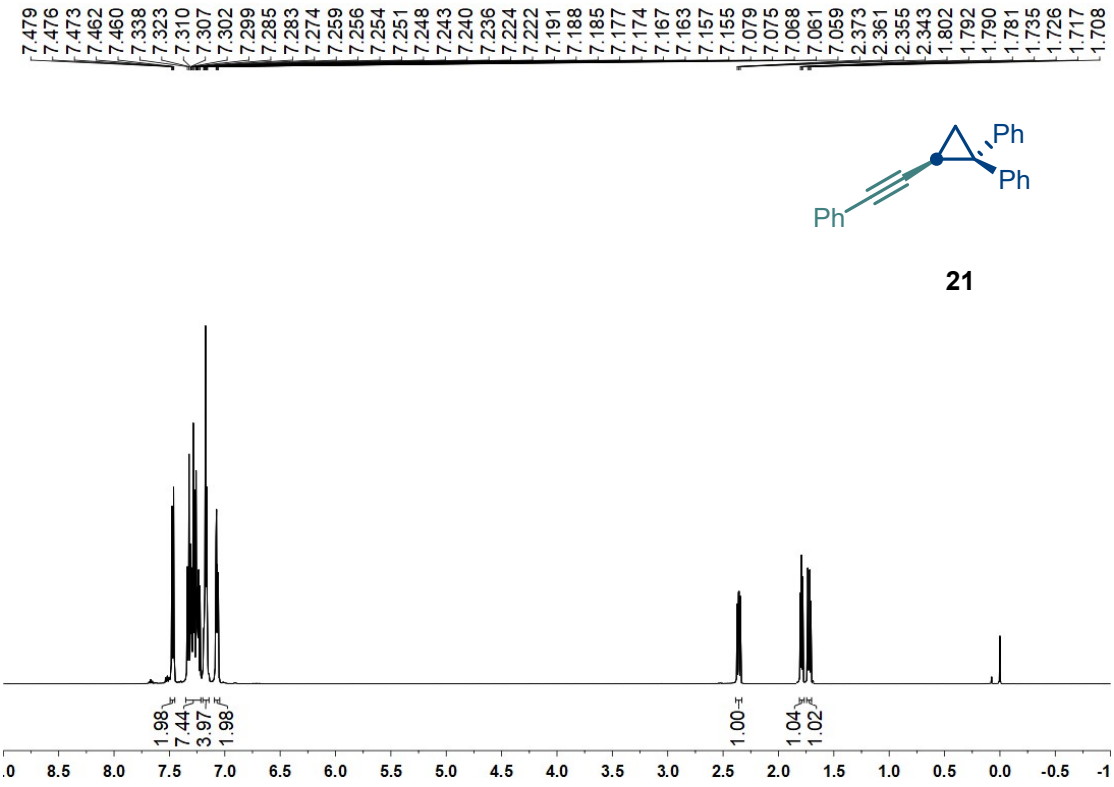
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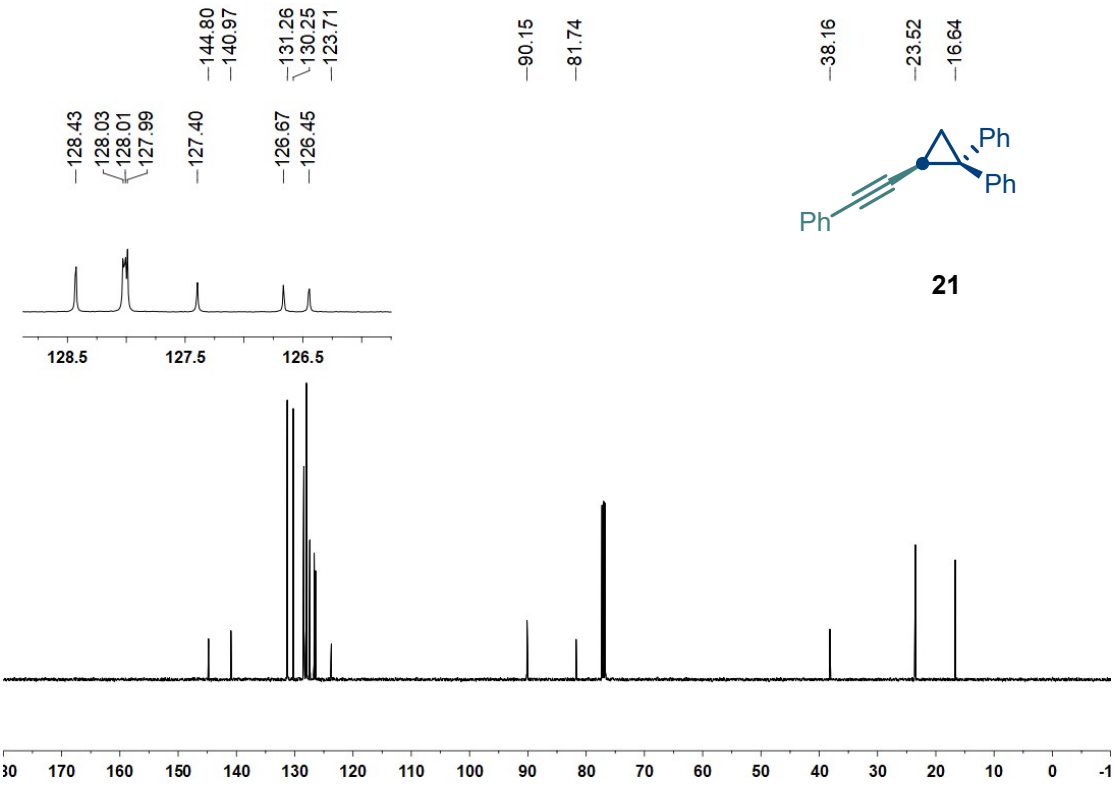
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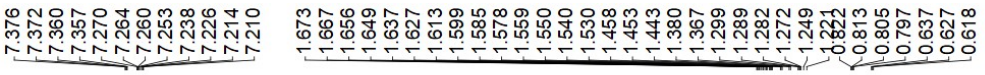
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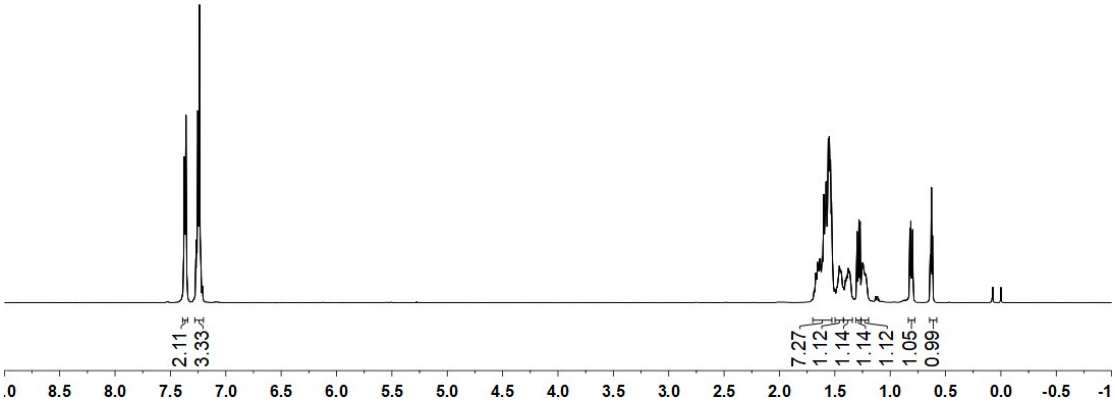
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Ph

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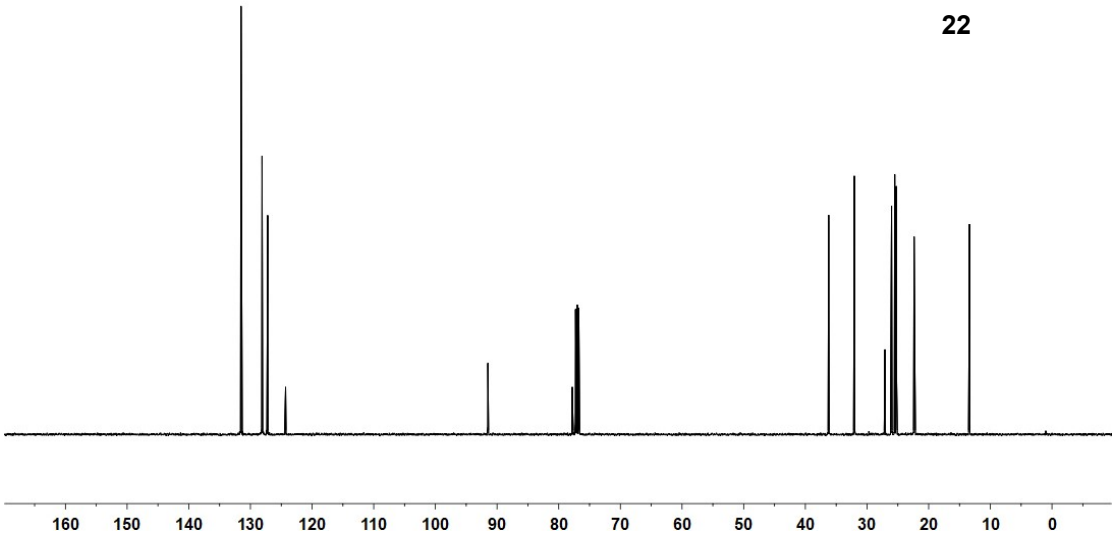
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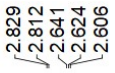
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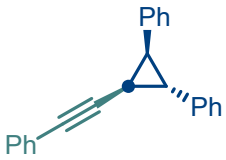
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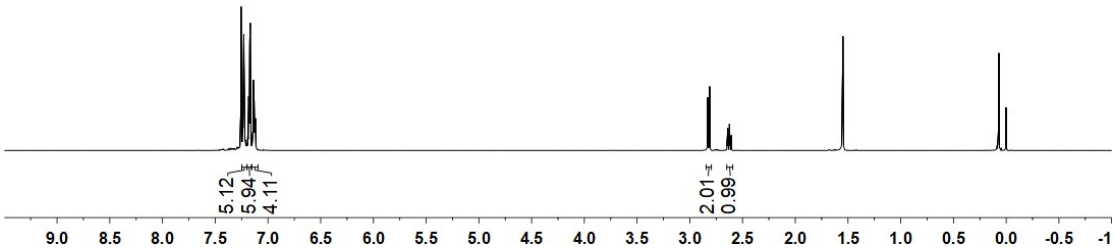
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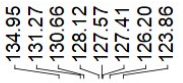
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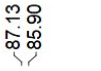
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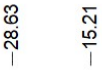
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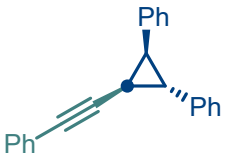
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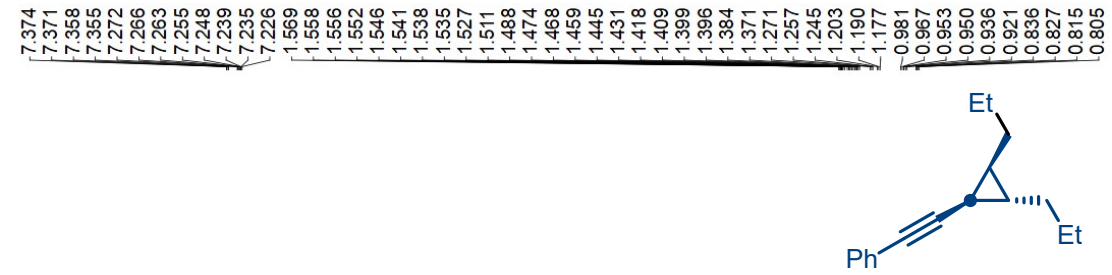
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Et
Et
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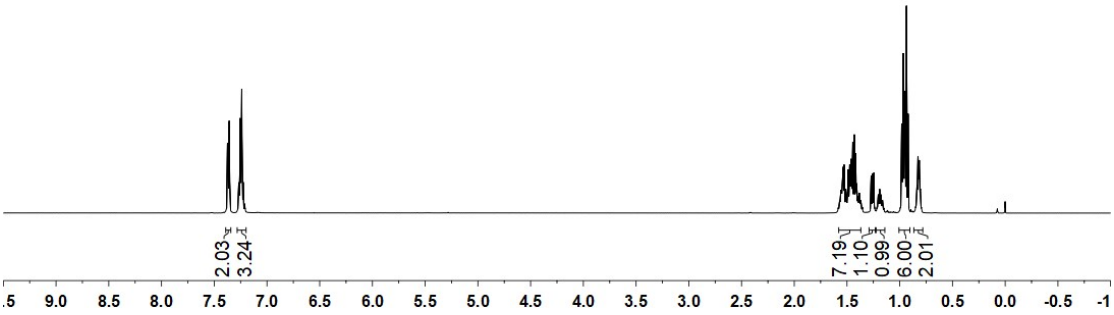
24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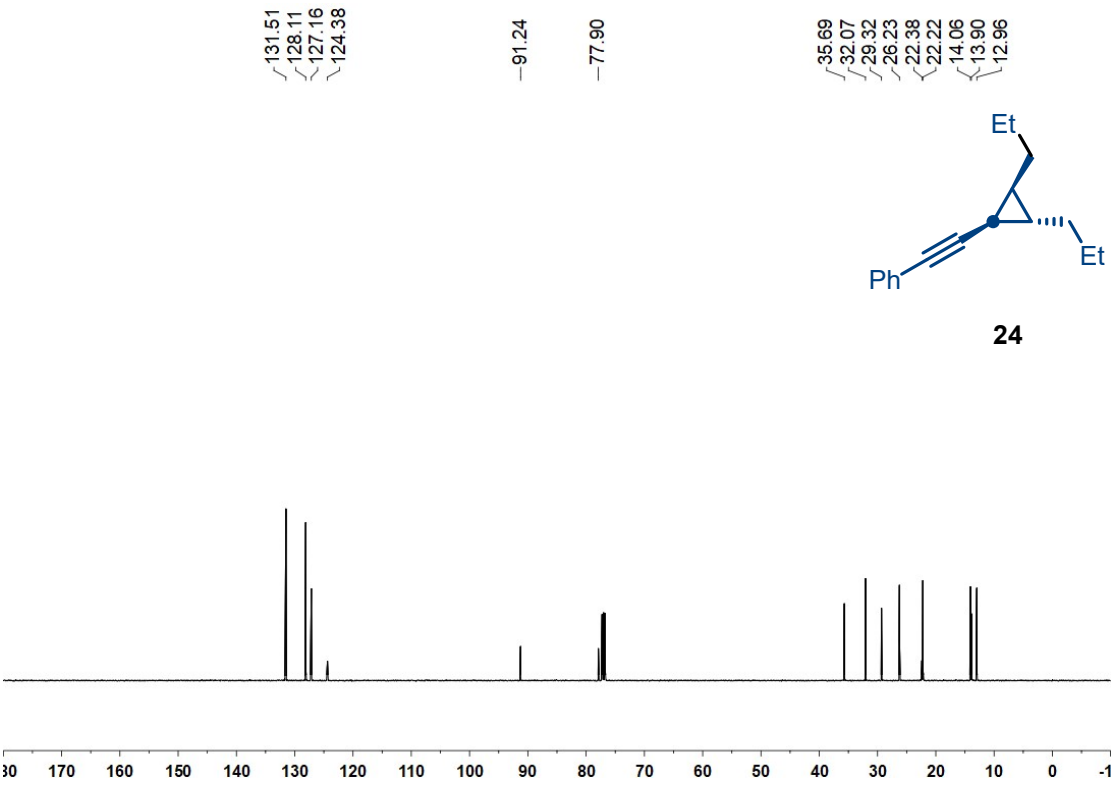
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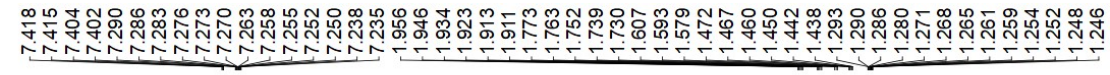
Et
Et
Ph

24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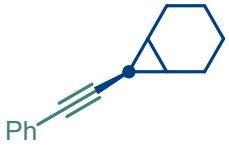
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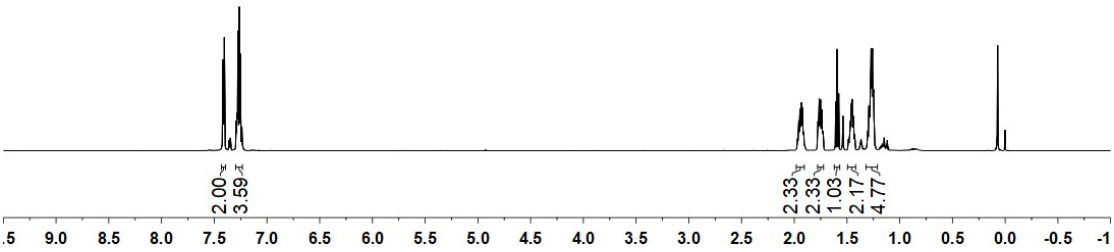
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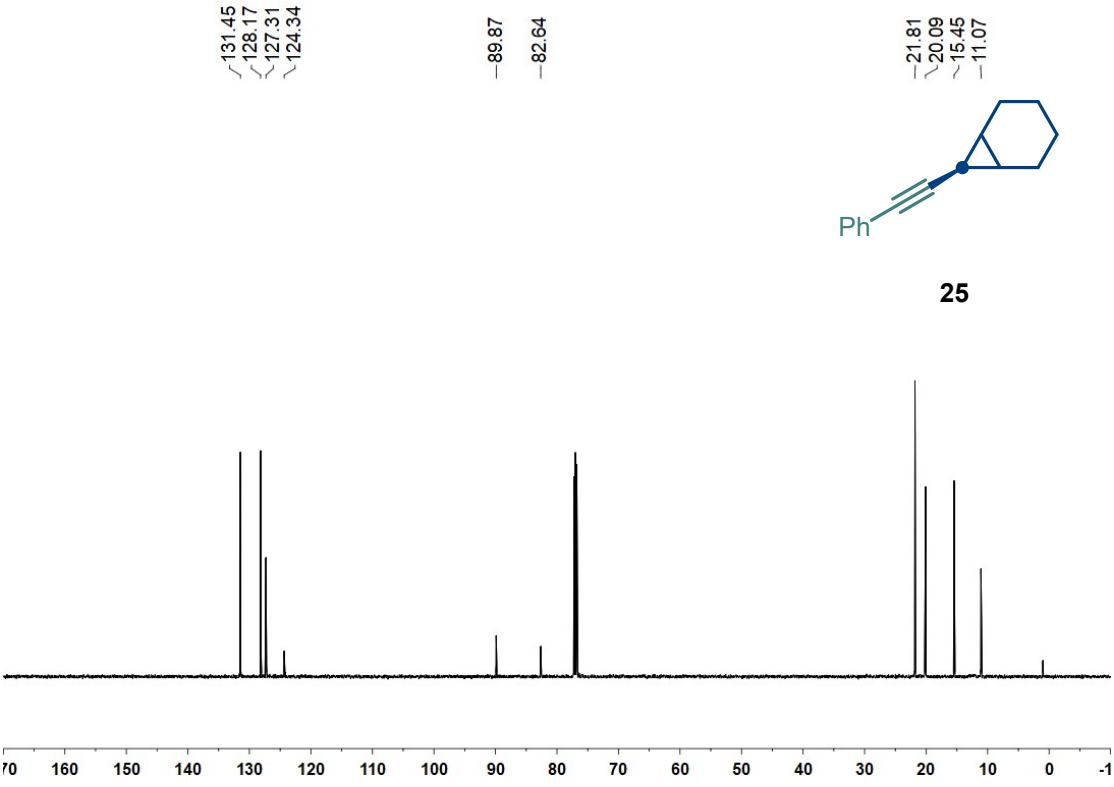
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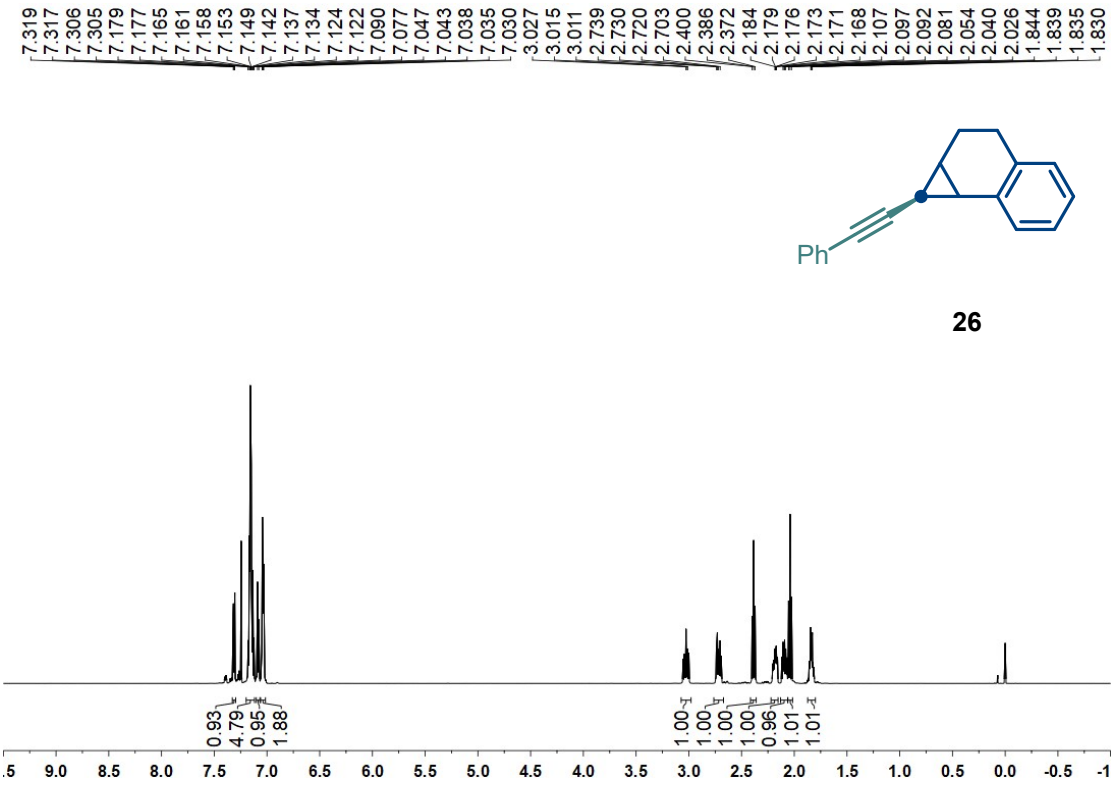
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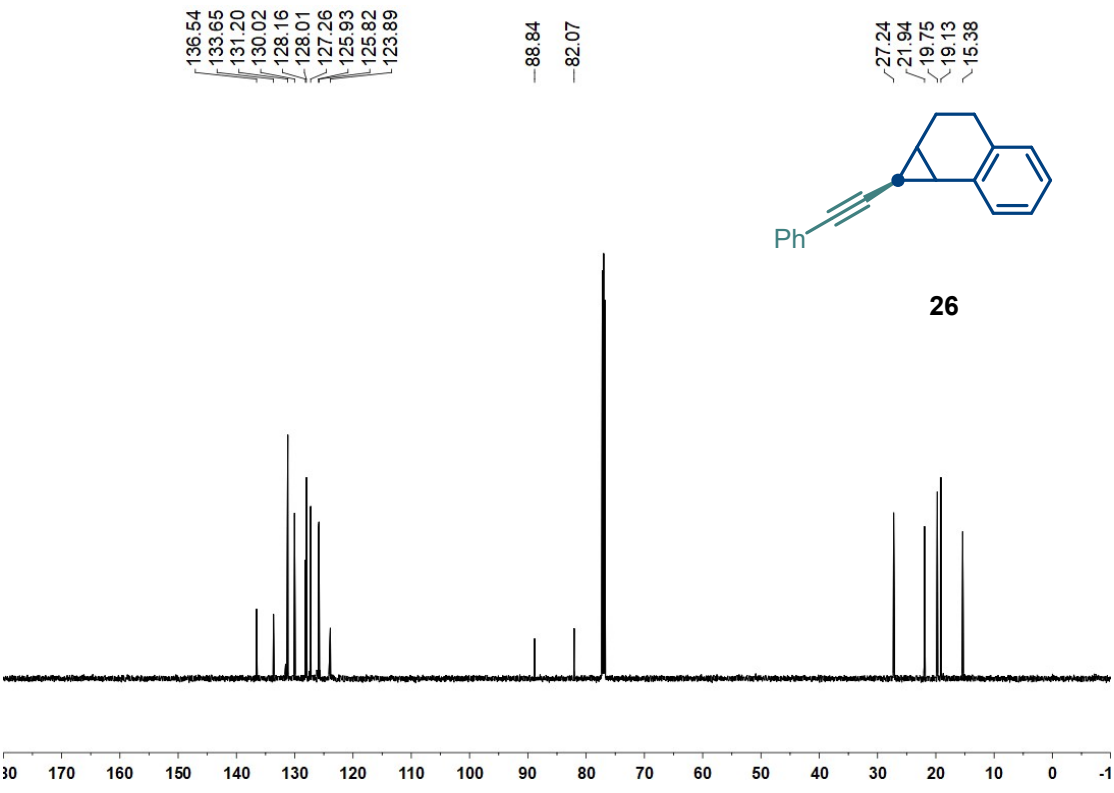
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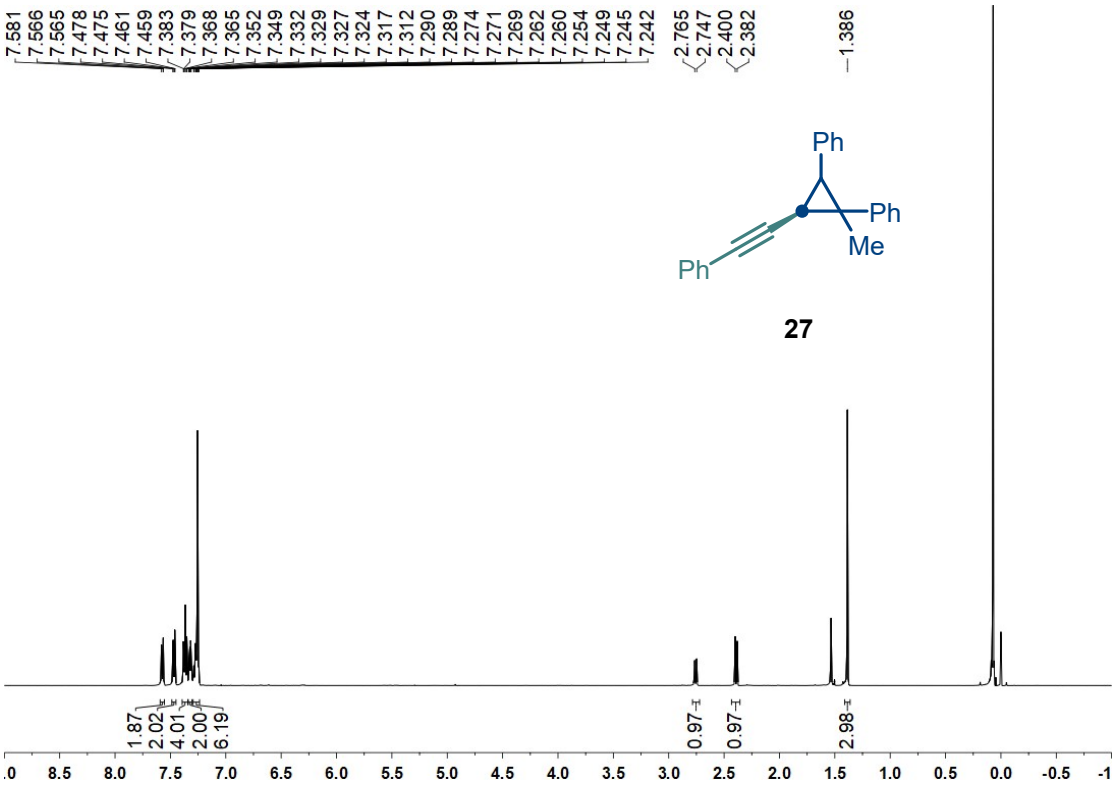
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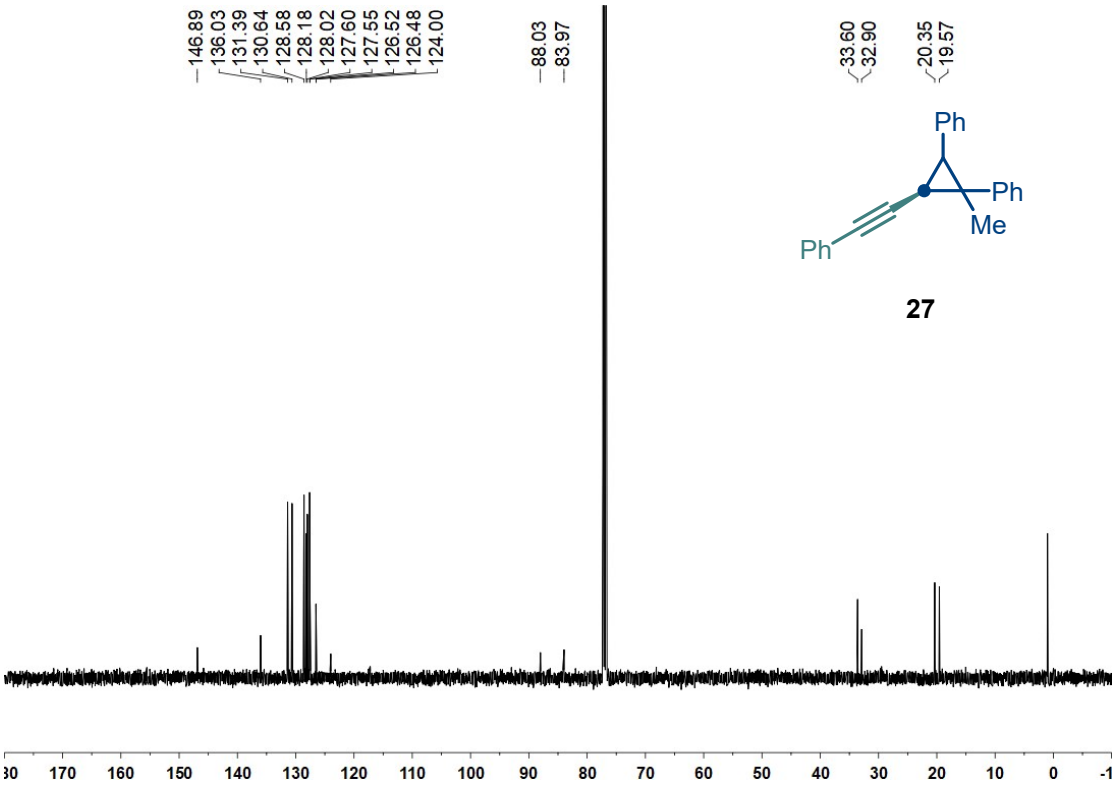
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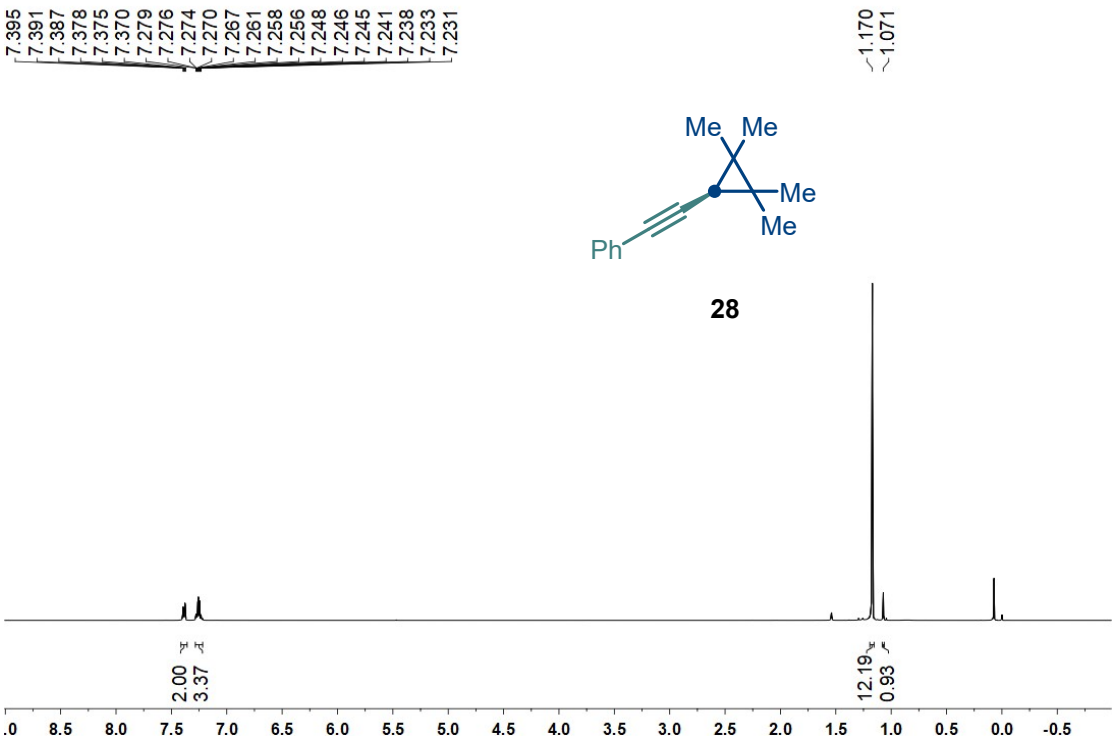
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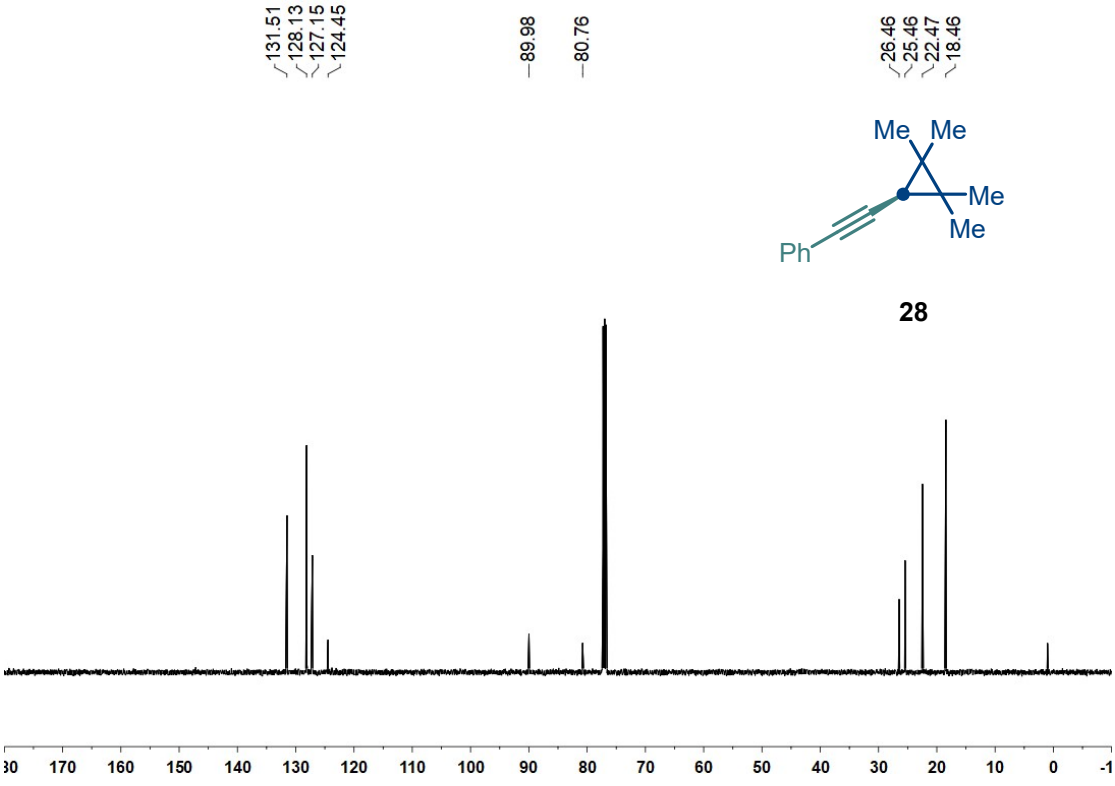
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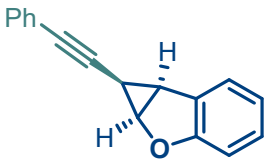
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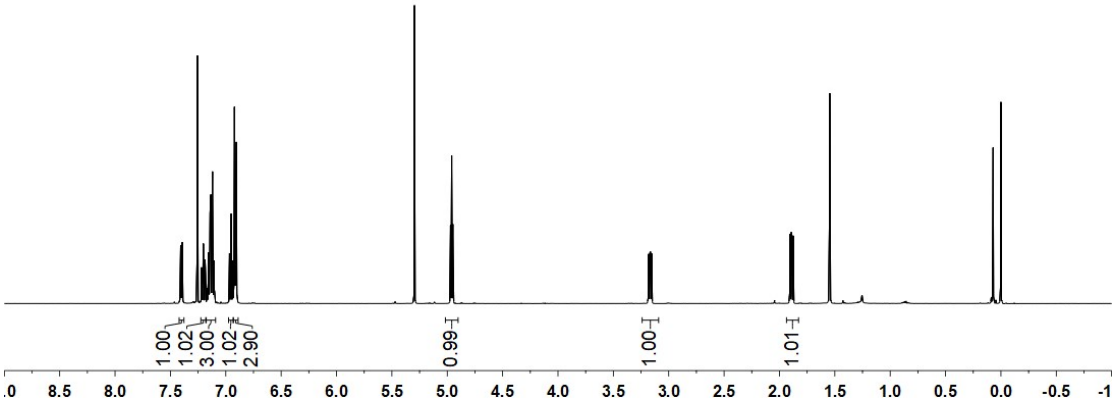
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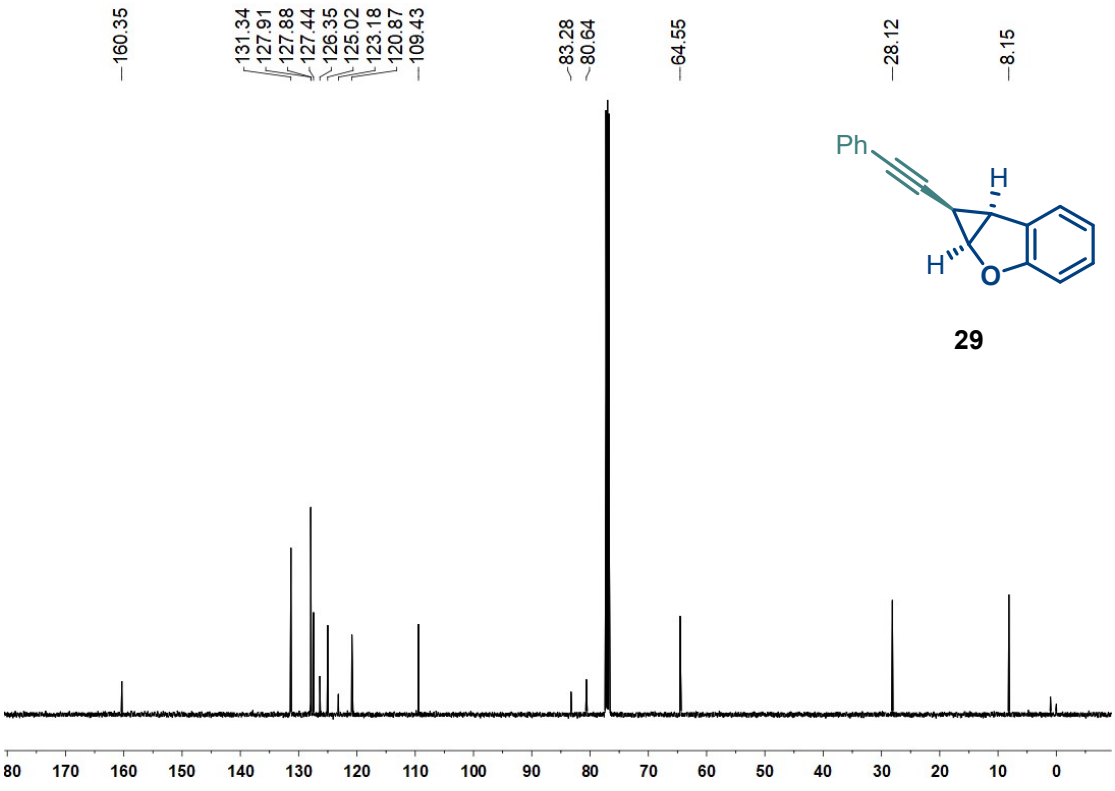
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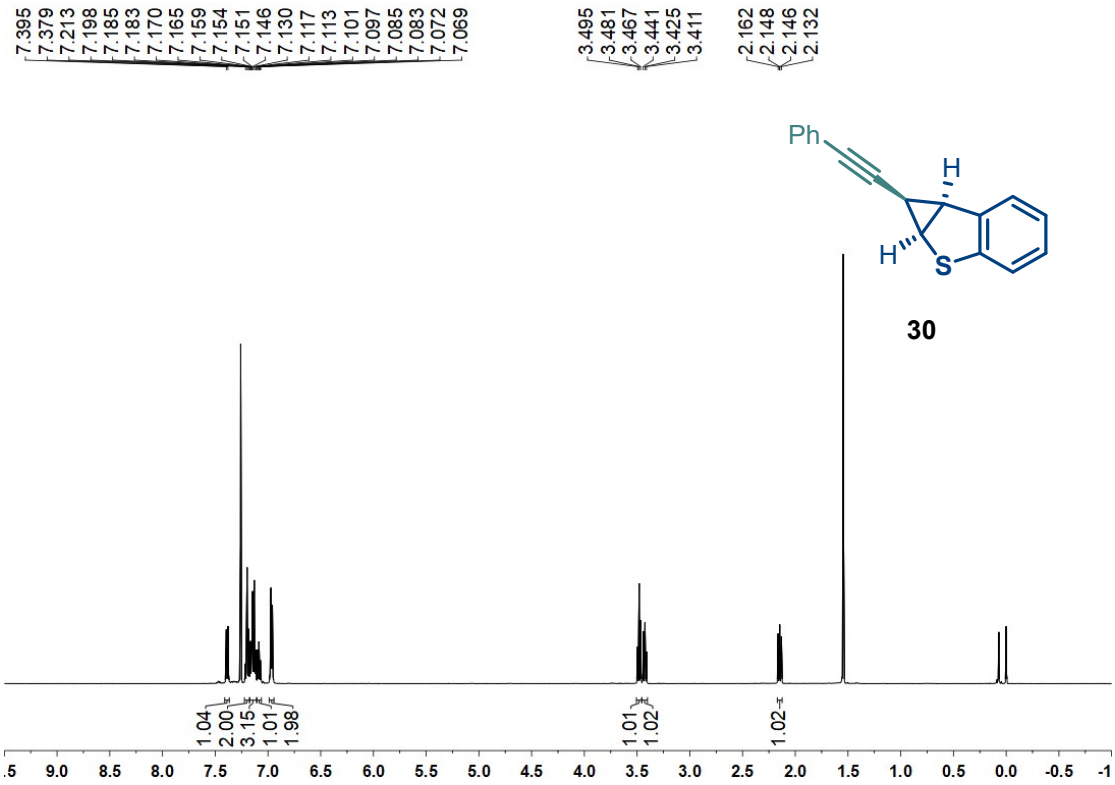
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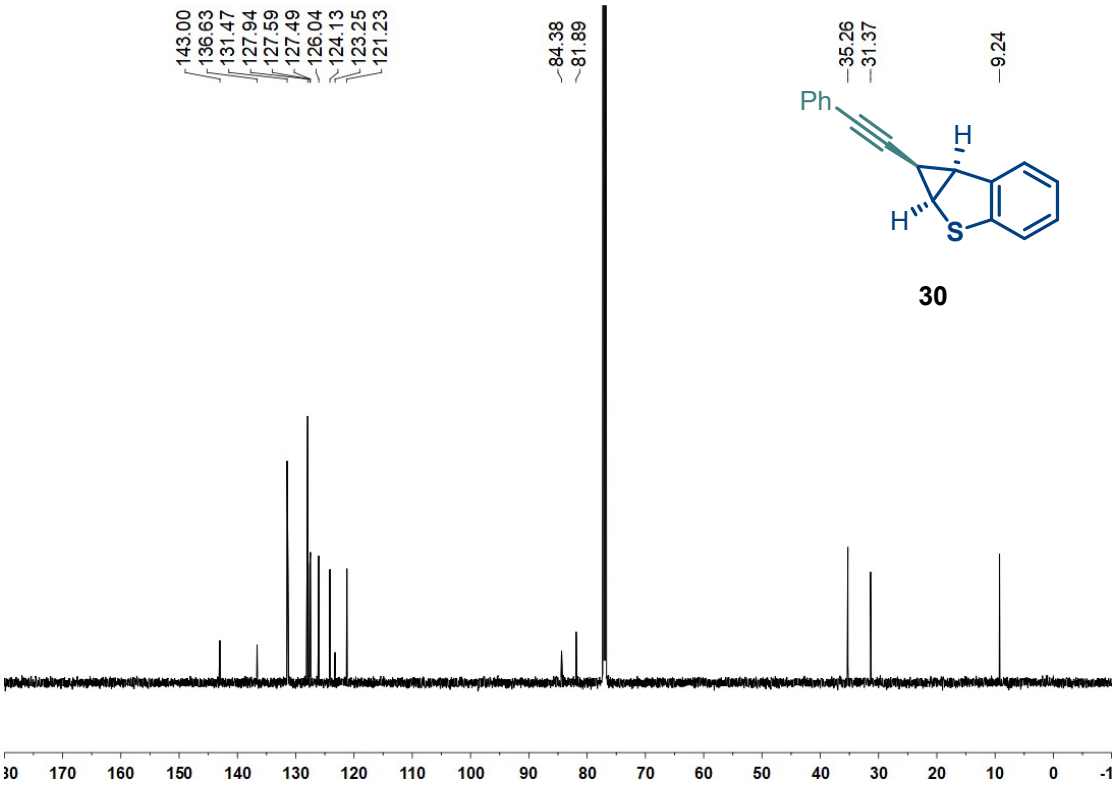
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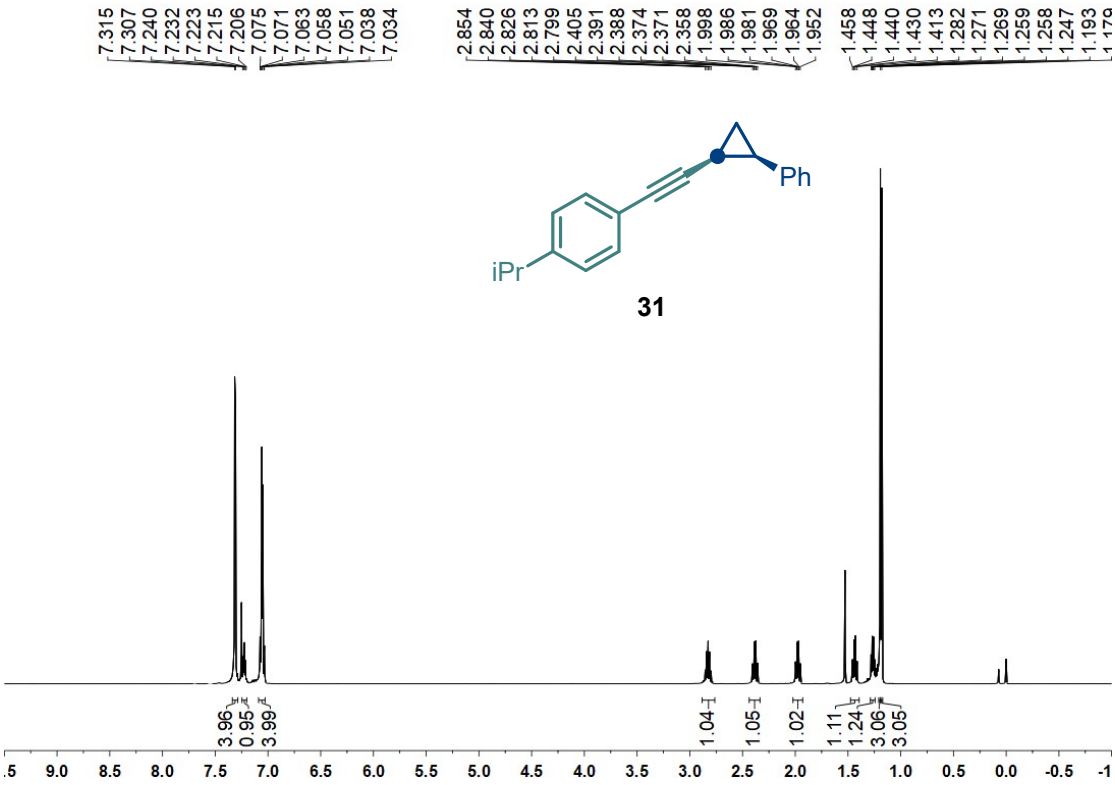
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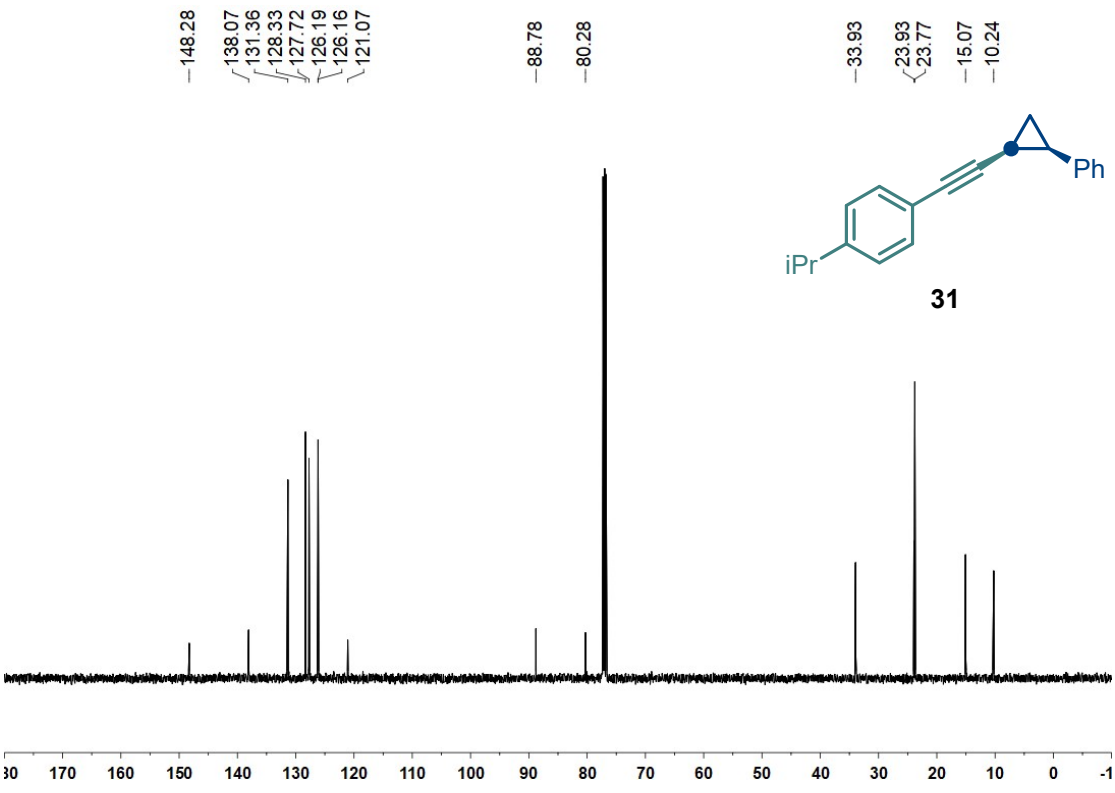
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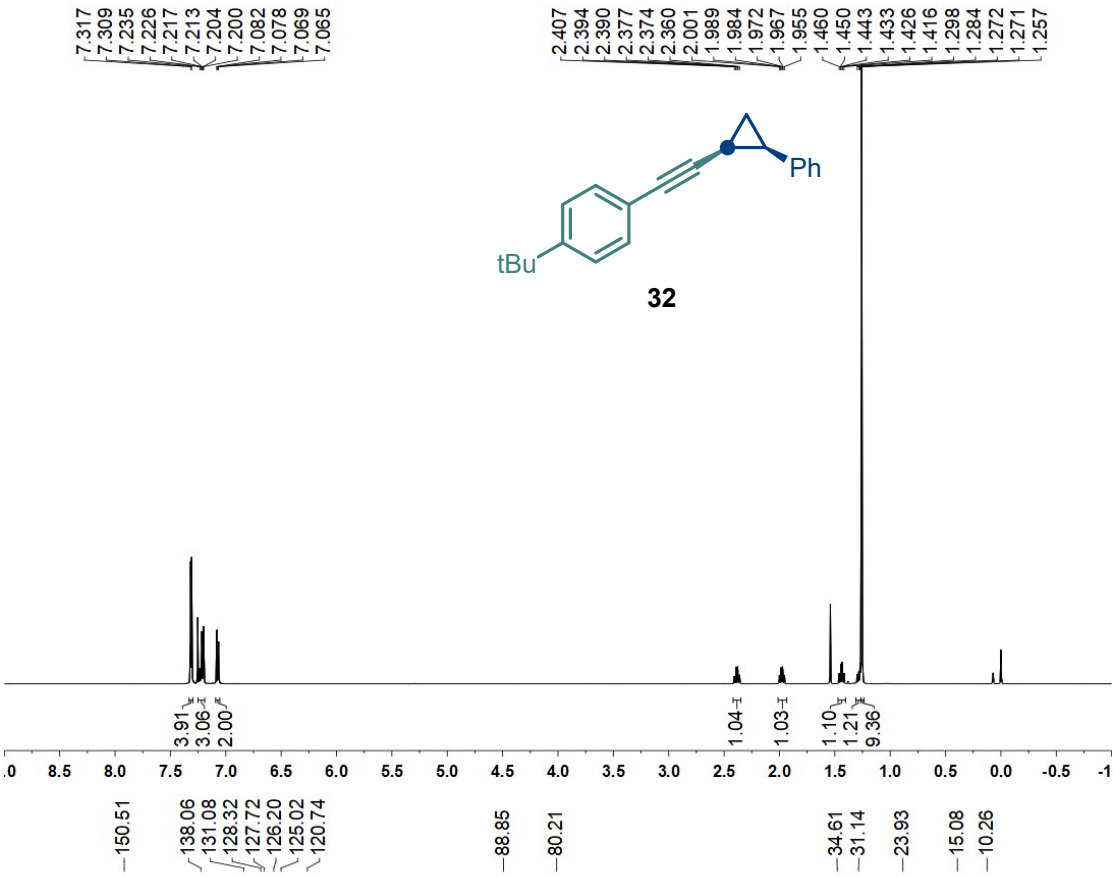
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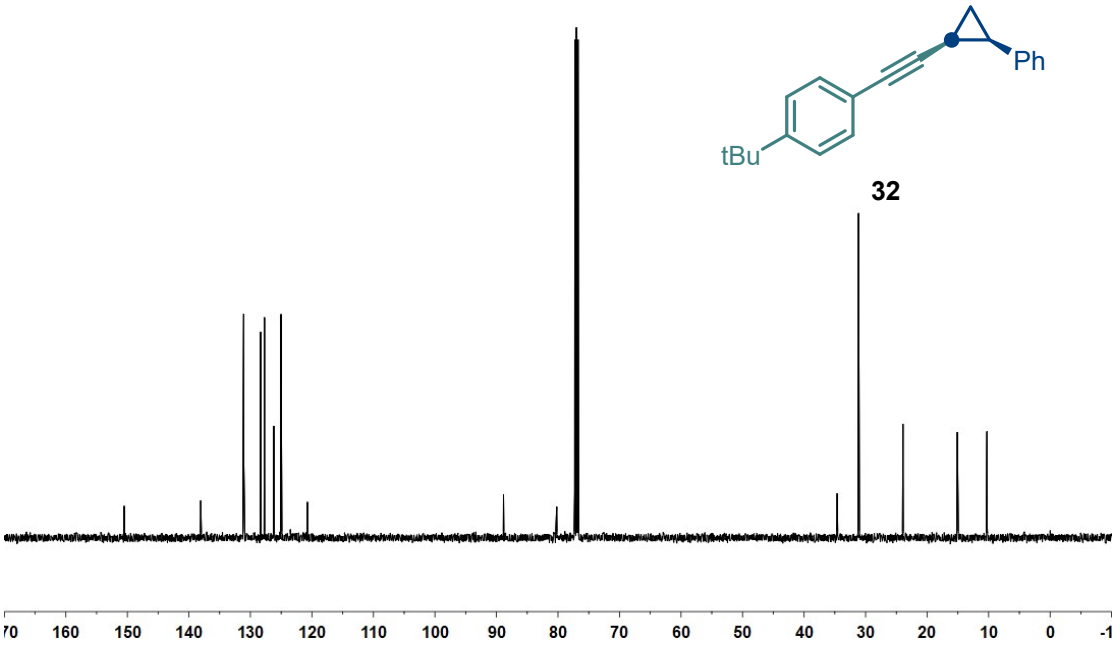
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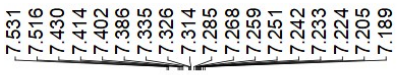
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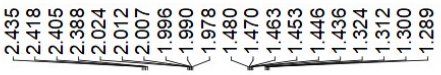
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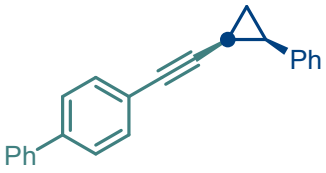
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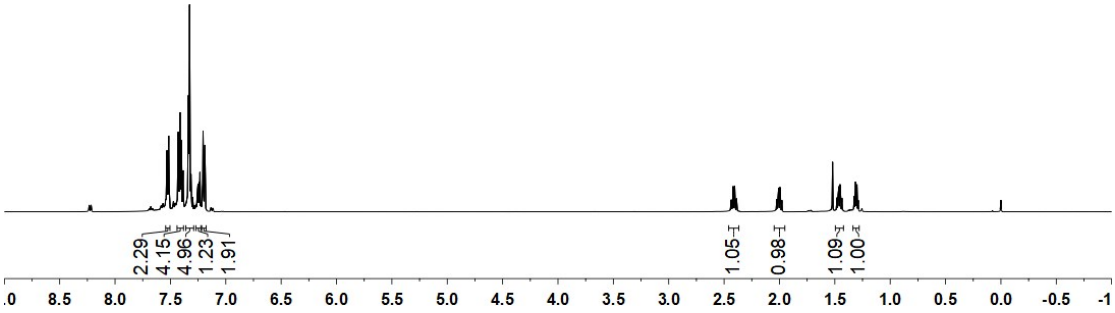
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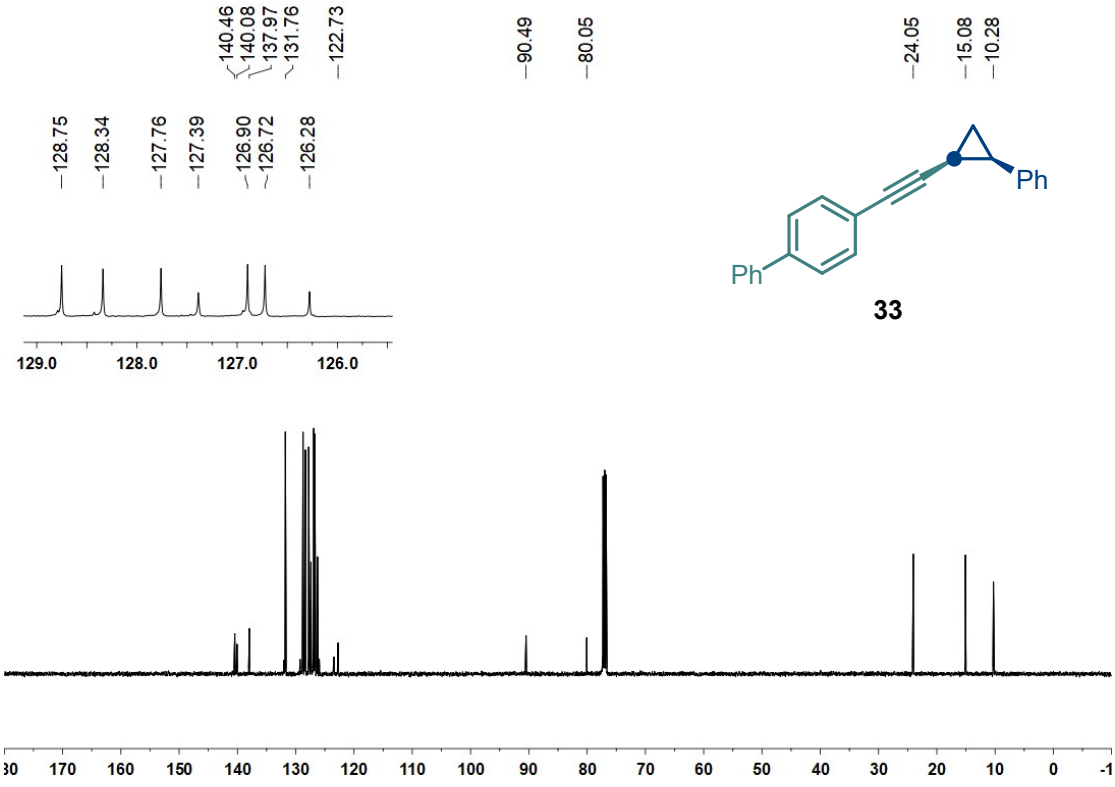
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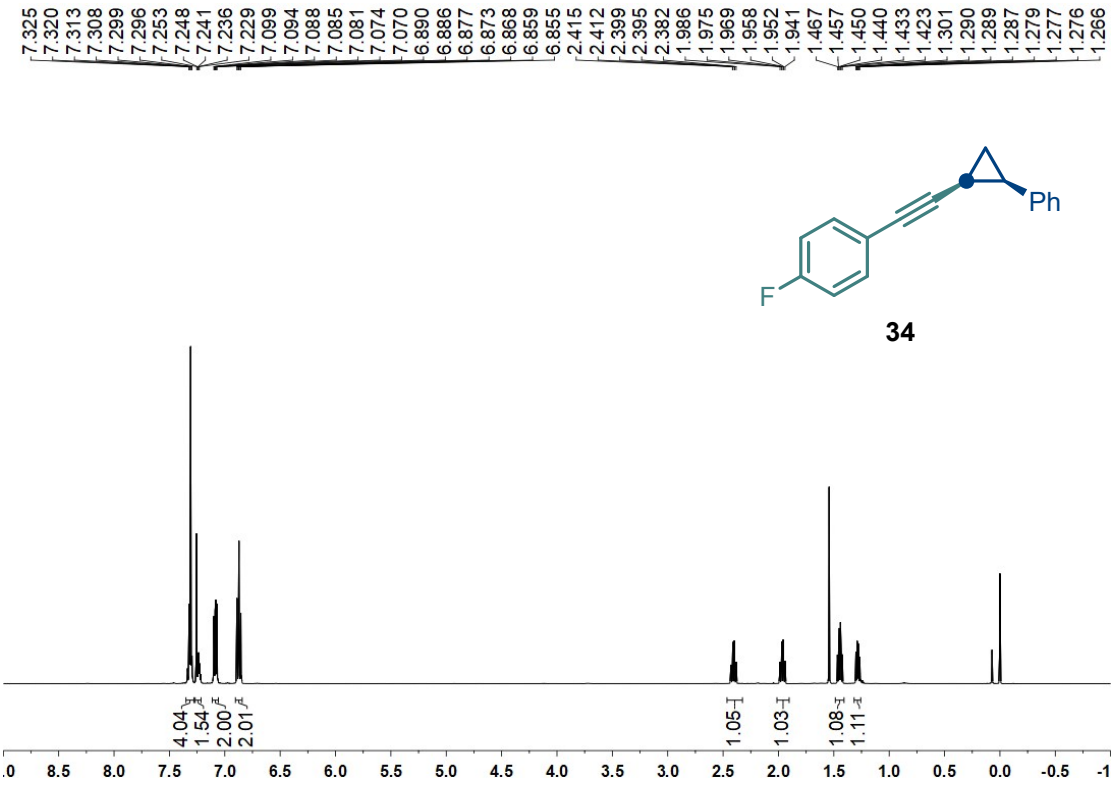
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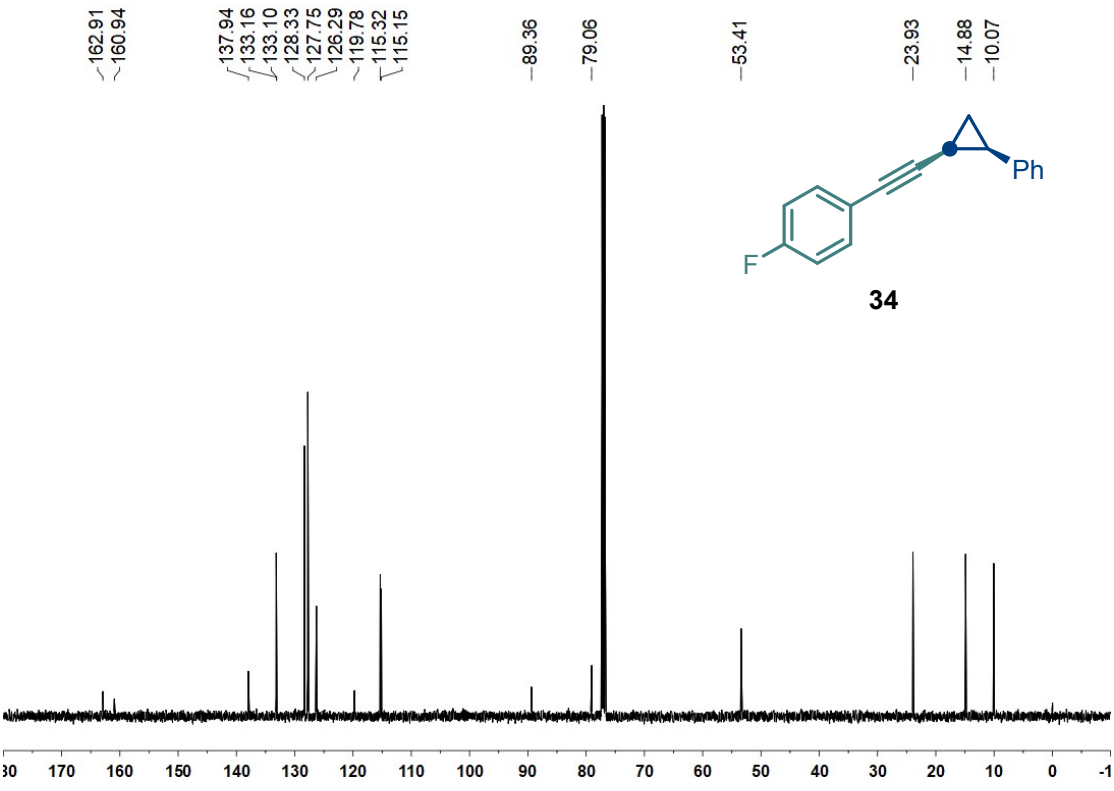
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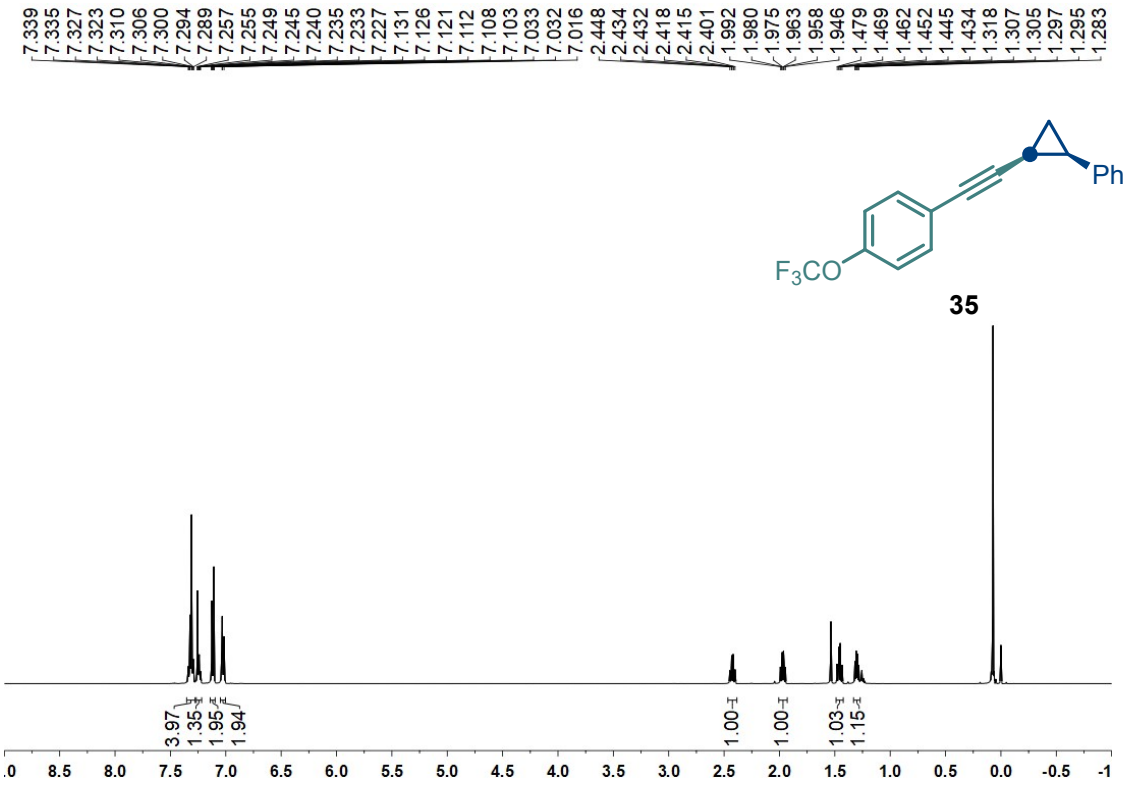
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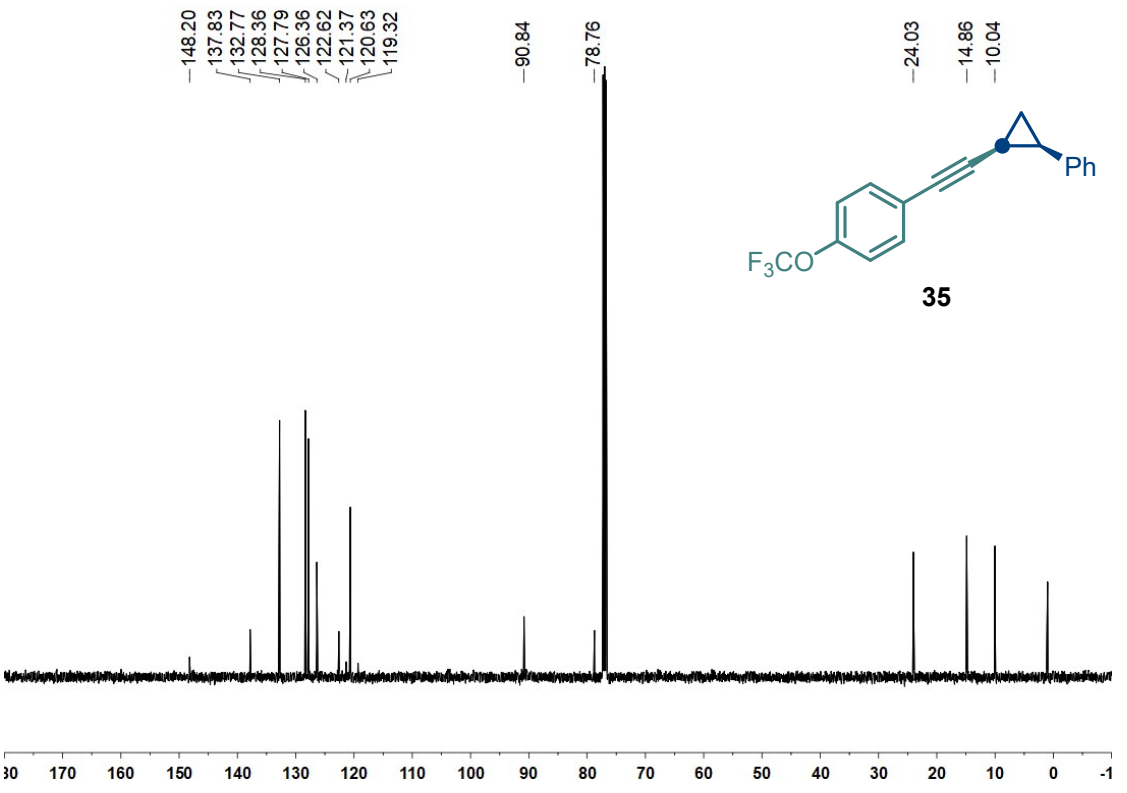
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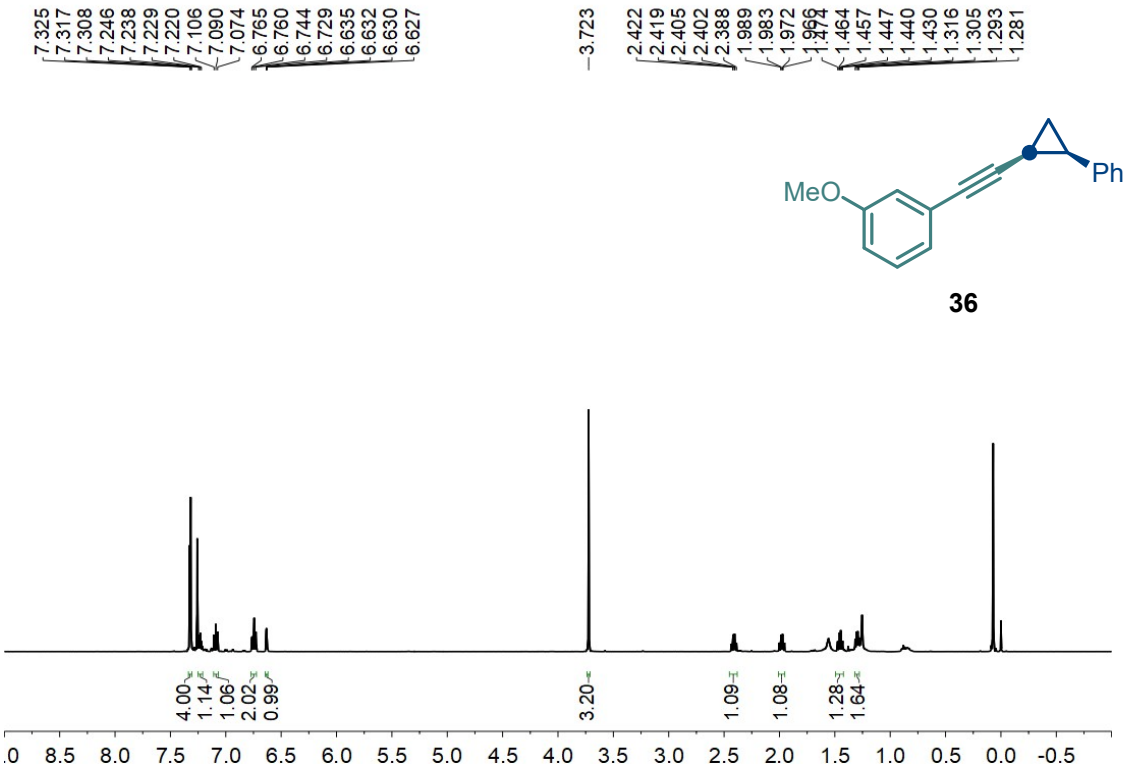
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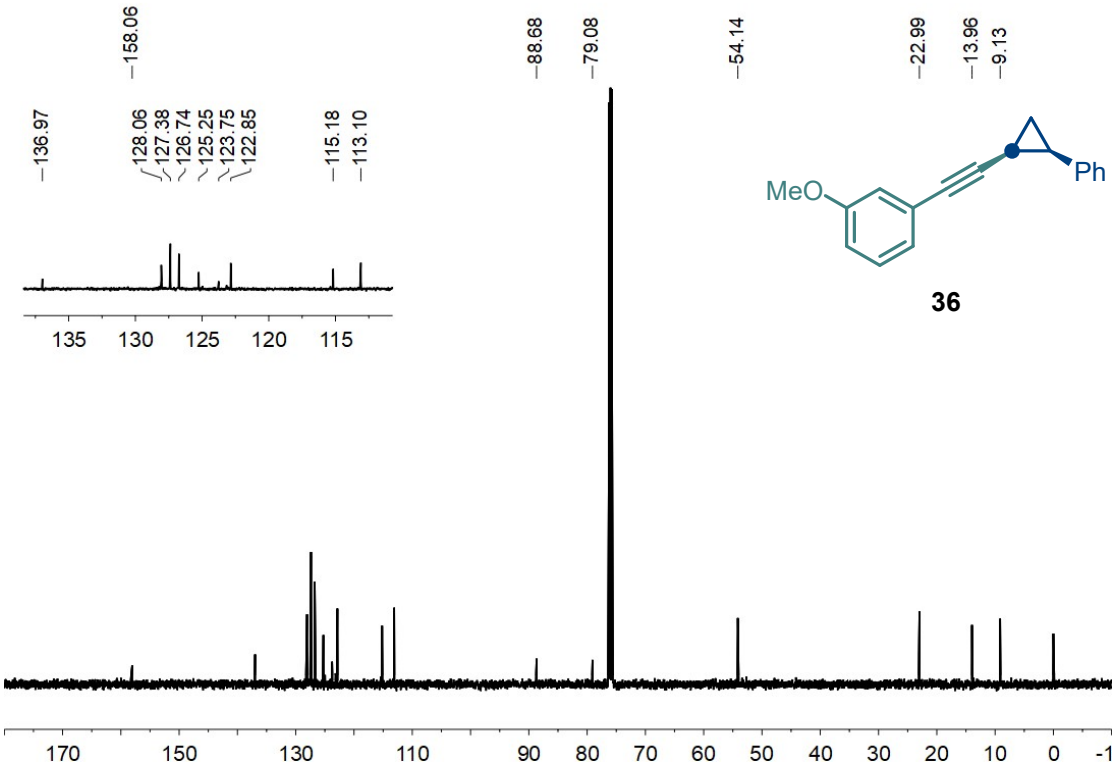
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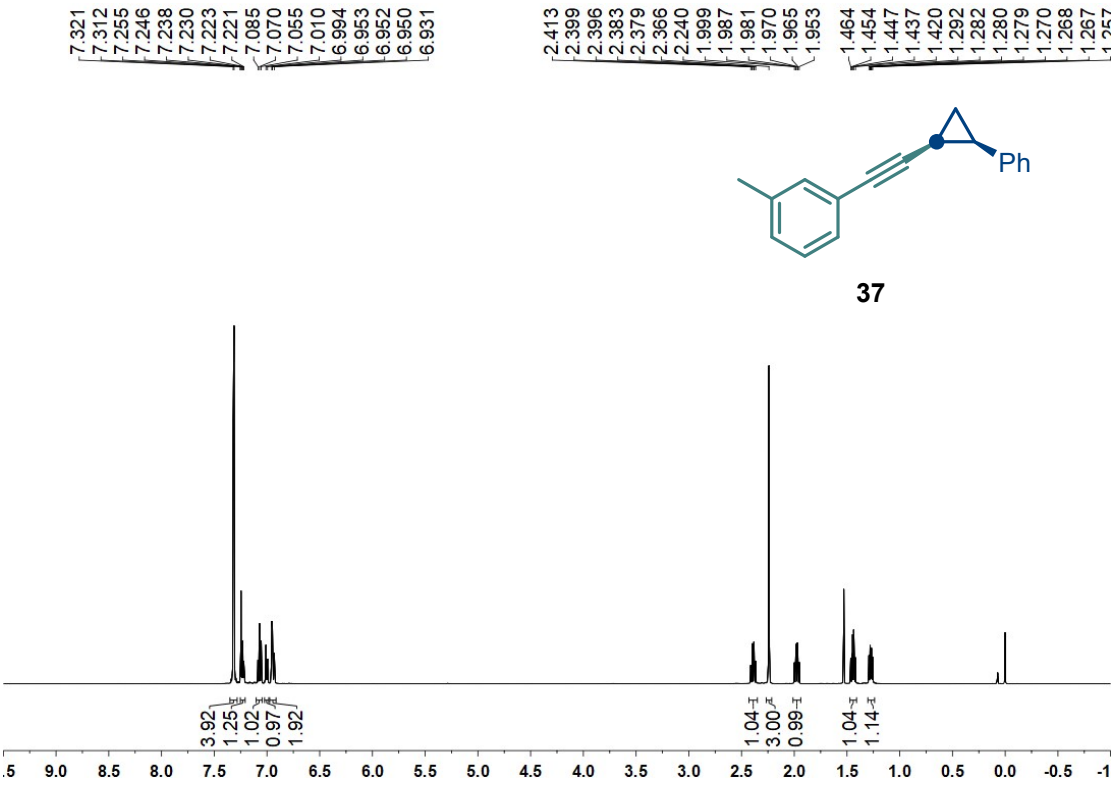
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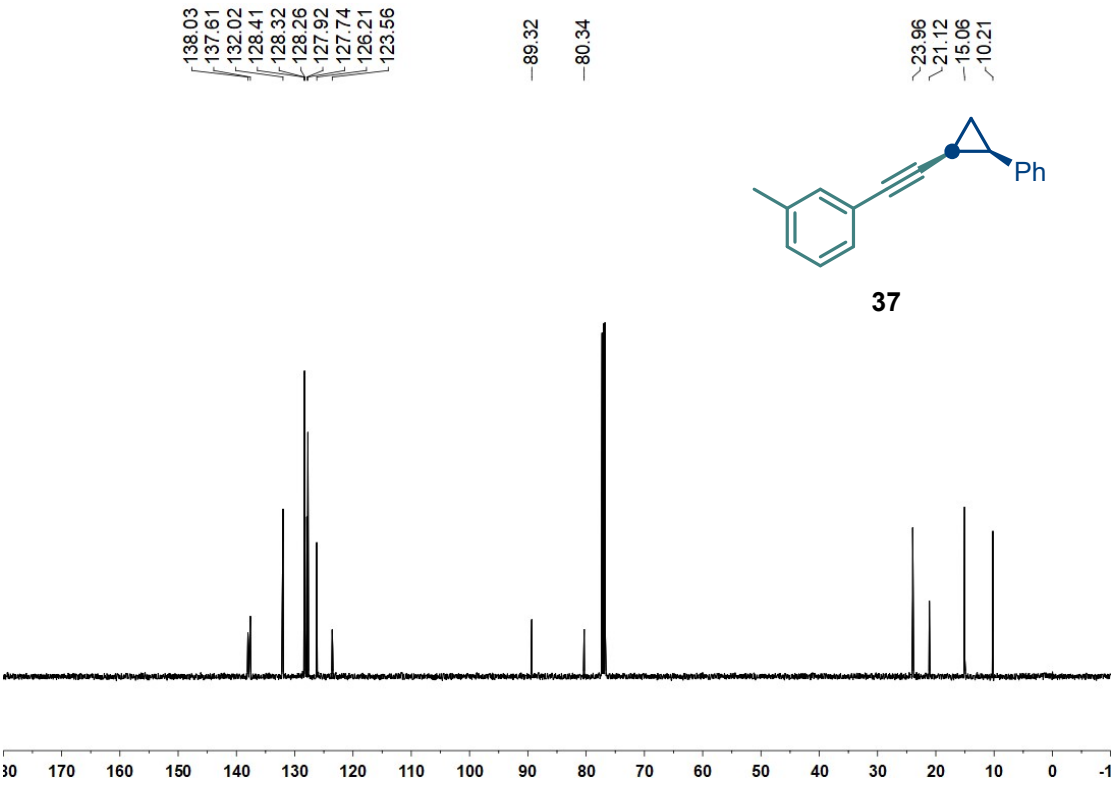
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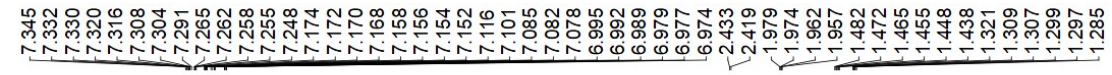
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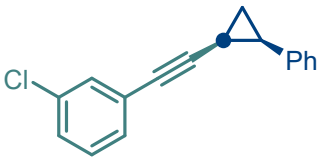
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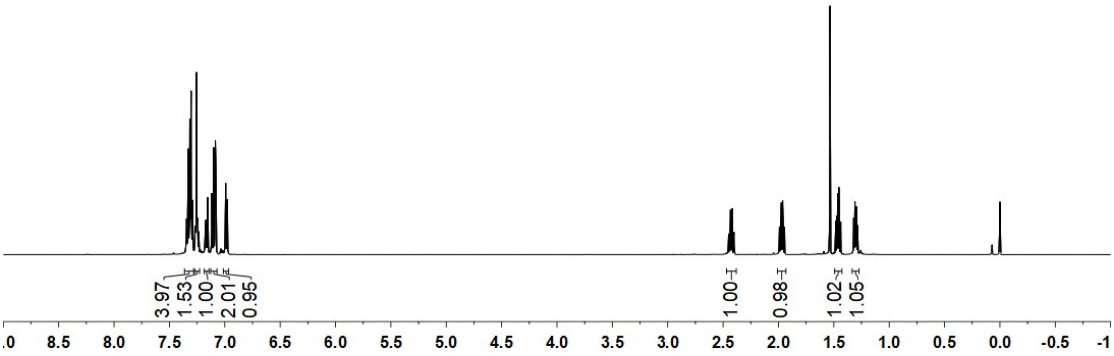
38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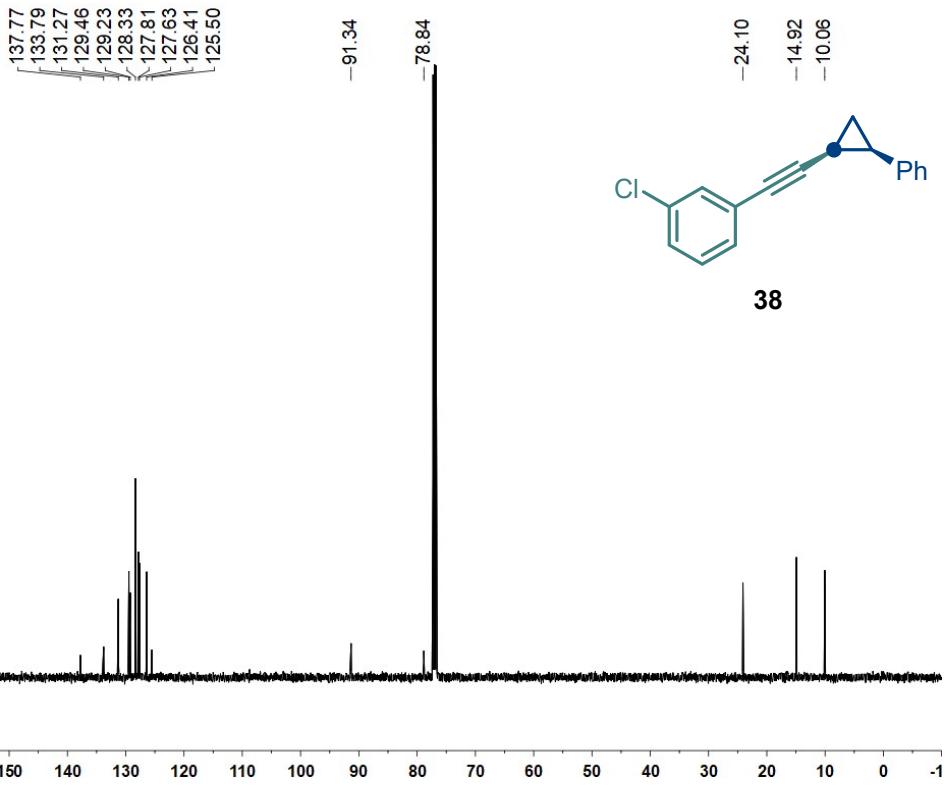
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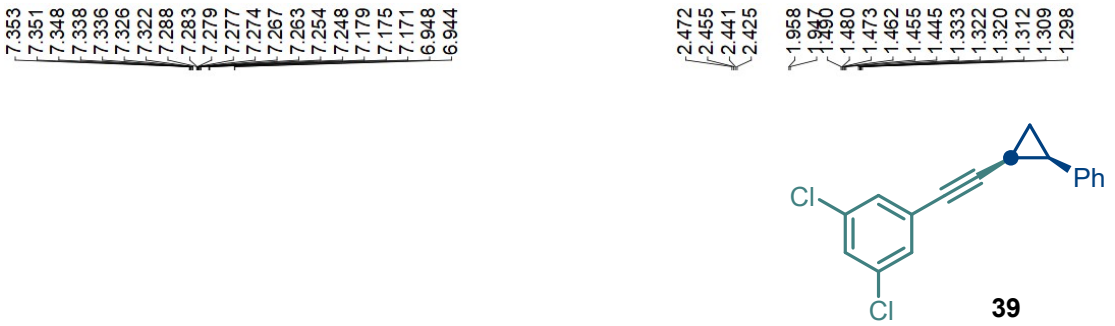
0

Cl
Ph

38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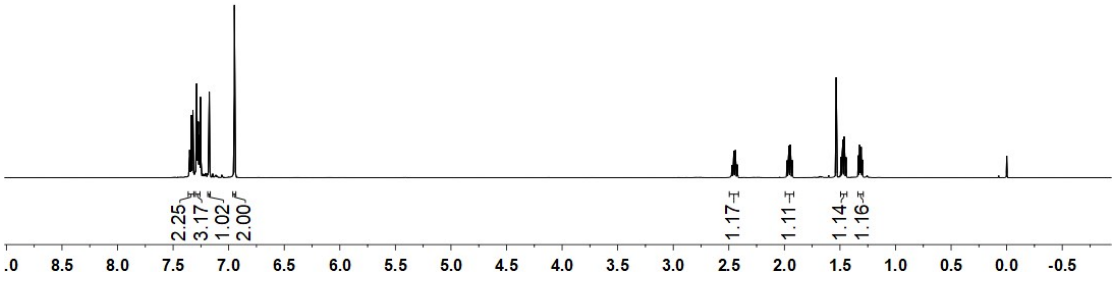
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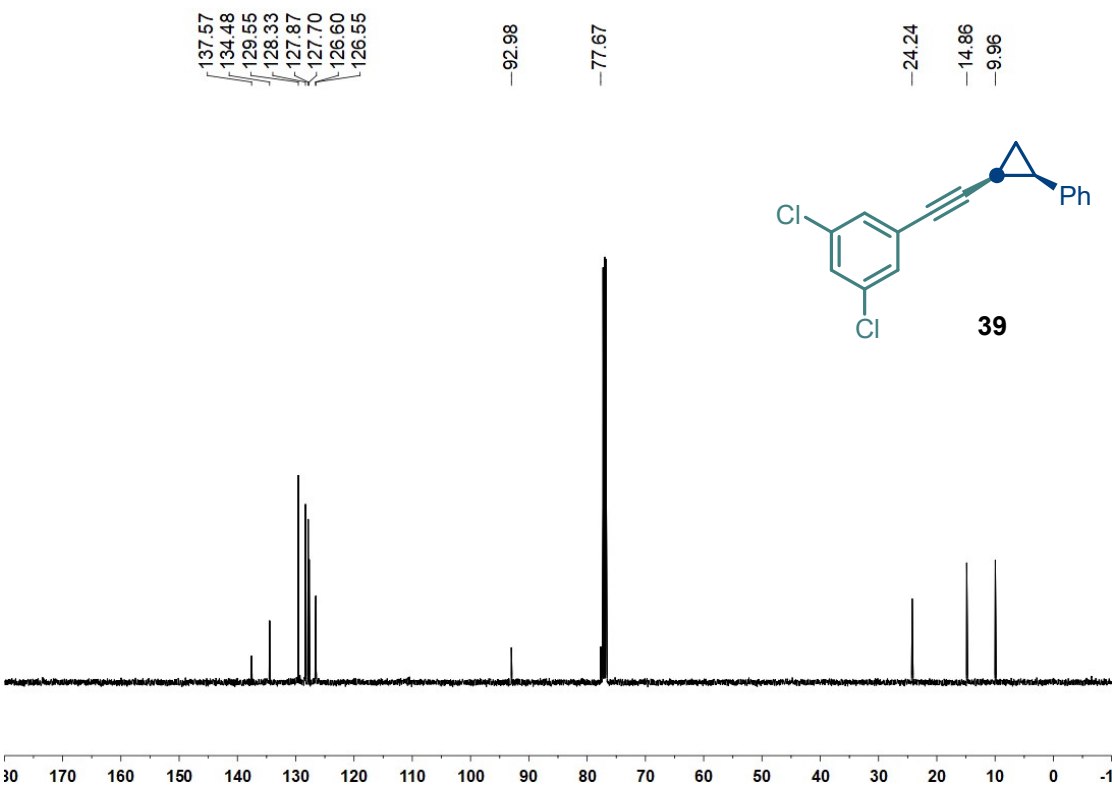
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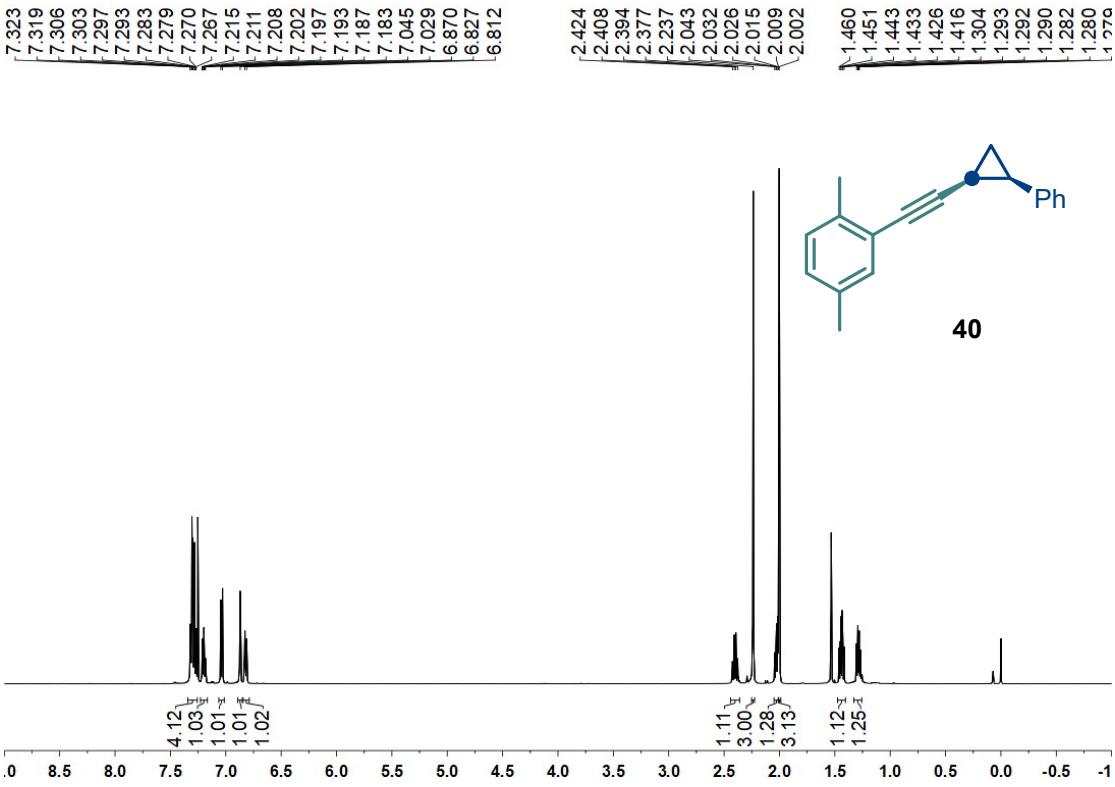
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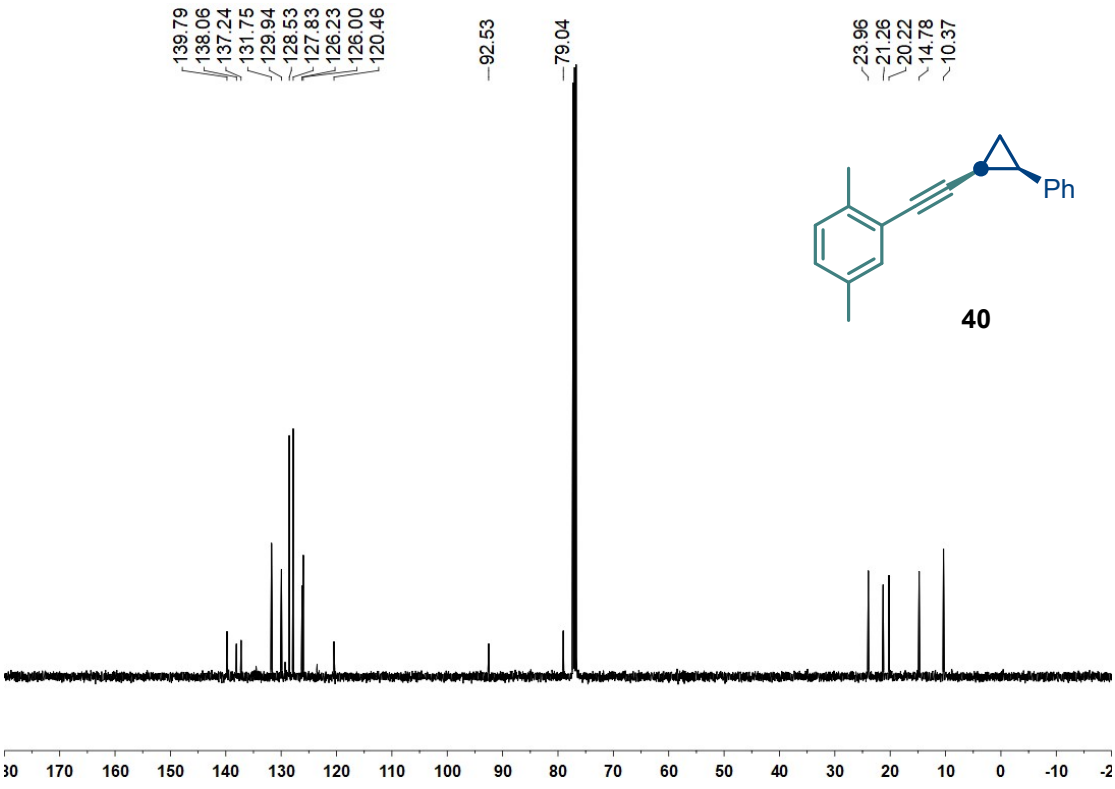
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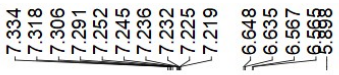
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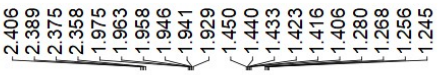
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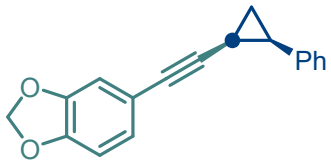
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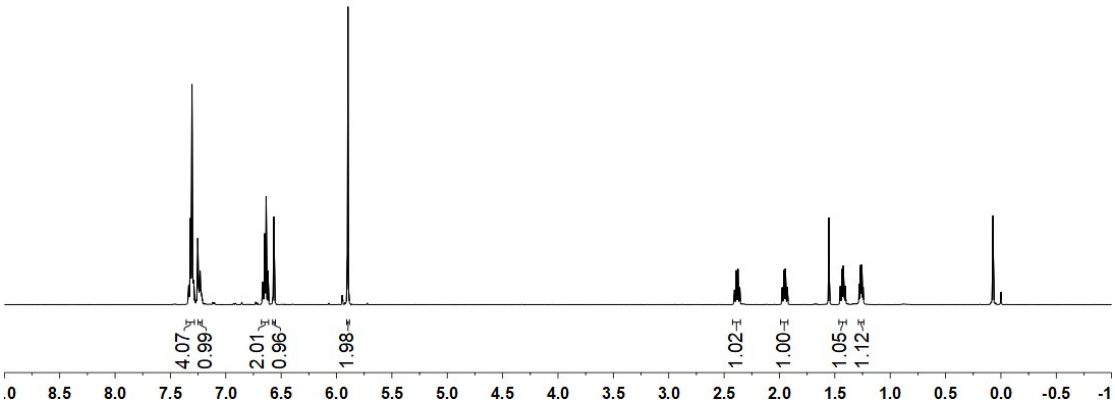
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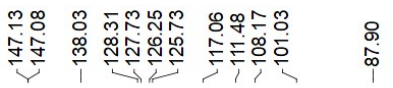
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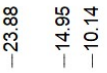
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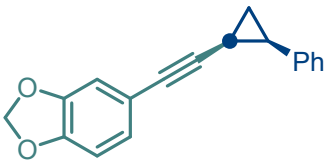
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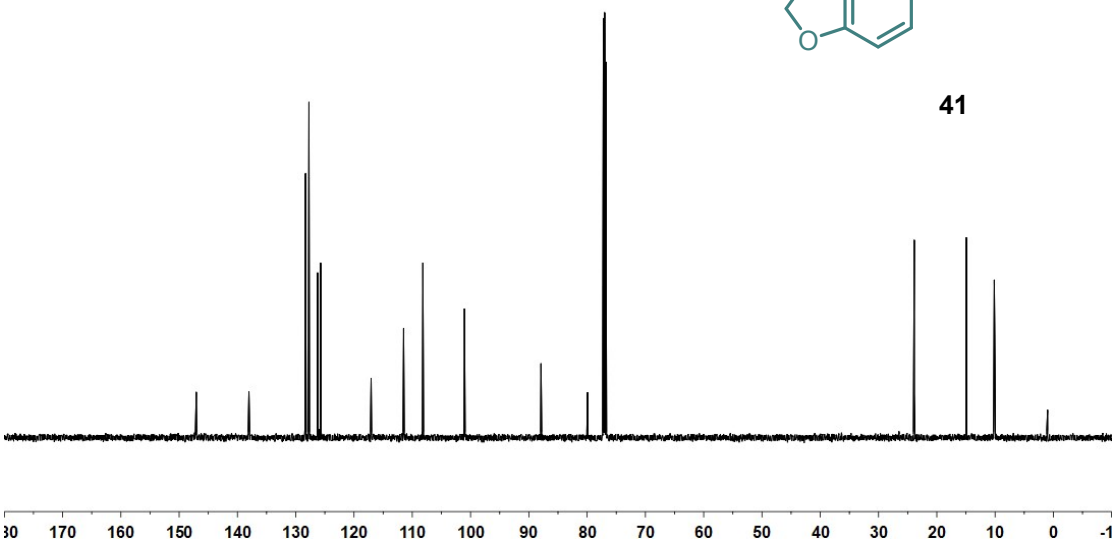
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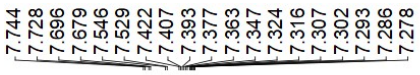
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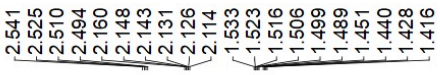
7.278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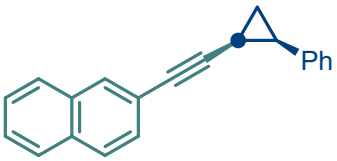
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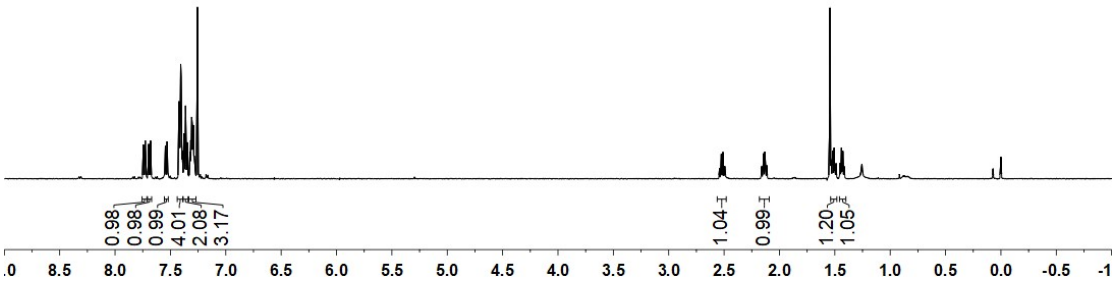
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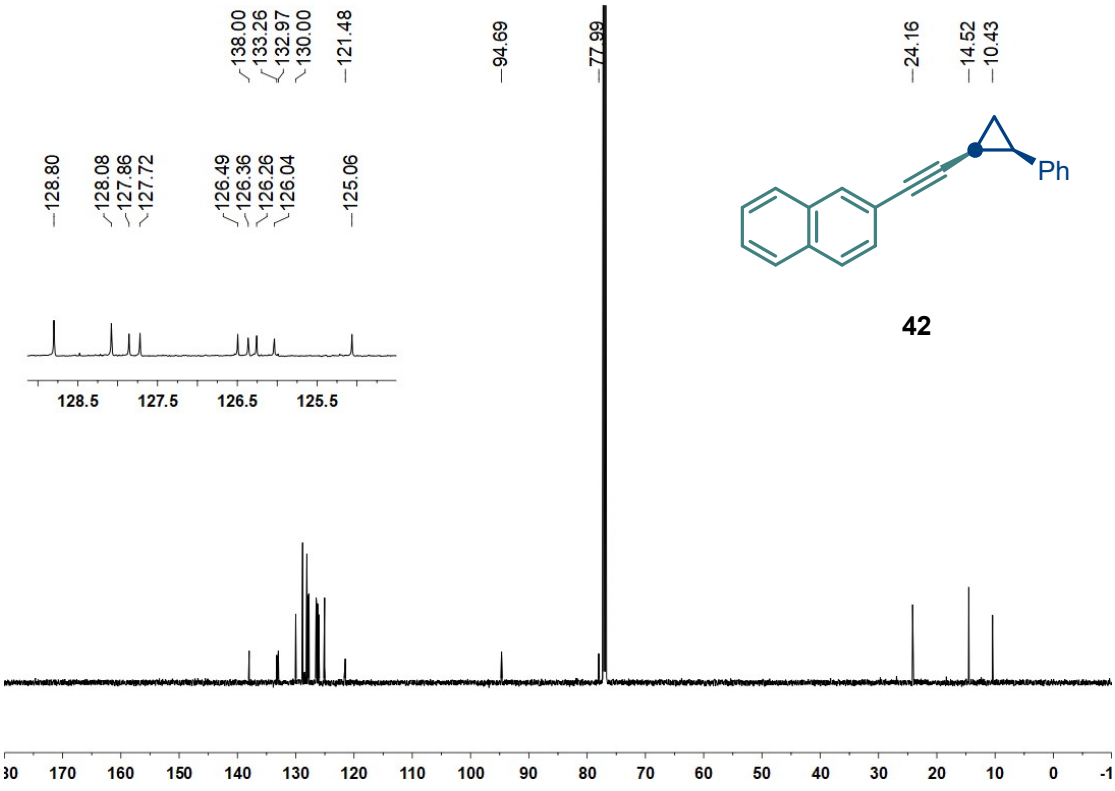
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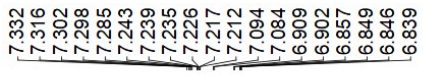
Ph

42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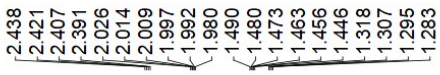
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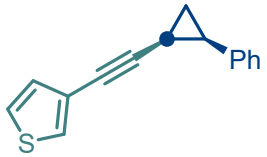
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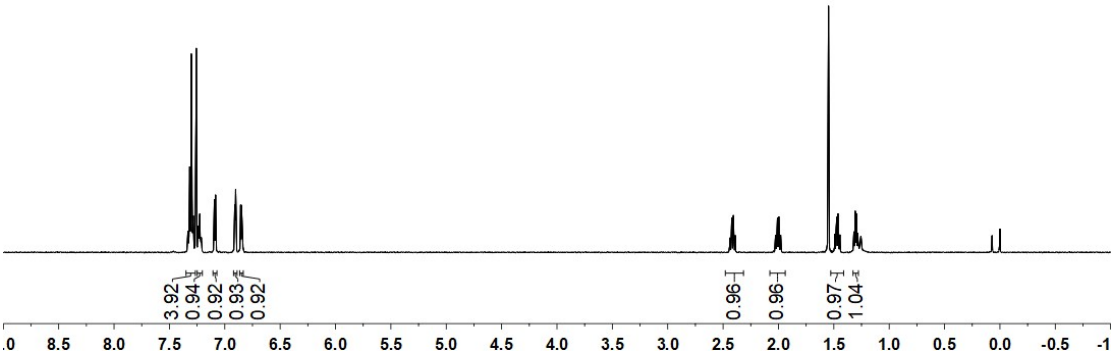
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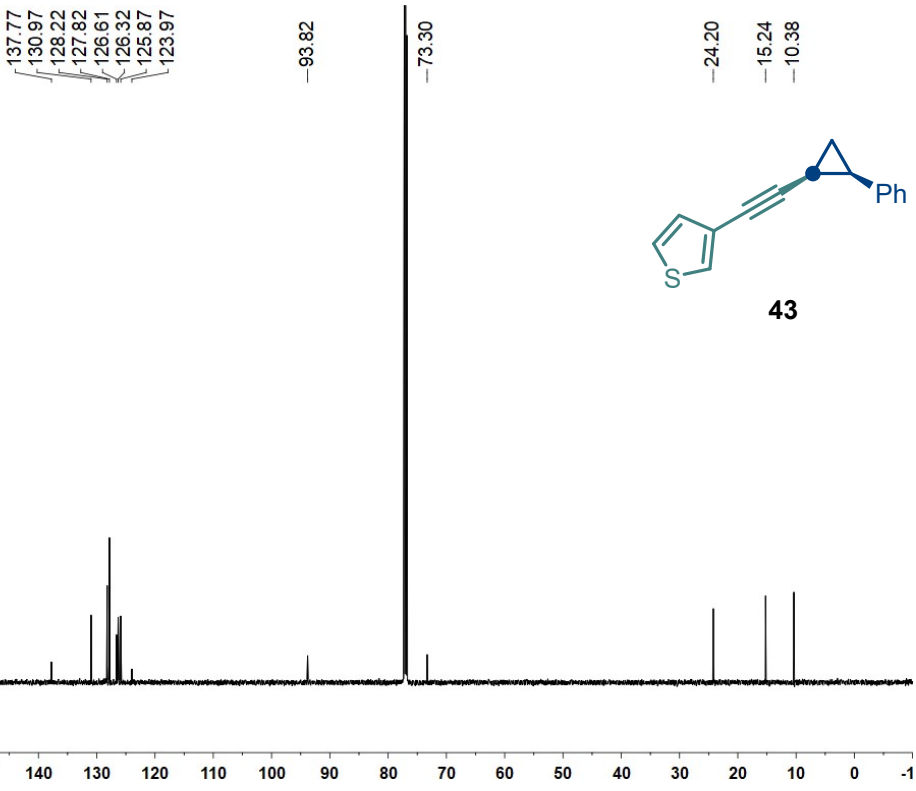
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0

Ph
S

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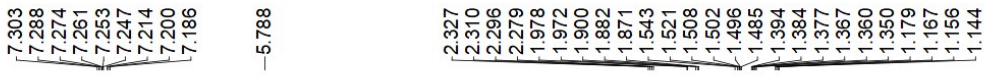
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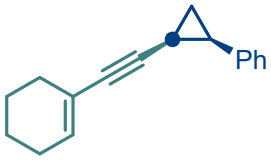
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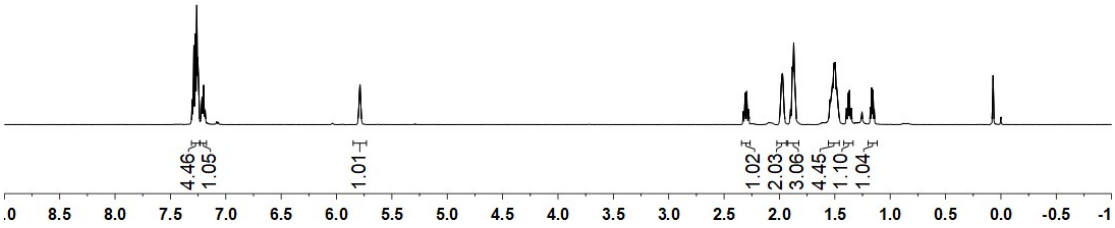
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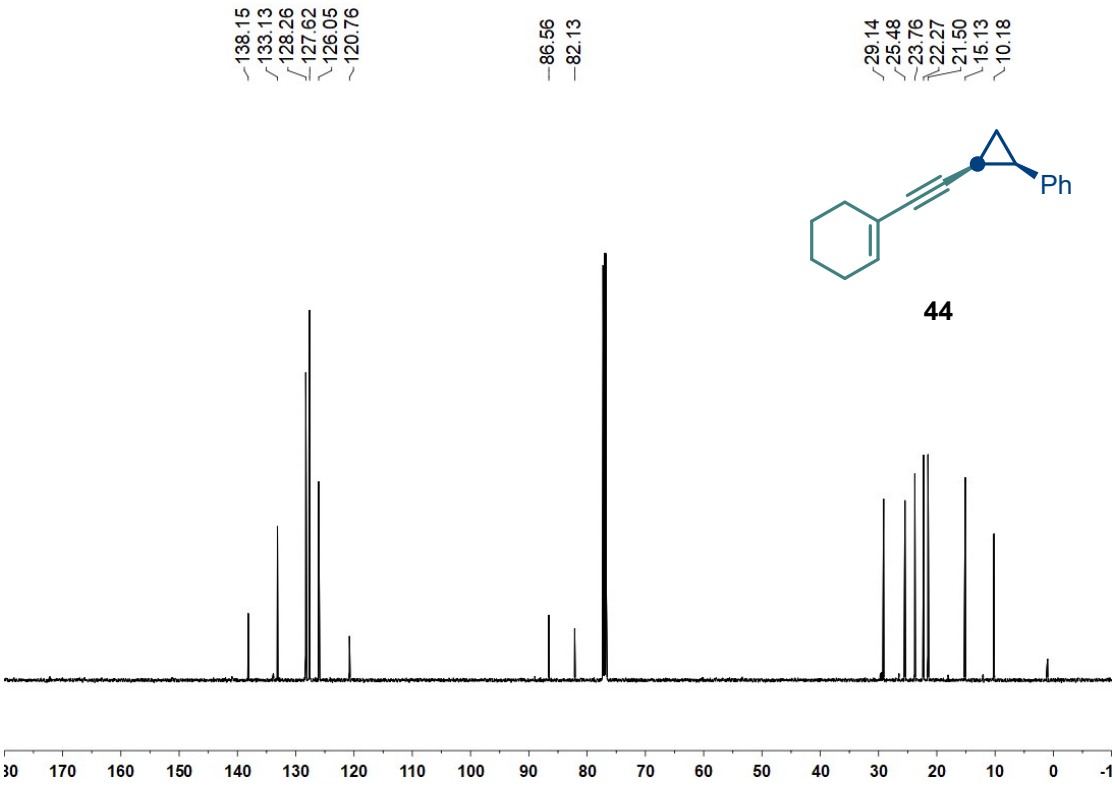
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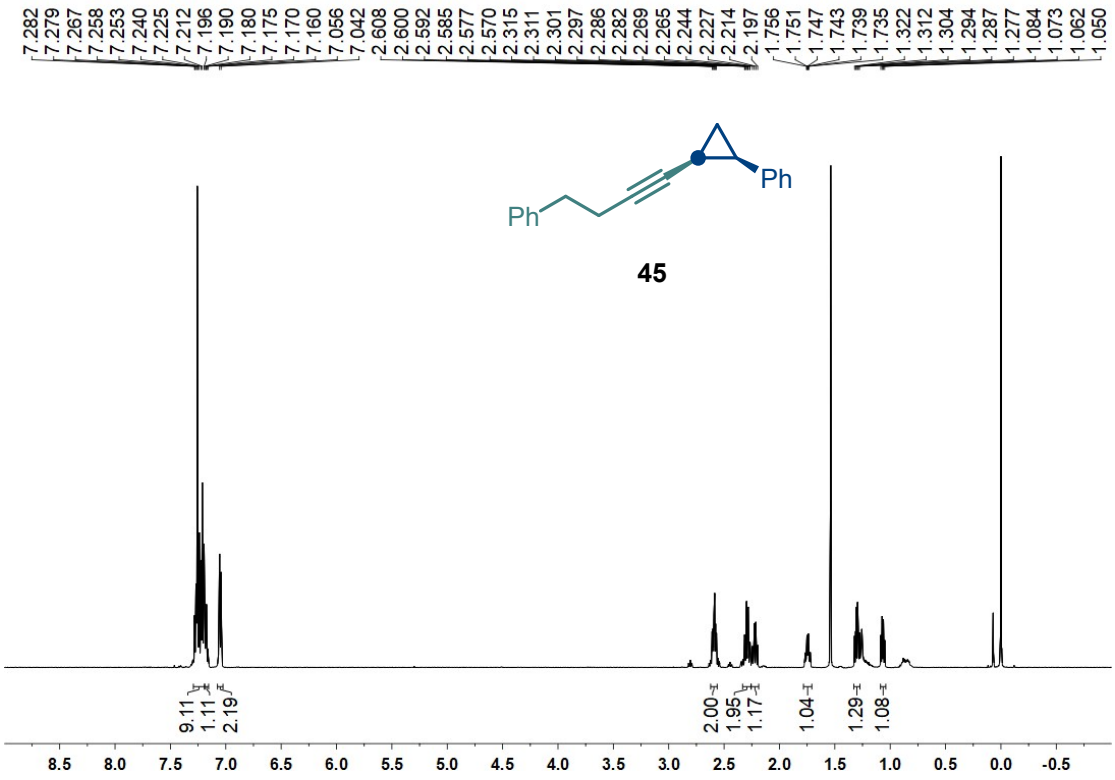
Ph

44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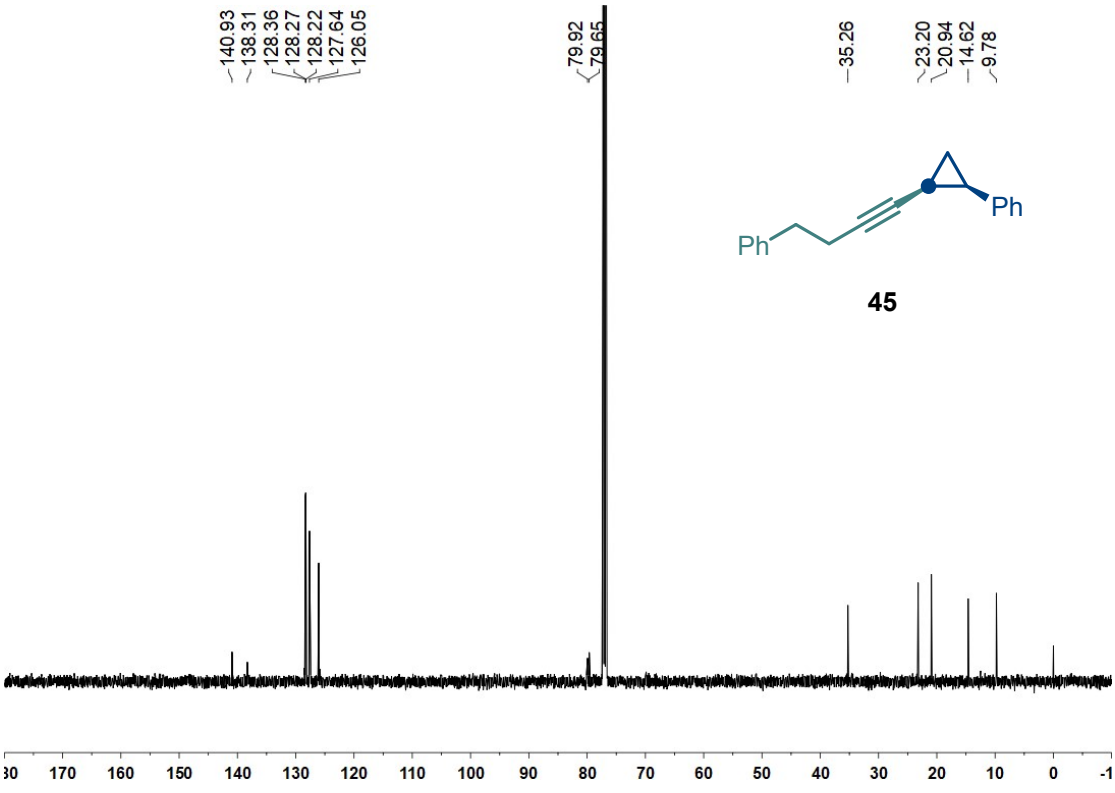
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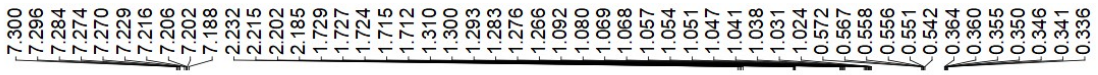
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2.202
2.185
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1.715
1.712
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1.300
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1.283
1.276
1.266
1.092
1.080
1.069
1.068
1.057
1.054
1.051
1.047
1.041
1.038
1.031
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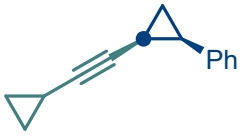
0.336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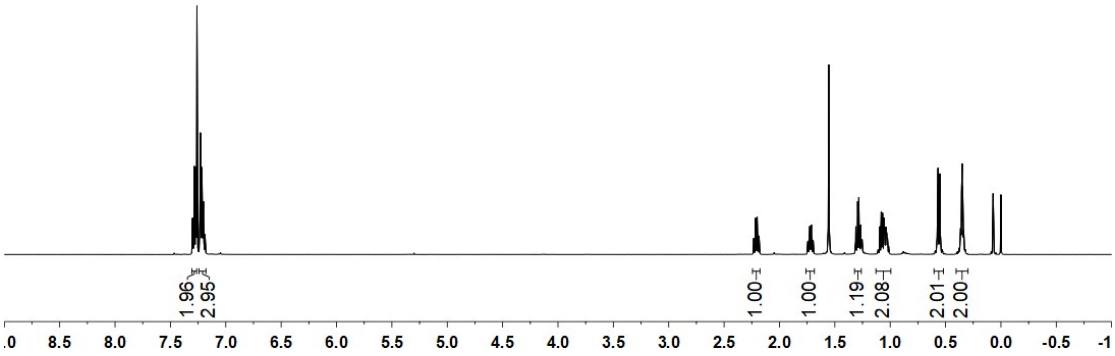
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2.00
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6.5
6.0
5.5
5.0
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3.5
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2.5
2.0
1.5
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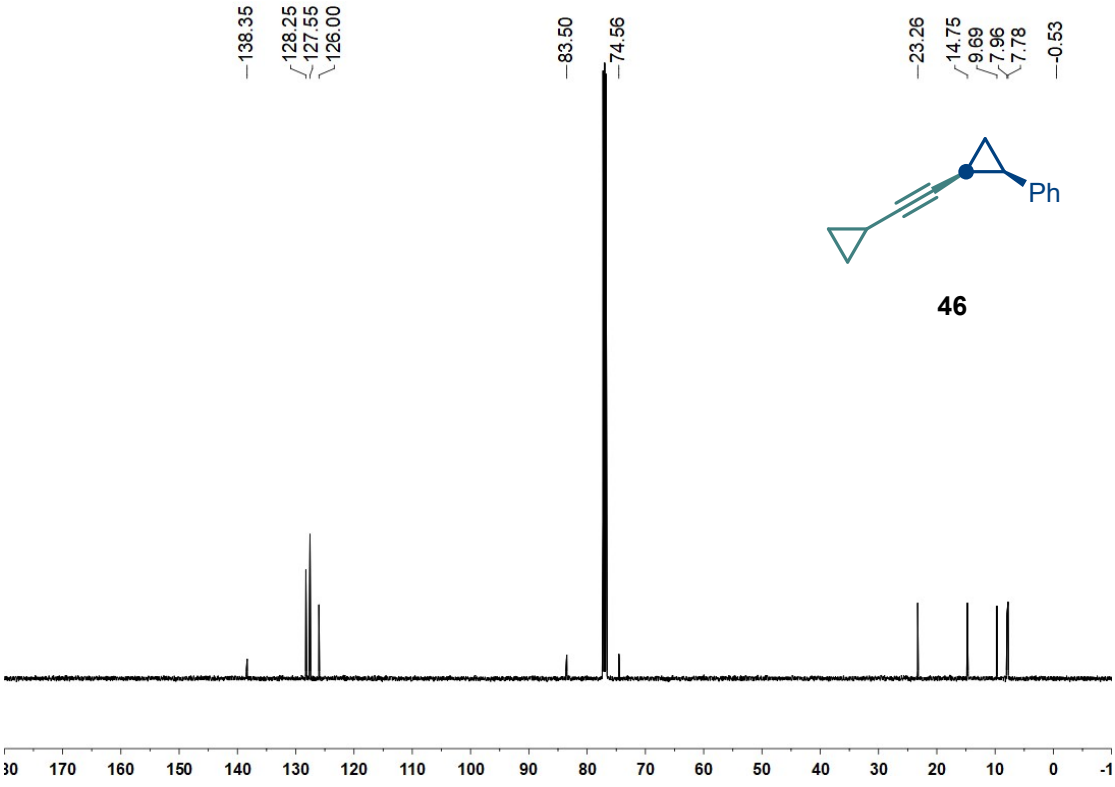
-0.5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127.55
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14.75
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7.78
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130
120
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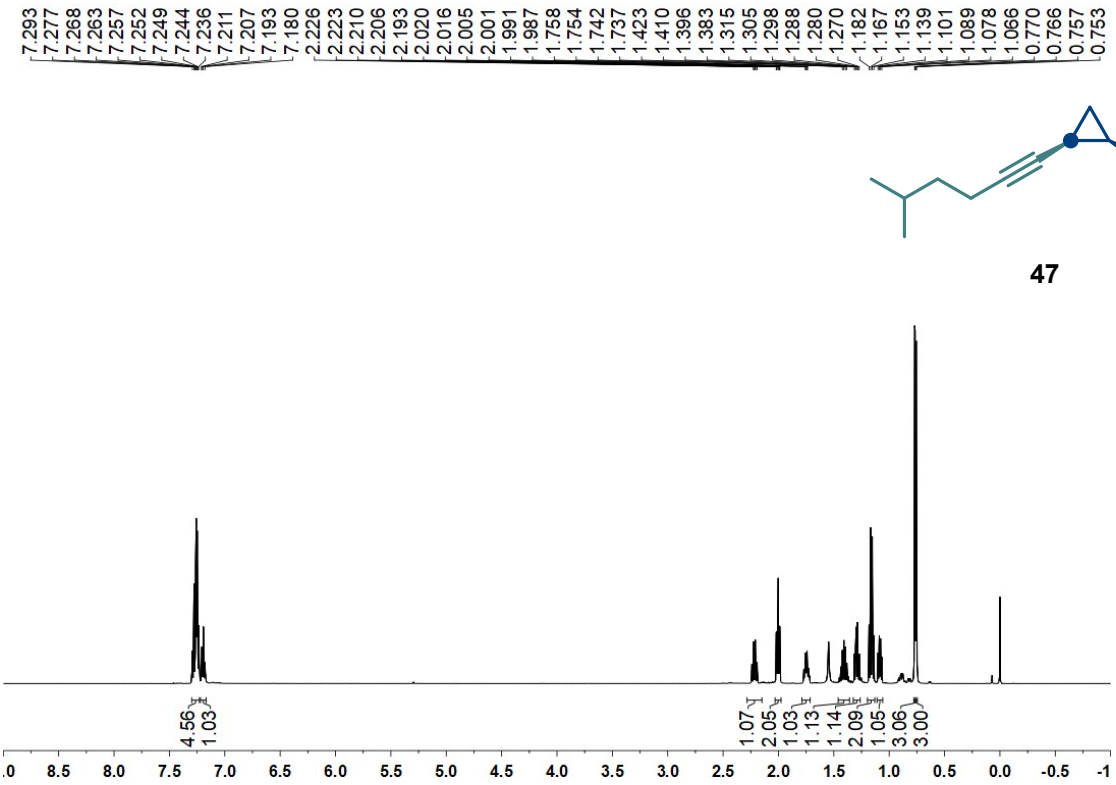
100
90
80
70
60
50
40
30
20
10
0

Ph

46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7.180
2.226
2.223
2.210
2.206
2.193
2.020
2.016
2.005
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1.078
1.066
0.770
0.766
0.757
0.753
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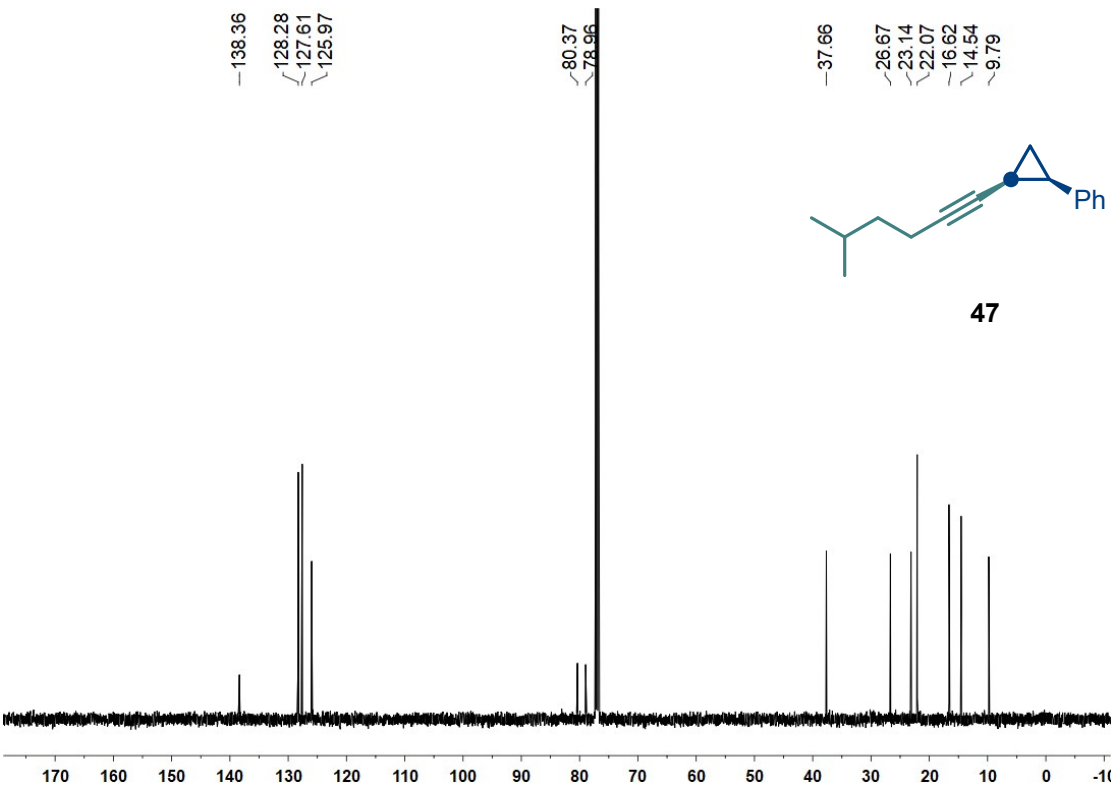
1.07
2.05
1.03
1.13
1.14
2.09
1.05
3.06
3.00

47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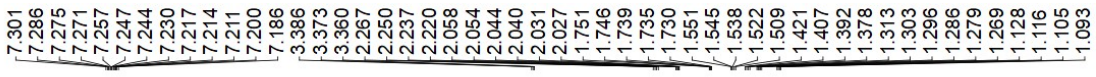
14.54
9.79
Ph

47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7.230
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7.214
7.211
7.200
7.186
3.386
3.373
3.360
2.267
2.250
2.237
2.220
2.058
2.054
2.044
2.040
2.031
2.027
1.751
1.746
1.739
1.735
1.730
1.551
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1.509
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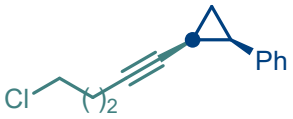
1.093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Cl
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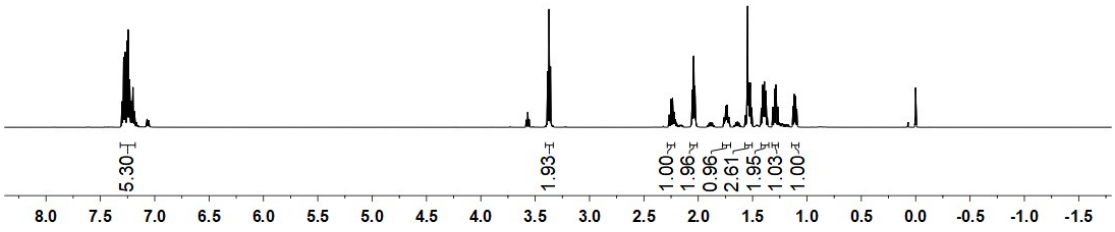
48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1.03
1.00
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7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
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2.5
2.0
1.5
1.0
0.5
0.0
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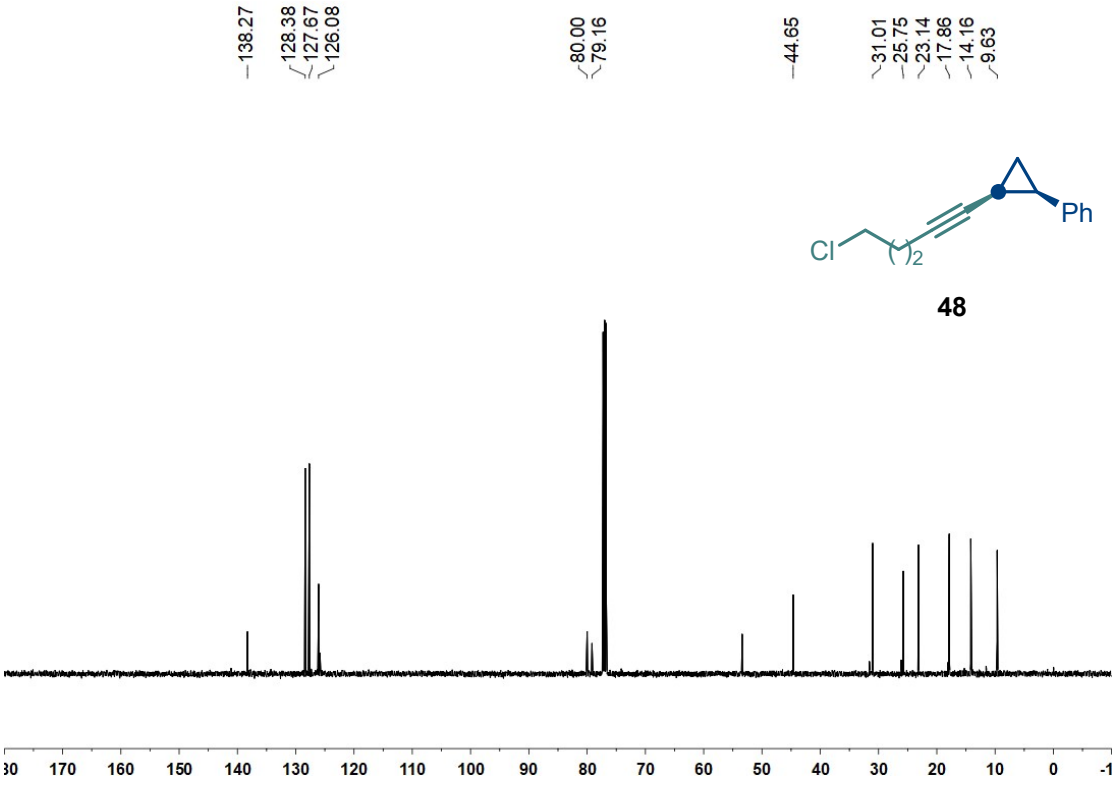
-1.0
-1.5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31.01
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23.14
17.86
14.16
9.63
170
160
150
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100
90
80
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60
50
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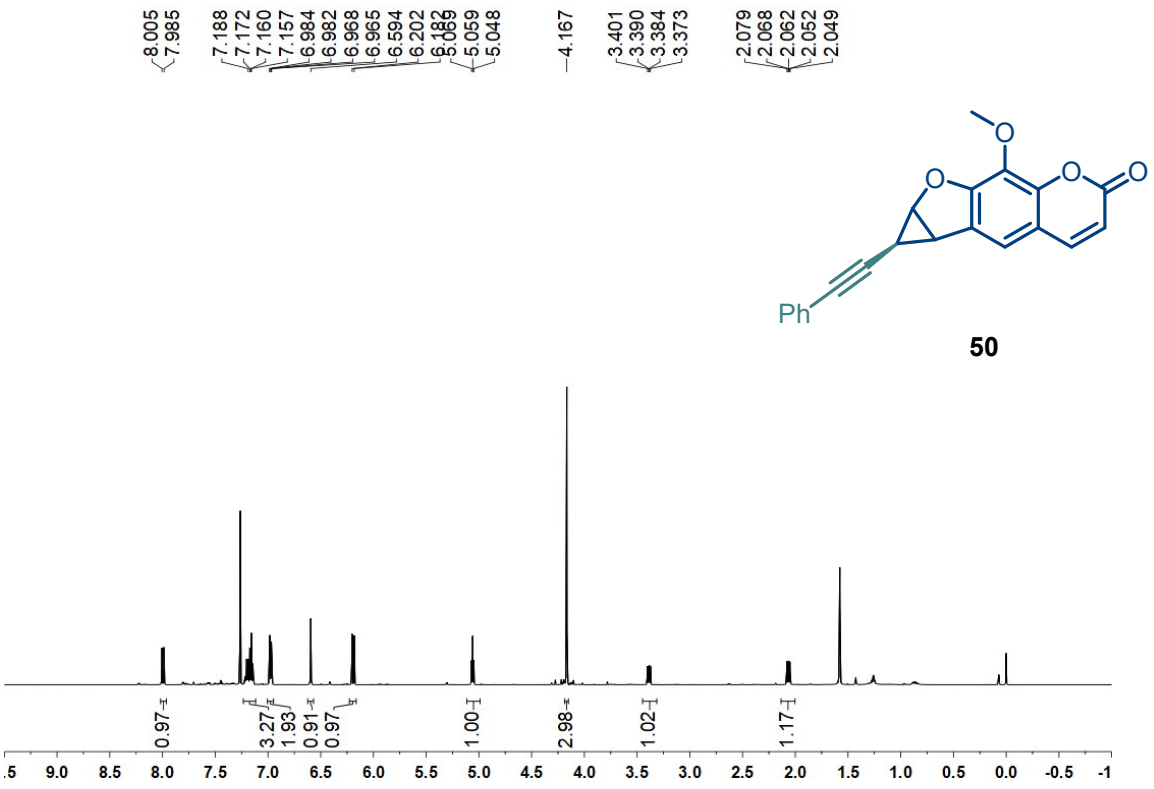
20
10
0
-1

Ph
Cl
2

48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6.594
6.202
5.059
5.048
4.167
3.401
3.390
3.384
3.373
2.079
2.068
2.062
2.052
2.049
O
O
O
O
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1.93
0.91
0.97
1.00
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1.02
1.17

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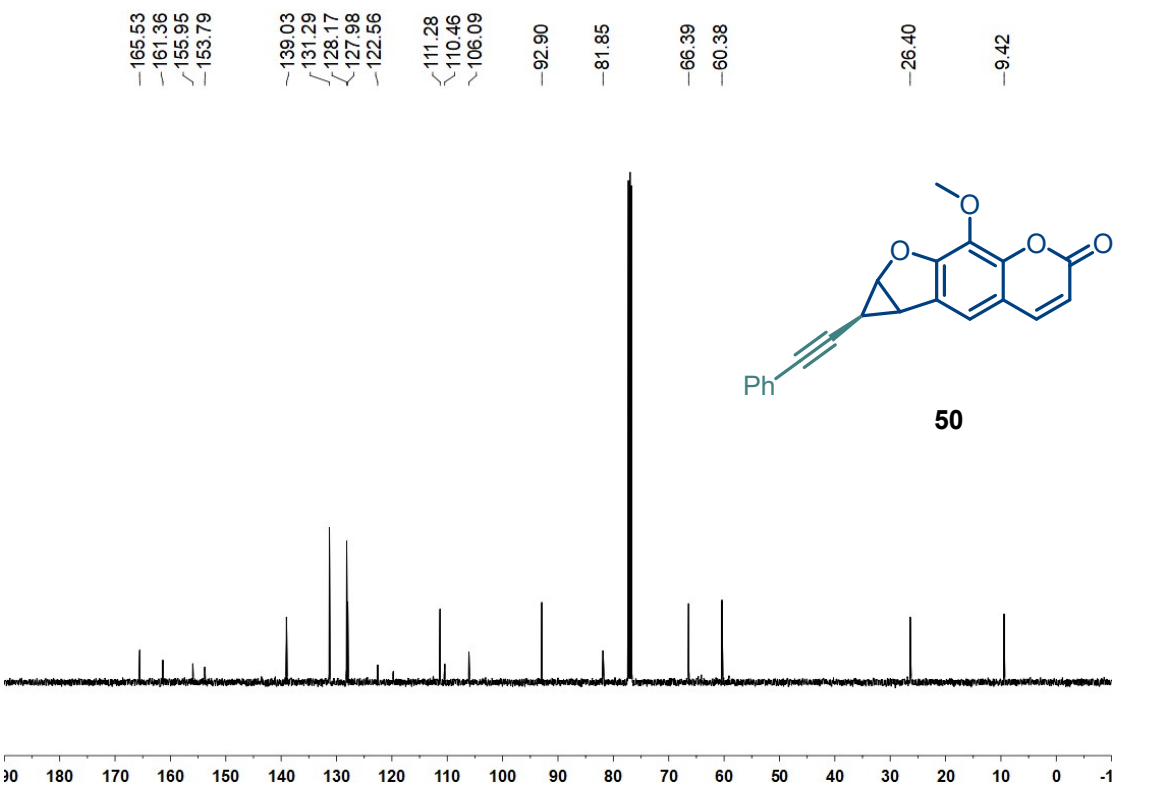
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128.17
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92.90
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66.39
60.38
26.40
9.42
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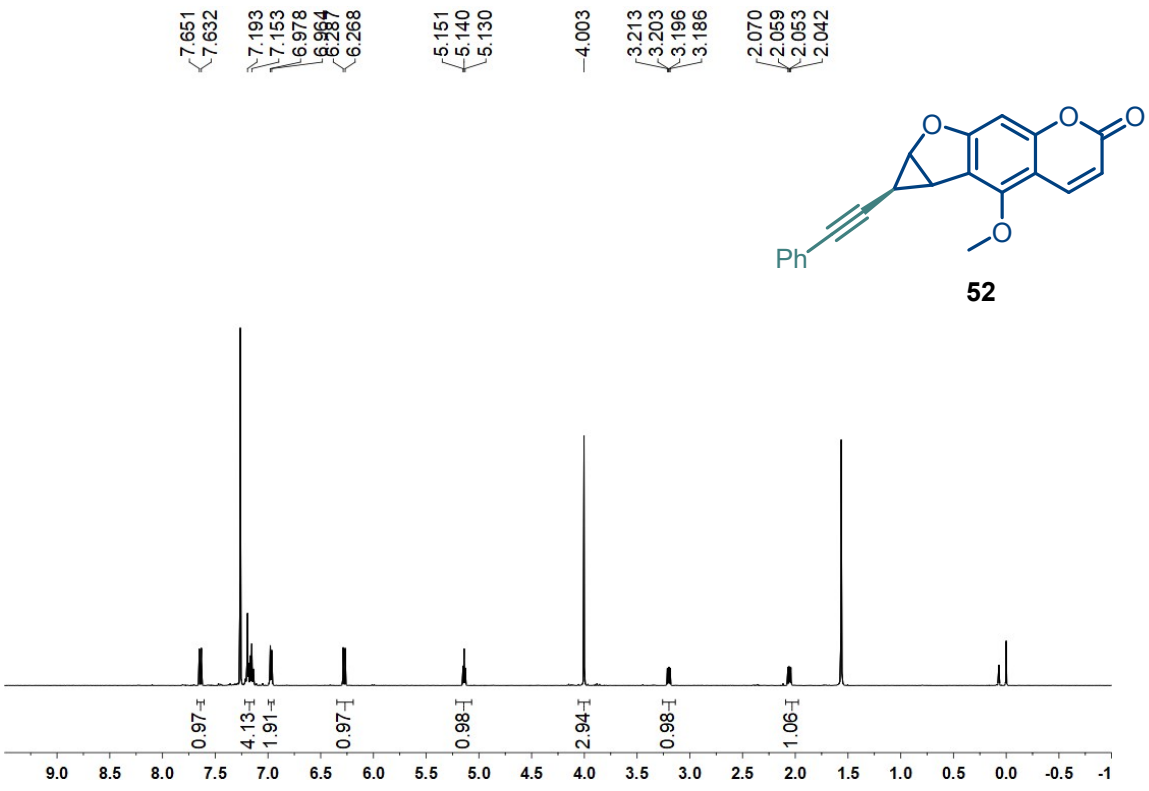
O
O
O
Ph

50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5.140
5.130
4.003
3.213
3.203
3.196
3.186
2.070
2.059
2.053
2.042
O
O
O
O
Ph
0.97
4.13
1.91
0.97
0.98
2.94
0.98
1.06
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
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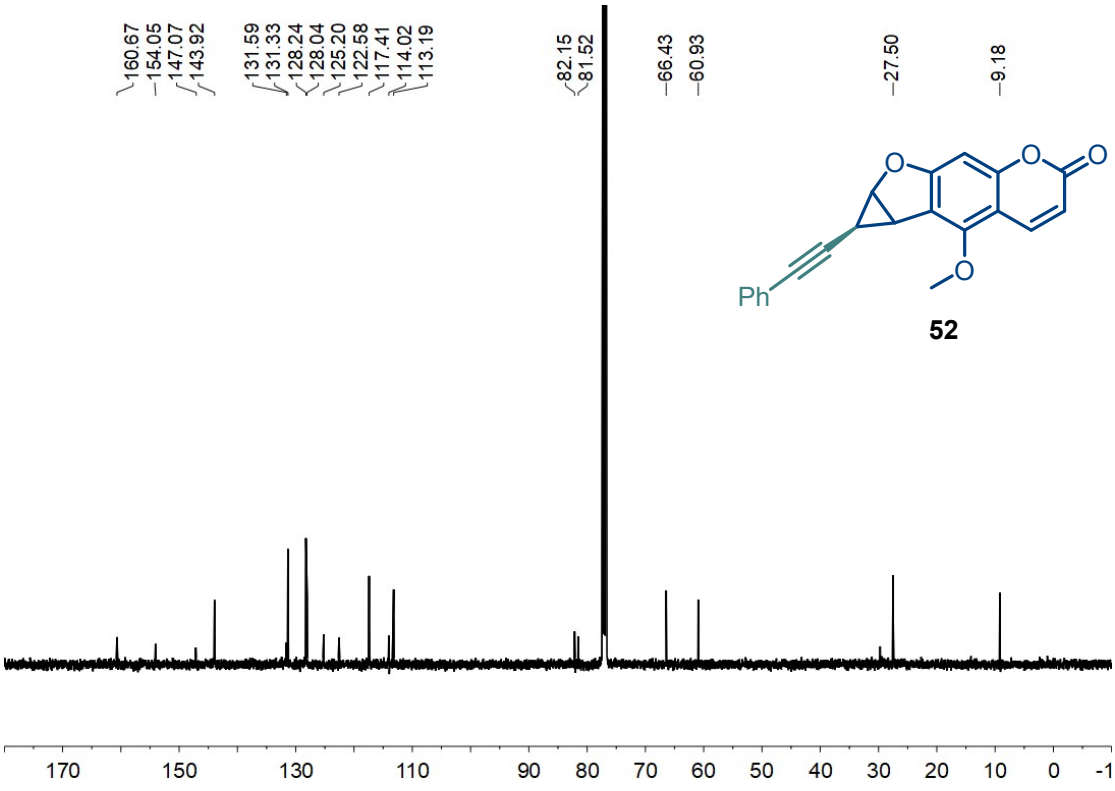
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
-1

52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147.07
143.92
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131.33
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128.04
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122.58
117.41
114.02
113.19
82.15
81.52
66.43
60.93
27.50
9.18
O
O
O
O
Ph
170
150
130
110
90
80
70
60
50
40
30
20
10
0
-1

52