

Supporting Information

Methanol as Hydrogen source: Room-temperature Highly-selective Transfer Hydrogenation of α,β -unsaturated Ketones

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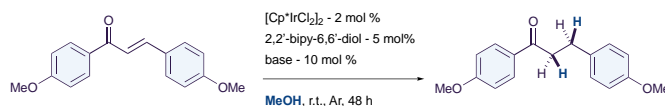
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1. General Information: Unless otherwise mentioned, all reactions were carried out under argon atmosphere. Reagent grade methanol was received from the commercial source (Merck) and used as such. ^1H and ^{13}C NMR were recorded on JEOL (500 MHz) using CDCl_3 as solvent. Chemical shifts (δ) are given in ppm relative to TMS, coupling constants (J) in Hz. The solvent signals used as references and the chemical shifts converted to TMS scale (CDCl_3 : $\delta_{\text{C}} = 77.0$ ppm; residual CHCl_3 in CDCl_3 : $\delta_{\text{H}} = 7.26$ ppm). All the reactions were monitored by analytical thin layer chromatography using commercial aluminium sheets precoated with silica gel. Column Chromatography was performed on silica gel (Merck, 200-400 mesh). EI-MS/ESI-MS was recorded on Agilent 6546LC/Q-TOF mass spectrometer. All other chemicals were received and used as such from the various commercial sources like sigma-aldrich, alfa-aesar, avra synthesis, TCI etc. CD_3OD (99.80% D) was purchased from Eurisotop.

Catalyst $[\text{Cp}^*\text{IrCl}_2]_2$ was prepared according to the reported procedure in literature.¹

Substrates **1a-1n**, **1q-1z** and **1ac-1ad** were prepared according to the reported literature procedures.²⁻⁴ Substrates **1o**⁵, **1p**⁶, **1aa**⁷, **1ab**⁸, **1ae**⁹, **1ah**¹⁰ were prepared according to the reported literature procedures.

2. Reaction Optimization:



Entry ^[a]	[M] (mol %)	Ligand (mol %)	Base (mol %)	Solvent (Conc.)	Yield(%) ^[b]
1	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	KOH (10)	MeOH (0.3 M)	86
2	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	NaOH (10)	MeOH (0.2 M)	82
3	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	K_2CO_3 (10)	MeOH (0.2 M)	83
4	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	KOH (10)	MeOH (0.2 M)	75
5	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	^t BuOK (10)	MeOH (0.2 M)	2.5
6	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.2 M)	90
7	-	L (5)	Cs_2CO_3 (10)	MeOH (0.2 M)	n.d.
8	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	-	Cs_2CO_3 (10)	MeOH (0.2 M)	traces
9	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	-	MeOH (0.2 M)	n.d.
10	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	Toluene (0.2 M)	n.d.
11	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	90
12	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.1 M)	32
13	$[\text{Cp}^*\text{IrCl}_2]_2$ (1)	L (2.5)	Cs_2CO_3 (10)	MeOH (0.5 M)	59
14	$[\text{Cp}^*\text{IrCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	24 ^[c]
15	$\text{Mn}(\text{CO})_5\text{Br}$ (4)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	n.d.
16	$\text{Fe}_2(\text{CO})_9$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	traces
17	$[\text{Ru}(\text{p-cymene})\text{Cl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	<5%
18	$[\text{Cp}^*\text{RhCl}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	n.d.
19	$[\text{Cp}^*\text{CoI}_2]_2$ (2)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	n.d.
20	$\text{Cp}^*\text{Co}(\text{CO})\text{I}_2$ (4)	L (5)	Cs_2CO_3 (10)	MeOH (0.5 M)	n.d.

^[a]All reagents were added under argon atmosphere unless otherwise stated using **1a**/[Ir]/L/Base in 0.2/0.004/0.01/0.02 mmol in CH_3OH and closed Schlenk tube is stirred at room temperature for 48 h. ^[b]Yields are isolated yields. ^[c]Reaction was performed in air.

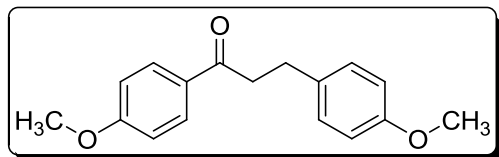
3. General procedure A:

Ir(III)-Catalyzed transfer hydrogenation of α,β -unsaturated ketones using methanol

An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with $[\text{Cp}^*\text{IrCl}_2]_2$ (4.7 mg, 0.006 mmol, 2 mol%), 2,2'-bipyridine-6,6'-diol (2.8 mg, 0.015 mmol, 5 mol%), Cs_2CO_3 (9.7 mg, 0.03 mmol, 10 mol%) and *(E)*-1,3-bis(4-methoxyphenyl)prop-2-en-1-one **1a** (0.3 mmol, 1.0 equiv.) followed by addition of 0.6 mL of methanol (0.5 M) under argon atmosphere. The closed tube was stirred at room temperature for 48 h. After completion of the reaction, the crude reaction mixture was purified by column chromatography using EtOAc/hexane as mobile phase to obtain analytically pure saturated ketones in moderate-to-excellent yields.

4. Analytical data of the products:

1,3-bis(4-methoxyphenyl)propan-1-one (2a): Compound **2a** was prepared according to the general

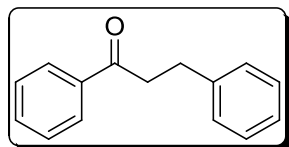


procedure **A** starting from **1a** (53.6 mg, 0.2 mmol) and purified by column chromatography (EtOAc:hexane =8:92) afforded **2a** in 90% yield (48.8 mg) as white solid. The NMR data is in accordance with the literature.^[12]

¹H NMR (500 MHz, CDCl₃): δ 7.94 (d, *J* = 8.5 Hz, 2H), 7.17 (d, *J* = 8.5 Hz, 2H), 6.92 (d, *J* = 8.8 Hz, 2H), 6.84 (d, *J* = 8.6 Hz, 2H), 3.86 (s, 3H), 3.78 (s, 3H), 3.21 (t, *J* = 7.6 Hz, 2H), 3.00 (t, *J* = 7.8 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 197.8, 163.3, 157.8, 133.3, 130.1, 129.9, 129.2, 113.8, 113.6, 55.3, 55.1, 40.2, 29.3.

1,3-diphenylpropan-1-one (2b): Compound **2b** was prepared according to the general procedure **A** starting

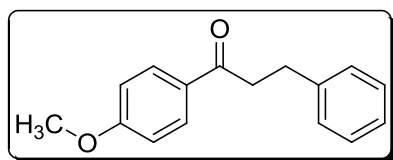


from **1b** (62.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2b** in 80% yield (50.4 mg) as white solid. The NMR data is in accordance with the literature.^[11]

¹H NMR (500 MHz, CDCl₃): δ 7.98-7.96 (m, 2H), 7.58-7.55 (m, 1H), 7.48-7.44 (m, 2H), 7.33-7.20 (m, 5H), 3.31 (t, *J* = 8.0 Hz, 2H), 3.08 (t, *J* = 7.9 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.2, 141.3, 136.8, 133.0, 128.6, 128.5, 128.4, 128.0, 126.1, 40.4, 30.1.

1-(4-methoxyphenyl)-3-phenylpropan-1-one (2c): Compound **2c** was prepared according to the

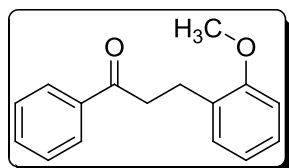


general procedure **A** starting from **1c** (71.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane=2:98) afforded **2c** in 91% yield (65.7 mg) as pale yellow solid. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 7.96-7.94 (m, 2H), 7.32-7.20 (m, 5H), 6.93-6.92 (m, 2H), 3.85 (s, 3H), 3.25 (t, *J* = 7.8 Hz, 2H), 3.06 (t, *J* = 7.9 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 197.7, 163.4, 141.4, 130.2, 129.9, 128.4, 128.3, 125.9, 113.6, 55.4, 39.9, 30.2.

3-(2-methoxyphenyl)-1-phenylpropan-1-one (2d): Compound **2d** was prepared according to the

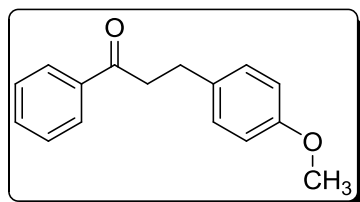


general procedure **A** starting from **1d** (71.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 3:97) afforded **2d** in 83% yield (59.8 mg) as yellow oil. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 8.00 (d, *J* = 7.2 Hz, 2H), 7.56 (t, *J* = 7.3 Hz, 1H), 7.46 (t, *J* = 7.9 Hz, 2H), 7.24-7.21 (m, 2H), 6.93-6.87 (m, 2H), 3.84 (s, 3H), 3.29 (t, *J* = 7.9 Hz, 2H), 3.08 (t, *J* = 8.0 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.9, 157.5, 136.9, 132.8, 130.1, 129.5, 128.4, 128.0, 127.4, 120.5, 110.2, 55.1, 38.9, 25.7.

3-(4-methoxyphenyl)-1-phenylpropan-1-one (2e): Compound **2e** was prepared according to the

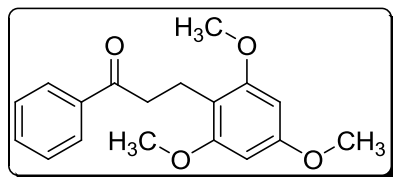


general procedure **A** starting from **1e** (71.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane = 2:98) afforded **2e** in 65% yield (47 mg) as white solid. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 7.97-7.96 (m, 2H), 7.57-7.54 (m, 1H), 7.47-7.44 (m, 2H), 7.19-7.17 (m, 2H), 6.86-6.84 (m, 2H), 3.79 (s, 3H), 3.28 (t, *J* = 7.5 Hz, 2H), 3.02 (t, *J* = 7.4 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.3, 157.9, 136.8, 133.4, 132.9, 129.3, 128.5, 127.9, 113.9, 55.2, 40.7, 29.2.

1-phenyl-3-(2,4,6-trimethoxyphenyl)propan-1-one (2f): Compound **2f** was prepared according to

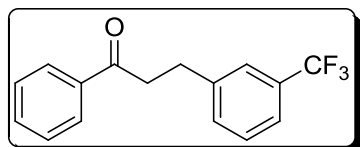


the general procedure **A** starting from **1f** (89.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane = 7:93) afforded **2f** in 43% yield (38.4 mg) as white solid. The NMR data is in accordance with the literature.^[14]

¹H NMR (500 MHz, CDCl₃): δ 8.01-7.99 (m, 2H), 7.55-7.52 (m, 1H), 7.46-7.43 (m, 2H), 6.14 (s, 2H), 3.82 (s, 3H), 3.78 (s, 6H), 3.12-3.09 (m, 2H), 3.02-2.99 (m, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 200.7, 159.6, 158.8, 137.0, 132.6, 128.3, 128.2, 109.8, 90.4, 55.5, 55.3, 38.7, 18.3.

1-phenyl-3-(3-(trifluoromethyl)phenyl)propan-1-one (2g): Compound **2g** was prepared according



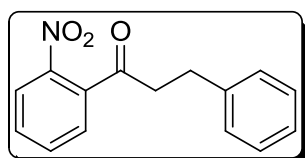
to the general procedure **A** starting from **1g** (82.8 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2g** in 56% yield (46.9 mg) as yellow oil. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 7.97-7.95 (m, 2H), 7.57 (t, *J* = 7.5 Hz, 1H), 7.52 (s, 1H), 7.48-7.39 (m, 5H), 3.33 (t, *J* = 7.5 Hz, 2H), 3.14 (t, *J* = 7.5 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.5, 142.2, 136.7, 133.2, 131.9, 131.2, 130.9, 130.7, 130.4, 128.9, 128.6, 127.9, 125.3, 125.2, 125.1, 125.1, 123.0, 123.0, 39.9, 29.8.

¹⁹F NMR (377 MHz, CDCl₃): δ -62.46.

1-(2-nitrophenyl)-3-phenylpropan-1-one (2h): Compound **2h** was prepared according to the general



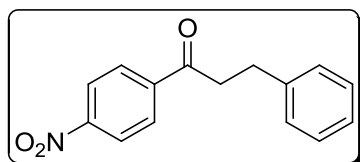
procedure **A** starting from **1h** (75.9 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 5:95) afforded **2h** in 65% yield (49.8 mg) as light brownish solid.

¹H NMR (500 MHz, CDCl₃): δ 7.96-7.93 (m, 3H), 7.56-7.52 (m, 2H), 7.47-7.43 (m, 3H), 7.38-7.35 (m, 1H), 3.41-3.38 (m, 2H), 3.33-3.30 (m, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.4, 149.2, 136.5, 136.4, 133.2, 132.5, 128.6, 127.9, 127.4, 124.8, 39.4, 27.7. (One carbon peak is overlapped)

ESI-MS calculated for C₁₅H₁₃NO₃ is [M+H]⁺ : 256.0974; found: 256.1115.

1-(4-nitrophenyl)-3-phenylpropan-1-one (2i): Compound **2i** was prepared according to the general

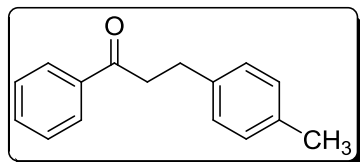


procedure **A** starting from **1i** (75.9 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2i** in 37% yield (28.2 mg) as white solid. The NMR data is in accordance with the literature.^[15]

¹H NMR (500 MHz, CDCl₃): δ 8.30-8.28 (m, 2H), 8.10-8.08 (m, 2H), 7.32-7.29 (m, 2H), 7.26-7.21 (m, 3H), 3.35 (t, *J* = 7.5 Hz, 2H), 3.10 (t, *J* = 7.6 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 197.6, 150.3, 141.2, 140.6, 129.0, 128.6, 128.4, 126.4, 123.8, 40.9, 29.8.

1-phenyl-3-(*p*-tolyl)propan-1-one (2j): Compound **2j** was prepared according to the general procedure

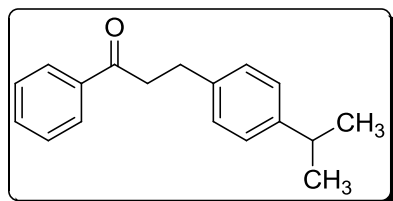


A starting from **1j** (66.6 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane=2:98) afforded **2j** in 72% yield (48.2 mg) as yellow oil. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 7.99-7.97 (m, 2H), 7.58-7.55 (m, 1H), 7.48-7.45 (m, 2H), 7.18-7.13 (m, 4H), 3.30 (t, *J* = 7.8 Hz, 2H), 3.06 (t, *J* = 7.9 Hz, 2H), 2.35 (s, 3H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.2, 138.1, 136.9, 135.5, 132.9, 129.2, 128.5, 128.2, 127.9, 40.5, 29.7, 20.9.

3-(4-isopropylphenyl)-1-phenylpropan-1-one (2k): Compound **2k** was prepared according to the

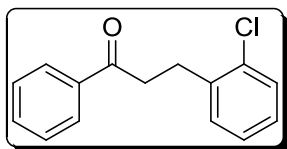


general procedure **A** starting from **1k** (75.03 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2k** in 69% yield (52 mg) as colorless oil. The NMR data is in accordance with the literature.^[16]

¹H NMR (500 MHz, CDCl₃): δ 7.99 (d, *J* = 8.0 Hz, 2H), 7.57 (t, *J* = 7.5 Hz, 1H), 7.47 (t, *J* = 7.4 Hz, 2H), 7.22 – 7.18 (m, 4H), 3.32 (t, *J* = 7.7 Hz, 2H), 3.07 (t, *J* = 7.7 Hz, 2H), 2.95-2.87 (m, 1H), 1.27 (d, *J* = 6.8 Hz, 6H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.3, 146.6, 138.5, 136.9, 132.9, 128.5, 128.3, 127.9, 126.5, 40.5, 33.7, 29.7, 24.0.

3-(2-chlorophenyl)-1-phenylpropan-1-one (2l): Compound **2l** was prepared according to the general



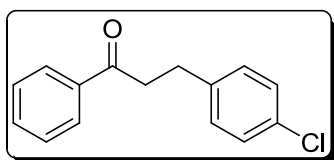
procedure **A** starting from **1l** (72.6 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane=2:98) afforded **2l** in 70% yield (51 mg) as white solid. The NMR data is in accordance with the literature.^[16]

¹H NMR (500 MHz, CDCl₃): δ 7.98-7.97 (m, 2H), 7.57-7.54 (m, 1H), 7.47-7.44 (m, 2H),

7.37-7.31 (m, 2H), 7.21-7.15 (m, 2H), 3.32 (t, *J* = 7.5 Hz, 2H), 3.19 (t, *J* = 7.3 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.9, 138.8, 136.7, 133.9, 133.1, 130.8, 129.5, 128.6, 128.0, 127.7, 126.9, 38.4, 28.3.

3-(4-chlorophenyl)-1-phenylpropan-1-one (2m): Compound **2m** was prepared according to the



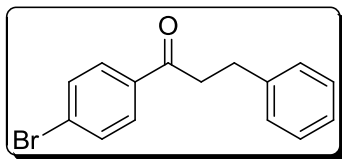
general procedure **A** starting from **1m** (72.6 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2m** in 75% yield (54.5 mg) as white solid. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 7.97-7.95 (m, 2H), 7.59-7.55 (m, 1H), 7.48-7.45 (m,

2H), 7.28-7.26 (m, 2H), 7.20-7.19 (m, 2H), 3.29 (t, *J* = 7.5 Hz, 2H), 3.06 (t, *J* = 7.4 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.7, 139.7, 136.7, 133.1, 131.8, 129.8, 128.6, 128.5, 127.9, 40.1, 29.3.

1-(4-bromophenyl)-3-phenylpropan-1-one (2n): Compound **2n** was prepared according to the



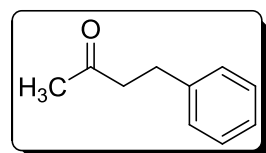
general procedure **A** starting from **1n** (85.7 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2n** in 72% yield (62.2 mg) as white solid. The NMR data is in accordance with the literature.^[12]

¹H NMR (500 MHz, CDCl₃): δ 7.83-7.81 (m, 2H), 7.60-7.58 (m, 2H), 7.32-7.20

(m, 5H), 3.27 (t, *J* = 7.4 Hz, 2H), 3.07 (t, *J* = 7.4 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.1, 140.9, 135.5, 131.9, 129.5, 128.5, 128.4, 128.2, 126.2, 40.3, 29.9.

4-phenylbutan-2-one (2o): Compound **2o** was prepared according to the general procedure **A** starting



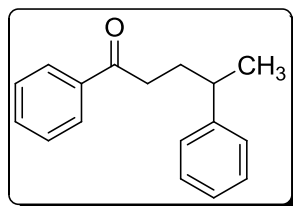
from **1o** (43.8 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 5:95) afforded **2o** in 47% yield (21 mg) as light brown liquid. The NMR data is in accordance with the literature.^[11]

¹H NMR (500 MHz, CDCl₃): δ 7.29-7.25 (m, 2H), 7.19-7.16 (3H), 2.88 (t, *J* = 7.8 Hz, 2H),

2.75 (t, *J* = 7.9 Hz, 2H), 2.12 (s, 3H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 207.9, 140.9, 128.5, 128.3, 126.1, 45.1, 30.0, 29.7.

1,4-diphenylpentan-1-one (2p): Compound **2p** was prepared according to the general procedure **A**



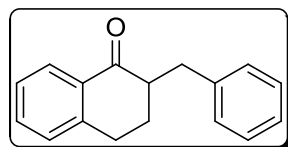
starting from **1p** (70.8 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2p** in 30% yield (21.7 mg) as pale yellow oil. The NMR data is in accordance with the literature.^[17]

¹H NMR (500 MHz, CDCl₃): δ 7.87-7.85 (m, 2H), 7.54-7.51 (m, 1H), 7.43-7.40 (m, 2H), 7.33-7.30 (m, 2H), 7.23-7.21 (m, 3H), 2.92-2.78 (m, 3H), 2.14-1.96 (m, 2H), 1.33

(d, *J* = 6.9 Hz, 3H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 200.3, 146.5, 136.9, 132.8, 128.5 (overlapped), 127.9, 127.0, 126.2, 39.5, 36.7, 32.4, 22.6.

2-benzyl-3,4-dihydronaphthalen-1(2H)-one (2q): Compound **2q** was prepared according to the

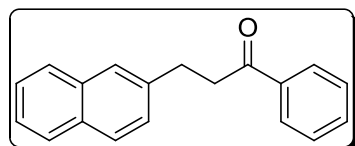


general procedure **A** starting from **1q** (70.2 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2q** in 99% yield (70 mg) as white solid. The NMR data is in accordance with the literature.^[13]

¹H NMR (500 MHz, CDCl₃): δ 8.14 (d, *J* = 7.9 Hz, 1H), 7.53-7.49 (m, 1H), 7.38-7.35 (m, 3H), 7.30-7.26 (m, 4H), 3.55 (dd, *J* = 13.7, 4.0 Hz, 1H), 3.03-2.92 (m, 2H), 2.83-2.77 (m, 1H), 2.73-2.69 (m, 1H), 2.19-2.14 (m, 1H), 1.88-1.77 (m, 1H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.2, 143.9, 139.9, 133.2, 132.4, 129.2, 128.6, 128.3, 127.5, 126.5, 126.1, 49.4, 35.6, 28.6, 27.6.

3-(naphthalen-2-yl)-1-phenylpropan-1-one (2r): Compound **2r** was prepared according to the

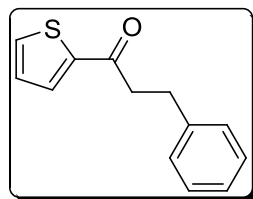


general procedure **A** starting from **1r** (51.6 mg, 0.2 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2r** in 62% yield (32 mg) as white solid. The NMR data is in accordance with the literature.^[12]

¹H NMR (500 MHz, CDCl₃): δ 8.00-7.98 (m, 2H), 7.83-7.80 (m, 3H), 7.71 (s, 1H), 7.58-7.55 (m, 1H), 7.48-7.40 (m, 5H), 3.40 (t, *J* = 7.7 Hz, 2H), 3.25 (t, *J* = 7.9 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.1, 138.8, 136.8, 133.6, 133.1, 132.1, 128.6, 128.1, 128.0, 127.6, 127.4, 127.1, 126.5, 125.9, 125.3, 40.3, 30.2.

3-phenyl-1-(thiophen-2-yl)propan-1-one (2s): Compound **2s** was prepared according to the general

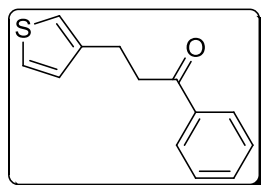


procedure **A** starting from **1s** (64.2 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2s** in 85% yield (54.8 mg) as yellow oil. The NMR data is in accordance with the literature.^[13]

^1H NMR (500 MHz, CDCl_3): δ 7.69 (dd, $J = 4.1, 1.1$ Hz, 1H), 7.62 (dd, $J = 4.8, 1.1$ Hz, 1H), 7.32-7.20 (m, 5H), 7.12-7.10 (m, 1H), 3.23 (t, $J = 7.5$ Hz, 2H), 3.08 (t, $J = 7.4$ Hz, 2H).

^{13}C NMR { ^1H } (125 MHz, CDCl_3): δ 192.1, 144.1, 140.9, 133.5, 131.8, 128.5, 128.4, 128.0, 126.1, 41.0, 30.3.

1-phenyl-3-(thiophen-3-yl)propan-1-one (2t): Compound **2t** was prepared according to the general

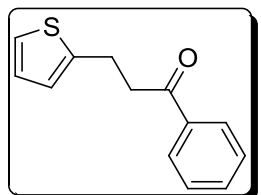


procedure **A** starting from **1t** (64.2, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2t** in 90% yield (58 mg) as colorless liquid. The NMR data is in accordance with the literature.^[18]

^1H NMR (500 MHz, CDCl_3): δ 7.98 (dd, $J = 8.1, 1.3$ Hz, 2H), 7.57 (t, $J = 7.5$ Hz, 1H), 7.47 (t, $J = 7.5$ Hz, 2H), 7.27-7.26 (m, 1H), 7.02-7.00 (m, 2H), 3.31 (t, $J = 7.6$ Hz, 2H), 3.11 (t, $J = 7.4$ Hz, 2H).

^{13}C NMR { ^1H } (125 MHz, CDCl_3): δ 199.1, 141.4, 136.8, 133.0, 128.5, 128.1, 127.9, 125.5, 120.5, 39.4, 24.5.

1-phenyl-3-(thiophen-2-yl)propan-1-one (2u): Compound **2u** was prepared according to the general

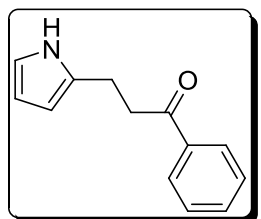


procedure **A** starting from **1u** (64.2 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2u** in 97% yield (62.5 mg) as colorless liquid. The NMR data is in accordance with the literature.^[13]

^1H NMR (500 MHz, CDCl_3): δ 7.98 (d, $J = 8.0$, 2H), 7.57 (t, $J = 7.3$ Hz, 1H), 7.47 (t, $J = 7.6$ Hz, 2H), 7.13 (dd, $J = 4.9, 0.9$ Hz, 1H), 6.94-6.92 (m, 1H), 6.88-6.87 (m, 1H), 3.39-3.36 (m, 2H), 3.32-3.29 (m, 2H).

^{13}C NMR { ^1H } (125 MHz, CDCl_3): δ 198.5, 143.8, 136.7, 133.1, 128.6, 127.9, 126.8, 124.6, 123.3, 40.5, 24.1.

1-phenyl-3-(1H-pyrrol-2-yl)propan-1-one (2v): Compound **2v** was prepared according to the general

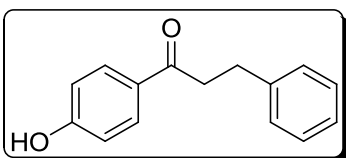


procedure **A** starting from **1v** (59.1 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 8:92) afforded **2v** in 25% yield (15 mg) as yellow solid. The NMR data is in accordance with the literature.^[19]

^1H NMR (500 MHz, CDCl_3): δ 8.61 (s, 1H), 7.98-7.96 (m, 2H), 7.57 (t, $J = 7.3$ Hz, 1H), 7.47 (t, $J = 7.6$ Hz, 2H), 6.68-6.67 (m, 1H), 6.11-6.09 (m, 1H), 5.96-5.95 (m, 1H), 3.35 (t, $J = 6.5$ Hz, 2H), 3.06 (t, $J = 6.5$ Hz, 2H).

^{13}C NMR { ^1H } (125 MHz, CDCl_3): δ 200.6, 136.7, 133.3, 131.7, 128.6, 127.9, 116.7, 107.8, 105.4, 39.4, 21.5.

1-(4-hydroxyphenyl)-3-phenylpropan-1-one (2w): Compound **2w** was prepared according to the

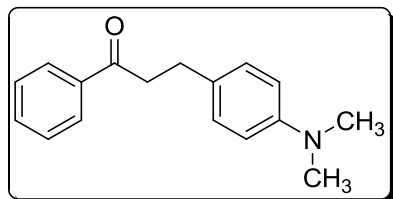


general procedure **A** starting from **1w** (67.2 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 11:89) afforded **2w** in 26% yield (17.4 mg) as white solid. The NMR data is in accordance with the literature.^[20]

^1H NMR (500 MHz, CDCl_3): δ 7.91-7.90 (m, 2H), 7.31-7.19 (m, 5H), 6.91-6.89 (m, 2H), 6.77 (br. s, 1H), 3.27 (t, $J = 7.5$ Hz, 2H), 3.06 (t, $J = 7.6$ Hz, 2H).

^{13}C NMR $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 198.9, 160.6, 141.2, 130.8, 129.6, 128.5, 128.4, 126.1, 115.5, 40.1, 30.4.

3-(4-(dimethylamino)phenyl)-1-phenylpropan-1-one (2x): Compound **2x** was prepared according

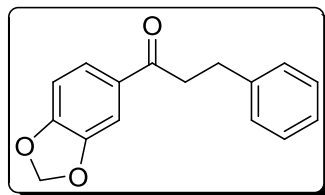


to the general procedure **A** starting from **1x** (75.3 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 8:92) afforded **2x** in 52% yield (39.5 mg) as pale oil liquid. The NMR data is in accordance with the literature.^[21]

^1H NMR (500 MHz, CDCl_3): δ 7.98 (d, J = 7.7 Hz, 2H), 7.56 (t, J = 7.3 Hz, 1H), 7.46 (t, J = 7.6 Hz, 2H), 7.15 (d, J = 8.6 Hz, 2H), 6.73 (d, J = 8.7 Hz, 2H), 3.28 (t, J = 7.9 Hz, 2H), 2.99 (t, J = 7.8 Hz, 2H), 2.93 (s, 6H).

^{13}C NMR $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 199.6, 149.2, 136.9, 132.9, 129.3, 128.9, 128.5, 128.0, 113.0, 40.9, 40.8, 29.2.

1-(benzo[d][1,3]dioxol-5-yl)-3-phenylpropan-1-one (2y): Compound **2y** was prepared according

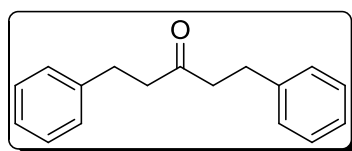


to the general procedure **A** starting from **1y** (75.6 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 3:97) afforded **2y** in 76% yield (58.1 mg) as white solid. The NMR data is in accordance with the literature.^[21]

^1H NMR (500 MHz, CDCl_3): δ 7.56 (dd, J = 8.1, 1.6 Hz, 1H), 7.45 (d, J = 1.7 Hz, 1H), 7.32 – 7.20 (m, 5H), 6.83 (d, J = 8.2 Hz, 1H), 6.02 (s, 2H), 3.22 (t, J = 7.9 Hz, 2H), 3.05 (t, J = 8.0 Hz, 2H).

^{13}C NMR $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 197.2, 151.6, 148.1, 141.3, 131.7, 128.4, 128.3, 126.0, 124.2, 107.8, 101.7, 40.1, 30.3. (one peak overlapped)

1,5-diphenylpentan-3-one (2z): Compound **2z** was prepared according to the general procedure **A**

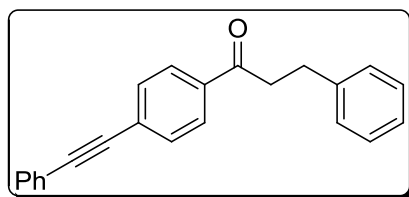


starting from **1z** (70.2 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 3:97) afforded **2z** in 66% yield (47.2 mg) as colorless liquid. The NMR data is in accordance with the literature.^[22]

^1H NMR (400 MHz, CDCl_3): δ 7.21-7.16 (m, 4H), 7.13-7.07 (m, 6H), 2.81 (t, J = 7.8 Hz, 4H), 2.63 (t, J = 7.6 Hz, 4H).

^{13}C NMR $\{^1\text{H}\}$ (100 MHz, CDCl_3): δ 209.0, 140.9, 128.4, 128.3, 126.0, 44.4, 29.7.

3-phenyl-1-(4-(phenylethynyl)phenyl)propan-1-one (2aa): Compound **2aa** was prepared

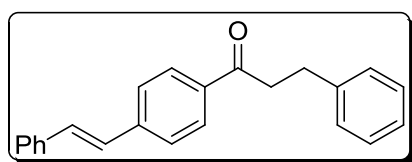


according to the general procedure **A** starting from **1aa** (92.4 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane = 2:98) afforded **2aa** in 93% yield (86.5 mg) as white shiny solid. The NMR data is in accordance with the literature.^[23]

¹H NMR (500 MHz, CDCl₃): δ 7.95 (d, *J* = 8.5 Hz, 2H), 7.63–7.57 (m, 4H), 7.39–7.23 (m, 8H), 3.31 (t, *J* = 7.8 Hz, 2H), 3.10 (t, *J* = 7.8 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.2, 141.1, 135.8, 131.7, 131.6, 128.7, 128.5, 128.4, 128.3, 128.0, 127.9, 126.1, 122.6, 92.7, 88.6, 40.4, 30.0.

(E)-3-phenyl-1-(4-styrylphenyl)propan-1-one (2ab): Compound **2ab** were prepared according to the

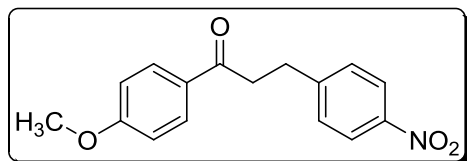


general procedure **A** starting from **1ab** (93.03 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2ab** in 44% yield (41.2 mg) as white solid. The NMR data is in accordance with the literature.^[11]

¹H NMR (500 MHz, CDCl₃): δ 7.96 (d, *J* = 8.3 Hz, 2H), 7.59–7.53 (m, 4H), 7.40–7.37 (m, 2H), 7.33–7.28 (m, 4H), 7.24–7.21 (m, 3H), 7.13 (d, *J* = 16.2 Hz, 1H), 3.31 (t, *J* = 8.0 Hz, 2H), 3.09 (t, *J* = 8.1 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 198.5, 141.9, 141.3, 136.7, 135.6, 131.4, 128.8, 128.6, 128.5, 128.4, 128.3, 127.4, 126.8, 126.5, 126.1, 40.4, 30.2.

1-(4-methoxyphenyl)-3-(4-nitrophenyl)propan-1-one (2ac): Compound **2ac** was prepared

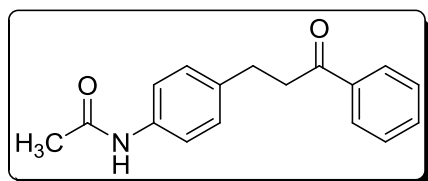


according to the general procedure **A** starting from **1ac** (84.9 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 12:88) afforded **2ac** in 81% yield (68.8 mg) as white solid. The NMR data is in accordance with the literature.^[24]

¹H NMR (500 MHz, CDCl₃): δ 8.10 (d, *J* = 8.6 Hz, 2H), 7.91 (d, *J* = 8.8 Hz, 2H), 7.39 (d, *J* = 8.6 Hz, 2H), 6.91 (d, *J* = 8.6 Hz, 2H), 3.84 (s, 3H), 3.28 (t, *J* = 7.2 Hz, 2H), 3.15 (t, *J* = 7.3 Hz, 2H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 196.6, 163.6, 149.4, 146.3, 130.2, 129.5, 129.2, 123.6, 113.7, 55.4, 38.9, 29.8.

N-(4-(3-oxo-3-phenylpropyl)phenyl)acetamide (2ad): Compound **2ad** was prepared according to

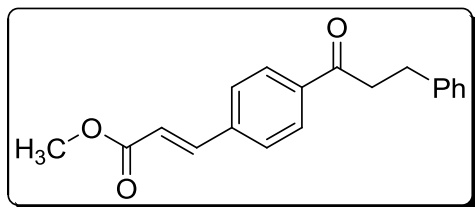


the general procedure **A** starting from **1ad** (79.5 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 30:70) afforded **2ad** in 56% yield (45.1 mg) as white solid. The NMR data is in accordance with the literature.^[25]

¹H NMR (500 MHz, CDCl₃): δ 7.94–7.93 (m, 3H), 7.56–7.53 (m, 1H), 7.45–7.42 (m, 4H), 7.16 (d, *J* = 8.3 Hz, 2H), 3.26 (t, *J* = 7.7 Hz, 2H), 3.01 (t, *J* = 7.7 Hz, 2H), 2.13 (s, 3H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 199.4, 168.6, 137.1, 136.7, 136.1, 133.1, 128.7, 128.6, 127.9, 120.3, 40.3, 29.4, 24.3.

methyl (*E*)-3-(4-cinnamoylphenyl)acrylate (2ae): Compound **2ae** was prepared according to the



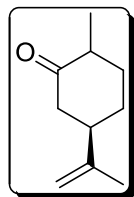
general procedure **A** starting from **1ae** (87.6 mg, 0.3 mmol) and purified by column chromatography (EtOAc:hexane = 9:91) afforded **2ae** in 45% yield (40 mg) as white solid.

$^1\text{H NMR}$ (500 MHz, CDCl_3): δ 7.96 (d, J = 8.5 Hz, 2H), 7.70 (d, J = 16.0 Hz, 1H), 7.59 (d, J = 8.3 Hz, 2H), 7.32-7.19 (m, 5H), 6.52 (d, J = 16.0 Hz, 1H), 3.82 (s, 3H), 3.29 (t, J = 7.8 Hz, 2H), 3.07 (t, J = 7.8 Hz, 2H).

$^{13}\text{C NMR}$ $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 198.4, 166.9, 143.3, 141.0, 138.6, 137.7, 128.5, 128.4, 128.1, 126.1, 120.2, 51.9, 40.5, 30.0. (one peak overlapped)

ESI-MS calculated for $\text{C}_{19}\text{H}_{18}\text{O}_3$ is $[\text{M}+\text{H}]^+$: 295.1334; found: 295.1323.

(5*R*)-2-methyl-5-(prop-1-en-2-yl)cyclohexanone (2af): Compound **2af** was prepared according to the general procedure **A** starting from **1af** (75.1 mg, 0.5 mmol) and purified by column chromatography (EtOAc:hexane = 2:98) afforded **2af** in 42% yield (32.1 mg) as colorless liquid.

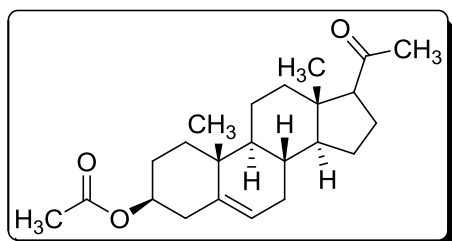


The NMR data is in accordance with the literature.^[26]

$^1\text{H NMR}$ (500 MHz, CDCl_3): δ 4.74 (m, 1H), 4.71 (s, 1H), 2.45-2.40 (m, 1H), 2.39-2.27 (m, 3H), 2.14-2.09 (m, 1H), 1.94-1.90 (m, 1H), 1.72 (s, 3H), 1.67-1.61 (m, 1H), 1.41-1.32 (m, 1H), 1.02 (d, J = 6.7 Hz, 3H).

$^{13}\text{C NMR}$ $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 212.7, 147.6, 109.6, 46.9, 46.8, 44.7, 34.9, 30.7, 20.5, 14.3.

(3*S*,8*S*,9*S*,10*R*,13*S*,14*S*)-17-acetyl-10,13-dimethyl-2,3,4,7,8,9,10,11,12,13,14,15,16,17-tetradecahydro-1*H*-cyclopenta[*a*]phenanthren-3-yl acetate (2ag): Compound **2ag** was prepared

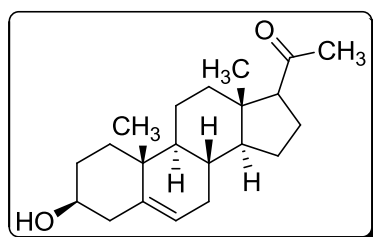


according to the general procedure **A** starting from **1ag** (107 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane=7:93) afforded **2ag** in 81% yield (87.1 mg) as white solid. The NMR data is in accordance with the literature.^[27]

$^1\text{H NMR}$ (500 MHz, CDCl_3): δ 5.33 (d, J = 4.0 Hz, 1H), 4.60-4.52 (m, 1H), 2.50 (t, J = 8.7 Hz, 1H), 2.29-2.27 (m, 2H), 2.17-2.15 (m, 1H), 2.08 (s, 3H), 2.02-1.94 (m, 1H), 1.99 (s, 3H), 1.84-1.82 (m, 2H), 1.68-1.38 (m, 9H), 1.23-1.07 (m, 4H), 0.98 (s, 3H), 0.59 (s, 3H).

$^{13}\text{C NMR}$ $\{^1\text{H}\}$ (125 MHz, CDCl_3): δ 209.3, 170.4, 139.6, 122.2, 73.7, 63.6, 56.7, 49.8, 43.9, 38.7, 37.9, 36.9, 36.5, 31.7, 31.7, 31.4, 27.6, 24.4, 22.7, 21.3, 20.9, 19.2, 13.1.

1-((3*S*,8*S*,9*S*,10*R*,13*S*,14*S*)-3-hydroxy-10,13-dimethyl-



2,3,4,7,8,9,10,11,12,13,14,15,16,17-tetradecahydro-1*H*-cyclopenta[*a*]phenanthren-17-yl)ethanone (**2ah**):

Compound **2ah** was prepared according to the general procedure **A** starting from **1ah** (94.3 mg, 0.3 mmol) and purified by column chromatography (EtOAc:Hexane=30:70) afforded **2ah** in 87% yield (82.8 mg) as white solid.

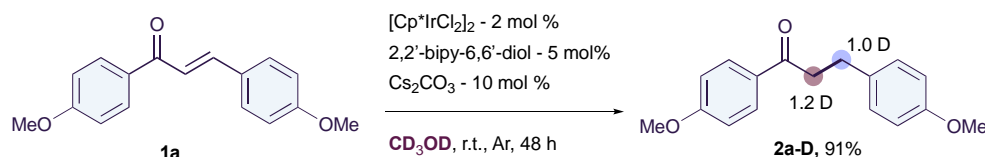
The NMR data is in accordance with the literature.^[28]

¹H NMR (500 MHz, CDCl₃): mixture of diastereomers: δ 5.32-5.30 (m, 1H), 3.52-3.48 (m, 1H), 2.50 (t, *J* = 8.9 Hz, 1H), 2.29-2.11 (m, 5H), 2.09 (s, 3H), 2.03-1.93 (m, 2H), 1.84-1.79 (m, 2H), 1.69-1.39 (m, 10H), 1.24-1.00 (m, 5H), 0.97 (s, 3H), 0.60 (s, 3H).

¹³C NMR {¹H} (125 MHz, CDCl₃): δ 209.6, 140.7, 121.2, 71.5, 63.6, 56.8, 49.9, 43.9, 42.1, 38.7, 37.2, 36.4, 31.8, 31.7, 31.5 (overlapped), 24.4, 22.7, 20.9, 19.3, 13.1.

5. Control Experiments:

5.1. H/D exchange experiment:



An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with [Cp*IrCl₂]₂ (4.78 mg, 0.006 mmol, 2 mol %), 2,2'-bipyridine-6,6'-diol (2.8 mg, 0.015 mmol, 5 mol %), Cs₂CO₃ (9.7 mg, 0.03 mmol, 10 mol %) and (*E*)-1,3-bis(4-methoxyphenyl)prop-2-en-1-one (0.3 mmol, 1.0 equiv.) followed by addition of 0.6 mL of CD₃OD (0.5 M) under argon atmosphere. The closed tube was stirred at room temperature for 48 h. After completion of the reaction, crude reaction mixture was purified by column chromatography using EtOAc:hexane (8:92) as mobile phase to obtain analytically pure deuterated saturated ketone **2a-D** in 91% yield (74.2 mg). ¹H NMR analysis revealed 120% deuterium incorporation at α position and 100% deuterium incorporation at β with respect to ketone moiety.

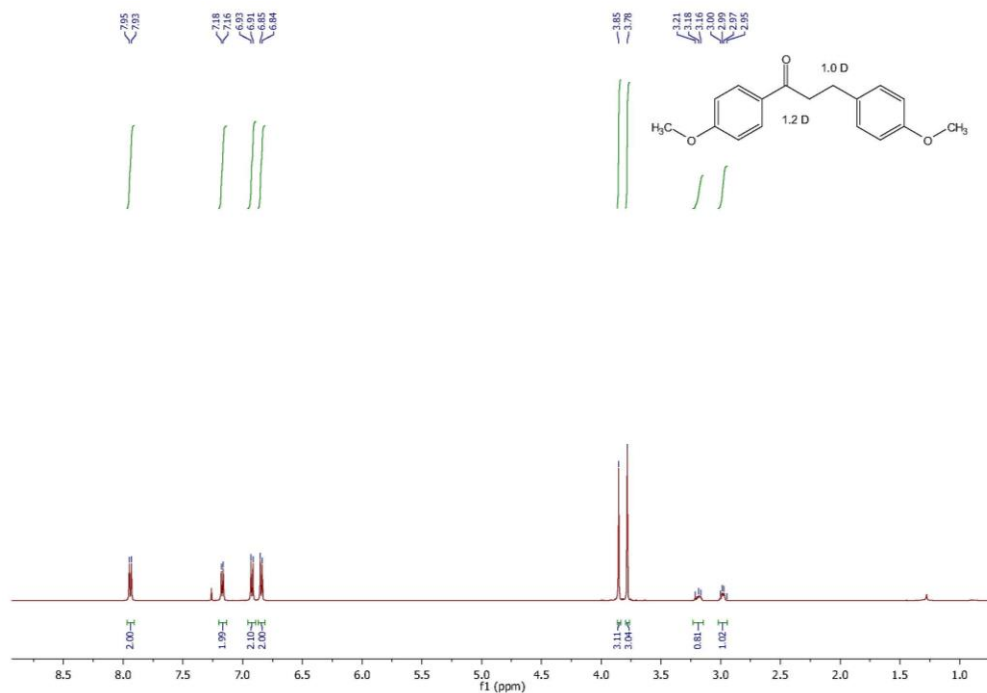


Figure S1: $^1\text{H-NMR}$ spectrum of isolated deuterated saturated ketone **2a-D**.

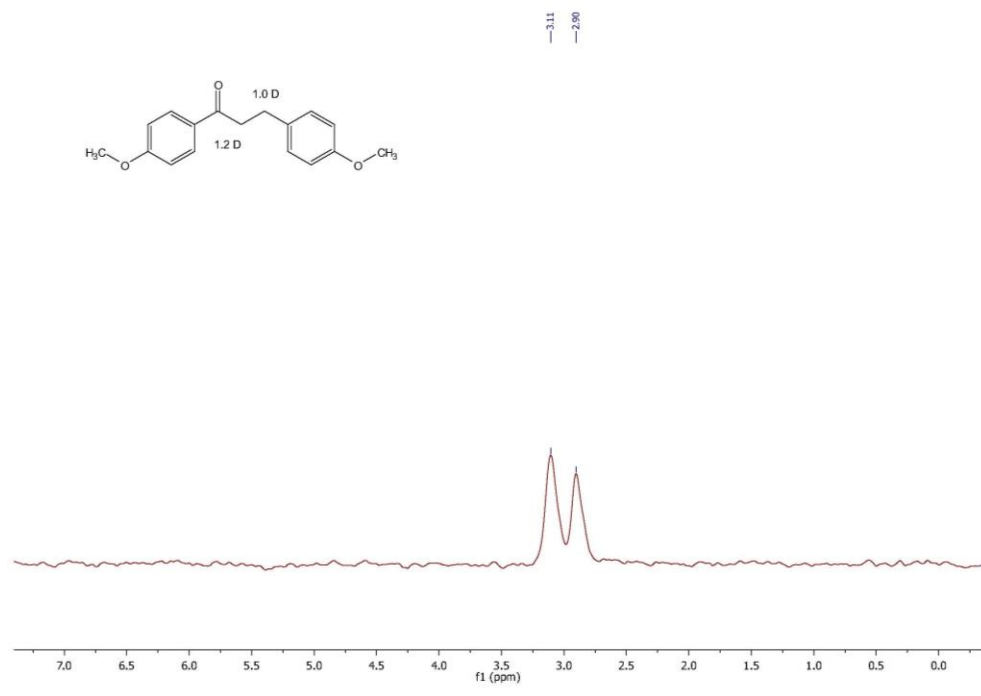


Figure S2: $^2\text{H-NMR}$ spectrum of isolated deuterated saturated ketone **2a-D**.

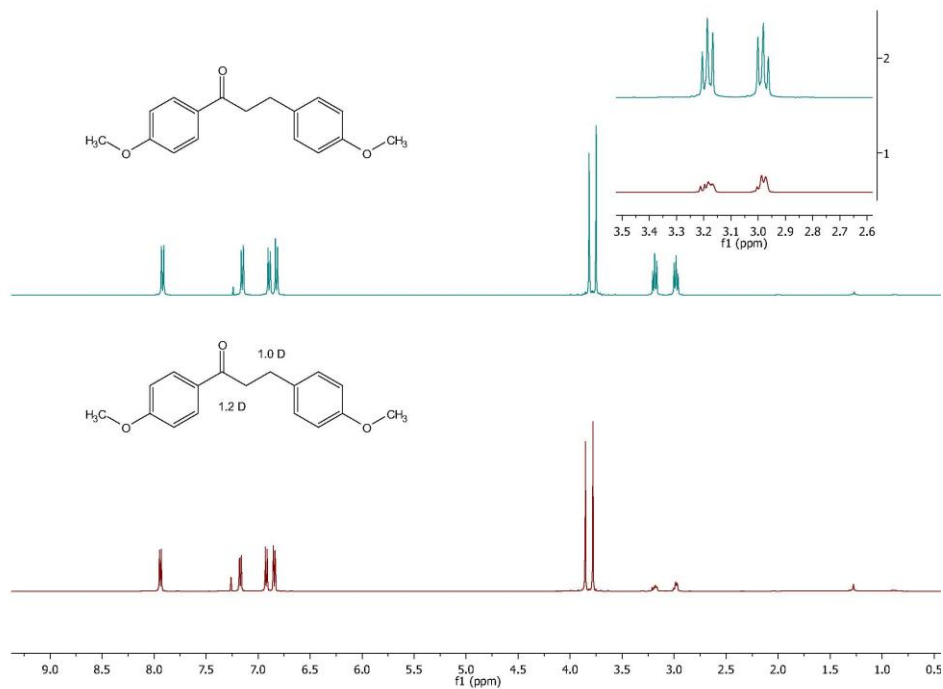


Figure S3: Stacked $^1\text{H-NMR}$ spectrums of **2a** and **2a-D** (enlarged splitting pattern).

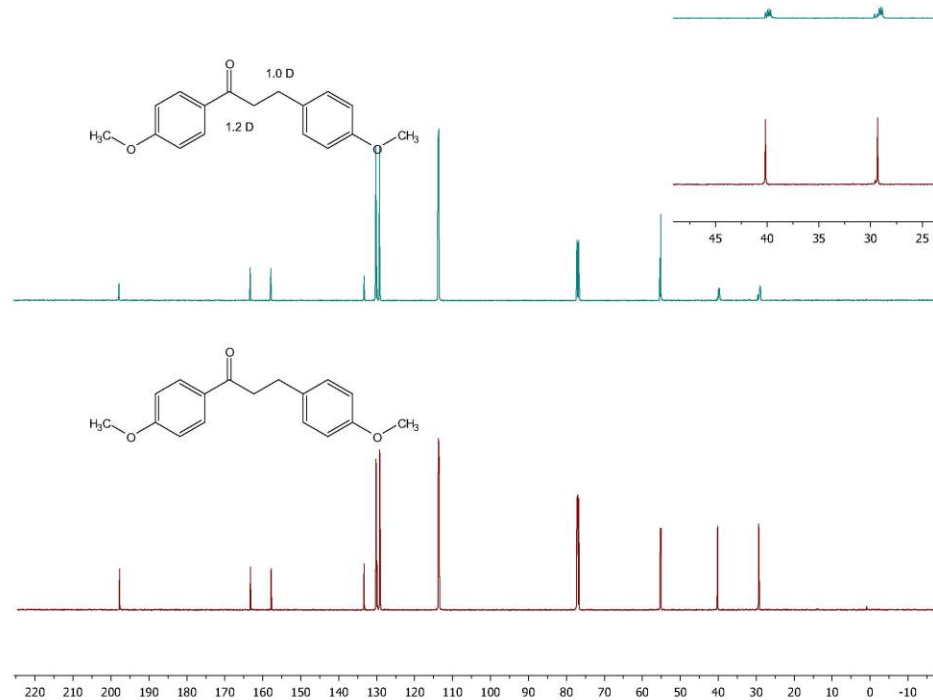


Figure S4: Stacked $^{13}\text{C-NMR}$ spectrums of **2a** and **2a-D** (enlarged splitting pattern).

5.2. Formation of Iridium-hydrido species: An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with $[\text{Cp}^*\text{IrCl}_2]_2$ (30 mg, 0.03 mmol, 1.0 equiv.), 2,2'-bipyridine-6,6'-diol (17.7 mg, 0.09 mmol, 2.5 equiv.) and NaOH (7.5 mg, 0.18 mmol, 5.0 equiv.) followed by addition of 0.6 mL of CH_3OH (0.05 M) under argon atmosphere. The closed tube was stirred at room temperature for 1 h. Solvent was evaporated under vacuum and the light brown residue was analyzed by ^1H -NMR in CD_3OD . ^1H -NMR spectrum is shown in Figure S6. The peak at -10.76 ppm strongly suggests the formation of iridium-hydrido species.

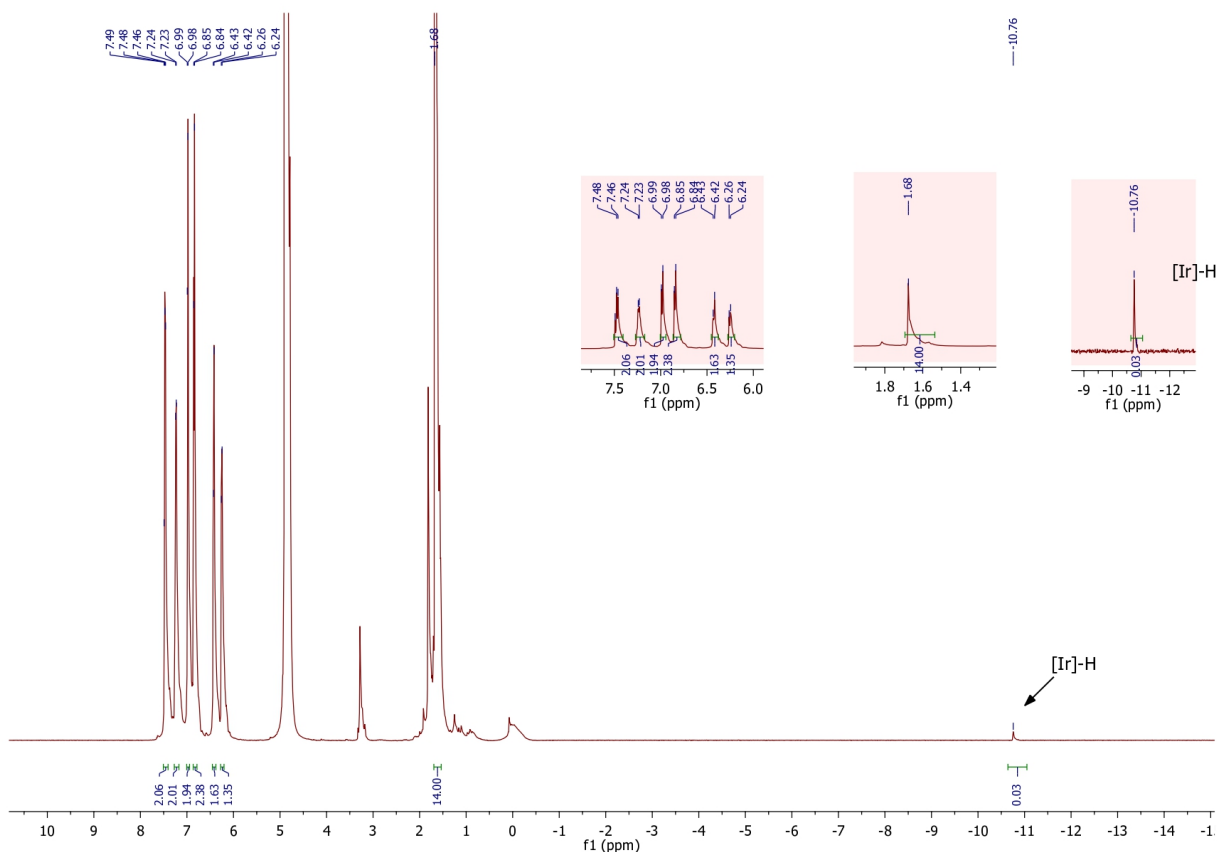
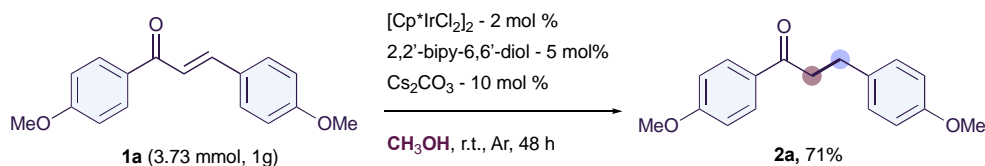


Figure S5: ^1H -NMR spectrum of iridium-hydrido species.

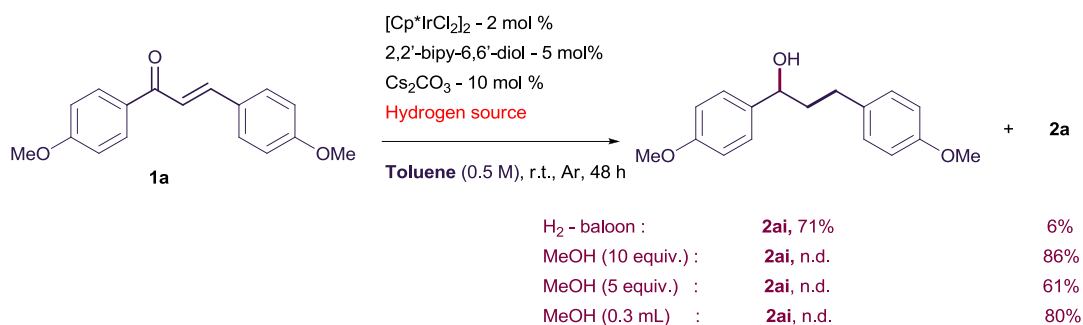
^1H NMR (400 MHz, CD_3OD): δ 7.49-7.46 (2H, pyridonate), 7.24-7.23 (2H, pyridonate), 6.99-6.98 (2H, pyridonate), 6.85-6.84 (2H, pyridonate), 6.43-6.42 (2H, pyridonate), 6.26-6.24 (2H, pyridonate), 1.68 (15H, Cp^*), -10.76 (Ir-H).

5.3. Gram scale synthesis:



An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with $[\text{Cp}^*\text{IrCl}_2]_2$ (60 mg, 0.07 mmol, 2 mol%), 2,2'-bipyridine-6,6'-diol (35 mg, 0.18 mmol, 5 mol%), Cs_2CO_3 (122 mg, 0.37 mmol, 10 mol%) and (*E*)-1,3-bis(4-methoxyphenyl)prop-2-en-1-one **1a** (1g, 3.72 mmol, 1.0 equiv.) followed by addition of 7.5 mL of methanol (0.5 M) under argon atmosphere. The sealed tube was stirred at room temperature for 48 h. After completion of the reaction, the crude reaction mixture was purified by column chromatography using EtOAc:Hexane (8:92) as mobile phase to obtain analytically pure saturated ketone **2a** in 71% yield (712 mg).

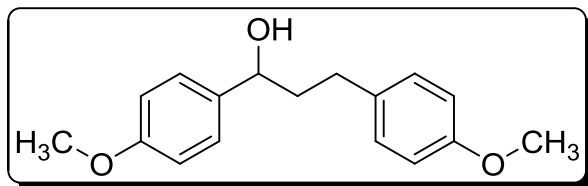
5.4. Hydrogenation using hydrogen balloon/methanol as LOHC:



H₂ – balloon experiment: An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with $[\text{Cp}^*\text{IrCl}_2]_2$ (4.78 mg, 0.006 mmol, 2 mol %), 2,2'-bipyridine-6,6'-diol (2.8 mg, 0.015 mmol, 5 mol %), Cs_2CO_3 (9.7 mg, 0.03 mmol, 10 mol %) and (*E*)-1,3-bis(4-methoxyphenyl)prop-2-en-1-one **1a** (0.3 mmol, 1.0 equiv.) followed by addition of 0.6 mL of toluene (0.5 M) under argon atmosphere. The resulting mixture was vacuumized quickly and purged with hydrogen gas (using direct "H₂" gas line). The closed tube was then connected to hydrogen balloon and stirred at room temperature for 48 h. After completion of the reaction, crude reaction mixture was purified by column chromatography using EtOAc/Hexane as mobile phase to obtain analytically pure **2a** (5 mg, 6% yield) and **2ai** (57.4 mg, 71% yield).

MeOH as hydrogen source in presence of toluene as solvent: An oven-dried Schlenk tube equipped with a magnetic stir bar was charged with $[\text{Cp}^*\text{IrCl}_2]_2$ (4.78 mg, 0.006 mmol, 2 mol %), 2,2'-bipyridine-6,6'-diol (2.8 mg, 0.015 mmol, 5 mol %), Cs_2CO_3 (9.7 mg, 0.03 mmol, 10 mol %) and (*E*)-1,3-bis(4-methoxyphenyl)prop-2-en-1-one (0.3 mmol, 1.0 equiv.) followed by addition of toluene (0.5 M) and 10 equiv./5 equiv./0.3 mL methanol under argon atmosphere. The closed tube was stirred at room temperature for 48 h. After completion of the reaction, crude reaction mixture was purified by column chromatography using EtOAc:hexane (8:92) as mobile phase to obtain analytically pure saturated ketone **2a** in good yields.

The NMR data of **2ai** is in accordance with the literature.^[29]



1,3-bis(4-methoxyphenyl)propan-1-ol (2ai):

¹H NMR (500 MHz, CDCl₃): δ 7.26-7.25 (m, 2H), 7.10-7.08 (m, 2H), 6.88-6.86 (m, 2H), 6.83-6.81 (m, 2H), 4.61-4.58 (m, 1H), 3.79 (s, 3H), 3.77 (s, 3H), 2.68-2.54

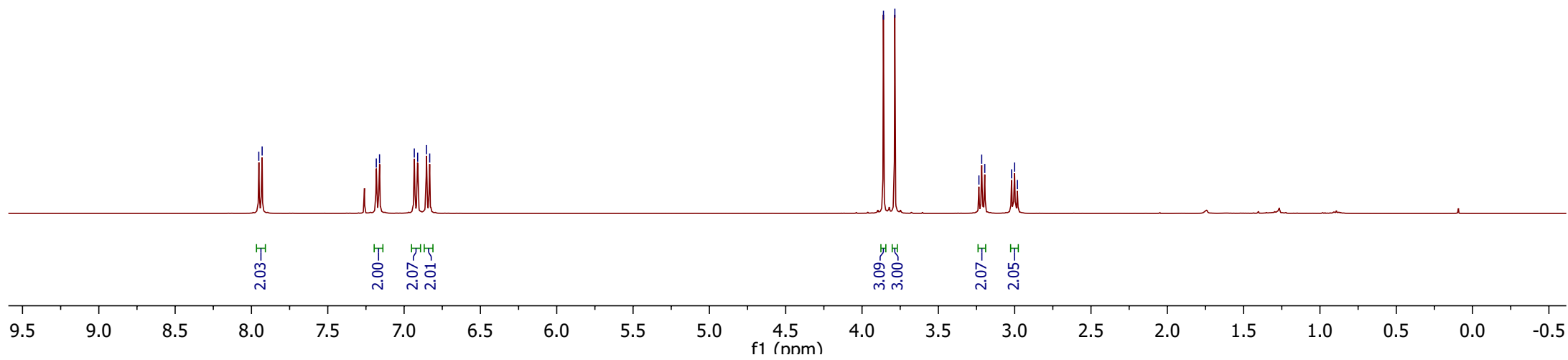
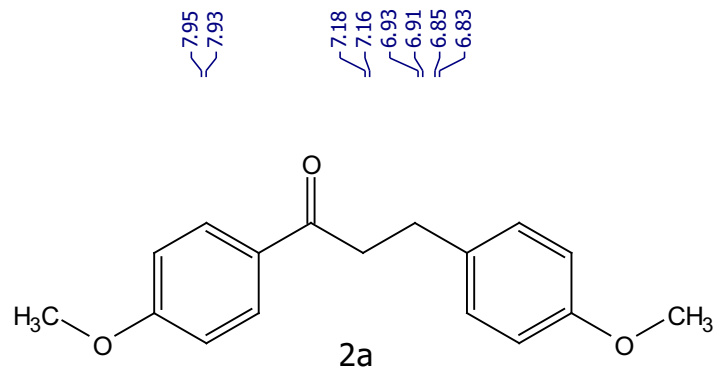
(m, 2H), 2.12-2.05 (m, 2H), 1.99-1.92 (m, 1H).

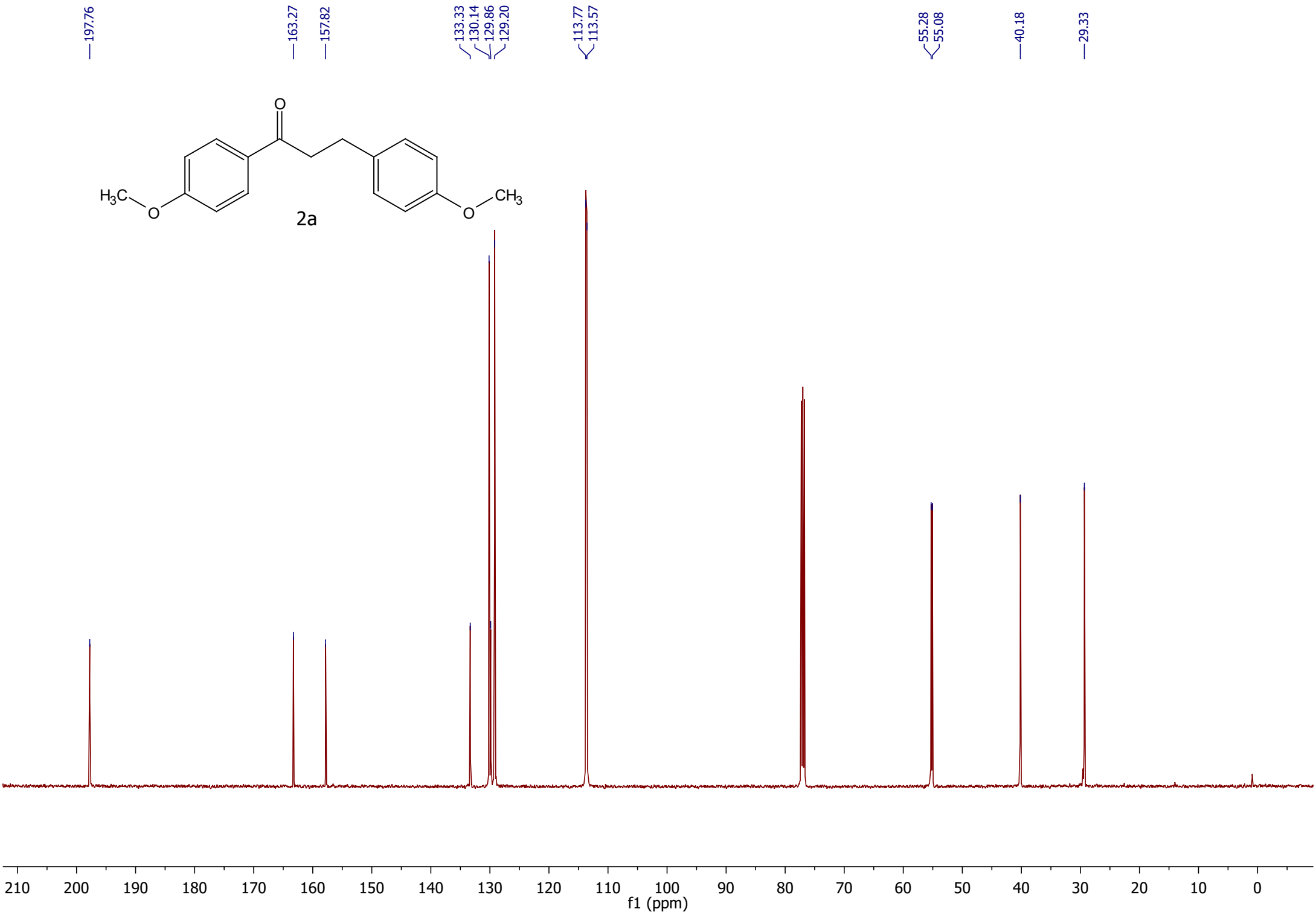
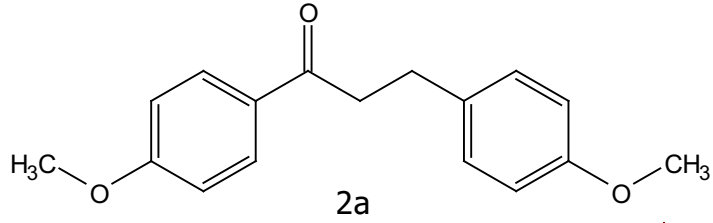
¹³C NMR {¹H} (125 MHz, CDCl₃): δ 158.9, 157.7, 136.7, 133.8, 129.2, 127.1, 113.8, 113.7, 73.3, 55.2, 55.2, 40.5, 31.1.

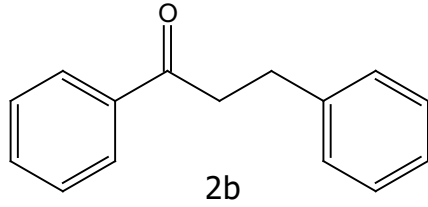
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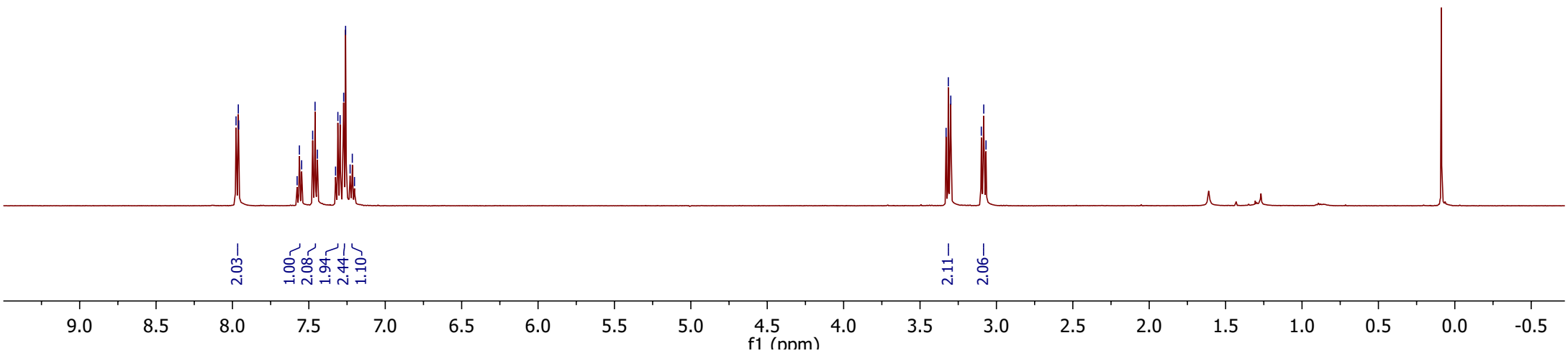


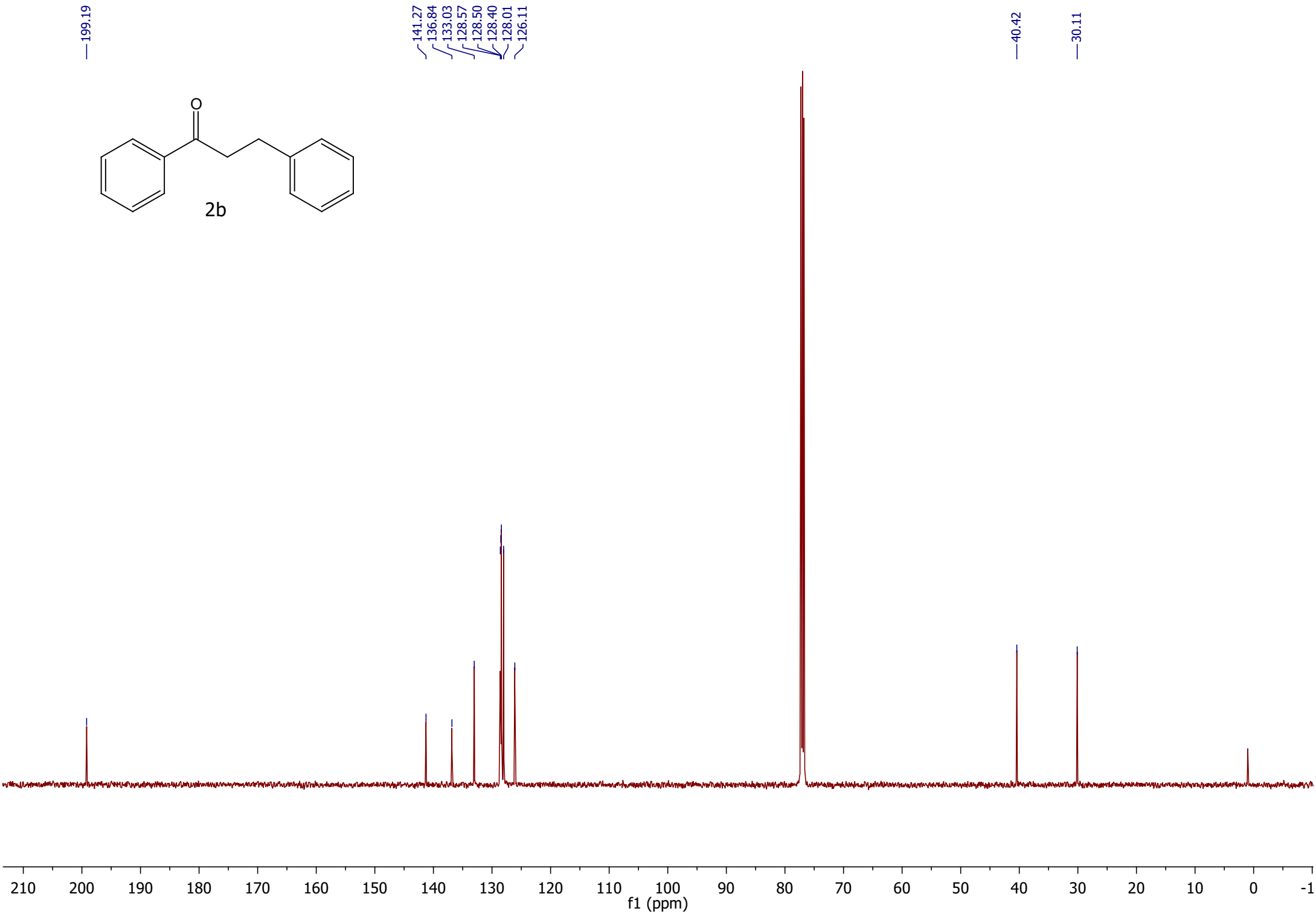
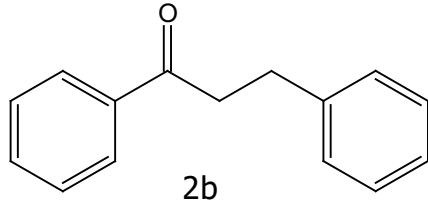


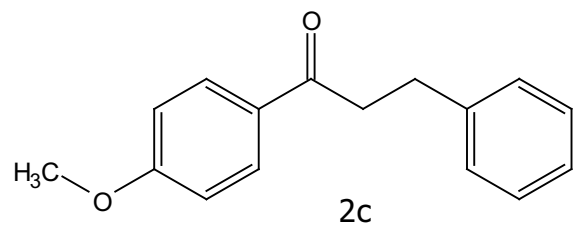


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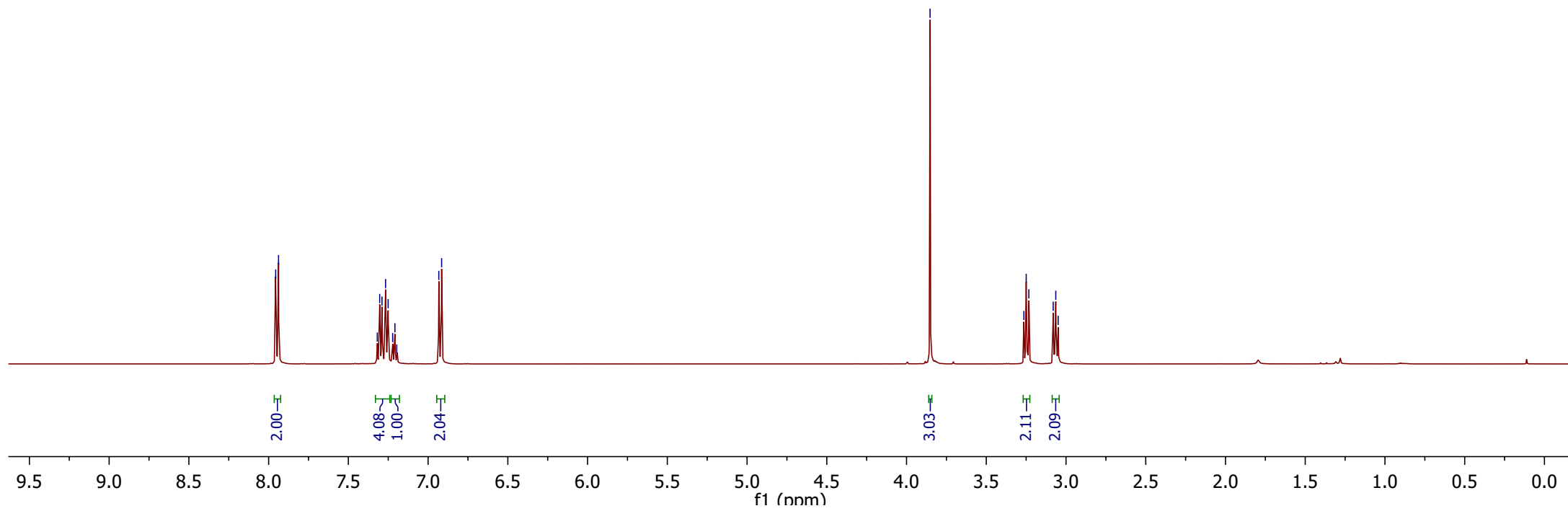


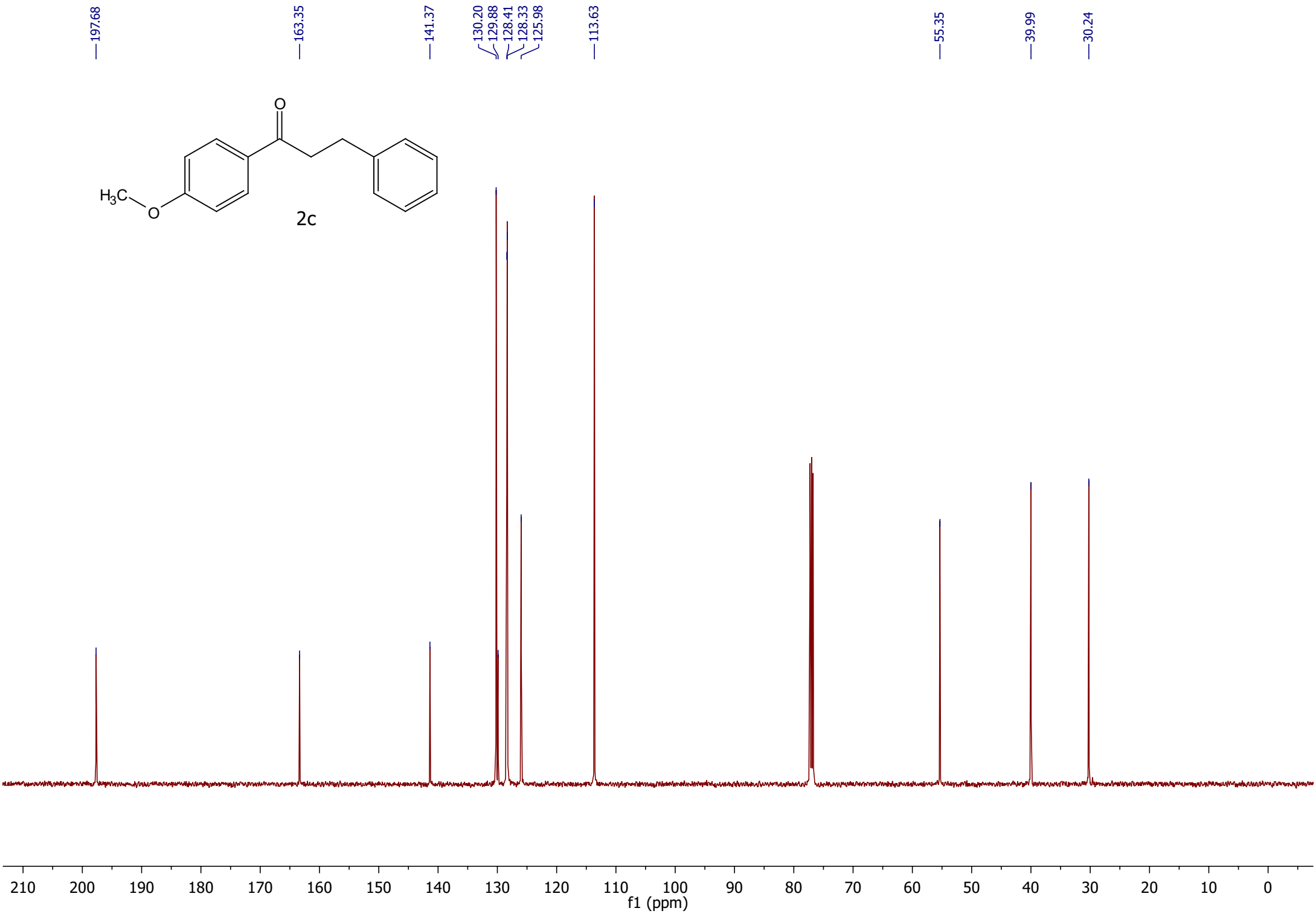
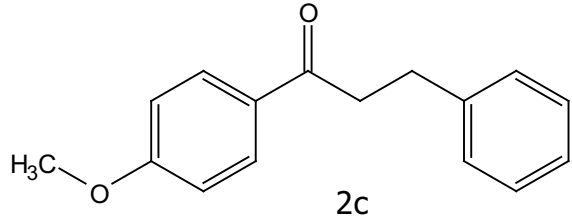




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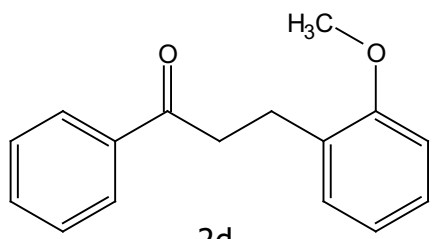




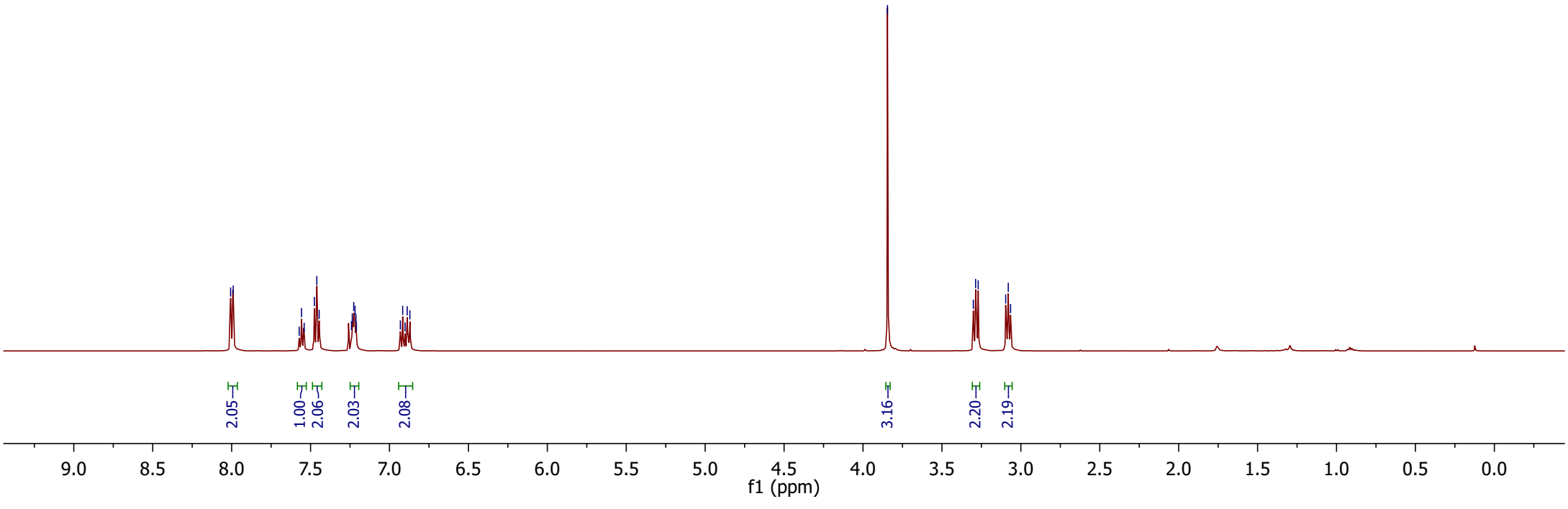
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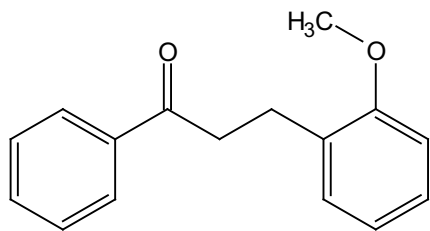
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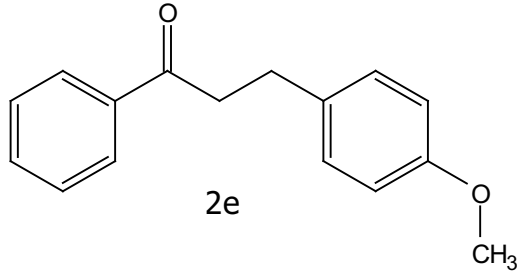
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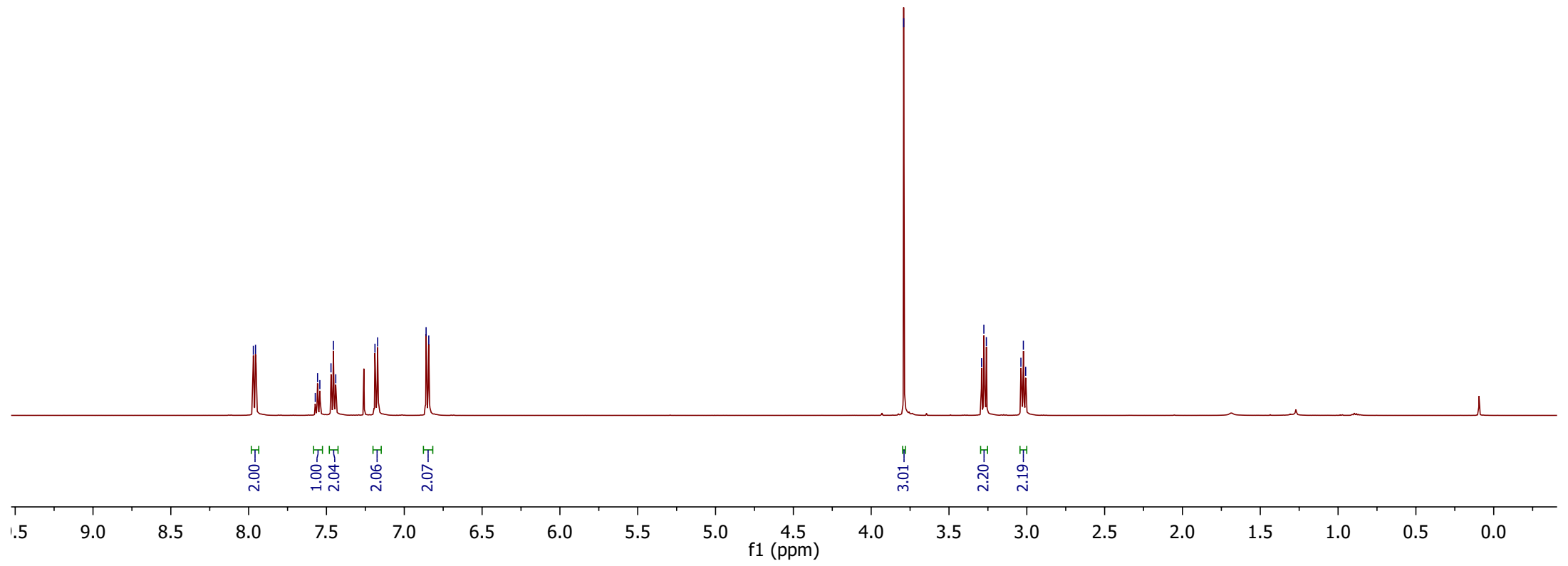
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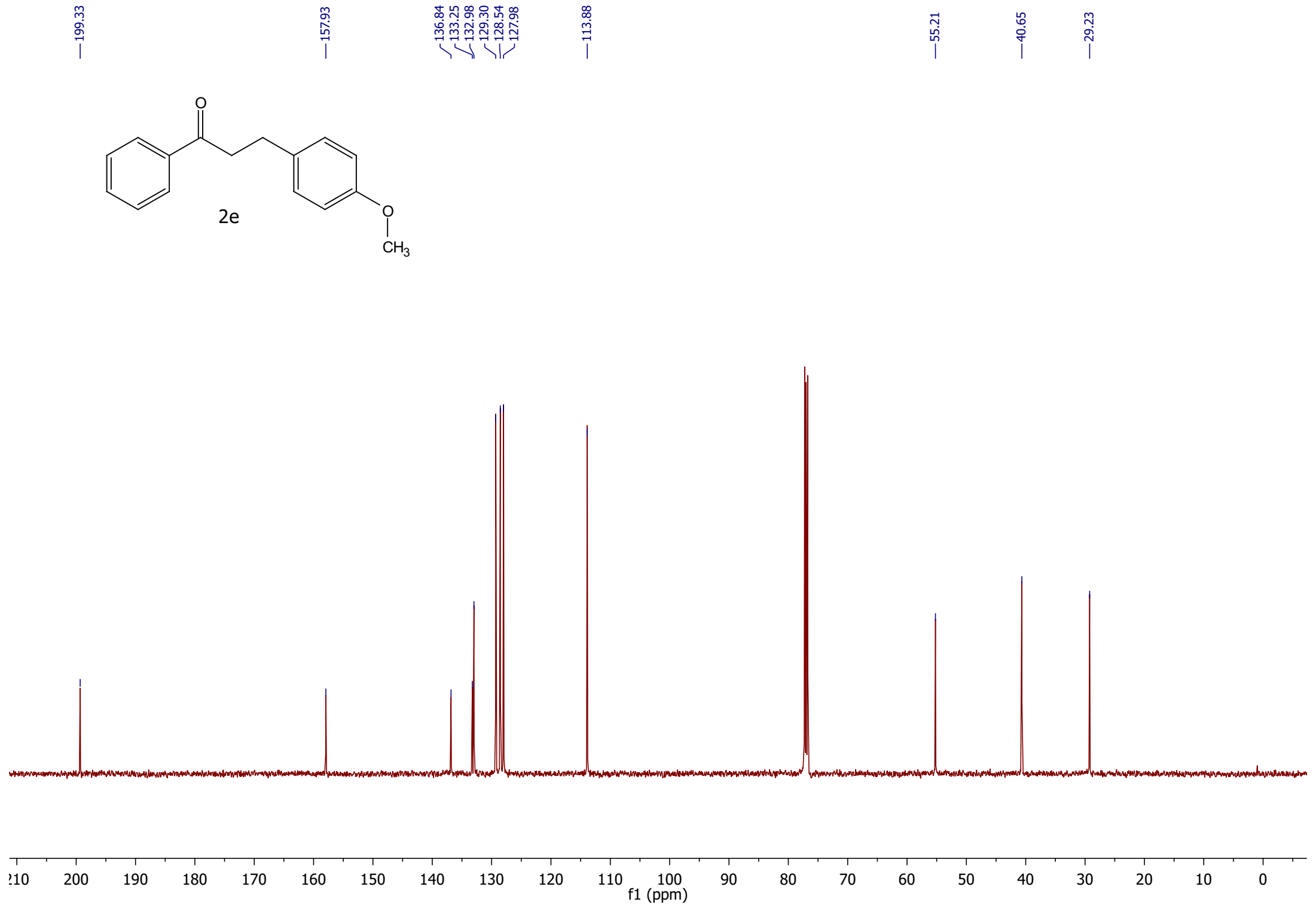
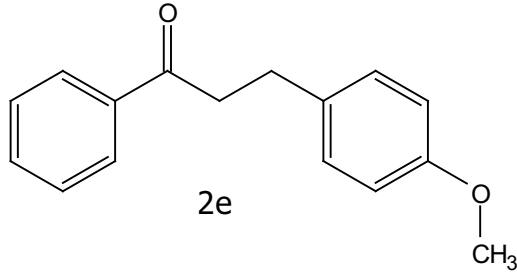
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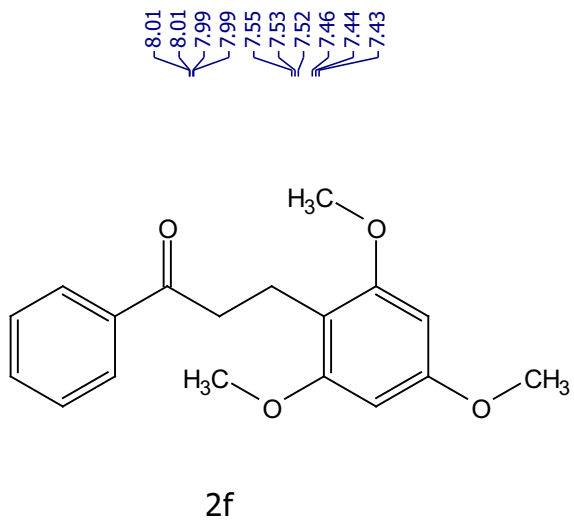


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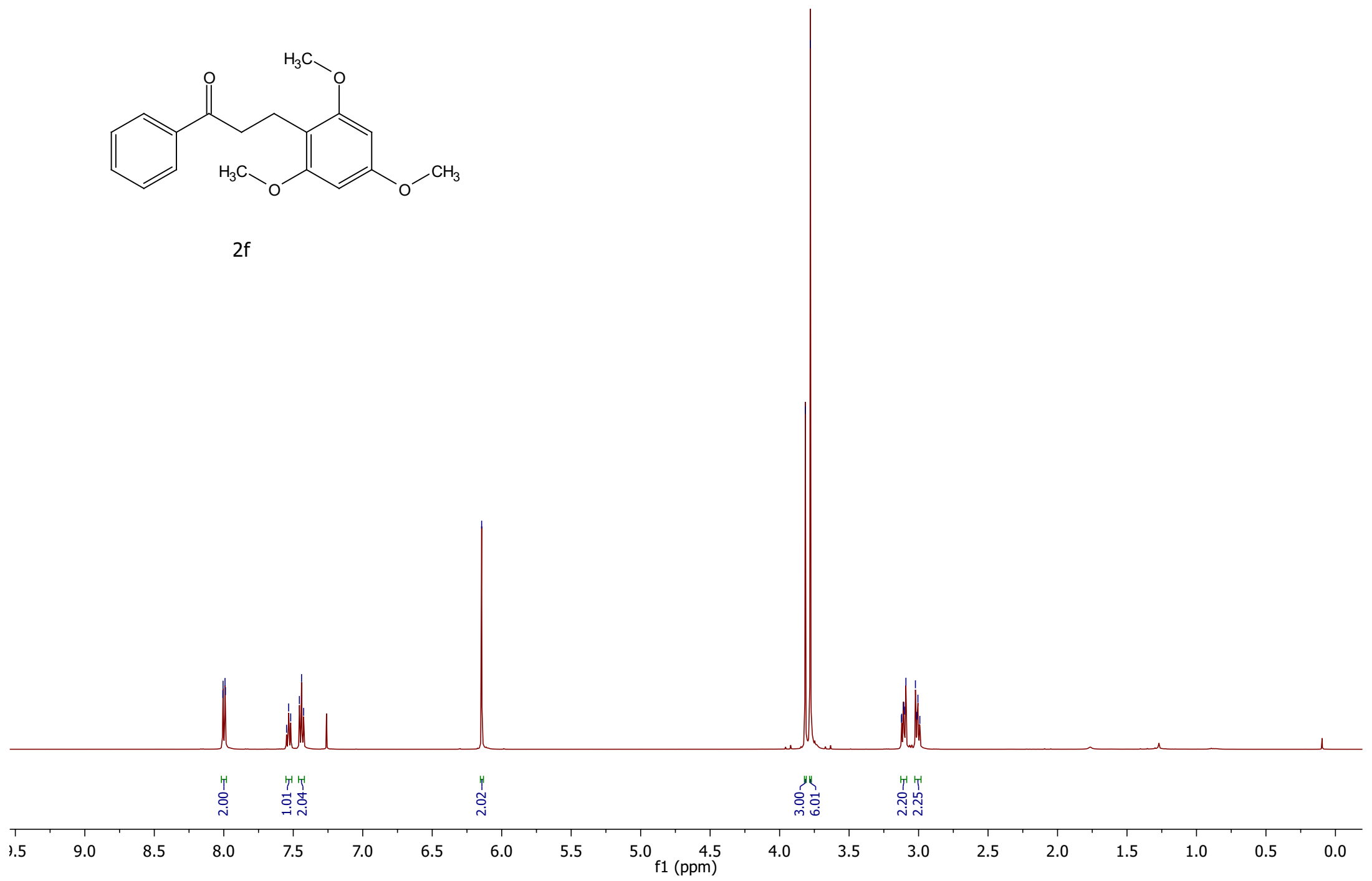




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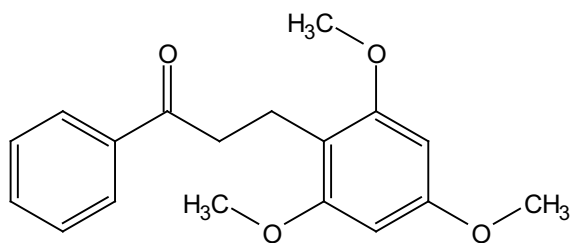
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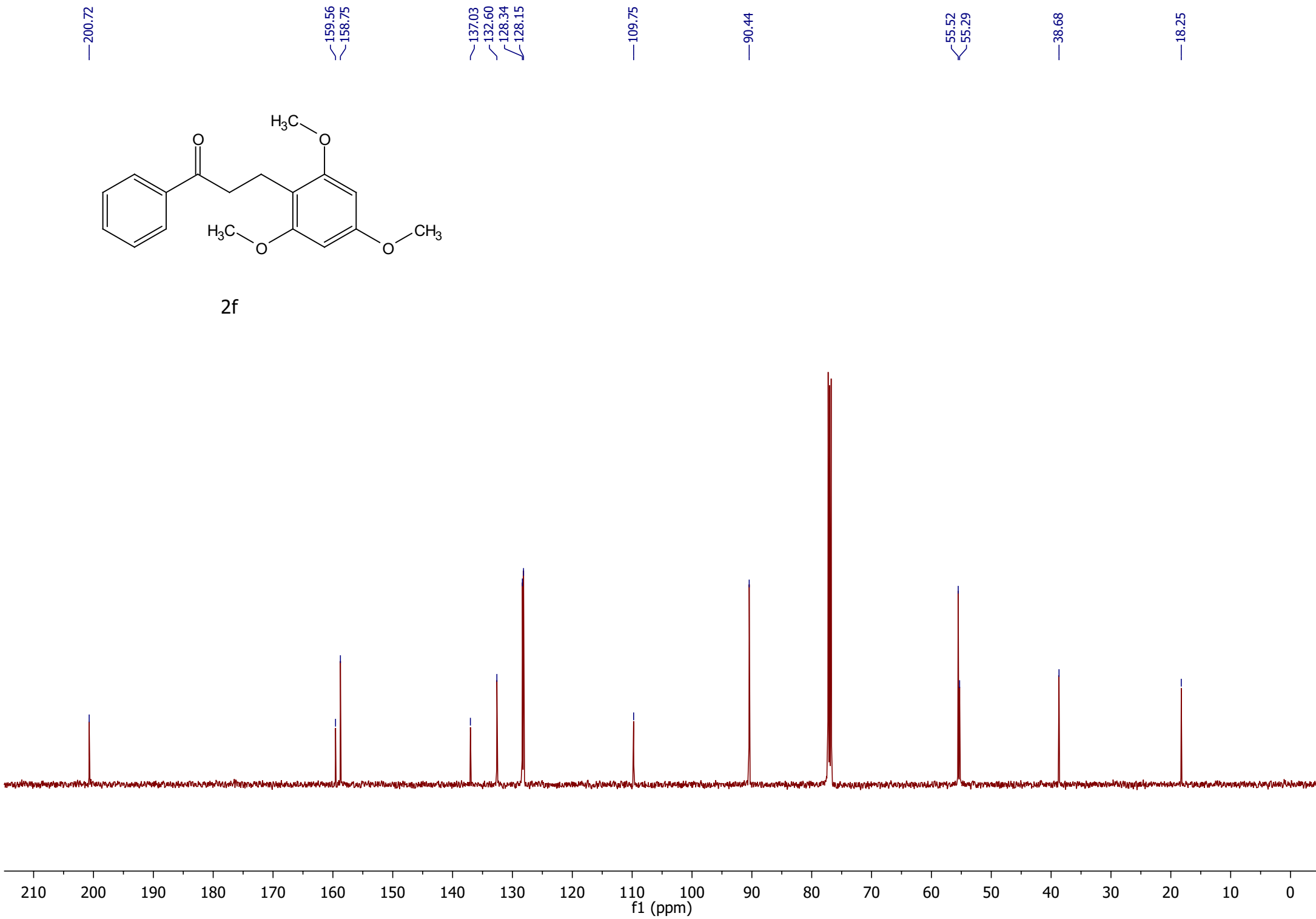
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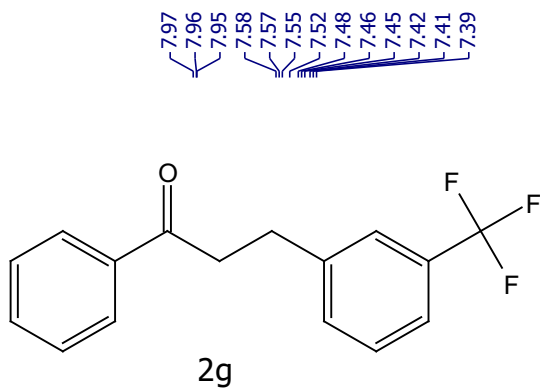
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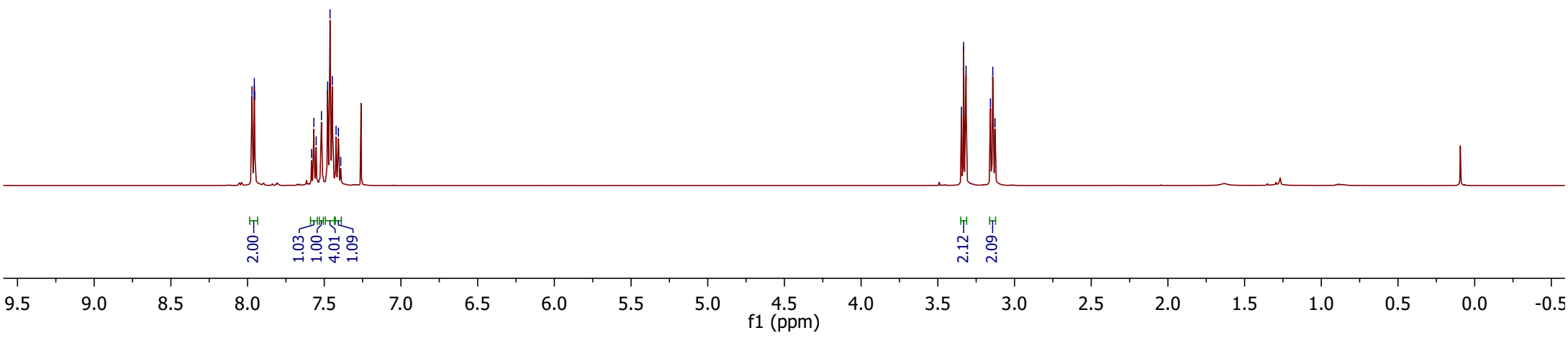
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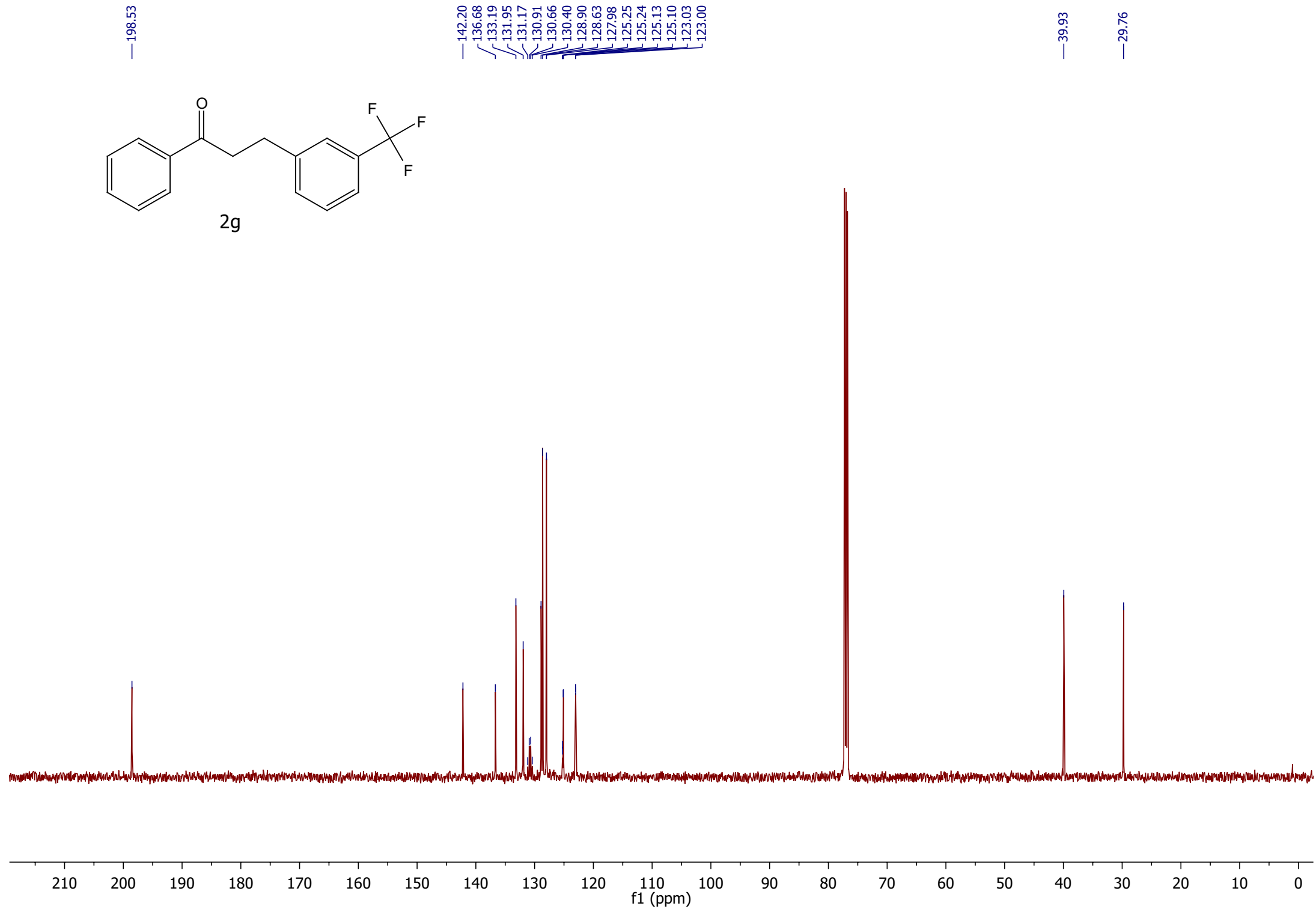
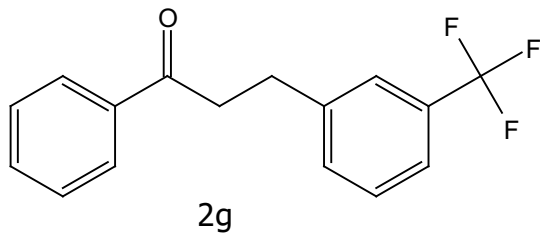


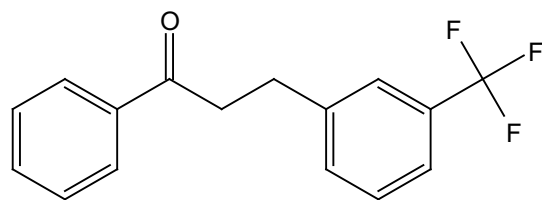


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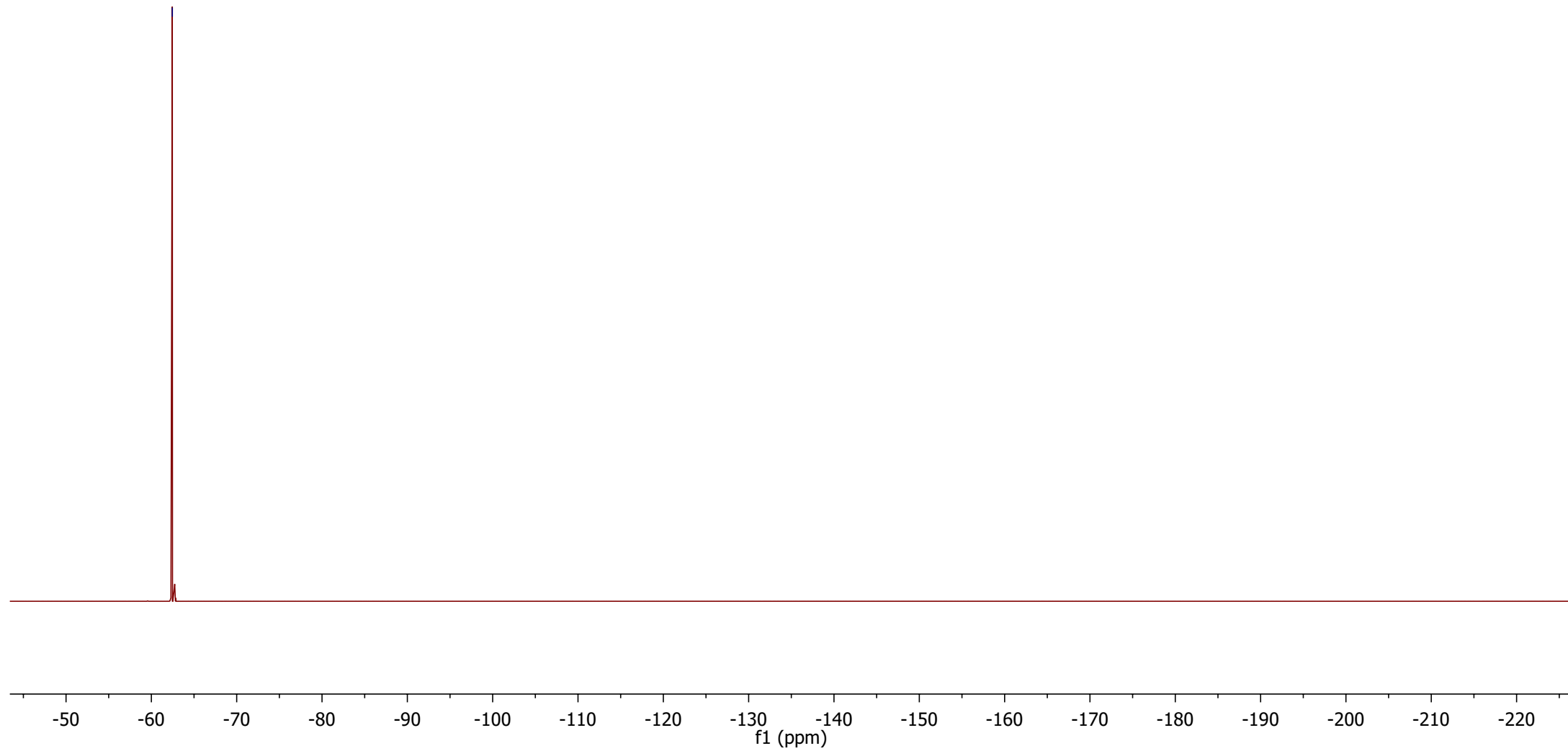


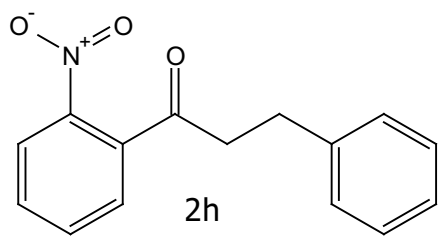




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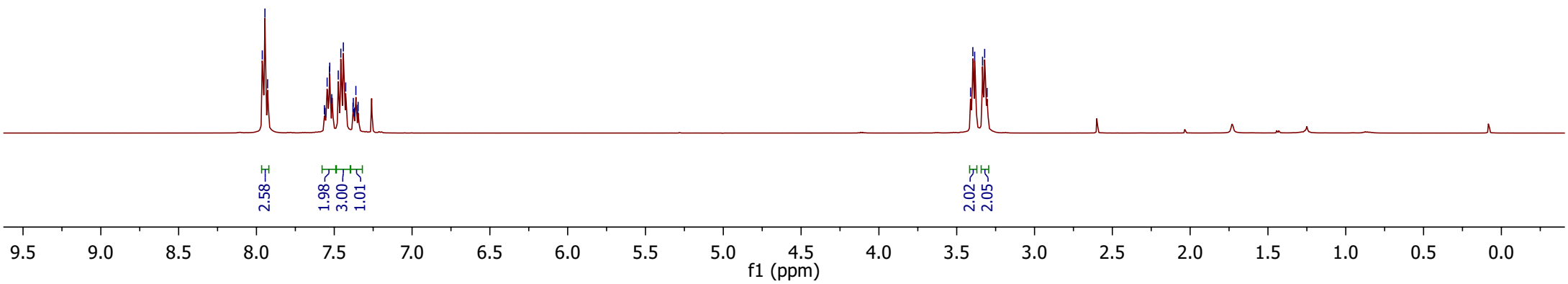
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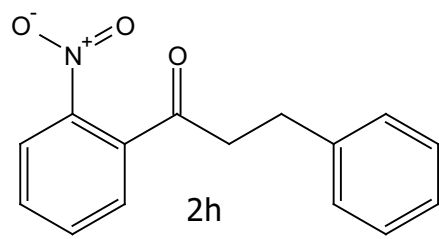




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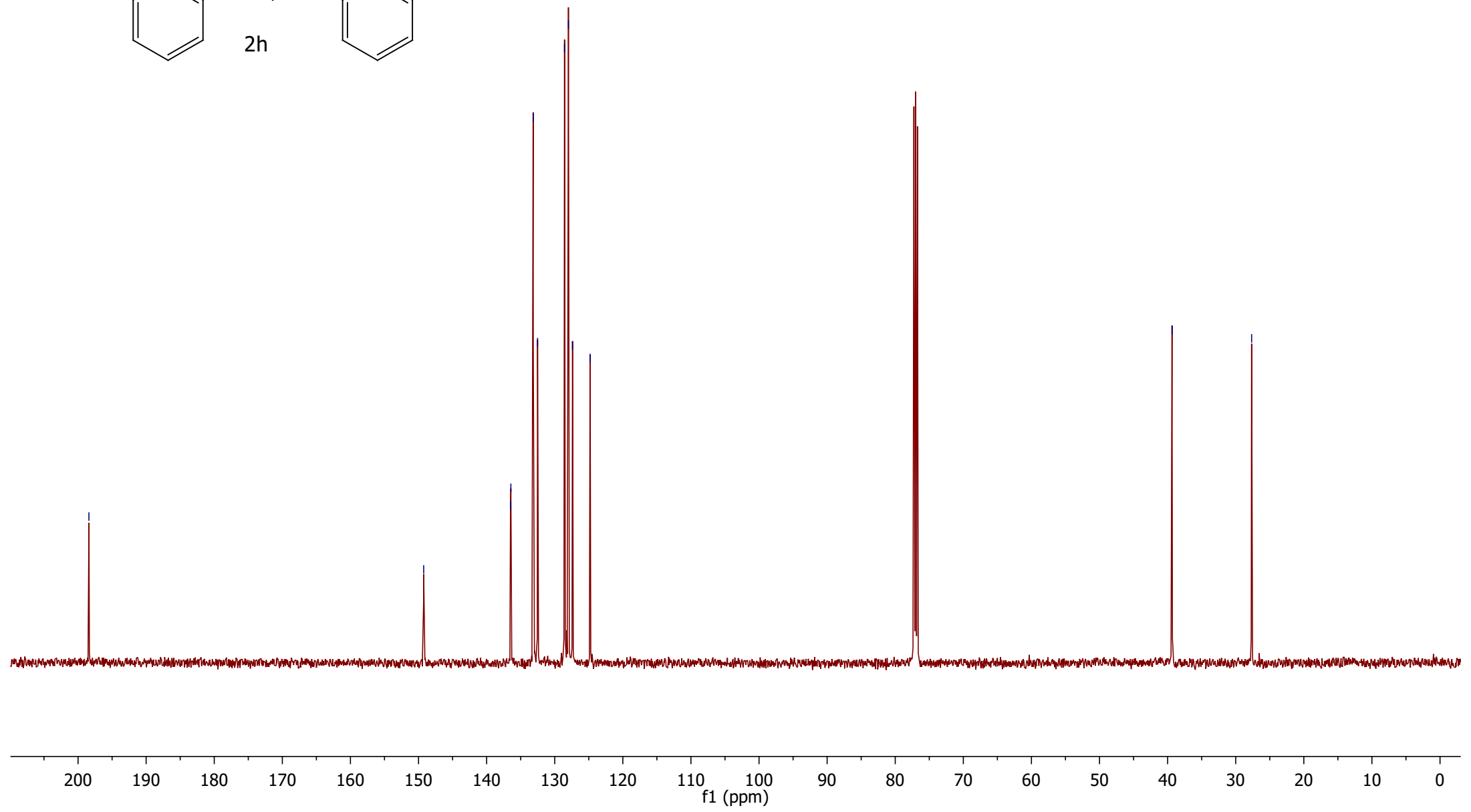
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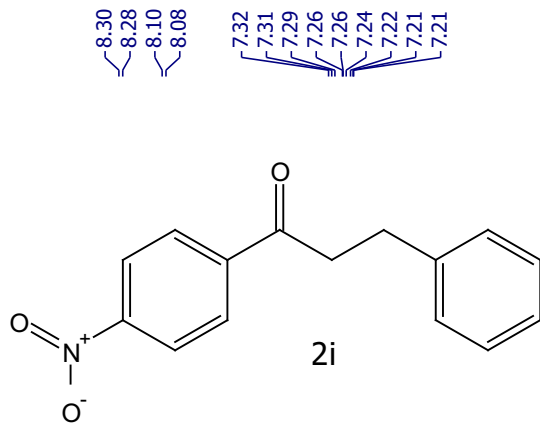
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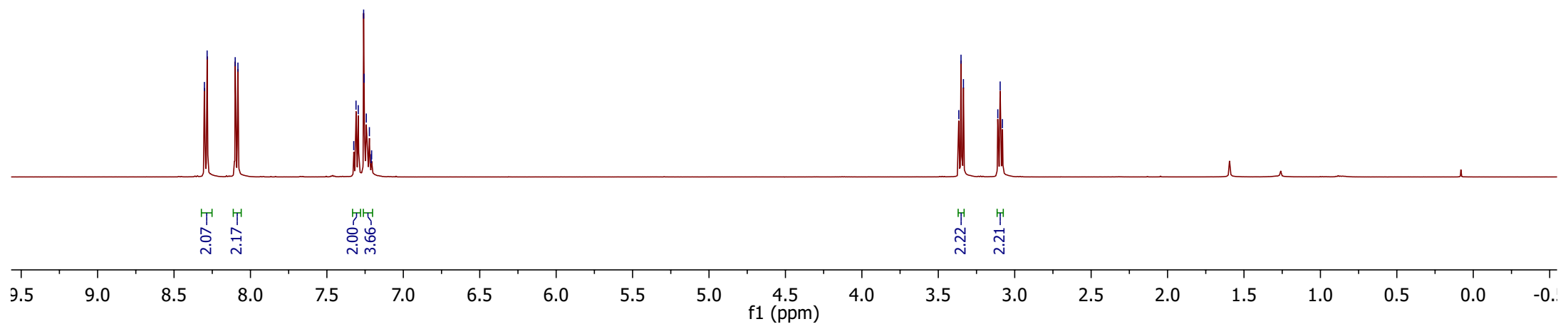
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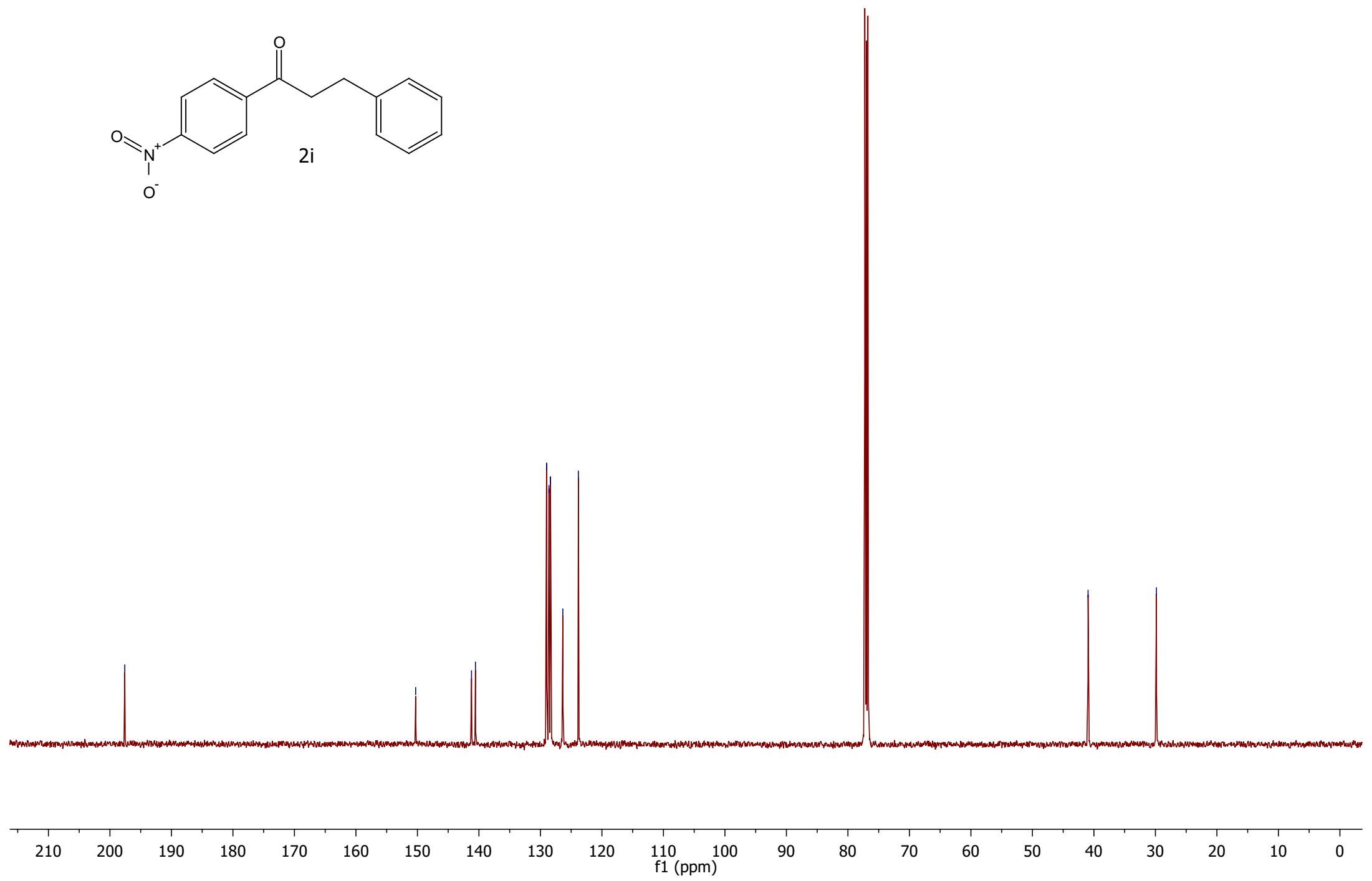
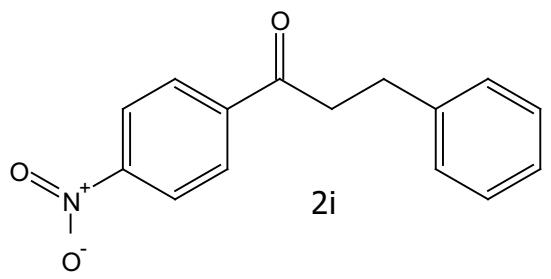
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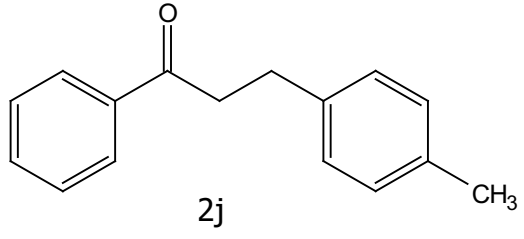
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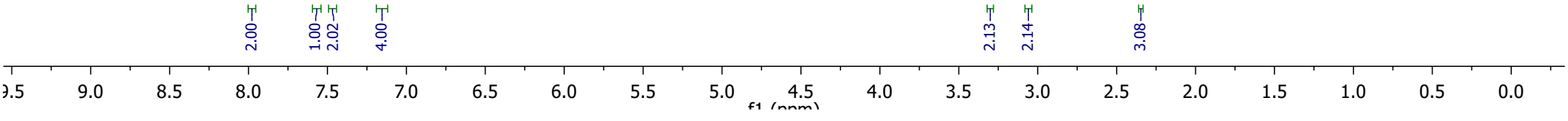


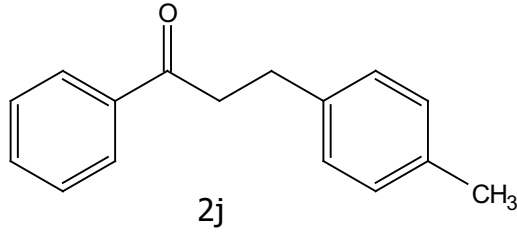


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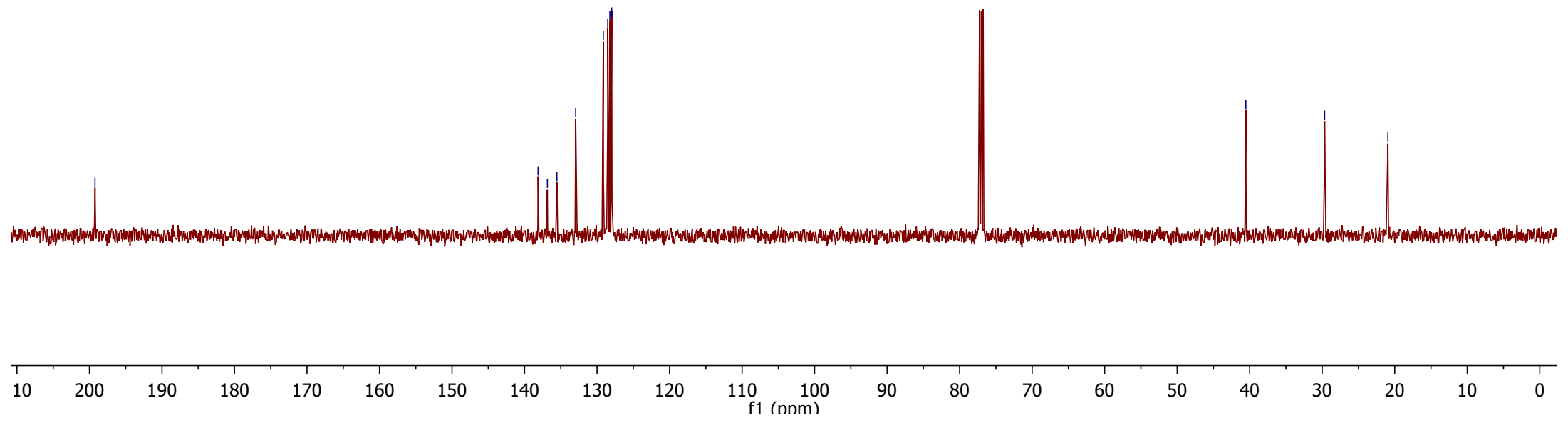
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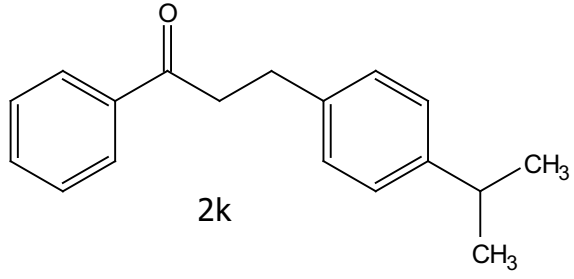
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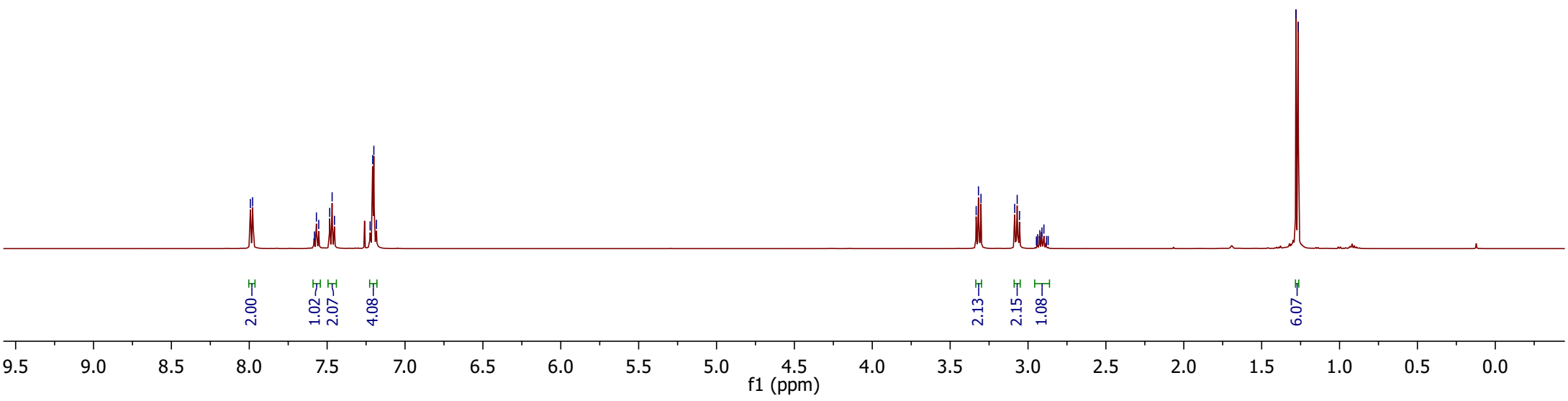


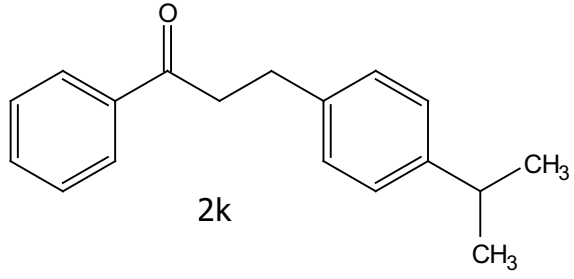


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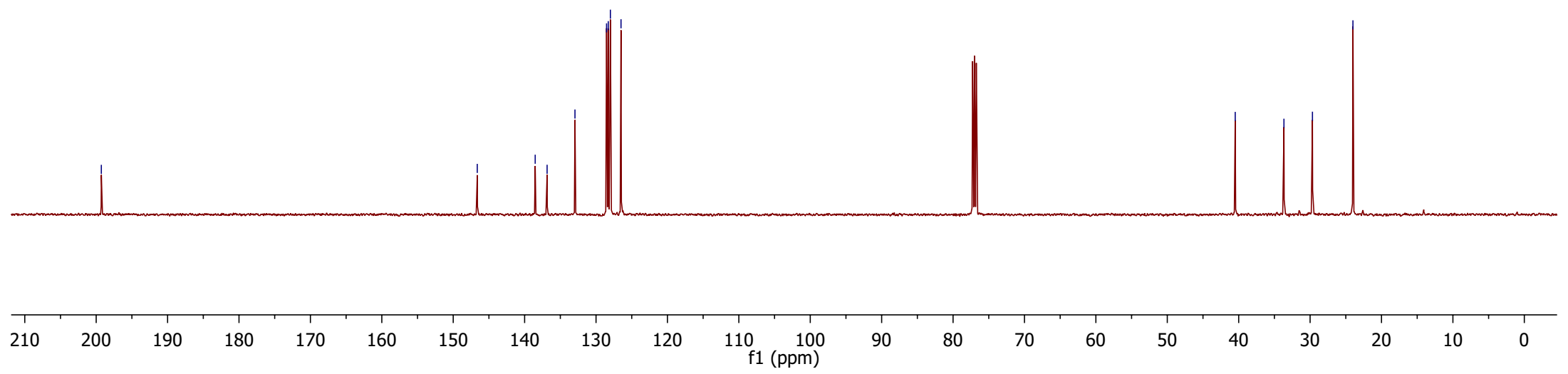
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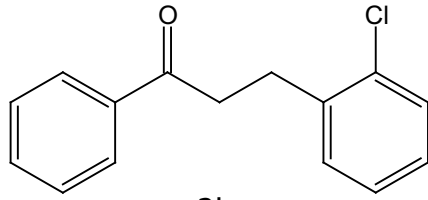
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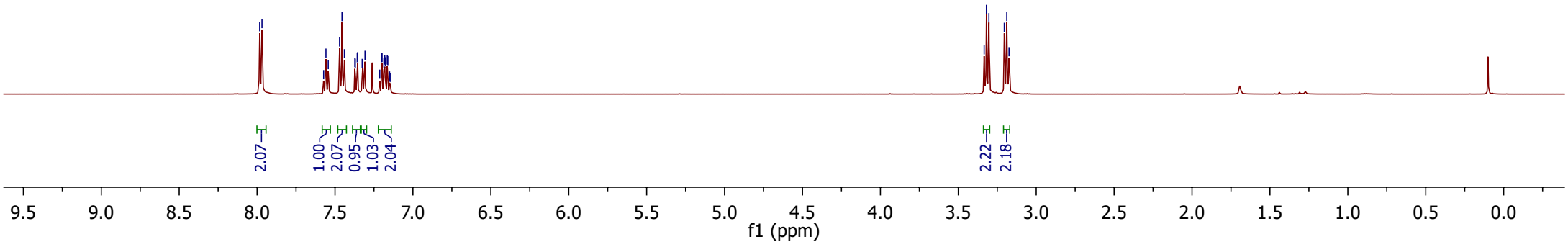


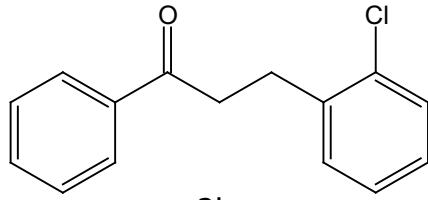


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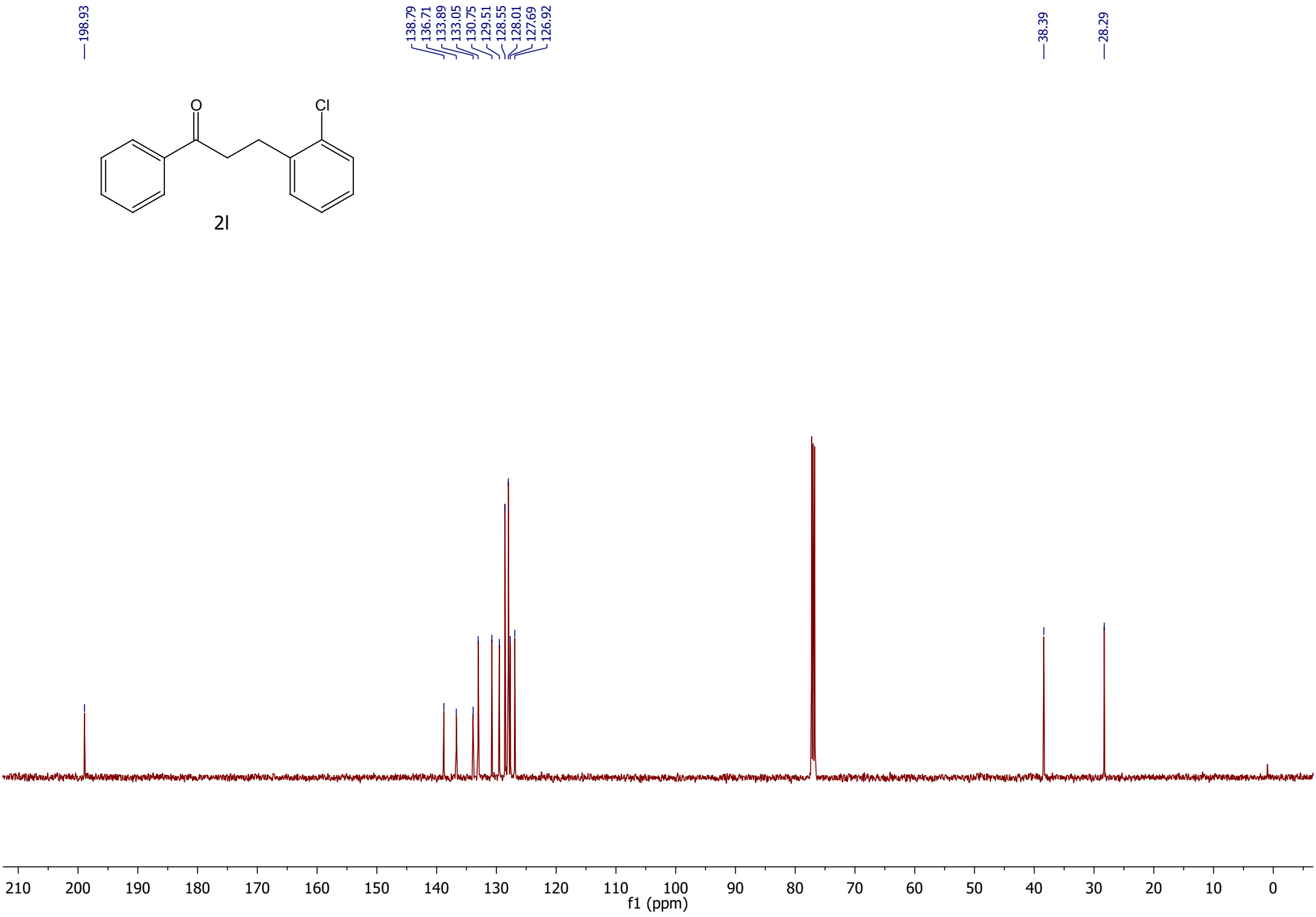
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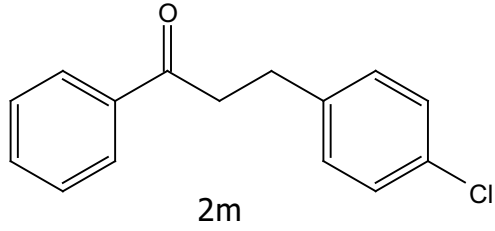
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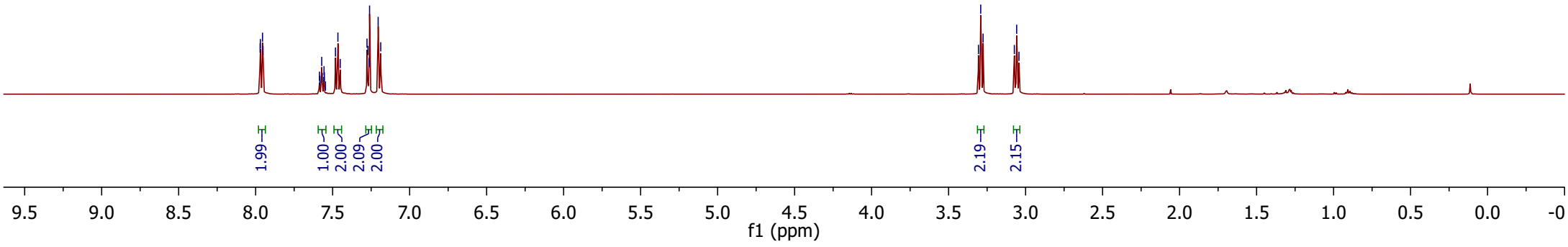
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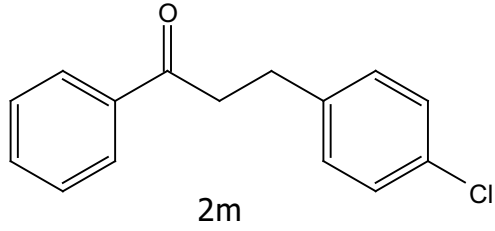




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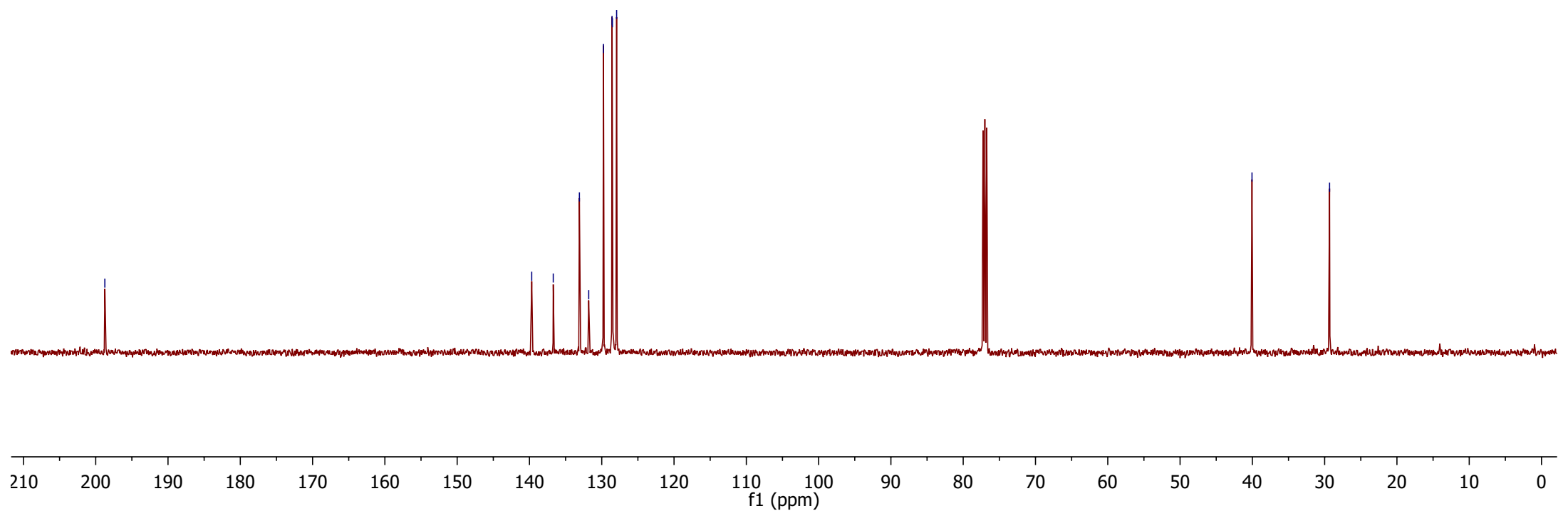


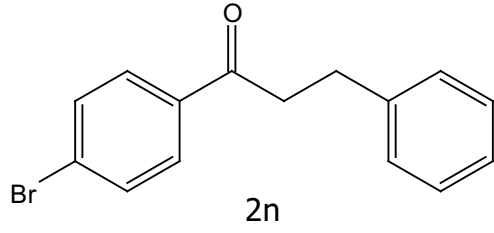
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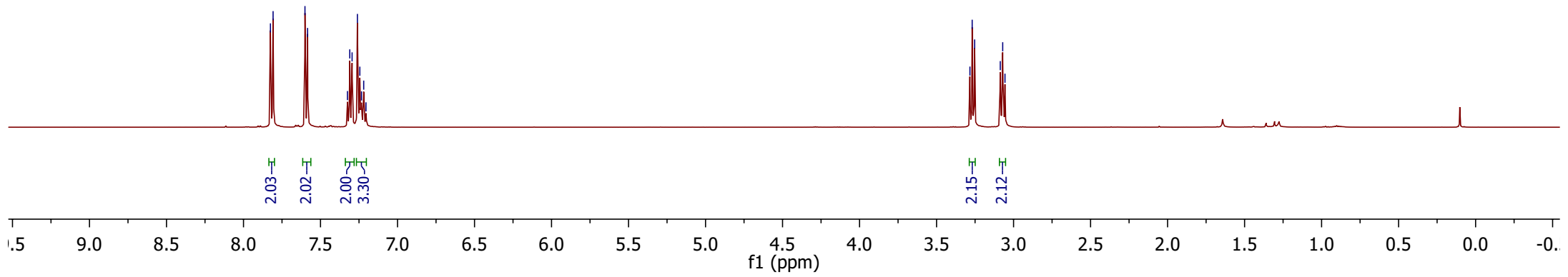
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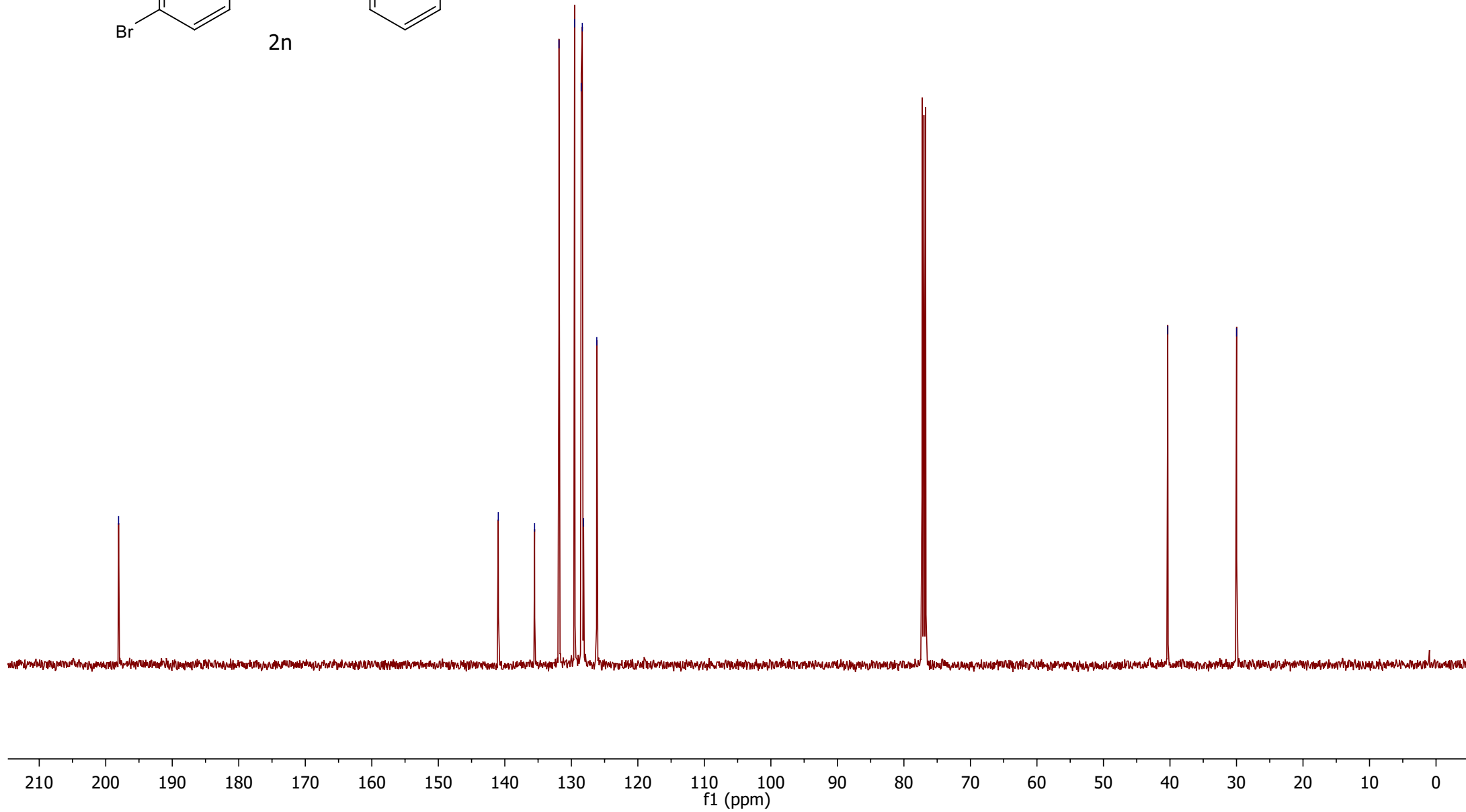
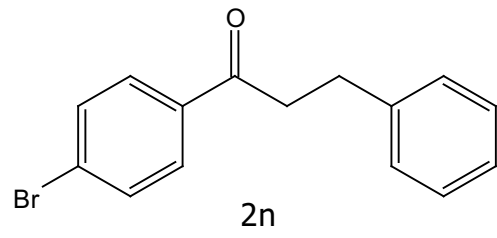


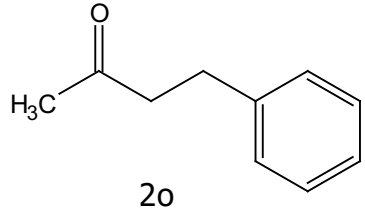
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— 40.34

— 29.99

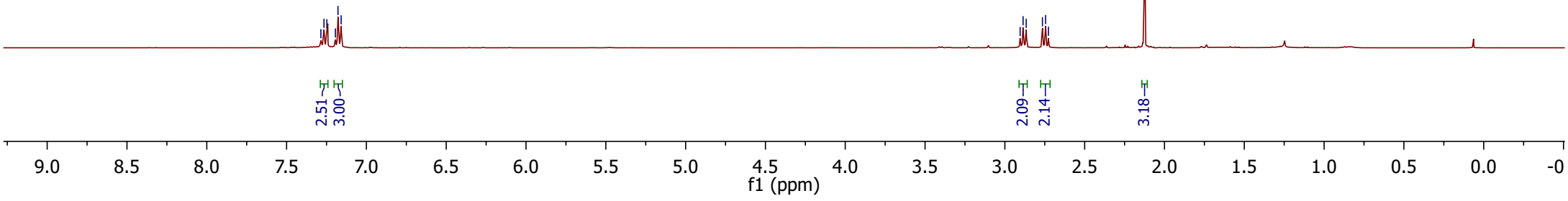


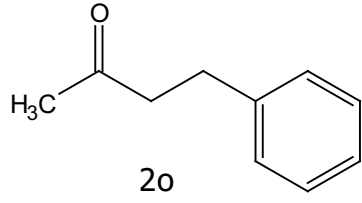


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—140.96

128.46

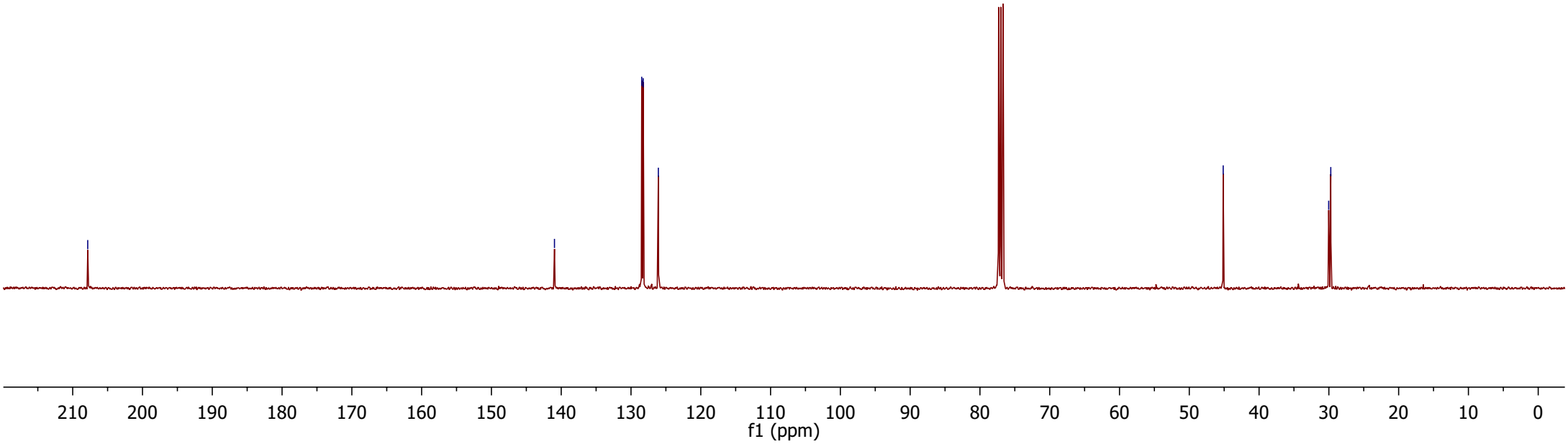
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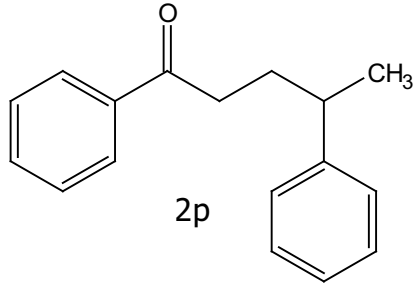
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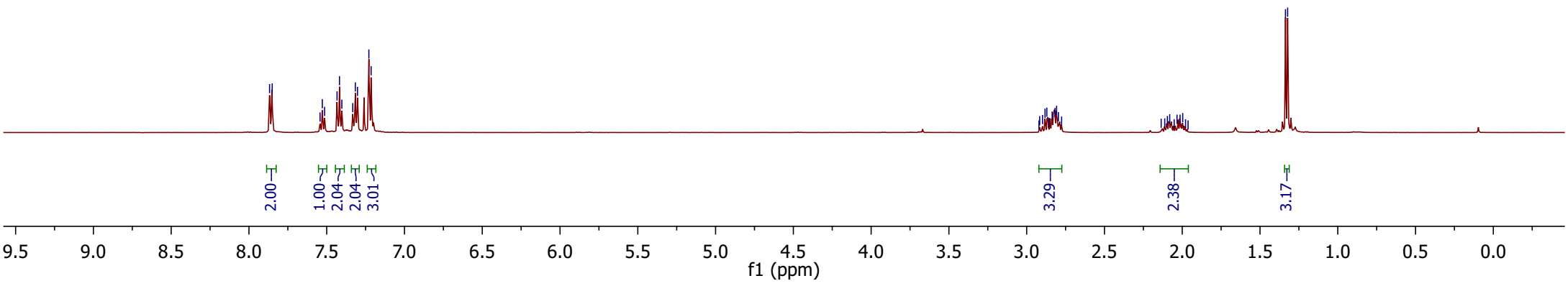
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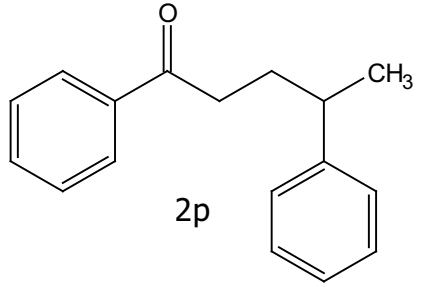


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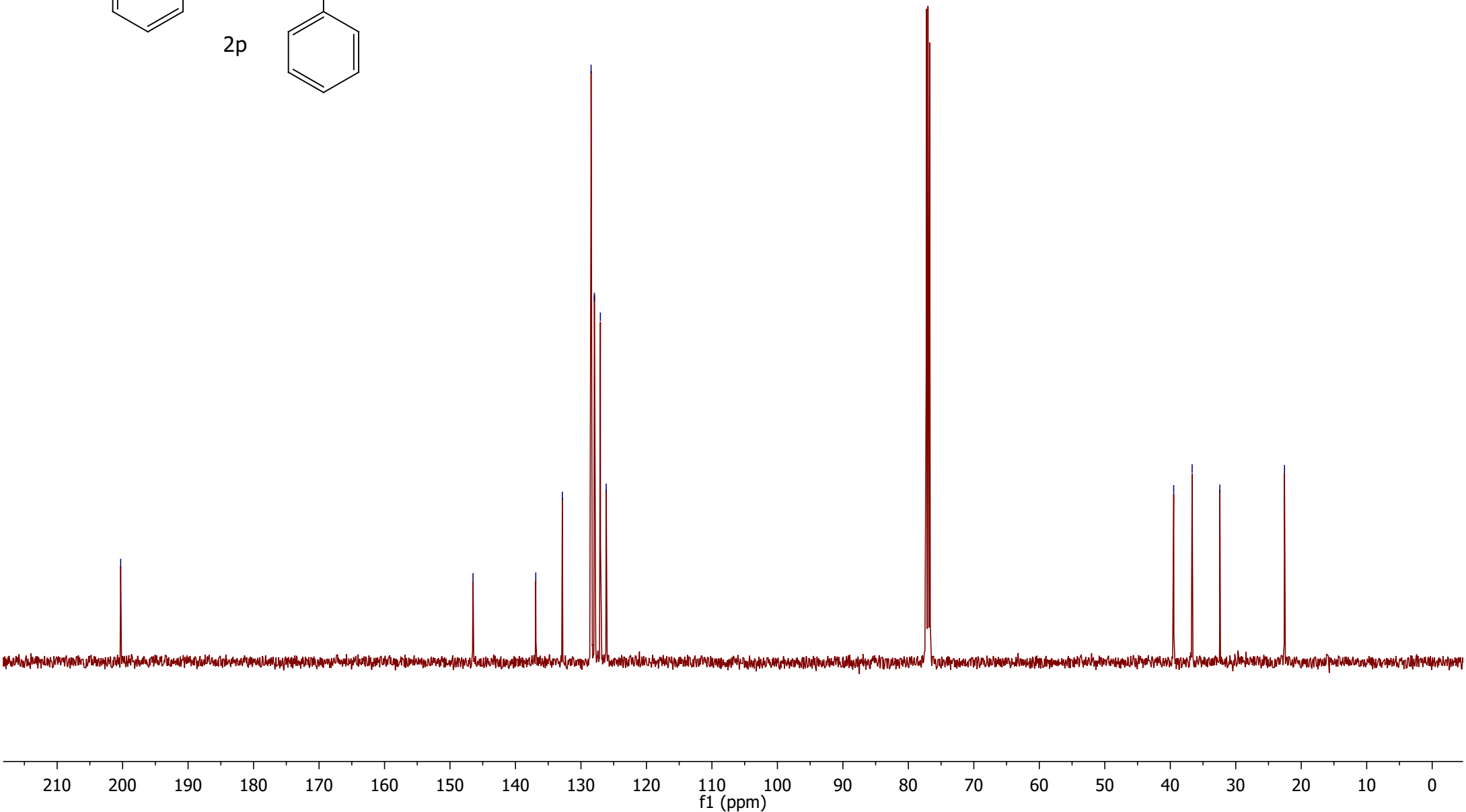


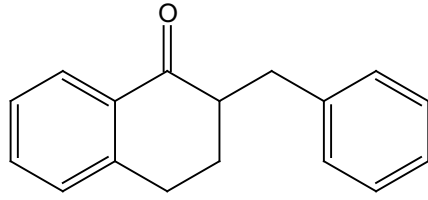
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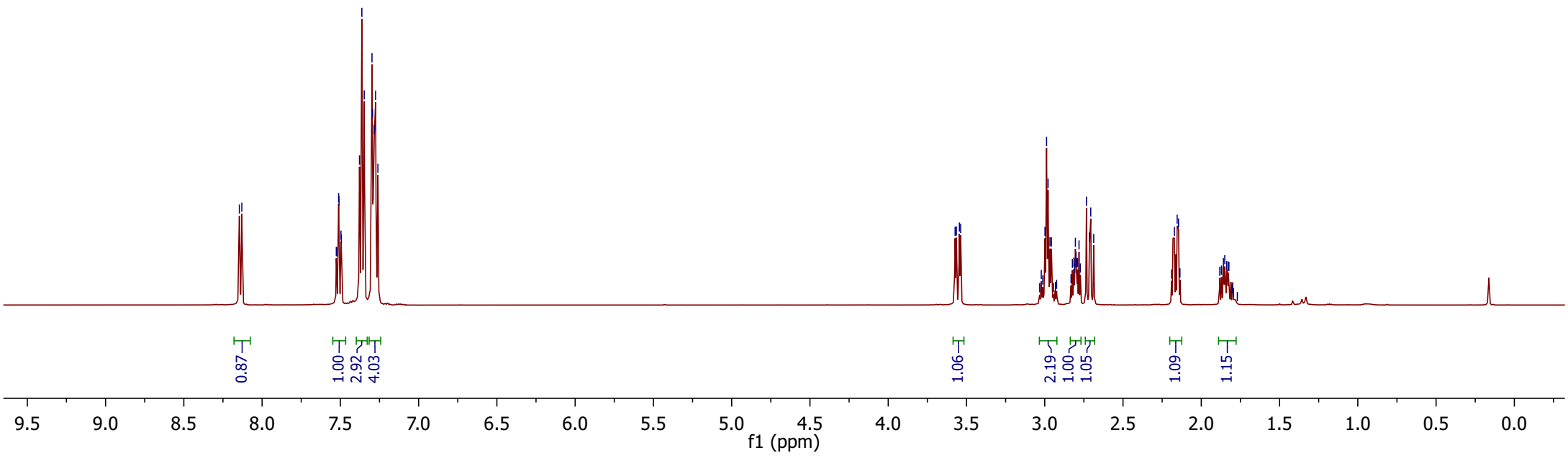


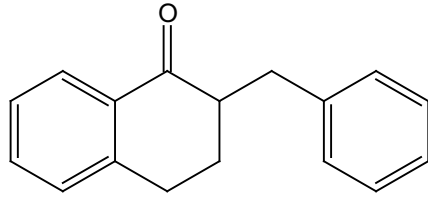


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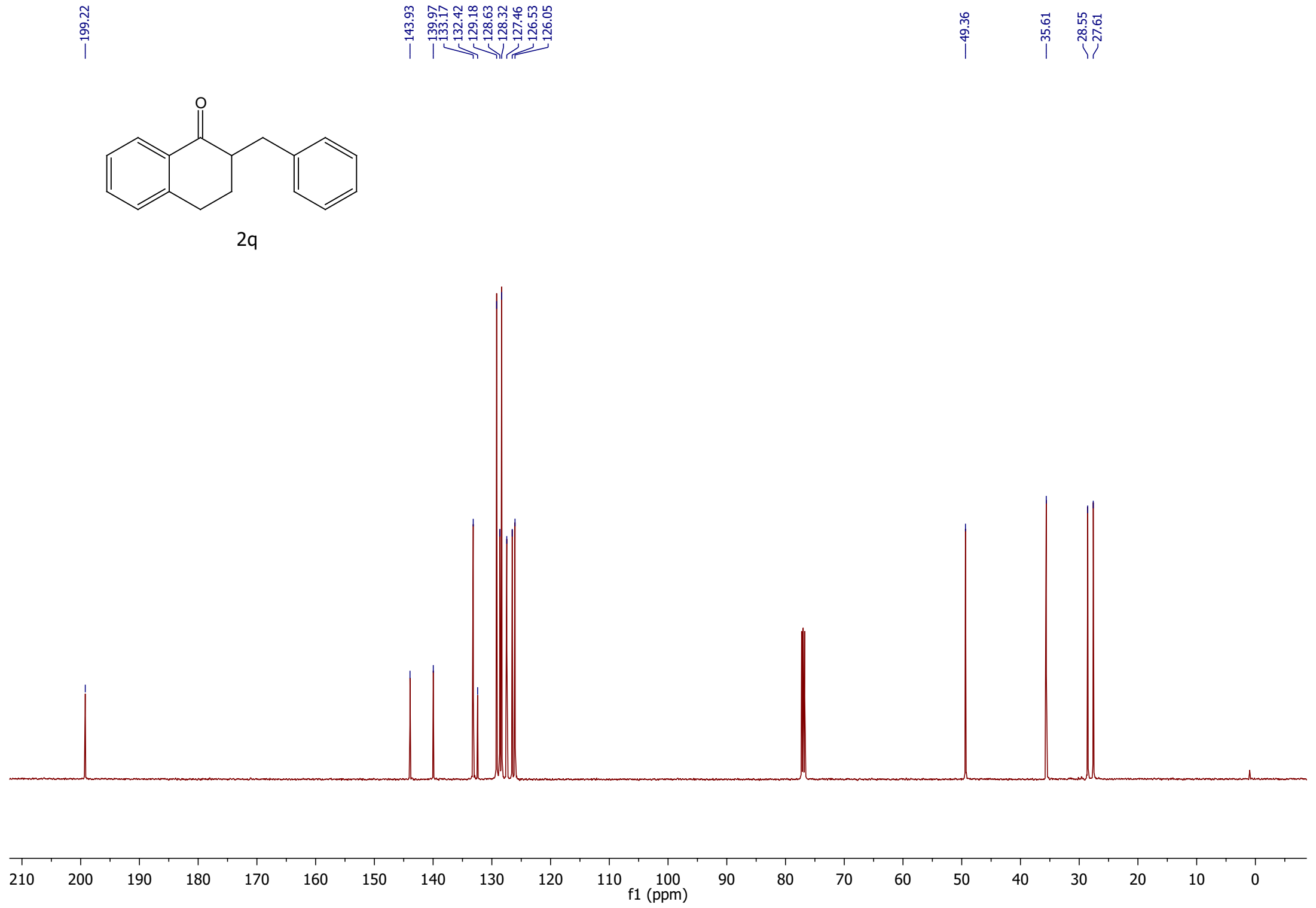
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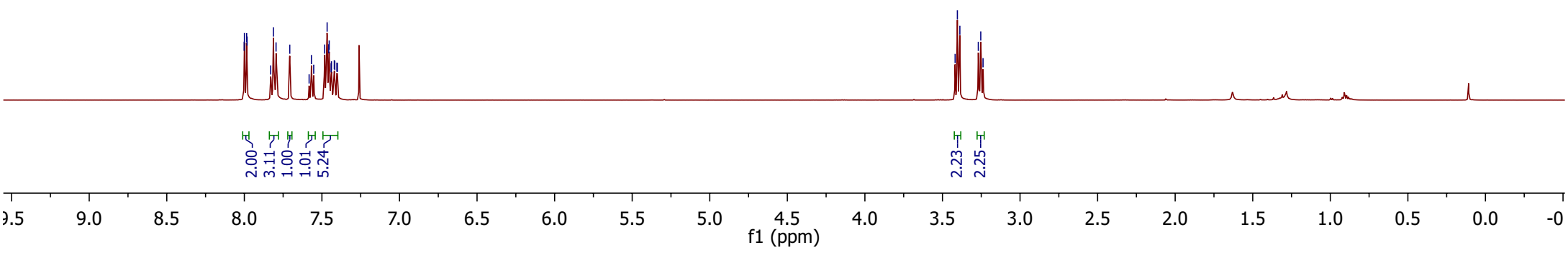
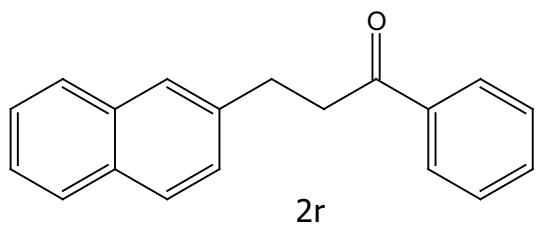


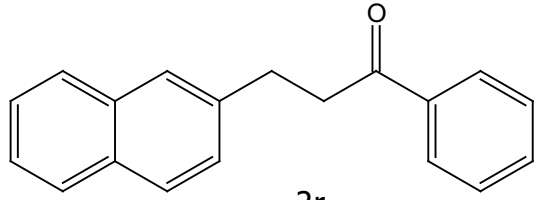
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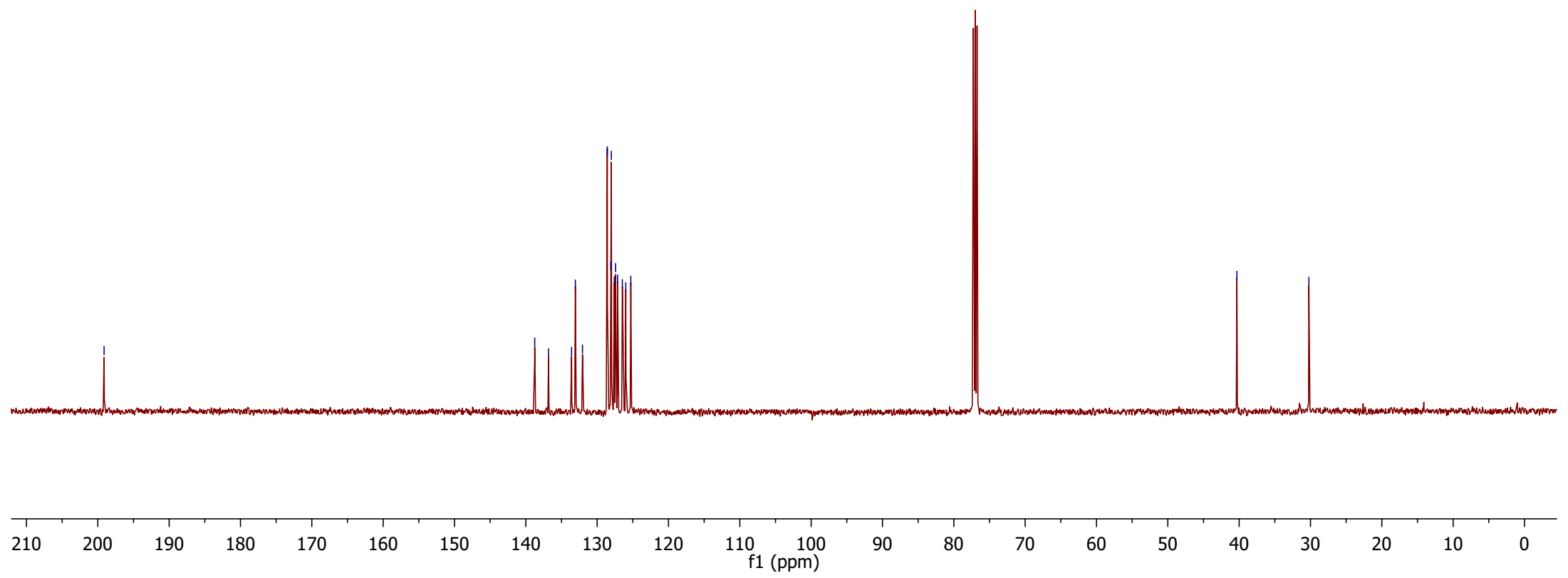


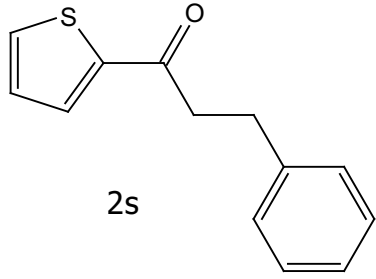
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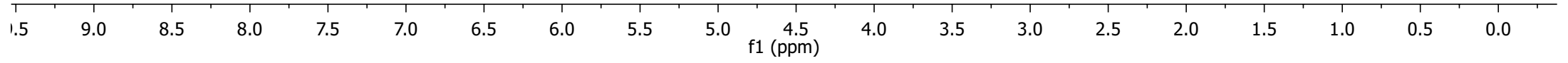
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—140.91

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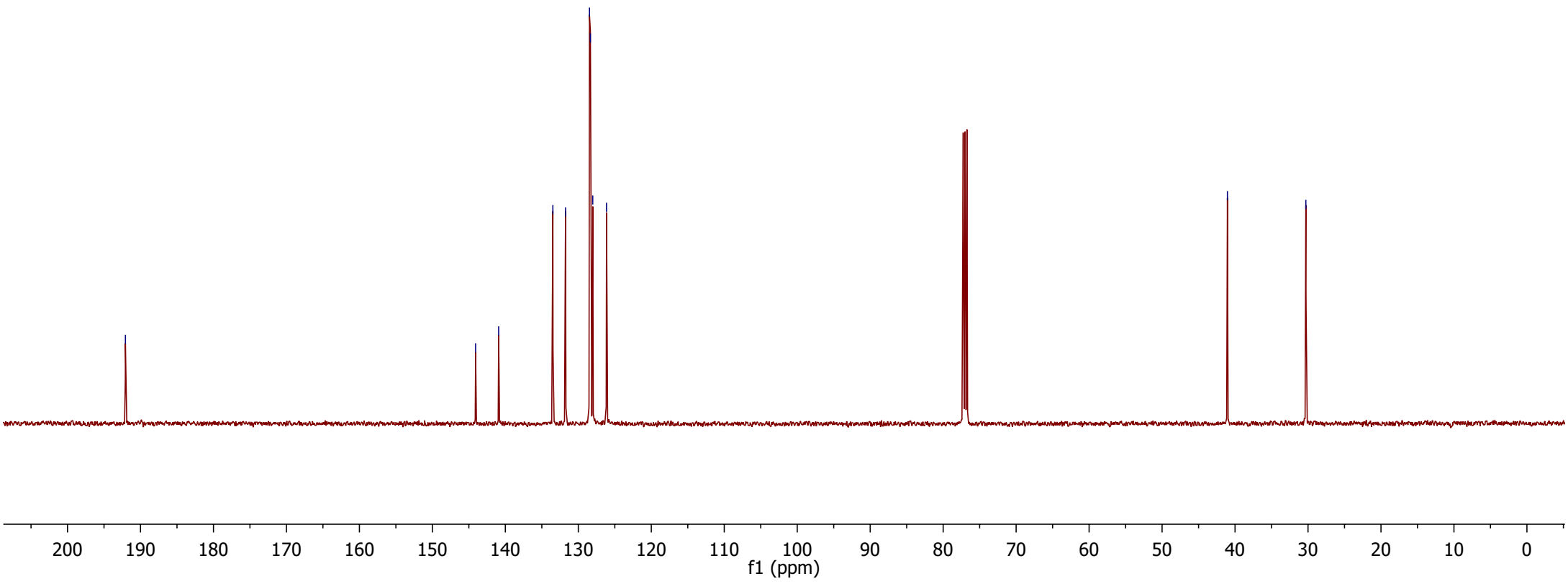
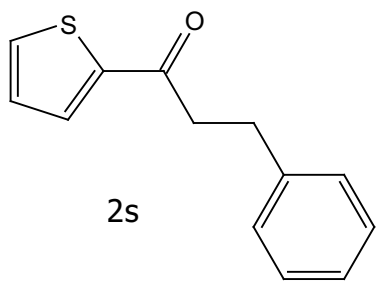
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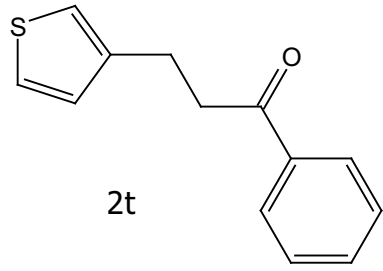
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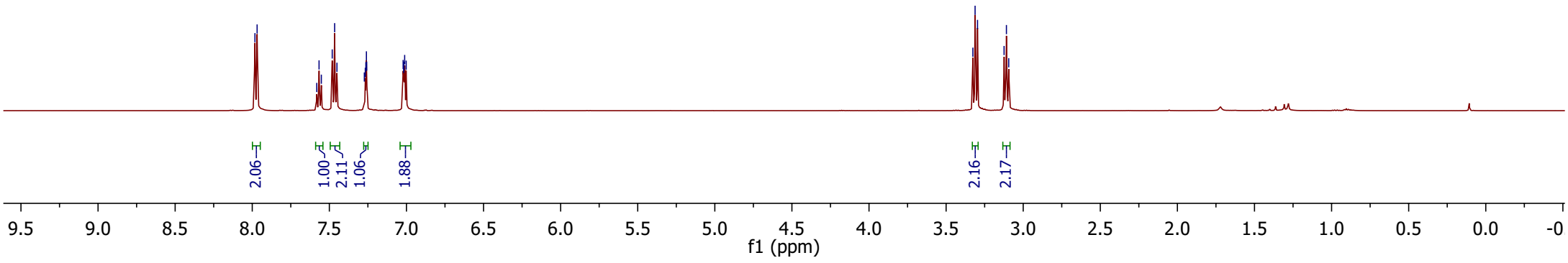
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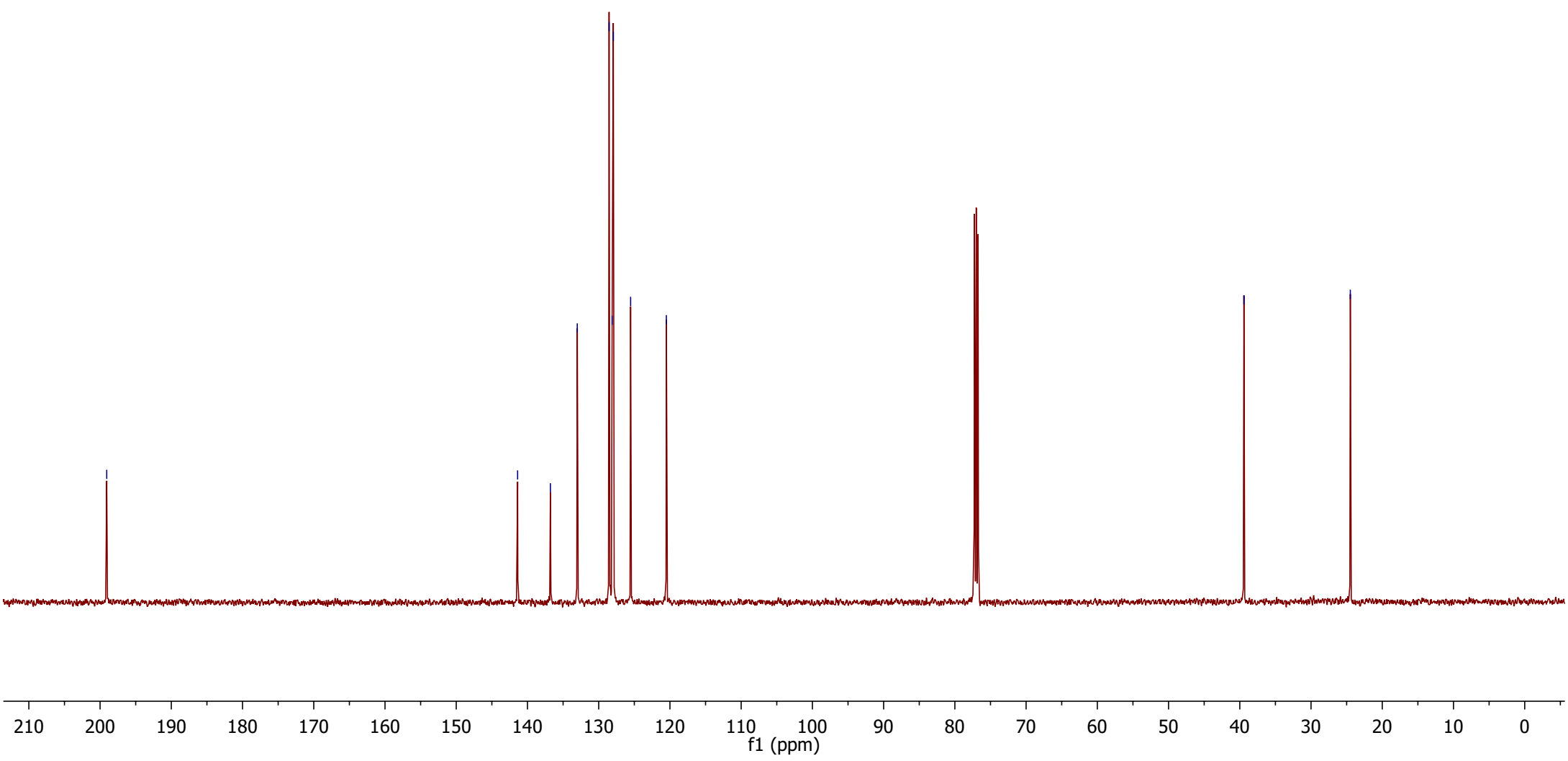
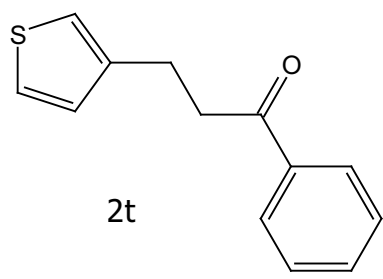


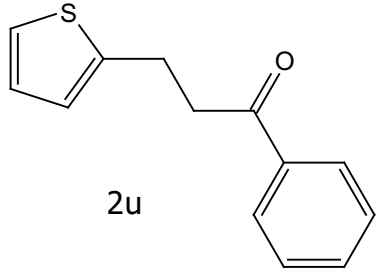
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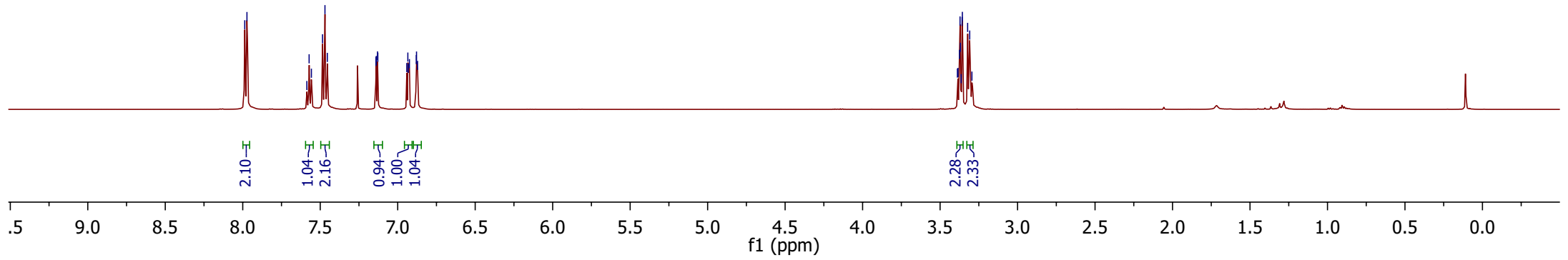
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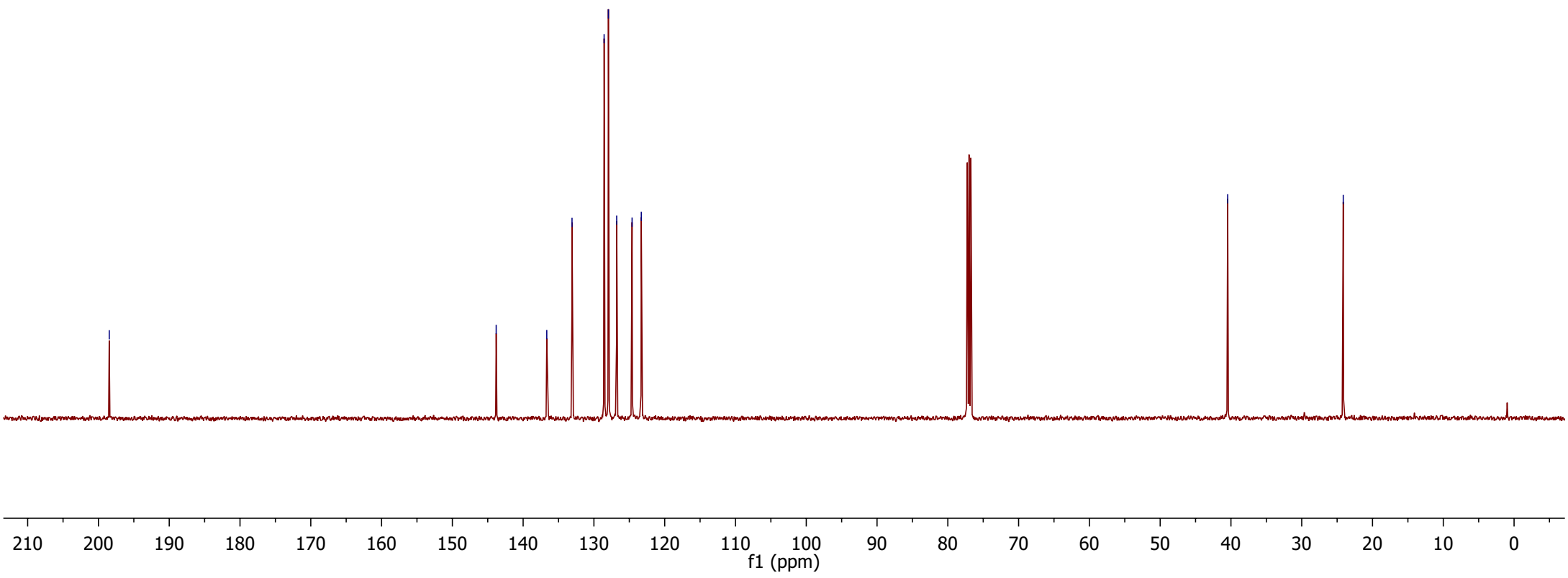
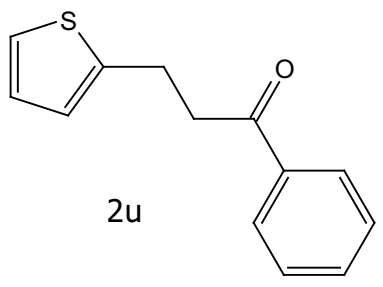
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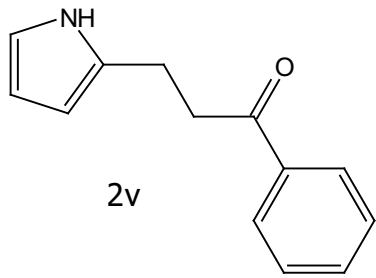
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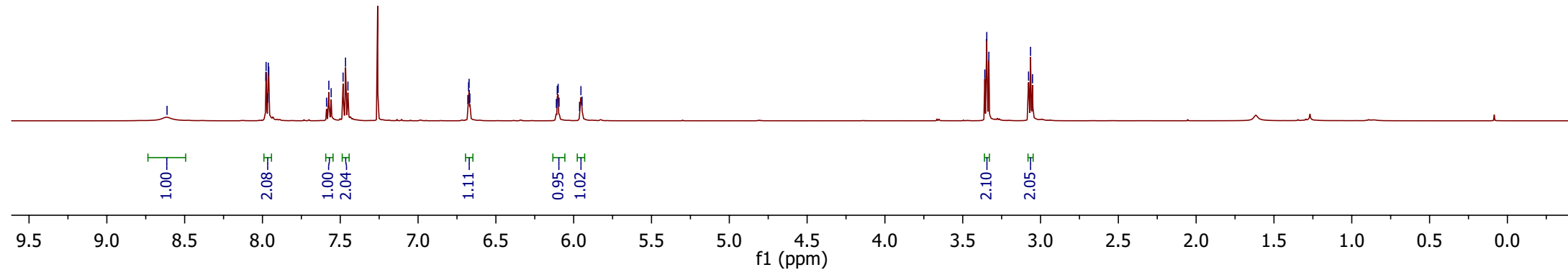
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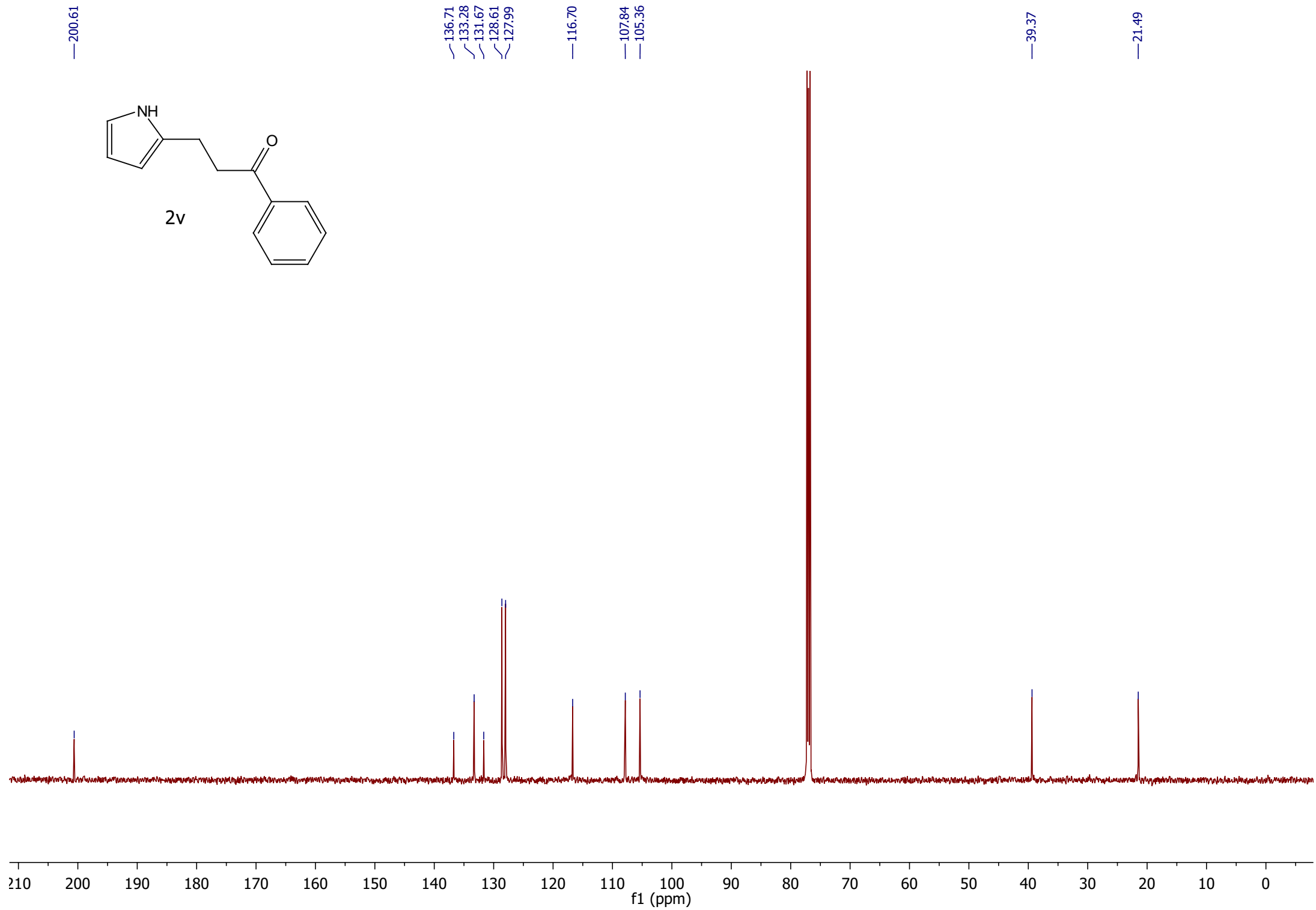
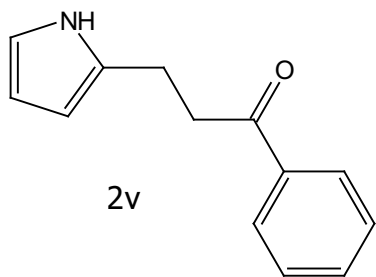
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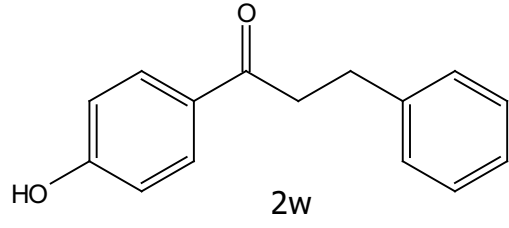




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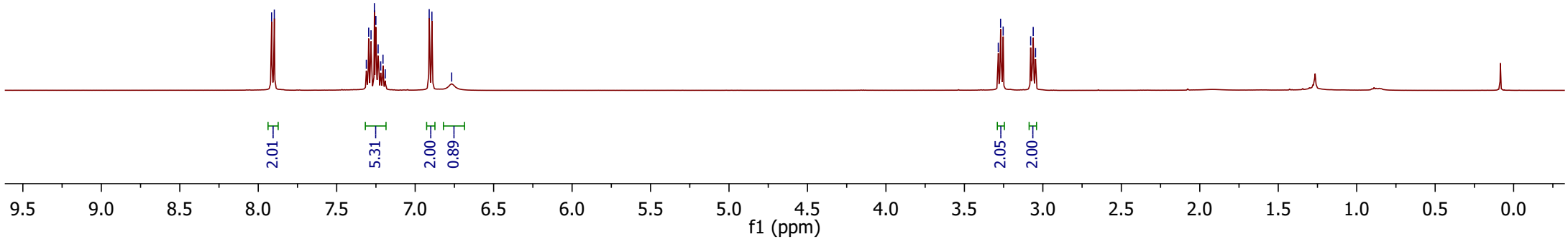


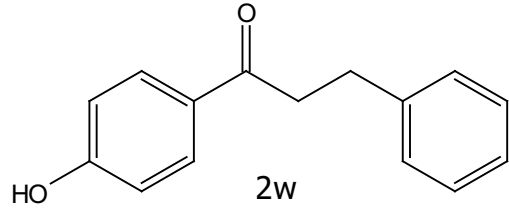




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—198.96

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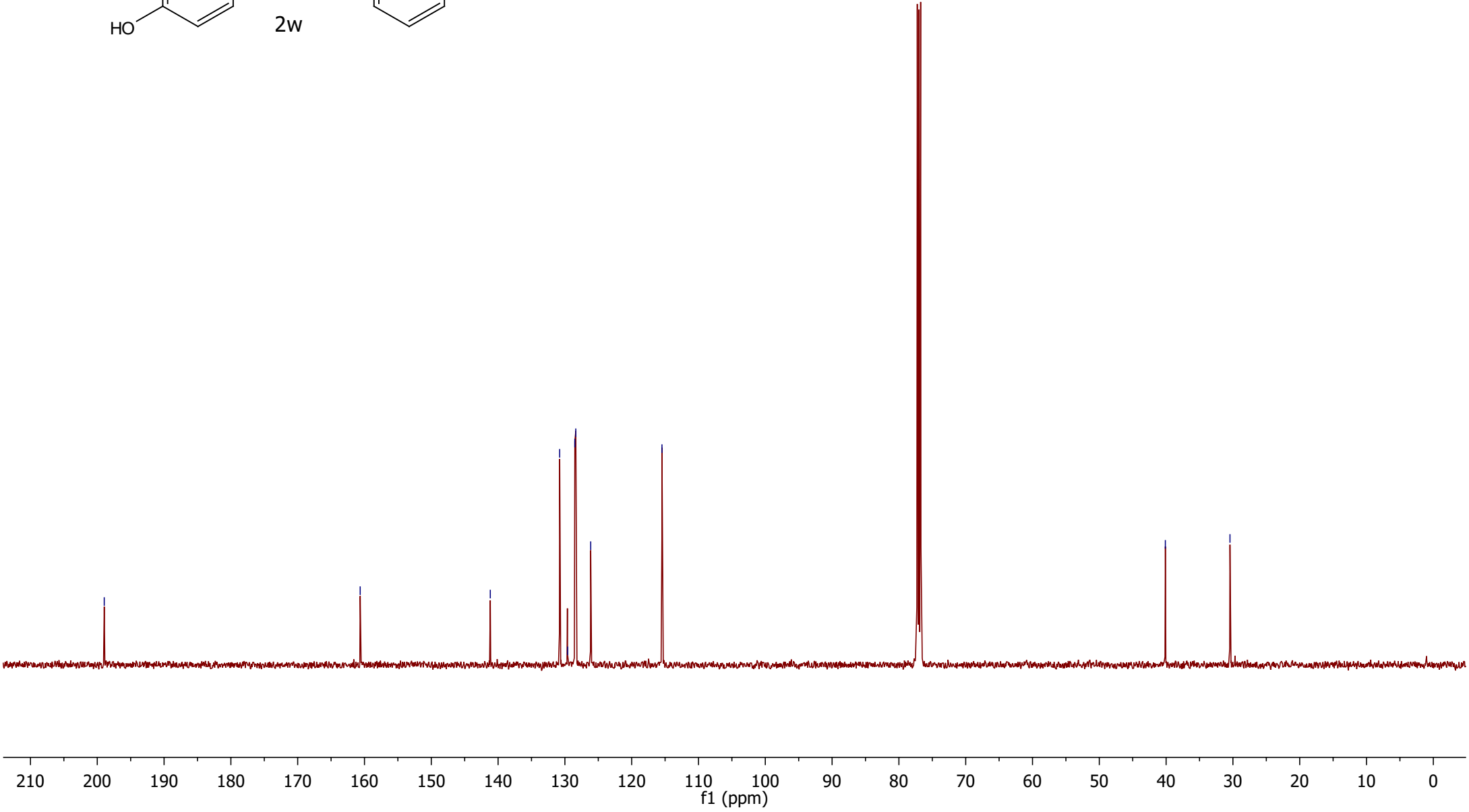
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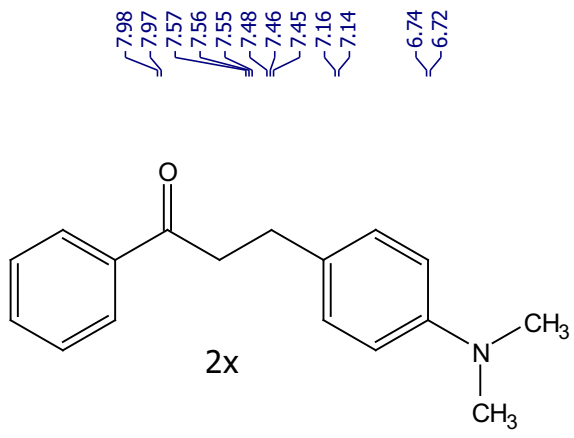
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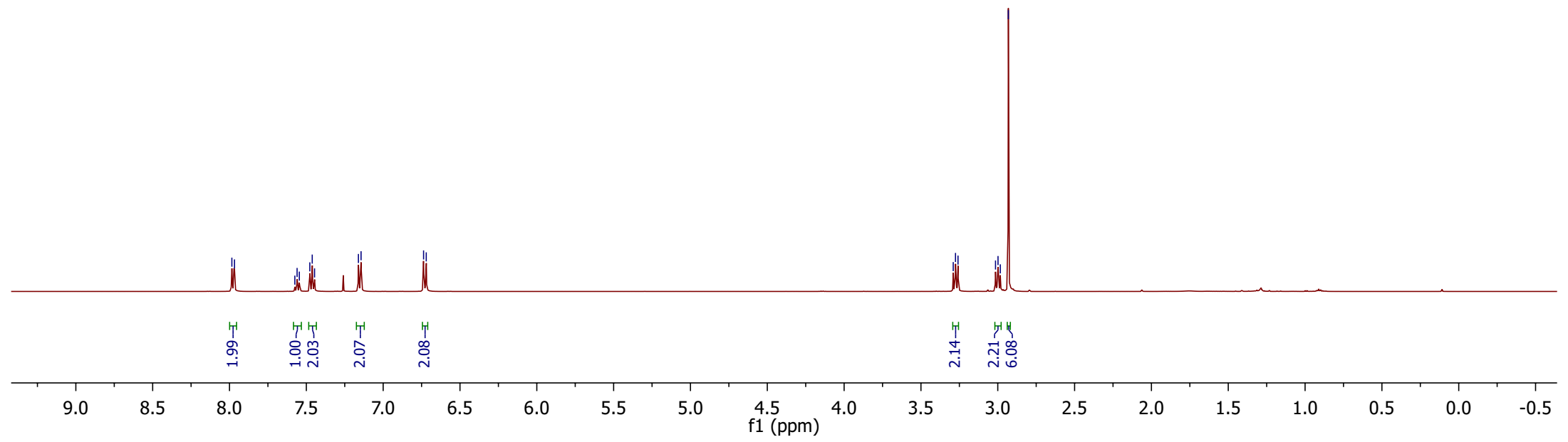
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— 199.61

— 149.21

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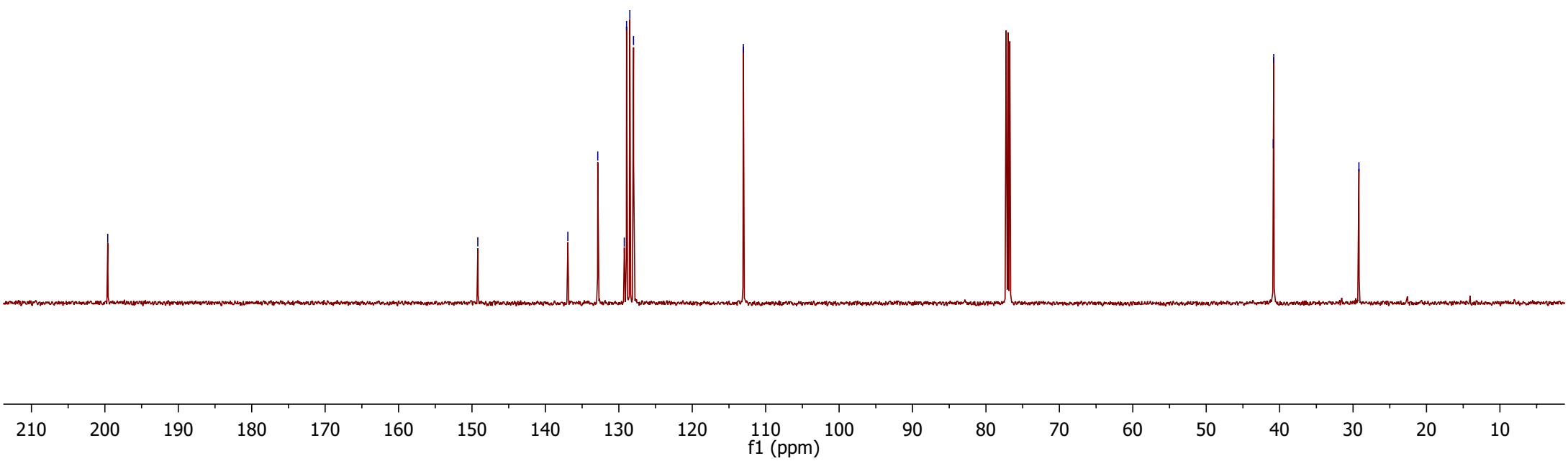
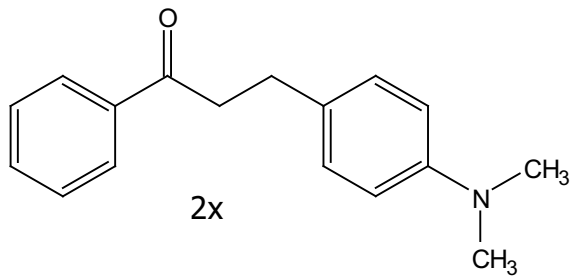
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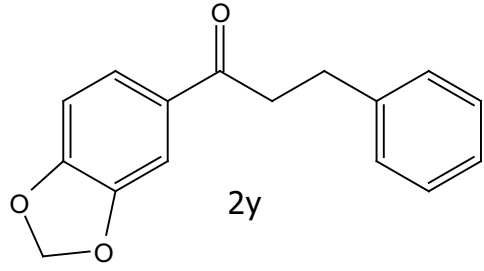
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— 29.20

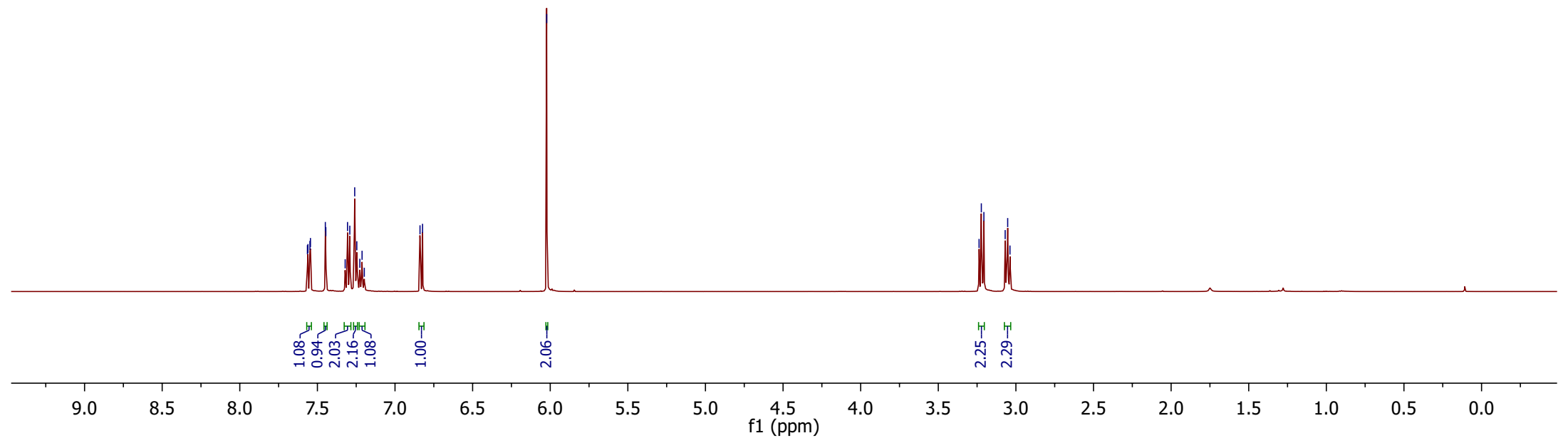




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—197.17

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—141.25

131.65

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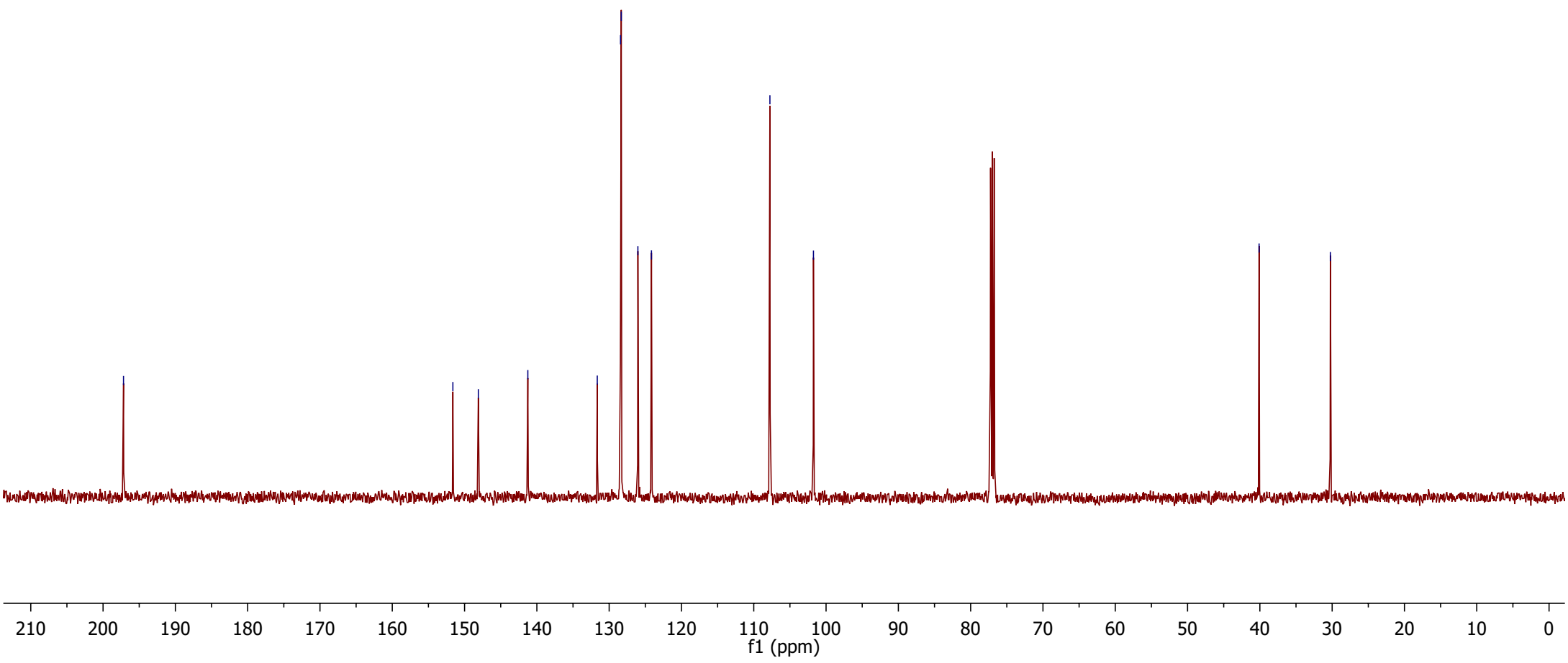
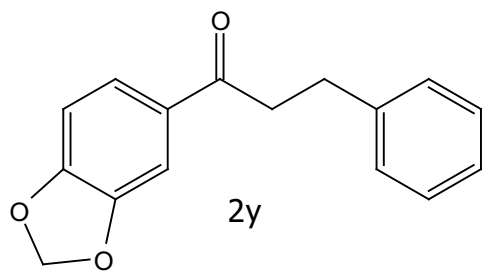
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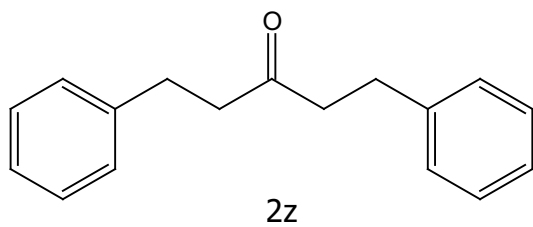
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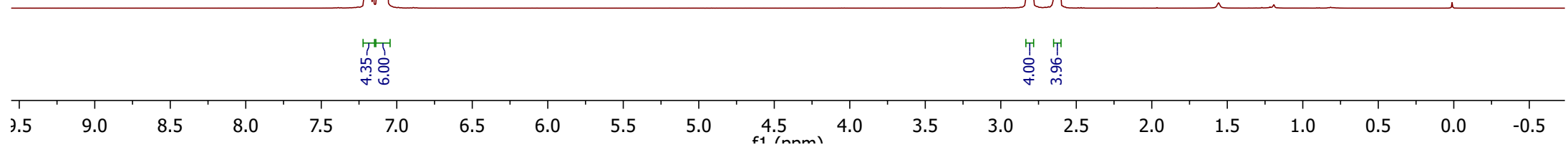
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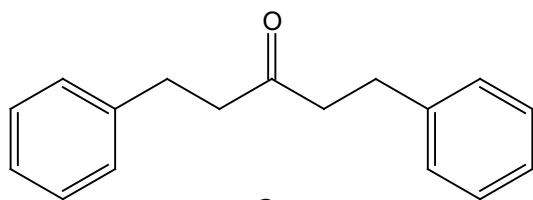




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2.61





2z

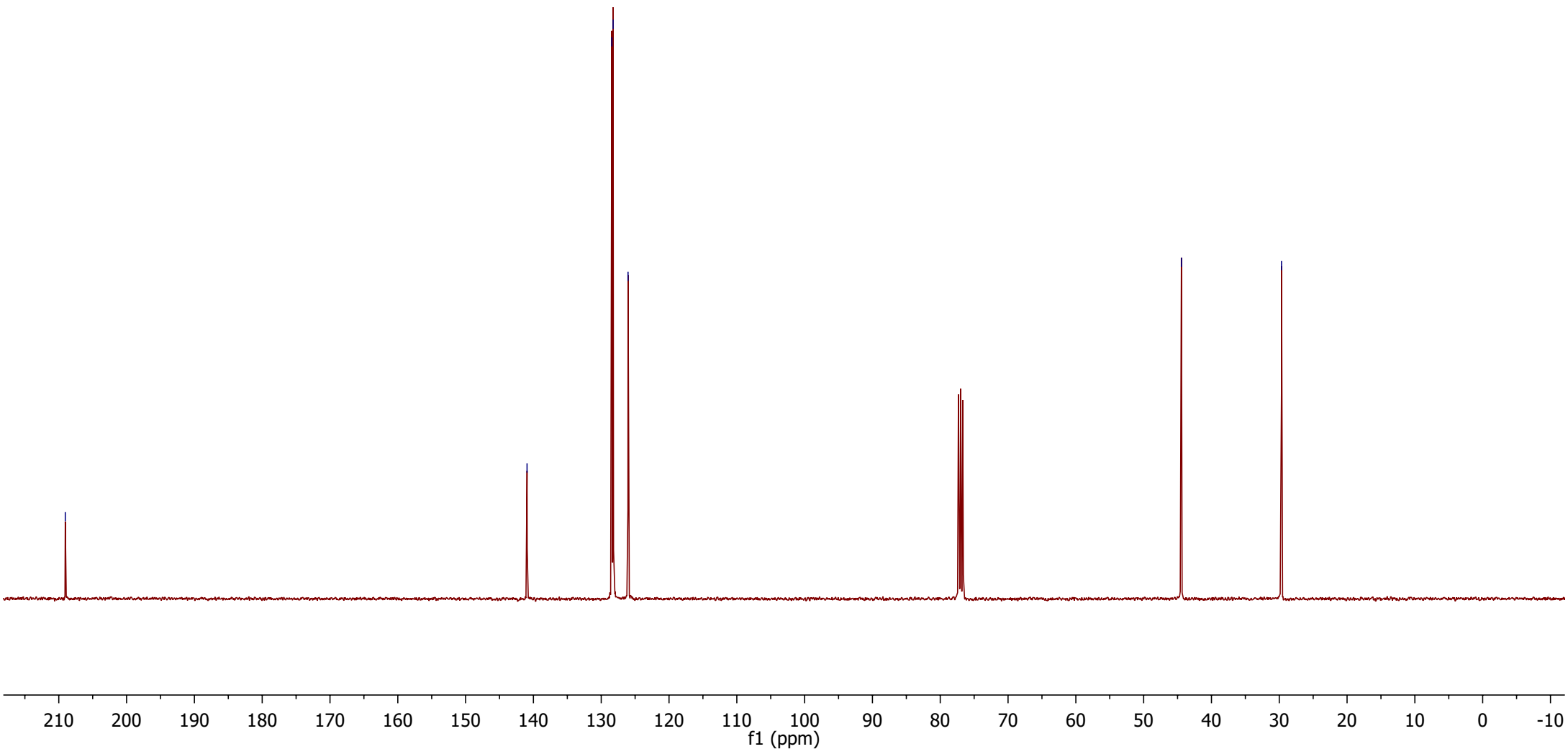
— 209.04

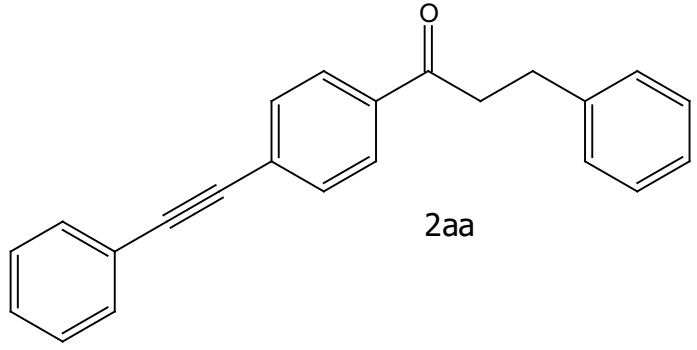
— 140.94

128.43
128.25
126.04

— 44.43

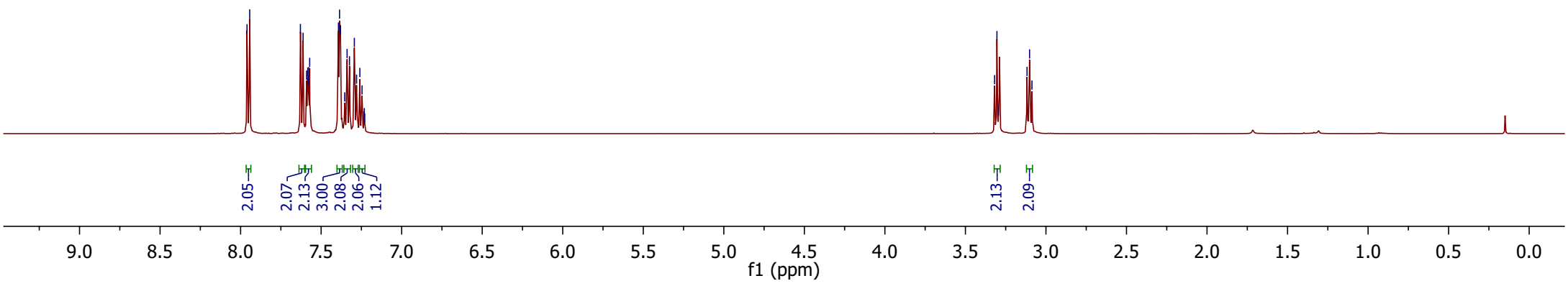
— 29.66

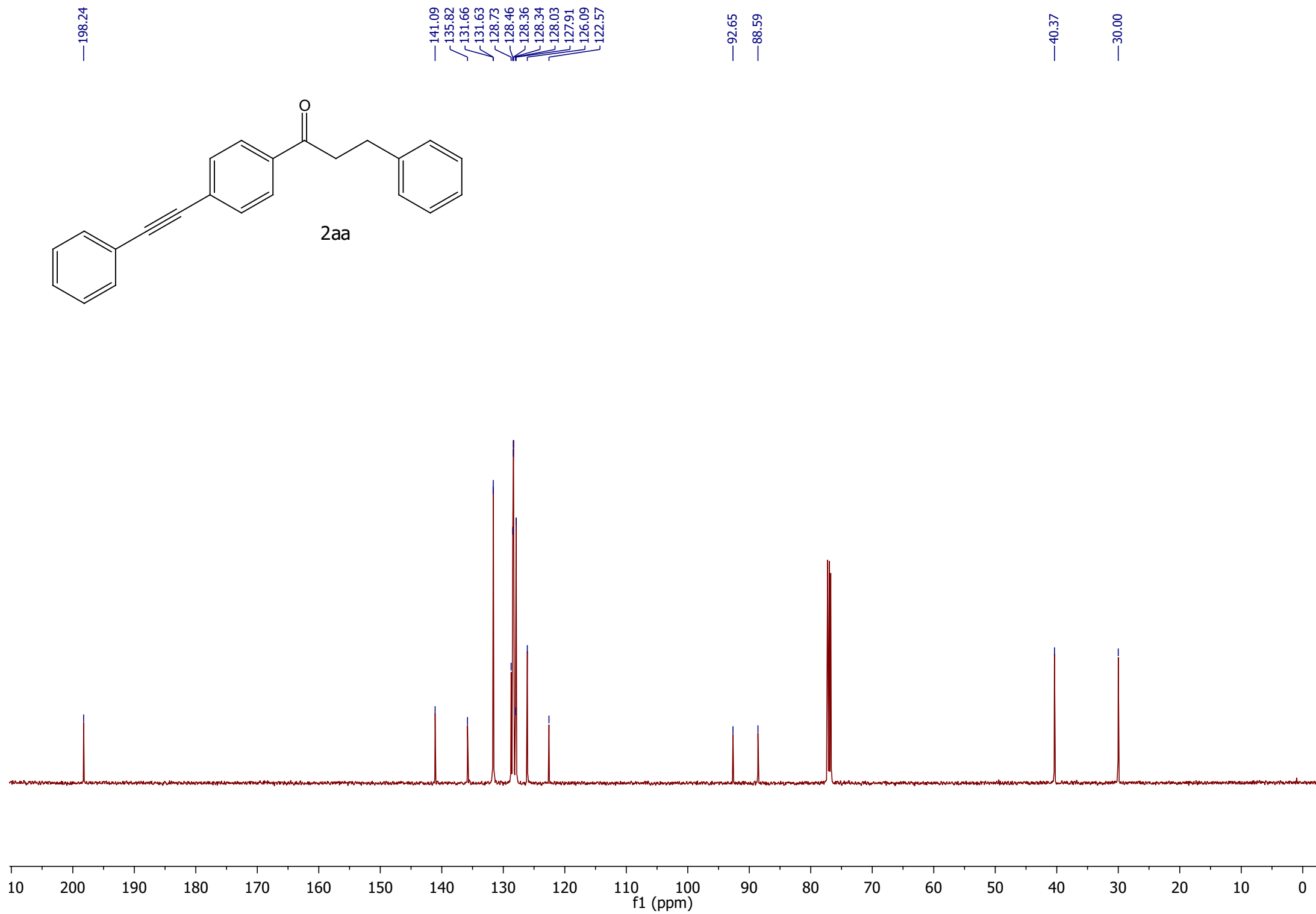
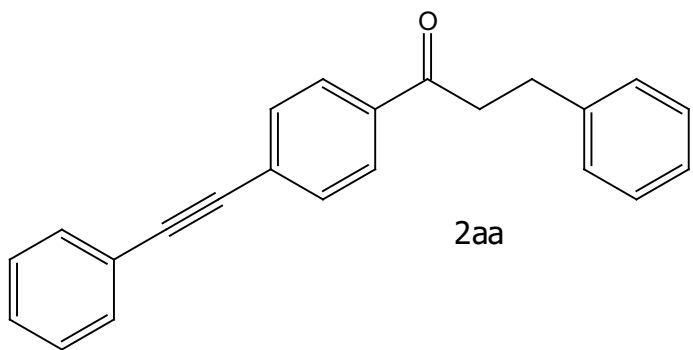


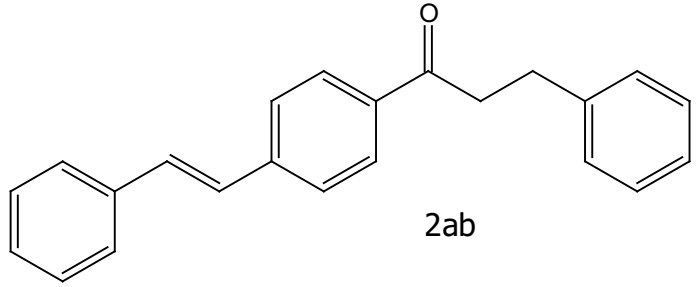


7.96
7.94
7.63
7.61
7.59
7.58
7.58
7.57
7.39
7.39
7.38
7.35
7.34
7.32
7.29
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7.25
7.23
7.23

3.32
3.30
3.30
3.12
3.10
3.09

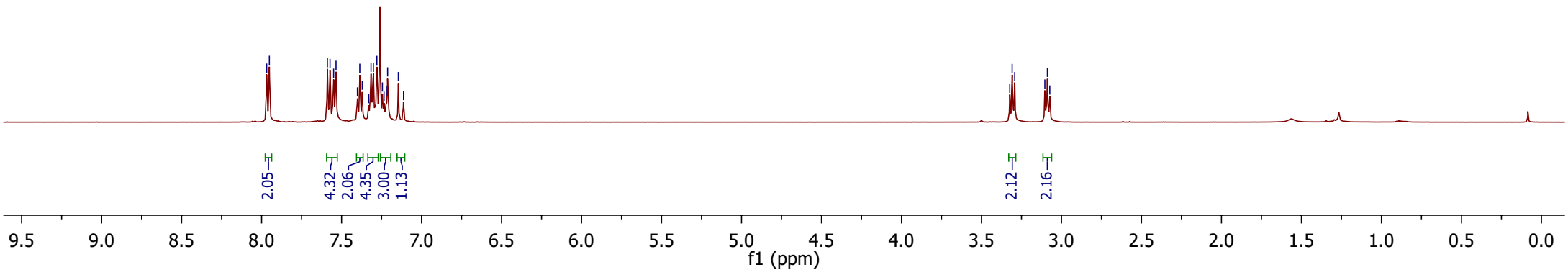


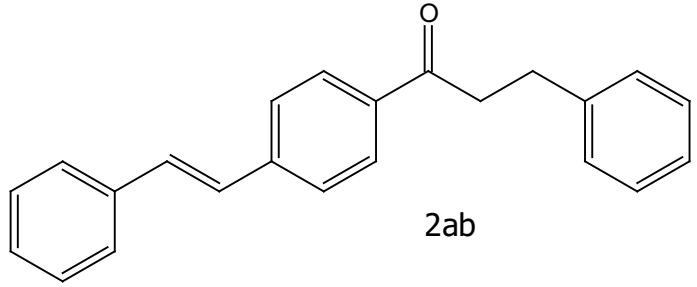




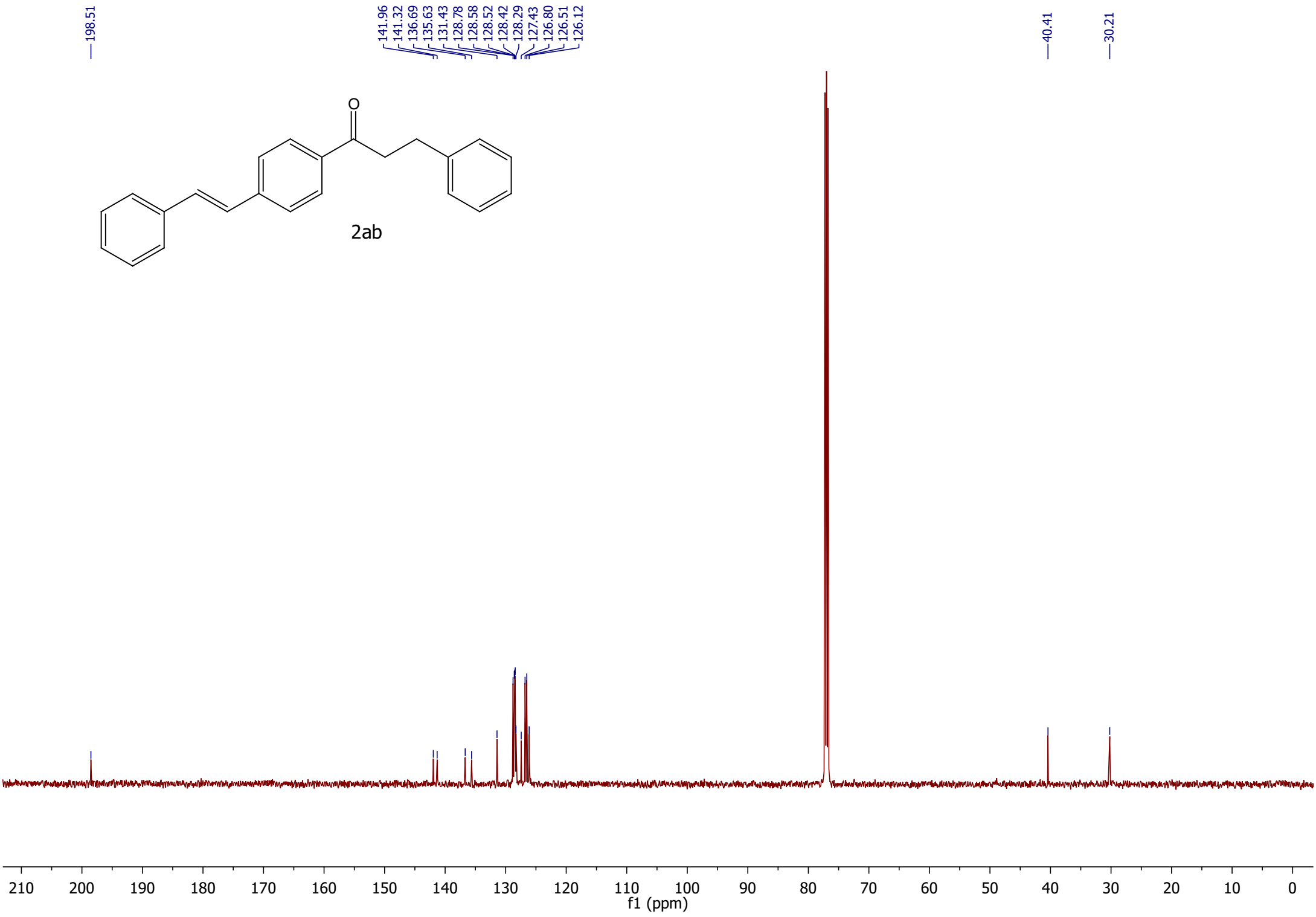
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7.57
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7.28
7.24
7.23
7.22
7.21
7.15
7.11

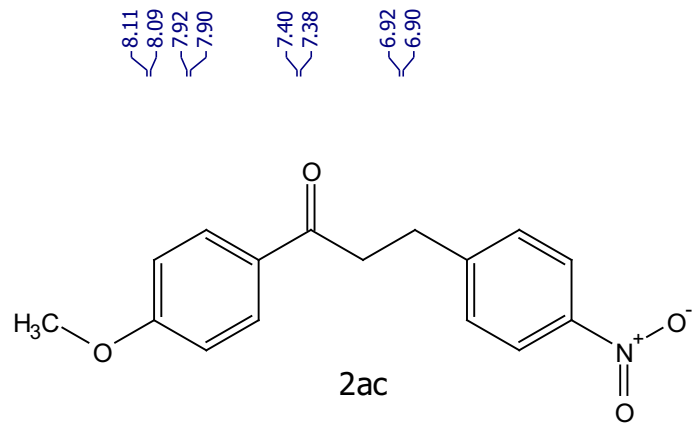
3.32
3.31
3.29
3.10
3.09
3.07





2ab





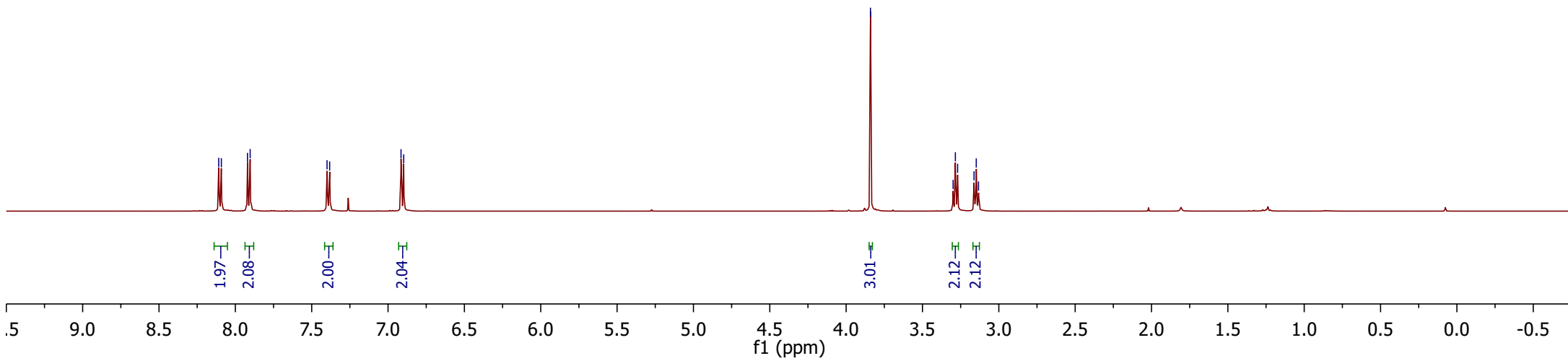
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8.09
7.92
7.90

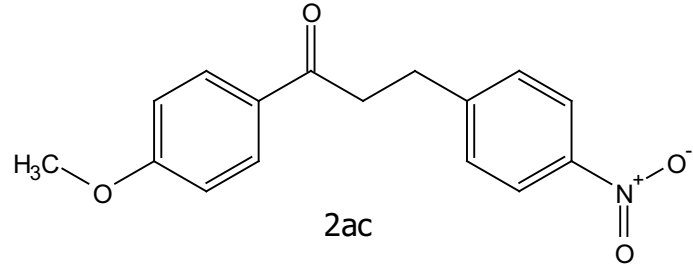
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7.38

6.92
6.90

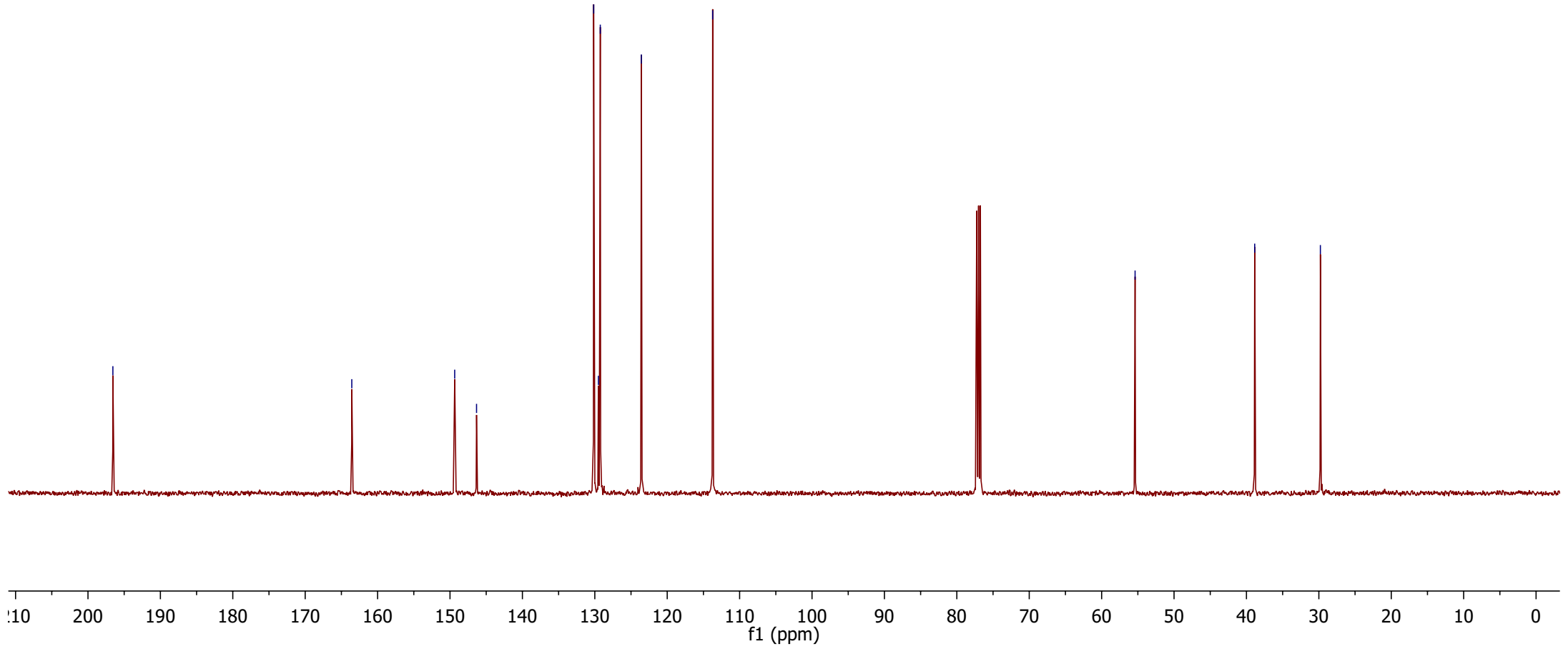
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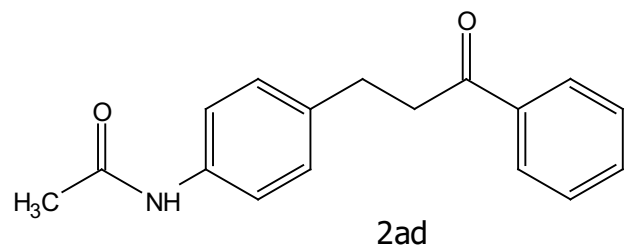
3.30
3.28
3.27
3.16
3.15
3.13





— 196.56 — 163.56 — 149.35 — 146.33 — 130.16 — 129.51 — 129.24 — 123.57 — 113.72 — 55.38 — 38.86 — 29.79

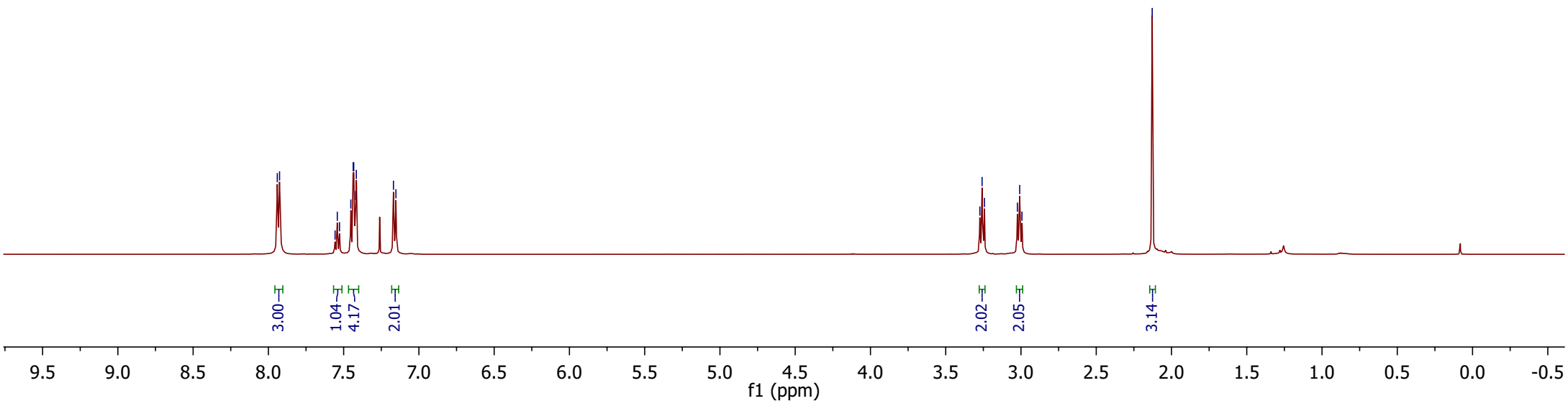


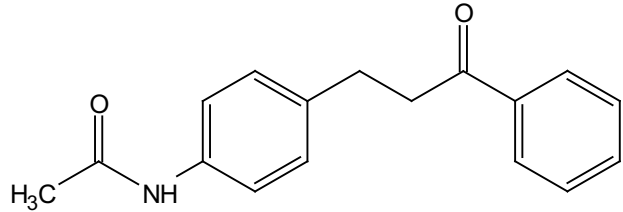


7.94
7.93
7.56
7.54
7.53
7.45
7.44
7.43
7.42
7.42
7.17
7.15

3.27
3.26
3.24
3.02
3.01
2.99

2.13

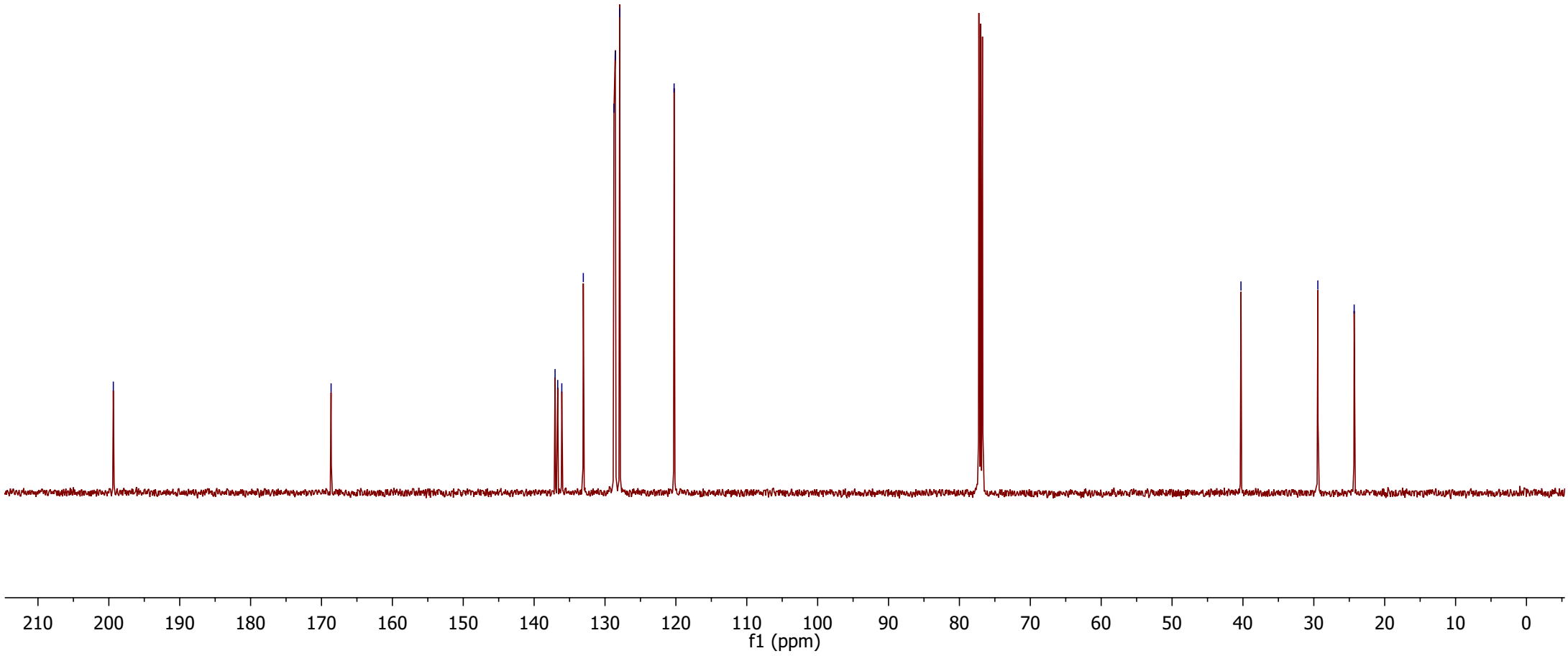


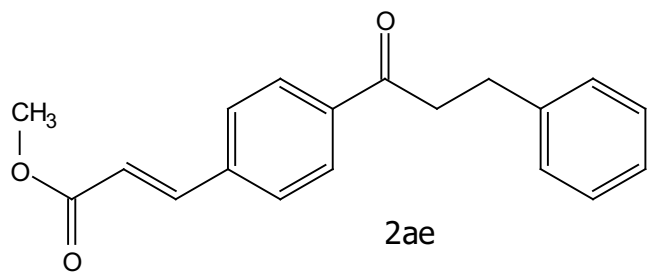


2ad

199.37
168.64
137.05
136.67
136.10
133.06
128.74
128.55
127.93
120.25

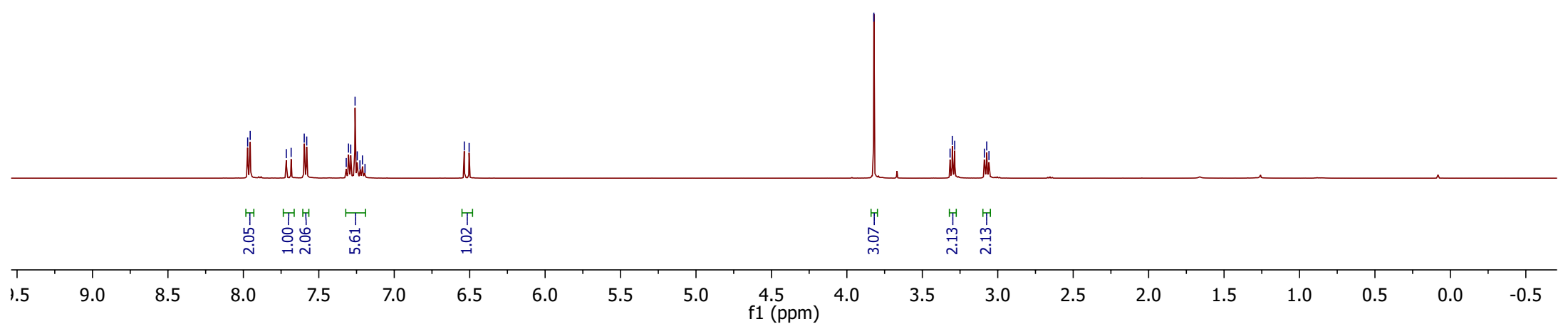
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24.31

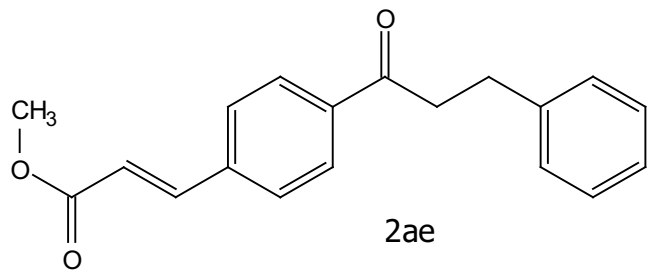




7.97
7.95
7.72
7.68
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7.58
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7.30
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7.26
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7.23
7.21
7.19
6.54
6.50

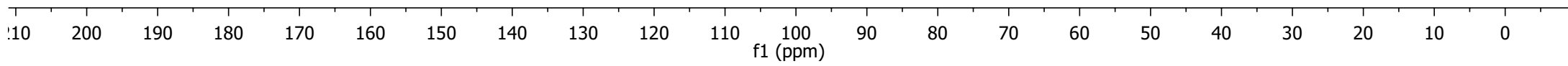
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3.07
3.06

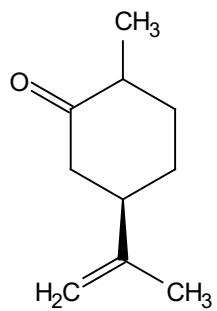




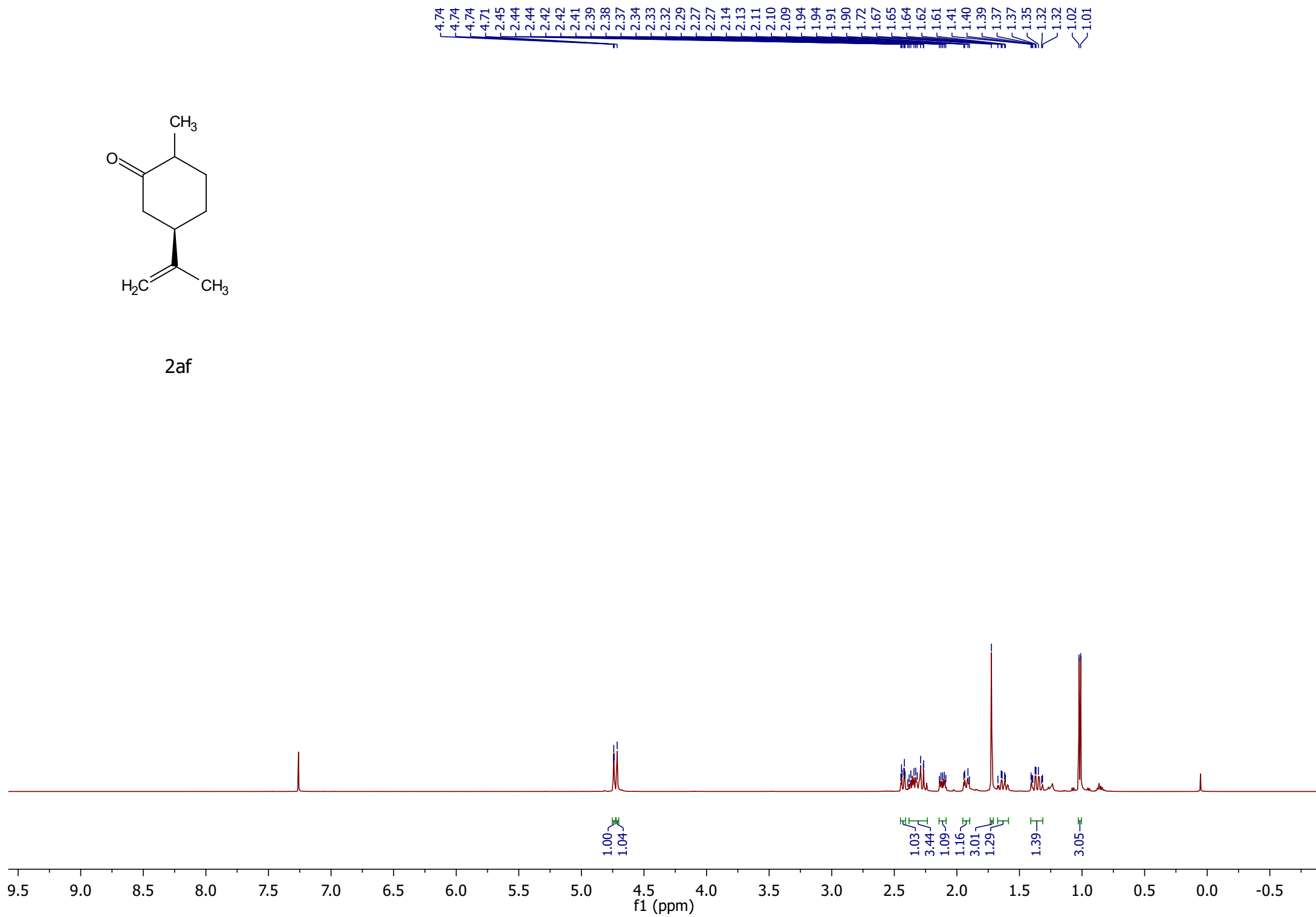
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143.25
141.04
138.58
137.69
128.52
128.36
128.12
126.16
120.23

51.87
40.53
30.00





2af



—212.67

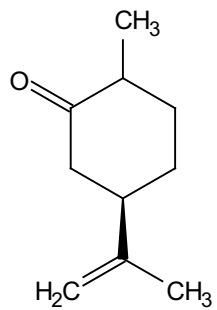
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—109.57

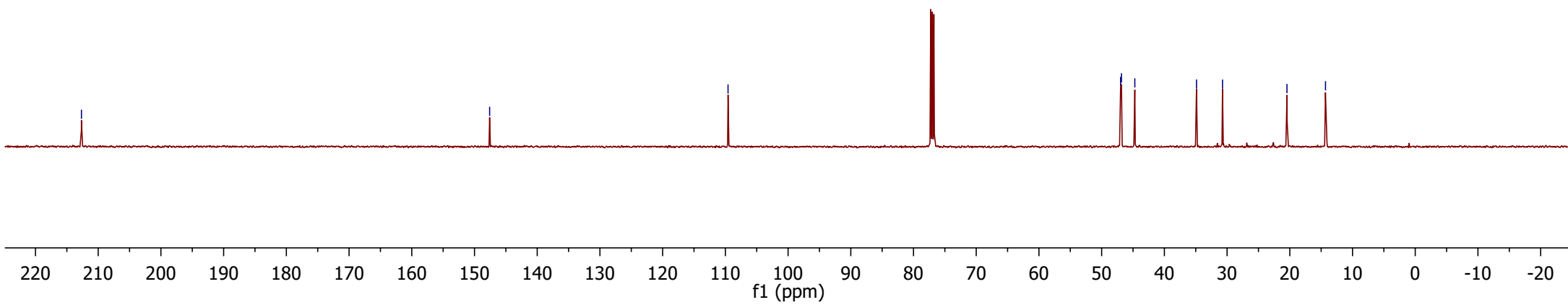
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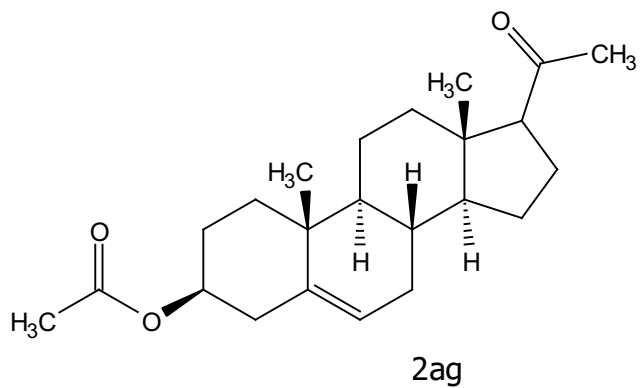
—34.87
—30.73

—20.45
—14.31

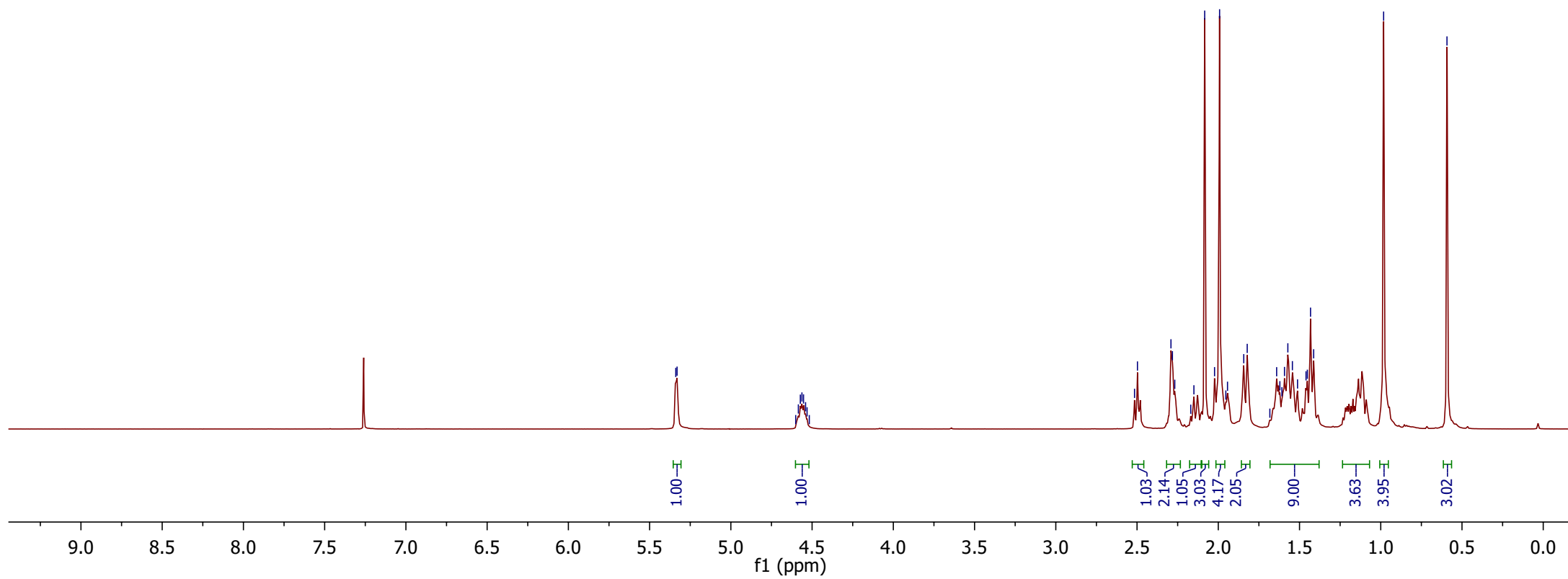


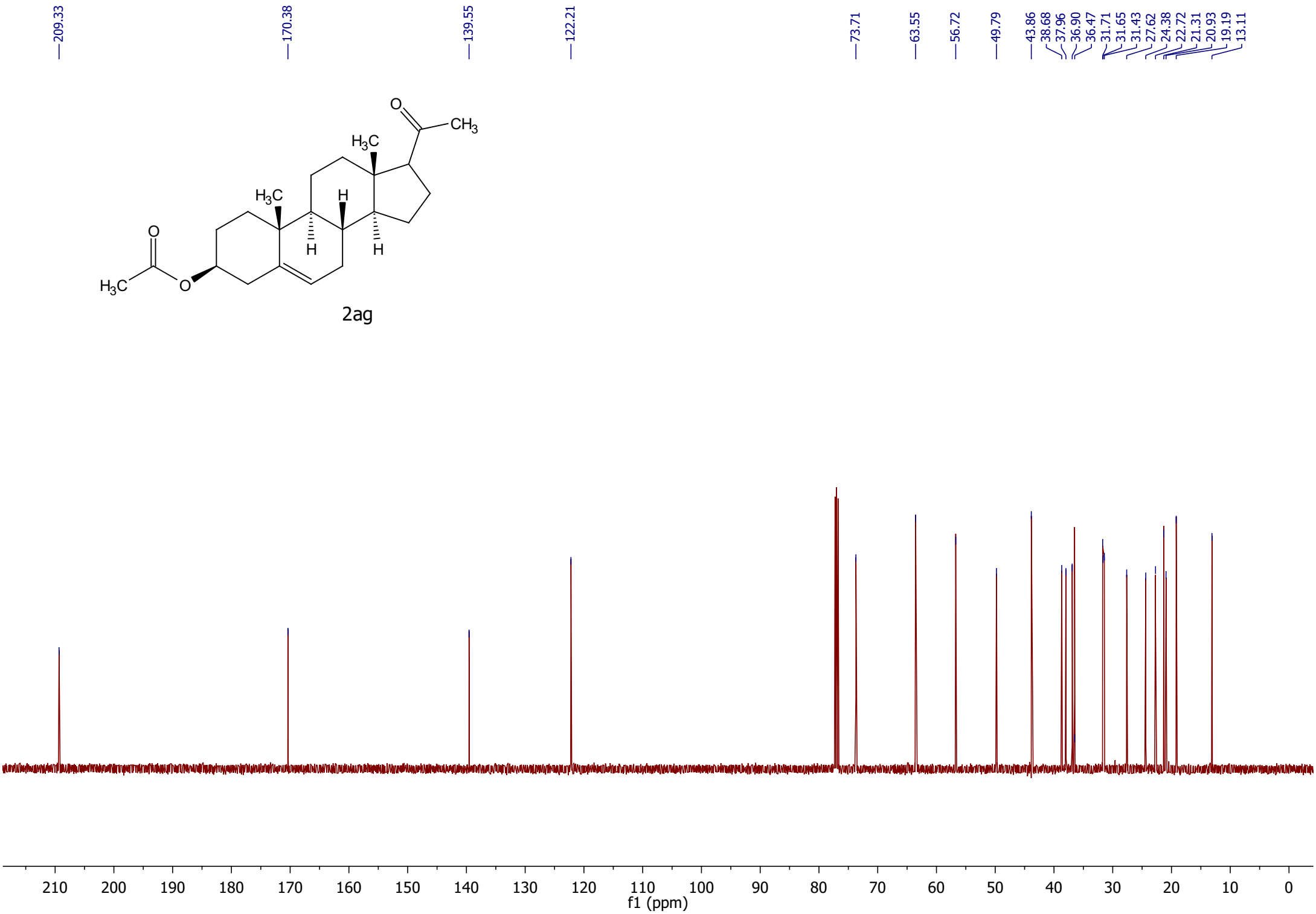
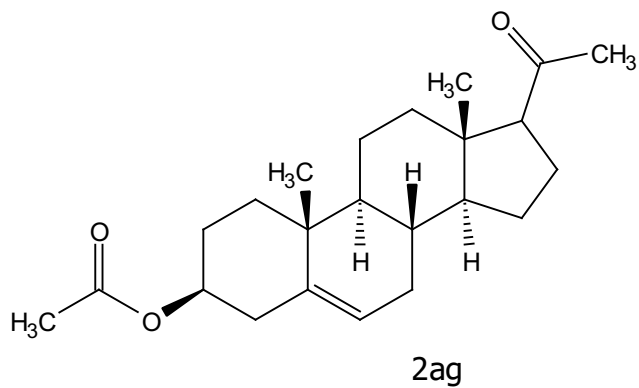
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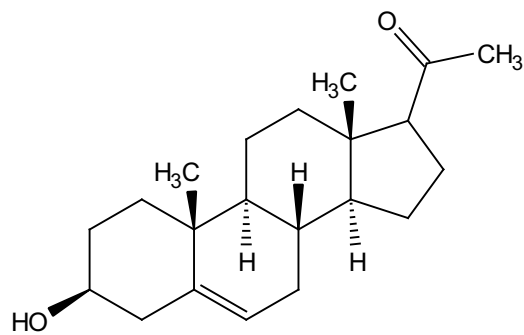




5.34
5.33
4.60
4.58
4.57
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4.54
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4.52
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2.50
2.29
2.28
2.27
2.17
2.15
2.08
2.02
1.99
1.95
1.94
1.84
1.82
1.68
1.64
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0.98
0.59



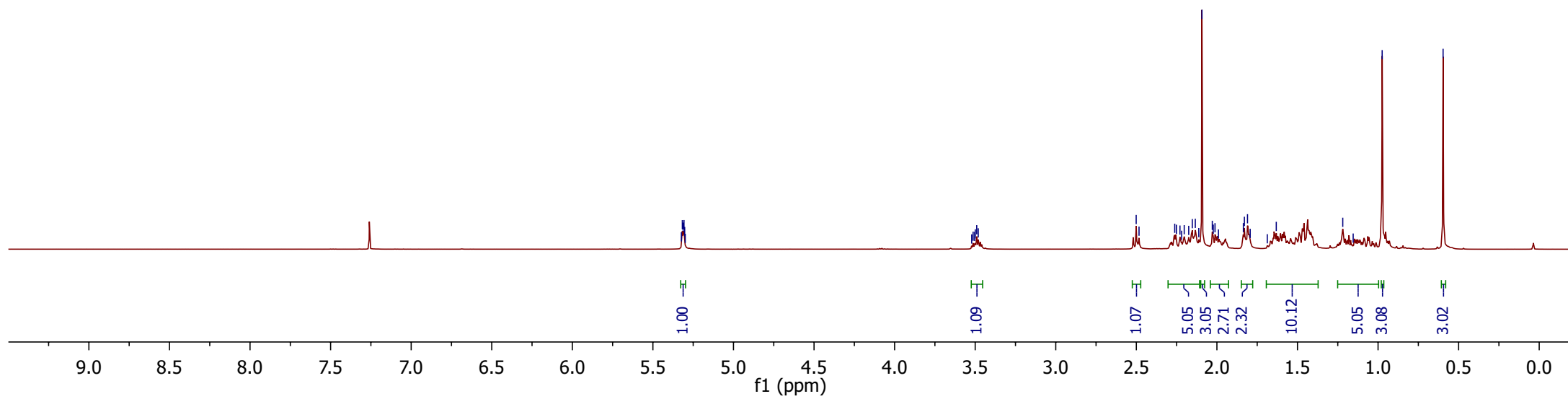




2ah

5.32
5.32
5.31
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5.30
5.30

3.52
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3.50
3.49
3.48
2.50
2.48
2.26
2.25
2.23
2.22
2.20
2.17
2.15
2.13
2.11
2.09
2.03
2.02
2.01
1.99
1.84
1.83
1.81
1.79
1.69
1.63
1.22
1.15
0.97
0.60



—209.64

—140.73

—121.22

—71.51

—63.60

—56.81

—49.88

43.92

42.11

38.74

37.17

36.42

31.76

31.68

31.46

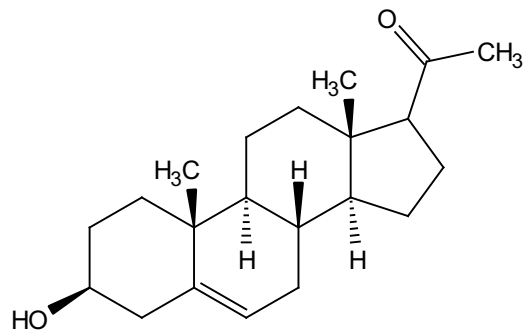
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22.72

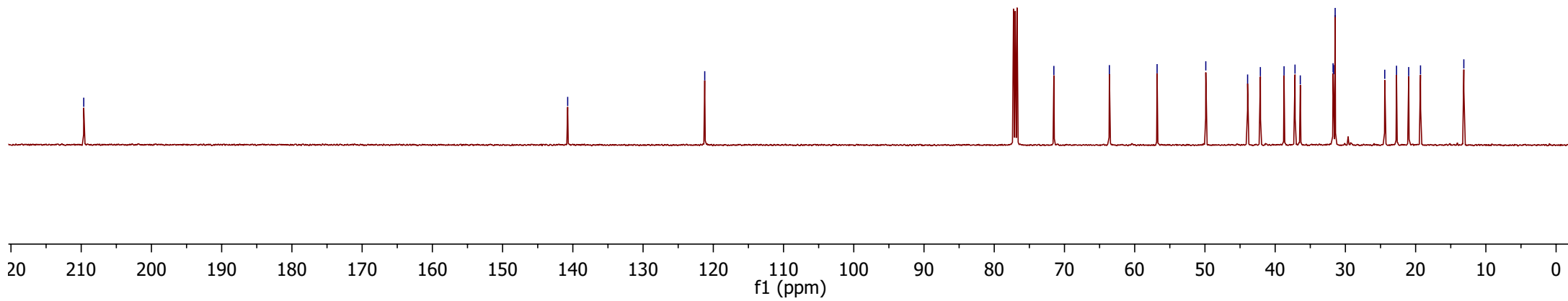
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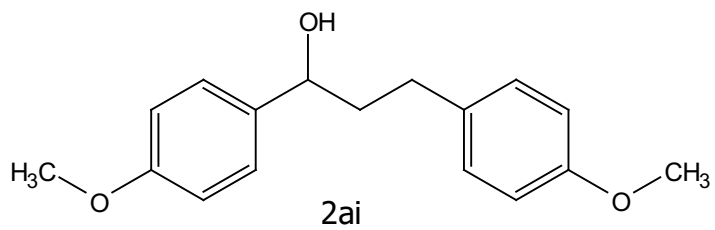
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—13.14



2ah

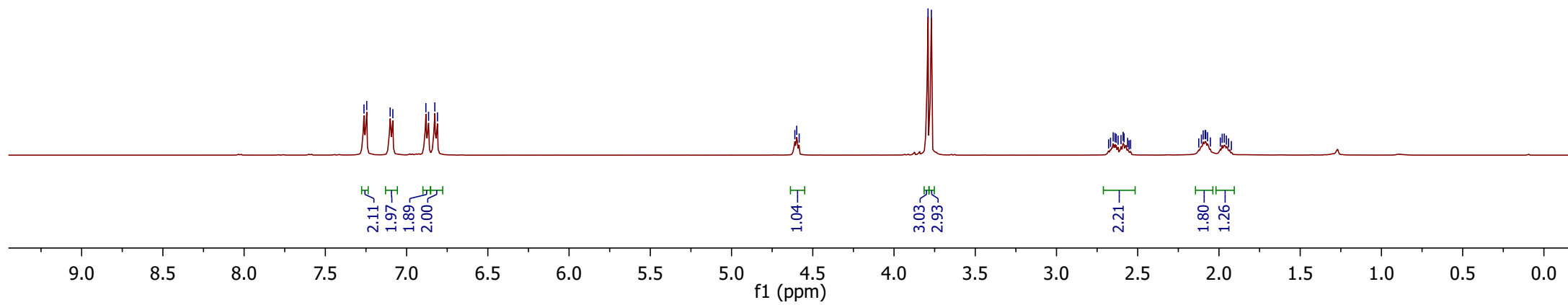


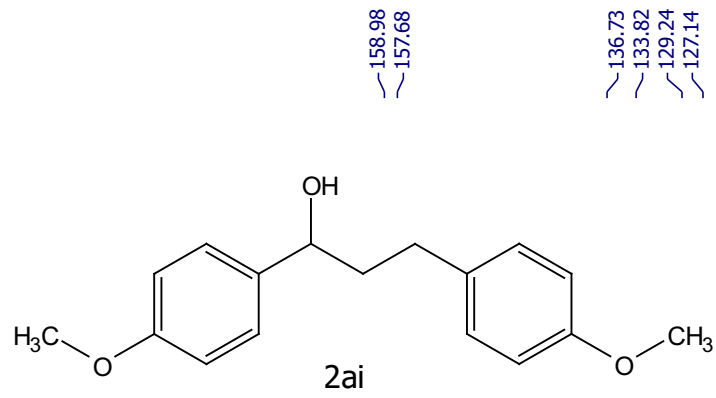


7.26
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6.86
6.83
6.81

4.61
4.60
4.58

3.79
3.77
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1.94
1.92





158.98
157.68

136.73
133.82
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127.14

113.78
113.73

73.34

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55.18

40.51

31.12

