Supplementary Information

Iodine-Catalyzed Cyclization-Allylation of *N*-Allyl-2-alkynylanilines via Iodocyclization-Rearrangement-Deiodination Sequence

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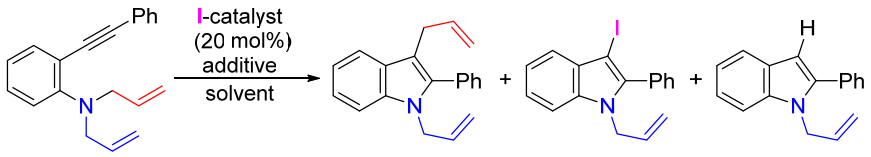
1. Optimization of Reaction Conditions

Table S1. Evaluation of iodine catalysts, additives and solvents



1a **2a** **3a** **5a**

entry	I-catalyst	additive (equiv.)	Solvent	(°C)	(h)	**2a**[a] (%)	**3a**[a] (%)	**5a**[a] (%)	**1a**[a] (%)
1	I_2		DCE	60	24	9	19	trace	58
2	I_2		toluene	60	24	2	15	0	74
3	I_2		MeCN	60	24	17	6	9	13
4	I_2		DMF	60	24	28	7	10	21
5	I_2		DMP	60	24	44	5	20	10
6	I_2		$MeNO_2$	60	24	41	4	12	6
7	I_2		$MeNO_2$	0/40	24	49	ND	15	16
8	I_2		$MeNO_2$	rt	24	29	8	0	53
9	I_2	NMP (5.2)	$MeNO_2$	40	24	62	14	14	0
10	NIS		$MeNO_2$	60	24	33	ND	0	49
11	ICl		$MeNO_2$	60	24	48	trace	13	0
12	Py_2IBF_4		$MeNO_2$	60	24	50	trace	16	0
13	Py_2IBF_4		$MeNO_2$	40	24	30	0	6	36
14	-	$HBF_4{\cdot}OEt_2$ (0.2)	$MeNO_2$	40	24	4	0	0	90
15	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	$MeNO_2$	40	24	69	6	3	0
16	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	Toluene	40	24	8	2	ND	ND
17	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	THF	40	24	50	ND	ND	ND
18	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	MeCN	40	24	38	4	6	42
19	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	MeCN	40	48	66	5	ND	9
20	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	DCE	40	24	45	5	ND	44
21	Py_2IBF_4	$HBF_4{\cdot}OEt_2$ (0.2)	DCM	40	48	40	4	2	30
22	Py_2IBF_4[b]	$HBF_4{\cdot}OEt_2$ (0.3)	DCM	40	24	54	10	ND	19

DCE = 1,2-dicloroethane, DMF = *N*,*N*-dimethylformamide, DMP = *N*-methylpyrrolidone, THF = tetrahedrofuran, DCM = dicloromethane, Py = pyridine.

[a] Determined by ^{1}H NMR analysis using an internal standard. ND = Not determined. [b] 30 mol%.

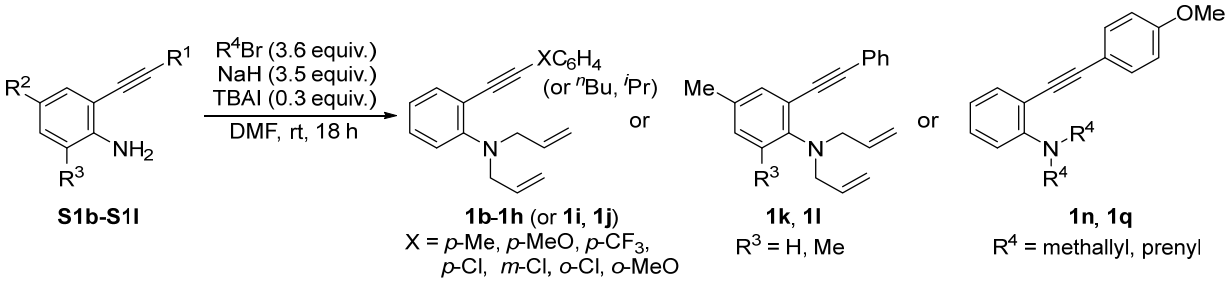
2. General Information

All reactions were carried out under an argon atmosphere. According to procedures reported in the literatures, *o*-alkynylanilines **1a** were prepared.[1] Molecular iodine, Iodine monochloride, *N*-iodosuccinimide (NIS), Barluenga's reagent (Py_2IBF_4, Py = pyridine) and $HBF_4{\cdot}OEt_2$ are commercially available. All solvents were purchased as the "anhydrous" and used without further purification. For the thin-layer chromatography (TLC) analysis, Merck precoated TLC plates (silica gel 60 F_{254}) were used. Column chromatography was performed on silica gel 60N (63-200 μm, neutral, Kanto Kagaku Co., Ltd.). Preparative thin layer chromatography (PTLC) was performed on Wakogel® B-5F (FUJIFILM Wako Pure Chemical Corp.). Medium pressure liquid chromatography (MPLC) was carried out with YAMAZEN EPCLC-Wprep 2XY.

^{1}H and ^{13}C NMR spectra were measured at 500 and 125 MHz in $CDCl_3$ and the chemical shifts are given in ppm using $CHCl_3$ (7.26 ppm) in $CDCl_3$ for ^{1}H NMR and $CDCl_3$ (77.0 ppm) for ^{13}C NMR as an internal standard, respectively. ^{19}F NMR spectra were measured at 470 MHz in CD_3Cl and the chemical shifts are given in ppm using C_6F_6 (-162.90 ppm) as an internal standard. Splitting patterns of an apparent multiplet associated with an averaged coupling constant were designed as s (singlet), d (doublet), t (triplet), q (quartet), m (multiplet), and br (broadened). Mass spectra and HRMS were recorded on double-focusing magnetic sector by FAB or ESI methods.

3. Preparation and Characterization of N,N-disubstituted o-alkynylanilines 1

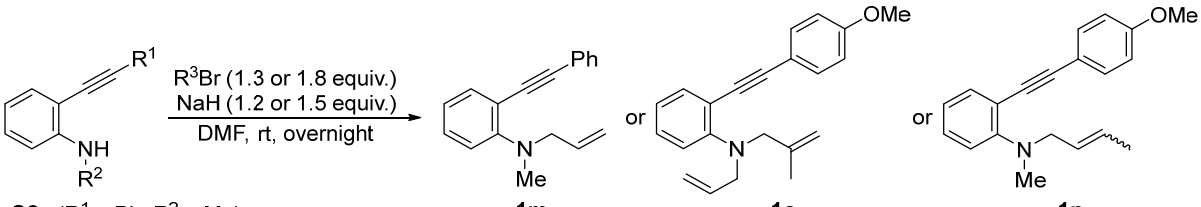
General procedure **A** (GP-**A**)



S1b-S1l → **1b-1h** (or **1i**, **1j**) X = *p*-Me, *p*-MeO, *p*-CF_3, *p*-Cl, *m*-Cl, *o*-Cl, *o*-MeO; **1k**, **1l** R^3 = H, Me; **1n**, **1q** R^4 = methallyl, prenyl

To a suspension of NaH (60% in oil, 3.5 equiv.) in DMF was added **S1b-S1l**[2] (0.985-8.11 mmol) in DMF at 0 ºC. After being stirred for 30 min at ambient temperature, allylic bromide R^4Br (R^4 = allyl, methallyl or prenyl, 3.6 equiv.) and tetrabutylammonium iodide (0.3 equiv.) was added. After being stirred at ambient temperature for 18h, the reaction mixture was quenched with sat. NH_4Cl aq. and 20 wt% $Na_2S_2O_3$ aq., and exacted with AcOEt. The organic layer was dried over $MgSO_4$ and concentrated in vacuo to dryness. In the reaction with **S1b**, **S1h** and **S1i**, since NMR analysis of the crude products indicated that the monoallylated products still remained, the crude products were treated with allyl bromide (1.2 equiv.) in a same manner to complete the allylation. The residue was purified by silica gel column chromatography (hexane:CH_2Cl_2 = 10:1) to give **1b-1l**, **1n** and **1q**.

General procedure **B** (GP-**B**)



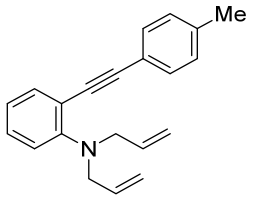
S2a (R^1 = Ph, R^2 = Me)
S2b (R^1 = 4-$MeOC_6H_4$, R^2 = allyl)
S2c (R^1 = 4-$MeOC_6H_4$, R^2 = Me)

→ **1m**, **1o**, **1p**

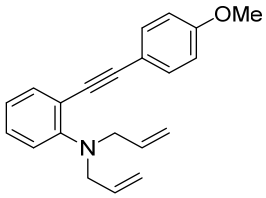
To a suspension of NaH (60% in oil, 1.5 equiv. for **S2a** and **S2c** or 1.2 equiv. for **S2b**) in DMF was added **S2a-S2c**[3] (1.64-3.14 mmol) in DMF at 0 ºC. After being stirred for 30 min at ambient temperature, allylic bromide R^3Br (R^3 = allyl or crotyl, 1.8 equiv.; R^3 = methallyl, 1.3 equiv.) was added 0 ºC. After being stirred at ambient temperature overnight, the reaction mixture was quenched with sat. NH_4Cl aq. and exacted with AcOEt. The organic layer was dried over $MgSO_4$ and concentrated in vacuo to dryness. The residue was purified by silica gel column chromatography (hexane:CH_2Cl_2 = 10:1) to give **1m**, **1o**

[1] S.-L. Niu, J. Hu, K. He, Y.-C. Chen and Q. Xiao, *Org. Lett.*, **2019**, *21*, 4250–4254.
[2] (*a*) C. Peng, Y. Wang, L. Liu, H. Wang, J. Zhao and Q. Zhu, *Eur. J. Org. Chem.*, **2010**, 818–822. (*b*) H. Liang, G. Zhu, X. Pu and L. Qiu, *Org. Lett.*, **2021**, *23*, 9246–9250. (*c*) A. S. K. Raj, A. S. Narode and R.-S. Liu, *Org. Lett.*, **2021**, *23*, 1378–1382. (*d*) J. I. Murray, N. J. Flodén, A. Bauer, N. D. Fessner, D. L. Dunklemann, O. Bob-Egbe, H. S. Rzepa, T. Bürgi, J. Richardson and A. C. Spivey, *Angew. Chem. Int. Ed.*, **2017**, *56*, 5760–5764.
[3] (*a*) L. Zhou, X. Liu, H. Lu, G. Deng, Y. Liang, Y. Yang and J.-H. Li, *Org. Chem. Front.*, **2021**, *8*, 5092–5097. (*b*) M. Mandal and R. Balamurugan, *Chem. Commun.*, **2022**, *58*, 9778–9781.

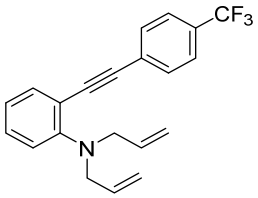
and **1p**.



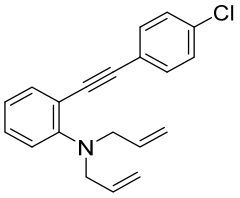
***N*,*N*-Diallyl-2-(*p*-tolylethynyl)aniline** (**1b**): 85% (1.34 g from **S1b** 1.14 g, GP-**A**). R_f = 0.35 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2209, 1591, 1511, 1485, 1441, 1276, 1216. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.56-7.52 (m, 2H), 7.39-7.30 (m, 4H), 7.04 (dd, *J* = 8.0, 1.7 Hz, 1H), 6.86 (d, *J* = 8.6 Hz, 1H), 5.92 (ddt, *J* = 17.2, 10.3, 5.5 Hz, 2H), 5.25 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.17 (dd, *J* = 10.3, 1.7 Hz, 2H), 3.94 (d, *J* = 5.5 Hz, 4H), 2.29 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 150.1, 135.4, 134.6, 131.4, 130.1, 129.6, 128.2, 127.8, 123.9, 119.7, 117.1, 115.9, 94.0, 88.9, 54.6, 20.3. HRMS (ESI): *m/z* calcd. for $C_{21}H_{22}N^+$ $[M+H]^+$ 288.1747; found 288.1750.



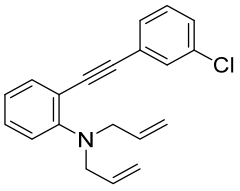
***N*,*N*-Diallyl-2-[(4-methoxyphenyl)ethynyl]aniline** (**1c**): 43% (1.06 g from **S1c** 1.81 g, GP-**A**). R_f = 0.28 (hexane: AcOEt = 20:1). Yellow solid. Mp 43-44 °C. IR (KBr) ν cm^{-1}; 2208, 1590, 1513, 1490, 1413, 1288, 1248, 1223, 1032. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.49-7.44 (m, 3H), 7.19 (ddd, *J* = 7.3, 7.3, 1.7 Hz, 1H), 6.92 (d, *J* = 8.0 Hz, 1H), 6.90-6.84 (m, 3H), 5.91 (ddt, *J* = 17.2, 10.3, 6.3 Hz, 2H), 5.23 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.17 (dd, *J* = 10.3, 1.7 Hz, 2H), 3.97 (d, *J* = 6.3 Hz, 4H), 3.83 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.3, 152.2, 135.3, 134.3, 132.8, 128.5, 120.5, 119.3, 117.2, 116.0, 115.8, 113.9, 94.3, 87.5, 55.3, 54.2. HRMS (ESI): *m/z* calcd. for $C_{21}H_{22}NO^+$ $[M+H]^+$ 304.1696; found 304.1703.



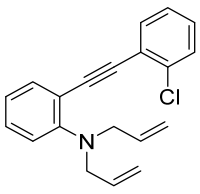
***N*,*N*-Diallyl-2-{[4-(trifluoromethyl)phenyl]ethynyl}aniline** (**1d**): 79% (1.78 g from **S1d** 1.72 g, GP-**A**). R_f = 0.30 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2211, 1614, 1593, 1486, 1416, 1321, 1277, 1219. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.65-7.58 (m, 4H), 7.51 (dd, *J* = 7.6, 1.7 Hz, 1H), 7.25 (ddd, *J* = 8.2, 7.5, 1.7 Hz, 1H), 6.95 (d, *J* = 8.2 Hz, 1H), 6.89 (ddd, *J* = 7.6, 7.5, 1.2 Hz, 1H), 5.91 (ddt, *J* = 17.2, 10.3, 5.7 Hz, 2H), 5.26 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.20 (dd, *J* = 10.3, 1.7 Hz, 2H), 3.98 (d, *J* = 5.7 Hz, 4H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.6, 135.0, 134.7, 131.5, 129.48 (q, $^2J_{C\text{-}F}$ = 32.4 Hz), 129.46, 127.7, 125.2 (q, $^3J_{C\text{-}F}$ = 3.6 Hz), 124.0 (q, $^1J_{C\text{-}F}$ = 272.3 Hz), 120.4, 119.2, 117.3, 114.4, 92.8, 91.6, 54.3. ^{19}F-NMR (470 MHz, CD_3Cl) δ ppm; -63.9 (s, 3F). HRMS (ESI): *m/z* calcd. for $C_{21}H_{19}F_3N^+$ $[M+H]^+$ 342.1464; found 342.1465.



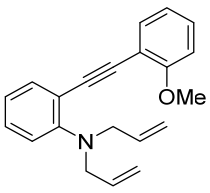
***N*,*N*-Diallyl-2-[(4-chlorophenyl)ethynyl]aniline** (**1e**): 81% (1.30 g from **S1e** 1.19 g, GP-**A**). R_f = 0.28 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2212, 1592, 1492, 1470, 1439, 1416, 1277, 1218, 1054. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.50 (dd, *J* = 7.5, 1.7 Hz, 1H), 7.46 (d, *J* = 8.6 Hz, 2H), 7.33 (d, *J* = 8.6 Hz, 2H), 7.24 (ddd, *J* = 7.5, 7.5, 1.7 Hz, 1H), 6.95 (d, *J* = 7.5 Hz, 1H), 6.90 (dd, *J* = 7.5, 7.5 Hz, 1H), 5.92 (ddt, *J* = 17.2, 9.7, 5.7 Hz, 2H), 5.26 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.20 (d, *J* = 9.7, 1.7 Hz, 2H), 3.99 (d, *J* = 5.7 Hz, 4H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.4, 135.1, 134.5, 133.8, 132.5, 129.1, 128.6, 122.4, 120.4, 119.2, 117.3, 114.9, 93.1, 90.0, 54.2. HRMS (ESI): *m/z* calcd. for $C_{20}H_{19}ClN^+$ $[M+H]^+$ 308.1201; found 308.1209.



***N*,*N*-Diallyl-2-[(3-chlorophenyl)ethynyl]aniline** (**1f**): 83% (1.77 g from **S1f** 1.57 g, GP-**A**). R_f = 0.33 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2217, 1591, 1491, 1473, 1440, 1416, 1276, 1218, 1089. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.53 (d, *J* = 1.4 Hz, 1H), 7.50 (ddd, *J* = 7.7, 1.4, 1.4 Hz, 1H), 7.41 (ddd, *J* = 7.2, 1.4, 1.4 Hz, 1H), 7.34-7.27 (m, 2H), 7.24 (ddd, *J* = 7.2, 1.4, 1.4 Hz, 1H), 6.95 (d, *J* = 8.3 Hz, 1H), 6.90 (dd, *J* = 7.5, 7.5 Hz, 1H), 5.92 (ddt, *J* = 17.2, 10.3, 6.0 Hz, 2H), 5.27 (ddt, *J* = 17.2, 1.4 Hz, 2H), 5.21 (ddt, *J* = 10.3, 1.4 Hz, 2H), 3.99 (d, *J* = 6.0 Hz, 4H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.5, 135.0, 134.6, 134.1, 131.1, 129.5, 129.2, 128.1, 125.6, 120.4, 119.2, 117.3, 114.7, 92.8, 90.3, 54.2 (note that two carbon peaks overlap with each other). HRMS (ESI): *m/z* calcd. for $C_{20}H_{19}ClN^+$ $[M+H]^+$ 308.1201; found 308.1207.

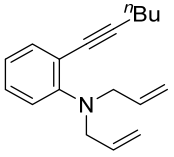


***N*,*N*-Diallyl-2-[(2-chlorophenyl)ethynyl]aniline** (**1g**): 97% (1.57 g from **S1g** 1.20 g, GP-**A**). R_f = 0.30 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2213, 1593, 1491, 1470, 1439, 1416, 1277, 1217, 1053. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.59-7.53 (m, 2H), 7.46-7.41 (m, 1H), 7.28-7.22 (m, 3H), 6.94 (d, *J* = 7.5 Hz, 1H), 6.90 (ddd, *J* = 7.5, 7.5, 1.2 Hz, 1H), 5.90 (ddt, *J* = 17.2, 10.3, 5.7 Hz, 2H), 5.23 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.17 (dd, *J* = 10.3, 1.7 Hz, 2H), 4.01 (d, *J* = 5.7 Hz, 4H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.3, 135.6, 135.0, 134.8, 133.0, 129.3, 129.2, 128.9, 126.4, 123.8, 120.4, 119.4, 117.2, 115.0, 94.1, 91.1, 54.3. HRMS (ESI): *m/z* calcd. for $C_{20}H_{19}ClN^+$ $[M+H]^+$ 308.1201; found 308.1202.

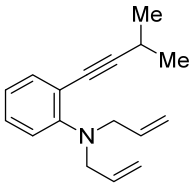


***N*,*N*-Diallyl-2-[(2-methoxyphenyl)ethynyl]aniline** (**1h**): 66% (0.477 g from **S1h** 0.534 g, GP-**A**). R_f = 0.26 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2208, 1591, 1496, 1484, 1434, 1274, 1247, 1217, 1025. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.52 (dd, *J* = 7.5, 1.7 Hz, 1H), 7.49 (dd, *J* = 7.5, 1.7 Hz, 1H), 7.29 (ddd, *J* = 7.5, 7.5, 1.7 Hz, 1H), 7.20 (ddd, *J* = 8.6, 7.5, 1.7 Hz, 1H), 6.96-6.86 (m, 4H), 5.92 (ddt, *J* = 17.2, 10.3, 5.7 Hz, 2H), 5.22 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.15 (dd, *J* = 10.3, 1.7 Hz, 2H), 4.01 (d, *J* = 5.7 Hz, 4H), 3.90 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ

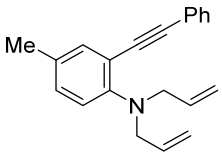
ppm; 159.8, 152.2, 135.4, 134.6, 133.2, 129.3, 128.7, 120.4, 119.4, 117.0, 115.9, 113.1, 110.6, 92.9, 90.8, 55.6, 54.2 (note that two carbon peaks overlap with each other). HRMS (ESI): *m/z* calcd. for $C_{21}H_{22}NO^+$ $[M+H]^+$ 304.1696; found 304.1696.



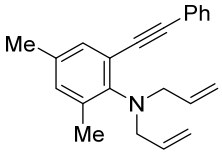
***N*,*N*-Diallyl-2-(hex-1-yn-1-yl)aniline** (**1i**): 62% (0.902 g from **S1i** 1.00 g, GP-**A**). R_f = 0.29 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2227, 1612, 1508, 1464, 1431, 1252, 1179. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.37 (dd, *J* = 7.4, 1.7 Hz, 1H), 7.16 (ddd, *J* = 8.0, 7.4, 1.7 Hz, 1H), 6.89 (d, *J* = 8.0 Hz, 1H), 6.84 (ddd, *J* = 7.4, 7.4, 1.2 Hz, 1H), 5.87 (ddt, *J* = 17.4, 10.3, 5.7 Hz, 2H), 5.21 (dd, *J* = 17.4, 1.4 Hz, 2H), 5.16 (dd, *J* = 10.3, 1.4 Hz, 2H), 3.91 (d, *J* = 5.7 Hz, 4H), 2.48 (t, *J* = 7.2 Hz, 2H), 1.67-1.58 (m, 2H), 1.55-1.46 (m, 2H), 0.96 (t, *J* = 7.4 Hz, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.3, 135.4, 134.3, 127.9, 120.6, 119.5, 117.0, 116.9, 95.5, 79.6, 54.2, 30.9, 22.1, 19.6, 13.7. HRMS (ESI): *m/z* calcd. for $C_{18}H_{24}N^+$ $[M+H]^+$ 254.1903; found 254.1905.



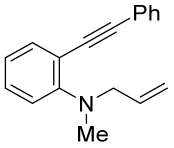
***N*,*N*-Diallyl-2-(3-methylbut-1-yn-1-yl)aniline** (**1j**): 61% (0.734 g from **S1j** 0.796 g, GP-**A**). R_f = 0.41 (hexane: AcOEt = 20:1). Colorless oil. IR (neat) ν cm^{-1}; 2224, 1592, 1488, 1442, 1411, 1321, 1214. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.37 (dd, *J* = 7.4, 1.7 Hz, 1H), 7.16 (ddd, *J* = 7.9, 7.6, 1.7 Hz, 1H), 6.89 (d, *J* = 7.9 Hz, 1H), 6.83 (dd, *J* = 7.6, 7.4 Hz, 1H), 5.88 (ddt, *J* = 17.4, 10.3, 6.0 Hz, 2H), 5.22 (dd, *J* = 17.4, 1.4 Hz, 2H), 5.16 (dd, *J* = 10.3, 1.4 Hz, 2H), 3.91 (d, *J* = 6.0 Hz, 4H), 2.84 (septet, *J* = 6.9 Hz, 1H), 1.29 (d, *J* = 6.9 Hz, 6H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 152.2, 135.4, 134.4, 127.9, 120.5, 119.4, 116.9, 116.6, 100.7, 78.9, 54.0, 23.0, 21.5. HRMS (ESI): m/z calcd. for $C_{17}H_{22}N^+$ $[M+H]^+$ 240.1747; found 240.1751.



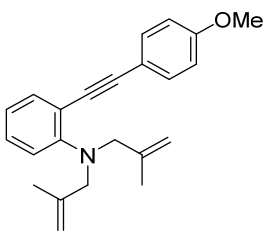
***N*,*N*-Diallyl-4-methyl-2-(phenylethynyl)aniline** (**1k**): 87% (1.25 g from **S1k** 1.04 g, GP-**A**). R_f = 0.33 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2202, 1598, 1499, 1488, 1442, 1416, 1277, 1216. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.57-7.53 (m, 2H), 7.39-7.30 (m, 4H), 7.04 (dd, *J* = 8.3, 1.7 Hz, 1H), 6.86 (d, *J* = 8.3 Hz, 1H), 5.92 (ddt, *J* = 17.2, 10.3, 5.7 Hz, 2H), 5.25 (dd, *J* = 17.2, 1.7 Hz, 2H), 5.17 (dd, *J* = 10.3, 1.7 Hz, 2H), 3.94 (d, *J* = 5.7 Hz, 4H), 2.29 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 150.1, 135.4, 134.6, 131.4, 130.1, 129.6, 128.3, 127.9, 123.9, 119.7, 117.1, 115.9, 94.0, 88.9, 54.6, 20.3. HRMS (ESI): *m/z* calcd. for $C_{21}H_{22}N^+$ $[M+H]^+$ 288.1747; found 288.1751.



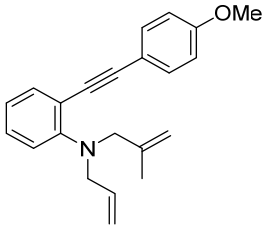
***N*,*N*-Diallyl-2,4-dimethyl-6-(phenylethynyl)aniline** (**1l**): 71% (1.10 g from **S1l** 1.13 g, GP-**A**). R_f = 0.35 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2208, 1598, 1492, 1472, 1442, 1415, 1268, 1216. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.59-7.55 (m, 2H), 7.42-7.33 (m, 3H), 7.18 (d, *J* = 2.3 Hz, 1H), 7.01 (s, 1H), 5.90 (ddt, *J* = 17.1, 10.5, 6.3 Hz, 2H), 5.18 (dd, *J* = 17.1, 1.7 Hz, 2H), 5.04 (dd, *J* = 10.5, 1.7 Hz, 2H), 3.87 (d, *J* = 6.3 Hz, 4H), 2.32 (s, 3H), 2.28 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 148.5, 137.8, 136.7, 134.1, 132.04, 132.00, 131.0, 128.4, 128.0, 123.9, 122.0, 116.2, 93.1, 89.5, 56.2, 20.6, 18.9. HRMS (ESI): *m/z* calcd. for $C_{22}H_{24}N^+$ $[M+H]^+$ 302.1903; found 302.1911.



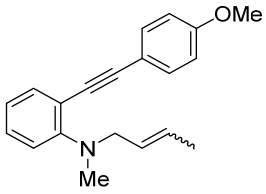
***N*-Allyl-*N*-methyl-2-(phenylethynyl)aniline** (**1m**): 96% (0.420 g from **S2a** 0.368 g, GP-**B**). R_f = 0.31 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2209, 1590, 1496, 1482, 1443, 1276, 1228, 1185. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.57-7.53 (m, 2H), 7.51 (dd, *J* = 8.0, 1.7 Hz, 1H), 7.38-7.30 (m, 3H), 7.26 (ddd, *J* = 7.7, 7.7, 1.7 Hz, 1H), 6.95 (d, *J* = 7.7 Hz, 1H), 6.91 (ddd, *J* = 7.7, 7.7, 1.2 Hz, 1H), 6.03 (ddt, *J* = 17.2, 10.3, 6.3 Hz, 1H), 5.29 (ddt, *J* = 17.2, 1.7 Hz, 1H), 5.21 (dd, *J* = 10.3, 1.7 Hz, 1H), 3.99 (d, *J* = 6.3 Hz, 2H), 2.87 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 153.9, 135.6, 134.4, 131.4, 129.2, 128.3, 127.9, 123.8, 120.4, 117.7, 117.3, 115.1, 94.6, 88.7, 59.2, 38.8. HRMS (ESI): *m/z* calcd. for $C_{18}H_{18}N^+$ $[M+H]^+$ 248.1434; found 248.1435.



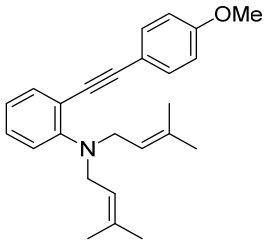
2-[(4-Methoxyphenyl)ethynyl]-*N*,*N*-bis(2-methylallyl)aniline (**1n**): 83% (0.271 g from **S1c** 0.220 g, GP-**A**). R_f = 0.29 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2211, 1606, 1512, 1486, 1441, 1287,1249, 1214, 1033. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.48 (dd, *J* = 7.5, 1.6 Hz, 1H), 7.46 (d, *J* = 8.9 Hz, 2H), 7.20 (ddd, *J* = 7.9, 7.5, 1.6 Hz, 1H), 6.94 (d, *J* = 7.9 Hz, 1H), 6.90 (d, *J* = 8.9 Hz, 2H), 6.88 (dd, *J* = 7.5, 7.5 Hz, 1H), 4.99 (s, 2H), 4.90 (s, 2H), 3.90 (s, 4H), 3.84 (s, 3H), 1.74 (s, 6H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.3, 152.5, 142.5, 134.3, 132.6, 128.4, 120.3, 119.5, 116.2, 115.7, 113.9, 112.3, 93.8, 87.7, 57.8, 55.2, 20.6. HRMS (ESI): *m/z* calcd. for $C_{23}H_{26}NO^+$ $[M+H]^+$ 332.2009; found 332.2010.



***N*-Allyl-2-[(4-methoxyphenyl)ethynyl]-*N*-(2-methylallyl)aniline** (**1o**): Quant. (0.532 g from **S2b** 0.432 g, GP-**B**). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2212, 1606, 1512, 1486, 1441, 1287,1249, 1215, 1033. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.47 (d, *J* = 7.6 Hz, 1H), 7.45 (d, *J* = 8.6 Hz, 2H), 7.19 (ddt, *J* = 7.9, 7.7, 1.2 Hz, 1H), 6.92 (d, *J* = 7.9 Hz, 1H), 6.88 (d, *J* = 8.6 Hz, 2H), 6.87 (dd, *J* = 7.7, 7.6 Hz, 1H), 5.91 (ddt, *J* = 17.2, 10.6, 6.0 Hz, 1H), 5.20 (d, *J* = 17.2 Hz, 1H), 5.15 (d, *J* = 10.6 Hz, 1H), 4.98 (s, 1H), 4.90 (s, 1H), 3.98 (d, *J* = 6.0 Hz, 2H), 3.83 (s, 5H), 1.73 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.3, 152.3, 142.4, 135.3, 134.2, 132.7, 128.4, 120.5, 119.6, 117.1, 116.2, 116.0, 113.9, 112.3, 94.0, 87.6, 57.1, 55.3, 54.9, 20.5. HRMS (ESI): *m/z* calcd. for $C_{22}H_{24}NO^+$ $[M+H]^+$ 318.1852; found 318.1850.

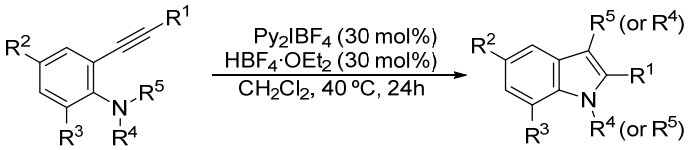


***N*-(But-2-en-1-yl)-2-[(4-methoxyphenyl)ethynyl]-*N*-methylaniline** (**1p**): 96% (0.880 g from **S2c** 0.746 g, GP-**B**). R_f = 0.23 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2210, 1606, 1512, 1487, 1447, 1287,1248, 1175, 1032. ^{1}H-NMR (500 MHz, $CDCl_3$, 83:17 mixture of geometrical isomers) δ ppm; 7.51-7.45 (m, 3H), 7.25-7.19 (m, 1H), 6.96-6.91 (m, 1H), 6.90-6.85 (m, 3H), 5.73-5.61 (m, 2H), 4.02 (d , *J* = 4.0 Hz, 0.34H), 3.89 (d, *J* = 4.0 Hz, 1.66H), 3.83 (s, 3H), 2.85 (s, 0.51H), 2.83 (s, 2.49H), 1.71 (d, *J* = 4.0 Hz, 2.49H), 1.67 (d, *J* = 5.2 Hz, 0.51H). ^{13}C-NMR (125 MHz, $CDCl_3$, 83:17 mixture of geometrical isomers) δ ppm; 159.3, 153.9, 134.13, 134.06 (minor), 132.7, 128.8, 128.4, 128.2, 127.8 (minor), 126.8 (minor), 120.6 (minor), 120.3, 117.8 (minor), 117.6, 116.0, 116.0 (minor), 115.9 (minor), 115.5, 113.9, 94.7 (minor), 94.5, 87.4, 87.3 (minor), 58.4, 55.2, 53.4 (minor), 52.6 (minor), 39.0 (minor), 38.6, 17.8, 13.0 (minor) [note that five sp^2 carbon peaks of the minor isomer overlap with the peaks of the major isomer]. HRMS (ESI): *m/z* calcd. for $C_{20}H_{22}NO^+$ $[M+H]^+$ 292.1696; found 292.1703.



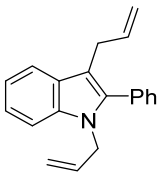
2-[(4-Methoxyphenyl)ethynyl]-*N*,*N*-bis(3-methylbut-2-en-1-yl)aniline (**1q**): 99% (1.80 g from **S1c** 1.13 g, GP-**A**). R_f = 0.31 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 2211, 1606, 1512, 1485, 1441, 1248, 1174. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.49-7.44 (m, 3H), 7.18 (ddd, *J* = 7.7, 7.7, 1.7 Hz, 1H), 6.90-6.83 (m, 4H), 5.32 (t, *J* = 6.3 Hz, 2H), 3.89 (d, *J* = 6.3 Hz, 4H), 3.83 (s, 3H), 1.71 (s, 6H), 1.64 (s, 6H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.3, 153.0, 134.2, 134.1, 132.8, 128.4, 122.3, 120.1, 119.1, 116.3, 116.1, 113.8, 94.3, 87.8, 55.3, 49.5, 25.8, 17.9. HRMS (ESI): *m/z* calcd. for $C_{25}H_{30}NO^+$ $[M+H]^+$ 360.2322; found 360.2319.

4. Preparation and Characterization of 3-allylindoles 2 and 4



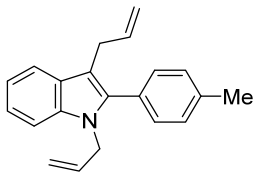
1a-1q **2a-2f**, **2h-2q**, **4p**, **4q**

After Barluenga's reagent (30 mol%) was treated with HBF_4·OEt_2 (30 mol%) in CH_2Cl_2 (1.5 mL) at 0 ºC for 15 min, a solution of **1a**-**1q** (0.49-0.60 mmol) in CH_2Cl_2 (3.0 mL) was added at 0 ºC. After being stirred at 40 ºC for 24 h, the reaction mixture was quenched with sat. $NaHCO_3$ aq. and 20 wt% $Na_2S_2O_3$ aq., and exacted with AcOEt. The organic layer was dried over $MgSO_4$ and concentrated in vacuo to dryness. The residue was purified by MPLC on silica gel modified with amino groups (hexane only) and by MPLC on silica gel modified with octadecylsilyl (ODS) groups (MeCN:H_2O = 9:1 to 1:0) in turn to give **2a-2f**, **2h-2q**, **4p** and **4q**.

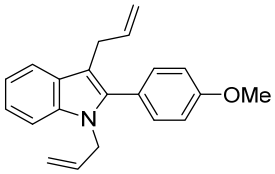


1,3-Diallyl-2-phenyl-1*H*-indole (**2a**): 53% (72.9 mg from **1a** 137.5 mg). R_f = 0.30 (hexane:AcOEt = 20:1). Yellow solid. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.66 (d, *J* = 8.0 Hz, 1H), 7.51-7.46 (m, 2H), 7.46-7.41 (m, 3H), 7.34 (d, *J* = 8.0 Hz, 1H), 7.25 (dd *J* = 8.0, 6.9 Hz, 1H), 7.17 (dd, *J* = 8.0, 6.9 Hz, 1H), 6.05 (ddt, *J* = 17.2, 9.7, 5.2 Hz, 1H), 5.93 (ddt, *J* = 16.9, 10.6, 4.6 Hz, 1H), 5.15 (dd, *J* = 10.6, 1.7 Hz, 1H), 5.08-5.01 (m, 2H), 4.93 (dd, *J* = 17.2, 1.2 Hz, 1H), 4.62 (ddd, *J* = 4.6, 1.7, 1.7 Hz, 2H), 3.47 (ddd, *J* = 5.2, 1.7, 1.7 Hz, 2H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 138.0, 137.9, 136.7, 133.9, 131.8, 130.4, 128.3, 128.1, 128.0, 121.7, 119.34, 119.30, 116.2, 114.6, 110.9, 110.1, 46.4, 29.2. ^{1}H and ^{13}C NMR spectra of **2a** were identical with those reported in literature.[4]

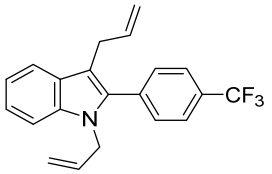
[4] M. E. Kieffer, L. M. Repka and S. E. Reisman, *J. Am. Chem. Soc.*, **2012**, *134*, 5131–5137.



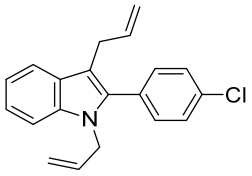
1,3-Diallyl-2-(*p*-tolyl)-1*H*-indole (**2b**): 58% (85.4 mg from **1b** 146.0 mg). R_f = 0.33 (hexane: AcOEt = 20:1). Yellow solid. Mp 41-42 °C. IR (KBr) ν cm^{-1}; 1462, 1435, 1415, 1358, 1189, 748. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.64 (d, *J* = 8.0 Hz, 1H), 7.33 (d, *J* = 8.0 Hz, 1H), 7.32 (d, *J* = 8.0 Hz, 2H), 7.28 (d, *J* = 8.0 Hz, 2H), 7.23 (dd, *J* = 8.0, 6.9 Hz, 1H), 7.15 (dd, *J* = 8.0, 6.9 Hz, 1H), 6.09 (ddt, *J* = 16.6, 10.5, 5.2 Hz, 1H), 5.92 (ddt, *J* = 16.6, 10.5, 5.2 Hz, 1H), 5.14 (d, *J* = 10.5 Hz, 1H), 5.06 (d, *J* = 16.6 Hz, 1H), 5.02 (d, *J* = 10.5 Hz, 1H), 4.93 (d, *J* = 16.6 Hz, 1H), 4.64-4.59 (m, 2H), 3.46 (d, *J* = 5.2 Hz, 2H), 2.45 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 138.1, 138.0, 137.9, 136.7, 134.0, 130.3, 129.0, 128.8, 128.1, 121.6, 119.3, 119.2, 116.2, 114.5, 110.7, 110.0, 46.4, 29.3, 21.3. HRMS (ESI): *m/z* calcd. for $C_{21}H_{20}N^+$ $[M-H]^+$ 286.1590; found 286.1594.



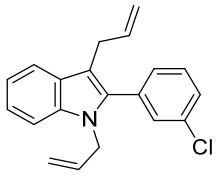
1,3-Diallyl-2-(4-methoxyphenyl)-1*H*-indole (**2c**)**:** 60% (93.8 mg from **1c** 155.7 mg). R_f = 0.28 (hexane: AcOEt = 20:1). Yellow solid. Mp 82-83 °C. IR (KBr) ν cm^{-1}; 1508, 1464, 1431, 1362, 1250, 1179, 1028,745. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.61 (d, *J* = 8.0 Hz, 1H), 7.33 (d, *J* = 8.6 Hz, 2H), 7.30 (d, *J* = 8.0 Hz, 1H), 7.21 (ddd, *J* = 8.0, 6.9, 1.2 Hz, 1H), 7.13 (dd, *J* = 8.0, 6.9 Hz, 1H), 6.99 (d, *J* = 8.6 Hz, 2H), 6.02 (ddt, *J* = 17.0, 10.1, 6.0 Hz,1H), 5.91 (ddt, *J* = 17.0, 10.1, 4.6 Hz, 1H), 5.12 (ddt, *J* = 10.1, 1.7, 1.7 Hz, 1H), 5.03 (ddt, *J* = 17.0, 1.7, 1.7 Hz, 1H), 5.00 (ddt, *J* = 10.1, 1.7, 1.7 Hz, 1H), 4.90 (ddt, *J* = 17.0, 1.7, 1.7 Hz, 1H), 4.59 (ddd, *J* = 4.6, 1.7, 1.7 Hz, 2H), 3.87 (s, 3H), 3.43 (ddd, *J* = 6.0, 1.7, 1.7 Hz, 2H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.5, 138.0, 137.8, 136.6, 134.0, 131.6, 128.0, 124.0, 121.5, 119.3, 119.2, 116.2, 114.5, 113.7, 110.6, 110.0, 55.3, 46.3, 29.3. HRMS (ESI): *m/z* calcd. for $C_{21}H_{20}NO^+$ $[M-H]^+$ 302.1539; found 302.1531.



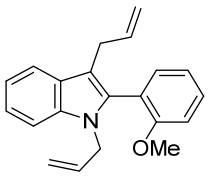
1,3-Diallyl-2-[4-(trifluoromethyl)phenyl]-1*H*-indole (**2d**)**:** 31% (53.7 mg from **1d** 172.8 mg). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow solid. Mp 55-56 °C. IR (KBr) ν cm^{-1}; 1462, 1436, 1420, 1361, 1327, 1161, 739. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.73 (d, *J* = 8.0 Hz, 2H), 7.65 (d, *J* = 8.0 Hz, 1H), 7.55 (d, *J* = 8.0 Hz, 2H), 7.33 (d, *J* = 8.0 Hz, 1H), 7.27 (ddd, *J* = 8.0, 7.3, 1.2 Hz, 1H), 7.17 (ddd, *J* = 8.0, 7.3, 1.2 Hz, 1H), 6.04 (ddt, *J* = 17.6, 9.6, 5.7 Hz, 1H), 5.92 (ddt, *J* = 172, 10.3, 4.6 Hz, 1H), 5.16 (ddt, *J* = 10.3, 1.7, 1.7 Hz, 1H), 5.05-5.00 (m, 2H), 4.90 (ddt, *J* = 17.2, 1.7, 1.7 Hz, 1H), 4.61 (ddd, *J* = 4.6, 1.9, 1.9 Hz, 2H), 3.44 (ddd, *J* = 5.7, 1.7, 1.7 Hz, 2H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 137.5, 137.1, 136.3, 135.5, 133.7, 130.6, 130.0 (q, $^2J_{C-F}$ = 32.5 Hz), 127.9, 125.3 ($^3J_{C-F}$ = 3.8 Hz), 124.1 (q, $^1J_{C-F}$ = 272.2 Hz), 122.4, 119.7, 119.6, 116.4, 114.9, 111.9, 110.2, 46.5, 29.1. ^{19}F-NMR (470 MHz, $CDCl_3$) δ ppm; -63.7 (s, 3F). HRMS (ESI): *m/z* calcd. for $C_{21}H_{17}F_3N^+$ $[M-H]^+$ 340.1308; found 340.1304.



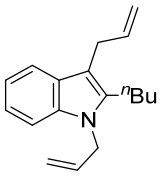
1,3-Diallyl-2-(4-chlorophenyl)-1*H*-indole (**2e**)**:** 39% (60.0 mg from **1e** 155.7 mg). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow solid. Mp 47-48 °C. IR (KBr) ν cm^{-1}; 1489, 1461, 1438, 1361, 1338, 1193, 1092, 1015, 750. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.63 (d, *J* = 8.0 Hz, 1H), 7.44 (d, *J* = 8.2 Hz, 2H), 7.35 (d, *J* = 8.2 Hz, 2H), 7.32 (d, *J* = 8.0 Hz, 1H), 7.25 (dd, *J* = 8.0, 7.8 Hz, 1H), 7.16 (dd, *J* = 8.0, 7.8 Hz, 1H), 6.02 (ddt, *J* = 17.2, 10.3, 6.0 Hz, 1H), 5.90 (ddt, *J* = 17.2, 10.3, 4.6 Hz, 1H), 5.14 (d, *J* = 10.3 Hz, 1H), 5.04-5.01 (m, 2H), 4.89 (d, *J* = 17.2 Hz, 1H), 4.59 (ddd, *J* = 4.6, 2.3, 2.3 Hz, 2H), 3.43 (d, *J* = 6.0 Hz, 2H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 137.7, 136.8, 136.6, 134.2, 133.8, 131.6, 130.2, 128.6, 127.9, 122.0, 119.5, 119.4, 116.3, 114.8, 111.3, 110.1, 46.4, 29.1. HRMS (ESI): *m/z* calcd. for $C_{20}H_{17}ClN^+$ $[M-H]^+$ 306.1044; found 306.1041.



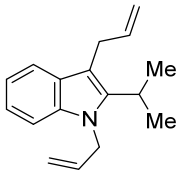
1,3-Diallyl-2-(3-chlorophenyl)-1*H*-indole (**2f**)**:** 26% (39.9 mg from **1f** 155.1 mg). R_f = 0.30 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 1460, 1439, 1361, 1193, 1118, 1078, 741. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.64 (d, *J* = 7.4 Hz, 1H), 7.45-7.37 (m, 3H), 7.32 (d, *J* = 7.4 Hz, 1H), 7.32-7.29 (m, 1H), 7.25 (dd, *J* = 7.4, 7.4 Hz, 1H), 7.16 (dd, *J* = 7.4, 7.4 Hz, 1H), 6.03 (ddt, *J* = 18.0, 11.2, 5.7 Hz, 1H), 5.91 (ddt, *J* = 17.2, 10.3, 4.6 Hz, 1H), 5.15 (d, *J* = 10.3 Hz, 1H), 5.05-5.02 (m, 2H), 4.90 (d, *J* = 17.2 Hz, 1H), 4.61 (d, *J* = 4.6 Hz, 2H), 3.44 (d, *J* = 5.7 Hz, 2H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 137.6, 136.9, 136.4, 134.1, 133.7, 133.6, 130.3, 129.5, 128.5, 128.2, 127.9, 122.1, 119.6, 119.5, 116.4, 114.8, 111.6, 110.1, 46.5, 29.1. HRMS (ESI): m/z calcd. for $C_{20}H_{17}ClN^+$ $[M-H]^+$ 306.1044; found 306.1038.



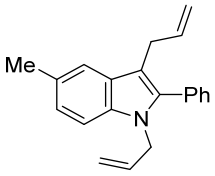
1,3-Diallyl-2-(2-methoxyphenyl)-1*H*-indole (**2h**)**:** 58% (90.2 mg from **1h** 156.2 mg). R_f = 0.26 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 1489, 1464, 1435, 1363, 1253, 1192, 1025, 741. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.64 (d, *J* = 7.4 Hz, 1H), 7.44 (ddd, *J* = 7.9, 7.9, 1.7 Hz, 1H), 7.34 (d, *J* = 7.9 Hz, 1H), 7.27 (dd, *J* = 7.4, 2.0 Hz, 1H), 7.21 (ddd, *J* = 7.9, 7.9, 1.7 Hz, 1H), 7.12 (dd, *J* = 7.4, 7.4 Hz, 1H), 7.04 (dd, *J* = 7.4, 7.4 Hz, 1H), 7.01 (d, *J* = 7.9 Hz, 1H), 5.97 (ddt, *J* = 17.1, 10.2, 6.1 Hz, 1H), 5.83 (ddt, *J* = 17.1, 10.2, 5.2 Hz, 1H), 5.05 (ddt, *J* = 10.2, 1.7, 1.7 Hz, 1H), 5.03 (ddt, *J* = 17.1, 1.7, 1.7 Hz, 1H), 4.96 (ddt, *J* = 10.2, 1.7, 1.7 Hz, 1H), 4.92 (ddt, *J* = 17.1, 1.7, 1.7 Hz, 1H), 4.58 (ddt, *J* = 16.9, 5.2, 1.7 Hz, 1H), 4.46 (ddt, *J* = 16.9, 5.2, 1.7 Hz, 1H), 3.76 (s, 3H), 3.42 (ddt, *J* = 15.8, 6.1, 1.7 Hz, 1H), 3.32 (ddt, *J* = 15.8, 6.1, 1.7 Hz, 1H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 158.0, 138.0, 136.5, 134.5, 134.3, 133.0, 130.2, 128.0, 121.3, 120.6, 120.4, 119.2, 118.9, 116.0, 114.2, 111.1, 110.9, 110.0, 55.3, 46.7, 29.5. HRMS (ESI): *m/z* calcd. for $C_{21}H_{20}NO^+$ $[M-H]^+$ 302.1539; found 302.1544.



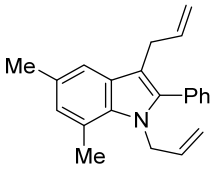
1,3-Diallyl-2-butyl-1*H*-indole (**2i**)**:** 62% (80.7 mg from **1i** 129.6 mg). R_f = 0.35 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 1468, 1438, 1415, 1364, 1180, 740. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.53 (d, *J* = 8.0 Hz, 1H), 7.23 (d, *J* = 8.0 Hz, 1H), 7.14 (dd, *J* = 8.0, 7.4 Hz, 1H), 7.07 (dd, *J* = 8.0, 7.4 Hz, 1H), 6.05-5.90 (m, 2H), 5.12 (dd, *J* = 10.3, 1.2 Hz, 1H), 5.08 (dd, *J* = 16.9, 1.9 Hz, 1H), 5.00 (dd, *J* = 10.2, 1.9 Hz, 1H), 4.85 (dd, *J* = 17.2, 1.2 Hz, 1H), 4.70 (ddd, *J* = 4.6, 1.7, 1.7 Hz, 2H), 3.50 (d, *J* = 5.9 Hz, 2H), 2.71 (t, *J* = 8.0 Hz, 2H), 1.56-1.52 (m, 2H), 1.46-1.36 (m, 2H), 0.95 (t, *J* = 7.4 Hz, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 137.8, 137.3, 136.1, 133.8, 128.0, 120.6, 118.8, 118.4, 116.0, 114.3, 109.2, 108.8, 45.4, 32.4, 29.0, 24.2, 22.7, 13.9. HRMS (ESI): *m/z* calcd. for $C_{18}H_{22}N^+$ $[M-H]^+$ 252.1747; found 252.1752.



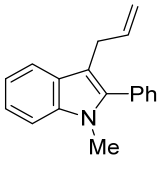
1,3-Diallyl-2-isopropyl-1*H*-indole (**2j**)**:** 47% (56.0 mg from **1j** 120.0 mg). R_f = 0.41 (hexane: AcOEt = 20:1). Colorless oil. IR (neat) ν cm^{-1}; 1469, 1416, 1362, 1189, 916, 742. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.52 (d, *J* = 8.0 Hz, 1H), 7.21 (d, *J* = 8.0 Hz, 1H), 7.14 (ddd, *J* = 8.0, 6.9, 1.2 Hz, 1H), 7.07 (ddd, *J* = 8.0, 6.9, 1.2 Hz, 1H), 6.07-5.91 (m, 2H), 5.13 (ddt, *J* = 10.3, 1.7, 1.7 Hz, 1H), 5.05 (ddt *J* = 17.2, 1.7, 1.7 Hz, 1H), 5.01 (ddt, *J* = 10.3, 1.7, 1.7 Hz, 1H), 4.86 (ddt, *J* = 17.2, 1.7, 1.7 Hz, 1H), 4.62 (ddd, *J* = 4.6, 1.7, 1.7 Hz, 2H), 3.61 (ddd, *J* = 5.7, 1.7, 1.7 Hz, 2H), 3.24 (sep, *J* = 7.1 Hz, 1H), 1.40 (d, *J* = 7.1 Hz, 6H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 141.6, 138.0, 136.0, 133.9, 128.5, 120.8, 118.9, 118.2, 116.0, 114.4, 109.1, 107.9, 45.7, 29.0, 25.8, 22.2. HRMS (ESI): *m/z* calcd. for $C_{17}H_{20}N^+$ $[M-H]^+$ 238.1590; found 238.1597.



1,3-Diallyl-5-methyl-2-phenyl-1*H*-indole (**2k**): 61% (87.0 mg from **1k** 142.7 mg). R_f = 0.31 (hexane:AcOEt = 20:1). Yellow solid. Mp 66-68 °C. IR (KBr) ν cm^{-1}; 1483, 1442, 1407, 1362, 1178, 761. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.48-7.40 (m, 6H), 7.21 (d, *J* = 8.3 Hz, 1H), 7.06 (d, *J* = 8.3 Hz, 1H), 6.03 (ddt, *J* = 16.7, 10.3, 5.7 Hz, 1H), 5.89 (ddt, *J* = 17.2, 10.3, 4.6 Hz, 1H), 5.12 (d, *J* = 10.3 Hz, 1H), 5.04 (dd, *J* = 16.7, 1.2 Hz, 1H), 5.01 (dd, *J* = 10.3, 1.2 Hz, 1H), 4.90 (dd, *J* = 17.2, 1.2 Hz, 1H), 4.58 (ddd, *J* = 4.6, 1.7, 1.7 Hz, 2H), 3.42 (d, *J* = 5.7 Hz, 2H), 2.48 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 138.1, 138.0, 135.2, 134.1, 131.9, 130.4, 128.6, 128.2, 128.0, 123.3, 119.0, 116.1, 114.5, 110.4, 109.8, 46.4, 29.2, 21.5 (note that two carbon peaks overlap with each other). HRMS (ESI): *m/z* calcd. for $C_{21}H_{20}N^+$ $[M-H]^+$ 286.1590; found 286.1583.

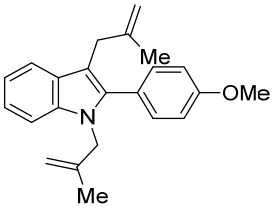


1,3-Diallyl-5,7-dimethyl-2-phenyl-1*H*-indole (**2l**): 67% (99.4 mg from **1l** 148.9 mg). R_f = 0.33 (hexane:AcOEt = 20:1). Yellow solid. Mp 48-49 °C. IR (KBr) ν cm^{-1}; 1468, 1446, 1417, 1364, 1176, 756. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.58-7.48 (m, 5H), 7.38 (s, 1H), 6.92 (s, 1H), 6.12 (ddt, *J* = 17.2, 10.3, 5.7 Hz, 1H), 5.93 (ddt, *J* = 17.2, 10.3, 3.7 Hz, 1H), 5.15-5.10 (m, 3H), 4.84 (ddd, *J* = 3.7, 1.7, 1.7 Hz, 2H), 4.69 (d, *J* = 17.2 Hz, 1H), 3.48 (d, *J* = 5.7 Hz, 2H), 2.78 (s, 3H), 2.56 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 139.2, 137.9, 136.0, 133.9, 132.2, 130.6, 129.1, 128.7, 128.1, 128.0, 126.6, 120.7, 116.8, 115.2, 114.4, 111.1, 47.5, 29.1, 21.2, 19.5. HRMS (ESI): *m/z* calcd. for $C_{22}H_{24}N^+$ $[M+H]^+$ 302.1903; found 302.1899.

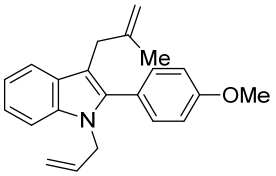


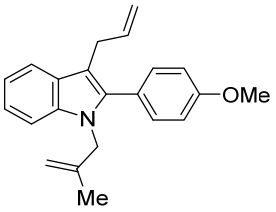
3-Allyl-1-methyl-2-phenyl-1*H*-indole (**2m**): 49% (59.1 mg from **1m** 120.4 mg). R_f = 0.29 (hexane:AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 1469, 1442, 1428, 1363, 1155, 741. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.63 (d, *J* = 8.0 Hz, 1H), 7.51-7.46 (m, 2H), 7.45-7.38 (m, 3H),7.35 (d, *J* = 8.0 Hz, 1H), 7.26 (ddd, *J* = 8.0, 8.0, 1.2 Hz, 1H), 7.15 (ddd, *J* = 8.0, 8.0, 1.2 Hz, 1H), 6.02 (ddt, *J* = 16.6, 10.3, 5.9 Hz, 1H), 5.05 (ddt, *J* = 16.6, 1.7, 1.7 Hz, 1H), 5.00 (ddt, *J* = 10.3, 1.7, 1.7 Hz, 1H), 3.63 (s, 3H), 3.45 (ddt, *J* = 5.9, 1.7, 1.7 Hz, 2H). ^{13}C-

NMR (125 MHz, $CDCl_3$) δ ppm; 138.1, 138.0, 137.3, 131.8, 130.5, 128.3, 128.0, 127.7, 121.7, 119.3, 119.2, 114.6, 110.6, 109.3, 30.9, 29.2. HRMS (ESI): m/z calcd. for $C_{18}H_{16}N^+$ $[M-H]^+$ 246.1277; found 246.1272.

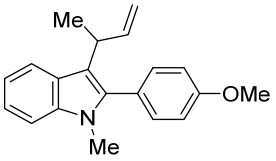


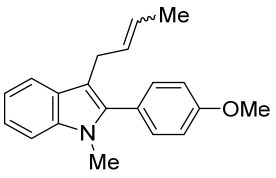
2-(4-Methoxyphenyl)-1,3-bis(2-methylallyl)-1*H*-indole (**2n**): 79% (131.6 mg from **1n** 166.2 mg). R_f = 0.29 (hexane:AcOEt = 20:1). Yellow solid. Mp 68-69 °C. IR (KBr) ν cm^{-1}; 1508, 1465, 1442, 1363, 1251, 1178, 1030, 742. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.62 (d, *J* = 8.0 Hz, 1H), 7.36 (d, *J* = 8.6 Hz, 2H), 7.29 (d, *J* = 8.0 Hz, 1H), 7.22 (dd, *J* = 8.0, 7.4 Hz, 1H), 7.14 (dd, *J* = 8.0, 7.4 Hz, 1H), 7.00 (d, *J* = 8.6 Hz, 2H), 4.85 (s, 1H), 4.79 (s, 1H), 4.69 (s, 1H), 4.52 (s, 2H), 4.49 (s, 1H), 3.89 (s, 3H), 3.40 (s, 2H), 1.75 (s, 3H), 1.63 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.4, 145.4, 141.5, 138.4, 136.7, 131.4, 128.3, 124.2, 121.4, 119.3, 119.2, 113.7, 111.1, 110.6, 110.5, 110.0, 55.2, 49.6, 33.3, 22.7, 20.0. HRMS (ESI): m/z calcd. for $C_{23}H_{24}NO^+$ $[M-H]^+$ 330.1852; found 330.1851.



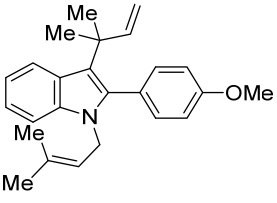


1-Allyl-2-(4-methoxyphenyl)-3-(2-methylallyl)-1*H*-indole (**2o**) and **3-allyl-2-(4-methoxyphenyl)-1-(2-methylallyl)-1*H*-indole** (**2o'**): 58% (**2o**:**2o'** = 64:36, 111.0 mg from **1n** 190.0 mg). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow oil. IR (neat) ν cm^{-1}; 1509, 1465, 1432, 1360, 1256, 1180, 1030, 746. ^{1}H-NMR (500 MHz, $CDCl_3$, 64:36 mixture of **2o** and **2o'**) δ ppm; 7.61 (d, *J* = 8.0 Hz, 0.36H), 7.60 (d, *J* = 7.6 Hz, 0.64H), 7.35-7.26 (m, 3H), 7.23-7.18 (m, 1H), 7.15-7.10 (m, 1H), 7.01-6.96 (m, 2H), 6.94 (ddt, *J* = 16.9, 10.0, 5.7 Hz, 0.36H), 5.90 (ddt, *J* = 17.2, 10.3, 4.6 Hz, 0.64H), 5.11 (ddt, *J* = 10.3, 1.2, 1.7 Hz, 0.64H), 5.05-4.99 (m, 0.72H), 4.87 (ddt, *J* = 17.2, 1.2, 1.7 Hz, 0.64H), 4.83 (s, 0.36H), 4.75 (s, 0.64H), 4.66 (s, 0.64H), 4.59 (ddd, *J* = 4.6, 2.3, 2.3 Hz, 1.28H), 4.48 (s, 1.08H), 3.87 (s, 3H), 3.43 (ddd, *J* = 5.7, 1.7, 1.7 Hz, 0.72H), 3.36 (s, 1.28H), 1.71 (s, 1.92H), 1.62 (s, 1.08H). ^{13}C-NMR (125 MHz, $CDCl_3$, 64:36 mixture of **2o** and **2o'**) δ ppm; 159.4, 145.4, 141.4 (minor), 138.3 (minor), 138.1, 136.8 (minor), 136.6, 134.1, 131.6, 131.5 (minor), 128.3, 128.0 (minor), 124.10, 124.06 (minor), 121.5 (minor), 121.4, 119.4, 119.22 (minor), 119.19, 119.1 (minor), 116.1, 114.5 (minor), 113.7, 111.1 (minor), 110.7, 110.6, 110.4 (minor), 110.1 (minor), 109.9, 55.3, 49.6 (minor), 46.3, 33.3, 29.2 (minor), 22.7, 20.1 (minor) [note that four carbon peaks of **2o'** overlap with the peaks of **2o**]. HRMS (ESI): m/z calcd. for $C_{22}H_{22}NO^+$ $[M-H]^+$ 316.1696; found 316.1691.

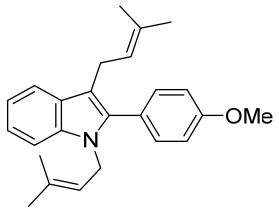




3-(But-3-en-2-yl)-2-(4-methoxyphenyl)-1-methyl-1*H*-indole (**2p**) and **3-(but-2-en-1-yl)-2-(4-methoxyphenyl)-1-methyl-1*H*-indole** (**4p**): 61% (**2p**:*E*-**4p**:*Z*-**4p** = 73:14:13, 87.0 mg from **1p** 142.0 mg). R_f = 0.25 (hexane:AcOEt = 20:1). Yellow solid. Mp 105-107 °C. IR (KBr) ν cm^{-1}; 1504, 1465, 1433, 1365, 1248, 1178, 1025, 745. ^{1}H-NMR (500 MHz, $CDCl_3$, 73:14:13 mixture of **2p**, *E*-**4p** and *Z*-**4p**) δ ppm; 7.70 (d, *J* = 8.0 Hz, 0.73H), 7.61 (dd, *J* = 8.0, 8.0 Hz, 0.27H), 7.35-7.28 (m, 3H), 7.22 (ddd, *J* = 8.0, 8.0, 1.2 Hz, 1H), 7.13 (dd, *J* = 7.4, 7.4 Hz, 0.27H), 7.09 (dd, *J* = 7.2, 7.2 Hz, 0.73H), 7.02 (d, *J* = 8.6 Hz, 2H), 6.21 (ddd, *J* = 17.5, 10.3, 5.2 Hz, 0.73H), 5.68-5.54 (m, 0.27H), 5.52-5.40 (m, 0.27H), 5.05 (ddd, *J* = 17.5, 1.7, 1.7 Hz, 0.73H), 5.00 (ddd, *J* = 10.3, 1.7, 1.7 Hz, 0.73H), 3.89 (s, 3H), 3.64-3.57 (m, 0.73H), 3.60 (s, 0.42H), 3.59 (s, 0.39H), 3.55 (s, 2.19H), 3.43 (d, *J* = 6.9 Hz, 0.27H), 3.35 (ddq, *J* = 5.7, 1.4, 1.5 Hz, 0.27H), 1.68 (dd, *J* = 7.2, 1.4 Hz, 0.39H), 1.63 (ddt, *J* = 7.3, 1.7, 1.5 Hz, 0.42H), 1.45 (d, *J* = 7.4 Hz, 2.19H). ^{13}C-NMR (125 MHz, $CDCl_3$, **2p** in 73:14:13 mixture of **2p**, *E*-**4p** and *Z*-**4p**) δ ppm; 159.5, 143.3, 137.2, 137.1, 132.0, 126.2, 124.3, 121.2, 120.4, 118.8, 115.8, 113.7, 112.4, 109.4, 55.3, 34.9, 30.6, 20.2. HRMS (ESI): *m/z* calcd. for $C_{20}H_{20}NO^+$ $[M-H]^+$ 290.1539; found 290.1541.



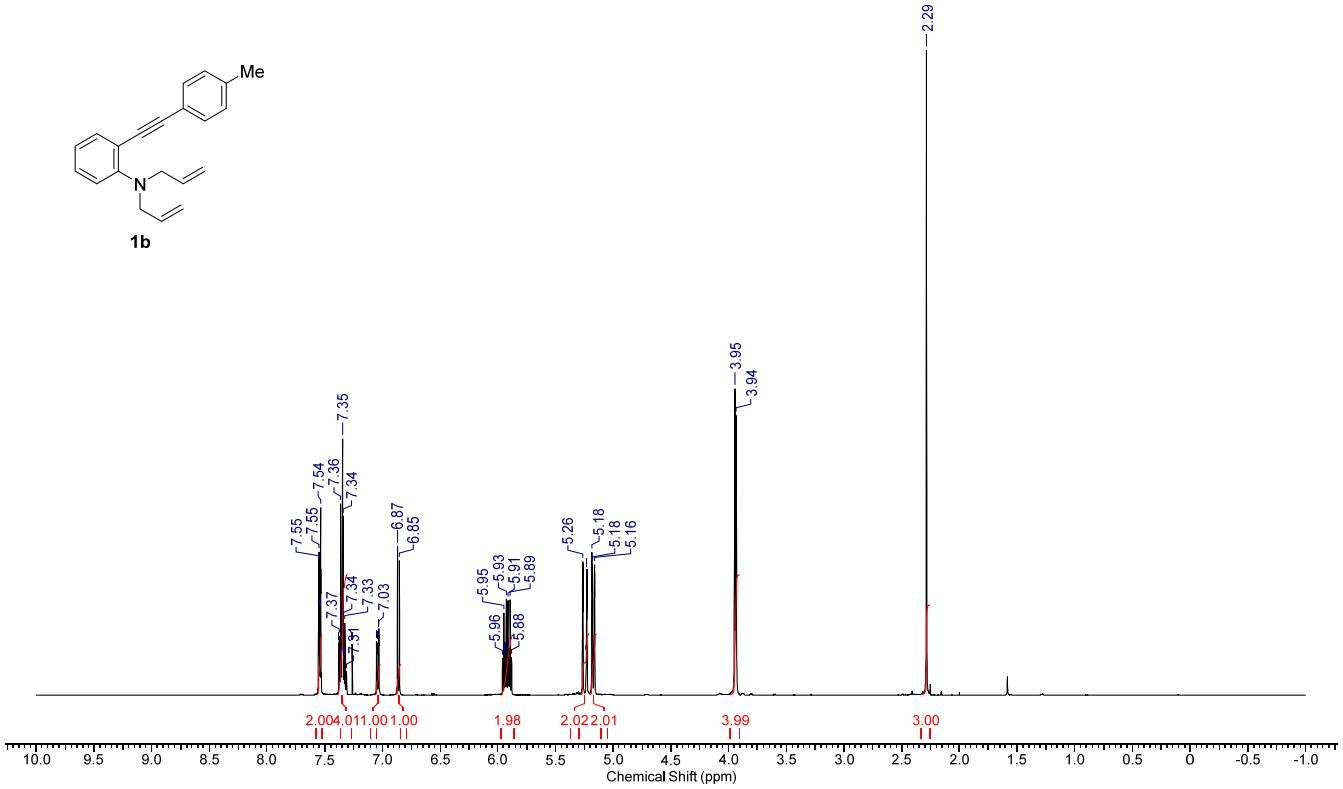
2-(4-Methoxyphenyl)-1-(3-methylbut-2-en-1-yl)-3-(2-methylbut-3-en-2-yl)-1*H*-indole (**2q**): 16% (28.7 mg from **1q** 181.4 mg). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow solid. Mp 103-104 °C. IR (KBr) ν cm^{-1}; 1502, 1463, 1439, 1358, 1246, 1181, 1033, 746. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.86 (d, *J* = 8.0 Hz, 1H), 7.31-7.23 (m, 3H), 7.20 (dd, *J* = 8.0, 6.9 Hz, 1H), 7.08 (dd, *J* = 7.5, 6.9 Hz, 1H), 6.94 (d, *J* = 8.6 Hz, 2H), 6.16 (dd, *J* = 17.2, 10.9 Hz, 1H), 5.13 (t, *J* = 6.3 Hz, 1H), 5.04 (dd, *J* = 17.2, 1.2 Hz, 1H), 4.93 (dd, *J* = 10.9, 1.2 Hz, 1H), 4.36 (d, *J* = 6.3 Hz, 2H), 3.89 (s, 3H), 1.64 (s, 3H), 1.50 (s, 3H), 1.32 (s, 6H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.5, 149.4, 136.0, 135.9, 133.4, 132.9, 126.83, 126.77, 122.1, 121.1, 121.0, 118.6, 118.3, 113.0, 109.5, 109.3, 55.3, 41.5, 39.0, 29.7, 25.5, 17.7. HRMS (ESI): *m/z* calcd. for $C_{25}H_{28}NO^+$ $[M-H]^+$ 358.2165; found 358.2155.



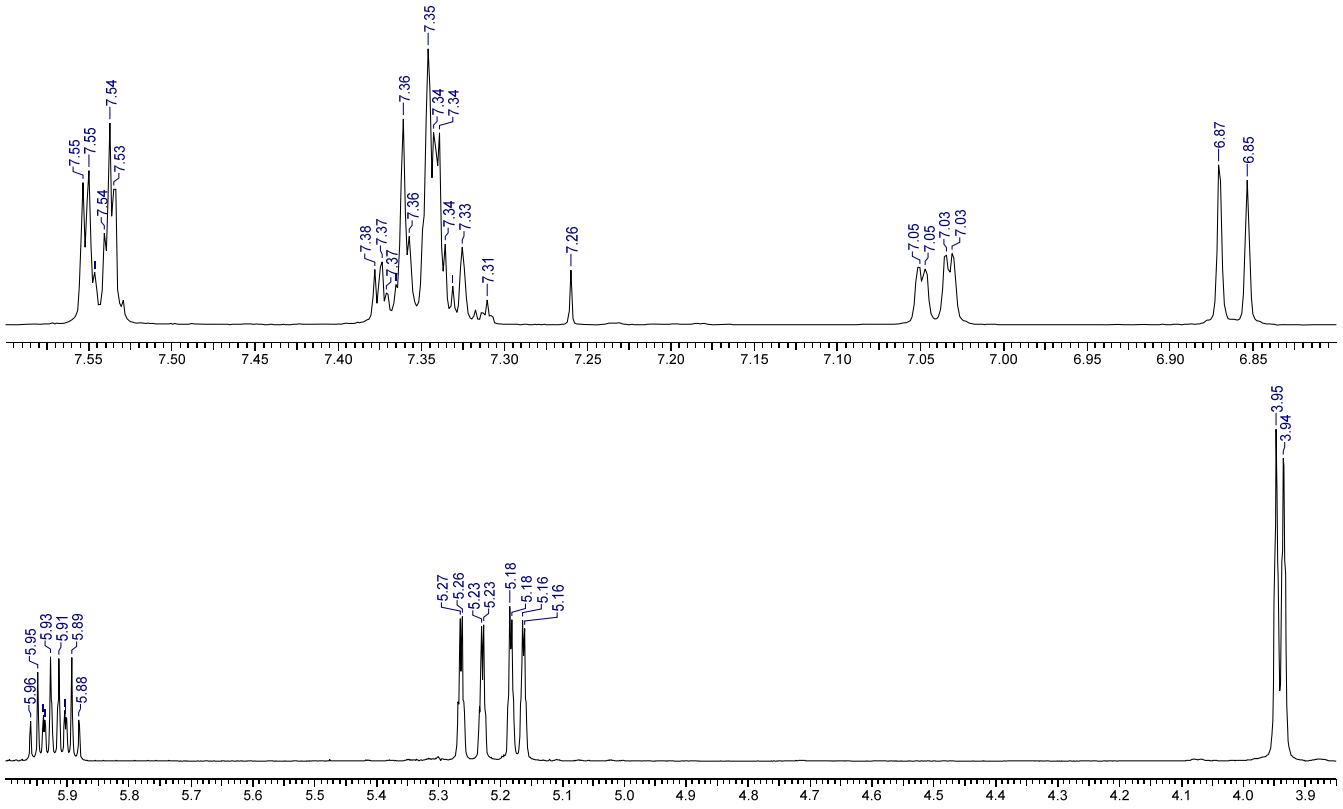
2-(4-Methoxyphenyl)-1,3-bis(3-methylbut-2-en-1-yl)-1*H*-indole (**4q**): 9% (17.0 mg from **1q** 181.4 mg). R_f = 0.29 (hexane: AcOEt = 20:1). Yellow solid. Mp 78-79 ℃. IR (KBr) ν cm^{-1}; 1505, 1466, 1442, 1359, 1249, 1176, 1034, 741. ^{1}H-NMR (500 MHz, $CDCl_3$) δ ppm; 7.60 (d, *J* = 8.0 Hz, 1H), 7.32-7.29 (m, 3H), 7.21 (dd, *J* = 8.0, 6.9 Hz, 1H), 7.12 (dd, *J* = 8.0, 6.9 Hz, 1H), 7.00 (d, *J* = 8.6 Hz, 2H), 5.32 (t, *J* = 6.9 Hz, 1H), 5.23 (t, *J* = 6.3 Hz, 1H), 4.57 (d, *J* = 6.3 Hz, 2H), 3.88 (s, 3H), 3.36 (d, *J* = 6.9 Hz, 2H), 1.67 (s, 6H), 1.66 (s, 3H), 1.58 (s, 3H). ^{13}C-NMR (125 MHz, $CDCl_3$) δ ppm; 159.3, 137.1, 136.3, 133.8, 132.1, 130.2, 127.9, 124.49, 124.47, 121.32, 121.28, 119.1, 119.0, 113.7, 112.6, 109.8, 55.3, 42.3, 25.7, 25.5, 23.9, 17.84, 17.76. HRMS (ESI): *m/z* calcd. for $C_{25}H_{28}NO^+$ $[M-H]^+$ 358.2165; found 358.2165.

5. ^{1}H and ^{13}C NMR Spectra of 1b-1q, 2a-2f, 2h-2q, 4p and 4q

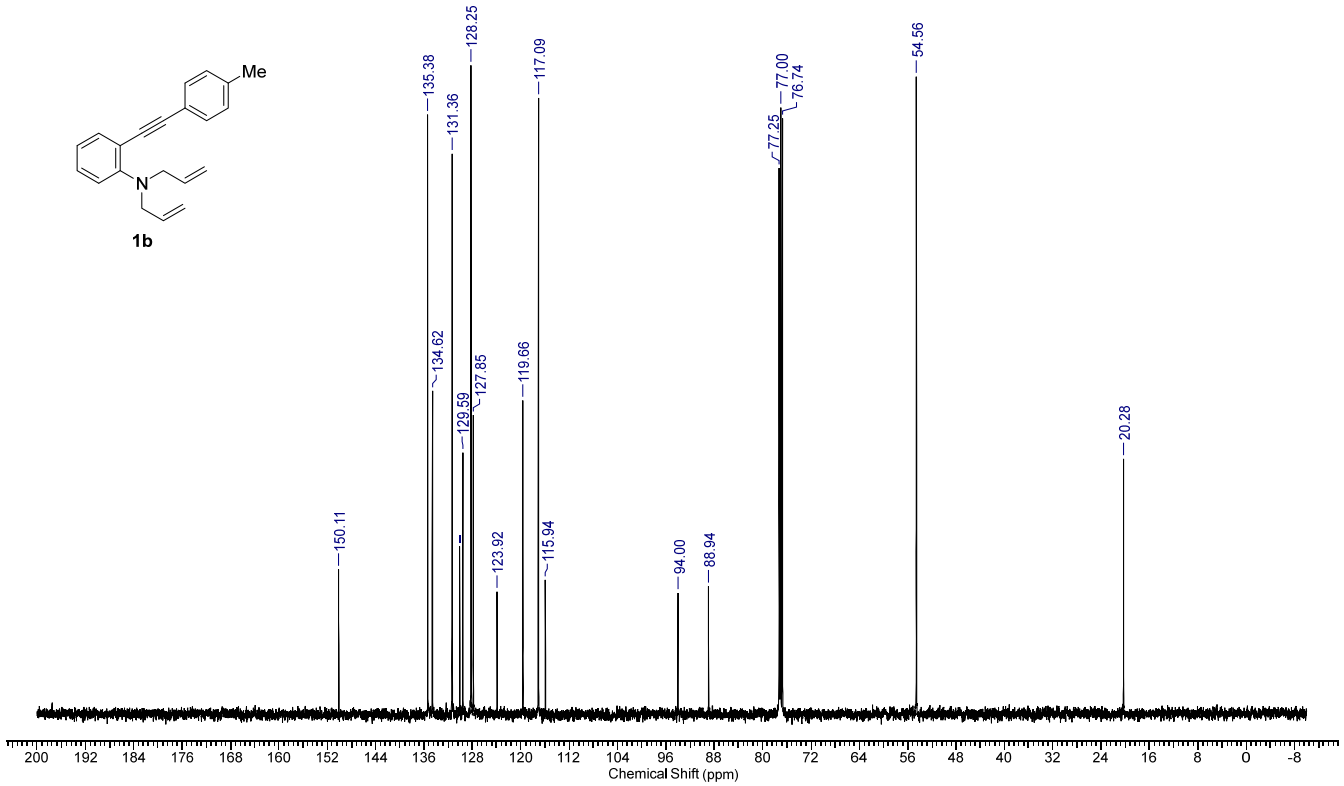
^{1}H NMR (500 MHz, $CDCl_3$) of **1b**



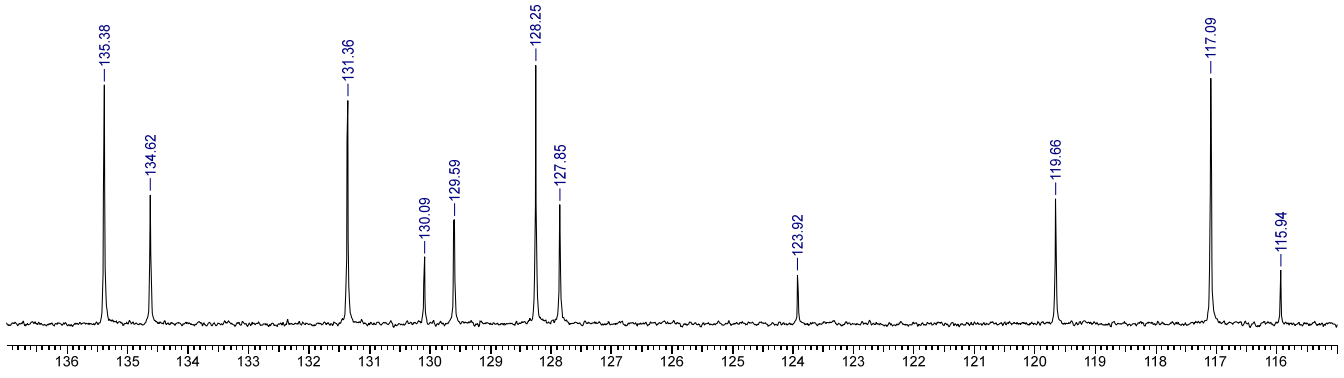
Enlarged view



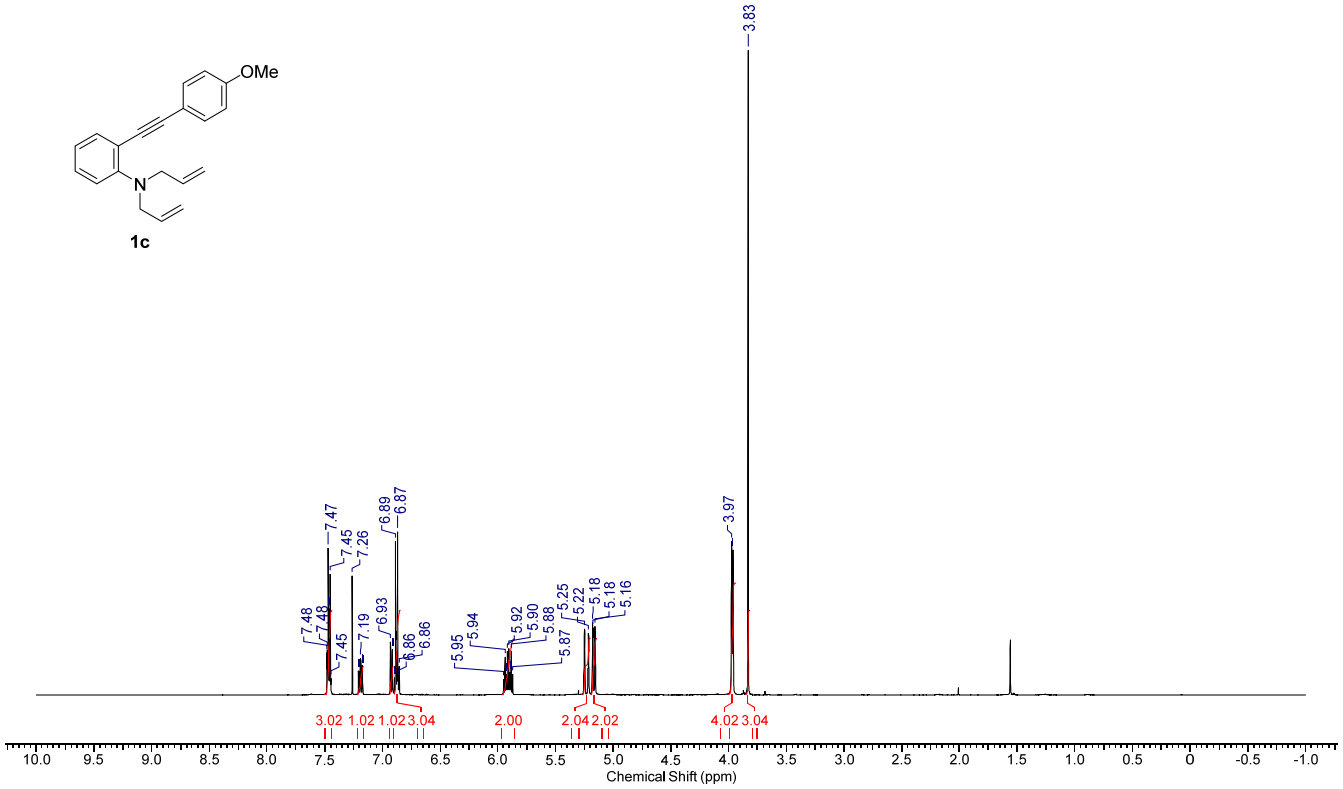
^{13}C NMR (125 MHz, $CDCl_3$) of **1b**



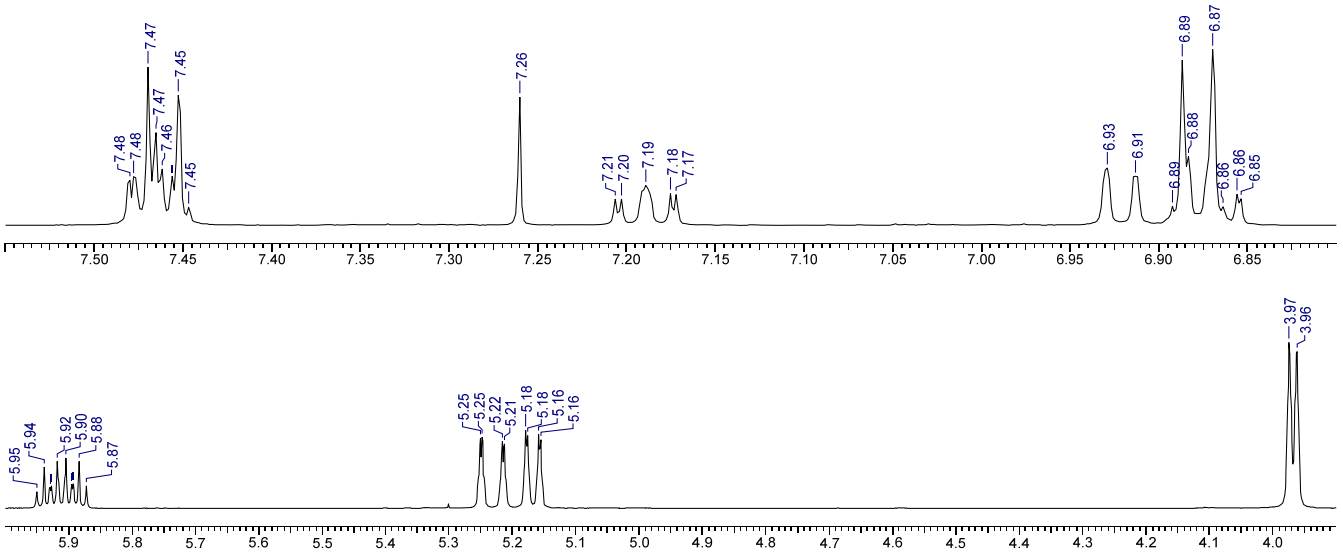
Enlarged view



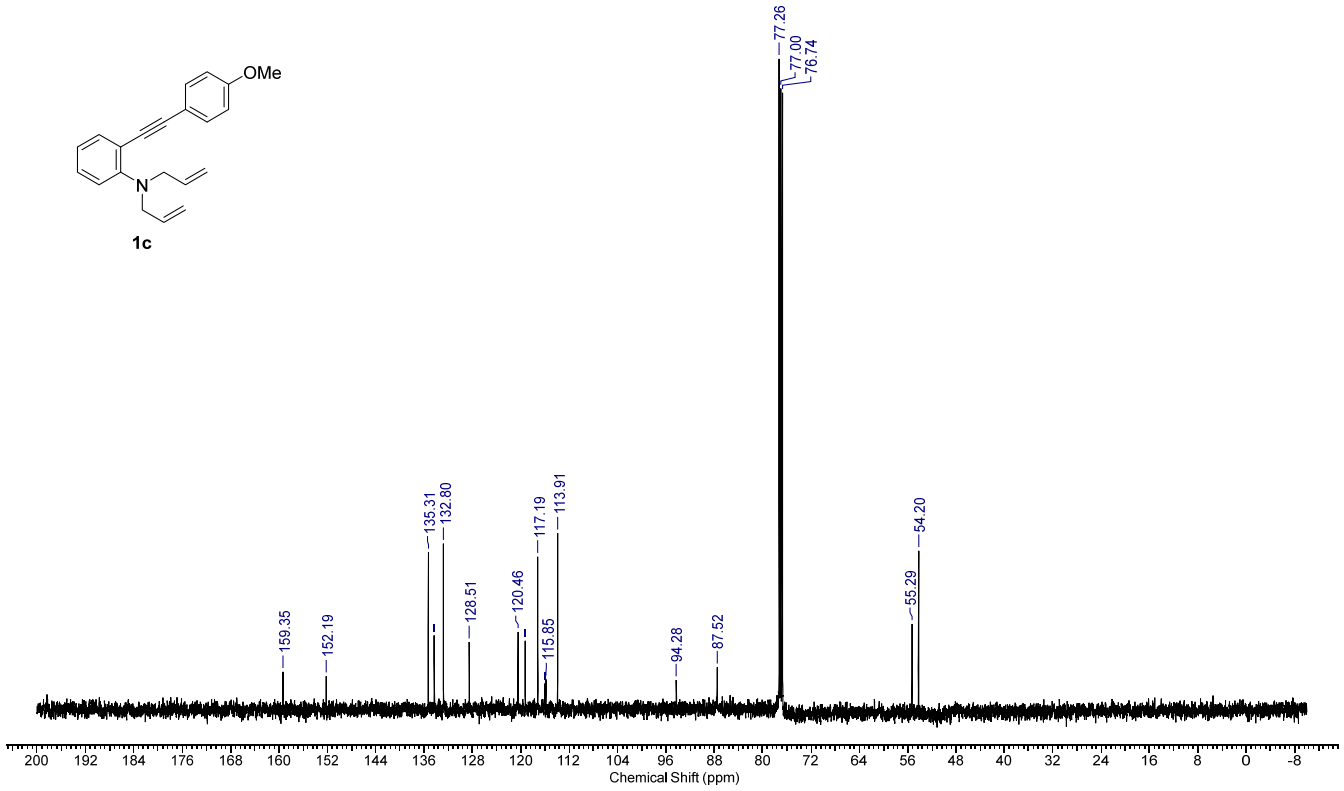
^{1}H NMR (500 MHz, $CDCl_3$) of **1c**



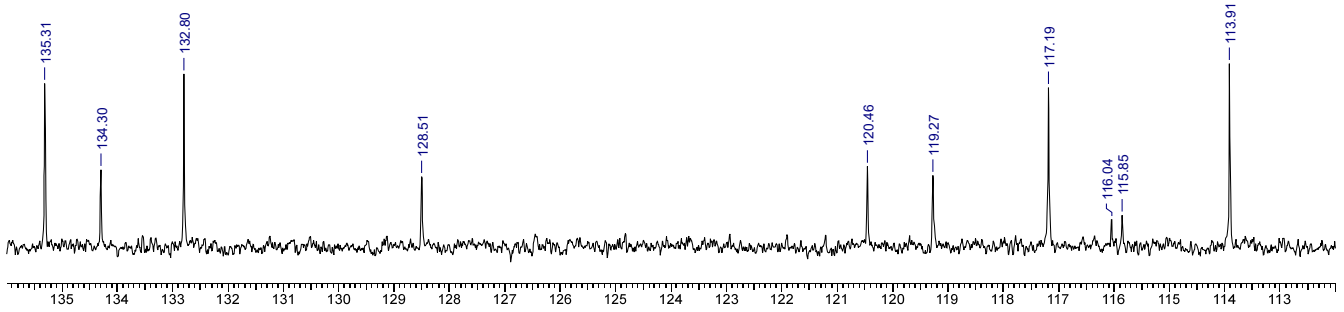
Enlarged view



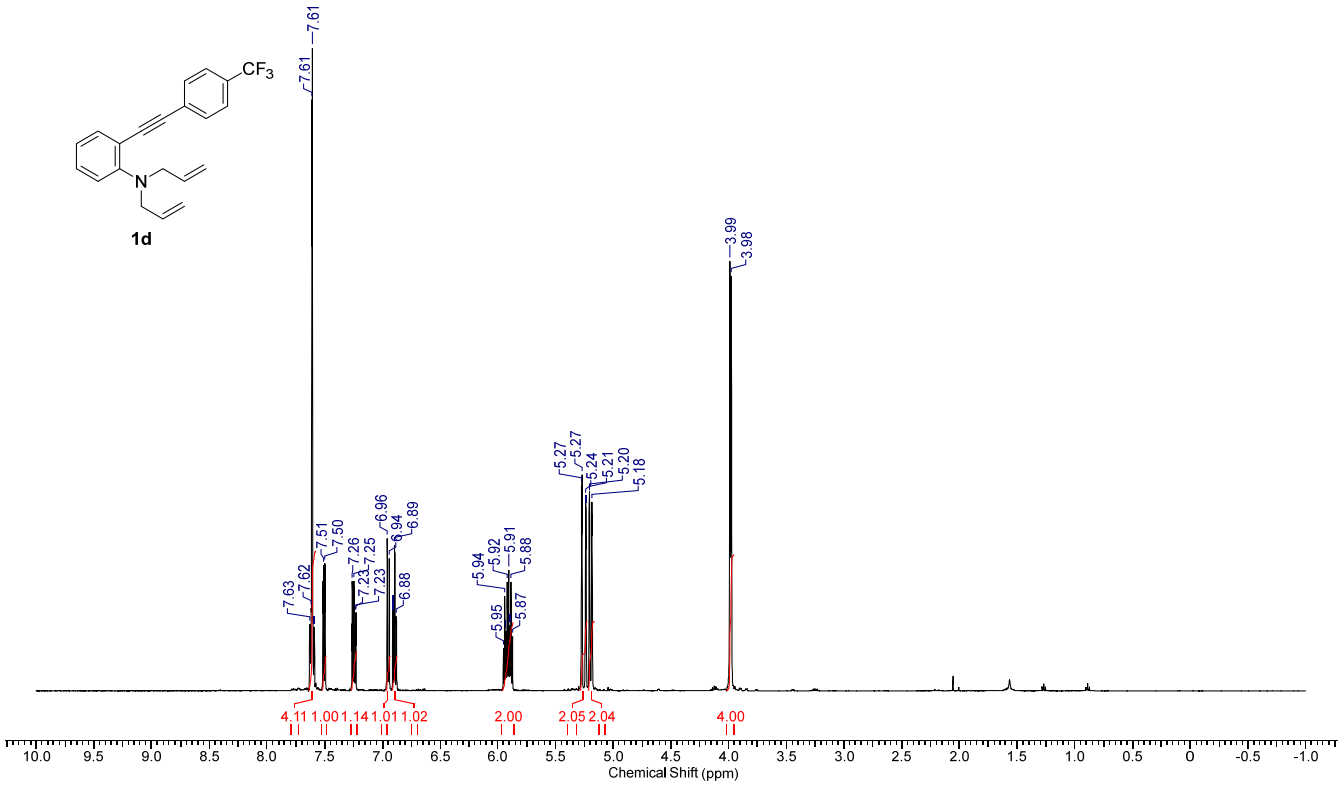
^{13}C NMR (125 MHz, $CDCl_3$) of **1c**



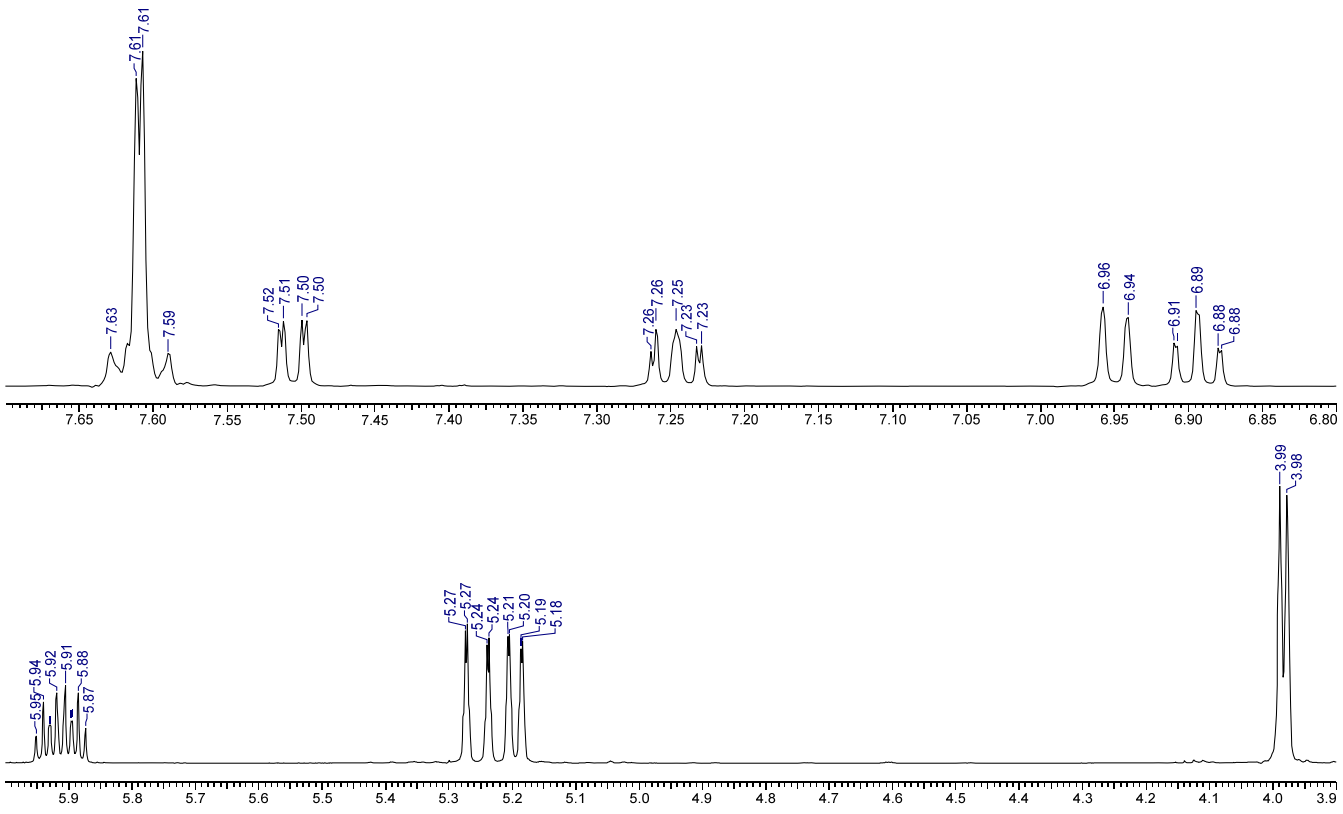
Enlarged view



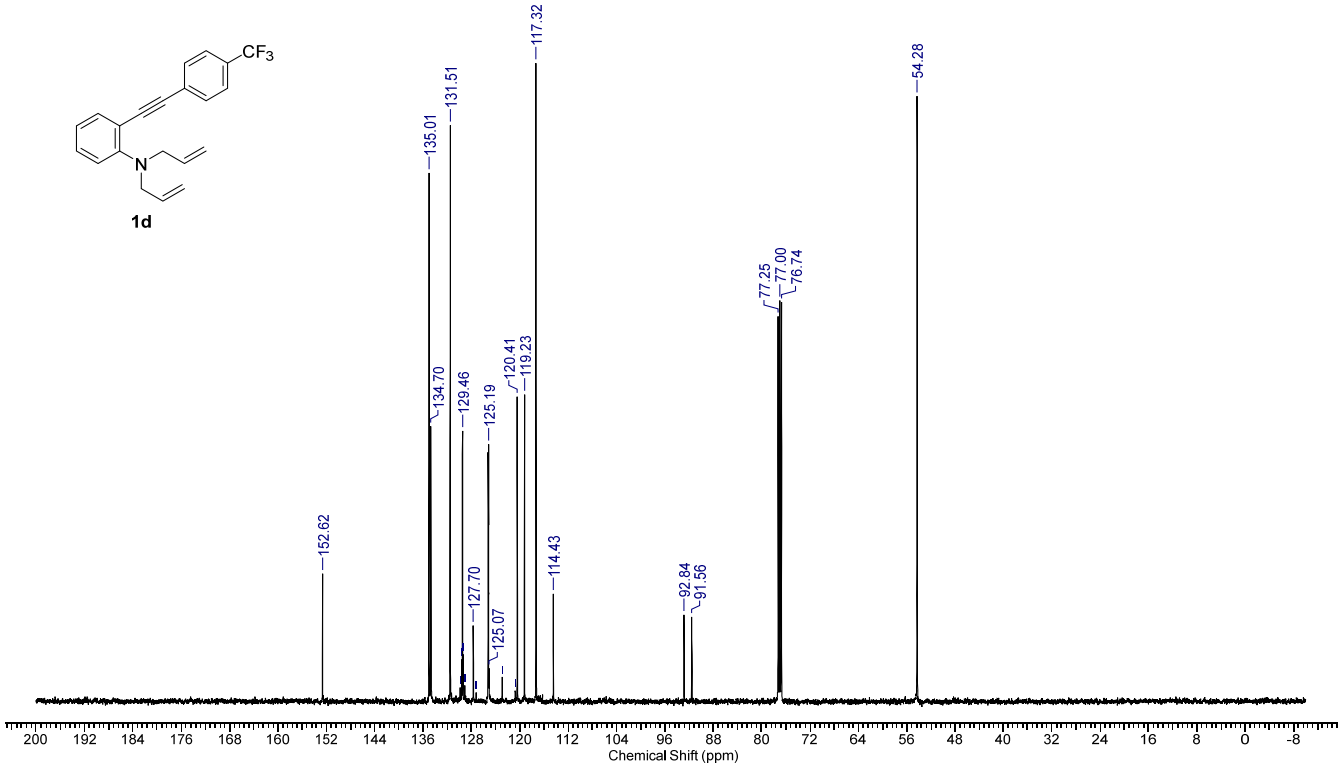
^{1}H NMR (500 MHz, $CDCl_3$) of **1d**



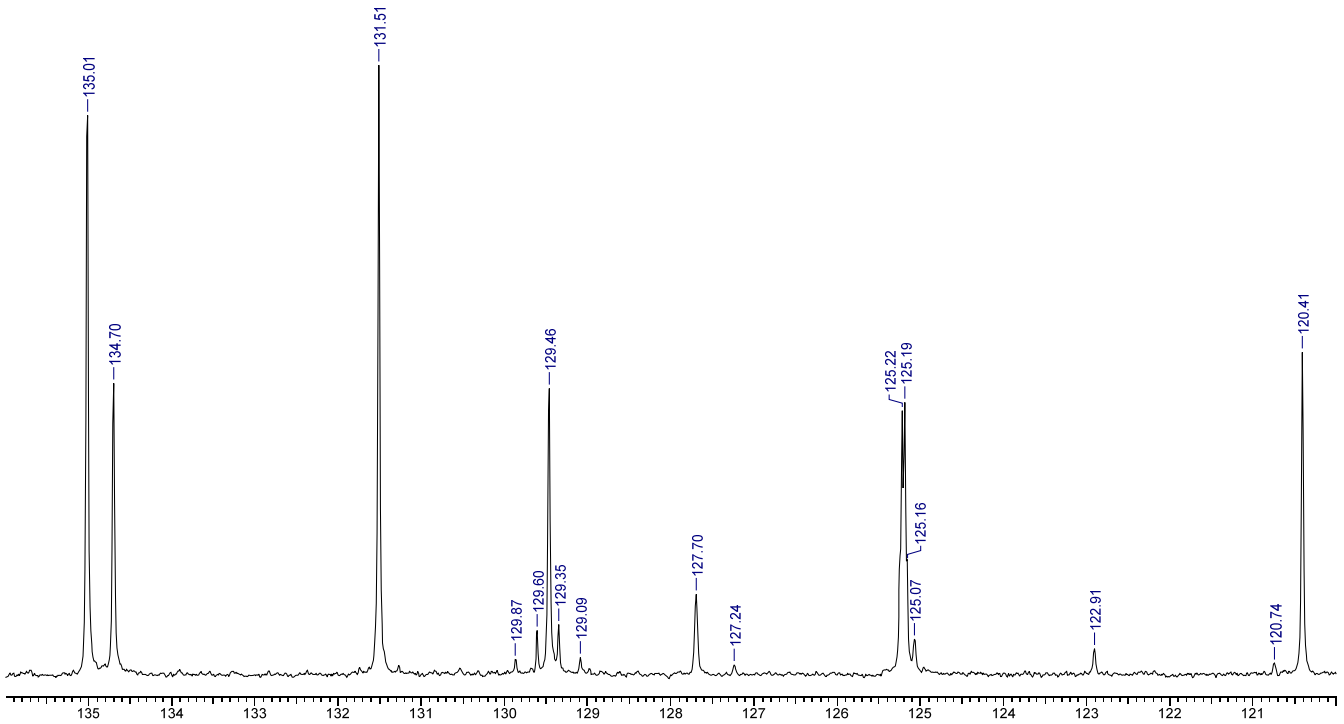
Enlarged view



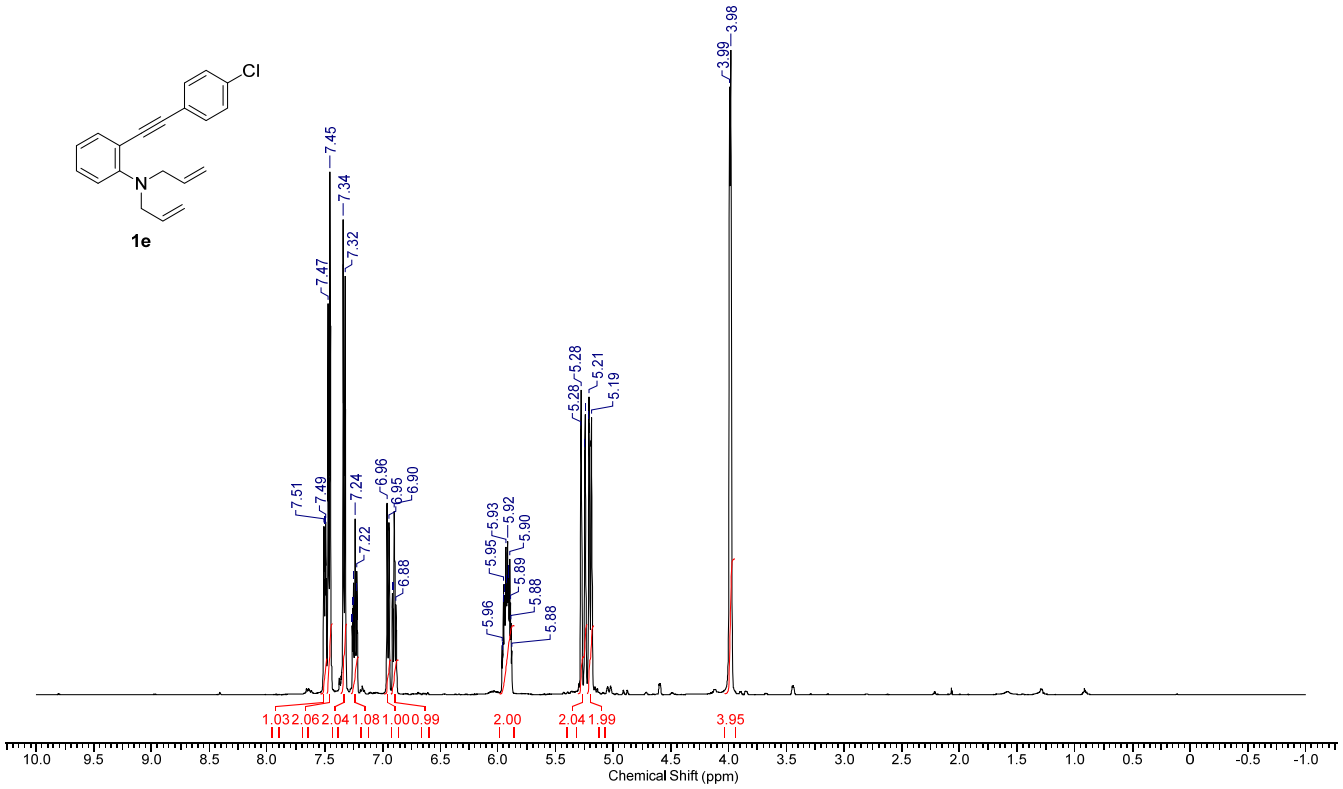
^{13}C NMR (125 MHz, $CDCl_3$) of **1d**



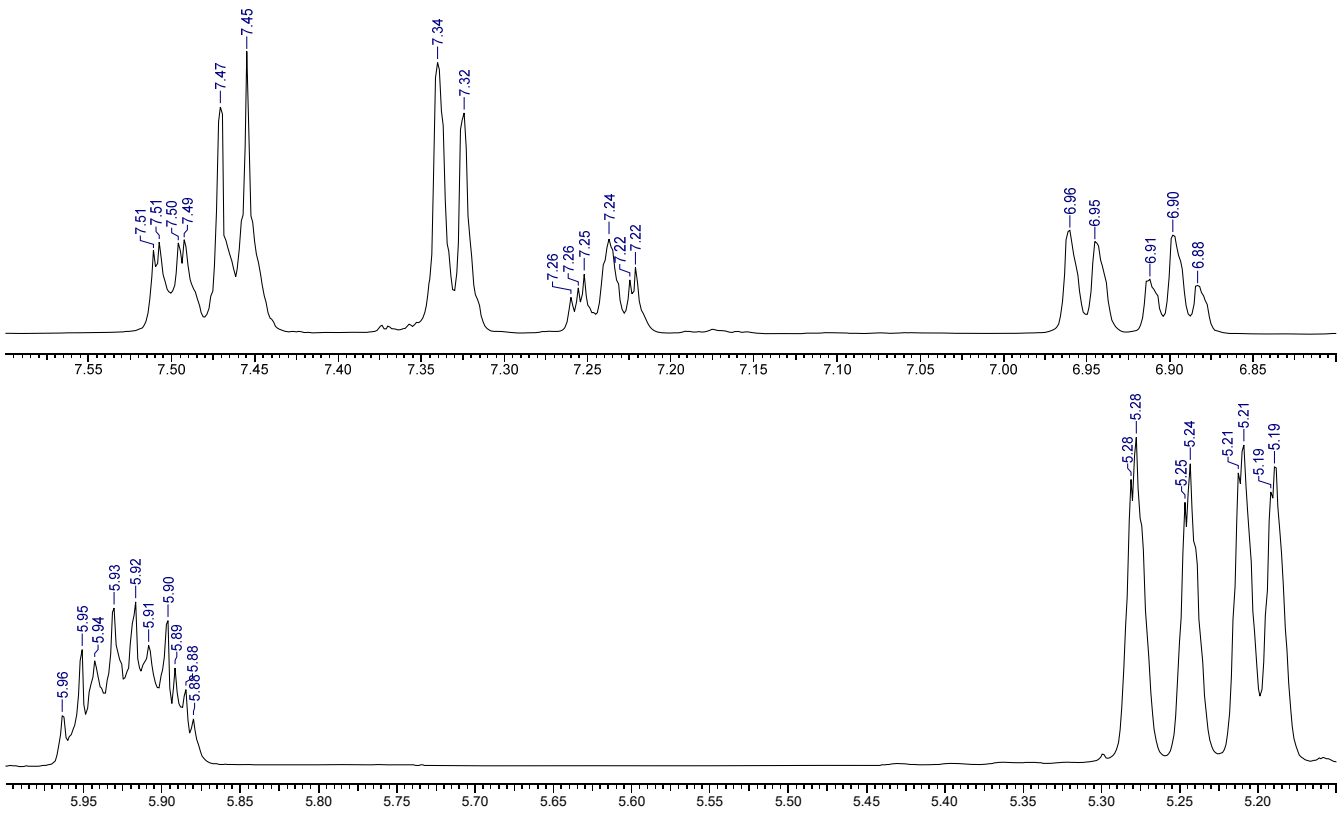
Enlarged view



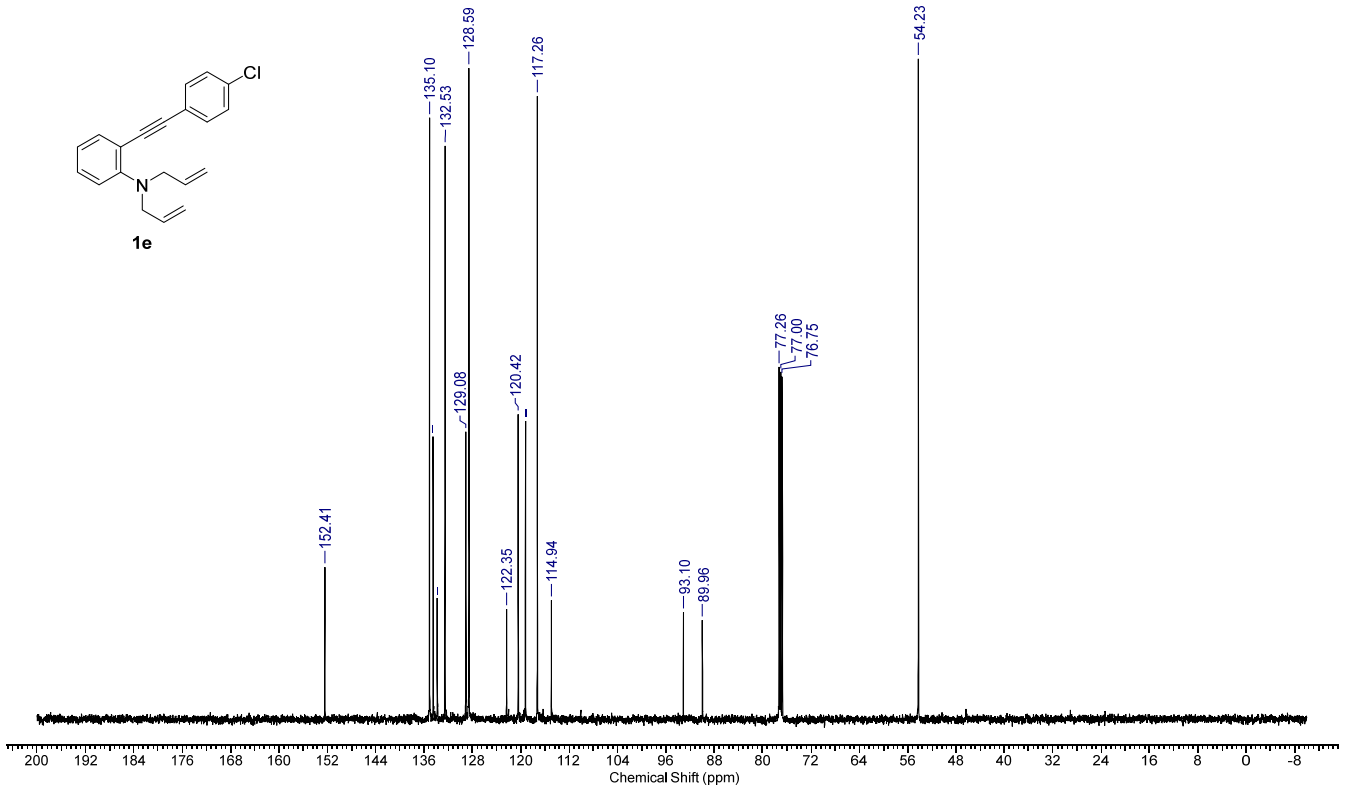
^{1}H NMR (500 MHz, $CDCl_3$) of **1e**



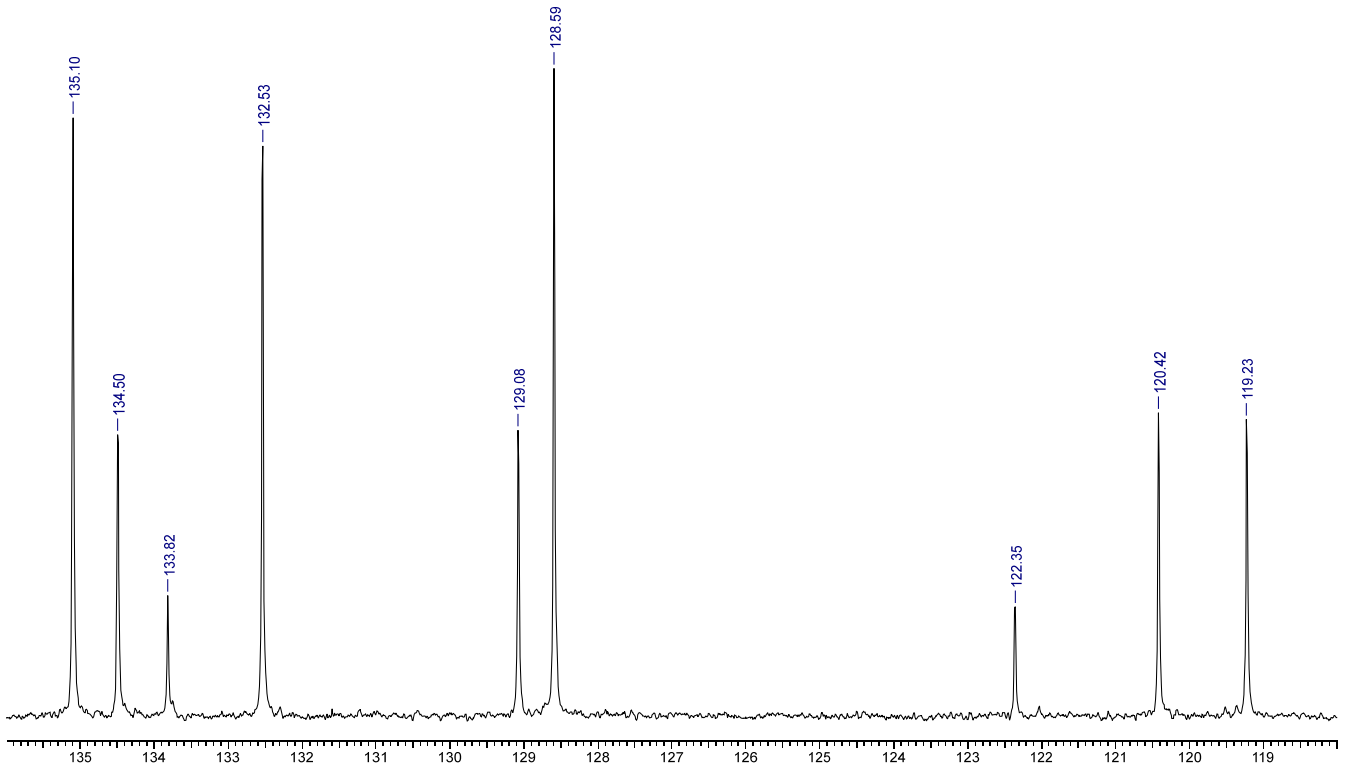
Enlarged view



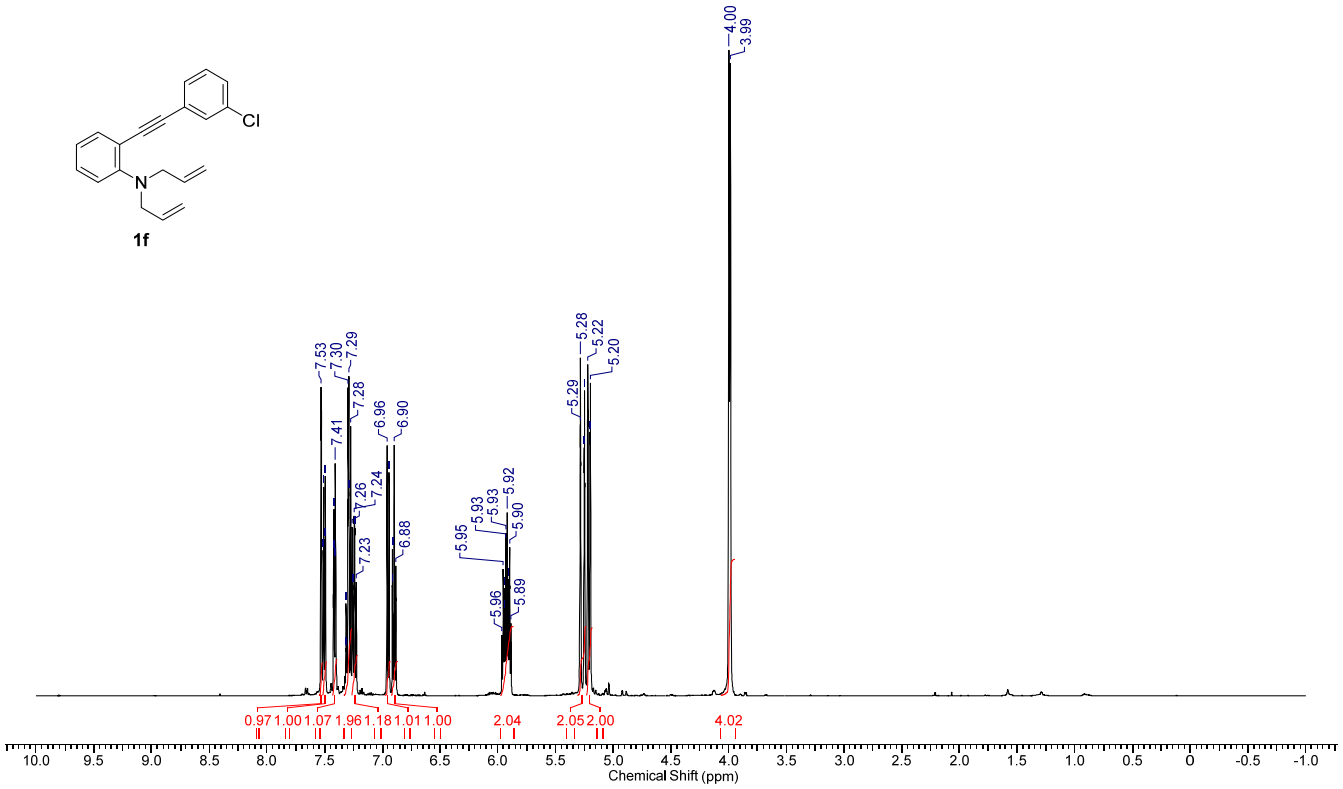
^{13}C NMR (125 MHz, $CDCl_3$) of **1e**



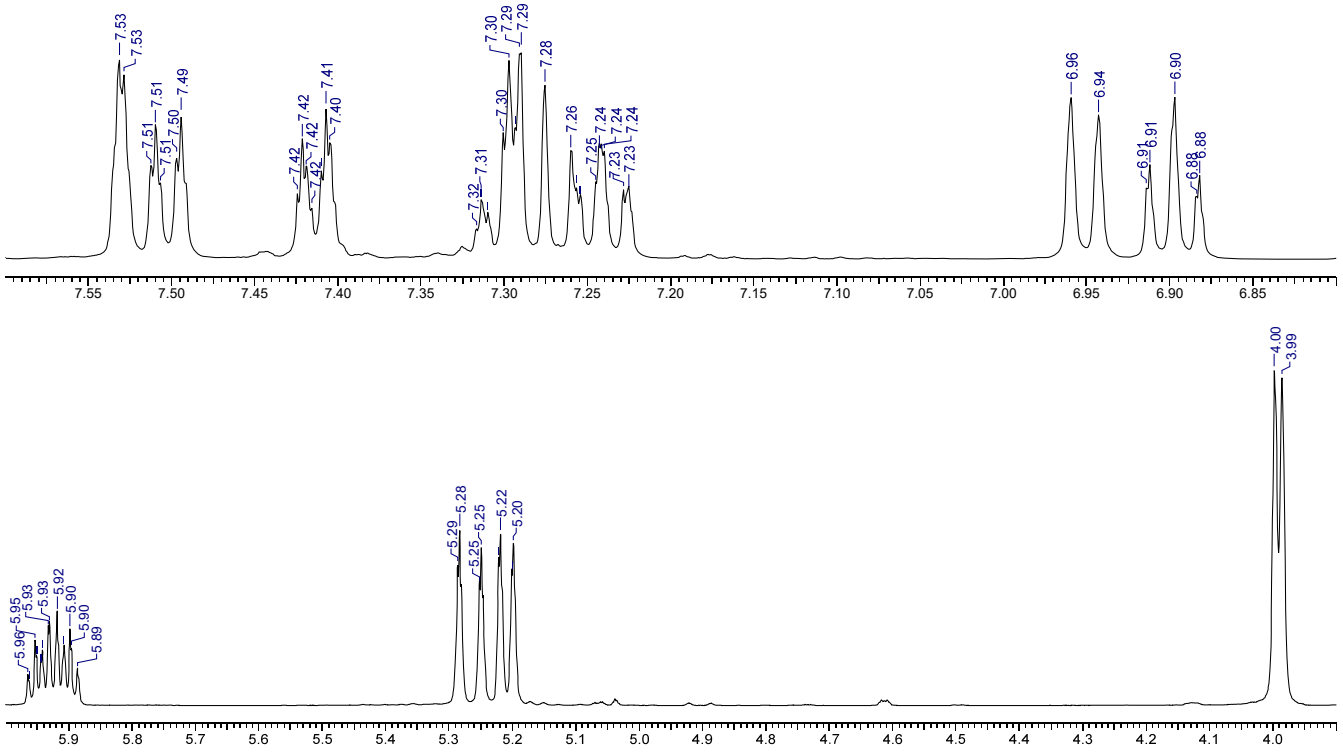
Enlarged view



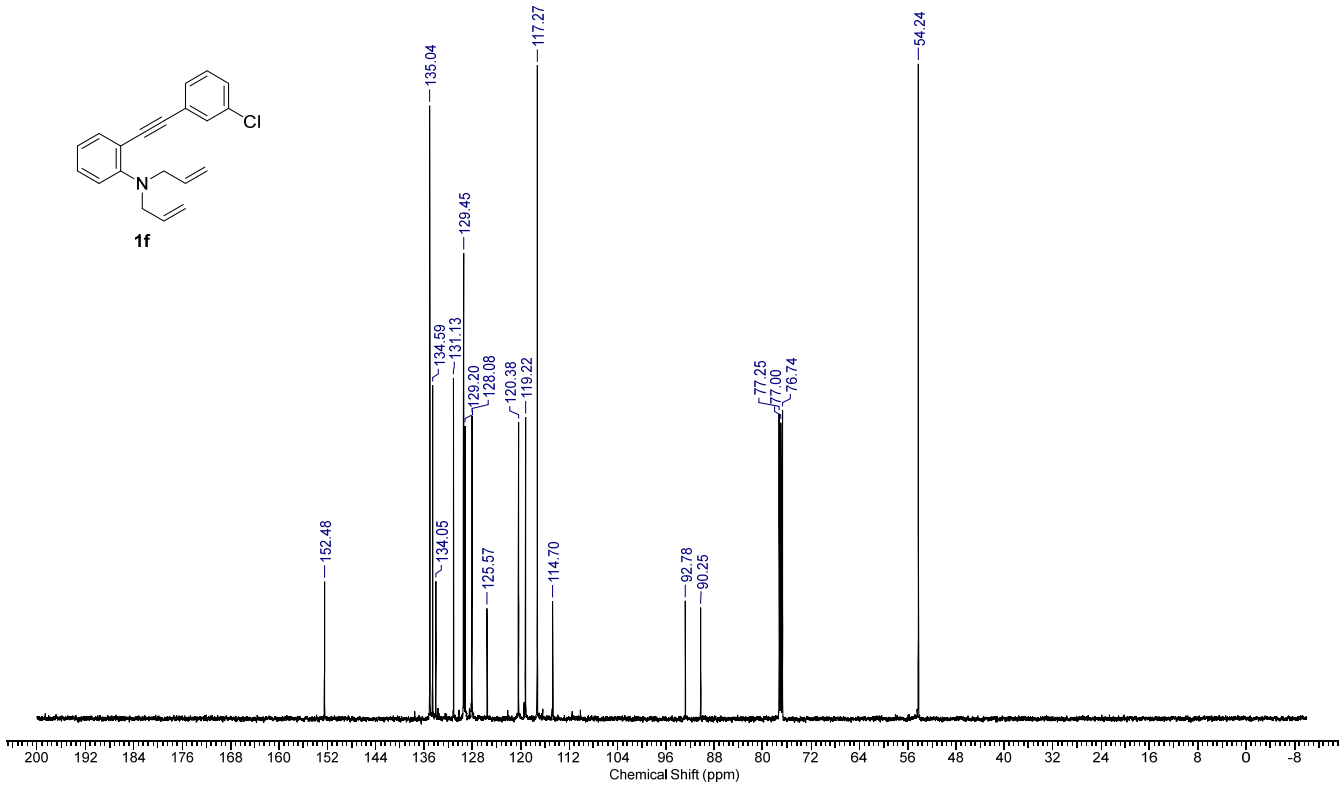
^{1}H NMR (500 MHz, $CDCl_3$) of **1f**



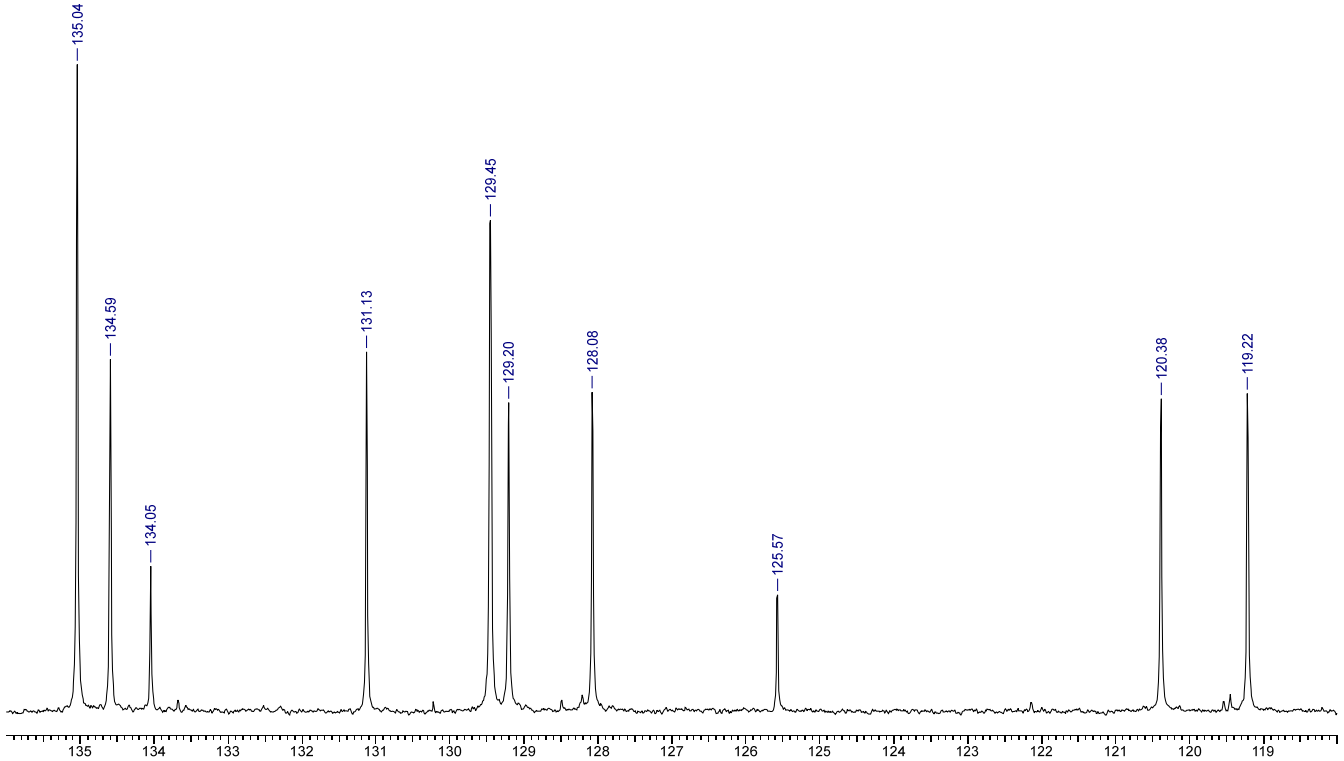
Enlarged view



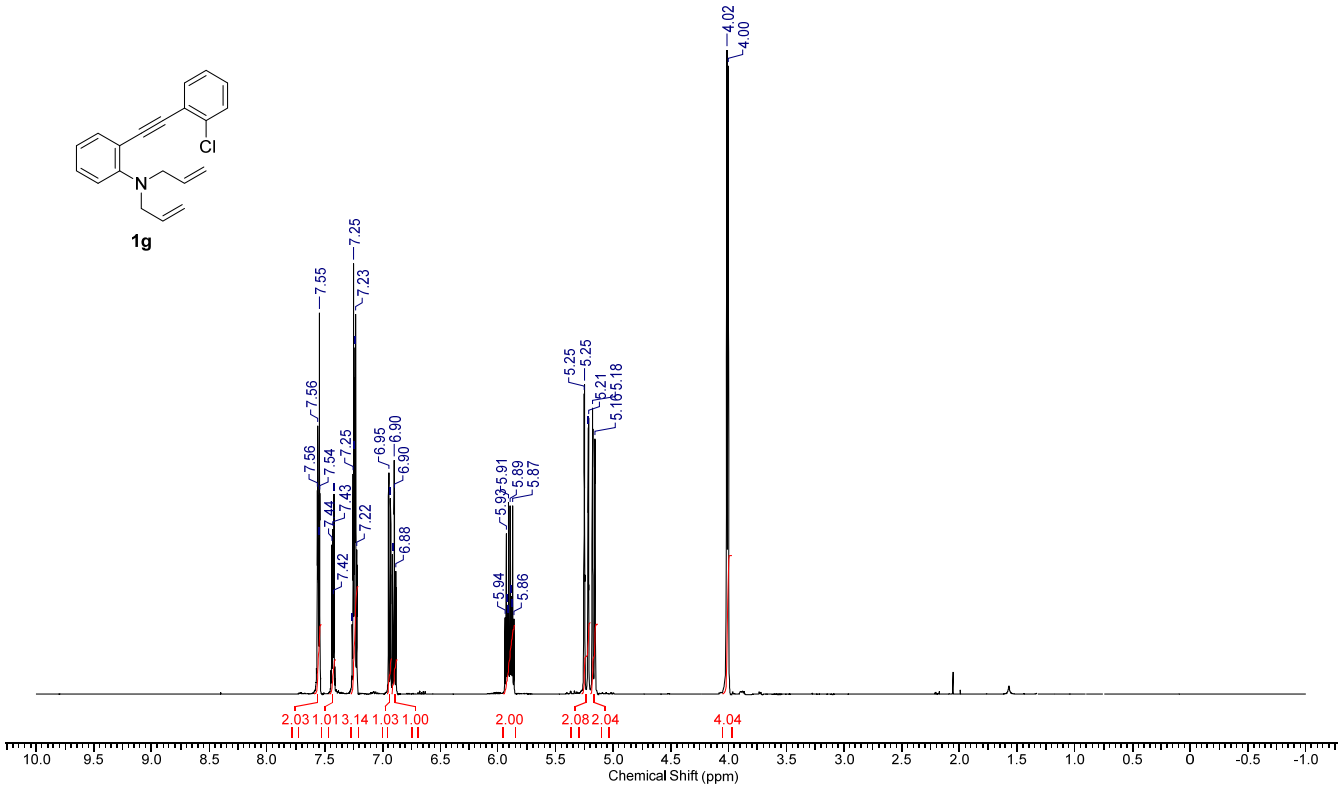
^{13}C NMR (125 MHz, $CDCl_3$) of **1f**



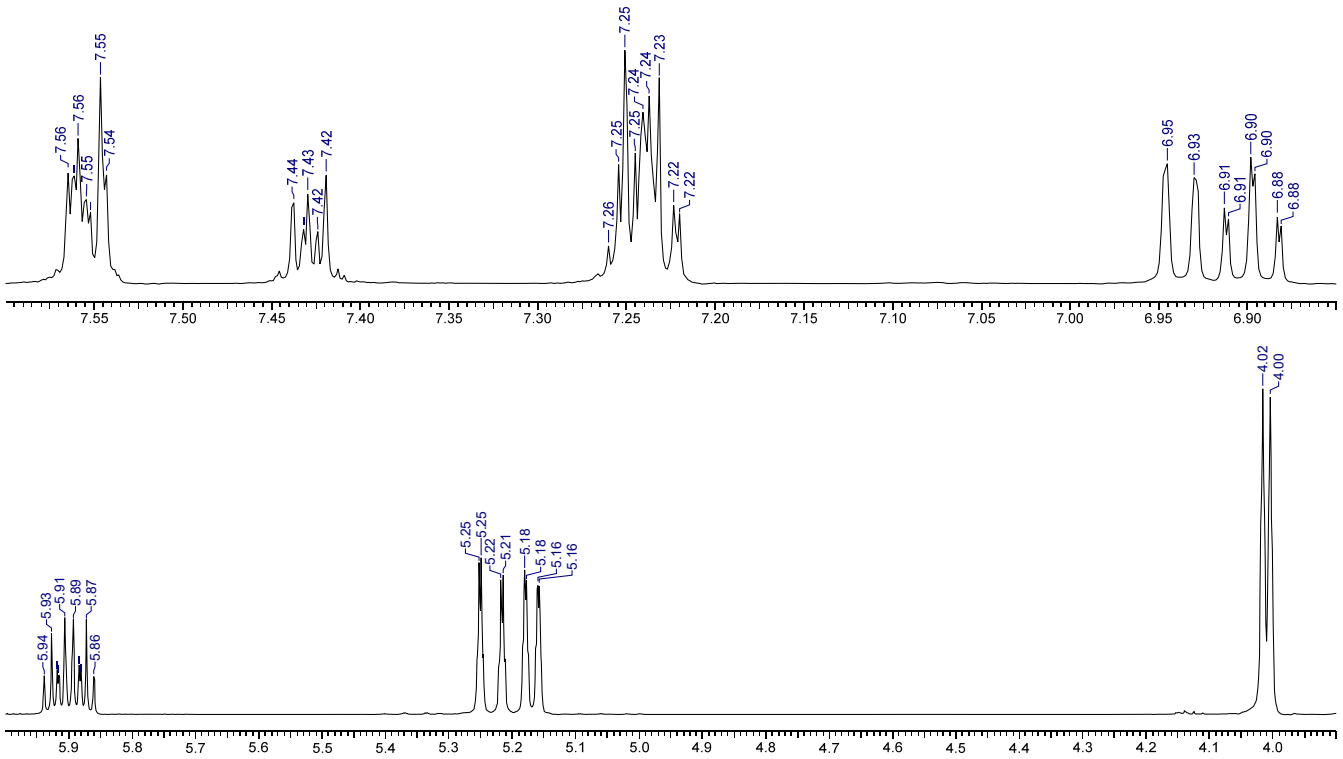
Enlarged view



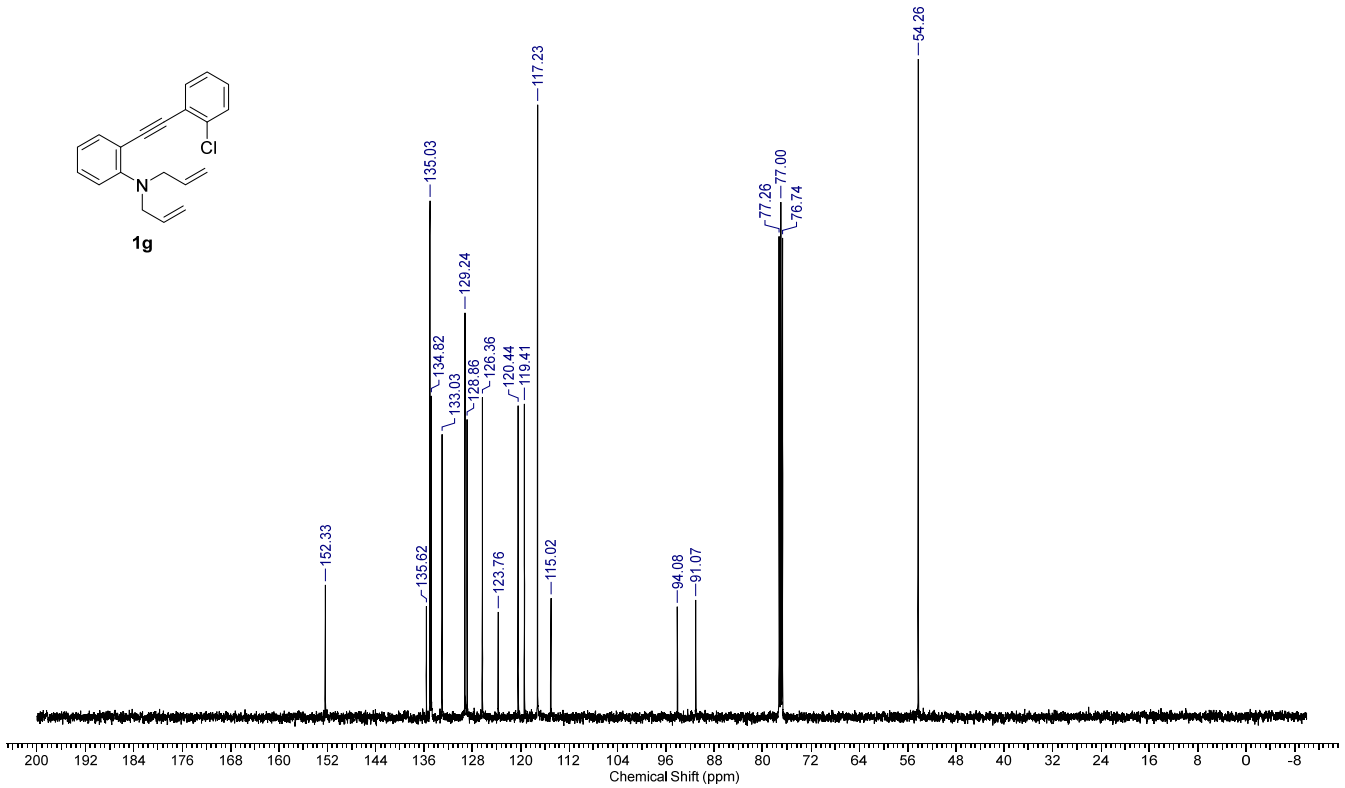
^{1}H NMR (500 MHz, $CDCl_3$) of **1g**



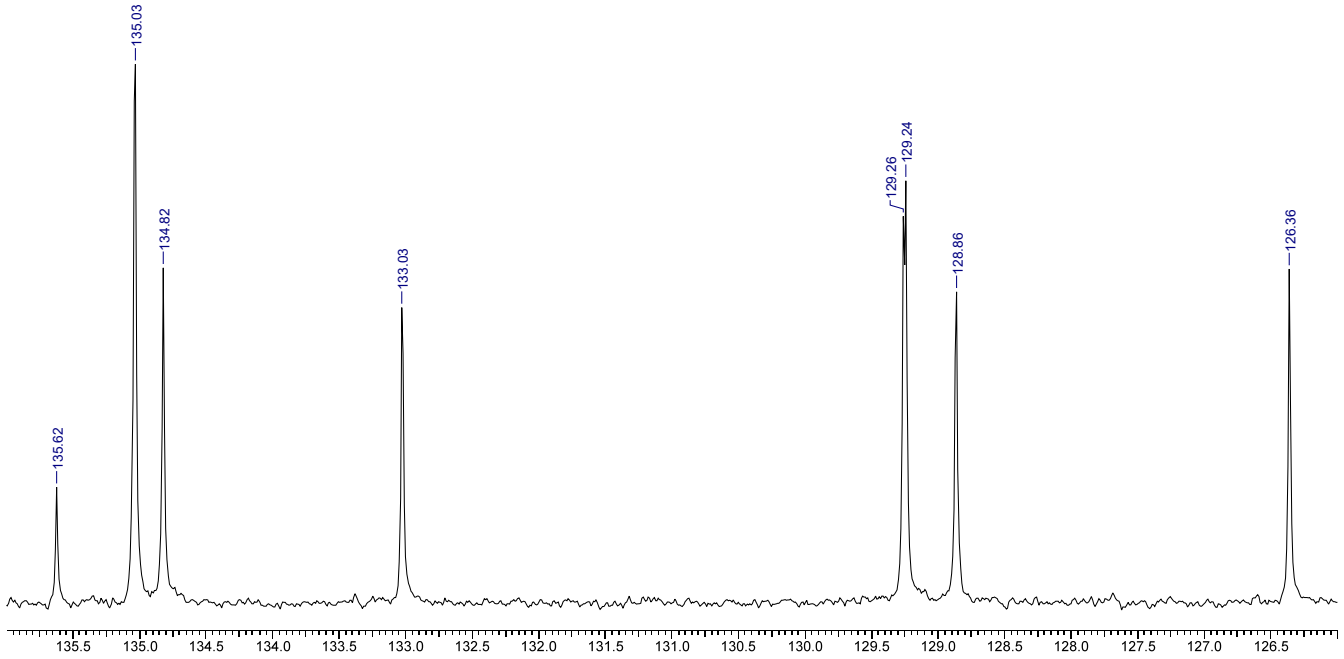
Enlarged view



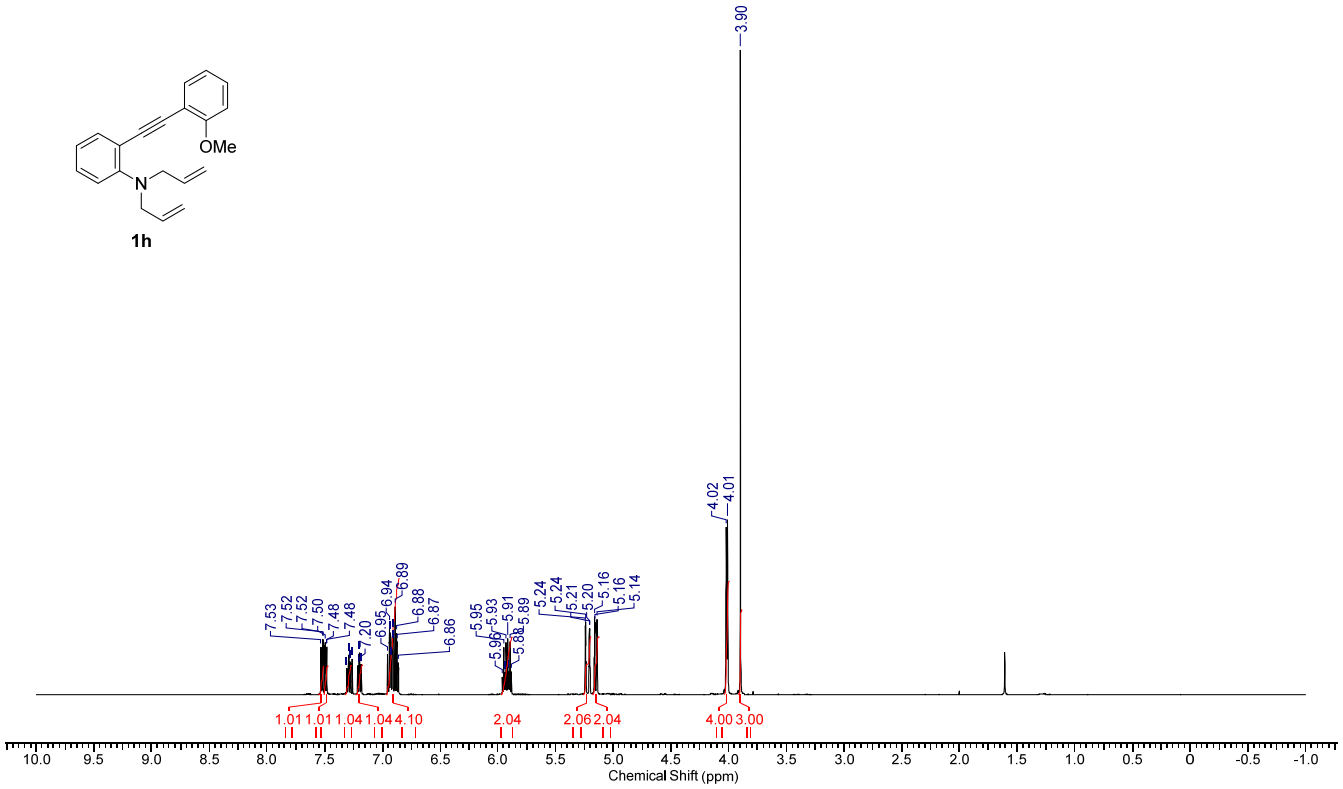
^{13}C NMR (125 MHz, $CDCl_3$) of **1g**



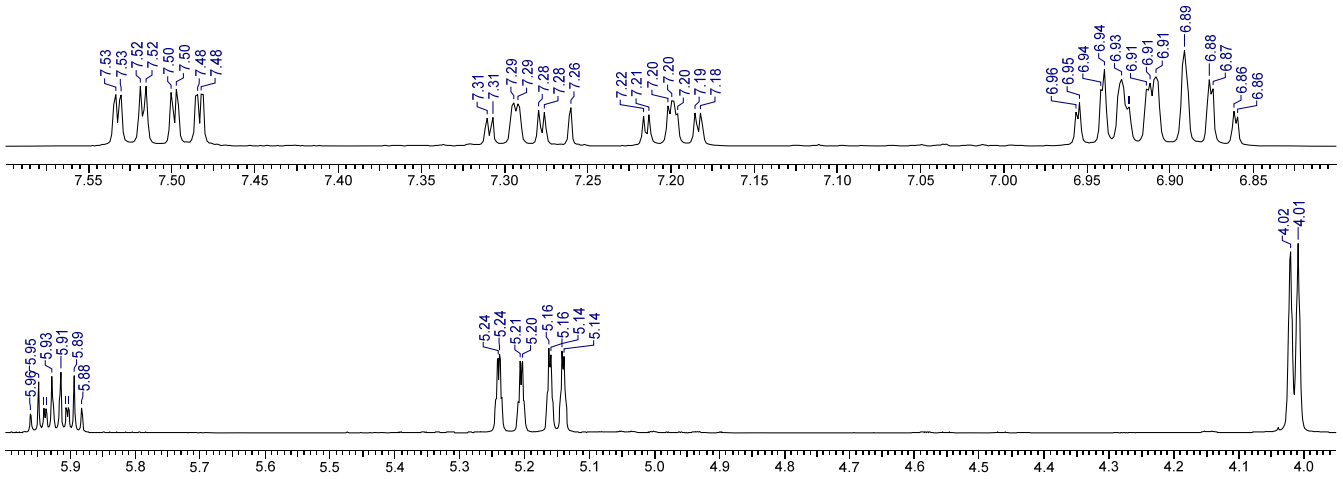
Enlarged view



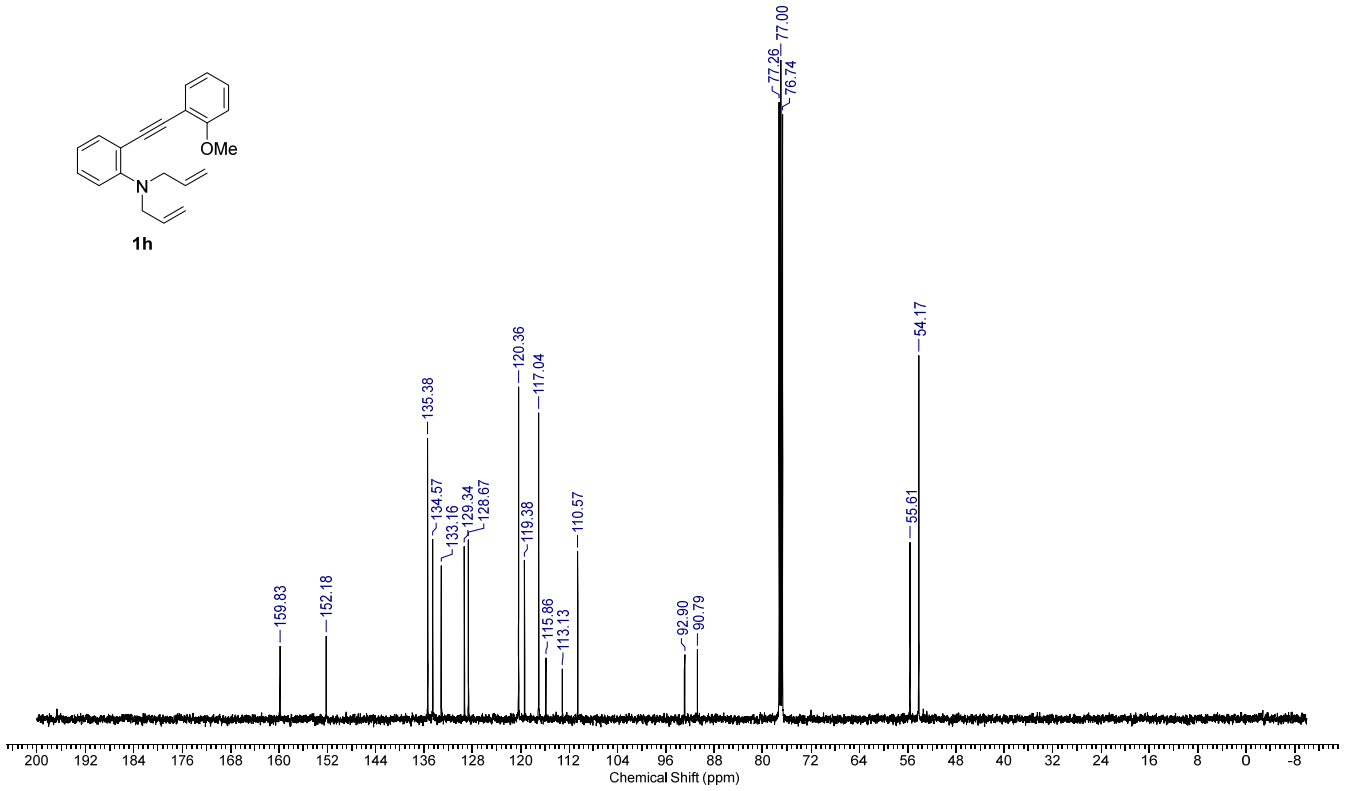
^{1}H NMR (500 MHz, $CDCl_3$) of **1h**



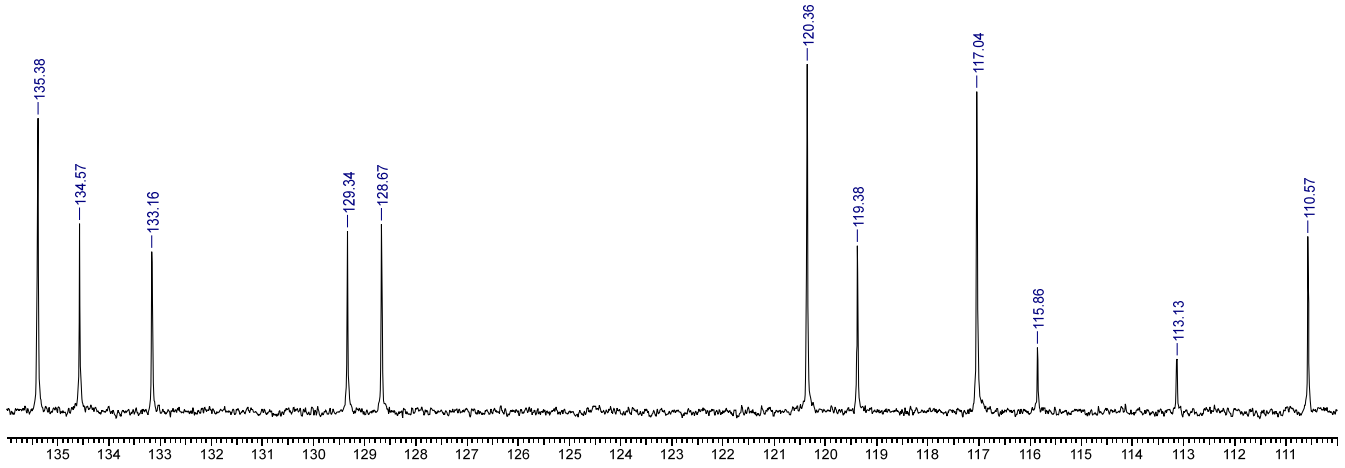
Enlarged view



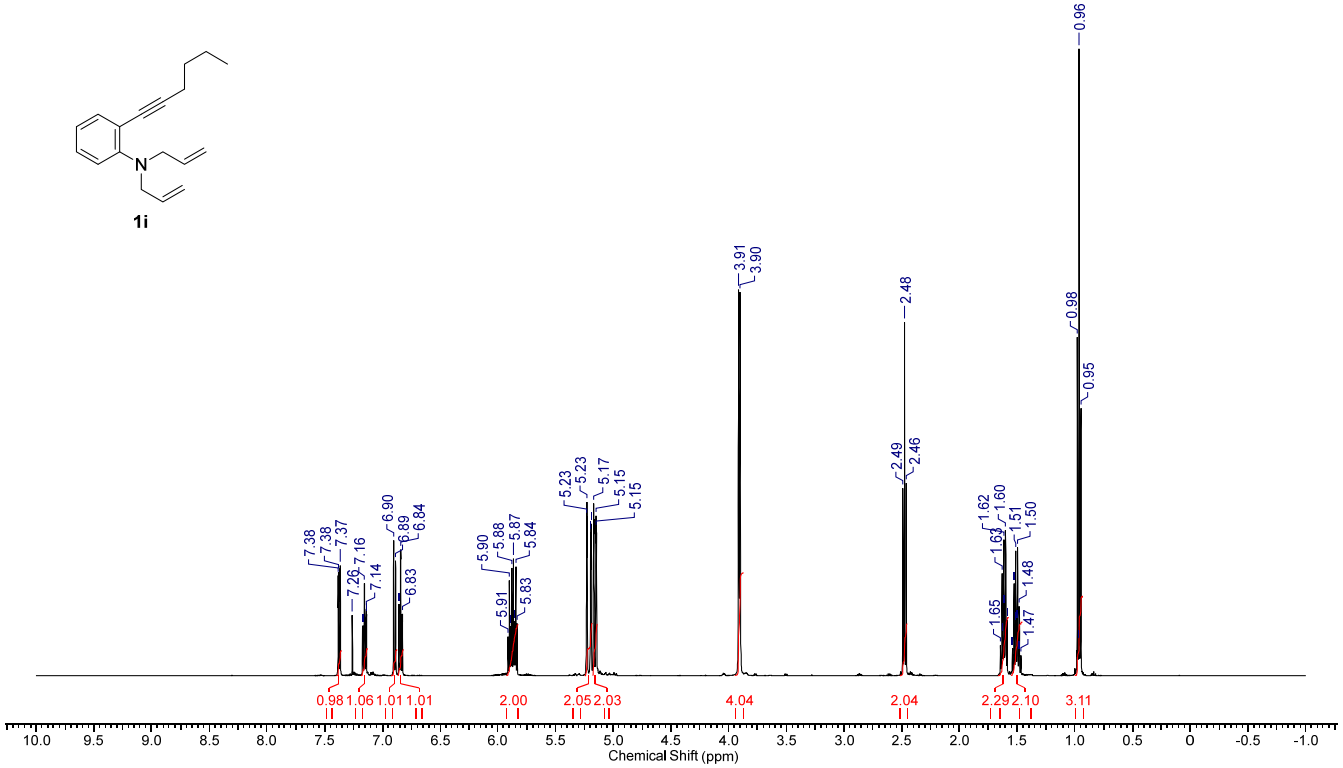
^{13}C NMR (125 MHz, $CDCl_3$) of **1h**



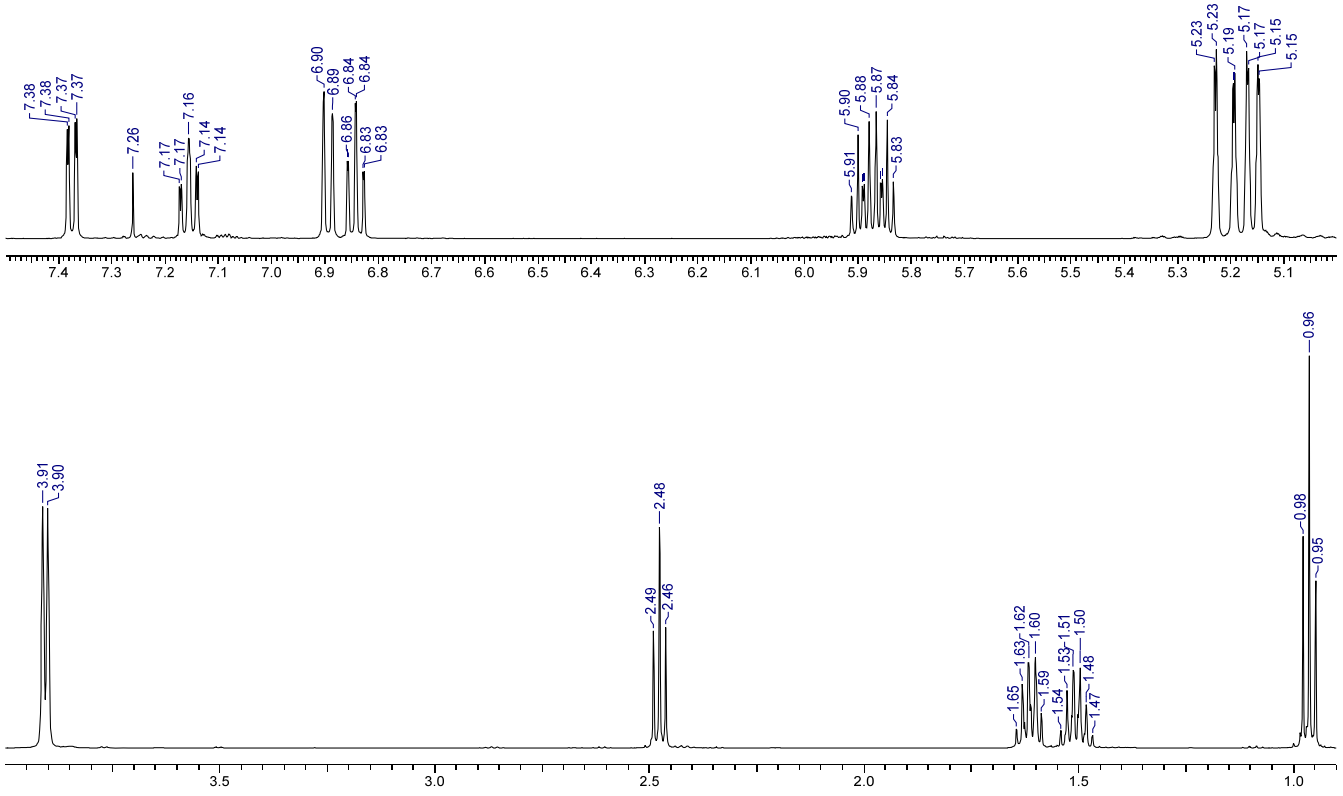
Enlarged view



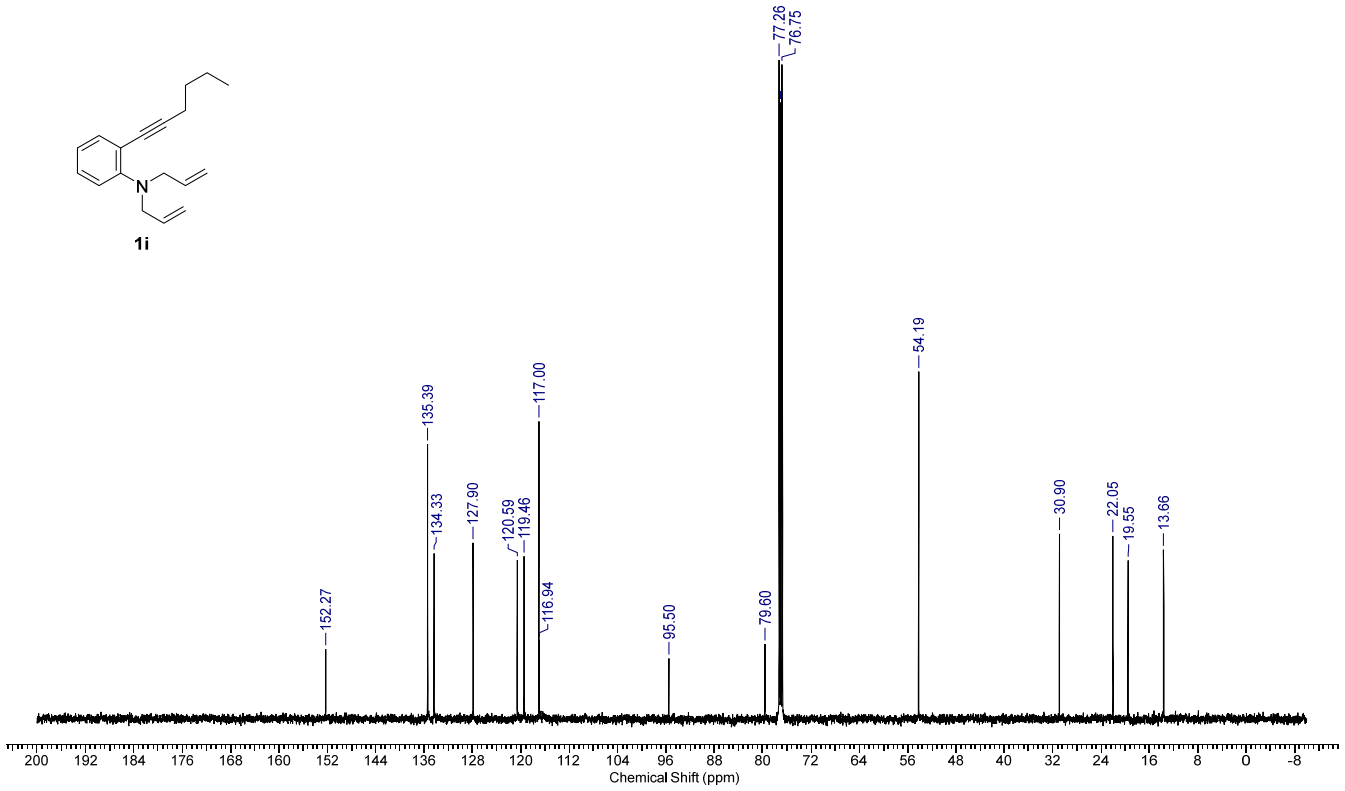
^{1}H NMR (500 MHz, $CDCl_3$) of **1i**



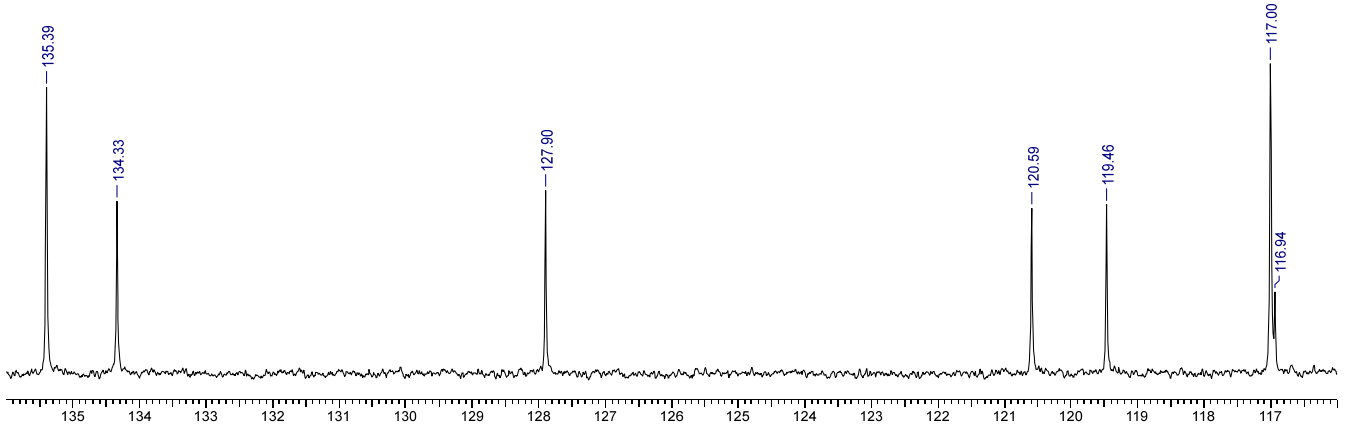
Enlarged view



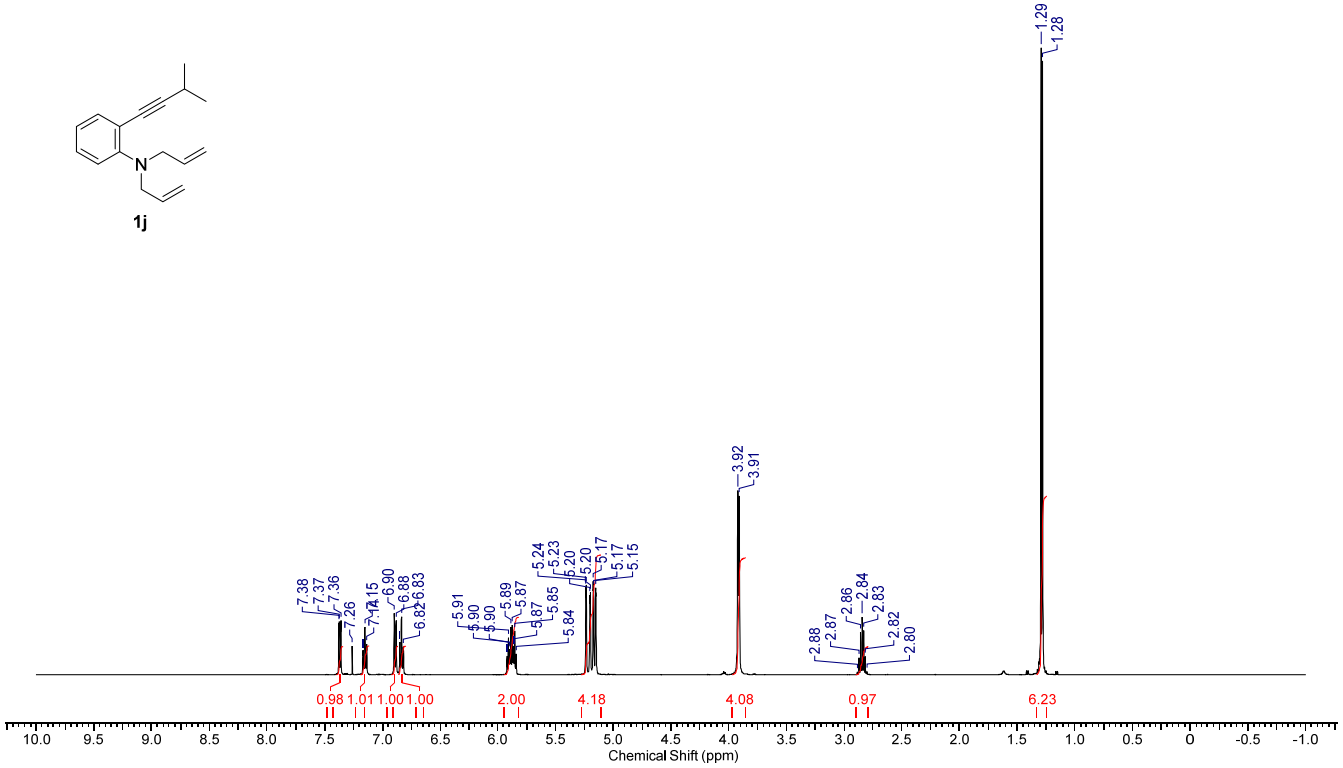
^{13}C NMR (125 MHz, $CDCl_3$) of **1i**



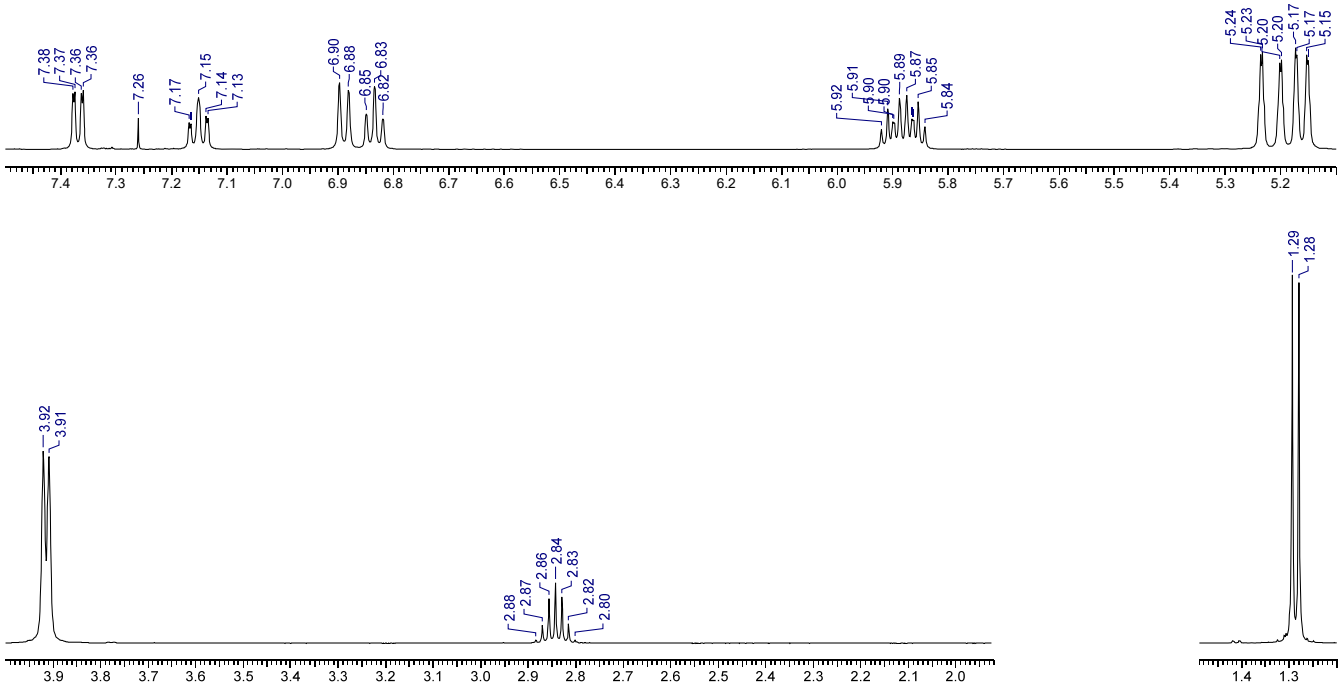
Enlarged view



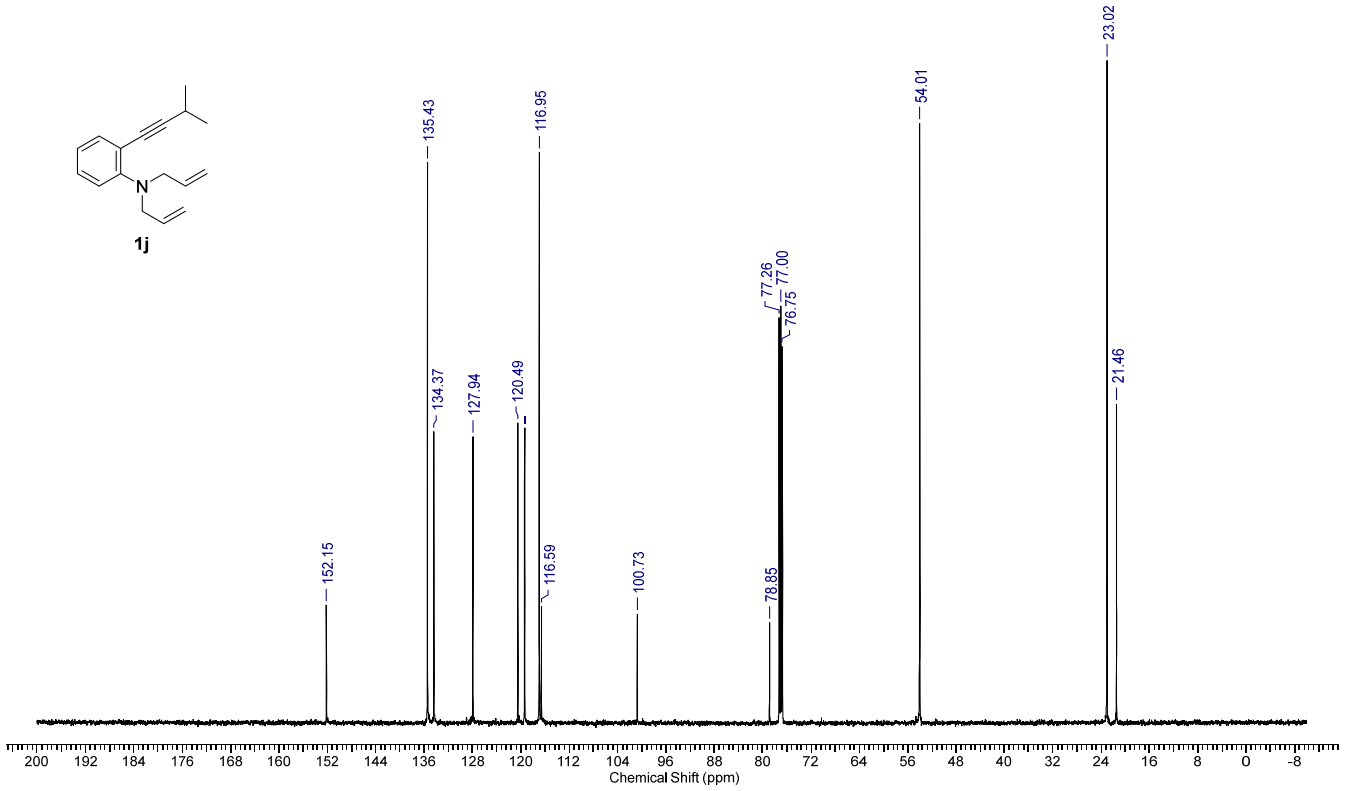
^{1}H NMR (500 MHz, $CDCl_3$) of **1j**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **1j**



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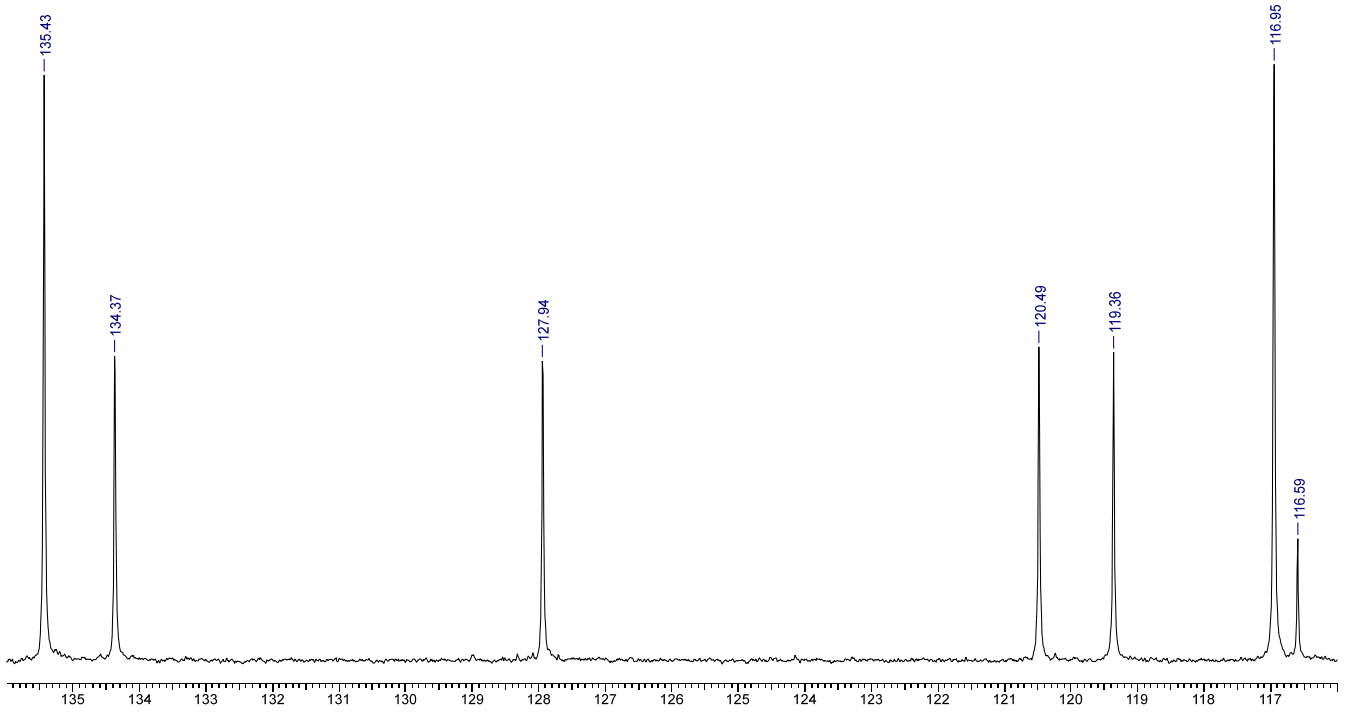
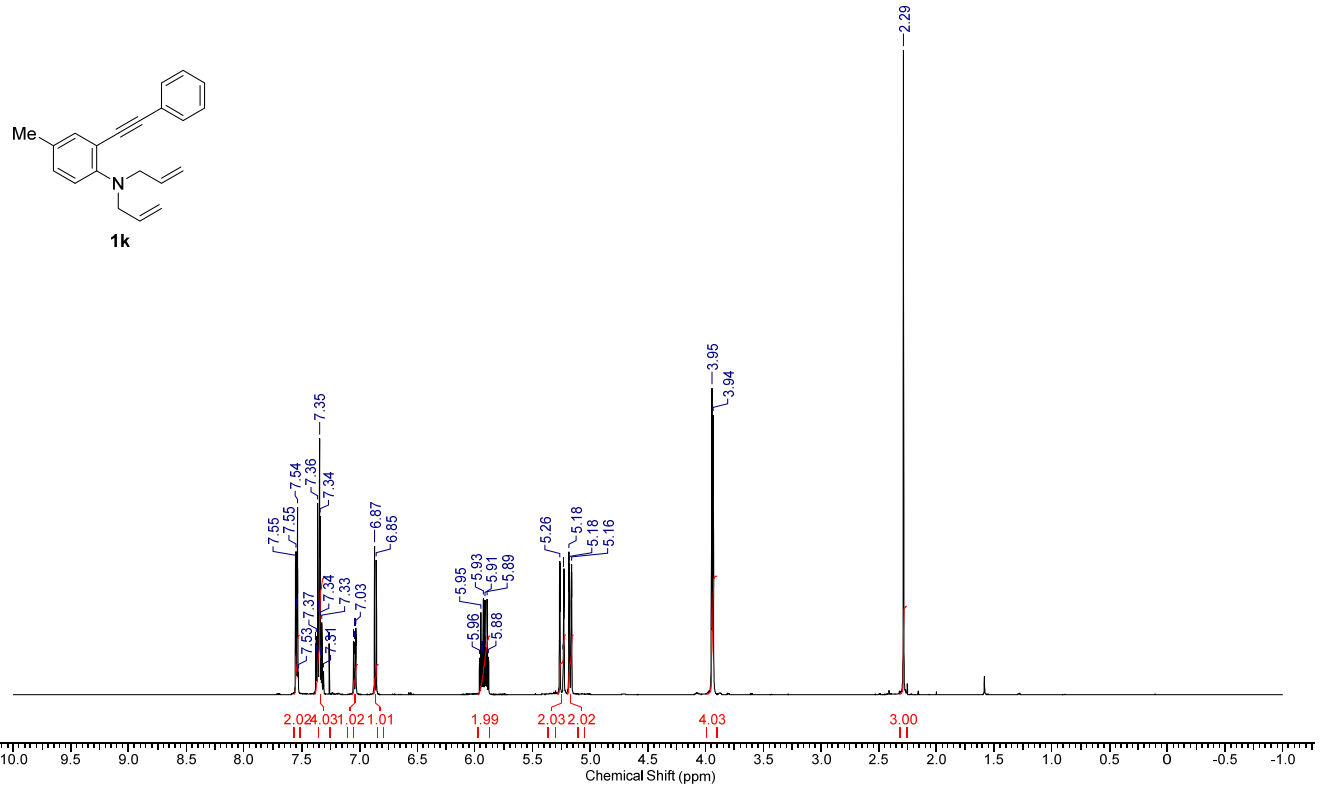


^{1}H NMR (500 MHz, $CDCl_3$) of **1k**



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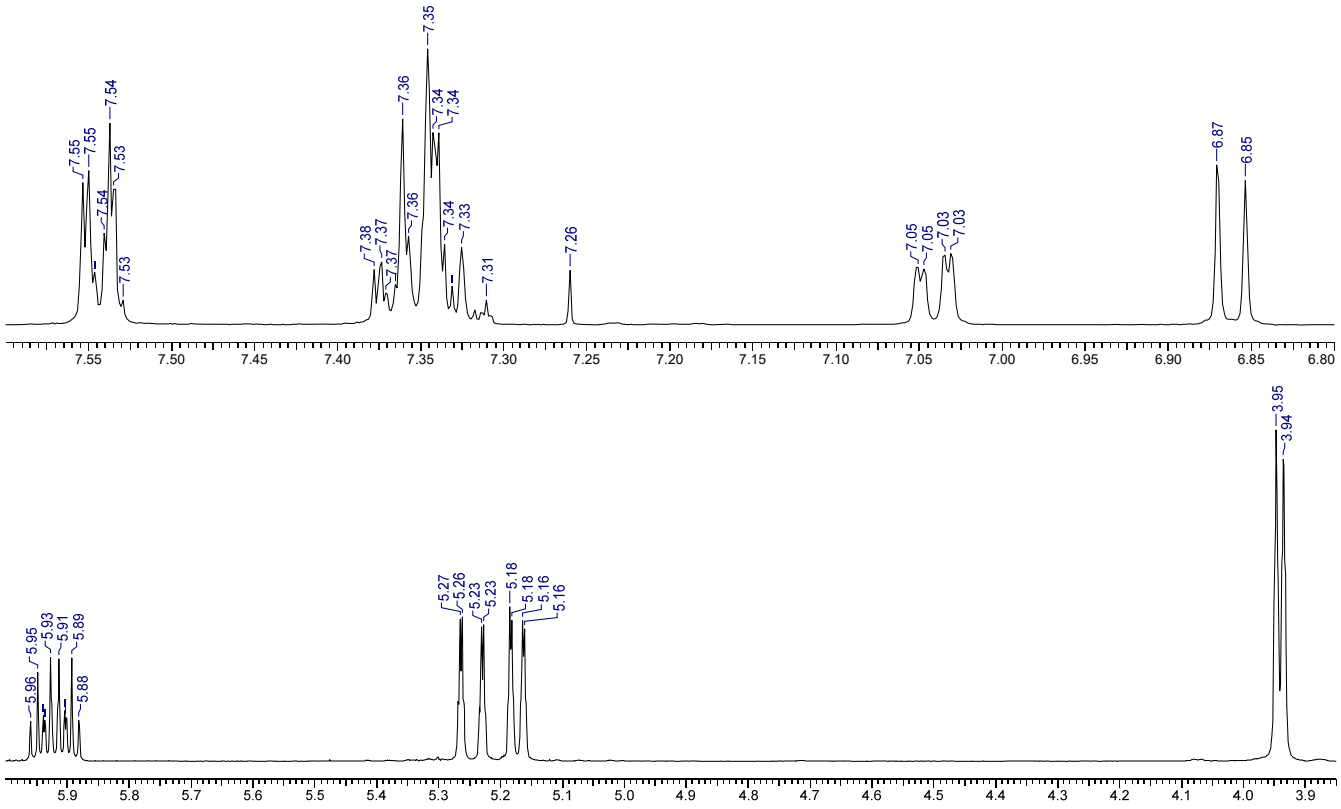
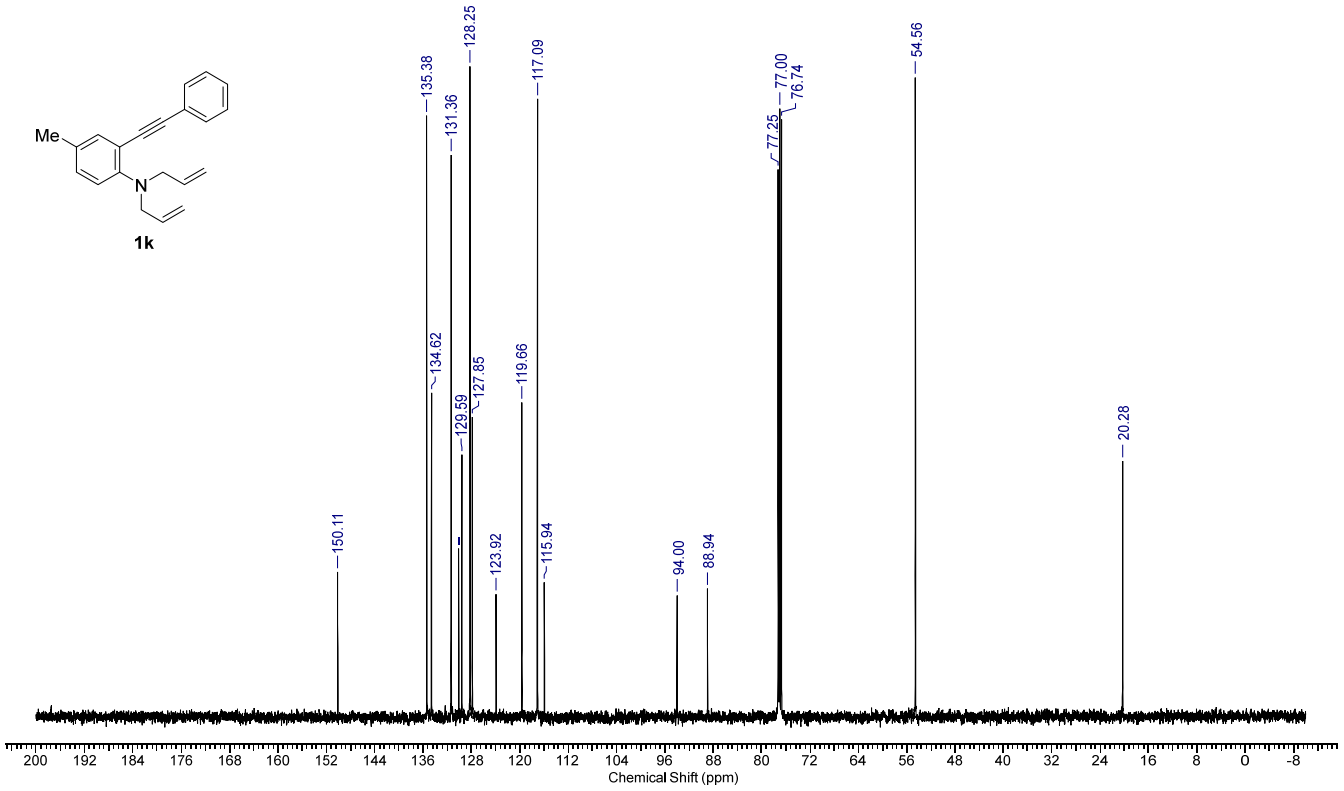


^{13}C NMR (125 MHz, $CDCl_3$) of **1k**



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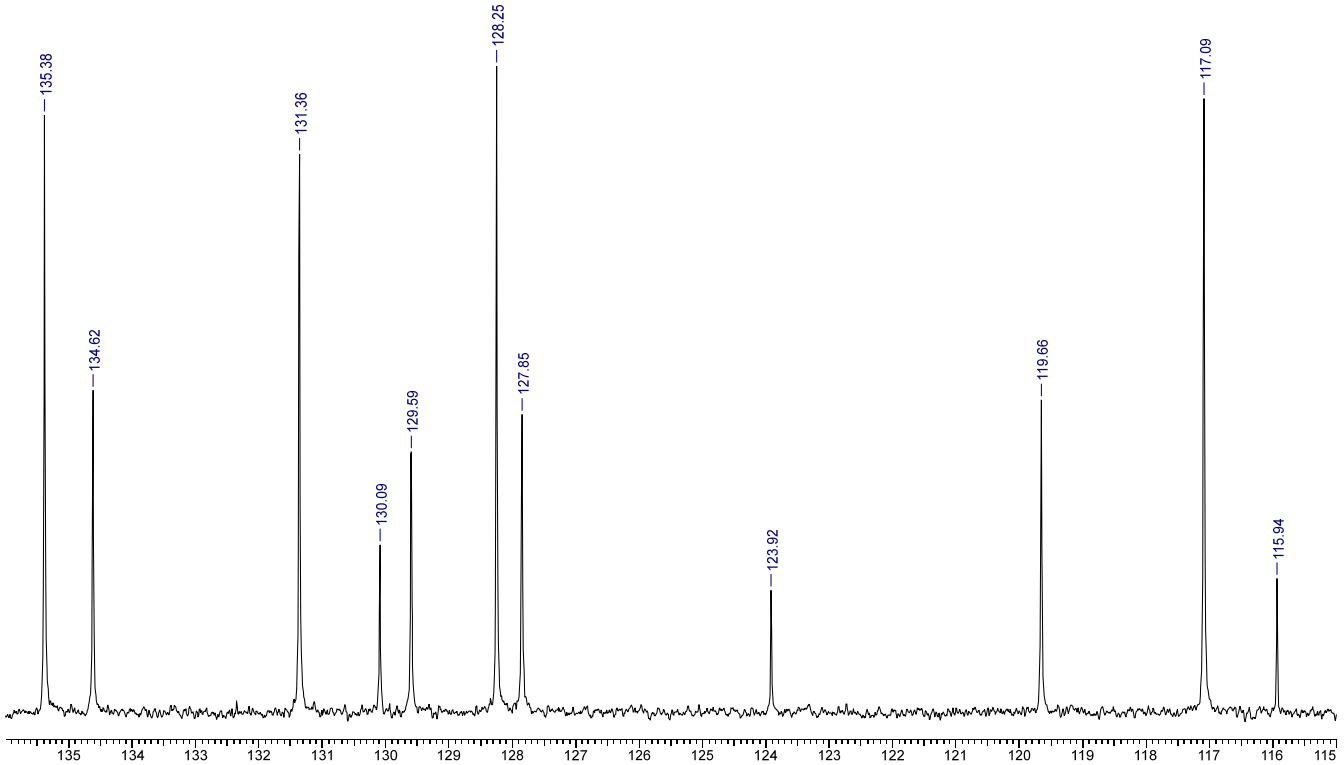
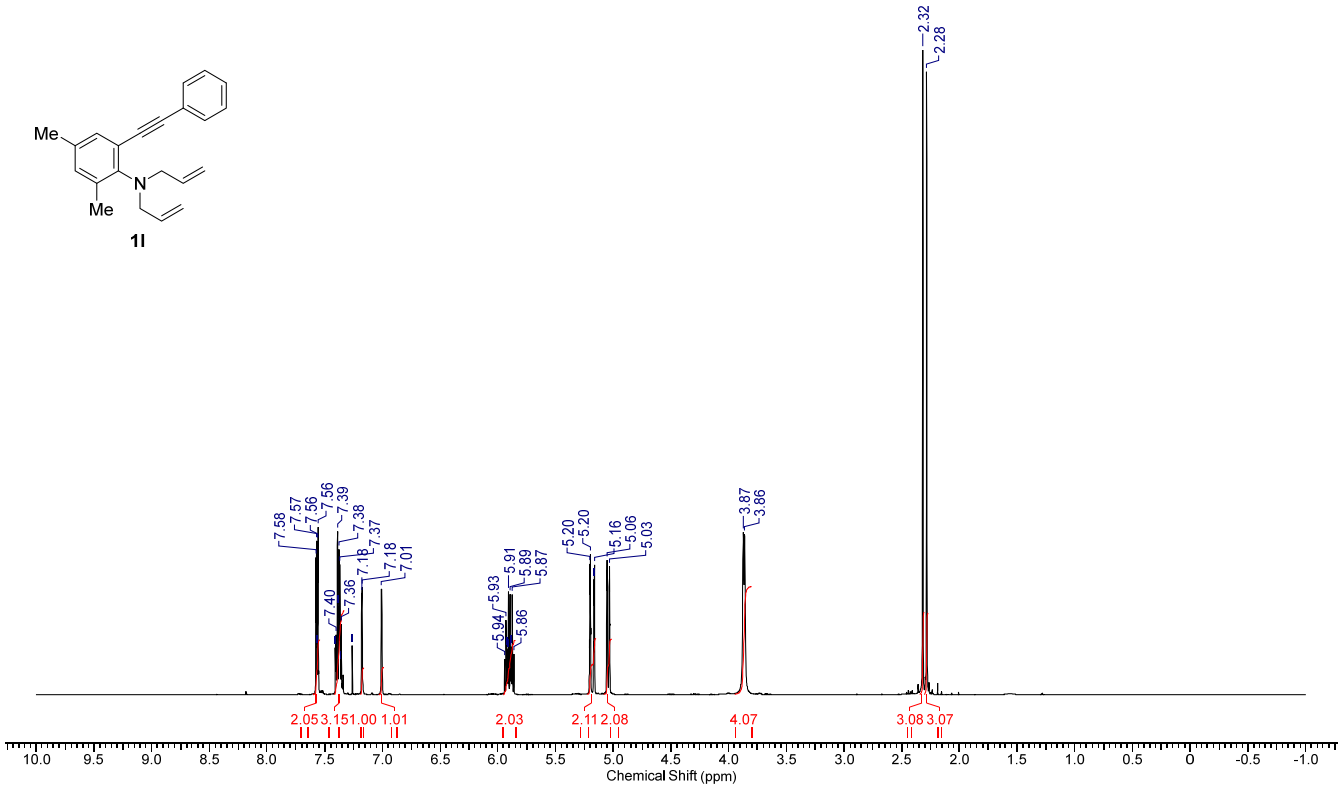


^{1}H NMR (500 MHz, $CDCl_3$) of **1l**



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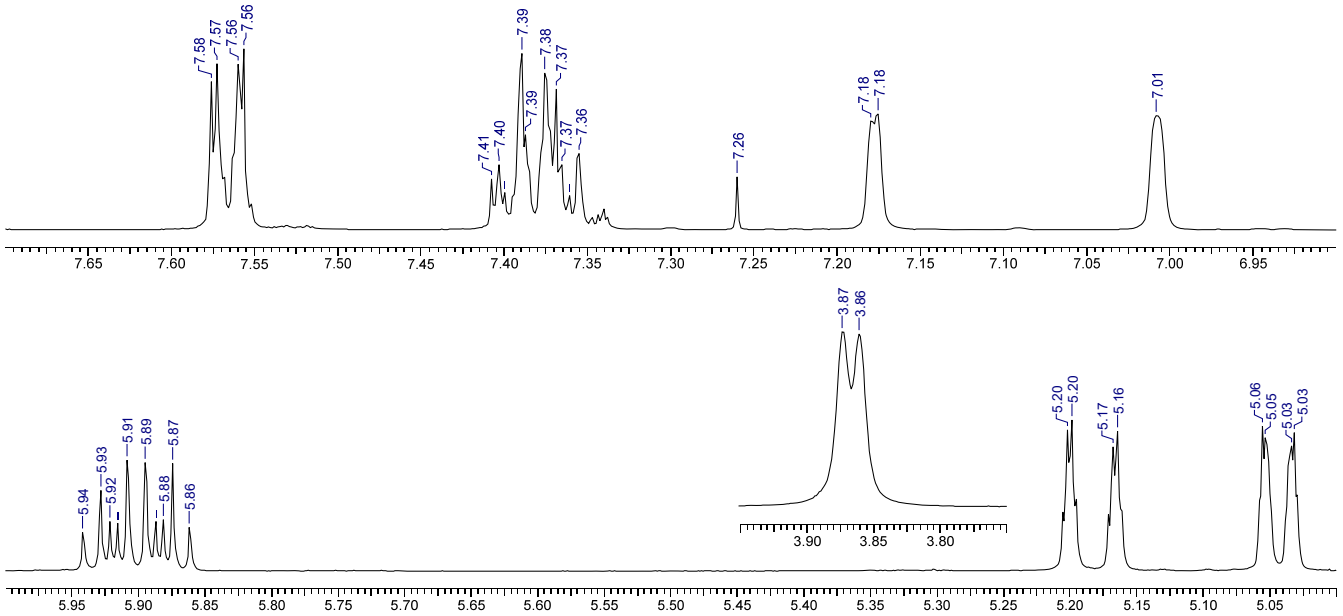
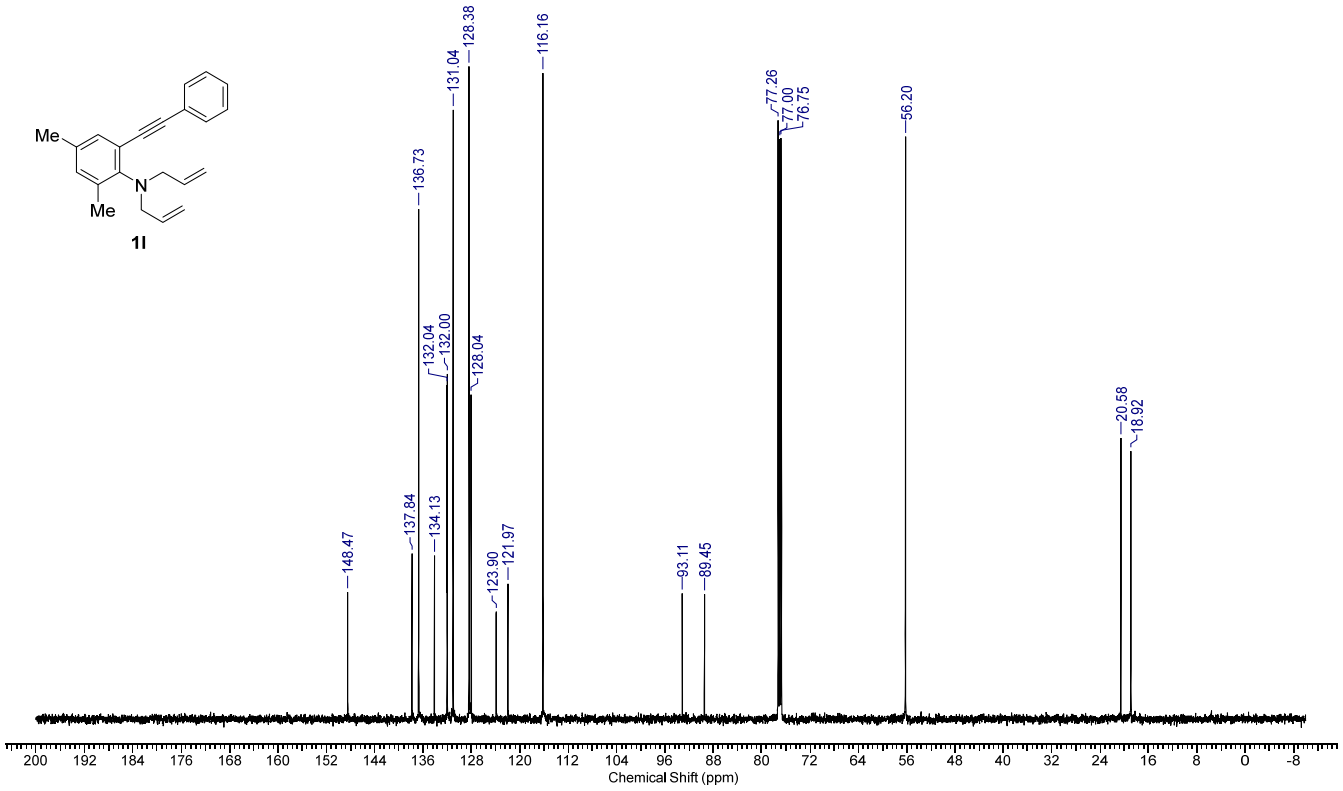
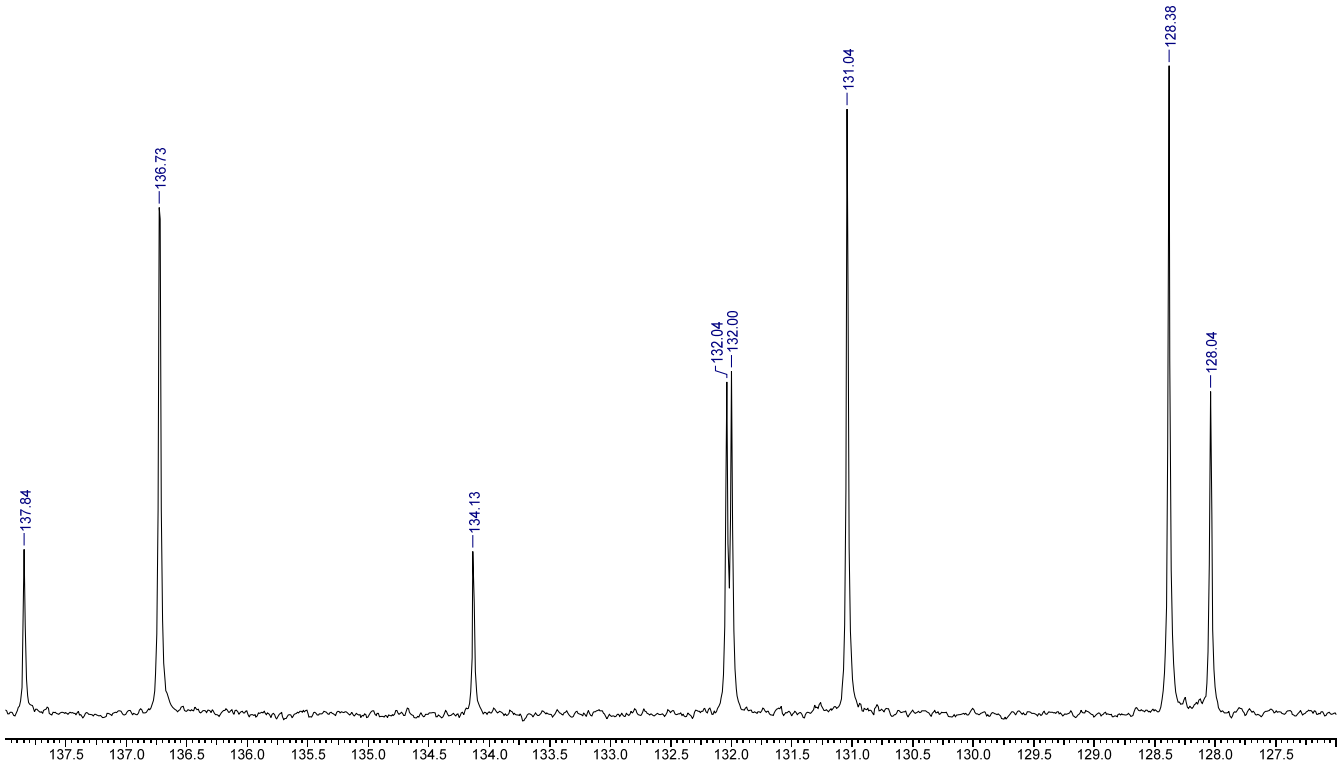


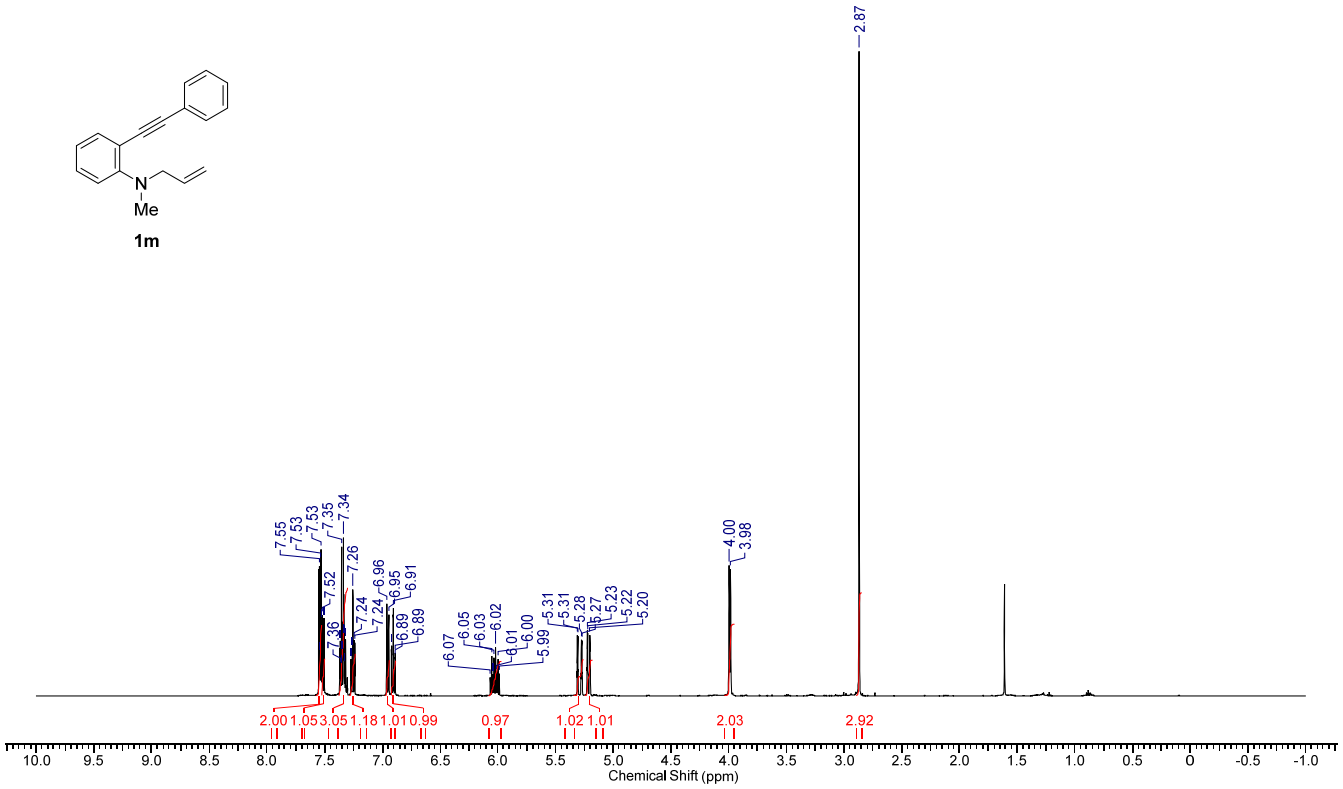
^{13}C NMR (125 MHz, $CDCl_3$) of **1l**



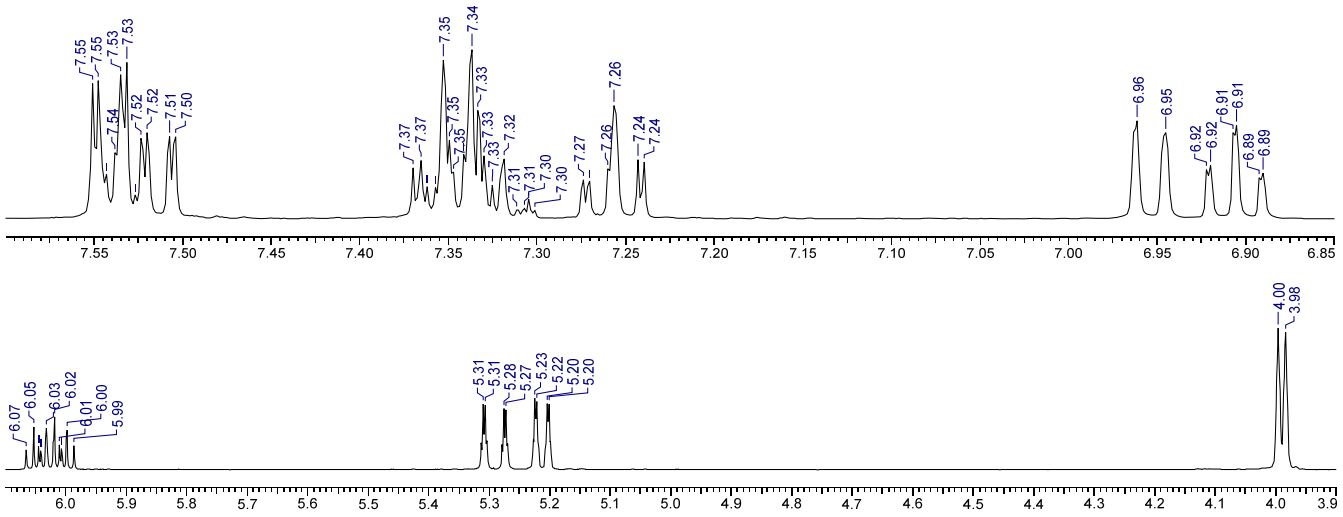
Enlarged view



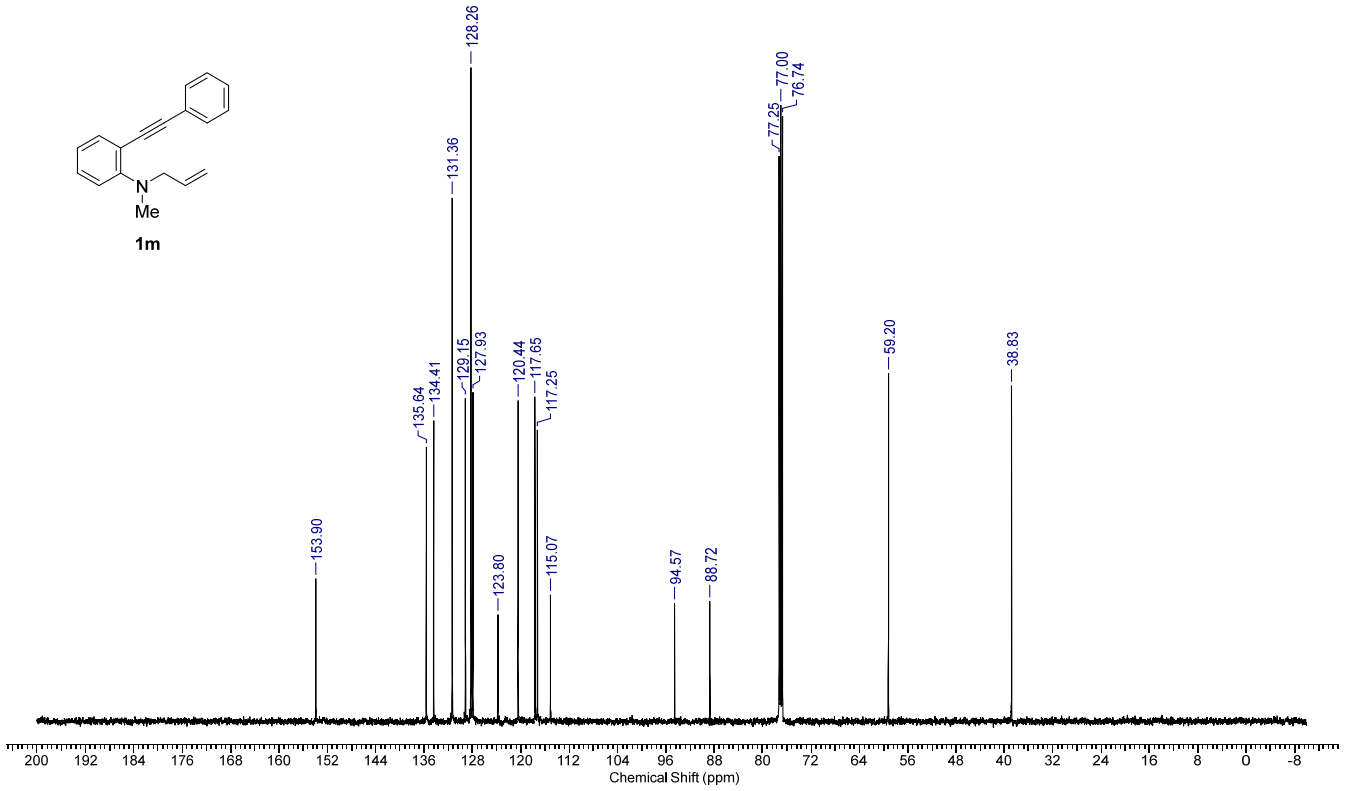
^{1}H NMR (500 MHz, $CDCl_3$) of **1m**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **1m**



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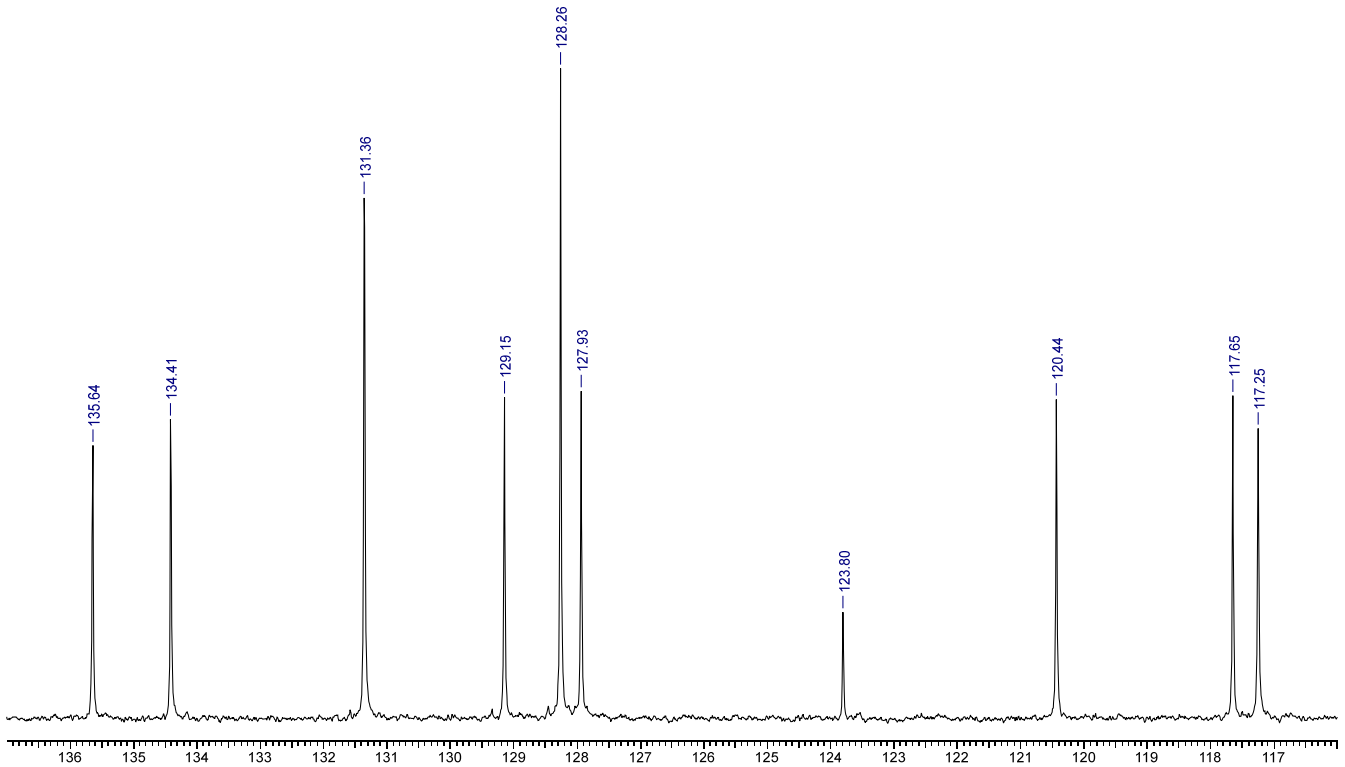
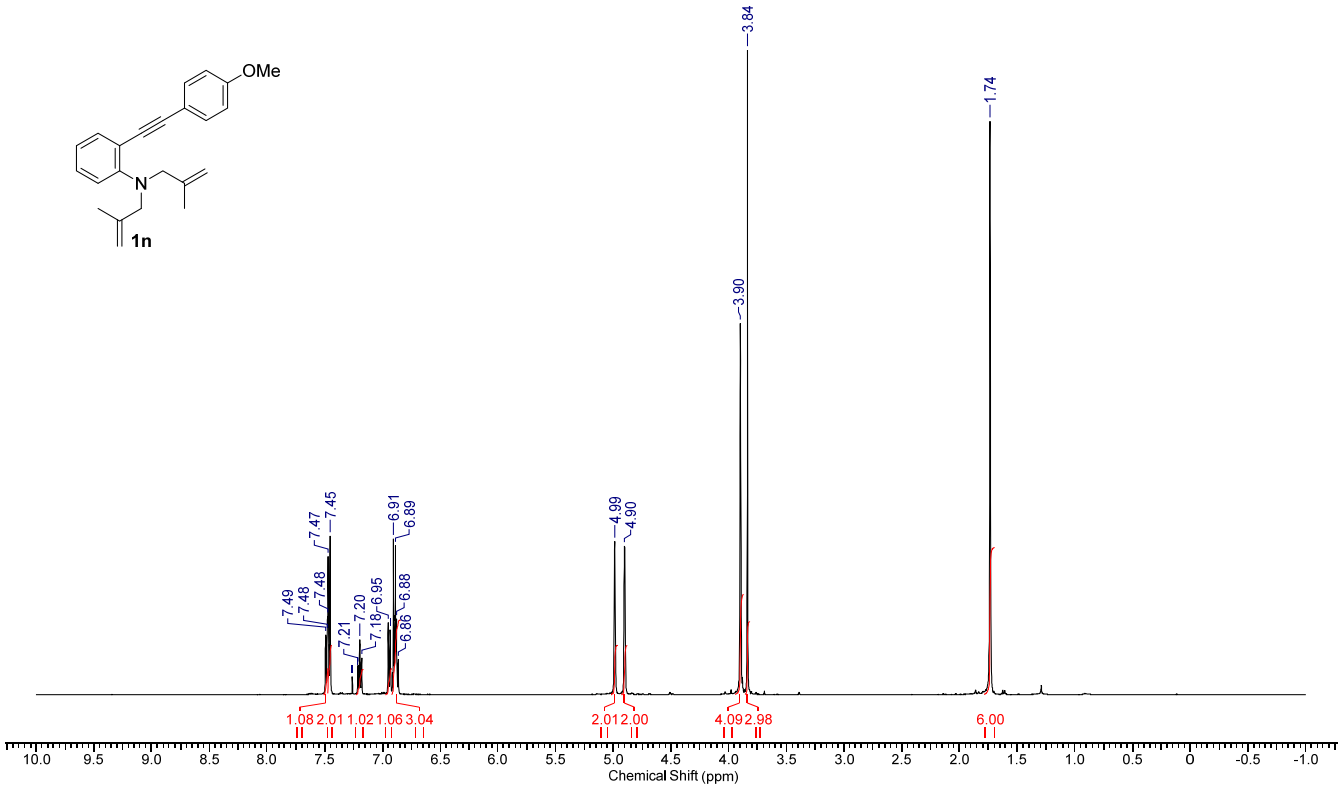
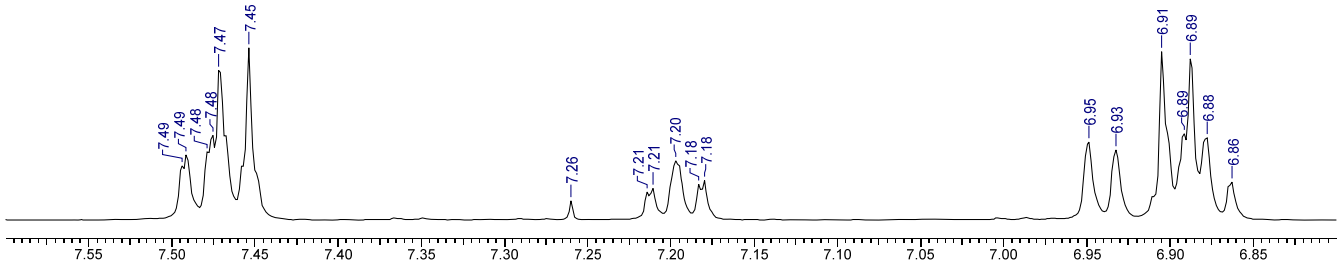


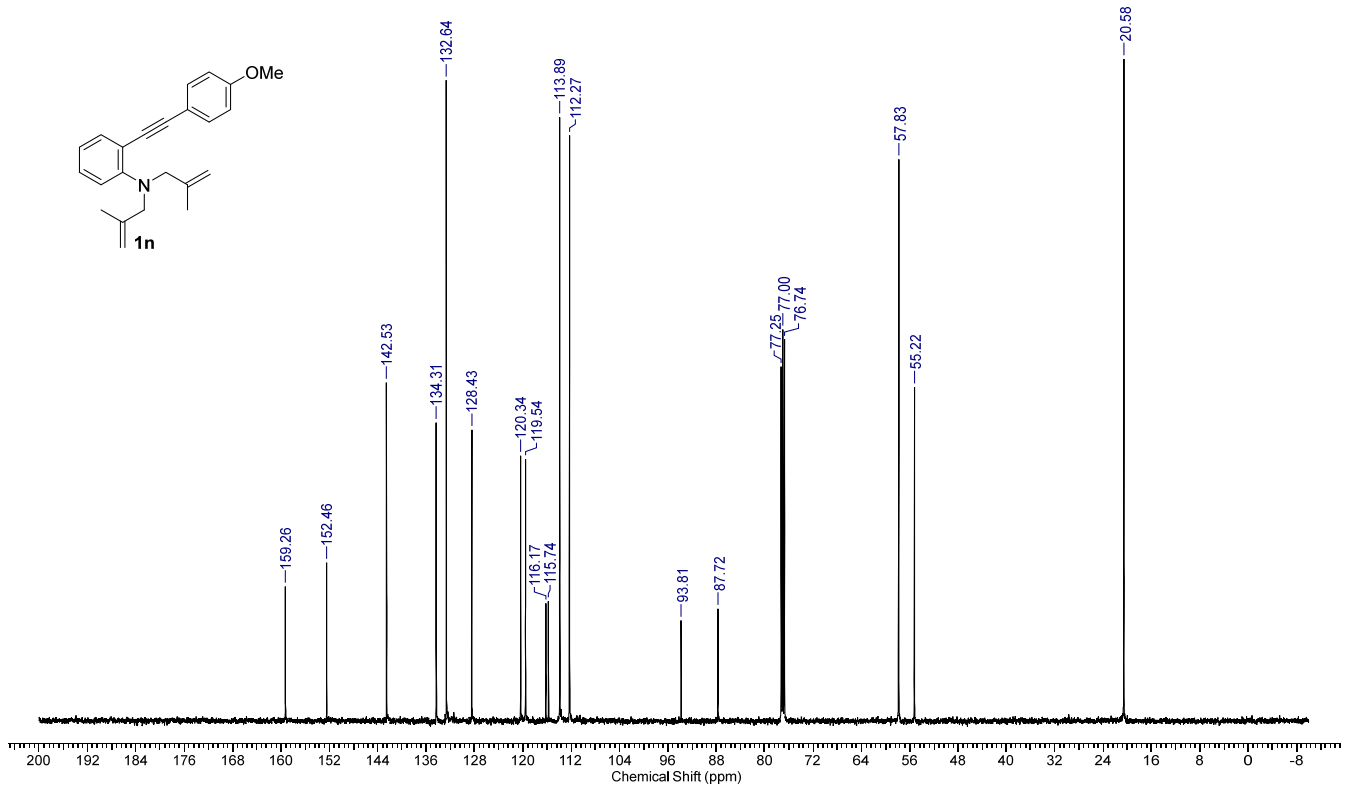
^{1}H NMR (500 MHz, $CDCl_3$) of **1n**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **1n**



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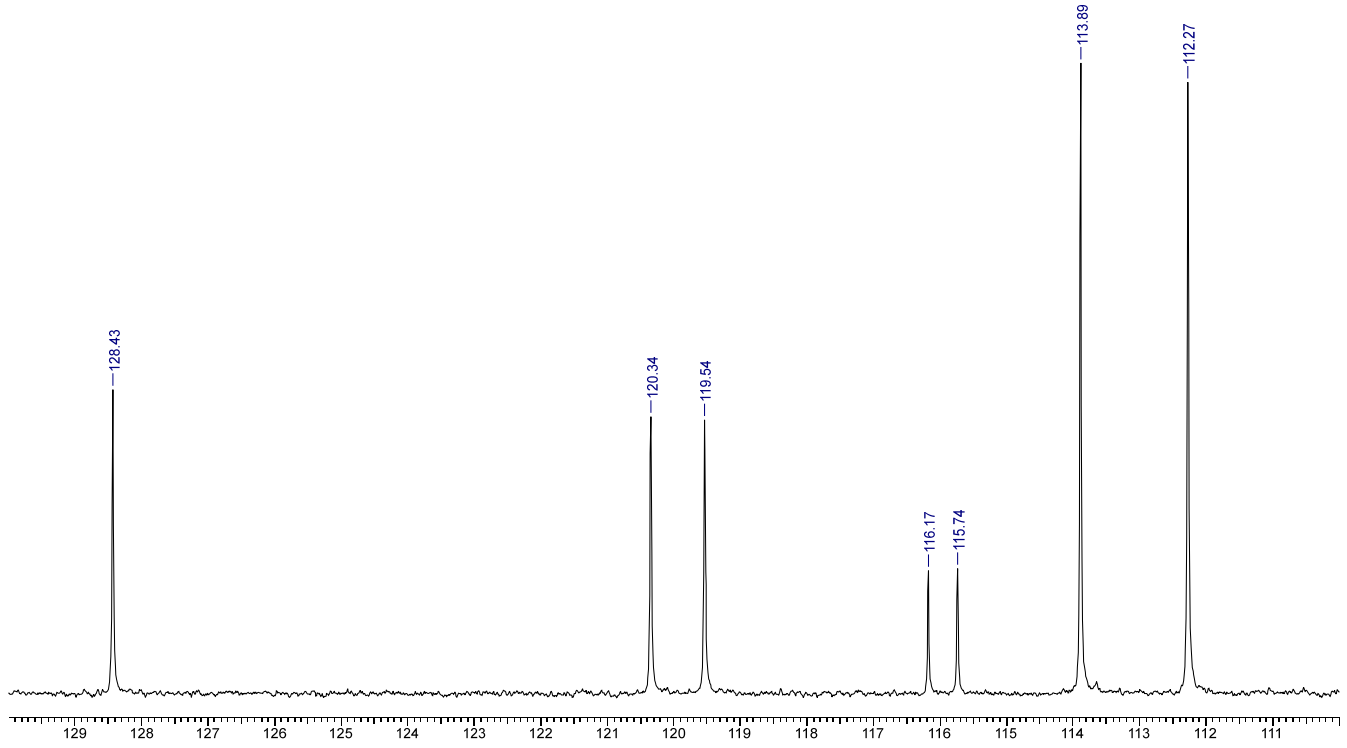
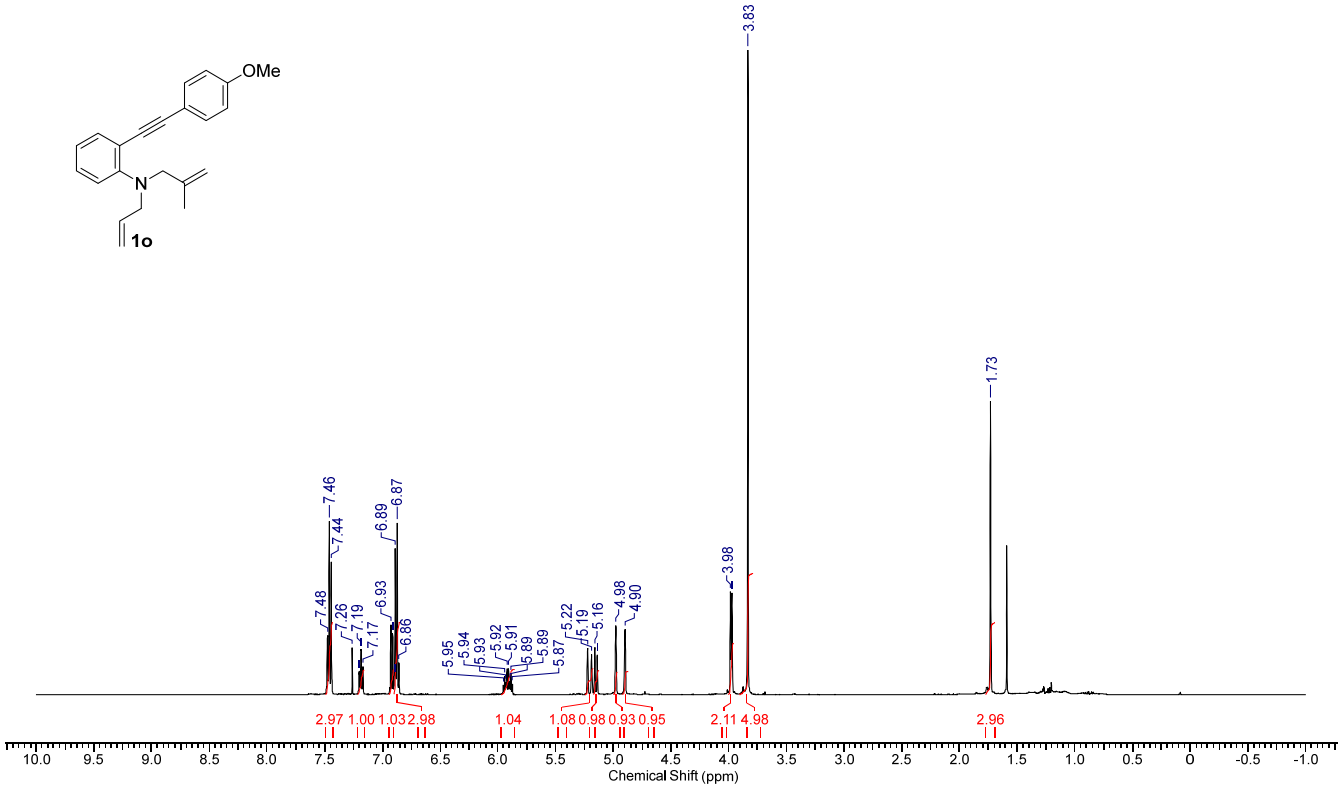
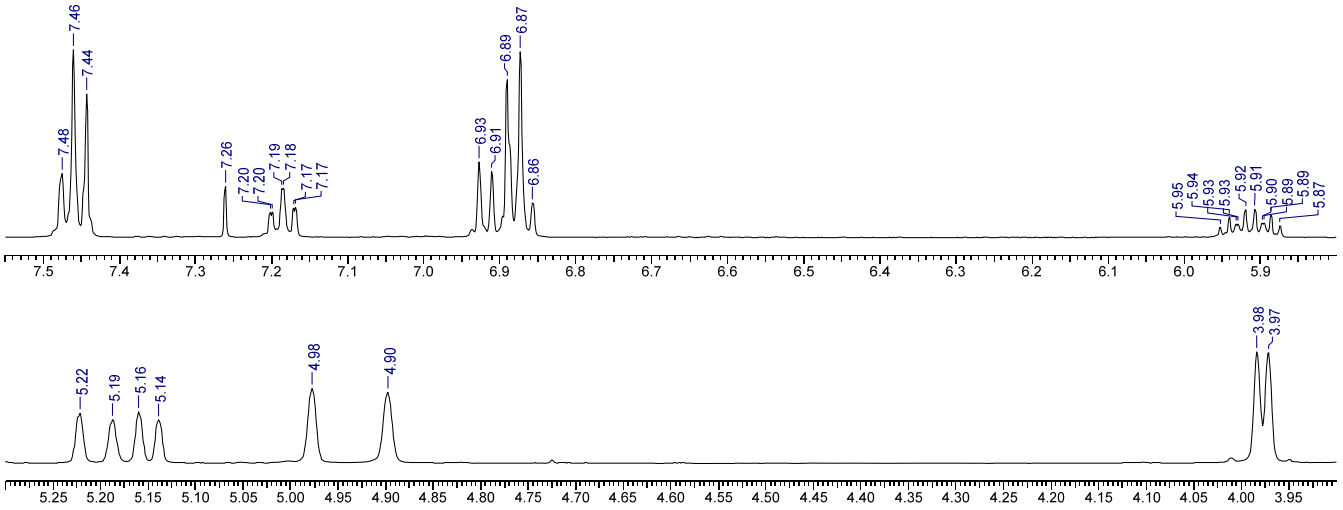


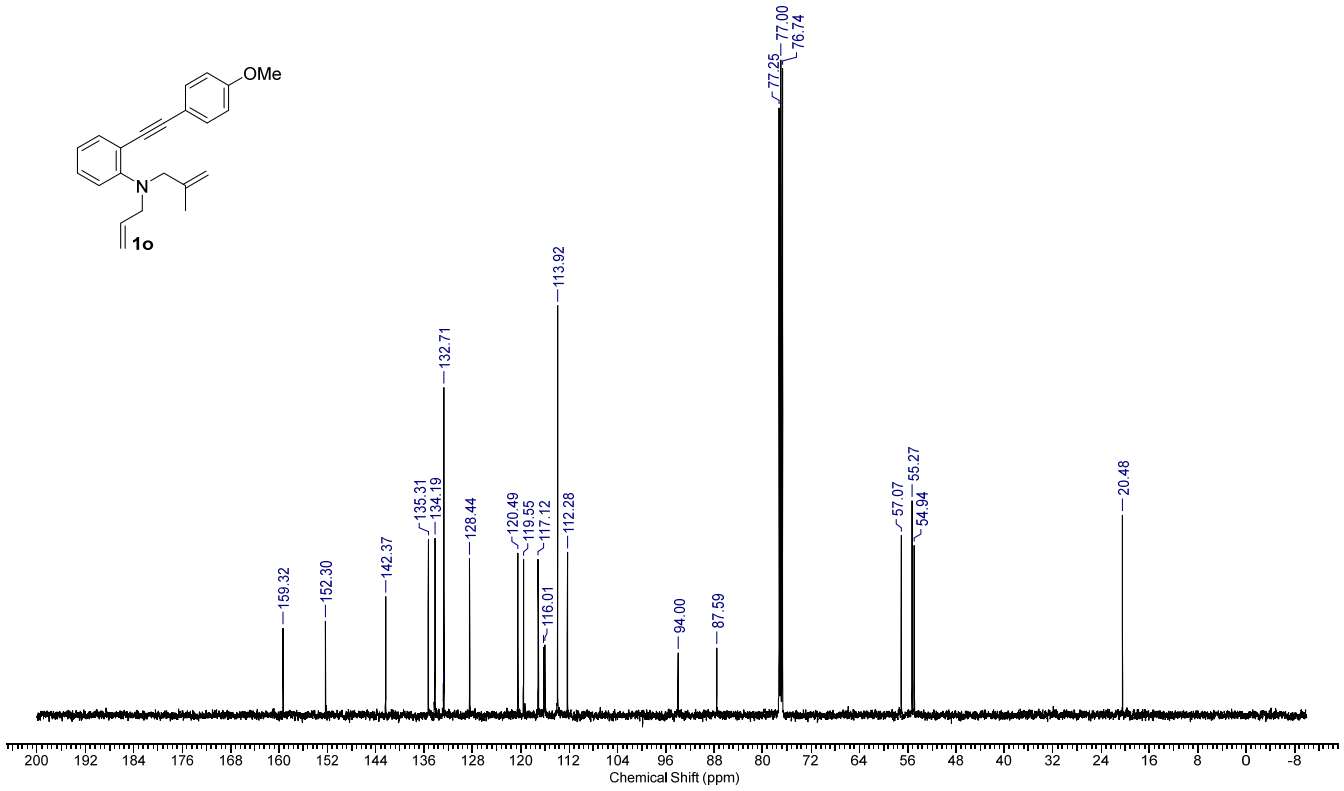
^{1}H NMR (500 MHz, $CDCl_3$) of **1o**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **1o**



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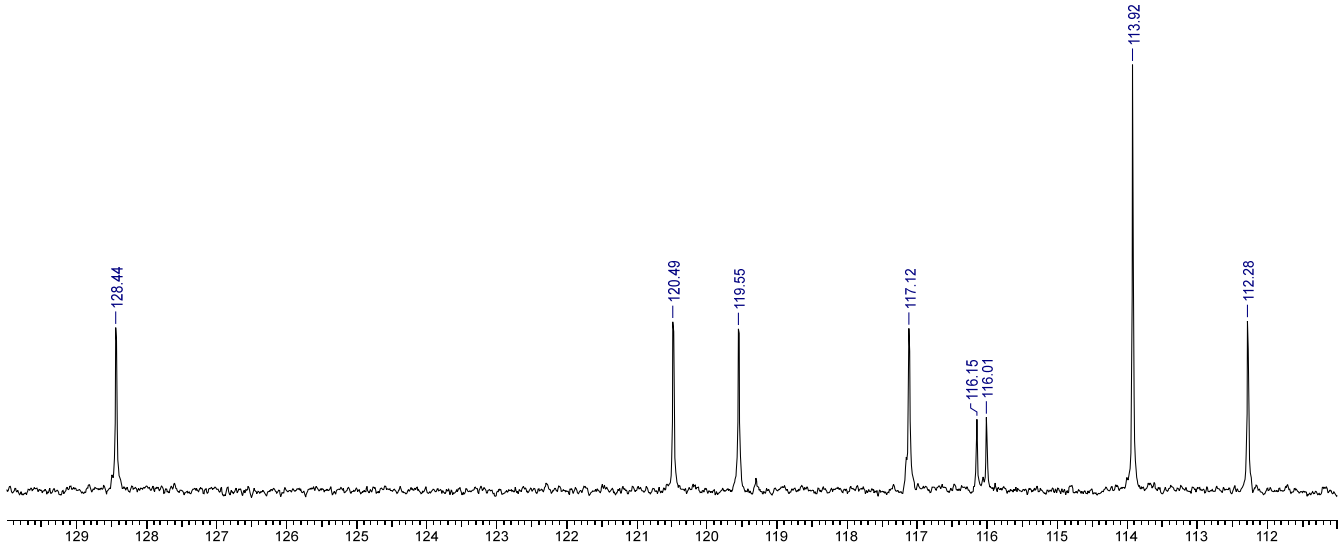
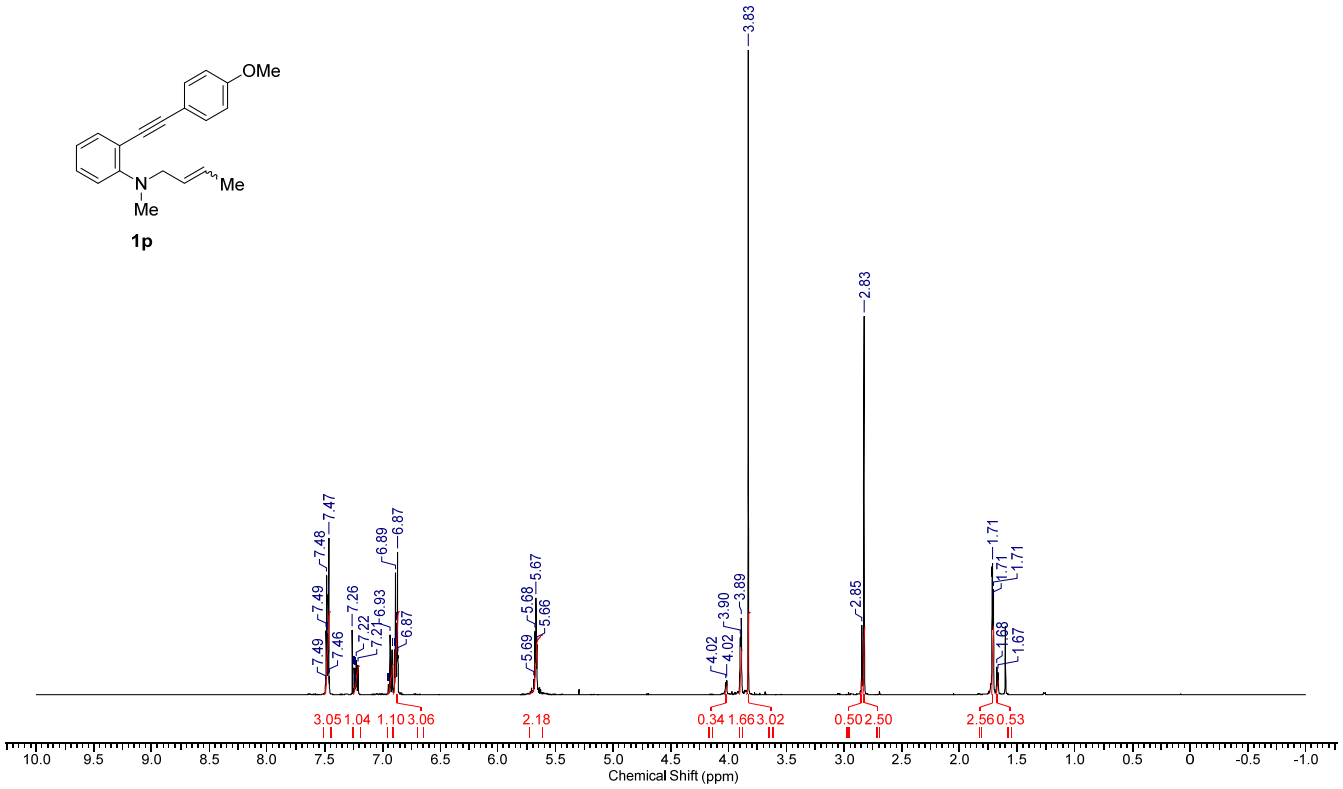


^{1}H NMR (500 MHz, $CDCl_3$) of **1p** (83:17 mixture of geometrical isomers)



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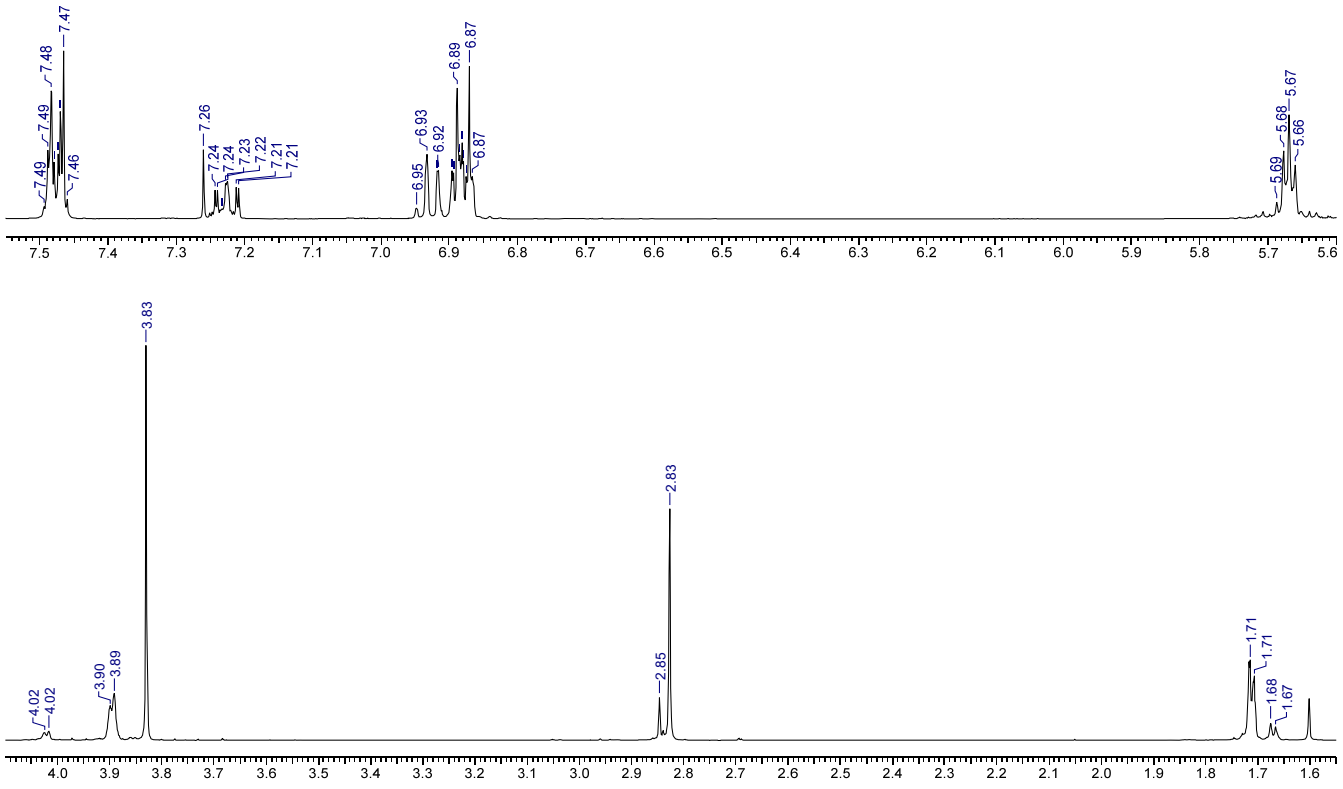
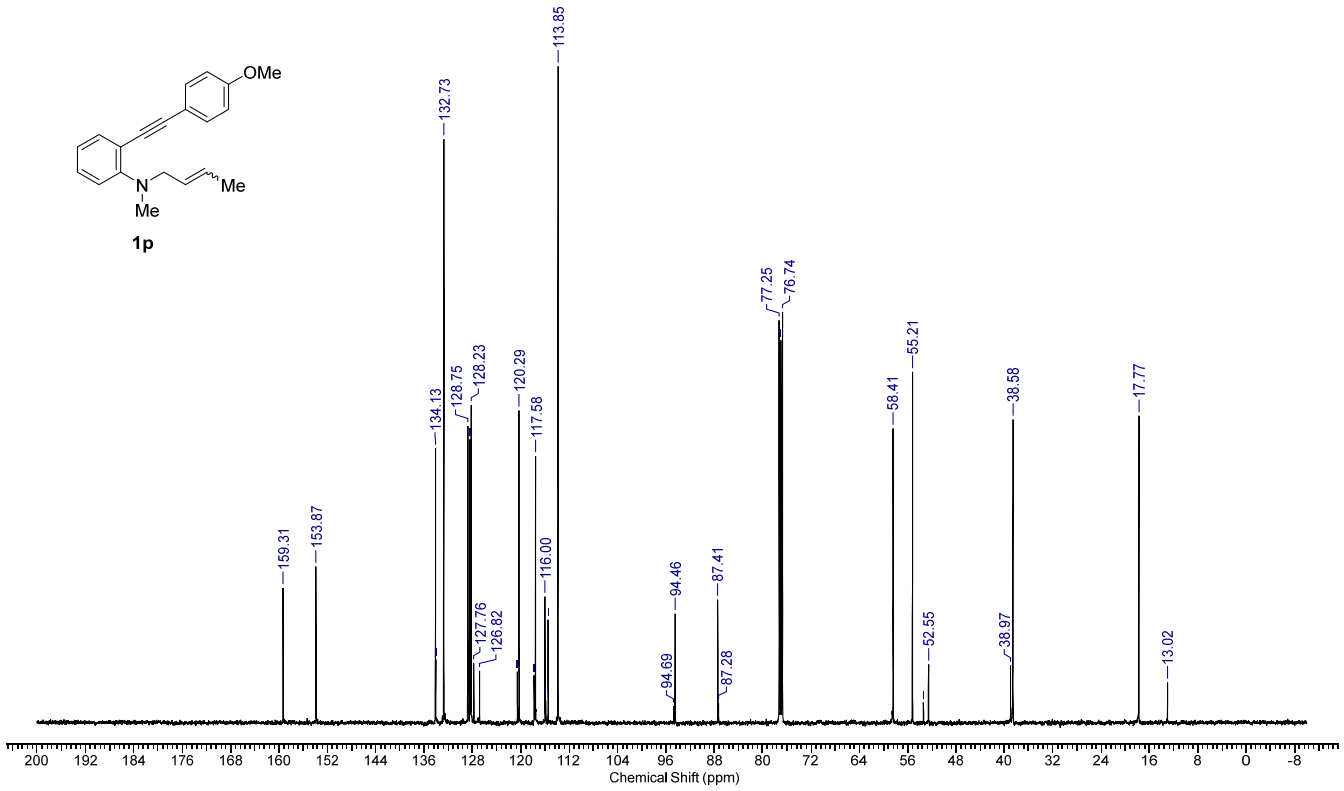


^{13}C NMR (125 MHz, $CDCl_3$) of **1p** (83:17 mixture of geometrical isomers)



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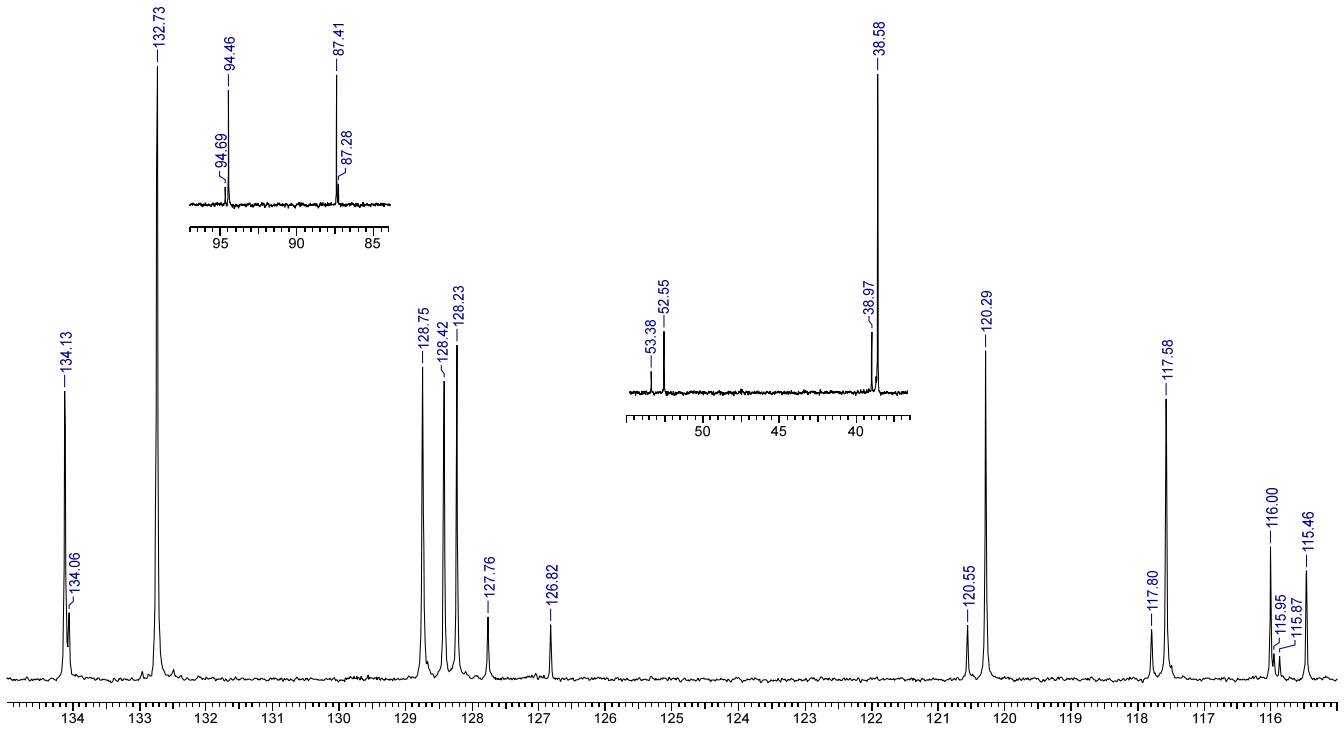
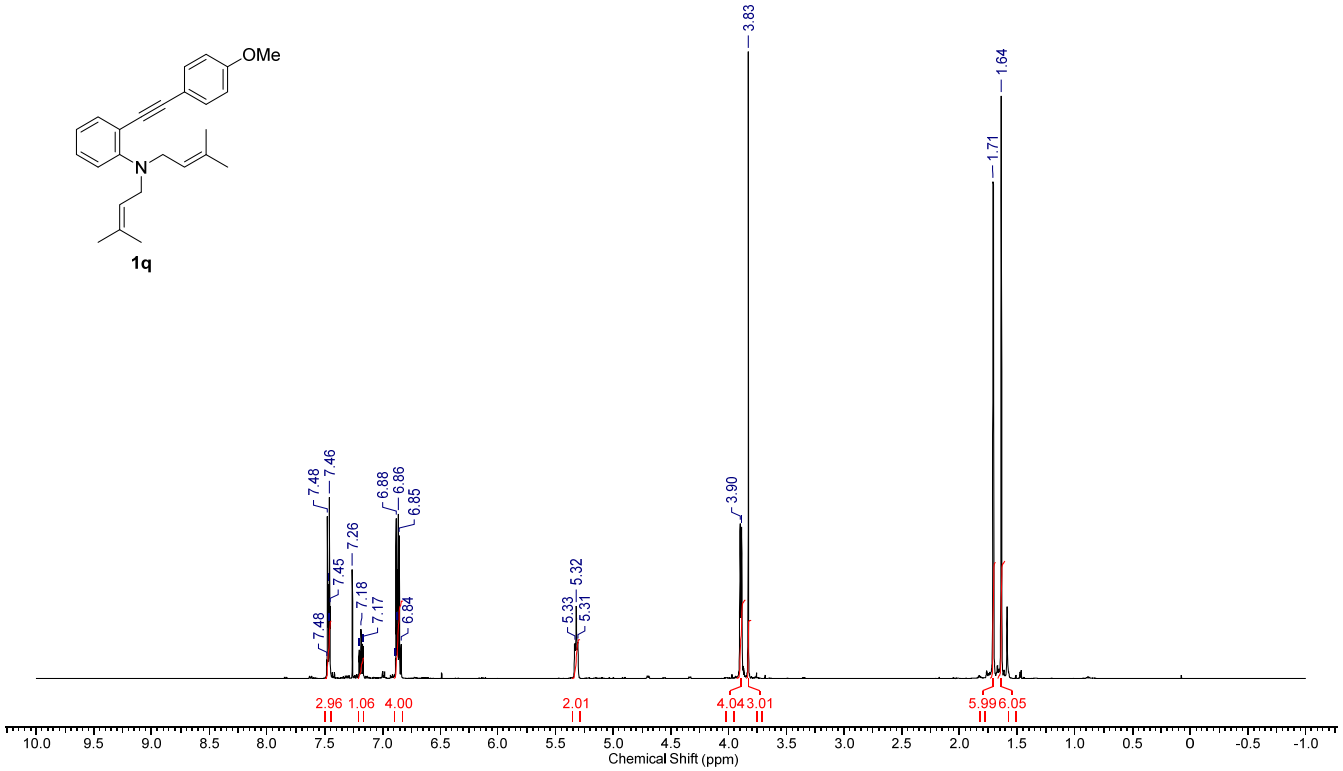
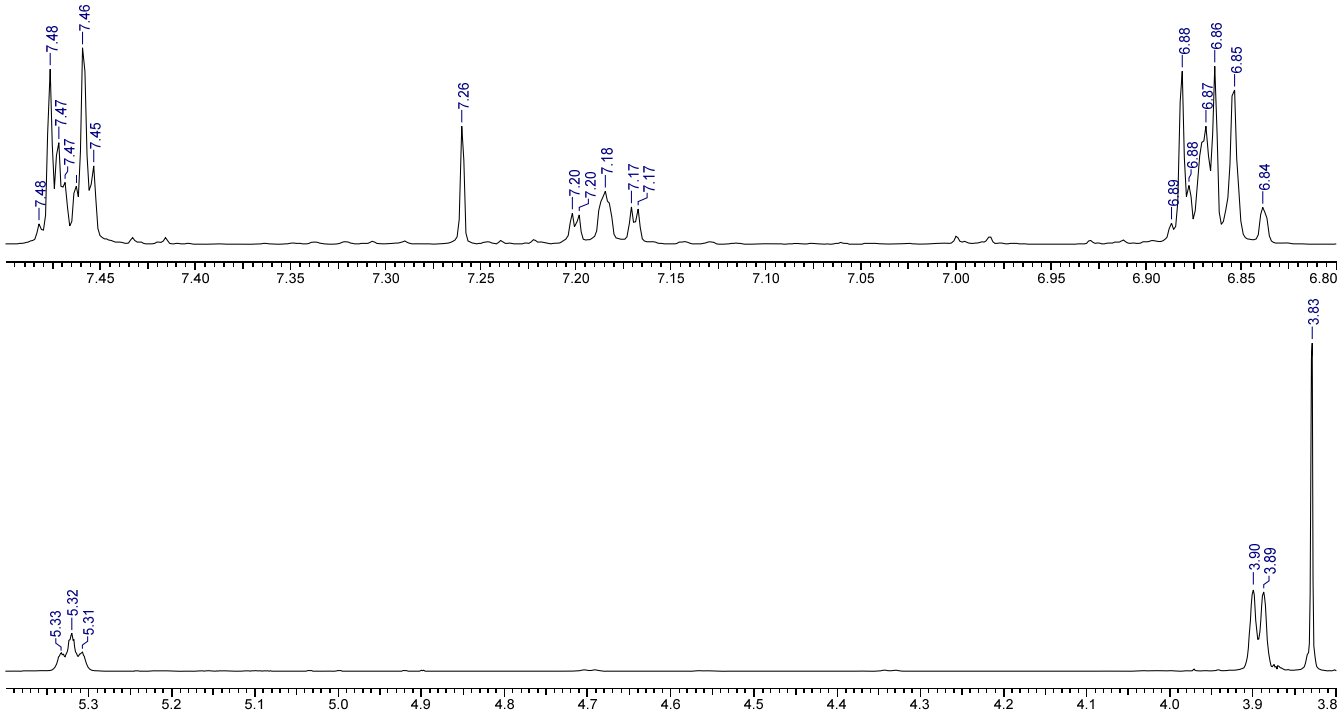


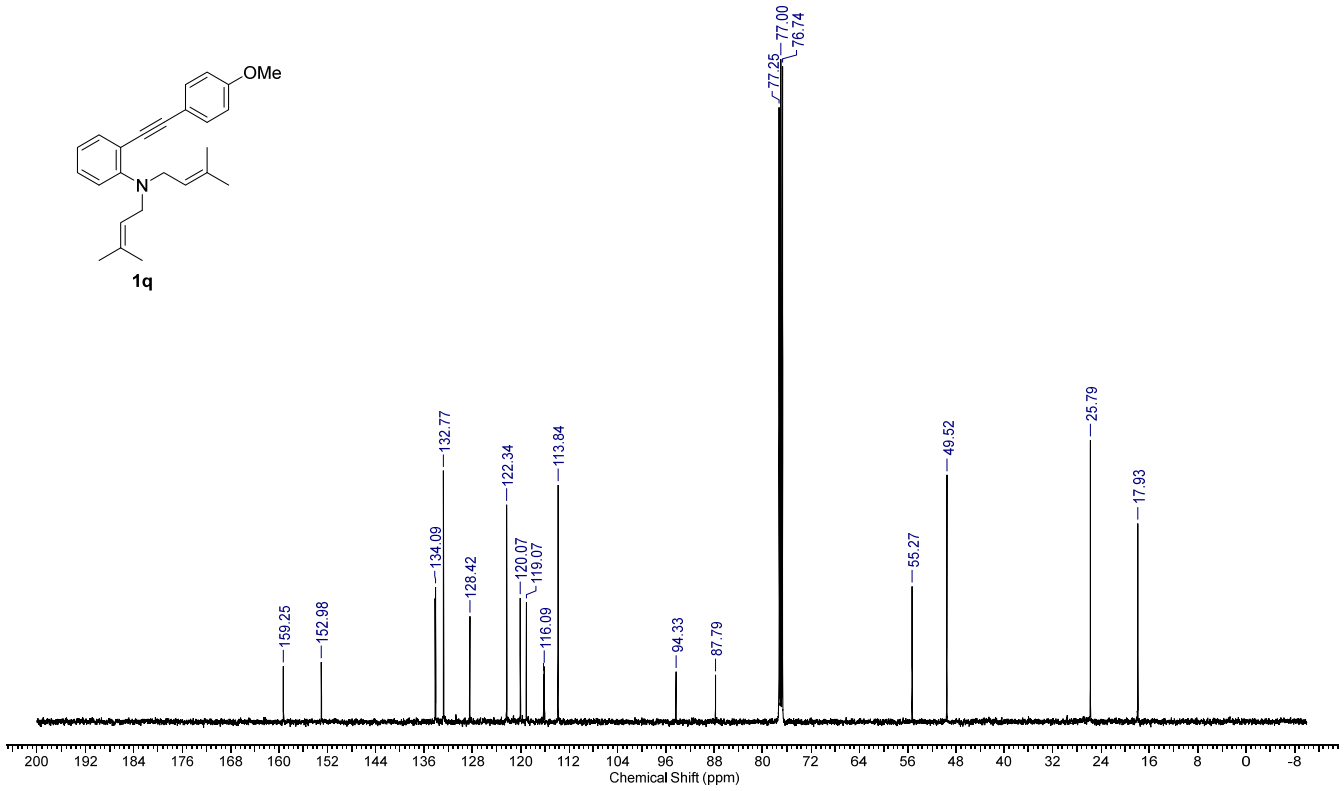
^{1}H NMR (500 MHz, $CDCl_3$) of **1q**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **1q**



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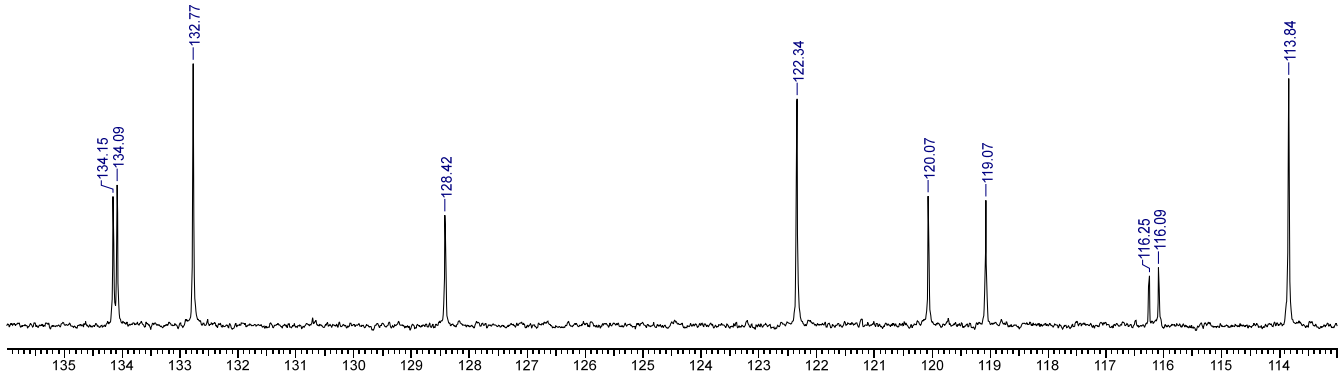
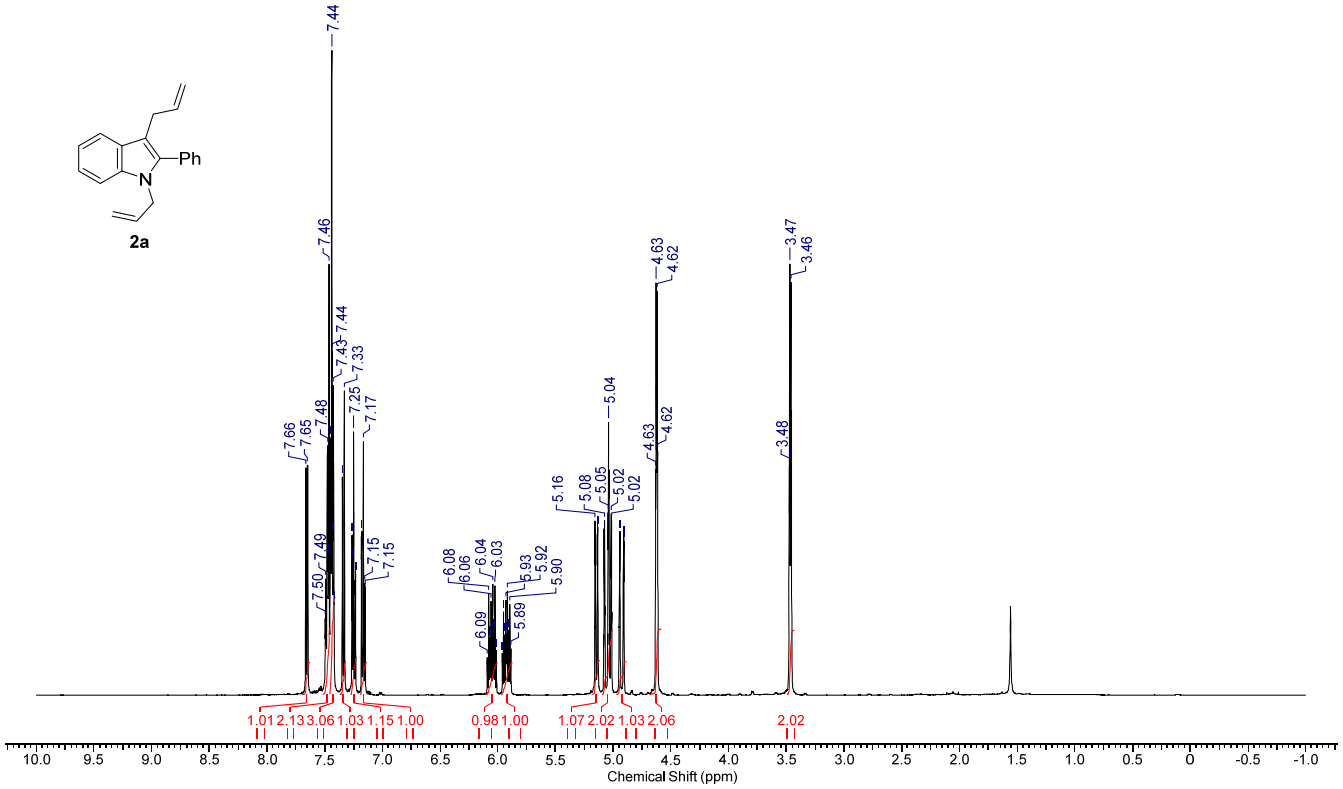


^{1}H NMR (500 MHz, $CDCl_3$) of **2a**



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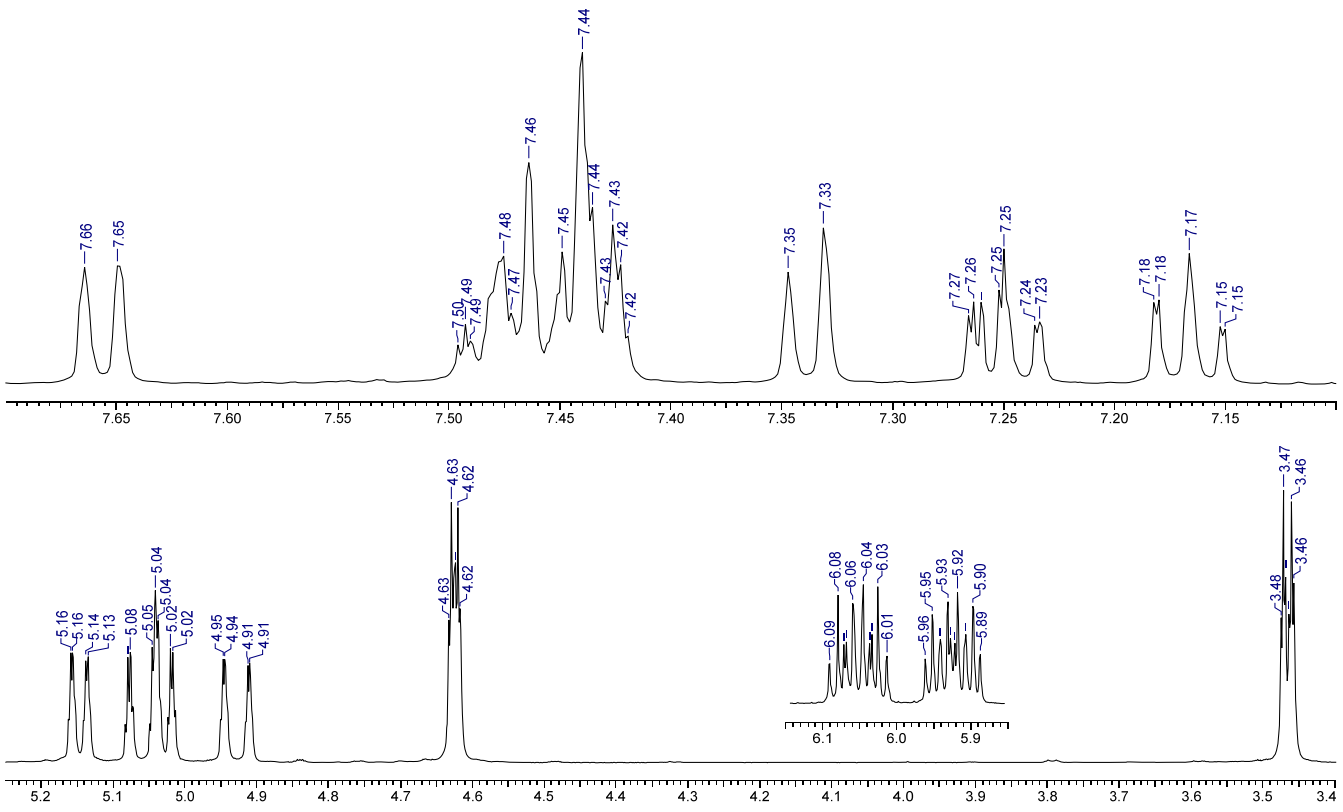
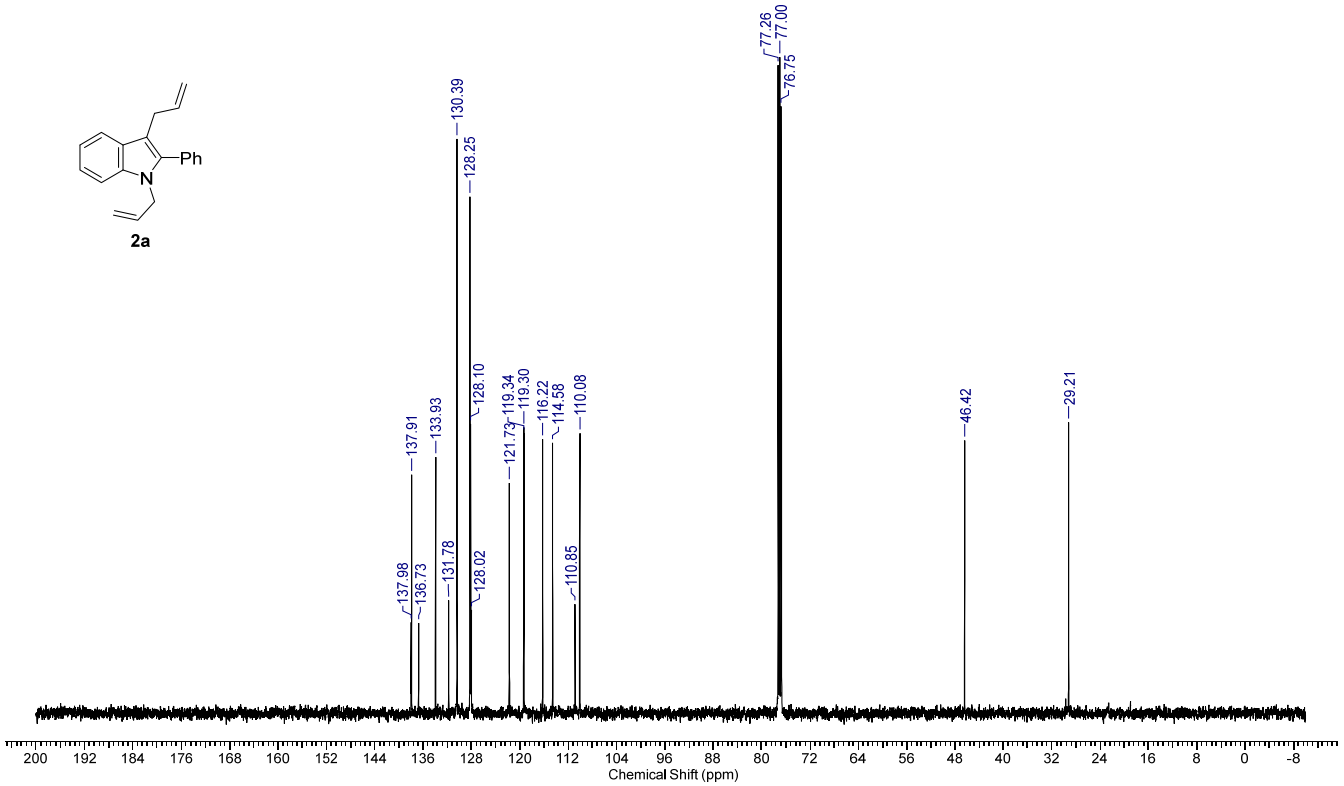


^{13}C NMR (125 MHz, $CDCl_3$) of **2a**



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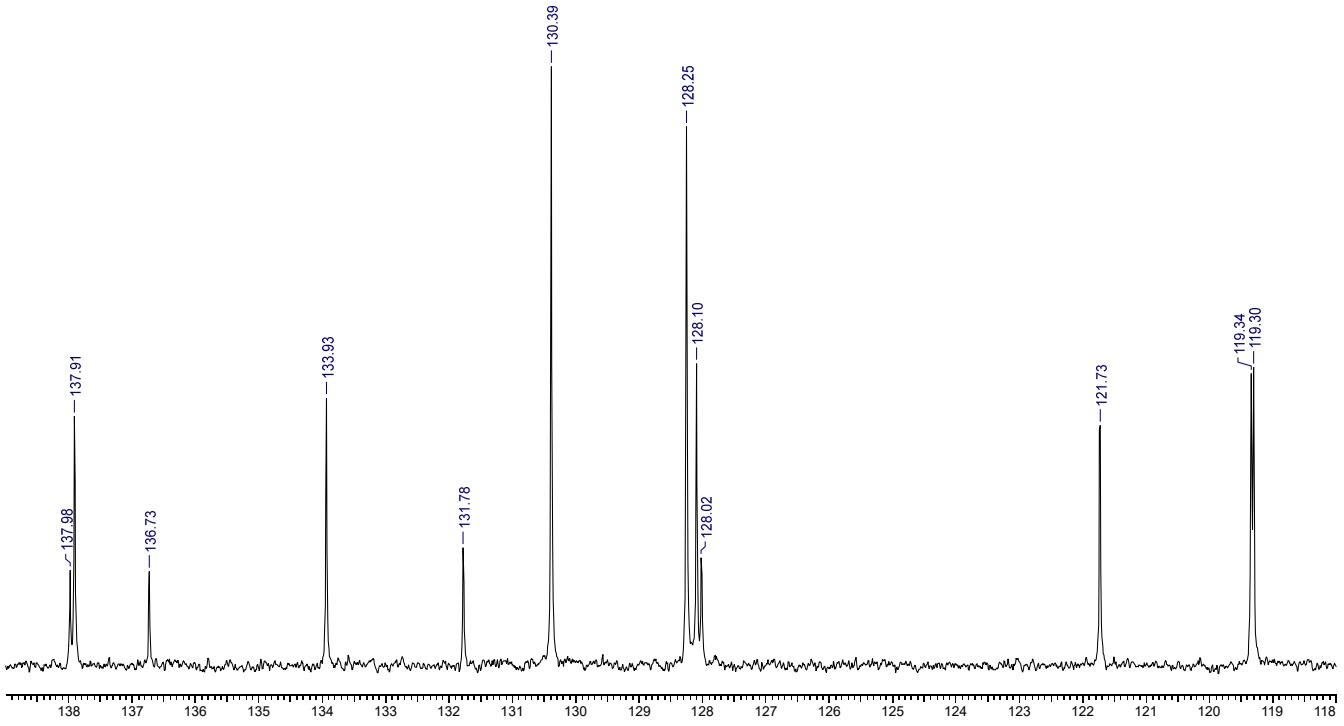
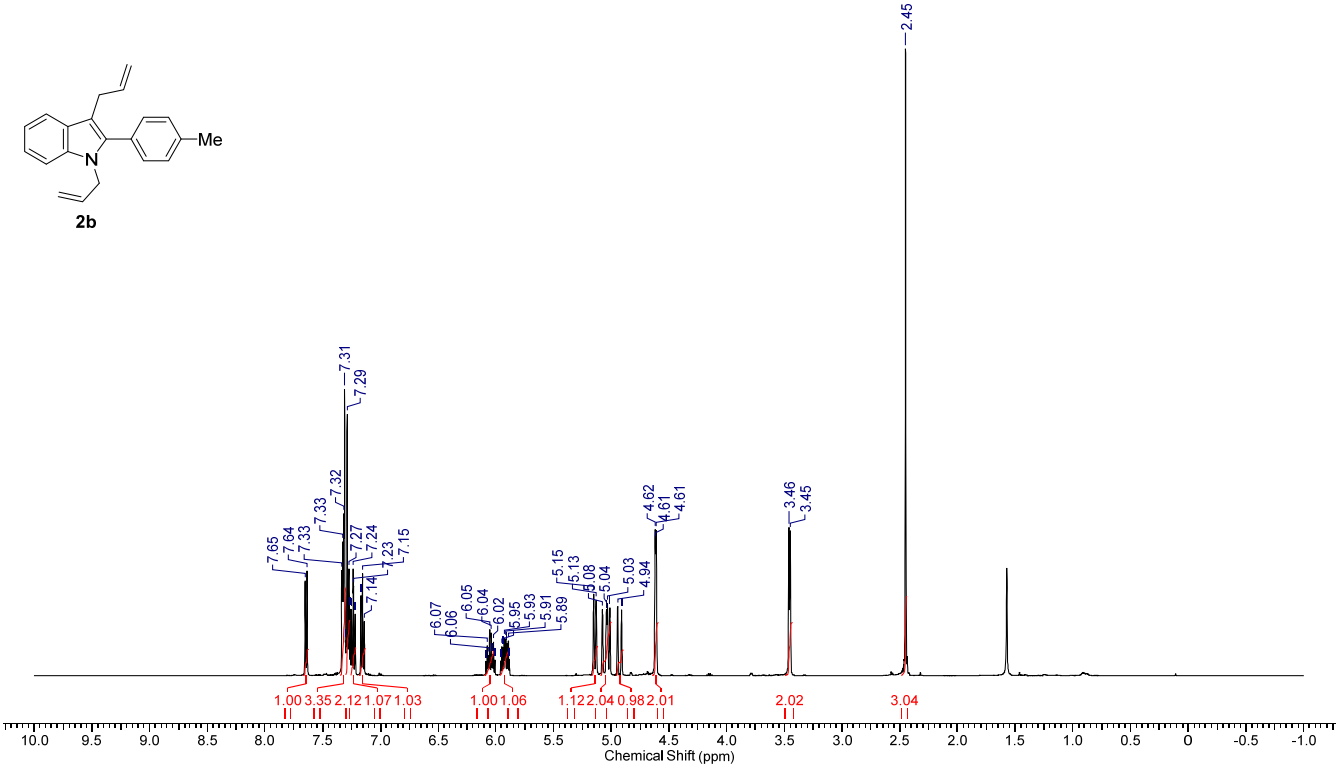


^{1}H NMR (500 MHz, $CDCl_3$) of **2b**



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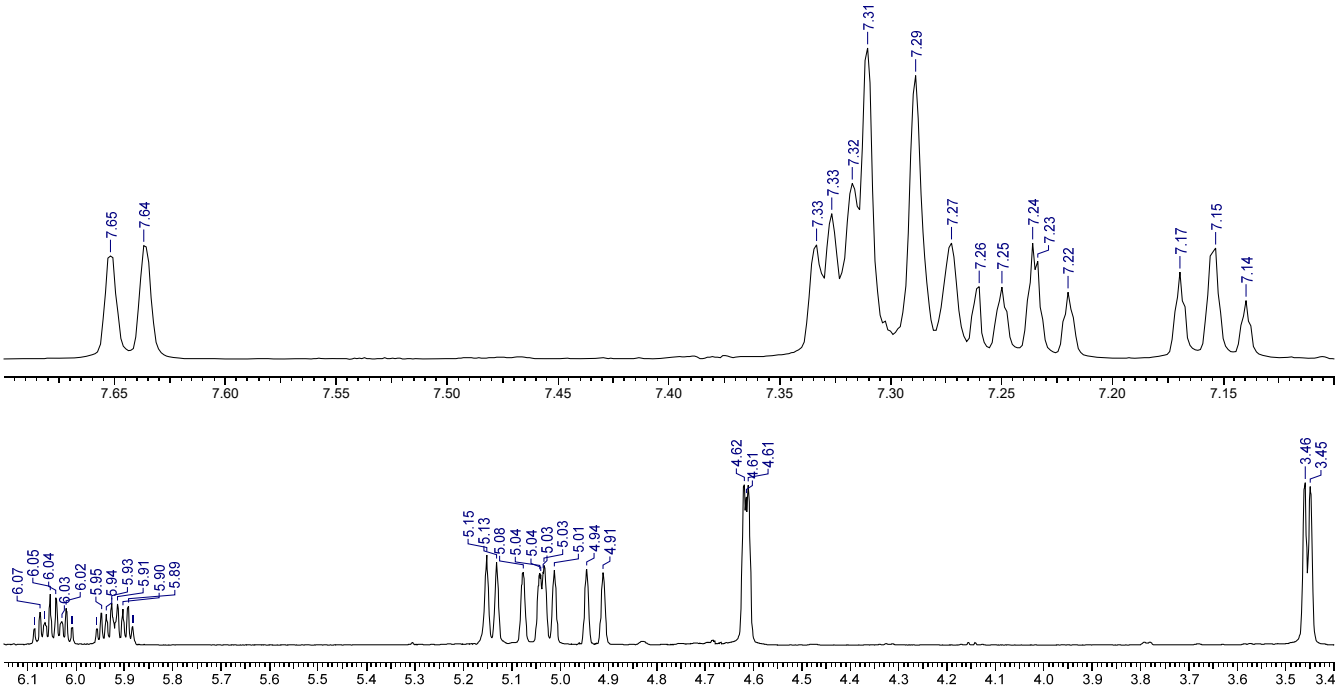
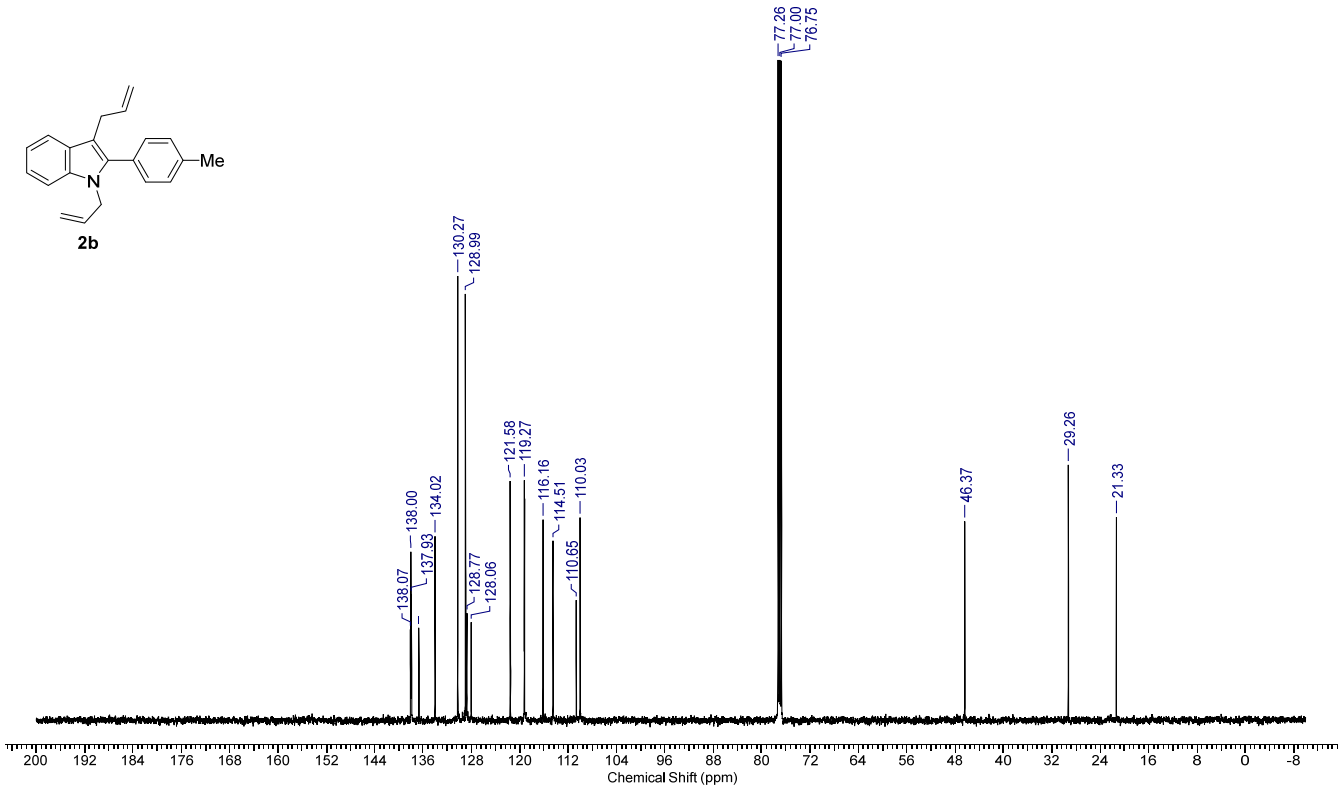


^{13}C NMR (125 MHz, $CDCl_3$) of **2b**



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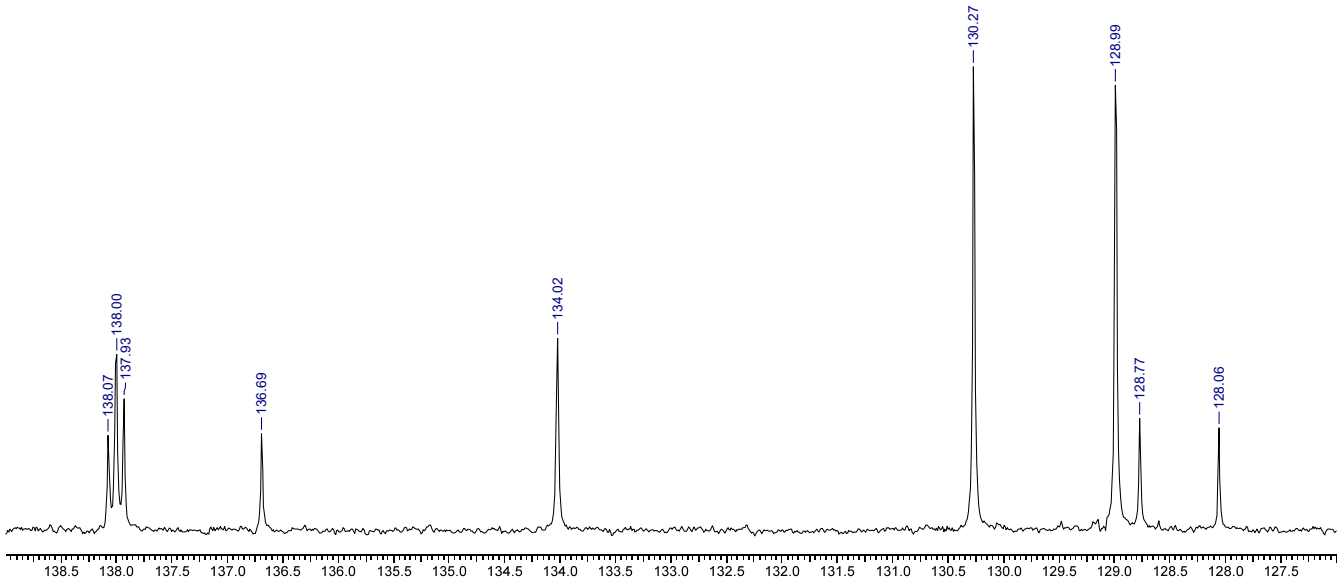
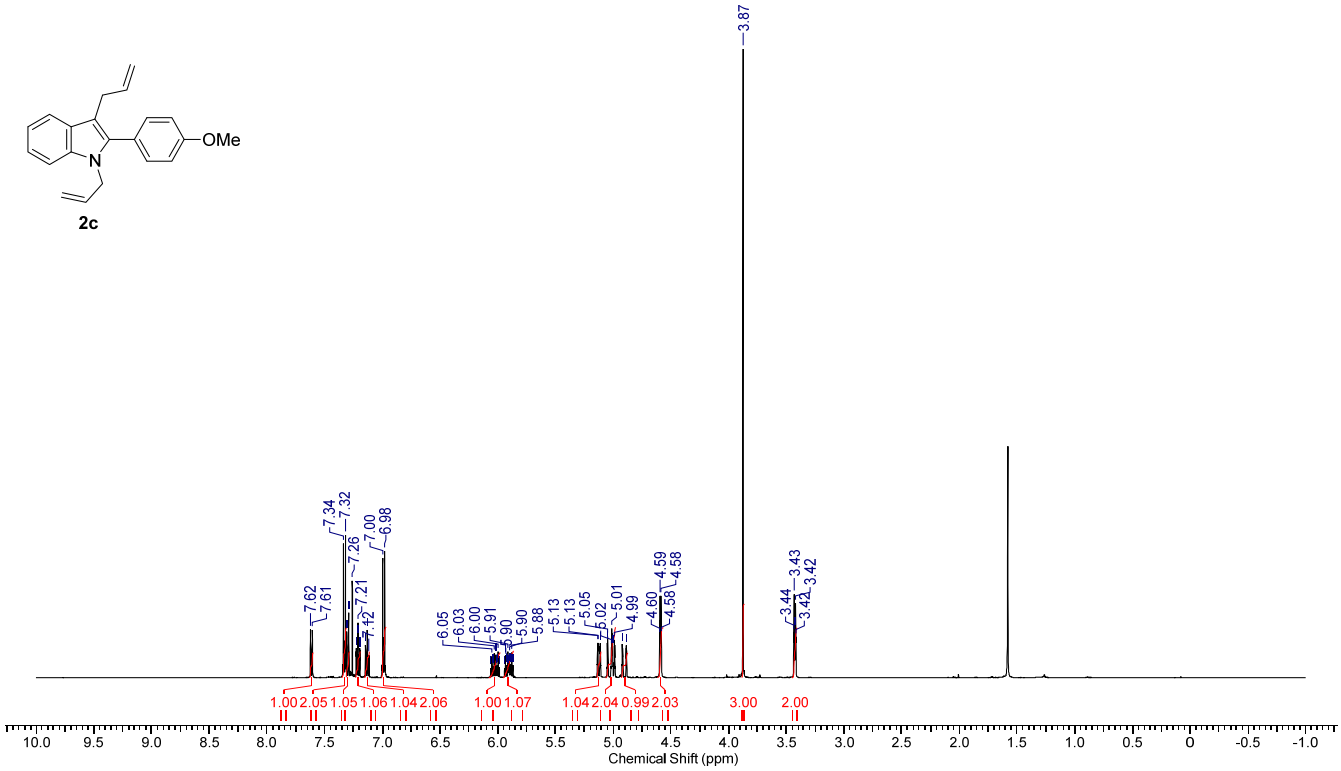


^{1}H NMR (500 MHz, $CDCl_3$) of **2c**



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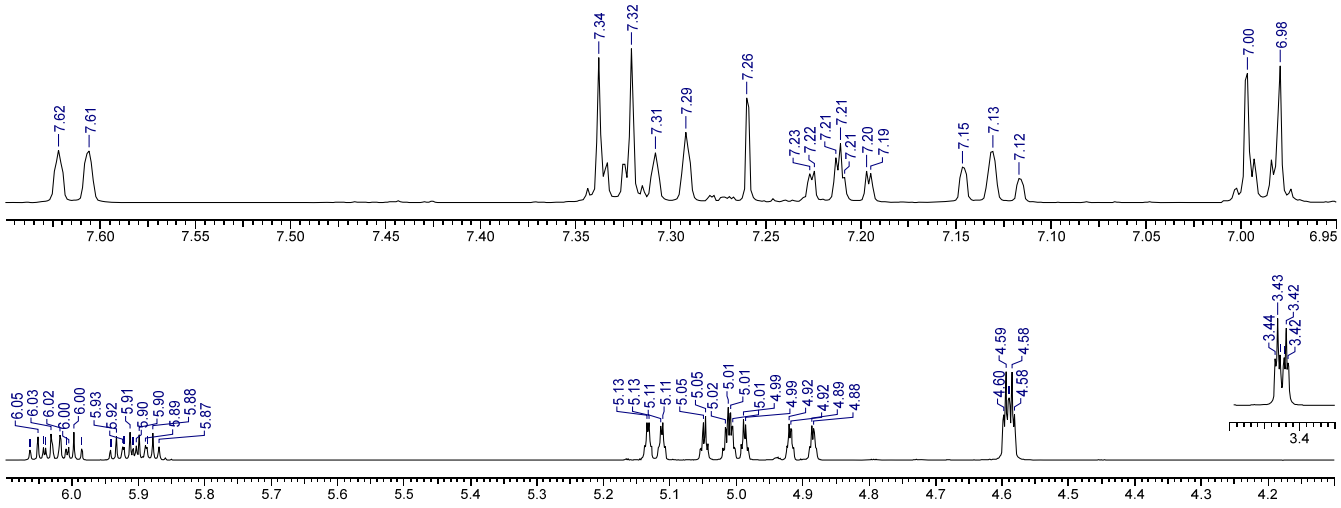
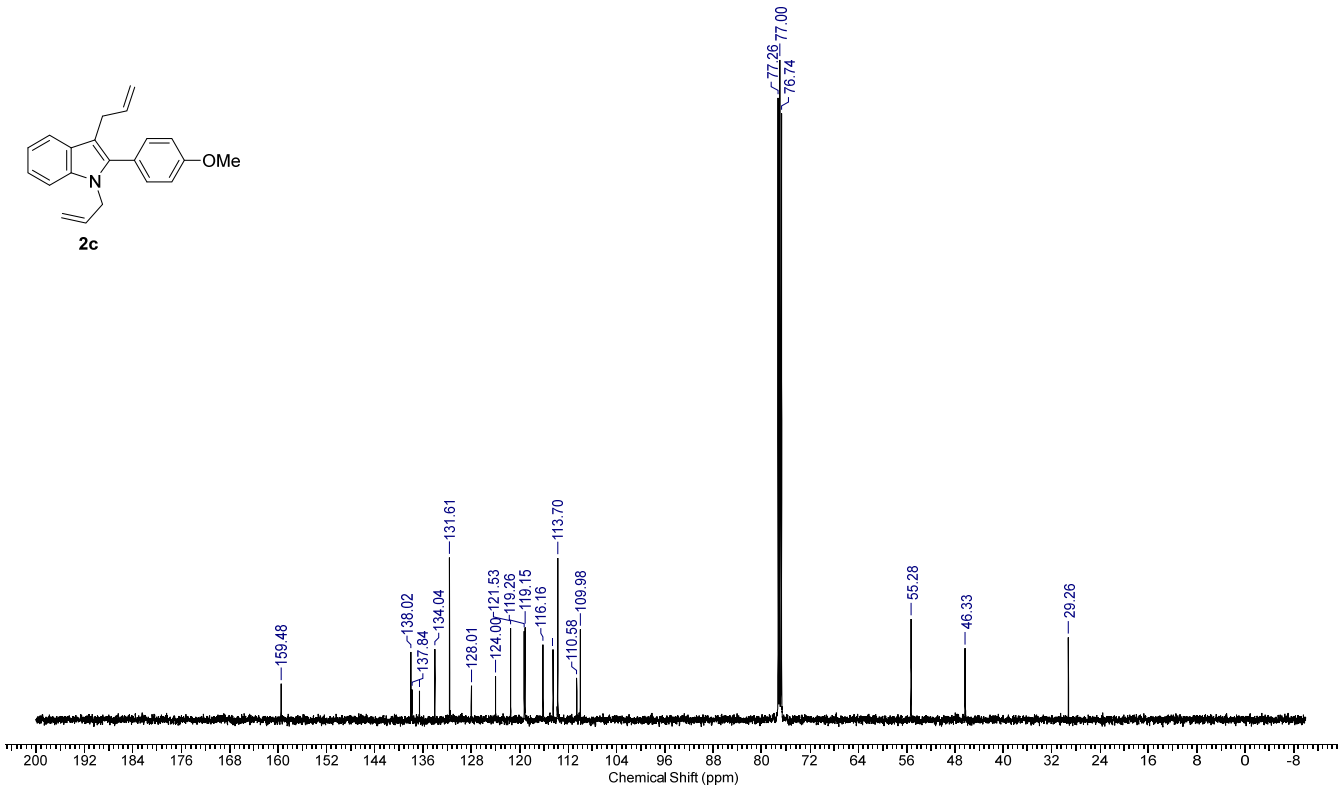


^{13}C NMR (125 MHz, $CDCl_3$) of **2c**



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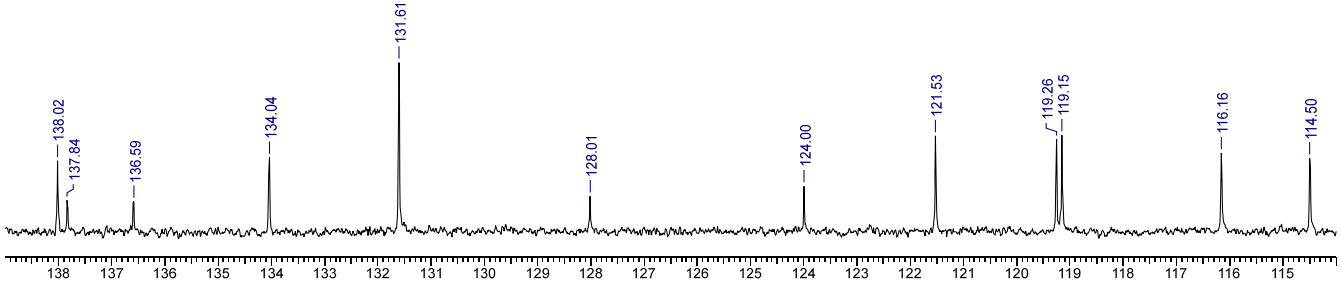
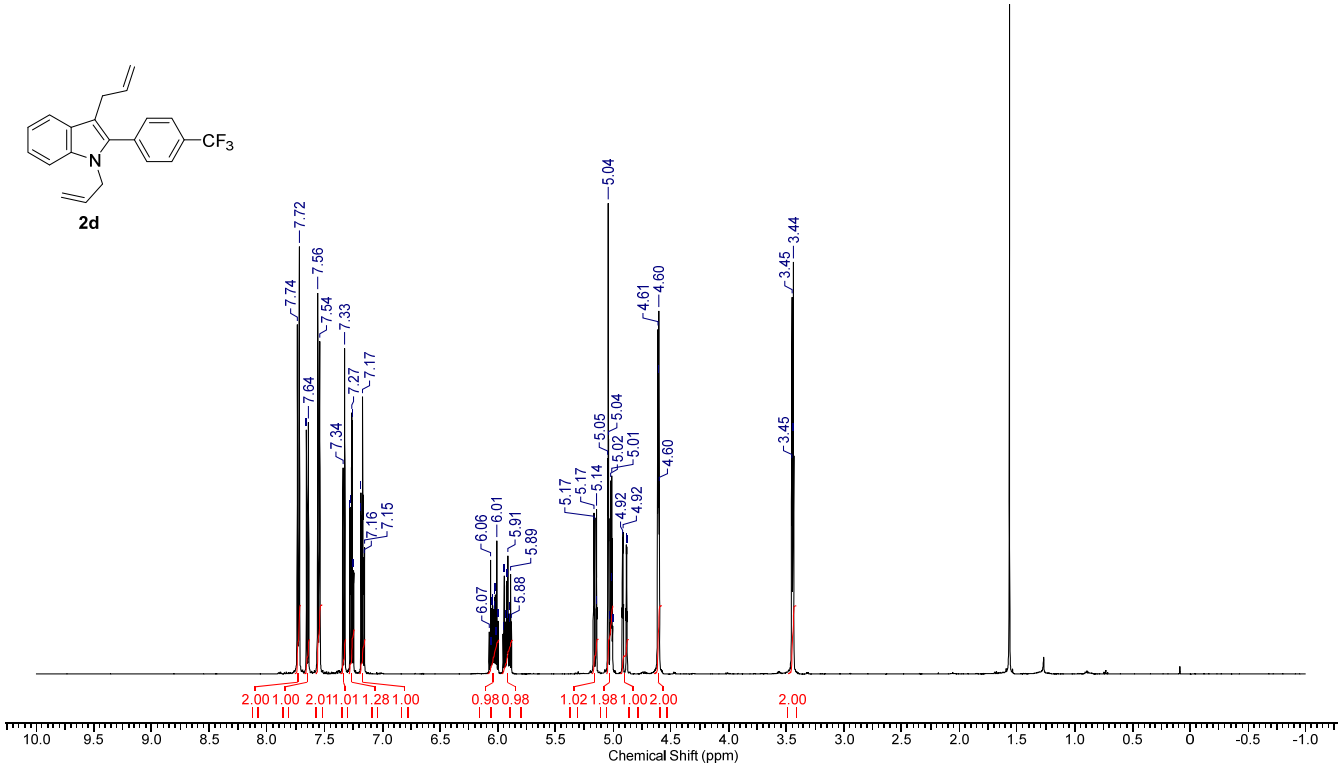
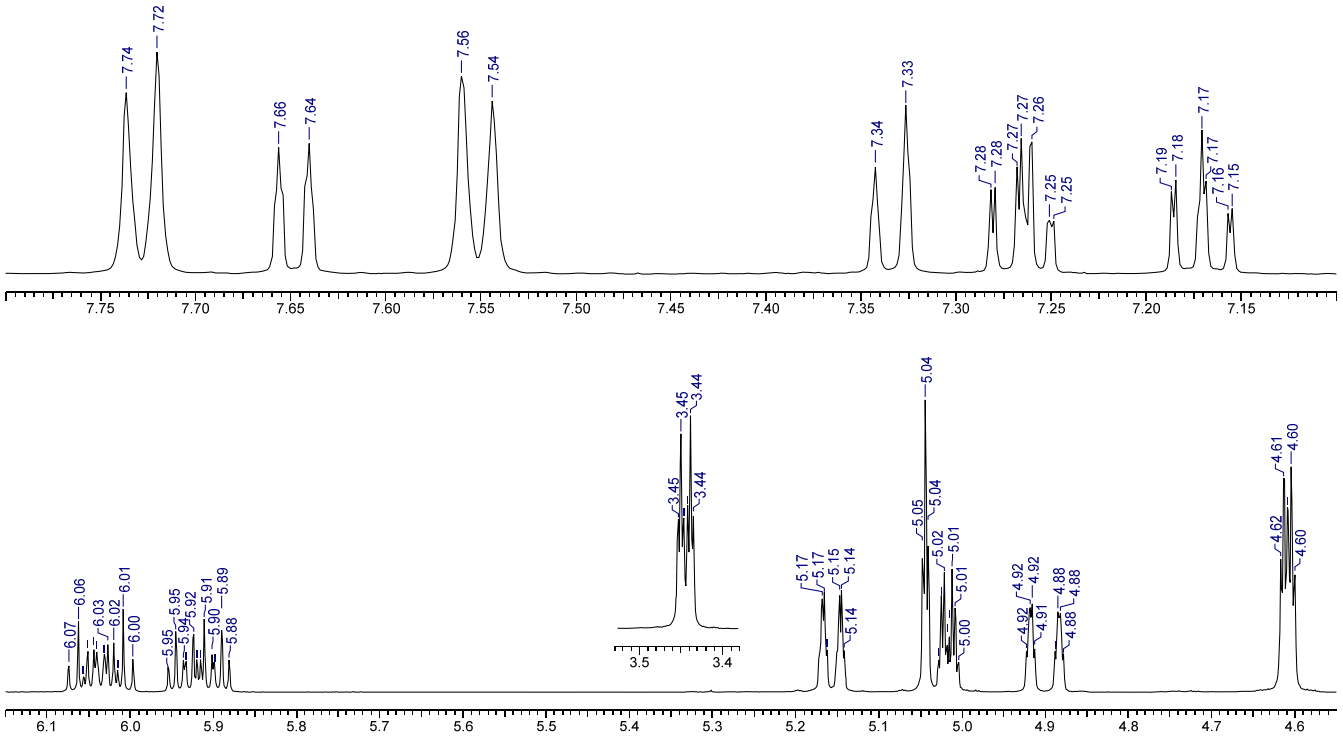


^{1}H NMR (500 MHz, $CDCl_3$) of **2d**



Enlarged view



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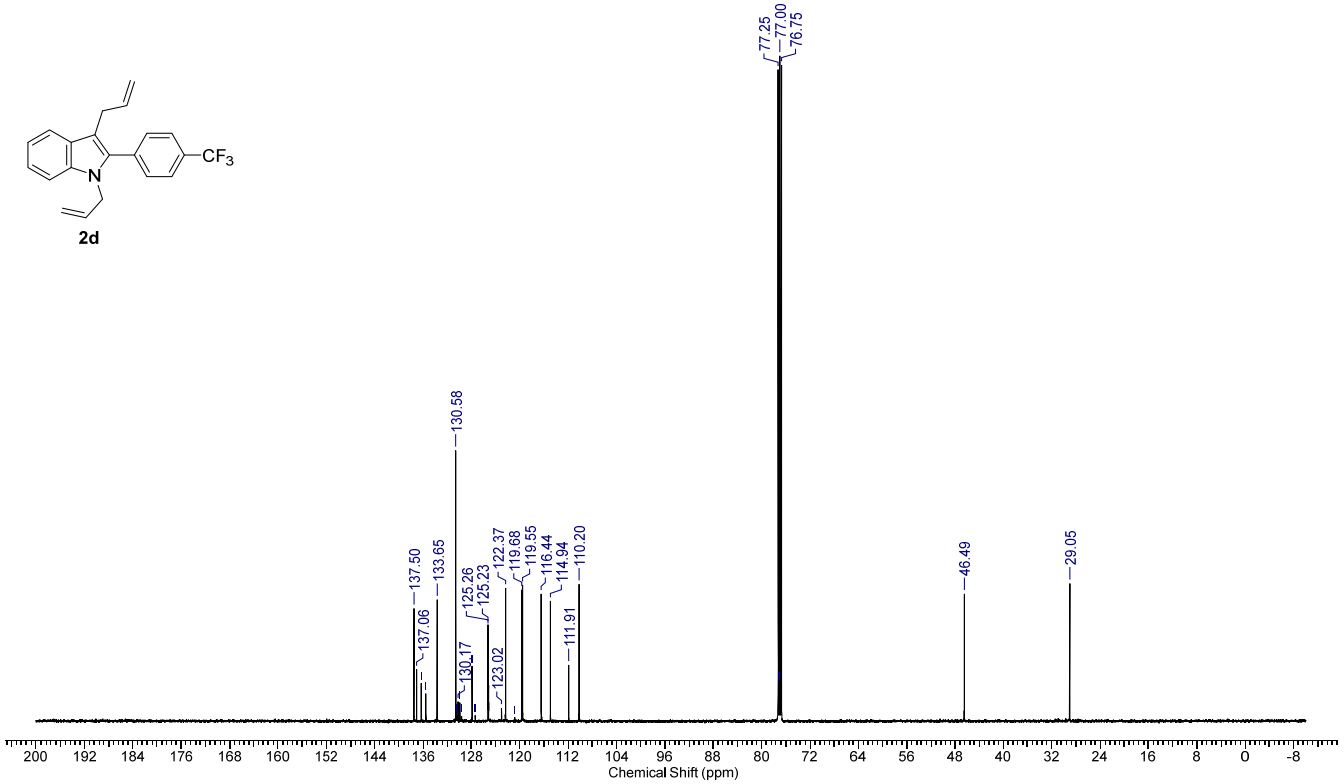

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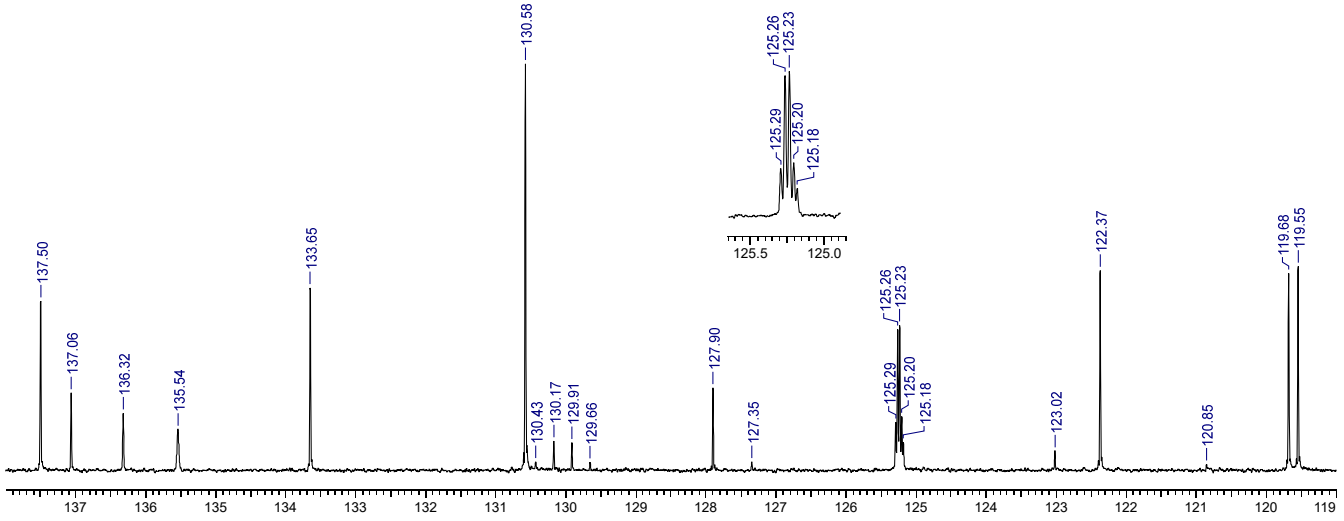

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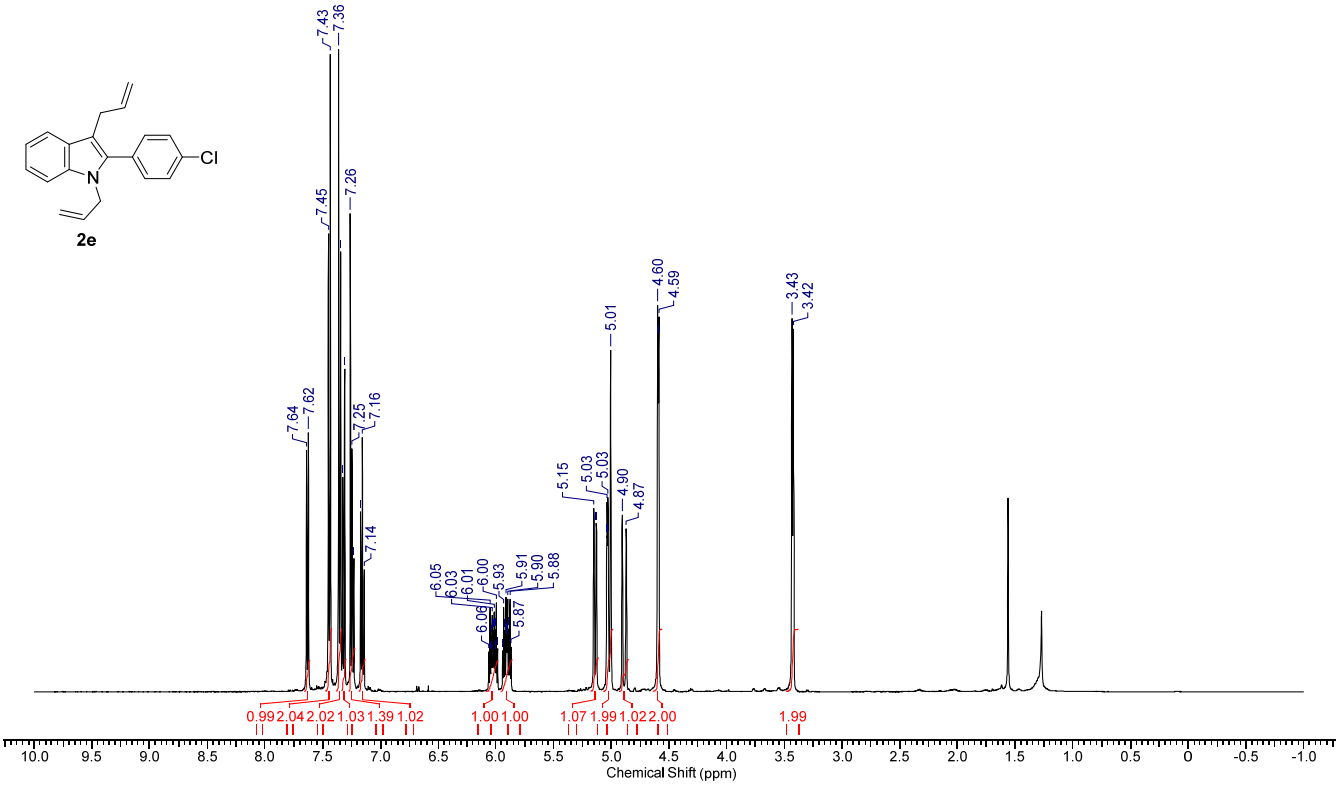

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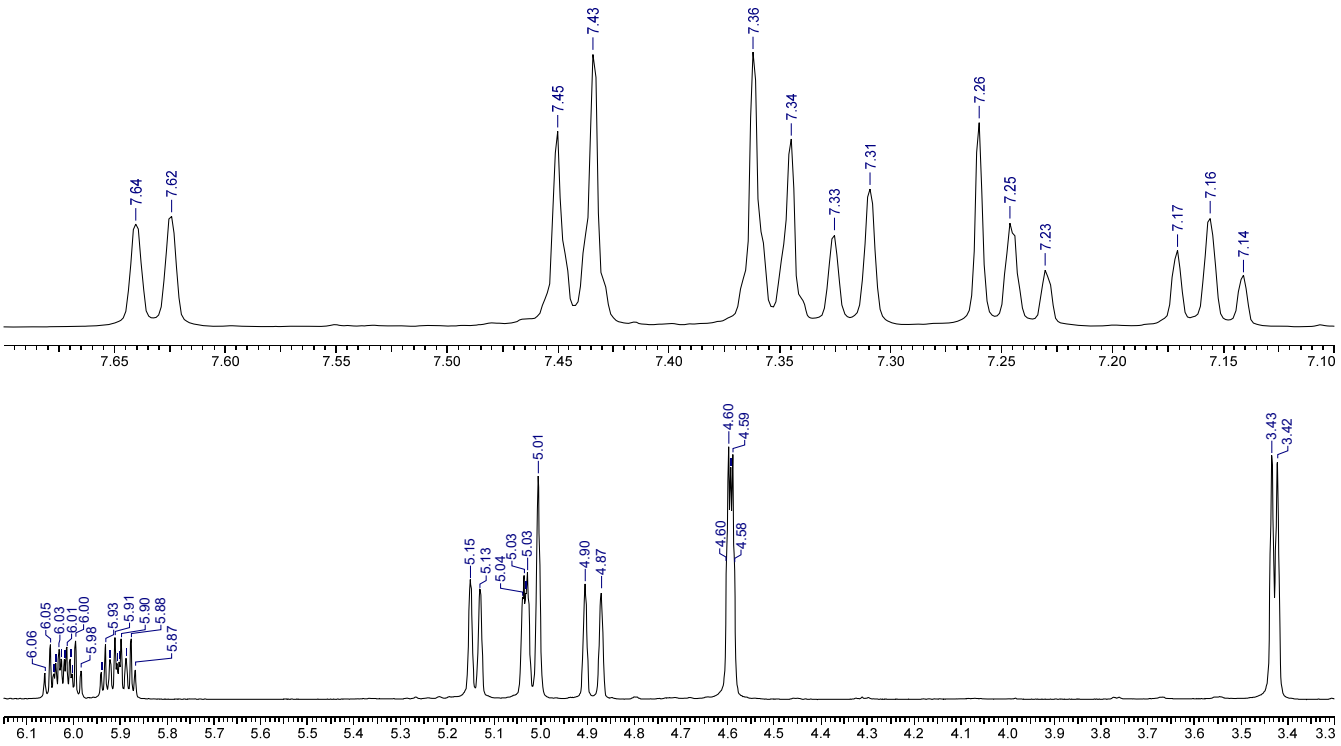
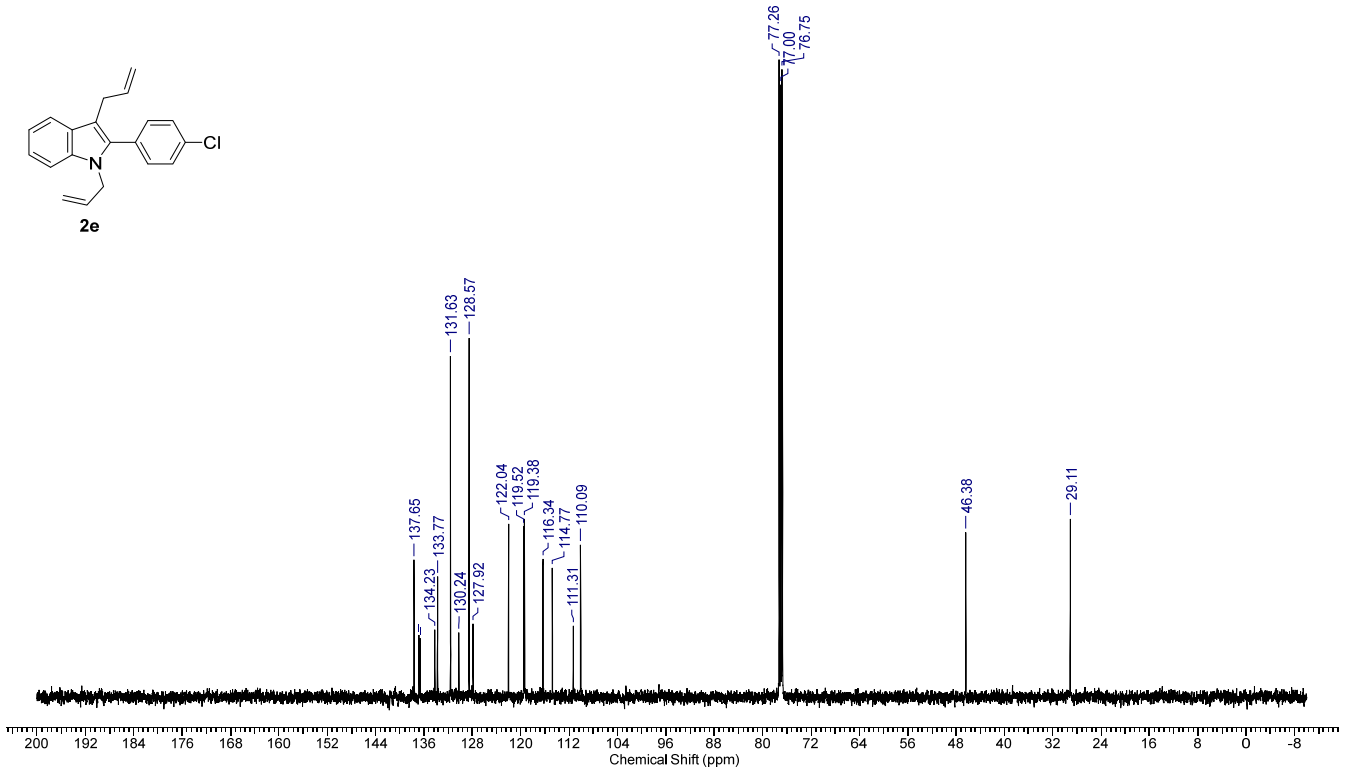


^{13}C NMR (125 MHz, $CDCl_3$) of **2e**



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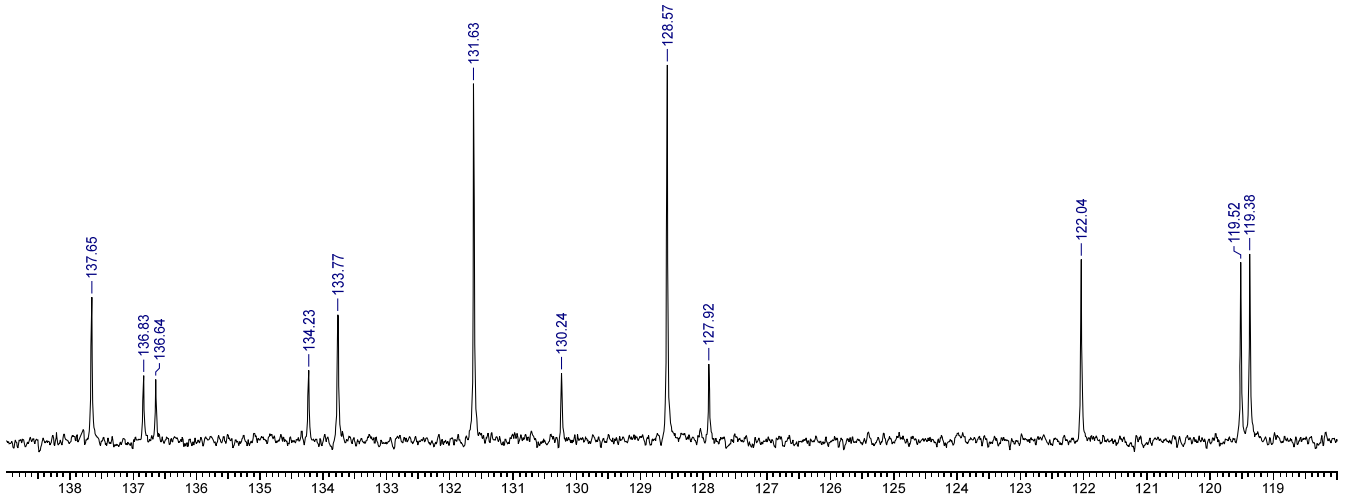
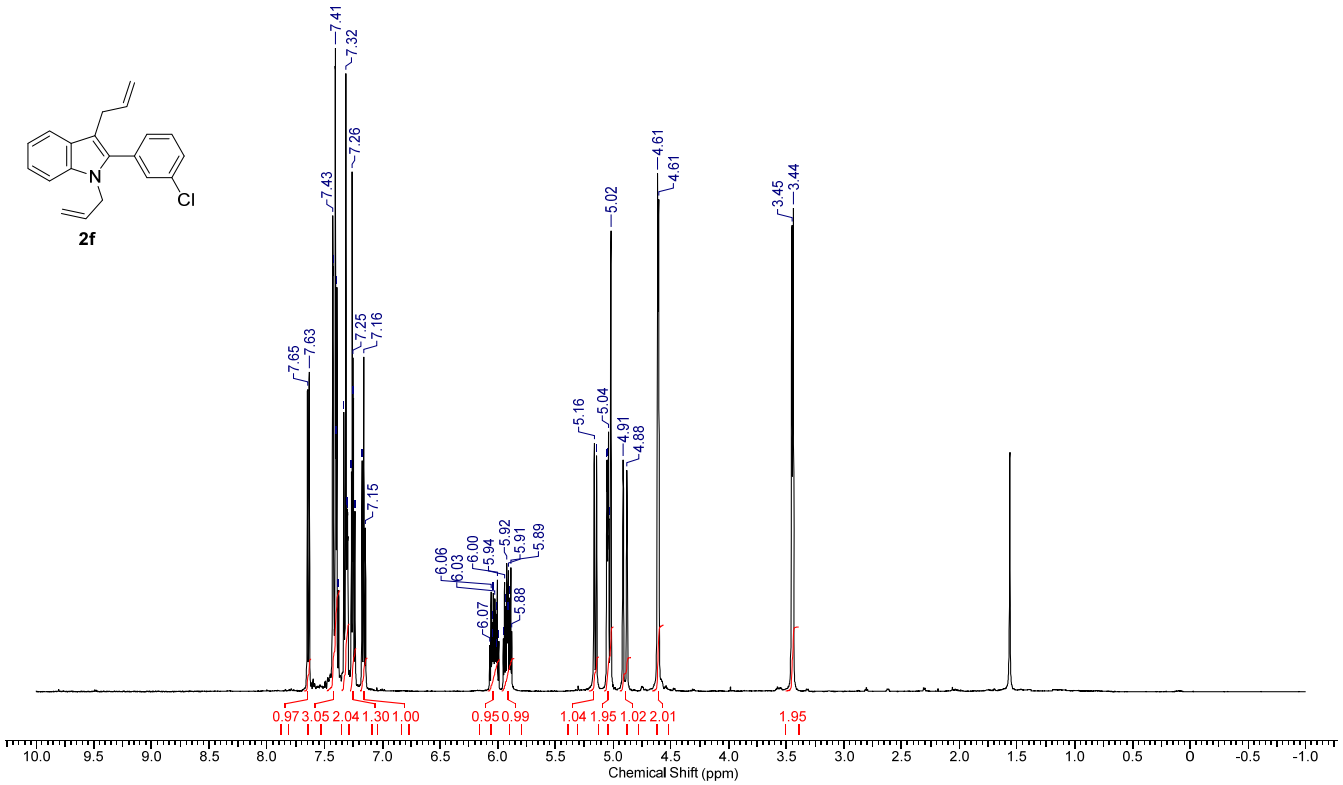
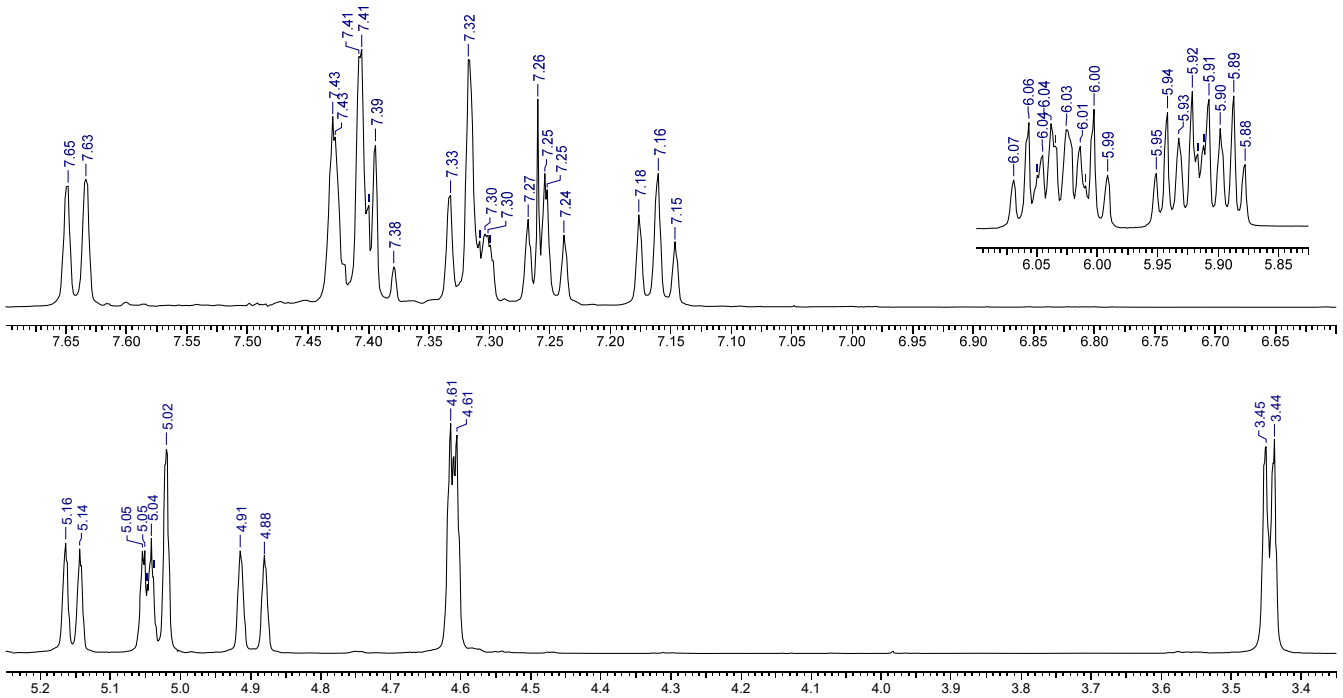


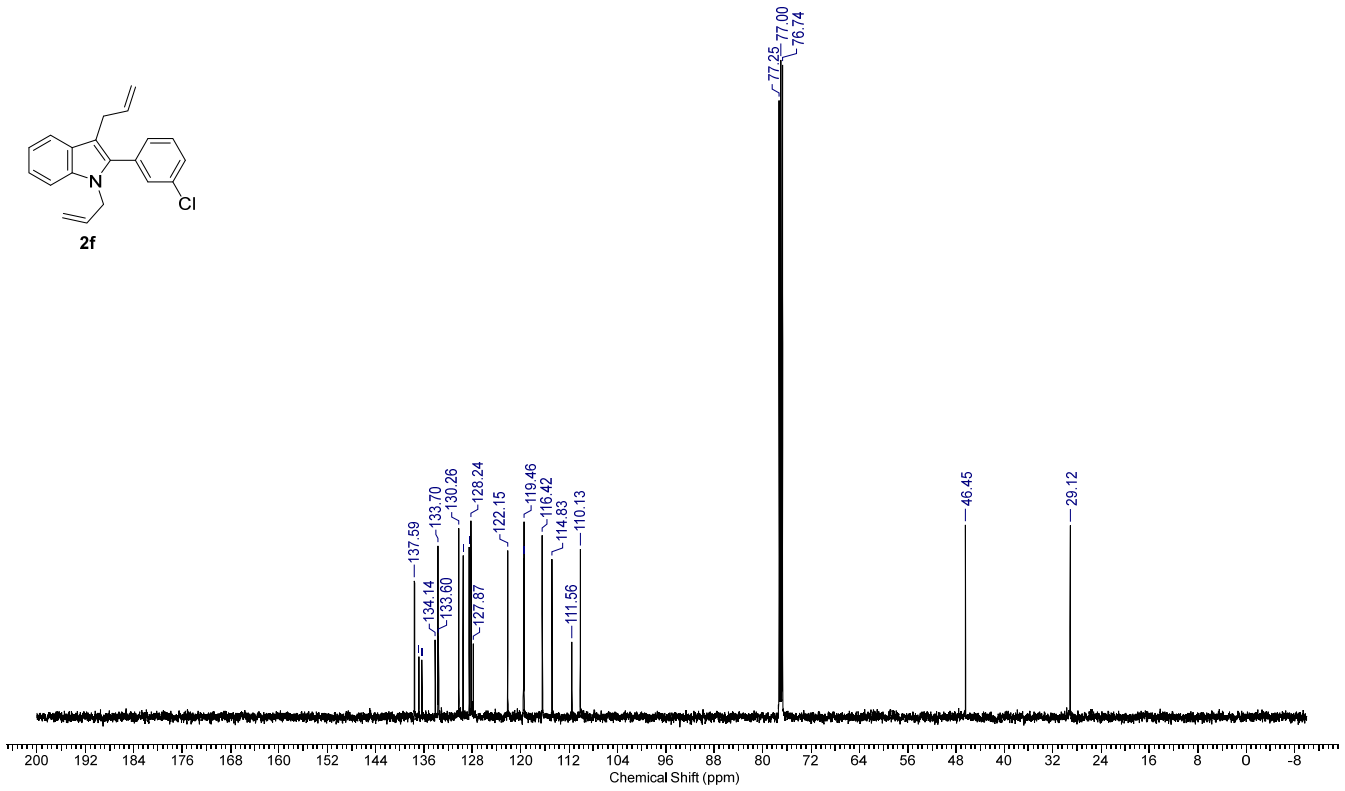
^{1}H NMR (500 MHz, $CDCl_3$) of **2f**



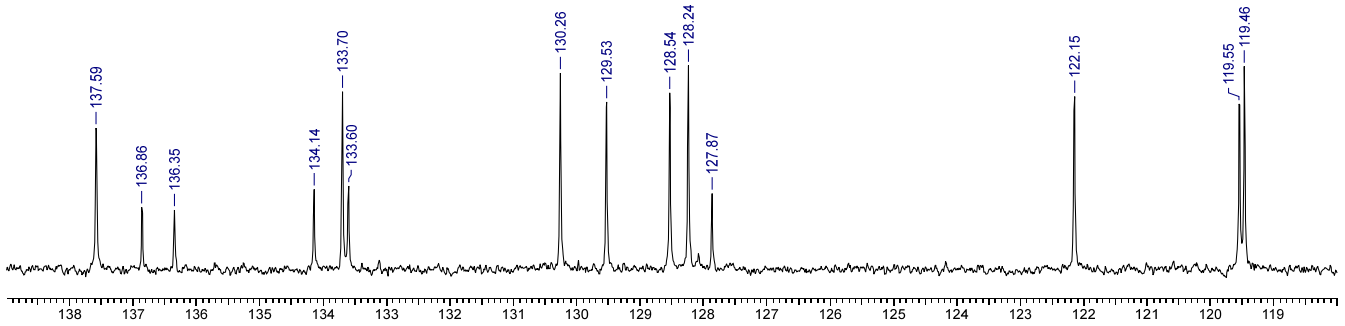
Enlarged view



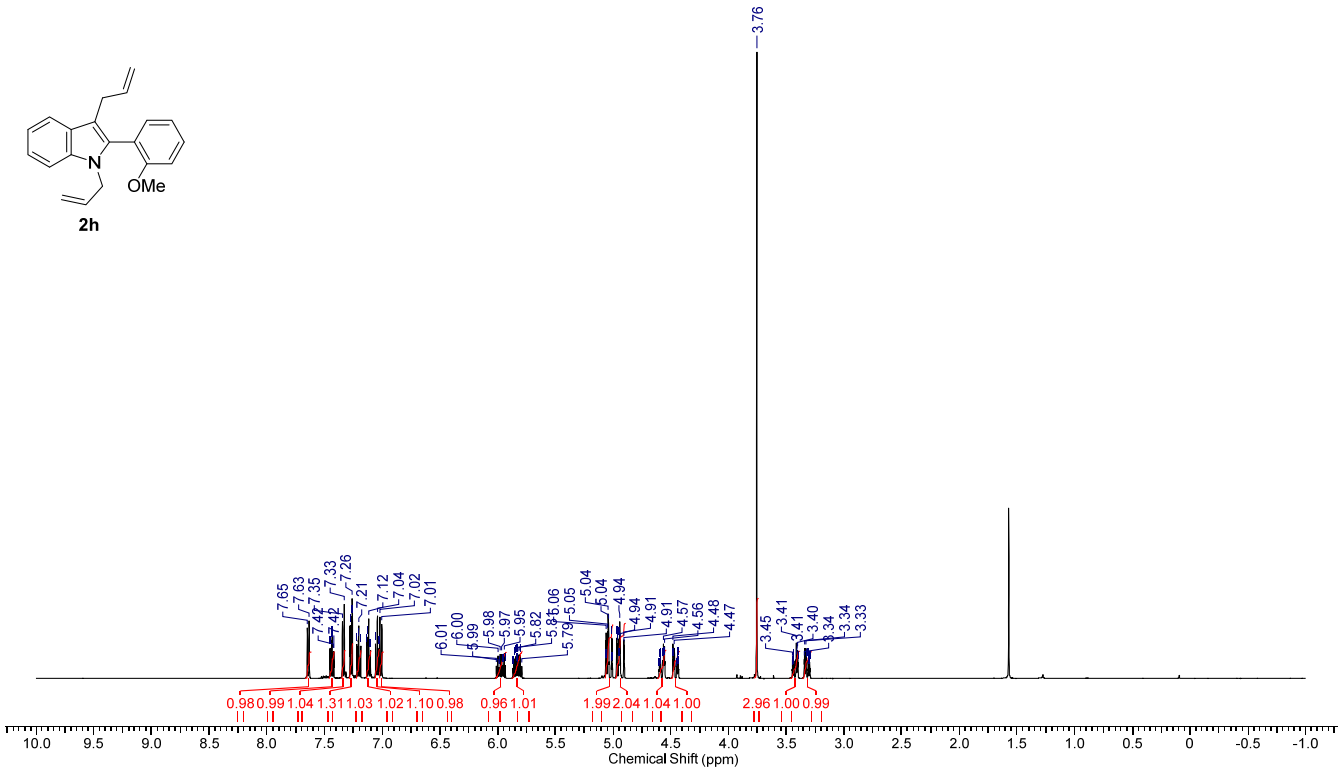
^{13}C NMR (125 MHz, $CDCl_3$) of **2f**



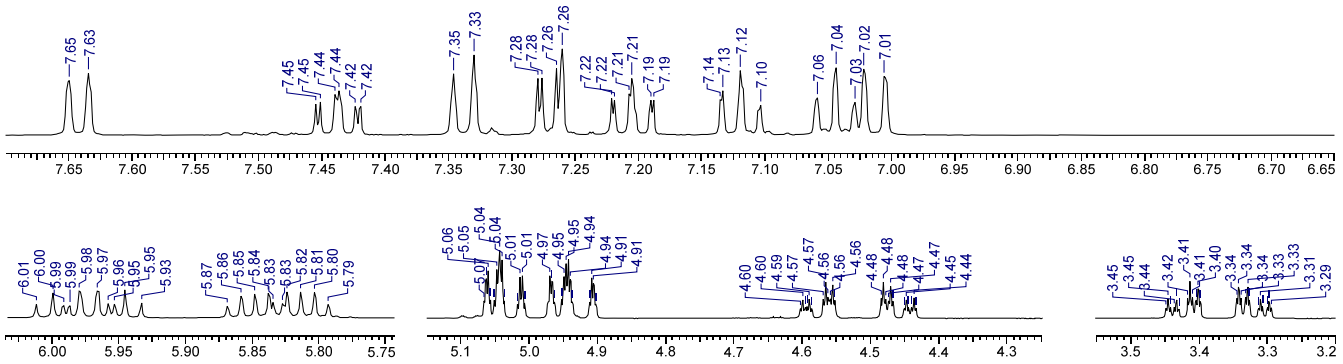
Enlarged view



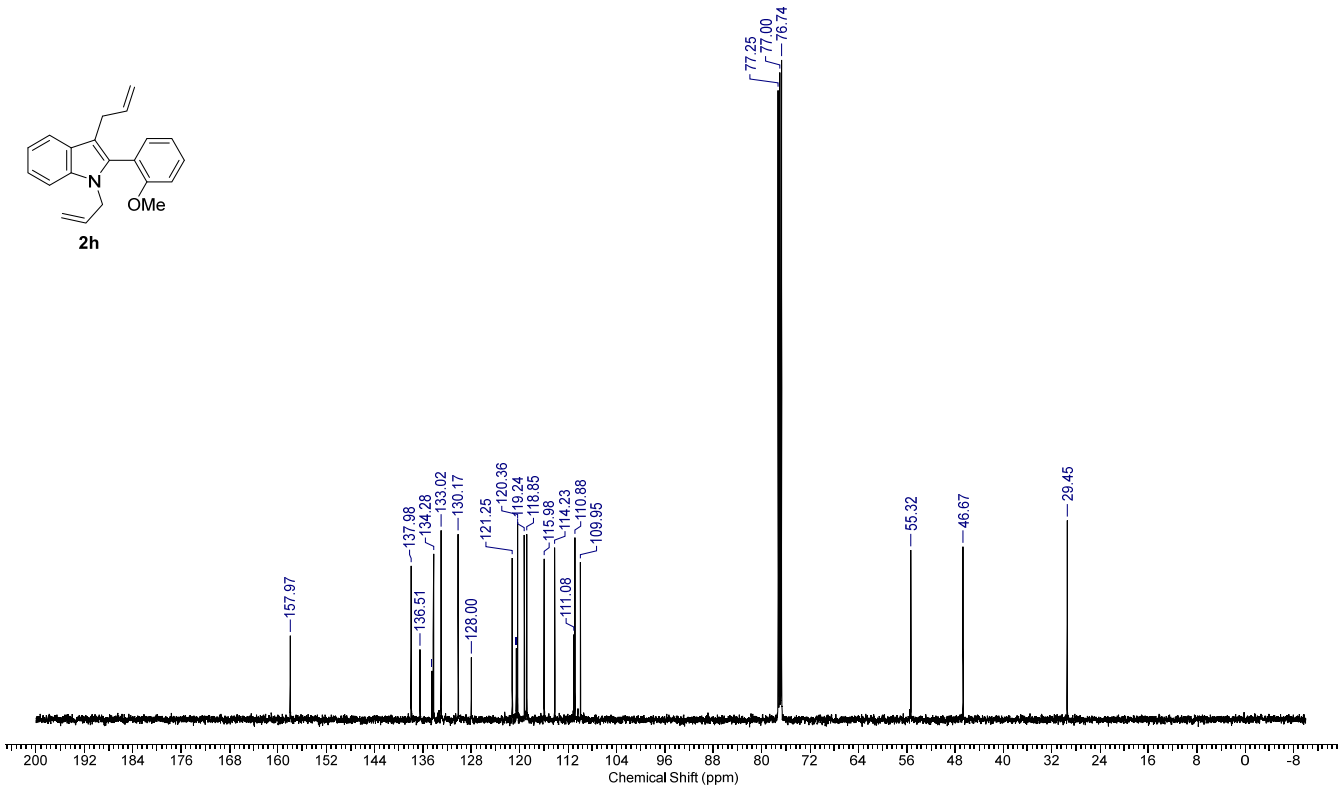
^{1}H NMR (500 MHz, $CDCl_3$) of **2h**



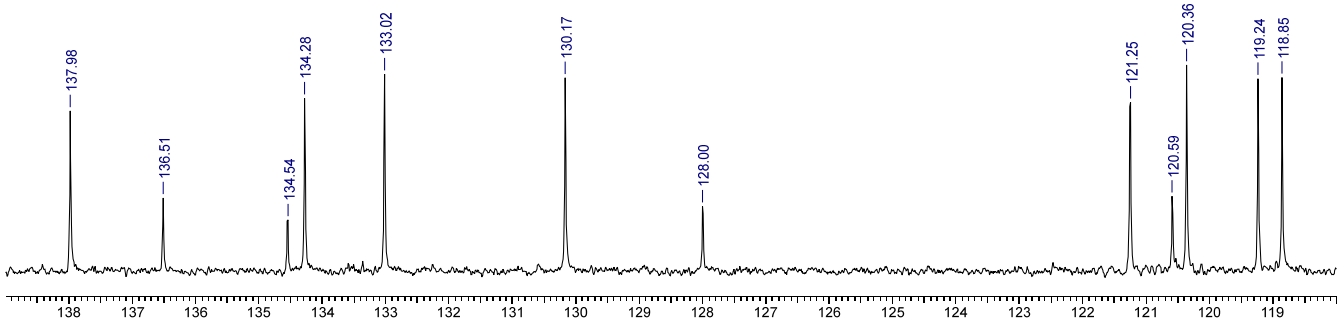
Enlarged view



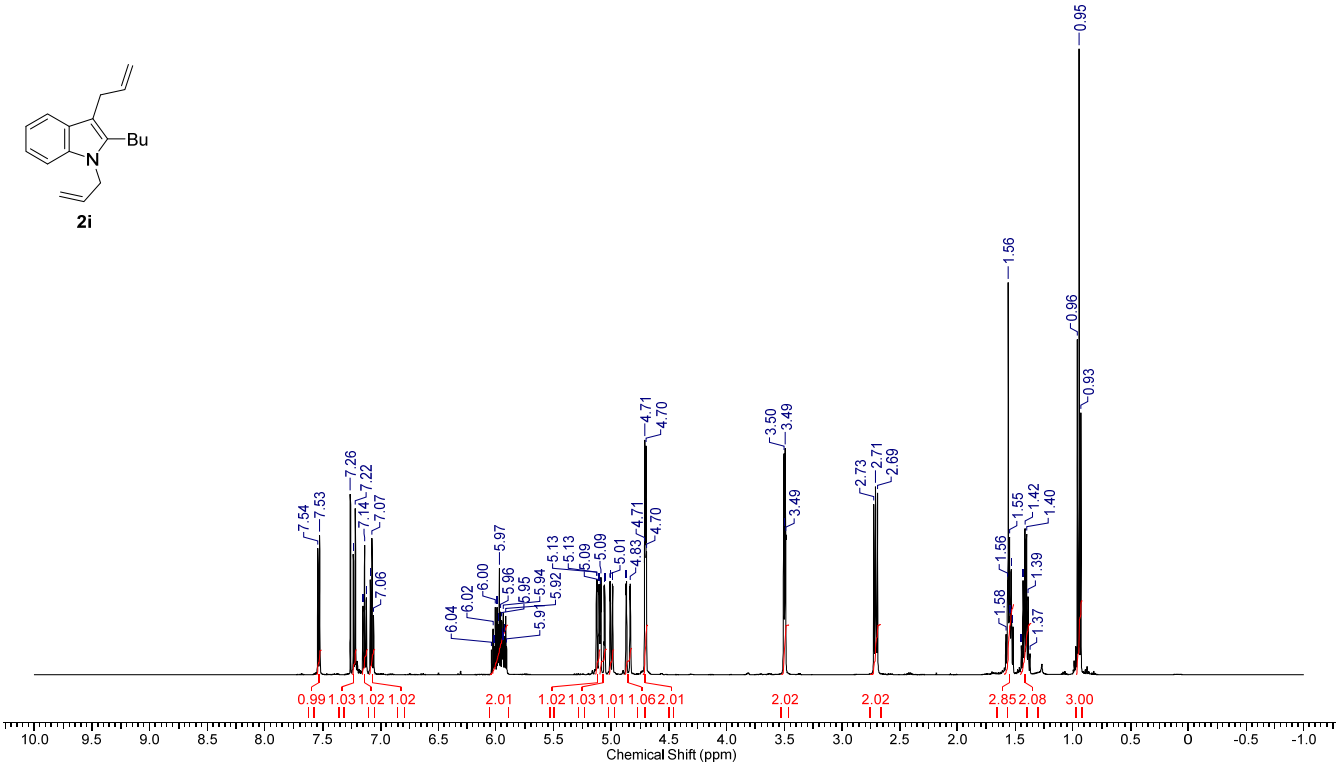
^{13}C NMR (125 MHz, $CDCl_3$) of **2h**



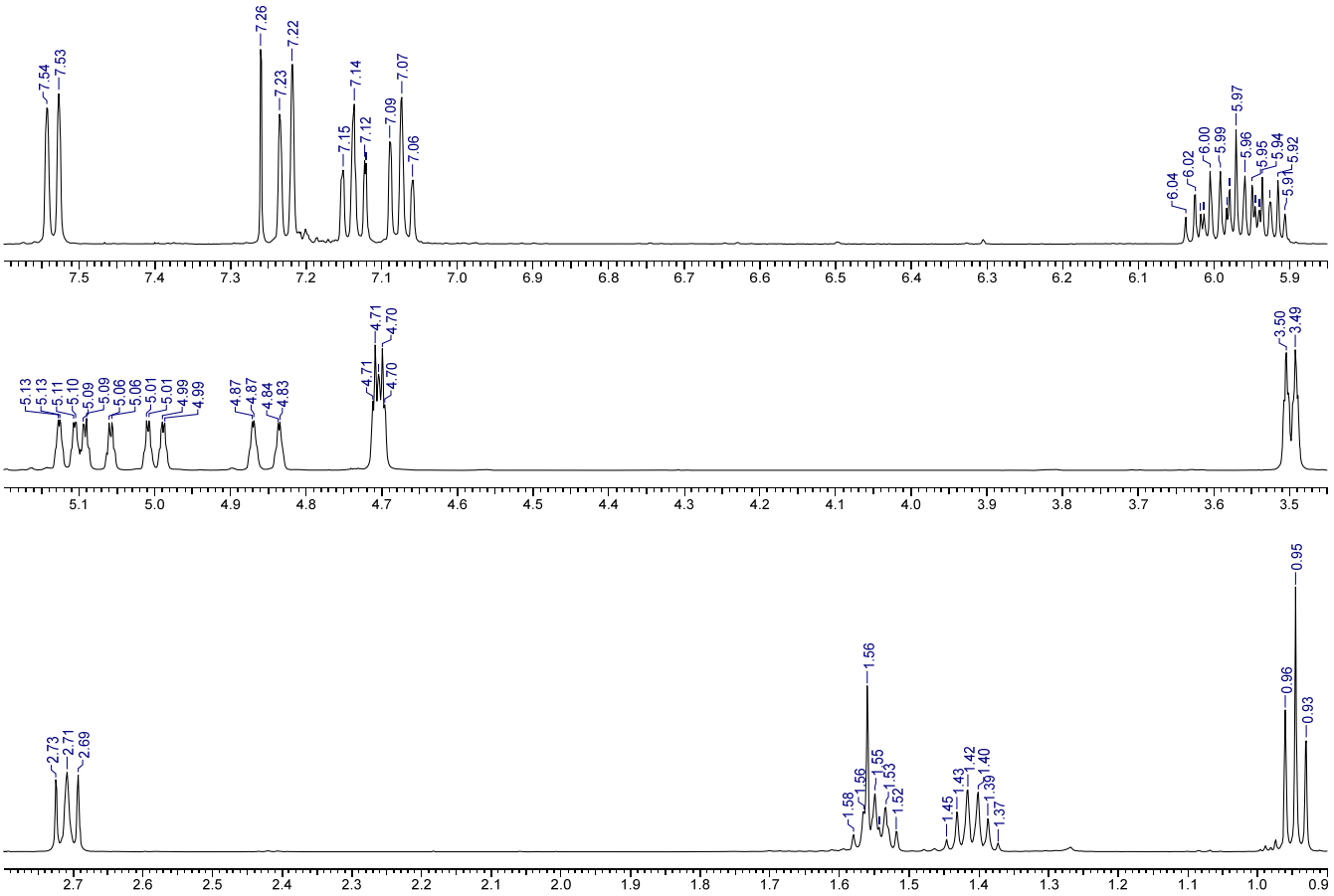
Enlarged view



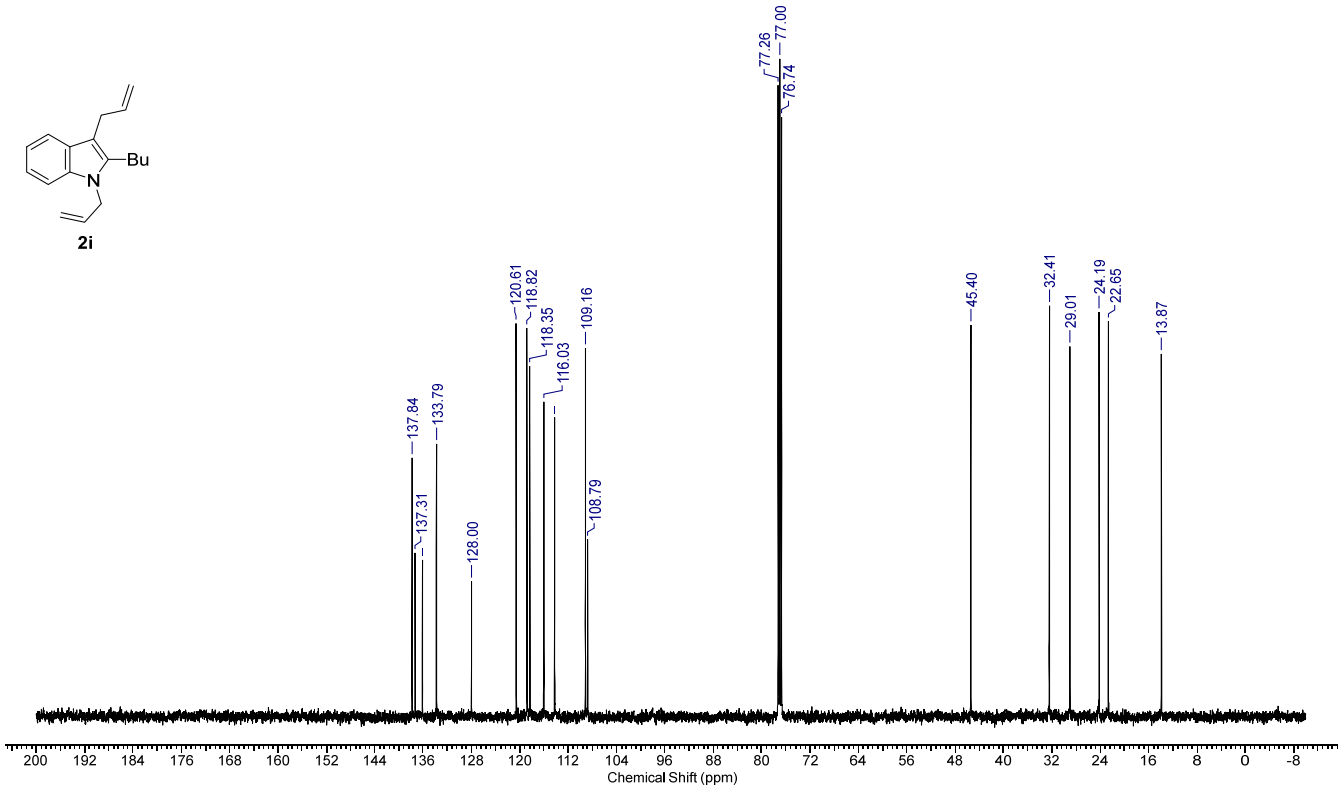
^{1}H NMR (500 MHz, $CDCl_3$) of **2i**



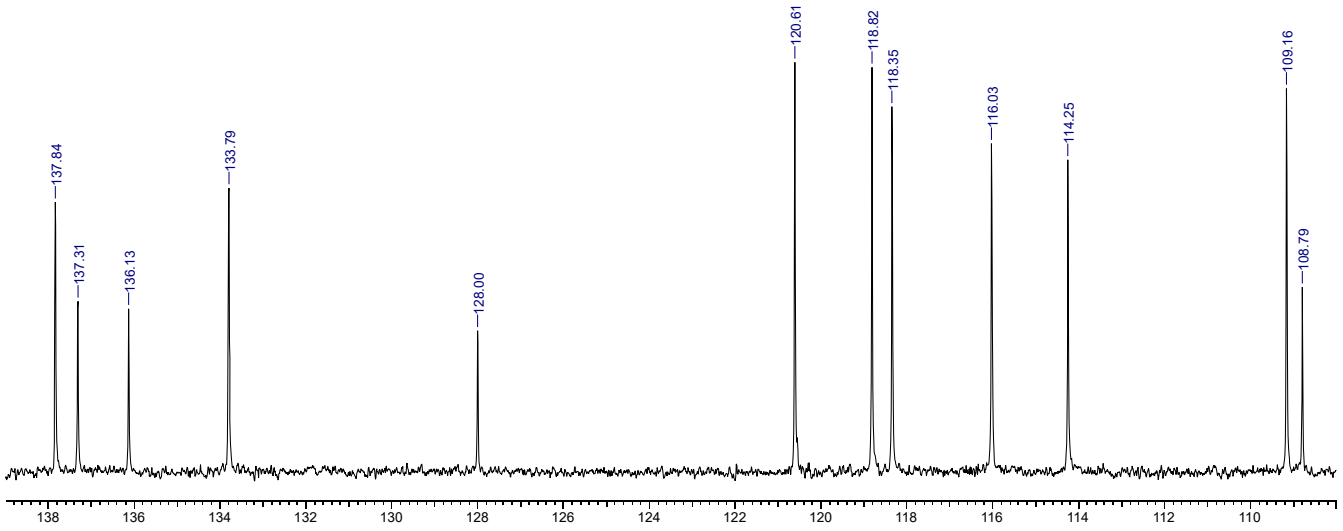
Enlarged view



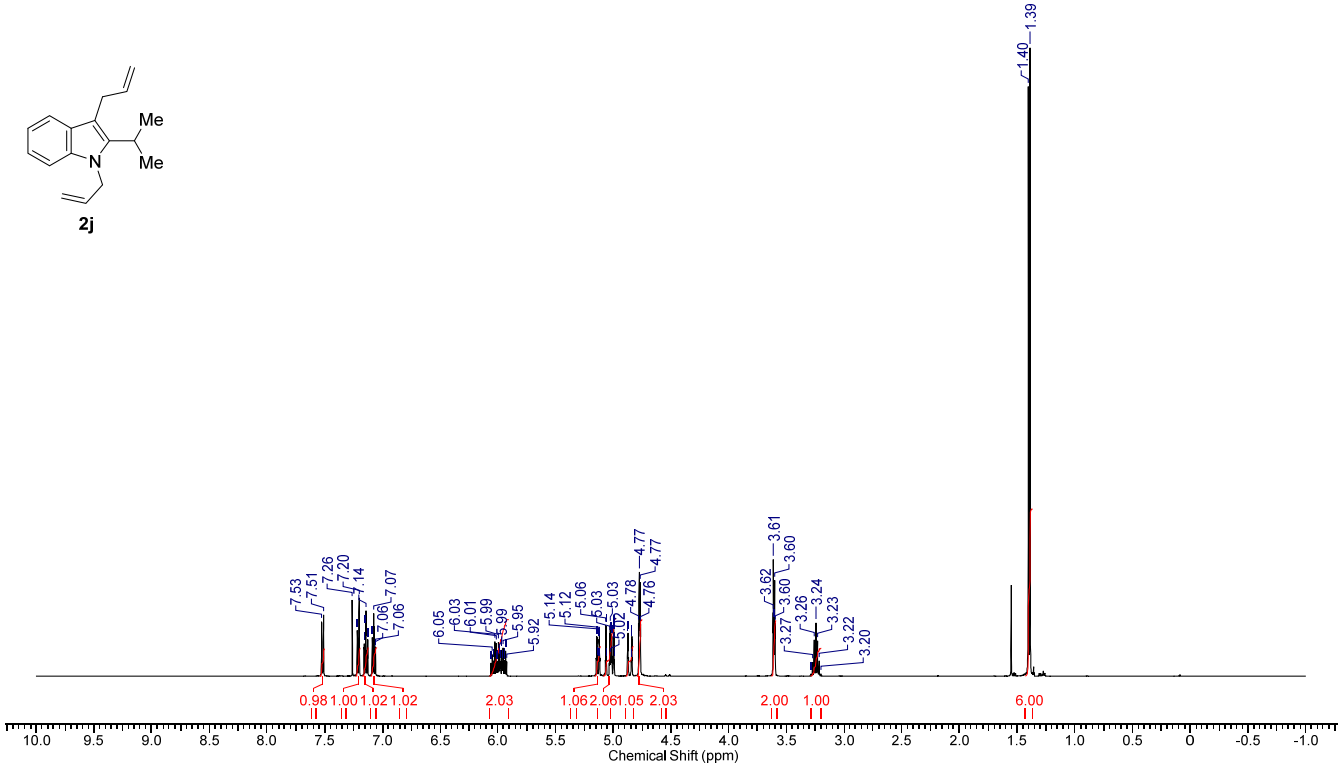
^{13}C NMR (125 MHz, $CDCl_3$) of **2i**



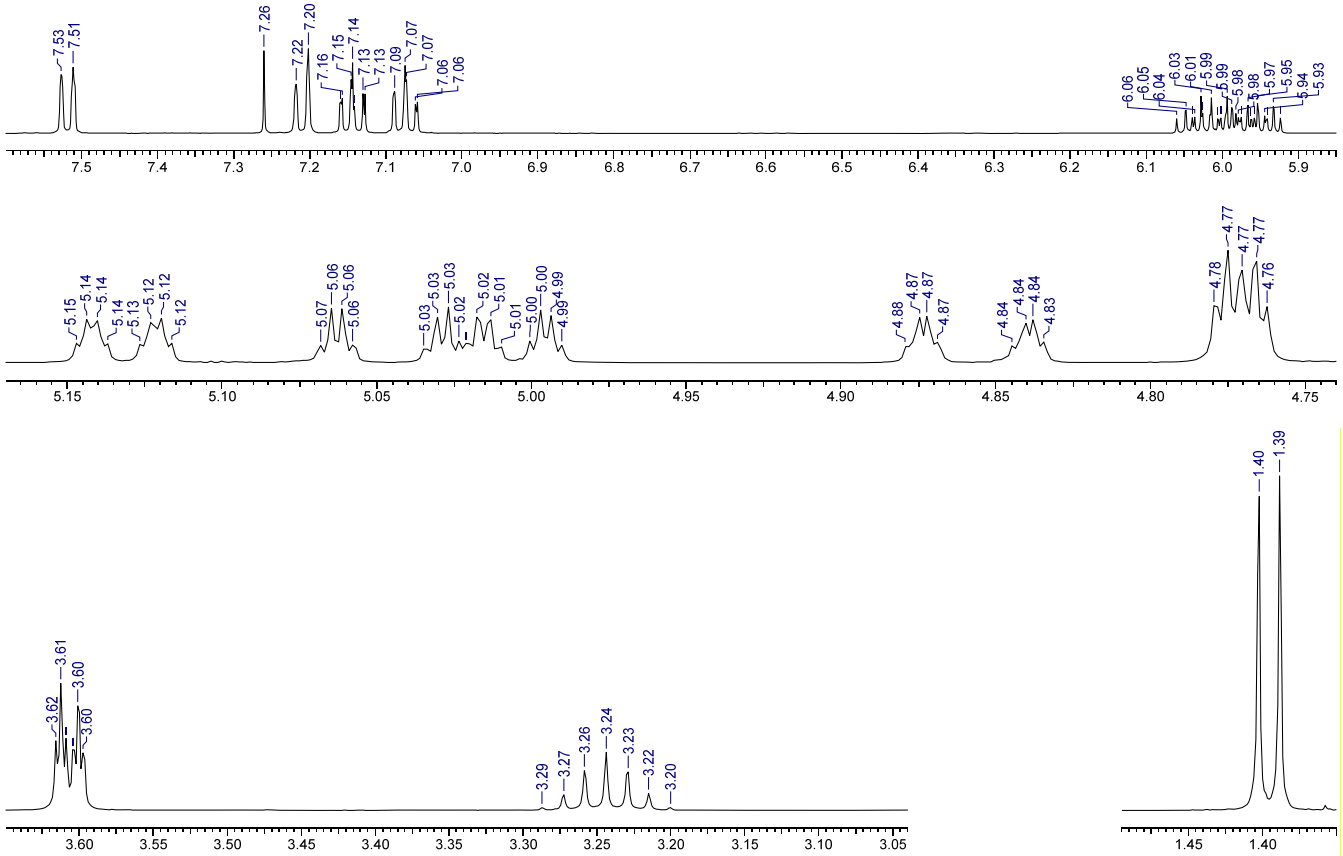
Enlarged view



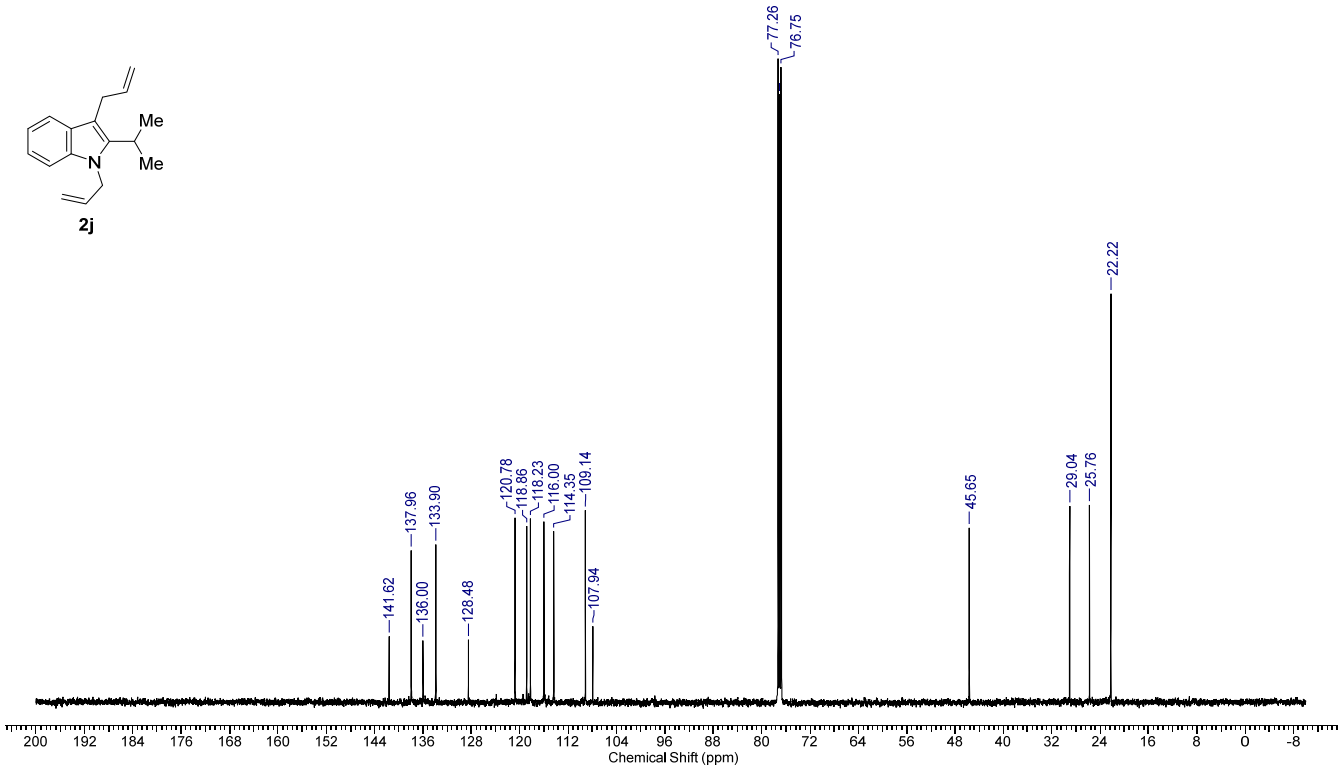
^{1}H NMR (500 MHz, $CDCl_3$) of **2j**



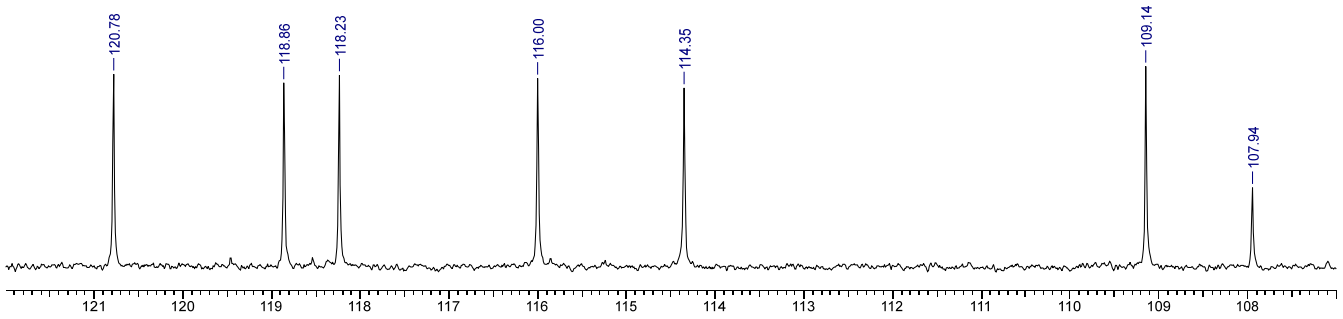
Enlarged view



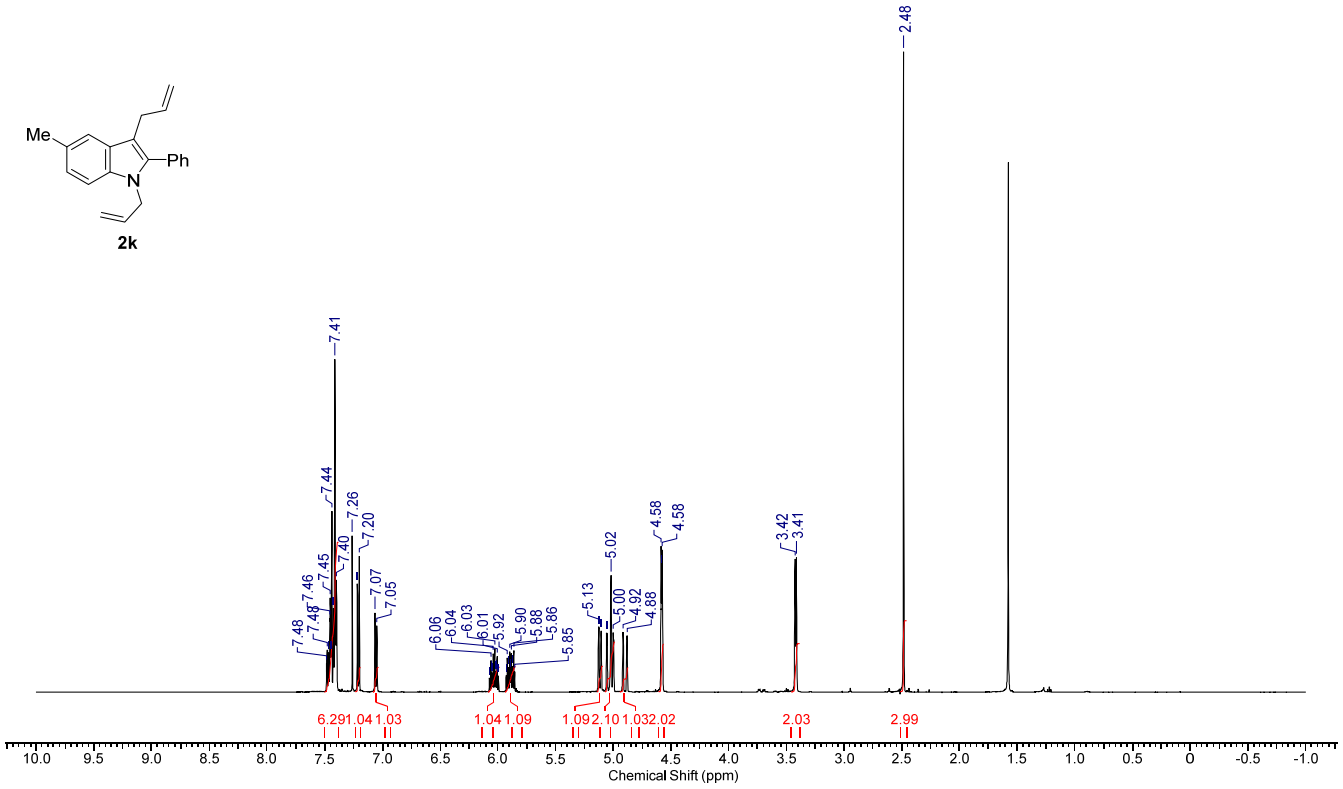
^{13}C NMR (125 MHz, $CDCl_3$) of **2j**



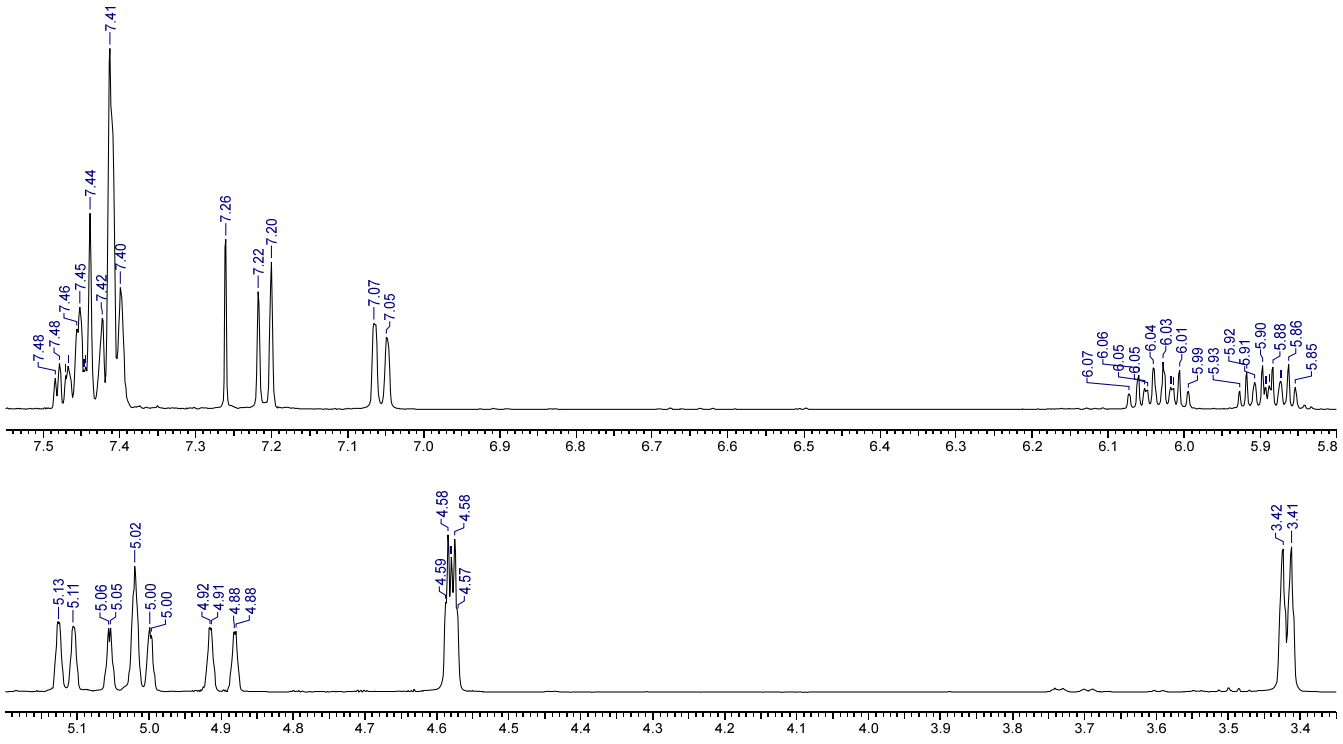
Enlarged view



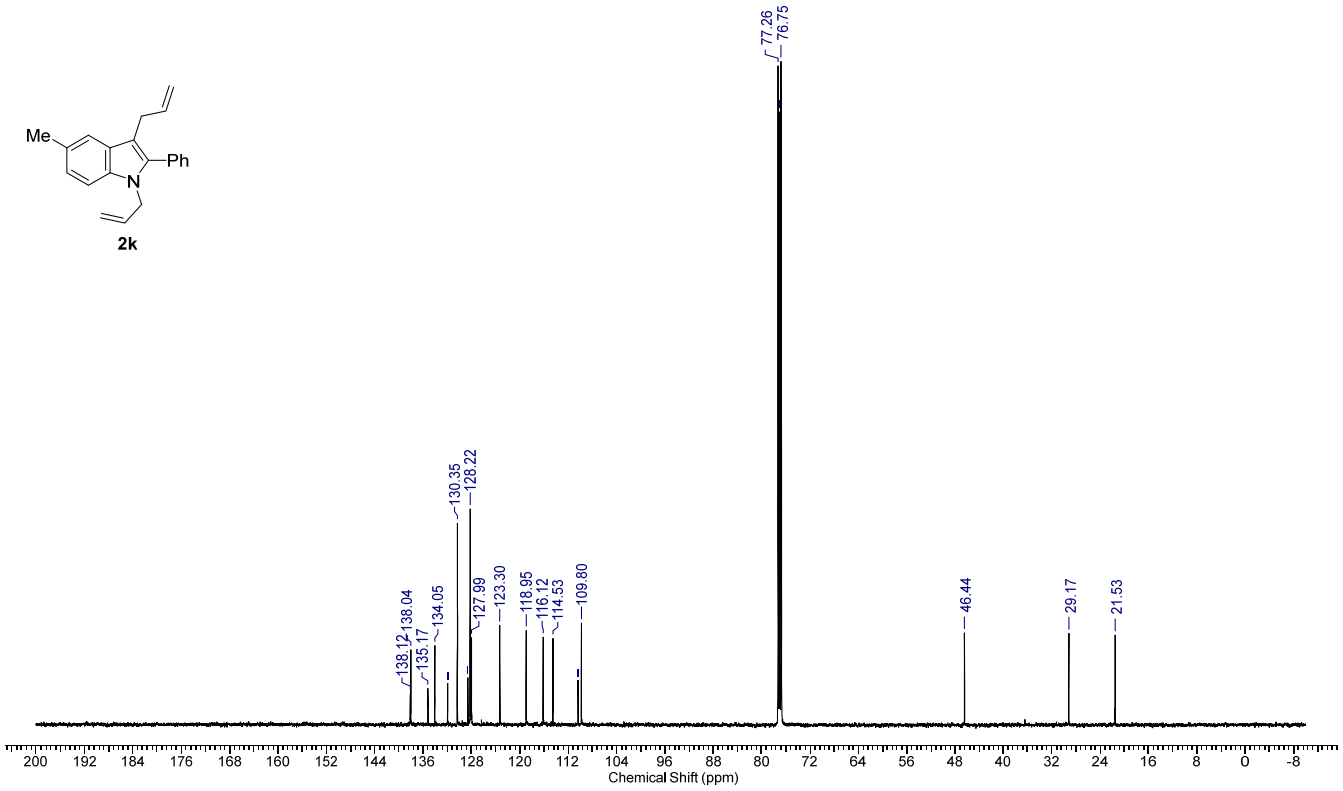
^{1}H NMR (500 MHz, $CDCl_3$) of **2k**



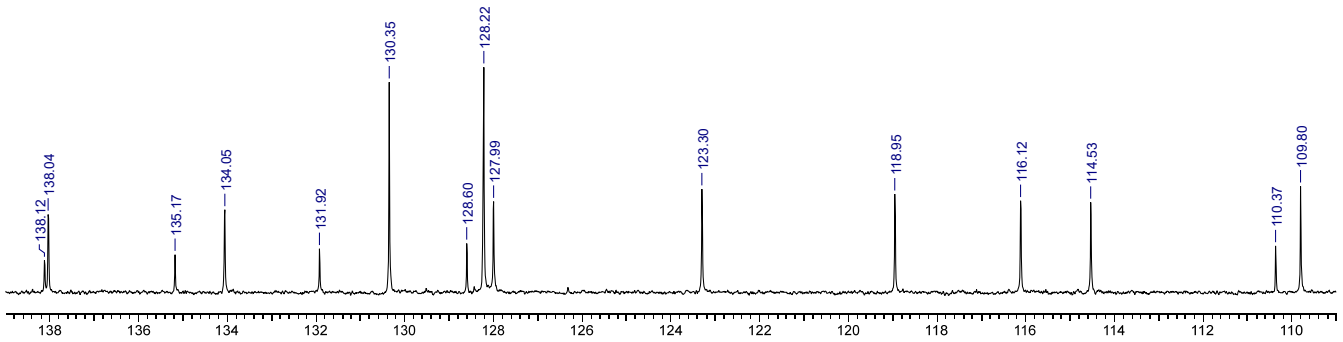
Enlarged view



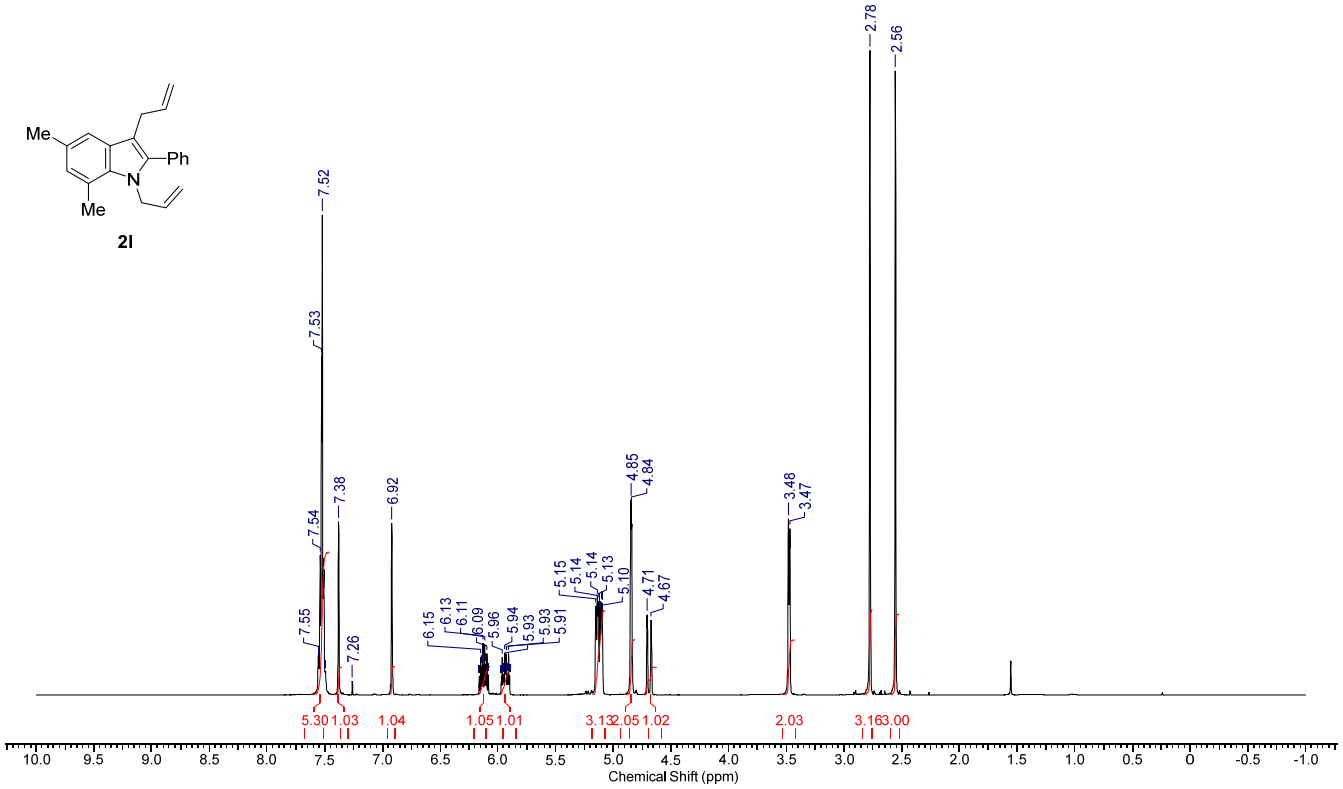
^{13}C NMR (125 MHz, $CDCl_3$) of **2k**



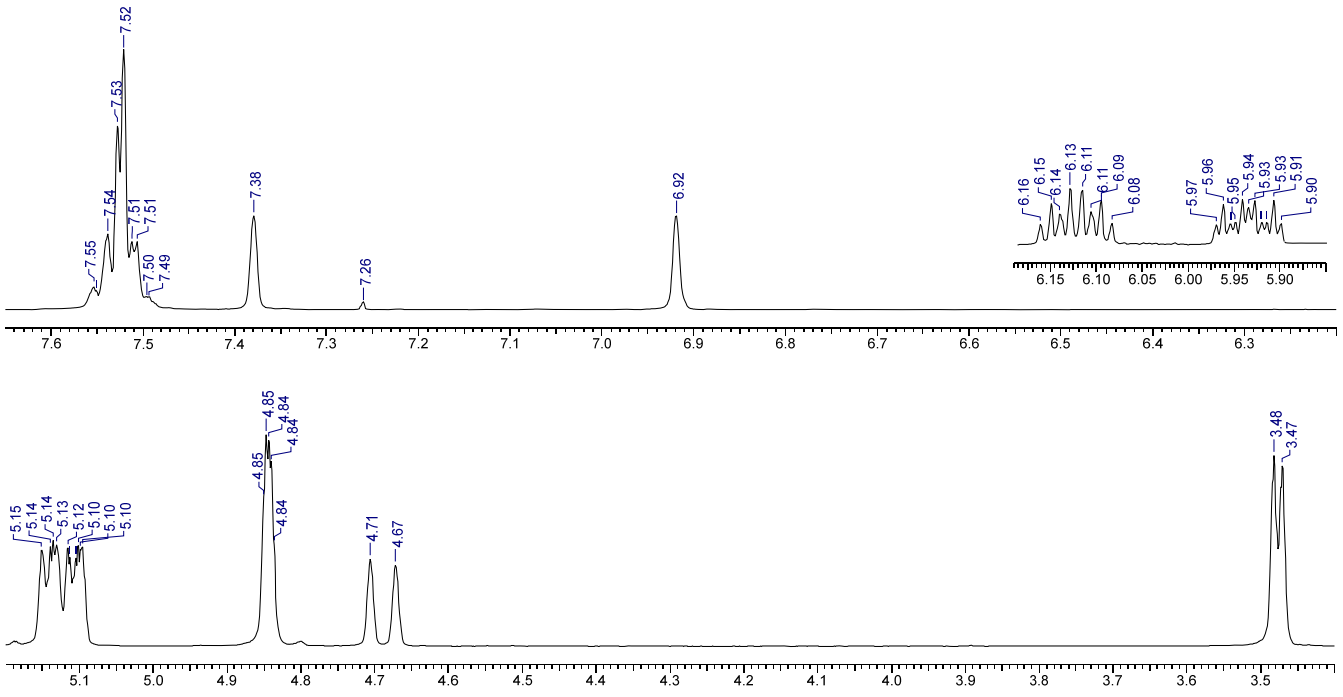
Enlarged view



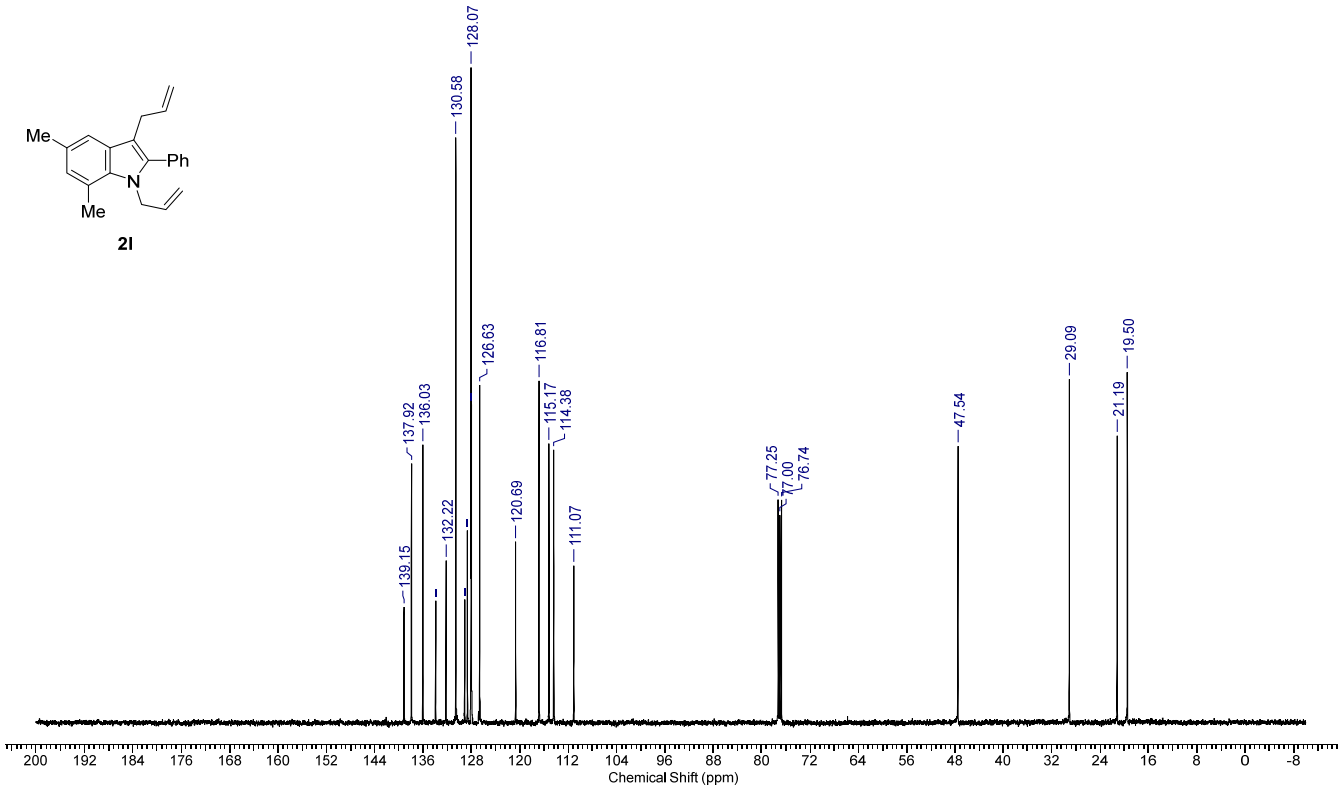
^{1}H NMR (500 MHz, $CDCl_3$) of **2l**



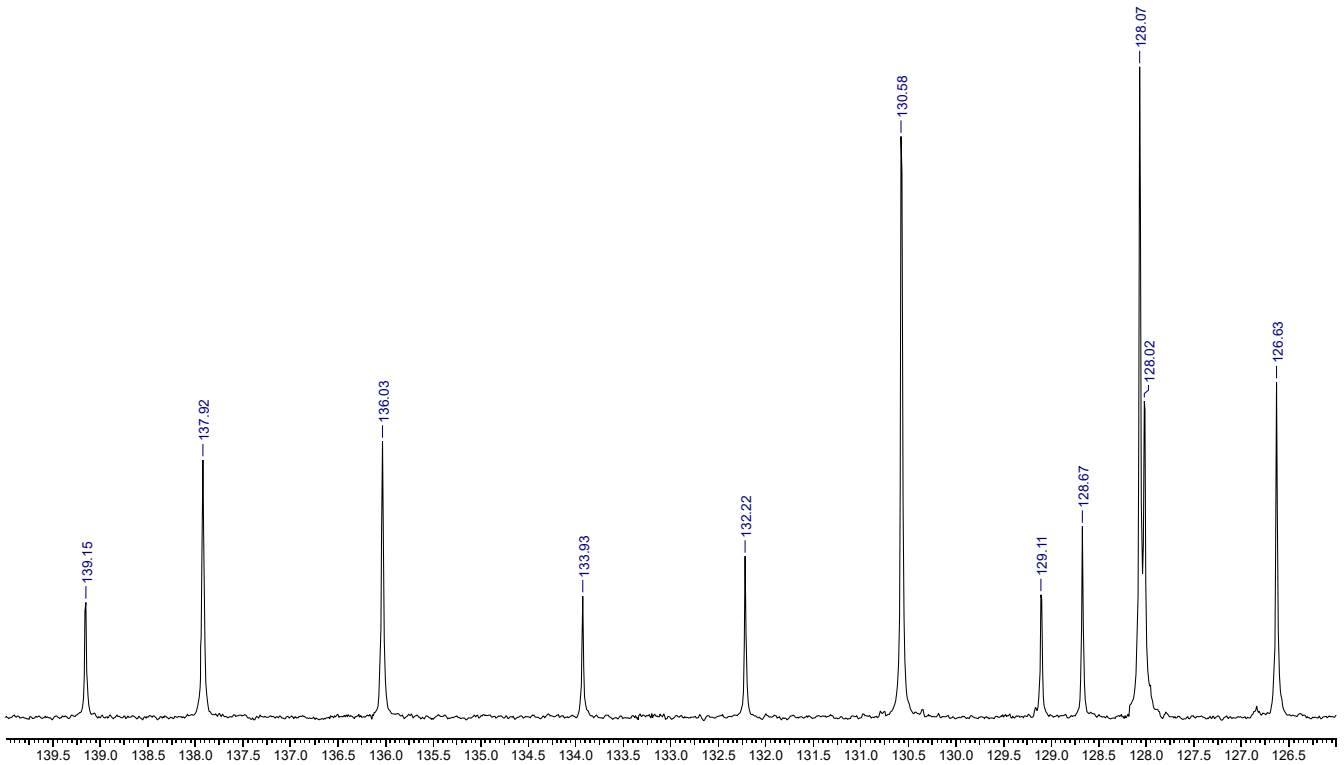
Enlarged view



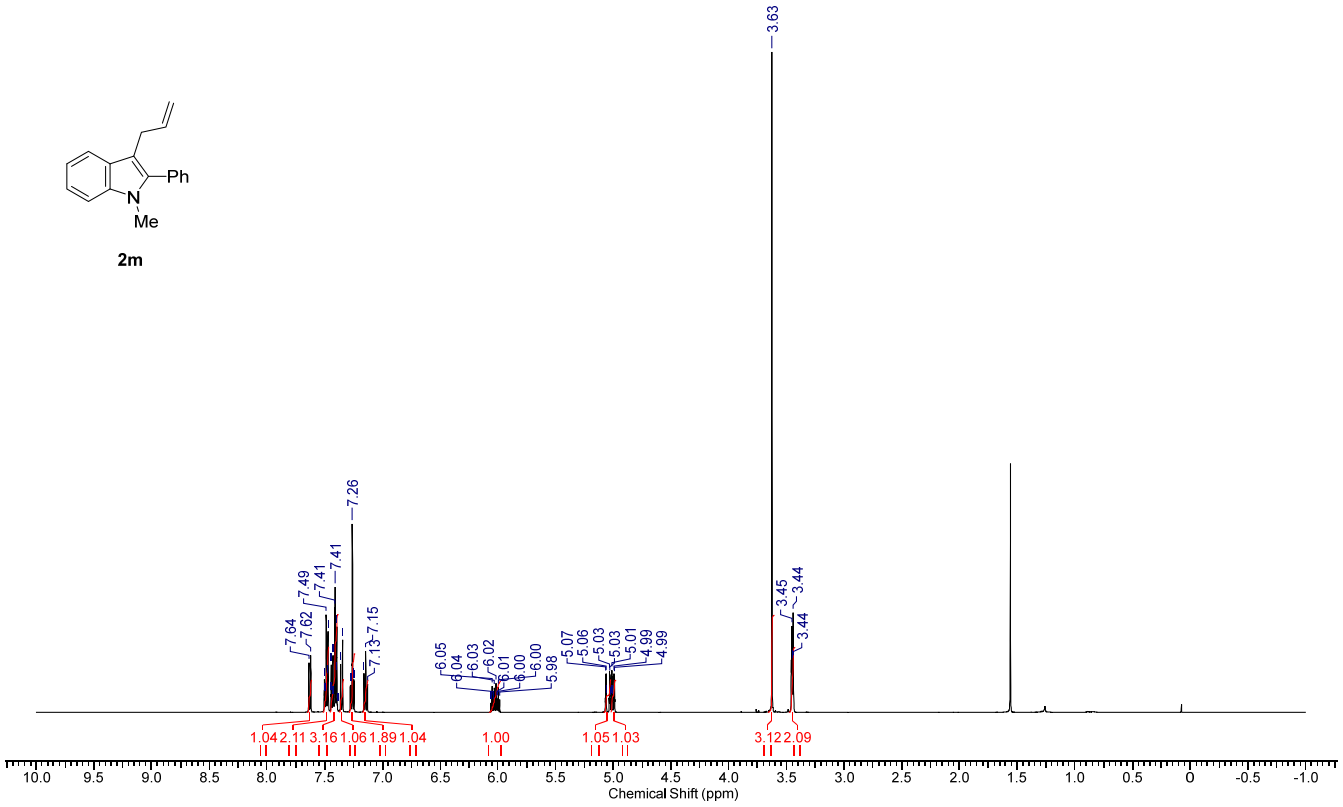
^{13}C NMR (125 MHz, $CDCl_3$) of **2l**



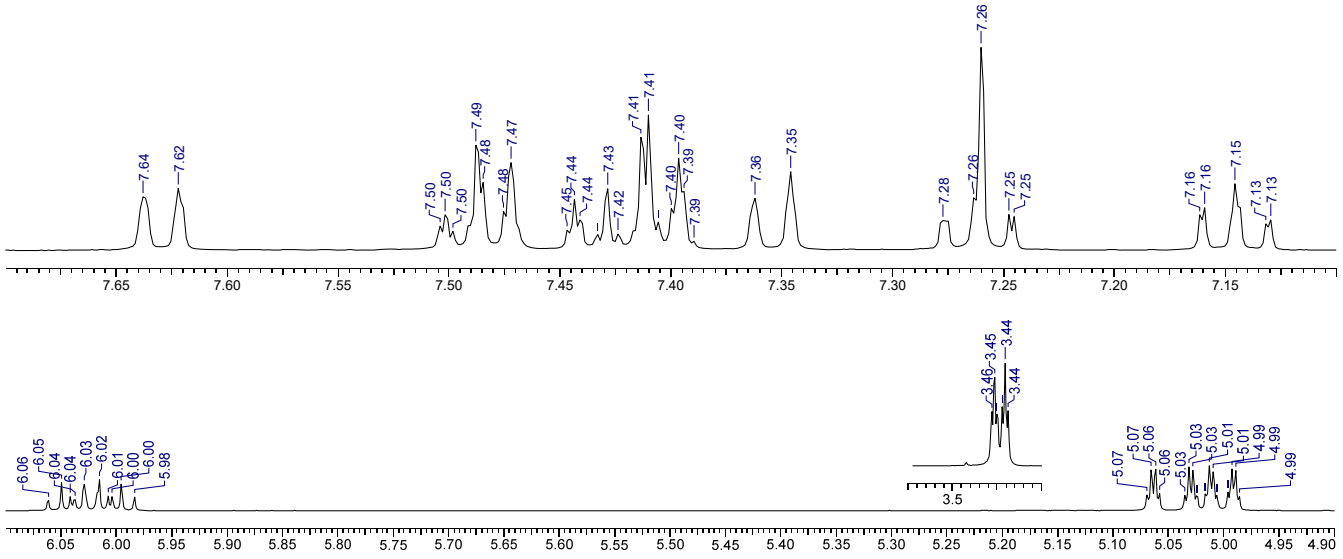
Enlarged view



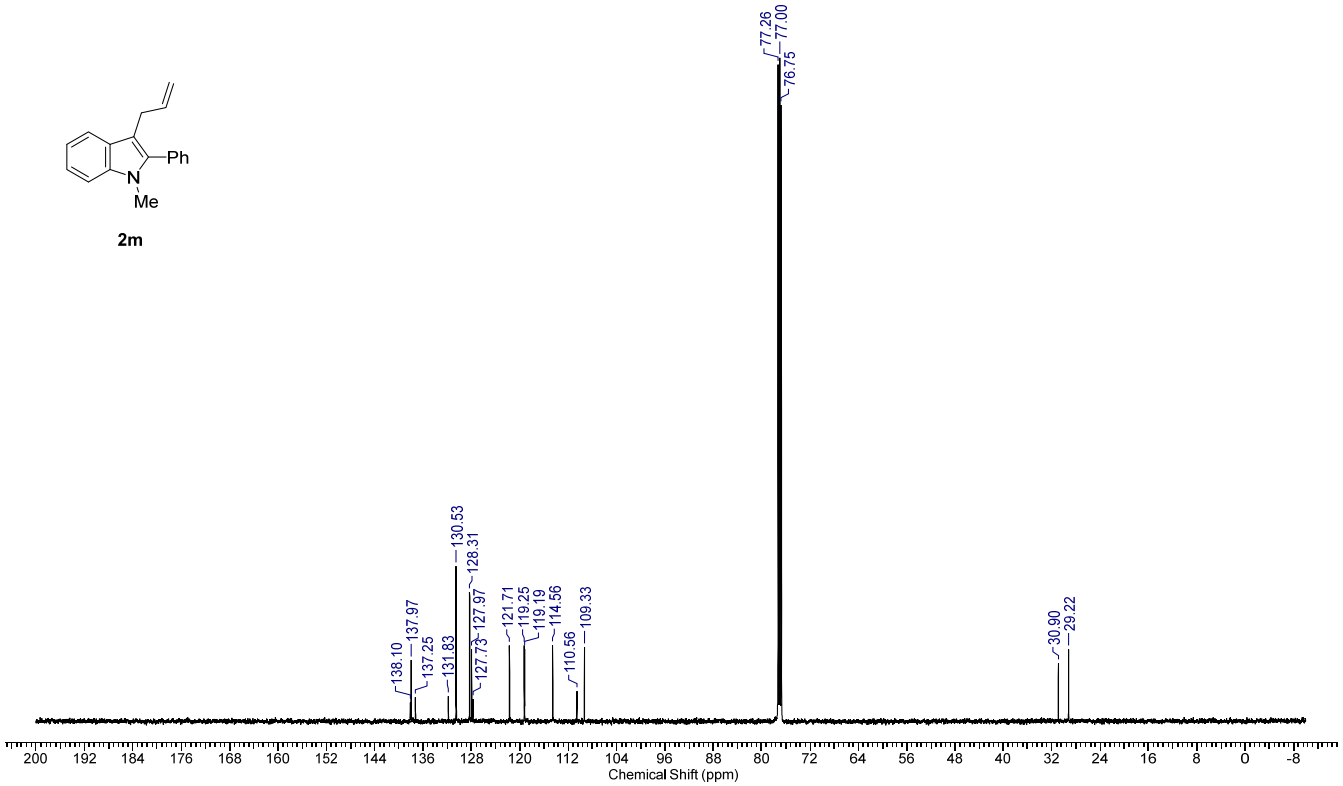
^{1}H NMR (500 MHz, $CDCl_3$) of **2m**



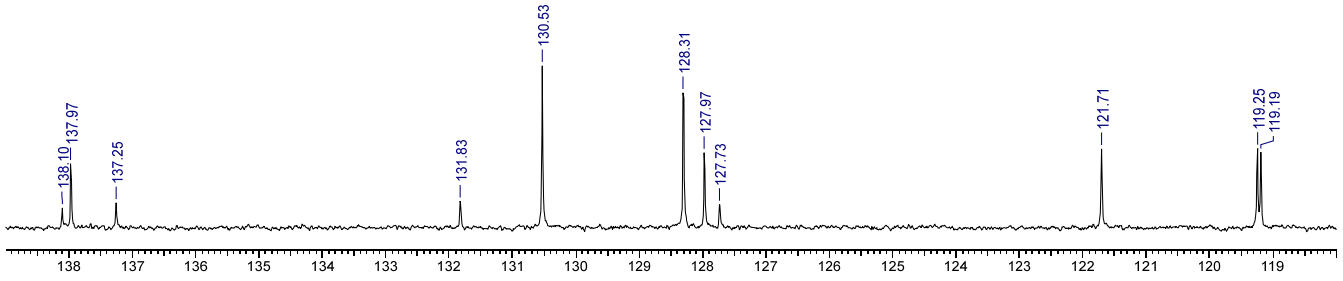
Enlarged view



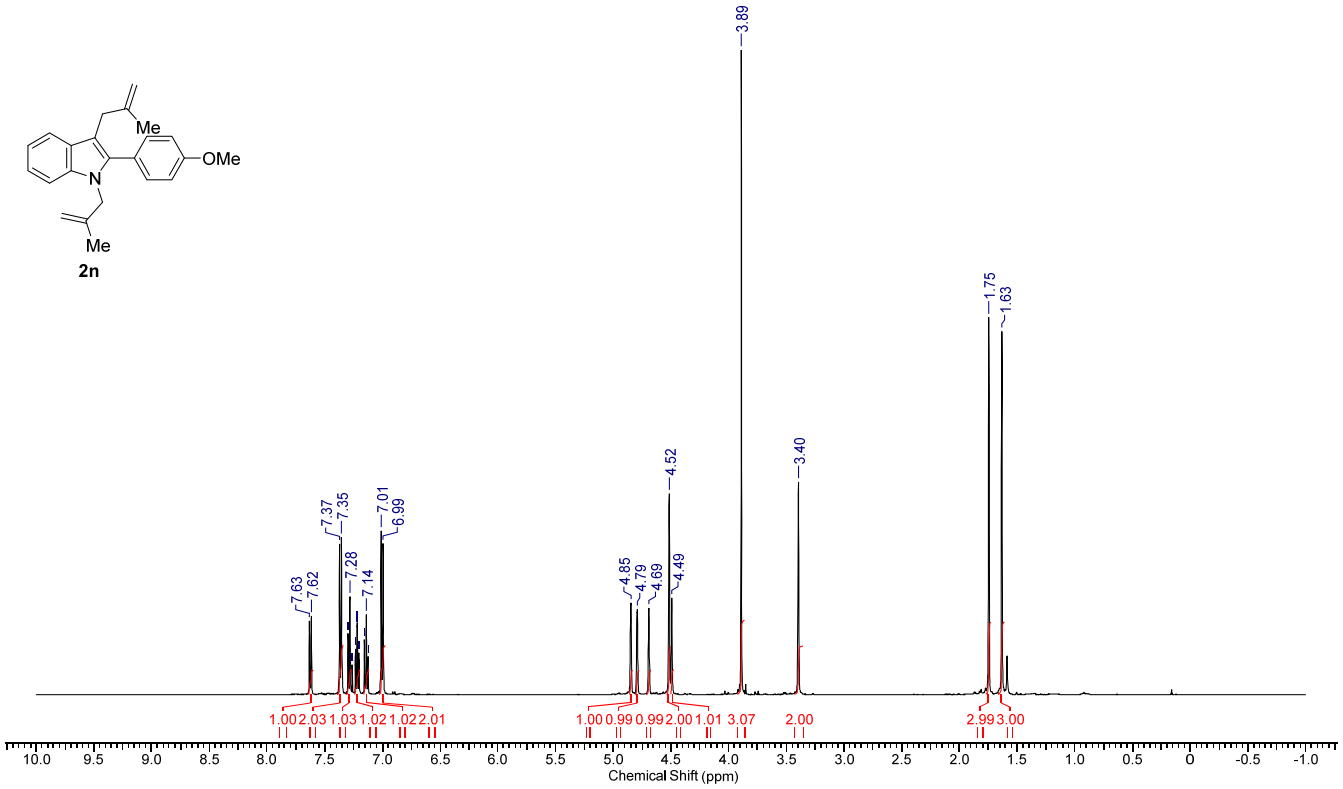
^{13}C NMR (125 MHz, $CDCl_3$) of **2m**



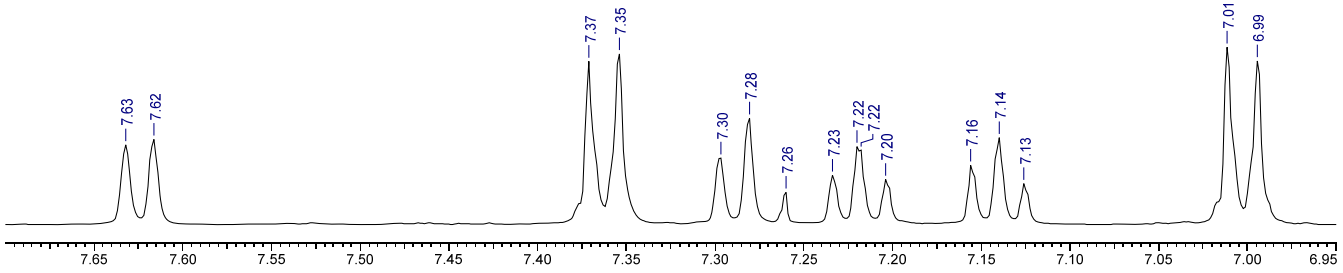
Enlarged view



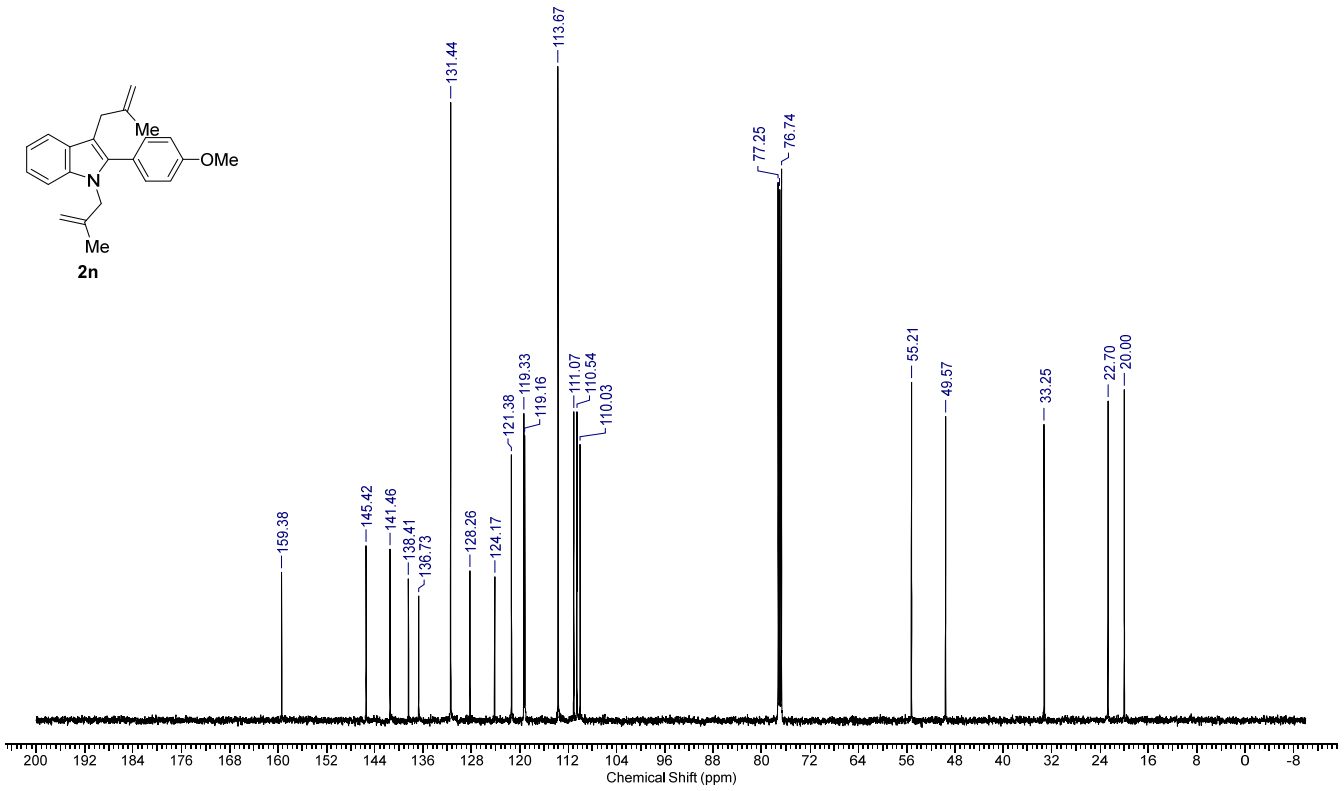
^{1}H NMR (500 MHz, $CDCl_3$) of **2n**



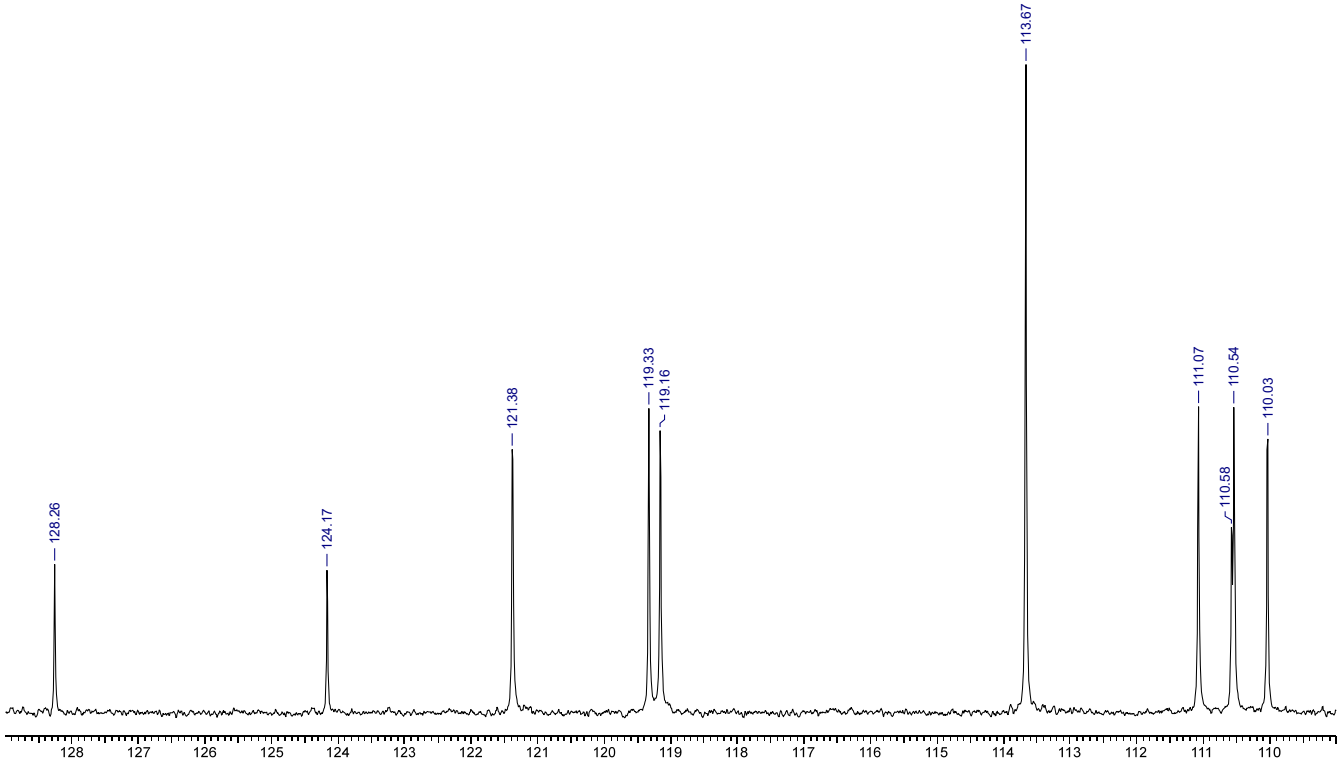
Enlarged view



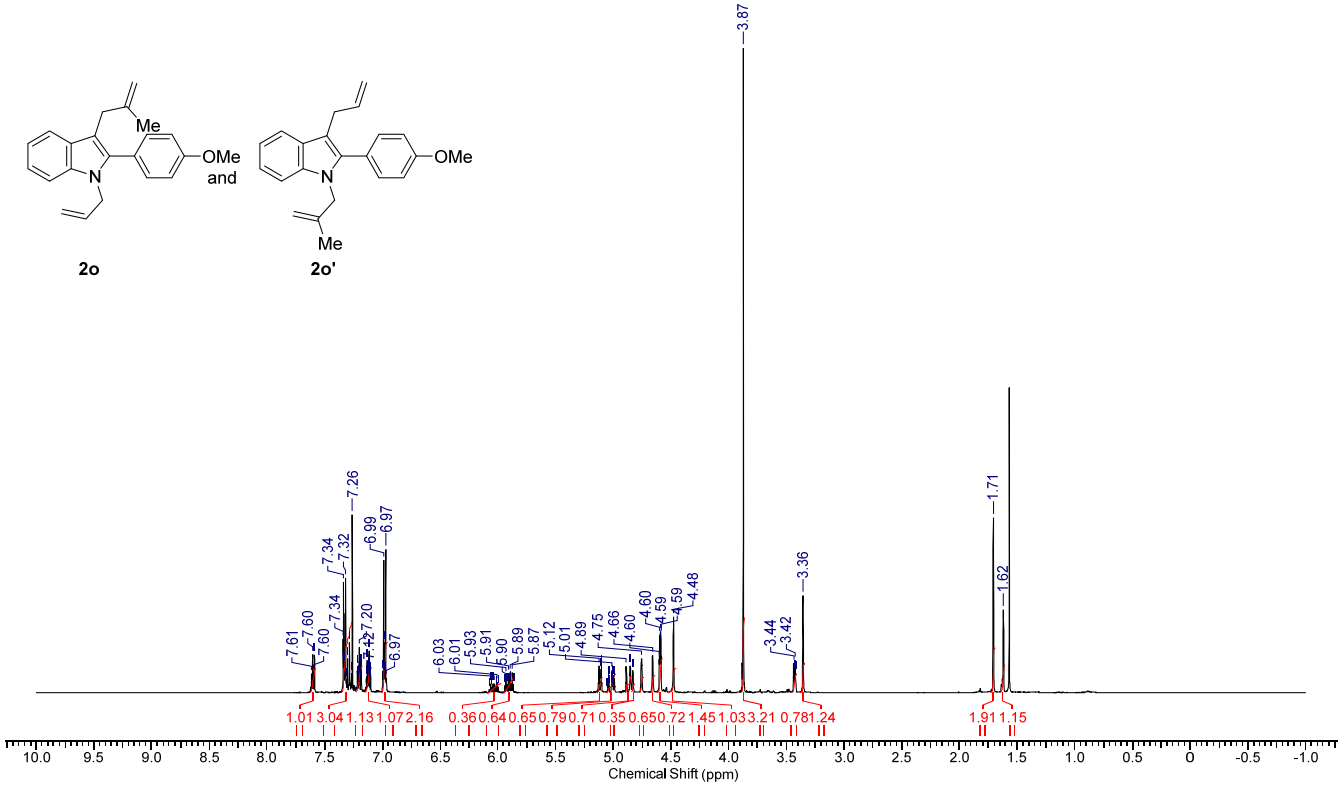
^{13}C NMR (125 MHz, $CDCl_3$) of **2n**



Enlarged view



^{1}H NMR (500 MHz, $CDCl_3$) of **2o** and **2o'** (**2o**:**2o'** = 64:36)



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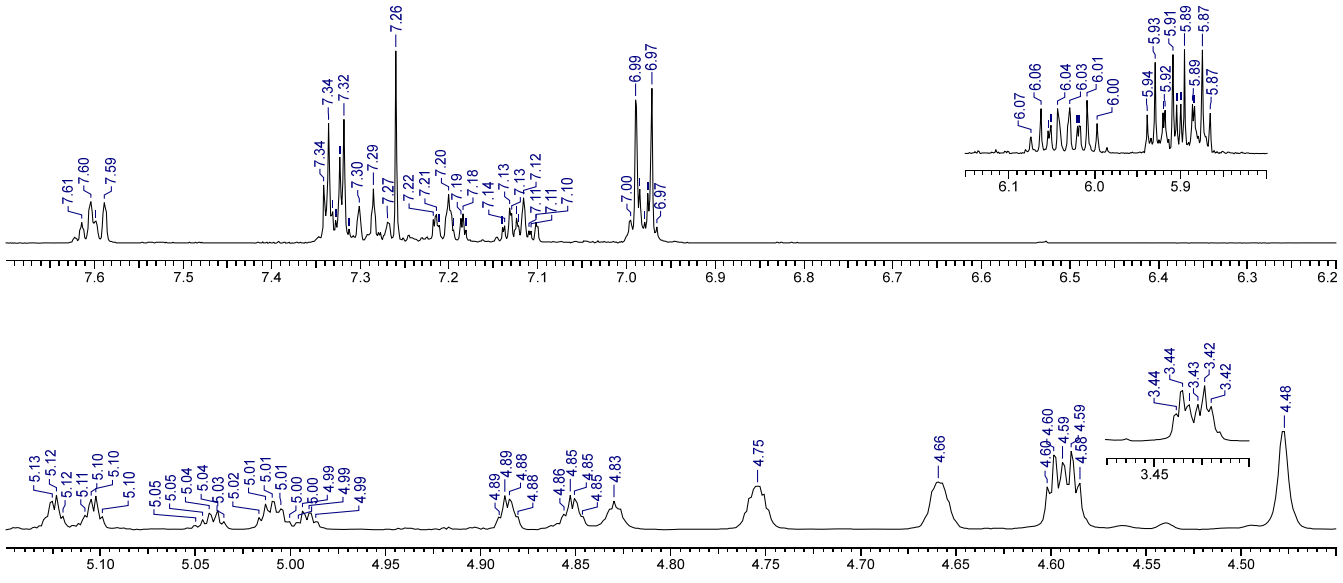
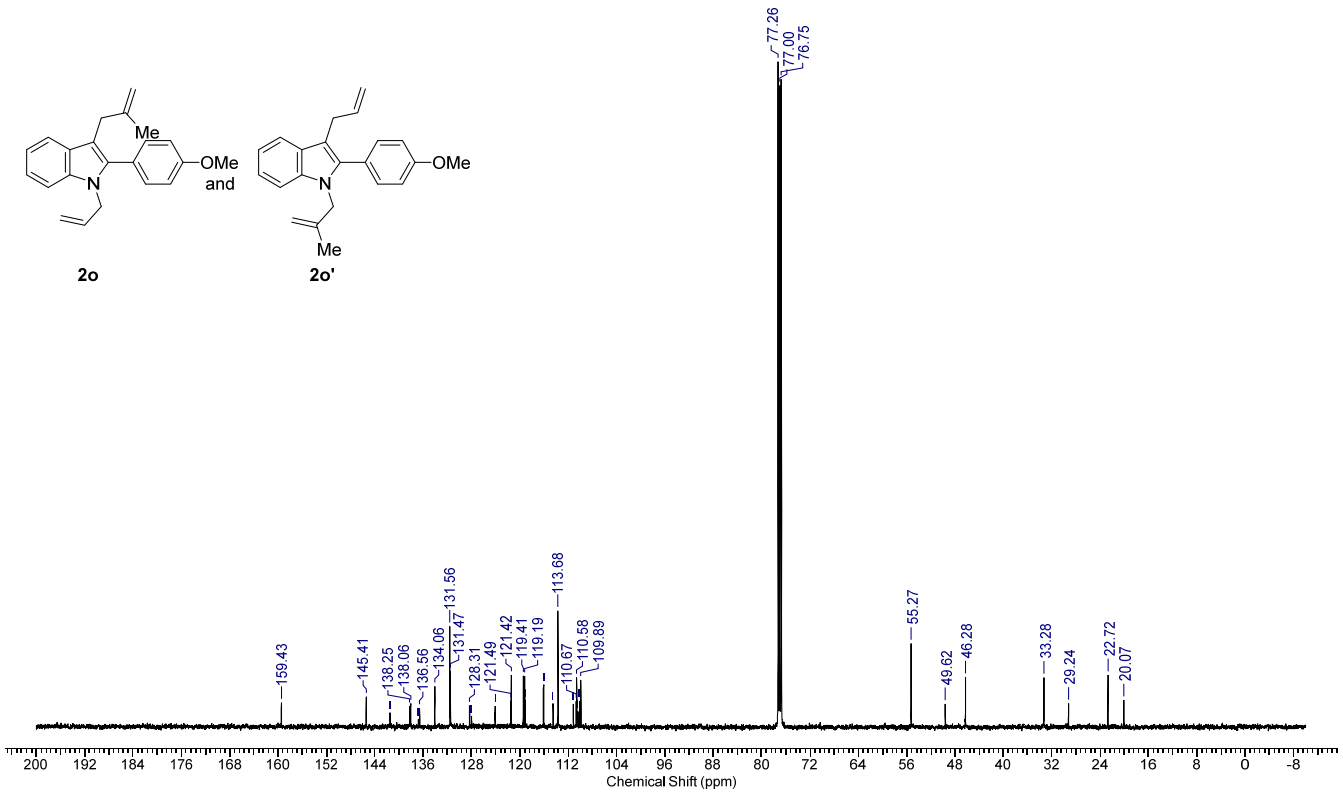
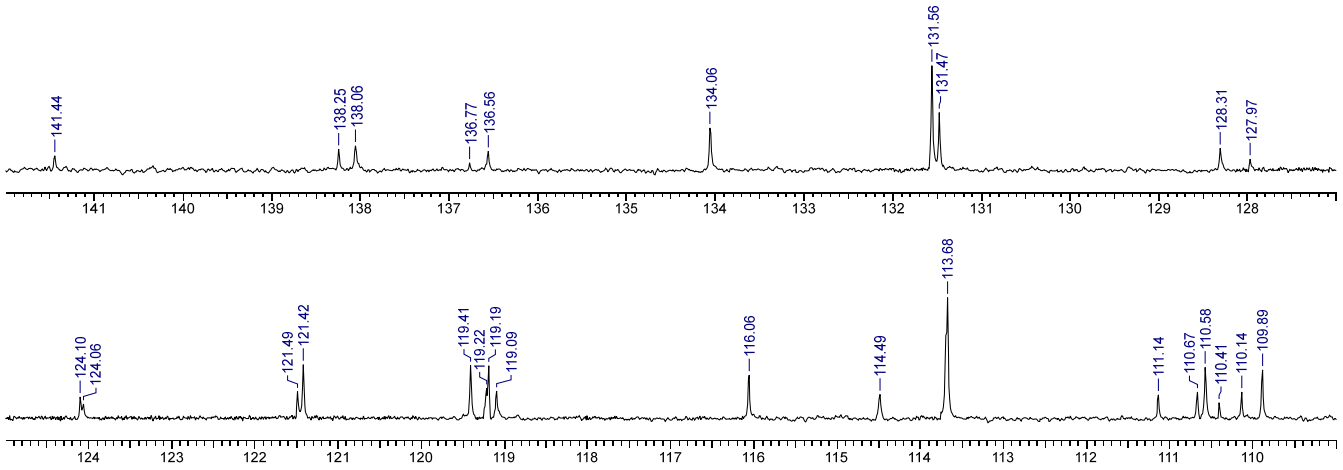


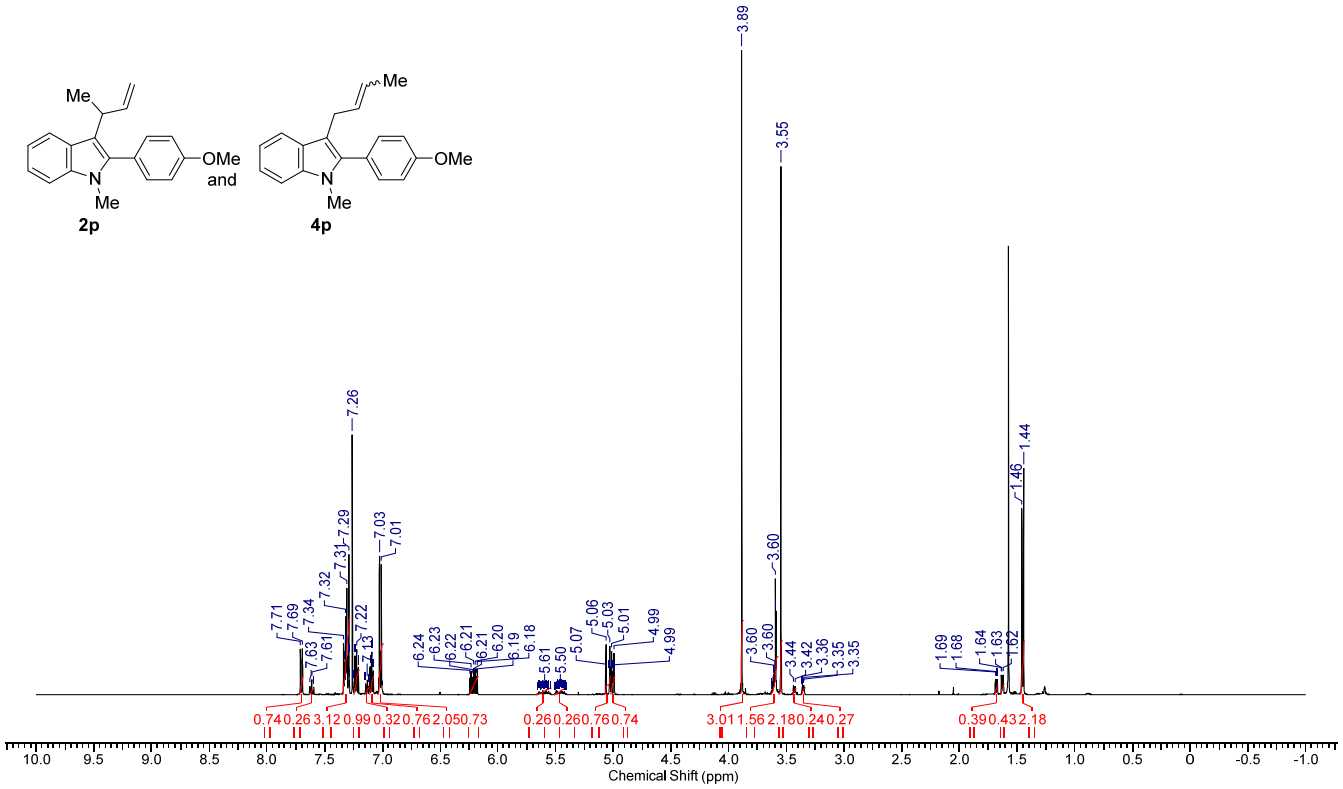
^{13}C NMR (125 MHz, $CDCl_3$) of **2o** and **2o'** (**2o**:**2o'** = 64:36)



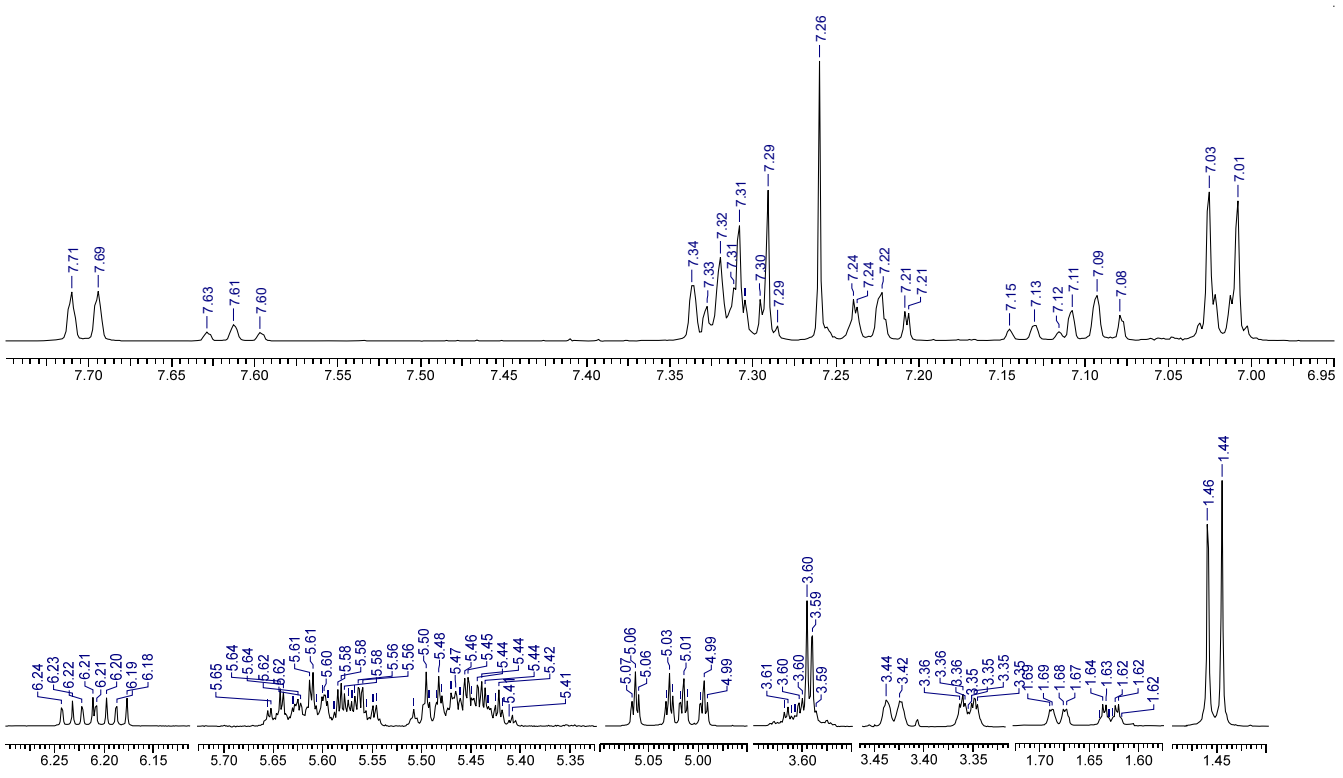
Enlarged view



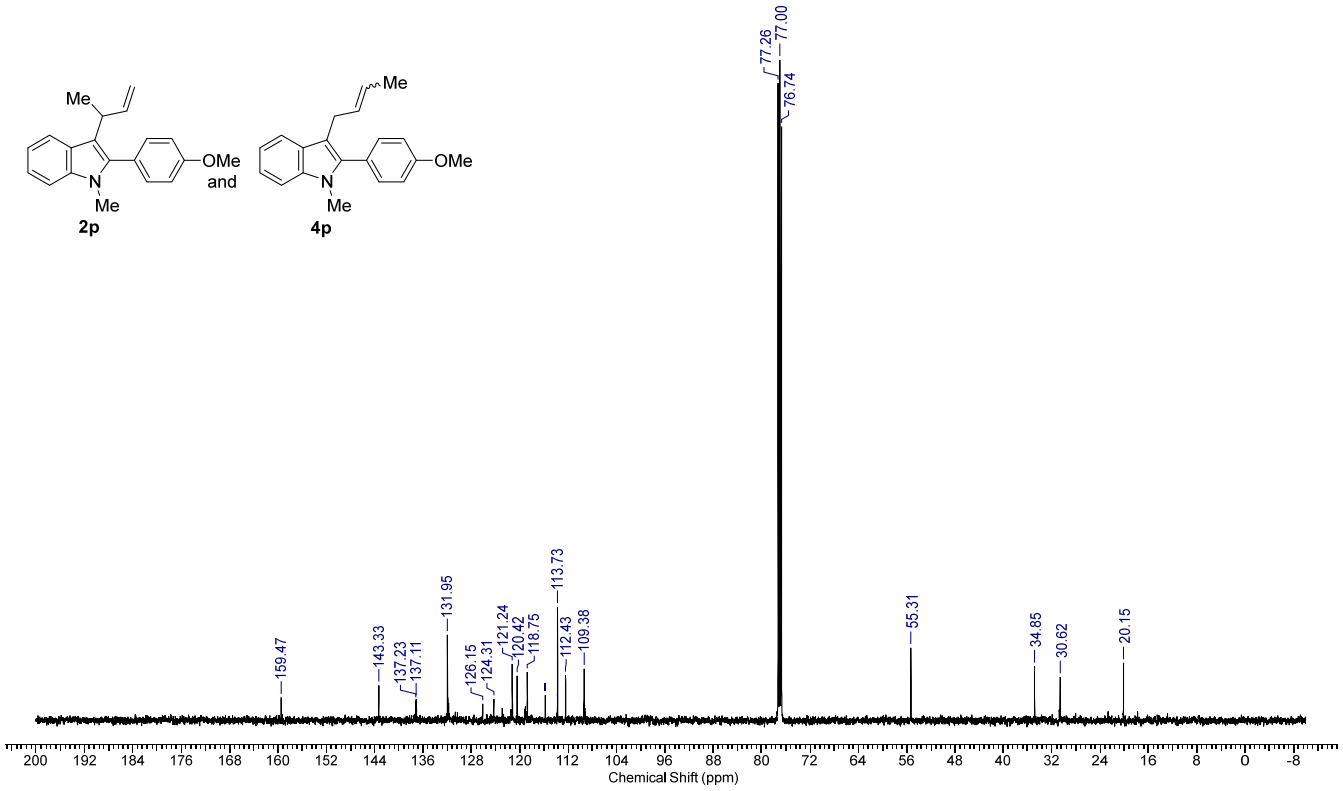
^{1}H NMR (500 MHz, $CDCl_3$) of **2p**, *E*-**4p** and *Z*-**4p** (**2p**:*E*-**4p**:*Z*-**4p** = 73:14:13)



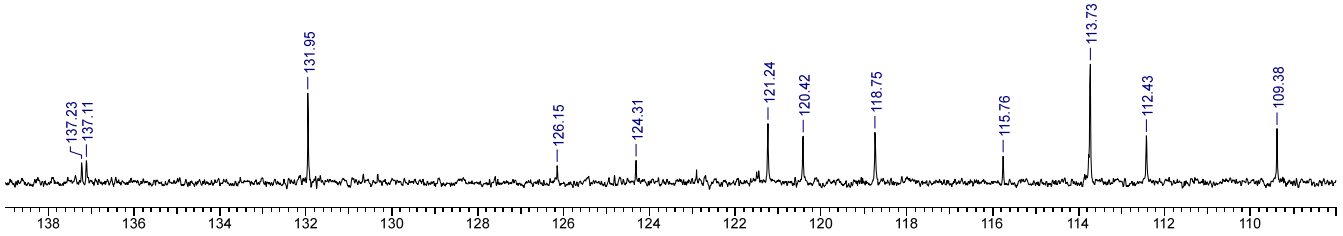
Enlarged view



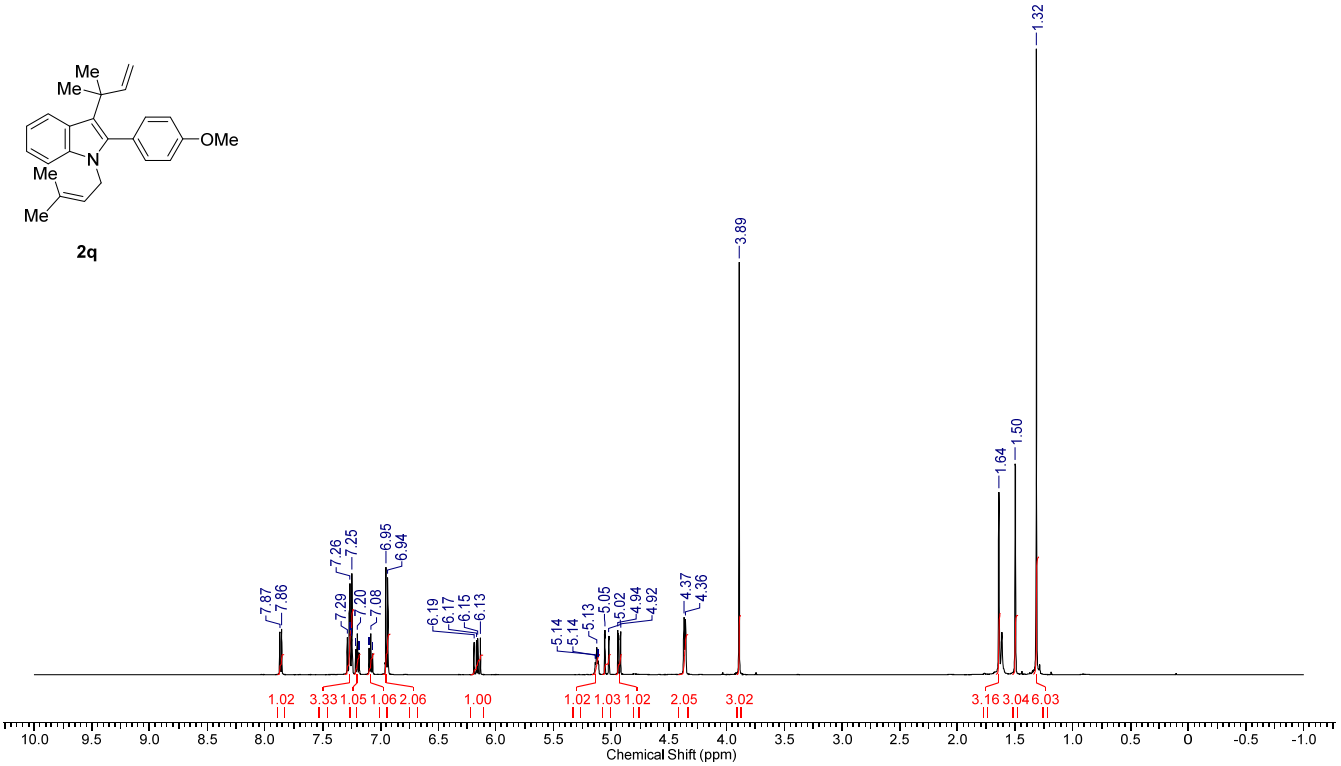
^{13}C NMR (125 MHz, $CDCl_3$) of **2p**, *E*-**4p** and *Z*-**4p** (**2p**:*E*-**4p**:*Z*-**4p** = 73:14:13)



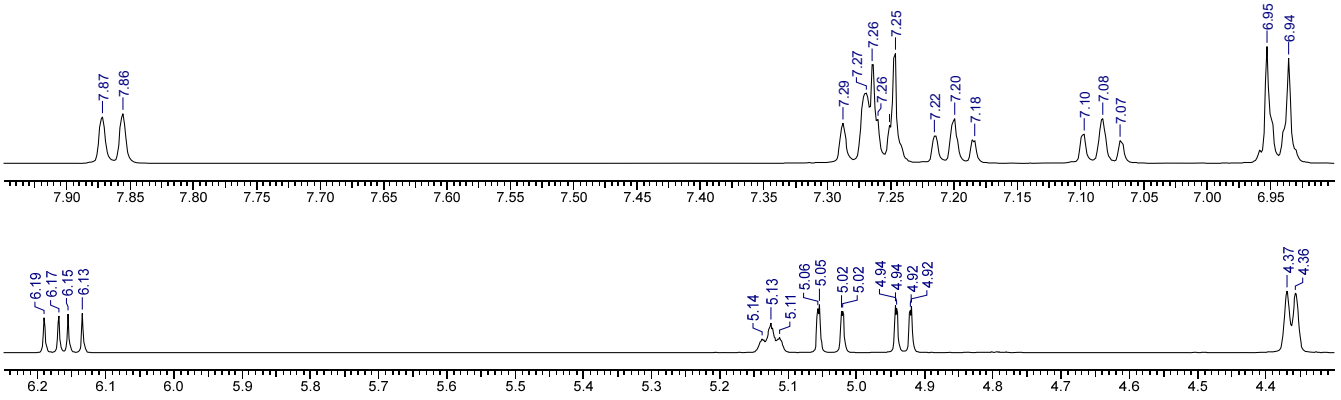
Enlarged view



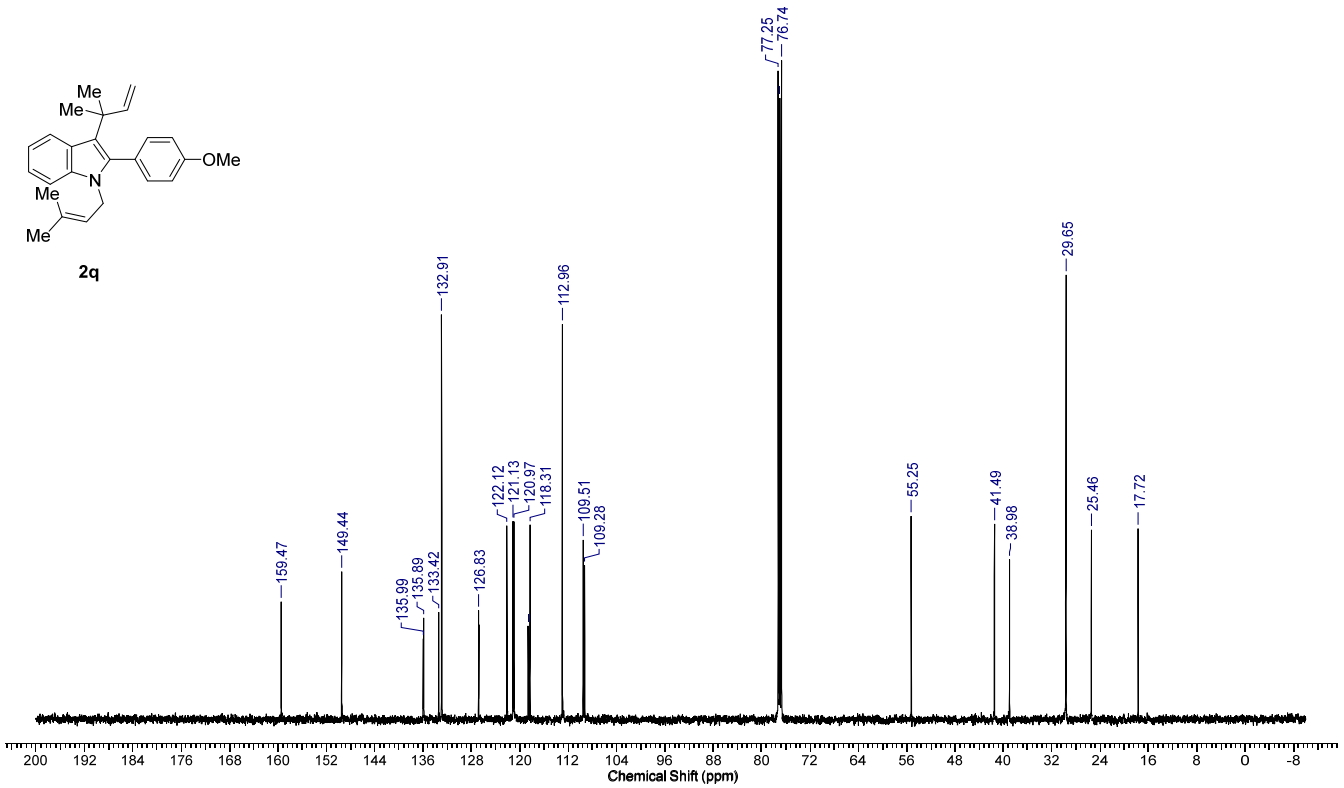
^{1}H NMR (500 MHz, $CDCl_3$) of **2q**



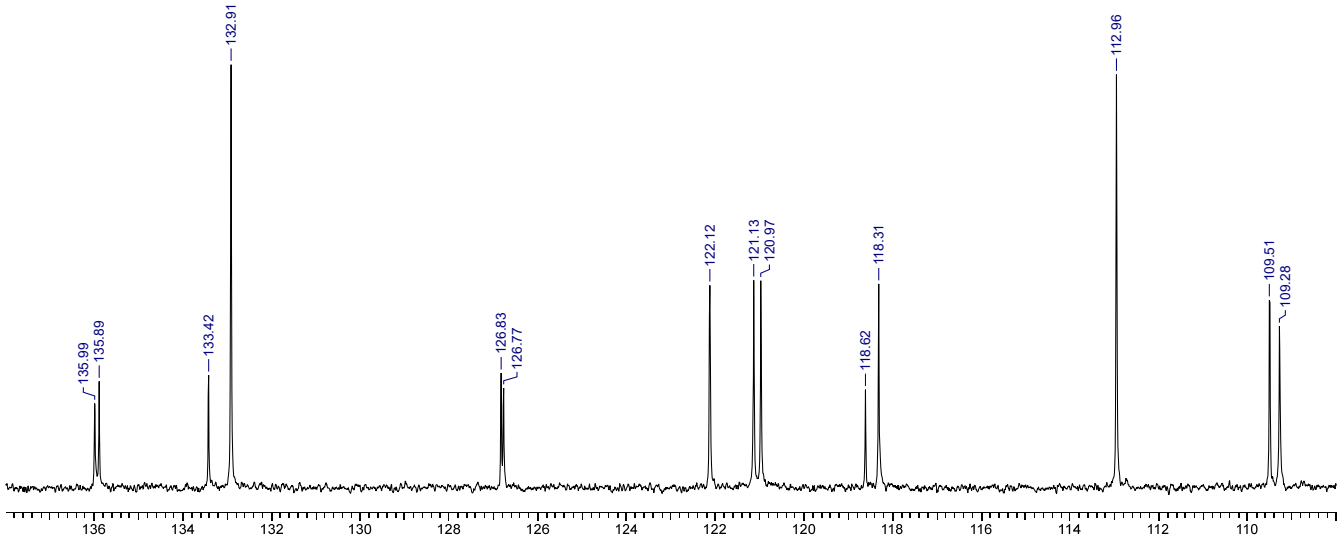
Enlarged view



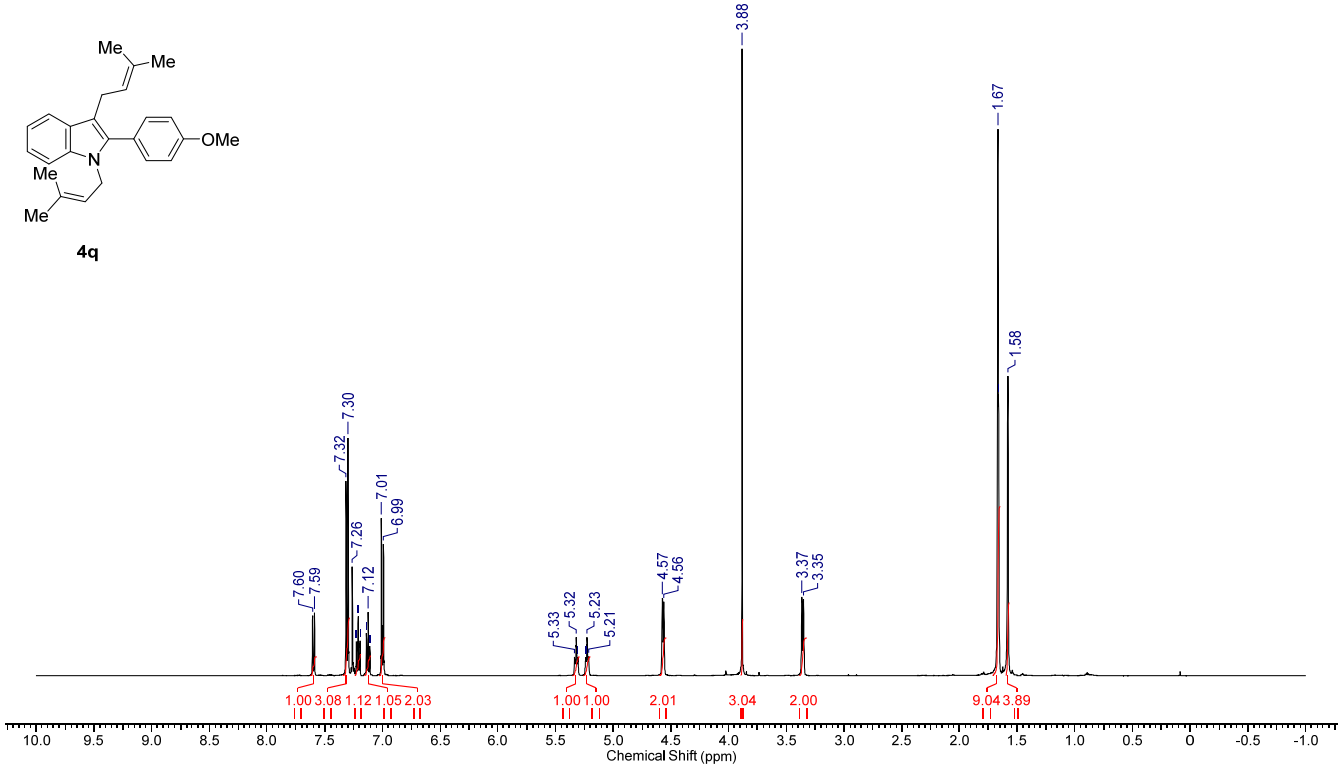
^{13}C NMR (125 MHz, $CDCl_3$) of **2q**



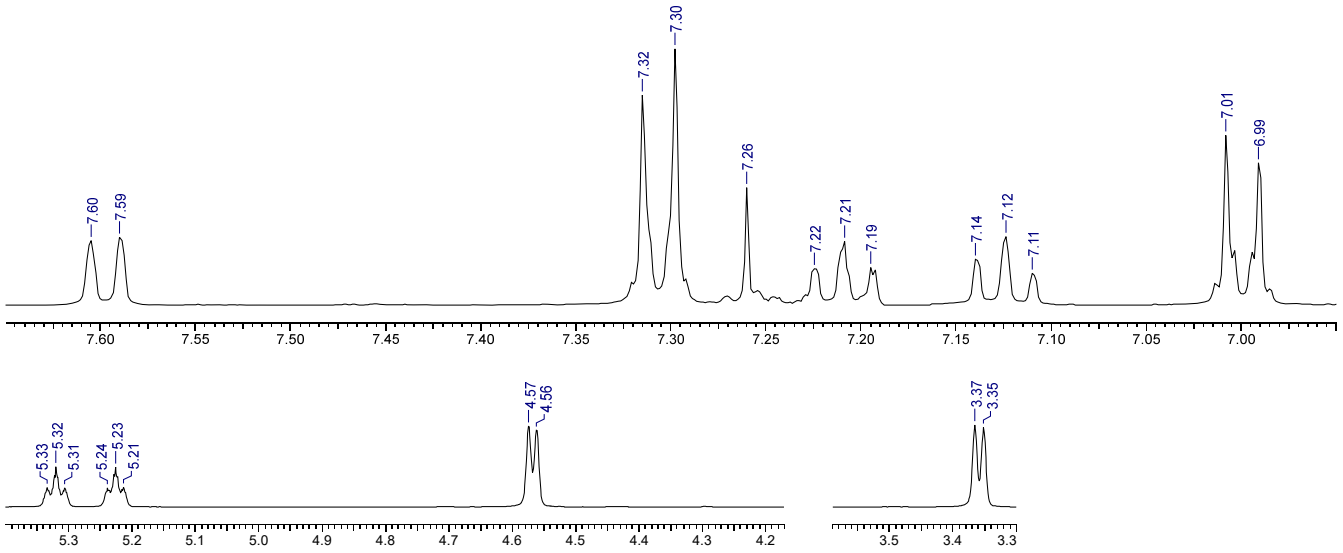
Enlarged view



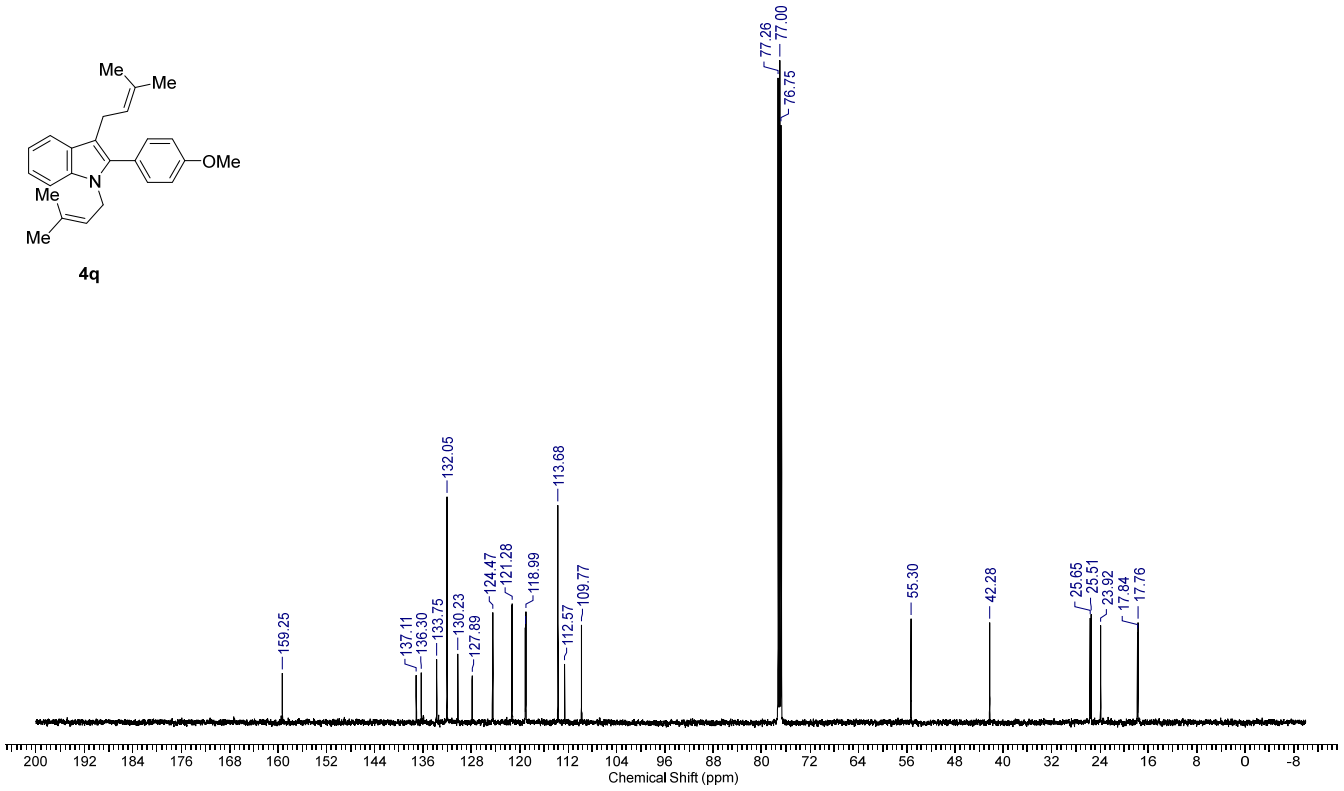
^{1}H NMR (500 MHz, $CDCl_3$) of **4q**



Enlarged view



^{13}C NMR (125 MHz, $CDCl_3$) of **4q**



Enlarged view