

Supporting Information

Metal-free dearomatization reactions of naphthol-ynamides for divergent and enantioselective synthesis of azaspirocycles

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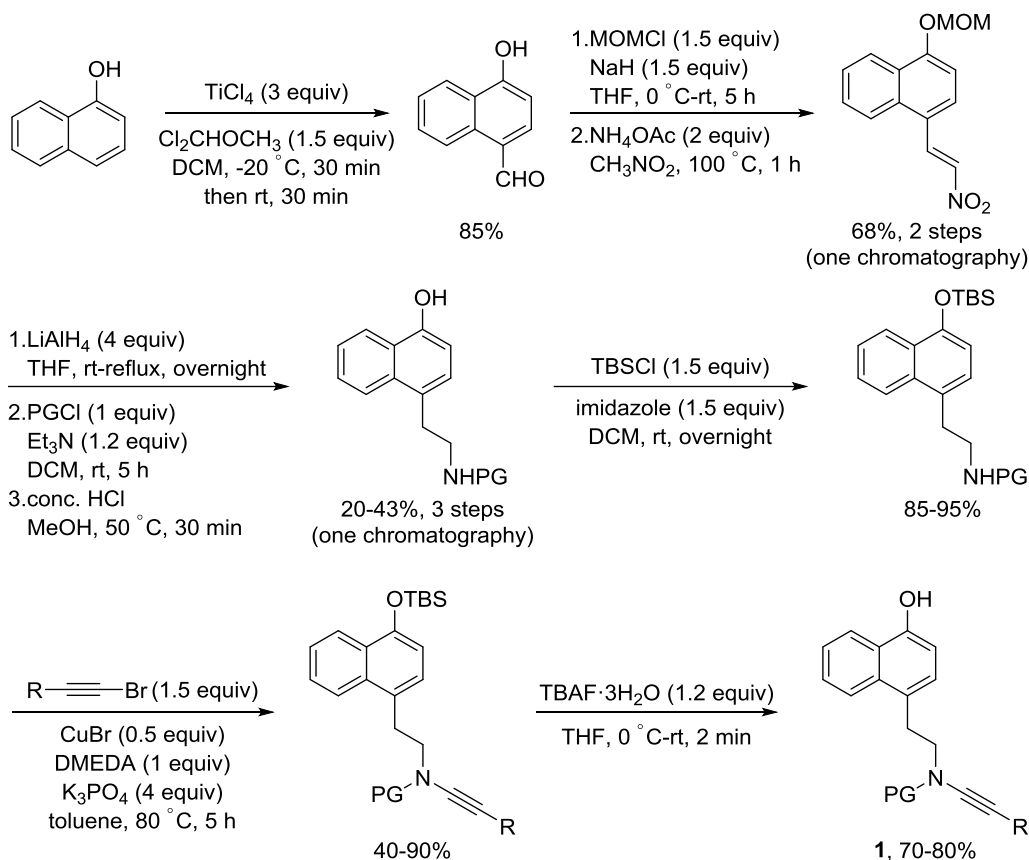
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General Information. Ethyl acetate (ACS grade), hexanes (ACS grade) and anhydrous 1,2-dichloroethane (ACS grade) were obtained commercially and used without further purification. Methylene chloride, tetrahydrofuran and diethyl ether were purified according to standard methods unless otherwise noted. Commercially available reagents were used without further purification. Reactions were monitored by thin layer chromatography (TLC) using silicycle pre-coated silica gel plates. Flash column chromatography was performed over silica gel (300-400 mesh). Infrared spectra were recorded on a Nicolet AVATER FTIR330 spectrometer as thin film and are reported in reciprocal centimeter (cm^{-1}). Mass spectra were recorded with Micromass QTOF2 Quadrupole/Time-of-Flight Tandem mass spectrometer using electron spray ionization.

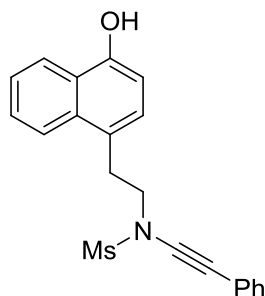
^1H NMR spectra were recorded on a Bruker AV-400 spectrometer and a Bruker AV-500 spectrometer in chloroform- d_3 . Chemical shifts are reported in ppm with the internal TMS signal at 0.0 ppm as a standard. The data is being reported as (s = singlet, d = doublet, t = triplet, m = multiplet or unresolved, brs = broad singlet, coupling constant(s) in Hz, integration).

^{13}C NMR spectra were recorded on a Bruker AV-400 spectrometer and a Bruker AV-500 spectrometer in chloroform- d_3 . Chemical shifts are reported in ppm with the internal chloroform signal at 77.0 ppm as a standard.

Representative synthetic procedures for the preparation of ynamides 1a-1o^{1,2,3}



N-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(phenylethynyl)methanesulfonamide (1a)

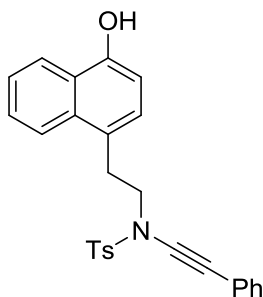


1a

Colorless oil. ¹H NMR (400 MHz, CDCl₃) δ 8.24 (d, *J* = 8.4 Hz, 1H), 8.02 (d, *J* = 8.4 Hz, 1H), 7.57 – 7.52 (m, 1H), 7.50 – 7.46 (m, 1H), 7.46 – 7.42 (m, 2H), 7.33 – 7.30 (m, 3H), 7.23 (s, 1H), 6.74 (d, *J* = 7.6 Hz, 1H), 5.96 (s, 1H), 3.86 (t, *J* = 7.6 Hz, 2H), 3.46 (t, *J* = 7.6 Hz, 2H), 2.79 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 151.2, 132.7, 131.5, 128.3, 128.1, 127.5, 126.9, 125.3, 125.0, 124.8, 123.1, 122.7, 122.4, 108.0, 81.3, 71.7, 52.4, 38.5,

31.6; IR (neat): 2955, 2930, 2858, 2236, 1585, 1462, 1361, 1164, 959, 841; HRESIMS Calcd for $[C_{21}H_{19}NNaO_3S]^+$ ($M + Na^+$) 388.0978, found 388.0983.

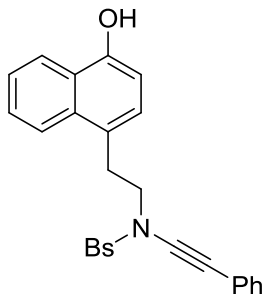
***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-4-methyl-*N*-(phenylethynyl)benzenesulfonamide (1b)**



1b

Colorless oil. 1H NMR (500 MHz, $CDCl_3$) δ 8.22 (d, $J = 8.0$ Hz, 1H), 7.95 (d, $J = 8.0$ Hz, 1H), 7.72 (d, $J = 8.5$ Hz, 2H), 7.54 – 7.46 (m, 2H), 7.42 – 7.39 (m, 2H), 7.35 – 7.28 (m, 3H), 7.23 (d, $J = 8.0$ Hz, 2H), 7.15 (d, $J = 8.0$ Hz, 1H), 6.69 (d, $J = 7.5$ Hz, 1H), 5.52 (s, 1H), 3.72 (t, $J = 8.0$ Hz, 2H), 3.40 (t, $J = 8.0$ Hz, 2H), 2.40 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 151.0, 144.6, 134.5, 132.7, 131.4, 129.7, 128.3, 127.8, 127.5, 127.2, 126.8, 125.6, 124.9, 124.8, 123.2, 122.8, 122.5, 108.1, 82.3, 71.3, 52.3, 31.5, 21.6; IR (neat): 2929, 2857, 2236, 1584, 1462, 1391, 1171, 1069, 755, 691; HRESIMS Calcd for $[C_{27}H_{23}NNaO_3S]^+$ ($M + Na^+$) 464.1291, found 464.1297.

4-bromo-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(phenylethynyl)benzenesulfonamide (1c)

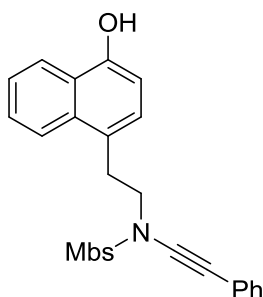


1c

Colorless oil. 1H NMR (400 MHz, $CDCl_3$) δ 8.23 (d, $J = 7.6$ Hz, 1H), 7.93 (d, $J = 7.6$ Hz, 1H), 7.57 – 7.48 (m, 6H), 7.44 – 7.40 (m, 2H), 7.36 – 7.32 (m, 3H), 7.12 (d, $J = 7.6$ Hz,

1H), 6.64 (d, $J = 7.6$ Hz, 1H), 5.58 (s, 1H), 3.79 (t, $J = 7.6$ Hz, 2H), 3.39 (t, $J = 7.6$ Hz, 2H); ^{13}C NMR (100 MHz, CDCl_3) δ 151.1, 136.5, 132.6, 132.3, 131.5, 128.7, 128.6, 128.4, 128.1, 127.3, 126.9, 125.3, 125.0, 124.8, 123.0, 122.6, 122.5, 108.0, 81.6, 71.7, 52.4, 31.3; IR (neat): 2995, 2926, 2236, 1587, 1369, 1247, 1170, 1068, 755, 618; HRESIMS Calcd for $[\text{C}_{26}\text{H}_{20}\text{BrNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 528.0239, found 528.0244.

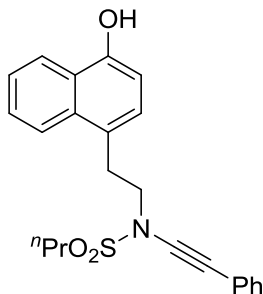
***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-4-methoxy-*N*-(phenylethynyl)benzenesulfonamide (1d)**



1d

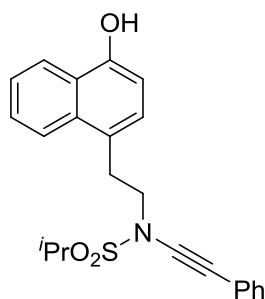
Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.21 (d, $J = 8.5$ Hz, 1H), 7.94 (d, $J = 8.5$ Hz, 1H), 7.72 (d, $J = 8.5$ Hz, 2H), 7.53 – 7.45 (m, 2H), 7.42 – 7.37 (m, 2H), 7.33 – 7.28 (m, 3H), 7.14 (d, $J = 7.5$ Hz, 1H), 6.86 (d, $J = 8.5$ Hz, 2H), 6.68 (d, $J = 7.5$ Hz, 1H), 5.61 (s, 1H), 3.83 (s, 3H), 3.73 (d, $J = 8.0$ Hz, 2H), 3.39 (d, $J = 8.0$ Hz, 2H); ^{13}C NMR (125 MHz, CDCl_3) δ 163.5, 151.0, 132.7, 131.3, 129.6, 129.0, 128.3, 127.8, 127.3, 126.8, 125.6, 124.9, 124.8, 123.1, 122.9, 122.5, 114.2, 108.1, 82.4, 71.4, 55.6, 52.2, 31.4; IR (neat): 3087, 2925, 2236, 1587, 1364, 1169, 1089, 821, 756, 594; HRESIMS Calcd for $[\text{C}_{27}\text{H}_{23}\text{NNaO}_4\text{S}]^+$ ($\text{M} + \text{Na}^+$) 480.1240, found 480.1243.

***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(phenylethynyl)propane-1-sulfonamide (1e)**



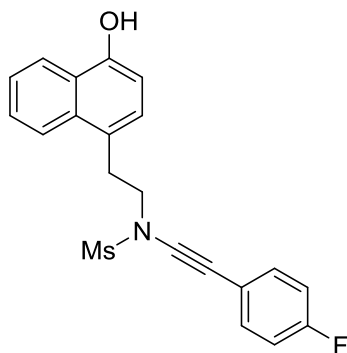
1e

Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.25 (d, $J = 8.0$ Hz, 1H), 8.05 (d, $J = 8.5$ Hz, 1H), 7.56 (t, $J = 7.5$ Hz, 1H), 7.50 (t, $J = 7.5$ Hz, 1H), 7.46 – 7.41 (m, 2H), 7.35 – 7.29 (m, 3H), 7.28 – 7.22 (m, 1H), 6.76 (d, $J = 7.5$ Hz, 1H), 5.56 (s, 1H), 3.86 (t, $J = 7.5$ Hz, 2H), 3.49 (t, $J = 7.5$ Hz, 2H), 2.96 (t, $J = 8.0$ Hz, 2H), 1.83 – 1.74 (m, 2H), 0.96 (t, $J = 7.5$ Hz, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 151.1, 132.8, 131.4, 128.3, 127.9, 127.4, 127.0, 125.6, 125.1, 124.8, 123.2, 122.7, 122.6, 108.1, 81.9, 71.2, 53.8, 52.5, 32.0, 16.9, 12.8; IR (neat): 2928, 2235, 1597, 1515, 1442, 1359, 1166, 1089, 755, 586; HRESIMS Calcd for $[\text{C}_{23}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 416.1291, found 416.1298.

***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(phenylethynyl)propane-2-sulfonamide (1f)****1f**

Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.24 (d, $J = 8.0$ Hz, 1H), 8.06 (d, $J = 8.5$ Hz, 1H), 7.56 (t, $J = 7.5$ Hz, 1H), 7.49 (t, $J = 7.5$ Hz, 1H), 7.44 – 7.41 (m, 2H), 7.34 – 7.28 (m, 3H), 7.23 (d, $J = 7.5$ Hz, 1H), 6.75 (d, $J = 7.5$ Hz, 1H), 5.72 (s, 1H), 3.84 (t, $J = 8.0$ Hz, 2H), 3.51 (t, $J = 8.0$ Hz, 2H), 3.48 – 3.42 (m, 1H), 1.40 (s, 3H), 1.38 (s, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 151.1, 132.8, 131.3, 128.3, 127.9, 127.3, 126.9, 125.5, 125.0, 124.9, 123.3, 122.8, 122.6, 108.1, 82.5, 70.6, 54.6, 53.3, 32.4, 16.5; IR (neat): 3065, 2926, 2236, 1587, 1367, 1248, 1170, 1068, 755, 594; HRESIMS Calcd for $[\text{C}_{23}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 416.1291, found 416.1294.

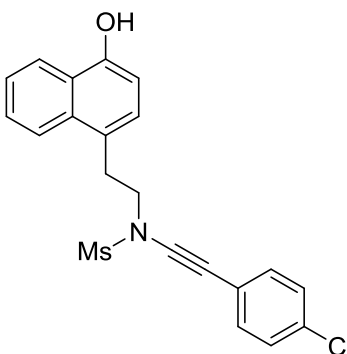
***N*-((4-fluorophenyl)ethynyl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1g)**



1g

Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.24 (d, $J = 8.0$ Hz, 1H), 8.03 (d, $J = 8.5$ Hz, 1H), 7.59 – 7.54 (m, 1H), 7.53 – 7.49 (m, 1H), 7.44 – 7.40 (m, 2H), 7.30 – 7.23 (m, 1H), 7.05 – 7.00 (m, 2H), 6.76 (d, $J = 7.5$ Hz, 1H), 5.45 (s, 1H), 3.88 (t, $J = 7.5$ Hz, 2H), 3.48 (d, $J = 7.5$ Hz, 2H), 2.82 (s, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 162.5 (d, $J = 247.5$ Hz), 151.1, 133.6 (d, $J = 7.5$ Hz), 132.7, 127.4, 127.0, 125.6, 125.2, 124.8, 123.2, 122.6, 118.5 (d, $J = 3.8$ Hz), 115.6 (d, $J = 22.5$ Hz), 108.1, 81.0, 70.5, 52.4, 38.6, 31.7; ^{19}F NMR (376 MHz, CDCl_3) δ -110.90 – -111.10 (m); IR (neat): 2935, 2239, 1509, 1355, 1228, 1157, 1114, 960, 765, 549; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{FNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 406.0884, found 406.0881.

***N*-((4-chlorophenyl)ethynyl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1h)**

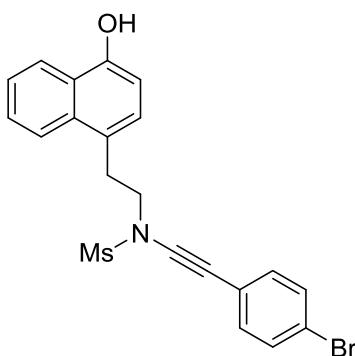


1h

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.25 (d, $J = 8.4$ Hz, 1H), 8.02 (d, $J = 8.4$ Hz, 1H), 7.59 – 7.54 (m, 1H), 7.53 – 7.48 (m, 1H), 7.36 – 7.28 (m, 4H), 7.27 – 7.22 (m, 1H), 6.77 (d, $J = 7.6$ Hz, 1H), 3.88 (t, $J = 7.6$ Hz, 2H), 3.48 (m, $J = 7.6$, 2H), 2.81 (s, 3H); ^{13}C

NMR (100 MHz, CDCl₃) δ 151.5, 134.0, 132.7(4), 132.6(5), 128.7, 127.5, 127.0, 125.2, 125.1, 125.0, 123.1, 122.8, 121.1, 108.1, 82.4, 70.7, 52.4, 38.7, 31.7; IR (neat): 2931, 2237, 1589, 1491, 1354, 1159, 1089, 959, 827, 763; HRESIMS Calcd for [C₂₁H₁₈ClNNaO₃S]⁺ (M + Na⁺) 422.0588, found 422.0583.

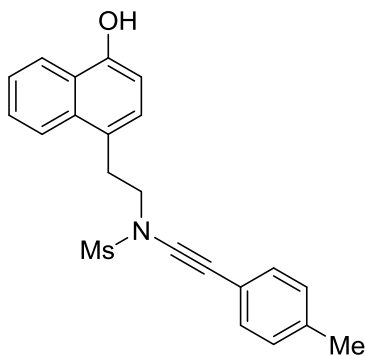
***N*-((4-bromophenyl)ethynyl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1i)**



1i

Brown oil. ¹H NMR (400 MHz, CDCl₃) δ 8.27 (d, *J* = 8.4 Hz, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.58 – 7.53 (m, 1H), 7.52 – 7.47 (m, 1H), 7.47 – 7.43 (m, 2H), 7.30 – 7.20 (m, 3H), 6.78 (d, *J* = 7.6 Hz, 1H), 3.88 (t, *J* = 7.6 Hz, 2H), 3.47 (t, *J* = 7.6 Hz, 2H), 2.79 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 151.6, 132.8, 132.7, 131.6, 127.5, 126.9, 125.1, 125.0, 123.1, 122.8, 122.1, 121.5, 108.1, 82.5, 70.8, 52.4, 38.7, 31.7; IR (neat): 2931, 2237, 1589, 1492, 1359, 1159, 959, 827, 764, 543; HRESIMS Calcd for [C₂₁H₁₈BrNNaO₃S]⁺ (M + Na⁺) 466.0083, found 466.0080.

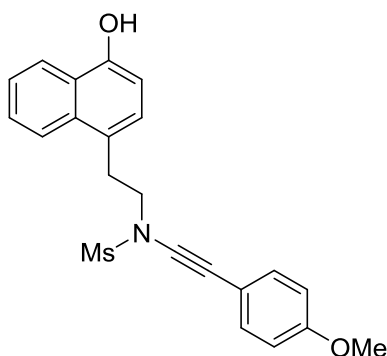
***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(*p*-tolylethynyl)methanesulfonamide (1j)**



1j

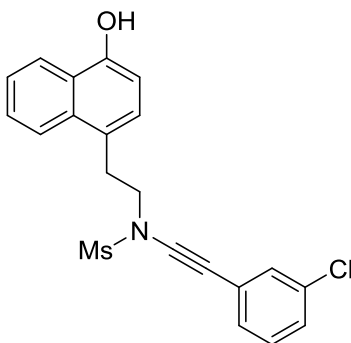
Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.24 (d, $J = 8.0$ Hz, 1H), 8.02 (d, $J = 8.4$ Hz, 1H), 7.57 – 7.52 (m, 1H), 7.51 – 7.46 (m, 1H), 7.35 (d, $J = 8.0$ Hz, 2H), 7.27 – 7.19 (m, 1H), 7.13 (d, $J = 8.0$ Hz, 2H), 6.74 (d, $J = 7.6$ Hz, 1H), 5.81 (s, 1H), 3.86 (t, $J = 7.6$ Hz, 2H), 3.46 (d, $J = 7.6$ Hz, 2H), 2.79 (s, 3H), 2.35 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 151.2, 138.3, 132.7, 131.6, 129.1, 127.4, 126.9, 125.4, 125.1, 124.8, 123.2, 122.6, 119.3, 108.0, 80.6, 71.6, 52.4, 38.4, 31.5, 21.4; IR (neat): 2995, 2928, 2238, 1588, 1355, 1247, 1160, 1053, 959, 767; HRESIMS Calcd for $[\text{C}_{22}\text{H}_{21}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 402.1134, found 402.1137.

***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-((4-methoxyphenyl)ethynyl)methanesulfonamide (1k)**

**1k**

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.25 (d, $J = 8.4$ Hz, 1H), 8.02 (d, $J = 8.4$ Hz, 1H), 7.56 – 7.51 (m, 1H), 7.50 – 7.45 (m, 1H), 7.43 – 7.38 (m, 2H), 7.23 (d, $J = 8.8$ Hz, 1H), 6.89 – 6.83 (m, 2H), 6.75 (d, $J = 7.6$ Hz, 1H), 6.10 (s, 1H), 3.85 (t, $J = 7.6$ Hz, 2H), 3.81 (s, 3H), 3.46 (t, $J = 7.6$ Hz, 2H), 2.80 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 159.7, 151.3, 133.6, 132.7, 127.4, 126.9, 125.4, 125.0, 124.9, 123.2, 122.7, 114.4, 114.0, 108.0, 79.9, 71.3, 55.3, 52.4, 38.4, 31.5; IR (neat): 2955, 2930, 2236, 1585, 1512, 1360, 1269, 1164, 959; HRESIMS Calcd for $[\text{C}_{22}\text{H}_{21}\text{NNaO}_4\text{S}]^+$ ($\text{M} + \text{Na}^+$) 418.1083, found 418.1088.

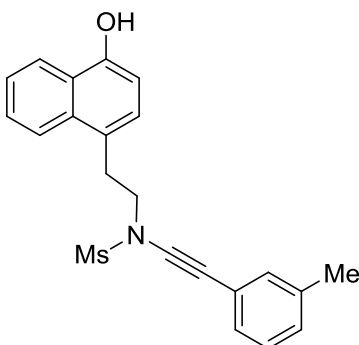
***N*-((3-chlorophenyl)ethynyl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1l)**



1l

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.24 (d, J = 8.0 Hz, 1H), 7.99 (d, J = 8.4 Hz, 1H), 7.55 (t, J = 7.6 Hz, 1H), 7.49 (t, J = 7.6 Hz, 1H), 7.38 (s, 1H), 7.28 – 7.19 (m, 4H), 6.74 (d, J = 7.6 Hz, 1H), 5.95 (s, 1H), 3.87 (t, J = 7.6 Hz, 2H), 3.45 (t, J = 7.6 Hz, 2H), 2.79 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 151.3, 134.1, 132.6, 131.0, 129.5, 129.3, 128.2, 127.5, 127.0, 125.1(4), 125.0(8), 124.8, 124.2, 123.0, 122.7, 108.0, 82.5, 70.6, 52.3, 38.7, 31.6; IR (neat): 3068, 2929, 2237, 1589, 1409, 1354, 1159, 1089, 960, 765; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{ClNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 422.0588, found 422.0585.

***N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)-*N*-(*m*-tolylethynyl)methanesulfonamide (1m)**

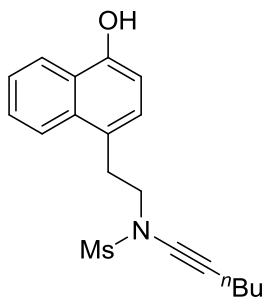


1m

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.24 (d, J = 8.4 Hz, 1H), 8.01 (d, J = 8.4 Hz, 1H), 7.56 – 7.51 (m, 1H), 7.49 – 7.44 (m, 1H), 7.31 – 7.23 (m, 2H), 7.22 – 7.17 (m, 2H), 7.11 (d, J = 7.6 Hz, 1H), 6.73 (d, J = 7.6 Hz, 1H), 6.03 (s, 1H), 3.85 (t, J = 7.6 Hz, 2H), 3.45 (d, J = 7.6 Hz, 2H), 2.78 (s, 3H), 2.31 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 151.3, 138.0, 132.7, 132.0, 129.0, 128.5, 128.2, 127.5, 126.9, 125.2, 125.0, 124.8, 123.1, 122.7, 122.2, 108.0, 80.9, 71.8, 52.3, 38.4, 31.5, 21.1; IR (neat): 2995, 2928, 2238, 1587, 1355,

1247, 1160, 1053, 959, 767; HRESIMS Calcd for $[\text{C}_{22}\text{H}_{21}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 402.1134, found 402.1132.

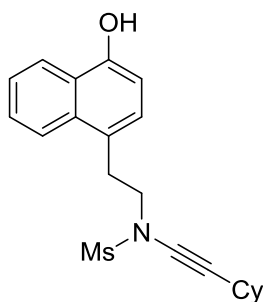
***N*-(hex-1-yn-1-yl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1n)**



1n

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.24 (d, $J = 8.0$ Hz, 1H), 7.99 (d, $J = 8.0$ Hz, 1H), 7.57 – 7.52 (m, 1H), 7.49 (t, $J = 7.2$ Hz, 1H), 7.21 (d, $J = 7.6$ Hz, 1H), 6.75 (d, $J = 7.6$ Hz, 1H), 5.78 (s, 1H), 3.73 (t, $J = 7.6$ Hz, 2H), 3.40 (t, $J = 7.6$ Hz, 2H), 2.79 (s, 3H), 2.36 (t, $J = 7.2$ Hz, 2H), 1.60 – 1.52 (m, 2H), 1.51 – 1.41 (m, 2H), 0.95 (t, $J = 7.2$ Hz, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 151.1, 132.7, 127.3, 126.8, 125.6, 125.0, 124.8, 123.2, 122.6, 108.0, 72.3, 71.4, 52.2, 37.9, 31.5, 31.0, 22.0, 18.2, 13.6; IR (neat): 2928, 2235, 1597, 1515, 1442, 1359, 1166, 1089, 964, 755; HRESIMS Calcd for $[\text{C}_{19}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 368.1291, found 368.1295.

***N*-(cyclohexylethynyl)-*N*-(2-(4-hydroxynaphthalen-1-yl)ethyl)methanesulfonamide (1o)**

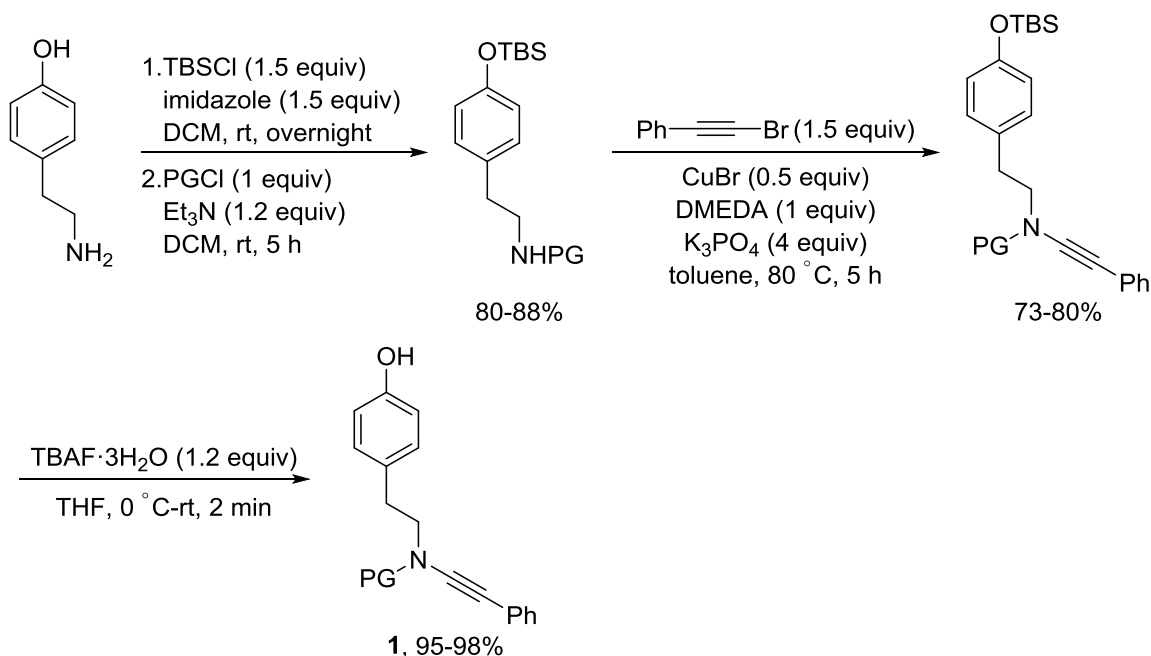


1o

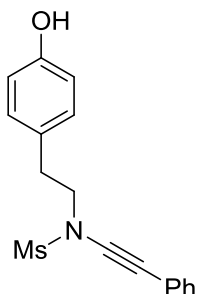
Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.24 (d, $J = 8.0$ Hz, 1H), 8.01 (d, $J = 8.5$ Hz, 1H), 7.55 (t, $J = 7.5$ Hz, 1H), 7.49 (t, $J = 7.5$ Hz, 1H), 7.21 (d, $J = 7.5$ Hz, 1H), 6.75 (d, J

= 7.5 Hz, 1H), 5.75 (s, 1H), 3.73 (t, J = 7.5 Hz, 2H), 3.40 (t, J = 7.5 Hz, 2H), 2.79 (s, 3H), 2.63 – 2.52 (m, 1H), 1.91 – 1.83 (m, 2H), 1.78 – 1.67 (m, 2H), 1.59 – 1.44 (m, 3H), 1.43 – 1.29 (m, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 151.1, 132.7, 127.3, 126.8, 125.6, 125.0, 124.8, 123.2, 122.6, 108.0, 75.4, 72.7, 52.2, 37.8, 32.9, 31.4, 28.9, 25.9, 24.8; IR (neat): 2929, 2849, 2255, 1541, 1481, 1351, 1158, 1120, 1043, 961; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{25}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 394.1447, found 394.1453.

Representative synthetic procedures for the preparation of ynamides 1p and 1q.



N-(4-hydroxyphenethyl)-*N*-(phenylethynyl)methanesulfonamide (1p)

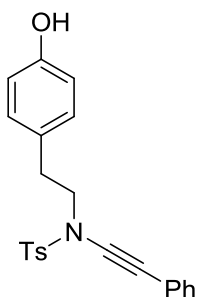


1p

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 7.44 – 7.40 (m, 2H), 7.34 – 7.28 (m, 3H), 7.13 – 7.09 (m, 2H), 6.81 – 6.76 (m, 2H), 5.92 (s, 1H), 3.76 (t, J = 7.2 Hz, 2H), 2.98 (t, J

= 7.2 Hz, 2H), 2.72 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 154.8, 131.4, 130.2, 129.3, 128.3, 128.1, 122.3, 115.5, 80.8, 71.9, 52.8, 38.3, 33.4; IR (neat): 3063, 2929, 2235, 1597, 1515, 1443, 1360, 1165, 963, 755; HRESIMS Calcd for $[\text{C}_{17}\text{H}_{17}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 338.0821, found 338.0824.

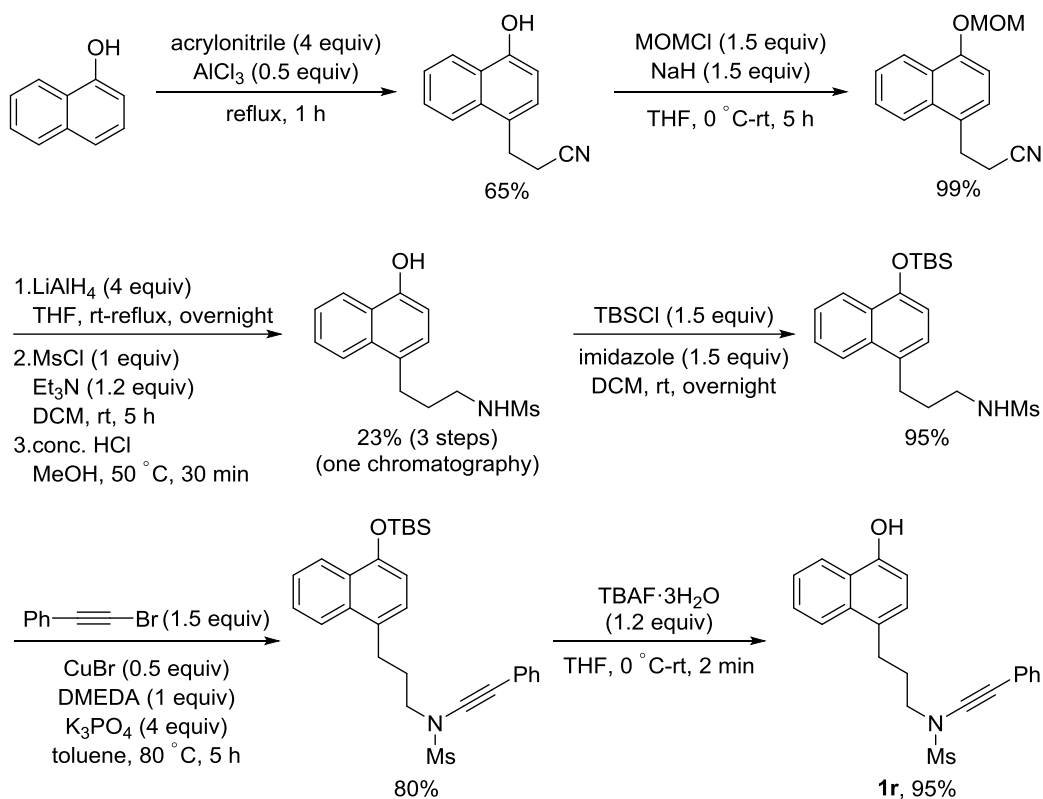
***N*-(4-hydroxyphenethyl)-4-methyl-*N*-(phenylethynyl)benzenesulfonamide (1q)**



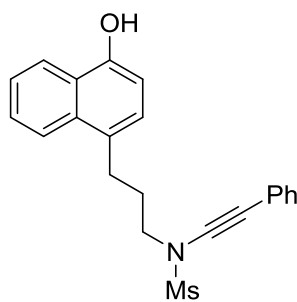
1q

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 7.75 (d, J = 8.4 Hz, 2H), 7.38 – 7.35 (m, 2H), 7.31 – 7.27 (m, 5H), 7.02 (d, J = 8.4 Hz, 2H), 6.73 (d, J = 8.4 Hz, 2H), 5.44 (s, 1H), 3.60 (t, J = 7.6 Hz, 2H), 2.96 – 2.85 (t, J = 7.6 Hz, 2H), 2.42 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 154.5, 144.7, 134.4, 131.4, 130.0, 129.7, 129.3, 128.3, 127.8, 127.6, 122.7, 115.4, 82.0, 71.3, 52.9, 33.5, 21.6; IR (neat): 3063, 2927, 2235, 1515, 1443, 1359, 1220, 1166, 1089, 965; HRESIMS Calcd for $[\text{C}_{23}\text{H}_{21}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 414.1134, found 414.1136.

Representative synthetic procedures for the preparation of ynamide 1r²



***N*-(3-(4-hydroxynaphthalen-1-yl)propyl)-*N*-(phenylethynyl)methanesulfonamide (**1r**)**

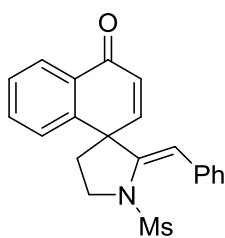


1r

Colorless oil. ^1H NMR (400 MHz, CDCl_3) δ 8.25 – 8.21 (m, 1H), 7.99 – 7.96 (m, 1H), 7.47 – 7.42 (m, 2H), 7.42 – 7.37 (m, 2H), 7.33 – 7.27 (m, 3H), 7.15 (d, J = 7.6 Hz, 1H), 6.76 (d, J = 7.6 Hz, 1H), 5.84 (s, 1H), 3.63 (t, J = 6.8 Hz, 2H), 3.12 (s, 3H), 3.11 (t, J = 8.0 Hz, 2H), 2.24 – 2.16 (m, 2H); ^{13}C NMR (100 MHz, CDCl_3) δ 150.5, 132.6, 131.5, 128.9, 128.3, 128.1, 126.4, 126.0, 124.9, 124.8, 123.5, 122.5, 122.4, 108.1, 81.3, 71.4, 51.3, 38.1, 29.2, 29.1; IR (neat): 2976, 2930, 2238, 1588, 1380, 1353, 1276, 1162, 961, 757; HRESIMS Calcd for $[\text{C}_{22}\text{H}_{21}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 402.1134, found 402.1131.

General procedure for the synthesis of the spirocyclic enones 2. To a mixture of the ynamide **1** (0.2 mmol) in dry MeCN (4 mL) at room temperature, Tf₂NH (0.01 mmol, 2.8 mg) was added. Then, the reaction mixture was stirred at 30 °C and the progress of the reaction was monitored by TLC. The reaction typically took 30 min. Upon completion, the mixture was quenched by Et₃N, then concentrated and the residue was purified by chromatography on silica gel (eluent: DCM) to afford the desired spirocyclic enone **2**.

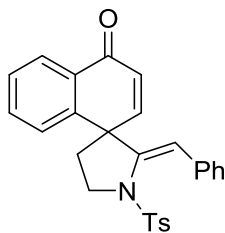
(Z)-2'-benzylidene-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2a)



2a

Compound **2a** was prepared in 82% yield (59.9 mg) according to the general procedure. White solid (mp 197-198 °C). ¹H NMR (400 MHz, CDCl₃) δ 8.21 (d, *J* = 8.0 Hz, 1H), 7.62 – 7.57 (m, 1H), 7.55 – 7.52 (m, 1H), 7.46 – 7.41 (m, 1H), 7.32 – 7.29 (m, 4H), 7.23 – 7.18 (m, 1H), 7.11 (d, *J* = 10.4 Hz, 1H), 6.45 (d, *J* = 10.4 Hz, 1H), 5.69 (s, 1H), 4.16 – 4.09 (m, 1H), 3.97 – 3.90 (m, 1H), 2.72 (s, 3H), 2.60 – 2.52 (m, 1H), 2.49 – 2.42 (m, 1H); ¹³C NMR (100 MHz, CDCl₃) δ 184.5, 150.0, 146.3, 142.7, 136.6, 133.2, 131.6, 128.4, 128.3, 128.0, 127.6, 127.4, 126.6, 125.8, 120.8, 51.7, 48.6, 41.8, 39.9; IR (neat): 3028, 2930, 1662(s), 1599, 1456, 1335, 1303, 1150, 965, 771; HRESIMS Calcd for [C₂₁H₁₉NNaO₃S]⁺ (M + Na⁺) 388.0978, found 388.0983.

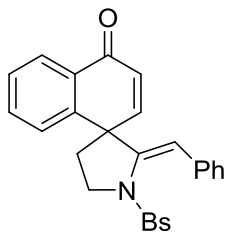
(Z)-2'-benzylidene-1'-tosyl-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2b)



2b

Compound **2b** was prepared in 78% yield (68.8 mg) according to the general procedure. White solid (mp 185-186 °C). ¹H NMR (500 MHz, CDCl₃) δ 8.17 (d, *J* = 8.0 Hz, 1H), 7.60 (d, *J* = 8.0 Hz, 2H), 7.53 (t, *J* = 7.5 Hz, 1H), 7.43 (d, *J* = 8.0 Hz, 1H), 7.40 (t, *J* = 7.5 Hz, 1H), 7.34 – 7.27 (m, 4H), 7.21 – 7.13 (m, 3H), 6.56 (d, *J* = 10.0 Hz, 1H), 6.28 (d, *J* = 10.0 Hz, 1H), 5.58 (s, 1H), 4.09 – 4.02 (m, 1H), 4.01 – 3.95 (m, 1H), 2.46 (s, 3H), 2.44 – 2.39 (m, 1H), 2.27 – 2.20 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 184.4, 149.9, 146.5, 144.4, 141.3, 136.3, 135.4, 133.2, 131.4, 129.6, 128.5, 128.0, 127.9, 127.5, 127.0, 126.6, 125.5, 121.4, 52.4, 49.2, 38.7, 21.6; IR (neat): 3028, 2930, 1662(s), 1599, 1456, 1335, 1150, 965, 771, 567; HRESIMS Calcd for [C₂₇H₂₃NNaO₃S]⁺ (*M* + Na⁺) 464.1291, found 464.1296.

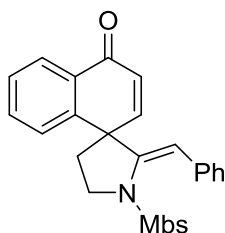
(*Z*)-2'-benzylidene-1'-((4-bromophenyl)sulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2c)



2c

Compound **2c** was prepared in 73% yield (73.7 mg) according to the general procedure. White solid (mp 204-205 °C). ¹H NMR (500 MHz, CDCl₃) δ 8.19 (d, *J* = 7.5 Hz, 1H), 7.60 – 7.54 (m, 3H), 7.46 – 7.40 (m, 4H), 7.19 – 7.13 (m, 5H), 6.78 (d, *J* = 10.0 Hz, 1H), 6.37 (d, *J* = 10.0 Hz, 1H), 5.63 (s, 1H), 4.16 – 4.10 (m, 1H), 4.02 – 3.96 (m, 1H), 2.55 – 2.48 (m, 1H), 2.39 – 2.33 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 184.4, 149.6, 146.3, 141.6, 137.8, 136.1, 133.2, 132.2, 131.5, 129.0, 128.3(8), 128.3(6), 128.0, 127.9, 127.6, 127.1, 126.7, 125.8, 121.6, 52.1, 49.1, 39.1; IR (neat): 2921, 2851, 1661(s), 1573, 1346, 1157, 1089, 1068, 757, 697; HRESIMS Calcd for [C₂₆H₂₀BrNNaO₃S]⁺ (*M* + Na⁺) 528.0239, found 528.0230.

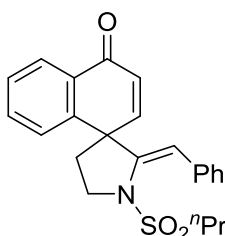
(*Z*)-2'-benzylidene-1'-((4-methoxyphenyl)sulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2d)



2d

Compound **2d** was prepared in 85% yield (77.7 mg) according to the general procedure. White solid (mp 194-195 °C). ^1H NMR (500 MHz, CDCl_3) δ 8.18 (d, J = 8.0 Hz, 1H), 7.65 (d, J = 9.0 Hz, 2H), 7.54 (t, J = 7.5 Hz, 1H), 7.44 (d, J = 8.0 Hz, 1H), 7.40 (t, J = 7.5 Hz, 1H), 7.31 (d, J = 7.0 Hz, 2H), 7.24 – 7.18 (m, 2H), 7.17 – 7.12 (m, 1H), 6.94 (d, J = 9.0 Hz, 2H), 6.62 (d, J = 10.0 Hz, 1H), 6.30 (d, J = 10.0 Hz, 1H), 5.58 (s, 1H), 4.08 – 4.02 (m, 1H), 4.00 – 3.93 (m, 1H), 3.89 (s, 3H), 2.48 – 2.41 (m, 1H), 2.29 – 2.22 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.5, 163.5, 150.0, 146.6, 141.5, 136.3, 133.2, 131.4, 130.1, 129.9, 128.5, 128.0, 127.9, 127.5, 127.1, 126.6, 125.5, 121.5, 114.2, 55.7, 52.5, 49.2, 38.8; IR (neat): 3060, 2926, 1663(s), 1596, 1497, 1302, 1262, 1155, 1091, 835; HRESIMS Calcd for $[\text{C}_{27}\text{H}_{23}\text{NNaO}_4\text{S}]^+$ ($\text{M} + \text{Na}^+$) 480.1240, found 480.1248.

(Z)-2'-benzylidene-1'-(propylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2e)

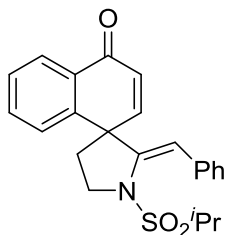


2e

Compound **2e** was prepared in 89% yield (70.0 mg) according to the general procedure. White solid (mp 214-215 °C). ^1H NMR (400 MHz, CDCl_3) δ 8.21 (d, J = 7.6 Hz, 1H), 7.62 – 7.54 (m, 2H), 7.46 – 7.41 (m, 1H), 7.32 – 7.26 (m, 4H), 7.24 – 7.17 (m, 1H), 7.11 (d, J = 10.0 Hz, 1H), 6.45 (d, J = 10.0 Hz, 1H), 5.70 (s, 1H), 4.15 – 4.08 (m, 1H), 3.94 – 3.87 (m, 1H), 2.58 – 2.49 (m, 2H), 2.47 – 2.38 (m, 2H), 1.78 – 1.68 (m, 2H), 0.90 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 184.6, 150.2, 146.4, 143.6, 136.8, 133.2, 131.6, 128.5, 128.2, 128.0, 127.5, 127.3, 126.6, 125.7, 120.5, 56.1, 51.6, 49.2, 40.1, 16.9,

12.6; IR (neat): 2921, 2851, 1660(s), 1456, 1332, 1139, 1095, 767, 696, 586; HRESIMS Calcd for $[C_{23}H_{23}NNaO_3S]^+$ ($M + Na^+$) 416.1291, found 416.1297.

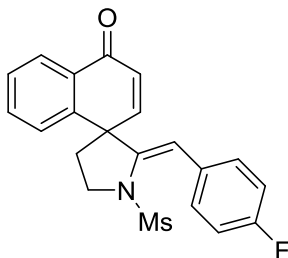
(Z)-2'-(benzylidene-1'-(isopropylsulfonyl))-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2f)



2f

Compound **2f** was prepared in 76% yield (59.8 mg) according to the general procedure. White solid (mp 209-210 °C). 1H NMR (400 MHz, $CDCl_3$) δ 8.20 (d, $J = 7.6$ Hz, 1H), 7.63 – 7.56 (m, 2H), 7.46 – 7.41 (m, 1H), 7.31 – 7.18 (m, 5H), 7.12 (d, $J = 10.0$ Hz, 1H), 6.45 (d, $J = 10.0$ Hz, 1H), 5.71 (s, 1H), 4.15 – 4.07 (m, 1H), 3.94 – 3.87 (m, 1H), 2.58 – 2.50 (m, 1H), 2.47 – 2.39 (m, 1H), 2.23 – 2.15 (m, 1H), 1.23 (d, $J = 6.8$ Hz, 3H), 1.18 (d, $J = 6.8$ Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 184.6, 150.3, 146.5, 144.2, 136.9, 133.1, 131.6, 128.5, 128.2, 128.1, 127.5, 127.2, 126.6, 125.6, 120.5, 55.2, 51.5, 49.9, 40.4, 16.4, 15.9; IR (neat): 2926, 2853, 1661(s), 1455, 1334, 1137, 758, 699, 583, 538; HRESIMS Calcd for $[C_{23}H_{23}NNaO_3S]^+$ ($M + Na^+$) 416.1291, found 416.1297.

(Z)-2'-(4-fluorobenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2g)

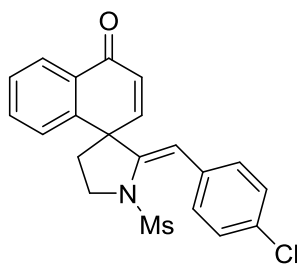


2g

Compound **2g** was prepared in 94% yield (72.0 mg) according to the general procedure. White solid (mp 197-198 °C). 1H NMR (400 MHz, $CDCl_3$) δ 8.21 (d, $J = 8.0$ Hz, 1H),

7.63 – 7.57 (m, 1H), 7.55 – 7.50 (m, 1H), 7.47 – 7.42 (m, 1H), 7.32 – 7.26 (m, 2H), 7.08 (d, $J = 10.4$ Hz, 1H), 7.01 – 6.96 (m, 2H), 6.45 (d, $J = 10.4$ Hz, 1H), 5.63 (s, 1H), 4.13 – 4.05 (m, 1H), 3.98 – 3.91 (m, 1H), 2.79 (s, 3H), 2.61 – 2.52 (m, 1H), 2.50 – 2.42 (m, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ 184.5, 161.7 (d, $J = 247.0$ Hz), 149.9, 146.3, 142.6, 133.3, 132.5 (d, $J = 3.0$ Hz), 131.5, 130.0 (d, $J = 8.0$ Hz), 127.9, 127.6, 126.7, 125.8, 119.8, 115.3 (d, $J = 21.0$ Hz), 51.8, 48.8, 41.8, 39.8; ^{19}F NMR (376 MHz, CDCl_3) δ -113.40 – -113.60 (m); IR (neat): 3068, 2929, 1661(s), 2236, 1508, 1456, 1338, 1225, 1153, 768; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{FNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 406.0884, found 406.0890.

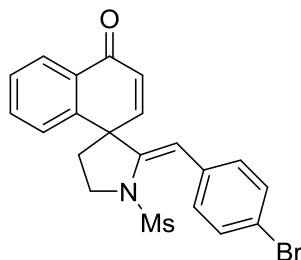
(Z)-2'-(4-chlorobenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2h)



2h

Compound **2h** was prepared in 94% yield (75.0 mg) according to the general procedure. White solid (mp 178-179 °C). ^1H NMR (400 MHz, CDCl_3) δ 8.21 (d, $J = 7.6$ Hz, 1H), 7.62 – 7.57 (m, 1H), 7.52 – 7.49 (m, 1H), 7.47 – 7.42 (m, 1H), 7.28 – 7.22 (m, 4H), 7.07 (d, $J = 10.0$ Hz, 1H), 6.45 (d, $J = 10.0$ Hz, 1H), 5.61 (s, 1H), 4.13 – 4.05 (m, 1H), 3.99 – 3.92 (m, 1H), 2.83 (s, 3H), 2.61 – 2.53 (m, 1H), 2.50 – 2.42 (m, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ 184.4, 149.7, 146.2, 143.0, 135.0, 133.3, 133.0, 131.5, 129.6, 128.5, 127.9, 127.7, 126.7, 125.9, 119.6, 51.8, 48.8, 41.8, 39.8; IR (neat): 3061, 2932, 1662(s), 1490, 1336, 1152, 1071, 967, 840, 768; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{ClNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 422.0588, found 422.0594.

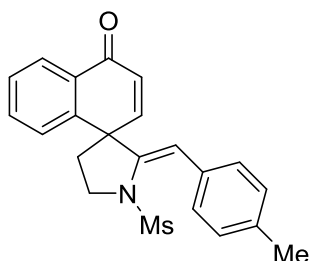
(Z)-2'-(4-bromobenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2i)



2i

Compound **2i** was prepared in 94% yield (83.3 mg) according to the general procedure. White solid (mp 192-193 °C). ^1H NMR (500 MHz, CDCl_3) δ 8.21 (d, J = 8.0 Hz, 1H), 7.62 – 7.57 (m, 1H), 7.50 (d, J = 7.5 Hz, 1H), 7.47 – 7.42 (m, 1H), 7.42 – 7.39 (m, 2H), 7.18 (d, J = 8.5 Hz, 2H), 7.07 (d, J = 10.0 Hz, 1H), 6.45 (d, J = 10.0 Hz, 1H), 5.58 (s, 1H), 4.11 – 4.05 (m, 1H), 3.98 – 3.92 (m, 1H), 2.83 (s, 3H), 2.60 – 2.53 (m, 1H), 2.49 – 2.43 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.4, 149.7, 146.1, 143.0, 135.5, 133.3, 131.5, 131.4, 129.9, 127.9, 127.7, 126.7, 125.9, 121.1, 119.6, 51.9, 48.8, 41.8, 39.7; IR (neat): 2954, 2926, 1662(s), 1486, 1336, 1152, 1071, 966, 768, 735; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{BrNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 466.0083, found 466.0074.

(Z)-2'-(4-methylbenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2j)

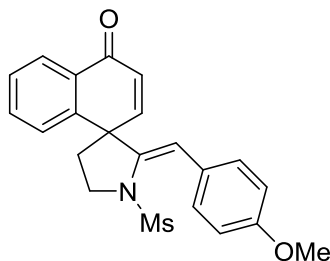


2j

Compound **2j** was prepared in 65% yield (49.3 mg) according to the general procedure. White solid (mp 209-210 °C). ^1H NMR (400 MHz, CDCl_3) δ 8.20 (d, J = 8.0 Hz, 1H), 7.61 – 7.55 (m, 1H), 7.54 – 7.51 (m, 1H), 7.45 – 7.40 (m, 1H), 7.21 (d, J = 8.0 Hz, 2H), 7.13 – 7.08 (m, 3H), 6.44 (d, J = 10.0 Hz, 1H), 5.65 (s, 1H), 4.15 – 4.08 (m, 1H), 3.96 – 3.89 (m, 1H), 2.78 (s, 3H), 2.58 – 2.50 (m, 1H), 2.49 – 2.41 (m, 1H), 2.30 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ 184.6, 150.3, 146.6, 141.9, 137.3, 133.5, 133.2, 131.5, 129.0, 128.2, 128.0, 127.5, 126.6, 125.6, 121.1, 51.7, 48.6, 41.8, 39.9, 21.2; IR (neat): 3023,

2923, 1661(s), 1456, 1337, 1158, 1085, 960, 768, 559; HRESIMS Calcd for $[C_{22}H_{21}NNaO_3S]^+$ ($M + Na^+$) 402.1134, found 402.1138.

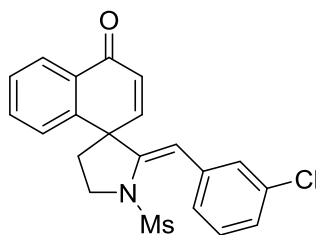
(Z)-2'-(4-methoxybenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2k)



2k

Compound **2k** was prepared in 79% yield (62.4 mg) according to the general procedure. White solid (mp 155-156 °C). 1H NMR (400 MHz, $CDCl_3$) δ 8.20 (d, $J = 8.0$ Hz, 1H), 7.61 – 7.56 (m, 1H), 7.54 – 7.51 (m, 1H), 7.45 – 7.40 (m, 1H), 7.30 – 7.22 (m, 2H), 7.11 (d, $J = 10.4$ Hz, 1H), 6.85 – 6.80 (m, 2H), 6.43 (d, $J = 10.4$ Hz, 1H), 5.63 (s, 1H), 4.15 – 4.07 (m, 1H), 3.96 – 3.89 (m, 1H), 3.78 (s, 3H), 2.81 (s, 3H), 2.58 – 2.50 (m, 1H), 2.49 – 2.42 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 184.6, 158.8, 150.5, 146.8, 141.3, 133.2, 131.5, 129.6, 128.8, 127.9, 127.5, 126.6, 125.6, 121.1, 113.7, 55.2, 51.8, 48.7, 41.8, 40.0; IR (neat): 3032, 2929, 1661(s), 1510, 1456, 1333, 1247, 1149, 1031, 767; HRESIMS Calcd for $[C_{22}H_{21}NNaO_4S]^+$ ($M + Na^+$) 418.1083, found 418.1086.

(Z)-2'-(3-chlorobenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2l)

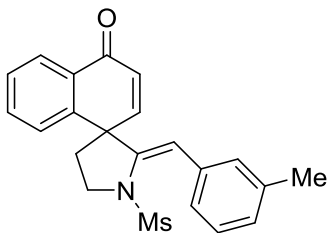


2l

Compound **2l** was prepared in 94% yield (75.0 mg) according to the general procedure. White solid (mp 206-207 °C). 1H NMR (500 MHz, $CDCl_3$) δ 8.21 (d, $J = 8.0$ Hz, 1H),

7.60 (t, $J = 7.5$ Hz, 1H), 7.51 (d, $J = 8.0$ Hz, 1H), 7.47 – 7.40 (m, 2H), 7.22 (t, $J = 7.5$ Hz, 1H), 7.17 (d, $J = 8.0$ Hz, 1H), 7.09 – 7.03 (m, 2H), 6.45 (d, $J = 10.0$ Hz, 1H), 5.59 (s, 1H), 4.12 – 4.06 (m, 1H), 4.00 – 3.95 (m, 1H), 2.89 (s, 3H), 2.62 – 2.55 (m, 1H), 2.49 – 2.43 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.4, 149.5, 146.0, 143.4, 138.4, 133.9, 133.3, 131.5, 129.7, 127.9, 127.6(9), 127.6(5), 127.3, 126.7, 126.6, 125.9, 119.1, 51.8, 48.8, 41.7, 39.7; IR (neat): 2957, 2927, 1661(s), 1595, 1445, 1335, 1149, 1093, 767, 566; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{ClNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 422.0588, found 422.0587.

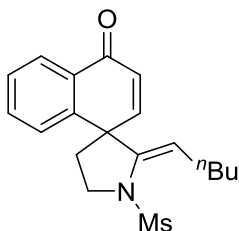
(Z)-2'-(3-methylbenzylidene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2m)



2m

Compound **2m** was prepared in 88% yield (66.7 mg) according to the general procedure. White solid (mp 137-138 °C). ^1H NMR (500 MHz, CDCl_3) δ 8.20 (d, $J = 8.0$ Hz, 1H), 7.61 – 7.56 (m, 1H), 7.54 (d, $J = 8.0$ Hz, 1H), 7.45 – 7.41 (m, 1H), 7.19 (t, $J = 7.5$ Hz, 1H), 7.14 (d, $J = 8.0$ Hz, 1H), 7.12 – 7.06 (m, 2H), 7.01 (d, $J = 7.5$ Hz, 1H), 6.44 (d, $J = 10.0$ Hz, 1H), 5.65 (s, 1H), 4.16 – 4.10 (m, 1H), 3.97 – 3.91 (m, 1H), 2.76 (s, 3H), 2.59 – 2.52 (m, 1H), 2.48 – 2.42 (m, 1H), 2.30 (s, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.5, 150.1, 146.4, 142.4, 137.8, 136.4, 133.2, 131.5, 129.0, 128.1(8), 128.1(6), 128.0, 127.5, 126.6, 125.7, 125.3, 120.9, 51.7, 48.6, 41.8, 39.9, 21.3; IR (neat): 2924, 2854, 1663(s), 1456, 1336, 1151, 965, 841, 768, 541; HRESIMS Calcd for $[\text{C}_{22}\text{H}_{21}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 402.1134, found 402.1139.

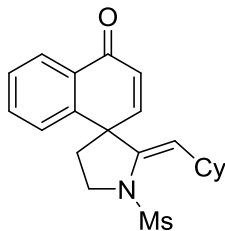
(Z)-1'-(methylsulfonyl)-2'-pentylidene-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2n)



2n

Compound **2n** was prepared in 64% yield (44.2 mg) according to the general procedure. Colorless oil. ^1H NMR (500 MHz, CDCl_3) δ 8.17 (d, $J = 7.5$ Hz, 1H), 7.58 (t, $J = 7.5$ Hz, 1H), 7.46 – 7.39 (m, 2H), 6.97 (d, $J = 10.0$ Hz, 1H), 6.38 (d, $J = 10.0$ Hz, 1H), 4.64 (t, $J = 7.0$ Hz, 1H), 3.97 – 3.90 (m, 1H), 3.89 – 3.83 (m, 1H), 3.19 (s, 3H), 2.53 – 2.46 (m, 1H), 2.41 – 2.34 (m, 1H), 2.15 – 2.10 (m, 2H), 1.26 – 1.16 (m, 4H), 0.84 (t, $J = 6.5$ Hz, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.6, 150.4, 146.7, 140.5, 133.0, 131.5, 127.9, 127.4, 126.5, 125.3, 122.5, 51.0, 48.5, 41.7, 39.9, 31.0, 29.3, 22.3, 13.8; IR (neat): 2956, 2928, 1661(s), 1456, 1334, 1151, 1094, 841, 768, 545; HRESIMS Calcd for $[\text{C}_{19}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 368.1291, found 368.1293.

(Z)-2'-(cyclohexylmethylene)-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-pyrrolidin]-4-one (2o)

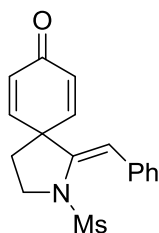


2o

Compound **2o** was prepared in 66% yield (49.0 mg) according to the general procedure. White solid (mp 110-111 °C), ^1H NMR (500 MHz, CDCl_3) δ 8.19 (d, $J = 7.5$ Hz, 1H), 7.60 (t, $J = 8.0$ Hz, 1H), 7.50 – 7.43 (m, 2H), 6.97 (d, $J = 10.5$ Hz, 1H), 6.46 (d, $J = 10.5$ Hz, 1H), 5.38 (d, $J = 10.5$ Hz, 1H), 4.04 – 3.99 (m, 1H), 3.79 – 3.73 (m, 1H), 3.05 (s, 3H), 2.44 – 2.37 (m, 1H), 2.29 – 2.23 (m, 1H), 1.56 – 1.46 (m, 1H), 1.40 – 1.28 (m, 4H), 0.96 – 0.83 (m, 3H), 0.77 – 0.64 (m, 2H), 0.45 – 0.37 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 184.0, 149.2, 146.8, 138.2, 133.2, 130.8, 127.7, 127.4, 126.7, 126.1, 117.2, 51.2, 48.3, 39.6, 36.5, 34.0, 33.6, 31.8, 25.8, 25.5, 25.4; IR (neat): 2925, 2850, 1662(s), 1449, 1337,

1160, 1074, 1024, 960, 768; HRESIMS Calcd for $[C_{21}H_{25}NNaO_3S]^+$ ($M + Na^+$) 394.1447, found 394.14549.

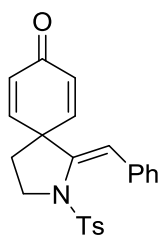
(Z)-1-benzylidene-2-(methylsulfonyl)-2-azaspiro[4.5]deca-6,9-dien-8-one (2p)



2p

Compound **2p** was prepared in 55% yield (34.7 mg) according to the general procedure. White solid (mp 124-125 °C). 1H NMR (400 MHz, $CDCl_3$) δ 7.38 – 7.30 (m, 4H), 7.24 – 7.20 (m, 1H), 6.98 (d, J = 10.0 Hz, 2H), 6.34 (d, J = 10.0 Hz, 2H), 6.00 (s, 1H), 3.91 (t, J = 6.8 Hz, 2H), 2.65 (s, 3H), 2.26 (t, J = 6.8 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 185.6, 149.7, 138.7, 136.0, 128.5, 128.4, 128.1, 127.7, 120.2, 51.0, 48.2, 41.6, 35.1; IR (neat): 2920, 2850, 1662(s), 1488, 1447, 1334, 1148, 1097, 963, 860; HRESIMS Calcd for $[C_{17}H_{17}NNaO_3S]^+$ ($M + Na^+$) 338.0821, found 338.0822.

(Z)-1-benzylidene-2-tosyl-2-azaspiro[4.5]deca-6,9-dien-8-one (2q)

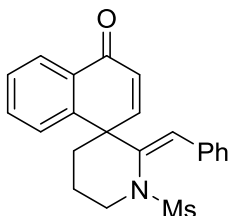


2q

Compound **2q** was prepared in 70% yield (54.8 mg) according to the general procedure. White solid (mp 149-150 °C). 1H NMR (500 MHz, $CDCl_3$) δ 7.62 (d, J = 8.5 Hz, 2H), 7.42 (d, J = 7.5 Hz, 2H), 7.29 (d, J = 8.0 Hz, 2H), 7.23 (d, J = 8.0 Hz, 2H), 7.21 – 7.16 (m, 1H), 6.56 (d, J = 10.0 Hz, 2H), 6.19 (d, J = 10.0 Hz, 2H), 5.88 (s, 1H), 3.88 (t, J = 7.0 Hz, 2H), 2.44 (s, 3H), 1.95 (t, J = 7.0 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 185.5, 149.9, 144.7, 136.5, 135.6, 135.0, 129.7, 128.7, 128.0, 127.9, 127.6, 127.4, 120.9, 51.7,

49.0, 33.9, 21.6; IR (neat): 3053, 2924, 1664(s), 1492, 1447, 1360, 1162, 1089, 859, 751; HRESIMS Calcd for $[C_{23}H_{21}NNaO_3S]^+$ ($M + Na^+$) 414.1134, found 414.1135.

(Z)-2'-benzylidene-1'-(methylsulfonyl)-4H-spiro[naphthalene-1,3'-piperidin]-4-one (2r)

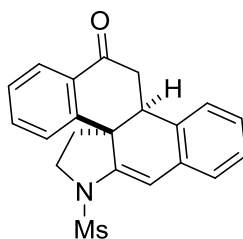


2r

Compound **2r** was prepared in 65% yield (49.3 mg) according to the general procedure. White solid (mp 244-245 °C). 1H NMR (400 MHz, $CDCl_3$) δ 8.27 (d, $J = 7.6$ Hz, 1H), 7.77 – 7.69 (m, 2H), 7.58 (t, $J = 7.2$ Hz, 1H), 7.46 (t, $J = 7.6$ Hz, 1H), 7.35 – 7.28 (m, 2H), 7.24 – 7.18 (m, 1H), 7.16 (d, $J = 7.2$ Hz, 2H), 6.54 (d, $J = 10.4$ Hz, 1H), 6.00 (s, 1H), 4.10 – 4.00 (m, 1H), 3.46 – 3.38 (m, 1H), 2.61 – 2.52 (m, 1H), 2.39 – 2.28 (m, 1H), 2.14 (s, 3H), 1.97 – 1.92 (m, 1H), 1.87 – 1.77 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 184.2, 149.1, 145.6, 139.1, 136.6, 132.1(1), 132.0(8), 130.1, 128.9, 128.5, 128.4, 127.6, 127.5, 127.2, 126.6, 48.5, 46.2, 40.4, 40.3, 22.0; IR (neat): 2930, 2868, 1663(s), 1455, 1329, 1147, 1013, 842, 772, 698; HRESIMS Calcd for $[C_{22}H_{21}NNaO_3S]^+$ ($M + Na^+$) 402.1134, found 402.1137.

General procedure for the synthesis of the azaspiropolycyclic compounds 3. To a mixture of the ynamide **1** (0.2 mmol) in dry DCM (4 mL) at 0 °C, Tf_2NH (0.01 mmol, 2.8 mg) was added. Then, the reaction mixture was stirred at 0 °C and the progress of the reaction was monitored by TLC. The reaction typically took 12-48 h. Upon completion, the mixture was quenched by Et_3N , then concentrated and the residue was purified by chromatography on silica gel (eluent: DCM) to afford the desired azaspiropolycyclic compound **3**.

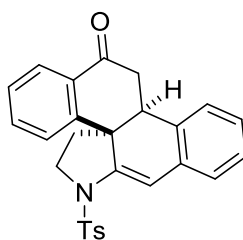
7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3a)



3a

Compound **3a** was prepared in 70% yield (51.1 mg) according to the general procedure. White solid (mp 242-243 °C). ^1H NMR (400 MHz, CDCl_3) δ 7.96 (d, J = 8.0 Hz, 1H), 7.42 (t, J = 7.6 Hz, 1H), 7.30 – 7.20 (m, 2H), 7.14 (d, J = 7.2 Hz, 1H), 7.07 – 6.95 (m, 3H), 6.46 (s, 1H), 4.15 – 4.07 (m, 1H), 3.92 – 3.84 (m, 1H), 3.53 – 3.47 (m, 2H), 3.20 (s, 3H), 3.17 – 3.11 (m, 1H), 2.48 – 2.39 (m, 1H), 2.33 – 2.27 (m, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ 195.1, 145.3, 142.9, 134.3, 134.2, 132.1, 131.6, 127.9, 127.3, 127.1, 126.4, 126.3, 125.8, 124.3, 103.2, 50.0, 48.8, 45.6, 37.8, 37.1, 36.7; IR (neat): 2955, 2923, 2853, 1684(s), 1594, 1342, 1246, 1157, 1088, 961; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{19}\text{NNaO}_3\text{S}]^+$ (M + Na^+) 388.0978, found 388.0981.

7-tosyl-6,7,12b,13-tetrahydrobenzo[f]naphtho[1,2-d]indol-14(5H)-one (3b)

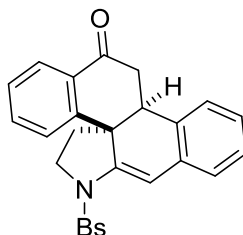


3b

Compound **3b** was prepared in 75% yield (66.2 mg) according to the general procedure. White solid (mp 265-266 °C). ^1H NMR (400 MHz, DMSO) δ 8.02 (d, J = 8.4 Hz, 2H), 7.70 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 8.0 Hz, 2H), 7.21 – 7.16 (m, 1H), 7.09 – 7.05 (m, 1H), 7.04 – 6.98 (m, 2H), 6.97 – 6.88 (m, 2H), 6.56 (s, 1H), 5.89 (d, J = 7.6 Hz, 1H), 4.15 – 4.10 (m, 1H), 3.62 – 3.54 (m, 1H), 3.41 – 3.36 (m, 1H), 3.32 – 3.26 (m, 1H), 3.24 – 3.17 (m, 1H), 2.48 (s, 3H), 2.38 – 2.29 (m, 1H), 2.25 – 2.19 (m, 1H); ^{13}C NMR (125 MHz, DMSO) δ 194.9, 145.6, 145.1, 143.6, 134.4, 133.9, 133.4, 132.9, 131.4, 130.3, 127.4, 127.3, 126.9, 126.2, 126.0, 125.8, 125.0, 124.2, 102.9, 49.5, 48.7, 44.5, 36.3, 36.0,

21.1; IR (neat): 2959, 2920, 2850, 1678(s), 1594, 1349, 1245, 1165, 1087, 805; HRESIMS Calcd for $[C_{27}H_{23}NNaO_3S]^+$ ($M + Na^+$) 464.1291, found 464.1299.

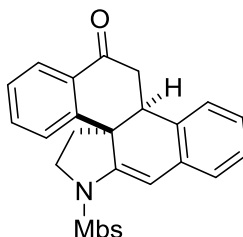
7-((4-bromophenyl)sulfonyl)-6,7,12b,13-tetrahydrobenzo[f]naphtho[1,2-d]indol-14(5H)-one (3c)



3c

Compound **3c** was prepared in 58% yield (58.6 mg) according to the general procedure. White solid (mp 260-261 °C). 1H NMR (400 MHz, DMSO) δ 8.07 (d, J = 8.8 Hz, 2H), 7.98 (d, J = 8.4 Hz, 2H), 7.72 (d, J = 8.0 Hz, 1H), 7.23 (t, J = 7.6 Hz, 1H), 7.09 – 6.94 (m, 5H), 6.57 (s, 1H), 6.02 (d, J = 7.6 Hz, 1H), 4.18 – 4.12 (m, 1H), 3.66 – 3.58 (m, 1H), 3.41 – 3.29 (m, 2H), 3.24 – 3.18 (m, 1H), 2.38 – 2.29 (m, 1H), 2.27 – 2.22 (m, 1H); ^{13}C NMR (125 MHz, DMSO) δ 194.9, 145.4, 143.5, 135.9, 134.2, 133.2, 133.0, 131.4, 129.2, 128.5, 127.6, 126.9, 126.3, 126.1, 126.0, 125.0, 124.2, 103.3, 49.4, 48.8, 44.5, 36.3, 36.1; IR (neat): 2962, 2923, 2852, 1678(s), 1595, 1350, 1261, 1169, 1088, 803; HRESIMS Calcd for $[C_{26}H_{20}BrNNaO_3S]^+$ ($M + Na^+$) 528.0239, found 528.0236.

7-((4-methoxyphenyl)sulfonyl)-6,7,12b,13-tetrahydrobenzo[f]naphtho[1,2-d]indol-14(5H)-one (3d)

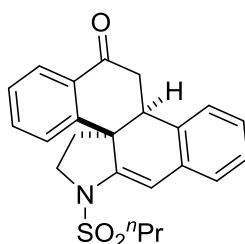


3d

Compound **3d** was prepared in 62% yield (56.7 mg) according to the general procedure. White solid (mp 285-286 °C). 1H NMR (400 MHz, DMSO) δ 8.07 (d, J = 8.8 Hz, 2H), 7.71 (d, J = 8.0 Hz, 1H), 7.30 – 7.23 (m, 2H), 7.19 (t, J = 7.2 Hz, 1H), 7.07 – 6.92 (m,

5H), 6.56 (s, 1H), 5.99 (d, $J = 7.6$ Hz, 1H), 4.14 – 4.08 (m, 1H), 3.90 (s, 3H), 3.62 – 3.54 (m, 1H), 3.42 – 3.27 (m, 2H), 3.24 – 3.17 (m, 1H), 2.37 – 2.28 (m, 1H), 2.25 – 2.19 (m, 1H); ^{13}C NMR (100 MHz, DMSO) δ 194.9, 163.7, 145.8, 143.7, 134.5, 133.4, 132.9, 131.4, 129.6, 128.5, 127.4, 126.9, 126.2, 126.0, 125.7, 125.1, 124.2, 114.9, 102.8, 55.9, 49.5, 48.6, 44.5, 36.3, 36.1; IR (neat): 2972, 2921, 1678(s), 1593, 1350, 1244, 1165, 1092, 908, 595; HRESIMS Calcd for $[\text{C}_{27}\text{H}_{23}\text{NNaO}_4\text{S}]^+$ ($\text{M} + \text{Na}^+$) 480.1240, found 480.1245.

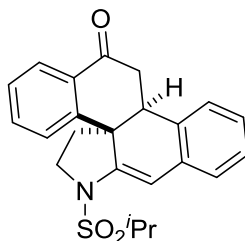
7-(propylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3e)



3e

Compound **3e** was prepared in 61% yield (48.0 mg) according to the general procedure. White solid (mp 174-175 °C). ^1H NMR (500 MHz, CDCl_3) δ 7.96 (d, $J = 7.5$ Hz, 1H), 7.43 (t, $J = 7.0$ Hz, 1H), 7.29 – 7.23 (m, 2H), 7.14 (d, $J = 7.0$ Hz, 1H), 7.08 – 7.03 (m, 1H), 7.02 – 6.95 (m, 2H), 6.40 (s, 1H), 4.12 – 4.06 (m, 1H), 3.94 – 3.87 (m, 1H), 3.53 – 3.48 (m, 2H), 3.36 – 3.32 (m, 2H), 3.17 – 3.12 (m, 1H), 2.47 – 2.40 (m, 1H), 2.33 – 2.28 (m, 1H), 2.08 – 1.93 (m, 2H), 1.15 (t, $J = 7.5$ Hz, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 195.1, 145.5, 142.9, 134.4, 134.3, 132.0, 131.6, 127.9, 127.3, 127.1, 126.3, 126.2, 125.9, 124.3, 102.7, 52.4, 50.1, 49.1, 45.7, 37.9, 36.8, 17.0, 13.1; IR (neat): 2930, 2868, 1663(s), 1455, 1329, 1147, 1013, 842, 772, 698; HRESIMS Calcd for $[\text{C}_{23}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 416.1291, found 416.1283.

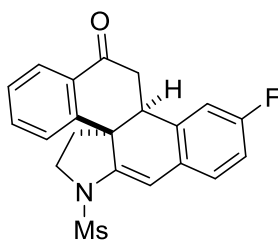
7-(isopropylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3f)



3f

Compound **3f** was prepared in 79% yield (62.1 mg) according to the general procedure. White solid (mp 169-170 °C). ^1H NMR (500 MHz, CDCl_3) δ 7.95 (d, $J = 7.5$ Hz, 1H), 7.42 (t, $J = 7.0$ Hz, 1H), 7.31 (d, $J = 8.0$ Hz, 1H), 7.26 – 7.20 (m, 1H), 7.13 (d, $J = 7.5$ Hz, 1H), 7.05 – 6.93 (m, 3H), 6.36 (s, 1H), 4.13 – 4.06 (m, 1H), 3.95 – 3.89 (m, 1H), 3.76 – 3.70 (m, 1H), 3.52 – 3.44 (m, 2H), 3.18 – 3.12 (m, 1H), 2.49 – 2.41 (m, 1H), 2.31 – 2.27 (m, 1H), 1.57 (d, $J = 6.5$ Hz, 3H), 1.48 (d, $J = 6.5$ Hz, 3H); ^{13}C NMR (125 MHz, CDCl_3) δ 195.1, 145.8, 142.9, 134.5, 134.3, 131.9, 131.5, 127.8, 127.2, 127.0, 126.2, 125.9(8), 125.9(5), 124.3, 102.5, 52.7, 50.1, 49.7, 45.7, 37.8, 36.7, 16.5, 16.1; IR (neat): 3062, 2925, 2854, 1686(s), 1595, 1482, 1336, 1247, 1145, 912; HRESIMS Calcd for $[\text{C}_{23}\text{H}_{23}\text{NNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 416.1291, found 416.1298.

11-fluoro-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[f]naphtho[1,2-d]indol-14(5H)-one (3g)

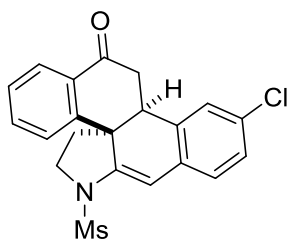


3g

Compound **3g** was prepared in 71% yield (54.4 mg) according to the general procedure. White solid (mp 244-245 °C). ^1H NMR (500 MHz, CDCl_3) δ 7.98 (d, $J = 8.0$ Hz, 1H), 7.44 (t, $J = 7.5$ Hz, 1H), 7.29 (t, $J = 7.5$ Hz, 1H), 7.25 (d, $J = 8.0$ Hz, 1H), 6.94 – 6.90 (m, 1H), 6.87 (d, $J = 9.5$ Hz, 1H), 6.76 – 6.71 (m, 1H), 6.43 (s, 1H), 4.13 – 4.08 (m, 1H), 3.91 – 3.85 (m, 1H), 3.51 – 3.45 (m, 1H), 3.44 – 3.39 (m, 1H), 3.20 (s, 3H), 3.19 – 3.13 (m, 1H), 2.47 – 2.40 (m, 1H), 2.33 – 2.28 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 194.5, 161.5 (d, $J = 243.8$ Hz), 144.7, 142.7, 134.5 (d, $J = 7.5$ Hz), 134.4, 131.5, 130.4 (d, $J =$

3.8 Hz), 128.0, 127.5 (d, $J = 8.8$ Hz), 127.3, 125.7, 113.8 (d, $J = 21.3$ Hz), 112.2 (d, $J = 23.8$ Hz), 102.3, 49.8, 48.8, 45.6, 37.8, 37.2, 36.7; ^{19}F NMR (376 MHz, CDCl_3) δ -114.80 – -115.00 (m); IR (neat): 2923, 2851, 1682(s), 1595, 1487, 1349, 1252, 1157, 961, 768; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{FNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 406.0884, found 406.0889.

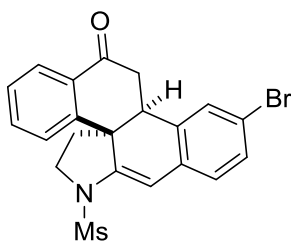
11-chloro-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3h)



3h

Compound **3h** was prepared in 68% yield (54.3 mg) according to the general procedure. White solid (mp 247-248 °C). ^1H NMR (500 MHz, CDCl_3) δ 7.99 (d, $J = 8.0$ Hz, 1H), 7.46 – 7.42 (m, 1H), 7.30 (t, $J = 7.5$ Hz, 1H), 7.23 (d, $J = 8.0$ Hz, 1H), 7.10 (s, 1H), 7.03 – 7.00 (m, 1H), 6.89 (d, $J = 8.0$ Hz, 1H), 6.42 (s, 1H), 4.12 – 4.07 (m, 1H), 3.91 – 3.84 (m, 1H), 3.51 – 3.46 (m, 1H), 3.46 – 3.41 (m, 1H), 3.21 (s, 3H), 3.17 – 3.12 (m, 1H), 2.47 – 2.39 (m, 1H), 2.33 – 2.28 (m, 1H); ^{13}C NMR (125 MHz, CDCl_3) δ 194.3, 145.6, 142.5, 134.4, 134.0, 132.9, 131.8, 131.5, 128.1, 127.4, 127.3, 125.7, 124.8, 102.2, 49.9, 48.8, 45.5, 37.8, 37.4, 36.5; IR (neat): 2925, 2854, 1685(s), 1595, 1477, 1350, 1248, 1158, 961, 736; HRESIMS Calcd for $[\text{C}_{21}\text{H}_{18}\text{ClNNaO}_3\text{S}]^+$ ($\text{M} + \text{Na}^+$) 422.0588, found 422.0590.

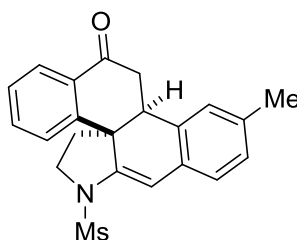
11-bromo-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3i)



3i

Compound **3i** was prepared in 58% yield (51.4 mg) according to the general procedure. White solid (mp 270-271 °C). ¹H NMR (400 MHz, CDCl₃) δ 8.00 (d, *J* = 7.6 Hz, 1H), 7.47 – 7.42 (m, 1H), 7.33 – 7.28 (m, 1H), 7.24 – 7.19 (m, 2H), 7.19 – 7.15 (m, 1H), 6.83 (d, *J* = 8.4 Hz, 1H), 6.40 (s, 1H), 4.13 – 4.07 (m, 1H), 3.91 – 3.83 (m, 1H), 3.52 – 3.47 (m, 1H), 3.47 – 3.40 (m, 1H), 3.20 (s, 3H), 3.17 – 3.11 (m, 1H), 2.47 – 2.38 (m, 1H), 2.33 – 2.27 (m, 1H); ¹³C NMR (100 MHz, CDCl₃) δ 194.3, 145.7, 142.4, 134.4, 134.3, 133.4, 131.5, 130.4, 128.1, 127.7, 127.5(2), 127.4(5), 125.6, 119.7, 102.2, 50.0, 48.8, 45.5, 37.8, 37.4, 36.4; IR (neat): 2925, 2854, 1684(s), 1595, 1475, 1349, 1247, 1158, 961, 735; HRESIMS Calcd for [C₂₁H₁₈BrNNaO₃S]⁺ (*M* + Na⁺) 466.0083, found 466.0073.

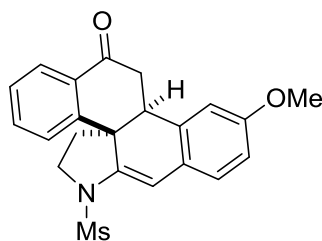
11-methyl-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3j)



3j

Compound **3j** was prepared in 72% yield (54.6 mg) according to the general procedure. White solid (mp 256-257 °C). ¹H NMR (400 MHz, CDCl₃) δ 7.97 (d, *J* = 8.4 Hz, 1H), 7.45 – 7.40 (m, 1H), 7.29 – 7.24 (m, 2H), 6.95 (s, 1H), 6.86 (s, 2H), 6.44 (s, 1H), 4.11 – 4.05 (m, 1H), 3.90 – 3.82 (m, 1H), 3.53 – 3.46 (m, 2H), 3.19 (s, 3H), 3.16 – 3.09 (m, 1H), 2.47 – 2.38 (m, 1H), 2.31 – 2.26 (m, 1H), 2.21 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 195.1, 144.3, 143.2, 136.1, 134.3, 132.0, 131.6, 131.5, 127.9, 127.8, 127.2, 126.3, 125.8, 125.2, 103.2, 50.0, 48.7, 45.6, 37.9, 37.0, 36.7, 21.3; IR (neat): 2924, 2854, 1684(s), 1595, 1342, 1249, 1158, 1065, 961, 735; HRESIMS Calcd for [C₂₂H₂₁NNaO₃S]⁺ (*M* + Na⁺) 402.1134, found 402.1139.

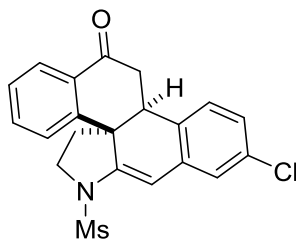
11-methoxy-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3k)



3k

Compound **3k** was prepared in 67% yield (52.9 mg) according to the general procedure. White solid (mp 220-221 °C). ¹H NMR (400 MHz, CDCl₃) δ 7.97 (d, *J* = 8.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.30 – 7.24 (m, 2H), 6.90 (d, *J* = 8.4 Hz, 1H), 6.73 (s, 1H), 6.59 – 6.56 (m, 1H), 6.43 (s, 1H), 4.11 – 4.05 (m, 1H), 3.90 – 3.82 (m, 1H), 3.70 (s, 3H), 3.49 – 3.43 (m, 2H), 3.18 (s, 3H), 3.17 – 3.10 (m, 1H), 2.47 – 2.37 (m, 1H), 2.31 – 2.26 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 194.9, 158.4, 143.3, 143.1, 134.3, 133.9, 131.6, 127.9, 127.3, 127.2, 127.1, 125.8, 111.7, 111.4, 103.0, 55.2, 49.9, 48.7, 45.8, 38.0, 36.9, 36.8; IR (neat): 2922, 2851, 1683(s), 1596, 1491, 1347, 1253, 1157, 961, 767; HRESIMS Calcd for [C₂₂H₂₁NNaO₄S]⁺ (*M* + Na⁺) 418.1083, found 418.1088.

10-chloro-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[f]naphtho[1,2-d]indol-14(5H)-one (3l)

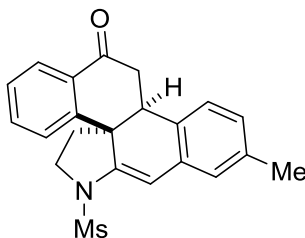


3l

Compound **3l** was prepared in 71% yield (56.7 mg) according to the general procedure. White solid (mp 245-246 °C). ¹H NMR (500 MHz, CDCl₃) δ 7.97 (d, *J* = 7.5 Hz, 1H), 7.44 (t, *J* = 7.0 Hz, 1H), 7.29 (t, *J* = 7.5 Hz, 1H), 7.21 (d, *J* = 7.5 Hz, 1H), 7.05 (d, *J* = 8.5 Hz, 1H), 6.98 – 6.91 (m, 2H), 6.38 (s, 1H), 4.13 – 4.07 (m, 1H), 3.92 – 3.85 (m, 1H), 3.47 – 3.42 (m, 2H), 3.21 (s, 3H), 3.17 – 3.11 (m, 1H), 2.47 – 2.40 (m, 1H), 2.33 – 2.28 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 194.6, 146.7, 142.4, 136.2, 134.4, 133.1, 131.5, 130.5, 128.1, 127.2, 126.1, 125.8, 125.6(4), 125.6(2), 101.9, 50.2, 48.9, 45.2, 37.6, 37.4,

36.7; IR (neat): 3063, 2926, 1685(s), 1594, 1348, 1247, 1158, 961, 805, 762; HRESIMS Calcd for $[C_{21}H_{18}ClNNaO_3S]^+$ ($M + Na^+$) 422.0588, found 422.0595.

10-methyl-7-(methylsulfonyl)-6,7,12b,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one (3m)

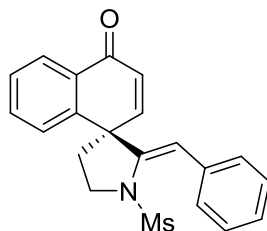


3m

Compound **3m** was prepared in 75% yield (56.9 mg) according to the general procedure. White solid (mp 269-270 °C). 1H NMR (400 MHz, $CDCl_3$) δ 7.96 (d, $J = 7.2$ Hz, 1H), 7.45 – 7.40 (m, 1H), 7.30 – 7.22 (m, 2H), 7.03 (d, $J = 7.6$ Hz, 1H), 6.85 – 6.77 (m, 2H), 6.42 (s, 1H), 4.12 – 4.06 (m, 1H), 3.92 – 3.84 (m, 1H), 3.51 – 3.43 (m, 2H), 3.19 (s, 3H), 3.15 – 3.08 (m, 1H), 2.47 – 2.38 (m, 1H), 2.32 – 2.26 (m, 1H), 2.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 195.2, 145.3, 143.1, 136.9, 134.3, 134.1, 131.6, 129.2, 127.8, 127.3, 127.1, 126.9, 125.8, 124.2, 103.3, 50.2, 48.8, 45.4, 37.8, 37.1, 36.8, 20.9; IR (neat): 2922, 2852, 1682(s), 1595, 1346, 1156, 1091, 960, 764, 546; HRESIMS Calcd for $[C_{22}H_{21}NNaO_3S]^+$ ($M + Na^+$) 402.1134, found 402.1138.

General procedure for the synthesis of the products (-)-2. To a mixture of the ynamide **1** (0.1 mmol) in Et_2O (2 mL) at 20 °C, NTP (0.0025 mmol, 2.1 mg) was added. Then, the reaction mixture was stirred at 20 °C for 12–36 h and the progress of the reaction was monitored by TLC. Upon completion, the mixture was quenched by Et_3N , then concentrated and the residue was purified by chromatography on silica gel (eluent: DCM) to afford the desired product (-)-2.

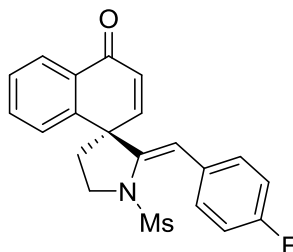
(*R,Z*)-2'-benzylidene-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2a)



(-)-2a

Compound **(-)-2a** was prepared in 60% yield (21.9 mg) according to the general procedure. White solid (mp 197-198 °C). $[\alpha]_D^{20} = -54.3^\circ$ (c = 1.0, CHCl₃). 90:10 e.r. (determined by HPLC: Chiralpak ODH Column, 20/80 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 19.47 min (minor), 22.23 min (major)).

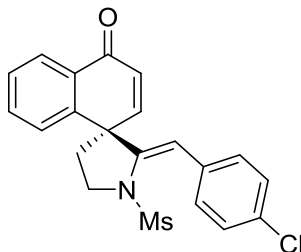
(*R,Z*)-2'-(4-fluorobenzylidene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2g)



(-)-2g

Compound **(-)-2g** was prepared in 65% yield (24.9 mg) according to the general procedure. White solid (mp 197-198 °C). $[\alpha]_D^{20} = -17.8^\circ$ (c = 1.0, CHCl₃). 90:10 e.r. (determined by HPLC: Chiralpak IC Column, 50/50 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 16.59 min (major), 18.36 min (minor)).

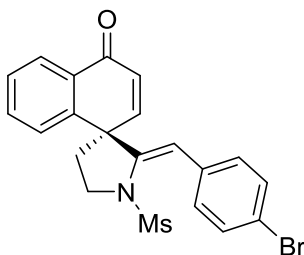
(*R,Z*)-2'-(4-chlorobenzylidene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2h)



(-)-2h

Compound **(-)-2h** was prepared in 55% yield (22.0 mg) according to the general procedure. White solid (mp 178-179 °C). $[\alpha]_D^{20} = -19.0^\circ$ (c = 1.0, CHCl₃). 91:9 e.r. (determined by HPLC: Chiralpak IC Column, 40/60 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 20.69 min (major), 22.18 min (minor)).

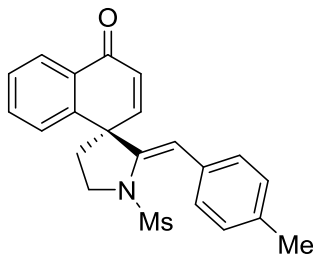
(*R,Z*)-2'-(4-bromobenzylidene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2i)



(-)-2i

Compound **(-)-2i** was prepared in 48% yield (21.3 mg) according to the general procedure. White solid (mp 192-193 °C). $[\alpha]_D^{20} = -83.0^\circ$ (c = 1.0, CHCl₃). 92.5:7.5 e.r. (determined by HPLC: Chiralpak IC Column, 30/70 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 37.16 min (major), 39.87 min (minor)).

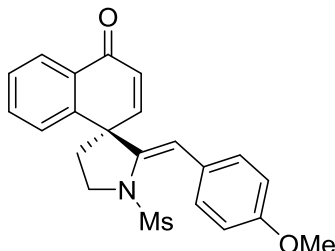
(*R,Z*)-2'-(4-methylbenzylidene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2j)



(-)-2j

Compound **(-)-2j** was prepared in 63% yield (23.9 mg) according to the general procedure. White solid (mp 209-210 °C). $[\alpha]_D^{20} = -46.3^\circ$ (c = 1.0, CHCl₃). 85:15 e.r. (determined by HPLC: Chiralpak IG Column, 50/50 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 15.75 min (minor), 19.47 min (major)).

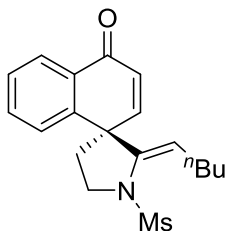
(*R,Z*)-2'-(4-methoxybenzylidene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2k)



(-)-2k

Compound **(-)-2k** was prepared in 58% yield (22.9 mg) according to the general procedure. White solid (mp 155-156 °C). $[\alpha]_D^{20} = -8.5^\circ$ ($c = 1.0$, CHCl_3). 87:13 e.r. (determined by HPLC: Chiralpak IG Column, 50/50 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 24.69 min (minor), 31.62 min (major)).

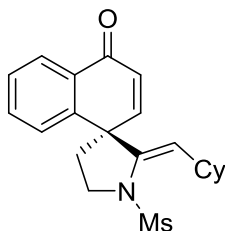
(*R,Z*)-1'-(methylsulfonyl)-2'-pentylidene-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2n)



(-)-2n

Compound **(-)-2n** was prepared in 48% yield (22.9 mg) according to the general procedure. Colorless oil. $[\alpha]_D^{20} = -14.8^\circ$ ($c = 1.0$, CHCl_3). 82.5:17.5 e.r. (determined by HPLC: Chiralpak IB Column, 10/90 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 15.83 min (major), 16.89 min (minor)).

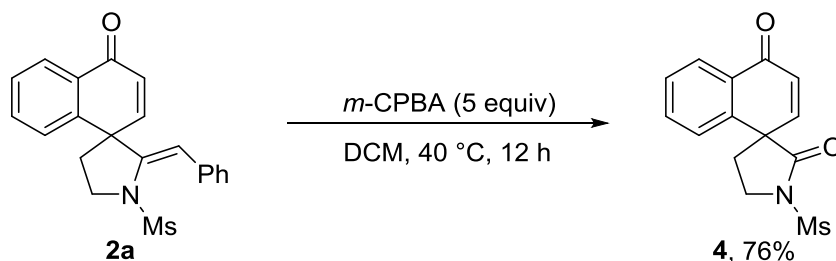
(*R,Z*)-2'-(cyclohexylmethylene)-1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidin]-4-one ((-)-2o)



(-)-2o

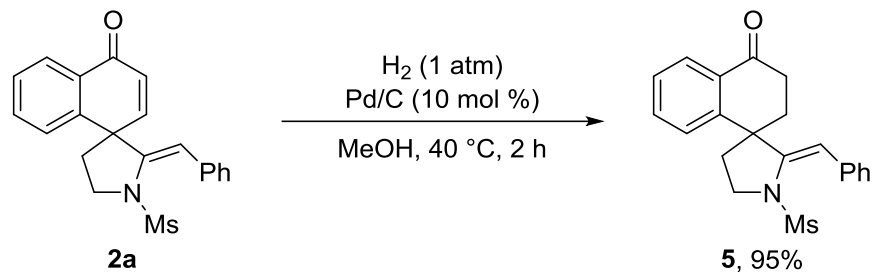
Compound **(-)-2o** was prepared in 38% yield (22.9 mg) according to the general procedure. White solid (mp 110-111 °C). $[\alpha]_D^{20} = -44.4^\circ$ (c = 1.0, CHCl₃). 83.5:16.5 e.r. (determined by HPLC: Chiralpak IB Column, 10/90 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 16.83 min (major), 17.92 min (minor)).

1'-(methylsulfonyl)-4*H*-spiro[naphthalene-1,3'-pyrrolidine]-2',4-dione (4**)⁴**



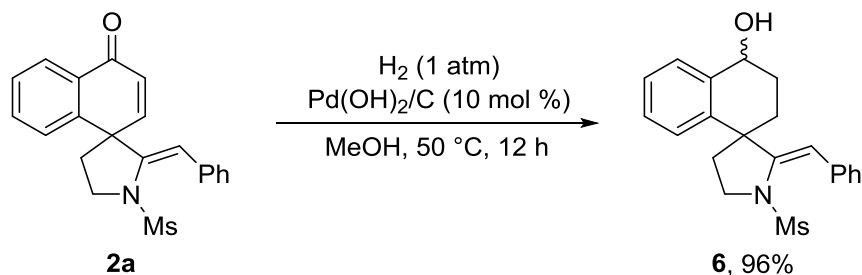
2a (0.1 mmol, 36.7 mg) was dissolved in DCM (2 mL) in a vial and stirred. *m*-CPBA (0.5 mmol, 86.3 mg) was added and the mixture was reacted at 40 °C in 12 h. Upon completion, the mixture was diluted by DCM, washed by NaHCO₃ three times and concentrated. The residue was purified by chromatography on silica gel (eluent: hexanes/ethyl acetate) to afford the desired product **4** (22.1 mg, 76% yield, white solid (mp 198-199 °C)). ¹H NMR (400 MHz, CDCl₃) δ 8.23 (d, *J* = 7.6 Hz, 1H), 7.66 (t, *J* = 7.2 Hz, 1H), 7.53 (t, *J* = 7.6 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 1H), 6.95 (d, *J* = 10.0 Hz, 1H), 6.63 (d, *J* = 10.0 Hz, 1H), 4.25 – 4.18 (m, 1H), 4.16 – 4.09 (m, 1H), 3.36 (s, 3H), 2.80 – 2.71 (m, 1H), 2.64 – 2.56 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 183.4, 172.3, 143.0, 141.4, 133.6, 131.8, 130.6, 128.8, 127.3, 126.2, 52.8, 43.3, 40.7, 33.9; IR (neat): 2920, 2850, 1725(s), 1662(s), 1457, 1354, 1167, 1105, 966, 775; HRESIMS Calcd for [C₁₄H₁₃NNaO₄S]⁺ (M + Na⁺) 314.0457, found 314.0467.

(Z)-2'-benzylidene-1'-(methylsulfonyl)-2H-spiro[naphthalene-1,3'-pyrrolidin]-4(3H)-one (5)



2a (0.1 mmol, 36.7 mg) was mixed with Pd/C (10% w/w Pd in activated carbon, 0.01 mmol, 10.6 mg) in a dry tube. Then, the mixtures were added MeOH (2 mL) and stirred under H₂ atmosphere at 40 °C in 2 h. Upon completion, the mixture was concentrated and purified by chromatography on silica gel (eluent: hexanes/ethyl acetate) to afford the desired product **5** (34.9 mg, 95% yield, white solid (mp 157-158 °C)). ¹H NMR (500 MHz, CDCl₃) δ 8.10 (d, *J* = 8.0 Hz, 1H), 7.58 – 7.54 (m, 1H), 7.49 (d, *J* = 7.5 Hz, 1H), 7.38 (t, *J* = 7.5 Hz, 1H), 7.34 – 7.28 (m, 4H), 7.23 – 7.18 (m, 1H), 5.66 (s, 1H), 4.05 – 3.98 (m, 1H), 3.76 – 3.70 (m, 1H), 2.94 – 2.87 (m, 1H), 2.78 – 2.71 (m, 1H), 2.59 (s, 3H), 2.44 – 2.37 (m, 2H), 2.35 – 2.29 (m, 1H), 2.26 – 2.19 (m, 1H); ¹³C NMR (125 MHz, CDCl₃) δ 197.5, 147.0, 146.2, 137.3, 134.2, 132.6, 128.7, 128.3, 127.5(1), 127.4(7), 127.4(5), 127.1, 119.1, 49.9, 47.5, 41.7, 37.0, 35.5, 33.5; IR (neat): 2924, 1634(s), 1334, 1146, 1094, 1047, 767, 695, 658, 543; HRESIMS Calcd for [C₂₁H₂₁NNaO₃S]⁺ (M + Na⁺) 390.1134, found 390.1145.

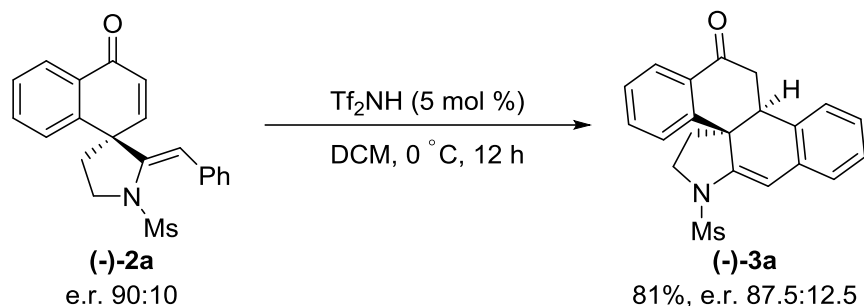
(Z)-2'-benzylidene-1'-(methylsulfonyl)-3,4-dihydro-2H-spiro[naphthalene-1,3'-pyrrolidin]-4-ol (6)



2a (0.1 mmol, 36.7 mg) was mixed with Pd(OH)₂/C (10% w/w Pd in activated carbon, 0.01 mmol, 14.1 mg) in a dry tube. Then, the mixtures were added MeOH (2 mL) and

stirred under H₂ atmosphere at 50 °C in 5 h. Upon completion, the mixture was concentrated and the residue was purified by chromatography on silica gel (eluent: hexanes/ethyl acetate) to afford the desired product **6** (35.4 mg, 96% yield, colorless oil). ¹H NMR (400 MHz, CDCl₃) δ 7.49 – 7.46 (m, 1H), 7.35 – 7.24 (m, 7H), 7.20 – 7.15 (m, 1H), 5.62 (s, 1H), 4.81 (t, *J* = 5.2 Hz, 1H), 3.98 – 3.90 (m, 1H), 3.72 – 3.65 (m, 1H), 2.59 (s, 3H), 2.31 – 2.15 (m, 3H), 2.10 – 2.00 (m, 2H), 1.95 – 1.90 (m, 1H), 1.84 – 1.77 (m, 1H); ¹³C NMR (100 MHz, CDCl₃) δ 148.5, 140.5, 139.6, 137.9, 128.8, 128.6, 128.4, 128.3, 128.1, 127.1, 126.7, 118.0, 68.2, 50.1, 47.5, 41.3, 39.2, 30.4, 29.0; IR (neat): 3025, 2933, 1488, 1447, 1334, 1148, 1064, 963, 761, 699; HRESIMS Calcd for [C₂₁H₂₃NNaO₃S]⁺ (*M* + Na⁺) 392.1291, found 392.1283.

(4*bS*,12*bR*)-7-(methylsulfonyl)-6,7,12*b*,13-tetrahydrobenzo[*f*]naphtho[1,2-*d*]indol-14(5*H*)-one ((-)-3a**)**



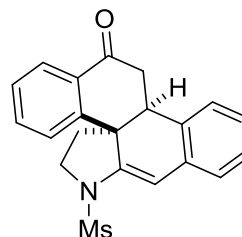
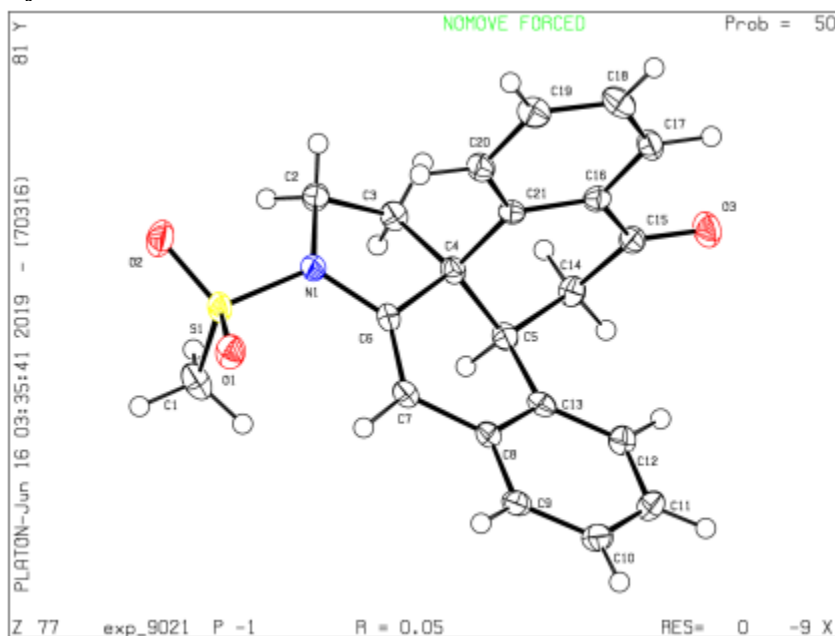
(-)-2a (0.1 mmol, 36.7 mg) was mixed with DCM (2 mL) in a dry tube. Then Tf₂NH (0.005 mmol, 1.4 mg) was added and stirred at 0 °C in 12 h. Upon completion, the mixture was quenched by Et₃N, then concentrated and purified by chromatography on silica gel (eluent: DCM) to afford the desired **(-)-3a** (14.8 mg, 81% yield, white solid (mp 242-243 °C)). [α]_D²⁰ = -37.1 ° (*c* = 1.0, CHCl₃). 87.5:12.5 e.r. (determined by HPLC: Chiralpak IC Column, 50/50 *i*-PrOH/hexane, 1.0 mL/min, 254 nm; TR = 14.23 min (major), 18.65 min (minor)).

Reference:

- [1] K. Antien, G. Viault, L. Pouys égu, P. A. Peixoto and S. Quideau, *Tetrahedron*, 2017, **73**, 3684.
- [2] R.-Q. Xu, Q. Gu and S.-L. You, *Angew. Chem., Int. Ed.*, 2017, **56**, 7252.

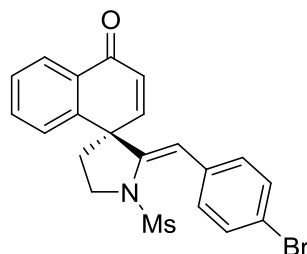
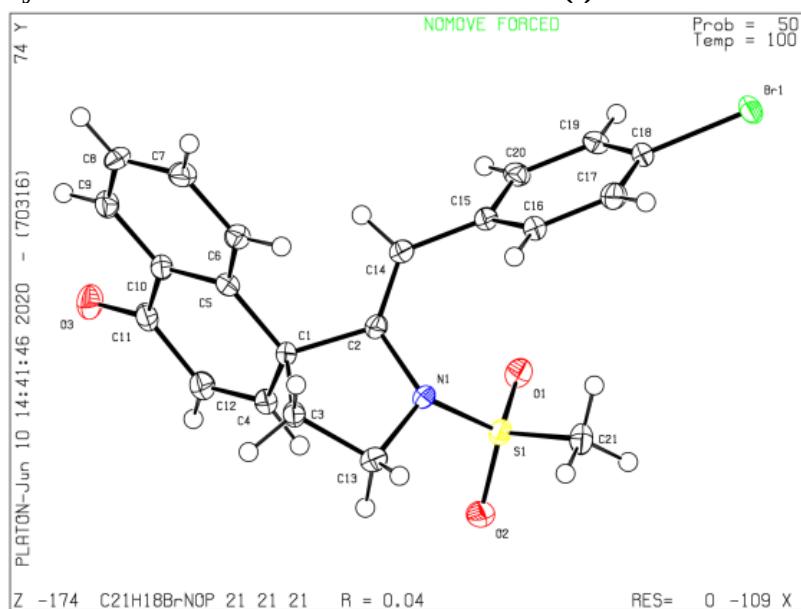
- [3] Y.-Q. Zhang, Y.-B. Chen, J.-R. Liu, S.-Q. Wu, X.-Y. Fan, Z.-X. Zhang, X. Hong and L.-W. Ye, *Nat. Chem.*, 2021, **13**, 1093.
- [4] Q. Wang, L. Zhang, J. Yao, G. Qiu, X. Li and H. Zhou, *J. Org. Chem.*, 2018, **83**, 4092.

Crystal data and structure refinement for 3a. CCDC Number = 2164054



Bond precision:	C-C = 0.0030 Å	Wavelength=1.54184
Cell:	a=5.8030 (3) alpha=91.966 (4)	b=11.2947 (6) beta=97.567 (4)
Temperature:	133 K	c=13.4472 (6) gamma=103.085 (5)
Volume	Calculated 849.13 (8)	Reported 849.13 (8)
Space group	P -1	P -1
Hall group	-P 1	-P 1
Moiety formula	C21 H19 N O3 S	C21 H19 N O3 S
Sum formula	C21 H19 N O3 S	C21 H19 N O3 S
Mr	365.43	365.43
Dx, g cm ⁻³	1.429	1.429
Z	2	2
Mu (mm ⁻¹)	1.874	1.874
F000	384.0	384.0
F000'	385.74	
h,k,lmax	6,12,15	6,12,15
Nref	2610	2598
Tmin,Tmax	0.911,0.945	0.917,1.000
Tmin'	0.911	
Correction method= # Reported T Limits: Tmin=0.917 Tmax=1.000		
AbsCorr = MULTI-SCAN		
Data completeness=	0.995	Theta(max)= 61.156
R(reflections)=	0.0518 (2386)	wR2(reflections)= 0.1386 (2598)
S =	1.062	Npar= 311

Crystal data and structure refinement for (-)-2i. CCDC Number = 2024760



Bond precision: C-C = 0.0071 Å

Wavelength=1.54184

Cell: a=5.6000(1)

b=18.0276(3)

c=18.3366(3)

alpha=90

beta=90

gamma=90

Temperature: 100 K

	Calculated	Reported
Volume	1851.16(5)	1851.16(5)
Space group	P 21 21 21	P 21 21 21
Hall group	P 2ac 2ab	P 2ac 2ab
Moiety formula	C ₂₁ H ₁₈ Br N O ₃ S	C ₂₁ H ₁₈ Br N O ₃ S
Sum formula	C ₂₁ H ₁₈ Br N O ₃ S	C ₂₁ H ₁₈ Br N O ₃ S
Mr	444.32	444.33
Dx, g cm ⁻³	1.594	1.594
Z	4	4
Mu (mm ⁻¹)	4.269	4.269
F ₀₀₀	904.0	904.0
F ₀₀₀ '	904.78	
h, k, lmax	6, 22, 22	6, 22, 22
Nref	3684[2149]	3662
Tmin, Tmax	0.950, 0.958	0.544, 1.000
Tmin'	0.808	

Correction method= # Reported T Limits: Tmin=0.544 Tmax=1.000

AbsCorr = MULTI-SCAN

Data completeness= 1.70/0.99

Theta(max)= 72.838

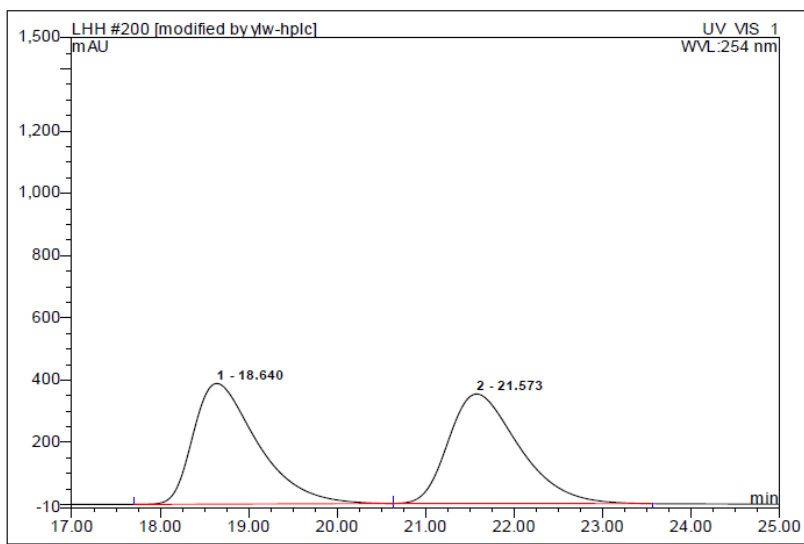
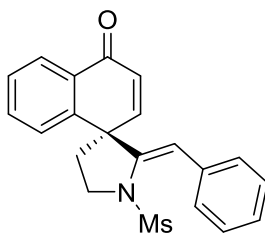
R(reflections)= 0.0428(3519)

wR2(reflections)= 0.1147(3662)

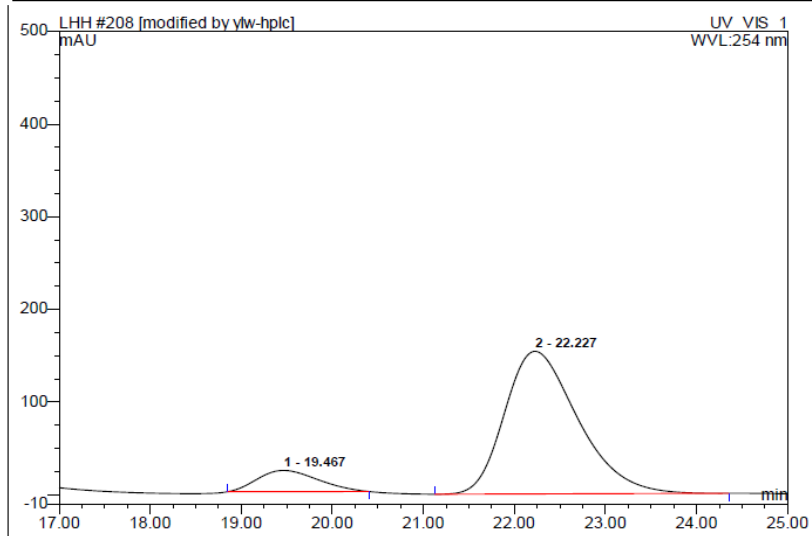
S = 1.060

Npar= 245

(-)-2a: ODH, *n*-hexane/2-propanol =80/20, $\nu = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

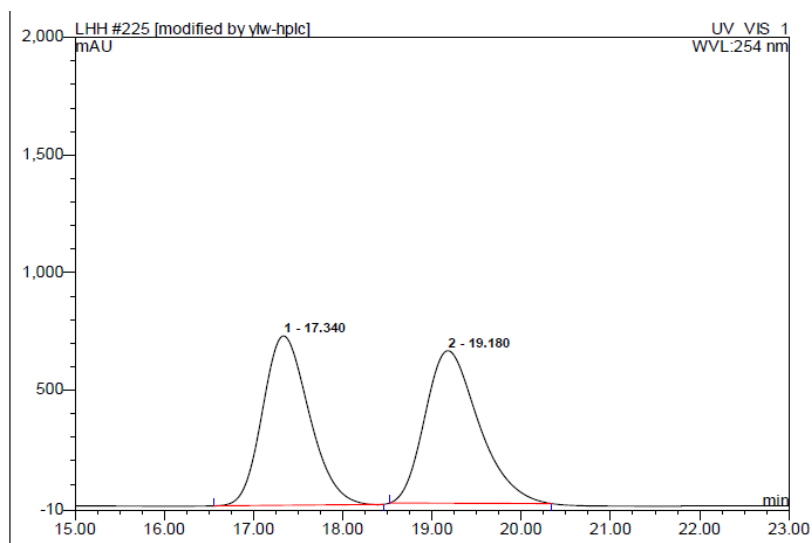
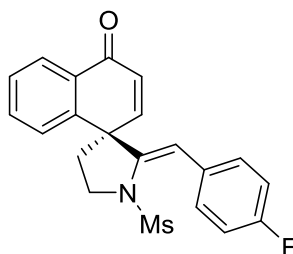


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	18.64	n.a.	387.567	325.725	50.10	n.a.	BMB*
2	21.57	n.a.	351.706	324.452	49.90	n.a.	bMB*
Total:			739.273	650.177	100.00	0.000	

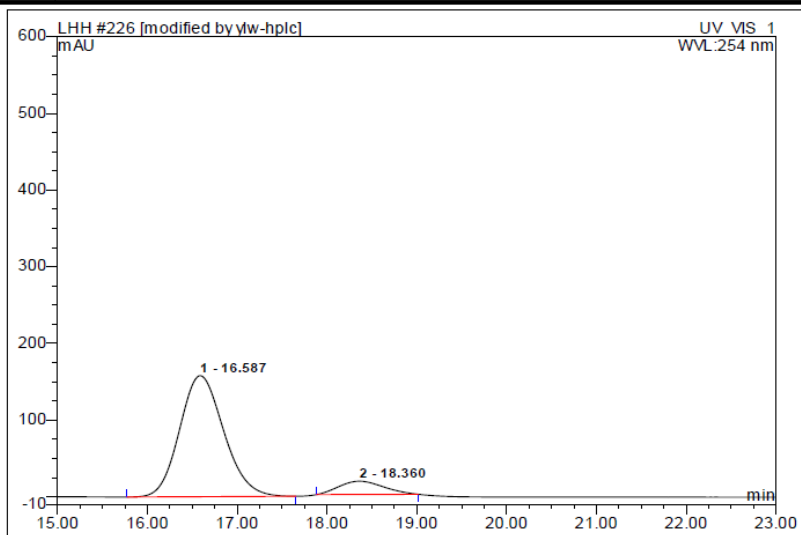


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	19.47	n.a.	22.895	17.549	10.90	n.a.	BMB*
2	22.23	n.a.	153.936	143.486	89.10	n.a.	BMB*
Total:			176.831	161.034	100.00	0.000	

(-)-2g: IC, *n*-hexane/2-propanol =50/50, $v = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

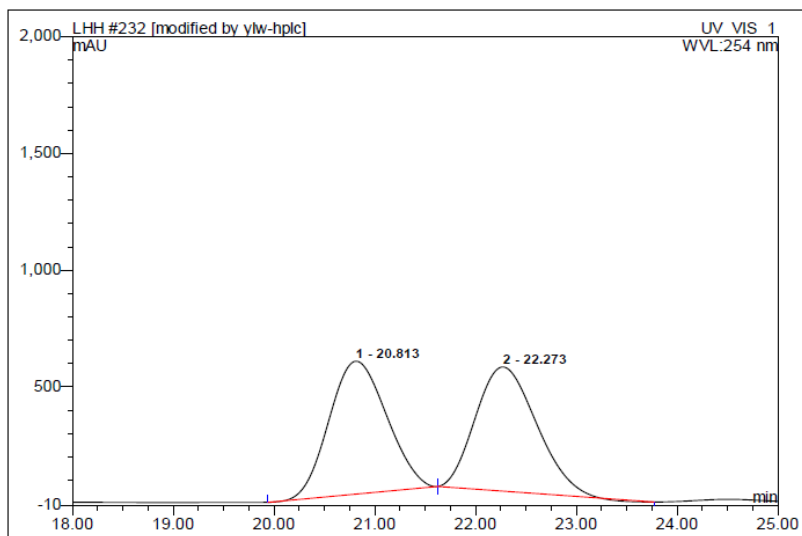
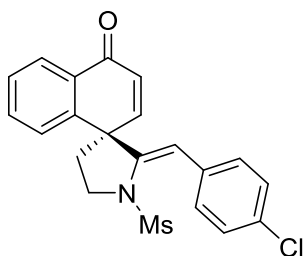


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	17.34	n.a.	719.876	435.252	48.98	n.a.	BMB*
2	19.18	n.a.	648.434	453.384	51.02	n.a.	BMB*
Total:			1368.311	888.636	100.00	0.000	

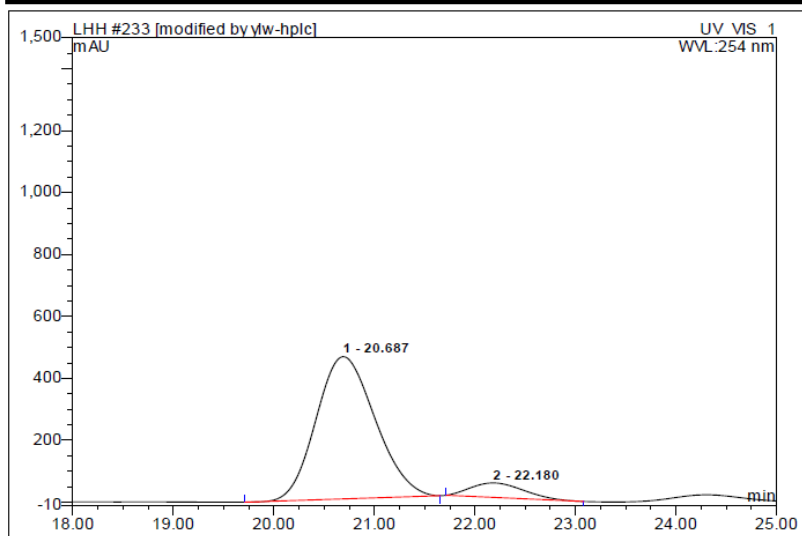


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	16.59	n.a.	157.786	89.964	89.98	n.a.	BMB*
2	18.36	n.a.	17.338	10.018	10.02	n.a.	BMB*
Total:			175.124	99.982	100.00	0.000	

(-)-2h: IC, *n*-hexane/2-propanol =60/40, $v = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

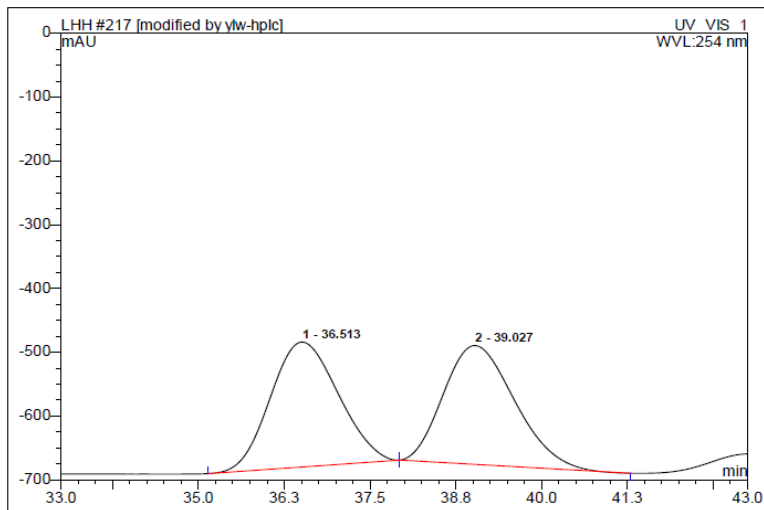
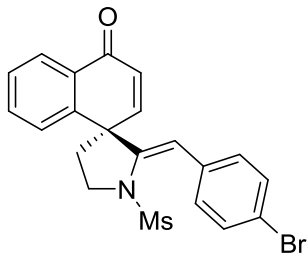


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	20.81	n.a.	569.760	376.496	50.29	n.a.	BMB*
2	22.27	n.a.	532.339	372.214	49.71	n.a.	bMB*
Total:			1102.099	748.710	100.00	0.000	

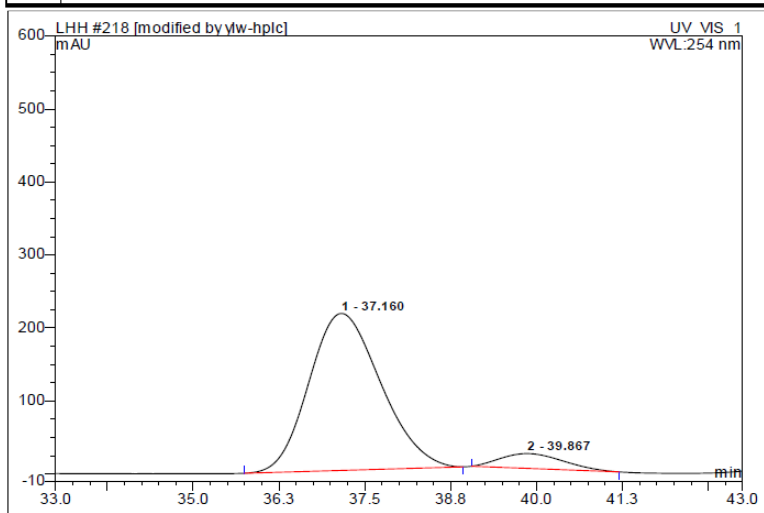


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	20.69	n.a.	459.265	312.193	91.59	n.a.	BMB*
2	22.18	n.a.	46.611	28.658	8.41	n.a.	BMB*
Total:			505.876	340.851	100.00	0.000	

(-)-2i: IC, *n*-hexane/2-propanol =70/30, $\nu = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

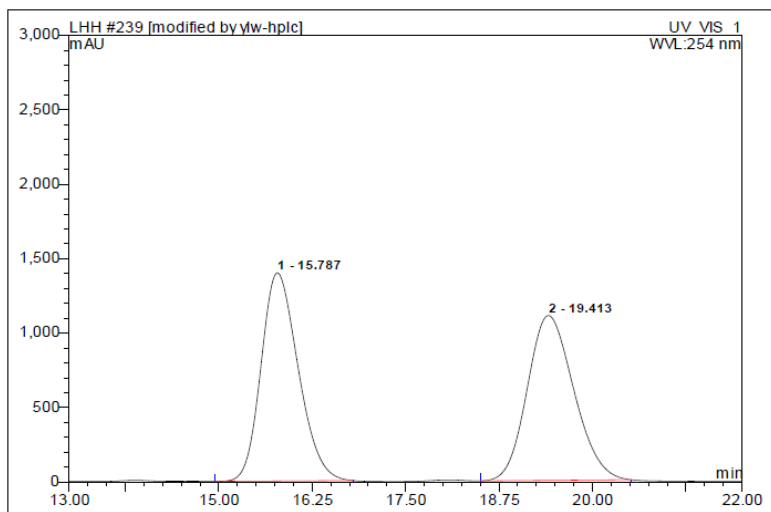
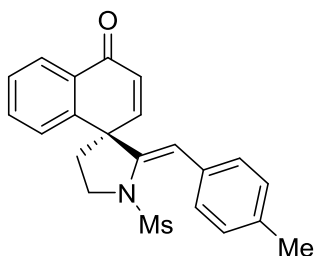


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	36.51	n.a.	195.707	223.603	50.00	n.a.	BMB*
2	39.03	n.a.	186.234	223.600	50.00	n.a.	bMB*
Total:			381.941	447.203	100.00	0.000	

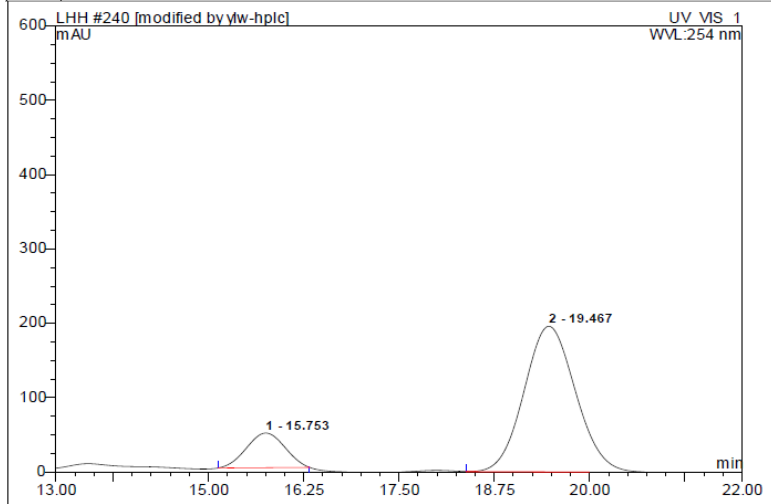


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	37.16	n.a.	215.106	260.020	92.31	n.a.	BMB*
2	39.87	n.a.	20.261	21.648	7.69	n.a.	BMB*
Total:			235.367	281.668	100.00	0.000	

(-)-2j: IG, *n*-hexane/2-propanol =50/50, $v = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

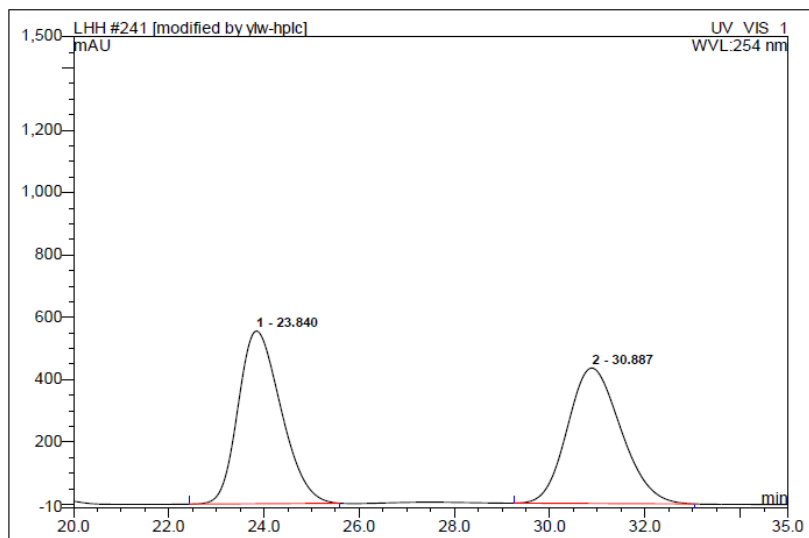
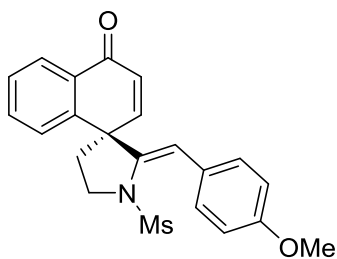


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	15.79	n.a.	1398.374	804.490	50.22	n.a.	BMB*
2	19.41	n.a.	1107.164	797.496	49.78	n.a.	BMB*
Total:			2505.539	1601.985	100.00	0.000	

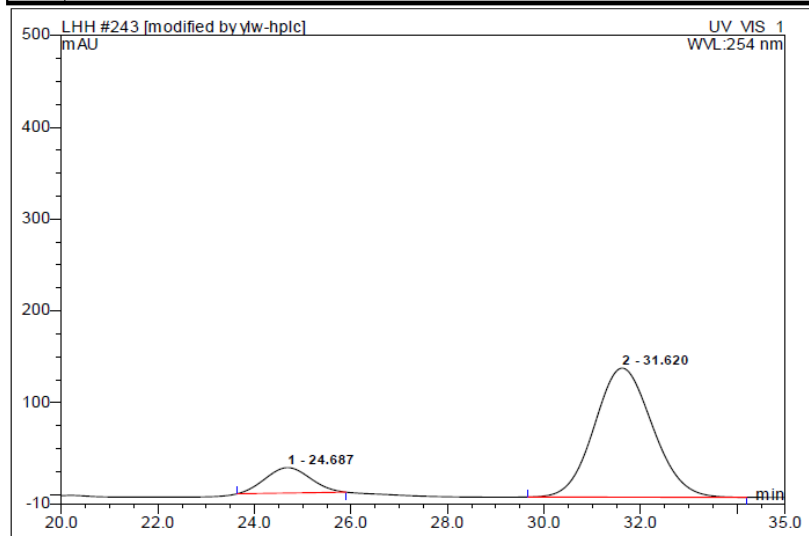


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	15.75	n.a.	46.578	27.093	15.03	n.a.	BMB*
2	19.47	n.a.	195.791	153.144	84.97	n.a.	BMB*
Total:			242.369	180.237	100.00	0.000	

(-)-**2k**: IG, *n*-hexane/2-propanol =50/50, $v = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

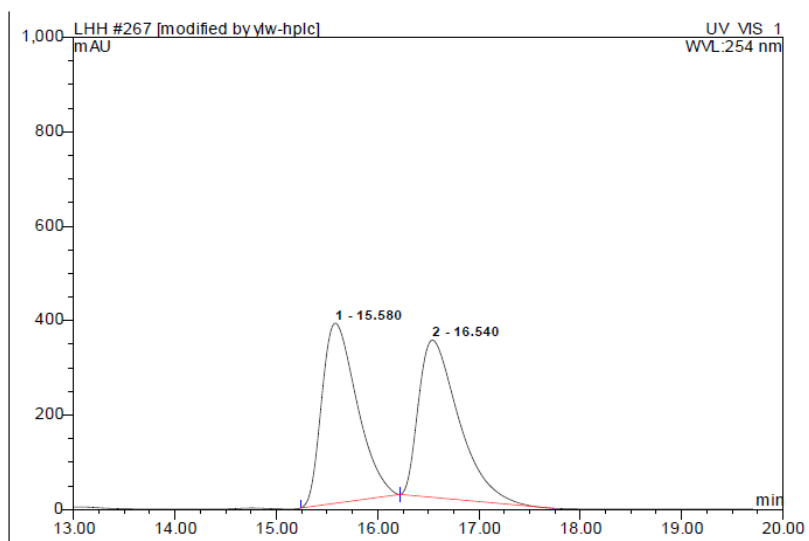
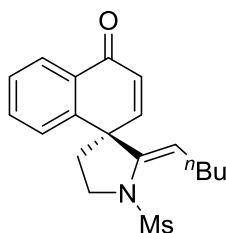


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	23.84	n.a.	553.843	578.295	50.58	n.a.	BMB*
2	30.89	n.a.	434.363	564.965	49.42	n.a.	BMB*
Total:			988.206	1143.260	100.00	0.000	

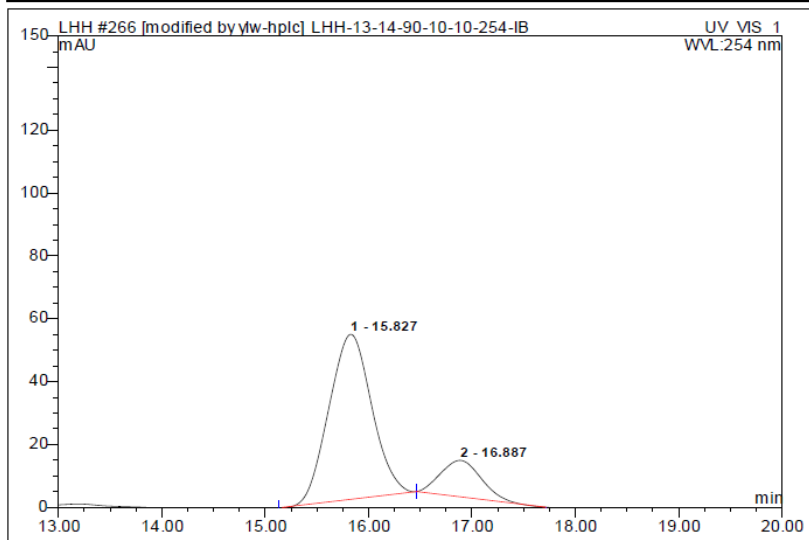


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	24.69	n.a.	27.617	29.820	13.19	n.a.	BMB*
2	31.62	n.a.	140.522	196.192	86.81	n.a.	BMB*
Total:			168.139	226.011	100.00	0.000	

(-)-2n: IB, *n*-hexane/2-propanol =90/10, $\nu = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

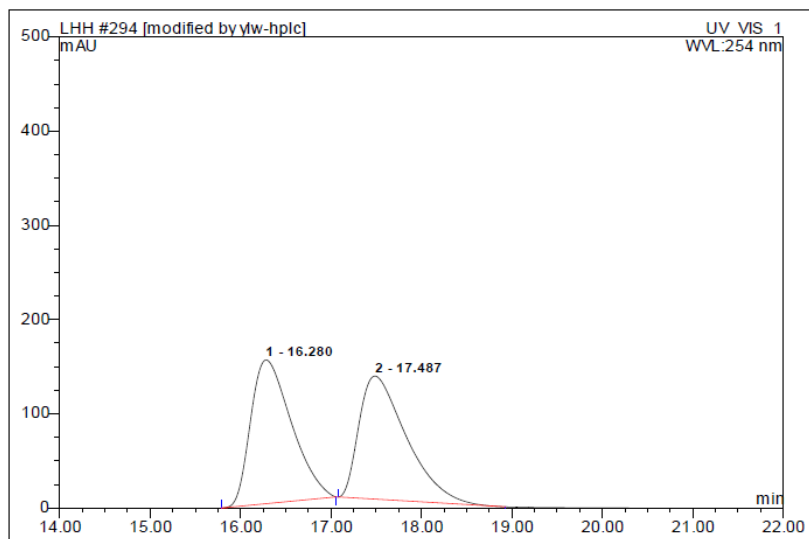
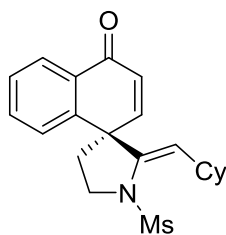


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	15.58	n.a.	381.569	153.888	50.04	n.a.	BMb*
2	16.54	n.a.	333.125	153.633	49.96	n.a.	bMB*
Total:			714.694	307.521	100.00	0.000	

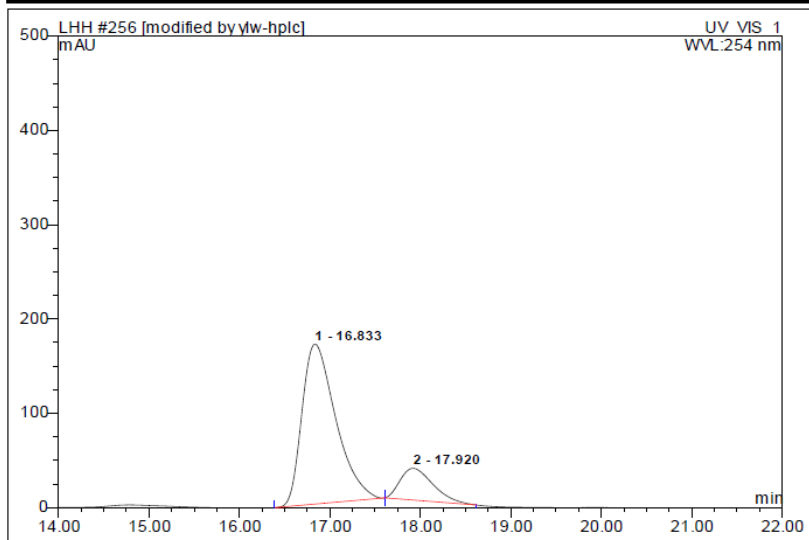


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	15.83	n.a.	52.517	25.139	82.62	n.a.	BMb*
2	16.89	n.a.	11.596	5.287	17.38	n.a.	bMB*
Total:			64.113	30.426	100.00	0.000	

(-)-2o: IB, *n*-hexane/2-propanol =90/10, $\nu = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$

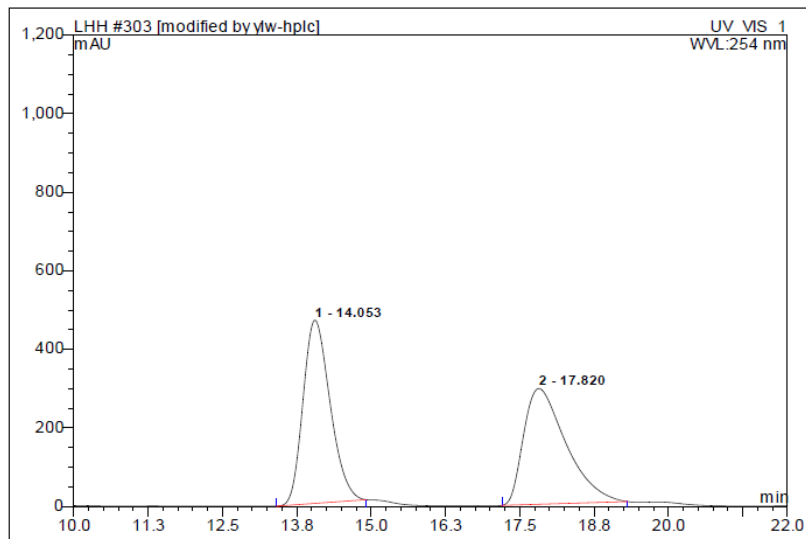
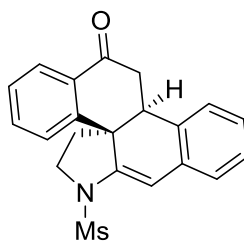


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	16.28	n.a.	152.546	79.683	50.13	n.a.	BMB*
2	17.49	n.a.	130.615	79.268	49.87	n.a.	BMB*
Total:			283.161	158.951	100.00	0.000	

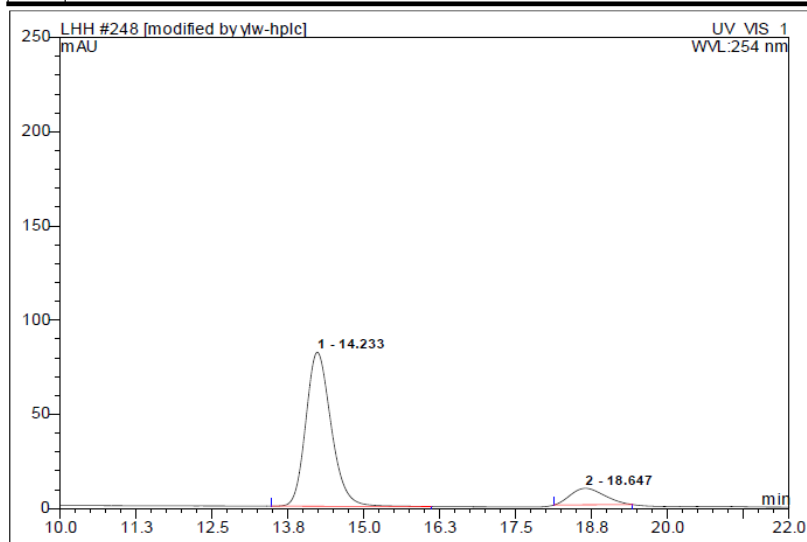


No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	16.83	n.a.	169.634	73.286	83.76	n.a.	BMB*
2	17.92	n.a.	33.660	14.205	16.24	n.a.	bMB*

(-)-3a: IC, *n*-hexane/2-propanol =50/50, $v = 1.0 \text{ mL min}^{-1}$, $\lambda = 254 \text{ nm}$



No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	14.05	n.a.	466.228	246.292	50.31	n.a.	BMB*
2	17.82	n.a.	294.551	243.266	49.69	n.a.	BMB*
Total:			760.779	489.558	100.00	0.000	



No.	Ret.Time min	Peak Name	Height mAU	Area mAU*min	Rel.Area %	Amount n.a.	Type
1	14.23	n.a.	81.767	39.607	87.54	n.a.	BMB*
2	18.65	n.a.	8.737	5.636	12.46	n.a.	BMB*
Total:			90.504	45.243	100.00	0.000	

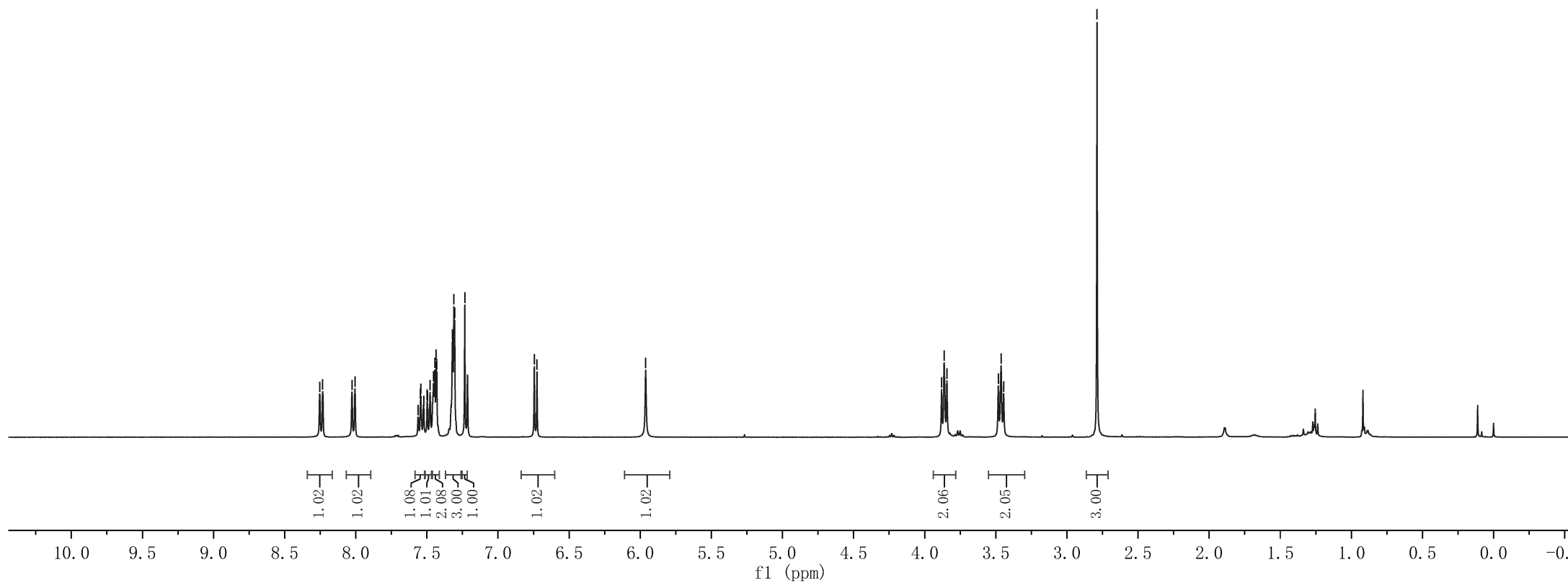
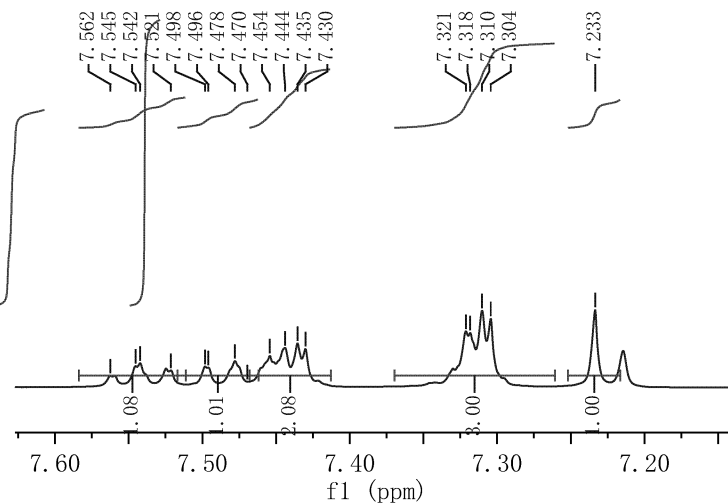
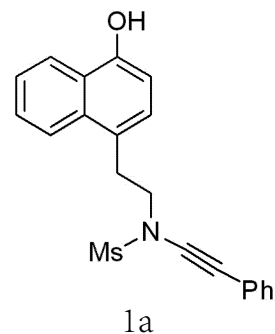
Parameter	Value
1 Title	LHH-19-108
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-02-04T18:31:24
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

8.254
8.233
8.027
8.006
7.454
7.444
7.435
7.430
7.321
7.318
7.310
7.304
7.233
6.743
6.726

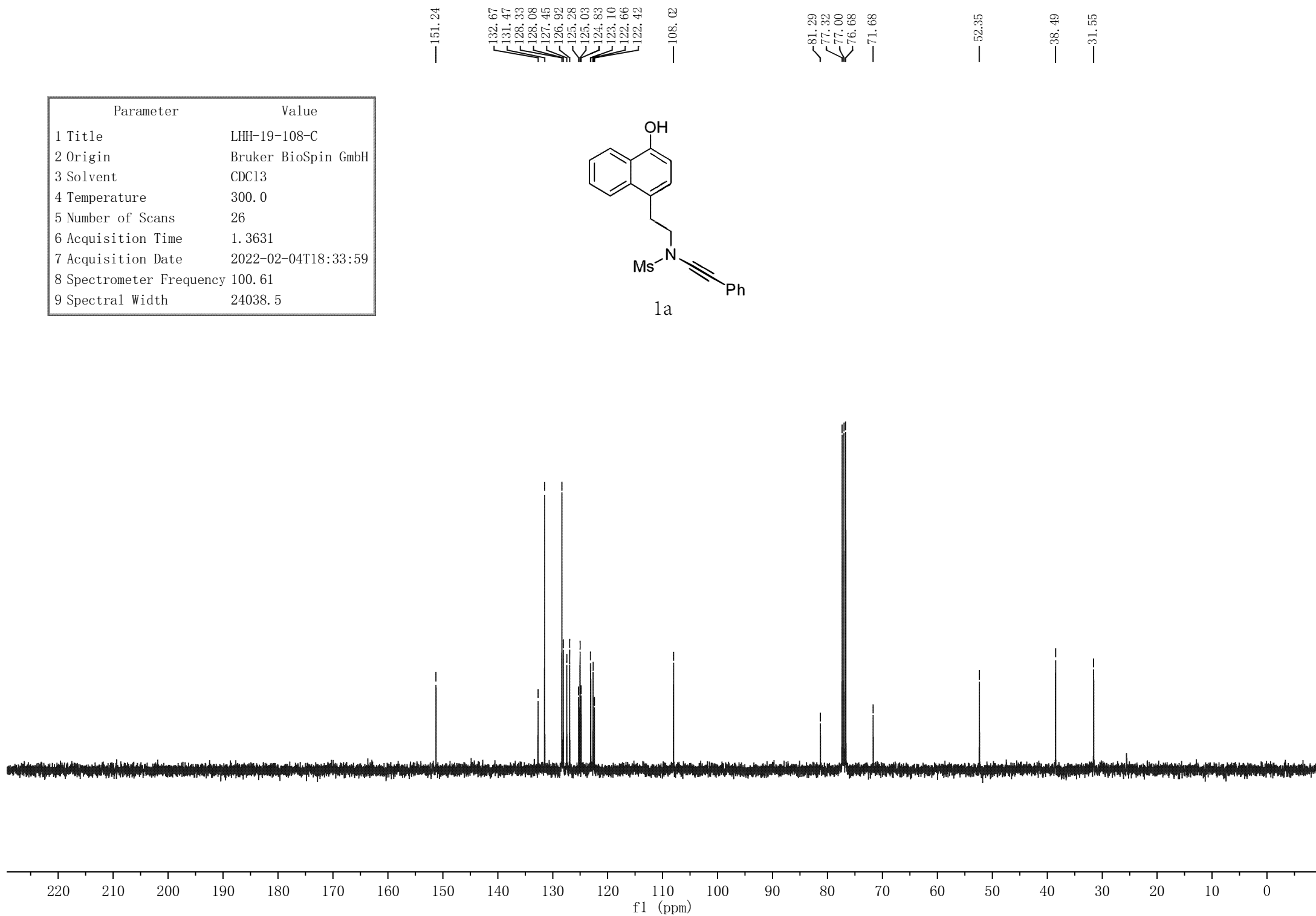
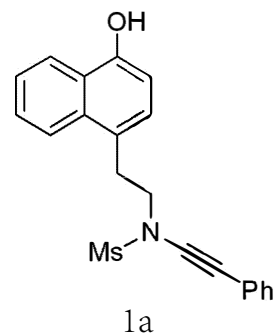
5.962

3.881
3.863
3.844
3.482
3.463
3.445

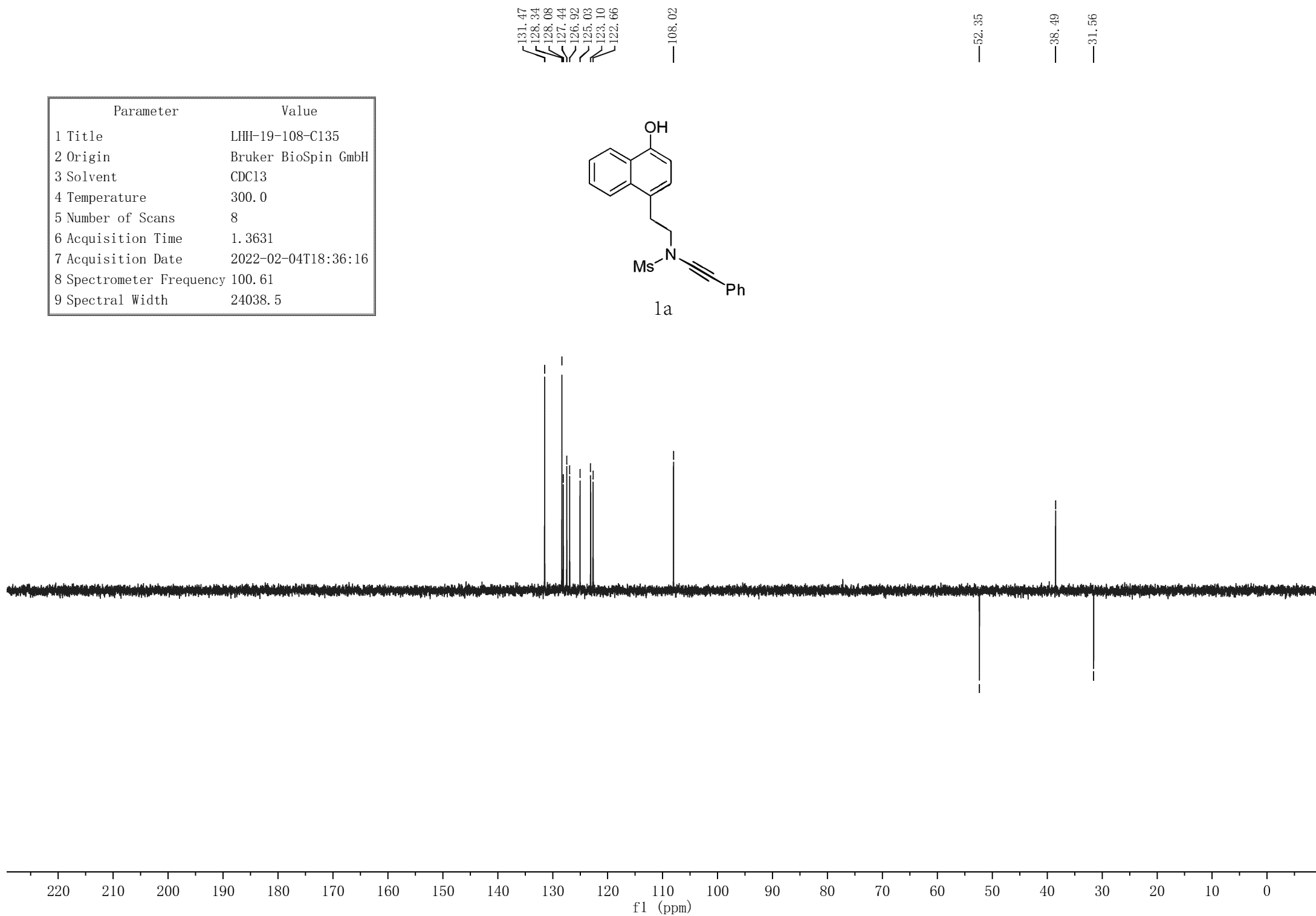
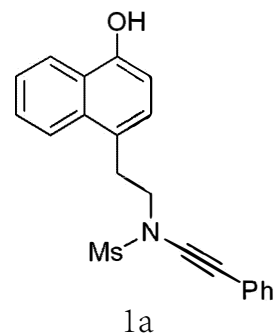
2.788



Parameter	Value
1 Title	LHH-19-108-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDCl3
4 Temperature	300.0
5 Number of Scans	26
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-04T18:33:59
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



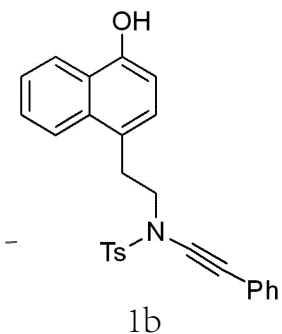
Parameter	Value
1 Title	LHH-19-108-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	8
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-04T18:36:16
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



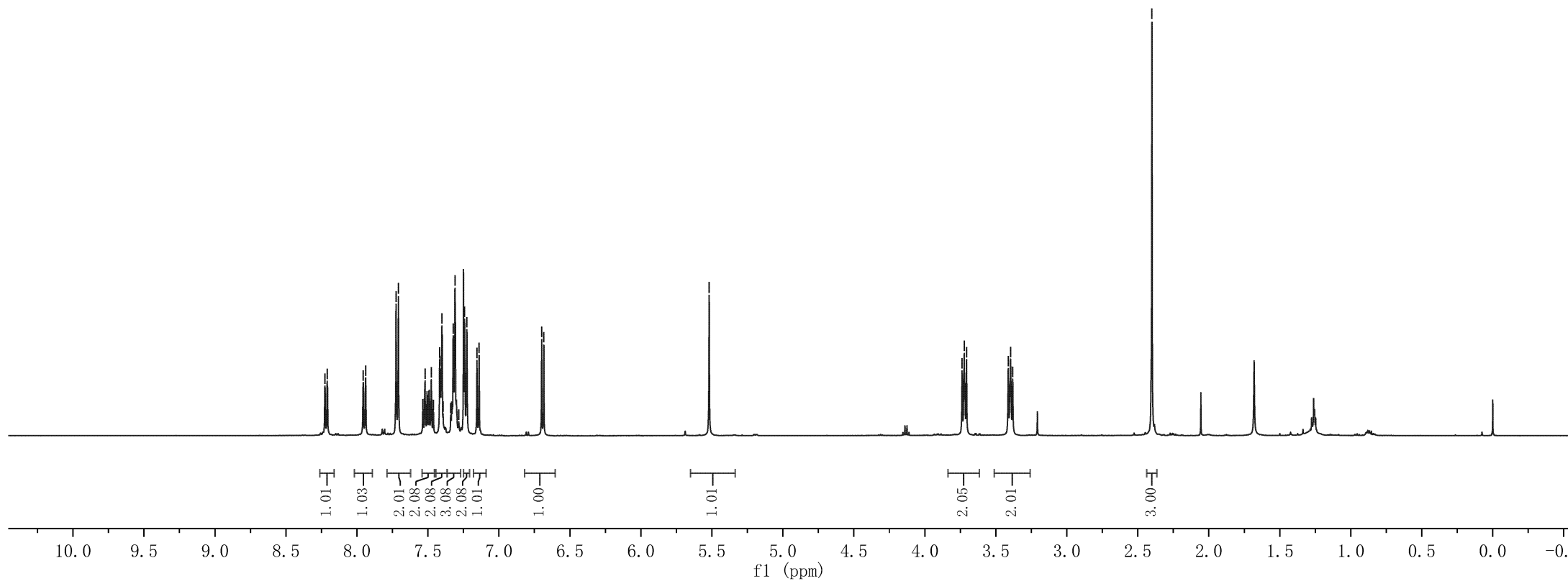
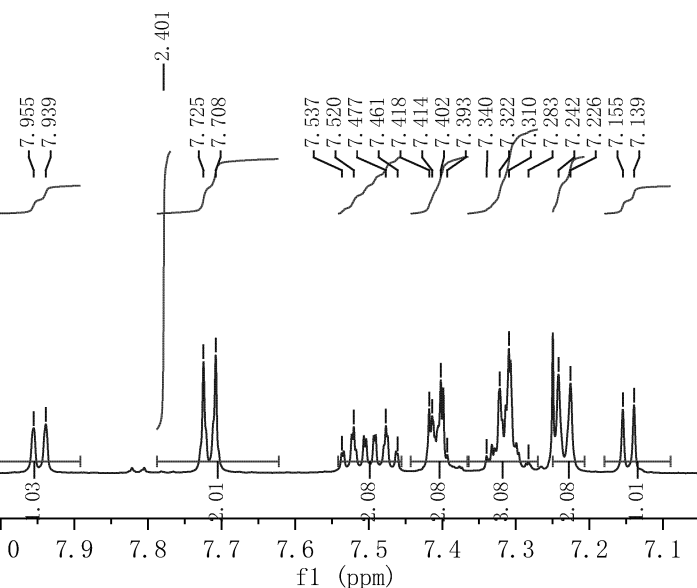
Parameter	Value
1 Title	LHH-19-Ts'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.0
5 Number of Scans	12
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-13T16:11:49
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.226
8.210
7.725
7.708
7.418
7.414
7.402
7.322
7.310
7.242
7.226
7.155
7.139
7.099
6.684

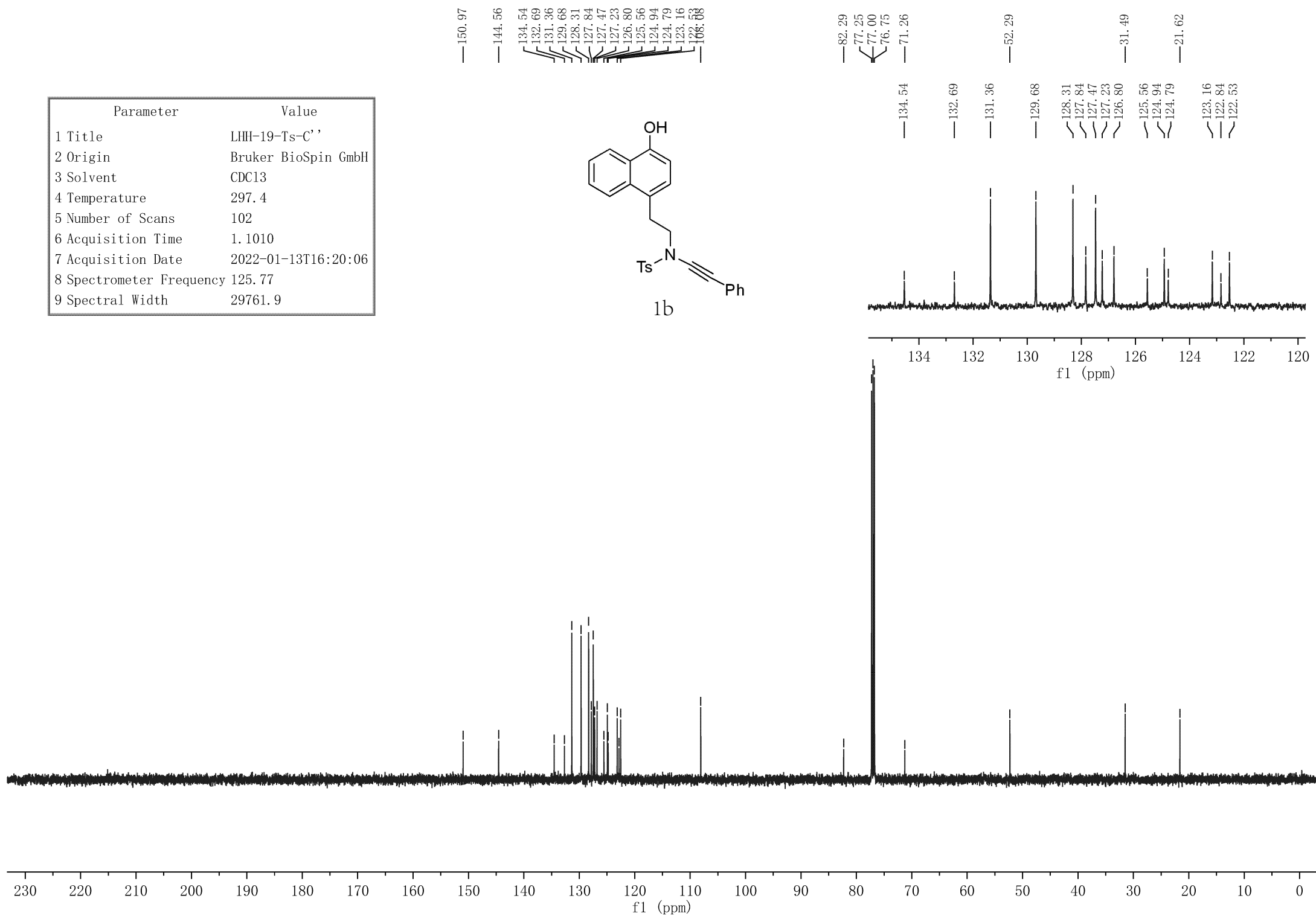
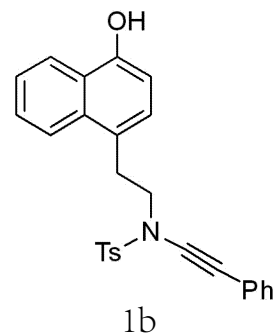
5.519



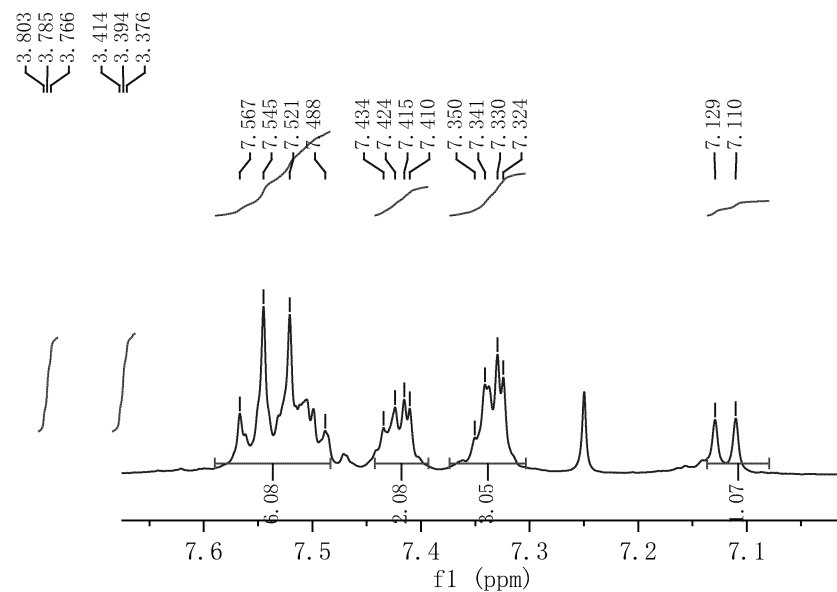
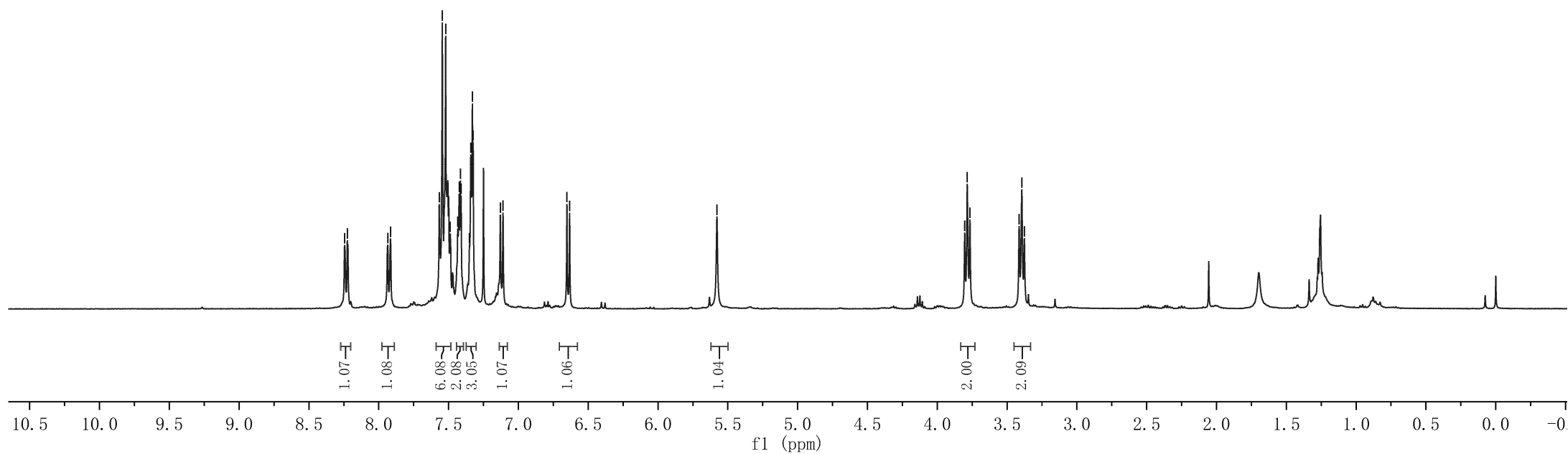
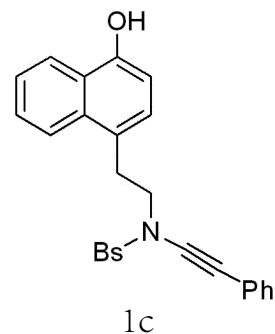
3.738
3.722
3.706
3.413
3.396
3.381



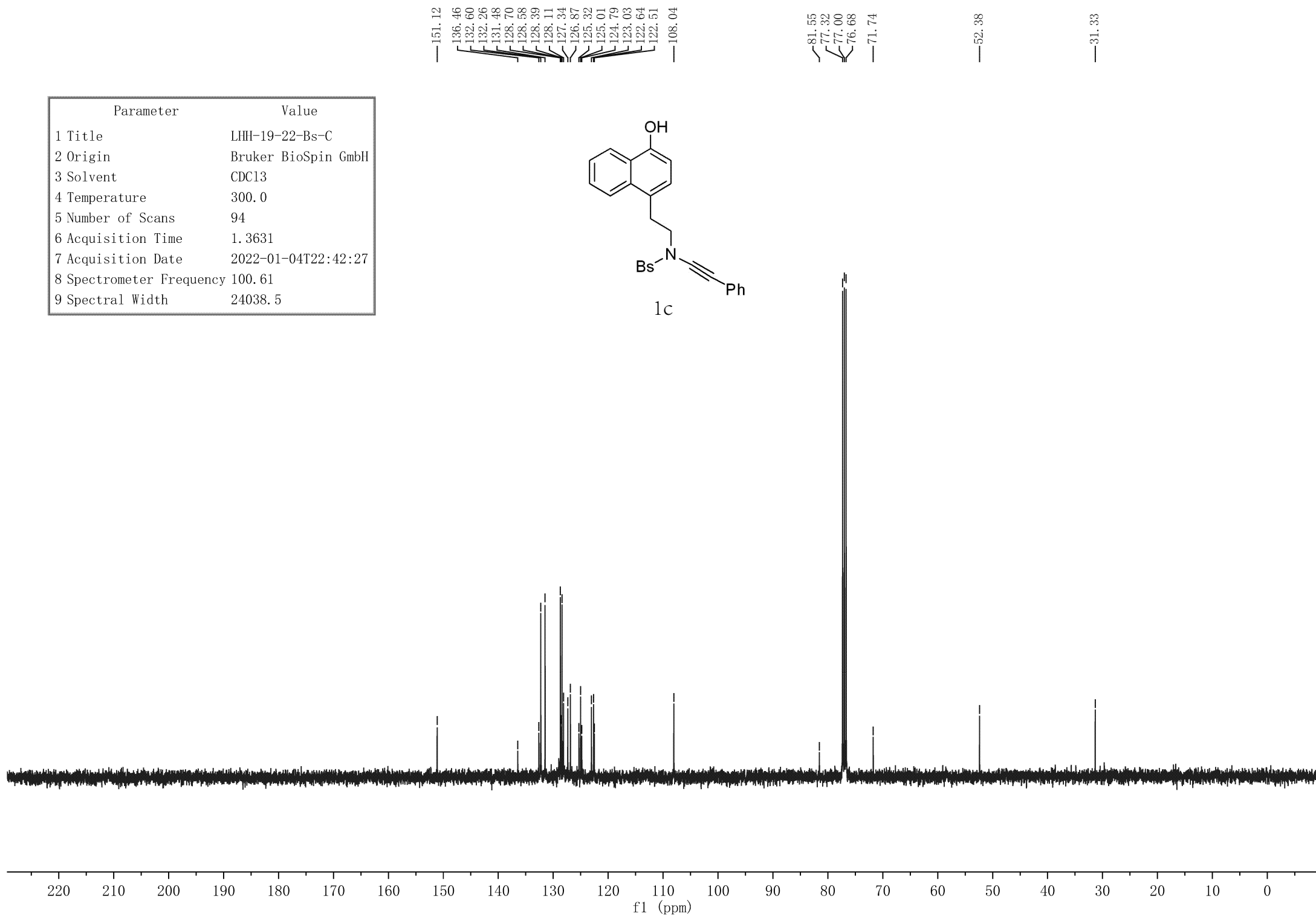
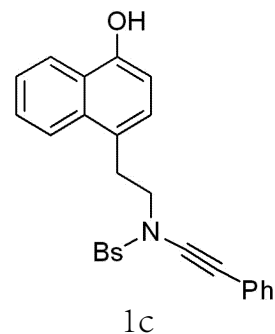
Parameter	Value
1 Title	LHH-19-Ts-C''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.4
5 Number of Scans	102
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-13T16:20:06
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



Parameter	Value
1 Title	LHH-19-22-Bs-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-04T22:40:04
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



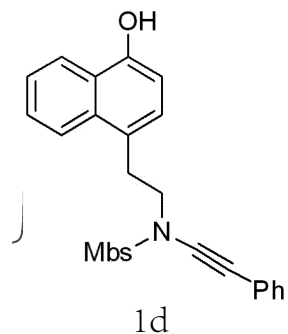
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1 Title	LHH-19-22-Bs-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	94
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-04T22:42:27
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



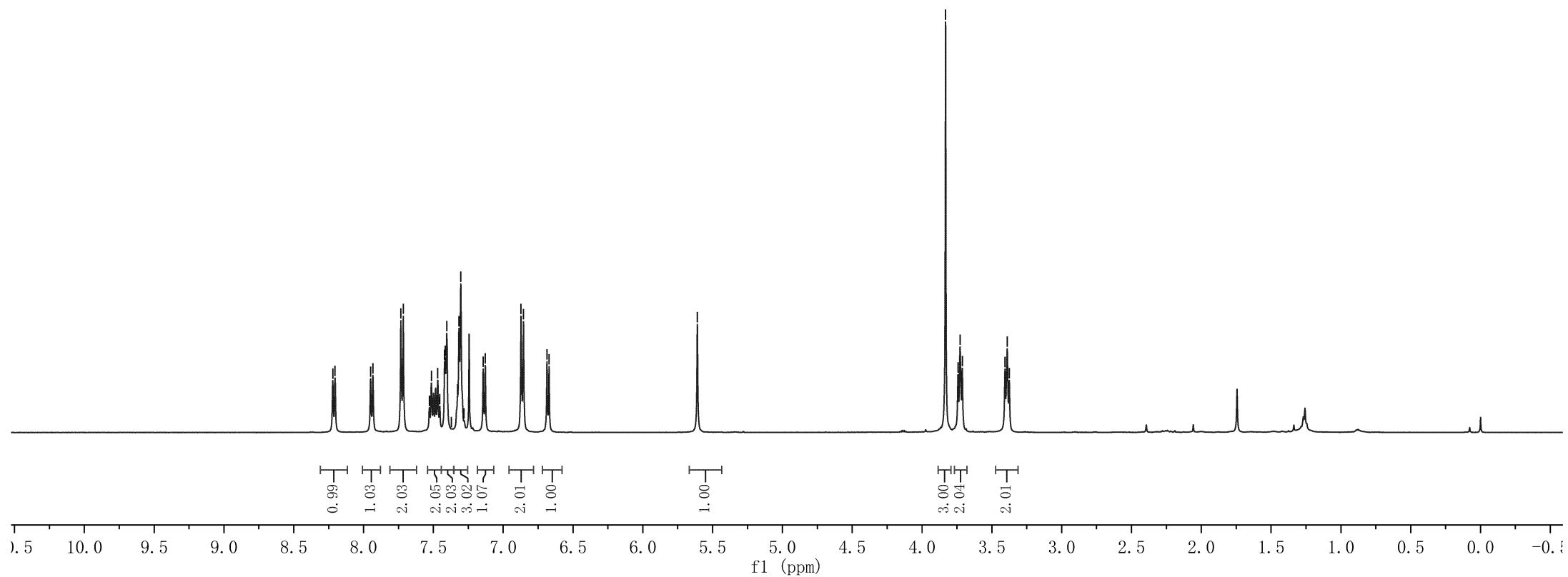
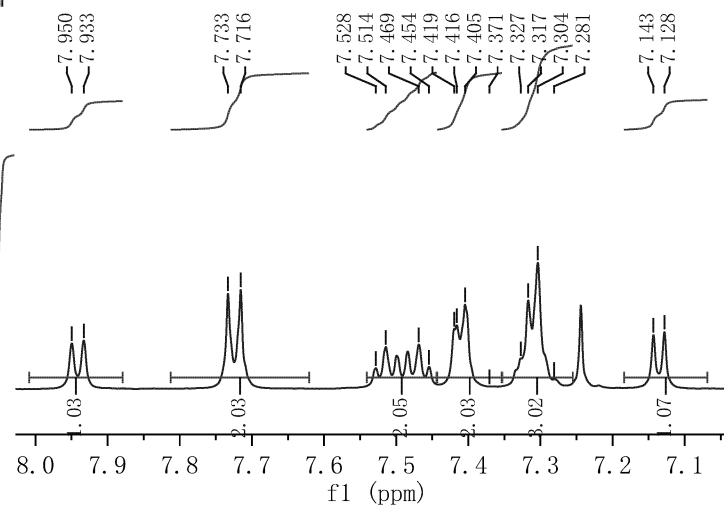
Parameter	Value
1 Title	LHH-19-MBS
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-14T16:39:06
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.221
8.204
7.733
7.716
7.419
7.416
7.405
7.317
7.304
7.143
6.898
6.855
6.686
6.671

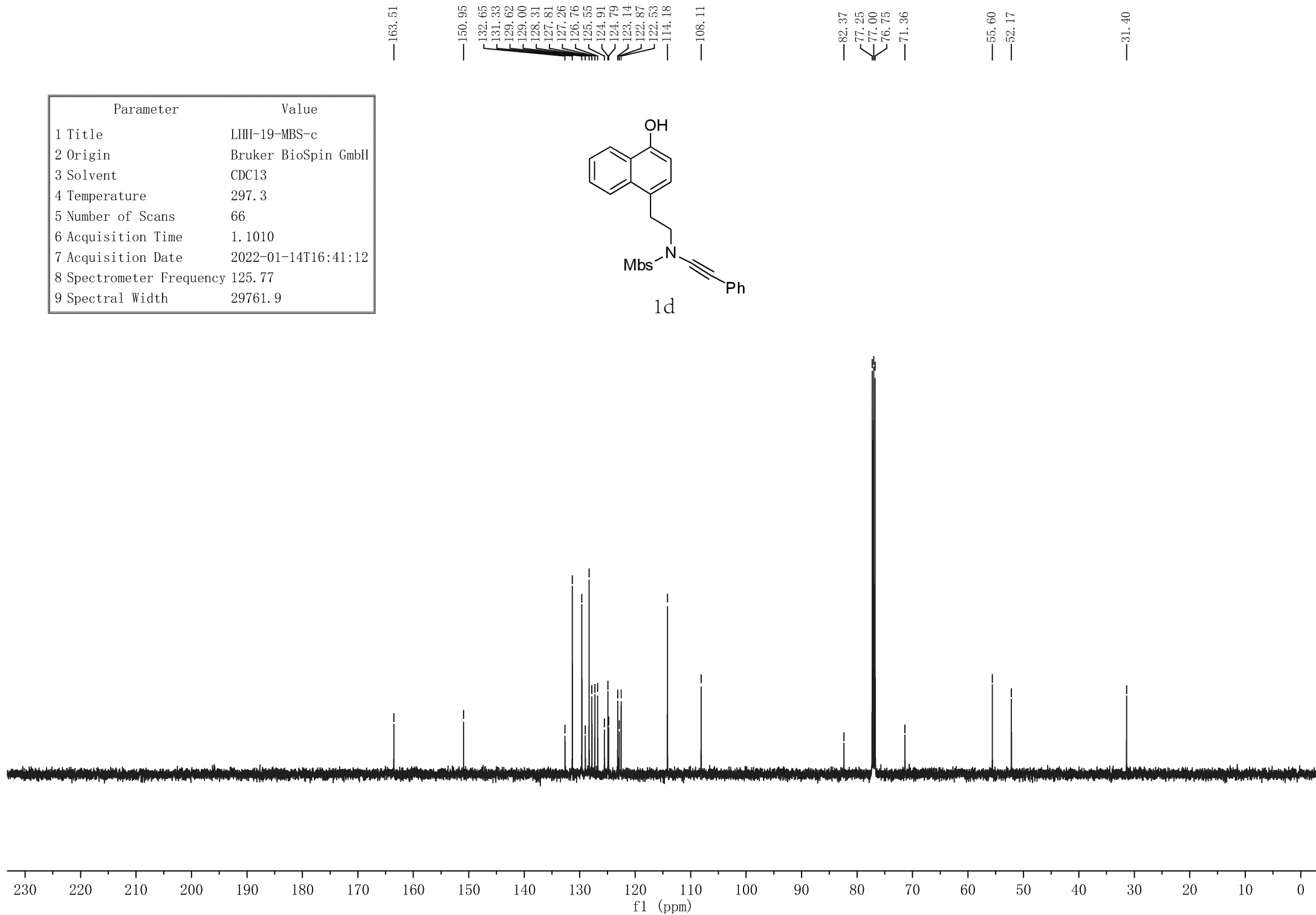
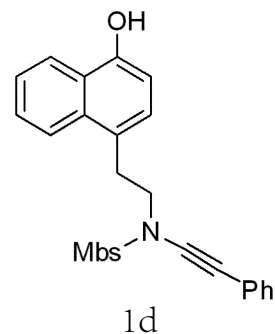
5.609



3.831
3.743
3.728
3.712
3.406
3.390
3.375



Parameter	Value
1 Title	LHH-19-MBS-c
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	66
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-14T16:41:12
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



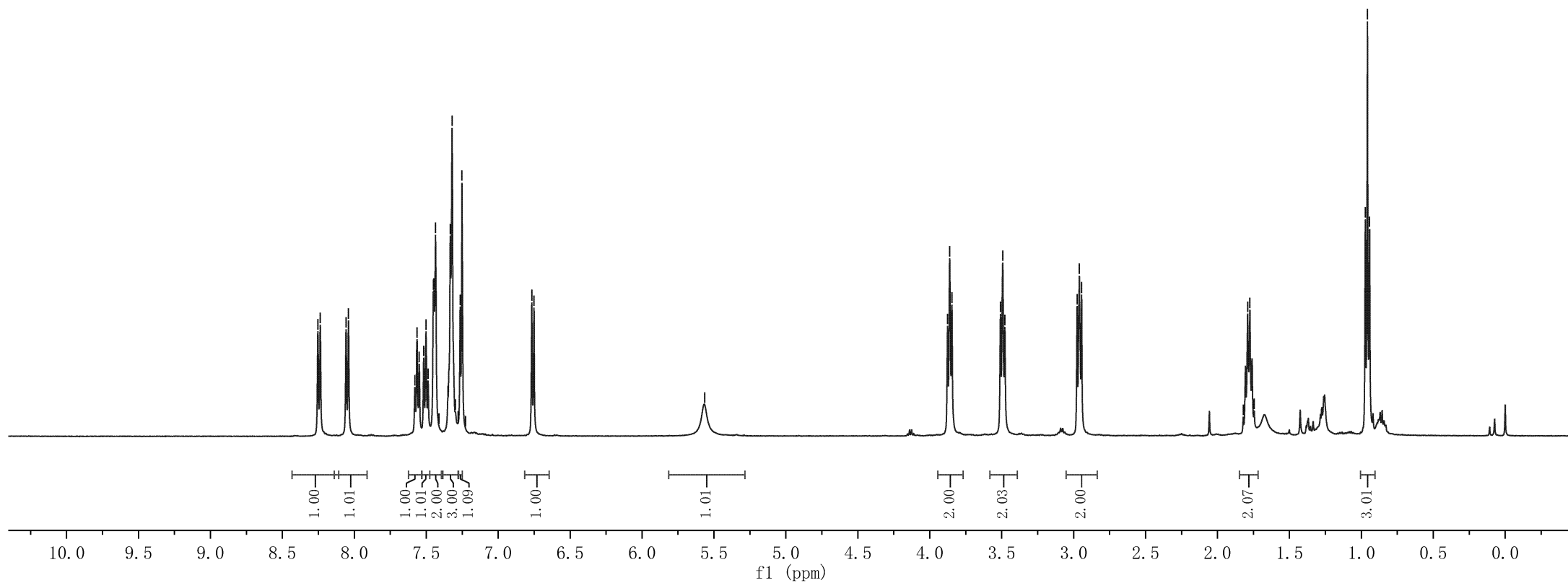
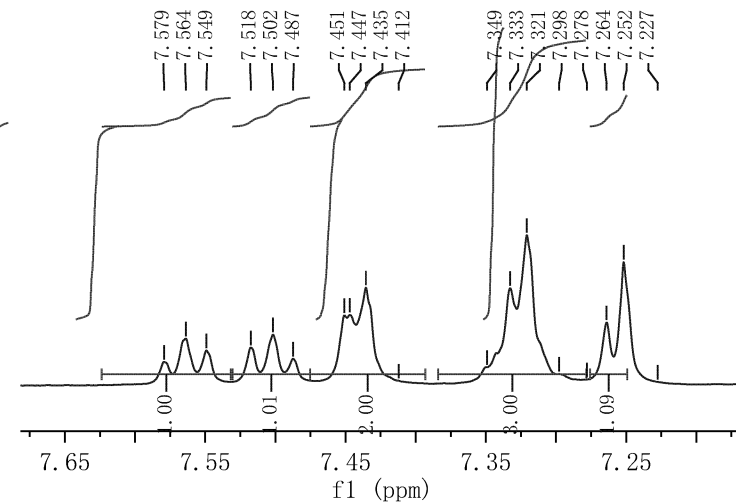
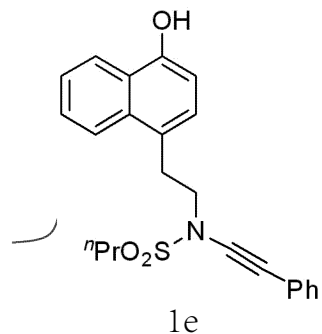
Parameter	Value
1 Title	LHH-19-77
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.8
5 Number of Scans	6
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-19T16:12:25
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.253
8.237
8.057
8.040
7.564
7.502
7.451
7.447
7.435
7.333
7.321
7.264
7.252
7.250
7.251
6.751

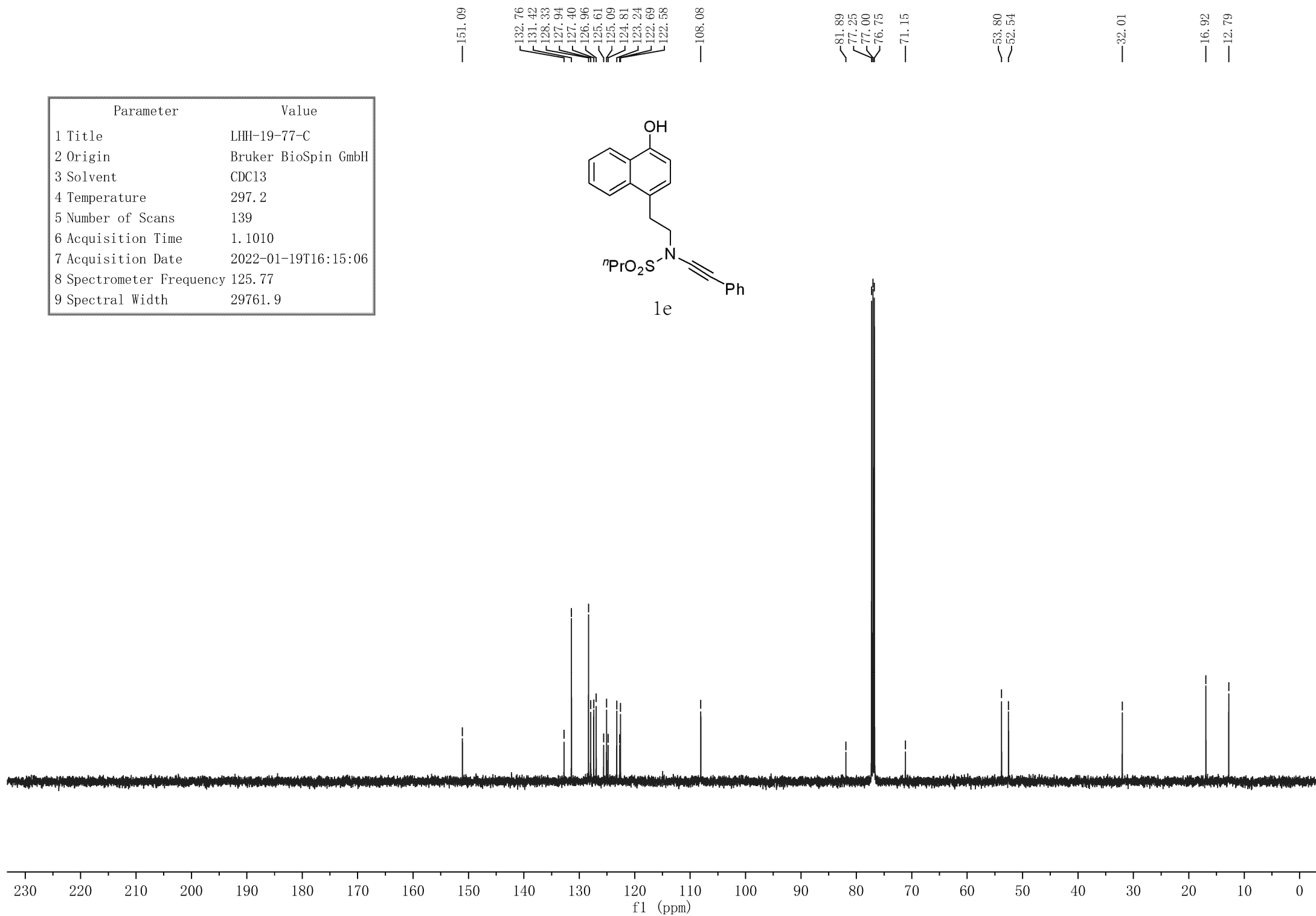
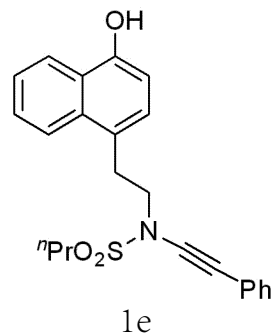
5.564

3.876
3.862
3.846
3.508
3.493
3.478
2.976
2.960
2.944

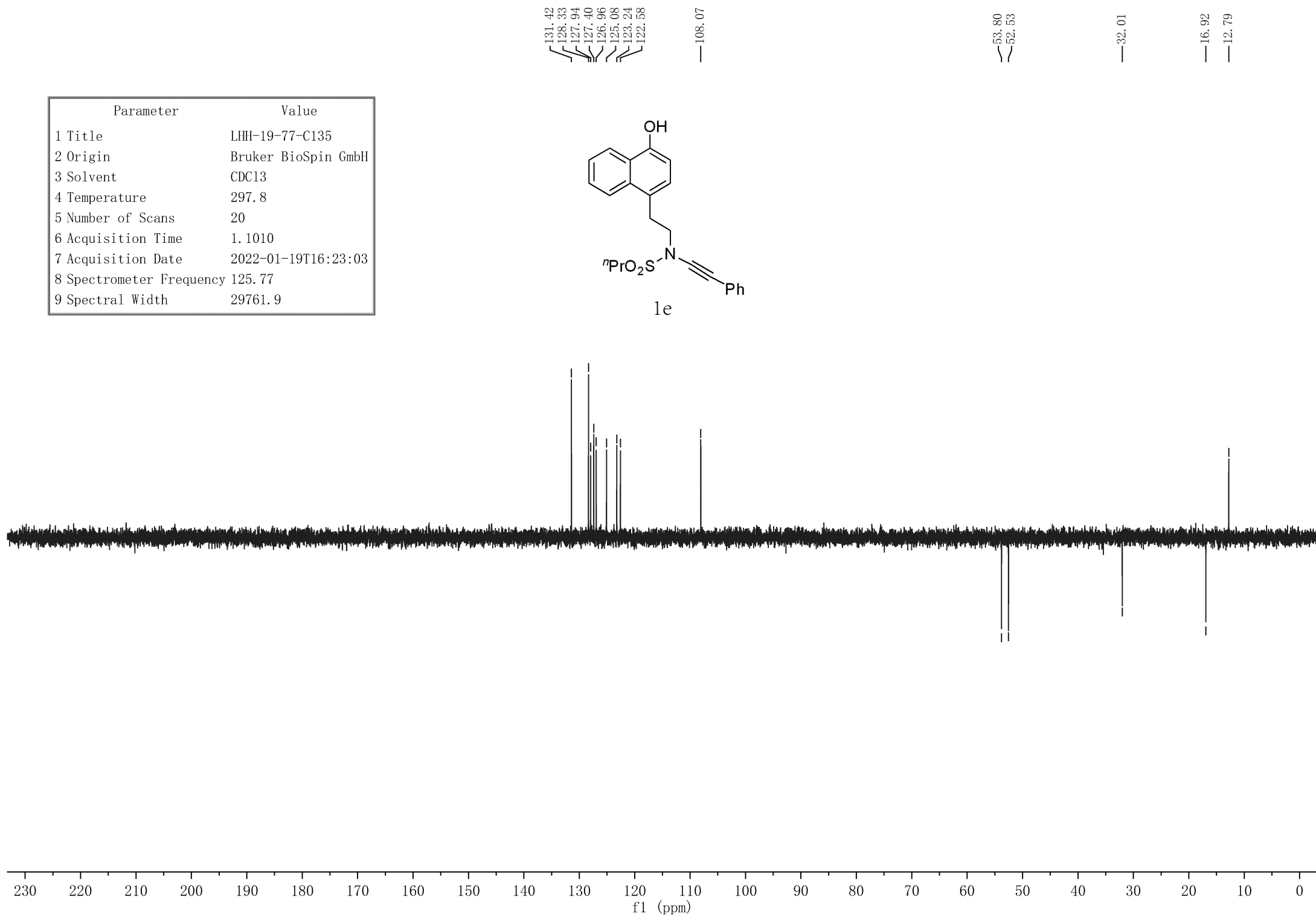
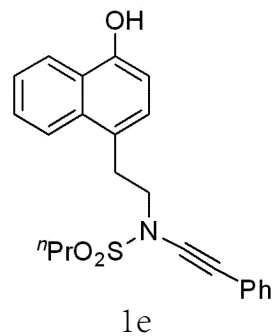
1.821
1.790
1.775
1.745
0.972
0.958
0.943



Parameter	Value
1 Title	LHH-19-77-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.2
5 Number of Scans	139
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-19T16:15:06
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



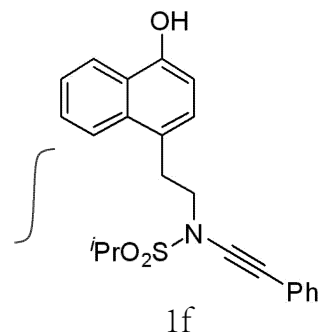
Parameter	Value
1 Title	LHH-19-77-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.8
5 Number of Scans	20
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-19T16:23:03
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



Parameter	Value
1 Title	LHH-19-85
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-21T19:26:57
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

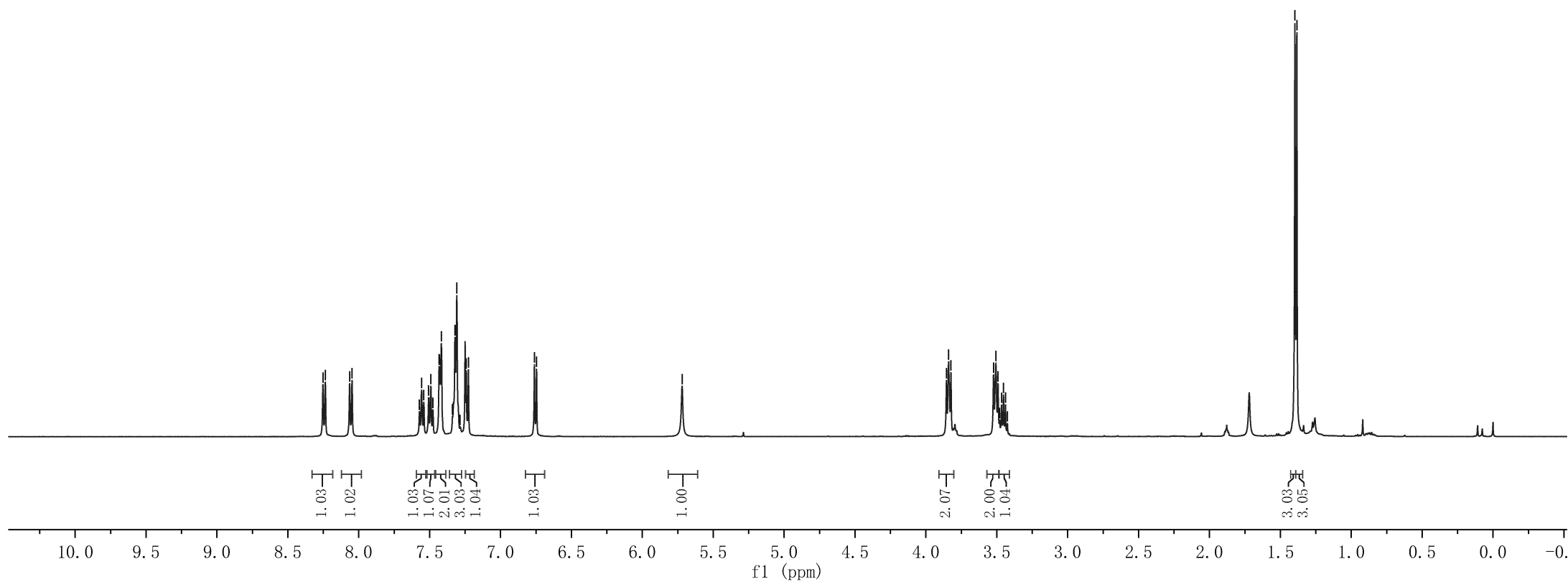
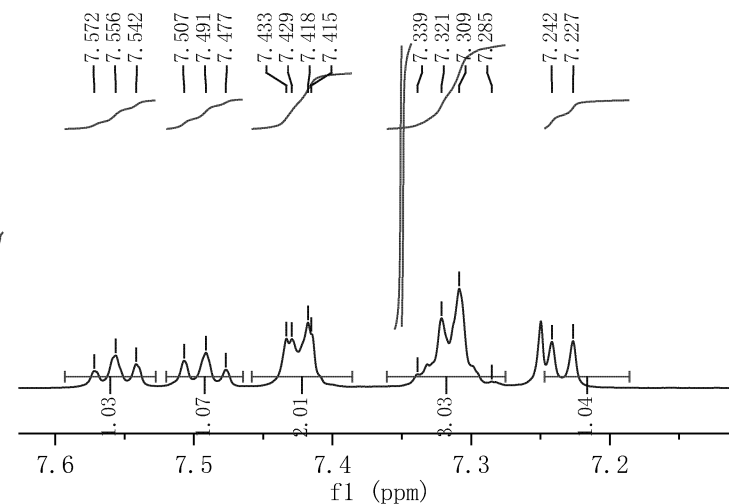
8.252
8.236
8.064
8.047
7.491
7.433
7.429
7.418
7.415
7.321
7.309
7.242
7.227
6.761
6.746

5.719

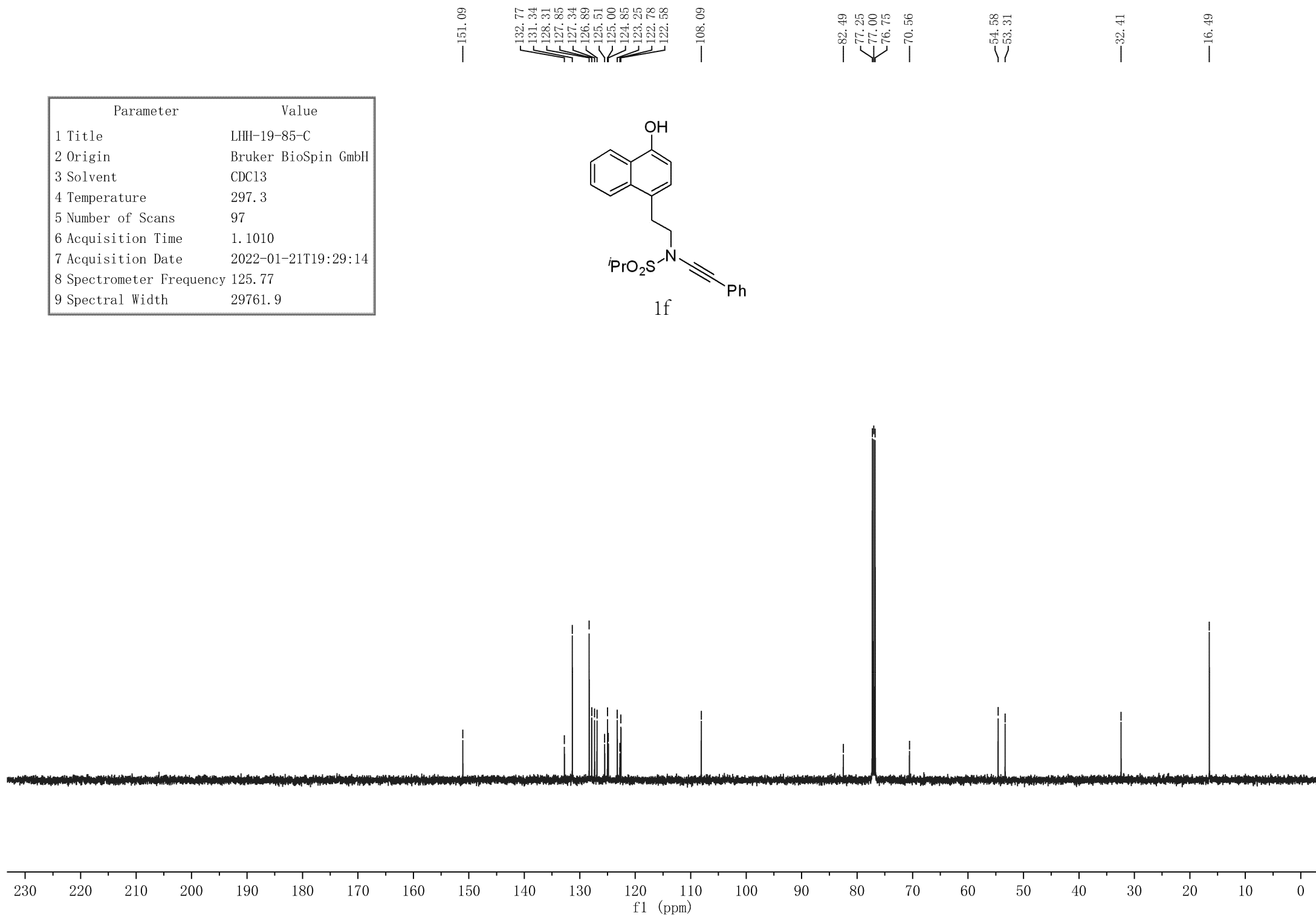
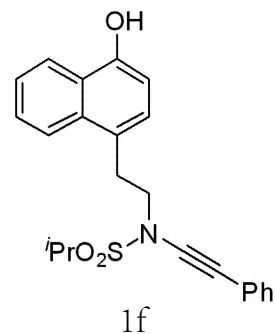


3.855
3.840
3.823
3.523
3.506
3.491
3.480
3.466
3.452
3.439
3.425

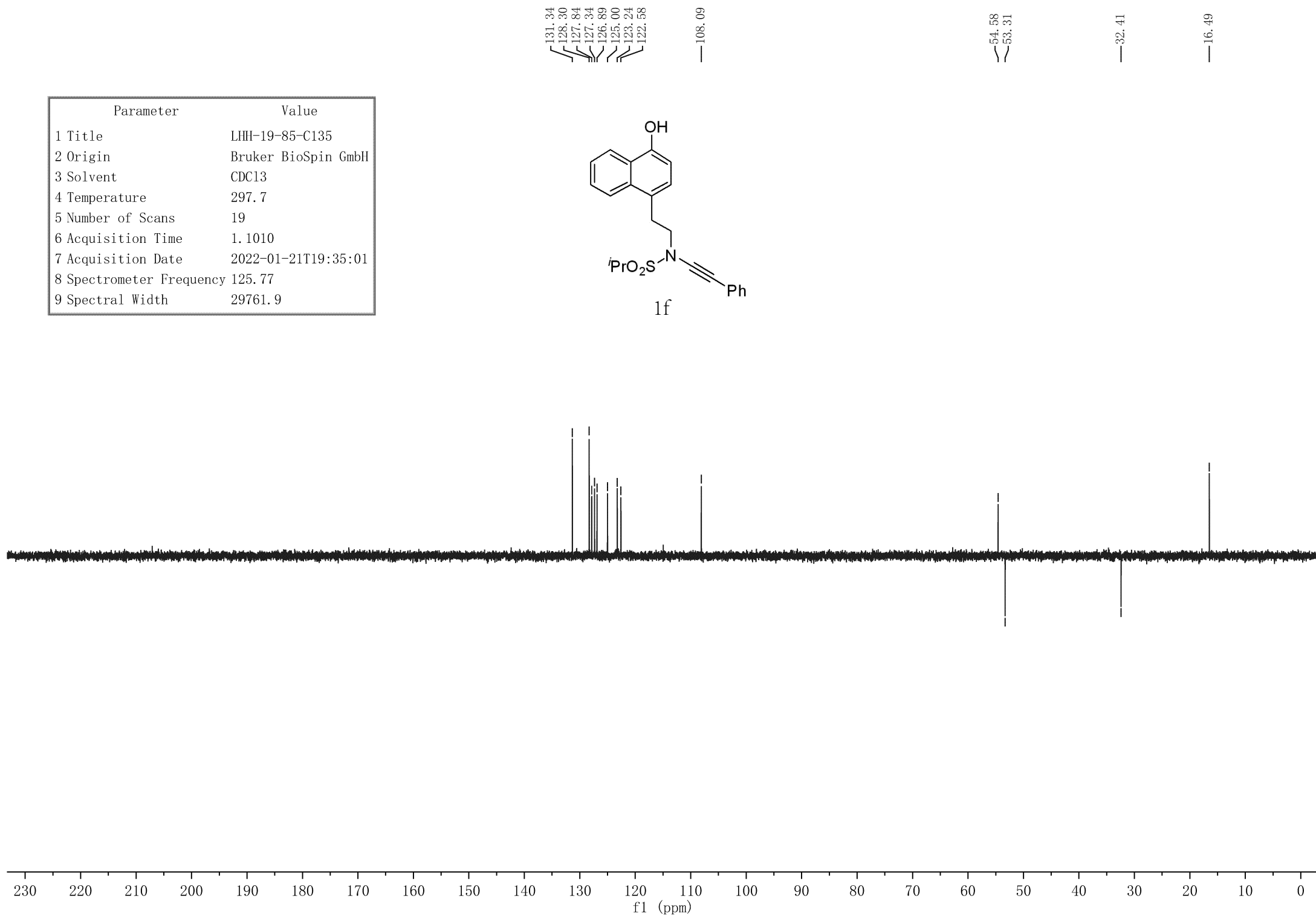
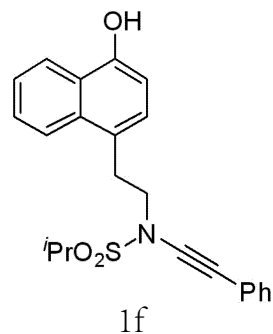
1.397
1.384



Parameter	Value
1 Title	LHH-19-85-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	97
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-21T19:29:14
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



Parameter	Value
1 Title	LHH-19-85-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.7
5 Number of Scans	19
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-21T19:35:01
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



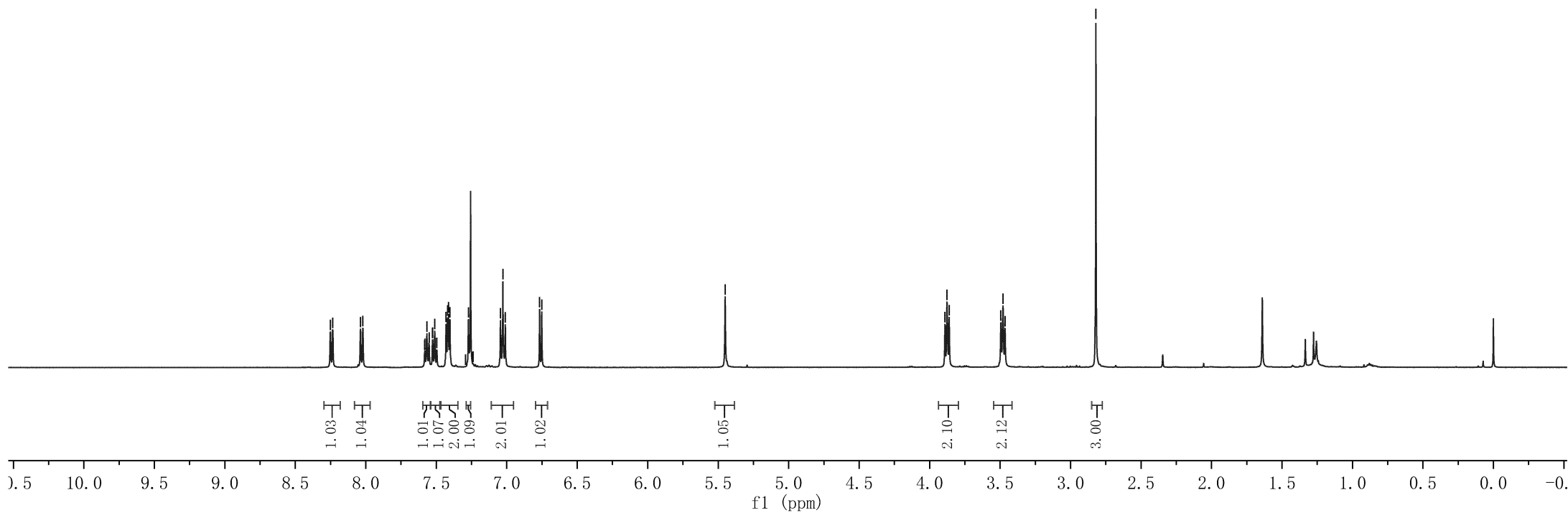
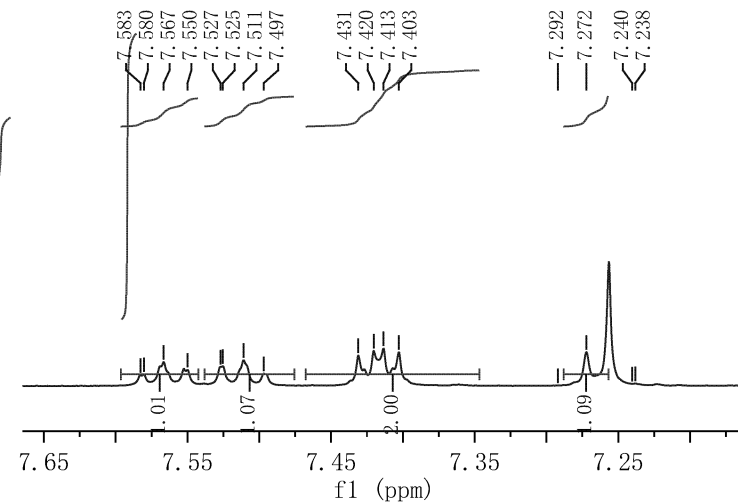
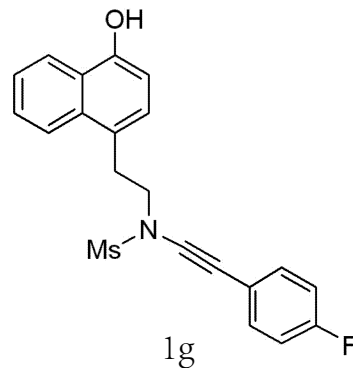
Parameter	Value
1 Title	LHH-19-105
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.8
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-29T17:22:02
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.251
8.235
8.038
8.021
7.511
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7.420
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7.403
7.272
7.045
7.028
6.969
6.969
6.752

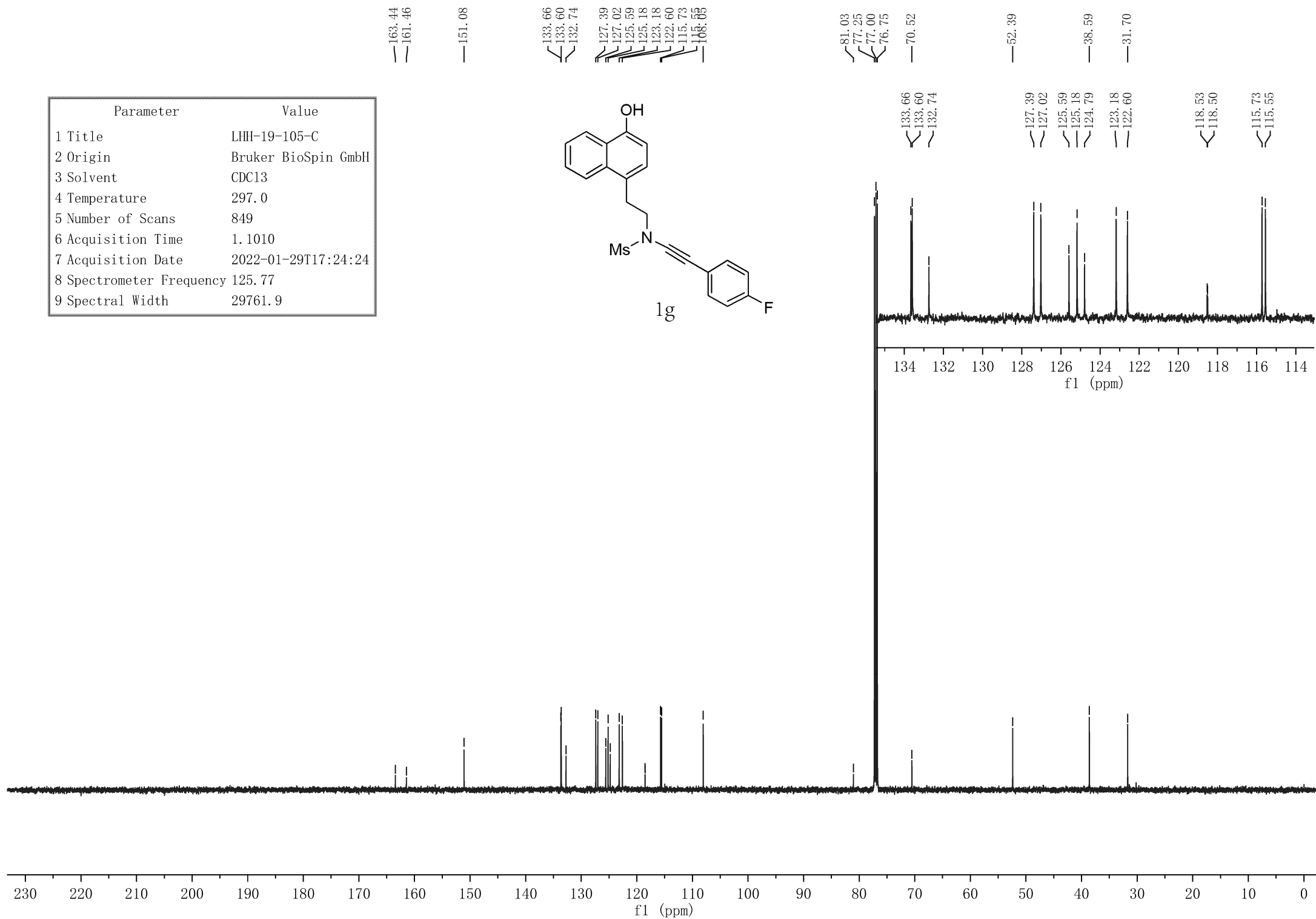
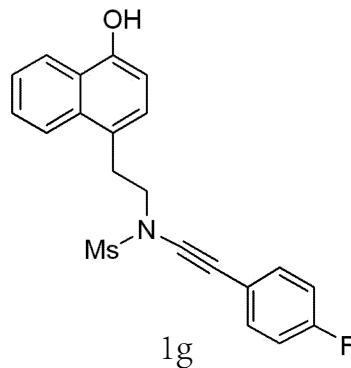
5.451

3.891
3.877
3.861
3.495
3.479
3.465

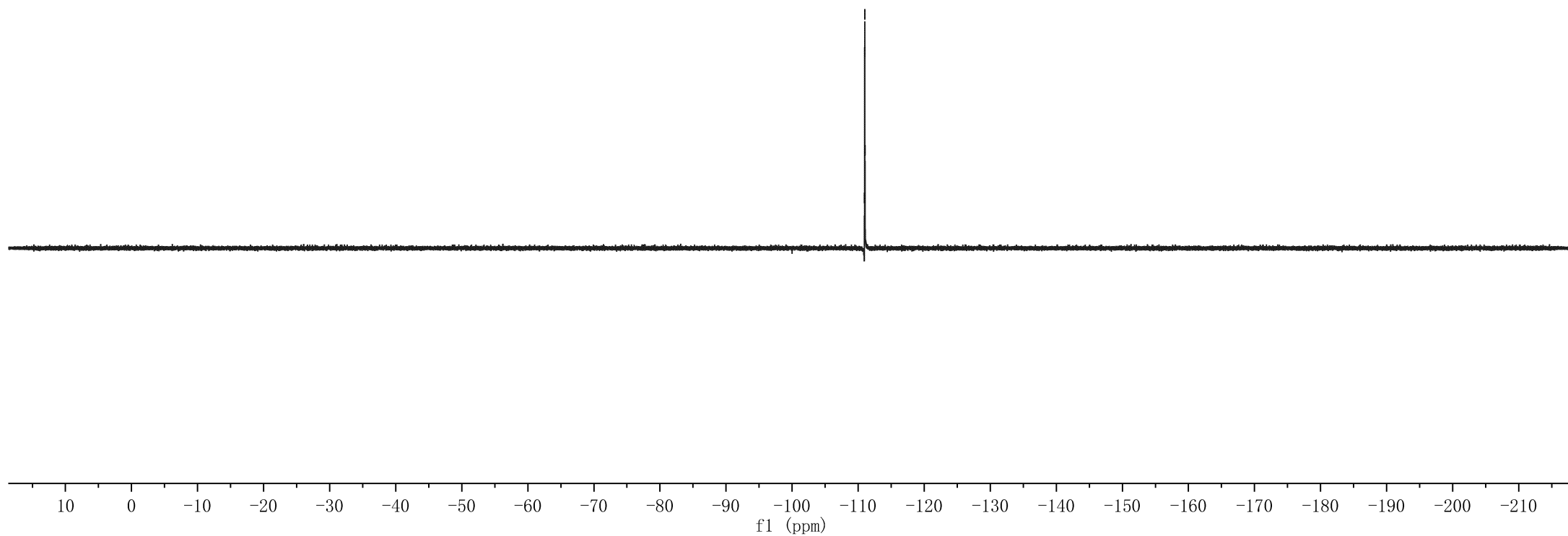
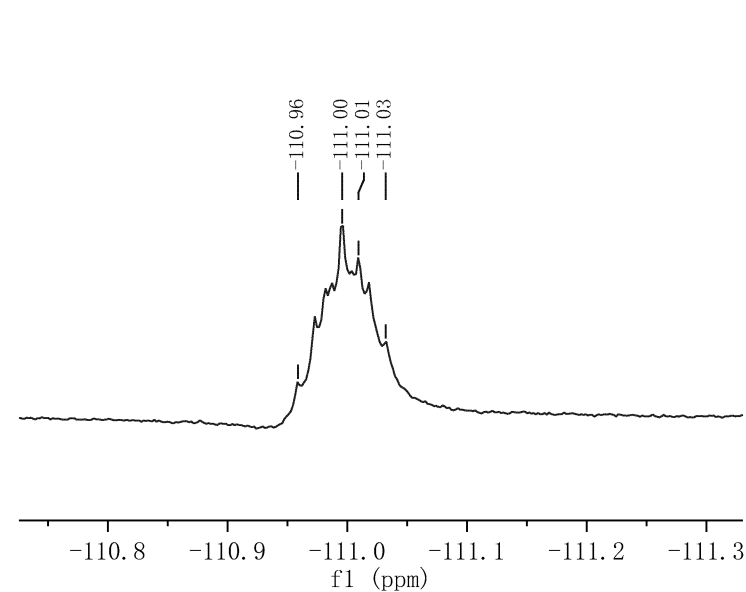
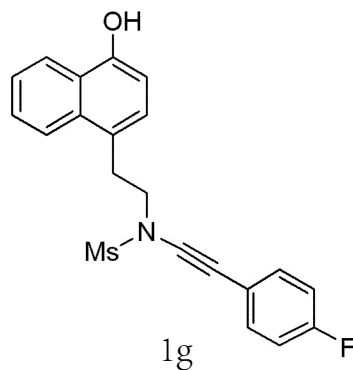
2.820



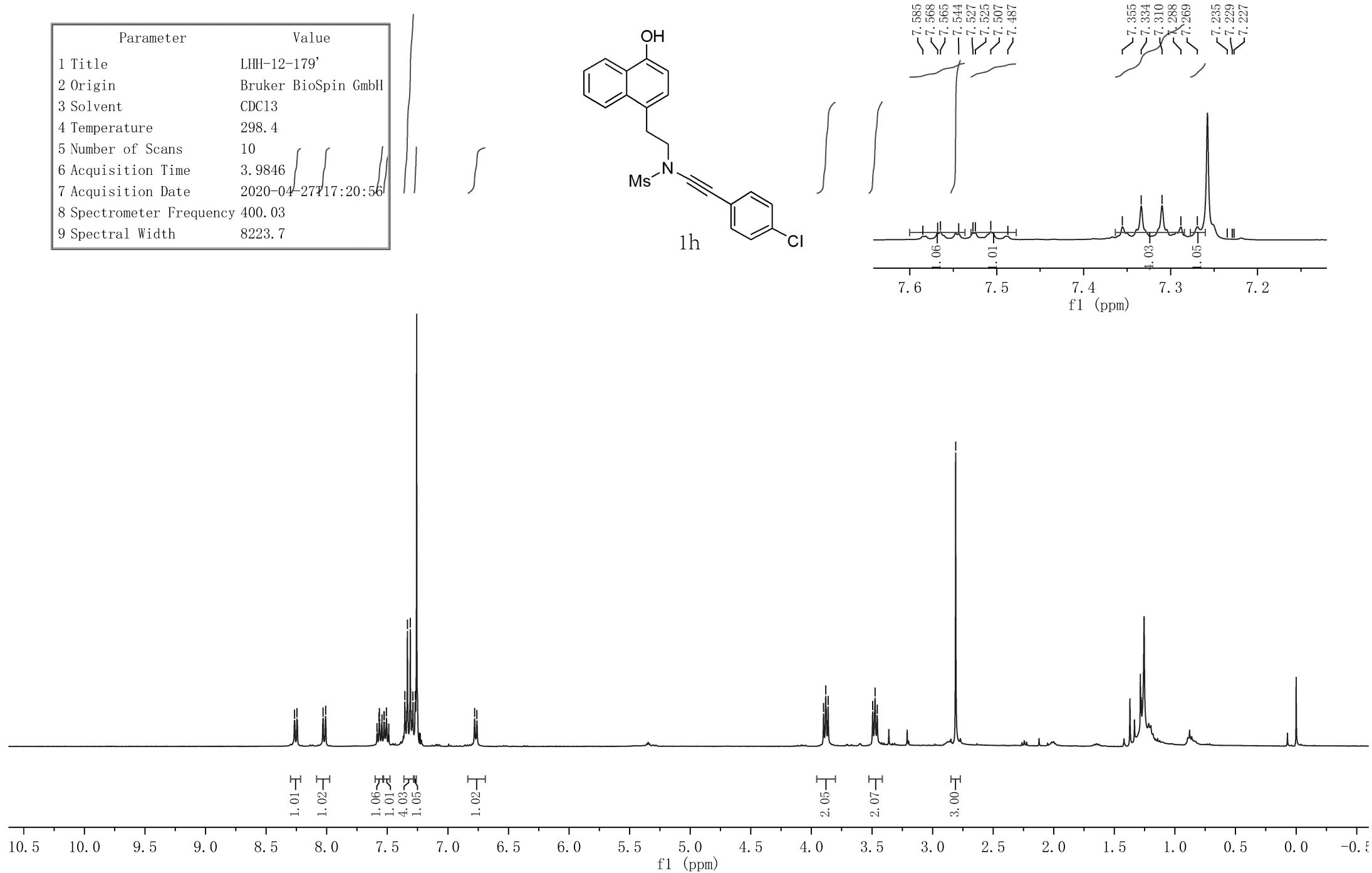
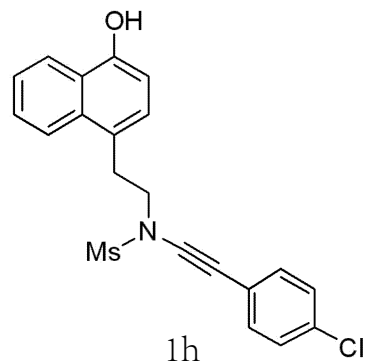
Parameter	Value
1 Title	LHH-19-105-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.0
5 Number of Scans	849
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-29T17:24:24
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



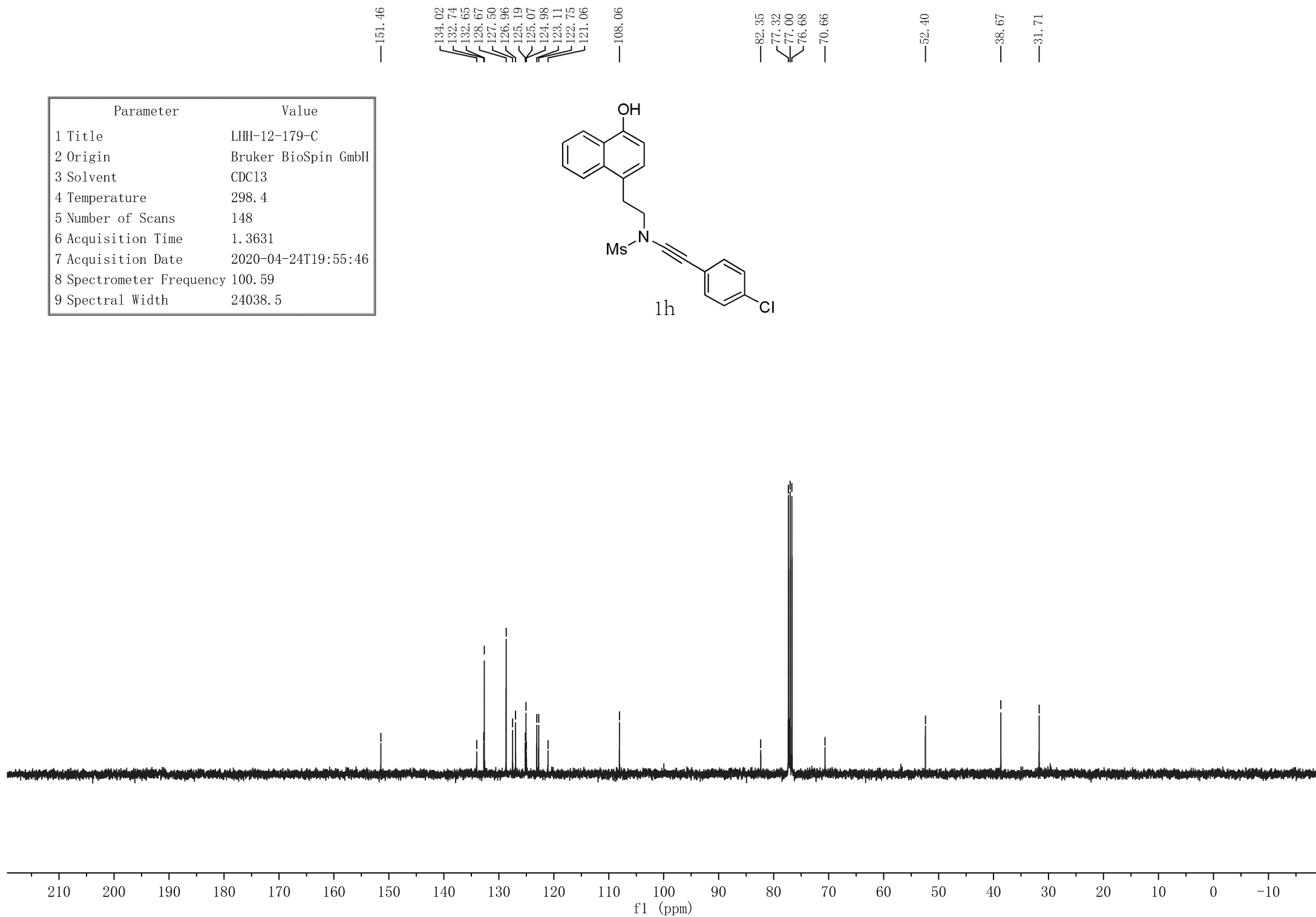
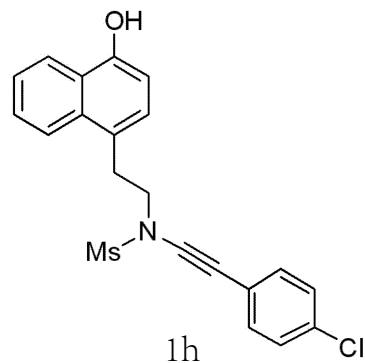
Parameter	Value
1 Title	LHH-19-34-F
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.3
5 Number of Scans	32
6 Acquisition Time	0.7340
7 Acquisition Date	2022-01-07T17:41:49
8 Spectrometer Frequency	376.31
9 Spectral Width	89285.7



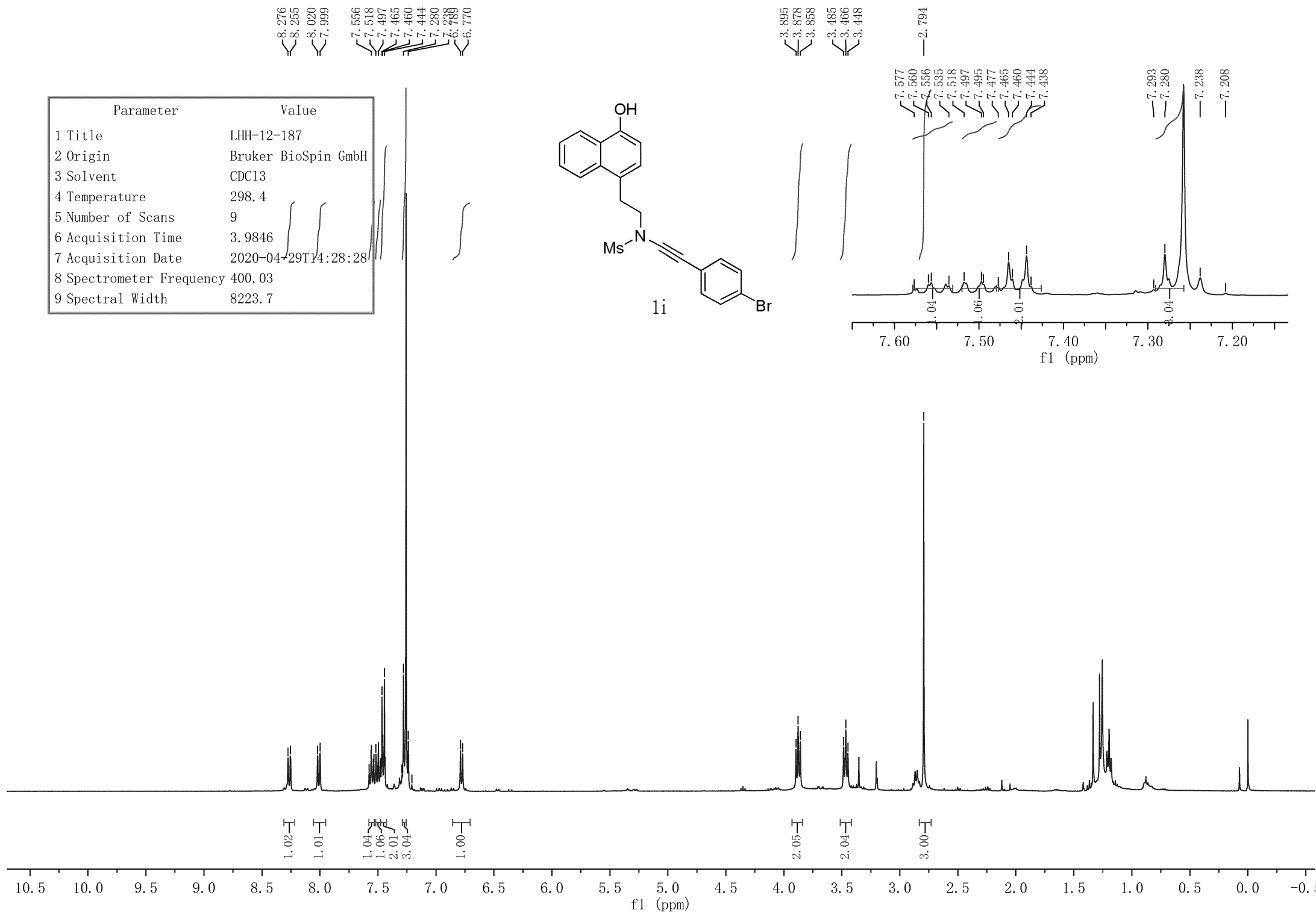
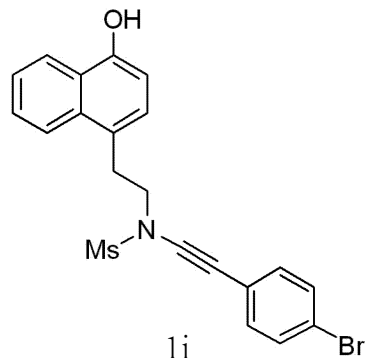
Parameter	Value
1 Title	LHH-12-179'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.4
5 Number of Scans	10
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-27T17:20:56
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7



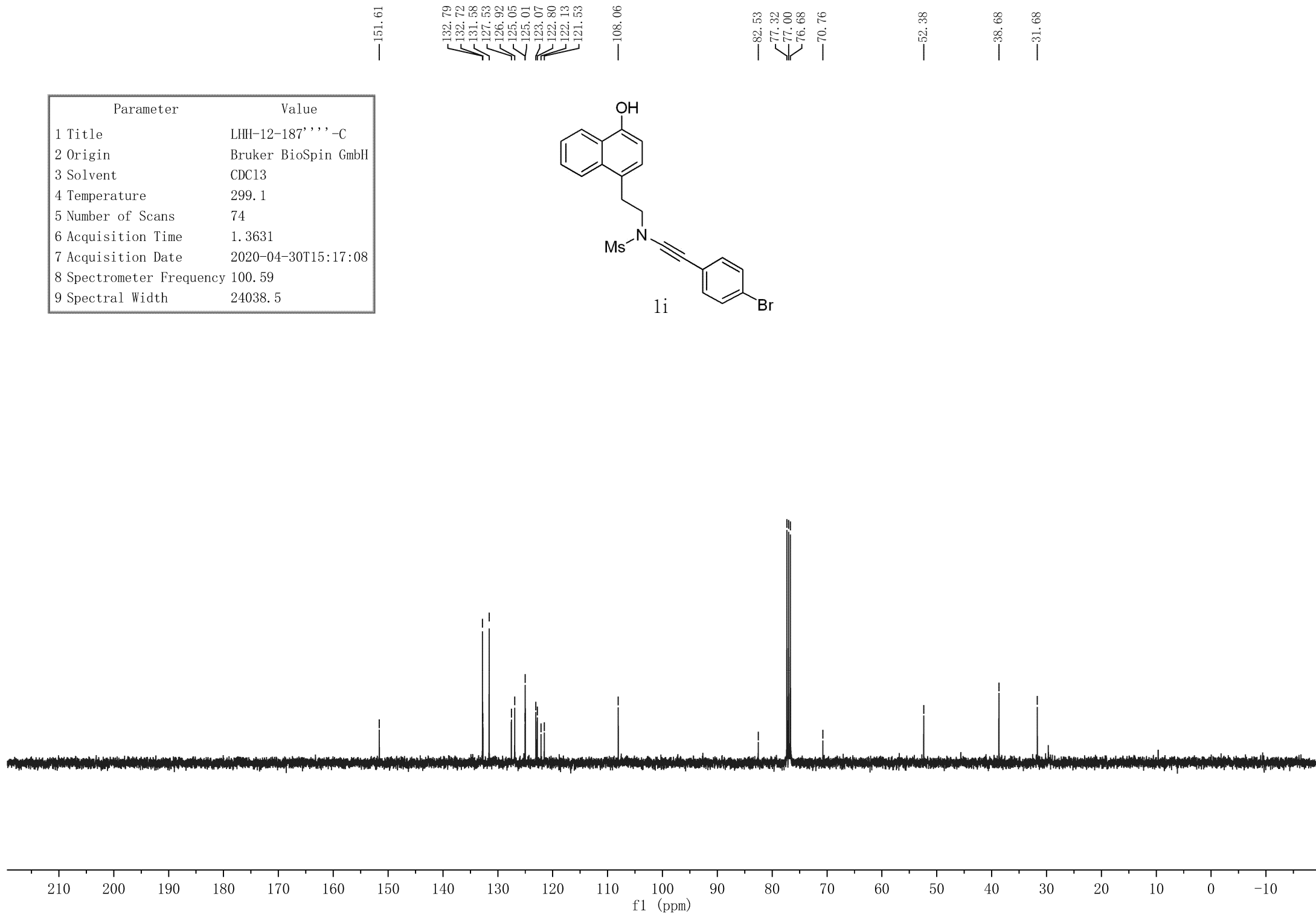
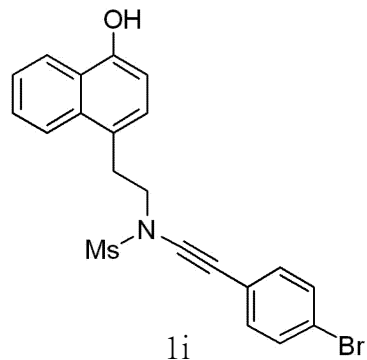
Parameter	Value
1 Title	LHH-12-179-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.4
5 Number of Scans	148
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-24T19:55:46
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-12-187
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.4
5 Number of Scans	9
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-29T14:28:28
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7



Parameter	Value
1 Title	LHH-12-187''''-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	299.1
5 Number of Scans	74
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-30T15:17:08
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-70
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.4
5 Number of Scans	5
6 Acquisition Time	3.9846
7 Acquisition Date	2022-01-18T12:37:20
8 Spectrometer Frequency	399.93
9 Spectral Width	8223.7

8.249
8.229
8.032
8.011
7.547
7.543
7.482
7.362
7.342
7.239
7.223
7.141
7.121
6.748
6.729

5.807

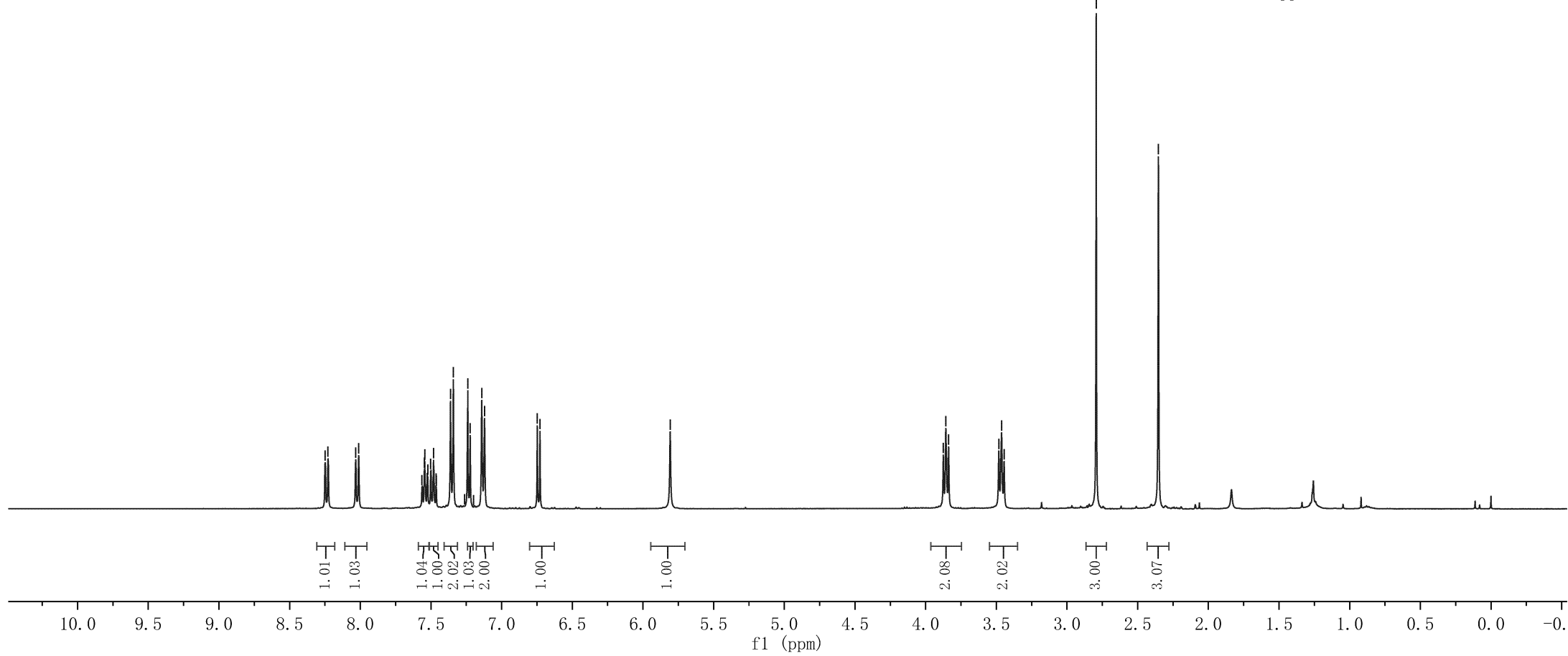
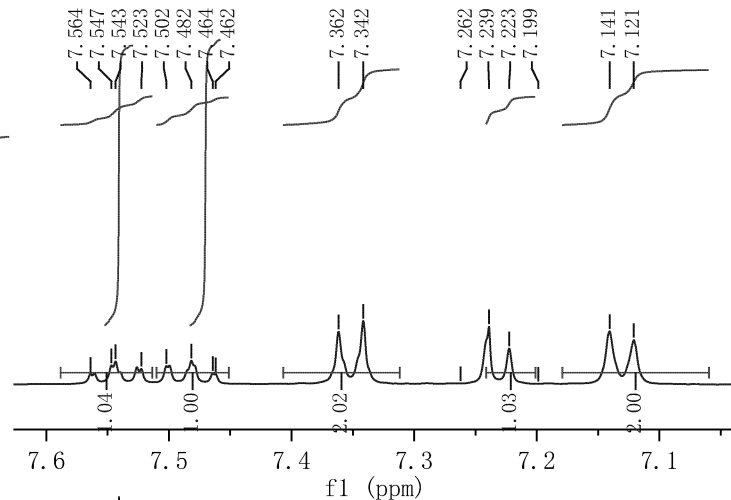
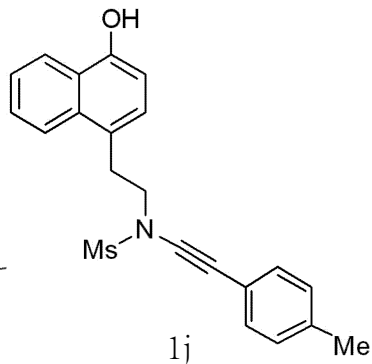
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3.837
3.483
3.463
3.445

2.793
2.353

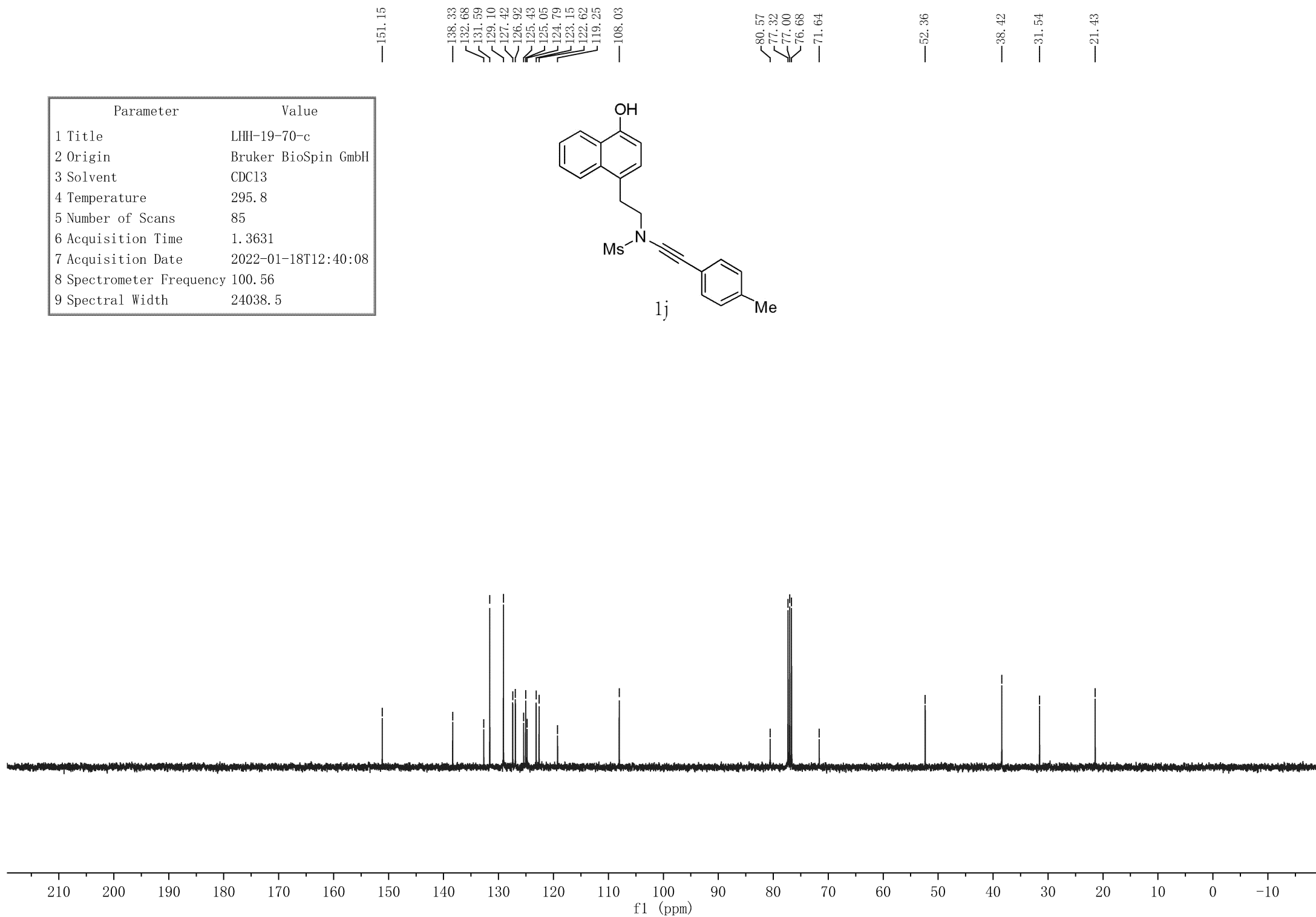
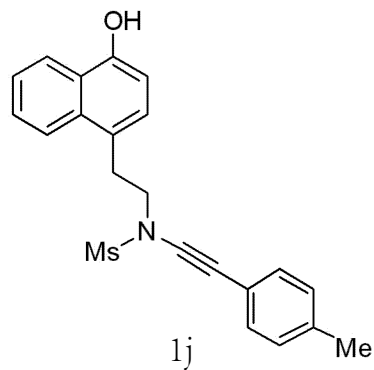
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7.342

7.262
7.239
7.223
7.199

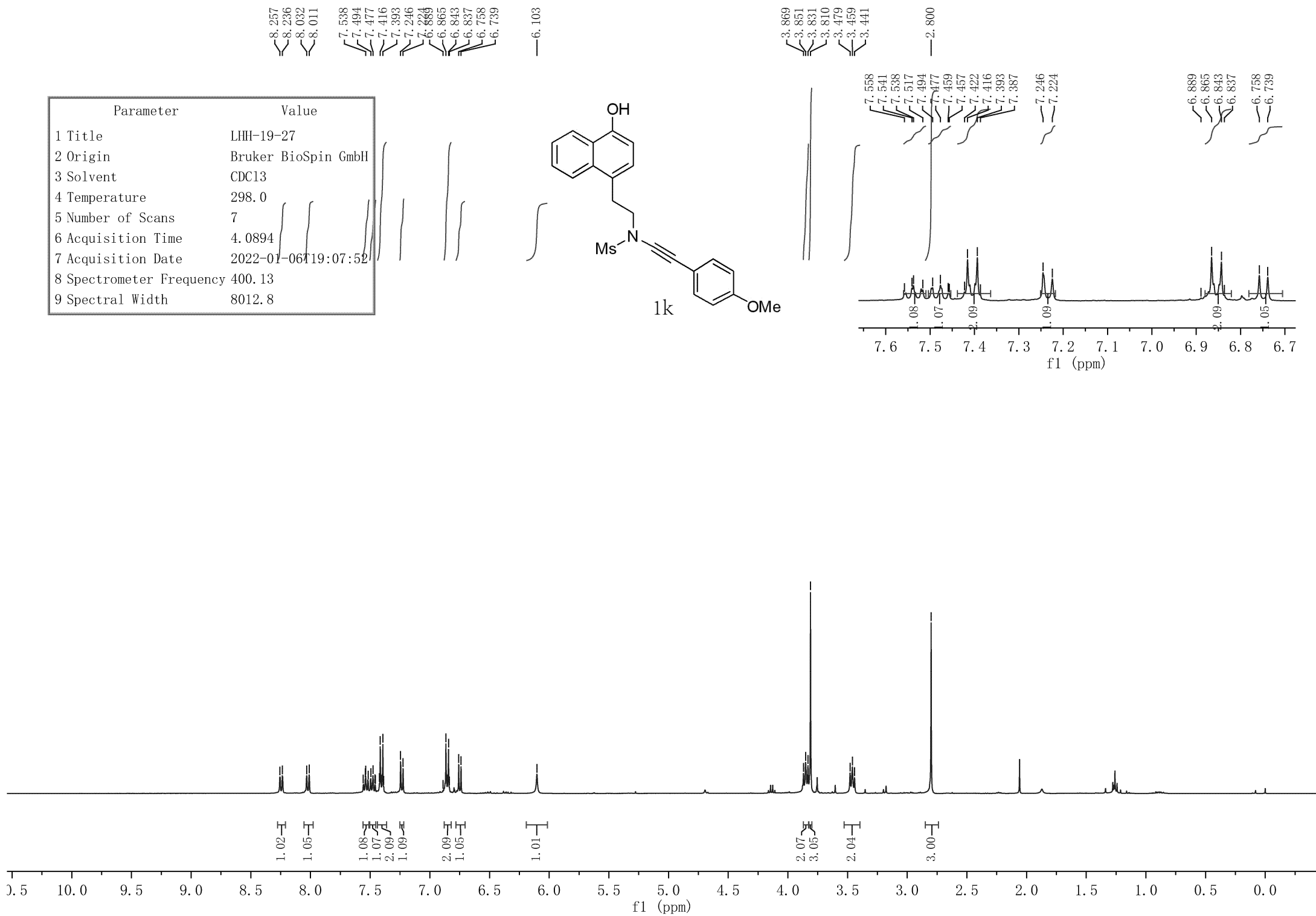
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7.121



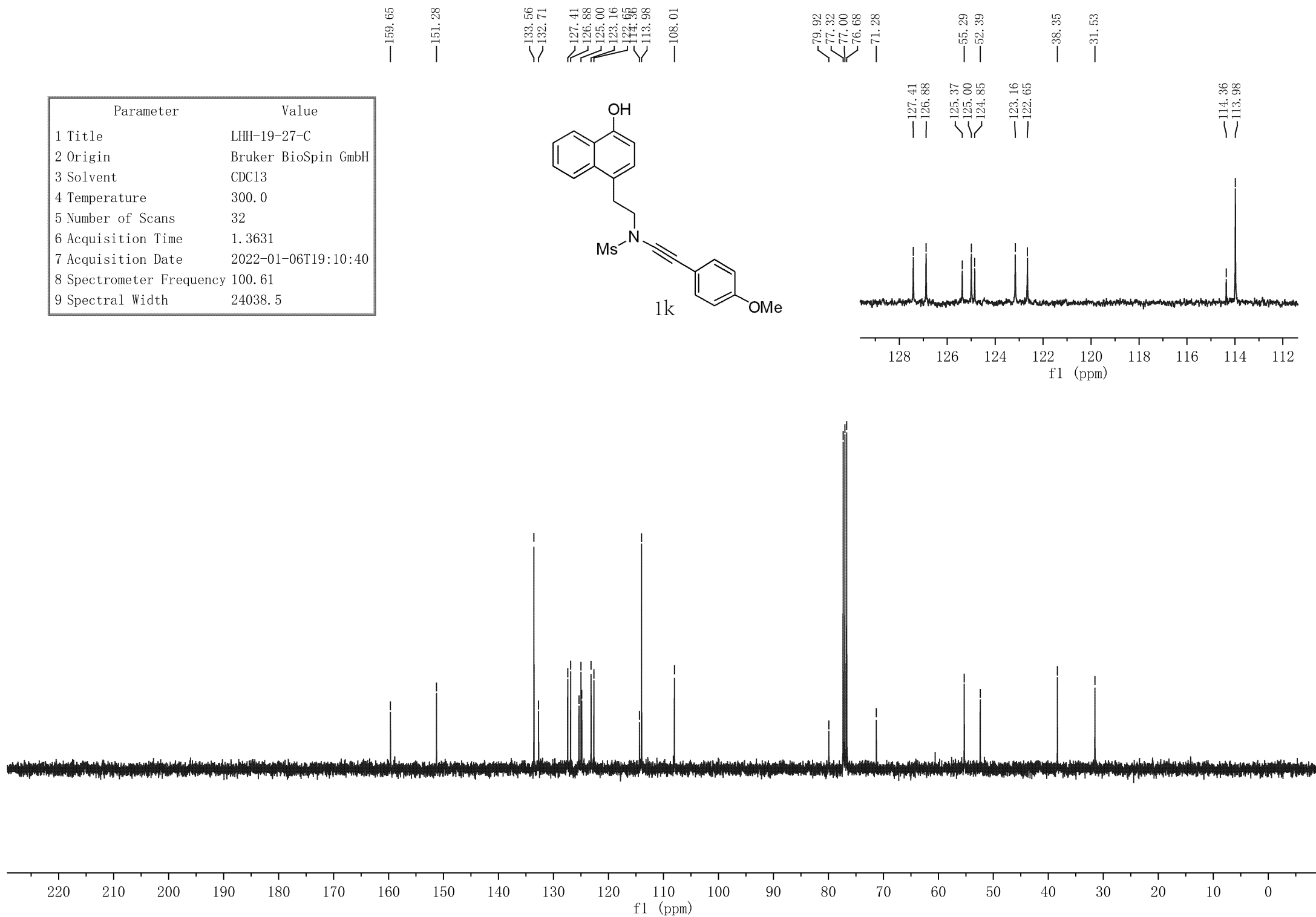
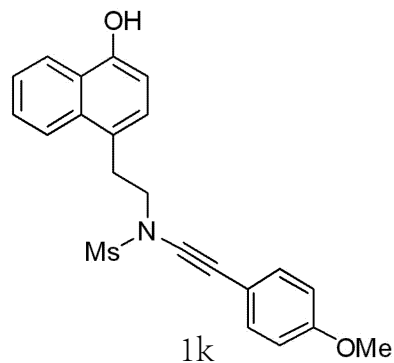
Parameter	Value
1 Title	LHH-19-70-c
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.8
5 Number of Scans	85
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-18T12:40:08
8 Spectrometer Frequency	100.56
9 Spectral Width	24038.5



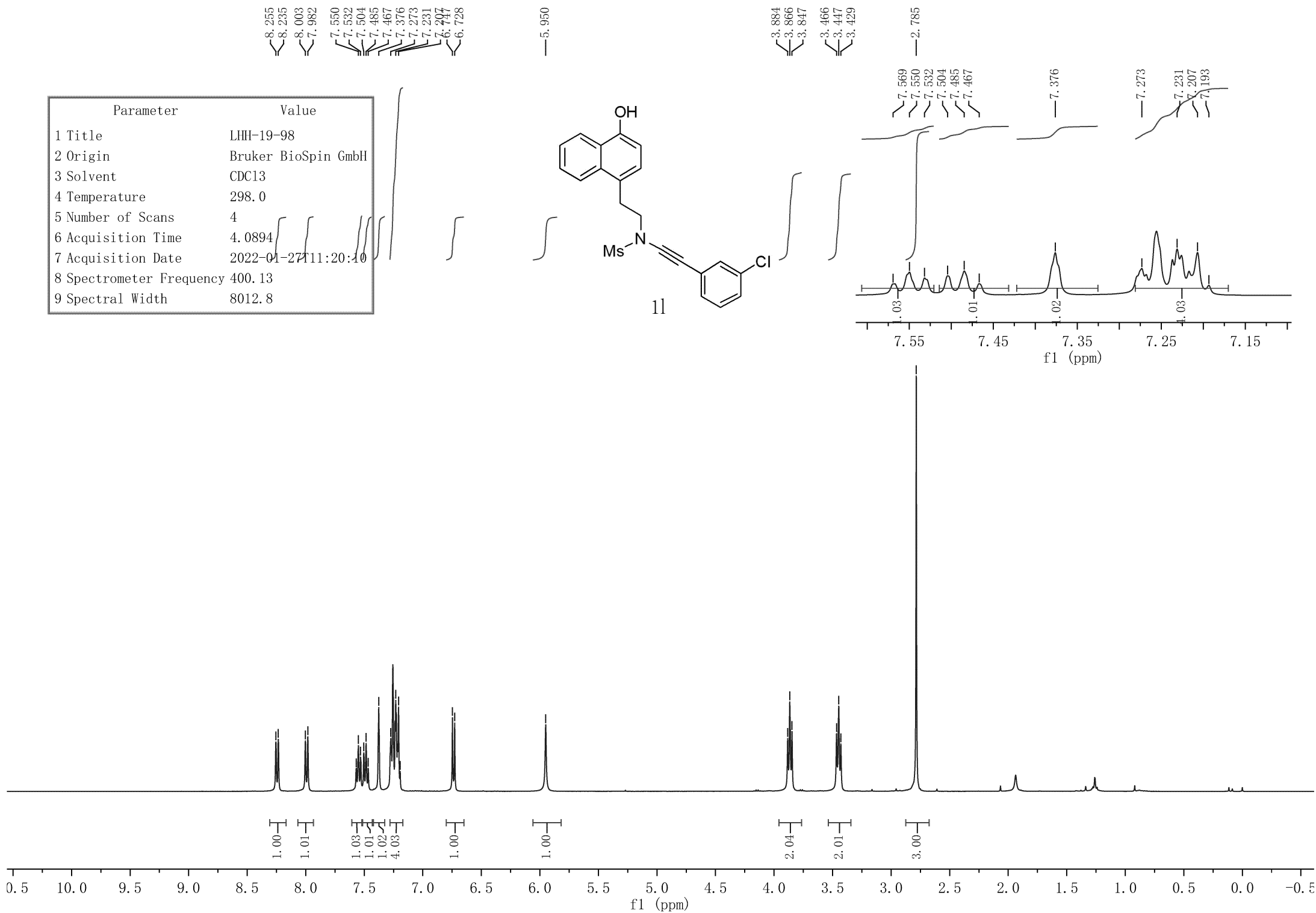
Parameter	Value
1 Title	LHH-19-27
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	7
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-06T19:07:52
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



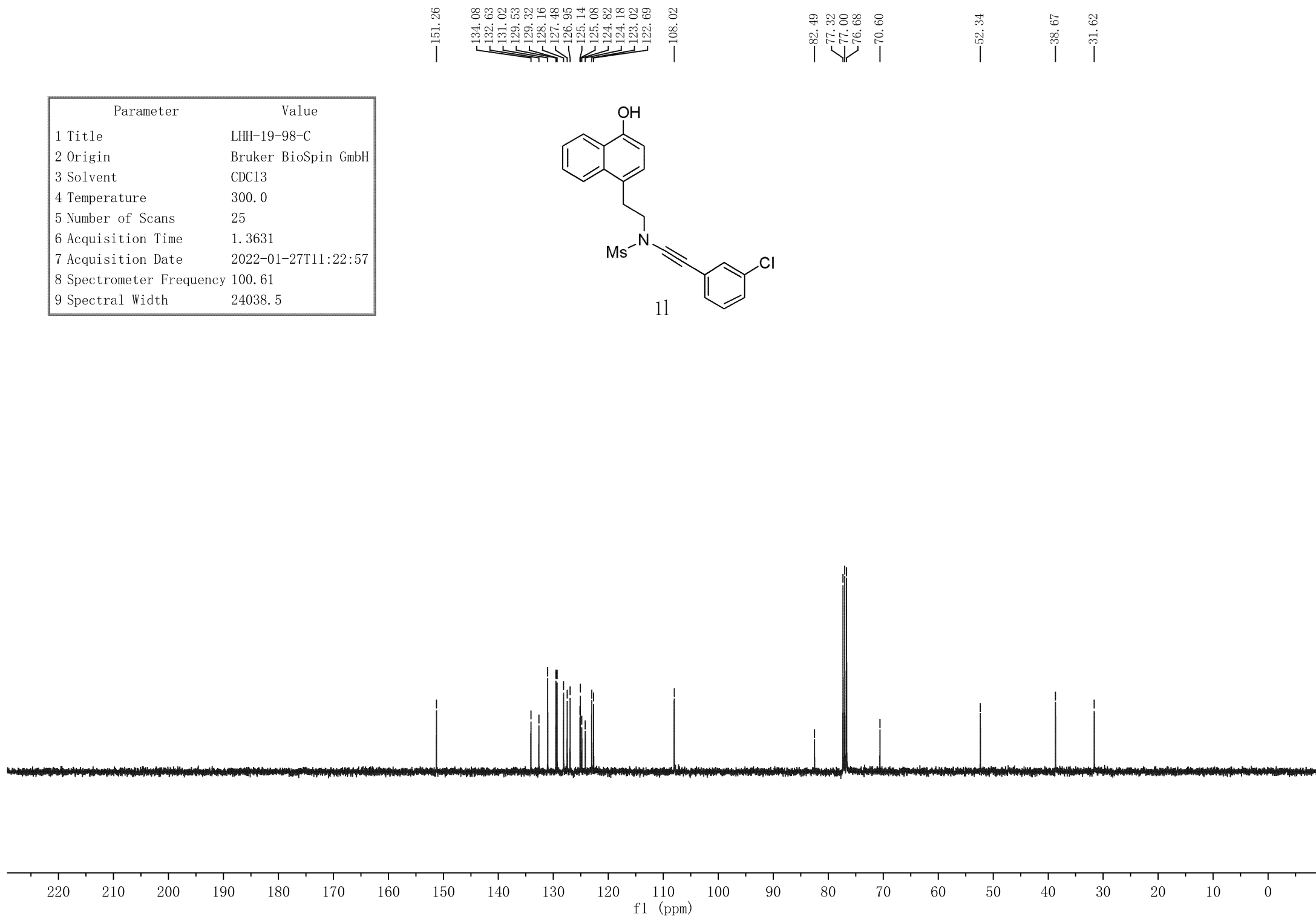
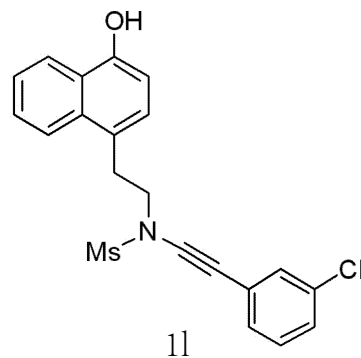
Parameter	Value
1 Title	LHH-19-27-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	32
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-06T19:10:40
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-98
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	4
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-27 11:20:10
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



Parameter	Value
1 Title	LHH-19-98-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	25
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-27T11:22:57
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	zyp-1-119
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-19T18:12:14
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

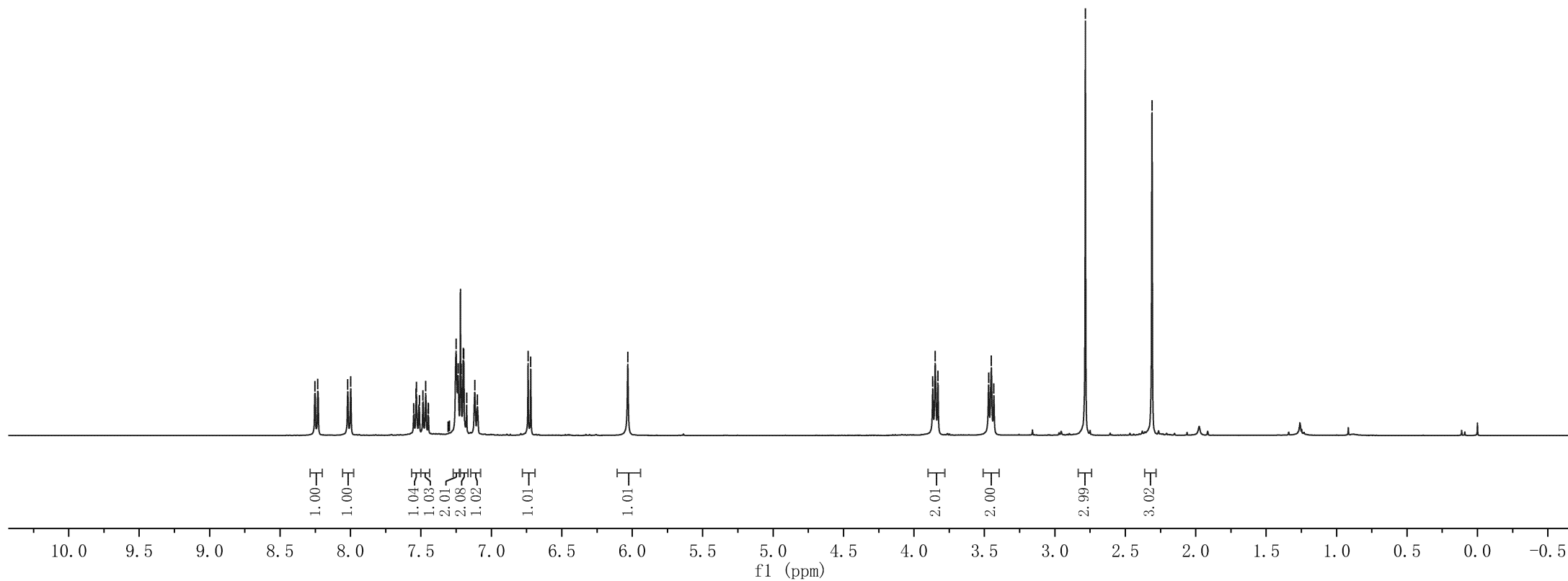
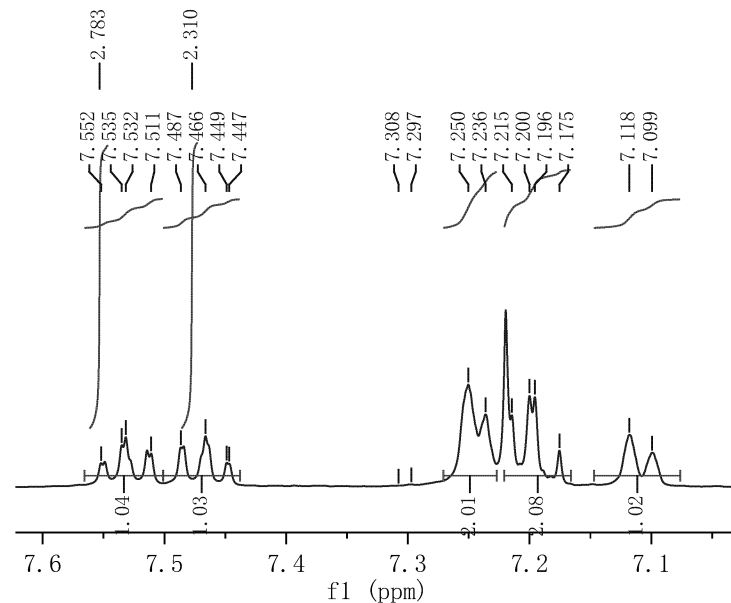
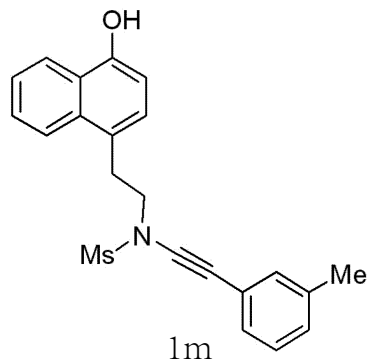
8.253
8.232
8.019
7.998
7.535
7.532
7.466
7.250
7.236
7.215
7.200
7.196
7.118
6.739
6.720

6.032

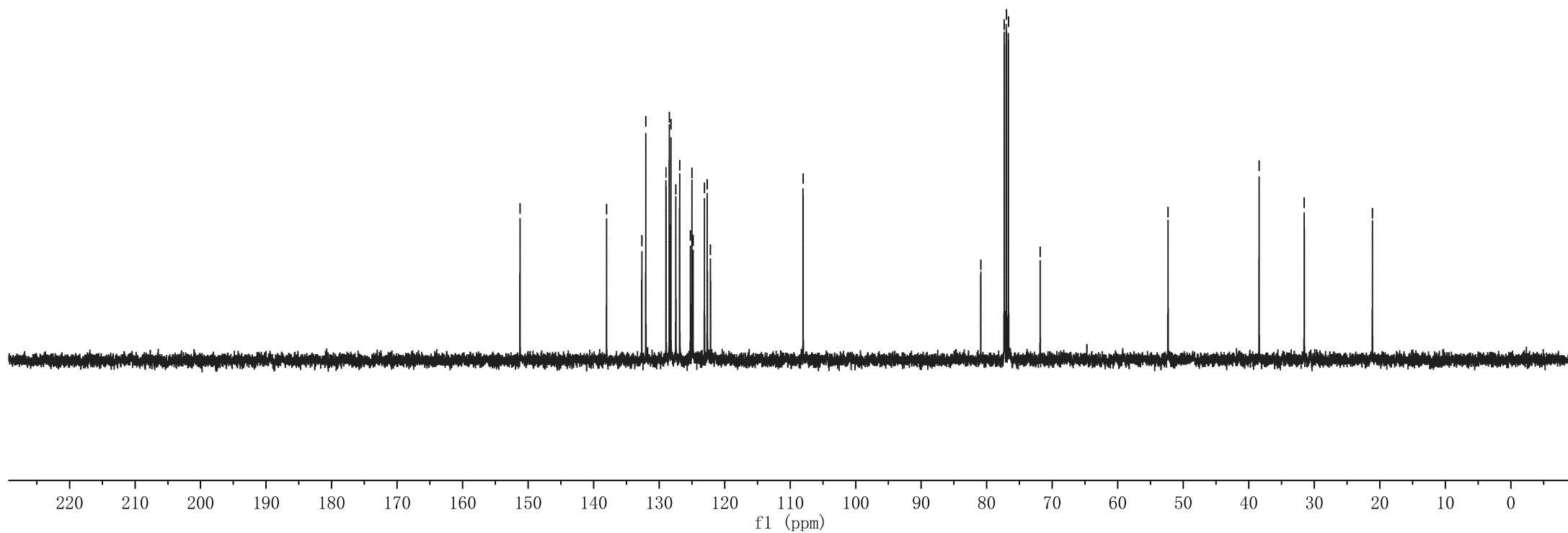
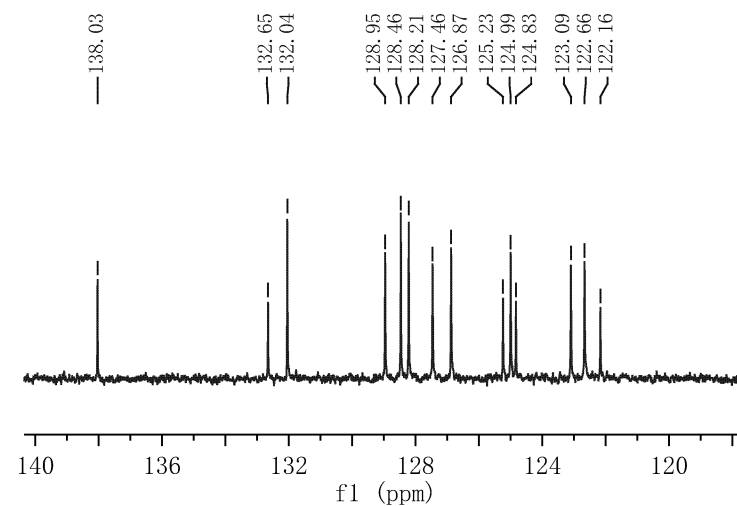
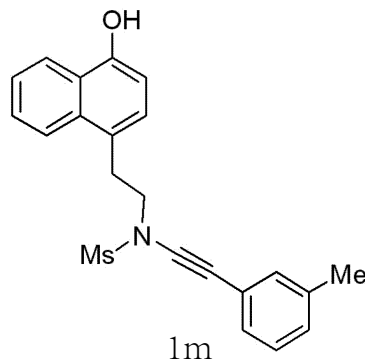
3.867
3.849
3.830
3.470
3.451
3.433

2.783
2.310
7.552
7.535
7.532
7.511
7.487
7.466
7.449
7.447

7.308
7.297
7.250
7.236
7.215
7.200
7.196
7.175
7.118
7.099



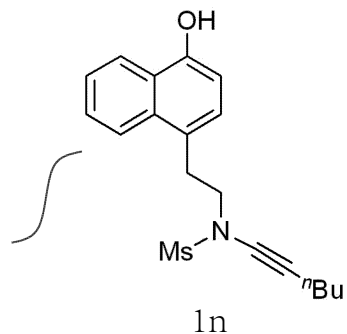
Parameter	Value
1 Title	zyp-1-119-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	31
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-19T18:14:44
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



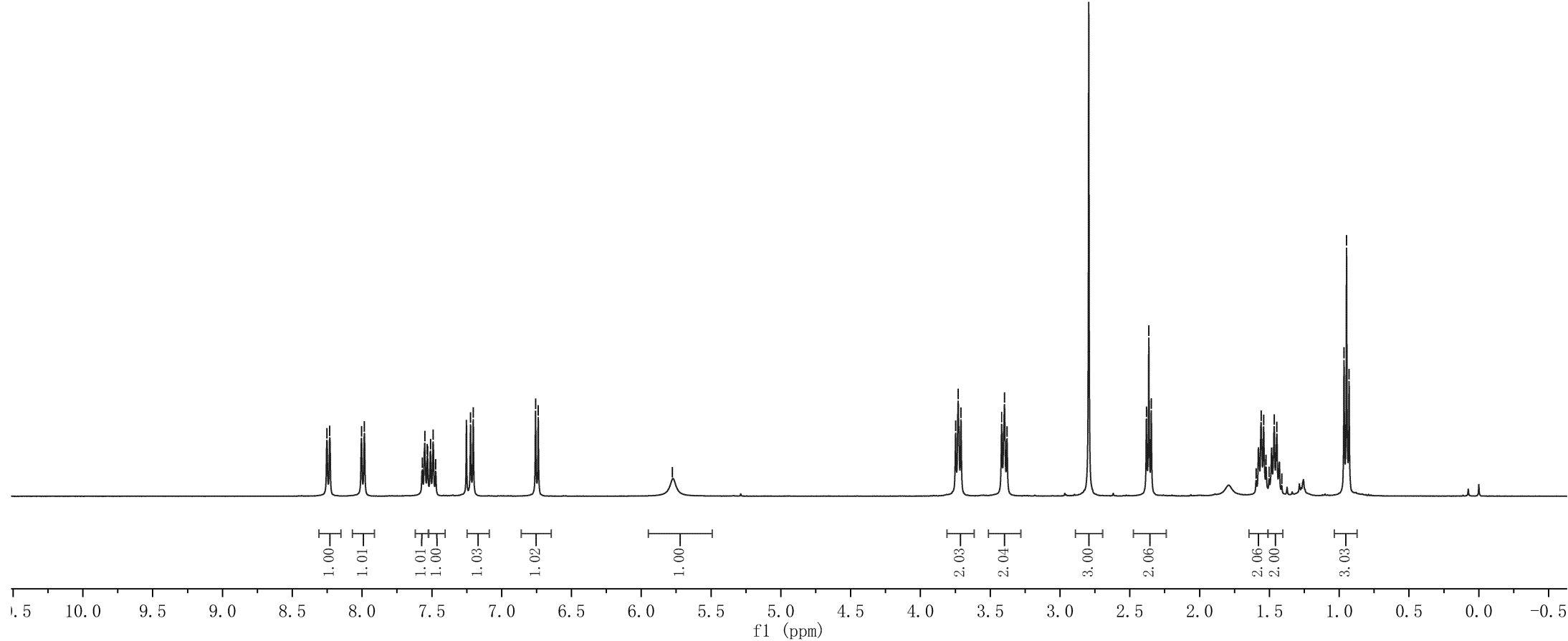
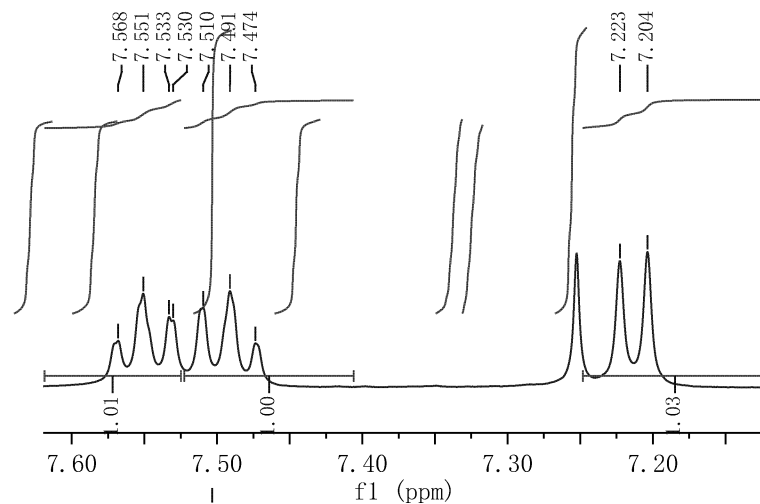
Parameter	Value
1 Title	LHH-19-90
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	4
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-24T17:01:30
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

8.252
8.232
8.004
7.984
7.551
7.533
7.530
7.510
7.491
7.483
7.204
6.757
6.738

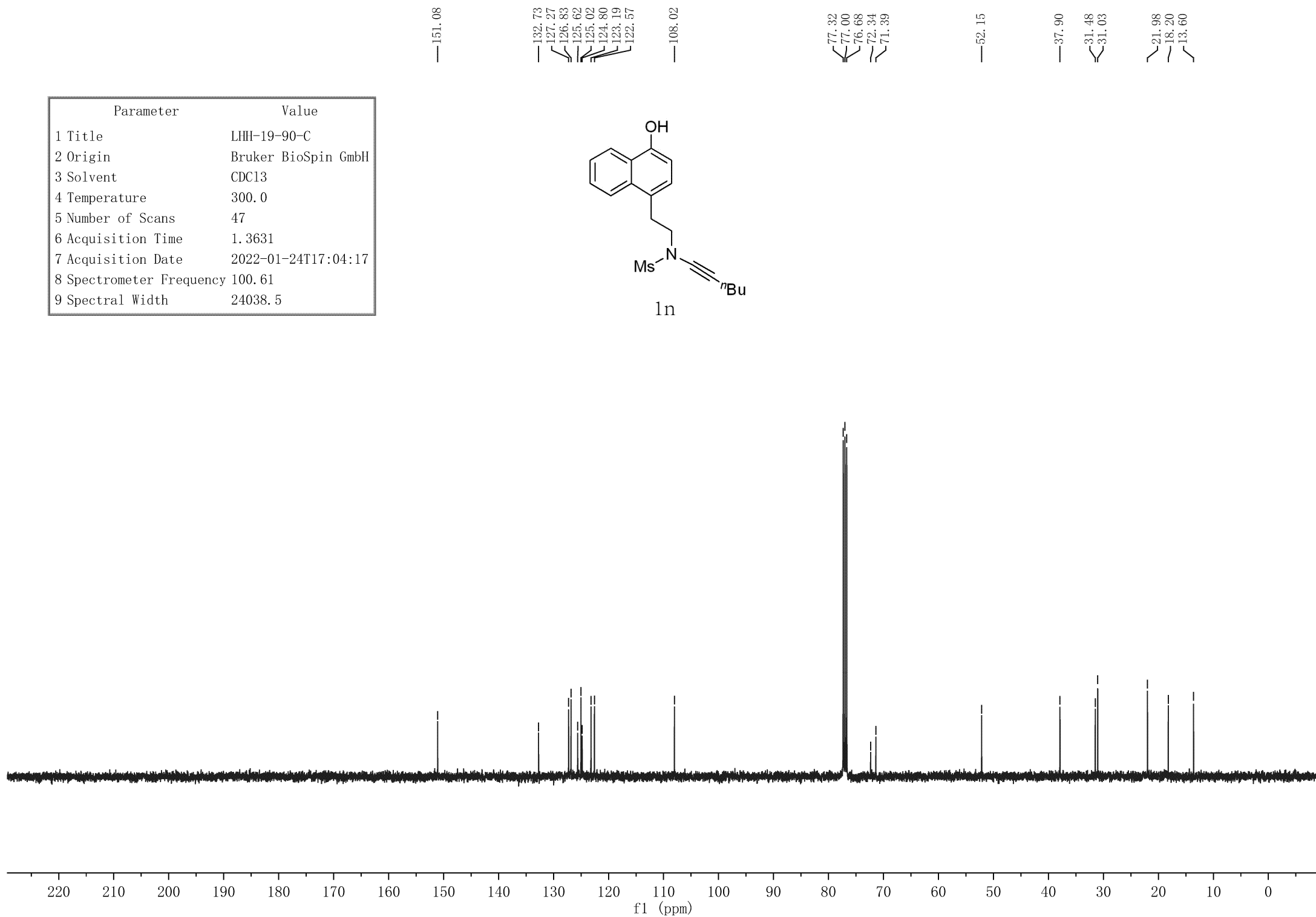
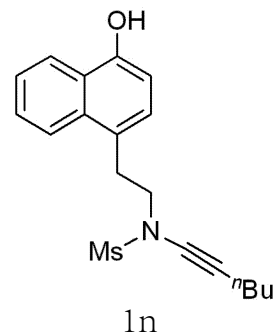
5.777



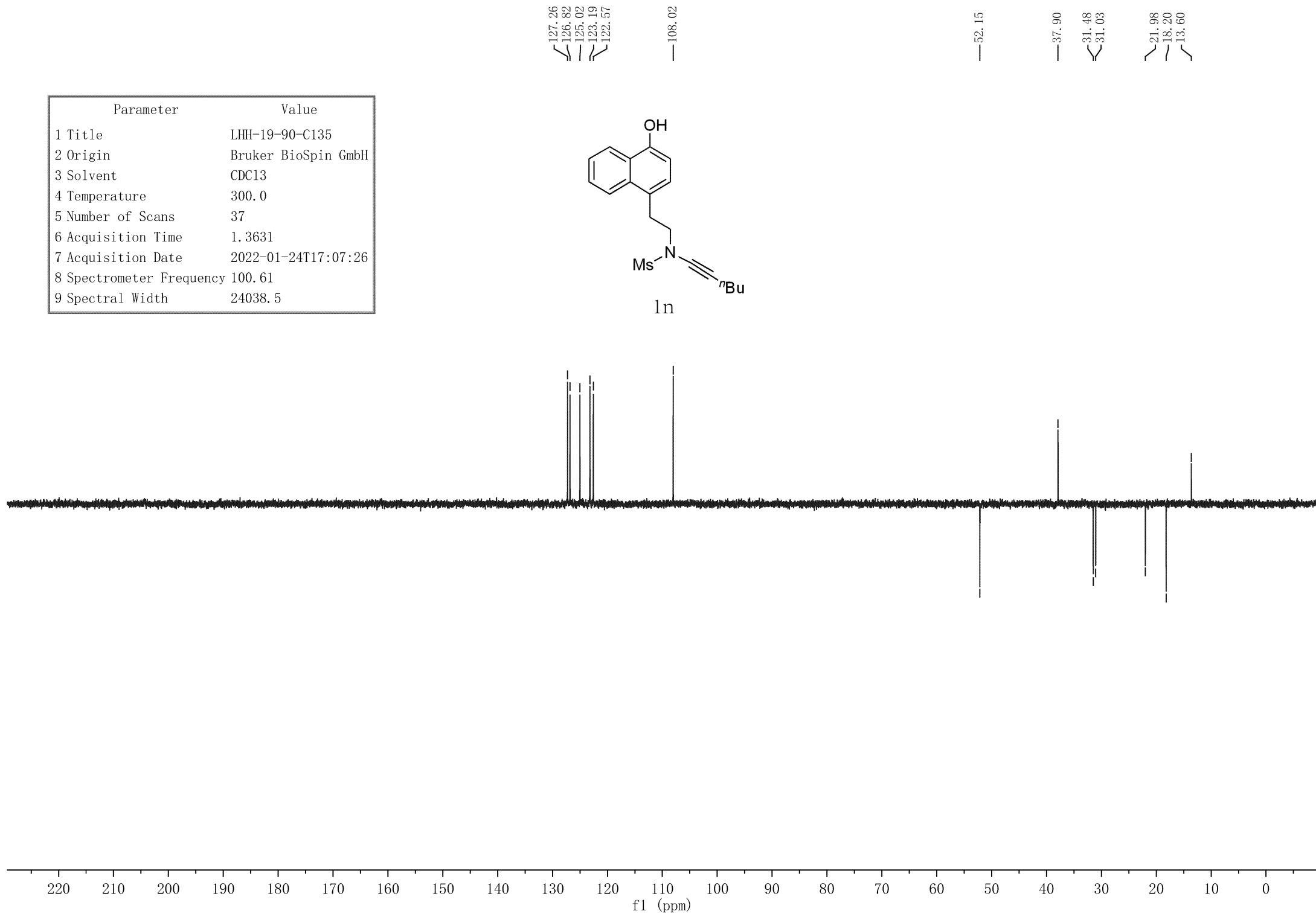
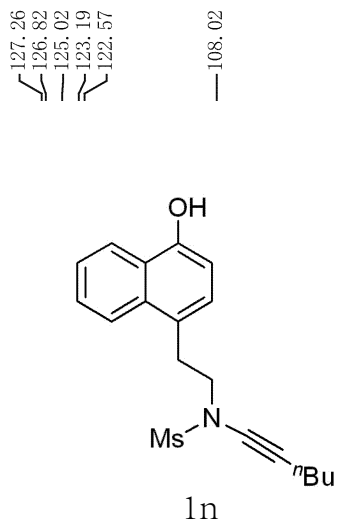
3.748
3.730
3.710
3.418
3.398
3.380
2.794
2.381
2.364
2.346
1.595
1.560
1.541
1.524
1.502
1.466
1.447
1.412
0.965
0.947
0.929



Parameter	Value
1 Title	LHH-19-90-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	47
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-24T17:04:17
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-90-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	37
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-24T17:07:26
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



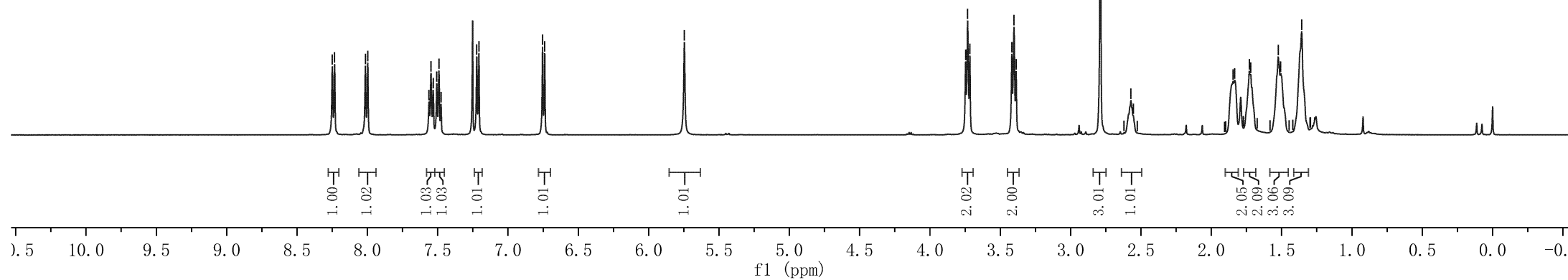
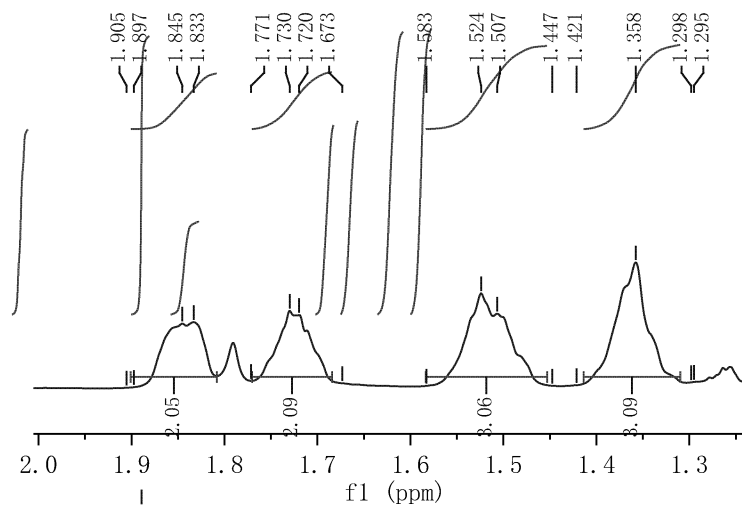
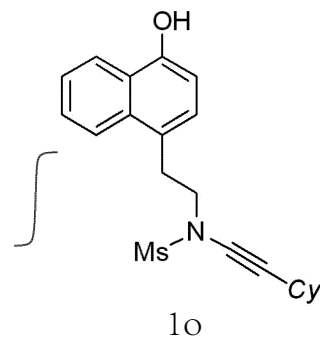
Parameter	Value
1 Title	zyp-1-109-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	153
6 Acquisition Time	1.1010
7 Acquisition Date	2022-04-15T12:33:18
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9

8.249
8.233
8.014
7.997
7.562
7.548
7.532
7.507
7.494
7.222
7.207
6.754
6.739

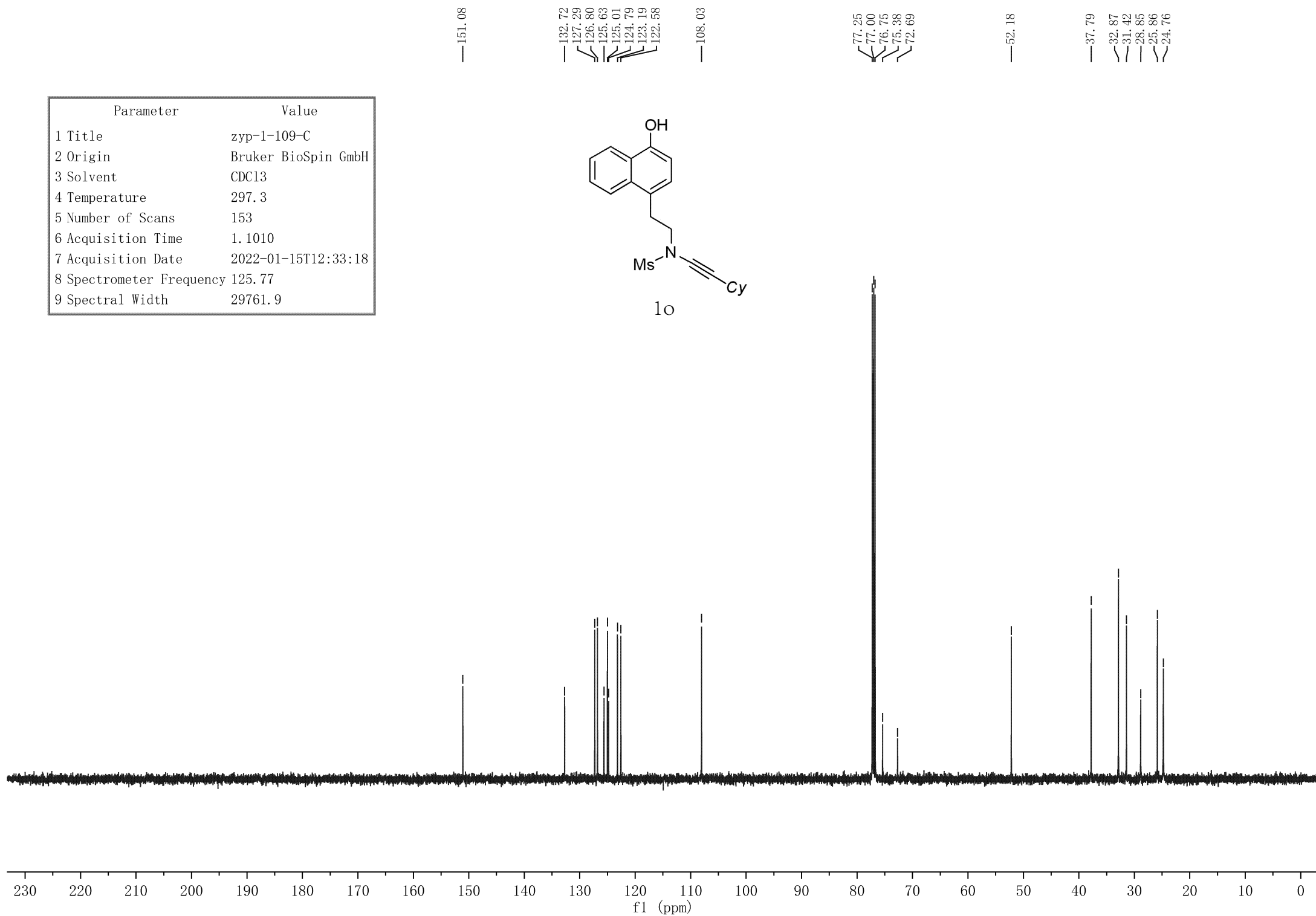
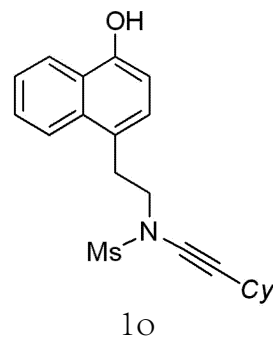
5.746

3.747
3.732
3.716
3.418
3.402
3.388

2.788
2.621
2.571
2.555
2.526
1.905
1.897
1.845
1.833
1.771
1.730
1.720
1.673
1.583
1.524
1.507
1.447
1.421
1.358
1.298
1.295

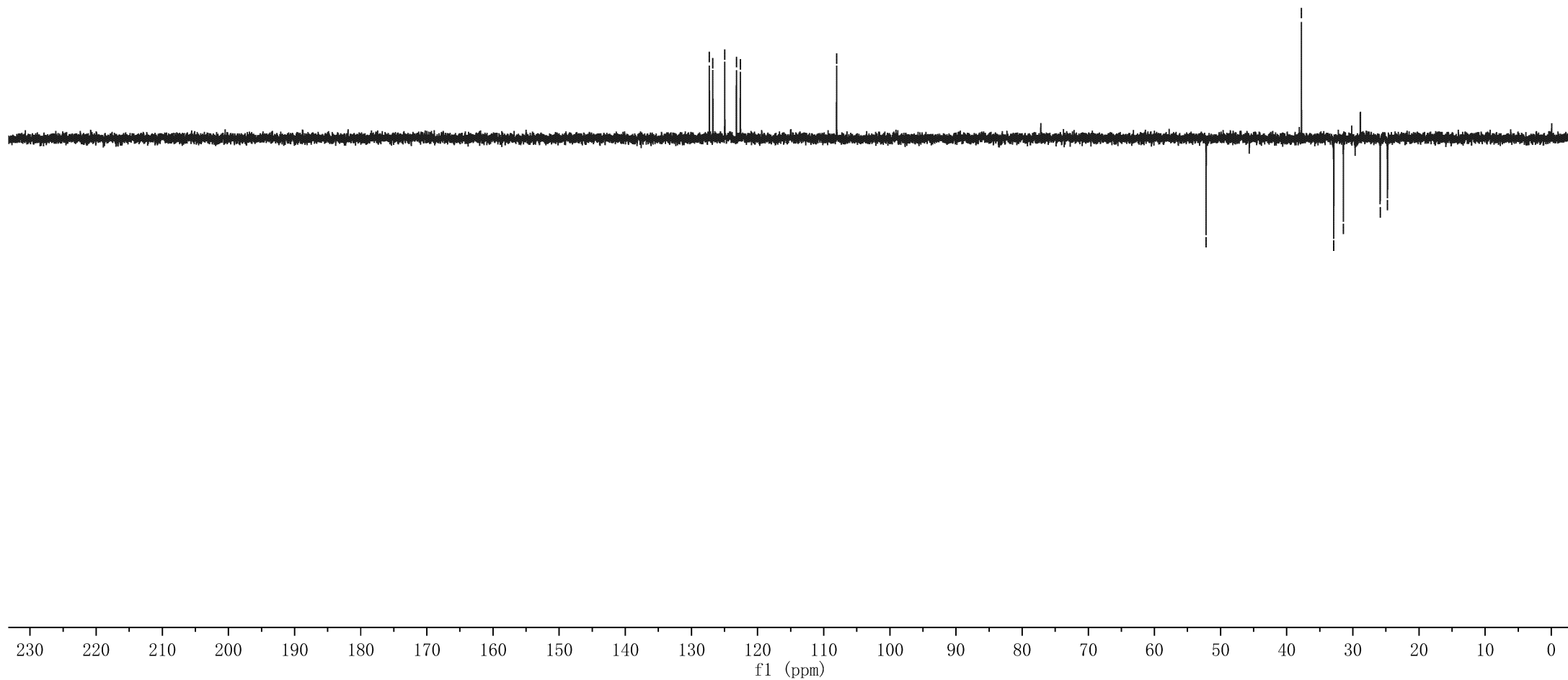
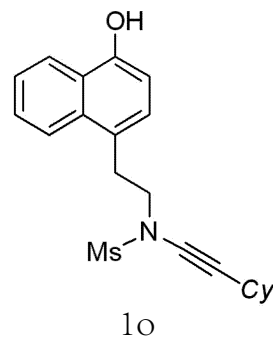


Parameter	Value
1 Title	zyp-1-109-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	153
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-15T12:33:18
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9

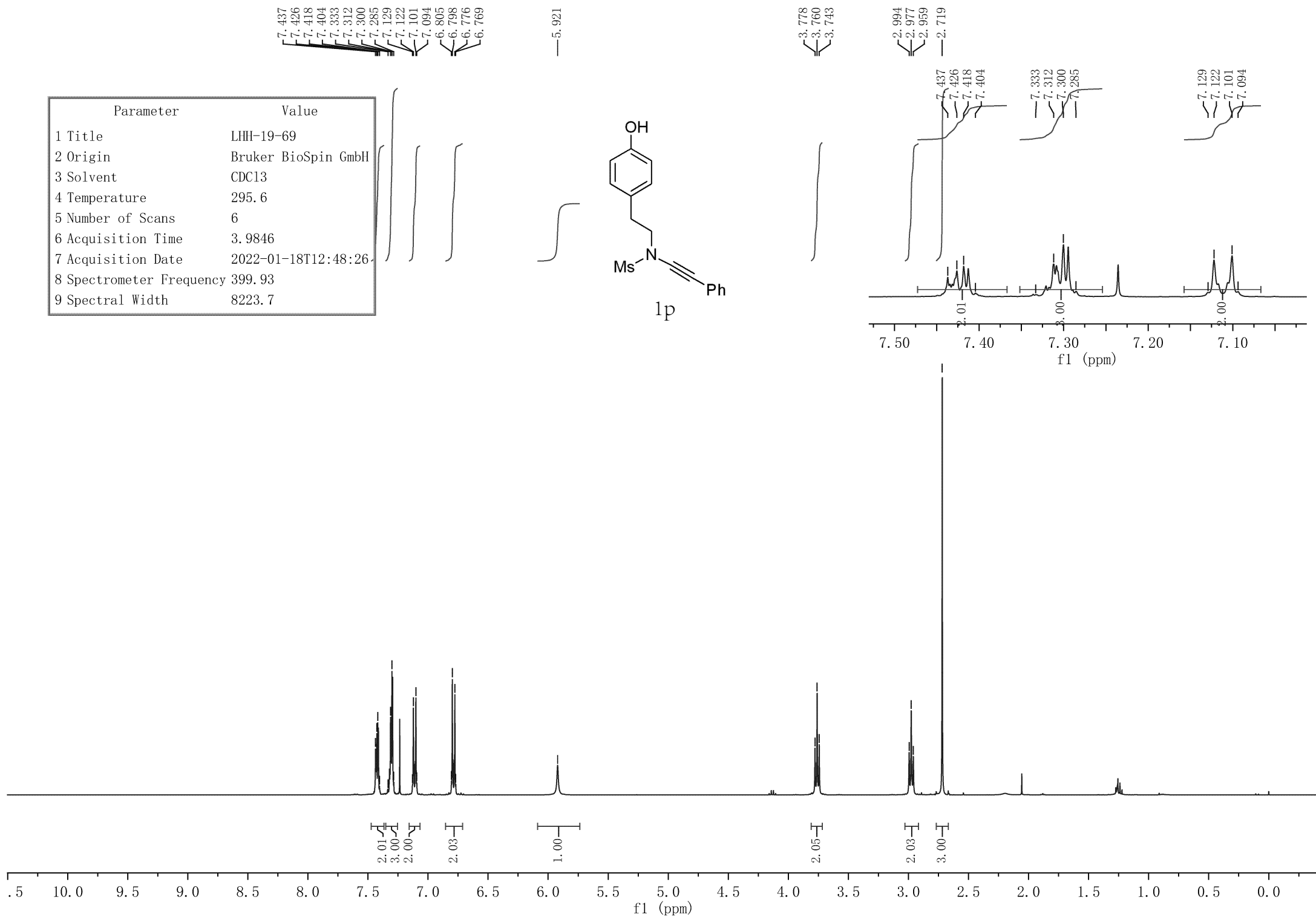


Parameter	Value
1 Title	LHH-13-60-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	301.5
5 Number of Scans	30
6 Acquisition Time	1.1010
7 Acquisition Date	2020-06-05T16:42:26
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9

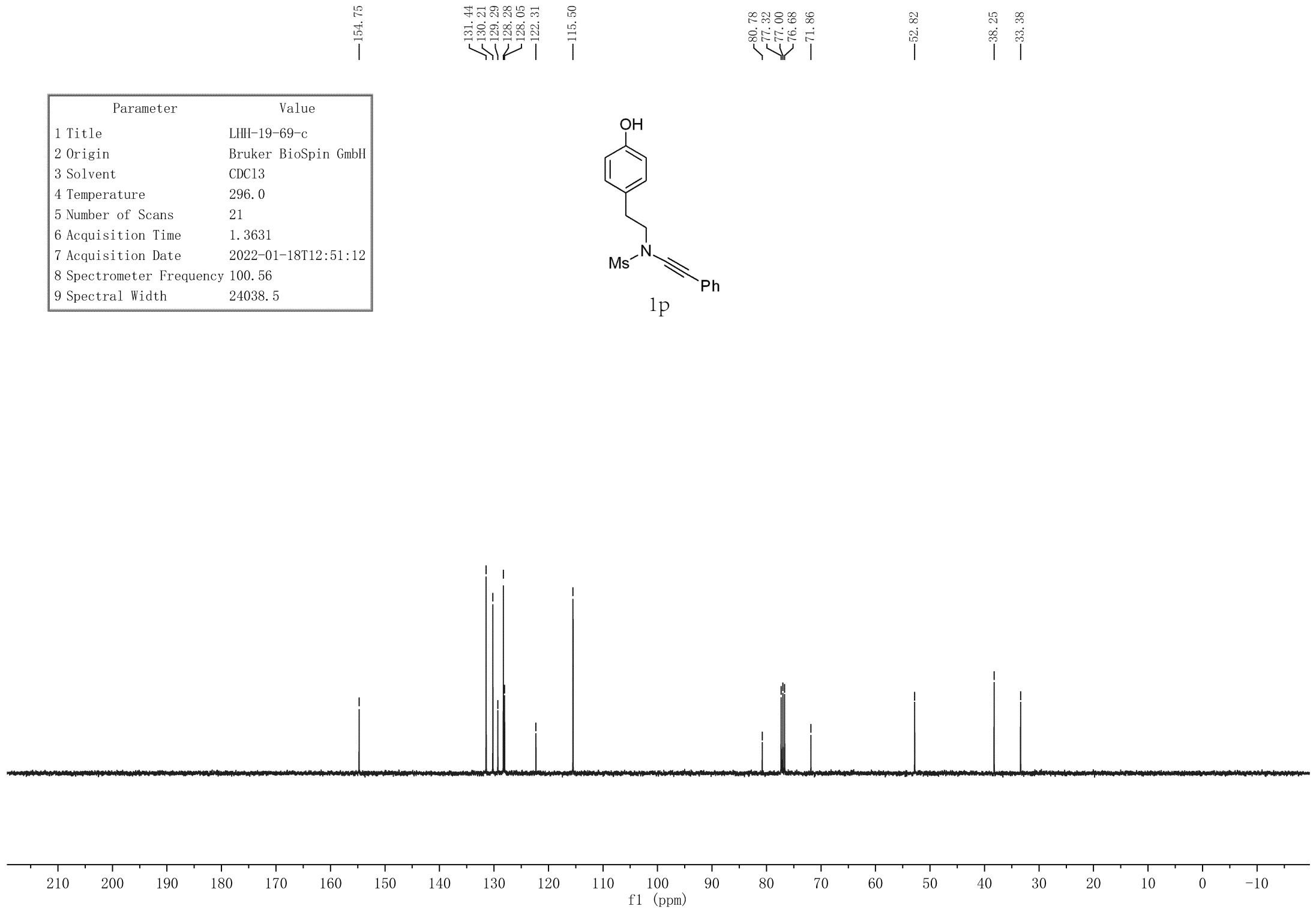
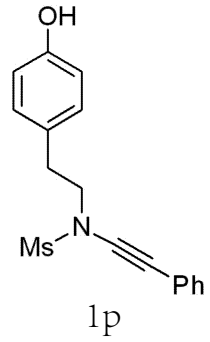
¹³C NMR chemical shifts (ppm):
 127.29, 126.77, 124.96, 123.20, 122.60, 108.05, 52.19, 37.78, 32.89, 31.44, 25.87, 24.76



Parameter	Value
1 Title	LHH-19-69
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.6
5 Number of Scans	6
6 Acquisition Time	3.9846
7 Acquisition Date	2022-01-18T12:48:26
8 Spectrometer Frequency	399.93
9 Spectral Width	8223.7



Parameter	Value
1 Title	LHH-19-69-c
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.0
5 Number of Scans	21
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-18T12:51:12
8 Spectrometer Frequency	100.56
9 Spectral Width	24038.5



Parameter	Value
1 Title	LIII-19-54
2 Origin	
3 Solvent	CDC13
4 Temperature	297.2
5 Number of Scans	16
6 Acquisition Time	4.0002
7 Acquisition Date	2022-01-14T03:46:20
8 Spectrometer Frequency	399.93
9 Spectral Width	8012.0

7.757
7.736
7.309
7.291
7.284
7.279
7.276
7.005
6.739
6.718

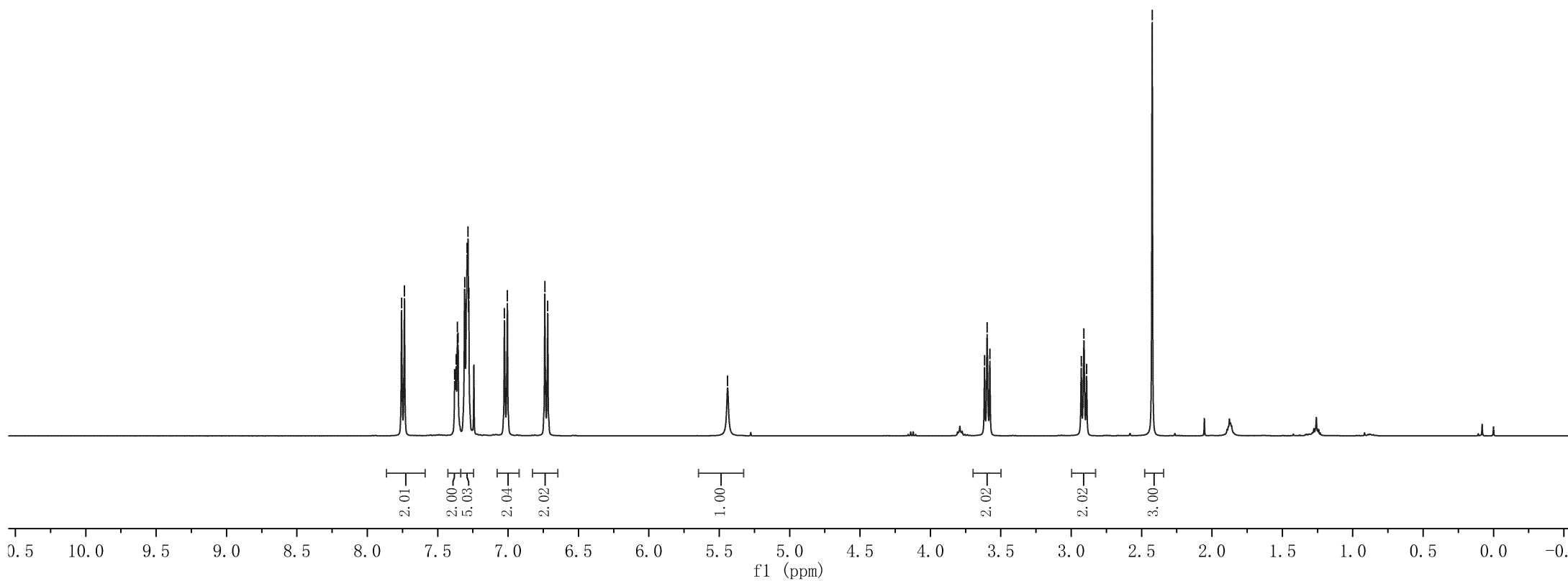
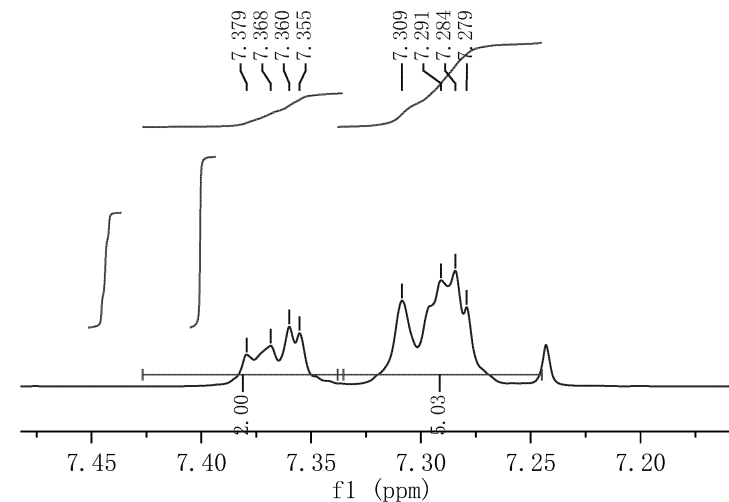
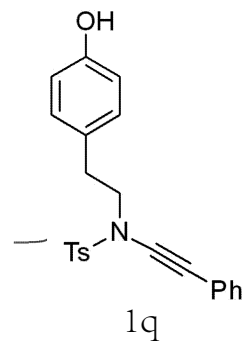
5.441

3.616
3.597
3.578

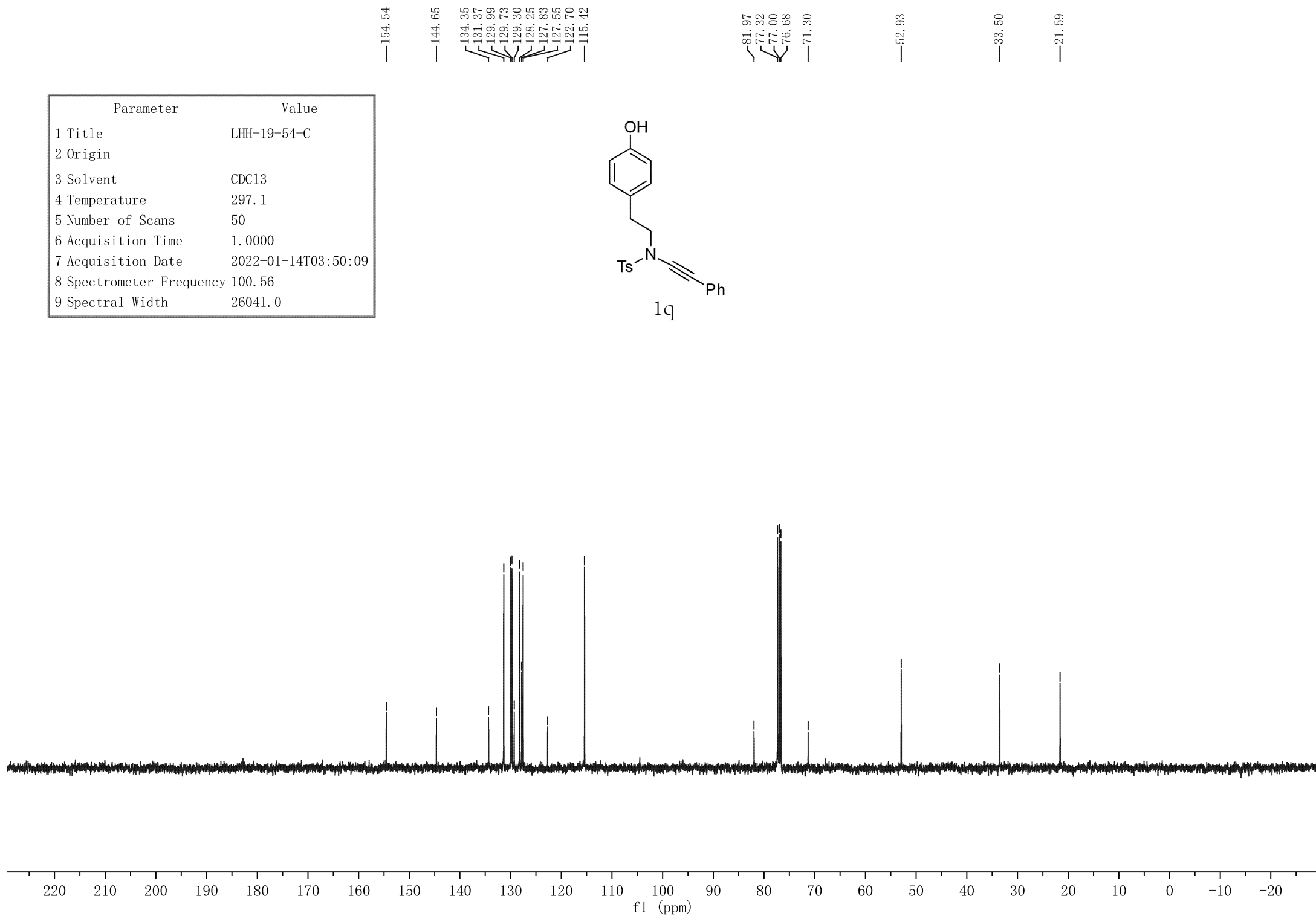
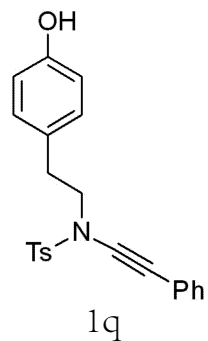
2.929
2.909
2.891

2.425

7.379
7.368
7.360
7.355
7.309
7.291
7.284
7.279



Parameter	Value
1 Title	LIHH-19-54-C
2 Origin	
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	50
6 Acquisition Time	1.0000
7 Acquisition Date	2022-01-14T03:50:09
8 Spectrometer Frequency	100.56
9 Spectral Width	26041.0



Parameter	Value
1 Title	LHH-10-178
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	9
6 Acquisition Time	4.0894
7 Acquisition Date	2019-09-19T09:23:03
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

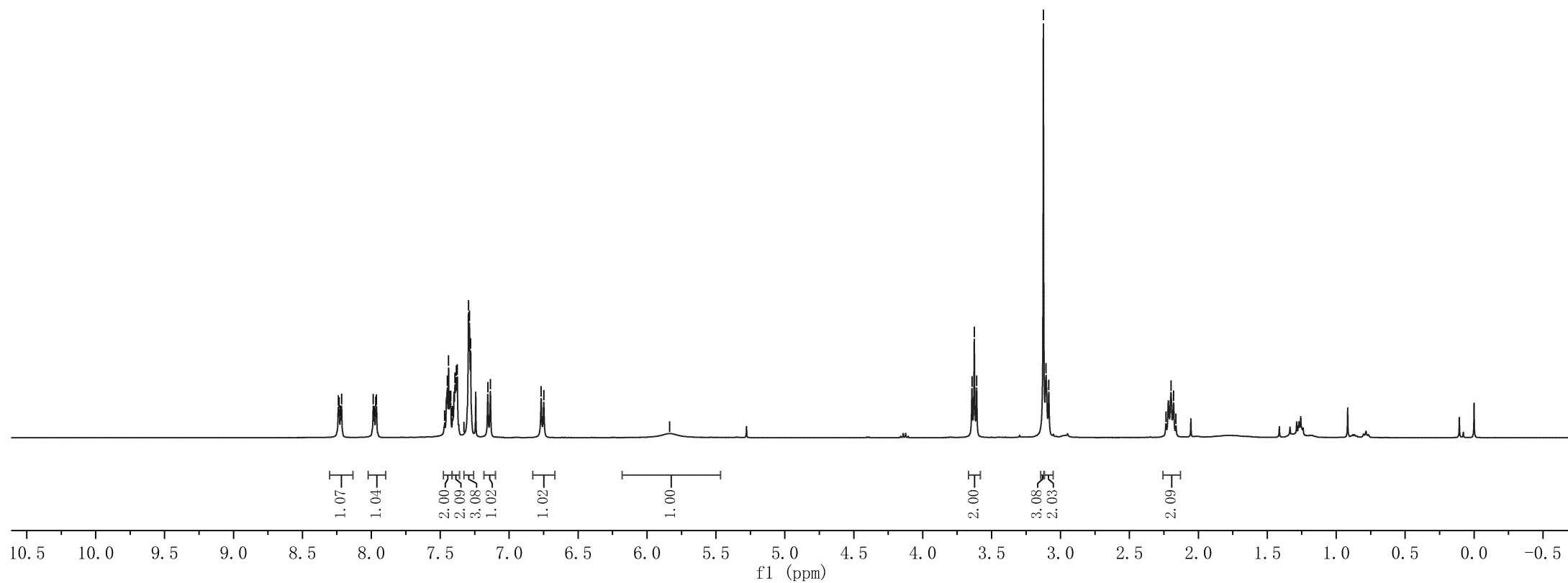
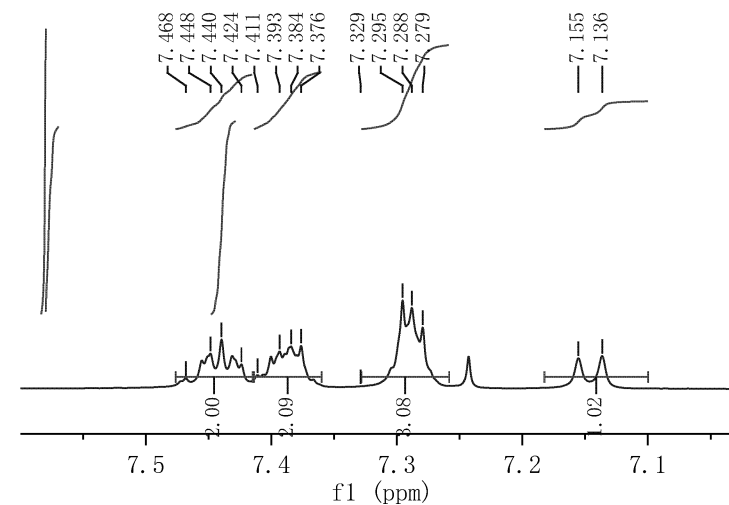
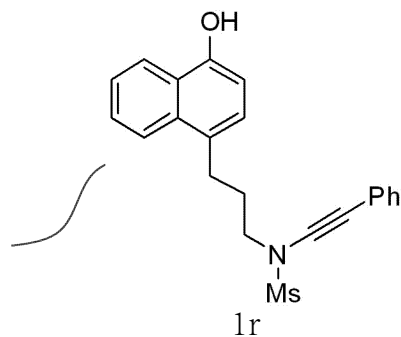
8.240
8.234
8.225
8.216
7.986
7.968
7.963
7.440
7.393
7.384
7.376
7.295
7.288
7.279
6.768
6.749

— 5.837

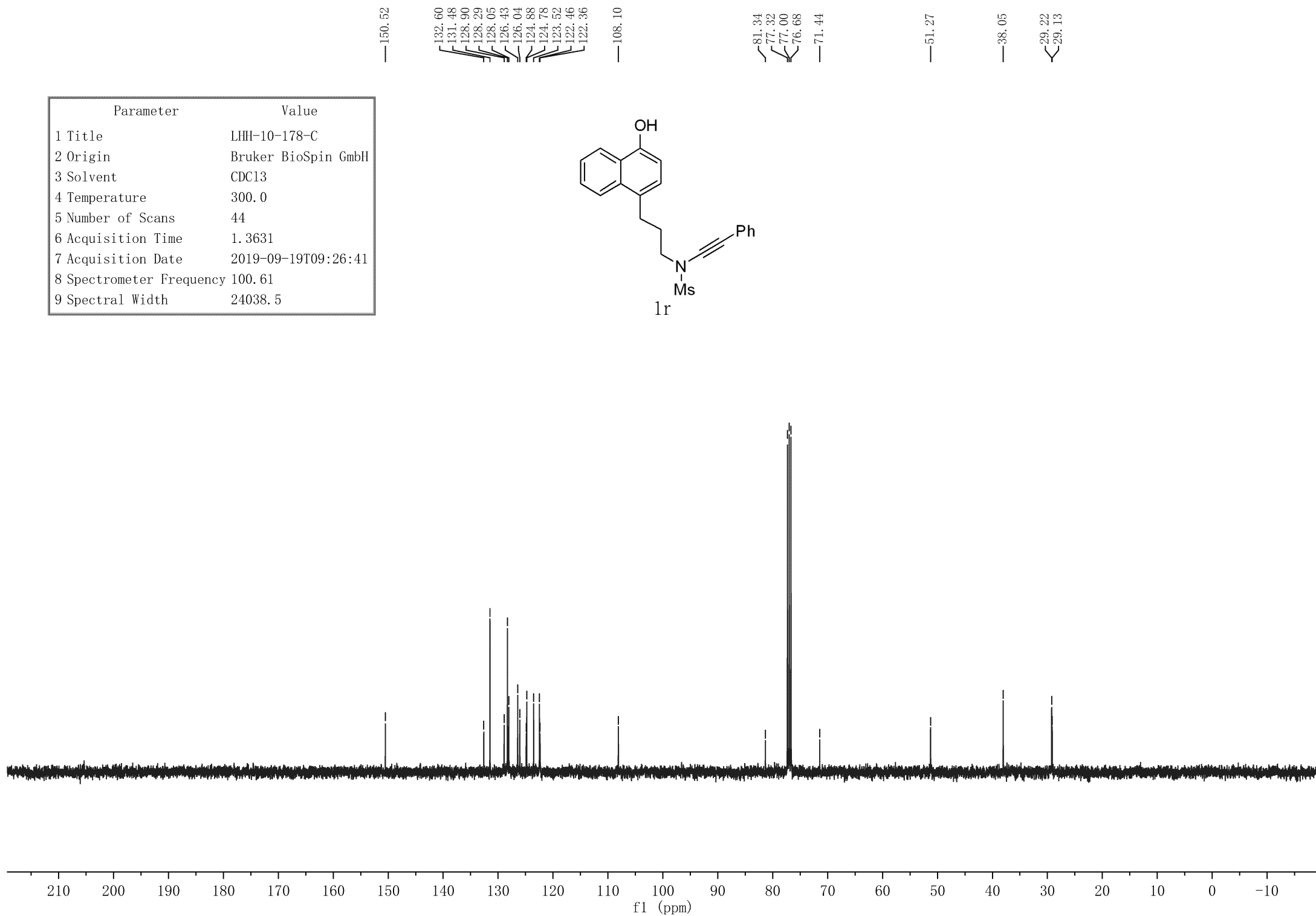
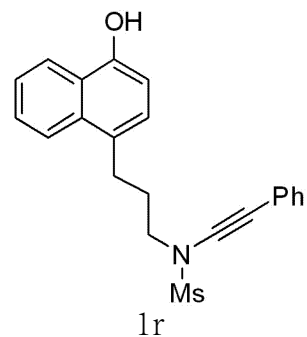
3.643
3.626
3.609

3.124
3.105
3.085

2.235
2.199
2.180
2.162



Parameter	Value
1 Title	LHH-10-178-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	44
6 Acquisition Time	1.3631
7 Acquisition Date	2019-09-19T09:26:41
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-12-104
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.1
5 Number of Scans	7
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-02T11:12:24
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7

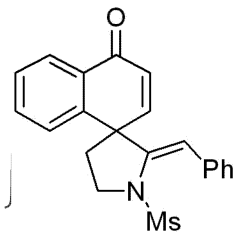
8.221
8.201
7.595
7.578
7.574
7.549
7.547
7.529
7.458
7.438
7.307
7.299
7.294
7.118
7.092
6.464
6.438

5.686

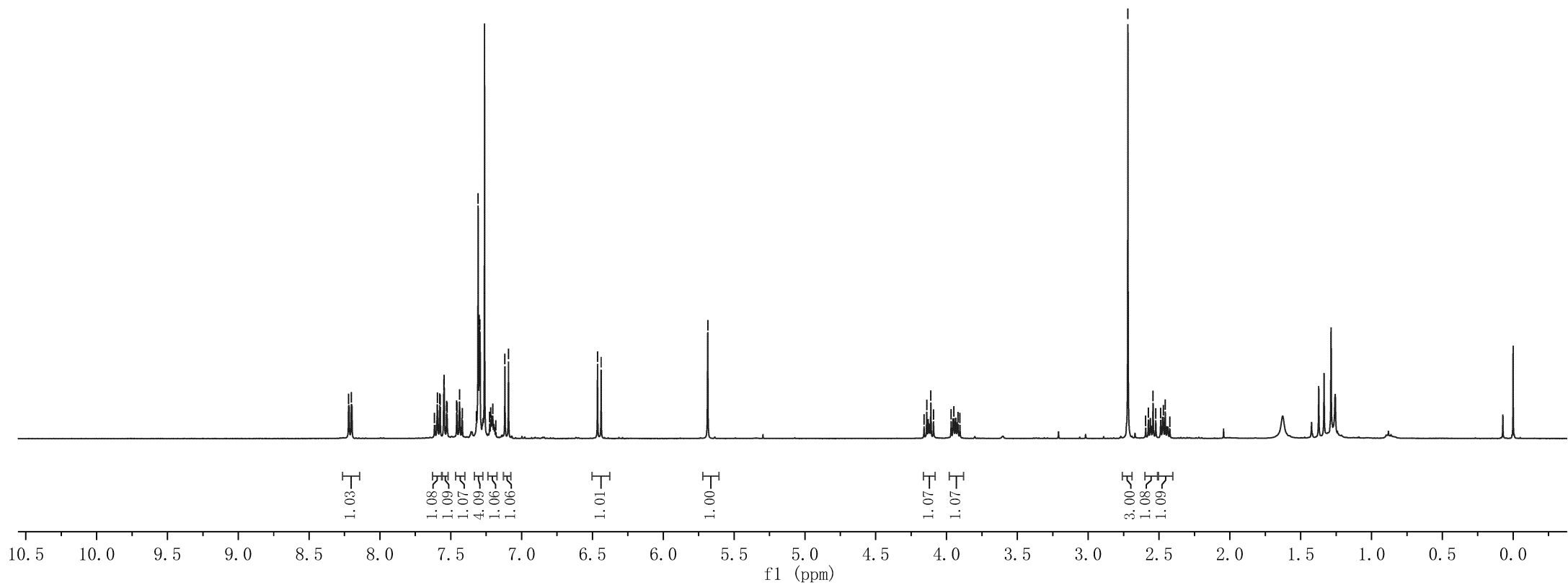
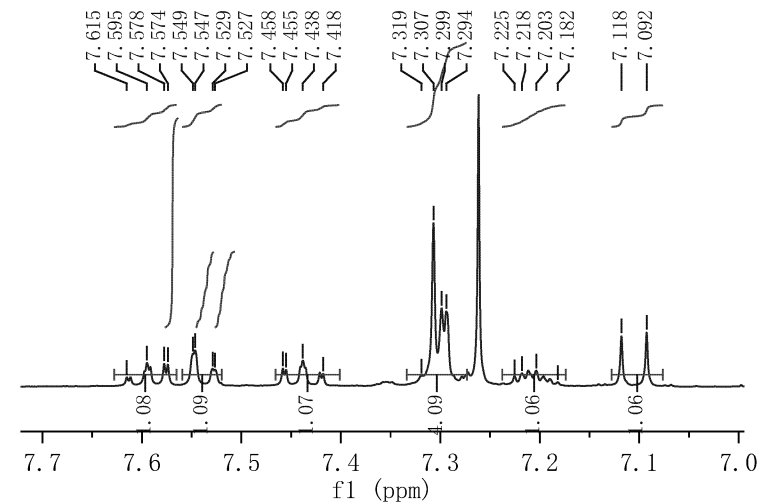
4.159
4.140
4.111
4.092
3.968
3.948
3.919
3.905

2.720
2.594
2.575
2.542
2.523
2.488
2.470
2.456
2.424

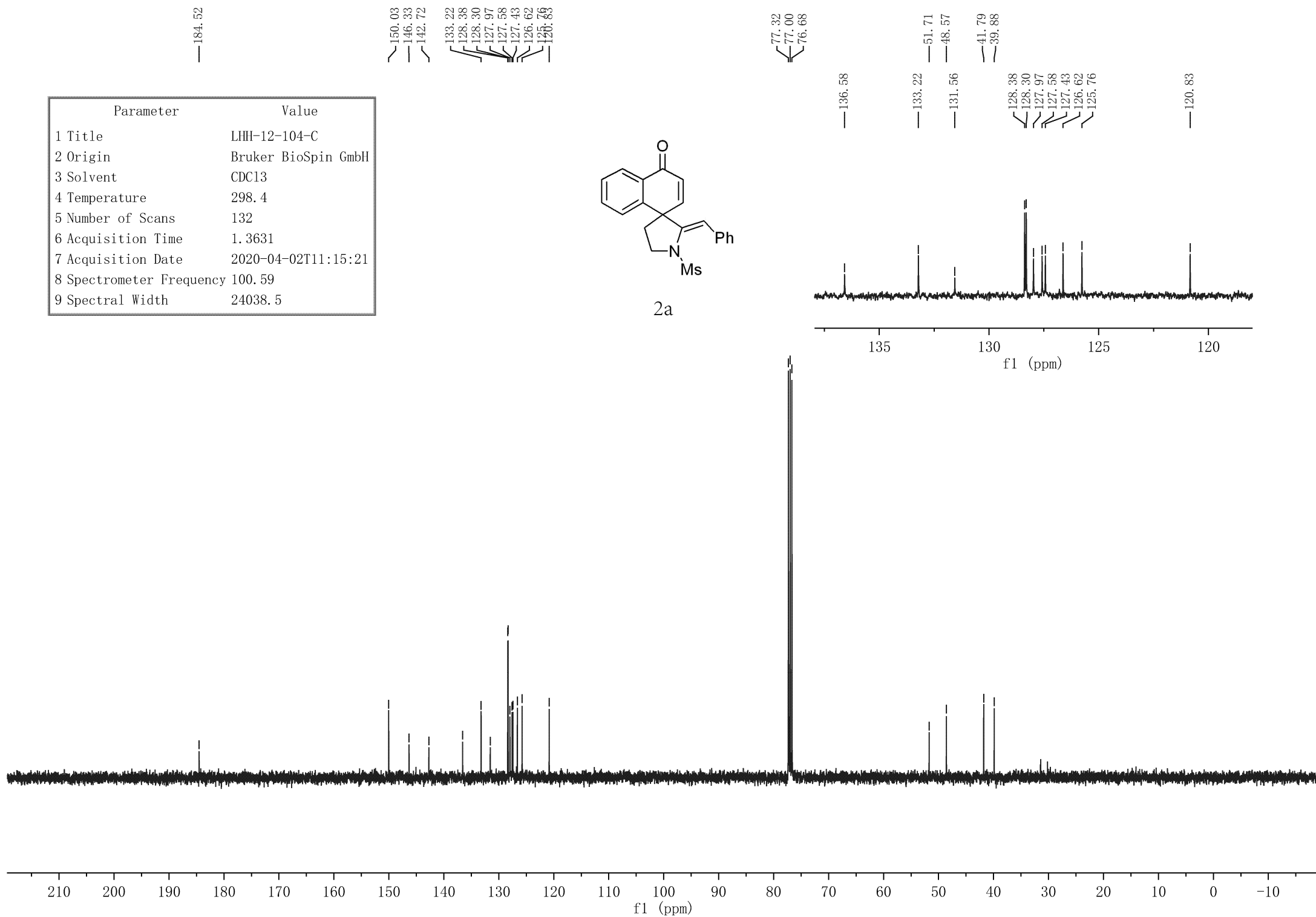
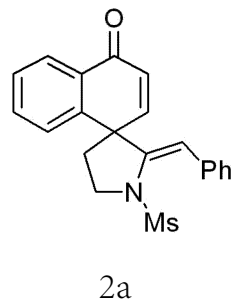
7.615
7.595
7.578
7.574
7.549
7.547
7.529
7.527
7.458
7.455
7.438
7.418
7.319
7.307
7.299
7.294
7.225
7.218
7.203
7.182
7.118
7.092



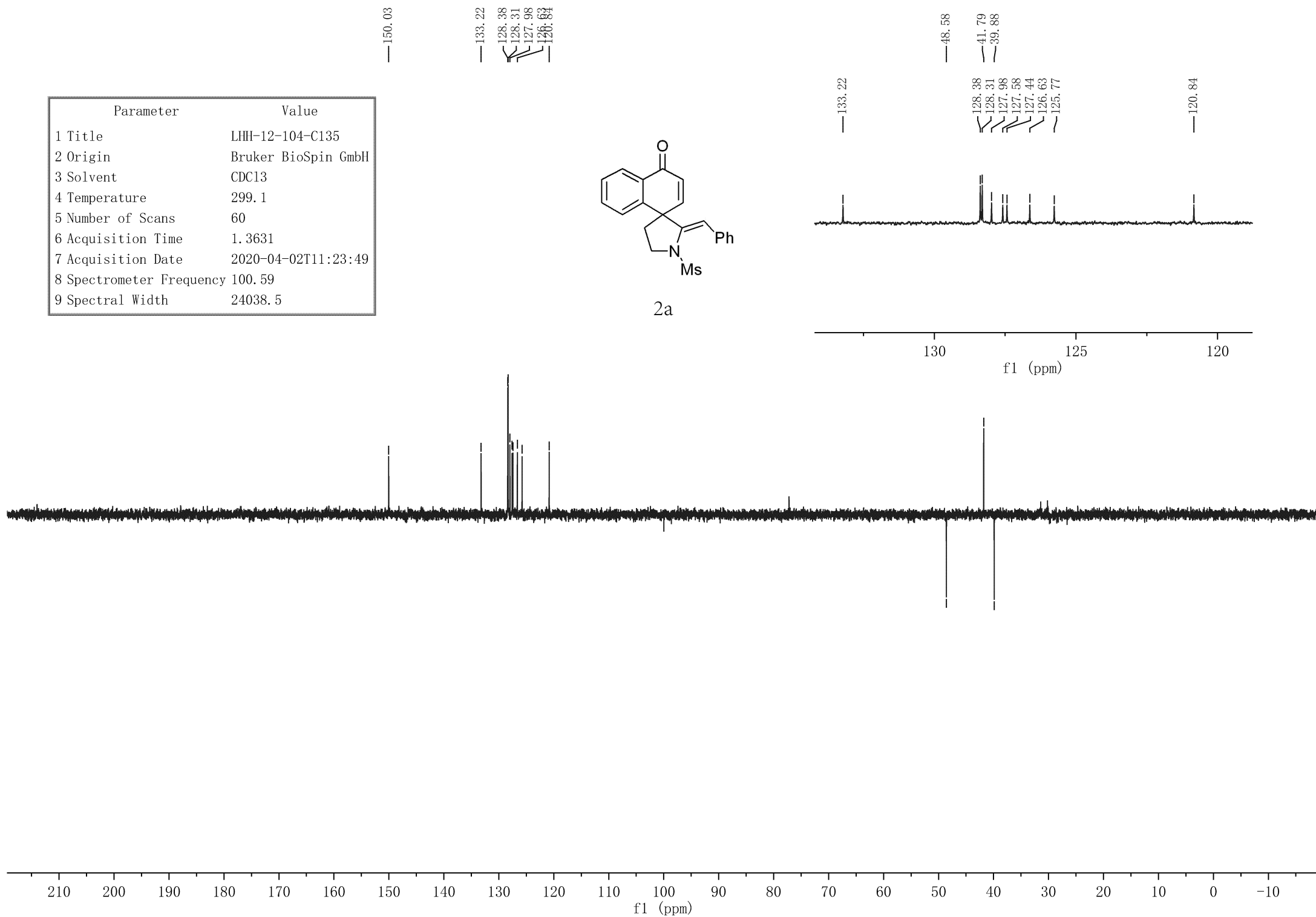
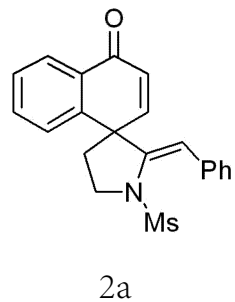
2a



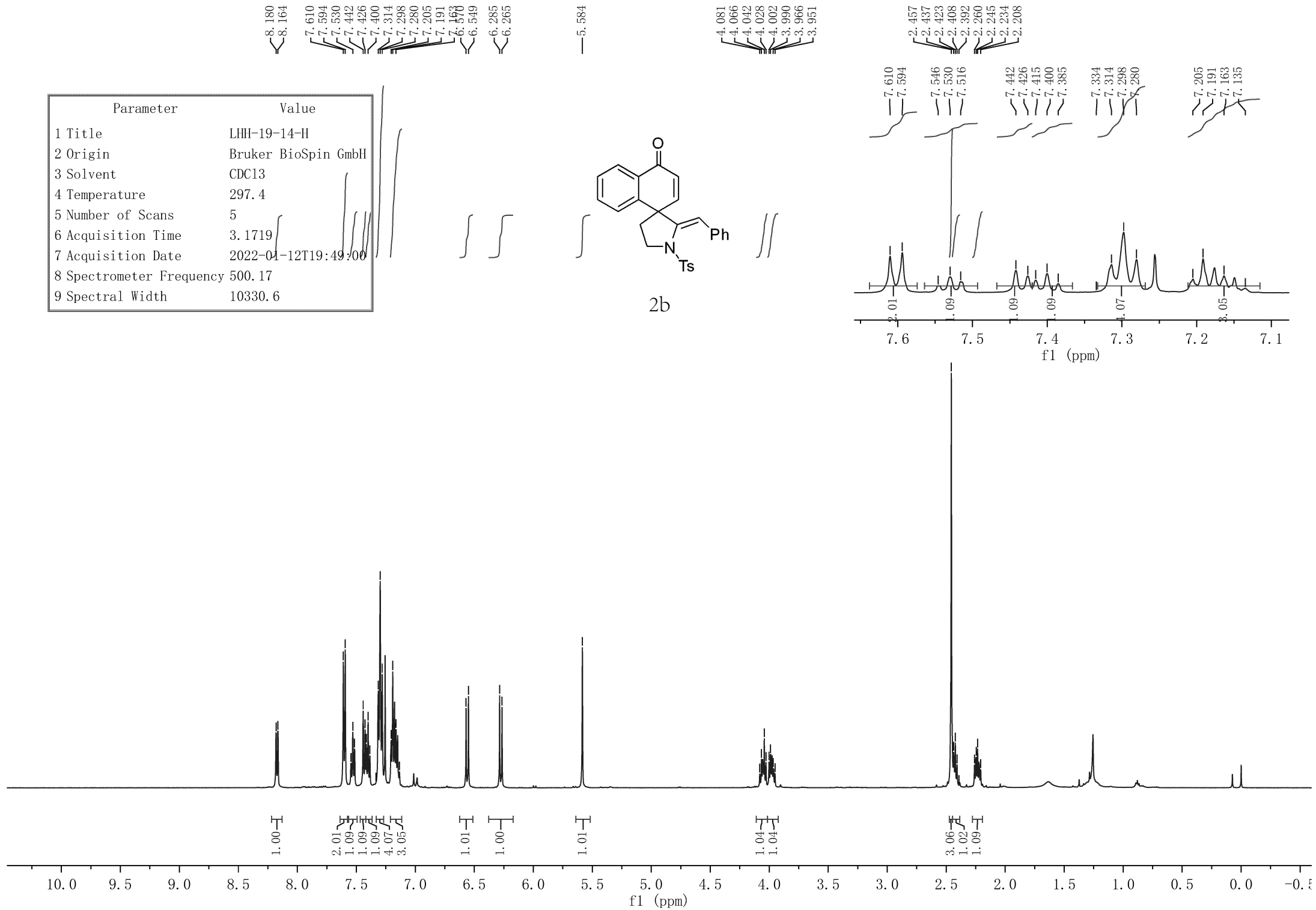
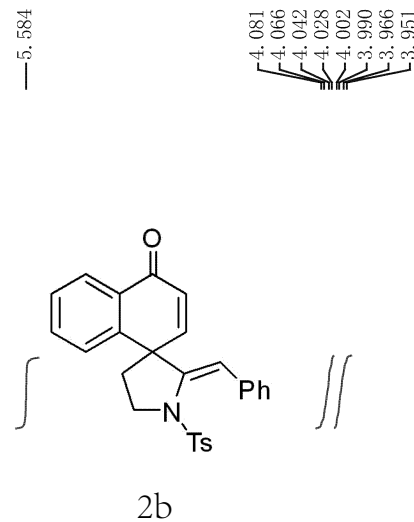
Parameter	Value
1 Title	LHH-12-104-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.4
5 Number of Scans	132
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-02T11:15:21
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



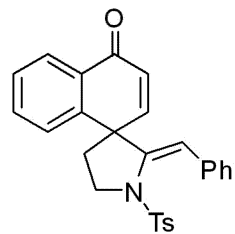
Parameter	Value
1 Title	LHH-12-104-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	299.1
5 Number of Scans	60
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-02T11:23:49
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



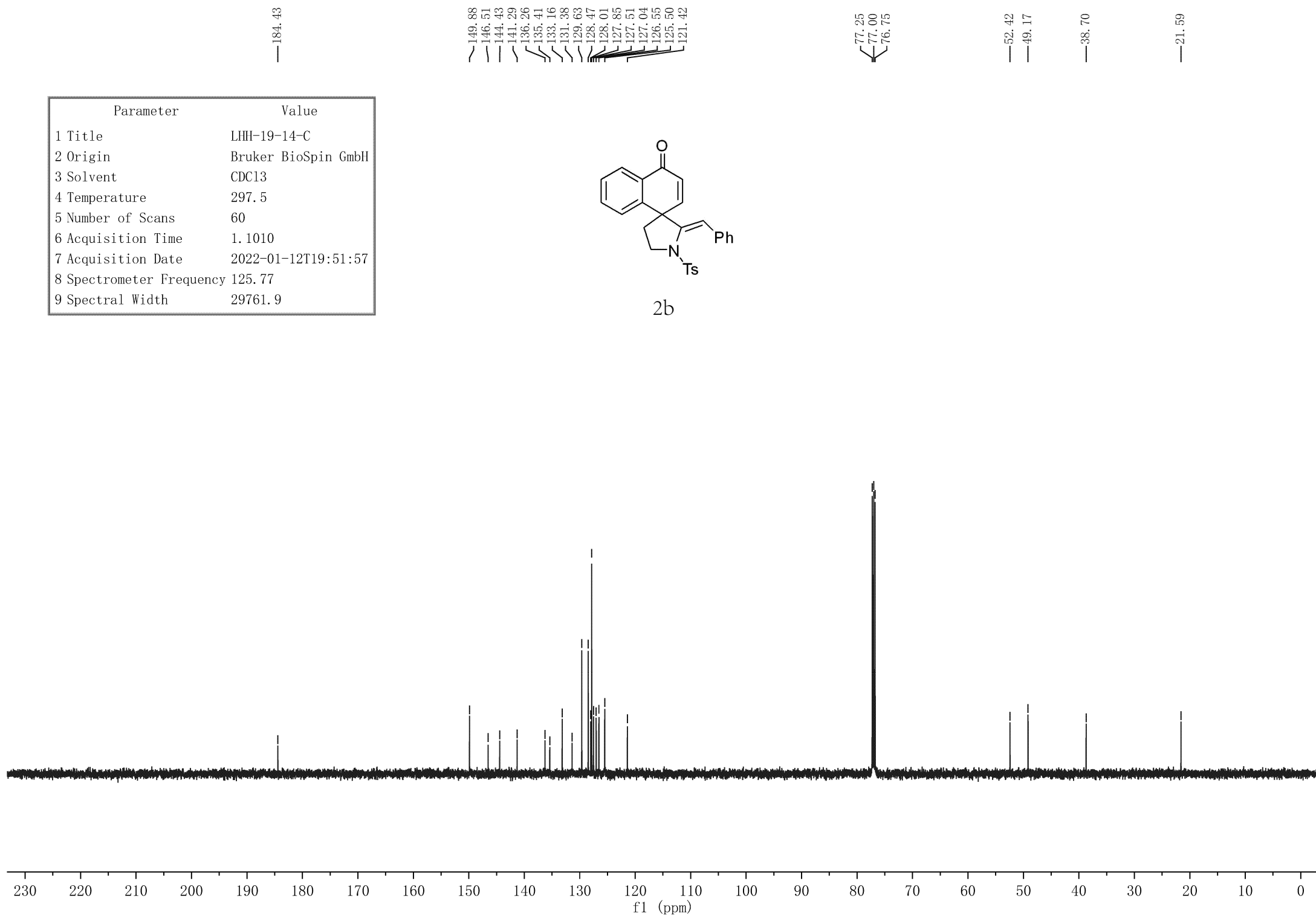
Parameter	Value
1 Title	LHH-19-14-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.4
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-04-12T19:49:00
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



Parameter	Value
1 Title	LHH-19-14-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.5
5 Number of Scans	60
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-12T19:51:57
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9

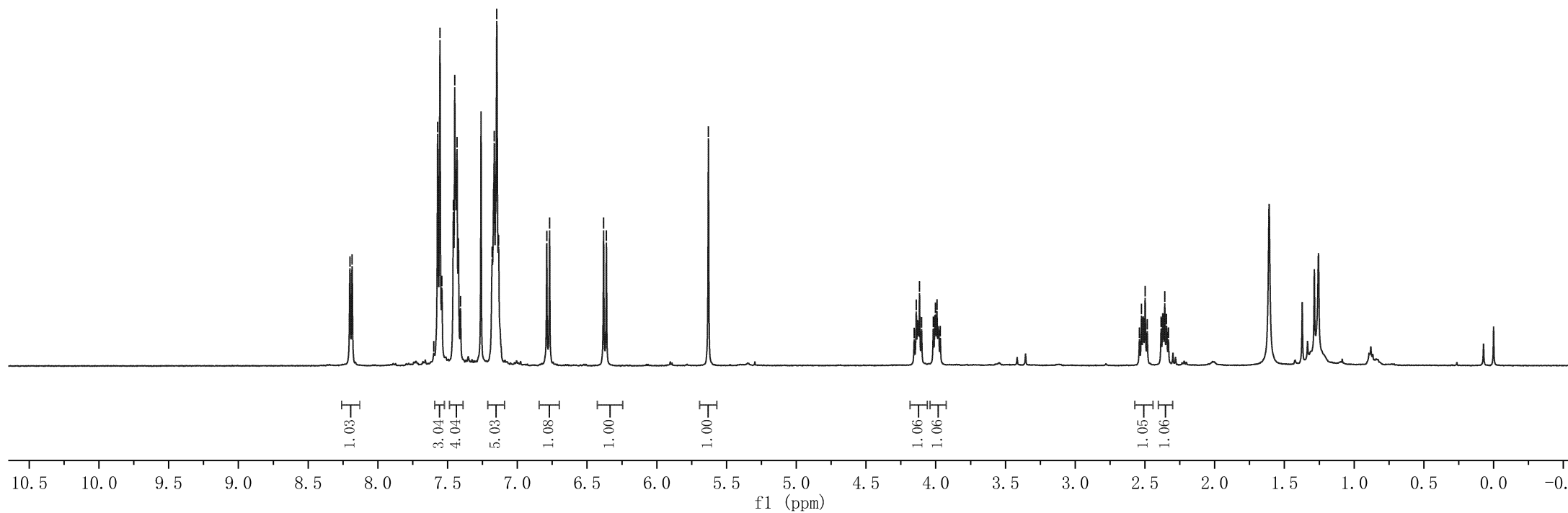
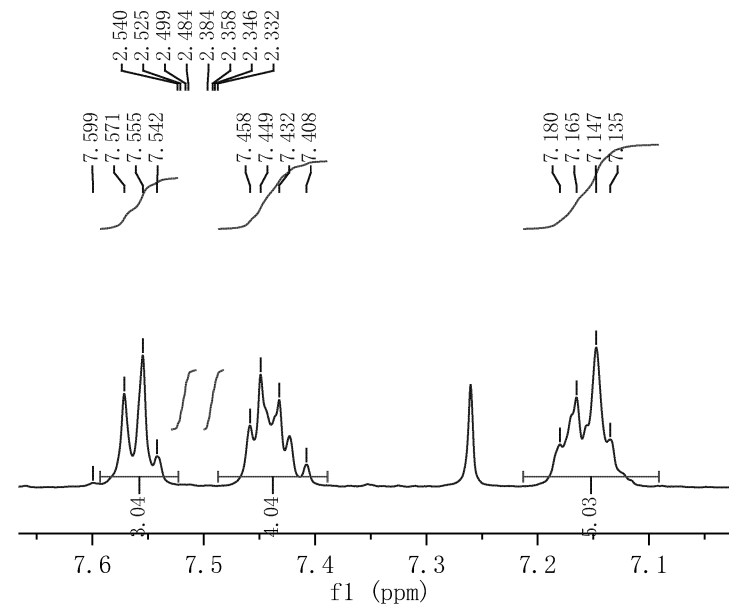
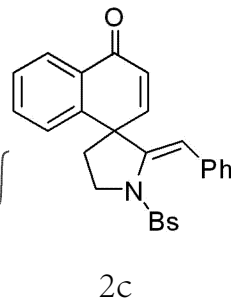


2b

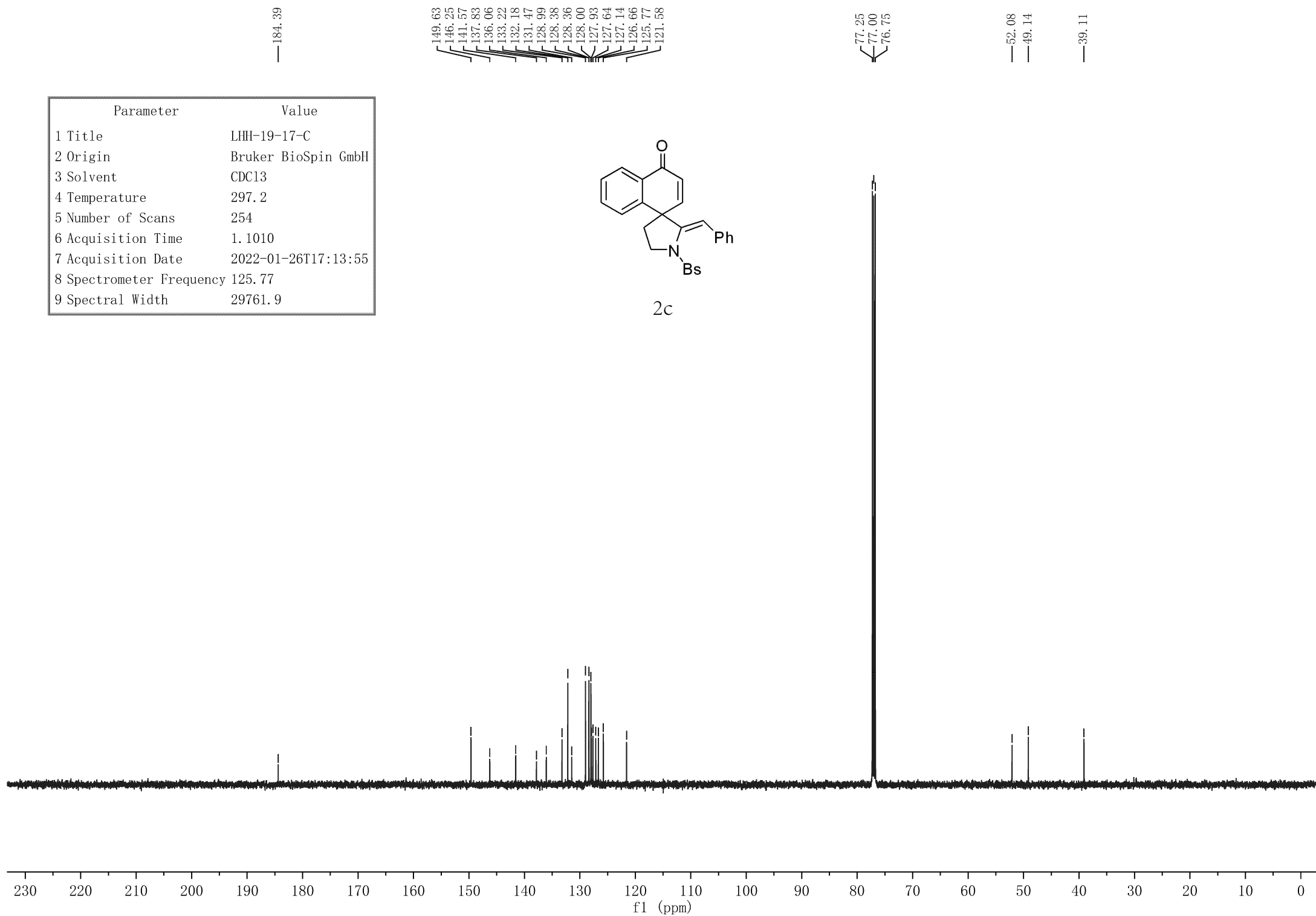
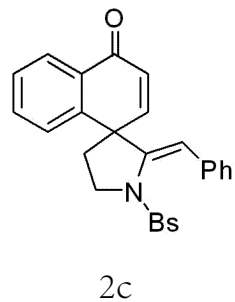


Parameter	Value
1 Title	LHH-19-17
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.0
5 Number of Scans	6
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-26T17:11:14
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.200 8.185 7.571 7.555 7.542 7.458 7.449 7.432 7.408 7.165 7.147 7.135 6.789 6.769 6.382 6.361 5.630 4.155 4.140 4.116 4.102 4.018 4.002 3.992 3.967



Parameter	Value
1 Title	LHH-19-17-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.2
5 Number of Scans	254
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-26T17:13:55
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



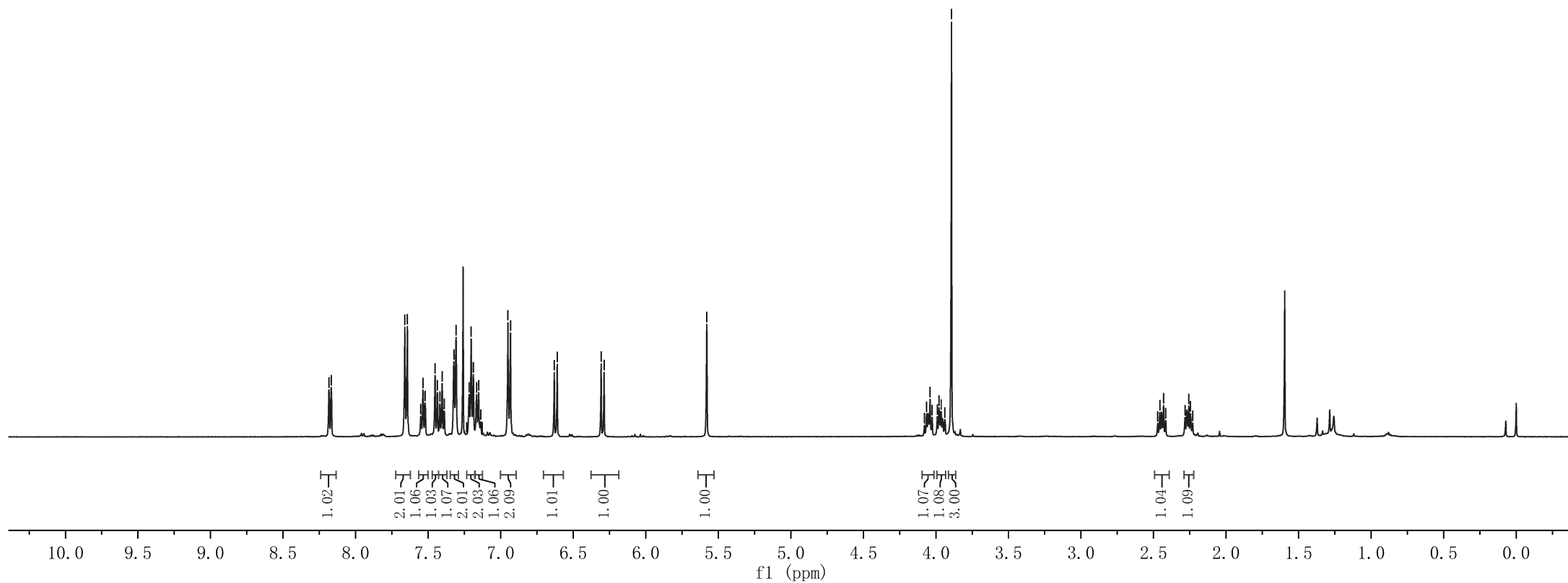
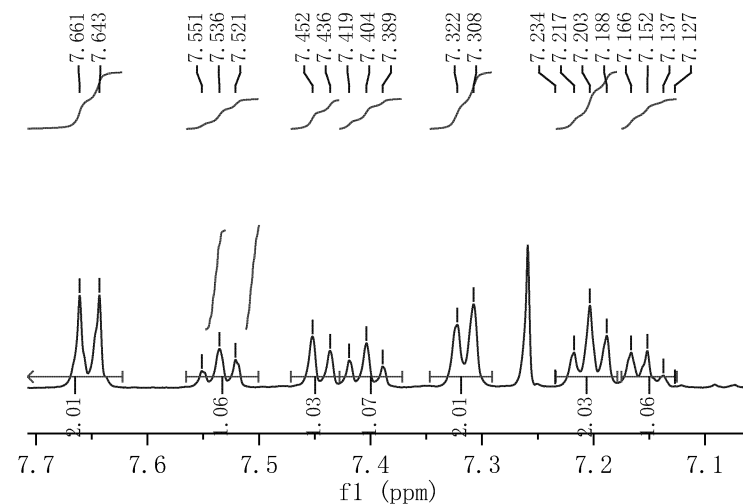
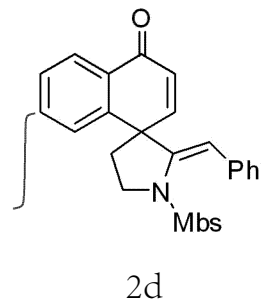
Parameter	Value
1 Title	LHH-19-59
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-15T14:22:12
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.184
8.168
7.661
7.643
7.536
7.452
7.436
7.404
7.322
7.308
7.203
7.188
6.950
6.832
6.817
6.610
6.307
6.287

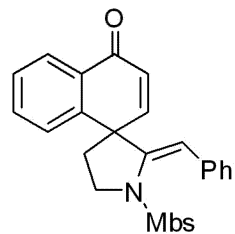
5.579

4.079
4.064
4.040
4.026
3.990
3.978
3.963
3.939
3.893

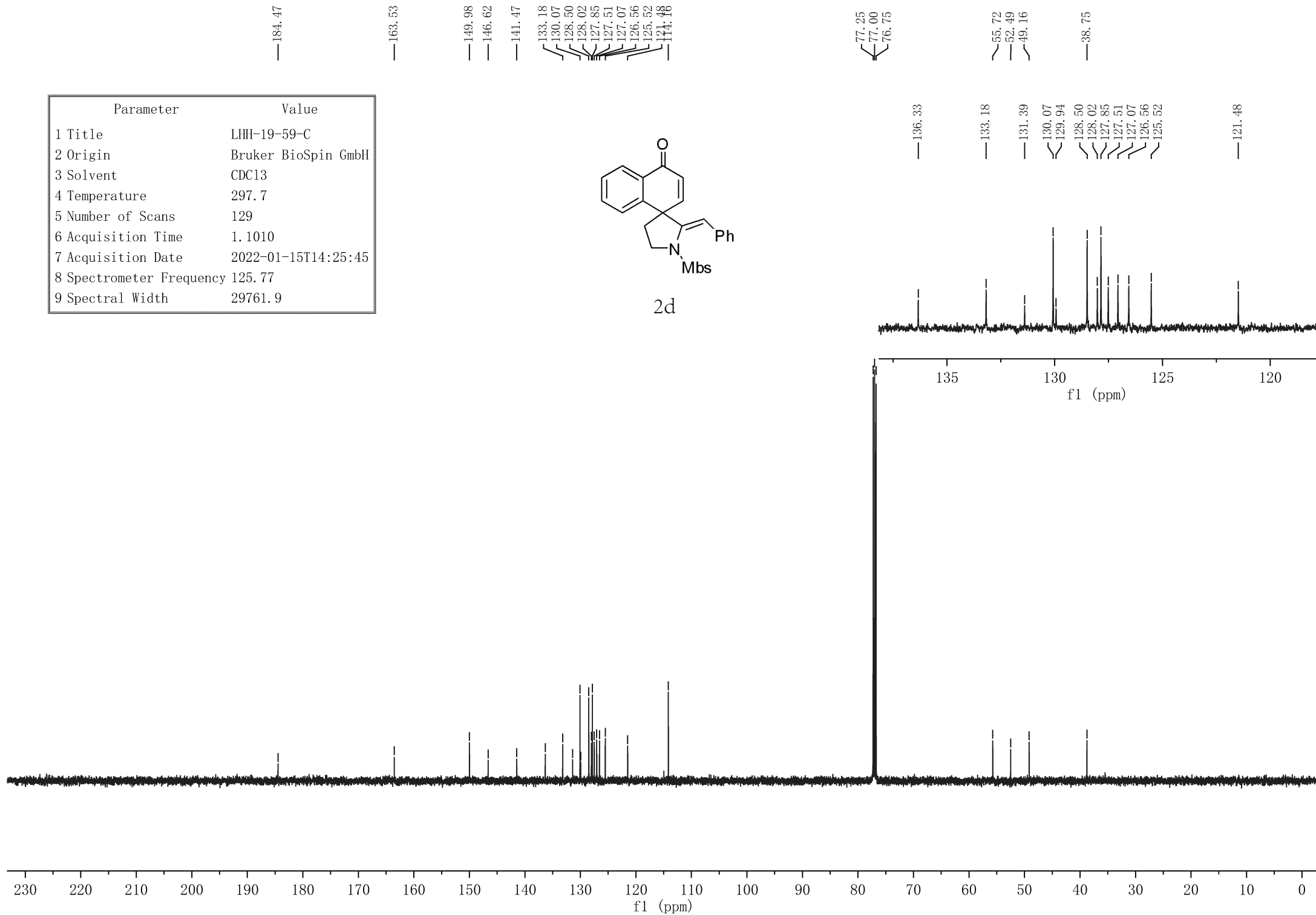
2.471
2.457
2.431
2.416
2.283
2.257
2.245
2.230



Parameter	Value
1 Title	LHH-19-59-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.7
5 Number of Scans	129
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-15T14:25:45
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9

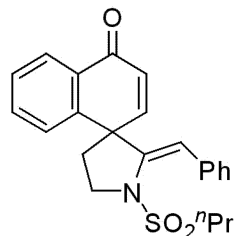


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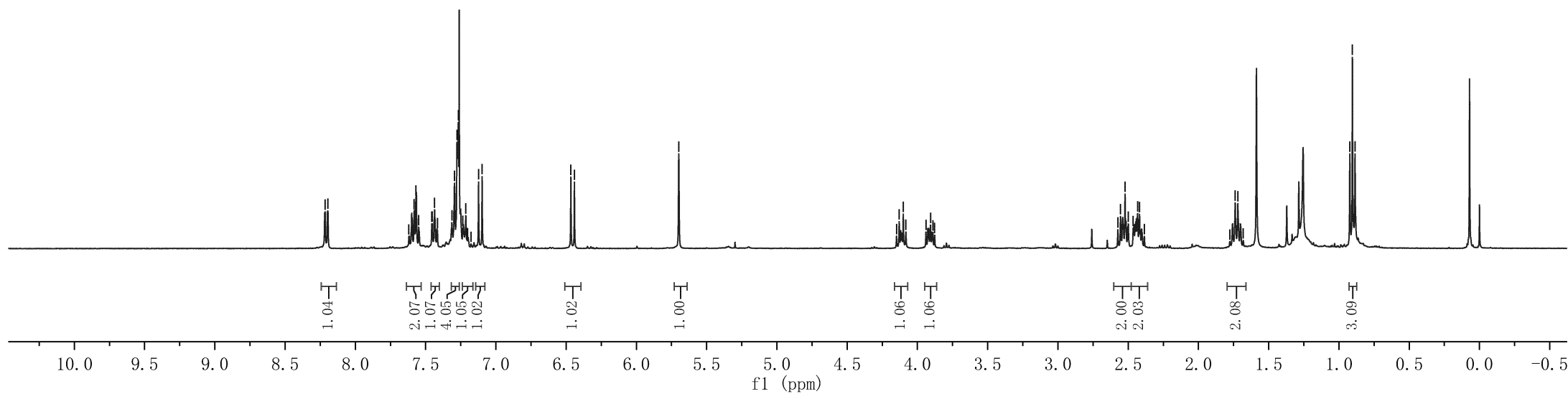
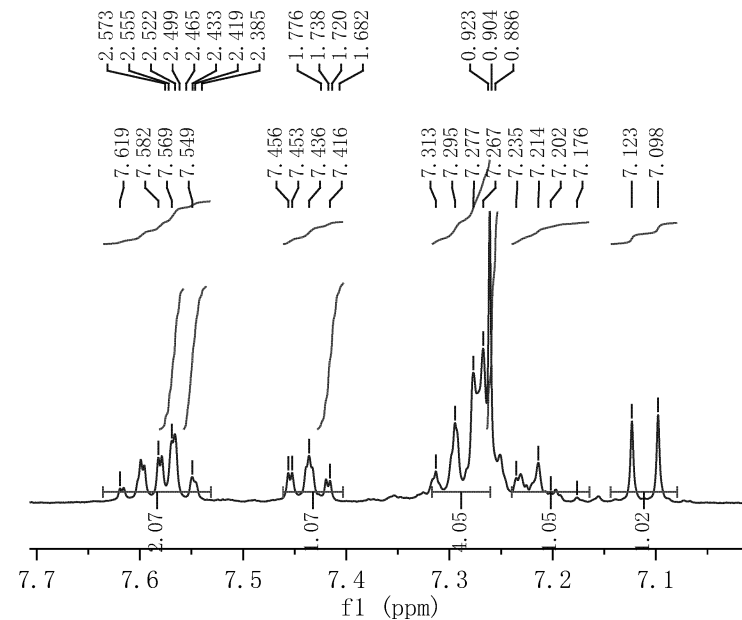


Parameter	Value
1 Title	LHH-19-78
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-20T09:21:48
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

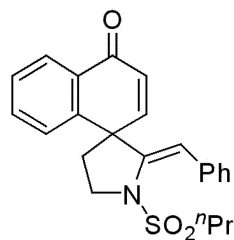
5.698



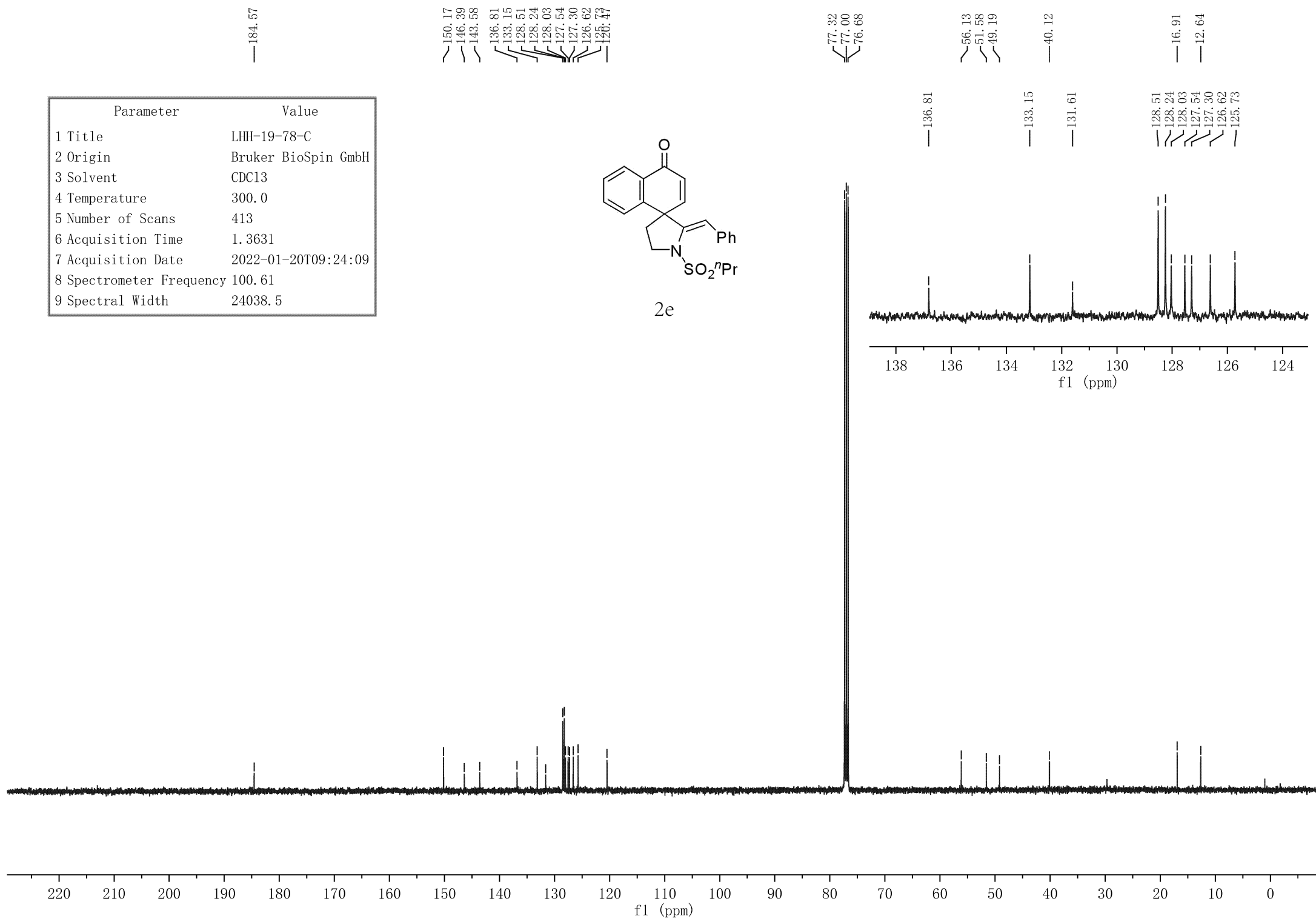
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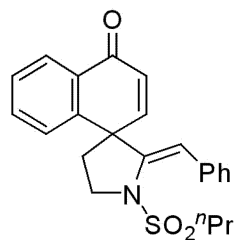
Parameter	Value
1 Title	LHH-19-78-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	413
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-20T09:24:09
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



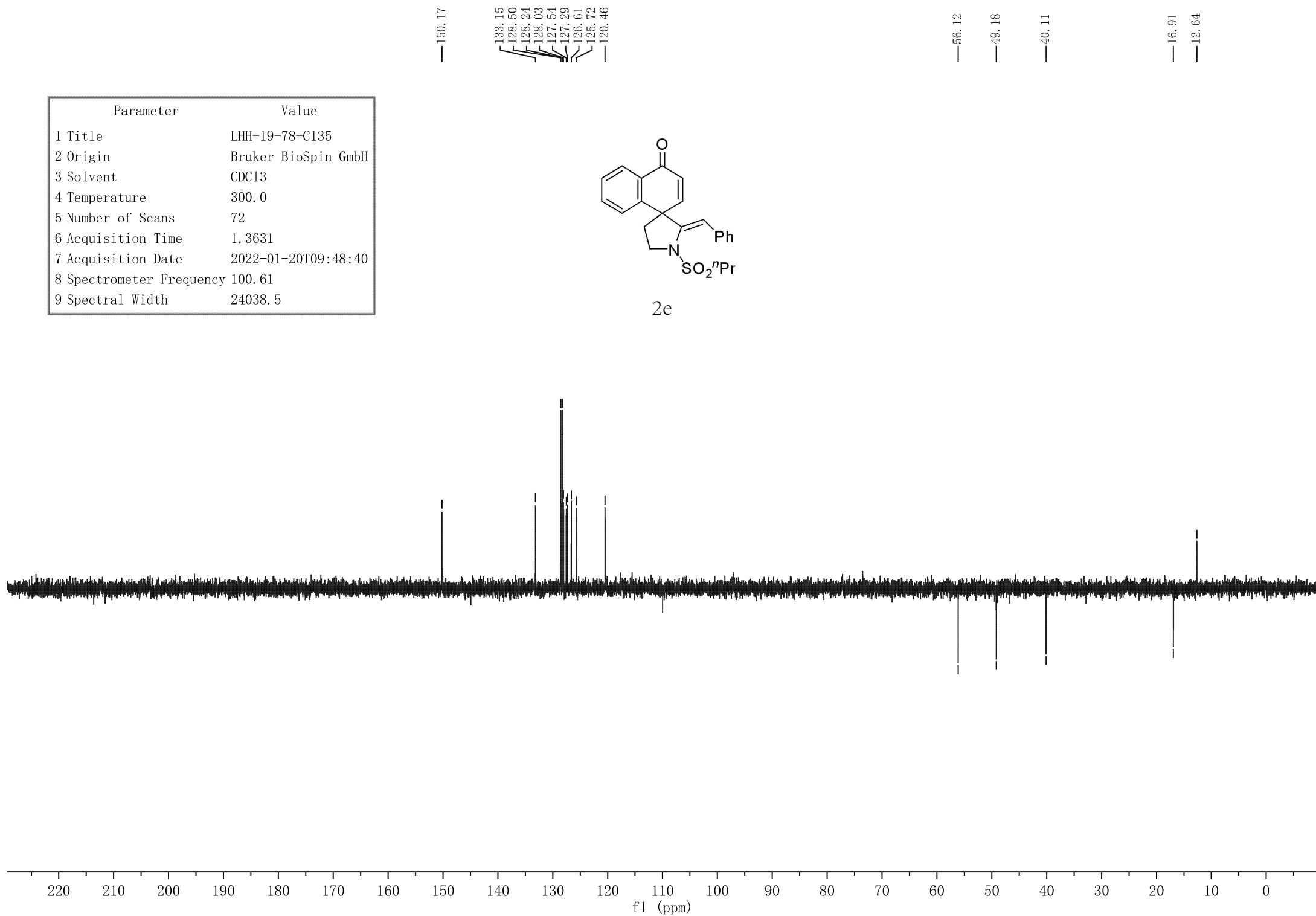
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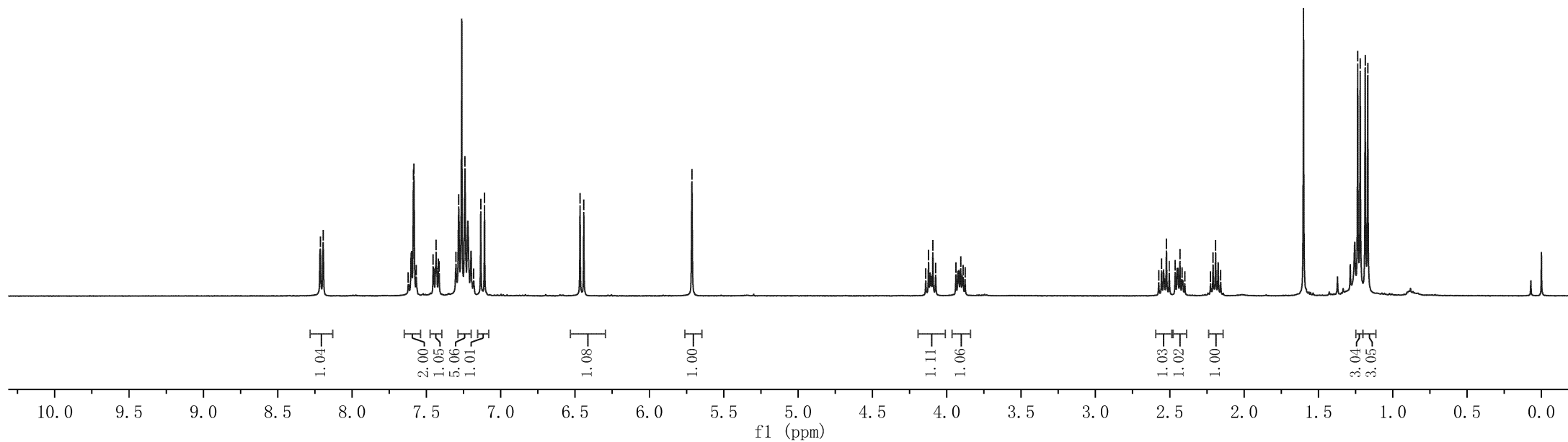
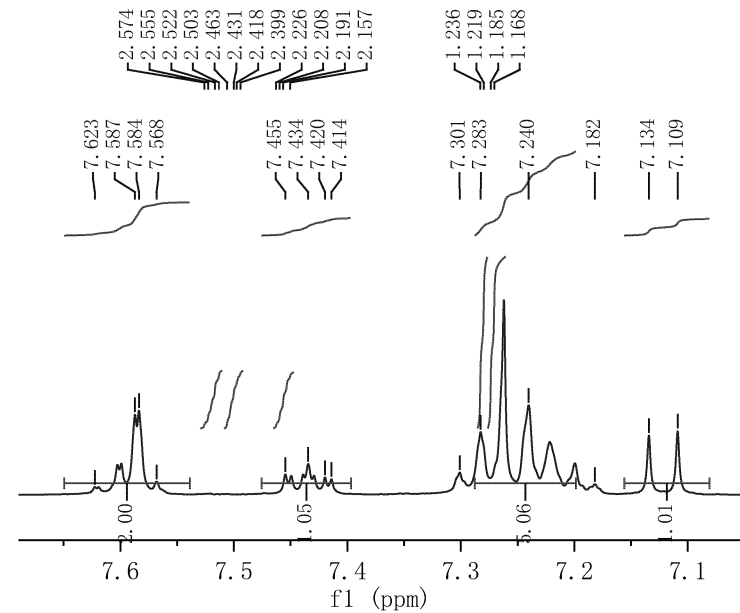
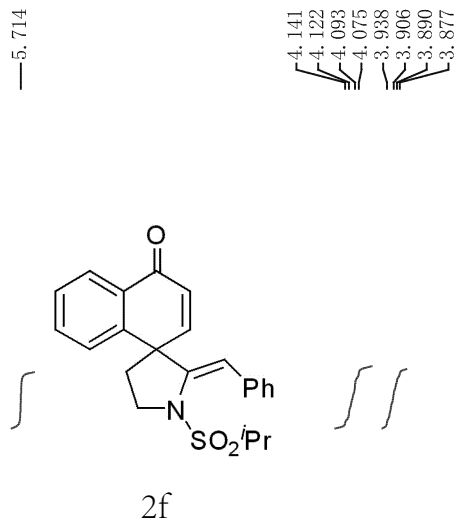
Parameter	Value
1 Title	LHH-19-78-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	72
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-20T09:48:40
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



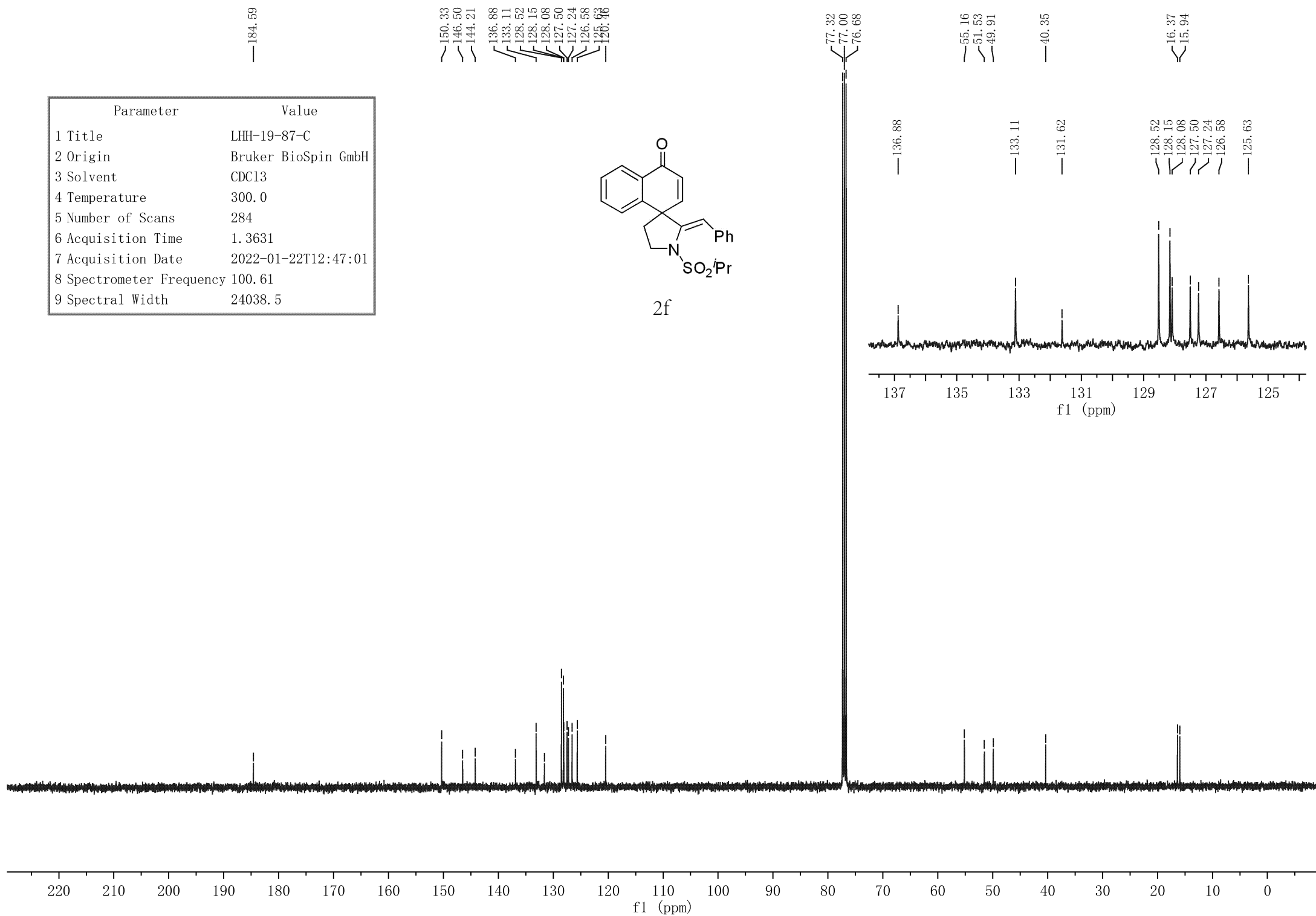
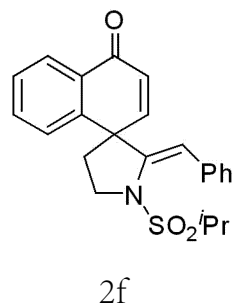
2e



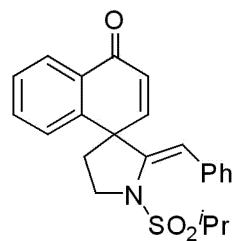
Parameter	Value
1 Title	LHH-19-87
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	6
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-22T12:40:53
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



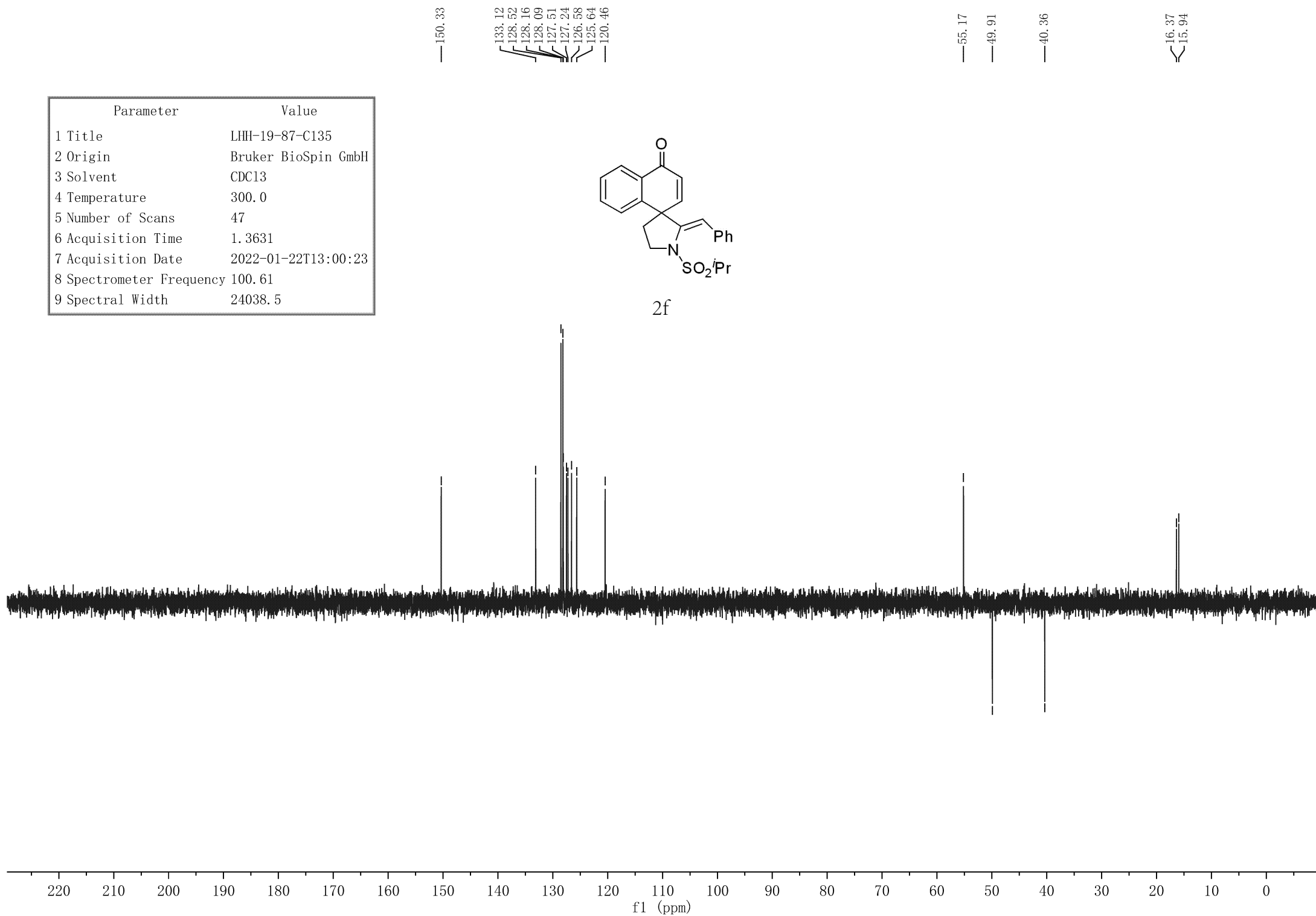
Parameter	Value
1 Title	LHH-19-87-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	284
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-22T12:47:01
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-87-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	47
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-22T13:00:23
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



2f

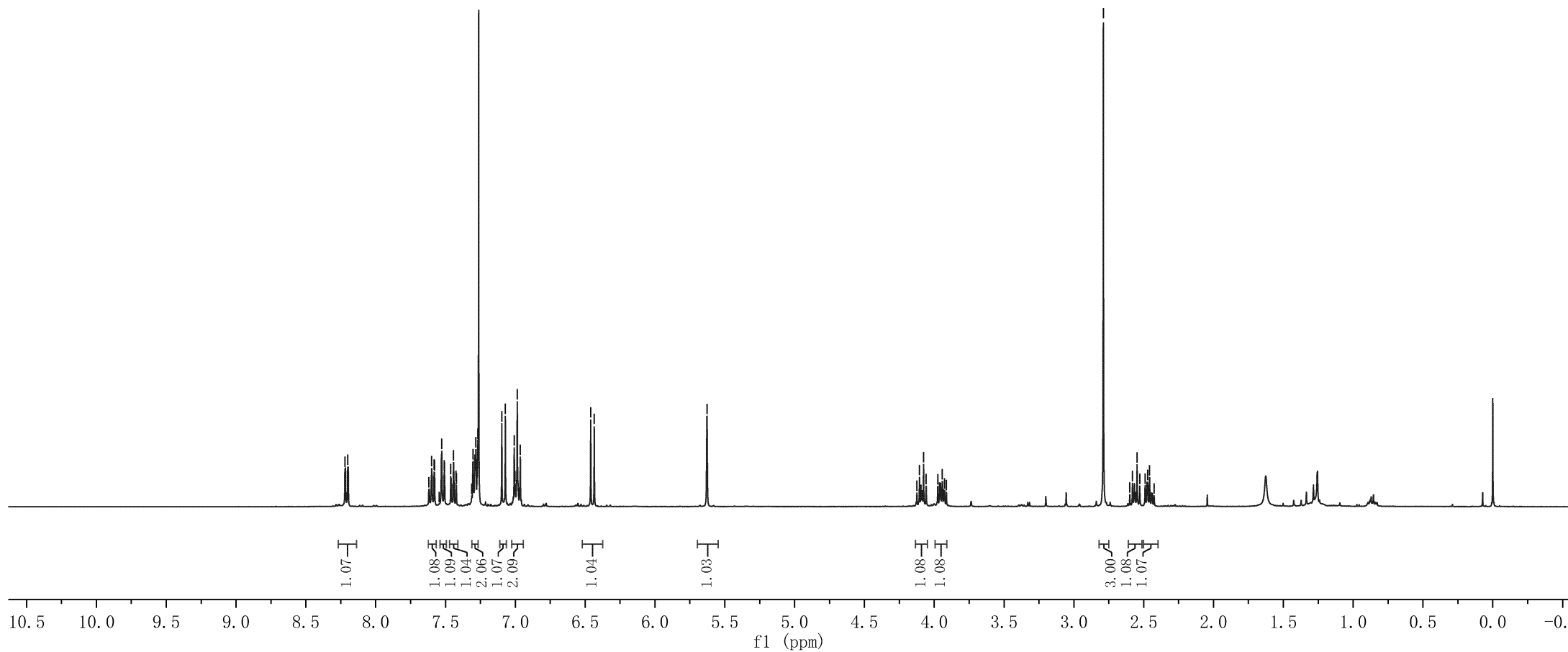
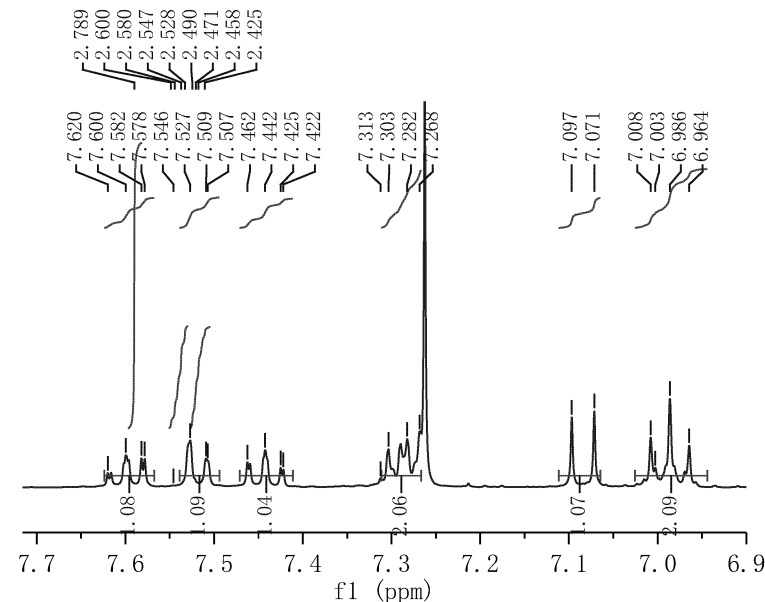
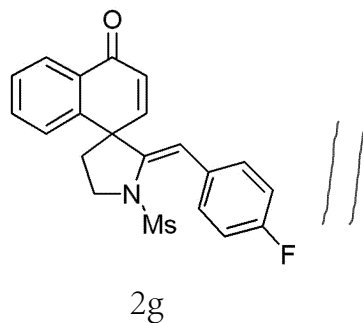


Parameter	Value
1 Title	LHH-12-142'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.5
5 Number of Scans	8
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-13T19:32:42
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7

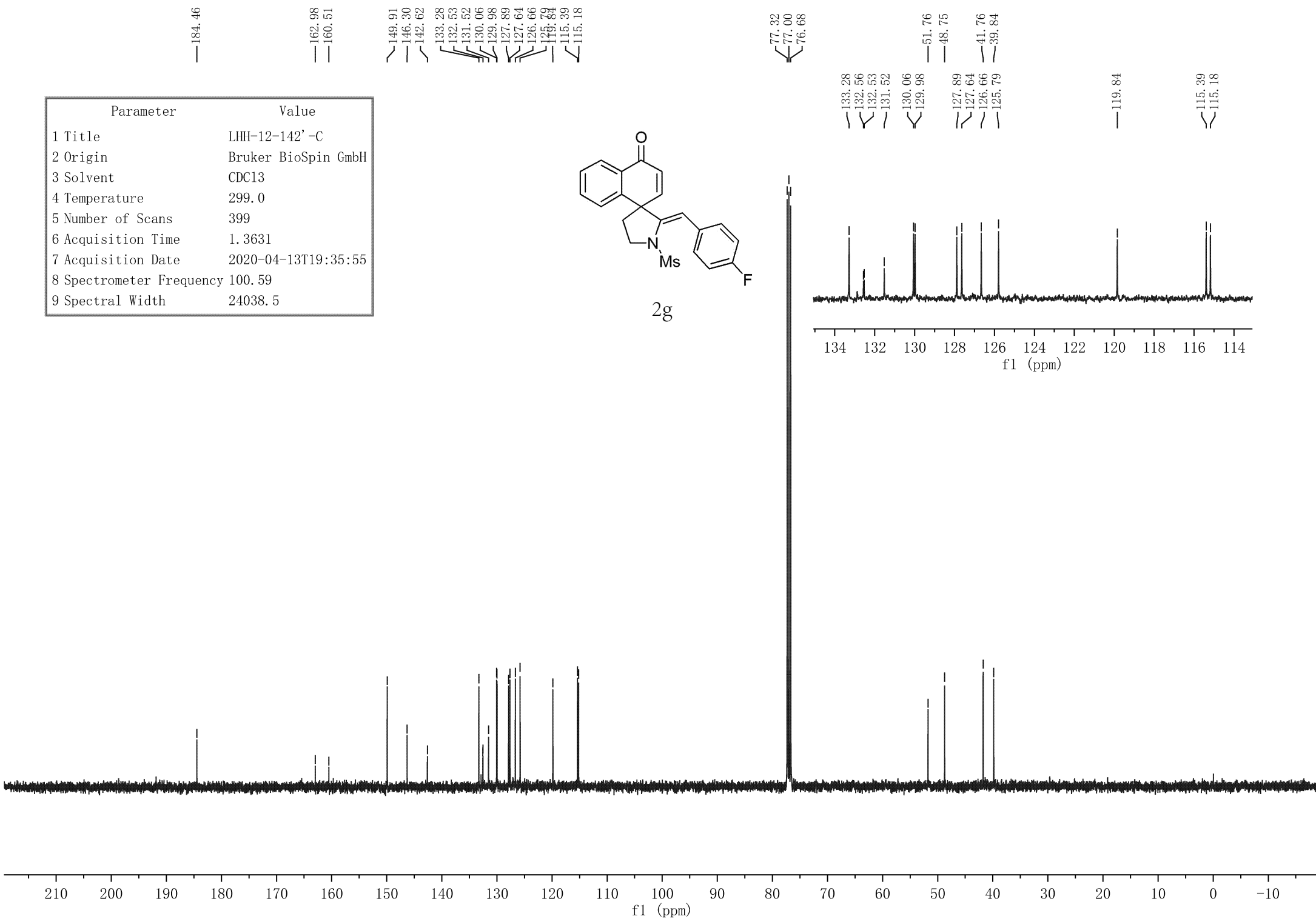
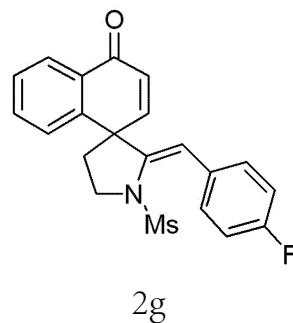
8.220
8.200
7.600
7.582
7.578
7.527
7.442
7.303
7.282
7.268
7.097
7.071
7.008
6.986
6.964
6.960
6.435

5.628

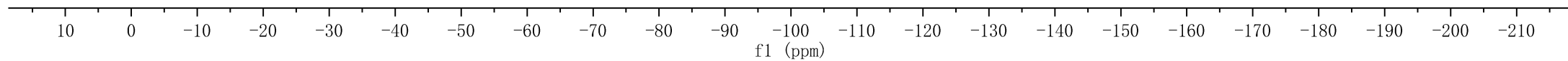
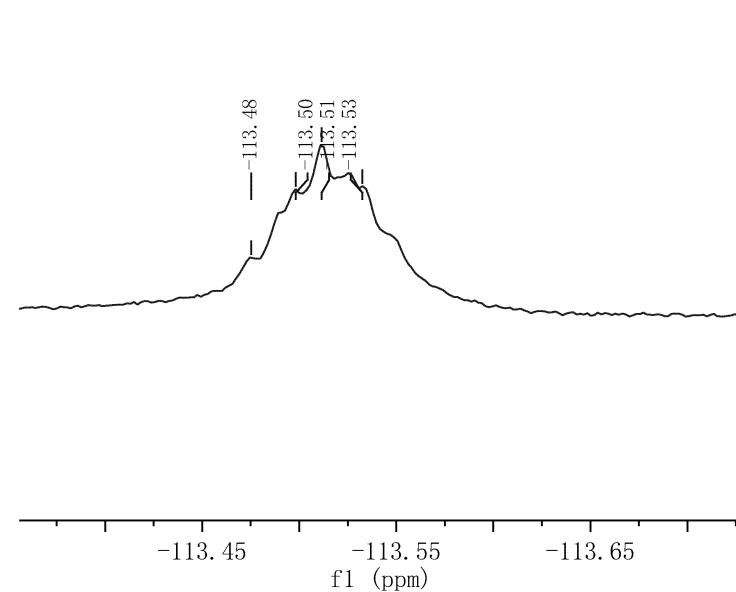
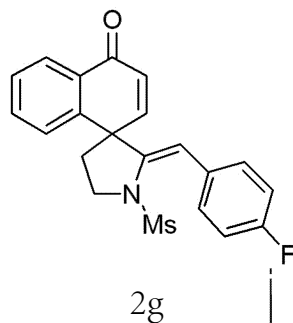
4.124
4.106
4.076
4.058
3.975
3.942
3.926
3.913



Parameter	Value
1 Title	LHH-12-142'-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	299.0
5 Number of Scans	399
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-13T19:35:55
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



Parameter	Value
1 Title	zyp-1-94-F'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.2
5 Number of Scans	16
6 Acquisition Time	0.7340
7 Acquisition Date	2022-01-04T21:15:17
8 Spectrometer Frequency	376.31
9 Spectral Width	89285.7



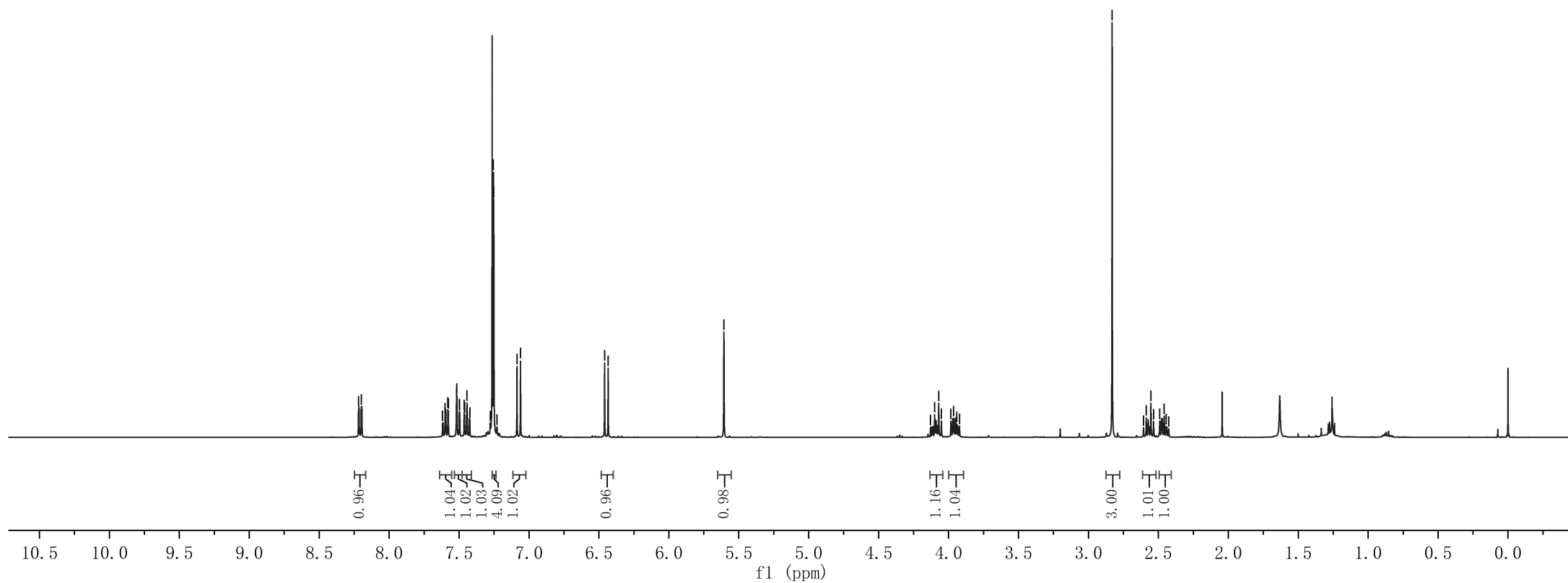
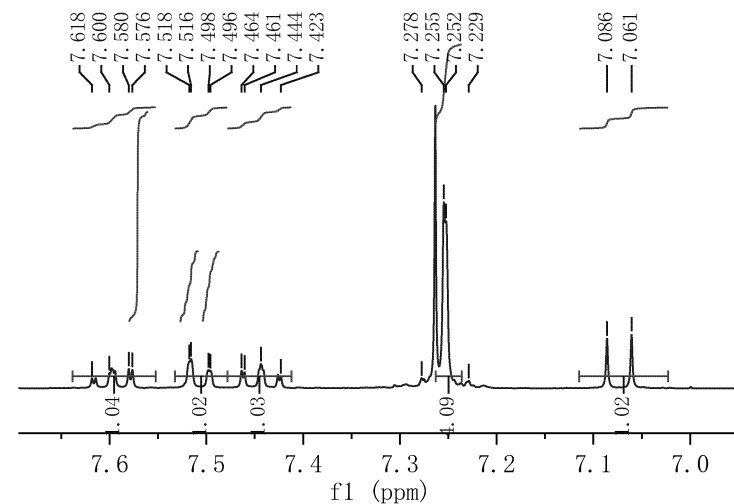
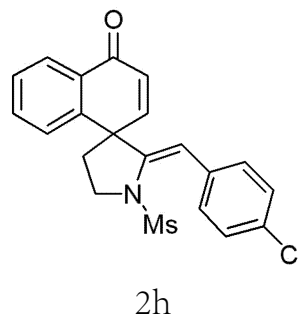
Parameter	Value
1 Title	LHH-12-155
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.4
5 Number of Scans	12
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-16T14:52:27
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7

8.218
8.199
7.580
7.576
7.518
7.516
7.498
7.496
7.464
7.461
7.444
7.255
7.252
7.086
7.051
6.434

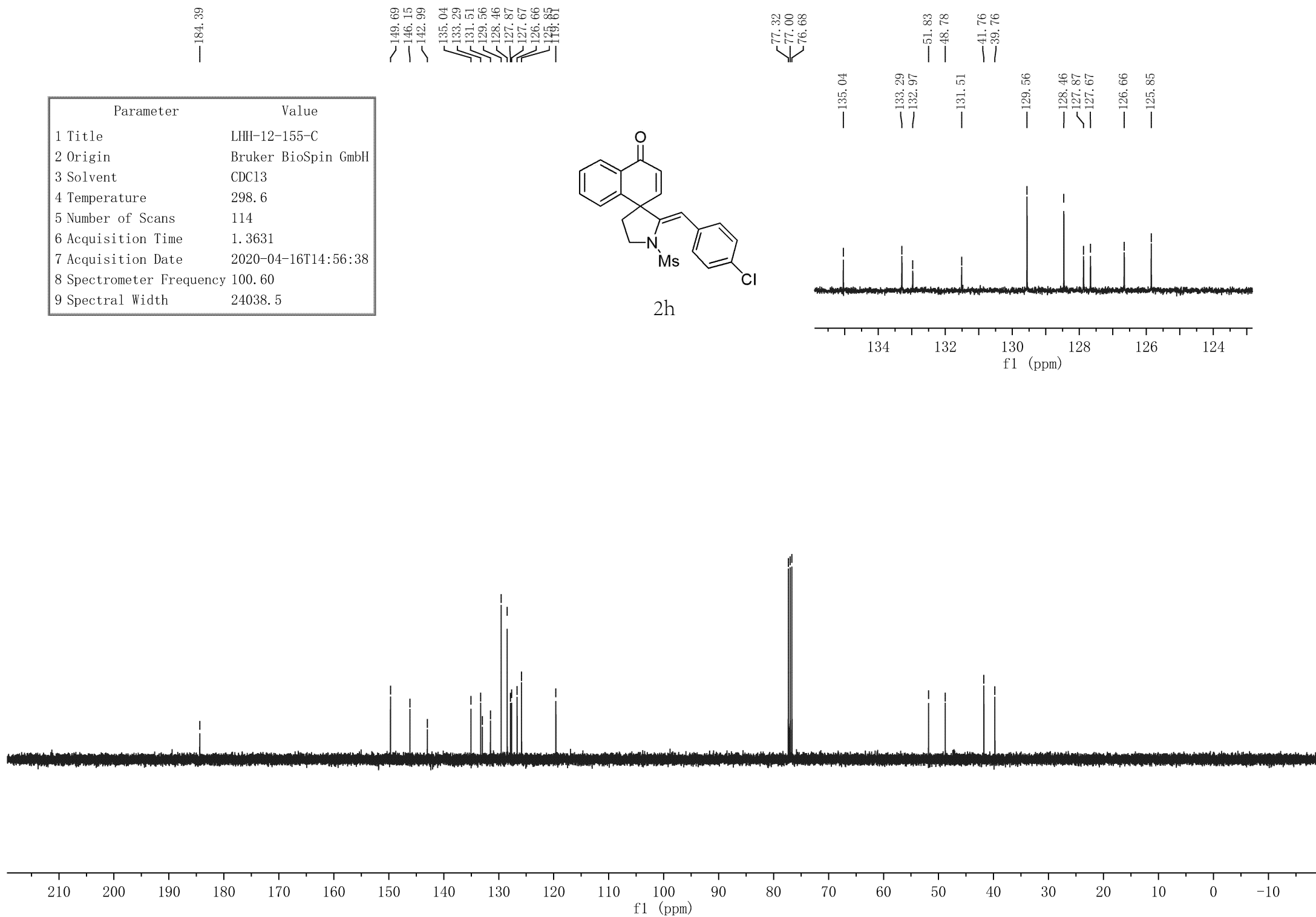
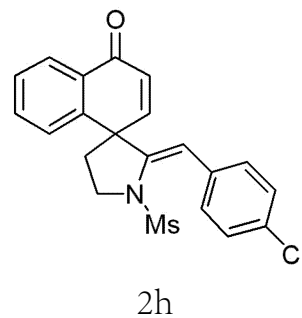
5.606

4.129
4.100
4.070
4.052
3.983
3.964
3.941
3.922

2.830
2.606
2.586
2.554
2.534
2.490
2.459
2.445
2.426



Parameter	Value
1 Title	LHH-12-155-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.6
5 Number of Scans	114
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-16T14:56:38
8 Spectrometer Frequency	100.60
9 Spectral Width	24038.5



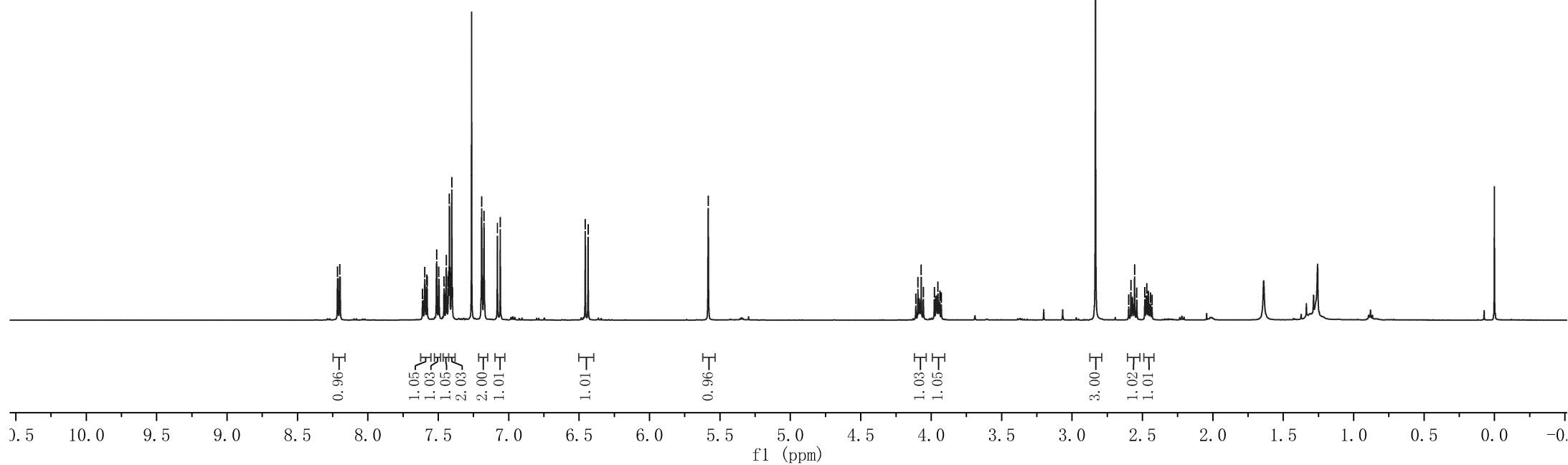
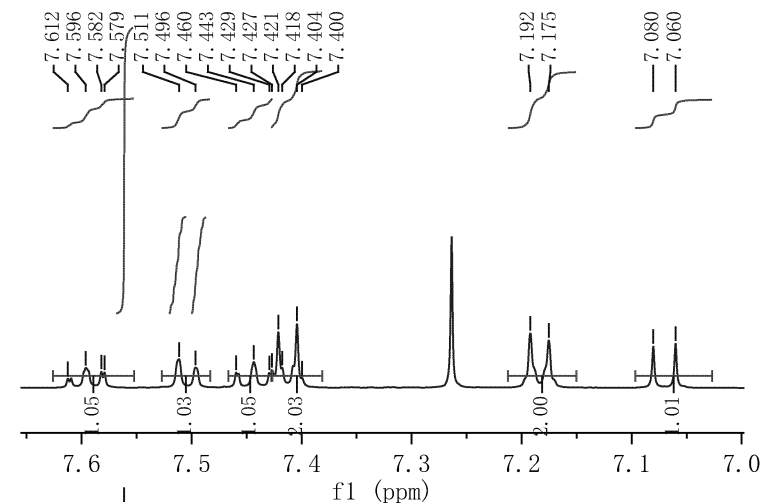
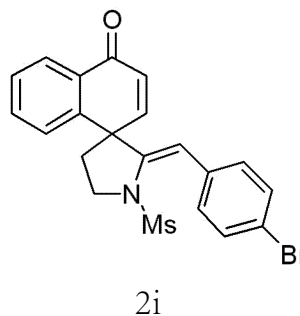
Parameter	Value
1 Title	LHH-12-140
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	299.1
5 Number of Scans	20
6 Acquisition Time	3.1719
7 Acquisition Date	2020-05-30T15:26:13
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

8.216
8.200
7.596
7.582
7.511
7.496
7.443
7.427
7.421
7.418
7.404
7.192
7.175
7.080
7.066
6.436

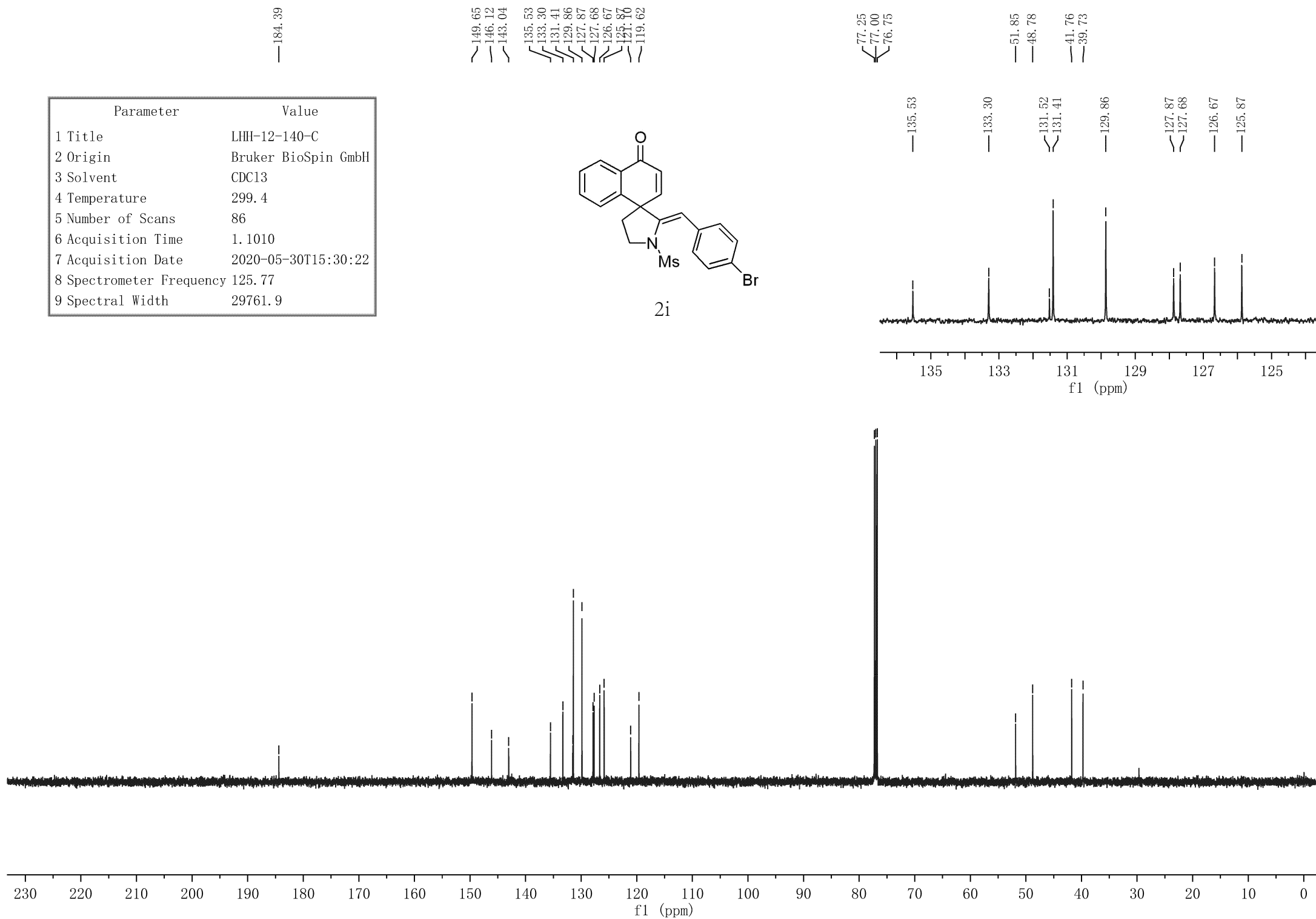
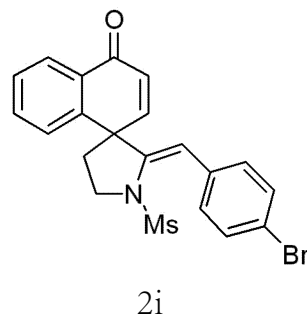
5.583

4.109
4.094
4.071
4.056
3.977
3.952
3.938
3.928

2.833
2.598
2.582
2.556
2.540
2.483
2.468
2.442
2.432



Parameter	Value
1 Title	LHH-12-140-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	299.4
5 Number of Scans	86
6 Acquisition Time	1.1010
7 Acquisition Date	2020-05-30T15:30:22
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



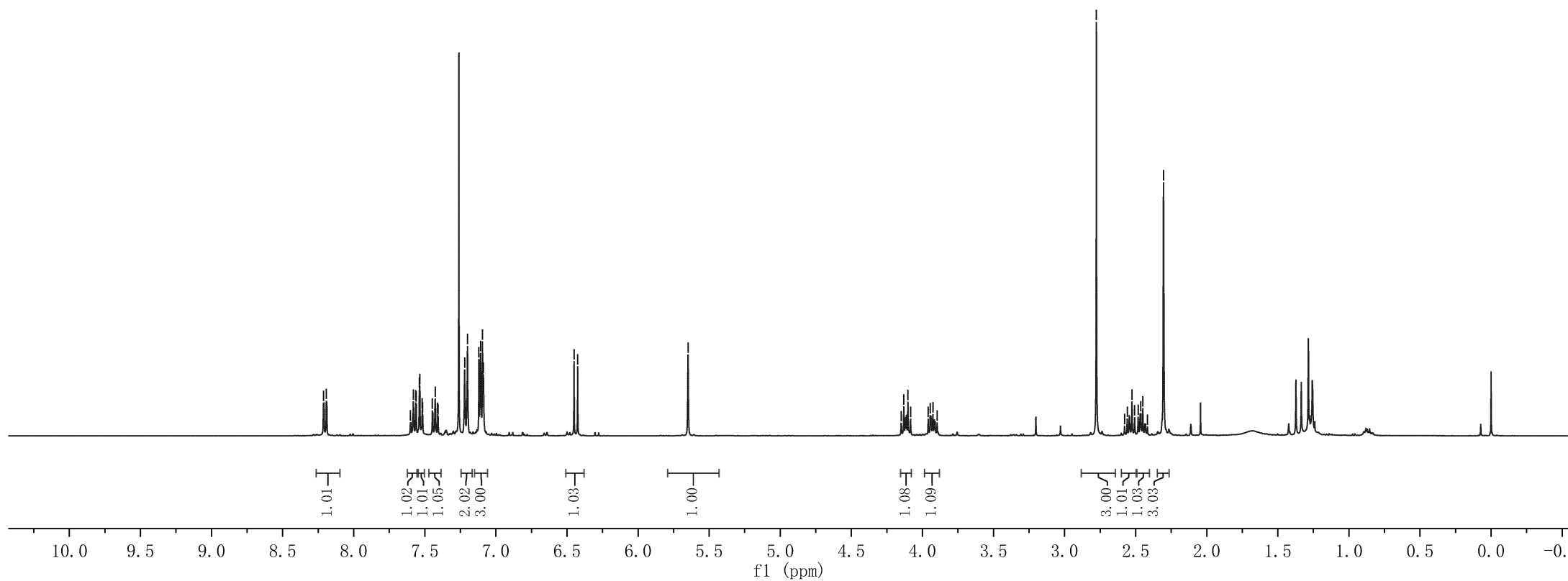
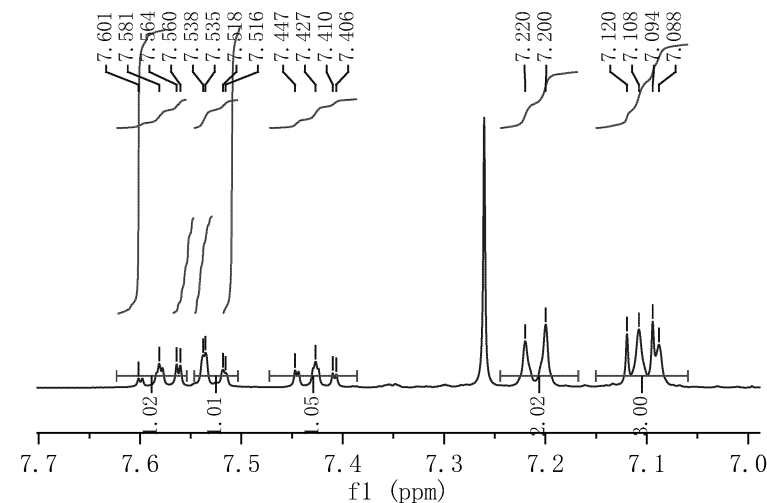
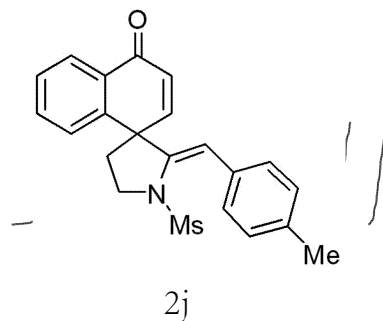
Parameter	Value
1 Title	LHH-12-178
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.1
5 Number of Scans	12
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-24T16:49:15
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7

8.213
8.193
7.581
7.564
7.560
7.538
7.535
7.518
7.447
7.427
7.220
7.200
7.120
7.108
7.094
7.088
6.425

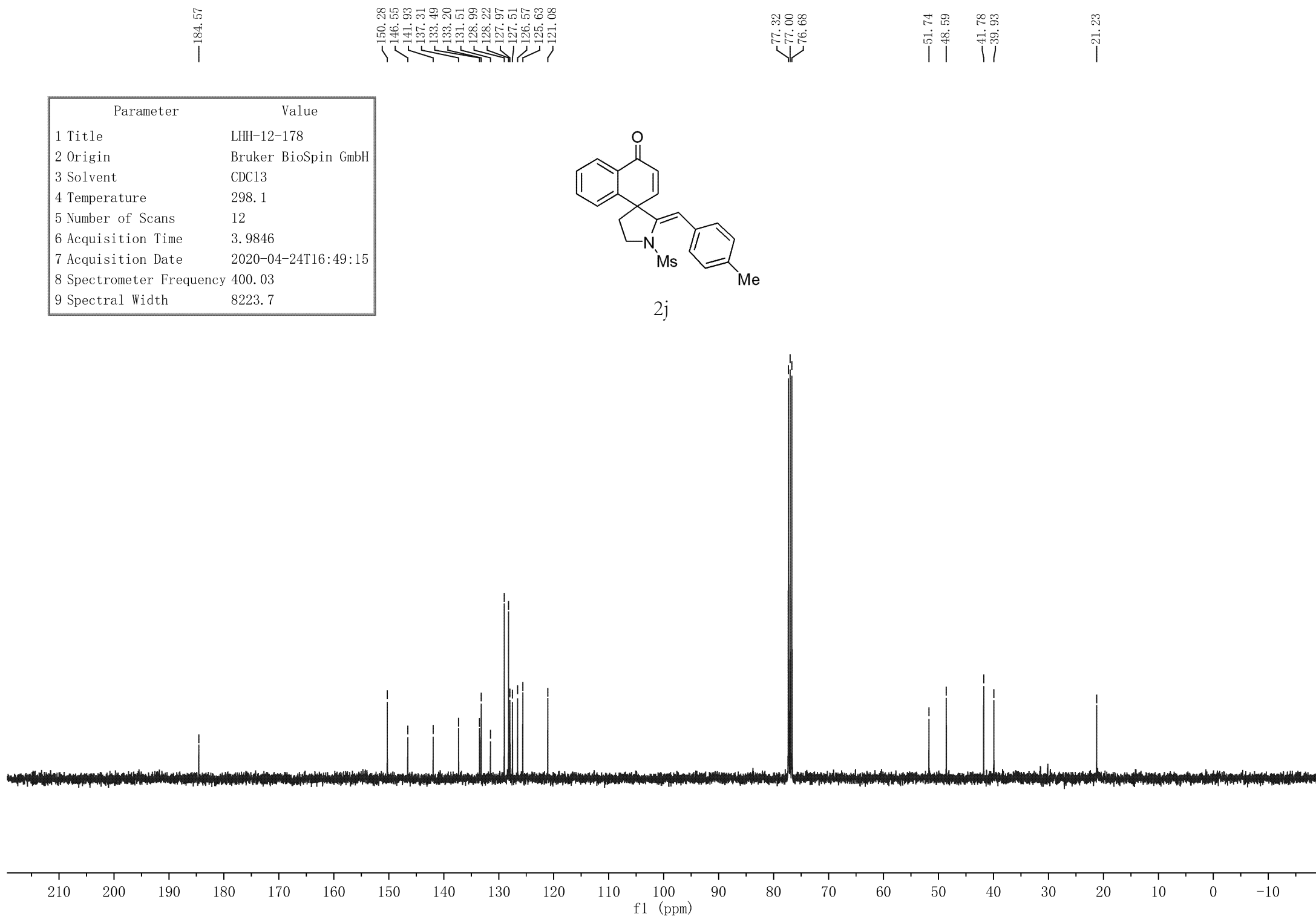
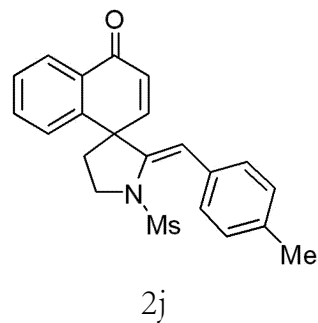
5.648

4.149
4.131
4.102
4.083
3.959
3.945
3.926
3.897

2.776
2.578
2.559
2.526
2.507
2.482
2.464
2.450
2.417
2.304



Parameter	Value
1 Title	LHH-12-178
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.1
5 Number of Scans	12
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-24T16:49:15
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7



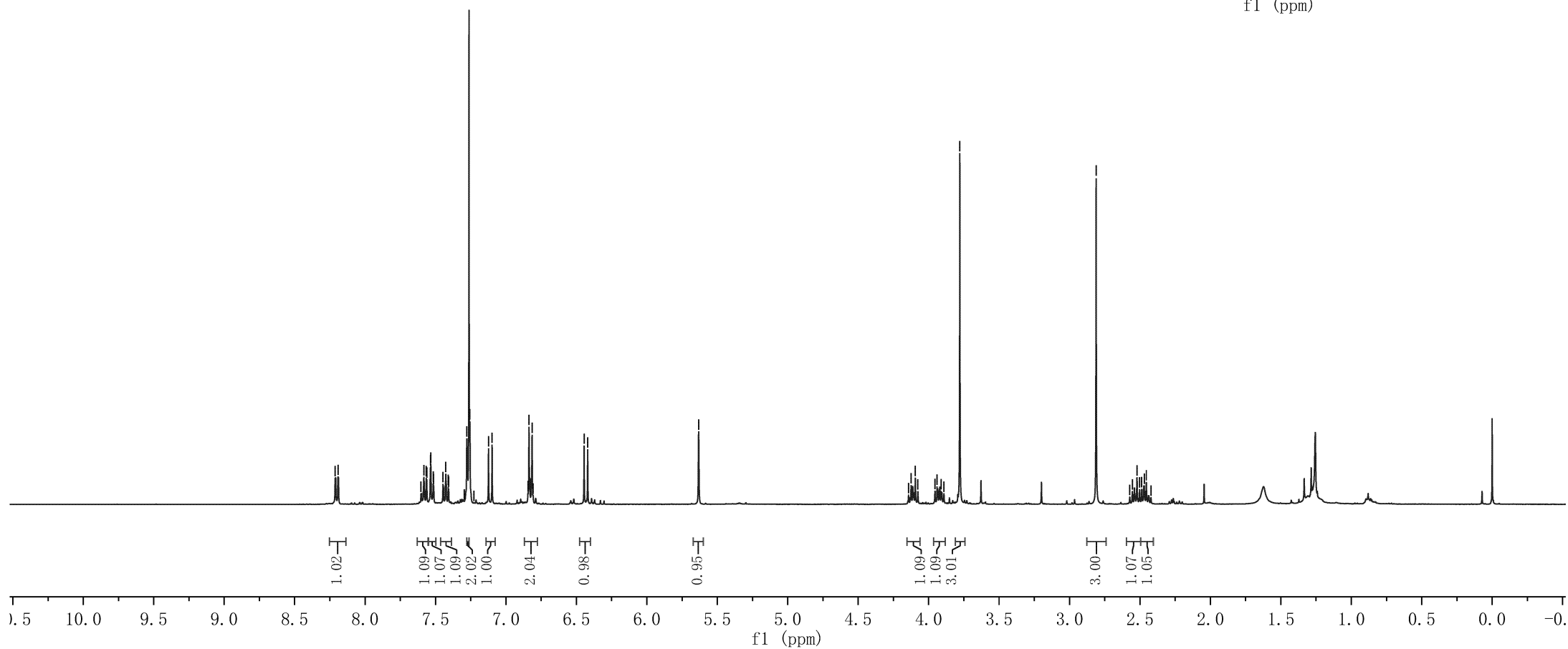
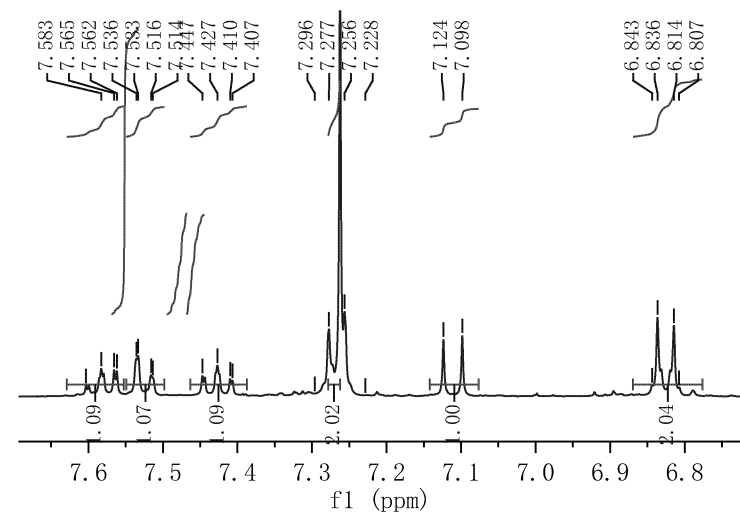
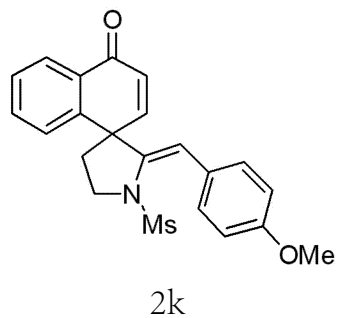
Parameter	Value
1 Title	LHH-12-159
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.2
5 Number of Scans	11
6 Acquisition Time	3.9846
7 Acquisition Date	2020-04-17T11:07:44
8 Spectrometer Frequency	400.03
9 Spectral Width	8223.7

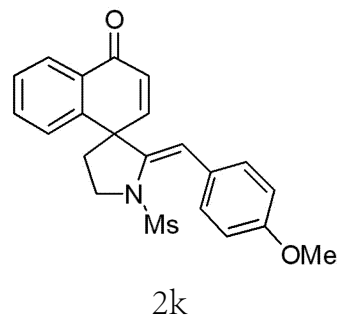
8.212
8.192
7.583
7.565
7.562
7.536
7.533
7.427
7.277
7.256
7.124
7.098
6.843
6.836
6.814
6.779
6.420

5.632

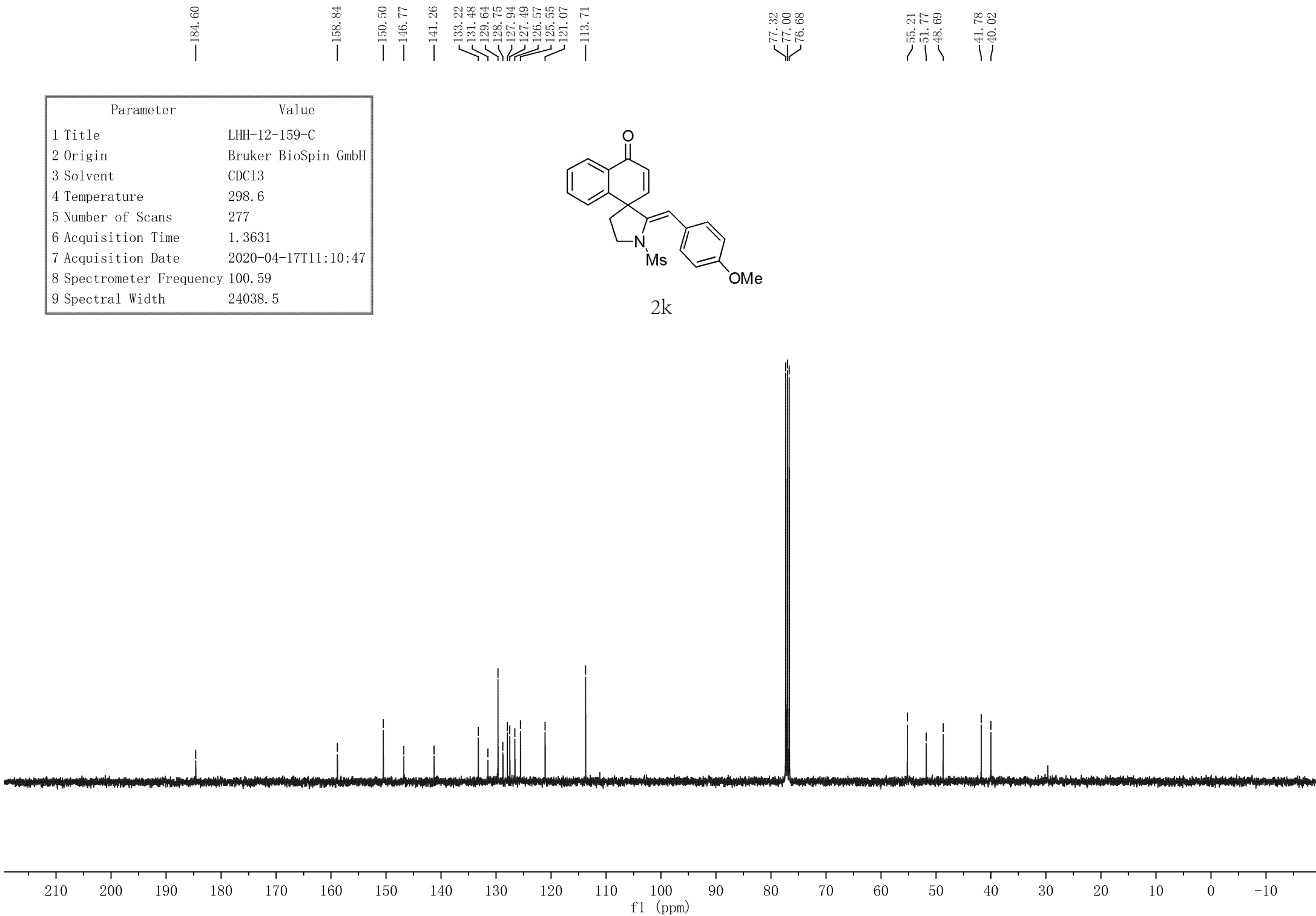
4.142
4.124
4.095
4.076
3.955
3.940
3.911
3.891
3.779

2.811
2.572
2.554
2.521
2.503
2.488
2.469
2.455
2.422

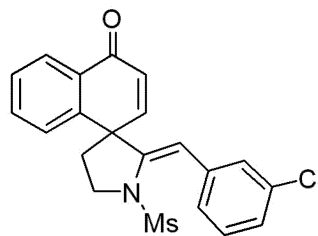




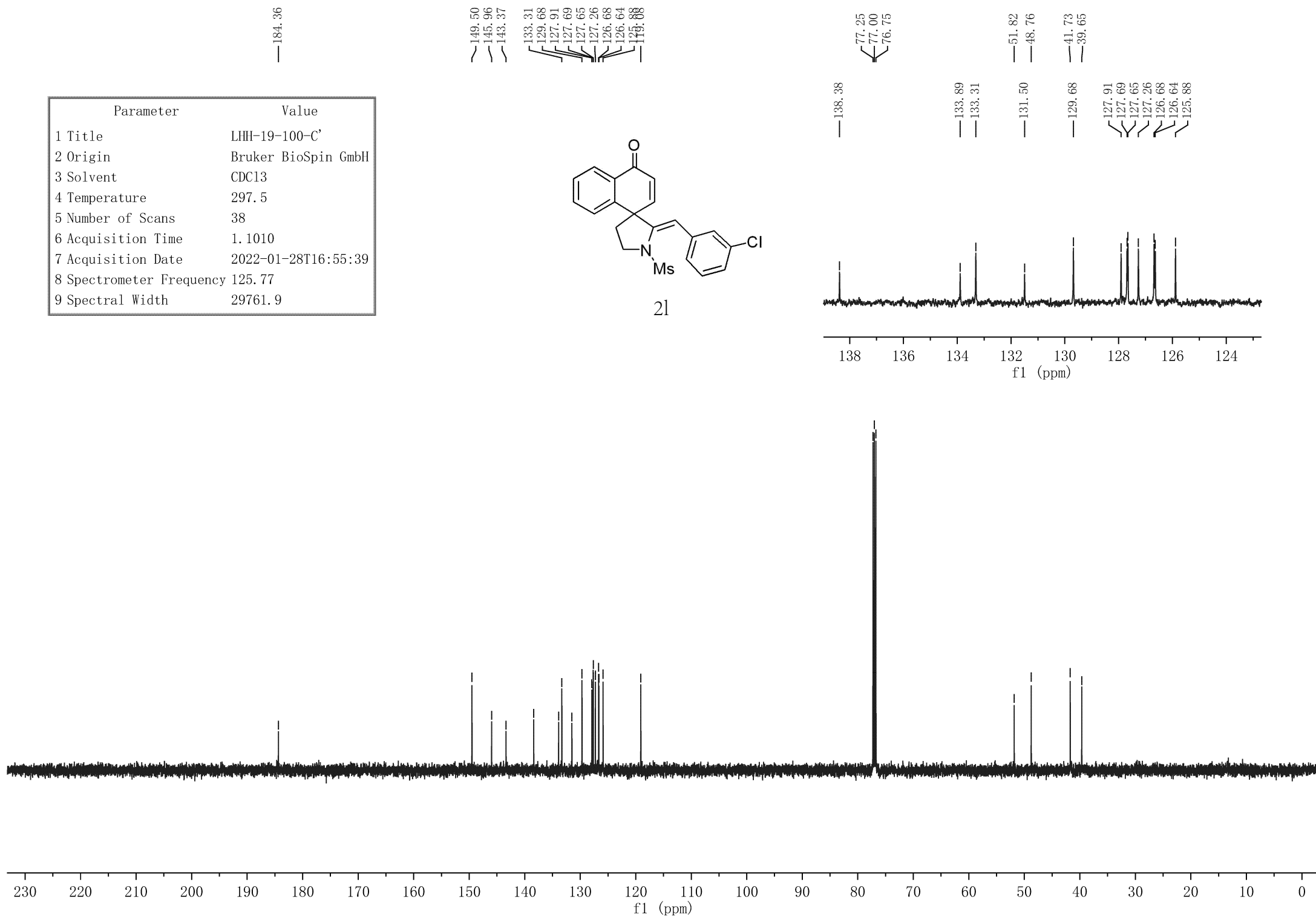
Parameter	Value
1 Title	LHH-12-159-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.6
5 Number of Scans	277
6 Acquisition Time	1.3631
7 Acquisition Date	2020-04-17T11:10:47
8 Spectrometer Frequency	100.59
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-100-C'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.5
5 Number of Scans	38
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-28T16:55:39
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



21



Parameter	Value
1 Title	ZYP-1-120-H'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.9
5 Number of Scans	23
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-20T15:19:08
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

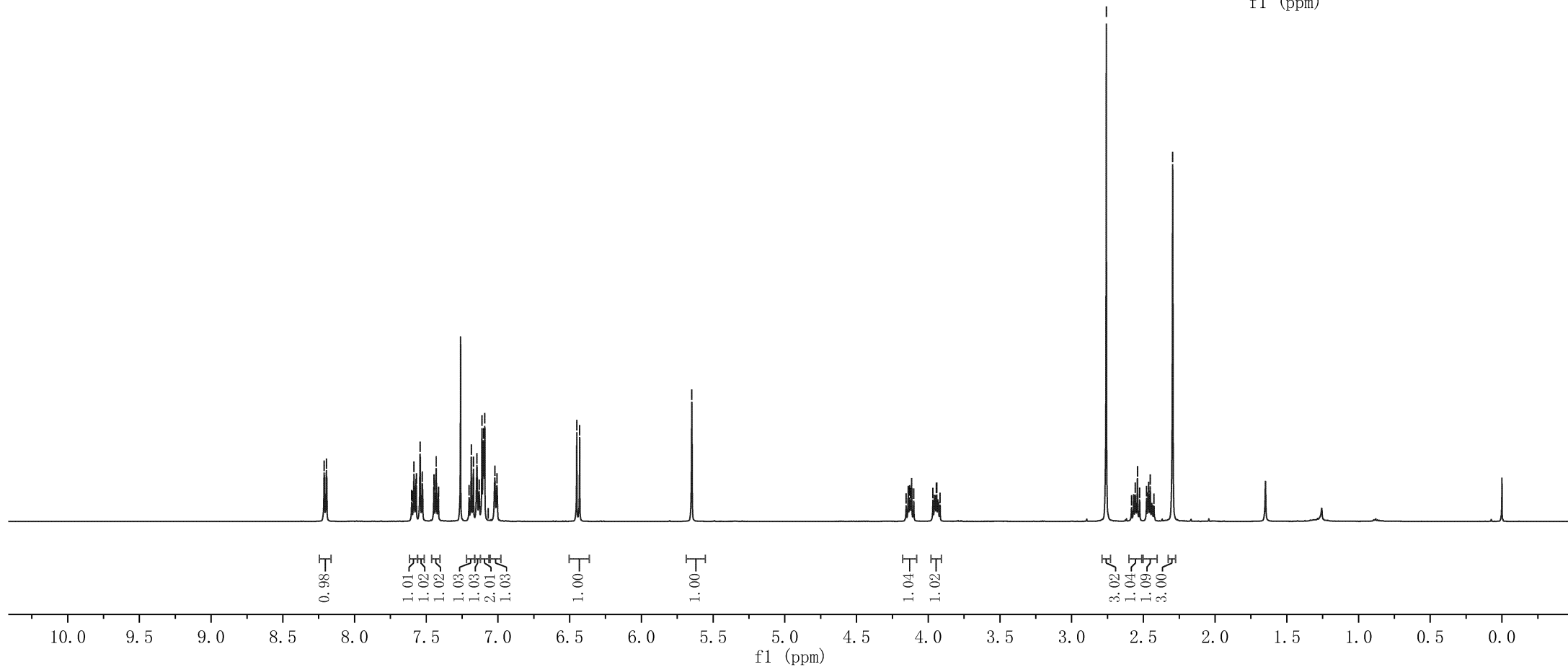
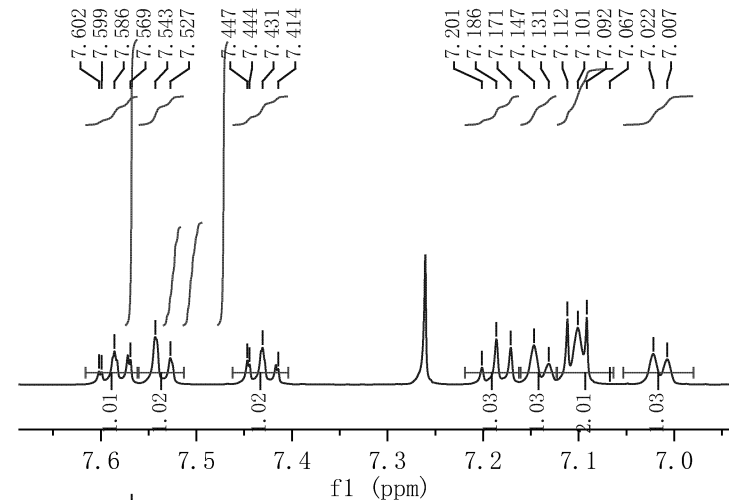
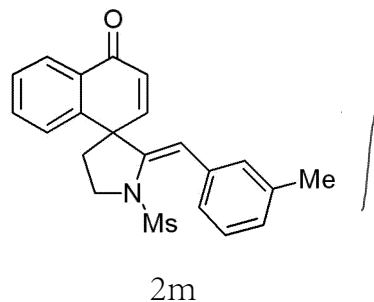
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8.196
7.586
7.569
7.543
7.527
7.447
7.431
7.186
7.171
7.147
7.112
7.101
7.092
7.022
6.987
6.966
6.430

5.648

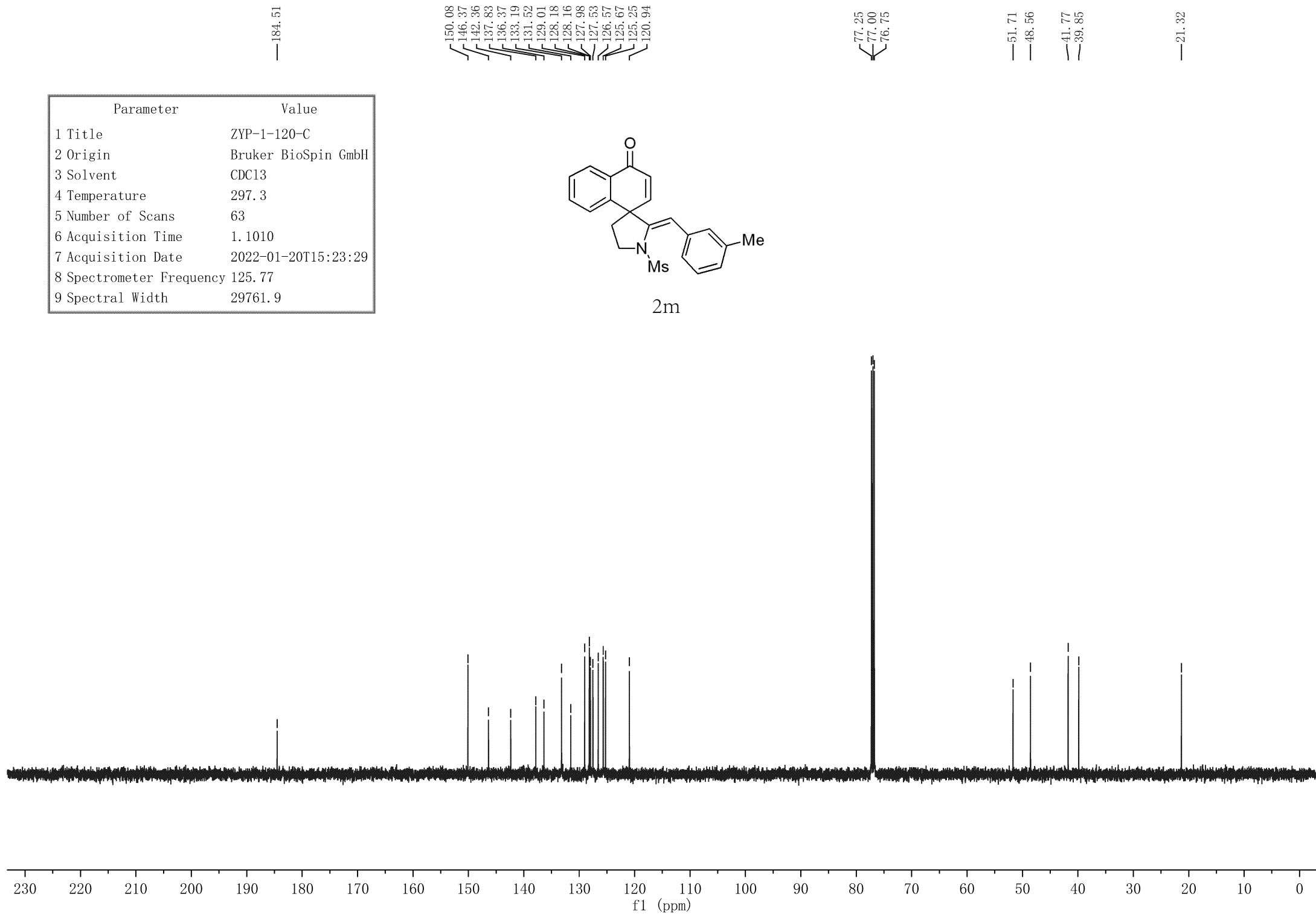
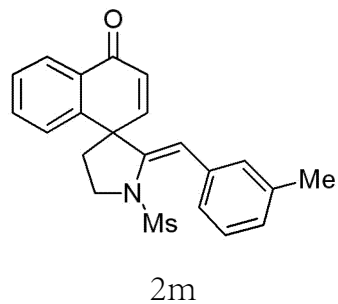
4.154
4.131
4.124
4.101
3.967
3.943
3.941
3.917

2.758
2.583
2.556
2.541
2.526
2.478
2.463
2.452
2.426
2.296

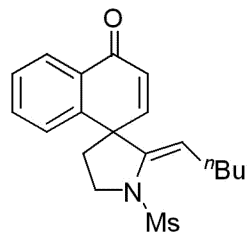
7.201
7.186
7.171
7.147
7.131
7.112
7.101
7.092
7.067
7.022
7.007



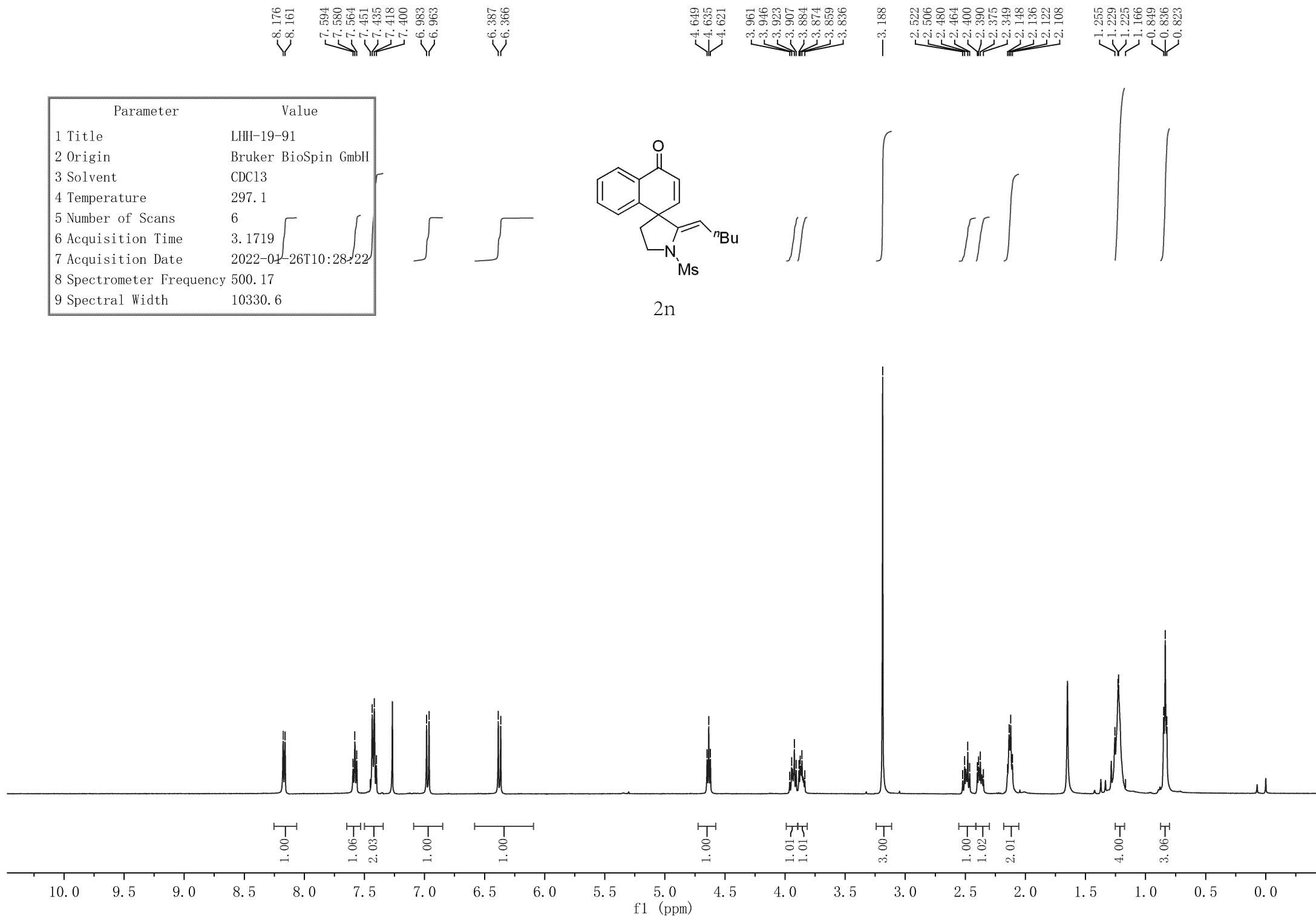
Parameter	Value
1 Title	ZYP-1-120-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	63
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-20T15:23:29
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



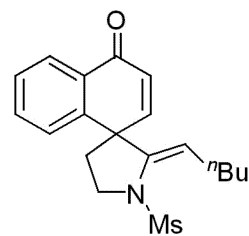
Parameter	Value
1 Title	LHH-19-91
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	6
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-26T10:28:22
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



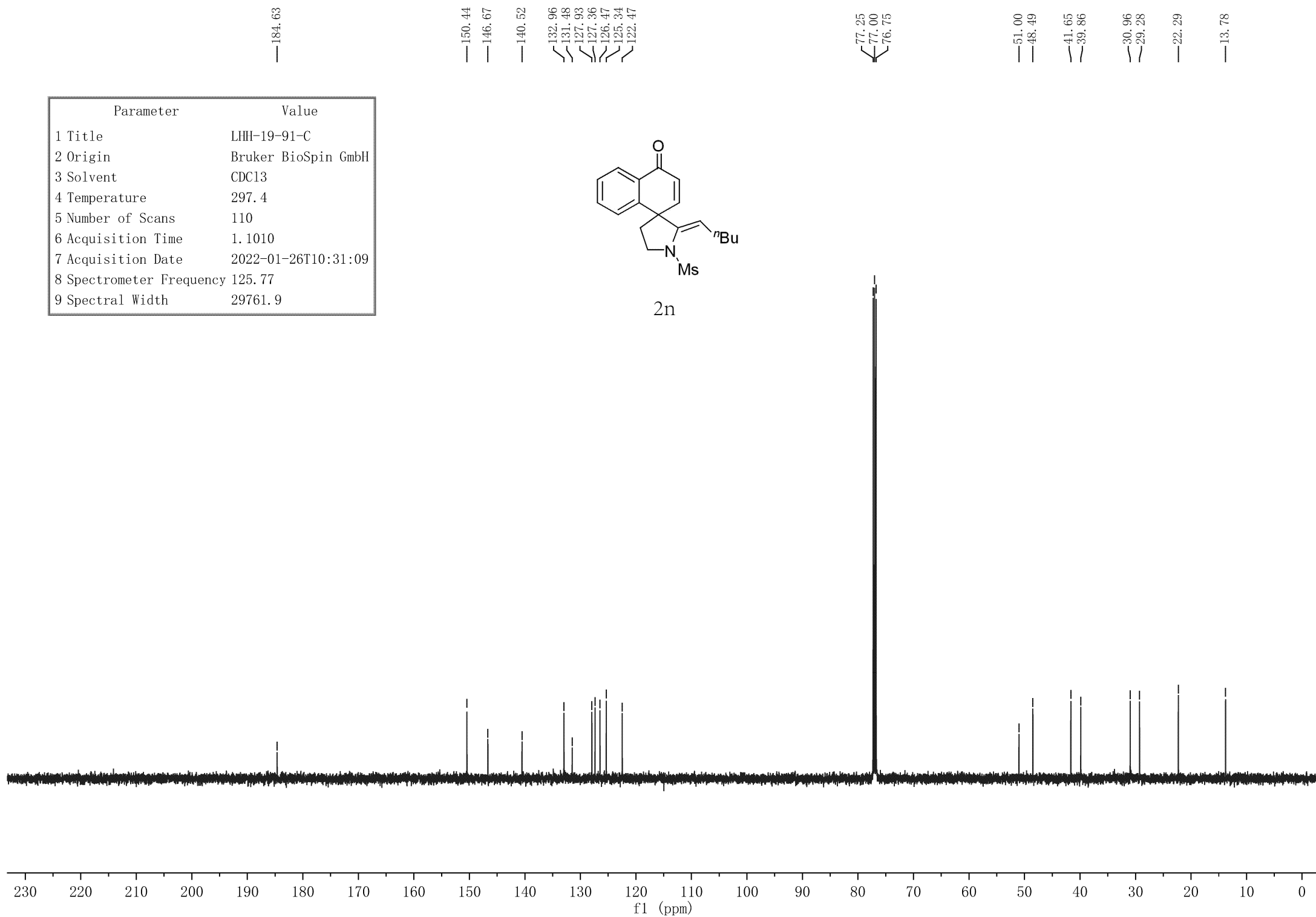
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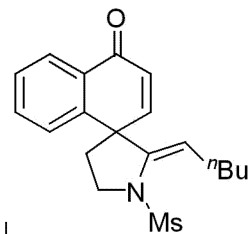
Parameter	Value
1 Title	LHH-19-91-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.4
5 Number of Scans	110
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-26T10:31:09
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



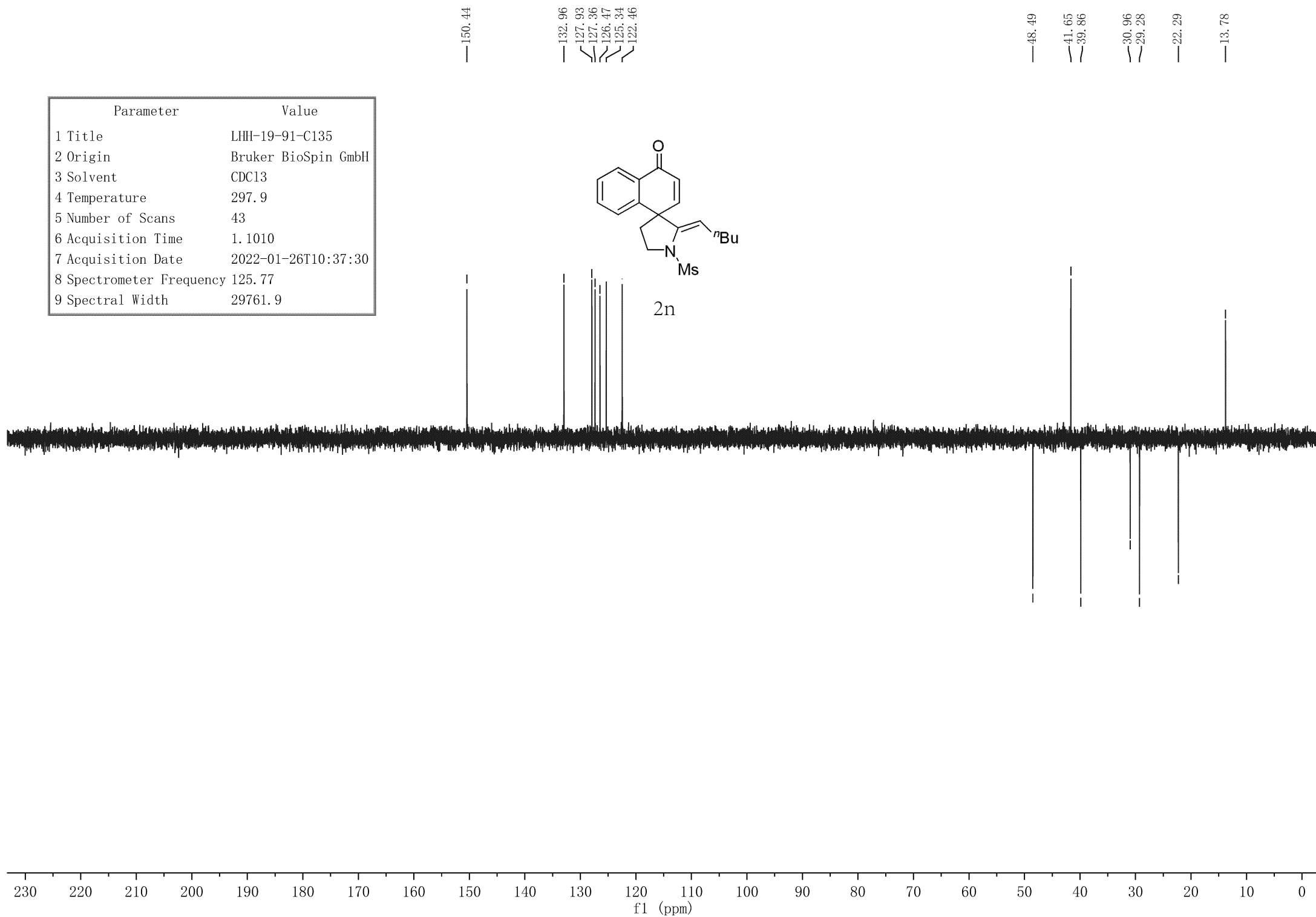
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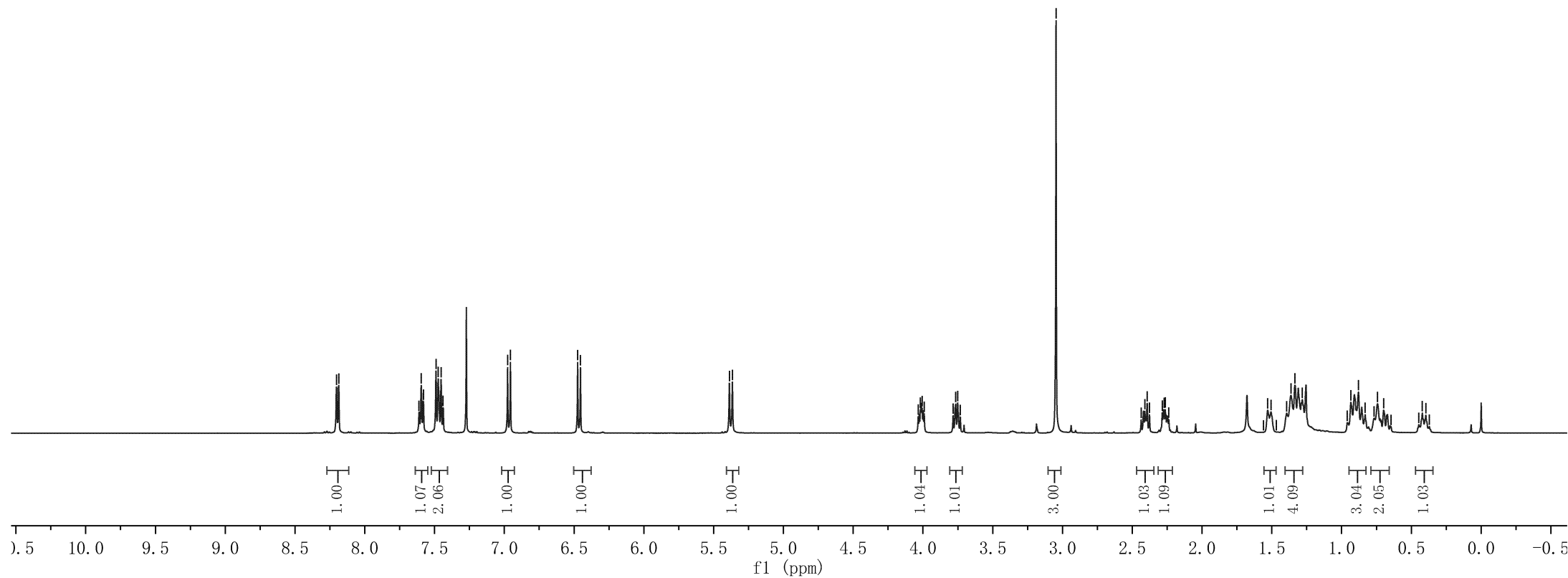
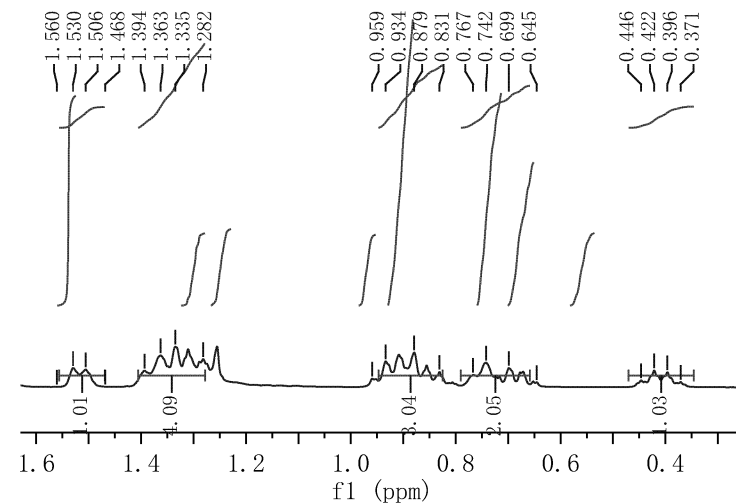
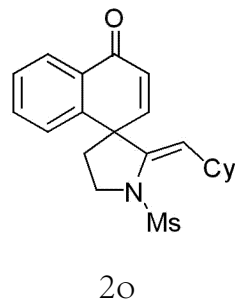
Parameter	Value
1 Title	LHH-19-91-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.9
5 Number of Scans	43
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-26T10:37:30
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



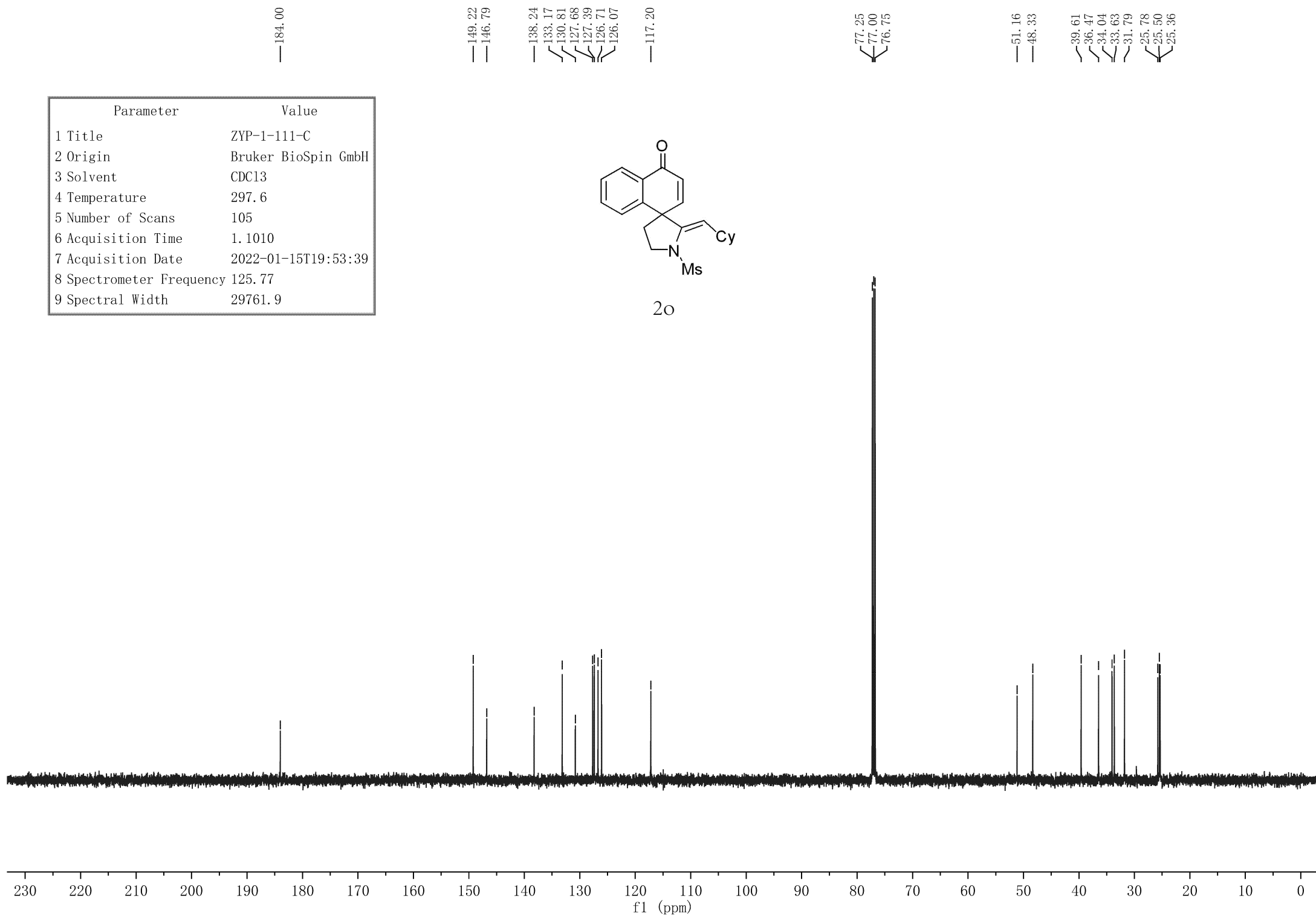
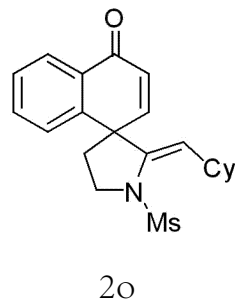
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Parameter	Value
1 Title	ZYP-1-111-H'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.2
5 Number of Scans	34
6 Acquisition Time	3.1719
7 Acquisition Date	2022-04-15T19:48:49
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



Parameter	Value
1 Title	ZYP-1-111-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.6
5 Number of Scans	105
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-15T19:53:39
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



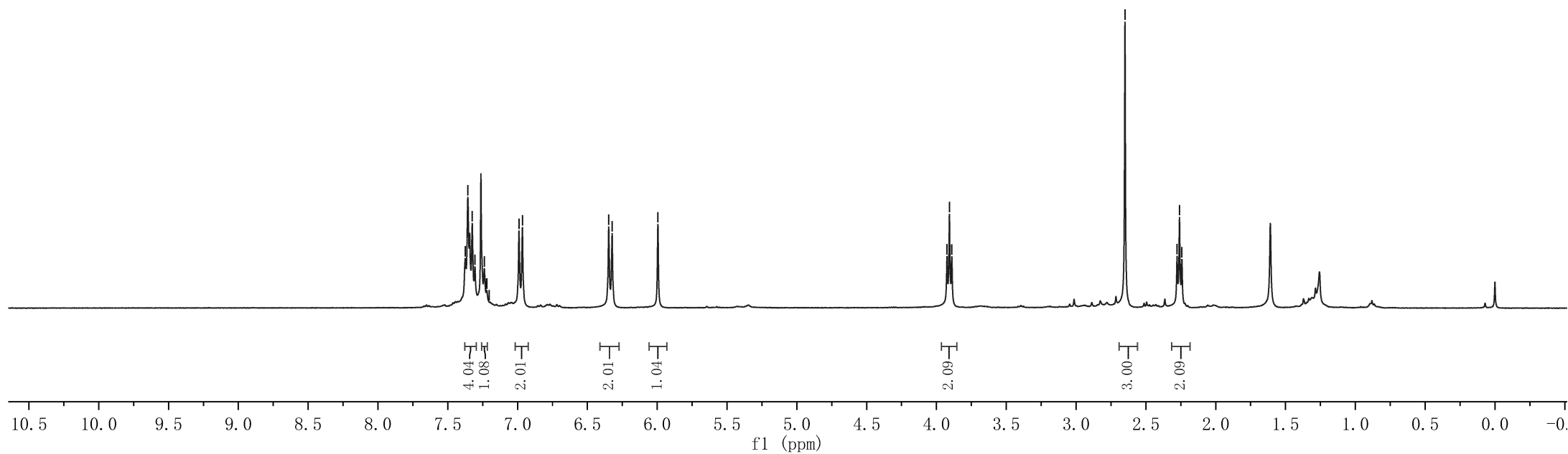
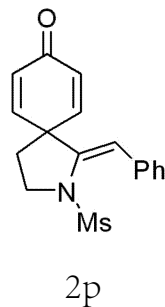
Parameter	Value
1 Title	LHH-19-79
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDCl3
4 Temperature	298.0
5 Number of Scans	6
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-20T00:39:03
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

7.375
7.356
7.325
7.306
7.238
7.222
7.221
7.203
6.990
6.965
6.348
6.323
5.995

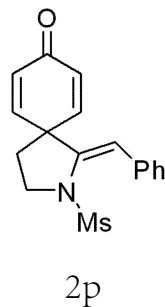
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3.907
3.890

2.650

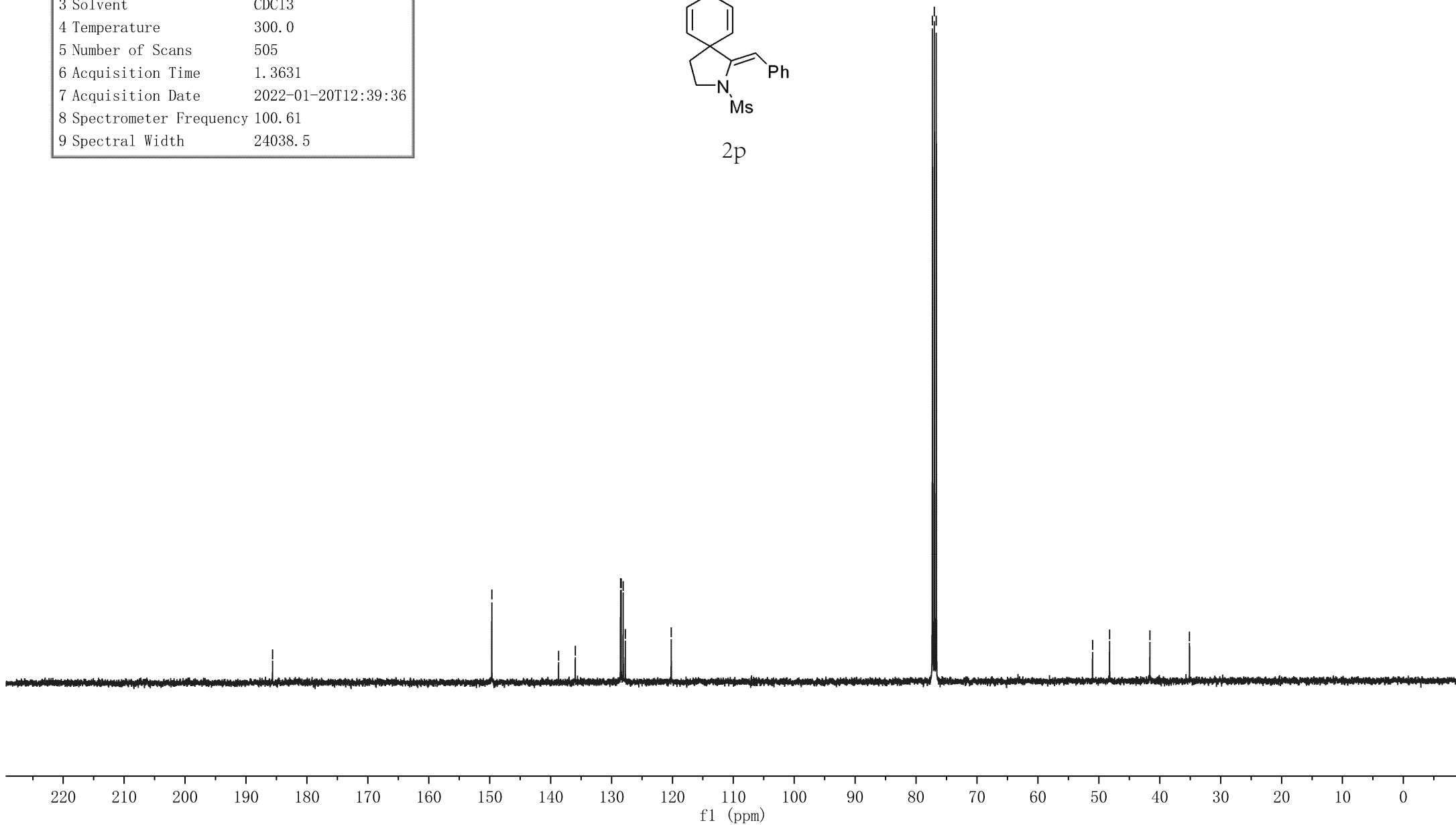
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2.259
2.242



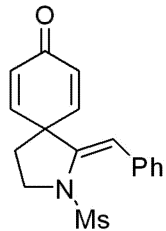
Parameter	Value
1 Title	LHH-19-79-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	505
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-20T12:39:36
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



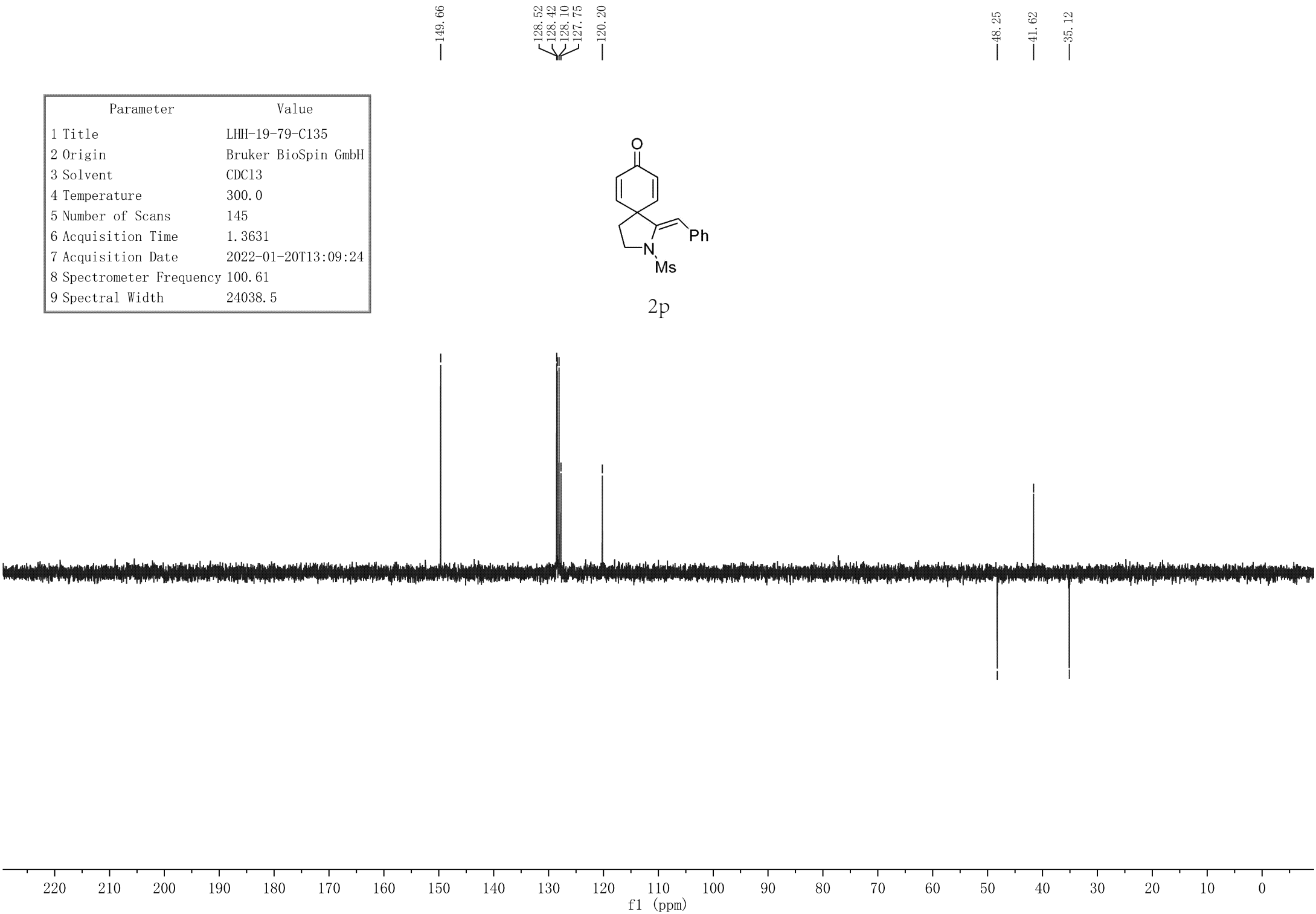
185.64
149.65
138.70
135.96
128.52
128.41
128.09
127.74
120.19
77.32
77.00
76.68
51.03
48.24
41.62
35.12



Parameter	Value
1 Title	LHH-19-79-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	145
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-20T13:09:24
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



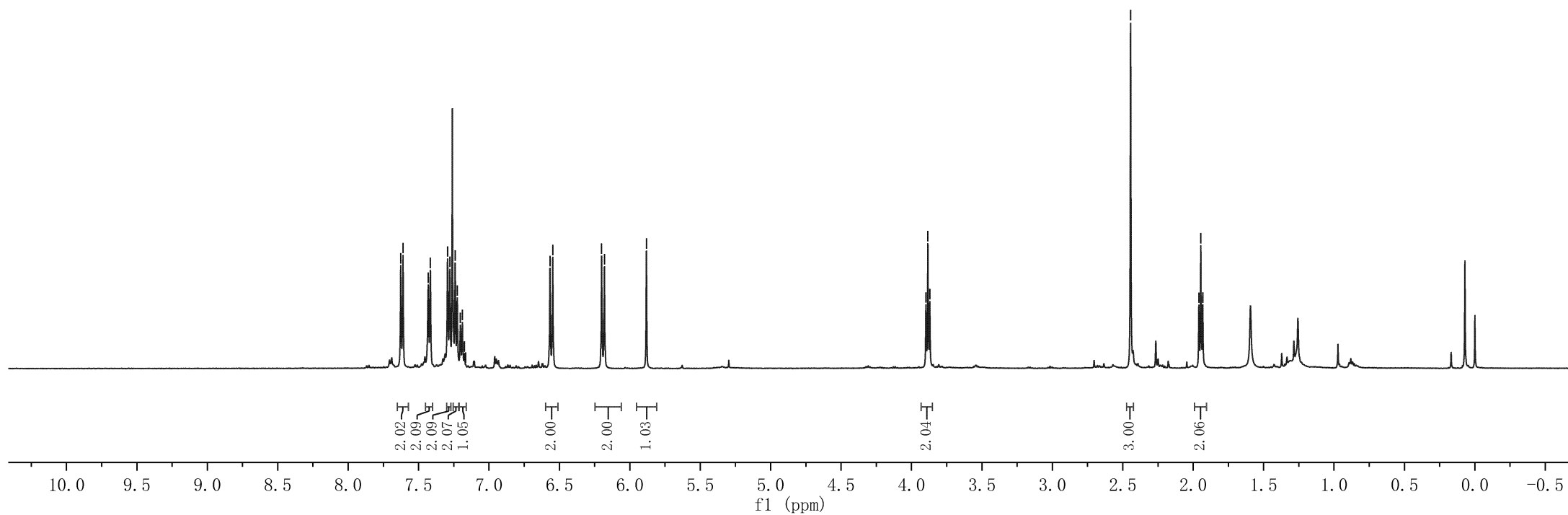
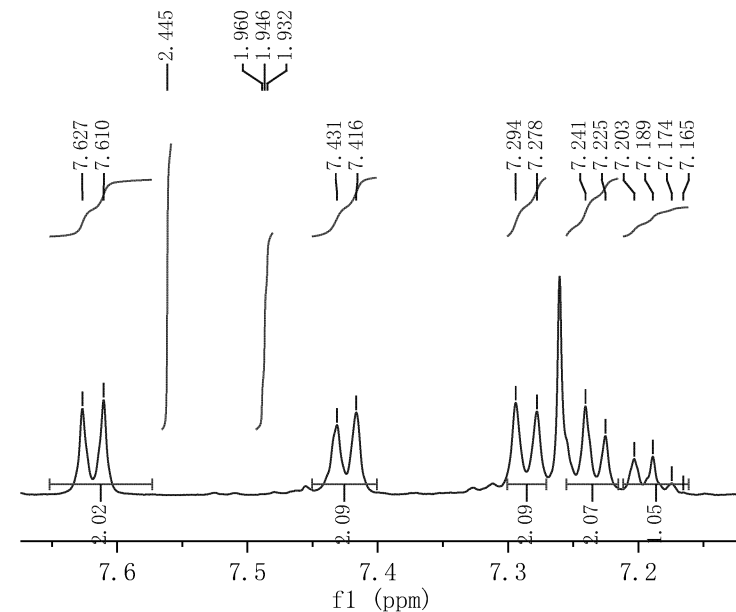
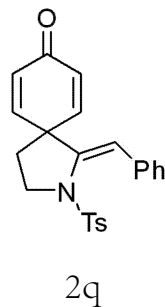
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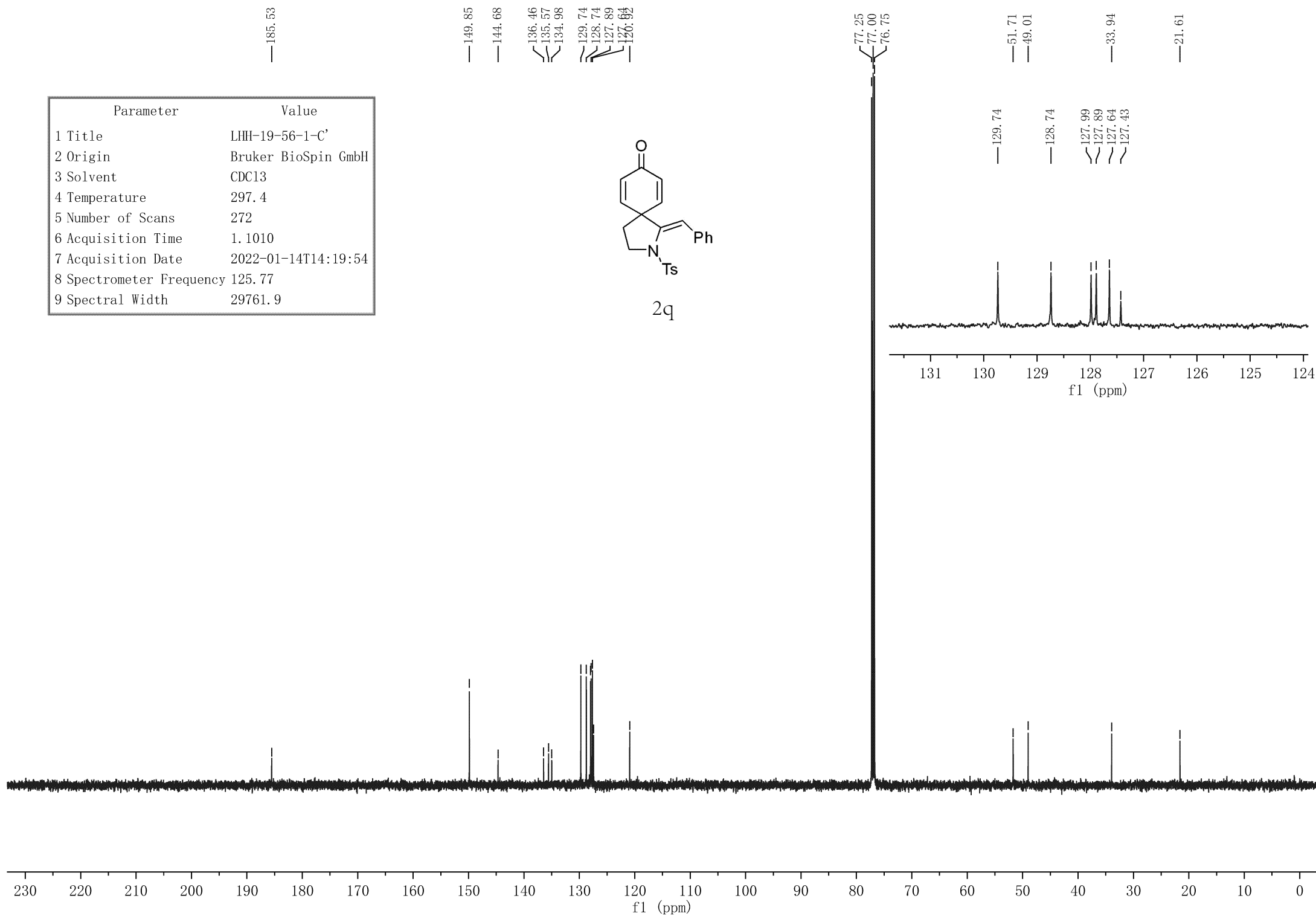
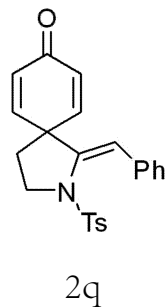
Parameter	Value
1 Title	LHH-19-56-1
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.0
5 Number of Scans	5
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-14T10:40:42
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

7.627
7.610
7.431
7.416
7.294
7.278
7.241
7.225
7.203
7.189
7.174
7.165
6.567
6.547
6.200
6.180
5.882

3.898
3.884
3.870

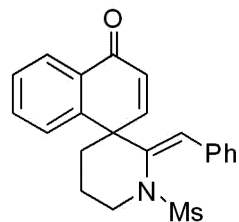


Parameter	Value
1 Title	LHH-19-56-1-C'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.4
5 Number of Scans	272
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-14T14:19:54
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



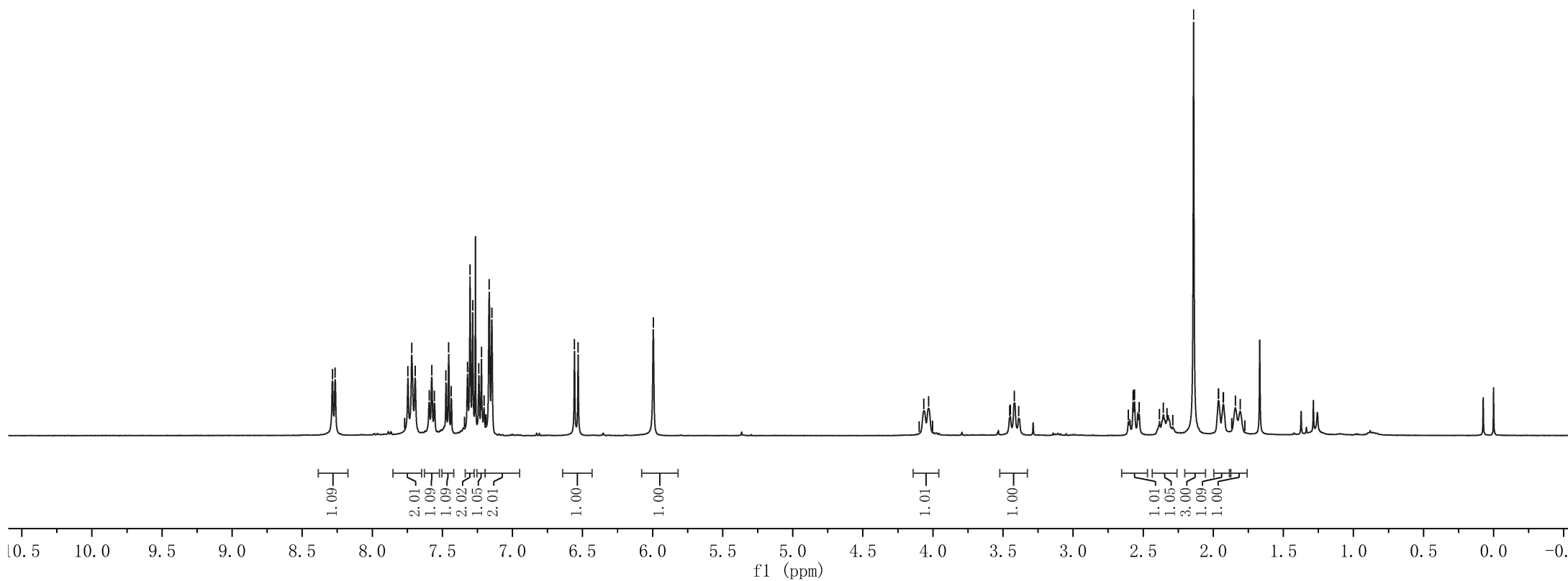
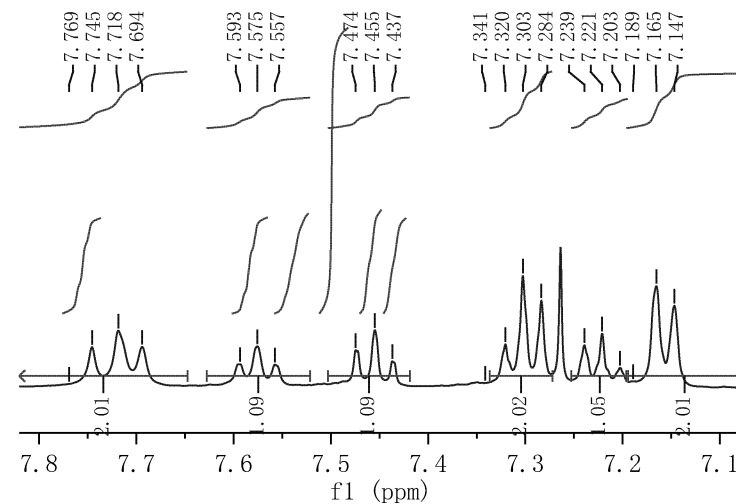
Parameter	Value
1 Title	LHH-19-132
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-02-19T10:00:38
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

8.284
8.265
7.745
7.718
7.694
7.575
7.474
7.455
7.320
7.303
7.284
7.239
7.221
7.165
7.147
6.531
5.995

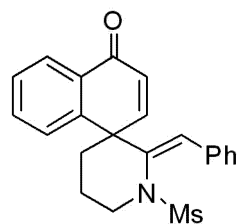


2r

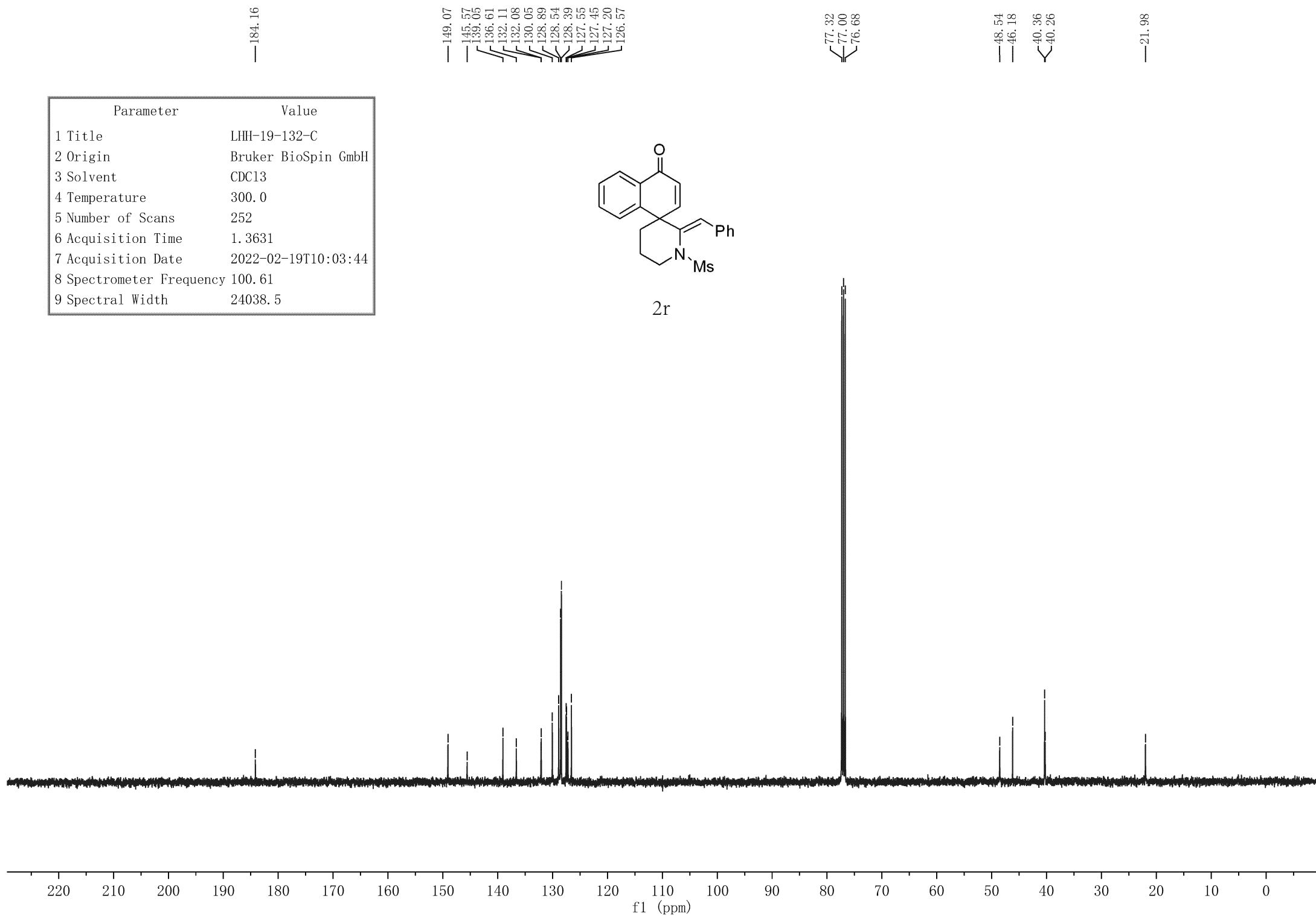
4.098
4.065
4.031
4.002
3.453
3.448
3.419
3.389
2.606
2.572
2.562
2.528
2.384
2.356
2.330
2.289
2.140
1.963
1.962
1.929
1.928
1.868
1.842
1.808
1.774



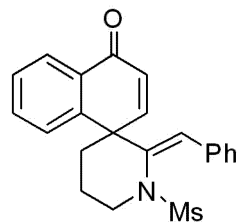
Parameter	Value
1 Title	LHH-19-132-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	252
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-19T10:03:44
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



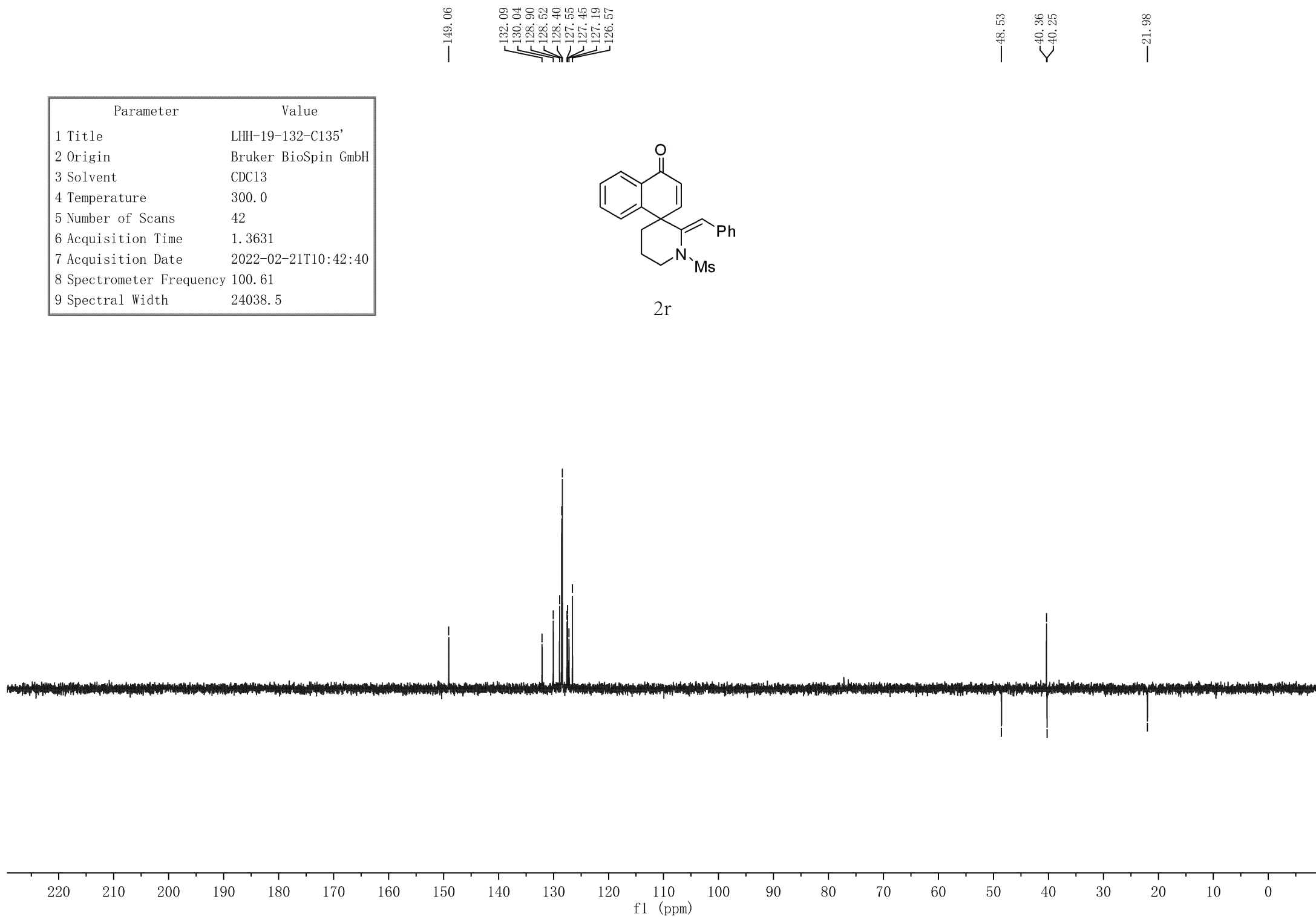
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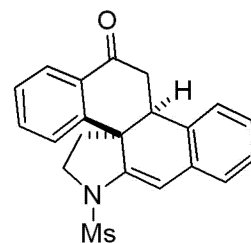
Parameter	Value
1 Title	LHH-19-132-C135'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	42
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-21T10:42:40
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



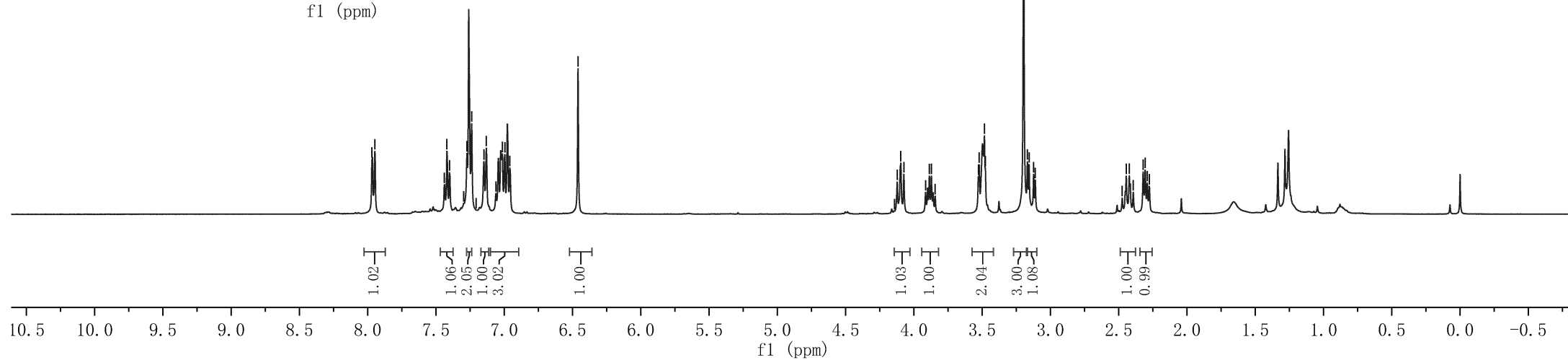
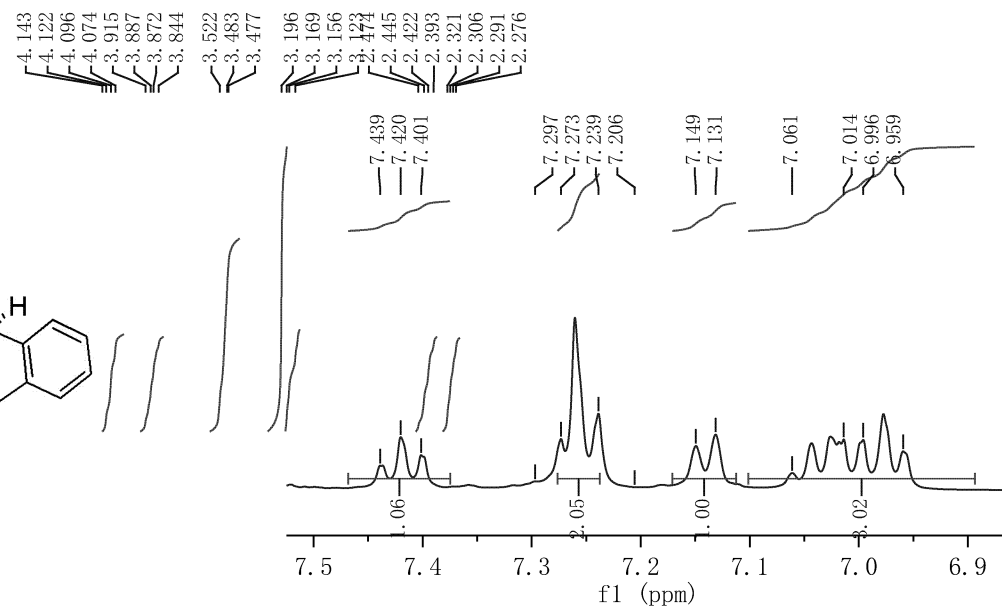
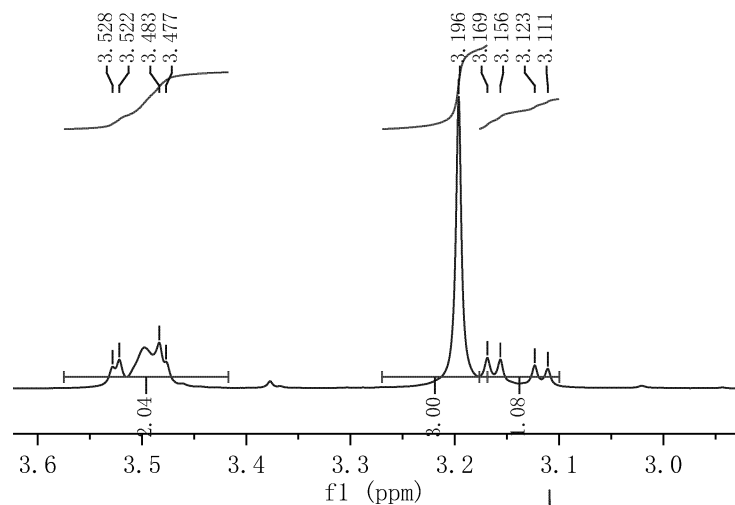
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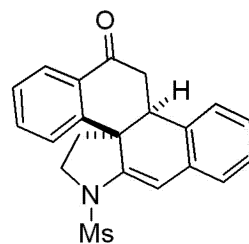
Parameter	Value
1 Title	zyp-1-80-h'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	16
6 Acquisition Time	4.0894
7 Acquisition Date	2021-12-20T17:01:30
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



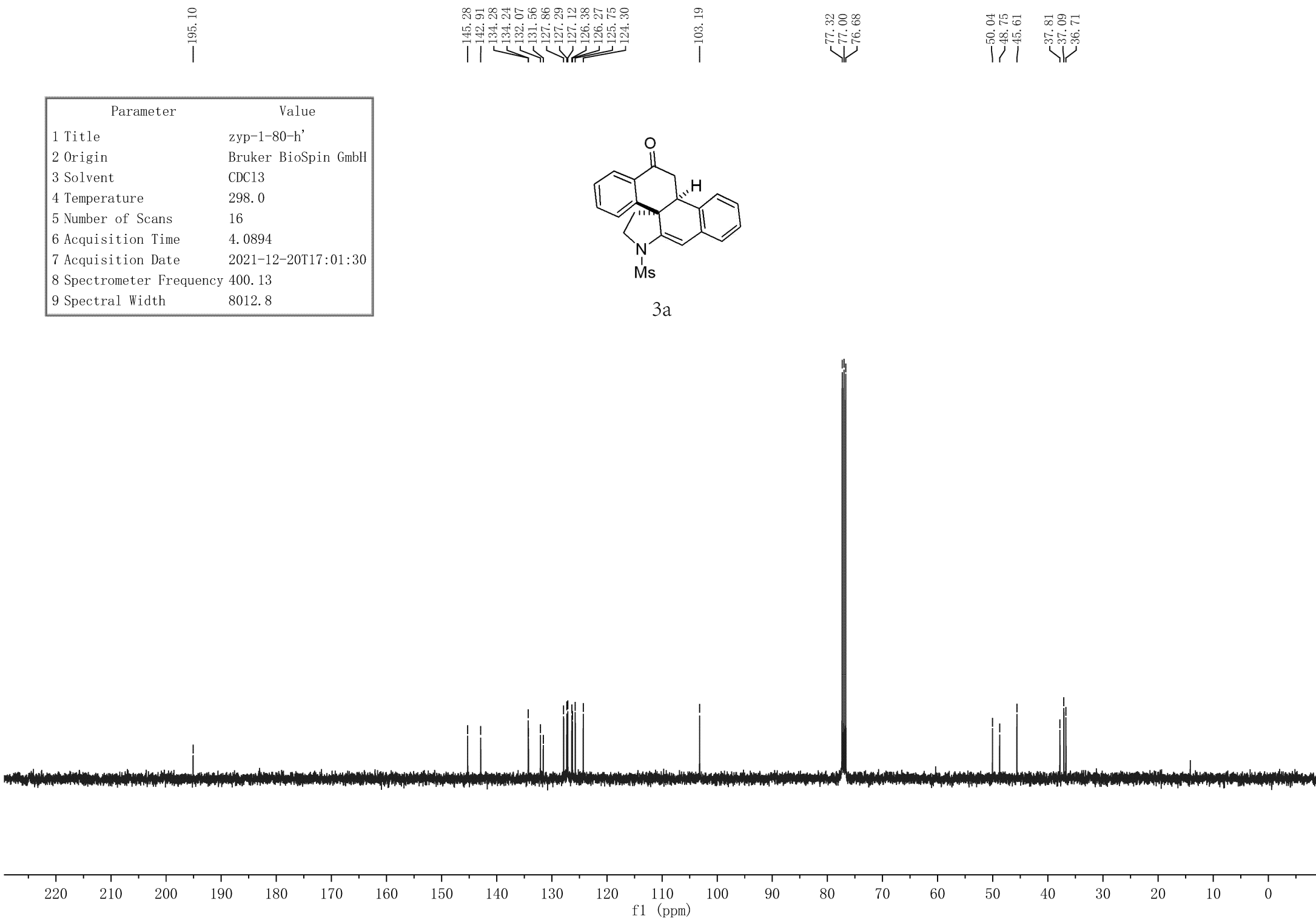
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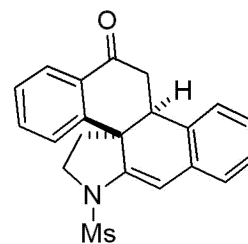
Parameter	Value
1 Title	zyp-1-80-h'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	16
6 Acquisition Time	4.0894
7 Acquisition Date	2021-12-20T17:01:30
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



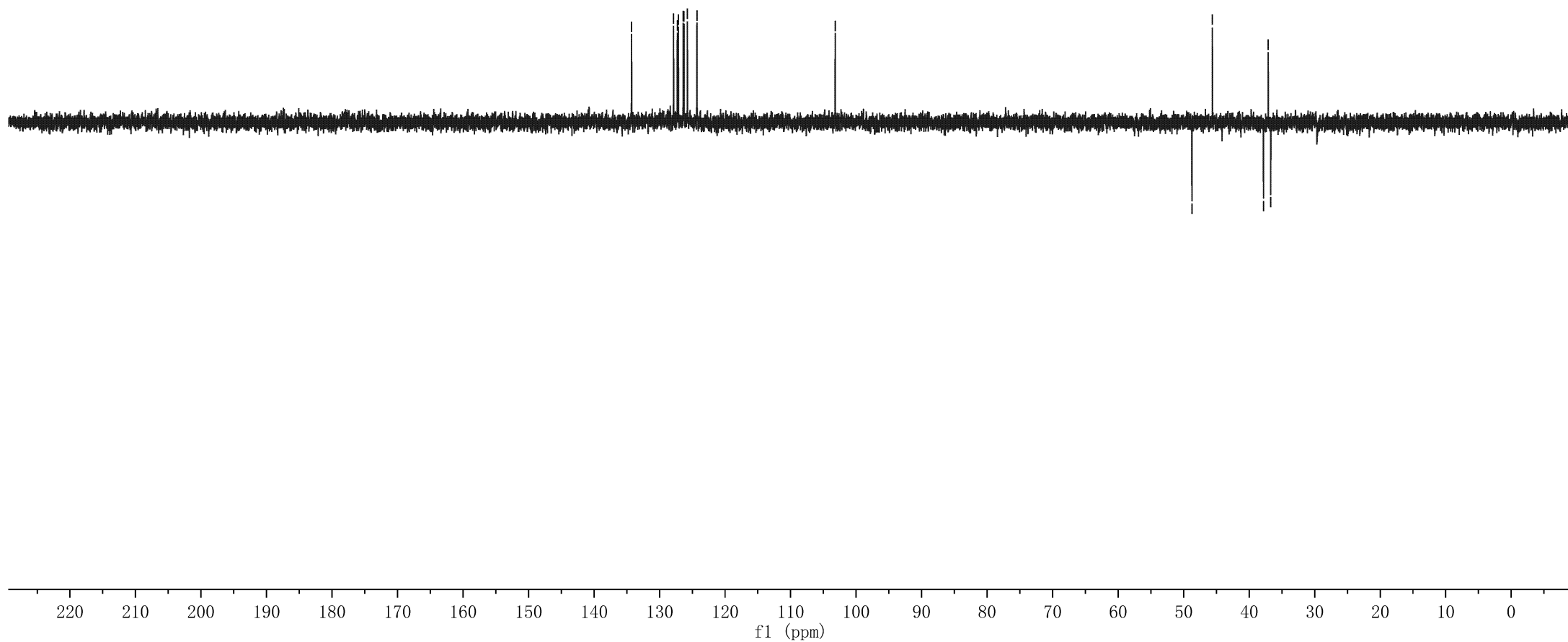
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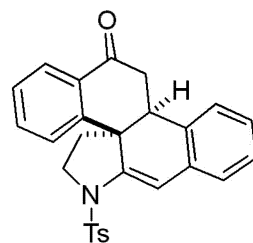
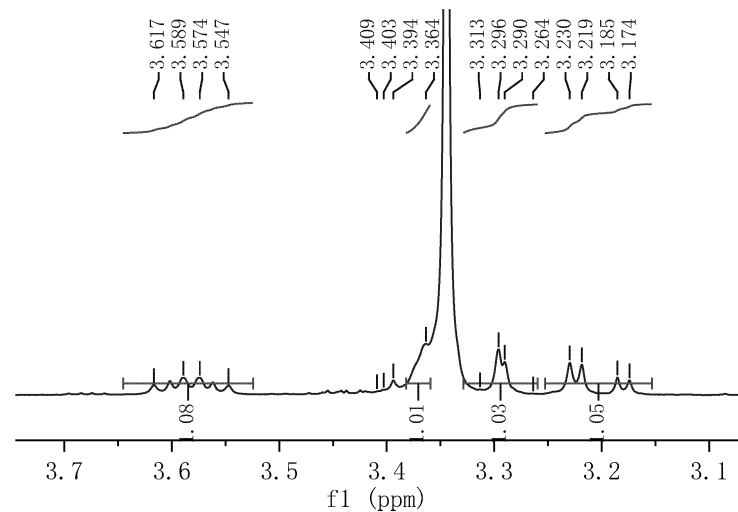
Parameter	Value
1 Title	LHH-19-C135''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	74
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-04T18:38:39
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



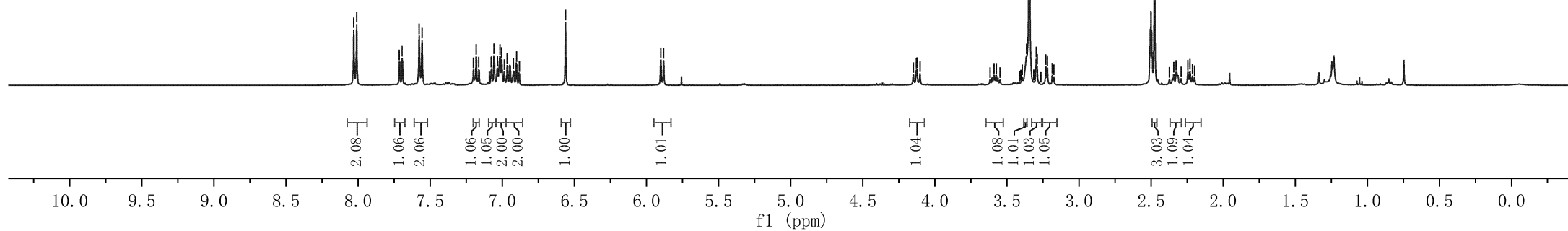
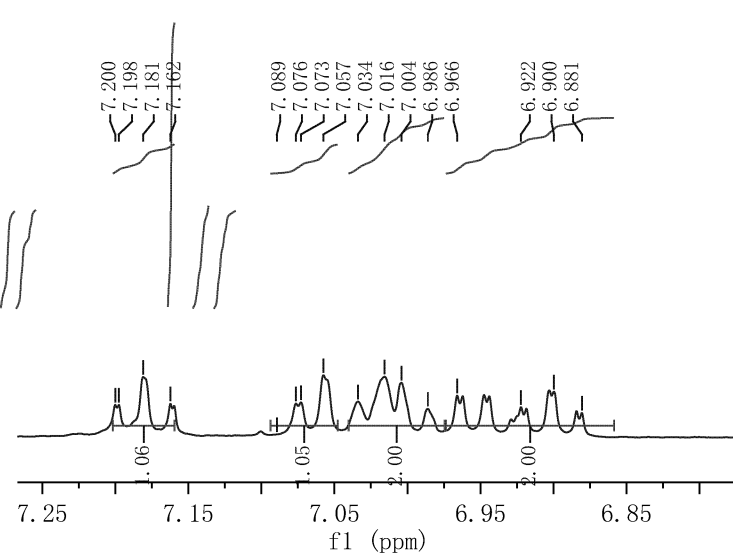
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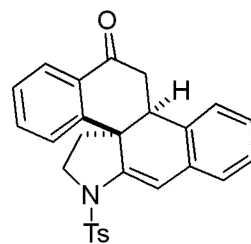
Parameter	Value
1 Title	LHH-19-65-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	295.4
5 Number of Scans	9
6 Acquisition Time	3.9846
7 Acquisition Date	2022-01-19T10:38:43
8 Spectrometer Frequency	399.93
9 Spectral Width	8223.7



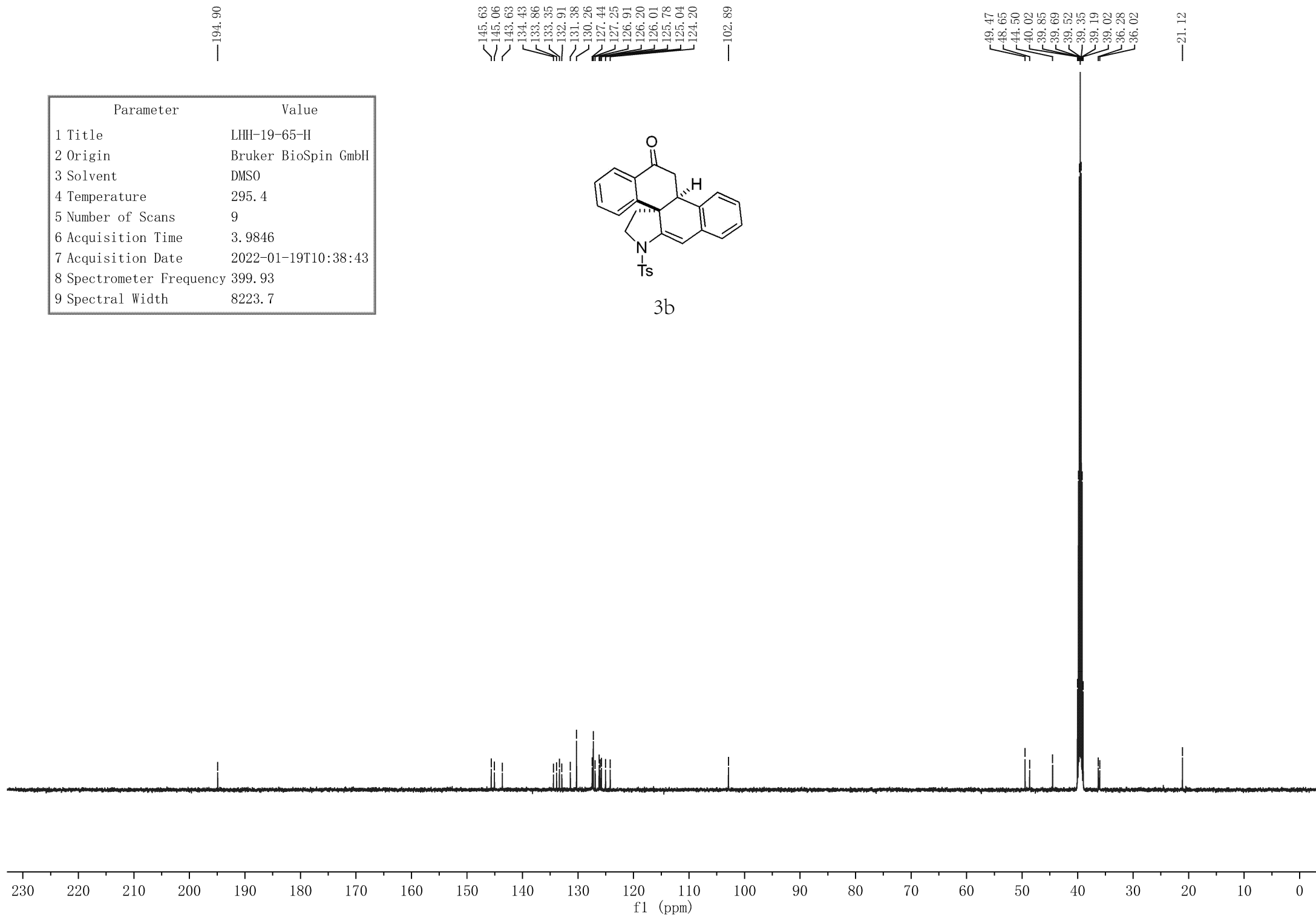
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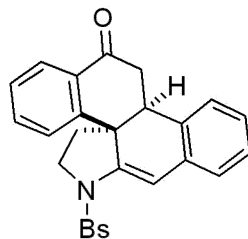
Parameter	Value
1 Title	LHH-19-65-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	295.4
5 Number of Scans	9
6 Acquisition Time	3.9846
7 Acquisition Date	2022-01-19T10:38:43
8 Spectrometer Frequency	399.93
9 Spectral Width	8223.7



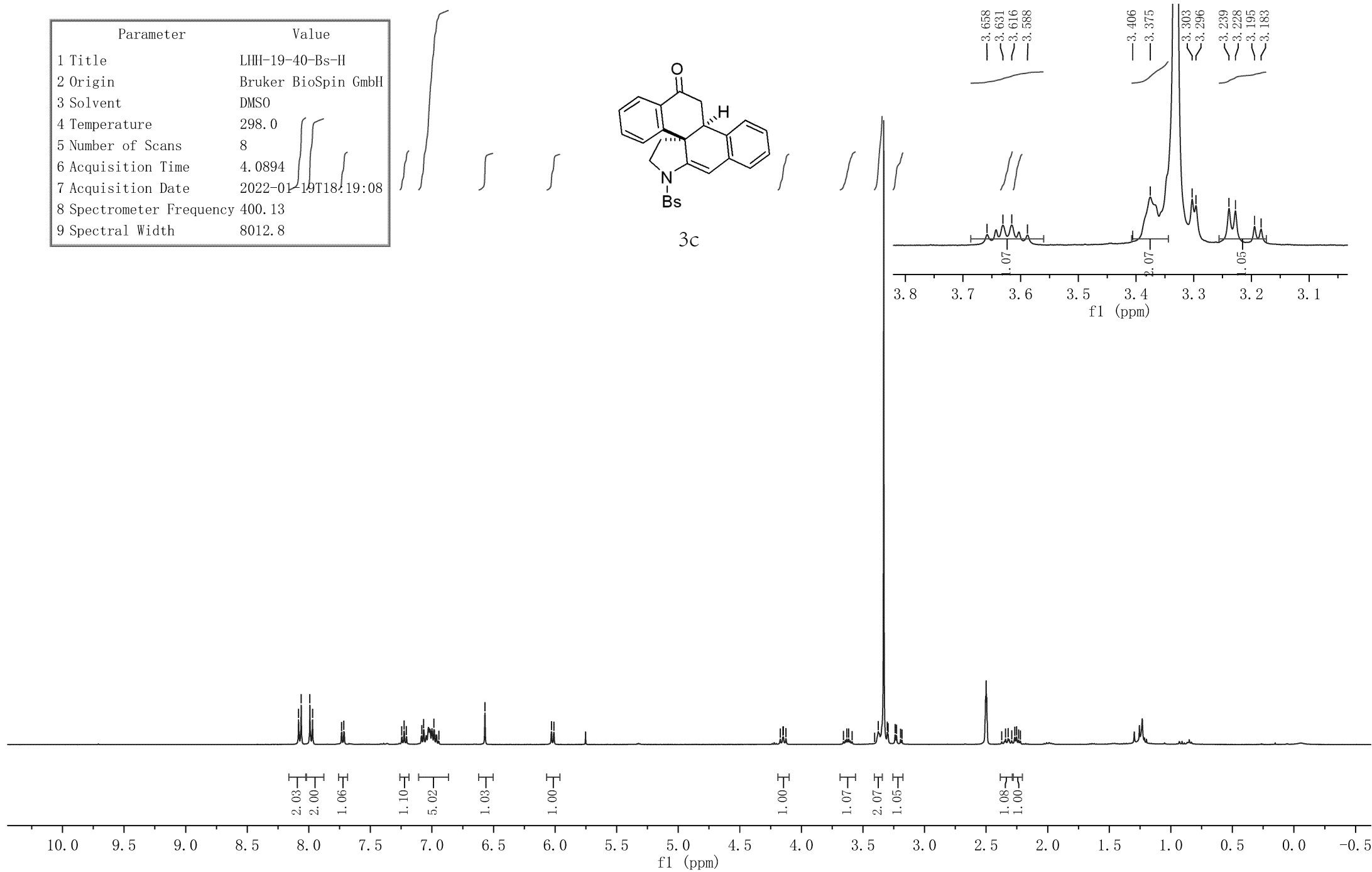
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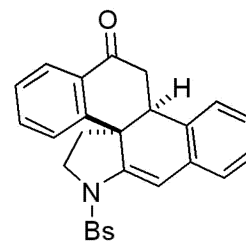
Parameter	Value
1 Title	LHH-19-40-Bs-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	298.0
5 Number of Scans	8
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-19T18:19:08
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



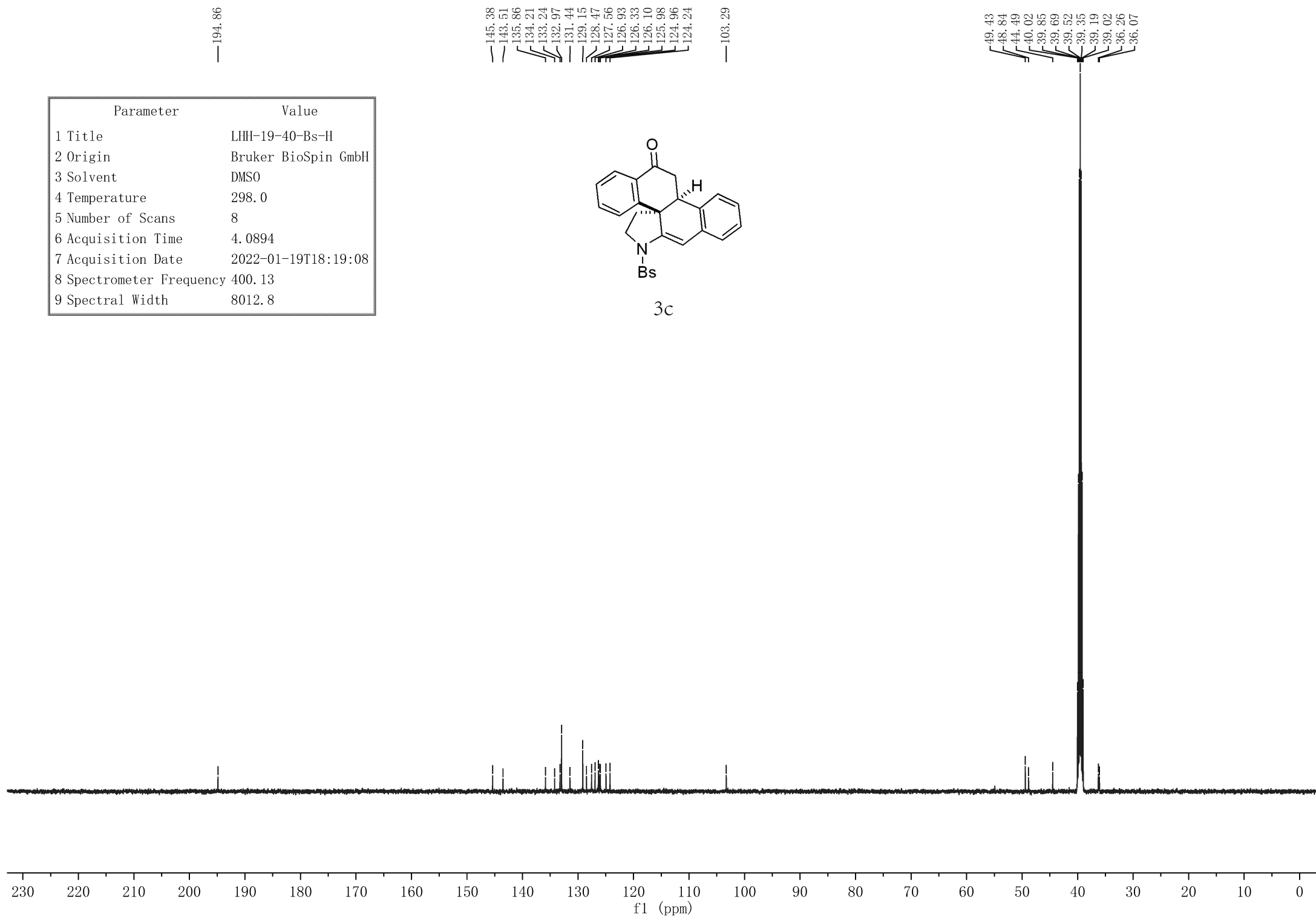
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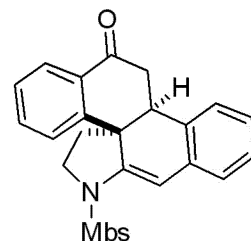
Parameter	Value
1 Title	LHH-19-40-Bs-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	298.0
5 Number of Scans	8
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-19T18:19:08
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



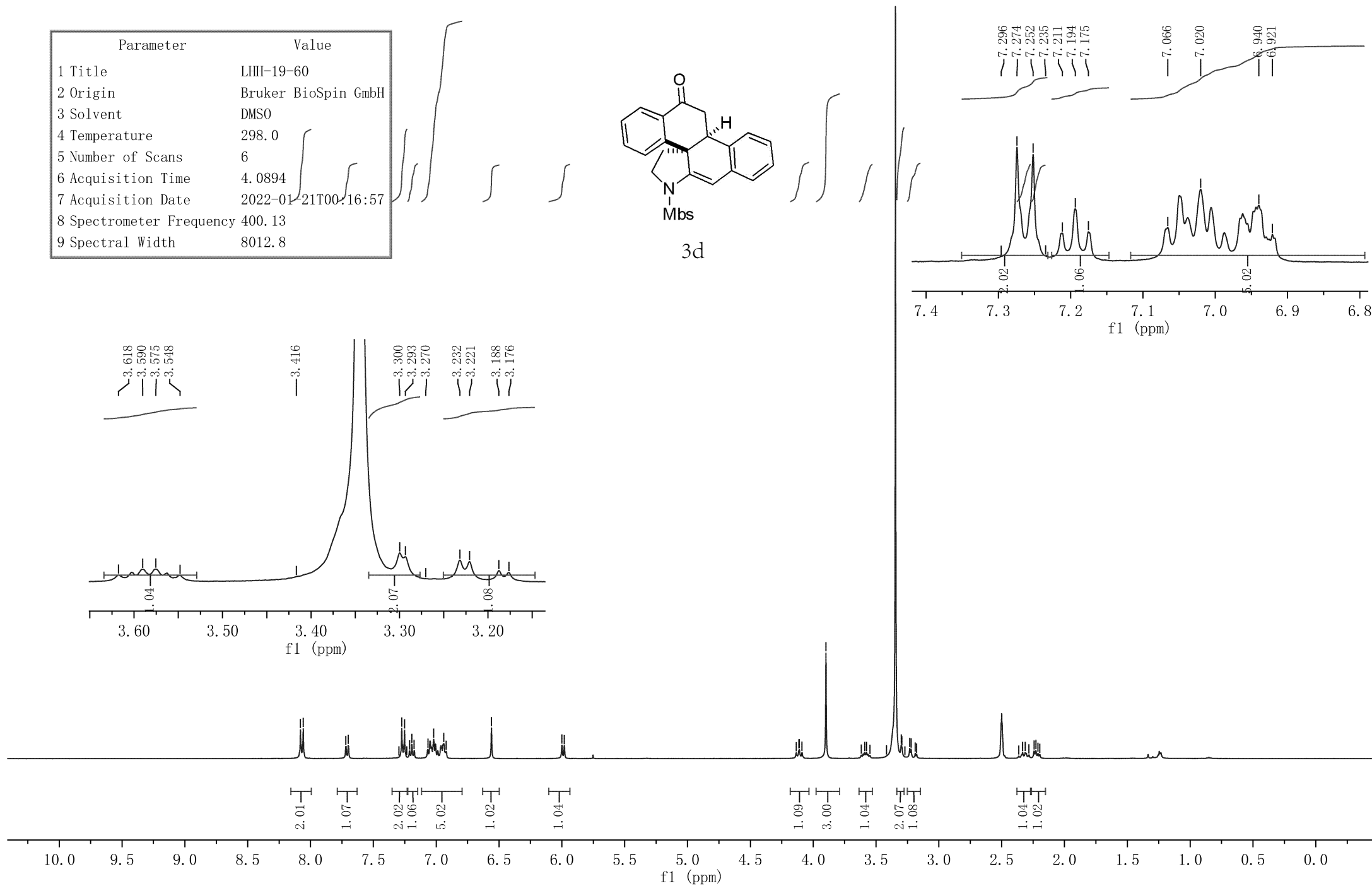
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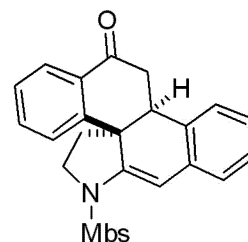
Parameter	Value
1 Title	LHH-19-60
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	298.0
5 Number of Scans	6
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-21T00:16:57
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



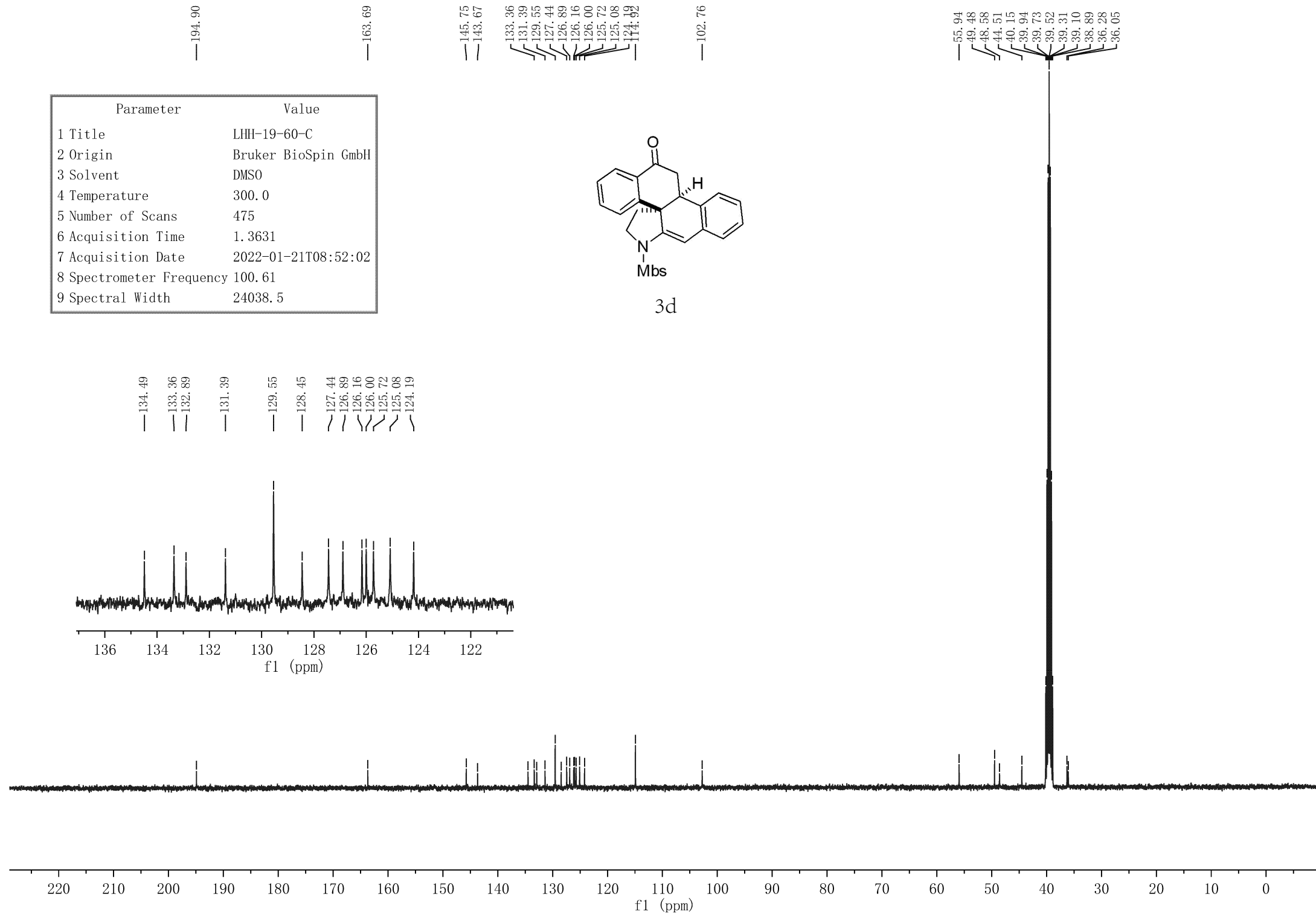
3d



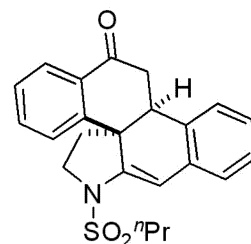
Parameter	Value
1 Title	LHH-19-60-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	DMSO
4 Temperature	300.0
5 Number of Scans	475
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-21T08:52:02
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



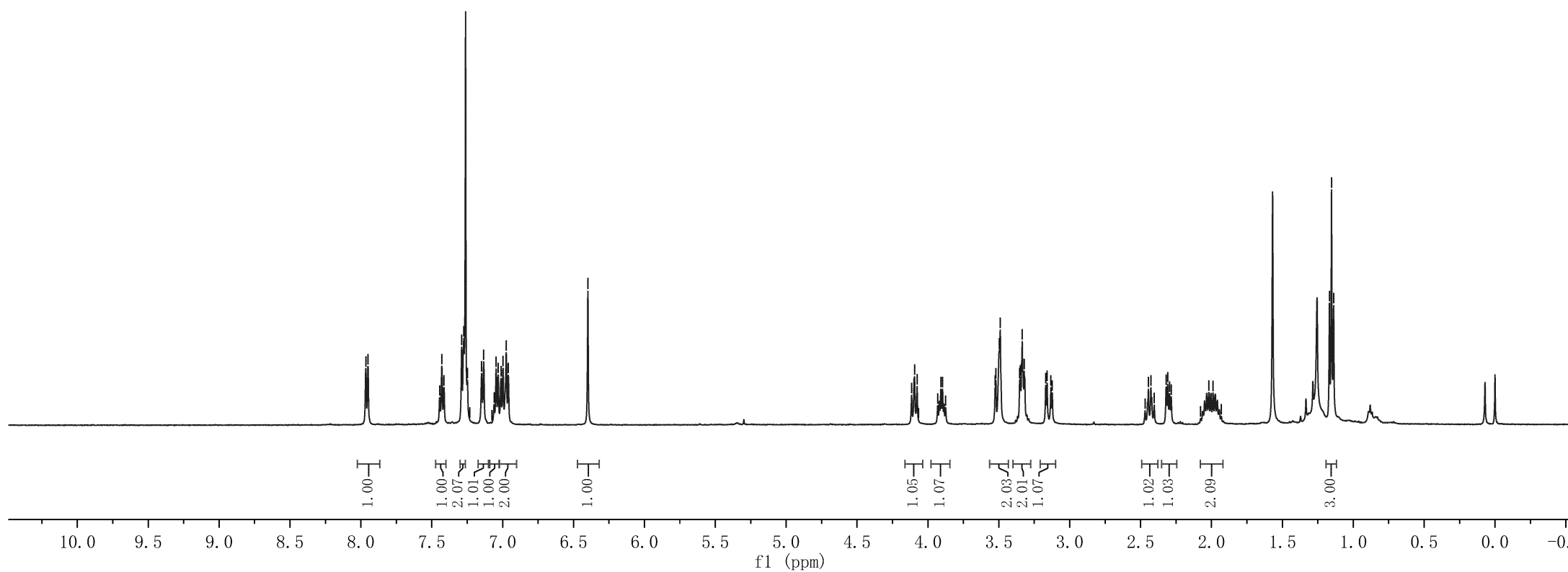
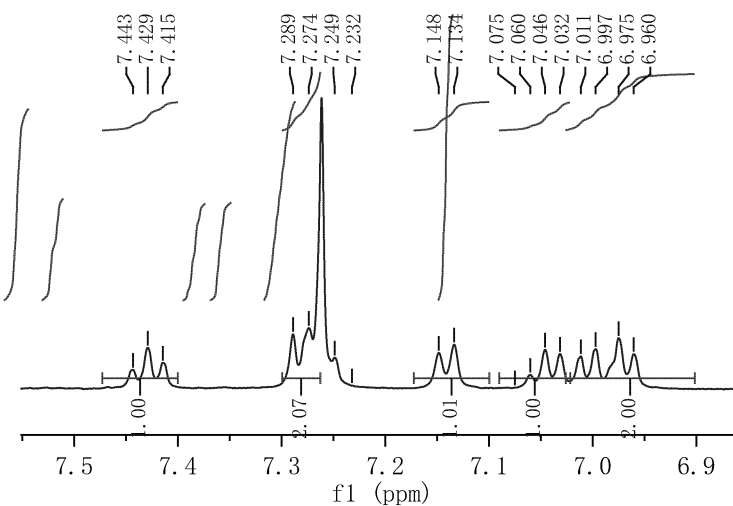
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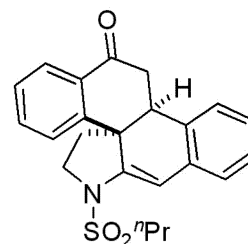
Parameter	Value
1 Title	LHH-19-81
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.1
5 Number of Scans	7
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-21 15:50:03
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



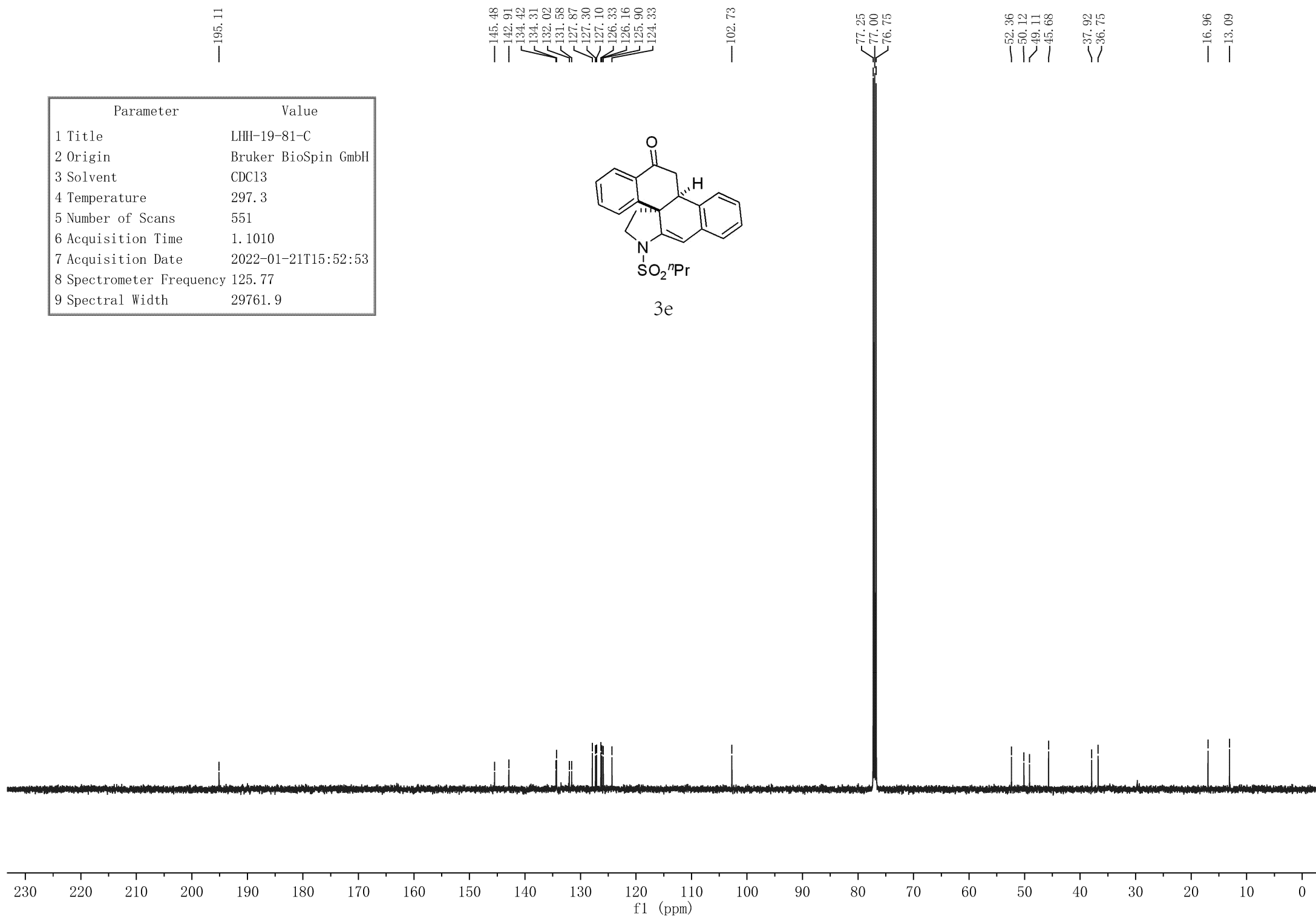
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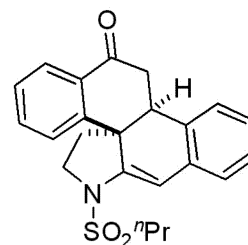
Parameter	Value
1 Title	LHH-19-81-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	551
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-21T15:52:53
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



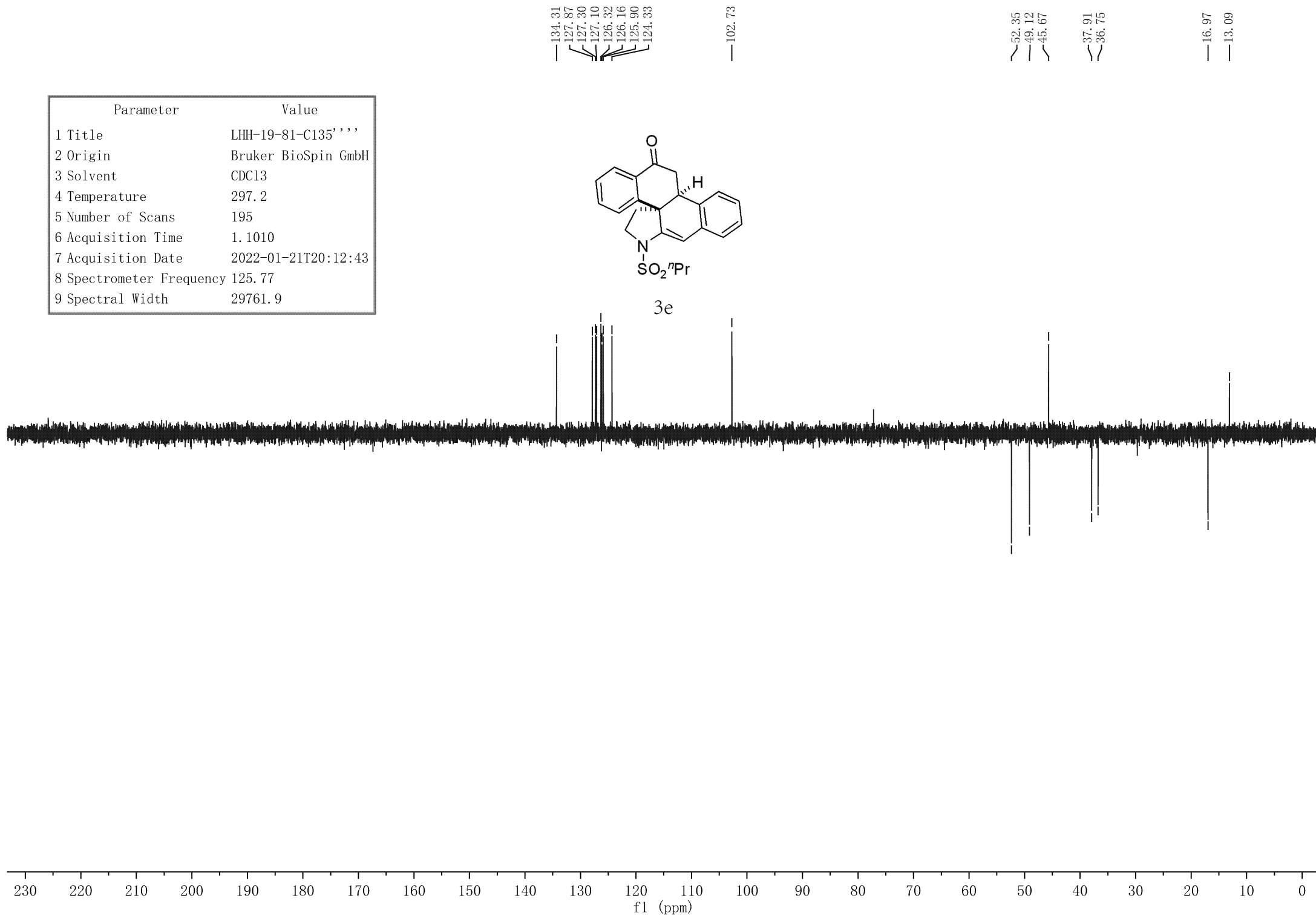
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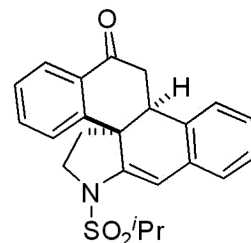
Parameter	Value
1 Title	LHH-19-81-C135''''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.2
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6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-21T20:12:43
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



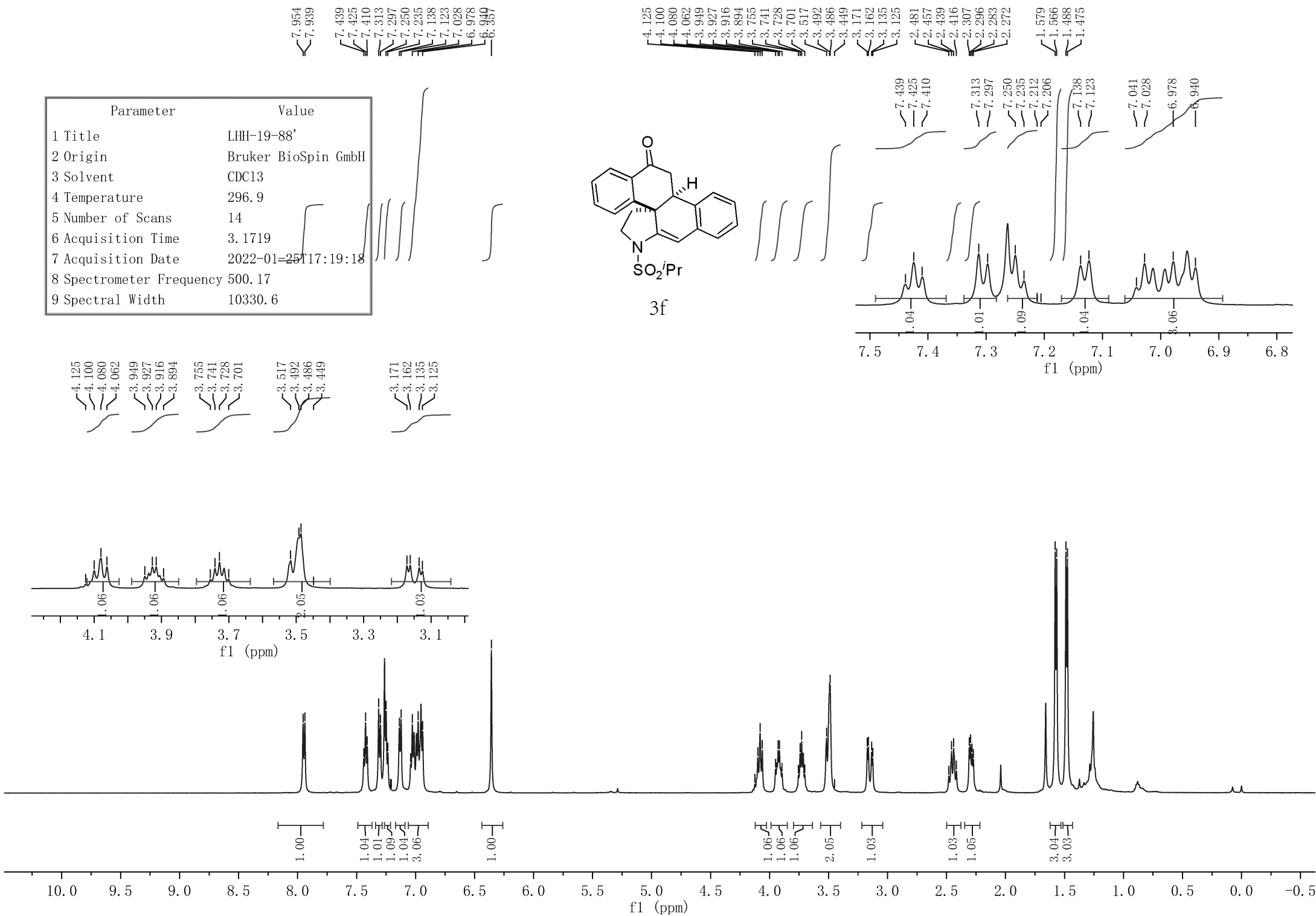
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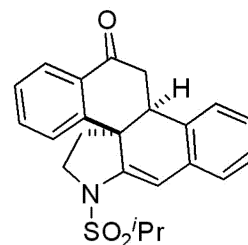
Parameter	Value
1 Title	LHH-19-88'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.9
5 Number of Scans	14
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-25 17:19:18
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



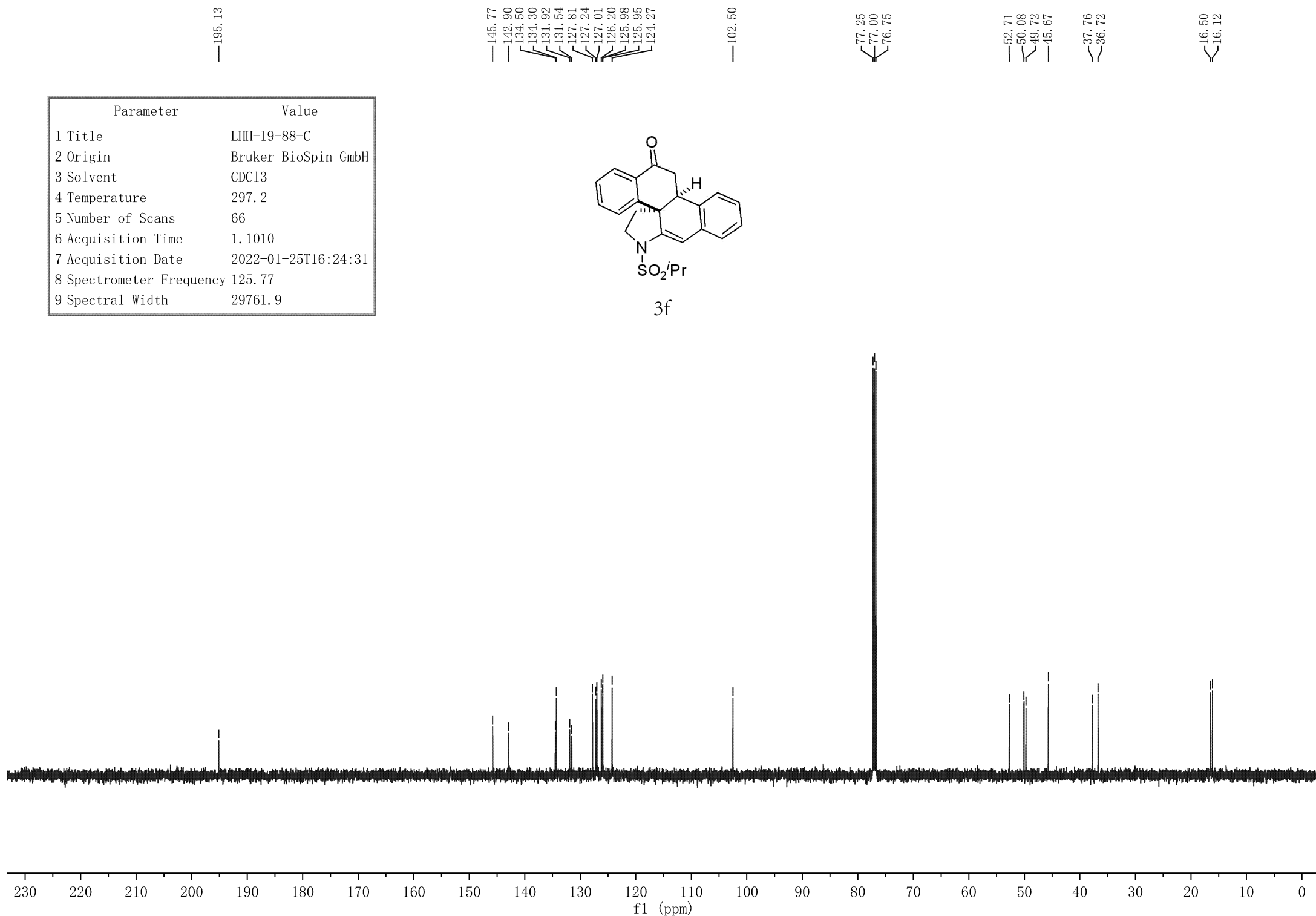
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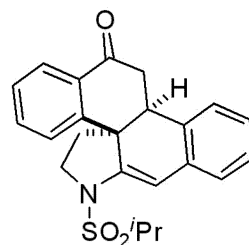
Parameter	Value
1 Title	LHH-19-88-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.2
5 Number of Scans	66
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-25T16:24:31
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



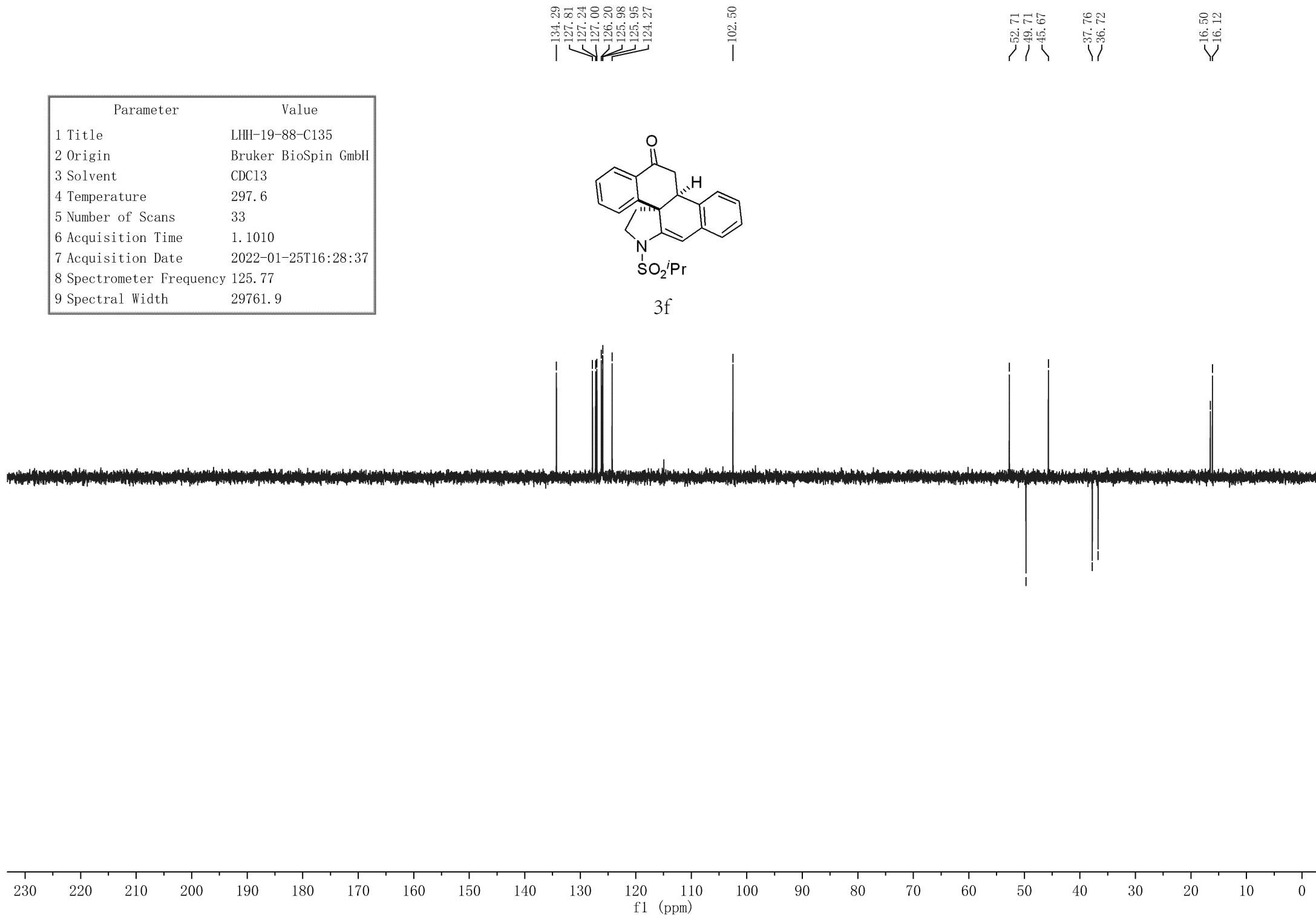
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Parameter	Value
1 Title	LHH-19-88-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.6
5 Number of Scans	33
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-25T16:28:37
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



3f

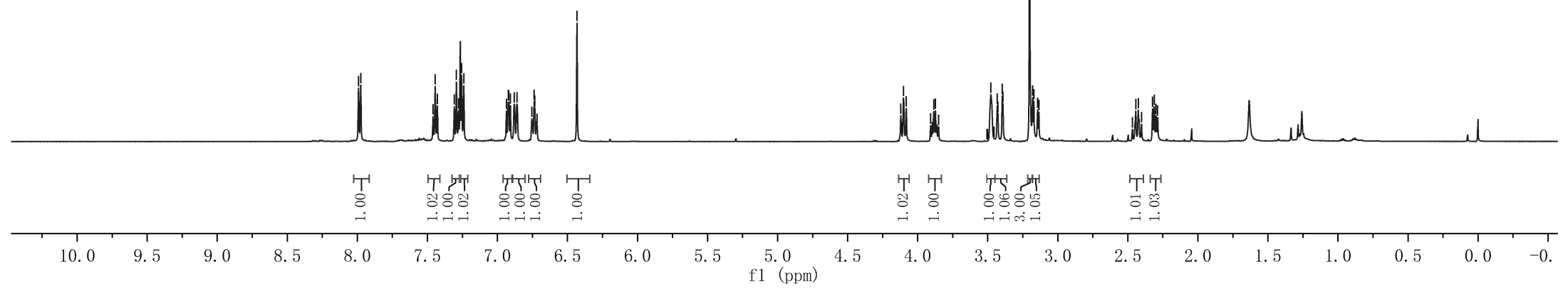
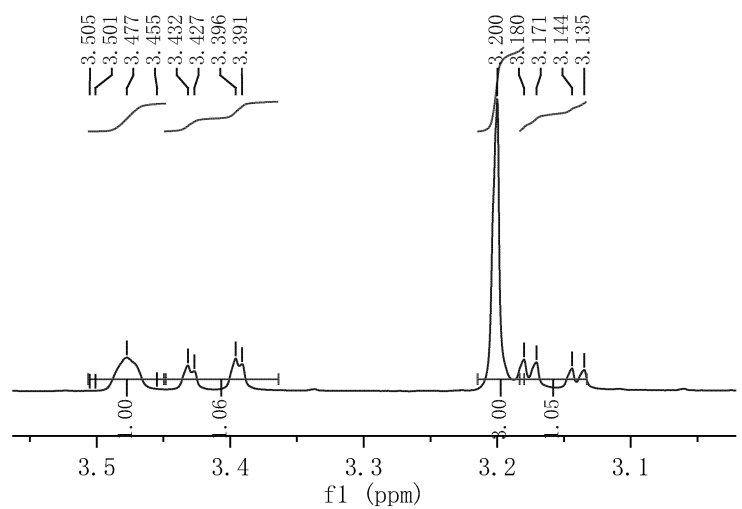
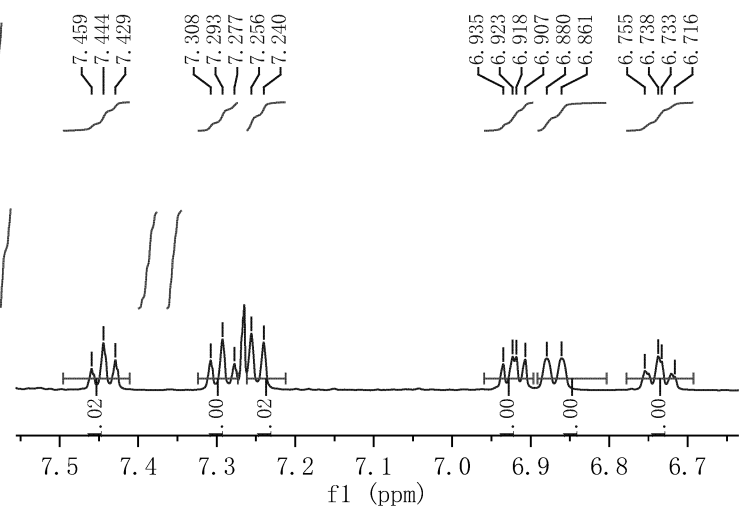
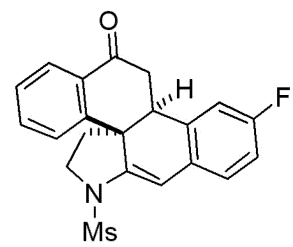


Parameter	Value
1 Title	ZYP-1-95-H'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.0
5 Number of Scans	9
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-06T15:38:49
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

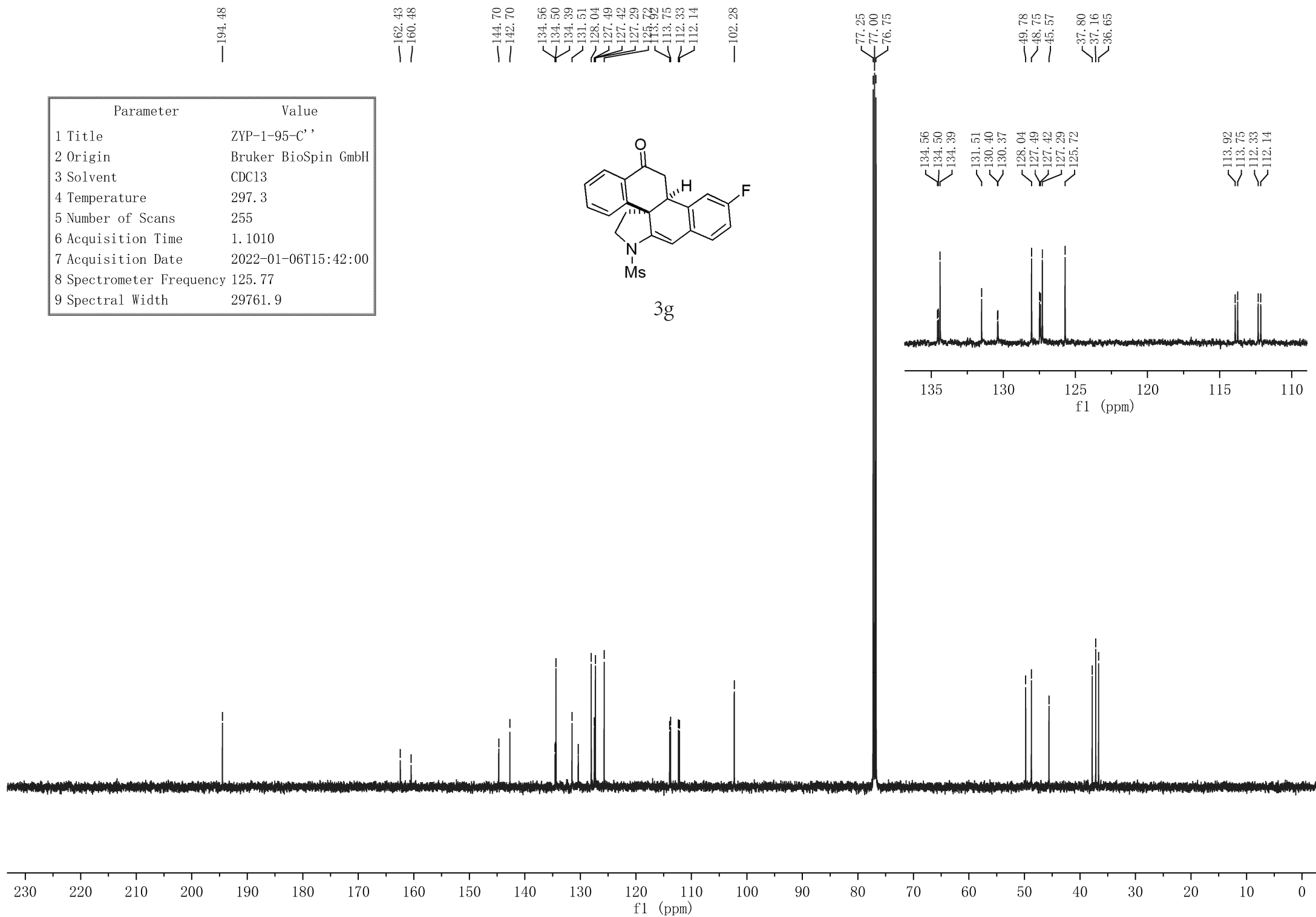
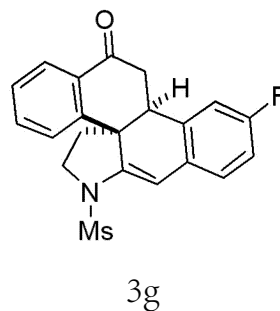
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7.293
7.277
7.256
7.240
6.923
6.918
6.880
6.861
6.733
6.716

4.120
4.113
4.102
4.081
3.907
3.885
3.873
3.851

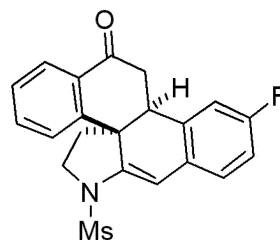
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3.432
3.396
3.391
3.200
3.180
3.467
2.443
2.426
2.402
2.323
2.312
2.299
2.288



Parameter	Value
1 Title	ZYP-1-95-C''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	255
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-06T15:42:00
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



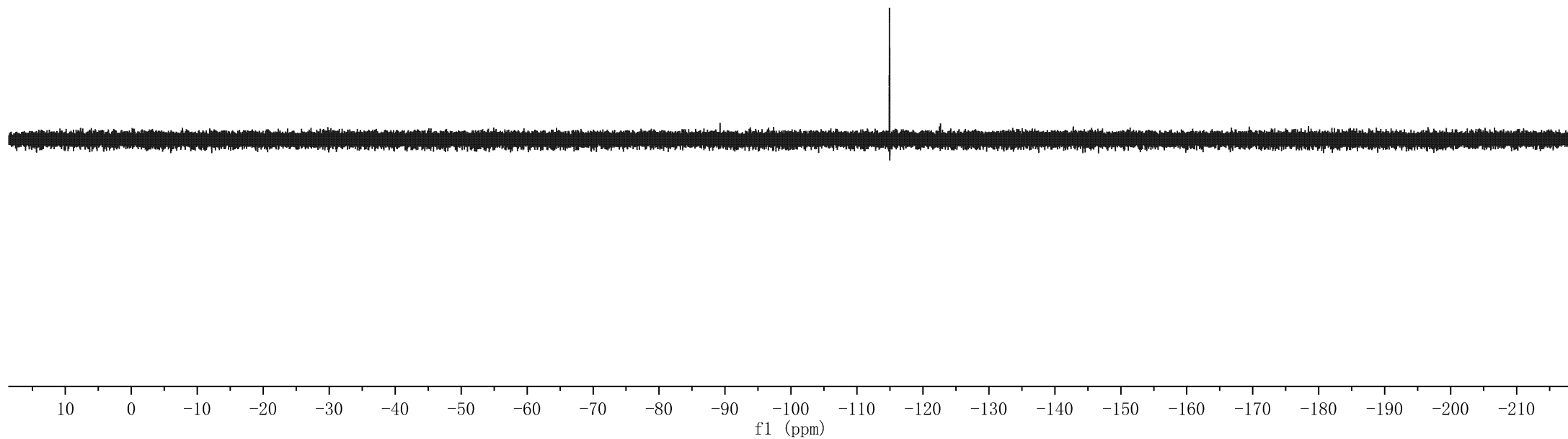
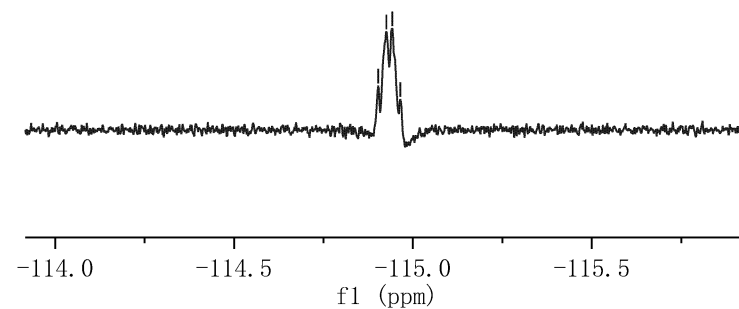
Parameter	Value
1 Title	zyp-1-95-F
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	294.9
5 Number of Scans	16
6 Acquisition Time	0.7340
7 Acquisition Date	2022-01-04T21:23:55
8 Spectrometer Frequency	376.31
9 Spectral Width	89285.7



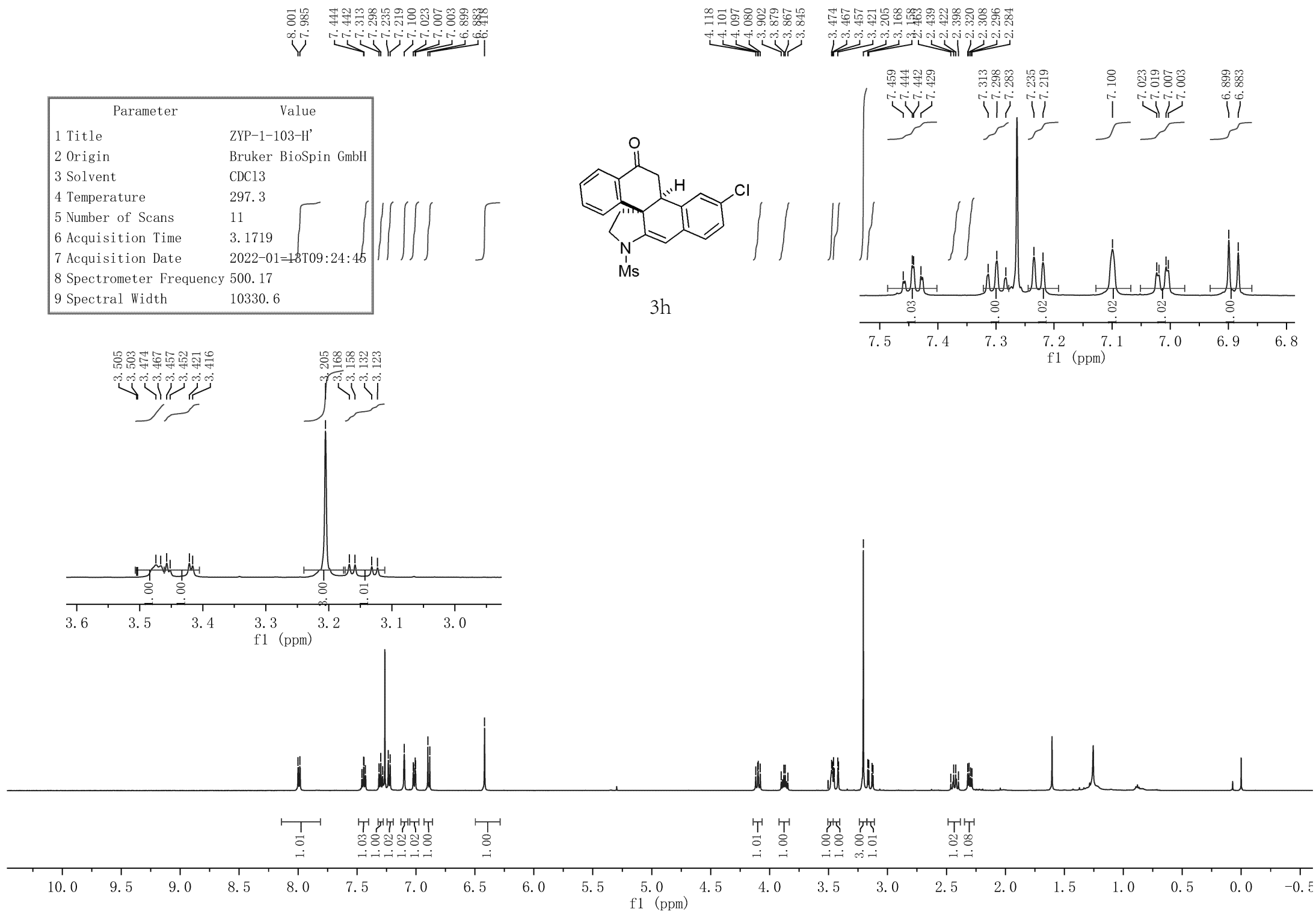
3g

-114.90
-114.93
-114.94
-114.97

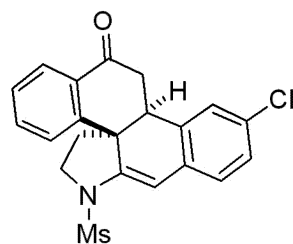
-114.90
-114.93
-114.94
-114.97



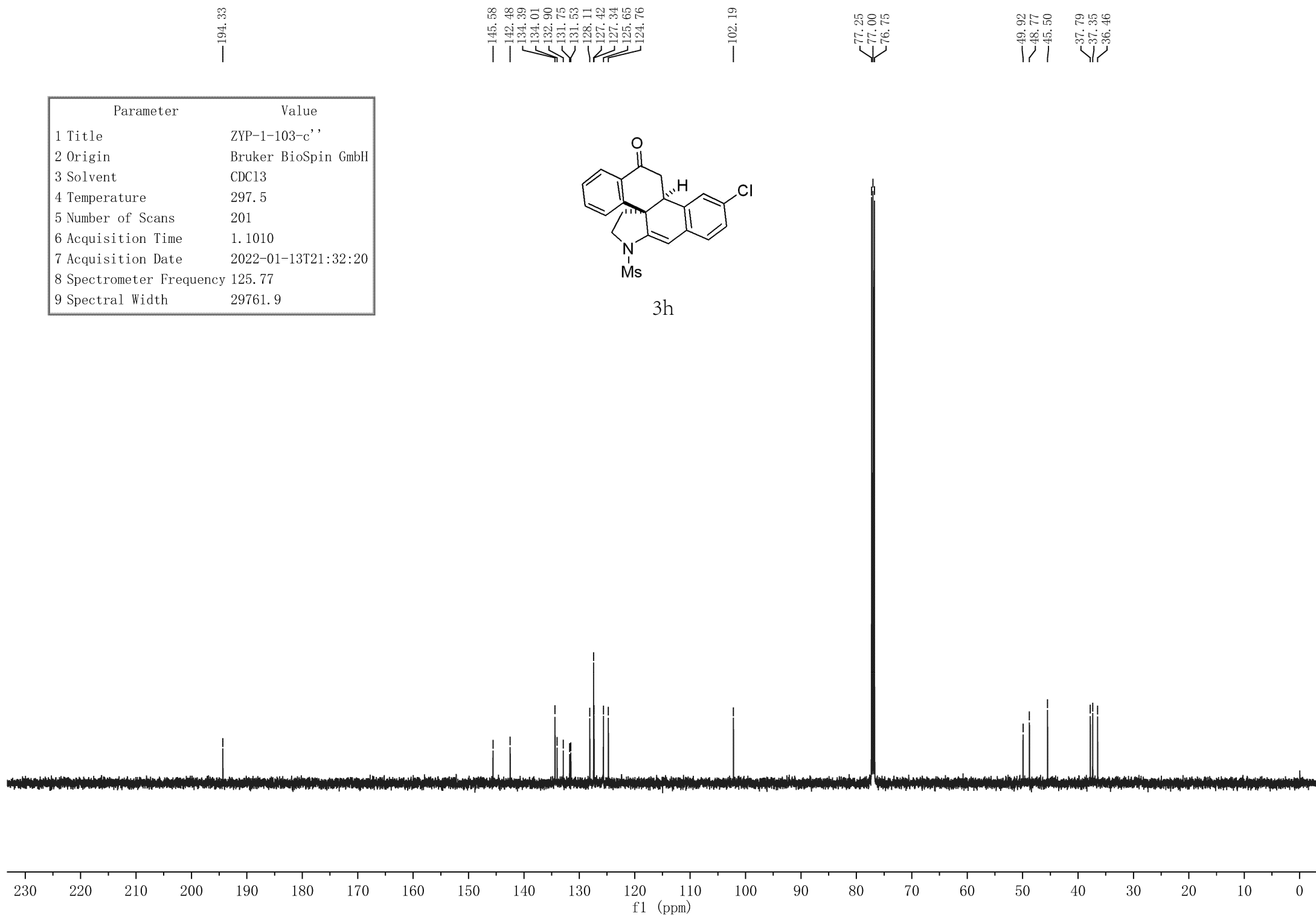
Parameter	Value
1 Title	ZYP-1-103-H'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	11
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-18T09:24:45
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



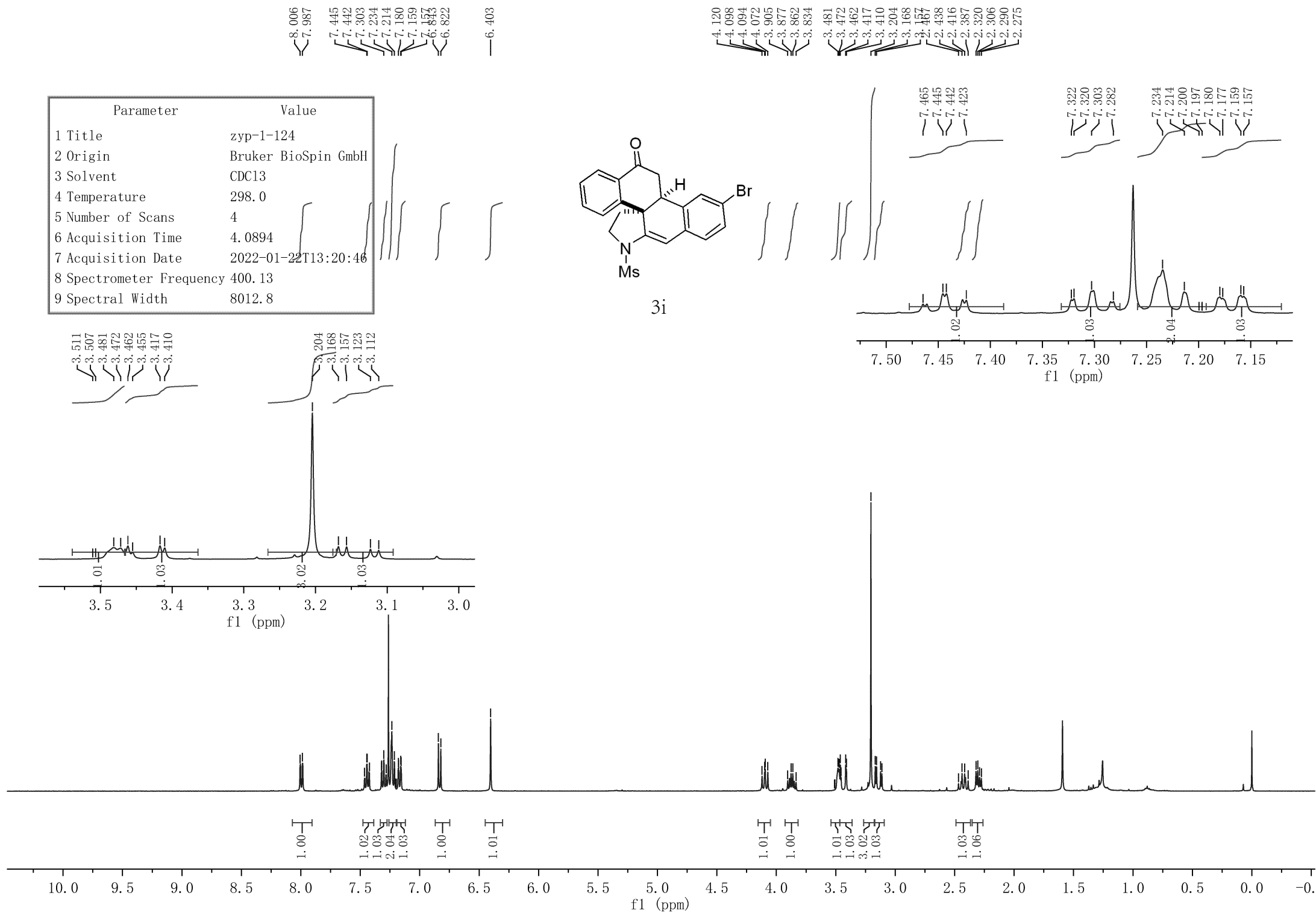
Parameter	Value
1 Title	ZYP-1-103-c''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.5
5 Number of Scans	201
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-13T21:32:20
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



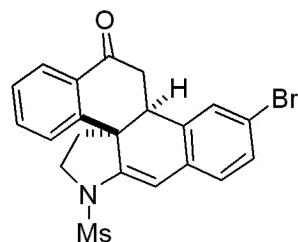
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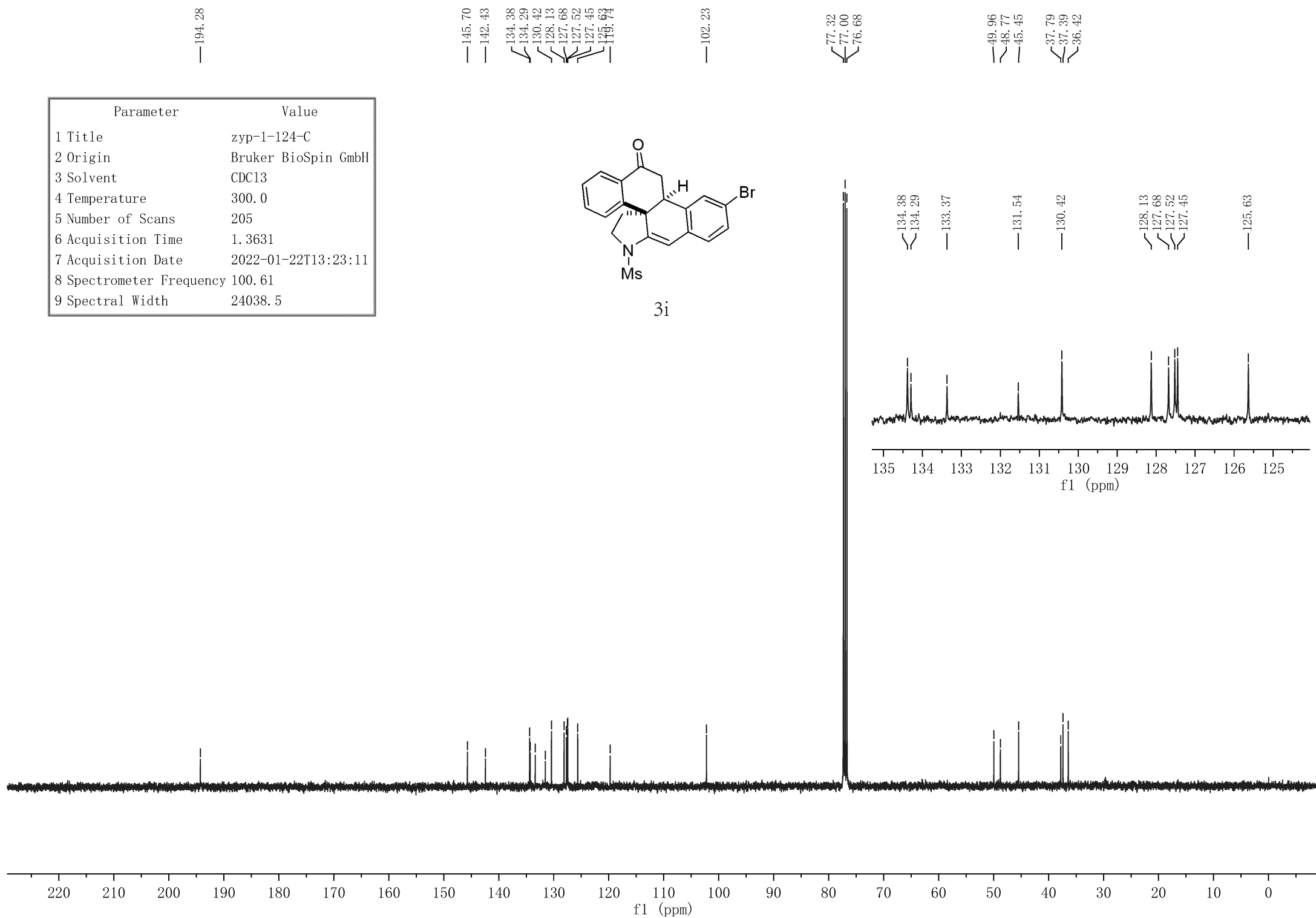
Parameter	Value
1 Title	zyp-1-124
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	4
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-22T13:20:46
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



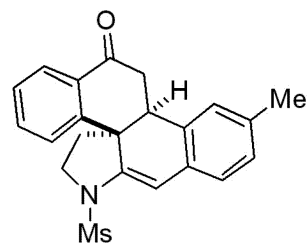
Parameter	Value
1 Title	zyp-1-124-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	205
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-22T13:23:11
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



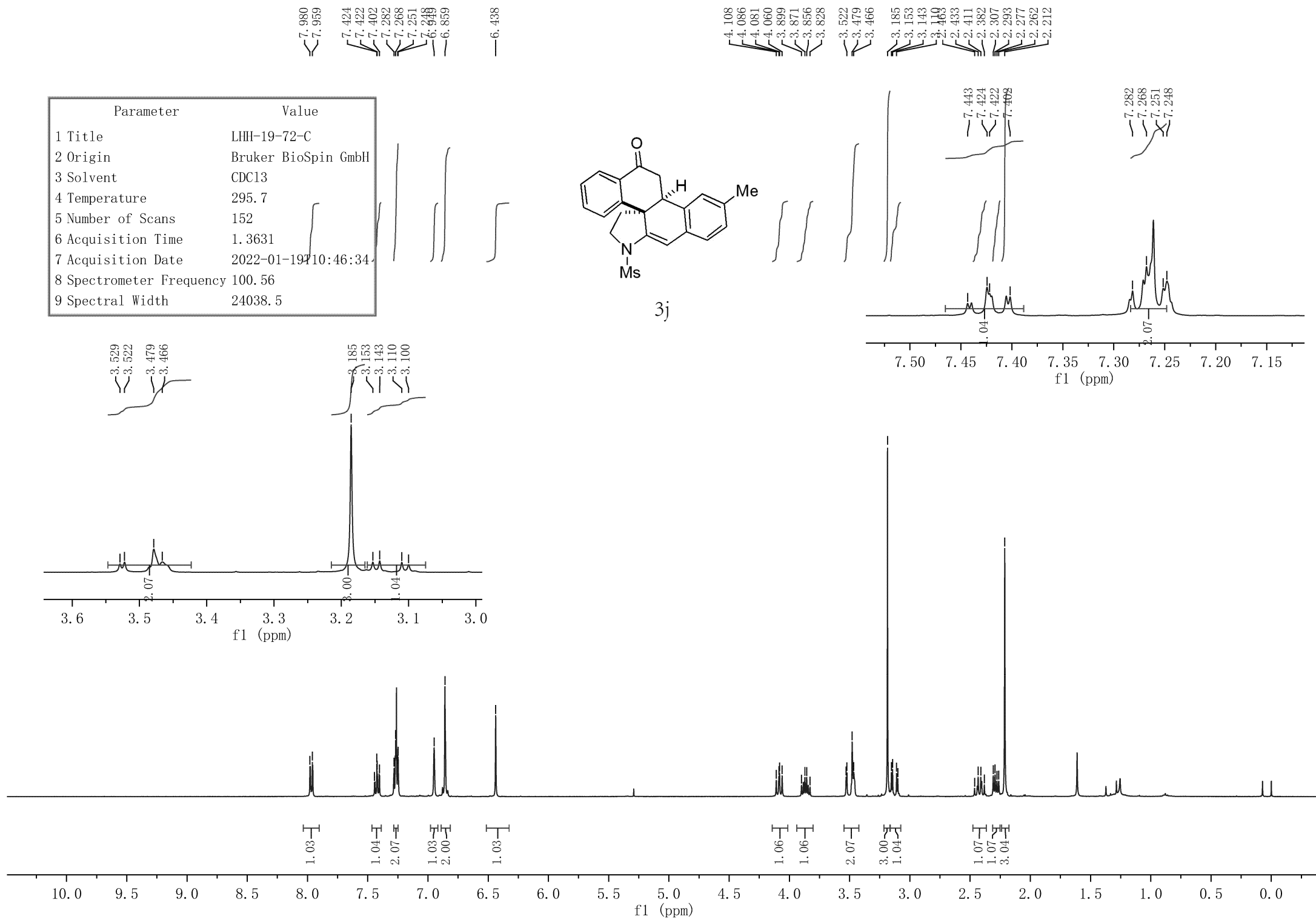
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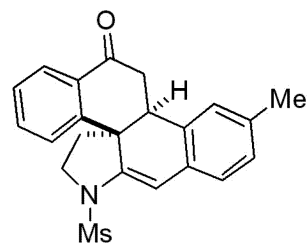
Parameter	Value
1 Title	LHH-19-72-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.7
5 Number of Scans	152
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-19T10:46:34
8 Spectrometer Frequency	100.56
9 Spectral Width	24038.5



3j



Parameter	Value
1 Title	LHH-19-72-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	295.7
5 Number of Scans	152
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-19T10:46:34
8 Spectrometer Frequency	100.56
9 Spectral Width	24038.5



3j

195.13

144.26
143.20
136.06
134.26
132.01
131.55
131.45
127.85
127.82
127.16
126.30
125.81
125.19

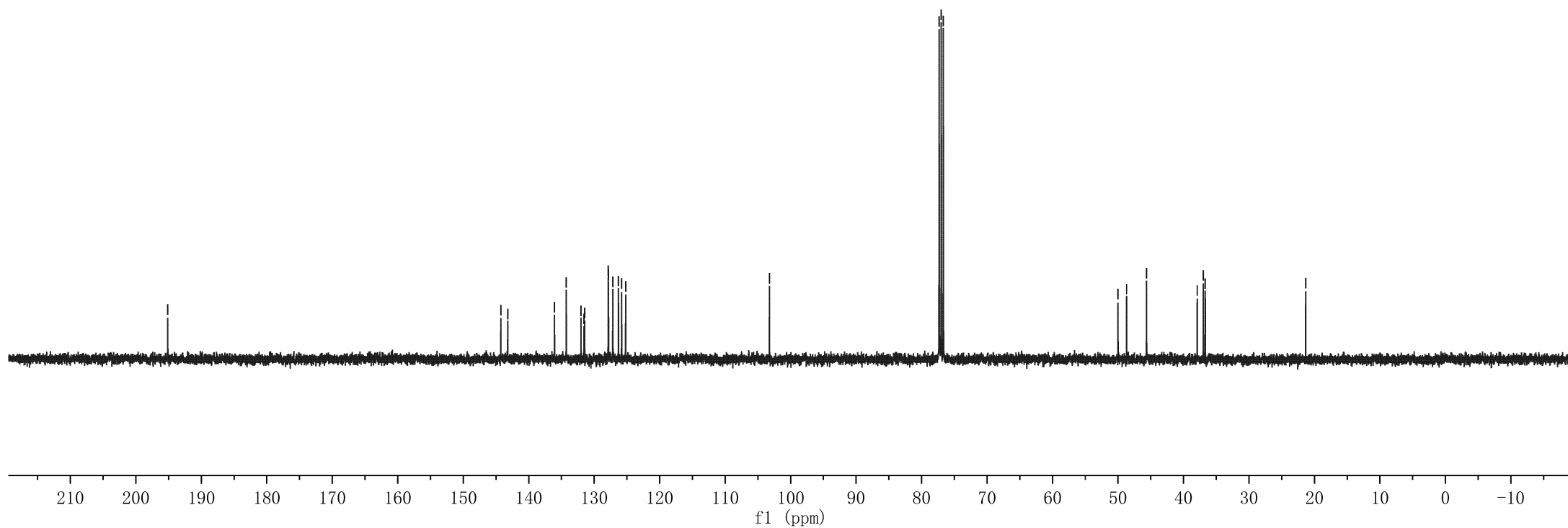
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77.32
77.00
76.68

50.01
48.69
45.64

37.91
36.97
36.68

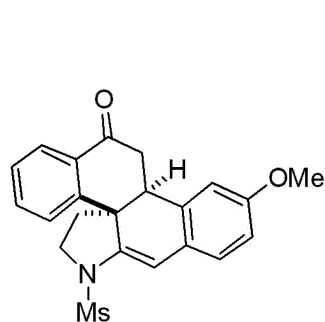
21.34



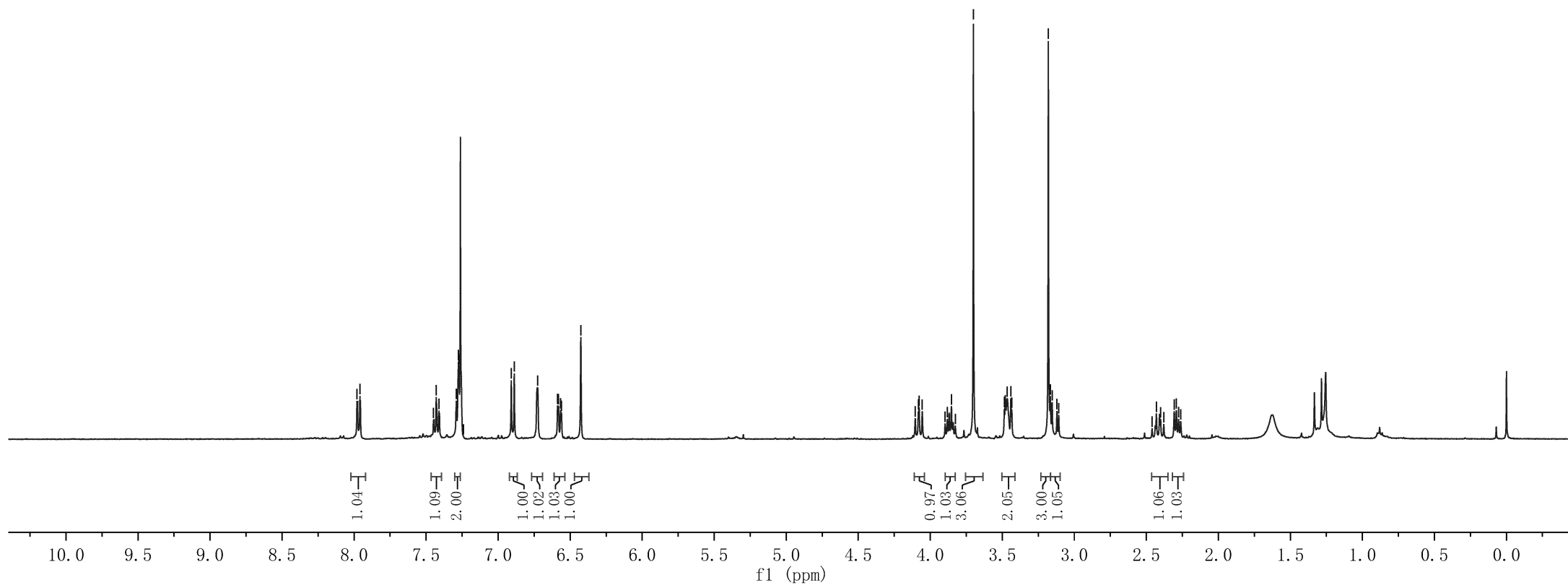
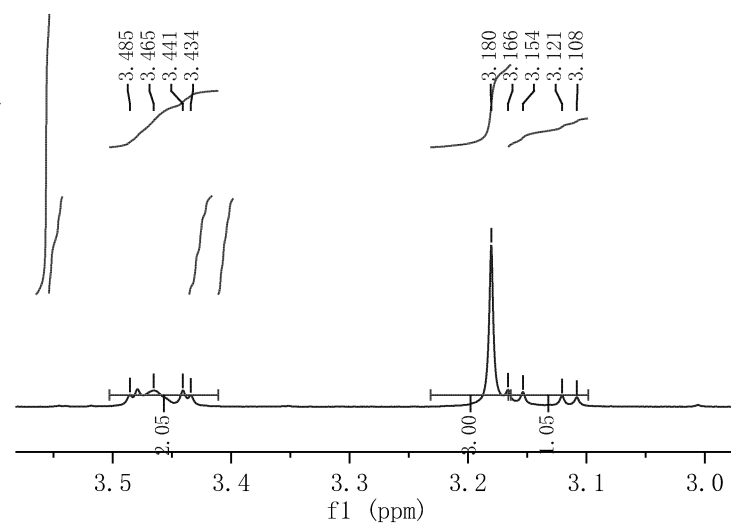
Parameter	Value
1 Title	LHH-19-29'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	3
6 Acquisition Time	4.0894
7 Acquisition Date	2022-01-09T17:01:49
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8

7.980
7.959
7.449
7.430
7.411
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7.241
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6.587
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6.561
6.426

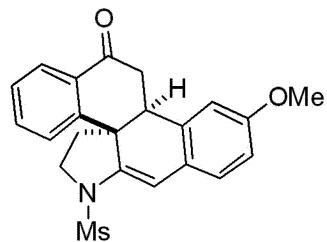
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2.430
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2.292
2.276
2.261



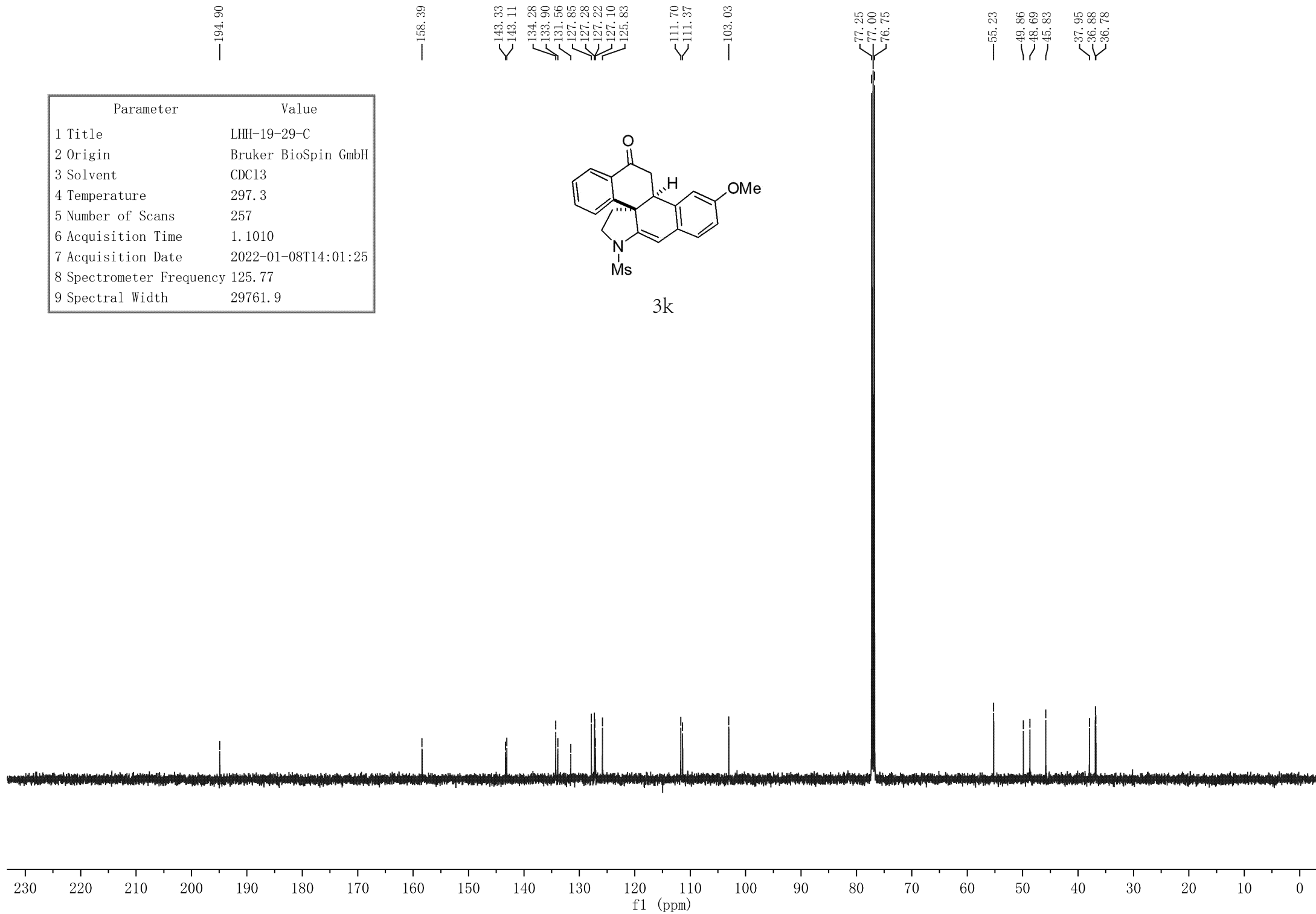
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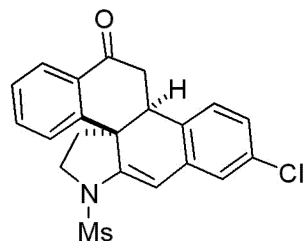
Parameter	Value
1 Title	LHH-19-29-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.3
5 Number of Scans	257
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-08T14:01:25
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



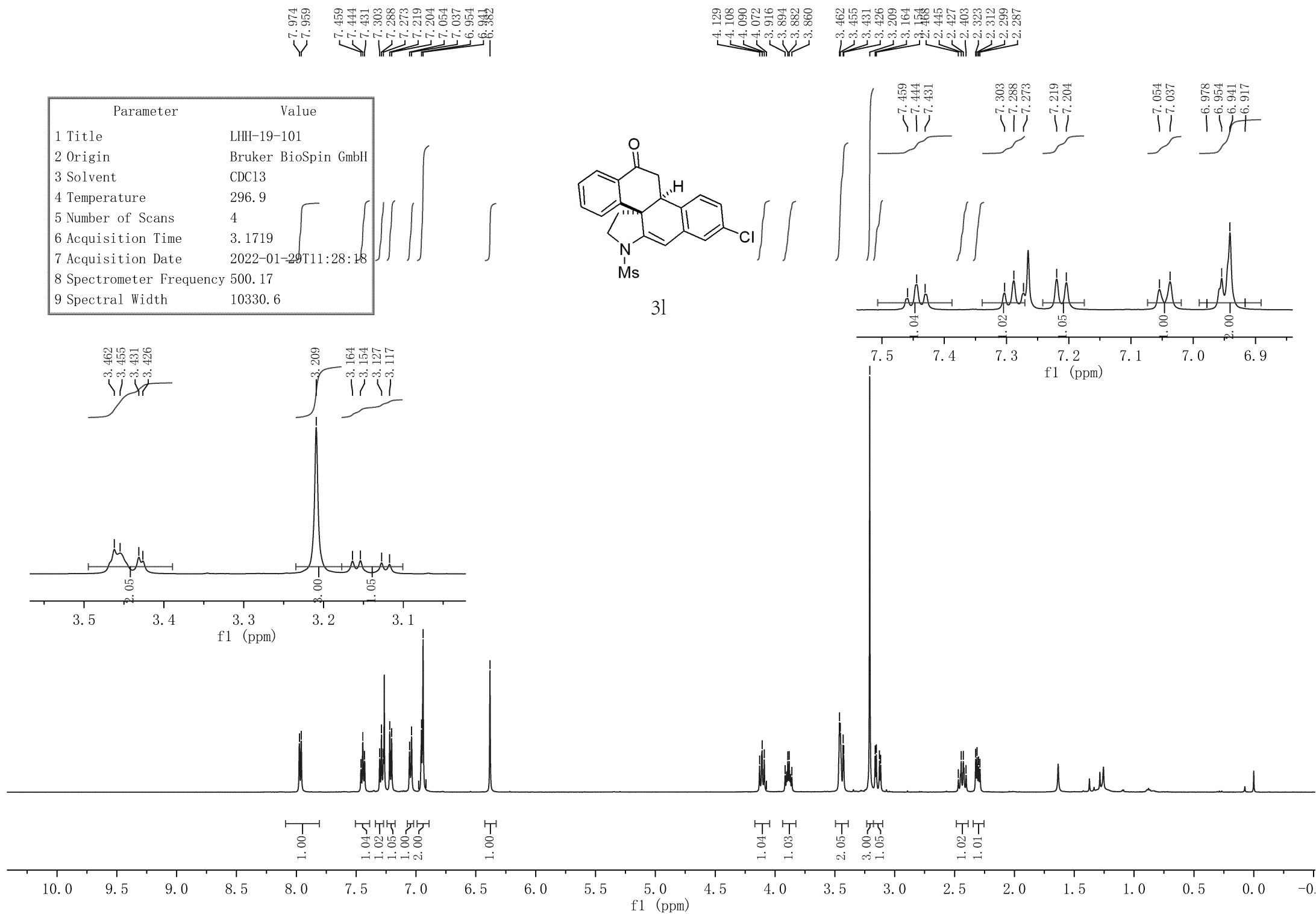
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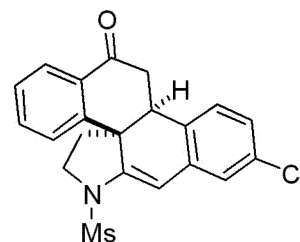
Parameter	Value
1 Title	LHH-19-101
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.9
5 Number of Scans	4
6 Acquisition Time	3.1719
7 Acquisition Date	2022-01-29T11:28:18
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6



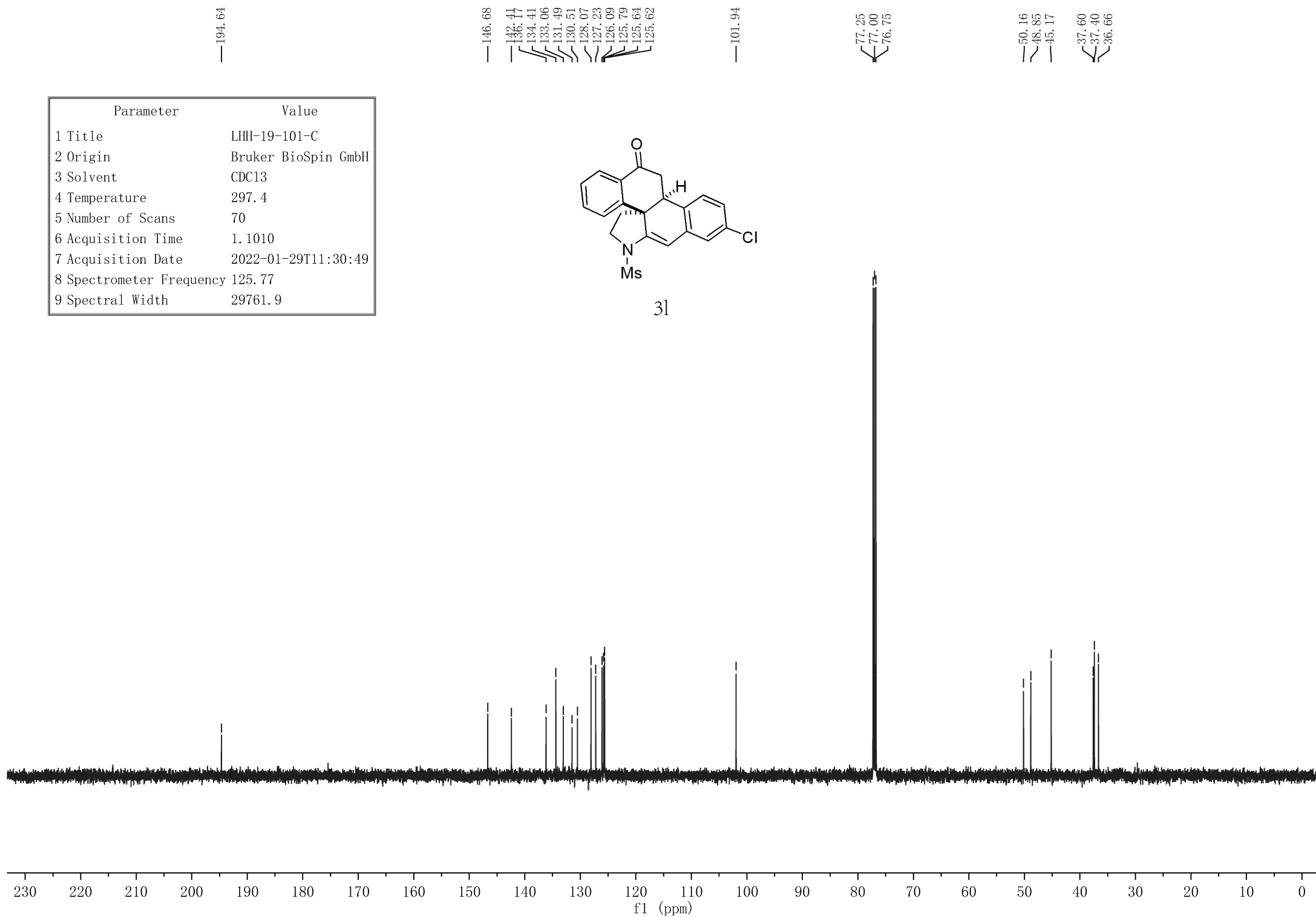
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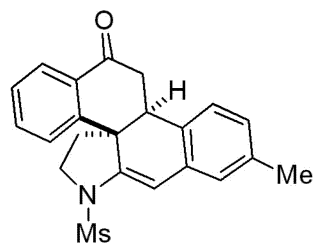
Parameter	Value
1 Title	LHH-19-101-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	297.4
5 Number of Scans	70
6 Acquisition Time	1.1010
7 Acquisition Date	2022-01-29T11:30:49
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



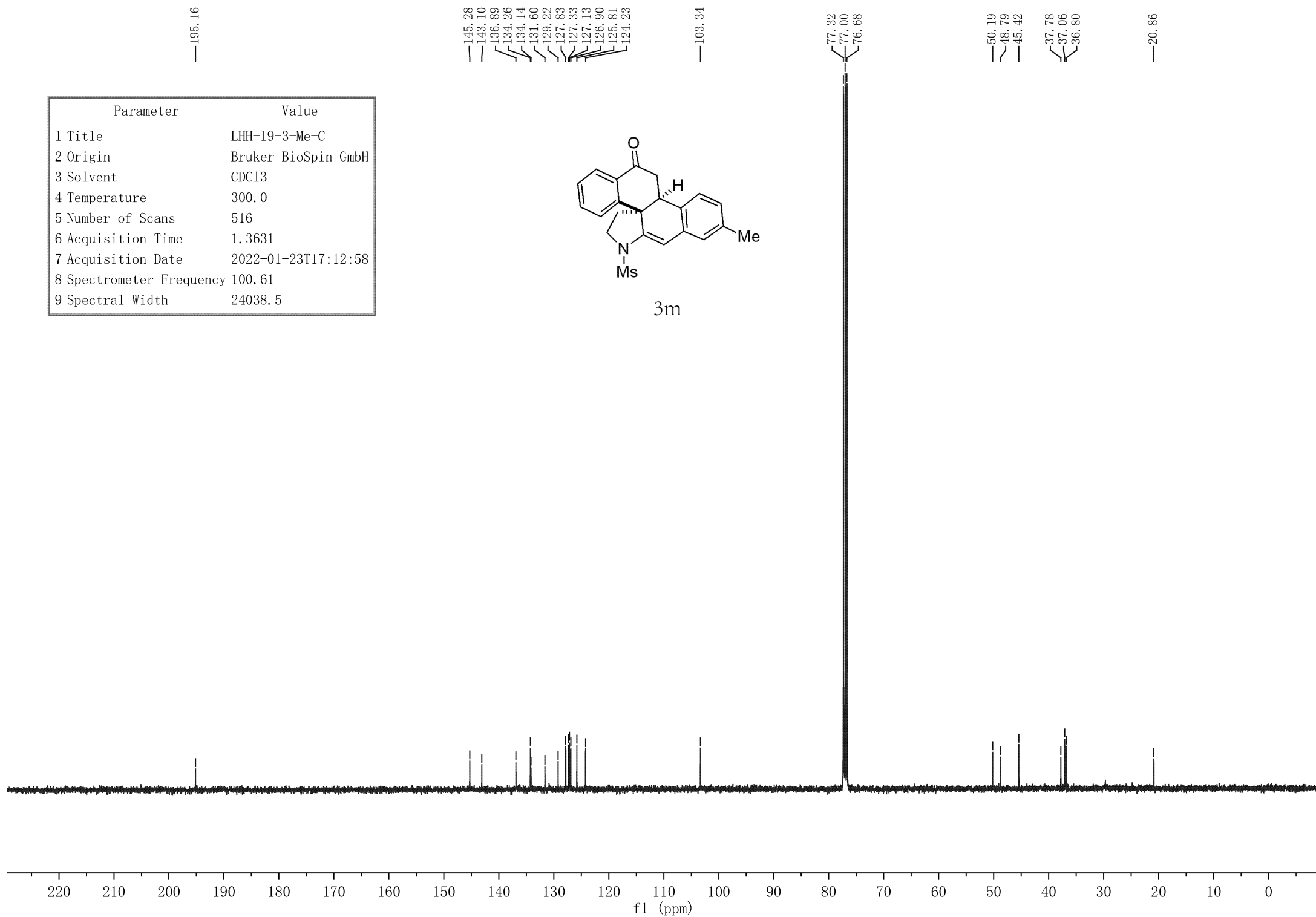
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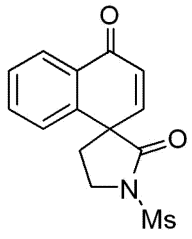
Parameter	Value
1 Title	LHH-19-3-Me-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	516
6 Acquisition Time	1.3631
7 Acquisition Date	2022-01-23T17:12:58
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



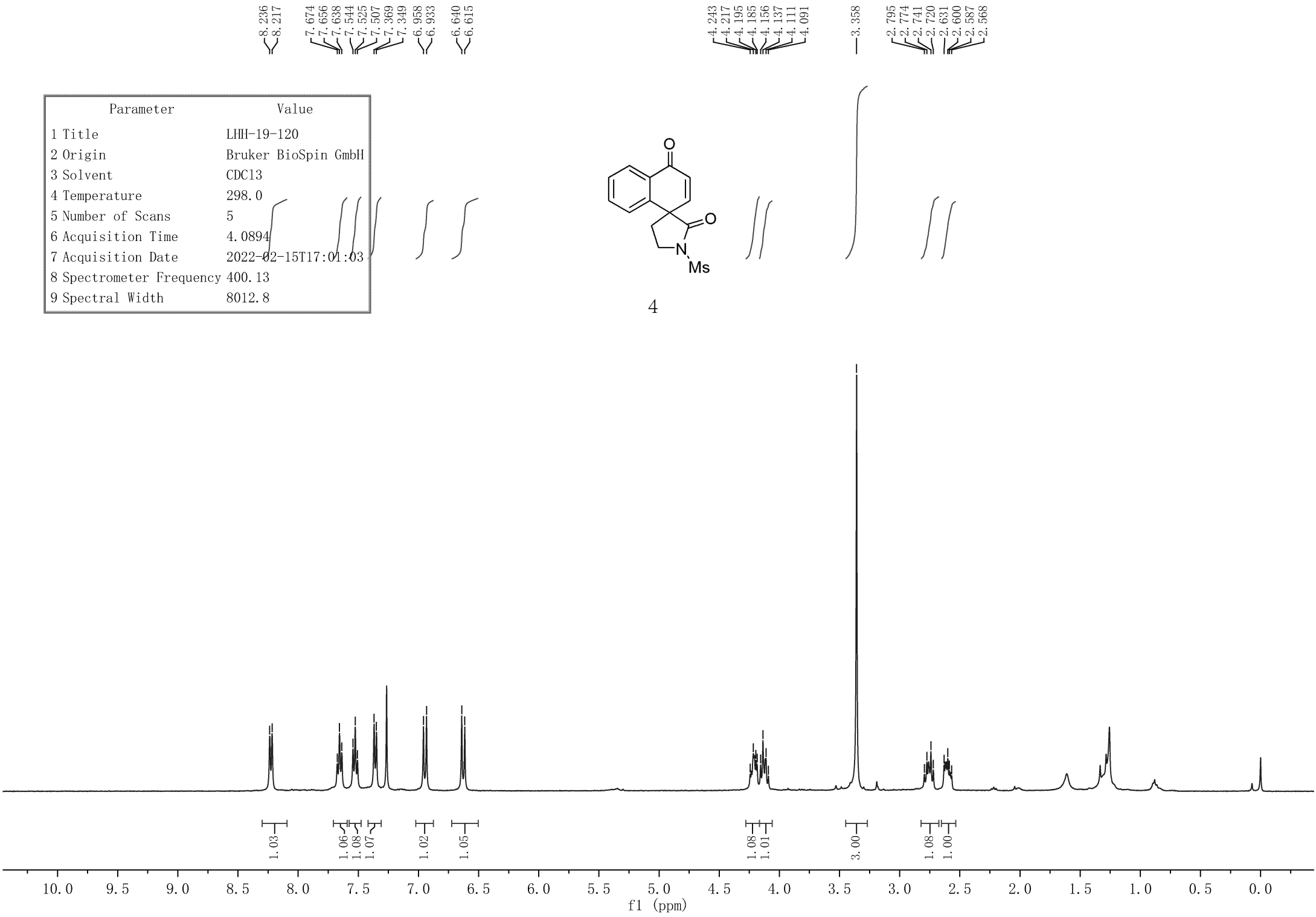
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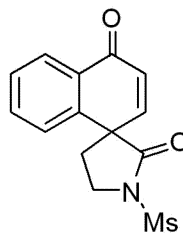
Parameter	Value
1 Title	LHH-19-120
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	5
6 Acquisition Time	4.0894
7 Acquisition Date	2022-02-15T17:01:03
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



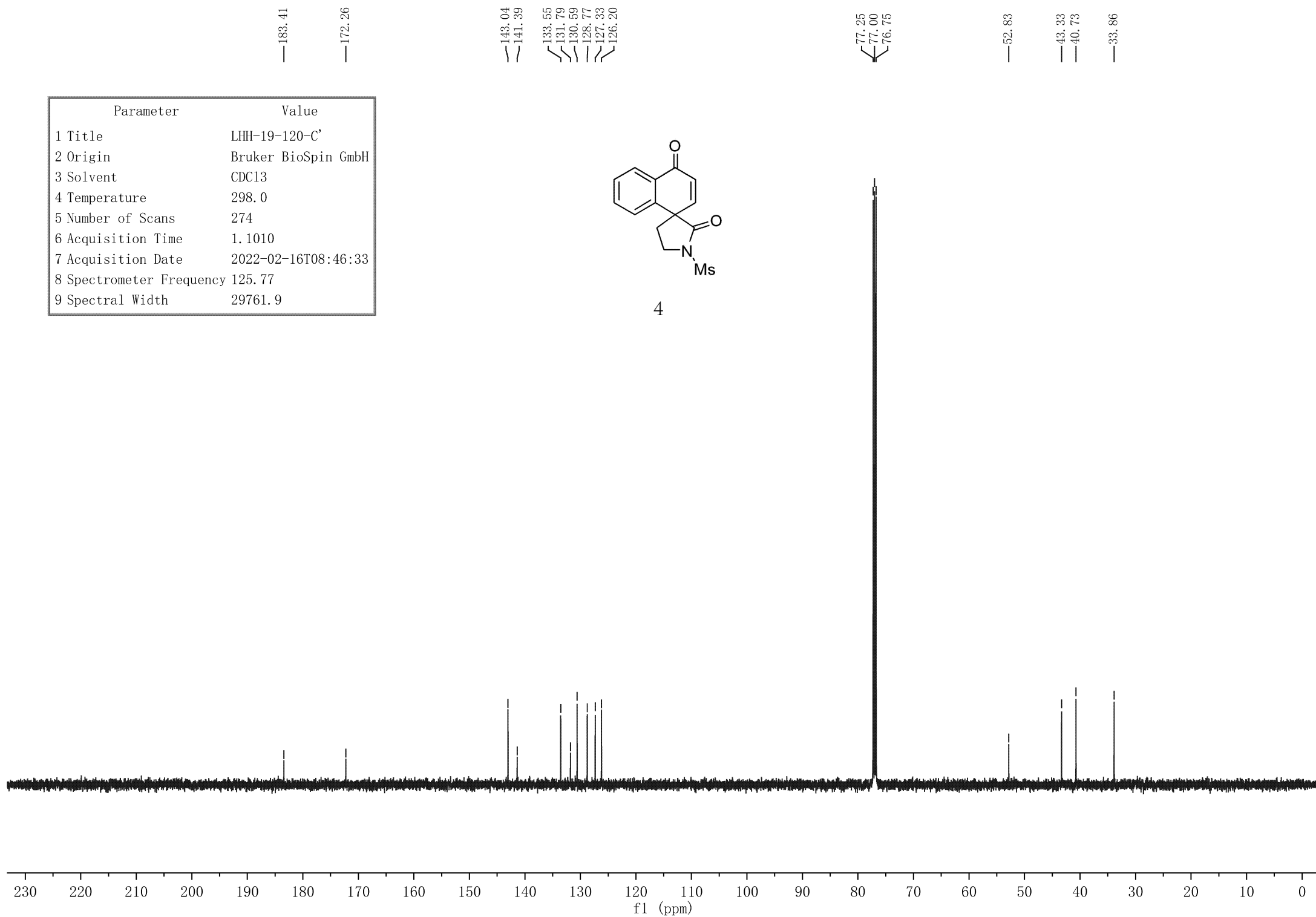
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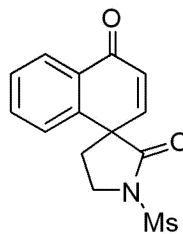
Parameter	Value
1 Title	LHH-19-120-C'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	274
6 Acquisition Time	1.1010
7 Acquisition Date	2022-02-16T08:46:33
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



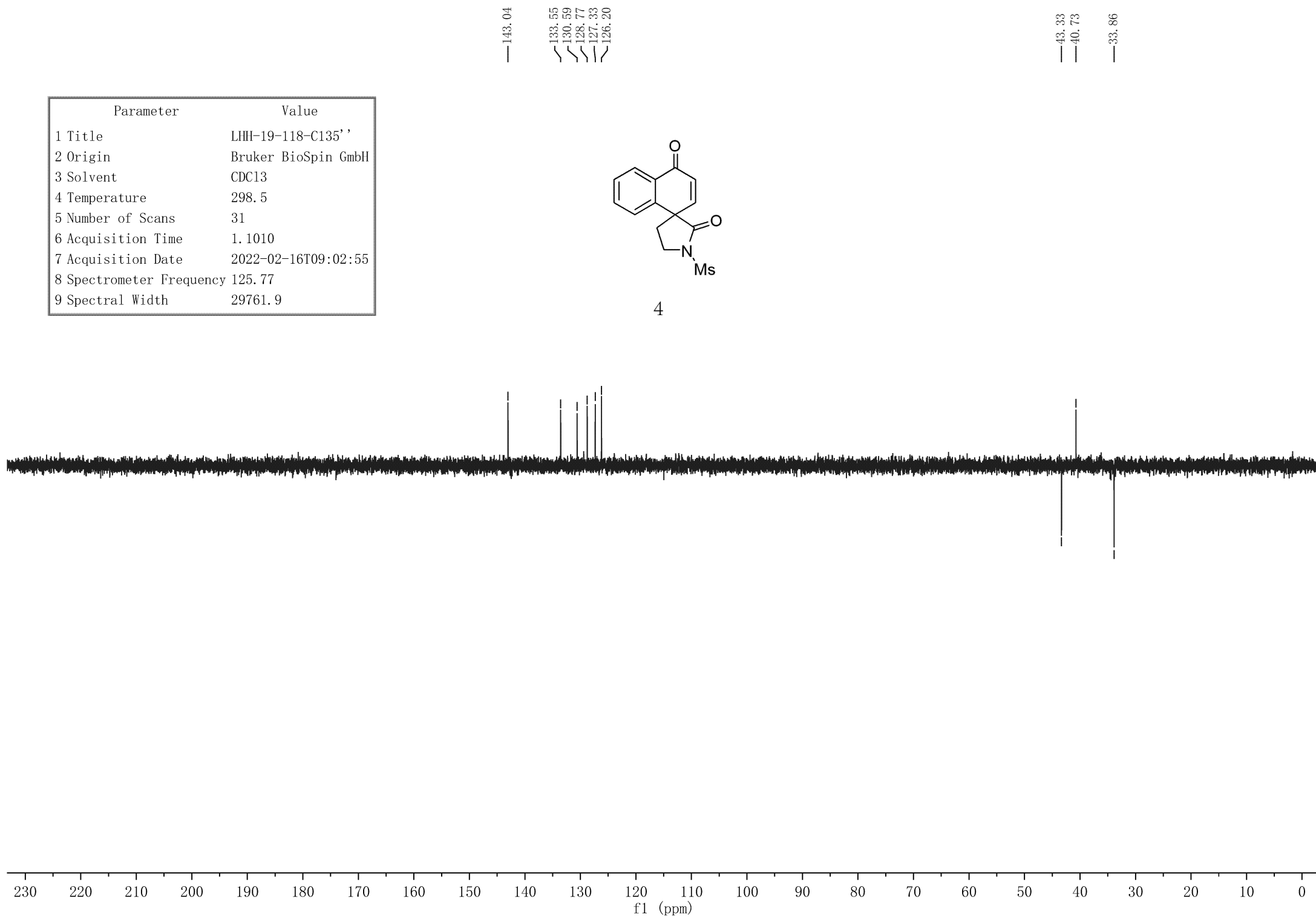
4



Parameter	Value
1 Title	LHH-19-118-C135''
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.5
5 Number of Scans	31
6 Acquisition Time	1.1010
7 Acquisition Date	2022-02-16T09:02:55
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



4

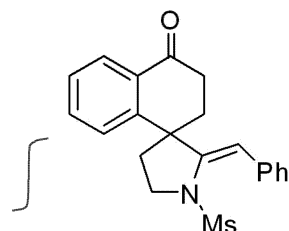


Parameter	Value
1 Title	LHH-19-130-H
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.7
5 Number of Scans	12
6 Acquisition Time	3.1719
7 Acquisition Date	2022-02-20T18:28:08
8 Spectrometer Frequency	500.17
9 Spectral Width	10330.6

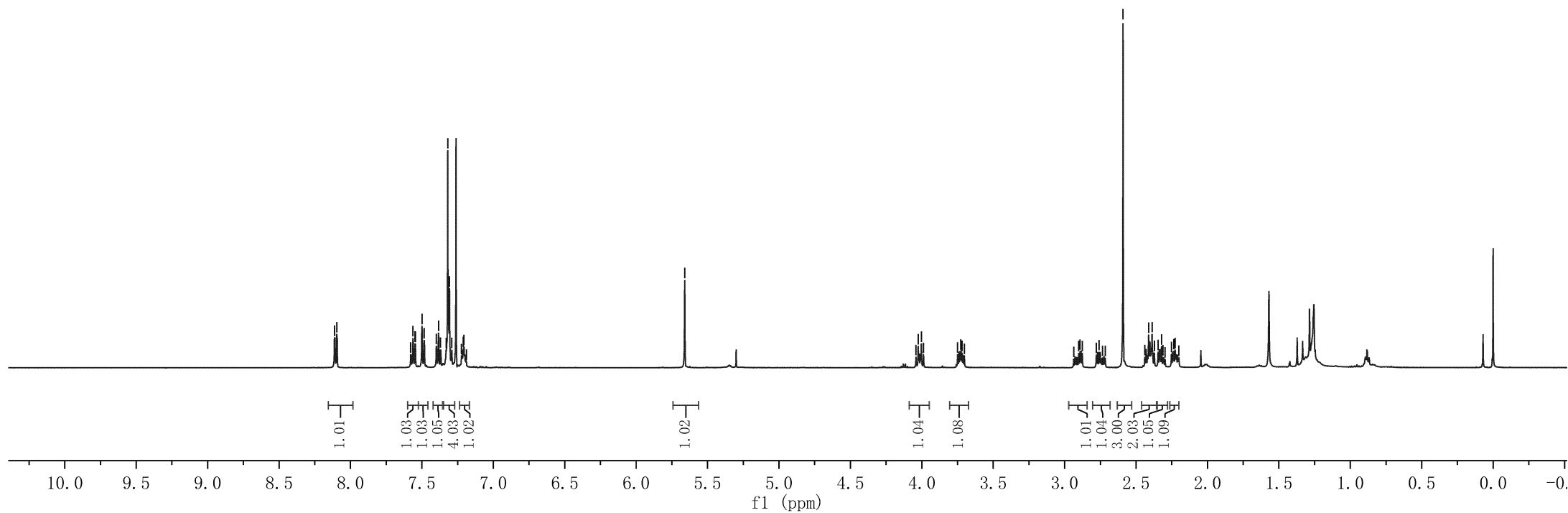
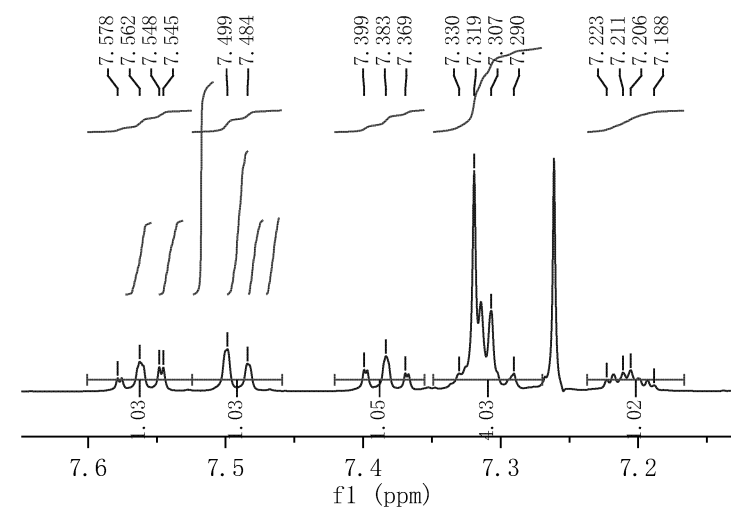
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7.223
7.211
7.206
7.188

5.660

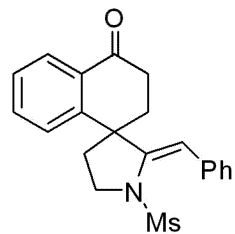
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2.778
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2.734
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2.591
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2.253
2.236
2.227
2.200



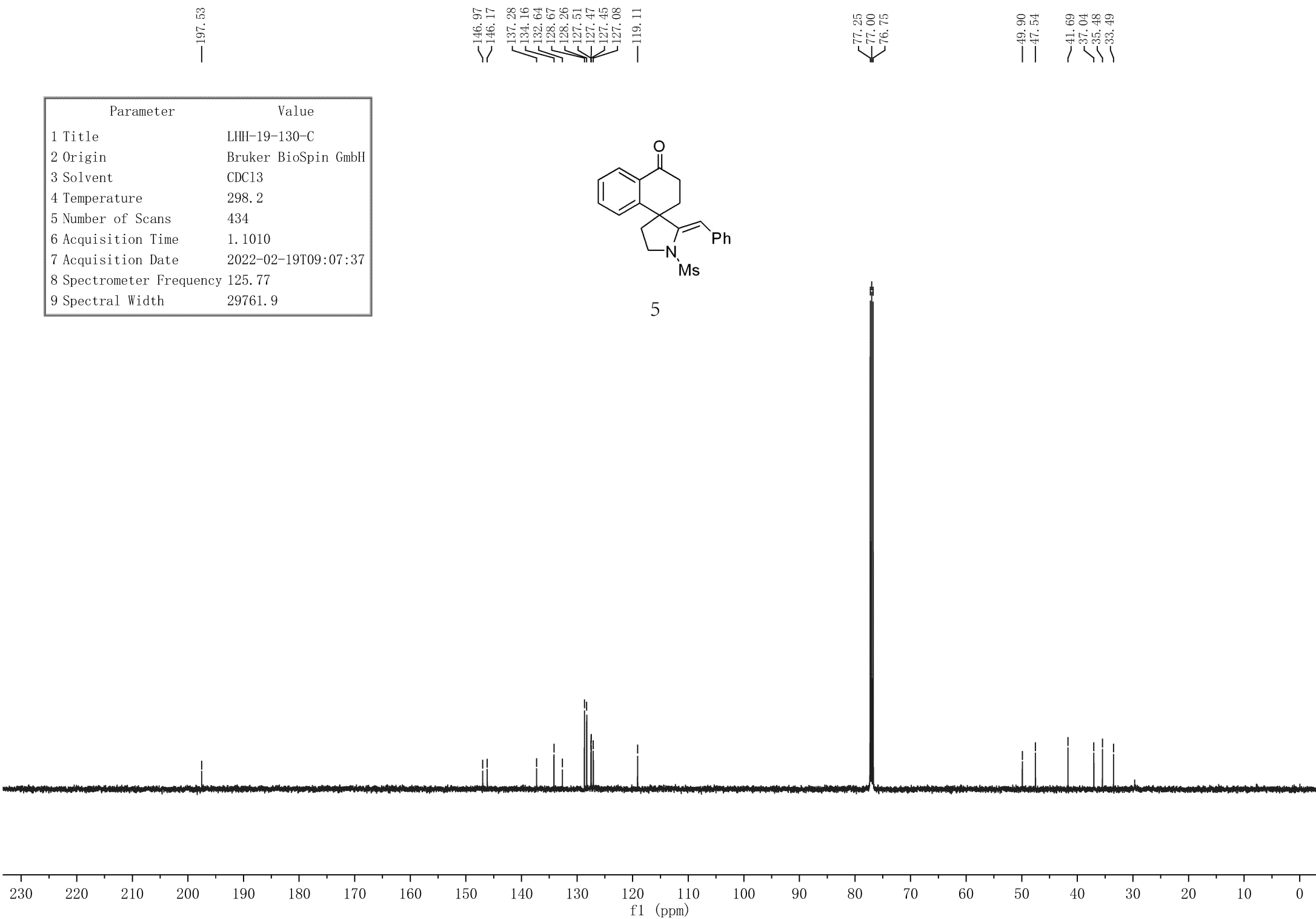
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Parameter	Value
1 Title	LHH-19-130-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.2
5 Number of Scans	434
6 Acquisition Time	1.1010
7 Acquisition Date	2022-02-19T09:07:37
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



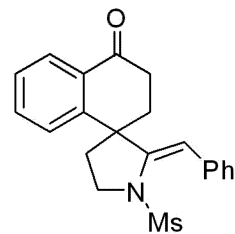
5



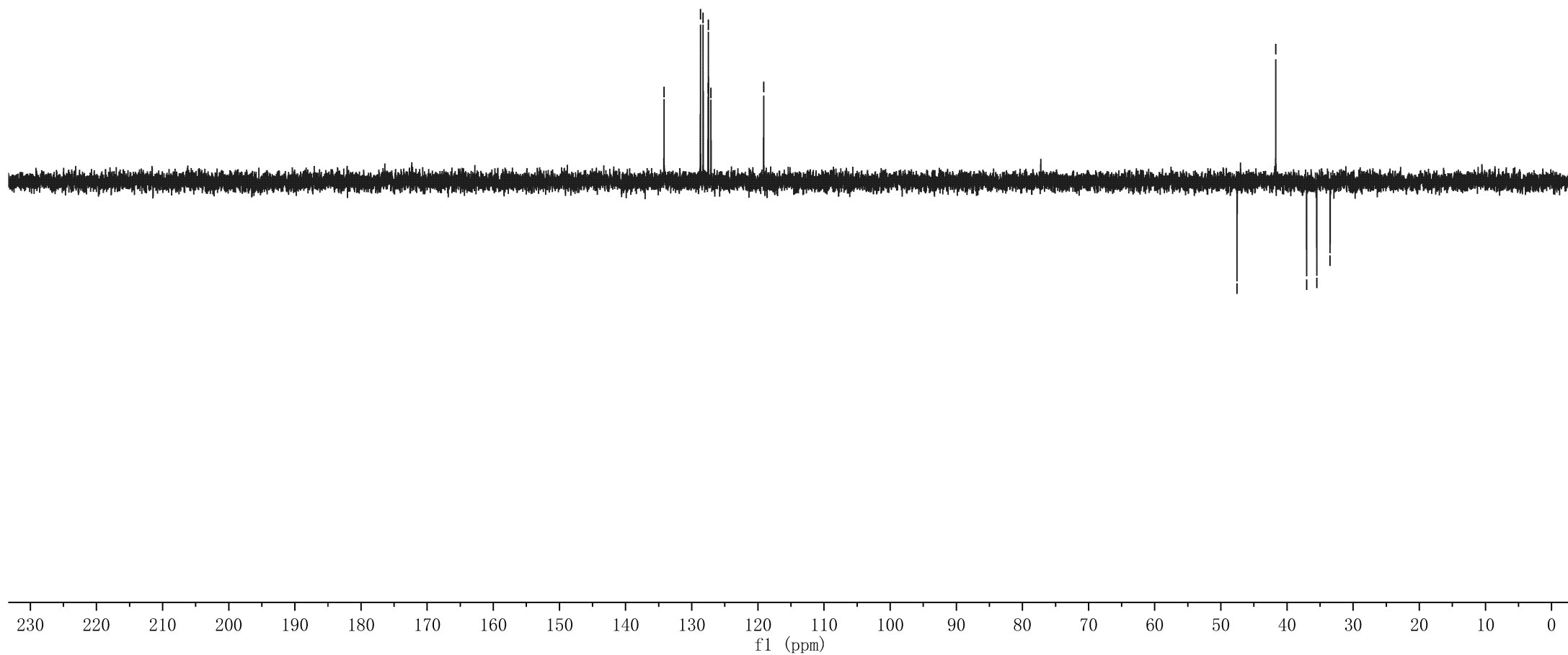
134.19
128.68
128.28
127.53
127.48
127.10
119.12

47.56
41.70
37.04
35.50
33.49

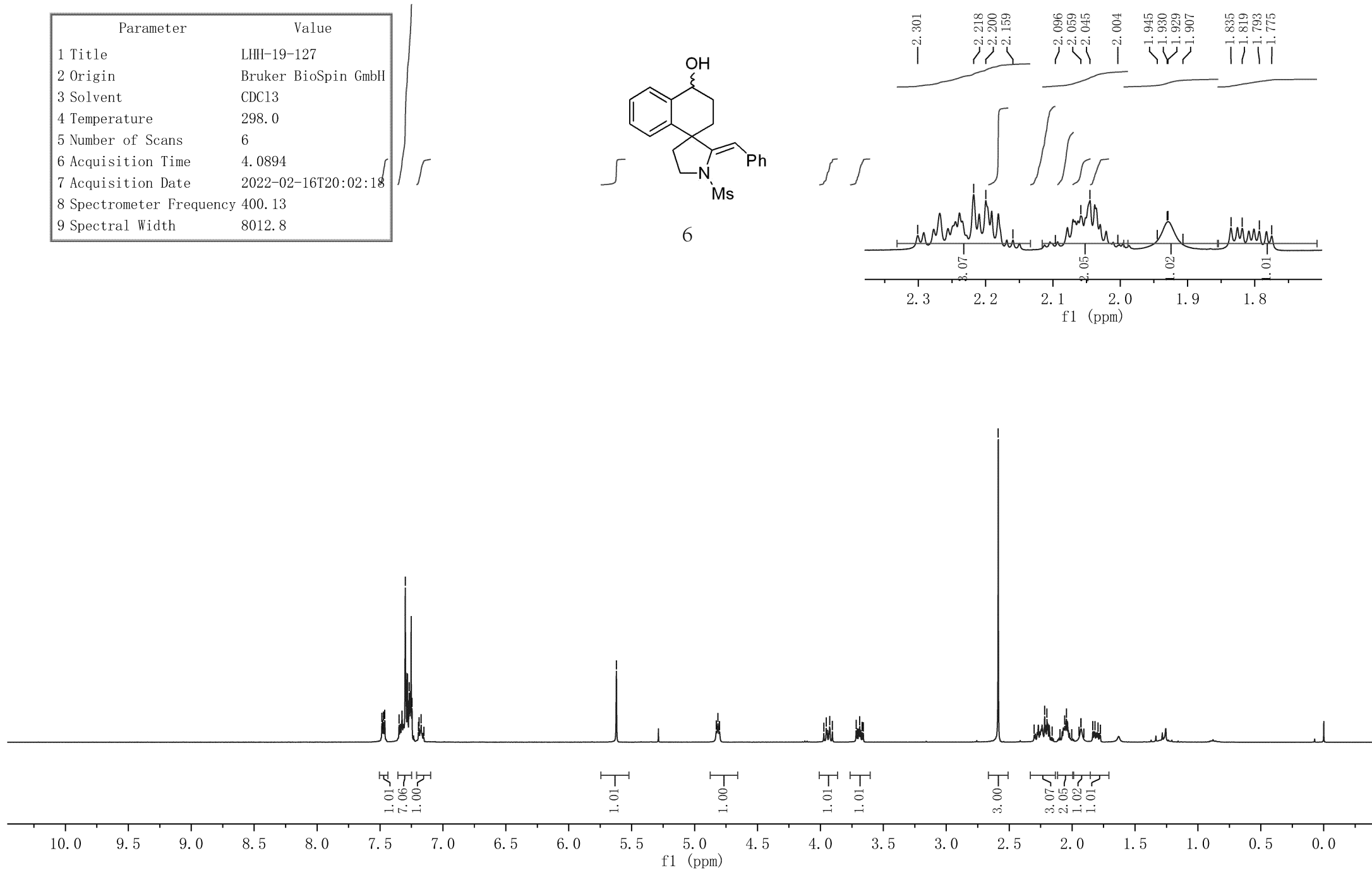
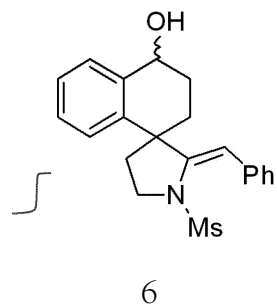
Parameter	Value
1 Title	LHH-19-130-C135'
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	296.9
5 Number of Scans	112
6 Acquisition Time	1.1010
7 Acquisition Date	2022-02-20T16:23:49
8 Spectrometer Frequency	125.77
9 Spectral Width	29761.9



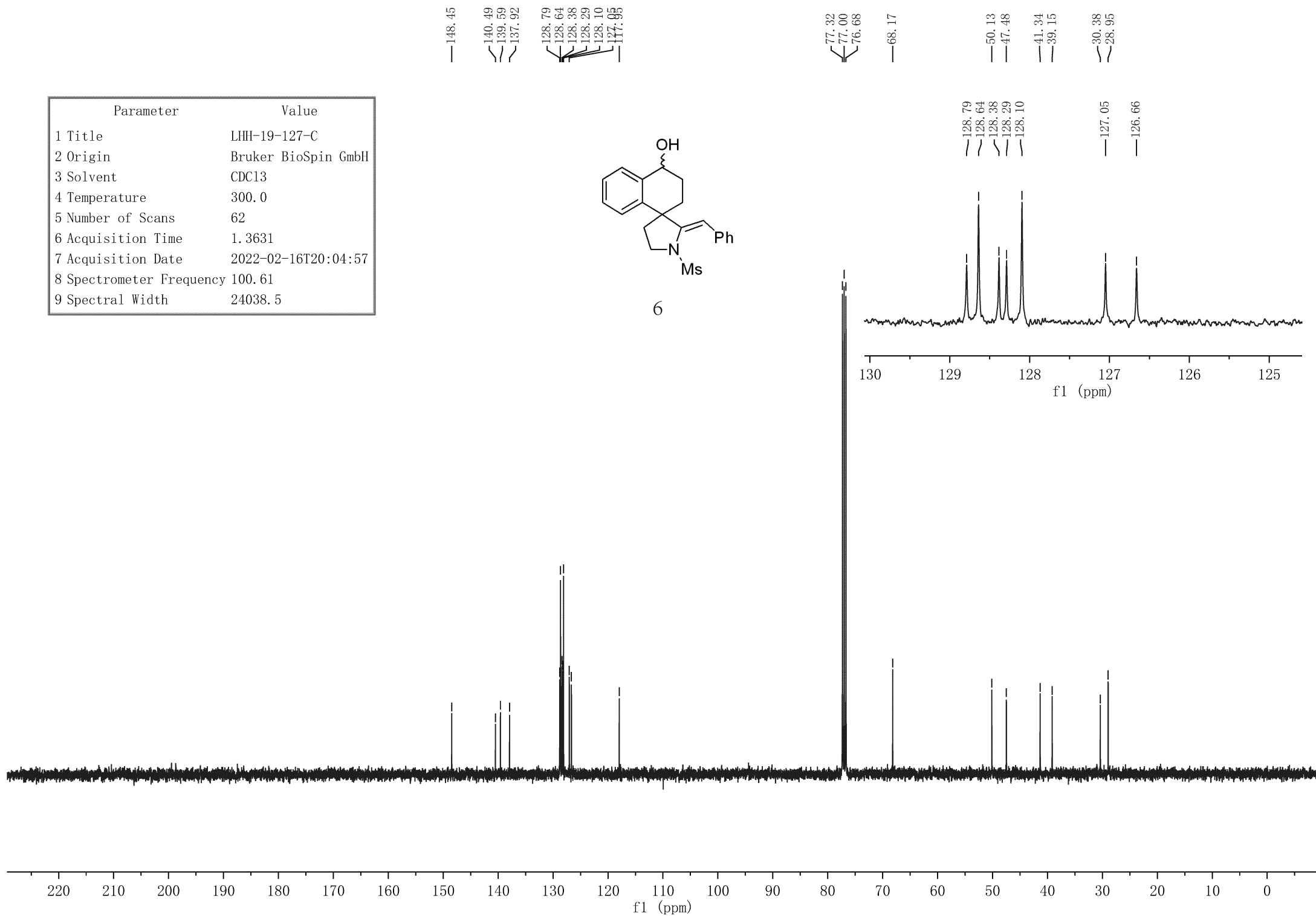
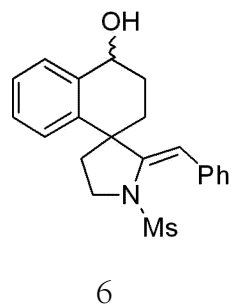
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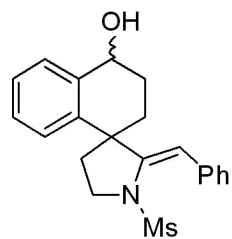
Parameter	Value
1 Title	LHH-19-127
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	298.0
5 Number of Scans	6
6 Acquisition Time	4.0894
7 Acquisition Date	2022-02-16T20:02:18
8 Spectrometer Frequency	400.13
9 Spectral Width	8012.8



Parameter	Value
1 Title	LHH-19-127-C
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	62
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-16T20:04:57
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



Parameter	Value
1 Title	LHH-19-127-C135
2 Origin	Bruker BioSpin GmbH
3 Solvent	CDC13
4 Temperature	300.0
5 Number of Scans	18
6 Acquisition Time	1.3631
7 Acquisition Date	2022-02-16T20:09:36
8 Spectrometer Frequency	100.61
9 Spectral Width	24038.5



6

