

Supporting Information

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Data of crystallography

Table 1. Data of crystal structure determination.

	3	4	5	7	8	9b	11b	13	14	16
Emp. formula	C ₂₄ H ₅₂ GeSi ₄	C ₃₂ H ₆₅ Br ₂ LiO ₂ Si ₄ Sn · 0.5 (C ₆ H ₁₄)	2 (C ₃₂ H ₆₅ Br ₂ LiO ₂ PbSi ₄), C ₅ H ₁₂	C ₂₄ H ₅₂ Si ₄ Sn	C ₆₀ H ₁₂₆ Ge ₂ Li ₂ O ₃ Si ₈	C ₄₀ H ₈₂ GeKO ₂ Si ₄	C ₃₆ H ₇₅ KO ₆ Si ₄ Sn	C ₁₆ AlF ₃₆ O ₄ , C ₃₂ H ₆₇ O ₂ Si ₄ Sn	C ₄₈ H ₁₀₂ Si ₈ Sn ₂ , C ₆ H ₆	C ₄₉ H ₆₈ GeN ₂
<i>M</i> [g/mol]	525.63	922.73	2008.43	571.73	1279.44	899.10	874.13	1682.07	1219.54	757.66
λ [Å]	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073
<i>T</i> [K]	100(2)	100(2)	100(2)	130(2)	100(2)	117(2)	100(2)	100(2)	170(2)	100(2)
crystal system	triclinic	triclinic	triclinic	monoclinic	triclinic	monoclinic	orthorhombic	triclinic	triclinic	triclinic
space group	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1	<i>P</i> 2 ₁ / <i>n</i>	<i>P</i> -1	<i>P</i> 2 ₁ / <i>c</i>	<i>P</i> <i>n</i> a 2 ₁	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1
<i>Z</i>	2	2	1	4	4	4	4	2	1	2
<i>a</i> [Å]	10.8467(4)	9.4333(3)	9.40780(10)	16.6242(4)	14.0474(4)	15.8808(5)	25.4342(3)	12.5225(4)	9.1345(2)	12.3798(4)
<i>b</i> [Å]	12.2159(4)	12.6264(4)	12.5125(2)	9.1360(2)	22.5485(6)	14.7774(5)	9.56500(10)	16.1355(5)	12.1875(3)	13.1930(4)
<i>c</i> [Å]	13.8073(4)	20.6027(6)	20.8396(3)	21.6779(5)	27.1022(7)	23.6977(8)	19.6290(3)	18.0799(5)	17.1264(3)	14.5014(5)
α [°]	66.376(2)	89.2150(10)	89.4300(10)	90	113.6040(10)	90	90	103.153(2)	108.1370(10)	76.764(2)
β [°]	75.966(2)	89.8210(10)	88.5550(10)	101.7770(10)	95.4590(10)	107.598(2)	90	100.644(2)	95.7350(10)	74.374(2)
γ [°]	76.691(2)	71.5940(10)	72.1210(10)	90	95.6420(10)	90	90	93.328(2)	104.1100(10)	86.728(2)
<i>V</i> [Å ³]	1607.83(10)	2328.20(13)	2333.92(6)	3223.10(13)	7742.6(4)	5301.0(3)	4775.31(10)	3476.95(19)	1724.96(7)	2220.31(13)
<i>D_c</i> [g/cm ³]	1.086	1.316	1.429	1.178	1.098	1.127	1.216	1.607	1.174	1.133
μ [mm ⁻¹]	1.111	2.394	5.454	0.950	0.936	0.785	0.759	0.588	0.892	0.723
F(000)	568	954	1010	1208	2768	1940	1856	1692	644	816
crystal size [mm]	0.220x0.170x0.140	0.210x0.180x0.170	0.312x0.267x0.178	0.230x0.210x0.190	0.220x0.200x0.170	0.220x0.200x0.160	0.190x0.170x0.160	0.180x0.170x0.150	0.220x0.210x0.180	0.440x0.331x0.188
θ range [°]	1.633 – 27.156	2.966 – 27.266	2.561 – 30.541	2.819 – 27.366	2.224 – 31.494	1.803 – 27.490	1.601 – 29.611	1.664 – 24.640	2.912 – 28.152	1.923 – 27.979
limiting indices	-13 ≤ <i>h</i> ≤ 13 -15 ≤ <i>k</i> ≤ 15 -17 ≤ <i>l</i> ≤ 17	-12 ≤ <i>h</i> ≤ 11 -16 ≤ <i>k</i> ≤ 16 -26 ≤ <i>l</i> ≤ 26	-13 ≤ <i>h</i> ≤ 13 -17 ≤ <i>k</i> ≤ 17 -29 ≤ <i>l</i> ≤ 29	-21 ≤ <i>h</i> ≤ 21 -11 ≤ <i>k</i> ≤ 11 -27 ≤ <i>l</i> ≤ 27	-20 ≤ <i>h</i> ≤ 20 -33 ≤ <i>k</i> ≤ 33 -39 ≤ <i>l</i> ≤ 38	-20 ≤ <i>h</i> ≤ 19 -19 ≤ <i>k</i> ≤ 19 -30 ≤ <i>l</i> ≤ 29	-33 ≤ <i>h</i> ≤ 35 -13 ≤ <i>k</i> ≤ 13 -26 ≤ <i>l</i> ≤ 27	-14 ≤ <i>h</i> ≤ 14 -18 ≤ <i>k</i> ≤ 18 -21 ≤ <i>l</i> ≤ 20	-12 ≤ <i>h</i> ≤ 12 -16 ≤ <i>k</i> ≤ 15 -22 ≤ <i>l</i> ≤ 22	-16 ≤ <i>h</i> ≤ 16 -17 ≤ <i>k</i> ≤ 17 -19 ≤ <i>l</i> ≤ 19
refl. coll.	27919	62977	80930	53525	291213	131244	72216	59241	35182	61443
ind. refl.	6995	10379	14169	7226	50738	11810	12871	11672	8352	10590
<i>R</i> _{int}	0.0323	0.0215	0.0432	0.0364	0.0288	0.0234	0.0489	0.0424	0.0215	0.0263
completeness	99.1	99.4	99.7	99.8	99.7	99.9	99.8	99.4	99.7	99.6
abs. correction	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan	multi-scan
max., min. trans.	0.7455, 0.6784	0.7455, 0.6413	0.7461, 0.5708	0.7456, 0.6442	0.7462, 0.6981	0.7456, 0.6921	0.7459, 0.7145	0.7451, 0.6928	0.7456, 0.6932	0.7456, 0.6618
Para./restr.	289/0	438/184	445/68	279/0	1466/0	549/0	455/1	888/2	312/0	489/0
<i>R</i> ₁ , <i>wR</i> ₂ [<i>I</i> > 2 σ (<i>I</i>)]	0.0358, 0.0908	0.0298, 0.0761	0.0365, 0.0879	0.0402, 0.0816	0.0331, 0.0740	0.0300, 0.0747	0.0266, 0.0505	0.0546, 0.1339	0.0219, 0.0583	0.0288, 0.0667
<i>R</i> ₁ , <i>wR</i> ₂ (all data)	0.0527, 0.0981	0.0359, 0.0792	0.0445, 0.0912	0.0529, 0.0849	0.0533, 0.0812	0.0420, 0.0816	0.0348, 0.0533	0.0739, 0.1469	0.0243, 0.0600	0.0361, 0.0704
Goof on <i>F</i> ²	1.060	1.018	1.049	1.122	1.008	1.055	1.018	1.032	1.065	1.037
peak / hole [<i>e</i> · Å ⁻³]	0.642, -0.744	1.219, -1.381	2.728, -2.238	0.881, -0.812	1.232, -0.854	0.496, -0.331	0.430, -0.247	1.707, -1.020	0.717, -0.536	0.462, -0.440
CCDC	2153532	2153539	2153540	2153561	2153534	2153536	2153537	2153538	2153535	2153533

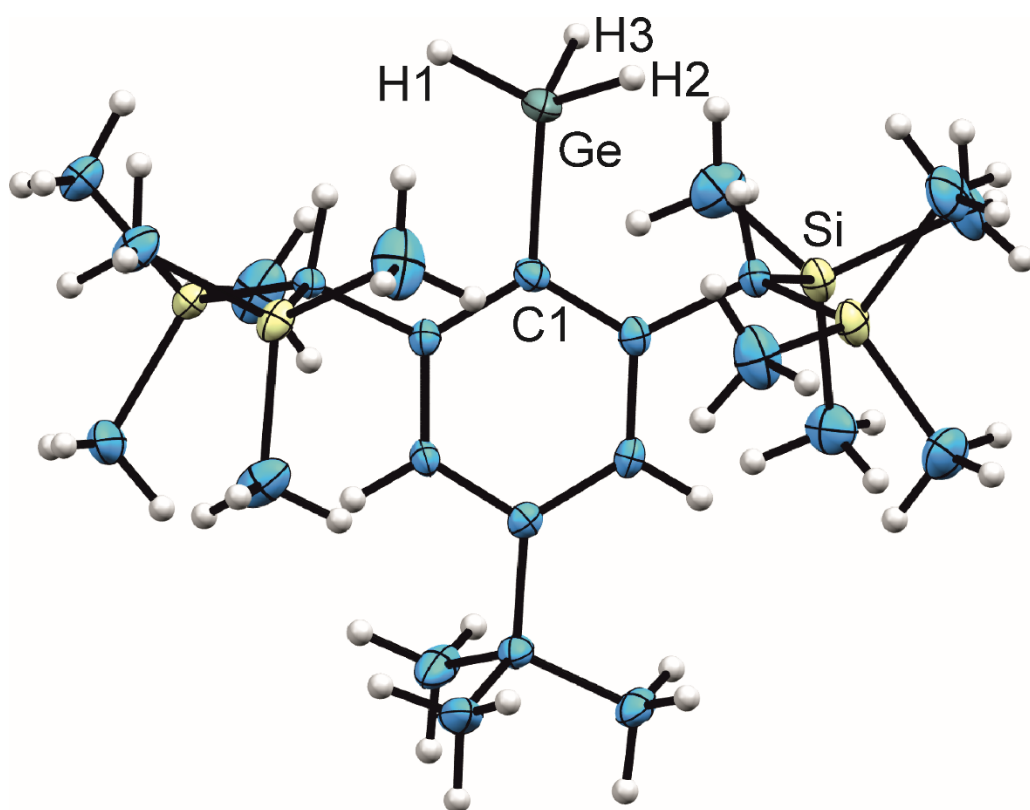


Figure S1. ORTEP of **3**. Hydrogen atoms connected to the germanium atom were found in the difference Fourier map. Other hydrogen atoms were placed in idealized positions. Ellipsoids at 50 % probability. Interatomic distance [Å]: Ge-C1 1.955(2).

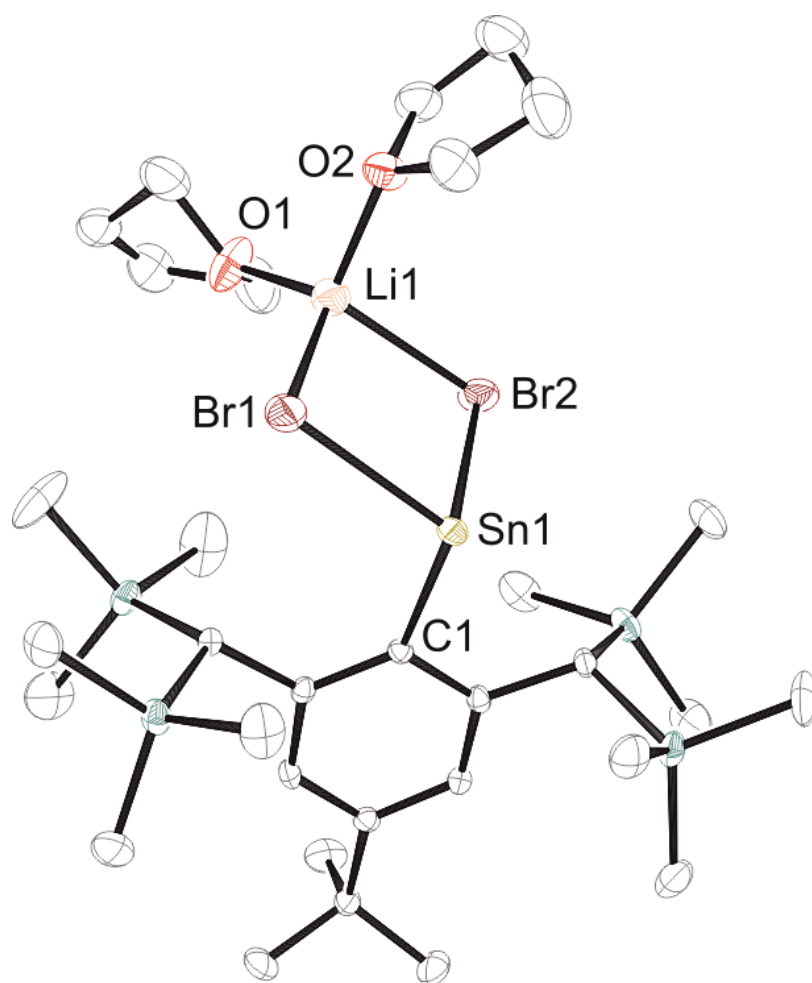


Figure S2. ORTEP of **4**. Hydrogen atoms, which are not shown, were placed in idealized positions. Ellipsoids at 50 % probability. Interatomic distance [Å], angles [°]: C1-Sn1 2.229(2), Br1-Sn1 2.7254(3), Br2-Sn1 2.7567(3), Li1-O1 1.874(5), Li1-O2 1.904(5), Li1-Br1 2.509(5), Li1-Br2 2.541(5), C1-Sn1-Br1 94.46(6), C1-Sn1-Br2 110.28(6), Br1-Sn1-Br2 87.48(1).

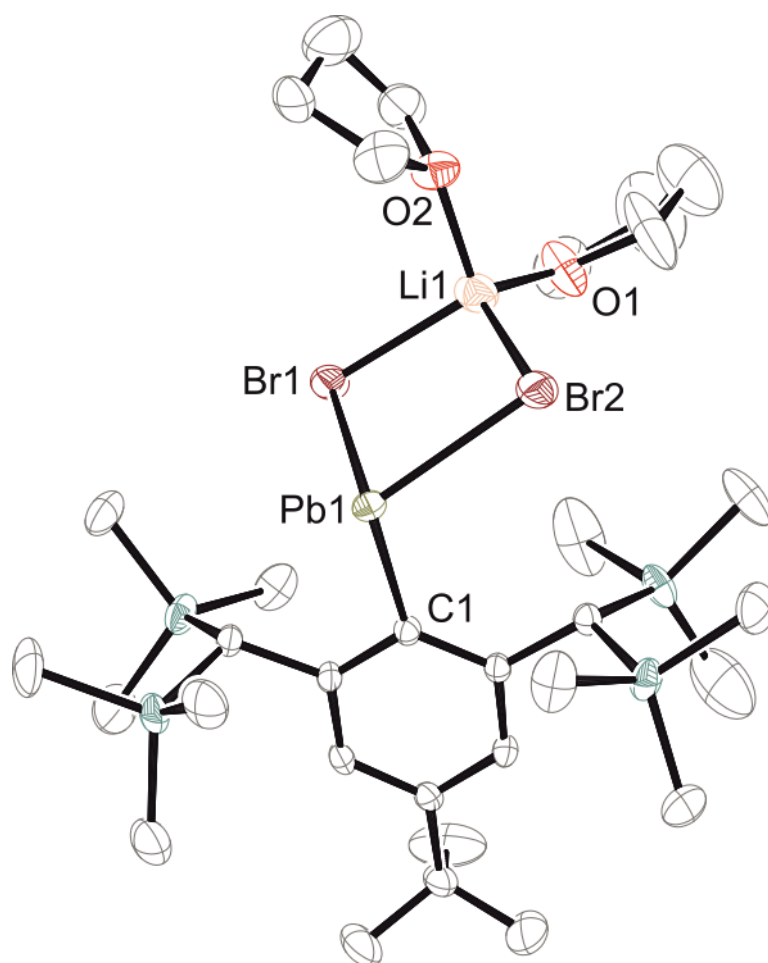


Figure S3. ORTEP of **5**. Hydrogen atoms, which are not shown, were placed in idealized positions. Ellipsoids at 50 % probability. Interatomic distance [Å], angles [°]: C1-Pb1 2.321(3), Pb1-Br1 2.8246(4), Pb1-Br2 2.8502(4), C1-Pb1-Br1 94.57(8), C1-Pb1-Br2 110.37(8), Br1-Pb1-Br2 86.74(1).

NMR spectra of compound 2

$^1\text{H-NMR}$ of TbbGeX_3 in C_6D_6 (#) at rt

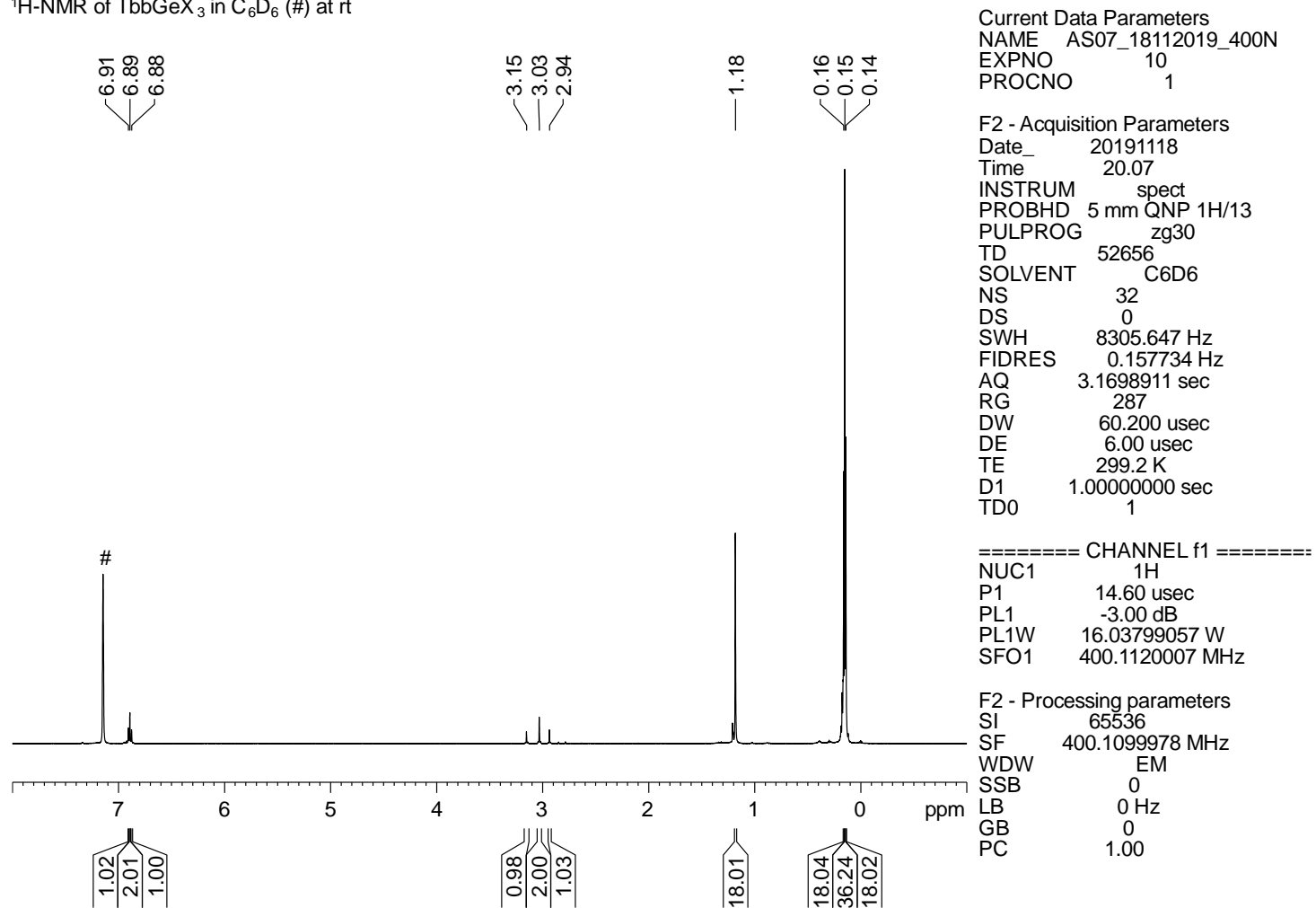
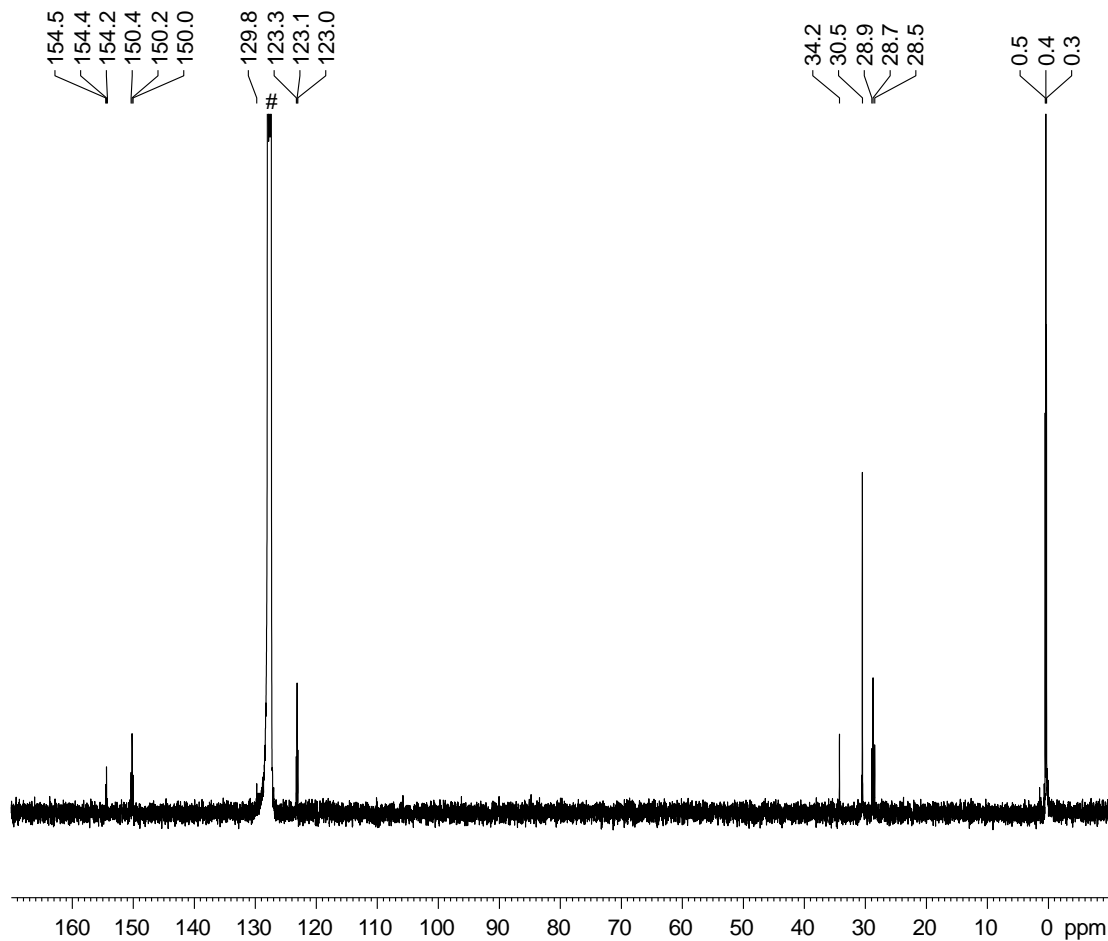


Figure S4. $^1\text{H-NMR}$ spectrum of compound 2.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of TbbGeX_3 in C_6D_6 (#) at rt



Current Data Parameters
NAME AS07_18112019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191119
Time 1.29
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 10000
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S5. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound 2.

²⁹Si-INEPTND-NMR of TbbGeX₃ in C₆D₆ at rt

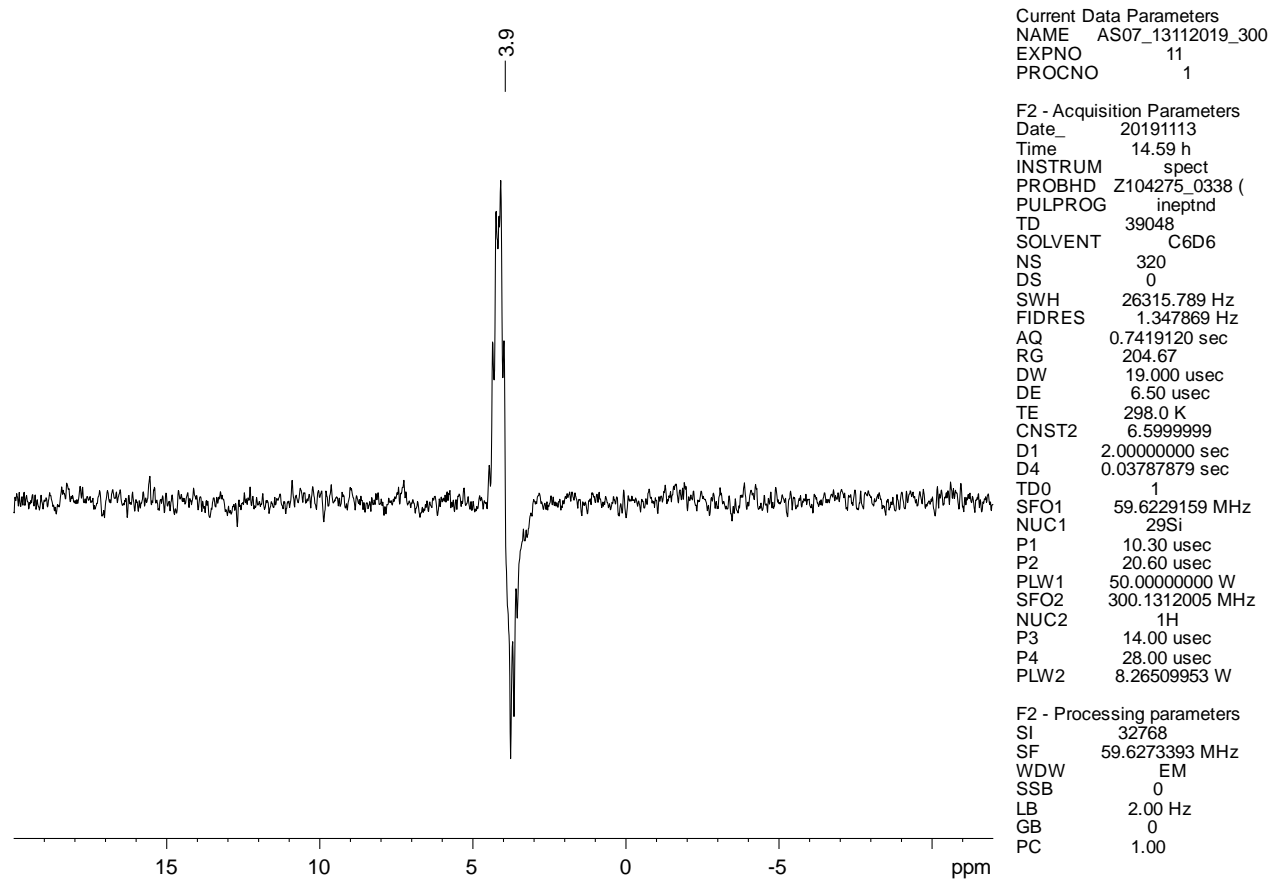
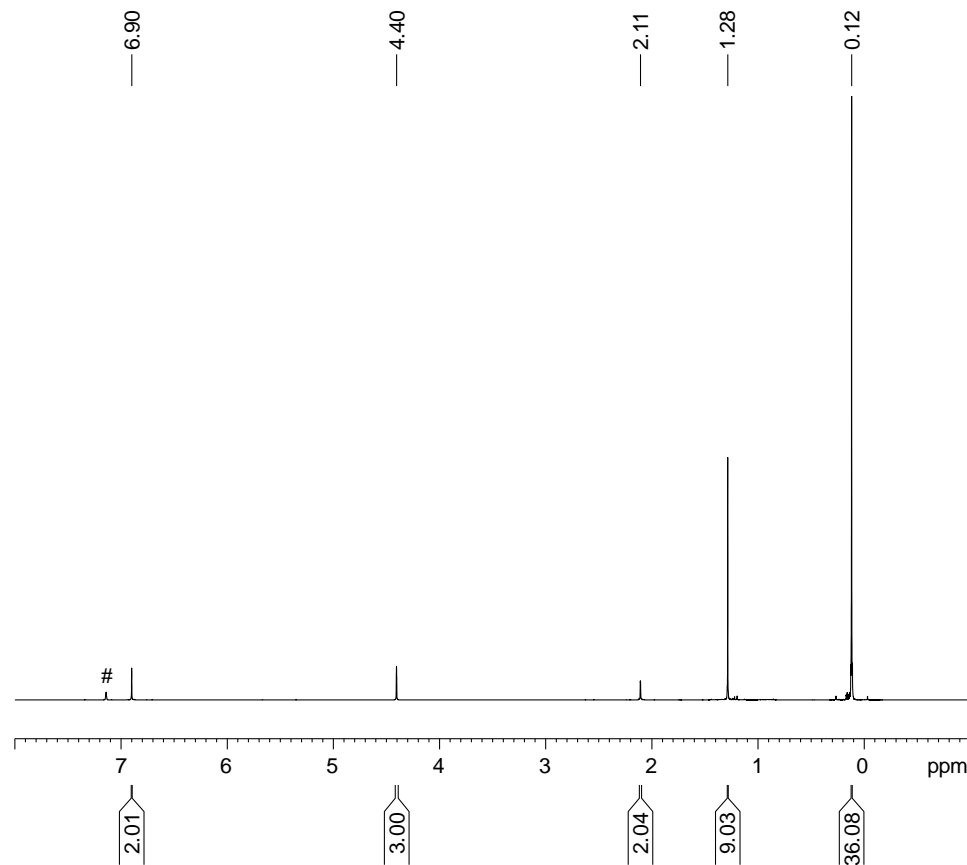


Figure S6. ²⁹Si-INEPTND-NMR spectrum of compound **2**.

NMR spectra of compound 3

¹H-NMR of TbbGeH₃ in C₆D₆ (#) at rt



Current Data Parameters
NAME MA179_13112019_400N
EXPNO 10
PROCNO 1

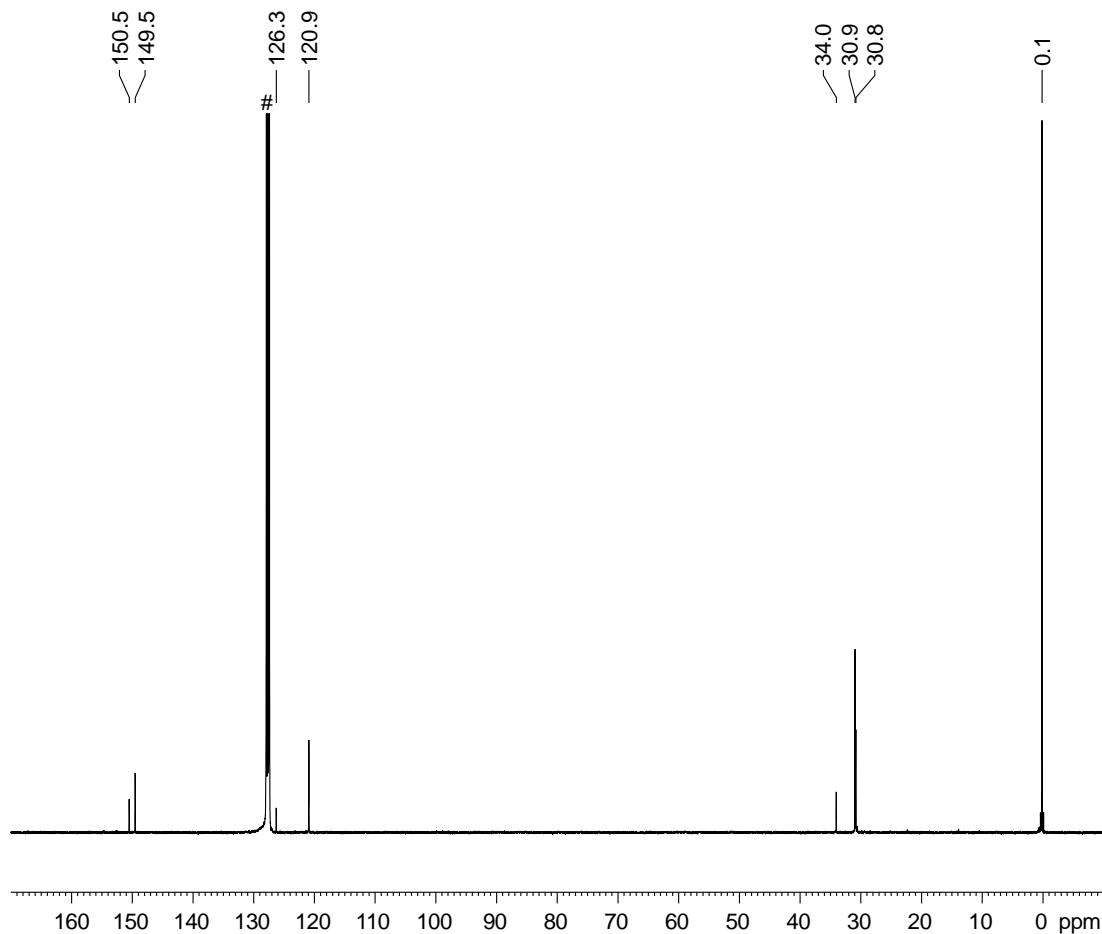
F2 - Acquisition Parameters
Date_ 20191113
Time 20.05
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zg30
TD 52656
SOLVENT C6D6
NS 32
DS 0
SWH 8305.647 Hz
FIDRES 0.157734 Hz
AQ 3.1698911 sec
RG 181
DW 60.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 1H
P1 14.60 usec
PL1 -3.00 dB
PL1W 16.03799057 W
SFO1 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 400.1100000 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

Figure S7. ¹H-NMR spectrum of compound 3.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of TbbGeH_3 in C_6D_6 (#) at rt



Current Data Parameters
NAME MA179_13112019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191114
Time_ 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.0000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====

NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

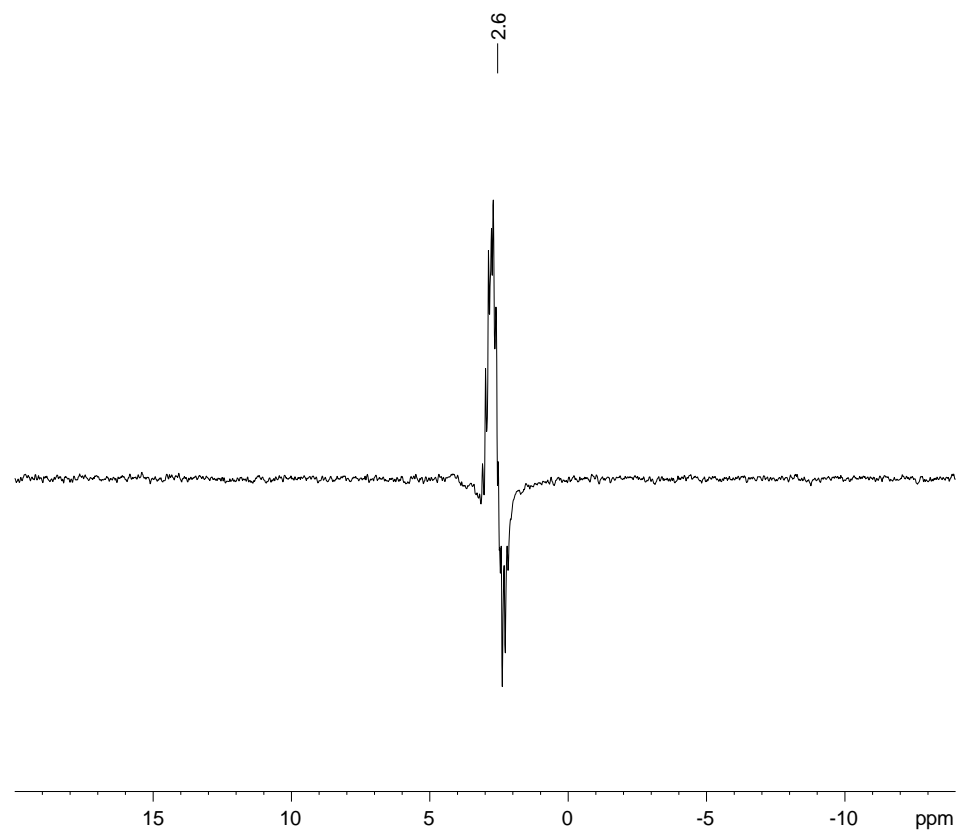
===== CHANNEL f2 =====

CPDPRG2 waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S8. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **3**.

^{29}Si -INEPTND-NMR of TbbGeH_3 in C_6D_6 at rt



Current Data Parameters
NAME MA179_13112019_300
EXPNO 11
PROCNO 1

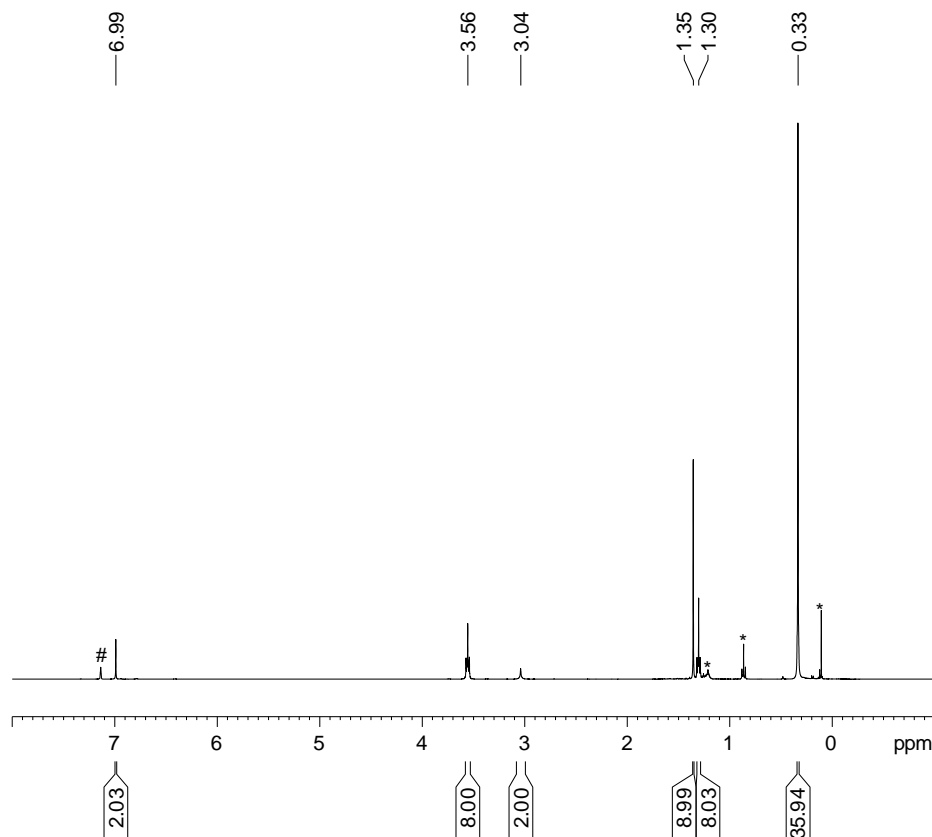
F2 - Acquisition Parameters
Date_ 20191113
Time 10.37 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 ^{29}Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273521 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S9. ^{29}Si -INEPTND-NMR spectrum of compound **3**.

NMR spectra of compound 4

¹H-NMR of ([TbbSnBr₂][Li{thf}₂]) in C₆D₆ (#) at rt, *: *n*-hexane and unknown impurity



Current Data Parameters
 NAME MA150K_18102019_400N
 EXPNO 10
 PROCNO 1

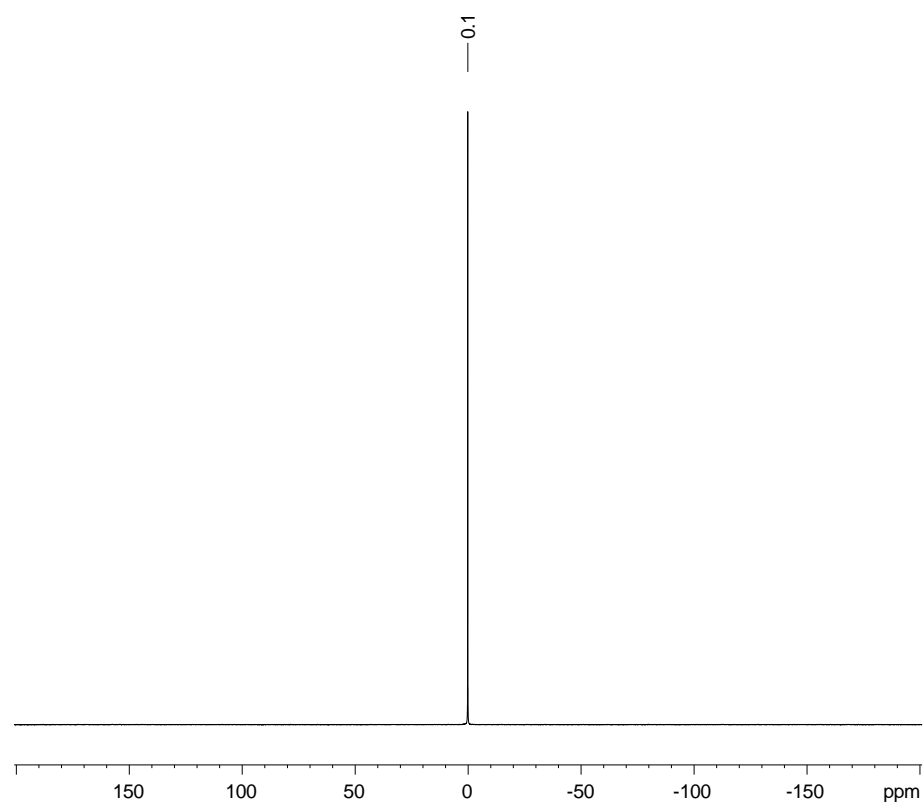
F2 - Acquisition Parameters
 Date_ 20191018
 Time 20.05
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 161
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100018 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S10. ¹H-NMR spectrum of compound 4.

^7Li -NMR of $([\text{TbbSnBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



Current Data Parameters
NAME MA150K_18102019_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191018
Time 10.49 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 32050
SOLVENT C6D6
NS 256
DS 0
SWH 46875.000 Hz
FIDRES 2.925117 Hz
AQ 0.3418667 sec
RG 204.67
DW 10.667 usec
DE 6.50 usec
TE 298.0 K
D1 0.20000000 sec
TD0 1
SFO1 116.6419139 MHz
NUC1 ^7Li
P1 8.81 usec
PLW1 34.00000000 W

F2 - Processing parameters
SI 32768
SF 116.6419139 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S11. ^7Li -NMR spectrum of compound **4**.

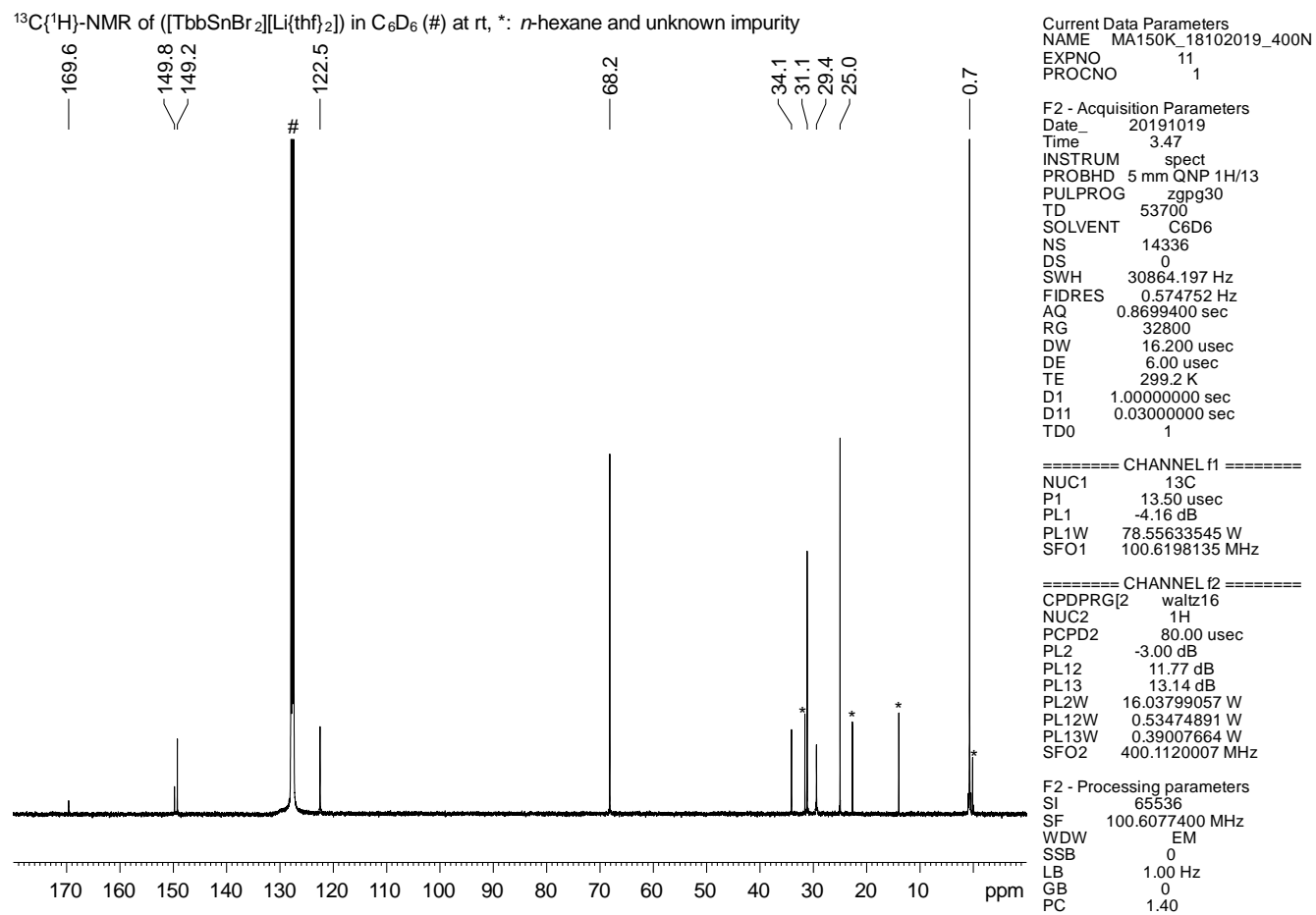
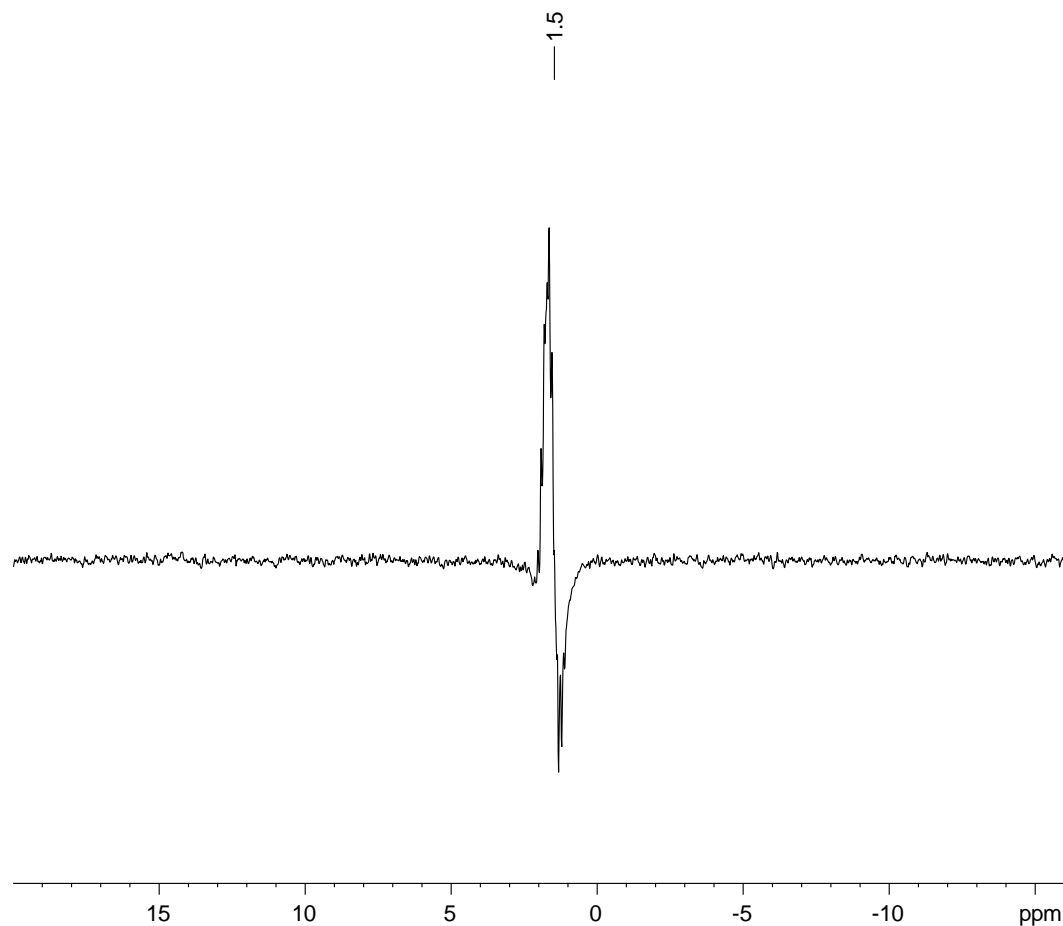


Figure S12. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **4**.

^{29}Si -INEPTND-NMR of $([\text{TbbSnBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



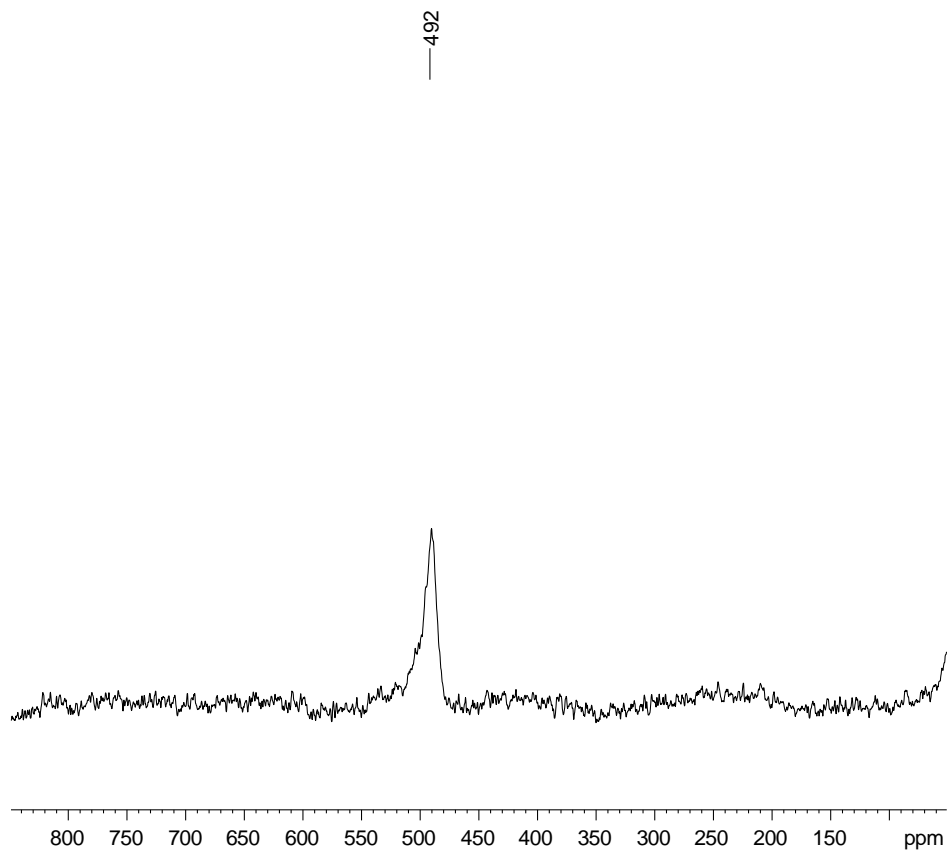
Current Data Parameters
NAME MA150_13112019_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
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Time 12.19 h
INSTRUM spect
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PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273839 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S13. ^{29}Si -INEPTND-NMR spectrum of compound 4.

^{119}Sn -NMR of $([\text{TbbSnBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



Current Data Parameters
NAME MA150_15102019_300
EXPNO 12
PROCNO 1

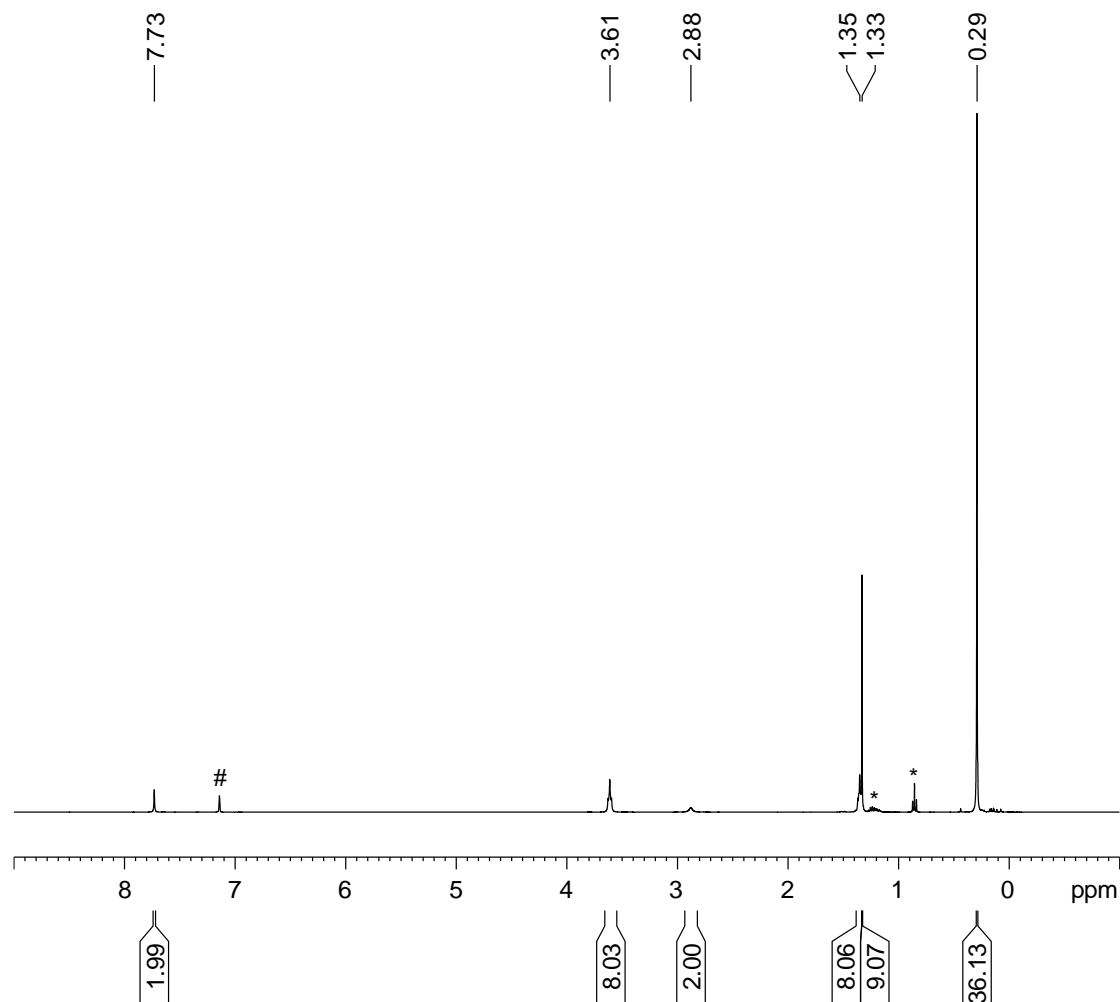
F2 - Acquisition Parameters
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Time 17.41 h
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PULPROG zg30
TD 4096
SOLVENT C6D6
NS 20480
DS 1
SWH 89285.711 Hz
FIDRES 43.596539 Hz
AQ 0.0229376 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.9707380 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 1.40

Figure S14. ^{119}Sn -NMR spectrum of compound **4**.

NMR spectra of compound 5

$^1\text{H-NMR}$ of $([\text{TbbPbBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 (#) at rt, *: *n*-pentane



Current Data Parameters
 NAME MA445K_22122020_400N
 EXPNO 10
 PROCNO 1

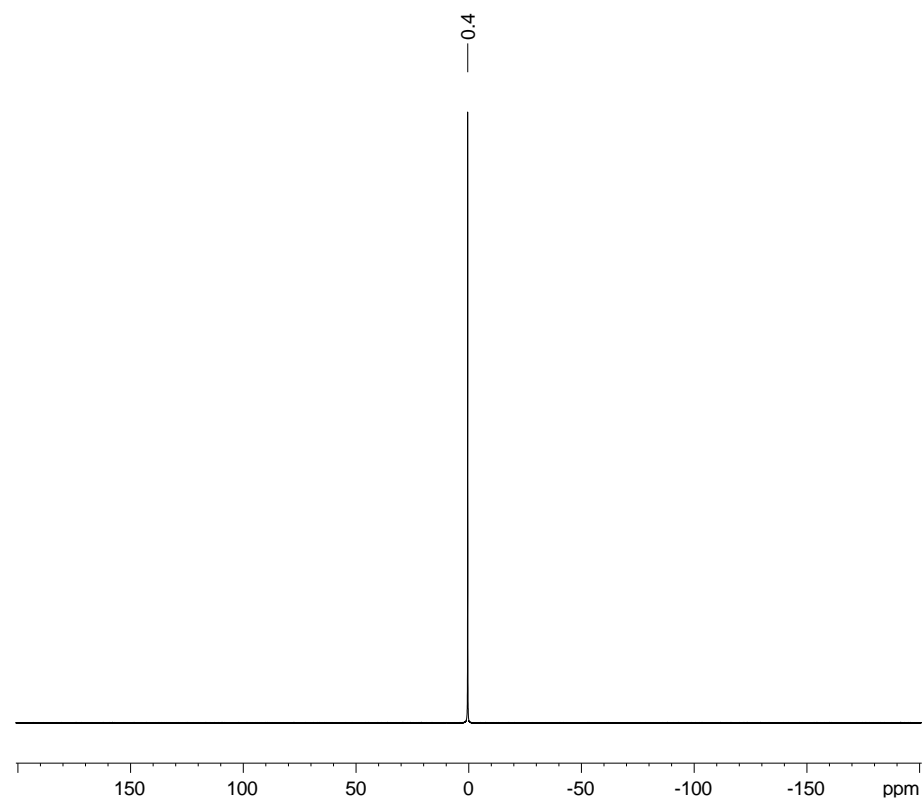
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 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 161
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
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 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S15. $^1\text{H-NMR}$ spectrum of compound 5.

^7Li -NMR of $([\text{TbbPbBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



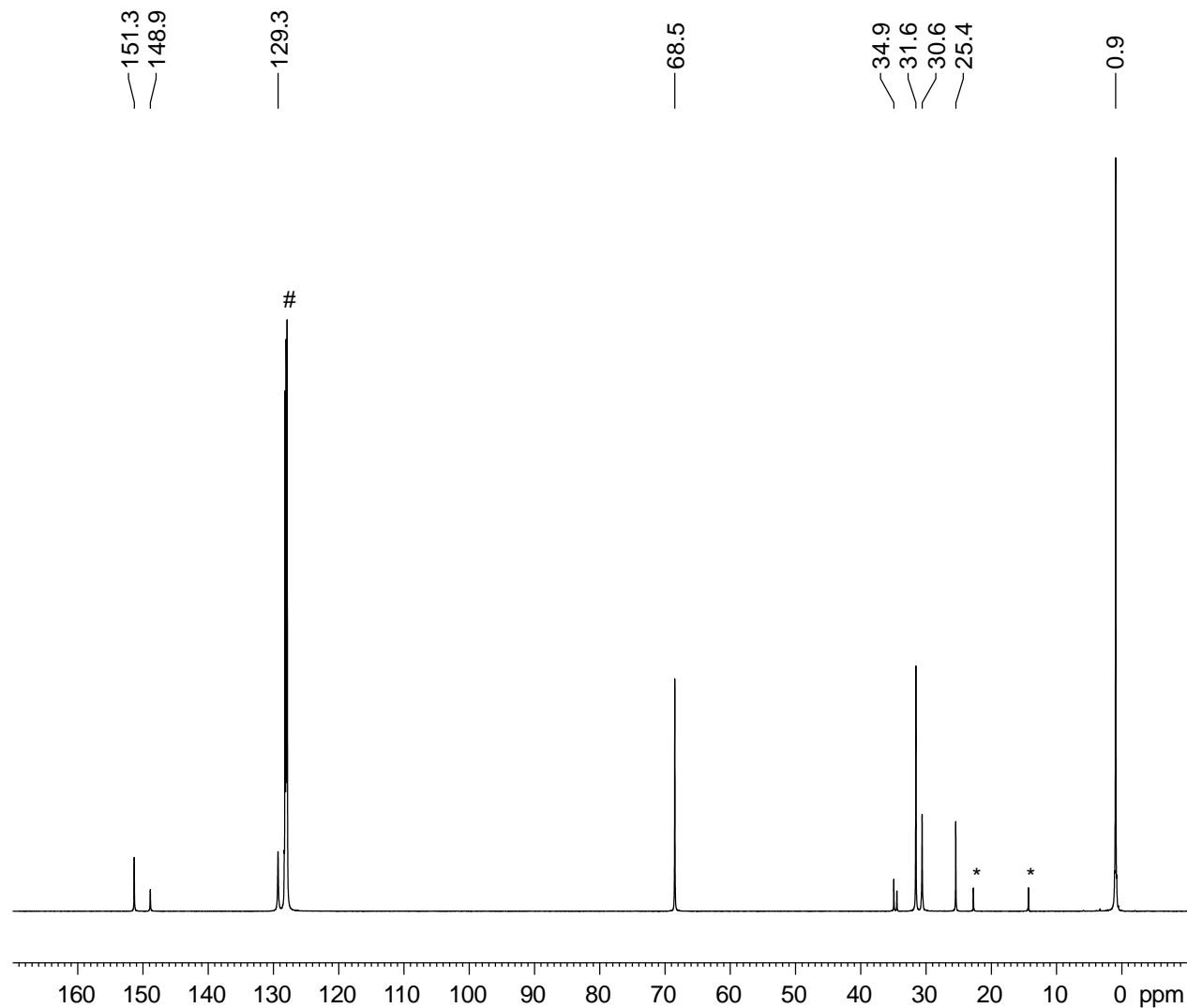
Current Data Parameters
NAME MA445K_25112020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20201125
Time 9.55 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 32050
SOLVENT C6D6
NS 256
DS 0
SWH 46875.000 Hz
FIDRES 2.925117 Hz
AQ 0.3418667 sec
RG 204.67
DW 10.667 usec
DE 6.50 usec
TE 298.0 K
D1 0.20000000 sec
TD0 1
SFO1 116.6419139 MHz
NUC1 ^7Li
P0 2.94 usec
P1 8.81 usec
PLW1 34.00000000 W

F2 - Processing parameters
SI 32768
SF 116.6419139 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S16. ^7Li -NMR spectrum of compound 5.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $([\text{TbbPbBr}_2][\text{Li}\{\text{thf}\}_2])$ in C_6D_6 (#) at rt, *: *n*-pentane



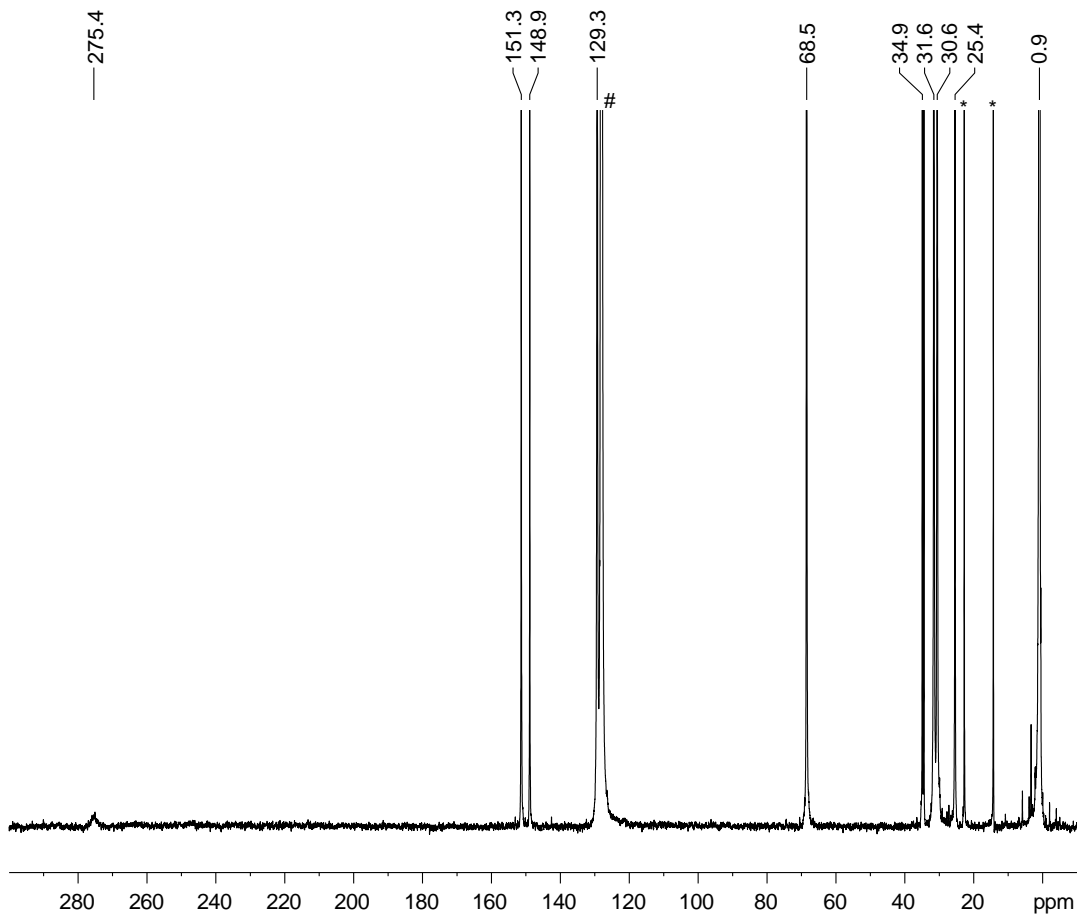
Current Data Parameters
NAME MA714K_30092021_60
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20210930
Time 20.06 h
INSTRUM spect
PROBHD Z126545_0027 (
PULPROG udef
TD 32768
SOLVENT C6D6
NS 4096
DS 0
SWH 53571.430 Hz
FIDRES 3.269741 Hz
AQ 0.3058347 sec
RG 189.6
DW 9.333 usec
DE 18.00 usec
TE 298.0 K
D1 4.00000000 sec
D12 0.00002000 sec
D20 20.00000000 sec
TD0 1
SFO1 150.9277075 MHz
NUC1 13C
P1 10.00 usec
P13 2000.00 usec
P26 500.00 usec
PLW1 57.02700043 W
SPNAM[5] Crp60comp.4
SPOAL5 0.500
SPOFFS5 0 Hz
SPW5 8.71310043 W
SPNAM[8] Crp60,0.5,20.1
SPOAL8 0.500
SPOFFS8 0 Hz
SPW8 8.71310043 W
SFO2 600.1324005 MHz
NUC2 1H
CPDPRG2 waltz16
PCPD2 70.00 usec
PLW2 23.41200066 W
PLW12 0.68803000 W

F2 - Processing parameters
SI 131072
SF 150.9027510 MHz
WDW EM
SSB 0
LB 7.00 Hz
GB 0
PC 1.40

Figure S17. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **5**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $([\text{TbbPbBr}_2][\text{Li}\{\text{thf}\}_2])$ in C_6D_6 (#) at rt, *: *n*-pentane



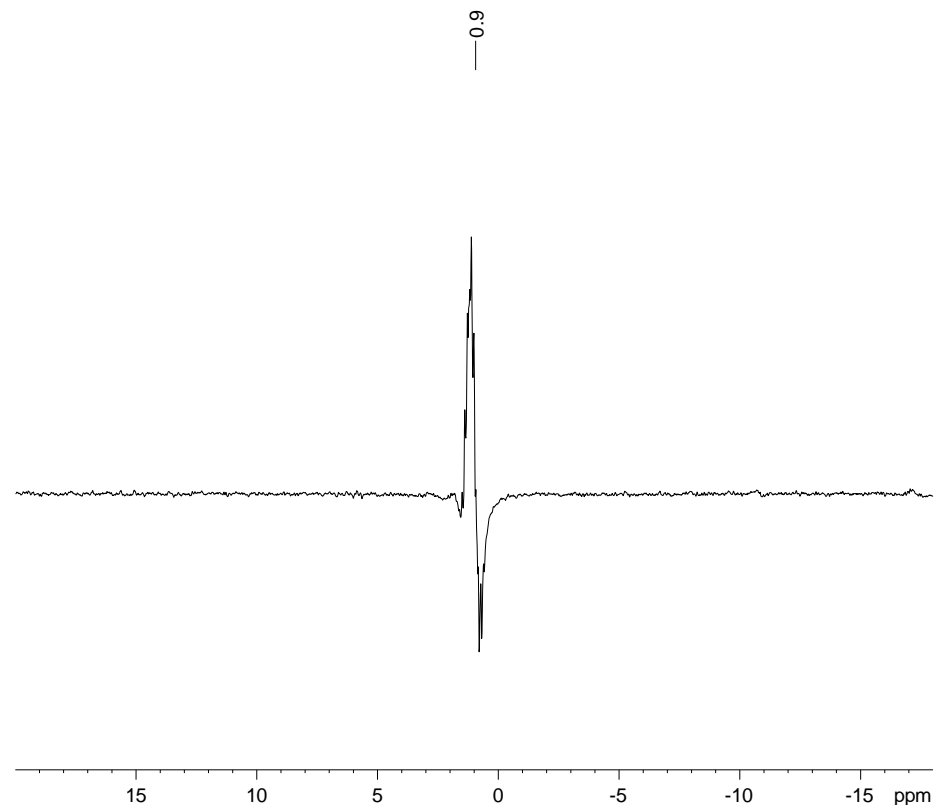
Current Data Parameters
NAME MA714K_30092021_600
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20210930
Time 20.06 h
INSTRUM spect
PROBHD Z126545_0027 (
PULPROG udef
TD 32768
SOLVENT C6D6
NS 4096
DS 0
SWH 53571.430 Hz
FIDRES 3.269741 Hz
AQ 0.3058347 sec
RG 189.6
DW 9.333 usec
DE 18.00 usec
TE 298.0 K
D1 4.00000000 sec
D12 0.00002000 sec
D20 20.00000000 sec
TD0 1
SFO1 150.9277075 MHz
NUC1 13C
P1 10.00 usec
P13 2000.00 usec
P26 500.00 usec
PLW1 57.02700043 W
SPNAM[5] Crp60comp.4
SPOAL5 0.500
SPOFFS5 0 Hz
SPW5 8.71310043 W
SPNAM[8] Crp60,0.5,20.1
SPOAL8 0.500
SPOFFS8 0 Hz
SPW8 8.71310043 W
SFO2 600.1324005 MHz
NUC2 1H
CPDPRG[2] waltz16
PCPD2 70.00 usec
PLW2 23.41200066 W
PLW12 0.68803000 W

F2 - Processing parameters
SI 131072
SF 150.9027510 MHz
WDW EM
SSB 0
LB 7.00 Hz
GB 0
PC 1.40

Figure S18. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound 5.

^{29}Si -INEPTND-NMR of $([\text{TbbPbBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



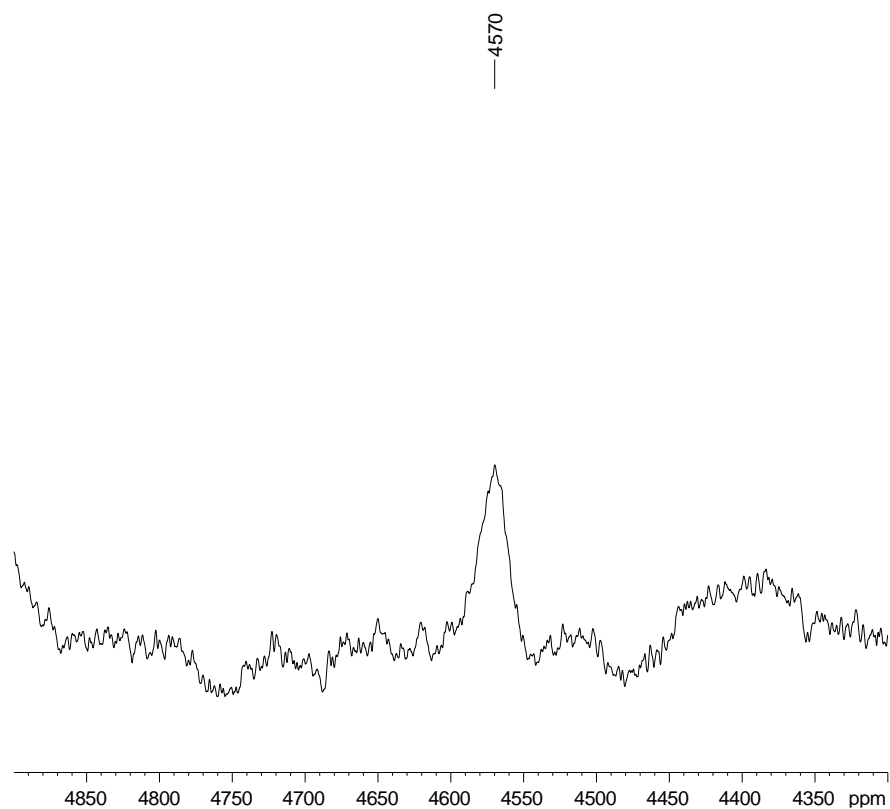
Current Data Parameters
NAME MA445K_24112020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20201124
Time 9.58 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.0000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273582 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S19. ^{29}Si -INEPTND-NMR spectrum of compound **5**.

^{207}Pb -NMR of $([\text{TbbPbBr}_2][\text{Li}(\text{thf})_2])$ in C_6D_6 at rt



Current Data Parameters
NAME MA445K_01032021_300N
EXPNO 18
PROCNO 1

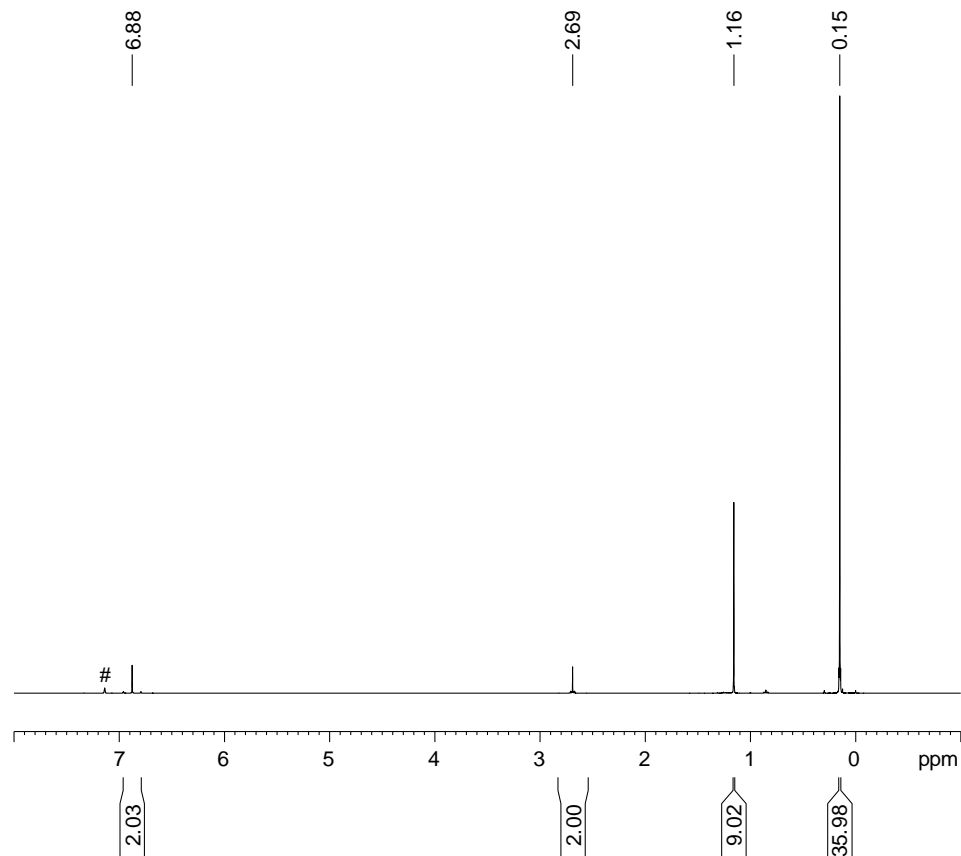
F2 - Acquisition Parameters
Date_ 20210301
Time 21.08 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 14998
SOLVENT C6D6
NS 128000
DS 0
SWH 125000.000 Hz
FIDRES 16.668890 Hz
AQ 0.0599920 sec
RG 204.67
DW 4.000 usec
DE 10.00 usec
TE 298.0 K
D1 0.20000000 sec
TD0 1
SFO1 63.0401498 MHz
NUC1 207Pb
P0 4.17 usec
P1 12.50 usec
PLW1 40.00000000 W

F2 - Processing parameters
SI 65536
SF 62.7889938 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 3.00

Figure S20. ^{207}Pb -NMR spectrum of compound **5**.

NMR spectra of compound 6

$^1\text{H-NMR}$ of TbSnBr_3 in C_6D_6 (#) at rt



Current Data Parameters
NAME MA164_31102019_400N
EXPNO 10
PROCNO 1

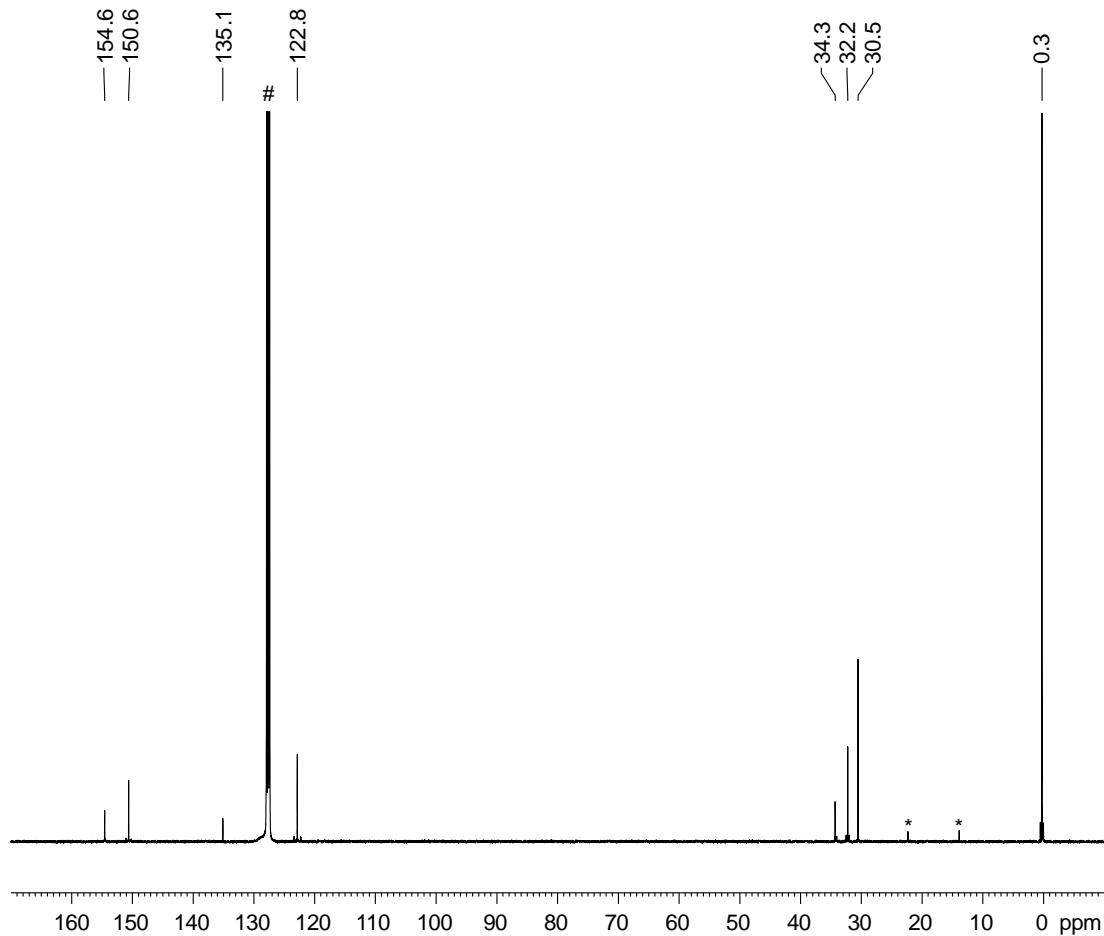
F2 - Acquisition Parameters
Date_ 20191031
Time 21.12
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zg30
TD 52656
SOLVENT C6D6
NS 32
DS 0
SWH 8305.647 Hz
FIDRES 0.157734 Hz
AQ 3.1698911 sec
RG 128
DW 60.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 1H
P1 14.60 usec
PL1 -3.00 dB
PL1W 16.03799057 W
SFO1 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 400.1100010 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

Figure S21. $^1\text{H-NMR}$ spectrum of compound 6.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of TbbSnBr_3 in C_6D_6 (#) at rt, *: *n*-pentane



Current Data Parameters
NAME MA164_31102019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191101
Time 2.10
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 9216
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

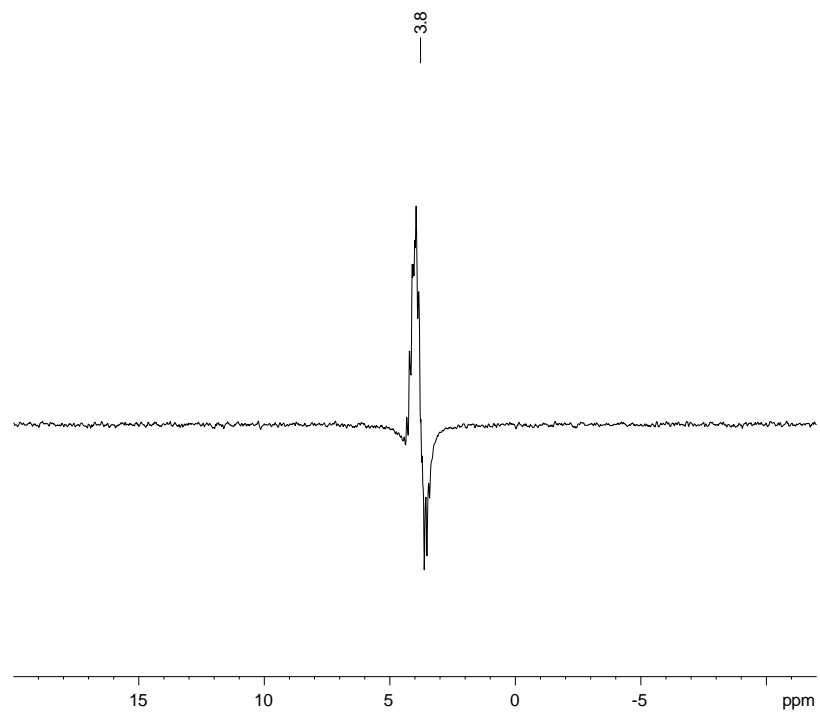
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077417 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S22. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **6**.

^{29}Si -INEPTND-NMR of TbSnBr_3 in C_6D_6 at rt



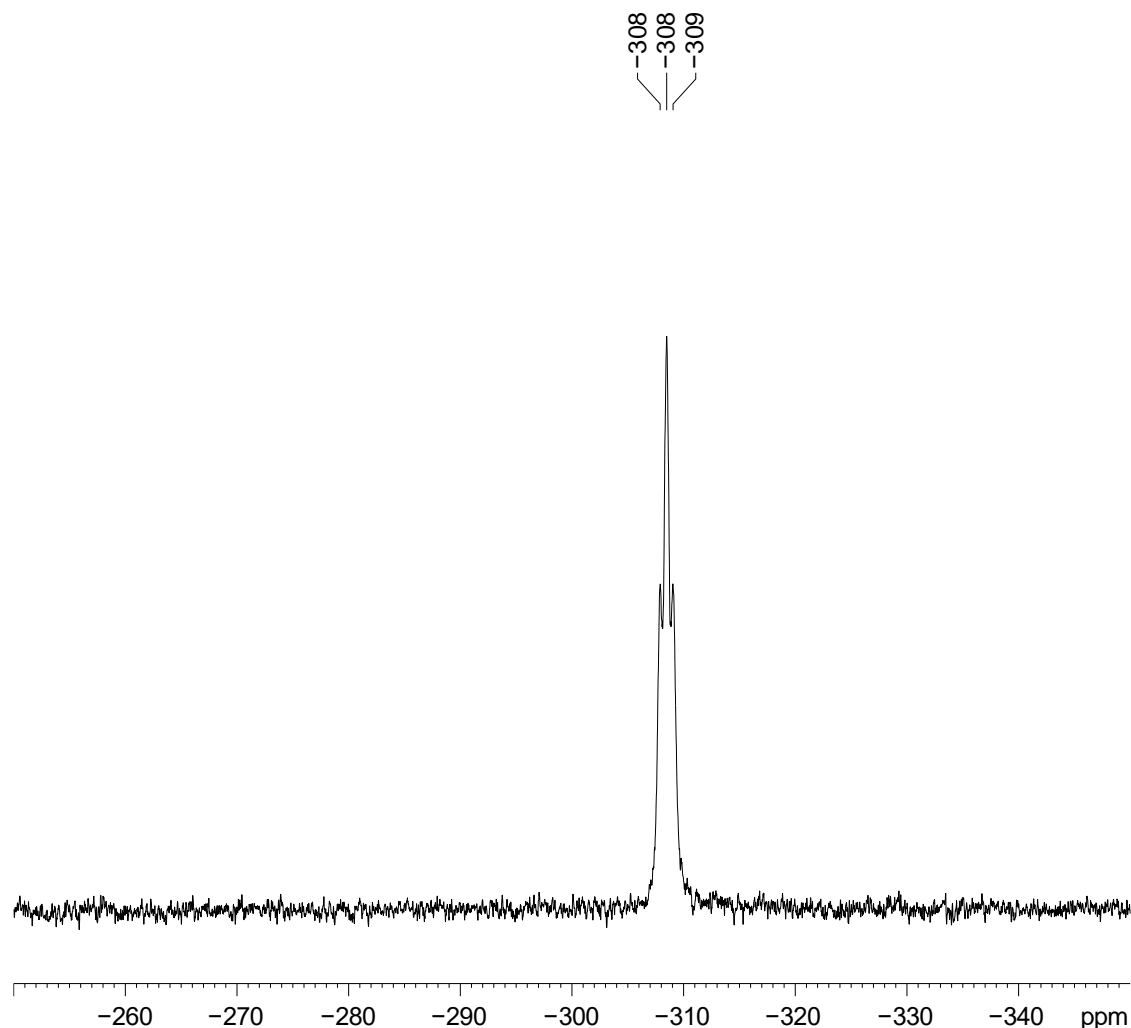
Current Data Parameters
NAME MA164_13112019_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191113
Time 11.35 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273498 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S23. ^{29}Si -INEPTND-NMR spectrum of compound **6**.

^{119}Sn -NMR of TbbSnBr_3 in C_6D_6 at rt



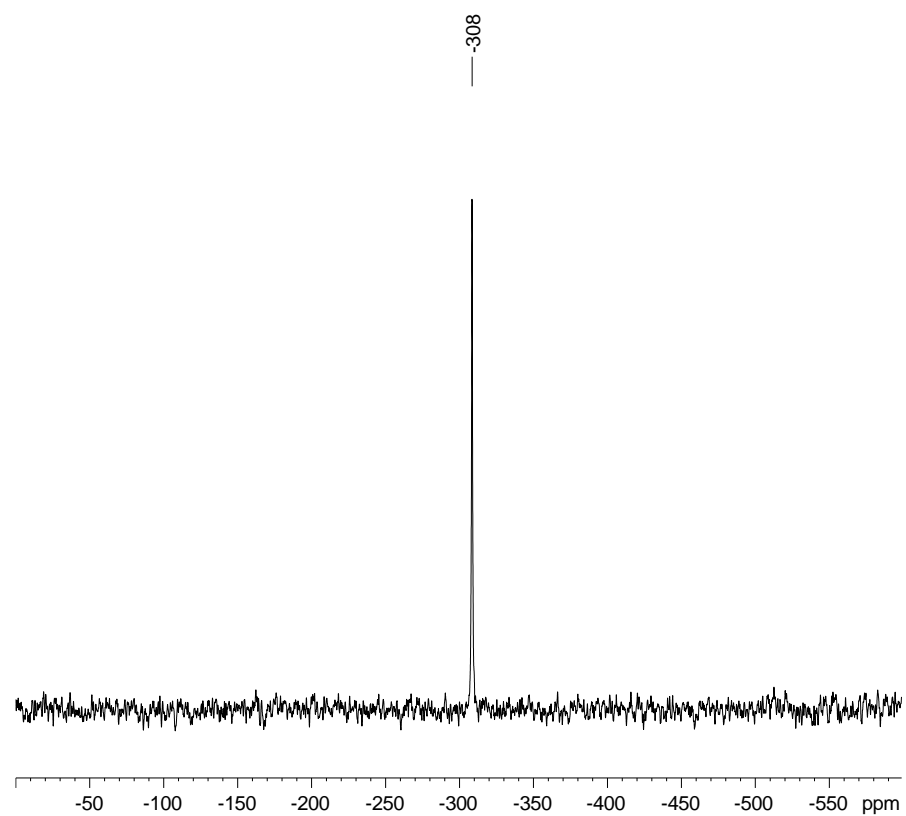
Current Data Parameters
NAME MA164_30102019_300N
EXPNO 17
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191030
Time 21.02 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 89276
SOLVENT C6D6
NS 73728
DS 1
SWH 89285.711 Hz
FIDRES 2.000218 Hz
AQ 0.4999456 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8979900 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 65536
SF 111.9203740 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S24. ^{119}Sn -NMR spectrum of compound **6**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of TbbSnBr_3 in C_6D_6 at rt



Current Data Parameters
NAME MA164K_25112019_300
EXPNO 11
PROCNO 1

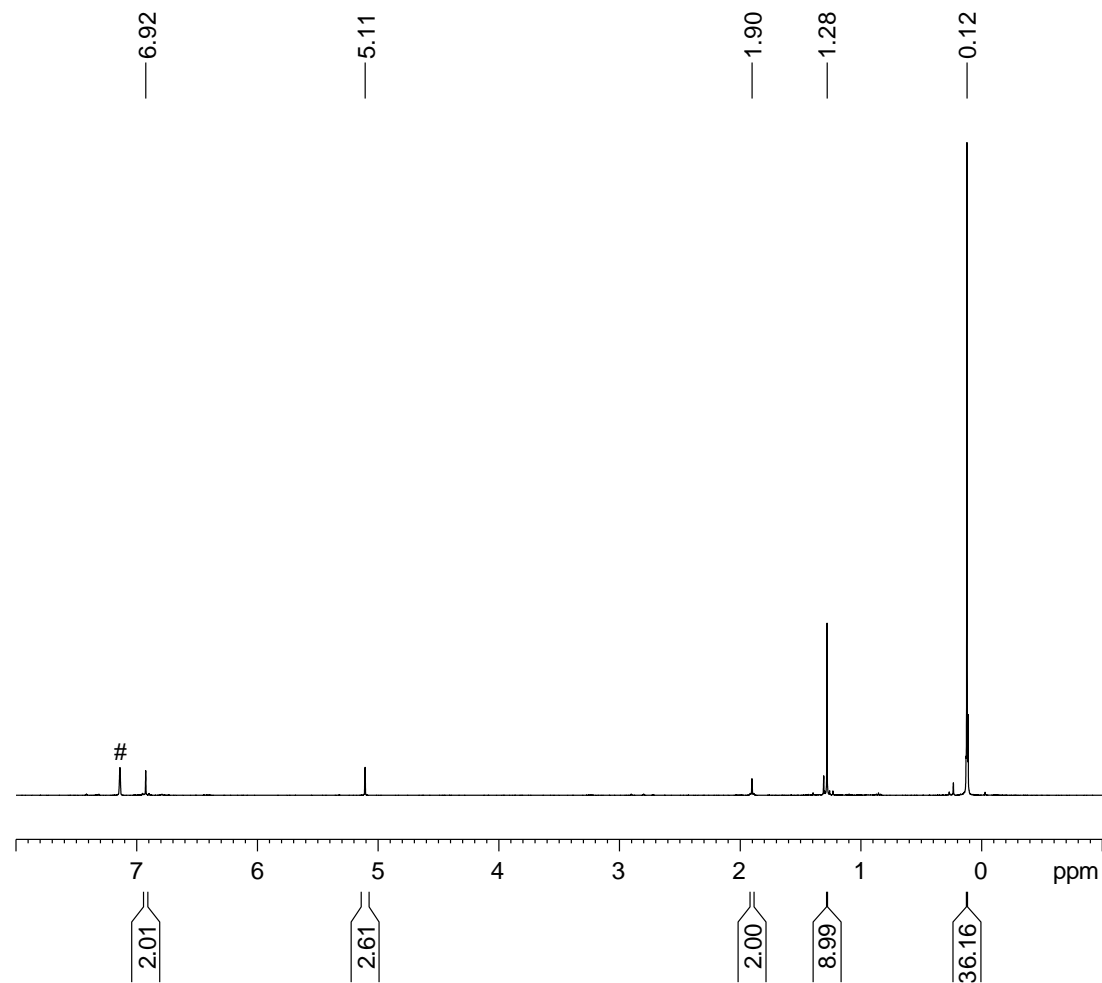
F2 - Acquisition Parameters
Date_ 20191125
Time 11.37 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT C6D6
NS 4900
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8979898 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 50.00 Hz
GB 0
PC 1.40

Figure S25. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **6**.

NMR spectra of compound 7

$^1\text{H-NMR}$ of TbbSnH_3 in C_6D_6 (#) at rt



Current Data Parameters
NAME MA232_10022020_400N
EXPNO 10
PROCNO 1

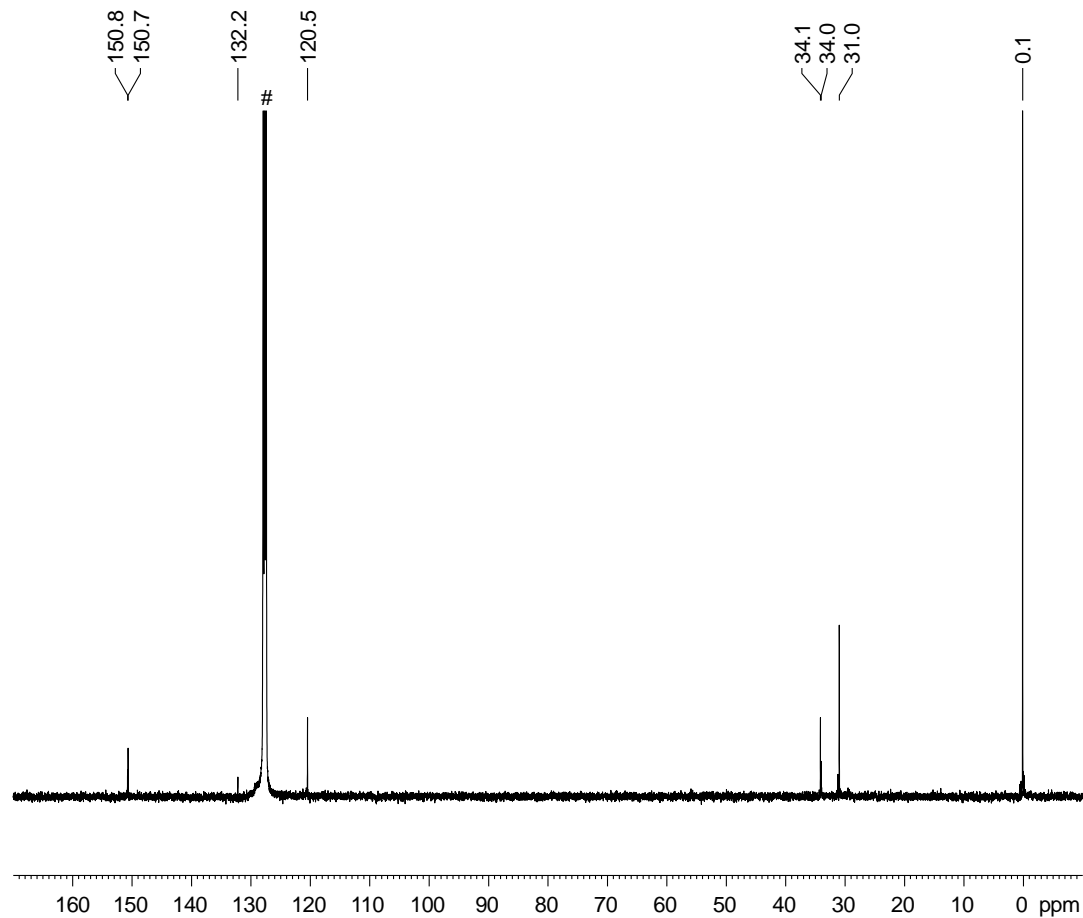
F2 - Acquisition Parameters
Date_ 20200210
Time_ 20.06
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zg30
TD 52656
SOLVENT C6D6
NS 32
DS 0
SWH 8305.647 Hz
FIDRES 0.157734 Hz
AQ 3.1698911 sec
RG 322
DW 60.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
TD0 1

==== CHANNEL f1 =====
NUC1 1H
P1 14.60 usec
PL1 -3.00 dB
PL1W 16.03799057 W
SFO1 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 400.1100000 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

Figure S26. ^1H NMR spectrum of compound 7.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $\text{Tb}(\text{SnH}_3)$ in C_6D_6 (#) at rt



Current Data Parameters
NAME MA232_10022020_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200211
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

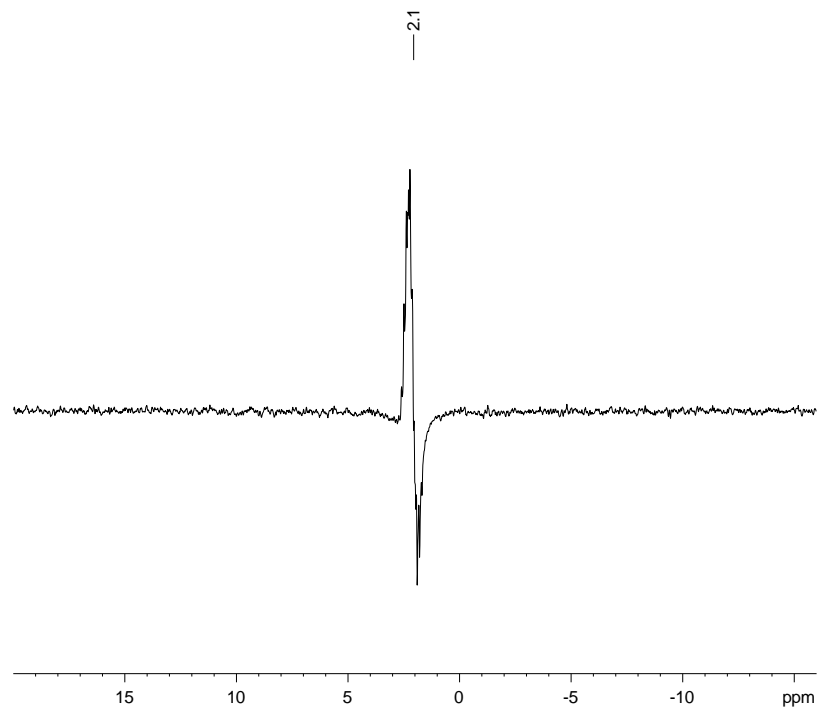
==== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

==== CHANNEL f2 =====
CPDPRG2 waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S27. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound 7.

^{29}Si -INEPTND-NMR of TbSnH_3 in toluene- d_8 at rt



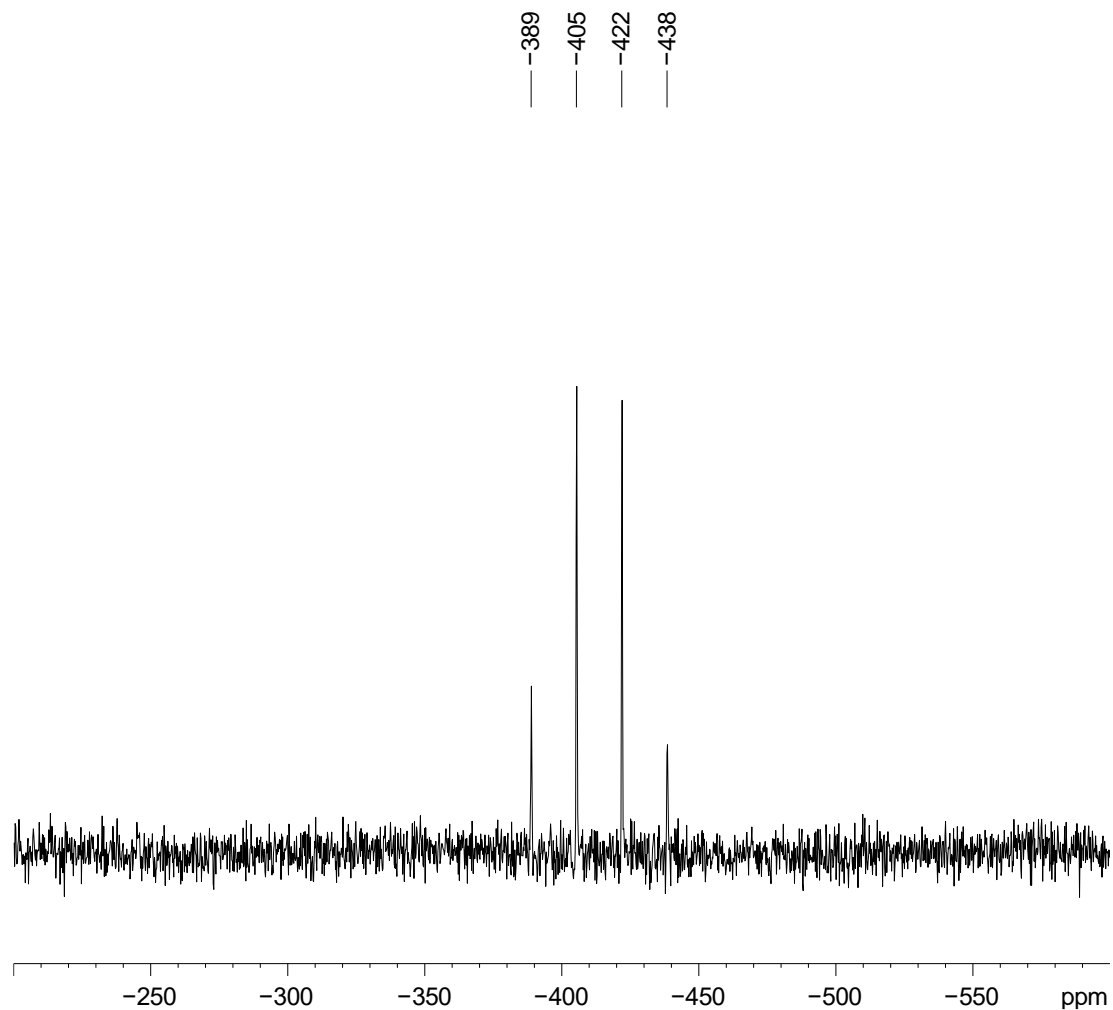
Current Data Parameters
NAME MA192_28112019_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191128
Time 15.20 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT Tol
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 ^{29}Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273880 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S28. ^{29}Si -INEPTND-NMR spectrum of compound **7**.

^{119}Sn -NMR of TbbSnH_3 in toluene- d_8 at rt



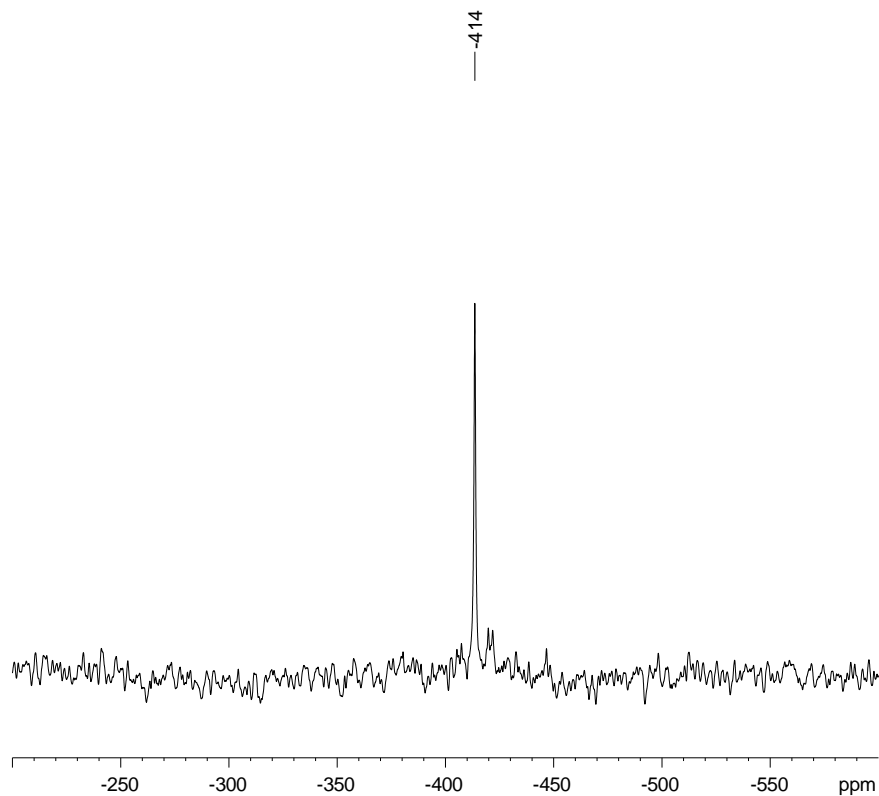
Current Data Parameters
NAME MA192_28112019_300
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191128
Time 15.55 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT Tol
NS 17408
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8867979 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.40

Figure S29. ^{119}Sn -NMR spectrum of compound **7**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of TbbSnH_3 in toluene- d_8 at rt



Current Data Parameters
NAME MA192_28112019_300
EXPNO 13
PROCNO 1

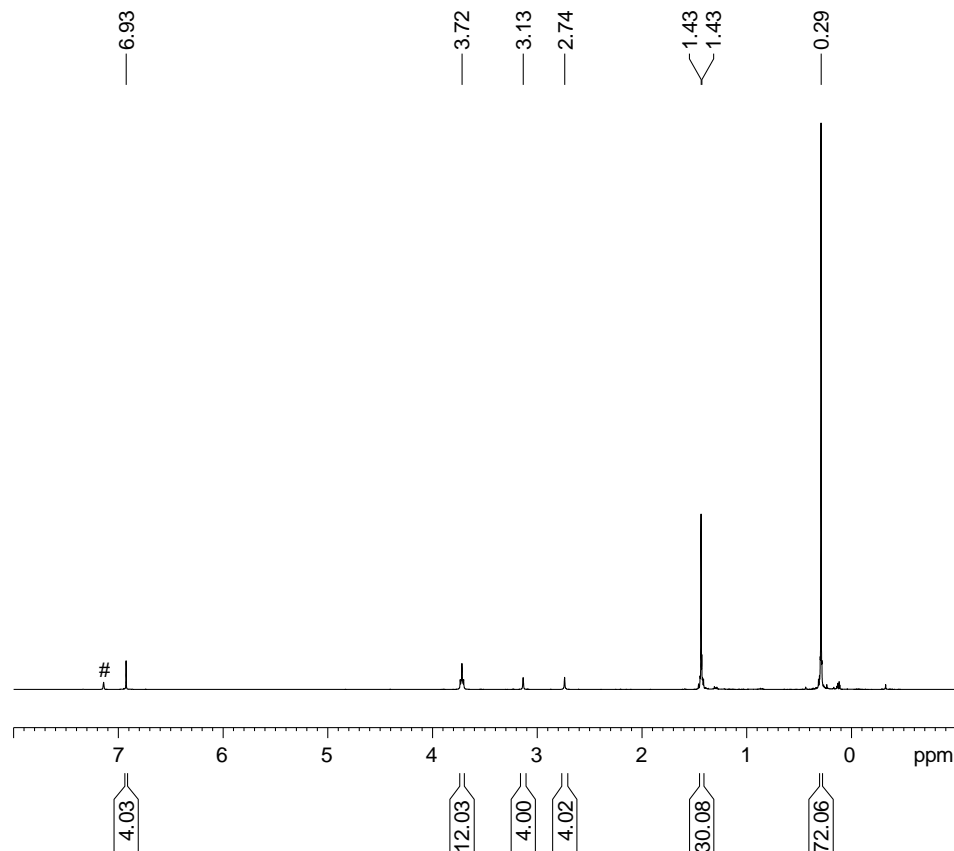
F2 - Acquisition Parameters
Date_ 20191128
Time 20.17 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT Tol
NS 5100
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8867977 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 50.00 Hz
GB 0
PC 1.40

Figure S30. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **7**.

NMR spectra of compound **8**

$^1\text{H-NMR}$ of $([\text{TbGeH}_2][\text{Li}(\text{thf})_{1.5}]_2)$ in C_6D_6 (#) at rt



Current Data Parameters
 NAME MA197_11122019_400N
 EXPNO 10
 PROCNO 1

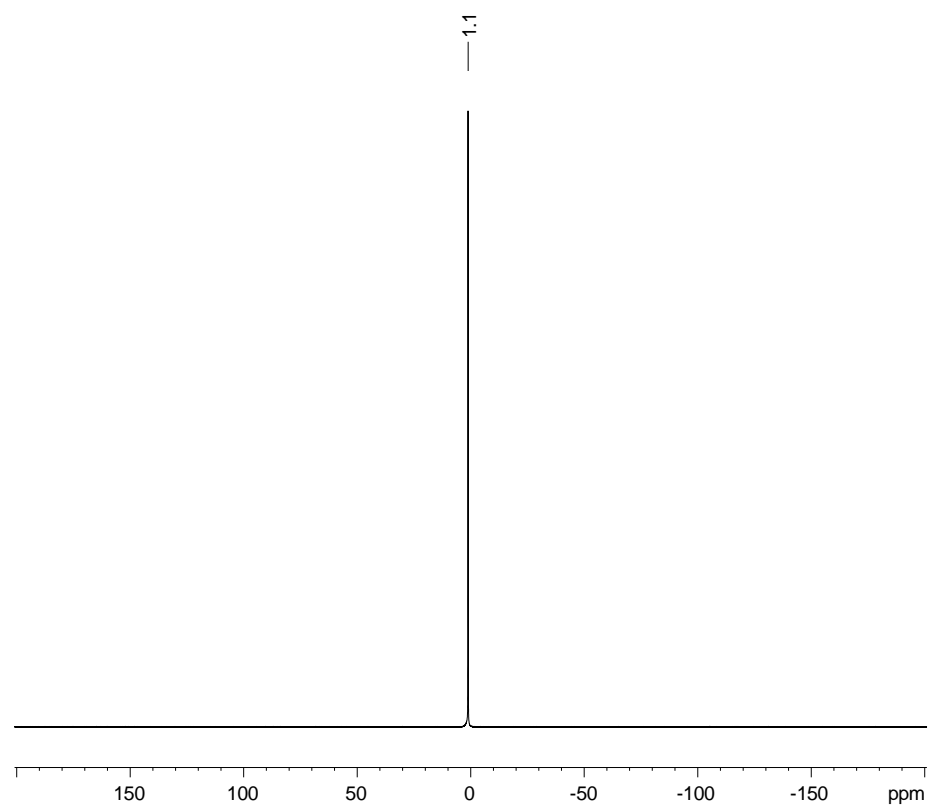
F2 - Acquisition Parameters
 Date_ 20191211
 Time 20.06
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 144
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S31. $^1\text{H-NMR}$ spectrum of compound **8**.

^7Li -NMR of $(\text{TbbGeH}_2)[\text{Li}(\text{thf})_{1.5}]_2$ in C_6D_6 at rt



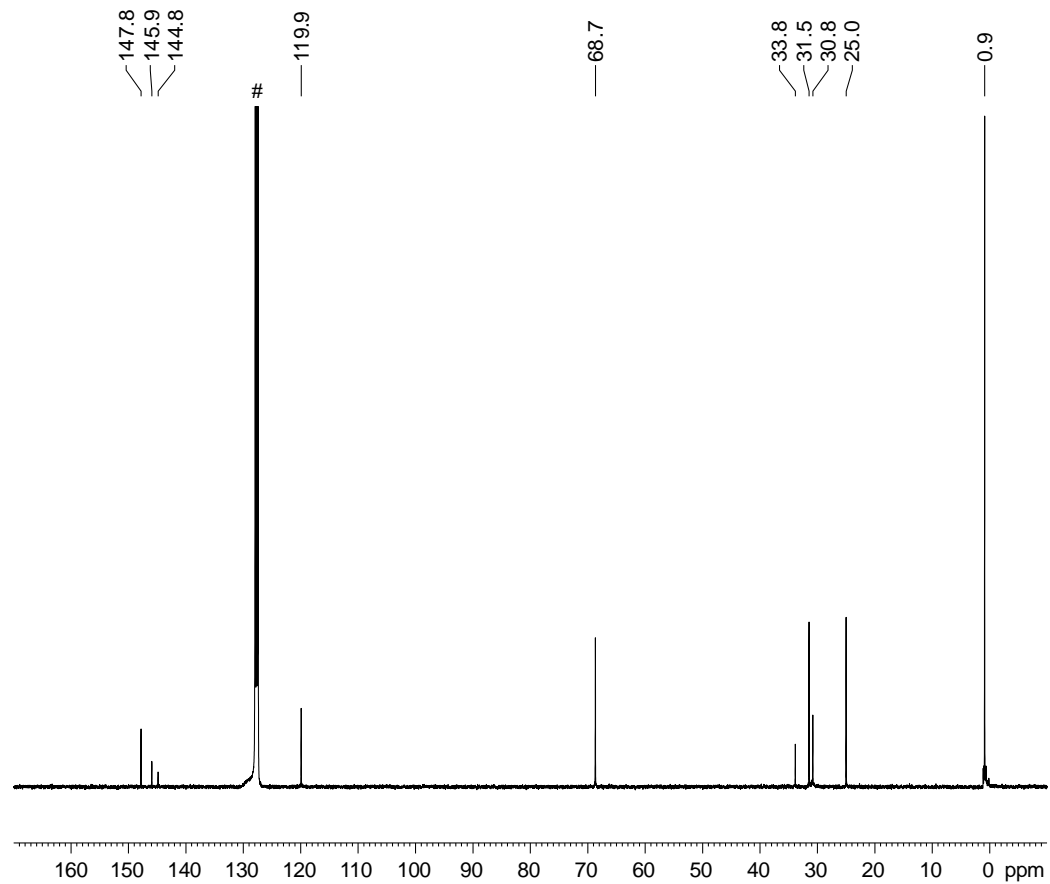
Current Data Parameters
NAME MA197K_11122019_300
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191211
Time_ 14.42 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 32050
SOLVENT C6D6
NS 256
DS 0
SWH 46875.000 Hz
FIDRES 2.925117 Hz
AQ 0.3418667 sec
RG 204.67
DW 10.667 usec
DE 6.50 usec
TE 298.0 K
D1 0.20000000 sec
TD0 1
SFO1 116.6419139 MHz
NUC1 ^7Li
P1 8.81 usec
PLW1 34.00000000 W

F2 - Processing parameters
SI 32768
SF 116.6419139 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S32. ^7Li -NMR spectrum of compound **8**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $[\text{TbbGeH}_2][\text{Li}(\text{thf})_{1.5}]_2$ in C_6D_6 (#) at rt



Current Data Parameters
NAME MA197_11122019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191212
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

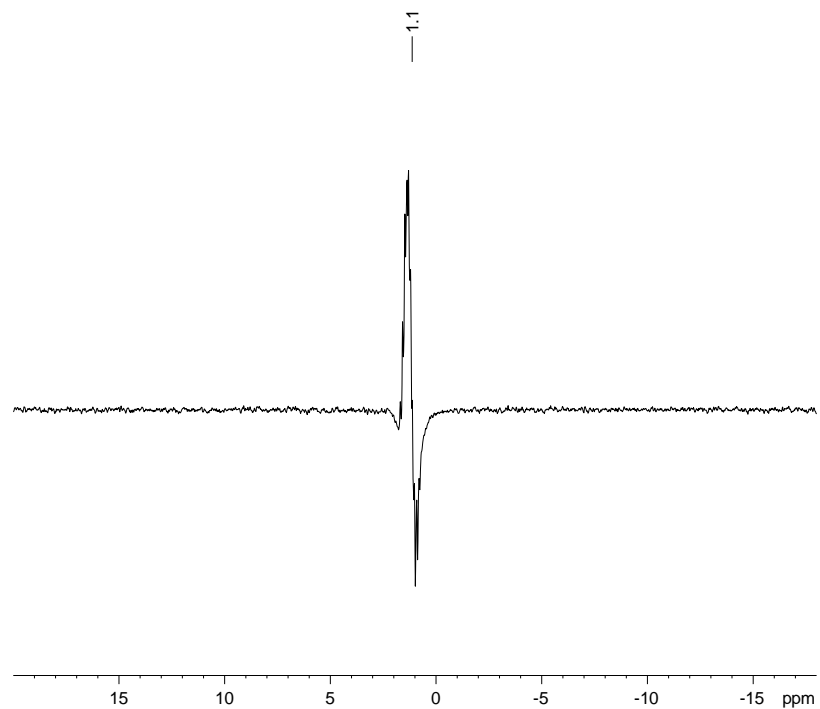
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S33. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **8**.

^{29}Si -INEPTND-NMR of $([\text{TbbGeH}_2[\text{Li}(\text{thf})_{1.5}]_2)_2$ in C_6D_6 at rt



Current Data Parameters
NAME MA197K_11122019_300
EXPNO 11
PROCNO 1

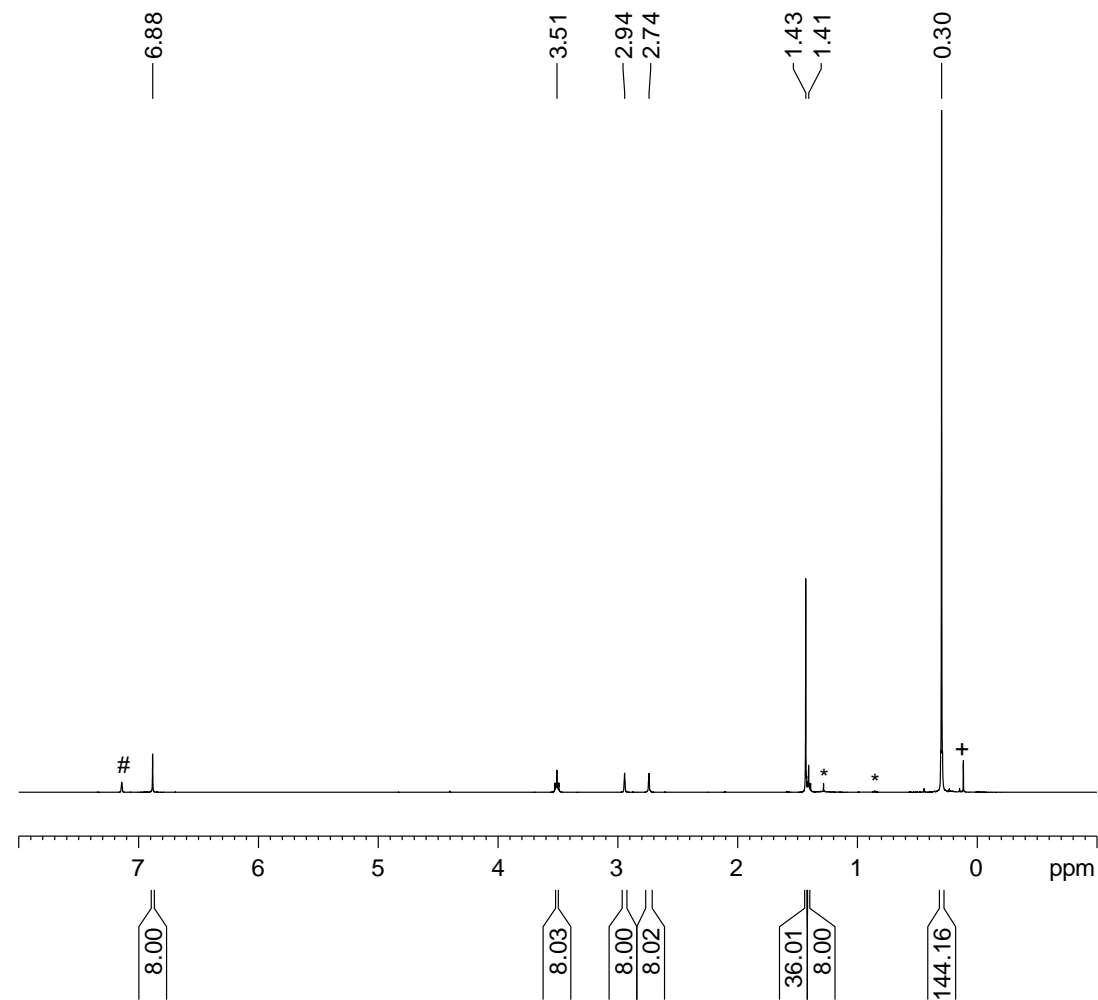
F2 - Acquisition Parameters
Date_ 20191211
Time 14.39 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273858 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S34. ^{29}Si -INEPTND-NMR spectrum of compound **8**.

NMR spectra of compound 9a

¹H-NMR of ([TbbGeH₂][K(thf)_{0.5}]₄) in C₆D₆ (#) at rt, *: *n*-pentane, +: unknown impurity



Current Data Parameters
 NAME AS17F_27112019_300
 EXPNO 10
 PROCNO 1

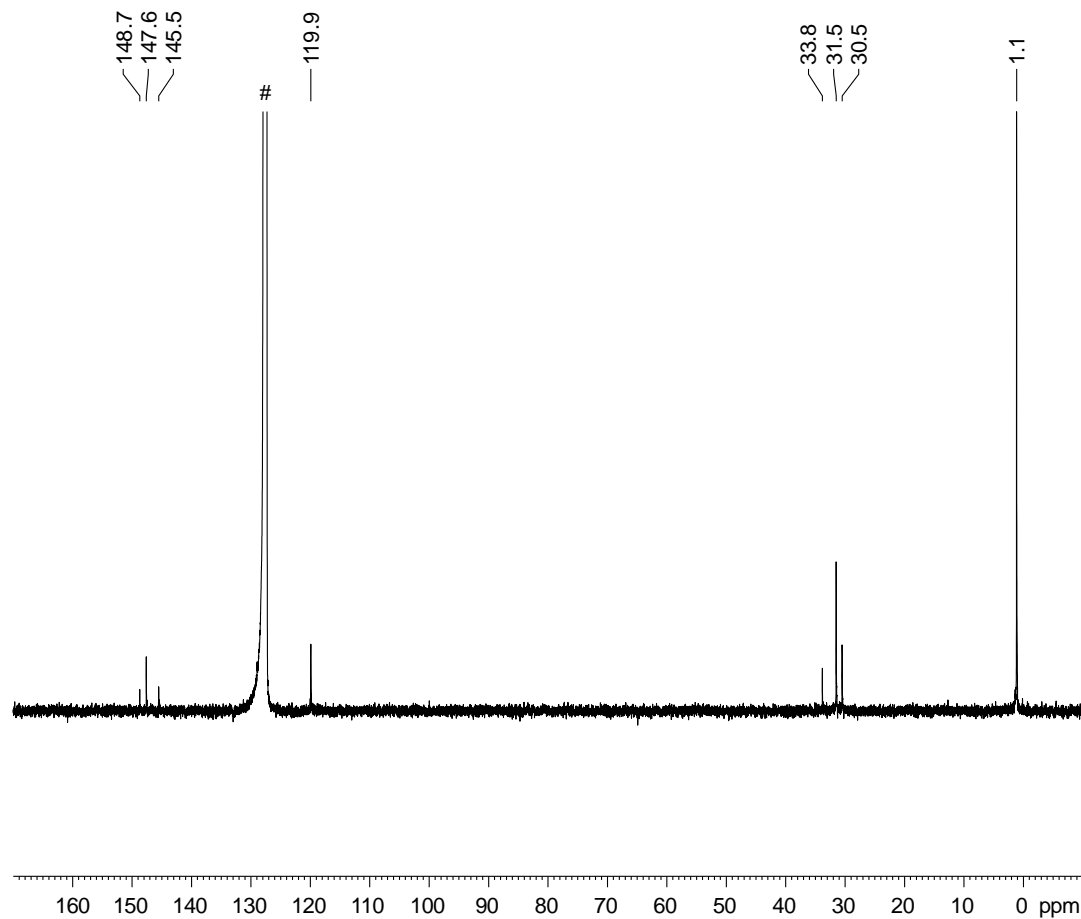
F2 - Acquisition Parameters
 Date_ 20191127
 Time 11.41
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 16
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 90.5
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

==== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S35. ¹H-NMR spectrum of compound 9a.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $(\text{TbbGeH}_2)[\text{K}(\text{thf})_{0.5}]_4$ in C_6D_6 (#) at rt



Current Data Parameters
NAME AS17,18_03122019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191204
Time 1.36
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 10240
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

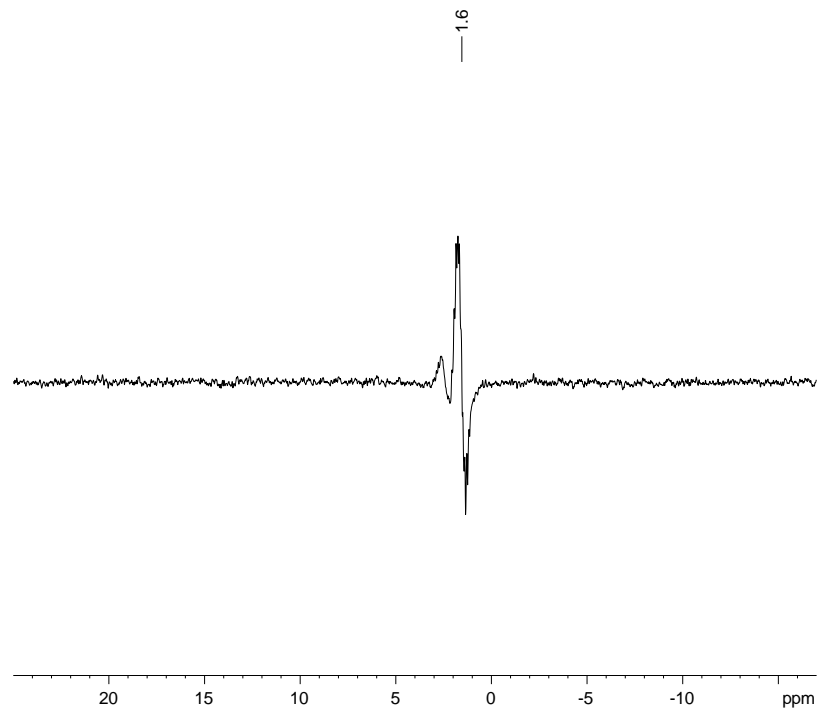
=====
CHANNEL f1
=====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

=====
CHANNEL f2
=====
CPDPRG2 waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S36. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **9a**.

^{29}Si -INEPTND-NMR of $([\text{TbbGeH}_2[\text{K}(\text{thf})_{0.5}]]_4)$ in C_6D_6 at rt



Current Data Parameters
NAME MA649K_10082021_300N
EXPNO 11
PROCNO 1

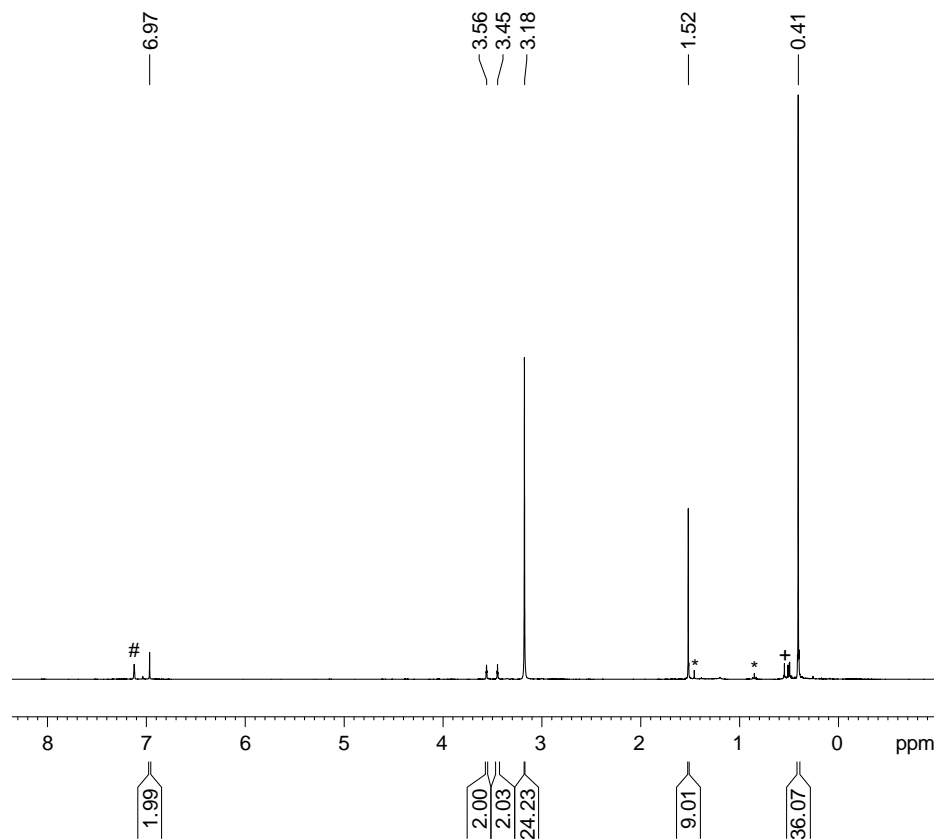
F2 - Acquisition Parameters
Date_ 20210810
Time 14.37 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 128
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273579 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S37. ^{29}Si -INEPTND-NMR spectrum of compound **9a**.

NMR spectra of compound **9b**

¹H-NMR of ([TbbGeH₂][K{[18]-c-6}]) in C₆D₆ (#) at rt, *: *n*-hexane, +: unknown impurities



Current Data Parameters
 NAME MA200K_17122019_400N
 EXPNO 10
 PROCNO 1

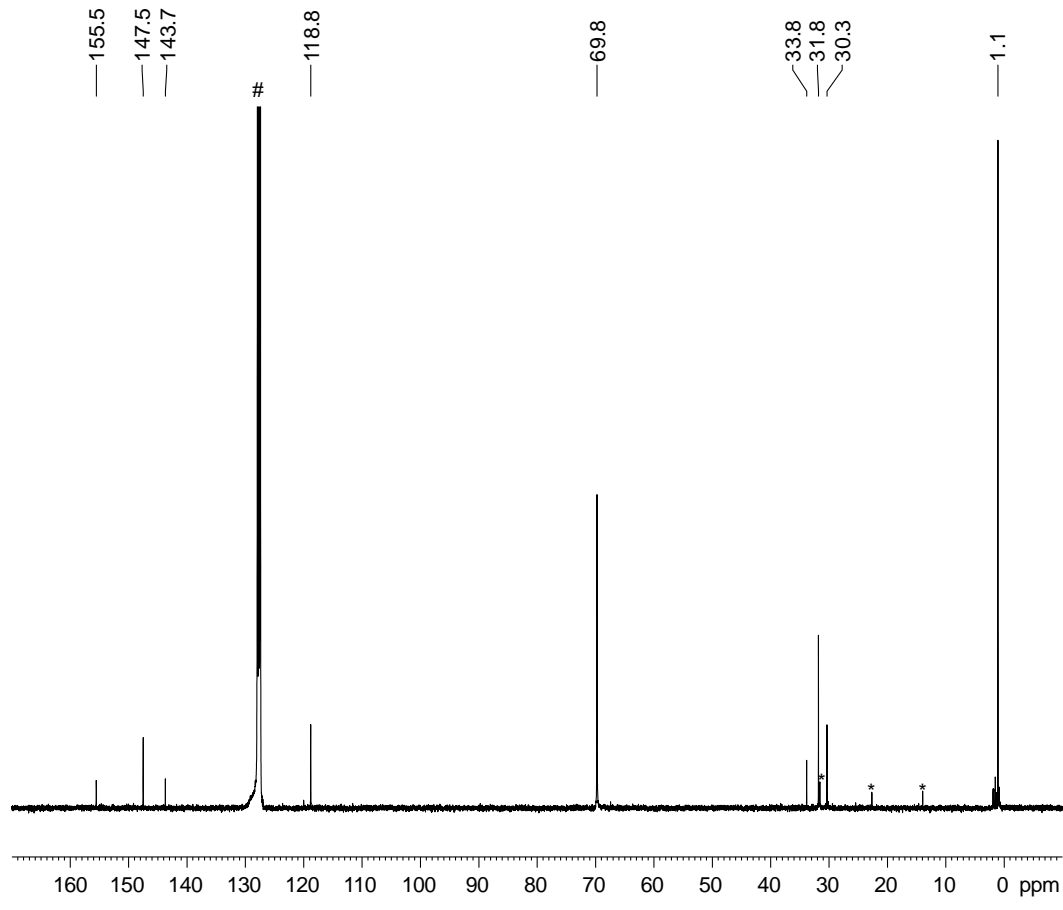
F2 - Acquisition Parameters
 Date_ 20191217
 Time 20.06
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 203
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100070 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S38. ¹H-NMR spectrum of compound **9b**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $([\text{TbbGeH}_2][\text{K}([\text{18-c-6})])$ in C_6D_6 (#) at rt, *: *n*-hexane



Current Data Parameters
NAME MA200K_17122019_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20191218
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

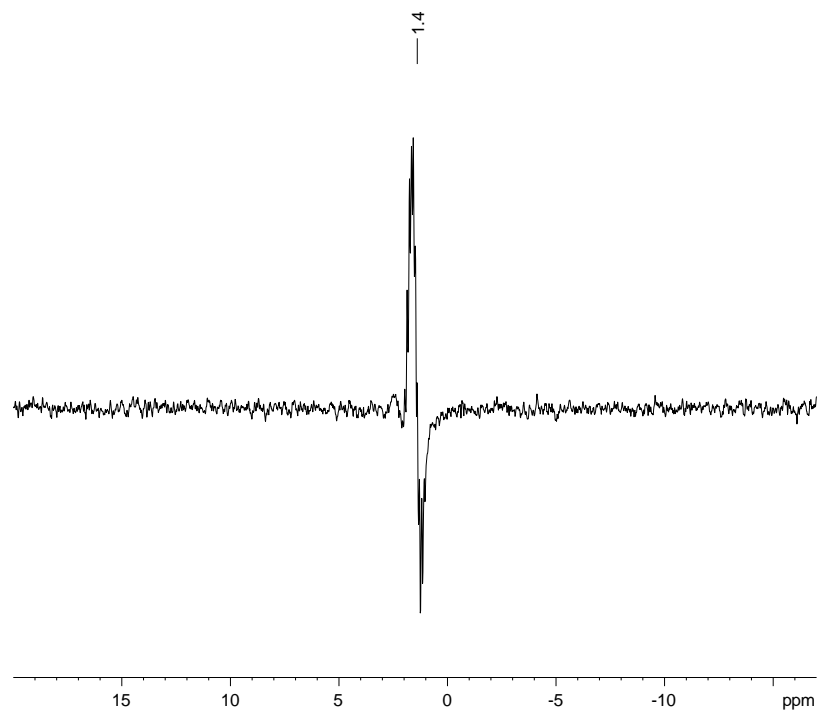
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S39. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **9b**.

²⁹Si-INEPTND-NMR of ([TbbGeH₂][K([18]-c-6)]) in C₆D₆ at rt



Current Data Parameters
NAME MA200K_18122019_300
EXPNO 11
PROCNO 1

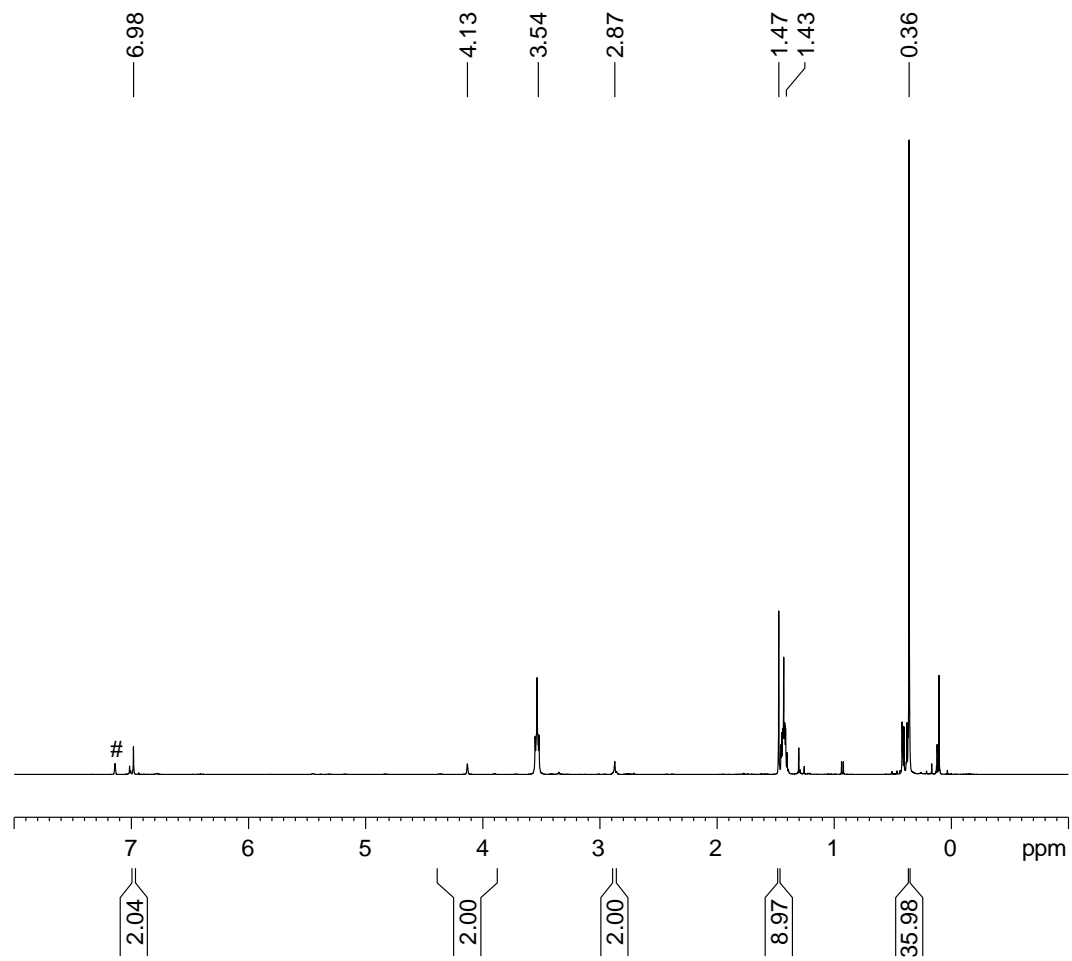
F2 - Acquisition Parameters
Date_ 20191218
Time 17.04 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.0000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.0000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273454 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S40. ²⁹Si-INEPTND-NMR spectrum of compound **9b**.

NMR spectra of compound 10

$^1\text{H-NMR}$ of $([\text{TbSnH}_2][\text{Li}\{\text{thf}\}_x])$ in C_6D_6 (#) at rt



Current Data Parameters
 NAME MA332_15062020_400N
 EXPNO 10
 PROCNO 1

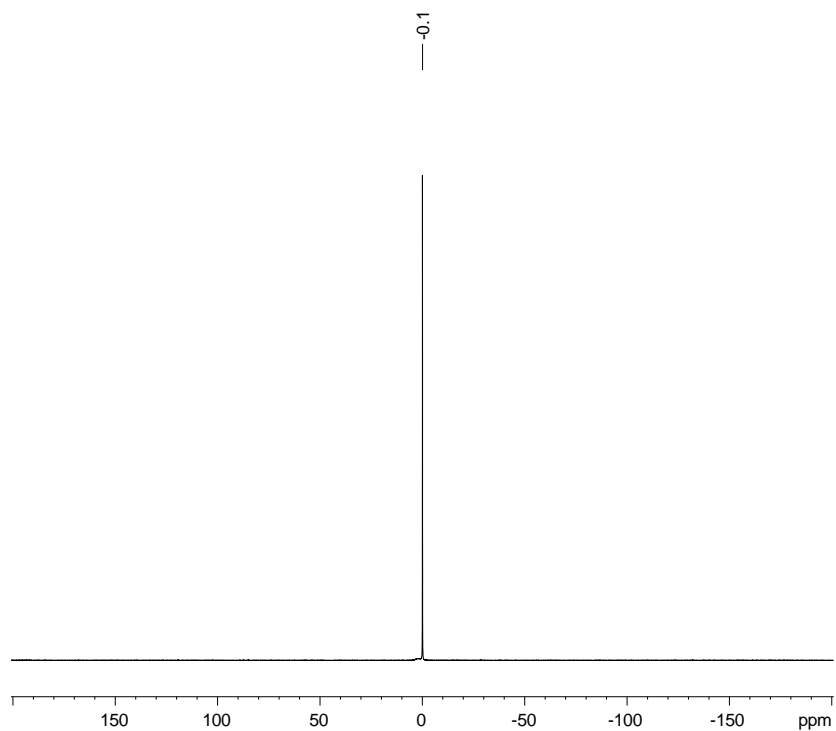
F2 - Acquisition Parameters
 Date_ 20200615
 Time 20.05
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 71.8
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

==== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100003 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S41. $^1\text{H-NMR}$ spectrum of compound 10.

^7Li -NMR of $([\text{TbbSnH}_2][\text{Li}(\text{thf})_x])$ in thf-d_8 at rt



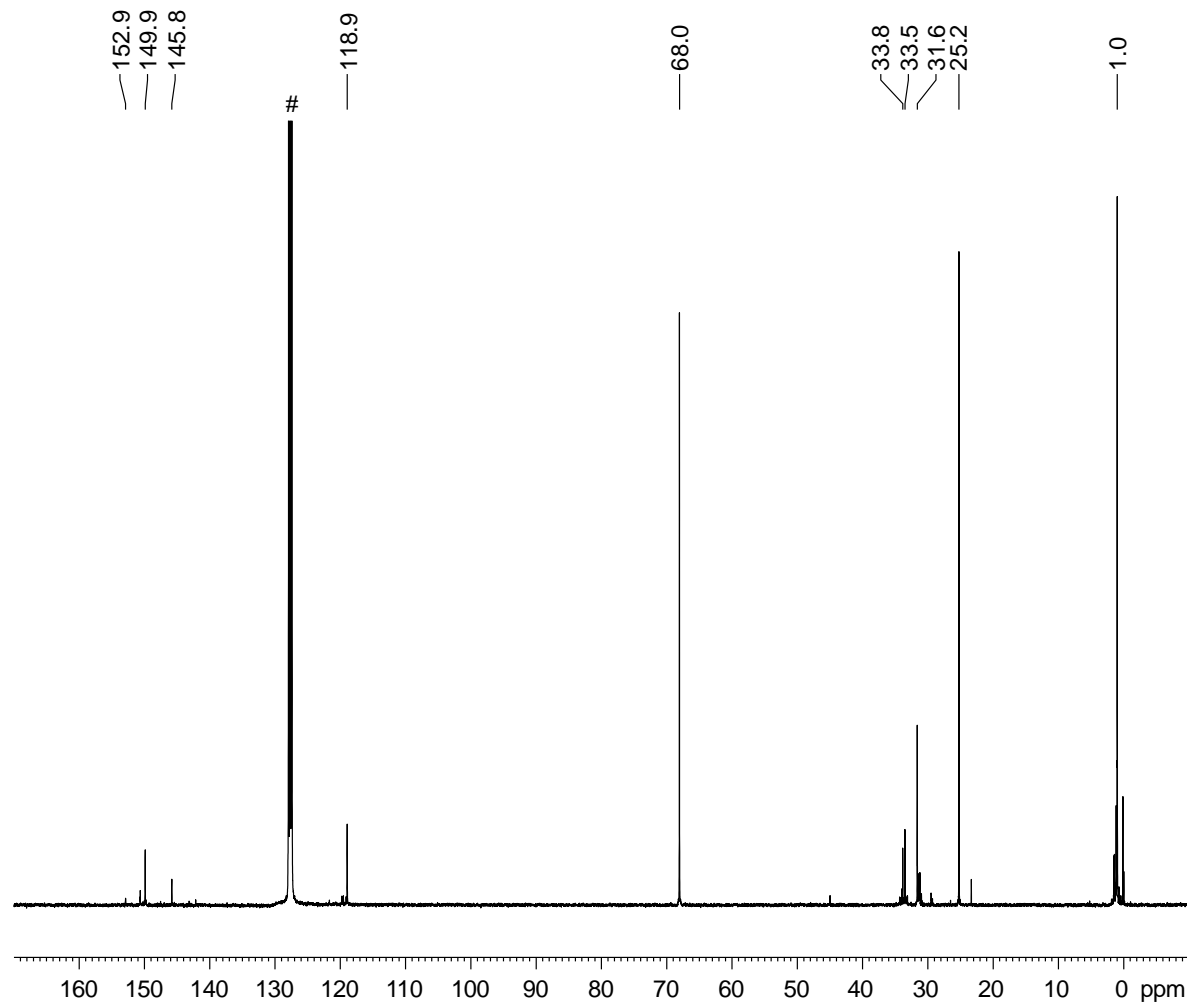
Current Data Parameters
NAME MA320_28052020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200528
Time 8.44 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 32050
SOLVENT THF
NS 256
DS 0
SWH 46875.000 Hz
FIDRES 2.925117 Hz
AQ 0.3418667 sec
RG 204.67
DW 10.667 usec
DE 6.50 usec
TE 298.0 K
D1 0.20000000 sec
TD0 1
SFO1 116.6419139 MHz
NUC1 ^7Li
P0 2.94 usec
P1 8.81 usec
PLW1 34.00000000 W

F2 - Processing parameters
SI 32768
SF 116.6419139 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S42. ^7Li -NMR spectrum of compound **10**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $([\text{TbbSnH}_2][\text{Li}\{\text{thf}\}_x])$ in C_6D_6 (#) at rt



Current Data Parameters
NAME MA332_15062020_400N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200616
Time 2.08
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

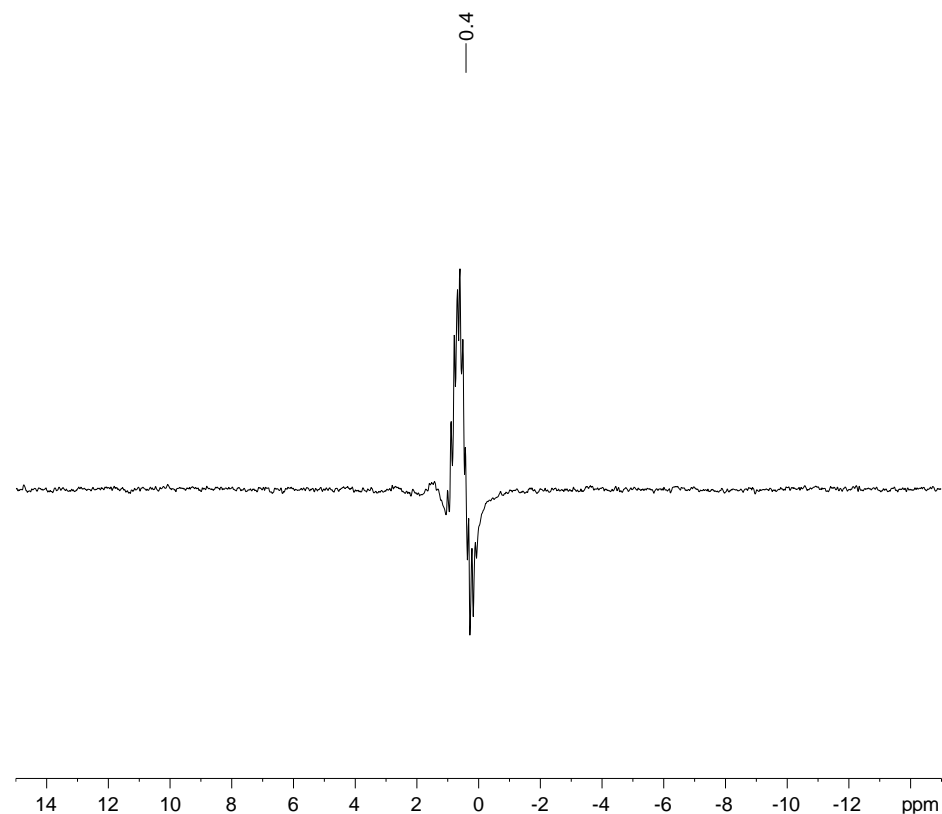
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S43. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **10**.

^{29}Si -INEPTND-NMR of $([\text{TbbSnH}_2][\text{Li}(\text{thf})_x])$ in thf-d_8 at rt



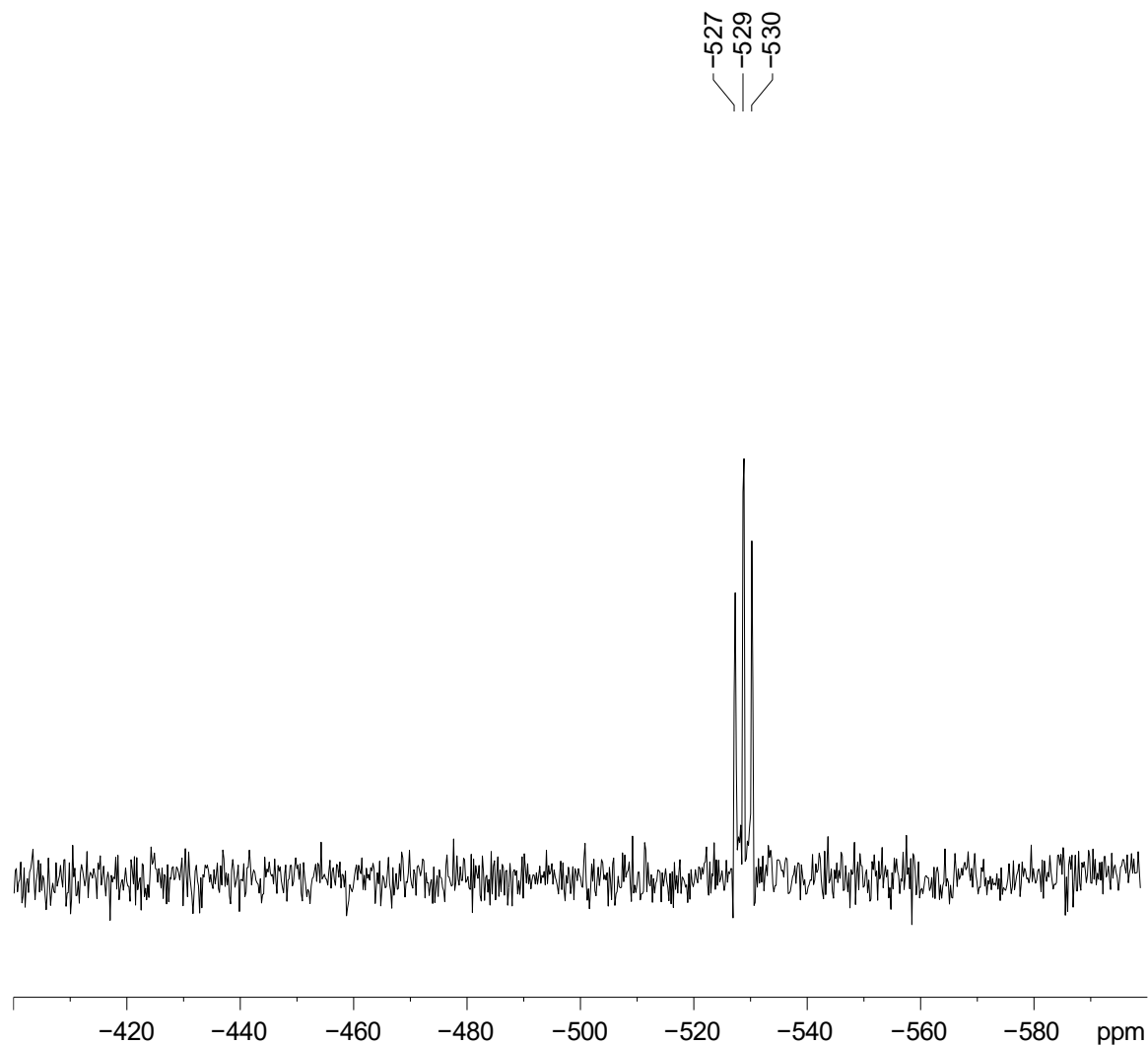
Current Data Parameters
NAME MA320_28052020_300
EXPNO 21
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200528
Time 10.24 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT THF
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273880 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S44. ^{29}Si -INEPTND-NMR spectrum of compound **10**.

^{119}Sn -NMR of $([\text{TbbSnH}_2][\text{Li}\{\text{thf}\}_x])$ in thf-d_8 at rt



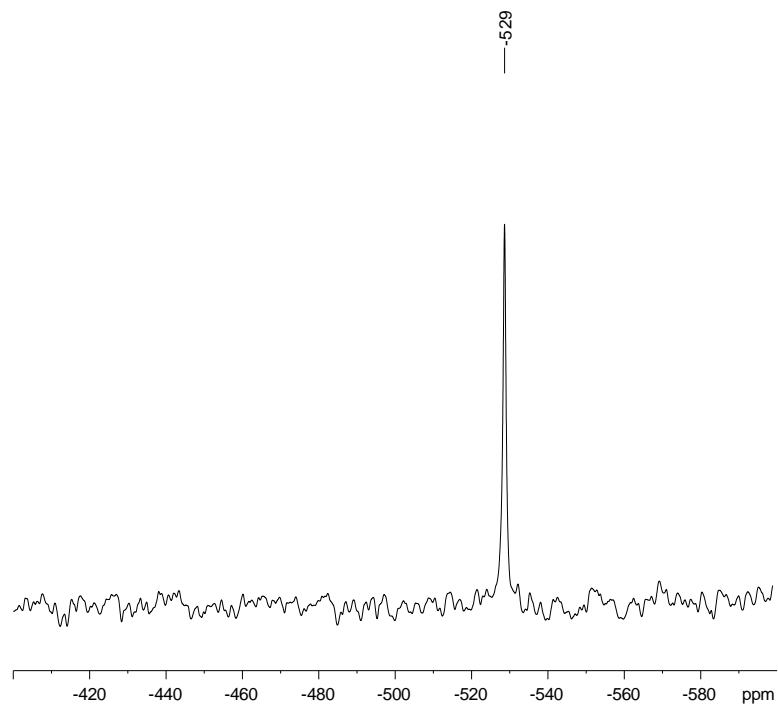
Current Data Parameters
NAME MA320_28052020_300
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200528
Time 9.11 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT THF
NS 15360
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8979900 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.40

Figure S45. ^{119}Sn -NMR spectrum of compound **10**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $([\text{TbbSnH}_2][\text{Li}(\text{thf})_x])$ in thf-d_8 at rt



Current Data Parameters
NAME MA320_28052020_300
EXPNO 22
PROCNO 1

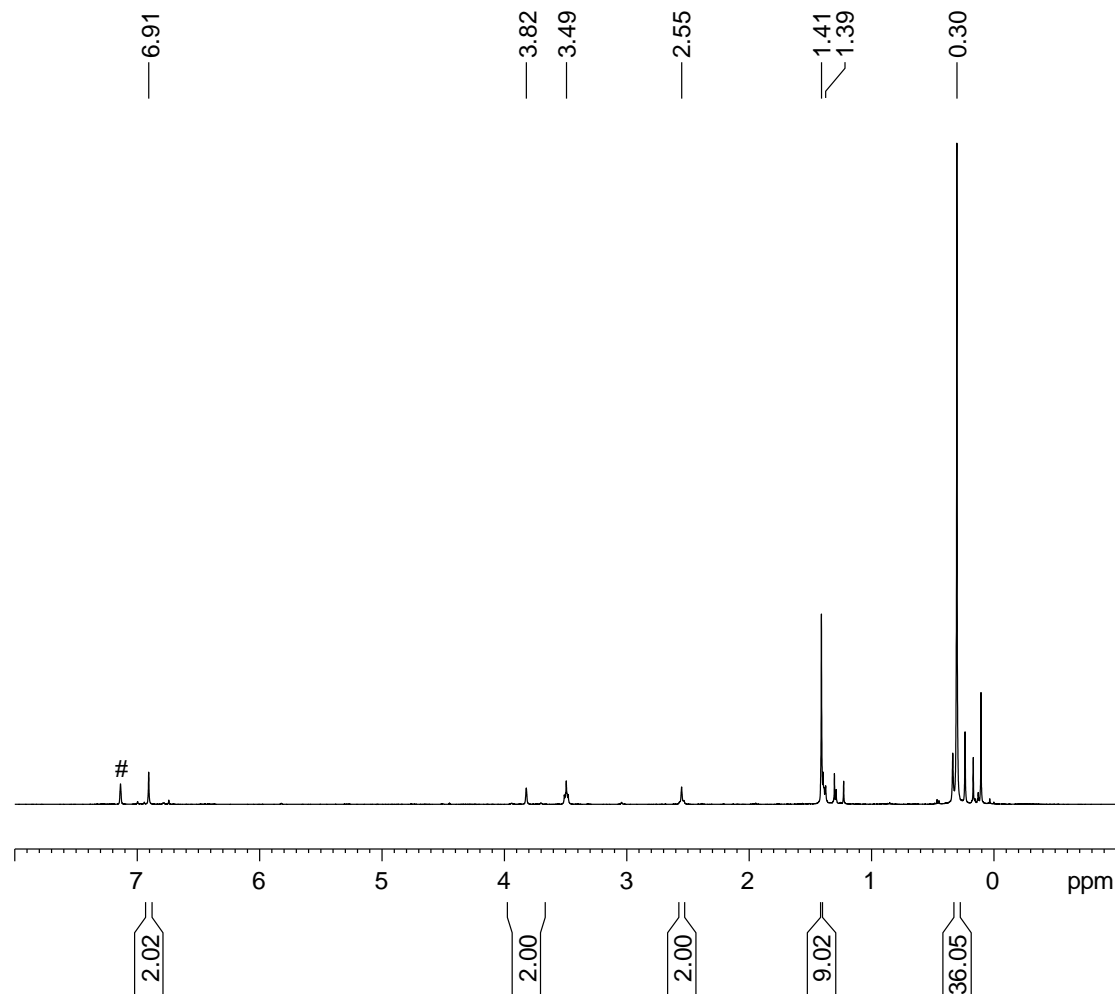
F2 - Acquisition Parameters
Date_ 20200528
Time 10.39 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgpg30
TD 39186
SOLVENT THF
NS 2253
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8979898 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG2 waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 1.40

Figure S46. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **10**.

NMR spectra of compound **11a**

$^1\text{H-NMR}$ of $([\text{TbbSnH}_2][\text{K}\{\text{thf}\}_x])$ in C_6D_6 (#) at rt



Current Data Parameters
 NAME MA344_29062020_400N
 EXPNO 10
 PROCNO 1

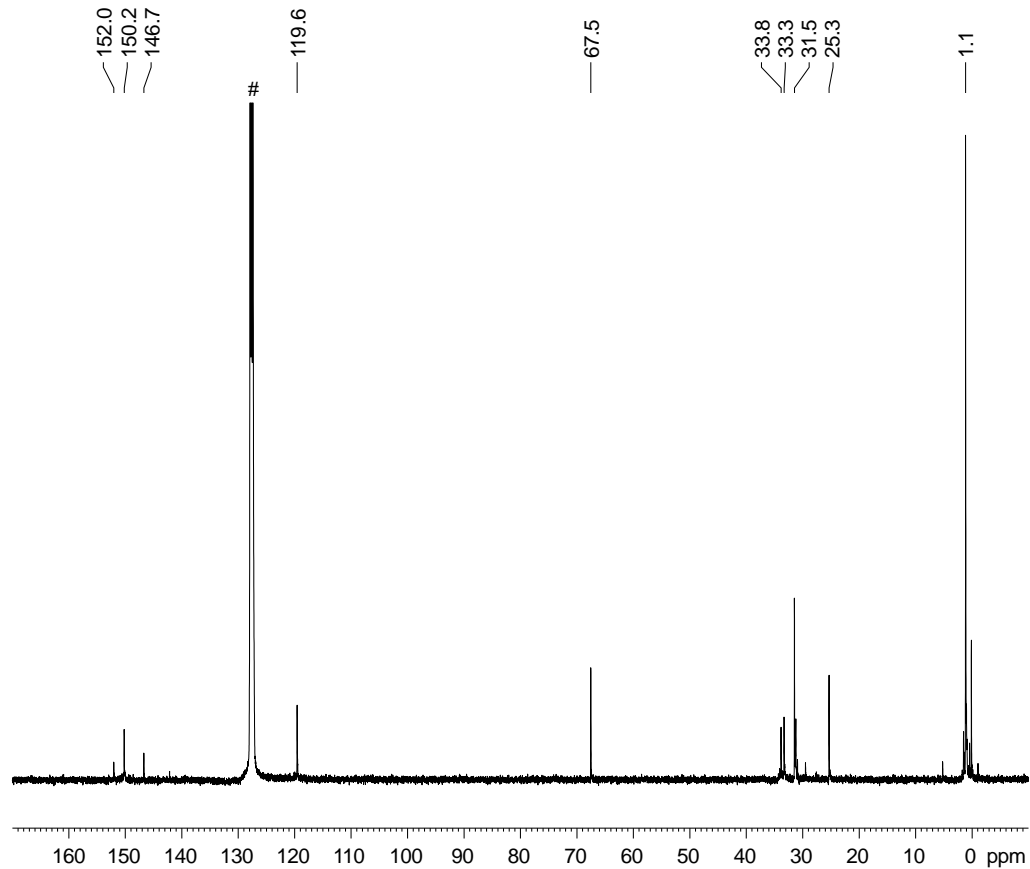
F2 - Acquisition Parameters
 Date_ 20200629
 Time 15.12
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 16
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 101
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100016 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S47. $^1\text{H-NMR}$ spectrum of compound **11a**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $([\text{TbbSnH}_2][\text{K}\{\text{thf}\}_x])$ in C_6D_6 (#) at rt



Current Data Parameters
NAME MA344_29062020_400N
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200630
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

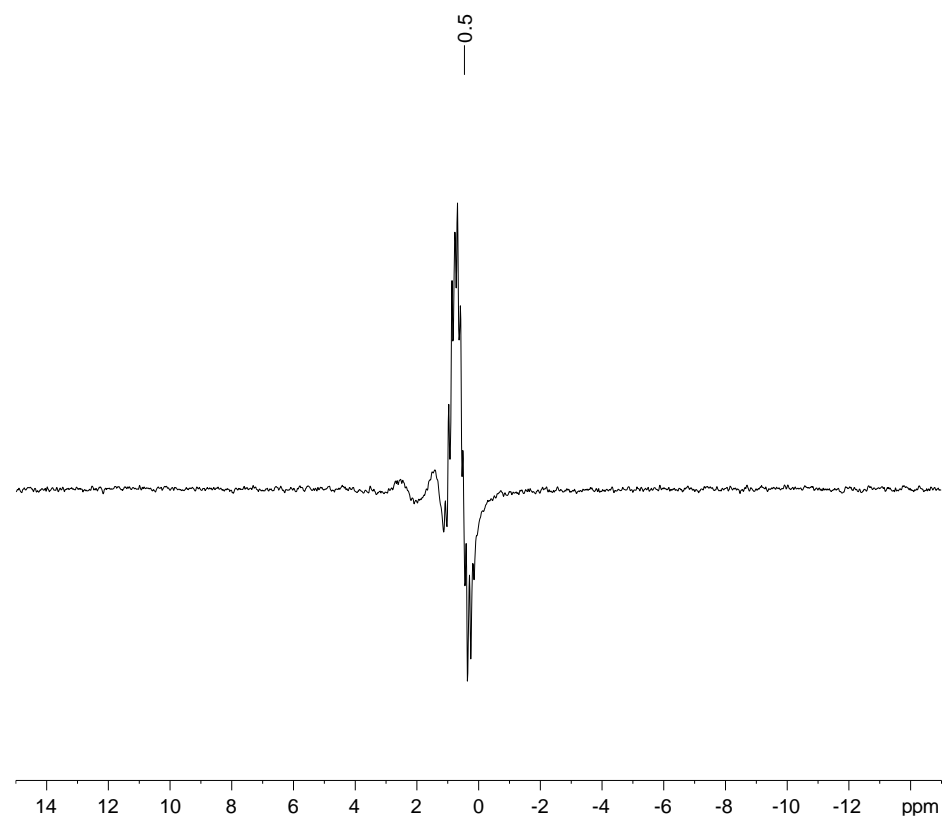
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S48. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **11a**.

^{29}Si -INEPTND-NMR of $([\text{TbbSnH}_2][\text{K}(\text{thf})_x])$ in thf-d_8 at rt



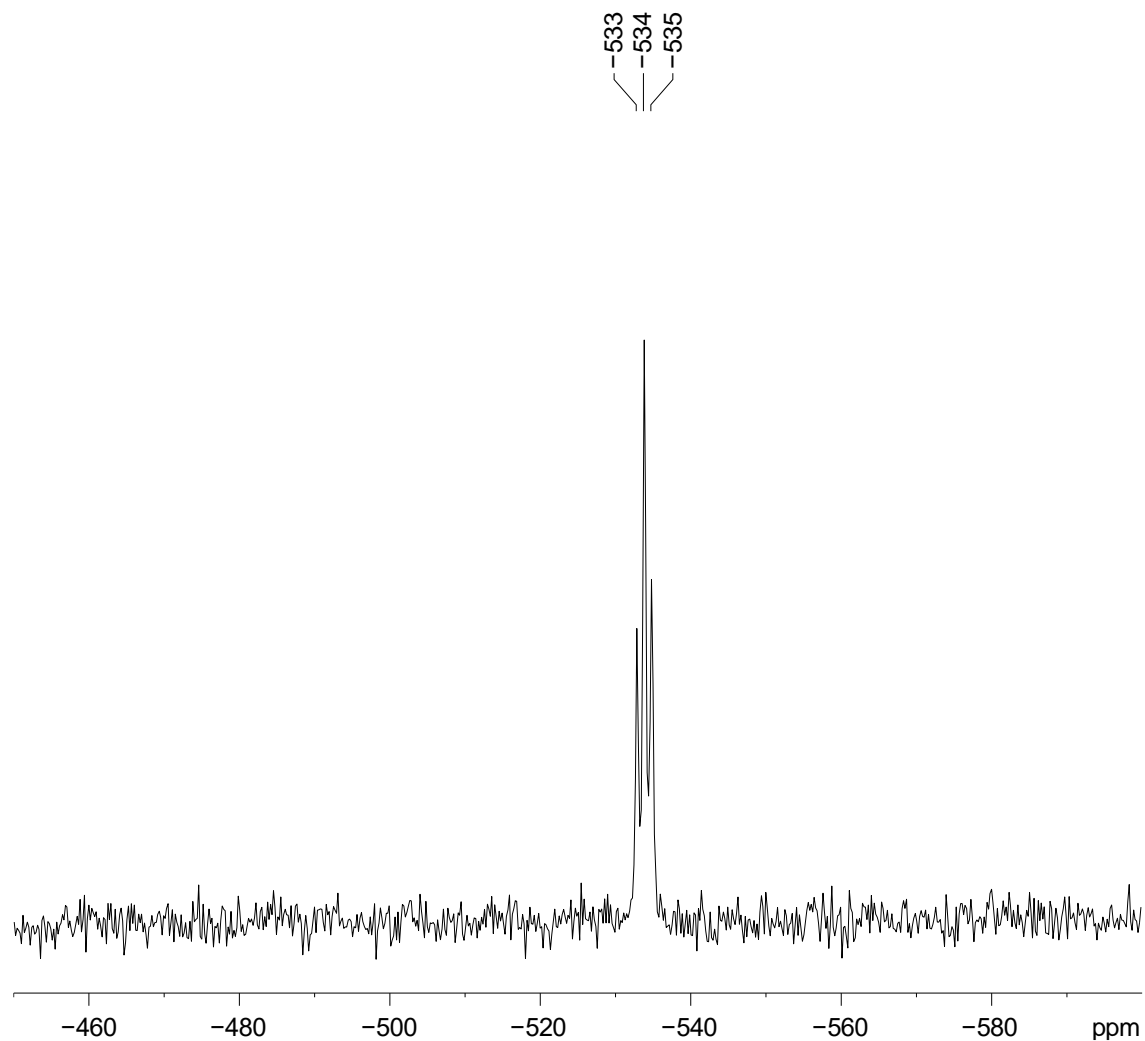
Current Data Parameters
NAME MA340_23062020_300
EXPNO 22
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200623
Time 11.25 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT THF
NS 300
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273880 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S49. ^{29}Si -INEPTND-NMR spectrum of compound **11a**.

^{119}Sn -NMR of $([\text{TbbSnH}_2][\text{K}(\text{thf})_x])$ in thf-d_8 at rt



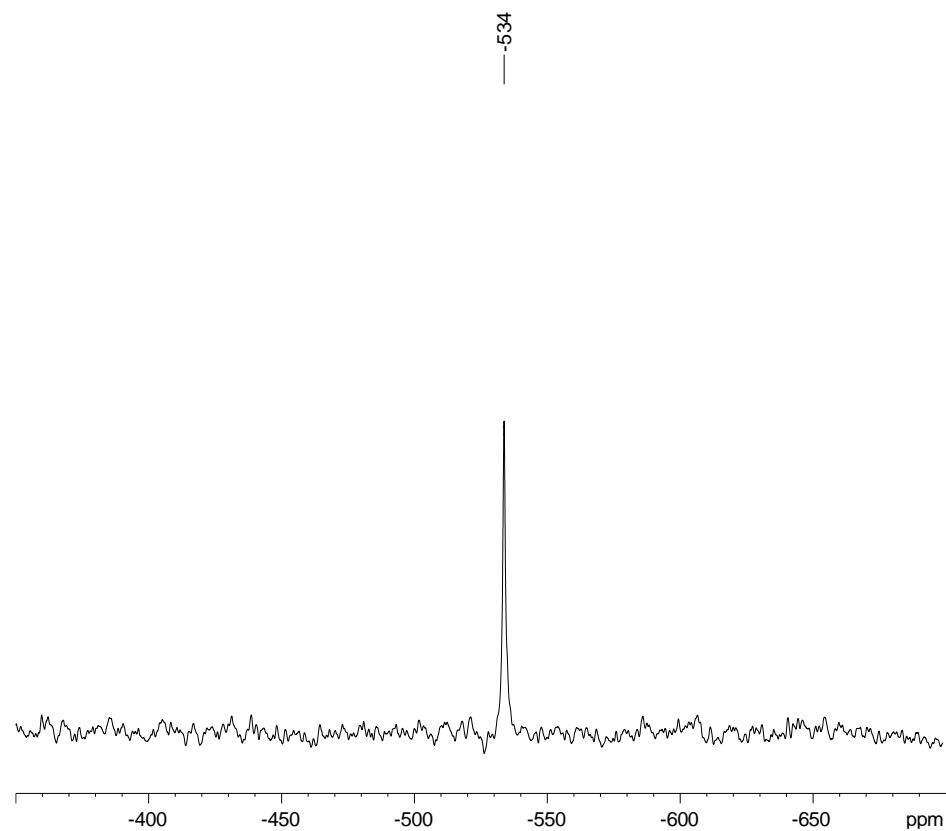
Current Data Parameters
NAME MA340_23062020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200623
Time 9.26 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT THF
NS 16384
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8867979 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S50. ^{119}Sn -NMR spectrum of compound **11a**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $([\text{TbbSnH}_2][\text{K}(\text{thf})_x])$ in thf-d_8 at rt



Current Data Parameters
NAME MA340_23062020_300
EXPNO 21
PROCNO 1

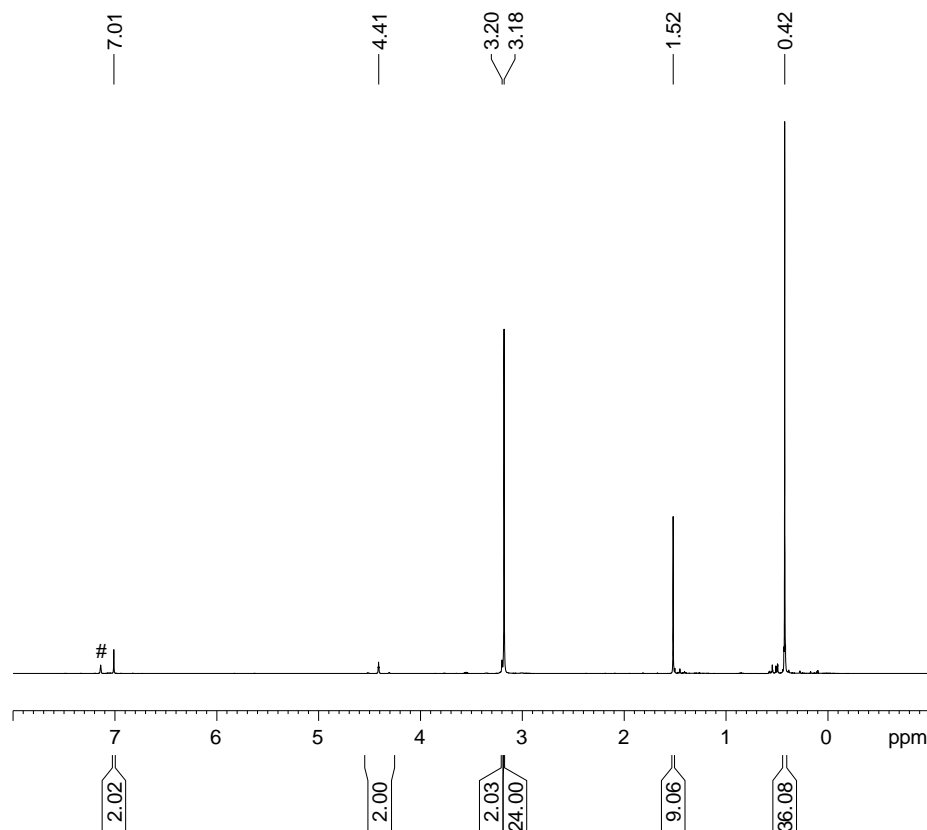
F2 - Acquisition Parameters
Date_ 20200623
Time 11.09 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT THF
NS 2458
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8867977 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 1.40

Figure S51. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **11a**.

NMR spectra of compound **11b**

¹H-NMR of ([TbbSnH₂][K{[18]crown-6}]) in C₆D₆ (#) at rt



Current Data Parameters
 NAME MA346_01072020_400N
 EXPNO 10
 PROCNO 1

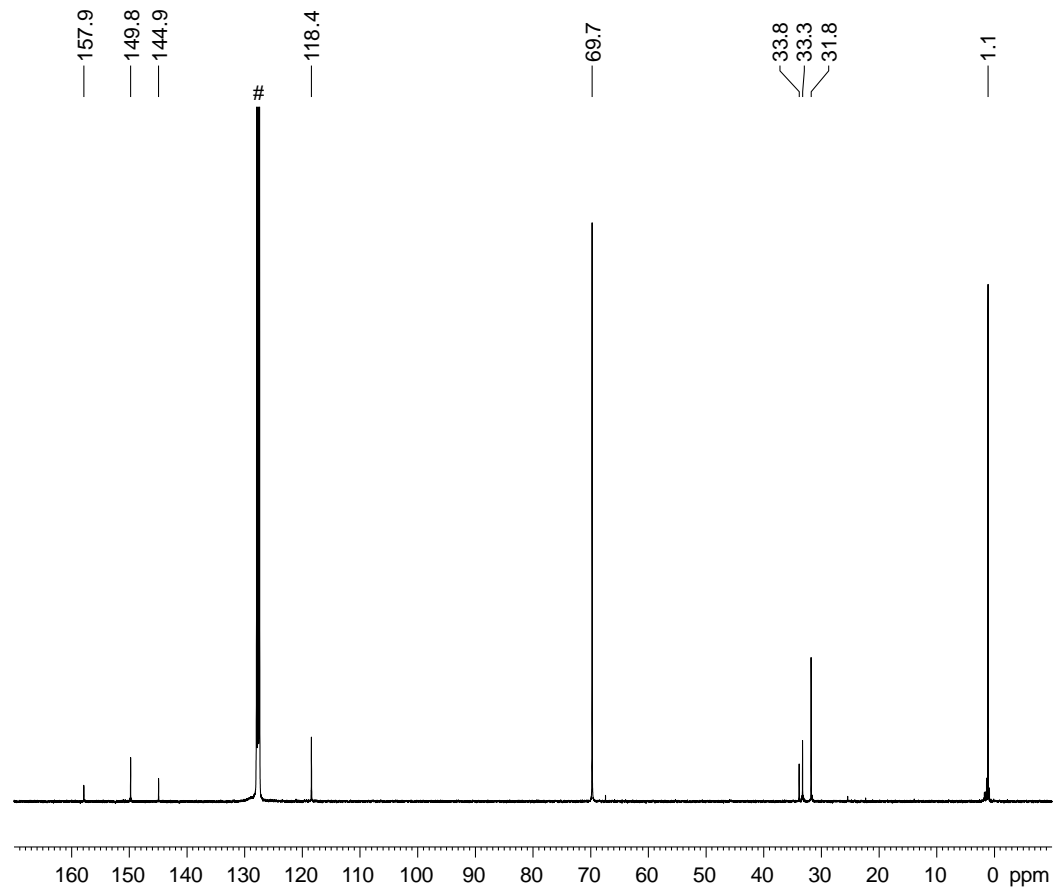
F2 - Acquisition Parameters
 Date_ 20200701
 Time 15.20
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 16
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 90.5
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

==== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S52. ¹H-NMR spectrum of compound **11b**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of ([TbbSnH₂][K{[18]crown-6}]) in C₆D₆ (#) at rt



Current Data Parameters
NAME MA346_01072020_400N
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200702
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

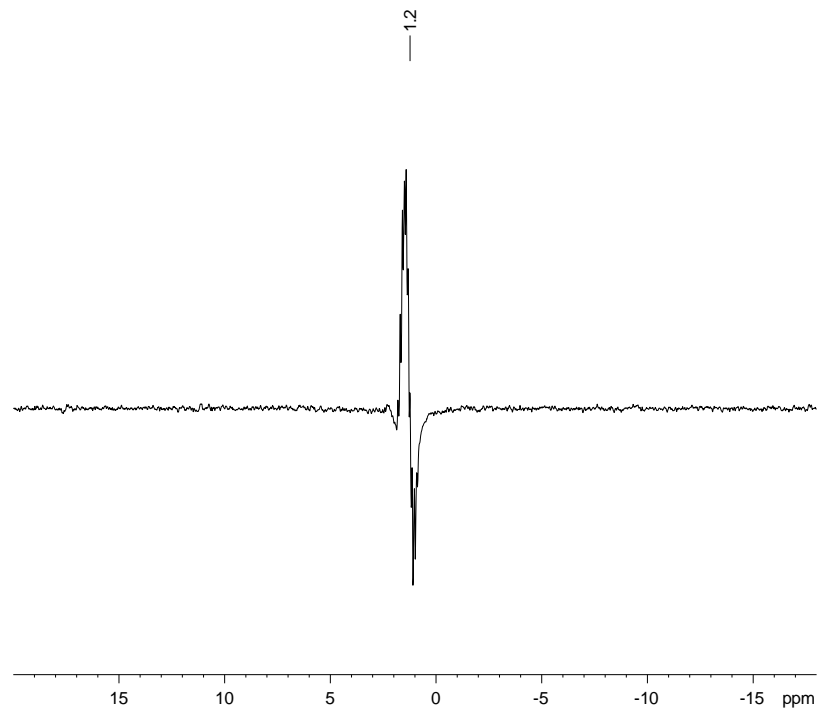
==== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

==== CHANNEL f2 =====
CPDPRG2 waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S53. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **11b**.

²⁹Si-INEPTND-NMR of ((TbbSnH₂)[K{[18]crown-6}]) in C₆D₆ at rt



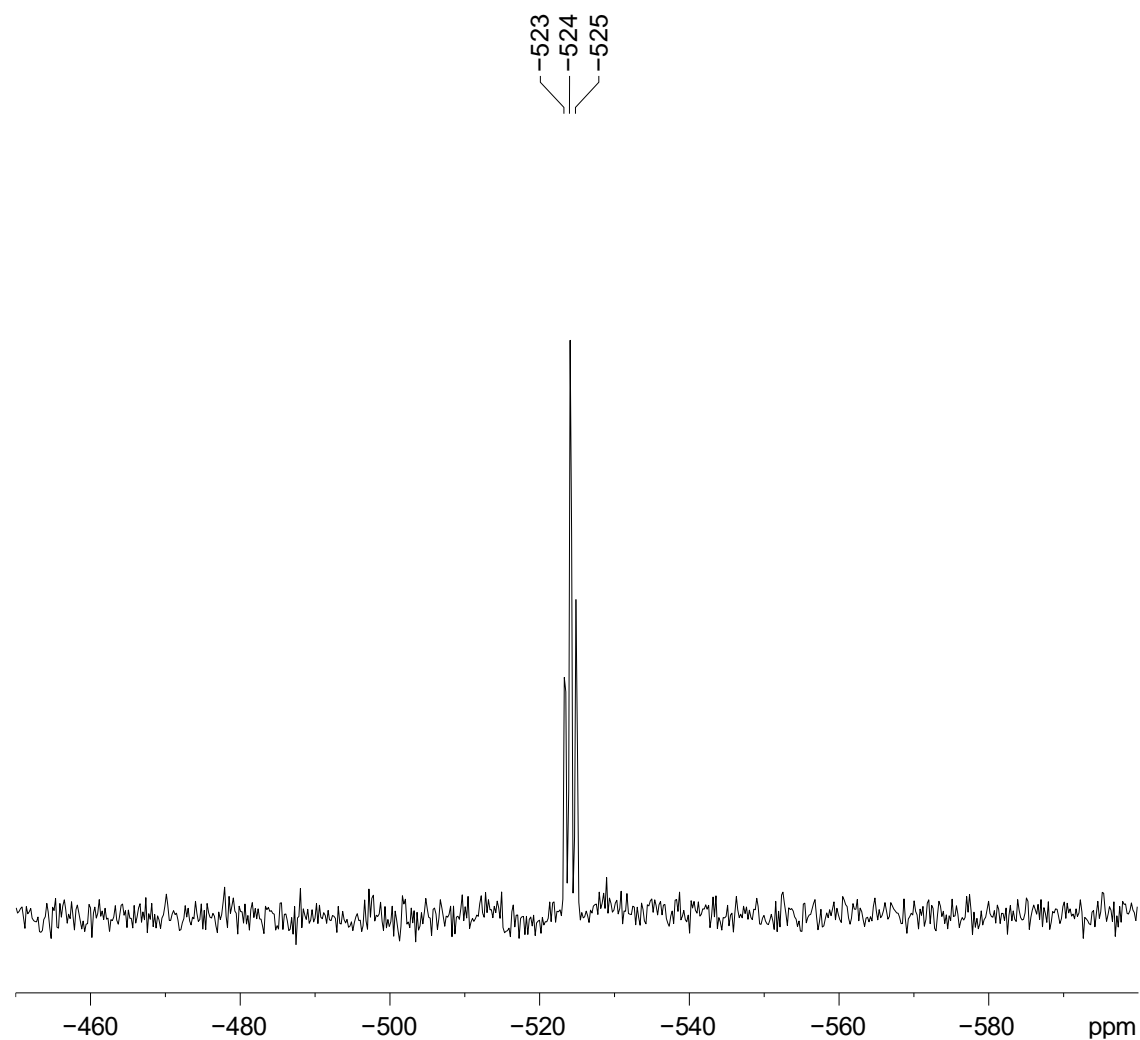
Current Data Parameters
NAME MA346_02072020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200702
Time 8.13 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273582 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S54. ²⁹Si-INEPTND-NMR spectrum of compound **11b**.

^{119}Sn -NMR of $([\text{TbbSnH}_2[\text{K}\{[18]\text{crown-6}}]])$ in C_6D_6 at rt



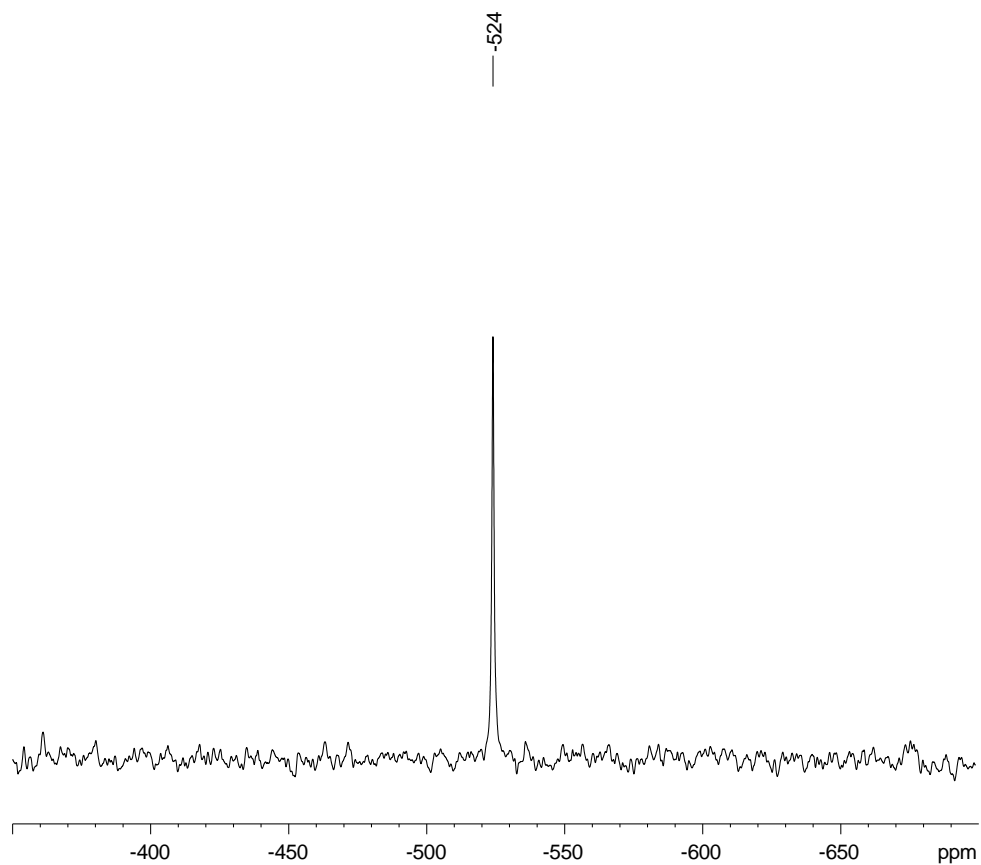
Current Data Parameters
NAME MA346_01072020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200701
Time 14.31 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT C6D6
NS 16384
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8867979 MHz
NUC1 119Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 3.00 Hz
GB 0
PC 1.40

Figure S55. ^{119}Sn -NMR spectrum of compound **11b**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $([\text{TbSnH}_2][\text{K}(\text{18-crown-6})])$ in C_6D_6 at rt



Current Data Parameters
NAME MA346_02072020_300
EXPNO 10
PROCNO 1

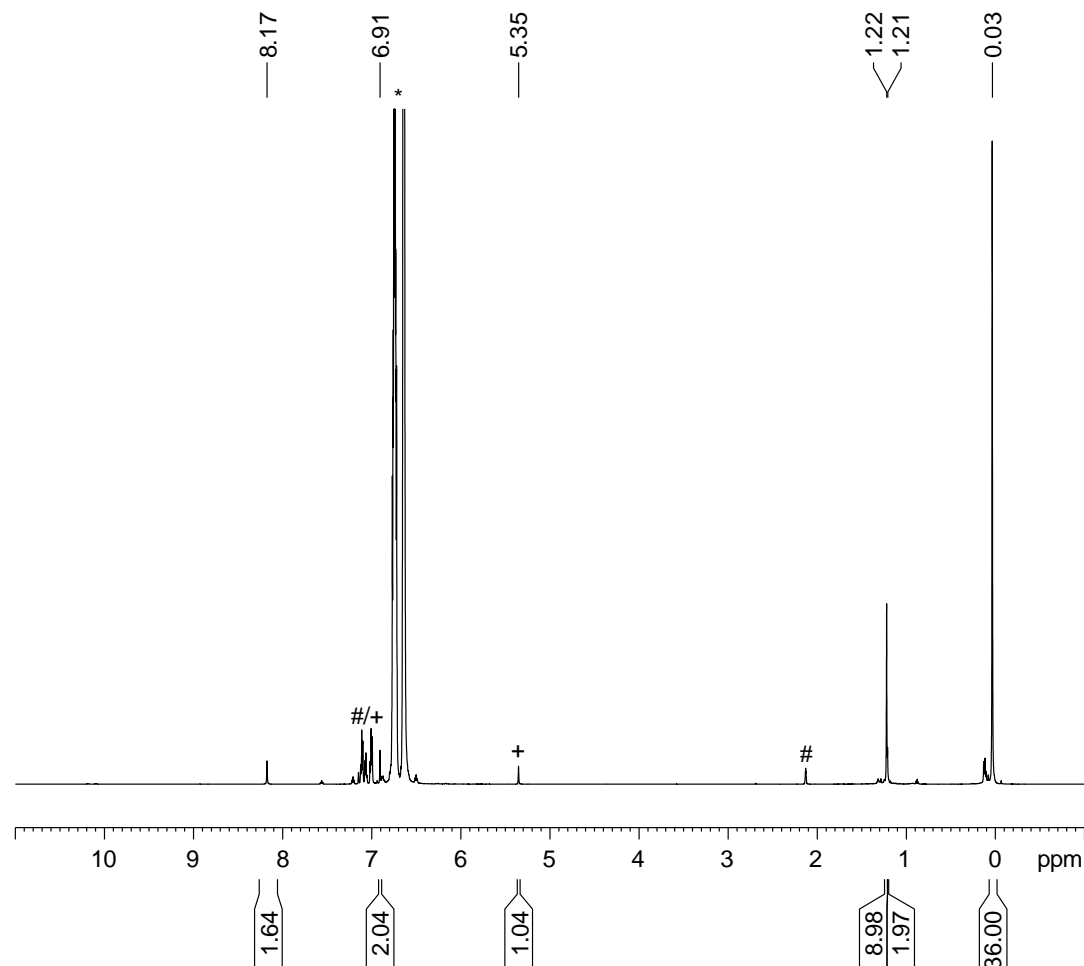
F2 - Acquisition Parameters
Date_ 20200702
Time 7.57 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT C6D6
NS 2458
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8867977 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 1.40

Figure S56. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **11b**.

NMR spectra of compound 12

$^1\text{H-NMR}$ of $[\text{TbbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 (#) + 1,2-difluorobenzene (*) at -40°C , +: HCPPh_3



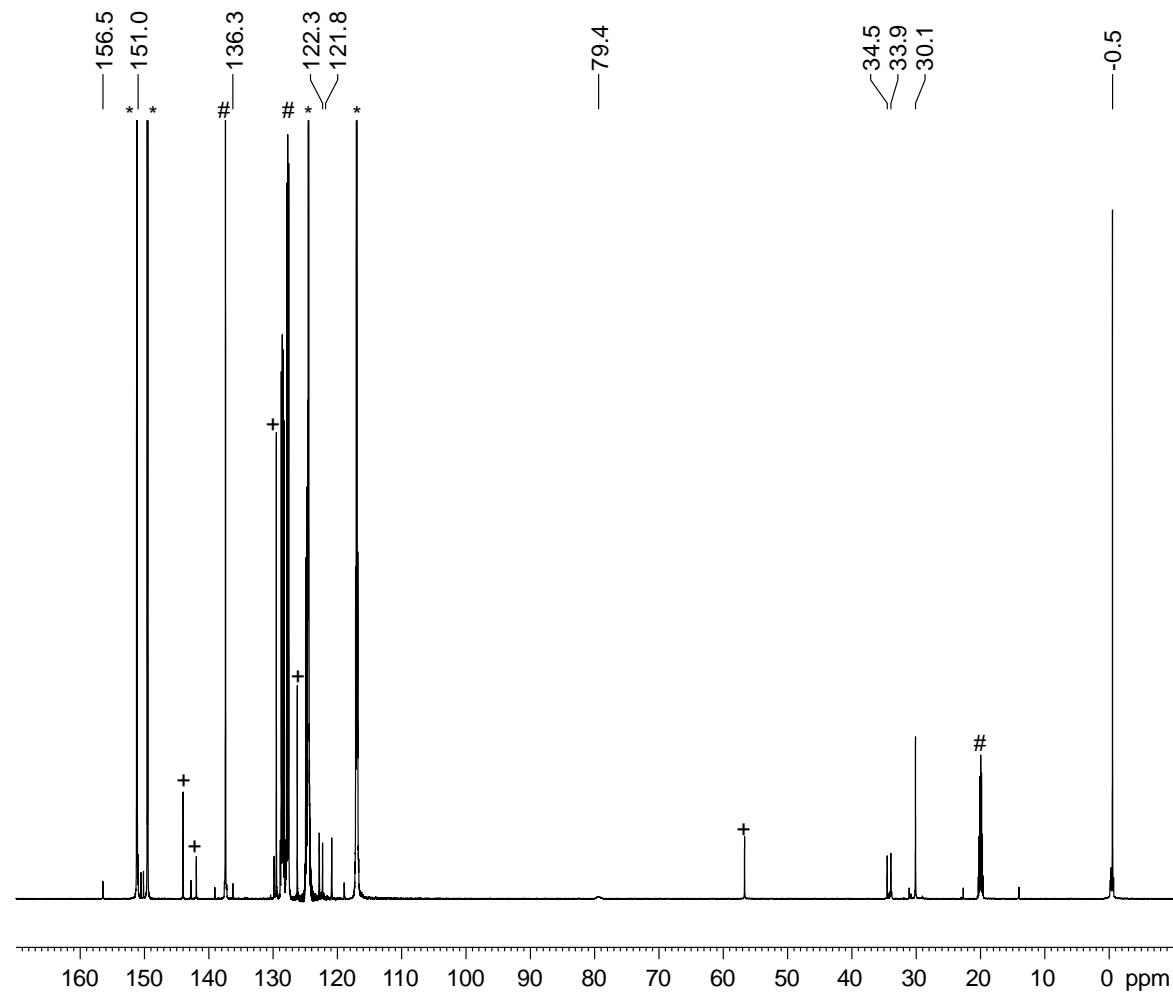
Current Data Parameters
 NAME MA241_18022020_600TT
 EXPNO 10
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 20200218
 Time_ 9.03 h
 INSTRUM spect
 PROBHD Z126545_0027 (
 PULPROG zg30
 TD 65536
 SOLVENT Tol
 NS 32
 DS 2
 SWH 12019.230 Hz
 FIDRES 0.366798 Hz
 AQ 2.7262976 sec
 RG 8.13
 DW 41.600 usec
 DE 10.00 usec
 TE 234.1 K
 D1 1.00000000 sec
 TD0 1
 SFO1 600.1337058 MHz
 NUC1 ^1H
 P1 12.00 usec
 PLW1 23.41200066 W

F2 - Processing parameters
 SI 65536
 SF 600.1300000 MHz
 WDW EM
 SSB 0
 LB 0.30 Hz
 GB 0
 PC 1.00

Figure S57. $^1\text{H-NMR}$ spectrum of compound 12.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 (#) + 1,2-difluorobenzene (*) at -40°C , +: HCPH_3



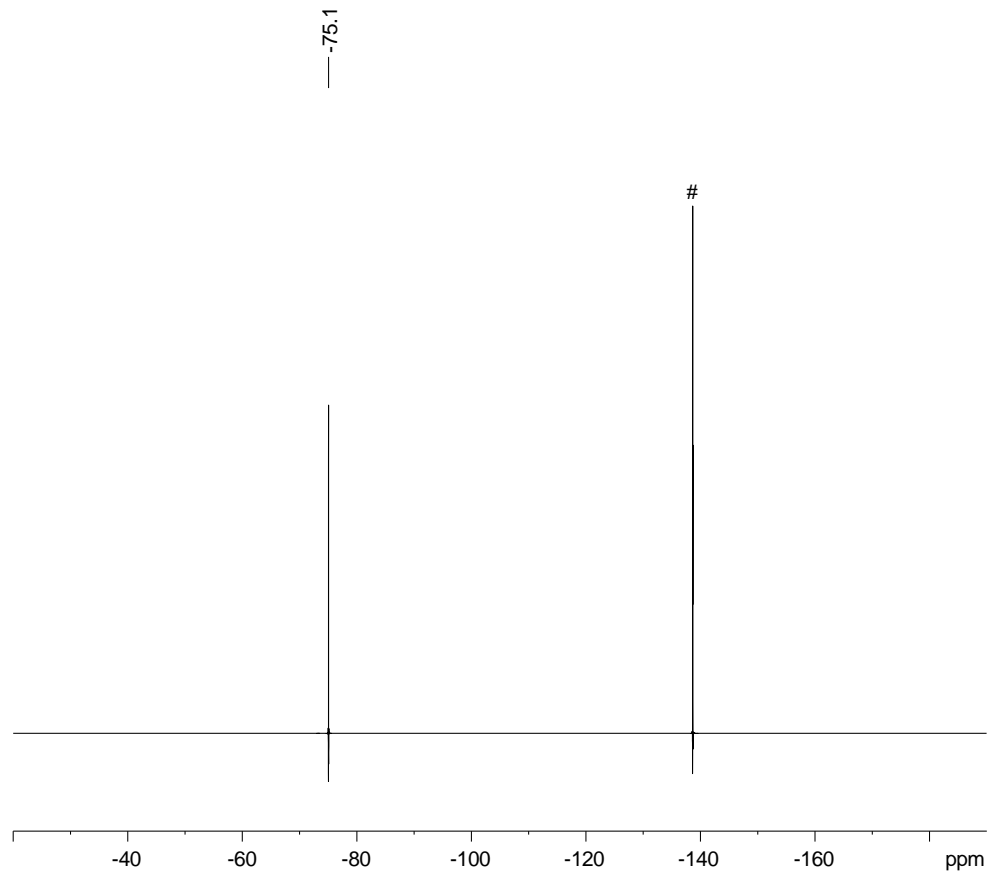
Current Data Parameters
 NAME MA241_18022020_600T
 EXPNO 11
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 20200218
 Time 10.33 h
 INSTRUM spect
 PROBHD Z126545_0027 (
 PULPROG udef
 TD 25902
 SOLVENT Tol
 NS 1024
 DS 8
 SWH 36231.883 Hz
 FIDRES 2.797613 Hz
 AQ 0.3574476 sec
 RG 189.6
 DW 13.800 usec
 DE 18.00 usec
 TE 234.1 K
 D1 4.00000000 sec
 D12 0.00002000 sec
 D20 20.00000000 sec
 TD0 1
 SFO1 150.9178988 MHz
 NUC1 13C
 P1 10.00 usec
 P13 2000.00 usec
 P26 500.00 usec
 PLW1 57.02700043 W
 SPNAM[5] Crp60comp.4
 SPOAL5 0.500
 SPOFFS5 0 Hz
 SPW5 8.71310043 W
 SPNAM[8] Crp60.0.5.20.1
 SPOAL8 0.500
 SPOFFS8 0 Hz
 SPW8 8.71310043 W
 SFO2 600.1324005 MHz
 NUC2 1H
 CPDPRG[2] waltz16
 PCPD2 70.00 usec
 PLW2 23.41200066 W
 PLW12 0.68803000 W

F2 - Processing parameters
 SI 131072
 SF 150.9028085 MHz
 WDW EM
 SSB 0
 LB 2.00 Hz
 GB 0
 PC 1.40

Figure S58. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **12**.

$^{19}\text{F}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2[\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene (#) at -40°C



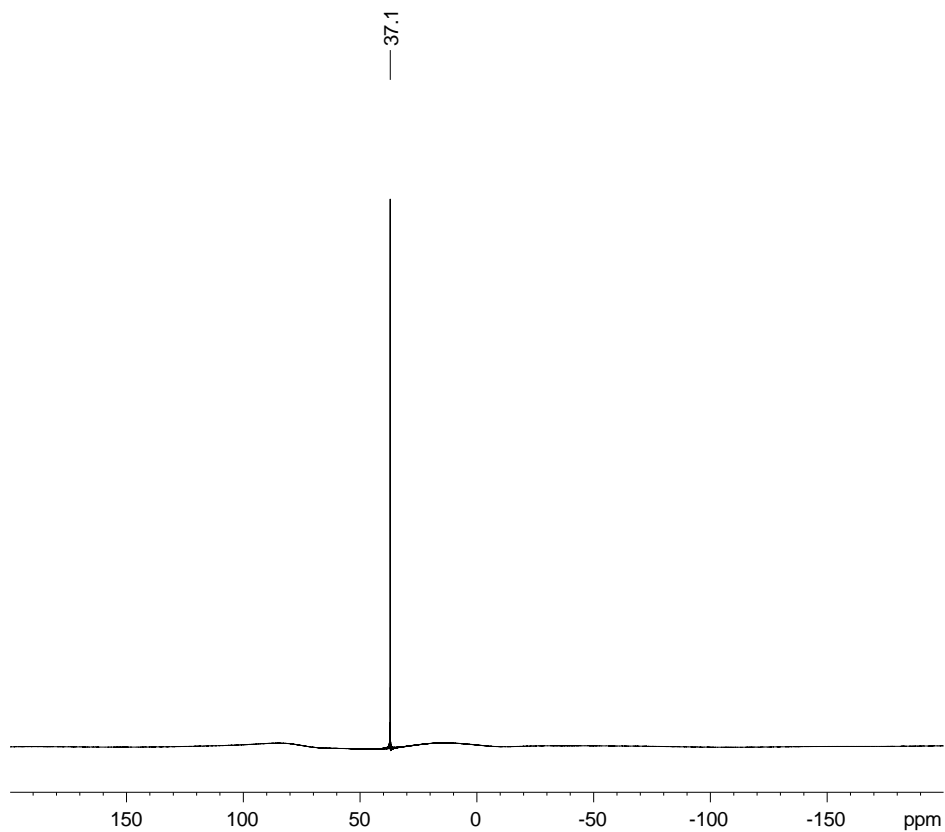
Current Data Parameters
NAME MA241_18022020_600TT
EXPNO 17
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200218
Time 14.43 h
INSTRUM spect
PROBHD Z126545_0027 (
PULPROG zgfgqn
TD 131072
SOLVENT Tol
NS 32
DS 4
SWH 133928.578 Hz
FIDRES 2.043588 Hz
AQ 0.4893355 sec
RG 116.78
DW 3.733 usec
DE 18.00 usec
TE 234.0 K
D1 1.00000000 sec
TD0 1
SFO1 564.6299196 MHz
NUC1 ^{19}F
P1 15.00 usec
PLW1 27.71999931 W

F2 - Processing parameters
SI 65536
SF 564.6863882 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

Figure S59. $^{19}\text{F}\{^1\text{H}\}$ -NMR spectrum of compound **12**.

^{27}Al -NMR of $[\text{TbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_4)_4]$ in toluene- d_8 + 1,2-difluorobenzene at -40°C



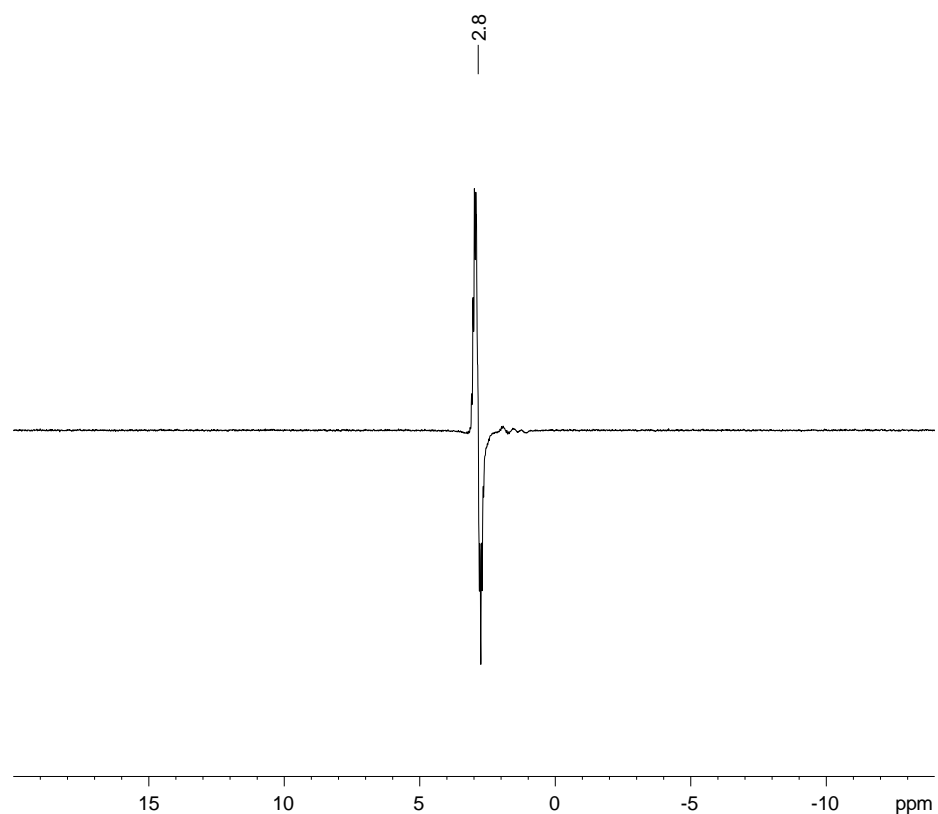
Current Data Parameters
NAME MA241_18022020_600TT
EXPNO 18
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200218
Time 14.45 h
INSTRUM spect
PROBHD Z126545_0027 (
PULPROG zg
TD 16384
SOLVENT Tol
NS 32
DS 4
SWH 62500.000 Hz
FIDRES 7.629395 Hz
AQ 0.1310720 sec
RG 189.6
DW 8.000 usec
DE 18.00 usec
TE 233.9 K
D1 1.00000000 sec
TD0 1
SFO1 156.3750279 MHz
NUC1 ^{27}Al
P1 11.50 usec
PLW1 60.00000000 W

F2 - Processing parameters
SI 32768
SF 156.3750279 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S60. ^{27}Al -NMR spectrum of compound **12**.

^{29}Si -INEPTND-NMR of $[\text{TbbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene at -40°C



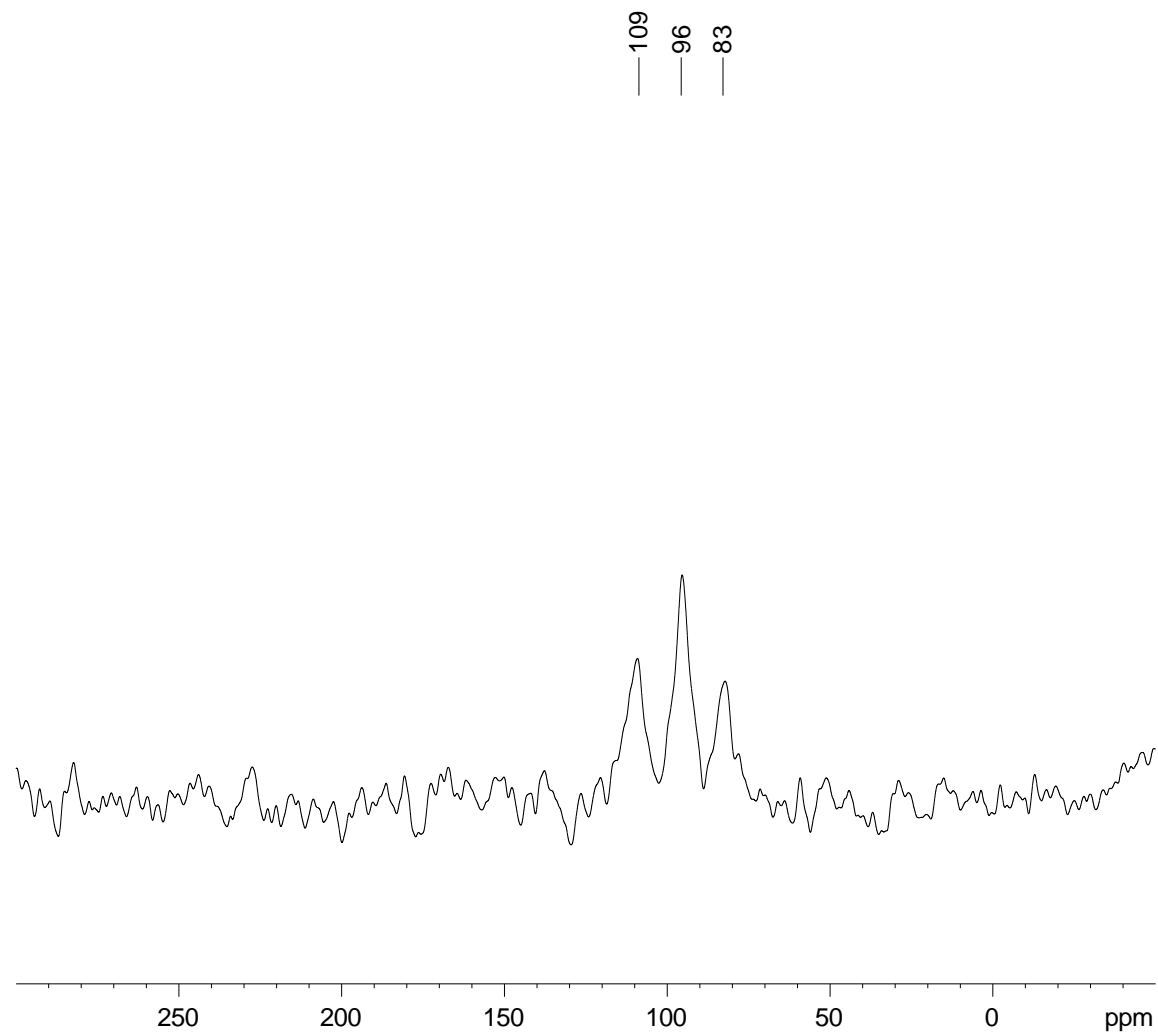
Current Data Parameters
NAME MA241_18022020_600TT
EXPNO 16
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200218
Time 14.40 h
INSTRUM spect
PROBHD Z126545_0027 (
PULPROG ineptnd
TD 16384
SOLVENT Tol
NS 128
DS 16
SWH 19132.652 Hz
FIDRES 2.335529 Hz
AQ 0.4281685 sec
RG 189.6
DW 26.133 usec
DE 18.00 usec
TE 233.9 K
CNST2 6.5999999
D1 5.0000000 sec
D4 0.03787879 sec
TD0 1
SFO1 119.2289493 MHz
NUC1 ^{29}Si
P1 14.50 usec
P2 29.00 usec
PLW1 50.0000000 W
SFO2 600.1324005 MHz
NUC2 ^1H
P3 12.00 usec
P4 24.00 usec
PLW2 23.41200066 W

F2 - Processing parameters
SI 32768
SF 119.2289493 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 4.00

Figure S61. ^{29}Si -INEPTND-NMR spectrum of compound **12**.

^{119}Sn -NMR of $[\text{TbbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene at $-40\text{ }^\circ\text{C}$



Current Data Parameters
NAME MA241_17022020_500TT
EXPNO 8
PROCNO 1

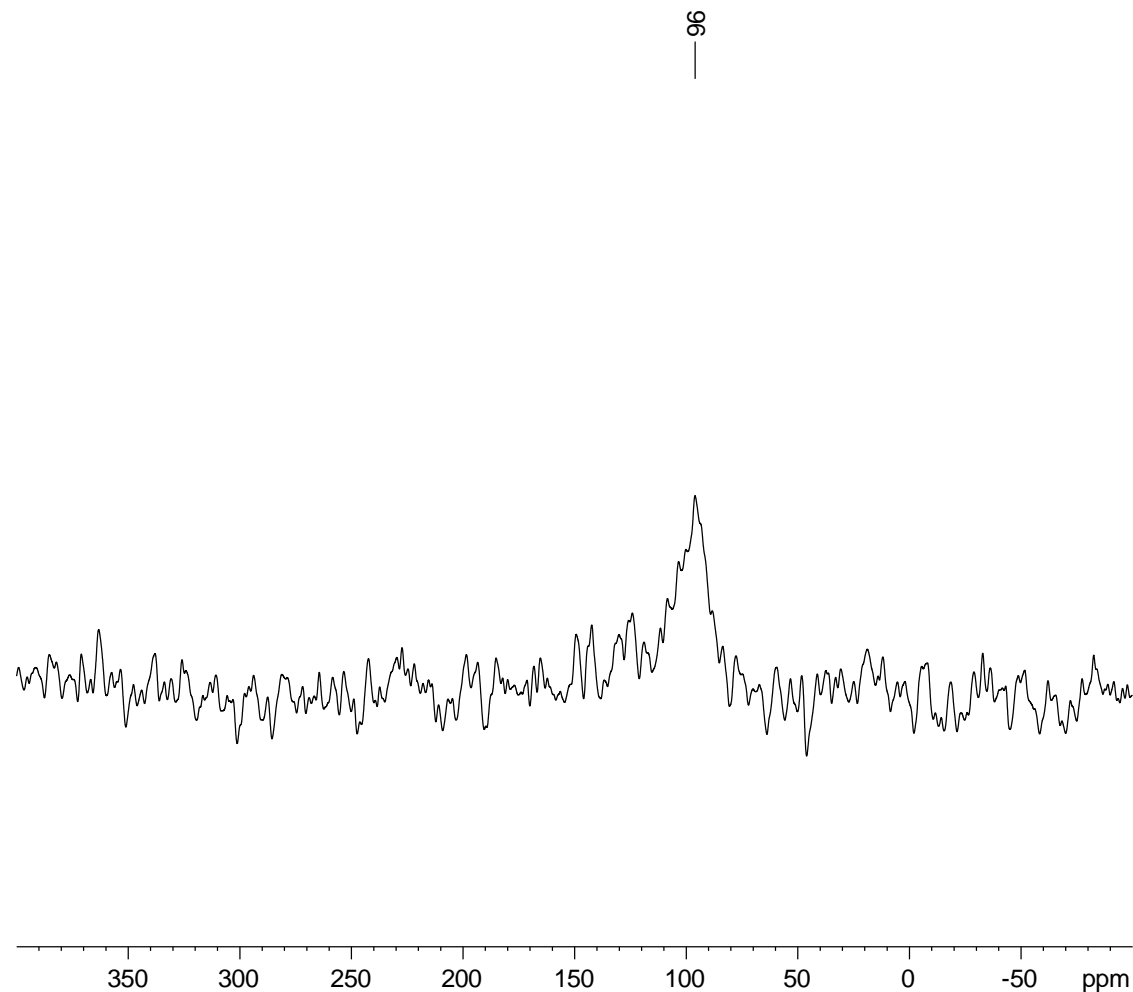
F2 - Acquisition Parameters
Date_ 20200217
Time 12.14
INSTRUM spect
PROBHD 5 mm TBO BB-1H
PULPROG zg30
TD 65536
SOLVENT Tol
NS 32893
DS 0
SWH 93750.000 Hz
FIDRES 1.430511 Hz
AQ 0.3495253 sec
RG 2050
DW 5.333 usec
DE 6.00 usec
TE 233.1 K
D1 0.20000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 119Sn
P1 11.93 usec
PL1 3.00 dB
PL1W 45.32131577 W
SFO1 186.5389383 MHz

F2 - Processing parameters
SI 131072
SF 186.5016380 MHz
WDW EM
SSB 0
LB 350.00 Hz
GB 0
PC 1.40

Figure S62. ^{119}Sn -NMR spectrum of compound **12**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene at -40°C



Current Data Parameters
NAME MA241_24022020_500
EXPNO 5
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200224
Time 15.11
INSTRUM spect
PROBHD 5 mm TBO BB-1H
PULPROG zgig30
TD 65536
SOLVENT Tol
NS 16279
DS 0
SWH 150000.000 Hz
FIDRES 2.288818 Hz
AQ 0.2184533 sec
RG 2050
DW 3.333 usec
DE 6.00 usec
TE 233.4 K
D1 0.20000000 sec
D11 0.03000000 sec
TD0 1

==== CHANNEL f1 =====
NUC1 119Sn
P1 11.93 usec
PL1 3.00 dB
PL1W 45.32131577 W
SFO1 186.5296132 MHz

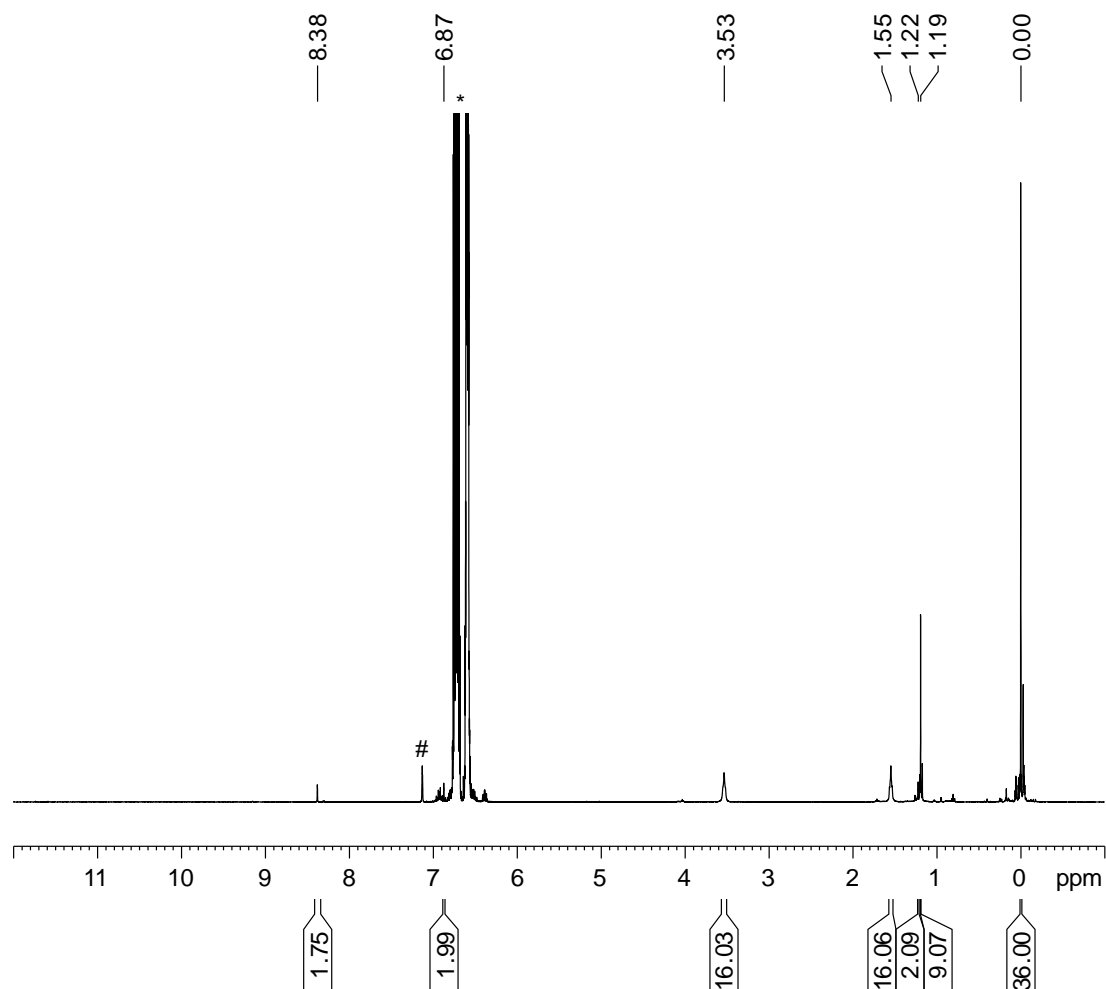
==== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -0.52 dB
PL12 15.00 dB
PL2W 24.34997177 W
PL12W 0.68312228 W
SFO2 500.1320005 MHz

F2 - Processing parameters
SI 65536
SF 186.5016380 MHz
WDW EM
SSB 0
LB 350.00 Hz
GB 0
PC 1.40

Figure S63. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **12**.

NMR spectra of compound **13**

^1H -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in C_6D_6 (#) + 1,2-difluorobenzene (*) at rt



Current Data Parameters
NAME MA246_08042020_400N
EXPNO 16
PROCNO 1

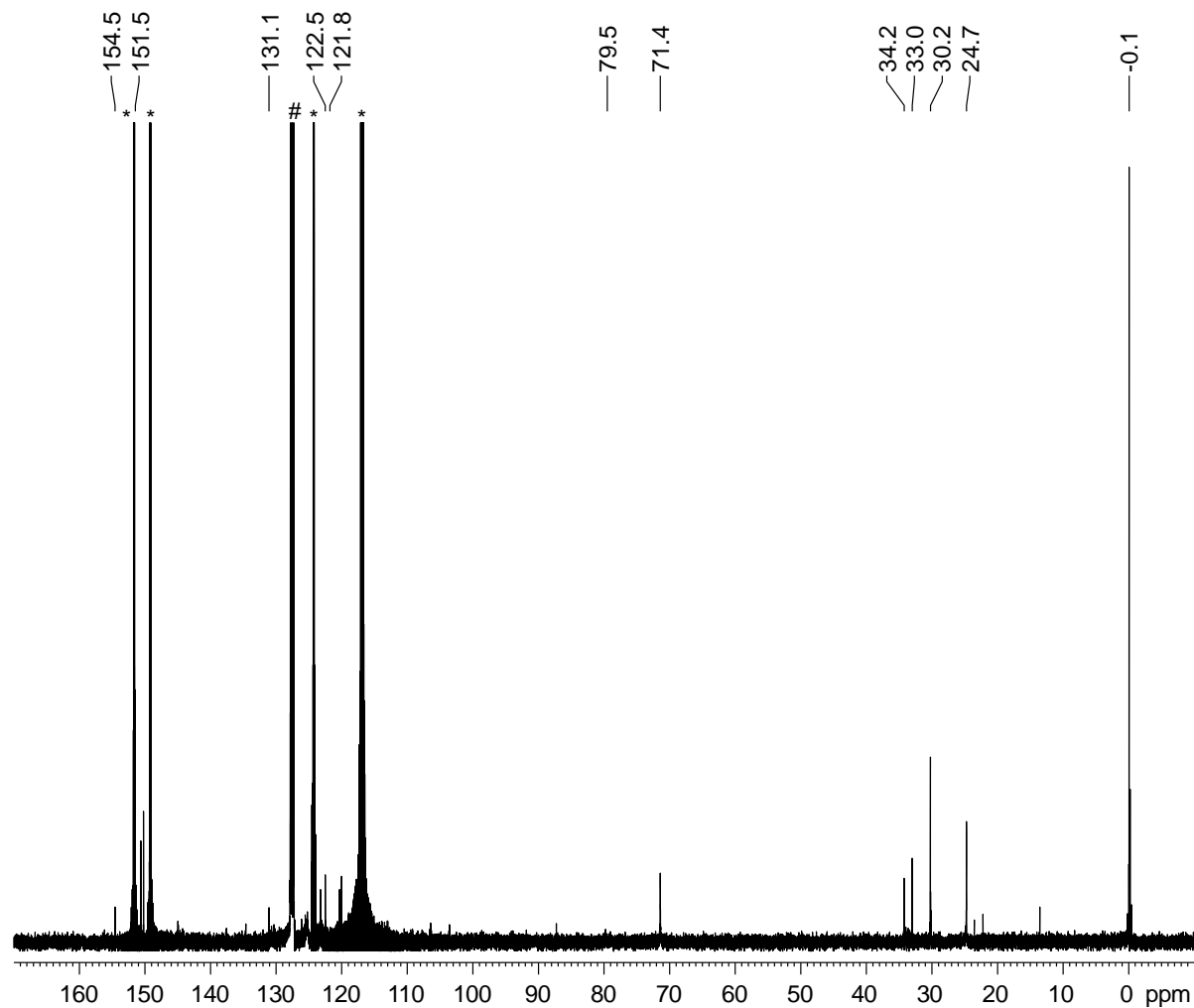
F2 - Acquisition Parameters
Date_ 20200409
Time 7.33
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zg30
TD 52656
SOLVENT C6D6
NS 32
DS 0
SWH 8305.647 Hz
FIDRES 0.157734 Hz
AQ 3.1698911 sec
RG 71.8
DW 60.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
TD0 1

==== CHANNEL f1 =====
NUC1 1H
P1 14.60 usec
PL1 -3.00 dB
PL1W 16.03799057 W
SFO1 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 400.1100032 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

Figure S64. ^1H -NMR spectrum of compound **13**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in C_6D_6 (#) + 1,2-difluorobenzene (*) at rt



Current Data Parameters
 NAME MA246_08042020_400N
 EXPNO 11
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 20200409
 Time 2.07
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zgpg30
 TD 53700
 SOLVENT C6D6
 NS 11264
 DS 0
 SWH 30864.197 Hz
 FIDRES 0.574752 Hz
 AQ 0.8699400 sec
 RG 32800
 DW 16.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 D11 0.03000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 13C
 P1 13.50 usec
 PL1 -4.16 dB
 PL1W 78.55633545 W
 SFO1 100.6198135 MHz

===== CHANNEL f2 =====
 CPDPRG[2] waltz16
 NUC2 1H
 PCPD2 80.00 usec
 PL2 -3.00 dB
 PL12 11.77 dB
 PL13 13.14 dB
 PL2W 16.03799057 W
 PL12W 0.53474891 W
 PL13W 0.39007664 W
 SFO2 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 100.6077400 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1 40

Figure S65. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **13**.

$^{19}\text{F}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in C_6D_6 + 1,2-difluorobenzene (#) at rt

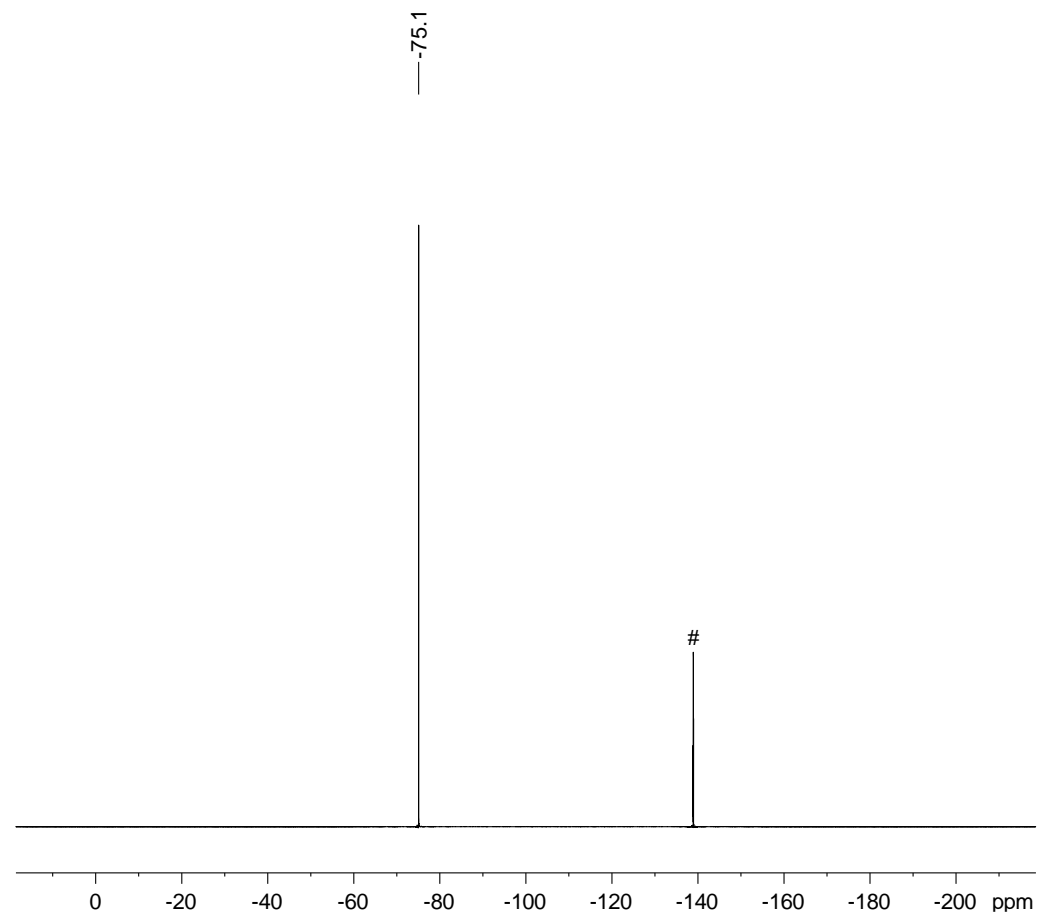


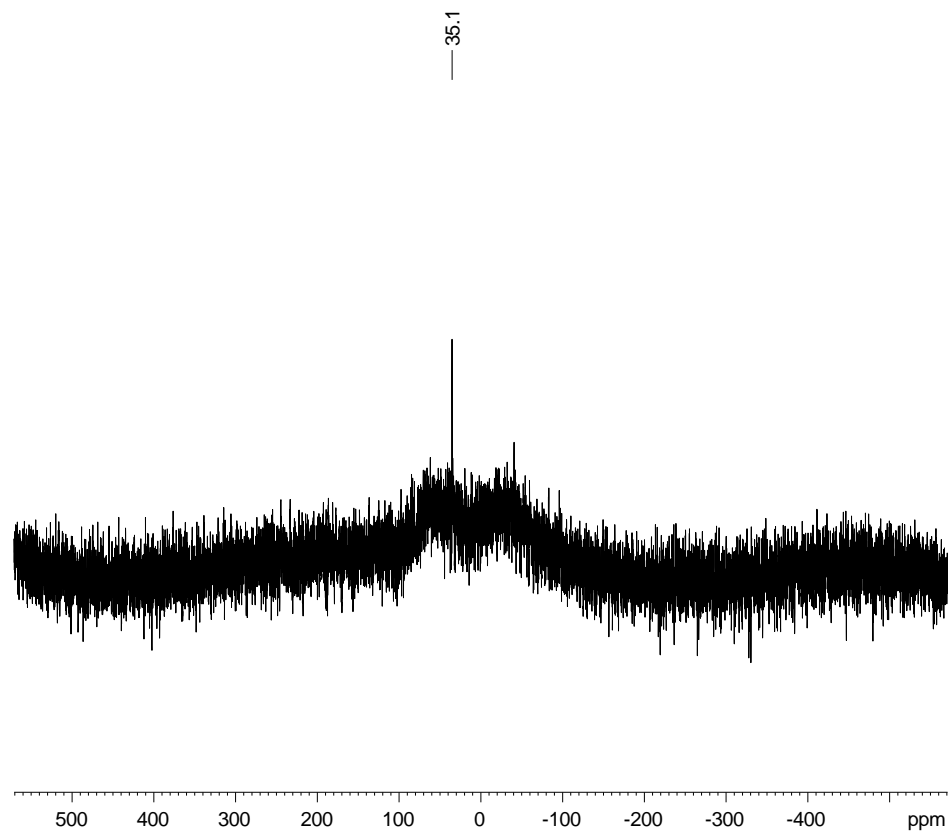
Figure S66. $^{19}\text{F}\{^1\text{H}\}$ -NMR spectrum of compound **13**.

Current Data Parameters
NAME MA246_08042020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200408
Time 8.06 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgbs
TD 262144
SOLVENT C6D6
NS 16
DS 4
SWH 66964.289 Hz
FIDRES 0.510897 Hz
AQ 1.9573419 sec
RG 204.67
DW 7.467 usec
DE 6.50 usec
TE 298.0 K
D1 1.00000000 sec
TD0 1
SFO1 282.3761148 MHz
NUC1 19F
P1 15.00 usec
P2 30.00 usec
PLW1 6.86250019 W

F2 - Processing parameters
SI 65536
SF 282.4043552 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

^{27}Al -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in C_6D_6 + 1,2-difluorobenzene at rt



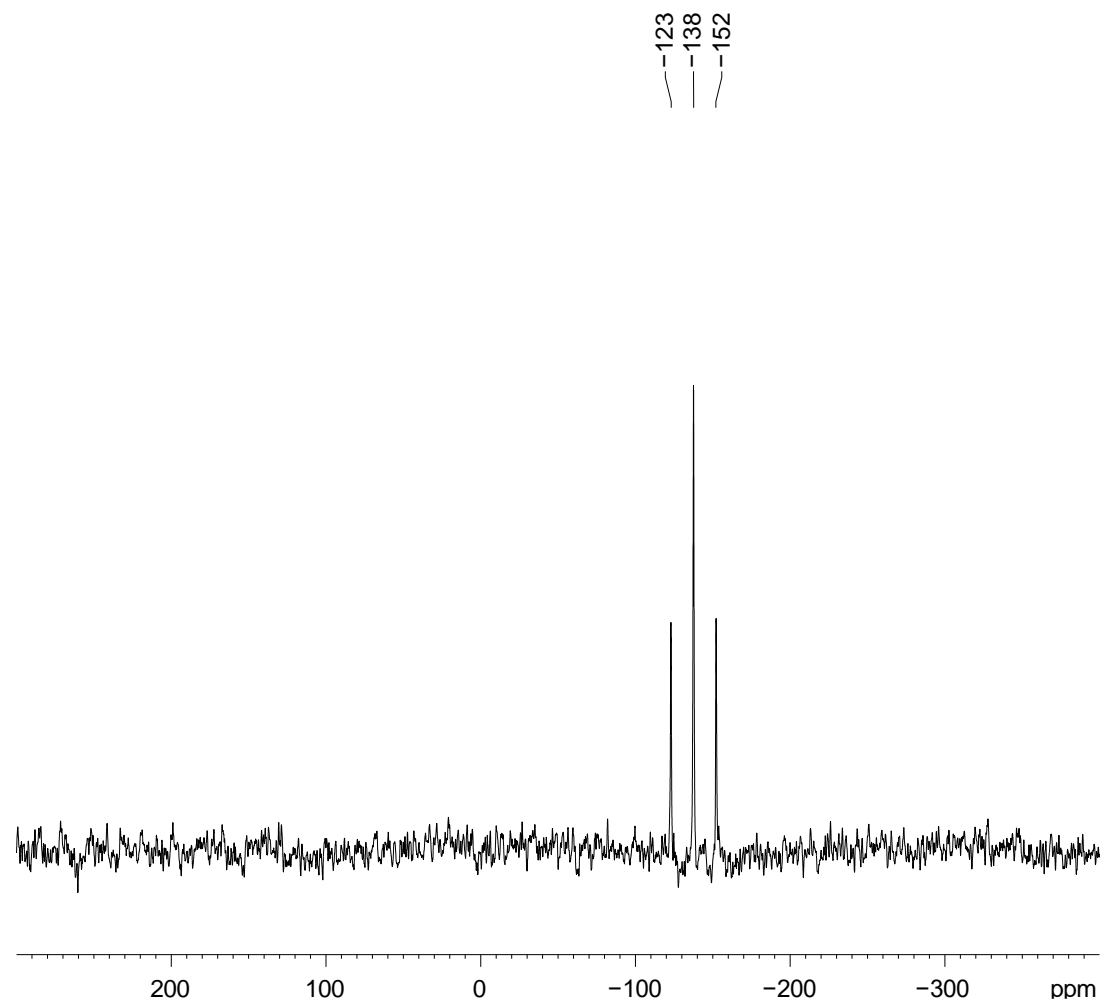
Current Data Parameters
NAME MA246_08042020_300
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200408
Time 8.08 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 16384
SOLVENT C6D6
NS 512
DS 0
SWH 89285.711 Hz
FIDRES 10.899135 Hz
AQ 0.0917504 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
TD0 1
SFO1 78.2044509 MHz
NUC1 27Al
P0 3.40 usec
P1 10.19 usec
PLW1 34.00000000 W

F2 - Processing parameters
SI 32768
SF 78.2044509 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S67. ^{27}Al -NMR spectrum of compound **13**.

^{119}Sn -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene at rt



Current Data Parameters
NAME MA242_24022020_500
EXPNO 2
PROCNO 1

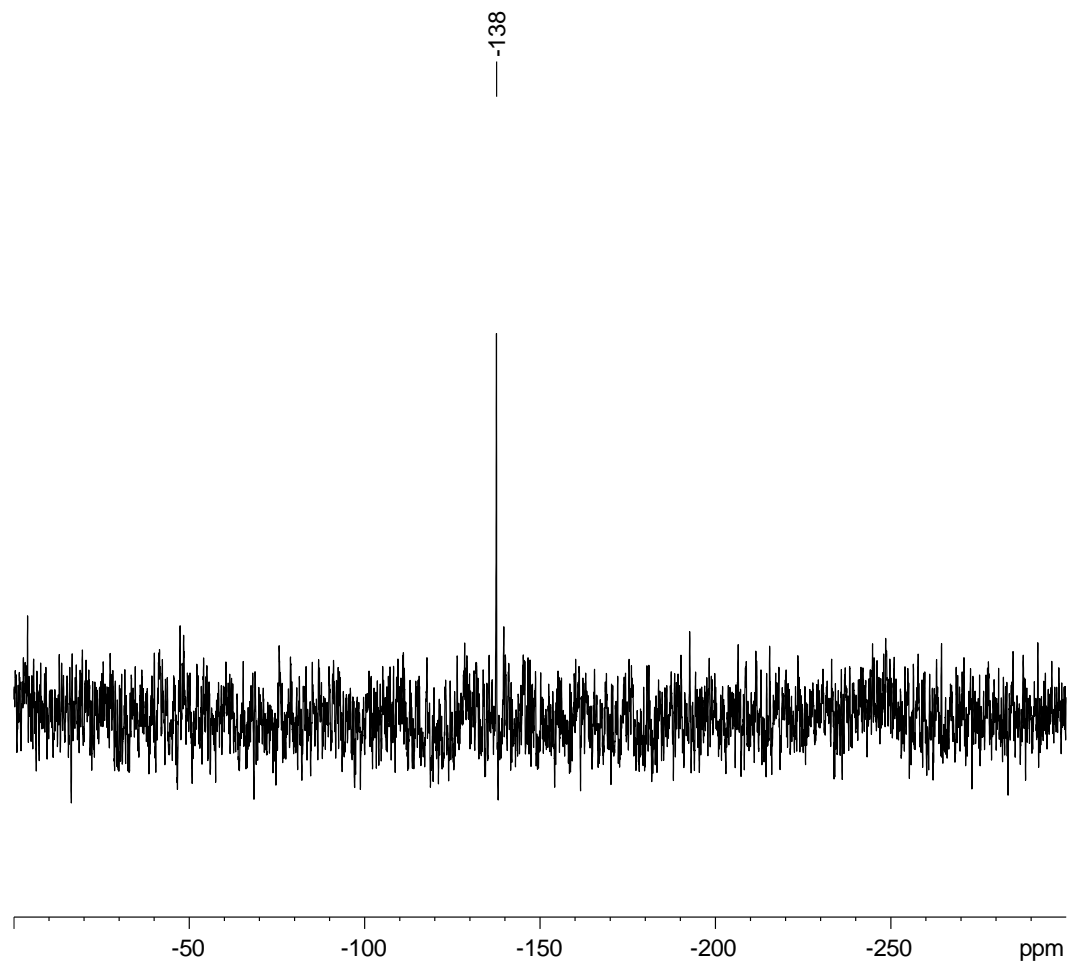
F2 - Acquisition Parameters
Date_ 20200225
Time 1.55
INSTRUM spect
PROBHD 5 mm TBO BB-1H
PULPROG zg30
TD 65536
SOLVENT Tol
NS 65000
DS 0
SWH 150000.000 Hz
FIDRES 2.288818 Hz
AQ 0.2184533 sec
RG 2050
DW 3.333 usec
DE 6.00 usec
TE 299.2 K
D1 0.20000000 sec
TD0 1

==== CHANNEL f1 =====
NUC1 119Sn
P1 11.93 usec
PL1 3.00 dB
PL1W 45.32131577 W
SFO1 186.5016380 MHz

F2 - Processing parameters
SI 131072
SF 186.5016380 MHz
WDW EM
SSB 0
LB 100.00 Hz
GB 0
PC 1.40

Figure S68. ^{119}Sn -NMR spectrum of compound **13**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2(\text{thf})_2][\text{Al}(\text{OC}(\text{CF}_3)_3)_4]$ in toluene- d_8 + 1,2-difluorobenzene at rt



Current Data Parameters
NAME MA242_24022020_500
EXPNO 3
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200225
Time 8.12
INSTRUM spect
PROBHD 5 mm TBO BB-1H
PULPROG zgig30
TD 65536
SOLVENT Tol
NS 50000
DS 0
SWH 150000.000 Hz
FIDRES 2.288818 Hz
AQ 0.2184533 sec
RG 2050
DW 3.333 usec
DE 6.00 usec
TE 299.2 K
D1 0.20000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 119Sn
P1 11.93 usec
PL1 3.00 dB
PL1W 45.32131577 W
SFO1 186.5016380 MHz

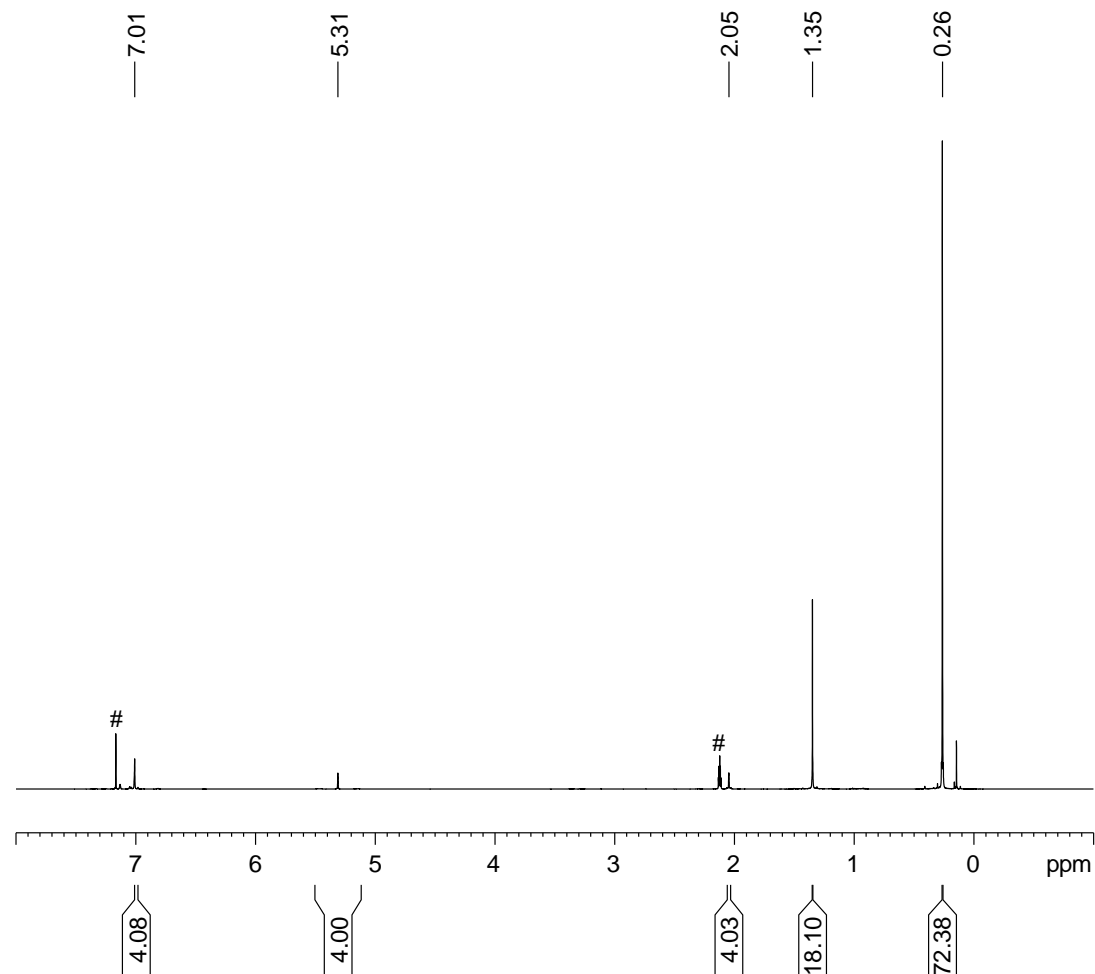
===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -0.52 dB
PL12 15.00 dB
PL2W 24.34997177 W
PL12W 0.68312228 W
SFO2 500.1320005 MHz

F2 - Processing parameters
SI 65536
SF 186.5016380 MHz
WDW EM
SSB 0
LB 50.00 Hz
GB 0
PC 1.40

Figure S69. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **13**.

NMR spectra of compound **14**

$^1\text{H-NMR}$ of $[\text{TbbSnH}_2]_2$ in toluene- d_8 (#) at rt



Current Data Parameters
 NAME MA263_06042020_400N
 EXPNO 11
 PROCNO 1

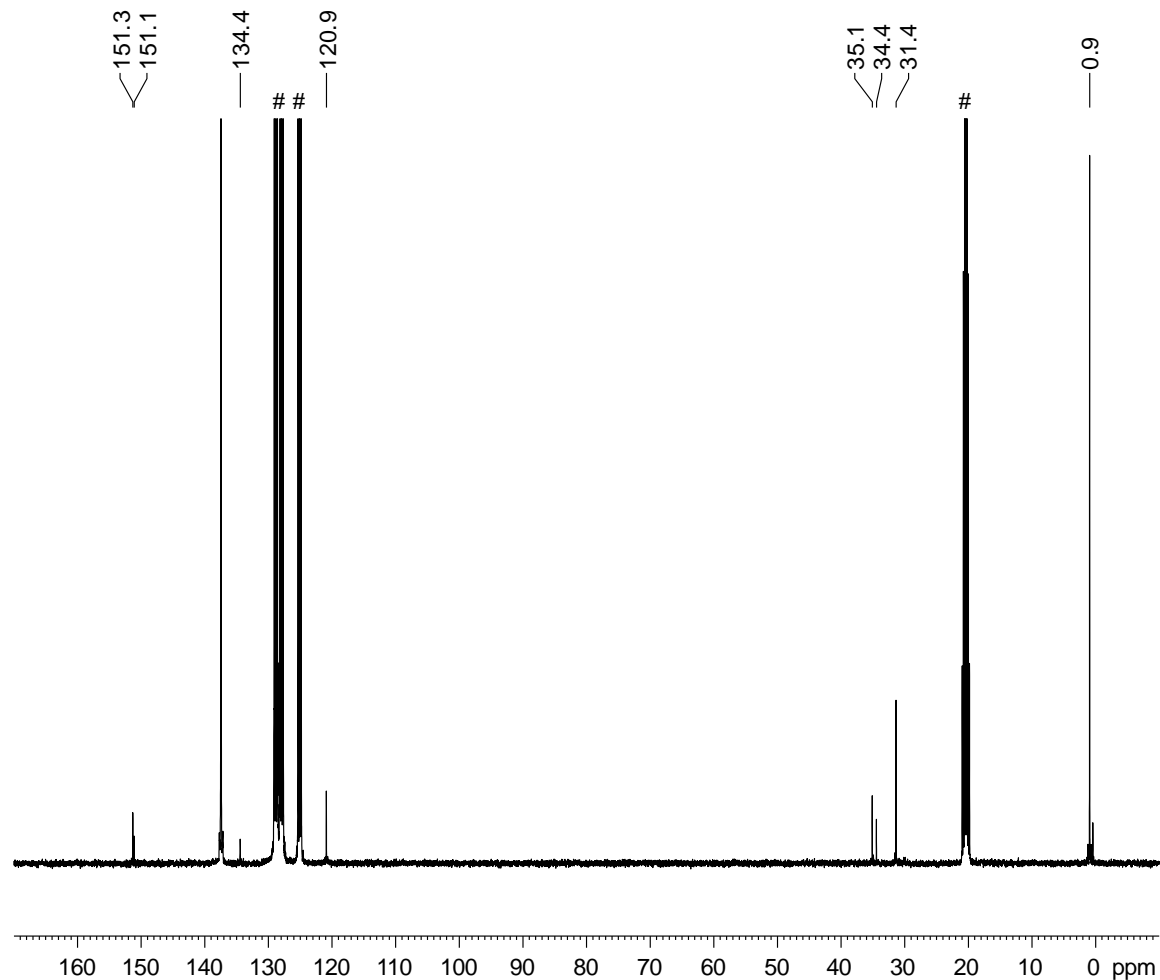
F2 - Acquisition Parameters
 Date_ 20200406
 Time 20.05
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT Tol
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 287
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

==== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S70. $^1\text{H-NMR}$ spectrum of compound **14**.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2]_2$ in toluene- d_8 (#) at rt



Current Data Parameters
NAME MA263_06042020_400N
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200407
Time 2.09
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT Tol
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

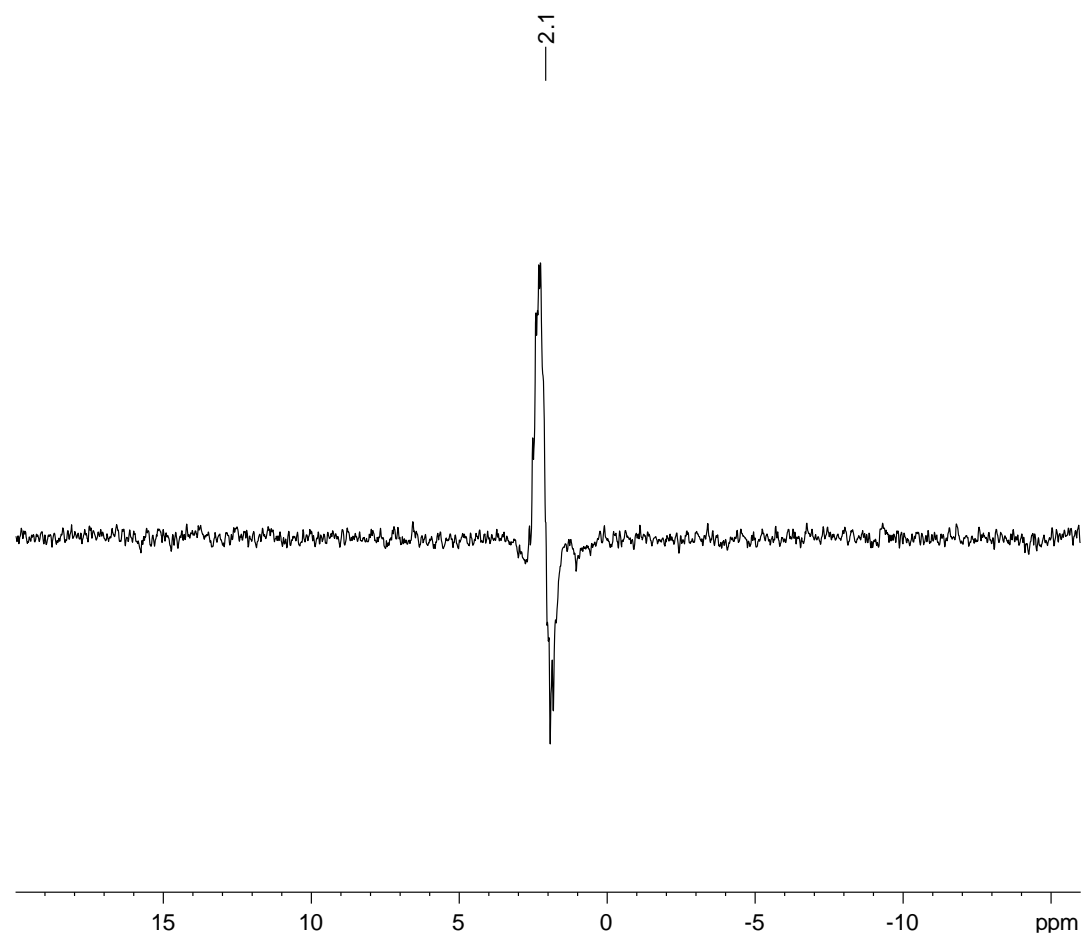
===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077048 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S71. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **14**.

^{29}Si -INEPTND-NMR of $[\text{TbbSnH}_2]_2$ in toluene- d_8 at rt



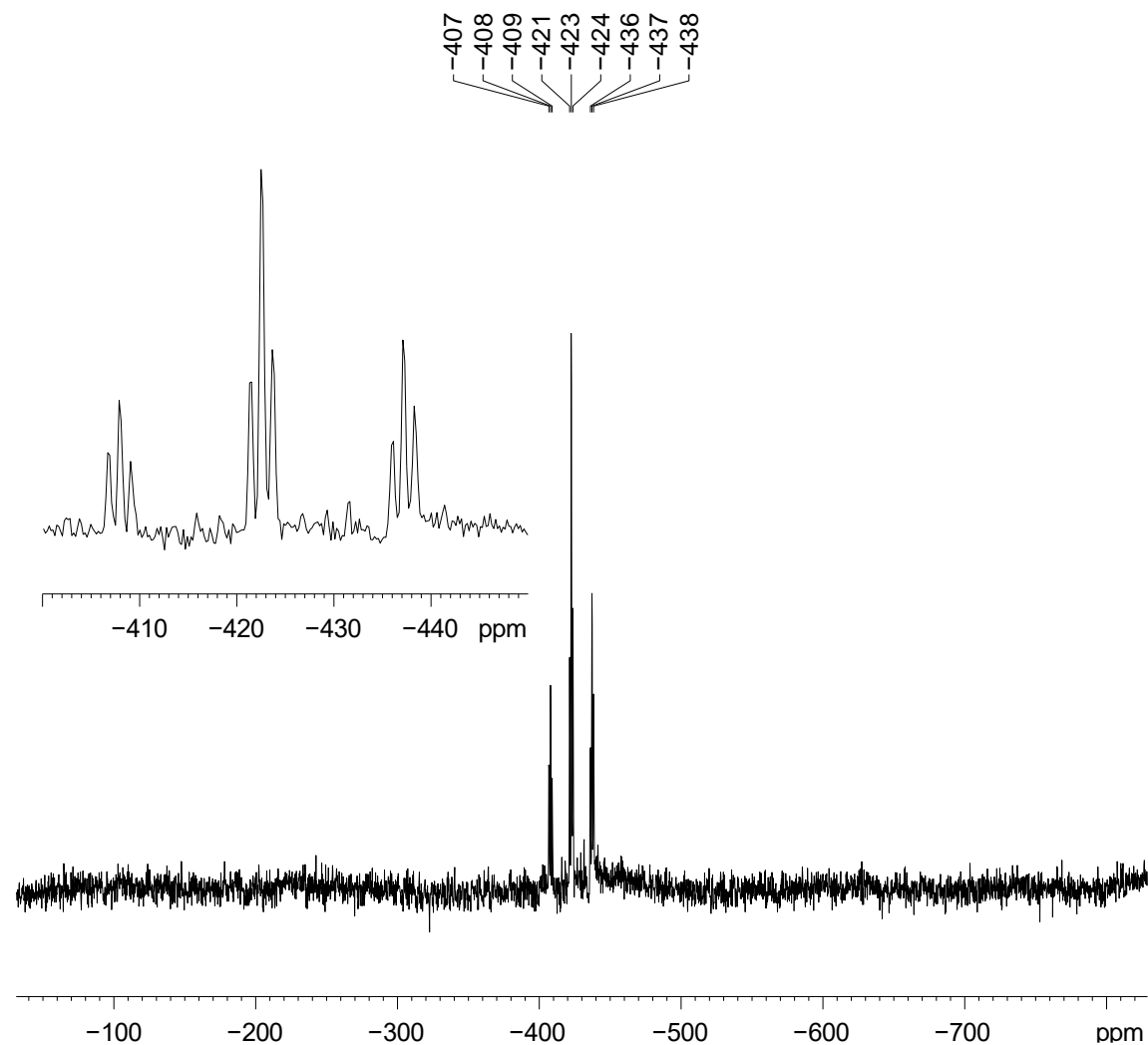
Current Data Parameters
NAME MA263_07042020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200407
Time 13.00 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT Tol
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.0000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 ^{29}Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273880 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S72. ^{29}Si -INEPTND-NMR spectrum of compound **14**.

^{119}Sn -NMR of $[\text{TbbSnH}_2]_2$ in thf-d_8 at rt



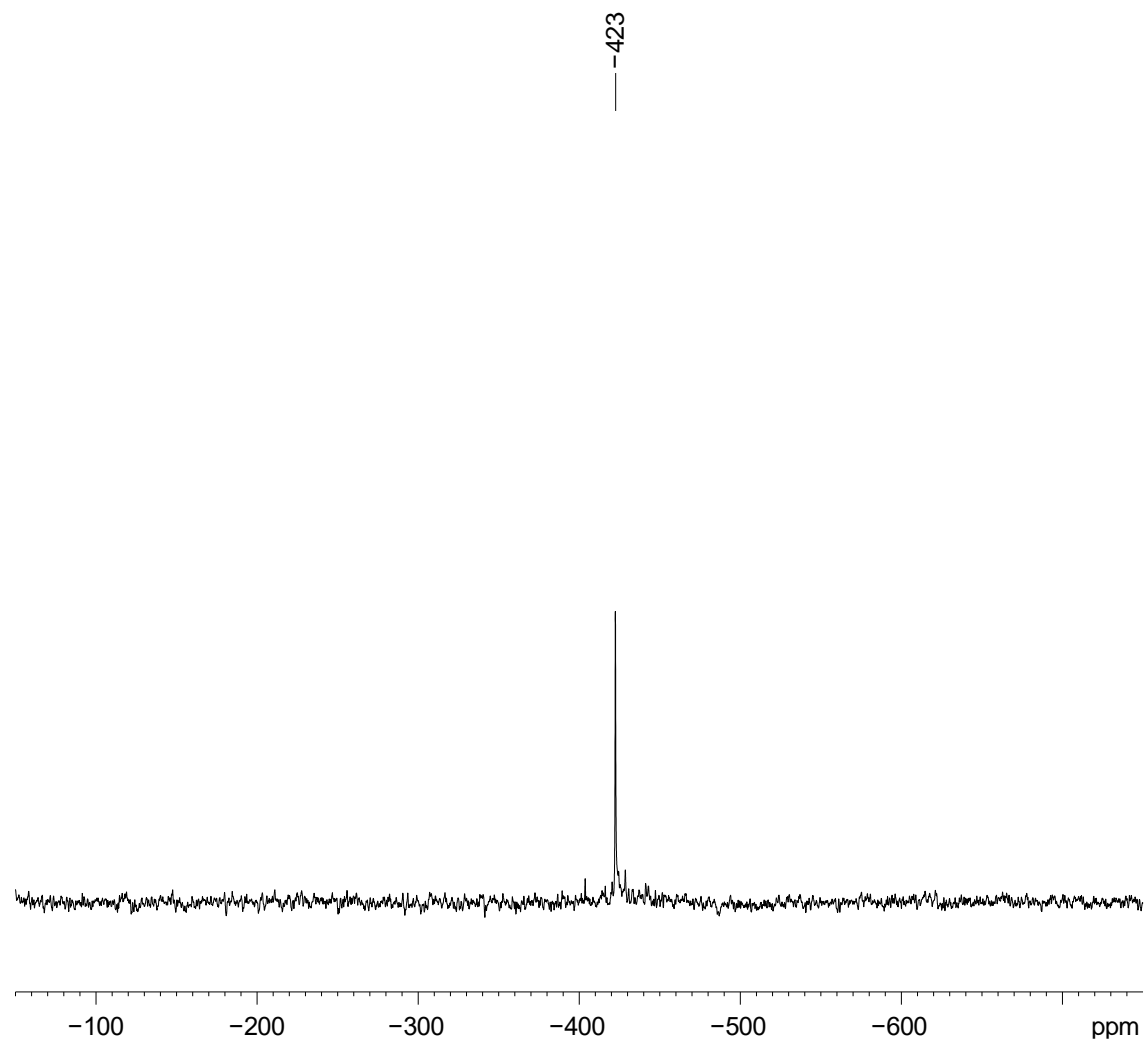
Current Data Parameters
NAME MA815_03022022_300N
EXPNO 24
PROCNO 1

F2 - Acquisition Parameters
Date_ 20220203
Time 20.35 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT THF
NS 204800
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8722484 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 10.00 Hz
GB 0
PC 1.40

Figure S73. ^{119}Sn -NMR spectrum of compound **14**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $[\text{TbbSnH}_2]_2$ in thf-d_8 at rt



Current Data Parameters
NAME MA815_03022022_300N
EXPNO 34
PROCNO 1

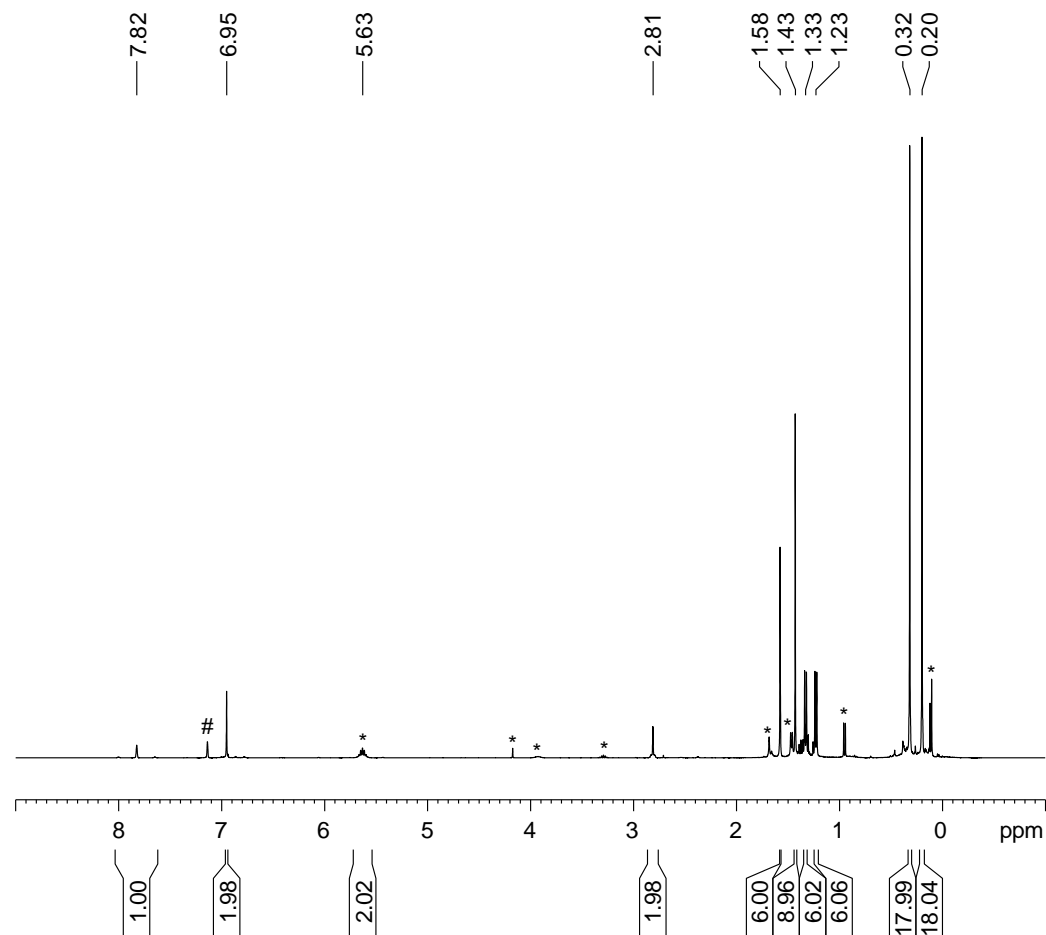
F2 - Acquisition Parameters
Date_ 20220204
Time 2.26 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT THF
NS 57344
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.8722482 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz

Figure S74. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **14**.

NMR spectra of compound 15

$^1\text{H-NMR}$ of $\text{TbbSnH}(\textit{i}\text{Pr-NHC})$ in C_6D_6 (#) at rt, *: $\textit{i}\text{Pr-NHC-H}_2$ + unknown impurity



Current Data Parameters
 NAME MA305_19052020_400N
 EXPNO 10
 PROCNO 1

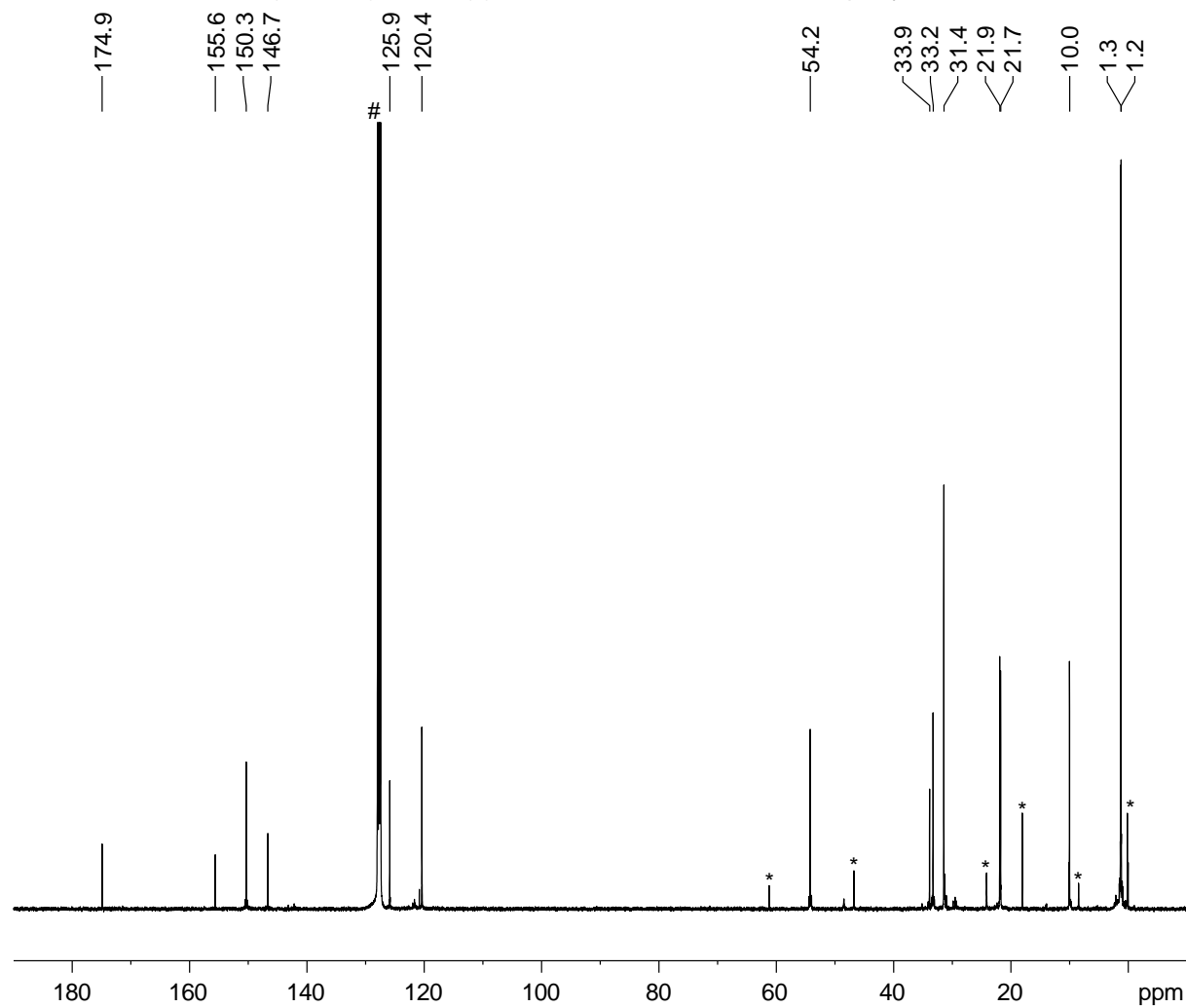
F2 - Acquisition Parameters
 Date_ 20200519
 Time 12.56
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 64
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TD0 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100011 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S75. $^1\text{H-NMR}$ spectrum of compound 15.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of TbbSnH(*i*Pr-NHC) in C_6D_6 (#) at rt, *: *i*Pr-NHC- H_2 + unknown impurity



Current Data Parameters
NAME MA305_19052020_400N
EXPNO 12
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200520
Time 2.08
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 11264
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

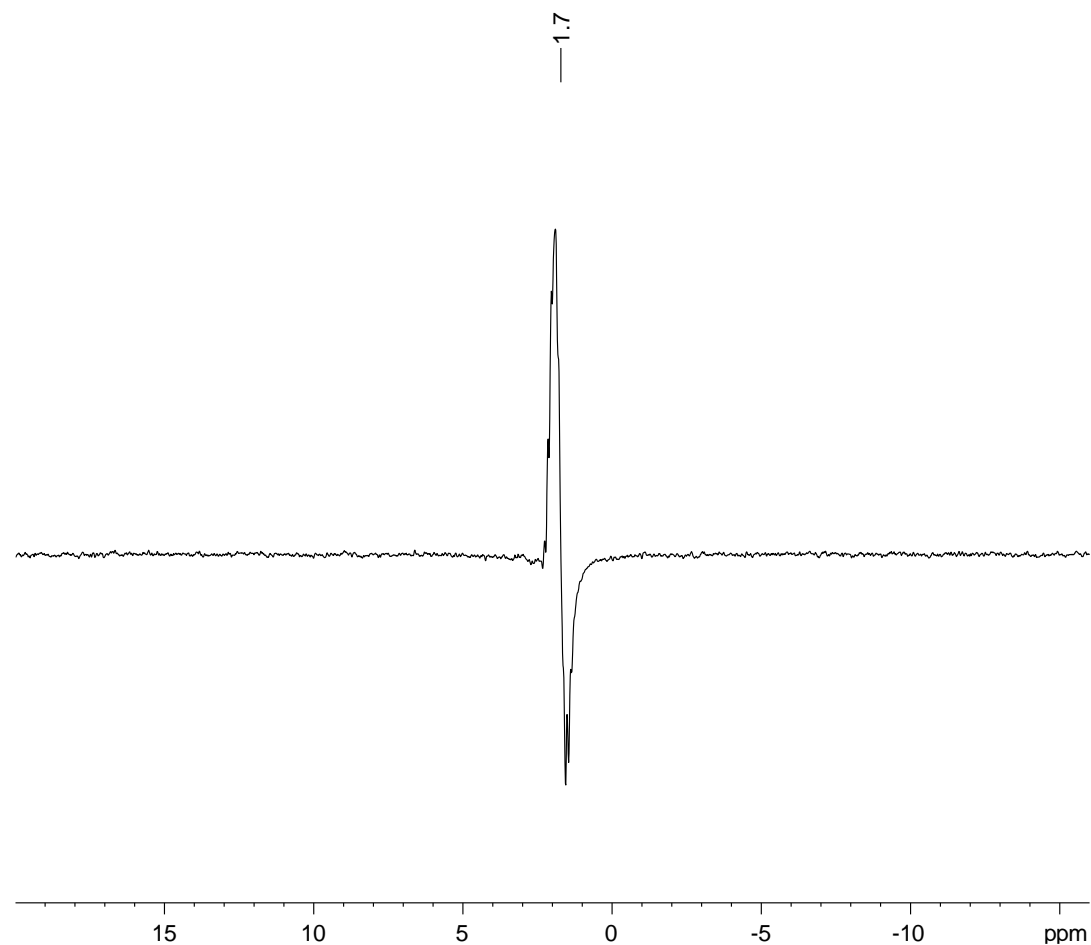
==== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

==== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077392 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S76. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **15**.

²⁹Si-INEPTND-NMR of TbbSnH(*i*Pr-NHC) in C₆D₆ at rt



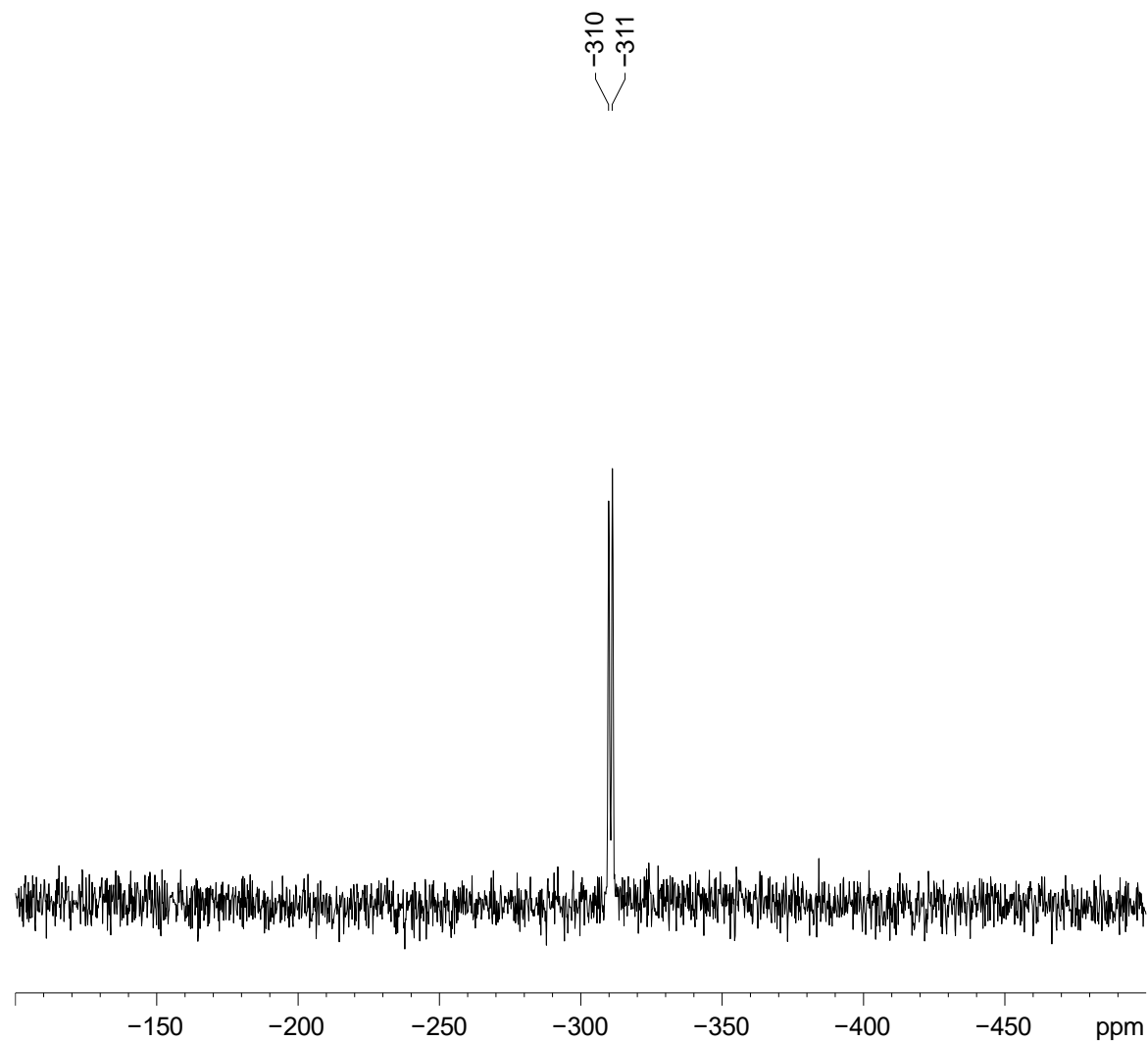
Current Data Parameters
NAME MA305_20052020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200520
Time 8.48 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.0000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.6229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273589 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S77. ²⁹Si-INEPTND-NMR spectrum of compound **15**.

^{119}Sn -NMR of $\text{TbbSnH}(\text{/Pr-NHC})$ in C_6D_6 at rt



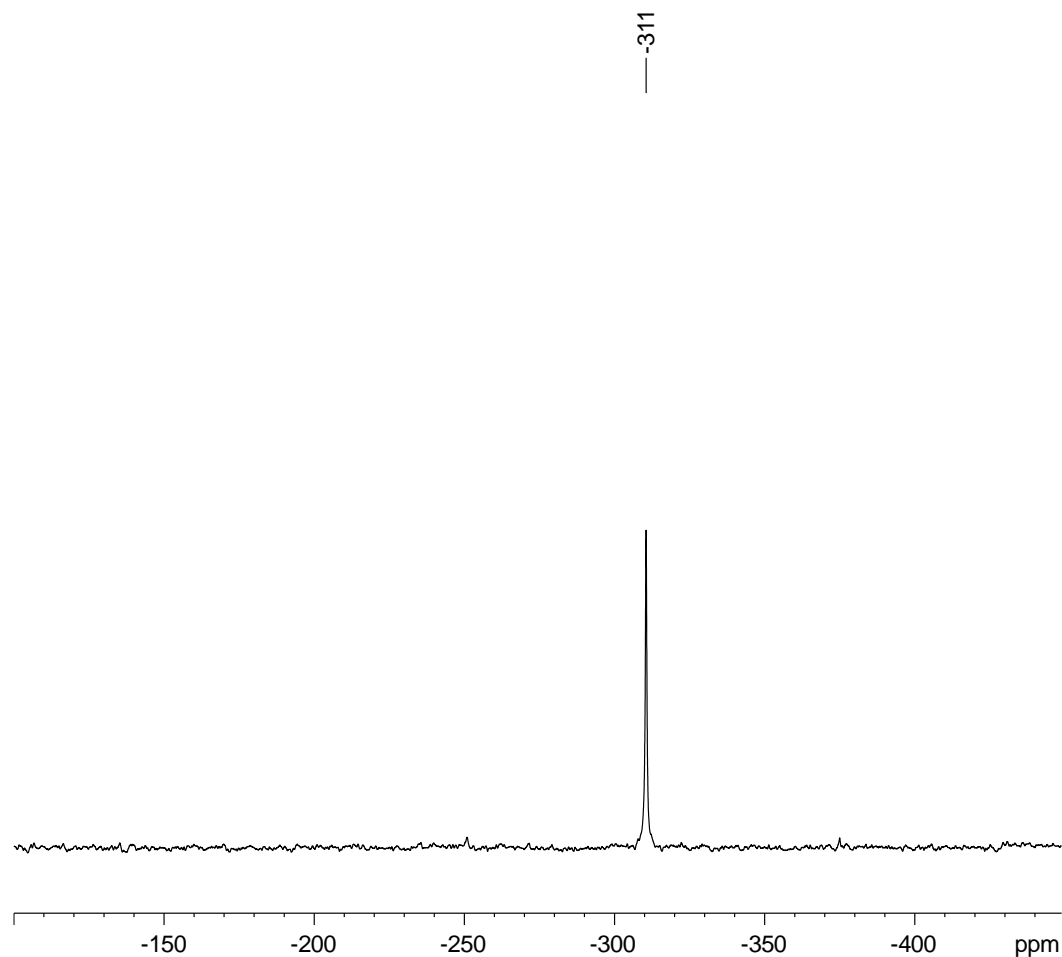
Current Data Parameters
NAME MA236_05022020_300
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20200205
Time 14.12 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zg30
TD 8918
SOLVENT C6D6
NS 16384
DS 1
SWH 89285.711 Hz
FIDRES 20.023708 Hz
AQ 0.0499408 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.02000000 sec
TD0 1
SFO1 111.8867979 MHz
NUC1 ^{119}Sn
P1 12.10 usec
PLW1 12.00000000 W

F2 - Processing parameters
SI 4096
SF 111.9203740 MHz
WDW EM
SSB 0
LB 5.00 Hz
GB 0
PC 1.40

Figure S78. ^{119}Sn -NMR spectrum of compound **15**.

$^{119}\text{Sn}\{^1\text{H}\}$ -NMR of $\text{TbbSnH}(\text{iPr-NHC})$ in C_6D_6 at rt



Current Data Parameters
NAME MA305_20052020_300
EXPNO 20
PROCNO 1

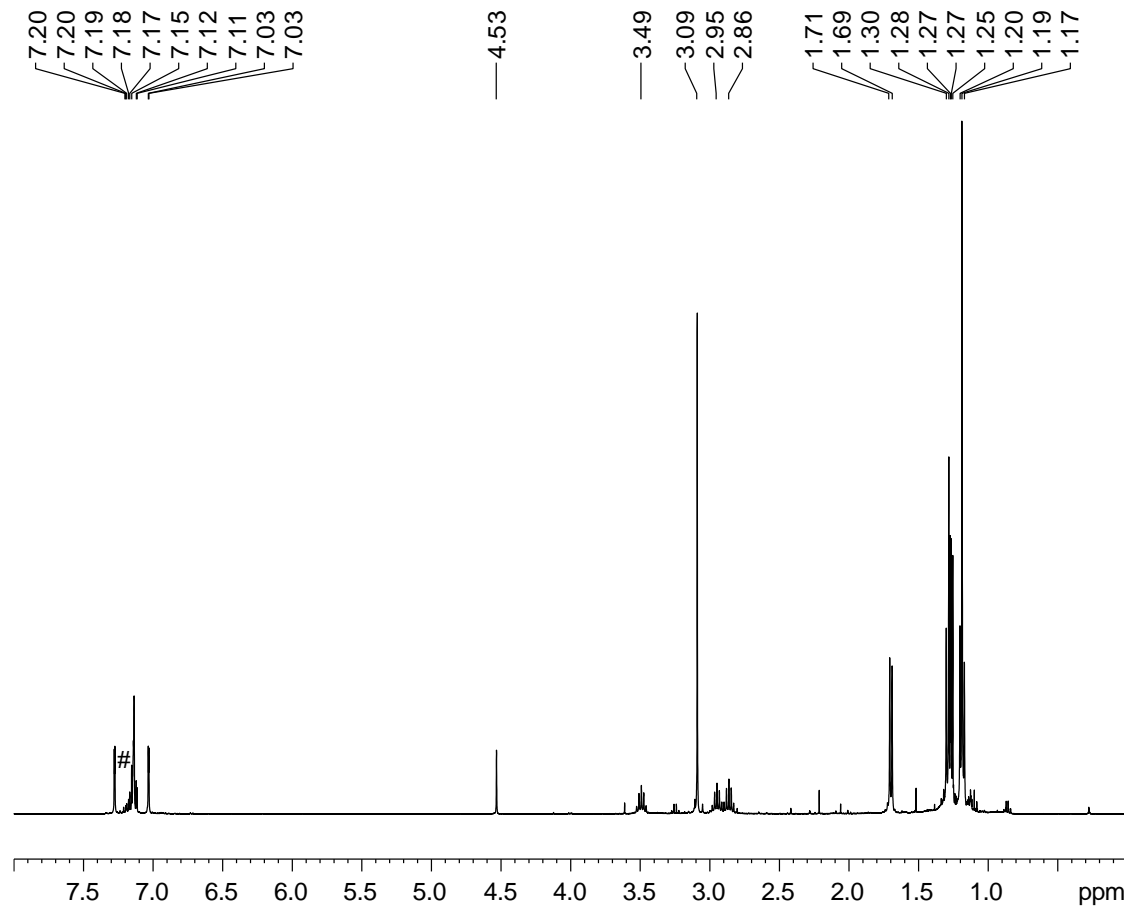
F2 - Acquisition Parameters
Date_ 20200520
Time 10.43 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG zgig30
TD 39186
SOLVENT C6D6
NS 5120
DS 4
SWH 89285.711 Hz
FIDRES 4.557021 Hz
AQ 0.2194416 sec
RG 204.67
DW 5.600 usec
DE 6.50 usec
TE 298.0 K
D1 0.10000000 sec
D11 0.03000000 sec
TD0 1
SFO1 111.9147778 MHz
NUC1 ^{119}Sn
P0 4.03 usec
P1 12.10 usec
PLW1 12.00000000 W
SFO2 300.1312005 MHz
NUC2 ^1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 65536
SF 111.9203738 MHz
WDW EM
SSB 0
LB 50.00 Hz
GB 0
PC 1.40

Figure S79. $^{119}\text{Sn}\{^1\text{H}\}$ -NMR spectrum of compound **15**.

NMR spectra of compound 16

$^1\text{H-NMR}$ of $\text{Ar}^*\text{GeH}(\text{Me-NHC})$ in C_6D_6 (#) at rt



Current Data Parameters
 NAME FD033_9-20160909-LW_Diab
 EXPNO 10
 PROCNO 1

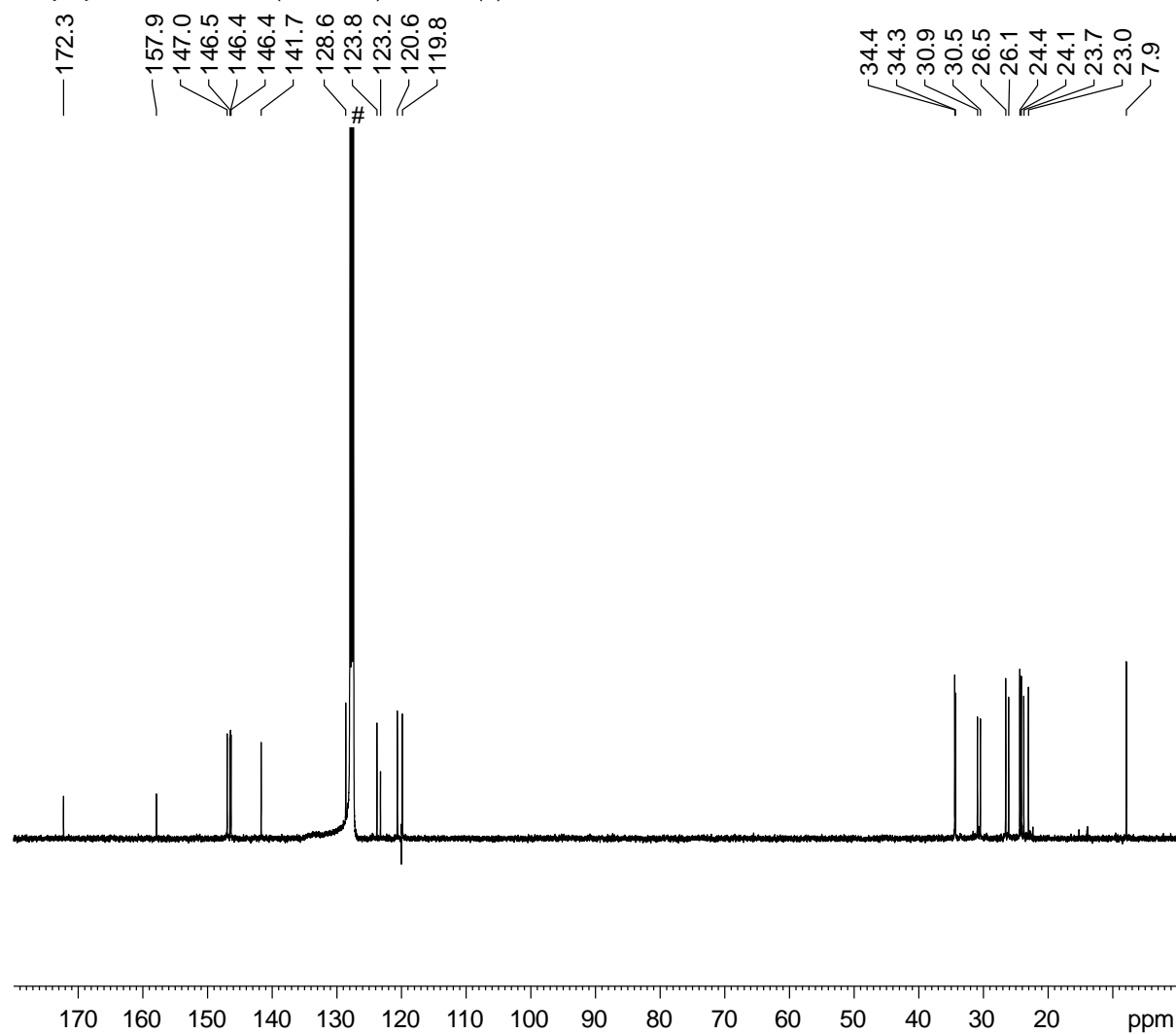
F2 - Acquisition Parameters
 Date_ 20160909
 Time 20.06
 INSTRUM spect
 PROBHD 5 mm QNP 1H/13
 PULPROG zg30
 TD 52656
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 8305.647 Hz
 FIDRES 0.157734 Hz
 AQ 3.1698911 sec
 RG 228
 DW 60.200 usec
 DE 6.00 usec
 TE 299.2 K
 D1 1.00000000 sec
 TDO 1

===== CHANNEL f1 =====
 NUC1 1H
 P1 14.60 usec
 PL1 -3.00 dB
 PL1W 16.03799057 W
 SFO1 400.1120007 MHz

F2 - Processing parameters
 SI 65536
 SF 400.1100000 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S80. $^1\text{H-NMR}$ spectrum of compound 16.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of Ar*GeH(Me-NHC) in C_6D_6 (#) at rt



Current Data Parameters
NAME FD033_9-20160909-LW_Diab
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20160910
Time 5.14
INSTRUM spect
PROBHD 5 mm QNP 1H/13
PULPROG zgpg30
TD 53700
SOLVENT C6D6
NS 17000
DS 0
SWH 30864.197 Hz
FIDRES 0.574752 Hz
AQ 0.8699400 sec
RG 32800
DW 16.200 usec
DE 6.00 usec
TE 299.2 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 13C
P1 13.50 usec
PL1 -4.16 dB
PL1W 78.55633545 W
SFO1 100.6198135 MHz

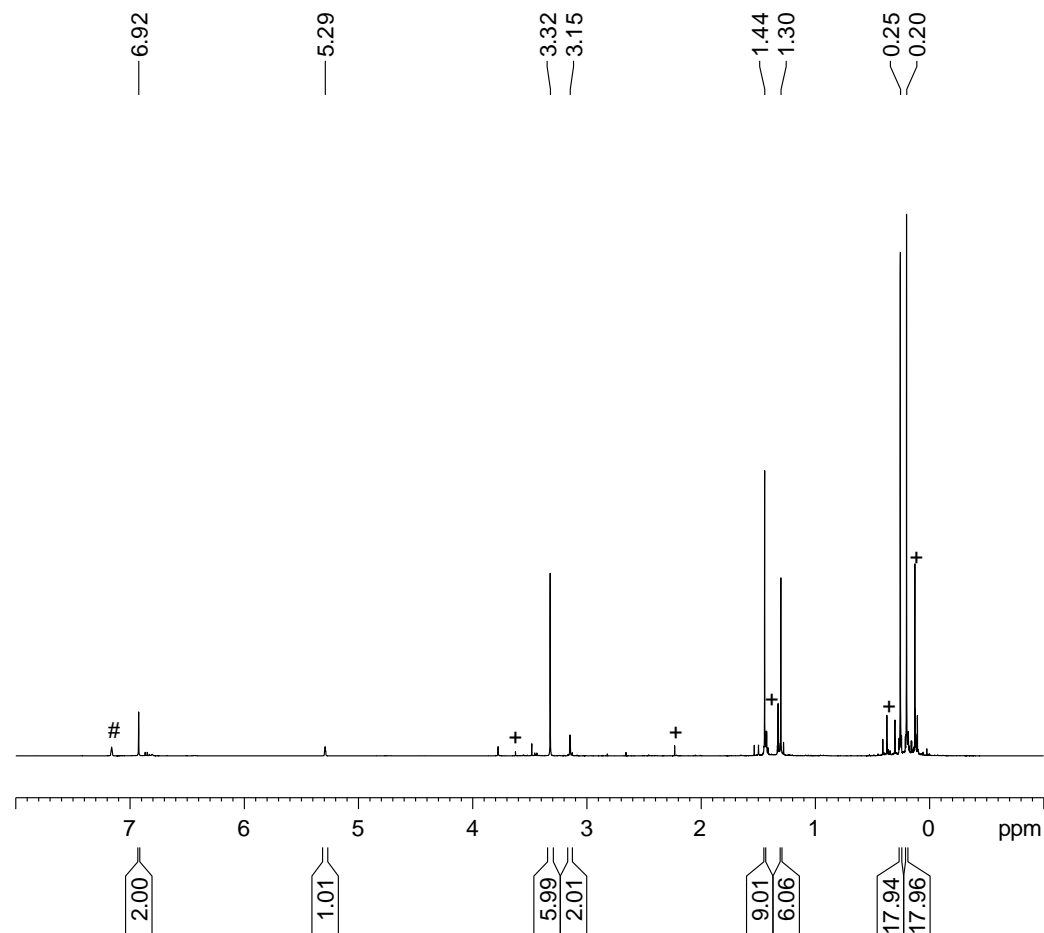
===== CHANNEL f2 =====
CPDPRG[2] waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 -3.00 dB
PL12 11.77 dB
PL13 13.14 dB
PL2W 16.03799057 W
PL12W 0.53474891 W
PL13W 0.39007664 W
SFO2 400.1120007 MHz

F2 - Processing parameters
SI 65536
SF 100.6077400 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S81. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **16**.

NMR spectra of compound 17

¹H-NMR of TbbGeH(Me-NHC) in C₆D₆ (#) at rt, +: unknown impurities



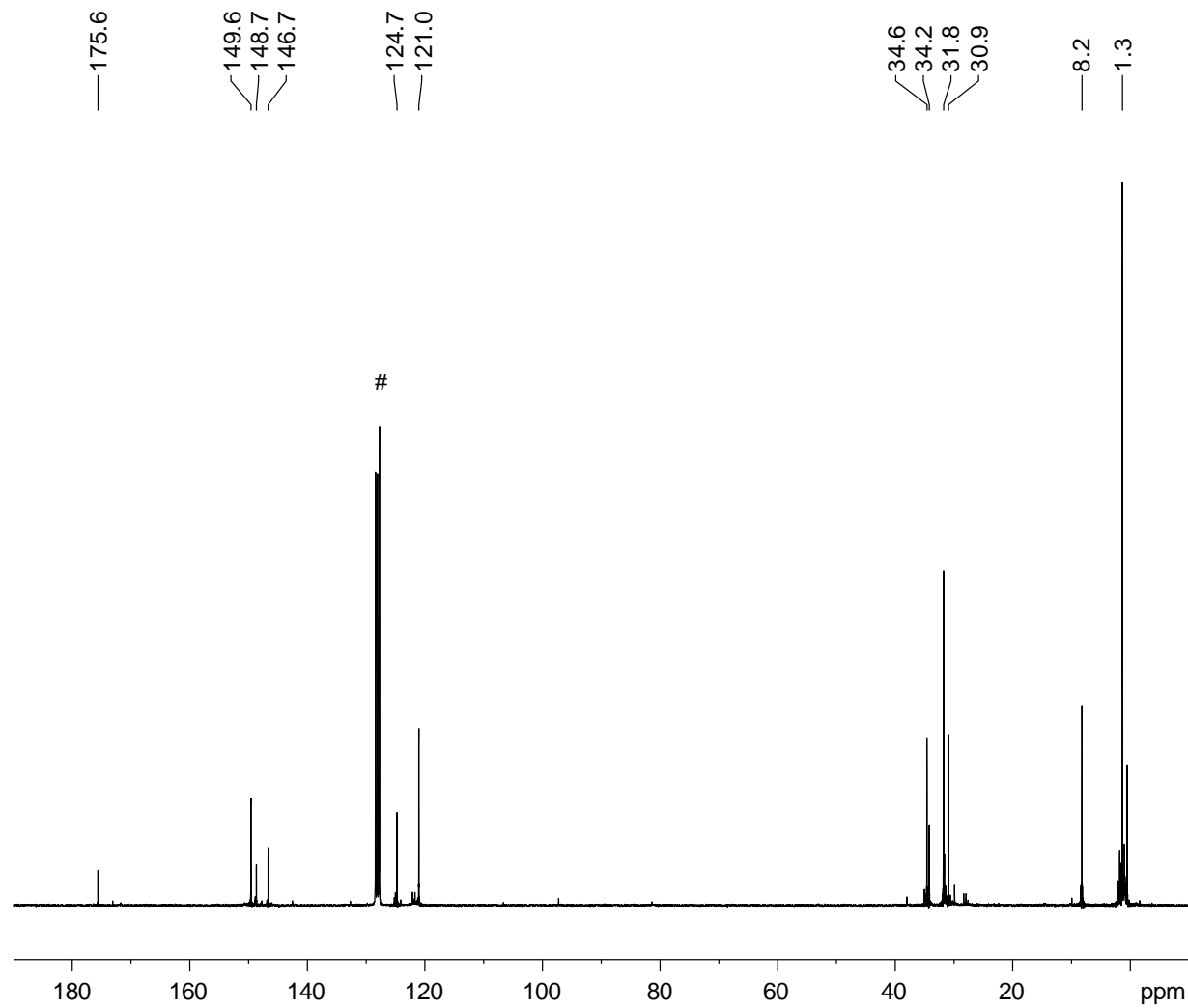
Current Data Parameters
 NAME MA427_30102020_300N
 EXPNO 10
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 20201030
 Time 16.41 h
 INSTRUM spect
 PROBHD Z104275_0338 (
 PULPROG zg30
 TD 38044
 SOLVENT C6D6
 NS 32
 DS 0
 SWH 6009.615 Hz
 FIDRES 0.315930 Hz
 AQ 3.1652608 sec
 RG 100.47
 DW 83.200 usec
 DE 6.50 usec
 TE 298.0 K
 D1 1.00000000 sec
 TD0 1
 SFO1 300.1318533 MHz
 NUC1 1H
 P0 4.67 usec
 P1 14.00 usec
 PLW1 8.26509953 W

F2 - Processing parameters
 SI 32768
 SF 300.1299975 MHz
 WDW EM
 SSB 0
 LB 0 Hz
 GB 0
 PC 1.00

Figure S82. ¹H-NMR spectrum of compound 17.

$^{13}\text{C}\{^1\text{H}\}$ -NMR of TbbGeH(Me-NHC) in C_6D_6 (#) at rt



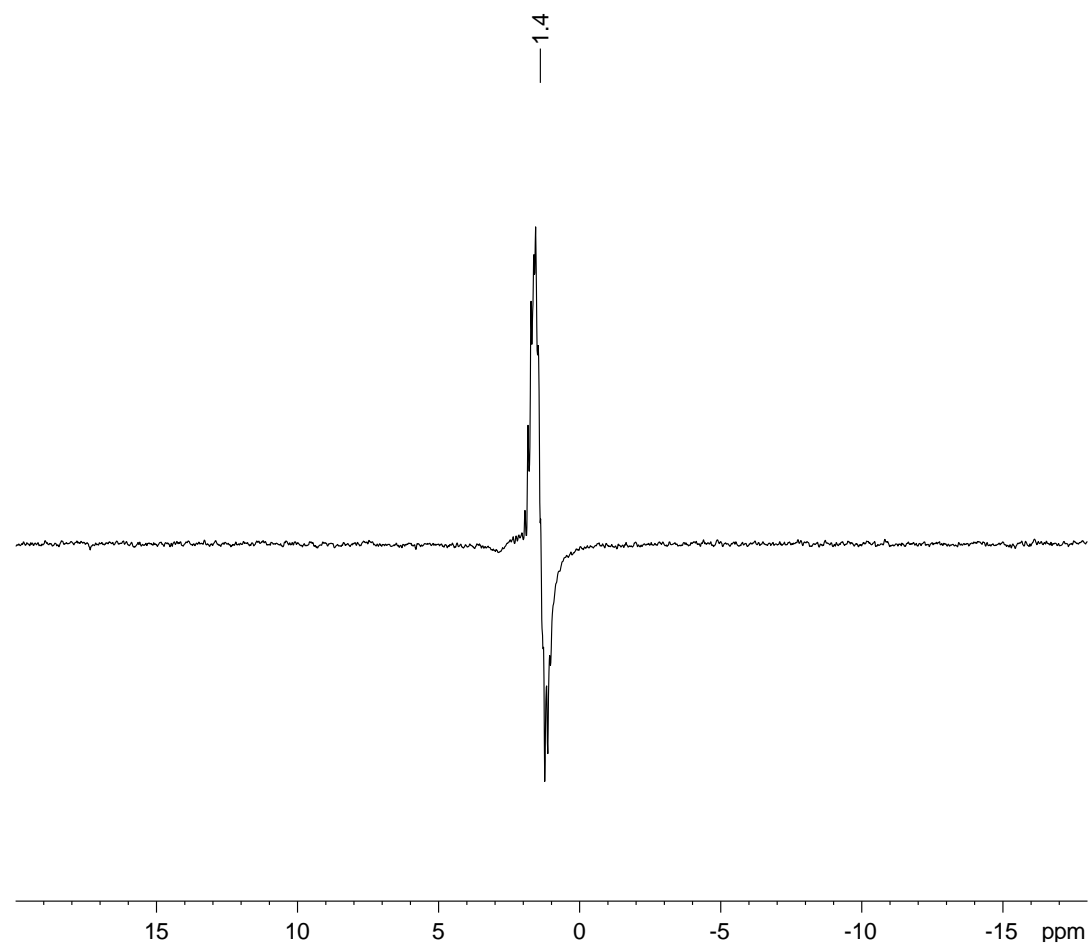
Current Data Parameters
NAME MA427_30102020_300I
EXPNO 13
PROCNO 1

F2 - Acquisition Parameters
Date_ 20201031
Time 0.09 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG udeflt
TD 13040
SOLVENT C6D6
NS 4608
DS 4
SWH 18115.941 Hz
FIDRES 2.778519 Hz
AQ 0.3599040 sec
RG 204.67
DW 27.600 usec
DE 6.50 usec
TE 298.0 K
D1 4.00000000 sec
D12 0.00002000 sec
D20 20.00000000 sec
TD0 1
SFO1 75.4752953 MHz
NUC1 13C
P1 10.00 usec
P13 2000.00 usec
P26 500.00 usec
PLW1 33.55099869 W
SPNAM[5] Crp60comp.4
SPOAL5 0.500
SPOFFS5 0 Hz
SPW5 5.12620020 W
SPNAM[8] Crp60,0,5,20.1
SPOAL8 0.500
SPOFFS8 0 Hz
SPW8 5.12620020 W
SFO2 300.1312005 MHz
NUC2 1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 8.26509953 W
PLW12 0.20000000 W

F2 - Processing parameters
SI 32768
SF 75.4677196 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

Figure S83. $^{13}\text{C}\{^1\text{H}\}$ -NMR spectrum of compound **17**.

^{29}Si -INEPTND-NMR of TbbGeH(Me-NHC) in C_6D_6 at rt



Current Data Parameters
NAME MA427_30102020_300N
EXPNO 11
PROCNO 1

F2 - Acquisition Parameters
Date_ 20201030
Time 16.57 h
INSTRUM spect
PROBHD Z104275_0338 (
PULPROG ineptnd
TD 39048
SOLVENT C6D6
NS 320
DS 0
SWH 26315.789 Hz
FIDRES 1.347869 Hz
AQ 0.7419120 sec
RG 204.67
DW 19.000 usec
DE 6.50 usec
TE 298.0 K
CNST2 6.5999999
D1 2.00000000 sec
D4 0.03787879 sec
TD0 1
SFO1 59.62229159 MHz
NUC1 29Si
P1 10.30 usec
P2 20.60 usec
PLW1 50.00000000 W
SFO2 300.1312005 MHz
NUC2 1H
P3 14.00 usec
P4 28.00 usec
PLW2 8.26509953 W

F2 - Processing parameters
SI 32768
SF 59.6273759 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.00

Figure S84. ^{29}Si -INEPTND-NMR spectrum of compound **17**.

NMR spectroscopic analysis of distannanes $\text{ArSnH}_2\text{-SnH}_2\text{Ar}$

Molecules $\text{RH}_2\text{SnSnH}_2\text{R}$ containing at least one NMR active tin atom constitute higher order spin systems due to the magnetic non-equivalence of the tin hydride protons. However, because the $^1J(\text{Sn,H})$ and $^2J(\text{Sn,H})$ coupling constants are much greater than the H,H coupling constants, the error is insignificant if their contributions to the multiplets are analyzed according to first-order rules (i.e., the two “doublets” of ^{119}Sn satellites in the ^1H NMR spectrum, Fig. S85, or the “triplet of triplets” of the major isotopologue in the ^{119}Sn NMR spectrum, Fig. S85, an A_2XB_2 spin system). An exception is the isotopologue containing two ^{119}Sn nuclei, an $\text{A}_2\text{XX}'\text{A}'_2$ spin system. Its subspectrum completely differs from the $^{119}\text{Sn}, ^{117}\text{Sn}$ isotopologue, which constitutes an A_2XYB_2 spin system. The $^1J(^{119}\text{Sn}, ^{117}\text{Sn})$ coupling constant can be obtained from the $^{119}\text{Sn}\{^1\text{H}\}$ NMR spectrum independently.

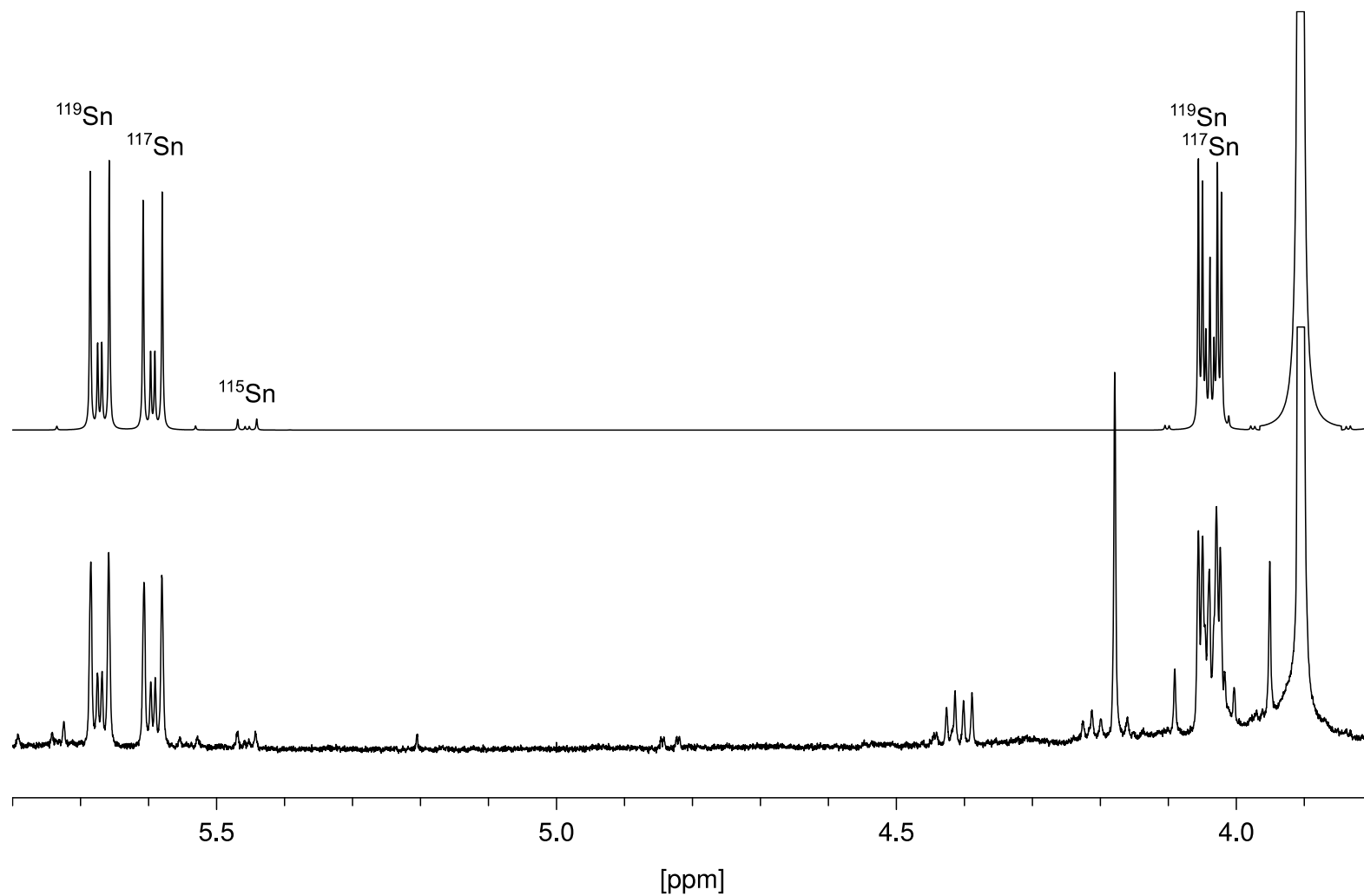


Figure S85. High-frequency half of the experimental and simulated ^1H NMR spectra due to the Sn-H protons of $[\text{Ar}'\text{SnH}_2]_2$ at 3.91 ppm, obtained at 500.13 MHz (r.t.). The satellites due to ^{119}Sn , ^{117}Sn or ^{115}Sn show the effect of ^1H - ^1H couplings.

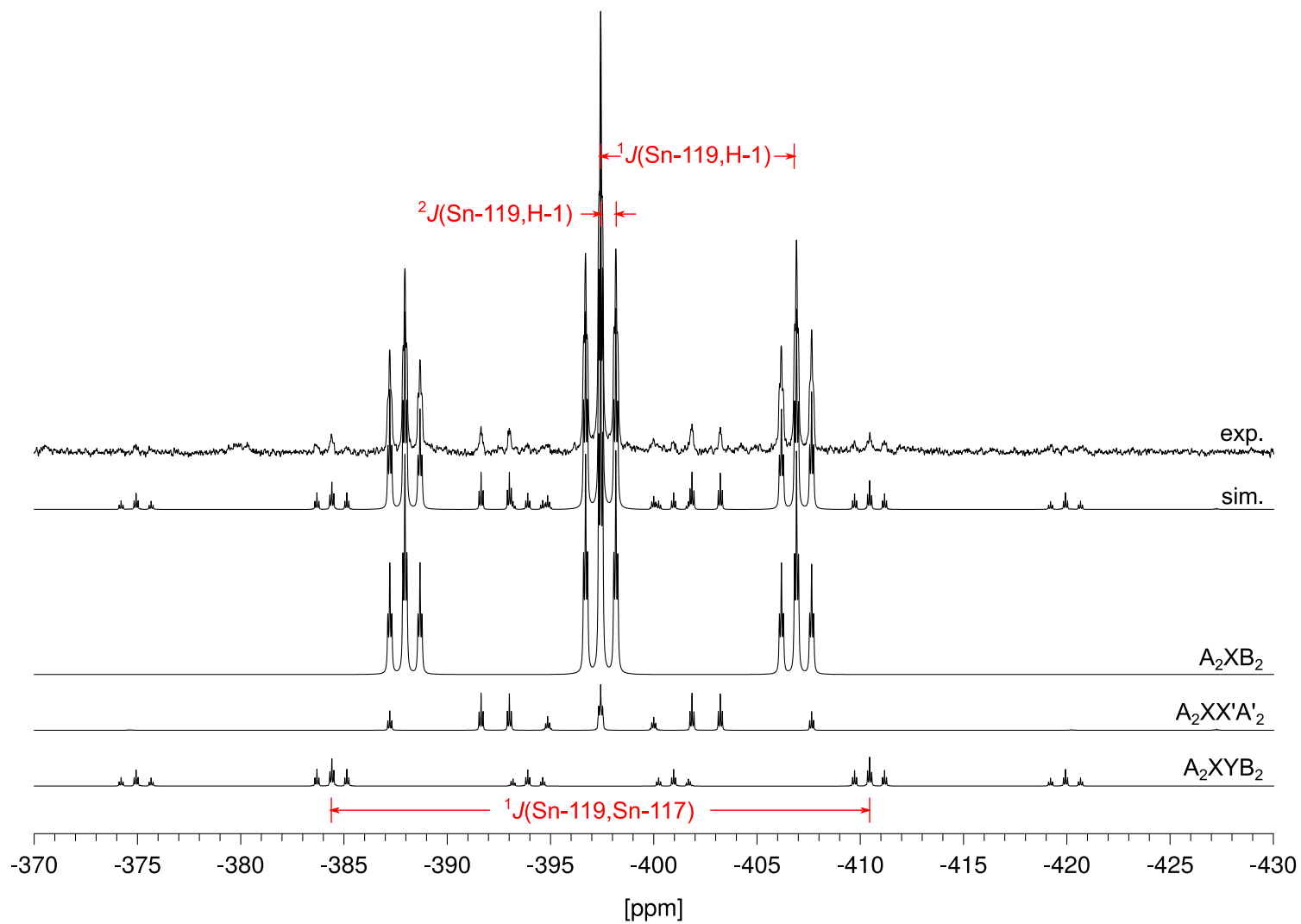


Figure S86. Experimental and calculated ^{119}Sn NMR spectra of $[\text{Ar}'\text{SnH}_2]_2$ obtained at 186.50 MHz (r.t.).

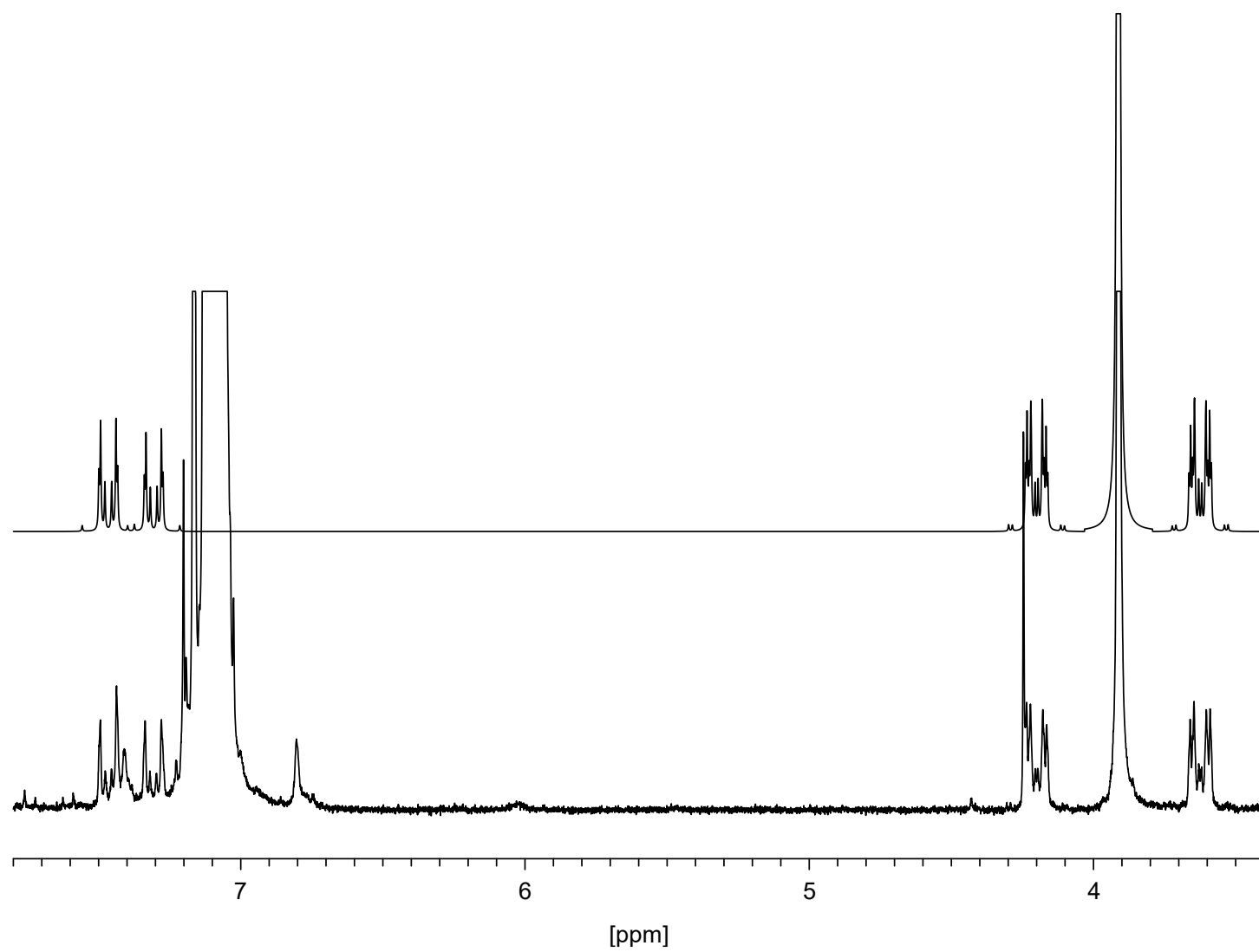


Figure S87. Experimental and calculated $^1\text{H-NMR}$ of $(\text{Ar}^*\text{SnH})_2$.

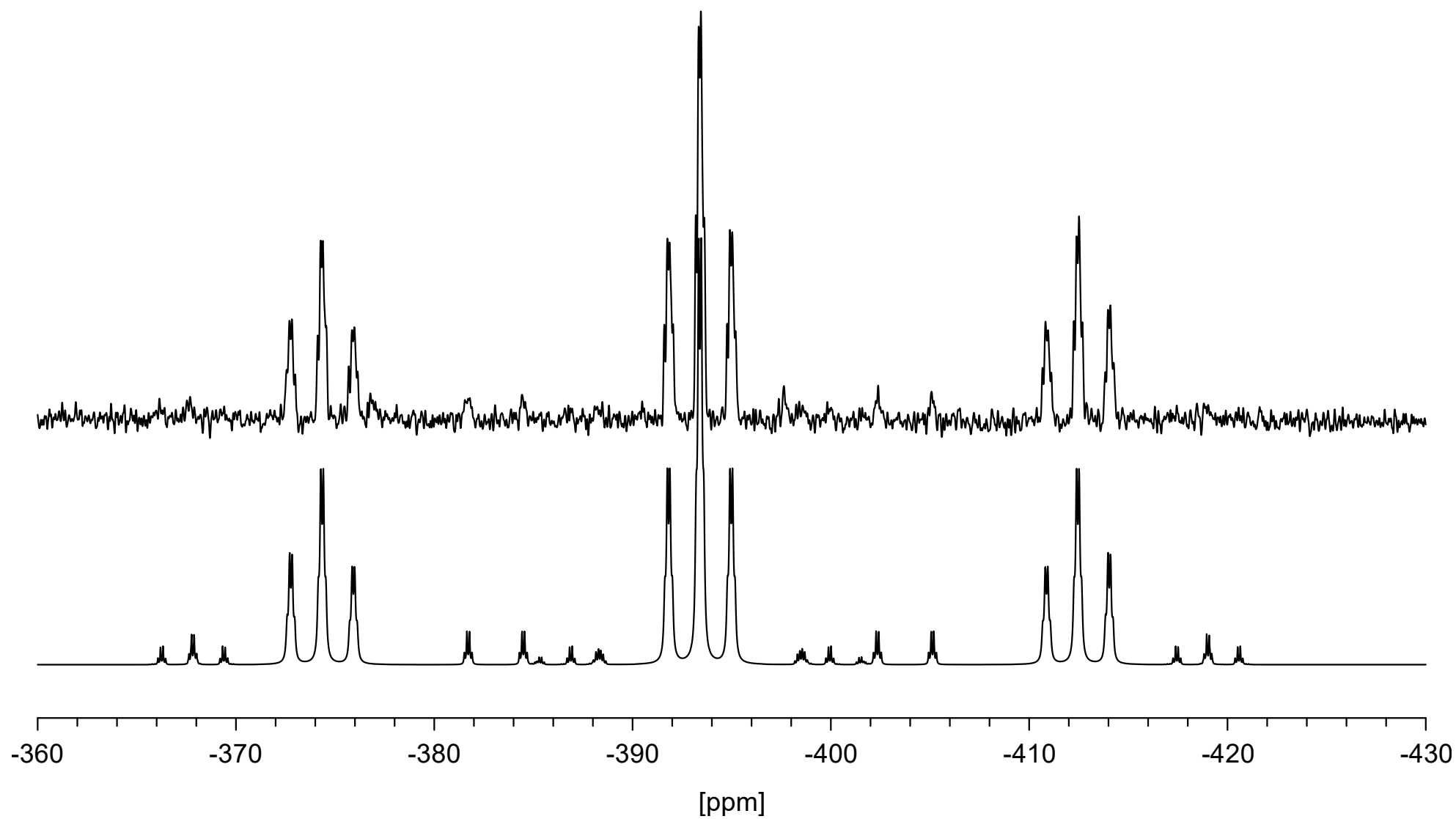


Figure S88. Experimental and calculated ^{119}Sn -NMR of $(\text{Ar}^*\text{SnH})_2$.

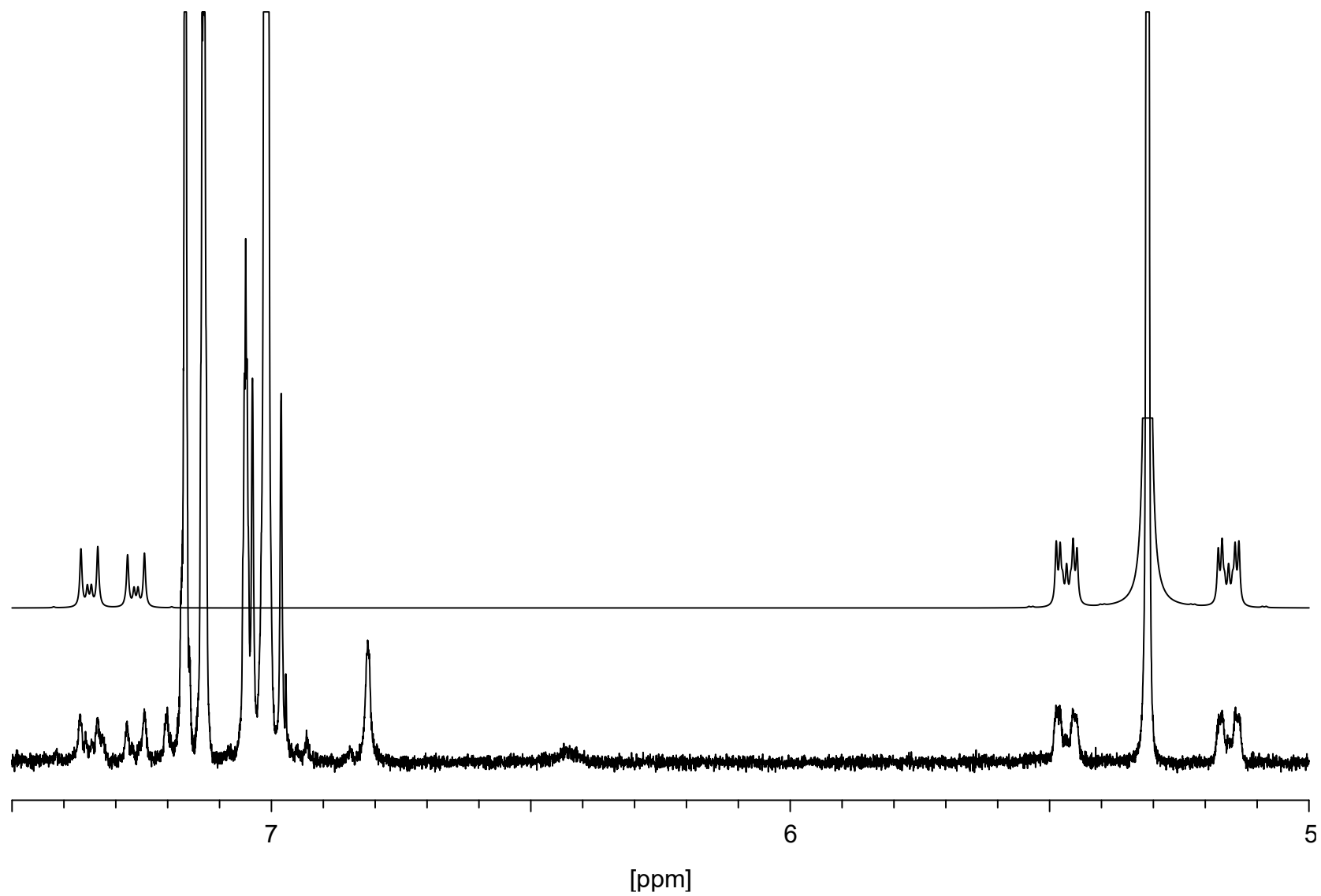


Figure S89. Experimental and calculated $^1\text{H-NMR}$ of $(\text{TbbSnH})_2$.