Supporting Information:

# An Ion Pair Scandium Hydride Supported by A Dianionic (NNNN)-Type Macrocycle Ligand

*Peng Cui, Thomas P. Spaniol, Laurent Maron*and Jun Okuda**

**List of Contents:**

**General.** All operations were performed under an inert atmosphere of argon using standard Schlenk-line or glovebox techniques. THF, pentane, $C_6D_6$ and THF-$d_8$ were dried over $LiAlH_4$, followed by vacuum transfer and stored in the glovebox. (1,7-$Me_2$TACD)$H_2$ was synthesized according to the literature procedure[1] and further purified by distillation at 80 °C in vacuo. [$(C_3H_5)_3Sc(THF)_2$] was prepared as we previously reported.[2] $^1H$, $^{13}C\{^1H\}$, $^{19}F\{^1H\}$ and $^{11}B$ NMR spectra were recorded at 25 °C on a Bruker DRX 400 spectrometer at 400 MHz, 100 MHz, 188 MHz and 128 MHz, respectively. All chemical shifts were reported in δ units with references to the residual solvent resonance of the deuterated solvents for proton and carbon chemical shifts, to external $BF_3{\cdot}OEt_2$ and $CFCl_3$ for boron and fluorine chemical shifts. Elemental analyses were performed by the microanalytical laboratory of this department.

**[(1,7-$Me_2$TACD)Sc($\eta^3$-$C_3H_5$)] (1):** (1,7-$Me_2$TACD)$H_2$ (52 mg, 0.256 mmol) in 1 mL of THF was slowly added to [$(C_3H_5)_3Sc(THF)_2$] (80 mg, 0.256 mmol) in 1 mL of THF at room temperature. The brown solution was then stirred for 20 min. The solvent was removed under vacuum and the pale-brown solid residue was extracted with pentane (8 mL × 3). After filtration, evaporation of pentane gave the product as pale-yellow crystals, yield 66 mg (82%). $^1H$ NMR ($C_6D_6$, 400 MHz, 25 °C): $\delta$ = 6.66 (quintet, $^3J_{HH}$ = 12.0 Hz, 1H, $CH_2CHCH_2$), 3.66 (dd, $^2J_{HH}$ = 12.6 Hz, $^3J_{HH}$ = 4.2 Hz, 2H, $CH_2$), 3.64 (dd, $^2J_{HH}$ = 12.4 Hz, $^3J_{HH}$ = 4.0 Hz, 2H, $CH_2$), 3.03 (br d, $^3J_{HH}$ = 11.6 Hz, 4H, $CH_2CHCH_2$), 2.64 (dd, $^2J_{HH}$ = 12.4 Hz, $^3J_{HH}$ = 4.8 Hz, 2H, $CH_2$), 2.62 (dd, $^2J_{HH}$ = 12.6 Hz, $^3J_{HH}$ = 4.8 Hz, 2H, $CH_2$), 2.56 (dd, $^2J_{HH}$ = 11.2 Hz, $^3J_{HH}$ = 4.6 Hz, 2H, $CH_2$), 2.55 (dd, $^2J_{HH}$ = 11.2 Hz, $^3J_{HH}$ = 5.0 Hz, 2H, $CH_2$), 2.39 (dd, $^2J_{HH}$ = 11.2 Hz, $^3J_{HH}$ = 4.2 Hz, 2H, $CH_2$), 2.37 (dd, $^2J_{HH}$ = 11.2 Hz, $^3J_{HH}$ = 4.2 Hz, 2H, $CH_2$), 2.31 ppm (s, 6H, $CH_3$); $^{13}C\{^1H\}$ NMR ($C_6D_6$, 100 MHz, 25 °C): $\delta$ = 147.91 ($CH_2CHCH_2$), 71.51 ($CH_2CHCH_2$), 59.54 ($CH_2$), 55.19 ($CH_2$), 46.95 ppm ($CH_3$). Anal. Calcd for $C_{13}H_{27}N_4Sc$ (284.34 g/mol): C, 54.91; H, 9.57; N, 19.70. Found: C, 54.86; H, 9.27; N, 19.46.

**[(1,7-$Me_2$TACD)$_3Sc_3H_2$][(1,7-$Me_2$TACD)$_3Sc_3H_4$] (2):** A solution of **1** (50 mg, 0.176 mmol) in 3 mL of THF was degassed and treated with 1 bar of $H_2$. The pale yellow solution was stirred at room temperature for 2 d. The solvent was removed under vacuum, the residue was washed with pentane and then recrystallized in THF/pentane at -35°C to give the product as an off-white

crystalline solid, yield 30 mg (70%). $^{1}$H NMR (THF-$d_8$, 400 MHz, 25 °C): $\delta$ = 4.37 (br s, 4H, $Sc_3H_4$), 3.48 (dd, $^{2}J_{HH}$ = 11.8 Hz, $^{3}J_{HH}$ = 3.8 Hz, 4H, C$H_2$), 3.46 (dd, $^{2}J_{HH}$ = 12.0 Hz, $^{3}J_{HH}$ = 3.6 Hz, 4H, C$H_2$), 3.37 (dd, $^{2}J_{HH}$ = 10.5 Hz, $^{3}J_{HH}$ = 2.6 Hz, 4H, C$H_2$), 3.34 (dd, $^{2}J_{HH}$ = 10.3 Hz, $^{3}J_{HH}$ = 2.6 Hz, 4H, C$H_2$), 3.27-3.22 (m, 6H, C$H_2$), 3.05-3.00 (m, 12H, C$H_2$), 2.85 (s, 22H, C$H_3$ and C$H_2$), 2.83-2.81 (m, 4H, C$H_2$), 2.79-2.75 (m, 10H, C$H_2$), 2.78 (s, 6H, C$H_3$), 2.71 (t, $^{3}J_{HH}$ = 3.4 Hz, 4H, C$H_2$), 2.67 (t, $^{3}J_{HH}$ = 3.4 Hz, 4H, C$H_2$), 2.65 (dd, $^{2}J_{HH}$ = 11.8 Hz, $^{3}J_{HH}$ = 3.8 Hz, 6H, C$H_2$), 2.63 (dd, $^{2}J_{HH}$ = 11.8 Hz, $^{3}J_{HH}$ = 3.8 Hz, 6H, C$H_2$), 2.56 (s, 12H, C$H_3$), 2.52 (dd, $^{2}J_{HH}$ = 10.6 Hz, $^{3}J_{HH}$ = 3.8 Hz, 6H, C$H_2$), 2.50 (dd, $^{2}J_{HH}$ = 10.2 Hz, $^{3}J_{HH}$ = 3.8 Hz, 6H, C$H_2$), 2.41 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 4.0 Hz, 4H, C$H_2$), 2.39 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 3.6 Hz, 4H, C$H_2$), 2.32 ppm (t, $^{3}J_{HH}$ = 3.0 Hz, 3H, C$H_2$), 2.29 (t, $^{3}J_{HH}$ = 3.0 Hz, 3H, C$H_2$), 2.16 (br s, 2H, $Sc_3H_2$); $^{13}$C{$^{1}$H} NMR (THF-$d_8$, 400 MHz, 25 °C): $\delta$ = 61.39 (*C*H$_2$), 60.57 (*C*H$_2$), 59.36 (*C*H$_2$), 56.82 (*C*H$_2$), 54.60 (*C*H$_2$), 53.45 (*C*H$_2$), 51.44 (*C*H$_2$), 49.03 (*C*H$_3$), 49.13 (*C*H$_3$), 48.04 ppm (*C*H$_3$). Anal. Calcd for $C_{60}H_{138}N_{24}Sc_6 \cdot C_4H_8O \cdot C_5H_{12}$ (1609.04 g/mol): C, 51.48; H, 9.89; N, 20.88. Found: C, 50.39; H, 9.88; N, 20.34. When crystallized from THF/pentane, the crystals always contain THF and pentane molecules in the crystal lattice, the relative low value for carbon may be attributed to the partial loss of these solvent molecules. The same holds for the corresponding deuteride **2-D$_6$**, which was synthesized by an analogous procedure by using $D_2$ instead of $H_2$. The $^{1}$H NMR spectrum of **2-D$_6$** is essentially the same as that of **2** except for the absence of resonances for the Sc*H* at $\delta$ = 4.37 and 2.16 ppm.

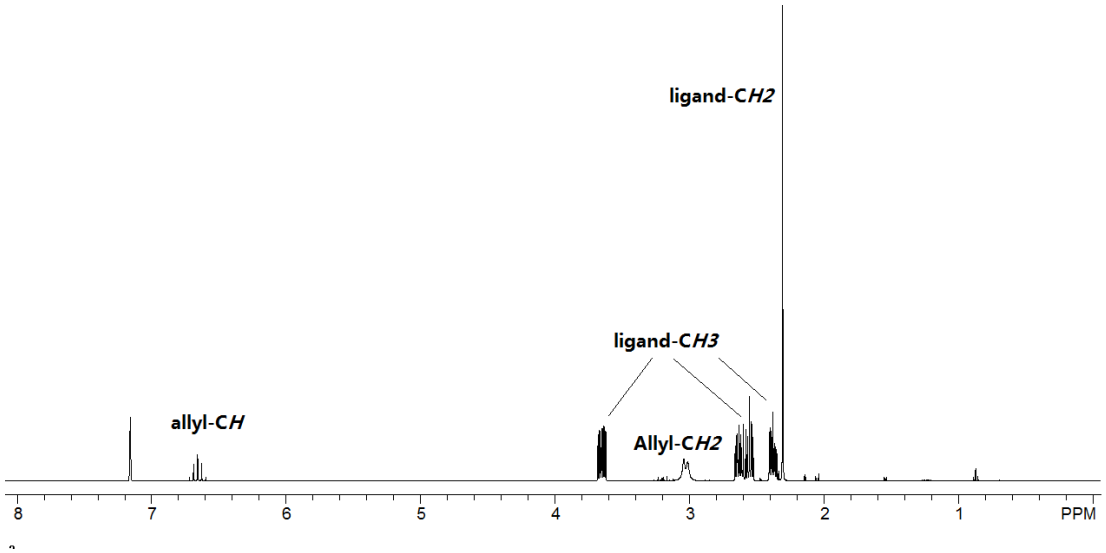
**[(1,7-Me$_2$TACD)$_3$Sc$_3$H$_2$][B{C$_6$H$_3$-3,5-(CF$_3$)$_2$}$_4$] (3):** A solution of [Me$_3$NH][B{C$_6$H$_3$-3,5-(CF$_3$)$_2$}$_4$] (50.4 mg, 0.056 mmol) in 1 mL of THF was slowly added to **1** (40 mg, 0.028 mmol) in 1 mL of THF. $H_2$ gas vigorously evolved. The pale-yellow solution was then stirred for 5 min. The solvent was removed under vacuum, the residue was washed with pentane and recrystallized from THF/pentane at room temperature to give the product as off-white crystals, yield 80 mg (92%). $^{1}$H NMR (THF-$d_8$, 400 MHz, 25 °C): $\delta$ = 7.79 (t, $^{4}J_{HH}$ = 2.4 Hz, 8H, *ortho*-aryl), 7.57 (s, 4H, *para*-aryl), 3.37 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 2.8 Hz, 3H, C$H_2$), 3.33 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 2.8 Hz, 3H, C$H_2$), 3.24 (dd, $^{2}J_{HH}$ = 12.6 Hz, $^{3}J_{HH}$ = 3.4 Hz, 3H, C$H_2$), 3.22 (dd, $^{2}J_{HH}$ = 11.4 Hz, $^{3}J_{HH}$ = 2.2 Hz, 3H, C$H_2$), 3.06-2.99 (m, 12H, C$H_2$), 2.84 (s, 18H, C$H_3$ and C$H_2$), 2.82-2.80 (m, 6H, C$H_2$), 2.78-2.73 (m, 6H, C$H_2$), 2.70 (t, $^{3}J_{HH}$ = 3.6 Hz, 3H, C$H_2$) , 2.66 (t, $^{3}J_{HH}$ = 3.4 Hz, 3H, C$H_2$), 2.30 (t,

$^{3}J_{HH}$ = 3.0 Hz, 3H, C$H_2$), 2.27 (t, $^{3}J_{HH}$ = 3.0 Hz, 3H, C$H_2$), 2.16 ppm (br s, 2H, Sc$_3H_2$); $^{13}$C{$^{1}$H} NMR (THF-$d_8$, 400 MHz, 25 °C): $\delta$ = 162.64 (q, $^{1}J_{BC}$ = 49.7 Hz, *ipso*-aryl), 135.42 (*ortho*-aryl), 129.84 (qq, $^{2}J_{FC}$ = 31.0 Hz, $^{4}J_{FC}$ = 2.6 Hz, *meta*-aryl), 125.34 (q, $^{1}J_{FC}$ = 270.6 Hz, $CF_3$), 118.00 (t, $^{3}J_{FC}$ = 3.5 Hz, *para*-aryl), 61.36 ($CH_2$), 59.33 ($CH_2$), 54.53 ($CH_2$), 53.41 ($CH_2$), 49.22 ($CH_3$); $^{19}$F NMR (THF-$d_8$, 188 MHz, 25 °C): δ = –63.39 ppm (s, $CF_3$), $^{11}$B NMR (THF-$d_8$, 128 MHz, 25 °C): δ = –6.51 ppm (s). Anal. Calcd for $C_{62}H_{80}BF_{24}N_{12}Sc_3$ (1595.02 g/mol): C, 46.69; H, 5.06; N, 10.54. Found: C, 46.88; H, 5.02; N, 9.98.

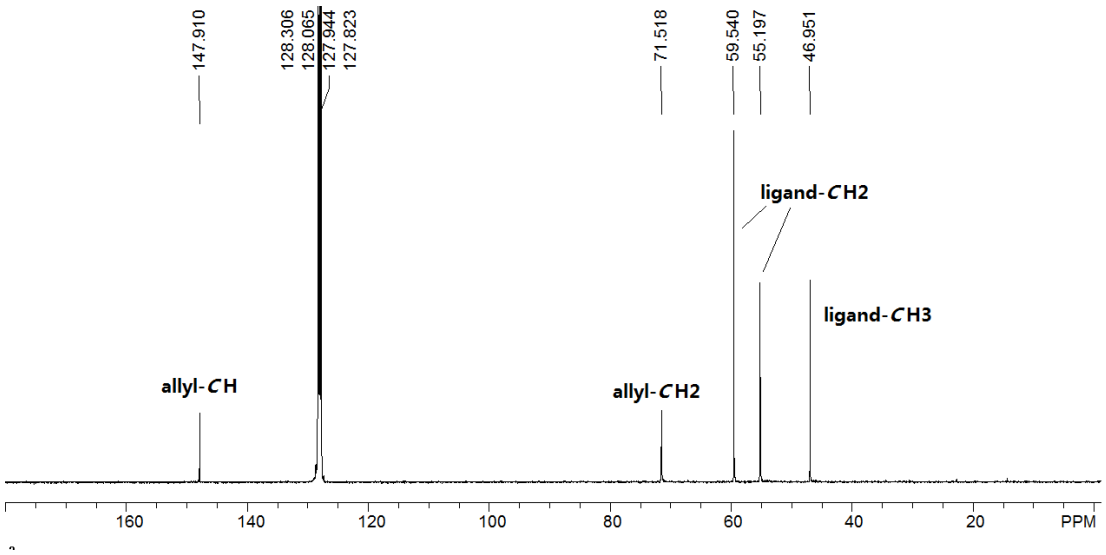
**[(1,7-Me$_2$TACD)$_3$Sc$_3$H$_2$][SPh] (4):** A solution of **2** (15 mg, 0.01 mmol) in 0.5 mL of THF was carefully layered with PhSSPh (2 mg, 0.01 mmol) in 0.5 mL of THF. The mixture was allowed to stand at room temperature for 24 h, during which time colorless blocks of **4** precipitated from the solution. The crystals were isolated, washed with pentane, and dried under vacuum; yield 14 mg (82%). $^{1}$H NMR (THF-$d_8$, 400 MHz, 25 °C): $\delta$ = 7.17 (d, $^{3}J_{HH}$ = 8.0 Hz, 2H, *ortho*-Ar), 6.45 (t, $^{3}J_{HH}$ = 7.0 Hz, 2H, *meta*-aryl), 6.17 (t, $^{3}J_{HH}$ = 6.4 Hz, 1H, *para*-aryl), 3.37 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 2.6 Hz, 3H, C$H_2$), 3.33 (dd, $^{2}J_{HH}$ = 10.4 Hz, $^{3}J_{HH}$ = 2.6 Hz, 3H, C$H_2$), 3.22 (dd, $^{2}J_{HH}$ = 12.4 Hz, $^{3}J_{HH}$ = 3.2 Hz, 3H, C$H_2$), 3.21 (dd, $^{2}J_{HH}$ = 11.2 Hz, $^{3}J_{HH}$ = 2.3 Hz, 3H, C$H_2$), 2.86 (s, 18H, C$H_3$), 2.83-2.79 (m, 6H, C$H_2$), 2.77-2.75 (m, 3H, C$H_2$), 2.75-2.71 (m, 3H, C$H_2$), 2.67 (t, $^{3}J_{HH}$ = 3.4 Hz, 3H, C$H_2$) , 2.63 (t, $^{3}J_{HH}$ = 3.6 Hz, 3H, C$H_2$), 2.33 (t, $^{3}J_{HH}$ = 3.2 Hz, 3H, C$H_2$), 2.30 (t, $^{3}J_{HH}$ = 3.2 Hz, 3H, C$H_2$), 2.14 (br s, 2H, Sc$_3H_2$). $^{13}$C NMR spectrum could not be recorded due to the poor solubility of the compound in THF and any other common solvents. Anal. Calcd for $C_{36}H_{73}N_{12}SSc_3 \cdot C_4H_8O$ (913.08 g/mol): C, 52.62; H, 8.94; N, 18.41. Found: C, 52.15; H, 9.02; N, 18.69.

**[(1,7-Me$_2$TACD)$_2$Sc$_2$(H)C$_2$H$_5$] (5):** A solution of **2** (35 mg, 0.024 mmol) in 2.0 mL of THF was degassed and treated with 1 bar of $C_2H_4$. The pale yellow solution was stirred at room temperature for 2 d. The solvent was then removed under vacuum and the pale-yellow residue was extracted with pentane (4 mL × 3). After filtration, evaporation of pentane gave the product as a pale-yellow crystals; yield 28 mg (82%). $^{1}$H NMR ($C_6D_6$, 400 MHz, 25 °C): $\delta$ = 4.94 (br s, 1H, Sc*H*), 3.75 (br s, 2H, C$H_2$), 3.60 (br s, 2H, C$H_2$), 3.43 (dd, $^{2}J_{HH}$ = 11.6 Hz, $^{3}J_{HH}$ = 6.0 Hz, 1H, C$H_2$), 3.40 (dd, $^{2}J_{HH}$ = 11.6 Hz, $^{3}J_{HH}$ = 6.0 Hz, 1H, C$H_2$), 3.19-3.12 (m, 4H, C$H_2$), 3.04 (dd, $^{2}J_{HH}$ = 11.8 Hz, $^{3}J_{HH}$ =

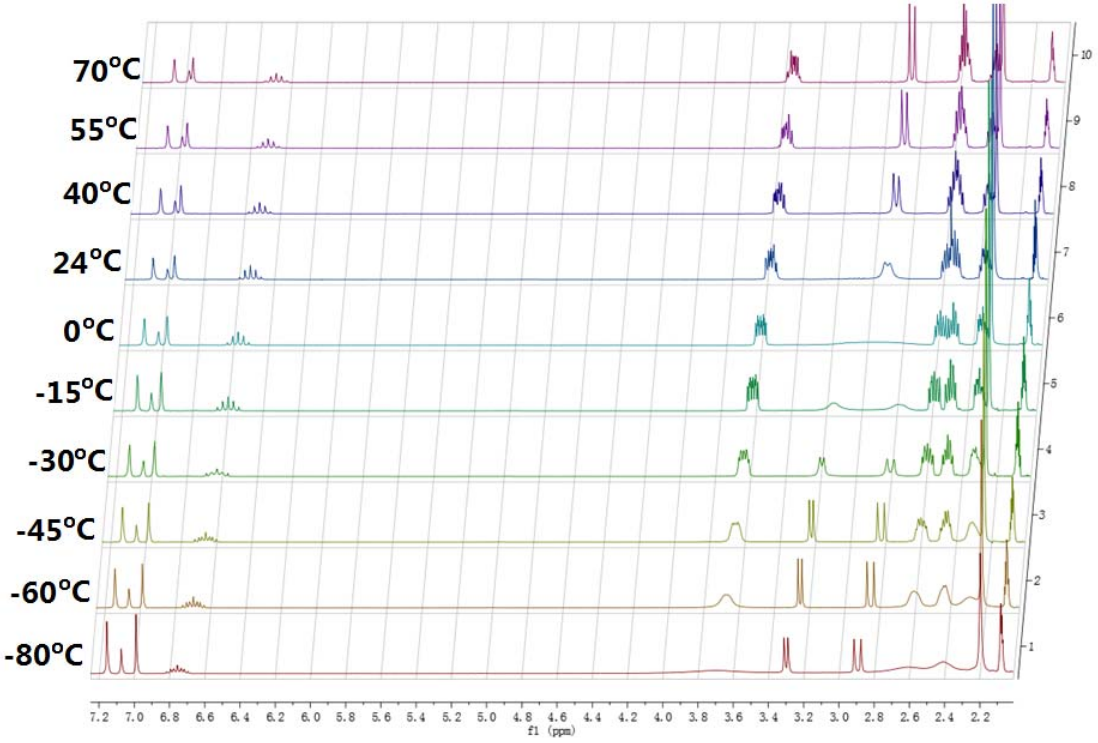
4.2 Hz, 1H, C$H_2$), 3.01 (dd, $^2J_{HH}$ = 11.4 Hz, $^3J_{HH}$ = 4.6 Hz, 1H, C$H_2$), 2.82-2.62 (m, 12H, C$H_2$), 2.47 (s, overlapped, 12H, C$H_3$), 2.28 (dd, $^2J_{HH}$ = 11.2 Hz, $^3J_{HH}$ = 4.4 Hz, 4H, C$H_2$), 2.23-2.15 (m, 4H, C$H_2$), 1.80 (t, $^3J_{HH}$ = 8.2 Hz, 3H, $CH_2CH_3$), 0.41 ppm (q, $^3J_{HH}$ = 8.0 Hz, 2H, $CH_2CH_3$); $^{13}C\{^1H\}$ NMR ($C_6D_6$, 100 MHz, 25 °C): $\delta$ = 61.82 ($CH_2$), 60.41 ($CH_2$), 58.80 ($CH_2$), 55.81 ($CH_3$), 55.74 ($CH_3$), 53.52 ($CH_2$), 48.71 ($CH_2$), 47.49 ($CH_2$), 46.18 ($CH_2$), 45.39 ($CH_2$), 28.44 ($CH_2CH_3$), 13.05 ppm ($CH_2CH_3$). Anal. Calcd for $C_{22}H_{50}N_8Sc_2$ (516.60 g/mol): C, 51.15; H, 9.76; N, 21.69. Found: C, 51.77; H, 10.00; N, 21.27. The deuteride **5-D$_2$** was synthesized by a similar procedure using **2-D$_6$** instead of **2**. The $^1$H NMR spectrum of **5-D$_2$** in $C_6D_6$ is essentially the same as that of **2** except for the absence of resonances for the Sc$H$ at $\delta$ = 4.37 ppm. The resonances for the Sc$CH_2CH_2$D are observed at $\delta$ = 1.80 (tt, $^3J_{HH}$ = 8.0 Hz, $^2J_{HD}$ = 4Hz, 2H, $CH_2CH_2$D) and 0.42 ppm (tt, $^3J_{HH}$ = 8.0 Hz, $^3J_{HD}$ = 1.6 Hz, 2H, $CH_2CH_2$D).



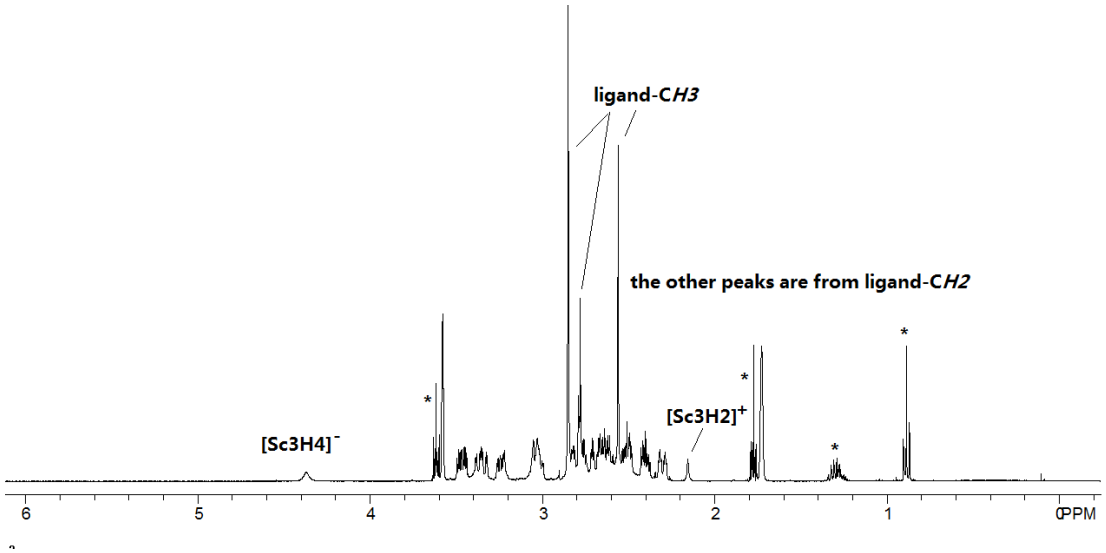

$^{1}$H NMR spectrum of **1** in $C_6D_6$ at 25 °C.



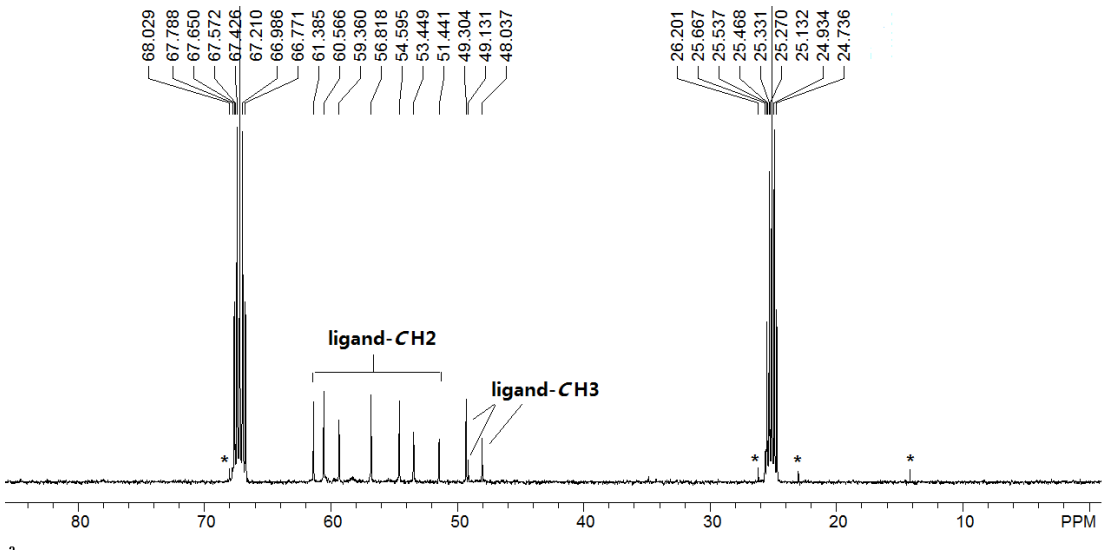

$^{13}$C{$^{1}$H} NMR spectrum of **1** in $C_6D_6$ at 25 °C.



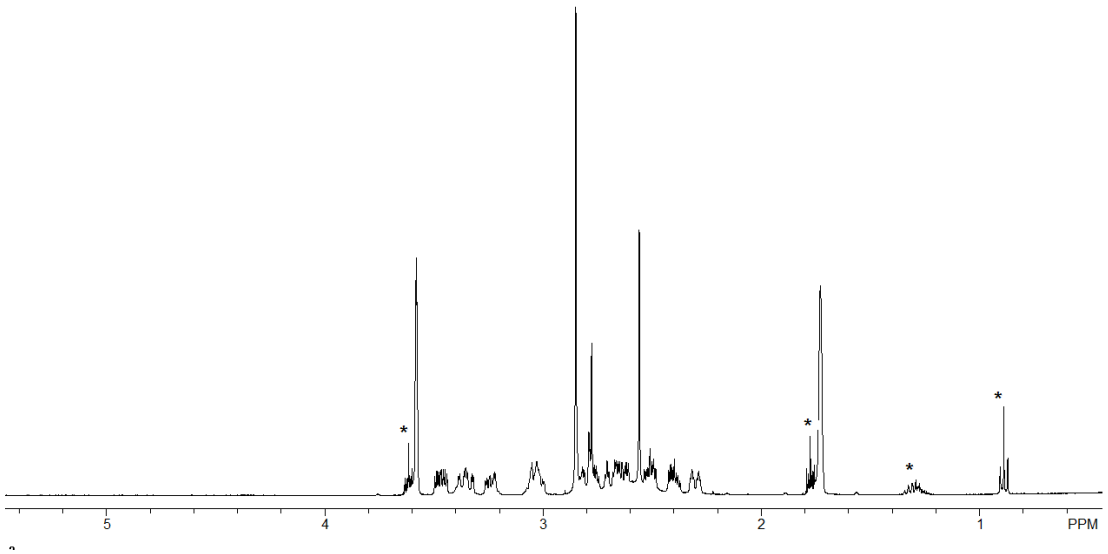

VT-NMR spectra of **1** in toluene-$d_8$.



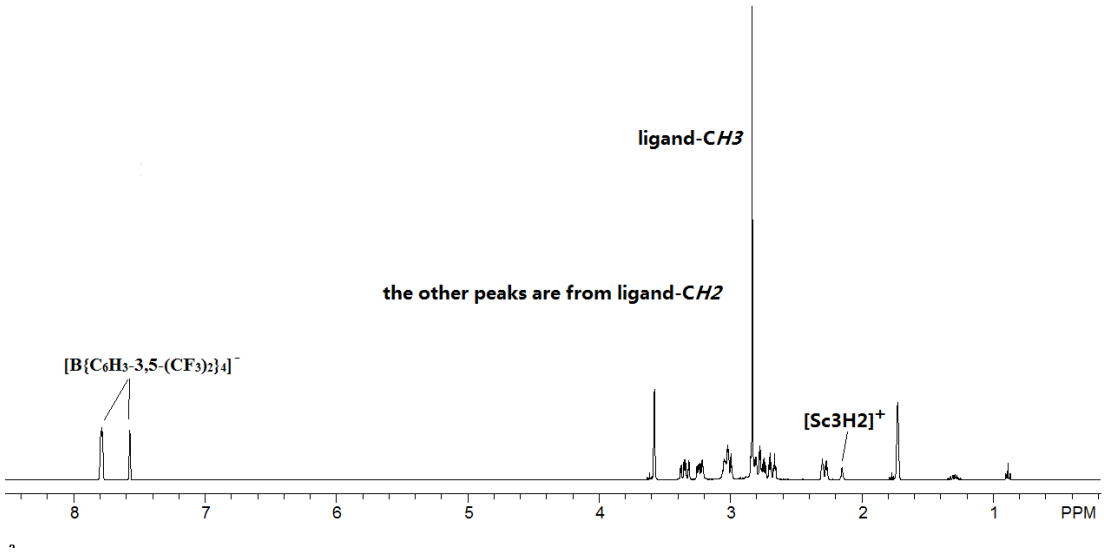

$^1$H NMR spectrum of **2** in THF-$d_8$ at 25 °C. * denotes co-crystallized THF and pentane.



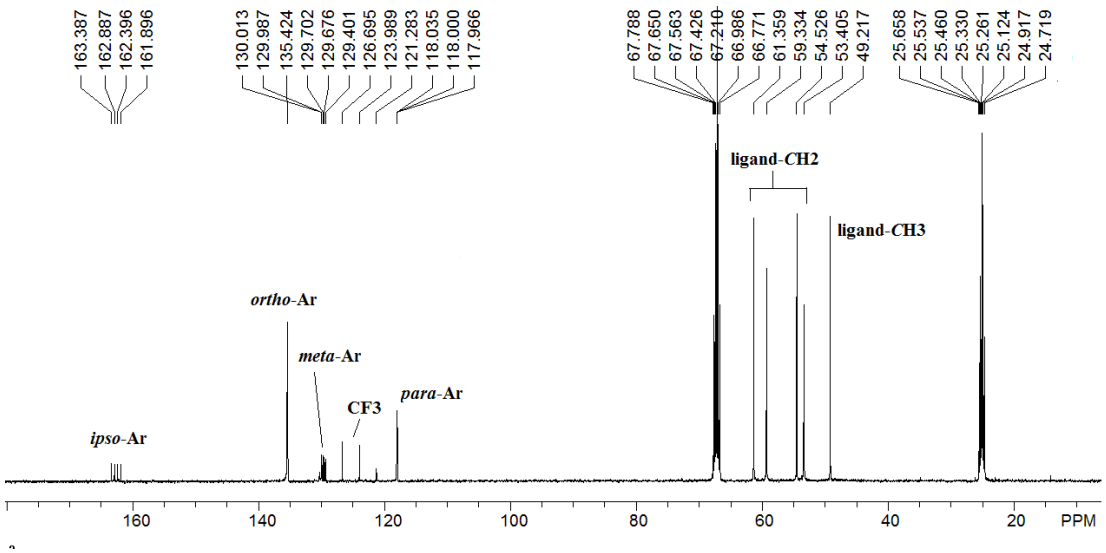

$^{13}C\{^{1}H\}$ NMR spectrum of **2** in THF-$d_8$ at 25 °C. * denotes co-crystallized THF and pentane.



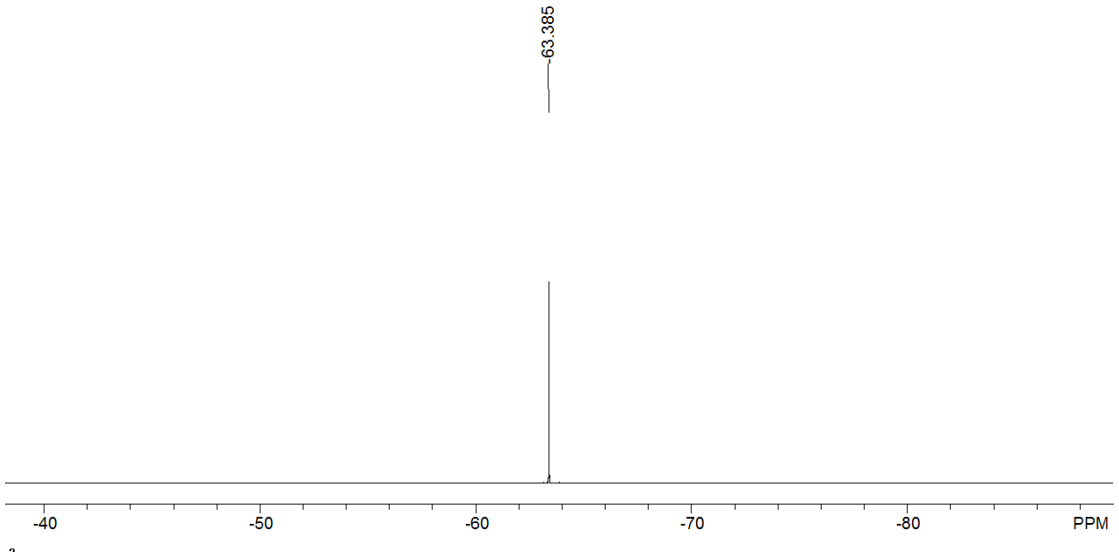

$^{1}H$ NMR spectrum of **2-D$_6$** in THF-$d_8$ at 25 °C. * denotes co-crystallized THF and pentane.



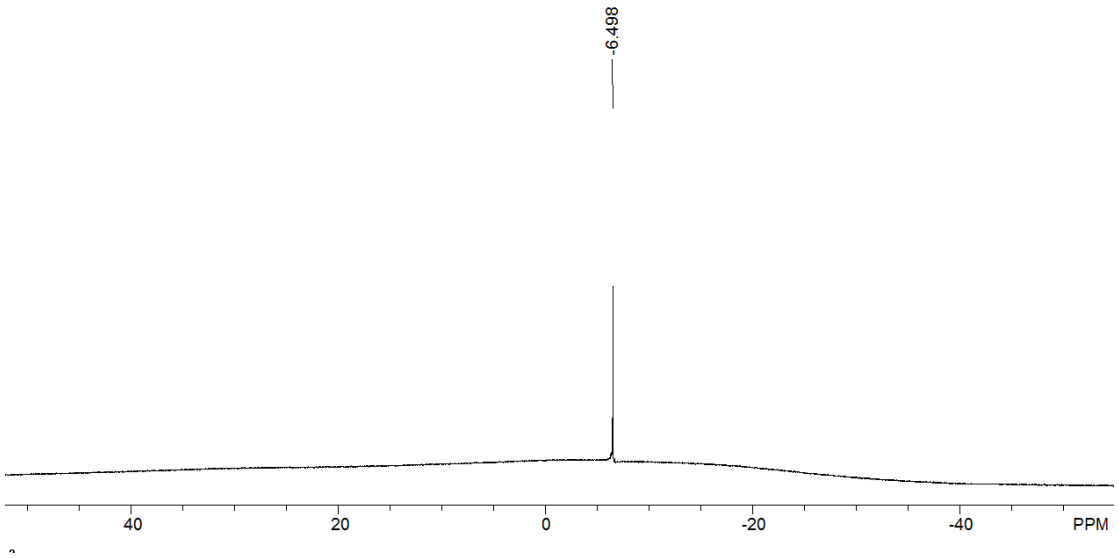

$^{1}$H NMR spectrum of **3** in THF-$d_8$ at 25 °C.



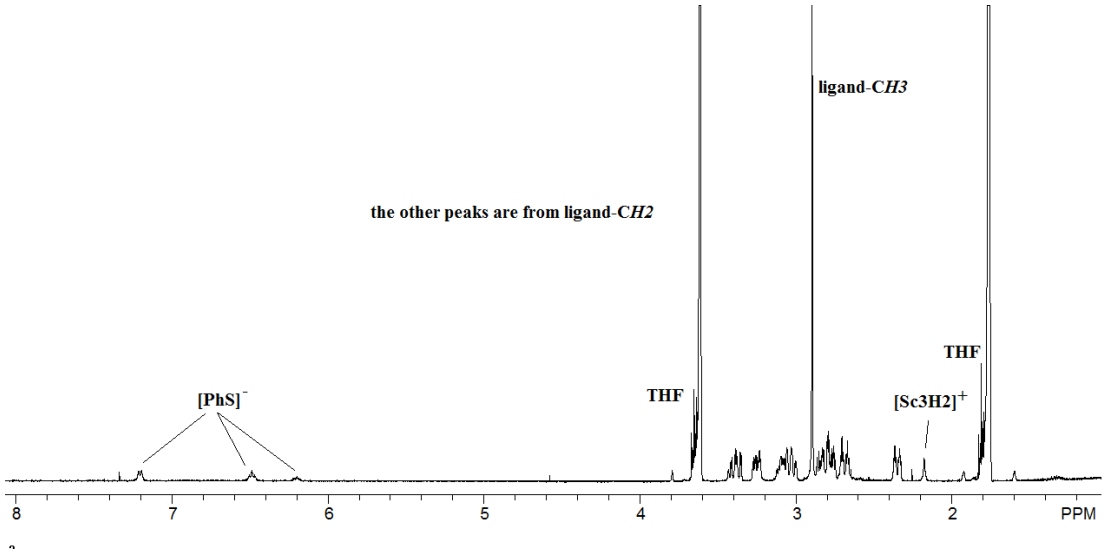

$^{13}$C{$^{1}$H} NMR spectrum of **3** in THF-$d_8$ at 25 °C.



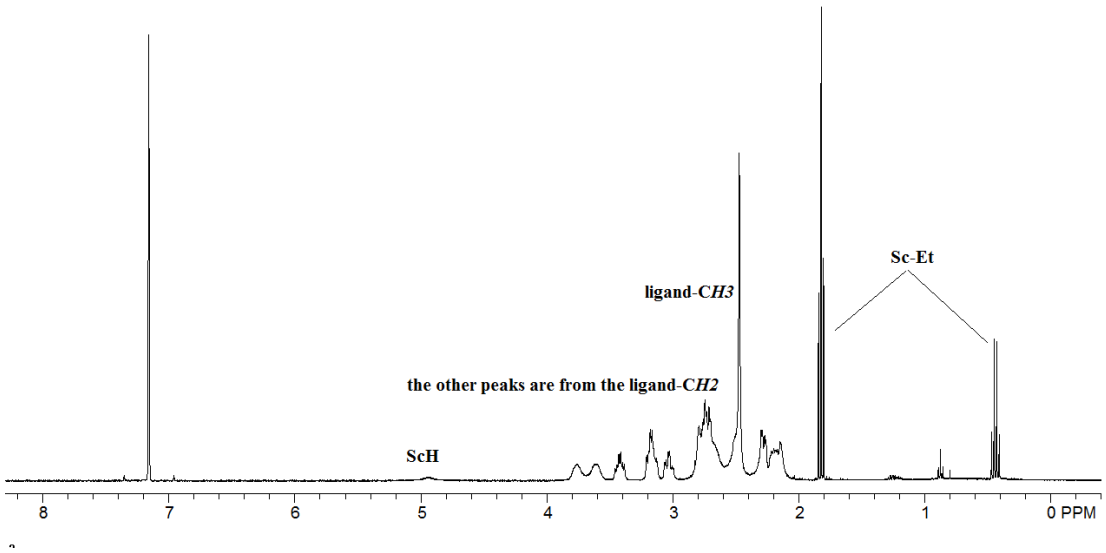

$^{19}$F NMR spectrum of **3** in THF-$d_8$ at 25 °C.



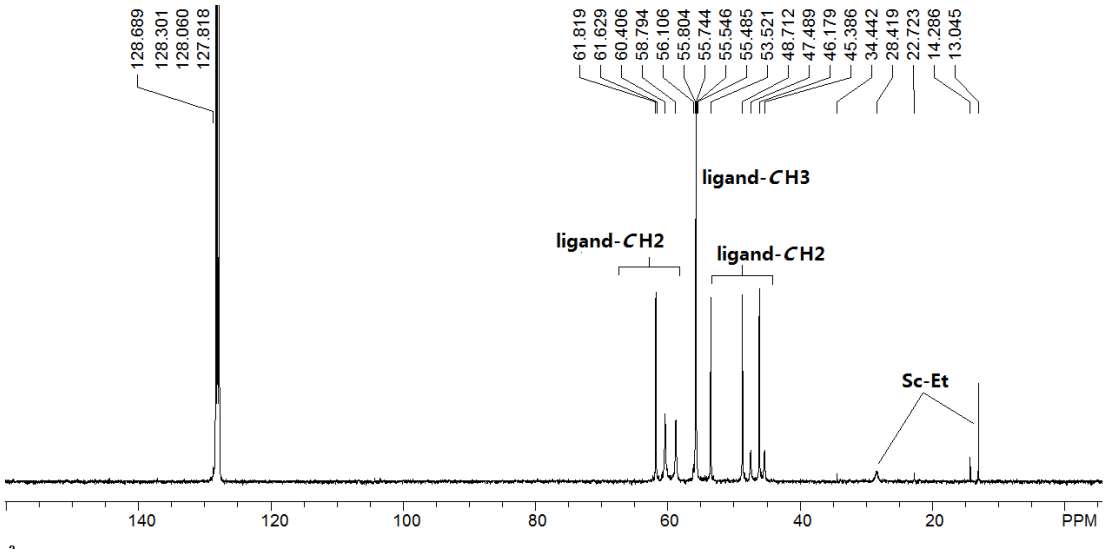

$^{11}$B NMR spectrum of **3** in THF-$d_8$ at 25 °C.



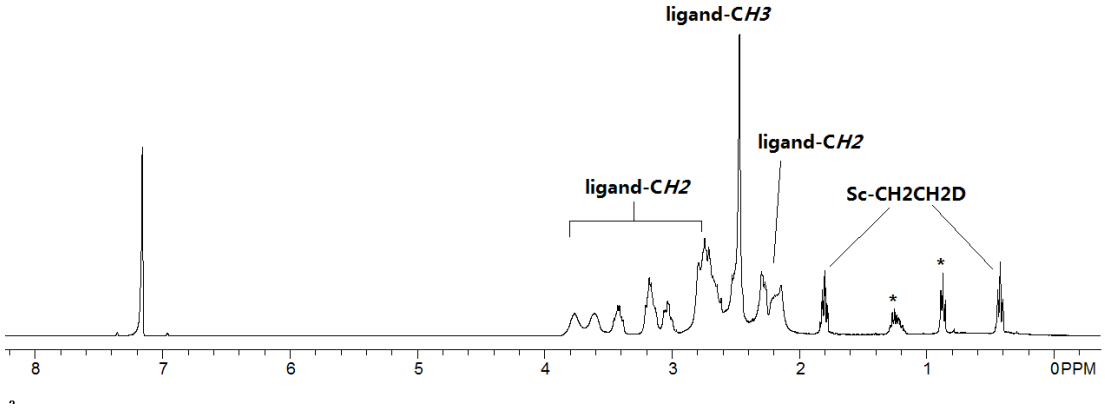

$^1$H NMR spectrum of **4** in THF-$d_8$ at 25 °C.




$^1$H NMR spectrum of **5** in $C_6D_6$ at 25 °C.




$^{13}C\{^{1}H\}$ NMR spectrum of **5** in $C_6D_6$ at 25 °C.




$^{1}H$ NMR spectrum of **5-D$_2$** in $C_6D_6$ at 25 °C. * denotes pentane.

***X-ray Crystallography.*** Data were collected on a Bruker CCD area-detector diffractometer with Mo $K_\alpha$ radiation (graphite monochromator, $\lambda = 0.71073$ Å) using ω scans. The SMART program package was used for the data collection and unit cell determination;[3a] processing of the raw frame data was performed using SAINT; absorption corrections were applied with SADABS.[3b] The structures were solved by direct methods and refined against $F^2$ using all reflections with the SHELXL-97 software as implemented in the program WinGX.[3c-e] Non-hydrogen atoms were refined anisotropically, and hydrogen atoms were placed in calculated positions. Compound **2** contains severely disordered hexane molecule that was included in the refinement with three split positions, C-C distance restraints and restrained isotropic displacement parameters. Compound **4** shows crystallographic $C_3$ symmetry and three crystallographically independent cationic fragments. The crystallographically imposed symmetry leads to disorder in the $C_6H_5S$ anion fragments. In this compound, the hydride atoms H1A, H1B, H2A, H2B, H3A, H3B were located in a Difference Fourier map and fixed in their positions. This structure includes thf molecule in the crystal lattice which is disordered against a $C_6H_5S$ anion. The atoms of the disordered groups could only be refined with isotropic displacement parameters. One of the three crystallographically independent cationic fragments shows a small disorder (C26, C27, C30). These atoms were included in the refinement with split positions and isotropic displacement parameters. Due to the strong disorder and the associated low quality, details of the molecular structure are not discussed for compound **4**. Crystal parameters and refinement results are given in Table 1. CCDC 961424 (**2**), 961425 (**5**), 961426 (**4**) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif .

**Table 1**. Crystallographic and Refinement Data for **2**, **4** and **5**.

| | **2** | **4** | **5** |
|---|---|---|---|
| formula | $(C_{60}H_{138}N_{24}Sc_6)\cdot(C_6H_{14})$ | $3[(C_{30}H_{68}N_{12}Sc_3)(C_6H_5S)]$ $\cdot 3(C_4H_8O)$ | $C_{22}H_{50}N_8Sc_2$ |
| fw | 1551.88 | 2739.32 | 516.62 |
| cryst size/mm | 0.29 × 0.23 × 0.14 | 0.35 × 0.25 × 0.22 | 0.35 × 0.20 × 0.18 |
| cryst syst. | monoclinic | trigonal | monoclinic |
| space group | $P2_1/c$ | $P6_3$ | $P2_1/c$ |
| $T$, K | 100(2) | 100(2) | 100(2) |
| $a$, Å | 18.0667(9) | 19.2442(17) | 14.1746(9) |
| $b$, Å | 19.2883(10) | 19.2442(17) | 11.2268(7) |
| $c$, Å | 28.4424(11) | 21.4089(19) | 17.1826(10) |
| $\alpha$, deg | | | |
| $\beta$, deg | 125.387(2) | | 90.8428(10) |
| $\gamma$, deg | | 120.00 | |
| $U$, Å$^3$ | 8080.4(7) | 6866.3(11) | 2734.1(3) |
| Z | 4 | 2 | 4 |
| $D_{calcd}$, (mg/m$^3$) | 1.276 | 1.325 | 1.255 |
| $F$(000) | 3368 | 2952 | 1120 |
| $\theta$ range deg | 1.48 - 25.10 | 1.90 - 26.46 | 1.44 - 28.43 |
| refns collected | 86450 | 82918 | 37144 |
| indep reflns ($R_{int}$) | 14355 (0.0935) | 9437 (0.0599) | 6838 (0.0834) |
| reflns obsd [$I > 2\sigma(I)$] | 10391 | 8291 | 5477 |
| data/restraints/params | 14355 /27/892 | 9437 / 1 / 492 | 37144 / 0/ 298 |
| $R_1$, $wR_2$ ($I > 2\sigma$(I)) | 0.0526, 0.1250 | 0.0665, 0.1758 | 0.0433, 0.1118 |
| $R_1$, $wR_2$ (all data) | 0.0793, 0.1414 | 0.0743, 0.1845 | 0.0542, 0.1187 |
| goodness-of-fit on $F^2$ | 1.039 | 1.041 | 1.002 |
| $\Delta\rho_{max, min}$, eÅ$^{-3}$ | 1.275, -0.762 | 0.418, -0.354 | 1.901, -0.633 |

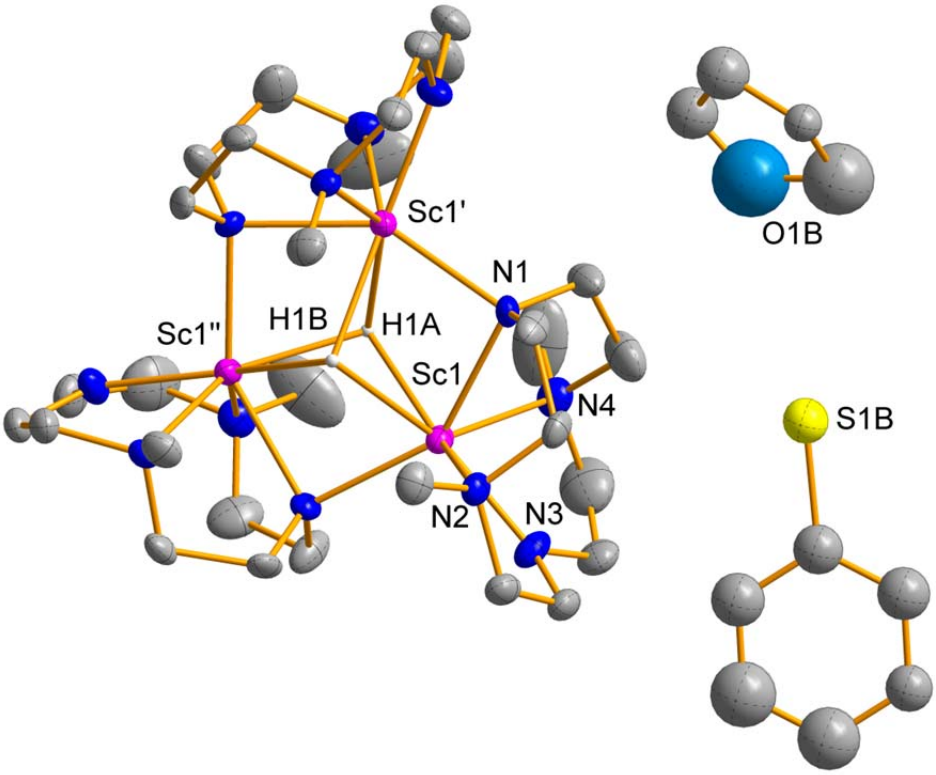



**Figure S1**. ORTEP representation of the molecular structure of **4** in the crystal. The $C_6H_5S$ anion that is disordered against a thf molecule is only shown with one split position.

***Computational Studies***. All calculations were performed using GAUSSIAN09 code.[4] The equilibrium and transition structures were fully optimized at the Becke's 3-parameter hybrid functional [5] combined with the non-local correlation functional provided by Perdew/Wang.[6] Scandium and phosphorous atoms were represented with the relativistic effective core potential SDD, augmented by *f* and *d* polarization functions respectively.[7,8] For the other atoms 6-31G(d,p) basis set were used.[9] The natural population analysis (NPA) was performed using Weinhold's methodology.[10]

Cartesian coordinates of optimizes structures:

**Cation in 2 $[(1,7\text{-}Me_2TACD)_3Sc_3H_2]^+$:**

scf done: -1981.037237

| | | | |
|---|---|---|---|
| Sc | -0.569403 | 8.658562 | 16.104976 |
| Sc | -3.575613 | 9.555528 | 17.096001 |
| Sc | -1.385130 | 11.853064 | 16.303103 |
| N | -2.414091 | 7.655302 | 16.829888 |
| N | 0.243956 | 7.635848 | 18.209233 |
| N | 0.889147 | 7.241443 | 15.609799 |
| N | -1.342275 | 7.362018 | 14.048423 |
| N | -3.456905 | 11.788701 | 17.179798 |
| N | -5.159412 | 10.384788 | 15.262930 |
| N | -5.459701 | 8.866407 | 17.594742 |
| N | -3.906857 | 10.000535 | 19.546295 |
| N | 0.418077 | 10.635167 | 15.796049 |
| N | -1.140469 | 12.266945 | 13.872764 |
| N | -1.005296 | 13.890551 | 16.023511 |
| N | 0.176463 | 12.570947 | 18.068162 |
| C | 0.947968 | 6.483720 | 14.382627 |
| H | 0.779976 | 5.405423 | 14.583871 |
| H | 1.940819 | 6.523994 | 13.898710 |
| C | -0.080630 | 6.969856 | 13.386243 |
| H | 0.287443 | 7.863198 | 12.867302 |
| H | -0.264003 | 6.203180 | 12.615576 |
| C | -1.983585 | 6.248703 | 14.789317 |
| H | -1.208071 | 5.696321 | 15.322924 |
| H | -2.469016 | 5.550889 | 14.086179 |
| C | -2.999741 | 6.788514 | 15.801288 |
| H | -3.780134 | 7.338167 | 15.250807 |
| H | -3.524591 | 5.935534 | 16.255697 |

| | | | |
|---|---|---|---|
| C | -2.141730 | 6.960097 | 18.093604 |
| H | -2.804601 | 6.090839 | 18.215986 |
| H | -2.376011 | 7.625838 | 18.936296 |
| C | -0.692800 | 6.486329 | 18.243079 |
| H | -0.562592 | 5.918557 | 19.179664 |
| H | -0.443824 | 5.822136 | 17.413110 |
| C | 1.637600 | 7.226986 | 17.928361 |
| H | 2.243208 | 8.139267 | 17.971670 |
| H | 2.012857 | 6.549830 | 18.712570 |
| C | 1.759261 | 6.591407 | 16.561577 |
| H | 2.822813 | 6.620437 | 16.262458 |
| H | 1.518965 | 5.510288 | 16.633471 |
| C | 0.242234 | 8.295675 | 19.519132 |
| H | -0.707458 | 8.790516 | 19.699267 |
| H | 1.038009 | 9.041906 | 19.552530 |
| H | 0.435887 | 7.571757 | 20.325915 |
| C | -2.257852 | 7.793860 | 12.986816 |
| H | -1.805897 | 8.594895 | 12.400416 |
| H | -3.193172 | 8.150647 | 13.406287 |
| H | -2.476958 | 6.961660 | 12.299551 |
| C | -4.549058 | 12.472318 | 16.484481 |
| H | -5.462993 | 12.481553 | 17.101077 |
| H | -4.307997 | 13.533652 | 16.299599 |
| C | -4.857625 | 11.824622 | 15.149358 |
| H | -3.976963 | 11.910440 | 14.505480 |
| H | -5.680652 | 12.358591 | 14.646233 |
| C | -6.507420 | 10.132027 | 15.829412 |
| H | -7.270448 | 10.185623 | 15.035389 |
| H | -6.737653 | 10.909732 | 16.563129 |
| C | -6.471230 | 8.793973 | 16.552160 |
| H | -7.468854 | 8.567846 | 16.956475 |
| H | -6.260225 | 7.985047 | 15.834342 |
| C | -6.059721 | 8.944785 | 18.904367 |
| H | -6.662126 | 8.052823 | 19.153403 |
| H | -6.765760 | 9.796621 | 18.989149 |
| C | -4.979898 | 9.066423 | 19.958765 |
| H | -4.503866 | 8.089016 | 20.095297 |
| H | -5.404862 | 9.366723 | 20.930289 |
| C | -4.387215 | 11.398358 | 19.448540 |
| H | -4.501009 | 11.834732 | 20.454883 |
| H | -5.376553 | 11.387454 | 18.982809 |
| C | -3.439938 | 12.227731 | 18.579596 |
| H | -3.706810 | 13.292516 | 18.668771 |
| H | -2.424662 | 12.136201 | 18.982834 |

| | | | |
|---|---|---|---|
| C | -5.112542 | 9.797147 | 13.919372 |
| H | -5.370934 | 8.735938 | 13.959794 |
| H | -4.108704 | 9.902862 | 13.510572 |
| H | -5.825829 | 10.290466 | 13.240432 |
| C | -2.852048 | 9.932709 | 20.564619 |
| H | -1.988239 | 10.524646 | 20.266466 |
| H | -2.535734 | 8.896625 | 20.701855 |
| H | -3.216856 | 10.301187 | 21.536153 |
| C | -0.306093 | 14.731348 | 16.965796 |
| H | -0.833213 | 15.680341 | 17.173136 |
| H | 0.678272 | 15.041812 | 16.557851 |
| C | -0.107330 | 14.007881 | 18.280927 |
| H | -1.026644 | 14.057981 | 18.874389 |
| H | 0.691438 | 14.479656 | 18.875393 |
| C | 1.443462 | 12.346022 | 17.330107 |
| H | 2.306489 | 12.525663 | 17.993113 |
| H | 1.494198 | 13.070702 | 16.514850 |
| C | 1.496234 | 10.930627 | 16.746597 |
| H | 2.483377 | 10.786648 | 16.282835 |
| H | 1.456962 | 10.213376 | 17.578836 |
| C | 0.799489 | 10.807447 | 14.389587 |
| H | 0.384209 | 9.982312 | 13.788253 |
| H | 1.888697 | 10.729070 | 14.259814 |
| C | 0.333482 | 12.132430 | 13.781131 |
| H | 0.784155 | 12.957375 | 14.336200 |
| H | 0.658746 | 12.214972 | 12.730266 |
| C | -1.594453 | 13.660122 | 13.674872 |
| H | -2.689280 | 13.641819 | 13.725183 |
| H | -1.321379 | 14.023516 | 12.671091 |
| C | -1.041004 | 14.570914 | 14.750906 |
| H | -0.026380 | 14.912659 | 14.459635 |
| H | -1.649853 | 15.492925 | 14.770486 |
| C | 0.266690 | 11.939824 | 19.389459 |
| H | 0.320655 | 10.860146 | 19.291909 |
| H | -0.618010 | 12.188433 | 19.979778 |
| H | 1.147876 | 12.301995 | 19.941400 |
| C | -1.758220 | 11.441613 | 12.831400 |
| H | -2.840833 | 11.577279 | 12.838801 |
| H | -1.546403 | 10.391554 | 13.011532 |
| H | -1.389130 | 11.722207 | 11.832612 |
| H | -1.495622 | 10.045850 | 17.548287 |
| H | -2.192435 | 9.993823 | 15.479921 |

**Anion in 2 $[(1,7\text{-}Me_2TACD)_3Sc_3H_4]^-$:**

scf done: -1982.346039

| | | | |
|---|---|---|---|
| Sc | -11.434917 | 10.416609 | 21.368691 |
| H | -11.831512 | 8.682772 | 22.404019 |
| H | -12.747108 | 9.169420 | 20.455510 |
| Sc | -11.907559 | 7.406348 | 20.859465 |
| Sc | -13.433247 | 4.730043 | 21.038791 |
| H | -13.703879 | 6.668726 | 20.367348 |
| H | -12.628789 | 6.146950 | 22.226874 |
| N | -10.874045 | 11.475479 | 23.136294 |
| N | -13.571546 | 11.145052 | 22.372724 |
| N | -12.139493 | 11.884899 | 19.980553 |
| N | -9.367940 | 12.279129 | 20.768237 |
| N | -10.122614 | 8.853662 | 20.458813 |
| N | -9.943403 | 6.456032 | 22.096512 |
| N | -11.539513 | 5.264098 | 19.979275 |
| N | -11.795630 | 7.695121 | 18.391482 |
| N | -13.308510 | 3.254156 | 22.574028 |
| N | -13.190919 | 2.130071 | 19.864504 |
| N | -15.168533 | 4.248054 | 19.878466 |
| N | -15.285284 | 5.275791 | 22.570710 |
| C | -11.883806 | 12.187013 | 23.868787 |
| H | -12.014473 | 13.242450 | 23.531129 |
| H | -11.651630 | 12.274313 | 24.953010 |
| C | -13.218036 | 11.462980 | 23.764438 |
| H | -13.122405 | 10.503608 | 24.284318 |
| H | -14.021837 | 12.042015 | 24.257881 |
| C | -13.954424 | 12.321534 | 21.569447 |
| H | -13.391951 | 13.188928 | 21.931645 |
| H | -15.030636 | 12.545761 | 21.703162 |
| C | -13.569228 | 12.071907 | 20.102609 |
| H | -14.095562 | 11.172818 | 19.750991 |
| H | -13.943540 | 12.913227 | 19.486258 |
| C | -11.472654 | 13.132768 | 19.738788 |
| H | -11.671781 | 13.909291 | 20.514930 |
| H | -11.806342 | 13.610642 | 18.791344 |
| C | -9.966588 | 12.952901 | 19.611931 |
| H | -9.772257 | 12.318573 | 18.738179 |
| H | -9.481406 | 13.932497 | 19.425152 |
| C | -9.403743 | 13.077137 | 22.004593 |
| H | -10.258749 | 13.758573 | 21.965514 |
| H | -8.491724 | 13.701534 | 22.088327 |
| C | -9.596797 | 12.138327 | 23.201940 |
| H | -8.772521 | 11.404117 | 23.201356 |
| H | -9.468556 | 12.717727 | 24.137156 |

| | | | |
|---|---|---|---|
| C | -14.639347 | 10.147441 | 22.352140 |
| H | -15.539766 | 10.514773 | 22.879082 |
| H | -14.277324 | 9.235265 | 22.826148 |
| H | -14.897085 | 9.897217 | 21.322544 |
| C | -7.990744 | 11.908328 | 20.473838 |
| H | -7.366147 | 12.791784 | 20.236847 |
| H | -7.954152 | 11.223553 | 19.623077 |
| H | -7.544369 | 11.402798 | 21.334539 |
| C | -8.995770 | 8.610485 | 21.349731 |
| H | -9.190643 | 9.078592 | 22.327542 |
| H | -8.060546 | 9.064622 | 20.977568 |
| C | -8.744496 | 7.118744 | 21.566844 |
| H | -8.499535 | 6.666074 | 20.600066 |
| H | -7.875730 | 6.947680 | 22.232119 |
| C | -9.943842 | 5.012240 | 21.812495 |
| H | -10.732914 | 4.565359 | 22.428598 |
| H | -8.980289 | 4.554138 | 22.106707 |
| C | -10.248267 | 4.715493 | 20.351846 |
| H | -9.433161 | 5.112652 | 19.715507 |
| H | -10.193096 | 3.616938 | 20.220911 |
| C | -11.752416 | 5.229944 | 18.536466 |
| H | -12.829350 | 5.152711 | 18.329162 |
| H | -11.286503 | 4.344721 | 18.066069 |
| C | -11.195059 | 6.471928 | 17.843062 |
| H | -10.114624 | 6.513092 | 18.018062 |
| H | -11.341197 | 6.422356 | 16.746370 |
| C | -10.944411 | 8.873772 | 18.158431 |
| H | -11.543037 | 9.762856 | 18.390059 |
| H | -10.636434 | 8.929314 | 17.096750 |
| C | -9.722877 | 8.885968 | 19.063687 |
| H | -9.056070 | 8.039818 | 18.807205 |
| H | -9.139870 | 9.790781 | 18.801291 |
| C | -10.057768 | 6.671384 | 23.542241 |
| H | -10.996549 | 6.230557 | 23.883267 |
| H | -10.099009 | 7.739531 | 23.760099 |
| H | -9.207284 | 6.212429 | 24.080246 |
| C | -13.120931 | 7.925819 | 17.808481 |
| H | -13.047556 | 8.106602 | 16.719926 |
| H | -13.567356 | 8.792195 | 18.300125 |
| H | -13.768065 | 7.068251 | 17.997723 |
| C | -12.758834 | 1.956482 | 22.272529 |
| H | -12.826627 | 1.259941 | 23.131311 |
| H | -11.681400 | 2.031711 | 22.045779 |
| C | -13.461111 | 1.323574 | 21.064587 |

| | | | |
|---|---|---|---|
| H | -13.162422 | 0.269123 | 20.898243 |
| H | -14.537840 | 1.327290 | 21.258755 |
| C | -14.309039 | 2.170120 | 18.917755 |
| H | -13.949719 | 2.694989 | 18.024020 |
| H | -14.605214 | 1.147857 | 18.605633 |
| C | -15.517659 | 2.923979 | 19.453378 |
| H | -16.266090 | 2.929320 | 18.630349 |
| H | -15.999066 | 2.303368 | 20.247156 |
| C | -16.324928 | 4.962500 | 20.374199 |
| H | -17.263457 | 4.658706 | 19.869468 |
| H | -16.207197 | 6.039277 | 20.183993 |
| C | -16.480251 | 4.743778 | 21.887384 |
| H | -17.398762 | 5.202262 | 22.302953 |
| H | -16.536925 | 3.666441 | 22.076597 |
| C | -14.936577 | 4.564092 | 23.809510 |
| H | -14.154547 | 5.158059 | 24.294374 |
| H | -15.803127 | 4.510907 | 24.495644 |
| C | -14.371356 | 3.177386 | 23.537925 |
| H | -14.038419 | 2.769146 | 24.517532 |
| H | -15.202134 | 2.500721 | 23.227617 |
| C | -11.995307 | 1.632225 | 19.200231 |
| H | -11.156247 | 1.606339 | 19.900841 |
| H | -11.722369 | 2.285838 | 18.368037 |
| H | -12.137801 | 0.607373 | 18.804311 |
| C | -15.452172 | 6.700104 | 22.850885 |
| H | -15.686414 | 7.234607 | 21.929365 |
| H | -14.507983 | 7.095043 | 23.229971 |
| H | -16.259287 | 6.872300 | 23.587078 |

**Neutral Tetramer [(1,7-$Me_2$TACD)$_4$$Sc_4H_4$]:**

SCF Done:-2642.2770736

| | | | | |
|---|---|---|---|---|
| Sc | 19.702248 | 2.768172 | 4.024575 | 0.670368 |
| Sc | 22.998585 | 3.535758 | 4.443875 | 0.676786 |
| H | 19.921306 | 4.205890 | 5.512805 | -0.171364 |
| H | 21.555790 | 2.093676 | 4.534841 | -0.169666 |
| N | 18.177309 | 2.421411 | 5.710782 | -0.643174 |
| N | 19.356660 | 0.308394 | 4.106975 | -0.481953 |
| N | 18.807367 | 2.099540 | 2.205280 | -0.543481 |
| N | 17.812502 | 4.330084 | 3.381133 | -0.470485 |
| N | 21.358188 | 4.114244 | 2.994931 | -0.645049 |
| N | 23.218976 | 6.335387 | 4.012517 | -0.427330 |
| N | 24.810707 | 4.089002 | 3.499968 | -0.558630 |
| N | 23.339063 | 1.964497 | 2.340710 | -0.449247 |

| | | | | |
|---|---|---|---|---|
| C | 17.845059 | 1.012072 | 5.898386 | -0.071980 |
| H | 16.821075 | 0.880526 | 6.287717 | 0.111402 |
| H | 18.510558 | 0.581876 | 6.652866 | 0.123301 |
| C | 17.984259 | 0.185045 | 4.626168 | -0.088105 |
| H | 17.743383 | -0.878451 | 4.813292 | 0.112820 |
| H | 17.294028 | 0.553020 | 3.863523 | 0.162909 |
| C | 19.454270 | -0.190314 | 2.731213 | -0.106530 |
| H | 19.087642 | -1.229385 | 2.654812 | 0.126660 |
| H | 20.516421 | -0.209186 | 2.469896 | 0.134514 |
| C | 18.694652 | 0.726884 | 1.786293 | -0.088840 |
| H | 19.074050 | 0.550146 | 0.758529 | 0.107353 |
| H | 17.628238 | 0.411830 | 1.741237 | 0.093813 |
| C | 17.942739 | 2.893535 | 1.374295 | -0.095288 |
| H | 16.930723 | 2.435456 | 1.311099 | 0.094251 |
| H | 18.278621 | 2.957025 | 0.318917 | 0.104650 |
| C | 17.812218 | 4.308384 | 1.909874 | -0.096467 |
| H | 18.664932 | 4.915246 | 1.588337 | 0.141829 |
| H | 16.901744 | 4.788459 | 1.507769 | 0.119849 |
| C | 16.705792 | 3.546526 | 3.967754 | -0.099335 |
| H | 16.608569 | 2.618498 | 3.401917 | 0.166870 |
| H | 15.753541 | 4.101176 | 3.871903 | 0.112761 |
| C | 16.984738 | 3.236778 | 5.439232 | -0.038667 |
| H | 17.060871 | 4.182136 | 5.990207 | 0.141217 |
| H | 16.085585 | 2.755749 | 5.857059 | 0.113753 |
| C | 20.283118 | -0.408594 | 4.974696 | -0.219775 |
| H | 20.384866 | 0.117062 | 5.924102 | 0.178359 |
| H | 21.267532 | -0.416298 | 4.507405 | 0.130820 |
| H | 19.959116 | -1.450088 | 5.147458 | 0.122188 |
| C | 17.701309 | 5.722227 | 3.814307 | -0.213066 |
| H | 18.537650 | 6.304977 | 3.425822 | 0.141736 |
| H | 17.716451 | 5.777987 | 4.901004 | 0.136173 |
| H | 16.761595 | 6.174326 | 3.452368 | 0.122124 |
| C | 21.121023 | 5.555897 | 2.976529 | -0.062725 |
| H | 20.643360 | 5.834375 | 3.923128 | 0.134872 |
| H | 20.422162 | 5.840187 | 2.167337 | 0.101912 |
| C | 22.389085 | 6.394445 | 2.801015 | -0.101850 |
| H | 22.969414 | 6.007528 | 1.960668 | 0.132456 |
| H | 22.137804 | 7.441659 | 2.552015 | 0.113589 |
| C | 24.645553 | 6.506075 | 3.722445 | -0.101388 |
| H | 25.165730 | 6.562496 | 4.685610 | 0.132434 |
| H | 24.831706 | 7.461661 | 3.196243 | 0.111081 |
| C | 25.209435 | 5.345976 | 2.928300 | -0.078193 |
| H | 24.918895 | 5.456244 | 1.859216 | 0.089620 |
| H | 26.311181 | 5.465468 | 2.923651 | 0.103300 |

| | | | | |
|---|---|---|---|---|
| C | 25.475174 | 2.992784 | 2.824034 | -0.083435 |
| H | 25.675215 | 2.167687 | 3.526424 | 0.121236 |
| H | 26.458480 | 3.285640 | 2.415776 | 0.101070 |
| C | 24.570810 | 2.480272 | 1.710022 | -0.087644 |
| H | 24.315948 | 3.319513 | 1.055340 | 0.139701 |
| H | 25.039109 | 1.704257 | 1.079595 | 0.110718 |
| C | 22.130997 | 2.234237 | 1.548603 | -0.085857 |
| H | 21.331526 | 1.597594 | 1.931535 | 0.154363 |
| H | 22.288271 | 1.954797 | 0.491035 | 0.121929 |
| C | 21.658997 | 3.674199 | 1.628656 | -0.048638 |
| H | 22.420327 | 4.316494 | 1.152544 | 0.100798 |
| H | 20.764653 | 3.746779 | 0.984028 | 0.138537 |
| C | 22.803938 | 7.394288 | 4.927181 | -0.227585 |
| H | 23.110576 | 8.388637 | 4.555208 | 0.118514 |
| H | 23.240226 | 7.243516 | 5.916736 | 0.132604 |
| H | 21.719819 | 7.398616 | 5.022655 | 0.133681 |
| C | 23.477721 | 0.522716 | 2.534532 | -0.225906 |
| H | 23.478093 | -0.018217 | 1.572200 | 0.121097 |
| H | 22.654094 | 0.155349 | 3.148103 | 0.136755 |
| H | 24.414063 | 0.300046 | 3.050419 | 0.145344 |
| Sc | 22.708339 | 2.676101 | 7.751240 | 0.632500 |
| Sc | 19.382508 | 3.516979 | 7.364726 | 0.609045 |
| H | 22.468872 | 4.144171 | 6.303064 | -0.181241 |
| H | 20.841718 | 2.020713 | 7.151942 | -0.203646 |
| N | 24.159387 | 2.229614 | 6.042277 | -0.645062 |
| N | 23.148052 | -0.039146 | 7.874331 | -0.439826 |
| N | 23.693576 | 2.110734 | 9.561595 | -0.544896 |
| N | 24.620272 | 4.224172 | 8.178681 | -0.474297 |
| N | 21.109279 | 3.960370 | 8.825822 | -0.647190 |
| N | 19.239511 | 6.025465 | 7.763250 | -0.474954 |
| N | 17.613106 | 3.964394 | 8.407800 | -0.545696 |
| N | 19.093252 | 1.798224 | 9.433311 | -0.441159 |
| C | 24.317187 | 0.787324 | 5.873340 | -0.062094 |
| H | 25.198307 | 0.551946 | 5.249038 | 0.105044 |
| H | 23.446317 | 0.405423 | 5.329182 | 0.117247 |
| C | 24.449181 | -0.002670 | 7.181609 | -0.099752 |
| H | 24.810698 | -1.030097 | 6.988405 | 0.111596 |
| H | 25.185741 | 0.480357 | 7.828194 | 0.159353 |
| C | 23.248714 | -0.298394 | 9.313904 | -0.108281 |
| H | 23.732316 | -1.274503 | 9.506518 | 0.116020 |
| H | 22.222150 | -0.381430 | 9.688460 | 0.136094 |
| C | 23.989833 | 0.792679 | 10.055683 | -0.088491 |
| H | 23.753540 | 0.673143 | 11.133374 | 0.104637 |
| H | 25.084416 | 0.590270 | 9.997736 | 0.090600 |

| | | | | |
|---|---|---|---|---|
| C | 24.506283 | 3.029748 | 10.324506 | -0.099007 |
| H | 25.526727 | 2.613438 | 10.464926 | 0.098365 |
| H | 24.134741 | 3.191320 | 11.356381 | 0.108609 |
| C | 24.631121 | 4.381736 | 9.639542 | -0.092729 |
| H | 23.788480 | 5.028968 | 9.896533 | 0.136500 |
| H | 25.548637 | 4.897244 | 9.976252 | 0.122216 |
| C | 25.720102 | 3.360397 | 7.706585 | -0.090441 |
| H | 25.790046 | 2.510935 | 8.386443 | 0.165787 |
| H | 26.679980 | 3.908403 | 7.750682 | 0.113982 |
| C | 25.469094 | 2.877020 | 6.281246 | -0.084207 |
| H | 25.579015 | 3.719072 | 5.585313 | 0.188838 |
| H | 26.290890 | 2.187158 | 6.029530 | 0.113343 |
| C | 22.350304 | -1.123789 | 7.305506 | -0.222739 |
| H | 22.332049 | -1.058018 | 6.220708 | 0.134288 |
| H | 21.324252 | -1.077907 | 7.675358 | 0.133763 |
| H | 22.773637 | -2.106927 | 7.577957 | 0.120365 |
| C | 24.723464 | 5.542118 | 7.556621 | -0.213950 |
| H | 23.917430 | 6.184065 | 7.917168 | 0.135177 |
| H | 24.632539 | 5.453159 | 6.475050 | 0.168989 |
| H | 25.686875 | 6.022085 | 7.801731 | 0.119902 |
| C | 21.322578 | 5.400473 | 8.915204 | -0.075335 |
| H | 21.883352 | 5.725614 | 8.032406 | 0.133669 |
| H | 21.930592 | 5.675361 | 9.796343 | 0.113759 |
| C | 20.020317 | 6.190029 | 9.001147 | -0.093760 |
| H | 19.422551 | 5.817910 | 9.836784 | 0.147921 |
| H | 20.213459 | 7.261381 | 9.191778 | 0.116527 |
| C | 17.817672 | 6.326821 | 7.982708 | -0.103902 |
| H | 17.333986 | 6.335055 | 6.999473 | 0.136685 |
| H | 17.689463 | 7.332822 | 8.420754 | 0.121787 |
| C | 17.168638 | 5.257967 | 8.844772 | -0.081349 |
| H | 17.385168 | 5.461464 | 9.917755 | 0.093317 |
| H | 16.071270 | 5.386100 | 8.756195 | 0.103380 |
| C | 16.947233 | 2.889125 | 9.111442 | -0.087169 |
| H | 16.642388 | 2.088865 | 8.416356 | 0.116827 |
| H | 16.019577 | 3.222377 | 9.606774 | 0.104496 |
| C | 17.903202 | 2.306295 | 10.144715 | -0.088792 |
| H | 18.206382 | 3.109865 | 10.822809 | 0.138083 |
| H | 17.449966 | 1.509803 | 10.760990 | 0.108899 |
| C | 20.338116 | 2.016433 | 10.180181 | -0.091686 |
| H | 21.111529 | 1.396576 | 9.719852 | 0.153467 |
| H | 20.231647 | 1.669522 | 11.224607 | 0.121055 |
| C | 20.814805 | 3.455932 | 10.170713 | -0.048397 |
| H | 20.058769 | 4.079637 | 10.678935 | 0.103571 |
| H | 21.714940 | 3.491965 | 10.808714 | 0.145226 |

| | | | | |
|---|---|---|---|---|
| C | 19.786114 | 6.907602 | 6.738069 | -0.210395 |
| H | 19.722641 | 7.967469 | 7.042320 | 0.121824 |
| H | 19.250653 | 6.771889 | 5.799420 | 0.136826 |
| H | 20.826959 | 6.646308 | 6.564726 | 0.146870 |
| C | 18.933118 | 0.367359 | 9.185488 | -0.239223 |
| H | 18.994996 | -0.217968 | 10.119907 | 0.118717 |
| H | 19.714010 | 0.027485 | 8.501230 | 0.146124 |
| H | 17.960525 | 0.169680 | 8.728892 | 0.147973 |

**Neutral Trimer [(1,7-$Me_2$TACD)$_3$$Sc_3H_3$]:**

scf done: -1981.707609

| | | | |
|---|---|---|---|
| Sc | 8.735358 | 3.649208 | 0.627220 |
| H | 9.177878 | 2.187018 | 2.002730 |
| Sc | 8.891664 | 2.671485 | 3.989199 |
| H | 7.179192 | 1.723314 | 3.754213 |
| Sc | 5.901096 | 1.801255 | 2.277444 |
| N | 5.430086 | 0.774112 | 0.528711 |
| C | 5.477750 | -0.661433 | 0.424999 |
| C | 6.364557 | -1.245911 | 1.515688 |
| N | 6.122914 | -0.594551 | 2.815088 |
| C | 7.165273 | -0.998794 | 3.764030 |
| N | 11.229917 | 1.913474 | 3.990658 |
| C | 11.934600 | 3.169279 | 4.345868 |
| C | 11.298363 | 4.353656 | 3.602682 |
| N | 9.889798 | 4.523215 | 3.930969 |
| C | 9.729037 | 5.203796 | 5.204065 |
| C | 8.251823 | 5.316788 | 5.569590 |
| N | 7.490323 | 4.055431 | 5.370349 |
| C | 6.141456 | 4.377581 | 4.880039 |
| N | 9.322408 | 2.008065 | 5.909531 |
| C | 8.658773 | 2.606296 | 7.038285 |
| C | 7.350927 | 3.241391 | 6.597971 |
| N | 9.416324 | 1.949907 | -0.953641 |
| C | 8.954485 | 0.592297 | -0.643874 |
| N | 7.543314 | 4.073363 | -1.035756 |
| N | 10.608745 | 4.255259 | -0.029055 |
| C | 11.388549 | 3.451258 | -0.932376 |
| C | 10.890092 | 2.009166 | -0.928819 |
| C | 11.796683 | 1.363529 | 2.758121 |
| C | 11.291855 | 0.897903 | 5.057683 |
| C | 10.545776 | 1.365601 | 6.298247 |
| C | 8.858220 | 2.397343 | -2.251665 |
| C | 7.473753 | 3.005682 | -2.012823 |

| | | | |
|---|---|---|---|
| C | 7.960671 | 5.334908 | -1.615642 |
| C | 7.939261 | 6.422628 | -0.543946 |
| N | 8.620083 | 6.028912 | 0.718869 |
| C | 7.919367 | 6.612902 | 1.873411 |
| C | 10.038329 | 6.455222 | 0.792041 |
| C | 10.939875 | 5.651941 | -0.131739 |
| C | 4.404889 | 1.316996 | -0.332399 |
| C | 4.291475 | 2.822990 | -0.155477 |
| N | 4.252883 | 3.248359 | 1.263968 |
| C | 4.482536 | 4.699762 | 1.292946 |
| C | 2.976122 | 2.939971 | 1.959022 |
| C | 2.999249 | 1.603822 | 2.692789 |
| N | 4.223100 | 1.510645 | 3.459933 |
| C | 4.303321 | 0.286860 | 4.233647 |
| C | 4.784557 | -0.895079 | 3.376813 |
| H | 11.202468 | 0.508385 | 2.424739 |
| H | 12.839929 | 1.038431 | 2.910336 |
| H | 11.767626 | 2.121478 | 1.974440 |
| H | 5.560226 | 3.461782 | 4.736044 |
| H | 6.222479 | 4.888705 | 3.918998 |
| H | 5.604486 | 5.028135 | 5.590819 |
| H | 10.389659 | 0.486220 | 6.954353 |
| H | 11.195590 | 2.044798 | 6.891860 |
| H | 10.808345 | -0.003609 | 4.660421 |
| H | 12.336311 | 0.632162 | 5.296047 |
| H | 13.012293 | 3.075528 | 4.124976 |
| H | 11.838433 | 3.324697 | 5.423215 |
| H | 11.399818 | 4.192876 | 2.520097 |
| H | 11.888680 | 5.259771 | 3.836141 |
| H | 10.266905 | 4.702402 | 6.030429 |
| H | 10.134431 | 6.236245 | 5.177956 |
| H | 8.128891 | 5.679507 | 6.602156 |
| H | 7.796236 | 6.063422 | 4.913269 |
| H | 6.913090 | 3.846799 | 7.410226 |
| H | 6.643802 | 2.442002 | 6.354632 |
| H | 9.300425 | 3.363099 | 7.537556 |
| H | 8.413101 | 1.871814 | 7.831216 |
| H | 9.338344 | 0.300192 | 0.336403 |
| H | 9.299605 | -0.131980 | -1.399939 |
| H | 7.861928 | 0.573800 | -0.598168 |
| H | 8.410334 | 6.258789 | 2.783171 |
| H | 6.880154 | 6.280794 | 1.880437 |
| H | 7.941810 | 7.714530 | 1.839821 |
| H | 12.467897 | 3.431095 | -0.679514 |

| | | | |
|---|---|---|---|
| H | 11.354372 | 3.858094 | -1.965877 |
| H | 11.204673 | 1.516788 | -0.004539 |
| H | 11.321919 | 1.441098 | -1.770344 |
| H | 8.842088 | 1.560698 | -2.971023 |
| H | 9.519208 | 3.166383 | -2.660094 |
| H | 6.802393 | 2.217538 | -1.644189 |
| H | 7.054540 | 3.341163 | -2.978785 |
| H | 8.963585 | 5.298432 | -2.082634 |
| H | 7.273723 | 5.665530 | -2.419753 |
| H | 8.362260 | 7.365166 | -0.924998 |
| H | 6.891959 | 6.612744 | -0.290675 |
| H | 10.120553 | 7.536172 | 0.587831 |
| H | 10.347694 | 6.279381 | 1.826490 |
| H | 10.844591 | 6.018009 | -1.175386 |
| H | 11.987156 | 5.874798 | 0.155343 |
| H | 8.144482 | -0.692848 | 3.386523 |
| H | 7.163175 | -2.092177 | 3.912197 |
| H | 7.018212 | -0.502604 | 4.723762 |
| H | 5.422486 | 4.916216 | 0.781604 |
| H | 4.543795 | 5.045750 | 2.327264 |
| H | 3.664631 | 5.243548 | 0.791381 |
| H | 5.864279 | -1.020218 | -0.550818 |
| H | 4.460347 | -1.099436 | 0.496413 |
| H | 7.415185 | -1.058483 | 1.267014 |
| H | 6.235024 | -2.340176 | 1.583187 |
| H | 4.811714 | -1.842213 | 3.943424 |
| H | 4.084981 | -1.032690 | 2.547544 |
| H | 5.001599 | 0.427903 | 5.070429 |
| H | 3.332677 | 0.018665 | 4.688956 |
| H | 2.865321 | 0.778582 | 1.966534 |
| H | 2.101100 | 1.572182 | 3.340577 |
| H | 2.132983 | 3.000578 | 1.252947 |
| H | 2.833167 | 3.720720 | 2.712518 |
| H | 3.406019 | 3.208131 | -0.690099 |
| H | 5.179979 | 3.299782 | -0.584345 |
| H | 3.424550 | 0.833869 | -0.142348 |
| H | 4.605945 | 1.134951 | -1.407206 |
| H | 7.166607 | 3.448469 | 2.029673 |

**Neutral Dimer [(1,7-$Me_2$TACD)$_2$$Sc_2H_2$]:**

scf done: -1321.1342546

| | | | | |
|---|---|---|---|---|
| H | 4.685591 | -2.222504 | 11.102813 | -0.130221 |
| N | 3.039510 | 0.055755 | 11.200623 | -0.625697 |

| | | | | |
|---|---|---|---|---|
| N | 1.267816 | -2.840170 | 13.050737 | -0.568993 |
| C | 1.671495 | -3.935675 | 13.904720 | -0.086010 |
| C | 2.420759 | -5.001218 | 13.091169 | -0.086437 |
| N | 3.564462 | -4.375708 | 12.393434 | -0.482870 |
| C | 4.710265 | -4.279534 | 13.301777 | -0.212597 |
| C | 2.937976 | -4.997241 | 10.061704 | -0.072352 |
| N | 2.394946 | -3.668273 | 9.967812 | -0.567207 |
| C | 1.295188 | -3.625933 | 9.047501 | -0.074374 |
| C | 0.556735 | -2.296719 | 9.152289 | -0.110557 |
| C | -0.466400 | -2.813520 | 11.327221 | -0.091311 |
| H | 1.144076 | -1.509285 | 8.663491 | 0.139279 |
| C | -0.204010 | -0.539877 | 10.594106 | -0.219352 |
| Sc | 2.624210 | -2.309082 | 11.551492 | 0.842103 |
| Sc | 4.999268 | -0.456704 | 11.799245 | 0.282337 |
| N | 5.350675 | -0.331610 | 9.410163 | -0.488875 |
| C | 5.685617 | -1.578943 | 8.697226 | -0.247651 |
| C | 4.023597 | 0.142624 | 8.939450 | -0.103418 |
| C | 3.191764 | 0.868313 | 9.994357 | -0.073499 |
| C | 2.386064 | 0.798519 | 12.285804 | -0.085787 |
| C | 3.335318 | 1.737048 | 13.044720 | -0.097207 |
| N | 4.470377 | 0.999253 | 13.642624 | -0.481900 |
| C | 4.077143 | 0.404132 | 14.925380 | -0.231661 |
| C | 5.664497 | 1.846908 | 13.831733 | -0.110941 |
| C | 6.215243 | 2.332078 | 12.497318 | -0.076462 |
| N | 6.165673 | 1.269733 | 11.526732 | -0.561461 |
| C | 6.457111 | 1.769549 | 10.207501 | -0.072491 |
| C | 6.455157 | 0.630437 | 9.201983 | -0.105319 |
| H | 3.945940 | -1.553676 | 13.012453 | -0.102351 |
| N | 0.381893 | -1.879716 | 10.554782 | -0.479319 |
| C | 3.980344 | -5.088349 | 11.170063 | -0.107581 |
| C | -0.150259 | -2.698530 | 12.820414 | -0.078839 |
| H | 3.648955 | 1.846223 | 10.217627 | 0.131556 |
| H | 2.217868 | 1.101268 | 9.522534 | 0.104173 |
| H | 3.465928 | -0.750904 | 8.642189 | 0.169988 |
| H | 4.136341 | 0.775340 | 8.044485 | 0.129760 |
| H | 6.448363 | 1.012361 | 8.166851 | 0.119633 |
| H | 7.382380 | 0.061599 | 9.331931 | 0.152934 |
| H | 7.446797 | 2.262948 | 10.137857 | 0.106316 |
| H | 5.729446 | 2.551278 | 9.899879 | 0.100787 |
| H | 5.636118 | 3.217026 | 12.157041 | 0.095784 |
| H | 7.237201 | 2.721309 | 12.673353 | 0.110236 |
| H | 6.414348 | 1.220413 | 14.331527 | 0.151347 |
| H | 5.451329 | 2.695901 | 14.503827 | 0.124486 |
| H | 3.739757 | 2.472443 | 12.344676 | 0.152040 |

| | | | | |
|---|---|---|---|---|
| H | 2.788048 | 2.296351 | 13.823002 | 0.123239 |
| H | 1.950556 | 0.079887 | 12.992665 | 0.176567 |
| H | 1.550315 | 1.410170 | 11.907588 | 0.116197 |
| H | 4.898139 | -2.315479 | 8.864177 | 0.209281 |
| H | 6.619522 | -1.987452 | 9.092851 | 0.148670 |
| H | 5.813155 | -1.395739 | 7.617351 | 0.120990 |
| H | 4.890365 | -0.220883 | 15.304774 | 0.152882 |
| H | 3.208307 | -0.239261 | 14.787039 | 0.197203 |
| H | 3.851975 | 1.184799 | 15.671248 | 0.117784 |
| H | 3.424943 | -5.347081 | 9.127802 | 0.098158 |
| H | 2.130863 | -5.740985 | 10.246157 | 0.093798 |
| H | 4.221647 | -6.143624 | 11.389279 | 0.117462 |
| H | 4.898150 | -4.595564 | 10.834065 | 0.165197 |
| H | 2.766056 | -5.846113 | 13.715755 | 0.104827 |
| H | 1.736452 | -5.397060 | 12.335372 | 0.153753 |
| H | 0.810105 | -4.423757 | 14.391675 | 0.096113 |
| H | 2.320512 | -3.594084 | 14.729908 | 0.114872 |
| H | -0.549266 | -1.737935 | 13.201600 | 0.095561 |
| H | -0.736647 | -3.475065 | 13.340871 | 0.097283 |
| H | -0.225674 | -3.830218 | 11.009565 | 0.159145 |
| H | -1.534605 | -2.628726 | 11.108267 | 0.103020 |
| H | -0.415058 | -2.348673 | 8.629580 | 0.120292 |
| H | 0.578502 | -4.457285 | 9.230733 | 0.091471 |
| H | 1.594447 | -3.754689 | 7.986697 | 0.098434 |
| H | 5.083088 | -5.281548 | 13.575345 | 0.114686 |
| H | 4.426889 | -3.744636 | 14.209048 | 0.156741 |
| H | 5.507268 | -3.715630 | 12.811699 | 0.163646 |
| H | 0.460519 | 0.171062 | 10.100051 | 0.140530 |
| H | -0.344218 | -0.220106 | 11.628391 | 0.151267 |
| H | -1.184721 | -0.516219 | 10.089480 | 0.121010 |

**Monmer [(1,7-Me$_2$TACD)ScH]:**

scf done: -660.549160

| | | | |
|---|---|---|---|
| C | 4.542340 | -2.217279 | 11.175920 |
| N | 3.731485 | -1.014600 | 11.408381 |
| C | 2.440126 | -1.358894 | 12.051803 |
| C | 1.432884 | -0.209215 | 11.959138 |
| N | 1.442826 | 0.335027 | 10.627823 |
| Sc | 3.100775 | 0.049218 | 9.452838 |
| N | 2.395262 | 2.134185 | 8.744552 |
| C | 2.592025 | 2.352626 | 7.305033 |
| C | 4.500626 | 0.011910 | 12.155290 |
| C | 5.419833 | 0.806957 | 11.213293 |
| N | 4.673472 | 1.213344 | 10.045576 |

| | | | |
|---|---|---|---|
| C | 4.657908 | 2.609024 | 9.673433 |
| C | 3.202078 | 3.088990 | 9.545560 |
| C | 0.690470 | 1.551468 | 10.476253 |
| C | 0.953166 | 2.150612 | 9.091649 |
| H | 3.157511 | -1.166860 | 7.979748 |
| H | 2.182491 | 3.323093 | 6.985589 |
| H | 3.659011 | 2.330093 | 7.069140 |
| H | 2.103737 | 1.554682 | 6.738557 |
| H | 5.483032 | -1.941170 | 10.692778 |
| H | 4.771189 | -2.737395 | 12.118625 |
| H | 4.008860 | -2.902010 | 10.510690 |
| H | 2.041644 | -2.220911 | 11.503982 |
| H | 2.592975 | -1.671167 | 13.097457 |
| H | 0.449925 | -0.615236 | 12.266796 |
| H | 1.673612 | 0.563289 | 12.720086 |
| H | -0.404367 | 1.411609 | 10.563808 |
| H | 0.947080 | 2.292958 | 11.262195 |
| H | 0.447611 | 1.532618 | 8.340296 |
| H | 0.542070 | 3.170102 | 9.014697 |
| H | 5.058394 | -0.461620 | 12.981469 |
| H | 3.778156 | 0.707554 | 12.590125 |
| H | 5.805499 | 1.668570 | 11.784160 |
| H | 6.310725 | 0.200355 | 10.965821 |
| H | 5.197415 | 2.808422 | 8.729096 |
| H | 5.147495 | 3.248320 | 10.427372 |
| H | 2.766906 | 3.122179 | 10.547812 |
| H | 3.132155 | 4.100116 | 9.109034 |

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