Electronic Supplementary Information for:

Rare Earth Arylsilazido Compounds with Inequivalent Secondary Interactions

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General. All manipulations were performed under a dry argon atmosphere using standard Schlenk techniques or under a nitrogen atmosphere in a glovebox unless otherwise indicated. Water and oxygen were removed from benzene, pentane, and ether solvents using an IT PureSolv system. Benzene-d_6 and toluene-d_8 were heated to reflux over Na/K alloy and vacuum-transferred. $ScCl_3THF_3$ was prepared by reaction of Sc_2O_3 with concentrated HCl followed by dehydration with $SOCl_2$.[1] YCl_3 and $LuCl_3$ were purchased from Strem Chemicals and used as received. Diisopropylaniline and Dimethylchlorosilane were purchased from Sigma-Aldrich and Gelest respectively and distilled, and *n*BuLi was purchased from Sigma-Aldrich and used as received. ^1H, $^{13}C\{^1H\}$, and $^{29}Si\{^1H\}$ HMBC NMR spectra were collected on a Bruker DRX-400 spectrometer or a Bruker Avance III-600 spectrometer. 15N NMR chemical shifts were measured by ^1H-^{15}N HMBC experiments and referenced to nitromethane as an external standard. Infrared spectra were measured on a Bruker Vertex 80, using KBr pellet (transmission mode). Elemental analyses were performed using a Perkin-Elmer 2400 Series II CHN/S. X-ray diffraction data was collected on a Bruker APEX II diffractometer.

HN(SiHMe$_2$)Dipp (**1**). *n*BuLi (21.2 mL, 0.0530 mol) was added dropwise to a solution of H$_2$NDipp (10.0 mL, 0.0530 mol) in pentane (300 mL) cooled to 0 °C. A white precipitate was observed immediately. The reaction mixture was warmed to room temperature and stirred overnight. The white precipitate was isolated by filtration, and then it was dissolved in diethyl ether (500 mL). The solution was cooled to 0 °C and ClSiHMe$_2$ (5.89 mL, 0.0530 mol) was added slowly to the solution. The reaction was warmed to room temperature and stirred for 3 h. The solution was filtered, and the filtrate was evaporated under vacuum to remove the volatile materials. The resulting yellowish solution distilled under dynamic vacuum at 103-105 °C to obtain the desired dimethylsilazane as a colorless product (9.88 g, 0.0420 mol, 83.3%). ^{1}H NMR (benzene-*d*$_6$, 600 MHz, 25 °C): δ 7.12 – 7.08 (3 H, *m*-C$_6$H$_5$ and *p*-C$_6$H$_5$ overlap), 4.89 (virtual octet, 1 H, $^1J_{\mathrm{SiH}}$ = 199.2 Hz, Si*H*Me$_2$), 3.44 (septet, 2 H, $^3J_{\mathrm{HH}}$ = 6.9 Hz, C*H*Me$_2$), 2.19 (s, 1 H, N*H*), 1.21 (d, 12 H, $^3J_{\mathrm{HH}}$ = 6.9 Hz, CH*Me$_2$*), 0.09 (d, 6 H, $^3J_{\mathrm{HH}}$ = 3.0 Hz, SiH*Me*$_2$). ^{13}C{^{1}H} NMR (benzene-*d*$_6$, 150 MHz, 25 °C): δ 144.15 (*o*-C$_6$H$_5$), 139.74 (*ipso*-C$_6$H$_5$), 124.48 (*p*-C$_6$H$_5$), 123.87 (*m*-C$_6$H$_5$), 28.88 (*C*HMe$_2$), 24.27 (CH*Me$_2$*), –1.14 (SiH*Me$_2$*). ^{15}N {^{1}H} NMR (benzene-*d*$_6$, 60.8 MHz, 25 °C): δ –343.6. ^{29}Si{^{1}H} NMR (benzene-*d*$_6$, 119.3 MHz, 25 °C): *δ* –9.9. IR (KBr, cm^{-1}): 3392 w (NH), 3066 w, 2963 s, 2870 m, 2112 m (SiH), 1462 m, 1441 m, 1326 m, 1253 s, 1196 w, 1108 w, 1056 w, 1044 w, 912 s, 831 m, 789 m, 747 m, 665 w.

LiN(SiHMe$_2$)Dipp (**2**). A solution of HN(SiHMe$_2$)Dipp (2.51 g, 0.0107 mol) in pentane (40 mL total volume) was cooled to –78 °C. *n*BuLi (4.26 mL, 0.0107 mol) was added to the solution, which was then stirred for 20 min at –78 °C. The reaction mixture became milky white, and then it was warmed to room temperature and stirred overnight. The solvent was evaporated for 2 h to directly provide the product as a white, highly air-sensitive solid (2.48 g, 0.0103 mol, 97.2%). ^{1}H NMR (benzene-d_6, 600 MHz, 25 °C): δ 7.03 (d, 2 H, $^3J_{HH}$ = 7.6 Hz, *m*-C_6H_5) 6.90 (t, 1 H, $^3J_{HH}$ = 7.6 Hz, *p*-C_6H_5), 5.09 (br m, 1 H, $^1J_{SiH}$ = 177 Hz, Si*H*Me_2), 3.54 (septet, 2 H, $^3J_{HH}$ = 6.7 Hz, C*H*Me_2), 1.13 (br d, 12 H, $^3J_{HH}$ = ~6 Hz, CH*Me*$_2$), 0.07 (d, 6 H, $^3J_{HH}$ = 2.7 Hz, SiH*Me*$_2$). $^{13}C\{^1H\}$ NMR (benzene-d_6, 150 MHz, 25 °C): δ 149.09 (*ipso*-C_6H_5), 142.71 (*o*-C_6H_5), 124.97 (*m*-C_6H_5), 120.67 (*p*-C_6H_5), 27.66 (*C*HMe_2), 25.28 (CH*Me*$_2$), 0.79 (SiH*Me*$_2$). $^{29}Si\{^1H\}$ NMR (benzene-d_6, 119.3 MHz, 25 °C): δ –20.21 (*Si*HMe_2). $^7Li\{^1H\}$ NMR (benzene-d_6, MHz, 25 °C): *δ* 0.99 (*Li*N). IR (KBr, cm^{-1}, small amounts of silazane are formed during measurement): 3390 w (silazane NH), 3050 w, 2961 s, 2868 m, 2108 w (silazane SiH), 2022 s (SiH), 1663 w, 1588 w, 1460 s, 1422 s, 1384 w, 1364 w, 1313 s, 1249 s, 1194 s, 1105 m, 1041 m, 936 s, 905 s, 821 m, 782 s, 751 s, 664 w 640 w. Anal. Calcd for $C_{14}H_{24}LiNSi$: C, 69.66; H, 10.02; N, 5.80. Found: C, 69.67; H, 10.21; N, 5.71. mp 139 – 141 °C.

Sc{N(SiHMe$_2$)Dipp}$_3$ (**3**). A solid mixture of $ScCl_3(THF)_3$ (0.100 g, 0.272 mmol) and LiN(SiHMe$_2$)Dipp (0.197 g, 0.816 mmol) was cooled to –78 °C for 30 min. Diethyl ether (7 mL) was cooled to –78 °C for 30 min in a separate vial. The solvent was added to the solid mixture, and the resulting mixture was stirred at –78 °C for 1 h. The reaction mixture was warmed to room temperature and stirred overnight. The volatile materials were evaporated under reduced pressure, and the remaining white solid was extracted with pentane (3 × 5 mL). The pentane extracts were combined and evaporated to obtain a sticky solid. The residue was extracted with pentane, concentrated, and cooled at –30 °C for 36 h. The pentane was decanted and the vial was dried under vacuum to obtain the desired product as a white crystalline solid (0.0879 g, 0.117 mmol, 43.2%). ^{1}H NMR (benzene-d_6, 600 MHz, 25 °C): δ 7.05 (d, 6 H, $^3J_{HH}$ = 7.6 Hz, *m*-C_6H_5) 6.96 (t, 3 H, $^3J_{HH}$ = 7.5 Hz, *p*-C_6H_5), 5.43 (br s, 3 H, $^1J_{SiH}$ = 142.6 Hz, Si*H*Me$_2$), 3.38 (septet, 6 H, $^3J_{HH}$ = 4.9 Hz, C*H*Me$_2$), 1.18 (d, 36 H, $^3J_{HH}$ = 6.6 Hz, CH*Me$_2$*), 0.27 (broad, 18 H, SiH*Me$_2$*). ^{13}C{^{1}H} NMR (benzene-d_6, 150 MHz, 25 °C): δ 148.81 (*ipso*-C_6H_5), 141.03 (*o*-C_6H_5), 124.64 (*m*-C_6H_5), 122.09 (*p*-C_6H_5), 29.22 (*C*HMe$_2$), 25.28 (CH*Me$_2$*), 1.77 (SiH*Me$_2$*). ^{15}N {^{1}H} NMR (benzene-d_6, 60.8 MHz, 25 °C): δ –221.0. ^{29}Si{^{1}H} NMR (benzene-d_6, 119.3 MHz, 25 °C): δ –28.08. IR (KBr, cm^{-1}): 3061 m, 3046 m, 2963 s, 2865 m, 2105 m (SiH), 2046 m (SiH), 1908 s (SiH), 1587 m, 1457 s, 1427 s, 1360 m, 1326 m, 1309 s, 1251 s, 1194 s, 1118 m, 1047 m, 1001 m, 950 s, 903 s, 878 s, 834 s, 812 s, 782 s, 760 s, 683 m, 670 m, 635 m. Anal. Calcd for $C_{42}H_{72}ScN_3Si_3$: C, 67.42; H, 9.70; N, 5.62. Found: C, 66.93; H, 9.54; N, 5.43. mp 186 – 188 °C.

Y{N(SiHMe$_2$)Dipp}$_3$ (**4**). A solid mixture of YCl_3 (0.0836 g, 0.428 mmol) and LiN(SiHMe$_2$)Dipp (0.310 g, 1.28 mmol) were allowed to react following the procedure of Sc{N(SiHMe$_2$)Dipp}$_3$ described above to provide the desired product as a white crystalline solid (0.190 g, 0.240 mmol, 56.2%). ^{1}H NMR (benzene-d_6, 600 MHz, 25 °C): δ 7.04 (d, 6 H, $^3J_{HH}$ = 7.7 Hz, *m*-C_6H_5) 6.93 (t, 3 H, $^3J_{HH}$ = 7.6 Hz, *p*-C_6H_5), 5.17 (septet, 3 H, $^1J_{SiH}$ = 129.2 Hz, $^3J_{HH}$ = 3 Hz, Si*H*Me$_2$), 3.42 (septet, 6 H, $^3J_{HH}$ = 6.5 Hz, C*H*Me$_2$), 1.16 (d, 36 H, $^3J_{HH}$ = 6.8 Hz, CH*Me$_2$*), 0.33 (d, 18 H, $^3J_{HH}$ = 2.7 Hz, SiH*Me$_2$*). ^{1}H NMR (toluene-d_8, 600 MHz, –68.15 °C): δ 7.26 – 6.76 (9 H), 5.42 (br s, 1 H, $^1J_{SiH}$ = 131.9 Hz, Si*H*Me$_2$), 5.26 (br s, 1 H, $^1J_{SiH}$ = 140.7 Hz, Si*H*Me$_2$), 4.89 (br s, 1 H, $^1J_{SiH}$ = 115.8 Hz, Si*H*Me$_2$), 4.18 (br s, 1 H, C*H*Me$_2$), 4.06 (br s, 1 H, C*H*Me$_2$), 3.59 (br s, 1 H, C*H*Me$_2$), 3.30 (br s, 1 H, C*H*Me$_2$), 3.00 (br s, 1 H, C*H*Me$_2$), 2.34 (br s, 1 H, C*H*Me$_2$), 1.63 (br s, 3 H, CH*Me$_2$*), 1.46 (br s, 3 H, CH*Me$_2$*), 1.43 (br s, 3 H, CH*Me$_2$*), 1.40 (br s, 3 H, CH*Me$_2$*), 1.21 (br s, 3 H, CH*Me$_2$*), 1.18

(br s, 3 H, CH*Me*$_2$), 1.11 (br s, 9 H, CH*Me*$_2$), 0.84 (br s, 3 H, CH*Me*$_2$), 0.77 (br s, 3 H, CH*Me*$_2$), 0.66 (br s, 3 H, CH*Me*$_2$), 0.57 (br s, 3 H, SiH*Me*$_2$), 0.54 (br s, 3 H, SiH*Me*$_2$), 0.42 (br s, 3 H, SiH*Me*$_2$), 0.37 (br s, 3 H, SiH*Me*$_2$), 0.22 (br s, 3 H, SiH*Me*$_2$), –0.06 (br s, 3 H, SiH*Me*$_2$). ^{13}C{^{1}H} NMR (benzene-*d*$_6$, 150 MHz, 25 °C): δ 147.82 (*ipso*-C_6H_5), 140.75 (*o*-C_6H_5), 124.45 (*m*-C_6H_5), 121.14 (*p*-C_6H_5), 28.93 (*C*HMe$_2$), 25.16 (CH*Me*$_2$), 1.96 (SiH*Me*$_2$). ^{15}N{^{1}H} NMR (benzene-*d*$_6$, 60.8 MHz, 25 °C): δ –239.61. ^{29}Si{^{1}H} NMR (benzene-*d*$_6$, 119.3 MHz, 25 °C): δ –28.18. IR (KBr, cm^{-1}): 3062 m, 3045 m, 2962 s, 2865 m, 2107 w (SiH), 1934 s (SiH), 1883 s (SiH), 1588 m, 1458 m, 1383 s, 1359 m, 1327 m, 1296 s, 1250 s, 1177 s, 1143 s, 1116 s, 1044 s, 1017 s, 951 s, 873 s, 833 s, 779 s, 758 s, 672 m, 626 m. Anal. Calcd for $C_{42}H_{72}YN_3Si_3$: C, 63.68; H, 9.16; N, 5.30. Found: C, 63.55; H, 9.55; N, 5.20. mp 193 - 195 °C.

Lu{N(SiHMe$_2$)Dipp}$_3$ (**5**). A solid mixture of LuCl$_3$ (0.235 g, 0.834 mmol) and LiN(SiHMe$_2$)Dipp (0.604 g, 2.50 mmol) were allowed to react, following the above procedure for the synthesis of Sc{N(SiHMe$_2$)Dipp}$_3$, to give the desired product as a white crystalline solid (0.334 g, 0.380 mmol, 45.5%). ^{1}H NMR (benzene-*d*$_6$, 600 MHz, 25 °C): δ 7.04 (d, 6 H, $^{3}J_{HH}$ = 7.5 Hz, *m*-C_6H_5) 6.92 (t, 3 H, $^{3}J_{HH}$ = 7.6 Hz, *p*-C_6H_5), 5.43 (br septet, 3 H, $^{1}J_{SiH}$ = 127.6 Hz, $^{3}J_{HH}$ = 2.9 Hz; Si*H*Me$_2$), 3.46 (septet, 6 H, $^{3}J_{HH}$ = 5.2 Hz, C*H*Me$_2$), 1.17 (d, 36 H, $^{3}J_{HH}$ = 6.7 Hz, CH*Me*$_2$), 0.35 (d, 18 H, $^{3}J_{HH}$ = 1.7 Hz, SiH*Me*$_2$). ^{1}H NMR (toluene-*d*$_8$, 600 MHz, –68.15 °C): δ 7.26 – 6.75 (9 H), 5.59 (br s, 1 H, $^{1}J_{SiH}$ = 132.1 Hz, Si*H*Me$_2$), 5.35 (br s, 2 H, $^{1}J_{SiH}$ = 151.5 Hz, Si*H*Me$_2$), 4.18 (br s, 1 H, C*H*Me$_2$), 4.07 (br s, 1 H, C*H*Me$_2$), 3.64 (br s, 1 H, C*H*Me$_2$), 3.31 (br s, 1 H, C*H*Me$_2$), 2.96 (br s, 1 H, C*H*Me$_2$), 2.51 (br s, 1 H, C*H*Me$_2$), 1.60 (br s, 3 H, CH*Me*$_2$), 1.45 (br s, 3 H, CH*Me*$_2$), 1.42 (br s, 3 H, CH*Me*$_2$), 1.40 (br s, 3 H, CH*Me*$_2$), 1.21 (br s, 6 H, CH*Me*$_2$), 1.17 (br s, 3 H, CH*Me*$_2$), 1.11 (br s, 6 H, CH*Me*$_2$), 0.80 (br s, 3 H, CH*Me*$_2$), 0.76 (br s, 3 H, CH*Me*$_2$), 0.69 (br s, 3 H, CH*Me*$_2$), 0.59 (br s, 3 H, SiH*Me*$_2$), 0.55 (br s, 3 H, SiH*Me*$_2$), 0.48 (br s, 3 H, SiH*Me*$_2$), 0.39 (br s, 3 H, SiH*Me*$_2$), 0.24 (br s, 3 H, SiH*Me*$_2$), –0.08 (br s, 3 H, SiH*Me*$_2$). ^{13}C{^{1}H} NMR (benzene-*d*$_6$, 150 MHz, 25 °C): δ 148.17 (*ipso*-C$_6$H$_5$), 141.23 (*o*-C$_6$H$_5$), 124.46 (*m*-C$_6$H$_5$), 121.28 (*p*-C$_6$H$_5$), 28.89 (*C*HMe$_2$), 25.11 (CH*Me*$_2$), 1.84 (SiH*Me*$_2$). ^{15}N {^{1}H} NMR (benzene-*d*$_6$, 60.8 MHz, 25 °C): δ –241.64. ^{29}Si{^{1}H} NMR (benzene-*d*$_6$, 119.3 MHz, 25 °C): δ –27.62. IR (KBr, cm^{-1}): 3062 m, 3045 m, 2962 s, 2864 s, 2108 w (SiH), 1942 s (SiH), 1877 s (SiH), 1587 m, 1457 s, 1428 s, 1383 m, 1360 m, 1311 s, 1250 s, 1199 s, 1105 m, 1046 s, 951 s, 874 s, 833 s, 780 s, 758 s, 671 m, 628 m. Anal. Calcd for $C_{42}H_{72}LuN_3Si_3$: C, 57.44; H, 8.26; N, 4.78. Found: C, 57.41; H, 8.28; N, 4.72. mp 189 - 191 °C.

Y{N(SiMe$_2$OCHMePh)Dipp}{N(SiHMe$_2$)Dipp}$_2$ (**6**). Y{N(SiHMe$_2$)Dipp} (0.101 g, 0.127 mmol) was dissolved in benzene (3 mL), and acetophenone (14.9 μL, 0.127 mmol) was added to the solution. The reaction mixture was stirred for 15 min., and the solvent was evaporated under reduced pressure. The resulting oily residue was extracted with pentane (3 × 5 mL), concentrated, and cooled to –30 °C to provide the desired product as a white crystalline solid (0.0761 g, 0.0774 mmol, 60.9%). ^{1}H NMR (benzene-*d*$_6$, 600 MHz, 25 °C): δ 7.17 – 6.96 (14 H, aromatic region), 5.33 (br s, 2 H, $^1J_{\mathrm{SiH}}$ = 135.5 Hz, Si*H*Me$_2$), 5.00 (q, 1 H, $^3J_{\mathrm{HH}}$ = 6.5 Hz, OC*H*MePh), 3.79 (br vt, 4 H, $^3J_{\mathrm{HH}}$ = 5.2 Hz, C*H*Me$_2$), 3.66 (v pentet, 2 H, $^3J_{\mathrm{HH}}$ = 6.6 Hz, C*H*Me$_2$), 1.42 (d, 3 H, $^3J_{\mathrm{HH}}$ = 6.4 Hz, OCH*Me*Ph), 1.32 (d, 12 H, $^3J_{\mathrm{HH}}$ = 6.0 Hz, CH*Me*$_2$), 1.26 (br, 12 H, CH*Me*$_2$), 1.18 (d, 6 H, $^3J_{\mathrm{HH}}$ = 6.4 Hz, CH*Me*$_2$), 1.02 (d, 6 H, $^3J_{\mathrm{HH}}$ = 5.2 Hz, CH*Me*$_2$), 0.37 (br s, 12 H, SiH*Me*$_2$), 0.22 (s, 3 H, Si*Me*$_2$), –0.24 (s, 3 H, Si*Me*$_2$). ^{13}C{^{1}H} NMR (benzene-*d*$_6$, 150 MHz, 25 °C): δ 147.99, 145.01, 144.05, 142.89, 141.97, 129.36, 128.68, 127.07, 124.82, 124.28, 122.22, 122.16 (aromatic region), 77.46 (O*C*HMePh), 28.01(*C*HMe$_2$), 27.80 (*C*HMe$_2$), 26.80 (CH*Me*$_2$), 26.57 (OCH*Me*Ph), 26.10

(CH*Me*$_2$), 25.55 (CH*Me*$_2$), 4.08 (Si*Me*$_2$), 2.57 (SiH*Me*$_2$), 2.35 (Si*Me*$_2$). ^{29}Si{^{1}H} NMR (benzene-*d*$_6$, 119.3 MHz, 40 °C): δ –23.4 (*Si*HMe$_2$), 3.94 (*Si*Me$_2$). IR (KBr, cm^{-1}): 3388 w, 3052 m, 2962 s, 2869 s, 2110 w (SiH, from hydrolysis), 1997 w (SiH), 1891 m (SiH), 1588 w, 1459 s, 1426 s, 1382 m, 1361 w, 1309 s, 1245 s, 1189 s, 1148 w, 1109 s, 1041 s, 918 s, 866 s, 836 s, 811 s, 779 s, 702 s, 675 w, 634 w. Anal. Calcd for $C_{55}H_{91}N_3OSi_3Y$ (with pentane): C, 67.17; H, 9.33; N, 4.27. Found: C, 67.07; H, 9.40; N, 4.17. mp 181 - 183 °C.

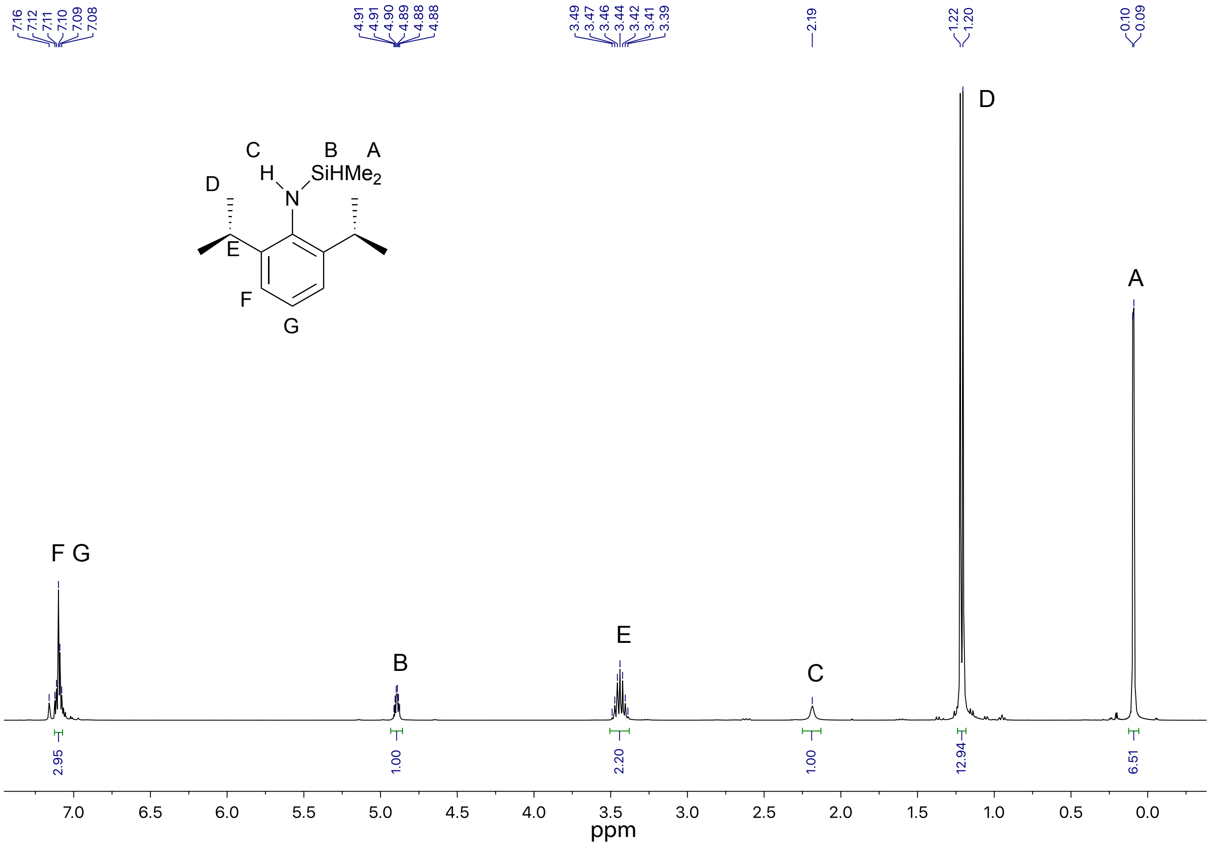

Figure S1. ^{1}H NMR spectrum of HN(SiHMe$_2$)Dipp (**1**) acquired in benzene-*d*$_6$ at room temperature.

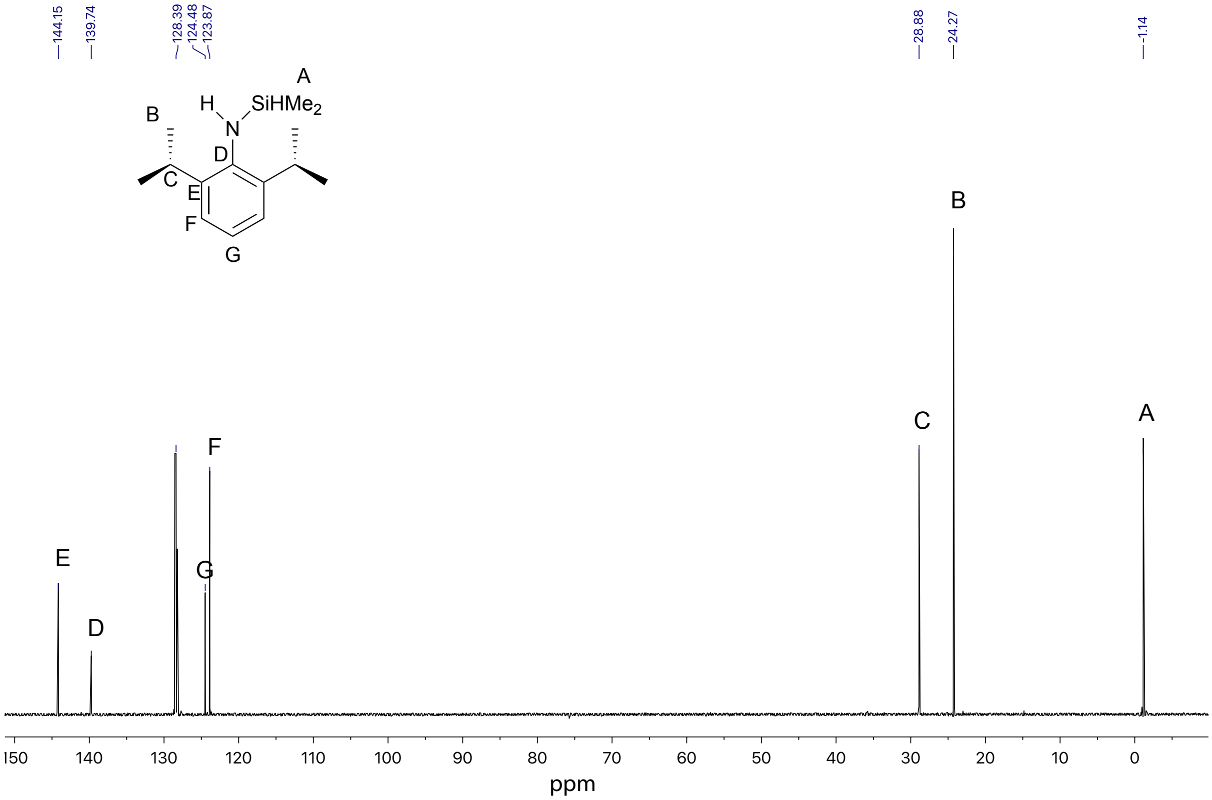

Figure S2. $^{13}C\{^1H\}$ NMR spectrum of HN(SiHMe$_2$)Dipp (**1**) acquired in benzene-d_6 at room temperature.

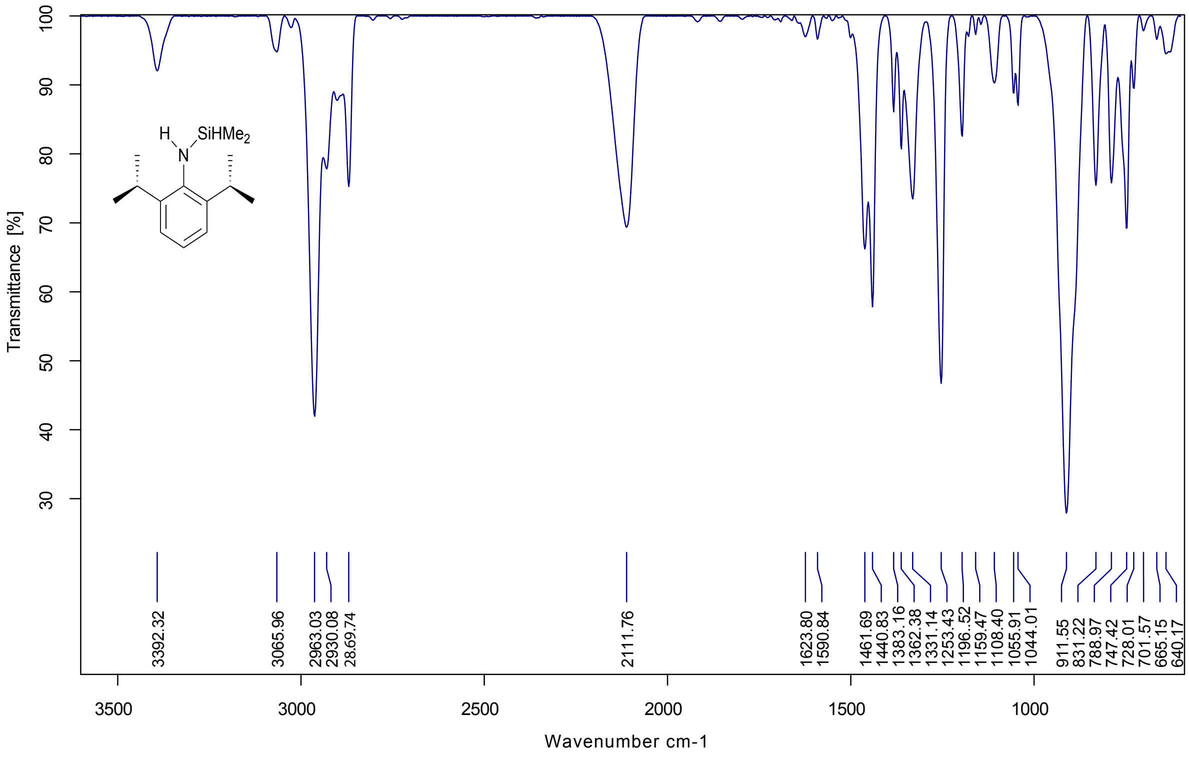

Figure S3. Infrared spectrum of HN(SiHMe$_2$)Dipp (**1**).

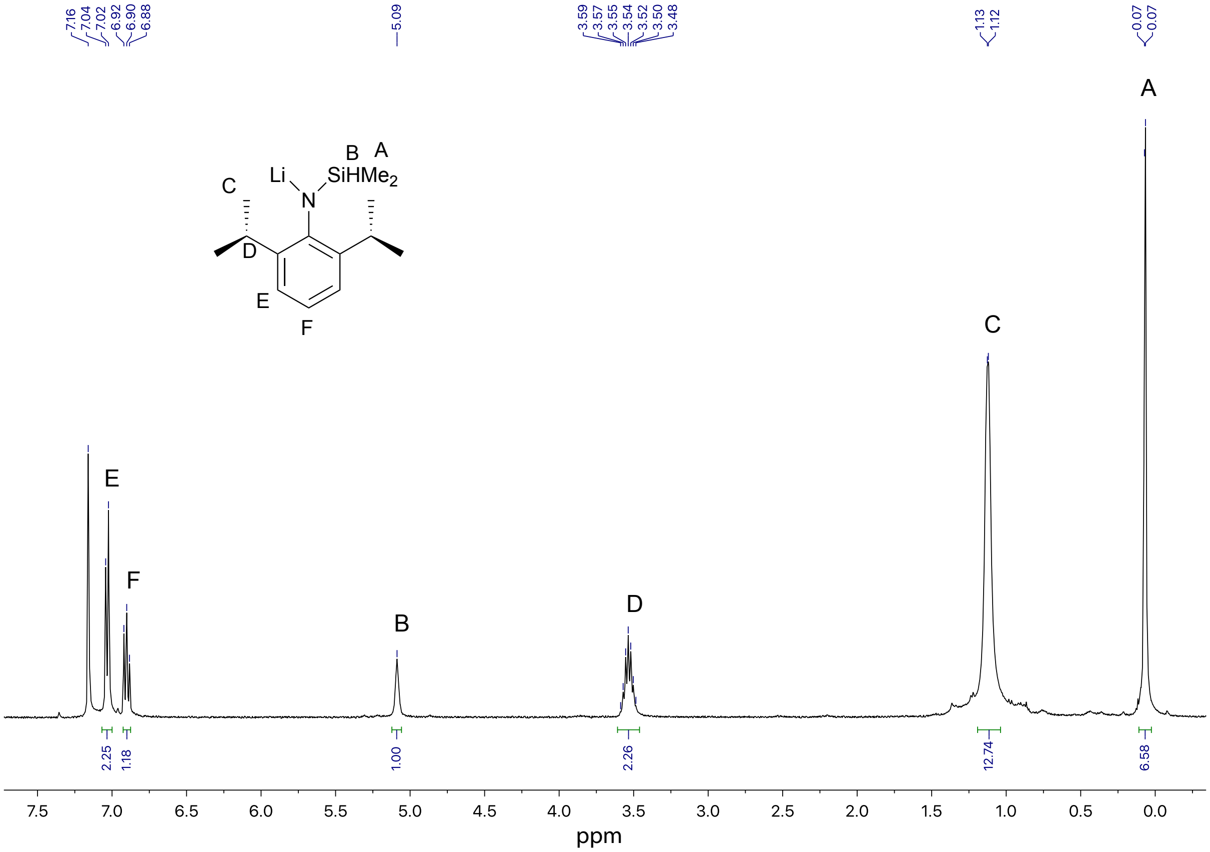

Figure S4. ^{1}H NMR spectrum of LiN(SiHMe$_2$)Dipp (**2**) acquired in benzene-d_6 at room temperature.

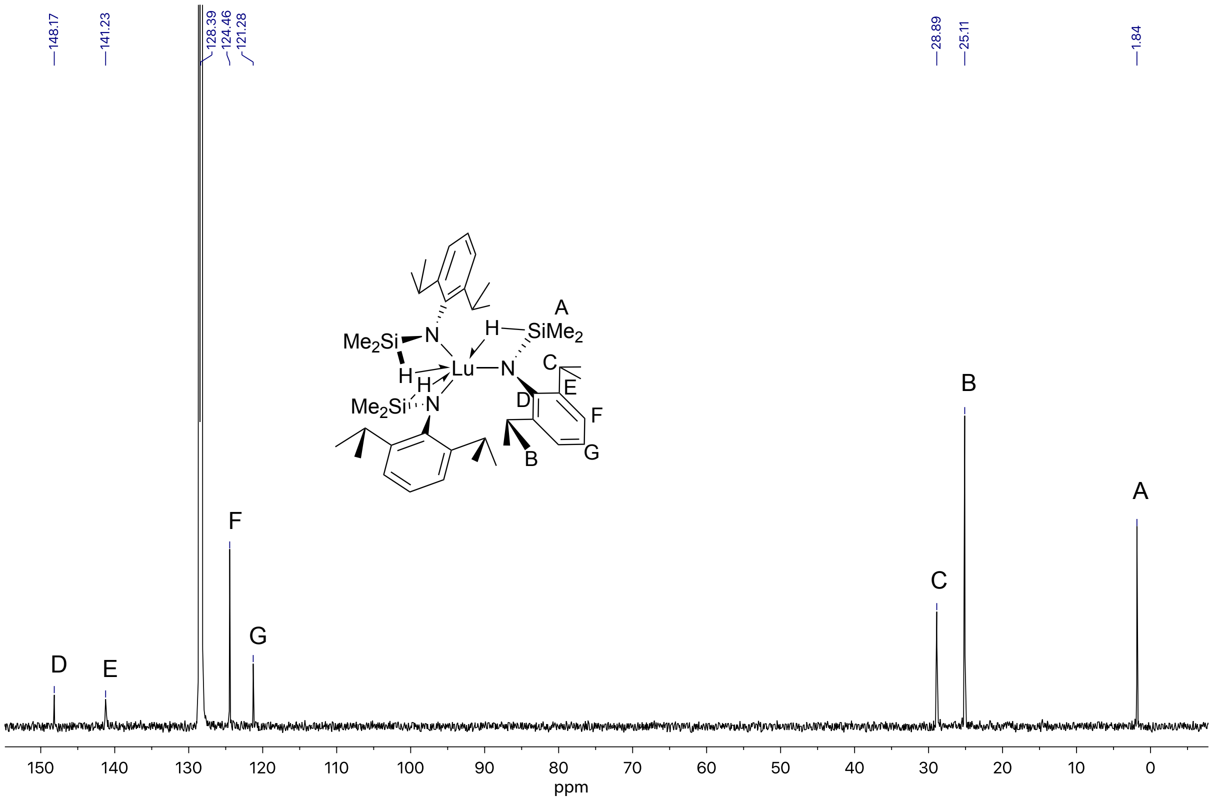

Figure S5. $^{13}C\{^1H\}$ NMR spectrum of LiN(SiHMe$_2$)Dipp (**2**) acquired in benzene-*d*$_6$ at room temperature.

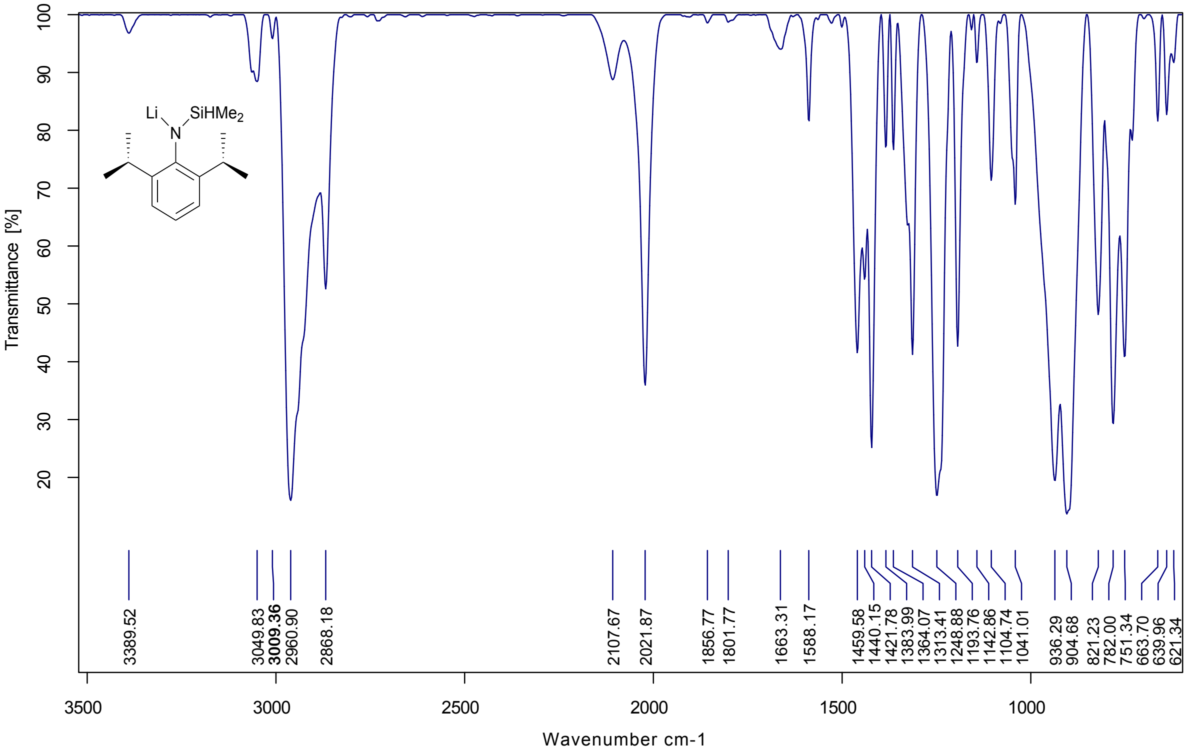

Figure S6. Infrared spectrum (KBr) of LiN(SiHMe$_2$)Dipp (**2**).

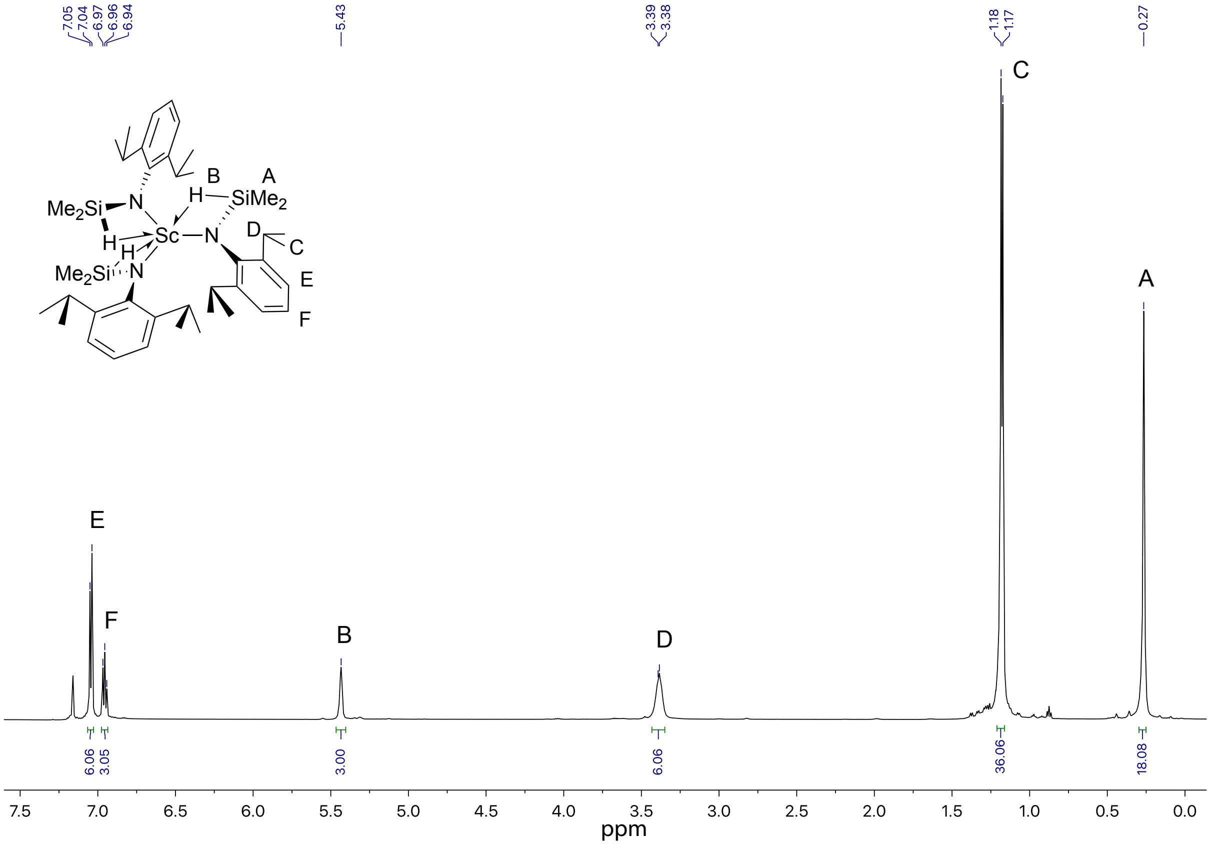

Figure S7. ^{1}H NMR spectrum of Sc{N(SiHMe$_2$)Dipp}$_3$ (**3**) acquired in benzene-d_6 at room temperature.

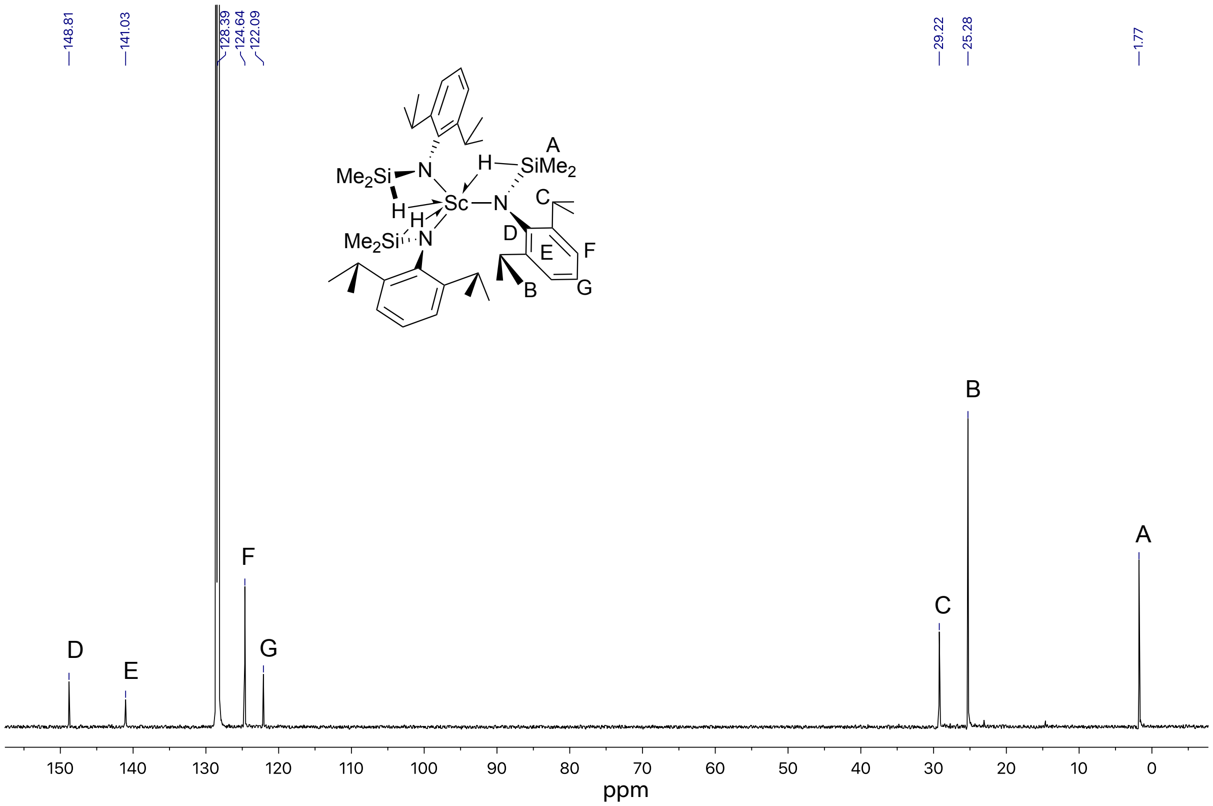

Figure S8. $^{13}C\{^{1}H\}$ NMR spectrum of Sc{N(SiHMe$_2$)Dipp}$_3$ (**3**) acquired in benzene-d_6 at room temperature.

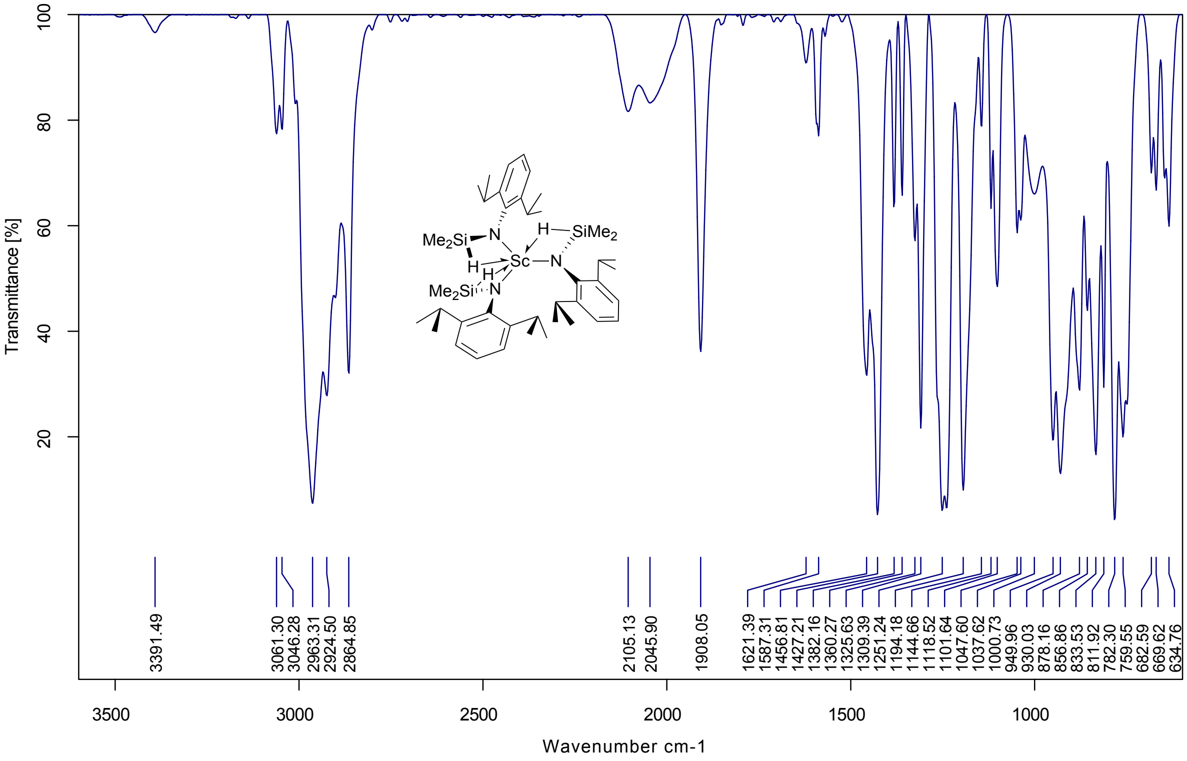

Figure S9. Infrared spectrum (KBr) of Sc{N(SiHMe$_2$)Dipp}$_3$ (**3**).

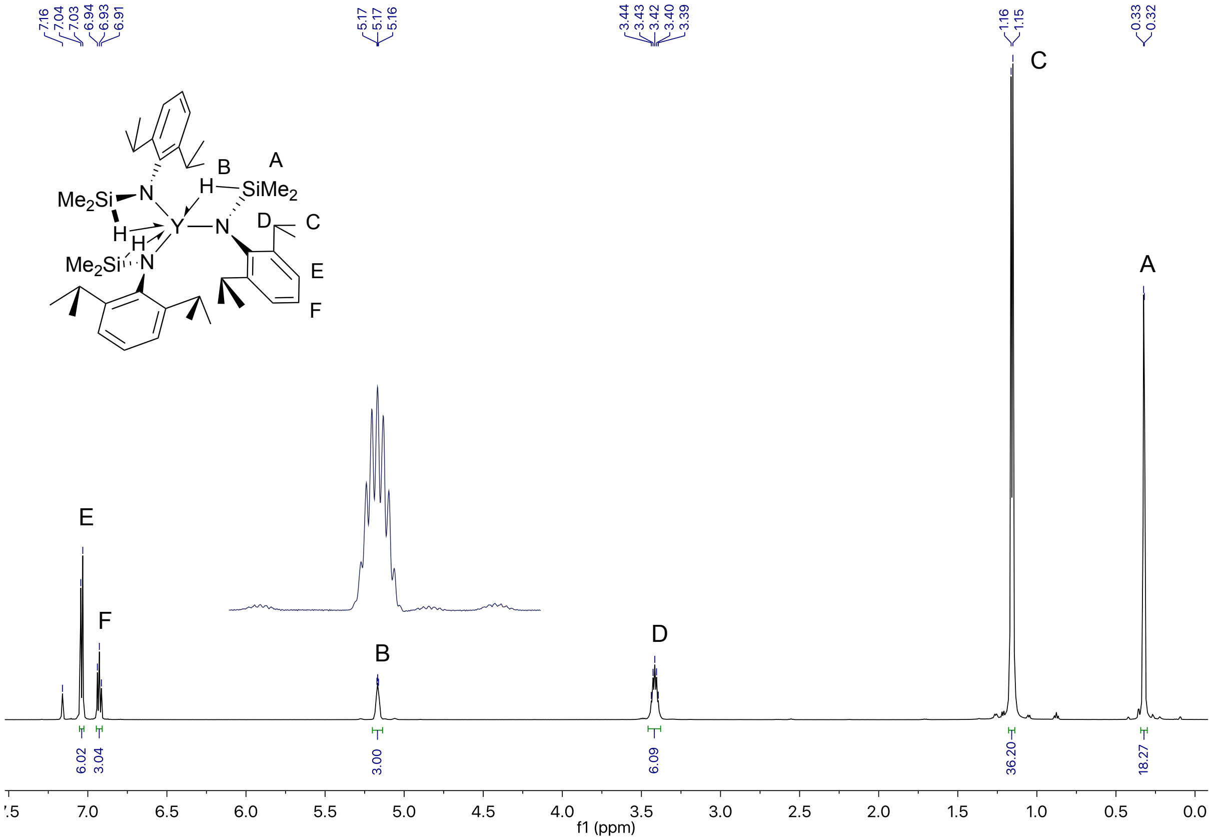

Figure S10. ^{1}H NMR spectrum of Y{N(SiHMe$_2$)Dipp}$_3$ (**4**) acquired in benzene-d_6 at room temperature.

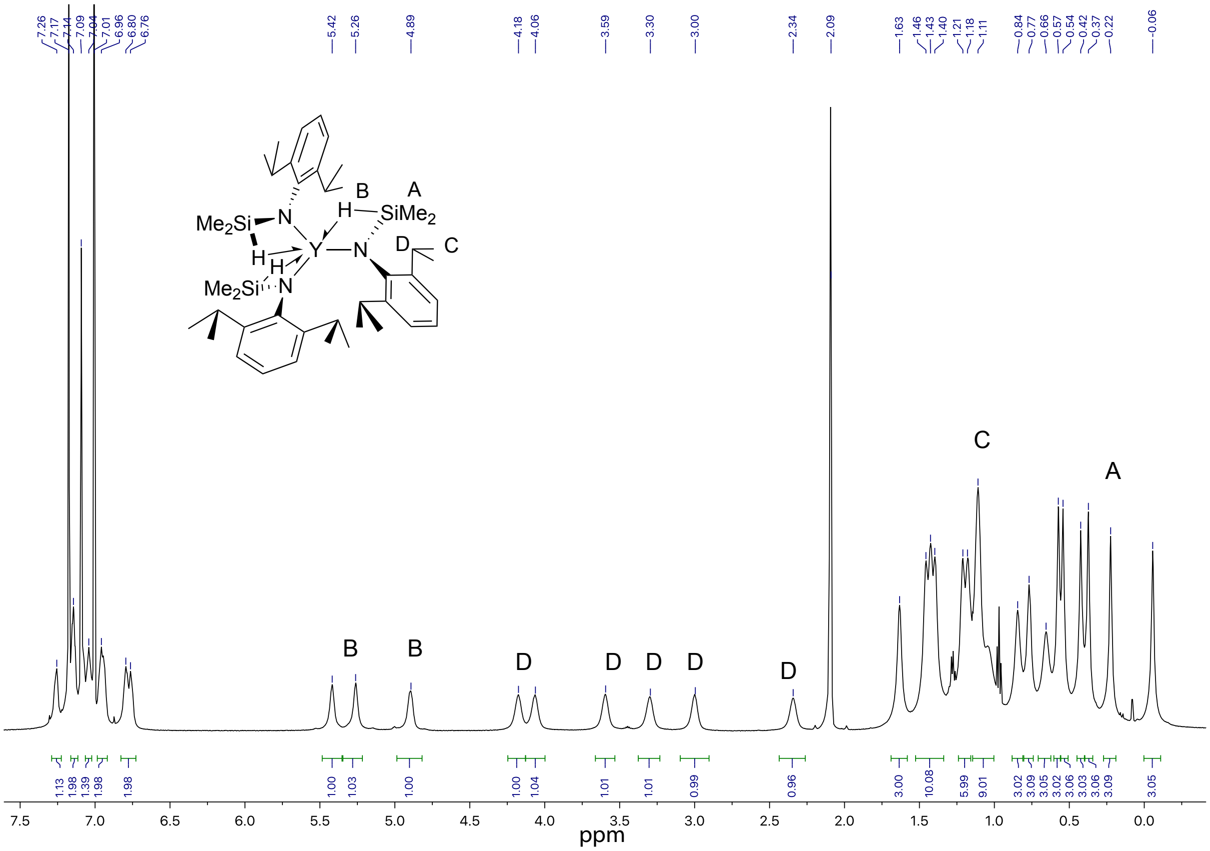

Figure S11. ^{1}H NMR spectrum of Y{N(SiHMe$_2$)Dipp}$_3$ (**4**) acquired in toluene-d_8 at 205 K.

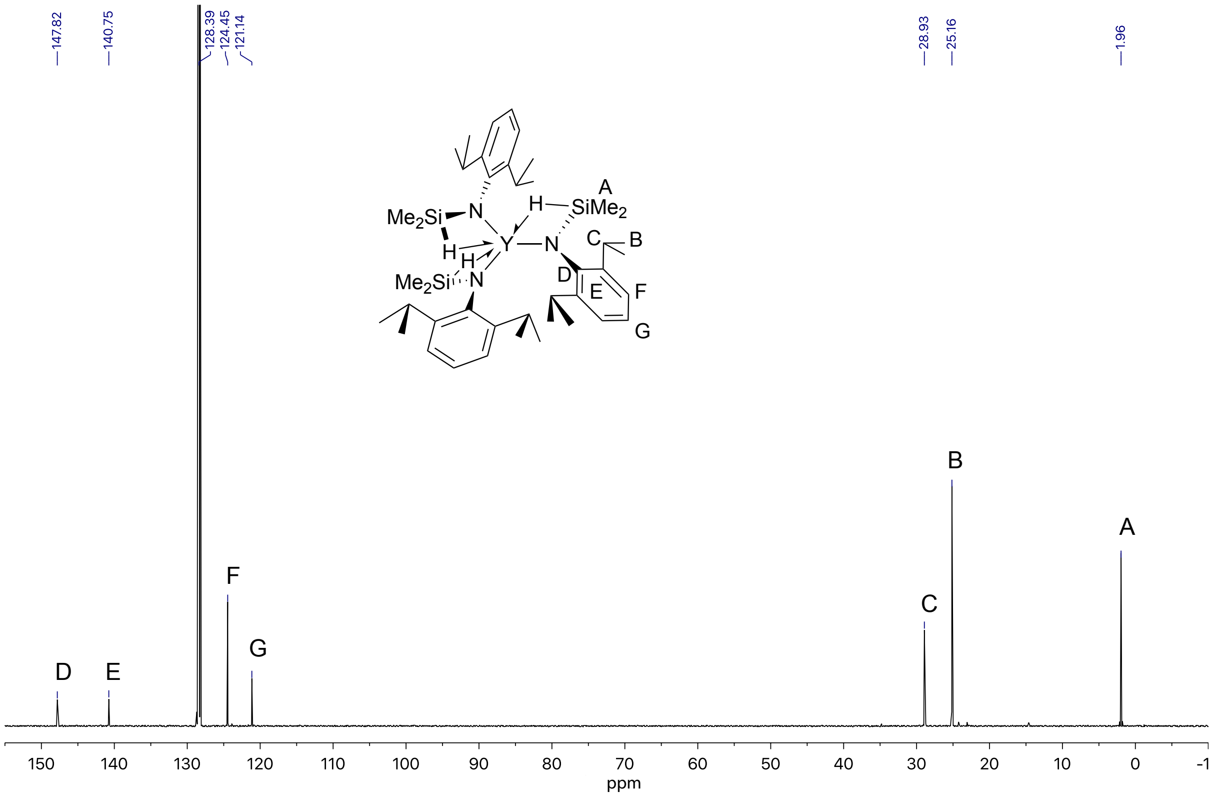

Figure S12. $^{13}C\{^1H\}$ NMR spectrum of Y{N(SiHMe$_2$)Dipp}$_3$ (**4**) acquired in benzene-*d*$_6$ at room temperature.

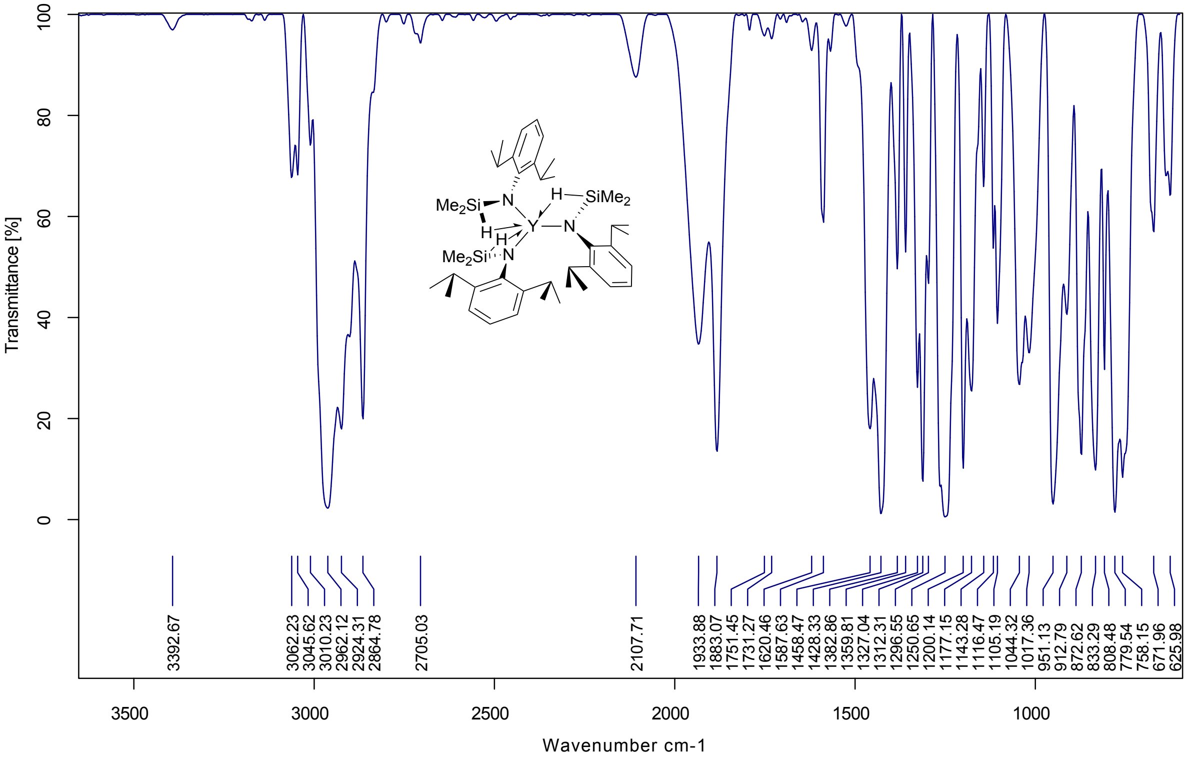

Figure S13. Infrared spectrum (KBr) of Y{N(SiHMe$_2$)Dipp}$_3$ (**4**).

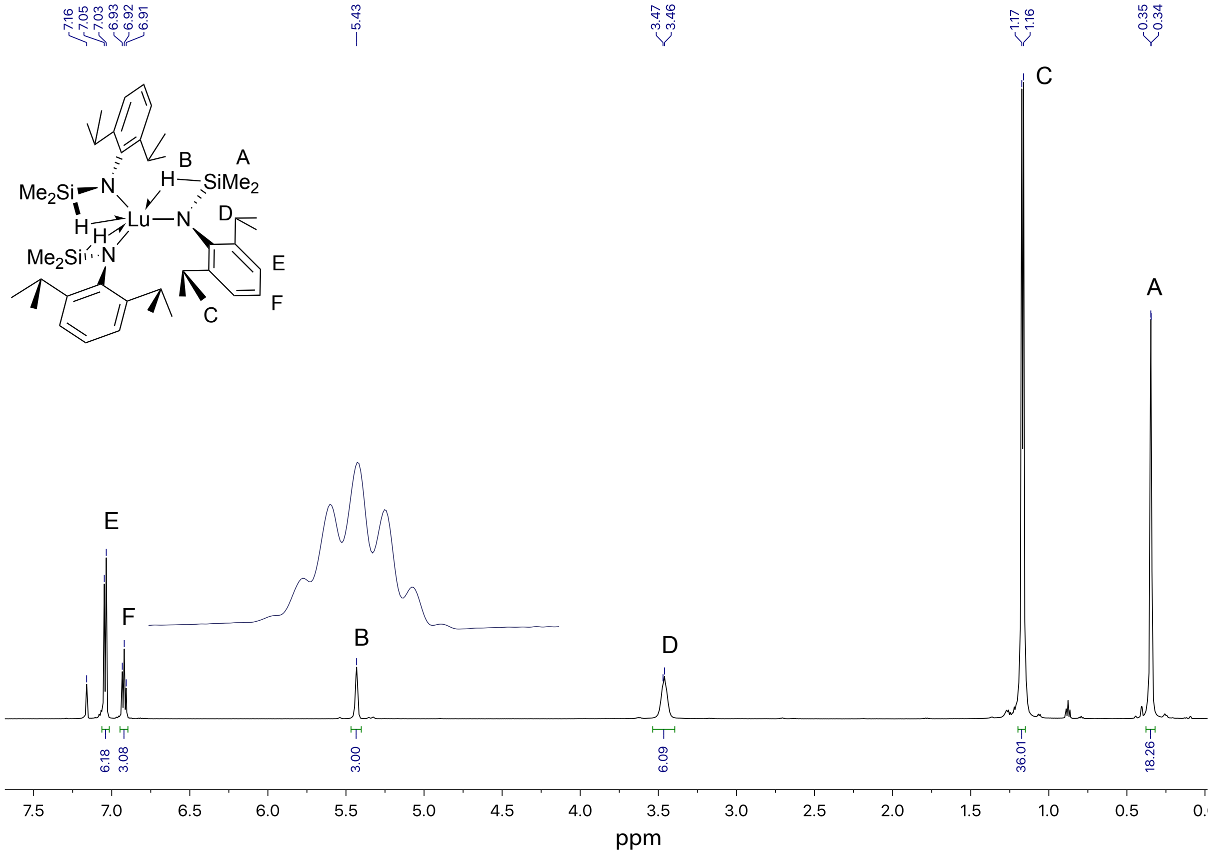

Figure S14. ^{1}H NMR spectrum of Lu{N(SiHMe$_2$)Dipp}$_3$ (**5**) acquired in benzene-*d*$_6$ at room temperature.

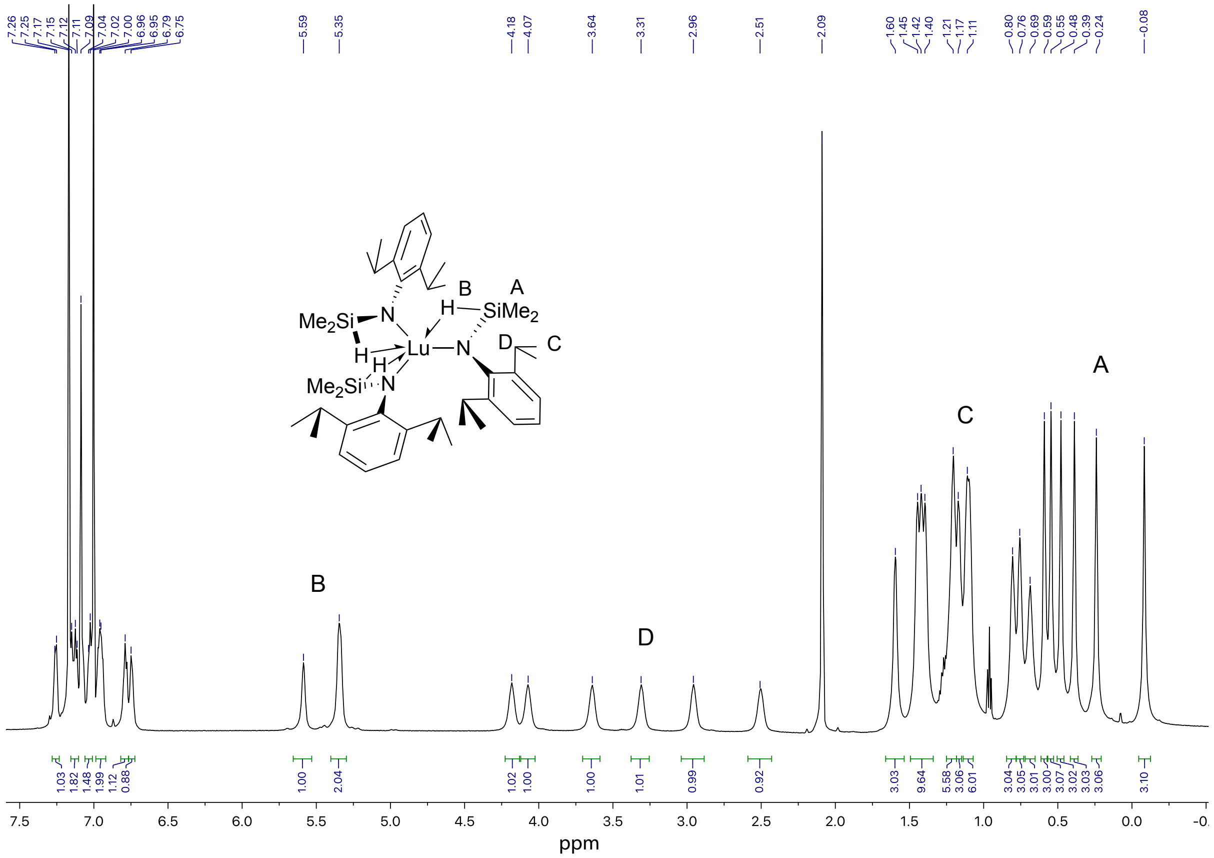

Figure S15. ^{1}H NMR spectrum of Lu{N(SiHMe$_2$)Dipp}$_3$ (**5**) acquired in toluene-d_8 at 205 K.

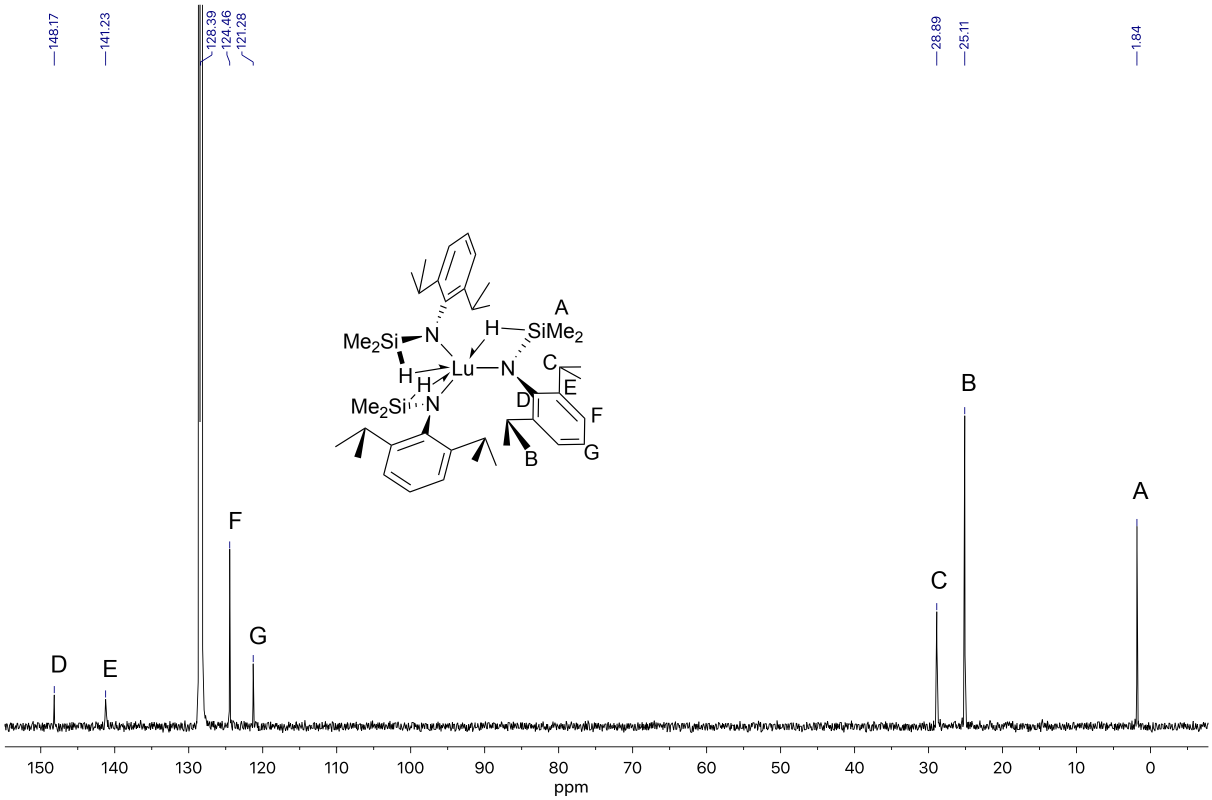

Figure S16. $^{13}C\{^1H\}$ NMR spectrum of Lu{N(SiHMe$_2$)Dipp}$_3$ (**5**) acquired in benzene-*d*$_6$ at room temperature.

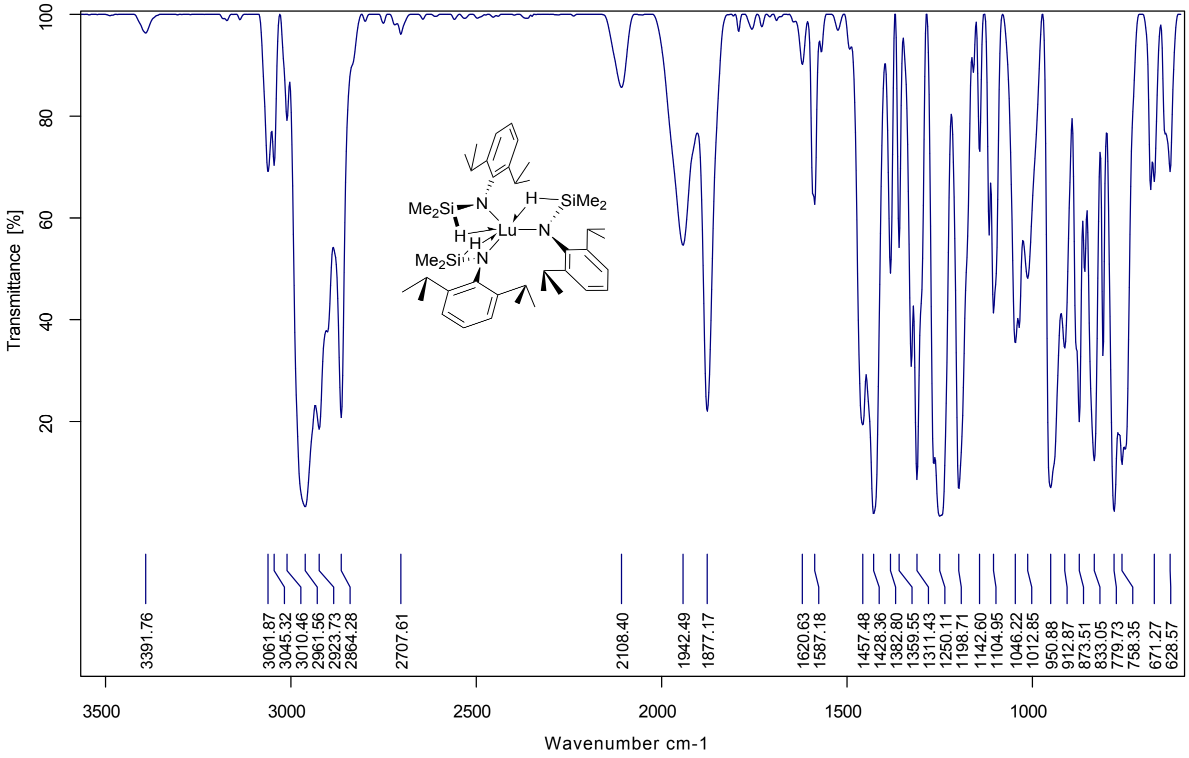

Figure S17. Infrared spectrum (KBr) of Lu{N(SiHMe$_2$)Dipp}$_3$ (**5**).

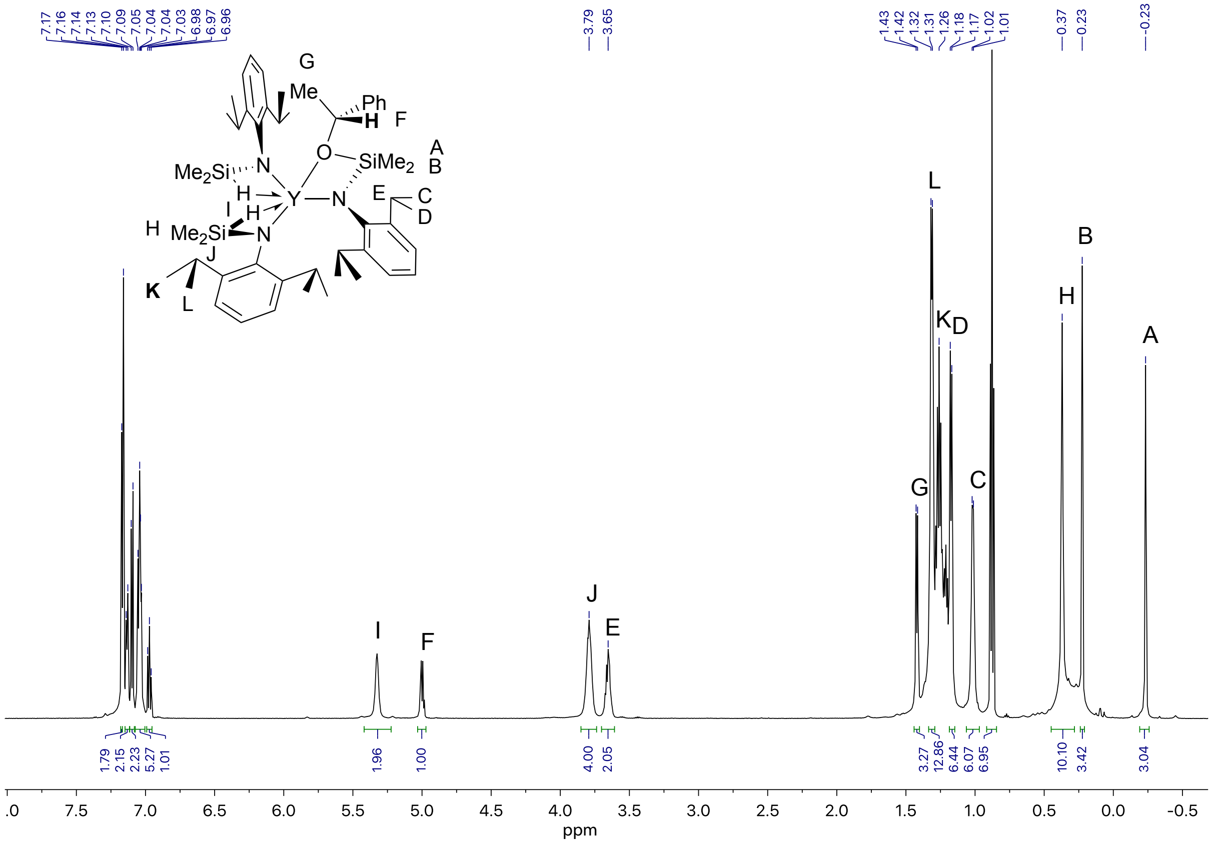

Figure S18. ^{1}H NMR spectrum of Y{N(SiMe$_2$OCHMePh)Dipp}{N(SiHMe$_2$)Dipp}$_2$ (**6**), which co-crystallizes with pentane, acquired in benzene-d_6 at room temperature.

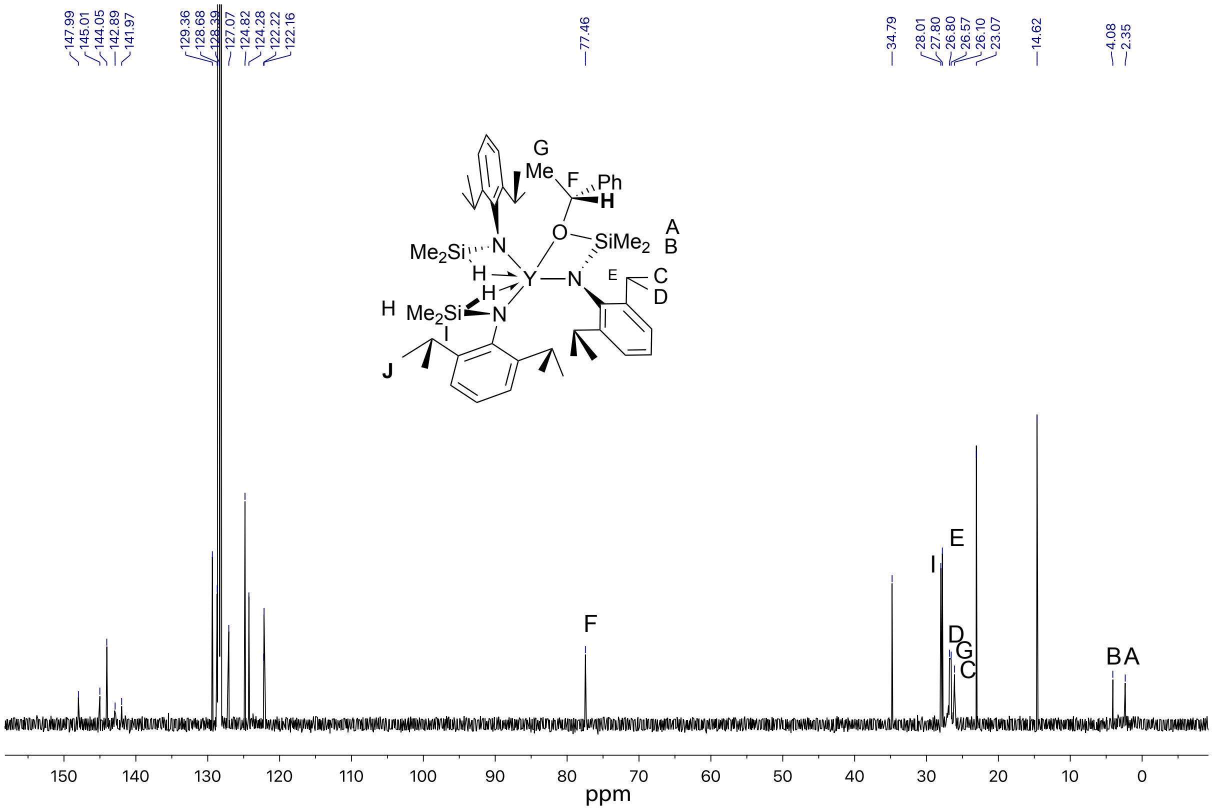

Figure S19. $^{13}C\{^1H\}$ NMR spectrum of Y{N(SiMe$_2$OCHMePh)Dipp}{N(SiHMe$_2$)Dipp}$_2$ (**6**) acquired in benzene-d_6 at room temperature. Signals H (2.57 ppm) and J (25.55 ppm) were obtained from ^1H-^{13}C HMQC, and the expected 12 aromatic ^{13}C signals were observed.

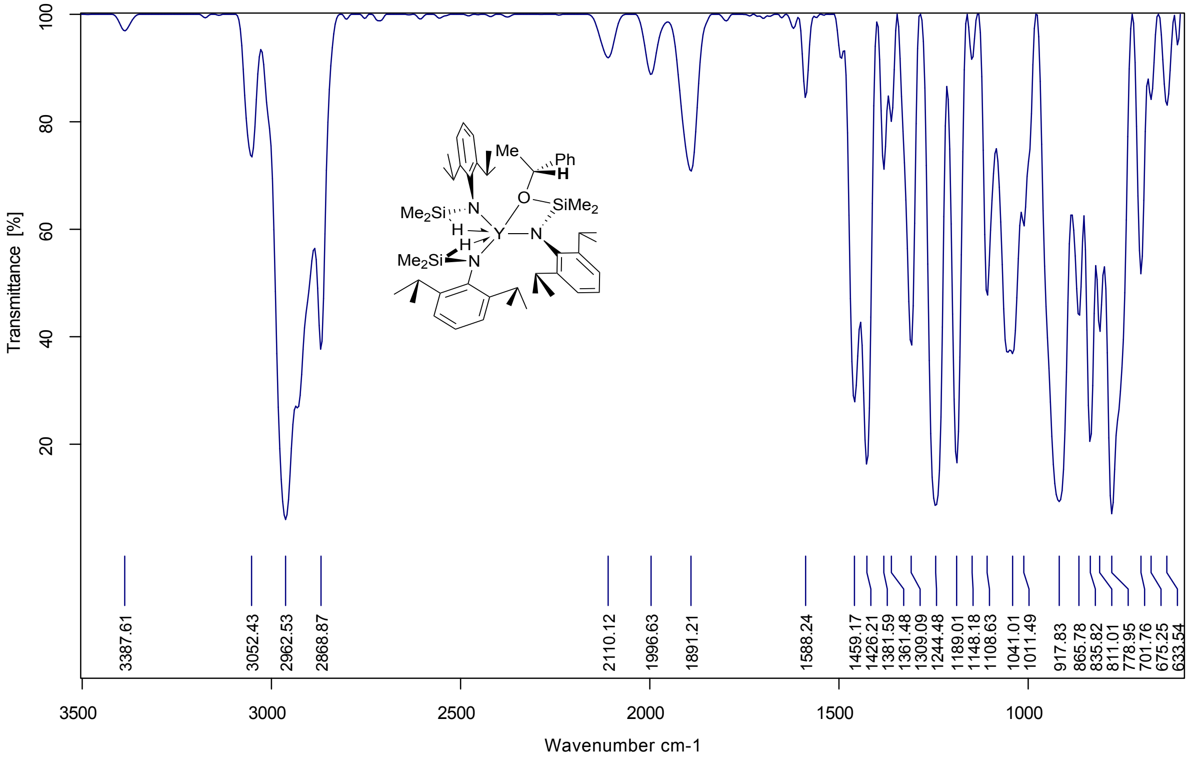

Figure S20. Infrared spectrum (KBr) of Y{N(SiMe$_2$OCHMePh)Dipp}{N(SiHMe$_2$)Dipp}$_2$ (**6**).

Reference

1. W. A. Herrmann, *Synthetic methods of organometallic and inorganic chemistry (Herrmann/Brauer)*, Georg Thieme Verlag ; Thieme Medical Publishers, Stuttgart, 1996.