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Supplementary Information

# Raman Spectroscopy of the N–N Bond in Rare Earth Dinitrogen Complexes

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**Raman Spectroscopy Glassware**

**Table S1** X-ray Data Collection Parameters of four crystals of **1-Lu**.

| | **1-Lu(A)** | **1-Lu(B)** | **1-Lu(C)** | **1-Lu(D)** |
|---|---|---|---|---|
| formula | $C_{40}H_{60}Lu_2N_2$ | $C_{40}H_{60}Lu_2N_2$ | $C_{40}H_{60}Lu_2N_2$ | $C_{40}H_{60}Lu_2N_2$ |
| fw | 918.84 | 918.84 | 918.84 | 918.84 |
| temp (K) | 93(2) | 143(2) | 88(2) | 143(2) |
| Wavelength | 0.71073 Å | 0.71073 Å | 0.71073 Å | 0.71073 Å |
| cryst syst | Triclinic | Tetragonal | Tetragonal | Tetragonal |
| space group | $P\bar{1}$ | $P\bar{4}2_1c$ | $P\bar{4}2_1c$ | $P\bar{4}2_1c$ |
| a (Å) | 10.6530(6) | 14.4028(16) | 14.3199(19) | 14.317(3) |
| b (Å) | 10.9963(6) | 14.4028(16) | 14.3199(19) | 14.317(3) |
| c (Å) | 16.6384(9) | 19.526(2) | 19.597(3) | 19.639(5) |
| Volume (Å$^3$) | 1862.31(18) | 4050.4(10) | 4018.4(12) | 4026(2) |
| α (deg) | 78.1853(6) | 90 | 90 | 90 |
| β (deg) | 78.1059(6) | 90 | 90 | 90 |
| γ (deg) | 83.7548(6) | 90 | 90 | 90 |
| Z | 2 | 4 | 4 | 4 |
| $\rho_{calc}$ (Mg/m$^3$) | 1.639 | 1.507 | 1.519 | 1.516 |
| μ (mm$^{-1}$) | 5.298 | 4.872 | 4.911 | 4.902 |
| R1 (I > 2σ(I))[a] | 0.0232 | 0.0325 | 0.0254 | 0.0274 |
| wR2 (all data)[a] | 0.0572 | 0.0911 | 0.0660 | 0.0758 |

[a]Definitions: $wR2 = [\sum[w(F_o^2 - F_c^2)^2]/ \sum[w(F_o^2)^2] ]^{1/2}$; $R1 = \sum||F_o| - |F_c||/\sum|F_o|$.

**Crystallographic Details**

**X-ray Data Collection, Structure Solution and Refinement for 1-Lu(A).** A red crystal of approximate dimensions 0.115 x 0.122 x 0.345 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[1] program package and the CELL_NOW[2] were used to determine the unit-cell parameters. Data was collected using a 25 sec/frame scan time for a sphere of diffraction data. The raw frame data was processed using SAINT[3] and TWINABS[4] to yield the reflection data file (HKLF5 format)[4]. Subsequent calculations were carried out using the SHELXTL[5] program. There were no systematic absences nor any diffraction symmetry other than the Friedel condition. The centrosymmetric triclinic space group $P\bar{1}$ was assigned and later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The lutetium atoms were disordered and included using multiple components, anisotropic thermal parameters and partial site-occupancy-factors (0.93/0.07). At convergence, wR2 = 0.0572 and Goof = 1.039 for 436 variables refined against 8933 data (0.74Å), R1 = 0.0232 for those 7977 data with $I > 2.0\sigma(I)$. The structure was refined as a two-component twin (HKLF5 format)[5].

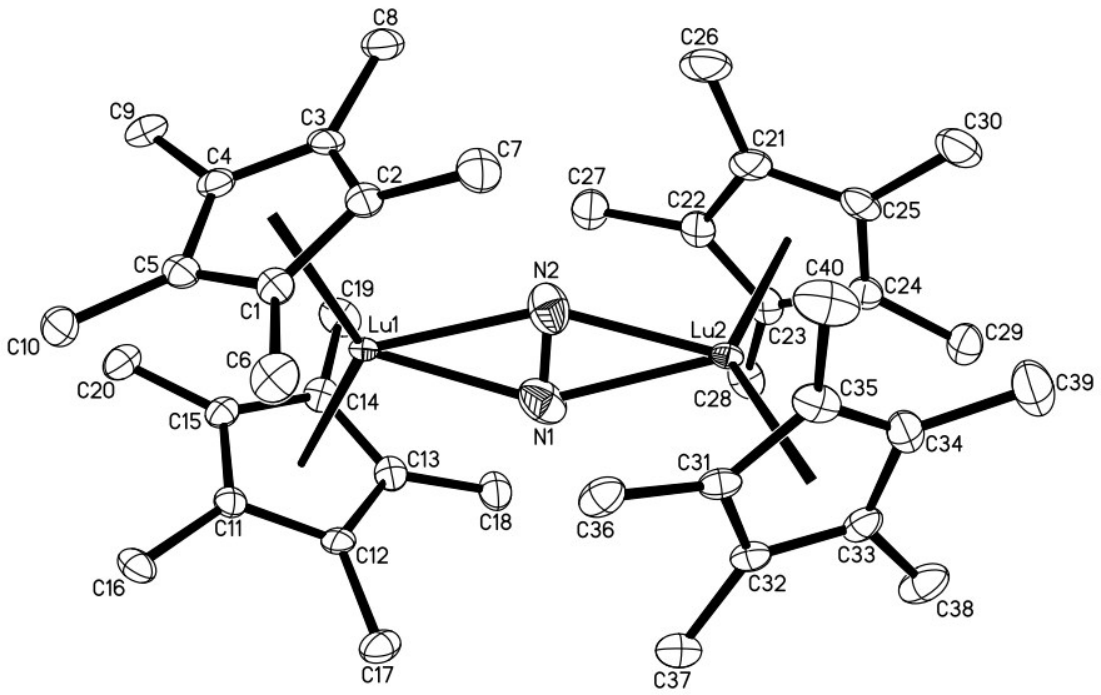


**Fig. S1** Thermal ellipsoid plot of $[(C_5Me_5)_2Lu]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Lu(A)**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S2** Bond lengths [Å] and angles [°] for **1-Lu(A)**.

| | |
|---|---|
| Lu(1)-Cnt1 | 2.335 |
| Lu(1)-Cnt2 | 2.334 |
| Lu(2)-Cnt1 | 2.332 |
| Lu(2)-Cnt1 | 2.320 |
| Lu(1)-N(1) | 2.227(4) |
| Lu(1)-N(2) | 2.265(4) |
| Lu(1)-C(14) | 2.593(3) |
| Lu(1)-C(1) | 2.595(3) |
| Lu(1)-C(13) | 2.604(3) |
| Lu(1)-C(3) | 2.614(3) |
| Lu(1)-C(2) | 2.614(3) |
| Lu(1)-C(12) | 2.618(3) |
| Lu(1)-C(5) | 2.655(3) |
| Lu(1)-C(15) | 2.656(3) |

| | |
|---|---|
| Lu(1)-C(11) | 2.658(3) |
| Lu(1)-C(4) | 2.661(3) |
| Lu(2)-N(2) | 2.205(4) |
| Lu(2)-N(1) | 2.267(4) |
| Lu(2)-C(21) | 2.582(3) |
| Lu(2)-C(32) | 2.590(3) |
| Lu(2)-C(33) | 2.605(3) |
| Lu(2)-C(22) | 2.611(3) |
| Lu(2)-C(31) | 2.618(3) |
| Lu(2)-C(35) | 2.625(3) |
| Lu(2)-C(34) | 2.634(4) |
| Lu(2)-C(25) | 2.635(3) |
| Lu(2)-C(23) | 2.635(3) |
| Lu(2)-C(24) | 2.662(3) |
| N(1)-N(2) | 1.111(5) |
| C(1)-C(2) | 1.414(5) |
| C(1)-C(5) | 1.423(5) |
| C(1)-C(6) | 1.502(5) |
| C(2)-C(3) | 1.410(5) |
| C(2)-C(7) | 1.511(5) |
| C(3)-C(4) | 1.421(6) |
| C(3)-C(8) | 1.506(5) |
| C(4)-C(5) | 1.426(5) |
| C(4)-C(9) | 1.500(5) |
| C(5)-C(10) | 1.499(5) |
| C(11)-C(15) | 1.406(5) |
| C(11)-C(12) | 1.417(5) |
| C(11)-C(16) | 1.503(5) |
| C(12)-C(13) | 1.415(5) |
| C(12)-C(17) | 1.498(5) |
| C(13)-C(14) | 1.416(5) |
| C(13)-C(18) | 1.503(5) |
| C(14)-C(15) | 1.420(5) |
| C(14)-C(19) | 1.503(5) |
| C(15)-C(20) | 1.503(5) |
| C(21)-C(22) | 1.416(6) |

| | |
|---|---|
| C(21)-C(25) | 1.435(6) |
| C(21)-C(26) | 1.488(6) |
| C(22)-C(23) | 1.396(5) |
| C(22)-C(27) | 1.512(6) |
| C(23)-C(24) | 1.419(5) |
| C(23)-C(28) | 1.499(5) |
| C(24)-C(25) | 1.425(5) |
| C(24)-C(29) | 1.504(6) |
| C(25)-C(30) | 1.499(6) |
| C(31)-C(35) | 1.407(5) |
| C(31)-C(32) | 1.423(4) |
| C(31)-C(36) | 1.500(5) |
| C(32)-C(33) | 1.416(5) |
| C(32)-C(37) | 1.492(5) |
| C(33)-C(34) | 1.428(6) |
| C(33)-C(38) | 1.497(5) |
| C(34)-C(35) | 1.412(6) |
| C(34)-C(39) | 1.510(6) |
| C(35)-C(40) | 1.509(5) |

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|---|---|
| Cnt1-Lu(1)-N(1) | 123.8 |
| Cnt1-Lu(1)-N(2) | 101.9 |
| Cnt2-Lu(1)-N(1) | 102.3 |
| Cnt2-Lu(1)-N(2) | 124.1 |
| Cnt1-Lu(1)-Cnt2 | 133.3 |
| Cnt3-Lu(2)-N(1) | 119.4 |
| Cnt3-Lu(2)-N(2) | 102.8 |
| Cnt4-Lu(2)-N(1) | 103.2 |
| Cnt4-Lu(2)-N(2) | 119.1 |
| Cnt3-Lu(2)-Cnt4 | 136.7 |
| N(1)-Lu(1)-N(2) | 28.62(13) |
| N(1)-Lu(1)-C(14) | 102.83(12) |
| N(2)-Lu(1)-C(14) | 112.87(12) |
| N(1)-Lu(1)-C(1) | 111.09(12) |
| N(2)-Lu(1)-C(1) | 100.51(12) |
| C(14)-Lu(1)-C(1) | 145.36(10) |

| | |
|---|---|
| N(1)-Lu(1)-C(13) | 76.75(13) |
| N(2)-Lu(1)-C(13) | 96.55(13) |
| C(14)-Lu(1)-C(13) | 31.62(11) |
| C(1)-Lu(1)-C(13) | 154.75(11) |
| N(1)-Lu(1)-C(3) | 112.87(13) |
| N(2)-Lu(1)-C(3) | 85.25(14) |
| C(14)-Lu(1)-C(3) | 119.93(12) |
| C(1)-Lu(1)-C(3) | 52.07(11) |
| C(13)-Lu(1)-C(3) | 148.80(11) |
| N(1)-Lu(1)-C(2) | 96.41(13) |
| N(2)-Lu(1)-C(2) | 75.79(13) |
| C(14)-Lu(1)-C(2) | 151.10(11) |
| C(1)-Lu(1)-C(2) | 31.51(11) |
| C(13)-Lu(1)-C(2) | 172.28(10) |
| C(3)-Lu(1)-C(2) | 31.30(11) |
| N(1)-Lu(1)-C(12) | 84.17(13) |
| N(2)-Lu(1)-C(12) | 111.59(14) |
| C(14)-Lu(1)-C(12) | 52.19(11) |
| C(1)-Lu(1)-C(12) | 123.58(11) |
| C(13)-Lu(1)-C(12) | 31.45(11) |
| C(3)-Lu(1)-C(12) | 162.95(11) |
| C(2)-Lu(1)-C(12) | 152.78(11) |
| N(1)-Lu(1)-C(5) | 142.50(12) |
| N(2)-Lu(1)-C(5) | 127.42(13) |
| C(14)-Lu(1)-C(5) | 114.40(11) |
| C(1)-Lu(1)-C(5) | 31.41(11) |
| C(13)-Lu(1)-C(5) | 136.03(11) |
| C(3)-Lu(1)-C(5) | 51.60(11) |
| C(2)-Lu(1)-C(5) | 51.65(11) |
| C(12)-Lu(1)-C(5) | 114.88(11) |
| N(1)-Lu(1)-C(15) | 128.20(12) |
| N(2)-Lu(1)-C(15) | 144.12(12) |
| C(14)-Lu(1)-C(15) | 31.37(11) |
| C(1)-Lu(1)-C(15) | 115.19(11) |
| C(13)-Lu(1)-C(15) | 51.51(11) |
| C(3)-Lu(1)-C(15) | 113.29(11) |

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|---|---|
| C(2)-Lu(1)-C(15) | 135.37(11) |
| C(12)-Lu(1)-C(15) | 51.28(10) |
| C(5)-Lu(1)-C(15) | 86.19(10) |
| N(1)-Lu(1)-C(11) | 115.01(13) |
| N(2)-Lu(1)-C(11) | 142.73(13) |
| C(14)-Lu(1)-C(11) | 51.77(10) |
| C(1)-Lu(1)-C(11) | 105.18(11) |
| C(13)-Lu(1)-C(11) | 51.60(11) |
| C(3)-Lu(1)-C(11) | 131.94(11) |
| C(2)-Lu(1)-C(11) | 135.86(11) |
| C(12)-Lu(1)-C(11) | 31.15(11) |
| C(5)-Lu(1)-C(11) | 86.43(10) |
| C(15)-Lu(1)-C(11) | 30.67(10) |
| N(1)-Lu(1)-C(4) | 144.02(13) |
| N(2)-Lu(1)-C(4) | 116.38(14) |
| C(14)-Lu(1)-C(4) | 102.41(11) |
| C(1)-Lu(1)-C(4) | 51.93(11) |
| C(13)-Lu(1)-C(4) | 133.50(11) |
| C(3)-Lu(1)-C(4) | 31.23(12) |
| C(2)-Lu(1)-C(4) | 51.65(11) |
| C(12)-Lu(1)-C(4) | 131.76(11) |
| C(5)-Lu(1)-C(4) | 31.11(10) |
| C(15)-Lu(1)-C(4) | 85.15(11) |
| C(11)-Lu(1)-C(4) | 100.73(10) |
| N(2)-Lu(2)-N(1) | 28.73(14) |
| N(2)-Lu(2)-C(21) | 85.02(15) |
| N(1)-Lu(2)-C(21) | 110.05(14) |
| N(2)-Lu(2)-C(32) | 106.73(14) |
| N(1)-Lu(2)-C(32) | 82.78(13) |
| C(21)-Lu(2)-C(32) | 166.94(13) |
| N(2)-Lu(2)-C(33) | 138.37(13) |
| N(1)-Lu(2)-C(33) | 113.28(14) |
| C(21)-Lu(2)-C(33) | 136.26(13) |
| C(32)-Lu(2)-C(33) | 31.64(12) |
| N(2)-Lu(2)-C(22) | 77.05(13) |
| N(1)-Lu(2)-C(22) | 92.17(13) |

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|---|---|
| C(21)-Lu(2)-C(22) | 31.64(13) |
| C(32)-Lu(2)-C(22) | 155.34(12) |
| C(33)-Lu(2)-C(22) | 139.13(12) |
| N(2)-Lu(2)-C(31) | 91.72(13) |
| N(1)-Lu(2)-C(31) | 79.01(12) |
| C(21)-Lu(2)-C(31) | 145.70(12) |
| C(32)-Lu(2)-C(31) | 31.71(10) |
| C(33)-Lu(2)-C(31) | 52.41(11) |
| C(22)-Lu(2)-C(31) | 168.34(11) |
| N(2)-Lu(2)-C(35) | 109.10(13) |
| N(1)-Lu(2)-C(35) | 106.45(13) |
| C(21)-Lu(2)-C(35) | 119.40(13) |
| C(32)-Lu(2)-C(35) | 51.95(11) |
| C(33)-Lu(2)-C(35) | 52.24(12) |
| C(22)-Lu(2)-C(35) | 150.99(12) |
| C(31)-Lu(2)-C(35) | 31.12(11) |
| N(2)-Lu(2)-C(34) | 139.99(13) |
| N(1)-Lu(2)-C(34) | 129.99(12) |
| C(21)-Lu(2)-C(34) | 115.33(13) |
| C(32)-Lu(2)-C(34) | 51.86(11) |
| C(33)-Lu(2)-C(34) | 31.62(12) |
| C(22)-Lu(2)-C(34) | 137.83(11) |
| C(31)-Lu(2)-C(34) | 51.61(11) |
| C(35)-Lu(2)-C(34) | 31.15(12) |
| N(2)-Lu(2)-C(25) | 116.72(14) |
| N(1)-Lu(2)-C(25) | 141.52(13) |
| C(21)-Lu(2)-C(25) | 31.90(13) |
| C(32)-Lu(2)-C(25) | 135.57(12) |
| C(33)-Lu(2)-C(25) | 104.35(12) |
| C(22)-Lu(2)-C(25) | 52.12(12) |
| C(31)-Lu(2)-C(25) | 133.65(12) |
| C(35)-Lu(2)-C(25) | 102.60(12) |
| C(34)-Lu(2)-C(25) | 87.00(12) |
| N(2)-Lu(2)-C(23) | 102.01(13) |
| N(1)-Lu(2)-C(23) | 106.12(12) |
| C(21)-Lu(2)-C(23) | 51.94(13) |

| | |
|---|---|
| C(32)-Lu(2)-C(23) | 128.25(11) |
| C(33)-Lu(2)-C(23) | 108.69(11) |
| C(22)-Lu(2)-C(23) | 30.87(12) |
| C(31)-Lu(2)-C(23) | 159.63(12) |
| C(35)-Lu(2)-C(23) | 146.99(11) |
| C(34)-Lu(2)-C(23) | 117.75(12) |
| C(25)-Lu(2)-C(23) | 51.86(12) |
| N(2)-Lu(2)-C(24) | 128.35(13) |
| N(1)-Lu(2)-C(24) | 137.19(12) |
| C(21)-Lu(2)-C(24) | 51.91(12) |
| C(32)-Lu(2)-C(24) | 120.12(11) |
| C(33)-Lu(2)-C(24) | 90.61(11) |
| C(22)-Lu(2)-C(24) | 51.29(12) |
| C(31)-Lu(2)-C(24) | 139.82(11) |
| C(35)-Lu(2)-C(24) | 116.14(12) |
| C(34)-Lu(2)-C(24) | 88.99(11) |
| C(25)-Lu(2)-C(24) | 31.21(12) |
| C(23)-Lu(2)-C(24) | 31.08(11) |
| N(2)-N(1)-Lu(1) | 77.6(4) |
| N(2)-N(1)-Lu(2) | 72.5(3) |
| Lu(1)-N(1)-Lu(2) | 150.0(2) |
| N(1)-N(2)-Lu(2) | 78.7(4) |
| N(1)-N(2)-Lu(1) | 73.8(3) |
| Lu(2)-N(2)-Lu(1) | 152.4(2) |
| C(2)-C(1)-C(5) | 108.0(3) |
| C(2)-C(1)-C(6) | 126.6(3) |
| C(5)-C(1)-C(6) | 125.2(3) |
| C(2)-C(1)-Lu(1) | 74.97(18) |
| C(5)-C(1)-Lu(1) | 76.61(18) |
| C(6)-C(1)-Lu(1) | 118.5(2) |
| C(3)-C(2)-C(1) | 108.1(3) |
| C(3)-C(2)-C(7) | 125.4(3) |
| C(1)-C(2)-C(7) | 126.3(3) |
| C(3)-C(2)-Lu(1) | 74.34(17) |
| C(1)-C(2)-Lu(1) | 73.52(18) |
| C(7)-C(2)-Lu(1) | 121.8(2) |

| | |
|---|---|
| C(2)-C(3)-C(4) | 108.6(3) |
| C(2)-C(3)-C(8) | 127.1(4) |
| C(4)-C(3)-C(8) | 123.8(4) |
| C(2)-C(3)-Lu(1) | 74.36(18) |
| C(4)-C(3)-Lu(1) | 76.24(19) |
| C(8)-C(3)-Lu(1) | 122.2(2) |
| C(3)-C(4)-C(5) | 107.4(3) |
| C(3)-C(4)-C(9) | 124.2(3) |
| C(5)-C(4)-C(9) | 127.6(4) |
| C(3)-C(4)-Lu(1) | 72.53(19) |
| C(5)-C(4)-Lu(1) | 74.19(19) |
| C(9)-C(4)-Lu(1) | 126.8(2) |
| C(1)-C(5)-C(4) | 107.9(3) |
| C(1)-C(5)-C(10) | 123.8(3) |
| C(4)-C(5)-C(10) | 127.1(4) |
| C(1)-C(5)-Lu(1) | 71.98(19) |
| C(4)-C(5)-Lu(1) | 74.70(19) |
| C(10)-C(5)-Lu(1) | 128.7(2) |
| C(15)-C(11)-C(12) | 107.9(3) |
| C(15)-C(11)-C(16) | 128.0(3) |
| C(12)-C(11)-C(16) | 123.3(3) |
| C(15)-C(11)-Lu(1) | 74.60(19) |
| C(12)-C(11)-Lu(1) | 72.88(18) |
| C(16)-C(11)-Lu(1) | 126.3(2) |
| C(13)-C(12)-C(11) | 108.0(3) |
| C(13)-C(12)-C(17) | 127.5(3) |
| C(11)-C(12)-C(17) | 124.2(3) |
| C(13)-C(12)-Lu(1) | 73.71(18) |
| C(11)-C(12)-Lu(1) | 75.98(18) |
| C(17)-C(12)-Lu(1) | 121.8(2) |
| C(12)-C(13)-C(14) | 108.1(3) |
| C(12)-C(13)-C(18) | 126.1(3) |
| C(14)-C(13)-C(18) | 125.6(3) |
| C(12)-C(13)-Lu(1) | 74.85(18) |
| C(14)-C(13)-Lu(1) | 73.76(18) |
| C(18)-C(13)-Lu(1) | 121.2(2) |

| | |
|---|---|
| C(13)-C(14)-C(15) | 107.4(3) |
| C(13)-C(14)-C(19) | 127.3(3) |
| C(15)-C(14)-C(19) | 125.0(3) |
| C(13)-C(14)-Lu(1) | 74.62(18) |
| C(15)-C(14)-Lu(1) | 76.79(18) |
| C(19)-C(14)-Lu(1) | 119.8(2) |
| C(11)-C(15)-C(14) | 108.5(3) |
| C(11)-C(15)-C(20) | 126.3(3) |
| C(14)-C(15)-C(20) | 123.8(3) |
| C(11)-C(15)-Lu(1) | 74.73(18) |
| C(14)-C(15)-Lu(1) | 71.84(18) |
| C(20)-C(15)-Lu(1) | 130.1(2) |
| C(22)-C(21)-C(25) | 107.9(4) |
| C(22)-C(21)-C(26) | 127.1(4) |
| C(25)-C(21)-C(26) | 124.9(4) |
| C(22)-C(21)-Lu(2) | 75.30(19) |
| C(25)-C(21)-Lu(2) | 76.07(19) |
| C(26)-C(21)-Lu(2) | 116.9(3) |
| C(23)-C(22)-C(21) | 108.7(3) |
| C(23)-C(22)-C(27) | 126.2(4) |
| C(21)-C(22)-C(27) | 125.1(4) |
| C(23)-C(22)-Lu(2) | 75.52(18) |
| C(21)-C(22)-Lu(2) | 73.06(19) |
| C(27)-C(22)-Lu(2) | 120.2(2) |
| C(22)-C(23)-C(24) | 108.3(3) |
| C(22)-C(23)-C(28) | 127.2(4) |
| C(24)-C(23)-C(28) | 124.1(4) |
| C(22)-C(23)-Lu(2) | 73.61(19) |
| C(24)-C(23)-Lu(2) | 75.52(19) |
| C(28)-C(23)-Lu(2) | 122.4(2) |
| C(23)-C(24)-C(25) | 108.2(3) |
| C(23)-C(24)-C(29) | 124.4(4) |
| C(25)-C(24)-C(29) | 126.0(4) |
| C(23)-C(24)-Lu(2) | 73.40(19) |
| C(25)-C(24)-Lu(2) | 73.3(2) |
| C(29)-C(24)-Lu(2) | 129.7(2) |

| | |
|---|---|
| C(24)-C(25)-C(21) | 106.8(3) |
| C(24)-C(25)-C(30) | 126.4(4) |
| C(21)-C(25)-C(30) | 126.1(4) |
| C(24)-C(25)-Lu(2) | 75.5(2) |
| C(21)-C(25)-Lu(2) | 72.03(19) |
| C(30)-C(25)-Lu(2) | 125.3(3) |
| C(35)-C(31)-C(32) | 107.6(3) |
| C(35)-C(31)-C(36) | 127.5(3) |
| C(32)-C(31)-C(36) | 124.7(3) |
| C(35)-C(31)-Lu(2) | 74.70(18) |
| C(32)-C(31)-Lu(2) | 73.04(17) |
| C(36)-C(31)-Lu(2) | 121.3(2) |
| C(33)-C(32)-C(31) | 108.6(3) |
| C(33)-C(32)-C(37) | 126.8(3) |
| C(31)-C(32)-C(37) | 124.5(3) |
| C(33)-C(32)-Lu(2) | 74.76(19) |
| C(31)-C(32)-Lu(2) | 75.25(18) |
| C(37)-C(32)-Lu(2) | 119.2(2) |
| C(32)-C(33)-C(34) | 106.9(3) |
| C(32)-C(33)-C(38) | 125.1(4) |
| C(34)-C(33)-C(38) | 127.7(4) |
| C(32)-C(33)-Lu(2) | 73.60(19) |
| C(34)-C(33)-Lu(2) | 75.3(2) |
| C(38)-C(33)-Lu(2) | 122.0(2) |
| C(35)-C(34)-C(33) | 108.4(3) |
| C(35)-C(34)-C(39) | 125.3(4) |
| C(33)-C(34)-C(39) | 124.9(4) |
| C(35)-C(34)-Lu(2) | 74.1(2) |
| C(33)-C(34)-Lu(2) | 73.1(2) |
| C(39)-C(34)-Lu(2) | 129.8(3) |
| C(31)-C(35)-C(34) | 108.4(3) |
| C(31)-C(35)-C(40) | 125.6(4) |
| C(34)-C(35)-C(40) | 125.8(4) |
| C(31)-C(35)-Lu(2) | 74.18(19) |
| C(34)-C(35)-Lu(2) | 74.78(19) |
| C(40)-C(35)-Lu(2) | 120.6(3) |

**X-ray Data Collection, Structure Solution and Refinement for 1-Lu(B).** A red crystal of approximate dimensions 0.107 x 0.111 x 0.390 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[7] program package was used to determine the unit-cell parameters and for data collection (25 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[9] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 4/*mmm* and the systematic absences were consistent with the tetragonal space group $P\bar{4}2_1c$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. The lutetium atom was disordered approximately 84% (Lu1), 16% (Lu2). Metric data involving the lutetium atom was referenced to the major component Lu(1). Carbon atoms C(16)-C(20) were disordered and included using multiple components, partial site-occupancy-factors and isotropic thermal parameters. At convergence, wR2 = 0.0911 and Goof = 1.145 for 219 variables refined against 4478 data (0.80Å), R1 = 0.0325 for those 3999 data with $I > 2.0\sigma(I)$. The absolute structure was assigned by refinement of the Flack parameter.[10]

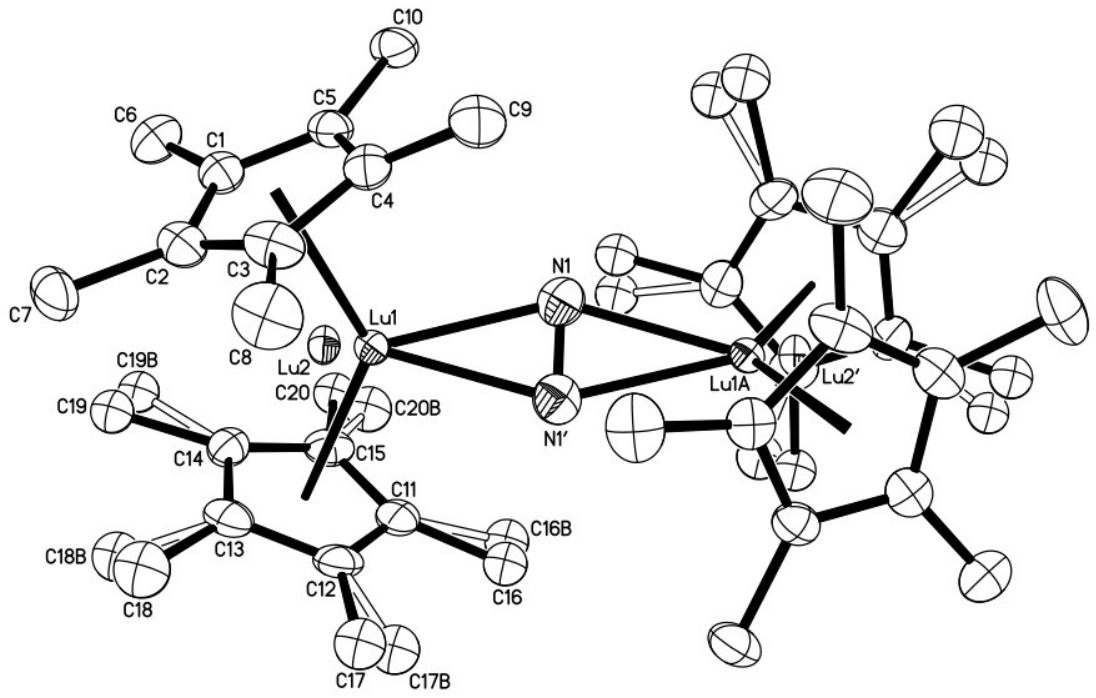


**Fig. S2** Thermal ellipsoid plot of $[(C_5Me_5)_2Lu]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Lu(B)**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S3** Bond lengths [Å] and angles [°] for **1-Lu(B)**.

| | |
|---|---|
| Lu(1)-Cnt1 | 2.352 |
| Lu(1)-Cnt2 | 2.349 |
| Lu(1)-N(1)#1 | 2.271(9) |
| Lu(1)-N(1) | 2.286(9) |
| Lu(1)-C(11) | 2.587(9) |
| Lu(1)-C(12) | 2.593(9) |
| Lu(1)-C(5) | 2.607(9) |
| Lu(1)-C(4) | 2.611(9) |
| Lu(1)-C(3) | 2.612(9) |
| Lu(1)-C(15) | 2.636(9) |
| Lu(1)-C(13) | 2.675(10) |
| Lu(1)-C(14) | 2.684(9) |
| Lu(1)-C(1) | 2.684(10) |

| | |
|---|---|
| Lu(1)-C(2) | 2.693(10) |
| N(1)-N(1)#1 | 1.190(16) |
| N(1)-Lu(1)#1 | 2.271(9) |
| C(1)-C(5) | 1.390(13) |
| C(1)-C(2) | 1.423(13) |
| C(1)-C(6) | 1.501(14) |
| C(2)-C(3) | 1.402(15) |
| C(2)-C(7) | 1.518(14) |
| C(3)-C(4) | 1.414(14) |
| C(3)-C(8) | 1.518(14) |
| C(4)-C(5) | 1.439(14) |
| C(4)-C(9) | 1.466(15) |
| C(5)-C(10) | 1.511(13) |
| C(11)-C(12) | 1.405(16) |
| C(11)-C(15) | 1.420(14) |
| C(11)-C(16) | 1.45(2) |
| C(11)-C(16B) | 1.54(3) |
| C(12)-C(13) | 1.396(16) |
| C(12)-C(17B) | 1.48(3) |
| C(12)-C(17) | 1.61(3) |
| C(13)-C(14) | 1.410(15) |
| C(13)-C(18B) | 1.51(3) |
| C(13)-C(18) | 1.61(3) |
| C(14)-C(15) | 1.383(15) |
| C(14)-C(19) | 1.55(2) |
| C(14)-C(19B) | 1.56(2) |
| C(15)-C(20B) | 1.51(3) |
| C(15)-C(20) | 1.54(3) |

| | |
|---|---|
| Cnt1-Lu(1)-N(1) | 104.9 |
| Cnt1-Lu(1)-N(1)#1 | 123.4 |
| Cnt2-Lu(1)-N(1) | 122.5 |
| Cnt2-Lu(1)-N(1)#1 | 102.7 |
| Cnt1-Lu(1)-Cnt2 | 132.1 |
| N(1)#1-Lu(1)-N(1) | 30.3(4) |
| N(1)#1-Lu(1)-C(11) | 77.0(3) |

| | |
|---|---|
| N(1)-Lu(1)-C(11) | 95.2(3) |
| N(1)#1-Lu(1)-C(12) | 84.9(3) |
| N(1)-Lu(1)-C(12) | 112.6(3) |
| C(11)-Lu(1)-C(12) | 31.5(4) |
| N(1)#1-Lu(1)-C(5) | 113.6(3) |
| N(1)-Lu(1)-C(5) | 86.8(3) |
| C(11)-Lu(1)-C(5) | 154.0(3) |
| C(12)-Lu(1)-C(5) | 160.6(4) |
| N(1)#1-Lu(1)-C(4) | 95.7(3) |
| N(1)-Lu(1)-C(4) | 79.3(3) |
| C(11)-Lu(1)-C(4) | 172.4(3) |
| C(12)-Lu(1)-C(4) | 146.7(3) |
| C(5)-Lu(1)-C(4) | 32.0(3) |
| N(1)#1-Lu(1)-C(3) | 110.3(3) |
| N(1)-Lu(1)-C(3) | 105.4(3) |
| C(11)-Lu(1)-C(3) | 149.2(3) |
| C(12)-Lu(1)-C(3) | 117.9(4) |
| C(5)-Lu(1)-C(3) | 52.0(3) |
| C(4)-Lu(1)-C(3) | 31.4(3) |
| N(1)#1-Lu(1)-C(15) | 103.0(3) |
| N(1)-Lu(1)-C(15) | 109.9(3) |
| C(11)-Lu(1)-C(15) | 31.5(3) |
| C(12)-Lu(1)-C(15) | 51.8(3) |
| C(5)-Lu(1)-C(15) | 124.2(3) |
| C(4)-Lu(1)-C(15) | 155.7(3) |
| C(3)-Lu(1)-C(15) | 144.2(4) |
| N(1)#1-Lu(1)-C(13) | 115.4(3) |
| N(1)-Lu(1)-C(13) | 143.0(3) |
| C(11)-Lu(1)-C(13) | 51.0(3) |
| C(12)-Lu(1)-C(13) | 30.7(3) |
| C(5)-Lu(1)-C(13) | 130.1(4) |
| C(4)-Lu(1)-C(13) | 132.4(4) |
| C(3)-Lu(1)-C(13) | 101.5(4) |
| C(15)-Lu(1)-C(13) | 50.7(4) |
| N(1)#1-Lu(1)-C(14) | 127.7(3) |
| N(1)-Lu(1)-C(14) | 140.1(3) |

| | |
|---|---|
| C(11)-Lu(1)-C(14) | 50.7(3) |
| C(12)-Lu(1)-C(14) | 50.8(3) |
| C(5)-Lu(1)-C(14) | 114.7(3) |
| C(4)-Lu(1)-C(14) | 136.4(3) |
| C(3)-Lu(1)-C(14) | 114.4(4) |
| C(15)-Lu(1)-C(14) | 30.1(3) |
| C(13)-Lu(1)-C(14) | 30.5(3) |
| N(1)#1-Lu(1)-C(1) | 143.8(3) |
| N(1)-Lu(1)-C(1) | 116.9(3) |
| C(11)-Lu(1)-C(1) | 136.3(3) |
| C(12)-Lu(1)-C(1) | 130.4(3) |
| C(5)-Lu(1)-C(1) | 30.4(3) |
| C(4)-Lu(1)-C(1) | 51.3(3) |
| C(3)-Lu(1)-C(1) | 51.0(3) |
| C(15)-Lu(1)-C(1) | 105.9(3) |
| C(13)-Lu(1)-C(1) | 99.8(3) |
| C(14)-Lu(1)-C(1) | 87.2(3) |
| N(1)#1-Lu(1)-C(2) | 140.9(3) |
| N(1)-Lu(1)-C(2) | 130.5(3) |
| C(11)-Lu(1)-C(2) | 134.3(3) |
| C(12)-Lu(1)-C(2) | 111.1(3) |
| C(5)-Lu(1)-C(2) | 51.0(3) |
| C(4)-Lu(1)-C(2) | 51.2(3) |
| C(3)-Lu(1)-C(2) | 30.6(3) |
| C(15)-Lu(1)-C(2) | 115.0(3) |
| C(13)-Lu(1)-C(2) | 84.2(3) |
| C(14)-Lu(1)-C(2) | 86.8(3) |
| C(1)-Lu(1)-C(2) | 30.7(3) |
| N(1)#1-N(1)-Lu(1)#1 | 75.6(8) |
| N(1)#1-N(1)-Lu(1) | 74.2(7) |
| Lu(1)#1-N(1)-Lu(1) | 149.6(4) |
| C(5)-C(1)-C(2) | 108.4(8) |
| C(5)-C(1)-C(6) | 123.4(9) |
| C(2)-C(1)-C(6) | 127.9(9) |
| C(5)-C(1)-Lu(1) | 71.7(5) |
| C(2)-C(1)-Lu(1) | 75.0(5) |

| | |
|---|---|
| C(6)-C(1)-Lu(1) | 124.5(6) |
| C(3)-C(2)-C(1) | 107.6(9) |
| C(3)-C(2)-C(7) | 124.0(9) |
| C(1)-C(2)-C(7) | 127.0(10) |
| C(3)-C(2)-Lu(1) | 71.5(6) |
| C(1)-C(2)-Lu(1) | 74.3(5) |
| C(7)-C(2)-Lu(1) | 130.0(7) |
| C(2)-C(3)-C(4) | 108.9(9) |
| C(2)-C(3)-C(8) | 124.4(10) |
| C(4)-C(3)-C(8) | 126.5(11) |
| C(2)-C(3)-Lu(1) | 77.9(5) |
| C(4)-C(3)-Lu(1) | 74.3(5) |
| C(8)-C(3)-Lu(1) | 118.3(8) |
| C(3)-C(4)-C(5) | 106.6(8) |
| C(3)-C(4)-C(9) | 128.3(10) |
| C(5)-C(4)-C(9) | 124.7(10) |
| C(3)-C(4)-Lu(1) | 74.3(5) |
| C(5)-C(4)-Lu(1) | 73.8(5) |
| C(9)-C(4)-Lu(1) | 122.7(7) |
| C(1)-C(5)-C(4) | 108.3(8) |
| C(1)-C(5)-C(10) | 126.1(10) |
| C(4)-C(5)-C(10) | 125.3(9) |
| C(1)-C(5)-Lu(1) | 77.9(5) |
| C(4)-C(5)-Lu(1) | 74.2(5) |
| C(10)-C(5)-Lu(1) | 119.4(6) |
| C(12)-C(11)-C(15) | 108.0(9) |
| C(12)-C(11)-C(16) | 112.2(14) |
| C(15)-C(11)-C(16) | 139.6(15) |
| C(12)-C(11)-C(16B) | 134.2(13) |
| C(15)-C(11)-C(16B) | 117.4(14) |
| C(12)-C(11)-Lu(1) | 74.5(5) |
| C(15)-C(11)-Lu(1) | 76.1(5) |
| C(16)-C(11)-Lu(1) | 117.9(11) |
| C(16B)-C(11)-Lu(1) | 120.9(10) |
| C(13)-C(12)-C(11) | 108.0(9) |
| C(13)-C(12)-C(17B) | 133.6(15) |

| | |
|---|---|
| C(11)-C(12)-C(17B) | 118.2(14) |
| C(13)-C(12)-C(17) | 113.1(14) |
| C(11)-C(12)-C(17) | 138.7(13) |
| C(13)-C(12)-Lu(1) | 77.9(6) |
| C(11)-C(12)-Lu(1) | 74.0(5) |
| C(17B)-C(12)-Lu(1) | 118.1(11) |
| C(17)-C(12)-Lu(1) | 111.1(11) |
| C(12)-C(13)-C(14) | 107.7(10) |
| C(12)-C(13)-C(18B) | 129.3(14) |
| C(14)-C(13)-C(18B) | 121.8(14) |
| C(12)-C(13)-C(18) | 111.4(14) |
| C(14)-C(13)-C(18) | 141.0(15) |
| C(12)-C(13)-Lu(1) | 71.4(5) |
| C(14)-C(13)-Lu(1) | 75.1(5) |
| C(18B)-C(13)-Lu(1) | 129.0(11) |
| C(18)-C(13)-Lu(1) | 118.0(12) |
| C(15)-C(14)-C(13) | 109.0(9) |
| C(15)-C(14)-C(19) | 136.4(13) |
| C(13)-C(14)-C(19) | 113.1(12) |
| C(15)-C(14)-C(19B) | 115.9(12) |
| C(13)-C(14)-C(19B) | 134.2(12) |
| C(15)-C(14)-Lu(1) | 73.0(5) |
| C(13)-C(14)-Lu(1) | 74.4(5) |
| C(19)-C(14)-Lu(1) | 128.6(10) |
| C(19B)-C(14)-Lu(1) | 126.7(10) |
| C(14)-C(15)-C(11) | 107.3(9) |
| C(14)-C(15)-C(20B) | 134.4(15) |
| C(11)-C(15)-C(20B) | 117.4(15) |
| C(14)-C(15)-C(20) | 115.8(13) |
| C(11)-C(15)-C(20) | 136.1(14) |
| C(14)-C(15)-Lu(1) | 76.9(6) |
| C(11)-C(15)-Lu(1) | 72.3(5) |
| C(20B)-C(15)-Lu(1) | 123.7(11) |
| C(20)-C(15)-Lu(1) | 124.1(10) |

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Symmetry transformations used to generate equivalent atoms:

#1 -x,-y+1,z

**X-ray Data Collection, Structure Solution and Refinement for 1-Lu(C).** A red crystal of approximate dimensions 0.083 x 0.096 x 0.230 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[7] program package was used to determine the unit-cell parameters and for data collection (25 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[9] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 4/*mmm* and the systematic absences were consistent with the tetragonal space group $P\bar{4}2_1c$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. The lutetium atom was disordered approximately 92% (Lu1), 8% (Lu2). Metric data involving the lutetium atom was referenced to the major component Lu(1). Carbon atoms C(16)-C(20) were disordered and included using multiple components, partial site-occupancy-factors and isotropic thermal parameters. At convergence, wR2 = 0.0660 and Goof = 1.082 for 218 variables refined against 5242 data (0.73Å), R1 = 0.0254 for those 4919 data with $I > 2.0\sigma(I)$. The absolute structure was assigned by refinement of the Flack parameter.[10]

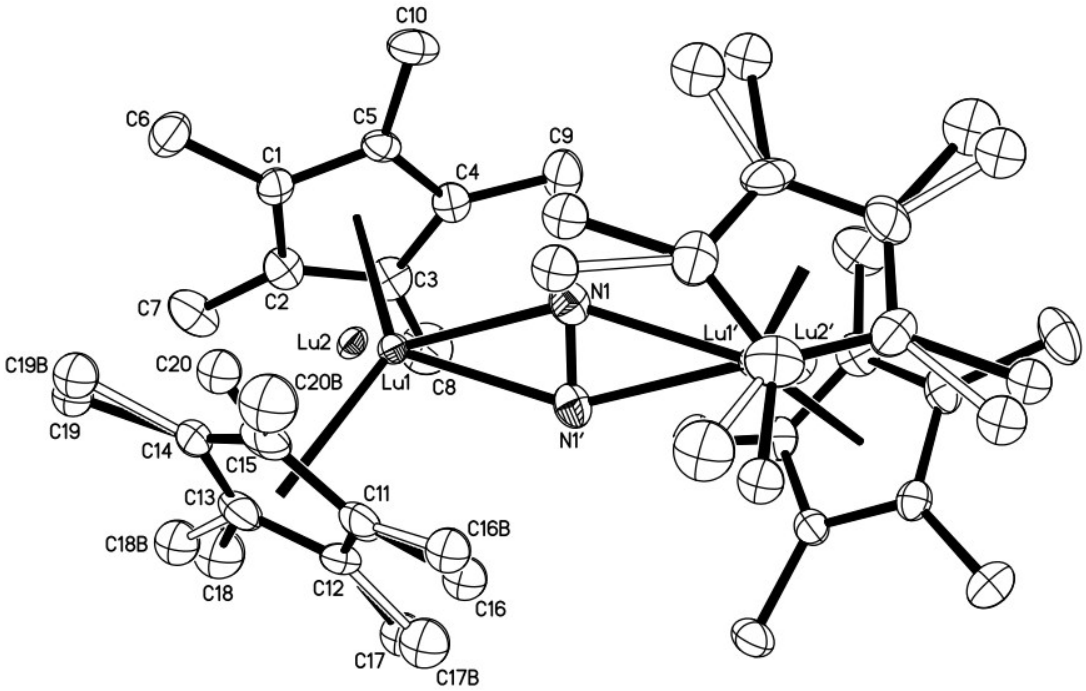


**Fig. S3** Thermal ellipsoid plot of $[(C_5Me_5)_2Lu]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Lu(C)**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S4** Bond lengths [Å] and angles [°] for **1-Lu(C)**.

| | |
|---|---|
| Lu(1)-Cnt1 | 2.336 |
| Lu(1)-Cnt2 | 2.351 |
| Lu(1)-N(1) | 2.267(5) |
| Lu(1)-N(1)#1 | 2.279(5) |
| Lu(1)-C(12) | 2.589(6) |
| Lu(1)-C(3) | 2.604(6) |
| Lu(1)-C(5) | 2.610(5) |
| Lu(1)-C(11) | 2.614(6) |
| Lu(1)-C(4) | 2.615(6) |
| Lu(1)-C(15) | 2.636(7) |
| Lu(1)-C(13) | 2.653(6) |
| Lu(1)-C(1) | 2.654(6) |
| Lu(1)-C(2) | 2.660(6) |

| | |
|---|---|
| Lu(1)-C(14) | 2.679(6) |
| N(1)-N(1)#1 | 1.233(10) |
| N(1)-Lu(1)#1 | 2.279(5) |
| C(1)-C(5) | 1.401(8) |
| C(1)-C(2) | 1.427(9) |
| C(1)-C(6) | 1.500(8) |
| C(2)-C(3) | 1.408(9) |
| C(2)-C(7) | 1.524(9) |
| C(3)-C(4) | 1.426(10) |
| C(3)-C(8) | 1.514(9) |
| C(4)-C(5) | 1.421(9) |
| C(4)-C(9) | 1.479(9) |
| C(5)-C(10) | 1.512(8) |
| C(11)-C(12) | 1.394(11) |
| C(11)-C(15) | 1.441(10) |
| C(11)-C(16) | 1.464(15) |
| C(11)-C(16B) | 1.589(16) |
| C(12)-C(13) | 1.390(11) |
| C(12)-C(17B) | 1.482(18) |
| C(12)-C(17) | 1.614(16) |
| C(13)-C(14) | 1.385(11) |
| C(13)-C(18B) | 1.514(17) |
| C(13)-C(18) | 1.66(2) |
| C(14)-C(15) | 1.380(11) |
| C(14)-C(19) | 1.530(15) |
| C(14)-C(19B) | 1.533(16) |
| C(15)-C(20B) | 1.48(2) |
| C(15)-C(20) | 1.516(16) |

| | |
|---|---|
| Cnt1-Lu(1)-N(1) | 104.2 |
| Cnt1-Lu(1)-N(1)#1 | 123.3 |
| Cnt2-Lu(1)-N(1) | 122.0 |
| Cnt2-Lu(1)-N(1)#1 | 101.8 |
| Cnt1-Lu(1)-Cnt2 | 133.1 |
| N(1)-Lu(1)-N(1)#1 | 31.5(2) |
| N(1)-Lu(1)-C(12) | 112.8(2) |

| | |
|---|---|
| N(1)#1-Lu(1)-C(12) | 84.3(2) |
| N(1)-Lu(1)-C(3) | 104.8(2) |
| N(1)#1-Lu(1)-C(3) | 109.7(2) |
| C(12)-Lu(1)-C(3) | 118.5(2) |
| N(1)-Lu(1)-C(5) | 86.04(18) |
| N(1)#1-Lu(1)-C(5) | 113.83(18) |
| C(12)-Lu(1)-C(5) | 161.1(2) |
| C(3)-Lu(1)-C(5) | 52.2(2) |
| N(1)-Lu(1)-C(11) | 94.9(2) |
| N(1)#1-Lu(1)-C(11) | 76.1(2) |
| C(12)-Lu(1)-C(11) | 31.1(2) |
| C(3)-Lu(1)-C(11) | 149.6(2) |
| C(5)-Lu(1)-C(11) | 154.3(2) |
| N(1)-Lu(1)-C(4) | 78.69(19) |
| N(1)#1-Lu(1)-C(4) | 95.86(19) |
| C(12)-Lu(1)-C(4) | 147.9(2) |
| C(3)-Lu(1)-C(4) | 31.7(2) |
| C(5)-Lu(1)-C(4) | 31.6(2) |
| C(11)-Lu(1)-C(4) | 171.8(2) |
| N(1)-Lu(1)-C(15) | 108.9(2) |
| N(1)#1-Lu(1)-C(15) | 102.2(2) |
| C(12)-Lu(1)-C(15) | 51.8(2) |
| C(3)-Lu(1)-C(15) | 145.8(2) |
| C(5)-Lu(1)-C(15) | 124.2(2) |
| C(11)-Lu(1)-C(15) | 31.9(2) |
| C(4)-Lu(1)-C(15) | 155.5(2) |
| N(1)-Lu(1)-C(13) | 143.0(2) |
| N(1)#1-Lu(1)-C(13) | 114.9(2) |
| C(12)-Lu(1)-C(13) | 30.7(2) |
| C(3)-Lu(1)-C(13) | 103.2(2) |
| C(5)-Lu(1)-C(13) | 130.6(2) |
| C(11)-Lu(1)-C(13) | 50.5(2) |
| C(4)-Lu(1)-C(13) | 134.2(2) |
| C(15)-Lu(1)-C(13) | 50.2(3) |
| N(1)-Lu(1)-C(1) | 116.49(18) |
| N(1)#1-Lu(1)-C(1) | 144.43(19) |

| | |
|---|---|
| C(12)-Lu(1)-C(1) | 130.5(2) |
| C(3)-Lu(1)-C(1) | 51.79(19) |
| C(5)-Lu(1)-C(1) | 30.86(18) |
| C(11)-Lu(1)-C(1) | 136.6(2) |
| C(4)-Lu(1)-C(1) | 51.61(19) |
| C(15)-Lu(1)-C(1) | 106.0(2) |
| C(13)-Lu(1)-C(1) | 99.8(2) |
| N(1)-Lu(1)-C(2) | 130.15(19) |
| N(1)#1-Lu(1)-C(2) | 140.7(2) |
| C(12)-Lu(1)-C(2) | 111.4(2) |
| C(3)-Lu(1)-C(2) | 31.0(2) |
| C(5)-Lu(1)-C(2) | 51.36(18) |
| C(11)-Lu(1)-C(2) | 134.9(2) |
| C(4)-Lu(1)-C(2) | 51.50(19) |
| C(15)-Lu(1)-C(2) | 116.0(2) |
| C(13)-Lu(1)-C(2) | 85.0(2) |
| C(1)-Lu(1)-C(2) | 31.16(19) |
| N(1)-Lu(1)-C(14) | 139.0(2) |
| N(1)#1-Lu(1)-C(14) | 126.9(2) |
| C(12)-Lu(1)-C(14) | 50.9(2) |
| C(3)-Lu(1)-C(14) | 115.9(2) |
| C(5)-Lu(1)-C(14) | 115.0(2) |
| C(11)-Lu(1)-C(14) | 50.8(2) |
| C(4)-Lu(1)-C(14) | 137.2(2) |
| C(15)-Lu(1)-C(14) | 30.1(2) |
| C(13)-Lu(1)-C(14) | 30.1(2) |
| C(1)-Lu(1)-C(14) | 87.19(19) |
| C(2)-Lu(1)-C(14) | 87.8(2) |
| N(1)#1-N(1)-Lu(1) | 74.8(4) |
| N(1)#1-N(1)-Lu(1)#1 | 73.7(4) |
| Lu(1)-N(1)-Lu(1)#1 | 148.5(2) |
| C(5)-C(1)-C(2) | 107.7(5) |
| C(5)-C(1)-C(6) | 124.5(6) |
| C(2)-C(1)-C(6) | 127.0(6) |
| C(5)-C(1)-Lu(1) | 72.9(3) |
| C(2)-C(1)-Lu(1) | 74.7(3) |

| | |
|---|---|
| C(6)-C(1)-Lu(1) | 125.9(4) |
| C(3)-C(2)-C(1) | 108.2(6) |
| C(3)-C(2)-C(7) | 124.2(6) |
| C(1)-C(2)-C(7) | 126.0(6) |
| C(3)-C(2)-Lu(1) | 72.3(3) |
| C(1)-C(2)-Lu(1) | 74.2(3) |
| C(7)-C(2)-Lu(1) | 130.7(4) |
| C(2)-C(3)-C(4) | 107.9(6) |
| C(2)-C(3)-C(8) | 124.6(7) |
| C(4)-C(3)-C(8) | 127.3(7) |
| C(2)-C(3)-Lu(1) | 76.7(3) |
| C(4)-C(3)-Lu(1) | 74.6(3) |
| C(8)-C(3)-Lu(1) | 119.0(5) |
| C(5)-C(4)-C(3) | 107.3(5) |
| C(5)-C(4)-C(9) | 124.9(6) |
| C(3)-C(4)-C(9) | 127.5(7) |
| C(5)-C(4)-Lu(1) | 74.0(3) |
| C(3)-C(4)-Lu(1) | 73.7(3) |
| C(9)-C(4)-Lu(1) | 122.7(4) |
| C(1)-C(5)-C(4) | 108.7(5) |
| C(1)-C(5)-C(10) | 125.6(6) |
| C(4)-C(5)-C(10) | 125.5(6) |
| C(1)-C(5)-Lu(1) | 76.3(3) |
| C(4)-C(5)-Lu(1) | 74.4(3) |
| C(10)-C(5)-Lu(1) | 119.5(4) |
| C(12)-C(11)-C(15) | 107.3(6) |
| C(12)-C(11)-C(16) | 113.3(9) |
| C(15)-C(11)-C(16) | 139.4(9) |
| C(12)-C(11)-C(16B) | 139.5(8) |
| C(15)-C(11)-C(16B) | 112.9(8) |
| C(12)-C(11)-Lu(1) | 73.5(4) |
| C(15)-C(11)-Lu(1) | 74.9(4) |
| C(16)-C(11)-Lu(1) | 118.6(6) |
| C(16B)-C(11)-Lu(1) | 121.9(6) |
| C(13)-C(12)-C(11) | 107.5(6) |
| C(13)-C(12)-C(17B) | 140.5(10) |

| | |
|---|---|
| C(11)-C(12)-C(17B) | 111.5(10) |
| C(13)-C(12)-C(17) | 115.1(9) |
| C(11)-C(12)-C(17) | 137.4(8) |
| C(13)-C(12)-Lu(1) | 77.2(4) |
| C(11)-C(12)-Lu(1) | 75.4(3) |
| C(17B)-C(12)-Lu(1) | 118.2(8) |
| C(17)-C(12)-Lu(1) | 112.5(7) |
| C(14)-C(13)-C(12) | 109.4(7) |
| C(14)-C(13)-C(18B) | 122.0(10) |
| C(12)-C(13)-C(18B) | 127.6(10) |
| C(14)-C(13)-C(18) | 141.3(10) |
| C(12)-C(13)-C(18) | 109.2(10) |
| C(14)-C(13)-Lu(1) | 76.0(4) |
| C(12)-C(13)-Lu(1) | 72.1(4) |
| C(18B)-C(13)-Lu(1) | 127.6(8) |
| C(18)-C(13)-Lu(1) | 115.1(8) |
| C(15)-C(14)-C(13) | 108.4(6) |
| C(15)-C(14)-C(19) | 136.7(9) |
| C(13)-C(14)-C(19) | 113.7(8) |
| C(15)-C(14)-C(19B) | 115.1(8) |
| C(13)-C(14)-C(19B) | 135.3(9) |
| C(15)-C(14)-Lu(1) | 73.2(4) |
| C(13)-C(14)-Lu(1) | 73.9(4) |
| C(19)-C(14)-Lu(1) | 127.5(6) |
| C(19B)-C(14)-Lu(1) | 127.4(7) |
| C(14)-C(15)-C(11) | 107.2(7) |
| C(14)-C(15)-C(20B) | 141.6(11) |
| C(11)-C(15)-C(20B) | 109.5(11) |
| C(14)-C(15)-C(20) | 116.8(8) |
| C(11)-C(15)-C(20) | 134.9(9) |
| C(14)-C(15)-Lu(1) | 76.7(4) |
| C(11)-C(15)-Lu(1) | 73.2(4) |
| C(20B)-C(15)-Lu(1) | 124.7(8) |
| C(20)-C(15)-Lu(1) | 124.7(6) |

---

Symmetry transformations used to generate equivalent atoms:

#1 -x+2,-y+1,z

**X-ray Data Collection, Structure Solution and Refinement for 1-Lu(D).** A red crystal of approximate dimensions 0.109 x 0.129 x 0.229 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[7] program package was used to determine the unit-cell parameters and for data collection (20 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[11] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 4/mmm and the systematic absences were consistent with the tetragonal space group $P\bar{4}2_1c$ which was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. The lutetium atom was disordered approximately 96% (Lu1), 4% (Lu2). Metric data involving the lutetium atom was referenced to the major component Lu(1). Carbon atoms C(16)-C(20) were disordered and included using multiple components with partial site-occupancy-factors and isotropic thermal parameters. At convergence, wR2 = 0.0758 and Goof = 1.096 for 213 variables refined against 4430 data (0.78Å), R1 = 0.0274 for those 4204 data with $I > 2.0\sigma(I)$. The absolute structure was assigned by refinement of the Flack parameter[10]. Solvent accessible voids were examined with the PLATON program SQUEEZE[12]. A model based on the application of the SQUEEZE routine was not deemed appropriate as no solvent molecules were observed.

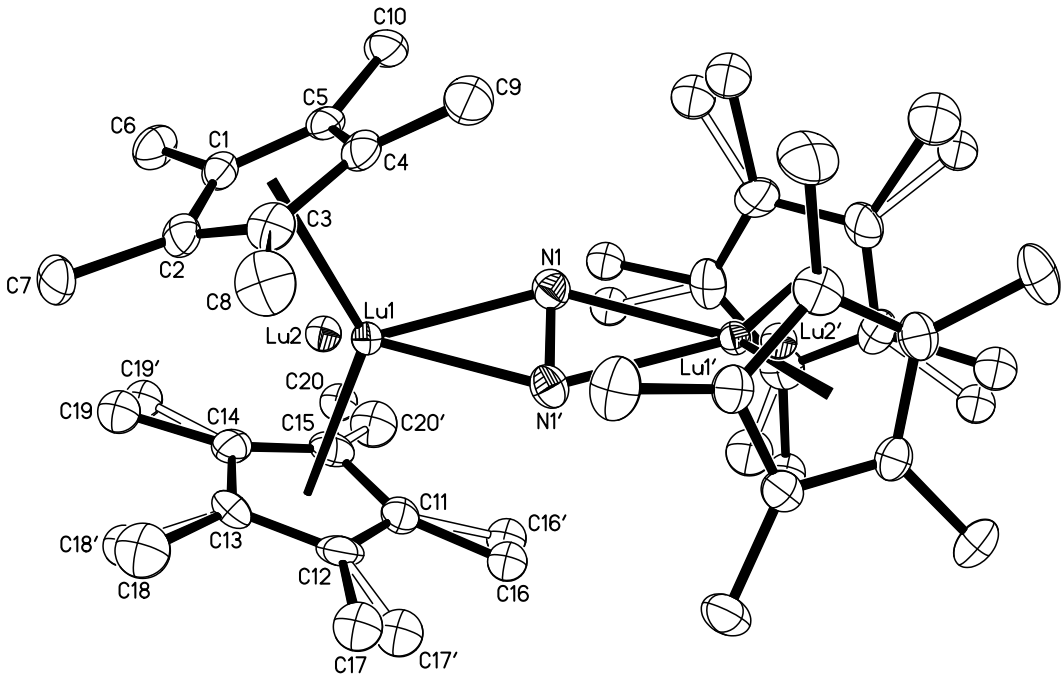


**Fig. S4** Thermal ellipsoid plot of $[(C_5Me_5)_2Lu]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Lu(D)**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S5** Bond lengths [Å] and angles [°] for **1-Lu(D)**.

| | |
|---|---|
| Lu(1)-Cnt1 | 2.330 |
| Lu(1)-Cnt1 | 2.338 |
| Lu(1)-N(1)#1 | 2.272(6) |
| Lu(1)-N(1) | 2.279(6) |
| Lu(1)-C(12) | 2.579(8) |
| Lu(1)-C(3) | 2.593(7) |
| Lu(1)-C(5) | 2.605(7) |
| Lu(1)-C(4) | 2.612(7) |
| Lu(1)-C(11) | 2.613(8) |
| Lu(1)-C(15) | 2.637(9) |
| Lu(1)-C(13) | 2.642(8) |

| | |
|---|---|
| Lu(1)-C(1) | 2.649(8) |
| Lu(1)-C(2) | 2.655(7) |
| Lu(1)-C(14) | 2.658(8) |
| N(1)-N(1)#1 | 1.239(12) |
| N(1)-Lu(1)#1 | 2.272(6) |
| C(1)-C(5) | 1.391(11) |
| C(1)-C(2) | 1.430(11) |
| C(1)-C(6) | 1.508(10) |
| C(2)-C(3) | 1.404(11) |
| C(2)-C(7) | 1.505(11) |
| C(3)-C(4) | 1.432(12) |
| C(3)-C(8) | 1.523(12) |
| C(4)-C(5) | 1.423(11) |
| C(4)-C(9) | 1.482(12) |
| C(5)-C(10) | 1.515(11) |
| C(11)-C(12) | 1.397(13) |
| C(11)-C(15) | 1.443(13) |
| C(11)-C(16) | 1.46(2) |
| C(11)-C(16A) | 1.55(2) |
| C(12)-C(13) | 1.399(13) |
| C(12)-C(17A) | 1.50(2) |
| C(12)-C(17) | 1.61(2) |
| C(13)-C(14) | 1.395(14) |
| C(13)-C(18A) | 1.492(19) |
| C(13)-C(18) | 1.68(2) |
| C(14)-C(15) | 1.391(13) |
| C(14)-C(19) | 1.54(2) |
| C(14)-C(19A) | 1.56(2) |
| C(15)-C(20A) | 1.44(3) |
| C(15)-C(20) | 1.53(2) |

| | |
|---|---|
| Cnt1-Lu(1)-N(1) | 104.2 |
| Cnt1#1-Lu(1)-N(1) | 123.3 |
| Cnt2-Lu(1)-N(1) | 121.6 |
| Cnt2#1-Lu(1)-N(1) | 101.3 |
| Cnt1-Lu(1)-Cnt2 | 133.6 |

| | |
|---|---|
| N(1)#1-Lu(1)-N(1) | 31.6(3) |
| N(1)#1-Lu(1)-C(12) | 83.6(3) |
| N(1)-Lu(1)-C(12) | 112.2(3) |
| N(1)#1-Lu(1)-C(3) | 109.8(3) |
| N(1)-Lu(1)-C(3) | 104.9(3) |
| C(12)-Lu(1)-C(3) | 118.8(3) |
| N(1)#1-Lu(1)-C(5) | 113.8(2) |
| N(1)-Lu(1)-C(5) | 85.9(2) |
| C(12)-Lu(1)-C(5) | 161.9(3) |
| C(3)-Lu(1)-C(5) | 52.4(3) |
| N(1)#1-Lu(1)-C(4) | 95.9(2) |
| N(1)-Lu(1)-C(4) | 78.6(2) |
| C(12)-Lu(1)-C(4) | 148.1(3) |
| C(3)-Lu(1)-C(4) | 31.9(3) |
| C(5)-Lu(1)-C(4) | 31.7(3) |
| N(1)#1-Lu(1)-C(11) | 75.6(2) |
| N(1)-Lu(1)-C(11) | 94.4(3) |
| C(12)-Lu(1)-C(11) | 31.2(3) |
| C(3)-Lu(1)-C(11) | 149.9(3) |
| C(5)-Lu(1)-C(11) | 154.0(3) |
| C(4)-Lu(1)-C(11) | 171.3(3) |
| N(1)#1-Lu(1)-C(15) | 101.9(3) |
| N(1)-Lu(1)-C(15) | 108.6(3) |
| C(12)-Lu(1)-C(15) | 52.0(3) |
| C(3)-Lu(1)-C(15) | 145.9(3) |
| C(5)-Lu(1)-C(15) | 124.2(3) |
| C(4)-Lu(1)-C(15) | 155.6(3) |
| C(11)-Lu(1)-C(15) | 31.9(3) |
| N(1)#1-Lu(1)-C(13) | 114.5(3) |
| N(1)-Lu(1)-C(13) | 142.8(3) |
| C(12)-Lu(1)-C(13) | 31.1(3) |
| C(3)-Lu(1)-C(13) | 102.9(3) |
| C(5)-Lu(1)-C(13) | 131.0(3) |
| C(4)-Lu(1)-C(13) | 134.1(3) |
| C(11)-Lu(1)-C(13) | 51.1(3) |
| C(15)-Lu(1)-C(13) | 50.9(3) |

| | |
|---|---|
| N(1)#1-Lu(1)-C(1) | 144.2(2) |
| N(1)-Lu(1)-C(1) | 116.2(2) |
| C(12)-Lu(1)-C(1) | 131.4(3) |
| C(3)-Lu(1)-C(1) | 51.5(3) |
| C(5)-Lu(1)-C(1) | 30.7(2) |
| C(4)-Lu(1)-C(1) | 51.4(2) |
| C(11)-Lu(1)-C(1) | 137.3(3) |
| C(15)-Lu(1)-C(1) | 106.4(3) |
| C(13)-Lu(1)-C(1) | 100.4(3) |
| N(1)#1-Lu(1)-C(2) | 140.8(2) |
| N(1)-Lu(1)-C(2) | 130.3(2) |
| C(12)-Lu(1)-C(2) | 111.8(3) |
| C(3)-Lu(1)-C(2) | 31.0(2) |
| C(5)-Lu(1)-C(2) | 51.8(2) |
| C(4)-Lu(1)-C(2) | 51.8(2) |
| C(11)-Lu(1)-C(2) | 135.2(2) |
| C(15)-Lu(1)-C(2) | 116.1(3) |
| C(13)-Lu(1)-C(2) | 84.8(3) |
| C(1)-Lu(1)-C(2) | 31.3(2) |
| N(1)#1-Lu(1)-C(14) | 126.8(3) |
| N(1)-Lu(1)-C(14) | 139.1(3) |
| C(12)-Lu(1)-C(14) | 51.3(3) |
| C(3)-Lu(1)-C(14) | 115.7(3) |
| C(5)-Lu(1)-C(14) | 115.2(3) |
| C(4)-Lu(1)-C(14) | 137.3(3) |
| C(11)-Lu(1)-C(14) | 51.2(3) |
| C(15)-Lu(1)-C(14) | 30.5(3) |
| C(13)-Lu(1)-C(14) | 30.5(3) |
| C(1)-Lu(1)-C(14) | 87.6(2) |
| C(2)-Lu(1)-C(14) | 87.6(3) |
| N(1)#1-N(1)-Lu(1)#1 | 74.5(5) |
| N(1)#1-N(1)-Lu(1) | 73.9(5) |
| Lu(1)#1-N(1)-Lu(1) | 148.4(3) |
| C(5)-C(1)-C(2) | 109.0(7) |
| C(5)-C(1)-C(6) | 124.0(8) |
| C(2)-C(1)-C(6) | 126.6(7) |

| | |
|---|---|
| C(5)-C(1)-Lu(1) | 72.9(4) |
| C(2)-C(1)-Lu(1) | 74.6(4) |
| C(6)-C(1)-Lu(1) | 124.6(5) |
| C(3)-C(2)-C(1) | 107.1(7) |
| C(3)-C(2)-C(7) | 124.6(8) |
| C(1)-C(2)-C(7) | 126.4(7) |
| C(3)-C(2)-Lu(1) | 72.1(4) |
| C(1)-C(2)-Lu(1) | 74.2(4) |
| C(7)-C(2)-Lu(1) | 131.4(6) |
| C(2)-C(3)-C(4) | 108.5(7) |
| C(2)-C(3)-C(8) | 125.1(9) |
| C(4)-C(3)-C(8) | 126.1(8) |
| C(2)-C(3)-Lu(1) | 76.9(4) |
| C(4)-C(3)-Lu(1) | 74.8(4) |
| C(8)-C(3)-Lu(1) | 119.8(6) |
| C(5)-C(4)-C(3) | 107.0(7) |
| C(5)-C(4)-C(9) | 125.7(8) |
| C(3)-C(4)-C(9) | 126.9(8) |
| C(5)-C(4)-Lu(1) | 73.9(4) |
| C(3)-C(4)-Lu(1) | 73.3(4) |
| C(9)-C(4)-Lu(1) | 122.9(5) |
| C(1)-C(5)-C(4) | 108.2(7) |
| C(1)-C(5)-C(10) | 126.1(8) |
| C(4)-C(5)-C(10) | 125.4(8) |
| C(1)-C(5)-Lu(1) | 76.4(4) |
| C(4)-C(5)-Lu(1) | 74.4(4) |
| C(10)-C(5)-Lu(1) | 120.1(5) |
| C(12)-C(11)-C(15) | 107.3(8) |
| C(12)-C(11)-C(16) | 113.7(11) |
| C(15)-C(11)-C(16) | 138.8(12) |
| C(12)-C(11)-C(16A) | 138.4(11) |
| C(15)-C(11)-C(16A) | 113.8(11) |
| C(12)-C(11)-Lu(1) | 73.0(5) |
| C(15)-C(11)-Lu(1) | 75.0(5) |
| C(16)-C(11)-Lu(1) | 119.7(8) |
| C(11)-C(12)-C(13) | 108.3(8) |

| | |
|---|---|
| C(11)-C(12)-C(17A) | 116.1(12) |
| C(13)-C(12)-C(17A) | 135.3(12) |
| C(11)-C(12)-C(17) | 139.0(11) |
| C(13)-C(12)-C(17) | 112.7(11) |
| C(11)-C(12)-Lu(1) | 75.7(4) |
| C(13)-C(12)-Lu(1) | 77.0(5) |
| C(17)-C(12)-Lu(1) | 112.2(9) |
| C(14)-C(13)-C(12) | 108.4(8) |
| C(14)-C(13)-C(18A) | 123.0(11) |
| C(12)-C(13)-C(18A) | 127.5(11) |
| C(14)-C(13)-C(18) | 140.7(12) |
| C(12)-C(13)-C(18) | 110.8(12) |
| C(14)-C(13)-Lu(1) | 75.4(4) |
| C(12)-C(13)-Lu(1) | 72.0(4) |
| C(18)-C(13)-Lu(1) | 114.5(10) |
| C(15)-C(14)-C(13) | 108.9(8) |
| C(15)-C(14)-C(19) | 136.4(12) |
| C(13)-C(14)-C(19) | 113.6(11) |
| C(15)-C(14)-C(19A) | 115.5(10) |
| C(13)-C(14)-C(19A) | 134.2(11) |
| C(15)-C(14)-Lu(1) | 74.0(5) |
| C(13)-C(14)-Lu(1) | 74.1(5) |
| C(19)-C(14)-Lu(1) | 126.8(9) |
| C(14)-C(15)-C(11) | 107.0(8) |
| C(14)-C(15)-C(20A) | 137.3(15) |
| C(11)-C(15)-C(20A) | 114.4(14) |
| C(14)-C(15)-C(20) | 116.4(10) |
| C(11)-C(15)-C(20) | 135.8(11) |
| C(14)-C(15)-Lu(1) | 75.6(5) |
| C(11)-C(15)-Lu(1) | 73.1(5) |
| C(20)-C(15)-Lu(1) | 124.3(8) |

---

Symmetry transformations used to generate equivalent atoms:

#1 -x,-y+1,z

**Table S6** X-ray Data Collection Parameters of **1-Gd** and **1-Tb**.

| | **1-Gd** | **1-Tb** |
|---|---|---|
| formula | $C_{40}H_{60}Gd_2N_2$ | $C_{40}H_{60}Tb_2N_2$ |
| fw | 883.40 | 886.74 |
| temp (K) | 88(2) | 88(2) |
| Wavelength | 0.71073 Å | 0.71073 Å |
| cryst syst | Tetragonal | Tetragonal |
| space group | $P\bar{4}2_1c$ | $P\bar{4}2_1c$ |
| a (Å) | 14.4239(19) | 14.4075(15) |
| b (Å) | 14.4239(19) | 14.4075(15) |
| c (Å) | 19.665(3) | 19.627(2) |
| Volume (Å$^3$) | 4091.3(12) | 4074.0(10) |
| α (deg) | 90 | 90 |
| β (deg) | 90 | 90 |
| γ (deg) | 90 | 90 |
| Z | 4 | 4 |
| $\rho_{calc}$ (Mg/m$^3$) | 1.434 | 1.446 |
| μ (mm$^{-1}$) | 3.240 | 3.469 |
| R1 ($I > 2\sigma(I)$)[a] | 0.0169 | 0.0159 |
| wR2 (all data)[a] | 0.0424 | 0.0377 |

[a]Definitions: $wR2 = [\sum[w(F_o^2 - F_c^2)^2]/ \sum[w(F_o^2)^2] ]^{1/2}$; $R1 = \sum||F_o| - |F_c||/\sum|F_o|$.

**X-ray Data Collection, Structure Solution and Refinement for 1-Gd.** A red crystal of approximate dimensions 0.248 x 0.296 x 0.618 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[13] program package was used to

determine the unit-cell parameters and for data collection (5 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[9] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 4/*mmm* and the systematic absences were consistent with the tetragonal space group $P\bar{4}2_1c$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. At convergence, wR2 = 0.0424 and Goof = 1.052 for 209 variables refined against 5262 data (0.73Å), R1 = 0.0169 for those 5068 data with $I > 2.0\sigma(I)$. The absolute structure was assigned by refinement of the Flack parameter.[10] There were high residuals present in the final difference-Fourier map. It was not possible to determine the nature of the residuals although it was probable that benzene or methylcyclohexane solvent was present. The SQUEEZE routine in the PLATON[12,14] program package was used to account for the electrons in the solvent accessible voids.

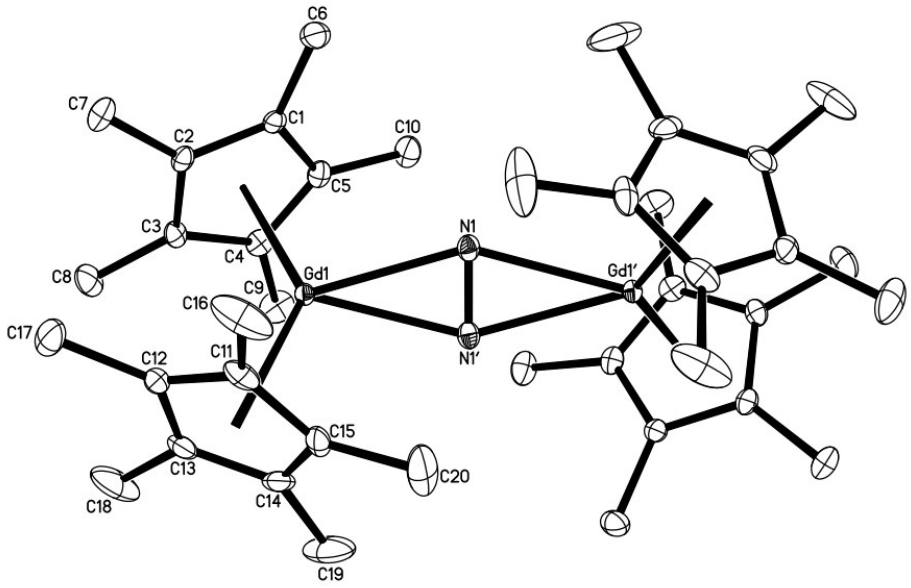


**Fig. S5** Thermal ellipsoid plot of $[(C_5Me_5)_2Gd]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Gd**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S7** Bond lengths [Å] and angles [°] for **1-Gd**.

| | |
|---|---|
| Gd(1)-Cnt1 | 2.409 |
| Gd(1)-Cnt2 | 2.429 |
| Gd(1)-N(1)#1 | 2.363(2) |
| Gd(1)-N(1) | 2.370(2) |
| Gd(1)-C(14) | 2.670(3) |
| Gd(1)-C(4) | 2.679(3) |
| Gd(1)-C(1) | 2.685(3) |
| Gd(1)-C(5) | 2.695(3) |
| Gd(1)-C(15) | 2.698(3) |
| Gd(1)-C(2) | 2.707(3) |
| Gd(1)-C(3) | 2.709(3) |
| Gd(1)-C(13) | 2.717(3) |
| Gd(1)-C(11) | 2.724(3) |
| Gd(1)-C(12) | 2.735(3) |
| N(1)-N(1)#1 | 1.236(5) |
| N(1)-Gd(1)#1 | 2.363(2) |

| | |
|---|---|
| C(1)-C(2) | 1.416(5) |
| C(1)-C(5) | 1.420(5) |
| C(1)-C(6) | 1.502(5) |
| C(2)-C(3) | 1.415(5) |
| C(2)-C(7) | 1.500(4) |
| C(3)-C(4) | 1.423(5) |
| C(3)-C(8) | 1.499(5) |
| C(4)-C(5) | 1.424(5) |
| C(4)-C(9) | 1.512(5) |
| C(5)-C(10) | 1.510(5) |
| C(11)-C(12) | 1.410(5) |
| C(11)-C(15) | 1.420(5) |
| C(11)-C(16) | 1.488(6) |
| C(12)-C(13) | 1.410(5) |
| C(12)-C(17) | 1.504(5) |
| C(13)-C(14) | 1.400(5) |
| C(13)-C(18) | 1.522(5) |
| C(14)-C(15) | 1.412(6) |
| C(14)-C(19) | 1.514(6) |
| C(15)-C(20) | 1.504(5) |

| | |
|---|---|
| Cnt1-Gd(1)-N(1) | 103.1 |
| Cnt1-Gd(1)-N(1)#1 | 120.6 |
| Cnt2-Gd(1)-N(1) | 121.3 |
| Cnt2-Gd(1)-N(1)#1 | 101.6 |
| Cnt1-Gd(1)-Cnt2 | 135.4 |
| N(1)#1-Gd(1)-N(1) | 30.28(11) |
| N(1)#1-Gd(1)-C(14) | 84.31(10) |
| N(1)-Gd(1)-C(14) | 111.58(11) |
| N(1)#1-Gd(1)-C(4) | 107.31(10) |
| N(1)-Gd(1)-C(4) | 102.99(10) |
| C(14)-Gd(1)-C(4) | 119.53(12) |
| N(1)#1-Gd(1)-C(1) | 111.93(9) |
| N(1)-Gd(1)-C(1) | 85.82(9) |
| C(14)-Gd(1)-C(1) | 162.44(11) |
| C(4)-Gd(1)-C(1) | 50.72(10) |

| | |
|---|---|
| N(1)#1-Gd(1)-C(5) | 94.03(10) |
| N(1)-Gd(1)-C(5) | 78.07(10) |
| C(14)-Gd(1)-C(5) | 147.55(12) |
| C(4)-Gd(1)-C(5) | 30.73(10) |
| C(1)-Gd(1)-C(5) | 30.61(10) |
| N(1)#1-Gd(1)-C(15) | 76.98(10) |
| N(1)-Gd(1)-C(15) | 94.84(10) |
| C(14)-Gd(1)-C(15) | 30.51(12) |
| C(4)-Gd(1)-C(15) | 150.04(11) |
| C(1)-Gd(1)-C(15) | 156.24(12) |
| C(5)-Gd(1)-C(15) | 170.68(11) |
| N(1)#1-Gd(1)-C(2) | 141.88(9) |
| N(1)-Gd(1)-C(2) | 116.03(9) |
| C(14)-Gd(1)-C(2) | 132.37(11) |
| C(4)-Gd(1)-C(2) | 50.38(10) |
| C(1)-Gd(1)-C(2) | 30.45(10) |
| C(5)-Gd(1)-C(2) | 50.26(10) |
| C(15)-Gd(1)-C(2) | 139.05(10) |
| N(1)#1-Gd(1)-C(3) | 137.92(10) |
| N(1)-Gd(1)-C(3) | 128.50(9) |
| C(14)-Gd(1)-C(3) | 113.06(12) |
| C(4)-Gd(1)-C(3) | 30.62(10) |
| C(1)-Gd(1)-C(3) | 50.41(9) |
| C(5)-Gd(1)-C(3) | 50.44(10) |
| C(15)-Gd(1)-C(3) | 136.39(10) |
| C(2)-Gd(1)-C(3) | 30.29(10) |
| N(1)#1-Gd(1)-C(13) | 114.17(10) |
| N(1)-Gd(1)-C(13) | 141.41(10) |
| C(14)-Gd(1)-C(13) | 30.10(11) |
| C(4)-Gd(1)-C(13) | 104.52(11) |
| C(1)-Gd(1)-C(13) | 132.72(11) |
| C(5)-Gd(1)-C(13) | 134.72(11) |
| C(15)-Gd(1)-C(13) | 49.81(11) |
| C(2)-Gd(1)-C(13) | 102.34(10) |
| C(3)-Gd(1)-C(13) | 87.17(11) |
| N(1)#1-Gd(1)-C(11) | 102.16(10) |

| | |
|---|---|
| N(1)-Gd(1)-C(11) | 109.27(10) |
| C(14)-Gd(1)-C(11) | 50.15(11) |
| C(4)-Gd(1)-C(11) | 147.59(11) |
| C(1)-Gd(1)-C(11) | 127.85(11) |
| C(5)-Gd(1)-C(11) | 158.24(11) |
| C(15)-Gd(1)-C(11) | 30.36(11) |
| C(2)-Gd(1)-C(11) | 109.86(11) |
| C(3)-Gd(1)-C(11) | 118.57(10) |
| C(13)-Gd(1)-C(11) | 49.67(11) |
| N(1)#1-Gd(1)-C(12) | 126.64(10) |
| N(1)-Gd(1)-C(12) | 139.19(10) |
| C(14)-Gd(1)-C(12) | 49.76(11) |
| C(4)-Gd(1)-C(12) | 117.79(11) |
| C(1)-Gd(1)-C(12) | 118.15(10) |
| C(5)-Gd(1)-C(12) | 139.11(10) |
| C(15)-Gd(1)-C(12) | 49.69(10) |
| C(2)-Gd(1)-C(12) | 90.65(10) |
| C(3)-Gd(1)-C(12) | 90.31(10) |
| C(13)-Gd(1)-C(12) | 29.99(11) |
| C(11)-Gd(1)-C(12) | 29.93(11) |
| N(1)#1-N(1)-Gd(1)#1 | 75.2(2) |
| N(1)#1-N(1)-Gd(1) | 74.5(2) |
| Gd(1)#1-N(1)-Gd(1) | 149.42(11) |
| C(2)-C(1)-C(5) | 108.0(3) |
| C(2)-C(1)-C(6) | 126.5(3) |
| C(5)-C(1)-C(6) | 125.4(3) |
| C(2)-C(1)-Gd(1) | 75.66(17) |
| C(5)-C(1)-Gd(1) | 75.09(18) |
| C(6)-C(1)-Gd(1) | 117.8(2) |
| C(3)-C(2)-C(1) | 108.4(3) |
| C(3)-C(2)-C(7) | 126.3(3) |
| C(1)-C(2)-C(7) | 124.9(3) |
| C(3)-C(2)-Gd(1) | 74.92(17) |
| C(1)-C(2)-Gd(1) | 73.90(17) |
| C(7)-C(2)-Gd(1) | 122.7(2) |
| C(2)-C(3)-C(4) | 107.8(3) |

| | |
|---|---|
| C(2)-C(3)-C(8) | 125.9(3) |
| C(4)-C(3)-C(8) | 125.1(3) |
| C(2)-C(3)-Gd(1) | 74.79(17) |
| C(4)-C(3)-Gd(1) | 73.55(17) |
| C(8)-C(3)-Gd(1) | 127.3(2) |
| C(3)-C(4)-C(5) | 107.9(3) |
| C(3)-C(4)-C(9) | 125.1(3) |
| C(5)-C(4)-C(9) | 126.9(3) |
| C(3)-C(4)-Gd(1) | 75.83(17) |
| C(5)-C(4)-Gd(1) | 75.23(18) |
| C(9)-C(4)-Gd(1) | 117.9(2) |
| C(1)-C(5)-C(4) | 107.8(3) |
| C(1)-C(5)-C(10) | 124.9(3) |
| C(4)-C(5)-C(10) | 127.1(3) |
| C(1)-C(5)-Gd(1) | 74.30(18) |
| C(4)-C(5)-Gd(1) | 74.04(18) |
| C(10)-C(5)-Gd(1) | 121.7(2) |
| C(12)-C(11)-C(15) | 107.6(3) |
| C(12)-C(11)-C(16) | 124.6(4) |
| C(15)-C(11)-C(16) | 127.3(4) |
| C(12)-C(11)-Gd(1) | 75.45(19) |
| C(15)-C(11)-Gd(1) | 73.8(2) |
| C(16)-C(11)-Gd(1) | 123.5(3) |
| C(11)-C(12)-C(13) | 108.3(3) |
| C(11)-C(12)-C(17) | 125.1(4) |
| C(13)-C(12)-C(17) | 125.6(4) |
| C(11)-C(12)-Gd(1) | 74.63(19) |
| C(13)-C(12)-Gd(1) | 74.32(19) |
| C(17)-C(12)-Gd(1) | 126.3(2) |
| C(14)-C(13)-C(12) | 108.1(3) |
| C(14)-C(13)-C(18) | 124.9(4) |
| C(12)-C(13)-C(18) | 126.4(4) |
| C(14)-C(13)-Gd(1) | 73.08(19) |
| C(12)-C(13)-Gd(1) | 75.69(18) |
| C(18)-C(13)-Gd(1) | 124.2(3) |
| C(13)-C(14)-C(15) | 108.4(3) |

| | |
|---|---|
| C(13)-C(14)-C(19) | 125.3(5) |
| C(15)-C(14)-C(19) | 126.3(4) |
| C(13)-C(14)-Gd(1) | 76.82(19) |
| C(15)-C(14)-Gd(1) | 75.82(19) |
| C(19)-C(14)-Gd(1) | 115.6(3) |
| C(14)-C(15)-C(11) | 107.7(3) |
| C(14)-C(15)-C(20) | 124.6(5) |
| C(11)-C(15)-C(20) | 127.6(5) |
| C(14)-C(15)-Gd(1) | 73.67(19) |
| C(11)-C(15)-Gd(1) | 75.9(2) |
| C(20)-C(15)-Gd(1) | 120.1(3) |

Symmetry transformations used to generate equivalent atoms:
#1 -x+2,-y+1,z

**X-ray Data Collection, Structure Solution and Refinement for 1-Tb.** A purple crystal of approximate dimensions 0.095 x 0.109 x 0.264 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[13] program package was used to determine the unit-cell parameters and for data collection (20 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[9] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 4/*mmm* and the systematic absences were consistent with the tetragonal space group $P\bar{4}2_1c$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. At convergence, wR2 = 0.0377 and Goof = 1.028 for 209 variables refined against 5254 data (0.73Å), R1 = 0.0159 for those 5092 data with $I > 2.0\sigma(I)$. The absolute structure was

assigned by refinement of the Flack parameter.[10] There were high residuals present in the final difference-Fourier map. It was not possible to determine the nature of the residuals although it was probable that benzene or methylcyclohexane solvent was present. The SQUEEZE routine in the PLATON[12,14] program package was used to account for the electrons in the solvent accessible voids.

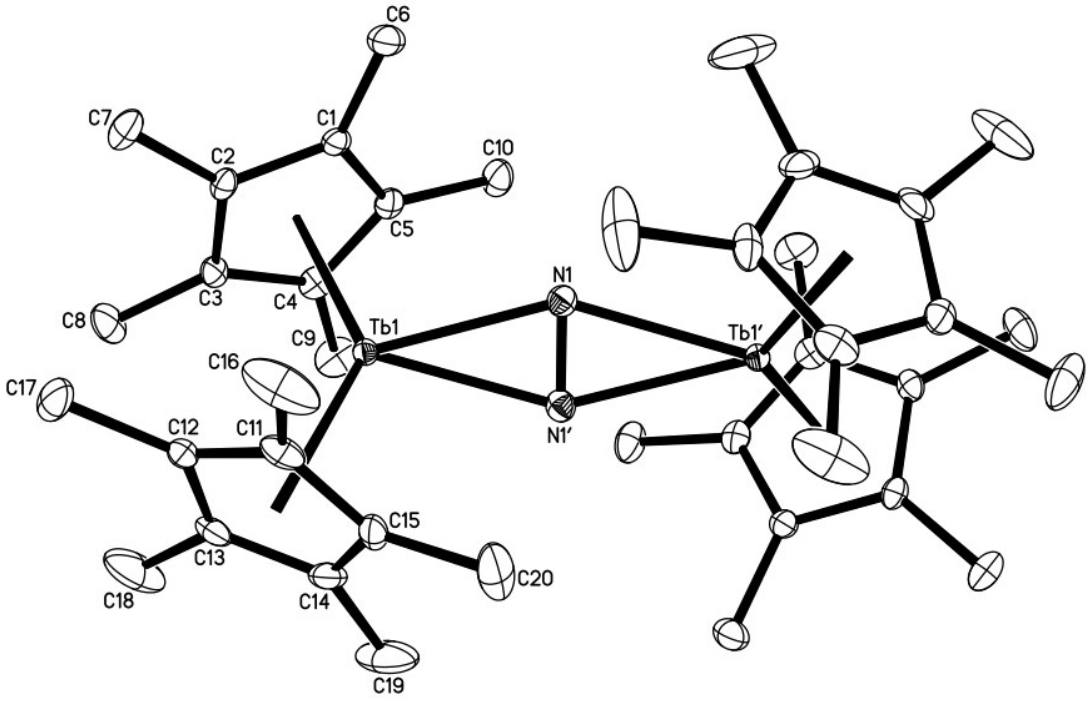


**Fig. S6** Thermal ellipsoid plot of $[(C_5Me_5)_2Tb]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **1-Tb**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S8** Bond lengths [Å] and angles [°] for **1-Tb**.

| | |
|---|---|
| Tb(1)-Cnt1 | 2.392 |
| Tb(1)-Cnt2 | 2.412 |
| Tb(1)-N(1)#1 | 2.347(2) |
| Tb(1)-N(1) | 2.350(2) |
| Tb(1)-C(14) | 2.656(3) |
| Tb(1)-C(4) | 2.663(3) |
| Tb(1)-C(1) | 2.669(3) |
| Tb(1)-C(5) | 2.673(3) |

| | |
|---|---|
| Tb(1)-C(15) | 2.679(3) |
| Tb(1)-C(2) | 2.693(3) |
| Tb(1)-C(3) | 2.695(3) |
| Tb(1)-C(13) | 2.705(3) |
| Tb(1)-C(11) | 2.708(3) |
| Tb(1)-C(12) | 2.718(3) |
| N(1)-N(1)#1 | 1.238(4) |
| N(1)-Tb(1)#1 | 2.347(2) |
| C(1)-C(5) | 1.414(4) |
| C(1)-C(2) | 1.420(4) |
| C(1)-C(6) | 1.502(4) |
| C(2)-C(3) | 1.414(4) |
| C(2)-C(7) | 1.503(4) |
| C(3)-C(4) | 1.418(4) |
| C(3)-C(8) | 1.508(4) |
| C(4)-C(5) | 1.421(5) |
| C(4)-C(9) | 1.512(4) |
| C(5)-C(10) | 1.512(4) |
| C(11)-C(12) | 1.402(5) |
| C(11)-C(15) | 1.415(5) |
| C(11)-C(16) | 1.496(6) |
| C(12)-C(13) | 1.420(5) |
| C(12)-C(17) | 1.506(5) |
| C(13)-C(14) | 1.402(5) |
| C(13)-C(18) | 1.520(5) |
| C(14)-C(15) | 1.404(5) |
| C(14)-C(19) | 1.516(5) |
| C(15)-C(20) | 1.504(5) |

| | |
|---|---|
| Cnt1-Tb(1)-N(1) | 103.2 |
| Cnt1-Tb(1)-N(1)#1 | 121.3 |
| Cnt2-Tb(1)-N(1) | 121.4 |
| Cnt2-Tb(1)-N(1)#1 | 101.5 |
| Cnt1-Tb(1)-Cnt2 | 135.1 |
| N(1)#1-Tb(1)-N(1) | 30.55(11) |
| N(1)#1-Tb(1)-C(14) | 84.13(10) |

| | |
|---|---|
| N(1)-Tb(1)-C(14) | 111.71(10) |
| N(1)#1-Tb(1)-C(4) | 108.17(9) |
| N(1)-Tb(1)-C(4) | 103.47(9) |
| C(14)-Tb(1)-C(4) | 119.62(11) |
| N(1)#1-Tb(1)-C(1) | 112.12(9) |
| N(1)-Tb(1)-C(1) | 85.54(8) |
| C(14)-Tb(1)-C(1) | 162.64(10) |
| C(4)-Tb(1)-C(1) | 50.95(9) |
| N(1)#1-Tb(1)-C(5) | 94.57(9) |
| N(1)-Tb(1)-C(5) | 78.21(9) |
| C(14)-Tb(1)-C(5) | 147.78(11) |
| C(4)-Tb(1)-C(5) | 30.88(10) |
| C(1)-Tb(1)-C(5) | 30.69(9) |
| N(1)#1-Tb(1)-C(15) | 76.74(9) |
| N(1)-Tb(1)-C(15) | 94.90(9) |
| C(14)-Tb(1)-C(15) | 30.52(11) |
| C(4)-Tb(1)-C(15) | 150.13(11) |
| C(1)-Tb(1)-C(15) | 155.75(11) |
| C(5)-Tb(1)-C(15) | 171.03(10) |
| N(1)#1-Tb(1)-C(2) | 142.43(9) |
| N(1)-Tb(1)-C(2) | 115.94(9) |
| C(14)-Tb(1)-C(2) | 132.29(10) |
| C(4)-Tb(1)-C(2) | 50.62(9) |
| C(1)-Tb(1)-C(2) | 30.70(9) |
| C(5)-Tb(1)-C(2) | 50.54(9) |
| C(15)-Tb(1)-C(2) | 138.43(9) |
| N(1)#1-Tb(1)-C(3) | 138.86(9) |
| N(1)-Tb(1)-C(3) | 128.84(9) |
| C(14)-Tb(1)-C(3) | 113.06(11) |
| C(4)-Tb(1)-C(3) | 30.70(9) |
| C(1)-Tb(1)-C(3) | 50.68(9) |
| C(5)-Tb(1)-C(3) | 50.65(9) |
| C(15)-Tb(1)-C(3) | 136.09(9) |
| C(2)-Tb(1)-C(3) | 30.44(9) |
| N(1)#1-Tb(1)-C(13) | 114.19(9) |
| N(1)-Tb(1)-C(13) | 141.75(9) |

| | |
|---|---|
| C(14)-Tb(1)-C(13) | 30.30(11) |
| C(4)-Tb(1)-C(13) | 104.11(10) |
| C(1)-Tb(1)-C(13) | 132.64(10) |
| C(5)-Tb(1)-C(13) | 134.48(10) |
| C(15)-Tb(1)-C(13) | 50.03(10) |
| C(2)-Tb(1)-C(13) | 102.02(10) |
| C(3)-Tb(1)-C(13) | 86.75(10) |
| N(1)#1-Tb(1)-C(11) | 101.94(10) |
| N(1)-Tb(1)-C(11) | 109.19(10) |
| C(14)-Tb(1)-C(11) | 50.30(10) |
| C(4)-Tb(1)-C(11) | 147.12(10) |
| C(1)-Tb(1)-C(11) | 127.32(10) |
| C(5)-Tb(1)-C(11) | 157.76(10) |
| C(15)-Tb(1)-C(11) | 30.46(10) |
| C(2)-Tb(1)-C(11) | 109.07(10) |
| C(3)-Tb(1)-C(11) | 117.96(10) |
| C(13)-Tb(1)-C(11) | 49.93(11) |
| N(1)#1-Tb(1)-C(12) | 126.61(9) |
| N(1)-Tb(1)-C(12) | 139.13(9) |
| C(14)-Tb(1)-C(12) | 50.12(10) |
| C(4)-Tb(1)-C(12) | 117.32(10) |
| C(1)-Tb(1)-C(12) | 117.67(9) |
| C(5)-Tb(1)-C(12) | 138.65(10) |
| C(15)-Tb(1)-C(12) | 49.89(9) |
| C(2)-Tb(1)-C(12) | 89.90(9) |
| C(3)-Tb(1)-C(12) | 89.69(10) |
| C(13)-Tb(1)-C(12) | 30.35(10) |
| C(11)-Tb(1)-C(12) | 29.95(10) |
| N(1)#1-N(1)-Tb(1)#1 | 74.9(2) |
| N(1)#1-N(1)-Tb(1) | 74.6(2) |
| Tb(1)#1-N(1)-Tb(1) | 149.24(11) |
| C(5)-C(1)-C(2) | 107.9(3) |
| C(5)-C(1)-C(6) | 126.0(3) |
| C(2)-C(1)-C(6) | 126.1(3) |
| C(5)-C(1)-Tb(1) | 74.81(16) |
| C(2)-C(1)-Tb(1) | 75.59(16) |

| | |
|---|---|
| C(6)-C(1)-Tb(1) | 118.2(2) |
| C(3)-C(2)-C(1) | 108.2(3) |
| C(3)-C(2)-C(7) | 126.3(3) |
| C(1)-C(2)-C(7) | 125.0(3) |
| C(3)-C(2)-Tb(1) | 74.84(16) |
| C(1)-C(2)-Tb(1) | 73.71(16) |
| C(7)-C(2)-Tb(1) | 123.8(2) |
| C(2)-C(3)-C(4) | 107.9(3) |
| C(2)-C(3)-C(8) | 125.7(3) |
| C(4)-C(3)-C(8) | 125.2(3) |
| C(2)-C(3)-Tb(1) | 74.72(15) |
| C(4)-C(3)-Tb(1) | 73.41(16) |
| C(8)-C(3)-Tb(1) | 127.6(2) |
| C(3)-C(4)-C(5) | 108.0(3) |
| C(3)-C(4)-C(9) | 125.2(3) |
| C(5)-C(4)-C(9) | 126.7(3) |
| C(3)-C(4)-Tb(1) | 75.90(16) |
| C(5)-C(4)-Tb(1) | 74.95(16) |
| C(9)-C(4)-Tb(1) | 118.1(2) |
| C(1)-C(5)-C(4) | 108.0(3) |
| C(1)-C(5)-C(10) | 124.7(3) |
| C(4)-C(5)-C(10) | 127.0(3) |
| C(1)-C(5)-Tb(1) | 74.50(16) |
| C(4)-C(5)-Tb(1) | 74.17(17) |
| C(10)-C(5)-Tb(1) | 121.7(2) |
| C(12)-C(11)-C(15) | 107.8(3) |
| C(12)-C(11)-C(16) | 124.3(4) |
| C(15)-C(11)-C(16) | 127.3(4) |
| C(12)-C(11)-Tb(1) | 75.45(18) |
| C(15)-C(11)-Tb(1) | 73.64(18) |
| C(16)-C(11)-Tb(1) | 123.6(2) |
| C(11)-C(12)-C(13) | 108.1(3) |
| C(11)-C(12)-C(17) | 125.4(4) |
| C(13)-C(12)-C(17) | 125.3(3) |
| C(11)-C(12)-Tb(1) | 74.60(17) |
| C(13)-C(12)-Tb(1) | 74.29(17) |

| | |
|---|---|
| C(17)-C(12)-Tb(1) | 126.9(2) |
| C(14)-C(13)-C(12) | 107.6(3) |
| C(14)-C(13)-C(18) | 125.0(4) |
| C(12)-C(13)-C(18) | 126.7(4) |
| C(14)-C(13)-Tb(1) | 72.92(18) |
| C(12)-C(13)-Tb(1) | 75.36(17) |
| C(18)-C(13)-Tb(1) | 124.3(3) |
| C(13)-C(14)-C(15) | 108.4(3) |
| C(13)-C(14)-C(19) | 124.5(4) |
| C(15)-C(14)-C(19) | 127.0(4) |
| C(13)-C(14)-Tb(1) | 76.77(18) |
| C(15)-C(14)-Tb(1) | 75.64(17) |
| C(19)-C(14)-Tb(1) | 115.8(3) |
| C(14)-C(15)-C(11) | 108.0(3) |
| C(14)-C(15)-C(20) | 124.8(4) |
| C(11)-C(15)-C(20) | 127.0(4) |
| C(14)-C(15)-Tb(1) | 73.85(18) |
| C(11)-C(15)-Tb(1) | 75.91(18) |
| C(20)-C(15)-Tb(1) | 120.4(2) |

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Symmetry transformations used to generate equivalent atoms:
#1 -x+2,-y+1,z

**Table S9** X-ray Data Collection Parameters of **2-Gd** and **2-Dy**.

| | **2-Gd** | **2-Dy** |
|---|---|---|
| formula | $C_{44}H_{68}Gd_2N_2O_2$ •2($C_7H_8$) | $C_{44}H_{68}Dy_2N_2O_2$ |
| fw | 1155.77 | 982.00 |
| temp (K) | 88(2) | 133(2) |
| Wavelength | 0.71073 Å | 0.71073 Å |
| cryst syst | Monoclinic | Monoclinic |
| space group | *C*2/*c* | *C*2/*c* |
| a (Å) | 15.2453(7) | 15.7376(15) |
| b (Å) | 13.9938(6) | 14.1959(14) |
| c (Å) | 25.6281(11) | 18.9717(18) |
| Volume (Å$^3$) | 5286.7(4) | 4157.5(7) |
| α (deg) | 90 | 90 |
| β (deg) | 104.7734(4) | 101.2185(11) |
| γ (deg) | 90 | 90 |
| Z | 4 | 4 |
| $\rho_{calc}$ (Mg/m$^3$) | 1.452 | 1.569 |
| μ (mm$^{-1}$) | 2.529 | 3.603 |
| R1 (I > 2σ(I))[a] | 0.0190 | 0.0316 |
| wR2 (all data)[a] | 0.0449 | 0.0790 |

[a]Definitions: $wR2 = [\sum[w(F_o^2 - F_c^2)^2]/ \sum[w(F_o^2)^2] ]^{1/2}$; $R1 = \sum||F_o| - |F_c||/\sum|F_o|$.

**X-ray Data Collection, Structure Solution and Refinement for 2-Gd.** A green crystal of approximate dimensions 0.237 x 0.280 x 0.374 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[13] program package was

used to determine the unit-cell parameters and for data collection (10 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[9] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 2/*m* and the systematic absences were consistent with the monoclinic space groups *Cc* and *C*2/*c*. It was later determined that space group *C*2/*c* was correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atom H(28) was located from a difference-Fourier map and refined (x,y,z and $U_{iso}$). The remaining hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. A methyl carbon atom on one tetramethylcyclopentadienyl ligand was disordered over two positions (C(16) / C(19)) and included using multiple components with partial site-occupancy factors. The hydrogen atom associated with the disordered tetramethylcyclopentadienyl ligand was not included in the refinement. There was a molecule of toluene solvent present (two per dimeric formula-unit). The solvent was disordered. Carbon atoms C(24)-C(27) were included as above. At convergence, wR2 = 0.0449 and Goof = 1.071 for 350 variables refined against 6785 data (0.73Å), R1 = 0.0190 for those 6008 data with $I > 2.0\sigma(I)$.

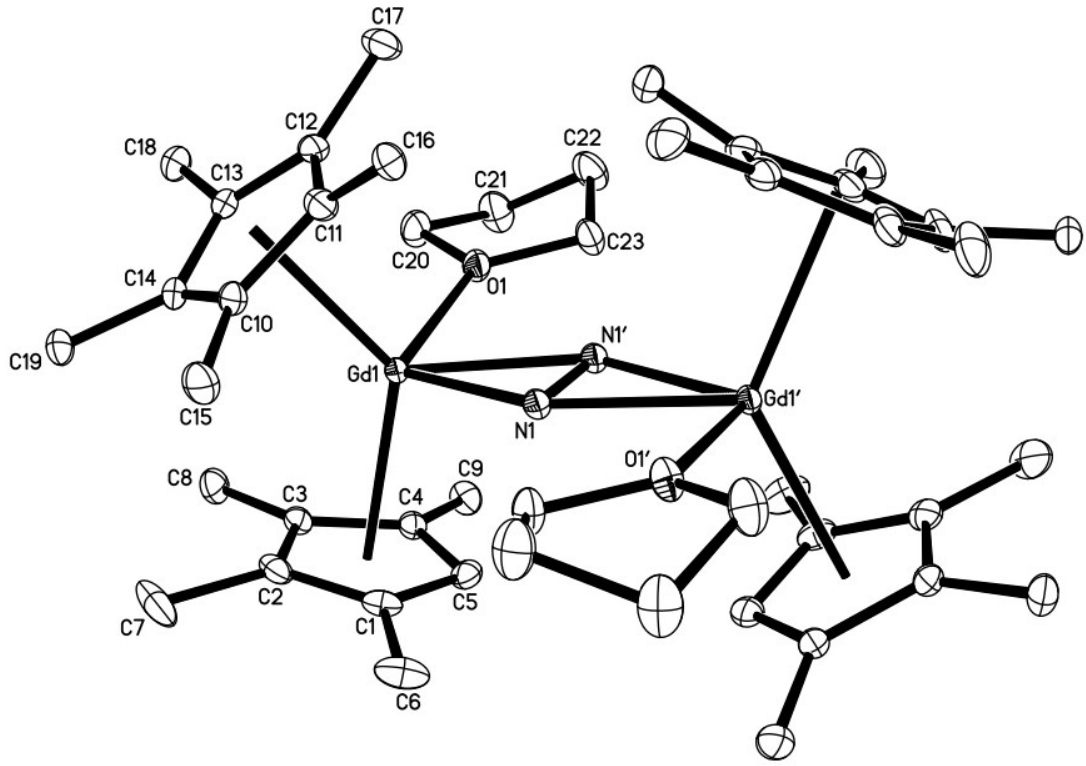


**Fig. S7** Thermal ellipsoid plot of $[(C_5Me_4H)_2Gd(THF)](\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **2-Gd**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S10** Bond lengths [Å] and angles [°] for **2-Gd**.

| | |
|---|---|
| Gd(1)-Cnt1 | 2.448 |
| Gd(1)-Cnt2 | 2.459 |
| Gd(1)-N(1) | 2.3703(16) |
| Gd(1)-N(1)#1 | 2.4057(15) |
| Gd(1)-O(1) | 2.4870(13) |
| Gd(1)-C(10) | 2.6840(19) |
| Gd(1)-C(5) | 2.6917(18) |
| Gd(1)-C(11) | 2.7048(18) |
| Gd(1)-C(1) | 2.7070(18) |
| Gd(1)-C(4) | 2.7338(18) |
| Gd(1)-C(14) | 2.7428(19) |
| Gd(1)-C(2) | 2.7516(18) |

| | |
|---|---|
| Gd(1)-C(3) | 2.7596(18) |
| Gd(1)-C(12) | 2.7695(18) |
| Gd(1)-C(13) | 2.7910(18) |
| O(1)-C(20) | 1.453(2) |
| O(1)-C(23) | 1.456(2) |
| N(1)-N(1)#1 | 1.247(3) |
| N(1)-Gd(1)#1 | 2.4057(15) |
| C(1)-C(2) | 1.408(3) |
| C(1)-C(5) | 1.423(3) |
| C(1)-C(6) | 1.503(3) |
| C(2)-C(3) | 1.422(3) |
| C(2)-C(7) | 1.512(3) |
| C(3)-C(4) | 1.419(3) |
| C(3)-C(8) | 1.507(3) |
| C(4)-C(5) | 1.415(3) |
| C(4)-C(9) | 1.501(3) |
| C(10)-C(14) | 1.407(3) |
| C(10)-C(11) | 1.421(3) |
| C(10)-C(15) | 1.508(3) |
| C(11)-C(16) | 1.391(4) |
| C(11)-C(12) | 1.415(3) |
| C(12)-C(13) | 1.416(3) |
| C(12)-C(17) | 1.511(3) |
| C(13)-C(14) | 1.424(3) |
| C(13)-C(18) | 1.507(3) |
| C(14)-C(19) | 1.445(4) |
| C(20)-C(21) | 1.516(3) |
| C(21)-C(22) | 1.524(3) |
| C(22)-C(23) | 1.518(3) |
| C(24)-C(25) | 1.401(17) |
| C(24)-C(29) | 1.463(5) |
| C(25)-C(26) | 1.382(13) |
| C(26)-C(27) | 1.383(6) |
| C(27)-C(28) | 1.470(6) |
| C(24B)-C(25B) | 1.399(10) |
| C(24B)-C(29) | 1.433(6) |

| | |
|---|---|
| C(25B)-C(26B) | 1.373(19) |
| C(26B)-C(27B) | 1.369(17) |
| C(27B)-C(28) | 1.396(7) |
| C(28)-C(29) | 1.360(4) |
| C(29)-C(30) | 1.491(4) |

| | |
|---|---|
| Cnt1-Gd(1)-O(1) | 103.9 |
| Cnt1-Gd(1)-N(1) | 103.3 |
| Cnt1-Gd(1)-N(1)#1 | 108.7 |
| Cnt2-Gd(1)-O(1) | 102.3 |
| Cnt2-Gd(1)-N(1) | 104.6 |
| Cnt2-Gd(1)-N(1)#1 | 116.2 |
| Cnt1-Gd(1)-Cnt2 | 130.1 |
| N(1)-Gd(1)-N(1)#1 | 30.25(7) |
| N(1)-Gd(1)-O(1) | 112.65(5) |
| N(1)#1-Gd(1)-O(1) | 82.61(5) |
| N(1)-Gd(1)-C(10) | 89.00(6) |
| N(1)#1-Gd(1)-C(10) | 111.56(6) |
| O(1)-Gd(1)-C(10) | 128.90(5) |
| N(1)-Gd(1)-C(5) | 82.92(5) |
| N(1)#1-Gd(1)-C(5) | 82.57(5) |
| O(1)-Gd(1)-C(5) | 96.84(5) |
| C(10)-Gd(1)-C(5) | 132.62(6) |
| N(1)-Gd(1)-C(11) | 79.24(6) |
| N(1)#1-Gd(1)-C(11) | 90.57(5) |
| O(1)-Gd(1)-C(11) | 106.14(5) |
| C(10)-Gd(1)-C(11) | 30.59(6) |
| C(5)-Gd(1)-C(11) | 154.99(6) |
| N(1)-Gd(1)-C(1) | 80.58(5) |
| N(1)#1-Gd(1)-C(1) | 95.35(6) |
| O(1)-Gd(1)-C(1) | 126.07(5) |
| C(10)-Gd(1)-C(1) | 102.07(6) |
| C(5)-Gd(1)-C(1) | 30.56(6) |
| C(11)-Gd(1)-C(1) | 127.79(6) |
| N(1)-Gd(1)-C(4) | 111.81(5) |
| N(1)#1-Gd(1)-C(4) | 103.08(5) |

| | |
|---|---|
| O(1)-Gd(1)-C(4) | 77.80(5) |
| C(10)-Gd(1)-C(4) | 137.77(6) |
| C(5)-Gd(1)-C(4) | 30.22(6) |
| C(11)-Gd(1)-C(4) | 166.23(6) |
| C(1)-Gd(1)-C(4) | 50.03(6) |
| N(1)-Gd(1)-C(14) | 119.01(6) |
| N(1)#1-Gd(1)-C(14) | 139.42(6) |
| O(1)-Gd(1)-C(14) | 111.65(5) |
| C(10)-Gd(1)-C(14) | 30.03(7) |
| C(5)-Gd(1)-C(14) | 129.83(6) |
| C(11)-Gd(1)-C(14) | 49.41(6) |
| C(1)-Gd(1)-C(14) | 104.30(6) |
| C(4)-Gd(1)-C(14) | 116.83(6) |
| N(1)-Gd(1)-C(2) | 107.68(6) |
| N(1)#1-Gd(1)-C(2) | 125.22(5) |
| O(1)-Gd(1)-C(2) | 122.45(5) |
| C(10)-Gd(1)-C(2) | 89.98(6) |
| C(5)-Gd(1)-C(2) | 49.61(6) |
| C(11)-Gd(1)-C(2) | 120.55(6) |
| C(1)-Gd(1)-C(2) | 29.88(6) |
| C(4)-Gd(1)-C(2) | 49.44(5) |
| C(14)-Gd(1)-C(2) | 80.23(6) |
| N(1)-Gd(1)-C(3) | 129.16(5) |
| N(1)#1-Gd(1)-C(3) | 131.25(5) |
| O(1)-Gd(1)-C(3) | 92.78(5) |
| C(10)-Gd(1)-C(3) | 108.76(6) |
| C(5)-Gd(1)-C(3) | 49.67(6) |
| C(11)-Gd(1)-C(3) | 136.44(6) |
| C(1)-Gd(1)-C(3) | 49.69(6) |
| C(4)-Gd(1)-C(3) | 29.95(5) |
| C(14)-Gd(1)-C(3) | 87.27(6) |
| C(2)-Gd(1)-C(3) | 29.90(6) |
| N(1)-Gd(1)-C(12) | 102.20(6) |
| N(1)#1-Gd(1)-C(12) | 100.98(5) |
| O(1)-Gd(1)-C(12) | 79.71(5) |
| C(10)-Gd(1)-C(12) | 49.92(6) |

| | |
|---|---|
| C(5)-Gd(1)-C(12) | 174.61(6) |
| C(11)-Gd(1)-C(12) | 29.92(6) |
| C(1)-Gd(1)-C(12) | 151.30(6) |
| C(4)-Gd(1)-C(12) | 144.39(6) |
| C(14)-Gd(1)-C(12) | 49.05(6) |
| C(2)-Gd(1)-C(12) | 129.02(6) |
| C(3)-Gd(1)-C(12) | 126.00(6) |
| N(1)-Gd(1)-C(13) | 128.29(6) |
| N(1)#1-Gd(1)-C(13) | 130.29(5) |
| O(1)-Gd(1)-C(13) | 82.98(5) |
| C(10)-Gd(1)-C(13) | 49.70(6) |
| C(5)-Gd(1)-C(13) | 146.40(6) |
| C(11)-Gd(1)-C(13) | 49.19(6) |
| C(1)-Gd(1)-C(13) | 130.80(6) |
| C(4)-Gd(1)-C(13) | 119.73(6) |
| C(14)-Gd(1)-C(13) | 29.80(6) |
| C(2)-Gd(1)-C(13) | 102.48(6) |
| C(3)-Gd(1)-C(13) | 96.73(6) |
| C(12)-Gd(1)-C(13) | 29.49(6) |
| C(20)-O(1)-C(23) | 108.99(15) |
| C(20)-O(1)-Gd(1) | 125.61(11) |
| C(23)-O(1)-Gd(1) | 125.39(11) |
| N(1)#1-N(1)-Gd(1) | 76.45(14) |
| N(1)#1-N(1)-Gd(1)#1 | 73.31(13) |
| Gd(1)-N(1)-Gd(1)#1 | 149.66(7) |
| C(2)-C(1)-C(5) | 107.60(17) |
| C(2)-C(1)-C(6) | 126.4(2) |
| C(5)-C(1)-C(6) | 125.7(2) |
| C(2)-C(1)-Gd(1) | 76.81(10) |
| C(5)-C(1)-Gd(1) | 74.13(10) |
| C(6)-C(1)-Gd(1) | 119.63(13) |
| C(1)-C(2)-C(3) | 108.54(17) |
| C(1)-C(2)-C(7) | 125.2(2) |
| C(3)-C(2)-C(7) | 125.1(2) |
| C(1)-C(2)-Gd(1) | 73.30(10) |
| C(3)-C(2)-Gd(1) | 75.36(10) |

| | |
|---|---|
| C(7)-C(2)-Gd(1) | 127.13(13) |
| C(4)-C(3)-C(2) | 107.68(17) |
| C(4)-C(3)-C(8) | 125.97(18) |
| C(2)-C(3)-C(8) | 125.97(17) |
| C(4)-C(3)-Gd(1) | 74.02(10) |
| C(2)-C(3)-Gd(1) | 74.74(10) |
| C(8)-C(3)-Gd(1) | 122.65(12) |
| C(5)-C(4)-C(3) | 107.82(17) |
| C(5)-C(4)-C(9) | 125.08(17) |
| C(3)-C(4)-C(9) | 126.77(18) |
| C(5)-C(4)-Gd(1) | 73.24(10) |
| C(3)-C(4)-Gd(1) | 76.03(10) |
| C(9)-C(4)-Gd(1) | 121.81(12) |
| C(4)-C(5)-C(1) | 108.36(17) |
| C(4)-C(5)-Gd(1) | 76.54(10) |
| C(1)-C(5)-Gd(1) | 75.32(10) |
| C(14)-C(10)-C(11) | 107.22(18) |
| C(14)-C(10)-C(15) | 126.84(18) |
| C(11)-C(10)-C(15) | 125.7(2) |
| C(14)-C(10)-Gd(1) | 77.30(11) |
| C(11)-C(10)-Gd(1) | 75.51(10) |
| C(15)-C(10)-Gd(1) | 117.56(13) |
| C(16)-C(11)-C(12) | 125.8(2) |
| C(16)-C(11)-C(10) | 124.6(2) |
| C(12)-C(11)-C(10) | 108.56(18) |
| C(16)-C(11)-Gd(1) | 123.92(19) |
| C(12)-C(11)-Gd(1) | 77.57(11) |
| C(10)-C(11)-Gd(1) | 73.90(11) |
| C(11)-C(12)-C(13) | 107.93(17) |
| C(11)-C(12)-C(17) | 125.77(19) |
| C(13)-C(12)-C(17) | 126.10(19) |
| C(11)-C(12)-Gd(1) | 72.50(10) |
| C(13)-C(12)-Gd(1) | 76.10(10) |
| C(17)-C(12)-Gd(1) | 121.34(13) |
| C(12)-C(13)-C(14) | 107.41(19) |
| C(12)-C(13)-C(18) | 127.12(18) |

| | |
|---|---|
| C(14)-C(13)-C(18) | 124.30(18) |
| C(12)-C(13)-Gd(1) | 74.41(11) |
| C(14)-C(13)-Gd(1) | 73.22(11) |
| C(18)-C(13)-Gd(1) | 127.63(14) |
| C(10)-C(14)-C(13) | 108.88(18) |
| C(10)-C(14)-C(19) | 120.0(2) |
| C(13)-C(14)-C(19) | 127.9(2) |
| C(10)-C(14)-Gd(1) | 72.67(10) |
| C(13)-C(14)-Gd(1) | 76.98(11) |
| C(19)-C(14)-Gd(1) | 132.7(2) |
| O(1)-C(20)-C(21) | 105.43(17) |
| C(20)-C(21)-C(22) | 101.42(19) |
| C(23)-C(22)-C(21) | 102.75(17) |
| O(1)-C(23)-C(22) | 105.95(16) |
| C(25)-C(24)-C(29) | 126.9(6) |
| C(26)-C(25)-C(24) | 119.8(10) |
| C(25)-C(26)-C(27) | 120.1(8) |
| C(26)-C(27)-C(28) | 114.3(4) |
| C(25B)-C(24B)-C(29) | 108.1(6) |
| C(26B)-C(25B)-C(24B) | 121.1(9) |
| C(27B)-C(26B)-C(25B) | 120.6(14) |
| C(26B)-C(27B)-C(28) | 127.4(10) |
| C(29)-C(28)-C(27B) | 103.1(5) |
| C(29)-C(28)-C(27) | 131.7(3) |
| C(28)-C(29)-C(24B) | 137.7(4) |
| C(28)-C(29)-C(24) | 105.9(3) |
| C(28)-C(29)-C(30) | 120.8(2) |
| C(24B)-C(29)-C(30) | 101.0(4) |
| C(24)-C(29)-C(30) | 133.1(3) |

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Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,-z+1/2

**X-ray Data Collection, Structure Solution and Refinement for 2-Dy.** A green crystal of approximate dimensions 0.137 x 0.176 x 0.307 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[13] program package and the CELL_NOW[15] program were used to determine the unit-cell parameters. Data was collected using a 15 sec/frame scan time for a sphere of diffraction data. The raw frame data was processed using SAINT[40] and TWINABS[16] to yield the reflection data file (HKLF 5 format).[16] Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 2/*m* and the systematic absences were consistent with the monoclinic space groups *Cc* and *C*2/*c*. It was later determined that space group *C*2/*c* was correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about a two-fold rotation axis. At convergence, wR2 = 0.0790 and Goof = 1.078 for 235 variables refined against 4928 data (0.75Å), R1 = 0.0316 for those 4482 with $I > 2.0\sigma(I)$. The structure was refined as a two-component twin, BASF[5] = 0.1165.

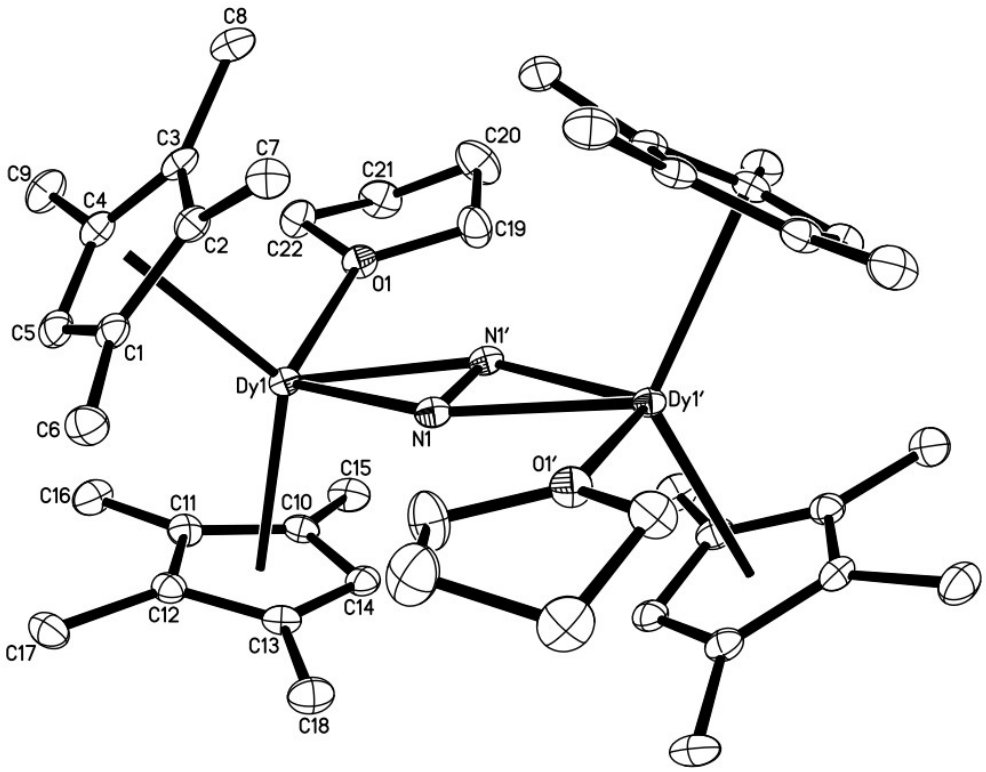


**Fig. S8** Thermal ellipsoid plot of $[(C_5Me_4H)_2Dy(THF)](\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **2-Dy**, drawn at the 30% probability level. Hydrogen atoms are omitted for clarity.

**Table S11** Bond lengths [Å] and angles [°] for **2-Dy**.

| | |
|---|---|
| Dy(1)-Cnt1 | 2.421 |
| Dy(1)-Cnt2 | 2.412 |
| Dy(1)-N(1) | 2.335(3) |
| Dy(1)-N(1)#1 | 2.366(3) |
| Dy(1)-O(1) | 2.486(3) |
| Dy(1)-C(1) | 2.657(4) |
| Dy(1)-C(5) | 2.659(4) |
| Dy(1)-C(14) | 2.660(4) |
| Dy(1)-C(10) | 2.690(4) |
| Dy(1)-C(13) | 2.692(4) |
| Dy(1)-C(11) | 2.706(4) |
| Dy(1)-C(2) | 2.716(4) |

| | |
|---|---|
| Dy(1)-C(4) | 2.728(4) |
| Dy(1)-C(12) | 2.730(4) |
| Dy(1)-C(3) | 2.763(4) |
| O(1)-C(19) | 1.448(5) |
| O(1)-C(22) | 1.454(5) |
| N(1)-N(1)#1 | 1.243(7) |
| N(1)-Dy(1)#1 | 2.366(3) |
| C(1)-C(5) | 1.417(6) |
| C(1)-C(2) | 1.422(6) |
| C(1)-C(6) | 1.496(6) |
| C(2)-C(3) | 1.422(6) |
| C(2)-C(7) | 1.502(5) |
| C(3)-C(4) | 1.422(6) |
| C(3)-C(8) | 1.503(6) |
| C(4)-C(5) | 1.412(6) |
| C(4)-C(9) | 1.514(6) |
| C(10)-C(14) | 1.412(6) |
| C(10)-C(11) | 1.418(6) |
| C(10)-C(15) | 1.506(6) |
| C(11)-C(12) | 1.411(6) |
| C(11)-C(16) | 1.505(6) |
| C(12)-C(13) | 1.410(6) |
| C(12)-C(17) | 1.509(6) |
| C(13)-C(14) | 1.429(6) |
| C(13)-C(18) | 1.512(6) |
| C(19)-C(20) | 1.502(7) |
| C(20)-C(21) | 1.516(7) |
| C(21)-C(22) | 1.511(6) |

| | |
|---|---|
| Cnt1-Dy(1)-O(1) | 102.9 |
| Cnt1-Dy(1)-N(1) | 105.8 |
| Cnt1-Dy(1)-N(1)#1 | 118.7 |
| Cnt2-Dy(1)-O(1) | 105.2 |
| Cnt2-Dy(1)-N(1) | 104.4 |
| Cnt2-Dy(1)-N(1)#1 | 109.5 |
| Cnt1-Dy(1)-Cnt2 | 127.0 |

| | |
|---|---|
| N(1)-Dy(1)-N(1)#1 | 30.66(16) |
| N(1)-Dy(1)-O(1) | 111.38(11) |
| N(1)#1-Dy(1)-O(1) | 81.05(11) |
| N(1)-Dy(1)-C(1) | 89.81(12) |
| N(1)#1-Dy(1)-C(1) | 113.62(12) |
| O(1)-Dy(1)-C(1) | 129.89(11) |
| N(1)-Dy(1)-C(5) | 120.68(13) |
| N(1)#1-Dy(1)-C(5) | 142.64(12) |
| O(1)-Dy(1)-C(5) | 112.56(12) |
| C(1)-Dy(1)-C(5) | 30.93(13) |
| N(1)-Dy(1)-C(14) | 84.00(11) |
| N(1)#1-Dy(1)-C(14) | 83.06(11) |
| O(1)-Dy(1)-C(14) | 97.19(12) |
| C(1)-Dy(1)-C(14) | 130.88(13) |
| C(5)-Dy(1)-C(14) | 126.48(13) |
| N(1)-Dy(1)-C(10) | 113.33(12) |
| N(1)#1-Dy(1)-C(10) | 103.87(12) |
| O(1)-Dy(1)-C(10) | 78.62(11) |
| C(1)-Dy(1)-C(10) | 134.92(13) |
| C(5)-Dy(1)-C(10) | 112.75(13) |
| C(14)-Dy(1)-C(10) | 30.60(12) |
| N(1)-Dy(1)-C(13) | 81.18(11) |
| N(1)#1-Dy(1)-C(13) | 95.83(12) |
| O(1)-Dy(1)-C(13) | 127.10(11) |
| C(1)-Dy(1)-C(13) | 99.93(13) |
| C(5)-Dy(1)-C(13) | 101.47(13) |
| C(14)-Dy(1)-C(13) | 30.97(12) |
| C(10)-Dy(1)-C(13) | 50.67(13) |
| N(1)-Dy(1)-C(11) | 130.55(12) |
| N(1)#1-Dy(1)-C(11) | 132.53(12) |
| O(1)-Dy(1)-C(11) | 94.46(12) |
| C(1)-Dy(1)-C(11) | 105.31(13) |
| C(5)-Dy(1)-C(11) | 82.69(13) |
| C(14)-Dy(1)-C(11) | 50.40(12) |
| C(10)-Dy(1)-C(11) | 30.47(12) |
| C(13)-Dy(1)-C(11) | 50.25(13) |

| | |
|---|---|
| N(1)-Dy(1)-C(2) | 80.42(11) |
| N(1)#1-Dy(1)-C(2) | 93.04(11) |
| O(1)-Dy(1)-C(2) | 106.50(11) |
| C(1)-Dy(1)-C(2) | 30.68(12) |
| C(5)-Dy(1)-C(2) | 50.22(13) |
| C(14)-Dy(1)-C(2) | 155.12(13) |
| C(10)-Dy(1)-C(2) | 162.96(13) |
| C(13)-Dy(1)-C(2) | 126.39(13) |
| C(11)-Dy(1)-C(2) | 132.69(13) |
| N(1)-Dy(1)-C(4) | 130.21(12) |
| N(1)#1-Dy(1)-C(4) | 132.78(12) |
| O(1)-Dy(1)-C(4) | 83.34(12) |
| C(1)-Dy(1)-C(4) | 50.74(13) |
| C(5)-Dy(1)-C(4) | 30.35(13) |
| C(14)-Dy(1)-C(4) | 143.22(13) |
| C(10)-Dy(1)-C(4) | 116.21(13) |
| C(13)-Dy(1)-C(4) | 128.44(13) |
| C(11)-Dy(1)-C(4) | 92.82(14) |
| C(2)-Dy(1)-C(4) | 49.96(13) |
| N(1)-Dy(1)-C(12) | 108.25(12) |
| N(1)#1-Dy(1)-C(12) | 125.96(12) |
| O(1)-Dy(1)-C(12) | 124.22(11) |
| C(1)-Dy(1)-C(12) | 86.90(13) |
| C(5)-Dy(1)-C(12) | 76.43(13) |
| C(14)-Dy(1)-C(12) | 50.15(12) |
| C(10)-Dy(1)-C(12) | 49.97(12) |
| C(13)-Dy(1)-C(12) | 30.14(12) |
| C(11)-Dy(1)-C(12) | 30.08(13) |
| C(2)-Dy(1)-C(12) | 117.57(12) |
| C(4)-Dy(1)-C(12) | 99.51(13) |
| N(1)-Dy(1)-C(3) | 103.33(11) |
| N(1)#1-Dy(1)-C(3) | 103.14(12) |
| O(1)-Dy(1)-C(3) | 80.14(11) |
| C(1)-Dy(1)-C(3) | 50.34(13) |
| C(5)-Dy(1)-C(3) | 49.78(13) |
| C(14)-Dy(1)-C(3) | 172.67(12) |

| | |
|---|---|
| C(10)-Dy(1)-C(3) | 142.28(13) |
| C(13)-Dy(1)-C(3) | 149.30(13) |
| C(11)-Dy(1)-C(3) | 122.75(12) |
| C(2)-Dy(1)-C(3) | 30.06(12) |
| C(4)-Dy(1)-C(3) | 30.01(12) |
| C(12)-Dy(1)-C(3) | 126.04(12) |
| C(19)-O(1)-C(22) | 108.3(3) |
| C(19)-O(1)-Dy(1) | 124.0(3) |
| C(22)-O(1)-Dy(1) | 127.7(2) |
| N(1)#1-N(1)-Dy(1) | 76.0(3) |
| N(1)#1-N(1)-Dy(1)#1 | 73.3(3) |
| Dy(1)-N(1)-Dy(1)#1 | 149.15(16) |
| C(5)-C(1)-C(2) | 106.9(4) |
| C(5)-C(1)-C(6) | 127.1(4) |
| C(2)-C(1)-C(6) | 125.8(4) |
| C(5)-C(1)-Dy(1) | 74.6(2) |
| C(2)-C(1)-Dy(1) | 77.0(2) |
| C(6)-C(1)-Dy(1) | 118.2(3) |
| C(3)-C(2)-C(1) | 108.4(4) |
| C(3)-C(2)-C(7) | 127.3(4) |
| C(1)-C(2)-C(7) | 124.0(4) |
| C(3)-C(2)-Dy(1) | 76.8(2) |
| C(1)-C(2)-Dy(1) | 72.4(2) |
| C(7)-C(2)-Dy(1) | 121.9(3) |
| C(2)-C(3)-C(4) | 107.9(4) |
| C(2)-C(3)-C(8) | 126.4(4) |
| C(4)-C(3)-C(8) | 125.3(4) |
| C(2)-C(3)-Dy(1) | 73.1(2) |
| C(4)-C(3)-Dy(1) | 73.6(2) |
| C(8)-C(3)-Dy(1) | 124.9(3) |
| C(5)-C(4)-C(3) | 107.4(4) |
| C(5)-C(4)-C(9) | 124.4(4) |
| C(3)-C(4)-C(9) | 127.0(4) |
| C(5)-C(4)-Dy(1) | 72.2(2) |
| C(3)-C(4)-Dy(1) | 76.4(2) |
| C(9)-C(4)-Dy(1) | 126.3(3) |

| | |
|---|---|
| C(4)-C(5)-C(1) | 109.3(4) |
| C(4)-C(5)-Dy(1) | 77.5(2) |
| C(1)-C(5)-Dy(1) | 74.4(2) |
| C(14)-C(10)-C(11) | 107.7(4) |
| C(14)-C(10)-C(15) | 125.1(4) |
| C(11)-C(10)-C(15) | 126.8(4) |
| C(14)-C(10)-Dy(1) | 73.5(2) |
| C(11)-C(10)-Dy(1) | 75.4(2) |
| C(15)-C(10)-Dy(1) | 122.7(3) |
| C(12)-C(11)-C(10) | 108.1(4) |
| C(12)-C(11)-C(16) | 124.7(4) |
| C(10)-C(11)-C(16) | 126.5(4) |
| C(12)-C(11)-Dy(1) | 75.9(2) |
| C(10)-C(11)-Dy(1) | 74.2(2) |
| C(16)-C(11)-Dy(1) | 123.5(3) |
| C(13)-C(12)-C(11) | 108.7(4) |
| C(13)-C(12)-C(17) | 125.6(4) |
| C(11)-C(12)-C(17) | 124.8(4) |
| C(13)-C(12)-Dy(1) | 73.4(2) |
| C(11)-C(12)-Dy(1) | 74.0(2) |
| C(17)-C(12)-Dy(1) | 127.2(3) |
| C(12)-C(13)-C(14) | 107.2(4) |
| C(12)-C(13)-C(18) | 126.7(4) |
| C(14)-C(13)-C(18) | 125.9(4) |
| C(12)-C(13)-Dy(1) | 76.4(2) |
| C(14)-C(13)-Dy(1) | 73.3(2) |
| C(18)-C(13)-Dy(1) | 119.7(3) |
| C(10)-C(14)-C(13) | 108.3(4) |
| C(10)-C(14)-Dy(1) | 75.9(2) |
| C(13)-C(14)-Dy(1) | 75.7(2) |
| O(1)-C(19)-C(20) | 107.4(4) |
| C(19)-C(20)-C(21) | 103.7(4) |
| C(22)-C(21)-C(20) | 102.0(4) |
| O(1)-C(22)-C(21) | 105.4(4) |

Symmetry transformations used to generate equivalent atoms:
#1 -x,y,-z+1/2

**Table S12** X-ray Data Collection Parameters of **3-Y** and **3-Dy**.

| | **3-Y** | **3-Dy** |
|---|---|---|
| formula | $C_{32}H_{44}N_2O_2Y_2$ | $C_{32}H_{44}N_2O_2Dy_2$ |
| fw | 666.51 | 813.69 |
| temp (K) | 88(2) | 133(2) |
| Wavelength | 0.71073 Å | 0.71073 Å |
| cryst syst | Monoclinic | Monoclinic |
| space group | $P2_1/n$ | $P2_1/n$ |
| a (Å) | 11.9100(9) | 11.9046(7) |
| b (Å) | 10.0376(7) | 10.0279(6) |
| c (Å) | 12.6988(9) | 12.7468(8) |
| Volume (Å$^3$) | 1510.39(19) | 1513.95(16) |
| α (deg) | 90 | 90 |
| β (deg) | 95.7802(9) | 95.7793(8) |
| γ (deg) | 90 | 90 |
| Z | 2 | 2 |
| $\rho_{calc}$ (Mg/m$^3$) | 1.466 | 1.785 |
| μ (mm$^{-1}$) | 3.853 | 4.927 |
| R1 (I > 2σ(I))[a] | 0.0193 | 0.0164 |
| wR2 (all data)[a] | 0.0453 | 0.0403 |

[a]Definitions: $wR2 = [\sum[w(F_o^2 - F_c^2)^2]/ \sum[w(F_o^2)^2] ]^{1/2}$; $R1 = \sum||F_o| - |F_c||/\sum|F_o|$.

**X-ray Data Collection, Structure Solution and Refinement for 3-Y.** A blue crystal of approximate dimensions 0.148 x 0.184 x 0.273 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[17] program package was used to determine the unit-cell parameters and for data collection (15 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT[8] and SADABS[18] to yield the reflection data file. Subsequent calculations were carried out using the SHELXTL[19] program. The diffraction symmetry was 2/*m* and the systematic absences were consistent with the monoclinic space group $P2_1/n$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were located from a difference-Fourier map and refined (x,y,z and $U_{iso}$). The molecule was located about an inversion center. At convergence, wR2 = 0.0453 and Goof = 1.034 for 260 variables refined against 3825 data (0.73Å), R1 = 0.0193 for those 3517 data with $I > 2.0\sigma(I)$.

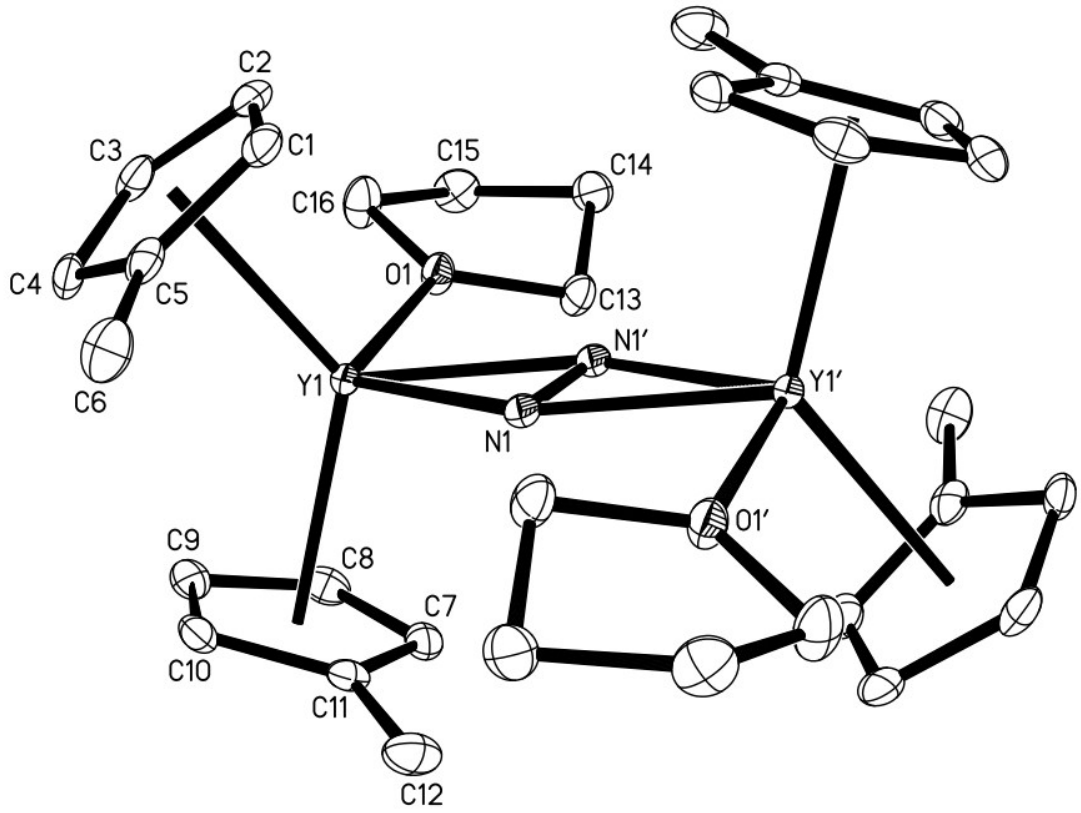


**Fig. S9** Thermal ellipsoid plot of $[(C_5H_4Me)_2(THF)Y]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **3-Y** drawn at the 50% probability level. Hydrogen atoms are omitted for clarity.

**Table S13** Bond lengths [Å] and angles [°] for **3-Y**.

| | |
|---|---|
| Y(1)-Cnt1 | 2.387 |
| Y(1)-Cnt2 | 2.381 |
| Y(1)-N(1) | 2.3023(11) |
| Y(1)-N(1)#1 | 2.3292(11) |
| Y(1)-O(1) | 2.4046(10) |
| Y(1)-C(10) | 2.6427(13) |
| Y(1)-C(9) | 2.6591(14) |
| Y(1)-C(3) | 2.6599(13) |
| Y(1)-C(2) | 2.6673(14) |
| Y(1)-C(11) | 2.6732(13) |

| | |
|---|---|
| Y(1)-C(4) | 2.6741(13) |
| Y(1)-C(1) | 2.6758(14) |
| Y(1)-C(7) | 2.6765(14) |
| Y(1)-C(8) | 2.6842(14) |
| O(1)-C(16) | 1.4556(17) |
| O(1)-C(13) | 1.4617(16) |
| N(1)-N(1)#1 | 1.250(2) |
| N(1)-Y(1)#1 | 2.3292(11) |
| C(1)-C(2) | 1.407(2) |
| C(1)-C(5) | 1.418(2) |
| C(2)-C(3) | 1.413(2) |
| C(3)-C(4) | 1.415(2) |
| C(4)-C(5) | 1.4135(19) |
| C(5)-C(6) | 1.504(2) |
| C(7)-C(11) | 1.4114(19) |
| C(7)-C(8) | 1.414(2) |
| C(8)-C(9) | 1.406(2) |
| C(9)-C(10) | 1.417(2) |
| C(10)-C(11) | 1.4114(19) |
| C(11)-C(12) | 1.5031(19) |
| C(13)-C(14) | 1.512(2) |
| C(14)-C(15) | 1.525(2) |
| C(15)-C(16) | 1.522(2) |

| | |
|---|---|
| Cnt1-Y(1)-N(1) | 104.0 |
| Cnt1-Y(1)-N(1') | 115.8 |
| Cnt1-Y(1)-O(1) | 103.0 |
| Cnt1-Y(1)-N(1) | 102.6 |
| Cnt1-Y(1)-N(1') | 109.4 |
| Cnt1-Y(1)-O(1) | 103.9 |
| Cnt1-Y(1)-Cnt2 | 129.1 |
| N(1)-Y(1)-N(1)#1 | 31.30(5) |
| N(1)-Y(1)-O(1) | 114.83(4) |
| N(1)#1-Y(1)-O(1) | 83.65(4) |
| N(1)-Y(1)-C(10) | 100.72(4) |
| N(1)#1-Y(1)-C(10) | 120.95(4) |

| | |
|---|---|
| O(1)-Y(1)-C(10) | 126.80(4) |
| N(1)-Y(1)-C(9) | 127.82(4) |
| N(1)#1-Y(1)-C(9) | 135.43(4) |
| O(1)-Y(1)-C(9) | 97.41(4) |
| C(10)-Y(1)-C(9) | 31.00(4) |
| N(1)-Y(1)-C(3) | 129.50(4) |
| N(1)#1-Y(1)-C(3) | 131.47(4) |
| O(1)-Y(1)-C(3) | 82.99(4) |
| C(10)-Y(1)-C(3) | 104.19(5) |
| C(9)-Y(1)-C(3) | 92.54(5) |
| N(1)-Y(1)-C(2) | 103.08(4) |
| N(1)#1-Y(1)-C(2) | 100.83(4) |
| O(1)-Y(1)-C(2) | 79.29(4) |
| C(10)-Y(1)-C(2) | 130.70(4) |
| C(9)-Y(1)-C(2) | 123.27(5) |
| C(3)-Y(1)-C(2) | 30.75(5) |
| N(1)-Y(1)-C(11) | 76.87(4) |
| N(1)#1-Y(1)-C(11) | 90.91(4) |
| O(1)-Y(1)-C(11) | 122.30(4) |
| C(10)-Y(1)-C(11) | 30.79(4) |
| C(9)-Y(1)-C(11) | 51.01(4) |
| C(3)-Y(1)-C(11) | 134.85(4) |
| C(2)-Y(1)-C(11) | 156.75(4) |
| N(1)-Y(1)-C(4) | 117.60(4) |
| N(1)#1-Y(1)-C(4) | 139.16(4) |
| O(1)-Y(1)-C(4) | 112.76(4) |
| C(10)-Y(1)-C(4) | 80.06(4) |
| C(9)-Y(1)-C(4) | 81.40(4) |
| C(3)-Y(1)-C(4) | 30.76(4) |
| C(2)-Y(1)-C(4) | 50.66(4) |
| C(11)-Y(1)-C(4) | 108.23(4) |
| N(1)-Y(1)-C(1) | 78.99(4) |
| N(1)#1-Y(1)-C(1) | 89.81(4) |
| O(1)-Y(1)-C(1) | 106.30(4) |
| C(10)-Y(1)-C(1) | 118.83(5) |
| C(9)-Y(1)-C(1) | 131.25(4) |

| | |
|---|---|
| C(3)-Y(1)-C(1) | 50.52(5) |
| C(2)-Y(1)-C(1) | 30.53(5) |
| C(11)-Y(1)-C(1) | 131.16(5) |
| C(4)-Y(1)-C(1) | 50.31(4) |
| N(1)-Y(1)-C(7) | 86.85(4) |
| N(1)#1-Y(1)-C(7) | 84.84(4) |
| O(1)-Y(1)-C(7) | 91.80(4) |
| C(10)-Y(1)-C(7) | 50.58(4) |
| C(9)-Y(1)-C(7) | 50.61(4) |
| C(3)-Y(1)-C(7) | 141.93(4) |
| C(2)-Y(1)-C(7) | 168.74(5) |
| C(11)-Y(1)-C(7) | 30.59(4) |
| C(4)-Y(1)-C(7) | 129.12(4) |
| C(1)-Y(1)-C(7) | 160.46(5) |
| N(1)-Y(1)-C(8) | 117.37(4) |
| N(1)#1-Y(1)-C(8) | 109.70(4) |
| O(1)-Y(1)-C(8) | 77.49(4) |
| C(10)-Y(1)-C(8) | 50.67(4) |
| C(9)-Y(1)-C(8) | 30.52(4) |
| C(3)-Y(1)-C(8) | 112.45(5) |
| C(2)-Y(1)-C(8) | 138.89(5) |
| C(11)-Y(1)-C(8) | 50.70(4) |
| C(4)-Y(1)-C(8) | 110.31(4) |
| C(1)-Y(1)-C(8) | 160.48(4) |
| C(7)-Y(1)-C(8) | 30.59(4) |
| C(16)-O(1)-C(13) | 109.29(10) |
| C(16)-O(1)-Y(1) | 127.91(8) |
| C(13)-O(1)-Y(1) | 122.44(8) |
| N(1)#1-N(1)-Y(1) | 75.54(10) |
| N(1)#1-N(1)-Y(1)#1 | 73.17(10) |
| Y(1)-N(1)-Y(1)#1 | 148.70(5) |
| C(2)-C(1)-C(5) | 108.97(13) |
| C(2)-C(1)-Y(1) | 74.40(8) |
| C(5)-C(1)-Y(1) | 75.09(8) |
| C(1)-C(2)-C(3) | 107.70(13) |
| C(1)-C(2)-Y(1) | 75.06(8) |

| | |
|---|---|
| C(3)-C(2)-Y(1) | 74.33(8) |
| C(2)-C(3)-C(4) | 107.84(13) |
| C(2)-C(3)-Y(1) | 74.91(8) |
| C(4)-C(3)-Y(1) | 75.17(8) |
| C(5)-C(4)-C(3) | 108.65(13) |
| C(5)-C(4)-Y(1) | 75.20(8) |
| C(3)-C(4)-Y(1) | 74.06(8) |
| C(4)-C(5)-C(1) | 106.84(13) |
| C(4)-C(5)-C(6) | 127.20(13) |
| C(1)-C(5)-C(6) | 125.93(13) |
| C(4)-C(5)-Y(1) | 74.22(8) |
| C(1)-C(5)-Y(1) | 74.24(8) |
| C(6)-C(5)-Y(1) | 118.74(9) |
| C(11)-C(7)-C(8) | 108.55(12) |
| C(11)-C(7)-Y(1) | 74.58(8) |
| C(8)-C(7)-Y(1) | 75.00(8) |
| C(9)-C(8)-C(7) | 107.93(13) |
| C(9)-C(8)-Y(1) | 73.76(8) |
| C(7)-C(8)-Y(1) | 74.40(8) |
| C(8)-C(9)-C(10) | 107.71(12) |
| C(8)-C(9)-Y(1) | 75.73(8) |
| C(10)-C(9)-Y(1) | 73.86(8) |
| C(11)-C(10)-C(9) | 108.55(12) |
| C(11)-C(10)-Y(1) | 75.80(8) |
| C(9)-C(10)-Y(1) | 75.14(8) |
| C(7)-C(11)-C(10) | 107.25(12) |
| C(7)-C(11)-C(12) | 127.14(13) |
| C(10)-C(11)-C(12) | 125.53(13) |
| C(7)-C(11)-Y(1) | 74.83(8) |
| C(10)-C(11)-Y(1) | 73.41(8) |
| C(12)-C(11)-Y(1) | 119.83(9) |
| O(1)-C(13)-C(14) | 104.31(11) |
| C(13)-C(14)-C(15) | 102.43(12) |
| C(16)-C(15)-C(14) | 102.18(12) |
| O(1)-C(16)-C(15) | 106.22(11) |

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y+2,-z+1

**X-ray Data Collection, Structure Solution and Refinement for 3-Dy.** A green crystal of approximate dimensions 0.169 x 0.199 x 0.265 mm was mounted on a glass fiber and transferred to a Bruker SMART APEX II diffractometer. The APEX2[20] program package and the CELL_NOW[15] were used to determine the unit-cell parameters. Data was collected using a 10 sec/frame scan time for a sphere of diffraction data. The raw frame data was processed using SAINT[8] and TWINABS[16] to yield the reflection data file (HKLF 5 format)[16]. Subsequent calculations were carried out using the SHELXTL[5] program. The diffraction symmetry was 2/*m* and the systematic absences were consistent with the monoclinic space group $P2_1/n$ that was later determined to be correct. The structure was solved by direct methods and refined on $F^2$ by full-matrix least-squares techniques. The analytical scattering factors[6] for neutral atoms were used throughout the analysis. Hydrogen atoms were included using a riding model. The molecule was located about an inversion center (Z = 2). At convergence, wR2 = 0.0403 and Goof = 1.036 for 175 variables refined against 3608 data (0.74Å), R1 = 0.0164 for those 3406 with $I > 2.0\sigma(I)$. The structure was refined as a two-component twin, BASF[5] = 0.4066.

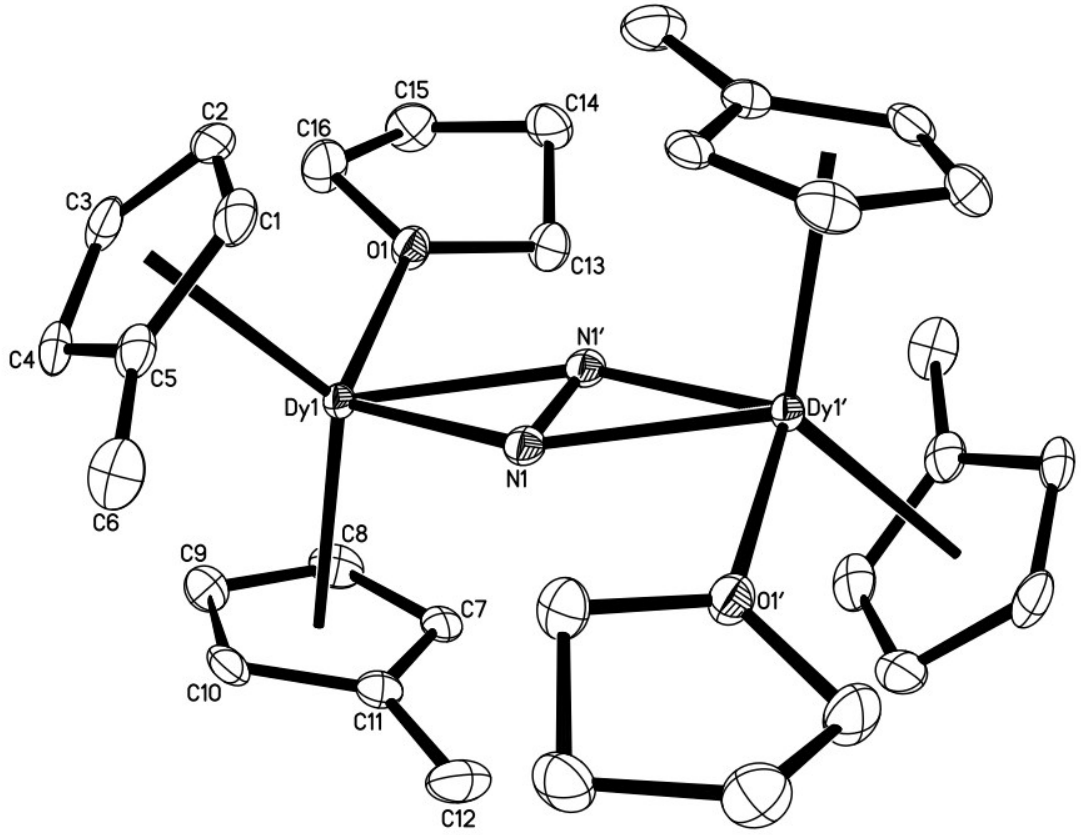


**Fig. S10** Thermal ellipsoid plot of $[(C_5H_4Me)_2(THF)Dy]_2(\mu\text{-}\eta^2\text{:}\eta^2\text{-}N_2)$, **3-Dy** drawn at the 50% probability level. Hydrogen atoms are omitted for clarity.

**Table S14** Bond lengths [Å] and angles [°] for **3-Dy**.

| | |
|---|---|
| Dy(1)-Cnt1 | 2.391 |
| Dy(1)-Cnt2 | 2.385 |
| Dy(1)-N(1) | 2.296(2) |
| Dy(1)-N(1)#1 | 2.334(2) |
| Dy(1)-O(1) | 2.4156(17) |
| Dy(1)-C(10) | 2.645(2) |
| Dy(1)-C(9) | 2.661(3) |
| Dy(1)-C(3) | 2.662(3) |
| Dy(1)-C(2) | 2.673(3) |
| Dy(1)-C(4) | 2.674(2) |
| Dy(1)-C(7) | 2.677(2) |

| | |
|---|---|
| Dy(1)-C(11) | 2.677(2) |
| Dy(1)-C(1) | 2.681(3) |
| Dy(1)-C(8) | 2.686(3) |
| O(1)-C(16) | 1.451(3) |
| O(1)-C(13) | 1.460(3) |
| N(1)-N(1)#1 | 1.250(4) |
| N(1)-Dy(1)#1 | 2.334(2) |
| C(1)-C(2) | 1.395(4) |
| C(1)-C(5) | 1.420(4) |
| C(2)-C(3) | 1.419(4) |
| C(3)-C(4) | 1.412(4) |
| C(4)-C(5) | 1.415(4) |
| C(5)-C(6) | 1.500(4) |
| C(7)-C(8) | 1.407(4) |
| C(7)-C(11) | 1.411(4) |
| C(8)-C(9) | 1.404(4) |
| C(9)-C(10) | 1.419(4) |
| C(10)-C(11) | 1.405(4) |
| C(11)-C(12) | 1.497(4) |
| C(13)-C(14) | 1.510(4) |
| C(14)-C(15) | 1.516(4) |
| C(15)-C(16) | 1.520(4) |

| | |
|---|---|
| Cnt1-Dy(1)-N(1) | 104.0 |
| Cnt1-Dy(1)-N(1)#1 | 116.0 |
| Cnt1-Dy(1)-O(1) | 103.1 |
| Cnt2-Dy(1)-N(1) | 102.6 |
| Cnt2-Dy(1)-N(1)#1 | 109.3 |
| Cnt2-Dy(1)-O(1) | 103.9 |
| Cnt1-Dy(1)-Cnt2 | 129.1 |
| N(1)-Dy(1)-N(1)#1 | 31.32(9) |
| N(1)-Dy(1)-O(1) | 114.57(6) |
| N(1)#1-Dy(1)-O(1) | 83.38(6) |
| N(1)-Dy(1)-C(10) | 100.44(8) |
| N(1)#1-Dy(1)-C(10) | 120.54(8) |
| O(1)-Dy(1)-C(10) | 127.02(8) |

| | |
|---|---|
| N(1)-Dy(1)-C(9) | 127.68(8) |
| N(1)#1-Dy(1)-C(9) | 135.25(8) |
| O(1)-Dy(1)-C(9) | 97.75(8) |
| C(10)-Dy(1)-C(9) | 31.02(8) |
| N(1)-Dy(1)-C(3) | 129.47(8) |
| N(1)#1-Dy(1)-C(3) | 131.60(8) |
| O(1)-Dy(1)-C(3) | 83.21(7) |
| C(10)-Dy(1)-C(3) | 104.47(9) |
| C(9)-Dy(1)-C(3) | 92.61(9) |
| N(1)-Dy(1)-C(2) | 102.86(9) |
| N(1)#1-Dy(1)-C(2) | 100.88(8) |
| O(1)-Dy(1)-C(2) | 79.62(8) |
| C(10)-Dy(1)-C(2) | 130.87(8) |
| C(9)-Dy(1)-C(2) | 123.45(9) |
| C(3)-Dy(1)-C(2) | 30.85(9) |
| N(1)-Dy(1)-C(4) | 117.74(8) |
| N(1)#1-Dy(1)-C(4) | 139.43(8) |
| O(1)-Dy(1)-C(4) | 112.89(7) |
| C(10)-Dy(1)-C(4) | 80.24(8) |
| C(9)-Dy(1)-C(4) | 81.27(8) |
| C(3)-Dy(1)-C(4) | 30.69(8) |
| C(2)-Dy(1)-C(4) | 50.63(8) |
| N(1)-Dy(1)-C(7) | 87.17(8) |
| N(1)#1-Dy(1)-C(7) | 84.93(8) |
| O(1)-Dy(1)-C(7) | 91.55(7) |
| C(10)-Dy(1)-C(7) | 50.38(8) |
| C(9)-Dy(1)-C(7) | 50.36(8) |
| C(3)-Dy(1)-C(7) | 141.66(9) |
| C(2)-Dy(1)-C(7) | 168.70(9) |
| C(4)-Dy(1)-C(7) | 128.93(8) |
| N(1)-Dy(1)-C(11) | 76.90(8) |
| N(1)#1-Dy(1)-C(11) | 90.71(8) |
| O(1)-Dy(1)-C(11) | 122.06(7) |
| C(10)-Dy(1)-C(11) | 30.61(8) |
| C(9)-Dy(1)-C(11) | 50.86(8) |
| C(3)-Dy(1)-C(11) | 134.98(8) |

| | |
|---|---|
| C(2)-Dy(1)-C(11) | 156.76(8) |
| C(4)-Dy(1)-C(11) | 108.39(8) |
| C(7)-Dy(1)-C(11) | 30.57(8) |
| N(1)-Dy(1)-C(1) | 79.15(8) |
| N(1)#1-Dy(1)-C(1) | 90.17(8) |
| O(1)-Dy(1)-C(1) | 106.38(8) |
| C(10)-Dy(1)-C(1) | 118.76(9) |
| C(9)-Dy(1)-C(1) | 130.99(8) |
| C(3)-Dy(1)-C(1) | 50.33(9) |
| C(2)-Dy(1)-C(1) | 30.21(9) |
| C(4)-Dy(1)-C(1) | 50.21(8) |
| C(7)-Dy(1)-C(1) | 160.75(9) |
| C(11)-Dy(1)-C(1) | 131.31(9) |
| N(1)-Dy(1)-C(8) | 117.53(8) |
| N(1)#1-Dy(1)-C(8) | 109.73(8) |
| O(1)-Dy(1)-C(8) | 77.60(7) |
| C(10)-Dy(1)-C(8) | 50.63(8) |
| C(9)-Dy(1)-C(8) | 30.44(8) |
| C(3)-Dy(1)-C(8) | 112.31(9) |
| C(2)-Dy(1)-C(8) | 139.00(10) |
| C(4)-Dy(1)-C(8) | 110.01(8) |
| C(7)-Dy(1)-C(8) | 30.42(8) |
| C(11)-Dy(1)-C(8) | 50.61(8) |
| C(1)-Dy(1)-C(8) | 160.09(9) |
| C(16)-O(1)-C(13) | 109.31(18) |
| C(16)-O(1)-Dy(1) | 127.92(14) |
| C(13)-O(1)-Dy(1) | 122.40(14) |
| N(1)#1-N(1)-Dy(1) | 76.03(18) |
| N(1)#1-N(1)-Dy(1)#1 | 72.65(17) |
| Dy(1)-N(1)-Dy(1)#1 | 148.68(9) |
| C(2)-C(1)-C(5) | 109.4(3) |
| C(2)-C(1)-Dy(1) | 74.56(15) |
| C(5)-C(1)-Dy(1) | 74.90(15) |
| C(1)-C(2)-C(3) | 107.7(2) |
| C(1)-C(2)-Dy(1) | 75.23(15) |
| C(3)-C(2)-Dy(1) | 74.15(15) |

| | |
|---|---|
| C(4)-C(3)-C(2) | 107.7(3) |
| C(4)-C(3)-Dy(1) | 75.13(14) |
| C(2)-C(3)-Dy(1) | 75.00(15) |
| C(3)-C(4)-C(5) | 108.7(2) |
| C(3)-C(4)-Dy(1) | 74.18(14) |
| C(5)-C(4)-Dy(1) | 75.21(14) |
| C(4)-C(5)-C(1) | 106.6(2) |
| C(4)-C(5)-C(6) | 127.2(2) |
| C(1)-C(5)-C(6) | 126.2(3) |
| C(4)-C(5)-Dy(1) | 74.17(14) |
| C(1)-C(5)-Dy(1) | 74.43(15) |
| C(6)-C(5)-Dy(1) | 118.91(17) |
| C(8)-C(7)-C(11) | 108.9(2) |
| C(8)-C(7)-Dy(1) | 75.15(15) |
| C(11)-C(7)-Dy(1) | 74.72(14) |
| C(9)-C(8)-C(7) | 107.8(2) |
| C(9)-C(8)-Dy(1) | 73.77(15) |
| C(7)-C(8)-Dy(1) | 74.43(15) |
| C(8)-C(9)-C(10) | 107.7(2) |
| C(8)-C(9)-Dy(1) | 75.78(15) |
| C(10)-C(9)-Dy(1) | 73.86(15) |
| C(11)-C(10)-C(9) | 108.5(2) |
| C(11)-C(10)-Dy(1) | 75.97(14) |
| C(9)-C(10)-Dy(1) | 75.11(14) |
| C(10)-C(11)-C(7) | 107.1(2) |
| C(10)-C(11)-C(12) | 126.1(2) |
| C(7)-C(11)-C(12) | 126.8(2) |
| C(10)-C(11)-Dy(1) | 73.42(14) |
| C(7)-C(11)-Dy(1) | 74.71(14) |
| C(12)-C(11)-Dy(1) | 120.03(17) |
| O(1)-C(13)-C(14) | 104.2(2) |
| C(13)-C(14)-C(15) | 102.7(2) |
| C(14)-C(15)-C(16) | 102.1(2) |
| O(1)-C(16)-C(15) | 106.2(2) |

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Symmetry transformations used to generate equivalent atoms:

#1 -x+1,-y+1,-z+1

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Definitions:

$wR2 = [\Sigma[w(F_o^2-F_c^2)^2] / \Sigma[w(F_o^2)^2] ]^{1/2}$

$R1 = \Sigma||F_o|-|F_c|| / \Sigma|F_o|$

$Goof = S = [\Sigma[w(F_o^2-F_c^2)^2] / (n-p)]^{1/2}$ where n is the number of reflections and p is the total number of parameters refined.