

Supplementary Information

Magnetization dynamics in a Dy based 2D coordination network bridged by a nitronyl nitroxide radical with a CN group

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Table S1. Selected bond distances (Å) and angles (°) for complex 1.

Bond distances			
Gd(1)-O(24)	2.342(7)	Gd(2)-O(6)	2.376(7)
Gd(1)-O(25)	2.325(7)	Gd(2)-O(20)	2.353(8)
Gd(1)-O(30)	2.335(7)	Gd(2)-O(21)	2.330(7)
Gd(1)-O(34)#3	2.363(7)	Gd(2)-O(28)	2.325(6)
Gd(1)-O(38)	2.335(8)	Gd(2)-O(35)	2.332(8)
Gd(1)-O(39)	2.320(8)	Gd(2)-O(36)	2.346(8)
Gd(1)-O(40)	2.343(8)	Gd(2)-O(42)	2.331(7)
Gd(1)-N(12)	2.541(10)	Gd(2)-N(9)#1	2.511(9)
Gd(3)-O(2)	2.363(7)	Gd(4)-O(1)	2.363(6)
Gd(3)-O(4)	2.308(6)	Gd(4)-O(11)	2.320(7)
Gd(3)-O(7)	2.349(6)	Gd(4)-O(12)	2.333(7)
Gd(3)-O(9)	2.345(7)	Gd(4)-O(14)	2.338(7)
Gd(3)-O(13)	2.337(7)	Gd(4)-O(19)	2.336(7)
Gd(3)-O(16)	2.324(7)	Gd(4)-O(26)	2.337(7)
Gd(3)-O(22)	2.341(7)	Gd(4)-O(41)	2.295(7)
Gd(3)-O(23)	2.363(6)	Gd(4)-N(8)	2.507(8)
Gd(5)-O(5)	2.341(7)	Gd(6)-O(3)	2.322(6)
Gd(5)-O(10)#2	2.353(6)	Gd(6)-O(8)	2.331(7)
Gd(5)-O(27)	2.341(7)	Gd(6)-O(15)	2.332(6)
Gd(5)-O(29)	2.328(7)	Gd(6)-O(17)	2.346(7)
Gd(5)-O(31)	2.330(7)	Gd(6)-O(18)	2.351(7)
Gd(5)-O(32)	2.315(7)	Gd(6)-O(1A)	2.341(6)
Gd(5)-O(33)	2.330(7)	Gd(6)-O(37)	2.351(7)
Gd(5)-N(10)	2.483(9)	Gd(6)-O(43)	2.323(7)
O(1)-N(3)	1.306(10)	O(6)-N(1)	1.292(10)
O(7)-N(2)	1.279(9)	O(10)-N(6)	1.313(10)
O(17)-N(4)	1.311(10)	O(1A)-N(7)	1.319(12)
O(23)-N(11)	1.283(10)	O(34)-N(5)	1.281(9)
Angles			
O(24)-Gd(1)-O(25)	73.4(3)	O(6)-Gd(2)-N(9)#1	140.7(3)
O(24)-Gd(1)-O(30)	141.1(2)	O(20)-Gd(2)-O(6)	71.7(3)
O(24)-Gd(1)-O(34)#3	73.1(3)	O(20)-Gd(2)-N(9)#1	72.4(3)
O(24)-Gd(1)-O(40)	79.7(3)	O(21)-Gd(2)-O(6)	104.3(3)
O(24)-Gd(1)-N(12)	143.0(3)	O(21)-Gd(2)-O(20)	75.6(3)
O(25)-Gd(1)-O(30)	143.4(3)	O(21)-Gd(2)-O(35)	74.5(3)
O(25)-Gd(1)-O(34)#3	107.9(3)	O(21)-Gd(2)-O(36)	138.4(3)
O(25)-Gd(1)-O(38)	75.6(3)	O(21)-Gd(2)-O(42)	73.8(3)
O(25)-Gd(1)-O(40)	74.6(3)	O(21)-Gd(2)-N(9)#1	81.5(3)
O(25)-Gd(1)-N(12)	79.7(3)	O(28)-Gd(2)-O(6)	81.8(2)
O(30)-Gd(1)-O(34)#3	80.6(3)	O(28)-Gd(2)-O(20)	74.0(3)
O(30)-Gd(1)-O(38)	73.2(3)	O(28)-Gd(2)-O(21)	145.2(3)

O(30)-Gd(1)-O(40)	115.6(3)	O(28)-Gd(2)-O(35)	116.0(3)
O(30)-Gd(1)-N(12)	72.9(3)	O(28)-Gd(2)-O(36)	73.6(3)
O(34)#3-Gd(1)-N(12)	140.8(3)	O(28)-Gd(2)-O(42)	139.1(2)
O(38)-Gd(1)-O(24)	123.1(3)	O(28)-Gd(2)-N(9)#1	73.5(3)
O(38)-Gd(1)-O(34)#3	72.8(3)	O(35)-Gd(2)-O(6)	151.2(3)
O(38)-Gd(1)-O(40)	134.2(3)	O(35)-Gd(2)-O(20)	133.0(3)
O(38)-Gd(1)-N(12)	72.2(3)	O(35)-Gd(2)-O(36)	72.1(3)
O(39)-Gd(1)-O(24)	76.2(3)	O(35)-Gd(2)-N(9)#1	68.0(3)
O(39)-Gd(1)-O(25)	138.4(3)	O(36)-Gd(2)-O(6)	93.6(3)
O(39)-Gd(1)-O(30)	75.3(3)	O(36)-Gd(2)-O(20)	146.0(3)
O(39)-Gd(1)-O(34)#3	89.2(3)	O(36)-Gd(2)-N(9)#1	107.6(3)
O(39)-Gd(1)-O(38)	145.8(3)	O(42)-Gd(2)-O(6)	73.4(2)
O(39)-Gd(1)-O(40)	72.6(3)	O(42)-Gd(2)-O(20)	125.3(3)
O(39)-Gd(1)-N(12)	110.7(3)	O(42)-Gd(2)-O(35)	78.8(3)
O(40)-Gd(1)-O(34)#3	150.2(3)	O(42)-Gd(2)-O(36)	76.0(3)
O(40)-Gd(1)-N(12)	68.9(3)	O(42)-Gd(2)-N(9)#1	142.9(3)
O(2)-Gd(3)-O(23)	72.3(2)	O(1)-Gd(4)-N(8)	143.0(3)
O(4)-Gd(3)-O(2)	73.5(3)	O(11)-Gd(4)-O(1)	107.3(2)
O(4)-Gd(3)-O(7)	90.7(2)	O(11)-Gd(4)-O(12)	74.8(3)
O(4)-Gd(3)-O(9)	75.7(3)	O(11)-Gd(4)-O(14)	143.5(2)
O(4)-Gd(3)-O(13)	73.3(2)	O(11)-Gd(4)-O(19)	75.8(3)
O(4)-Gd(3)-O(16)	139.6(2)	O(11)-Gd(4)-O(26)	73.7(3)
O(4)-Gd(3)-O(22)	146.1(3)	O(11)-Gd(4)-N(8)	79.7(3)
O(4)-Gd(3)-O(23)	104.9(2)	O(12)-Gd(4)-O(1)	148.2(3)
O(7)-Gd(3)-O(2)	73.1(2)	O(12)-Gd(4)-O(14)	116.8(3)
O(7)-Gd(3)-O(23)	135.9(2)	O(12)-Gd(4)-O(19)	135.7(2)
O(9)-Gd(3)-O(2)	132.9(2)	O(12)-Gd(4)-O(26)	76.9(3)
O(9)-Gd(3)-O(7)	72.4(2)	O(12)-Gd(4)-N(8)	68.8(3)
O(9)-Gd(3)-O(23)	151.1(3)	O(14)-Gd(4)-O(1)	81.0(2)
O(13)-Gd(3)-O(2)	122.7(3)	O(14)-Gd(4)-N(8)	73.9(3)
O(13)-Gd(3)-O(7)	150.8(2)	O(19)-Gd(4)-O(1)	72.9(2)
O(13)-Gd(3)-O(9)	79.9(3)	O(19)-Gd(4)-O(14)	72.9(2)
O(13)-Gd(3)-O(22)	133.4(2)	O(19)-Gd(4)-O(26)	124.5(3)
O(13)-Gd(3)-O(23)	72.9(3)	O(19)-Gd(4)-N(8)	73.9(3)
O(16)-Gd(3)-O(2)	146.7(2)	O(26)-Gd(4)-O(1)	73.5(2)
O(16)-Gd(3)-O(7)	103.4(2)	O(26)-Gd(4)-O(14)	141.0(2)
O(16)-Gd(3)-O(9)	73.0(2)	O(26)-Gd(4)-N(8)	141.0(3)
O(16)-Gd(3)-O(13)	76.6(3)	O(41)-Gd(4)-O(1)	89.5(3)
O(16)-Gd(3)-O(22)	73.9(3)	O(41)-Gd(4)-O(11)	140.8(3)
O(16)-Gd(3)-O(23)	91.0(2)	O(41)-Gd(4)-O(12)	72.9(3)
O(22)-Gd(3)-O(2)	73.5(2)	O(41)-Gd(4)-O(14)	72.7(3)
O(22)-Gd(3)-O(7)	72.1(2)	O(41)-Gd(4)-O(19)	143.4(3)
O(22)-Gd(3)-O(9)	123.2(2)	O(41)-Gd(4)-O(26)	77.9(3)
O(22)-Gd(3)-O(23)	72.4(2)	O(41)-Gd(4)-N(8)	108.0(3)

O(5)-Gd(5)-O(10)#2	78.7(2)	O(3)-Gd(6)-O(8)	139.7(2)
O(5)-Gd(5)-N(10)	74.2(3)	O(3)-Gd(6)-O(15)	77.6(2)
O(10)#2-Gd(5)-N(10)	141.5(3)	O(3)-Gd(6)-O(17)	89.9(2)
O(27)-Gd(5)-O(5)	120.3(3)	O(3)-Gd(6)-O(18)	72.9(2)
O(27)-Gd(5)-O(10)#2	149.1(3)	O(3)-Gd(6)-O(1A)	103.3(2)
O(27)-Gd(5)-N(10)	69.4(3)	O(3)-Gd(6)-O(37)	76.6(3)
O(29)-Gd(5)-O(5)	146.0(3)	O(3)-Gd(6)-O(43)	149.3(2)
O(29)-Gd(5)-O(10)#2	106.0(2)	O(8)-Gd(6)-O(15)	72.7(2)
O(29)-Gd(5)-O(27)	72.7(3)	O(8)-Gd(6)-O(17)	106.5(2)
O(29)-Gd(5)-O(31)	75.9(3)	O(8)-Gd(6)-O(18)	75.6(3)
O(29)-Gd(5)-O(33)	73.8(3)	O(8)-Gd(6)-O(1A)	90.1(3)
O(29)-Gd(5)-N(10)	83.3(3)	O(8)-Gd(6)-O(37)	143.3(3)
O(31)-Gd(5)-O(5)	73.5(2)	O(15)-Gd(6)-O(17)	73.0(2)
O(31)-Gd(5)-O(10)#2	72.5(3)	O(15)-Gd(6)-O(18)	80.0(3)
O(31)-Gd(5)-O(27)	133.7(3)	O(15)-Gd(6)-O(1A)	150.4(3)
O(31)-Gd(5)-O(33)	125.3(2)	O(15)-Gd(6)-O(37)	135.8(3)
O(31)-Gd(5)-N(10)	73.9(3)	O(17)-Gd(6)-O(18)	150.5(3)
O(32)-Gd(5)-O(5)	72.1(3)	O(17)-Gd(6)-O(37)	71.7(2)
O(32)-Gd(5)-O(10)#2	93.9(3)	O(18)-Gd(6)-O(37)	124.4(3)
O(32)-Gd(5)-O(27)	72.1(3)	O(1A)-Gd(6)-O(17)	136.2(3)
O(32)-Gd(5)-O(29)	139.1(3)	O(1A)-Gd(6)-O(18)	72.3(3)
O(32)-Gd(5)-O(31)	144.9(3)	O(1A)-Gd(6)-O(37)	71.1(3)
O(32)-Gd(5)-O(33)	78.4(3)	O(43)-Gd(6)-O(8)	70.9(3)
O(32)-Gd(5)-N(10)	103.0(3)	O(43)-Gd(6)-O(15)	120.4(3)
O(33)-Gd(5)-O(5)	137.5(2)	O(43)-Gd(6)-O(17)	74.3(2)
O(33)-Gd(5)-O(10)#2	73.5(3)	O(43)-Gd(6)-O(18)	131.4(3)
O(33)-Gd(5)-O(27)	76.6(3)	O(43)-Gd(6)-O(1A)	73.6(3)
O(33)-Gd(5)-N(10)	143.4(3)	O(43)-Gd(6)-O(37)	73.5(3)
N(3)-O(1)-Gd(4)	136.6(6)	N(1)-O(6)-Gd(2)	136.0(6)
N(2)-O(7)-Gd(3)	142.4(6)	N(6)-O(10)-Gd(5)#4	137.3(6)
N(4)-O(17)-Gd(6)	143.0(6)	N(7)-O(1A)-Gd(6)	142.9(7)
N(11)-O(23)-Gd(3)	140.6(6)	N(5)-O(34)-Gd(1)#5	139.8(6)

Symmetry transformations used to generate equivalent atoms:

#1 x-1, y, z+1; #2 x-1, y, z; #3 x, y, z+1; #4 x+1, y, z; #5 x, y, z-1

Table S2. Selected bond distances (Å) and angles (°) for complex 2.

Bond distances			
Tb(1)-O(24)	2.369(9)	Tb(2)-O(6)	2.388(9)
Tb(1)-O(25)	2.359(10)	Tb(2)-O(20)	2.364(9)
Tb(1)-O(30)	2.373(9)	Tb(2)-O(21)	2.338(9)
Tb(1)-O(34)#3	2.381(9)	Tb(2)-O(28)	2.356(9)
Tb(1)-O(38)	2.357(10)	Tb(2)-O(35)	2.362(11)
Tb(1)-O(39)	2.351(12)	Tb(2)-O(36)	2.370(10)
Tb(1)-O(40)	2.372(11)	Tb(2)-O(42)	2.370(9)
Tb(1)-N(12)	2.550(12)	Tb(2)-N(9)#1	2.540(11)
Tb(3)-O(2)	2.375(10)	Tb(4)-O(1)	2.381(8)
Tb(3)-O(4)	2.355(8)	Tb(4)-O(11)	2.334(9)
Tb(3)-O(7)	2.353(8)	Tb(4)-O(12)	2.350(9)
Tb(3)-O(9)	2.375(8)	Tb(4)-O(14)	2.361(9)
Tb(3)-O(13)	2.357(11)	Tb(4)-O(19)	2.355(9)
Tb(3)-O(16)	2.356(9)	Tb(4)-O(26)	2.360(9)
Tb(3)-O(22)	2.373(9)	Tb(4)-O(41)	2.323(9)
Tb(3)-O(23)	2.384(8)	Tb(4)-N(8)	2.541(12)
Tb(5)-O(5)	2.372(10)	Tb(6)-O(3)	2.355(8)
Tb(5)-O(10)#2	2.380(8)	Tb(6)-O(8)	2.358(9)
Tb(5)-O(27)	2.367(10)	Tb(6)-O(15)	2.374(9)
Tb(5)-O(29)	2.357(9)	Tb(6)-O(17)	2.365(8)
Tb(5)-O(31)	2.350(9)	Tb(6)-O(18)	2.360(10)
Tb(5)-O(32)	2.343(9)	Tb(6)-O(1A)	2.368(8)
Tb(5)-O(33)	2.351(10)	Tb(6)-O(37)	2.372(9)
Tb(5)-N(10)	2.526(13)	Tb(6)-O(43)	2.357(8)
O(1)-N(3)	1.313(13)	O(6)-N(1)	1.326(12)
O(7)-N(2)	1.323(12)	O(10)-N(6)	1.284(13)
O(17)-N(4)	1.279(13)	O(1A)-N(7)	1.306(15)
O(23)-N(11)	1.310(14)	O(34)-N(5)	1.286(13)
Angles			
O(24)-Tb(1)-O(30)	141.8(3)	O(6)-Tb(2)-N(9)#1	140.5(3)
O(24)-Tb(1)-O(34)#3	73.6(3)	O(20)-Tb(2)-O(6)	71.4(3)
O(24)-Tb(1)-O(40)	80.0(4)	O(20)-Tb(2)-O(36)	145.6(3)
O(24)-Tb(1)-N(12)	142.6(4)	O(20)-Tb(2)-O(42)	125.2(3)
O(25)-Tb(1)-O(24)	72.6(3)	O(20)-Tb(2)-N(9)#1	72.4(4)
O(25)-Tb(1)-O(30)	143.6(3)	O(21)-Tb(2)-O(6)	104.8(3)
O(25)-Tb(1)-O(34)#3	108.3(3)	O(21)-Tb(2)-O(20)	76.9(3)
O(25)-Tb(1)-O(40)	75.5(4)	O(21)-Tb(2)-O(28)	145.3(3)
O(25)-Tb(1)-N(12)	79.6(4)	O(21)-Tb(2)-O(35)	74.8(4)
O(30)-Tb(1)-O(34)#3	80.5(3)	O(21)-Tb(2)-O(36)	137.5(3)
O(30)-Tb(1)-O(38)	73.2(4)	O(21)-Tb(2)-O(42)	73.3(3)
O(30)-Tb(1)-N(12)	72.8(4)	O(21)-Tb(2)-N(9)#1	81.7(4)

O(34)#3-Tb(1)-N(12)	140.4(4)	O(28)-Tb(2)-O(6)	81.4(3)
O(38)-Tb(1)-O(24)	123.0(4)	O(28)-Tb(2)-O(20)	72.9(3)
O(38)-Tb(1)-O(25)	76.0(4)	O(28)-Tb(2)-O(35)	115.5(4)
O(38)-Tb(1)-O(30)	73.2(4)	O(28)-Tb(2)-O(36)	74.4(3)
O(38)-Tb(1)-O(34)#3	72.9(3)	O(28)-Tb(2)-O(42)	139.4(3)
O(38)-Tb(1)-O(40)	134.6(4)	O(28)-Tb(2)-N(9)#1	73.3(3)
O(38)-Tb(1)-N(12)	71.7(4)	O(35)-Tb(2)-O(6)	151.6(3)
O(39)-Tb(1)-O(24)	76.1(4)	O(35)-Tb(2)-O(20)	133.8(3)
O(39)-Tb(1)-O(25)	137.5(4)	O(35)-Tb(2)-O(36)	71.3(4)
O(39)-Tb(1)-O(30)	75.9(4)	O(35)-Tb(2)-O(42)	79.7(3)
O(39)-Tb(1)-O(34)#3	89.2(4)	O(35)-Tb(2)-N(9)#1	67.9(4)
O(39)-Tb(1)-O(38)	146.3(4)	O(36)-Tb(2)-O(6)	93.6(3)
O(39)-Tb(1)-O(40)	71.4(4)	O(36)-Tb(2)-O(42)	76.0(3)
O(39)-Tb(1)-N(12)	111.4(4)	O(36)-Tb(2)-N(9)#1	107.8(4)
O(40)-Tb(1)-O(30)	114.5(4)	O(42)-Tb(2)-O(6)	73.3(3)
O(40)-Tb(1)-O(34)#3	150.4(3)	O(42)-Tb(2)-N(9)#1	143.3(4)
O(2)-Tb(3)-O(9)	133.4(3)	O(1)-Tb(4)-N(8)	143.5(3)
O(2)-Tb(3)-O(23)	72.3(3)	O(11)-Tb(4)-O(1)	107.3(3)
O(4)-Tb(3)-O(2)	74.1(3)	O(11)-Tb(4)-O(12)	75.1(3)
O(4)-Tb(3)-O(9)	76.5(3)	O(11)-Tb(4)-O(14)	143.9(3)
O(4)-Tb(3)-O(13)	72.4(3)	O(11)-Tb(4)-O(19)	76.3(3)
O(4)-Tb(3)-O(22)	145.9(3)	O(11)-Tb(4)-O(26)	73.0(3)
O(4)-Tb(3)-O(23)	104.4(3)	O(11)-Tb(4)-N(8)	79.9(4)
O(7)-Tb(3)-O(2)	72.3(3)	O(12)-Tb(4)-O(1)	147.2(3)
O(7)-Tb(3)-O(4)	91.0(3)	O(12)-Tb(4)-O(14)	116.5(3)
O(7)-Tb(3)-O(9)	72.9(3)	O(12)-Tb(4)-O(19)	135.8(3)
O(7)-Tb(3)-O(13)	150.3(3)	O(12)-Tb(4)-O(26)	76.7(3)
O(7)-Tb(3)-O(16)	103.1(3)	O(12)-Tb(4)-N(8)	69.2(4)
O(7)-Tb(3)-O(22)	72.1(3)	O(14)-Tb(4)-O(1)	81.4(3)
O(7)-Tb(3)-O(23)	135.4(3)	O(14)-Tb(4)-O(26)	141.2(3)
O(9)-Tb(3)-O(23)	151.1(3)	O(14)-Tb(4)-N(8)	74.0(4)
O(13)-Tb(3)-O(2)	123.4(3)	O(19)-Tb(4)-O(1)	74.0(3)
O(13)-Tb(3)-O(7)	150.8(3)	O(19)-Tb(4)-O(14)	72.7(3)
O(13)-Tb(3)-O(9)	79.3(3)	O(19)-Tb(4)-O(26)	124.9(3)
O(13)-Tb(3)-O(22)	134.1(3)	O(19)-Tb(4)-N(8)	73.2(3)
O(13)-Tb(3)-O(23)	73.7(3)	O(26)-Tb(4)-O(1)	73.1(3)
O(16)-Tb(3)-O(2)	146.1(3)	O(26)-Tb(4)-N(8)	140.8(3)
O(16)-Tb(3)-O(4)	139.7(3)	O(41)-Tb(4)-O(1)	89.6(3)
O(16)-Tb(3)-O(7)	103.4(3)	O(41)-Tb(4)-O(11)	140.7(3)
O(16)-Tb(3)-O(9)	72.1(3)	O(41)-Tb(4)-O(12)	72.1(3)
O(16)-Tb(3)-O(13)	77.4(3)	O(41)-Tb(4)-O(14)	72.2(3)
O(16)-Tb(3)-O(22)	74.1(3)	O(41)-Tb(4)-O(19)	143.1(3)
O(16)-Tb(3)-O(23)	91.7(3)	O(41)-Tb(4)-O(26)	78.8(3)
O(22)-Tb(3)-O(2)	72.6(3)	O(41)-Tb(4)-N(8)	107.5(4)

O(22)-Tb(3)-O(7)	72.1(3)	N(3)-O(1)-Tb(4)	137.8(7)
O(22)-Tb(3)-O(9)	123.3(3)	N(2)-O(7)-Tb(3)	142.8(7)
O(22)-Tb(3)-O(23)	72.0(3)	N(1)-O(6)-Tb(2)	135.7(7)
O(5)-Tb(5)-O(10)#2	78.9(3)	O(3)-Tb(6)-O(8)	139.8(3)
O(5)-Tb(5)-N(10)	74.1(4)	O(3)-Tb(6)-O(15)	78.0(3)
O(10)#2-Tb(5)-N(10)	142.1(4)	O(3)-Tb(6)-O(17)	89.7(3)
O(27)-Tb(5)-O(5)	120.3(3)	O(3)-Tb(6)-O(18)	72.3(3)
O(27)-Tb(5)-O(10)#2	148.9(3)	O(3)-Tb(6)-O(1A)	103.3(3)
O(27)-Tb(5)-N(10)	69.0(4)	O(3)-Tb(6)-O(37)	76.6(3)
O(29)-Tb(5)-O(5)	145.9(3)	O(3)-Tb(6)-O(43)	148.3(3)
O(29)-Tb(5)-O(10)#2	106.1(3)	O(8)-Tb(6)-O(15)	72.3(3)
O(29)-Tb(5)-O(27)	72.5(3)	O(8)-Tb(6)-O(17)	106.5(3)
O(29)-Tb(5)-O(31)	75.9(3)	O(8)-Tb(6)-O(18)	76.1(3)
O(29)-Tb(5)-O(33)	73.8(3)	O(8)-Tb(6)-O(1A)	89.8(3)
O(29)-Tb(5)-N(10)	83.4(4)	O(8)-Tb(6)-O(37)	143.1(3)
O(31)-Tb(5)-O(5)	73.0(3)	O(8)-Tb(6)-O(43)	71.8(3)
O(31)-Tb(5)-O(10)#2	73.4(3)	O(17)-Tb(6)-O(15)	72.9(3)
O(31)-Tb(5)-O(27)	133.1(3)	O(17)-Tb(6)-O(1A)	136.9(3)
O(31)-Tb(5)-O(29)	76.2(3)	O(17)-Tb(6)-O(37)	72.0(3)
O(31)-Tb(5)-O(33)	125.3(3)	O(18)-Tb(6)-O(15)	79.6(3)
O(31)-Tb(5)-N(10)	73.5(3)	O(18)-Tb(6)-O(17)	149.8(3)
O(32)-Tb(5)-O(5)	73.1(3)	O(18)-Tb(6)-O(1A)	72.3(4)
O(32)-Tb(5)-O(10)#2	94.2(3)	O(18)-Tb(6)-O(37)	124.2(3)
O(32)-Tb(5)-O(27)	71.5(3)	O(1A)-Tb(6)-O(15)	149.7(4)
O(32)-Tb(5)-O(29)	138.1(3)	O(1A)-Tb(6)-O(37)	71.4(4)
O(32)-Tb(5)-O(31)	145.6(4)	O(37)-Tb(6)-O(15)	136.4(3)
O(32)-Tb(5)-O(33)	78.3(3)	O(43)-Tb(6)-O(15)	121.0(3)
O(32)-Tb(5)-N(10)	102.8(4)	O(43)-Tb(6)-O(17)	74.5(3)
O(33)-Tb(5)-O(5)	137.8(3)	O(43)-Tb(6)-O(18)	132.3(3)
O(33)-Tb(5)-O(10)#2	73.0(3)	O(43)-Tb(6)-O(1A)	73.3(3)
O(33)-Tb(5)-O(27)	77.1(3)	O(43)-Tb(6)-O(37)	72.5(3)
O(33)-Tb(5)-O(29)	73.5(3)	N(6)-O(10)-Tb(5)#4	137.5(8)

Symmetry transformations used to generate equivalent atoms:

#1 x-1, y, z+1; #2 x-1, y, z; #3 x, y, z+1; #4 x+1, y, z; #5 x, y, z-1

Table S3. Selected bond distances (Å) and angles (°) for complex 3.

Bond distances			
Dy(1)-O(24)	2.375(9)	Dy(2)-O(6)	2.389(9)
Dy(1)-O(25)	2.361(10)	Dy(2)-O(20)	2.368(10)
Dy(1)-O(30)	2.374(9)	Dy(2)-O(21)	2.342(9)
Dy(1)-O(34)#3	2.377(9)	Dy(2)-O(28)	2.364(9)
Dy(1)-O(38)	2.362(10)	Dy(2)-O(35)	2.368(11)
Dy(1)-O(39)	2.352(12)	Dy(2)-O(36)	2.371(10)
Dy(1)-O(40)	2.375(11)	Dy(2)-O(42)	2.372(9)
Dy(1)-N(12)	2.551(12)	Dy(2)-N(9)#1	2.544(10)
Dy(3)-O(2)	2.377(10)	Dy(4)-O(1)	2.381(8)
Dy(3)-O(4)	2.355(9)	Dy(4)-O(11)	2.338(9)
Dy(3)-O(7)	2.353(8)	Dy(4)-O(12)	2.355(9)
Dy(3)-O(9)	2.377(8)	Dy(4)-O(14)	2.360(10)
Dy(3)-O(13)	2.356(10)	Dy(4)-O(19)	2.360(9)
Dy(3)-O(16)	2.352(9)	Dy(4)-O(26)	2.364(9)
Dy(3)-O(22)	2.380(9)	Dy(4)-O(41)	2.329(9)
Dy(3)-O(23)	2.387(8)	Dy(4)-N(8)	2.549(11)
Dy(5)-O(5)	2.376(10)	Dy(6)-O(3)	2.354(8)
Dy(5)-O(10)#2	2.381(8)	Dy(6)-O(8)	2.362(9)
Dy(5)-O(27)	2.375(10)	Dy(6)-O(15)	2.380(8)
Dy(5)-O(29)	2.359(10)	Dy(6)-O(17)	2.366(8)
Dy(5)-O(31)	2.355(9)	Dy(6)-O(18)	2.365(10)
Dy(5)-O(32)	2.344(9)	Dy(6)-O(1A)	2.374(8)
Dy(5)-O(33)	2.354(10)	Dy(6)-O(37)	2.379(9)
Dy(5)-N(10)	2.522(12)	Dy(6)-O(43)	2.357(8)
O(1)-N(3)	1.322(13)	O(6)-N(1)	1.326(12)
O(7)-N(2)	1.326(12)	O(10)-N(6)	1.284(13)
O(17)-N(4)	1.282(13)	O(1A)-N(7)	1.307(15)
O(23)-N(11)	1.310(14)	O(34)-N(5)	1.290(13)
Angles			
O(24)-Dy(1)-O(34)#3	73.6(3)	O(6)-Dy(2)-N(9)#1	140.4(3)
O(24)-Dy(1)-N(12)	142.7(4)	O(20)-Dy(2)-O(6)	71.4(3)
O(25)-Dy(1)-O(24)	72.6(4)	O(20)-Dy(2)-O(36)	145.7(3)
O(25)-Dy(1)-O(30)	143.7(3)	O(20)-Dy(2)-O(42)	125.5(3)
O(25)-Dy(1)-O(34)#3	108.2(3)	O(20)-Dy(2)-N(9)#1	72.4(4)
O(25)-Dy(1)-O(38)	75.9(4)	O(21)-Dy(2)-O(6)	104.7(3)
O(25)-Dy(1)-O(40)	75.5(4)	O(21)-Dy(2)-O(20)	77.0(3)
O(25)-Dy(1)-N(12)	79.7(4)	O(21)-Dy(2)-O(28)	145.4(3)
O(30)-Dy(1)-O(24)	141.8(3)	O(21)-Dy(2)-O(35)	75.0(4)
O(30)-Dy(1)-O(34)#3	80.5(3)	O(21)-Dy(2)-O(36)	137.3(3)
O(30)-Dy(1)-O(40)	114.6(4)	O(21)-Dy(2)-O(42)	73.3(3)
O(30)-Dy(1)-N(12)	72.7(4)	O(21)-Dy(2)-N(9)#1	81.9(4)

O(34)#3-Dy(1)-N(12)	140.3(4)	O(28)-Dy(2)-O(6)	81.3(3)
O(38)-Dy(1)-O(24)	123.0(4)	O(28)-Dy(2)-O(20)	72.8(3)
O(38)-Dy(1)-O(30)	73.3(4)	O(28)-Dy(2)-O(35)	115.5(4)
O(38)-Dy(1)-O(34)#3	72.9(3)	O(28)-Dy(2)-O(36)	74.6(3)
O(38)-Dy(1)-O(40)	134.6(4)	O(28)-Dy(2)-O(42)	139.3(3)
O(38)-Dy(1)-N(12)	71.6(4)	O(28)-Dy(2)-N(9)#1	73.3(3)
O(39)-Dy(1)-O(24)	76.1(4)	O(35)-Dy(2)-O(6)	151.6(3)
O(39)-Dy(1)-O(25)	137.5(4)	O(35)-Dy(2)-O(20)	133.8(3)
O(39)-Dy(1)-O(30)	75.9(4)	O(35)-Dy(2)-O(36)	71.1(4)
O(39)-Dy(1)-O(34)#3	89.2(4)	O(35)-Dy(2)-O(42)	79.5(3)
O(39)-Dy(1)-O(38)	146.4(4)	O(35)-Dy(2)-N(9)#1	67.9(4)
O(39)-Dy(1)-O(40)	71.3(4)	O(36)-Dy(2)-O(6)	93.7(3)
O(39)-Dy(1)-N(12)	111.3(4)	O(36)-Dy(2)-O(42)	75.7(3)
O(40)-Dy(1)-O(24)	79.8(4)	O(36)-Dy(2)-N(9)#1	107.8(4)
O(40)-Dy(1)-O(34)#3	150.3(3)	O(42)-Dy(2)-O(6)	73.5(3)
O(40)-Dy(1)-N(12)	69.2(4)	O(42)-Dy(2)-N(9)#1	143.2(4)
N(5)-O(34)-Dy(1)#5	138.9(8)	N(1)-O(6)-Dy(2)	135.6(7)
O(2)-Dy(3)-O(22)	72.6(3)	O(1)-Dy(4)-N(8)	143.5(3)
O(2)-Dy(3)-O(23)	72.3(3)	O(11)-Dy(4)-O(1)	107.3(3)
O(4)-Dy(3)-O(2)	74.0(3)	O(11)-Dy(4)-O(12)	75.3(3)
O(4)-Dy(3)-O(9)	76.5(3)	O(11)-Dy(4)-O(14)	143.7(3)
O(4)-Dy(3)-O(13)	72.4(3)	O(11)-Dy(4)-O(19)	76.1(3)
O(4)-Dy(3)-O(22)	145.8(3)	O(11)-Dy(4)-O(26)	73.0(3)
O(4)-Dy(3)-O(23)	104.4(3)	O(11)-Dy(4)-N(8)	80.0(4)
O(7)-Dy(3)-O(2)	72.4(3)	O(12)-Dy(4)-O(1)	147.3(3)
O(7)-Dy(3)-O(4)	91.0(3)	O(12)-Dy(4)-O(14)	116.4(3)
O(7)-Dy(3)-O(9)	72.8(3)	O(12)-Dy(4)-O(19)	135.9(3)
O(7)-Dy(3)-O(13)	150.3(3)	O(12)-Dy(4)-O(26)	76.7(3)
O(7)-Dy(3)-O(22)	72.2(3)	O(12)-Dy(4)-N(8)	69.1(4)
O(7)-Dy(3)-O(23)	135.4(3)	O(14)-Dy(4)-O(1)	81.4(3)
O(9)-Dy(3)-O(2)	133.3(3)	O(14)-Dy(4)-O(26)	141.5(3)
O(9)-Dy(3)-O(22)	123.4(3)	O(14)-Dy(4)-N(8)	73.8(4)
O(9)-Dy(3)-O(23)	151.1(3)	O(19)-Dy(4)-O(1)	74.0(3)
O(13)-Dy(3)-O(2)	123.3(3)	O(19)-Dy(4)-O(14)	72.7(3)
O(13)-Dy(3)-O(9)	79.4(3)	O(19)-Dy(4)-O(26)	124.8(3)
O(13)-Dy(3)-O(22)	134.1(3)	O(19)-Dy(4)-N(8)	73.3(3)
O(13)-Dy(3)-O(23)	73.7(3)	O(26)-Dy(4)-O(1)	73.3(3)
O(16)-Dy(3)-O(2)	146.3(3)	O(26)-Dy(4)-N(8)	140.7(3)
O(16)-Dy(3)-O(4)	139.6(3)	O(41)-Dy(4)-O(1)	89.7(3)
O(16)-Dy(3)-O(7)	103.2(3)	O(41)-Dy(4)-O(11)	140.6(3)
O(16)-Dy(3)-O(9)	72.2(3)	O(41)-Dy(4)-O(12)	71.8(3)
O(16)-Dy(3)-O(13)	77.2(3)	O(41)-Dy(4)-O(14)	72.5(3)
O(16)-Dy(3)-O(22)	74.3(3)	O(41)-Dy(4)-O(19)	143.3(3)
O(16)-Dy(3)-O(23)	91.6(3)	O(41)-Dy(4)-O(26)	78.8(3)

O(22)-Dy(3)-O(23)	72.0(3)	O(41)-Dy(4)-N(8)	107.3(4)
N(2)-O(7)-Dy(3)	143.0(7)	N(3)-O(1)-Dy(4)	137.7(7)
N(11)-O(23)-Dy(3)	139.3(8)	N(7)-O(1A)-Dy(6)	143.0(9)
O(5)-Dy(5)-O(10)#2	78.9(3)	O(3)-Dy(6)-O(8)	139.6(3)
O(5)-Dy(5)-N(10)	74.1(4)	O(3)-Dy(6)-O(15)	77.9(3)
O(10)#2-Dy(5)-N(10)	142.1(4)	O(3)-Dy(6)-O(17)	89.5(3)
O(27)-Dy(5)-O(5)	120.4(3)	O(3)-Dy(6)-O(18)	72.4(3)
O(27)-Dy(5)-O(10)#2	149.0(3)	O(3)-Dy(6)-O(1A)	103.5(3)
O(27)-Dy(5)-N(10)	68.9(4)	O(3)-Dy(6)-O(37)	76.6(3)
O(29)-Dy(5)-O(5)	145.9(3)	O(3)-Dy(6)-O(43)	148.3(3)
O(29)-Dy(5)-O(10)#2	106.0(3)	O(8)-Dy(6)-O(15)	72.2(3)
O(29)-Dy(5)-O(27)	72.5(3)	O(8)-Dy(6)-O(17)	106.4(3)
O(29)-Dy(5)-N(10)	83.6(4)	O(8)-Dy(6)-O(18)	76.0(3)
O(31)-Dy(5)-O(5)	73.0(3)	O(8)-Dy(6)-O(1A)	89.9(3)
O(31)-Dy(5)-O(10)#2	73.4(3)	O(8)-Dy(6)-O(37)	143.3(3)
O(31)-Dy(5)-O(27)	133.0(3)	O(15)-Dy(6)-O(17)	73.0(3)
O(31)-Dy(5)-O(29)	76.1(4)	O(15)-Dy(6)-O(18)	79.6(3)
O(31)-Dy(5)-N(10)	73.6(3)	O(15)-Dy(6)-O(1A)	149.7(4)
O(32)-Dy(5)-O(5)	73.1(3)	O(15)-Dy(6)-O(37)	136.4(3)
O(32)-Dy(5)-O(10)#2	94.4(3)	O(17)-Dy(6)-O(18)	149.8(3)
O(32)-Dy(5)-O(27)	71.4(3)	O(17)-Dy(6)-O(1A)	136.8(3)
O(32)-Dy(5)-O(29)	138.1(3)	O(17)-Dy(6)-O(37)	72.1(3)
O(32)-Dy(5)-O(31)	145.6(4)	O(18)-Dy(6)-O(1A)	72.3(4)
O(32)-Dy(5)-O(33)	78.4(3)	O(18)-Dy(6)-O(37)	124.2(3)
O(32)-Dy(5)-N(10)	102.5(4)	O(1A)-Dy(6)-O(37)	71.4(3)
O(33)-Dy(5)-O(5)	138.0(3)	O(43)-Dy(6)-O(8)	71.9(3)
O(33)-Dy(5)-O(10)#2	73.1(3)	O(43)-Dy(6)-O(15)	121.1(3)
O(33)-Dy(5)-O(27)	77.0(4)	O(43)-Dy(6)-O(17)	74.5(3)
O(33)-Dy(5)-O(29)	73.3(3)	O(43)-Dy(6)-O(18)	132.3(3)
O(33)-Dy(5)-O(31)	125.4(3)	O(43)-Dy(6)-O(1A)	73.2(3)
O(33)-Dy(5)-N(10)	143.3(4)	O(43)-Dy(6)-O(37)	72.5(3)
N(6)-O(10)-Dy(5)#4	137.8(8)	N(4)-O(17)-Dy(6)	143.4(8)
N(2)-O(7)-Dy(3)	143.0(7)	N(6)-O(10)-Dy(5)#4	137.8(8)
N(4)-O(17)-Dy(6)	143.4(8)	N(7)-O(1A)-Dy(6)	143.0(9)
N(11)-O(23)-Dy(3)	139.3(8)	N(5)-O(34)-Dy(1)#5	138.9(8)

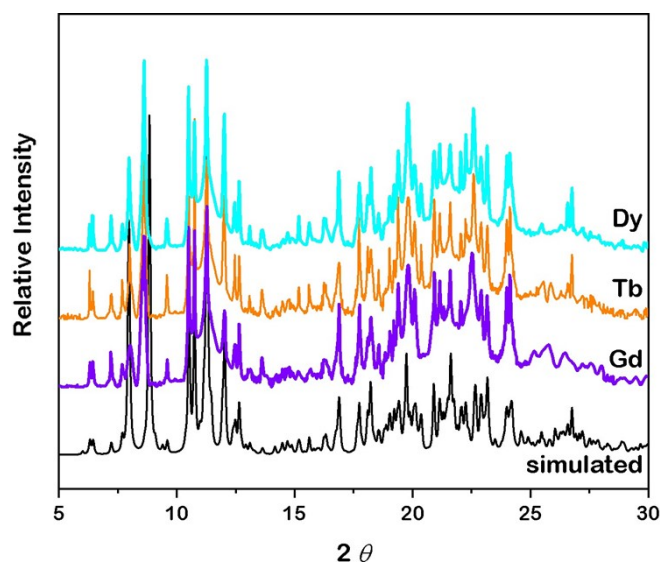
Symmetry transformations used to generate equivalent atoms:

#1 x-1, y, z+1; #2 x-1, y, z; #3 x, y, z+1; #4 x+1, y, z; #5 x, y, z-1

Table S4. SHAPE analyses for complexes **1–3**.

Complex 1	Gd1	Gd2	Gd3	Gd4	Gd5	Gd6
SAPR-8	0.620	1.025	1.335	0.667	1.252	1.115
TDD-8	1.411	1.005	0.417	1.271	0.563	0.676
BTPR-8	1.770	1.806	2.096	1.762	1.974	2.028
Complex 2	Tb1	Tb2	Tb3	Tb4	Tb5	Tb 6
SAPR-8	0.634	1.054	1.387	0.621	1.226	1.136
TDD-8	1.423	1.004	0.458	1.254	0.554	0.658
BTPR-8	1.752	1.854	2.069	1.736	1.997	2.057
Complex 3	Dy1	Dy2	Dy3	Dy4	Dy5	Dy6
SAPR-8	0.639	1.035	1.353	0.684	1.251	1.153
TDD-8	1.478	1.005	0.423	1.235	0.532	0.673
BTPR-8	1.758	1.824	2.013	1.773	1.942	2.027

SAPR-8: Square antiprism (D_{4d}); TDD-8: Triangular dodecahedron (D_{2d}); BTPR-8: Biaugmented trigonal (C_{2v})

**Figure S1.** Powder X-ray diffraction patterns of complexes **1-3**.

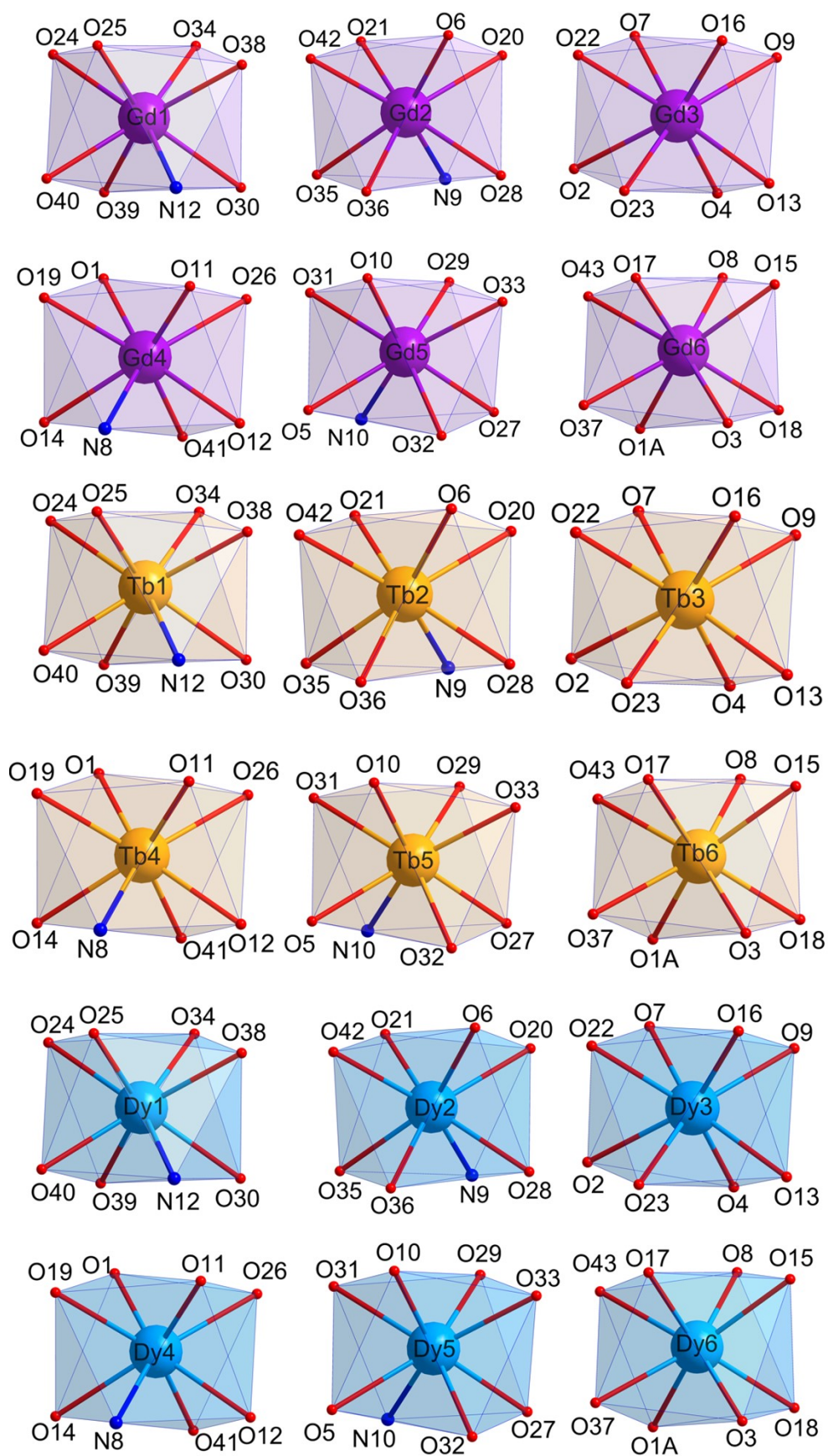
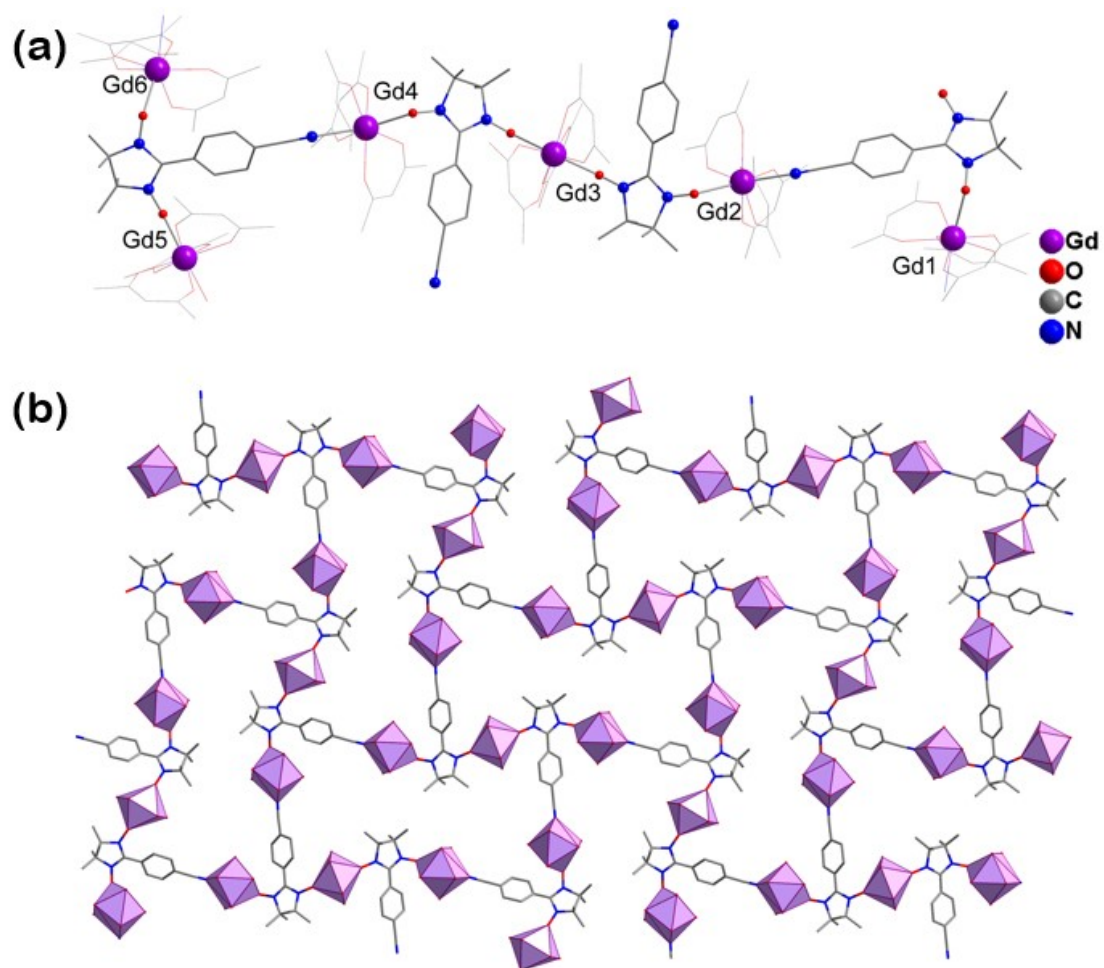


Figure S2. Coordination polyhedrons of Ln(III) ions in complexes 1-3.



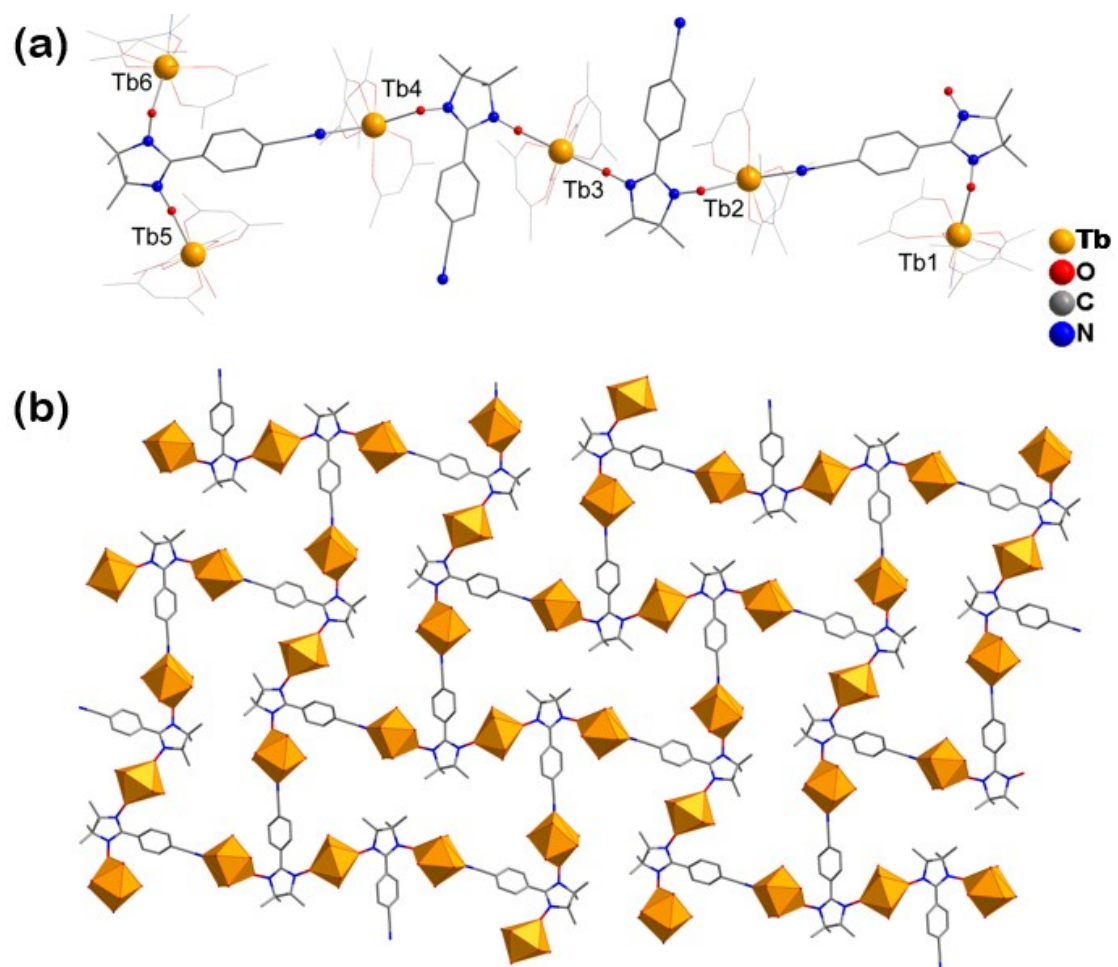


Figure S4. (a) The asymmetric unit of **2** (Fluorine and hydrogen atoms are omitted for clarity); (b) View of 2D coordination network of **2**.

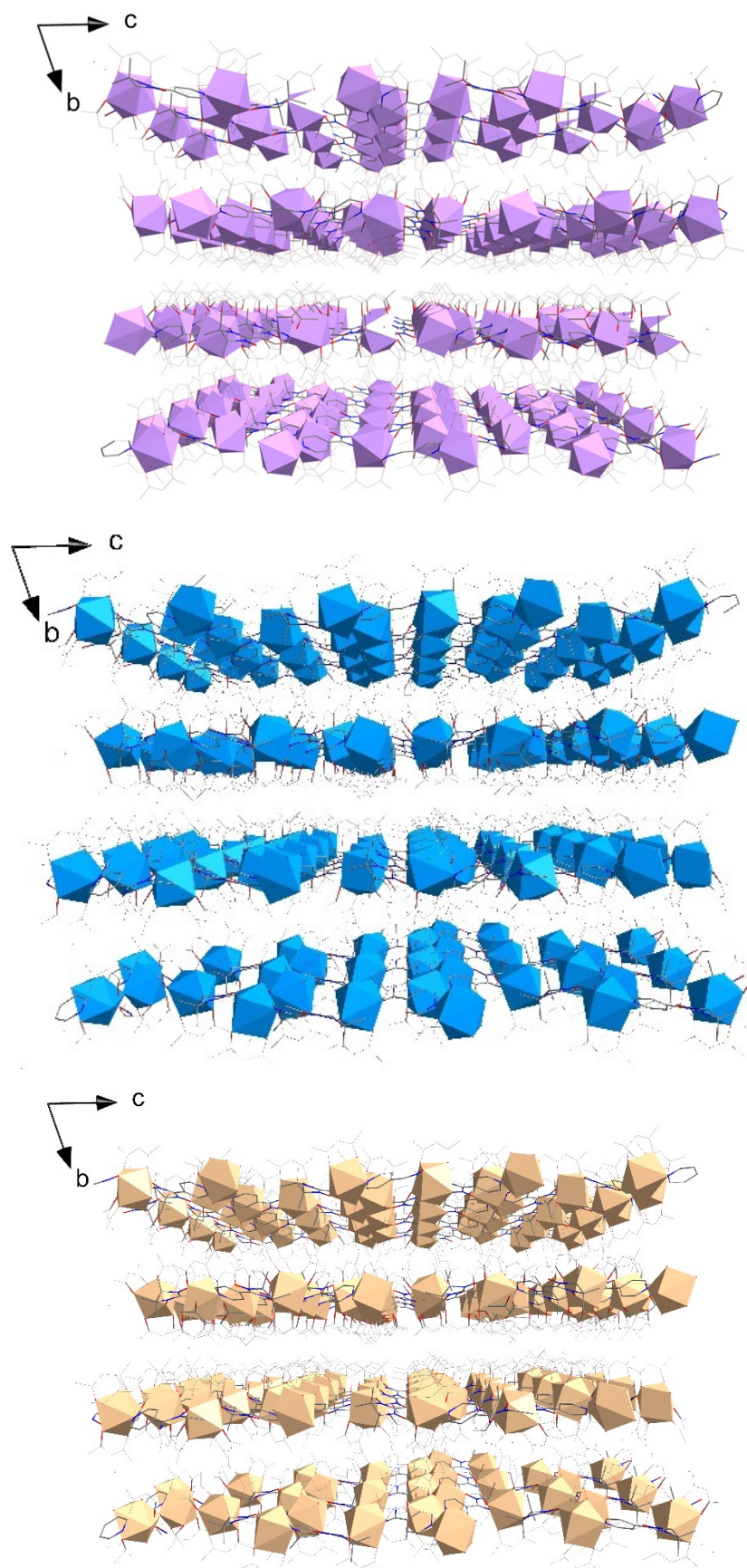


Figure S5. Packing patterns of 2D networks of complexes 1-3 (top to bottom for 1-3, respectively).

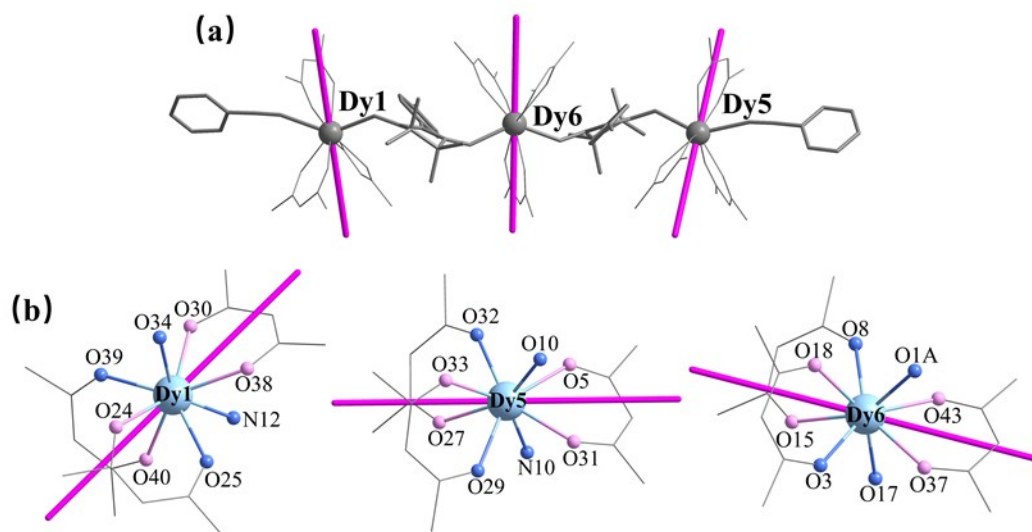


Figure S6. (a) Magellan predicted magnetic axes for the Dy1, Dy6 and Dy5 centers in **3**; (b) The orientations of magnetic axes and the coordination atoms in the first spheres of Dy1, Dy6 and Dy5 centers in **3**. (H and F atoms are omitted for clarity).

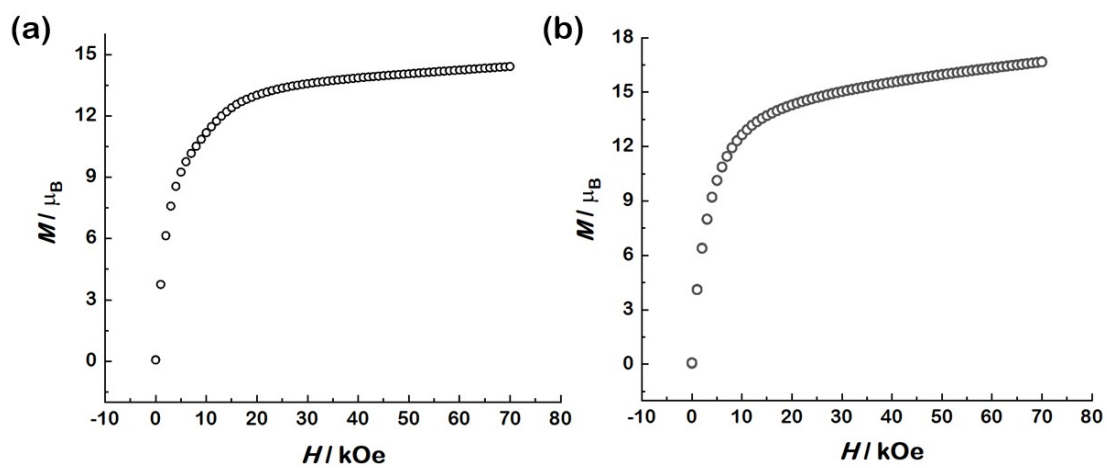


Figure S7. M versus H plots for (a) **2** and (b) **3** at 2 K.

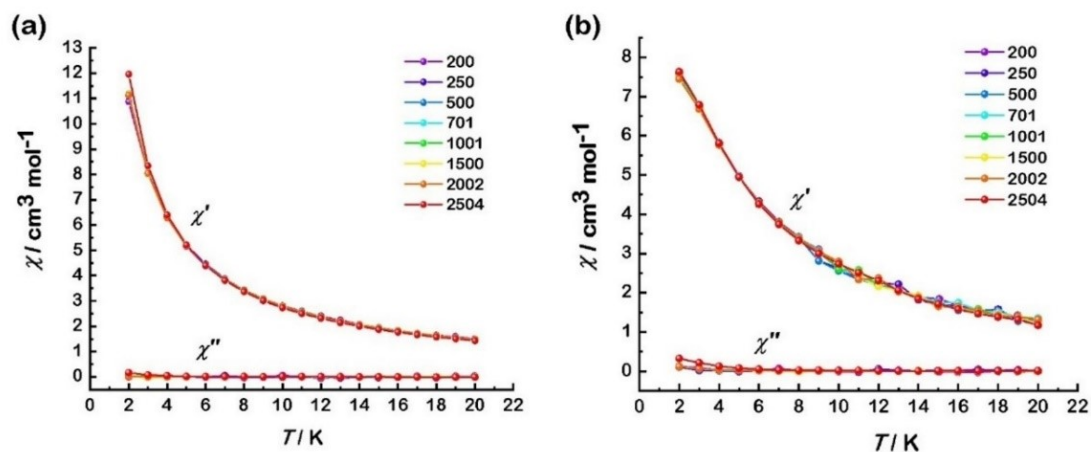


Figure S8. Temperature dependence of the ac magnetic susceptibility for **2** under zero dc field (a) and a dc field of 2500Oe (b).

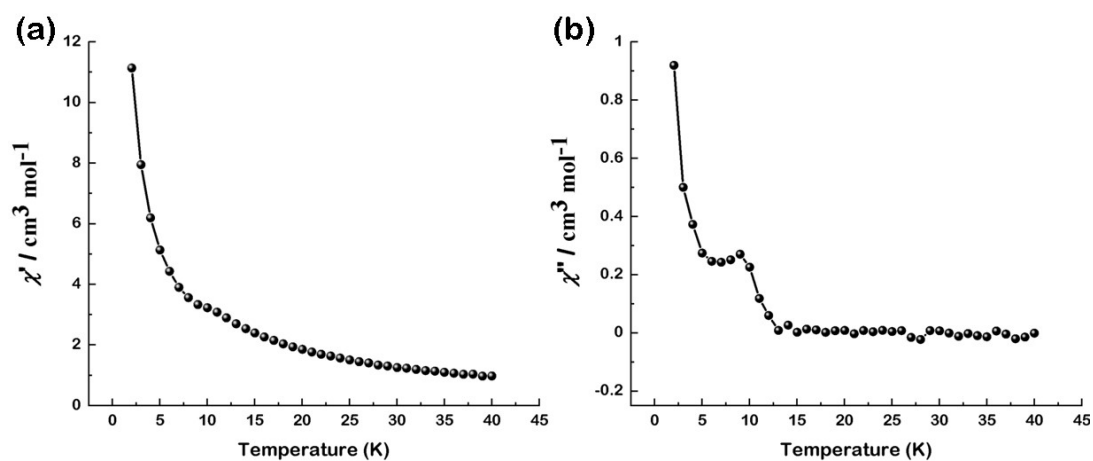


Figure S9. Temperature dependences of in-phase (χ'_{M}) (a) and out-of-phase (χ''_{M}) (b) magnetic susceptibility for **3** in zero dc field with an oscillation of 3 Oe at 997 Hz.