

## Supporting Information

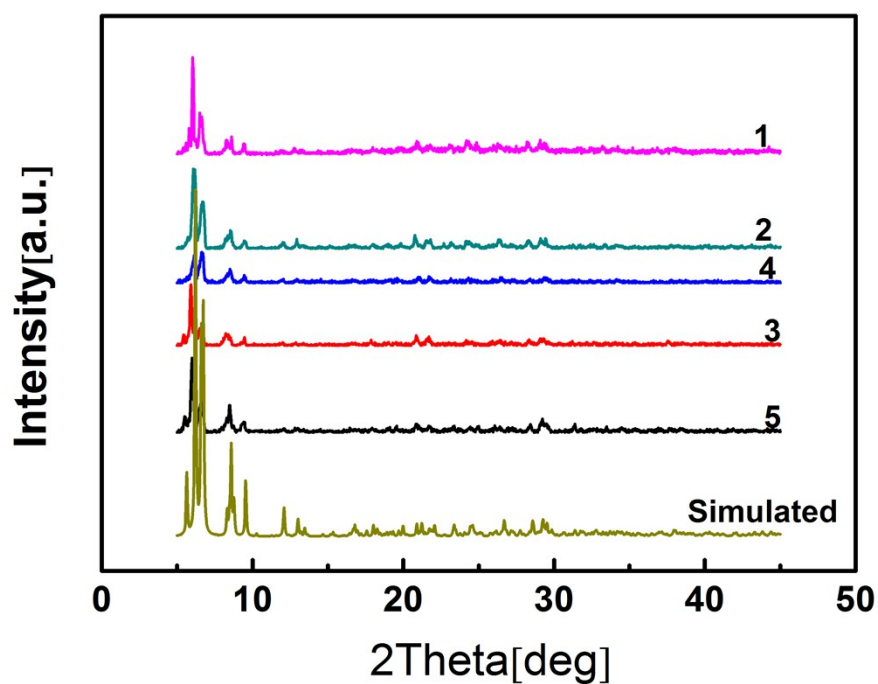


Figure S1. The simulated XRPD pattern and the experimental patterns for complexes 1-5.

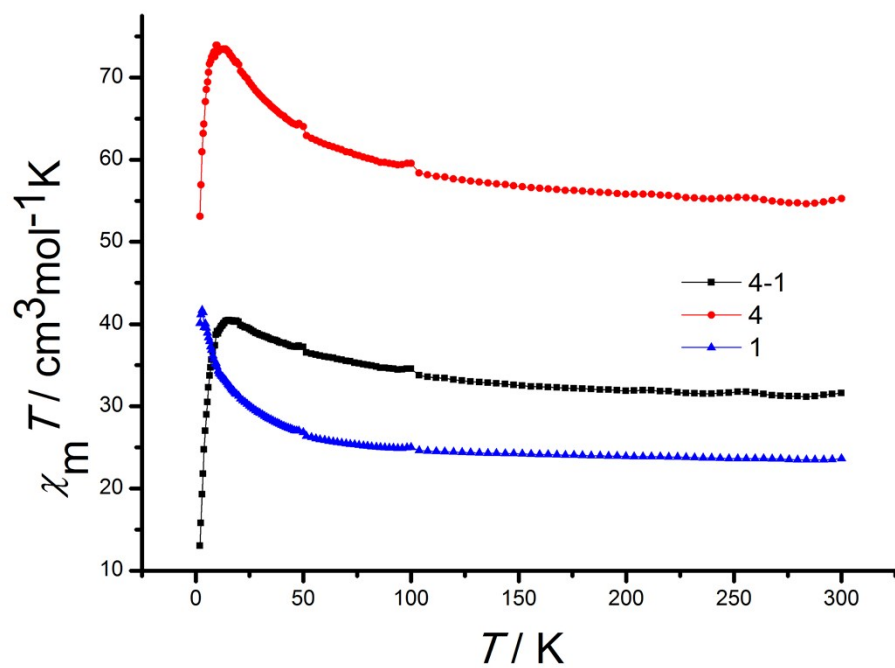


Figure S2. The ‘subtracted’  $\chi_m T$  versus  $T$  plots at 0.1 T for complex 4.

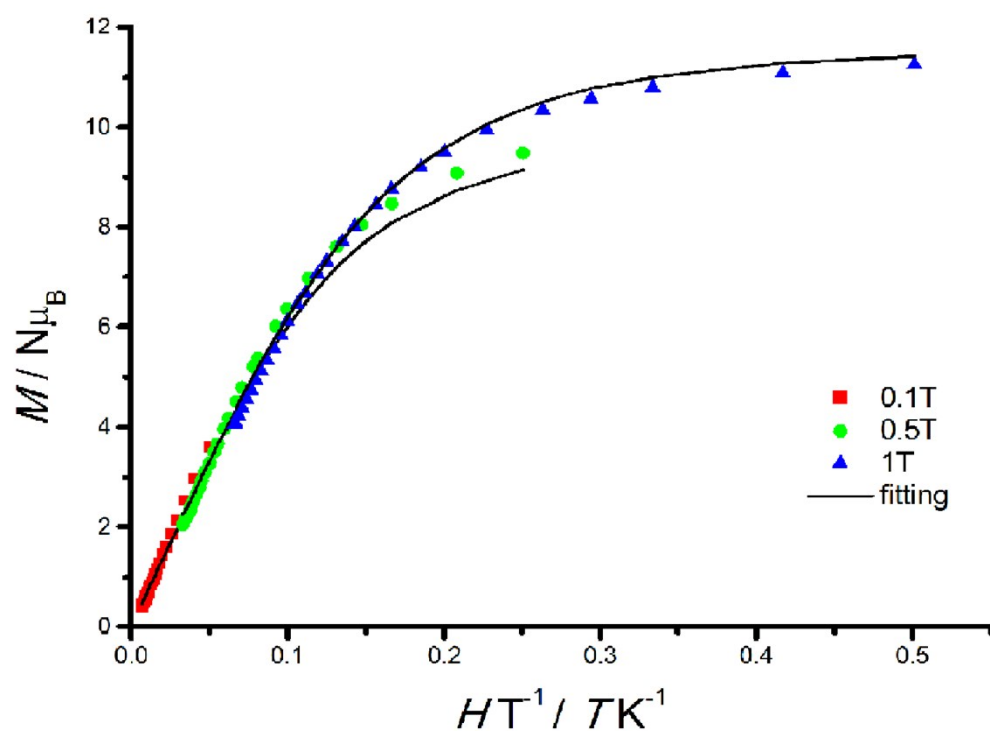


Figure S3. Plots of  $M$  vs.  $H/T$  for complex 1 at the indicated applied field.

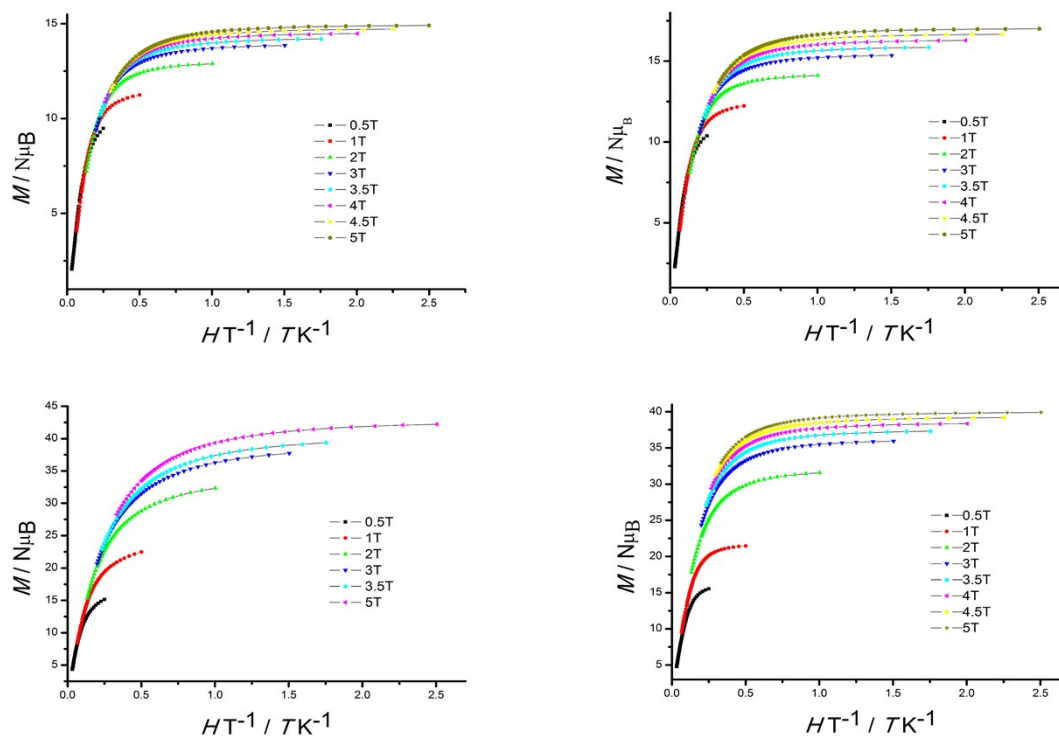


Figure S4. Plots of  $M/N\mu_B$  vs  $H/T$  for complexes 1 (upleft), 2 (upright), 4 (left bottom) and 5 (right bottom). at the indicated applied fields. Solid lines are eye guides.

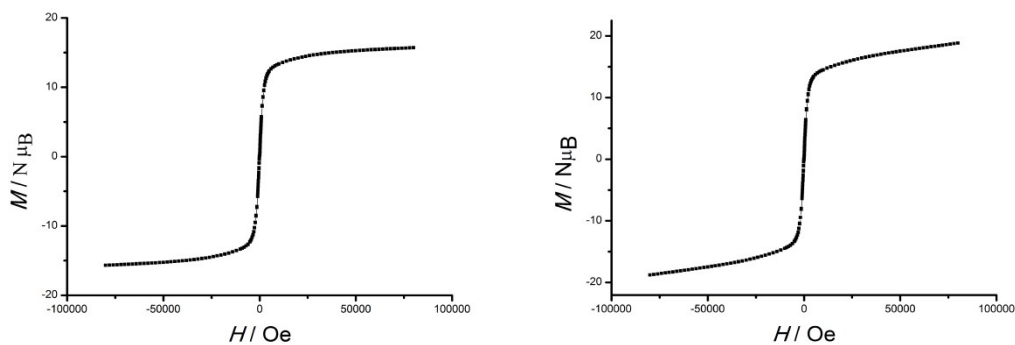


Figure S5. Plots of  $M / N\mu_B$  vs  $H$  for complexes 1 (left) and 2 (right) at 2 K.

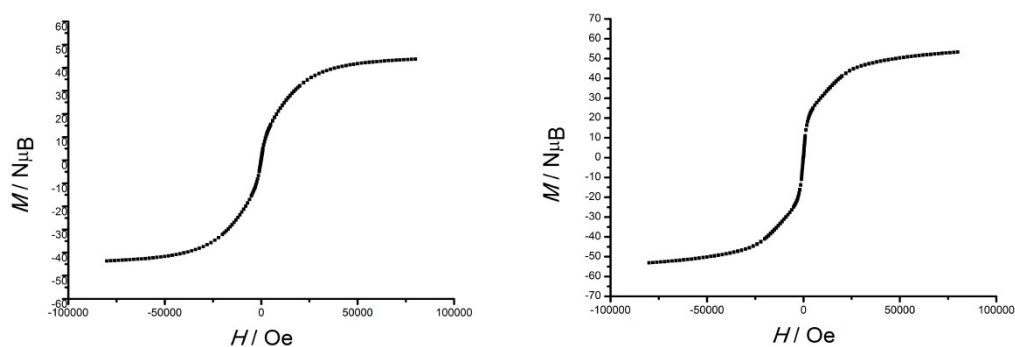


Figure S6. Plots of  $M / N\mu_B$  vs  $H$  for complexes 3 (left) and 4 (right) at 2K.

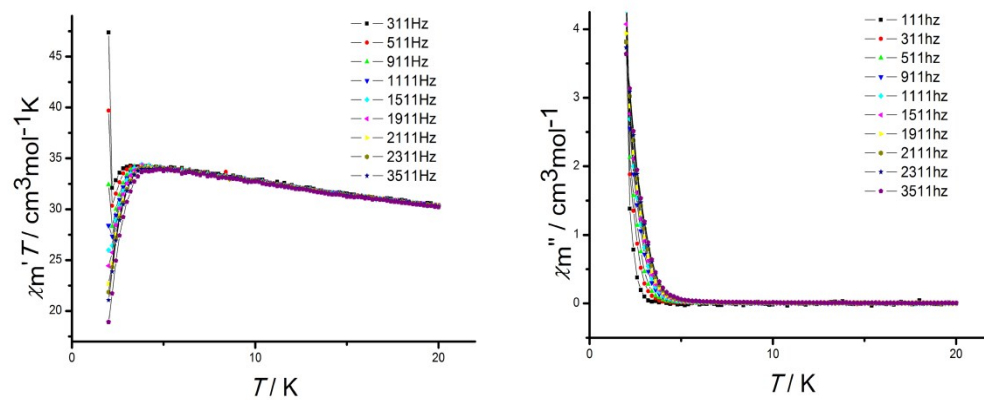


Figure S7. ( left ) In-phase  $\chi'_m T$  and (right) out-of-phase  $\chi''_m$  ac susceptibility signals of complex 1 in a 3.0 G field oscillating.

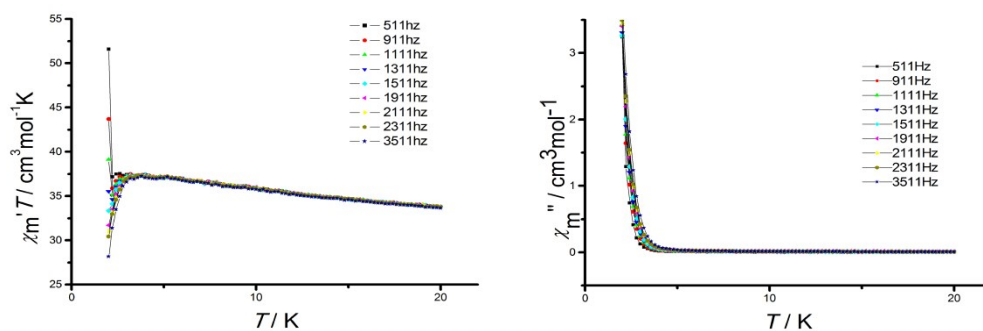


Figure S8. ( left ) In-phase  $\chi'_m T$  and (right) out-of-phase  $\chi''_m$  ac susceptibility signals of complex 2 in a 3.0 G field oscillating.

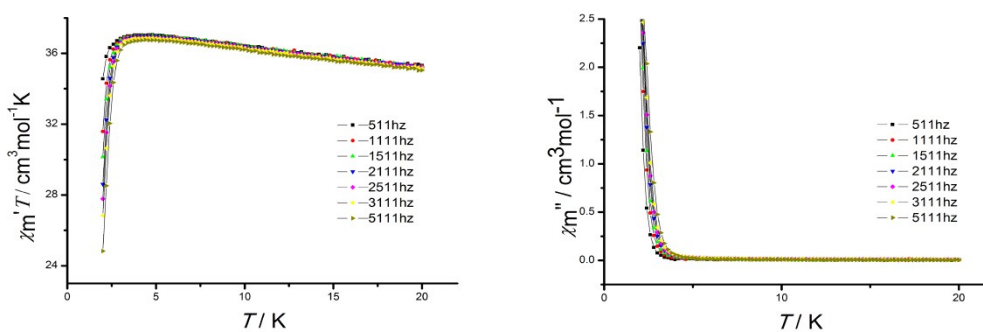


Figure S9. ( left ) In-phase  $\chi'_m T$  and (right) out-of-phase  $\chi''_m$  ac susceptibility signals of complex 3 in a 3.0 G field oscillating.

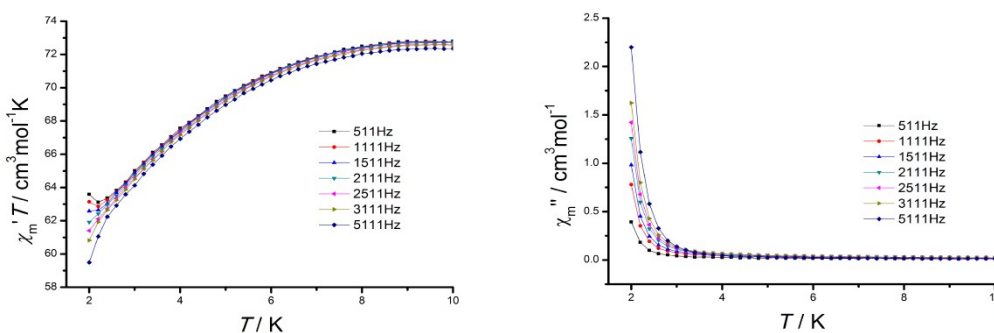


Figure S10. ( left ) In-phase  $\chi'_m T$  and (right) out-of-phase  $-\chi''_m$  ac susceptibility signals of complex 4 in a 3.0 G field oscillating

Table 1. Bond-valence sums for the Mn atoms of complex 4.

Atom	Mn <sup>2+</sup>	Mn <sup>3+</sup>	Mn <sup>4+</sup>
Mn1	3.30	3.04	
Mn2	3.27	3.02	
Mn3	3.85	3.55	3.48
Mn4	3.13	2.86	

Table 2. Bond-valence sums for the O atoms of complex 4.

Atom	BVS	Assignment
O11	1.76	$\mu_4$ -O
O16	2.16	$\mu_3$ -O
O17	2.15	$\mu_3$ -O
O20	2.00	$\mu_4$ -O

Table 3. Selected bond lengths (Å) and angles (deg) for complex 1

La(1)-O(5)	2.447(7)	Mn(3)-O(8)	2.092(4)	O(18)-Mn(1)-Mn(4)	116.82(11)
La(1)-O(2)	2.445(8)	Mn(3)-O(10)	2.114(5)	O(16)-Mn(1)-Mn(4)	41.43(13)
La(1)-O(17)	2.472(4)	Mn(4)-O(12)	1.890(4)	O(9)#1-Mn(1)-Mn(2)	83.75(12)
La(1)-O(26)	2.484(5)	Mn(4)-O(13)	1.942(5)	O(17)-Mn(1)-Mn(2)	99.20(13)
La(2)-O(25)	2.480(4)	Mn(4)-O(14)	1.956(4)	O(25)-Mn(1)-Mn(2)	32.42(13)
La(2)-O(4)	2.515(6)	O(2)-La(1)-Mn(1)	76.90(16)	O(26)-Mn(2)-Mn(4)	143.61(16)
La(2)-O(7)	2.587(4)	O(17)-La(1)-Mn(1)	30.63(10)	O(17)-Mn(1)-Mn(2)	99.20(13)
La(2)-O(18)	2.654(4)	O(26)-La(1)-Mn(1)	80.16(10)	O(25)-Mn(1)-Mn(2)	32.42(13)
Mn(1)-O(18)	2.217(4)	O(25)-La(2)-Mn(2)	28.34(9)	O(26)-Mn(2)-Mn(4)	143.61(16)
Mn(1)-O(16)	2.357(5)	O(4)-La(2)-Mn(2)	99.51(19)	O(12)-Mn(2)-Mn(4)	33.16(12)
Mn(2)-O(26)	1.870(4)	O(7)-La(2)-Mn(2)	37.09(9)	O(12)-Mn(3)-Mn(4)	37.93(11)
Mn(2)-O(12)	1.873(4)	O(18)-La(2)-Mn(2)	88.34(9)	O(11)-Mn(3)-Mn(4)	42.39(13)
Mn(3)-O(9)	1.890(4)	O(1)-La(2)-Mn(1)	99.0(2)	O(12)-Mn(4)-Mn(3)	37.97(13)
Mn(3)-O(12)	1.892(4)	O(3)-La(2)-Mn(1)	107.74(11)	O(13)-Mn(4)-Mn(3)	98.93(16)
Mn(3)-O(11)	2.030(4)	O(6)-La(2)-Mn(1)	147.19(11)		

Table 4. Selected bond lengths (Å) and angles (deg) for complex 2

Pr(1)-O(13)	2.371(6)	Mn(2)-O(16)	1.874(4)	O(9)-Pr(2)-O(6)	75.68(15)
Pr(1)-O(2)	2.409(8)	Mn(2)-O(17)	1.884(4)	O(9)-Pr(2)-O(10)	74.29(15)
Pr(1)-O(2')	2.411(15)	Mn(2)-O(26)	1.886(4)	O(6)-Pr(2)-O(10)	76.27(14)
Pr(1)-O(17)	2.427(4)	Mn(2)-N(4)	2.030(5)	O(9)-Pr(2)-O(12)	72.2(2)
Pr(1)-O(1)	2.442(4)	Mn(2)-O(15)	2.200(4)	O(9)-Pr(2)-Mn(1)	146.99(11)
Pr(1)-O(26)	2.523(4)	Mn(3)-O(11)#1	1.894(4)	O(6)-Pr(2)-Mn(1)	107.97(11)
Pr(1)-O(25)	2.555(5)	Mn(3)-O(8)#1	1.937(5)	O(10)-Pr(2)-Mn(1)	74.88(10)
Pr(1)-O(23)	2.590(5)	Mn(3)-O(7)#1	1.949(4)	O(12)-Pr(2)-Mn(1)	130.08(17)
Pr(2)-O(10)	2.424(4)	Mn(3)-O(10)#1	2.018(4)	O(26)-Pr(2)-Mn(1)	28.81(9)
Pr(2)-O(12)	2.429(6)	Mn(4)-O(16)	1.882(4)	O(9)-Pr(2)-Mn(2)	108.03(11)
Pr(2)-O(26)	2.445(4)	Mn(4)-O(11)	1.889(4)	O(6)-Pr(2)-Mn(2)	148.41(11)
Pr(2)-O(3)	2.480(6)	Mn(4)-O(10)	2.054(4)	O(10)-Pr(2)-Mn(2)	74.83(9)
Pr(2)-O(4)	2.563(4)	O(13)-Pr(1)-O(17)	79.18(19)	O(12)-Pr(2)-Mn(2)	97.32(15)
Mn(1)-O(1)	1.871(4)	O(2)-Pr(1)-O(17)	147.1(3)	O(9)-Pr(2)-Pr(1)	140.81(12)
Mn(1)-O(11)#1	1.872(4)	O(2')-Pr(1)-O(17)	126.5(4)	O(6)-Pr(2)-Pr(1)	139.59(11)
Mn(1)-O(26)	1.900(4)	O(13)-Pr(1)-O(1)	137.1(2)	O(10)-Pr(2)-Pr(1)	122.17(10)
O(17)-Mn(2)-Mn1	98.97(12)	O(17)-Mn(2)-O(18)	100.46(17)	O(16)-Mn(2)-Mn(3)	33.78(12)
O(26)-Mn(2)-Mn1	32.99(12)	O(16)-Mn(2)-O(18)	75.27(16)	O(16)-Mn(2)-Mn(1)	83.69(11)

Table 5. Selected bond lengths (Å) and angles (deg) for complex 3

Nd(1)-O(7)	2.404(5)	Nd(1)-O(1)	2.418(5)	O(16)-Nd(2)-O(22)	73.00(14)
Nd(1)-O(12)	2.407(4)	Nd(2)-O(15)	2.413(4)	O(15)-Nd(2)-O(20)	70.19(14)
Nd(1)-O(23)	2.416(4)	Nd(2)-O(16)	2.423(4)	O(16)-Nd(2)-O(20)	74.87(15)
Nd(2)-O(4)#1	2.390(8)	Nd(2)-O(26)#1	2.508(4)	O(11)-Mn(1)-O(23)	104.44(18)
Mn(1)-O(8)	1.934(5)	O(7)-Nd(1)-O(12)	75.60(16)	O(24)-Mn(1)-O(13)	81.78(18)
Mn(1)-O(11)	1.953(4)	O(7)-Nd(1)-O(23)	74.45(15)	O(8)-Mn(1)-Mn(2)	149.06(15)
Mn(1)-O(23)	2.023(4)	O(12)-Nd(1)-O(23)	76.55(14)	O(11)-Mn(1)-Mn(2)	98.48(15)
Mn(1)-O(13)	2.102(5)	O(7)-Nd(1)-O(1)	72.3(2)	O(23)-Mn(1)-Mn(2)	43.56(12)
Mn(2)-O(24)	1.878(4)	O(7)-Nd(1)-Mn(3)#1	147.10(11)	O(8)-Mn(1)-Mn(4)	141.15(15)
Mn(2)-O(25)	1.884(4)	O(12)-Nd(1)-Mn(3)#1	108.19(11)	O(11)-Mn(1)-Mn(4)	95.33(14)
Mn(2)-O(23)#1	2.050(4)	O(23)-Nd(1)-Mn(3)#1	74.84(10)	O(23)-Mn(1)-Mn(4)	110.95(12)
Mn(2)-O(23)	2.060(4)	O(1)-Nd(1)-Mn(3)#1	129.91(18)	O(24)-Mn(2)-O(23)	80.39(16)
Mn(2)-O(9)	2.101(4)	O(12)-Nd(1)-Mn(4)#1	148.72(11)	O(25)-Mn(2)-O(23)	94.29(17)
Mn(3)-O(16)	1.869(4)	O(23)-Nd(1)-Mn(4)#1	74.76(10)	O(10)#1-Mn(3)-Mn(1)	115.10(11)
Mn(3)-O(25)#1	1.876(4)	O(1)-Nd(1)-Mn(4)#1	97.23(15)	O(13)-Mn(3)-Mn(1)	41.88(13)
Mn(3)-O(26)#1	1.903(4)	O(7)-Nd(1)-Nd(2)#1	140.76(11)	O(16)-Mn(3)-Mn(4)	98.86(13)
Mn(4)-O(24)	1.875(4)	O(12)-Nd(1)-Nd(2)#1	139.67(11)	O(25)#1-Mn(3)-Mn(4)	83.77(12)
Mn(4)-O(26)#1	1.884(4)	O(23)-Nd(1)-Nd(2)#1	121.93(10)	O(13)-Mn(3)-Nd(2)	89.74(12)
Mn(4)-O(15)	1.886(4)	O(15)-Nd(2)-O(22)	119.50(16)	O(15)-Mn(4)-Mn(3)	99.05(12)

Table 6. Selected bond lengths (Å) and angles (deg) for complex 5

Dy(1)-O(3)	2.270(5)	Mn(4)-O(15)	1.883(4)	O(4)-Dy(2)-Mn(1)	121.65(13)
Dy(1)-O(1)	2.282(5)	Mn(4)-O(12)#1	1.892(4)	O(5)-Dy(2)-Mn(1)	151.95(10)
Dy(1)-O(20)	2.342(4)	Mn(4)-O(8)#1	1.923(4)	O(15)-Mn(1)-O(20)	171.05(18)
Dy(1)-O(18)	2.363(4)	Mn(4)-O(6)#1	1.947(4)	O(15)-Mn(1)-O(19)	100.08(16)
Dy(1)-O(19)	2.451(3)	O(3)-Dy(1)-O(1)	81.5(2)	O(20)-Mn(1)-O(19)	86.31(16)
Dy(2)-O(11)	2.318(4)	O(3)-Dy(1)-O(20)	147.02(17)	O(20)-Mn(1)-Mn(4)	141.99(13)
Dy(2)-O(7)	2.339(4)	O(1)-Dy(1)-O(20)	79.55(19)	O(19)-Mn(1)-Mn(4)	88.67(11)
Dy(2)-O(2)	2.339(5)	O(3)-Dy(1)-O(18)	75.83(17)	O(15)-Mn(1)-Mn(3)	83.61(12)
Dy(2)-O(4)	2.342(5)	O(1)-Dy(1)-O(18)	140.93(18)	O(20)-Mn(1)-Mn(3)	98.68(12)
Dy(2)-O(5)	2.353(6)	O(3)-Dy(1)-Mn(1)	117.86(14)	O(9)-Mn(1)-Dy(1)	93.08(11)
Mn(1)-O(15)	2.345(4)	O(1)-Dy(1)-Mn(1)	80.53(15)	O(16)-Mn(1)-Dy(1)	89.12(18)
Mn(1)-O(20)	1.880(4)	O(20)-Dy(1)-Mn(1)	32.13(9)	O(15)-Mn(1)-Dy(2)	84.42(12)
Mn(1)-O(19)	1.882(4)	O(18)-Dy(1)-Mn(1)	82.52(10)	O(20)-Mn(1)-Dy(2)	104.37(13)
Mn(2)-O(15)	1.890(4)	O(19)-Dy(1)-Mn(1)	32.99(9)	O(15)-Mn(2)-O(12)	172.22(17)
Mn(2)-O(12)	1.875(4)	O(23)-Dy(1)-Mn(1)	136.05(10)	O(15)-Mn(2)-O(11)	93.58(16)
Mn(2)-O(11)	1.875(4)	O(11)-Dy(2)-O(7)	76.96(15)	O(13)-Mn(2)-Mn(4)	94.76(12)
Mn(2)-O(11)#1	2.040(4)	O(11)-Dy(2)-O(2)	142.22(17)	O(10)-Mn(2)-Mn(4)	132.20(12)
Mn(2)-O(13)	2.069(4)	O(7)-Dy(2)-O(2)	71.88(17)	Mn(1)-Mn(3)-Dy(1)	62.12(2)
Mn(3)-O(18)	2.086(4)	O(7)-Dy(2)-Mn(3)	150.16(11)	Mn(4)-Mn(3)-Dy(1)	120.85(3)
Mn(3)-O(12)#1	1.871(4)	O(2)-Dy(2)-Mn(3)	94.09(13)	O(18)-Mn(3)-Dy(2)	104.47(13)
Mn(3)-O(19)	1.886(4)	O(4)-Dy(2)-Mn(3)	92.05(12)	O(16)-Mn(4)-Mn(2)	116.74(15)
Mn(3)-O(16)	1.901(4)	O(2)-Dy(2)-Mn(1)	126.63(14)	Mn(2)-Mn(4)-Mn(3)	97.51(3)