Electronic Supplementary Material (ESI) for Dalton Transactions.

Table S1. Crystallographic data for complexes **3** and **4**.

	3·6MeCN·0.5H_2O	**4**·6MeCN·0.5H_2O
Formula	$C_{102}H_{115}Dy_2Mn_6N_{18}O_{42.5}$	$C_{102}H_{115}Er_2Mn_6N_{18}O_{42.5}$
M_W	2927.75	2937.27
Crystal System	Triclinic	Triclinic
Space group	P-1	P-1
a/Å	13.533(3)	13.514(4)
b/ Å	14.266(3)	14.234(5)
c/Å	16.867(4)	16.827(5)
α/°	91.18(2)	90.99(3)
β/°	108.18(2)	108.18(3)
γ/°	111.57(2)	111.68(4)
V/Å^3	2843.2(12)	2825.3(18)
Z	1	1
T/K	100	100
λ/Å	0.71073	0.71073
D_c/g cm^{-3}	1.710	1.726
μ(Mo-Ka)/ mm^{-1}	2.04	2.21
Meas./indep. (R_{int}) refl.	30039 / 15646 (0.036)	35579 /13963 (0.109)
Obs. refl. [$I>2\sigma(I)$]	12214	7765
$wR2$	0.110	0.097
$R1$	0.046	0.058
Goodness of fit on F^2	1.04	1.01
$\Delta\rho_{max,min}$/ eÅ^{-3}	2.26, -1.12	1.21, -0.86

Table S2. Selected bond distances (Å) and angles (°) for **3**.

Dy—O14A	2.277(3)	Mn2—O1B	1.889(3)
Dy—O15C	2.315(3)	Mn2—O14B	1.890(3)
Dy—O14B	2.390(2)	Mn2—O1	1.906(3)
Dy—O2	2.395(2)	Mn2—N1B	1.974(3)
Dy—O21	2.428(3)	Mn2—O1C	2.244(3)
Dy—O12	2.510(3)	Mn2—O13	2.436(3)
Dy—O1	2.519(3)	Mn3—O14C^{i}	1.882(3)
Dy—O11	2.535(3)	Mn3—O15C^{i}	1.902(3)
Dy—O22	2.547(3)	Mn3—O1	1.917(2)
Dy—N1	2.909(4)	Mn3—O2^{i}	1.959(2)
Dy—Mn3^{i}	3.4141(11)	Mn3—O1W	2.248(3)
Dy—Mn2	3.530(9)	Mn3—O2	2.336(3)
Mn1—O1A	1.873(3)	Mn1—O14C^{i}	2.162(2)
Mn1—O14A	1.874(3)	Mn1—O13	2.198(3)
Mn1—N1A	1.982(3)	Mn1—Mn3	3.1187(11)
Mn1—O1	2.104(2)		
Mn1—Mn3—Dy^{i}	174.95(2)	O14A—Dy—O15C	154.68(9)
Dy^{i}—Mn3—Dy	122.43(3)	O14A—Dy—O14B	96.96(9)
Mn2—O1—Mn3	133.90(14)	O15C—Dy—O14B	77.09(8)
Mn2—O1—Mn1	109.81(11)	O14A—Dy—O2	98.00(9)
Mn3—O1—Mn1	101.63(11)	O15C—Dy—O2	65.98(9)
Mn2—O1—Dy	101.70(11)	O14B—Dy—O2	121.07(9)
Mn3—O1—Dy	106.46(10)	O14A—Dy—O21	73.97(10)
Mn1—O1—Dy	97.94(10)	O15C—Dy—O21	129.41(10)
Mn3^{i}—O2—Mn3	102.88(11)	O14B—Dy—O21	87.30(9)
Mn3^{i}—O2—Dy	102.82(11)	O2—Dy—O21	151.50(9)
Mn3—O2—Dy	98.03(9)	O14A—Dy—O12	118.32(9)
O14A—Dy—O1	65.74(9)	O15C—Dy—O12	78.69(9)
O15C—Dy—O1	89.88(10)	O14B—Dy—O12	138.54(9)
O14B—Dy—O1	64.27(8)	O2—Dy—O12	77.22(9)
O2—Dy—O1	71.21(8)	O21—Dy—O12	82.71(9)

Symmetry codes: (i) -*x*+1, -*y*+1, -*z*+1.

Table S3. Selected bond distances (Å) and angles (°) for **4**.

Er—O14A	2.266(4)	Mn2—O14B	1.893(4)
Er—O15C	2.307(4)	Mn2—O1	1.901(4)
Er—O14B	2.353(4)	Mn2—O1B	1.909(4)
Er—O2	2.383(4)	Mn2—N1B	1.970(5)
Er—O21	2.408(4)	Mn2—O1C	2.243(4)
Er—O1	2.468(4)	Mn2—O13	2.435(4)
Er—O12	2.470(4)	Mn3—O14C^{i}	1.889(4)
Er—O11	2.510(4)	Mn3—O15C^{i}	1.912(4)
Er—O22	2.518(4)	Mn3—O1	1.929(4)
Er—N1	2.878(6)	Mn3—O2^{i}	1.952(4)
Er—Mn3^{i}	3.3933(16)	Mn3—O1W	2.241(4)
Er—Mn2	3.4228(15)	Mn3—O2	2.331(4)
Mn1—O14A	1.868(4)	Mn1—O14C^{i}	2.153(4)
Mn1—O1A	1.876(4)	Mn1—O13	2.193(4)
Mn1—N1A	1.984(5)	Mn1—Mn3	3.1212(17)
Mn1—O1	2.108(4)		
Mn1—Mn3—Er^{i}	175.25(4)	O14A—Er—O15C	154.99(14)
Er^{i}—Mn3—Er	122.33(4)	O14A—Er—O14B	97.21(14)
Mn2—O1—Mn3	132.8(2)	O15C—Er—O14B	76.84(13)
Mn2—O1—Mn1	109.55(16)	O14A—Er—O2	97.85(13)
Mn3—O1—Mn1	101.22(16)	O15C—Er—O2	66.72(13)
Mn2—O1—Er	102.36(16)	O14B—Er—O2	122.02(14)
Mn3—O1—Er	107.44(16)	O14A—Er—O21	74.03(15)
Mn1—O1—Er	98.73(15)	O15C—Er—O21	129.01(14)
Mn3^{i}—O2—Mn3	102.81(17)	O14B—Er—O21	87.21(15)
Mn3^{i}—O2—Er	102.57(15)	O2—Er—O21	150.67(14)
Mn3—O2—Er	98.03(13)	O14A—Er—O1	66.04(14)
O15C—Er—O1	89.81(14)	O14B—Er—O1	64.75(13)

Symmetry code: (i) -*x*+1, -*y*+1, -*z*+1.

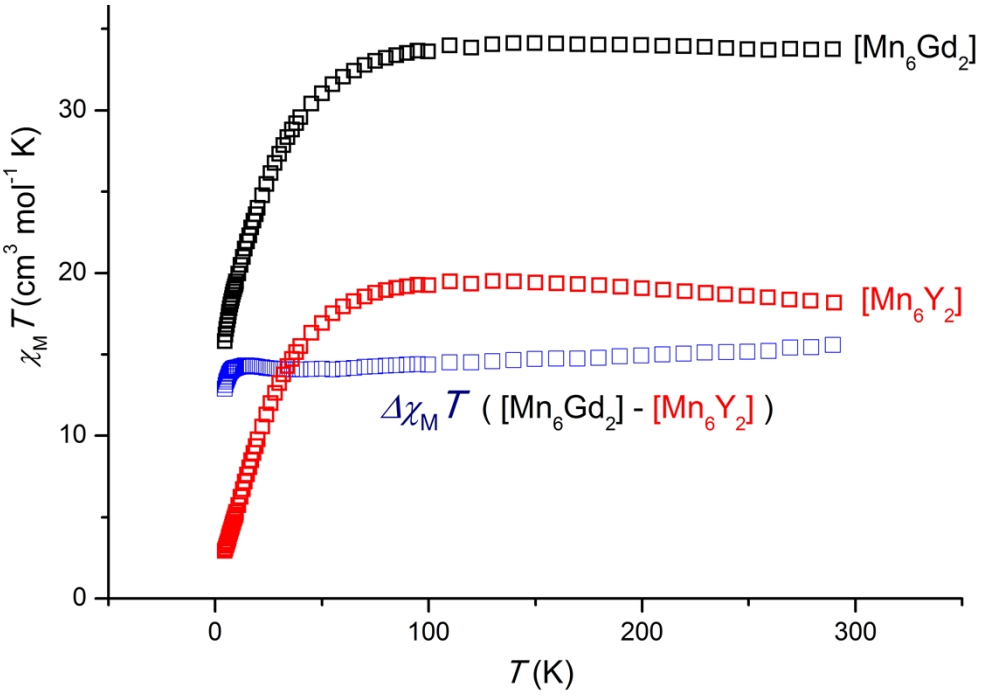

Figure S1. Plot of $\chi_M T$ *vs.* T and $\Delta\chi_M T$ *vs.* T for complexes **1** and **5**.