

## Supporting Information

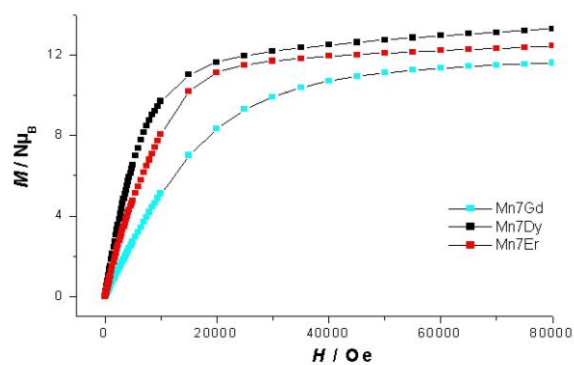


Figure S1. Plots of  $M/N\mu_B$  vs  $H$  for complexes 1-3 at 2K. Solid lines are eye guides.

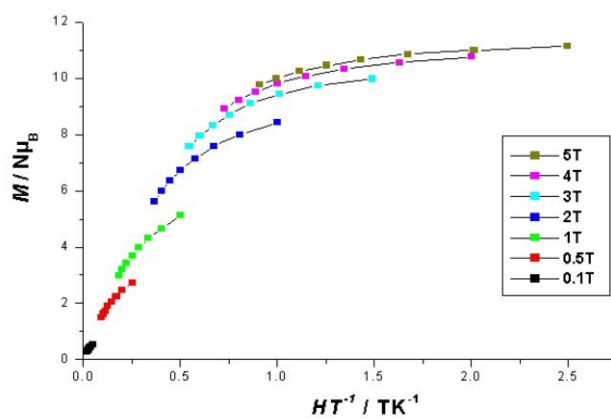


Figure S2. Plots of  $M/N\mu_B$  vs  $H/T$  for complex 1 (Mn<sub>7</sub>Gd) at the indicated applied fields. Solid lines are eye guides.

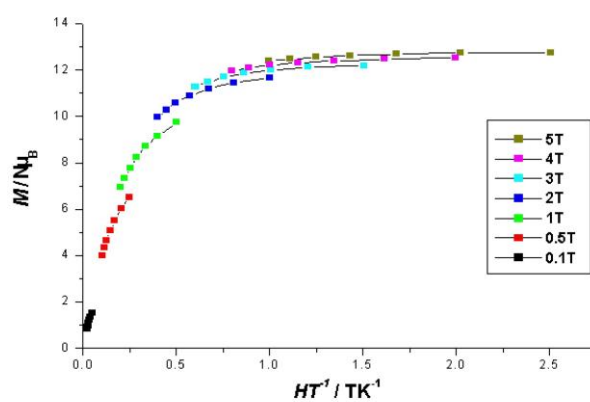
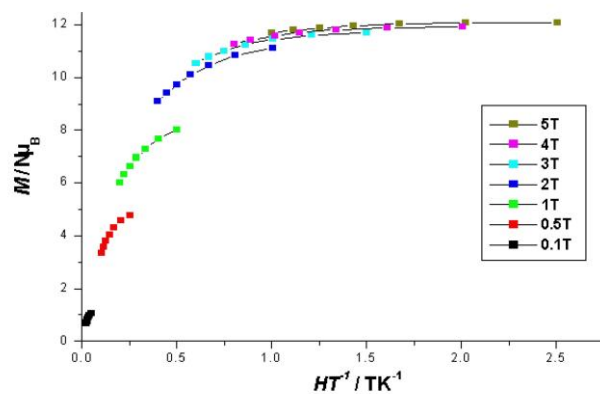
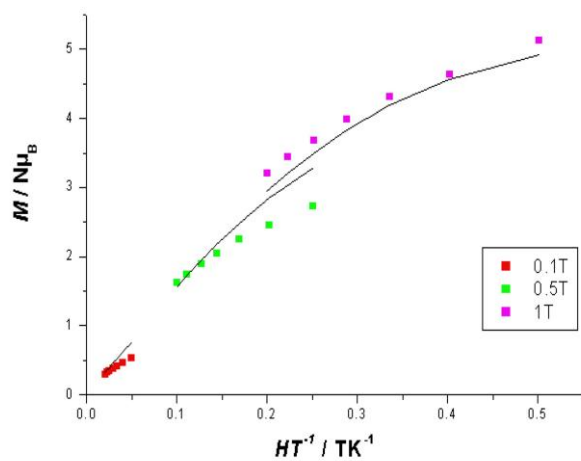


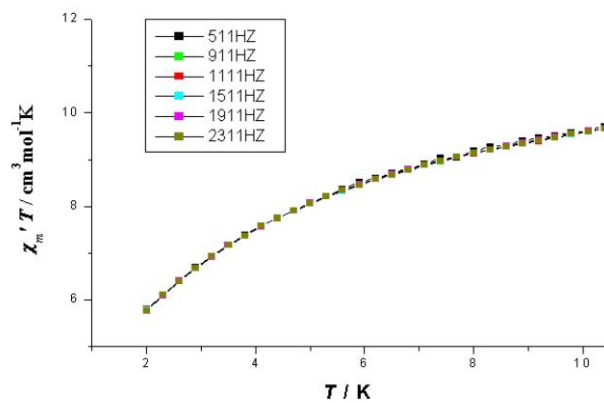
Figure S3. Plots of  $M/N\mu_B$  vs  $H/T$  for complex 2 (Mn<sub>7</sub>Dy) at the indicated applied fields. Solid lines are eye guides.



**Figure S4.** Plots of  $M/N\mu_B$  vs  $H/T$  for complex **3** ( $\text{Mn}_7\text{Er}$ ) at the indicated applied fields. Solid lines are eye guides.



**Figure S5.** Plots of  $M/N\mu_B$  vs  $H/T$  for complex **1** ( $\text{Mn}_7\text{Gd}$ ) at the indicated applied fields. Solid lines represent the best fits of the data.



**Figure S6.** In-phase ( $\chi'_m$ ) (as  $\chi'_m T$ ) ac susceptibility signals of complex **1** in a 3.0 G field oscillating at the indicated frequencies.

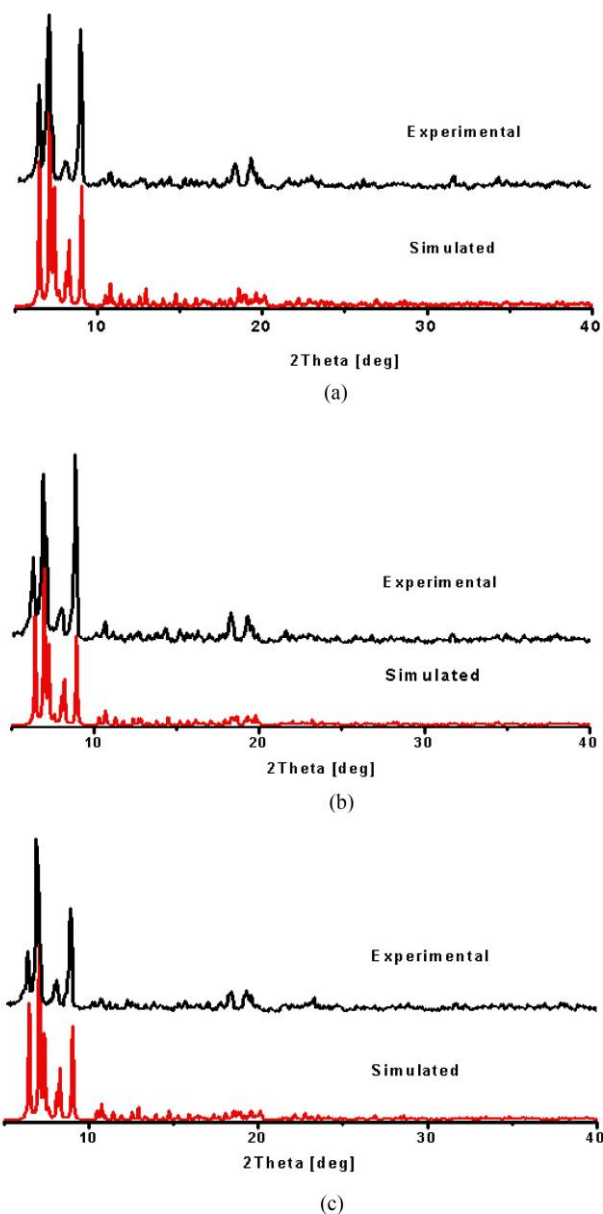


Figure S7. XRPD patterns for complexes 1-3

Table S1. Bond-Valence Sums for the O Atoms of Complex 3<sup>a</sup>.

atom	BVS	assignment	group	atom	BVS	assignment	group
O1	1.82	RO <sup>-</sup>	Piv <sup>-</sup>	O16	1.81	RO <sup>-</sup>	Piv <sup>-</sup>
O2	1.68	RO <sup>-</sup>	Piv <sup>-</sup>	O17	1.69	RO <sup>-</sup>	Mdea <sup>2-</sup>
O3	1.8	RO <sup>-</sup>	Piv <sup>-</sup>	O18	1.86	RO <sup>-</sup>	Mdea <sup>2-</sup>
O4	1.71	RO <sup>-</sup>	Piv <sup>-</sup>	O19	1.94	RO <sup>-</sup>	Piv <sup>-</sup>
O5	1.93	RO <sup>-</sup>	Mdea <sup>2-</sup>	O20	1.83	RO <sup>-</sup>	Piv <sup>-</sup>
O6	1.98	RO <sup>-</sup>	Mdea <sup>2-</sup>	O21	2.24	RO <sup>-</sup>	Mdea <sup>2-</sup>
O7	1.93	RO <sup>-</sup>	Piv <sup>-</sup>	O22	2.23	RO <sup>-</sup>	Mdea <sup>2-</sup>
O8	1.49	RO <sup>-</sup>	Piv <sup>-</sup>	O23	1.96	RO <sup>-</sup>	Piv <sup>-</sup>

O9	1.9	RO <sup>-</sup>	Piv <sup>-</sup>	O24	1.75	RO <sup>-</sup>	Piv <sup>-</sup>
O10	1.93	RO <sup>-</sup>	Piv <sup>-</sup>	O25	1.11	OH	OH
O11	1.85	RO <sup>-</sup>	Piv <sup>-</sup>	O26	0.98	OH	OH
O12	1.88	RO <sup>-</sup>	Piv <sup>-</sup>	O27	2.02	O <sup>2-</sup>	O <sup>2-</sup>
O13	1.87	RO <sup>-</sup>	MdeaH <sup>-</sup>	O28	0.99	OH	OH
O14	1.23	ROH	MdeaH <sup>-</sup>	O29	2.03	O <sup>2-</sup>	O <sup>2-</sup>
O15	2.06	RO <sup>-</sup>	Piv <sup>-</sup>				

<sup>a</sup> BVS values for O atoms of RO<sup>-</sup> and ROH groups are typically 1.8–2.0 and 1.0–1.2, respectively, but can be affected by hydrogen bonding.

Table S2. Bond-Valence Sums for the Mn<sup>a</sup> Atoms of Complex **3**.

atom	Mn <sup>2+</sup>	Mn <sup>3+</sup>	Mn <sup>4+</sup>
Mn1	3.2	<u>2.95</u>	2.92
Mn2	3.38	<u>3.12</u>	3.06
Mn3	<u>1.91</u>	1.76	1.73
Mn4	3.40	<u>3.14</u>	3.08
Mn5	3.38	<u>3.11</u>	3.06
Mn6	3.23	<u>2.98</u>	2.92
Mn7	3.34	<u>3.08</u>	3.02

<sup>a</sup>The underlined value is the closest to the charge for which it was calculated, and the nearest whole number can be taken as the oxidation state of the atom.