Supplementary Information

Structures and Magnetic Properties of $Mn^{III}_4Ln^{III}_4$ Aggregates with a "Square-in-Square" Topology

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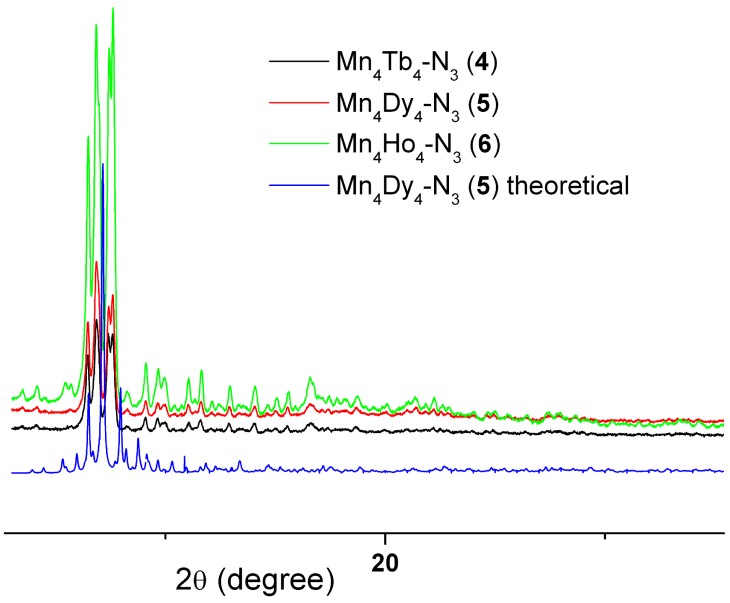

Figure S1. X-ray powder diffraction patterns measured at room temperature for complexes **4, 5** and **6**, and calculated powder diffraction pattern for **5** from the data measured at 100 K.

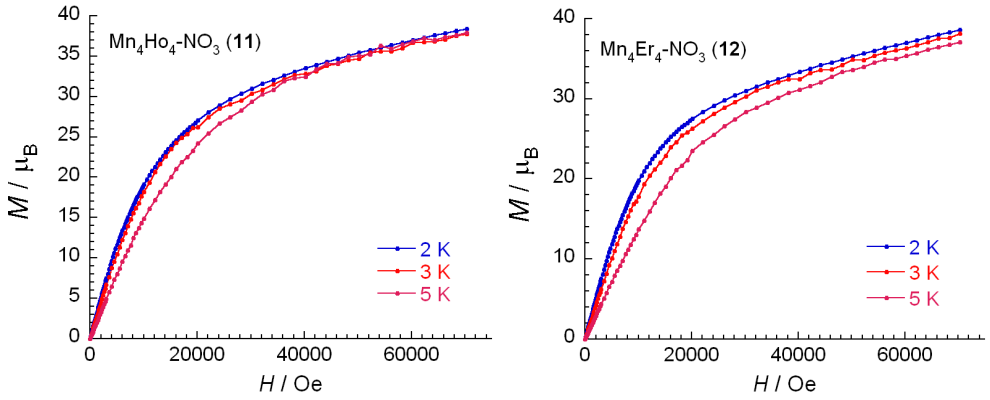

Figure S2. Field dependence of magnetization at low temperatures for all compounds. Compound codes are indicated in the inset of figures.

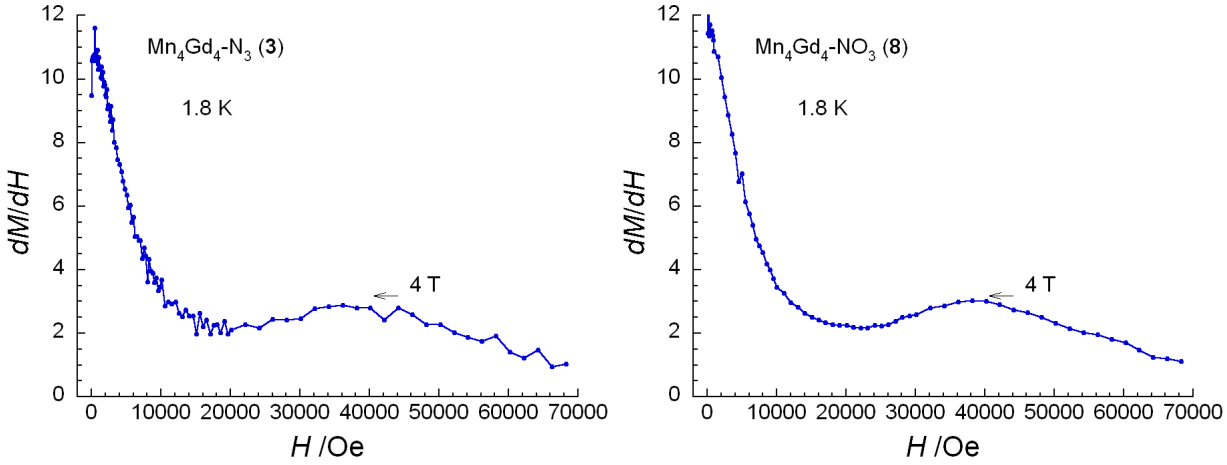

Figure S3. *dM/dH* vs *H* for **3** and **8** obtained at 1.8 K.

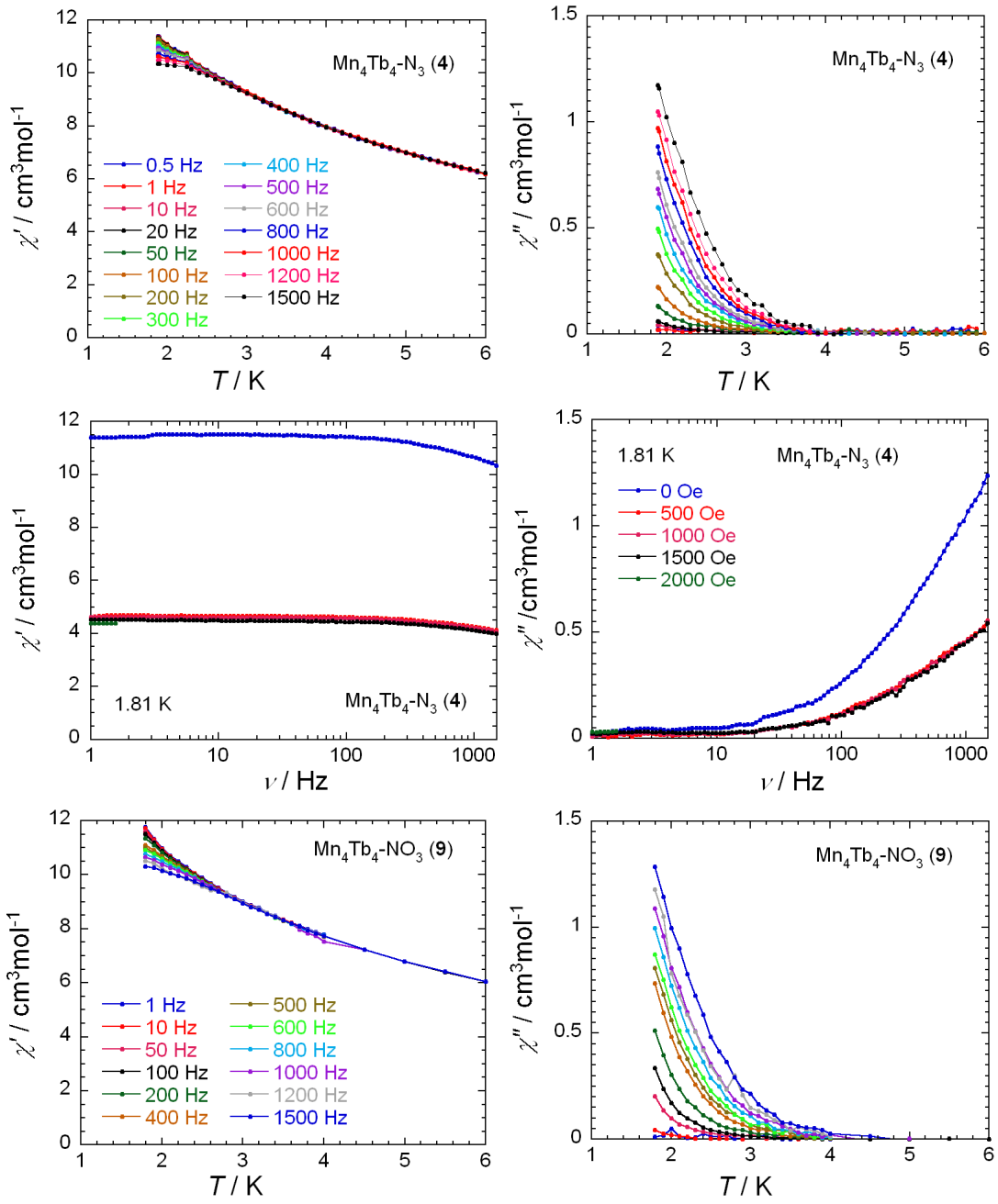

Figure S4. ac data for Mn_4Tb_4 analogues (**4** and **9**).

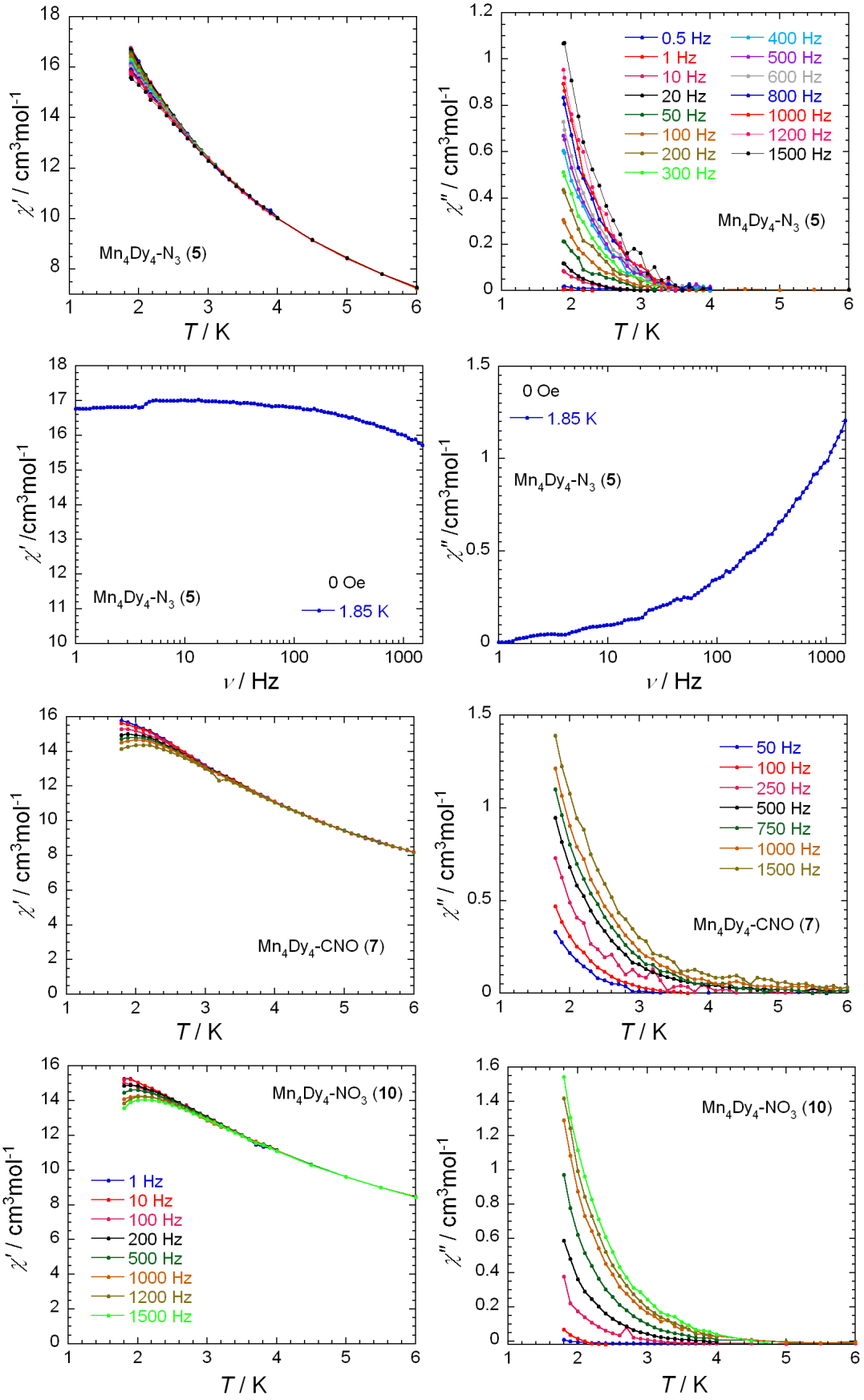

Figure S5. ac data for Mn_4Dy_4 analogues (**5**, **7** and **10**).

Table S1 Selected bond distances (Å) and angles (deg) for complex **7**

Bond lengths

Dy1—O1^{i}	2.324(3)	Dy1—N2^{ii}	2.509(3)
Dy1—O1	2.333(3)	Mn1—O3	1.893(3)
Dy1—O2	2.354(3)	Mn1—O2	1.912(3)
Dy1—O3^{i}	2.372(3)	Mn1—O4	1.940(3)
Dy1—O5	2.374(3)	Mn1—O6	1.945(3)
Dy1—O7^{i}	2.387(3)	Mn1—O1	2.163(3)
Dy1—N2	2.494(3)	Mn1—N1	2.476(3)
		Dy1—Dy1^{i}	3.9575(3)

Bond angles

Mn1—O3—Dy1^{ii}	104.13(11)	Mn1—O1—Dy1^{ii}	97.58(10)
Dy1—N2—Dy1^{i}	104.57(11)	Mn1—O1—Dy1	97.78(10)
Mn1—O2—Dy1	104.74(11)	Dy1^{ii}—O1—Dy1	116.38(11)
		Dy1^{i}—Dy1—Dy1^{ii}	90.000

(i) -0.25+y, 1.25-x, 1.75-z; (ii) 0.25-y, 0.25+x, 1.75-z.

Table S2 Selected bond distances (Å) and angles (deg) for complex **10**

Bond lengths

Dy1—O4	2.324(5)	Dy4—O26	2.362(7)
Dy1—O1	2.324(5)	Dy4—O24	2.385(6)
Dy1—O5	2.333(5)	Dy4—O38	2.402(5)
Dy1—O12	2.348(5)	Dy4—O35	2.459(5)
Dy1—O28	2.384(5)	Mn1—O6	1.905(5)
Dy1—O14	2.397(5)	Mn1—O5	1.898(5)
Dy1—O38	2.435(5)	Mn1—O13	1.949(5)
Dy1—O29	2.452(5)	Mn1—O15	1.934(5)
Dy2—O6	2.376(5)	Mn1—O1	2.144(5)
Dy2—O7	2.381(6)	Mn1—N1	2.458(6)
Dy2—O2	2.332(5)	Mn2—O8	1.887(6)
Dy2—O1	2.321(5)	Mn2—O7	1.896(6)
Dy2—O16	2.389(6)	Mn2—O19	1.956(6)
Dy2—O18	2.391(6)	Mn2—O17	1.928(7)
Dy2—O32	2.425(5)	Mn2—O2	2.141(5)
Dy2—O29	2.430(5)	Mn2—N2	2.491(9)
Dy3—O2	2.335(5)	Mn3—O9	1.901(5)
Dy3—O9	2.363(5)	Mn3—O10	1.891(6)
Dy3—O8	2.341(5)	Mn3—O21	1.942(7)
Dy3—O3	2.320(5)	Mn3—O23	1.947(6)
Dy3—O20	2.387(6)	Mn3—O3	2.136(5)
Dy3—O22	2.395(6)	Mn3—N3	2.473(7)
Dy3—O35	2.430(5)	Mn4—O12	1.899(6)
Dy3—O32	2.432(5)	Mn4—O11	1.901(6)
Dy4—O3	2.322(6)	Mn4—O25	1.960(7)
Dy4—O11	2.369(6)	Mn4—O27	1.956(6)
Dy4—O4	2.322(5)	Mn4—O4	2.134(5)
Dy4—O10	2.358(5)	Mn4—N4	2.456(7)

Bond angles

Mn1—O1—Dy1	97.3(2)	Dy4— O3 —Dy3	115.9(2)
Mn1—O1—Dy2	98.70(19)	Dy4— O4 —Dy1	115.3(2)
Dy1—O1—Dy2	116.1(2)	Mn1—O5—Dy1	104.5(2)
Mn2—O2—Dy3	97.4(2)	Mn1—O6—Dy2	104.1(2)
Mn2—O2—Dy2	98.2(2)	Mn2—O7—Dy2	104.1(2)
Dy3—O2—Dy2	115.9(2)	Mn2—O8—Dy3	104.9(2)
Mn3—O3—Dy4	98.2(2)	Mn3—O9—Dy3	104.2(2)
Mn3—O3—Dy3	98.5(2)	Mn3—O10—Dy4	104.5(2)
Dy4—O3—Dy3	115.9(2)	Mn4—O11—Dy4	103.6(2)
Mn4—O4—Dy4	98.11(19)	Mn4—O12—Dy1	104.5(2)
Mn4—O4—Dy1	98.1(2)	Dy1—Dy2—Dy3	89.762(10)
Dy4—O4—Dy1	115.3(2)	Dy2—Dy3—Dy4	89.797(11)
Dy2— O1 —Dy1	116.1(2)	Dy3—Dy4—Dy1	90.291(10)
Dy2— O2 —Dy3	115.9(2)	Dy4—Dy1—Dy2	90.143(11)