Supporting Information for

A Family of Binuclear Dysprosium(III)-Radical Compounds with Magnetic Relaxation in ON and OFF States

Tian Han, Wei Shi,* Xiao-Ping Zhang, Lei-Lei Li and Peng Cheng*

Department of Chemistry and Key Laboratory of Advanced Energy Materials Chemistry (MOE) and TKL of Metal and Molecule Based Material Chemistry, Nankai University, Tianjin 300071, P. R. China

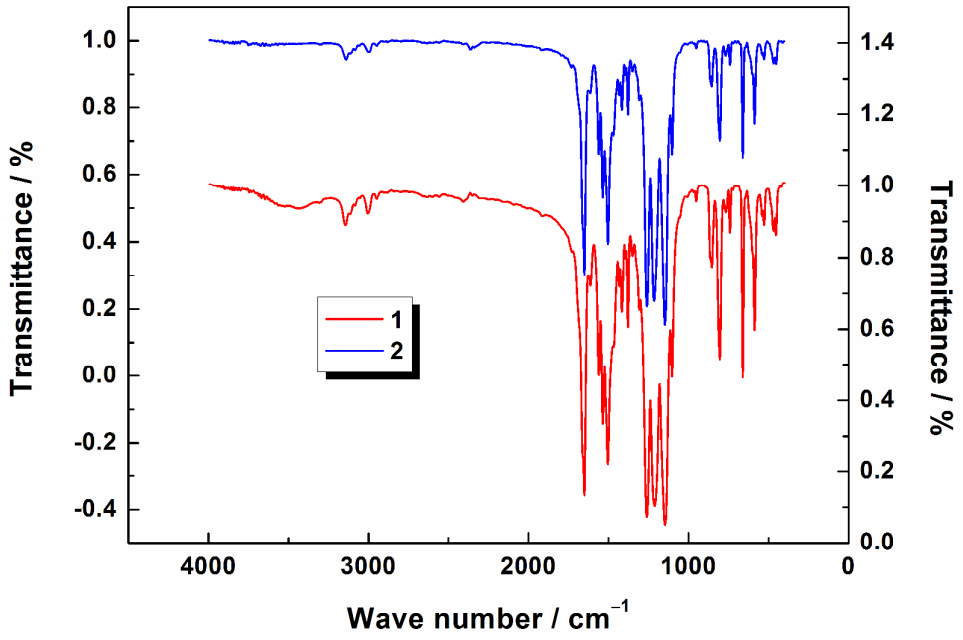

Figure S1. The IR spectra of compounds **1** and **2**.

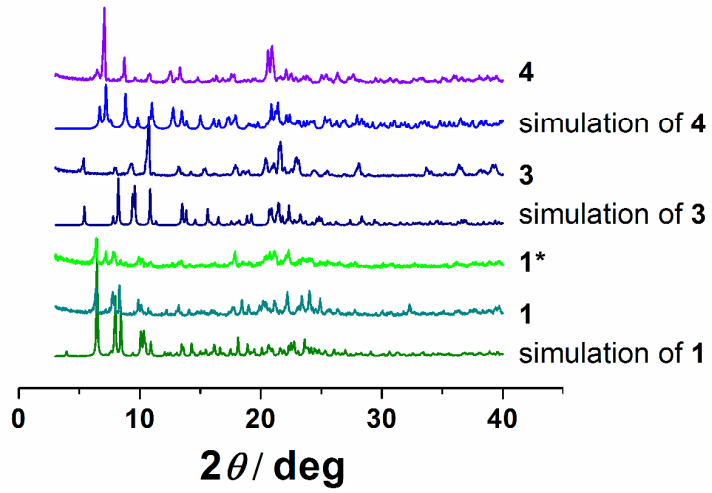

Figure S2. XRPD patterns of simulation from single-crystal diffraction data, **1**, **1***, **3** and **4**.

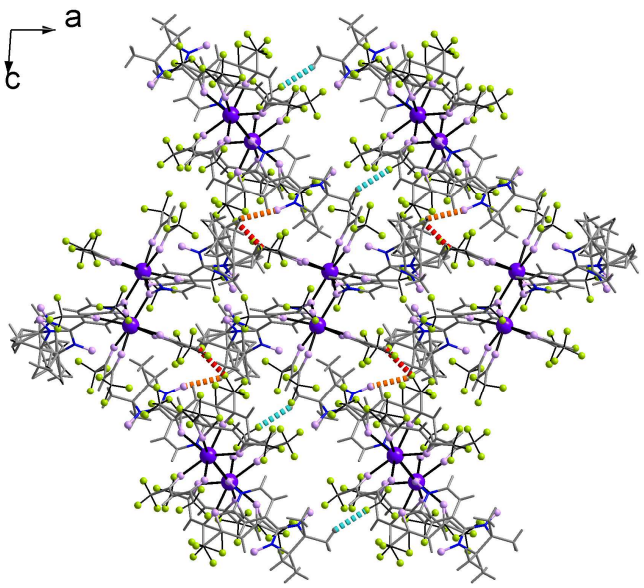

Figure S3. 3D packing diagram of **1** through the weak intermolecular interactions: C_{12B}–H_{12E}···O_{17} (orange), C_{19}–H_{19}···F_{25A} (red) and C_{38}–H_{38B}···F_{33} (aqua) with the distances of 3.05, 3.33 and 3.39 Å, respectively.

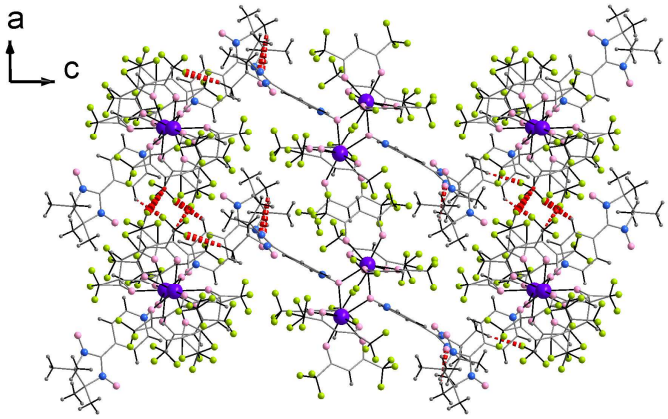

Figure S4. 3D packing diagram of **2**·2CH_2Cl_2 through weak C–H···O and C–H···F intermolecular hydrogen bonds: C_2–H_2···F_6, C_9–H_{9C}···O_{17}, C_{19A}–H_{19A}···F_{12A}, C_{37}–H_{37C}···F_4, C_{55}–H_{55A}···O_{18} and C_{55}–H_{55B}···O_9 with the distances of 3.09, 3.42, 3.31, 3.41, 3.16 and 3.30 Å, respectively.

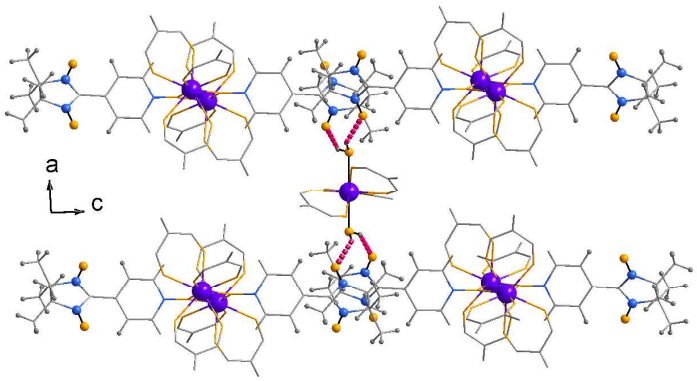

Figure S5. 3D packing diagram of **3** through classic O–H···O intermolecular hydrogen bonds.

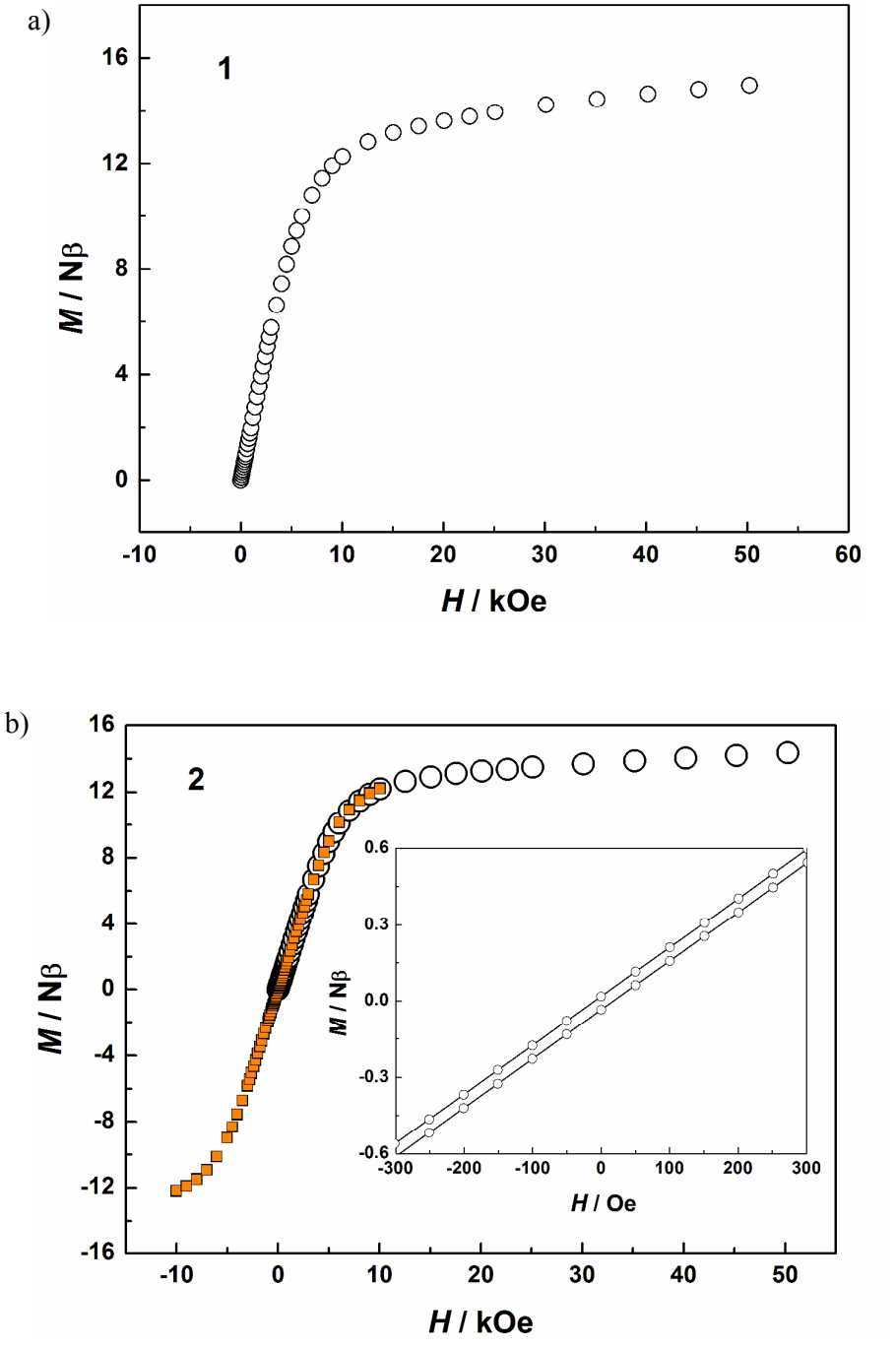

Figure S6. a) Magnetization versus field of **1** at 2 K. b) Magnetization versus field (circle) and magnetic hysteresis loop (square) of **2** at 2 K. In the inset, we show the enlargement of the low field region.

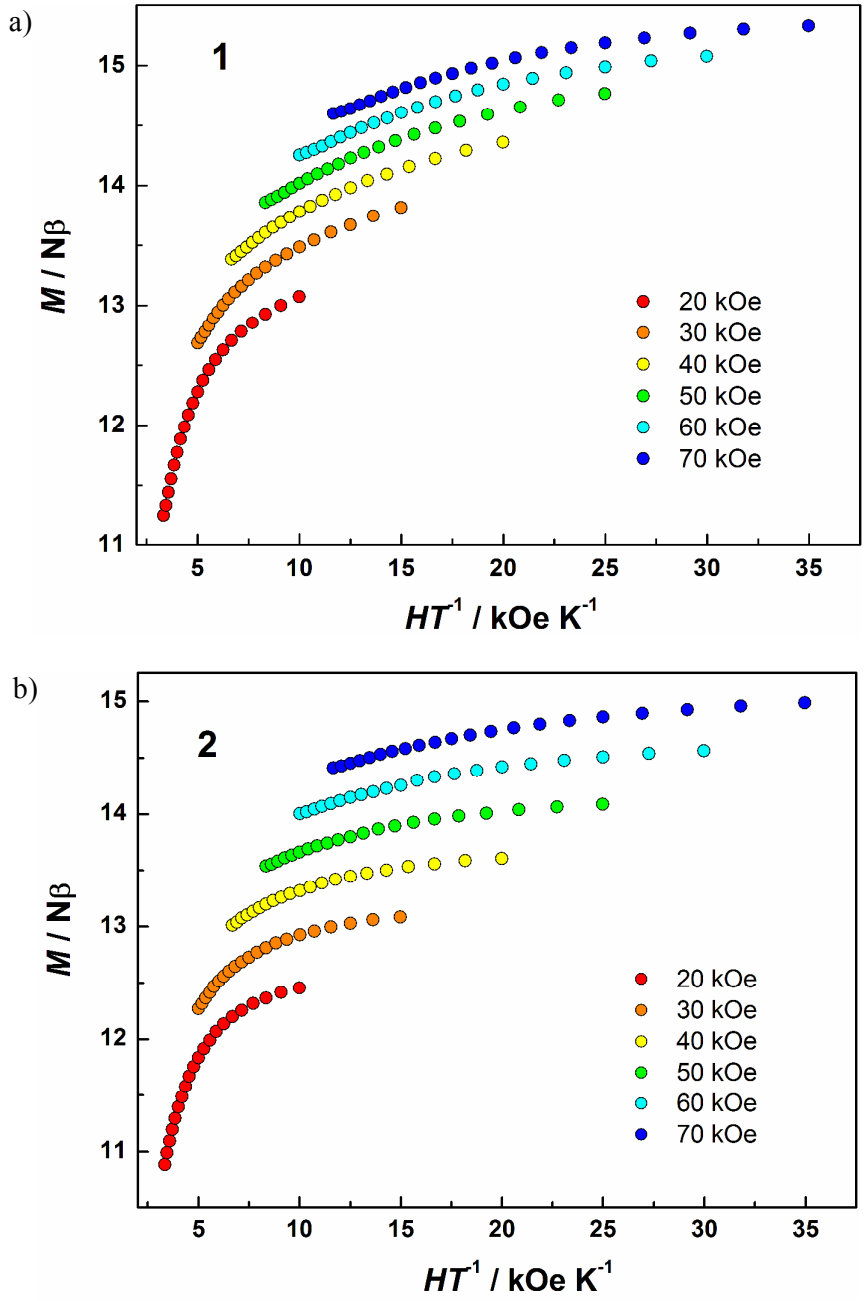

Figure S7. *M*/Nβ versus HT^{-1} plots at various magnetic fields for **1** (a) and **2** (b).

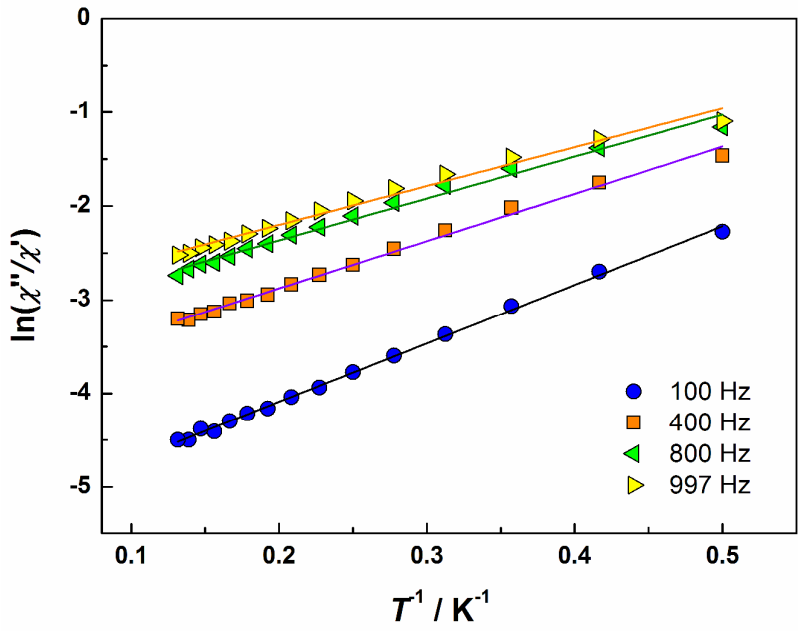

Figure S8. Plot of ln(χ''/χ') *vs.* 1/*T* for compound **2**. The solid line represents the fitting results.

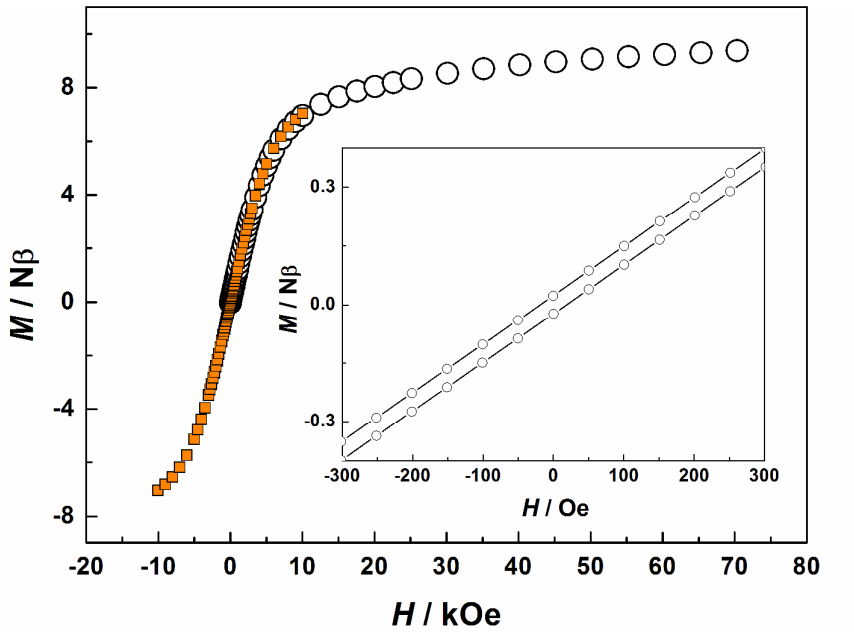

Figure S9. Magnetization versus field (circle) and magnetic hysteresis loop (square) of **3** at 2 K. In the inset, we show the enlargement of the low field region.

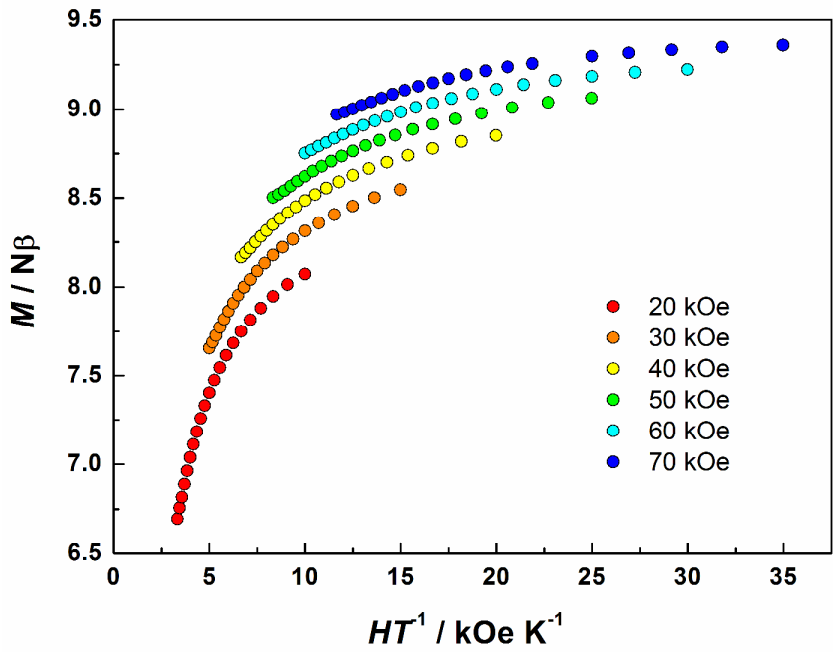

Figure S10. *M*/Nβ versus HT^{-1} plots at various magnetic fields for **3**.

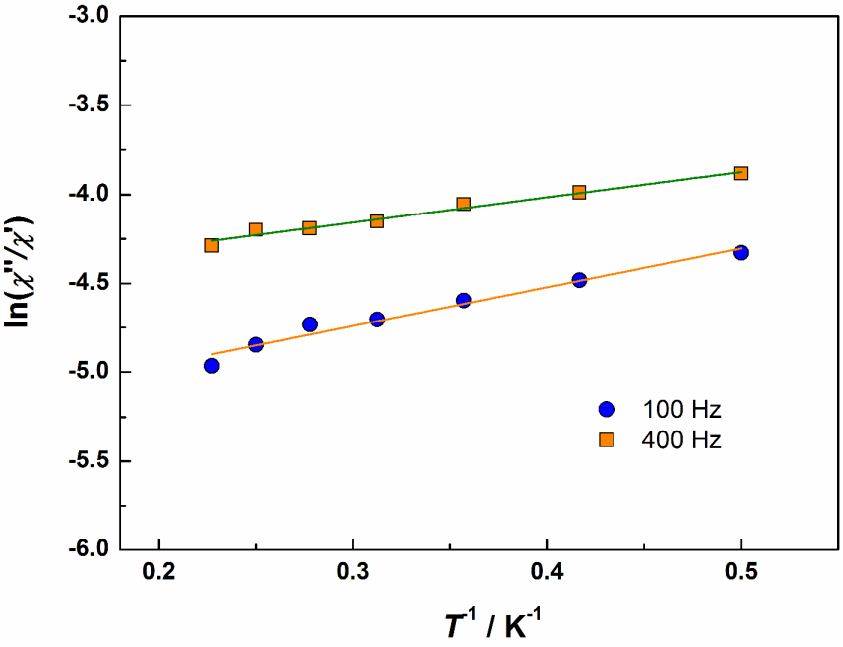

Figure S11. Plot of ln(χ''/χ') *vs.* 1/*T* for compound **3**. The solid line represents the fitting results.

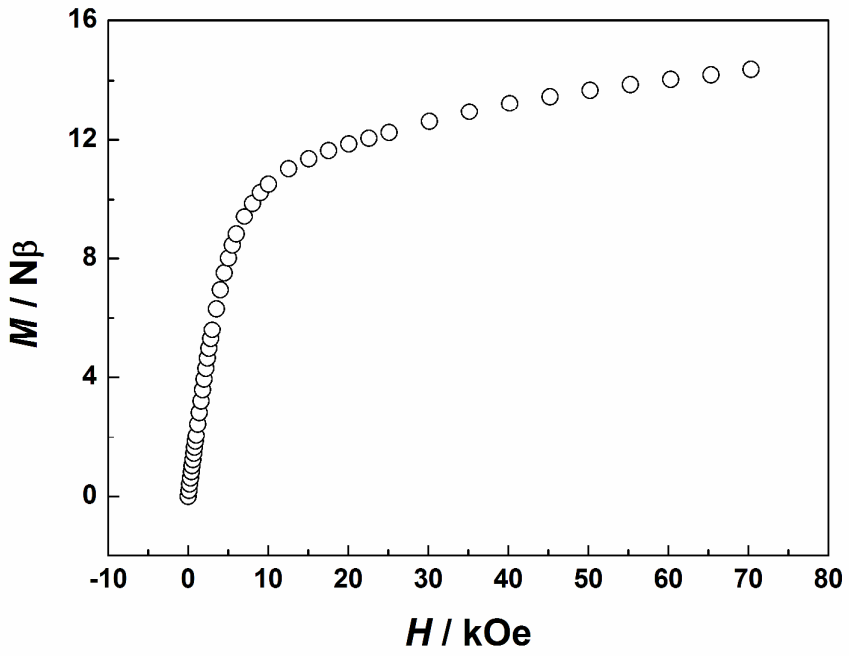

Figure S12. Magnetization versus field at 2 K for **4**.

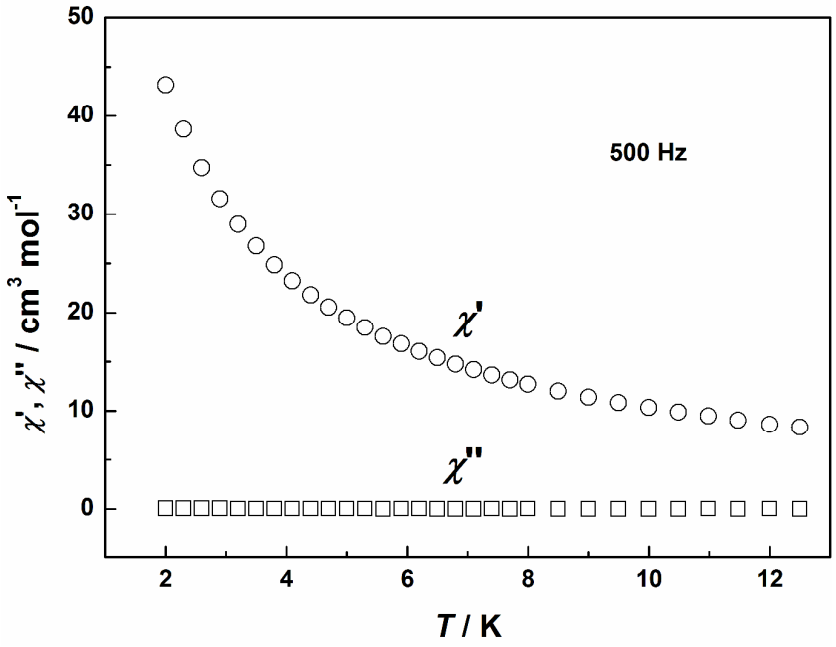

Figure S13. Temperature dependence of the imaginary χ'' and real χ' components of the *ac* susceptibility measured in zero applied field for **4**.

Table S1. Selected bonds and angles for **1** and **2**·$2CH_2Cl_2$. Symmetry code: #1 -x+1,-y,-z+1; #2 -x,-y+2,-z; #3 -x,-y+1,-z+2; #4 -x,-y+1,-z+1.

1				**2**·$2CH_2Cl_2$			
Bonds/Angles	Å / º	Bonds/Angles	Å / º	Bonds/Angles	Å / º	Bonds/Angles	Å / º
Dy1-O5	2.356(5)	Dy2-O13	2.312(5)	Dy1-O3	2.348(5)	Dy2-O11	2.334(6)
Dy1-O7	2.347(4)	Dy2-O10	2.395(5)	Dy1-O4	2.337(6)	Dy2-O10	2.415(5)
Dy1-O6	2.316(5)	Dy2-O10#2	2.379(5)	Dy1-O5	2.325(6)	Dy2-O10#4	2.399(5)
Dy1-O4	2.314(5)	Dy2-O14	2.350(5)	Dy1-O2	2.331(5)	Dy2-O12	2.334(6)
Dy1-O1	2.396(5)	Dy2-O11	2.360(5)	Dy1-O1	2.370(5)	Dy2-O16	2.325(6)
Dy1-O2	2.331(5)	Dy2-O16	2.333(5)	Dy1-O7	2.325(6)	Dy2-O15	2.347(6)
Dy1-O3	2.348(5)	Dy2-O15	2.306(6)	Dy1-O6	2.373(6)	Dy2-O14	2.363(6)
Dy1-O1#1	2.400(4)	Dy2-O12	2.315(6)	Dy1-O1#3	2.444(5)	Dy2-O13	2.323(5)
O1-Dy1-O1#1	63.63(17)	O10#2-Dy2-O10	64.52(18)	O1-Dy1-O1#3	63.81(19)	O10#4-Dy2-O10	63.03(19)
O6-Dy1-O7	72.89(16)	O12-Dy2-O11	73.5(2)	O2-Dy1-O3	71.54(18)	O12-Dy2-O11	71.8(2)
O2-Dy1-O3	71.53(17)	O13-Dy2-O14	72.94(19)	O5-Dy1-O6	72.4(2)	O13-Dy2-O14	72.57(19)
O4-Dy1-O5	71.87(16)	O15-Dy2-O16	72.3(2)	O7-Dy1-O4	73.7(2)	O16-Dy2-O15	71.9(2)
O5-Dy1-O1	76.82(16)	O16-Dy2-O10	75.54(18)	O3-Dy1-O1	72.27(18)	O15-Dy2-O10	86.77(19)
O4-Dy1-O1#1	78.78(16)	O15-Dy2-O10#2	81.15(19)	O2-Dy1-O1#3	81.97(18)	O14-Dy2-O10#4	79.78(17)

Table S2. Selected bonds and angles for **3** and **4**. Symmetry code: #1 -x-1,y,-z-1/2; #2 -x-1/2,-y+1/2,-z; #3 -x,-y-1,-z.

3				**4**	
Bonds/Angles	Å / º	Bonds/Angles	Å / º	Bonds/Angles	Å / º
Dy1-O1	2.385(5)	Dy2-O10	2.300(5)	Dy1-O1	2.425(3)
Dy1-O2	2.329(5)	Dy2-O10#1	2.300(5)	Dy1-O4	2.345(4)
Dy1-O3	2.334(5)	Dy2-O11	2.403(5)	Dy1-O5	2.386(4)
Dy1-O4	2.336(5)	Dy2-O11#1	2.403(5)	Dy1-O6	2.383(4)
Dy1-O5	2.312(5)	Dy2-O12	2.373(5)	Dy1-O7	2.358(4)
Dy1-O6	2.335(5)	Dy2-O12#1	2.373(5)	Dy1-O8	2.393(4)
Dy1-O7	2.369(4)	Dy2-O13	2.332(5)	Dy1-O9	2.457(4)
Dy1-O1#2	2.412(5)	Dy2-O13#1	2.332(5)	Dy1-O10	2.446(4)
O1-Dy1-O1#2	62.87(17)	O10-Dy2-O11	72.25(18)	Dy1-O1#3	2.509(4)
O6-Dy1-O7	71.21(16)	O12-Dy2-O12#1	72.1(2)	O1-Dy1-O1#3	60.15(14)
O2-Dy1-O3	70.67(17)	O10#1-Dy2-O11#1	72.25(18)	O4-Dy1-O5	70.81(14)
O4-Dy1-O5	72.51(18)	O13-Dy2-O13#1	147.8(3)	O6-Dy1-O7	72.81(14)
O3-Dy1-O1	80.92(18)	O11-Dy2-O13	71.06(18)	O8-Dy1-O9	68.77(13)
O4-Dy1-O1#2	74.61(16)	O12-Dy2-O13#1	73.40(18)	O10-Dy1-O1	77.20(13)

Table S3. δ and φ values for compound **3**.

	3		DD	TP	SAP
δ_1	O1#2 [O1 O2] O3	20.26	29.5	0.0	0.0
δ_2	O4 [O2 O5] O3	35.35	29.5	21.8	0.0
δ_3	O1#2 [O1 O6] O7	39.03	29.5	48.2	52.4
δ_4	O4 [O5 O6] O7	30.39	29.5	48.2	52.4
φ_1	O1-O5-O1#2-O4	6.64	0	14.1	24.5
φ_2	O2-O6-O3-O7	5.97			

A [B C] D is the dihedral angle between the ABC plane and the BCD plane. A-B-C-D is the dihedral angle between the (AB)CD plane and the AB(CD) where (AB) signifies the midpoint of the AB edge. Symmetry code: #2 -x-1/2,-y+1/2,-z.