Supplementary Information

Synthesis, structural characterization and magnetic properties study of Cr(III)/Ln(III), Ln=Gd and Dy complexes

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Tables S1. Main bond distances (Å) and angles (°) of complexes **1**-**3**.

1			**2**			**3**		
Gd1	O5	2.346(8)	Dy1	O27	2.38(1)	Y1	O5	2.29(2)
Gd1	O4	2.425(9)	Dy1	O21	2.370(9)	Y1	O11	2.35(2)
Gd1	O11	2.40(1)	Dy1	O20	2.359(7)	Y1	O22	2.35(2)
Gd1	O22	2.397(8)	Dy1	O4	2.422(7)	Y1	O10	2.34(3)
Gd1	O10	2.35(1)	Dy1	O10	2.315(9)	Y1	O4	2.34(2)
Gd1	O3	2.434(8)	Dy1	O1	2.390(8)	Y1	O23	2.37(2)
Gd1	O23	2.41(1)	Dy1	O5	2.322(7)	Y1	O2	2.37(2)
Gd1	O1	2.417(9)	Dy1	O2	2.400(8)	Y1	O5	2.29(2)
Gd2	O6	2.354(9)	Dy2	O2	2.377(7)	Y2	O14	2.37(3)
Gd2	O4	2.413(8)	Dy2	O1	2.402(8)	Y2	O24	2.39(3)
Gd2	O14	2.43(1)	Dy2	O3	2.394(7)	Y2	O15	2.37(3)
Gd2	O1	2.42(1)	Dy2	O13	2.386(9)	Y2	O2	2.38(2)
Gd2	O2	2.416(8)	Dy2	O7	2.323(8)	Y2	O6	2.29(2)
Gd2	O7	2.35(1)	Dy2	O6	2.333(8)	Y2	O7	2.33(2)
Gd2	O15	2.40(1)	Dy2	O12	2.406(8)	Y2	O15	2.37(3)
Gd2	O24	2.38(1)	Dy2	O23	2.35(1)	Y2	O3	2.37(2)
Gd3	O1	2.457(8)	Dy3	O1	2.418(7)	Y3	O1	2.40(2)
Gd3	O3	2.415(8)	Dy3	O4	2.386(8)	Y3	O4	2.38(2)
Gd3	O19	2.420(8)	Dy3	O9	2.35(1)	Y3	O9	2.35(3)
Gd3	O9	2.37(1)	Dy3	O17	2.388(7)	Y3	O19	2.38(2)
Gd3	O26	2.40(1)	Dy3	O25	2.35(1)	Y3	O26	2.35(3)
Gd3	O2	2.432(9)	Dy3	O16	2.388(8)	Y3	O18	2.36(2)
Gd3	O8	2.345(8)	Dy3	O3	2.405(8)	Y3	O3	2.38(2)
Gd3	O18	2.422(9)	Dy3	O8	2.316(7)	Y3	O8	2.30(2)

Cr1	O13	1.99(1)	Cr1	N1	2.09(1)	Cr1	N1	2.09(3)
Cr1	O4	1.942(9)	Cr1	O22	1.979(9)	Cr1	O6	1.97(2)
Cr1	O5	1.96(1)	Cr1	O5	1.956(8)	Cr1	O5	1.97(3)
Cr1	O12	2.01(1)	Cr1	O2	1.937(7)	Cr1	O13	2.01(2)
Cr1	O6	1.943(9)	Cr1	O6	1.943(8)	Cr1	O2	1.95(2)
Cr1	N1	2.09(1)	Cr1	O11	1.976(8)	Cr1	O12	1.98(2)
Cr2	O2	1.951(9)	Cr2	O7	1.958(7)	Cr2	N2	2.11(3)
Cr2	O7	1.952(8)	Cr2	O3	1.949(8)	Cr2	O16	1.98(3)
Cr2	O8	1.95(1)	Cr2	O8	1.953(9)	Cr2	O7	2.01(2)
Cr2	O17	1.988(9)	Cr2	O15	1.984(8)	Cr2	O3	2.00(2)
Cr2	O16	2.00(1)	Cr2	O14	1.98(1)	Cr2	O8	1.99(3)
Cr2	N2	2.10(1)	Cr2	N2	2.08(1)	Cr2	O17	2.01(2)
Cr3	O3	1.95(1)	Cr3	O10	1.974(7)	Cr3	N3	2.04(4)

Cr3	O10	1.976(8)	Cr3	O19	1.972(9)	Cr3	O21	1.98(2)
Cr3	O21	1.99(1)	Cr3	N3	2.10(2)	Cr3	O10	1.95(2)
Cr3	N3	2.10(2)	Cr3	O18	1.979(7)	Cr3	O4	1.99(3)
Cr3	O20	1.969(8)	Cr3	O9	1.964(8)	Cr3	O9	1.95(2)
Cr3	O9	1.955(9)	Cr3	O4	1.93(1)	Cr3	O20	1.98(2)

Gd1	Cr1	3.353(2)	Dy1	Dy2	3.952(1)	Y1	Cr3	3.315(7)
Gd1	Gd3	4.0160(7)	Dy1	Dy3	3.966(1)	Y1	Cr1	3.322(5)
Gd1	Cr3	3.359(2)	Dy1	Cr1	3.327(2)	Y2	Y1	3.937(4)
Gd2	Cr2	3.351(2)	Dy1	Cr3	3.326(2)	Y2	Y3	3.983(4)
Gd2	Gd1	3.9993(7)	Dy2	Dy3	3.997(1)	Y2	Cr2	3.312(6)
Gd2	Cr1	3.372(2)	Dy2	Cr1	3.345(2)	Y2	Cr1	3.327(6)
Gd3	Cr3	3.367(2)	Dy2	Cr2	3.321(2)	Y3	Y1	3.943(3)
Gd3	Gd2	4.0451(7)	Dy3	Cr2	3.325(2)	Y3	Cr3	3.332(6)
Gd3	Cr2	3.350(2)	Dy3	Cr3	3.341(2)	Y3	Cr2	3.313(5)

1				**2**				**3**			
O13	Cr1	O12	87.6(4)	N1	Cr1	O22	94.3(4)	N1	Cr1	O6	85(1)
N1	Cr1	O12	94.4(5)	O5	Cr1	O22	90.1(4)	N1	Cr1	O13	94(1)
O5	Cr1	O12	90.2(4)	O2	Cr1	O22	96.0(4)	O2	Cr1	O6	85.9(9)
O4	Cr1	O12	95.0(4)	O11	Cr1	O22	87.8(4)	O5	Cr1	O6	92(1)
O13	Cr1	O6	90.0(4)	O11	Cr1	O6	90.0(4)	O13	Cr1	O12	88(1)
O4	Cr1	O6	86.4(4)	O2	Cr1	O6	86.0(3)	N1	Cr1	O12	94(1)
N1	Cr1	O6	84.6(5)	O5	Cr1	O6	92.0(4)	O5	Cr1	O12	90(1)
O5	Cr1	O6	92.1(4)	N1	Cr1	O6	84.1(4)	O13	Cr1	O6	90(1)
N2	Cr2	O7	84.8(4)	O10	Cr3	O19	90.7(4)	O10	Cr3	O21	91(1)
N2	Cr2	O16	94.0(4)	N3	Cr3	O19	94.7(5)	N3	Cr3	O21	95(1)
N2	Cr2	O17	93.9(4)	O18	Cr3	O19	87.9(4)	O20	Cr3	O21	88(1)
N2	Cr2	O8	84.3(4)	O4	Cr3	O19	94.7(4)	O4	Cr3	O21	95(1)
O17	Cr2	O2	94.5(4)	O10	Cr3	O9	91.2(4)	O10	Cr3	O9	91(1)
O16	Cr2	O2	94.3(4)	O4	Cr3	O9	87.0(4)	O4	Cr3	O9	87.2(9)
O7	Cr2	O2	87.2(4)	O18	Cr3	O9	90.1(4)	O20	Cr3	O9	90(1)
O8	Cr2	O2	87.8(4)	N3	Cr3	O9	83.9(5)	N3	Cr3	O9	83(1)
O10	Cr3	O21	90.7(4)	O3	Cr2	O14	94.6(3)	O8	Cr2	O17	89(1)
O3	Cr3	O21	94.8(4)	O15	Cr2	O14	86.3(4)	N2	Cr2	O17	97(1)
N3	Cr3	O21	94.6(5)	N2	Cr2	O14	94.7(4)	O3	Cr2	O17	93.3(9)
O20	Cr3	O21	87.5(4)	O7	Cr2	O14	91.1(3)	O16	Cr2	O17	88(1)
O3	Cr3	O9	87.2(4)	O3	Cr2	O8	87.5(3)	O7	Cr2	O3	87.7(9)
O20	Cr3	O9	90.3(4)	O7	Cr2	O8	92.4(3)	O7	Cr2	O16	89(1)
N3	Cr3	O9	83.8(5)	N2	Cr2	O8	83.7(4)	O7	Cr2	N2	82(1)
O10	Cr3	O9	91.4(4)	O15	Cr2	O8	90.2(4)	O8	Cr2	O7	94(1)

Table S2. *Ab-initio* (MOLCAS-SINGLE_ANISO) computed single ion parameters of Cr(III) sites in complex **2**.

	Cr(1)	**Cr(2)**	**Cr(3)**
g (g_x, g_y, g_z)	1.969, 1.967, 1.964	1.969, 1.968, 1.964	1.969, 1.967, 1.964
D / cm^{-1}	-0.29	-0.35	-0.29
$\|E/D\|$	0.157	0.07	0.09

Table S3. *Ab-initio* (MOLCAS-SINGLE_ANISO) computed single ion energies and *g*-tensors of low lying energy KDs of Dy(III) sites in complex **2**.

Dy(1)			**Dy(2)**			**Dy(3)**		
energy /cm^{-1}	***g* tensor g_x, g_y, g_z**	**comp. / % (m_J)**	**energy /cm^{-1}**	***g* tensor g_x, g_y, g_z**	**comp. / % (m_J)**	**energy /cm^{-1}**	***g* tensor g_x, g_y, g_z**	**comp. / % (m_J)**
0.00	0.94, 1.76, 14.5	41% (13/2)	0.00	0.05, 0.06, 19.33	90% (15/2)	0.00	0.05, 0.09, 19.23	88% (15/2)
0.00			0.00			0.00		
20.83	1.17, 3.22, 10.73	29% (11/2)	79.53	0.65, 0.90, 15.63	66% (13/2)	72.06	0.26, 0.35, 16.32	73% (13/2)
20.83			79.53			72.06		
46.05	1.95, 5.14, 10.23	44% (9/2)	123.75	3.46, 5.70, 9.01	35% (11/2)	119.23	1.59, 3.14, 13.47	30% (9/2)
46.05			123.75			119.23		
60.23	0.17, 3.32, 11.73	22% (11/2)	146.07	0.75, 3.11, 12.34	30% (5/2)	141.23	0.85, 3.27, 13.10	28% (3/2)
60.23			146.07			141.23		
67.98	1.70, 4.55, 10.86	32% (5/2)	156.16	2.69, 3.47, 11.22	29% (1/2)	162.16	3.31, 5.79, 10.50	30% (7/2)
67.98			156.16			162.16		
92.18	2.24, 3.32, 13.35	34% (3/2)	170.65	1.36, 2.14, 13.02	36% (3/2)	188.91	0.14, 0.90, 17.18	55% (1/2)
92.18			170.65			188.91		
118.41	0.38, 0.92, 18.47	52% (1/2)	191.81	0.78, 3.53, 15.95	31% (7/2)	207.44	0.28, 1.73, 16.19	39% (5/2)
118.41			191.81			207.44		
378.27	0.00, 0.00, 19.96	58% (15/2)	465.52	0.00, 0.00, 19.95	25% (9/2)	473.32	0.00, 0.00, 19.93	27% (9/2)
378.27			465.52			473.32		

Table S4. CHELPG-DFT computed charges of Dy(III) coordinate oxygen atoms and Dy-O bond distances in complex **2**.

Dy1				**Dy2**				**Dy3**			
	charge	**%**	**Dy-O / Å**		**charge**	**%**	**Dy-O / Å**		**charge**	**%**	**Dy-O / Å**
μ-OH (O1)	-1.438	22	2.390(8)	μ-OH (O1)	-1.438	23	2.402(8)	μ-OH (O1)	-1.438	23	2.418(7)
aqua (O27)	-0.964	15	2.38(1)	μ-piv (O23)	-0.853	13	2.35(1)	μ-OH (O4)	-0.823	13	2.386(8)
μ-OH (O2)	-0.829	13	2.400(8)	μ-OH (O2)	-0.829	13	2.377(7)	μ-piv (O25)	-0.787	12	2.35(1)
μ-OH (O4)	-0.823	13	2.422(7)	tea (O7)	-0.778	12	2.323(8)	μ_2-piv (O16)	-0.667	10	2.388(8)
tea (O10)	-0.703	11	2.315(9)	tea (O6)	-0.754	12	2.333(8)	μ_2-piv (O17)	-0.659	10	2.388(7)
μ_2-piv (O21)	-0.693	11	2.370(9)	μ_2-piv (O13)	-0.734	12	2.386(9)	tea (O8)	-0.655	10	2.316(7)
μ_2-piv (O20)	-0.669	10	2.359(7)	μ_2-piv (O12)	-0.686	11	2.406(8)	μ-OH (O3)	-0.555	9	2.405(8)
tea (O5)	-0.661	10	2.322(7)	μ-OH (O3)	-0.555	9	2.394(7)	tea (O9)	-0.536	8	2.35(1)
Total	-6.627				-6.120				-6.780		

Table S5. Matrix main values (after diagonalization) of projected (onto effective Dy(III) *s*=1/2 ground KD) exchange and dipolar magnetic interactions as arising from MOLCAS/POLY_ANISO based experimental data fitting.

	Dy1-Dy2	**Dy1- Dy3**	**Dy2- Dy3**	**Cr1-Dy2**	**Cr1-Dy1**	**Cr2-Dy2**	**Cr2-Dy3**	**Cr3-Dy3**	**Cr3-Dy1**
J_{exc} / cm^{-1}									
x	-0.058	-0.024	0.366	1.428	-2.087	2.355	1.299	-1.486	0.816
y	0.017	0.001	0.000	-0.342	0.020	-2.014	-0.015	-0.011	0.290
z	0.000	0.024	-0.129	0.000	0.520	0.013	-2.916	2.534	-1.742
isotropic contribution	**-0.014**	**0.001**	**0.079**	**0.362**	**-0.516**	**0.118**	**-0.544**	**0.346**	**-0.212**
J_{dip} / cm^{-1}									
x	0.377	0.406	-0.900	0.017	0.061	-0.101	0.031	-0.053	0.061
y	-0.303	0.001	0.000	-0.018	-0.028	0.061	0.000	0.062	0.010
z	0.000	-0.200	0.318	0.000	0.001	0.000	-0.087	0.001	-0.058
isotropic contribution	**0.025**	**0.069**	**-0.194**	**0.000**	**0.012**	**-0.013**	**-0.019**	**0.003**	**0.004**
J_{total} / cm^{-1} **isotropic contribution**	**0.070**	**-0.115**	**0.362**	**-0.504**	**0.105**	**-0.563**	**0.349**	**-0.208**	**0.000**

Table S6. Fitting parameters of AC magnetic data frequency dependence under variable temperature and variable DC magnetic field (2 K) , through a generalized Debye model.

	T / K	χ_{S1} / cm^3mol^{-1}	χ_{T1} / cm^3mol^{-1}	χ_{S2} / cm^3mol^{-1}	χ_{T2} / cm^3mol^{-1}	α_1	α_2	τ_1 / s	τ_2 / s
0 Oe	2	1.29	3.82	1.81	10.26	0.19	0.23	7.43E-04	4.74E-05
	2.2	1.29	3.20	1.54	10.20	0.17	0.26	3.80E-04	2.32E-05
	2.3	1.13	2.47	1.40	10.20	0.13	0.28	1.94E-04	1.14E-05
	2.5	1.13	1.95	0.01	10.05	0.06	0.35	8.45E-05	4.25E-06
	2.7	1.13	1.77	0.01	9.54	0.00	0.32	4.76E-05	2.24E-06
	3	1.13	1.67	0.01	9.04	0.02	0.29	2.72E-05	1.23E-06
	3.2	1.13	1.58	0.01	8.53	0.07	0.20	1.89E-05	8.50E-07
	3.4	1.05	1.55	0.00	8.03	0.00	0.23	8.37E-06	3.29E-07
1500 Oe	2.0	1.09	3.48	5.08	10.03	0.28	0.18	7.58E-04	4.11E-05
	2.2	0.90	3.16	5.08	10.27	0.30	0.20	3.29E-04	1.96E-05
	2.3	0.55	2.84	5.08	10.40	0.33	0.22	1.22E-04	8.61E-06
	2.5	0.55	2.86	4.25	10.08	0.31	0.23	4.70E-05	3.20E-06
	2.7	0.14	3.45	4.19	9.23	0.30	0.00	1.59E-05	1.39E-06
	3.0	0.14	2.86	3.32	9.49	0.24	0.00	9.74E-06	6.48E-07
	3.2	0.00	2.88	3.31	9.17	0.23	0.00	4.28E-06	2.63E-07
3000 Oe	2.0	0.43	1.26	0.23	2.89	0.23	0.24	6.86E-04	2.88E-05
	2.2	0.39	1.51	0.19	2.92	0.36	0.23	2.93E-04	1.36E-05
	2.3	0.37	1.25	0.19	3.28	0.27	0.23	1.67E-04	7.54E-06
	2.5	0.30	1.71	0.19	2.96	0.35	0.15	4.58E-05	3.42E-06
	2.7	0.00	2.08	0.00	2.65	0.34	0.00	1.22E-05	1.36E-06
	3.0	0.00	1.73	0.00	3.02	0.30	0.00	8.31E-06	8.28E-07

H/ Oe	χ_{S1} /cm^3mol^{-1}	χ_{T1} /cm^3mol^{-1}	χ_{S2} /cm^3mol^{-1}	χ_{T2} /cm^3mol^{-1}	α_1	α_2	τ_1 / s	τ_2 / s
0	0.00	3.44	4.74	11.69	0.17	0.23	8.39E-04	5.43E-05
200	0.00	3.44	4.71	11.50	0.18	0.22	7.93E-04	5.67E-05
400	0.08	3.44	4.71	11.28	0.15	0.20	9.39E-04	6.18E-05
600	0.08	3.44	4.71	10.80	0.18	0.19	8.79E-04	6.19E-05
800	0.12	3.44	4.71	10.30	0.17	0.17	8.37E-04	5.92E-05
1000	0.10	3.44	4.71	10.09	0.18	0.18	1.05E-03	5.83E-05
1200	0.13	2.90	4.69	9.71	0.14	0.18	8.56E-04	5.40E-05
1400	0.12	2.90	4.69	9.27	0.19	0.19	9.31E-04	5.01E-05
1600	0.14	2.50	4.67	8.84	0.18	0.19	7.66E-04	4.57E-05
1800	0.14	2.50	4.67	8.44	0.22	0.20	8.16E-04	4.23E-05
2000	0.21	2.13	4.62	8.11	0.18	0.20	7.13E-04	4.01E-05

Table S7. DFT computed high spin (HS) and broken symmetry (BS) states spin density and energy values of complexes **1**-**3**. Spin densities in bold type shows the sites where spin was flipped.

1 Gd_3Cr_3							
	spin density						**energy /hartrees**
	Gd1	**Gd2**	**Gd3**	**Cr1**	**Cr2**	**Cr3**	
HS	7.030636	7.019895	7.022084	3.103535	3.103966	3.104018	-42171. 26435364
BS1	7.025827	7.029480	7.018649	**-3.100190**	3.103821	3.103884	-42171.26461822
BS2	7.027005	7.024539	7.022595	3.103345	**-3.101652**	3.103895	-42171.26463101
BS3	7.033495	7.016355	7.032389	3.103685	3.103691	**-3.099802**	-42171.26463770
BS4	**-7.021467**	7.020622	7.021698	3.101442	3.104122	3.101727	-42171.26465029
BS5	7.028550	**-7.030868**	7.021890	3.102295	3.102384	3.104133	-42171.26460572
BS6	7.029108	7.019447	**-7.028892**	3.103547	3.103483	3.102225	-42171.26462936
BS7	7.022198	7.034130	7.019156	**-3.100373**	**-3.101794**	3.103761	-42171.26489414
BS8	7.029871	7.020996	7.032903	3.103496	**-3.101919**	**-3.099923**	-42171.26491406
BS9	7.028697	7.025938	7.028949	**-3.100035**	3.103548	**-3.099930**	-42171.26490169
BS10	**-7.023550**	**-7.030139**	7.021505	3.100205	3.102541	3.101843	-42171.26490073
BS11	7.027025	**-7.031317**	**-7.029081**	3.102309	3.101903	3.102339	-42171.26488297

BS12	**-7.022988**	7.020177	**-7.029279**	3.101455	3.103641	3.099935	-42171.26492424
2 Dy_3Cr_3							
	spin density						**energy /hartrees**
	Dy1	**Dy2**	**Dy3**	**Cr1**	**Cr2**	**Cr3**	
HS	5.012296	5.008149	5.008876	3.096606	3.100598	3.102621	-44860.53125569
BS1	5.011670	5.008145	5.004826	**-3.098220**	3.100359	3.102516	-44860.53133291
BS2	5.008482	5.009443	5.009557	3.096468	**-3.101294**	3.102492	-44860.53138059
BS3	5.015293	5.003634	5.016460	3.096752	3.100346	**-3.101914**	-44860.53143496
BS4	**-5.006657**	5.006200	5.009662	3.097846	3.100599	3.102276	-44860.53139673
BS5	5.011540	**-4.997276**	5.005657	3.096919	3.101236	3.102520	-44860.53138684
BS6	5.008869	5.002439	**-5.010655**	3.096643	3.101135	3.102591	-44860.53135976
BS7	**-5.007282**	5.006189	5.005611	**-3.096971**	3.100360	3.102177	-44860.53139249
BS8	5.007723	**-4.995979**	5.006337	3.096787	**-3.100651**	3.102391	-44860.53131721
BS9	5.011863	4.997927	**-5.003073**	3.096790	3.100889	**-3.101944**	-44860.53137956
3 Y_3Cr_3							
	spin density						**energy /hartrees**
	Cr1		**Cr2**		**Cr3**		
HS	3.110680		3.130028		3.113920		-7681.58091298
BS1	**-3.110330**		3.129907		3.113777		-7681.58091320
BS2	3.110513		**-3.129396**		3.113690		-7681.58091349
BS3	3.110498		3.129516		**-3.113547**		-7681.58091317

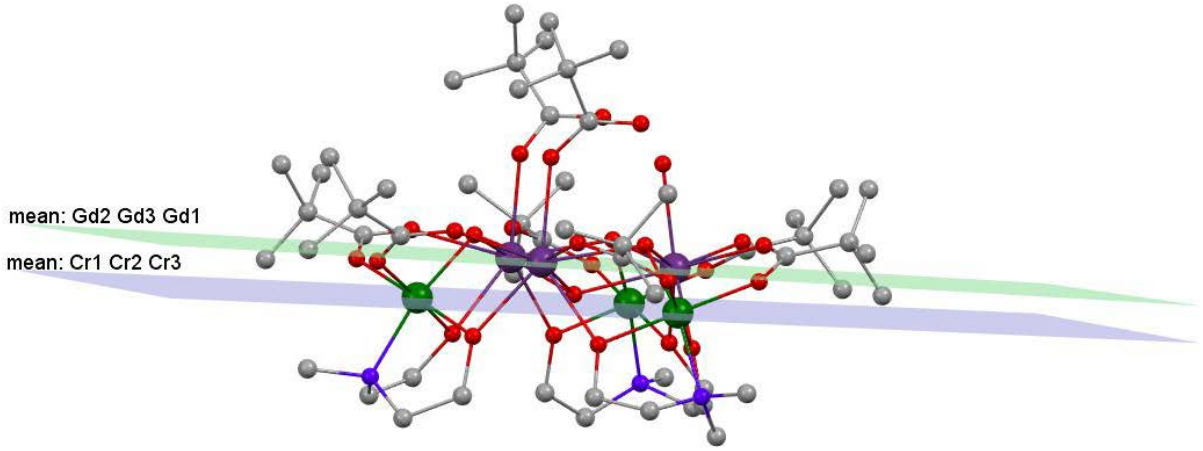


Figure S1. Molecular planes containing all Gd and all Cr atoms running parallel (angle between planes *ca*. 1°) and shifted *ca*. 1 Å. Complex **1** is chosen as example of all isostructural complexes **1**-**3**. Hydrogen atoms as well as disordered moieties have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.

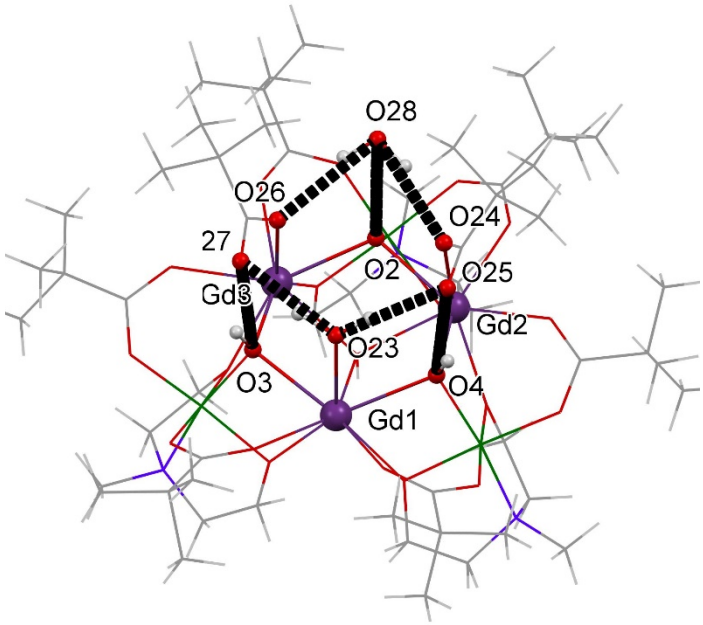
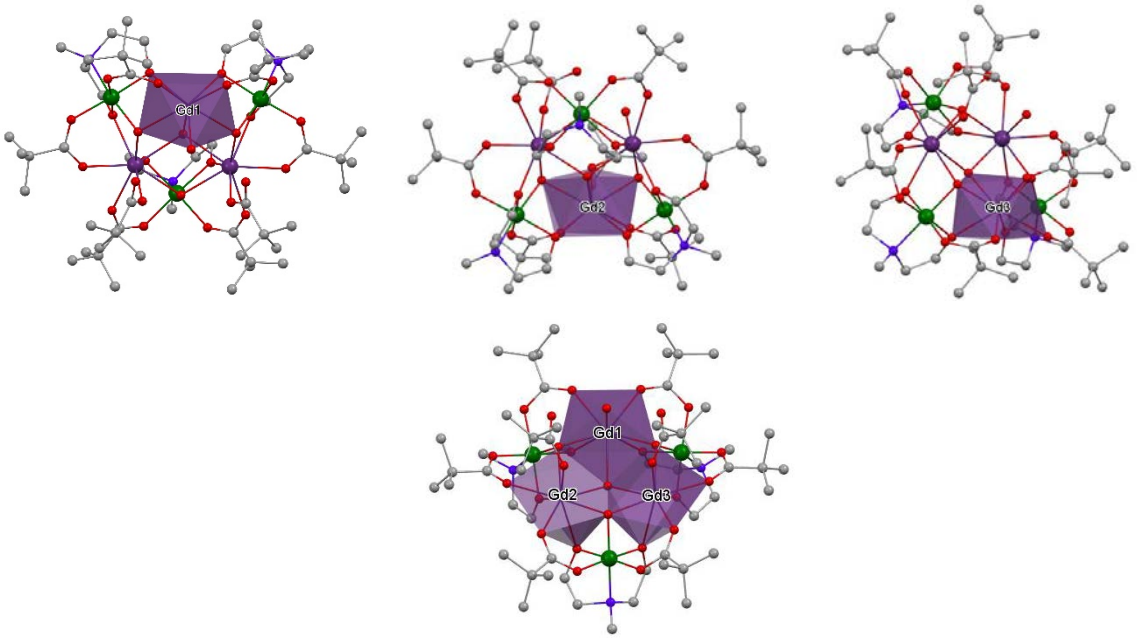
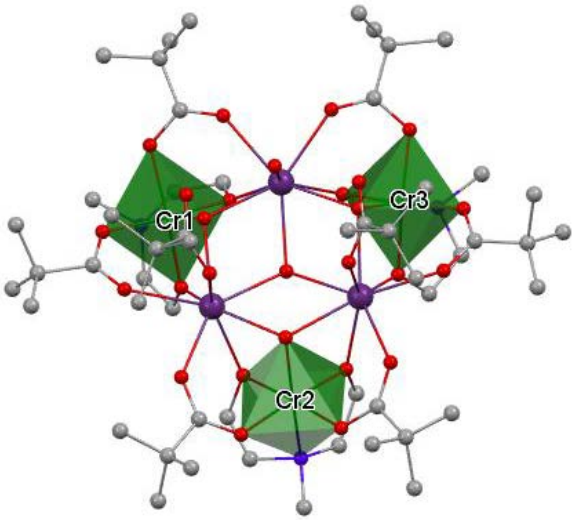


Figure S2. Intra-molecular H-bonding network (involving one solvent water molecule, O28) in complex **1** structure. Complex **1** is chosen as example of all isostructural complexes **1**-**3**. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.



Figures S3. Polyhedral representation of Gd sites in complex **1**. Complex **1** is chosen as example of all isostructural complexes **1-3**. Hydrogen atoms as well as disordered moieties have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.



Figures S4. Polyhedral representation of Cr sites in complex **1**. Complex **1** is chosen as example of all isostructural complexes **1-3**. Hydrogen atoms as well as disordered moieties have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.

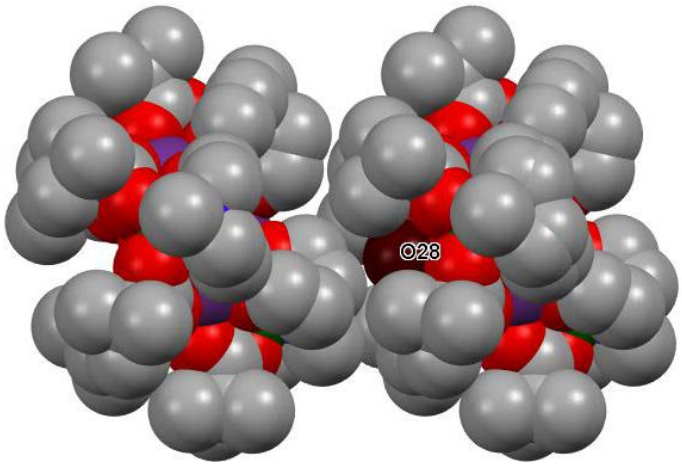


Figure S5. Spacefill representation of adjacent complex **1** molecules, showing the encapsulated solvent water molecules (O28). Complex **1** is chosen as example of all isostructural complexes **1**-**3**. Hydrogen atoms as well as disordered moieties have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.

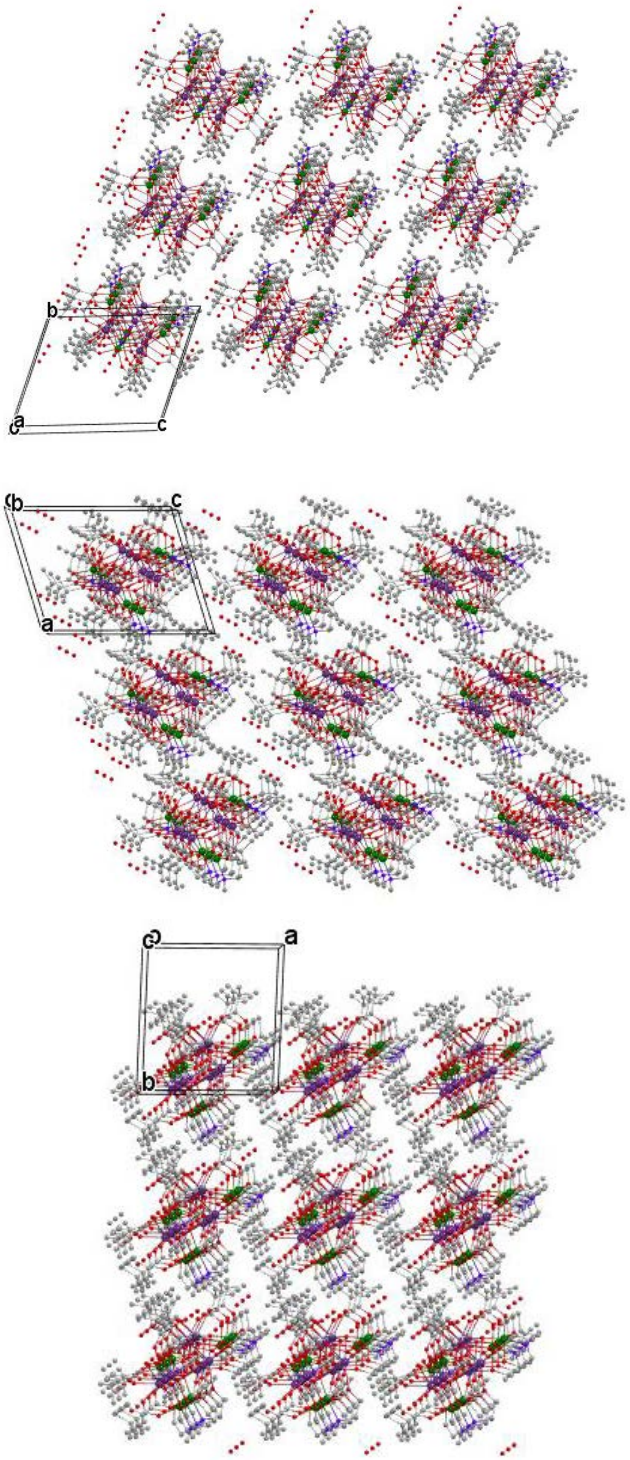


Figure S6. Crystal packing of complex **1** view along unit cell directions: *a*, *b* and *c* axis from top to bottom respectively. Complex **1** is chosen as example of all isostructural complexes **1**-**3**. Hydrogen atoms have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.

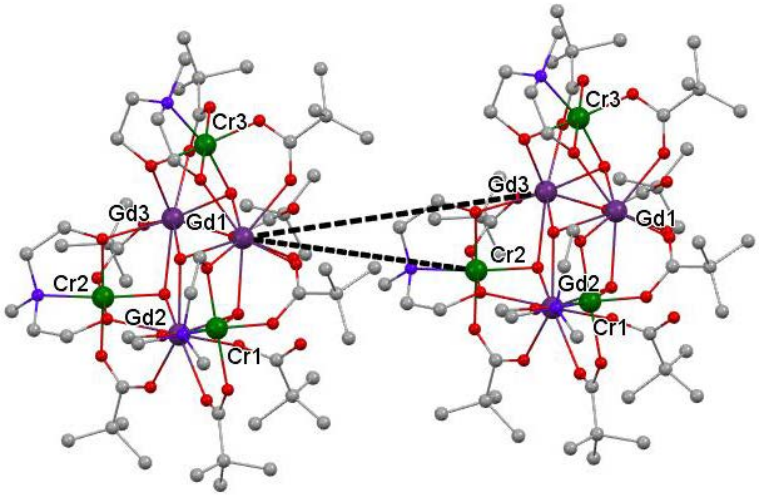


Figure S7. Shortest inter-molecular metal-metal distance and Ln·····Ln distance. Complex **1** is chosen as example of all isostructural complexes **1**-**3**. Hydrogen atoms as well as disordered moieties have been omitted to improve visualization. Colour code: C: grey; O: red; N: blue; Cr: green and Gd: violet.

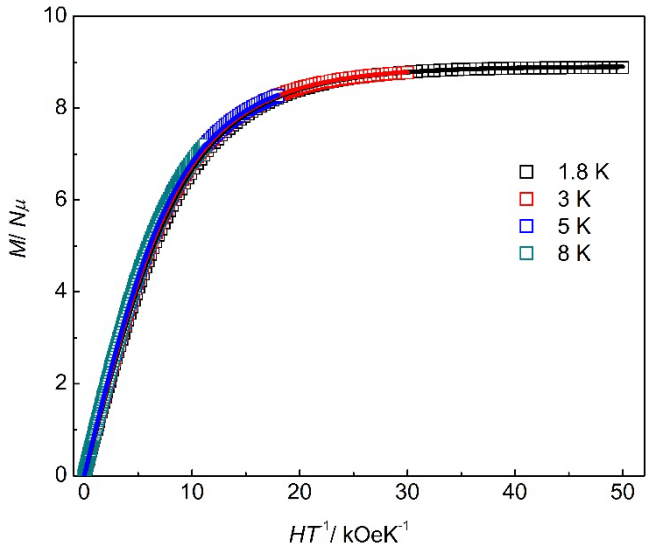


Figure S8. Reduced magnetization data plot of complex **3**. Open symbols: experimental data; Full lines: simulated data with model described in the text.

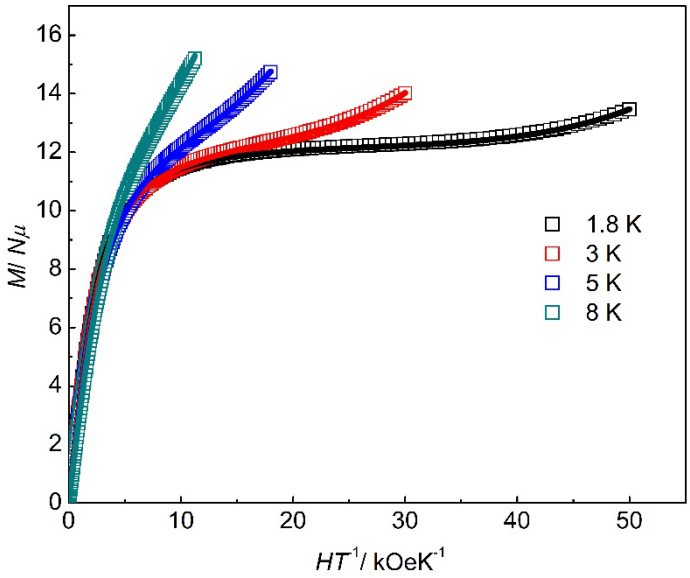


Figure S9. Reduced magnetization data plot of complex **1**. Open symbols: experimental data; Full lines: simulated data with model described in the text.

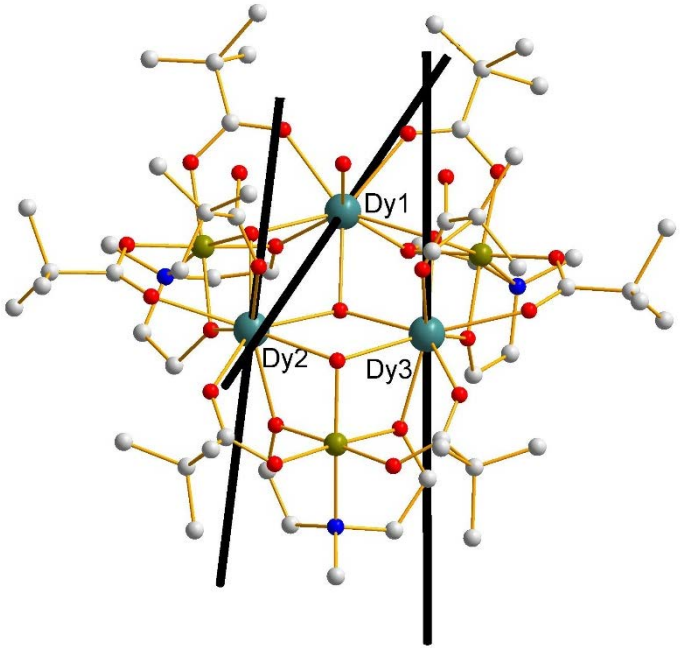
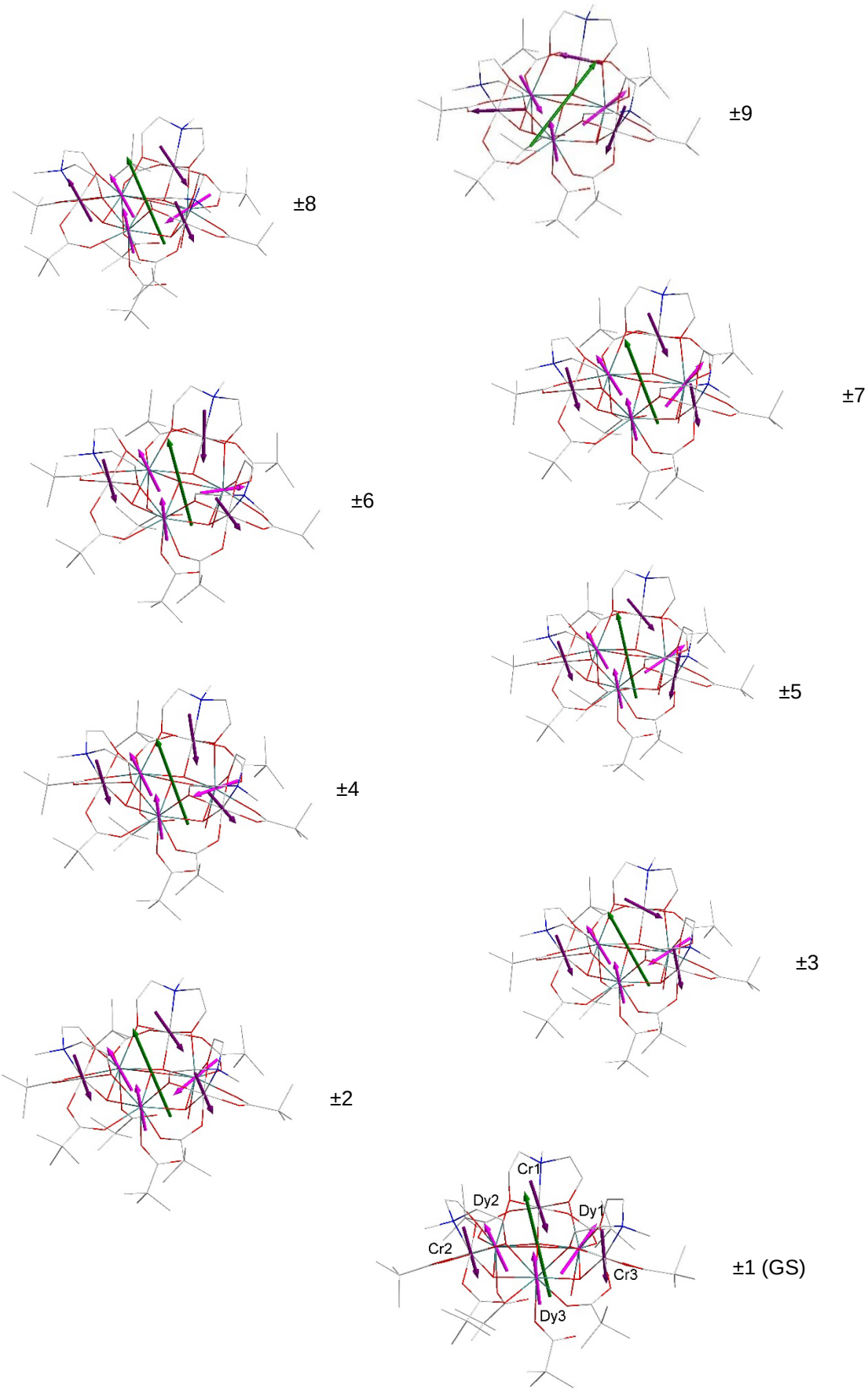
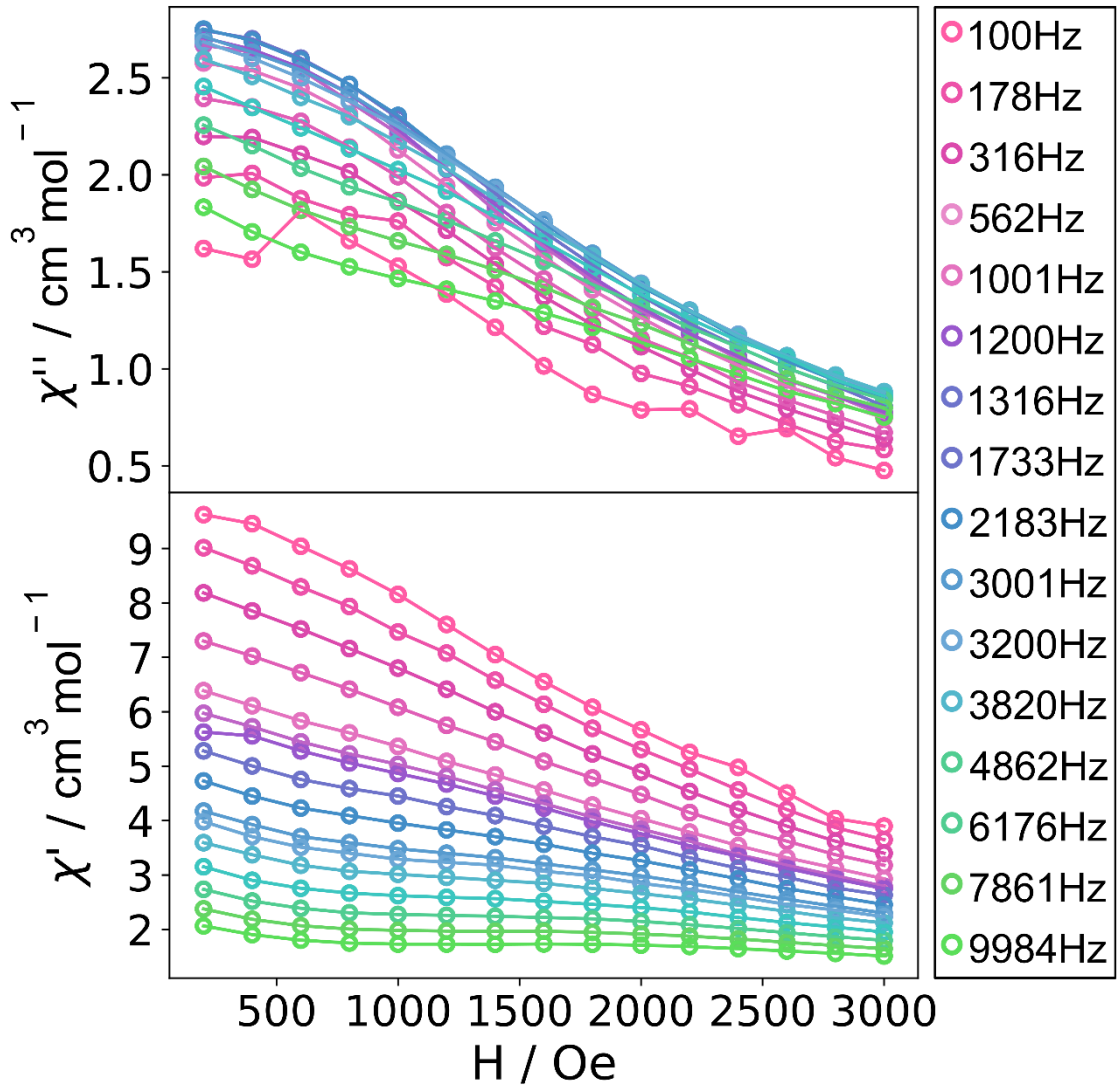


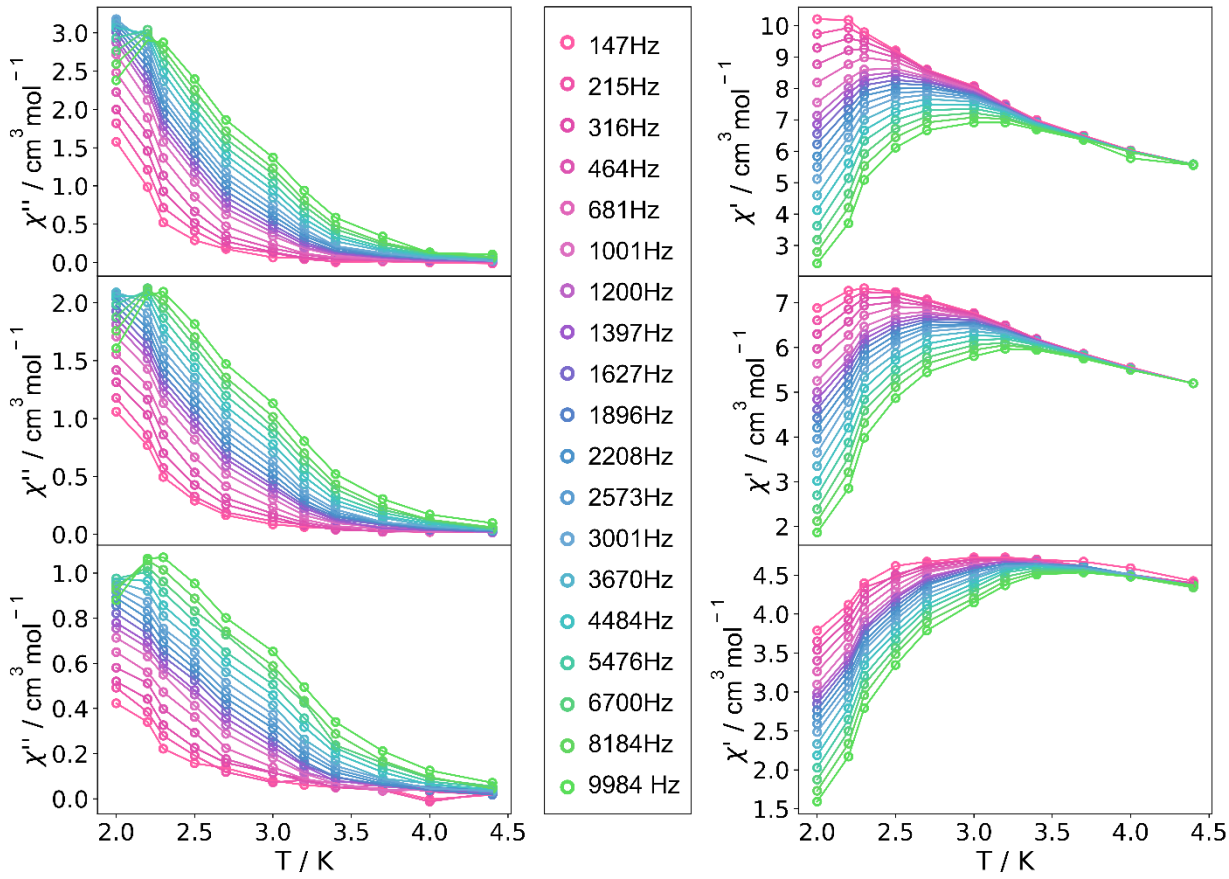
Figure S10. Local main magnetic moment quantization axis orientation at each Dy(III) site in complex **2**, as arising from MOLCAS/SINGLE_ANISO quantum computations.



FigureS11. Local magnetic moment orientations within low lying exchange states in complex **2** as arising from MOLCAS/POLY_ANISO computation.



FigureS12. AC magnetic susceptibility data of complex **2** at 2 K under DC applied magnetic field (0-3 kOe) and variable frequency (up to 10 kHz). Lines are only drawn as eye guideline.



Figures S13. χ'' (left) and χ' (right) temperature dependence with frequencies up to 10 kHz at 0 Oe (top), 1500 Oe (middle) and 3000 Oe (bottom) applied DC magnetic fields for complex **2**. Open symbols: experimental data; Full lines: eye guidelines.

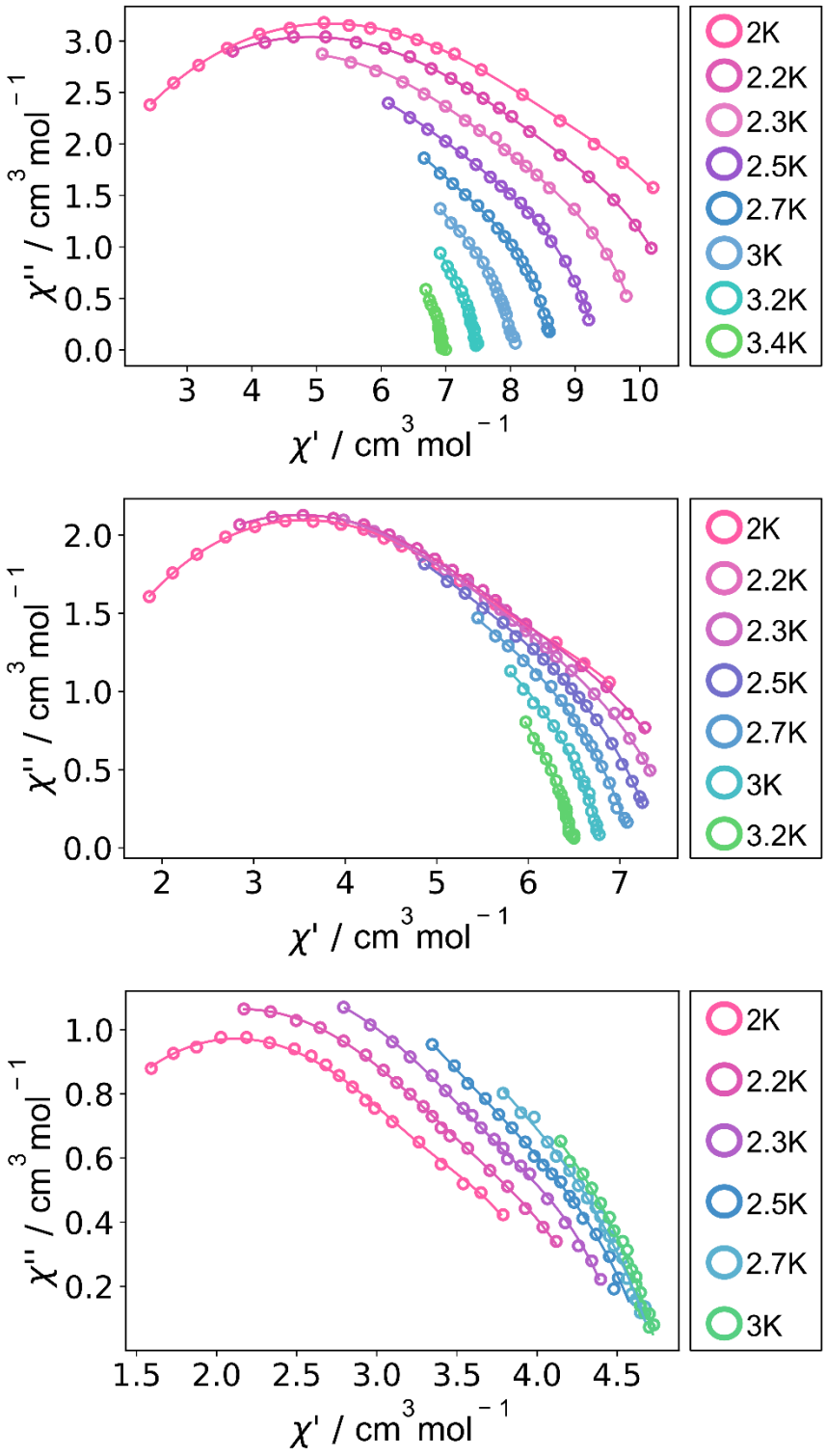
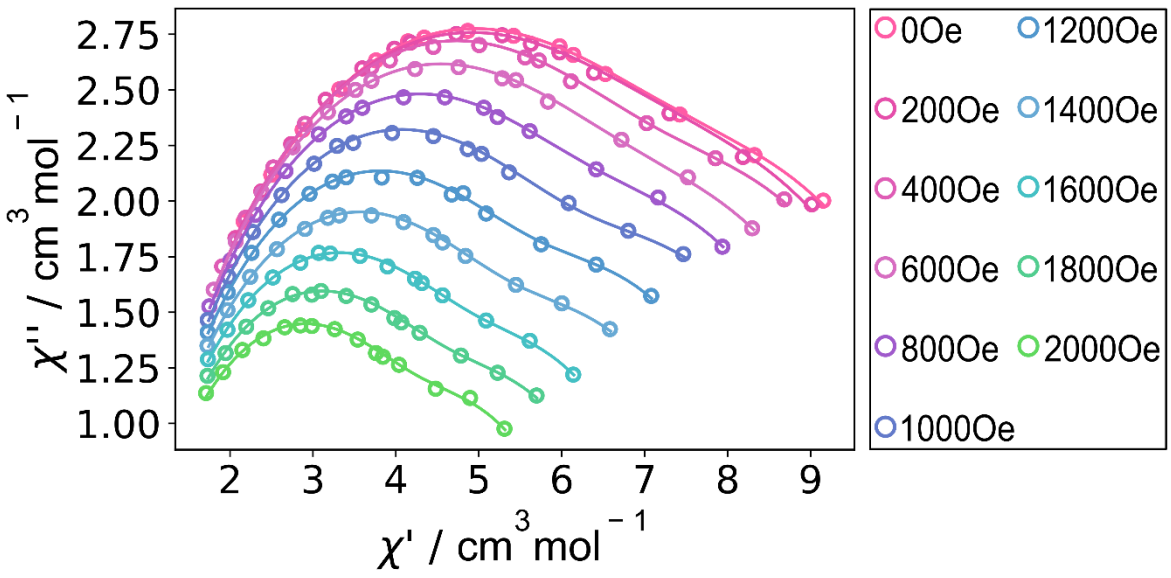
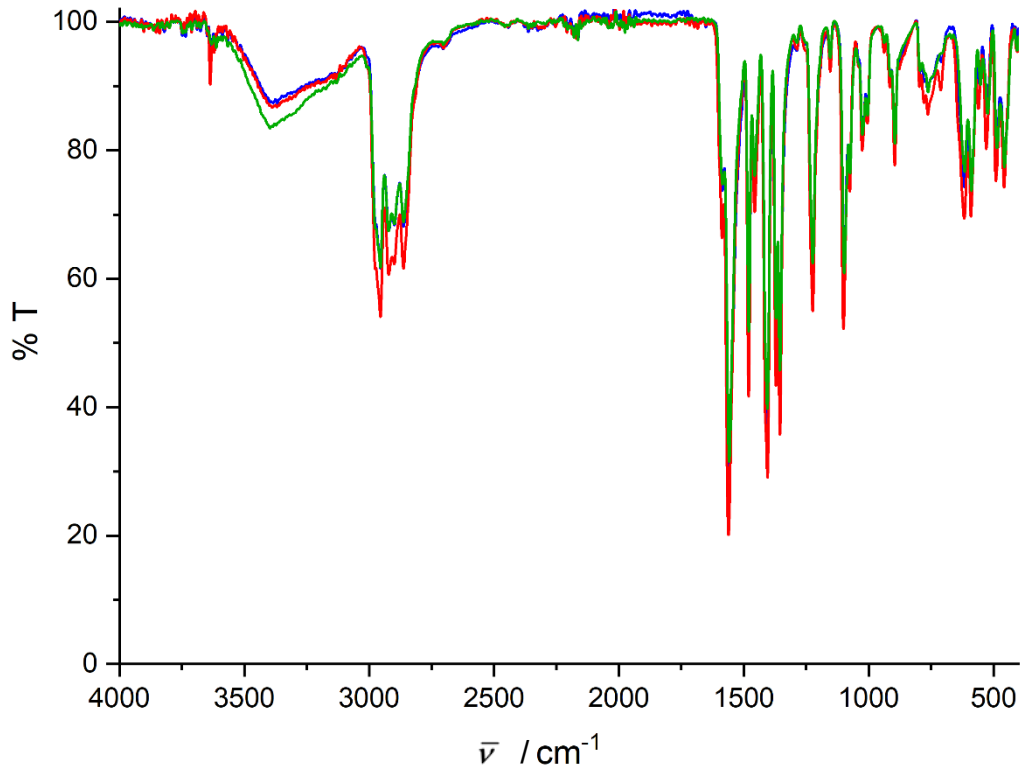


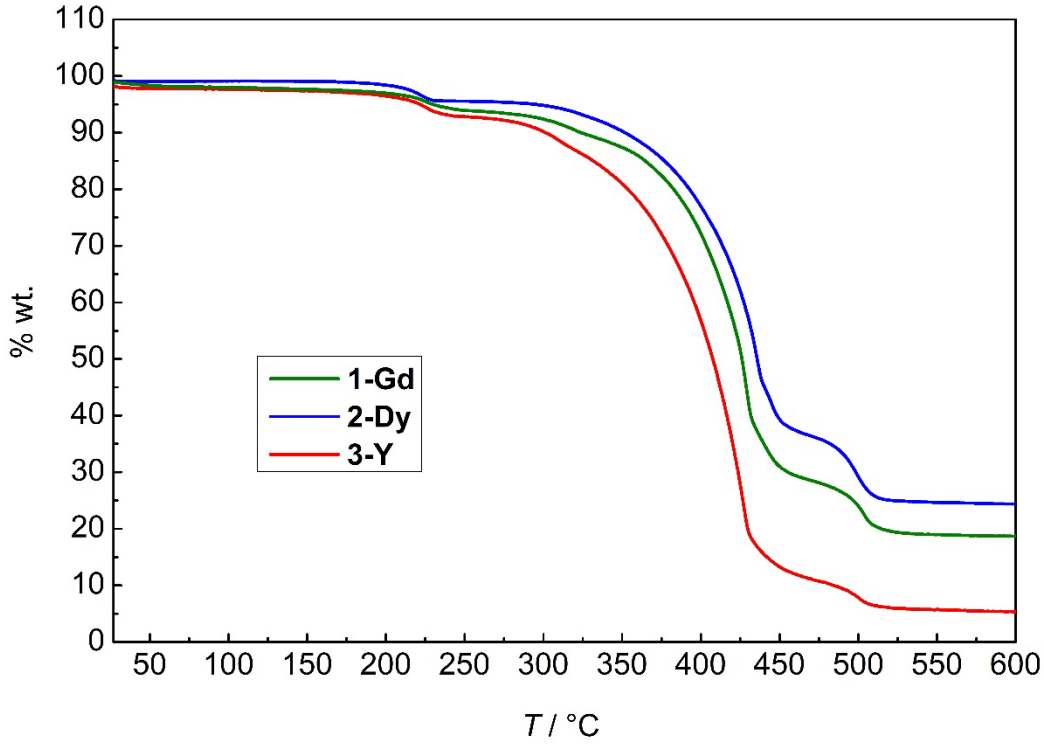
Figure S14. Cole-Cole plots of AC magnetic data for complex **2** at 0 Oe (top); 1500 Oe (middle) and 3000 Oe (bottom) DC magnetic field. Open symbols: experimental data; full lines: simulated data with generalized Debye model (see text).



Figures S15. Cole-Cole plots of AC magnetic data for complex **2** at 2K under DC magnetic fields between 0 and 2000 Oe. Open symbols: experimental data; full lines: simulated data with generalized Debye model (see text).



Figures S16. Infrared spectra of reported complexes **1-Gd** (green), **2-Dy** (blue) and **3-Y** (red).



Figures S17. Thermogravimetric (TGA) data of reported complexes **1-3**.

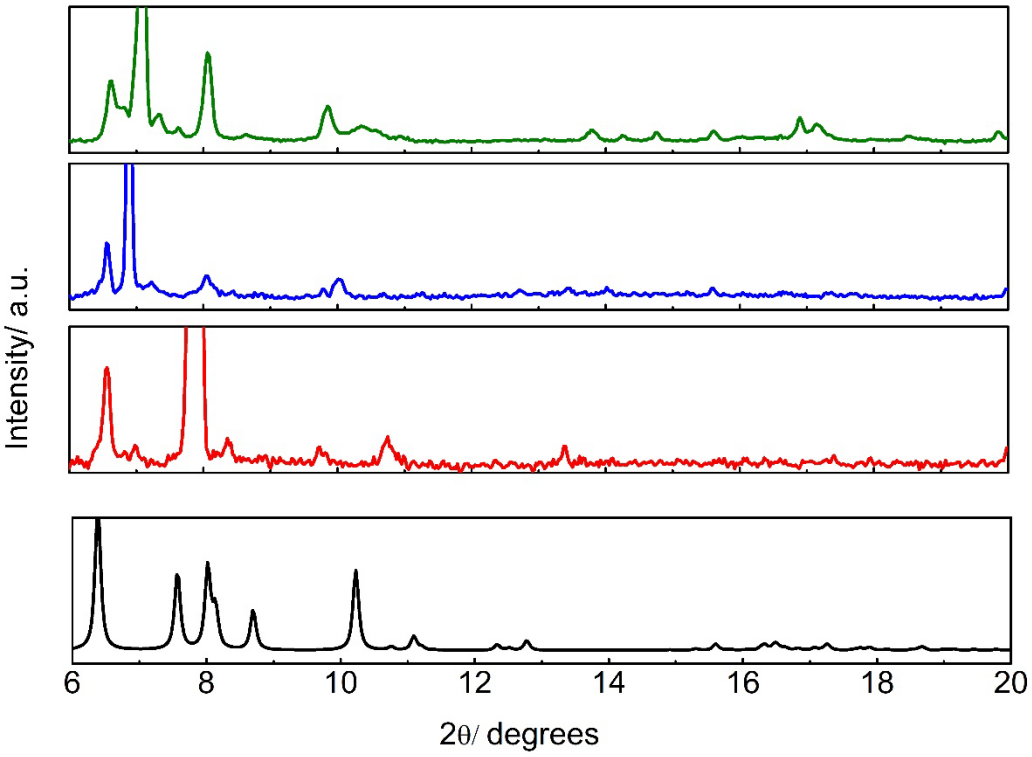


Figure S18. Powder *X*-ray diffractograms at room temperature of complexes **1-Gd** (red), **2-Dy** (blue) and **3-Y** (green) and simulated diffractogram from single crystal *X*-ray data (black).

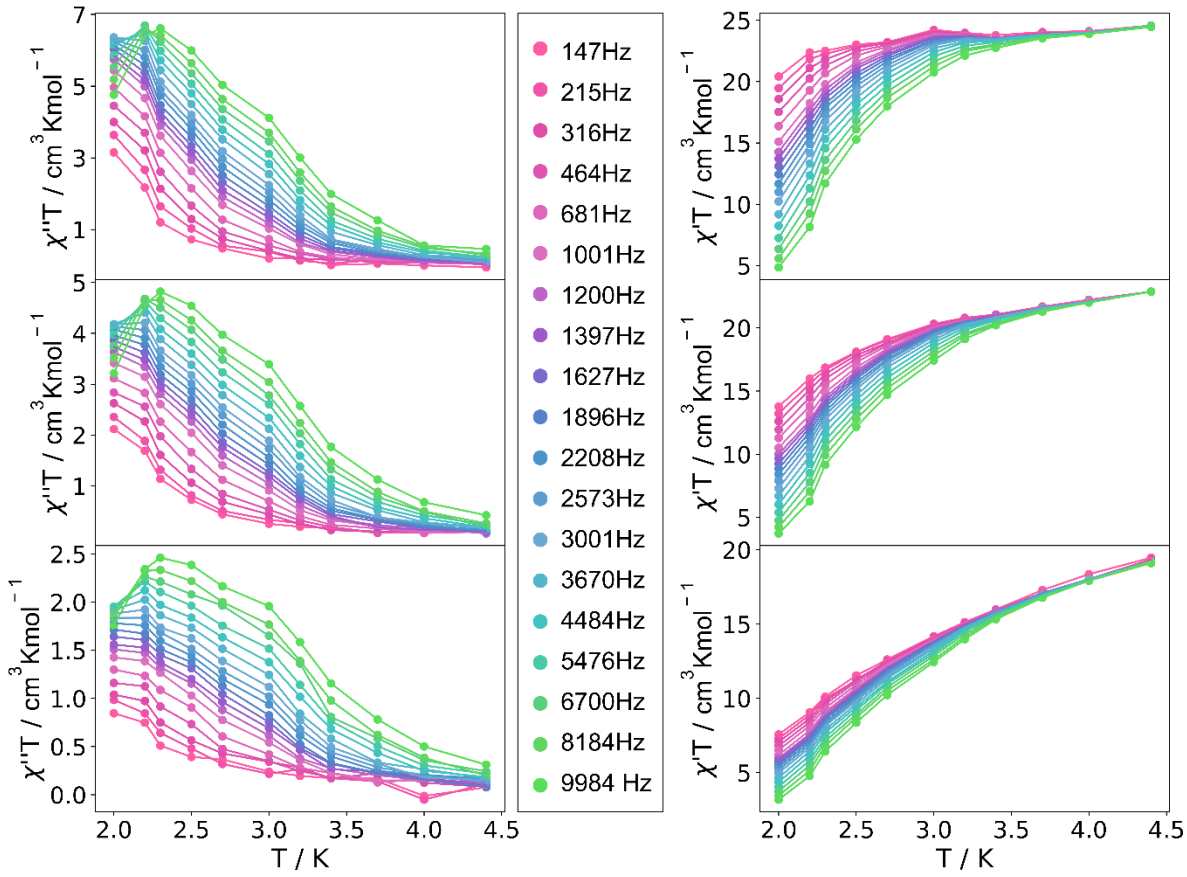


Figure S19. χT vs. T AC data at 0 DC (bottom), 1500 Oe (middle) and 3000 Oe (top) external DC field of complex **2-Dy**.

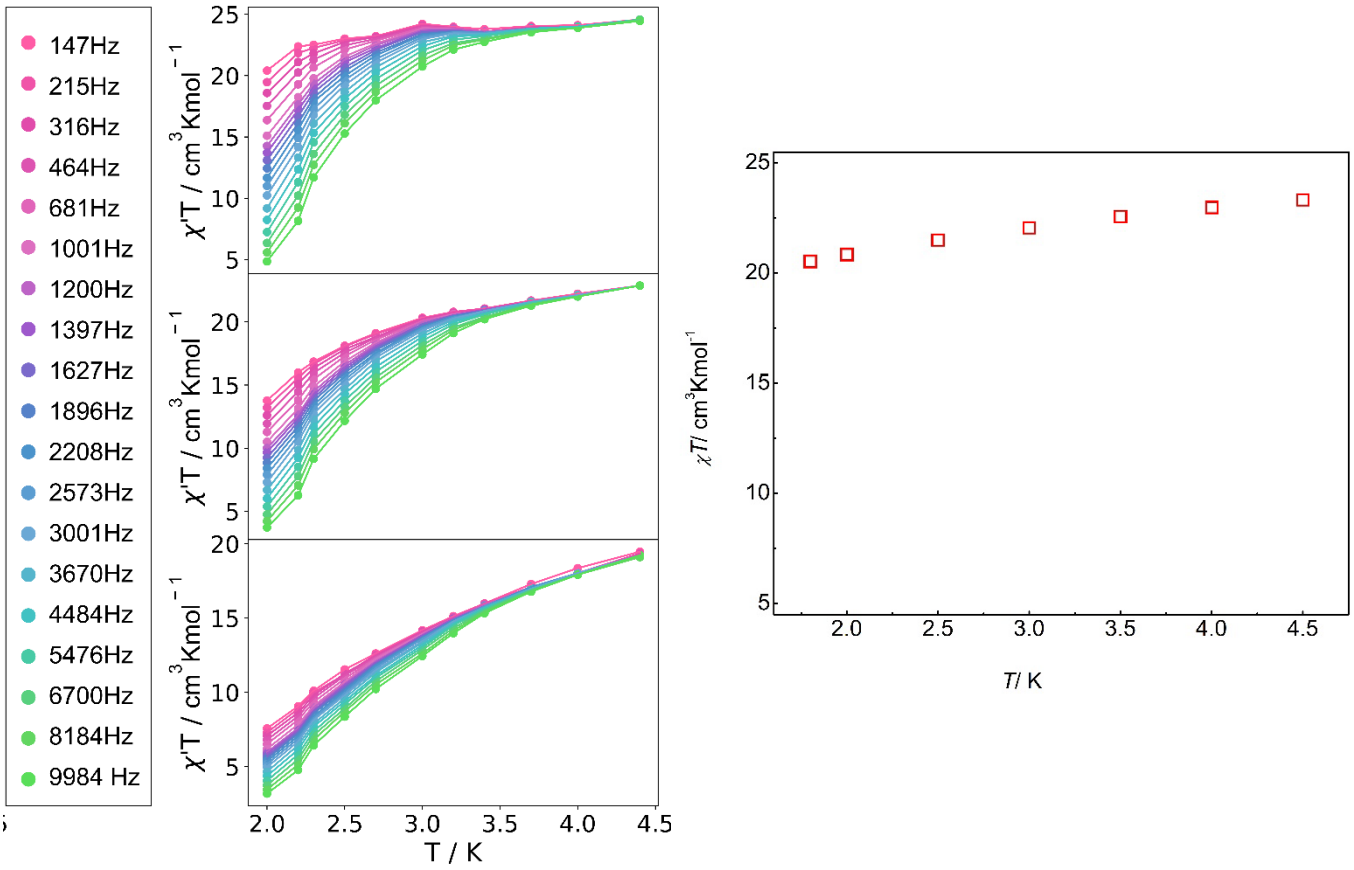


Figure S20. Left: χT vs. T AC data at 0 DC (bottom), 1500 Oe (middle) and 3000 Oe (top) external DC field of complex **2-Dy**. Right: DC χT vs. T data below 4.5 K of complex **2** at 1000 Oe.