SUPPORTING INFORMATION

Modulating Magnetic Anisotropy in Linear Tetranuclear Dysprosium(III) Complexes via Coordinated Anions.

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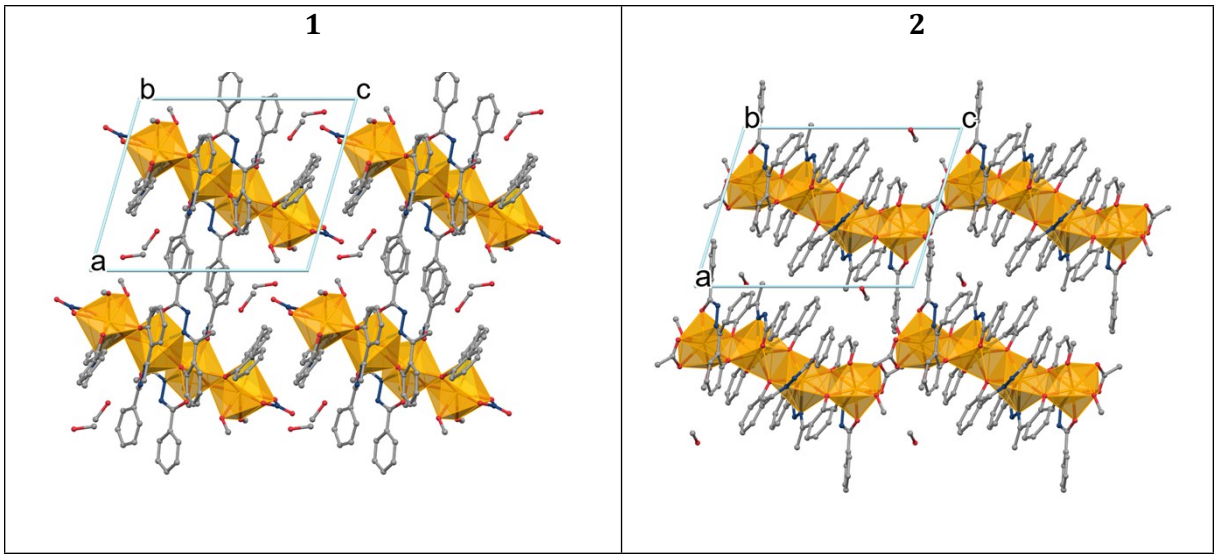

Figure S1: Views of the packing arrangements for complexes **1** and **2** along the *b* crystallographic axis. The hydrogen atoms have been omitted for clarity.

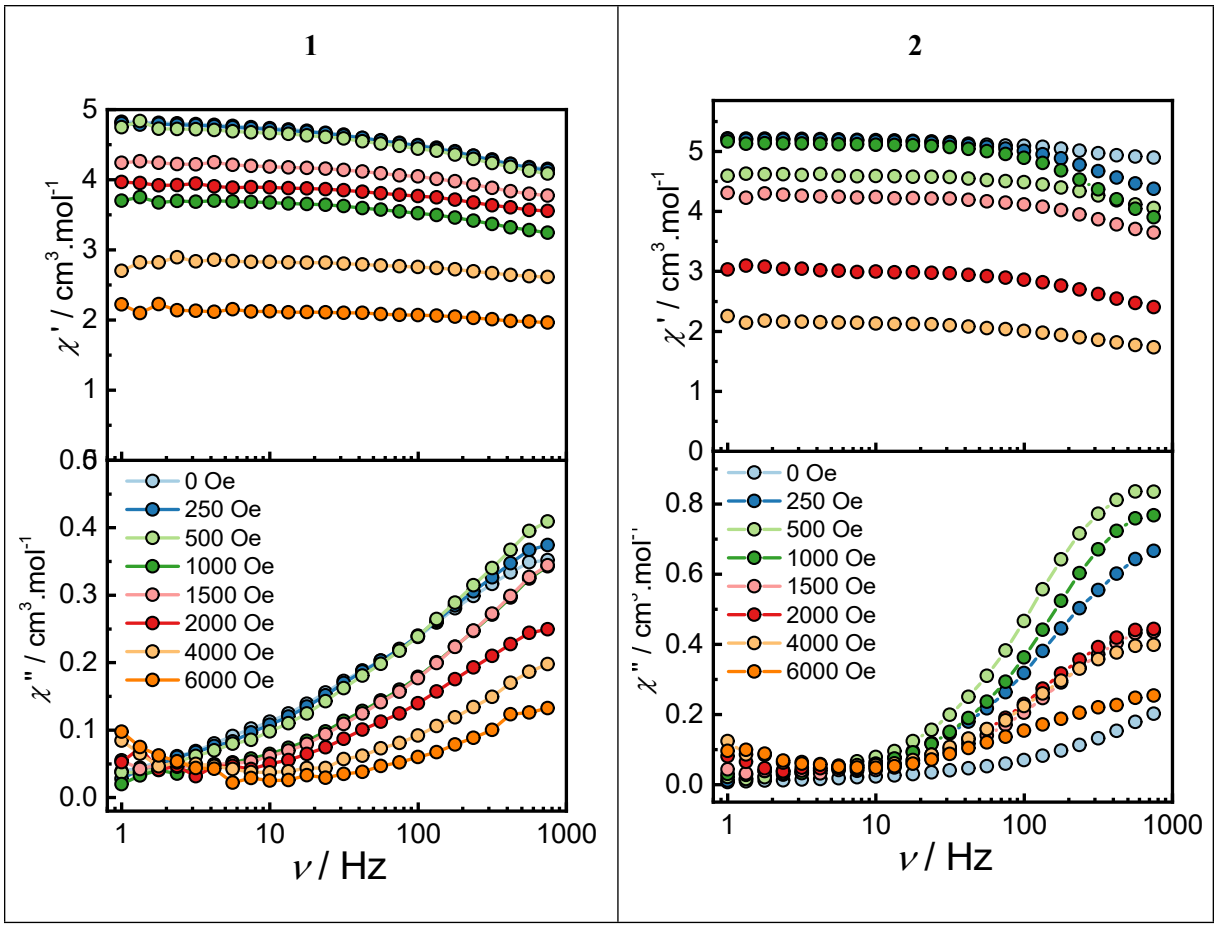

Figure S2: Frequency dependence of χ' and χ'' for **1** and **2** for various dc fields at 2 K.

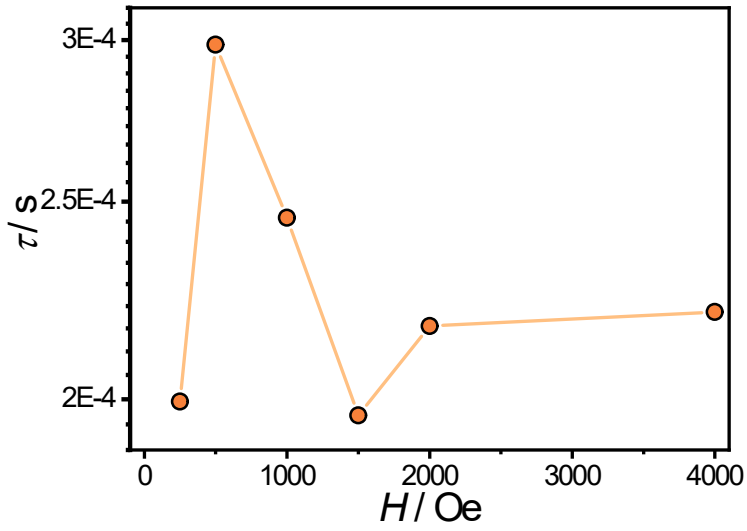

Figure S3: Field dependence of the relaxation time for **1** and **2** at 2 K. Line is a guide for eyes.

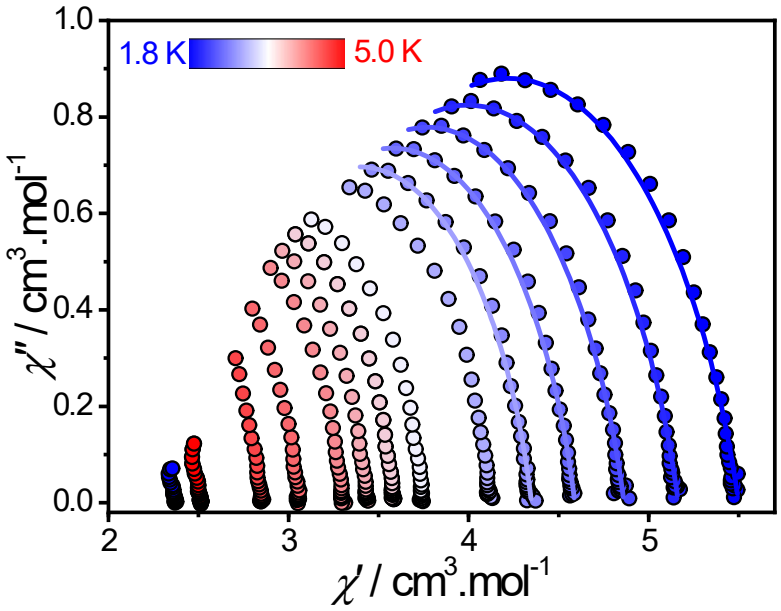

Figure S4: Cole-Cole (Argand) plots obtained using the ac susceptibility data for **2** under a 500 Oe field.

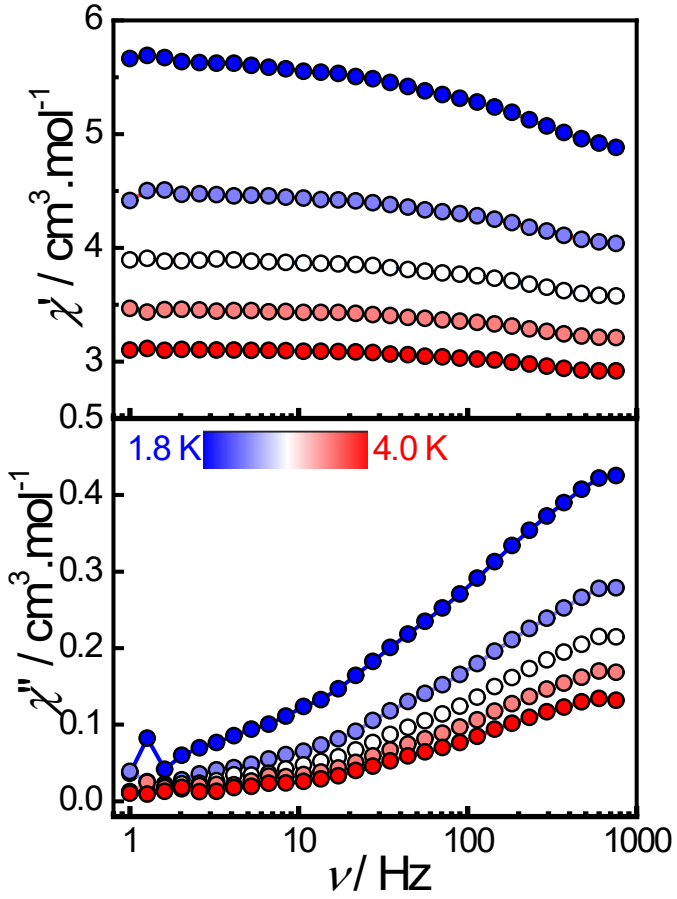

Figure S5: Frequency dependence of the out-of phase (χ″) susceptibilities for **1** under a 500 Oe dc-field.

Table S1: Crystal data, data collection and structure refinement details for **1-2**.

	1	**2**
Formula	$C_{98}H_{106}Dy_4N14_{O26}$	$C_{100}H_{104}Dy_4N_{12}O_{22}$
M	2545.96	2475.95
T, K	150.15 K	150.15 K
Crystal system	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$
Z (*Z*')	1	1
a, Å	13.8482(6)	13.1922(8)
b, Å	14.9319(7)	14.1360(9)
c, Å	14.9564(7)	15.6765(10)
α, °	93.119(2)°	101.295(2)°
β, °	102.607(2)°	99.543(2)°
γ, °	117.440(2)°	117.467(2)°.
V, Å^3	2635.3(2)	2430.9(3)
d_{calcd}, g.cm^{-3}	1.604	1.691
Mu, mm^{-1}	2.881	3.117
F_{000}	1264	1228
wR_2 (all data)	0.0433	0.0511
$S(F^2)$	1.095	1.044

Table S2: SHAPE analysis for **1** and **2**.

		OP	HPY	HBPY	CU	SAPR	TDD	JGBF	JETBPY	JBTPR	BTPR	JSD	TT	ETBPY
1	Dy1	31.470	21.671	16.080	9.920	1.874	**1.214**	15.949	28.006	3.045	2.161	4.423	10.313	24.224
	Dy2	34.187	22.134	16.724	9.805	2.735	**1.657**	15.678	28.296	3.945	2.934	4.784	10.554	23.932
2	Dy1	33.511	21.253	15.23	9.937	**1.318**	2.101	16.268	27.007	3.086	2.253	5.236	10.774	23.149
	Dy2	31.121	20.100	12.921	10.293	3.291	3.185	11.612	24.804	3.689	**2.985**	5.084	11.069	20.184

OP: Octagon
HPY: Heptagonal pyramid
HBPY: Hexagonal bipyramid
CU: Cube
SAPR: Square antiprism
TDD: Triangular dodecahedron
JGBF: Johnson gyrobifastigium
JETBPY: Johnson elongated triangular bipyramid
JBTPR: Johnson biaugmented trigonal prism
BTPR: Biaugmented trigonal prism
JSD: Snub diphenoid J84
TT: Triakis tetrahedron
ETBPY: Elongated trigonal bipyramid

Table S3: Fitting of the Cole-Cole plots with a generalized Debye model under a 500 Oe dc field for 2.

T (K)	χ_S (cm^3. mol^{-1})	χ_T (cm^3. mol^{-1})	α
1.80003	8.81555	16.49864	0.23319
1.99998	8.48569	15.49298	0.21726
2.19988	8.03302	14.58852	0.21163
2.39999	7.70871	13.76918	0.19876
2.60016	7.35575	13.04703	0.1938

Table S4: *Ab initio* calculated energies, *g*-tensor main values of the ground doublet and the nth KD doublet for the ground multiplet J = 15/2 obtained for the individual fragment **Dy1** in **1**.

KD	Energy (cm^{-1})	g_x	g_y	g_z	Wavefunction (only component with > 20 % are given)
1	0	10.5207026	9.34367039	1.62771667	43.8% |±3/2>>; 33.5% |±1/2>
2	14	7.10525321	5.30881065	1.80895843	38.0% |±5/2>; 34.0% |±1/2>
3	45	1.3922696	5.76276394	11.4865372	33.8 % |±7/2>;24.2% |±5/2>
4	84	8.47129535	7.3915465	3.15687154	35.4 % |±7/2>;23.8 % |±9/2>
5	144	1.36488508	3.78719637	12.3550548	37.4 % |±9/2>;29.5 %| ±11/2>
6	257	2.70365051	5.72520814	9.43800796	27.1% |±13/2>; 25.2 %| ±11/2>

7	305	1.80875076	6.80029146	12.0673329	31.1% | ±11/2>
8	493	0.0419435	0.09272241	19.4784586	82.7 | ±15/2>

Table S5: *Ab initio* calculated energies, *g*-tensor main values of the ground doublet and the nth KD doublet for the ground multiplet $J = 15/2$ obtained for the individual fragment **Dy2** in **1**.

KD	Energy (cm^{-1})	g_x	g_y	g_z	Wavefunction (only component with > 20 % are given)
1	0	0.00162983	0.0171375	19.7318171	97.1 % | ±15/2>>
2	66	1.03706486	1.72228401	15.905214	86.4% | ±13/2>
3	114	1.71041302	3.53998884	13.8437617	22.8 % | ±5/2>
4	160	10.0906727	5.41666892	0.17424847	47.7 % | ±11/2>
5	214	3.37321391	5.89830627	10.4540394	34.5 % | ±9/2>;20.9 %| ±11/2>
6	302	1.13128092	1.93976414	16.0474142	29.7% | ±9/2>; 27.8 %| ±7/2>
7	435	0.71890113	1.21749458	17.4542283	33.5% | ±3/2>; 21.9 %| ±5/2>; 20.7% | ±1/2>
8	475	0.37322956	1.92185715	18.118985	26.0% | ±5/2>; 23.1 %| ±1/2>; 22.2% | ±1/2>

Table S6: *Ab initio* calculated energies, *g*-tensor main values of the ground doublet and the nth KD doublet for the ground multiplet $J = 15/2$ obtained for the individual fragment **Dy1** in **2**.

KD	Energy (cm^{-1})	g_x	g_y	g_z	Wavefunction (only component with > 20 % are given)
1	0	0.60016941	1.10377075	16.75342281	61.9% | ±15/2>
2	29	2.08632926	3.40394824	13.73168298	26.1% | ±3/2>; 21.4% | ±7/2>; 21.3% | ±1/2>;
3	48	9.75597866	7.20354648	1.30051558	26.3 % | ±9/2>;
4	84	9.07173042	6.11402172	1.13390603	29.2 % | ±13/2>;24.0 % | ±9/2>;
5	153	1.79118542	4.28130216	12.30925322	No components > 20%
6	266	2.32520653	3.58804312	12.28138672	20.1% | ±7/2>;
7	317	1.61863692	4.29257961	14.84762711	24.7% | ±13/2>; 21.0% | ±11/2>

8	439	0.0960058	0.22691975	19.25800192	37.0% |±1/2>; 30.1% |±3/2>

Table S7: *Ab initio* calculated energies, *g*-tensor main values of the ground doublet and the nth KD doublet for the ground multiplet $J = 15/2$ obtained for the individual fragment **Dy2** in **2**.

KD	Energy (cm^{-1})	g_x	g_y	g_z	Wavefunction (only component with > 20 % are given)
1	0	0.03531591	0.06450346	19.6552356	97.0% |±15/2>
2	116	1.55272665	6.71466905	12.208662	47.6% |±13/2>
3	147	2.30861678	4.86349329	9.55702568	20.9 % |±9/2>
4	218	4.04923077	4.40300107	10.815582	24.7 % |±7/2>;21.4 % |±13/2>
5	269	0.97201679	4.94903047	10.9284212	21.2 % |±1/2>
6	316	0.51350407	1.92928421	16.6053338	20.3% |±5/2>;
7	355	0.53631204	2.19662127	16.1295451	26.2% |±9/2>; 21.3% |±7/2>
8	672	0.01071432	0.01582752	19.8326802	33.7% |±1/2>; 28.0% |±3/2>

Table S8: Weight of individual crystal-field parameters on the crystal-field splitting **Dy1** in **1**. Only values larger than 1% are given.

k	*q*	B_k^q	*Weight in %*
2	0	1.94E+00	14.01
4	-1	6.57E-03	8.59
4	3	-6.51E-03	8.51
4	2	5.73E-03	7.50
2	-2	-9.10E-01	6.56

4	4	-4.69E-03	6.13
4	0	3.68E-03	4.81
6	-4	-3.59E-05	4.40
6	6	3.17E-05	3.89
6	1	2.95E-05	3.62
2	-1	5.01E-01	3.61
4	-2	2.35E-03	3.08
6	-5	2.43E-05	2.98
6	2	2.39E-05	2.94
4	-3	-1.65E-03	2.16
6	-6	1.72E-05	2.10
4	-4	1.50E-03	1.97
6	-1	1.52E-05	1.86
6	5	1.27E-05	1.55
2	2	-1.83E-01	1.32
4	1	-9.04E-04	1.18
6	0	8.85E-06	1.09
6	-2	-8.27E-06	1.01

Table S9: Weight of individual crystal-field parameters on the crystal-field splitting obtained for **Dy1** in **2.** Only values larger than 1% are given.

k	***q***	B_k^q	***Weight in %***
2	0	-1.93E+00	14.2164186
4	-4	9.25E-03	12.3608476
2	2	9.88E-01	7.27444863
4	-2	4.95E-03	6.61590483
4	-1	-4.67E-03	6.23500341

2	-1	8.02E-01	5.90489908
4	4	-3.93E-03	5.25478747
4	3	3.26E-03	4.36076978
6	-1	2.39E-05	2.99977576
4	-3	2.22E-03	2.97077274
6	5	-2.35E-05	2.94612635
4	2	-2.16E-03	2.88635666
2	-2	3.74E-01	2.75362726
6	-4	-1.84E-05	2.3085846
4	0	-1.71E-03	2.28886999
6	-3	-1.83E-05	2.28819592
6	3	-1.80E-05	2.25621055
6	0	1.68E-05	2.10688923
6	-2	-1.65E-05	2.06345088
6	1	-1.45E-05	1.81871883
4	1	1.13E-03	1.51073449
6	-6	9.21E-06	1.15482399

Table S10: Weight of individual crystal-field parameters on the crystal-field splitting obtained for Dy1 in 2 Only values larger than 1% are given.

k	***q***	B_k^q	***Weight in %***
2	2	-5.34E+00	12.46
4	-1	-3.89E-02	7.33
4	0	3.75E-02	7.08
6	5	9.03E-04	6.41
4	1	3.15E-02	5.95
6	1	8.01E-04	5.68

4	2	2.95E-02	5.58
4	3	-2.53E-02	4.77
6	6	5.97E-04	4.23
2	-2	-1.63E+00	3.80
2	1	1.46E+00	3.40
4	-4	1.72E-02	3.25
6	-4	-4.24E-04	3.01
6	-6	-4.21E-04	2.99
4	4	-1.42E-02	2.67
6	0	3.74E-04	2.65
6	4	-3.74E-04	2.65
6	-2	-3.29E-04	2.34
2	0	-9.73E-01	2.27
6	3	-3.14E-04	2.23
6	2	-2.51E-04	1.78
4	-2	-9.21E-03	1.74
6	-1	-1.61E-04	1.14

Table S11: Weight of individual crystal-field parameters on the crystal-field splitting obtained for **Dy2** in **2** Only values larger than 1% are given.

k	*q*	B_k^q	*Weight in %*
2	2	6.67E+00	14.06
2	0	-4.59E+00	9.68
4	-4	4.49E-02	7.66
4	3	-3.73E-02	6.36
4	4	3.66E-02	6.23
6	-1	8.54E-04	5.47
6	1	-8.05E-04	5.15
6	-5	-6.98E-04	4.47
6	-6	6.84E-04	4.38
6	6	5.08E-04	3.26
6	4	5.01E-04	3.21
4	2	-1.84E-02	3.13
2	-1	-1.43E+00	3.01
6	0	-4.35E-04	2.79
4	0	-1.47E-02	2.51
4	-3	-1.40E-02	2.39
6	3	3.47E-04	2.22
6	-2	3.21E-04	2.05
6	2	3.12E-04	2.00
4	-1	-1.09E-02	1.85
4	1	9.57E-03	1.63
2	-2	5.48E-01	1.15