Supplementary Information

Influence of bridging and chelating co-ligands on the distinct slow magnetic relaxation behaviours in ZnDy complexes

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Table of Contents

Table S1. Crystallographic data and structure refinement parameters of **1**−**4**.

	1	**2**	**3**	**4**
Empirical formula	$C_{30}H_{27}N_6O_{13}Cl_2ZnDy$	$C_{30}H_{37}N_2O_{13}ZnDy$	$C_{28}H_{28}N_3O_{11}ZnDy$	$C_{75}H_{95}N_4O_{21.50}Zn_2Dy_2$
Formula weight	978.34	861.48	810.40	1852.28
Temperature/K	150(2)	150(2)	150(2)	150(2)
Crystal system	monoclinic	monoclinic	monoclinic	monoclinic
Space group	$P2_1/n$	C2/c	$P2_1/c$	$P2_1/c$
a/Å	12.5542(7)	22.205(2)	13.183(4)	15.6478(12)
b/Å	18.0783(10)	10.8751(11)	14.076(4)	22.9141(16)
c/Å	15.5861(9)	27.261(3)	15.695(6)	22.4930(18)
α/°	90	90	90	90
β/°	97.757(2)	91.454(4)	95.718(14)	101.537(3)
γ/°	90	90	90	90
Volume/Å^3	3505.0(3)	6580.8(11)	2898.0(16)	7902.0(10)
Z	4	8	4	4
ρ_{calc} g/cm^3	1.854	1.739	1.857	1.557
μ/mm^{-1}	3.026	3.050	3.453	2.542
F(000)	1932	3448	1604	3748
2*Θ* range for data collection/°	5.008 to 58.39	6.09 to 61.044	4.818 to 61.342	4.966 to 54.482
Reflections collected	80102	91442	78459	154491
Independent reflections	9476 [R_{int} = 0.0235, R_{sigma} = 0.0122]	10022 [R_{int} = 0.0269, R_{sigma} = 0.0137]	8940 [R_{int} = 0.0457, R_{sigma} = 0.0224]	17535 [R_{int} = 0.0627, R_{sigma} = 0.0328]
Data/restraints/parameter	9476/0/492	10022 /2/444	8940 /0/403	17535 /0/986
Goodness-of-fit on F^2	1.141	1.083	1.091	1.101
Final *R* indexes [I>=2σ (I)]	R_1 = 0.0279, wR_2 = 0.0617	R_1 = 0.0163, wR_2 = 0.0393	R_1 = 0.0212, wR_2 = 0.0483	R_1 = 0.0769, wR_2 = 0.1739
Final R indexes [all data]	R_1 = 0.0340, wR_2 = 0.0673	R_1 = 0.0187, wR_2 = 0.0404	R_1 = 0.0278, wR_2 = 0.0536	R_1 = 0.1195, wR_2 = 0.2062
Largest diff. peak/hole / e Å^{-3}	1.386/–1.591	0.665/–0.646	0.874/–1.261	2.553/–2.041

Table S2. SHAPE analysis of the Dy(III) ion for complex **1**.

Label	Shape	Symmetry	**1** (Distortion)
			Dy1
DP-10	Decagon	D_{10h}	36.905
EPY-10	Enneagonal pyramid	C_{9v}	24.341
OBPY-10	Octagonal bipyramid	D_{8h}	16.713
PPR-10	Pentagonal prism	D_{5h}	9.170
PAPR-10	Pentagonal antiprism	D_{5d}	11.286
JBCCU-10	Bicapped cube (Elongated square bipyramid J15)	D_{4h}	11.519
JBCSAPR-10	Bicapped square antiprism (Gyroelongated square bipyramid J17)	D_{4d}	6.404
JMBIC-10	Metabidiminished icosahedron (J62)	C_{2v}	8.158
JATDI-10	Augmented tridiminished icosahedron (J64)	C_{3v}	17.535
JSPC-10	Sphenocorona (J87)	C_{2v}	**3.022**
SDD-10	Staggered dodecahedron	D_2	5.108
TD-10	Tetradecahedron	C_{2v}	4.965
HD-10	Hexadecahedron	D_{4h}	7.884

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Table S3. SHAPE analysis of the Dy(III) ion for complexes **2**, **3** and **4**.

Label	Shape	Symmetry	**2** (Distortion)	**3** (Distortion)	**4** (Distortion)	
			Dy1	Dy1	DyA	DyB
EP-9	Enneagon	D_{9h}	33.888	34.886	33.516	34.110
OPY-9	Octagonal pyramid	C_{8v}	24.778	25.224	22.066	25.280
HBPY-9	Heptagonal bipyramid	D_{7h}	16.858	16.348	15.707	16.133
JTC-9	Johnson triangular cupola J3	C_{3v}	14.051	15.711	15.033	15.573
JCCU-9	Capped cube J8	C_{4v}	8.556	9.107	7.438	8.646
CCU-9	Spherical-relaxed capped cube	C_{4v}	6.867	7.387	5.957	6.834
JCSAPR-9	Capped square antiprism J10	C_{4v}	4.354	4.432	3.700	4.761
CSAPR-9	Spherical capped square antiprism	C_{4v}	3.863	3.929	**2.640**	4.312
JTCTPR-9	Tricapped trigonal prism J51	D_{3h}	4.216	5.419	4.234	5.438
TCTPR-9	Spherical tricapped trigonal prism	D_{3h}	3.309	4.398	3.476	4.623
JTDIC-9	Tridiminished icosahedron J63	C_{3v}	10.827	11.404	9.506	10.974
HH-9	Hula-hoop	C_{2v}	6.553	5.839	7.376	5.315
MFF-9	Muffin	C_s	**2.947**	**2.990**	**3.105**	**3.263**

Table S4. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **1** by the single-component generalized Debye model at zero and 1000 Oe dc fields.

T (K)	χ_S (emu/mol)	$\Delta\chi$ (emu/mol)	τ (s)	α
At zero dc field				
1.9	2.30697	3.04097	6.02062E-5	0.05937
2.09	2.11335	2.70905	6.02577E-5	0.05208
2.29	2.01893	2.42551	6.28703E-5	0.04155
2.48	1.78118	2.30819	5.92629E-5	0.05257
2.67	1.71971	2.06475	6.19188E-5	0.03877
2.87	1.603	1.92383	6.13093E-5	0.03741
3.06	1.50696	1.79674	6.12411E-5	0.04246
3.25	1.34893	1.76024	5.73886E-5	0.04736
3.45	1.31004	1.61969	5.89545E-5	0.04163
3.64	1.21221	1.55859	5.71967E-5	0.04351
3.83	1.13565	1.49432	5.60818E-5	0.04762
4.03	1.13565	1.49432	5.60818E-5	0.04762
4.22	1.07129	1.3183	5.75306E-5	0.03782
4.42	0.98964	1.29668	5.47705E-5	0.04772
4.61	0.99401	1.19614	5.73041E-5	0.03291
4.8	0.98482	1.11982	5.78966E-5	0.03476
5	0.89619	1.12595	5.47014E-5	0.0359
5.5	0.81253	1.0267	5.16622E-5	0.04078
6	0.67194	1.01341	4.44275E-5	0.03785
6.5	0.70981	0.85446	4.56941E-5	0.03169
At 1000 Oe dc field				
1.9	0.15646	6.83504	0.53286	0.39895
2.09	0.14555	5.90392	0.36651	0.39309
2.29	0.14145	5.22739	0.25381	0.38087
2.48	0.13538	4.67736	0.17567	0.36954
2.67	0.13498	4.16811	0.11828	0.34721
2.87	0.13602	3.74413	0.08066	0.3238
3.06	0.13695	3.39498	0.05627	0.29923
3.25	0.13808	3.11331	0.04046	0.27412
3.45	0.1359	2.88849	0.02963	0.25563
3.64	0.13798	2.68855	0.0218	0.23554
3.83	0.13665	2.52344	0.01629	0.21527
4.03	0.13339	2.38952	0.0123	0.20222
4.22	0.13248	2.26505	0.00935	0.18459
4.42	0.12923	2.16774	0.00726	0.17339

4.61	0.13179	2.06266	0.00562	0.15762
4.8	0.12925	1.97693	0.00439	0.14773
5	0.12659	1.89416	0.00342	0.14008
5.5	0.1207	1.7171	0.00187	0.12194
6	0.11703	1.57108	0.00106	0.10543
6.5	0.11538	1.44692	6.31104E-4	0.09046
7	0.12665	1.32866	3.92451E-4	0.07883
7.5	0.13071	1.23129	2.47604E-4	0.07642
8	0.13135	1.1457	1.60013E-4	0.07223
8.5	0.13709	1.0654	1.05583E-4	0.06775
9	0.20331	0.92987	7.82507E-5	0.04267
9.5	0.24896	0.82633	5.85144E-5	0.02713
10	0.27448	0.74842	4.1608E-5	0.02412

Table S5. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **2** by the single-component generalized Debye model at zero and 1000 Oe dc fields.

T (K)	χ_S (emu/mol)	$\Delta\chi$ (emu/mol)	τ (s)	α
At zero dc field				
1.9	1.87668	3.26283	1.04119E-4	0.03877
2.09	1.54462	3.11523	9.49454E-5	0.0492
2.29	1.358	2.9421	9.07515E-5	0.05263
2.48	1.16613	2.78527	8.5878E-5	0.05331
2.67	1.11951	2.53459	8.73524E-5	0.05117
2.87	0.92985	2.53763	7.9977E-5	0.05992
3.06	0.85826	2.39586	7.87465E-5	0.06008
3.25	0.78312	2.28515	7.67839E-5	0.05997
3.45	0.73065	2.16865	7.53212E-5	0.05914
3.64	0.68459	2.0598	7.36752E-5	0.05745
4.03	0.68165	1.80012	7.39844E-5	0.04751
4.22	0.66601	1.70568	7.29151E-5	0.04342
4.42	0.60524	1.66773	6.85771E-5	0.04717
4.61	0.58735	1.59301	6.6307E-5	0.04256
4.8	0.54817	1.54664	6.27471E-5	0.04013
5	0.51205	1.50187	5.88085E-5	0.03846
5.5	0.40164	1.4325	4.75439E-5	0.03174
6	0.48881	1.20989	4.43168E-5	0.02466
6.5	0.38361	1.19177	3.22233E-5	0.03201
7	0.34127	1.12668	2.41133E-5	0.03288

7.5	0.34644	1.02832	1.80517E-5	0.03882
8	0.35881	0.93323	1.32779E-5	0.04733
8.5	0.06756	1.15186	6.56051E-6	0.0681
9	0.60437	0.55052	9.25159E-6	0.07755
9.5	0.4154	0.68054	4.29508E-6	0.10082
At 1000 Oe dc field				
1.9				
2.09	0.26589	8.15878	1.0065	0.28934
2.29	0.26171	4.64832	0.23192	0.19795
2.48	0.25302	3.73233	0.10374	0.14689
2.67	0.24226	3.37631	0.05952	0.12323
2.87	0.23585	3.09192	0.03526	0.10079
3.06	0.22403	2.91349	0.02302	0.09897
3.25	0.21564	2.74421	0.01521	0.08845
3.45	0.20753	2.59271	0.01031	0.07825
3.64	0.20007	2.45695	0.00717	0.06899
3.83	0.19113	2.34108	0.0051	0.06276
4.03	0.18392	2.23292	0.00369	0.05646
4.22	0.177	2.1379	0.00272	0.05234
4.42	0.16847	2.06092	0.00204	0.05204
4.61	0.16255	1.9792	0.00154	0.04874
4.8	0.15585	1.90768	0.00117	0.04787
5	0.14925	1.83834	8.88631E-4	0.04694
5.5	0.13476	1.69584	4.55044E-4	0.04486
6	0.11684	1.56966	2.40953E-4	0.04725
6.5	0.09217	1.47428	1.3259E-4	0.05236
7	0.0724	1.38862	7.45851E-5	0.0549
7.5	0.02524	1.34332	4.11133E-5	0.06431
8	0.04898	1.23783	2.45695E-5	0.06493

Table S6. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **4** by the single-component generalized Debye model at zero dc field.

T (K)	χ_S (emu/mol)	$\Delta\chi$ (emu/mol)	τ (s)	α
1.9	1.73238	4.92454	1.41148E-4	0.11624
2.1	1.63916	4.32748	1.40498E-4	0.09385
2.3	1.49929	4.01055	1.38204E-4	0.09602
2.5	1.37036	3.70237	1.35747E-4	0.09627
2.7	1.26303	3.44233	1.3367E-4	0.09623
2.87	1.17797	3.21006	1.32253E-4	0.09499
3.06	1.09237	3.01979	1.2992E-4	0.09636
3.25	1.02511	2.84576	1.28073E-4	0.09598
3.45	0.96401	2.68879	1.26297E-4	0.09547
3.64	0.90369	2.55008	1.24209E-4	0.09467
3.83	0.8718	2.40367	1.2394E-4	0.09208
4.03	0.86629	2.24657	1.25121E-4	0.08469
4.22	0.83429	2.13524	1.23642E-4	0.08119
4.42	0.78709	2.05604	1.20507E-4	0.08341
4.61	0.74904	1.97559	1.17242E-4	0.08223
4.8	0.72585	1.88918	1.15157E-4	0.07898
5	0.68836	1.82258	1.11383E-4	0.07855
5.5	0.62528	1.65652	1.02828E-4	0.07203
6	0.56549	1.52566	9.26699E-5	0.06696
6.5	0.51936	1.41483	8.29143E-5	0.06155
7	0.55946	1.25185	8.14543E-5	0.04719
7.5	0.51612	1.1752	7.06286E-5	0.04661
8	0.50993	1.0758	6.33766E-5	0.04016
8.5	0.50503	0.98831	5.61362E-5	0.03715
9	0.48805	0.92247	4.83038E-5	0.03604
9.5	0.50778	0.82864	4.33621E-5	0.0304
10	0.44471	0.825	3.3811E-5	0.03911

Table S7. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **4** by the double-component generalized Debye model at 1000 Oe dc field.

T (K)	χ_S (emu/mol)	$\Delta\chi_1$ (emu/mol)	τ_1 (s)	α_1	$\Delta\chi_2$ (emu/mol)	τ_2 (s)	α_2
2	0.15882	0.85965	0.75917	0*	5.3854	0.28936	0.3937
2.2	0.14796	0.86504	0.76553	0*	4.82582	0.19614	0.37908
2.4	0.13095	0.80571	0.64823	0*	4.35007	0.12615	0.37099
2.6	0.12269	0.86632	0.57717	0*	3.83818	0.07618	0.35058
2.8	0.11397	0.87563	0.50063	0.00982	3.49207	0.04999	0.33483
3.0	0.10609	1.00595	0.38273	0.04404	3.07018	0.03007	0.31649
3.2	0.10141	1.08007	0.2919	0.07405	2.75863	0.01935	0.29857
3.4	0.09954	1.17595	0.21452	0.10907	2.45615	0.01245	0.27834
3.6	0.09315	1.1213	0.15604	0.10168	2.28548	0.0088	0.27065
3.8	0.08734	1.09566	0.11704	0.10207	2.13192	0.00628	0.26003
4.0	0.07991	1.01846	0.09078	0.09303	2.04916	0.00477	0.25846
4.2	0.07482	1.02272	0.06675	0.09927	1.90527	0.0035	0.25257
4.4	0.06349	0.92802	0.05317	0.08664	1.87641	0.00281	0.26027
4.6	0.06268	0.95342	0.03921	0.10191	1.73423	0.00209	0.2504
4.8	0.0555	0.92509	0.03081	0.08966	1.65434	0.00162	0.24829
5	0.04772	0.84216	0.02543	0.0692	1.63886	0.00135	0.25611
5.5	0.02978	0.75765	0.01497	0.04907	1.51576	7.93367E-4	0.26566
6	-0.00221	0.6705	0.00922	0.03124	1.4513	4.9278E-4	0.28881
6.5	-0.0172	0.64168	0.00574	0.0462	1.33831	3.05222E-4	0.29698
7	0.01111	0.61321	0.00378	0.03573	1.19859	2.05933E-4	0.27886
7.5	0.0374	0.58511	0.0026	0.03386	1.08821	1.46739E-4	0.27082
8	0.10005	0.55627	0.00182	0.04615	0.95954	1.1822E-4	0.25717
8.5	0.16436	0.52865	0.00133	0.04813	0.82841	9.67172E-5	0.22268

***** The values were fixed to avoid overparameterization

Table S8. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **3** by the single-component generalized Debye model at 600 Oe dc field.

T (K)	χ_S (emu/mol)	$\Delta\chi$ (emu/mol)	τ (s)	α
At 600 Oe dc field				
2	1.60544	4.08057	2.80178E-4	0.21775
2.18	1.3853	3.81145	1.33514E-4	0.23704
2.37	1.22632	3.68233	7.38287E-5	0.23294
2.56	1.16392	3.48997	4.75645E-5	0.22106
2.75	1.27223	3.14885	3.59682E-5	0.20336
2.93	1.34165	2.85851	2.91363E-5	0.17514
3.12	1.12217	2.88231	2.0777E-5	0.15874
3.31	1.85551	1.96847	2.51867E-5	0.11767
3.49	1.34429	2.31208	1.32998E-5	0.13366

Table S9. Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **3** by the single-component generalized Debye model at 2000 Oe dc field.

T (K)	χ_S (emu/mol)	$\Delta\chi_1$ (emu/mol)	τ_1 (s)	α_1	$\Delta\chi_2$ (emu/mol)	τ_2 (s)	α_2	$\Delta\chi_3$ (emu/mol)	τ_3 (s)	α_3
2	0.05*	1.68788	1.6171E-4	0.33373	1.59521	0.01499	0.34588	1.61549	0.41128	0.05209
2.2	0.05*	1.75646	1.1900E-4	0.33884	1.55478	0.01836	0.37595	1.45713	0.38138	0.01568
2.4	0.05*	2.04636	1.0149E-4	0.36345	1.05726	0.01678	0.29654	1.38027	0.32196	0.05367
2.6	0.05584	2.17175	8.1836E-5	0.33972	1.00073	0.01971	0.31571	1.04371	0.30008	0.00833
2.8	0.11104	2.25702	7.0186E-5	0.3087	0.73033	0.01618	0.27242	0.94807	0.26982	0.04565
3	0.26446	2.18544	6.3284E-5	0.25831	0.61273	0.01499	0.27877	0.77649	0.25797	0.05019
3.2	0.13822	2.38051	4.3297E-5	0.24321	0.5317	0.01612	0.30438	0.59418	0.25598	0.02763
3.4	0.05*	2.50095	2.9456E-5	0.21995	0.43899	0.01452	0.33159	0.49145	0.25089	0.02454
3.6	0.05*	2.47533	2.0296E-5	0.17306	0.42696	0.0108	0.45348	0.3875	0.24467	0.00697
3.8	0.05*	2.49337	1.3393E-5	0.18225	0.32158	0.01068	0.45931	0.32542	0.24333	0*
4	0.05*	2.46844	8.1228E-6	0.20491	0.28841	0.00994	0.5318	0.24789	0.24983	0*
4.2	0.05*	2.00248	5.4876E-6	0.2682	0.22495	0.00884	0.55535	0.20021	0.25655	0*
4.4	0.05*	2.49255	9.6547E-7	0.48193	0.09677	0.02614	0.3956	0.16478	0.28522	0*
4.6	0.05*	2.44774	4.2254E-7	0.53342	0.06624	0.03577	0.38084	0.13123	0.30348	0*
4.8	0.05*	2.40888	1.1955E-7	0.61033	0.02829	0.05052	0.15898	0.10917	0.32759	0*
5	0.05*	2.33448	1.3388E-7	0.56917	0.03311	0.05749	0.32429	0.08664	0.33935	0*

***** The values were fixed to avoid overparameterization

Table S10. The splitting of ^{6}H15/2 the lowest multiplets for **1** calculated by CASSCF/SINGLE_ANISO together with *g*-values for each Kramers doublets

E (cm^{-1})	g_x	g_y	g_z
0	0.001	0.019	18.847
96	0.110	0.136	17.752
143	0.110	1.122	17.766
163	0.415	1.914	13.404
236	0.244	1.023	12.899
280	9.168	8.150	4.272
372	0.037	0.290	16.036
536	0.032	0.113	19.085

Table S11. The splitting of ^{6}H15/2 the lowest multiplets for **2** calculated by CASSCF/SINGLE_ANISO together with *g*-values for each Kramers doublets

E (cm^{-1})	g_x	g_y	g_z
0	0.008	0.023	19.070
96	0.665	1.606	15.215
138	0.829	1.422	14.482
177	2.594	4.641	11.331
208	3.175	4.461	12.139
265	1.558	2.026	13.374
432	0.061	0.075	16.148
611	0.003	0.016	18.821

Table S12. The splitting of ^{6}H15/2 the lowest multiplets for **3** calculated by CASSCF/SINGLE_ANISO together with *g*-values for each Kramers doublets

E (cm^{-1})	g_x	g_y	g_z
0	1.292	6.997	12.922
17	2.538	4.107	9.432
90	3.812	4.486	10.731
132	0.834	4.071	13.679
183	1.153	3.326	12.274
222	0.942	1.308	16.554
390	0.130	0.180	16.202
568	0.022	0.056	18.709

Table S13. The splitting of ^{6}H15/2 the lowest multiplets for **4-A** calculated by CASSCF/SINGLE_ANISO together with *g*-values for each Kramers doublets

E (cm^{-1})	g_x	g_y	g_z
0	0.080	0.197	19.066
86	0.844	1.205	16.091
154	2.033	3.176	13.962
210	1.908	5.126	10.863
284	2.187	4.845	10.255
351	1.502	1.715	15.352
472	0.180	0.276	13.737
645	0.016	0.020	19.193

Table S14. The splitting of ^{6}H15/2 the lowest multiplets for **4-B** calculated by CASSCF/SINGLE_ANISO together with *g*-values for each Kramers doublets

E (cm^{-1})	g_x	g_y	g_z
0	0.009	0.015	19.435
100	1.003	1.842	17.746
134	0.336	1.738	14.638
181	3.852	5.045	11.565
215	2.146	2.532	14.997
261	1.008	1.280	13.595
451	0.071	0.080	16.049
635	0.013	0.044	18.667

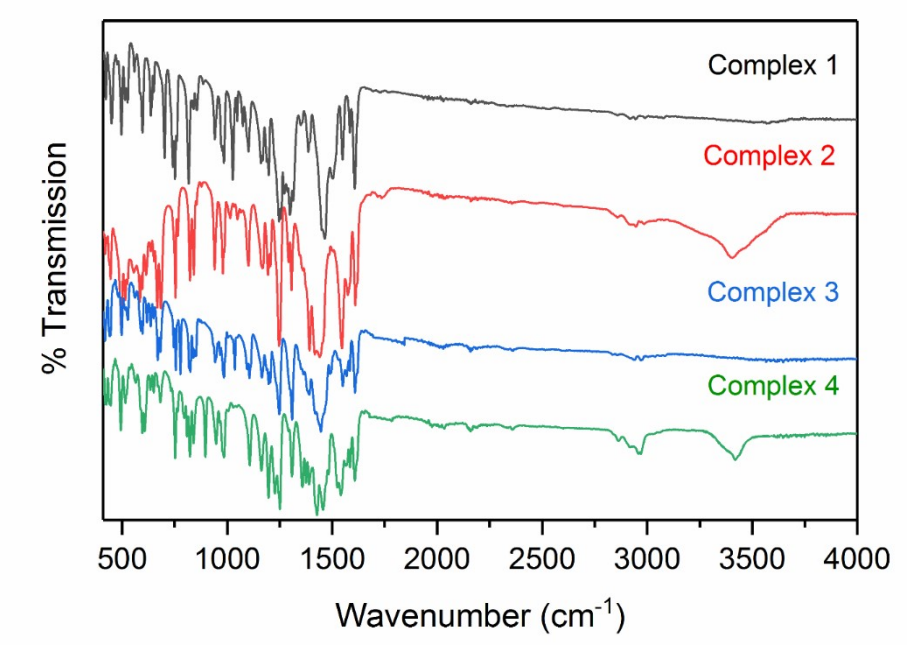


Fig. S1. IR spectra of **1-4**.

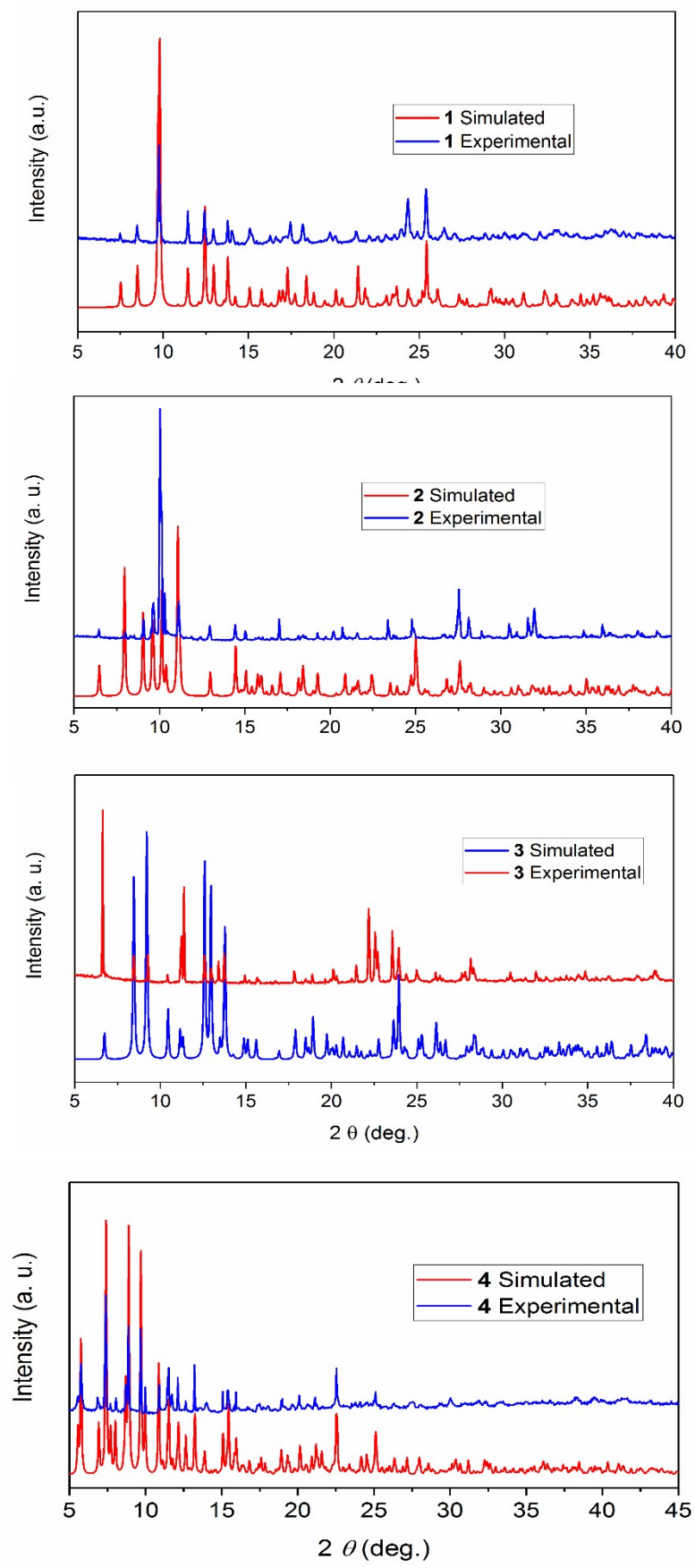


Fig. S2. Experimental and simulated PXRD patterns of **1-4**

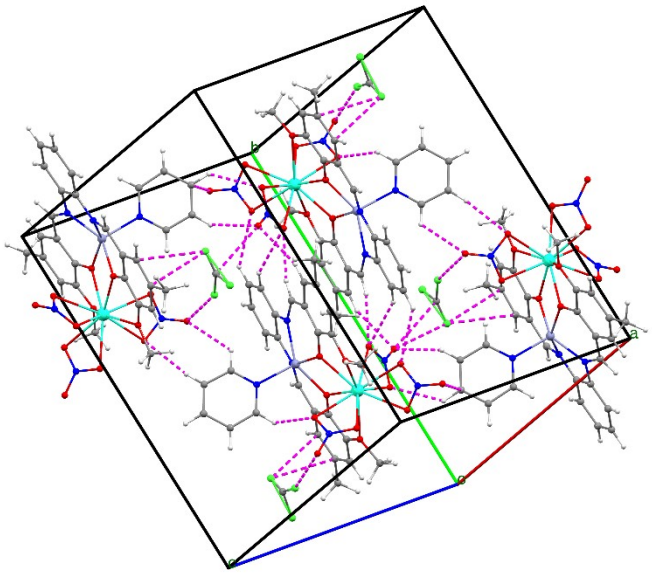


Fig. S3. A part of crystal packing for **1**.

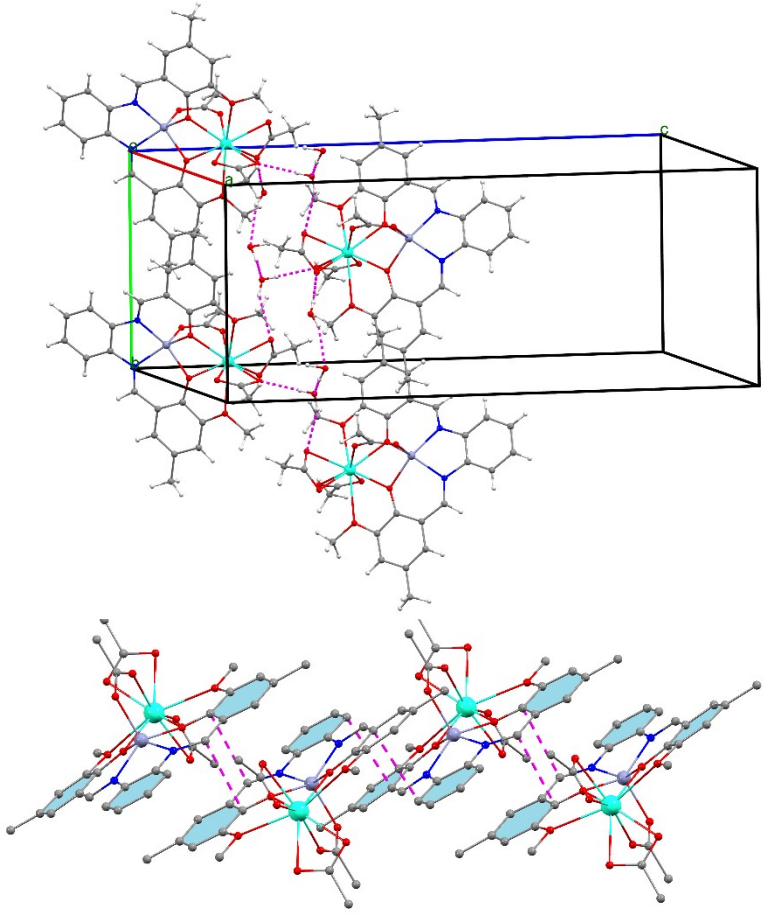


Fig. S4. A part of crystal packing showing hydrogen bonding interaction (top) and π···π stacking interaction (bottom) for **2**.

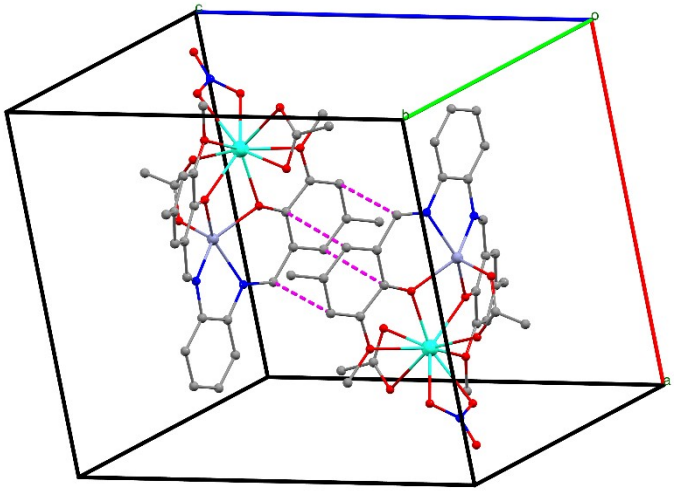


Fig. S5. A part of crystal packing for **3**.

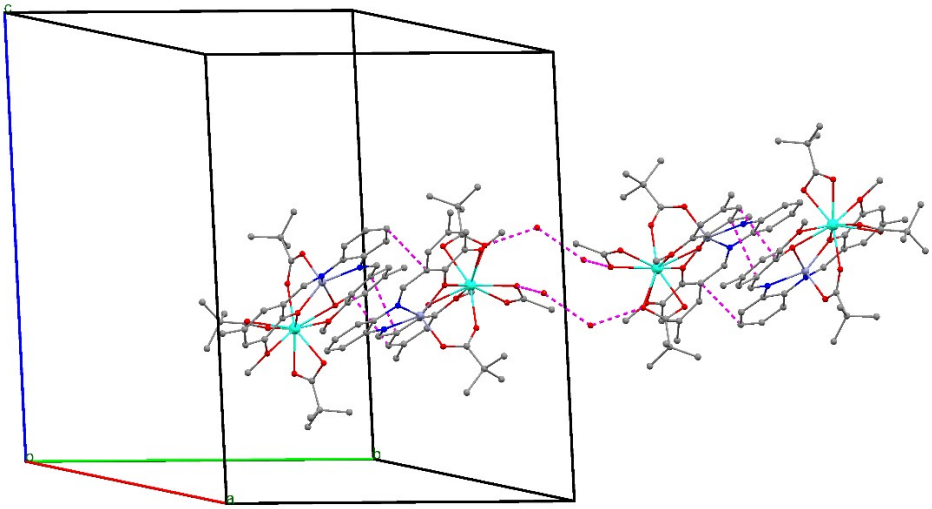


Fig. S6. A part of crystal packing for **4**.

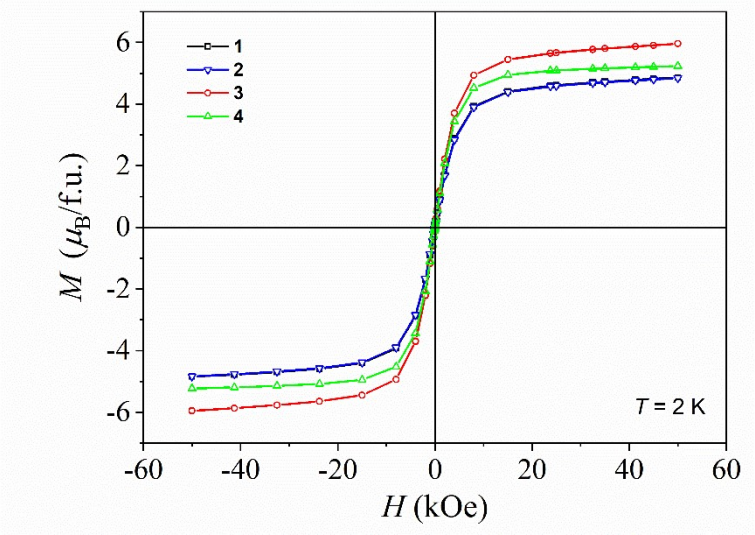


Fig. S7. Magnetization plots of **1–4** at 2 K.

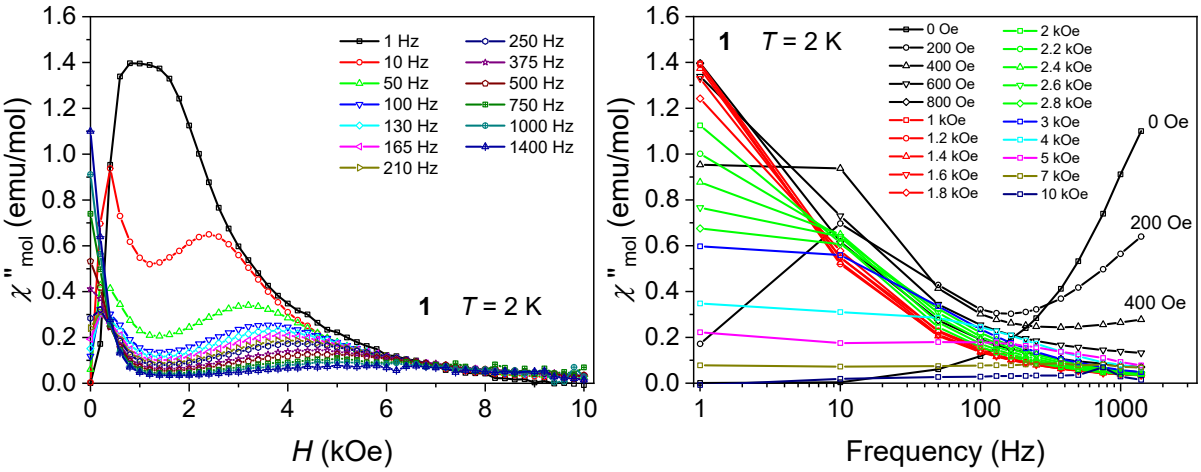


Fig. S8. Out-of-phase ac susceptibility as a function of dc magnetic field (left) and frequency (right) for **1** measured at 2 K.

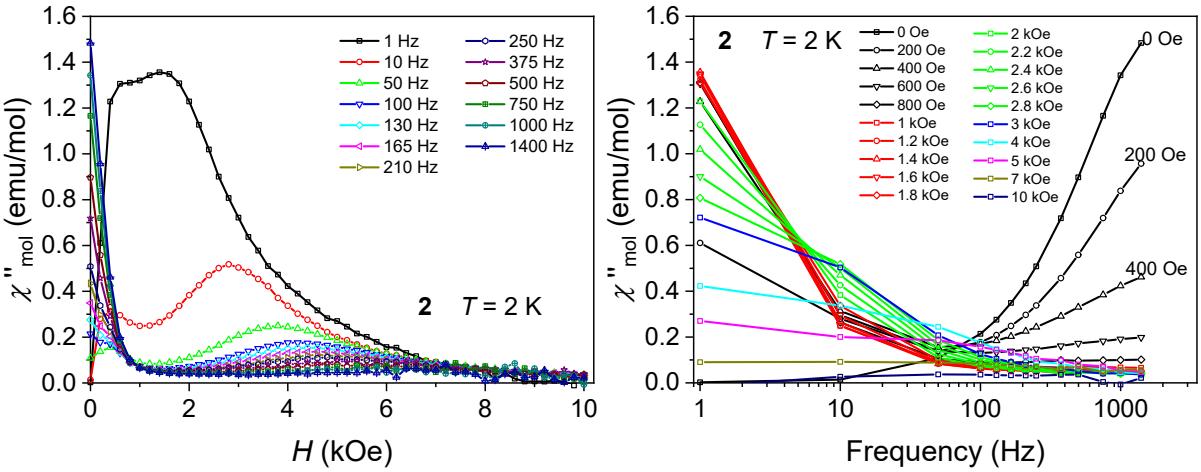


Fig. S9. Out-of-phase ac susceptibility as a function of dc magnetic field (left) and frequency (right) for **2** measured at 2 K.

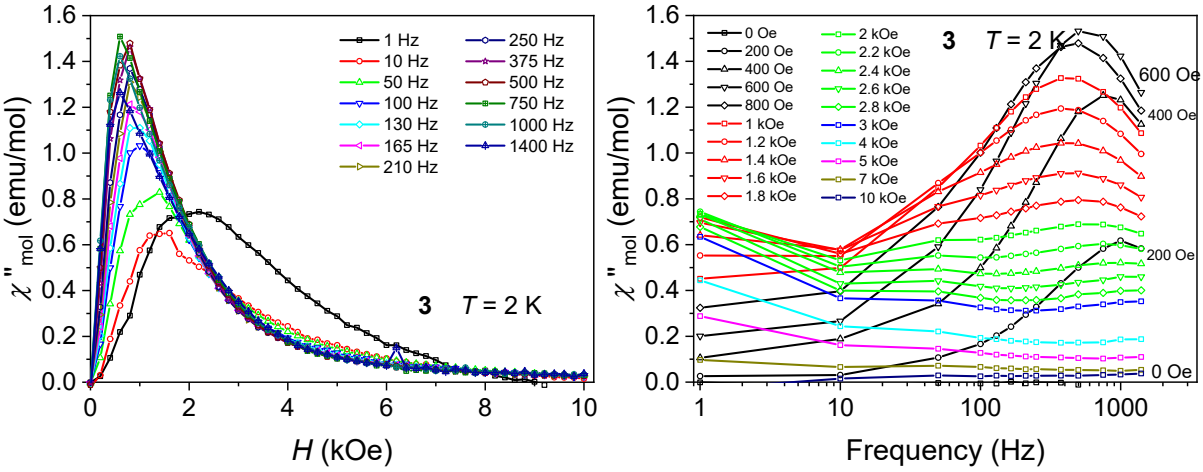


Fig. S10. Out-of-phase ac susceptibility as a function of dc magnetic field (left) and frequency (right) for **3** measured at 2 K.

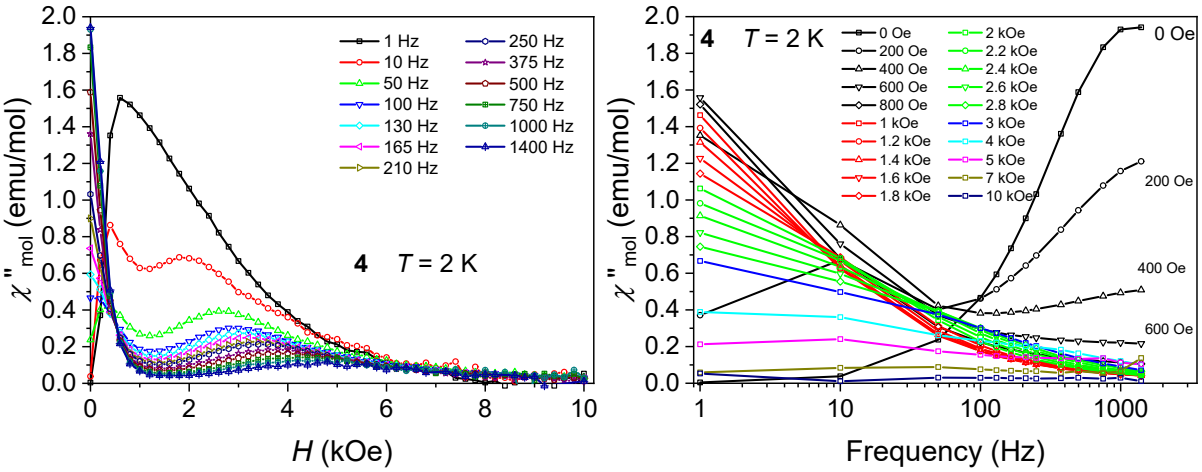


Fig. S11. Out-of-phase ac susceptibility as a function of dc magnetic field (left) and frequency (right) for **4** measured at 2 K. The magnetic data are scaled per one Dy^{III} ion.

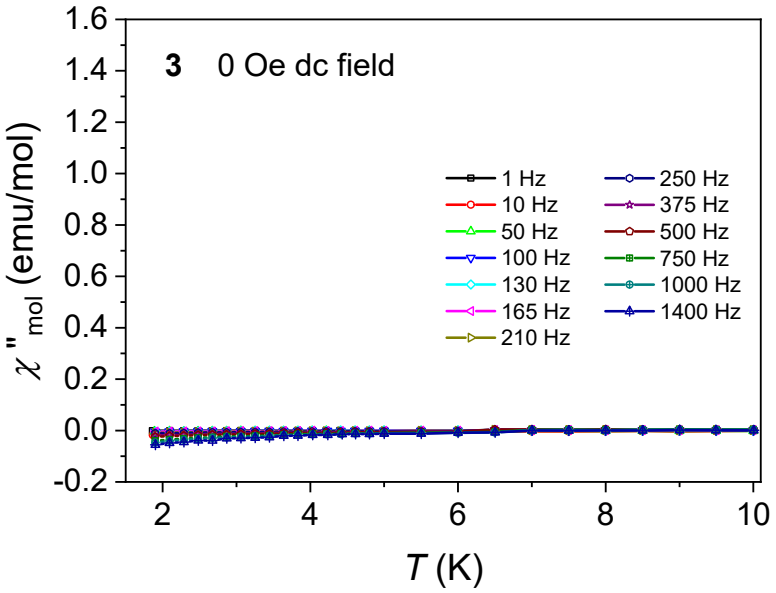


Fig. S12. The variable temperature out-of-phase ac susceptibility data of **-3** at zero dc field.

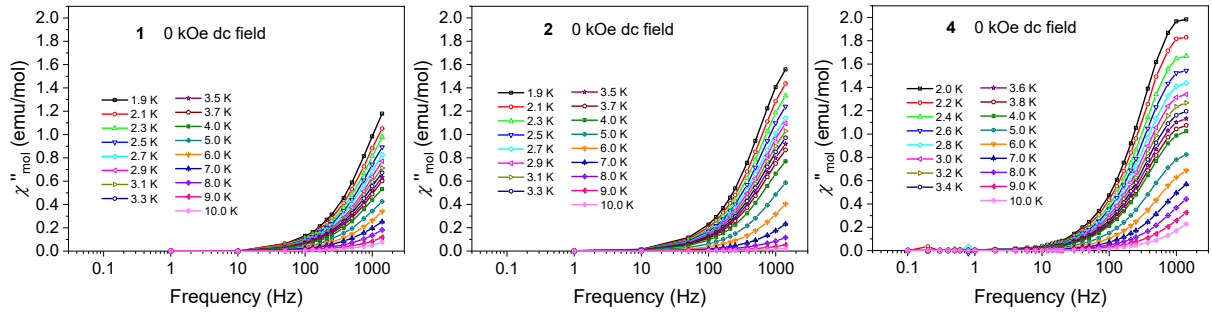


Fig. S13. Frequency dependence of the out-of-phase ac susceptibility component of **1**, **2** and **4** measured in the temperature range 1.9–10 K and at zero dc field. The magnetic data are scaled per one Dy^{III} ion.

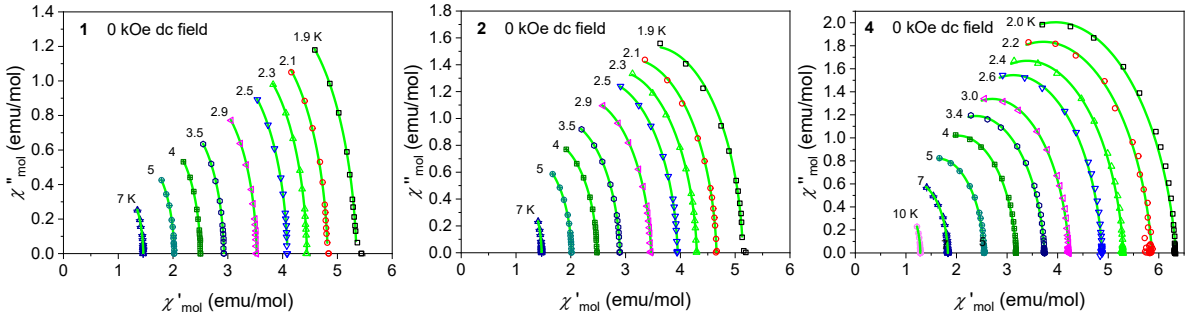


Fig. S14. Cole-Cole plots for **1**, **2** and **4** at zero dc field. The solid lines are the best fits to the experiments with the generalized Debye model.

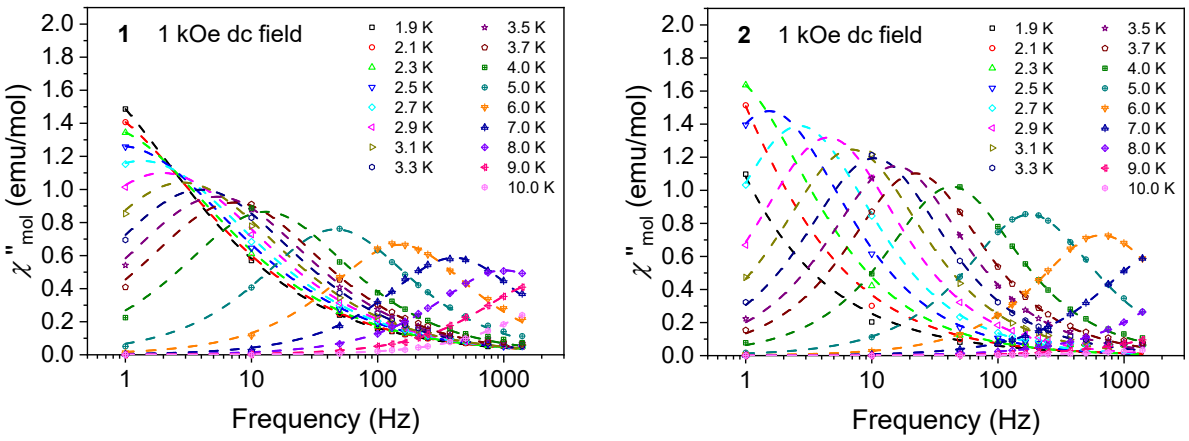


Fig. S15. Frequency dependence of the out-of-phase ac susceptibility component of **1** and **2** measured in the temperature range 1.9–10 K and at 1000 Oe dc field. Solid lines represent the best fit by the generalised Debye model.

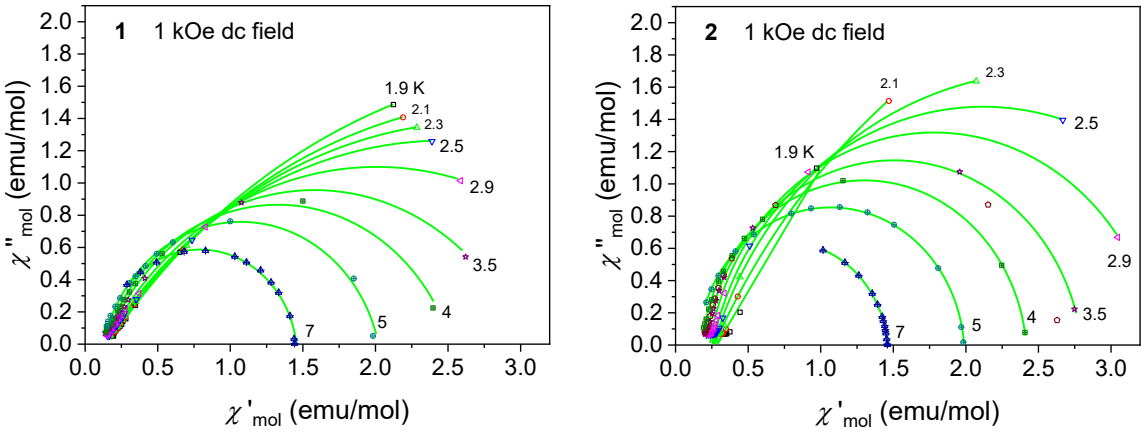


Fig. S16. Cole-Cole plots for **1** and **2** at 1000 Oe dc field. The solid lines are the best fits to the experiments with the generalized Debye model.

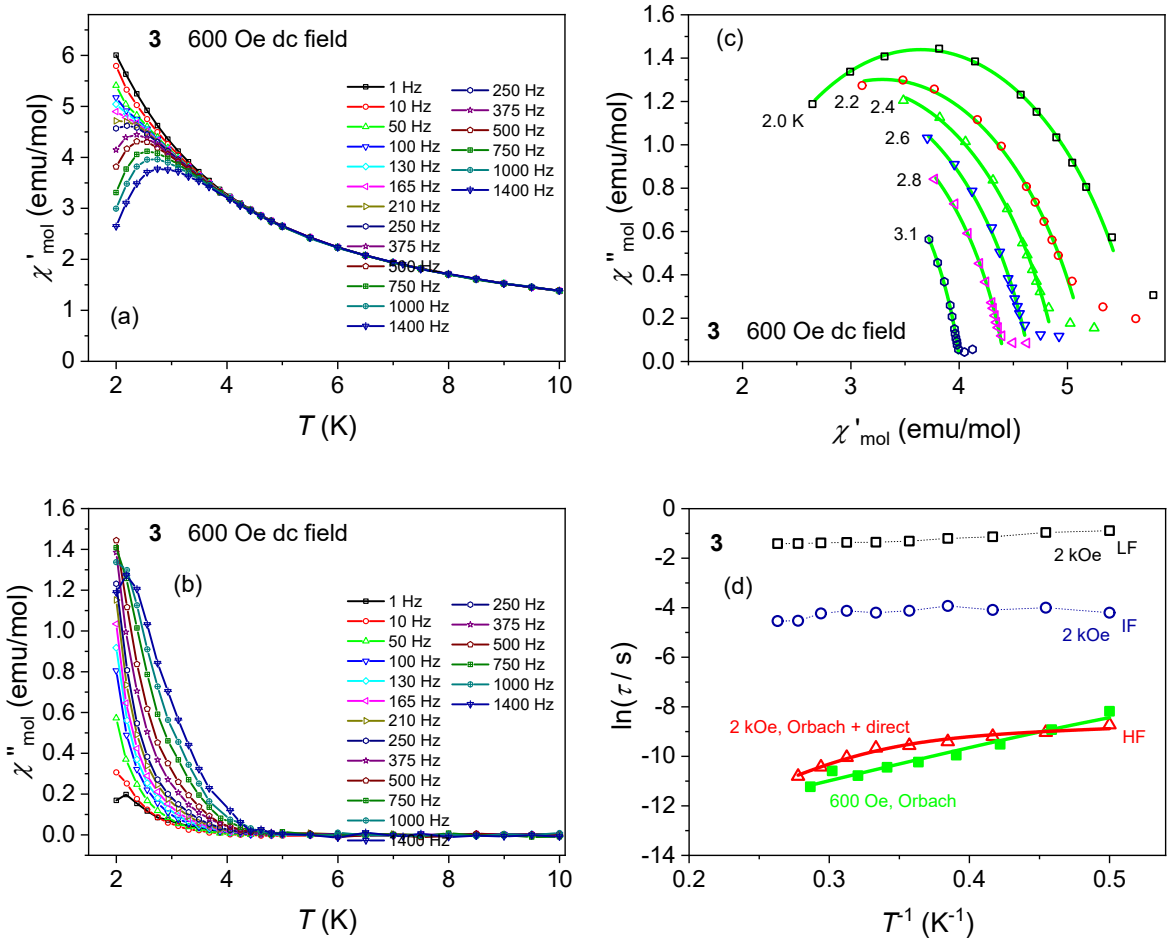


Fig. S17. Temperature dependence of in-phase (a) and out-of-phase (b) ac susceptibilities, Cole-Cole plots fitted by the generalized Debye model (c) at 600 Oe dc field and the logarithmic magnetisation relaxation time versus T–1 plots at 600 and 2000 Oe dc fields for **3**.

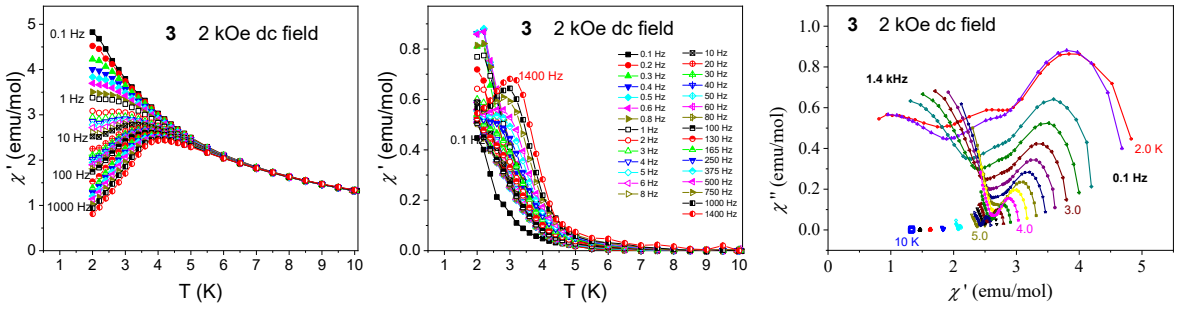


Fig. S18. Temperature dependence of in-phase (a) and out-of-phase (b) ac susceptibilities and Cole-Cole plots fitted by three-set Debye model (c) for **3** at 2000 Oe dc field.

1

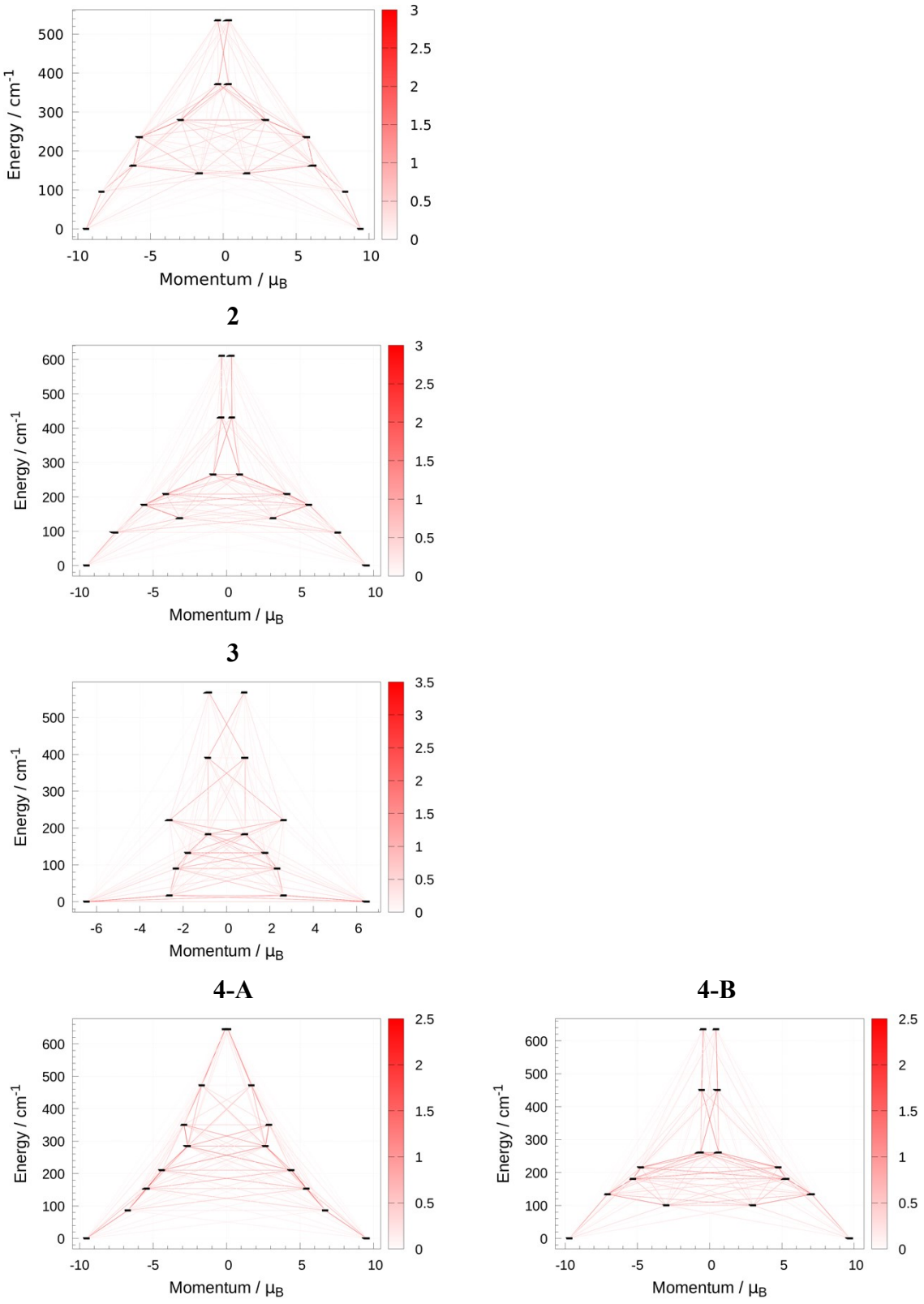


Fig. S19. Plots made by SINGLE_ANISO module based on CASSCF calculations for Dy^{III} complexes of **1-4**. The red lines represent the transition matrix elements of the magnetic moment connecting two states.

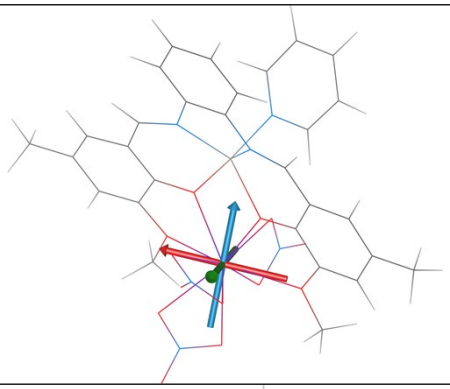
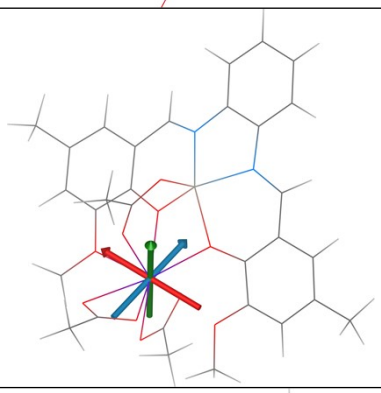
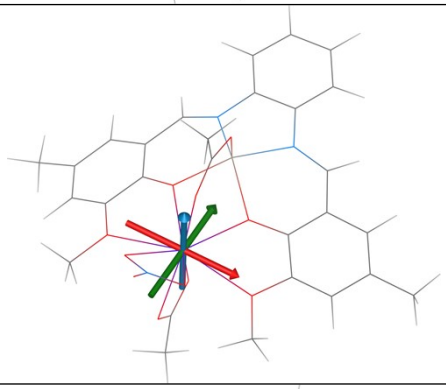
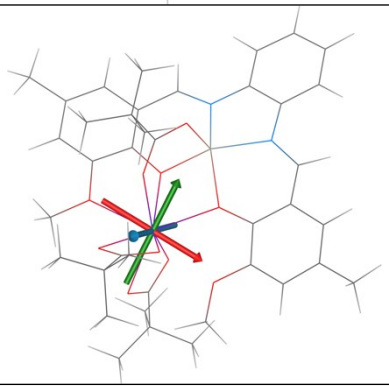
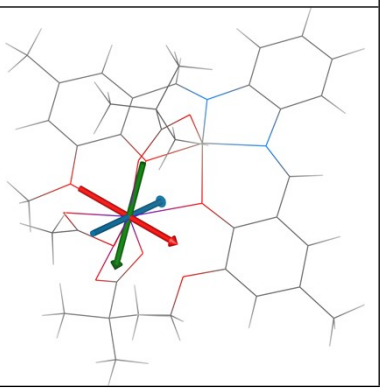
1			
2			
3			
4-A		**4-B**	

Fig. S20. The molecular structures of **1-4** derived from the experimental X-ray geometry used for CASSCF calculations overlaid with of g-tensor of the first Kramers doublet (x/y/z-axis are showed as red/green/blue arrows).