Supporting Information (SI)

Single molecule magnetism in a family of related $\{Co^{III}_2Dy^{III}_2\}$ butterfly complexes: Effects of ligand replacement on the dynamics of magnetic relaxation.

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Packing Diagrams (Figures S1-S3).

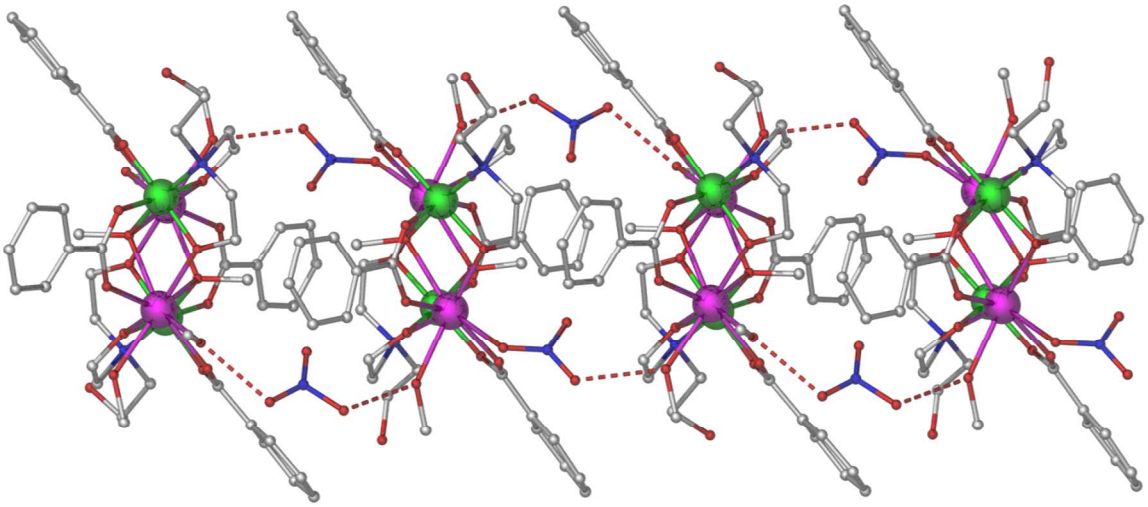

Figure S1. 1-D H-bonded chains via the nitrate and MeOH molecules for **1**. H-bonds are indicated by the dashed red lines.

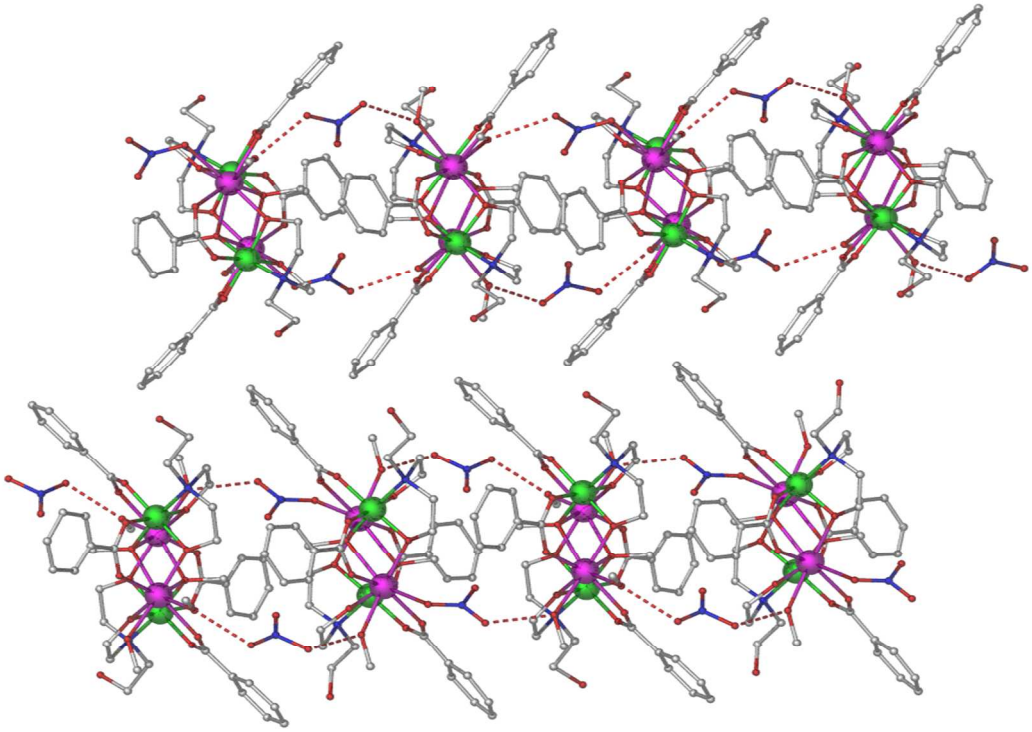

Figure S2. Aromatic C-H···π interactions between the 1-D H-bonded chains for **1**, shown along the c-axis.

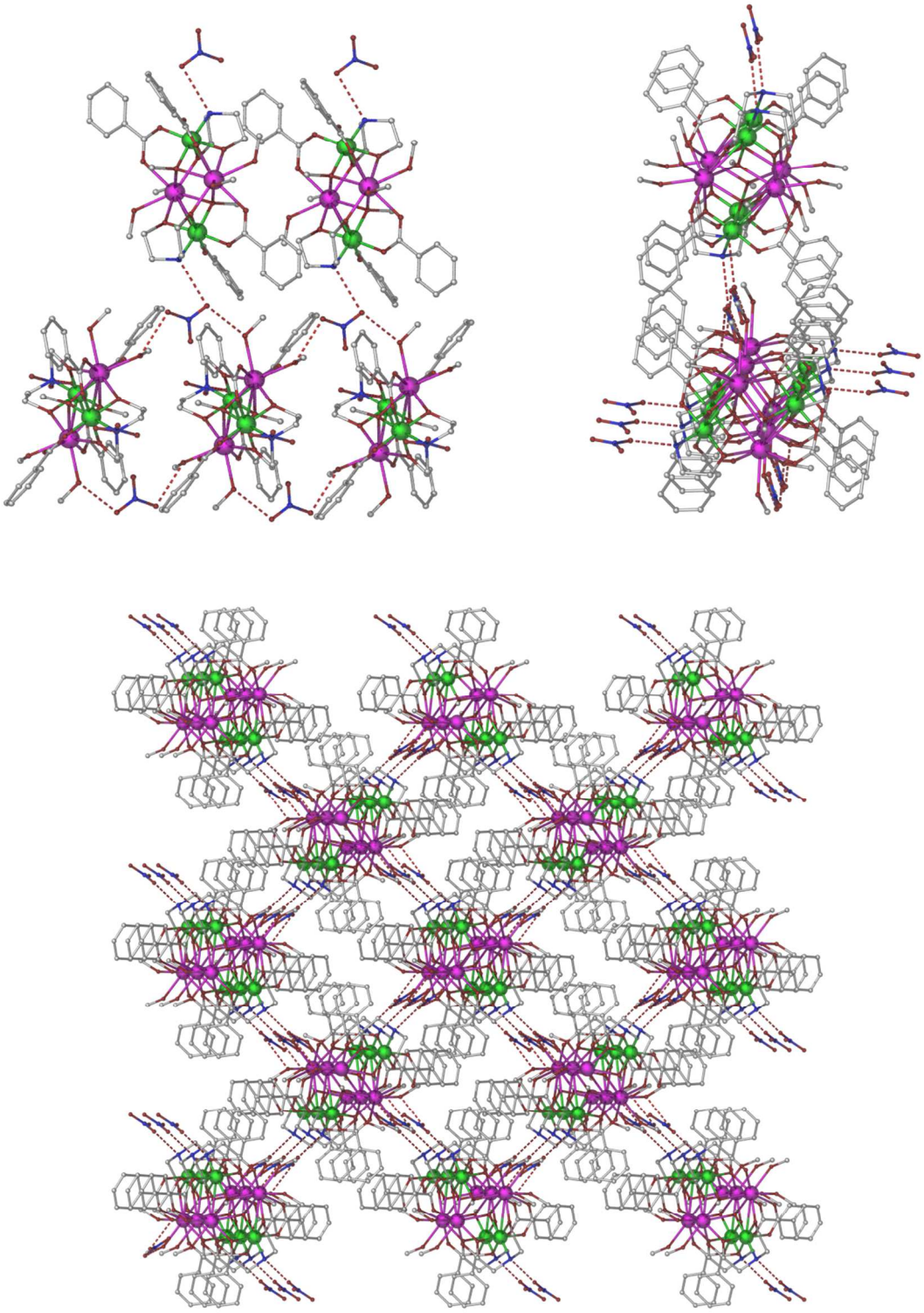

Figure S3. (top right and left) Highlighting the H-bond interactions via the NO_3^- anion; (bottom) 3-D H-bonded Packing arrangement of **2**.

Magnetic data (Figures S4-S8).

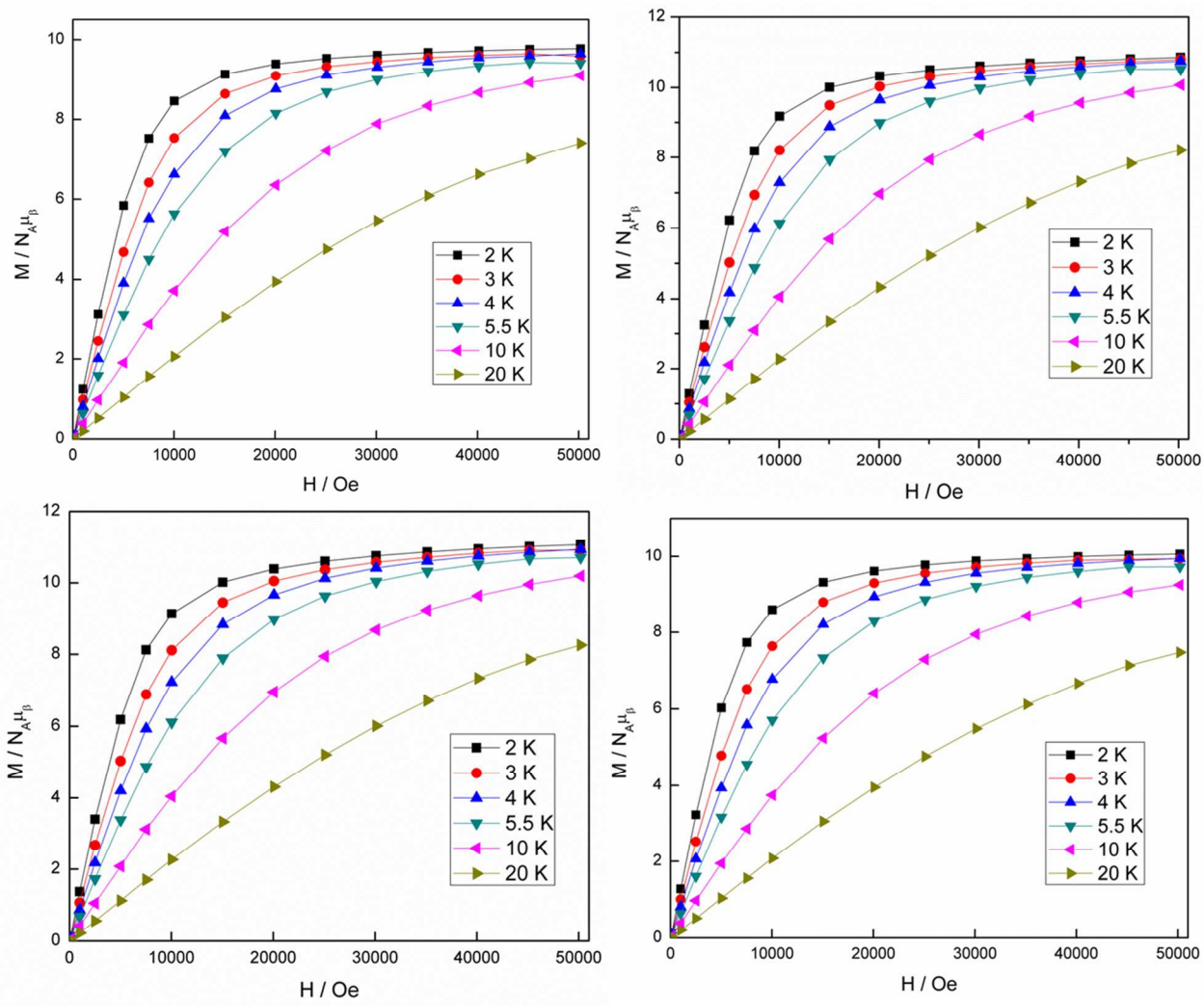

Figure S4. Isothermal *M vs H* plot for compound **1** (top left), **2** (top right), **3** (bottom left) and **4** (bottom right). The solid lines just join the points.

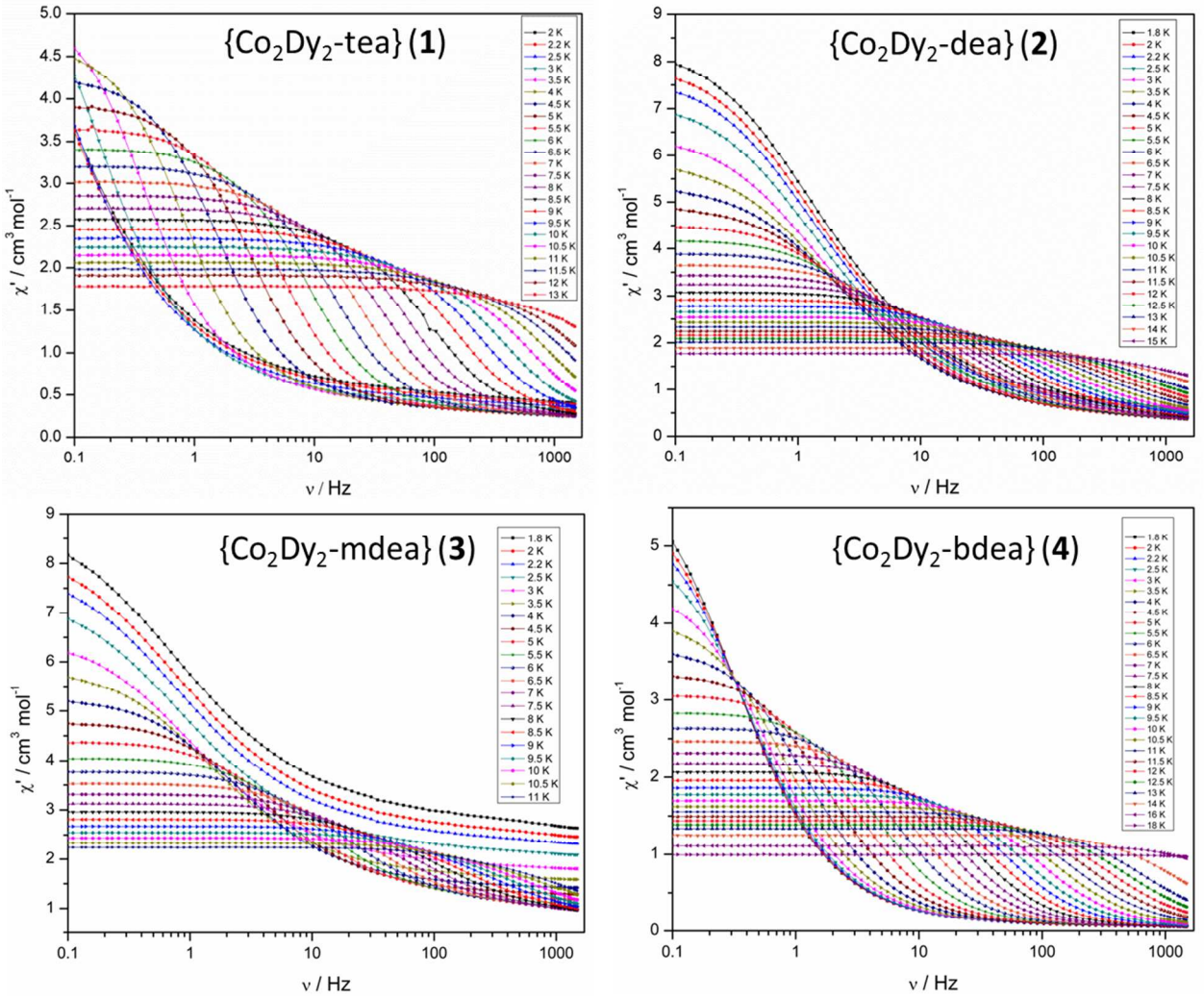

Figure S5. Ac susceptibility data for **1** (top left), **2** (top right), **3** (bottom left) and **4** (bottom right) plotted as χ_M' *vs frequency*.

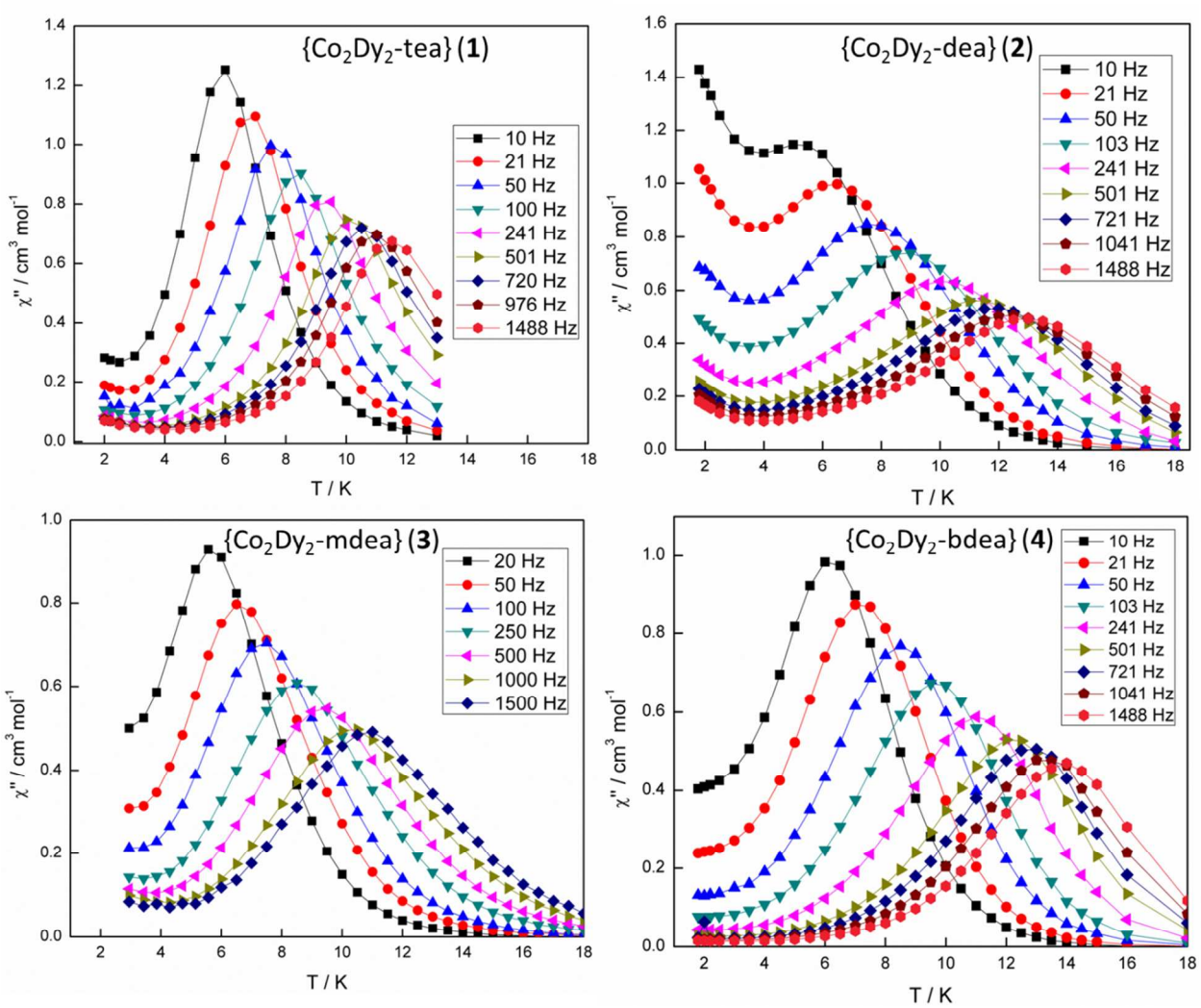

Figure S6. Temperature dependence of the out-of-phase ac susceptibilities, χ_M'', of **1** (top, left), **2** (top right), **3** (bottom left) and **4** (bottom right).

Cole-Cole plots and analysis

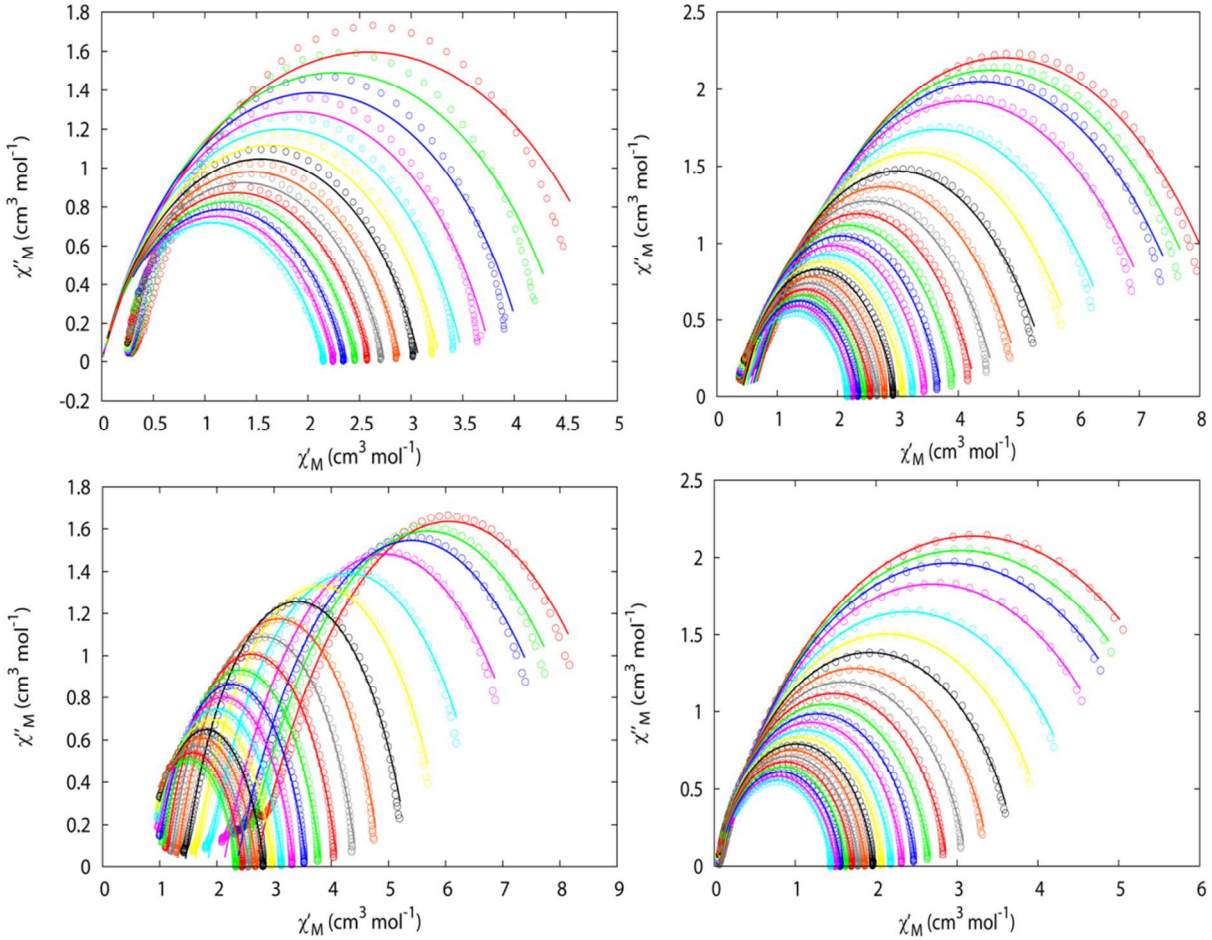

Figure S7. Cole-Cole plots for **1** (top left), **2** (top right), **3** (bottom left) and **4** (bottom right), between 1.8 – 14 K. The solid lines are best fits to the experimental data with the generalized Debye model as detailed below.

Cole-Cole plots were fitted using formulae describing χ' and χ'' in terms of frequency, constant temperature susceptibility (χ_T), adiabatic susceptibility (χ_S), relaxation time (τ), and a variable representing the distribution of relaxation times (α).

Cole-Cole plots, fitted to a generalized Debye for **1** in the temperature range 4 − 10.5 K, revealed compressed semicircular plots, indicating a single relaxation process (Figure S7, top left). It was found, however, that the α values ranged from 0.29 − 0.24, indicating a reasonably broad distribution of relaxation times in this single relaxation process.

Cole-Cole plots for **2** fitted to a generalized Debye model in the temperature range 1.8 – 12 K (Figure S7, top right), reveal rather compressed semicircles, which leads to moderately large α values ranging from 0.38 – 0.28, indicating a significant distribution of relaxation times.

Cole-Cole plots fitted to a generalized Debye model for **3** in the temperature range 1.8 – 10.5 K (Figure S7, bottom left) take the form of compressed semicircles and, at temperatures below 8 K, multiple relaxation processes are observable. Unfortunately there is not enough data to accurately model the dynamics for the second relaxation process. However, for the main thermally activated process, α values of 0.43 – 0.30 are determined, indicating a broad distribution of relaxation times.

Cole-Cole plots fitted to a generalized Debye model for **4** in the temperature range 1.8 – 14 K (Figure S7, bottom right), reveal relatively symmetrical semicircles, indicating a single relaxation process, with α values ranging from 0.26 – 0.15, indicating a narrow distribution of relaxation times for this single relaxation process.

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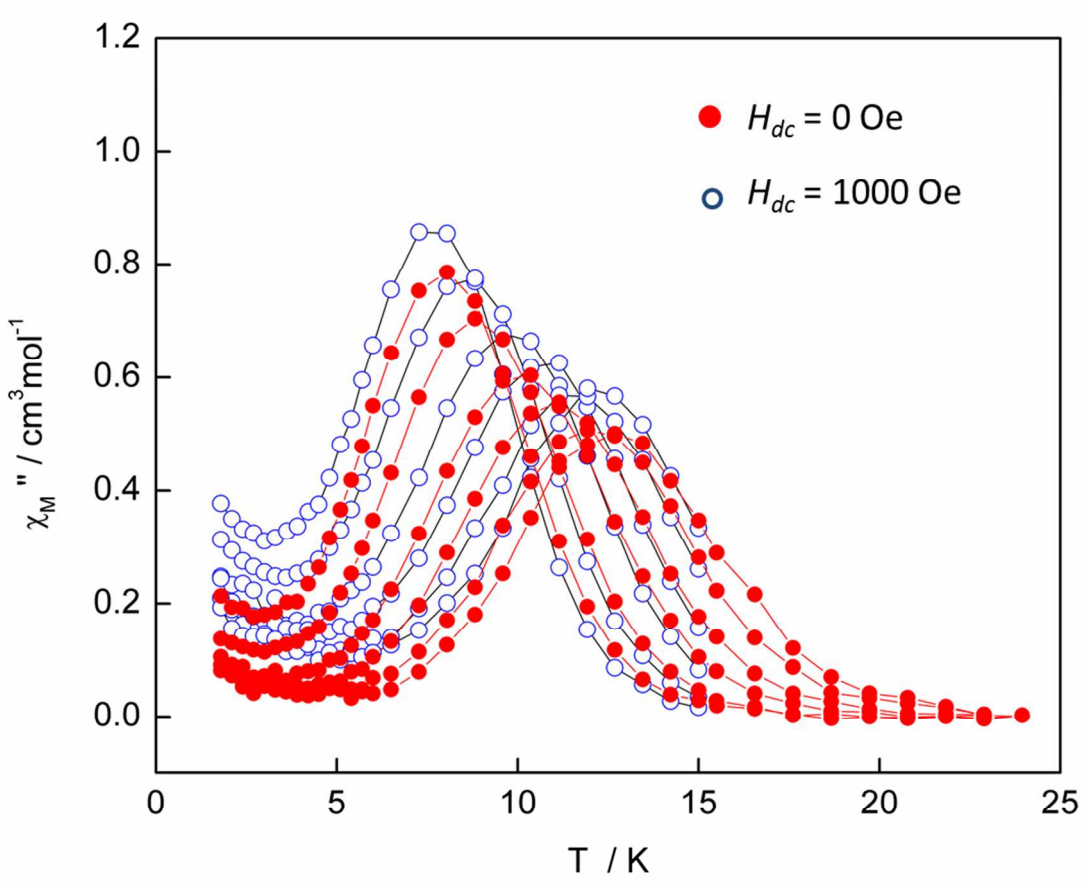

Figure S8. Temperature dependence of the out-of-phase, χ_M'', ac susceptibility, of **1**, in zero applied dc field (red circles) and H_{dc} = 1000 Oe (open circles) at several frrquencies (250 - 1500 Hz).

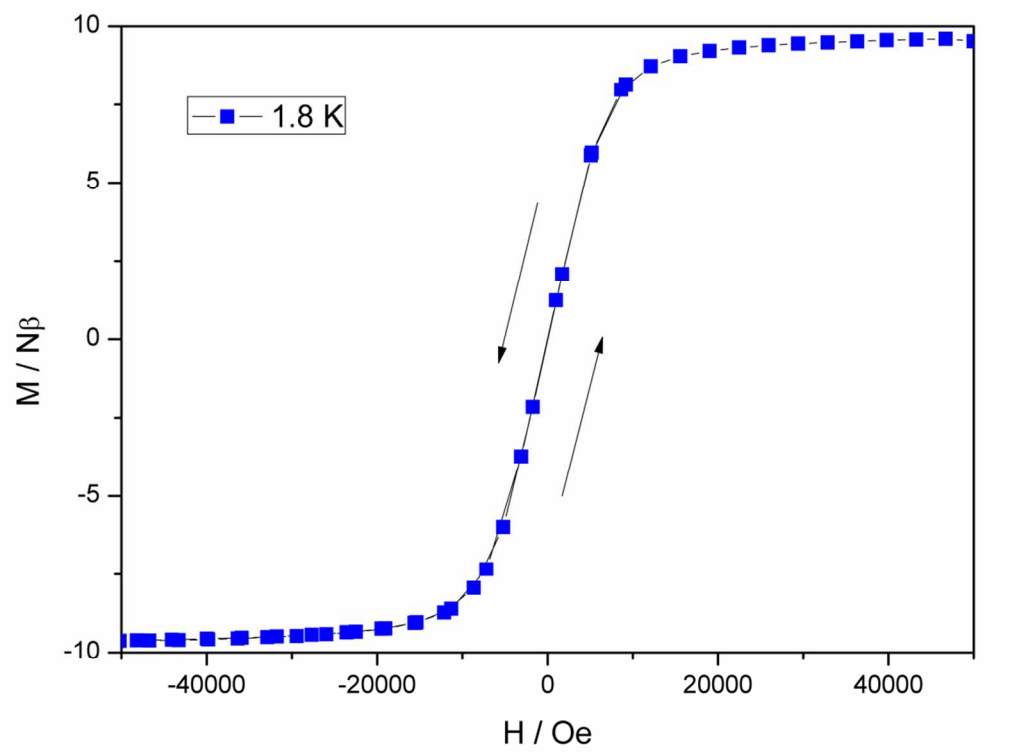

Figure S9. Plot of magnetization (*M*) *versus* field (*H*) for **1**, sweeping the field with an average sweep rate of 0.003 T/s, at the temperature indicated.

Ab-initio Calculations

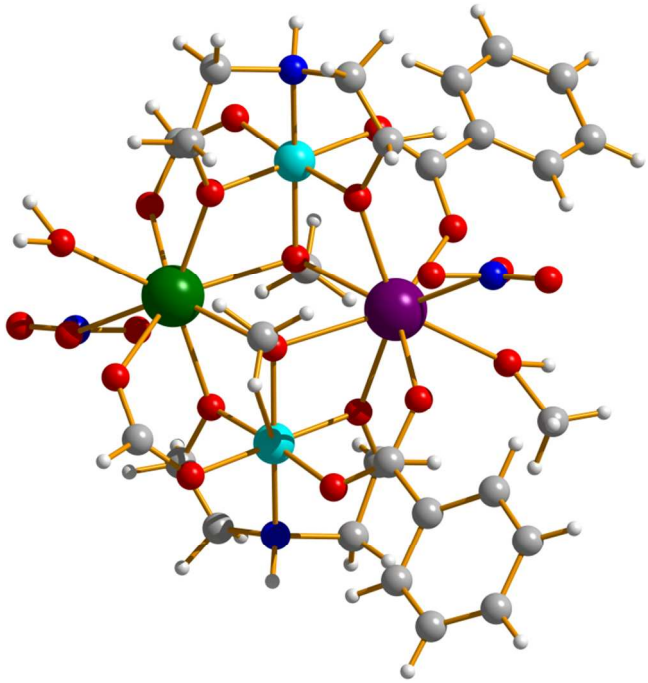

Figure S10. Structure of the fragment A of molecule 1 in compound **1**. Fragment A of molecule 2 in compound **1** is similar.

Table S1. Contractions of the employed basis sets in computational approximations A and B.

Basis α	Basis β
Dy.ANO-RCC...7s6p4d3f1g.	Dy.ANO-RCC...8s7p5d4f2g1h.
Lu.ANO-RCC...7s6p4d3f1g.	Lu.ANO-RCC...7s6p4d3f1g.
Co.ANO-RCC...5s4p2d1f.	Co.ANO-RCC...5s4p2d1f.
O.ANO-RCC...3s2p1d. (close)	O.ANO-RCC...4s3p2d. (close)
O.ANO-DK3.Tsuchiya.12s8p.2s1p. (distant)	O.ANO-RCC...3s2p. (distant)
N.ANO-RCC...3s2p1d. (close)	N.ANO-RCC...4s3p2d. (close)
N.ANO-DK3.Tsuchiya.12s8p.2s1p. (distant)	N.ANO-RCC...3s2p. (distant)
C.ANO-DK3.Tsuchiya.12s8p.2s1p.	C.ANO-RCC...3s2p.
H.ANO-DK3.Tsuchiya.6s.1s.	H.ANO-RCC...2s.

Ab-initio results

Tables S2-S9 display the results of the *ab-initio* calculated electronic and magnetic properties of the individual Dy^{III} free-ions for **1** – **4**, using the fragment and basis set indicated.

Table S2. Energies of the lowest Kramers doublets (cm^{-1}) for **1** (molecule 1).

Aα	Aβ	Bα	Bβ
0.000	0.000	0.000	0.000
91.168	102.930	90.960	102.608
190.425	214.880	189.937	214.728
261.566	298.374	260.840	297.419
325.581	370.009	324.691	367.808
377.240	406.936	376.416	403.613
490.428	504.759	489.678	502.437
690.116	734.554	689.504	734.189
3577.674	3580.834	3577.557	3580.537
3678.701	3692.227	3678.333	3691.966
3769.824	3797.231	3769.179	3795.869
3842.013	3875.907	3841.222	3874.103
3912.312	3944.481	3911.520	3942.392
3972.777	3998.649	3972.062	3996.772
4065.234	4097.132	4064.664	4096.408
6139.259	6145.304	6139.035	6144.975
6224.801	6241.787	6224.308	6240.854
6323.947	6349.649	6323.246	6348.080
6410.195	6441.608	6409.395	6439.671
6453.751	6487.522	6453.065	6486.383
6537.054	6558.708	6536.490	6557.506
8120.865	8130.777	8120.533	8130.257
8199.728	8219.003	8199.141	8217.771
8322.810	8349.597	8322.016	8347.658
8381.101	8412.579	8380.471	8411.626
8466.834	8487.547	8466.252	8486.284
9648.241	9660.208	9647.838	9659.498
9773.833	9795.312	9773.125	9793.825
9896.399	9925.048	9895.732	9923.628
9986.776	10009.397	9986.174	10008.274
…	…	…	…

Table S3. Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) (molecule 1) for **1**.

KD		Aα		Aβ		Bα		Bβ	
		E	*g*	E	*g*	E	*g*	E	*g*
1	g_X		0.0339		0.0216		0.0342		0.0217
	g_Y	0.000	0.0569	0.000	0.0331	0.000	0.0574	0.000	0.0330
	g_Z		19.6360		19.6957		19.6349		19.6972
2	g_X		0.2097		0.1542		0.2111		0.1552
	g_Y	91.168	0.2427	102.930	0.1687	90.960	0.2446	102.608	0.1698
	g_Z		16.7262		16.8427		16.7244		16.8491
3	g_X		0.7543		0.3096		0.7617		0.3119
	g_Y	190.425	1.2089	214.880	0.5050	189.937	1.2240	214.728	0.5072
	g_Z		14.5772		14.8105		14.5722		14.8172
4	g_X		2.2728		1.2894		2.2827		1.2415
	g_Y	261.566	4.1054	298.374	2.1934	260.840	4.1281	297.419	2.1411
	g_Z		10.4371		11.5965		10.4197		11.6299
5	g_X		2.8007		1.3878		2.7981		1.2800
	g_Y	325.581	4.1689	370.009	3.3887	324.691	4.1588	367.808	3.2360
	g_Z		11.7779		10.9704		11.7843		11.0556
6	g_X		1.7752		3.0735		1.7634		3.0744
	g_Y	377.240	2.2311	406.936	4.6248	376.416	2.2186	403.613	4.7985
	g_Z		14.7835		12.9040		14.7862		12.6138
7	g_X		0.3171		0.4068		0.3154		0.4010
	g_Y	490.428	0.4294	504.759	0.7050	489.678	0.4265	502.437	0.6968
	g_Z		17.4166		17.3512		17.4186		17.3389
8	g_X		0.0182		0.0149		0.0181		0.0144
	g_Y	690.116	0.0357	734.554	0.0282	689.504	0.0353	734.189	0.0272
	g_Z		19.2622		19.3644		19.2631		19.3685

Table S4. Energies of the lowest Kramers doublets (cm^{-1}) for **1** (molecule 2).

Aα	Aβ	Bα	Bβ
0.000	0.000	0.000	0.000
83.692	94.489	84.058	94.528
186.129	208.872	187.447	209.620
271.868	290.392	271.701	288.822
334.461	344.972	332.485	338.817
368.970	380.544	366.686	375.385
426.724	442.522	424.828	438.701
705.078	746.598	704.777	745.924
3572.175	3573.956	3572.125	3573.660
3675.532	3690.600	3676.764	3691.241
3775.164	3783.283	3774.426	3780.795
3842.304	3852.960	3840.888	3849.177
3897.655	3905.434	3895.372	3900.005
3949.996	3971.217	3948.980	3968.672
4060.772	4098.019	4060.668	4097.377
6131.369	6135.876	6131.717	6135.824
6224.157	6233.694	6224.191	6232.436
6319.186	6327.293	6318.031	6324.105
6401.886	6408.263	6399.832	6403.451
6465.769	6483.178	6464.536	6480.555
6507.485	6540.359	6507.780	6539.852
8113.102	8118.697	8113.288	8118.211
8196.344	8205.427	8195.850	8203.369
8313.478	8318.416	8311.813	8314.028
8393.537	8407.756	8392.163	8405.043
8440.290	8467.348	8440.447	8466.542

9640.050	9646.083	9640.015	9645.134
9771.414	9777.950	9770.636	9775.102
9895.764	9902.393	9893.799	9898.450
9969.161	9997.090	9969.311	9996.346
…	…	…	…

Table S5. Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) (molecule 2) for **1**.

KD		Aα		Aβ		Bα		Bβ	
		E	*g*	E	*g*	E	*g*	E	*g*
1	g_X		0.0317		0.0212		0.0313		0.0208
	g_Y	0.000	0.0517	0.000	0.0303	0.000	0.0504	0.000	0.0291
	g_Z		19.6438		19.7124		19.6513		19.7167
2	g_X		0.1944		0.1746		0.1964		0.1778
	g_Y	83.692	0.2078	94.489	0.1869	84.058	0.2104	94.528	0.1914
	g_Z		16.7947		16.9536		16.8202		16.9736
3	g_X		0.1676		0.1114		0.1732		0.1371
	g_Y	186.129	0.3513	208.872	0.2479	187.447	0.3506	209.620	0.2592
	g_Z		14.9559		14.9115		14.9772		14.9017
4	g_X		1.0226		0.3259		1.0072		0.3615
	g_Y	271.868	1.5846	290.392	0.7062	271.701	1.5757	288.822	0.8019
	g_Z		11.8617		12.0253		11.8659		12.0482
5	g_X		2.4883		2.4975		2.5170		2.5727
	g_Y	334.461	5.3086	344.972	5.0287	332.485	5.2378	338.817	4.8776
	g_Z		12.5342		12.7501		12.7851		13.2309
6	g_X		0.7788		1.8740		0.6689		1.9929
	g_Y	368.970	3.1502	380.544	2.9879	366.686	2.9366	375.385	2.7861
	g_Z		9.3410		8.8280		9.2623		8.8247
7	g_X		1.0899		0.9198		1.0893		0.8864
	g_Y	426.724	3.4655	442.522	2.6769	424.828	3.3943	438.701	2.4860
	g_Z		14.6910		15.1327		14.7522		15.2449
8	g_X		0.0110		0.0069		0.0106		0.0065
	g_Y	705.078	0.0186	746.598	0.0113	704.777	0.0178	745.924	0.0106
	g_Z		19.4886		19.5369		19.4933		19.5408

Table S6. Energies of the lowest Kramers doublets (cm^{-1}) for **2** (left); Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) for **2** (right).

Bα	Bβ
0.000	0.000
112.734	108.276
260.314	249.142
349.722	335.951
401.734	394.423
442.264	434.800
484.238	477.653
821.959	811.199
3580.541	3576.884
3741.509	3727.773
3818.488	3810.659
3897.077	3889.182

3956.452	3949.239
4034.789	4023.597
4174.229	4160.789
6157.951	6149.659
6279.133	6267.315
6368.046	6359.280
6448.495	6441.450
6537.710	6528.049
6623.169	6605.862
8149.719	8139.531
8253.066	8241.166
8357.588	8349.144
8452.681	8444.093
8548.052	8530.152
9681.022	9669.831
9827.002	9815.066
9937.392	9929.891
10075.349	10058.118
…	

KD		Bα		Bβ	
		E	*g*	E	*g*
1	g_X	0.000	0.00405	0.000	0.00458
	g_Y		0.00435		0.00518
	g_Z		19.84541		19.83905
2	g_X	112.734	0.09606	108.276	0.10496
	g_Y		0.10880		0.11702
	g_Z		17.18326		17.15578
3	g_X	260.314	0.20352	249.142	0.24839
	g_Y		0.24712		0.31790
	g_Z		14.63571		14.75502
4	g_X	349.722	0.71326	335.951	0.40008
	g_Y		1.37374		1.07285
	g_Z		13.20686		13.03589
5	g_X	401.734	3.30531	394.423	3.50630
	g_Y		5.97459		6.03073
	g_Z		10.97800		10.51169
6	g_X	442.264	0.09355	434.800	0.52324
	g_Y		3.34219		3.79039
	g_Z		9.30512		9.21609
7	g_X	484.238	0.92980	477.653	0.83740
	g_Y		3.70534		3.59072
	g_Z		14.25437		14.20400
8	g_X	821.959	0.00239	811.199	0.00261
	g_Y		0.00478		0.00514
	g_Z		19.67152		19.66209

Table S7. Energies of the lowest Kramers doublets (cm^{-1}) for **3** (top); Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) for **3** (bottom).

Bα	Bβ
0.000	0.000
84.119	87.511
207.023	203.390
232.210	225.657
281.603	277.465
349.241	347.518
423.013	426.171
751.031	759.910
3565.057	3565.201
3684.323	3675.854
3746.060	3742.224
3804.790	3800.416
3854.162	3851.465
3963.712	3965.107
4106.412	4111.948
6129.295	6125.608
6206.554	6199.466
6284.580	6279.958
6363.389	6359.179
6468.222	6469.031
6549.396	6550.331
8109.026	8103.984
8172.515	8165.885
8270.477	8264.741
8391.349	8391.651
8470.971	8469.010
9629.147	9623.425
9739.591	9731.613
9869.787	9867.945
10000.587	9999.757
…	…

KD		Bα		Bβ	
		E	*g*	E	*g*
1	g_X	0.000	0.00992	0.000	0.01066
	g_Y		0.01043		0.01152
	g_Z		19.85684		19.85923
2	g_X	84.119	0.12368	87.511	0.13371
	g_Y		0.13944		0.14946
	g_Z		17.12494		17.15037
3	g_X	207.023	0.91202	203.390	0.94589
	g_Y		3.90265		4.03245
	g_Z		11.93642		11.85614
4	g_X	232.210	0.16715	225.657	0.06030
	g_Y		4.80272		5.17574
	g_Z		13.93834		13.74929
5	g_X	281.603	2.49993	277.465	2.49215
	g_Y		3.66873		4.08613
	g_Z		11.74083		11.04565
6	g_X	349.241	2.97527	347.518	3.16638
	g_Y		4.30736		4.12487
	g_Z		9.30802		9.57518

7	g_X	423.013	0.83206	426.171	0.75677
	g_Y		1.82700		1.57840
	g_Z		14.56702		14.80559
8	g_X	751.031	0.00526	759.910	0.00451
	g_Y		0.00682		0.00542
	g_Z		19.61381		19.62324

Table S8. Energies of the lowest Kramers doublets (cm^{-1}) for **4** (molecule 1) (top); Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) for **4** (molecule 1) (bottomt).

Bα	Bβ
0.000	0.000
117.097	119.852
257.754	262.020
336.891	348.859
371.810	394.413
421.217	440.531
476.919	497.484
821.268	827.607
3582.718	3583.768
3737.369	3735.157
3809.996	3821.462
3881.735	3899.530
3928.564	3951.950
4025.994	4036.205
4173.692	4177.629
6158.496	6158.607
6268.573	6272.418
6355.380	6367.857
6430.013	6450.291
6526.410	6536.778
6620.729	6622.466
8146.595	8148.061
8239.130	8245.065
8342.908	8360.076
8443.117	8452.920
8542.672	8544.457
9674.251	9676.813
9812.548	9821.844
9926.753	9940.054
10069.810	10071.280
…	…

KD		Bα		Bβ	
		E	*g*	E	*g*
1	g_X		0.00512		0.00711
	g_Y	0.000	0.00598	0.000	0.00876
	g_Z		19.84436		19.83008
2	g_X		0.13521		0.12487
	g_Y	117.097	0.16387	119.852	0.14129
	g_Z		17.17445		17.13936

3	g_X g_Y g_Z	257.754	0.59343 0.85034 14.19933	262.020	0.22304 0.29007 14.56935
4	g_X g_Y g_Z	336.891	9.75791 6.71550 3.16157	348.859	1.87279 3.20122 11.80441
5	g_X g_Y g_Z	371.810	10.45889 6.77873 0.30686	394.413	11.15190 6.87506 2.22471
6	g_X g_Y g_Z	421.217	2.64043 3.03202 10.08946	440.531	1.35781 2.17162 9.99299
7	g_X g_Y g_Z	476.919	0.96034 2.29480 15.36585	497.484	0.99087 2.69706 15.18277
8	g_X g_Y g_Z	821.268	0.00278 0.00407 19.66980	827.607	0.00377 0.00586 19.63823

Table S9. Energies of the lowest Kramers doublets (cm^{-1}) for **4** (molecule 2) (top); Energies (cm^{-1}) and *g* tensors of the lowest Kramers doublets (KD) for **4** (molecule 2) (bottom).

Bα	Bβ
0.000	0.000
100.582	104.607
220.482	230.164
278.881	296.383
321.801	330.477
351.579	366.720
414.915	435.729
765.185	770.081
3573.250	3574.690
3714.752	3714.458
3763.569	3774.723
3822.194	3839.244
3864.228	3882.960
3975.490	3983.515
4124.249	4128.483
6143.530	6144.660
6233.734	6238.378
6306.370	6317.644
6366.523	6384.255
6477.373	6483.823
6574.792	6578.362
8124.140	8126.741
8199.411	8204.499
8284.000	8300.161
8397.045	8403.002
8496.919	8500.188
9645.871	9649.230
9765.760	9774.929
9877.323	9886.736
10024.417	10026.850
…	…

KD		Bα		Bβ	
		E	*g*	E	*g*
1	g_X g_Y g_Z	0.000	0.00739 0.00952 19.84821	0.000	0.00754 0.00932 19.83854
2	g_X g_Y g_Z	100.582	0.19084 0.27326 17.05789	104.607	0.13681 0.18323 17.07626
3	g_X g_Y g_Z	220.482	2.13499 4.08387 12.23361	230.164	1.47530 2.24005 13.35999
4	g_X g_Y g_Z	278.881	1.16555 5.57167 10.17543	296.383	2.98592 5.44666 10.18313
5	g_X g_Y g_Z	321.801	0.35017 5.60995 13.15722	330.477	11.10339 7.47260 0.53240
6	g_X g_Y g_Z	351.579	2.62678 5.96104 10.29390	366.720	2.44654 5.36095 10.58417
7	g_X g_Y g_Z	414.915	0.49778 1.03922 16.29325	435.729	0.52323 1.02867 16.70920
8	g_X g_Y g_Z	765.185	0.00298 0.00442 19.71806	770.081	0.00349 0.00527 19.69575

Magnetic fits

Figures S9- S16 show fits of the magnetic data, using the Hamiltonian described in the text. The ionic anisotropy on the metal sites is calculated *ab initio*. The dipolar magnetic interaction between metal sites is based only on *ab initio* data, so requires no fitting parameters. Fitting parameters are used to simulate the anisotropic exchange interaction. The latter is only a fraction of the total magnetic interaction between metal sites, therefore, will not change qualitatively the results even if would be described by different parameters than the ones which we used in the text.

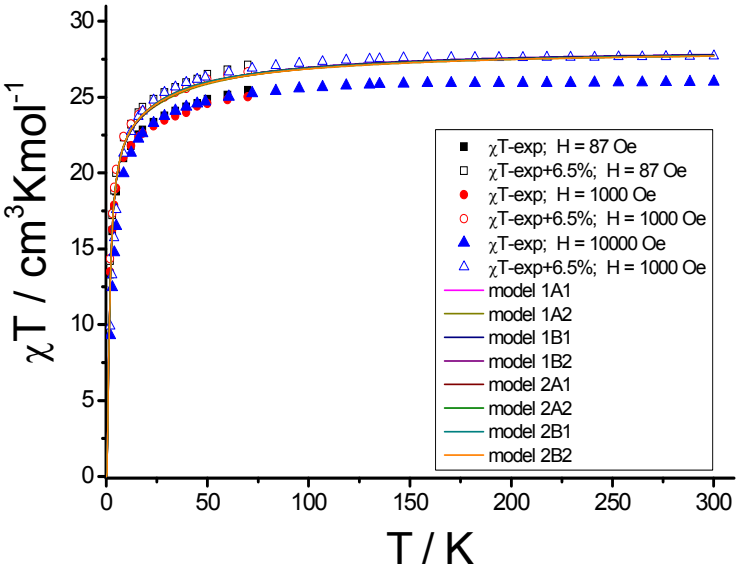

Figure S11. A comparison between measured and calculated magnetic susceptibility of **1**. An up-scaling of the experimental curve with 6.5% leads to an almost perfect agreement.

Rescaling is performed in all cases because of several unavoidable errors (paramagnetic impurities, mass errors, solvent, water entry/exit with variation of temperature etc). We do not expect large ab initio errors for the description of the room temperature $\chi_M T$ for individual metal sites (there is sufficient evidence for that). On the other hand at 300K the $\chi_M T$ is just a sum from individual metal centers, so our rescaling is done in the way to reconcile experiment and theory at 300K.

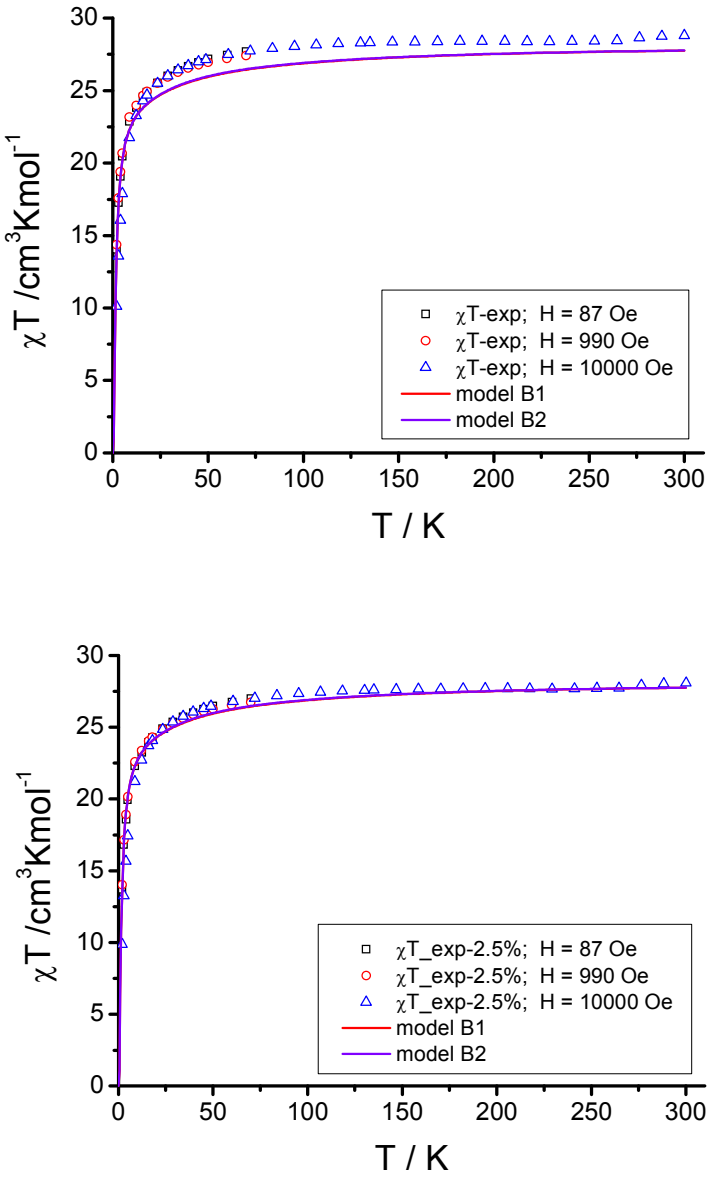

Figure S12. A comparison between measured and calculated magnetic susceptibility of **2** (top). A comparison between downscaled experimental data by 2.5% and calculated magnetic susceptibility of **2**. Downscaling was done only to estimate the discrepancy.

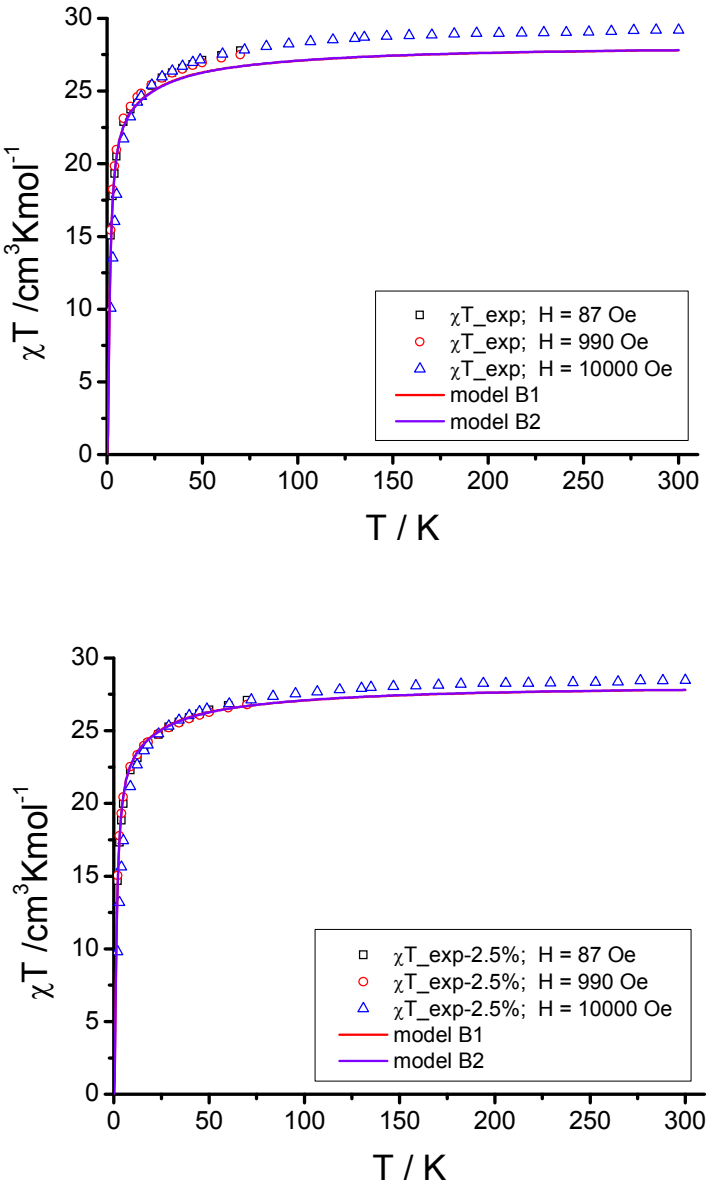

Figure S13. A comparison between measured and calculated magnetic susceptibility of **3** (top). A comparison between downscaled experimental data by 2.5% and calculated magnetic susceptibility of **3**. Downscaling was done only to estimate the discrepancy.

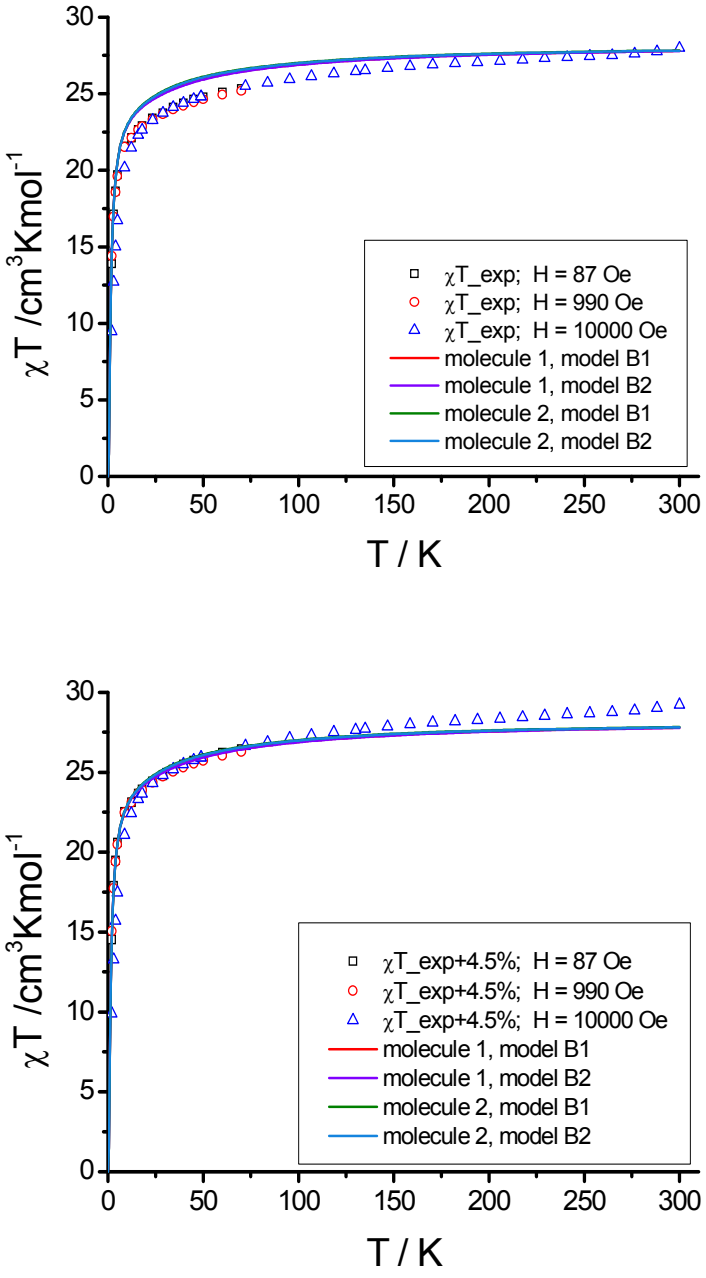

Figure S14. A comparison between measured and calculated magnetic susceptibility of **4.**(top). A comparison between downscaled experimental data by 2.5% and calculated magnetic susceptibility of **4**. Downscaling was done only to estimate the discrepancy.

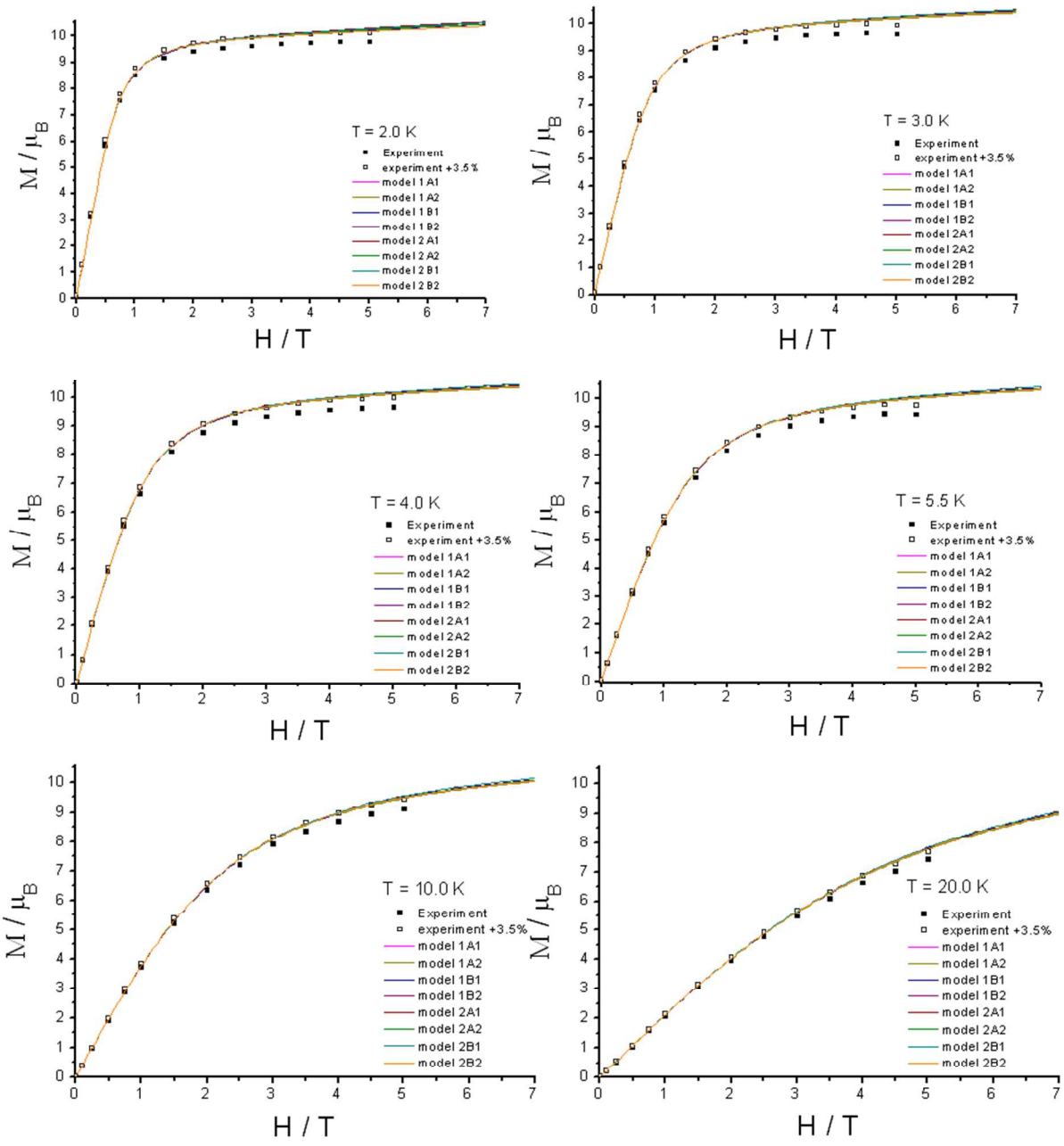

Figure S15. Measured and calculated molar magnetization of **1** between 2.0 – 20 K. The perfect agreement is achieved if one up-scales the experiment with about 3.5%.

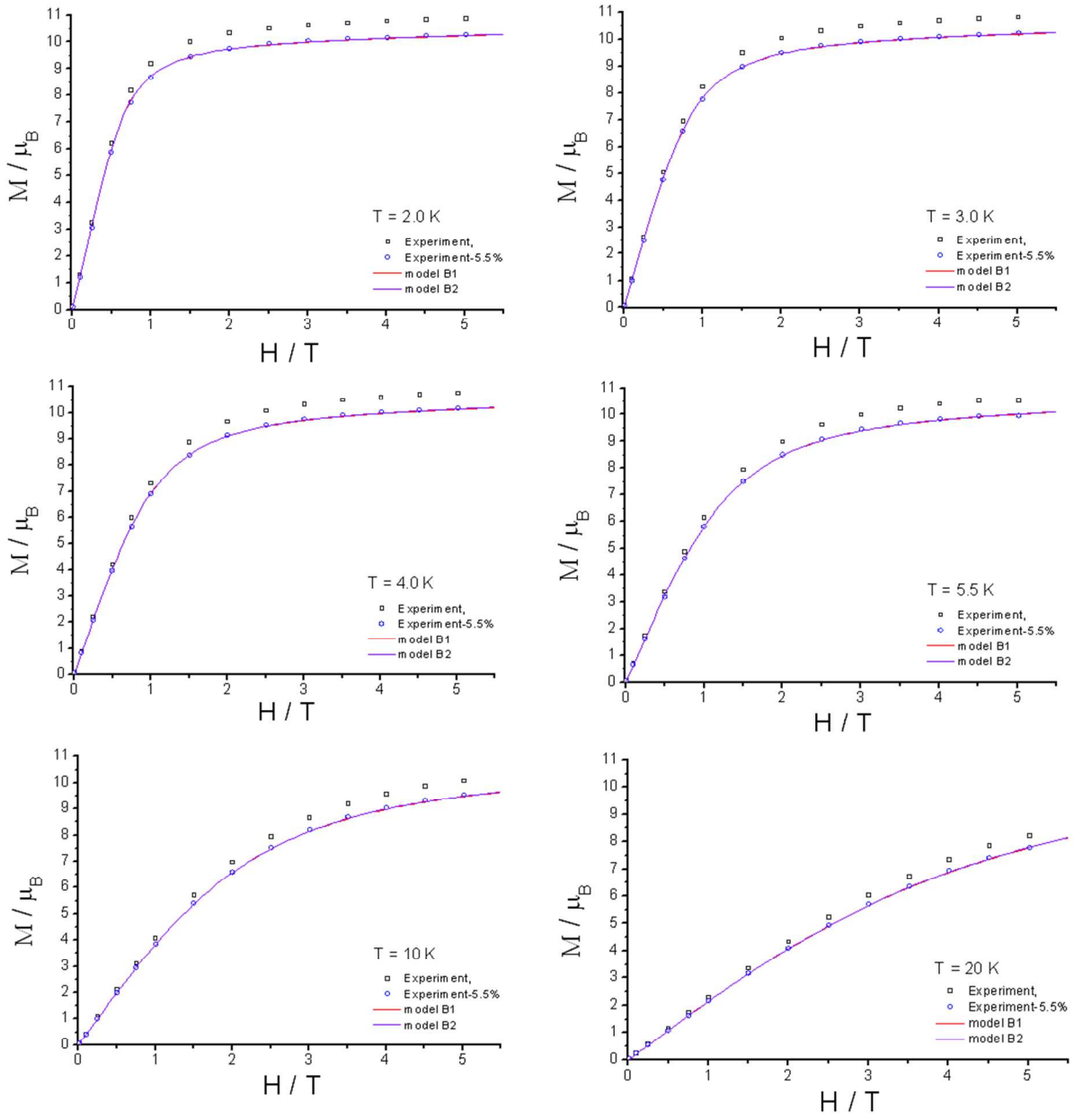

Figure S16. Measured and calculated molar magnetization of **2** between 2.0 – 20 K. The perfect agreement is achieved if one up-scales the experiment with about 5.5%.

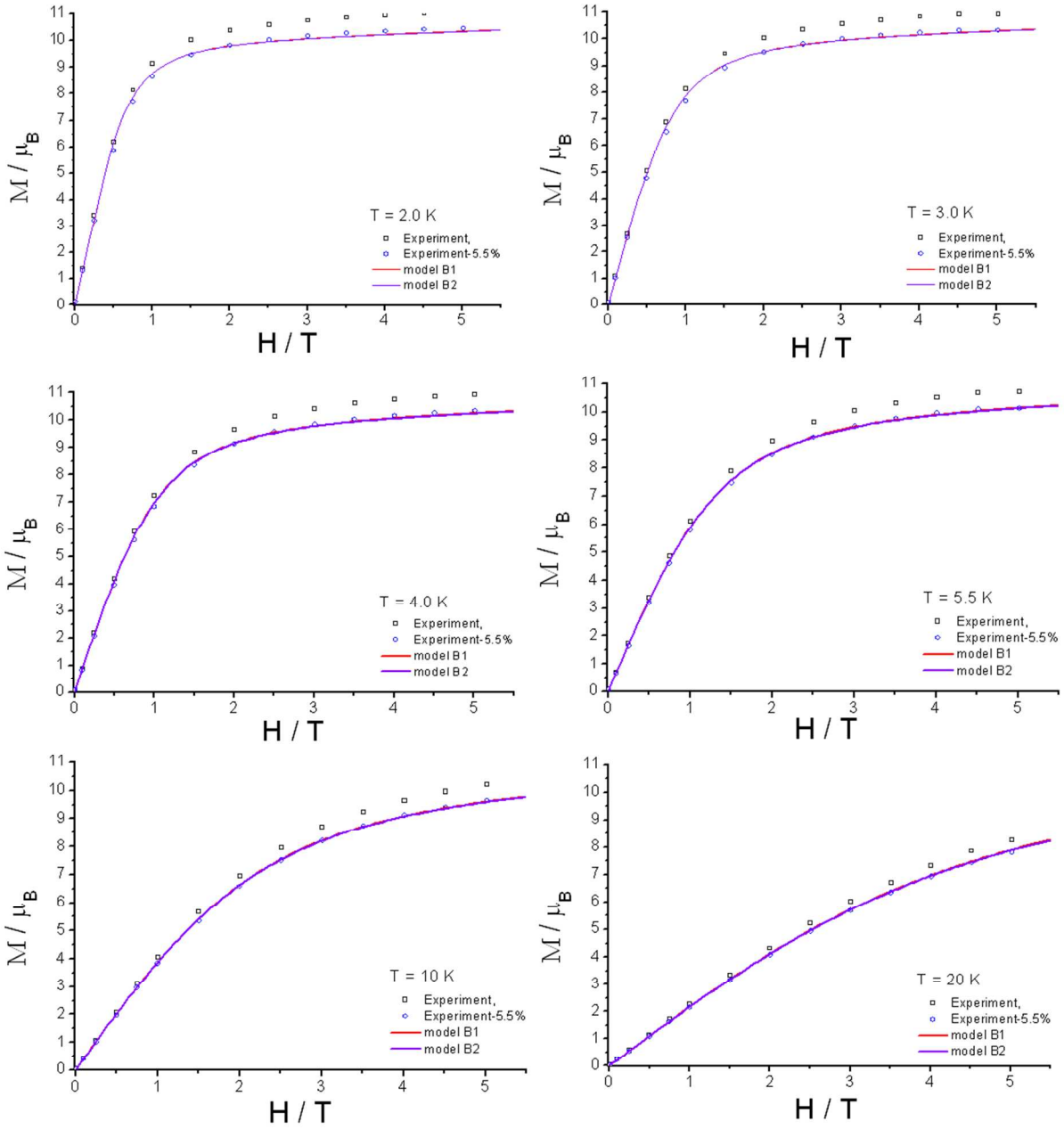

Figure S17. Measured and calculated molar magnetization of **3** between 2.0 – 20 K. The perfect agreement is achieved if one up-scales the experiment with about 5.5%.

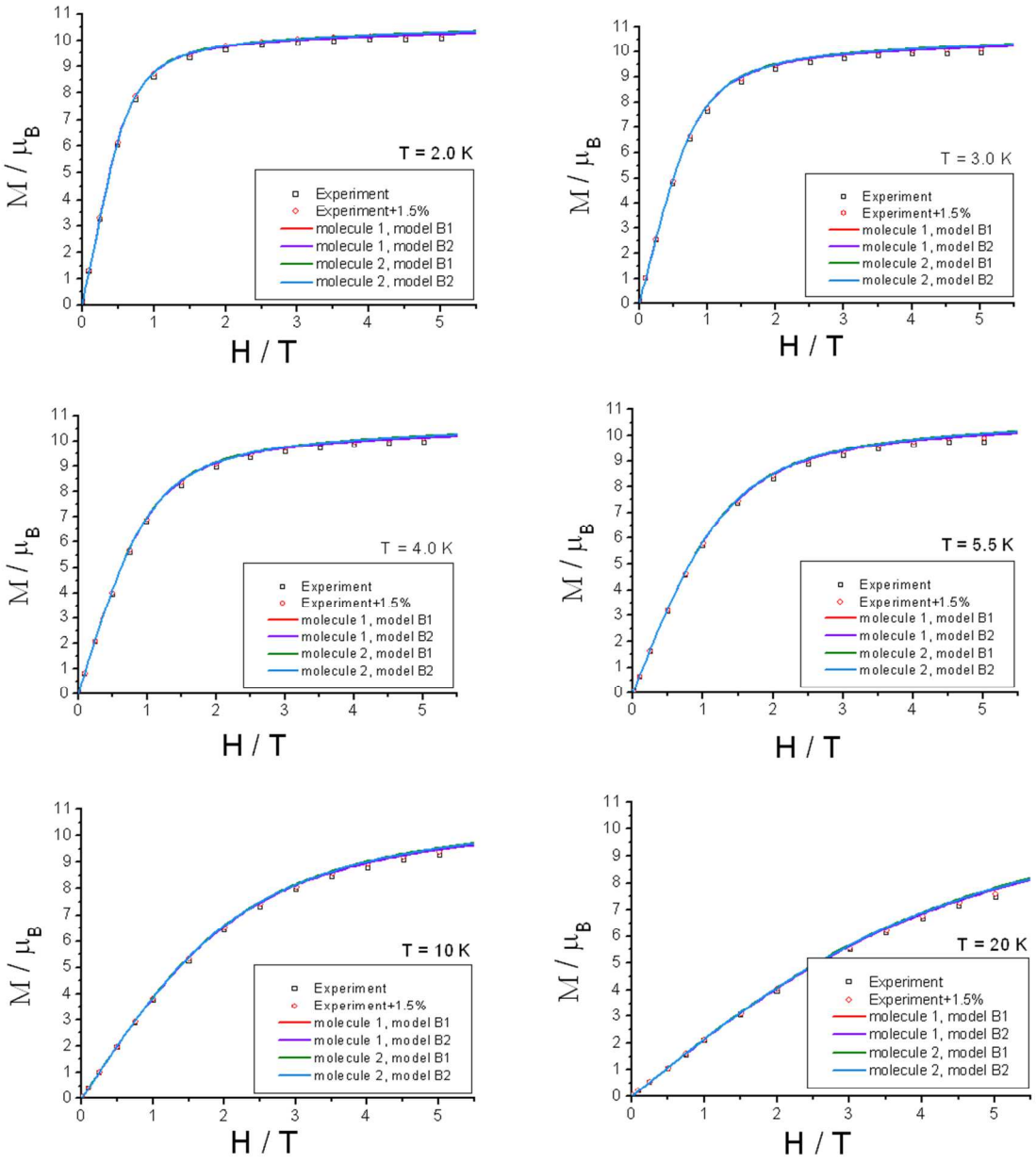

Figure S18. Measured and calculated molar magnetization of **4** between 2.0 – 20 K. The perfect agreement is achieved if one up-scales the experiment with about 1.5%.

Table S10. Energies (cm^{-1}) and the corresponding tunneling gaps and g_Z values of the lowest 4 exchange doublet states of the complex **1**.

molecule 1 model Aα		
Energy	Δ_{tun}	g_Z
0.000000000000000	4.797464E-06	0.00000
0.000004797464236		
1.440863445448618	1.952817E-05	39.27094
1.440882973616502		
91.142406299825709	1.014505E-04	0.00000
91.142507750373028		
91.395980257161710	9.607161E-05	0.00000
91.396076328769041		

molecule 1 model Aβ		
energy	Δ_{tun}	g_Z
0.000000000000000	1.297680E-06	0.00000
0.000001297679559		
1.452122477654919	7.007646E-06	39.39075
1.452129485300851		
102.907162794638609	3.661065E-05	0.00000
102.907199405284345		
103.157597701783473	3.447852E-05	0.00000
103.157632180305058		

molecule 1 model Bα		
Energy	Δ_{tun}	g_Z
0.000000000000000	4.881695E-06	0.00000
0.000004881694807		
1.440674942959495	1.986503E-05	0.00000
1.440694807984924		
90.934481510808581	1.032349E-04	39.26891
90.934584745710126		
91.188038769683743	9.764593E-05	0.00000
91.188136415612789		

molecule 1 model Bβ		
energy	Δ_{tun}	g_Z
0.000000000000000	1.263523E-06	0.00000
0.000001263522885		
1.452927994830413	6.990304E-06	0.00000
1.452934985134879		
102.584064459846260	3.651383E-05	39.39381
102.584100973676684		
102.834264759072198	3.412666E-05	0.00000
102.834298885735706		

molecule 2 model Aα		
Energy	Δ_{tun}	g_Z
0.000000000000000	4.258016E-06	0.00000
0.000004258015721		
1.450797897066219	1.653994E-05	39.28653
1.450814437006260		
83.669694263781381	8.555609E-05	0.00000
83.669779819867088		
83.922944556585094	8.284923E-05	0.00000
83.923027405810103		

molecule 2 model Aβ		
energy	Δ_{tun}	g_Z
0.000000000000000	8.872979E-07	0.00000
0.000000887297932		
1.463957470902943	6.320130E-06	39.42413
1.463963791032483		
94.467098868387566	3.517934E-05	0.00000
94.467134047728408		
94.713363808748824	2.856343E-05	0.00000
94.713392372178646		

molecule 2 model Bα		
Energy	Δ_{tun}	g_Z
0.000000000000000	3.908623E-06	0.00000
0.000003908622788		
1.452924685648653	1.587040E-05	39.30167
1.452940556050323		
84.036379432969824	8.332615E-05	0.00000
84.036462759123864		
84.288235365412461	7.943411E-05	0.00000
84.288314799523249		

molecule 2 model Bβ		
energy	Δ_{tun}	g_Z
0.000000000000000	7.396901E-07	0.00000
0.000000739690065		
1.465044597499337	5.940796E-06	39.43291
1.465050538295236		
94.507111344767665	3.390922E-05	0.00000
94.507145253990217		
94.751888650019410	2.641006E-05	0.00000
94.751915060076584		

Table S11. Energies (cm^{-1}) and the corresponding tunneling gaps and g_Z values of the lowest 4 exchange doublet states of the complex **2**.

model Bα		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000004761867	4.76187E-08	0.00000
1.32464411317311 1.32464428930562	1.76133E-07	39.69075
112.69987913120548 112.69988010971026	9.78505E-07	0.00000
112.95283230927218 112.95283242625953	1.16987E-07	0.00000

model Bβ		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000004826545	4.82655E-08	0.00000
1.32562801070123 1.32562824356435	2.32863E-07	39.67799
108.24177933031484 108.24178117514261	1.84483E-06	0.00000
108.49398094803567 108.49398120927681	2.61241E-07	0.00000

Table S12. Energies (cm^{-1}) and the corresponding tunneling gaps and g_Z values of the lowest 4 exchange doublet states of the complex **3**.

model Bα		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000039920273	3.99203E-07	0.00000
1.25706154356534 1.25706262771785	1.08415E-06	39.71362
84.07216052413625 84.07216279818819	2.27405E-06	0.00000
84.33867671438499 84.33868544170637	8.72732E-06	0.00000

model Bβ		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000055434312	5.54343E-07	0.00000
1.25454157899791 1.25454289730359	1.31831E-06	39.71840
87.46561575132330 87.46561974064839	3.98932E-06	0.00000
87.73089318312503 87.73090286118394	9.67806E-06	0.00000

Table S13. Energies (cm^{-1}) and the corresponding tunneling gaps and g_Z values of the lowest 4 exchange doublet states of the complex **4**.

molecule 1 model Bα		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000005723484	5.72348E-08	0.00000
1.21385918137810 1.21385946191920	2.80541E-07	39.68862
117.06824442979638 117.06824747000501	3.04021E-06	0.00000
117.29960009486585 117.29960100248610	9.07621E-07	0.00000

molecule 1 model Bβ		
energy	Δ_{tun}	g_Z
0.00000000000000 0.00000008723848	8.72385E-08	0.00000
1.20699361413294 1.20699417871135	5.64578E-07	39.66002
119.82093146348839 119.82093652383389	5.06035E-06	0.00000
120.05293841697198 120.05293862728553	2.10314E-07	0.00000

molecule 2 model Bα		
Energy	Δ_{tun}	g_Z
0.00000000000000 0.00000026588193	2.65882E-07	0.00000
1.22716948864658 1.22717019204271	7.03396E-07	39.69633
100.55061976702430 100.55062101568763	1.24866E-06	0.00000
100.79093888327334 100.79095049562031	1.16123E-05	0.00000

molecule 2 model Bβ		
energy	Δ_{tun}	g_Z
0.00000000000000 0.00000024564535	2.45645E-07	0.00000
1.22067205819697 1.22067274949856	6.91302E-07	39.67697
104.57360194911446 104.57360492771639	2.97860E-06	0.00000
104.81331516846775 104.81331979061581	4.62215E-06	0.00000