Electronic Supporting Information

Quenching Quantum Tunnelling of the Magnetization Utilizing 4d-4f Exchange Interactions in Butterfly-shaped {$Ru^{III}_2Ln^{III}_2$} (Ln = Gd, Tb, Dy, Ho, and Er) Single-Molecule Magnets

Imon J. Dutta,[a] Vignesh Kasi,[b] Deepanshu Chauhan,[b] Keith S. Murray,[c] Stuart K. Langley,[d] Maheswaran Shanmugam,[b,*] Kuduva R. Vignesh[a,*]

a) Department of Chemical Sciences, Indian Institute of Science Education and Research Mohali, Sector-81, Knowledge City, S.A.S. Nagar, Mohali-140306, Punjab, India.
b) Department of Chemistry, Indian Institute of Technology Bombay, Powai, Mumbai-400076, Maharashtra, India.
c) School of Chemistry, Monash University, Clayton, VIC, Australia 3800.
d) Department of Natural Sciences, Chemistry, Manchester Metropolitan University, Manchester, UK.

Table of Contents

Table S1. Crystallographic data for complexes…….3
Table S2. The *CShM* value calculation by *SHAPE* analysis…….4
Table S3. Selected bond and angle parameters……5-6
Figure S1. X-ray powder diffraction data…….7
Figure S2. The M vs. H curves at different temperatures…….8
Figure S3. Magnetization (M) vs field hysteresis loops……9
Figure S4. χ''_M vs. frequency plots for **4-Ho** at H_{dc} = 1600 Oe…….10
Figure S5. χ'_M vs. frequency plots…….11
Figure S6. χ''_M vs. χ'_M plots……...12
Figure S7. χ''_M vs. frequency, χ'_M vs. frequency, and χ''_M vs. χ'_M plots for **4-Ho** at H_{dc} = 1600 Oe……..13
Table S4. Cole-Cole fit values of **1-Gd**…….14
Table S5. Cole-Cole fit values of **2-Tb**…….14
Table S6. Cole-Cole fit values of **3-Dy**…….15
Table S7. Cole-Cole fit values of **4-Ho** at 0 Oe…….15
Table S8. Cole-Cole fit values of **4-Ho** at 1600 Oe…….15
Table S9. Cole-Cole fit values of **5-Er**…….16
Figure S8. Magnetization relaxation time (τ), plotted as ln(τ) versus T^{-1} for complex **1-Gd**, **2-Tb**, and **5-Er**……16
Figure S9. Magnetization relaxation time (τ), plotted as ln(τ) versus T^{-1} for complex **4-Ho** at 1600 Oe…….17
Figure S10. Magnetic cores with possible exchange pathways in all the complexes……17
Table S10. Energies of the lowest Kramers doublets (KDs)/ Ising doublets (IDs) (cm^{-1}) of Ln^{III} centers and the *g*-tensor of the ground Kramers doublet/ Ising doublets……18
Table S11. *g*-Tensors of the Tb^{III}, Dy^{III}, Ho^{III}, and Er^{III} Fragments of complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**…….18
Table S12. Energies of the lowest Kramers doublets of Ru^{III} centers and the *g*-tensor of the ground Kramers doublets of complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**…….19
Table S13. RASSI energies of the lowest spin-orbit states on each Ln^{III} center in complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**……19-23
Figure S11. The *ab initio* computed magnetization blocking barrier for Tb2; Dy2, Ho2; and Er2 ions……..24
Table S14. SINGLE_ANISO computed crystal field parameters for complex **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**……25
Table S15. Different spin configurations are employed for extracting *J* values of complex **1-Gd**…….26
Figure S12. DFT-computed spin-density plots of complex **1-Gd**……27
Figure S13. (a) Eigenvalue plots for computed *J* values of **1-Gd**. (b) DFT-computed spin-density plots of complex **1-Gd**…….27
Figure S14. Eigenvalue plots for experimental *J* values and of **1-Gd**…….28
Figure S15. Low-lying exchange spectra in **5-Er**…….28
Table S16-S20. Lowest exchange doublets corresponding to tunnel splitting, and the g_z value of each doublet for complex **1-Gd**, **2-Tb**, **3-Dy**, **4-Ho,** and **5-Er**……29-31
References……31

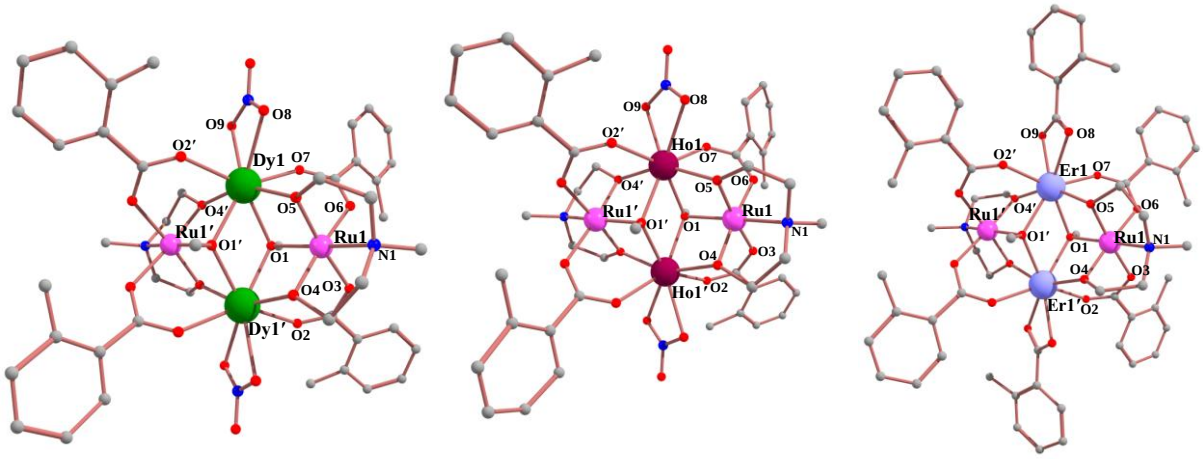
Table S1. Crystallographic data for complexes **3-Dy**, **4-Ho**, and **5-Er**.

Identification code	**3-Dy**	**4-Ho**	**5-Er**
Empirical formula	$Ru_2Dy_2C_{44}H_{56}N_4O_{20}$	$Ru_2Ho_2C_{44}H_{56}N_4O_{20}$	$Ru_2Er_2C_{61}H_{74}N_2O_{19}$
Formula weight	1488.06	1492.92	1675.88
Temperature/K	100(2)	103(2)	100(2)
Crystal system	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a/Å	10.3957(12)	10.4168(4)	8.9130(2)
b/Å	10.7243(13)	10.7457(8)	15.9402(5)
c/Å	11.0456(15)	11.0695(5)	21.6355(7)
α/°	89.649(10)	89.667(4)	93.117(3)
β/°	89.588(10)	89.915(3)	93.548(2)
γ/°	86.740(10)	85.949(4)	95.630(2)
Volume/Å^3	1229.4(3)	1235.96(12)	3047.51(15)
Z	1	1	2
ρ_{calc}g/cm^3	2.010	2.006	1.826
μ/mm^{-1}	3.689	3.847	3.287
F(000)	728	730	1660
Radiation	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)
2Θ range for data collection/°	1.844 to 25.000	1.840 to 24.997	2.220 to 24.999
Index ranges	-11 ≤ h ≤ 12, -12 ≤ k ≤ 12, -13≤ l ≤ 13	-12 ≤ h ≤ 12, -12 ≤ k ≤ 12, -13≤ l ≤ 13	-10 ≤ h ≤ 10, -18 ≤ k ≤ 18, -25 ≤ l ≤ 25
Reflections collected	21107	21342	130286
Independent reflections	21107 [R_{int}=0.1928, R_{sigma}=0.1979]	21342 [R_{int}=0.0418, R_{sigma}=0.0621]	130286 [R_{int} = 0.0615, R_{sigma} = 0.1360]
Data/restraints/parameters	4328/18/251	4351/40/316	10735/ 20/784
Goodness-of-fit on F^2	1.059	1.055	1.022
Final R indexes [I>=2σ (I)]	R_1 = 0.1027, wR_2 = 0.2507	R_1 = 0.0422, wR_2 = 0.1083	R_1 = 0.0456, wR_2 = 0.0982
Final R indexes [all data]	R_1 = 0.2039, wR_2 = 0.3227	R_1 = 0.0537, wR_2 = 0.1176	R_1 = 0.0750, wR_2 = 0.1122
Largest diff. peak/hole / e Å^{-3}	2.566, -1.553	2.009, -1.161	1.463, -1.062

Table S2. The *CShM* values were calculated by *SHAPE* analysis for complex **3-Dy**, **4-Ho**, and **5-Er**.

Complex	Metal ions	Coordination Geometry	*CShM* values
3-Dy	**Dy**	Octagon (D_{8h})	24.556
		Heptagonal pyramid (C_{7v})	23.543
		Hexagonal bipyramid (D_{6h})	13.625
		Cube (O_h)	10.628
		Square antiprism (D_{4d})	**1.747**
		Triangular dodecahedron (D_{2d})	2.348
		Johnson gyrobifastigium J26 (D_{2d})	11.888
		Johnson elongated triangular bipyramid J14 (D_{3h})	27.226
		Biaugmented trigonal prism J50 (C_{2v})	2.500
		Biaugmented trigonal prism (C_{2v})	1.997
	Ru	Hexagon (D_{6h})	32.398
		Pentagonal pyramid (C_{5v})	25.977
		Octahedron (*Oh*)	**0.411**
		Trigonal prism (D_{3h})	13.889
		Johnson pentagonal pyramid J2 (C_{5v})	30.210
4-Ho	**Ho**	Octagon (D_{8h})	25.200
		Heptagonal pyramid (C_{7v})	23.431
		Hexagonal bipyramid (D_{6h})	13.606
		Cube (O_h)	10.620
		Square antiprism (D_{4d})	**1.719**
		Triangular dodecahedron (D_{2d})	2.174
		Johnson gyrobifastigium J26 (D_{2d})	12.030
		Johnson elongated triangular bipyramid J14 (D_{3h})	27.035
		Biaugmented trigonal prism J50 (C_{2v})	2.431
		Biaugmented trigonal prism (C_{2v})	2.048
	Ru	Hexagon (D_{6h})	32.370
		Pentagonal pyramid (C_{5v})	26.131
		Octahedron (*Oh*)	**0.400**
		Trigonal prism (D_{3h})	14.118
		Johnson pentagonal pyramid J2 (C_{5v})	30.369
5-Er	**Er**	Octagon (D_{8h})	25.289
		Heptagonal pyramid (C_{7v})	23.190
		Hexagonal bipyramid (D_{6h})	13.382
		Cube (O_h)	10.539
		Square antiprism (D_{4d})	**1.671**
		Triangular dodecahedron (D_{2d})	2.146
		Johnson gyrobifastigium J26 (D_{2d})	11.740
		Johnson elongated triangular bipyramid J14 (D_{3h})	27.257
		Biaugmented trigonal prism J50 (C_{2v})	2.431
		Biaugmented trigonal prism (C_{2v})	1.985
	Ru	Hexagon (D_{6h})	32.726
		Pentagonal pyramid (C_{5v})	26.330
		Octahedron (*Oh*)	**0.399**
		Trigonal prism (D_{3h})	14.104
		Johnson pentagonal pyramid J2 (C_{5v})	30.557

Table S3. Selected bond and angle parameters for **3-Dy**, **4-Ho**, and **5-Er** using the labelling scheme given below.



Complex **3-Dy**			
Atoms	Bond length (Å)	Atoms	Bond angle (deg)
Dy1-O5	2.281	Ru1-O1-Dy1	92.76
Dy1-O4’	2.268	Ru1-O1-Dy1’	92.32
Dy1-O2’	2.325	Dy1-O1-Dy1’	113.60
Dy1-O7	2.341	Ru1-O5-Dy1	99.85
Dy1-O1	2.371	Ru1-O4-Dy1’	98.41
Dy1-O1’	2.375		
Dy1-O9	2.449		
Dy1-O8	2.488		
Ru1-O5	1.956		
Ru1-O4	2.005		
Ru1-O3	2.033		
Ru1-O6	2.052		
Ru1-N1	2.054		
Ru1-O1	2.110		
Dy1-Ru1	3.249		
Dy1-Ru1’	3.240		
Dy1-Dy1’	3.971		
Ru1-Ru1’	5.132		

Complex **4-Ho**			
Atoms	Bond length (Å)	Atoms	Bond angle (deg)
Ho1-O5	2.261	Ru1-O1- Ho1	91.87
Ho1-O4’	2.263	Ru1-O1- Ho1’	91.69
Ho1-O2’	2.318	Ho1-O1- Ho1’	112.71
Ho1-O7	2.326	Ru1-O5- Ho1	99.28
Ho1-O1	2.380	Ru1-O4- Ho1’	98.55
Ho1-O1’	2.365		
Ho1-O9	2.445		
Ho1-O8	2.447		
Ru1-O5	1.985		

Ru1-O4	1.985		
Ru1-O3	2.050		
Ru1-O6	2.053		
Ru1-N1	2.078		
Ru1-O1	2.123		
Ho1-Ru1	3.240		
Ho1-Ru1’	3.224		
Ho1- Ho1’	3.950		
Ru1-Ru1’	5.117		

Complex **5-Er**			
Atoms	Bond length (Å)	Atoms	Bond angle (deg)
Er1-O5	2.275	Ru1-O1- Er1	92.84
Er1-O4’	2.264	Ru1-O1- Er1’	92.37
Er1-O2’	2.310	Er1-O1- Er1’	113.85
Er1-O7	2.320	Ru1-O5- Er1	100.79
Er1-O1	2.408	Ru1-O4- Er1’	100.28
Er1-O1’	2.394		
Er1-O9	2.388		
Er1-O8	2.412		
Ru1-O5	1.984		
Ru1-O4	1.981		
Ru1-O3	2.074		
Ru1-O6	2.051		
Ru1-N1	2.089		
Ru1-O1	2.122		
Er1-Ru1	3.287		
Er1-Ru1’	3.264		
Er1- Er1’	4.023		
Ru1-Ru1’	5.170		

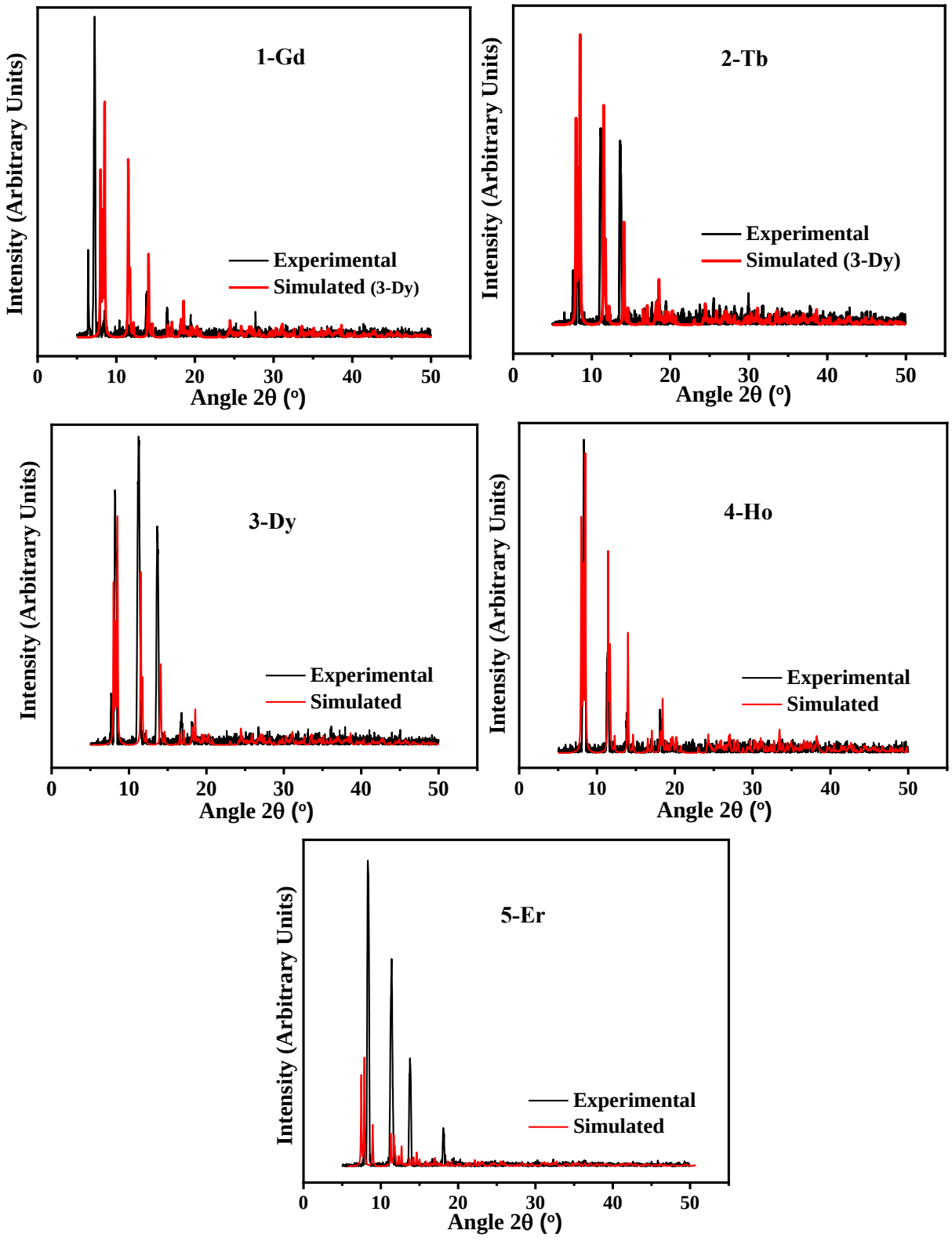


Figure S1. X-ray powder diffraction data for complexes (a) **1-Gd**, (b) **2-Tb**, (c) **3-Dy**, (d) **4-Ho**, and (e) **5-Er**. The data are compared to the simulated single-crystal X-ray diffractogram.

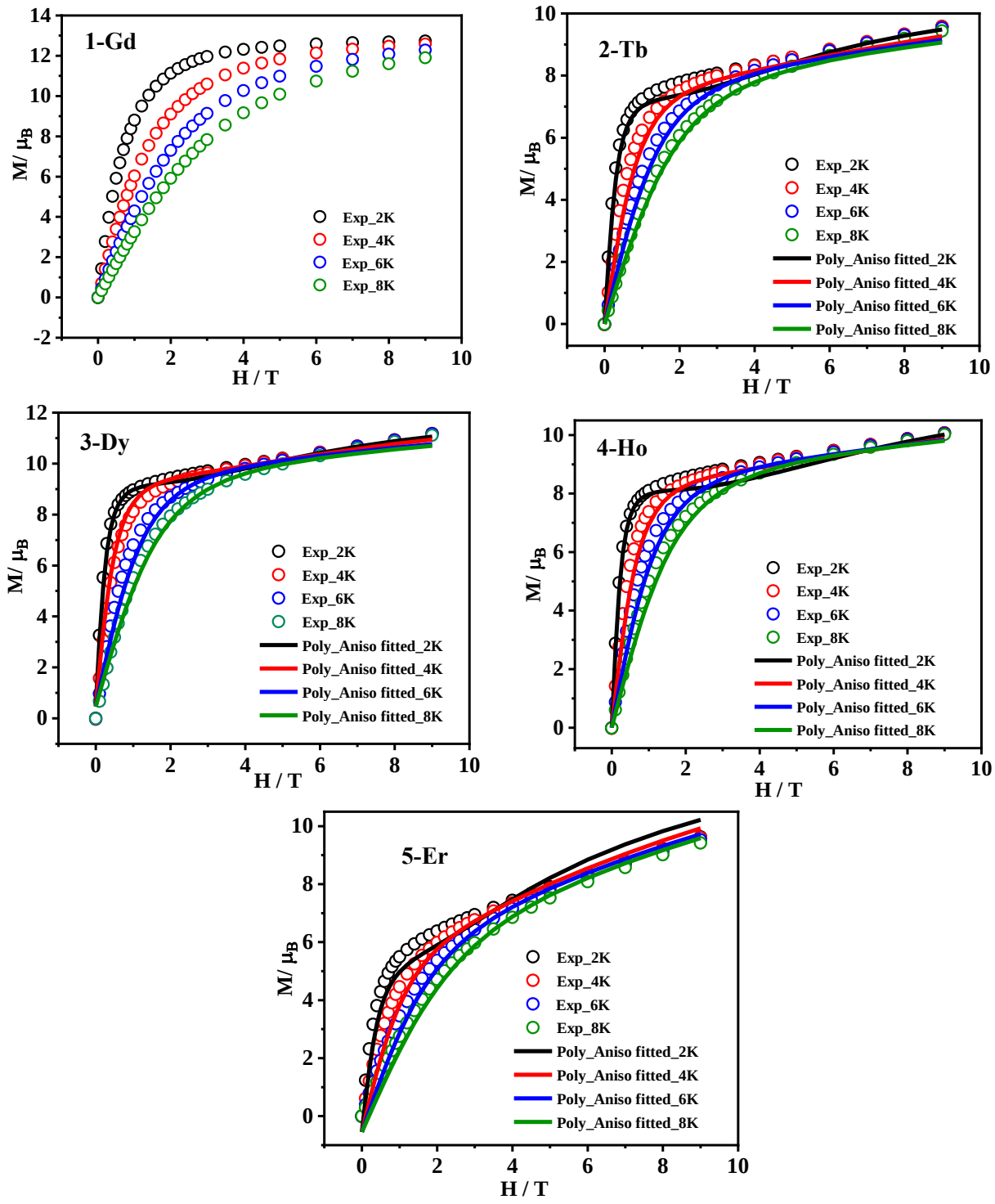


Figure S2. The M vs. H curves at different temperatures for complexes **1-Gd**, **2-Tb**, **3-Dy**, **4-Ho**, and **5-Er**. The fitted lines are from POLY_ANISO (see the computational part in the manuscript).

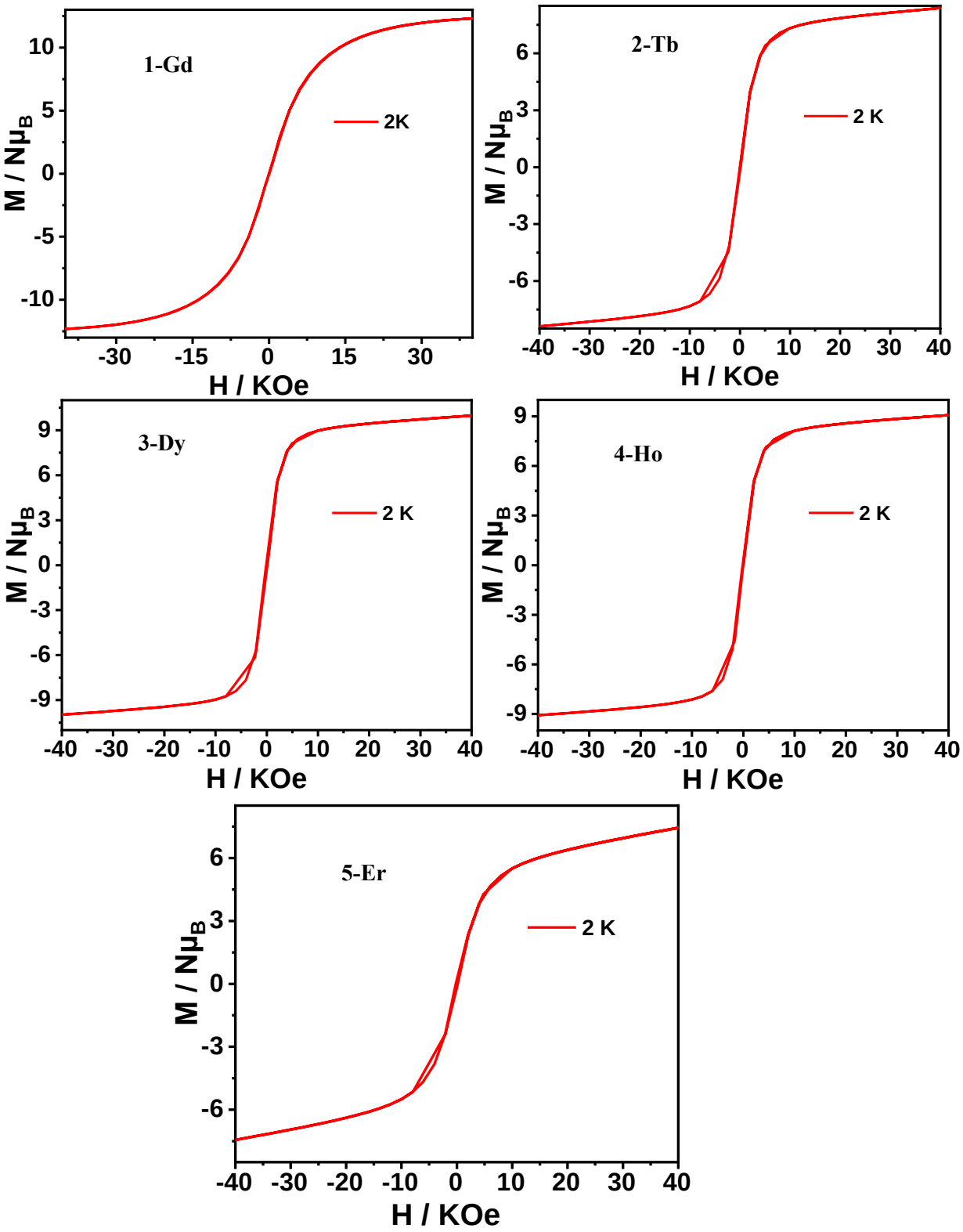


Figure S3. Magnetization (M) vs field hysteresis loops for complexes **1-Gd**, **2-Tb**, **3-Dy**, **4-Ho**, and **5-Er** at the indicated temperature.

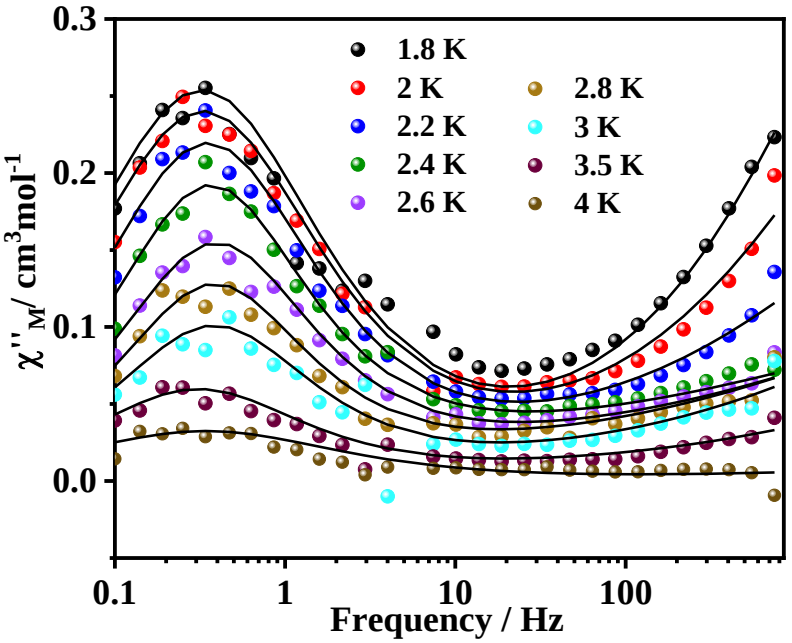


Figure S4. χ''_M vs. frequency plots for **4-Ho** at H_{dc} = 1600 Oe. The solid lines are fitted values obtained from the CC-FIT[1] program using the parameters given in the text.

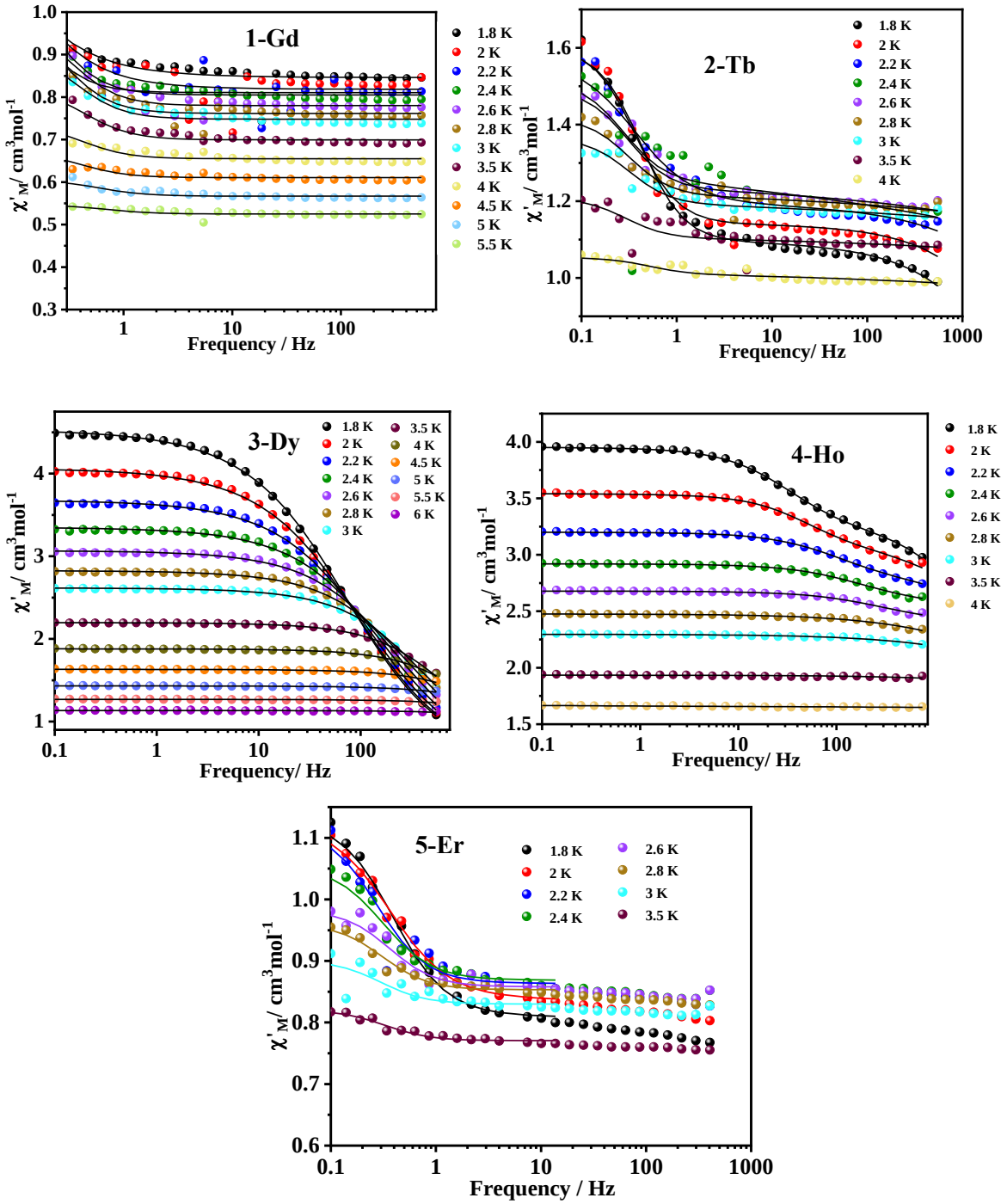


Figure S5. χ'_M vs. frequency plots for **3-Dy** and **4-Ho** (middle-left and middle-right) at H_{dc} = 0 Oe and **1-Gd**, **2-Tb**, and **5-Er** (top-left and top-right and bottom), at H_{dc} = 3600, 2000, and 2400 Oe, respectively. The solid lines are fitted values obtained from the CC-FIT[1] program using the parameters given in the text.

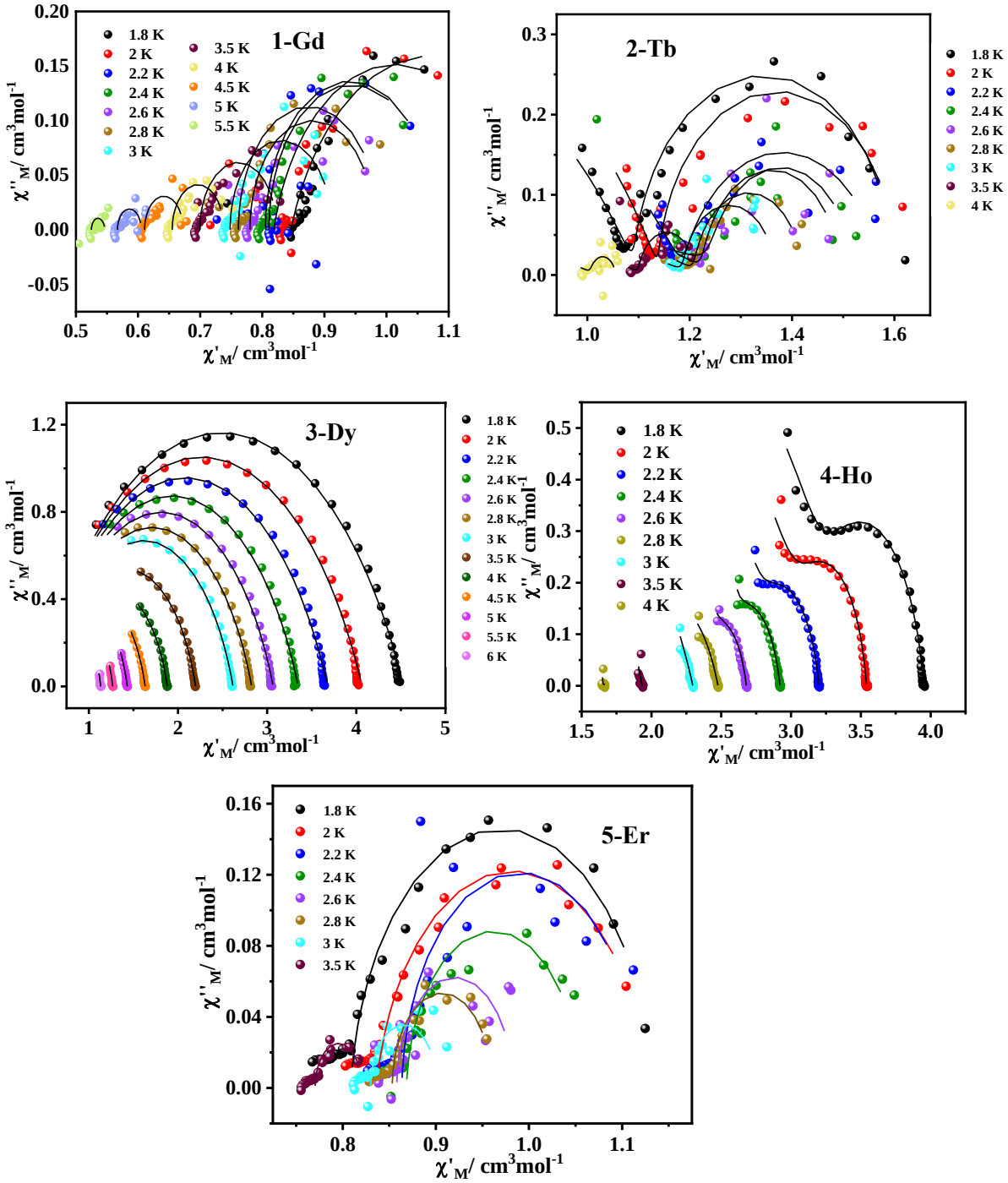


Figure S6. χ''_M vs. χ'_M plots for **3-Dy** and **4-Ho** (middle-left and middle-right) at H_{dc} = 0 Oe and **1-Gd**, **2-Tb**, and **5-Er** (top-left and top-right and bottom), at H_{dc} = 3600, 2000, and 2400 Oe, respectively. The solid lines are fitted values obtained from the CC-FIT[1] program using the parameters given in the text.

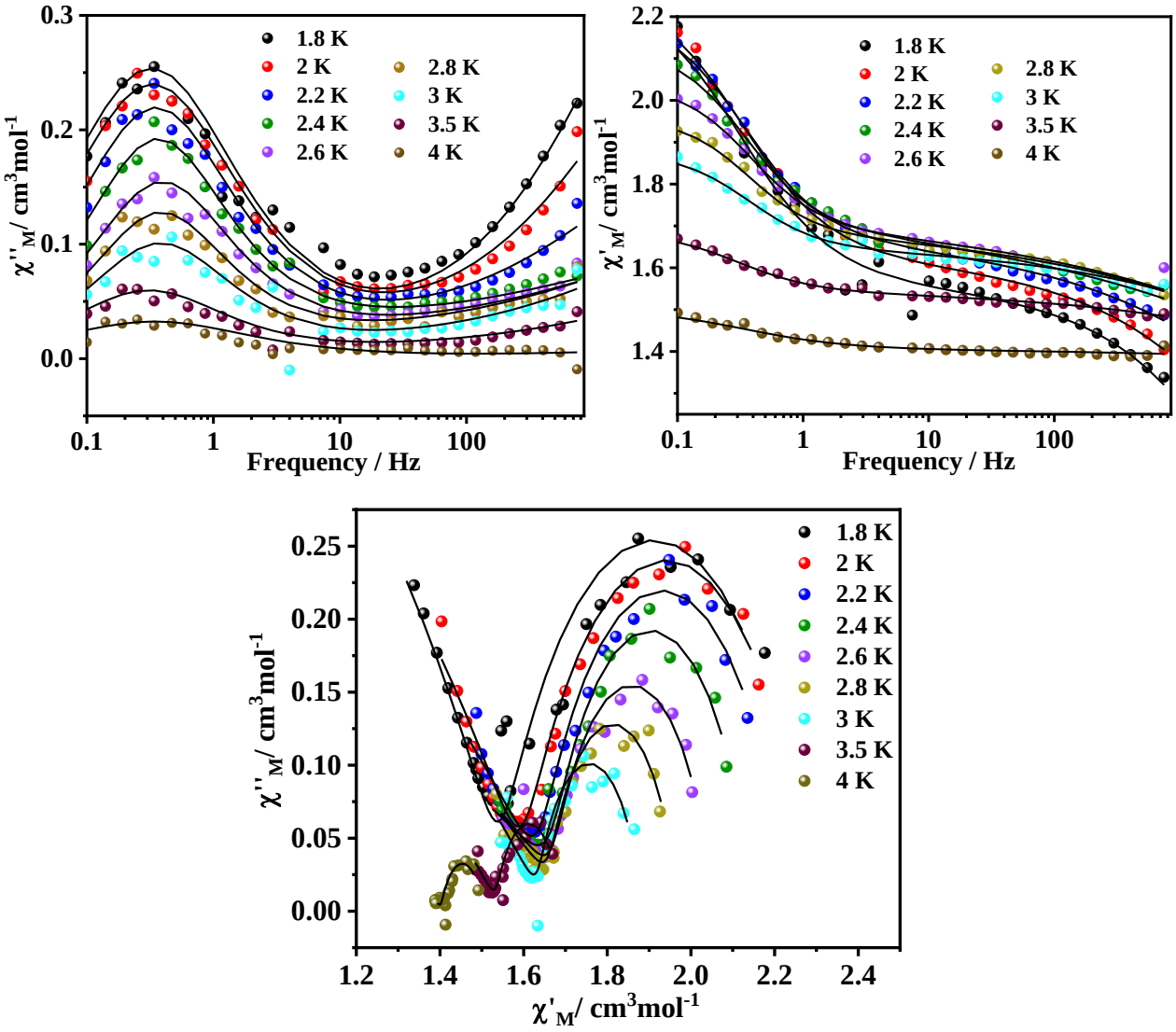


Figure S7. χ''_M vs. frequency (top, left), χ'_M vs. frequency (top, right), and χ''_M vs. χ'_M (bottom) plots for **4-Ho** at H_{dc} = 1600 Oe. The solid lines are fitted values obtained from the CC-FIT[1] program using the parameters given in the text.

Table S4. Cole-Cole fit values of **1-Gd** between 1.8–5.5 K under a dc field of 3600 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	Residual
1.8	0.8461	1.30	1.77	0.21	0.015
2	0.8185	1.21	1.11	0.16	0.033
2.2	0.8108	1.08	0.98	0.29E-12	0.034
2.4	0.8051	1.08	0.69	0.58E-12	0.004
2.6	0.7801	0.98	0.48	0.86E-12	0.009
2.8	0.7616	0.99	0.55	0.83E-12	0.008
3	0.7482	0.91	0.43	0.87E-12	0.006
3.5	0.6995	0.82	0.36	0.15E-11	0.003
4	0.6551	0.74	0.38	0.23E-11	0.003
4.5	0.6105	0.67	0.39	0.28E-11	0.003
5	0.5669	0.61	0.26	0.23E-11	0.002
5.5	0.5249	0.55	0.22	0.31E-11	0.002

Table S5. Cole-Cole fit values of **2-Tb** between 1.8–4 K under a dc field of 2000 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	χ2/cm^3 mol^{-1}	τ2/s	α2	Residual
1.8	0.24E-11	1.10	0.23E-4	0.34	0.50	0.37	0.01	0.02
2	0.36E-11	1.14	0.18E-4	0.30	0.46	0.43	0.01	0.02
2.2	0.28E-17	1.22	0.11E-5	0.61	0.33	0.43	0.03	0.03
2.4	0.16E-16	1.27	0.46E-8	0.79	0.27	0.58	0.06	0.16
2.6	0.25E-17	1.24	0.21E-7	0.70	0.26	0.52	0.03	0.03
2.8	0.69E-17	1.23	0.48E-10	0.81	0.20	0.51	0.25E-6	0.01
3	0.26E-16	1.23	0.69E-15	0.90	0.16	0.50	0.59E-7	0.006
3.5	0.63E-16	1.95	0.11E-14	0.99	0.09	0.57	0.53E-9	0.02
4	0.85E-16	1.74	0.93E-15	0.99	0.03	0.38	0.13E-8	0.005

Table S6. Cole-Cole fit values of **3-Dy** between 1.8–6 K under a dc field of 0 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	Residual
1.8	0.45	4.53	0.002	0.34	0.03
2	0.46	4.07	0.002	0.33	0.03
2.2	0.49	3.68	0.001	0.31	0.03
2.4	0.52	3.35	0.0009	0.30	0.02
2.6	0.55	3.07	0.0007	0.28	0.01
2.8	0.59	2.83	0.0005	0.26	0.01
3	0.62	2.62	0.0004	0.25	0.007
3.5	0.67	2.20	0.0002	0.22	0.003
4	0.65	1.88	0.0001	0.21	0.002
4.5	0.44	1.63	0.00006	0.21	0.0009
5	0.26E-4	1.43	0.00002	0.21	0.0007
5.5	0.28E-4	1.27	0.00001	0.19	0.0006
6	0.31E-4	1.13	0.000009	0.18	0.0006

Table S7. Cole-Cole fit values of **4-Ho** between 1.8–4 K under a dc field of 0 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	χ2/cm^3 mol^{-1}	τ2/s	α2	Residual
1.8	1.59	1.63	0.57E-4	0.18	0.74	0.005	0.19	0.005
2	0.34E-6	3.11	0.81E-5	0.38	0.43	0.003	0.13	0.006
2.2	0.11E-6	2.79	0.60E-5	0.26	0.41	0.002	0.20	0.002
2.4	0.20E-6	2.55	0.85E-5	0.23E-13	0.37	0.0009	0.23	0.002
2.6	0.11E-9	2.39	0.53E-5	0.56E-13	0.29	0.0005	0.25	0.002
2.8	0.53E-9	2.03	0.15E-5	0.18E-13	0.45	0.00008	0.38	0.002
3	0.26E-8	1.21	0.65E-6	0.96E-20	1.08	0.000005	0.43	0.001
3.5	0.44E-16	1.70	0.16E-5	0.82E-20	0.23	0.000009	0.44	0.002
4	0.80E-16	1.53	0.19E-5	0.13E-19	0.22	0.00002	0.97	0.001

Table S8. Cole-Cole fit values of **4-Ho** between 1.8–4 K under a dc field of 1600 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	χ2/cm^3 mol^{-1}	τ2/s	α2	Residual
1.8	0.33E-7	1.55	0.20E-4	0.41	0.73	0.50	0.23	0.03
2	0.43E-7	1.61	0.84E-5	0.49	0.67	0.49	0.22	0.005
2.2	0.83E-7	1.67	0.17E-5	0.61	0.56	0.46	0.17	0.004
2.4	0.12E-7	1.73	0.53E-7	0.77	0.42	0.44	0.11	0.004
2.6	0.13E-7	1.72	0.70E-7	0.73	0.34	0.41	0.10	0.005
2.8	0.17E-7	1.70	0.92E-7	0.71	0.27	0.42	0.08	0.002
3	0.16E-7	1.65	0.16E-6	0.65	0.23	0.43	0.11	0.005
3.5	0.78E-7	1.55	0.18E-7	0.67	0.14	0.53	0.15	0.001
4	0.15E-6	1.41	0.13E-10	0.71	0.10	0.47	0.28	0.002

Table S9. Cole-Cole fit values of **5-Er** between 1.8–3.5 K under a dc field of 2400 Oe.

T (K)	χS(Total) /cm^3 mol^{-1}	χ1/cm^3 mol^{-1}	τ1/s	α1	Residual
1.8	0.79	1.15	0.40	0.15	0.01
2	0.82	1.15	0.44	0.20	0.005
2.2	0.85	1.14	0.59	0.15	0.02
2.4	0.85	1.07	0.51	0.17	0.006
2.6	0.85	0.99	0.44	0.08	0.006
2.8	0.84	0.99	0.58	0.26	0.002
3	0.82	0.94	0.91	0.37	0.004
3.5	0.76	0.84	0.66	0.37	0.0005

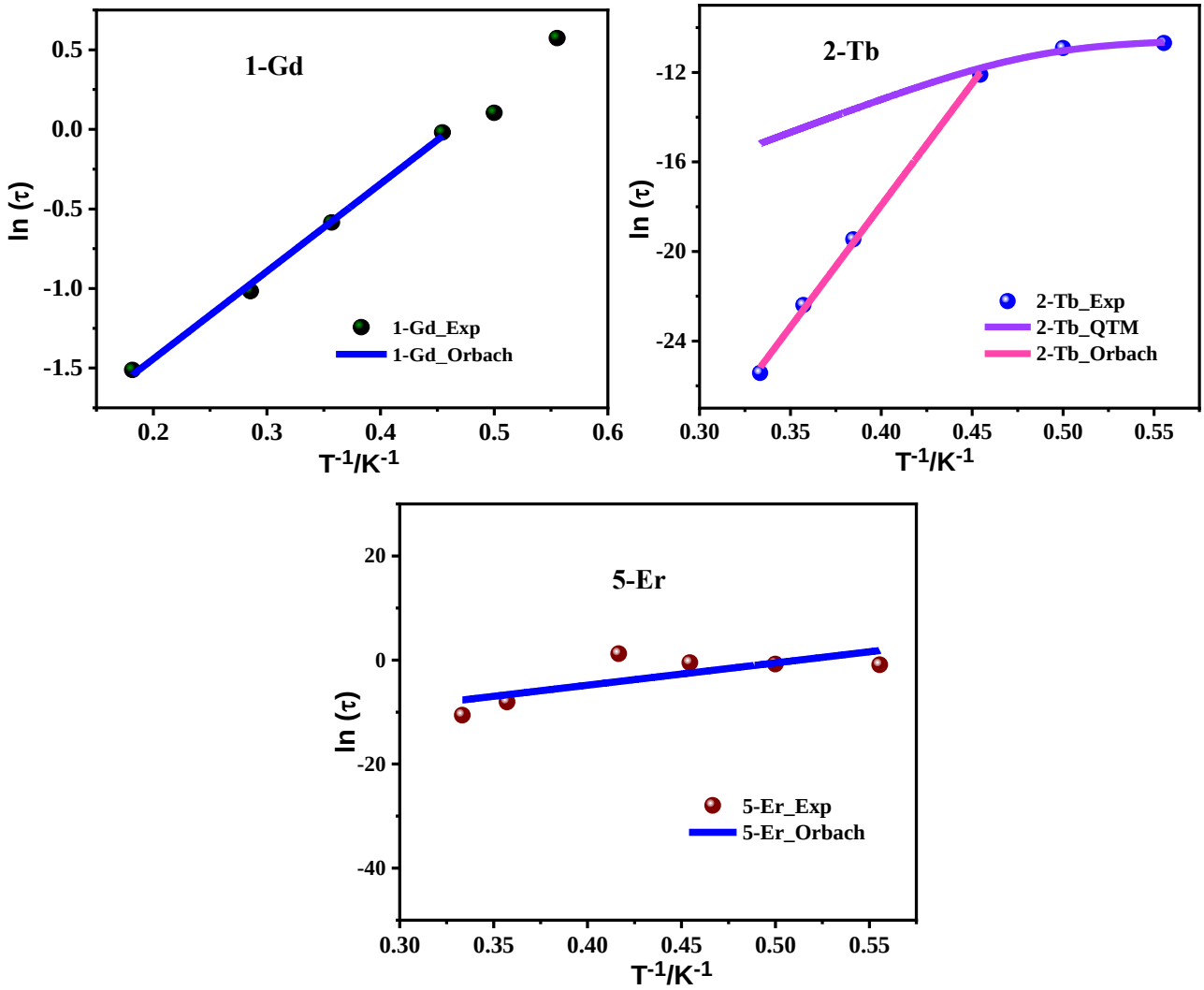


Figure S8. Magnetization relaxation time (τ), plotted as ln(τ) versus T^{-1} for complex **1-Gd**, **2-Tb**, and **5-Er**.

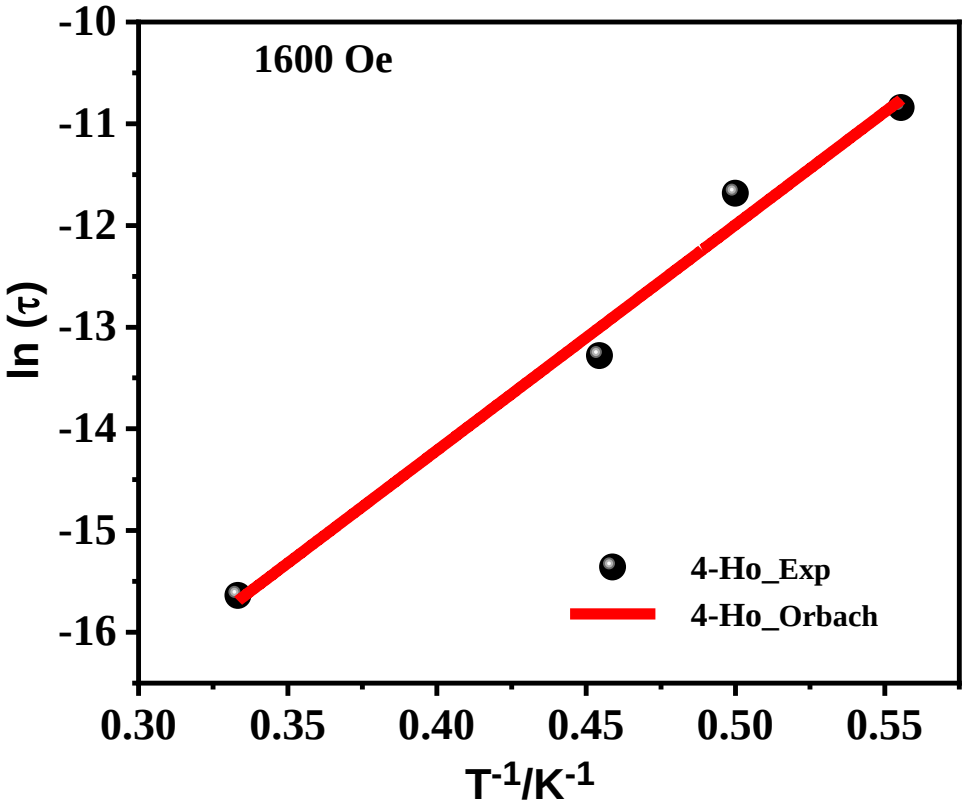


Figure S9. Magnetization relaxation time (τ), plotted as ln(τ) versus T^{-1} for complex **4-Ho** at 1600 Oe.

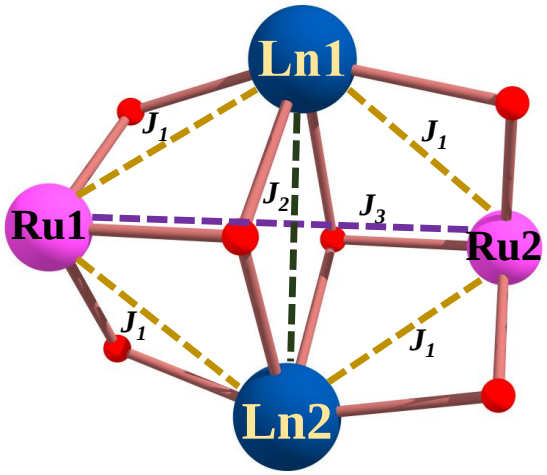


Figure S10. Magnetic cores with possible exchange interactions in all the complexes.

Table S10. Energies of the lowest Kramers doublets (KDs)/ Ising doublets (IDs) (cm^{-1}) of Ln^{III} centers and the *g*-tensor of the ground Kramers doublet/ Ising doublets of complexes **2-Tb**, **3-Dy**, **4-Ho**, and **5-Er**.

	2-Tb		**3-Dy**		**4-Ho**		**5-Er**	
	Tb1	**Tb2**	**Dy1**	**Dy2**	**Ho1**	**Ho2**	**Er1**	**Er2**
KDs/ IDs	0.4	0.4	0.0	0.0	1.0	1.0	0.0	0.0
	118.3	118.3	101.8	101.8	70.1	70.1	36.8	36.8
	234.3	234.3	136.9	136.9	116.7	116.7	54.8	54.8
	310.0	310.0	172.4	172.4	167.7	167.7	108.9	108.9
	359.9	359.9	208.2	208.2	231.4	231.4	174.8	174.8
	393.7	393.7	260.1	260.1	240.5	240.5	220.7	220.7
	478.2	478.2	351.2	351.2	256.0	256.0	273.2	273.2
			551.1	551.1	263.2	263.2	338.1	338.1
g_x	0.0000	0.0000	0.0050	0.0050	0.0000	0.0000	1.9402	1.9400
g_y	0.0000	0.0000	0.0146	0.0146	0.0000	0.0000	2.3804	2.3812
g_z	17.2084	17.2126	19.7554	19.7701	19.0756	19.0840	13.6903	13.6868

Table S11. *g*-Tensors of the Tb^{III}, Dy^{III}, Ho^{III}, and Er^{III} fragments of complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er** That Originate from the corresponding ground Kramers Doublets/ Ising Doublets:

		2-Tb		**3-Dy**		**4-Ho**		**5-Er**	
KD /ID		**Tb1**	**Tb2**	**Dy1**	**Dy2**	**Ho1**	**Ho2**	**Er1**	**Er2**
1	g_x	0.0000	0.0000	0.0050	0.0050	0.0000	0.0000	1.9402	1.9400
	g_y	0.0000	0.0000	0.0146	0.01456	0.0000	0.0000	2.3804	2.3812
	g_z	17.2084	17.2126	19.7554	19.7701	19.0756	19.0840	13.6903	13.6868
2	g_x	0.0000	0.0000	0.5421	0.5393	0.0000	0.0000	2.5820	2.5823
	g_y	0.0000	0.0000	0.7975	0.7977	0.0000	0.0000	3.8440	3.8431
	g_z	13.2876	13.2945	17.2960	17.2919	15.1488	15.1482	10.0216	10.0252
3	g_x	0.0000	0.0000	1.0504	1.0517	0.0000	0.0000	0.1503	0.1512
	g_y	0.0000	0.0000	2.9782	2.9762	0.0000	0.0000	2.3086	2.3099
	g_z	9.8469	9.8491	15.5823	15.5826	11.1683	11.1717	13.7700	13.7692
4	g_x	0.0000	0.0000	0.9382	0.9406	0.0000	0.0000	2.7606	2.7608
	g_y	0.0000	0.0000	3.7319	3.7415	0.0000	0.0000	3.7345	3.7332
	g_z	10.7105	10.7088	11.2496	11.2658	12.2590	12.2518	10.6054	0.6080
5	g_x	0.0000	0.0000	8.4528	8.4541	0.0000	0.0000	9.0317	9.0393
	g_y	0.0000	0.0000	6.3095	6.3140	0.0000	0.0000	6.4878	6.4825
	g_z	10.3484	10.3469	1.2298	1.2316	9.6594	9.6645	1.2245	1.2250
6	g_x	0.0000	0.0000	2.6709	2.6749	0.0000	0.0000	0.7680	0.7693
	g_y	0.0000	0.0000	3.5113	3.5109	0.0000	0.0000	1.6818	1.6829
	g_z	16.1718	16.1714	13.4222	13.4328	13.6160	13.6145	7.4883	7.4902
7	g_x			0.6690	0.6699	0.0000	0.0000	9.6692	9.6689
	g_y			0.7901	0.7896	0.0000	0.0000	6.8351	6.8341
	g_z			16.9103	16.9140	16.1450	16.1459	2.4302	2.4306
8	g_x			0.0102	0.0103			0.5218	0.5223
	g_y			0.0209	0.0210			0.7695	0.7679
	g_z			19.3829	19.3783			16.5222	16.5235

Table S12. Energies of the lowest Kramers doublets (cm^{-1}) of Ru^{III} centers and the *g*-tensor of the ground Kramers doublets of complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**.

	2-Tb		3-Dy		4-Ho		5-Er	
	Ru1	**Ru2**	**Ru1**	**Ru2**	**Ru1**	**Ru2**	**Ru1**	**Ru2**
KDs	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	4057.7	4057.8	4179.5	4057.8	4344.6	4177.2	3854.4	3854.2
	5176.4	5176.4	5313.3	5176.4	5405.4	5227.4	4909.4	4909.3
	15089.5	15090.1	15745.7	15090.0	15707.9	15035.1	15143.0	15142.6
	15308.9	15309.5	15964.2	15309.4	15818.5	15146.2	15354.1	15354.0
	16338.2	16338.8	16995.0	16338.7	15984.8	15313.2	15465.3	15465.5
g_x	2.6158	2.5926	2.6051	2.5926	2.5906	2.5851	2.6571	2.6260
g_y	2.4868	2.4880	2.4778	2.4880	2.4745	2.4848	2.5174	2.5168
g_z	1.5450	1.5608	1.5583	1.5608	1.5715	1.5687	1.5071	1.5263

Table S13. RASSI energies of the lowest spin-orbit states (cm^{-1}) on each Ln^{III} center in complexes **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**.

2-Tb		3-Dy		4-Ho		5-Er	
Tb1	**Tb2**	**Dy1**	**Dy2**	**Ho1**	**Ho2**	**Er1**	**Er2**
0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.730	0.730	101.821	101.819	1.926	1.926	36.814	36.814
115.624	115.624	136.852	136.851	62.045	62.044	54.784	54.784
121.058	121.058	172.390	172.389	78.074	78.074	108.862	108.862
227.484	227.484	208.231	208.229	96.109	96.109	174.795	174.794
241.070	241.070	260.085	260.083	137.294	137.296	220.722	220.722
307.359	307.359	351.155	351.154	148.913	148.914	273.151	273.151
312.549	312.550	551.136	551.135	186.407	186.407	338.080	338.080
351.423	351.424	3576.847	3576.848	231.370	231.368	6599.072	6599.072
368.446	368.446	3668.505	3668.504	240.466	240.467	6643.129	6643.129
393.715	393.715	3703.706	3703.706	256.040	256.038	6664.909	6664.908
473.686	473.686	3744.026	3744.026	260.164	260.164	6712.468	6712.468
482.704	482.704	3784.323	3784.323	266.169	266.167	6733.578	6733.577
2210.070	2210.070	3829.414	3829.414	289.241	289.241	6755.593	6755.593
2217.156	2217.156	3923.737	3923.737	305.042	305.042	6819.817	6819.817
2276.422	2276.423	6134.273	6134.273	326.710	326.711	10649.870	10649.870
2284.215	2284.216	6177.055	6177.056	332.145	332.146	10695.454	10695.454
2308.256	2308.256	6235.813	6235.814	5200.007	5200.007	10715.410	10715.410
2314.726	2314.726	6299.197	6299.198	5200.049	5200.049	10736.570	10736.569
2342.526	2342.525	6332.705	6332.705	5248.294	5248.294	10748.933	10748.933
2390.879	2390.879	6393.669	6393.670	5250.156	5250.157	10794.440	10794.440
2445.027	2445.028	8092.519	8092.519	5272.941	5272.941	13396.370	13396.370
2546.758	2546.758	8130.445	8130.445	5278.751	5278.750	13462.360	13462.360
2562.161	2562.161	8220.142	8220.143	5284.262	5284.261	13512.938	13512.938
3545.581	3545.581	8268.499	8268.500	5291.890	5291.889	13571.390	13571.390
3580.851	3580.851	8334.336	8334.336	5300.155	5300.155	13628.199	13628.199
3606.169	3606.170	9603.950	9603.951	5304.292	5304.291	18997.725	18997.725
3652.122	3652.122	9698.655	9698.656	5310.299	5310.299	19038.589	19038.589
3693.566	3693.566	9780.911	9780.912	5326.264	5326.264	19084.827	19084.827
3751.905	3751.906	9851.484	9851.485	5327.560	5327.560	19101.560	19101.560
3767.381	3767.382	10019.850	10019.852	5375.913	5375.913	19172.361	19172.361
3825.866	3825.866	10046.477	10046.479	5375.928	5375.928	22662.594	22662.594
3927.711	3927.711	10068.069	10068.071	8938.977	8938.977	22672.446	22672.446
4642.877	4642.878	10109.241	10109.242	8939.311	8939.311	22692.013	22692.013
4691.423	4691.423	10157.042	10157.044	8958.606	8958.606	22721.978	22721.978

4710.740	4710.741	10224.033	10224.035	8961.029	8961.030	22735.702	22735.702
4752.610	4752.611	10785.809	10785.810	8975.207	8975.206	22768.508	22768.508
4759.156	4759.156	10874.982	10874.982	8980.304	8980.304	23607.309	23607.309
4806.214	4806.214	11072.490	11072.490	8984.514	8984.513	23656.097	23656.097
4818.781	4818.781	11467.380	11467.380	8995.940	8995.939	25171.728	25171.728
5359.433	5359.434	11514.814	11514.814	9006.486	9006.485	25200.924	25200.923
5385.907	5385.908	11538.124	11538.124	9016.659	9016.658	25250.715	25250.715
5568.761	5568.761	11554.769	11554.770	9027.948	9027.947	25312.266	25312.265
5606.753	5606.753	11570.665	11570.666	9080.775	9080.776	27261.248	27261.248
5615.630	5615.630	13393.904	13393.905	9082.723	9082.723	27285.935	27285.935
5880.743	5880.744	13447.292	13447.294	11694.484	11694.484	27336.824	27336.824
6011.560	6011.560	13492.882	13492.883	11694.588	11694.588	27567.579	27567.579
6089.740	6089.741	13511.609	13511.611	11715.791	11715.791	27653.557	27653.557
6236.889	6236.889	14863.810	14863.812	11717.762	11717.761	27776.042	27776.042
23586.683	23586.687	14909.587	14909.589	11726.939	11726.938	27829.848	27829.848
23588.051	23588.055	14948.291	14948.293	11737.892	11737.890	27870.822	27870.822
23626.003	23626.007	15768.404	15768.406	11746.750	11746.748	27913.163	27913.163
23635.397	23635.400	15780.578	15780.580	11753.619	11753.617	27936.462	27936.462
23645.529	23645.533	16304.349	16304.351	11774.051	11774.049	31843.103	31843.104
23663.778	23663.782	24508.501	24508.495	11818.220	11818.220	32083.168	32083.169
23665.912	23665.916	24598.096	24598.091	11825.000	11825.000	32119.956	32119.956
23718.611	23718.615	24638.132	24638.126	13813.902	13813.901	32180.825	32180.825
23719.308	23719.312	24702.616	24702.606	13823.269	13823.267	32193.272	32193.273
29593.638	29593.641	24844.522	24844.514	13880.770	13880.769	32247.491	32247.491
29606.612	29606.615	25094.061	25094.057	13928.011	13928.008	32274.323	32274.323
29611.442	29611.445	25205.639	25205.637	13942.395	13942.395	32413.607	32413.607
29615.738	29615.741	25257.503	25257.502	13966.067	13966.068	32709.547	32709.547
29619.729	29619.732	25303.100	25303.099	13985.956	13985.955	32734.884	32734.884
29621.646	29621.649	25321.648	25321.647	14015.552	14015.550	32783.323	32783.322
29625.391	29625.394	25349.016	25349.012	14069.394	14069.391	32859.144	32859.144
30165.439	30165.442	25389.147	25389.144	20566.200	20566.186	32887.903	32887.903
30165.485	30165.488	25427.791	25427.786	20574.463	20574.449	32940.062	32940.061
30219.289	30219.292	27242.768	27242.760	20594.288	20594.276	33242.943	33242.943
30223.954	30223.957	27304.962	27304.953	20615.372	20615.360	33256.498	33256.499
30261.595	30261.597	27347.926	27347.918	20621.658	20621.646	33263.716	33263.716
30285.784	30285.787	27373.225	27373.218	20640.545	20640.532	33294.511	33294.511
30303.145	30303.147	27410.028	27410.021	20641.906	20641.893	33953.010	33953.010
30339.549	30339.552	27442.636	27442.624	20688.764	20688.752	33973.250	33973.249
30353.764	30353.767	27519.317	27519.315	20691.230	20691.219	33986.239	33986.239
30356.341	30356.344	27530.772	27530.769	20741.281	20741.269	33994.505	33994.505
30384.773	30384.776	27568.489	27568.483	20741.652	20741.640	34020.425	34020.425
30427.109	30427.112	27605.795	27605.789	23370.271	23370.258	37420.157	37420.157
30436.290	30436.293	27671.166	27671.163	23370.843	23370.830	37630.011	37630.011
30465.534	30465.537	27709.370	27709.365	23380.064	23380.052	37663.640	37663.640
30468.738	30468.741	27722.161	27722.156	23384.043	23384.031	37712.431	37712.431
30553.497	30553.501	27761.204	27761.198	23385.933	23385.920	37737.921	37737.921
30555.086	30555.089	27780.578	27780.574	23831.429	23831.416	37773.460	37773.460
30561.202	30561.205	27876.003	27875.996	23839.937	23839.924	37822.367	37822.367
30562.350	30562.353	27967.197	27967.191	23877.403	23877.391	37907.075	37907.075
30691.531	30691.535	28790.952	28790.946	23895.164	23895.151	37970.645	37970.645
30691.563	30691.566	28853.033	28853.027	23906.121	23906.108	40393.422	40393.422
31208.138	31208.141	28897.412	28897.408	23912.454	23912.442	40607.157	40607.158
31215.360	31215.362	28921.463	28921.462	23946.769	23946.757	40636.687	40636.687
31333.848	31333.851	28947.537	28947.534	23951.535	23951.523	40688.054	40688.054
31340.070	31340.073	28965.734	28965.731	23958.062	23958.054	40739.177	40739.177
31365.189	31365.192	28988.548	28988.546	23959.295	23959.286	40753.458	40753.458
31389.218	31389.221	29011.529	29011.526	23961.433	23961.422	40798.133	40798.134
31391.349	31391.352	29038.555	29038.550	23969.314	23969.308	40846.175	40846.176
31427.709	31427.711	29078.745	29078.739	23972.430	23972.425	40900.507	40900.507
31440.010	31440.013	29102.913	29102.909	23974.303	23974.293	40962.274	40962.274

31465.844	31465.847	29114.018	29114.014	23977.598	23977.587	41268.994	41268.994
31474.382	31474.385	29127.964	29127.958	23978.779	23978.769	41337.690	41337.690
31500.179	31500.182	29141.086	29141.079	23986.584	23986.572	41394.992	41394.992
31504.390	31504.392	29146.957	29146.952	23989.562	23989.551	42028.989	42028.989
31656.468	31656.471	29165.231	29165.222	23997.574	23997.564	42076.556	42076.557
31657.490	31657.493	29177.228	29177.221	24006.859	24006.847	42160.484	42160.484
31711.477	31711.480	29190.619	29190.612	24011.713	24011.703	49410.936	49410.937
31713.782	31713.785	29213.555	29213.547	24020.054	24020.044	49426.420	49426.420
31734.830	31734.833	29238.289	29238.280	24020.617	24020.604	49665.233	49665.233
31755.188	31755.191	29259.240	29259.234	24027.205	24027.192	49758.769	49758.769
31764.549	31764.551	29300.817	29300.810	24037.744	24037.736	49826.764	49826.764
31786.531	31786.534	29313.036	29313.028	24038.250	24038.243	49972.044	49972.044
31789.419	31789.422	29353.271	29353.264	25756.550	25756.537	50010.539	50010.540
31807.441	31807.444	29369.827	29369.825	25765.974	25765.961	50019.023	50019.023
31827.677	31827.680	29414.073	29414.069	25789.880	25789.866	50048.227	50048.227
31860.938	31860.941	29605.866	29605.862	25840.439	25840.426	50073.712	50073.713
31863.042	31863.045	29660.204	29660.198	25851.521	25851.509	50114.182	50114.182
31946.313	31946.316	29678.964	29678.959	25860.017	25860.004	50179.076	50179.076
31955.882	31955.886	29739.589	29739.582	25881.373	25881.361	50198.100	50198.101
31969.493	31969.496	31200.545	31200.548	26264.087	26264.075	50293.430	50293.431
31977.475	31977.478	31215.614	31215.612	26268.353	26268.340	50382.537	50382.537
32070.764	32070.767	31281.127	31281.126	26323.006	26322.994	50459.355	50459.355
32071.182	32071.186	31323.627	31323.625	26334.123	26334.111	50525.858	50525.857
32252.786	32252.789	31371.802	31371.800	26344.604	26344.590	50573.474	50573.474
32271.777	32271.780	31406.004	31406.001	27454.683	27454.669	50642.845	50642.846
32276.033	32276.036	31427.653	31427.651	27508.354	27508.342	50718.030	50718.031
32294.516	32294.519	31483.001	31483.001	27542.073	27542.061	51404.091	51404.091
32313.857	32313.859	31537.162	31537.160	28924.883	28924.876	51552.494	51552.494
32377.922	32377.925	31557.966	31557.963	28925.137	28925.129	51658.970	51658.970
32379.106	32379.108	31581.021	31581.018	28929.134	28929.123	51770.363	51770.363
32445.364	32445.367	31605.586	31605.582	28929.972	28929.962	51873.052	51873.053
32450.354	32450.357	31633.495	31633.492	28930.297	28930.287	51976.486	51976.486
32479.647	32479.650	31676.349	31676.341	28937.220	28937.209	51996.071	51996.072
32481.598	32481.601	32910.871	32910.870	28938.351	28938.340	52030.868	52030.868
32483.141	32483.143	32925.887	32925.883	28939.415	28939.403	52083.020	52083.020
32491.538	32491.541	32957.588	32957.584	28942.081	28942.071	52166.293	52166.293
32505.212	32505.215	32963.687	32963.684	28943.467	28943.456	52274.536	52274.536
32512.925	32512.927	32977.878	32977.875	28945.495	28945.485	54665.805	54665.805
32528.120	32528.123	32994.521	32994.518	28948.603	28948.592	54707.048	54707.048
32540.946	32540.948	33017.356	33017.353	28950.671	28950.656	56173.272	56173.273
32545.316	32545.319	33038.839	33038.835	28959.038	28959.026	56331.242	56331.243
32566.844	32566.847	33054.935	33054.930	28962.841	28962.827	56452.617	56452.618
32577.045	32577.048	33065.377	33065.372	29453.052	29453.034	56547.608	56547.608
32587.107	32587.110	33075.831	33075.827	29453.546	29453.527	56626.810	56626.810
32604.178	32604.181	33092.338	33092.333	29463.082	29463.066	56689.100	56689.100
32622.312	32622.315	33113.056	33113.054	29464.563	29464.546	56763.037	56763.037
32633.037	32633.040	33217.257	33556.271	29471.902	29471.884	56848.424	56848.425
32641.095	32641.098	33556.271	33605.838	29493.376	29493.362	58578.362	58578.363
32647.100	32647.103	33605.839	33656.769	29501.862	29501.848	58624.651	58624.652
32653.271	32653.274	33656.773	34016.156	29506.429	29506.416	58631.084	58631.084
32729.866	32729.869	34016.155	34038.180	29523.191	29523.175	58659.897	58659.897
32749.628	32749.631	34038.181	34123.877	29523.741	29523.726	58733.949	58733.949
32769.498	32769.501	34123.876	34165.308	29530.650	29530.637	59339.803	59339.803
32778.755	32778.758	34165.311	34197.026	29539.349	29539.335	60962.093	60962.093
32822.810	32822.813	34197.030	34231.819	29555.232	29555.218	60978.723	60978.724
32824.896	32824.898	34231.822	34272.168	29814.586	29814.574	61071.233	61071.233
32841.402	32841.405	34272.172	34805.220	29823.699	29823.688	61096.572	61096.572
32845.928	32845.931	34805.220	34832.048	29840.043	29840.030	61122.328	61122.328
32855.892	32855.895	34832.048	34843.412	29850.028	29850.014	61135.831	61135.831
32865.316	32865.319	34843.414		29862.984	29862.971	61192.755	61192.755

32867.758	32867.761	34856.736	34856.733	29878.640	29878.626	61239.048	61239.048
32903.616	32903.619	34865.977	34865.975	29903.026	29903.014	61260.262	61260.263
32911.402	32911.405	34879.021	34879.019	29921.260	29921.247	67000.379	67000.379
32927.209	32927.211	34895.628	34895.627	29926.915	29926.903	67143.947	67143.946
32935.684	32935.687	34913.128	34913.126	29961.567	29961.557	67233.075	67233.075
32940.889	32940.891	34941.658	34941.657	29962.412	29962.403	67289.631	67289.631
32969.027	32969.030	34989.789	34989.785	32847.997	32847.982	67329.444	67329.444
32976.573	32976.576	35173.914	35173.912	32856.920	32856.906	67373.643	67373.644
32999.672	32999.675	35191.626	35191.621	32863.310	32863.295	76087.761	76087.761
33015.116	33015.119	35204.779	35204.777	32877.040	32877.025	76128.273	76128.273
33031.168	33031.171	35224.824	35224.820	32884.298	32884.283	76149.087	76149.088
33048.254	33048.257	35234.704	35234.703	32887.957	32887.944	83111.494	83111.495
33077.174	33077.177	35265.365	35265.363	32895.366	32895.353	83213.474	83213.475
33096.008	33096.011	35293.850	35293.851	32897.888	32897.875	83253.084	83253.084
33115.126	33115.129	35367.913	35367.908	32907.488	32907.472	83312.891	83312.891
33121.669	33121.672	35900.367	35900.371	32914.991	32914.979	87051.381	87051.382
33158.543	33158.545	36024.313	36024.317	32918.114	32918.102	87149.373	87149.374
33164.392	33164.395	36062.833	36062.834	32927.053	32927.041	87227.161	87227.161
33185.964	33185.967	36134.122	36134.124	32928.548	32928.534	87263.987	87263.987
33198.490	33198.493	37001.937	37001.941	32930.663	32930.650	87330.110	87330.110
33215.230	33215.233	37028.697	37028.697	32939.208	32939.195		
33240.110	33240.113	37064.541	37064.542	32942.818	32942.806		
33321.026	33321.029	37369.271	37369.275	32944.645	32944.632		
33334.161	33334.164	37393.481	37393.484	32952.650	32952.637		
33354.734	33354.737	37401.788	37401.785	32957.184	32957.171		
33379.227	33379.230	37428.562	37428.564	32959.080	32959.068		
33390.360	33390.363	37458.838	37458.837	32977.937	32977.924		
33397.055	33397.059	37469.354	37469.355	32981.407	32981.394		
33420.013	33420.016	37481.579	37481.579	33087.769	33087.749		
33432.537	33432.540	37506.470	37506.470	33087.844	33087.824		
33437.241	33437.244	37547.022	37547.022	33183.645	33183.632		
33466.445	33466.448	37576.178	37576.177	33185.834	33185.821		
33480.451	33480.454	37614.244	37614.245	33199.053	33199.038		
33824.806	33824.809	37632.956	37632.956	33214.900	33214.887		
33841.526	33841.529	37668.160	37668.161	33220.678	33220.665		
33848.067	33848.070	37725.705	37725.705	33230.893	33230.879		
33864.955	33864.958	37745.639	37745.643	33235.190	33235.176		
33881.482	33881.485	37801.340	37801.342	33242.579	33242.564		
33890.654	33890.657	37832.702	37832.701	33254.255	33254.241		
33898.207	33898.210	38653.390	38653.390	33262.148	33262.132		
34108.496	34108.499	38670.878	38670.880	33270.840	33270.825		
34114.938	34114.941	38682.173	38682.173	33271.290	33271.274		
34137.213	34137.216	38689.301	38689.302	33288.920	33288.903		
34161.512	34161.514	38700.659	38700.659	33291.971	33291.955		
34169.484	34169.487	38706.633	38706.633	33306.370	33306.353		
34537.829	34537.832	38718.030	38718.031	33314.258	33314.242		
34597.562	34597.564	38732.217	38732.219	33324.718	33324.704		
34638.357	34638.359	38761.731	38761.733	33330.048	33330.032		
35705.305	35705.308	39115.536	39115.541	33340.607	33340.591		
36532.141	36532.144	39244.687	39244.688	33348.883	33348.870		
36534.261	36534.264	39359.915	39359.914	33355.247	33355.233		
36541.664	36541.666	39429.381	39429.377	33360.397	33360.381		
36544.924	36544.926	39444.522	39444.519	33368.319	33368.304		
36564.061	36564.063	39470.707	39470.704	33370.918	33370.905		
36576.129	36576.131	39495.833	39495.829	33377.964	33377.957		
36618.438	36618.440	39516.013	39516.012	33380.319	33380.307		
36639.179	36639.181	39535.364	39535.364	33413.343	33413.333		
36693.938	36693.940	39578.399	39578.399	33413.933	33413.921		
36707.754	36707.756	40062.306	40062.311	33517.595	33517.599		
36747.629	36747.631	40102.215	40102.221	33517.716	33517.720		

36768.055	36768.058	40279.448	40279.450	34224.487	34224.477		
36769.553	36769.557	40325.063	40325.065	34227.094	34227.082		
36788.729	36788.731	40359.358	40359.358	34302.064	34302.054		
36798.369	36798.372	40380.360	40380.358	34317.900	34317.889		
38589.984	38589.986	40380.361	40385.060	34338.315	34338.303		
38598.985	38598.987	40385.062	40396.261	34390.792	34390.775		
38604.565	38604.567	40396.263	40413.799	34402.745	34402.728		
38619.392	38619.394	40413.800	40421.370	34427.877	34427.861		
38622.102	38622.104	40421.372	40435.889	34442.678	34442.661		
38635.348	38635.350	40435.891	40464.471	34445.898	34445.881		
38651.004	38651.006	40464.471	40532.097	34450.487	34450.471		
38674.156	38674.158	40532.096	40606.466	34469.386	34469.369		
38709.593	38709.596	40606.464	40931.127	34471.501	34471.484		
38719.845	38719.847	40931.120	40936.408	34484.855	34484.837		
38729.464	38729.466	40936.402	40948.559	34487.371	34487.354		
38764.796	38764.798	40948.554	40959.846	34493.045	34493.028		
38773.166	38773.168	40959.842	40986.752	36503.466	36503.449		
39732.169	39732.171	40986.747	41009.475	36507.808	36507.791		
39734.741	39734.743	40986.748	41024.711	36531.634	36531.619		
39738.356	39738.358	41009.470	41042.535	36545.772	36545.755		
39752.053	39752.055	41009.470	41187.300	36561.690	36561.679		
39755.728	39755.730	41024.707	41211.423	36568.472	36568.455		
39779.962	39779.964	41042.534	41229.919	36574.507	36574.496		
39804.984	39804.986	41187.300	41251.628	36607.949	36607.936		
39808.069	39808.071	41211.423	41300.935	36640.654	36640.641		
39811.715	39811.717	41229.918	41548.723	36648.241	36648.230		
39824.212	39824.214	41251.626	41559.245	36693.062	36693.047		
39830.768	39830.770	41300.935	41562.882	36712.955	36712.940		
40260.839	40260.841	41548.717	41567.036	36723.258	36723.244		
40292.642	40292.644	41559.241	41571.135	36746.329	36746.314		
40323.211	40323.213	41562.879	41575.387	36748.667	36748.651		
40324.724	40324.726	41567.031	41578.531	36783.259	36783.242		
40345.302	40345.304	41571.130	41582.911	38390.394	38390.375		
40353.845	40353.847	41575.382	41587.926	38390.515	38390.496		
40359.713	40359.715	41578.528	41594.063	38392.345	38392.329		
40387.933	40387.935	41582.906	41647.878	38420.250	38420.234		
40394.539	40394.542	41587.922	41899.397	38430.454	38430.437		
40602.256	40602.258	41594.057	41929.937	38442.463	38442.445		
40638.387	40638.389	41647.872	41977.540	38459.575	38459.557		

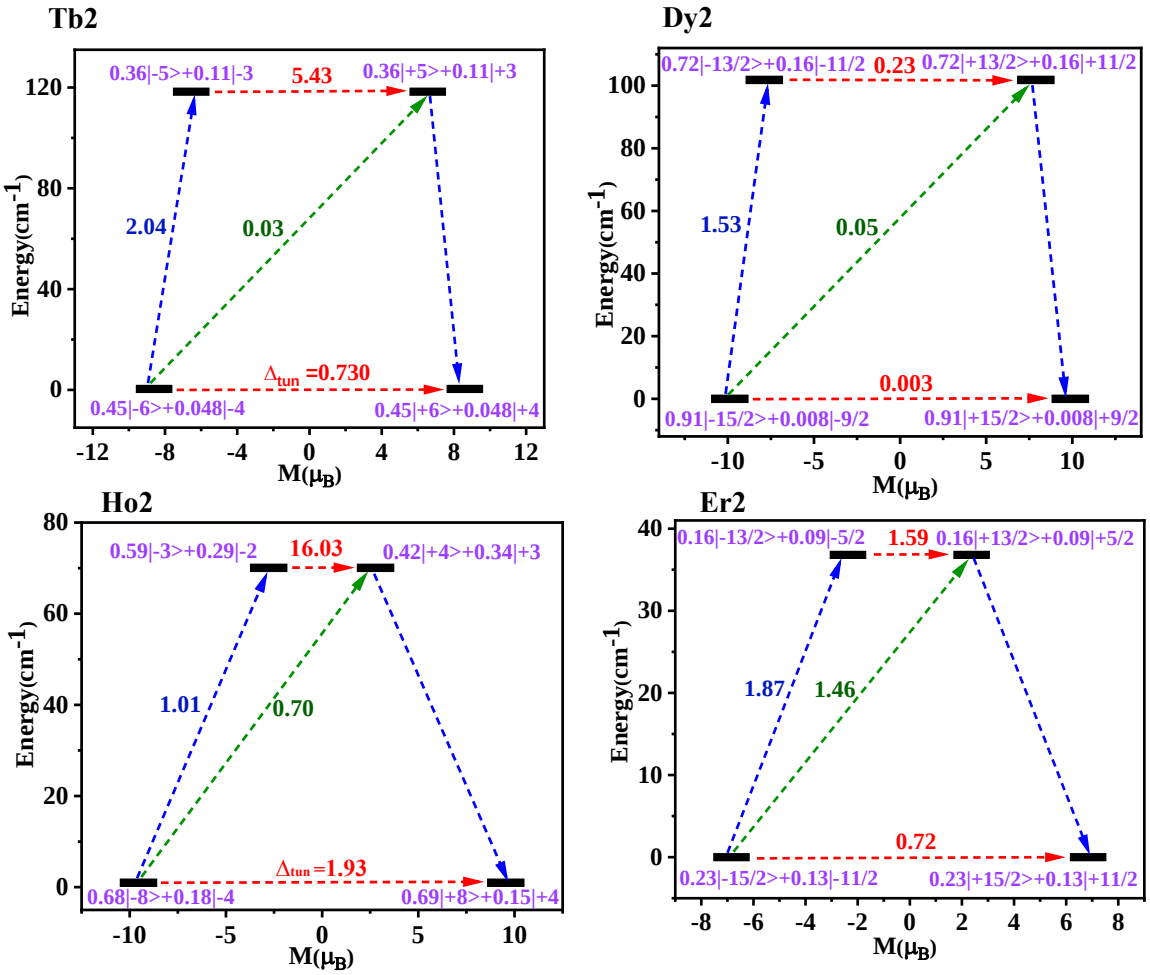


Figure S11. The *ab initio* computed magnetization blocking barrier for the Tb2 ion in **2-Tb** (top, left); Dy2 ion in **3-Dy** (top, right), Ho2 ion in **4-Ho** (bottom, left); Er2 ion in **5-Er** (bottom, right). The thick black line indicates the KDs/IDs as a function of computed magnetic moment. The green/blue arrows show the possible pathway through Orbach/Raman relaxation. The dotted red lines represent the presence of QTM/TA-QTM between the connecting pairs. The numbers provided at each arrow are the mean absolute value for the corresponding matrix element of the transition magnetic moment.

Table S14. SINGLE_ANISO computed crystal field parameters for complex **2-Tb, 3-Dy**, **4-Ho**, and **5-Er**. B_k^q is the crystal field parameter.

k	q	B_k^q	B_k^q	B_k^q	B_k^q	B_k^q	B_k^q	B_k^q	B_k^q
		2-Tb		**3-Dy**		**4-Ho**		**5-Er**	
		Tb1	Tb2	Dy1	Dy2	Ho1	Ho2	Er1	Er2
2	-2	-2.44	-2.45	-0.65	-0.67	-0.04	-0.04	0.18	0.18
	-1	-1.64	-1.64	3.04	3.09	1.31	1.31	0.18	0.18
	0	-2.94	-2.94	-1.10	-1.10	-0.41	-0.41	-0.74	-0.74
	1	1.63	1.62	-4.20	-4.16	0.13	0.13	0.18	0.18
	2	-0.40	-0.40	0.67	0.66	-0.28	-0.27	0.35	0.35
4	-4	-0.02	-0.02	0.002	0.001	-0.002	-0.002	0.01	0.01
	-3	0.04	0.04	0.02	0.02	-0.03	-0.02	0.02	0.02
	-2	-0.06	-0.06	-0.007	-0.007	-0.00001	-0.00004	0.003	0.003
	-1	0.03	0.03	-0.007	-0.008	-0.01	-0.01	-0.02	-0.02
	0	-0.007	-0.007	-0.008	-0.008	-0.003	-0.003	-0.002	-0.002
	1	-0.04	-0.04	0.02	0.02	-0.002	-0.002	-0.02	-0.02
	2	-0.04	-0.04	-0.0003	-0.0004	0.0001	0.00008	0.0007	0.0008
	3	0.07	0.07	0.07	0.07	-0.02	-0.02	-0.01	-0.009
	4	-0.01	-0.01	-0.0005	-0.0004	0.001	0.001	-0.009	-0.009
6	-6	0.0004	0.0004	0.0002	0.0001	-0.0003	-0.0003	0.00004	0.00004
	-5	0.0004	0.0004	-0.0005	-0.0005	-0.0003	-0.0003	0.0003	0.0003
	-4	0.00004	0.00004	0.00008	0.00009	0.0001	0.0002	0.0001	0.0001
	-3	-0.0002	-0.0001	0.0001	0.0001	0.00008	0.00008	-0.0003	-0.0003
	-2	0.0003	0.0003	0.00003	0.00003	-0.00002	-0.00002	0.0003	0.0003
	-1	-0.0002	-0.0002	-0.0002	-0.0002	0.00008	0.00008	0.0003	0.0004
	0	0.00003	0.00003	0.00003	0.00003	-0.00004	-0.00004	0.00002	0.00002
	1	0.0003	0.0003	0.00009	0.00009	0.00002	0.00002	0.0003	0.0003
	2	0.0002	0.0002	-0.00003	-0.00003	-0.00003	-0.00003	0.0002	0.0002
	3	-0.0002	-0.0002	0.0001	0.0001	0.00007	0.00008	-0.0001	-0.0001
	4	0.0000009	0.000002	-0.00005	-0.0002	-0.0001	-0.0001	-0.0001	-0.0001
	5	-0.00002	-0.00001	0.0002	0.0002	-0.0004	-0.0004	-0.0007	-0.0007
	6	-0.0003	-0.0003	0.0002	0.0002	0.00007	0.00006	-0.0004	-0.0004

Table S15. Different spin configurations are employed for extracting *J* values of complex **1-Gd**.

	Gd1	Gd2	Ru1	Ru2	Energy (Hartree)
HS	↑	↑	↑	↑	**–3847.77352**
BS1	↑	↓	↑	↑	**–3847.77361**
BS2	↓	↑	↑	↑	**3847.77361**
BS3	↑	↑	↓	↓	**3847.77371**
BS4	↑	↑	↓	↑	**3847.77362**
BS5	↑	↑	↑	↓	**3847.77362**

Keeping the spin alignments according to the DFT-calculated/PHI-extracted exchange parameters, the spin ground state is predicted to be $S = 6$ (See Figure S13, left and S14), where the spins on the Ru^{III} centers are oriented "down" and the spins on the Gd^{III} centers are oriented "up". The various spin density plots used are shown in Figures S12 and S13, where Figure S13, right, represents the spin ground state of the system. The presence of an unpaired electron in δ-type d_{xy}-orbital of the Ru^{III} ion facilitates spin delocalization through the bridging oxygen atoms, which gains significant spin density; whereas the spin polarization on the Gd^{III} ions opens up a super-exchange pathway, which leads to an antiferromagnetic exchange interaction. The angle between Gd^{III}-O-Gd^{III} is observed to be 113.6°, which is anticipated to exhibit ferromagnetic interaction based on the established correlation by Rajaraman and co-workers,[2,3] and agrees with the DFT computed *J* value of +0.01 cm^{-1}. The exchange interaction between Ru^{III}…Ru^{III} results in antiferromagnetic exchange, which is a 1,3 interaction.[4] The magneto-structural correlation established for these 1,3 interactions involving Gd^{III} ions shows a parabolic trend, where Ru–Gd–Ru angles below 120° result in antiferromagnetic coupling, with smaller angles increasing the coupling strength.[5,6] In **1-Gd**, the Ru–Gd–Ru angle is 104.5°, and hence, the antiferromagnetic exchange interaction shows strong agreement with the previously developed correlation.

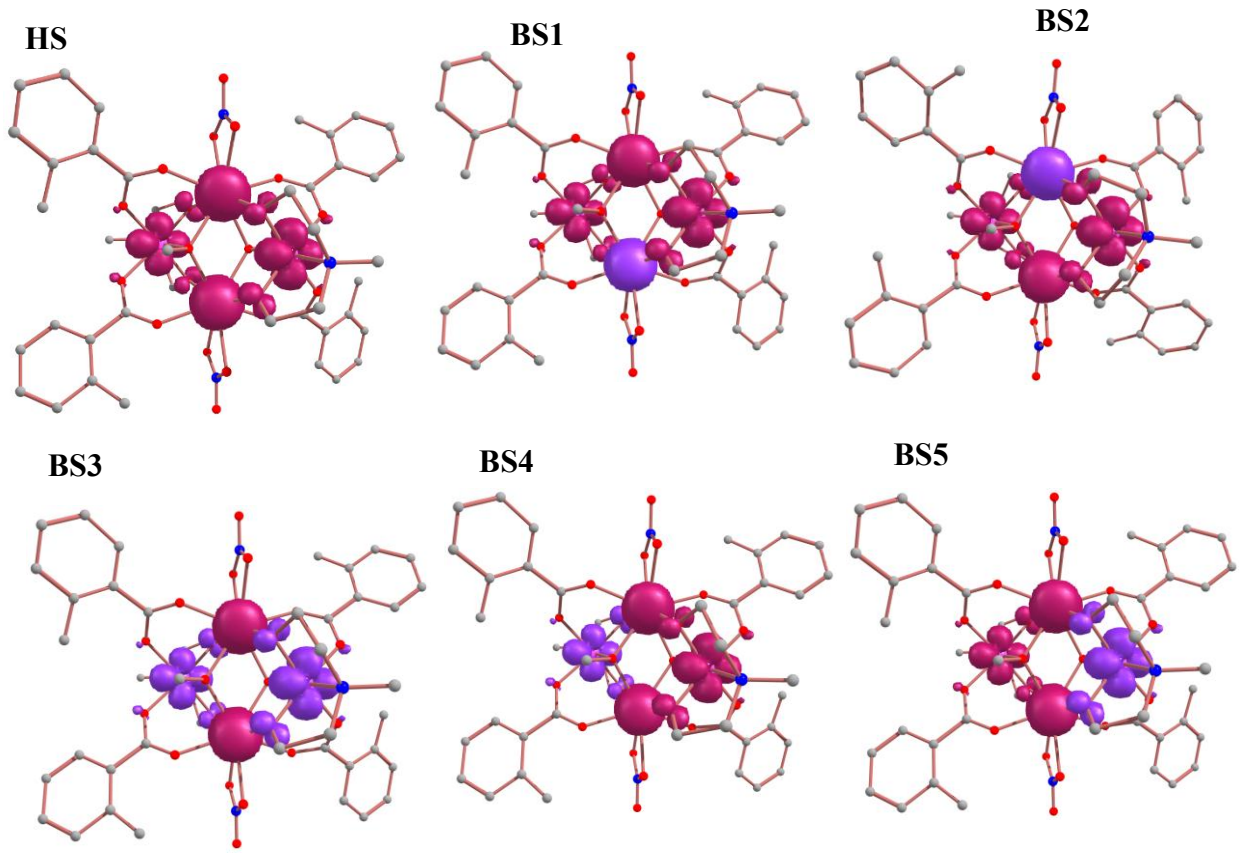


Figure S12. DFT-computed spin-density plots of complex **1-Gd**.

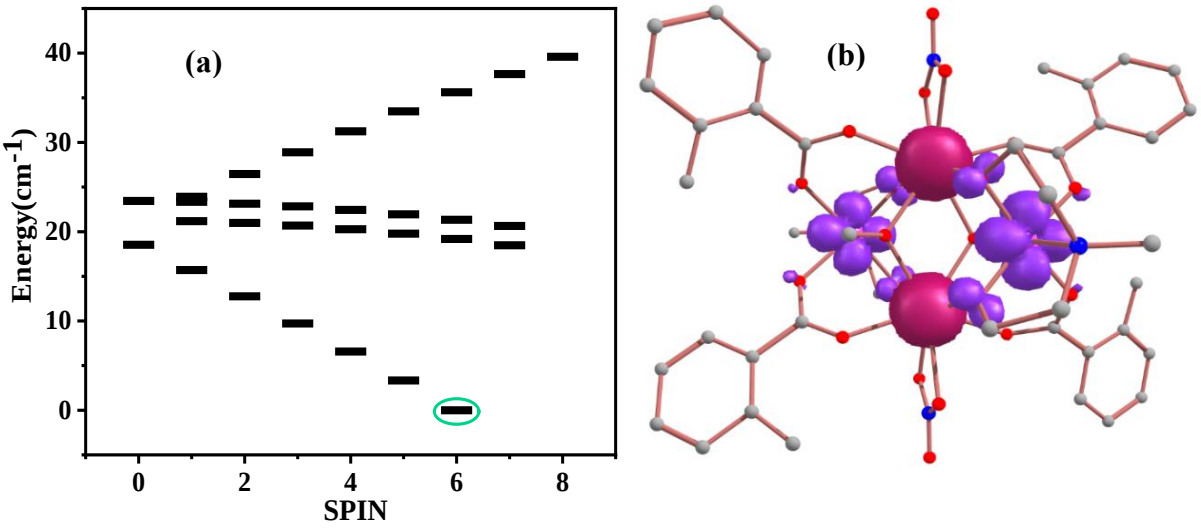


Figure S13. (a) Eigenvalue plots for computed *J* values and (spin ground state is highlighted) of **1-Gd**. (b) DFT-computed spin-density plots of complex **1-Gd**, representing $S = 6$ ground state. The magenta and violet colors represent positive and negative spin densities, respectively.

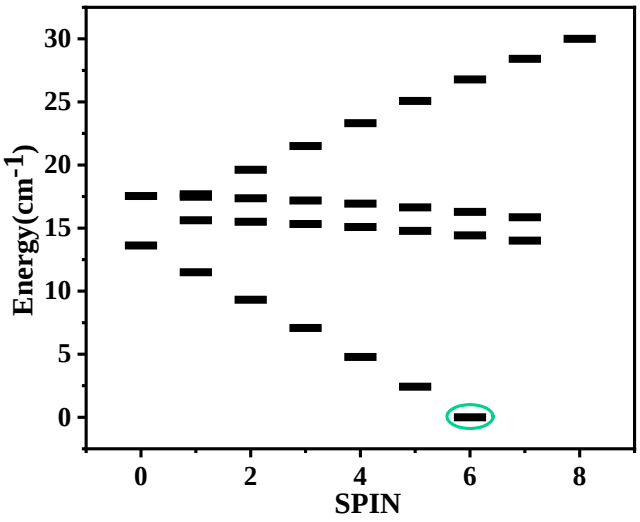


Figure S14. Eigenvalue plots for experimental *J* values and (spin ground state is highlighted) of **1-Gd**.

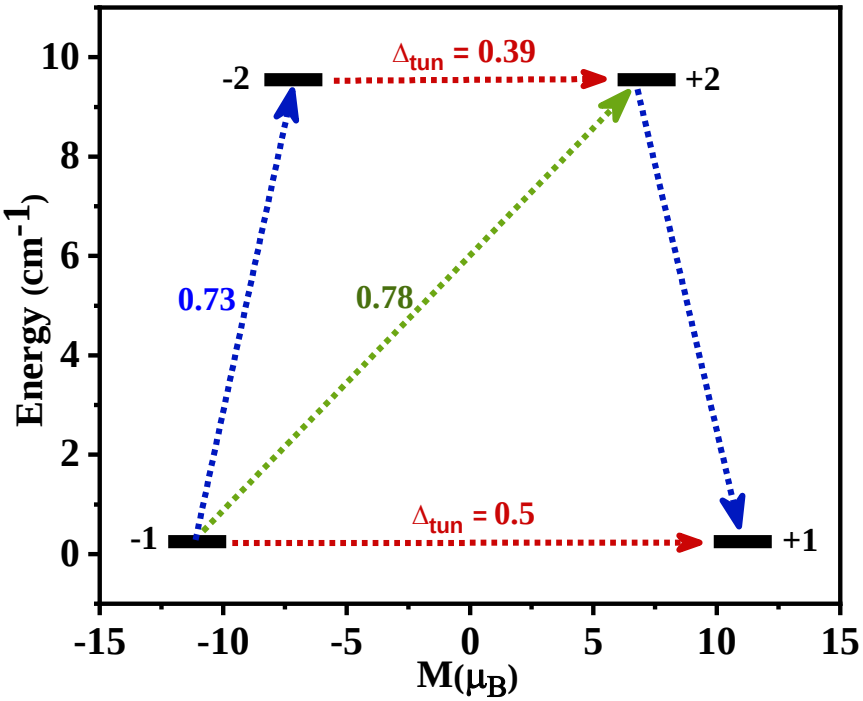


Figure S15. Low-lying exchange spectra in **5-Er**. The exchange states are placed on the diagram according to their magnetic moments (bold black lines). The red arrows show the tunneling transitions (energy splitting) within each doublet state, while the green/blue arrows show the possible pathway through Raman/Orbach relaxation. The numbers at the paths are averaged transition moments in μ_B, connecting the corresponding states.

Table S16. Lowest exchange doublets (cm^{-1}) corresponding to tunnel splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex **1-Gd**.

No.	E (cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.000019032540	1.9×10^{-5}	18.585177263
2	0.012044285975 0.012044359361	7.3×10^{-8}	19.980651662
3	0.013749552092 0.013788088144	3.8×10^{-5}	20.863306650
4	0.066269771854 0.066270543801	7.7×10^{-8}	16.394845994
5	0.168674592223 0.171797825668	3.1×10^{-3}	15.269016745
6	0.202870298367 0.209913251104	7.0×10^{-3}	13.705496420
7	0.218862739980 0.218863080584	3.4×10^{-7}	15.981879356
8	0.281269943729 0.282831732594	1.6×10^{-3}	6.467402546
9	0.298261685925 0.305806375277	7.5×10^{-3}	10.077380408
10	0.311628784265 0.359267702205	4.7×10^{-2}	2.681905829

Table S17. Lowest exchange doublets (cm^{-1}) corresponding to tunnel splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex **2-Tb**.

No.	E (cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.000690517011	6.9×10^{-4}	30.368240002
2	3.773842615291 3.930814763636	0.16	0.027354452
3	4.371983418247 4.757464432473	0.39	0.124379649
4	4.766529006760 4.774464158850	0.008	0.146593215
5	5.144080829265 5.522673338076	0.38	21.408291877

Table S18. Lowest exchange doublets (cm^{-1}) corresponding to tunnel splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex **3-Dy**.

No.	E (cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.000006466542	6.4×10^{-6}	36.381731581
2	8.806919717718 8.806933669572	1.4×10^{-5}	0.016978202
3	9.586874638924 9.586876683484	2.0×10^{-6}	0.527671756
4	9.622539224663 9.622562373023	2.3×10^{-5}	0.537637839
5	10.400766226913 10.400931142370	1.6×10^{-4}	0.023344408
6	10.544101824540 10.544219709625	1.2×10^{-4}	39.513987201
7	10.599492843448 10.599557179314	6.4×10^{-5}	39.515238607
8	21.178392940132 21.178394139949	1.2×10^{-6}	42.671756927
9	102.653331765777 102.654021990019	6.9×10^{-4}	33.076407425
10	102.813239961505 102.813662797930	4.2×10^{-4}	33.072431433
11	108.979294485172 108.980179275008	8.8×10^{-4}	0.160042958
12	109.003359378156 109.003660900997	3.0×10^{-4}	0.171595072
13	111.387536414215 111.391418925327	3.9×10^{-3}	0.234613262

Table S19. Lowest exchange doublets (cm^{-1}) corresponding to tunnel splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex **4-Ho**.

No.	E (cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.011509029799	0.01	34.058542909
2	6.761679219802 7.028334712146	0.27	0.269683613
3	7.674908983753 8.455229405606	0.78	0.027835752
4	8.467030005322 8.509972196211	0.04	0.075825931
5	8.619895727630 9.267380459104	0.65	0.033809126

Table S20. Lowest exchange doublets (cm^{-1}) corresponding to tunnel splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex **5-Er**.

No.	E (cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.502856082568	0.50	22.095849779
2	9.351731456867 9.746696671669	0.39	14.322550904
3	10.731907245972 10.829865495745	0.09	0.004823948
4	10.999737544880 11.014823506218	0.02	5.078034351
5	11.036936892738 11.310723931633	0.27	6.190436885

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