Electronic Supporting Information

In situ Hydrolysis of a Carbophosphazene Ligand Leads to One-Dimensional Lanthanide Coordination Polymers. Synthesis, Structure and Dynamic Magnetic Studies.

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Scheme S1. General synthetic route adopted to isolate the tetrameric Cu^{II} ensemble. [1]

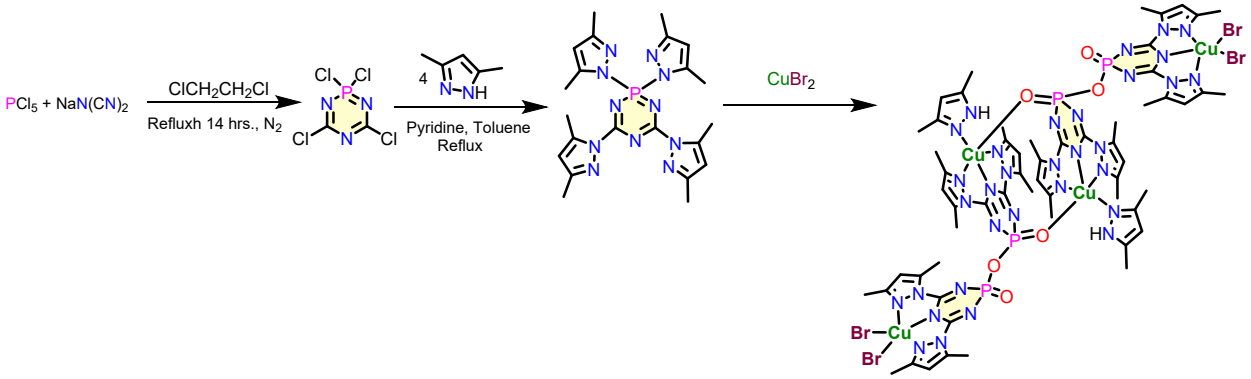

Table S1. Crystallographic and hydrogen bond parameters for **1-4**

	1	**2**	**3**	**4**
Formula	$C_{14}H_{40}ClGdN_{12}O_{14}P_2$	$C_{15}H_{42}ClN_{12}O_{14}P_2Tb$	$C_{15}H_{42}ClDyN_{12}O_{14}P_2$	$C_{15}H_{42}ClErN_{12}O_{14}P_2$
Size [mm]	0.220 × 0.210 × 0.190	0.240 × 0.200 × 0.140	0.240 × 0.210 × 0.190	0.260 × 0.210 × 0.170
System	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$
a [Å]	12.9642(6)	12.9417(3)	12.9515(4)	12.9252(3)
b [Å]	13.9446(8)	13.8877(4)	13.8286(5)	13.7997(3)
c [Å]	18.4472(12)	18.2069(5)	18.1752(7)	18.2660(5))
α [°]	90	90	90	90
β [°]	96.725(5)	97.507(2)	97.685(3)	97.052(2)
γ [°]	90	90	90	90
V [Å^3]	3312.0(3)	3244.29(15)	3226.0(2)	3233.35(14)
Z	4	4	4	4
ρ_{calcd}[g/cm^{-3}]	1.715	1.783	1.801	1.806
$2\Theta_{max}$	49.998	59.226	52.738	57.838
Radiation	Mo K_α	Mo K_α	MoKα	Mo K_α
λ [Å]	0.71073	0.71073	0.71073	0.71073
T [K]	293(2)	293(2)	293(2)	293(2)
Reflns	33646	41564	38110	29965
Ind. Reflns	5770	7739	6581	7610
reflns with *I>2σ(I)*	16297	5617	5209	5857
R1	0.0485	0.0351	0.0591	0.0404
wR2	0.1173	0.0801	0.1624	0.1055

Hydrogen-bond geometry (Å, º) for **1-4**

Complex	**Interaction**	**Type**	**dD–H (Å)**	**H···A (Å)**	**dD···A (Å)**	**<DHA (°)**
1	N1–H1·····O5	Intra	0.86	2.21	2.978(7)	148
	O1–H1A·····Cl1		0.85	2.18	2.990(7)	160
	O1W–H1WB·····O3	Intra	0.93	1.93	2.725(7)	143
	N7–H7·····O3	Intra	0.86	2.16	2.860(7)	138
2	O1–H1·····Cl1		0.87	2.21	3.031(3)	157
	O1W–H1WA·····O3	Intra	0.85	1.84	2.685(3)	169
	N1–H1A·····O5	Intra	0.86	2.17	2.932(4)	148
	O1W–H1WB·····O12		0.95	1.76	2.699(4)	169
	N8–H8·····O3	Intra	0.86	2.14	2.839(4	137
	O12–H12·····Cl1		0.82	2.17	2.976(4)	166
	O14–H14·····N3		0.82	2.02	2.817(5)	165
3	O1–H1·····Cl1		0.82	2.28	3.024(7)	150
	O1W–H1WA·····O5	Intra	0.85	1.90	2.695(9)	155
	O1S–H1S·····N8		0.84	2.00	2.800(11)	160
	N3–H3·····O5	Intra	0.88	2.14	2.847(9)	137
	N6–H6·····O3	Intra	0.88	2.14	2.928(9)	148
4	O1–H1·····Cl1		0.82	2.27	3.037(3)	156
	O1W–H1WA·····O3	Intra	0.85	1.84	2.677(4)	169
	O1W–H1WB·····O14		0.85	1.89	2.712(5)	163
	N3–H3·····O3S		0.86	2.03	2.824(5)	153
	O3S–H3S·····Cl1		0.82	2.27	3.094(4)	179
	N6–H6·····O3	Intra	0.86	2.16	2.850(5)	137
	O14–H14·····Cl1		0.82	2.15	2.961(6)	170

Table S2. Continuous Shape Measurement (CShM) analysis for **1**-**4**

	1	**2**	**3**	**4**
EP-9	36.221	35.843	35.691	36.116
OPY-9	21.303	21.154	21.196	21.313
HBPY-9	16.678	17.076	17.002	17.303
JTC-9	16.065	15.819	15.671	15.975
JCCU-9	11.538	11.072	11.014	10.942
CCU-9	9.696	9.503	9.422	9.821
JCSAPR-9	3.108	2.407	2.401	2.281
CSAPR-9	**2.252**	1.742	1.719	1.648
JTCTPR-9	4.049	3.804	3.825	3.487
TCTPR-9	2.701	2.512	2.470	2.334
JTDIC-9	12.941	12.814	12.700	12.797
HH-9	8.351	8.760	8.695	9.135
MFF-9	**1.655**	**1.357**	**1.339**	**1.315**

EP-9, D_{9h} Enneagon, **OPY-9**C_{8v} Octagonal pyramid, HBPY-D_{7h}Heptagonal bipyramid, **JTC-9**C_{3v} Johnson triangular cupola J3, **JCCU-9**C_{4v} Capped cube J8, **CCU-9**C_{4v} Spherical-relaxed capped cube, **JCSAPR-9**C_{4v} Capped square antiprism J10, **CSAPR-9**C_{4v} Spherical capped square antiprism, **JTCTPR-9**D_{3h} Tricapped trigonal prism J51, **TCTPR-9**D_{3h} Spherical tricapped trigonal prism, **JTDIC-9**C_{3v} Tridiminished icosahedron J63, **HH-9** C_{2v} Hula-hoop**MFF-9**C_s Muffin

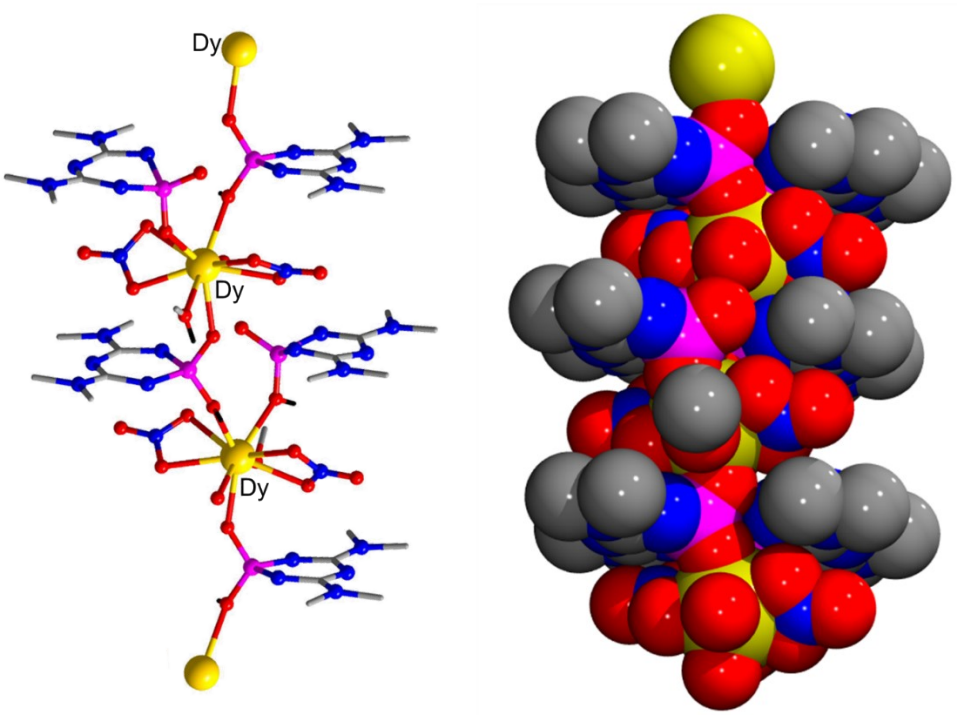

Figure S1. 1D extended structure of representative coordination polymer **3** and its space filling diagram.

Table S3. Selected bond lengths and bond angle parameters for **1-4**.

Bond Length (Å)	**1**	**2**	**3**	**4**
Ln1-O1	2.462(5)	2.448(3)	2.437(7)	2.405(3)
Ln1-O2	2.327(5)	2.296(2)	2.291(7)	2.264(2)
Ln1-O4	2.384(5)	2.370(2)	2.357(7)	2.338(2)
Ln1-O5	2.446(4)	2.432(2)	2.421(6)	2.417(2)
Ln1-O6	2.532(6)	2.546(3)	2.426(7)	2.401(3)
Ln1-O7	2.487(5)	2.439(3)	2.534(8)	2.522(3)
Ln1-O9	2.488(5)	2.474(3)	2.468(7)	2.482(3)
Ln1-O10	2.542(6)	2.515(3)	2.494(7)	2.438(3)
Ln1-O1W	2.420(5)	2.370(2)	2.358(7)	2.331(3)
Bond Angle (˚)				
P1-O2-Ln1	139.1(3)	138.07(14)	138.1(3)	137.97(15)
P2-O4-Ln1	155.8(3)	154.37(13)	154.7(4)	154.49(17)
P2-O5-Ln2	140.7(3)	139.41(13)	139.0(3)	139.63(16)
Intermetallic Distance (Å)				
Ln1-Ln2	7.172	7.136	7.111	7.094

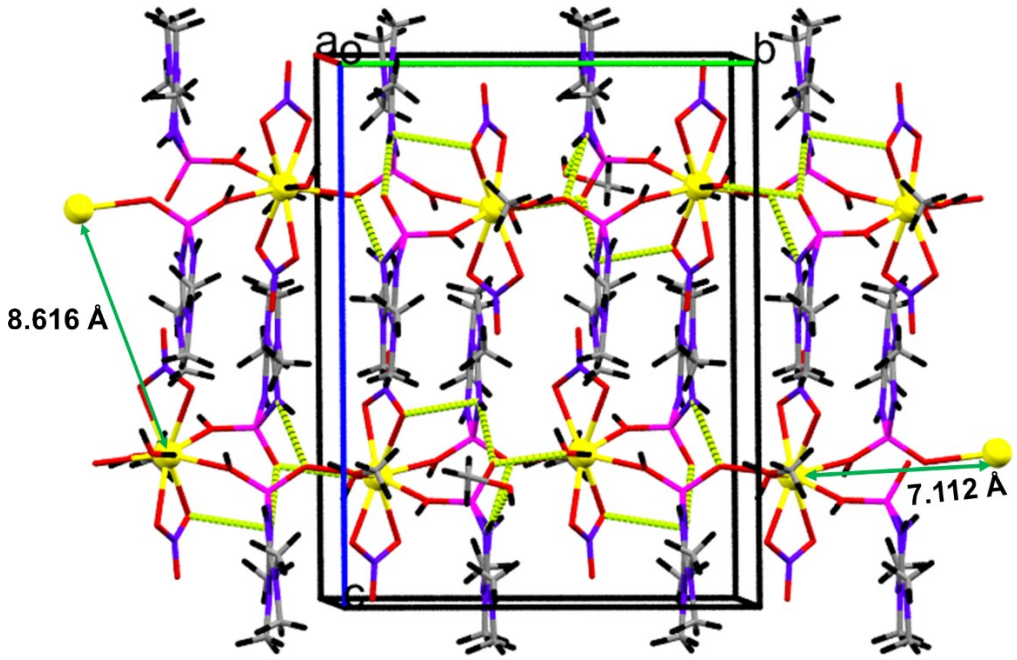

Figure S2. Packing structure of representative complex **3** along a-axis. The fragmented light green bonds represent hydrogen bonding.

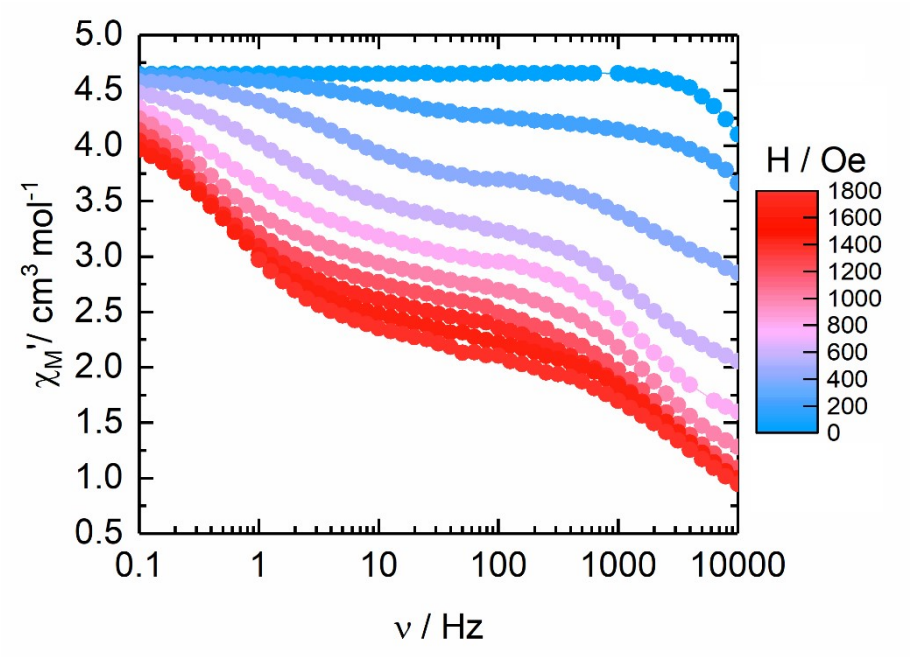

Figure S3. Field dependence of the in-phase component (χ_M') of the magnetic susceptibility at 2 K for **3**

Extended Debye model used for two relaxation contributions (Eq. S1).

$$\chi_M{}' = \chi_S + (\chi_T - \chi_S) \left\{ \frac{\beta\left[1 + (\omega\tau_1)^{1-\alpha_1} \sin\left(\frac{\pi}{2}\alpha_1\right)\right]}{1 + 2(\omega\tau_1)^{1-\alpha_1} \sin\left(\frac{\pi}{2}\alpha_1\right) + (\omega\tau_1)^{2(1-\alpha_1)}} + \frac{(1-\beta)\left[1 + (\omega\tau_2)^{1-\alpha_2} \sin\left(\frac{\pi}{2}\alpha_2\right)\right]}{1 + 2(\omega\tau_2)^{1-\alpha_2} \sin\left(\frac{\pi}{2}\alpha_2\right) + (\omega\tau_2)^{2(1-\alpha_2)}} \right\}$$

$$\chi_M{}'' = (\chi_T - \chi_S) \left\{ \frac{\beta\left[(\omega\tau_1)^{1-\alpha_1} \cos\left(\frac{\pi}{2}\alpha_1\right)\right]}{1 + 2(\omega\tau_1)^{1-\alpha_1} \sin\left(\frac{\pi}{2}\alpha_1\right) + (\omega\tau_1)^{2(1-\alpha_1)}} + \frac{(1-\beta)\left[(\omega\tau_2)^{1-\alpha_2} \cos\left(\frac{\pi}{2}\alpha_2\right)\right]}{1 + 2(\omega\tau_2)^{1-\alpha_2} \sin\left(\frac{\pi}{2}\alpha_2\right) + (\omega\tau_2)^{2(1-\alpha_2)}} \right\}$$

With χ_T the isothermal susceptibility, χ_S the adiabatic susceptibility, τ the relaxation time and α an empiric parameter which describe the distribution of the relaxation time. For SMM with only one relaxing object α is close to zero. The extended Debye model was applied to fit simultaneously the experimental variations of χ_M' and χ_M'' with the frequency ν of the oscillating field ($\omega = 2\pi\nu$). The best fitted parameters τ_1, α_1, χ_{1T}, χ_{1S}, τ_2, α_2, χ_{2T} and χ_{2S} are listed in Tables S4 and S11 with the coefficient of determination R^2. τ_1, α_1, χ_{1T}, χ_{1S} are parameters for the high frequency contribution while τ_2, α_2, χ_{2T} and χ_{2S} are for the low frequency contribution.

Extended Debye model used for a single relaxation contribution (Eq. S2).

$$\chi_M{}' = \chi_S + (\chi_T - \chi_S) \frac{1 + (\omega\tau)^{1-\alpha} \sin\left(\alpha\frac{\pi}{2}\right)}{1 + 2(\omega\tau)^{1-\alpha} \sin\left(\alpha\frac{\pi}{2}\right) + (\omega\tau)^{2-2\alpha}}$$

$$\chi_M{}'' = (\chi_T - \chi_S) \frac{(\omega\tau)^{1-\alpha} \cos\left(\alpha\frac{\pi}{2}\right)}{1 + 2(\omega\tau)^{1-\alpha} \sin\left(\alpha\frac{\pi}{2}\right) + (\omega\tau)^{2-2\alpha}}$$

With χ_T the isothermal susceptibility, χ_S the adiabatic susceptibility, τ the relaxation time and α an empiric parameter which describe the distribution of the relaxation time. For SMM with only one relaxation time, α is close to zero. The extended Debye model was applied to fit simultaneously the experimental variations of χ_M' and χ_M'' with the frequency ν of the oscillating field ($\omega = 2\pi\nu$). Typically, only the temperatures for which a maximum on the χ'' vs. f curves, have been considered. The best fitted parameters τ, α, χ_T, χ_S are listed in Tables S3, S5-S6 and S8-S10 with the coefficient of determination R^2.

Table S4. Best fitted parameters ($\chi_{T,1}$, $\chi_{S,1}$, τ_1, α_1, ($\chi_{T,2}$, $\chi_{S,2}$, τ_2 and α_2) with the extended Debye model (Eq. S1) for compound **3** at 2 K in the magnetic field range 200-1600 Oe.

H / Oe	$\chi_{T,1}$ / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	τ_1 / s	α_1	$\chi_{T,2}$ / cm^3 mol^{-1}	α_2	τ_2 / s	R^2
200	3.37825	2.95761	0.01921	0.33654	4.22332	0.26027	1.10158E-5	0.99998
400	3.66654	2.73227	0.04393	0.30613	3.68578	0.23894	7.55752E-5	0.99999
600	3.45039	2.02836	0.11814	0.39292	3.23137	0.21123	1.11113E-4	0.99985
800	3.6194	1.63867	0.34566	0.47904	2.90569	0.17314	1.11327E-4	0.99977
1000	3.13061	1.11681	0.41693	0.40683	2.75402	0.26921	9.45152E-5	0.99951
1200	2.75637	0.73791	0.45268	0.36031	2.63551	0.35437	7.9492E-5	0.99961
1400	2.54155	0.59352	0.42027	0.31904	2.54596	0.43427	6.80281E-5	0.99973
1600	2.58741	0.60431	0.33618	0.31579	2.38037	0.43	5.58487E-5	0.99976

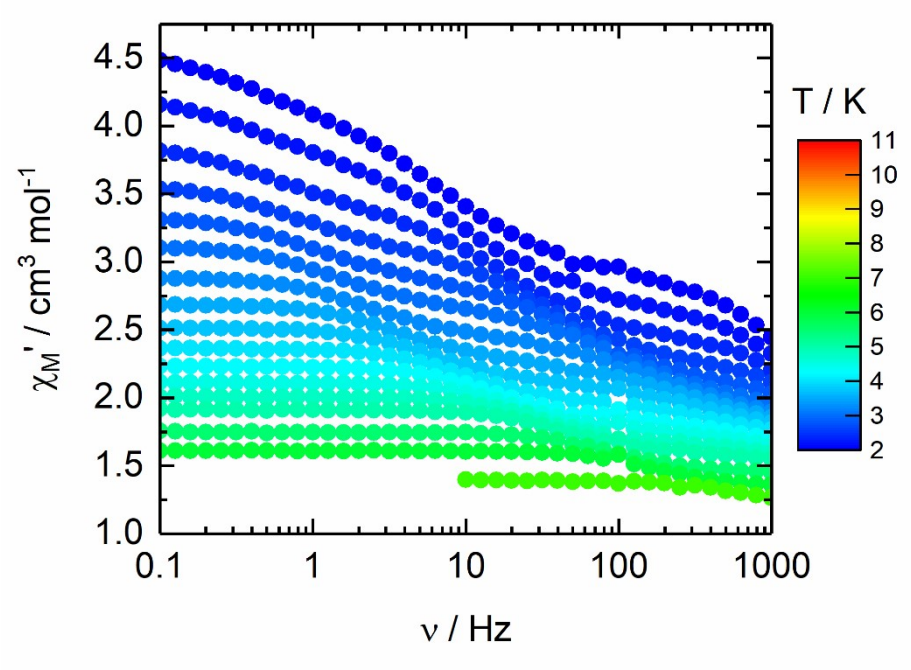

Figure S4. Thermal dependence of the in-phase component (χ_M') of the magnetic susceptibility under an applied dc field of 800 Oe in the 0.1-1000 Hz frequency range for **3**.

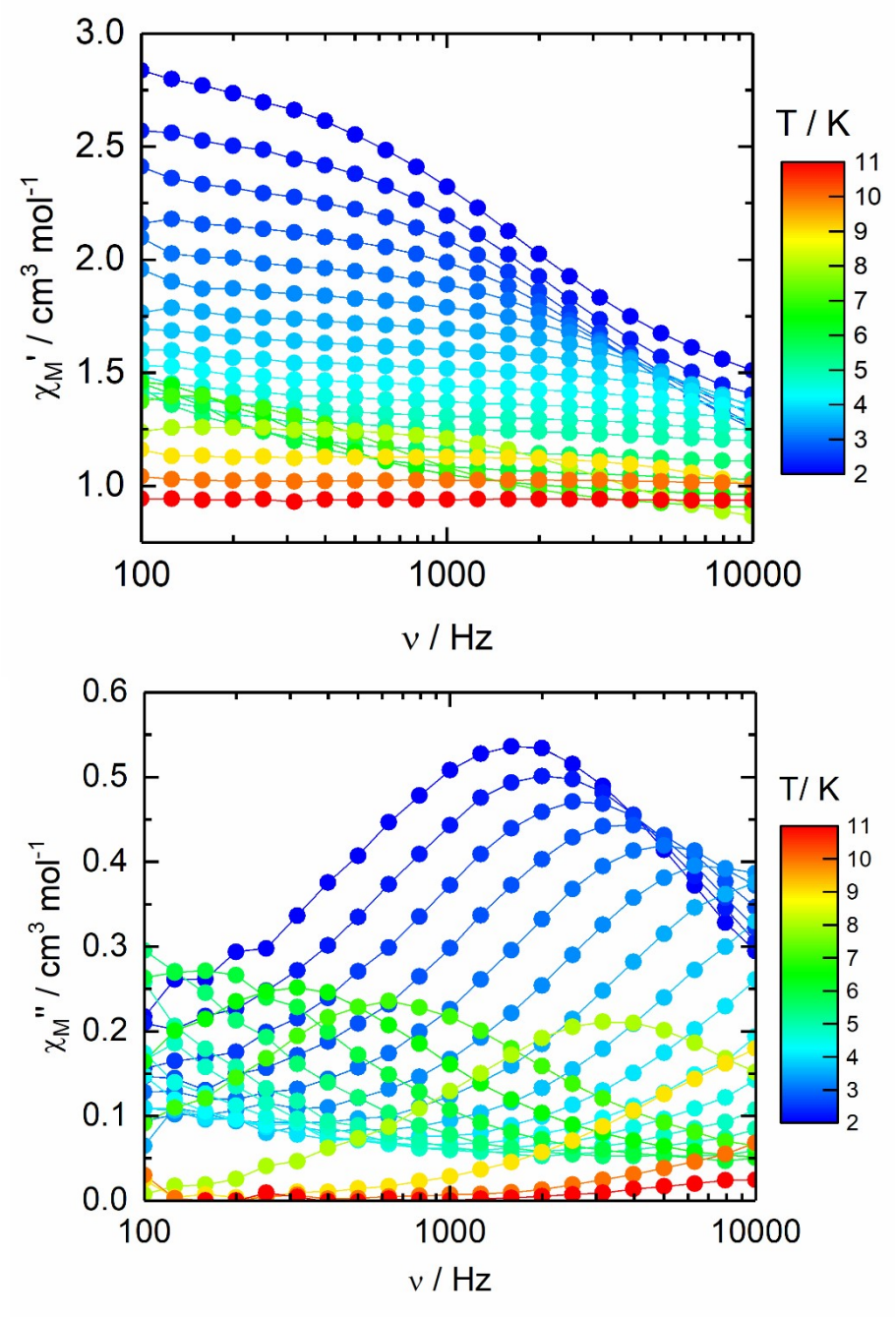

Figure S5. Thermal dependence of the in-phase (χ_M') and out-of-phase (χ_M'') component of the magnetic susceptibility under an applied dc field of 800 Oe in the 100-10000 Hz frequency range for **3**.

Table S5. Best fitted parameters ($\chi_{T,1}$, $\chi_{S,1}$, τ_1, α_1, ($\chi_{T,2}$, $\chi_{S,2}$, τ_2 and α_2) with the extended Debye model (Eq. S1) for compound **3** at 2 K in the temperature range of 2-7 K for LF contribution and 2-4.5 K for the MF.

T / K	$\chi_{T,1}$ / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	τ_1 / s	α_1	$\chi_{T,2}$ / cm^3 mol^{-1}	α_2	τ_2 / s	R^2
2	2.87334	2.6969	0.64708	2.65722E-12	4.4083	0.38874	0.02419	0.99999
2.2	2.73898	2.48942	0.56009	0.07503	4.0081	0.38507	0.01624	0.99996
2.4	2.60518	2.27802	0.36345	0.10316	3.54299	0.34962	0.00838	0.99998
2.6	2.54714	2.13409	0.23596	0.14882	3.16489	0.28245	0.00444	0.99999
2.8	2.40253	1.98506	0.16235	0.12556	2.91091	0.26481	0.00249	0.99999
3	2.21623	1.82167	0.11976	0.11288	2.72297	0.30907	0.00134	0.99998
3.25	1.9127	1.59109	0.07931	0.04831	2.56744	0.43534	5.85658E-4	0.99998
3.5	1.88318	1.55491	0.04692	0.05049	2.36207	0.43296	3.67898E-4	0.99999
3.75	1.89377	1.56785	0.0289	0.04819	2.19084	0.40776	2.97115E-4	0.99999
4	1.93511	1.59501	0.01778	0.06575	2.02565	0.33973	2.82937E-4	0.99999
4.25	1.96669	1.61946	0.01129	0.06812	1.88442	0.20395	3.1418E-4	0.99995
4.5	1.74884	1.49086	0.00836	0	1.85644	0.44932	2.05141E-4	0.99998
4.75	2.64768	2.27598	0.00427	0.11581				0.99995
5	2.08201	1.7452	0.00322	0.10163				0.99995
5.5	2.19791	1.89936	0.00173	0.07417				0.99998
6	1.9685	1.71861	9.45639E-4	0.01537				0.99995
7	1.22032	1.04729	2.67959E-4	0.05541				0.99994

Table S6. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model (Eq. S2) for **3** at 800 Oe in the temperature range 2-3.5 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
2	2.94814	1.2773	0.28189	9.99227E-5	0.99976
2.25	2.66193	1.13817	0.27163	7.6471E-5	0.99959
2.5	2.43364	1.02706	0.26206	5.81079E-5	0.9996
2.75	2.22317	0.9181	0.25322	4.09385E-5	0.99941
3	2.07516	0.75307	0.28964	2.56523E-5	0.99911
3.25	1.87405	0.81613	0.1803	2.01954E-5	0.99994
3.5	1.75297	0.67576	0.20326	1.18228E-5	0.99993

Table S7. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model (Eq. S2) for **3** at 800 Oe in the temperature range 6-9 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
6	1.7731	1.02586	0.19471	0.00128	0.99964
6.5	1.57341	0.95614	0.13808	5.30576E-4	0.99975
7	1.43929	0.89099	0.11282	2.33685E-4	0.99975
8	1.25997	0.78884	0.06655	4.58688E-5	0.99991
9	1.1325	0.7028	0.05777	1.00361E-5	0.99987

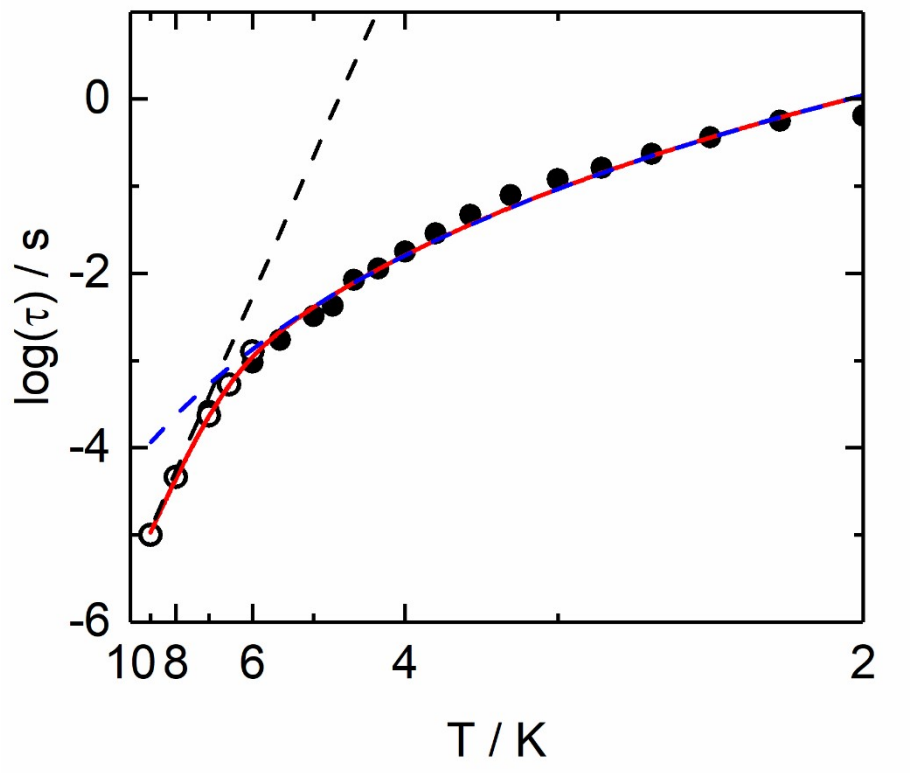

Figure S6. Thermal dependence of the LF relaxation time for **3** from 2 to 9 K at 800 Oe is depicted in full black dots from squid measurements and in open black circles from ppms measurements. The red line depicts the best fit with parameters given in the main text. Dashed black and blue lines are the separated Orbach and Raman processes, respectively.

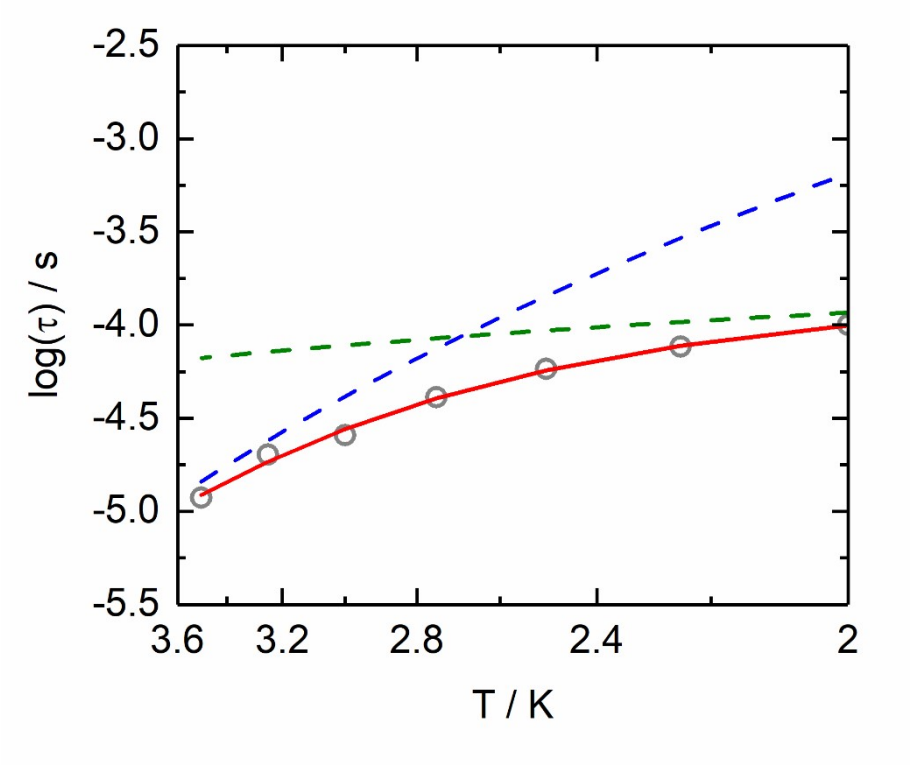

Figure S7. Thermal dependence of the HF relaxation time for **3** from 2 to 3.5 K at 800 Oe. The red line depicts the best fit with parameters given in the main text. Dashed blue and green lines are the separated Raman and direct processes, respectively.

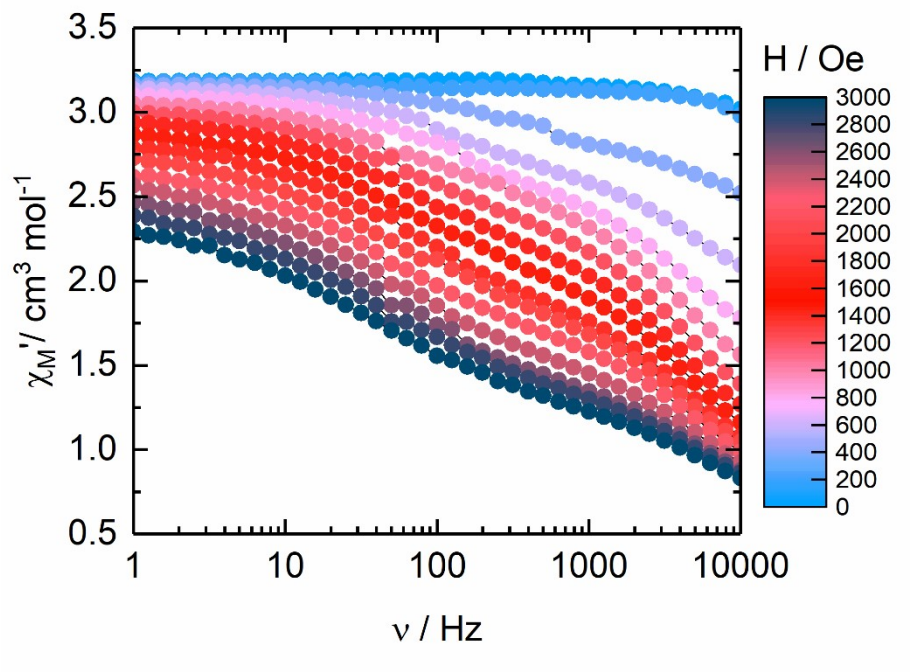

Figure S8. Field dependence of the in-phase component (χ_M') of the magnetic susceptibility at 2 K for **4**.

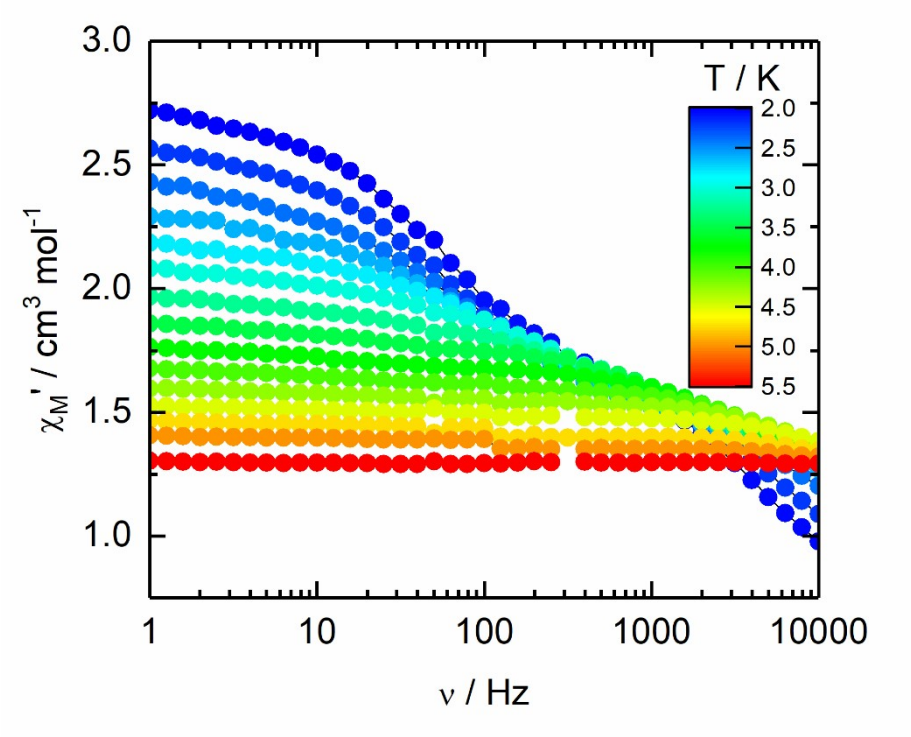

Figure S9. Thermal dependence of the in-phase component (χ_M') of the magnetic susceptibility under an applied dc field of 2200 Oe in the 1-10000 Hz frequency range for **4**.

Table S8. Best fitted parameters ($\chi_{T,1}$, $\chi_{S,1}$, τ_1, α_1, ($\chi_{T,2}$, $\chi_{S,2}$, τ_2 and α_2) with the extended Debye model (Eq. S1) for compound **4** at 2 K in the temperature range of 2-4.5 K for LF contribution and 2-2.4 K for the HF.

T / K	$\chi_{T,1}$ / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	τ_1 / s	α_1	$\chi_{T,2}$ / cm^3 mol^{-1}	α_2	τ_2 / s	R^2
2	1.56883	0.44954	0.00323	0.29073	1.624	0.26281	2.18237E-5	0.99983
2.2	1.57185	0.49511	0.00292	0.32998	1.51913	0.22898	1.35073E-5	0.99987
2.4	1.36414	0.37956	0.0023	0.36322	1.46308	0.24293	6.47715E-6	0.99992
2.6	1.56296	0.61061	0.00144	0.41102	1.37083	0.13301		0.99991
2.8	1.72703	0.92397	8.91798E-4	0.40427	1.3842	0.08962		0.99996
3	1.89328	1.04835	3.53743E-4	0.5027	1.25791	0		0.99991
3.25	2.8482	1.60646	1.51815E-4	0.53005	0.85936	0.52366		0.99997
3.5	2.96376	2.34791	5.8825E-5	0.35681	0	1		0.99993
3.75	2.82506	2.25706	2.9618E-5	0.31365	0	1		0.99998
4	2.69354	2.15882	1.57407E-5	0.27608	0	1		0.99997
4.25	2.63594	2.17602	1.04962E-5	0.21941	0	1		0.99996

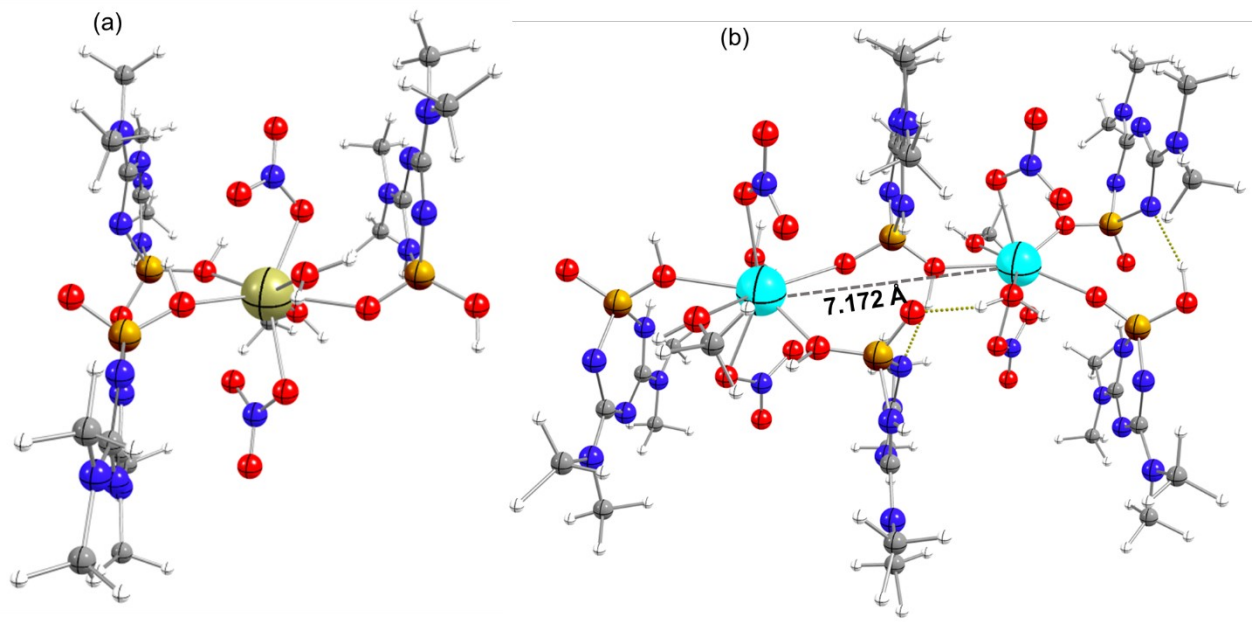

Figure S10. (a) Mononuclear model for complexes **2-4** that have been used in the *Ab-initio* calculations; (b) Dimeric model structure obtained from the crystal structure of complex **1** to compute the intermolecular exchange interaction employing BS-DFT.

Table S9. Basis functions used in *ab initio* CASSCF calculations using MOLCAS 8.2 code.

Atoms	ANO-RCC basis functions
H	2s
C	3s2p
N, P, O	3s2p1d
Dy, Tb, Er	8s7p5d3f2g1h

Table S10. RASSI-SO computed low-lying 7 septet (in red) and 140 quintet states (in black) along with spin-orbit coupled states (in blue, Kramer pseudo doublets) for Tb centre in **2**. All the values are reported here in cm^{-1}.

Tb			
0.0	35285.8	45903.7	75193.6
90.6	35368.7	45930.1	75212.4
145.6	35375.6	45960.4	75224.4
169.5	35382.1	45969.9	75244.3
383.8	35406.6	45992.4	75274.2
560.5	35434.9	46000.2	75293.4
565.3	35442.5	50103.1	75313.3
25660.2	35473.6	50114.4	75351.5
25674.3	35532.0	50169.3	77503.4
25715.1	35536.8	50187.3	77570.1
25788.2	40221.9	50218.9	77629.7
25798.2	40262.5	50222.7	77686.7
29220.3	40267.2	50280.9	77714.7
29220.5	40307.3	50286.1	77742.3
29286.0	40347.1	51775.4	77778.8
29292.0	40377.4	51808.2	93188.6
29315.5	40393.9	51920.6	93209.2
29322.3	40410.3	51943.0	93384.1
29336.4	40439.3	52054.7	93395.5
29347.3	40451.5	54537.8	93526.9
29356.6	40459.1	54678.9	93625.1
29392.7	40459.6	54803.2	93629.0
29400.3	40503.3	57660.2	93816.4
29439.2	40519.0	57663.9	93821.0
29448.6	40529.2	57690.5	95220.5
29474.6	40565.4	57748.5	95243.3
29479.5	40588.1	57772.1	95438.2
29507.2	40627.7	57775.8	95574.2
29508.3	40689.4	57802.7	3389952726.9
30617.2	40707.8	57837.9	3389952726.9
30630.9	45728.8	57898.9	
30668.1	45730.0	57926.8	
30685.1	45806.8	57941.2	
30699.8	45816.8	68512.9	
30709.9	45818.0	75067.4	
30755.1	45824.8	75103.7	
30810.2	45840.7	75115.2	
30839.5	45855.5	75147.8	
35271.5	45877.5	75159.3	

SPIN-ORBIT STATES			
0.0	5857.0	31336.3	32524.4
0.3	5967.9	31344.9	32534.7
34.7	6110.6	31352.9	32543.6
35.7	23506.4	31369.0	32548.9
116.8	23507.0	31396.8	32556.6
121.4	23536.7	31449.5	32578.9
181.1	23541.7	31460.7	32579.7
197.8	23549.3	31597.5	32602.3
228.3	23571.8	31597.6	32605.7
296.1	23579.3	31668.0	32615.7
300.5	23611.7	31672.4	32618.7
363.7	23612.7	31675.8	32629.0
364.2	29449.8	31680.1	32631.2
2086.3	29455.5	31688.6	32684.9
2088.6	29459.4	31699.6	32688.0
2207.5	29461.5	31706.7	32728.7
2223.9	29464.4	31709.4	32739.8
2241.4	29469.0	31716.2	32748.6
2242.0	29474.7	31741.2	32755.6
2250.2	30089.2	31742.4	32757.1
2296.2	30089.3	31774.5	32771.7
2306.5	30153.1	31777.1	32781.6
2354.6	30153.9	31781.7	32790.7
2365.1	30170.2	31784.7	32796.8
3460.3	30172.1	31789.7	32827.6
3539.0	30189.5	31792.5	32841.8
3551.5	30191.3	31958.1	32879.2
3572.3	30205.3	31977.4	32884.5
3596.4	30207.4	31986.9	32903.0
3615.6	30211.6	32020.7	32907.1
3635.4	30216.0	32036.0	32922.8
3688.6	30222.9	32129.4	32934.0
3727.0	30256.9	32130.9	32943.9
4577.8	30259.5	32198.7	32949.3
4582.8	30294.7	32205.1	32957.0
4618.6	30296.0	32210.5	32967.7
4633.9	30322.8	32223.1	32971.9
4653.6	30323.1	32229.0	32988.0
4666.2	30339.9	32239.5	32991.0
4679.3	30339.9	32245.9	33013.5
5266.6	31190.9	32289.3	33039.5
5329.0	31192.2	32294.0	33050.4
5346.7	31268.5	32447.4	33068.0
5429.9	31271.9	32447.5	33073.8
5449.5	31322.0	32510.1	33112.9

5745.6	31328.6	32510.2	33125.1
33133.1	38322.8	41851.1	44412.7
33143.6	38331.0	41856.4	44428.1
33162.3	39043.8	41889.4	44441.3
33168.0	39045.8	41905.6	44443.7
33357.7	39074.5	41979.3	44471.9
33372.5	39077.0	41998.6	44486.0
33395.6	39085.6	42027.1	44494.5
33404.6	39087.4	42038.2	44506.4
33425.9	39100.2	42080.7	44508.1
33438.1	39109.9	42138.8	44528.2
33449.9	39116.0	42230.4	44536.6
34005.5	39125.0	42888.6	44540.8
34014.1	39127.4	42904.9	44558.1
34059.9	39541.2	42965.4	44561.7
34076.3	39543.9	42985.4	44577.3
34100.4	39554.3	43037.7	44582.6
34323.4	39557.5	43082.4	44590.5
34372.8	39568.7	43159.8	44602.6
34422.3	39570.0	43208.8	44610.0
35641.2	39581.1	43334.1	44615.8
36464.6	39587.5	43341.1	44622.4
36465.4	39595.5	43341.9	44624.6
36503.5	39963.1	43369.1	44639.7
36505.2	39976.6	43370.0	44643.5
36540.0	39988.4	43389.2	44649.3
36542.9	40023.1	43393.3	44663.5
36582.0	40043.6	43403.0	44852.8
36583.5	40060.7	43410.8	44864.0
36590.5	40081.2	43414.2	44894.4
36596.4	41694.5	43426.7	44895.6
36599.1	41696.2	43428.7	44919.8
36607.6	41709.1	43445.2	44939.1
36612.5	41714.9	43445.7	44943.3
36645.6	41727.9	43459.8	44960.3
36646.4	41733.1	43459.9	44962.6
38181.4	41742.5	43892.6	44976.5
38182.9	41754.5	43909.9	44977.7
38229.9	41768.5	43964.1	46847.3
38233.5	41784.1	44019.1	46853.1
38243.9	41788.9	44024.8	46877.2
38247.9	41792.8	44079.1	46891.2
38269.2	41795.2	44133.9	46891.2
38273.4	41812.0	44352.5	47706.4
38287.5	41813.7	44357.1	47706.4
38298.6	41832.7	44373.8	47742.3

38302.6	41841.8	44403.5	47743.5
47767.6	49178.0	50825.8	51200.7
47773.8	49189.0	50826.1	51208.7
47785.4	49205.4	50840.1	51212.6
47789.7	49206.6	50847.6	51213.4
47805.2	49209.3	50899.8	51229.4
47812.0	49211.2	50900.9	51230.7
47835.8	49212.4	50902.1	51236.6
47847.8	49219.5	50929.4	51239.4
47850.4	49231.3	50936.3	51241.3
47860.5	49237.3	50955.3	51246.8
47869.8	49239.7	50955.4	51250.5
47896.9	49240.8	50956.3	51255.9
47901.2	49244.9	50964.0	51257.9
47912.4	49248.1	50976.3	51259.3
47915.0	49254.6	50978.8	51267.6
48227.3	49255.3	50992.2	51272.2
48227.4	49258.3	51002.1	51274.5
48289.2	49259.5	51002.6	51278.3
48297.3	49272.6	51004.9	51281.4
48306.7	49281.1	51006.2	51285.4
48312.2	49298.5	51020.3	51291.5
48314.5	49300.6	51021.3	51293.4
48320.0	49308.9	51031.3	51294.7
48332.6	49339.2	51036.3	51296.0
48337.4	49343.9	51048.6	51319.3
48345.2	50513.1	51057.0	51321.0
48382.0	50513.2	51063.1	51423.2
48386.4	50577.7	51063.3	51423.5
49089.2	50586.5	51064.7	51478.6
49090.5	50604.2	51075.4	51509.3
49096.7	50608.8	51076.4	51569.7
49098.1	50620.1	51077.5	51598.8
49110.2	50633.6	51092.4	51631.3
49110.3	50636.2	51102.8	51667.0
49122.0	50639.3	51105.7	51681.8
49124.0	50666.9	51121.4	51686.4
49137.7	50678.1	51125.2	51824.6
49138.6	50680.1	51135.0	51859.5
49143.7	50703.4	51135.8	51865.9
49150.3	50710.9	51139.0	51871.3
49153.6	50725.9	51151.5	51889.8
49155.4	50726.6	51156.8	51904.2
49160.5	50752.9	51163.3	51914.4
49166.1	50753.5	51164.0	51939.3
49168.2	50779.4	51192.4	51954.4

49172.1	50781.0	51199.0	52042.3
52046.2	52920.1	53675.3	55191.5
52046.3	52928.1	53680.5	55268.0
52046.7	52930.3	53681.9	55395.6
52068.5	52941.9	53702.2	55536.5
52074.3	52942.7	53706.9	55594.9
52075.8	52947.4	53707.3	55609.1
52090.2	52957.8	53712.1	55609.3
52098.4	52970.0	53712.2	55628.2
52100.6	52970.5	53719.4	55711.2
52122.2	52994.9	53724.8	55720.9
52154.6	52995.8	53727.1	55726.6
52158.6	53006.4	53754.9	55730.2
52541.9	53021.0	53906.5	55743.8
52642.8	53025.4	53989.7	55752.3
52701.7	53026.8	54035.8	55759.1
52706.5	53047.6	54043.0	55761.5
52708.2	53048.0	54072.0	55782.5
52717.4	53134.1	54073.0	55783.8
52725.2	53342.1	54102.6	55858.3
52725.5	53362.7	54329.3	55862.2
52738.8	53364.1	54349.5	55931.9
52750.7	53371.7	54362.3	55937.8
52766.9	53390.9	54385.8	55955.0
52768.6	53402.1	54394.7	55961.7
52780.3	53417.8	54403.4	55972.2
52790.3	53427.0	54415.8	55982.4
52796.2	53462.0	54425.7	55984.1
52799.8	53469.6	54434.1	56013.5
52803.1	53479.6	54435.3	56018.0
52813.9	53484.9	54460.9	56197.4
52819.6	53492.8	54463.4	56346.0
52822.6	53503.4	54477.0	56681.3
52829.4	53524.4	54489.4	56721.3
52834.7	53528.3	54502.0	56959.9
52844.4	53544.6	54502.1	57513.4
52857.2	53572.0	54520.3	57539.6
52857.8	53572.1	54520.6	57555.4
52858.1	53580.8	54607.3	57573.8
52860.4	53629.2	54735.1	57631.5
52868.9	53629.9	54929.7	58425.4
52873.1	53630.0	55000.6	58425.5
52876.1	53641.6	55105.9	58427.1
52896.4	53652.4	55138.2	58427.8
52898.9	53653.2	55152.7	58449.0
52900.2	53653.8	55182.3	58462.7

52904.0	53670.3	55182.8	58469.4
58472.9	58703.4	59493.5	60421.4
58475.1	58705.8	59496.6	60722.0
58491.6	58727.1	59512.0	60753.8
58505.9	58729.3	59520.9	60776.8
58508.5	58734.0	59524.2	60799.4
58518.4	58734.6	59534.9	60811.7
58518.8	58747.7	59550.2	60969.0
58528.3	58779.0	59555.1	60986.6
58528.6	58779.1	59558.0	60992.6
58529.0	58813.2	59562.8	61014.4
58540.7	58814.9	59571.2	61021.1
58544.9	58817.3	59583.1	61027.4
58563.8	58820.0	59588.6	61033.5
58563.8	58828.2	59594.8	61037.8
58564.8	58834.8	59606.0	61090.7
58567.6	58834.8	59620.6	61097.5
58570.8	58854.5	59623.3	61127.9
58573.4	58872.7	59641.8	61133.8
58575.5	58887.2	59649.7	61144.2
58584.5	58898.3	59662.5	61169.0
58584.5	58899.5	59699.8	61183.7
58586.9	58907.9	59707.1	61210.1
58592.5	58911.3	59713.8	61221.7
58593.0	58915.1	59725.0	61400.1
58593.2	58926.1	59748.5	61479.6
58609.0	58930.0	59754.1	61487.6
58619.8	58930.4	59757.2	61513.1
58628.2	58951.4	59759.6	61513.6
58630.2	58951.6	59777.9	61525.9
58633.8	58960.6	59780.0	61526.6
58641.2	58995.7	59781.6	61531.1
58641.9	59014.4	59794.7	61531.4
58642.3	59018.1	59799.3	61532.1
58644.7	59025.9	59804.7	61545.6
58645.5	59028.7	59804.8	61556.8
58651.8	59038.0	59808.9	61565.9
58652.3	59046.7	60349.1	61566.0
58653.2	59048.9	60362.4	61577.7
58653.9	59078.0	60372.2	61578.3
58654.4	59084.8	60373.0	61583.3
58670.7	59127.9	60374.5	61586.3
58671.1	59128.9	60385.3	61595.9
58671.2	59159.5	60390.9	61601.4
58672.3	59164.8	60391.1	61602.0
58672.4	59176.6	60403.5	61602.3

58685.2	59195.5	60415.3	61603.9
61609.4	63527.9	65981.3	66902.3
61637.8	63584.0	66042.1	66917.2
61637.9	63585.5	66042.2	66938.4
61642.6	63831.4	66042.2	66972.4
61737.9	64729.1	66042.7	66981.6
61763.5	64769.8	66042.9	67023.5
61792.2	64804.1	66042.9	67032.9
61794.1	64843.6	66068.1	67244.9
61802.7	64843.6	66070.0	67247.2
61807.5	64890.2	66072.5	67283.9
61812.2	64890.7	66101.5	67294.7
61819.1	64921.4	66107.8	67338.1
62218.1	64927.4	66144.9	67348.0
62219.2	64929.5	66148.9	67370.3
62254.0	64929.9	66149.8	67372.8
62256.7	64966.2	66153.5	67387.5
62266.0	64978.0	66172.3	67414.6
62271.1	65007.7	66185.4	67415.0
62273.1	65008.9	66186.8	67423.5
62283.4	65020.5	66197.1	67424.9
62284.1	65055.3	66197.1	67452.7
62288.3	65059.9	66197.8	67452.9
62302.1	65098.3	66215.8	67473.0
62309.4	65099.1	66216.0	67473.5
62310.0	65439.4	66221.8	67479.6
62383.1	65444.4	66225.4	67481.0
62383.6	65457.5	66236.6	67501.7
63133.0	65477.5	66245.3	67506.1
63224.1	65493.9	66245.5	67517.7
63287.6	65495.9	66270.3	67520.6
63361.8	65523.7	66292.6	67532.8
63391.4	65535.5	66411.6	67538.3
63392.2	65552.2	66447.7	67546.5
63409.3	65566.0	66483.4	67552.8
63437.8	65576.8	66530.9	67556.5
63467.6	65608.5	66540.0	67563.1
63477.0	65632.1	66540.9	67565.3
63491.0	65642.9	66559.9	67594.7
63491.9	65685.5	66760.4	67595.5
63501.5	65698.8	66760.6	68019.3
63509.6	65719.1	66822.3	68020.0
63513.0	65725.2	66824.2	68059.1
63514.2	65965.3	66855.5	68059.5
63516.1	65976.4	66856.4	68090.8
63523.7	65977.0	66860.1	68091.1

63527.6	65981.3	66876.0	68123.2
68136.2	69073.4	79981.1	81580.9
68139.4	69077.9	79991.6	81602.3
68170.1	69142.8	80000.3	81632.0
68170.8	69143.9	80004.4	81644.8
68205.6	73544.5	80037.1	81696.3
68207.4	73546.7	80037.5	81700.5
68238.9	73560.2	80054.3	82092.7
68239.0	73572.4	80055.0	82095.7
68266.2	73574.7	80535.7	82161.9
68267.7	77645.5	80537.9	82177.0
68393.2	77654.9	80559.3	82187.4
68399.9	77678.4	80560.7	82220.0
68427.8	77704.3	80574.2	82237.2
68476.3	77719.2	80578.7	82270.8
68501.2	77729.4	80596.6	82277.7
68546.0	77740.1	80598.0	82325.0
68556.9	77760.3	80598.8	82325.7
68611.2	77765.2	80617.3	95231.8
68621.9	77780.0	80647.4	95264.4
68667.7	77797.9	80653.2	95415.7
68670.1	77809.9	80656.6	95448.6
68683.8	77825.8	80665.3	95569.0
68713.5	77847.0	80747.4	95602.2
68716.0	77851.2	80771.2	95714.8
68736.1	77862.2	80789.4	95947.9
68751.8	77862.6	80802.3	95971.5
68774.4	79013.7	80813.6	96108.0
68787.9	79013.9	80942.4	96148.5
68810.9	79038.9	80960.1	96158.5
68820.4	79041.0	80976.6	96183.2
68826.4	79056.4	80996.6	96289.2
68853.6	79063.8	81004.8	96303.4
68854.0	79070.1	81017.6	96734.7
68876.6	79082.1	81023.1	96736.0
68887.6	79083.4	81032.6	96830.3
68917.9	79100.5	81035.6	96877.6
68918.8	79101.0	81065.3	96931.1
68962.9	79117.5	81097.4	96949.3
68964.6	79117.6	81123.4	96977.3
69011.8	79128.2	81127.6	96994.3
69015.8	79128.2	81157.3	97083.0
69029.2	79933.6	81167.6	97131.8
69032.0	79935.3	81190.4	97575.9
69053.9	79947.2	81525.7	97583.0
69055.5	79950.7	81531.2	97681.8

69065.9	79972.0	81550.5	97721.2
97748.3			
97760.6			
97807.9			
97838.4			
97869.2			
97887.1			
97912.8			
97947.0			
97974.3			
98041.6			
98050.5			
98453.1			
98453.7			
98601.8			
98602.5			
98723.2			
98727.5			
98778.6			
98827.4			
98838.3			
98945.0			
98946.1			
99052.9			
99067.6			
99229.0			
99233.5			
99331.8			
99375.2			
100203.7			
100794.2			
101321.6			
101354.1			
101951.2			
101953.2			
102102.6			
102106.2			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			
3389956457.0			

3389956457.0

Table S11. RASSI-SO computed low-lying 21 spin-free sextet states and the spin-orbit coupled (Kramer doublets) for Dy centre in **3**. All the values are reported here in cm^{-1}. The values in red colour are for the spin-free sextet states while values in the blue colour are for the spin-orbit coupled states.

Dy		
SPIN-FREE STATES	**SPIN-ORBIT STATES**	
0.0	0.0	9765.5
9.1	152.1	9826.6
216.4	201.1	9912.3
231.7	235.2	11023.7
248.0	289.3	11142.9
305.3	325.5	11279.8
343.2	391.0	11845.4
438.1	485.9	11857.1
455.3	3077.2	11888.1
536.0	3139.7	11898.8
548.8	3175.6	11930.3
7654.4	3226.3	13624.6
7683.5	3269.8	13646.8
7719.7	3327.5	13676.2
7794.2	3403.6	13699.2
7813.9	5668.0	15030.8
7824.3	5733.1	15041.4
7846.1	5760.7	15066.5
34849.4	5822.1	16028.4
35038.2	5883.1	16044.6
35348.6	5954.8	16632.2
	7863.3	38855.0
	7915.3	38942.6
	7961.0	39009.0
	8042.7	39107.4
	8112.3	40248.7
	9599.3	40367.7
	9643.5	40560.4
	9654.5	41367.3
	9684.1	41422.3
	9697.3	
	9726.8	
	9749.9	

Table S12. RASSI-SO computed low-lying 35 quartets (in red) and 112 doublet states (in black) along with spin-orbit coupled states (in blue, Kramer doublets) for Er centre in **4**. All the values are reported here in cm^{-1}.

Er			
0.0	18115.4	33358.7	50340.6
53.2	18116.8	33400.3	50352.3
99.0	18146.0	33405.0	50379.2
150.7	18151.0	43876.5	50381.8
151.7	18154.5	43894.5	51439.7
206.0	18157.9	43933.5	51458.3
270.5	18165.7	43977.6	51599.4
286.3	18171.7	43999.4	51724.0
308.1	18181.8	44041.7	51746.4
330.5	18188.7	44045.7	60769.0
371.0	18191.8	44101.1	60800.5
433.0	23977.2	44104.4	60849.0
438.5	23977.9	44116.1	60873.5
18300.8	24054.3	44117.1	60893.4
18337.3	24062.0	44171.8	60913.5
18376.5	24069.4	44171.9	60939.3
18388.0	24100.1	44602.4	77207.1
18425.1	24112.6	44602.4	77216.6
18467.3	24162.9	44721.3	77294.6
18531.4	24165.9	44721.7	77320.9
18563.7	27020.8	44837.8	77350.9
28676.2	27021.0	44841.0	77399.9
28718.7	27106.5	44942.0	77411.2
28763.3	27106.7	44948.4	77496.8
28785.8	27181.1	44998.8	77500.8
28841.7	27184.1	45036.3	110456.9
28853.2	27235.8	45045.7	110512.0
28876.7	27244.5	45112.4	110717.1
28927.0	27270.4	45114.5	110821.3
28954.3	27277.0	45206.8	110901.0
46733.9	27285.5	45206.9	111088.0
46874.2	27351.4	45364.0	111096.1
47015.2	27351.6	45364.1	
47193.5	27494.3	50204.1	
47229.2	27494.3	50222.0	
	33115.5	50254.3	
	33118.0	50265.9	
	33152.6	50277.7	
	33228.9	50312.4	
	33321.2	50318.4	

SPIN-ORBIT STATES

0.0	27903.5	42199.7	61058.8
42.4	27937.6	49456.6	61115.5
96.4	27975.6	49479.0	61161.7
171.8	31987.6	49724.3	61170.1
180.3	32062.6	49809.0	61206.1
223.2	32139.0	49881.4	61241.3
252.3	32195.4	50013.0	61291.2
329.1	32232.3	50069.6	61306.6
6623.7	32253.4	50090.2	67055.9
6661.1	32319.4	50108.9	67212.3
6696.3	32470.2	50146.1	67304.0
6715.0	32790.0	50171.9	67357.2
6746.4	32829.3	50211.2	67396.3
6766.9	32855.7	50220.6	67435.1
6815.0	32872.1	50323.6	76155.1
10676.5	32881.9	50410.5	76196.2
10706.7	32925.2	50481.1	76227.4
10728.9	33266.4	50556.3	83199.8
10749.0	33296.7	50627.6	83280.6
10769.9	33310.7	50714.0	83328.1
10806.5	33329.2	50868.4	83415.6
13426.4	33985.0	51447.2	87135.5
13463.9	34010.9	51603.3	87228.6
13554.1	34015.8	51713.7	87287.6
13586.0	34028.6	51835.9	87351.1
13633.0	34041.9	51950.8	87443.5
19023.8	37535.7	52018.0	116542.5
19064.1	37621.7	52074.3	116816.2
19107.7	37686.1	52118.6	117048.4
19140.2	37749.4	52162.0	120963.9
19193.0	37767.0	52190.0	121212.4
22701.9	37836.7	52238.4	121401.8
22723.2	37850.3	54715.2	121603.4
22734.3	37961.5	54761.5	
22745.6	37987.1	56235.6	
22751.9	40444.1	56369.5	
22776.0	40671.4	56487.5	
23630.4	40701.3	56579.8	
23688.2	40736.3	56649.3	
25187.4	40767.1	56732.2	
25237.5	40797.9	56833.4	
25288.7	40830.8	56977.6	
25342.6	40866.0	58633.8	
27297.2	40943.0	58678.0	

27323.6	40991.0	58709.2
27358.5	41336.2	58733.7
27588.4	41368.2	58760.6
27689.3	41390.3	59392.6
27825.0	42051.8	61033.5
27840.1	42128.9	61058.8

Table S13. SINGLE_ANISO computed g-tensors, and the tunnel splitting from ground state g_{zz} orientation for Tb centre in **2**.

Tb				
Pseudo-KDs Energy (cm^{-1})	g_{xx}	g_{yy}	g_{zz}	Δ_{tun} (cm^{-1})
0.0	0.0002	0.0005	17.6265	0.3
0.3				
34.7	0.0003	0.0005	16.4244	0.9
35.7				
116.8	0.0005	0.0014	8.2102	4.6
121.4				
181.1	0.0000	0.0002	9.8936	16.7
197.8				
-				
296.1	0.0001	0.0006	14.3596	4.4
300.5				

Table S14. SINGLE_ANISO computed g-tensors, the angle of deviation from ground state g_{zz} orientation and relative energies of eight low-lying Kramers' doublets for Dy centre in complex **3**.

Dy				
KDs Energy(cm^{-1})	g_{xx}	g_{yy}	g_{zz}	θ (° angle)
0.0	0.0298	0.0440	19.5632	0.0
152.1	0.9274	1.7941	16.2973	39.0
201.1	10.2639	6.9153	3.1207	59.0
235.2	0.9086	2.0811	10.8589	72.5
289.3	1.6572	4.0687	11.1803	62.5
325.5	1.9910	5.3975	12.1462	72.0

391.0	0.0871	0.2387	18.1467	65.9
485.9	0.0179	0.0341	19.8171	88.0

Table S15. SINGLE_ANISO computed g-tensors, the angle of deviation from ground state g_{zz}orientation and relative energies of eight low-lying Kramers doublets for Er centre in **4**.

Er				
KDs Energy (cm^{-1})	g_{xx}	g_{yy}	g_{zz}	θ (° angle)
0.0	0.9655	1.9316	15.0560	0.0
42.4	1.3515	3.3578	12.7371	110.4
96.4	0.2539	3.5666	9.7213	72.6
171.8	5.3683	4.2753	0.0387	98.0
180.3	1.1524	2.4267	12.4983	43.2
223.2	0.4572	3.2031	11.5457	66.9
252.3	0.1223	3.1282	11.6367	65.3
329.1	0.1408	0.3750	15.8410	79.4

Table S16. SINGLE_ANISO computed crystal field parameters for the Tb centre in **2**; Dy centre in **3** and Er centre in **4** respectively. The major dominating values are kept in bold.

k	**q**	B_k^q	B_k^q	B_k^q
		Tb	**Dy**	**Er**
2	-2	-0.39E+00	1.20E+00	6.80E-01
	-1	-0.38E+01	2.50E+00	1.30E+00
	0	**-0.24E+01**	**-3.70E+00**	**-6.50E-01**
	1	0.17E+01	3.90E+00	5.10E-01
	2	0.30E+01	5.30E+00	3.80E-01
4	-4	0.38E-02	2.10E-02	1.50E-03
	-3	-0.24E-01	-2.90E-01	2.90E-03
	-2	-0.73E-02	-6.90E-03	-2.00E-03
	-1	0.47E-01	-4.20E-02	-7.50E-03
	0	-0.80E-03	-1.90E-02	-9.40E-04
	1	-0.72E-02	4.10E-02	-3.00E-03
	2	0.79E-02	-1.30E-02	4.50E-03
	3	0.31E-01	5.20E-02	-1.10E-02
	4	0.16E-02	4.30E-02	-9.40E-03
6	-6	0.15E-03	1.80E-03	2.10E-04
	-5	-0.25E-03	-3.10E-03	5.50E-04
	-4	-0.81E-04	-1.00E-03	9.30E-06
	-3	0.45E-04	-7.20E-03	2.00E-04
	-2	0.76E-04	5.00E-04	2.80E-04
	-1	0.13E-03	-3.20E-03	2.00E-04
	0	-0.12E-04	-7.80E-04	-3.00E-05

	1	-0.19E-03	-2.70E-03	2.00E-04
	2	-0.30E-03	2.90E-03	-6.50E-05
	3	-0.13E-03	1.50E-03	-4.70E-04
	4	0.21E-03	3.40E-03	1.10E-04
	5	-0.37E-03	-6.20E-03	9.10E-04
	6	-0.52E-04	-1.30E-03	4.20E-04

Table S17. SINGLE_ANISO computed wave function decomposition analysis for Tb centre in **2**. The major dominating values are kept in bold.

±mJ	*wave function decomposition analysis* **Tb**
1	**95.9%|±6⟩ +** 3.4% |±4⟩
2	**33.4% |±5⟩**+ 27.3% |±4⟩+ 23.5% |±3⟩+ 10.2% |±2⟩
3	**57.6% |±5⟩**+ 16.1% |±2⟩ + 8.9% |±4⟩+ 8.0% |±1⟩ + 6.4% |±3⟩
4	**44.2% |±4⟩** + 24.4% |±1⟩ + 11.7%|±3⟩+ 8.0% |0⟩ + 7.9% |±2⟩
5	**40.5% |±3⟩** + 21.3% |0⟩ + 17.8% |±1⟩ + 14.0% |±2⟩
6	**41.0% |±2⟩** + 20.5% |±1⟩ + 17.4% |0⟩ + 10.3% |±3⟩ + 9.0% |±4⟩
7	**52.6% |±1⟩** + 21.4% |±2⟩ + 14.9% |±3⟩ + 9.0% |±4⟩

Table S18. SINGLE_ANISO computed wave function decomposition analysis for Dy centre in **3**. The major dominating values are kept in bold.

±mJ	*wave function decomposition analysis* **Dy**
KD1	**94.1% |±15/2⟩ +** 3.0% |±9/2⟩
KD2	**40.3% |±13/2⟩** + 24.6% |±7/2⟩+ 13.7% |±9/2⟩+ 10.1% |±11/2⟩**+** 5.0% |±3/2⟩
KD3	**31.2 % |±5/2⟩** + 19.4% |±13/2⟩ + 16.9% |±1/2⟩+ 13.6% |±11/2⟩**+** 9.2% |±3/2⟩
KD4	**24.7% |±3/2⟩ +** 17.5% |±11/2⟩ + 15.5% |±9/2⟩ + 13.3% |±1/2⟩ + 9.2% |±7/2⟩
KD5	**23.7% |±5/2⟩**+ 20.0% |±9/2⟩ + 16.9% |±11/2⟩+ 12.9% |±5/2⟩ + 11.5% |±13/2⟩
KD6	**39.2% |±3/2⟩** + 28.0% |±1/2⟩ + 15.3% |±5/2⟩ + 7.3% |±11/2⟩ + 5.8% |±9/2⟩
KD7	**26.6% |±7/2⟩**+ 23.4% + |±9/2⟩ + 18.3% |±1/2⟩ + 12.8% |±11/2⟩ + 11.8% |±5/2⟩
KD8	**45.4% |±1/2⟩**+ 31.0% |±3/2⟩ + 14.8% |±5/2⟩ + 5.3|±7/2⟩

Table S19. SINGLE_ANISO computed wave function decomposition analysis for Er centre in **4**. The major dominating values are kept in bold.

±mJ	*wave function decomposition analysis* **Er**
KD1	**67.8% |±15/2⟩**+ 11.3% |±9/2⟩ + 7.5% |±11/2⟩+ 4.4% |±13/2⟩
KD2	**32.5% |±7/2⟩** + 27.3% |±5/2⟩ + 9.7% |±3/2⟩ + 9.1% |±1/2⟩+ 7.3% |±11/2⟩
KD3	**26.2% |±9/2⟩** + 21.7% |±3/2⟩+ 13.8% |±1/2⟩**+** 11.5% |±7/2⟩ + 10.6% |±13/2⟩
KD4	**33.1% |±13/2⟩ +** 13.5% |±9/2⟩ + 13.0% |±3/2⟩ + 10.9% |±11/2⟩ + 9.3% |±5/2⟩ + 7.4% |±1/2⟩ + 6.9% |±15/2⟩**+** 5.9% |±7/2⟩
KD5	**34.0% |±11/2⟩**+ 23.8% |±13/2⟩ + 10.2% |±15/2⟩ + 10.2% |±5/2⟩ + 7.7% |±9/2⟩ + 7.7%|±1/2⟩
KD6	**22.6% |±9/2⟩** + 20.3% |±7/2⟩ + 13.0% |±5/2⟩ + 12.4% |±3/2⟩+ 12.3% |±11/2⟩+ 11.1% |±13/2⟩
KD7	**32.3% |±7/2⟩**+ 24.0% + |±9/2⟩ + 23.4% |±5/2⟩ + 15.3% |±1/2⟩
KD8	**39.5% |±1/2⟩**+ 34.0% |±3/2⟩ + 12.0% |±5/2⟩ + 5.0%|±13/2⟩

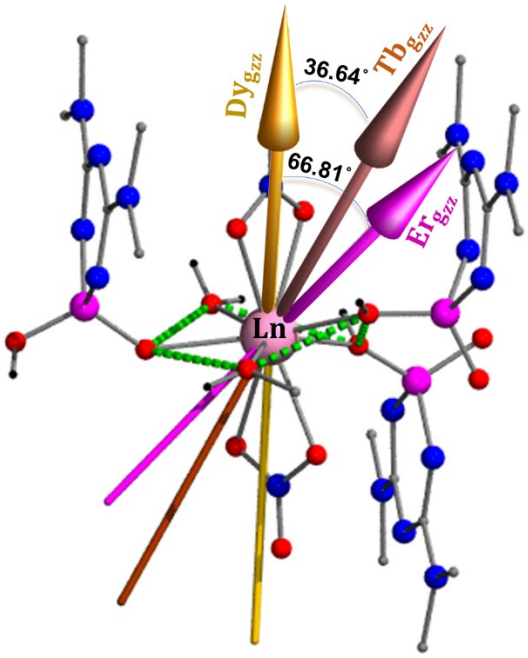

Figure S11. Comparison of computed ground state g_{zz} orientations in **2**-**4**. The angle shows the relative orientations of ground state g_{zz} onTb^{III},$Dy^{III}$and Er^{III} ions in **2**-**4**.

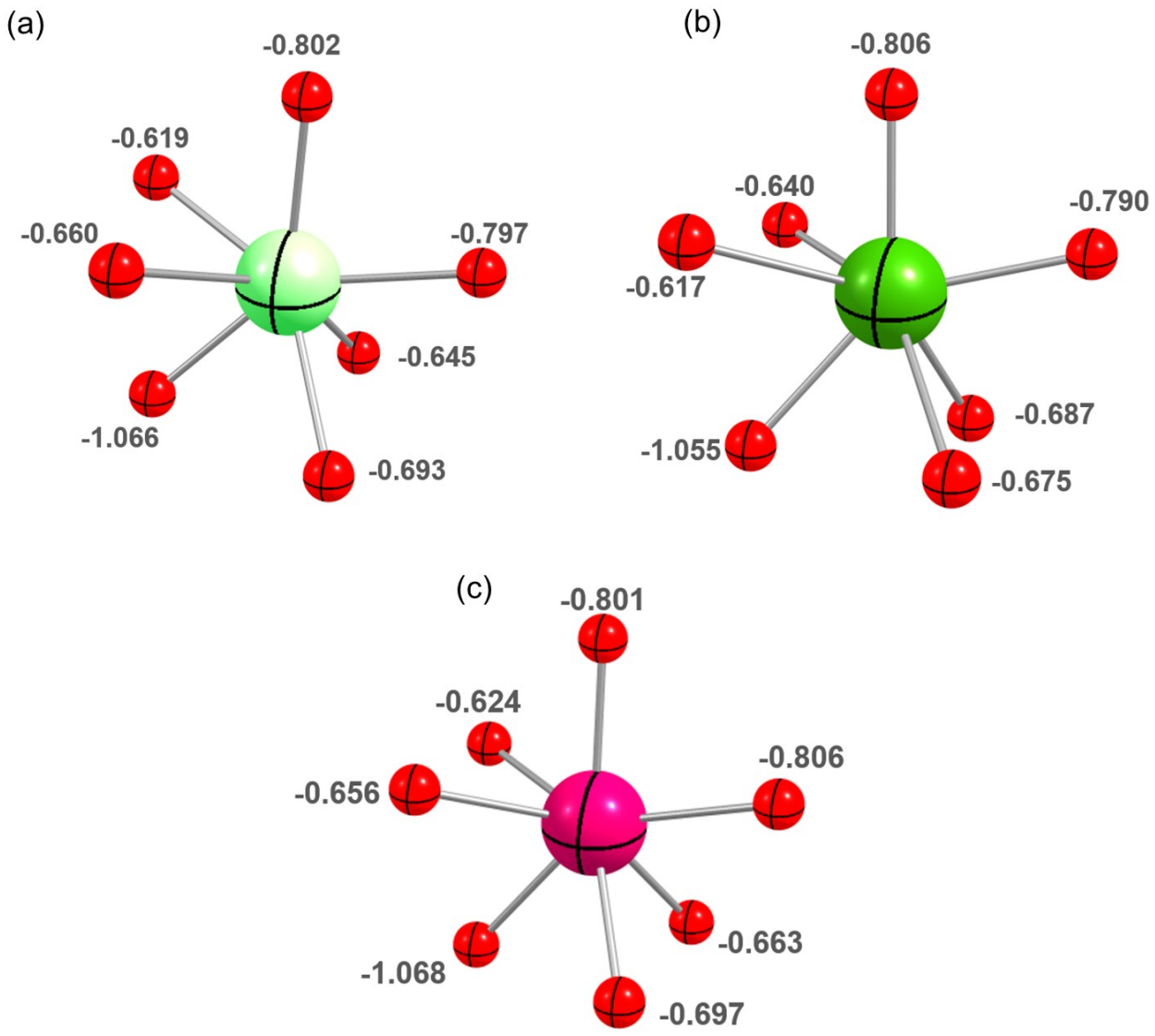

Figure S12. CASSCF computed LoProp charges on the first coordinating atoms of (a) Tb, (b) Dy and (c) Er in complexes **2**-**4**.

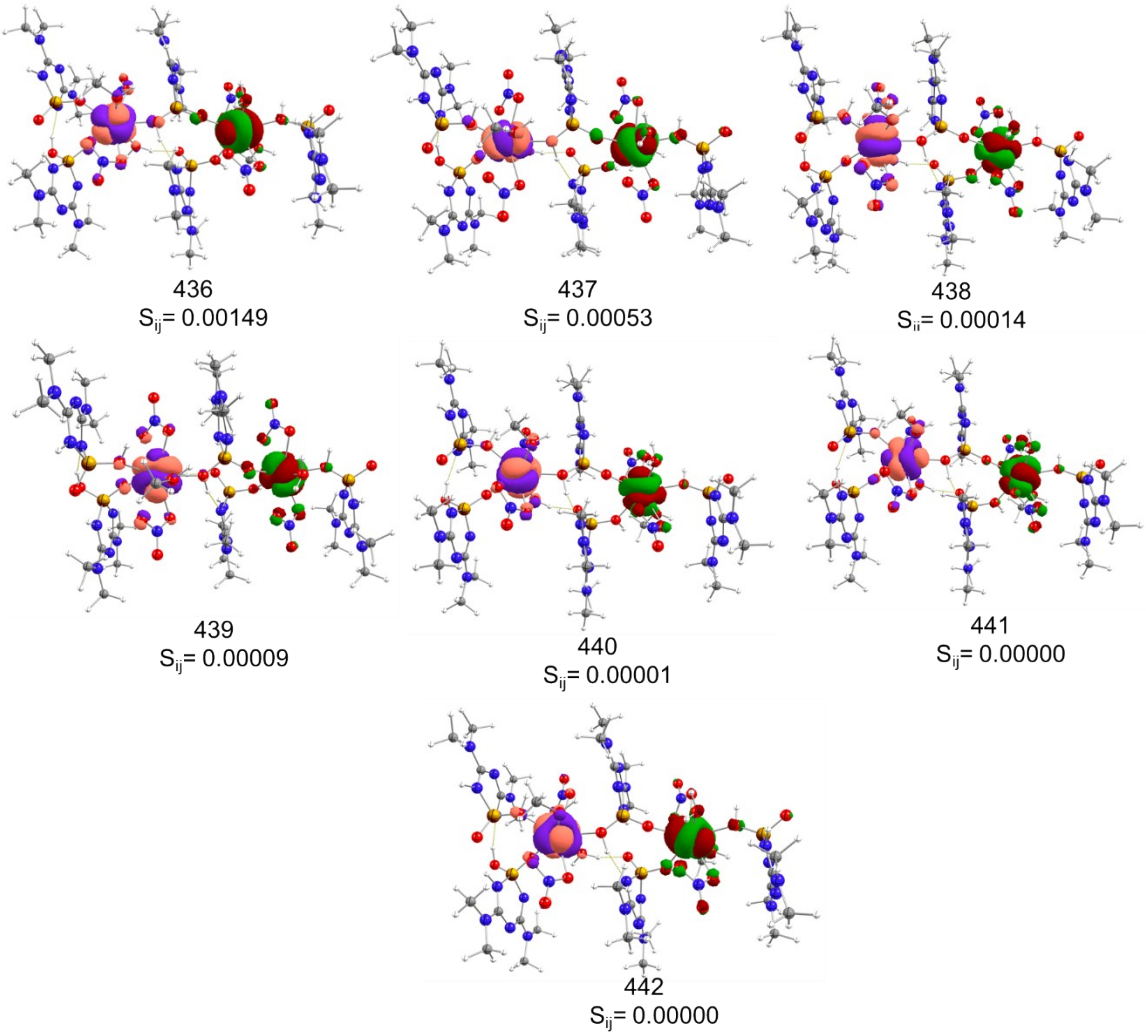

Figure S13. DFT computed overlap integral between the seven 4*f*-orbitals of Gd^{III} centres in Gd-dimer obtained from **1**. The orbitals were constructed using the contour value of 0.03 e^{-}/bohr^3.

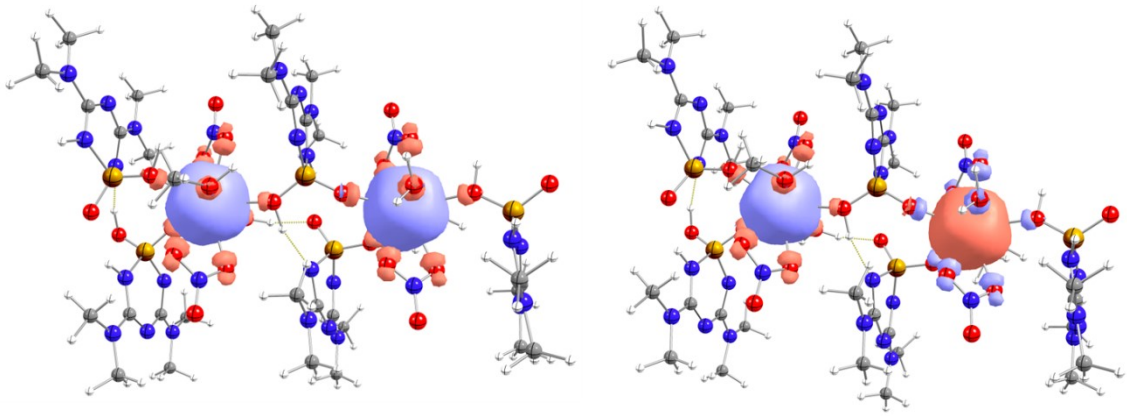

Figure S14. DFT computed spin-density plot for S=7 state of Gd^{III} centres. The iso-density surface here is constructed with a contour value of 0.0005e^-/bohr^3. The purple and orange colour represents the positive and negative spin densities respectively.

Table S20. BS-DFT computed energies of high-spin and broken-symmetry solution of Gd_2 using H= -2JS_1S_2 formalism.

Solution	**Energy (E_h)**	ρ^{Gd1}	ρ^{Gd2}	$<S^{**2}>$	**J (cm^{-1})**
HS	-29049.398021413388	7.0202	7.0217	56.0122	-0.00049
BS	-29049.398021464465	7.0202	-7.0217	7.0122	

J values are estimated using the following equation,

$$J = -\frac{E_{HS} - E_{BS}}{<S^{**2}>_{HS} - <S^{**2}>_{BS}}$$

Table S21. DFT computed overlap integral values for the seven corresponding orbitals between Gd^{III} centres in **1**.

Corresponding Orbital 4f-orbitals	Overlap Integral value (S_{ab})
436	0.00149
437	0.00053
438	0.00014
439	0.00009
440	0.00001
441	0.00000
442	0.00000

Table S22. Hydrogen optimized cartesian coordinates of monomeric complexes **2-4** and dimeric complex **1** respectively.

Monomeric complex **2**			
Tb	0.000000000000	0.000000000000	0.000000000000
P	-0.102791000000	-3.662325000000	-0.531056000000
P	-1.387522000000	3.281525000000	1.222042000000
P	1.586260000000	3.054738000000	-0.858318000000
O	-0.084769000000	-2.412016000000	0.298561000000
O	-0.349529000000	-4.918746000000	0.229606000000
O	-1.405544000000	4.531834000000	0.392425000000
O	1.680469000000	3.767594000000	0.438094000000
O	1.166476000000	0.793404000000	2.082706000000

O	2.262508000000	-0.930198000000	0.103792000000
O	1.457622000000	1.569449000000	-0.826910000000
O	-1.140784000000	2.025104000000	0.461379000000
O	0.440550000000	-0.885480000000	-2.229461000000
O	-2.271430000000	-0.670220000000	-0.085200000000
O	-0.978988000000	0.740492000000	-2.230724000000
O	-0.622890000000	-0.378718000000	2.363938000000
O	0.463467000000	0.376773000000	4.098083000000
O	-0.360460000000	-0.155126000000	-4.125522000000
N	-1.213716000000	-3.527059000000	-1.775301000000
N	0.298120000000	-3.647882000000	-3.603852000000
N	1.293516000000	-3.768705000000	-1.444610000000
N	2.565629000000	-3.560390000000	-3.359263000000
N	-1.957481000000	-3.497895000000	-3.948623000000
N	0.337364000000	0.255950000000	2.874777000000
N	-0.276598000000	3.416791000000	2.466287000000
N	-1.788433000000	3.295968000000	4.294838000000
N	-2.783829000000	3.175145000000	2.135595000000
N	2.877626000000	3.454288000000	-1.857433000000
N	-0.303733000000	-0.096797000000	-2.896259000000
N	0.373831000000	3.730653000000	-1.791908000000
N	1.532145000000	3.458454000000	-3.834903000000
N	-4.055942000000	3.383460000000	4.050249000000
N	0.467168000000	3.445955000000	4.639609000000
N	3.823352000000	3.602886000000	-3.956746000000
N	-0.764730000000	3.666769000000	-3.797718000000
C	-0.932499000000	-3.564556000000	-3.104024000000
C	1.353639000000	-3.645105000000	-2.804380000000
C	2.724768000000	-3.413180000000	-4.802609000000
C	-1.727282000000	-3.595109000000	-5.391066000000
C	3.767375000000	-3.575666000000	-2.549863000000
C	-3.343820000000	-3.486785000000	-3.484897000000
C	-0.557815000000	3.379294000000	3.795010000000
C	0.392445000000	3.609830000000	-3.150956000000
C	-2.843952000000	3.298745000000	3.495366000000
C	2.719343000000	3.507061000000	-3.219550000000
C	-2.053145000000	3.684823000000	-3.094999000000
C	3.741433000000	3.705655000000	-5.407312000000
C	3.524032000000	-0.249562000000	0.210111000000
C	-4.215082000000	3.530670000000	5.493594000000
C	0.236968000000	3.348741000000	6.082052000000
C	-0.830724000000	3.663992000000	-5.261100000000
C	5.158337000000	3.587610000000	-3.353126000000
C	-5.257688000000	3.368184000000	3.240849000000
C	1.853507000000	3.457065000000	4.175882000000
H	0.416169000000	-5.304685000000	0.273289000000

H	-2.038864000000	-3.422901000000	-1.553456000000
H	3.074835000000	-4.228388000000	-5.170846000000
H	3.332702000000	-2.693797000000	-4.986727000000
H	1.872712000000	-3.222919000000	-5.201533000000
H	-0.928335000000	-3.111817000000	-5.620312000000
H	-2.474292000000	-3.217363000000	-5.860388000000
H	-1.624808000000	-4.517252000000	-5.638363000000
H	3.742016000000	-2.847951000000	-1.925304000000
H	4.537035000000	-3.485396000000	-3.116660000000
H	3.819283000000	-4.406151000000	-2.069711000000
H	-3.496874000000	-4.242276000000	-2.912685000000
H	-3.935240000000	-3.534003000000	-4.241227000000
H	-3.511578000000	-2.677132000000	-2.997524000000
H	2.329755000000	-1.711937000000	0.453798000000
H	2.231869000000	1.239199000000	-0.954168000000
H	-1.906483000000	1.639165000000	0.417696000000
H	-2.791481000000	-0.000972000000	0.101806000000
H	-2.441858000000	-1.245310000000	0.542247000000
H	0.548551000000	3.520949000000	2.244442000000
H	3.644836000000	3.629273000000	-1.511939000000
H	-2.023842000000	3.075153000000	-2.354914000000
H	-2.751255000000	3.422346000000	-3.699702000000
H	-2.227729000000	4.572247000000	-2.771889000000
H	3.835283000000	2.833507000000	-5.795405000000
H	4.445095000000	4.276439000000	-5.728617000000
H	2.891397000000	4.076457000000	-5.654609000000
H	3.725575000000	0.183734000000	-0.622033000000
H	4.213885000000	-0.885619000000	0.415891000000
H	3.475741000000	0.405937000000	0.910484000000
H	-4.565148000000	2.715462000000	5.861832000000
H	-4.823015000000	4.250053000000	5.677713000000
H	-3.363026000000	3.720931000000	5.892518000000
H	-0.561978000000	3.832033000000	6.311298000000
H	0.983979000000	3.726487000000	6.551374000000
H	0.134495000000	2.426598000000	6.329349000000
H	0.058060000000	3.697322000000	-5.622117000000
H	-1.325730000000	4.430593000000	-5.560744000000
H	-1.268787000000	2.862672000000	-5.560744000000
H	5.240051000000	4.318103000000	-2.735787000000
H	5.822520000000	3.675102000000	-4.042668000000
H	5.289798000000	2.758514000000	-2.887414000000
H	-5.232329000000	4.095899000000	2.616289000000
H	-6.027348000000	3.458454000000	3.807645000000
H	-5.309596000000	2.537699000000	2.760696000000
H	2.006561000000	2.701574000000	3.603671000000
H	2.444927000000	3.409847000000	4.932213000000

H	2.021265000000	4.266718000000	3.688510000000

Monomeric complex **3**

Dy	0.000000000000	0.000000000000	0.000000000000
P	-3.284534250000	-1.436413470000	0.862992350000
P	2.244886520000	2.698208450000	-1.340521350000
P	3.508922930000	-0.254799930000	0.374838410000
O	-2.253380260000	-0.879839400000	-0.078074350000
O	-4.621461010000	-1.659437340000	0.249502100000
O	4.026825840000	0.049510570000	-0.963957410000
O	3.475969880000	3.171934320000	-0.620325090000
O	-1.488555320000	1.787406040000	0.384984490000
O	1.316092300000	1.889453810000	-0.505811750000
O	0.901319810000	-0.569181490000	-2.254150930000
O	2.112994790000	-0.748225910000	0.477555870000
O	-0.330387360000	-0.925379800000	2.218932540000
O	-0.912282940000	0.598108820000	-2.213174090000
O	0.039389490000	-2.417702290000	-0.297663210000
O	0.579943070000	1.036071980000	2.238462230000
O	-0.017108880000	0.027284100000	-4.128935860000
O	0.266031690000	-0.055244530000	4.119976640000
N	-2.705142550000	-2.015632390000	3.856082890000
N	-2.707492740000	-2.802709370000	1.613999630000
N	-1.758496210000	-4.033918560000	3.325874060000
N	-3.438715070000	-0.462776060000	2.210448290000
N	-3.433267190000	0.064933370000	4.446179860000
N	0.000000000000	0.000000000000	-2.909319880000
N	4.505777750000	-1.345426000000	1.185311690000
N	3.745405950000	1.057831610000	1.387638070000
N	1.703931870000	3.294930220000	-4.337364630000
N	1.473127820000	3.975832590000	-2.072516360000
N	0.189411080000	0.000000000000	2.890139190000
N	4.220140230000	-0.266250630000	3.293153570000
N	3.478376480000	1.917922190000	3.508220840000
N	0.887653480000	5.372571600000	-3.819783840000
N	2.658147320000	1.842552220000	-2.713271930000
N	5.284716100000	-2.295008090000	3.140097410000
N	2.697857060000	1.335393100000	-4.953312690000
C	-3.192856490000	-0.839496590000	3.487810880000
C	-2.402688350000	-2.904154170000	2.950922460000
C	-4.042690200000	1.364487670000	4.171904800000
C	-1.375056720000	-4.247552470000	4.709917230000
C	-1.379333410000	-5.063924880000	2.389423500000
C	-3.270001270000	-0.305687850000	5.858531330000
C	3.804289270000	0.909019830000	2.749613610000
C	4.668245850000	-1.266979240000	2.542937680000

C	2.329435080000	2.181541870000	-3.982545640000
C	5.521551340000	-2.289391350000	4.577371710000
C	2.895546540000	3.153992560000	2.976336320000
C	1.378815400000	4.173132150000	-3.429990520000
C	3.638854450000	1.867903940000	4.965999180000
C	3.322327080000	0.042691910000	-4.680507820000
C	5.722089130000	-3.479048040000	2.394383900000
C	1.142894960000	-3.300319590000	-0.627780290000
C	0.752403090000	5.699240480000	-5.228160190000
C	0.511016610000	6.403741190000	-2.883680700000
C	2.298886290000	1.598691190000	-6.342750660000
H	-4.744931320000	-1.462148390000	-0.700515750000
H	-2.470442170000	-3.627902080000	1.054499320000
H	-3.328214620000	2.188222000000	4.388196530000
H	-4.939508210000	1.506297220000	4.810931730000
H	-4.341996310000	1.401855840000	3.111054960000
H	-1.761786480000	-3.414234240000	5.318433480000
H	-0.269197970000	-4.282577310000	4.812227450000
H	-1.798168280000	-5.201470450000	5.088933370000
H	-2.257096050000	-5.551830330000	1.904717670000
H	-0.835389710000	-5.860390600000	2.927495720000
H	-0.700375210000	-4.682628400000	1.593151980000
H	-4.181938790000	-0.800626900000	6.258710490000
H	-3.079730240000	0.611059480000	6.449316070000
H	-2.411770200000	-0.988319390000	5.974085180000
H	-1.654876210000	2.595808070000	-0.139262210000
H	-2.338263010000	1.536210330000	0.813813810000
H	1.809987540000	1.899919920000	0.395040060000
H	1.920056600000	-1.232321310000	1.320265990000
H	-0.704202960000	-2.578848290000	-0.913344550000
H	5.035656610000	-2.046443300000	0.659076120000
H	3.262761790000	1.022078470000	-2.550589090000
H	5.200450540000	-1.316148010000	4.980825130000
H	6.598885280000	-2.444045830000	4.794968590000
H	4.941697440000	-3.097706670000	5.072684130000
H	3.326363030000	4.016193400000	3.522594930000
H	1.795474830000	3.146625930000	3.125524100000
H	3.139702280000	3.252338630000	1.899968030000
H	2.650789220000	1.986720830000	5.458133230000
H	4.303261560000	2.690242310000	5.304951200000
H	4.070731550000	0.896834610000	5.255202110000
H	4.202860390000	0.142648540000	-4.011012350000
H	3.685177280000	-0.379361360000	-5.634843080000
H	2.604839280000	-0.673991570000	-4.226534750000
H	4.906349990000	-3.892419810000	1.762783080000
H	6.014737060000	-4.264257160000	3.114389620000

H	6.606632860000	-3.274282980000	1.750169440000
H	1.690525080000	-2.929122310000	-1.517489400000
H	0.774942220000	-4.329315880000	-0.814447640000
H	1.821259200000	-3.331069790000	0.243706960000
H	1.003244980000	4.814639250000	-5.835743420000
H	1.426627320000	6.539024010000	-5.512502890000
H	-0.290116520000	6.014307920000	-5.458017270000
H	-0.572087150000	6.648910210000	-2.975396050000
H	1.084882230000	7.337588980000	-3.076487590000
H	0.726971100000	6.044299830000	-1.862344970000
H	1.324739300000	1.117696980000	-6.578709420000
H	3.074139500000	1.199212590000	-7.025259320000
H	2.196950040000	2.686128220000	-6.491236180000

Monomeric complex **4**

Er	0.000000000000	0.000000000000	0.000000000000
P	-1.561990000000	3.024620000000	0.883740000000
P	1.372910000000	3.257420000000	-1.227610000000
P	0.124770000000	-3.642430000000	0.538760000000
O	1.406280000000	4.503950000000	-0.393010000000
O	-1.665470000000	3.749100000000	-0.408050000000
O	-1.433450000000	1.538530000000	0.840410000000
O	1.120390000000	1.997920000000	-0.474580000000
O	0.576270000000	-0.396330000000	-2.334680000000
O	-0.398010000000	-0.883460000000	2.197640000000
O	0.996580000000	0.754570000000	2.187490000000
O	-1.180730000000	0.813910000000	-2.027960000000
O	-2.210810000000	-0.938660000000	-0.105860000000
O	2.239400000000	-0.664040000000	0.060730000000
O	0.415530000000	-0.141030000000	4.084380000000
O	-0.521950000000	0.373690000000	-4.062080000000
O	0.091400000000	-2.395900000000	-0.295840000000
O	0.377300000000	-4.901930000000	-0.214270000000
N	-0.346300000000	3.704940000000	1.810970000000
N	-0.381960000000	0.263300000000	-2.842800000000
N	2.766380000000	3.143300000000	-2.140890000000
N	0.263470000000	3.395830000000	-2.470460000000
N	0.337750000000	-0.088590000000	2.853320000000
N	-2.850110000000	3.412390000000	1.896170000000
N	-1.507950000000	3.411010000000	3.866670000000
N	1.785120000000	3.284050000000	-4.301730000000
N	0.796380000000	3.623520000000	3.826790000000
N	4.052500000000	3.347530000000	-4.053380000000
N	-0.470600000000	3.430330000000	-4.644340000000
N	-3.788370000000	3.557290000000	3.995370000000
N	-1.268700000000	-3.756560000000	1.452040000000

N	1.234210000000	-3.504020000000	1.781610000000
N	-0.287440000000	-3.615800000000	3.612880000000
N	-2.554810000000	-3.552320000000	3.364530000000
N	1.968280000000	-3.469520000000	3.955490000000
C	-0.366920000000	3.566950000000	3.168750000000
C	0.537760000000	3.358570000000	-3.799590000000
C	-2.696240000000	3.463450000000	3.253950000000
C	2.829480000000	3.270250000000	-3.507730000000
C	-3.706290000000	3.651120000000	5.452850000000
C	2.074250000000	3.656640000000	3.119800000000
C	5.258370000000	3.330970000000	-3.248500000000
C	-3.507890000000	-0.277650000000	-0.204840000000
C	4.205150000000	3.495190000000	-5.496350000000
C	0.865310000000	3.600070000000	5.286080000000
C	-1.863630000000	3.448270000000	-4.176650000000
C	-5.133780000000	3.544870000000	3.398970000000
C	-0.232190000000	3.322690000000	-6.090940000000
C	-1.331800000000	-3.629600000000	2.818880000000
C	0.959920000000	-3.541280000000	3.110740000000
C	-3.760690000000	-3.568880000000	2.559650000000
C	-2.707470000000	-3.404660000000	4.807500000000
C	3.361310000000	-3.451580000000	3.487800000000
C	1.729870000000	-3.577160000000	5.402090000000
H	-2.231230000000	1.252740000000	0.706990000000
H	1.610380000000	1.974460000000	0.213910000000
H	-2.295810000000	-1.718340000000	-0.453190000000
H	2.748080000000	-0.016840000000	-0.195780000000
H	2.377420000000	-1.280890000000	-0.522080000000
H	3.503020000000	2.991500000000	-1.722140000000
H	-3.999010000000	2.824520000000	5.844410000000
H	-4.267880000000	4.365950000000	5.761030000000
H	-2.797400000000	3.823620000000	5.712080000000
H	2.090630000000	2.965280000000	2.454510000000
H	2.789190000000	3.515890000000	3.745210000000
H	2.183370000000	4.510850000000	2.697420000000
H	5.235070000000	4.056840000000	-2.621280000000
H	6.025680000000	3.427570000000	-3.819530000000
H	5.316170000000	2.498850000000	-2.775370000000
H	-3.467350000000	0.406810000000	-0.877380000000
H	-3.731310000000	0.117020000000	0.639920000000
H	-4.177320000000	-0.922100000000	-0.445940000000
H	4.598980000000	2.698940000000	-5.858910000000
H	4.771110000000	4.247270000000	-5.683070000000
H	3.343260000000	3.635940000000	-5.895160000000
H	-0.023560000000	3.602820000000	5.648630000000
H	1.340090000000	4.374230000000	5.596060000000

H	1.324910000000	2.806580000000	5.572500000000
H	-2.027840000000	2.675480000000	-3.632810000000
H	-2.454930000000	3.438610000000	-4.934390000000
H	-2.016500000000	4.243130000000	-3.661820000000
H	-5.219770000000	4.280390000000	2.788060000000
H	-5.792250000000	3.624900000000	4.093260000000
H	-5.266540000000	2.719640000000	2.925830000000
H	0.519090000000	3.870540000000	-6.333860000000
H	-1.010990000000	3.622150000000	-6.565890000000
H	-0.048830000000	2.409150000000	-6.319350000000
H	-3.489480000000	3.451030000000	1.631510000000
H	-0.112700000000	-4.925390000000	-0.902760000000
H	-2.005340000000	-3.908350000000	1.033290000000
H	-3.737390000000	-2.843020000000	1.932430000000
H	-4.528000000000	-3.472280000000	3.130680000000
H	-3.818490000000	-4.401000000000	2.086520000000
H	-3.101300000000	-4.200910000000	5.170060000000
H	-3.273430000000	-2.652580000000	4.994220000000
H	-1.845580000000	-3.263910000000	5.206310000000
H	3.525520000000	-4.224360000000	2.943960000000
H	3.952610000000	-3.461240000000	4.245540000000
H	3.514180000000	-2.656720000000	2.972970000000
H	0.978590000000	-3.029310000000	5.645010000000
H	2.508670000000	-3.277710000000	5.877040000000
H	1.546510000000	-4.490700000000	5.630500000000

Dimeric complex **1**

Gd	-0.003687000000	0.001315000000	-0.005878000000
Gd	1.578985000000	-6.972212000000	-0.560177000000
P	0.198183000000	-3.660380000000	0.637489000000
P	3.172093000000	-3.899863000000	-1.476728000000
P	1.379848000000	3.309537000000	-1.210353000000
P	-1.594258000000	3.076857000000	0.904350000000
P	1.374093000000	-10.635055000000	-1.196012000000
O	0.148359000000	-2.432148000000	-0.204778000000
O	0.455488000000	-4.917107000000	-0.111837000000
O	3.271839000000	-3.140449000000	-0.225300000000
O	3.055831000000	-5.374080000000	-1.381276000000
O	2.050822000000	-7.826635000000	-2.846382000000
O	2.759348000000	-6.165794000000	1.542871000000
O	-0.750471000000	-7.565109000000	-0.518228000000
O	1.033820000000	-7.404464000000	1.828334000000
O	0.558701000000	-6.312962000000	-2.781298000000
O	3.821585000000	-7.976833000000	-0.387773000000
O	2.149766000000	-6.686408000000	3.542771000000
O	1.198457000000	-7.118399000000	-4.690002000000

O	1.424931000000	-9.405134000000	-0.356273000000
O	1.121505000000	2.054566000000	-0.458443000000
O	-1.479213000000	1.602351000000	0.811931000000
O	-0.476227000000	-0.848013000000	2.282079000000
O	-1.183386000000	0.804380000000	-2.110581000000
O	2.325280000000	-0.593589000000	-0.046605000000
O	0.541120000000	-0.436298000000	-2.393495000000
O	1.017143000000	0.664290000000	2.213883000000
O	-2.247115000000	-1.001807000000	-0.176218000000
O	1.430686000000	4.539457000000	-0.370614000000
O	-1.693378000000	3.833777000000	-0.348638000000
O	-0.574235000000	0.279151000000	-4.109406000000
O	0.376724000000	-0.136690000000	4.124240000000
O	1.115751000000	-11.890026000000	-0.444101000000
N	1.949420000000	-3.269048000000	-2.411587000000
N	1.329688000000	-3.468004000000	1.846857000000
N	-1.179990000000	-3.794952000000	1.569216000000
N	1.986592000000	-6.764272000000	2.340712000000
N	4.452890000000	-3.528152000000	-2.505257000000
N	1.273856000000	-7.078130000000	-3.469760000000
N	-0.140092000000	-3.636999000000	3.710941000000
N	3.115376000000	-3.563086000000	-4.468164000000
N	0.815076000000	-3.391969000000	-4.425294000000
N	-2.424407000000	-3.583271000000	3.512135000000
N	5.383411000000	-3.374531000000	-4.619557000000
N	2.136986000000	-3.505164000000	4.002975000000
N	0.248501000000	3.500358000000	-2.420115000000
N	2.757908000000	3.171911000000	-2.141800000000
N	-0.371063000000	3.708584000000	1.837910000000
N	-2.874747000000	3.451739000000	1.932112000000
N	0.301358000000	-0.098869000000	2.903917000000
N	-0.411125000000	0.203625000000	-2.907189000000
N	1.718138000000	3.326316000000	-4.283845000000
N	-1.537261000000	3.419739000000	3.895086000000
N	0.763180000000	3.588870000000	3.851867000000
N	4.002497000000	3.378568000000	-4.085148000000
N	-0.558830000000	3.459429000000	-4.576153000000
N	-3.805139000000	3.610477000000	4.046091000000
N	0.242746000000	-10.444233000000	-2.405773000000
N	2.752153000000	-10.772680000000	-2.127458000000
N	1.712384000000	-10.618275000000	-4.269504000000
N	3.996743000000	-10.566024000000	-4.070807000000
N	-0.564584000000	-10.485162000000	-4.561812000000
C	1.083287000000	-3.547402000000	3.187495000000
C	1.953775000000	-3.428029000000	-3.778111000000
C	4.277742000000	-3.483452000000	-3.855861000000

C	-1.211558000000	-3.659677000000	2.924718000000
C	5.257116000000	-3.254657000000	-6.074566000000
C	-3.648040000000	-3.612833000000	2.751263000000
C	-0.480083000000	-3.372608000000	-3.746381000000
C	-2.507581000000	-3.432538000000	4.962784000000
C	3.500697000000	-3.443461000000	3.531135000000
C	5.089221000000	-7.333065000000	-0.338275000000
C	6.724526000000	-3.352186000000	-4.048976000000
C	1.934661000000	-3.652809000000	5.440869000000
C	0.766905000000	-3.463161000000	-5.872622000000
C	-0.375549000000	3.552418000000	3.204759000000
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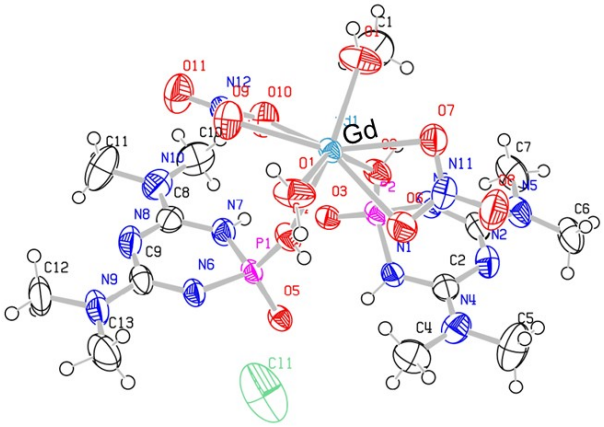

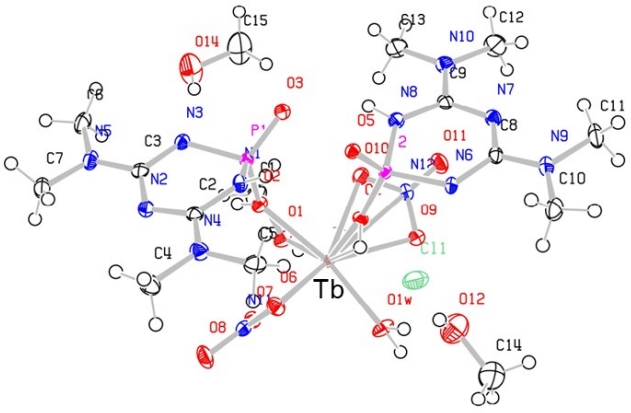

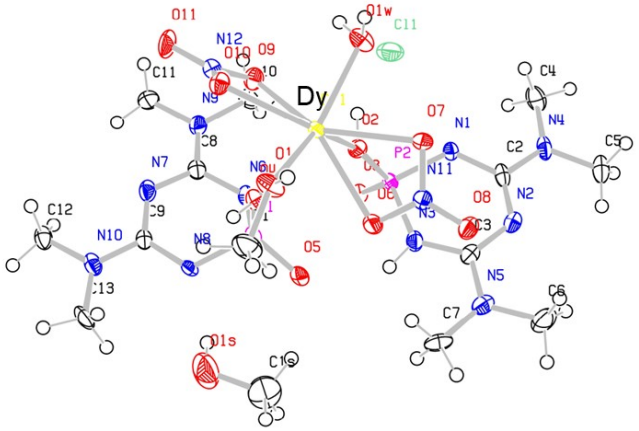

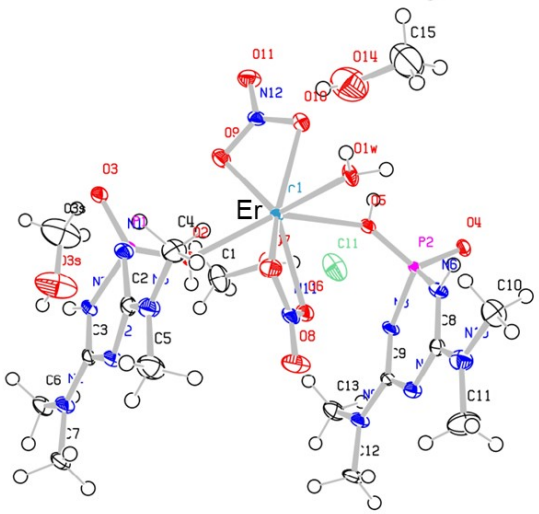

Figure S15. ORTEP diagrams drawn (using ORTEP-3 software[2]) for complexes **1-4** at 50% thermal ellipsoid probability.

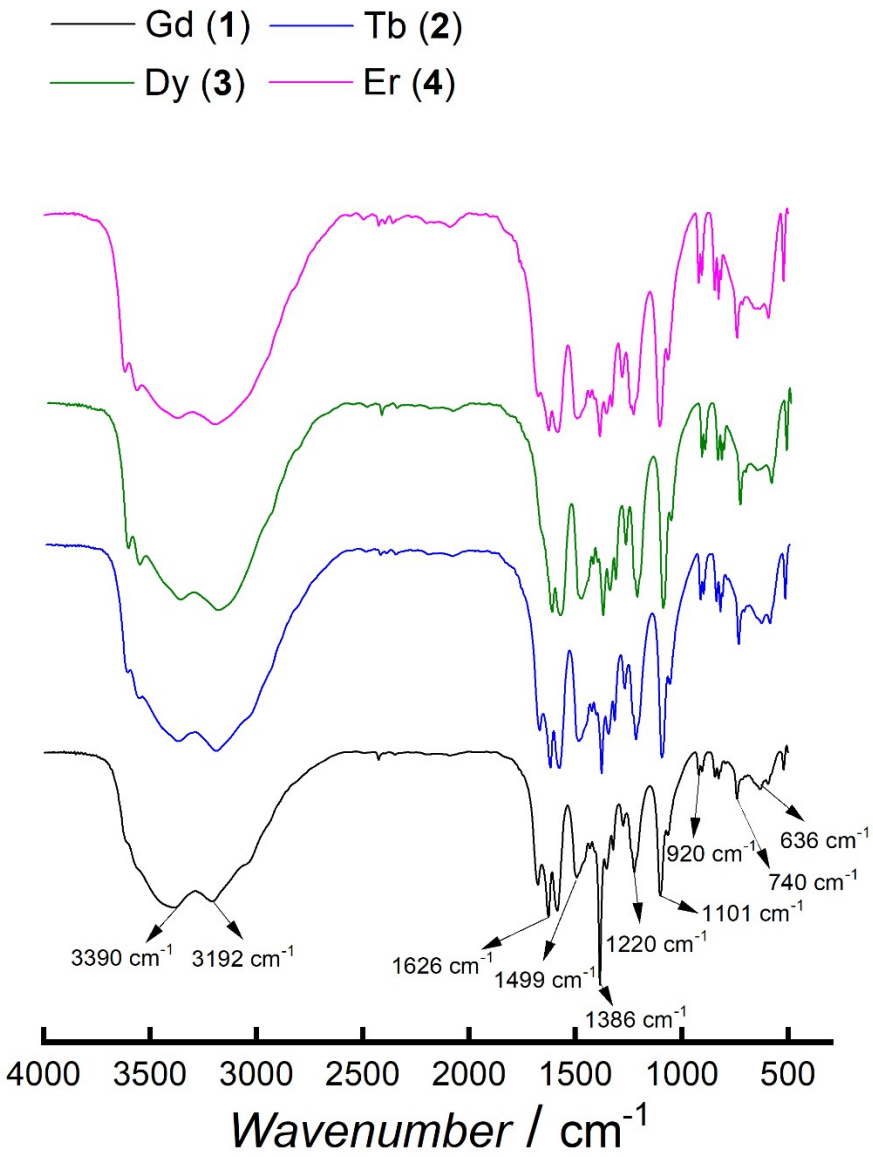

Figure S16. FTIR spectra of complexes **1-4**. The characteristic bands are listed in main text.

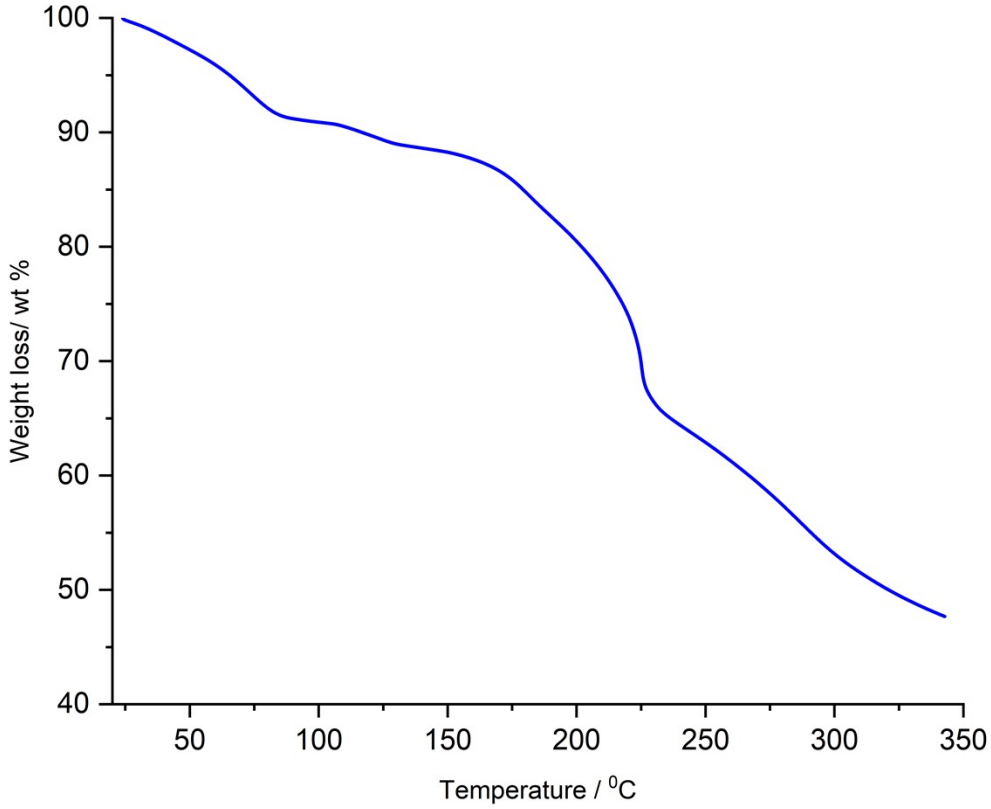

Figure S17. Thermogravimetric analysis (TGA) of **3**.

The thermal gravimetric analysis (TGA) data indicates that when the temperature reaches 70°C, the sample experiences a weight loss of around 8.5%, equivalent to the departure of two methanol molecules from the crystal lattice. While one of these methanol molecules has been accounted for in the updated cif (Crystallographic Information File), the other molecule was masked due to its significant disorder.

References

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2. L. J. Farrugia, WinGX and ORTEP for Windows: an update *J. Appl. Cryst.* (2012), 45, 849-854.