Supporting Information

Optimization of the interatomic distance in the [Gd-O]$^+$ cation

In order to ensure ourselves that the obtained short bond length in Dy-O$^+$ (Figure 1) is not accidental, we performed the geometry optimization of a similar complex Gd-O$^+$, and check the result against the size of the used active spaces of the multiconfigurational self-consistent field method (CASSCF) and additional account of dynamical correlation (CASPT2).
Optimization of the distance in the Gd-O$^+$ cation was performed with MOLCAS at the CASSCF/CASPT2 level of theory, using the Triple-Zeta Polarized (TZP) basis sets for both atoms from ANO-RCC Basis Set Library available in MOLCAS. The following contractions were employed:

Gd – ANO-RCC…8s7p5d4f2g1h.
O – ANO-RCC…4s3p2d1f.

Three active spaces were considered for the CASSCF method:

AS1 — restricted active space – 7 electrons in 7 orbitals (the 4f shell of the Gd) — CAS(7 in 7).
AS2 — Gd(4f shell) + O{2px,2py,2pz}+3 virtual orbitals — CAS(13 in 13).
AS3 — Gd(4f shell) + O{2s,2px,2py,2pz}+4 virtual orbitals — CAS(15 in 15).

Dynamic correlation was added by means of the CASPT2 program.

	Level of theory	Active space	R (in Å)	Charge on Gd	Charge on O	Natural Bond Order*
1	CASSCF	**AS1**	1.75	1.7454	-0.7454	4.985
2	CASSCF/CASPT2	**AS1**	1.76	1.6907	-0.6907	5.987
3	CASSCF	**AS2**	1.79	1.5910	-0.5910	1.983
4	CASSCF/CASPT2	**AS2**	1.76	1.5510	-0.5510	3.971
5	CASSCF	**AS3**	1.78	1.5983	-0.5983	5.844
6	CASSCF/CASPT2	**AS3**	1.75	1.5411	-0.5411	5.817

* -- obtained from the CASSCF program.

For the cation [Dy-O]$^+$ a slightly smaller bond length is expected (Figure 1) because of a smaller radius of Dy^{3+} compared to Gd^{3+}.

Computational details:

The entire Dy_3 triangle was computed ab initio: no optimization of the structure was done.
In order to compute the energy spectrum of the individual Dy centers, the neighboring Dy ions were computationally simulated by a diamagnetic closed-shell La^{3+}.
Active space of the Complete Active Space Self-Consistent field method included the 4f shell of the Dy^{3+} ion: CAS(9 in 7).
Spin-orbit coupling was done by the Restricted Active Space State Interaction (RASSI) method. The number of states included in the spin-orbit coupling (RASSI) was 21 sextets (all) 108 quartets (224 in total) and 32 doublets (490 in total). Including more states would probably change the states which are very high in energy, while the energies and magnetic properties of the lowest states remain the same.
All basis sets were taken from the ANO-RCC basis set library available in MOLCAS. The following contractions were used:
Dy, La -- 7s6p4d3f1g.
Cl -- 4s3p.
O, N, C -- 3s2p.
H.-- 2s.
This afforded a set of 664 basis functions which were further used in the CASSCF/RASSI programs.

Table S1: CASSCF (spin-free) and RASSI (spin-orbit) energies (cm^{-1}) for Dy1-Dy3 centers in Dy_3 triangle.

Spin Multiplicity		CASSCF			RASSI		
		1	2	3	1	2	3
6	6H	0.000	0.000	0.000	0.000	0.000	0.000
		6.914	2.091	34.429	223.555	130.353	103.476
		310.476	197.261	100.608	357.842	273.744	141.038
		380.549	217.861	177.968	443.316	390.084	171.100
		485.781	413.776	219.825	506.003	464.282	208.691
		498.163	468.672	266.042	541.690	513.565	228.586
		555.171	525.403	283.091	582.487	577.534	287.051
		619.799	589.381	332.093	622.393	699.696	395.074
		626.464	605.677	334.203	3631.505	3575.763	3566.062
		664.782	743.297	469.169	3767.427	3728.404	3628.893
		673.407	750.799	485.901	3880.100	3825.851	3667.381
	6F	7748.439	7716.354	7569.958	3951.925	3902.917	3689.112
		7772.399	7740.964	7634.101	3992.281	3968.580	3721.390
		7864.434	7854.917	7660.064	4033.476	4027.186	3754.292
		7933.689	7901.437	7721.728	4079.714	4113.021	3797.647
		7959.947	7921.467	7746.345	6245.970	6172.193	6129.318
		7989.872	7977.258	7781.261	6328.442	6306.240	6175.260
		8001.254	7995.910	7783.440	6410.074	6383.502	6201.756
	6P	34785.778	34722.573	34849.737	6479.141	6440.971	6219.078
		35292.229	35267.587	34987.968	6526.664	6513.050	6267.326

	35495.540	35535.525	35168.419	6612.469	6620.108	6318.432
	24969.288	24960.051	24886.147	8236.342	8166.178	8085.637
	24973.403	24963.039	24894.519	8296.722	8288.602	8125.956
	25124.651	25065.113	24959.322	8386.182	8349.757	8156.256
	25133.550	25067.648	24967.050	8443.696	8421.077	8198.904
	25202.574	25140.083	24975.852	8543.882	8544.870	8263.910
	25211.226	25157.396	24977.316	9779.193	9716.942	9611.188
	25221.475	25178.115	24984.643	9860.103	9847.666	9676.577
	25236.700	25215.366	25013.649	9961.883	9927.103	9723.917
	25248.175	25238.706	25024.665	10061.544	10064.545	9784.673
	25260.750	25249.512	25058.927	10146.624	10119.223	9967.826
4	25278.689	25273.308	25066.958	10211.087	10178.168	10009.150
	25308.500	25278.310	25105.642	10249.551	10218.951	10025.167
	25320.212	25291.745	25118.389	10272.599	10259.982	10061.698
	25386.639	25361.738	25212.631	10331.645	10299.857	10073.209
	25396.893	25405.608	25235.076	10343.643	10342.830	10103.747
	25413.836	25411.184	25248.800	10938.349	10882.896	10803.037
	25432.651	25428.549	25280.983	11185.953	11140.801	10911.163
	25442.530	25434.897	25295.715	11263.296	11292.775	10992.428
	25457.743	25440.680	25304.905	11741.275	11702.593	11518.547
	25465.146	25451.914	25309.835	11750.240	11724.653	11543.097
	…	…	…	11757.030	11738.483	11550.133
	37513.109	37493.317	37337.482	11780.641	11775.471	11562.359
	37514.249	37494.871	37347.137	11836.501	11815.478	11588.881
	37559.787	37526.057	37363.862	13666.025	13642.379	13445.736
	37571.512	37532.184	37368.460	13675.400	13656.386	13460.655
	37577.972	37548.100	37380.434	13730.243	13703.776	13508.642
	37585.397	37576.799	37399.119	13754.708	13736.568	13525.798
	37597.500	37601.369	37413.278	15127.373	15111.572	14913.240
	37655.921	37612.800	37457.156	15162.077	15134.783	14938.245
	37677.228	37644.754	37475.092	15199.709	15178.376	14973.922
	37683.821	37683.477	37484.813	16057.699	16036.400	15836.563
2	37710.886	37712.838	37547.491	16063.673	16042.509	15840.672
	37720.716	37718.719	37555.628	16530.233	16511.127	16305.118
	37756.930	37728.746	37556.944	25275.996	25231.763	25159.085
	37765.355	37729.987	37566.883	25357.352	25336.479	25187.021
	37768.643	37744.505	37587.548	25409.704	25380.788	25199.628
	37775.554	37766.615	37599.714	25464.686	25426.796	25242.674
	37787.437	37781.623	37603.563	25487.525	25463.087	25294.315
	39144.750	39142.990	39011.362	25554.200	25542.096	25332.405
	39145.247	39143.765	39014.485	25580.130	25566.312	25347.233
	39254.330	39213.066	39054.890	25609.162	25613.468	25380.689
	…	…	…	…	…	…

Table S2: Comparative g_X, g_Y, g_Z for 8 lowest Kramers doublets of the Dy1-Dy3 centers of the Dy_3 triangle.

KD		1		2		3	
		E (cm^{-1})	g	E (cm^{-1})	g	E (cm^{-1})	g
1	g_X	0.000	0.0015	0.000	0.0048	0.000	0.0088
	g_Y		0.0026		0.0072		0.0341
	g_Z		19.7285		19.5877		19.6673
2	g_X	223.555	0.0302	130.353	0.0263	103.476	0.3985
	g_Y		0.0430		0.0295		0.6451
	g_Z		16.6940		16.7704		17.7500
3	g_X	357.842	0.9851	273.744	0.1991	141.038	3.4955
	g_Y		1.1679		0.2423		3.6706
	g_Z		13.6251		13.9970		11.3661
4	g_X	443.316	4.2030	390.084	0.8390	171.100	2.2148
	g_Y		5.4340		1.0588		4.9693
	g_Z		8.9738		11.0519		11.7486
5	g_X	506.003	2.2538	464.282	3.7772	208.691	0.0690
	g_Y		3.0835		5.3005		3.2231
	g_Z		13.9778		8.5234		14.0985
6	g_X	541.690	0.1392	513.565	2.0094	228.586	1.2158
	g_Y		1.7831		3.6592		3.0479
	g_Z		13.6022		14.2754		16.4501
7	g_X	582.487	0.2282	577.534	0.2872	287.051	0.1044
	g_Y		0.6579		0.7102		0.2814
	g_Z		17.4705		18.0290		18.7437
8	g_X	622.393	0.0738	699.696	0.0182	395.074	0.0022
	g_Y		0.1792		0.0383		0.0087
	g_Z		18.8487		19.5646		19.5569

Table S3: *g*-factors and main magnetic axes (3×3) for 8 lowest Kramers doublets of the Dy1 center from Dy_3 triangle.

KD	E (cm^{-1})		g	*x*	*y*	*z*
1	0.000	g_X	0.0015	0.9883	0.0310	-0.1495
		g_Y	0.0026	-0.1527	0.1821	-0.9714
		g_Z	19.7285	-0.0029	0.9828	0.1847
2	223.555	g_X	0.0302	0.9960	0.0313	0.0836
		g_Y	0.0430	0.0824	0.0365	-0.9959
		g_Z	16.6940	-0.0342	0.9988	0.0338
3	357.842	g_X	0.9851	0.9438	0.0367	-0.3285
		g_Y	1.1679	0.3146	0.2056	0.9267
		g_Z	13.6251	0.1015	-0.9780	0.1825
4	443.316	g_X	4.2030	0.3368	0.3735	0.8643
		g_Y	5.4340	-0.9345	0.0202	0.3555
		g_Z	8.9738	0.1153	-0.9274	0.3558
5	506.003	g_X	2.2538	-0.6853	-0.3473	0.6402
		g_Y	3.0835	0.1742	-0.9316	-0.3189
		g_Z	13.9778	0.7071	-0.1070	0.6989
6	541.690	g_X	0.1392	-0.2467	-0.9453	-0.2134
		g_Y	1.7831	0.8441	-0.3177	0.4318
		g_Z	13.6022	-0.4760	-0.0736	0.8764
7	582.487	g_X	0.2282	0.3345	-0.9199	-0.2049
		g_Y	0.6579	0.4096	-0.0539	0.9107
		g_Z	17.4705	-0.8487	-0.3885	0.3587
8	622.393	g_X	0.0738	0.1884	0.9613	-0.2013
		g_Y	0.1792	-0.4824	-0.0879	-0.8715
		g_Z	18.8487	-0.8554	0.2613	0.4472

Table S4: Relative angles between the main anisotropy axes in different doublet states of the Dy1 center from Dy_3 triangle.

KD	1	2	3	4	5	6	7	8
1	0.00	8.41	20.62	29.66	89.27	86.44	74.24	71.62
2	8.41	0.00	12.20	21.23	82.28	87.98	71.67	73.64
3	20.62	12.20	0.00	9.21	71.65	80.90	69.78	76.02
4	29.66	21.23	9.21	0.00	65.27	73.97	66.67	80.39
5	89.27	82.28	71.65	65.27	0.00	82.75	75.80	69.44
6	86.44	87.98	80.90	73.97	82.75	0.00	29.31	31.81
7	74.24	71.67	69.78	66.67	75.80	29.31	0.00	34.73
8	71.62	73.64	76.02	80.39	69.44	31.81	34.73	0.00

Table S5: *g*-factors and main magnetic axes (3×3) for 8 lowest Kramers doublets of the Dy2 center from Dy_3 triangle.

KD	E (cm^{-1})		g	*x*	*y*	*z*
1	0.000	g_X	0.0048	-0.8920	-0.1358	0.4312
		g_Y	0.0072	0.3713	-0.7642	0.5273
		g_Z	19.5877	0.2579	0.6305	0.7321
2	130.353	g_X	0.0263	0.9624	-0.2687	0.0389
		g_Y	0.0295	0.2165	0.6731	-0.7072
		g_Z	16.7704	0.1638	0.6890	0.7060
3	273.744	g_X	0.1991	-0.8093	-0.3135	0.4967
		g_Y	0.2423	0.5745	-0.5987	0.5582
		g_Z	13.9970	0.1224	0.7371	0.6646
4	390.084	g_X	0.8390	-0.4714	0.5332	-0.7025
		g_Y	1.0588	-0.8806	-0.2414	0.4078
		g_Z	11.0519	0.0478	0.8108	0.5833
5	464.282	g_X	3.7772	-0.8255	-0.2033	-0.5266
		g_Y	5.3005	-0.4727	-0.2608	0.8417
		g_Z	8.5234	-0.3084	0.9437	0.1192
6	513.565	g_X	2.0094	0.4670	0.5980	0.6514
		g_Y	3.6592	-0.8837	0.3424	0.3191
		g_Z	14.2754	-0.0322	-0.7246	0.6884
7	577.534	g_X	0.2872	0.4596	0.5503	0.6971
		g_Y	0.7102	-0.5348	0.7981	-0.2774
		g_Z	18.0290	-0.7090	-0.2454	0.6611
8	699.696	g_X	0.0182	0.1244	-0.8781	-0.4620
		g_Y	0.0383	0.0276	0.4685	-0.8830
		g_Z	19.5646	0.9919	0.0971	0.0825

Table S6: Relative angles between the main anisotropy axes in different doublet states of the Dy2 center from Dy_3 triangle.

KD	1	2	3	4	5	6	7	8
1	0.00	5.66	9.30	15.97	45.09	88.68	82.86	69.30
2	5.66	0.00	4.00	10.89	40.13	88.98	80.35	74.33
3	9.30	4.00	0.00	6.96	36.17	85.75	80.66	76.52
4	15.97	10.89	6.96	0.00	29.26	79.96	81.60	80.21
5	45.09	40.13	36.17	29.26	0.00	57.40	85.22	82.29
6	88.68	88.98	85.75	79.96	57.40	0.00	51.41	87.67
7	82.86	80.35	80.66	81.60	85.22	51.41	0.00	44.83
8	69.30	74.33	76.52	80.21	82.29	87.67	44.83	0.00

Table S7: *g*-factors and main magnetic axes (3×3) for 8 lowest Kramers doublets of the Dy3 center from Dy_3 triangle.

KD	E (cm^{-1})		g	*x*	*y*	*z*
1	0.000	g_X	0.0088	0.9358	0.3501	-0.0414
		g_Y	0.0341	-0.2162	0.6628	0.7169
		g_Z	19.6673	0.2784	-0.6619	0.6959
2	103.476	g_X	0.3985	0.8841	0.3838	-0.2665
		g_Y	0.6451	-0.3729	0.9233	0.0925
		g_Z	17.7500	0.2815	0.0176	0.9594
3	141.038	g_X	3.4955	0.1532	0.9880	0.0202
		g_Y	3.6706	-0.8300	0.1176	0.5452
		g_Z	11.3661	0.5363	-0.1003	0.8381
4	171.100	g_X	2.2148	0.0546	0.0617	0.9966
		g_Y	4.9693	0.4811	-0.8729	0.0804
		g_Z	11.7486	0.8749	0.4839	0.0180
5	208.691	g_X	0.0690	0.5153	0.6266	-0.5847
		g_Y	3.2231	-0.8373	0.2226	-0.4994
		g_Z	14.0985	-0.1828	0.7469	0.6393
6	228.586	g_X	1.2158	-0.5445	0.4187	-0.7268
		g_Y	3.0479	-0.7972	0.0111	0.6036
		g_Z	16.4501	0.2608	0.9081	0.3278
7	287.051	g_X	0.1044	-0.1898	-0.1922	0.9628
		g_Y	0.2814	-0.2326	-0.9439	-0.2343
		g_Z	18.7437	0.9539	-0.2684	0.1344
8	395.074	g_X	0.0022	0.6627	-0.7400	0.1151
		g_Y	0.0087	0.3303	0.4267	0.8419
		g_Z	19.5569	-0.6721	-0.5199	0.5272

Table S8: Relative angles between the main anisotropy axes in different doublet states of the Dy3 center from Dy_3 triangle.

KD	1	2	3	4	5	6	7	8
1	0.00	43.65	37.03	83.52	84.42	72.93	57.41	58.39
2	43.65	0.00	18.47	74.90	53.72	64.68	67.11	72.37
3	37.03	18.47	0.00	65.77	68.54	70.92	48.81	82.57
4	83.52	74.90	65.77	0.00	75.01	46.19	48.38	32.74
5	84.42	53.72	68.54	75.01	0.00	31.01	73.95	86.19
6	72.93	64.68	70.92	46.19	31.01	0.00	87.94	63.12
7	57.41	67.11	48.81	48.38	73.95	87.94	0.00	64.59
8	58.39	72.37	82.57	32.74	86.19	63.12	64.59	0.00

Table S9: CASSCF (spin-free) and RASSI (spin-orbit) energies (cm^{-1}) for Tb1-Tb3 centers in Tb_3 molecule.

Spin Multiplicity	CASSCF			RASSI		
	1	2	3	1	2	3
7	0.0	0.0	0.0	0.000	0.000	0.000
	92.0	38.8	157.6	0.131	0.029	0.090
	103.5	195.8	188.2	22.579	142.214	106.225
	162.5	345.2	282.1	26.390	144.587	107.496
	310.1	526.8	495.2	59.529	235.247	171.446
	578.3	677.1	631.0	80.723	242.520	178.386
	674.6	819.9	685.3	100.832	292.435	214.780
5	25644.3	25699.8	25706.9	148.959	330.380	235.191
	25646.3	25709.9	25717.0	154.370	356.951	242.988
	25723.0	25818.2	25798.4	275.726	390.849	275.819
	25739.4	25876.2	25854.7	279.138	402.843	277.512
	25753.8	25905.0	25868.1	347.074	623.244	485.772
	29132.8	29238.4	29263.5	348.359	623.458	485.818
	29141.1	29243.1	29263.7	2095.135	2227.202	2153.189
	29164.6	29258.4	29314.4	2097.977	2232.951	2175.865
	29172.8	29264.8	29315.6	2127.122	2259.951	2194.253
	29236.3	29337.7	29356.9	2150.358	2288.122	2219.502
	…	…	…	2193.445	2326.271	2249.338
3	47102.5	47199.8	47192.0	2258.577	2384.119	2301.420
	47104.6	47205.8	47198.8	2288.914	2397.278	2312.073
	47112.1	47219.5	47208.2	2297.216	2473.503	2387.739
	48215.5	48304.1	48269.4	2302.357	2486.533	2392.897
	48216.6	48305.1	48269.5	2348.493	2540.629	2439.139
	48231.2	48328.0	48295.2	2365.063	2554.125	2442.956
	48239.1	48328.8	48295.6	3446.275	3577.850	3478.595
	48252.9	48344.5	48349.7	3452.561	3636.194	3566.922
	48275.2	48345.6	48352.0	3517.782	3648.473	3603.186
	48279.8	48374.2	48365.6	3534.211	3688.533	3650.930
	…	…	…	3565.813	3717.299	3655.089
1	56773.3	56875.8	56871.4	3593.375	3780.941	3686.517
	57535.1	57590.5	57612.5	3683.947	3809.001	3715.080
	57536.5	57590.7	57613.0	3701.164	3840.996	3756.333
	57554.2	57638.3	57637.7	3722.555	3894.382	3774.043
	57555.4	57638.3	57638.8	4539.100	4676.831	4624.012
	57588.5	57683.2	57687.1	4561.129	4700.755	4644.695
	57592.5	57684.3	57687.8	4574.003	4720.701	4654.673
	57602.6	57713.9	57716.3	4622.909	4739.823	4686.317
	57624.1	57714.6	57718.5	4643.222	4814.801	4717.431
	57637.1	57763.0	57747.1	4662.720	4835.722	4735.595
	…	…	…	4676.932	4863.014	4759.839
				5242.003	5369.507	5329.227

	5323.938	5458.651	5371.789
	5333.295	5520.962	5422.958
	5439.980	5600.705	5490.967
	5512.622	5689.231	5557.024
	5738.918	5860.734	5790.181
	5885.420	6045.610	5939.565
	5933.952	6170.127	6047.993
	6093.551	6284.161	6177.464
	23481.562	23607.903	23550.890
	…	…	…

Table S10 Comparative g_X, g_Y, g_Z for 8 lowest Kramers doublets of the Tb1-Tb3 centers of the Tb_3 molecule.

ID		1		2		3	
		E (cm^{-1})	g	E (cm^{-1})	g	E (cm^{-1})	g
1	g_X		0.000		0.000		0.000
	g_Y	0.000	0.000	0.000	0.000	0.000	0.000
	g_Z	0.131	16.833	0.029	17.839	0.090	17.837
2	g_X		0.000		0.000		0.000
	g_Y	22.579	0.000	142.214	0.000	106.225	0.000
	g_Z	26.390	14.229	144.587	14.415	107.496	14.711
3	g_X		0.000		0.000		0.000
	g_Y	59.529	0.000	235.247	0.000	171.446	0.000
	g_Z	80.723	10.958	242.520	10.822	178.386	11.853
4		100.832	---	292.435	---	214.780	---
5	g_X		0.000		0.000		0.000
	g_Y	148.959	0.000	330.380	0.000	235.191	0.000
	g_Z	154.370	10.675	356.951	11.827	242.988	9.904
6	g_X		0.000		0.000		0.000
	g_Y	275.726	0.000	390.849	0.000	275.819	0.000
	g_Z	279.138	12.771	402.843	16.196	277.512	14.113
7	g_X		0.000		0.000		0.000
	g_Y	347.074	0.000	623.244	0.000	485.772	0.000
	g_Z	348.359	15.384	623.458	17.720	485.818	17.685

Table S11: *g*-factors and main magnetic axes (3×3) for 8 lowest Ising doublets of the Tb1 center from Tb_3 complex.

1	0.000	g_X	0.000	0.2174	-0.0449	0.9750
		g_Y	0.000	-0.5114	0.8455	0.1530
	0.131	g_Z	16.833	-0.8313	-0.5319	0.1608
2		g_X	0.000	0.0821	-0.4489	-0.8897
	22.579	g_Y	0.000	-0.8012	0.5012	-0.3268
	26.390	g_Z	14.229	-0.5927	-0.7397	0.3184
3		g_X	0.000	0.7879	-0.5836	-0.1964
	59.529	g_Y	0.000	0.2842	0.0615	0.9567
	80.723	g_Z	10.958	-0.5462	-0.8096	0.2144
4	100.832		---	---	---	---
5		g_X	0.000	0.4098	0.9121	0.0000
	148.959	g_Y	0.000	0.4493	-0.2019	-0.8702
	154.370	g_Z	10.675	-0.7937	0.3566	-0.4926
6		g_X	0.000	-0.6947	-0.5557	0.4565
	275.726	g_Y	0.000	0.1970	-0.7576	-0.6222
	279.138	g_Z	12.771	0.6916	-0.3423	0.6359
7		g_X	0.000	0.1530	-0.3204	-0.9348
	347.074	g_Y	0.000	-0.6540	-0.7419	0.1472
	348.359	g_Z	15.384	0.7407	-0.5889	0.3231

Table S12: Relative angles between the main anisotropy axes in different doublet states of the Tb1 center from Tb_3 complex.

1	0.00	20.36	23.17	66.99	73.10	75.49
2	20.36	0.00	7.67	88.12	84.34	84.93
3	23.17	7.67	0.00	87.75	87.96	81.87
5	66.99	88.12	87.75	0.00	10.12	16.81
6	73.10	84.34	87.96	10.12	0.00	23.15
7	75.49	84.93	81.87	16.81	23.15	0.00

Table S13: *g*-factors and main magnetic axes (3×3) for 8 lowest Ising doublets of the Tb2 center from Tb_3 complex.

1	0.000	g_X	0.000	-0.6164	-0.1168	-0.7787
	0.000	g_Y	0.000	0.7502	-0.3875	-0.5357
	0.029	g_Z	17.839	-0.2392	-0.9144	0.3265
2	142.214	g_X	0.000	0.8973	-0.4413	0.0000
		g_Y	0.000	-0.1495	-0.3039	-0.9408
	144.587	g_Z	14.415	0.4152	0.8442	-0.3387
3	235.247	g_X	0.000	0.3285	-0.7074	-0.6257
		g_Y	0.000	-0.7529	0.2037	-0.6257
	242.520	g_Z	10.822	-0.5701	-0.6767	0.4657
4	292.435		---	---	---	---
5	330.380	g_X	0.000	0.4345	0.7028	-0.5631
		g_Y	0.000	-0.4206	0.7112	0.5631
	356.951	g_Z	11.827	0.7963	-0.0078	0.6047
6	390.849	g_X	0.000	0.6535	0.7568	0.0000
		g_Y	0.000	-0.5667	0.4893	-0.6628
	402.843	g_Z	16.196	-0.5016	0.4331	0.7487
7	623.244	g_X	0.000	-0.3652	-0.9237	0.1150
		g_Y	0.000	0.5135	-0.3029	-0.8028
	623.458	g_Z	17.720	0.7764	-0.2341	0.5850

Table S14: Relative angles between the main anisotropy axes in different doublet states of the Tb2 center from Tb_3 complex.

1	0.00	10.90	24.87	89.19	88.19	77.33
2	10.90	0.00	15.00	78.05	79.78	74.81
3	24.87	15.00	0.00	80.38	70.02	89.33
5	89.19	78.05	80.38	0.00	87.14	13.10
6	88.19	79.78	70.02	87.14	0.00	86.97
7	77.33	74.81	89.33	13.10	86.97	0.00

Table S15: *g*-factors and main magnetic axes (3×3) for 8 lowest Kramers doublets of the Tb3 center from Tb_3 complex.

1	0.000	g_X	0.000	-0.0303	-0.0830	-0.9960
		g_Y	0.000	0.9391	-0.3434	0.0000
	0.090	g_Z	17.837	0.3421	0.9354	-0.0884
2		g_X	0.000	0.0257	-0.0364	-0.9990
	106.225	g_Y	0.000	-0.8815	0.4704	-0.0398
	107.496	g_Z	14.711	-0.4714	-0.8816	0.0199
3		g_X	0.000	0.8011	-0.5985	0.0000
	171.446	g_Y	0.000	0.0589	0.0789	-0.9951
	178.386	g_Z	11.853	0.5955	0.7972	0.0984
4	214.780		---	---	---	---
5		g_X	0.000	-0.0505	-0.9966	-0.0648
	235.191	g_Y	0.000	0.9986	-0.0495	-0.0158
	242.988	g_Z	9.904	0.0125	-0.0655	0.9977
6		g_X	0.000	-0.8362	0.5145	0.1898
	275.819	g_Y	0.000	0.4435	0.8380	-0.3177
	277.512	g_Z	14.113	0.3225	0.1814	0.9289
7		g_X	0.000	0.4999	0.8660	0.0000
	485.772	g_Y	0.000	0.2984	-0.1723	-0.9387
	485.818	g_Z	17.685	-0.8129	0.4693	-0.3446

Table S16: Relative angles between the main anisotropy axes in different doublet states of the Tb3 center from Tb_3 complex.

1	0.00	8.94	19.81	81.65	81.86	78.97
2	8.94	0.00	10.98	88.84	87.54	81.27
3	19.81	10.98	0.00	86.93	82.01	81.72
5	81.65	88.84	86.93	0.00	19.46	67.36
6	81.86	87.54	82.01	19.46	0.00	48.12
7	78.97	81.27	81.72	67.36	48.12	0.00