Supporting Information

An investigation into the magnetic interactions in a series of Dy₂ single-molecule magnets

Chaoyi Jin,[a, b] Xiao-Lei Li,[a] Zhiliang Liu,[b] Akseli Mansikkamäki,*[c] and Jinkui Tang*[a,d]

[a]State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, P. R. China
[b]College of Chemistry and Chemical Engineering, Inner Mongolia University, Hohhot 010021, China
[c]NMR Research Unit, University of Oulu, P.O. Box 8000, Finland
[d]School of Applied Chemistry and Engineering, University of Science and Technology of China, Hefei, 230026, P. R. China

Corresponding Authors

*E-mail: Akseli.Mansikkamaki@oulu.fi
*E-mail: tang@ciac.ac.cn

Table S1. Crystallographic data and structure refinement details of complexes **1**, **2** and **3**.

	1	**2**	**3**
mpirical formula	$C_{49}H_{65}ClDy_2F_3N_{11}O_{15}$	$C_{41}H_{51}Dy_2N_9O_{12}S$	$C_{41}H_{56}Cl_2Dy_2N_8O_{13}$
Formula Weight	1465.57	1218.97	1264.83
Temperature/K	173	173	173
Crystal system	monoclinic	monoclinic	monoclinic
Space group	$P2_1/c$	$P2_1/c$	$P2_1/c$
a/Å	17.6969(6)	14.5797(6)	10.7089(4)
b/Å	15.0291(5)	21.9434(9)	28.6894(12)
c/Å	22.5627(7)	15.8475(7)	15.6149(6)
α/°	90	90	90
β/°	112.191(2)	116.407(2)	105.994(2)
γ/°	90	90	90
Volume/Å^3	5556.5(3)	4541.0(3)	4611.7(3)
Z	4	4	4
ρ_{calc}g/cm^3	1.752	1.783	1.822
μ/mm^{-1}	15.409	18.436	18.816
reflns collected	32439	27644	30217
R_{int}	0.0627	0.0462	0.0461
GOF on F^2	1.029	1.029	1.048
*R_1, wR_2 [I>=2σ (I)]	0.0442, 0.1088	0.0323, 0.0755	0.0359, 0.0895
*R_1, wR_2 [all data]	0.0544, 0.1176	0.0415, 0.0804	0.0454, 0.0947

*$R_1=\Sigma||Fo|-|Fc||/\Sigma|Fo|$ for $Fo>2\sigma(Fo)$; $wR_2=(\Sigma w(Fo^2-Fc^2)^2/\Sigma(wFc^2)^2)^{1/2}$ all reflections, $w=1/[\sigma^2(Fo^2)+(0.1557P)^2]$ where P = $(Fo^2+2Fc^2)/3$

Table S2. Selected bond distances [Å] for complexes **1**, **2**, and **3**.

1		**2**		**3**	
Dy1-O1	2.527(4)	Dy1-O2	2.353(3)	Dy1-O1	2.199(4)
Dy1-O2	2.342(4)	Dy1-O3	2.346(3)	Dy1-O2	2.515(4)
Dy1-O3	2.194(4)	Dy1-O4	2.471(3)	Dy1-O3	2.489(4)
Dy1-O4	2.455(4)	Dy1-O5	2.223(3)	Dy1-O4	2.351(4)
Dy1-O7	2.415(4)	Dy1-O9	2.468(3)	Dy1-O5	2.476(4)
Dy1-O10	2.460(4)	Dy1-N1	2.491(4)	Dy1-O7	2.432(4)
Dy1-N1	2.509(5)	Dy1-N3	2.501(4)	Dy1-N2	2.500(4)
Dy1-N3	2.691(5)	Dy1-N8	2.633(4)	Dy1-N4	2.685(5)
Dy1-N5	2.545(5)	Dy1-N9	2.543(4)	Dy1-N5	2.528(5)
Dy2-O1	2.324(4)	Dy2-O1	2.338(3)	Dy2-Cl1	2.7644(16)
Dy2-O2	2.520(4)	Dy2-O3	2.540(3)	Dy2-O2	2.342(4)
Dy2-O5	2.183(4)	Dy2-O4	2.356(3)	Dy2-O4	2.558(4)
Dy2-O6	2.427(4)	Dy2-O6	2.247(3)	Dy2-O6	2.472(4)
Dy2-O8	2.456(4)	Dy2-O7	2.526(3)	Dy2-O8	2.212(4)
Dy2-O11	2.409(4)	Dy2-O8	2.463(3)	Dy2-O10	2.444(4)
Dy2-N6	2.502(5)	Dy2-N2	2.532(4)	Dy2-N1	2.525(5)
Dy2-N7	2.697(5)	Dy2-N5	2.576(4)	Dy2-N6	2.474(5)
Dy2-N8	2.502(5)	Dy2-N7	2.633(4)	Dy2-N7	2.652(5)

Table S3. Accurate geometry analysis by SHAPE 2.0 software.

complex	1	2	3
Dy1	1.427	1.428	1.740
Dy2	1.373	2.041	1.861

Table S4. Characteristic dynamic parameters for **1**, **2** and **3**.

	1	2 FR	2 SR	3
U_{eff}/K	90.57	72.03	54.02	81.39
τ_0 (Orbach)	3.01×10^{-7}	1.19×10^{-8}	3.06×10^{-6}	9.18×10^{-8}
U_{eff}/K	87.25	78.81	53.41	81.39
τ_0 (fit)	2.48×10^{-7}	5.38×10^{-9}	3.75×10^{-6}	1.91×10^{-7}
C $(s^{-1}K^{-n})$	2.59	11.72	5.05	5.05
n	1.72	1.52	1.07	1.07
α	0.03-0.23	0.17-0.19	< 0.08	0.08-0.2
τ (1.9–10 K)	0.001–0.11	10^{-5}–0.027	0.001–0.094	10^{-4}–0.07

Table S5. Properties of the eight lowest Kramers doublets (KDs) of each Dy^{III} ion in **1**.

	Dy1					Dy2			
	E/cm^{-1}	g_x	g_y	g_z	θ_a	E/cm^{-1}	g_x	g_y	g_z	θ_a
KD 1	0	0.0302	0.0643	19.6051	0.0°	0	0.0257	0.0549	19.5943	0.0°
KD 2	147	1.1035	3.1208	14.4679	7.6°	136	1.1345	2.5965	15.2341	8.2°
KD 3	200	0.7856	2.4868	12.1192	138.3°	190	0.7057	3.2518	13.1546	57.4°
KD 4	283	1.3508	1.6048	11.9522	21.2°	254	0.8310	4.2267	10.6270	154.8°
KD 5	351	3.9066	5.7331	10.3442	46.1°	316	3.1668	4.2837	11.8896	128.1°
KD 6	385	1.6824	2.7985	15.9031	92.5°	358	8.5726	6.4501	1.4961	40.4°
KD 7	419	0.4226	0.8814	16.6808	99.9°	375	2.0992	5.5468	14.1902	95.6°
KD 8	562	0.0163	0.0394	19.6694	104.5°	498	0.0309	0.0496	19.5169	74.3°

[a] The angle between the principal magnetic axis of the doublet and that of the ground doublet.

Table S6. Properties of the eight lowest Kramers doublets (KDs) of each Dy^{III} ion in **2**.

	Dy1					Dy2			
	E/cm^{-1}	g_x	g_y	g_z	θ_a	E/cm^{-1}	g_x	g_y	g_z	θ_a
KD 1	0	0.0455	0.0662	19.4346	0.0°	0	0.3067	1.1655	17.3799	0.0°
KD 2	110	1.0438	2.0482	15.3855	7.4°	69	0.4876	1.0211	13.5709	18.8°
KD 3	166	1.6525	3.6387	12.2049	120.7°	109	0.2929	0.5279	17.7239	36.6°
KD 4	224	8.3467	6.8996	0.6906	97.1°	141	3.3107	5.7153	9.3171	124.7°
KD 5	280	9.8743	5.9968	1.5192	153.9°	202	2.5843	4.0092	10.5912	73.0°
KD 6	318	3.2537	5.6318	10.7723	103.8°	289	0.1876	0.5357	16.3934	56.1°
KD 7	373	1.2474	2.0281	15.6755	92.3°	348	0.2179	0.2705	17.5037	94.5°
KD 8	485	0.0215	0.0594	19.4335	79.6°	477	0.0225	0.0532	19.1894	91.9°

[a] The angle between the principal magnetic axis of the doublet and that of the ground doublet.

Table S7. Properties of the eight lowest Kramers doublets (KDs) of each Dy^{III} ion in **3**.

	Dy1					Dy2			
	E/cm^{-1}	g_x	g_y	g_z	θ_a	E/cm^{-1}	g_x	g_y	g_z	θ_a
KD 1	0	0.0305	0.0612	19.6699	0.0°	0	0.0259	0.0625	19.5576	0.0°
KD 2	148	1.0409	3.2962	14.2075	20.3°	131	0.9223	1.9579	15.8873	18.8°
KD 3	201	1.4058	2.1405	10.8921	11.0°	197	1.3266	2.8546	13.1811	120.6°
KD 4	288	0.7102	3.1154	11.6462	23.1°	258	0.9756	4.0923	10.1631	18.6°
KD 5	366	8.5943	7.7417	4.1465	43.5°	325	9.1623	8.8510	3.2952	141.1°
KD 6	427	1.4938	2.3520	14.1464	98.4°	372	1.3433	4.9926	11.9116	83.4°
KD 7	460	0.0422	0.1539	18.0354	85.3°	401	1.9423	5.8174	12.6832	108.2°
KD 8	568	0.0260	0.0618	19.5884	78.1°	482	0.1364	0.4062	19.1253	76.8°

[a] The angle between the principal magnetic axis of the doublet and that of the ground doublet.
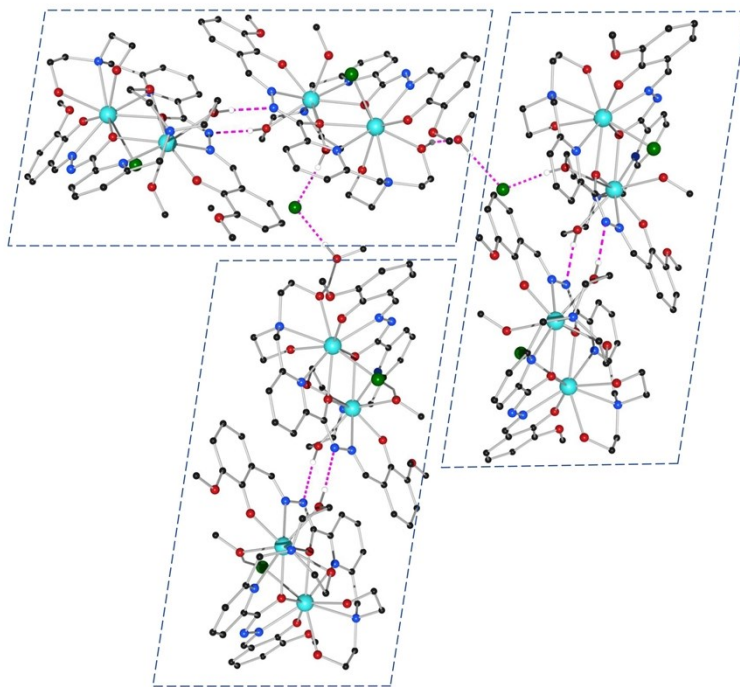
Fig. S1. Multiple intermolecular hydrogen bondings (pink line) in **3**.
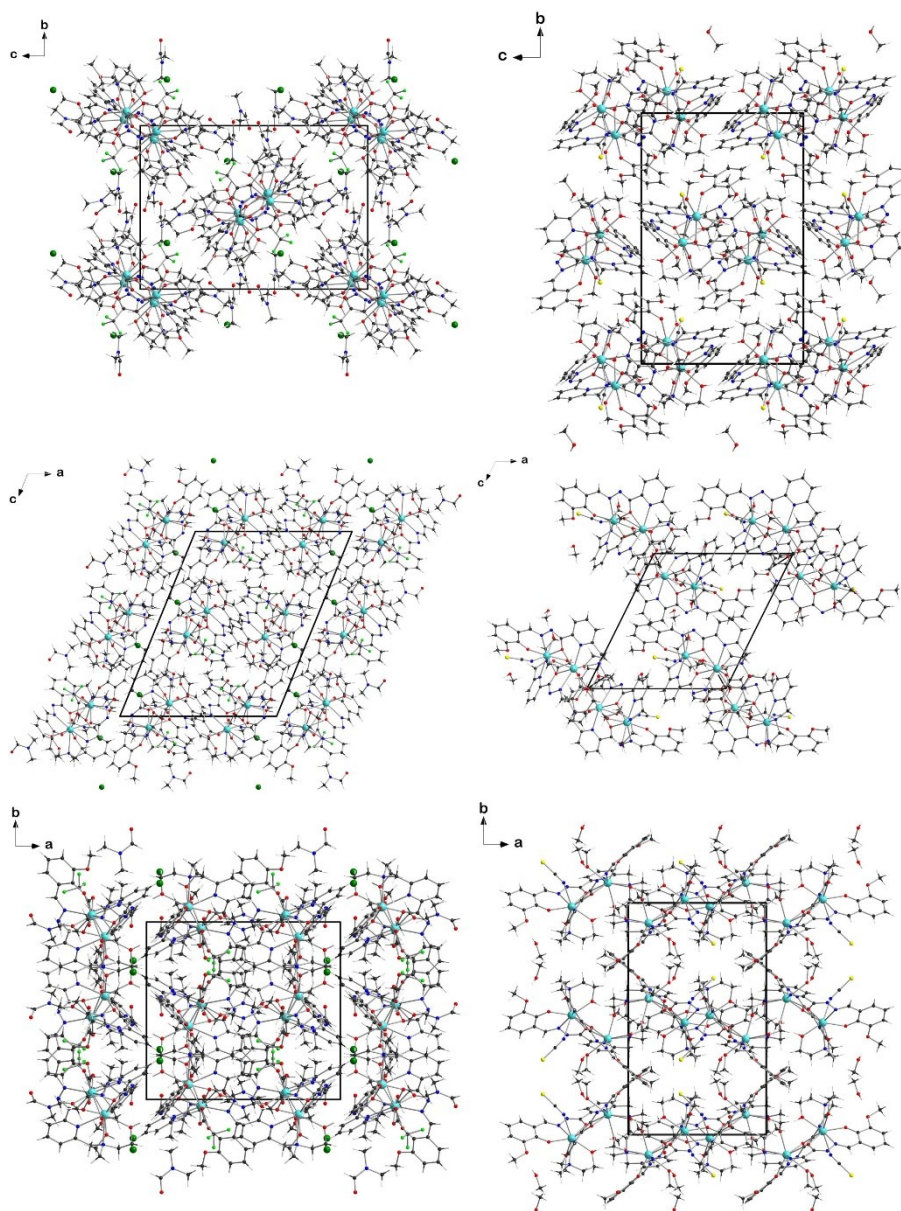
Fig. S2. Packing arrangement along the crystallographic a (top), b (middle), and c-axis (bottom) for complexes **1** (left) and **2** (right). Color code: turquiose, Dy; blue, N; red, O; bright green, F; green, Cl; yellow, S; gray, C; white (wires / sticks), H.
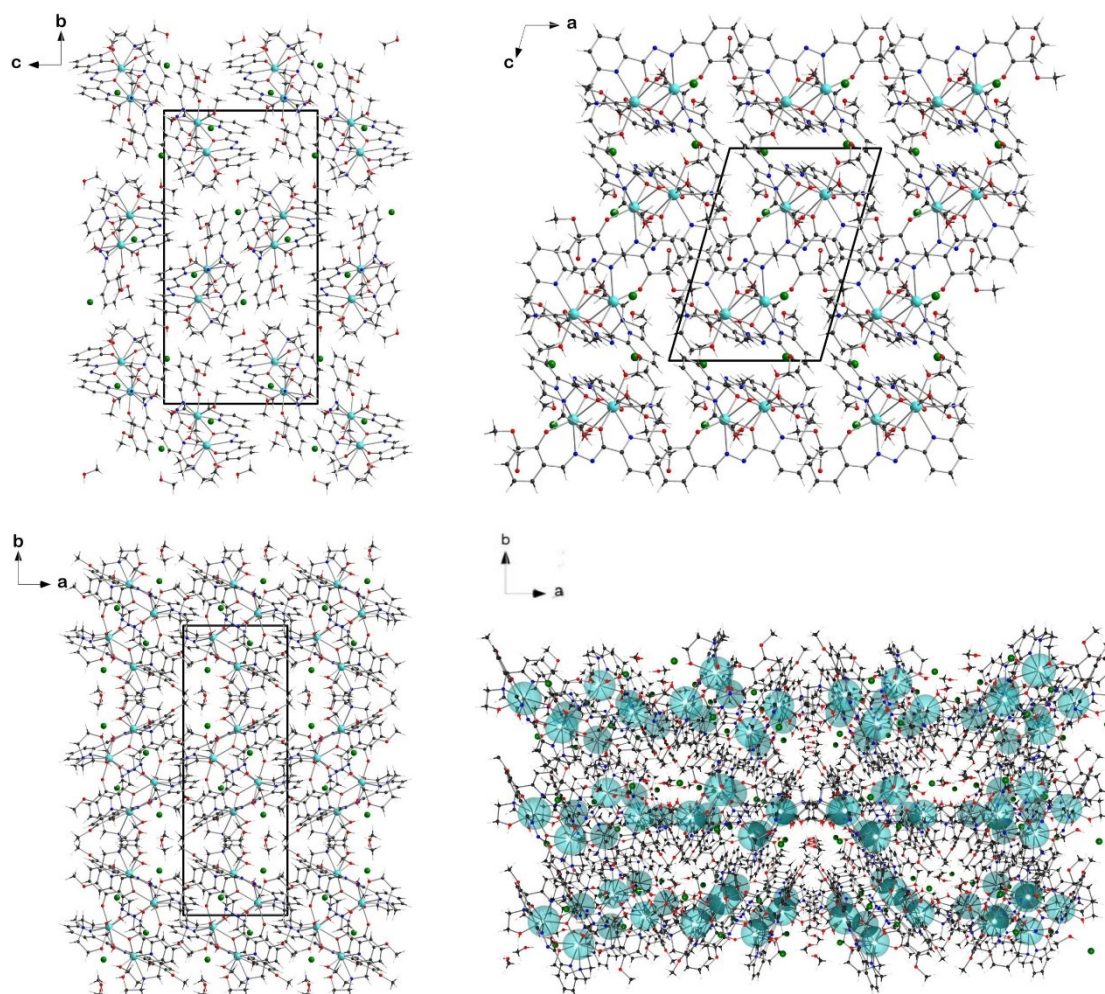
Fig. S3. Packing arrangement along the crystallographic a (top right), b (top left), and c-axis (bottom left in normal style and bottom right in central projection style) for complexes **3**. Color code: turquiose, Dy; blue, N; red, O; bright green, F; green, Cl; gray, C; white (wires / sticks), H.
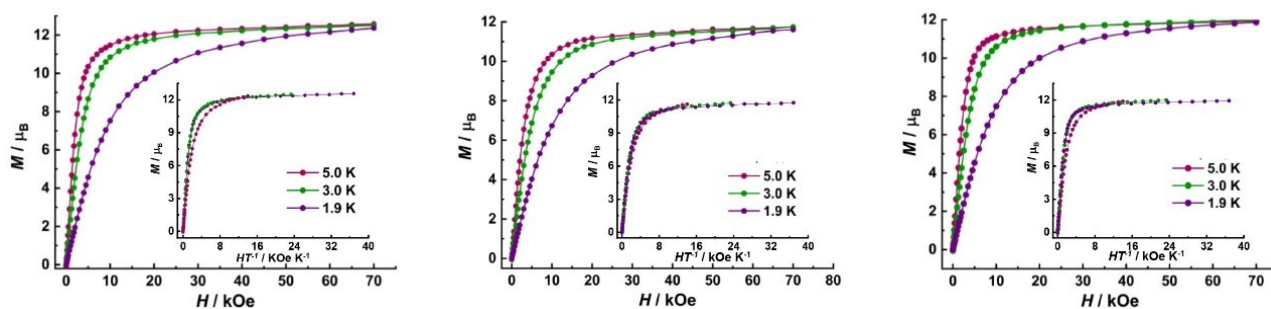
Fig. S4. Field dependences of magnetization in the field range 0−70 kOe and at the temperature range of 1.9−5.0 K. Insets: Plots of the reduced magnetization M vs. H/T for complexes **1** (left), **2** (middle), **3** (right). The solid lines are just guides for the eye.
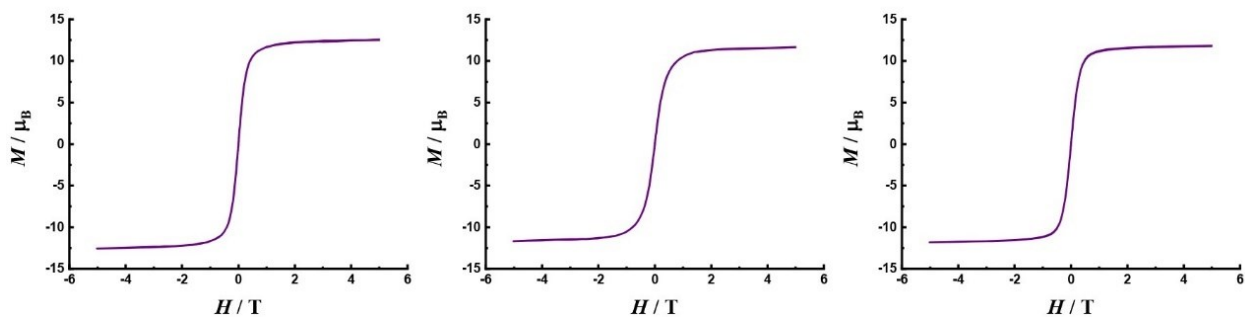
Fig. S5. Magnetization plots of complexes **1−3** show no observable hysteresis at 1.9 K.
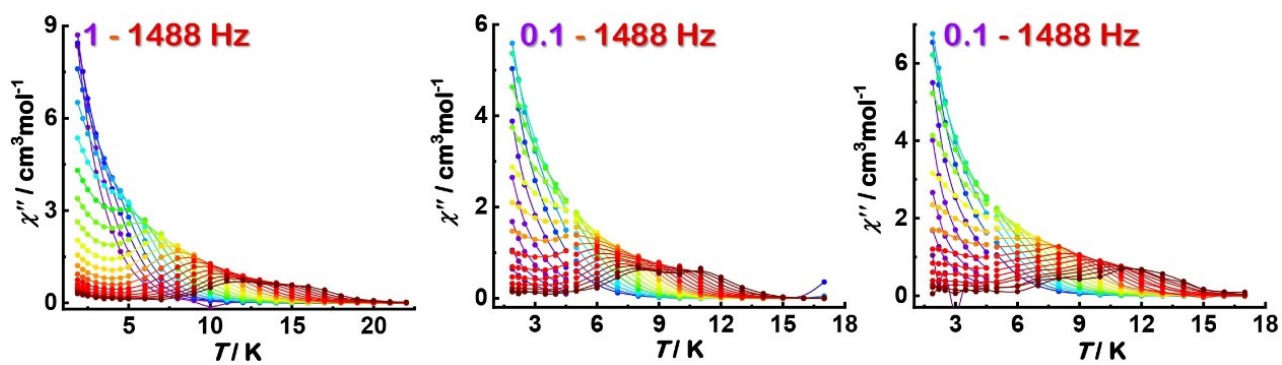
Fig. S6. Temperature dependence of the out-of-phase ac susceptibilities under zero dc field for three complexes (**1−3** from left to right).

Fig. S7. 3D graph of χ'' vs. frequency v in logarithmic scale in the temperature range 1.9−17 K for **2**.

Additional computational data for the supporting information

Table S8. The ab initio CF parameters for the two Dy[III] ions in **1** given in Iwahara-Chibotaru notation.[1,2]

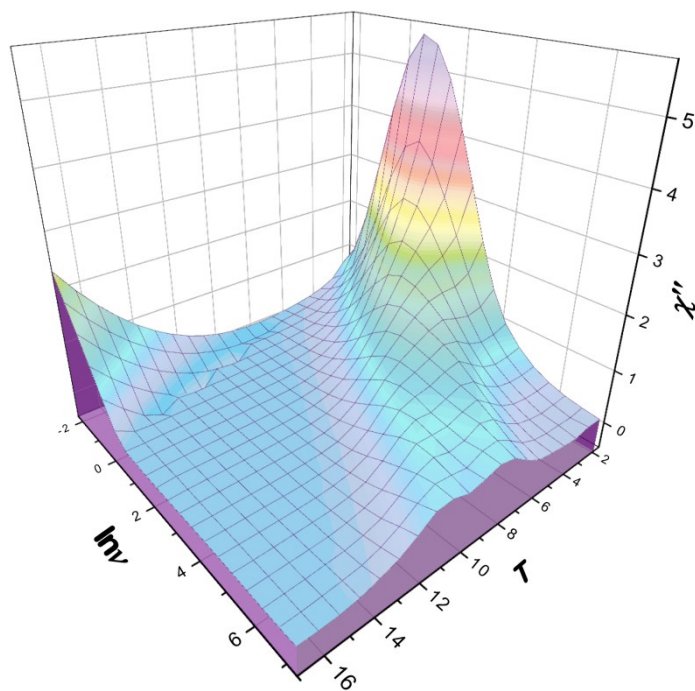| k	q [a]	Re(Bkq)	Im(Bkq)	|Bkq|	Re(Bkq)	Im(Bkq)	|Bkq|
			Dy1			Dy2	
2	0	–227.527436	0.000000	227.527436	–205.934140	0.000000	205.934140
2	1	37.943018	1.297149	37.965184	–30.114491	–6.432165	30.793754
2	2	95.856761	24.053742	98.828646	71.211407	14.380530	72.648910
4	0	–52.620590	0.000000	52.620590	–48.882603	0.000000	48.882603
4	1	–21.338139	6.684112	22.360535	19.460838	–2.548276	19.626970
4	2	9.677035	1.557041	9.801499	8.224756	6.873201	10.718559
4	3	7.311721	21.379531	22.595256	–8.511394	–23.357174	24.859634
4	4	–14.626035	28.553557	32.081560	–9.402747	26.655346	28.265157
6	0	–3.748779	0.000000	3.748779	–1.140073	0.000000	1.140073
6	1	–2.571494	–8.606822	8.982759	2.976646	7.656332	8.214611
6	2	–3.897546	5.871811	7.047626	0.478664	2.003315	2.059707
6	3	–3.888340	–20.800298	21.160614	8.585159	21.695533	23.332405
6	4	–8.861874	9.713114	13.148285	0.538819	9.573391	9.588542
6	5	–10.224231	–11.699037	15.537129	14.314347	10.961486	18.029274
6	6	4.185059	11.745306	12.468638	9.295960	11.332696	14.657587
8	0	–0.111543	0.000000	0.111543	–0.137407	0.000000	0.137407
8	1	0.108400	0.029944	0.112459	–0.104400	–0.011873	0.105073
8	2	–0.044884	0.106618	0.115681	–0.048114	0.108972	0.119121
8	3	0.052065	0.226508	0.232415	–0.049600	–0.183071	0.189671
8	4	0.114198	0.008404	0.114507	0.049259	–0.002847	0.049341
8	5	0.029149	–0.010487	0.030979	–0.009181	0.017237	0.019530
8	6	–0.009341	0.015212	0.017851	0.013403	0.001403	0.013476
8	7	–0.066576	–0.044132	0.079875	0.068610	0.010626	0.069428
8	8	0.008595	0.031477	0.032630	0.023623	0.005265	0.024203
10	0	0.022888	0.000000	0.022888	0.016172	0.000000	0.016172
10	1	0.008718	0.006522	0.010888	–0.012299	–0.014077	0.018692
10	2	0.028527	–0.025780	0.038450	0.010752	–0.018162	0.021106
10	3	–0.001660	0.043368	0.043399	–0.014777	–0.042884	0.045358
10	4	0.005848	–0.008966	0.010705	–0.007836	–0.005955	0.009842
10	5	0.004487	0.007137	0.008430	–0.012108	–0.003955	0.012737
10	6	0.005612	–0.006162	0.008335	0.005876	–0.008401	0.010252
10	7	0.001190	–0.007565	0.007658	–0.000371	0.003884	0.003902
10	8	–0.001587	–0.011552	0.011661	–0.001470	–0.007980	0.008114

10	9	0.012838	−0.003037	0.013192	−0.011683	0.007471	0.013867
10	10	−0.016997	−0.007194	0.018457	−0.017596	0.002272	0.017742
12	0	−0.002921	0.000000	0.002921	−0.004039	0.000000	0.004039
12	1	0.000417	0.003046	0.003074	−0.000507	−0.000652	0.000826
12	2	−0.002724	−0.000435	0.002759	−0.003292	0.000324	0.003308
12	3	0.001496	0.001489	0.002111	−0.001207	−0.000139	0.001215
12	4	−0.000992	−0.000077	0.000995	−0.000849	0.000437	0.000955
12	5	0.000890	−0.000125	0.000899	−0.000364	0.000214	0.000423
12	6	−0.000808	−0.000263	0.000850	−0.000778	0.000259	0.000820
12	7	0.000446	0.000777	0.000896	−0.000449	−0.000039	0.000450
12	8	−0.000524	0.000335	0.000622	−0.000486	0.001028	0.001138
12	9	0.000990	−0.000110	0.000996	−0.000516	0.000868	0.001010
12	10	−0.000485	0.000454	0.000664	−0.000059	0.001050	0.001051
12	11	0.000442	−0.000762	0.000881	0.000054	0.001215	0.001216
12	12	−0.000427	0.000336	0.000543	−0.000148	0.000723	0.000738
14	0	−0.000009	0.000000	0.000009	0.000002	0.000000	0.000002
14	1	0.000000	−0.000002	0.000002	0.000003	0.000008	0.000008
14	2	−0.000008	−0.000007	0.000011	0.000002	−0.000008	0.000008
14	3	−0.000012	−0.000020	0.000024	0.000006	0.000007	0.000009
14	4	0.000025	−0.000014	0.000028	0.000014	−0.000016	0.000021
14	5	−0.000011	0.000002	0.000011	0.000003	−0.000005	0.000006
14	6	0.000000	−0.000001	0.000001	−0.000002	−0.000003	0.000004
14	7	0.000000	−0.000004	0.000004	0.000002	0.000002	0.000002
14	8	0.000004	−0.000002	0.000004	0.000003	−0.000003	0.000004
14	9	−0.000004	0.000002	0.000004	0.000001	−0.000004	0.000004
14	10	0.000002	−0.000001	0.000003	−0.000001	−0.000003	0.000003
14	11	−0.000001	0.000001	0.000001	−0.000001	−0.000002	0.000002
14	12	0.000000	−0.000001	0.000001	0.000000	−0.000001	0.000001
14	13	0.000000	0.000001	0.000001	−0.000001	0.000000	0.000001
14	14	0.000003	0.000000	0.000003	0.000002	−0.000001	0.000002

a The parameters are only listed for positive q. Values with negative q are related by Bk–q = (–1)qBkq*.

Table S9. The ab initio CF parameters for the two Dy^{III} ions in **2** given in Iwahara-Chibotaru notation.[1,2]

		Dy1			Dy2		
k	q[a]	Re(Bkq)	Im(Bkq)	|Bkq|	Re(Bkq)	Im(Bkq)	|Bkq|
2	0	−197.206778	0.000000	197.206778	−157.473526	0.000000	157.473526
2	1	8.687197	−18.898049	20.799126	39.160732	7.696743	39.909934
2	2	55.587897	−67.085513	87.123363	91.974126	−64.914518	112.575017
4	0	−33.489146	0.000000	33.489146	27.690717	0.000000	27.690717

4	1	−6.556341	2.581691	7.046328	8.566342	−12.851124	15.444533
4	2	−0.496744	−4.812062	4.837633	11.865698	11.644659	16.625068
4	3	−6.327684	−22.271133	23.152601	24.135383	−3.528694	24.391974
4	4	−23.655787	−16.142671	28.638822	−48.151363	4.430567	48.354769
6	0	1.228650	0.000000	1.228650	−24.439992	0.000000	24.439992
6	1	−4.725096	7.691051	9.026561	−17.025467	−7.669450	18.673163
6	2	−0.274176	−9.712037	9.715906	14.069872	4.157616	14.671301
6	3	−4.622857	23.352693	23.805862	4.111050	−3.183018	5.199263
6	4	−3.500001	−5.457376	6.483283	−13.897193	−4.976333	14.761296
6	5	−5.006070	20.067136	20.682134	1.202131	−6.790529	6.896116
6	6	4.541673	−11.120082	12.011786	−7.912386	15.408624	17.321419
8	0	−0.051198	0.000000	0.051198	0.322255	0.000000	0.322255
8	1	0.042697	0.002923	0.042797	0.020055	−0.190718	0.191770
8	2	−0.046807	0.095966	0.106772	−0.145334	0.031162	0.148637
8	3	0.010626	−0.199535	0.199818	0.380822	−0.104595	0.394925
8	4	0.076645	0.008662	0.077133	−0.034580	0.083157	0.090060
8	5	0.023311	−0.050700	0.055802	−0.003867	0.066959	0.067070
8	6	0.002624	0.012107	0.012388	0.049679	−0.086840	0.100046
8	7	0.002222	0.059129	0.059171	−0.038150	−0.002235	0.038215
8	8	0.007313	−0.032663	0.033471	0.012698	0.007898	0.014954
10	0	0.009827	0.000000	0.009827	0.017438	0.000000	0.017438
10	1	0.011347	−0.020234	0.023199	−0.018538	−0.009985	0.021056
10	2	0.010503	0.006824	0.012525	0.002633	−0.008734	0.009122
10	3	−0.005624	−0.033648	0.034114	−0.016836	0.016250	0.023399
10	4	−0.006705	0.005347	0.008576	0.007188	0.002414	0.007583
10	5	−0.002645	−0.008725	0.009117	−0.001411	0.004448	0.004666
10	6	0.010309	−0.000382	0.010316	−0.003813	−0.005265	0.006501
10	7	0.008785	0.001901	0.008989	−0.000395	−0.009456	0.009464
10	8	0.005763	−0.000043	0.005763	0.003377	0.002511	0.004209
10	9	0.011151	−0.006632	0.012974	−0.002592	0.018672	0.018851
10	10	−0.006280	0.010089	0.011884	0.016183	−0.031251	0.035192
12	0	−0.003484	0.000000	0.003484	−0.001601	0.000000	0.001601
12	1	−0.000186	−0.001533	0.001544	0.007840	0.000260	0.007844
12	2	−0.003481	0.000515	0.003518	−0.002579	−0.001337	0.002906
12	3	0.000946	−0.000034	0.000947	−0.003529	0.000494	0.003563
12	4	−0.001258	0.000399	0.001320	0.001602	0.000094	0.001604
12	5	0.001498	0.000432	0.001559	−0.000084	0.000341	0.000351
12	6	−0.000823	−0.000039	0.000824	0.000063	−0.001107	0.001109

12	7	0.000279	–0.000371	0.000464	–0.001240	0.000485	0.001331
12	8	–0.001092	–0.000416	0.001168	0.000489	0.000117	0.000503
12	9	0.000720	0.000060	0.000722	–0.000184	0.000608	0.000635
12	10	–0.001112	–0.000499	0.001219	0.000401	–0.000567	0.000694
12	11	0.000890	0.000675	0.001117	0.000382	0.000353	0.000520
12	12	–0.000359	–0.000316	0.000478	–0.000528	–0.000782	0.000943
14	0	0.000001	0.000000	0.000001	0.000010	0.000000	0.000010
14	1	0.000005	0.000008	0.000010	–0.000036	0.000001	0.000036
14	2	0.000005	0.000006	0.000007	0.000007	–0.000013	0.000014
14	3	0.000000	0.000004	0.000004	–0.000005	0.000016	0.000017
14	4	0.000023	0.000008	0.000024	–0.000001	–0.000007	0.000007
14	5	–0.000007	–0.000013	0.000015	0.000007	–0.000003	0.000008
14	6	–0.000001	0.000005	0.000005	0.000002	0.000002	0.000002
14	7	0.000002	0.000000	0.000002	–0.000002	0.000000	0.000002
14	8	0.000005	0.000001	0.000005	–0.000002	0.000001	0.000002
14	9	–0.000004	–0.000003	0.000005	0.000000	–0.000003	0.000003
14	10	0.000003	0.000000	0.000003	0.000000	0.000002	0.000002
14	11	–0.000001	–0.000001	0.000002	0.000000	–0.000001	0.000001
14	12	0.000001	0.000000	0.000001	0.000002	0.000000	0.000002
14	13	–0.000001	–0.000002	0.000002	0.000001	–0.000002	0.000002
14	14	0.000001	–0.000002	0.000002	–0.000004	0.000003	0.000005

a The parameters are only listed for positive q. Values with negative q are related by Bk–q = (–1)qBkq*.

Table S10. The ab initio CF parameters for the two Dy^{III} ions in **3** given in Iwahara-Chibotaru notation.[1, 2]

| | | Dy1 | | | Dy2 | | |
k	q [a]	Re(Bkq)	Im(Bkq)	|Bkq|	Re(Bkq)	Im(Bkq)	|Bkq|
2	0	–242.578613	0.000000	242.578613	–204.284464	0.000000	204.284464
2	1	–23.791257	–6.537954	24.673240	–16.433649	–27.985058	32.453479
2	2	106.339326	–3.697890	106.403603	75.406177	11.024038	76.207749
4	0	–49.665446	0.000000	49.665446	–55.101500	0.000000	55.101500
4	1	18.367594	–9.740517	20.790530	6.109510	4.952908	7.864948
4	2	8.717286	7.907171	11.769215	6.946280	6.532216	9.535232
4	3	–2.465684	–29.413430	29.516596	2.803538	–21.046166	21.232073
4	4	–15.600705	30.902922	34.617518	–19.761656	22.761631	30.143240
6	0	–8.343477	0.000000	8.343477	–0.131228	0.000000	0.131228
6	1	–3.342524	8.192830	8.848442	5.375274	10.742991	12.012719
6	2	–12.290044	7.580891	14.440051	–2.609126	10.568969	10.886259
6	3	–1.690696	14.006351	14.108024	10.589220	18.831554	21.604606
6	4	–13.152667	11.555996	17.508104	–4.909646	8.226819	9.580458

6	5	8.880950	14.378021	16.899667	14.200514	12.385108	18.842651
6	6	0.514401	12.086749	12.097691	9.846583	9.151488	13.442654
8	0	−0.191037	0.000000	0.191037	0.002064	0.000000	0.002064
8	1	0.049842	−0.090616	0.103419	−0.060196	−0.020994	0.063752
8	2	−0.005679	0.068629	0.068863	−0.008349	−0.114068	0.114373
8	3	0.022760	−0.187512	0.188888	−0.093972	−0.156125	0.182225
8	4	0.146419	−0.002330	0.146438	0.086778	−0.053267	0.101823
8	5	−0.047302	−0.040480	0.062258	−0.012767	−0.009358	0.015829
8	6	−0.030606	0.021146	0.037201	0.007417	0.012591	0.014613
8	7	0.058704	0.064042	0.086876	0.051124	0.038290	0.063873
8	8	0.022647	0.043187	0.048765	0.038253	0.022239	0.044248
10	0	0.044654	0.000000	0.044654	0.008493	0.000000	0.008493
10	1	0.013897	0.014811	0.020310	−0.018897	−0.023989	0.030538
10	2	0.048895	−0.025954	0.055356	0.018819	−0.013643	0.023244
10	3	0.009548	−0.017758	0.020162	−0.013887	−0.048349	0.050304
10	4	0.010302	−0.020432	0.022882	−0.006054	−0.003859	0.007179
10	5	−0.005058	−0.006714	0.008406	−0.006778	−0.005961	0.009026
10	6	0.000841	−0.005636	0.005699	0.003082	−0.004247	0.005247
10	7	−0.002370	0.009016	0.009322	−0.004880	0.012294	0.013227
10	8	0.002320	−0.018485	0.018630	0.002424	−0.005010	0.005566
10	9	−0.009708	0.000729	0.009735	−0.010440	0.005374	0.011742
10	10	−0.014742	−0.013818	0.020206	−0.018200	−0.003205	0.018480
12	0	0.000998	0.000000	0.000998	−0.004075	0.000000	0.004075
12	1	−0.001036	−0.004136	0.004264	−0.000909	−0.003414	0.003533
12	2	−0.000013	−0.000740	0.000740	−0.004072	0.000675	0.004128
12	3	−0.001816	−0.002478	0.003072	−0.001572	−0.000604	0.001684
12	4	−0.000494	−0.000514	0.000712	−0.001698	0.000162	0.001706
12	5	−0.001042	−0.000224	0.001066	−0.001320	0.001054	0.001689
12	6	0.000082	−0.000753	0.000757	−0.000487	0.000042	0.000489
12	7	−0.000111	−0.000684	0.000693	−0.000449	−0.000524	0.000690
12	8	−0.000412	−0.000206	0.000461	−0.000233	0.000787	0.000821
12	9	−0.001240	−0.000360	0.001291	−0.000685	0.000513	0.000856
12	10	−0.000726	0.000245	0.000766	−0.000176	0.000949	0.000965
12	11	−0.000772	0.000516	0.000928	0.000112	0.001121	0.001127
12	12	−0.000514	0.000061	0.000518	0.000022	0.000612	0.000612
14	0	−0.000011	0.000000	0.000011	−0.000014	0.000000	0.000014
14	1	−0.000002	−0.000011	0.000011	−0.000005	0.000008	0.000009
14	2	−0.000015	−0.000002	0.000015	0.000000	−0.000010	0.000010

14	3	0.000020	0.000029	0.000036	0.000015	0.000019	0.000024
14	4	0.000013	0.000002	0.000013	0.000030	–0.000026	0.000040
14	5	0.000013	0.000010	0.000017	0.000009	–0.000014	0.000017
14	6	–0.000002	0.000006	0.000007	–0.000004	–0.000001	0.000004
14	7	0.000000	0.000004	0.000004	–0.000001	0.000004	0.000004
14	8	0.000002	0.000001	0.000003	0.000003	–0.000005	0.000006
14	9	0.000004	0.000001	0.000004	0.000002	–0.000005	0.000005
14	10	0.000003	0.000001	0.000003	0.000002	–0.000003	0.000003
14	11	0.000000	–0.000001	0.000001	0.000000	–0.000001	0.000001
14	12	0.000001	–0.000001	0.000002	0.000000	–0.000001	0.000001
14	13	–0.000001	0.000000	0.000001	–0.000002	–0.000001	0.000002
14	14	0.000003	0.000000	0.000003	0.000002	–0.000001	0.000002

a The parameters are only listed for positive q. Values with negative q are related by $B_{k-q} = (-1)^q B_{kq}^*$.

Table S11. Squared projections of the the CF eigenstates corresponding to the eigth lowest Kramers doublets (KDs) of **1** onto angular momentum eigenstates with total angular momentum $J = 15/2$ and various angular momentum projections M

	Dy1							
M	KD 1		KD 2		KD 3		KD 4	
–15/2	0.074	0.891	0.001	0.000	0.006	0.004	0.000	0.016
–13/2	0.000	0.001	0.237	0.491	0.001	0.102	0.004	0.098
–11/2	0.001	0.015	0.009	0.000	0.153	0.212	0.006	0.323
–9/2	0.001	0.007	0.039	0.079	0.010	0.023	0.008	0.319
–7/2	0.001	0.007	0.005	0.001	0.083	0.117	0.002	0.032
–5/2	0.000	0.002	0.022	0.043	0.016	0.049	0.009	0.076
–3/2	0.000	0.000	0.011	0.032	0.035	0.065	0.015	0.051
–1/2	0.000	0.000	0.029	0.002	0.072	0.053	0.026	0.014
+1/2	0.000	0.000	0.002	0.029	0.053	0.072	0.014	0.026
+3/2	0.000	0.000	0.032	0.011	0.065	0.035	0.051	0.015
+5/2	0.002	0.000	0.043	0.022	0.049	0.016	0.076	0.009
+7/2	0.007	0.001	0.001	0.005	0.117	0.083	0.032	0.002
+9/2	0.007	0.001	0.079	0.039	0.023	0.010	0.319	0.008
+11/2	0.015	0.001	0.000	0.009	0.212	0.153	0.323	0.006
+13/2	0.001	0.000	0.491	0.237	0.102	0.001	0.098	0.004
+15/2	0.891	0.074	0.000	0.001	0.004	0.006	0.016	0.000
M	KD 5		KD 6		KD 7		KD 8	
–15/2	0.002	0.001	0.002	0.000	0.000	0.001	0.000	0.000
–13/2	0.034	0.002	0.002	0.012	0.001	0.010	0.001	0.004
–11/2	0.121	0.008	0.018	0.022	0.077	0.002	0.019	0.014

M								
−9/2	0.132	0.011	0.068	0.014	0.123	0.060	0.033	0.074
−7/2	0.266	0.066	0.041	0.046	0.150	0.004	0.098	0.082
−5/2	0.094	0.088	0.258	0.010	0.050	0.071	0.072	0.142
−3/2	0.026	0.031	0.147	0.185	0.005	0.169	0.093	0.135
−1/2	0.084	0.034	0.061	0.115	0.164	0.113	0.137	0.096
+1/2	0.034	0.084	0.115	0.061	0.113	0.164	0.096	0.137
+3/2	0.031	0.026	0.185	0.147	0.169	0.005	0.135	0.093
+5/2	0.088	0.094	0.010	0.258	0.071	0.050	0.142	0.072
+7/2	0.066	0.266	0.046	0.041	0.004	0.150	0.082	0.098
+9/2	0.011	0.132	0.014	0.068	0.060	0.123	0.074	0.033
+11/2	0.008	0.121	0.022	0.018	0.002	0.077	0.014	0.019
+13/2	0.002	0.034	0.012	0.002	0.010	0.001	0.004	0.001
+15/2	0.001	0.002	0.000	0.002	0.001	0.000	0.000	0.000

Dy2

M	KD 1		KD 2		KD 3		KD 4	
−15/2	0.868	0.095	0.001	0.001	0.007	0.000	0.018	0.002
−13/2	0.002	0.000	0.665	0.137	0.000	0.061	0.045	0.022
−11/2	0.015	0.002	0.007	0.011	0.305	0.037	0.319	0.064
−9/2	0.010	0.001	0.067	0.014	0.036	0.022	0.271	0.043
−7/2	0.004	0.000	0.002	0.008	0.143	0.024	0.021	0.002
−5/2	0.002	0.000	0.023	0.001	0.044	0.050	0.049	0.014
−3/2	0.000	0.000	0.038	0.003	0.065	0.023	0.043	0.062
−1/2	0.000	0.000	0.002	0.020	0.120	0.063	0.011	0.015
+1/2	0.000	0.000	0.020	0.002	0.063	0.120	0.015	0.011
+3/2	0.000	0.000	0.003	0.038	0.023	0.065	0.062	0.043
+5/2	0.000	0.002	0.001	0.023	0.050	0.044	0.014	0.049
+7/2	0.000	0.004	0.008	0.002	0.024	0.143	0.002	0.021
+9/2	0.001	0.010	0.014	0.067	0.022	0.036	0.043	0.271
+11/2	0.002	0.015	0.011	0.007	0.037	0.305	0.064	0.319
+13/2	0.000	0.002	0.137	0.665	0.061	0.000	0.022	0.045
+15/2	0.095	0.868	0.001	0.001	0.000	0.007	0.002	0.018

M	KD 5		KD 6		KD 7		KD 8	
−15/2	0.004	0.000	0.004	0.000	0.001	0.000	0.000	0.000
−13/2	0.033	0.002	0.007	0.011	0.007	0.003	0.001	0.004
−11/2	0.080	0.013	0.003	0.064	0.036	0.004	0.007	0.032
−9/2	0.143	0.006	0.087	0.097	0.053	0.026	0.021	0.104
−7/2	0.395	0.019	0.025	0.065	0.074	0.024	0.027	0.166
−5/2	0.188	0.022	0.246	0.097	0.021	0.041	0.034	0.167

M								
−3/2	0.004	0.043	0.105	0.137	0.263	0.003	0.006	0.206
−1/2	0.045	0.003	0.002	0.051	0.280	0.164	0.139	0.084
+1/2	0.003	0.045	0.051	0.002	0.164	0.280	0.084	0.139
+3/2	0.043	0.004	0.137	0.105	0.003	0.263	0.206	0.006
+5/2	0.022	0.188	0.097	0.246	0.041	0.021	0.167	0.034
+7/2	0.019	0.395	0.065	0.025	0.024	0.074	0.166	0.027
+9/2	0.006	0.143	0.097	0.087	0.026	0.053	0.104	0.021
+11/2	0.013	0.080	0.064	0.003	0.004	0.036	0.032	0.007
+13/2	0.002	0.033	0.011	0.007	0.003	0.007	0.004	0.001
+15/2	0.000	0.004	0.000	0.004	0.000	0.001	0.000	0.000

Table S12. Squared projections of the the CF eigenstates corresponding to the eigth lowest Kramers doublets (KDs) of **2** onto angular momentum eigenstates with total angular momentum J = 15/2 and various angular momentum projections M

	Dy1							
M	KD 1		KD 2		KD 3		KD 4	
−15/2	0.924	0.011	0.001	0.002	0.022	0.000	0.018	0.000
−13/2	0.004	0.000	0.175	0.626	0.008	0.048	0.052	0.030
−11/2	0.040	0.000	0.003	0.017	0.326	0.007	0.316	0.032
−9/2	0.014	0.000	0.024	0.066	0.018	0.014	0.220	0.092
−7/2	0.001	0.000	0.004	0.007	0.154	0.000	0.001	0.009
−5/2	0.002	0.000	0.003	0.008	0.034	0.056	0.038	0.063
−3/2	0.001	0.000	0.013	0.025	0.067	0.032	0.019	0.087
−1/2	0.001	0.000	0.014	0.013	0.135	0.078	0.015	0.007
+1/2	0.000	0.001	0.013	0.014	0.078	0.135	0.007	0.015
+3/2	0.000	0.001	0.025	0.013	0.032	0.067	0.087	0.019
+5/2	0.000	0.002	0.008	0.003	0.056	0.034	0.063	0.038
+7/2	0.000	0.001	0.007	0.004	0.000	0.154	0.009	0.001
+9/2	0.000	0.014	0.066	0.024	0.014	0.018	0.092	0.220
+11/2	0.000	0.040	0.017	0.003	0.007	0.326	0.032	0.316
+13/2	0.000	0.004	0.626	0.175	0.048	0.008	0.030	0.052
+15/2	0.011	0.924	0.002	0.001	0.000	0.022	0.000	0.018
M	KD 5		KD 6		KD 7		KD 8	
−15/2	0.001	0.007	0.007	0.001	0.001	0.003	0.001	0.000
−13/2	0.014	0.000	0.011	0.008	0.016	0.004	0.002	0.004
−11/2	0.000	0.120	0.056	0.008	0.019	0.031	0.005	0.020
−9/2	0.089	0.036	0.205	0.011	0.119	0.015	0.019	0.058
−7/2	0.029	0.317	0.093	0.086	0.118	0.048	0.005	0.127
−5/2	0.054	0.114	0.057	0.152	0.081	0.170	0.047	0.121

M								
−3/2	0.051	0.103	0.076	0.009	0.076	0.189	0.025	0.227
−1/2	0.015	0.050	0.096	0.125	0.012	0.098	0.216	0.124
+1/2	0.050	0.015	0.125	0.096	0.098	0.012	0.124	0.216
+3/2	0.103	0.051	0.009	0.076	0.189	0.076	0.227	0.025
+5/2	0.114	0.054	0.152	0.057	0.170	0.081	0.121	0.047
+7/2	0.317	0.029	0.086	0.093	0.048	0.118	0.127	0.005
+9/2	0.036	0.089	0.011	0.205	0.015	0.119	0.058	0.019
+11/2	0.120	0.000	0.008	0.056	0.031	0.019	0.020	0.005
+13/2	0.000	0.014	0.008	0.011	0.004	0.016	0.004	0.002
+15/2	0.007	0.001	0.001	0.007	0.003	0.001	0.000	0.001

Dy2

M	KD 1		KD 2		KD 3		KD 4	
−15/2	0.050	0.590	0.028	0.015	0.157	0.002	0.003	0.086
−13/2	0.005	0.031	0.093	0.272	0.218	0.002	0.022	0.016
−11/2	0.015	0.199	0.008	0.027	0.332	0.004	0.002	0.006
−9/2	0.005	0.019	0.078	0.301	0.165	0.003	0.001	0.029
−7/2	0.005	0.066	0.010	0.012	0.012	0.000	0.025	0.418
−5/2	0.002	0.002	0.026	0.100	0.057	0.005	0.022	0.000
−3/2	0.000	0.011	0.003	0.003	0.017	0.008	0.008	0.224
−1/2	0.001	0.000	0.004	0.020	0.015	0.002	0.136	0.001
+1/2	0.000	0.001	0.020	0.004	0.002	0.015	0.001	0.136
+3/2	0.011	0.000	0.003	0.003	0.008	0.017	0.224	0.008
+5/2	0.002	0.002	0.100	0.026	0.005	0.057	0.000	0.022
+7/2	0.066	0.005	0.012	0.010	0.000	0.012	0.418	0.025
+9/2	0.019	0.005	0.301	0.078	0.003	0.165	0.029	0.001
+11/2	0.199	0.015	0.027	0.008	0.004	0.332	0.006	0.002
+13/2	0.031	0.005	0.272	0.093	0.002	0.218	0.016	0.022
+15/2	0.590	0.050	0.015	0.028	0.002	0.157	0.086	0.003
M	KD 5		KD 6		KD 7		KD 8	
−15/2	0.004	0.031	0.027	0.001	0.005	0.000	0.002	0.000
−13/2	0.075	0.074	0.147	0.015	0.002	0.017	0.000	0.011
−11/2	0.005	0.079	0.267	0.000	0.020	0.006	0.028	0.000
−9/2	0.014	0.066	0.211	0.033	0.013	0.023	0.003	0.036
−7/2	0.093	0.040	0.090	0.050	0.081	0.048	0.039	0.010
−5/2	0.108	0.202	0.009	0.071	0.089	0.197	0.026	0.085
−3/2	0.048	0.020	0.016	0.025	0.254	0.077	0.218	0.067
−1/2	0.089	0.052	0.029	0.009	0.021	0.148	0.084	0.390
+1/2	0.052	0.089	0.009	0.029	0.148	0.021	0.390	0.084

M								
+3/2	0.020	0.048	0.025	0.016	0.077	0.254	0.067	0.218
+5/2	0.202	0.108	0.071	0.009	0.197	0.089	0.085	0.026
+7/2	0.040	0.093	0.050	0.090	0.048	0.081	0.010	0.039
+9/2	0.066	0.014	0.033	0.211	0.023	0.013	0.036	0.003
+11/2	0.079	0.005	0.000	0.267	0.006	0.020	0.000	0.028
+13/2	0.074	0.075	0.015	0.147	0.017	0.002	0.011	0.000
+15/2	0.031	0.004	0.001	0.027	0.000	0.005	0.000	0.002

Table S13. Squared projections of the the CF eigenstates corresponding to the eigth lowest Kramers doublets (KDs) of **3** onto angular momentum eigenstates with total angular momentum J = 15/2 and various angular momentum projections M

	Dy1							
M	KD 1		KD 2		KD 3		KD 4	
−15/2	0.938	0.040	0.000	0.001	0.003	0.008	0.002	0.000
−13/2	0.000	0.000	0.388	0.237	0.106	0.028	0.177	0.034
−11/2	0.008	0.000	0.004	0.008	0.146	0.309	0.101	0.002
−9/2	0.001	0.000	0.091	0.057	0.001	0.001	0.310	0.066
−7/2	0.009	0.000	0.006	0.017	0.064	0.152	0.028	0.008
−5/2	0.002	0.000	0.059	0.048	0.007	0.019	0.125	0.016
−3/2	0.001	0.000	0.028	0.025	0.048	0.039	0.025	0.021
−1/2	0.000	0.000	0.000	0.032	0.005	0.065	0.060	0.023
+1/2	0.000	0.000	0.032	0.000	0.065	0.005	0.023	0.060
+3/2	0.000	0.001	0.025	0.028	0.039	0.048	0.021	0.025
+5/2	0.000	0.002	0.048	0.059	0.019	0.007	0.016	0.125
+7/2	0.000	0.009	0.017	0.006	0.152	0.064	0.008	0.028
+9/2	0.000	0.001	0.057	0.091	0.001	0.001	0.066	0.310
+11/2	0.000	0.008	0.008	0.004	0.309	0.146	0.002	0.101
+13/2	0.000	0.000	0.237	0.388	0.028	0.106	0.034	0.177
+15/2	0.040	0.938	0.001	0.000	0.008	0.003	0.000	0.002
M	KD 5		KD 6		KD 7		KD 8	
−15/2	0.000	0.002	0.003	0.000	0.001	0.000	0.000	0.001
−13/2	0.004	0.003	0.005	0.003	0.002	0.007	0.003	0.003
−11/2	0.011	0.283	0.065	0.010	0.004	0.020	0.002	0.027
−9/2	0.057	0.012	0.039	0.209	0.053	0.020	0.027	0.055
−7/2	0.011	0.189	0.199	0.030	0.119	0.011	0.018	0.138
−5/2	0.023	0.103	0.065	0.103	0.056	0.143	0.066	0.165
−3/2	0.030	0.138	0.084	0.080	0.182	0.046	0.049	0.206
−1/2	0.077	0.057	0.029	0.075	0.025	0.311	0.096	0.144
+1/2	0.057	0.077	0.075	0.029	0.311	0.025	0.144	0.096

M								
+3/2	0.138	0.030	0.080	0.084	0.046	0.182	0.206	0.049
+5/2	0.103	0.023	0.103	0.065	0.143	0.056	0.165	0.066
+7/2	0.189	0.011	0.030	0.199	0.011	0.119	0.138	0.018
+9/2	0.012	0.057	0.209	0.039	0.020	0.053	0.055	0.027
+11/2	0.283	0.011	0.010	0.065	0.020	0.004	0.027	0.002
+13/2	0.003	0.004	0.003	0.005	0.007	0.002	0.003	0.003
+15/2	0.002	0.000	0.000	0.003	0.000	0.001	0.001	0.000

Dy2

M	KD 1		KD 2		KD 3		KD 4	
−15/2	0.078	0.878	0.004	0.000	0.000	0.016	0.010	0.000
−13/2	0.000	0.002	0.706	0.024	0.039	0.011	0.172	0.008
−11/2	0.002	0.021	0.069	0.000	0.013	0.316	0.177	0.012
−9/2	0.001	0.010	0.093	0.005	0.007	0.024	0.317	0.011
−7/2	0.000	0.005	0.024	0.002	0.002	0.157	0.030	0.001
−5/2	0.000	0.001	0.029	0.004	0.035	0.045	0.065	0.042
−3/2	0.000	0.001	0.010	0.010	0.014	0.128	0.058	0.021
−1/2	0.000	0.000	0.015	0.005	0.096	0.096	0.017	0.060
+1/2	0.000	0.000	0.005	0.015	0.096	0.096	0.060	0.017
+3/2	0.001	0.000	0.010	0.010	0.128	0.014	0.021	0.058
+5/2	0.001	0.000	0.004	0.029	0.045	0.035	0.042	0.065
+7/2	0.005	0.000	0.002	0.024	0.157	0.002	0.001	0.030
+9/2	0.010	0.001	0.005	0.093	0.024	0.007	0.011	0.317
+11/2	0.021	0.002	0.000	0.069	0.316	0.013	0.012	0.177
+13/2	0.002	0.000	0.024	0.706	0.011	0.039	0.008	0.172
+15/2	0.878	0.078	0.000	0.004	0.016	0.000	0.000	0.010

M	KD 5		KD 6		KD 7		KD 8	
−15/2	0.002	0.002	0.004	0.000	0.000	0.003	0.001	0.000
−13/2	0.003	0.007	0.000	0.008	0.001	0.011	0.000	0.008
−11/2	0.176	0.036	0.064	0.033	0.019	0.025	0.026	0.010
−9/2	0.018	0.075	0.083	0.014	0.090	0.126	0.043	0.082
−7/2	0.254	0.039	0.040	0.115	0.092	0.060	0.105	0.076
−5/2	0.202	0.019	0.087	0.044	0.067	0.181	0.107	0.072
−3/2	0.100	0.045	0.168	0.070	0.028	0.120	0.044	0.185
−1/2	0.003	0.021	0.167	0.103	0.137	0.041	0.232	0.007
+1/2	0.021	0.003	0.103	0.167	0.041	0.137	0.007	0.232
+3/2	0.045	0.100	0.070	0.168	0.120	0.028	0.185	0.044
+5/2	0.019	0.202	0.044	0.087	0.181	0.067	0.072	0.107
+7/2	0.039	0.254	0.115	0.040	0.060	0.092	0.076	0.105

+9/2	0.075	0.018	0.014	0.083	0.126	0.090	0.082	0.043
+11/2	0.036	0.176	0.033	0.064	0.025	0.019	0.010	0.026
+13/2	0.007	0.003	0.008	0.000	0.011	0.001	0.008	0.000
+15/2	0.002	0.002	0.000	0.004	0.003	0.000	0.000	0.001

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