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Electronic Supporting Information

Theoretical Exploration of Single-Molecule Magnetic and Single-Molecule Toroic Behavior in Peroxide-Bridged Double-triangular $\{M^{II}_3Ln^{III}_3\}$ (M = Ni, Cu and Zn; Ln = Gd, Tb and Dy) Complexes

Amit Gharu[a] and Kuduva R. Vignesh[a,*]

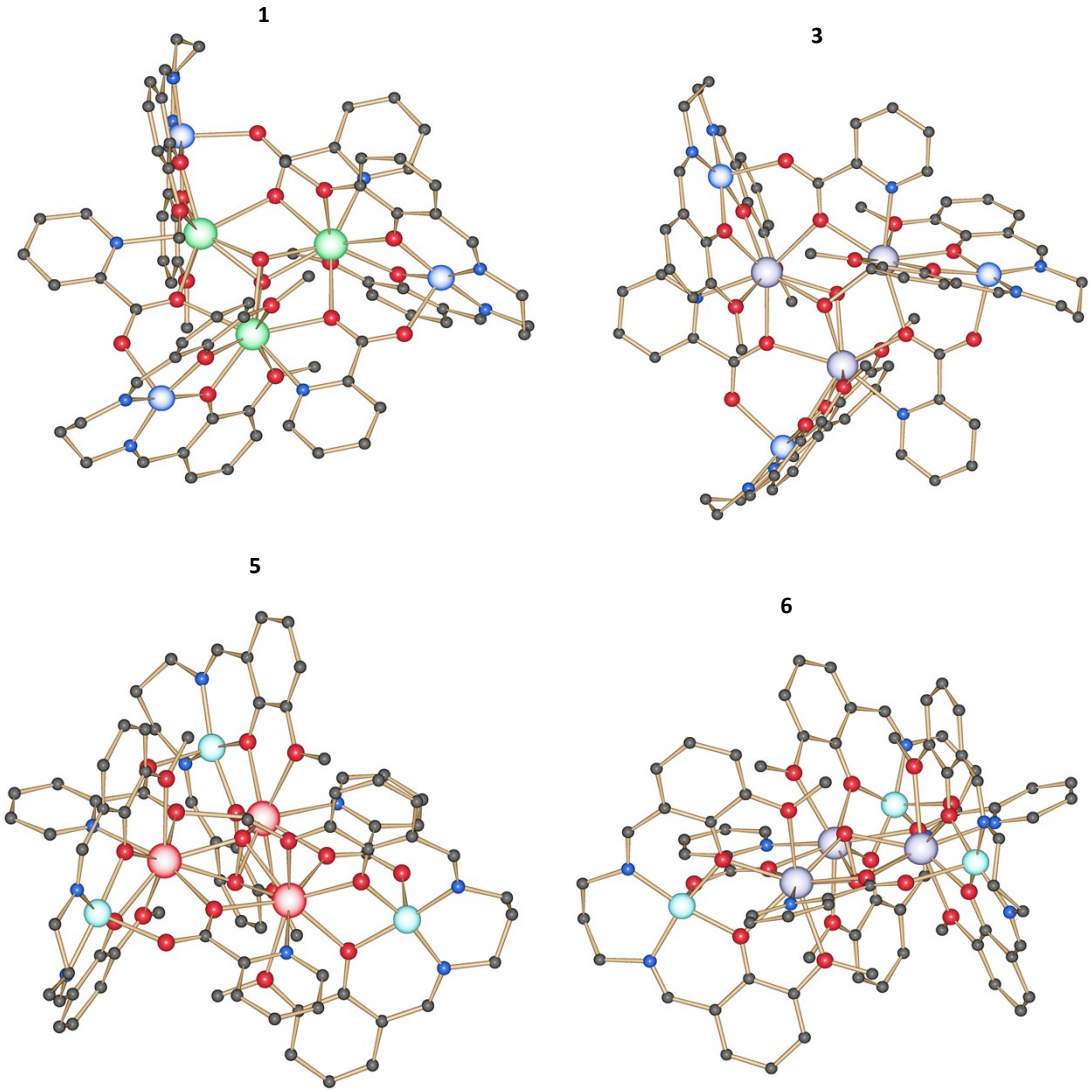

Figure S1: Molecular structure of complexes **1**, **3**, **5**, and **6**.

Table S1: Selected structural parameters of complexes **1-9**.

Structural parameters in relation to the J_1 (Dy^{III}… Dy^{III}) interaction {Zn_3Dy_3} complex					
*Peroxide oxygen atom.					
	Bond Length(Å)		Bond Angle (º)		Dihedral Angle (º)
Zn2-μ2 O3	2.059	Zn2-μ2 O3-Dy1	105.9	Zn2-μ2 O3-Dy1-μ2 O4	17.4
Zn2-μ2 O4	2.061	Zn2-μ2 O4-Dy1	105.7		
Dy1-μ2 O3	2.301				
Dy1-μ2 O4	2.301				
Zn9-μ2 O10	2.059	Zn9-μ2 O10-Dy8	105.8	Zn9-μ2 O10-Dy8-μ2 O11	18.4
Zn 9-μ2 O11	2.061	Zn9-μ2 O11-Dy8	105.7		
Dy8-μ2 O10	2.301				
Dy8-μ2 O11	2.301				
Zn14-μ2 O15	2.059	Zn14-μ2 O15-Dy13	105.8	Zn14-μ2 O15-Dy13-μ2 O16	17.4
Zn14-μ2 O16	2.061	Zn14-μ2 O16-Dy13	105.7		
Dy13-μ2 O15	2.301				
Dy13-μ2 O16	2.301				
Dy1-μ3 O6*	2.358	Dy1-μ3 O6*-Dy8	109.4	Dy1-μ2 O5-Dy13-μ3 O6*	154.5
Dy1-μ3 O7*	2.351	Dy1-μ3 O7*-Dy8	110.1	Dy1-μ2 O5-Dy13-μ3 O7*	20.2
Dy8-μ3 O6*	2.358	Dy1-μ3 O6*-Dy13	109.4	Dy1-μ2 O12-Dy8-μ3 O6*	20.7
Dy8-μ3 O7*	2.351	Dy1-μ3 O7*-Dy13	110.1	Dy1-μ2 O12-Dy8-μ3 O7*	20.6
Dy13-μ3 O6*	2.358	Dy8-μ3 O6*-Dy13	109.4	Dy8-μ2 O17-Dy13-μ3 O6*	22.9
Dy13-μ3 O7*	2.351	Dy8-μ3 O7*-Dy13	110.1	Dy8-μ2 O17-Dy13-μ3 O7*	23.1
Dy1-μ2 O5	2.556	Dy1-μ2 O5-Dy13	102.2		
Dy1-μ2 O12	2.389	Dy1-μ2 O12-Dy8	102.2		
Dy8-μ2 O12	2.556	Dy8-μ2 O17-Dy13	102.2		
Dy8-μ2 O17	2.389				
Dy13-μ2 O5	2.389				
Dy13-μ2 O17	2.556				
Structural parameters in relation to the J_1 (Dy^{III}… Dy^{III}) & J_2 (Cu^{II}…Dy^{III}) interaction {Cu_3Dy_3} complex					
Cu2-μ2 O3	1.973	Cu2-μ2 O3-Dy1	107.1	Cu2-μ2 O3-Dy1-μ2 O4	14.4
Cu2-μ2 O4	1.974	Cu2-μ2 O4-Dy1	106.8		
Dy1-μ2 O3	2.315				
Dy1-μ2 O4	2.321				
Cu9-μ2 O10	1.973	Cu9-μ2 O10-Dy8	107.1	Cu9-μ2 O10-Dy8-μ2 O11	13.2
Cu9-μ2 O11	1.974	Cu9-μ2 O11-Dy8	106.8		
Dy8-μ2 O10	2.315				
Dy8-μ2 O11	2.321				
Cu14-μ2 O15	1.973	Cu14-μ2 O15-Dy13	107.1	Cu14-μ2 O15-Dy13-μ2 O16	13.2
Cu14-μ2 O16	1.974	Cu14-μ2 O16-Dy13	106.8		
Dy13-μ2 O15	2.315				
Dy13-μ2 O16	2.321				
Dy1-μ3 O6*	2.345	Dy1-μ3 O6*-Dy8	109.4	Dy1-μ2 O5-Dy8-μ3O6*	20.3
Dy1-μ3 O7*	2.345	Dy1-μ3 O7*-Dy8	109.4	Dy1-μ2 O5-Dy8-μ3O7*	20.9
Dy8-μ3 O6*	2.346	Dy1-μ3 O6*-Dy13	109.4	Dy1-μ2 O17-Dy13-μ3O6*	20.7
Dy8-μ3 O7*	2.345	Dy1-μ3 O7*-Dy13	109.4	Dy1-μ2 O17-Dy13-μ3O7*	21.4
Dy13-μ3 O6*	2.346	Dy8-μ3 O6*-Dy13	109.4	Dy8-μ2 O12-Dy13-μ3O6*	-20.3
Dy13-μ3 O7*	2.345	Dy8-μ3 O7*-Dy13	109.4	Dy8-μ2 O12-Dy13-μ3O7*	20.9

Dy1-μ2 O5	2.527	Dy1-μ2 O5- Dy 8	102.8		
Dy1-μ2 O17	2.371	Dy1-μ2 O17- Dy 13	102.8		
Dy8-μ2 O5	178.469	Dy8-μ2 O12- Dy13	102.8		
Dy8-μ2 O12	2.527				
Dy13-μ2 O12	2.371				
Dy13-μ2 O17	2.527				
Structural parameters in relation to the J_1 (Dy^{III}… Dy^{III}) & J_2 (Ni^{II}…Dy^{III}) interaction {Ni_3Dy_3} complex					
Ni2-N16	2.007	Ni2-N16-O3	120.4	Ni2-N22-μ2 O5-Dy1	55.2
Ni2-N19	2.011	Ni2-N19-O4	123.6	Ni2-N10-μ2 O4-Dy1	45.9
Ni2-N22	2.017	Ni2-N22-O5	122.9	Ni2-N16-μ2 O3-Dy1	55.8
Dy1-O3	2.411	Dy1-O3-N16	116.1	Ni61-N73-μ2 O62-Dy60	55.8
Dy1-O4	2.366	Dy1-O4-N19	119.4	Ni61-N76-μ2 O63-Dy60	45.9
Dy1-O5	2.384	Dy1-O5-N22	115.6	Ni61-N79-μ2 O64-Dy60	55.2
Ni61-N73	2.007	Ni61-N73-O62	120.4	Ni118-N130-μ2 O119-Dy117	55.8
N61-N76	2.011	N61-N76-O63	123.6	Ni118-N136-μ2 O121-Dy117	55.2
Ni61-N79	2.017	Ni61-N79-O64	122.9	Ni118-N133-μ2 O120-Dy117	45.9
Dy60-O62	2.411	Dy60-O62-N73	122.9		
Dy60-O63	2.366	Dy60-O63-N76	119.4		
Dy60-O64	2.384	Dy60-O64-N79	115.6		
Ni118-N130	2.007	Ni118-N130-O119	120.4		
Ni118-N133	2.011	Ni118-N133-O120	123.6		
Ni118-N136	2.017	Ni118-N136-O121	122.9		
Dy117-O119	2.411	Dy117-O119-N130	116.1		
Dy60-O120	2.366	Dy60-O120-N133	119.5		
Dy60-O121	2.384	Dy60-O121-N136	115.7		
Dy1-μ3 $O9^*$	2.323	Dy1-μ3 $O9^*$-Dy60	110.3	Dy1-μ2 O3-Dy117-μ3 $O9^*$	11.3
Dy1-μ3 $O10^*$	2.328	Dy1-μ3 $O9^*$-Dy117	110.3	Dy1-μ2 O3-Dy117-μ3 $O10^*$	28.6
Dy60-μ3 $O9^*$	2.323	Dy60-μ3 $O9^*$-Dy117	110.3	Dy1-μ2 O62-Dy60-μ3 $O9^*$	11.3
Dy60-μ3 $O10^*$	2.328	Dy1-μ3 $O10^*$-Dy60	109.9	Dy1-μ2 O62-Dy60-μ3 10^*	30.9
Dy117-μ3 $O9^*$	2.323	Dy1-μ3 $O10^*$-Dy117	109.9	Dy60-μ2 O119-Dy117-μ3 $O9^*$	12.2
Dy117-μ3 $O10^*$	2.328	Dy60-μ3 $O10^*$-Dy117	109.9	Dy60-μ2O119-Dy117-μ3$O10^*$	28.6
Dy1-μ2 O3	2.411	Dy1-μ2 O62-Dy60	104.2		
Dy1-μ2 O62	2.422	Dy1-μ2 O3-Dy117	104.2		
Dy60-μ2 O62	2.411	Dy60-μ2 O119-Dy117	104.2		
Dy60-μ2 O119	2.422				
Dy117-μ2 O3	2.422				
Dy117-μ2 O119	2.411				
Structural parameters in relation to the J_1 (Tb^{III}… Tb^{III}) interaction {Zn_3Tb_3} complex					
Zn2-μ2 O3	2.059	Zn2-μ2 O3-Tb1	106.3	Zn2-μ2 O3-Tb1-μ2 O4	18.1
Zn2-μ2 O4	2.056	Zn2-μ2 O4-Tb1	106.1		
Tb1-μ2 O3	2.291				
Tb1-μ2 O4	2.311				
Zn9-μ2 O10	2.059	Zn9-μ2 O10-Tb8	106.3	Zn9-μ2 O10-Tb8-μ2 O11	19.1
Zn 9-μ2 O11	2.056	Zn9-μ2 O11-Tb8	106.1		
Tb8-μ2 O10	2.291				
Tb8-μ2 O11	2.302				
Zn14-μ2 O15	2.059	Zn14-μ2 O15-Tb13	106.3	Zn14-μ2 O15-Tb13-μ2 O16	19.1
Zn14-μ2 O16	2.056	Zn14-μ2 O16-Tb13	106.1		

Tb13-μ2 O15	2.291				
Tb13-μ2 O16	2.302				
Tb1-μ3 O6*	2.374	Tb1-μ3 O6*-Tb8	109.2	Tb1-μ2 O17-Tb13-μ3 O6*	21.1
Tb1-μ3 O7*	2.358	Tb1-μ3 O7*-Tb8	110.3	Tb1-μ2 O17-Tb13-μ3 O7*	21.5
0Tb8-μ3 O6*	2.374	Tb1-μ3 O6*-Tb13	109.2	Tb1-μ2 O5-Tb8-μ3 O6*	20.6
Tb8-μ3 O7*	2.358	Tb1-μ3 O7*-Tb13	110.3	Tb1-μ2 O5-Tb8-μ3 O7*	19.8
Tb13-μ3 O6*	2.374	Tb8-μ3 O6*-Tb13	109.2	Tb8-μ2 O12-Tb13-μ3 O6*	20.6
Tb13-μ3 O7*	2.358	Tb8-μ3 O7*-Tb13	110.3	Tb8-μ2 O12-Tb13-μ3 O7*	19.8
Tb1-μ2 O5	2.584	Tb1-μ2 O17-Tb13	102.1		
Tb1-μ2 O17	2.391	Tb1-μ2 O5-Tb8	102.1		
Tb8-μ2 O12	2.584	Tb8-μ2 O12-Tb13	102.1		
Tb8-μ2 O5	2.391				
Tb13-μ2 O17	2.584				
Tb13-μ2 O12	2.391				
Structural parameters in relation to the J_1 (Tb^{III}…Tb^{III}) & J_2 (Cu^{II}…Tb^{III}) interaction for {Cu_3Tb_3} complex					
Cu9-μ2 O11	1.963	Cu9-μ2 O11-Tb8	107	Cu9-μ2 O11-Tb8-μ2O10	13.6
Cu9-μ2 O10	1.980	Cu9-μ2 O10-Tb8	107.2		
Tb8-μ2 O11	2.331				
Tb8-μ2 O10	2.311				
Cu2-μ2 O3	1.981	Cu2-μ2 O3-Tb1	107.2	Cu2-μ2 O3-Tb1-μ2O4	13.6
Cu2-μ2 O4	1.963	Cu2-μ2 O4-Tb1	107.1		
Tb1-μ2 O3	2.311				
Tb1-μ2 O4	2.331				
Cu14-μ2 O15	1.981	Cu14-μ2 O15-Tb13	107.2	Cu14-μ2 O15-Tb13-μ2O16	14.9
Cu14-μ2 O16	1.963	Cu14-μ2 O16-Tb13	107.1		
Tb13-μ2 O15	2.311				
Tb13-μ2 O16	2.331				
Tb1-μ3 O6*	2.363	Tb1-μ3 O6*-Tb8	109.4	Tb1-μ2 O5-Tb8-μ3O7*	20.8
Tb1-μ3 O7*	2.353	Tb1-μ3 O7*-Tb8	110.1	Tb1-μ2 O5-Tb8-μ3O6*	21.4
Tb8-μ3 O6*	2.363	Tb1-μ3 O6*-Tb13	109.4	Tb1-μ2 O5-Tb13-μ3O6*	22.6
Tb8-μ3 O7*	2.353	Tb1-μ3 O7*-Tb13	110.1	Tb1-μ2 O5-Tb13-μ3O7*	20.4
Tb13-μ3 O6*	2.363	Tb8-μ3 O6*-Tb13	109.4	Tb1-μ2 O17-Tb13-μ3O6*	20.2
Tb13-μ3 O7*	2.353	Tb8-μ3 O7*-Tb13	110.1	Tb8-μ2 O17-Tb13-μ3O7*	20.4
Tb1-μ2 O5	2.551	Tb1-μ2 O12-Tb8	102.9		
Tb1-μ2 O12	2.378	Tb1-μ2 O5-Tb13	102.9		
Tb8-μ2 O17	2.378	Tb8-μ2 O17-Tb13	102.9		
Tb8-μ2 O12	2.551				
Tb13-μ2 O5	2.378				
Tb13-μ2 O17	2.551				
Structural parameters in relation to the J_1 (Gd^{III}…Gd^{III}) interaction for {Zn_3Gd_3} complex					
	Bond Length(Å)		Bond Angle (º)		Dihedral Angle (º)
Zn9-μ2 O11	2.043	Zn9-μ2 O11-Gd8	106.2	Zn9-μ2 O11-Gd8-μ2 O10	17.5
Zn9-μ2 O10	2.054	Zn9-μ2 O10-Gd8	106.5		
Gd8-μ2 O11	2.326				
Gd8-μ2 O10	2.307				
Zn2-μ2 O3	2.054	Zn2-μ2 O3-Gd1	106.5	Zn2-μ2 O3-Gd1-μ2 O4	158.5
Zn2-μ2 O4	2.043	Zn2-μ2 O4-Gd1	106.2		
Gd1-μ2 O3	2.307				

Gd1-μ2 O4	2.326				
Zn14-μ2 O15	2.054	Zn14-μ2 O15-Gd13	106.5	Zn14-μ2 O15-Gd13-μ2 O16	158.5
Zn14-μ2 O16	2.043	Zn14-μ2 O16-Gd13	106.2		
Gd13-μ2 O15	2.307				
Gd13-μ2 O16	2.326				
Gd1-μ3 O6*	2.386	Gd1-μ3 O6*-Gd8	109.5	Gd1-μ2 O5-Gd8-μ3 O6*	20.3
Gd1-μ3 O7*	2.371	Gd1-μ3 O7*-Gd8	110.5	Gd1-μ2 O5-Gd8-μ3 O7*	22.6
Gd8-μ3 O6*	2.386	Gd1-μ3 O6*-Gd13	109.5	Gd1-μ2 O17-Gd13-μ3O6*	20.6
Gd8-μ3 O7*	2.371	Gd1-μ3 O7*-Gd13	110.5	Gd1-μ2 O17-Gd13-μ3O7*	21.5
Gd13-μ3 O6*	2.386	Gd8-μ3 O6*-Gd13	109.5	Gd8-μ2 O12-Gd13-μ2 O6*	20.6
Gd13-μ3 O7*	2.371	Gd8-μ3 O7*-Gd13	110.5	Gd8-μ2 O12-Gd13-μ2 O7*	21.5
Gd1-μ2 O5	2.585	Gd1-μ2 O5-Gd8	102.3		
Gd1-μ2 O17	2.415	Gd1-μ2 O17-Gd13	102.3		
Gd8-μ2 O12	2.585	Gd8-μ2 O12-Gd13	102.3		
Gd8-μ2 O5	2.415				
Gd13-μ2 O12	2.415				
Gd13-μ2 O17	2.585				
Structural parameters in relation to the J_1 (Gd^{III}… Gd^{III}) & J_2 (Ni^{II}…Gd^{III}) interaction {Ni_3Gd_3} complex					
Ni2-N10	2.0353	Ni2-N10-O3	123.7	Ni2-N10-μ2 O3-Gd1	53.1
Ni2-N11	2.014	Ni2-N11-O4	122.8	Ni2-N11-μ2 O4-Gd1	47.1
Ni2-N12	2.014	Ni2-N12-O5	121.5	Ni2-N12-μ2 O5-Dy1	56.8
Gd1-O3	2.406	Gd1-O3-N10	116.3	Ni32-N38-μ2 O33-Gd31	53.1
Gd1-O4	2.379	Gd1-O4-N11	115.4	Ni32-N39-μ2 O34-Gd31	47.1
Gd1-O5	2.441	Gd1-O5-N12	119.9	Ni32-N40-μ2 O35-Gd31	56.8
Ni32-N38	2.035	Ni32-N38-O33	123.7	Ni60-N66-μ2 O61- Gd59	53.1
Ni32-N39	2.014	N32-N39-O34	122.8	Ni60-N67-μ2 O62-Gd59	47.1
Ni32-N40	2.014	Ni32-N40-O34	121.5	Ni60-N68-μ2 O63-Gd59	56.8
Gd31-O33	2.406	Gd31-O33-N38	116.3		
Gd31-O34	2.379	Gd31-O34-N39	119.9		
Gd31-O35	2.441	Gd31-O35-N40	115.4		
Ni60-N66	2.014	Ni60-N66-O61	123.7		
Ni60-N67	2.014	Ni60-N67-O62	122.8		
Ni60-N68	2.035	Ni60-N68-O63	121.5		
Gd59-O61	2.406	Gd59-O61-N66	116.3		
Gd59-O62	2.379	Gd59-O62-N67	119.9		
Gd59-O63	2.441	Gd59-O63-N68	115.4		
Gd1-μ3 O6*	2.358	Gd1-μ3 O6*Gd31	110.1	Gd1-μ2 O5-Gd59-μ3 O6*	11.9
Gd1-μ3 O7*	2.364	Gd1-μ3 O7*Gd31	109.7	Gd1-μ2 O5-Gd59-μ3 O7*	30.6
Gd31-μ3 O6*	2.358	Gd59-μ3 O6*Gd31	110.1	Gd1-μ2 O35-Gd31-μ3 O6*	12.8
Gd31-μ3 O7*	2.364	Gd59-μ3 O7*Gd31	109.7	Gd1-μ2 O35-Gd31-μ3 O7*	30.7
Gd59-μ3 O6*	2.357	Gd59-μ3 O6*Gd1	110.1	Gd31-μ2 O63-Gd59-μ3 O6*	12.8
Gd59-μ3 O7*	2.364	Gd59-μ3 O7*Gd1	109.7	Gd31-μ2 O63-Gd59-μ3 O7*	30.7
Gd1-μ2 O5	2.441				
Gd1-μ2 O35	2.451				
Gd31μ3 O63	2.451				
Gd31-μ2 O35	2.441				
Gd59-μ2 O5	2.451				
Gd59-μ2 O63	2.441				

Table S2: The *g*-tensors for the lowest Kramers doublets of Dy(III) ions in Zn_3Dy_3(**1**):

Kramer Doublets	Dy1	Dy2	Dy3
1	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.003$	$g_y = 0.003$	$g_y = 0.003$
	$g_z = 19.688$	$g_z = 19.688$	$g_z = 19.712$
2	$g_x = 0.503$	$g_x = 0.499$	$g_x = 0.534$
	$g_y = 1.411$	$g_y = 1.429$	$g_y = 1.496$
	$g_z = 16.500$	$g_z = 16.387$	$g_z = 16.367$
3	$g_x = 0.760$	$g_x = 0.679$	$g_x = 0.633$
	$g_y = 1.858$	$g_y = 1.806$	$g_y = 1.868$
	$g_z = 13.156$	$g_z = 13.031$	$g_z = 13.004$
4	$g_x = 0.218$	$g_x = 0.171$	$g_x = 0.123$
	$g_y = 3.134$	$g_y = 3.052$	$g_y = 3.139$
	$g_z = 11.035$	$g_z = 11.052$	$g_z = 11.052$
5	$g_x = 9.076$	$g_x = 8.979$	$g_x = 9.095$
	$g_y = 6.644$	$g_y = 6.645$	$g_y = 6.628$
	$g_z = 2.639$	$g_z = 2.642$	$g_z = 2.641$
6	$g_x = 1.825$	$g_x = 1.825$	$g_x = 1.818$
	$g_y = 2.585$	$g_y = 2.543$	$g_y = 2.632$
	$g_z = 17.116$	$g_z = 17.193$	$g_z = 16.986$
7	$g_x = 0.843$	$g_x = 0.830$	$g_x = 0.779$
	$g_y = 1.413$	$g_y = 1.418$	$g_y = 1.414$
	$g_z = 12.836$	$g_z = 12.756$	$g_z = 12.832$
8	$g_x = 0.332$	$g_x = 0.343$	$g_x = 0.332$
	$g_y = 1.215$	$g_y = 1.260$	$g_y = 1.246$
	$g_z = 16.924$	$g_z = 16.897$	$g_z = 16.891$

Table S3: The *g*-tensors for the lowest Ising doublets of Tb(III) ions in Zn_3Tb_3(**2**):

Kramer Doublets	Tb_1	Tb_2	Tb_3
1	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.003$
	$g_z = 17.908$	$g_z = 17.908$	$g_z = 17.914$
2	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.534$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 1.496$
	$g_z = 14.623$	$g_z = 14.625$	$g_z = 14.622$
3	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.633$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 1.868$
	$g_z = 11.222$	$g_z = 11.223$	$g_z = 11.181$
4	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.123$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 3.139$
	$g_z = 7.419$	$g_z = 7.395$	$g_z = 7.180$
5	$g_x = 0.000$	$g_x = 0.000$	$g_x = 9.095$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 6.628$
	$g_z = 4.282$	$g_z = 4.236$	$g_z = 4.177$
6	$g_x = 0.000$	$g_x = 0.000$	$g_x = 1.818$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 2.632$
	$g_z = 17.195$	$g_z = 17.228$	$g_z = 17.271$

Table S4: The *g*-tensors for the lowest Kramers doublets of Dy(III) ions in Cu_3Dy_3(**3**):

Kramer Doublets	Dy1	Dy2	Dy3
1	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.002$	$g_y = 0.002$	$g_y = 0.002$
	$g_z = 19.619$	$g_z = 19.623$	$g_z = 19.654$
2	$g_x = 0.244$	$g_x = 0.225$	$g_x = 0.227$
	$g_y = 0.570$	$g_y = 0.521$	$g_y = 0.507$
	$g_z = 16.959$	$g_z = 17.016$	$g_z = 17.079$
3	$g_x = 1.502$	$g_x = 1.444$	$g_x = 1.438$
	$g_y = 1.655$	$g_y = 1.622$	$g_y = 1.583$
	$g_z = 13.523$	$g_z = 13.595$	$g_z = 13.617$
4	$g_x = 0.711$	$g_x = 0.645$	$g_x = 0.521$
	$g_y = 2.635$	$g_y = 2.590$	$g_y = 2.646$
	$g_z = 11.022$	$g_z = 11.087$	$g_z = 11.207$
5	$g_x = 1.031$	$g_x = 1.174$	$g_x = 1.407$
	$g_y = 3.512$	$g_y = 3.866$	$g_y = 4.351$
	$g_z = 10.294$	$g_z = 10.001$	$g_z = 9.899$
6	$g_x = 3.122$	$g_x = 3.037$	$g_x = 3.016$
	$g_y = 4.336$	$g_y = 4.163$	$g_y = 3.974$
	$g_z = 14.189$	$g_z = 14.595$	$g_z = 14.958$
7	$g_x = 1.057$	$g_x = 1.077$	$g_x = 0.971$
	$g_y = 1.171$	$g_y = 1.202$	$g_y = 1.108$
	$g_z = 13.954$	$g_z = 13.897$	$g_z = 13.947$
8	$g_x = 0.194$	$g_x = 0.201$	$g_x = 0.198$
	$g_y = 0.574$	$g_y = 0.593$	$g_y = 0.601$
	$g_z = 17.605$	$g_z = 17.58$	$g_z = 17.553$

Table S5: The *g*-tensors for the lowest Ising doublets of Tb(III) ions in Cu_3Tb_3(**4**):

Kramer Doublets	Tb_1	Tb_2	Tb_3
1	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.003$
	$g_z = 17.891$	$g_z = 17.923$	$g_z = 17.927$
2	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 14.615$	$g_z = 14.627$	$g_z = 14.636$
3	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 11.224$	$g_z = 11.170$	$g_z = 11.216$
4	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 7.483$	$g_z = 7.279$	$g_z = 7.418$
5	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 4.534$	$g_z = 4.333$	$g_z = 4.488$
6	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 15.620$	$g_z = 15.953$	$g_z = 15.735$

Table S6: The *g*-tensors for the lowest Kramers doublets of Dy(III) ions in Ni_3Dy_3(**5**):

Kramer Doublets	Dy1	Dy2	Dy3
1	$g_x = 0.005$	$g_x = 0.005$	$g_x = 0.005$
	$g_y = 0.006$	$g_y = 0.006$	$g_y = 0.005$
	$g_z = 19.451$	$g_z = 19.516$	$g_z = 19.478$
2	$g_x = 0.336$	$g_x = 0.378$	$g_x = 0.342$
	$g_y = 0.633$	$g_y = 0.749$	$g_y = 0.681$
	$g_z = 17.428$	$g_z = 17.133$	$g_z = 17.217$
3	$g_x = 2.275$	$g_x = 2.524$	$g_x = 2.183$
	$g_y = 4.593$	$g_y = 5.401$	$g_y = 4.231$
	$g_z = 11.295$	$g_z = 10.588$	$g_z = 11.465$
4	$g_x = 0.552$	$g_x = 1.009$	$g_x = 0.285$
	$g_y = 5.121$	$g_y = 5.393$	$g_y = 5.051$
	$g_z = 10.998$	$g_z = 10.137$	$g_z = 11.067$
5	$g_x = 8.461$	$g_x = 3.102$	$g_x = 8.965$
	$g_y = 6.428$	$g_y = 5.723$	$g_y = 6.011$
	$g_z = 2.171$	$g_z = 9.244$	$g_z = 2.382$
6	$g_x = 1.683$	$g_x = 1.048$	$g_x = 1.191$
	$g_y = 2.388$	$g_y = 1.582$	$g_y = 2.112$
	$g_z = 15.753$	$g_z = 15.842$	$g_z = 15.347$
7	$g_x = 1.102$	$g_x = 1.044$	$g_x = 1.049$
	$g_y = 1.248$	$g_y = 1.229$	$g_y = 1.278$
	$g_z = 16.908$	$g_z = 17.032$	$g_z = 16.981$
8	$g_x = 0.026$	$g_x = 0.026$	$g_x = 0.028$
	$g_y = 0.056$	$g_y = 0.054$	$g_y = 0.059$
	$g_z = 19.631$	$g_z = 19.693$	$g_z = 19.678$

Table S7: The *g*-tensors for the lowest Ising doublets of Tb(III) ions in Ni_3Tb_3(**6**):

Kramer Doublets	Tb1	Tb2	Tb3
1	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 17.434$	$g_z = 17.421$	$g_z = 17.446$
2	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 14.313$	$g_z = 14.227$	$g_z = 14.338$
3	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 10.921$	$g_z = 11.006$	$g_z = 11.021$
4	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 5.431$	$g_z = 5.386$	$g_z = 5.437$
5	$g_x = 0.000$	$g_x = 0.000$	$g_x = 0.000$
	$g_y = 0.000$	$g_y = 0.000$	$g_y = 0.000$
	$g_z = 4.731$	$g_z = 4.333$	$g_z = 4.712$

Table S8: RASSI energies of the lowest spin-orbit states (in cm^{-1}) on each Dy^{III} center.

$\{Zn_3Dy_3\}$(**1**)			$\{Cu_3Dy_3\}$(**3**)			$\{Ni_3Dy_3\}$(**5**)		
Dy1	Dy1	Dy1	Dy1	Dy1	Dy3	Dy1	Dy1	Dy3
0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
128.509	149.195	148.812	148.774	128.450	127.821	141.603	145.025	145.392
169.742	200.109	201.826	203.271	169.652	168.883	185.910	188.064	187.017
235.522	251.860	254.626	257.904	237.262	237.345	224.993	226.093	227.167
351.165	349.755	354.425	356.183	354.812	352.725	295.325	296.231	296.397
408.716	376.045	381.279	386.156	413.354	411.997	325.705	328.843	324.960
485.036	493.507	497.126	501.852	488.001	489.138	374.145	376.904	376.492
579.086	612.036	615.632	617.599	582.197	581.914	530.126	539.580	528.038
3586.161	3599.096	3598.785	3598.768	3585.723	3585.529	3608.219	3609.524	3609.682
3658.744	3667.304	3669.310	3670.097	3659.933	3658.671	3691.295	3691.178	3690.710
3787.460	3787.383	3790.310	3791.964	3789.875	3788.370	3742.226	3744.140	3744.916
3864.857	3865.867	3869.287	3872.362	3867.284	3866.751	3782.482	3784.958	3782.898
3913.218	3938.402	3941.802	3944.053	3915.216	3914.622	3826.137	3829.017	3826.079
3953.415	3971.749	3975.213	3977.925	3956.227	3956.242	3880.971	3886.351	3875.509
3989.207	3989.671	3993.512	6150.351	3993.131	3992.473	3977.265	3983.512	3947.619
6136.698	6149.390	6149.738	6221.057	6136.600	6136.480	6152.830	6155.628	6153.557
6214.008	6217.262	6219.833	6342.834	6216.065	6214.574	6218.475	6218.363	6216.212
6339.054	6337.361	6340.111	6431.673	6341.070	6340.856	6269.227	6271.429	6268.993
6409.160	6425.308	6428.690	6462.770	6410.907	6410.812	6334.386	6336.734	6331.223
6430.808	6458.351	6460.765	6494.493	6433.063	6432.429	6384.139	6388.603	6373.641
6489.253	6487.427	6491.882	8134.152	6493.393	6492.012	6455.692	6461.624	6426.398
8119.746	8132.311	8133.205	8194.394	8120.098	8119.826	8123.758	8126.962	8122.459
8189.680	8189.263	8192.129	8325.522	8191.948	8191.137	8173.084	8173.749	8169.573
8317.725	8318.752	8321.654	8395.853	8319.417	8320.178	8246.182	8248.526	8242.650
8360.753	8392.083	8394.280	8445.558	8362.491	8361.757	8321.290	8325.194	8314.337
8437.614	8439.502	8443.827	9665.641	8441.510	8439.517	8394.722	8399.645	8376.908
9652.595	9663.413	9664.905	9749.438	9653.562	9652.646	9660.589	9663.848	9659.158
9748.753	9742.280	9744.918	9895.167	9750.438	9751.858	9707.019	9707.874	9703.212
9860.703	9890.682	9892.644	9980.727	9861.803	9861.898	9806.863	9811.081	9803.611
9973.881	9974.711	9978.932	10135.773	9977.698	9975.891	9933.586	9938.290	9924.350
10111.711	10132.763	10134.890	10143.100	10113.067	10111.730	10086.745	10087.149	10086.907
10139.955	10138.921	10141.753	10186.538	10142.522	10141.347	10109.395	10112.403	10109.919
10172.656	10182.215	10185.025	10200.220	10174.670	10173.668	10131.675	10134.124	10132.240
10185.899	10196.067	10198.625	10239.970	10188.032	10186.701	10156.798	10159.363	10157.502
10222.797	10234.926	10238.346	10318.774	10225.753	10224.093	10194.196	10198.312	10194.549
10301.809	10312.613	10316.491	10814.458	10305.131	10304.245	10222.733	10226.846	10219.706
10803.584	10812.714	10813.993	11013.993	10804.329	10803.349	10815.672	10816.504	10812.215
11003.832	11006.917	11010.429	11169.651	11006.376	11006.007	10935.738	10938.573	10936.641
11147.606	11162.795	11166.261	11557.754	11150.525	11151.064	11091.507	11098.374	11091.123
11546.229	11552.990	11555.675	11621.684	11548.249	11547.548	11521.849	11523.888	11522.063
11602.107	11617.883	11620.499	11628.682	11604.207	11602.768	11554.498	11557.452	11555.319
11616.313	11625.168	11628.316	11654.381	11619.064	11616.909	11582.061	11585.245	11582.429
11641.124	11649.251	11652.420	11681.158	11643.764	11642.731	11590.107	11593.481	11590.197
11664.025	11676.070	11679.525	13512.413	11666.959	11665.494	11617.733	11620.328	11617.338
13497.965	13507.554	13510.319	13538.658	13500.028	13499.409	13461.513	13464.364	13462.027
13524.672	13533.866	13536.701	13577.954	13526.853	13525.920	13501.557	13504.637	13502.631
13561.445	13573.051	13576.091	13608.887	13564.068	13562.930	13519.000	13522.319	13520.008
13595.235	13604.764	13607.818	14981.779	13597.783	13596.336	13541.426	13544.366	13541.507
14967.045	14977.049	14979.906	15007.996	14969.231	14968.252	14935.145	14937.907	14936.370
14992.219	15003.272	15006.233	15030.517	14994.674	14993.500	14953.035	14955.977	14954.049
15017.203	15025.885	15028.799	15867.044	15019.554	15018.720	14973.124	14976.715	14973.642
15852.348	15862.324	15865.225	15874.882	15854.616	15853.670	15810.901	15813.823	15811.852
15860.460	15870.170	15873.085	16404.937	15862.856	15861.887	15823.563	15826.996	15824.424
16390.166	16400.029	16403.034	24650.882	16392.586	16391.633	16349.666	16352.954	16350.570

24634.365	24646.350	24648.614	24716.010	24635.841	24635.148	24624.518	24627.496	24627.354
24698.964	24710.886	24712.775	24768.495	24699.889	24700.482	24655.937	24658.760	24656.370
24763.270	24762.451	24764.773	24812.237	24764.663	24765.477	24742.634	24744.168	24744.468
24799.240	24809.481	24812.219	24913.305	24801.436	24798.927	24797.180	24799.605	24797.917
24895.505	24910.156	24912.446	24913.305	24897.232	24895.594	24851.589	24799.605	24851.983
25152.778	25163.589	25163.071	25166.223	25151.591	25153.443	25171.456	24854.669	25173.023
25208.755	25222.492	25224.331	25166.223	25209.537	25209.415	25242.302	25172.982	25242.915
25364.085	25357.998	25361.353	25226.268	25366.797	25365.259	25275.557	25243.921	25280.042
25375.806	25374.518	25377.291	25226.268	25378.158	25377.091	25340.575	25279.729	25339.939
25405.562	25429.188	25432.038	25362.330	25407.612	25408.675	25364.787	25343.158	25365.867
25436.880	25447.080	25449.901	25362.330	25438.805	25437.277	25395.140	25368.322	25397.025
25496.857	25511.204	25514.367	25379.516	25524.763	25498.459	25439.076	25397.364	25440.528
25521.406	25526.562	25530.167	25379.516	27268.134	25522.977	25501.836	25444.269	25500.993
27268.324	27275.518	27276.063	25435.766	27379.267	27268.331	27300.325	25505.336	27299.792
27378.233	27387.914	27389.595	25435.766	27422.780	27379.396	27354.957	27302.017	27356.409
27421.433	27431.620	27434.076	25451.302	27474.681	27423.194	27401.835	27358.175	27401.513
27472.755	27483.177	27486.071	25451.302	27527.902	27475.010	27428.401	27403.727	27429.519
27525.710	27538.665	27542.382	25515.735	27560.329	27527.522	27471.270	27432.068	27472.426
27557.221	27559.168	27562.353	25515.735	27647.748	27560.744	27515.867	27475.821	27515.661
27646.408	27649.983	27652.540	25532.251	27670.111	27644.110	27601.806	27520.035	27602.664
27667.818	27676.881	27678.740	25532.251	27702.005	27702.094	27633.735	27606.013	27637.554
27701.466	27715.329	27716.675	27277.720	27720.398	27719.810	27680.523	27633.768	27686.910
27719.386	27727.117	27729.846	27277.720	27743.268	27742.736	27710.085	27682.313	27713.829
27742.335	27756.437	27757.971	27391.572	27787.128	27785.007	27738.179	27709.749	27739.597
27784.784	27791.011	27793.816	27391.572	27799.948	27799.201	27764.670	27737.076	27771.896
27798.675	27818.204	27819.962	27437.242	27846.545	27844.905	27785.518	27767.589	27795.038
27843.735	27849.173	27851.082	27437.242	27863.062	27860.242	27827.321	27791.524	27833.095
27861.283	27865.304	27868.579	27489.580	27922.138	27917.940	27839.698	27829.315	27843.416
27917.415	27933.685	27938.403	27489.580	28106.273	28105.357	27921.632	27841.908	27919.811
28102.421	28114.125	28118.615	27545.867	28856.102	28856.149	27947.247	27926.015	27957.815
28856.584	28870.392	28870.156	27545.867	28909.425	28908.534	28877.205	27952.905	28879.521
28908.979	28919.743	28921.405	27566.000	28998.380	28998.619	28943.315	28878.997	29068.769
……	……	……	……	……	……	……	……	……

Table S9: RASSI energies of the lowest spin-orbit states (in cm^{-1}) on each Tb^{III} center.

{Zn_3Tb_3}(**2**)			{Cu_3Tb_3}(**4**)			{Ni_3Tb_3}(**6**)		
Tb1	Tb2	Tb3	Tb3	Tb2	Tb3	Tb1	Tb2	Tb3
0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.010	0.026	0.031	0.026	0.009	0.016	0.680	0.661	0.632
178.483	173.455	171.848	172.334	177.669	177.128	84.266	82.894	83.080
179.330	173.743	172.105	172.585	178.470	177.928	90.590	88.858	88.825
324.282	319.761	317.439	318.306	323.080	322.117	158.459	159.017	158.678
337.008	331.952	330.411	330.489	335.761	336.000	180.338	180.303	179.224
427.279	425.457	422.269	424.870	427.271	425.144	239.880	240.246	239.069
432.183	439.653	440.017	438.489	430.741	431.605	289.226	289.097	286.582
472.661	478.055	475.535	477.263	472.103	469.334	315.705	316.089	313.678
498.794	538.330	540.273	539.682	498.779	498.988	366.838	364.849	363.639
509.521	548.862	548.610	549.530	509.089	507.735	372.988	371.037	369.946
601.273	576.701	579.774	578.333	603.536	604.500	456.187	456.917	450.245
603.780	582.047	584.213	583.389	605.898	606.932	456.913	457.812	450.994
2287.916	2276.573	2252.543	2253.116	2286.827	2264.463	2134.672	2134.409	2132.533
2293.581	2282.589	2258.272	2259.022	2292.465	2269.760	2135.839	2136.950	2134.336
2324.208	2337.087	2313.792	2315.503	2324.542	2300.772	2224.001	2223.597	2223.088
2358.250	2372.231	2350.220	2350.351	2358.950	2336.089	2242.987	2243.876	2242.598
2366.216	2377.859	2356.343	2355.196	2364.411	2343.511	2259.572	2260.127	2258.178
2422.688	2432.406	2408.258	2410.360	2421.858	2398.693	2281.308	2280.862	2280.081
2467.810	2464.007	2443.343	2443.595	2469.972	2447.391	2322.811	2320.561	2320.243
2486.064	2471.280	2448.626	2450.022	2486.331	2463.360	2339.328	2339.720	2336.982
2525.263	2506.705	2482.499	2484.446	2525.042	2500.825	2369.166	2367.825	2366.931
2609.385	2621.521	2600.454	2598.588	2608.823	2588.036	2445.164	2445.910	2440.725
2645.800	2657.286	2637.619	2634.412	2645.603	2625.417	2460.868	2462.517	2456.911
3640.164	3636.464	3628.126	3630.471	3641.969	3632.639	3535.758	3535.722	3535.040
3686.694	3691.050	3683.498	3683.119	3685.033	3678.878	3573.774	3572.817	3571.612
3735.742	3729.099	3717.274	3722.546	3736.344	3724.406	3591.633	3589.161	3588.339
3749.722	3753.239	3742.986	3745.172	3748.427	3739.279	3620.208	3619.555	3618.079
3790.418	3799.108	3790.323	3788.974	3788.350	3780.930	3669.095	3669.531	3668.369
3813.484	3805.653	3792.991	3797.580	3811.741	3801.159	3690.774	3694.518	3689.589
3894.920	3906.548	3904.830	3900.139	3896.974	3892.141	3718.992	3719.574	3716.277
3902.771	3911.714	3908.423	3904.351	3904.631	3899.062	3795.792	3793.309	3792.144
3970.758	3976.768	3973.914	3968.606	3971.008	3966.707	3817.847	3816.681	3814.218
4717.229	4716.822	4700.225	4701.673	4716.723	4700.403	4612.682	4613.041	4610.745
4754.859	4755.845	4740.468	4740.514	4754.053	4739.351	4650.351	4648.574	4648.430
4802.206	4809.164	4795.549	4794.238	4802.469	4788.107	4672.115	4669.508	4669.606
4859.593	4857.493	4843.964	4842.336	4859.647	4845.263	4706.208	4706.738	4704.703
4901.602	4905.385	4893.692	4890.921	4902.439	4889.565	4729.884	4729.858	4727.120
4930.317	4936.857	4923.038	4921.614	4931.177	4915.980	4758.309	4758.198	4755.776
4934.662	4942.909	4928.436	4927.711	4935.259	4919.733	4764.489	4763.142	4760.909
5380.837	5385.312	5385.542	5385.612	5381.325	5380.523	5348.982	5348.470	5348.063
5553.628	5547.386	5544.299	5544.203	5552.875	5550.353	5384.311	5381.387	5380.689
5590.549	5596.976	5597.446	5593.725	5590.256	5590.848	5466.105	5469.063	5463.354
5685.680	5689.678	5686.146	5686.655	5685.752	5681.552	5510.445	5509.771	5508.366
5741.209	5751.193	5751.790	5750.352	5742.854	5741.635	5565.359	5567.160	5562.699
5896.730	5893.489	5908.633	5907.495	5896.138	5911.446	5833.299	5833.285	5830.764
6160.261	6184.186	6199.769	6201.214	6160.705	6176.059	5976.804	5977.342	5975.256
6213.207	6198.405	6217.171	6214.057	6214.360	6230.864	6095.354	6094.391	6090.517
6354.012	6356.343	6384.574	6383.727	6354.663	6381.840	6234.342	6233.970	6231.018
23662.565	23663.343	24434.671	24435.289	23662.425	24434.308	24340.817	24339.977	24338.560
23662.952	23663.630	24434.918	24435.509	23662.761	24434.774	24340.970	24340.147	24338.719
23720.947	23721.477	24500.962	24502.005	23721.281	24497.320	24391.233	24390.532	24389.080
23724.279	23726.360	24504.636	24505.694	23724.480	24502.517	24394.830	24394.371	24392.542
23779.104	23777.781	24546.363	24548.062	23778.876	24546.973	24410.974	24409.785	24408.703

23791.754	23794.054	24561.283	24562.524	23792.088	24558.817	24428.607	24426.920	24426.294
23819.056	23816.035	24581.388	24583.413	23819.410	24586.960	24432.597	24431.069	24430.161
23821.668	23830.879	24613.757	24614.234	23820.717	24604.316	24486.354	24485.413	24483.332
23831.048	23836.546	24618.114	24618.873	23830.600	24612.426	24487.754	24486.852	24484.764
29703.874	29703.729	29990.698	29992.186	29703.966	29990.457	29878.296	29877.570	29876.026
29707.187	29706.211	29994.264	29995.829	29707.155	29993.896	29879.964	29879.268	29877.700
29720.445	29723.508	30002.196	30003.904	29720.378	30002.969	29884.449	29883.261	29882.110
29723.623	29724.597	30011.716	30013.358	29723.162	30010.001	29891.384	29890.636	29889.038
29730.199	29732.082	30014.888	30016.438	29730.385	30011.565	29893.110	29892.728	29890.822
29731.356	29733.876	30021.385	30022.849	29731.271	30018.449	29901.641	29900.799	29899.376
29737.901	29737.084	30026.807	30028.007	29737.817	30026.363	29909.489	29908.515	29907.118
30365.482	30373.760	31330.256	31335.187	30366.561	31322.232	31226.367	31224.605	31224.129
30366.615	30375.045	31331.366	31336.203	30367.814	31323.254	31226.628	31224.951	31224.381
30376.646	30385.246	31342.097	31346.387	30376.788	31332.482	31262.447	31256.944	31261.091
30377.936	30386.497	31343.381	31347.641	30378.092	31333.718	31264.819	31259.907	31263.403
30435.560	30429.752	31387.994	31393.865	30431.279	31394.438	31274.618	31269.455	31273.722
30437.424	30431.760	31388.368	31394.709	30433.419	31395.001	31276.846	31274.956	31276.181
30457.950	30461.478	31426.849	31425.027	30459.518	31425.439	31293.793	31287.443	31291.988
30461.642	30464.075	31429.694	31428.261	30463.506	31429.482	31297.993	31295.552	31296.416
30504.053	30506.769	31470.492	31469.261	30504.236	31466.997	31322.405	31322.006	31319.619
30518.744	30518.329	31477.523	31477.927	30520.425	31481.644	31324.739	31324.833	31322.158
30528.560	30527.010	31487.779	31487.887	30528.204	31483.608	31348.212	31344.316	31345.389
30535.392	30535.635	31491.580	31491.568	30536.608	31494.714	31359.652	31356.850	31356.446
30536.864	30543.284	31503.439	31502.943	30538.430	31497.996	31379.213	31375.228	31375.511
30546.193	30546.589	31506.200	31507.182	30543.790	31500.522	31415.133	31414.906	31412.790
30551.586	30550.699	31509.600	31508.604	30550.296	31506.161	31420.685	31419.911	31418.170
30568.058	30568.724	31535.013	31535.285	30568.356	31533.116	31456.356	31456.701	31452.681
30569.969	30569.945	31537.657	31537.414	30570.436	31536.938	31457.436	31457.515	31453.780
30646.651	30647.061	31611.398	31612.039	30646.921	31610.157	31488.936	31490.039	31486.668
30647.389	30647.677	31613.329	31614.013	30647.622	31612.057	31489.066	31490.155	31486.806
30693.483	30692.142	31668.176	31665.844	30692.869	31667.834	31499.510	31508.726	31496.722
30693.648	30692.295	31668.784	31666.493	30693.027	31668.424	31499.807	31508.941	32110.850
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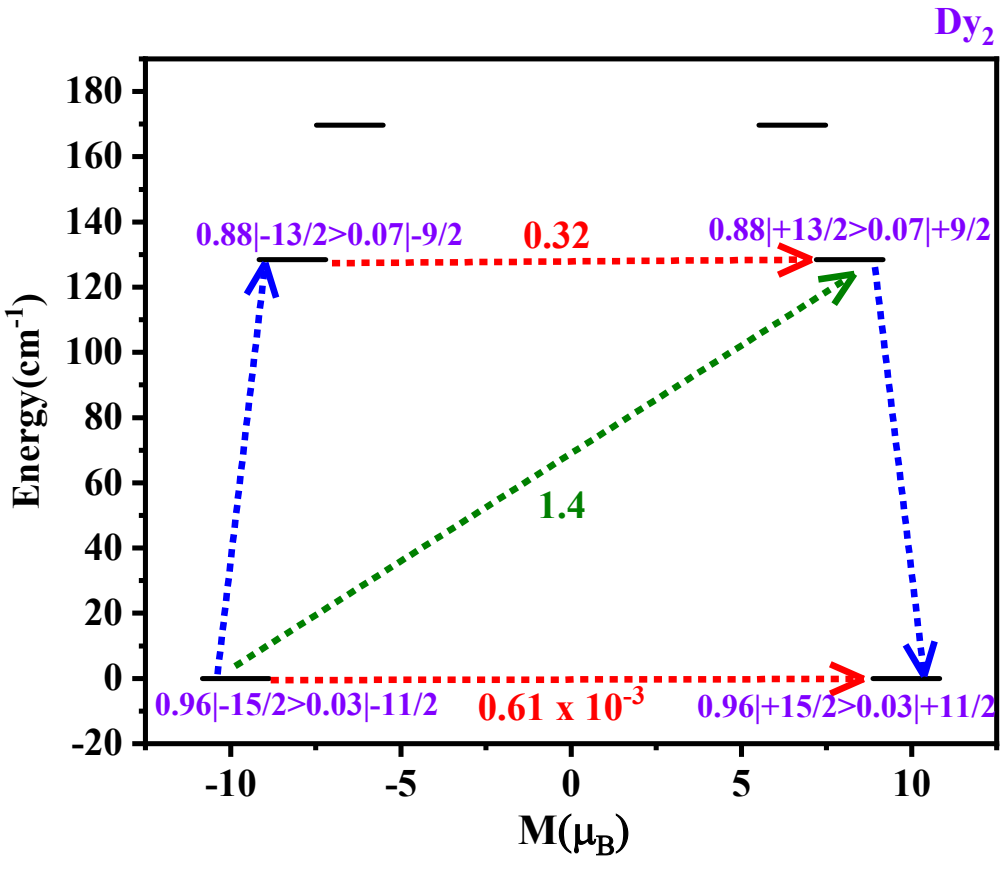

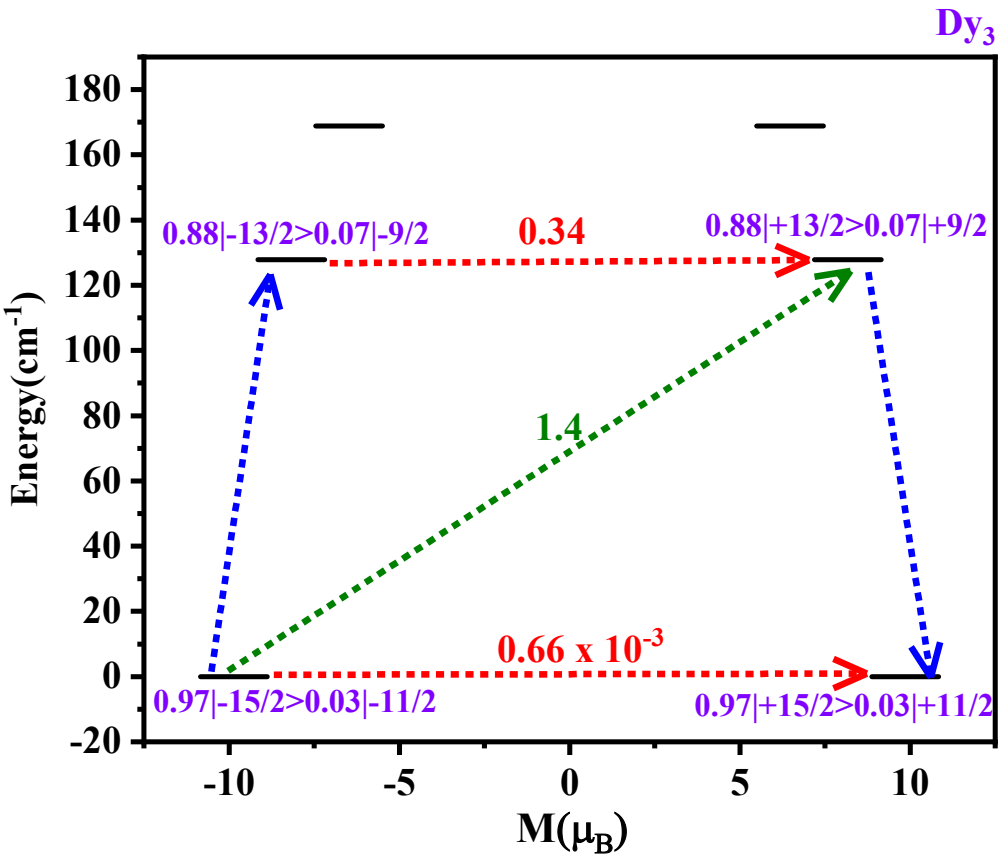

Figure S2: Magnetic relaxation mechanism of Dy2 and Dy3 centers in **1.**

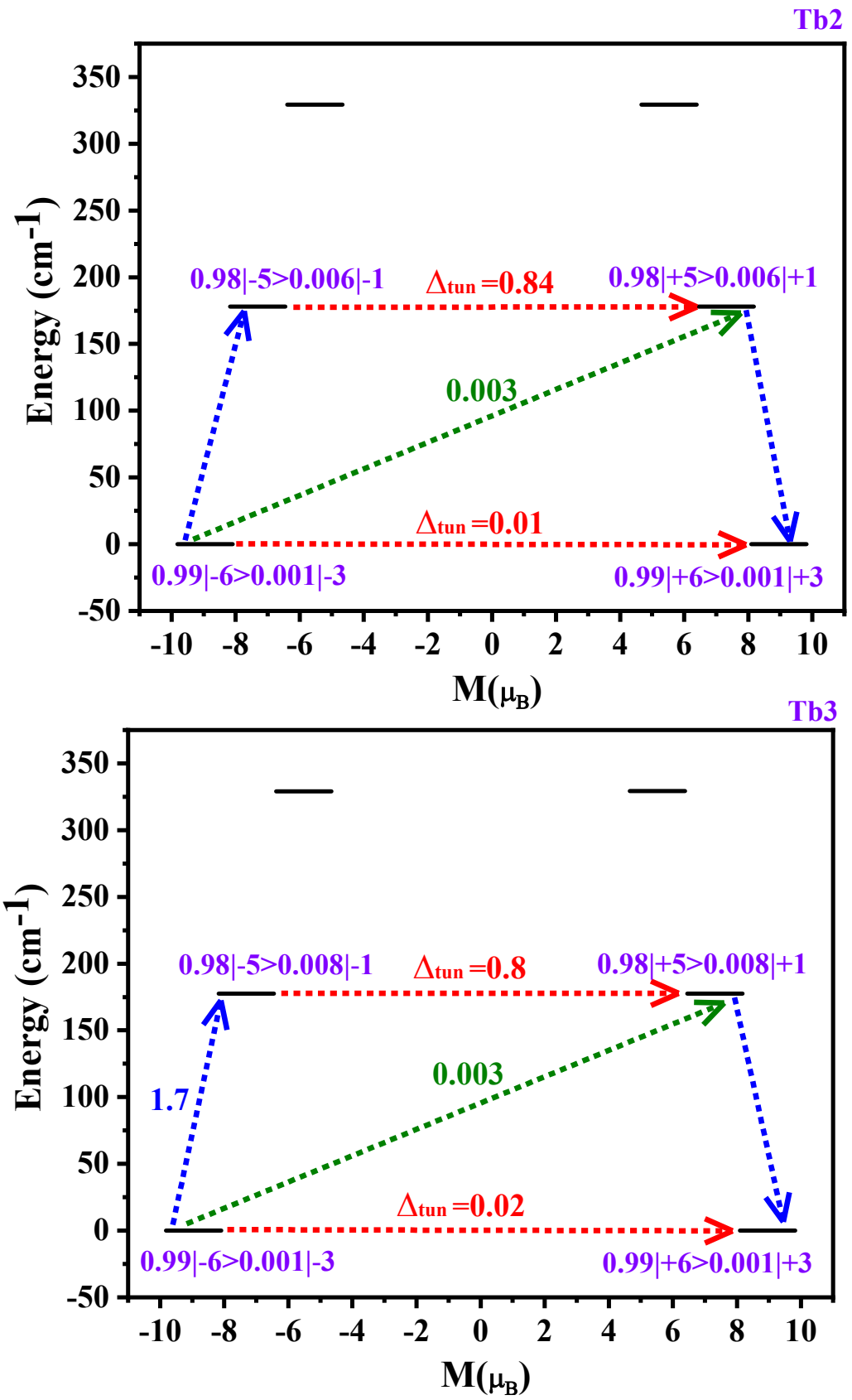

Figure S3: Magnetic relaxation mechanism of Tb2 and Tb3 centers in **2.**

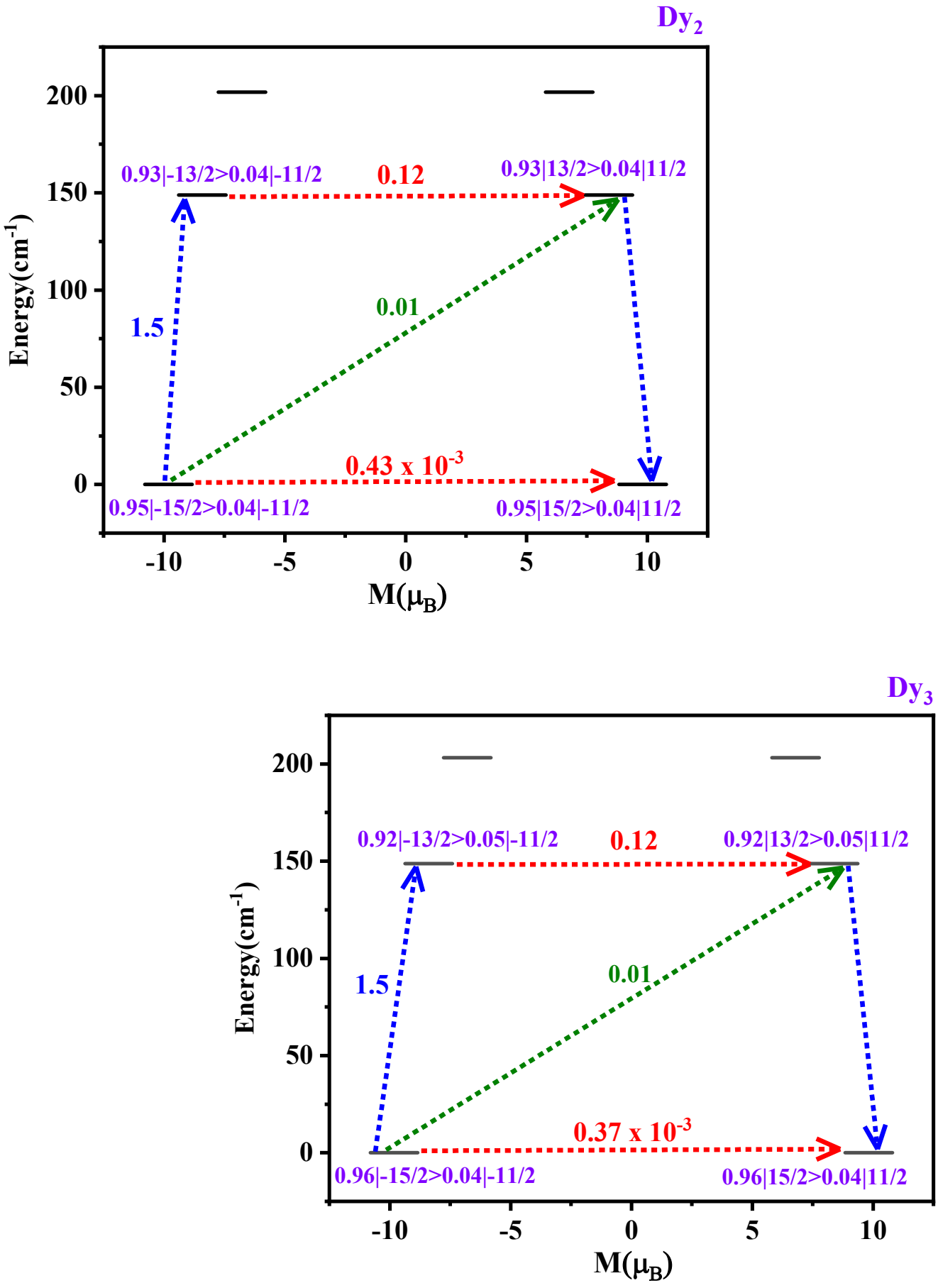

Figure S4: Magnetic relaxation mechanism of Dy2 and Dy3 centers in **4.**

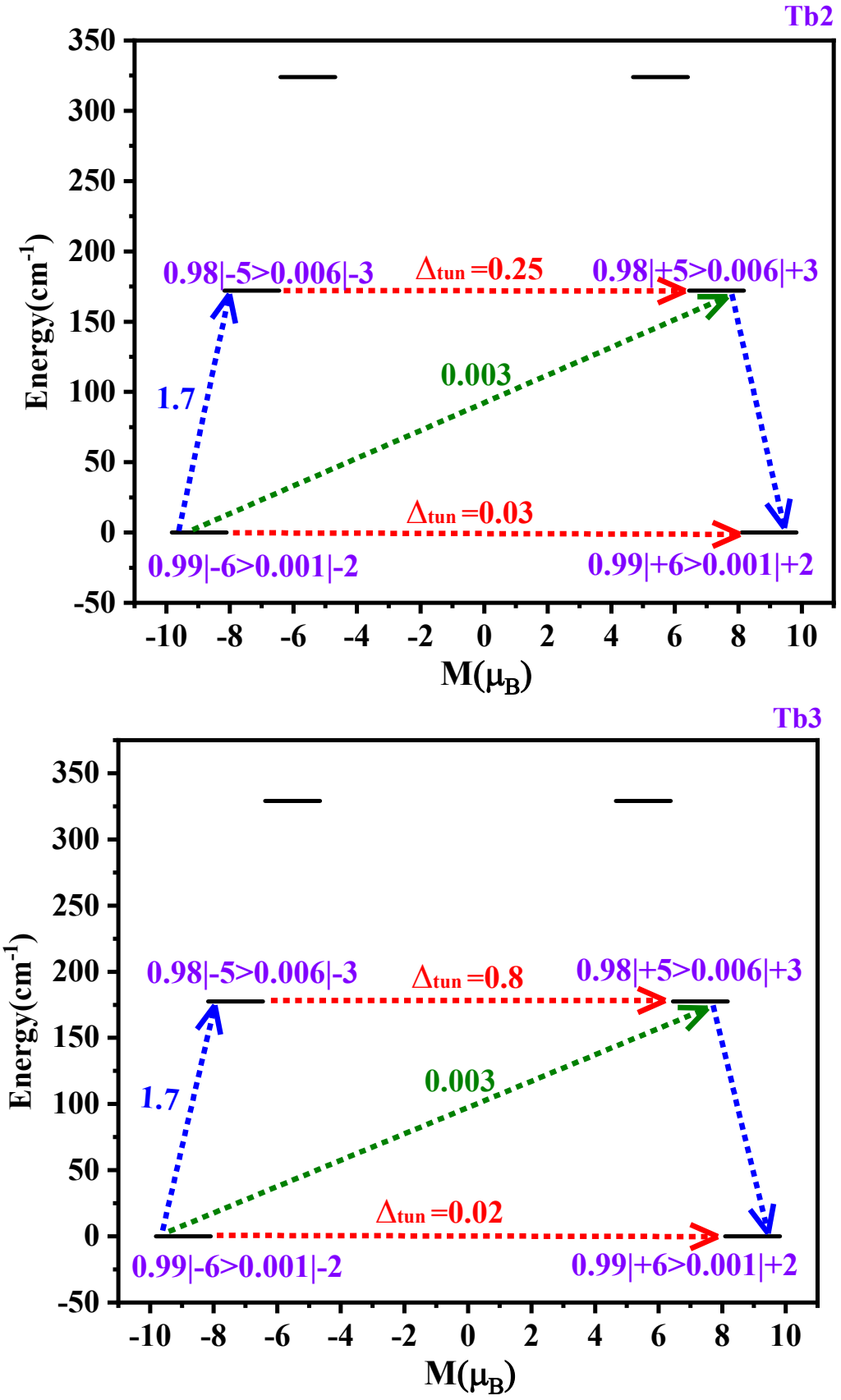

Figure S5: Magnetic relaxation mechanism of Tb2 and Tb3 centers in **5.**

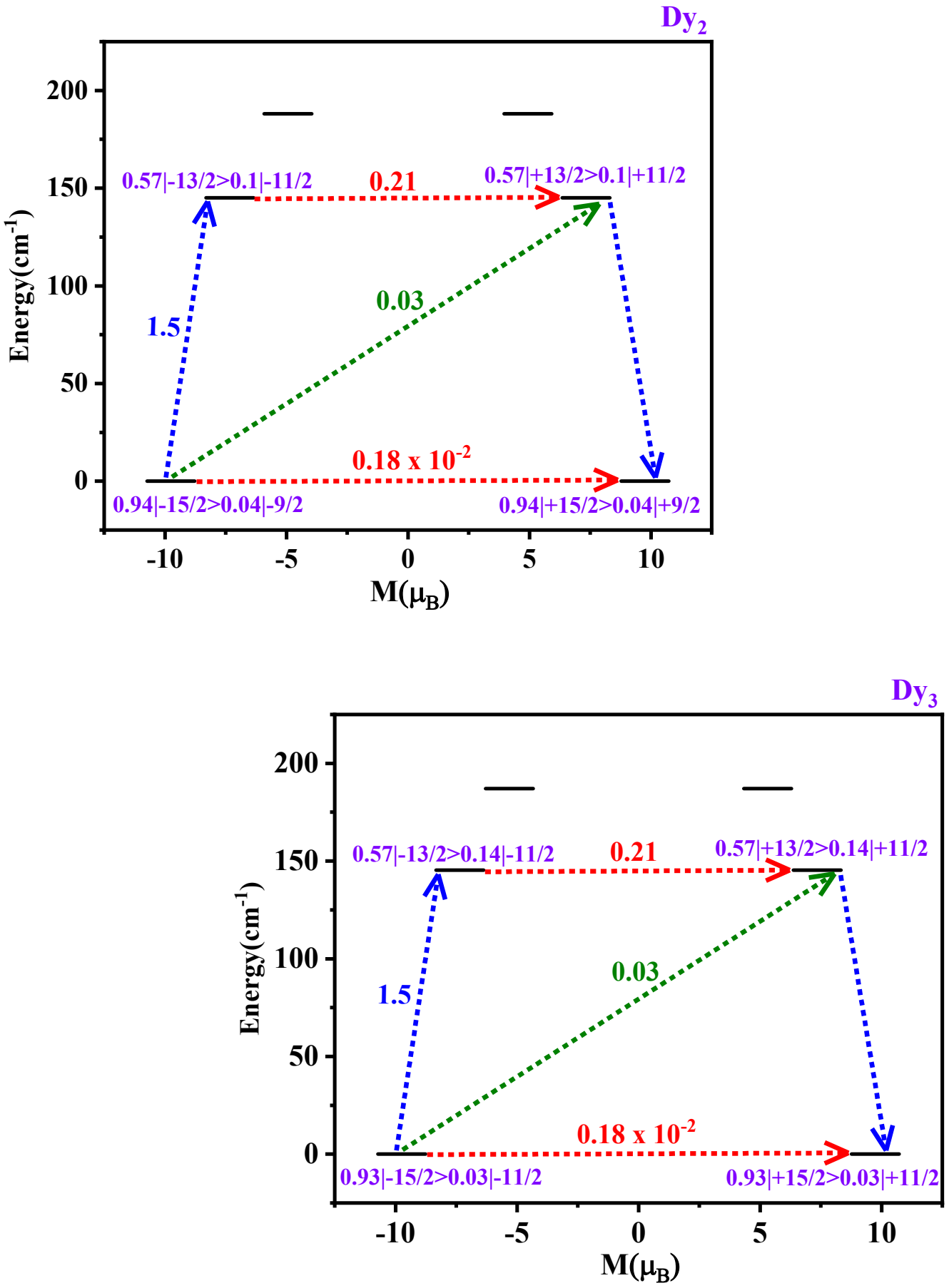

Figure S6: Magnetic relaxation mechanism of Dy2 and Dy3 centers in **7.**

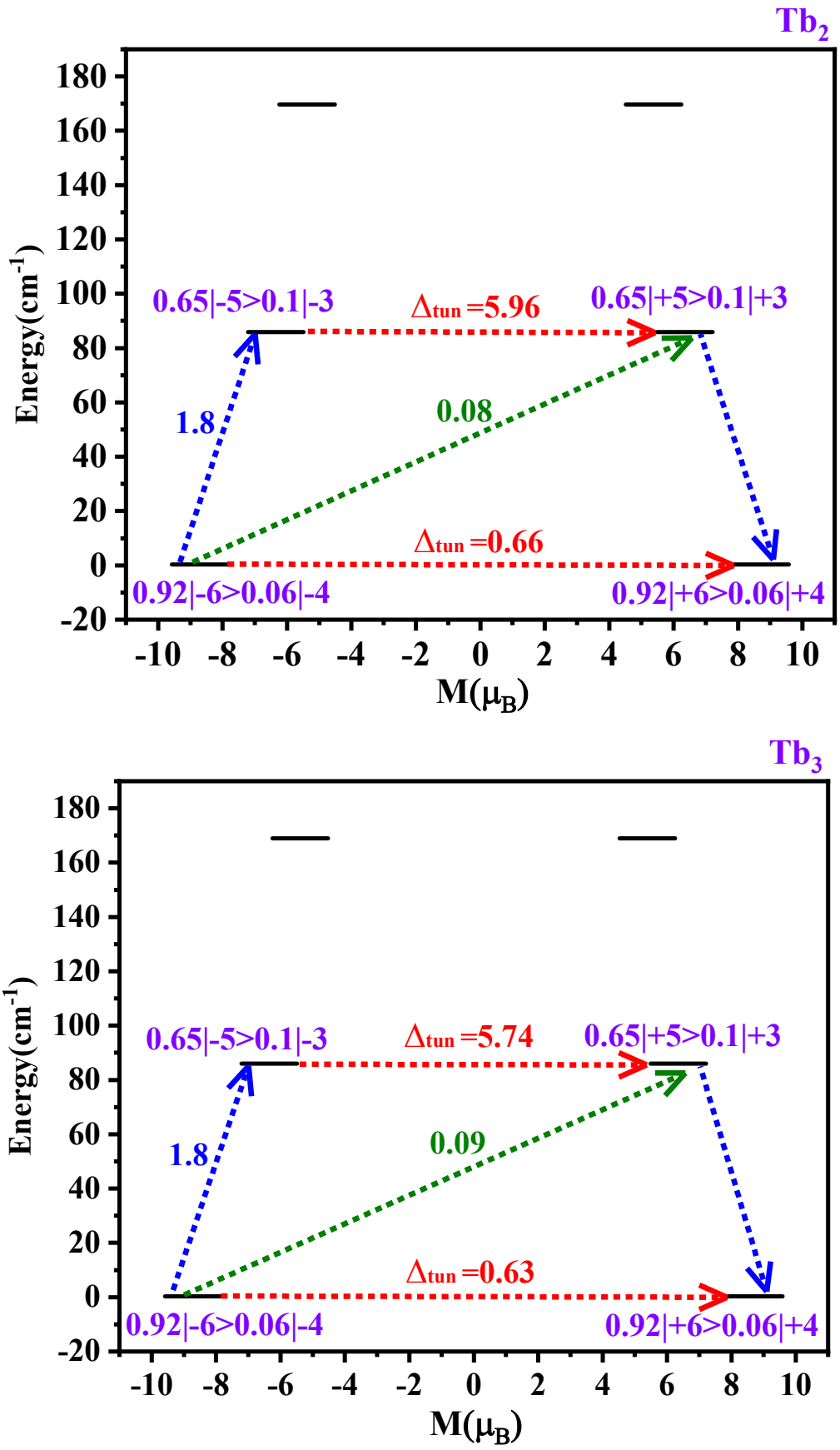

Figure S7: Magnetic relaxation mechanism of Tb2 and Tb3 centers in **8.**

Table S10: SINGLE_ANISO computed crystal field parameters for each Dy(III) ion in complex $\{Zn_3Dy_3\}$(**1**). The major components in the Table are in bold. B_k^q is the crystal field parameter and Q_k^q is the extended Stevens operator. The quantization axis is chosen to be the main magnetic axis of the ground pseudo-doublet.

k	q	B_k^q	B_k^q	B_k^q
		Dy1	Dy2	Dy3
2	-2	0.005	0.207	0.093
	-1	-0.367	0.337	-0.338
	0	-2.866	-2.88	-2.893
	1	0.078	0.095	-0.062
	2	-0.448	-0.432	-0.478
4	-4	-0.001	0.012	0.011
	-3	-0.039	0.034	-0.034
	-2	-0.727×10^{-3}	0.004	0.004
	-1	0.008	-0.008	0.008
	0	0.002	0.002	0.002
	1	0.939×10^{-3}	-0.001	0.976×10^{-3}
	2	-0.011	-0.01	-0.011
	3	0.002	0.021	-0.018
	4	-0.018	-0.013	-0.014
6	-6	-0.273×10^{-4}	0.173×10^{-3}	0.167×10^{-3}
	-5	-0.154×10^{-4}	0.311×10^{-4}	-0.288×10^{-4}
	-4	-0.111×10^{-4}	0.296×10^{-4}	0.231×10^{-4}
	-3	0.965×10^{-4}	-0.863×10^{-4}	0.785×10^{-4}
	-2	0.168×10^{-4}	-0.152×10^{-3}	-0.124×10^{-3}
	-1	-0.135×10^{-3}	0.134×10^{-3}	-0.14×10^{-3}
	0	-0.244×10^{-4}	-0.239×10^{-4}	-0.244×10^{-4}
	1	-0.234×10^{-4}	0.167×10^{-4}	-0.109×10^{-4}
	2	0.389×10^{-3}	0.366×10^{-3}	0.362×10^{-3}
	3	0.143×10^{-4}	-0.568×10^{-4}	0.483×10^{-4}
	4	-0.277×10^{-4}	-0.130×10^{-4}	-0.174×10^{-4}
	5	-0.481×10^{-4}	-0.233×10^{-4}	0.691×10^{-4}
	6	$-0.189\ 10^{-3}$	-0.678×10^{-4}	-0.992×10^{-4}

Table S11: SINGLE_ANISO computed crystal field parameters for each Dy(III) ion in complexes $\{Cu_3Dy_3\}$(**3**). The major components in the Table are in bold. B_k^q is the crystal field parameter and Q_k^q is the extended Stevens operator. The quantization axis is chosen to be the main axis of the pseudo-magnetic ground doublet.

k	q	B_k^q	B_k^q	B_k^q
		Dy1	Dy2	Dy3
2	-2	0.429	0.351	-0.121
	-1	-1.111	-0.987	-0.104
	0	-6.864	-6.92	-2.932
	1	-0.737×10^{-3}	0.631	0.195
	2	1.039	0.978	0.313
4	-4	-0.042	-0.103	0.005
	-3	-0.197	-0.153	-0.027
	-2	-0.014	-0.035	0.002
	-1	0.048	0.049	0.006
	0	0.012	0.012	0.002
	1	-0.011	-0.002	0.002
	2	-0.076	-0.071	-0.011
	3	0.054	0.141	-0.006
	4	-0.118	-0.069	-0.0192
6	-6	-0.002	-0.004	0.866×10^{-4}
	-5	-0.001	0.233×10^{-4}	-0.215×10^{-4}
	-4	-0.63×10^{-3}	-0.644×10^{-3}	0.128×10^{-4}
	-3	0.001	0.002	0.527×10^{-4}
	-2	0.002	0.004	-0.557×10^{-4}
	-1	-0.003	-0.003	-0.146×10^{-3}
	0	-0.677×10^{-3}	-0.666×10^{-3}	-0.276×10^{-4}
	1	0.414×10^{-3}	0.557×10^{-3}	-0.392×10^{-4}
	2	0.011	0.009	0.385×10^{-3}
	3	-0.533×10^{-3}	-0.002	0.436×10^{-4}
	4	-0.832×10^{-3}	-0.852×10^{-3}	-0.155×10^{-4}
	5	-0.691×10^{-4}	0.002	0.523×10^{-4}
	6	-0.004	-0.843×10^{-3}	-0.181×10^{-3}

Table S12: SINGLE_ANISO computed crystal field parameters for each Dy(III) ion in complexes{Ni_3Dy_3}(**5**). The major components in the Table are in bold. B_k^q is the crystal field parameter and Q_k^q is the extended Stevens operator. The quantization axis is chosen to be the main magnetic axis of the ground pseudo-doublet.

k	q	B_k^q	B_k^q	B_k^q
		Dy1	Dy2	Dy3
2	-2	1.642	-1.808	-1.73
	-1	-1.938	-1.879	-1.789
	0	-1.891	-1.921	-1.906
	1	-0.978	0.873	0.791
	2	1.142	1.029	0.981
4	-4	0.319 x 10^{-2}	0.483 x 10^{-3}	-0.395 x 10^{-3}
	-3	-0.4 x 10^{-2}	-0.515 x 10^{-2}	-0.182 x 10^{-2}
	-2	-0.942 x 10^{-2}	0.993 x 10^{-2}	0.972 x 10^{-2}
	-1	0.857 x 10^{-2}	0.945 x 10^{-2}	0.822 x 10^{-2}
	0	-0.201 x 10^{-2}	-0.209 x 10^{-2}	-0.194 x 10^{-2}
	1	0.011	-0.011	-0.011
	2	-0.323 x 10^{-2}	-0.186 x 10^{-2}	-0.271 x 10^{-2}
	3	-0.109 x 10^{-2}	-0.528 x 10^{-3}	0.155 x 10^{-2}
	4	-0.015	-0.015	-0.015
6	-6	-0.239 x 10^{-4}	0.51 x 10^{-4}	0.403 x 10^{-4}
	-5	0.464 x 10^{-3}	0.384 x 10^{-3}	0.376 x 10^{-3}
	-4	0.129 x 10^{-3}	-0.107 x 10^{-3}	-0.105 x 10^{-3}
	-3	0.103 x 10^{-3}	0.468 x 10^{-4}	0.126 x 10^{-4}
	-2	0.214 x 10^{-3}	-0.197 x 10^{-3}	-0.181 x 10^{-3}
	-1	0.106 x 10^{-3}	0.861 x 10^{-4}	0.1 x 10^{-3}
	0	-0.143 x 10^{-4}	-0.138 x 10^{-4}	-0.149 x 10^{-4}
	1	-0.651 x 10^{-4}	0.777 x 10^{-4}	0.774 x 10^{-4}
	2	-0.217 x 10^{-3}	-0.232 x 10^{-3}	-0.245 x 10^{-3}
	3	-0.356 x 10^{-3}	0.362 x 10^{-3}	0.365 x 10^{-3}
	4	-0.774 x 10^{-4}	-0.999 x 10^{-4}	-0.996 x 10^{-4}
	5	-0.246 x 10^{-3}	0.321 x 10^{-3}	0.353 x 10^{-3}
	6	-0.453 x 10^{-4}	-0.369 x 10^{-4}	-0.377 x 10^{-4}

Table S13: Lowest exchange doublets (cm^{-1}) corresponding to QTM/TAQT-M, and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Zn_3Dy_3\}$(**1**).

No.	E(cm^{-1})	QTM/TA-QTM	g_z
1	0	0.19×10^{-8}	39.547112309
2	0.30488 0.30488	0.26×10^{-8}	39.360131826
3	3.58402 3.58402	0.21×10^{-10}	39.553463369
4	4.23419 4.23419	0.18×10^{-9}	0.151399626
5	128.09638 128.09638	0.41×10^{-5}	36.495787817
6	128.29706 128.29706	0.28×10^{-3}	38.459259410
7	128.42693 128.42693	0.11×10^{-3}	37.921969120
8	128.76485 128.76485	0.25×10^{-3}	36.393454898
9	128.92082 128.92082	0.12×10^{-3}	37.216826185
10	129.14869 129.14869	0.14×10^{-4}	36.150019038
11	131.19299 131.19299	0.10×10^{-5}	37.063775026
12	131.67494 131.67494	0.97×10^{-4}	38.435960845
13	131.72672 131.72672	0.58×10^{-3}	3.500606505
14	132.14048 132.14048	0.95×10^{-5}	35.646659909
15	132.41837 132.41837	0.34×10^{-4}	3.191274357

Table S14: Lowest exchange doublets (cm^{-1}) corresponding to tunnel Splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Cu_3Dy_3\}$(**4**).

No.	Exchange doublets	Δ_{tun}	g_z
0	4.443100953866 4.443100953866	0.4 x 10^{-9}	39.320637111
1	0.000000000000 0.000000111608	1.1 x 10^{-9}	43.473095458
2	0.004753750736 0.004753902392	1.5 x 10^{-9}	43.480099934
3	0.017871284790 0.017871324858	0.4 x 10^{-9}	43.283661363
4	2.210967236501 2.210967362933	1.2 x 10^{-9}	39.495483401
5	2.212571546887 2.212571633058	0.8 x 10^{-9}	42.048574725
6	2.215428644228 2.215428808536	1.6 x 10^{-9}	42.019559631
7	2.218466339200 2.218466598040	2.5 x 10^{-9}	41.174763154
8	2.218909295197 2.218909427149	1.3 x 10^{-9}	41.233870636
9	2.221915598849 2.221915926485	3.2 x 10^{-9}	39.488266589
10	2.221915598849 2.221915926485	3.2 x 10^{-9}	41.036895119
11	2.230161385630 2.230161423526	0.3 x 10^{-9}	39.308175719
12	2.230719152089 2.230719197887	0.4 x 10^{-9}	41.852863471
13	2.234775971324 2.234776040175	0.6 x 10^{-9}	38.132163573
14	4.423539179599 4.423539275756	0.9 x 10^{-9}	37.166921771
15	4.429433658412 4.429433957083	2.9 x 10^{-9}	39.494027764
16	4.429583793421 4.429583932411	1.3 x 10^{-9}	39.483031438
17	4.431038375939 4.431038558290	1.8 x 10^{-9}	38.079952960
18	4.432591019086 4.432591382250	3.6 x 10^{-9}	37.229587283
19	4.436070713907 4.436070974960	2.6 x 10^{-9}	37.036549415

	4.443009581705		
21	4.447065706753 4.447065771312	0.6×10^{-9}	37.937626929
22	5.430173586395 5.430173588219	0.1×10^{-9}	0.132219462
23	6.642006259390 6.642006464158	2.0×10^{-9}	35.512563962
24	6.646745581225 6.646745858953	0.7×10^{-9}	35.520008254
25	6.659914120677 6.659914193684	0.7×10^{-9}	35.351609398
26	7.642175440123 7.642175441917	0.1×10^{-8}	4.096355223
27	7.644057548534 7.644057550301	0.1×10^{-8}	3.880648703
28	7.648390773569 7.648390775785	0.2×10^{-8}	4.032970660
29	9.856059119077 9.856059120815	0.1×10^{-8}	3.985641952
30	9.860392357454 9.860392359640	0.2×10^{-8}	4.103521396
31	9.862274462189 9.862274464347	0.2×10^{-8}	3.921000955
32	12.074275762886 12.074275765016	0.2×10^{-8}	0.107486358
33	127.964899602970 127.964973804412	0.7×10^{-4}	40.836010852

Table S15: Lowest exchange doublets (cm^{-1}) corresponding to QTM/TAQT-M, and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Ni_3Dy_3\}$(**7**).

No.	E(cm^{-1})	QTM/TA-QTM	g_z
1	1E-7 1E-7	0.32×10^{-6}	30.3739
2	0.00671 0.00671	0.56×10^{-6}	30.2461
3	0.04196 0.04196	0.45×10^{-7}	29.8817
4	5.50119 5.50119	0.67×10^{-9}	4.4953
5	5.62264 5.62264	0.96×10^{-6}	31.8414
6	5.62853 5.62853	0.13×10^{-4}	34.8076
7	5.63112 5.63112	0.23×10^{-4}	31.7918
8	5.6342 5.6342	0.13×10^{-5}	33.5408
9	5.6467 5.6467	0.28×10^{-5}	33.7377
10	5.65263 5.65263	0.43×10^{-5}	34.6829
11	5.66884 5.66884	0.75×10^{-6}	34.3383
12	5.67116 5.67116	0.79×10^{-6}	33.1601
13	5.68388 5.68388	0.66×10^{-6}	31.2975
14	11.12447 11.12447	0.53×10^{-6}	5.9766
15	11.12632 11.12632	0.16×10^{-6}	5.6868
16	11.14393 11.14393	0.11×10^{-7}	5.7766
17	11.25118 11.25118	0.14×10^{-4}	36.3406
18	11.2586 11.2586	0.23×10^{-4}	34.6633
19	11.26933 11.26933	0.75×10^{-5}	34.801
20	11.27523 11.27523	0.18×10^{-3}	38.0386
21	11.27704 11.27704	0.29×10^{-3}	36.312

Table S16: Lowest exchange doublets (cm^{-1}) corresponding to tunnel Splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Zn_3Tb_3\}$(**2**).

No.	E(cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.002517970366	2.5×10^{-3}	30.081602288
2	0.010019007416 0.014246048909	4.2×10^{-3}	28.563295461
3	0.026956640611 0.028665551808	1.7×10^{-3}	31.910219291
4	3.675911896234 3.675912056562	1.6×10^{-9}	0.391974788
5	177.129845351287 177.129953525399	1.0×10^{-4}	30.942110347
6	177.649879801049 177.650006051460	1.3×10^{-4}	30.784587758
7	177.819749453082 177.819900371870	1.5×10^{-4}	30.934316908

Table S17: Lowest exchange doublets (cm^{-1}) corresponding to QTM/TAQT-M, and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Cu_3Tb_3\}$(**5**).

No.	E(cm^{-1})	QTM/TA-QTM	g_z
1	0 0.22684	0.94×10^{-12}	4.0276
2	0.22729 0.23057	0.76×10^{-8}	4.0484
3	0.45413 0.4574	0.62×10^{-8}	4.1863
4	0.45786 0.68469	0.99×10^{-9}	4.0158
5	10.20186 10.30177	0.27×10^{-8}	4.1388
6	10.4041 10.4269	0.37×10^{-8}	4.1878
7	10.42958 10.44382	0.37×10^{-8}	1.1201
8	10.52579 10.53336	0.65×10^{-12}	34.8255
9	10.53784 10.63202	0.91×10^{-6}	34.7169
10	10.63528 10.6378	0.64×10^{-4}	32.4112
11	10.6546 10.66923	0.72×10^{-5}	38.1122
12	10.67271 10.75728	0.19×10^{-5}	35.2263
13	10.76171 10.76939	0.43×10^{-5}	29.6446
14	10.86356 10.86546	0.63×10^{-6}	38.0225
15	10.86356 10.86546	0.77×10^{-5}	35.0931
16	10.87005 10.89823	0.15×10^{-4}	31.6503
17	10.99317 11.09825	0.27×10^{-4}	32.8392
18	172.92158 173.11264	0.16×10^{-4}	38.4995
19	173.14476 173.14857	0.17×10^{-4}	28.8671
20	173.3365 173.33962	0.59×10^{-5}	38.7488
21	173.37176 173.55349	0.47×10^{-6}	32.6328

Table S18: Lowest exchange doublets (cm^{-1}) corresponding to tunnel Splitting (Δ_{tun}, cm^{-1}), and the g_z value of each doublet (g_x and $g_y = 0$) for complex $\{Ni_3Tb_3\}$(**8**).

No.	E(cm^{-1})	Δ_{tun}	g_z
1	0.000000000000 0.216718263089	0.22	17.7119
2	0.234133591441 0.436642499288	0.20	0.27332

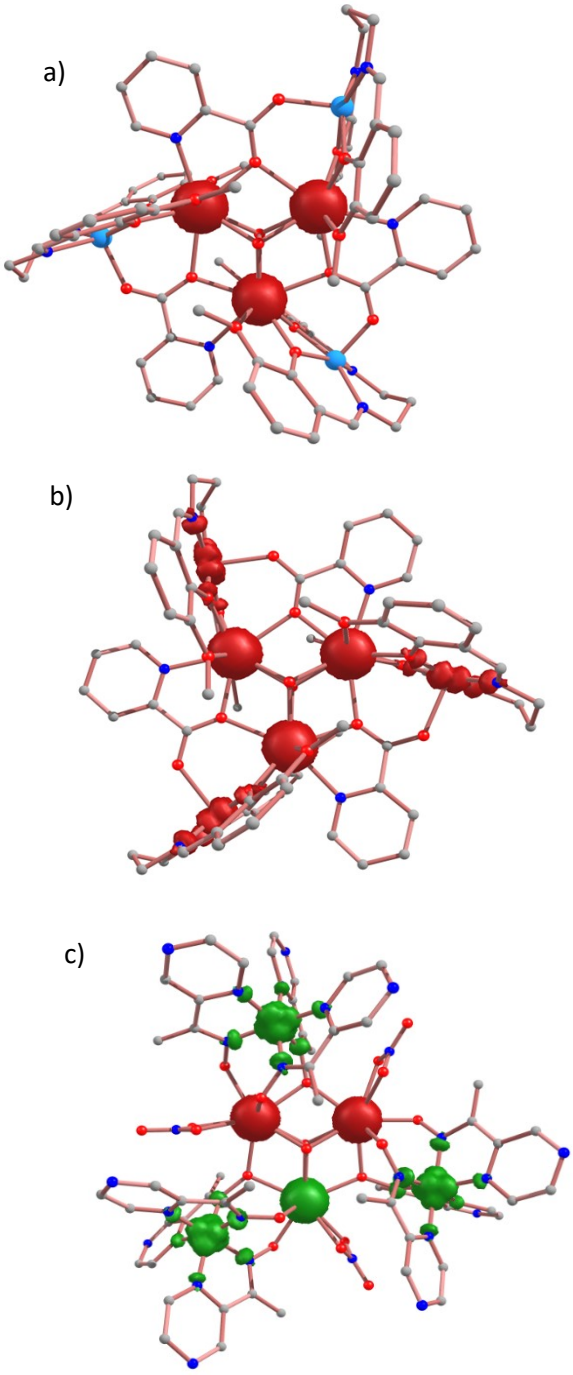

Figure S8: Spin density plot computed for complex a) **3,** b) **6,** and c) **9**. The red and green colors represent positive and negative spin densities, respectively.

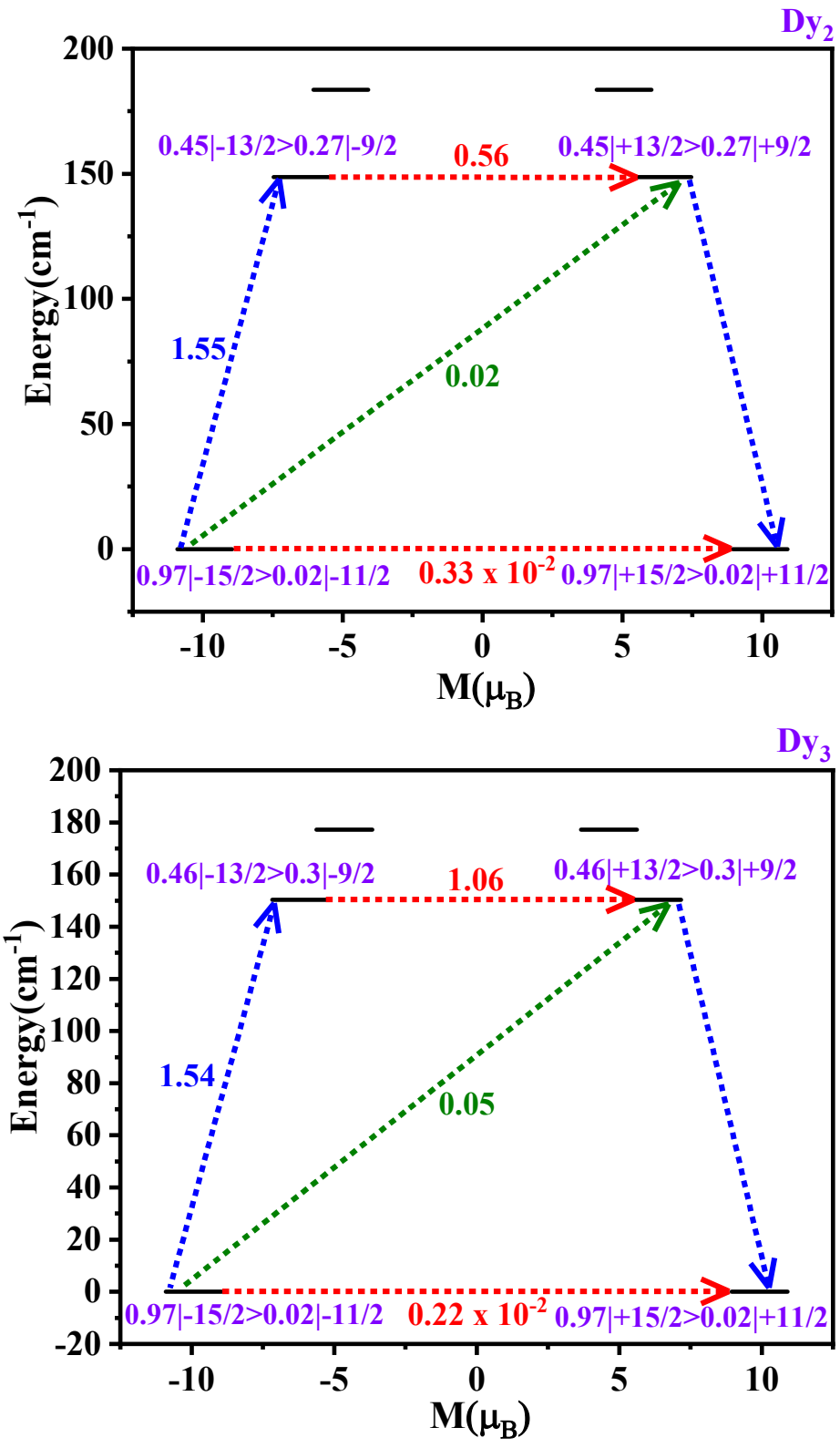

Figure S9: Magnetic relaxation mechanism of Dy2 and Dy3 centers in **1a.**

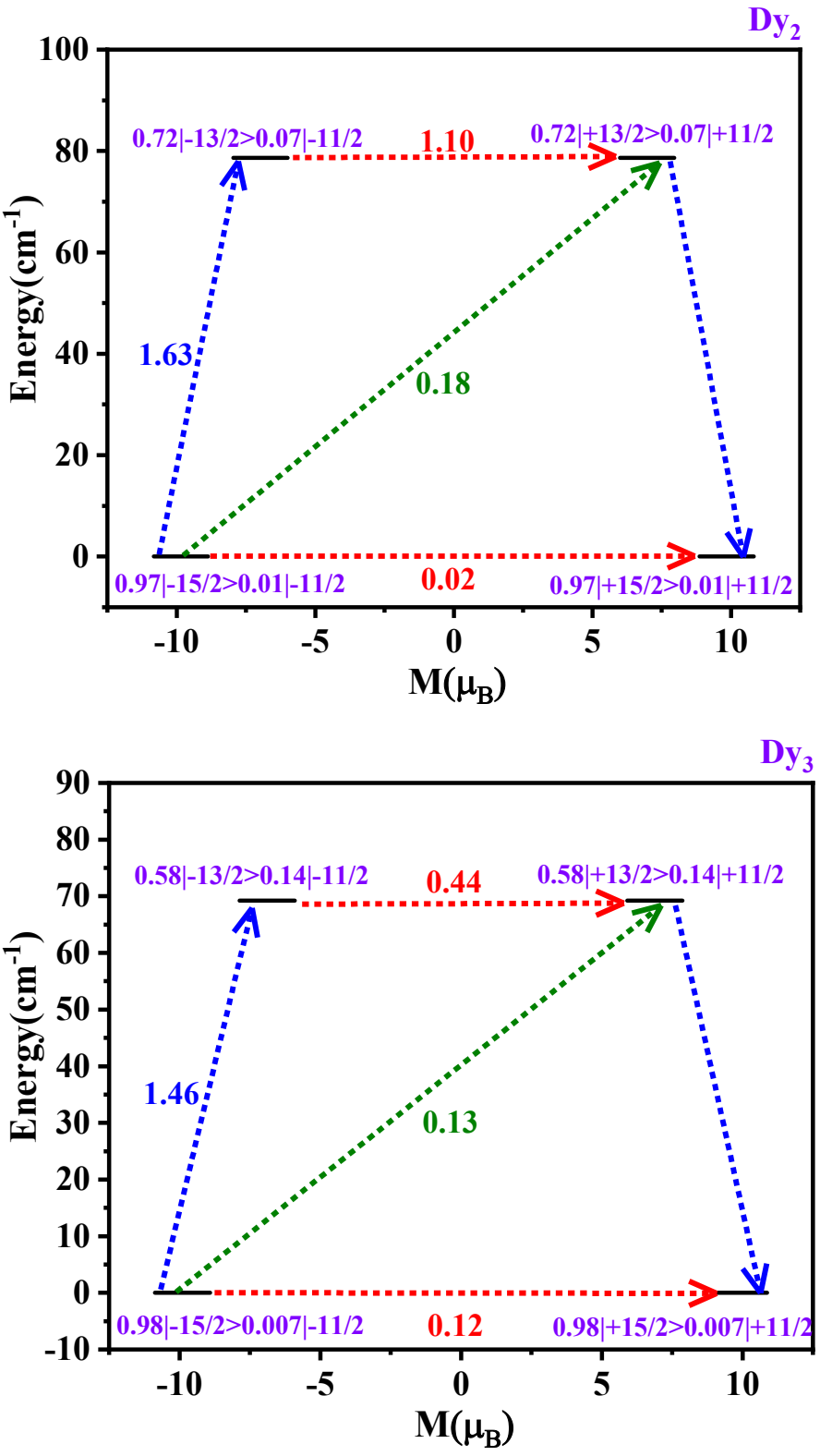

Figure S10: Magnetic relaxation mechanism of Dy2 and Dy3 centers in **10.**