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Supporting Information

The Role of Radical Bridges in Polynuclear Single-Molecule Magnets

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Table S1: Energies of the ground J-multiplet of Tb ion (cm^{-1}).

State	**1-Cp**	**1-N**	**1-Tb**	**2-Tb**
1	0.00	0.00	0.00	0.00
2	2.03E-05	7.72E-03	4.93E-02	5.37E-06
3	323.52	257.40	206.31	529.52
4	323.53	257.93	206.82	529.52
5	630.87	491.36	439.80	1082.40
6	630.94	504.44	443.56	1082.46
7	912.77	647.82	639.80	1652.68
8	913.46	739.92	682.19	1655.06
9	1139.81	779.21	815.49	2201.16
10	1165.40	1031.73	842.18	2247.82
11	1287.78	1032.33	899.80	2604.17
12	1378.00	1568.37	1007.83	3111.97
13	1407.05	1568.69	1013.10	3163.81

Table S2: ground g-tensor of the Tb ions.

	1-Cp	**1-N**	**1-Tb**	**2-Tb**
g_x	0.0000	0.0000	0.0000	0.0000
g_y	0.0000	0.0000	0.0000	0.0000
g_z	17.9516	17.8976	17.8712	17.9402

Note that the transverse components of g-tensor ($g_{x,y}$) of Tb ion are zero because of the Griffith's theorem.[1,2] As such, the magnetisation blocking performance is dictated by the magnitude of the tunnel splitting, i.e. the energy gap between the two low-lying spin-orbit energy states. These values are indicated in Table 1, main article.

Table S3: Modified CF states by exchange model (cm^{-1}).

State	**1-Tb**	**2-Tb**
1	0.00	0.00
2	0.01	8E-5
3	253.77	575.97
4	253.98	576.70
5	493.27	1136.52
6	498.82	1136.77
7	688.09	1695.06
8	718.10	1696.62
9	840.51	2246.77
10	866.72	2251.14
11	915.18	2610.49
12	1010.88	3124.79
13	1014.76	3172.82

Table S4: Energies of low-lying exchange doublets (cm^{-1}).

Doublet	**1-Tb**	**2-Tb**
1	0.000	0.000
2	225.272	235.764
3	225.272	235.764
4	312.660	480.171
5	329.546	638.442
6	443.523	649.165
7	443.523	756.236
8	482.681	756.236

Table S5: g-tensor of the ground exchange doublet.

	1-Tb	**2-Tb**
g_x	0.0000	0.0000
g_y	0.0000	0.0000
g_z	33.7960	33.8836

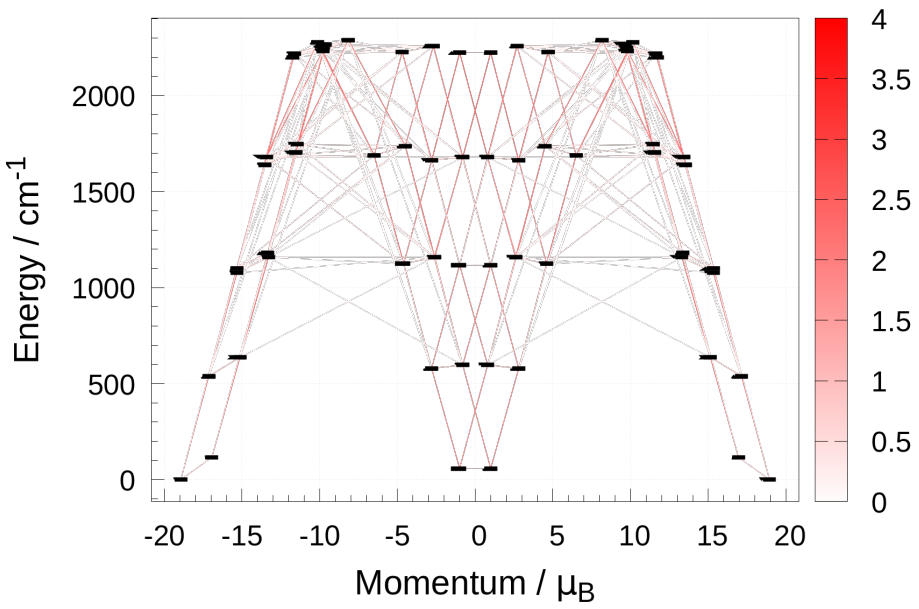

Figure S1: The simulation of magnetization blocking diagram in **2-Tb** using the Lines model with $J_{ex} = +20$ cm^{-1} ($\mathcal{H}_{eff}^{Lines} = \sum_i^2 -J_i \tilde{s}_i \hat{S}_R$ where $\tilde{s}_i$ is the speudospin of Ising doublets of the Tb ions and $\hat{S}_R$ is the spin operator of the radical). The red lines represent the mean absolute value of the transition magnetic dipole moment connecting respective states. The magnitude of the transition dipole moment is directly linked with the intensity of the color line, following the legend on the right of the plot.

Table S6: Contribution of local CF doublets on each Tb site to exchange doublets for **1-Tb** (only the contribution larger or equal to 5% is reported).

Local CF doublet	Exchange doublet					
	1	2	3	4	5	6
$\|\pm6\rangle$	99%	96%	96%	49%	50%	53%
$\|\pm5\rangle$				51%	50%	47%
$\|\pm4\rangle$						
$\|\pm3\rangle$						
$\|\pm2\rangle$						
$\|\pm1\rangle$						
$\|0\rangle$						

Table S7: Contribution of local CF doublets on each Tb site to exchange doublets for **2-Tb** (only the contribution larger or equal to 5% is reported).

Local CF doublet	Exchange doublet					
	1	2	3	4	5	6
$\|\pm6\rangle$	100%	99%	99%	100%	49%	50%
$\|\pm5\rangle$					51%	50%
$\|\pm4\rangle$						
$\|\pm3\rangle$						
$\|\pm2\rangle$						
$\|\pm1\rangle$						
$\|0\rangle$						

Table S8: Atomic coordinates for **1-Tb** in *ab initio* calculations

Atom	Cartesian coordinates (Å)		
	x	y	z
Tb1	0.00698	0	2.10869
Lu1	-0.00698	0	-2.10869
N1	0.68893	0	0.00228
N2	-0.702	0.05521	0.00741
C1	-1.0998	2.07491	3.47191
C2	-0.59366	2.67871	2.31027
C3	0.82775	2.59863	2.37339
C4	1.17725	1.95008	3.58404
C5	-0.0025	1.62946	4.27013
C6	-2.56389	1.99412	3.86471
C7	-1.39045	3.32051	1.20901
C8	1.78869	3.16399	1.3972
C9	2.57773	1.72659	4.10037
C10	1.06943	-2.36675	2.84504
C11	0.67637	-1.81388	4.09311
C12	-0.73631	-1.74445	4.10831
C13	-1.21308	-2.2716	2.9044
C14	-0.11598	-2.64414	2.12204
C15	2.47241	-2.66595	2.42177
C16	1.58526	-1.52796	5.25234
C17	-1.56994	-1.36723	5.28856
C18	-2.65786	-2.40676	2.52155
C19	1.55502	2.04433	-2.88183

C20	1.03101	1.5836	-4.11024
C21	-0.3653	1.87393	-4.13578
C22	-0.68335	2.51542	-2.91788
C23	0.4778	2.62852	-2.1397
C24	2.97494	1.91793	-2.38748
C25	1.7974	1.07386	-5.2921
C26	-1.28451	1.61422	-5.30302
C27	-2.04501	3.04862	-2.53885
C28	1.06692	-2.12806	-3.41081
C29	0.5562	-2.67036	-2.18938
C30	-0.86287	-2.60585	-2.27944
C31	-1.21237	-2.02417	-3.49938
C32	-0.0317	-1.74161	-4.20483
C33	2.53409	-2.05014	-3.8045
C34	1.37592	-3.24846	-1.06973
C35	-1.81589	-3.12318	-1.24164
C36	-2.59677	-1.8756	-4.05343
H1	-0.05766	1.19439	5.11354
H2	-3.05573	1.47335	3.19508
H3	-2.9382	2.89895	3.91295
H4	-2.6413	1.5607	4.7397
H5	-1.46544	2.7005	0.45487
H6	-0.93778	4.13954	0.91677
H7	-2.28404	3.54297	1.53897
H8	2.63215	3.3758	1.85442
H9	1.41556	3.98125	1.00499
H10	1.95686	2.50922	0.68853
H11	2.55109	1.5692	5.06692
H12	3.12441	2.51904	3.91556
H13	2.96901	0.9478	3.65407
H14	-0.16102	-3.01901	1.24973
H15	2.77788	-1.97448	1.79899
H16	2.49913	-3.54001	1.97827
H17	3.05763	-2.68009	3.20877
H18	2.51076	-1.47447	4.93835
H19	1.50833	-2.24809	5.91362
H20	1.33109	-0.67705	5.66492
H21	-1.03032	-0.84067	5.91548
H22	-1.89018	-2.17844	5.73407

H23	-2.3374	-0.83438	4.98792
H24	-3.01983	-1.52467	2.29442
H25	-3.15874	-2.78348	3.27239
H26	-2.73708	-3.00073	1.74528
H27	0.54195	3.02147	-1.27695
H28	2.97371	1.7528	-1.42087
H29	3.4607	2.74727	-2.57454
H30	3.41334	1.17044	-2.8437
H31	2.68714	0.78369	-5.00222
H32	1.88404	1.78943	-5.95595
H33	1.31724	0.31704	-5.68983
H34	-1.38703	0.6485	-5.42967
H35	-0.90463	2.01222	-6.11267
H36	-2.16127	2.01513	-5.12578
H37	-2.63667	2.30173	-2.31062
H38	-2.42629	3.54528	-3.29498
H39	-1.95719	3.64558	-1.76675
H40	0.01569	-1.3544	-5.07139
H41	2.60071	-1.90438	-4.77121
H42	2.98113	-2.88841	-3.56656
H43	2.96124	-1.30567	-3.33042
H44	2.27044	-3.46774	-1.39933
H45	0.94255	-4.06183	-0.73688
H46	1.44642	-2.59399	-0.34449
H47	-1.83219	-2.50783	-0.47952
H48	-1.52095	-4.00838	-0.93949
H49	-2.71383	-3.19156	-1.62966
H50	-3.07466	-1.1802	-3.55595
H51	-3.07601	-2.7261	-3.96961
H52	-2.54822	-1.6222	-4.9994

Table S9: Atomic coordinates for **1-Cp** in *ab initio* calculations

Atom	Cartesian coordinates (Å)		
	x	y	z
Tb1	0.00698	0	2.10869
C1	-1.0998	2.07491	3.47191
C2	-0.59366	2.67871	2.31027

C3	0.82775	2.59863	2.37339
C4	1.17725	1.95008	3.58404
C5	-0.0025	1.62946	4.27013
C6	-2.56389	1.99412	3.86471
C7	-1.39045	3.32051	1.20901
C8	1.78869	3.16399	1.3972
C9	2.57773	1.72659	4.10037
C10	1.06943	-2.36675	2.84504
C11	0.67637	-1.81388	4.09311
C12	-0.73631	-1.74445	4.10831
C13	-1.21308	-2.2716	2.9044
C14	-0.11598	-2.64414	2.12204
C15	2.47241	-2.66595	2.42177
C16	1.58526	-1.52796	5.25234
C17	-1.56994	-1.36723	5.28856
C18	-2.65786	-2.40676	2.52155
H1	-0.05766	1.19439	5.11354
H2	-3.05573	1.47335	3.19508
H3	-2.9382	2.89895	3.91295
H4	-2.6413	1.5607	4.7397
H5	-1.46544	2.7005	0.45487
H6	-0.93778	4.13954	0.91677
H7	-2.28404	3.54297	1.53897
H8	2.63215	3.3758	1.85442
H9	1.41556	3.98125	1.00499
H10	1.95686	2.50922	0.68853
H11	2.55109	1.5692	5.06692
H12	3.12441	2.51904	3.91556
H13	2.96901	0.9478	3.65407
H14	-0.16102	-3.01901	1.24973
H15	2.77788	-1.97448	1.79899
H16	2.49913	-3.54001	1.97827
H17	3.05763	-2.68009	3.20877
H18	2.51076	-1.47447	4.93835
H19	1.50833	-2.24809	5.91362
H20	1.33109	-0.67705	5.66492
H21	-1.03032	-0.84067	5.91548
H22	-1.89018	-2.17844	5.73407
H23	-2.3374	-0.83438	4.98792

H24	-3.01983	-1.52467	2.29442
H25	-3.15874	-2.78348	3.27239
H26	-2.73708	-3.00073	1.74528

Table S10: Atomic coordinates for **1-N** in *ab initio* calculations

Atom	Cartesian coordinates (Å)		
	x	y	z
Tb1	0.00698	0	2.10869
Lu1	-0.00698	0	-2.10869
N1	0.68893	0	0.00228
N2	-0.702	0.05521	0.00741

Table S11: Atomic coordinates for **2-Tb** in *ab initio* calculations

Atom	Cartesian coordinates (Å)		
	x	y	z
Tb1	0.00698	0	2.10869
Lu1	-0.00698	0	-2.10869
N1	0.68893	0	0.00228
N2	-0.702	0.05521	0.00741
O5	-0.02880	0.00000	-4.34390
O6	0.02850	0.00000	4.34390

References

[1] J. S. Griffith, *Phys. Rev.*, 1963, **132**, 316–319.

[2] J. S. Griffith, *The Theory of Transition-Metal Ions*, Cambridge University Press, 1964.