Electronic Supplementary Information

Elemental analyses (Elemental vario MICRO cube Elemental Analyzer, silver boats) calc. (found) for **1** ($C_{10}H_{14}DyNO_{12}$, M_r = 502.72): 23.89 (23.73) % C, 2.79 (2.69) % N, 2.81 (2.61) % H.

FT-IR (Bruker Tensor 27 FT-IR (Bruker Optik GmbH., Germany) spectrometer using the KBr technique in the range of 4000 – 400 cm^{-1})
1: 3500(m); 3376(s); 3080(vw); 3001(vw); 2927(vw); 1606(ssh); 1597(s); 1500(s); 1458(m); 1433(m); 1408(m); 1382(m); 1343(m); 1315(m); 1246(m); 1191(m); 1122(m); 1034(m); 946(w); 830(m); 817(w); 761(m); 696(w); 584(w); 532(w); 437(w).

Single-crystal X-ray structure analysis (Oxford Diffraction XCalibur equipped with Sapphire 3 CCD detector). The multi-scan method[1] was used for absorption correction of **1**.

Table S1. Crystal data and structure refinement for **1**

Empirical formula	$C_{10}H_{14}DyNO_{12}$
Formula weight	502.72
Temperature (K)	123(2)
Crystal system, space group	monoclinic, $P2_1/c$
Unit cell dimensions (Å, °)	a = 14.53519(15)
	b = 11.48172(11) β = 99.0238(9)
	c = 8.90846(8)
V (Å^3)	1468.32(2)
Z, Calculated density (Mg/m^3)	4, 2.274
μ (mm^{-1})	5.158
Crystal size (mm)	0.227 x 0.165 x 0.082
Crystal color / form	colorless / block
θ range for data collection (°)	3.081 - 25.995
Limiting indices	$-17 \le h \le 17$, $-14 \le k \le 14$, $-10 \le l \le 10$
Reflections collected / unique	46504 / 2875 [R_{int} = 0.0550]
Completeness to θ = 25.242°	99.8 %
Data / restraints / parameters	2875 / 9 / 235
S on F^2	1.079
Final R indices [$I > 2\sigma_I$]	R_1 = 0.0179, wR_2 = 0.0435
R indices (all data)	R_1 = 0.0194, $wR2$ = 0.0440
Largest diff. peak and hole (e.Å^3)	0.633 and –1.010

[1] Blessing, R. H. *Acta Cryst. A.* **1995**, 51, 33-38.

Table S2 Selected geometric parameters (Å, °) for **1**.

Dy1-O1	2.3398(18)	O1-Dy1-O3	64.94(6)
Dy1-O3	2.4699(17)	O3-Dy1-O4	63.57(6)
Dy1-O4	2.4321(17)	O4-Dy1-O5	66.03(6)
Dy1-O5	2.3350(17)	O7-Dy1-O8	49.98(6)
Dy1-O6^{i}	2.3308(18)	O5-Dy1-O7	146.92(6)
Dy1-O7	2.4898(19)	O7-Dy1-O6^{i}	73.30(6)
Dy1-O8	2.606(2)	O8-Dy1-O6^{i}	66.29(6)
Dy1-O10	2.3150(18)	O10-Dy1-O11	89.50(7)
Dy1-O11	2.4024(18)		
O1-C1	1.265(3)		
O2-C1	1.240(3)		
O5-C10	1.258(3)		
O6-C10	1.250(3)		
O7-N1	1.283(3)		
O8-N1	1.254(3)		
O9-N1	1.222(3)		

Symmetry code: i: x, 0.5 – y, –0.5 + z.

Table S3 Possible hydrogen bonds for **1** (Å, °).

D---H···A	D-H	H···A	D···A	D-H···A
O10-H110···O21^{iv}	0.834(10)	1.909(16)	2.701(3)	158(3)
O10-H210···O12	0.839(10)	1.974(12)	2.802(3)	169(3)
O11-H111···O2^{v}	0.832(10)	2.013(10)	2.844(3)	175(3)
O11-H211···O12^{ii}	0.832(10)	2.005(13)	2.825(3)	168(3)
O12-H112···O1^{vi}	0.835(10)	2.002(13)	2.804(3)	161(3)
O12- H212···O7^{ii}	0.837(10)	2.065(13)	2.891(3)	169(3)

Symmetry codes: ii: x, 0.5 – y, 0.5 + z; iv: –x, –y, –z; v: x, –0.5 – y, 0.5 + z; vi: –x, 0.5 + y, 0.5 – z.

Table S4. Results of the fitting procedure for AC susceptibility components of **1** at B_{DC} = 0.15 with a three-set Debye model.

T/K	$R(\chi')$ /%	$R(\chi'')$ /%	χ_S	χ_{LF}	α_{LF}	τ_{LF} / s	χ_{IF}	α_{IF}	τ_{IF} /10^{-3} s	χ_{HF}	α_{HF}	τ_{HF} /10^{-6} s	x_{LF}	x_{IF}	x_{HF}
1.9	1.8	0.60	8.7	37(7)	.11	**1.31**(13)	67(2)	.25	85(20)	72(3)	.26	438(111)	.45	.47	.07
2.1	1.1	0.82	7.9	27(3)	.00	0.87(4)	57(1)	.26	72(9)	64(1)	.31	516(80)	.36	.53	.11
2.3	1.4	1.6	7.6	36(5)	.15	0.62(7)	55(1)	.23	43(11)	62(1)	.29	606(115)	.51	.35	.13
2.7	1.4	0.80	6.3	23(4)	.04	0.47(4)	43(2)	.29	38(12)	53(1)	.29	641(89)	.37	.42	.22
3.1	1.1	1.8	5.8	17(4)	.01	0.35(3)	33(2)	.34	30(14)	47(1)	.27	647(46)	.27	.39	.34
3.5	0.75	1.1	5.1	12(4)	.03	0.35(5)	23(1)	.32	37(20)	42(1)	.27	595(22)	.18	.30	.52
3.9	0.85	1.4	[5.1]	9(3)	.00	0.32(7)	16(1)	.27	34	37(1)	.22	450(27)	.13	.21	.66
4.3	0.56	1.4	[5.1]	7(2)	.00	0.40(8)	13(1)	.35	35	34(1)	.15	283(7)	.08	.19	.73
4.7	0.44	1.1	[5.1]	6(2)	.00	0.35(6)	11(1)	.44	29	32(1)	.09	150(2)	.05	.17	.78
5.1c	0.38	0.87	[5.1]	6(1)	.00	0.32(10)	10(1)	.57	27	29(1)	.05	72.7(8)	.02	.17	.81
5.5c	0.33	2.0	[5.1]	5.6(11)	.00	0.40(10)	8.2(9)	.55	19	27(1)	.04	32.6(11)	.02	.12	.86
5.9	0.28	3.0	[5.1]	5.5(8)	.02	0.38(11)	7.4(11)	.60	12	25(1)	.01	14.9(16)	.02	.09	.89

Table S5 Electronic g-matrix from effective Hamiltonian.

g-matrix:

1.897916	0.596428	0.493404
0.950541	3.293627	1.196481
0.230923	1.392603	2.963724

g-factors:

1.510550 1.999175 4.677841 iso = 2.729189

g-shifts:

-0.491770 -0.003144 2.675522 iso = 0.726869

Orientation:

X	0.8243412	0.4938292	0.2767567
Y	-0.5129985	0.4449468	0.7340674
Z	0.2393619	-0.7470977	0.6201216

Table S6 SOC corrected transition energies

	ΔE/cm^{-1}	ΔE/eV	Boltzmann populations at T = 300 K
0:	0.00	0.0000	2.24e-01
1:	0.00	0.0000	2.24e-01
2:	154.79	0.0192	1.07e-01
3:	154.79	0.0192	1.07e-01
4:	290.24	0.0360	5.57e-02
5:	290.24	0.0360	5.57e-02
6:	369.92	0.0459	3.80e-02
7:	369.92	0.0459	3.80e-02
8:	432.52	0.0536	2.81e-02
9:	432.52	0.0536	2.81e-02
10:	511.11	0.0634	1.93e-02
11:	511.11	0.0634	1.93e-02
12:	563.83	0.0699	1.50e-02
13:	563.83	0.0699	1.50e-02
14:	585.85	0.0726	1.35e-02
15:	585.85	0.0726	1.35e-02
16:	2993.23	0.3711	1.31e-07
17:	2993.23	0.3711	1.31e-07
18:	3144.25	0.3898	6.33e-08
19:	3144.25	0.3898	6.33e-08
20:	3240.38	0.4018	3.99e-08
21:	3240.38	0.4018	3.99e-08
22:	3307.21	0.4100	2.90e-08
23:	3307.21	0.4100	2.90e-08
24:	3360.89	0.4167	2.24e-08

25:	3360.89	0.4167	2.24e-08
26:	3396.90	0.4212	1.88e-08
27:	3396.90	0.4212	1.88e-08
28:	3451.89	0.4280	1.45e-08
29:	3451.89	0.4280	1.45e-08
30:	5572.70	0.6909	5.53e-13
31:	5572.70	0.6909	5.53e-13
32:	5704.21	0.7072	2.94e-13
33:	5704.21	0.7072	2.94e-13
34:	5770.49	0.7154	2.14e-13
35:	5770.49	0.7154	2.14e-13
36:	5836.46	0.7236	1.56e-13
37:	5836.46	0.7236	1.56e-13
38:	5872.09	0.7280	1.32e-13
39:	5872.09	0.7280	1.32e-13
40:	5968.29	0.7400	8.30e-14
41:	5968.29	0.7400	8.30e-14
42:	7746.97	0.9605	1.64e-17
43:	7746.97	0.9605	1.64e-17
44:	7847.30	0.9729	1.01e-17
45:	7847.30	0.9729	1.01e-17
46:	7929.18	0.9831	6.84e-18
47:	7929.18	0.9831	6.84e-18
48:	7973.16	0.9885	5.54e-18
49:	7973.16	0.9885	5.54e-18
50:	8082.71	1.0021	3.27e-18
51:	8082.71	1.0021	3.27e-18
52:	9489.37	1.1765	3.85e-21
53:	9489.37	1.1765	3.85e-21
54:	9609.81	1.1915	2.16e-21
55:	9609.81	1.1915	2.16e-21
56:	9656.29	1.1972	1.73e-21
57:	9656.29	1.1972	1.73e-21
58:	9695.78	1.2021	1.43e-21
59:	9695.78	1.2021	1.43e-21
60:	9735.33	1.2070	1.18e-21
61:	9735.33	1.2070	1.18e-21
62:	9764.38	1.2106	1.03e-21
63:	9764.38	1.2106	1.03e-21
64:	9784.44	1.2131	9.35e-22
65:	9784.44	1.2131	9.35e-22
66:	9817.84	1.2173	7.96e-22
67:	9817.84	1.2173	7.96e-22
68:	9833.85	1.2192	7.37e-22
69:	9833.85	1.2192	7.37e-22
70:	9868.68	1.2236	6.24e-22
71:	9868.68	1.2236	6.24e-22
72:	10826.92	1.3424	6.30e-24
73:	10826.92	1.3424	6.30e-24
74:	11085.23	1.3744	1.83e-24
75:	11085.23	1.3744	1.83e-24
76:	11167.33	1.3846	1.23e-24
77:	11167.33	1.3846	1.23e-24
78:	11852.14	1.4695	4.61e-26

79:	11852.14	1.4695	4.61e-26
80:	11876.59	1.4725	4.10e-26
81:	11876.59	1.4725	4.10e-26
82:	11897.30	1.4751	3.71e-26
83:	11897.30	1.4751	3.71e-26
84:	11932.20	1.4794	3.14e-26
85:	11932.20	1.4794	3.14e-26
86:	11961.14	1.4830	2.73e-26
87:	11961.14	1.4830	2.73e-26
88:	13599.25	1.6861	1.06e-29
89:	13599.25	1.6861	1.06e-29
90:	13624.76	1.6893	9.37e-30
91:	13624.76	1.6893	9.37e-30
92:	13659.48	1.6936	7.93e-30
93:	13659.48	1.6936	7.93e-30
94:	13695.97	1.6981	6.66e-30
95:	13695.97	1.6981	6.66e-30
96:	14971.87	1.8563	1.47e-32
97:	14971.87	1.8563	1.47e-32
98:	15005.40	1.8604	1.25e-32
99:	15005.40	1.8604	1.25e-32
100:	15022.12	1.8625	1.15e-32
101:	15022.12	1.8625	1.15e-32
102:	15964.61	1.9794	1.25e-34
103:	15964.61	1.9794	1.25e-34
104:	15976.09	1.9808	1.19e-34
105:	15976.09	1.9808	1.19e-34
106:	16554.23	2.0525	7.42e-36
107:	16554.23	2.0525	7.42e-36
108:	38840.47	4.8156	2.83e-82
109:	38840.47	4.8156	2.83e-82
110:	38896.89	4.8226	2.16e-82
111:	38896.89	4.8226	2.16e-82
112:	38996.57	4.8350	1.34e-82
113:	38996.57	4.8350	1.34e-82
114:	39233.15	4.8643	4.30e-83
115:	39233.15	4.8643	4.30e-83
116:	40218.00	4.9864	3.82e-85
117:	40218.00	4.9864	3.82e-85
118:	40434.59	5.0133	1.35e-85
119:	40434.59	5.0133	1.35e-85
120:	40588.88	5.0324	6.45e-86
121:	40588.88	5.0324	6.45e-86
122:	41355.63	5.1274	1.63e-87
123:	41355.63	5.1274	1.63e-87
124:	41437.37	5.1376	1.10e-87
125:	41437.37	5.1376	1.10e-87

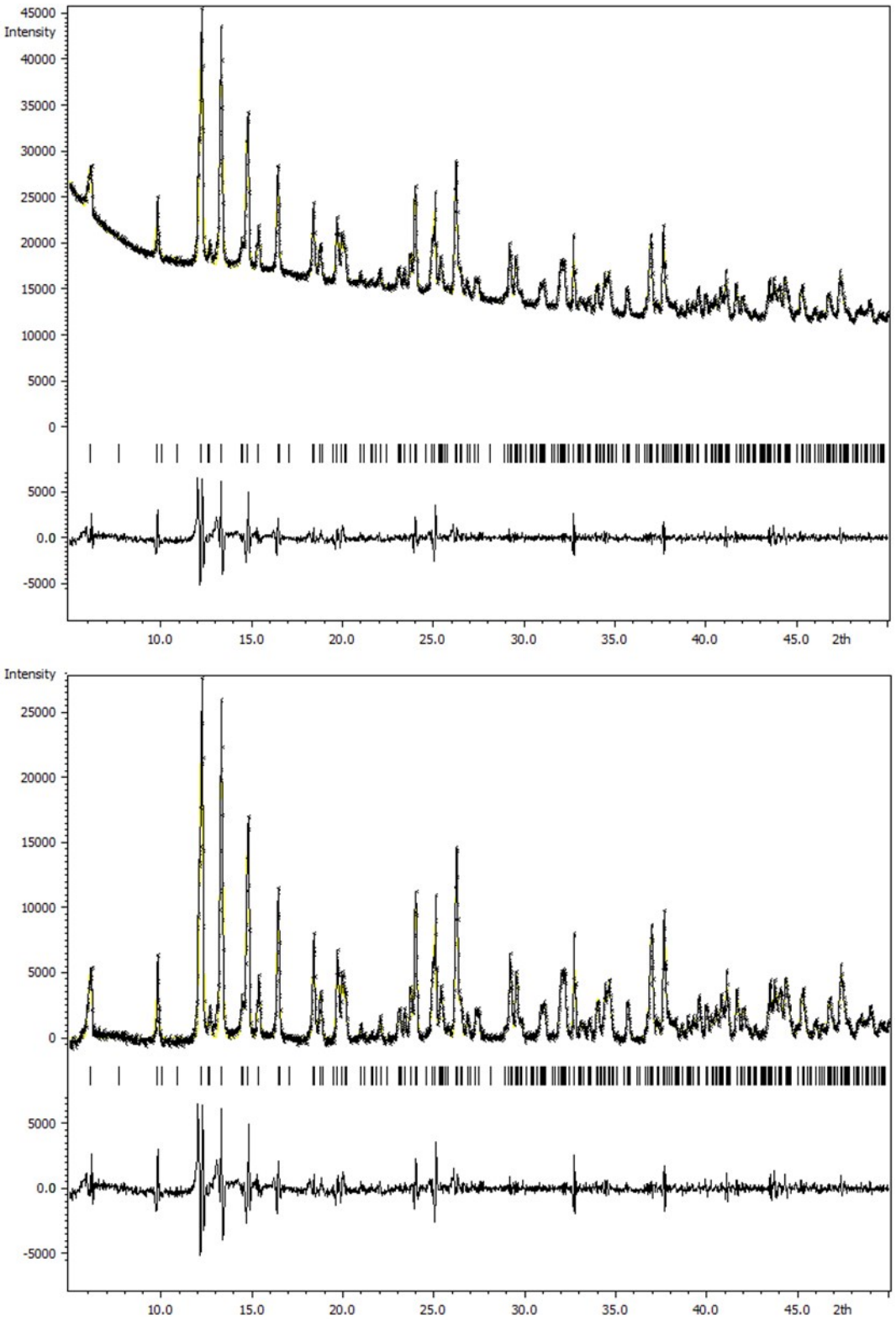

Figure S1 Results of Le Bail refinement of the powder diffraction pattern of **1**, with the unit-cell parameters taken from the known crystal structure of **1**, see **Table S1**). The refined cell parameters are $a = 14.6562\ (12)$ Å, $b = 11.5035\ (8)$ Å, $c = 8.9000\ (8)$ Å, $\beta = 99.407\ (5)°$, $R(p) = 0.0198$, $R(wp) = 0.0321$, goodness of fit = 4.10. Upper figure: uncorrected base line; bottom figure: with corrected base line.

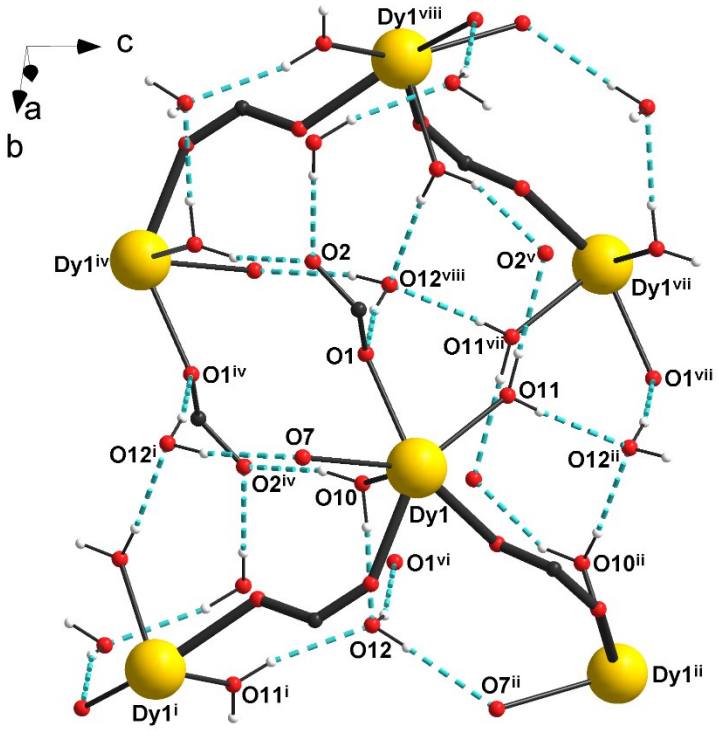

Figure S2 Hydrogen bonding system in **1**. Only relevant atoms are shown for clarity. The zig-zag chains of Dy(III) atoms are shown by wire model and hydrogen bonds are shown as blue dashed lines. Symmetry codes: i: *x*, 0.5 – *y*, –0.5 + *z*; ii: *x*, 0.5 – *y*, 0.5 + *z*; iii: *x*, *y*, 1 + *z*; iv: -*x*, -*y*, -*z*; v: *x*, –0.5 – *y*, 0.5 + *z*; vi: -*x*, 0.5 + *y*, 0.5 – *z*; vii: -*x*, -*y*, 1- *z*; viii: -*x*, -0.5 + *y*, 0.5 - *z*.

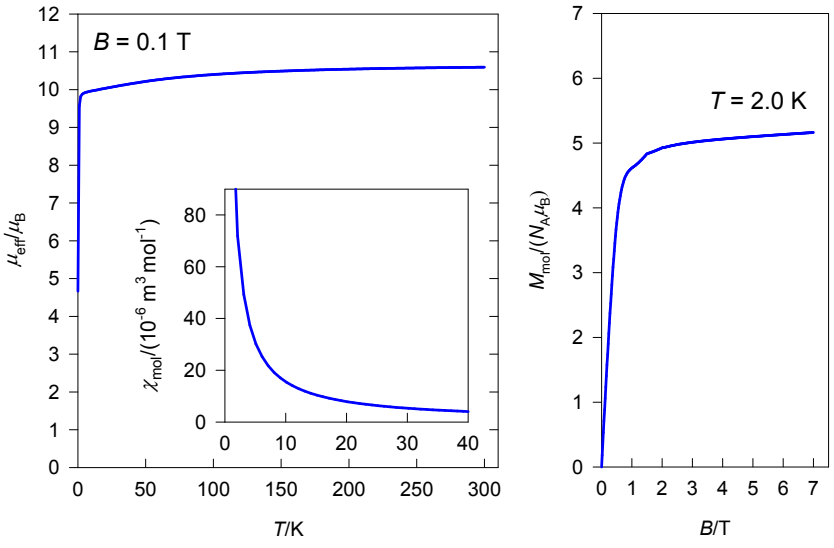

Figure S3 *Ab initio* magnetic data for complex **1**.

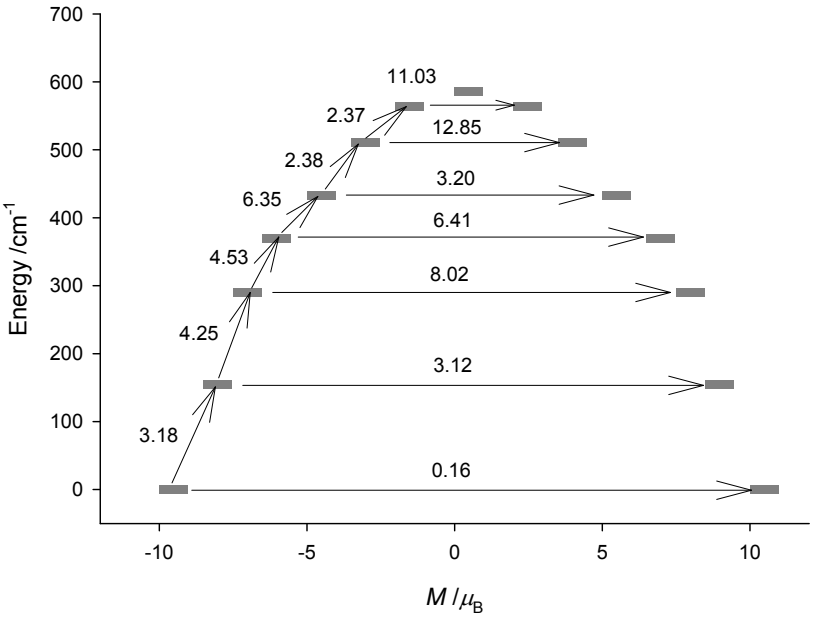

Figure S4. Energies of Kramers doublets and transition moments for **1**. Arrows correspond to the direct and quantum tunneling relaxation processes.