Supporting Information

Effect of axial coordination environment on quantum tunnelling magnetization for dysprosium single ion magnet with theoretical insight

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Table S1. Crystal data and structure refinement for Complexes **1** and **2**.

Identification code	Complex **1**	Complex **2**
Empirical formula	$C_{51}H_{59}B_2ClDyN_8O_7$	$C_{28}H_{31}ClDyN_8O_4S_2$
Formula weight	1115.63	805.68
Temperature/K	296.15	140.01
Crystal system	monoclinic	monoclinic
Space group	$P2_1/n$	$P2_1/c$
a/Å	11.9371(2)	21.4811(16)
b/Å	19.0224(4)	9.7468(9)
c/Å	22.7876(4)	15.2004(13)
α/°	90	90
β/°	91.5360(10)	100.532(2)
γ/°	90	90
Volume/Å^3	5172.57(17)	3128.9(5)
Z	4	4
ρ_{calc}g/cm^3	1.433	1.710
μ/mm^{-1}	1.554	2.655
F(000)	2280.0	1608.0
Crystal size/mm^3	0.72 × 0.56 × 0.47	0.71 × 0.57 × 0.43
Radiation	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)
2θ range for data collection/°	2.79 to 51.652	4.602 to 51.55
Index ranges	-14 ≤ h ≤ 14, -23 ≤ k ≤ 19, -27 ≤ l ≤ 27	-26 ≤ h ≤ 26, -11 ≤ k ≤ 11, -18 ≤ l ≤ 18
Reflections collected	30468	58760
Independent reflections	9924 [R_{int} = 0.0373, R_{sigma} = 0.0449]	5993 [R_{int} = 0.0525, R_{sigma} = 0.0265]
Data/restraints/parameters	9924/0/639	5993/6/407
Goodness-of-fit on F^2	1.027	1.056
Final R indexes [I>=2σ (I)]	R_1 = 0.0320, wR_2 = 0.0615	R_1 = 0.0224, wR_2 = 0.0415
Final R indexes [all data]	R_1 = 0.0413, wR_2 = 0.0648	R_1 = 0.0297, wR_2 = 0.0435
Largest diff. peak/hole / e Å^{-3}	0.53/-0.48	0.62/-0.59

Table S2. Shape analysis for the studied complexes.

1	EP-9	D9h	Enneagon
2	OPY-9	C8v	Octagonal pyramid
3	HBPY-9	D7h	Heptagonal bipyramid
4	JTC-9	C3v	Johnson triangular cupola J3
5	JCCU-9	C4v	Capped cube J8
6	CCU-9	C4v	Spherical-relaxed capped cube
7	JCSAPR-9	C4v	Capped square antiprism J10
8	CSAPR-9	C4v	Spherical capped square antiprism
9	JTCTPR-9	D3h	Tricapped trigonal prism J51
10	TCTPR-9	D3h	Spherical tricapped trigonal prism
11	JTDIC-9	C3v	Tridiminished icosahedron J63
12	HH-9	C2v	Hula-hoop
13	MFF-9	Cs	Muffin

Complex **1**											
EP-9	OPY-9	HBPY-9	JTC-9	JCCU-9	CCU-9	JCSAPR-9	CSAPR-9	JTCTPR-9	TCTPR-9	JTDIC-9	HH-9	MFF-9
37.386	22.997	14.592	16.309	5.121	3.734	3.942	**3.235**	5.432	4.033	9.918	8.005	3.929
Complex **2**											
EP-9	OPY-9	HBPY-9	JTC-9	JCCU-9	CCU-9	JCSAPR-9	CSAPR-9	JTCTPR-9	TCTPR-9	JTDIC-9	HH-9	MFF-9
32.125	23.590	15.366	15.574	7.501	6.348	3.332	2.597	3.071	**2.594**	12.511	6.840	2.604

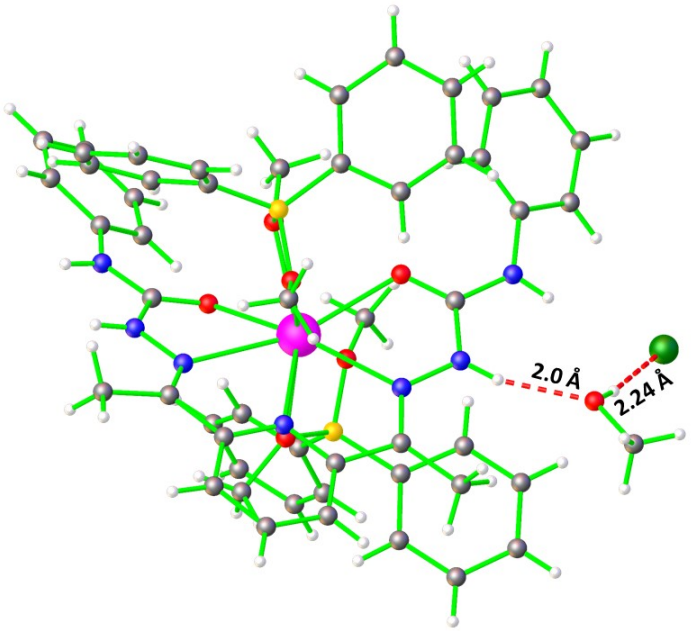

Fig S1. Intermolecular H-bonding of free MeOH molecule with the ligand and chloride ion for **1**.

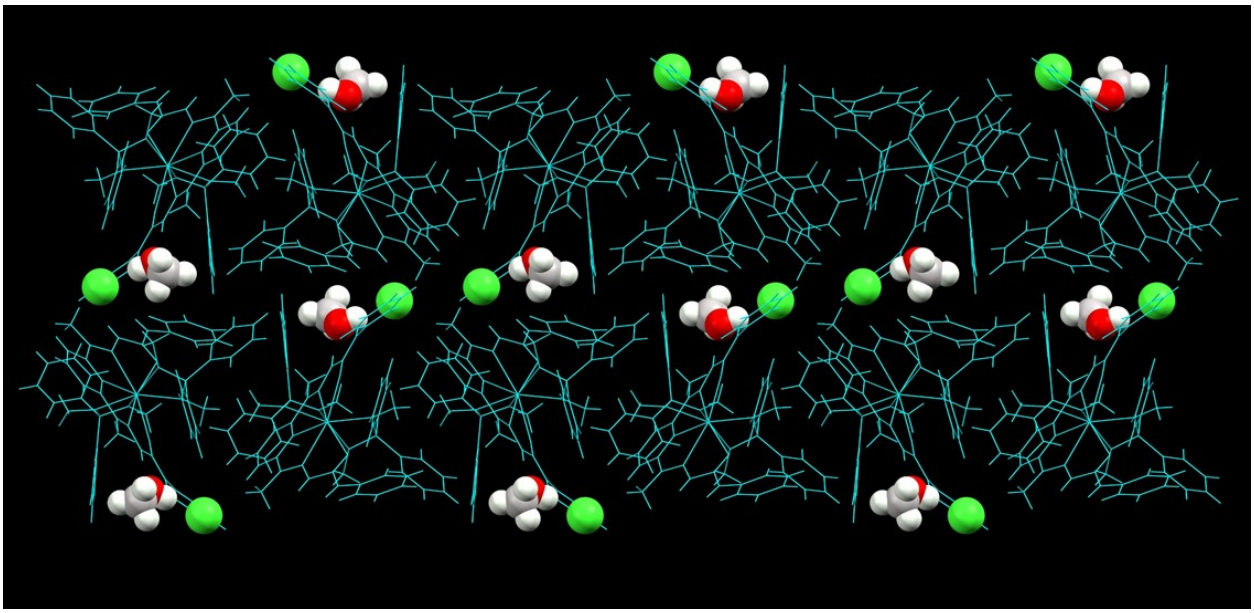

Fig S2. 2D supramolecular arrangement for complex **1**.

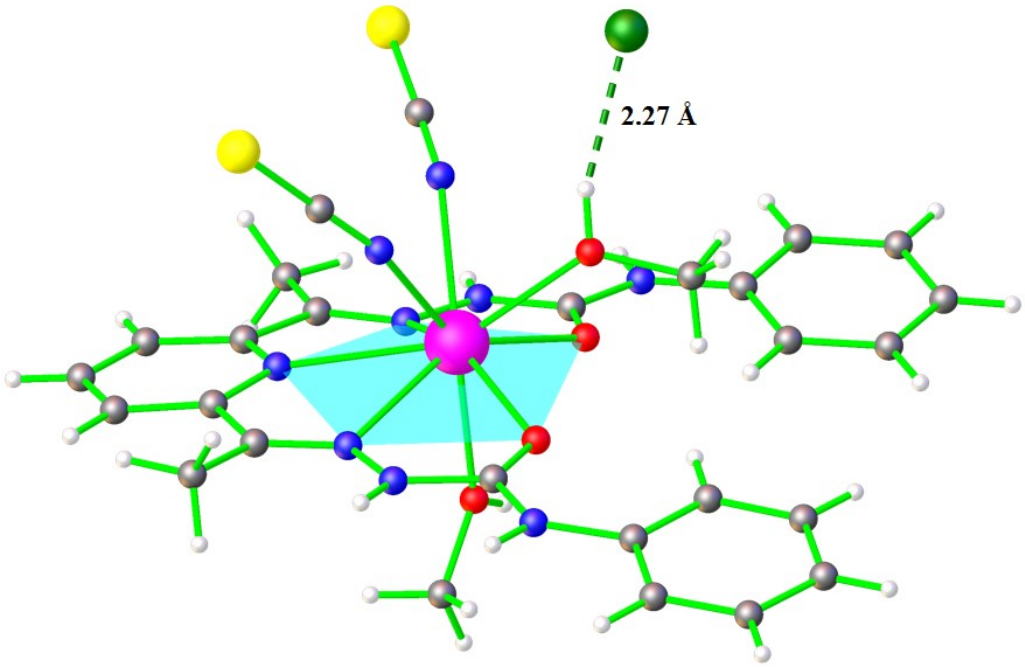

Fig S3. Intermolecular H-bonding between MeOH and chloride ion for complex **2**.

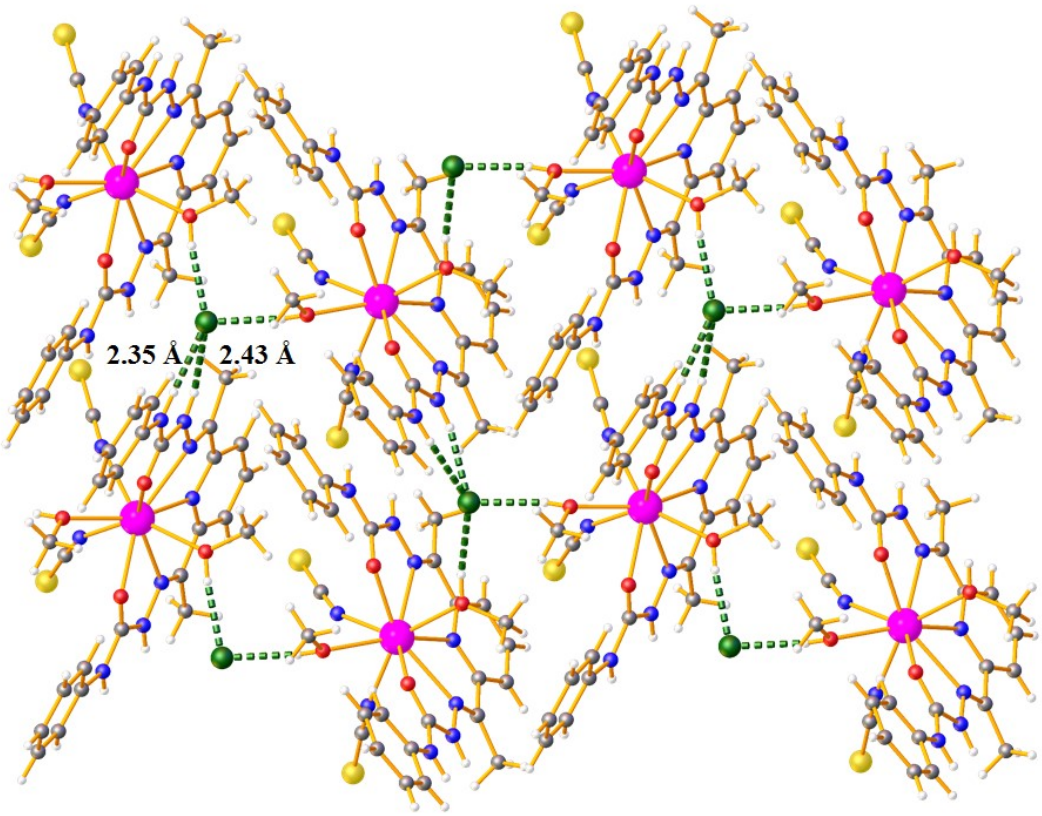

Fig S4. H-bonding between free –NH and Cl^- ion with 2D supramolecular arrangement for complex **2**.

Table S3. Bond lengths for complex **1**.

Atom	Atom	Length/Å		Atom	Atom	Length/Å
Dy1	O2	2.4646(17)		C40	C45	1.402(4)
Dy1	O5	2.351(2)		C40	B2	1.624(5)
Dy1	O1	2.4086(18)		C40	C41	1.381(5)
Dy1	O3	2.3343(19)		C19	C20	1.380(4)
Dy1	O6	2.3489(19)		C6	C1	1.367(5)
Dy1	O4	2.365(2)		C6	C5	1.390(4)
Dy1	N5	2.509(2)		C33	C34	1.392(4)
Dy1	N4	2.554(2)		C46	B2	1.624(4)
Dy1	N3	2.541(2)		C46	C47	1.390(4)
Dy1	B1	3.068(3)		C46	C51	1.384(5)
O2	C17	1.239(3)		C1	C2	1.386(5)
O5	B2	1.507(4)		C23	C22	1.389(4)
O5	C38	1.418(4)		C45	C44	1.373(5)
O1	C7	1.230(3)		C10	C11	1.389(4)
O3	C24	1.409(4)		C10	C8	1.479(4)
O3	B1	1.504(4)		C11	C12	1.380(4)
O6	C39	1.420(3)		C13	C12	1.380(4)

O6	B2	1.509(4)		C26	C27	1.402(4)
O4	B1	1.515(4)		C26	C31	1.390(4)
O4	C25	1.415(4)		C26	B1	1.631(4)
N7	C18	1.413(4)		C8	C9	1.497(4)
N7	C17	1.340(3)		C2	C3	1.372(5)
N5	N6	1.359(3)		C41	C42	1.410(5)
N5	C15	1.287(4)		C27	C28	1.388(4)
N6	C17	1.369(4)		C34	C35	1.373(5)
N2	N3	1.367(3)		C20	C21	1.382(5)
N2	C7	1.368(4)		C31	C30	1.392(5)
N4	C14	1.348(3)		C47	C48	1.383(5)
N4	C10	1.354(3)		C37	C36	1.381(5)
N3	C8	1.282(4)		C50	C51	1.389(5)
O7	C52	1.393(4)		C50	C49	1.362(6)
N1	C6	1.423(4)		C28	C29	1.374(5)
N1	C7	1.351(4)		C22	C21	1.377(4)
C15	C14	1.476(4)		C30	C29	1.365(5)
C15	C16	1.494(4)		C49	C48	1.355(6)
C18	C19	1.388(4)		C44	C43	1.357(6)
C18	C23	1.377(4)		C35	C36	1.369(5)
C14	C13	1.382(4)		C5	C4	1.377(5)
C32	C33	1.384(4)		C3	C4	1.377(5)
C32	B1	1.622(4)		C43	C42	1.378(6)
C32	C37	1.394(4)				

Table S4. Bond angles for complex **1**.

Atom	**Atom**	**Atom**	**Angle/°**		**Atom**	**Atom**	**Atom**	**Angle/°**
O2	Dy1	N5	63.32(7)		O2	C17	N6	121.1(2)
O2	Dy1	N4	121.67(7)		N7	C17	N6	112.7(2)
O2	Dy1	N3	163.27(7)		N4	C14	C15	115.5(3)
O2	Dy1	B1	82.69(8)		N4	C14	C13	122.3(3)
O5	Dy1	O2	69.51(7)		C13	C14	C15	122.2(3)
O5	Dy1	O1	72.37(6)		C33	C32	B1	123.8(3)
O5	Dy1	O4	150.78(6)		C33	C32	C37	115.8(3)
O5	Dy1	N5	89.82(7)		C37	C32	B1	120.5(3)
O5	Dy1	N4	127.04(7)		C45	C40	B2	121.2(3)
O5	Dy1	N3	123.14(7)		C41	C40	C45	115.7(3)
O5	Dy1	B1	137.30(8)		C41	C40	B2	123.1(3)
O1	Dy1	O2	117.08(6)		C20	C19	C18	119.9(3)
O1	Dy1	N5	159.46(7)		C1	C6	N1	122.3(3)
O1	Dy1	N4	121.21(7)		C1	C6	C5	120.2(3)
O1	Dy1	N3	62.81(7)		C5	C6	N1	117.5(3)
O1	Dy1	B1	93.92(8)		C32	C33	C34	122.7(3)

O3	Dy1	O2	78.93(6)		C47	C46	B2	122.8(3)
O3	Dy1	O5	111.80(7)		C51	C46	B2	121.6(3)
O3	Dy1	O1	71.29(7)		C51	C46	C47	115.2(3)
O3	Dy1	O6	154.88(7)		C6	C1	C2	119.2(3)
O3	Dy1	O4	56.77(7)		O1	C7	N2	120.9(3)
O3	Dy1	N5	126.47(7)		O1	C7	N1	123.8(3)
O3	Dy1	N4	121.09(7)		N1	C7	N2	115.3(2)
O3	Dy1	N3	85.65(7)		C18	C23	C22	118.8(3)
O3	Dy1	B1	28.40(8)		C44	C45	C40	123.4(4)
O6	Dy1	O2	112.48(6)		N4	C10	C11	121.6(3)
O6	Dy1	O5	56.80(6)		N4	C10	C8	116.0(3)
O6	Dy1	O1	83.64(7)		C11	C10	C8	122.3(2)
O6	Dy1	O4	144.32(7)		C12	C11	C10	118.9(3)
O6	Dy1	N5	77.93(7)		C12	C13	C14	118.6(3)
O6	Dy1	N4	73.23(7)		O5	B2	O6	95.7(2)
O6	Dy1	N3	84.24(7)		O5	B2	C40	113.6(3)
O6	Dy1	B1	164.05(8)		O5	B2	C46	110.6(2)
O4	Dy1	O2	81.52(7)		O6	B2	C40	110.4(2)
O4	Dy1	O1	120.30(7)		O6	B2	C46	111.6(3)
O4	Dy1	N5	80.22(7)		C46	B2	C40	113.7(3)
O4	Dy1	N4	71.63(7)		C27	C26	B1	122.6(3)
O4	Dy1	N3	84.64(7)		C31	C26	C27	115.3(3)
O4	Dy1	B1	28.84(8)		C31	C26	B1	122.1(3)
N5	Dy1	N4	61.71(7)		N3	C8	C10	114.7(2)
N5	Dy1	N3	123.35(8)		N3	C8	C9	124.9(3)
N5	Dy1	B1	106.27(8)		C10	C8	C9	120.4(3)
N4	Dy1	B1	94.94(9)		C3	C2	C1	121.1(4)
N3	Dy1	N4	61.68(7)		C40	C41	C42	121.5(4)
N3	Dy1	B1	80.66(8)		C28	C27	C26	122.0(3)
C17	O2	Dy1	117.98(17)		C35	C34	C33	119.7(3)
B2	O5	Dy1	103.61(17)		C19	C20	C21	120.1(3)
C38	O5	Dy1	131.07(19)		C26	C31	C30	122.6(3)
C38	O5	B2	120.6(3)		C48	C47	C46	123.0(4)
C7	O1	Dy1	122.1(2)		O3	B1	Dy1	47.57(12)
C24	O3	Dy1	131.7(2)		O3	B1	O4	95.5(2)
C24	O3	B1	121.5(2)		O3	B1	C32	112.4(2)
B1	O3	Dy1	104.03(16)		O3	B1	C26	109.9(2)
C39	O6	Dy1	137.28(19)		O4	B1	Dy1	48.85(13)
C39	O6	B2	118.6(2)		O4	B1	C32	112.9(2)
B2	O6	Dy1	103.65(16)		O4	B1	C26	110.8(2)
B1	O4	Dy1	102.30(17)		C32	B1	Dy1	132.75(19)
C25	O4	Dy1	135.36(19)		C32	B1	C26	113.9(3)
C25	O4	B1	121.7(3)		C26	B1	Dy1	113.33(19)
C17	N7	C18	129.5(2)		C13	C12	C11	119.8(3)
N6	N5	Dy1	114.75(17)		C36	C37	C32	122.0(3)

C15	N5	Dy1	124.16(17)		C49	C50	C51	120.1(4)
C15	N5	N6	118.8(2)		C29	C28	C27	120.7(3)
N5	N6	C17	116.0(2)		C21	C22	C23	121.2(3)
N3	N2	C7	114.9(2)		C46	C51	C50	122.1(3)
C14	N4	Dy1	120.82(17)		C29	C30	C31	120.5(4)
C14	N4	C10	118.7(2)		C48	C49	C50	120.0(3)
C10	N4	Dy1	120.37(18)		C30	C29	C28	118.8(3)
N2	N3	Dy1	115.37(17)		C43	C44	C45	119.4(4)
C8	N3	Dy1	123.40(18)		C36	C35	C34	119.1(3)
C8	N3	N2	119.9(2)		C22	C21	C20	119.4(3)
C7	N1	C6	124.8(2)		C49	C48	C47	119.5(4)
N5	C15	C14	114.0(2)		C4	C5	C6	119.8(3)
N5	C15	C16	122.8(3)		C2	C3	C4	119.4(4)
C14	C15	C16	123.1(3)		C44	C43	C42	120.3(4)
C19	C18	N7	115.7(3)		C43	C42	C41	119.5(4)
C23	C18	N7	123.7(3)		C35	C36	C37	120.7(3)
C23	C18	C19	120.5(3)		C3	C4	C5	120.3(3)
O2	C17	N7	126.2(3)					

Table S5. Bond lengths for complex **2**.

Atom	**Atom**	**Length/Å**		**Atom**	**Atom**	**Length/Å**
Dy1	O1	2.4132(18)		N4	C10	1.347(3)
Dy1	O3	2.4136(18)		N4	C14	1.349(3)
Dy1	O2	2.4014(17)		N9	C25	1.163(3)
Dy1	O4	2.3936(18)		N8	C24	1.158(3)
Dy1	N3	2.552(2)		C5	C4	1.385(4)
Dy1	N5	2.578(2)		C5	C6	1.383(4)
Dy1	N4	2.570(2)		C15	C14	1.479(4)
Dy1	N9	2.446(2)		C15	C16	1.498(4)
Dy1	N8	2.407(2)		C8	C10	1.471(4)
S2	C25	1.643(3)		C8	C9	1.494(4)
S1	C24	1.628(3)		C12	C13	1.380(4)
O1	C7	1.234(3)		C12	C11	1.374(4)
O3	C26	1.433(3)		C10	C11	1.390(4)
O2	C17	1.235(3)		C14	C13	1.390(4)
O4	C27	1.437(3)		C4	C3	1.386(4)
N1	C7	1.349(3)		C6	C1	1.390(4)
N1	C5	1.416(3)		C18	C23	1.375(4)
N2	N3	1.368(3)		C18	C19	1.382(4)
N2	C7	1.378(3)		C1	C2	1.374(4)
N6	N5	1.366(3)		C23	C22	1.387(4)
N6	C17	1.378(3)		C3	C2	1.375(4)
N3	C8	1.286(3)		C22	C21	1.374(4)

N5	C15	1.287(3)		C20	C21	1.368(5)
N7	C17	1.341(3)		C20	C19	1.381(4)
N7	C18	1.417(3)				

Table S6. Bond angles for complex **2**.

Atom	Atom	Atom	Angle/°		Atom	Atom	Atom	Angle/°
O1	Dy1	O3	72.41(6)		C15	N5	Dy1	124.06(17)
O1	Dy1	N3	63.23(6)		C15	N5	N6	118.5(2)
O1	Dy1	N5	148.06(6)		C17	N7	C18	128.8(2)
O1	Dy1	N4	123.09(6)		C10	N4	Dy1	120.46(17)
O1	Dy1	N9	143.67(7)		C10	N4	C14	118.0(2)
O3	Dy1	N3	121.98(7)		C14	N4	Dy1	121.52(16)
O3	Dy1	N5	119.44(7)		C25	N9	Dy1	160.1(2)
O3	Dy1	N4	153.21(6)		C24	N8	Dy1	156.7(2)
O3	Dy1	N9	77.01(7)		O2	C17	N6	120.5(2)
O2	Dy1	O1	98.96(6)		O2	C17	N7	127.0(2)
O2	Dy1	O3	70.68(6)		N7	C17	N6	112.5(2)
O2	Dy1	N3	148.23(6)		O1	C7	N1	125.8(2)
O2	Dy1	N5	62.74(6)		O1	C7	N2	121.3(2)
O2	Dy1	N4	122.00(6)		N1	C7	N2	112.9(2)
O2	Dy1	N9	88.81(7)		C4	C5	N1	116.7(2)
O2	Dy1	N8	141.15(7)		C6	C5	N1	122.8(2)
O4	Dy1	O1	70.62(6)		C6	C5	C4	120.4(2)
O4	Dy1	O3	123.72(6)		N5	C15	C14	113.7(2)
O4	Dy1	O2	74.92(6)		N5	C15	C16	123.6(2)
O4	Dy1	N3	74.30(6)		C14	C15	C16	122.6(2)
O4	Dy1	N5	79.03(6)		N3	C8	C10	114.7(2)
O4	Dy1	N4	83.06(6)		N3	C8	C9	125.0(3)
O4	Dy1	N9	144.97(7)		C10	C8	C9	120.3(2)
O4	Dy1	N8	138.22(7)		C11	C12	C13	119.6(2)
N3	Dy1	N5	118.06(7)		N4	C10	C8	116.6(2)
N3	Dy1	N4	61.36(7)		N4	C10	C11	122.4(2)
N4	Dy1	N5	60.67(6)		C11	C10	C8	120.9(2)
N9	Dy1	N3	121.47(7)		N4	C14	C15	116.3(2)
N9	Dy1	N5	65.95(7)		N4	C14	C13	122.3(2)
N9	Dy1	N4	79.68(7)		C13	C14	C15	121.3(2)
N8	Dy1	O1	80.97(7)		C12	C13	C14	118.8(3)
N8	Dy1	O3	72.41(7)		C5	C4	C3	119.6(3)
N8	Dy1	N3	65.62(7)		N9	C25	S2	177.5(3)
N8	Dy1	N5	130.11(7)		C5	C6	C1	118.8(3)
N8	Dy1	N4	87.79(7)		C12	C11	C10	118.8(2)
N8	Dy1	N9	71.37(8)		C23	C18	N7	124.1(2)
C7	O1	Dy1	121.82(16)		C23	C18	C19	119.9(3)

C26	O3	Dy1	127.23(16)		C19	C18	N7	116.0(3)
C17	O2	Dy1	123.80(16)		C2	C1	C6	121.1(3)
C27	O4	Dy1	132.08(16)		N8	C24	S1	177.2(3)
C7	N1	C5	129.1(2)		C18	C23	C22	119.3(3)
N3	N2	C7	115.0(2)		C2	C3	C4	120.4(3)
N5	N6	C17	115.3(2)		C21	C22	C23	121.1(3)
N2	N3	Dy1	115.30(14)		C21	C20	C19	120.8(3)
C8	N3	Dy1	124.07(18)		C20	C21	C22	119.1(3)
C8	N3	N2	118.9(2)		C20	C19	C18	119.9(3)
N6	N5	Dy1	114.45(15)		C1	C2	C3	119.5(3)

Table S7. Field and relaxation time at 2 K temperature for complex **1**.

Complex 1	
Field (T)	τ (s)
0.02	4.42934E-4
0.04	9.4954E-4
0.08	0.002
0.1	0.0022
0.12	**0.00227**
0.15	0.00195
0.18	0.00153
0.2	0.00127
0.22	0.00105
0.25	7.98161E-4
0.28	6.10354E-4
0.3	4.65323E-4
0.32	3.81452E-4
0.35	2.77419E-4
0.38	2.1371E-4
0.4	1.83065E-4
0.45	1.25806E-4
0.5	9.19355E-5
0.55	8.30645E-5
0.6	7.90323E-5
0.65	6.69355E-5
0.7	6.20968E-5
0.75	5.64516E-5
0.8	4.91935E-5
0.85	5.80645E-5
0.9	4.67742E-5
0.95	4.35484E-5
1	4.83871E-5

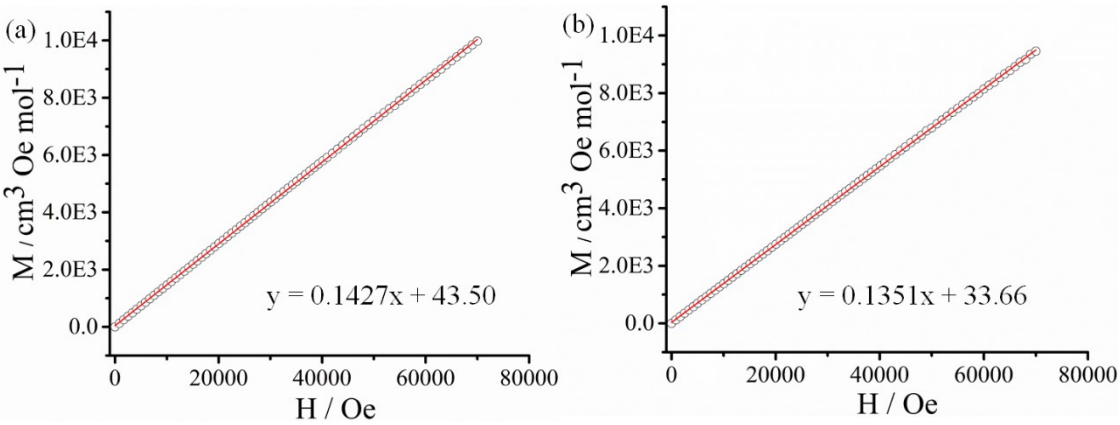

Figure S5. The magnetization plot at 100 K in range of magnetic field 0-7 T for complexes **1** (a) and **2** (b). The linear fit of the magnetization data at 100 K indicates the absence of ferromagnetic impurity

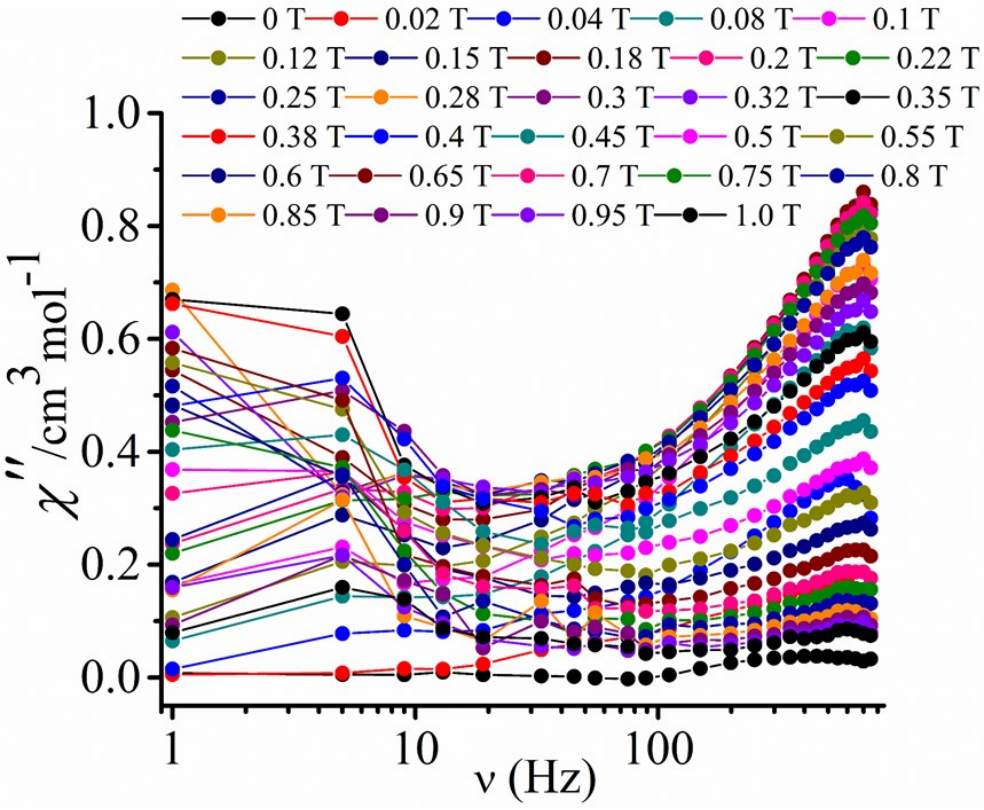

Fig S6. The frequency dependence of the out-of-phase susceptibility under different external field at 2 K temperature for complex **2**.

Table S8. The parameters obtained from the fitting of frequency dependent data using generalized Debye model for complex **1**.

T (K)	χ_S	χ_T	τ (s)	α
2	1.10365	18.68543	0.00226	0.13251
2.3	1.07589	16.71428	0.00177	0.14736
2.6	1.086	15.14969	0.00114	0.15586
2.9	1.12425	13.83332	7.92464E-4	0.16204
3.2	1.18169	12.7259	5.73405E-4	0.1681
3.5	1.22299	11.79867	4.14656E-4	0.17779
3.8	1.28295	10.9982	3.02808E-4	0.18834
4.1	1.40364	10.27728	2.29869E-4	0.19438
4.4	1.49458	9.66193	1.76504E-4	0.20348
4.7	1.59888	9.10693	1.37944E-4	0.20894
5	1.5216	8.61689	1.03894E-4	0.22125
5.3	1.5298	8.15988	8.10086E-5	0.2224
5.6	1.07203	7.76192	6.09885E-5	0.23752
5.9	1.3124	7.38379	4.78372E-5	0.22483

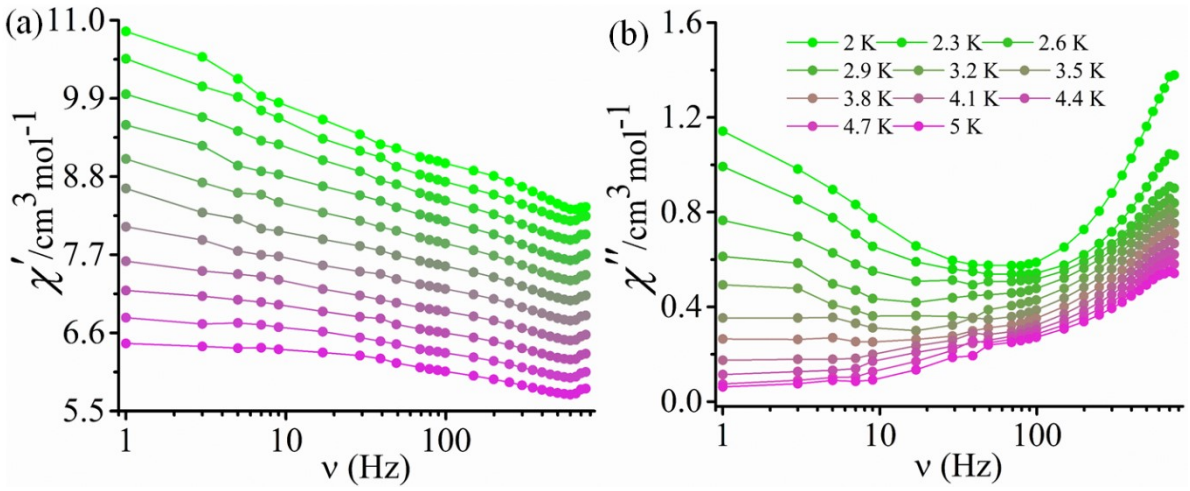

Fig S7. The frequency dependence of in-phase (a) and out-of-phase (b) susceptibility under 0.18 T external field for complex **2**.

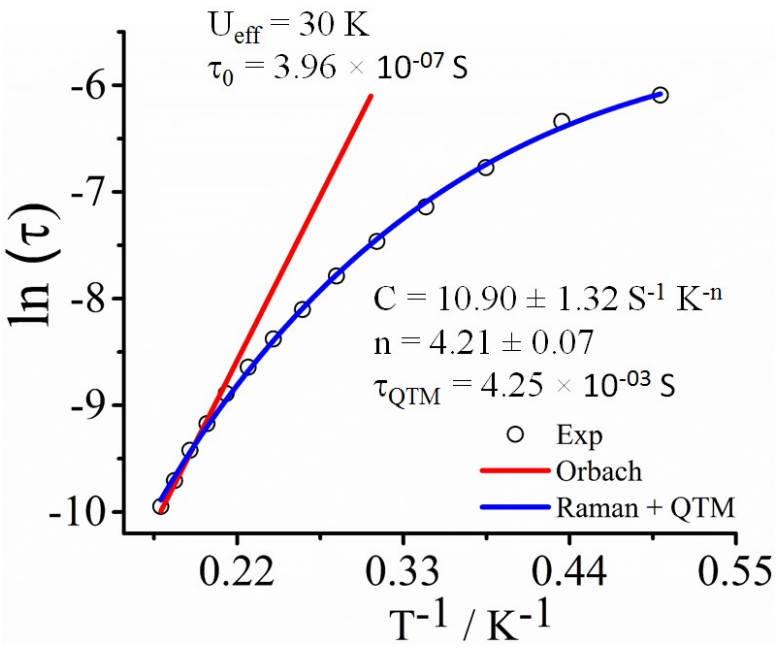

Fig S8. ln(τ) *vs*. 1/T plot for complex **1**.

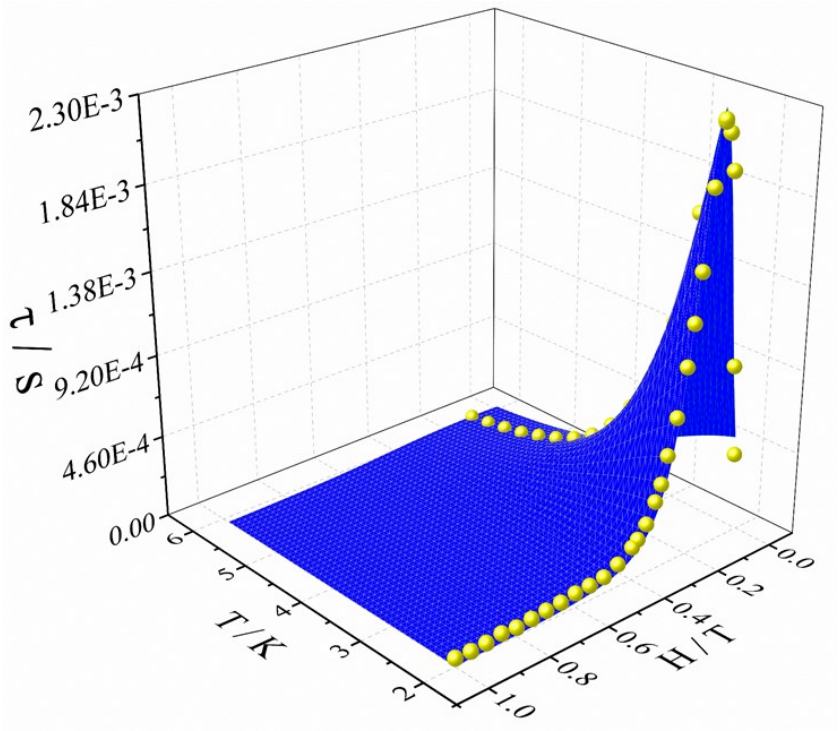

Fig S9. 3D isosurface plot for temperature and field dependence of relaxation time for **1**.

Table S9. SINGLE_ANISO computed crystal-field parameters for MOF **1** and **2**.

k	Q	Complex **1**	Complex **2**
	-2	-0.4101E+00	-0.4912E+00
	-1	-0.8202E+00	-0.9825E+00
2	0	-0.2815E+01	-0.9916E+00
	1	0.2002E+01	-0.2851E+01
	2	0.1300E+01	0.1611E+01
	-4	-0.1005E-01	-0.9335E-02
	-3	-0.2844E-01	-0.2640E-01
	-2	-0.7603E-02	-0.7056E-02
	-1	-0.1075E-01	-0.9979E-02
4	0	-0.1668E-02	-0.2189E-02
	1	-0.6935E-03	0.4176E-02
	2	0.1471E-01	0.1493E-01
	3	0.2603E-02	-0.3536E-01
	4	-0.7315E-02	-0.2845E-02
	-6	-0.2577E-03	-0.5191E-04
	-5	-0.8928E-03	-0.1798E-03
	-4	-0.1903E-03	-0.3833E-04
	-3	-0.3475E-03	-0.6999E-04
	-2	-0.1737E-03	-0.3499E-04
	-1	-0.2198E-03	-0.4427E-04
6	0	-0.3770E-04	0.4003E-06
	1	-0.2312E-03	0.8753E-04
	2	-0.1523E-03	-0.4016E-04
	3	-0.1845E-04	-0.2218E-03
	4	-0.1004E-04	-0.1043E-03
	5	-0.3232E-03	-0.2009E-03
	6	0.6209E-03	0.4572E-03

Table S10. Ab initio calculated low-lying spin-free energy states for the investigated complexes **1**-**2**.

Complex **1**	Complex **2**
0.000	0.000
14.809	45.239
124.890	80.002
224.397	179.591
353.750	202.473
418.905	280.320
490.147	317.621

Table S11. Ab initio calculated low-lying spin-orbit energy states for the investigated complexes **1**-**2**.

Complex **1**	Complex **2**
0.000	0.000
0.000	0.000
189.194	43.555
189.194	43.555
239.840	84.180
239.840	84.180
315.349	126.628
315.349	126.628
417.879	203.911
417.879	203.911
491.449	221.356
491.449	221.356
591.904	243.127
591.904	243.127
643.729	415.840
643.729	415.840

Table S12. Single_aniso computed energy of the KDs, g and wavefuctions composition for Complex **1**.

Kramers doublets (KDs)	Energy (cm^{-1})	g_x	g_y	g_z	Wave function composition
1	0.000	0.042	0.059	19.85	98.11%|±15/2>+1.49%|±11/2> +0.17%|±9/2>
2	189.194	1.388	3.546	14.95	60.17%|±13/2>+20.44%|±9/2> +7.98%|±11/2>+5.91%|±7/2>+ 2.92%|±5/2>+1.15%|±1/2>+1.2 2%|±3/2>
3	239.840	1.491	5.164	10.66	44.93%|±11/2>+17.55%|±9/2> +17.65%|±7/2>+10.03%|±13/2 >+4.0%|±5/2>+2.16%|±3/2>+2. 52%|±1/2>+1.13%|±15/2>
4	315.349	2.973	6.203	10.46	22.63%|±9/2>+23.78%|±7/2>+ 13.18%|±11/2>+14.62%|±5/2> +20.05%|±13/2>+4.91%|±3/2> 0.55%|±1/2>0.24%|±15/2>
5	417.879	1.222	3.108	13.41	0.074%|±15/2>+4.21%|±13/2>

					+11.76%|±11/2>+12.18%|±9/2>+22.49%|±7/2>+35.52%|±5/2>+8.28%|±3/2>+5.43%|±1/2>
6	491.449	0.420	2.668	13.50	0.20%|±15/2>+1.17%|±13/2>+8.88%|±11/2>+6.40%|±9/2>+6.19%|±7/2>+9.45%|±5/2>+42.54%|±3/2>+24.53%|±1/2>
7	591.904	1.389	1.850	12.13	0.024%|±15/2>+2.95%|±13/2>+4.41%|±11/2>+9.23%|±9/2>+13.23%|±7/2>+26.96%|±5/2>+16.33%|±3/2>+26.83%|±1/2>
8	643.729	0.685	4.401	14.97	0.020%|±15/2>+0.776%|±13/2>+7.33%|±11/2>+11.36%|±9/2>+10.66%|±7/2>+6.46%|±5/2>+24.41%|±3/2>+38.95%|±1/2>

Table S13. Single_aniso computed energy of the KDs, g and wavefuctions composition for Complex **2**.

Kramers doublets (KDs)	Energy (cm^{-1})	g_x	g_y	g_z	Wave function composition
1	0.000	0.907	3.350	17.13	74.56%|±15/2>+0.77%|±13/2>+11.40%|±11/2>+2.50%|±9/2>+6.29%|±7/2>+1.17%|±5/2>+2.28%|±3/2>+0.99%|±1/2>
2	43.555	3.522	4.152	9.989	8.74%|±15/2>+41.41%|±13/2>+1.15%|±11/2>+21.57%|±9/2>+4.87%|±7/2>+13.01%|±5/2>+3.94%|±3/2>+5.27%|±1/2>
3	84.180	0.370	3.289	10.94	9.03%|±15/2>+24.03%|±13/2>+29.47%|±11/2>+5.71%|±9/2>+13.13%|±7/2>+2.32%|±5/2>+7.43%|±3/2>+8.82%|±1/2>
4	126.628	1.703	2.271	13.05	2.95%|±15/2>+12.43%|±13/2>+18.21%|±11/2>+18.79%|±9/2>+15.00%|±7/2>+15.68%|±5/2>+13.08%|±3/2>+3.83%|±1/2>
5	203.911	0.236	1.143	17.22	1.53%|±15/2>+5.29%|±13/2>+7.08%|±11/2>+4.38%|±9/2>+11.42%|±7/2>+13.37%|±5/2>+20.06%|±3/2>+36.83%|±1/2>
6	221.356	0.477	1.899	15.65	1.49%|±15/2>+3.79%|±13/2>+7.98%|±11/2>+19.69%|±9/2>+20.88%|±7/2>+12.47%|±5/2>+17.15%|±3/2>+16.51%|±1/2>
7	243.127	0.220	1.936	16.89	0.33%|±15/2>+5.10%|±13/2>+

					7.69%|±11/2>+3.56%|±9/2>+7.08%|±7/2>+28.26%|±5/2>+27.10%|±3/2>+20.83%|±1/2>
8	415.840	0.004	0.008	19.56	1.32%|±15/2>+7.14%|±13/2>+16.97%|±11/2>+23.76%|±9/2>+21.28%|±7/2>+13.68%|±5/2>+8.93%|±3/2>+6.89%|±1/2>

Table S14. Single_aniso computed energy of the KDs, g, angle between the anisotropic axes and wave functions composition for model complex **1a**.

Kramers doublets (KDs)	Energy (cm^{-1})	g_x	g_y	g_z	Angle (°)	Wave function composition
1	0.000	0.000	0.000	19.96		99.28%|±15/2>
2	411.301	0.000	0.000	16.85	1.16	95.50%|±13/2>+4.29%|±9/2>
3	712.464	0.010	0.012	13.90	2.95	89.57%|±11/2>+9.15%|±7/2>
4	991.199	0.166	0.167	11.26	6.21	82.41%|±9/2>+12.23%|±5/2>+4.16%|±13/2>
5	1260.985	1.132	1.339	8.742	10.01	74.78%|±7/2>+13.26%|±3/2>+0.81%|±5/2>+8.56%|±11/2>
6	1491.424	3.706	5.065	6.427	129.21	64.68%|±5/2>+4.58%|±3/2>+16.52%|±1/2>+10.50%|±9/2>+3.27%|±7/2>
7	1674.750	2.495	3.983	12.725	88.70	57.29%|±3/2>+17.16%|±1/2>+13.19%|±5/2>+10.80%|±7/2>+1.04%|±9/2>
8	1825.802	0.501	1.508	18.752	87.66	64.63%|±1/2>+24.22%|±3/2>+8.93%|±5/2>+1.70%|±7/2>

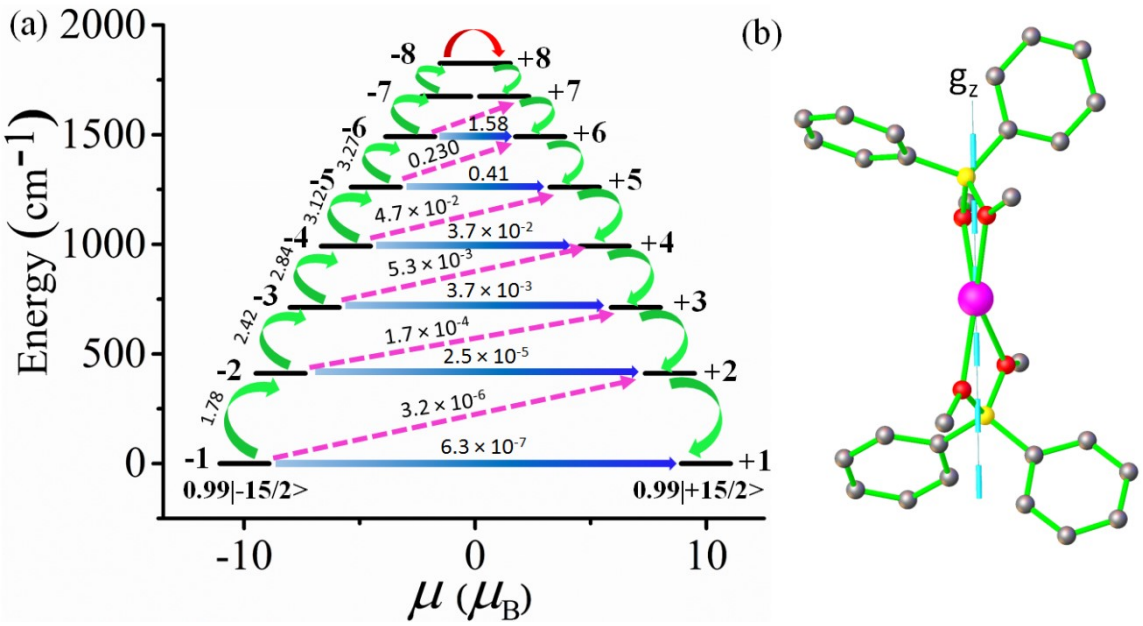

Fig S10. Single_aniso calculated blocking barrier (a) and orientation of the main magnetic axis for model complex **1a**.