Supporting Information

Coordination microenvironment perturbed single-ion magnet behavior in *β*-diketone Dy(III) complex

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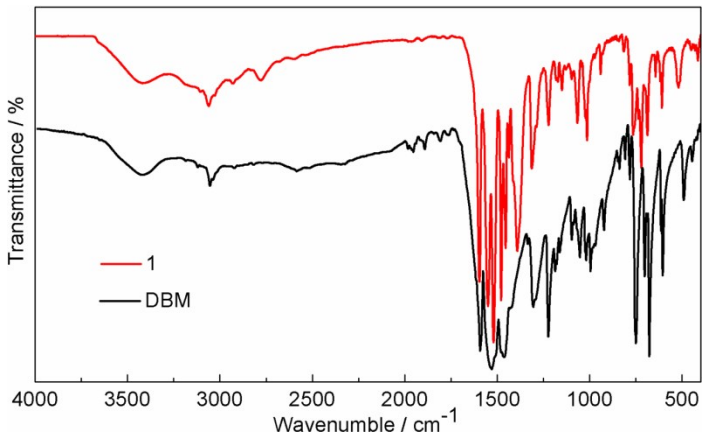

Fig. S1 IR curves of **1.**

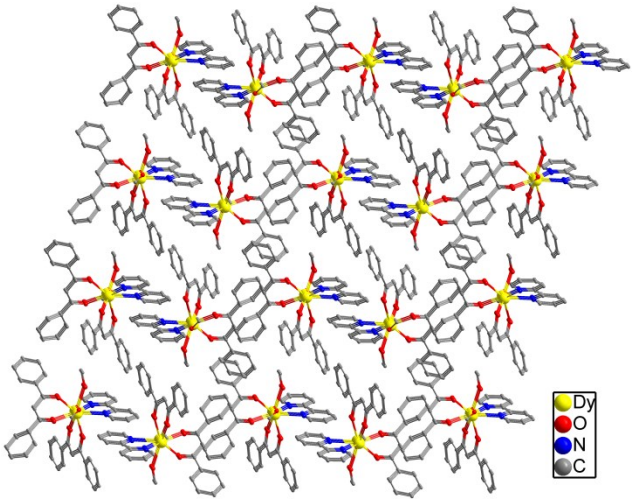

Fig. S2 Molecular stacking charts of complex **1.** H atoms and free chloride ion are omitted for clarity.

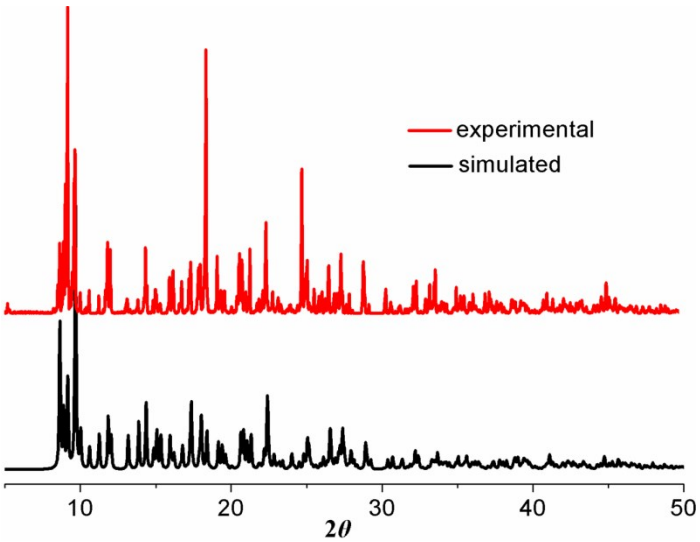

Fig. S3 PXRD curves of **1**.

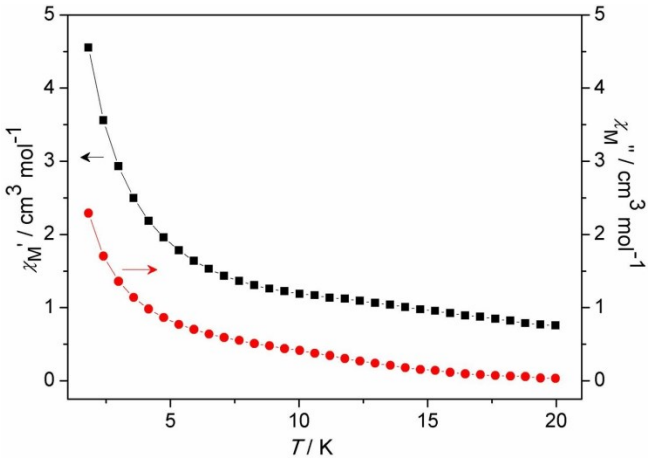

Fig. S4 Temperature dependence of χ' and χ'' susceptibilities for **1** without static field.

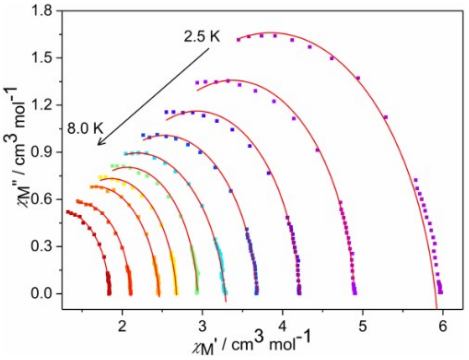

Fig. S5 Cole-Cole plots at H_{dc} = 0 Oe for **1**. The solid lines represent the best fit to the measured results.

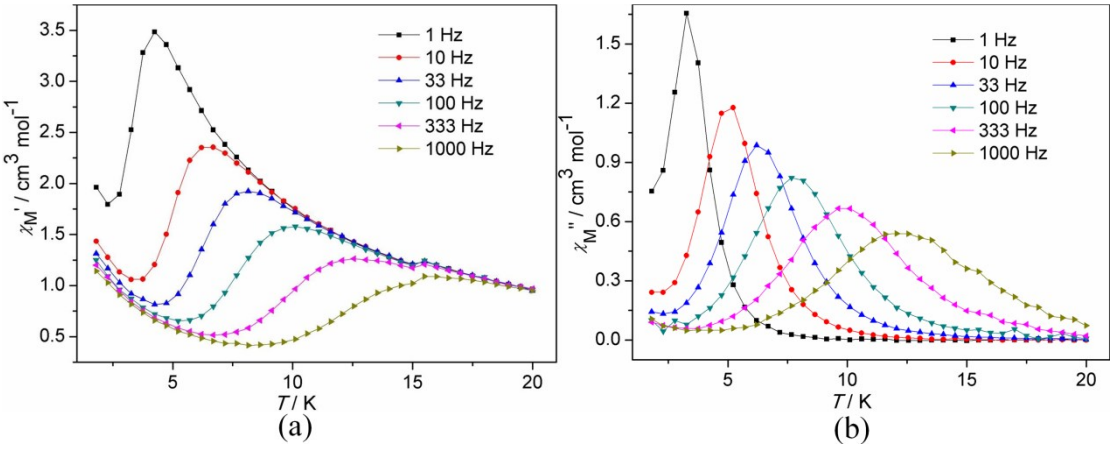

Fig. S6 Temperature dependence of χ' and χ'' susceptibilities for **1** at applied dc fields of 1200 Oe.

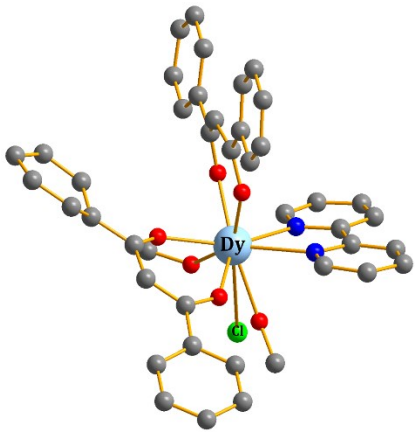

Fig. S7 Calculated model structures of Dy^{III} fragment of **1**. H atoms are omitted.

Table S1. Selected bond lengths (Å) and bond angles (°) for **1**.

Complex **1**			
Dy(1)-O(1)	2.295(3)	Dy(1)-O(2)	2.293(2)
Dy(1)-O(3)	2.295(2)	Dy(1)-O(4)	2.275(3)
Dy(1)-O(5)	2.446(3)	Dy(1)-O(6)	2.402(3)
Dy(1)-N(1)	2.545(3)	Dy(1)-N(2)	2.569(3)
O(1)-Dy(1)-O(2)	73.57(8)	O(1)-Dy(1)-O(3)	146.70(9)
O(1)-Dy(1)-O(4)	79.15(10)	O(1)-Dy(1)-O(5)	136.71(9)
O(1)-Dy(1)-O(6)	77.43(9)	O(1)-Dy(1)-N(1)	112.08(9)
O(1)-Dy(1)-N(2)	72.00(9)	O(2)-Dy(1)-O(3)	83.72(8)
O(2)-Dy(1)-O(4)	81.99(10)	O(2)-Dy(1)-O(5)	142.98(8)
O(2)-Dy(1)-O(6)	145.70(9)	O(5)-Dy(1)-N(2)	74.59(9)
O(3)-Dy(1)-N(2)	139.02(9)	O(4)-Dy(1)-O(5)	118.91(10)
O(4)-Dy(1)-O(6)	74.86(11)	O(4)-Dy(1)-N(1)	147.39(11)
O(4)-Dy(1)-N(2)	146.21(10)	O(5)-Dy(1)-O(6)	71.28(8)
O(5)-Dy(1)-N(1)	74.82(8)	O(5)-Dy(1)-N(2)	74.59(9)
O(6)-Dy(1)-N(1)	136.45(10)	O(6)-Dy(1)-N(2)	81.91(10)
N(1)-Dy(1)-N(2)	63.12(10)	Dy(1)-O(1)-C(17)	136.1(2)
C(43)-O(6)-H(6A)	114.00	Dy(1)-N(1)-C(1)	119.8(2)
Dy(1)-N(1)-C(5)	122.5(3)	C(1)-N(1)-C(5)	117.7(3)

Table S2. Dy (III) ion geometry analysis of **1** by SHAPE 2.1 software.

Dy (III) ion geometry analysis of **1**

```
HBPY-8          3 D6h   Hexagonal bipyramid
CU-8            4 Oh    Cube
SAPR-8          5 D4d   Square antiprism
TDD-8           6 D2d   Triangular dodecahedron
JGBF-8          7 D2d   Johnson gyrobifastigium J26
JETBPY-8        8 D3h   Johnson elongated triangular bipyramid J14
JBTPR-8         9 C2v   Biaugmented trigonal prism J50
BTPR-8         10 C2v   Biaugmented trigonal prism
JSD-8          11 D2d   Snub diphenoid J84
TT-8           12 Td    Triakis tetrahedron
ETBPY-8        13 D3h   Elongated trigonal bipyramid

Structure [ML8 ]    HBPY-8     CU-8     SAPR-8     TDD-8     JGBF-8    JETBPY-8    JBTPR-8    BTPR-8     JSD-8     TT-8     ETBPY-8
 ABOXIY         ,   16.954,    10.092,    0.535,     2.053,    16.149,    28.292,     2.801,     2.132,     4.907,   10.880,    24.113
```

Configuration	ABOXIY, **1**
Hexagonal bipyramid (D_{6h})	16.954
Cube (O_h)	10.092
Square antiprism (D_{4d})	0.535
Triangular dodecahedron (D_{2d})	2.053
Johnson gyrobifastigium J26 (D_{2d})	16.149
Johnson elongated triangular bipyramid J14 (D_{3h})	28.292
Biaugmented trigonal prism J50 (C_{2v})	2.801
Biaugmented trigonal prism (C_{2v})	2.132
Snub siphenoid J84 (D_{2d})	4.907
Triakis tetrahedron(T_d)	10.880
Elongated trigonal bipyramid(D_{3h})	24.113

Table S3. Relevant structural parameters in square anti-prism geometry, α angle and ϕ angle.

α angle (°)	**1**	ϕ angle (°)	**1**
O1	60.536	O1-Dy-O4	42.886
O2	58.114	O1-Dy-O6	45.198
N1	51.539	O2-Dy-O3	47.434
N2	47.675	O2-Dy-O4	44.612
O3	58.114	O3-Dy-N1	46.072
O4	61.815	O5-Dy-N1	41.992
O5	57.085	O5-Dy-N2	43.992
O6	54.675	O6-Dy-N2	48.061
Deviation	56.194	Deviation	45.031
Sum of deviation to ideal 54.74°	1.454	Sum of deviation to ideal 45°	0.031

Table S4. Crystal field parameters and g-factors for **1** fitted from *M vs. H*.

	1
B_2^0	362.8
B_4^0	685.7
B_6^0	337.1

Table S5. Energy levels and eigenstates for **1**.

Energy / cm^{-1}	Eigenstate
0	±1/2
85	±15/2
189	±13/2
374	±3/2
943	±5/2
966	±11/2
1407	±7/2
1460	±9/2

Table S6. Relaxation fitting parameters from least-squares fitting of $\chi(f)$ data under zero dc field of **1.**

T(K)	χ_T	χ_S	α
2.5	5.906	1.796	0.135
3	4.897	1.859	0.071
3.5	4.211	1.649	0.062
4	3.684	1.364	0.089
4.5	3.284	1.099	0.126
5	2.940	1.192	0.053
5.5	2.676	1.051	0.066
6	2.459	0.881	0.093
7	2.108	0.822	0.067
8	1.842	0.691	0.063

Table S7. Relaxation fitting parameters from least-squares fitting of $\chi(f)$ data under 1200 Oe dc field of **1.**

T(K)	χ_T	χ_S	α
3.5	5.871	0.781	0.212

3.8	4.676	0.775	0.118
4	4.379	0.753	0.096
4.2	4.197	0.778	0.077
4.5	3.922	0.669	0.082
5	3.524	0.607	0.073
5.5	3.221	0.533	0.081
6	2.958	0.448	0.074
6.5	2.042	0.346	0.043
7	2.750	0.430	0.054
7.5	2.579	0.455	0.065
8	2.442	0.409	0.046
8.5	2.279	0.389	0.045
9	2.151	0.375	0.039
9.5	1.938	0.351	0.029
10	1.849	0.331	0.035
10.5	1.769	0.277	0.036
11	1.687	0.305	0.036