Designing a mononuclear Dy^{III} Single-Molecule Magnet (SMM) by Using a N,O,N,O-based multichelating Schiff base ligand and a *β*-diketonate ligand

Sheng Zhang,[*1,3] Wenjiao Mo,[1] Bing Yin,[*2] Xingqiang Lü,[3] Jiangwei Zhang,[*4]

1. *College of Chemistry and Chemical Engineering, Baoji University of Arts and Sciences, Baoji 721013, China.*
2. *Key Laboratory of Synthetic and Natural Functional Molecule Chemistry of Ministry of Education, College of Chemistry and Materials Science, Northwest University, Xi'an, Shaanxi 710069, China.*
3. *School of Chemical Engineering, Shaanxi Key Laboratory of Degradable Medical Material, Northwest University, Xi'an 710069, Shaanxi, China.*
4. *Dalian Institute of Chemical Physics, Chinese Academy of Sciences (CAS), Dalian 116023, P.R. China.*

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

Dy(1)-N(1)	2.568(10)
Dy(1)-N(2)	2.633(16)
Dy(1)-N(3)	2.674(13)
Dy(1)-N(4)	2.588(10)
Dy(1)-O(1)	2.369(12)
Dy(1)-O(2)	2.283(14)
Dy(1)-O(6)	2.241(11)
Dy(1)-O(7)	2.259(11)
N(1)-Dy(1)-N(2)	66.6(5)
N(1)-Dy(1)-N(3)	124.7(5)
N(1)-Dy(1)-N(4)	148.5(5)
N(2)-Dy(1)-N(3)	68.8(6)
N(4)-Dy(1)-N(2)	94.3(5)
N(4)-Dy(1)-N(3)	63.6(4)
O(1)-Dy(1)-N(1)	80.6(5)
O(1)-Dy(1)-N(2)	141.2(5)
O(1)-Dy(1)-N(3)	150.0(6)
O(1)-Dy(1)-N(4)	104.3(4)
O(2)-Dy(1)-N(1)	137.7(5)
O(2)-Dy(1)-N(2)	148.5(4)
O(2)-Dy(1)-N(3)	79.7(5)
O(2)-Dy(1)-N(4)	70.7(5)
O(2)-Dy(1)-O(1)	70.3(4)
O(6)-Dy(1)-N(1)	76.1(4)

O(6)-Dy(1)-N(2)	76.0(5)
O(6)-Dy(1)-N(3)	121.9(5)
O(6)-Dy(1)-N(4)	75.0(5)
O(6)-Dy(1)-O(1)	76.4(4)
O(6)-Dy(1)-O(2)	123.4(4)
O(6)-Dy(1)-O(7)	152.4(4)
O(7)-Dy(1)-N(1)	76.3(4)
O(7)-Dy(1)-N(2)	92.7(5)
O(7)-Dy(1)-N(3)	74.9(4)
O(7)-Dy(1)-N(4)	131.7(4)
O(7)-Dy(1)-O(1)	99.3(4)
O(7)-Dy(1)-O(2)	78.8(4)

Table S2 Selected bond lengths (Å) and angles (°) for **2**.

Er(1)-O(6)	2.219(4)
Er(1)-O(5)	2.231(5)
Er(1)-O(2)	2.299(5)
Er(1)-O(1)	2.321(5)
Er(1)-N(1)	2.517(6)
Er(1)-N(4)	2.537(5)
Er(1)-N(3)	2.618(5)
Er(1)-N(2)	2.618(6)
O(6)-Er(1)-O(5)	151.27(17)
O(6)-Er(1)-O(2)	79.36(16)
O(5)-Er(1)-O(2)	124.05(16)
O(6)-Er(1)-O(1)	99.40(17)
O(5)-Er(1)-O(1)	76.25(18)
O(2)-Er(1)-O(1)	71.40(17)
O(6)-Er(1)-N(1)	75.12(17)
O(5)-Er(1)-N(1)	76.17(17)
O(2)-Er(1)-N(1)	137.14(19)
O(1)-Er(1)-N(1)	79.45(19)
O(6)-Er(1)-N(4)	132.85(18)
O(5)-Er(1)-N(4)	74.94(18)
O(2)-Er(1)-N(4)	70.21(18)
O(1)-Er(1)-N(4)	103.73(18)
N(1)-Er(1)-N(4)	149.2(2)
O(6)-Er(1)-N(3)	75.02(16)
O(5)-Er(1)-N(3)	122.49(17)
O(2)-Er(1)-N(3)	78.34(17)
O(1)-Er(1)-N(3)	149.73(19)

N(1)-Er(1)-N(3)	125.70(17)
N(4)-Er(1)-N(3)	64.34(16)
O(6)-Er(1)-N(2)	92.97(17)
O(5)-Er(1)-N(2)	75.12(18)
O(2)-Er(1)-N(2)	148.04(17)
O(1)-Er(1)-N(2)	140.53(19)
N(1)-Er(1)-N(2)	67.74(19)
N(4)-Er(1)-N(2)	94.42(18)
N(3)-Er(1)-N(2)	69.72(18)

Table S3 Dy^{III} (**1**) /Er^{III} (**2**) ion geometry analysis by SHAPE 2.1 software.

Configuration	ABOXIY, **1**	ABOXIY, **2**
Square antiprism (D_{4d})	3.103	3.010
Triangular dodecahedron (D_{2d})	0.773	0.615
Biaugmented trigonal prism J50 (C_{2v})	2.886	2.858
Biaugmented trigonal prism (C_{2v})	2.977	2.860
Snub siphenoid J84 (D_{2d})	4.127	3.843

S H A P E v2.1 Continuous Shape Measures calculation

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Contact: llunell@ub.edu

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1 structures

SAPR-8 5 D4d Square antiprism

TDD-8 6 D2d Triangular dodecahedron

JBTPR-8 9 C2v Biaugmented trigonal prism J50

BTPR-8 10 C2v Biaugmented trigonal prism

JSD-8 11 D2d Snub diphenoid J84

Structure [ML8]	SAPR-8	TDD-8	JBTPR-8	BTPR-8	JSD-8
ABOXIY ,	3.103,	0.773,	2.886,	2.977,	4.127

S H A P E v2.1 Continuous Shape Measures calculation

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2 structures

SAPR-8	5 D4d	Square antiprism
TDD-8	6 D2d	Triangular dodecahedron
JBTPR-8	9 C2v	Biaugmented trigonal prism J50
BTPR-8	10 C2v	Biaugmented trigonal prism
JSD-8	11 D2d	Snub diphenoid J84

Structure [ML8]	SAPR-8	TDD-8	JBTPR-8	BTPR-8	JSD-8
ABOXIY,	3.010,	0.615,	2.858,	2.860,	3.843

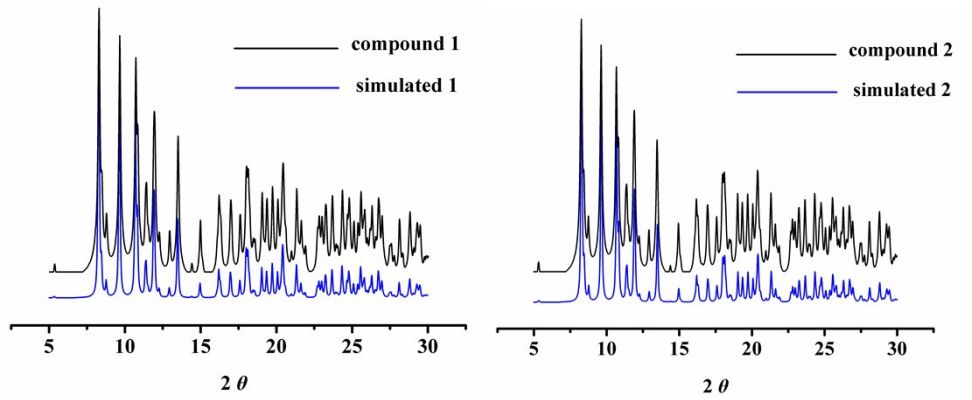

Fig. S1 XRPD curves of **1** (a) and **2** (b)**.**

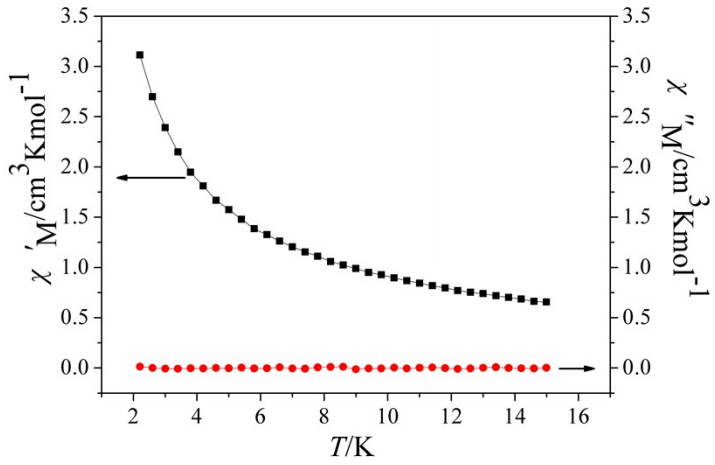

Fig. S2 Temperature dependence of the in-of-phase (χ′) and out-of-phase (χ″) of the ac susceptibility for **2** under a zero applied dc field.

The magnetic susceptibility data of **1** under a zero dc field were described by the modified Debye functions:[1]

$$\chi'(\omega) = \chi_S + (\chi_T - \chi_S)\frac{1+(\omega\tau)^{1-\alpha}\sin(\frac{\pi}{2}\alpha)}{1+2(\omega\tau)^{1-\alpha}\sin(\frac{\pi}{2}\alpha)+(\omega\tau)^{(2-2\alpha)}}$$

$$\chi''(\omega) = (\chi_T - \chi_S)\frac{(\omega\tau)^{1-\alpha}\cos(\frac{\pi}{2}\alpha)}{1+2(\omega\tau)^{1-\alpha}\sin(\frac{\pi}{2}\alpha)+(\omega\tau)^{(2-2\alpha)}}$$

$$\chi''_{\omega=\tau^{-1}} = (\chi_T - \chi_S)\frac{\cos(\frac{\pi}{2}\alpha)}{2+2\sin(\frac{\pi}{2}\alpha)} = \frac{1}{2}(\chi_T - \chi_S)\tan\frac{\pi}{4}(1-\alpha)$$

1 (a) H. X. Zhang, S. Y. Lin, S. F. Xue, C. Wang and J. K. Tang, *Dalton Trans.*, 2014, **43**, 6262−6268; (b) K. Suzuki, R. Sato and N. Mizuno, *Chem. Sci.*, 2013, **4**, 596−600; (c) K. S. Cole and R. H. Cole, *J. Chem. Phys.*, 1941, **9**, 341−351.

Table S4 Relaxation fitting parameters from Least-Squares Fitting of $\chi(\omega)$ data of **1** under a zero applied dc field.

T(K)	$\Delta\chi_1$ (cm^3mol^{-1})	$\Delta\chi_2$ (cm^3mol^{-1})	τ(s)	α
2.0	0.272	3.247	0.106×10^{-02}	0.188
2.2	0.248	2.942	0.105×10^{-02}	0.187
2.4	0.2264	2.691	0.103×10^{-02}	0.188
2.6	0.208	2.481	0.102×10^{-02}	0.189
2.8	0.194	2.298	0.997×10^{-03}	0.188
3.0	0.193	2.251	0.990×10^{-03}	0.188
3.2	0.171	2.009	0.966×10^{-03}	0.187
3.6	0.154	1.783	0.935×10^{-03}	0.188
4.0	0.141	1.601	0.903×10^{-03}	0.187
4.4	0.129	1.454	0.870×10^{-03}	0.187
4.8	0.119	1.332	0.838×10^{-03}	0.187
5.2	0.110	1.228	0.806×10^{-03}	0.187
5.6	0.102	1.140	0.774×10^{-03}	0.187
6.0	0.0972	1.063	0.745×10^{-03}	0.186
6.5	0.09231	0.981	0.709×10^{-03}	0.185
7.0	0.0871	0.910	0.674×10^{-03}	0.184
7.5	0.0825	0.849	0.641×10^{-03}	0.183
8.0	0.0787	0.796	0.609×10^{-03}	0.181
9.0	0.0723	0.707	0.549×10^{-03}	0.177
10.0	0.0672	0.636	0.494×10^{-03}	0.170
12.0	0.0587	0.530	0.397×10^{-03}	0.154
15.0	0.0454	0.424	0.278×10^{-03}	0.126
18.0	0.0256	0.354	0.187×10^{-03}	0.104

Table S5. *Ab Initio* Computed Crystal-Field Parameters up to rank=6.

		B(k,q)		
k	q	**1**	**2**	
2	-2	0.60510565359442E+00	0.17515459282406E+00	
	-1	-0.10487651939680E+01	0.15355991599981E+00	
	0	-0.41043350293965E+01	-0.86641980084076E+00	
	1	0.25180931108187E+01	-0.54987752619537E-01	
	2	0.66029427274511E+00	0.21641445673634E+01	
4	-4	0.50189290916581E-02	0.20489194469896E-01	
	-3	-0.74425136992390E-02	-0.28951637760740E-01	
	-2	-0.14441480522047E-02	0.87284903743358E-02	
	-1	0.55829387946827E-02	-0.86333218309293E-02	
	0	-0.66291571507294E-02	0.56158581805032E-03	
	1	-0.15901066224980E-01	0.58835461222758E-02	
	2	-0.69230133377082E-02	-0.11825145884374E-01	
	3	-0.83653010914577E-01	-0.35542934268359E-01	
	4	-0.11501487187023E-01	0.11385131508978E-01	
6	-6	-0.40456000061462E-04	0.19817047095936E-03	
	-5	0.12341880377796E-03	0.28418118648947E-03	
	-4	0.10435257787274E-03	-0.10858681382344E-03	
	-3	-0.30189656887200E-04	0.39390082796733E-03	
	-2	-0.20295478572625E-04	0.10531977419516E-03	
	-1	0.92370127503163E-05	-0.29200142994898E-03	
	0	0.34086906547381E-04	-0.39054832961069E-04	
	1	0.11689543956830E-04	-0.20157183139439E-03	
	2	0.37285823578108E-04	0.34821274039758E-04	
	3	-0.22628821801487E-03	-0.44711312362080E-04	
	4	-0.14339824032593E-03	-0.40119944202828E-04	
	5	0.19285394856755E-03	0.90683344561452E-05	
	6	0.15786488308522E-03	0.41623813136293E-03	