Supplementary Information

A series of dinuclear lanthanide complexes with slow magnetic relaxation for Dy_2 and Ho_2

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1. ^{1}H and ^{13}C NMR spectra of the H_2L ligand

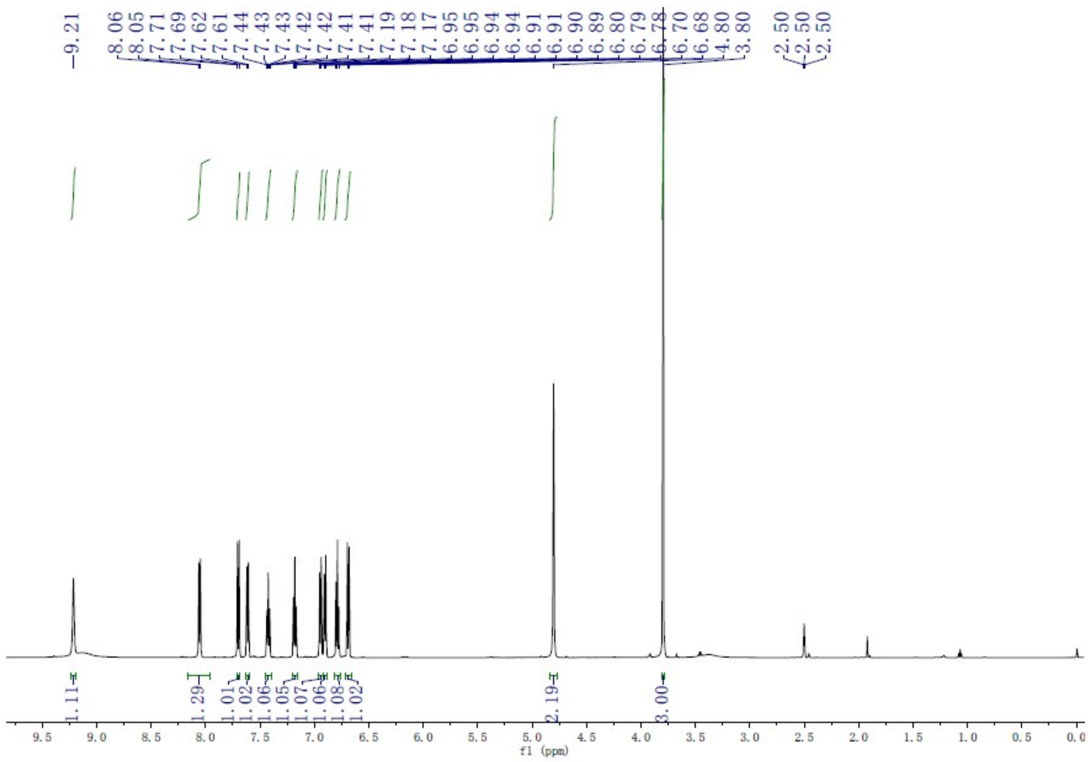


Fig. S1 ^{1}H NMR (300 MHz) spectrum of H_2L in DMSO.

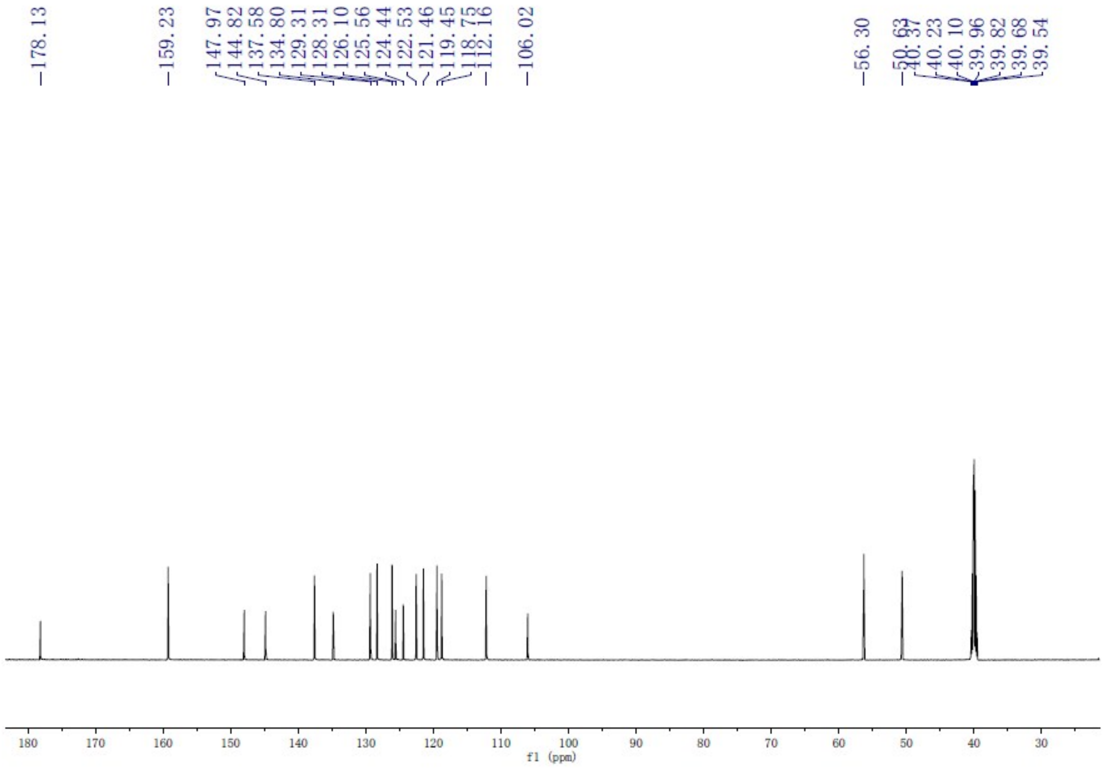


Fig. S2 ^{13}C NMR (300 MHz) spectrum of H_2L in DMSO.

2. Selected bond lengths and angles for **1-7**

Table S1 Selected Bond distances and bond angles for **1**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Eu1	N1	2.487(9)	O7	Eu1	N1	107.36(19)
Eu1	O1	2.219(8)	O8	Eu1	N1	76.5(3)
Eu1	O2	2.353(7)	O1	Eu1	N1	72.9(3)
Eu1	O5	2.392(7)	O4	Eu2	N2	71.6(3)
Eu1	O6	2.534(9)	O12	Eu2	N2	108.9(3)
Eu1	O7	2.3786(14)	O5	Eu2	N2	78.0(3)
Eu1	O8	2.470(7)	Eu1	O2	Eu2	109.8(3)
Eu1	O9	2.501(8)	Eu2	O5	Eu1	109.1(3)
Eu2	O2	2.361(7)	O1	Eu1	O6	81.1(3)
Eu2	N2	2.496(9)	O1	Eu1	O7	92.1(3)
Eu2	O3	2.560(7)	O4	Eu2	O3	79.2(2)
Eu2	O4	2.201(7)	O4	Eu2	O11	82.0(2)
Eu2	O5	2.342(7)				
Eu2	O11	2.4195(12)				
Eu2	O12	2.486(9)				
Eu2	O13	2.494(8)				
Eu1	Eu2	3.8560(13)				

Table S2 Selected Bond distances and bond angles for **2**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Gd1	N1	2.457(7)	N1	Gd1	O7	97.1(4)
Gd1	O1	2.216(7)	O1	Gd1	N1	72.4(2)
Gd1	O2	2.331(5)	O8	Gd1	N1	77.1(2)
Gd1	O5	2.366(6)	Gd1	O2	Gd2	109.6(2)
Gd1	O6	2.520(7)	Gd2	O5	Gd1	108.9(2)
Gd1	O7	2.473(4)	O4	Gd2	N2	71.9(2)
Gd1	O8	2.442(7)	O5	Gd2	N2	77.7(2)
Gd1	O9	2.492(7)	O12	Gd2	N2	109.7(3)
Gd2	O2	2.356(6)	O1	Gd1	O6	80.5(3)
Gd2	N2	2.475(8)	O1	Gd1	O7	87.7(3)
Gd2	O3	2.531(6)	O4	Gd2	O3	78.4(2)
Gd2	O4	2.190(7)	O4	Gd2	O11	82.7(3)
Gd2	O5	2.341(5)				
Gd2	O11	2.411(3)				
Gd2	O12	2.440(7)				
Gd2	O13	2.468(7)				

Gd1	Gd2	3.8312(11)

Table S3 Selected Bond distances and bond angles for **3**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Tb1	N1	2.443(9)	N1	Tb1	O8	77.5(3)
Tb1	O1	2.204(8)	O1	Tb1	N1	73.2(3)
Tb1	O2	2.319(6)	O7	Tb1	N1	95.1(4)
Tb1	O5	2.348(6)	Tb1	O2	Tb2	110.3(3)
Tb1	O6	2.517(8)	Tb2	O5	Tb1	109.7(3)
Tb1	O7	2.435(10)	O5	Tb2	N2	78.5(2)
Tb1	O8	2.448(8)	O12	Tb2	N2	108.0(3)
Tb1	O9	2.486(8)	O5	Tb2	N2	78.5(2)
Tb2	O2	2.332(7)	O1	Tb1	O6	121.8(2)
Tb2	N2	2.432(9)	O1	Tb1	O7	164.3(3)
Tb2	O3	2.518(7)	O4	Tb2	O3	78.4(2)
Tb2	O4	2.184(7)	O4	Tb2	O1	82.7(3)
Tb2	O5	2.320(6)				
Tb2	O11	2.399(9)				
Tb2	O12	2.428(8)				
Tb2	O13	2.467(8)				
Tb1	Tb2	3.8157(11)				

Table S4 Selected Bond distances and bond angles for **4**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Dy1	O1	2.186(6)	N1	Dy1	O7	98.8(3)
Dy1	O2	2.314(5)	N1	Dy1	O8	77.3(2)
Dy1	O8	2.429(7)	O1	Dy1	N1	73.5(2)
Dy1	O9	2.492(7)	O4	Dy2	N2	72.6(2)
Dy2	O2	2.330(5)	O5	Dy2	N2	78.2(2)
Dy2	N2	2.448(7)	O12	Dy2	N2	108.3(3)
Dy2	O3	2.523(6)	Dy2	O5	Dy1	109.5(2)
Dy2	O4	2.171(5)	Dy1	O2	Dy2	109.9(2)
Dy2	O5	2.317(5)	O1	Dy1	O6	78.8(2)
Dy2	O11	2.387(2)	O1	Dy1	O7	86.2(3)
Dy2	O12	2.424(7)	O4	Dy2	O3	77.5(2)
Dy2	O13	2.430(7)	O4	Dy2	O11	83.3(3)
Dy1	O6	2.513(7)				
Dy1	O7	2.447(3)				
Dy1	O5	2.339(6)				

Dy1	N1	2.416(7)
Dy1	Dy2	3.8024(11)

Table S5 Selected Bond distances and bond angles for **5**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Ho1	N1	2.413(7)	N1	Ho1	O7	98.8(3)
Ho1	O1	2.157(6)	N1	Ho1	O8	76.7(2)
Ho1	O2	2.285(5)	O1	Ho1	N1	74.1(2)
Ho1	O5	2.316(5)	O5	Ho2	N2	78.3(2)
Ho1	O6	2.488(7)	O4	Ho2	N2	72.2(2)
Ho1	O7	2.451(3)	O12	Ho2	N2	108.5(3)
Ho1	O8	2.414(7)	Ho1	O2	Ho2	109.6(2)
Ho1	O9	2.455(7)	Ho2	O5	Ho1	109.2(2)
Ho2	O2	2.310(5)	O1	Ho1	O6	78.5(2)
Ho2	N2	2.439(7)	O1	Ho1	O7	87.3(3)
Ho2	O3	2.501(6)	O4	Ho2	O3	77.5(2)
Ho2	O4	2.159(6)	O4	Ho2	O11	83.3(3)
Ho2	O5	2.291(5)				
Ho2	O11	2.394(2)				
Ho2	O12	2.410(7)				
Ho2	O13	2.438(7)				
Ho1	Ho2	3.7554(10)				

Table S6 Selected Bond distances and bond angles for **6**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Er1	N1	2.386(9)	N1	Er1	O7	97.5(4)
Er1	O1	2.161(9)	N1	Er1	O8	77.3(3)
Er1	O5	2.314(8)	O1	Er1	N1	74.6(3)
Er1	O6	2.489(8)	O4	Er2	N2	72.4(3)
Er1	O7	2.463(4)	O5	Er2	N2	78.4(3)
Er1	O8	2.419(8)	Er1	O2	Er2	110.1(3)
Er1	O9	2.432(8)	O12	Er2	N2	107.6(3)
Er2	O2	2.297(7)	Er2	O5	Er1	109.4(3)
Er2	N2	2.429(10)	O1	Er1	O6	78.0(3)
Er2	O3	2.504(7)	O1	Er1	O7	85.1(4)
Er2	O4	2.156(8)	O4	Er2	O3	77.3(3)
Er2	O5	2.292(7)	O4	Er2	O11	84.3(3)
Er2	O11	2.384(3)				
Er2	O12	2.386(9)				

Er2	O13	2.449(8)
Er1	O2	2.289(7)
Er1	Er2	3.7596(11)

Table S7 Selected Bond distances and bond angles for **7**

Atom	Atom	Length/Å	Atom	Atom	Atom	Angle/°
Yb1	O1	2.141(4)	O1	Yb1	N1	74.71(16)
Yb1	O2	2.276(4)	N1	Yb1	O8	76.92(16)
Yb1	O5	2.284(4)	N1	Yb1	O7	97.5(2)
Yb1	N1	2.383(5)	O5	Yb2	N2	78.86(15)
Yb1	O8	2.399(5)	O4	Yb2	N2	73.23(16)
Yb1	O7	2.467(2)	O12	Yb2	N2	106.89(19)
Yb1	O9	2.433(5)	Yb2	O2	Yb1	110.03(15)
Yb1	O6	2.481(5)	Yb2	O5	Yb1	109.89(15)
Yb2	O4	2.126(4)	O1	Yb1	O6	77.28(17)
Yb2	O2	2.280(4)	O1	Yb1	O7	85.7(2)
Yb2	O5	2.276(4)	O4	Yb2	O3	76.07(15)
Yb2	O11	2.3801(17)	O4	Yb2	O11	83.81(18)
Yb2	O12	2.378(5)				
Yb2	N2	2.408(5)				
Yb2	O13	2.432(5)				
Yb2	O3	2.488(4)				
Yb1	Yb2	3.7332(8)				

3. XRD patterns of **1-7**

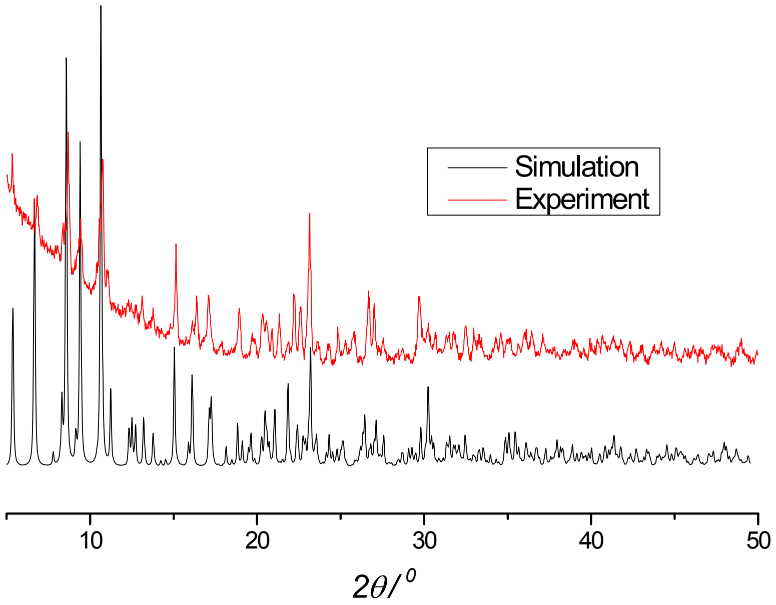


Fig. S3 PXRD patterns for **1**.

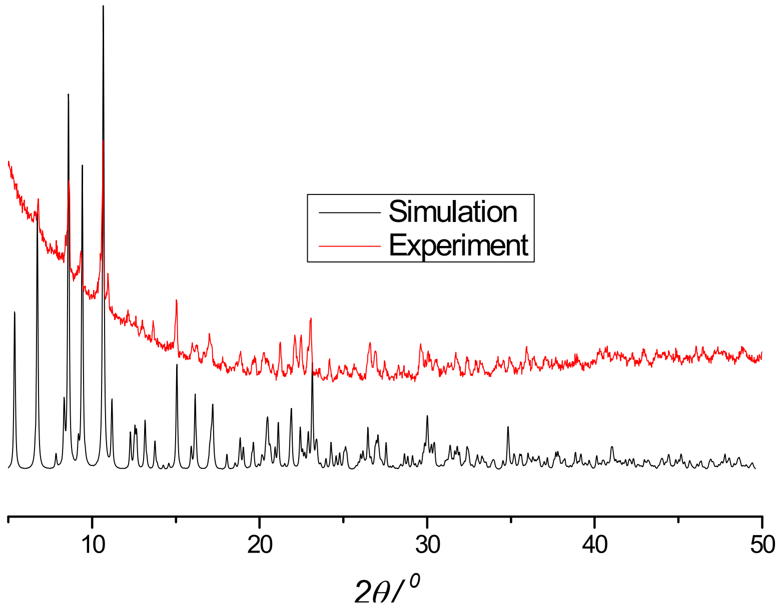


Fig. S4 PXRD patterns for **2**.

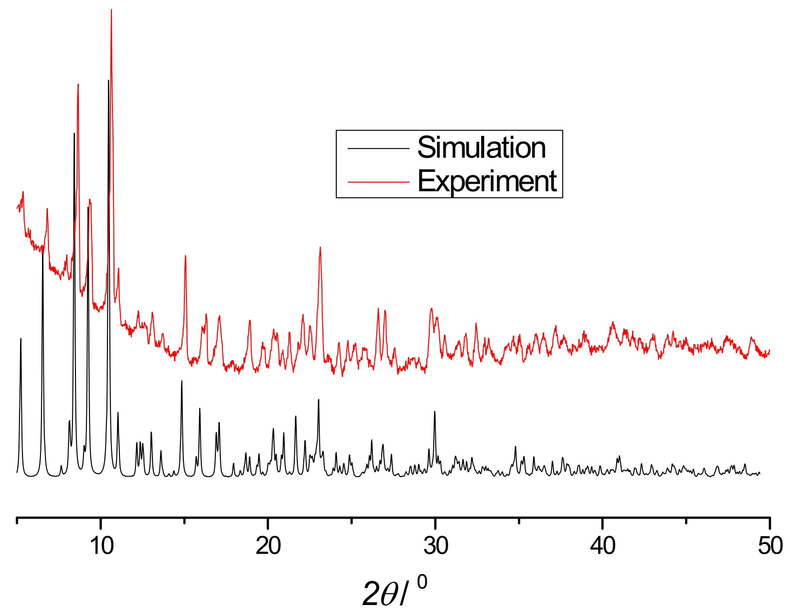


Fig. S5 PXRD patterns for **3**.

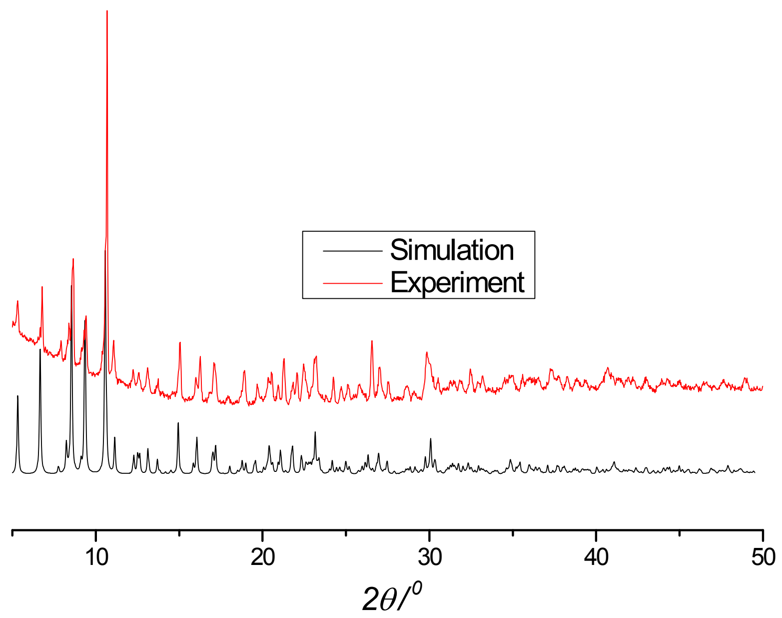


Fig. S6 PXRD patterns for **4**.

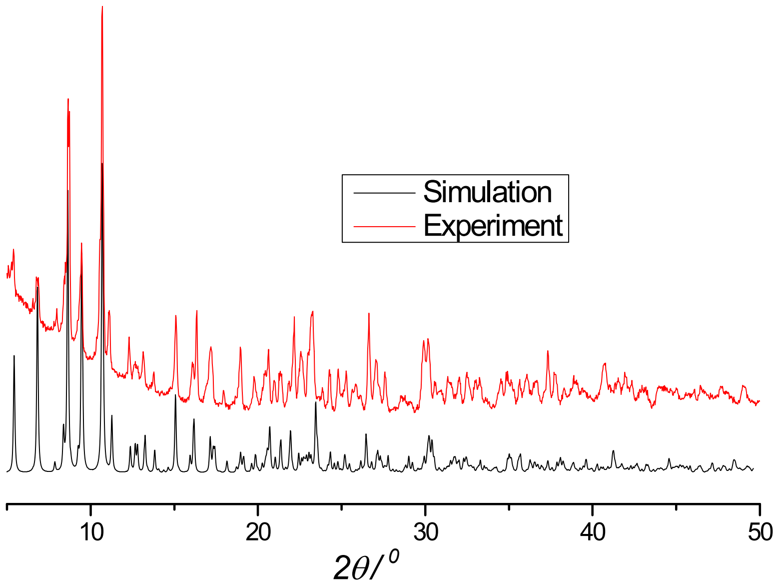


Fig. S7 PXRD patterns for **5**.

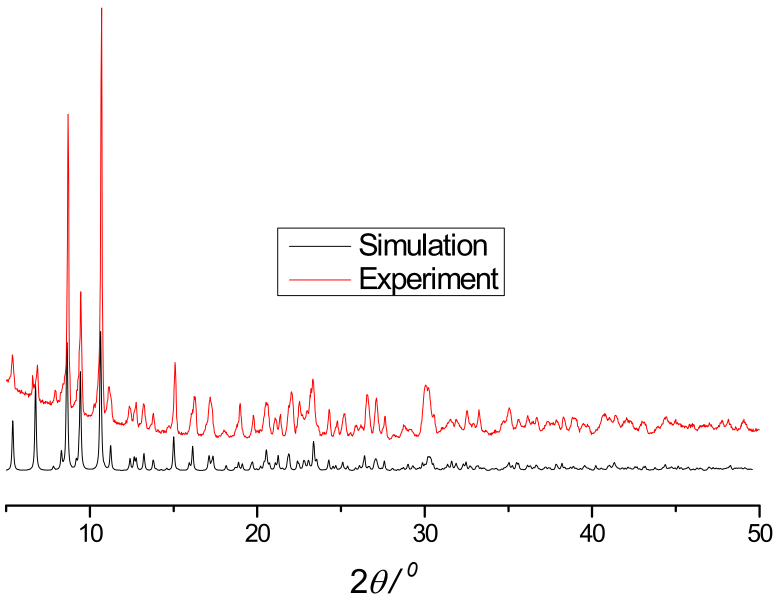


Fig. S8 PXRD patterns for **6**.

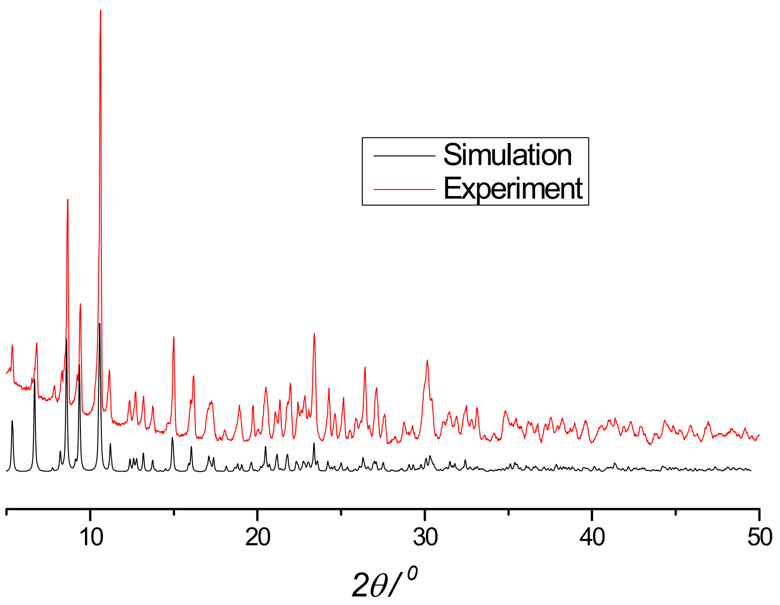


Fig. S9 PXRD patterns for **7**.

4. IR spectra of the H_2L ligand and **1-7**

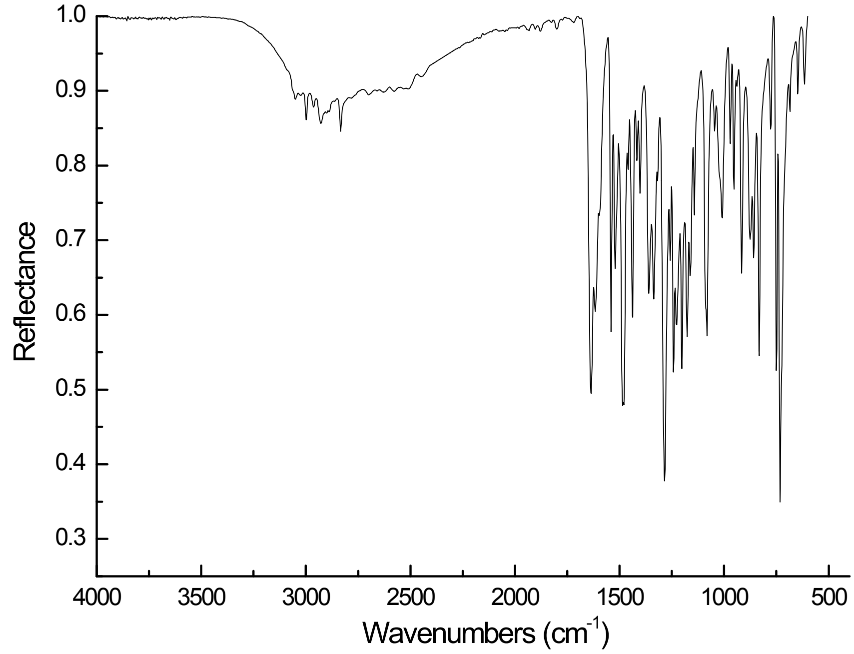


Fig. S10 The IR spectrum of H_2L.

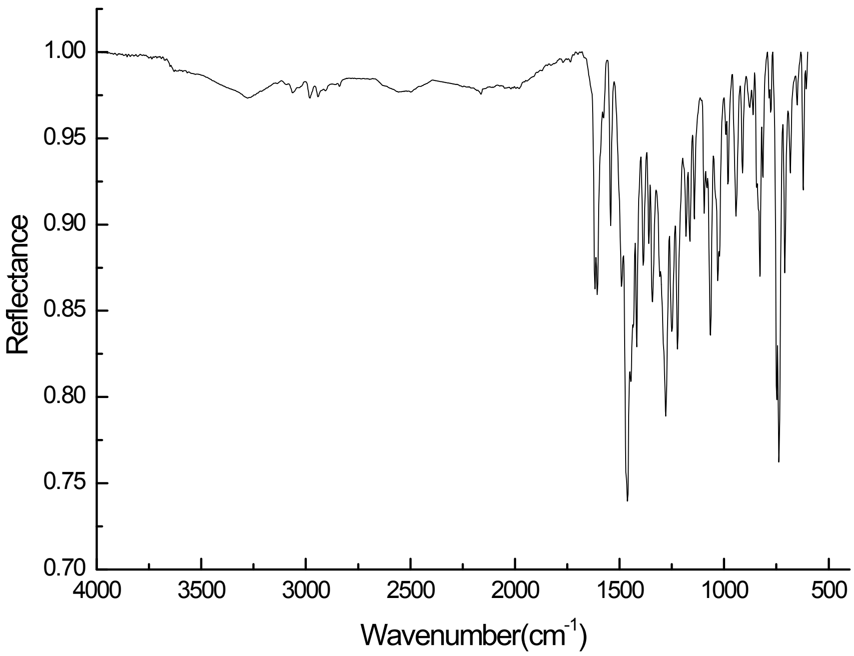


Fig. S11 The IR spectrum of **1**.

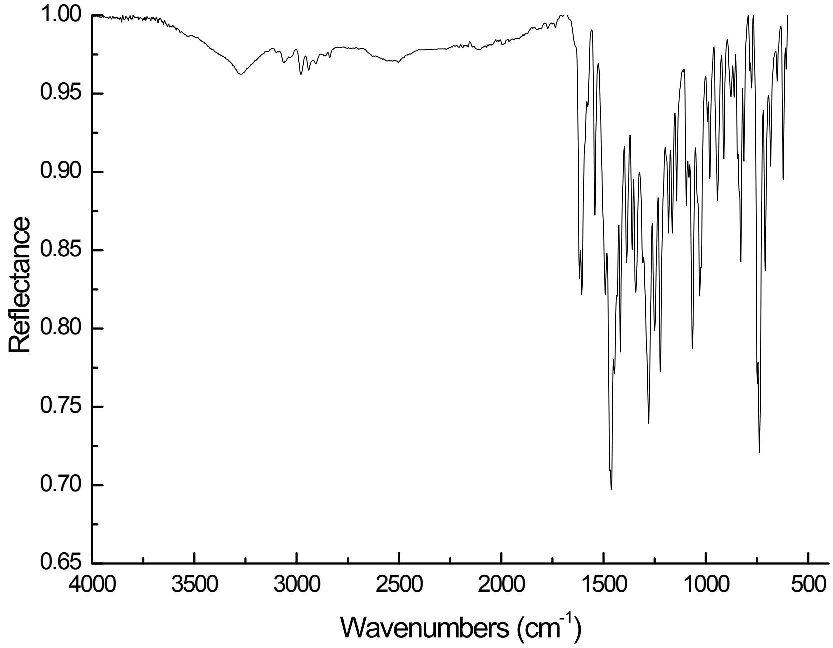


Fig. S12 The IR spectrum of **2**.

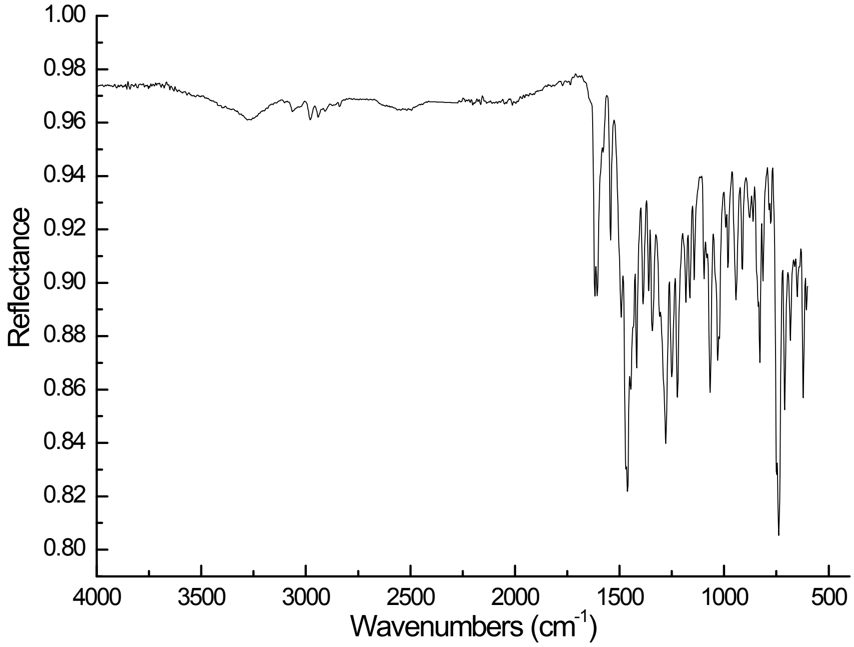


Fig. S13 The IR spectrum of **3**.

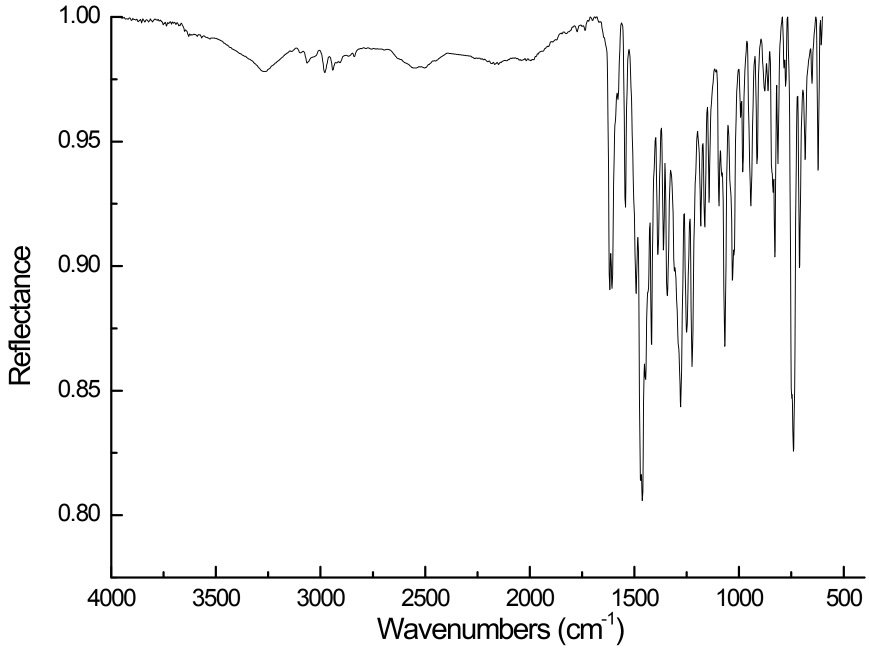


Fig. S14 The IR spectrum of **4**.

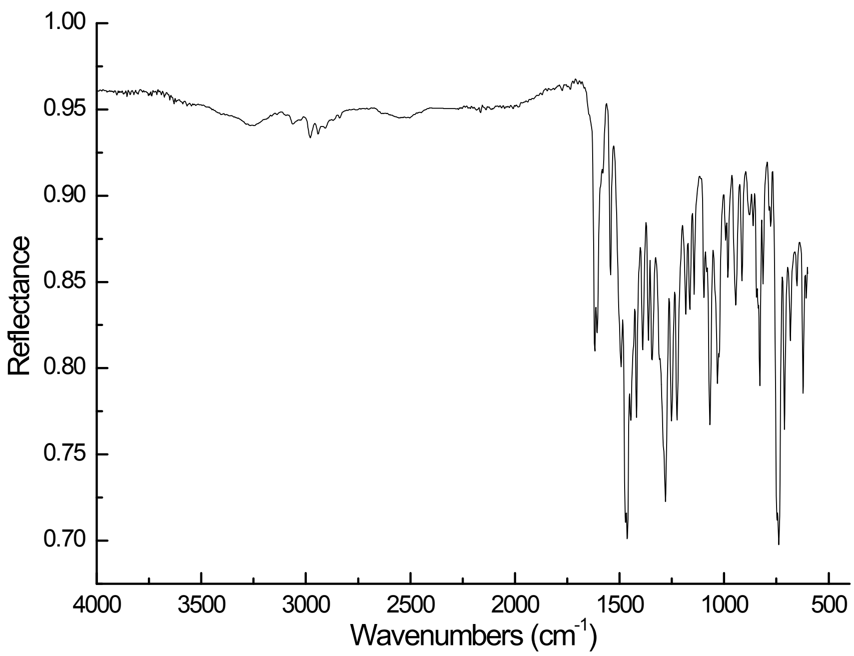


Fig. S15 The IR spectrum of **5**.

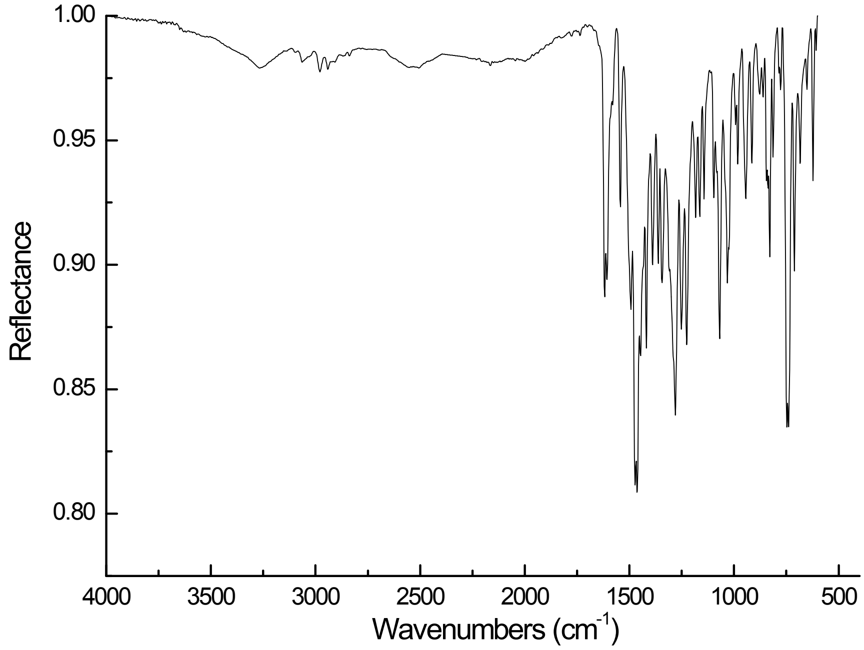


Fig. S16 The IR spectrum of **6**.

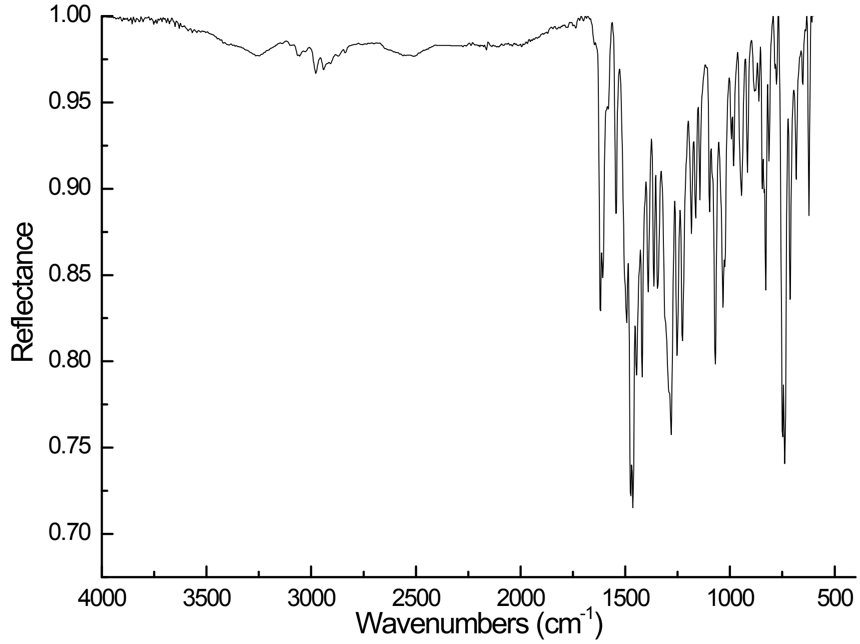


Fig. S17 The IR spectrum of **7**.

5. Dc magnetic properties of **2, 3, 4** and **6**

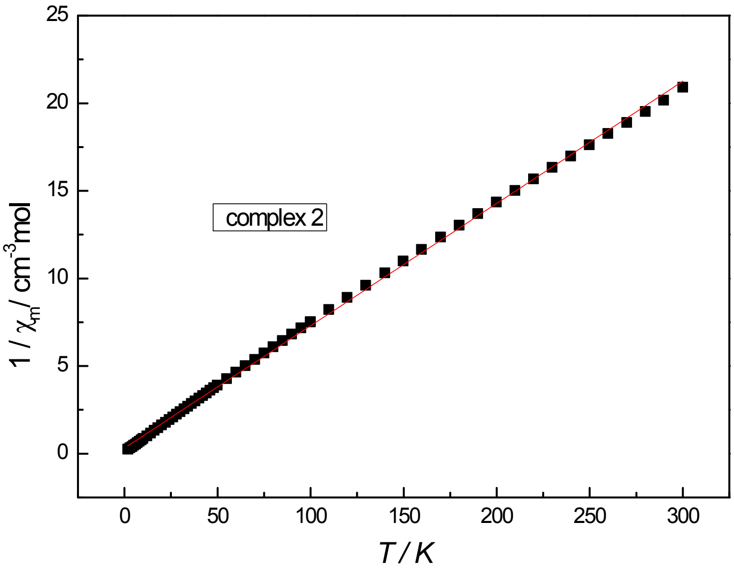


Fig. S18 The plot of $1/\chi_M$ versus T for **2** and the linear fit of Curie-Weiss law at 1000 Oe field.

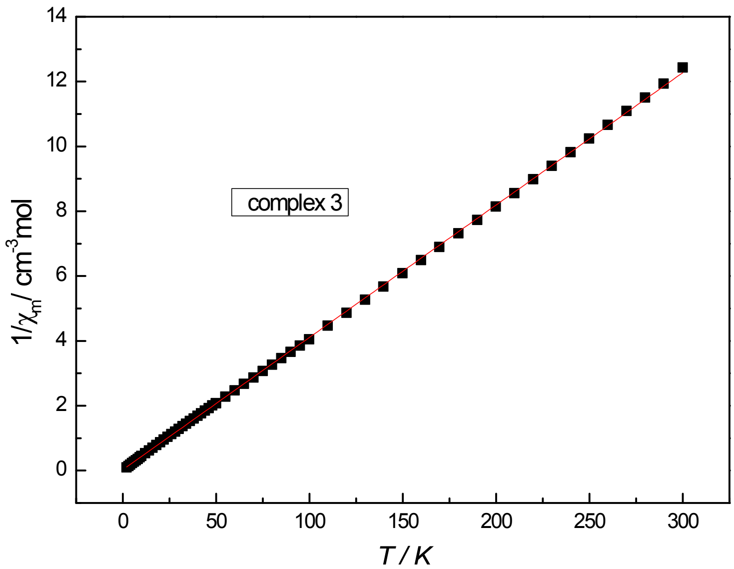


Fig. S19 The plot of $1/\chi_M$ versus T for **3** and the linear fit of Curie-Weiss law at 1000 Oe field.

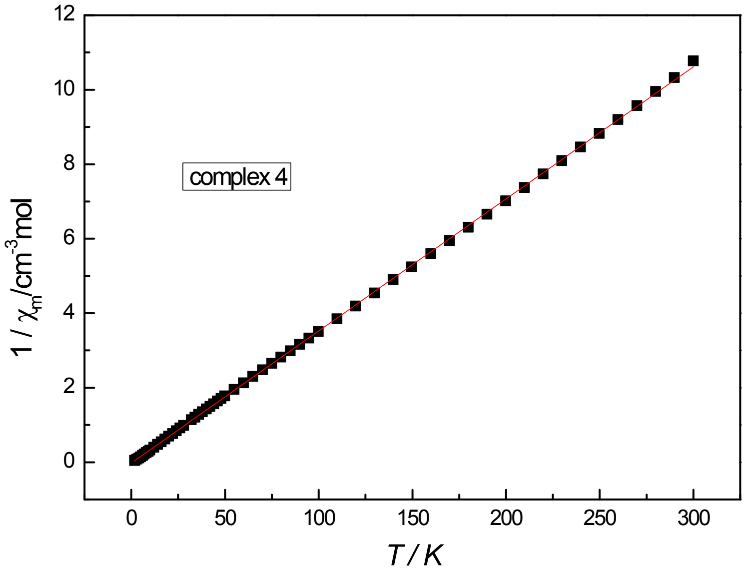


Fig. S20 The plot of 1/χM versus *T* for **4** and the linear fit of Curie-Weiss law at 1000 Oe field.

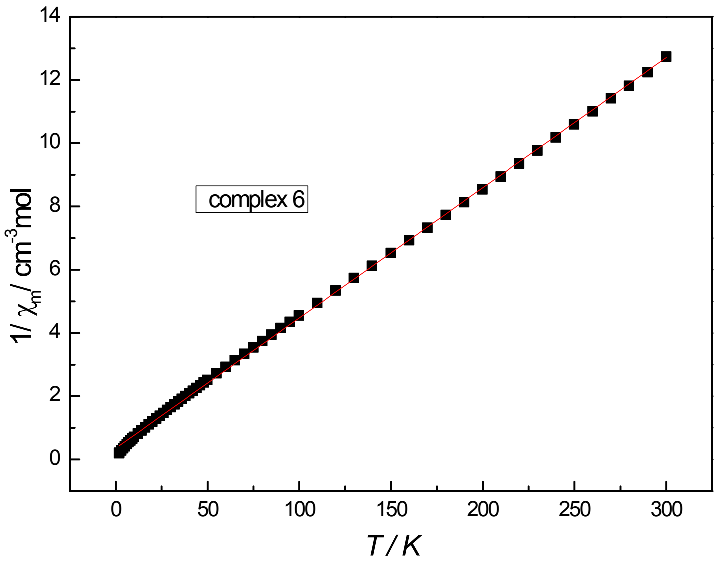


Fig. S21 The plot of 1/χM versus *T* for **6** and the linear fit of Curie-Weiss law at 1000 Oe field.

6. Parameters obtained from fitting the Cole-Cole plots of **4**

Table S8 Parameters from the least-square fitting of the Cole-Cole plots of **4** according to the generalized Debye model.

Temperature / K	X_s / $cm^3mol^{-1}K$	X_T / $cm^3mol^{-1}K$	τ / s	α
2	6.72E-02	2.04E+01	1.95E-01	1.91E-01
2.5	6.03E-02	1.41E+01	1.24E-01	1.75E-01
3	5.39E-02	1.10E+01	9.39E-02	1.69E-01
3.5	4.85E-02	8.92E+00	7.32E-02	1.66E-01
4	4.24E-02	7.75E+00	6.01E-02	1.69E-01
4.5	3.54E-02	6.94E+00	4.91E-02	1.75E-01
5	2.60E-02	6.43E+00	4.01E-02	1.86E-01
5.5	1.49E-02	6.18E+00	3.34E-02	2.03E-01
6	2.90E-03	5.87E+00	2.67E-02	2.18E-01
6.5	9.70E-16	5.54E+00	2.04E-02	2.28E-01
7	1.59E-15	5.17E+00	1.52E-02	2.33E-01
8	1.79E-15	4.37E+00	7.78E-03	2.30E-01
9	4.22E-15	3.73E+00	4.19E-03	2.23E-01
10	7.82E-15	3.25E+00	2.42E-03	2.17E-01
11	1.29E-14	2.89E+00	1.48E-03	2.12E-01
12	2.05E-14	2.60E+00	9.57E-04	2.04E-01
13	4.54E-14	2.36E+00	6.43E-04	1.96E-01
14	7.33E-14	2.16E+00	4.48E-04	1.86E-01
15	1.13E-13	1.99E+00	3.22E-04	1.76E-01
16	2.38E-13	1.85E+00	2.39E-04	1.65E-01

7. Plot of ln τ versus T^{-1} of **4** considering the Raman and Orbach processes

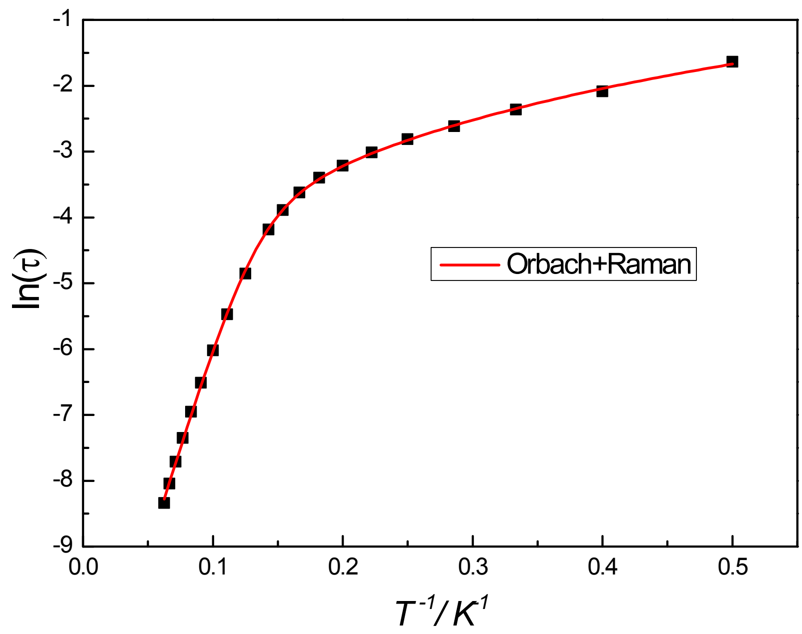


Fig. S22 The plot of ln τ versus T^{-1} of **4**. The red solid line represents the fitting by the Arrhenius law considering the Raman and Orbach processes; C = 1.67, n =1.66, U_{eff} = 67 K and $\tau_0 = 4.8 \times 10^{-6}$ s.

8. Ac magnetic properties of **4**

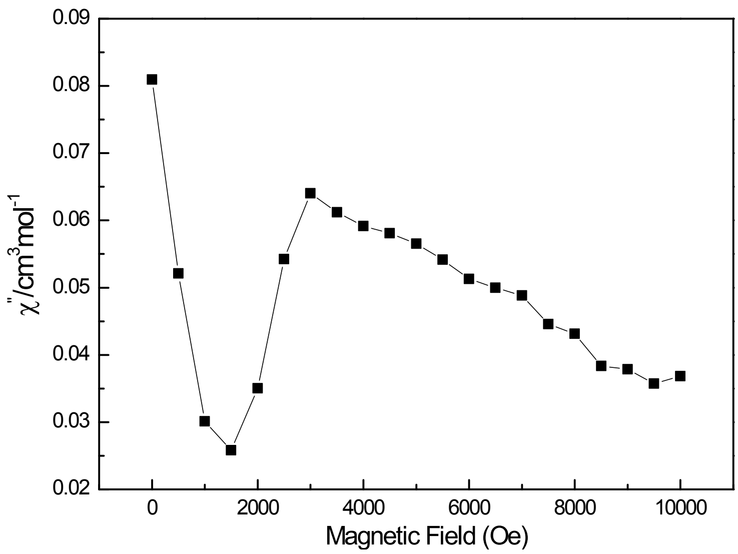


Fig. S23 The out-of-phase (χ'') ac susceptibility for **4** (2 K, f = 999Hz) under the applied static field from 0-10000 Oe.

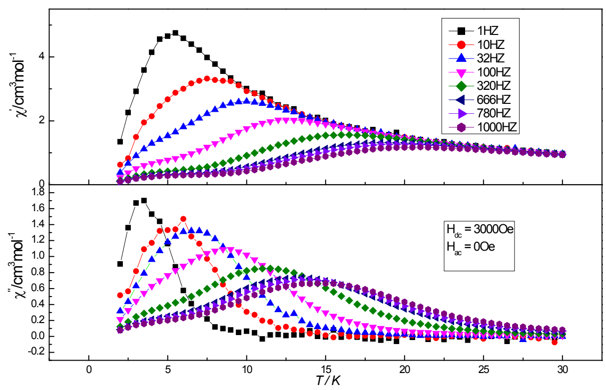


Fig. S24 Temperature dependence of the in-phase and out-of-phase ac susceptibility data for **4** under 3000 Oe dc field range from 2 to 30 K.

9. Ac Magnetic properties of **5**

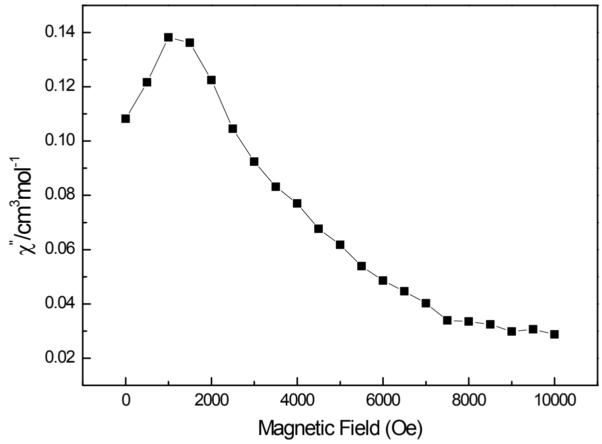


Fig. S25 The out-of-phase (χ'') ac susceptibility for **5** (2 K, f = 999Hz) under the applied static field from 0-10000 Oe.

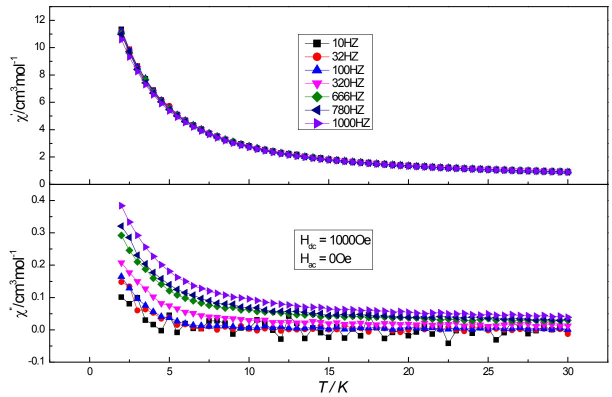
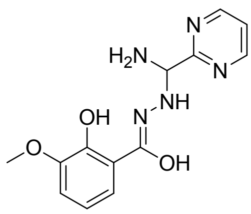
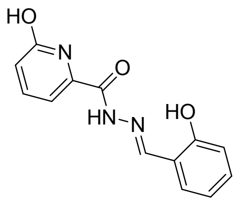
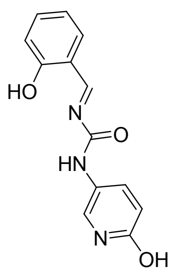
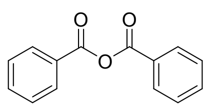
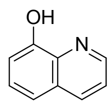
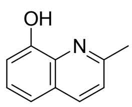
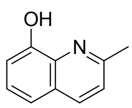
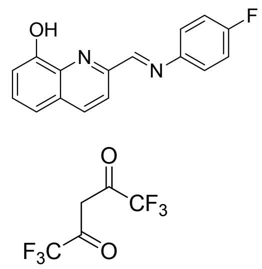
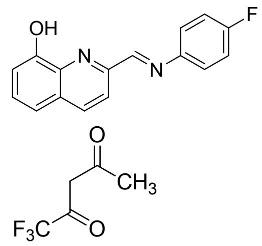
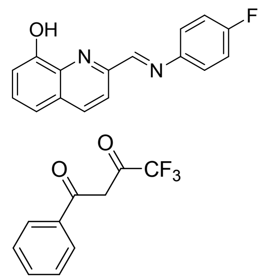
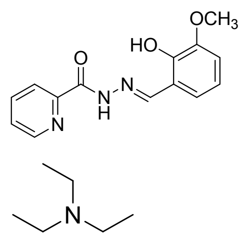
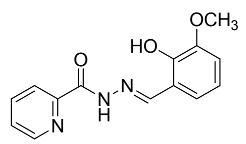
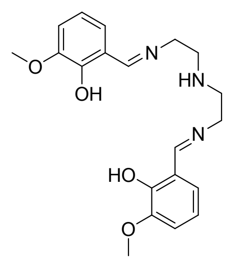
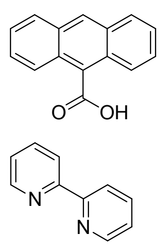


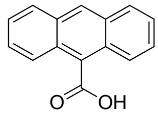
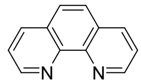
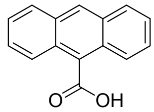
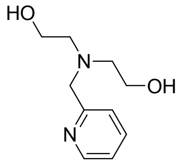
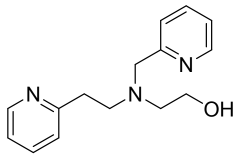
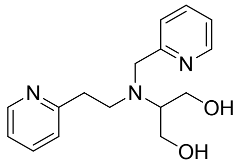
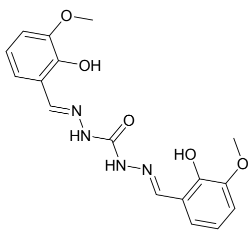
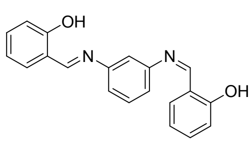
Fig. S26 Temperature dependence of the in-phase and out-of-phase ac susceptibility data for **5** under 1000 Oe dc field range from 2 to 30 K.

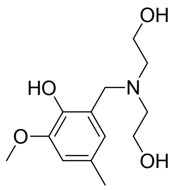
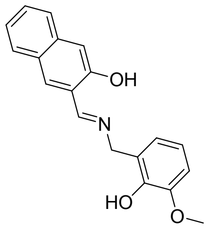
10. Dy_2 SMMs reported in the literature

Table S9 Examples of O-bridged dinuclear dysprosium complexes having SMM properties

Complexes	Ligand	Coordination Numbers and configurations	Dy…Dy distance (Å)	Dy–O–Dy bond angle (°)	Magnetic properties Ueff (K), τ_0 (s)	Ref.
$[Dy_2(a'povh)_2(OAc)_2$ $(DMF)_2]$		8,a hula-hoop-like geometry	3.6768(5)	102.62(13)	$U_{eff} = 322.1$ $\tau_0 = 3.4 \times 10^{-9}$ antiferromagnetic	1*(a)*
$[Dy_2(HL)_2Cl_2(H_2O)_3]$ $\cdot 2H_2O\cdot MeCN$		8, distorted pentagonal-bipyramidal	3.9557(8)	114.6(3)	$U_{eff1} = 204$ $\tau_{01} = 5.93 \times 10^{-9}$ $U_{eff2} = 103$ $\tau_{02} = 1.76 \times 10^{-8}$ ferromagnetic	1 (*b*)
$[Dy_2(ovph)_2Cl_2$ $(MeOH)_3]\cdot MeCN$		8,pentagonal bipyramidal	3.8644 (5)	111.5(2) - 112.3(2)	$U_{eff1} = 150$ $\tau_{01} = 2.3 \times 10^{-8}$ $U_{eff2} = 198$ $\tau_{02} = 7.3 \times 10^{-9}$ ferromagnetic	1 (*c*)
$[Dy_2(dbm)_4(OQ)_2$ $(CH_3OH)_2]$	 	8,triangular square antiprism	3.908(2)	111.45(10)	$U_{eff} = 109.5$ $\tau_0 = 4.23 \times 10^{-9}$ ferromagnetic	1 (*d*)
$[Dy_2(Mq)_4(NO3)_6]$		9,distorted trigonal prism	3.914(5)	112.26(1)	$U_{eff} = 40.01$ $\tau_0 = 5.44 \times 10^{-6}$ antiferromagnetic	1 (*e*)
$[Dy_2(Mq)_4Cl_6]$ $(EtOH)_2$		6,distorted octahedral	3.836(8)	110.80(1)	$U_{eff} = 102.4$ $\tau_0 = 3.54 \times 10^{-5}$ antiferromagnetic	1 (*e*)
$[Dy_2(hfac)_4(L)_2]$		8,distorted dodecahedral	3.7633(8	107.572(90)	$U_{eff} = 6.77$ $\tau_0 = 9.12 \times 10^{-6}$	1 (*f*)

					antiferromag- netic	
$[Dy_2(tfac)_4L_2]$		8,distorted dodecahedral	3.8800(10)	110.1(3)	$U_{eff} = 19.83$ $\tau_0 = 7.62 \times 10^{-8}$ antiferromag- netic	1 (*f*)
$[Dy_2(bfac)_4(L)_2]$ $\cdot C_7H_{16}$		8,distorted dodecahedral	3.8326(10)	108.88(10)	$U_{eff} = 25.65$ $\tau_0 = 1.64 \times 10^{-6}$ antiferromag- netic	1 (*f*)
$[Dy_2(hmi)_2(NO_3)_2$ $(MeOH)_2]$		8,distorted dodecahedral	---	106.41	$U_{eff} = 56$ $\tau_0 = 3 \times 10^{-7}$ ferromag- netic	1 (*g*)
$[Dy_2(hmi)_2(NO_3)_2$ $(MeOH)_2]\cdot MeCN$		8,distorted dodecahedral	---	107.68	$U_{eff} = 71$ $\tau_0 = 7 \times 10^{-8}$ ferromag- netic	1(*g*)
$[Dy_2(valdien)_2$ $(NO_3)_2]$		8,distorted dodecahedral	3.768(3)	108.22(3)	$U_{eff} = 76$ $\tau_0 = 6.04 \times 10^{-7}$ antiferromag- netic	1 (*h*)
$[Dy_2(\mu_2\text{-anthc})_4$ $(anthc)_2(L)_2]$ (1)		9,distorted monocapped square antiprism	3.9490(2)	105.18(6)	$U_{eff} = 51.2(3)$ $\tau_0 = 3.2(1) \times 10^{-8}$ ferromagnetic	1 (*i*)

$[Dy_2(\mu_2\text{-anthc})_4(anthc)_2(L)_2]$ (2)	 	9,distorted monocapped square antiprism	3.9176(4)	106.20(10)	U_{eff} = 49.4 τ_0 = 4.6(2) × 10^{-8} ferromagnetic	1*(i)*
$[Dy_2(\mu_2\text{-anthc})_4(anthc)_2(L)_2]$ (3)	 	9,distorted monocapped square antiprism	3.9224(6)	105.18(6)	U_{eff} = 31.6 τ_0 = 3.4(2) × 10^{-8} ferromagnetic	1 (*i*)
$[Dy_2(HL1)_2(NO_3)_4]$		9, distorted monocapped square antiprism	3.709	109.83	U_{eff} = 72.48 τ_0 = 8.504 × 10^{-8} antiferromagnetic	1 (*j*)
$[Dy_2(L2)_2(NO_3)_4]$		9, distorted monocapped square antiprism	3.706	109.71	U_{eff} = 41.55 τ_0 = 8.5 × 10^{-7} antiferromag-netic	1 (*j*)
$[Dy_2(HL3)_2(NO_3)_4]$		9, distorted monocapped square antiprism	3.719	110.65	U_{eff} = 6.77 τ_0 = 9.12 × 10^{-6} ferromagnetic	1 (*j*)
$[Dy_2(H_3L)_2(PhCOO)_4]\cdot 4H_2O$		9,tricapped trigonal prism	3.695	101.78	U_{eff} = 42.7 τ_0 = 1.37 × 10^{-7} antiferromag-netic	1 (*k*)
$[Dy_2(L)_2(acac)_2(H_2O)]\cdot 2CH_2Cl_2$		9,distorted square antiprism	3.84	110.8	$U_{eff\,1}$= 37 τ_{01} = 4.2 × 10^{-7} $U_{eff\,2}$= 80 τ_{02} = 8.3 × 10^{-8} ferromagnetic	1 (*l*)

$[Dy_2(H_2L)_2(\mu\text{-}piv)_2(piv)_2]\cdot{}_2CHCl_3$		9, distorted monocapped square-antiprism	3.633	103.02	U_{eff1} = 8.96 τ_{01} = 8.81 × 10^{-5} U_{eff2} = 35.51 τ_{02} = 1.48 × 10^{-6} antiferromag-netic	1(*m*)
$[Dy_2L_2(C_2H_5OH)_2(NO_3)_2]\cdot 0.5C_5H_5N$		8,distorted dodecahedral	3.8024(11)	109.5(2) - 109.9(2)	U_{eff} = 66.7 τ_0 = 3.76 × 10^{-6} ferromagnetic	This work

References

1 (*a*) S. Xue, Y. Guo, L. Ungur, J. Tang and L. F. Chibotaru, *Chem. Eur. J.*, 2015, **21**, 14099; (*b*) L. Zhang, J. Jung, P. Zhang, M. Guo, L. Zhao, J. Tang and B. Le Guennic, *Chem. Eur. J.*, 2016, **22**, 1392; (*c*) Y. Guo, G. Xu, W. Wernsdorfer, L. Ungur, Y. Guo, J. Tang, H. Zhang, L. F. Chibotaru and A. K. Powell, *J. Am. Chem. Soc.*, 2011, **133**, 1194; (*d*) H. Shen, W. Wang, H. Gao and J. Cui, *RSC. Adv.*, 2016, **6**, 34165; (*e*) F. Yang, Q. Zhou, G. Zeng, G. Li, L. Gao, Z. Shi and S. Feng, *Dalton Trans.*, 2014, **43**, 1238; (*f*) W. Wang, H. Zhang, S. Wang, H. Shen, H. Gao, J. Cui and B. Zhao, *Inorg. Chem.*, 2015, **54**, 10610; (*g*) P. Lin, T. J. Burchell, R. Clérac and M. Murugesu, *Angew. Chem., Int. Ed.*, 2008, **47**, 8848; (*h*) J. Long, F. Habib, P.-H. Lin, I. Korobkov, G. Enright, L. Ungur, W. Wernsdorfer, L. F. Chibotaru and M. Murugesu, *J. Am. Chem. Soc.*, 2011, **133**, 5319; (*i*) Y. Wang, C. Han, Y. Zhang, Q. Liu, C. Liu and S. Yin, *Inorg. Chem.*, 2016, **55**, 5578; (*j*) Y. Peng, V. Mereacre, A. Baniodeh, Y. Lan, M. Schlageter, G. E. Kostakis and A. K. Powell, *Inorg. Chem.*, 2016, **55**, 68; (*k*) H. Ke, S. Zhang, X. Li, Q. Wei, G. Xie, W. Wang and S. Chen, *Dalton Trans.*, 2015, **44**, 21025; (*l*) P. Lin, W. Sun, M. Yu, G. Li, P. Yan and M. Murugesu, *Chem. Commun.*, 2011, **47**, 10993; (*m*) P. Bag, C. K. Rastogi, S. Biswas, S. Sivakumar, V. Mereacre and V. Chandrasekhar, *Dalton Trans.*, 2015, **44**, 4328.