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Supporting Information

Role of Molecular Symmetry on the Magnetic Relaxation Dynamics of Five-Coordinate Dy(III) Complexes

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Table of Contents

Table S1 Selected crystallographic data and structure refinement for complexes **1-3**.

Identification code	**1**	**2**	**3**
Empirical formula	$C_{68}H_{106}Dy_2N_8O_2Si_6$	$C_{35}H_{50}DyN_5Si_3$	$C_{33}H_{54}DyN_5OSi_3$
Formula weight	1561.14	787.57	783.58
Temperature/K	150.0	170	170
Wavelength/Å	0.71073	1.54178	0.71073
Crystal system	monoclinic	monoclinic	monoclinic
Space group	$P2_1$	$P2_1/c$	$P2_1/c$
Unit cell dimensions	a = 10.0282(5) Å	a = 15.7628(5) Å	a = 10.176(2)Å
	b = 19.3323(9) Å	b = 12.1123(4) Å	b = 19.387(6) Å
	c = 19.4044(10) Å	c = 19.7909(7) Å	c = 20.093(5) Å
	α = 90°	α = 90°	α = 90°
	β = 101.790(2)°	β = 90.746(2)°	β = 102.988(9)°
	γ = 90°	γ = 90°	γ = 90°
Volume/Å^3	3682.5(3)	3776.8(2)	3862.3(17)
Z	2	4	4
Density (calculated)/g/cm^3	1.408	1.385	1.348
Absorption coefficient/mm^{-1}	2.158	11.719	2.058
F(000)	1604.0	1612.0	1612.0
Theta range for data collection	3.006 to 55.04°	5.61 to 136.88	5.428 to 54.924
Index ranges	-13 <=h<= 13	-19 <=h<= 18	-13 <=h<= 13
	-25 <=k<= 25	-14 <=k<= 14	-25 <=k<= 25
	-25 <=l<= 25	-23 <=l<= 23	-26 <=l<= 26
Reflections collected	76565	40780	86187
Independent reflections	16866 [R_{int} = 0.0556 R_{sigma} = 0.0492]	6905[R_{int} = 0.0470 R_{sigma} = 0.0314]	8828[R_{int}= 0.0396 R_{sigma} = 0.0181]
Data/restraints/parameters	16866/19/793	6905/24/406	8828/0/400
Goodness-of-fit on F^2	1.036	1.025	1.078
Final R indexes [I>=2σ (I)]	R_1 = 0.0301 wR_2 = 0.0643	R_1 = 0.0291 wR_2 = 0.0665	R_1 = 0.0211 wR_2 = 0.0443
Final R indexes [all data]	R_1 = 0.0365 wR_2 = 0.0666	R_1 = 0.0394 wR_2 = 0.0716	R_1 = 0.0255 wR_2 = 0.0459
Largest diff. peak/hole / e Å^{-3}	1.14/-0.61	0.73/-0.69	0.47/-0.67

Table S2 Selected Bond Lengths (Å) and Bond Angles (°) for **1-3**.

Complex 1			
Dy1-O1	2.425(4)	O1-Dy1-N4	172.00(14)
Dy1-N4	2.499(5)	O1-Dy1-C8	101.97(15)
Dy1-N1	2.249(5)	N4-Dy1-C8	70.18(15)
Dy1-C8	2.999(5)	N1-Dy1-O1	101.28(16)
Dy1-N3	2.249(4)	N1-Dy1-N4	83.95(16)
Dy1-N2	2.263(5)	N1-Dy1-C8	135.05(16)
Dy2-N8	2.527(5)	N1-Dy1-N3	117.54(17)
Dy2-O2	2.409(5)	N1-Dy1-N2	114.39(17)
Dy2-N7	2.242(5)	N3-Dy1-O1	88.72(15)
Dy2-N5	2.265(5)	N3-Dy1-N4	83.47(16)
Dy2-N6	2.247(5)	N3-Dy1-C8	26.76(16)
Si1-N1	1.719(5)	N3-Dy1-N2	124.50(17)
Si1-C30	1.865(7)	N2-Dy1-O1	99.38(16)
Si1-C29	1.860(7)	N2-Dy1-N4	83.73(16)
Si1-C31	1.888(7)	N2-Dy1-C8	99.01(16)
Si6-N7	1.719(5)	O2-Dy2-N8	171.40(16)
Si6-C67	1.855(7)	N7-Dy2-N8	82.78(16)
Si6-C66	1.861(7)	N7-Dy2-O2	102.63(16)
Si6-C68	1.887(8)	N7-Dy2-N5	115.50(17)
Si2-N2	1.720(5)	N7-Dy2-N6	116.39(17)
Si2-C33	1.890(7)	N5-Dy2-N8	83.44(18)
Si2-C34	1.875(7)	N5-Dy2-O2	99.92(18)
Si3-N3	1.714(5)	N6-Dy2-N8	82.89(17)
Si5-N6	1.723(5)	N6-Dy2-O2	88.69(16)
Si4-N5	1.707(5)	N6-Dy2-N5	123.75(18)

Complex 2			
Dy1-N3	2.253(3)	N3-Dy1-N4	83.90(9)
Dy1-N4	2.500(2)	N3-Dy1-N1	118.59(10)
Dy1-N1	2.258(3)	N3-Dy1-N5	93.77(10)
Dy1-N2	2.242(3)	N3-Dy1-C27	135.46(9)
Dy1-N5	2.532(3)	N1-Dy1-N4	85.18(9)
Si1-N1	1.715(3)	N1-Dy1-N5	100.86(10)
Si1-C29	1.875(4)	N1-Dy1-C27	26.77(9)
Si1-C30	1.862(4)	N2-Dy1-N3	117.83(9)
Si1-N2	1.718(3)	N2-Dy1-N4	83.20(9)
Si2-C9	1.868(4)	N2-Dy1-N1	120.44(10)
Si2-C8	1.870(4)	N2-Dy1-N5	92.98(10)
Si2-C10	1.867(4)	N2-Dy1-C27	95.50(9)
Si3-N3	1.717(3)	N5-Dy1-C27	114.31(10)
Si3-C18	1.870(4)	N1-Si1-C29	113.49(17)

Si3-C20	1.866(4)	N1-Si1-C28	110.85(17)
Si3-C19	1.863(4)	N1-Si1-C30	110.09(16)
N3-C13	1.412(4)	N2-Si2-C9	107.29(15)
N4-C11	1.491(4)	N2-Si2-C8	111.68(16)
N4-C21	1.491(4)	N2-Si2-C10	113.40(16)
N4-C1	1.491(4)	N3-Si3-C18	111.23(16)
N1-C27	1.419(4)	N3-Si3-C20	109.09(16)
N2-C3	1.416(4)	N3-Si3-C19	114.02(17)
N5-C31	1.326(5)	Si3-N3-Dy1	130.84(14)
N5-C35	1.287(6)	C13-N3-Dy1	109.75(19)
C1-N4-Dy1	110.61(18)	C11-N4-Dy1	109.88(17)
Si1-N1-Dy1	135.42(14)	C21-N4-Dy1	108.21(18)
C27-N1-Dy1	107.4(2)	Si2-N2-Dy1	129.61(14)
C3-N2-Dy1	111.05(19)	C31-N5-Dy1	122.5(3)
C35-N5-Dy1	121.5(3)	N1-C27-Dy1	45.79(14)
C22-C27-Dy1	94.2(2)	C26-C27-Dy1	129.3(2)

Complex 3			
Dy1-O1	2.1686(14)	N1-C13	1.407(3)
Dy1-N4	2.5884(17)	O1-Dy1-N4	178.67(6)
Dy1-N3	2.2720(17)	O1-Dy1-N3	100.50(6)
Dy1-N2	2.2904(18)	O1-Dy1-N2	99.08(6)
Dy1-N1	2.2684(17)	O1-Dy1-N1	97.71(6)
Si2-N2	1.7208(18	N3-Dy1-N4	80.82(6)
Si1-N1	1.7188(18)	N3-Dy1-N2	118.06(6)
Si3-N3	1.7150(19)	N2-Dy1-N4	80.46(6)
Si2-C9	1.875(2)	N1-Dy1-N4	81.50(6)
Si2-C8	1.879(2)	N1-Dy1-N3	113.35(6)
Si2-C7	1.881(2)	N1-Dy1-N2	121.23(6)
Si1-C6	1.869(2)	N2-Si2-C9	109.13(10)
Si1-C5	1.873(3)	N2-Si2-C8	114.95(10)
Si1-C4	1.874(3)	N2-Si2-C7	110.76(10)
Si3-C12	1.874(3	N1-Si1-C6	110.69(10)
Si3-C11	1.875(2)	N1-Si1-C5	108.17(10)
Si3-C10	1.888(3)	N1-Si1-C4	114.65(11)
O1-N5	1.378(2)	N3-Si3-C12	109.15(10)
N4-C26	1.495(3)	N3-Si3-C11	110.78(11)
N4-C19	1.495(3)	N3-Si3-C10	113.45(11)
N4-C33	1.496(3)	N5-O1-Dy1	175.81(13)
N5-C1	1.495(3)	Si1-N1-Dy1	129.04(9)
N5-C3	1.490(3)	Si3-N3-Dy1	124.80(9)
N5-C2	1.497(3)	C33-N4-Dy1	110.98(12)
N3-C27	1.418(3)	C13-N1-Dy1	109.08(12)
N2-C20	1.414(3)	C27-N3-Dy1	114.34(13)

Table S3 Relaxation fitting parameters from least-squares fitting of $\chi(f)$ data under 500 Oe dc field of **1**.

T(K)	χ_T	χ_S	α
2	3.991	1.416	0.266
2.3	3.769	1.261	0.184
2.6	3.512	1.147	0.111
3	3.167	1.008	0.051
3.3	2.947	0.946	0.016
3.6	2.743	0.841	0.031
4	2.476	0.764	0.014
4.3	2.325	0.762	0.023
4.6	2.175	0.724	0.017
5	2.011	0.719	0.019
5.5	1.837	0.741	0.021
6	1.686	0.838	0.015

Table S4 Relaxation fitting parameters from least-squares fitting of $\chi(f)$ data under 500 Oe dc field of **2**.

T(K)	χ_T	χ_S	α
2.6	3.278	0.510	0.437
3	2.926	0.872	0.338
3.3	2.692	1.110	0.212
3.6	2.488	1.070	0.160
4	2.244	1.012	0.078
4.3	2.101	0.841	0.109
4.6	1.967	0.675	0.074
5	1.818	0.130	0.106
5.5	1.659	2.24×10^{-11}	0.145
6	1.528	3.81×10^{-11}	0.231

Table S5 Relaxation fitting parameters from least-squares fitting of $\chi(f)$ data under 500 Oe dc field of **3**.

T(K)	χ_T	χ_S	α
2	4.453	0.657	0.372
2.3	4.084	0.613	0.304
2.6	3.748	0.542	0.288
3	3.370	0.477	0.296
3.3	3.102	0.447	0.317
3.6	2.842	0.488	0.323
4	2.570	0.703	0.302
4.3	2.394	0.833	0.297
4.6	2.222	0.931	0.272
5	2.053	1.014	0.258
5.5	1.871	1.036	0.273
6	1.720	1.079	0.299

Table S6 Calculated energy levels (cm^{-1}), ***g*** (g_x, g_y, g_z) tensors and predominant m_J values of the lowest eight Kramers doublets (KDs) of complexes **1*a***, **1*b***, **2** and **3**, using CASSCF/RASSI-SO with OpenMolcas.

KDs	**1*a***			**1*b***		
	E	***g***	m_J	*E*	***g***	m_J
0	0.0	0.180 0.437 19.284	±15/2	0.0	0.124 0.279 19.410	±15/2
1	140.7	10.385 8.330 2.506	±13/2	160.7	1.945 4.763 13.550	±13/2
2	213.2	6.646 4.333 1.681	±5/2	246.3	7.781 5.448 2.258	±9/2
3	295.2	1.051 3.490 12.742	±7/2	334.2	0.263 3.509 10.837	±5/2
4	343.2	0.118 1.868 15.017	±3/2	378.0	0.607 1.546 17.046	±1/2
5	368.8	1.223 1.924 16.719	±1/2	396.3	1.595 2.182 15.897	±3/2
6	469.3	0.250 0.336 19.110	±11/2	534.5	0.185 0.467 18.461	±11/2
7	574.7	0.036 0.080 19.681	±9/2	579.6	0.166 0.610 19.073	±7/2
KDs	**2**			**3**		
	E	***g***	m_J	*E*	***g***	m_J
0	0.0	0.902 3.871 16.514	±15/2	0	0.0	0.127 0.316 19.054
1	71.7	3.556 5.248 7.447	±9/2	1	83.8	0.575 1.851 14.941
2	160.4	6.167 5.012 3.482	±5/2	2	112.2	0.530 1.007 17.069

3	260.7	1.859 4.121 11.939	±11/2	3	161.3	3.925 5.182 11.268
4	288.1	1.513 1.899 16.835	±3/2	4	203.0	1.533 2.297 13.292
5	393.1	0.334 1.754 17.172	±1/2	5	261.1	0.075 2.161 14.872
6	407.8	0.511 2.421 15.505	±7/2	6	274.6	0.529 2.279 14.878
7	419.0	0.174 1.224 17.290	±13/2	7	403.5	0.007 0.020 19.732

Table S7 Wave functions with definite projection of the total moment | m_J > for the lowest eight KDs of complexes **1*a***, **1*b***, **2** and **3** using CASSCF/RASSI-SO with OpenMolcas.

	states	E/cm^{-1}	wave functions
1*a*	KD_0	0.0	92.9%|±15/2>
	KD_1	140.7	42.2%|±13/2>+22.9%|±9/2>+15.7%|±5/2>
	KD_2	213.2	19.5%|±13/2>+27.1%|±11/2>+27.7%|±7/2>+11.2%|±3/2>
	KD_3	295.2	20.8%|±13/2>+22.9%|±5/2>+24.9%|±3/2>+12.6%|±1/2>+9.6%|±11/2>
	KD_4	343.2	11.1%|±13/2>+20.8%|±11/2>+23.2%|±9/2>+27.2%|±1/2>+6.8%|±7/2>+6.7%|±3/2>
	KD_5	368.8	17.7%|±11/2>+16.0%|±9/2>+19.9%|±7/2>+14.8%|±5/2>+10.9%|±3/2>+18.1%|±1/2>
	KD_6	469.3	13.8%|±11/2>+19.5%|±9/2>+18.4%|±7/2>+18.7%|±5/2>+19.1%|±3/2>+7.2%|±1/2>
	KD_7	574.7	11.0%|±9/2>+17.7%|±7/2>+20.5%|±5/2>+21.3%|±3/2>+23.9%|±1/2>
1*b*	KD_0	0.0	94.0%|±15/2>
	KD_1	160.7	54.6%|±13/2>+19.9%|±9/2>+11.1%|±5/2>
	KD_2	246.3	16.4%|±13/2>+30.9%|±11/2>+25.3%|±7/2>+11.5%|±3/2>
	KD_3	334.2	19.5%|±13/2>+15.0%|±11/2>+14.0%|±9/2>+22.8%|±5/2>+21.9%|±3/2>
	KD_4	378.0	15.1%|±11/2>+17.7%|±9/2>+10.6%|±7/2>+9.0%|±5/2>+12.9%|±3/2>+30.0%|±1/2>
	KD_5	396.3	17.9%|±11/2>+20.0%|±9/2>+20.9%|±7/2>+10.0%|±5/2>+22.5%|±1/2>
	KD_6	534.5	10.4%|±11/2>+13.8%|±9/2>+11.6%|±7/2>+20.4%|±5/2>+29.2%|±3/2>+12.2%|±1/2>
	KD_7	579.6	13.8%|±9/2>+23.9%|±7/2>+21.3%|±5/2>+14.8%|±3/2>+21.3%|±1/2>
2	KD_0	0.0	70.8%|±15/2>+12.1%|±11/2>+9.2%|±7/2>
	KD_1	71.7	12.2%|±15/2>+22.6%|±13/2>+24.4%|±9/2>+20.9%|±5/2>+9.4%|±1/2>
	KD_2	160.4	13.2%|±15/2>+20.7%|±13/2>+17.7%|±11/2>+23.1%|±7/2>+13.7%|±3/2>
	KD_3	260.7	29.1%|±13/2>+17.1%|±11/2>+10.3%|±9/2>+9.3%|±7/2>+11.0%|±5/2>+19.9%|±1/2>
	KD_4	288.1	21.8%|±5/2>+29.3%|±3/2>+22.0%|±1/2>+9.0%|±7/2>
	KD_5	393.1	12.2%|±9/2>+17.1%|±7/2>+22.1%|±5/2>+19.5%|±3/2>+24.2%|±1/2>

	KD_6	407.8	17.3%|±11/2>+15.8%|±9/2>+14.8%|±5/2>+22.3%|±3/2>+16.0%|±1/2>
	KD_7	419.0	11.9%|±13/2>+26.8%|±11/2>+24.2%|±9/2>+22.5%|±7/2>+6.9%|±5/2>
3	KD_0	0.0	88.1%|±15/2>+8.4%|±11/2>
	KD_1	83.8	49.7%|±13/2>+21.0%|±9/2>+8.7%|±7/2>+6.3%|±11/2>
	KD_2	112.2	35.1%|±1/2>+29.5%|±3/2>+17.7%|±5/2>+5.8%|±13/2>
	KD_3	161.3	12.4%|±13/2>+21.5%|±11/2>+16.5%|±7/2>+19.1%|±3/2>+16.5%|±1/2>
	KD_4	203.0	23.2%|±7/2>+34.4%|±5/2>+12.9%|±3/2>+8.7%|±9/2>+9.2%|±13/2>
	KD_5	261.0	12.8%|±13/2>+28.2%|±11/2>+25.2%|±9/2>+19.4%|±7/2>+8.7%|±5/2>
	KD_6	274.7	9.6%|±13/2>+23.5%|±11/2>+32.7%|±9/2>+16.1%|±7/2>+8.2%|±5/2>
	KD_7	403.5	11.0%|±7/2>+18.2%|±5/2>+27.7%|±3/2>+34.9%|±1/2>

Table S8 Calculated crystal-field parameters B (k, q) and the corresponding weights for compounds **1*a***, **1*b***, **2** and **3** using CASSCF/RASSI-SO with OpenMolcas.

1*a*				**1*b***			
k	q	B (k, q)	Weight (%)	k	q	B (k, q)	Weight (%)
2	–2	-0.8059×10^{0}	5.89	2	–2	0.3451×10^{0}	2.58
	–1	-0.4799×10^{-1}	0.35		–1	0.1266×10^{0}	0.95
	0	-0.1900×10^{1}	13.89		0	-0.2173×10^{1}	16.23
	1	0.9764×10^{0}	7.14		1	-0.9865×10^{0}	7.37
	2	0.1540×10^{1}	11.26		2	0.1422×10^{1}	10.62
4	–4	0.4223×10^{-4}	0.05	4	–4	0.6635×10^{-4}	0.08
	–3	-0.2564×10^{-3}	0.34		–3	-0.3541×10^{-3}	0.48
	–2	0.2679×10^{-2}	3.55		–2	-0.1019×10^{-2}	1.38
	–1	0.3140×10^{-3}	0.41		–1	-0.4204×10^{-3}	0.56
	0	-0.4347×10^{-2}	5.76		0	-0.4542×10^{-2}	6.15
	1	-0.4345×10^{-2}	5.76		1	0.4517×10^{-2}	6.12
	2	0.5615×10^{-2}	7.44		2	0.5926×10^{-2}	8.03
	3	-0.3294×10^{-2}	4.36		3	0.3513×10^{-2}	4.76
	4	-0.8820×10^{-2}	11.70		4	-0.9211×10^{-2}	12.48
6	–6	0.1678×10^{-4}	2.08	6	–6	-0.9665×10^{-5}	1.22
	–5	0.3630×10^{-5}	0.45		–5	0.1843×10^{-5}	0.23
	–4	0.4637×10^{-5}	0.57		–4	-0.1155×10^{-5}	0.14
	–3	-0.1048×10^{-5}	0.13		–3	-0.1020×10^{-4}	1.29
	–2	-0.8241×10^{-5}	1.02		–2	0.4034×10^{-5}	0.51
	–1	-0.6025×10^{-5}	0.74		–1	-0.1394×10^{-4}	1.77
	0	-0.1555×10^{-4}	1.93		0	-0.1150×10^{-4}	1.46
	1	0.1437×10^{-4}	1.78		1	-0.1550×10^{-4}	1.97
	2	-0.1195×10^{-4}	1.48		2	-0.9078×10^{-5}	1.15
	3	0.1034×10^{-4}	1.28		3	-0.1115×10^{-4}	1.41
	4	-0.2470×10^{-4}	3.07		4	-0.2328×10^{-4}	2.96
	5	-0.2602×10^{-4}	3.23		5	0.2755×10^{-4}	3.50

	6	0.2290×10^{-4}	2.84		6	0.2373×10^{-4}	3.01
2				**3**			
k	q	$B(k, q)$	Weight (%)	k	q	$B(k, q)$	Weight (%)
2	–2	-0.1223×10^{0}	1.03	2	–2	0.7437×10^{0}	7.65
	–1	0.8921×10^{-1}	0.75		–1	0.2953×10^{0}	3.04
	0	-0.1188×10^{1}	9.96		0	-0.1055×10^{1}	10.86
	1	0.2054×10^{0}	1.72		1	0.1601×10^{0}	1.64
	2	0.1917×10^{1}	16.07		2	0.7596×10^{-1}	0.78
4	–4	-0.8416×10^{-3}	1.28	4	–4	-0.9016×10^{-3}	1.68
	–3	0.2079×10^{-3}	0.31		–3	-0.3429×10^{-3}	0.64
	–2	-0.2304×10^{-2}	3.50		–2	-0.6220×10^{-2}	11.62
	–1	-0.1594×10^{-3}	0.24		–1	-0.1420×10^{-2}	2.65
	0	-0.3845×10^{-2}	5.84		0	-0.2959×10^{-2}	5.52
	1	0.2602×10^{-2}	3.95		1	-0.5365×10^{-3}	1.00
	2	0.6851×10^{-2}	10.42		2	0.6164×10^{-2}	11.51
	3	0.4256×10^{-2}	6.47		3	-0.1279×10^{-2}	2.39
	4	-0.9098×10^{-2}	13.84		4	-0.7803×10^{-2}	14.58
6	–6	-0.6745×10^{-5}	0.96	6	–6	-0.1792×10^{-4}	3.14
	–5	-0.3191×10^{-5}	0.45		–5	-0.4628×10^{-5}	0.81
	–4	-0.1238×10^{-4}	1.76		–4	-0.1221×10^{-4}	2.13
	–3	0.3479×10^{-5}	0.49		–3	-0.1663×10^{-4}	2.91
	–2	0.1096×10^{-4}	1.56		–2	0.1842×10^{-4}	3.22
	–1	0.2523×10^{-5}	0.35		–1	-0.2215×10^{-4}	3.88
	0	-0.2138×10^{-4}	3.05		0	-0.7196×10^{-5}	1.26
	1	-0.2162×10^{-4}	3.08		1	0.5511×10^{-5}	0.96
	2	-0.6993×10^{-5}	0.99		2	0.2163×10^{-5}	0.37
	3	0.9557×10^{-6}	0.13		3	0.5988×10^{-6}	0.10
	4	-0.2889×10^{-4}	4.12		4	-0.7199×10^{-5}	1.26
	5	0.1705×10^{-4}	2.43		5	-0.4380×10^{-5}	0.76
	6	0.2344×10^{-4}	3.34		6	0.1066×10^{-4}	1.86

Table S9 Continuous Shape Measures (*CShM*) calculations for complexes **1**-**3**.

	Structure	**PP-5**	**vOC-5**	**TBPY-5**	**SPY-5**	**JTBPY-5**
1 (Dy01)	**CShM**	34.261	5.781	0.835	4.306	2.207
1 (Dy02)	**CShM**	34.407	5.892	0.890	4.412	2.185
2	**CShM**	35.542	7.113	0.737	5.250	1.735
3	**CShM**	35.217	6.537	0.531	5.027	2.512

PP-5 Pentagon (D_{5h})

vOC-5 Vacant octahedron (C_{4v})

TBPY-5 Trigonal bipyramid (D_{3h})

SPY-5 Spherical square pyramid (C_{4v})

JTBPY-5 Johnson trigonal bipyramid J12 (D_{3h})

Table S10 Crystal field parameters for **1-3** fitted from magnetic data.

	B_0^2	B_0^4	B_2^4	B_0^6
1(Dy1)	301	-16	208	52
2	187	37	44	38
3	79	56	221	283

Table S11 Energy levels and ***g*** for **1-3** fitted from magnetic data.

KDs	**1(Dy1)**		**2**		**3**	
	E/cm^{-1}	***g***	E/cm^{-1}	***g***	E/cm^{-1}	***g***
1	0	0.0000 0.0000 19.7734	0	0.0000 0.0000 18.8536	0	0.0004 0.0066 19.0064
2	128	0.0000 0.0000 17.2346	91	0.0000 0.0000 15.9878	107	0.6460 2.4864 17.1728

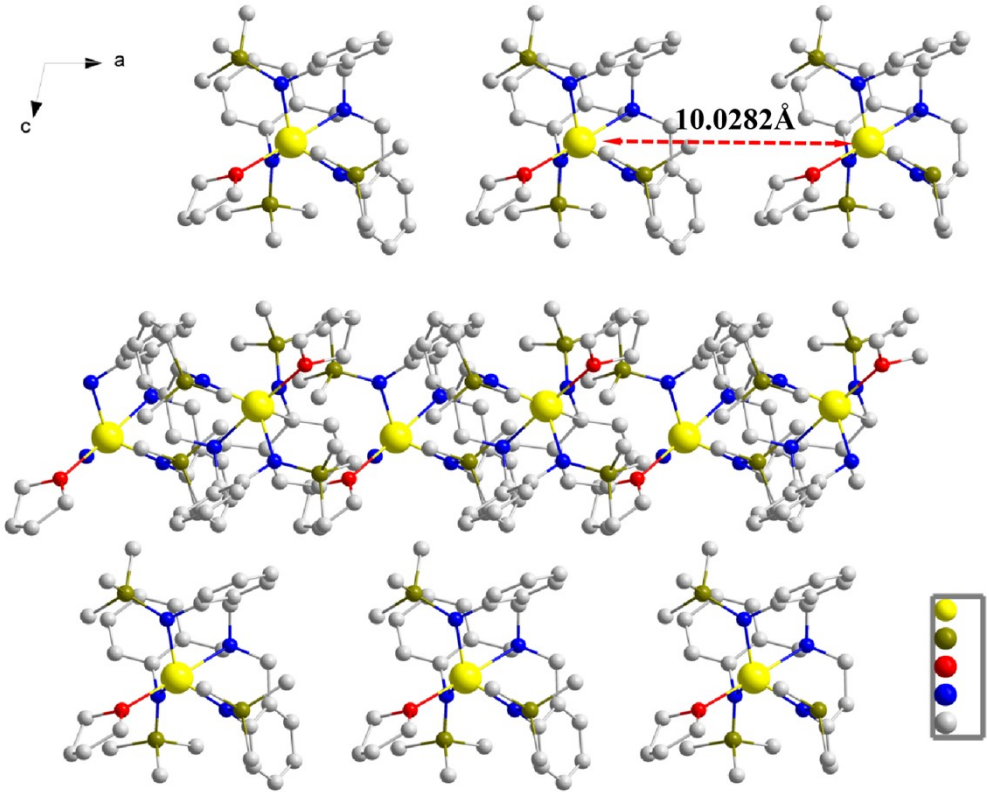

Fig. S1 Packing arrangement of **1** along the crystallographic *b* axis.

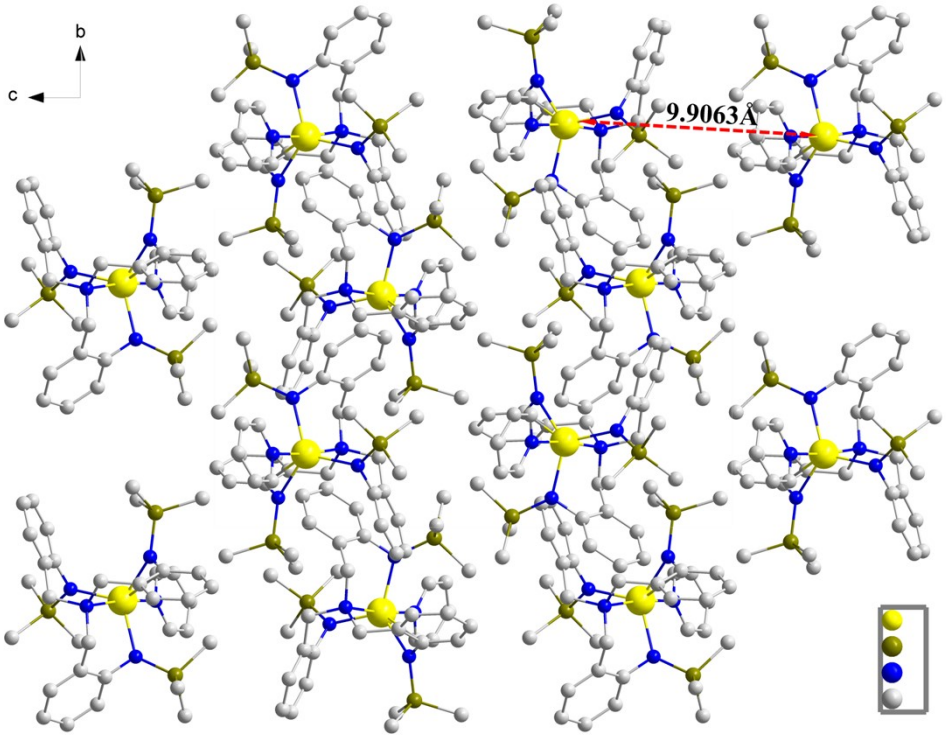

Fig. S2 Packing arrangement of **2** along the crystallographic *a* axis.

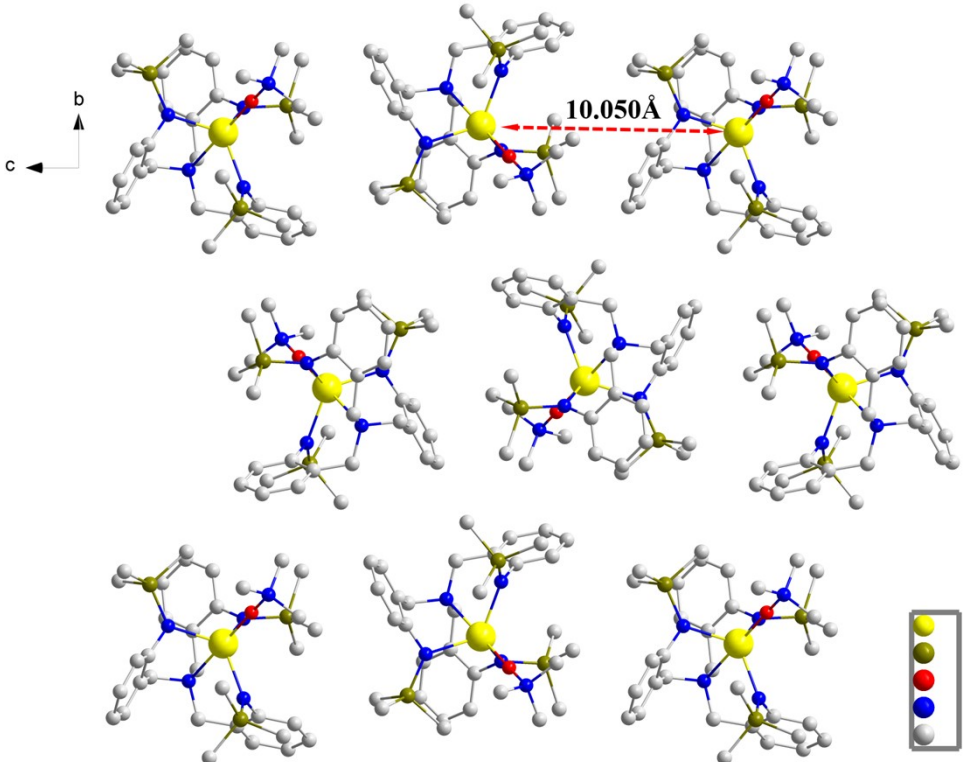

Fig. S3 Packing arrangement of **3** along the crystallographic *a* axis.

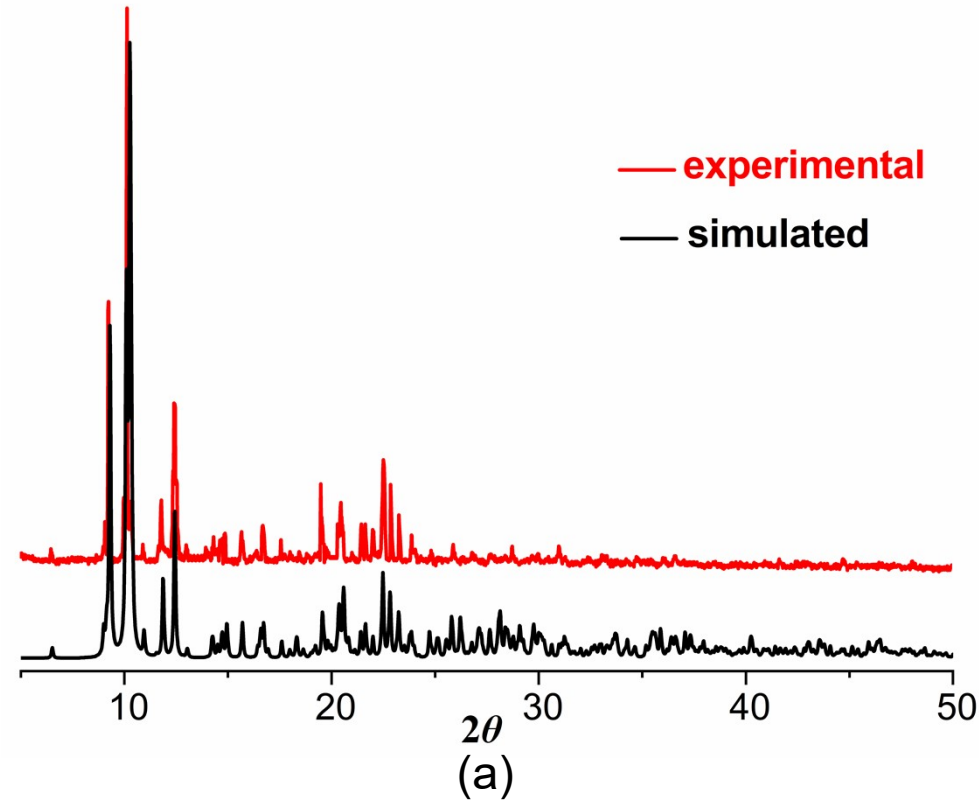

Fig. S4 PXRD patterns for complex **1**.

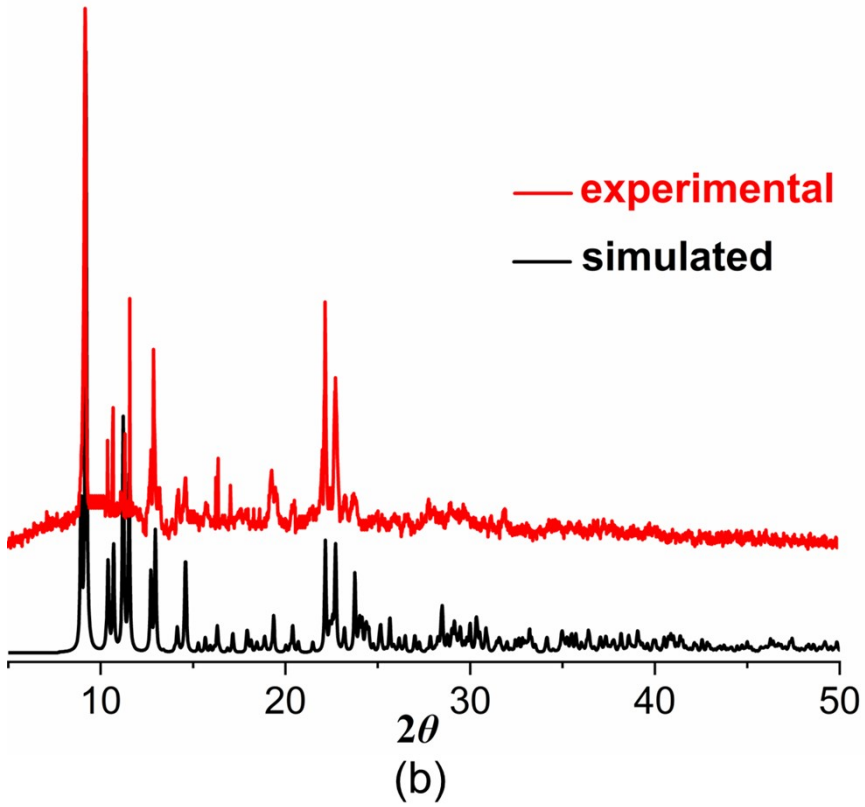

Fig. S5 PXRD patterns for complex **2**.

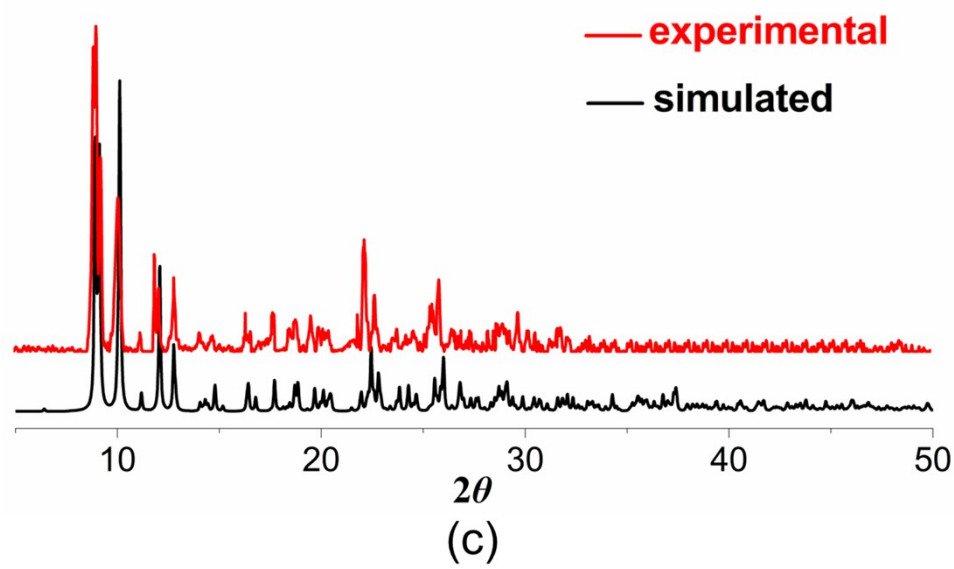

Fig. S6 PXRD patterns for complex **3**.

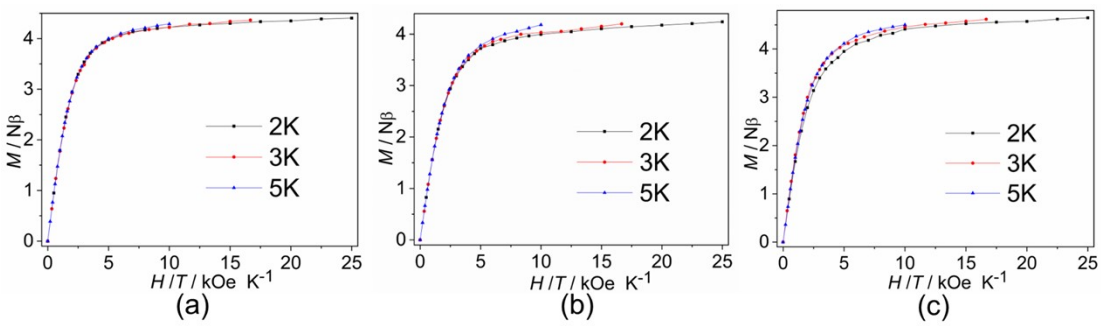

Fig. S7 Plots of *M* vs. *H*/*T* for **1-3** at different temperature.

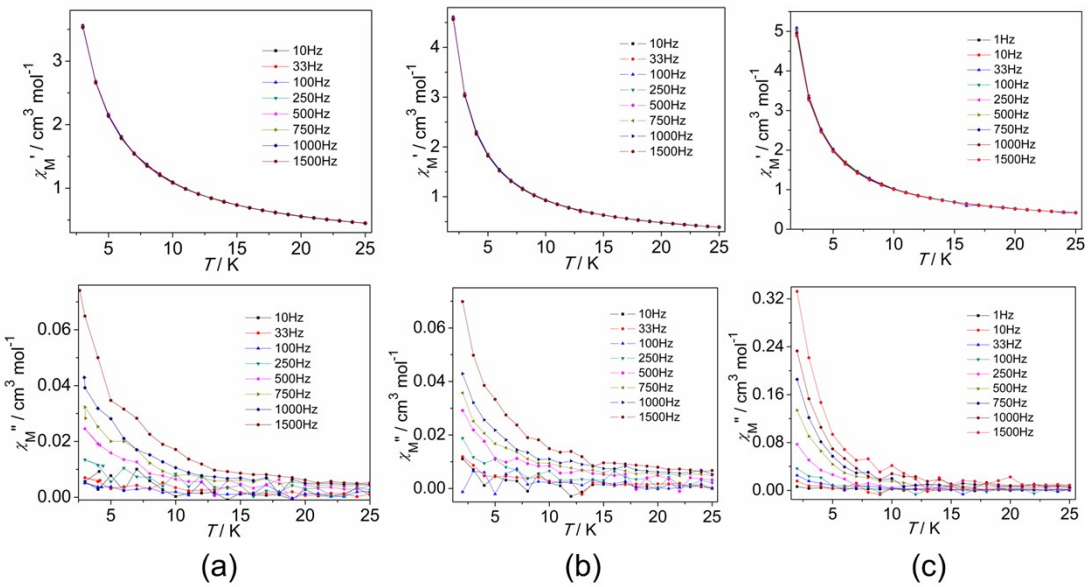

Fig. S8 Temperature dependence of the in-phase (top) and out (bottom) ac susceptibilities at different frequencies without a dc field for complexes **1-3**.

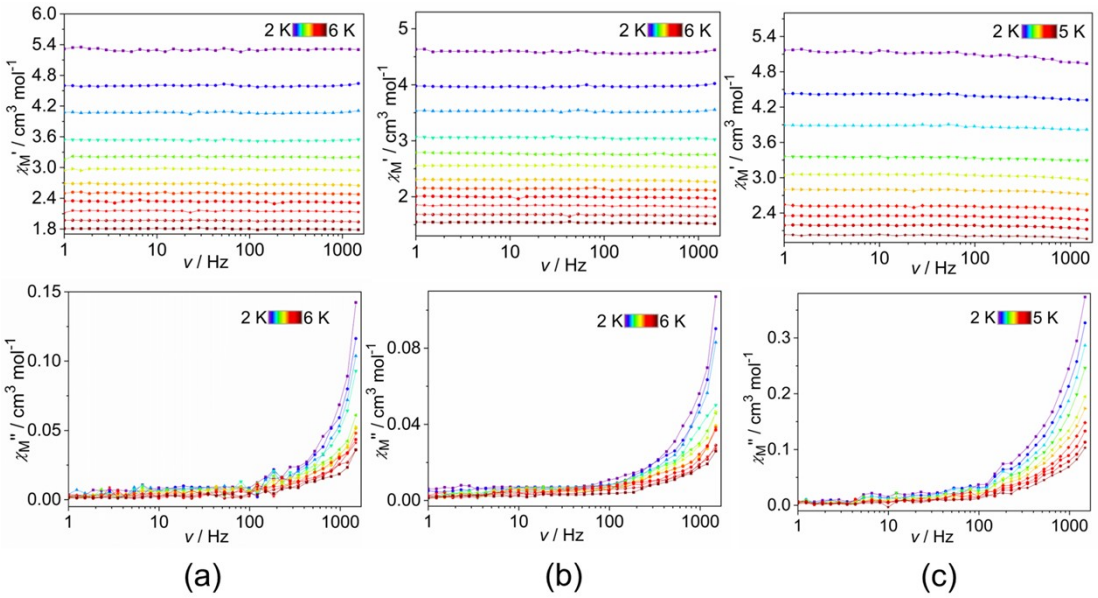

Fig. S9 Frequency dependence of the in-phase (top) and out (bottom) ac susceptibilities at different frequencies without a dc field for complexes **1**-**3**.

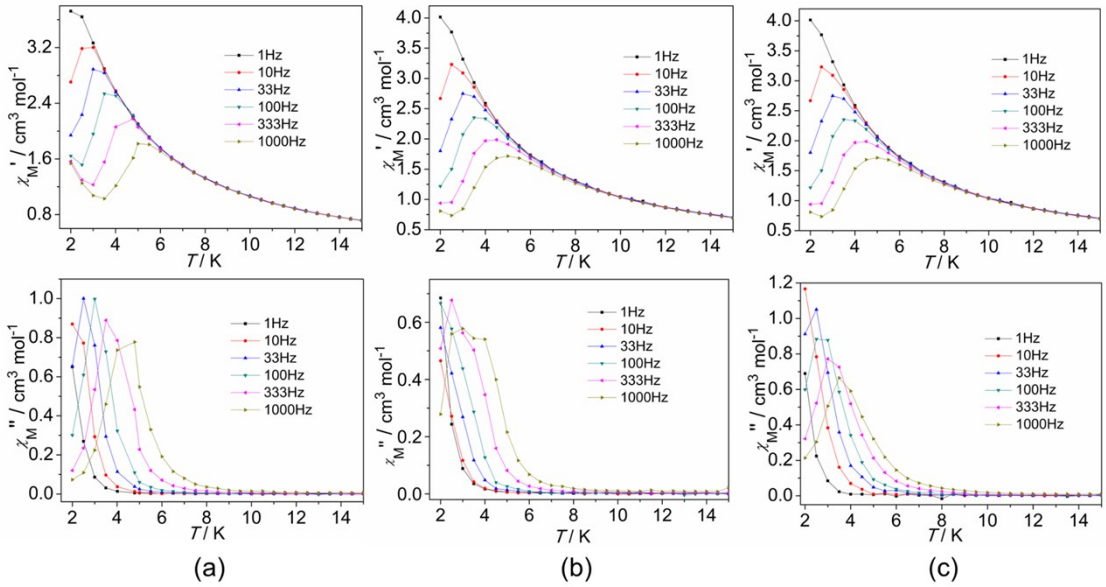

Fig. S10 Temperature dependence of the in-phase (top) and out-of-phase (bottom) ac susceptibilities at different frequencies with a dc field of 500 Oe for complexes **1** (a, a′), **2** (b, b′) and **3** (c, c′).

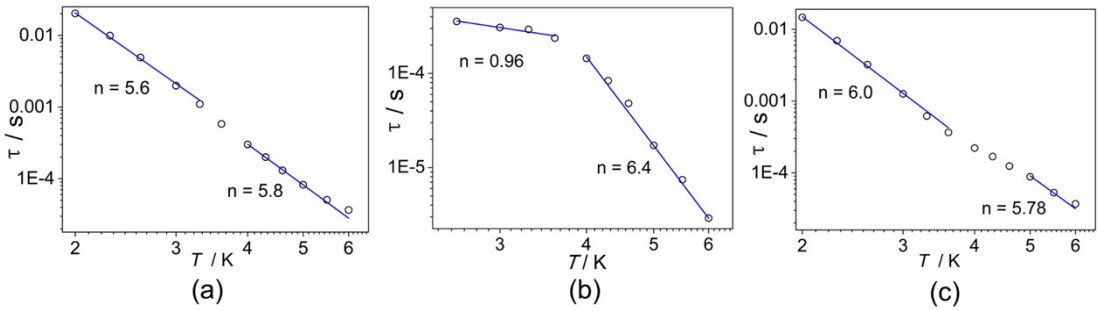

Fig. S11 Temperature dependent relaxation times of **1** (a), **2** (b) and **3** (c) (log-log scale) under a 500Oe dc field. The lines were fitted to the equation of $\tau = T^{-n}$ to give *n* values.

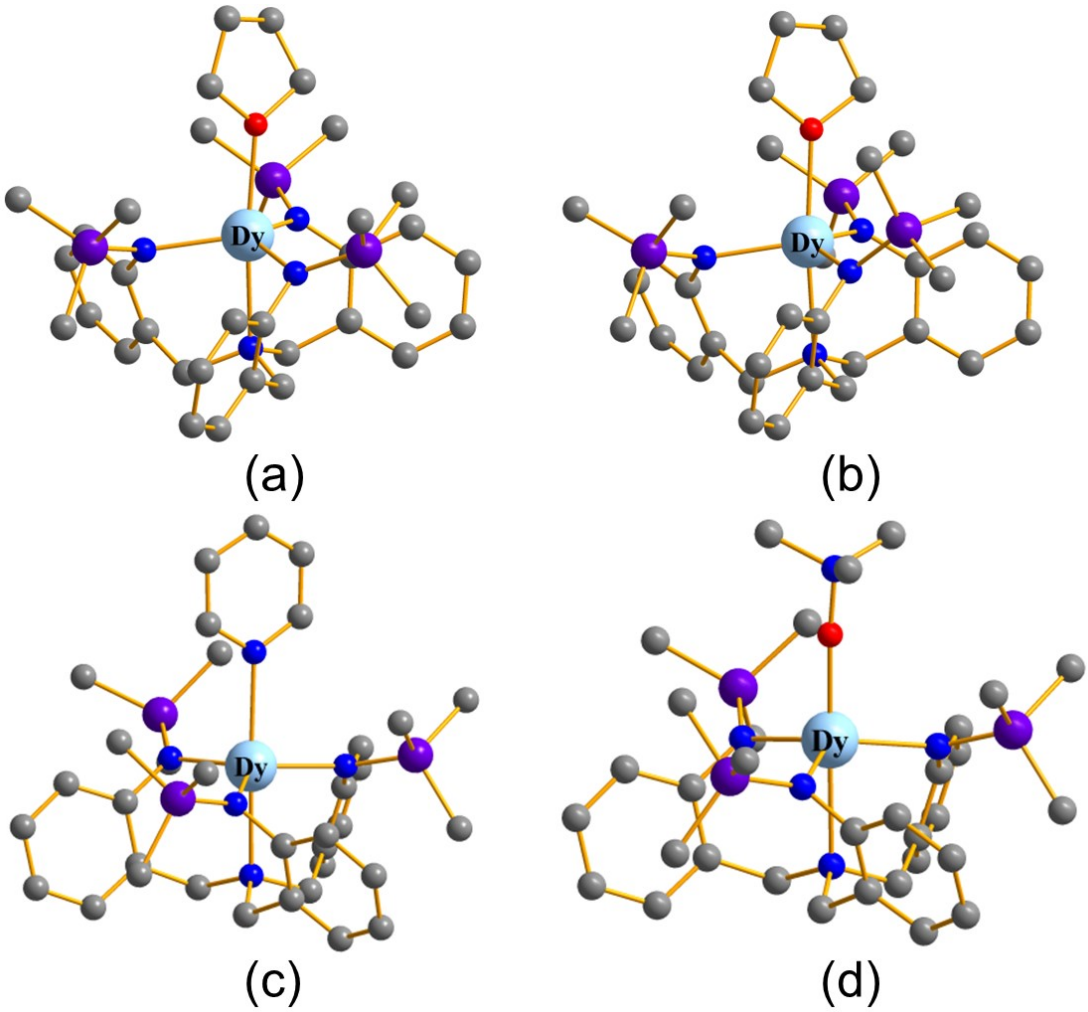

Fig. S12 Calculated molecular structures of complexes **1*a*** (a), **1*b*** (b), **2** (c) and **3** (d); H atoms are omitted for clarify.

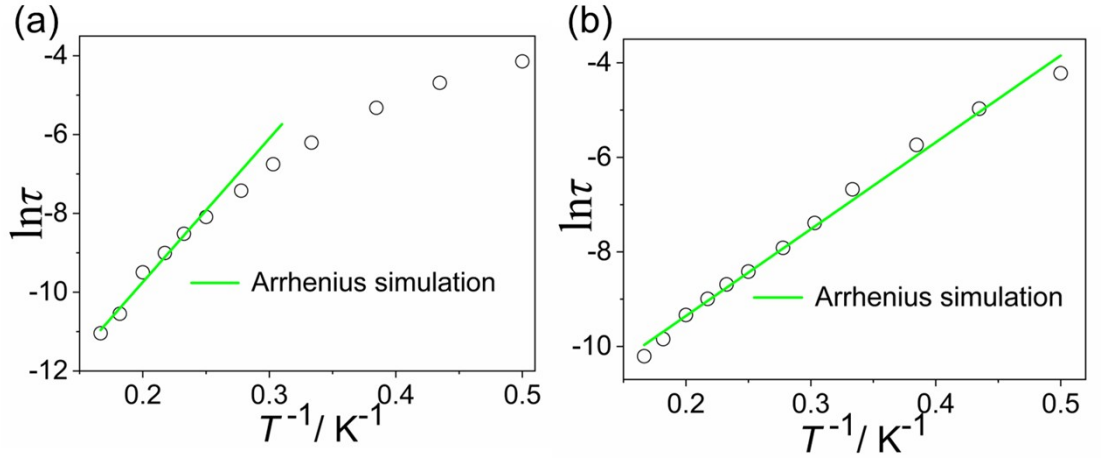

Fig. S13 Magnetization relaxation time, ln τ vs. T^{-1} plot under 500Oe dc field for **1** (a), **3** (b). The green line represents the Arrhenius fit.

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