Supporting Information

Investigation of magneto-structural correlation based on a series of seven-coordinated β-diketone Dy(III) single-ion magnets with C_{2v} and C_{3v} local symmetry

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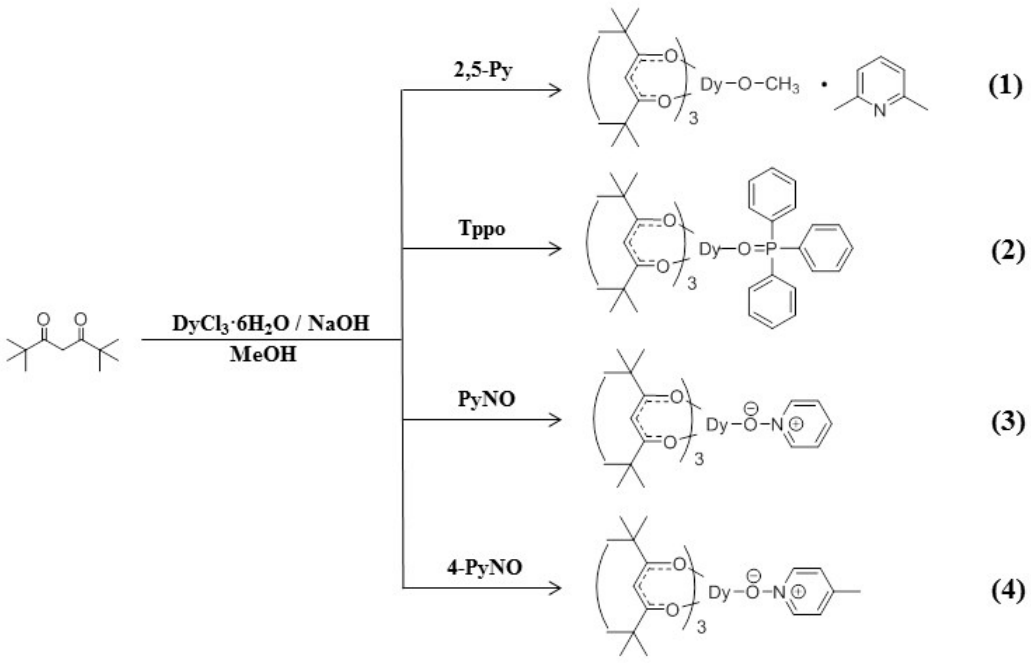
Scheme 1. Synthesis of complexes **1–4.**



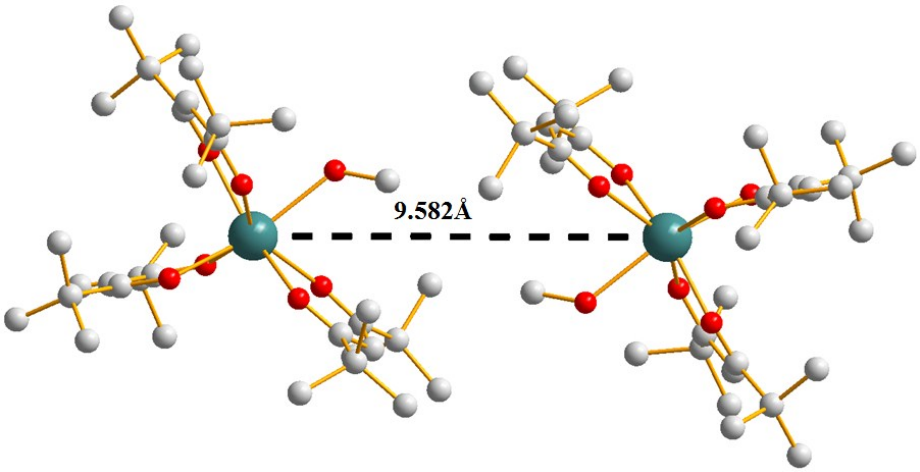


Figure S1. The shortest Dy···Dy distance between the two molecules for complexes **1**.

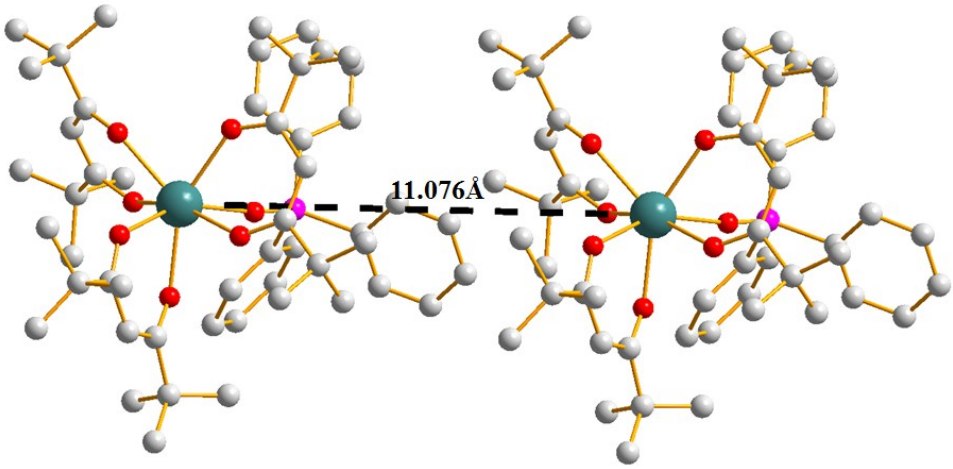


Figure S2. The shortest Dy···Dy distance between the two molecules for complexes **2**.

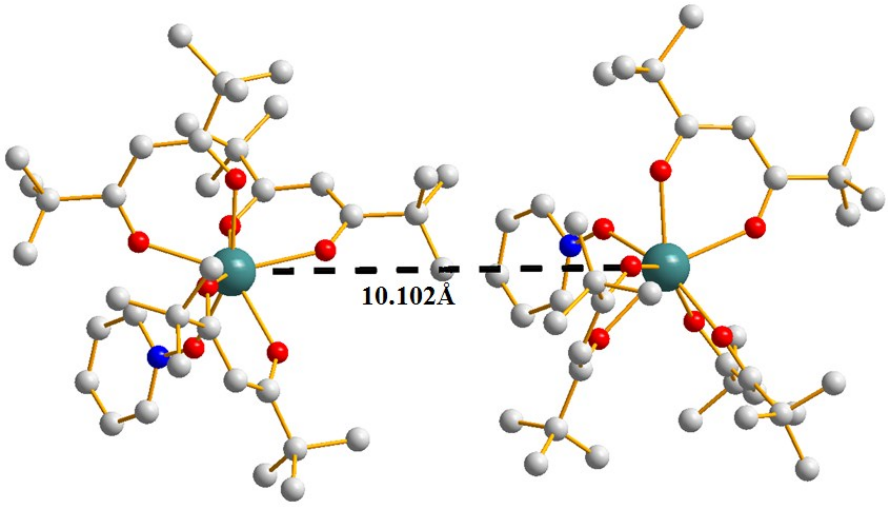


Figure S3. The shortest Dy···Dy distance between the two molecules for complexes **3**.

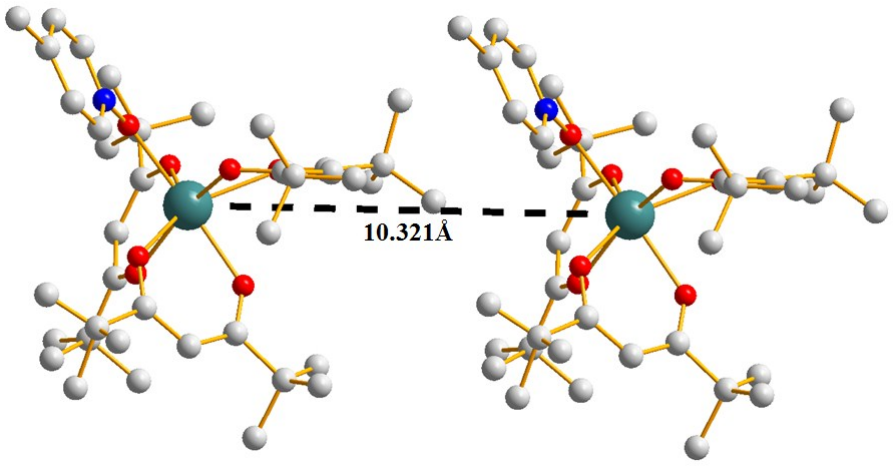


Figure S4. The shortest Dy···Dy distance between the two molecules for complexes **4**.

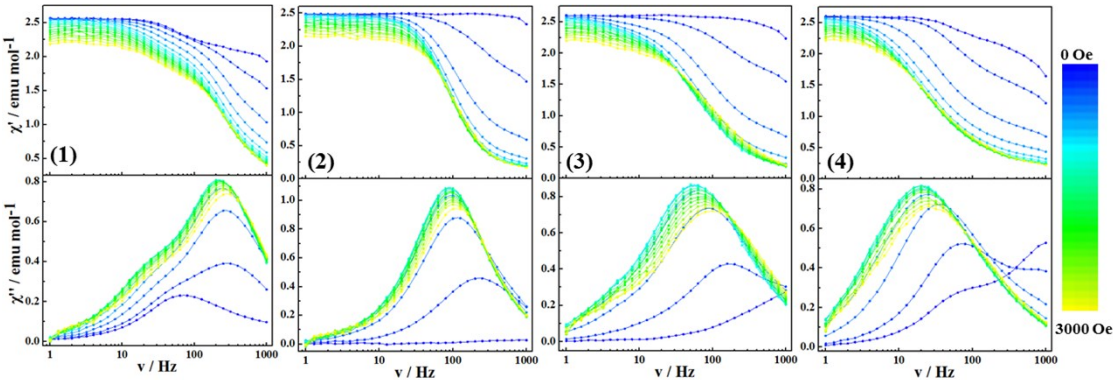


Figure S5. Frequency dependence at 5K of in-phase (χ′) and out-phase (χ″) ac susceptibility component at different dc fields for complexes **1**−**4**.

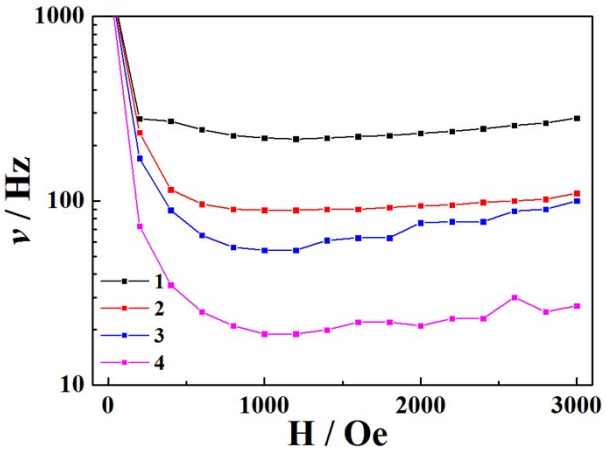


Figure S6 Field dependence of the characteristic frequency as a function of the applied dc field for complexes **1**−**4** at 5K.

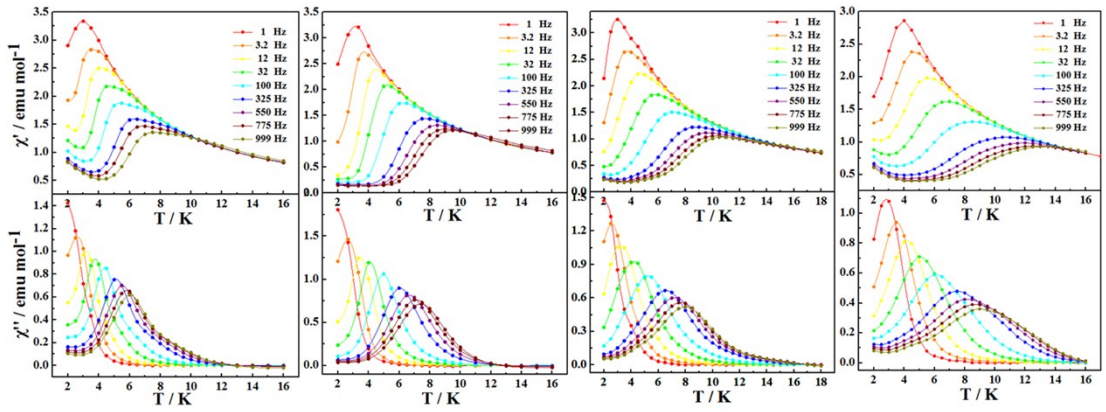


Figure S7. Temperature dependence of the in-phase (χ′) and out-of-phase (χ″) ac susceptibility under 1000 Oe in the frequency range 1—999 Hz for complexes **1**–**4**.

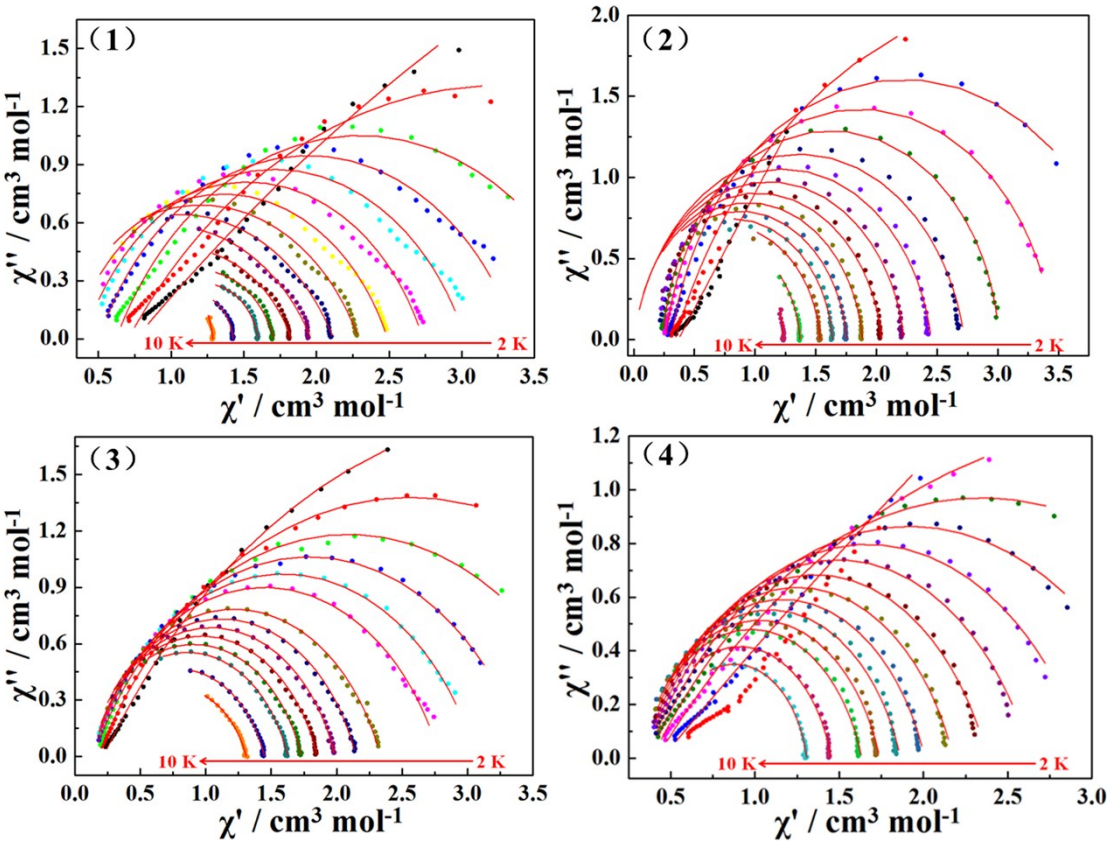


Figure S8. Cole–Cole plots of complexes **1**–**4**. The solid lines represent the fitting using the generalized Debye model.

Table S1. Selected bond lengths (Å) and angles (o) for complexes **1**−**4**.

Complex	**1**	**2**	**3**	**4**
Dy(1)–O(1)	2.289(4)	2.314(3)	2.275(7)	2.281(3)
Dy(1)–O(2)	2.271(4)	2.280(4)	2.286(5)	2.304(4)
Dy(1)–O(3)	2.246(4)	2.281(3)	2.282(6)	2.244(3)
Dy(1)–O(4)	2.277(4)	2.294(4)	2.283(6)	2.323(3)
Dy(1)–O(5)	2.281(4)	2.314(4)	2.303(6)	2.330(4)
Dy(1)–O(6)	2.278(4)	2.277(4)	2.280(5)	2.289(3)
Dy(1)–O(7)	2.406(4)	2.297(4)	2.330(5)	2.317(4)
O(2)-Dy(1)-O(1)	73.85(14)	72.76(13)	76.2(2)	72.21(14)
O(3)-Dy(1)-O(1)	82.61(16)	112.39(13)	79.7(3)	123.90(15)
O(4)-Dy(1)-O(1)	143.23(16)	151.03(14)	147.0(2)	138.65(12)
O(5)-Dy(1)-O(1)	141.34(16)	121.17(15)	80.3(3)	133.95(14)
O(6)-Dy(1)-O(1)	80.68(14)	75.89(14)	134.7(2)	78.78(12)
O(4)-Dy(1)-O(2)	80.37(16)	81.56(15)	80.0(2)	77.70(15)
O(4)-Dy(1)-O(3)	73.46(15)	74.61(13)	74.6(2)	76.02(12)
O(2)-Dy(1)-O(3)	94.11(18)	79.16(13)	82.1(3)	79.52(15)
O(4)-Dy(1)-O(5)	75.42(15)	79.56(15)	117.7(2)	77.17(15)
O(2)-Dy(1)-O(5)	127.76(14)	156.83(14)	80.0(3)	153.39(14)
O(2)-Dy(1)-O(6)	154.34(15)	130.81(14)	130.3(2)	124.70(15)
O(3)-Dy(1)-O(6)	85.61(15)	78.96(14)	132.5(2)	79.28(13)

Table S2 Energy levels and eigenstates for **1** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	26%| ∓ 5/2> + 22%| ± 3/2> + 22%| ∓ 7/2> +18%| ∓ 1/2> +…
102	42%| ∓ 7/2> + 32%| ± 9/2> + 15%| ∓ 5/2> +10%| ∓ 11/2> +…
159	43%| ∓ 9/2> + 19%| ± 5/2> + 17%| ∓ 3/2> +6%| ∓ 11/2> +…
193	49%| ∓ 3/2> + 37%| ± 5/2> + 5%| ∓ 11/2> +…
196	70%| ∓ 1/2> + 12%| ± 11/2> +10%| ± 3/2>+6%| ∓ 9/2> +…
201	54%| ∓ 11/2> + 23%| ± 7/2> + 12%| ± 15/2>+9%| ∓ 1/2> +…
253	87%| ∓ 15/2> + 9%| ± 11/2> + 3%| ±7/2> +…
258	91%| ∓ 13/2> + 7%| ± 9/2> + 2%| ± 5/2> +…

Table S3 Energy levels and eigenstates for **2** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	89%| ∓ 15/2> + 10%| ± 9/2> + 0.31%| ∓ 3/2> +…
148	66%| ∓ 7/2> + 18%| ± 1/2> + 12%| ∓ 13/2> +…
185	56%| ∓ 9/2> + 35%| ± 3/2> + 8%| ∓ 15/2> +…
195	71%| ∓ 5/2> + 9%| ± 11/2> + 7%| ∓ 1/2> +7%| ± 15/2> +…
236	56%| ∓ 13/2> + 32%| ± 11/2> ++7%| ± 1/2>+5%| ∓ 7/2> +…
242	59%| ∓ 11/2> + 21%| ± 13/2> + 16%| ± 5/2>+4%| ∓ 7/2> +…
287	64%| ∓ 3/2> + 34%| ± 9/2> + 2%| ±1 5/2> +…
324	67%| ∓ 1/2> + 19%| ± 7/2> + 10%| ± 5/2>+3.5%| ∓ 13/2> +…

Table S4 Energy levels and eigenstates for **3** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	83%| ∓ 15/2> + 16%| ± 9/2> + 0.11%| ∓ 3/2> +…
96	86%| ∓ 7/2> + 10%| ± 5/2> + 4%| ∓ 13/2> +…
177	80%| ∓ 9/2> + 16%| ± 15/2> + 4%| ∓ 3/2> +…
179	84%| ∓ 5/2> + 9%| ± 7/2> + 6%| ∓ 11/2> +…
278	86%| ∓ 11/2> + 9%| ± 1/2> +5%| ± 5/2> +…
283	95%| ∓ 3/2> + 4%| ± 9/2> +…
345	60%| ∓1/2> + 32%| ± 13/2> + 6%| ±11/2> +…
378	63%| ∓ 13/2> + 30%| ± 1/2> + 5%| ± 7/2>+2%| ∓ 11/2> +…

Table S5 Energy levels and eigenstates for **4** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	89%| ∓ 15/2> + 10%| ± 9/2 +…
27	89%| ∓ 9/2> + 10%| ± 15/2> +…
32	98%| ∓ 9/2> + 1.6%| ± 5/2> +…
90	95%| ∓ 11/2> + 5%| ± 5/2> +…
110	93%| ∓ 5/2> + 5%| ± 11/2> +2%| ± 7/2> +…
156	99.7%| ∓ 13/2> +…
203	99%| ∓3/2> +…
266	99%| ∓ 1/2> +…

Table S6 Generalised Debye model fitting parameters for complex **1**.

T (K)	χ_S	χ_T	τ (s)	α
3	0.62237	3.97285	0.03418	0.28690
3.5	0.50513	3.38384	0.01075	0.25913
4	0.39965	3.02369	0.00396	0.25089
4.5	0.31234	2.73276	0.00164	0.24909
5	0.22598	2.4895	7.4229E-4	0.25661
5.5	0.11865	2.28939	3.5318E-4	0.27785
6	0.00907	2.11705	1.7308E-4	0.30288
6.5	0.01675	1.96338	9.9217E-5	0.3134
7	0.36674	1.82693	9.4206E-5	0.28031
7.5	0.71734	1.70528	1.1415E-4	0.21243
8	0.89218	1.59991	1.2517E-4	0.1423
9	0.91856	1.42579	8.7255E-5	0.07099
10	0.02860	1.28748	1.2706E-5	0.09559

Table S7 Generalised Debye model fitting parameters for complex **2**.

T (K)	χ_S	χ_T	τ (s)	α
3	0.30954	4.14276	0.05755	0.1124
3.5	0.26782	3.46277	0.01836	0.07402
4	0.2419	3.00816	0.00721	0.04488
4.5	0.22061	2.67943	0.00338	0.02964
5	0.20673	2.42101	0.00178	0.02122
5.5	0.19742	2.20565	0.00102	0.01335
6	0.19591	2.02646	6.26205E-4	0.00827
6.5	0.20337	1.87358	4.07185E-4	0.00163
7	0.21744	1.74532	2.76176E-4	2.42476E-11
7.5	0.23722	1.63214	1.93096E-4	3.64615E-11
8	0.26099	1.53128	1.37664E-4	5.60551E-11
9	0.30803	1.36658	7.15911E-5	5.82728E-11
10	1.46302E-6	1.23177	2.59181E-5	2.62848E-10

Table S8 Generalised Debye model fitting parameters for complex **3**.

T (K)	χ_S	χ_T	τ (s)	α
3	0.17306	4.94639	0.10012	0.33332
3.5	0.13561	4.04617	0.03716	0.30794
4	0.12631	3.454	0.01641	0.27668
4.5	0.11862	3.06511	0.00849	0.25800
5	0.11341	2.7847	0.00481	0.24515
5.5	0.1001	2.33309	0.00174	0.22039
6	0.09506	2.14676	0.0011	0.21052
6.5	0.0949	1.98574	7.26065E-4	0.20067
7	0.08612	1.84979	4.88418E-4	0.19989
7.5	0.08782	1.72907	3.37745E-4	0.19914
8	0.0964	1.62515	2.39603E-4	0.2007
9	0.14591	1.44799	1.27484E-4	0.21029
10	0.25412	1.30966	7.60816E-5	0.22974

Table S9 Generalised Debye model fitting parameters for complex **4**.

T (K)	χ_S	χ_T	τ (s)	α
3	0.41424	6.19965	0.49186	0.48694
3.5	0.3875	4.30034	0.09248	0.41417
4	0.37441	3.46303	0.03098	0.35082
4.5	0.36228	2.9773	0.01397	0.30329
5	0.35111	2.6406	0.00738	0.27021
5.5	0.34032	2.3794	0.00429	0.24779
6	0.32804	2.17784	0.00266	0.23229
6.5	0.31599	2.00776	0.00174	0.22258
7	0.3154	1.86367	0.0012	0.21064
7.5	0.31376	1.7405	8.48537E-4	0.20432
8	0.32142	1.6329	6.24634E-4	0.19501
9	0.37121	1.45107	3.77552E-4	0.16337
10	0.44895	1.30639	2.56249E-4	0.12545

Table S10 The curves are fitted by the modified Arrhenius relationship the Raman process is taken account $\tau^{-1} = CT^n + \tau_0^{-1} \exp(-U_{eff}/k_B T)$.

complex	C($s^{-1}\cdot K^{-n}$)	n	$\tau 0$ (s)	U_{eff} (K)
1	0.012	5.36	2.76×10^{-6}	26.6
2	0.011	5.82	1.12×10^{-6}	35.9
3	0.087	5.13	5.53×10^{-6}	42.7
4	0.024	5.16	1.58×10^{-6}	55.8