Supplementary Information (SI) for Dalton Transactions.

Supplementary Information for:

A Dysprosium Alkylidene Single Molecule Magnet

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Contents

Experimental details

General. All operations were carried out under an atmosphere of argon using Schlenk techniques or in an argon-filled glovebox. Toluene, THF, hexane, and C_6D_6 were dried over Na/K alloy, transferred under vacuum, and stored in the glovebox. Anhydrous $DyCl_3$ and Li^{n}Bu were commercially available and used without further purification. The raw materials LiL(L= $[MeC(NDIPP)CHC(Me)(NCH_2CH_2N(Me)_2)]^-$, DIPP = 2,6-(^{i}Pr)$_2C_6H_3$)[1] and $CH_2(SiMe_3)PPh_2Se$[2] were synthesized following the literature. These compounds were stored in the glovebox. ^1H and $^{31}P\{^1H\}$ NMR spectra were recorded on a Bruker Avance III HD 500 MHz spectrometer. Chemical shifts were reported in δ units with references to the residual solvent resonance of the deuterated solvents for proton chemical shifts, and to external H_3PO_4 (85%) for phosphorus chemical shifts. EA values were obtained from Elementar UNICUBE.

Li{CH(SiMe$_3$)PPh$_2$Se}(THF) was synthesized by slowly adding Li^{n}Bu (2.5 M in hexane, 1.42 mmol, 0.57 mL) to a solution of $CH_2(SiMe_3)PPh_2Se$ (500 mg, 1.42 mmol) in THF (ca. 10 mL) under -78 °C. After stirring for 1.5 h at room temperature, the $LiCH(SiMe_3)PPh_2Se(THF)$ was isolated by removing the volatiles under vacuum and repeatedly washed with hexane. The white solid of $Li\{CH(SiMe_3)PPh_2Se\}(THF)$ (400 mg, 0.93 mmol, 65% yield) was dried in vacuum. ^1H NMR (500 MHz, C_6D_6, 25 °C): δ = 8.19 (*m*, 4H, *o*-Ph*H* of PPh_2), 7.16-7.03 (*m*, *p*-Ph*H* and *m*-Ph*H* of PPh_2, overlapped with the residual solvent resonance of the deuterated solvent), 3.56 (*m*, 4H, THF-*H*), 1.22 (*m*, 4H, THF-*H*), 0.57 (*d*, $^2J_{P\text{-}H}$=10.0 Hz, 1H, PC*H*Si), 0.06ppm (*s*, 9H, Si*Me$_3$*); $^{31}P\{^1H\}$ NMR (200 MHz, C_6D_6, 25 °C) : δ=29.1 (*t*, $^2J_{P\text{-}Se}$= 230 Hz).

[DyLCl$_2$(THF)](1): To a mixture of anhydrous $DyCl_3$ (400 mg, 1.49 mmol) and LiL (500 mg, 1.49 mmol), 10 mL of THF was added. After stirring at room temperature for 6 hours, the volatiles were removed under vacuum, then the residue was extracted with toluene (15 mL). the complex **1** was isolated by removing the volatiles under vacuum and repeatedly washed with hexane(3×3 mL). 775 mg powder of **1** was obtained after drying under vacuum. **Crystallization**: These solids were solved in a mixture of toluene(4 mL) and THF(1 mL). The solution was concentrated to 2 mL at room temperature under reduced pressure. After standing at room temperature for one day, yellow crystals of **1** (400 mg, 0.63 mmol, 42% yield) were obtained. Anal. Calcd. for $C_{25}H_{42}N_3DyCl_2O$: C 47.36; H 6.68; N 6.63. Found: C 46.64; H 6.55; N 6.31.

[DyL{CH(SiMe$_3$)PPh$_2$Se}Cl](2): Complex **1** (127 mg, 0.20 mmol) and $Li\{CH(SiMe_3)PPh_2Se\}(THF)$(87 mg, 0.20 mmol) were mixed in toluene (4 mL). After stirring at room temperature for 1.5 h, the toluene solution was separated by centrifugation. The complex **2** was isolated by removing the volatiles under vacuum and repeatedly washed with hexane(3×3 mL). 123 mg powder of **2** was obtained after drying under vacuum. **Crystallization**: These solids were solved in hot toluene (0.5 mL) heated by hairdryer. After standing at room temperature overnight, yellow crystals of **2** (68 mg, 0.08 mmol, 39% yield) were obtained. Anal. Calcd. for $C_{37}H_{54}N_3DySiPSeCl$: C 50.68; H 6.21; N 4.79. Found: C 51.07; 6.30; N 4.72.

[DyL{C(SiMe$_3$)PPh$_2$Se}](3): MeLi (1.6 M in diethoxymethane, 0.07 mL, 0.11 mmol) was added to a solution of **2** (95 mg, 0.11 mmol) in toluene (3 mL) at room temperature. After stirring at 50^{o}C for 2 hours, the toluene solution was separated by centrifugation. The complex **2** was isolated by removing the volatiles under vacuum and repeatedly washed with hexane(3×3 mL). 86 mg powder of **2** was obtained after drying under vacuum. **Crystallization**: These solids were solved in toluene(1 mL) and concentrated to 0.5 mL at room temperature. After standing at room temperature overnight, yellow crystals of **3** (63 mg, 0.07 mmol, 68% yield) were obtained. Anal. Calcd. for $C_{37}H_{53}N_3DySiPSe$：C 52.88；H 6.36；N 5.00. Found：C 53.16；H 6.65；N 4.54.

X-ray Crystallography

The single crystal of **1-3** covered in grease was mounted under nitrogen atmosphere on a glass fiber. Data collections of **1** and **2** were performed at 200 K on Bruker APEX-II CCD diffractometer with Ga *Kα* radiation (λ = 1.34138 Å). Data collections of **3** were performed at 293 K on XtaLAB AFC12 (RINC): Kappa single diffractometer with Mo *Kα* radiation (λ = 0.71073 Å). Using Olex2, the structures were solved with the ShelXS or ShelXT structure solution program using Direct Methods or Patterson Method and refined with the ShelXL refinement package using Least Squares minimisation[3-5]. Hydrogen atoms were placed at calculated positions and were included in the structure calculation. The solvent in complexs **2** and **3** were masked using solvent mask function in Olex2. Crystallographic data and refinement for **1–3** are listed in Table S1.

Table S1. Crystallographic Data and Refinement parameters.

	1	**2**	**3**
Formula	$C_{25}H_{42}Cl_2DyN_3O$	$C_{37}H_{54}ClDyN_3PSeSi$	$C_{37}H_{53}DyN_3PSeSi$
Formula weight	634.01	876.80	840.34
color	yellow	yellow	yellow
Temperature/K	200.00	200.00	293.00
Wavelength(Å)	1.34138	1.34138	0.71073
Crystal system	triclinic	triclinic	monoclinic
Space group	P-1	P-1	$P2_1/n$
a/Å	10.3923(4)	12.0201(3)	12.6239(2)
b/Å	10.9758(4)	12.9019(4)	11.4942(2)
c/Å	13.5542(5)	16.9940(5)	31.4518(5)
α/°	75.400(2)	108.3600(10)	90
β/°	73.024(2)	95.0090(10)	94.681(2)
γ/°	75.3690(10)	110.0140(10)	90
V/Å^3	1403.92(9)	2293.31(12)	4548.48(13)
Z	2	2	4
D_{calcd} (g/cm^3)	1.500	1.270	1.227
F(000)	642.0	886.0	1700.0
θ range (°)	3.022 to 54.998	3.489 to 72.399	2.400 to 29.869
no. of refns collected	28925	90474	126995
no. of unique refns	5300	13717	12331
no. of obsd refns ($I > 2\sigma$(I))	4876	13433	10048
No. of params.	298	417	419
Final R, R_w ($I > 2\sigma$(I))	0.0430, 0.1142	0.0353, 0.0990	0.0364, 0.0677
Goodness-of-fit on F^2	1.080	1.034	1.042
$\Delta\rho_{\text{max, min}}$, eÅ^{-3}	1.701/-1.870	1.425/-1.898	0.976/-0.930

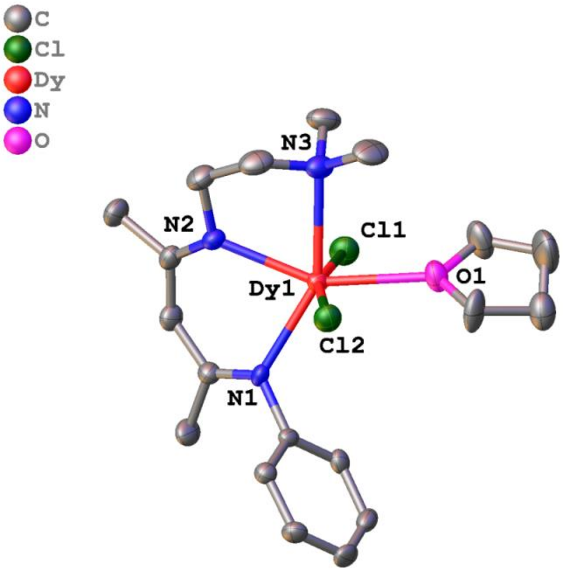

Figure S1 Molecular structure of **1** with thermal ellipsoids at 30% probability. Isopropyl groups and all hydrogen atoms are omitted for clarity. Selected bond lengths (Å) and angles (deg): Dy1−N1 2.354(4), Dy1−N2 2.324(4), Dy1−N3 2.535(4), Dy1−Cl1 2.5864(13), Dy1−Cl2 2.5925(13),Dy1−O1 2.475(4), Cl1−Dy1−Cl2 162.92(5), O1−Dy1−Cl1 81.50(9), O1−Dy1−Cl2 81.74(9), O1−Dy1−N3 84.95(15), N1−Dy1−Cl1 93.25(10), N1−Dy1−Cl2 93.47(11), N1−Dy1−O1 124.32(13), N1−Dy1−N3 150.47(15), N2−Dy1−Cl1 94.19(11), N2−Dy1−Cl2 102.41(11), N2−Dy1−O1 154.88(14), N2−Dy1−N1 80.48(14), N2−Dy1−N3 70.74(15), N3−Dy1−Cl1 95.27(12), N3−Dy1−Cl2 86.52(11).

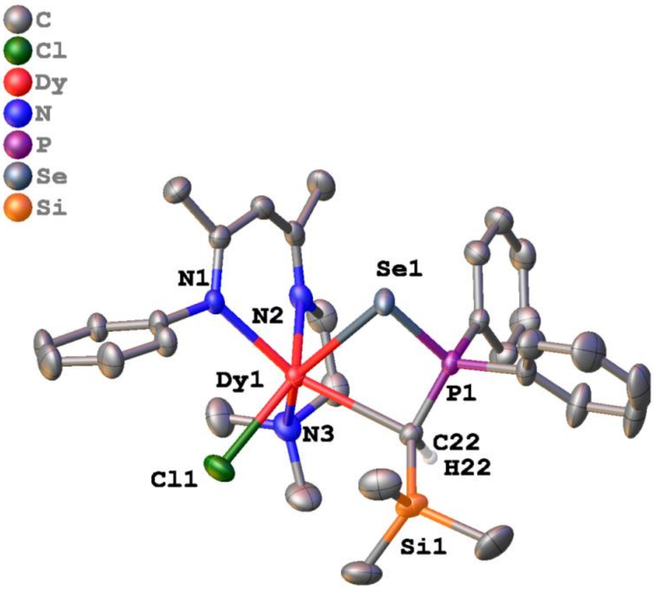

Figure S2 Molecular structure of **2** with thermal ellipsoids at 30% probability. Isopropyl groups and all hydrogen atoms(except for H22) are omitted for clarity. Selected bond lengths (Å) and angles (deg): Dy1−N1 2.390(2), Dy1−N2 2.314(2), Dy1−N3 2.518(2), Dy1−C22 2.665(2), Dy1−Cl1 2.5667(7), Dy1−Se1 2.9366(3), Se1−P1 2.1586(6), P1−C22 1.744(3), Si1−C22 1.870(3), Cl1−Dy1−Se1 126.46(2), Cl1−Dy1−C22 101.27(6), N2−Dy1−Se1 91.42(6), N2−Dy1−Cl1 138.81(6), N2−Dy1−N1 80.04(8), N2−Dy1−C22 108.06(8), N2−Dy1−N3 68.98(9), N1−Dy1−Se1 84.67(5), N1−Dy1−Cl1 87.36(5), N1−Dy1−C22 152.22(7), N1−Dy1−N3 121.19(8), C22−Dy1−Se1 68.89(5), N3−Dy1−Se1 141.96(6), N3−Dy1−Cl1 85.20(6), N3−Dy1−C22 86.08(8), Dy1−C22−H22 109.7, P1−C22−Dy1 93.63(10), P1−C22−Si1 119.66(14), P1−C22−H22 109.7, Si1−C22−Dy1 113.25(12), Si1−C22−H22 109.7.

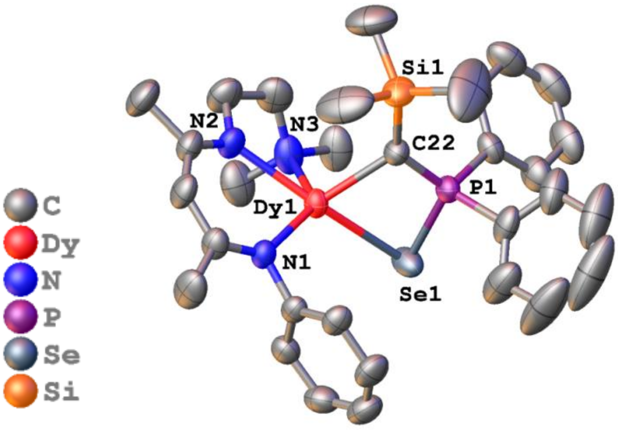

Figure S3 Molecular structure of **3** with thermal ellipsoids at 30% probability. Isopropyl groups and all hydrogen atoms are omitted for clarity. Selected bond lengths (Å) and angles (deg): Dy1−Se1 2.8058(3), Dy1−N1 2.319(2), Dy1−N2 2.383(2), Dy1−N3 2.506(2), Dy1−C22 2.266(3), Se1−P1 2.2265(7), P1−C22 1.662(3),N1−Dy1−Se1 118.42(5), N1−Dy1−N2 77.08(7), N1−Dy1−N3 134.16(8), N2−Dy1−Se1 159.26(5), N2−Dy1−N3 70.49(8), N3−Dy1−Se1 88.89(6), C22−Dy1−Se1 77.83(6), C22−Dy1−N1 117.60(8), C22−Dy1−N2 108.29(9), C22−Dy1−N3 103.05(9),P1−C22−Dy1 99.03(12), P1−C22−Si1 133.82(16), Si1−C22−Dy1 127.04(13).

Table S2 Selected bond distances [Å] and angles [deg] of **1-3** for compare

bond lengths(Å) or angles(deg)	**1**	**2**	**3**
Dy1−N1	2.354(4)	2.390(2)	2.319(2)
Dy1−N2	2.324(4)	2.314(2)	2.383(2)
Dy1−N3	2.535(4)	2.518(2)	2.506(2)
Dy1−Cl1	2.5864(13)	2.5667(7)	-
Dy1−Cl2	2.5925(13)	-	-
Dy1−O1	2.475(4)	-	-
Dy1−C22	-	2.665(2)	2.266(3)
Dy1−Se1	-	2.9366(3)	2.8058(3)
Se1−P1	-	2.1586(6)	2.2265(7)
P1−C22	-	1.744(3)	1.662(3)
Si1−C22	-	1.870(3)	1.809(3)
N1−Dy1−N2	80.48(14)	80.04(8)	77.08(7)
N2−Dy1−N3 (deg)	70.74(15)	68.98(9)	70.49(8)
N1−Dy1−N3	150.47(15)	121.19(8)	134.16(8)
P1−C22−Dy1	-	93.63(10) .	99.03(12)
P1−C22−Si1	-	119.66(14)	133.82(16)
Si1−C22−Dy1	-	113.25(12)	127.04(13)
Latest distance of Dy1⋯Dy1	7.548	8.330	9.178

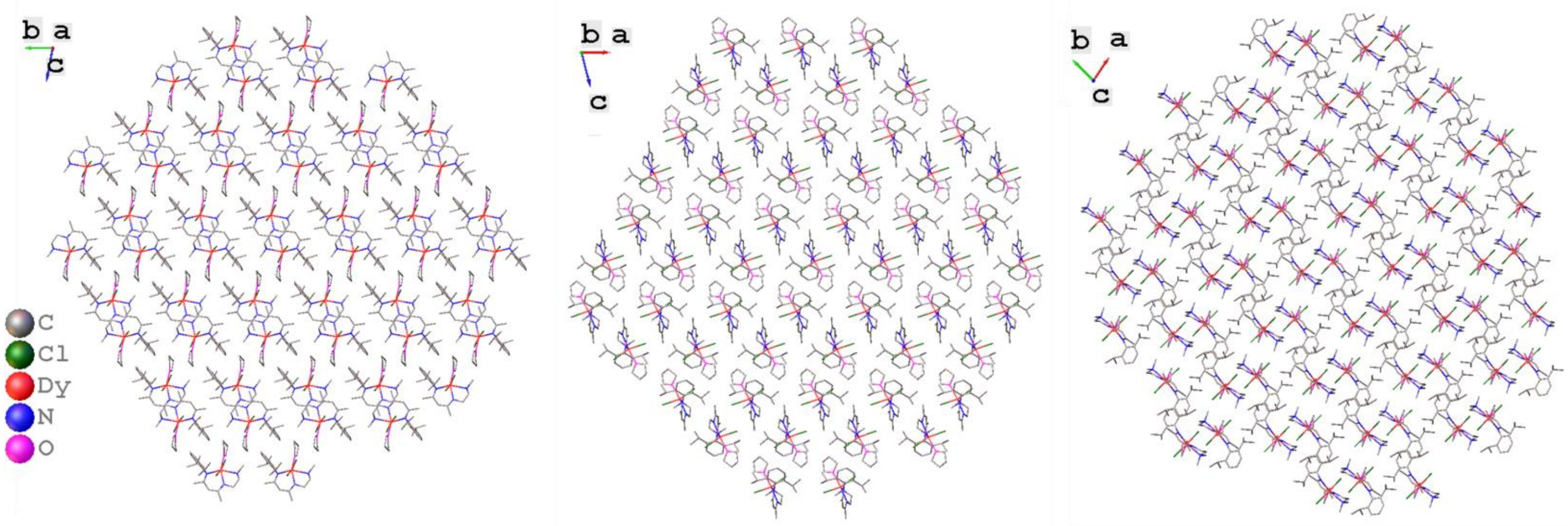

Figure S4 Packing diagrams for **1** viewed along the corresponding crystalline axis. H atoms are omitted for clarity.

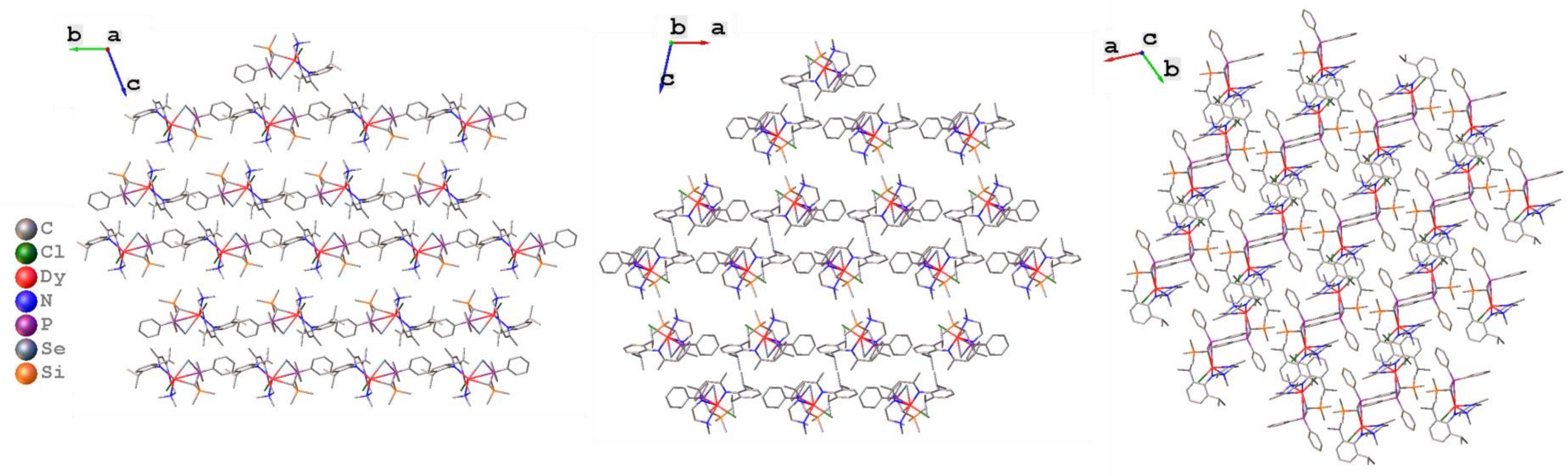

Figure S5 Packing diagrams for **2** viewed along the corresponding crystalline axis. H atoms are omitted for clarity.

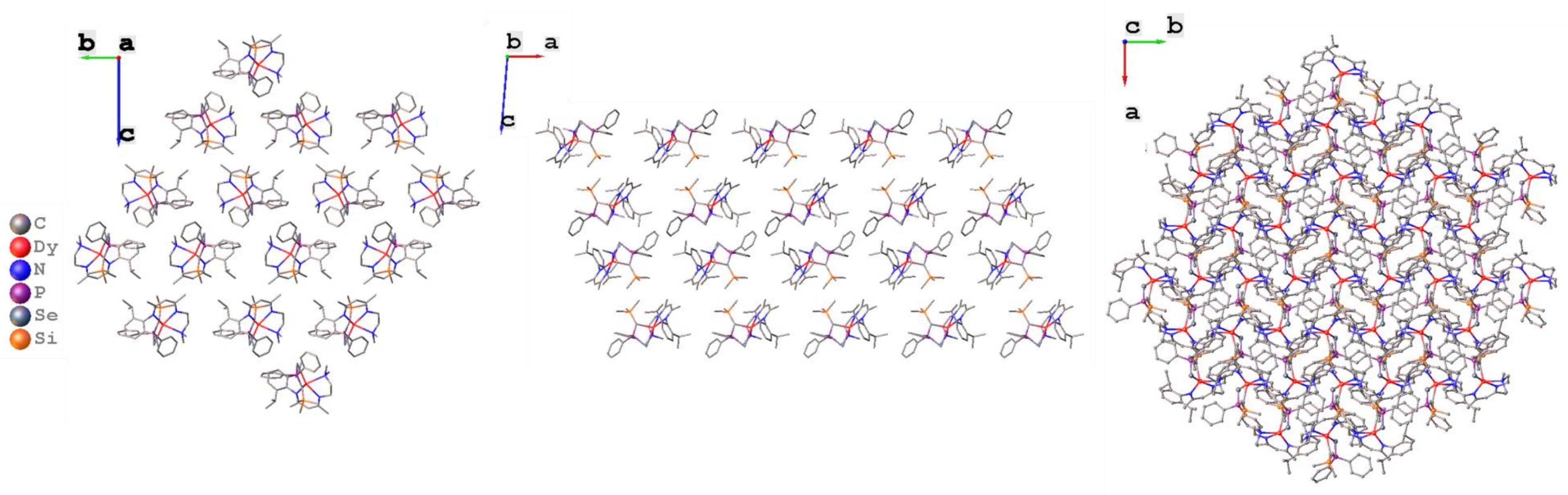

Figure S6 Packing diagrams for **3** viewed along the corresponding crystalline axis. H atoms are omitted for clarity.

Magnetic Characterization

The samples were stuffed in capsules with parafilm covered to protect them from air and water. Direct current (dc) magnetism and alternating current (ac) magnetic measurement of poly crystals were performed on Quantum Design MPMS3 magnetometer. Capsule, parafilm and molecule formula (Pascal constant)[6] were considered when the diamagnetic correction was carried on the data.

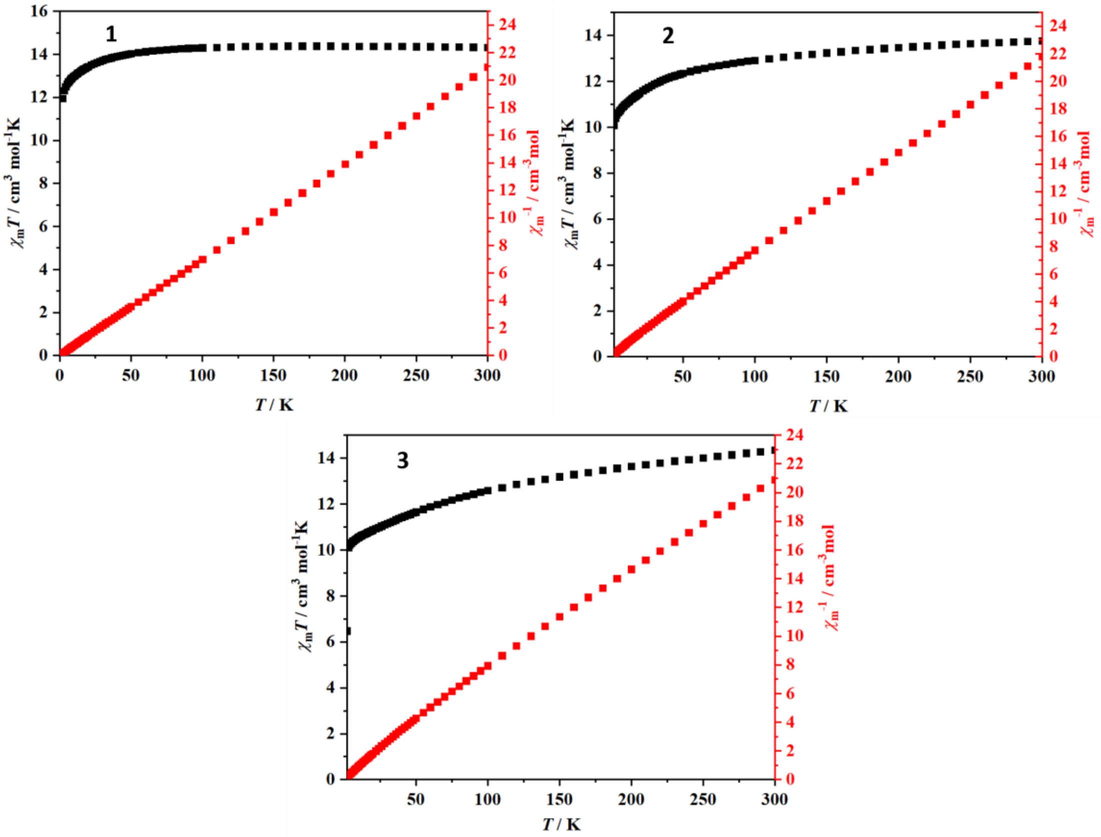

Figure S7 Temperature dependence of χ_mT and χ_m^{-1} for **1-3**.

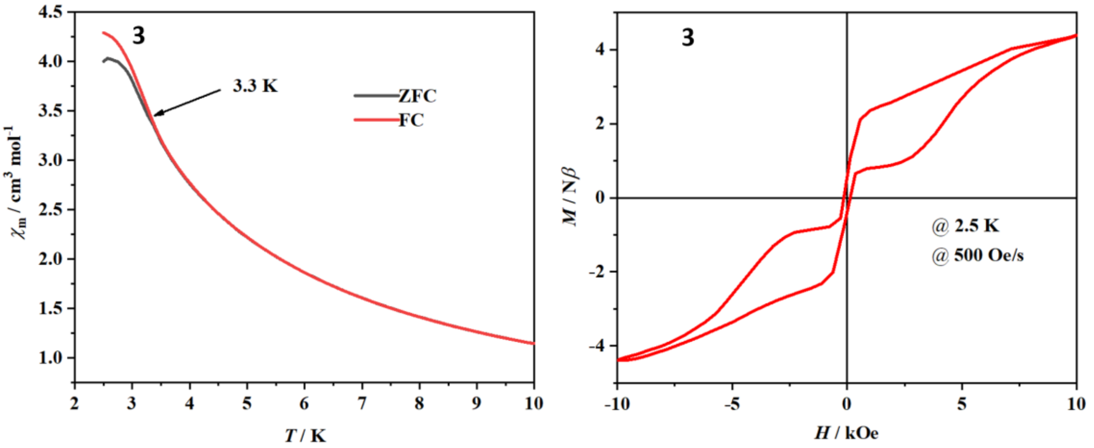

Figure S8 Left, the ZFC/FC magnetic susceptibilities under 1 kOe dc field were measured for **3**. Right, hysteresis loops measured at 2.5 K with a sweep rate of 500 Oes^{-1} for **3**.

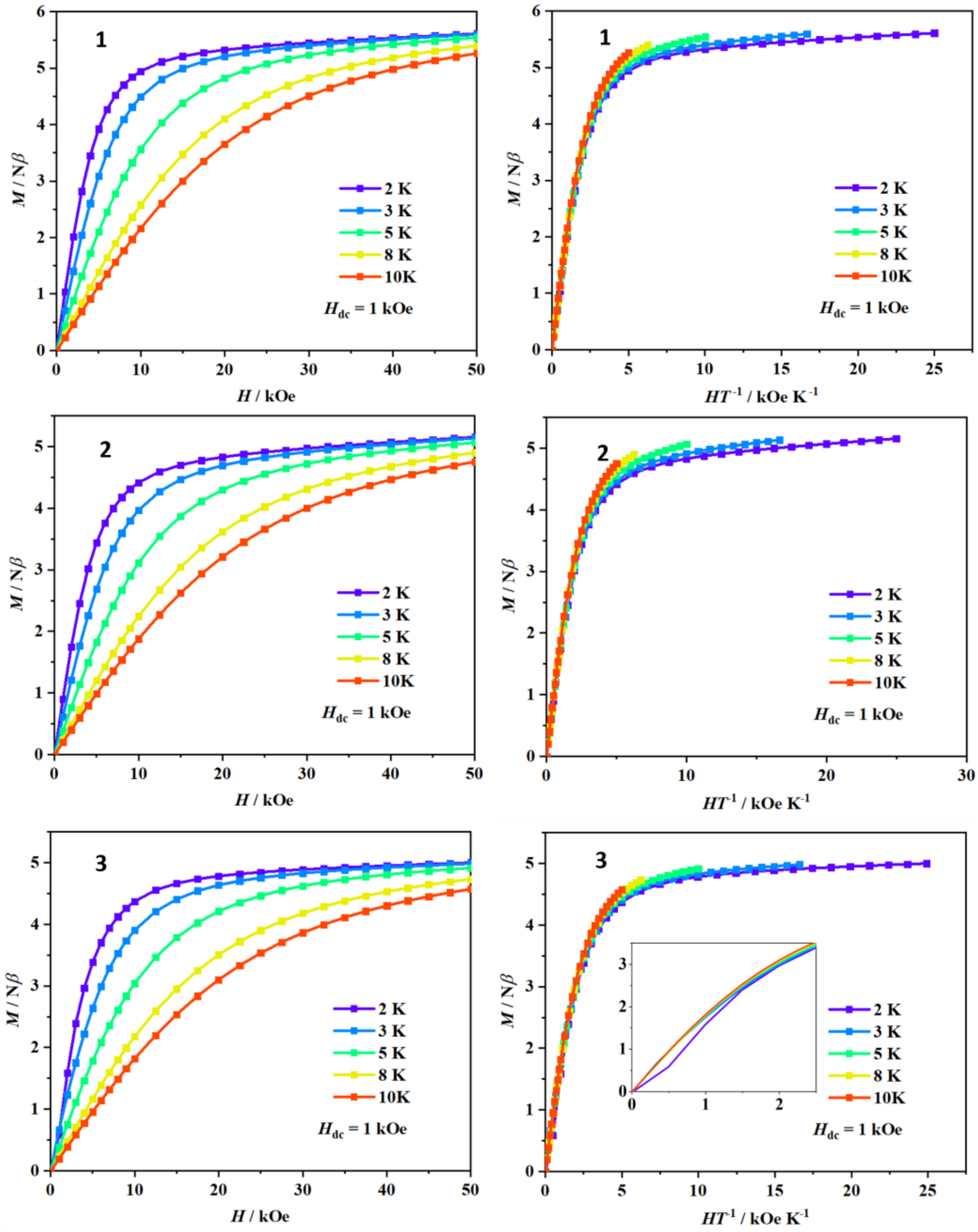

Figure S9 Left, M vs H plots at different temperature for **1-3**. Right, M vs HT^{-1} plots at different temperature for **1-3**. The partial enlarged drawing was inserted in M vs HT^{-1} plot of **3**.

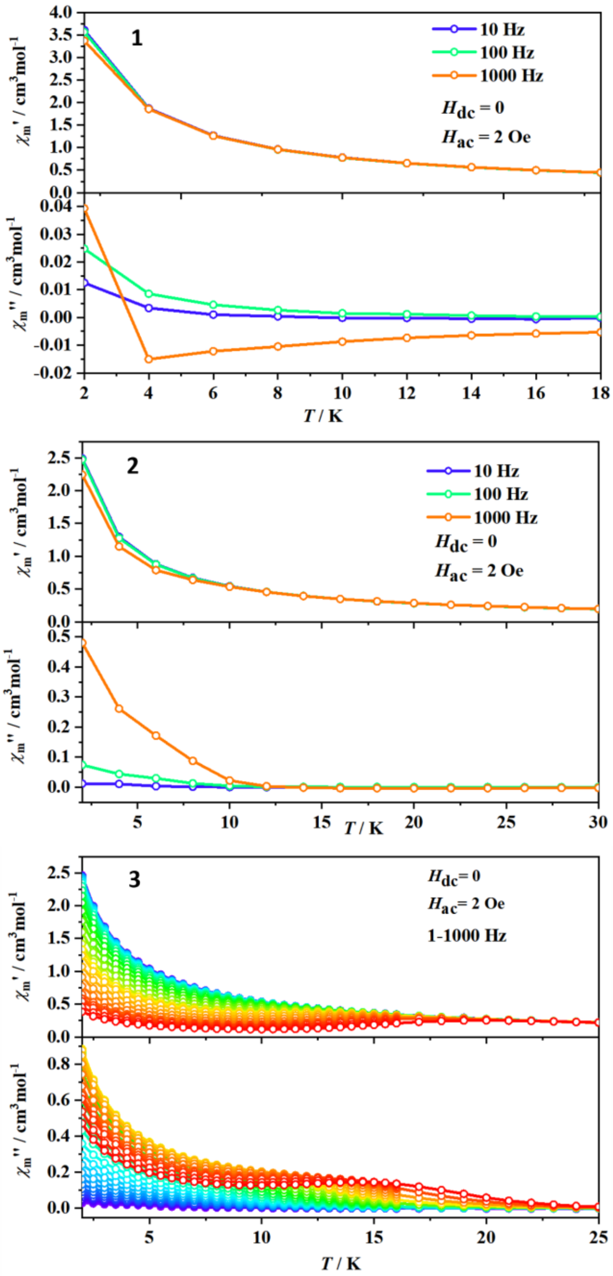

Figure S10 Temperature dependence of the alternating-current molar magnetic susceptibilities for **1-3** in zero dc field.

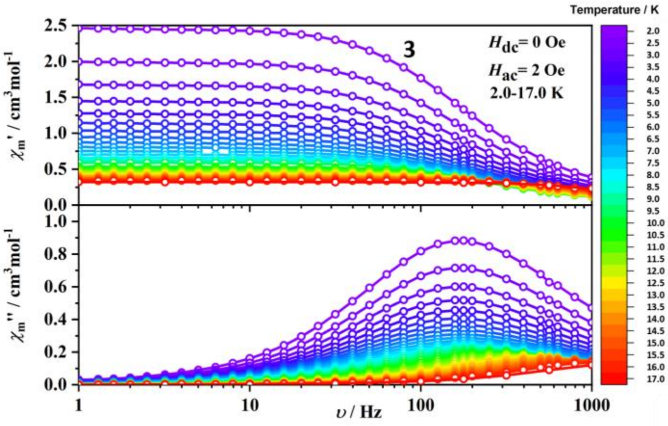

Figure S11 frequency dependence of the alternating-current molar magnetic susceptibilities for **3** under zero dc field.

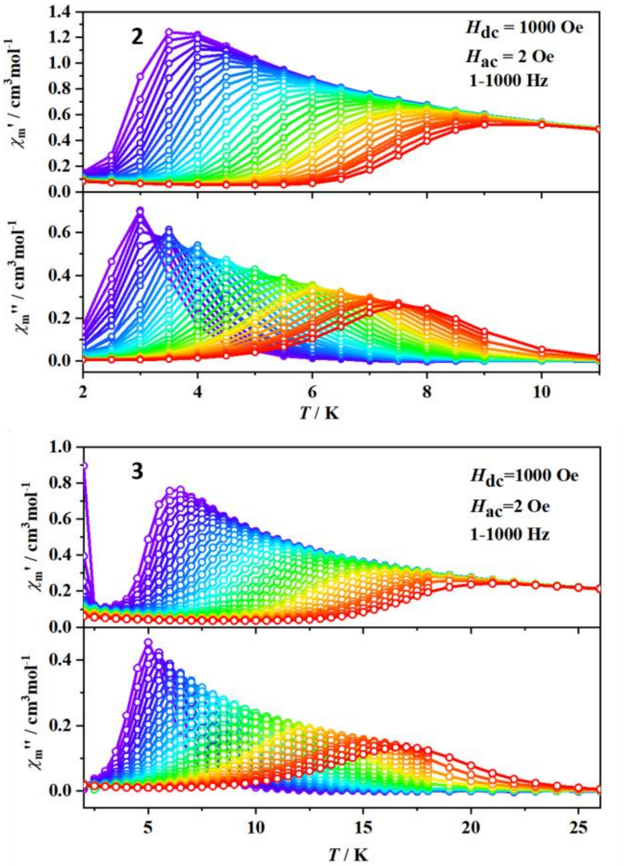

Figure S12 Temperature dependence of the alternating-current molar magnetic susceptibilities for **2** and **3** in 1000 Oe dc field.

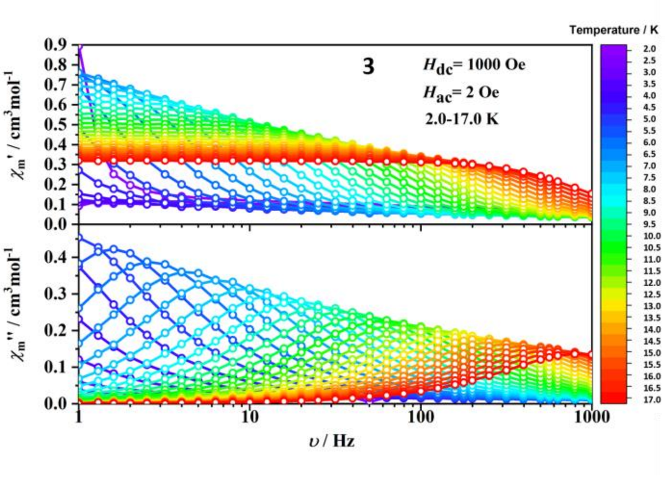

Figure S13 frequency dependence of the alternating-current molar magnetic susceptibilities for **3** in 1000 Oe dc field.

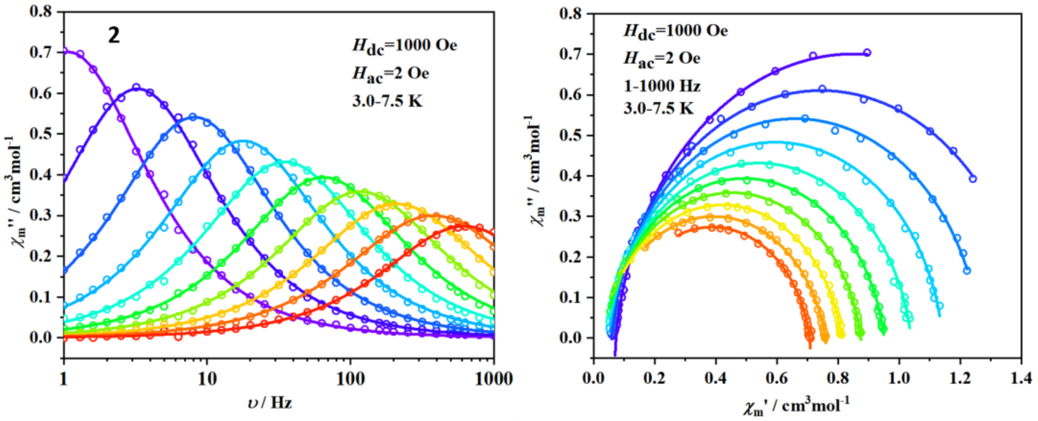

Figure S14 Left, frequency dependence of the alternating-current molar magnetic susceptibilities for **2** in 1000 Oe dc field. Right, Cole–Cole plots of **2** measured at 3.0–7.5 K under 1000 Oe dc field, with the best fit to the generalized Debye model.

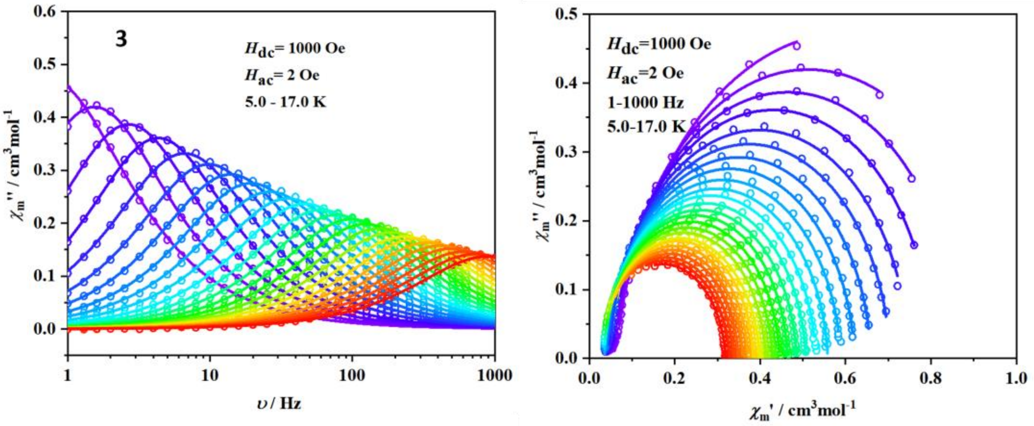

Figure S15 Left, frequency dependence of the alternating-current molar magnetic susceptibilities for **3** in 1000 Oe dc field. Right, Cole–Cole plots of **3** measured at 5.0–17.0 K under 1000 Oe dc field, with the best fit to the generalized Debye model.

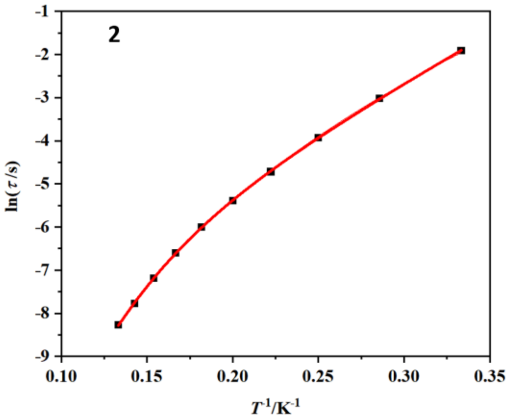

Figure S16 Plot of ln(τ) versus T^{-1} for **2** under 1000 Oe dc field. The solid lines represent the fit results with the consideration of Orbach and Raman processes.

Table S3 Relaxation fitting parameters of **2** at 3.0–7.5 K from Least-Squares Fitting of χ(f) data under 1000 Oe dc field (1-1000 Hz).

Temperature (K)	χ_t (cm^3 mol^{-1})	χ_s (cm^3 mol^{-1})	α	τ (s)
3.0	1.63	0.0737	0.067	0.148
3.5	1.43	0.0590	0.073	0.0487
4.0	1.27	0.0534	0.072	0.0196
4.5	1.14	0.0477	0.079	0.00894
5.0	1.04	0.0415	0.091	0.00457
5.5	0.950	0.0368	0.094	0.00247
6.0	0.875	0.0317	0.102	0.00136
6.5	0.812	0.0289	0.112	0.00076
7.0	0.758	0.0288	0.123	0.00042
7.5	0.706	0.0688	0.097	0.00026

Table S4 Relaxation fitting parameters of **3** at 5.0–17.0 K from Least-Squares Fitting of χ(f) data under zero dc field (1-1000 Hz).

Temperature (K)	χ_t (cm^3 mol^{-1})	χ_s (cm^3 mol^{-1})	α	τ (s)
5.0	1.14	0.0619	0.048	0.214
5.5	0.965	0.0597	0.033	0.103
6.0	0.872	0.0577	0.024	0.0594
6.5	0.805	0.0553	0.038	0.0368
7.0	0.750	0.0452	0.037	0.0241
7.5	0.704	0.0438	0.037	0.0163
8.0	0.660	0.0402	0.036	0.0114
8.5	0.620	0.0374	0.042	0.00822
9.0	0.589	0.0349	0.039	0.00605
9.5	0.558	0.0343	0.029	0.00452
10.0	0.530	0.0343	0.030	0.00343
10.5	0.506	0.0335	0.029	0.00264
11.0	0.484	0.0331	0.022	0.00206
11.5	0.463	0.0334	0.025	0.00162
12.0	0.445	0.0322	0.028	0.00129
12.5	0.428	0.0309	0.028	0.00104
13.0	0.412	0.0306	0.037	0.00085
13.5	0.398	0.0282	0.020	0.00069
14.0	0.383	0.0318	0.027	0.00057
14.5	0.371	0.0301	0.027	0.00047
15.0	0.359	0.0306	0.029	0.00039
15.5	0.348	0.0303	0.048	0.00032
16.0	0.338	0.0223	0.042	0.00026
16.5	0.328	0.0254	0.042	0.00022
17.0	0.319	0.0259	0.048	0.00018

Ab initio Calculation

All calculations were done with CASSCF/RASSI/SINGLE_ANISO implanted in MOLCAS 8.0 package[7-9]. The calculation fragment is built based on the X-ray determined molecule structure of complex **3**. The basis sets for all atoms are atomic natural orbitals from the MOLCAS ANO-RCC library: ANO-RCC-VTZP for Dy^{III} ions; VTZ for close N, Se and C; VDZ for distant atoms. The calculations employed the second order Douglas-Kroll-Hess Hamiltonian, where scalar relativistic contractions were taken into account in the basis set and the spin-orbit couplings were handled separately in the restricted active space state interaction (RASSI-SO) procedure. The active electrons in 7 active spaces include all f electrons CAS (9 in 7) for complexes **3** in the CASSCF calculation. To exclude all the doubts, we calculated all the roots in the active space. We have mixed the maximum number of spin-free state which was possible with our hardware (21 sextets, 128 quadruplets and 130 doublets). The Single_Aniso program was then used to obtain electronic states, *g* tensors and so forth.

Table S5 Relative energy (***E***), effective ***g*** values and crystal field wavefunctions of the lowest 8 Kramers Doublets (**KD**s) obtained respectively from ab initio calculation of **3**.

KDs	***E***(cm^{-1})	***E***(K^{-1})	g_x	g_y	g_z	Wavefunction
1	0	0	0.002	0.002	19.931	99.52%|±15/2⟩+0.35%|±11/2⟩+0.06%|±9/2⟩ +0.04%|±7/2⟩+0.03%|±5/2⟩
2	118.76	170.87	0.030	0.036	18.020	0.05%|±15/2⟩+69.93%|±13/2⟩+19.74%|±11/2⟩ +6.54%|±7/2⟩+2.73%|±7/2⟩+0.84%|±5/2⟩ +0.14%|±3/2⟩+0.03%|±1/2⟩
3	190.75	274.44	0.057	0.093	14.356	0.17%|±15/2⟩+27.88%|±13/2⟩+48.41%|±11/2⟩ +13.64%|±9/2⟩+5.60%|±7/2⟩+3.04%|±5/2⟩ +1.13%|±3/2⟩+1.13%|±1/2⟩
4	273.62	393.68	0.551	0.607	12.539	0.22%|±15/2⟩+0.68%|±13/2⟩+26.84%|±11/2⟩ +61.91%|±9/2⟩+2.73%|±7/2⟩+3.60%|±5/2⟩ +2.93%|±3/2⟩+1.10%|±1/2⟩
5	359.04	516.57	1.345	2.115	9.317	1.10%|±13/2⟩+2.26%|±11/2⟩+11.54%|±9/2⟩ +72.52%|±7/2⟩+1.84%|±5/2⟩+5.43%|±3/2⟩ +5.32%|±1/2⟩
6	441.11	634.67	3.712	6.259	9.492	0.02%|±15/2⟩+0.09%|±13/2⟩+1.16%|±11/2⟩ +2.65%|±9/2⟩+5.93%|±7/2⟩+58.19%|±5/2⟩ +13.65%|±3/2⟩+18.31%|±1/2⟩
7	572.98	824.39	0.759	1.209	14.876	0.01%|±15/2⟩+0.26%|±13/2⟩+0.90%|±11/2⟩ +2.81%|±9/2⟩+8.60%|±7/2⟩+26.25%|±5/2⟩ +49.63%|±3/2⟩+11.54%|±1/2⟩
8	716.83	1031.36	0.102	0.272	18.542	0.06%|±13/2⟩+0.35%|±11/2⟩+0.86%|±9/2⟩ +1.84%|±7/2⟩+6.21%|±5/2⟩+27.10%|±3/2⟩ +63.57%|±1/2⟩

Table S6 The *ab initio* computed crystal field parameters of complex **3**.

k	q	B_k^q
2	-2	0.85
	-1	-0.50
	0	-3.5
	1	-1.2
	2	-1.7
4	-4	0.0073
	-3	0.042
	-2	-0.016
	-1	0.0026
	0	0.0035
	1	0.00057
	2	0.0043
	3	-0.056
	4	0.0011
6	-6	-0.00012
	-5	-0.00032
	-4	0.000031
	-3	-0.00013
	-2	0.00014
	-1	0.0000089
	0	-0.000030
	1	0.00011
	2	0.00011
	3	0.00018
	4	-0.000011
	5	-.00034
	6	-0.000030

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