Supporting Information

A series of counter cation dependent tetra β-diketonate mononuclear lanthanide(III) single-molecule magnets and immobilization on pre-functionalised GaN substrates by anion exchange reaction

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Table S1-S6. Selected bond bengths (Å) and Angles (°) for **1**-**5**

Table S1 Complex **1Tb**

Tb1-O1	2.375(6)	Tb1-O5	2.345(6)
Tb1-O2	2.346(5)	Tb1-O6	2.348(5)
Tb1-O3	2.324(6)	Tb1-O7	2.357(6)
Tb1-O4	2.341(5)	Tb1-O8	2.352(5)
O2-Tb1-O1	70.0(2)	O4-Tb1-O6	146.0(2)
O2-Tb1-O6	75.22(18)	O4-Tb1-O7	140.25(19)
O2-Tb1-O7	79.90(19)	O4-Tb1-O8	76.97(18)
O2-Tb1-O8	144.2(2)	O5-Tb1-O1	74.8(2)
O3-Tb1-O1	112.55(18)	O5-Tb1-O2	134.21(19)
O3-Tb1-O2	75.95(18)	O5-Tb1-O6	71.1(2)
O3-Tb1-O4	71.3(2)	O5-Tb1-O7	116.14(19)
O3-Tb1-O5	146.47(18)	O5-Tb1-O8	79.2(2)
O3-Tb1-O6	141.1(2)	O6-Tb1-O1	81.06(18)
O3-Tb1-O7	78.3(2)	O6-Tb1-O7	71.3(2)
O3-Tb1-O8	77.27(19)	O6-Tb1-O8	114.13(16)
O4-Tb1-O1	73.79(18)	O7-Tb1-O1	143.43(18)
O4-Tb1-O2	115.49(17)	O8-Tb1-O1	143.5(2)
O4-Tb1-O5	80.4(2)	O8-Tb1-O7	71.77(19)

Table S2 Complex **1Nd**

Nd1-O1	2.415(3)	Nd1-O5	2.408(3)
Nd1-O2	2.426(3)	Nd1-O6	2.450(3)
Nd1-O3	2.413(3)	Nd1-O7	2.410(3)
Nd1-O4	2.433(3)	Nd1-O8	2.415(3)
O1-Nd1-O2	68.91(11)	O5-Nd1-O4	77.43(11)
O1-Nd1-O4	81.84(11)	O5-Nd1-O6	75.38(11)
O1-Nd1-O6	74.61(12)	O5-Nd1-O7	117.03(12)
O2-Nd1-O4	112.28(11)	O5-Nd1-O8	69.42(11)
O2-Nd1-O6	81.20(11)	O7-Nd1-O1	132.32(11)
O3-Nd1-O1	116.43(12)	O7-Nd1-O2	76.16(12)
O3-Nd1-O2	72.02(12)	O7-Nd1-O3	80.40(12)
O3-Nd1-O4	69.25(11)	O7-Nd1-O4	142.87(13)
O3-Nd1-O6	143.16(11)	O7-Nd1-O6	68.74(11)
O3-Nd1-O8	80.54(12)	O7-Nd1-O8	76.63(11)
O4-Nd1-O6	146.47(11)	O8-Nd1-O1	146.43(11)
O5-Nd1-O1	80.37(11)	O8-Nd1-O2	143.97(12)
O5-Nd1-O2	145.35(11)	O8-Nd1-O4	77.70(11)
O5-Nd1-O3	139.04(11)	O8-Nd1-O6	110.02(11)

Table S3 Complex **2**

Dy1-O2	2.328(2)	Dy1-O5	2.331(2)
Dy1-O3	2.338(2)	Dy1-O1	2.350(2)
Dy1-O8	2.351(2)	Dy1-O4	2.370(2)
Dy1-O6	2.389(2)	Dy1-O7	2.368(2)
Dy2-O13	2.347(2)	Dy2-O12	2.354(2)
Dy2-O15	2.336(2)	Dy2-O11	2.346(2)
Dy2-O10	2.356(2)	Dy2-O9	2.364(3)
Dy2-O14	2.384(2)	Dy2-O16	2.390(2)
O2-Dy1-O5	131.92(8)	O2-Dy1-O3	79.46(8)
O5-Dy1-O3	144.76(8)	O2-Dy1-O1	70.43(8)
O5-Dy1-O1	77.08(8)	O3-Dy1-O1	106.02(8)
O2-Dy1-O8	75.15(9)	O5-Dy1-O8	120.91(9)
O3-Dy1-O8	77.64(9)	O1-Dy1-O8	143.85(8)
O2-Dy1-O7	143.20(8)	O5-Dy1-O7	79.70(8)
O3-Dy1-O7	79.10(8)	O1-Dy1-O7	144.82(8)
O8-Dy1-O7	71.27(8)	O2-Dy1-O4	124.64(9)
O5-Dy1-O4	75.75(8)	O3-Dy1-O4	71.86(9)
O1-Dy1-O4	73.49(8)	O8-Dy1-O4	138.21(8)
O7-Dy1-O4	75.41(8)	O2-Dy1-O6	74.31(8)
O5-Dy1-O6	70.02(8)	O3-Dy1-O6	144.01(8)

O1-Dy1-O6	88.02(8)	O8-Dy1-O6	72.16(8)
O7-Dy1-O6	108.37(8)	O4-Dy1-O6	143.98(8)
O11-Dy2-O15	92.23(9)	O15-Dy2-O13	101.26(8)
O11-Dy2-O13	145.97(8)	O15-Dy2-O12	142.35(9)
O11-Dy2-O12	75.01(8)	O13-Dy2-O12	75.32(8)
O15-Dy2-O10	75.29(9)	O11-Dy2-O10	70.28(9)
O13-Dy2-O10	143.31(8)	O12-Dy2-O10	129.29(9)
O15-Dy2-O9	146.68(8)	O11-Dy2-O9	100.90(9)
O13-Dy2-O9	84.73(8)	O12-Dy2-O9	70.96(9)
O10-Dy2-O9	80.54(8)	O15-Dy2-O14	77.10(8)
O11-Dy2-O14	143.33(8)	O13-Dy2-O14	70.61(8)
O12-Dy2-O14	132.70(8)	O10-Dy2-O14	73.07(8)
O9-Dy2-O14	74.11(9)	O15-Dy2-O16	70.28(9)
O11-Dy2-O16	74.85(8)	O13-Dy2-O16	80.53(8)
O12-Dy2-O16	72.20(9)	O10-Dy2-O16	129.17(8)
O9-Dy2-O16	142.69(8)	O14-Dy2-O16	130.69(8)

Table S4 Complex **3**

Dy1-O4	2.3750(18)	Dy1-O2	2.3454(18)
Dy1-O6	2.3044(18)	Dy1-O5	2.3783(17)
Dy1-O3	2.3224(19)	Dy1-O7	2.3299(19)
Dy1-O1	2.3579(18)	Dy1-O8	2.3709(18)
O4-Dy1-O5	70.92(6)	O1-Dy1-O4	139.74(6)
O6-Dy1-O4	76.78(7)	O1-Dy1-O5	122.35(6)
O6-Dy1-O3	142.80(6)	O1-Dy1-O8	77.88(6)
O6-Dy1-O1	141.81(7)	O7-Dy1-O4	75.38(6)
O6-Dy1-O7	112.44(7)	O7-Dy1-O1	77.21(6)
O6-Dy1-O2	80.42(7)	O7-Dy1-O2	141.30(6)
O6-Dy1-O5	72.47(6)	O7-Dy1-O5	143.76(7)
O6-Dy1-O8	71.25(6)	O7-Dy1-O8	71.73(7)
O3-Dy1-O4	73.12(6)	O2-Dy1-O4	142.73(6)
O3-Dy1-O1	73.73(6)	O2-Dy1-O1	72.06(6)
O3-Dy1-O7	80.50(7)	O2-Dy1-O5	74.30(6)
O3-Dy1-O2	111.86(7)	O2-Dy1-O8	79.22(6)
O3-Dy1-O5	77.36(6)	O8-Dy1-O4	119.54(6)
O3-Dy1-O8	143.88(6)	O8-Dy1-O4	137.87(6)

Table S5 Complex **4**

Dy1—O23	2.318(6)	Dy1—O22	2.344(6)
Dy1—O21	2.319(6)	Dy1—O24	2.357(5)
Dy1—O19	2.323(5)	Dy1—O18	2.360(5)
Dy1—O20	2.343(6)	Dy1—O17	2.365(5)
Dy2—O13	2.346(6)	Dy2—O12	2.333(6)
Dy2—O16	2.354(6)	Dy2—O9	2.340(6)
Dy2—O14	2.366(5)	Dy2—O10	2.340(5)
Dy2—O11	2.402(6)	Dy2—O15	2.343(6)
Dy3—O8	2.323(5)	Dy3—O5	2.351(6)
Dy3—O3	2.327(6)	Dy3—O6	2.367(5)
Dy3—O2	2.338(6)	Dy3—O7	2.371(5)
Dy3—O1	2.347(6)	Dy3—O4	2.379(7)
O23—Dy1—O21	134.07(19)	O13—Dy2—O16	137.49(19)
O23—Dy1—O19	74.0(2)	O12—Dy2—O14	72.07(19)
O21—Dy1—O19	148.05(19)	O9—Dy2—O14	151.8(2)
O23—Dy1—O20	116.5(2)	O10—Dy2—O14	129.6(2)
O21—Dy1—O20	80.6(2)	O15—Dy2—O14	76.8(2)
O19—Dy1—O20	71.22(19)	O13—Dy2—O14	73.18(19)
O23—Dy1—O22	75.8(2)	O16—Dy2—O14	74.4(2)
O21—Dy1—O22	71.1(2)	O12—Dy2—O11	71.4(2)
O19—Dy1—O22	139.8(2)	O9—Dy2—O11	73.6(2)
O20—Dy1—O22	147.7(2)	O10—Dy2—O11	79.9(2)
O23—Dy1—O24	71.0(2)	O15—Dy2—O11	133.7(2)
O21—Dy1—O24	74.54(19)	O13—Dy2—O11	148.43(19)
O19—Dy1—O24	110.21(19)	O16—Dy2—O11	73.6(2)

O20—Dy1—O24	73.65(19)	O14—Dy2—O11	122.87(19)
O22—Dy1—O24	83.9(2)	O8—Dy3—O3	144.8(2)
O23—Dy1—O18	146.6(2)	O8—Dy3—O2	75.2(2)
O21—Dy1—O18	76.82(19)	O3—Dy3—O2	80.2(2)
O19—Dy1—O18	81.63(19)	O8—Dy3—O1	116.3(2)
O20—Dy1—O18	75.2(2)	O3—Dy3—O1	77.7(3)
O22—Dy1—O18	111.9(2)	O2—Dy3—O1	71.3(2)
O24—Dy1—O18	140.4(2)	O8—Dy3—O5	136.7(2)
O23—Dy1—O17	81.4(2)	O3—Dy3—O5	75.6(2)
O21—Dy1—O17	116.8(2)	O2—Dy3—O5	144.5(2)
O19—Dy1—O17	76.9(2)	O1—Dy3—O5	78.4(2)
O20—Dy1—O17	136.05(18)	O8—Dy3—O6	76.0(2)
O22—Dy1—O17	72.9(2)	O3—Dy3—O6	111.6(2)
O24—Dy1—O17	147.57(19)	O2—Dy3—O6	141.93(19)
O18—Dy1—O17	70.9(2)	O1—Dy3—O6	145.3(2)
O12—Dy2—O9	135.8(2)	O5—Dy3—O6	72.3(2)
O12—Dy2—O10	75.6(2)	O8—Dy3—O7	71.41(19)
O9—Dy2—O10	72.3(2)	O3—Dy3—O7	142.4(3)
O12—Dy2—O15	148.3(2)	O2—Dy3—O7	111.8(2)
O9—Dy2—O15	75.8(2)	O1—Dy3—O7	73.4(2)
O10—Dy2—O15	122.0(2)	O5—Dy3—O7	75.3(2)
O12—Dy2—O13	91.5(2)	O6—Dy3—O7	81.44(19)
O9—Dy2—O13	105.1(2)	O8—Dy3—O4	77.4(2)
O10—Dy2—O13	70.1(2)	O3—Dy3—O4	71.9(3)
O15—Dy2—O13	73.3(2)	O2—Dy3—O4	74.0(2)
O12—Dy2—O16	103.9(2)	O1—Dy3—O4	136.9(2)
O9—Dy2—O16	91.1(2)	O5—Dy3—O4	121.0(2)
O10—Dy2—O16	151.9(2)	O6—Dy3—O4	75.8(2)
O15—Dy2—O16	73.1(2)	O7—Dy3—O4	145.0(2)

Table S6 Complx **5**

Dy1—O1	2.509(4)	Dy1—O7	2.406(4)
Dy1—O5	2.433(4)	Dy1—O4	2.518(4)
Dy1—O8	2.264(4)	Dy1—O6	2.276(4)
Dy1—O3	2.300(4)	Dy1—O2	2.326(4)
O1—Dy1—O4	153.89(15)	O7—Dy1—O1	66.65(15)
O7—Dy1—O5	76.64(16)	O7—Dy1—O4	138.89(15)
O5—Dy1—O1	137.63(14)	O5—Dy1—O4	67.17(14)
O8—Dy1—O1	117.68(15)	O8—Dy1—O7	77.66(15)
O8—Dy1—O5	71.72(14)	O8—Dy1—O4	73.11(15)
O8—Dy1—O6	140.13(16)	O8—Dy1—O3	135.27(15)
O8—Dy1—O2	79.77(15)	O6—Dy1—O1	74.08(15)
O6—Dy1—O7	73.04(15)	O6—Dy1—O5	75.62(15)
O6—Dy1—O4	114.00(15)	O6—Dy1—O3	81.21(15)
O6—Dy1—O2	137.87(15)	O3—Dy1—O1	84.00(15)
O3—Dy1—O7	145.05(15)	O3—Dy1—O5	119.60(15)
O3—Dy1—O4	73.37(15)	O3—Dy1—O2	69.15(16)
O2—Dy1—O1	73.65(15)	O2—Dy1—O7	116.62(15)
O2—Dy1—O5	145.23(14)	O2—Dy1—O4	86.01(15)

Table S7 Continuous Shape Measures (CShMs) of the coordination geometry for Dy(III) ion in complex 1 (S values calculated with the Shape program). The *S* values indicated the proximity to the ideal polyhedron, thus, a *S* = 0 corresponds to the non-distorted polyhedron. The three closer ideal geometries to the real complexes are listed and below is the symmetry and description for each polyhedron.

Complexes	**s**	**polyhedron**

1Nd		0.488	SAPR-8, D_{4d}, Square antiprism
		1.943	TDD-8, D_{2d}, Triangular dodecahedron
		2.198	BTPR-8, C_{2v}, Biaugmented trigonal prism
1Tb		0.290	SAPR-8, D_{4d}, Square antiprism
		2.123	TDD-8, D_{2d}, Triangular dodecahedron
		2.265	BTPR-8, C_{2v}, Biaugmented trigonal prism
2	Dy1	0.818	SAPR-8, D_{4d}, Square antiprism
		1.076	TDD-8, D_{2d}, Triangular dodecahedron
		1.725	BTPR-8, C_{2v}, Biaugmented trigonal prism
	Dy2	0.457	TDD-8, D_{2d}, Triangular dodecahedron
		1.989	SAPR-8, D_{4d}, Square antiprism
		2.140	BTPR-8, C_{2v}, Biaugmented trigonal prism
3		0.343	SAPR-8, D_{4d}, Square antiprism
		1.735	TDD-8, D_{2d}, Triangular dodecahedron
		1.895	BTPR-8, C_{2v}, Biaugmented trigonal prism
4	Dy1	0.478	SAPR-8, D_{4d}, Square antiprism
		1.554	TDD-8, D_{2d}, Triangular dodecahedron
		2.214	BTPR-8, C_{2v}, Biaugmented trigonal prism
	Dy2	0.471	TDD-8, D_{2d}, Triangular dodecahedron
		1.473	SAPR-8, D_{4d}, Square antiprism
		2.369	BTPR-8, C_{2v}, Biaugmented trigonal prism
	Dy3	0.298	SAPR-8, D_{4d}, Square antiprism
		1.615	TDD-8, D_{2d}, Triangular dodecahedron
		2.007	BTPR-8, C_{2v}, Biaugmented trigonal prism

5	0.746	SAPR-8, D_{4d}, Square antiprism
	2.464	TDD-8, D_{2d}, Triangular dodecahedron
	2.580	BTPR-8, C_{2v}, Biaugmented trigonal prism

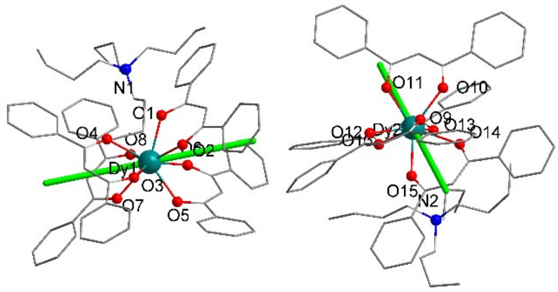


Fig. S1 Orientations of the anisotropy axes for each of the two Dy(III) ions in complexes **2** as calculated by MAGELLAN.

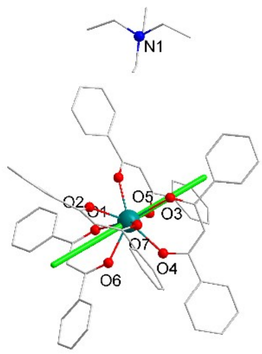


Fig. S2 Orientations of the anisotropy axes for Dy(III) ion in complexes **3** as calculated by MAGELLAN.

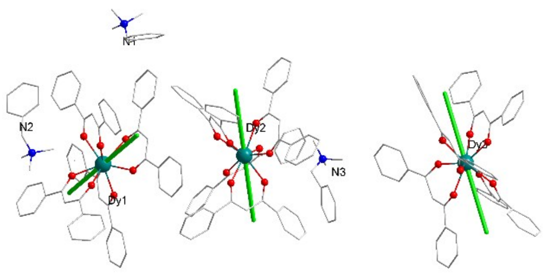


Fig. S3 Orientations of the anisotropy axes for each of the three Dy(III) ions in complexes **4** as calculated by MAGELLAN.

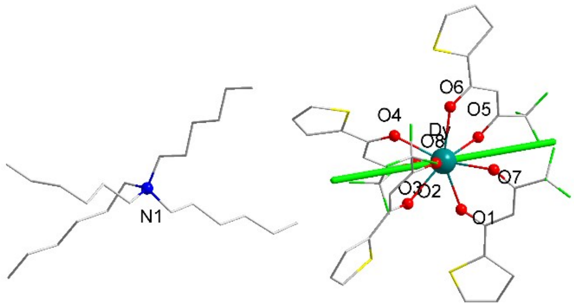


Fig. S4 Orientations of the anisotropy axes for each of Dy(III) ion in complexes **5** as calculated by MAGELLAN.

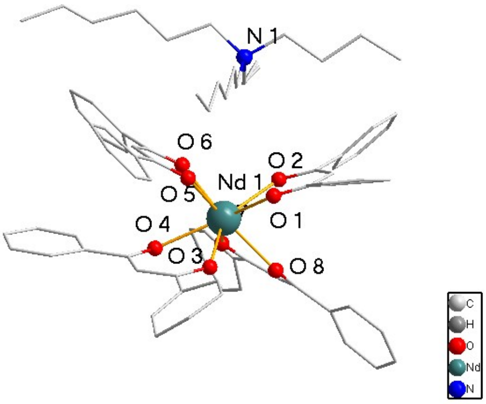


Fig. S5 The crystal structure of complex **1Nd** (hydrogen atoms are omitted for clarity).

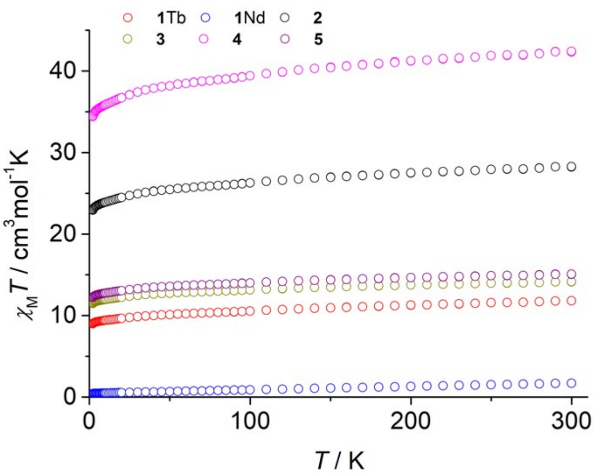


Fig. S6 experimental data of temperature dependence of the $\chi_M T$ values of complexes **1-5**.

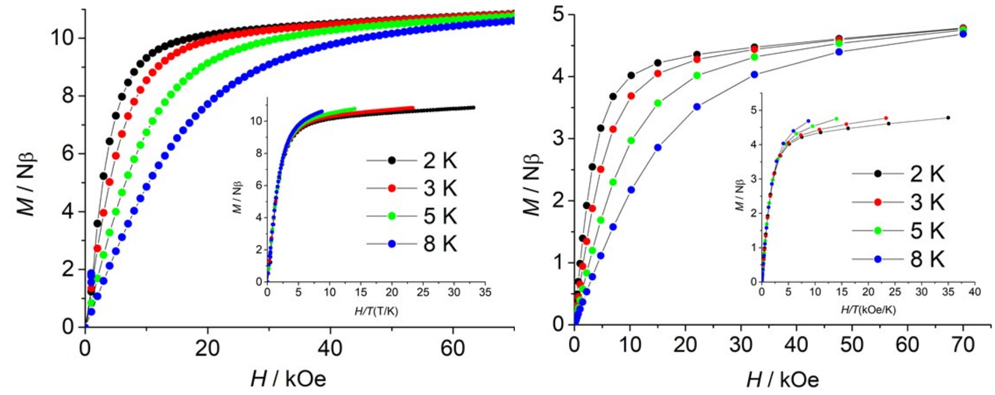


Fig. S7 Field dependence of the magnetization between 2 and 8 K for **2** and **3**.

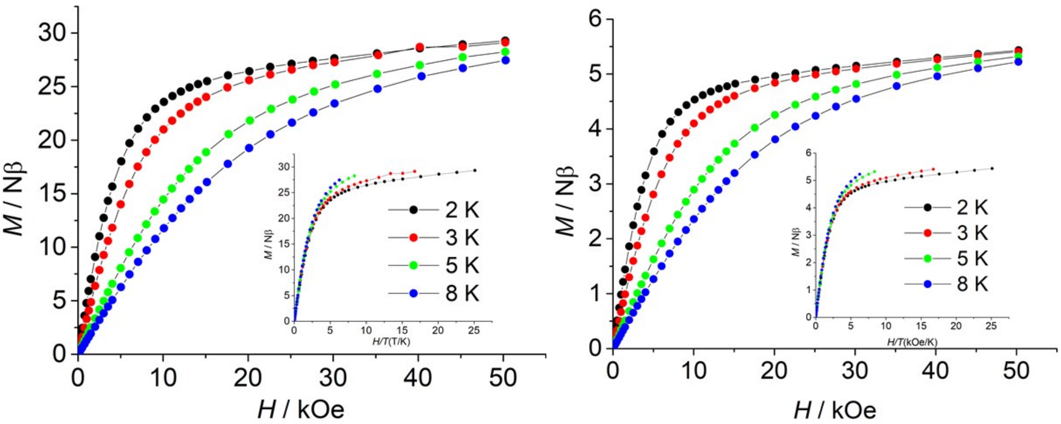


Fig. S8 Field dependence of the magnetization between 2 and 8 K for **4** and **5**.

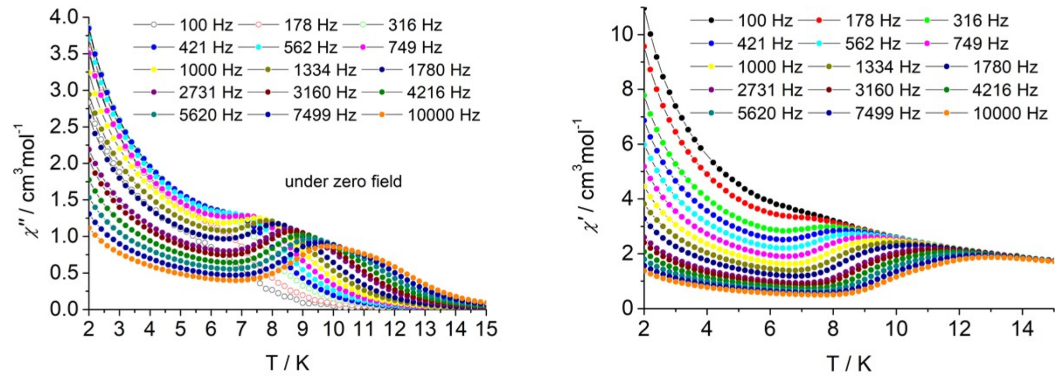


Fig. S9 The temperature dependence of the out-of-phase (χ'') ac susceptibility and in-phase (χ') ac susceptibility of complex **2** under 0 Oe in the frequency range 1-10000 Hz.

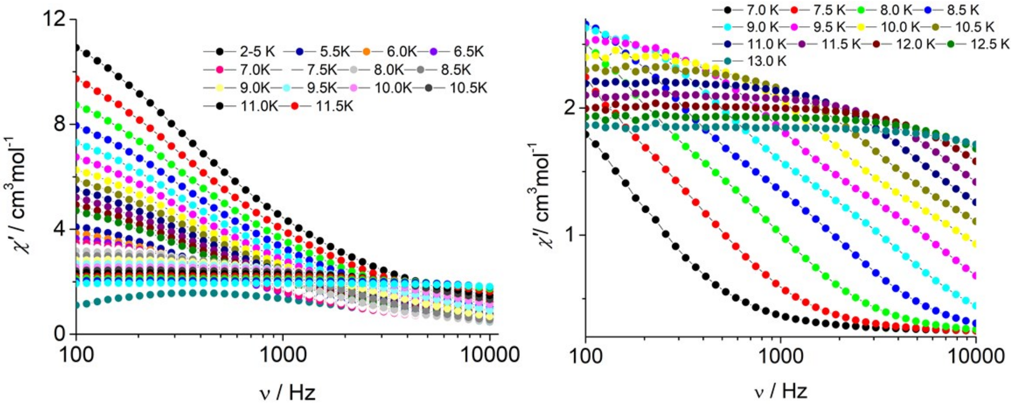


Fig. S10 The frequency dependence of the in-phase (χ') ac susceptibility of complex **2** under 0 Oe and 2000 Oe in the frequency range 1-10000 Hz.

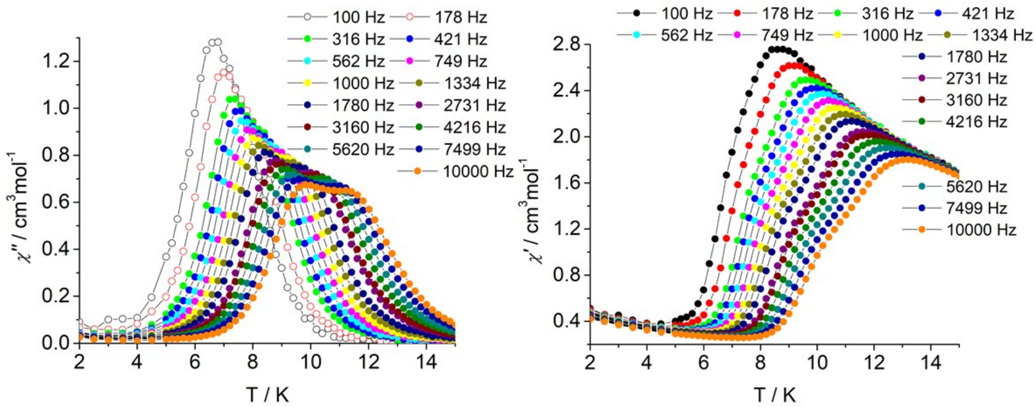


Fig. S11 The temperature dependence of the out-of-phase (χ'') ac susceptibility and in-phase (χ') ac susceptibility of complex **2** under 2000 Oe in the frequency range 1-10000 Hz.

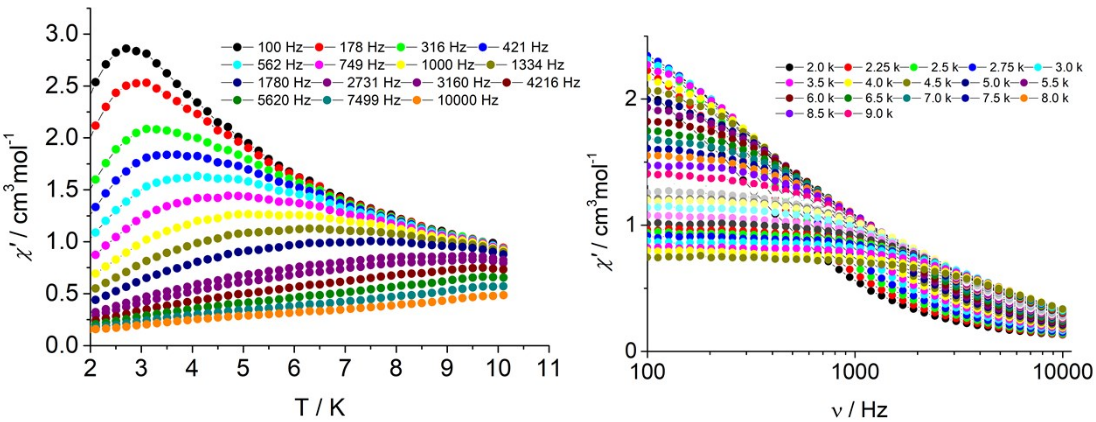


Fig. S12 The temperatue dependence and frequency dependence of the in-phase (χ') ac susceptibility of complex **1Tb** under 1500 Oe in the frequency range 1-10000 Hz.

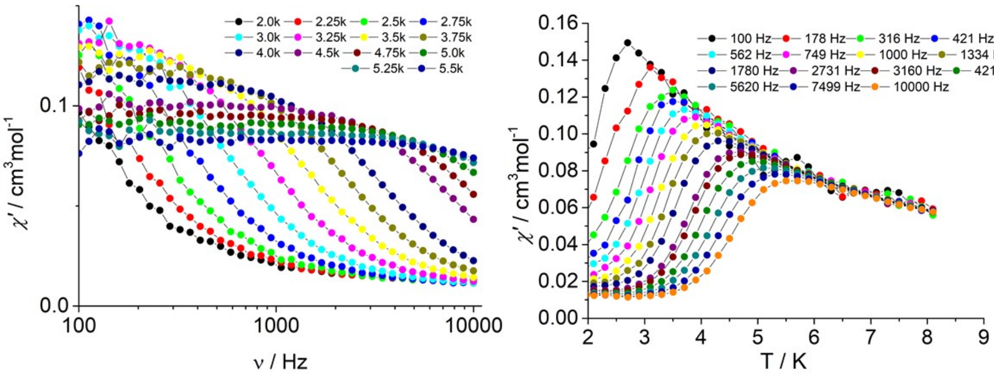


Fig. S13 The temperatue dependence and frequency dependence of the in-phase (χ') ac susceptibility of complex **1Nd** under 2000 Oe in the frequency range 1-10000 Hz.

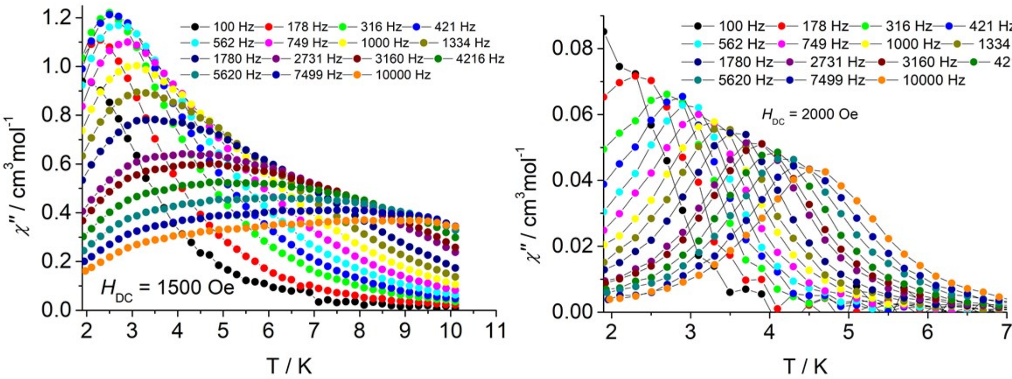


Fig. S14 The temperature dependence of the out-of-phase (χ'') ac susceptibility of complex **1(Tb)** under 1500 Oe and the out-of-phase (χ'') ac susceptibility of complex **1(Nd)** under 2000 Oe in the frequency range 1-10000 Hz.

Fig. S15 The temperature dependence of the in-phase (χ') and the frequency dependence of the in-phase (χ') ac susceptibility component under 1500 Oe for complex **1**(Tb).

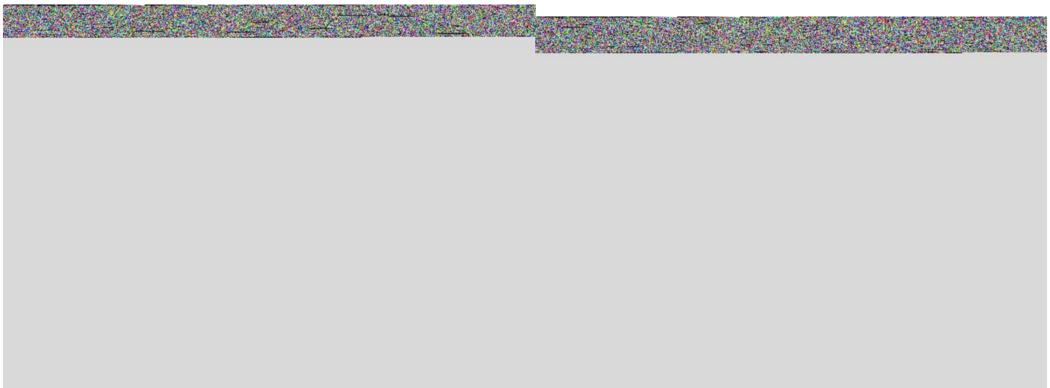


Fig. S16 The temperature dependence of the in-phase (χ') and the frequency dependence of the in-phase (χ') ac susceptibility component under 2000 Oe for complex **1**(Nd).

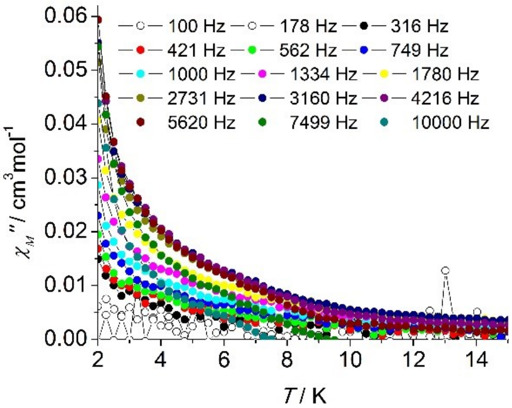


Fig. S17 The temperature dependence of the in-phase (χ'') ac susceptibility component under 0 Oe for complex **3**.

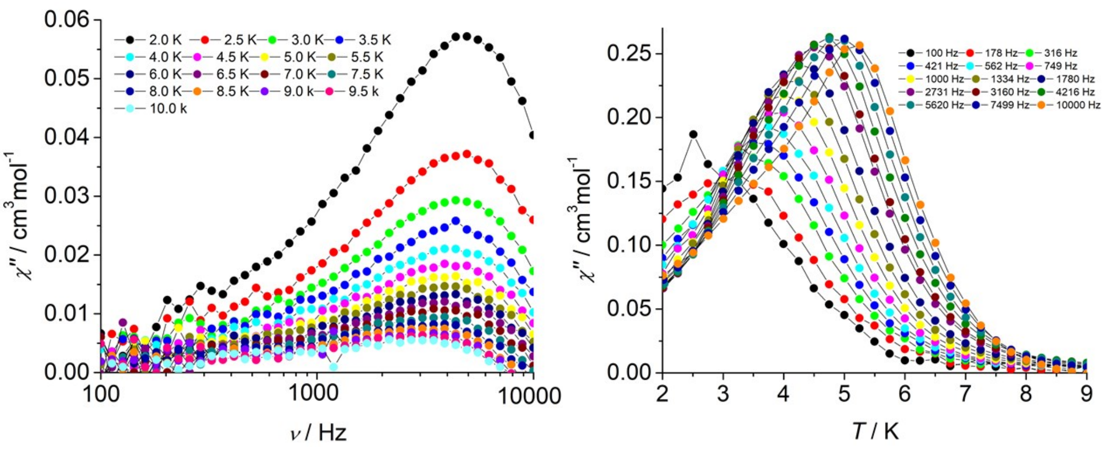


Fig. S18 The frequency dependence of the out-of-phase (χ'') and the temperature dependence of the out-of-phase (χ'') ac susceptibility component under 4000 Oe for complex **3**.

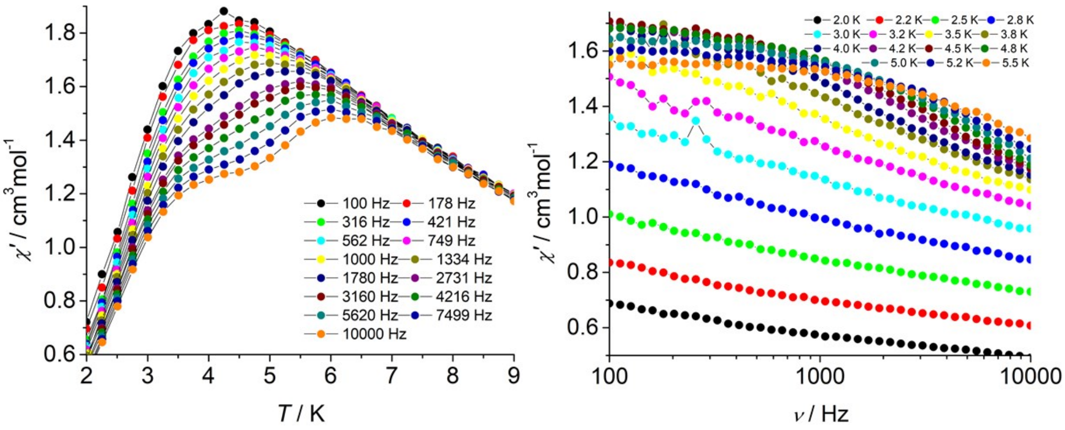


Fig. S19 The temperature dependence of the in-phase (χ') and the frequency dependence of the in-phase (χ') ac susceptibility component under 4000 Oe for complex **3**.

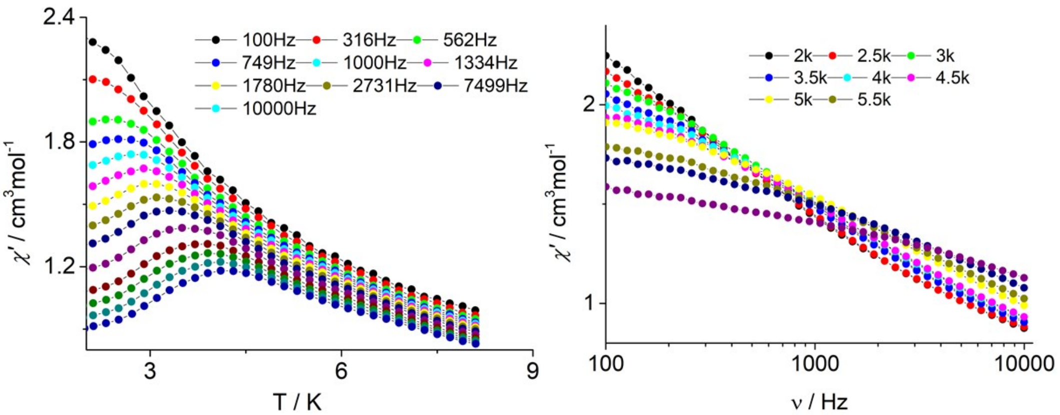


Fig. S20 The temperature dependence of the in-phase (χ') and the frequency dependence of the in-phase (χ') ac susceptibility component under 900 Oe for complex **4**.

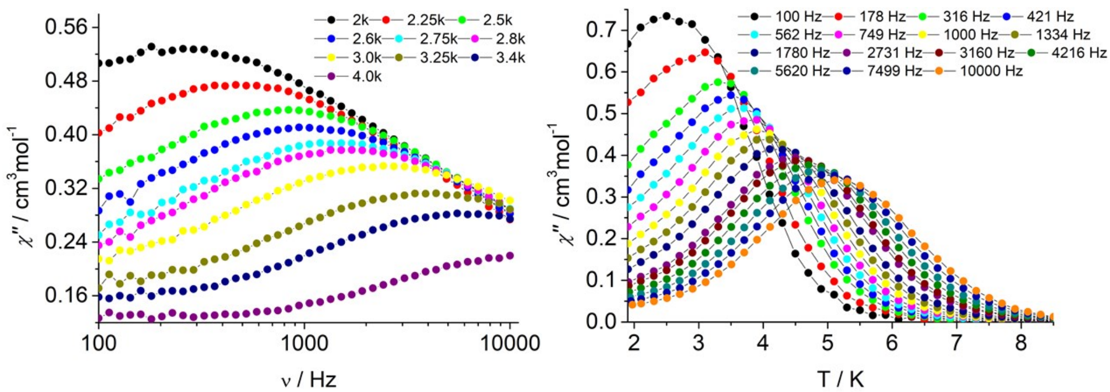


Fig. S21 The frequency dependence of the out-of-phase (χ'') ac susceptibility component under 900 Oe for complex **4** and the temperature dependence of the out-of-phase (χ'') ac susceptibility component under 700 Oe for complex **5**.

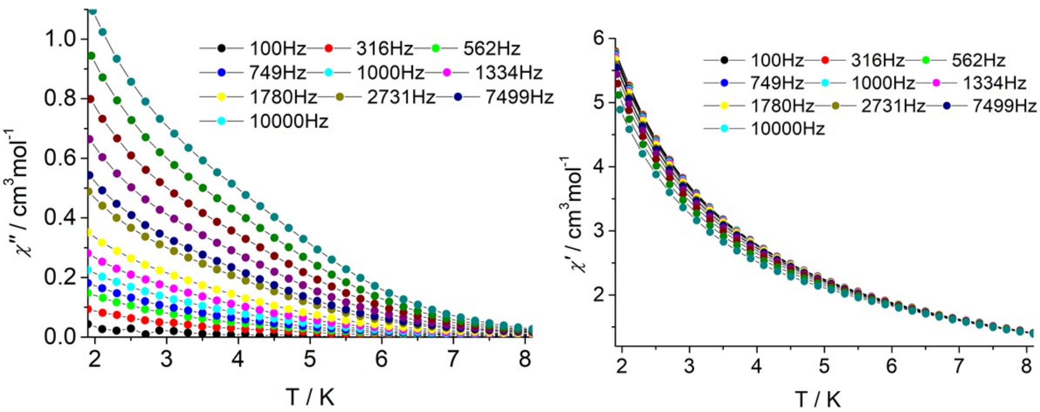


Fig. S22 The temperature dependence of the out-of-phase (χ'') and the in-phase (χ') ac susceptibility component under 0 Oe for complex **5**.

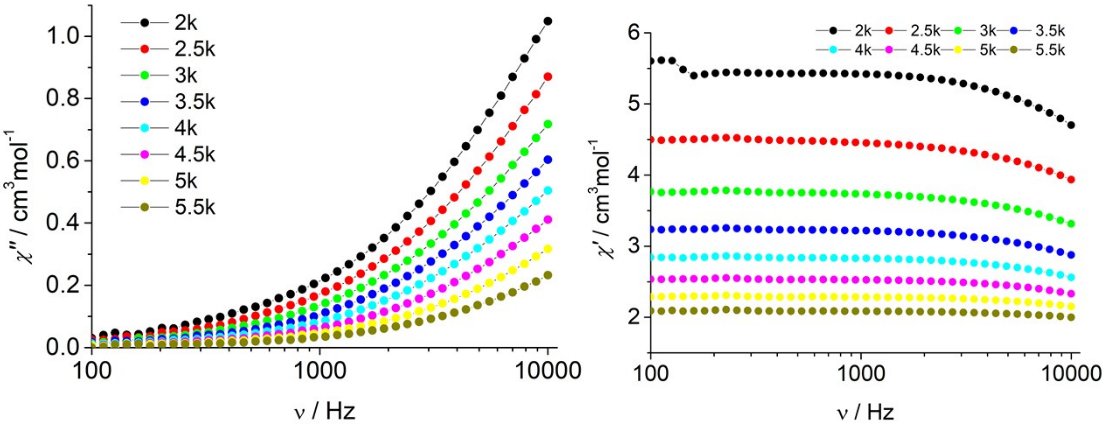


Fig. S23 The frequency dependence of the out-of-phase (χ'') and the in-phase (χ') ac susceptibility component under 0 Oe for complex **5**.

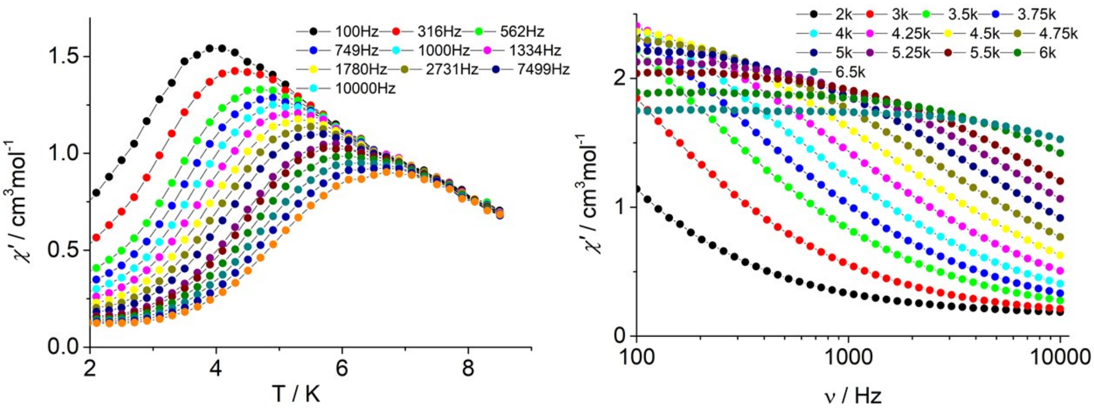


Fig. S24 The temperature dependence of the in-phase (χ') and the frequency dependence of the in-phase (χ') ac susceptibility component under 700 Oe for complex **5**.

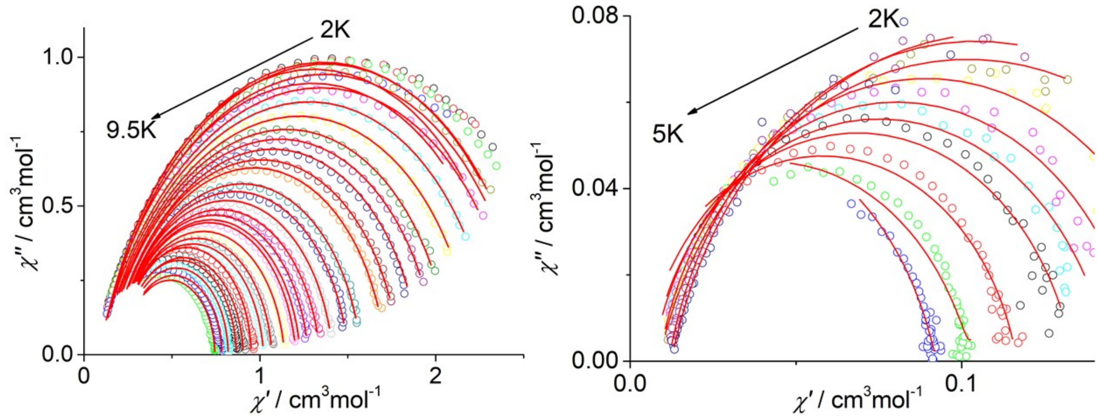


Fig. S25 Cole-Cole (Argand) plot for **1**Tb and **1**Nd obtained using the ac susceptibility data. The solid lines correspond to the best fit obtained with a generalized Debye model under 1500 Oe and 2000 Oe.

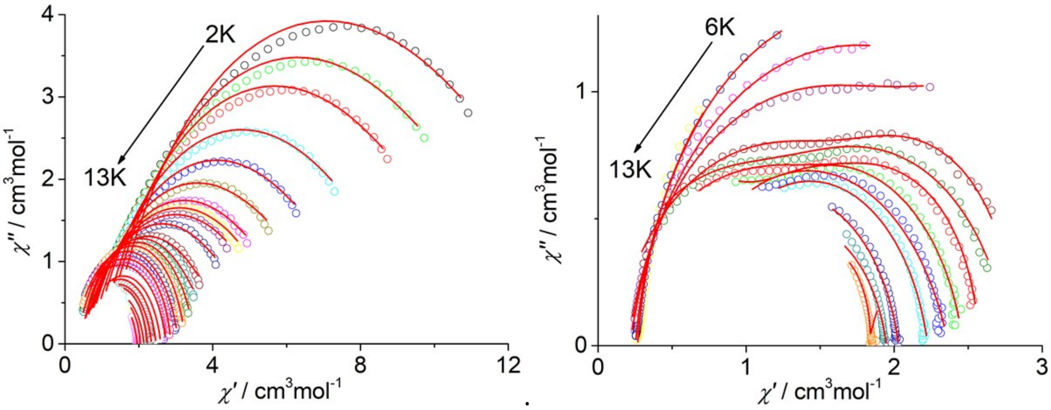


Fig. S26 Cole-Cole (Argand) plot for **2** obtained using the ac susceptibility data. The solid lines correspond to the best fit obtained with a generalized Debye model under zero field and under 2000 Oe.

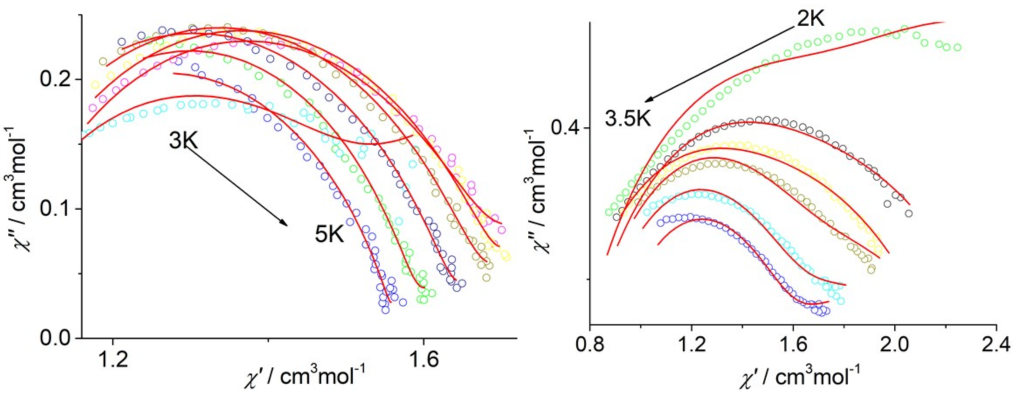


Fig. S27 Cole-Cole (Argand) plot for **3** and **4** obtained using the ac susceptibility data. The solid lines correspond to the best fit obtained with a generalized Debye model under 700 Oe and 4000 Oe.

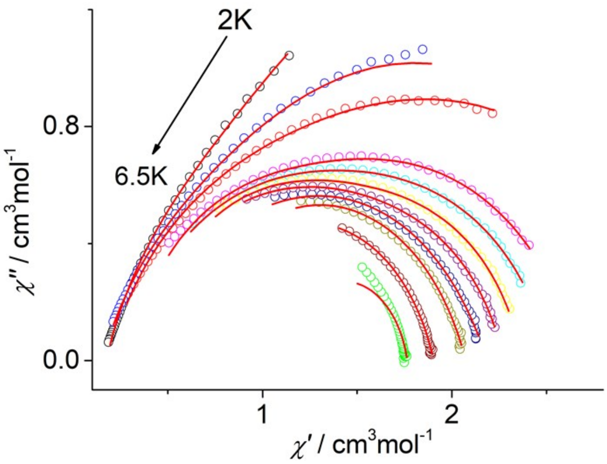


Fig. S28 Cole-Cole (Argand) plot for **5** obtained using the ac susceptibility data. The solid lines correspond to the best fit obtained with a generalized Debye model under 900 Oe.

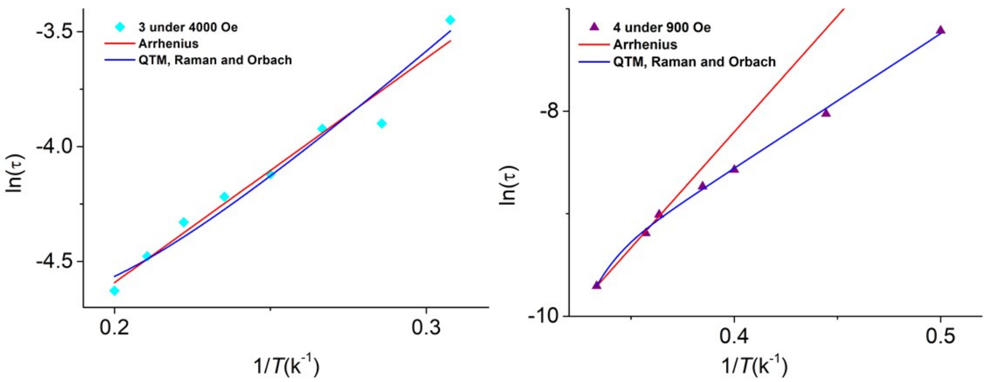
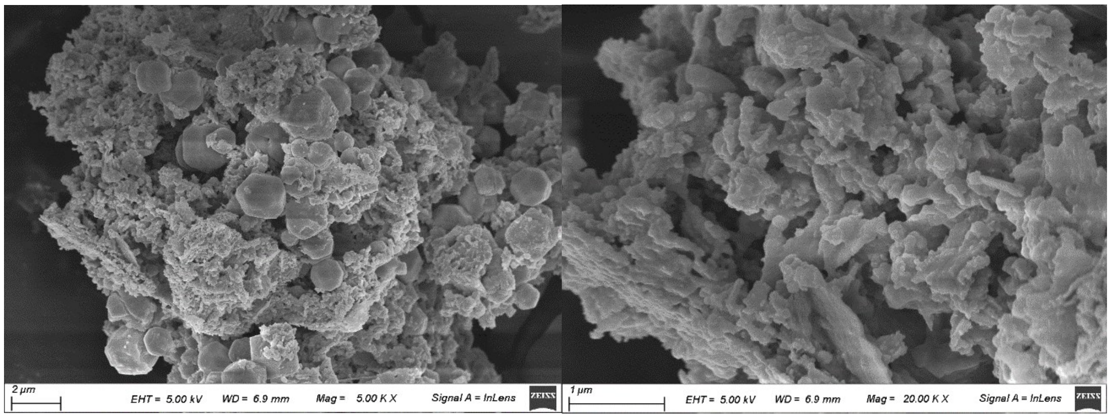


Fig. S29 The ln(τ) vs. 1/T plot for **3** and **4** based on the ac susceptibility data under optimum field



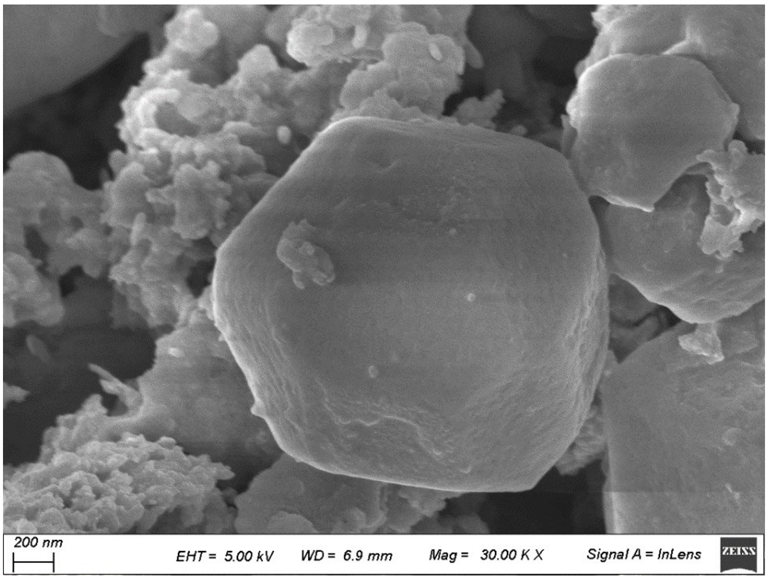


Fig. S30 SEM image of the GaN-IM$^+$-[Dy(DBM)$_4$]$^-$.

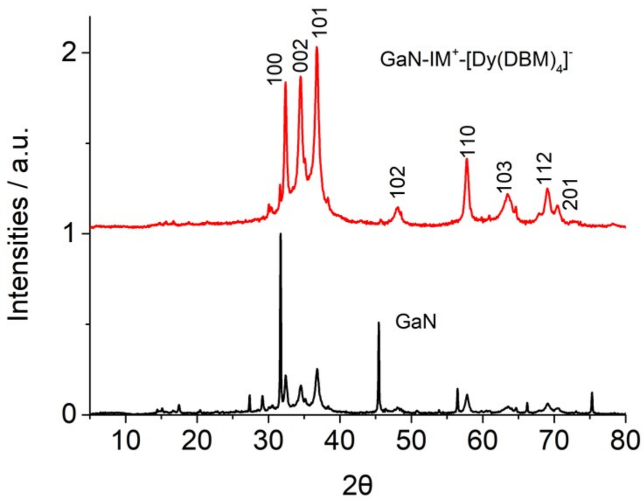


Fig. S31 X-ray Diffraction (XRD) patterns for the pure GaN and GaN-IM$^+$ -[Dy(DBM)$_4$]$^-$.

Table S8 Energy barriers obtained from the Arrhenius law fitting and Equation 1 of the out-of-phase (χ'') ac susceptibility data under optimum dc field.

Relaxation processes	Orbach processes		Raman and Orbach processes			
	U_{eff}/κ_B (K)	τ_0 (s)	C ($s^{-1}\cdot K^{-n}$)	n	U_{eff}/κ_B (K)	τ_0 (s)
1(Tb)	24.1	1×10^{-7}	20.796	5.5	37.5	2.243×10^{-7}
1(Nd)	23.7	2×10^{-5}	2.0098	6	27.766	1.956×10^{-7}
2	116.98	6.1×10^{-10}	0.152	4.179	129.25	1.172×10^{-10}
3	9.7	1.7×10^{-3}	2.0354	2.43	13.12	1.107×10^{-6}

4	22.6	3.2×10^{-8}	2	5.845	14.127	7.0744×10^{-6}
5	42.6	7.1×10^{-9}	14.232	3.393	55.58	5.85×10^{-10}

Table S9 Energy barriers obtained from the Arrhenius law fitting and Equation 1 of the out-of-phase (χ'') ac susceptibility data under optimum 0 field.

Relaxation processes	Orbach processes		Raman, QTM and Orbach processes				
	U_{eff}/κ_B (K)	τ_0 (s)	q	C ($s^{-1}\cdot K^{-n}$)	n	U_{eff}/κ_B (K)	τ_0 (s)
2	57.7	8.3×10^{-8}	4.481×10^{-4}	0.73	5.168	77.6	2×10^{-8}

Table S10 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **2** at 0 Oe in the temperature range 2-13 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1	τ_2/s	α_2	β
2	1.19100	13.66910	0.00034	0.22722	0.00435	0.41695	0.71
2.25	1.045	12.19109	0.00034	0.23	0.00441	0.42758	0.71
2.5	0.939	10.89701	0.00034	0.23	0.00389	0.42987	0.71
3	0.75	8.79003	0.00032	0.23	0.00146	0.39859	0.64673
3.5	0.67	7.67003	0.00032	0.23	0.00212	0.41456	0.67183
4	0.568	7.08799	0.00031	0.22999	0.00315	0.44	0.63914
4.5	0.513	6.03011	0.00031	0.23	0.00169	0.43382	0.64116
4.75	0.46001	5.52	0.00029	0.22999	0.00068	0.36224	0.4768
5	0.447	5.23305	0.00024	0.22072	0.0006	0.32438	0.31092
5.5	0.45262	5.41513	0.00031	0.22972	0.01205	0.43997	0.70513
6.5	0.41720	4.67600	0.00027	0.19667	0.01673	0.44	0.71
7	0.43145	4.198	0.00022	0.14118	0.01222	0.43678	0.71
7.5	0.33802	3.17748	0.0002	0.13001	0.00011	0.19342	0.71
8	0.37313	2.87512	0.00008	0.13	0.00015	0.08417	0.35785

8.5	0.28569	2.77983	0.0001	0.13001	0.00006	0.11188	0.65875
9	0.28801	2.57572	0.00004	0.13258	0.0001	0.01571	0.70763
9.5	0.12323	2.61189	0.00005	0.13023	0.00001	0.14399	0.70957
10	0.56305	2.85397	0.00003	0.13002	0.50289	0.09290	0.71000
10.5	0.53028	2.72095	0.00002	0.13001	0.49744	0.09714	0.70999
11	0.56610	2.5	0.00002	0.13	0.49683	0.09553	0.70997
11.5	0.04884	2.16616	0.00002	0.13	0.00001	0.05724	0.31049
12	0.65500	2.08744	0.00001	0.13001	0.48120	0.09422	0.7097
12.5	0.30001	1.73601	0.00001	0.23	0.00001	0.05435	0.31042
13	0.19742	2.15106	0.00001	0.13124	0.00001	0.10000	0.70995

Table S11 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **1(Tb)** at 1500 Oe in the temperature range 2-9.5 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1
2	0.08444	2.40689	0.0005	0.15203
2.25	0.09755	2.43482	0.00043	0.12446
2.5	0.09186	2.53117	0.0004	0.13626
2.75	0.09898	2.54486	0.00036	0.1415
3	0.07956	2.52368	0.00032	0.16352
3.25	0.08204	2.47491	0.0003	0.18047
3.5	0.09618	2.34148	0.00026	0.17457
3.75	0.10241	2.22521	0.00023	0.1763
4	0.11589	2.08889	0.0002	0.16619
4.25	0.11883	2.01025	0.00018	0.16750
4.5	0.14265	1.91547	0.00016	0.15669

4.75	0.145	1.79692	0.00014	0.14606
5	0.15509	1.71746	0.00013	0.13773
5.5	0.16538	1.57695	0.0001	0.12993
5.75	0.169	1.50248	0.00009	0.12385
6	0.17122	1.41458	0.00009	0.11487
6.25	0.17151	1.3419	0.00008	0.1151
6.5	0.17916	1.27544	0.00007	0.09804
6.75	0.191	1.22979	0.00007	0.08608
7	0.19735	1.20401	0.00006	0.0794
7.25	0.19794	1.1392	0.00006	0.07987
7.5	0.19932	1.06787	0.00005	0.06558
7.75	0.20444	1.02029	0.00005	0.05763
8	0.213	0.95052	0.00004	0.01421
8.25	0.21904	0.93168	0.00004	0.0029
8.5	0.22325	0.90521	0.00004	0.02735
8.75	0.22575	0.86513	0.00004	0.00445
9	0.233	0.81921	0.00003	0.0033
9.25	0.23524	0.78315	0.00003	0.01761
9.5	0.23472	0.74365	0.00003	0.00969

Table S12 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **1(Nd)** at 2000 Oe in the temperature range 2-9.5 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1	τ_2/s	α_2	β
2	0.01248	0.3314	0.00185	0.15437	0.1982	0.11555	0.56742
2.25	0.01162	0.41357	0.00127	0.13724	0.25127	0.04242	0.43668

2.5	0.0086	0.35664	0.00088	0.17427	0.17143	0.2202	0.49674
2.75	0.00868	0.36435	0.00051	0.13038	0.30761	0.0586	0.44034
3	0.00670	0.24089	0.0003	0.13012	0.505	0.01792	0.61159
3.25	0.00609	0.20248	0.00018	0.13028	0.505	0.27731	0.67881
3.5	0.0037	0.39802	0.00011	0.13009	0.505	0.1	0.32629
4	0.00024	0.21956	0.00004	0.13	0.33986	0.07185	0.53198
4.5	0.01742	0.24748	0.00001	0.16494	0.18428	0.21098	0.48372
5	0.04664	0.34899	0.00004	0.149	0.34255	0.08459	0.39739

Table S13 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **2** at 2000 Oe in the temperature range 6-13 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1	τ_2/s	α_2	β
6	0.2651	2.8	0.00494	0.13	0.00314	0.09978	0.70923
6.5	0.25094	3.18792	0.00378	0.15807	0.00202	0.02491	0.53207
7	0.21661	4.48037	0.00113	0.1355	0.01753	0.02062	0.57483
7.5	0.20466	3.20016	0.00044	0.13	0.00307	0.08704	0.66179
8	0.17783	3	0.00017	0.13012	0.00132	0.08592	0.52614
8.5	0.09828	2.71688	0.00009	0.18914	0.00072	0.09976	0.68876
9	0.055	2.72471	0.00029	0.13612	0.00003	0.12252	0.5709
9.5	0.08001	2.50002	0.00003	0.22997	0.00023	0.06544	0.71
10	0.05272	2.41333	0.00008	0.13	0.00001	0.07977	0.65444
10.5	0.46999	2.80436	0.00003	0.19965	0.50141	0.09661	0.67602
11	0.63467	2.46410	0.00002	0.13	0.50271	0.09744	0.64610
11.5	0.56238	2.22622	0.00001	0.13001	0.50010	0.09656	0.69976
12	0.57005	2.05537	0.00001	0.13	0.50496	0.04817	0.70999

12.5	0.78230	1.641	0.00001	0.13	0.32479	0.09669	0.70997
13	1.11002	1.546	0.01111	0.13	0.00001	0.07279	0.53285

Table S14 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **3** at 4000 Oe in the temperature range 3.5-5.5 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1	τ_2/s	α_2	β
3.5	1.01803	1.41454	0.00008	0.22946	0.02616	0.43981	0.32385
4	1.05383	1.3821	0.00007	0.22998	0.04379	0.25876	0.45144
4.2	1.03755	1.71303	0.00006	0.22896	0.20071	0.41099	0.37583
4.5	1.02128	1.74027	0.00005	0.23	0.12435	0.2042	0.39236
4.7	0.98962	1.42558	0.00003	0.22993	0.19369	0.35329	0.48239
5	0.96419	1.94891	0.00003	0.23	0.45076	0.27747	0.34857
5.2	1.00293	1.59057	0.00002	0.17971	0.505	0.38228	0.36901
5.5	0.97331	1.50795	0.00002	0.23	0.07894	0.08592	0.39295

Table S15 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **4** at 900 Oe in the temperature range 3.5-5.5 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1	τ_2/s	α_2	β
2	0.70632	2.692	0.00008	0.22999	0.00273	0.38863	0.31
2.6	0.70903	1.78123	0.00006	0.21907	0.0007	0.43467	0.35895
2.8	0.64	1.63823	0.00003	0.22243	0.00036	0.43996	0.33005
3	0.72321	1.61001	0.00004	0.23	0.0014	0.43987	0.51445
3.25	0.75605	1.49220	0.00004	0.22997	0.00327	0.43964	0.54108
3.5	0.82298	1.41533	0.00003	0.22973	0.00581	0.43995	0.5148

Table S16 Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for complex **5** at 700 Oe in the temperature range 3.5-6.5 K.

T/ K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ_1/s	α_1
3.5	0.22304	2.42685	0.00043	0.18998
4	0.309	2.61593	0.00027	0.25664
4.25	0.40904	2.56423	0.0002	0.25412
4.5	0.52808	2.43881	0.00013	0.22184
5	0.82278	2.22392	0.00007	0.13549
5.5	1.10409	2.0344	0.00004	0.0202
6	1.321	1.90283	0.00003	0.00035
6.5	1.42866	1.7654	0.00002	0.0003

Table S17 Parameters involved in the SAP geometry for complexes **1-5** compared to the ideal geometry

Parameters	1Tb	1Nd	2	3	4	5	Ideal SAP
Skew angle (Ø)	42.87°	44.64°	47.33°	45.36°	45.98°	45.87°	45°
γ	114.58°	113.93°	113.26°	116.54°	113.95°	116.97°	109.48°
magic angles (*α*)	56.34°	56.86°	57.47°	60.61°	57.27°	59.47°	54.7356°
d_{in} (Å)	2.79	2.86	2.78	2.82	2.77	2.86	d_{in}= d_{pp}
d_{pp}/ d^*_{pp}(Å)	2.54/2.58	2.63/2.61	2.15/1.97	2.44/2.33	2.55/2.45	2.44/2.42	
$\sigma_Ø{}^2$	2.13	2.65	17.07	4.10	1.75	4.41	0
$\sigma_\alpha{}^2$	4.87	7.32	10.33	42.91	9.211	33.35	0

Table S18 The details for parameters involved in SAP geometry for **1Tb** and **1Nd**

Skew angle (Ø) of complex 1Tb (°)		**Skew angle (Ø) of complex 1Nd (°)**	
Ø1 = 42.87(1)	Ø2 = 45.71(1)	Ø1 = 42.96(1)	Ø2 = 45.98(1)
Ø3 = 43.33(2)	Ø4 = 45.87(2)	Ø3 = 43.16(1)	Ø4 = 46.46(1)
Mean value of Ø = 42.87(1)		Mean value of Ø = 44.64(2)	
Parameter *γ* and magic angles *α* for complex 1Tb (°)		**Parameter *γ* and magic angles *α* for complex 1Nd (°)**	

γ(O1-Dy1-O3) = 112.55(2)	α(O1-O3) = 55.32(1)	γ(O1-Dy1-O3) = 112.27(2)	α(O1-O3) = 56.27(1)
γ(O8-Dy1-O6) = 114.14(2)	α(O2-O4) = 57.84(1)	γ(O2-Dy1-O4) = 116.43(2)	α(O2-O4) = 58.25(1)
γ(O2-Dy1-O4) = 115.47(2)	α(O5-O7) = 57.81(1)	γ(O5-Dy1-O7) = 117.03(2)	α(O5-O7) = 58.53(1)
γ(O7-Dy1-O5) = 116.16(2)	α(O6-O8) = 54.42(1)	γ(O6-Dy1-O8) = 110.02(2)	α(O6-O8) = 54.42(1)
Mean value of γ = 114.58	Mean value of α = 56.34	Mean value of γ = 113.93	Mean value of α = 56.86
Values of d_{in} for 1Tb (Å)		**Values of d_{in} for 1Nd (Å)**	
d_{in}(O1-O4) = 2.83(8)	d_{in}(O5-O6) = 2.99(6)	d_{in}(O1-O2) = 2.75(8)	din(O5-O6) = 2.74(6)
d_{in}(O2-O3) = 2.87(8)	d_{in}(O6-O7) = 2.76(6)	d_{in}(O3-O4) = 2.74(8)	din(O7-O8) = 2.74(6)
d_{in}(O1-O2) = 2.71(8)	d_{in}(O7-O8) = 2.74(6)	d_{in}(O2-O3) = 2.84(8)	din(O5-O8) = 2.97(6)
d_{in}(O3-O4) = 2.71(8)	d_{in}(O5-O8) = 2.73(6)	d_{in}(O1-O4) = 3.17(8)	din(O6-O7) = 2.99(6)
Mean value of d_{in} = 2.79(6)		Mean value of d_{in} = 2.86(6)	
d_{pp}(A-B) in 1Tb (Å)		**d_{pp}(A-B) in 1Nd (Å)**	
d_{pp}(O1-B) = 2.58(5)	d_{pp}(O2-B) = 2.55(5)	d_{pp}(O1-B) = 2.67(5)	d_{pp}(O2-B) = 2.62(5)
d_{pp}(O3-B) = 2.55(5)	d_{pp}(O4-B) = 2.48(5)	d_{pp}(O3-B) = 2.73(5)	d_{pp}(O4-B) = 2.51(5)
Mean value of d_{pp}(A-B) = 2.54(5)		Mean value of d_{pp}(A-B) = 2.63(5)	
$\sigma_{Ø}^{2}$ and σ_{α}^{2} for 1Tb		**$\sigma_{Ø}^{2}$ and σ_{α}^{2} for 1Nd**	
$\sigma_{Ø}^{2}$ = 2.13, σ_{α}^{2} = 4.87		$\sigma_{Ø}^{2}$ = 2.65, σ_{α}^{2} = 7.32	

Table S19 The details for parameters involved in SAP geometry for **2** and **3**

Skew angle (Ø) of complex 2 (°)		**Skew angle (Ø) of complex 3 (°)**	
Ø1 = 43.877(1)	Ø2 = 50.335(1)	Ø1 = 44.32(1)	Ø2 = 46.05(1)
Ø3 = 43.993(2)	Ø4 = 51.128(2)	Ø3 = 44.07(1)	Ø4 = 47.02(1)
Mean value of Ø = 47.335(1)		Mean value of Ø = 45.36(2)	
Parameter γ and magic angles α for complex 2 (°)		**Parameter γ and magic angles α for complex 3 (°)**	
γ(O1-Dy1-O3) = 108.37(2)	α(O1-O3) = 54.76(1)	γ(O1-Dy1-O3) = 111.86(2)	α(O1-O3) = 55.51(1)
γ(O2-Dy1-O4) = 118.21(2)	α(O2-O4) = 57.51(1)	γ(O2-Dy1-O4) = 122.35(2)	α(O2-O4) = 60.73(1)

γ(O5-Dy1-O7) = 108.62(2)	α(O5-O7) = 59.28 (1)	γ(O5-Dy1-O7) = 112.44(2)	α(O6-O8) = 56.88(1)
γ(O6-Dy1-O8) = 117.85(2)	α(O6-O8) = 58.32(1)	γ(O6-Dy1-O8) = 119.54(2)	α(O5-O7) = 69.32(1)
Mean value of γ =113.26	Mean value of α = 57.47	Mean value of γ = 116.54	Mean value of α = 60.61
Values of d_{in} for 2 (Å)		**Values of d_{in} for 3 (Å)**	
d_{in}(O1-O2) = 2.72(8)	d_{in}(O5-O6) = 2.87(6)	d_{in}(O1-O2) = 2.93(8)	din(O5-O6) = 2.88(6)
d_{in}(O3-O4) = 2.70(6)	d_{in}(O7-O8) = 2.86(6)	d_{in}(O3-O4) = 2.76(8)	din(O7-O8) = 2.72(6)
d_{in}(O2-O3) = 2.81(8)	d_{in}(O6-O7) = 2.73(6)	d_{in}(O2-O3) = 2.85(8)	din(O6-O7) = 2.91(6)
d_{in}(O1-O4) = 2.88(8)	d_{in}(O5-O8) = 2.73(6)	d_{in}(O1-O4) = 2.81(8)	din(O5-O8) = 2.75(6)
Mean value of d_{in} = 2.78(6)		Mean value of d_{in} = 2.82(6)	
d_{pp}(A-B) in 2 (Å)		**d_{pp}(A-B) in 3 (Å)**	
d_{pp}(O1-B) = 2.468(5)	d_{pp}(O2-B) = 2.457(5)	d_{pp}(O1-B) = 2.46(5)	d_{pp}(O2-B) = 2.23(5)
d_{pp}(O3-B) = 1.852(5)	d_{pp}(O4-B) = 1.853(5)	d_{pp}(O3-B) = 2.65(5)	d_{pp}(O4-B) = 2.44(5)
Mean value of d_{pp}(A-B) = 2.157(5)		Mean value of d_{pp}(A-B) = 2.44(5)	
$\sigma_{\varnothing}{}^2$ and $\sigma_{\alpha}{}^2$ for 2		**$\sigma_{\varnothing}{}^2$ and $\sigma_{\alpha}{}^2$ for 3**	
$\sigma_{\varnothing}{}^2$ = 17.07, $\sigma_{\alpha}{}^2$ = 10.33		$\sigma_{\varnothing}{}^2$ = 4.10, $\sigma_{\alpha}{}^2$ = 42.91	

Table S20 The details for parameters involved in SAP geometry for **4** and **5**

Skew angle (Ø) of complex 4 (°)		**Skew angle (Ø) of complex 5 (°)**	
Ø1 = 45.11(1)	Ø2 = 46.19(1)	Ø1 = 43.85(1)	Ø2 = 47.51(1)
Ø3 = 45.31(1)	Ø4 = 47.34(1)	Ø3 = 44.09(1)	Ø4 = 48.03(1)
Mean value of Ø =45.98(2)		Mean value of Ø = 45.87(2)	
Parameter γ and magic angles α for complex 4 (°)		**Parameter γ and magic angles α for complex 5 (°)**	
γ(O1-Dy1-O3) = 117.00(2)	α(O1-O3) = 59.77(1)	γ(O1-Dy1-O3) = 114.00(2)	α(O1-O3) = 61.45(1)
γ(O2-Dy1-O4) = 111.91(2)	α(O2-O4) = 55.68(1)	γ(O2-Dy1-O4) = 119.59(2)	α(O2-O4) = 62.56(1)
γ(O5-Dy1-O7) = 110.17(2)	α(O5-O7) = 55.86 (1)	γ(O5-Dy1-O7) = 116.62(2)	α(O6-O8) = 54.00(1)
γ(O6-Dy1-O8) = 116.73(2)	α(O6-O8) = 57.77(1)	γ(O6-Dy1-O8) = 117.68(2)	α(O5-O7) = 59.87(1)

Mean value of γ = 113.95	Mean value of α = 57.27	Mean value of γ = 116.97	Mean value of α = 59.47
Values of d_{in} for 4 (Å)		**Values of d_{in} for 5 (Å)**	
d_{in}(O1-O2) = 2.71(8)	d_{in}(O5-O6) = 2.71(6)	d_{in}(O1-O2) = 2.97(8)	din(O5-O6) = 2.70(6)
d_{in}(O3-O4) = 2.75(6)	d_{in}(O7-O8) = 2.71(6)	d_{in}(O3-O4) = 2.74(8)	din(O7-O8) = 2.94(6)
d_{in}(O2-O3) = 2.82(8)	d_{in}(O6-O7) = 2.79(6)	d_{in}(O2-O3) = 2.88(8)	din(O6-O7) = 2.90(6)
d_{in}(O1-O4) = 2.90(8)	d_{in}(O5-O8) = 2.83(6)	d_{in}(O1-O4) = 2.88(8)	din(O5-O8) = 2.93(6)
Mean value of d_{in} = 2.77(6)		Mean value of d_{in} =2.86(6)	
d_{pp}(A-B) in 4 (Å)		**d_{pp}(A-B) in 5 (Å)**	
d_{pp}(O1-B) = 2.46(5)	d_{pp}(O2-B) = 2.59(5)	d_{pp}(O1-B) = 2.48(5)	d_{pp}(O2-B) = 2.45(5)
d_{pp}(O3-B) = 2.57(5)	d_{pp}(O4-B) = 2.58(5)	d_{pp}(O3-B) = 2.54(5)	d_{pp}(O4-B) = 2.32(5)
Mean value of d_{pp}(A-B) = 2.55(5)		Mean value of d_{pp}(A-B) = 2.44(5)	
$\sigma_{\emptyset}{}^2$ and $\sigma_{\alpha}{}^2$ for 4		**$\sigma_{\emptyset}{}^2$ and $\sigma_{\alpha}{}^2$ for 1Nd**	
$\sigma_{\emptyset}{}^2$ = 1.75, $\sigma_{\alpha}{}^2$ = 9.21		$\sigma_{\emptyset}{}^2$ = 4.41, $\sigma_{\alpha}{}^2$ = 33.35	

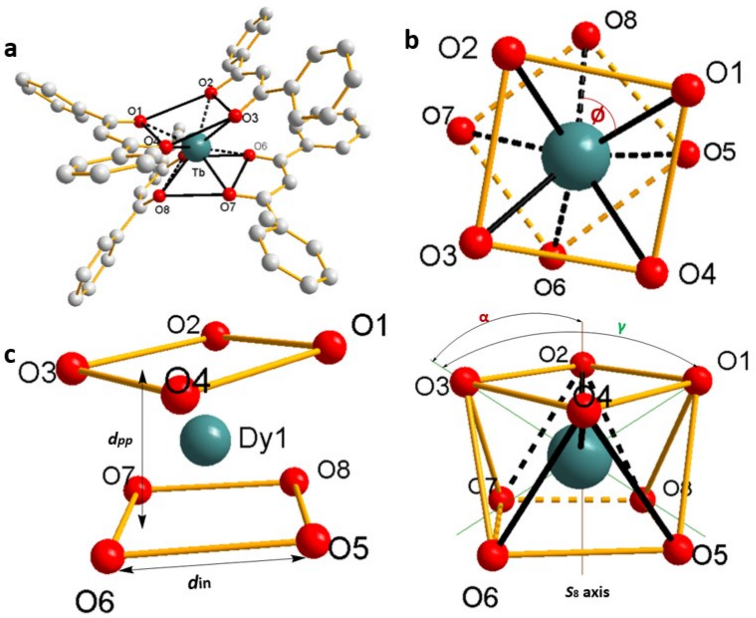


Fig. S32 (a) Crystal structure of **1Tb**. (b) Skew angle (Ø) for the SAP coordination geometry. (c) Scheme showing the parameters d_{in}, d_{pp}, α and γ for the SAP coordination geometry.

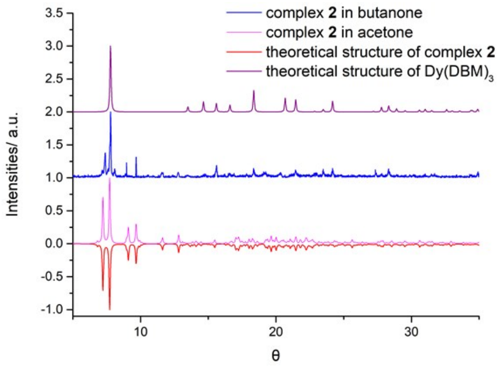


Fig. S33 X-ray Diffraction (XRD) patterns for **2** stirring in butanone and acetone, and the theoretical structure of **2** and $Dy(DBM)_3$.