Supplementary Materials

Slow Relaxation of the Magnetization in Bis-Decorated Chiral Helicene-Based Coordination Complexes of Lanthanides

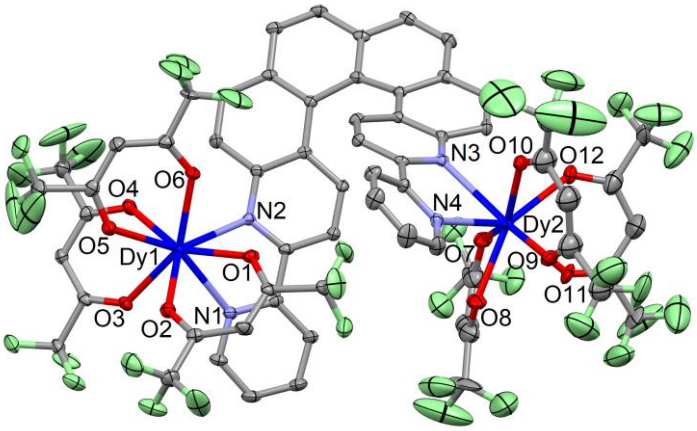

Figure S1. ORTEP view of **1**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and solvent molecules of crystallization are omitted for clarity.

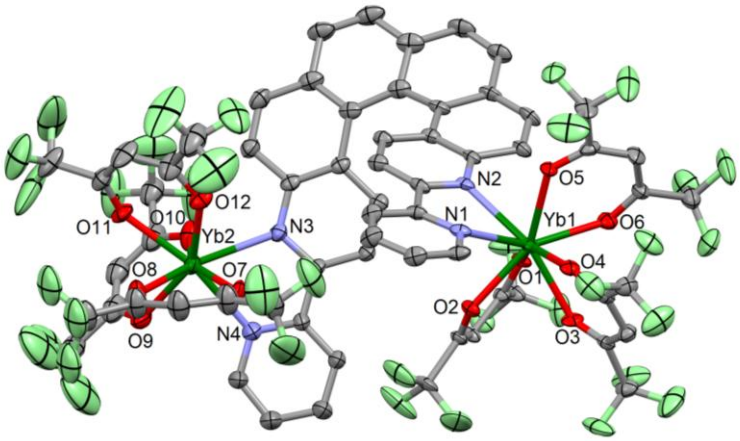

Figure S2. ORTEP view of **2**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms and solvent molecules of crystallization are omitted for clarity.

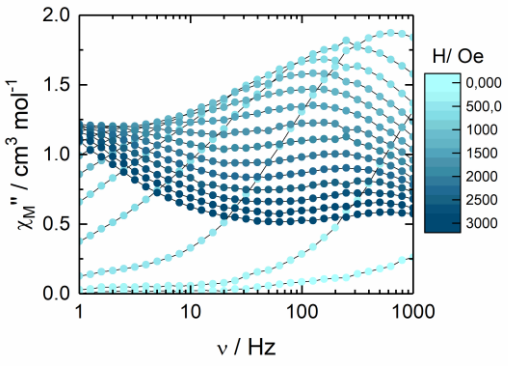

Figure S3. Field dependence of the out-of-phase component of the magnetic susceptibility for **1** at 2 K.

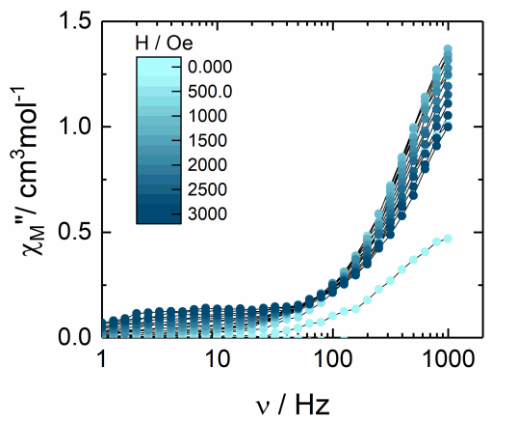

Figure S4. Field dependence of the out-of-phase component of the magnetic susceptibility for **2** at 2 K.

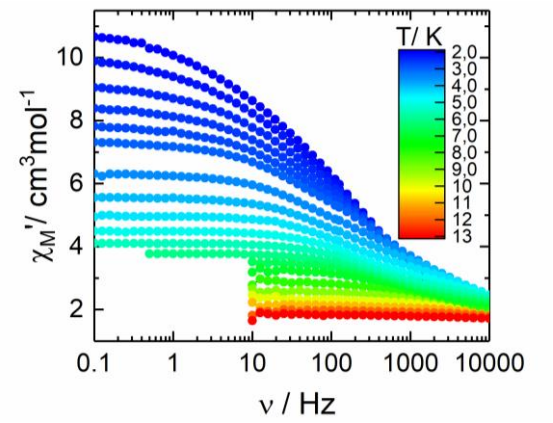

Figure S5. Frequency dependence of the in-phase component of the magnetic susceptibility for **1**.

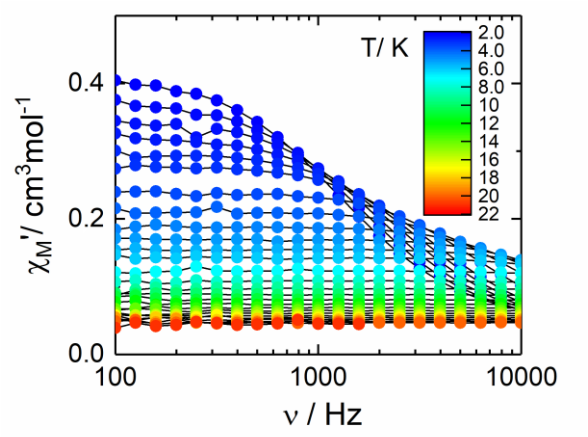

Figure S6. Frequency dependence of the in-phase component of the magnetic susceptibility for **2**.

Extended Debye model.

$$\chi_M' = \chi_S + (\chi_T - \chi_S)\frac{1+(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)}{1+2(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)+(\omega\tau)^{2-2\alpha}}$$

$$\chi_M'' = (\chi_T - \chi_S)\frac{(\omega\tau)^{1-\alpha}\cos\left(\alpha\frac{\pi}{2}\right)}{1+2(\omega\tau)^{1-\alpha}\sin\left(\alpha\frac{\pi}{2}\right)+(\omega\tau)^{2-2\alpha}}$$

With χ_T the isothermal susceptibility, χ_S the adiabatic susceptibility, τ the relaxation time and α an empiric parameter which describe the distribution of the relaxation time. For SMM with only one relaxing object α is close to zero. The extended Debye model was applied to fit simultaneously the experimental variations of χ_M' and χ_M'' with the frequency f of the oscillating field ($\omega = 2\pi f$). Typically, only the temperatures for which a maximum on the χ'' vs. f curves, have been considered. The best fitted parameters τ, α, χ_T, χ_S are listed in Table S3 and S4 with the coefficient of determination R^2.

Table S1. X-ray crystallographic data of **1** and **2**.

Compounds	**1**	**2**
Formula	**$C_{64}H_{26}Dy_2F_{36}N_4O_{12}$**	**$C_{70}H_{40}Yb_2F_{36}N_4O_{12}$**
M / g.mol^{-1}	**2051.89**	**2159.14**
Crystal system	**Triclinic**	**Triclinic**
Space group	**P-1 (N°2)**	**P-1 (N°2)**
Cell parameters	a = 12.2629(16) Å b = 16.534(2) Å c = 18.118(3) Å α = 79.414(5) ° β = 77.667(5) ° γ = 87.190(5) °	a = 12.2906(17) Å b = 17.962(3) Å c = 18.439(3) Å α = 90.015(5) ° β = 99.337(5) ° γ = 100.196(5) °
Volume/Å^3	3527.4(8)	3951.7(10)
Z	2	2
T/K	150 (2)	150 (2)
2θ range /°	4.24 ≤ 2θ ≤ 55.20	5.85 ≤ 2θ ≤ 54.97
ρ_{calc} / g.cm^{-3}	1.932	1.815
μ / mm^{-1}	2.261	2.498
Number of reflections	75302	136147
Independent reflections	15733	17903
$Fo^2 > 2\sigma(Fo)^2$	11594	14464
Number of variables	1027	1025
R_{int}, R_1, ωR_2	0.0758, 0.0672, 0.1585	0.0489, 0.1070, 0.2547

Table S2. SHAPE analysis for **1** and **2**.

		$CShM_{SAPR-8}$ (square antiprism) D_{4d}	**$CShM_{BTPR-8}$ (biaugmented trigonal prism) C_{2v}**	**$CShM_{TDD-8}$ (triangular dodecahedron) D_{2d}**
	Dy1	**0.589**	**2.616**	**2.626**
1	**Dy2**	**1.704**	**1.480**	**1.677**
	Yb1	**1.632**	**1.671**	**0.600**
2	**Yb2**	**1.268**	**2.612**	**1.165**

Table S3. Best-fitted parameters (χT, χS, τ and α) with the extended Debye model 2 at 1000 Oe field in the temperature range 2.0-4 K.

T/K	χ_T/cm^3 mol^{-1}	χ_S/cm^3 mol^{-1}	α	τ/s	R^2
2	0,41509	0,04357	0,12382	1,19997E-4	0,99975
2.2	0,37815	0,04077	0,11062	9,3418E-5	0,9996
2.4	0,34762	0,03734	0,10312	7,35554E-5	0,99897
2.6	0,32366	0,03498	0,10083	5,89835E-5	0,99934
2.8	0,29902	0,03472	0,08331	4,7948E-5	0,99948
3	0,28054	0,03121	0,08488	3,93158E-5	0,99954
3.5	0,23989	0,03147	0,05495	2,51774E-5	0,99925
4	0,21184	0,02786	0,05772	1,65059E-5	0,99941

Table S4. Computed energy levels (the ground state is set at zero), component values of the Lande factor g and wavefunction composition for each MJ state of the ground-state multiplet for Dy1 of **1**.

KD	Energy (cm^{-1})	g_X	g_Y	g_Z	Wavefunction composition *
1	0.0	1.34	2.01	14.94	0.30 |±13/2> + 0.27 |±11/2> + 0.23 |±15/2> + 0.11 |±7/2>
2	55.9	4.98	6.34	9.80	0.22 |±5/2> + 0.19 |±11/2> + 0.16 |±3/2> + 0.13 |±13/2> + 0.12 |±9/2>
3	143.9	4.20	5.04	9.49	0.29 |±13/2> + 0.27 |±9/2> + 0.10 |±3/2> + 0.10 |±1/2>
4	297.4	4.00	5.62	8.84	0.25 |±15/2> + 0.20 |±1/2> + 0.16 |±9/2> + 0.14 |±13/2> + 0.11 |±7/2>
5	383.5	1.39	3.91	13.17	0.30 |±7/2> + 0.18 |±5/2> + 0.13 |±11/2> + 0.11 |±3/2> + 0.10 |±1/2>
6	412.3	0.60	3.58	12.30	0.28 |±1/2> + 0.16 |±9/2> + 0.13 |±15/2> + 0.13 |±11/2> + 0.12 |±5/2>
7	510.8	0.80	3.37	12.45	0.25 |±3/2> + 0.23 |±15/2> + 0.16 |±11/2> + 0.14 |±9/2>
8	662.7	0.80	0.75	17.87	0.35 |±5/2> + 0.22 |±7/2> + 0.19 |±3/2> + 0.11 |±1/2>

* Only the contributions ≥ 10% are given.

Table S5. Computed energy levels (the ground state is set at zero), component values of the Lande factor g and wavefunction composition for each MJ state of the ground-state multiplet for Dy2 of **1**.

KD	Energy (cm^{-1})	g_X	g_Y	g_Z	Wavefunction composition *
1	0.0	0.17	0.33	18.97	0.86 |±15/2> + 0.11 |±11/2>
2	96.0	0.83	1.45	15.46	0.54 |±13/2> + 0.29 |±9/2>
3	164.0	0.82	2.60	13.48	0.32 |±7/2> + 0.30 |±11/2> + 0.14 |±5/2>
4	204.6	0.08	0.74	17.35	0.21 |±11/2> + 0.20 |±1/2> + 0.14 |±5/2> + 0.14 |±3/2> + 0.13 |±13/2>
5	230.9	8.95	6.55	3.19	0.23 |±9/2> + 0.21 |±13/2> + 0.20 |±5/2> + 0.12 |±3/2>
6	273.5	2.48	3.22	13.56	0.25 |±1/2> + 0.23 |±3/2> + 0.17 |±7/2> + 0.12 |±11/2> + 0.10 |±9/2>
7	327.2	0.32	0.63	18.43	0.27 |±5/2> + 0.21 |±7/2> + 0.17 |±9/2> + 0.15 |±3/2> + 0.11 |±11/2>
8	652.7	0.01	0.01	19.76	0.43 |±1/2> + 0.31 |±3/2> + 0.16 |±5/2>

* Only the contributions ≥ 10% are given.