

Supporting Information for

A Single Molecule Magnet Behaviour in a D_{3h} Symmetry Dy(III) Complex Involving a Quinone-Tetrathiafulvalene-Quinone Bridge

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Absorption spectra were recorded on a Varian Cary 5000 UV-Visible-NIR spectrometer. The dc and ac magnetic susceptibility measurements were performed on grinded single crystals pelletized in Teflon tape with a Quantum Design MPMS-XL SQUID magnetometer. For χ_{MT} vs. T the applied magnetic field was equal 0.2 kOe between 2 and 20 K, 2 kOe between 20 and 70 K and 10 kOe above 70 K. The experimental data have been corrected from the diamagnetism of the sample holder, and the intrinsic diamagnetism of the materials was evaluated with Pascal's tables.

To perform satisfying electrochemistry, DMF and MeCN solvents must be used leading to the dissociation of the coordination complexes due to their weak stability in such solvents. Thus the results were similar than those obtained for the free ligand and previously published in *Tetrahedron* **2010**, *66*, 7605.

Single crystals of **1** and **2** were mounted on a Nonius four circles diffractometer equipped with a CCD camera and a graphite monochromated MoK α radiation source ($\lambda = 0.71073 \text{ \AA}$) while single crystal of **3** was mounted on a APEXII Bruker-AXS diffractometer for data collection (MoK α radiation source, $\lambda = 0.71073 \text{ \AA}$), from the Centre de Diffractométrie (CDIFX), Université de Rennes 1, France. Structures were solved with a direct method using the SIR-97 program and refined with a full matrix least-squares method on F² using the SHELXL-97 program.¹

[1] SHELX97 - Programs for Crystal Structure Analysis (Release 97-2). G. M. Sheldrick, Institut für Anorganische Chemie der Universität, Tammanstrasse 4, D-3400 Göttingen, Germany, 1998. SIR97 - A. Altomare, M. C. Burla, M. Camalli, G. L. Casciarano, C. Giacovazzo, A. Guagliardi, A. G. G. Moliterni, G. Polidori, R. Spagna, *J. Appl. Cryst.* **1999** *32*, 115-119.

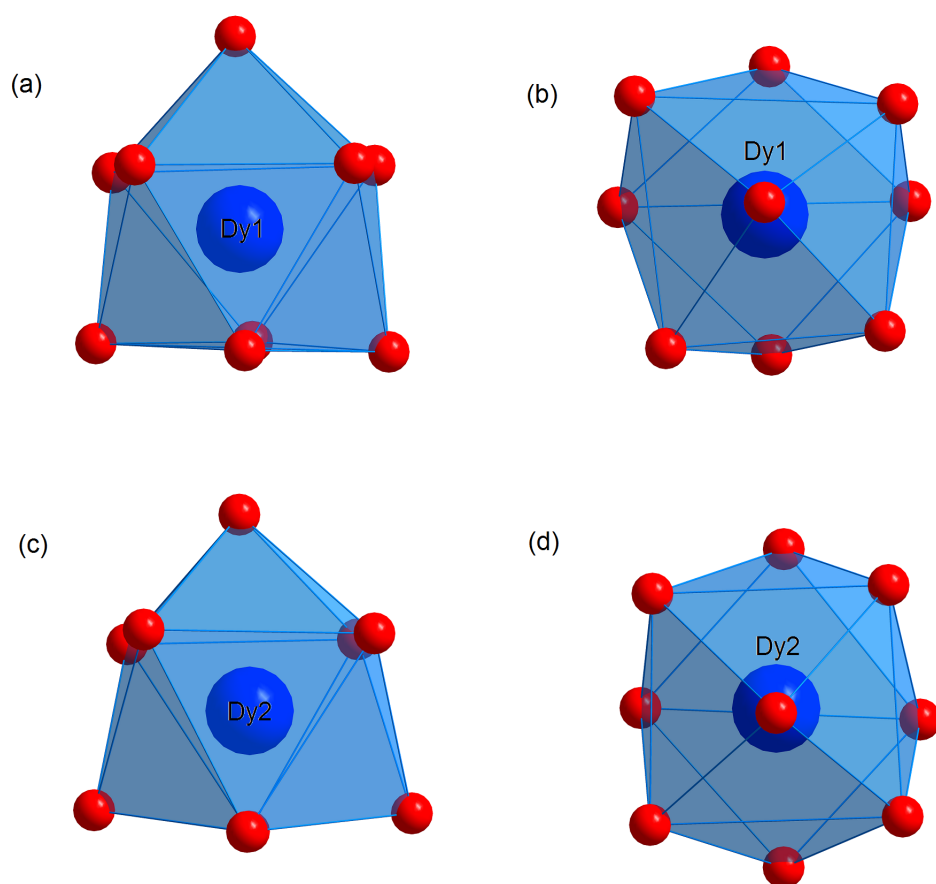


Fig. S1 Coordination polyhedron along the three fold axis and four fold axis for the Dy1 (a) and (b), and for the Dy2 (c) and (d).

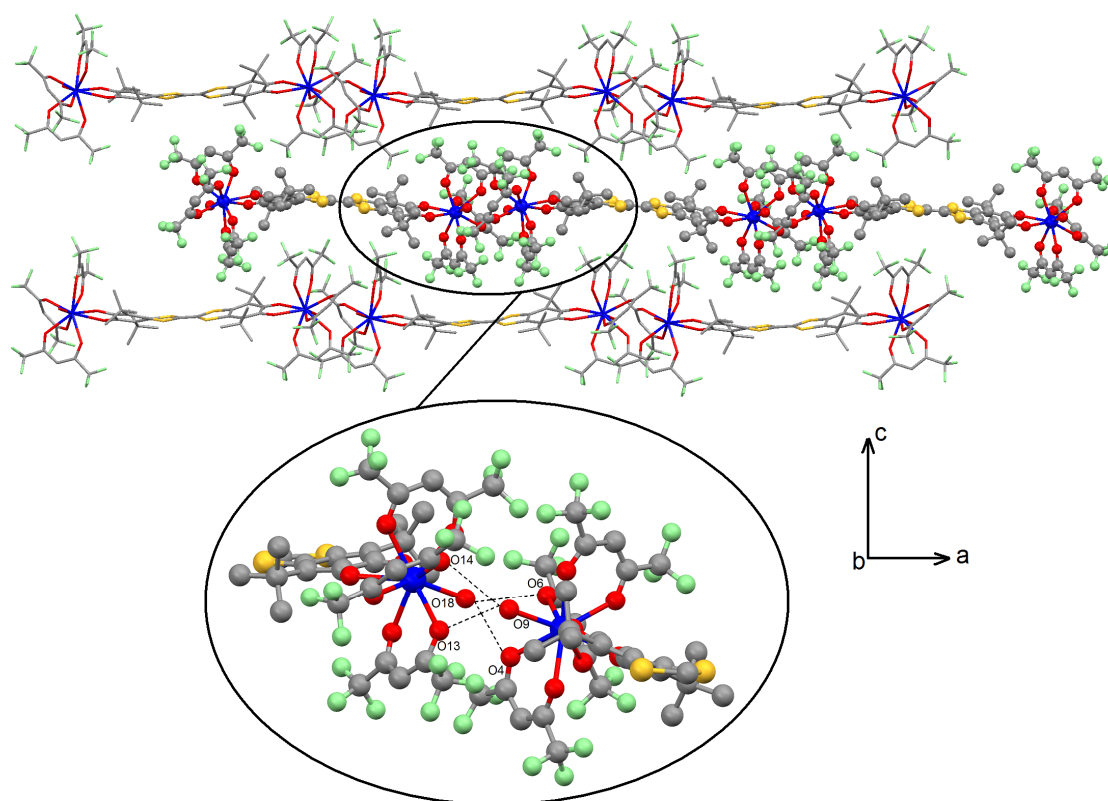


Fig. S2 Crystal packing of the dinuclear complexes in **1** highlighting the formation of pseudo-dimers (a) and chains (b). Hydrogen atoms are omitted for clarity.

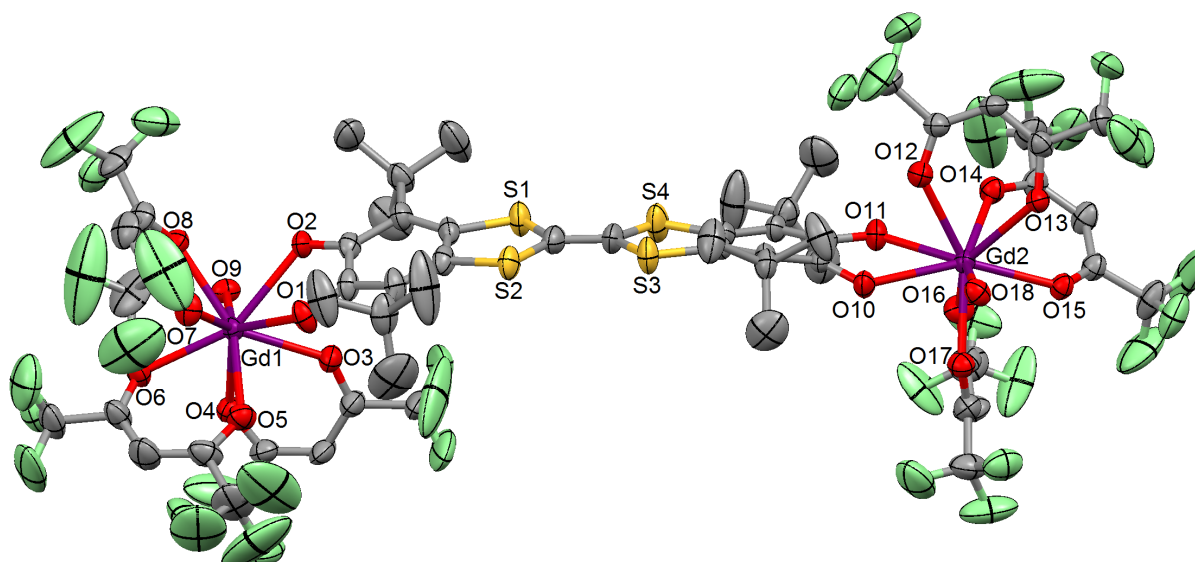


Fig. S3 ORTEP view of the dinuclear complex **2**. Thermal ellipsoids are drawn at 30% probability. Hydrogen atoms are omitted for clarity.

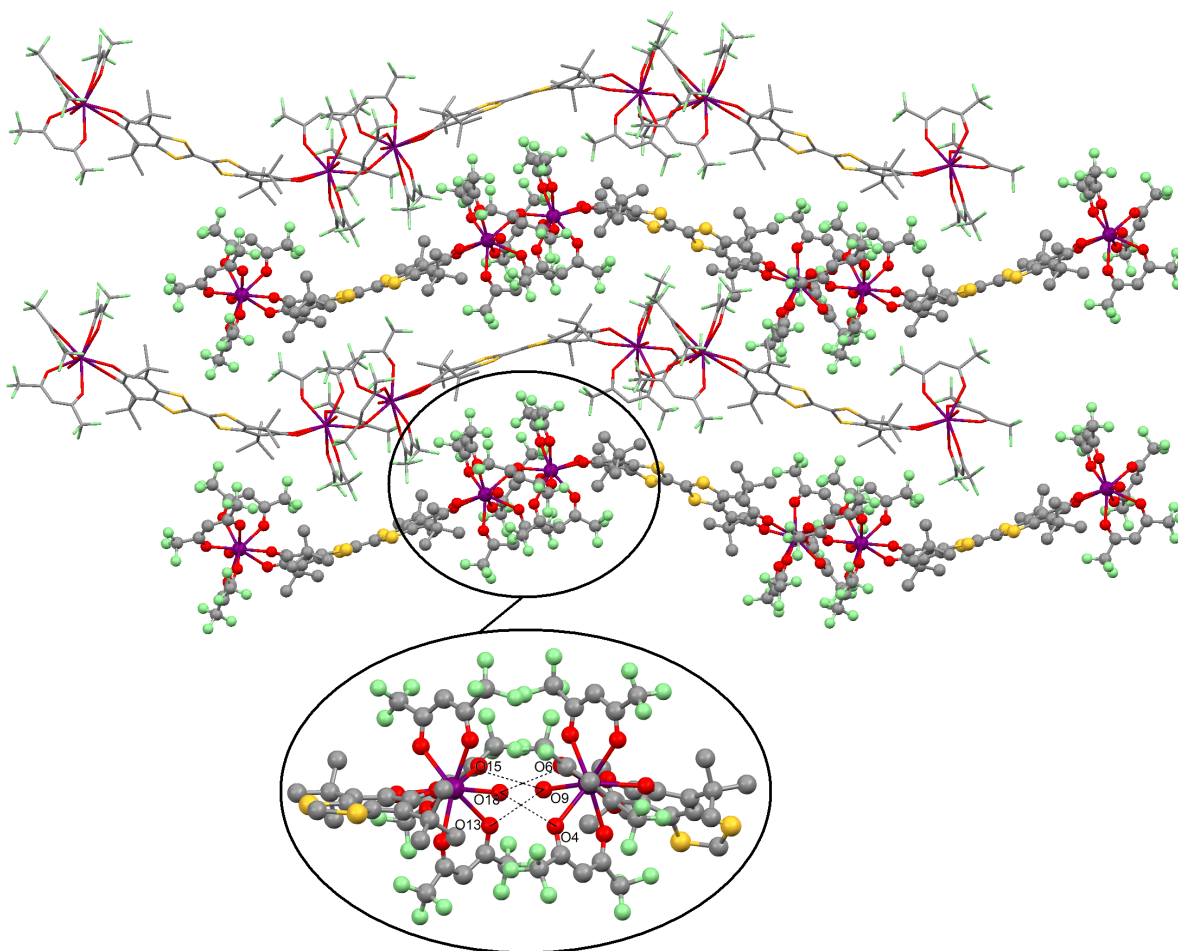
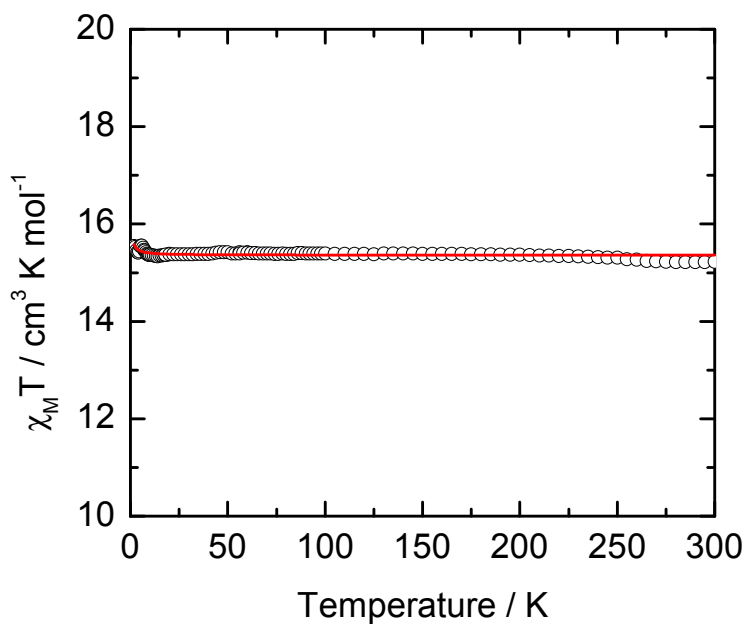
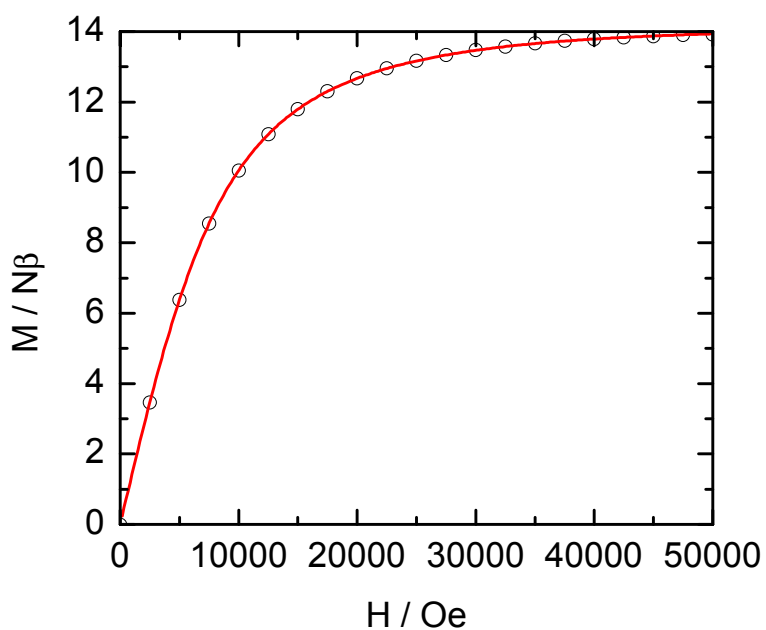


Fig. S4 Packing crystal of **2** highlighting the formation of pseudo-dimers and chains.



a)



b)

Fig. S5 Temperature dependence of $\chi_M T$ for **2** (circles) with best fitted curve (red line) with Curie law ($\chi_M T = (N\beta^2/3k)g^2(63/4)$ ($g=1.97$)) (a). Field dependence of the magnetization for **2** (circles) recorded at 2 K with the best fitted curve with the Brillouin function ($g=2.01$) (b).

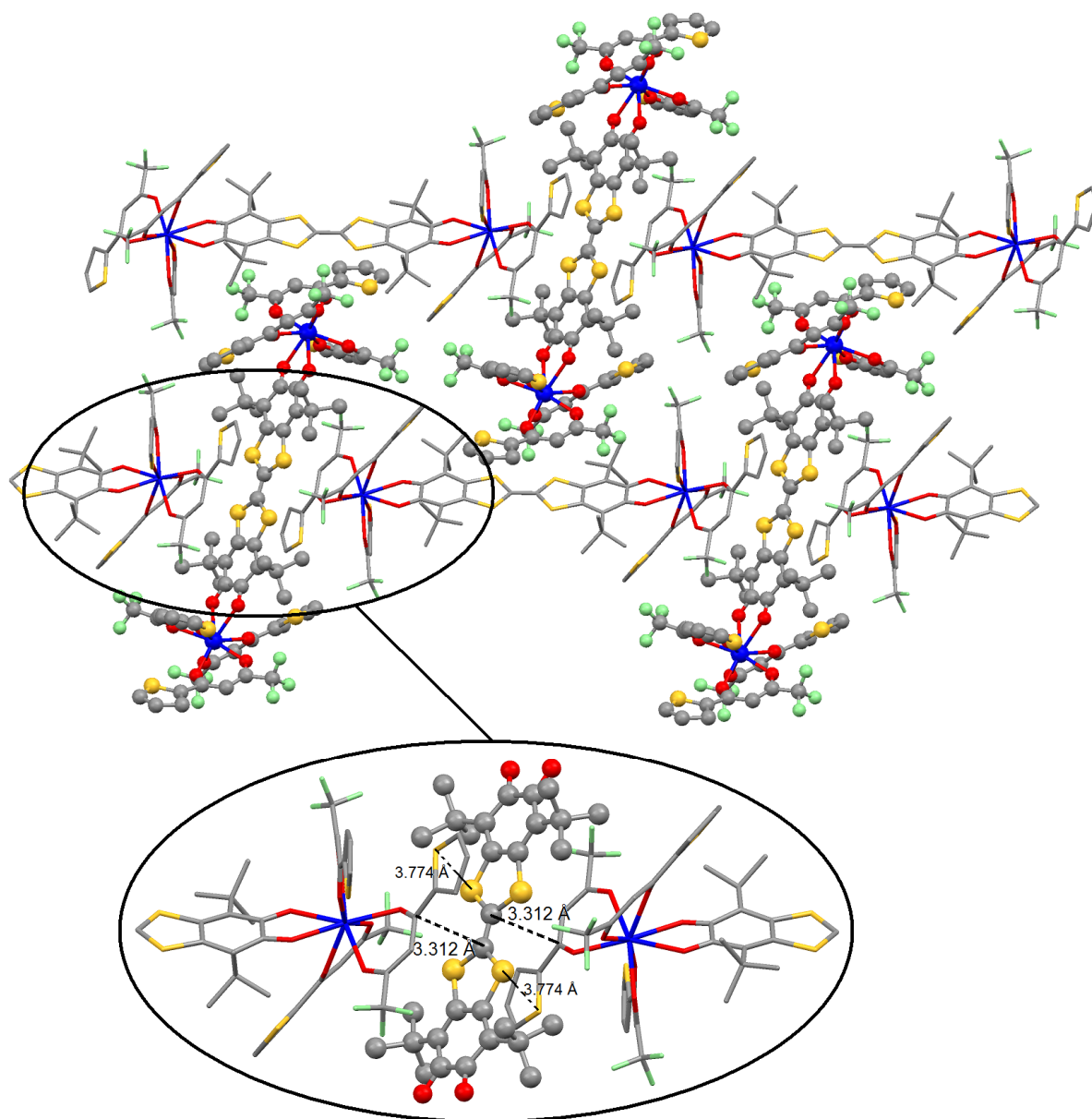


Fig. S6 Packing crystal of **3** highlighting the short contacts between the tta⁻ anion and the TTF core.

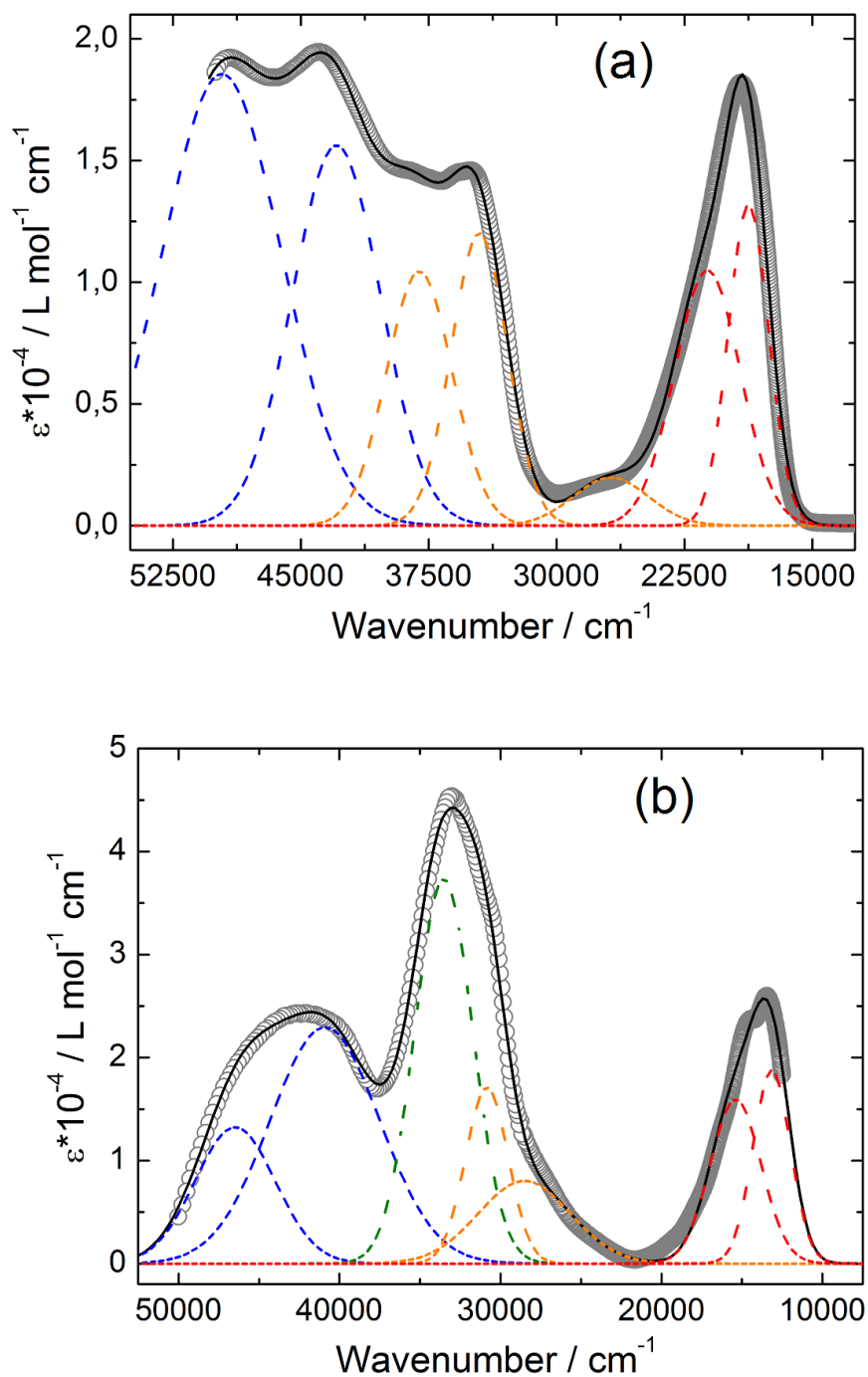


Fig. S7 Experimental *n*-heptane solution ($4 \cdot 10^{-5}$ M) UV-visible absorption spectra (open circles), respective Gaussian deconvolutions (dashed lines) and best fit (full black line) $R = 0.9983$ (for **L**) (a), $R = 0.9960$ (for **1**) (b).

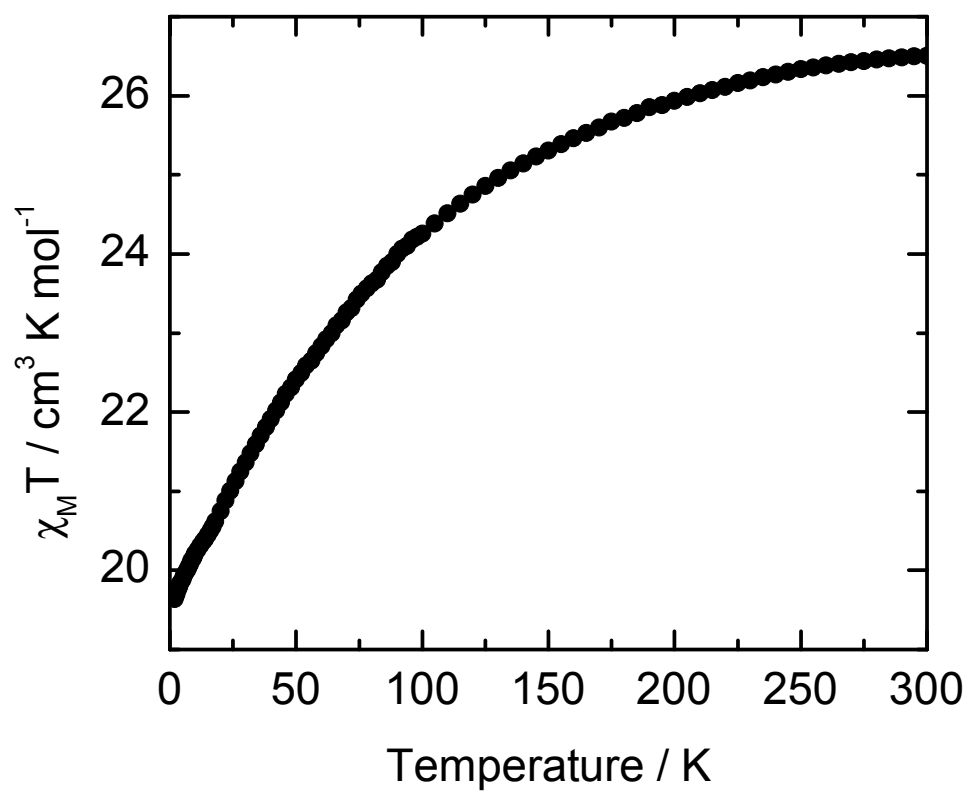
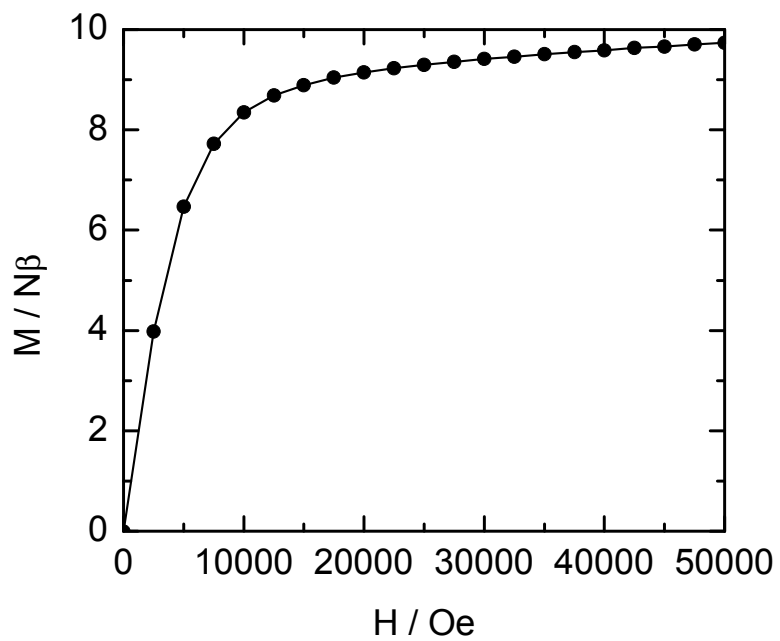
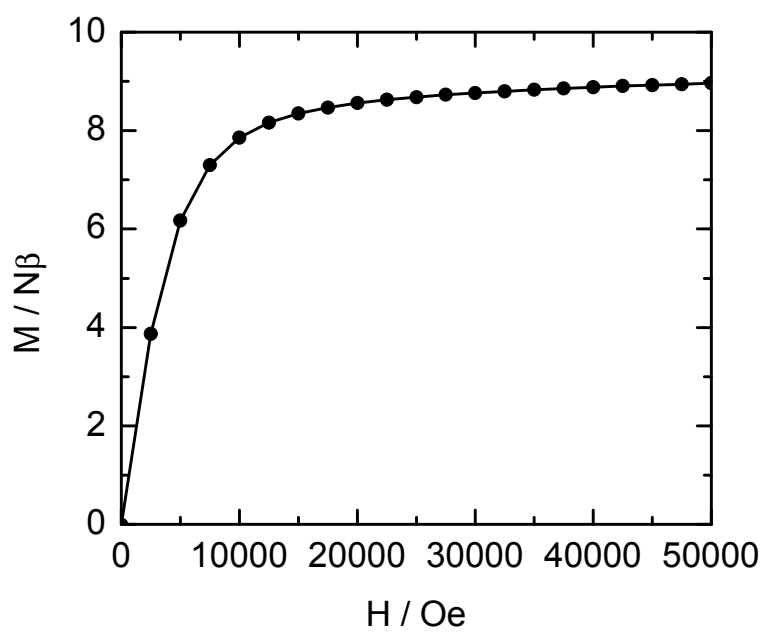


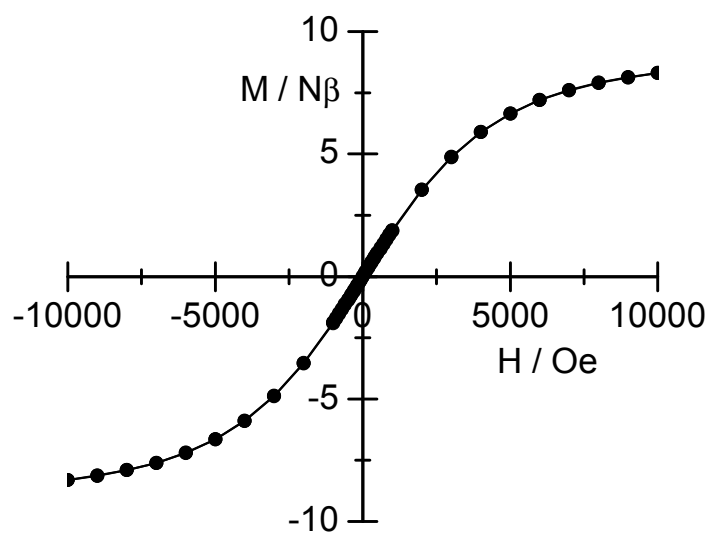
Fig. S8 Temperature dependences of $\chi_M T$ for **3**.



a)



b)



c)

Fig. S9 Field dependence of the magnetization at 2 K for **1** (a), **3** (b). Magnetic hysteresis loop for compound **1** at 1.8 K.

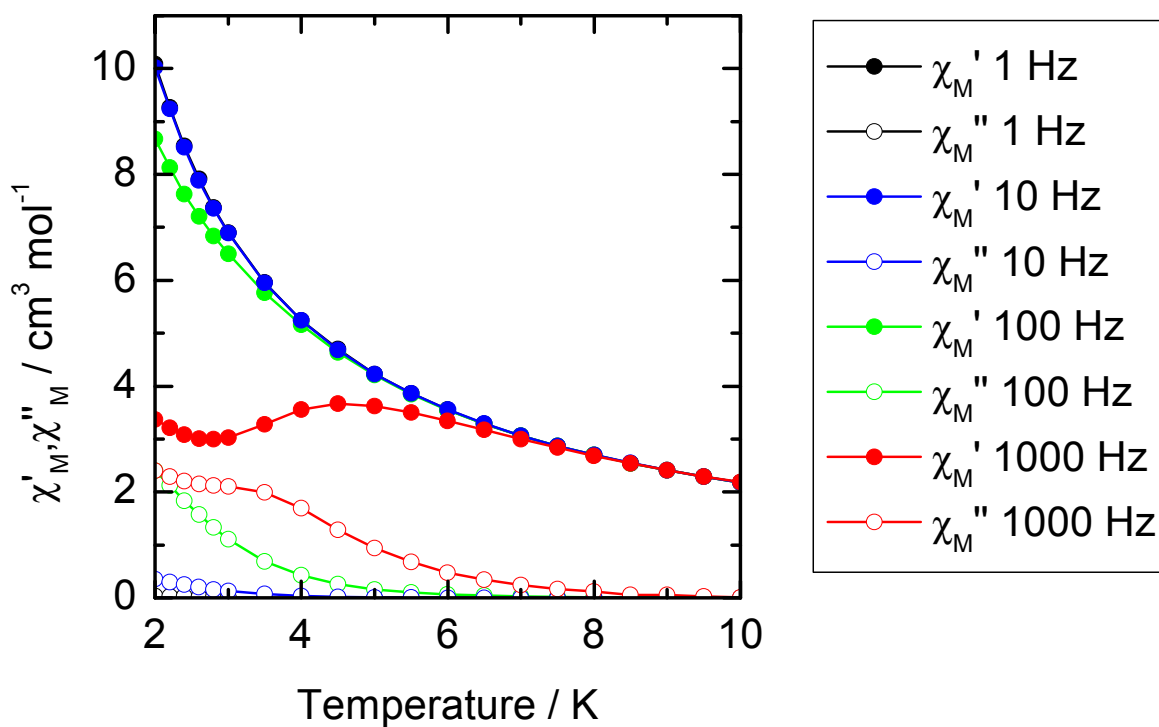


Fig. S10 Temperature dependence of the in-phase (χ_M') and out-of-phase (χ_M'') components of the magnetic molar ac susceptibility at 1, 10, 100 and 1000 Hz for compound **1**.

Extended Debye model:

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

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

With χ_T the isothermal susceptibility and χ_S the adiabatic susceptibility. The extended Debye model was applied to fit simultaneously the experimental χ' and χ'' data around the maximum in χ . The best fitted parameters χ_T , χ_S , α and τ are listed in Table S2. The best fitted curves (red lines) at 1.8 K are represented here below.

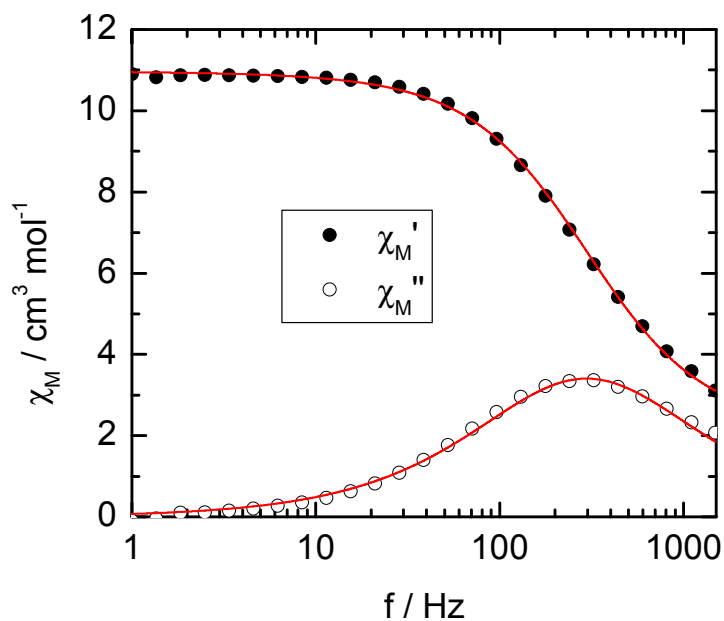


Table S1. Selected bond lengths for compounds **1-3**.

	1	2	3
Ln-O1	2.435(5)	2.464(5)	2.440(7)
Ln-O2	2.508(5)	2.516(5)	2.501(7)
Ln-O3	2.364(5)	2.438(5)	2.341(8)
Ln-O4	2.498(5)	2.512(6)	2.307(8)
Ln-O5	2.328(5)	2.353(6)	2.331(8)
Ln-O6	2.336(5)	2.386(5)	2.326(8)
Ln-O7	2.384(5)	2.398(6)	2.334(7)
Ln-O8	2.393(5)	2.382(5)	2.316(7)
Ln-O9	2.397(5)	2.403(5)	
Ln-O10	2.523(5)	2.561(5)	
Ln-O11	2.414(4)	2.490(5)	
Ln-O12	2.357(5)	2.436(6)	
Ln-O13	2.469(5)	2.462(5)	
Ln-O14	2.361(5)	2.349(5)	
Ln-O15	2.327(5)	2.397(5)	
Ln-O16	2.372(6)	2.410(6)	
Ln-O17	2.378(5)	2.390(6)	
Ln-O18	2.396(5)	2.410(5)	

Table S2. Best fitted parameters (χ_T , χ_S , τ and α) with the extended Debye model for compound **1** between 1.8 and 3.8 K measured in zero external field. The agreement factor R^2 is given in the last column.

T / K	$\chi_T / \text{cm}^3 \text{mol}^{-1}$	$\chi_S / \text{cm}^3 \text{mol}^{-1}$	α	τ / s	R^2
3.8	5.21916	0.73913	0.18177	1.12424E-4	0.99994
3.7	5.33652	0.71721	0.18479	1.25555E-4	0.99998
3.6	5.4681	0.84169	0.1805	1.50354E-4	0.99994
3.5	5.59975	0.85829	0.18593	1.73259E-4	0.99995
3.4	5.74073	0.77883	0.19142	1.93566E-4	0.99995
3.3	5.8893	0.92233	0.19227	2.31327E-4	0.99995
3.2	6.04372	0.98502	0.19461	2.72528E-4	0.99995
3.1	6.21491	1.02141	0.20136	3.20085E-4	0.99994
3	6.39942	0.98344	0.21534	3.64708E-4	0.99987
2.9	6.5801	1.07225	0.21904	4.37674E-4	0.9999
2.8	6.80112	1.041	0.23605	5.05916E-4	0.99981
2.7	7.01008	1.07755	0.24654	6.00003E-4	0.9998
2.6	7.21627	1.14812	0.25234	7.19144E-4	0.99975
2.5	7.50814	1.12423	0.27586	8.48863E-4	0.99974
2.4	7.75769	1.19329	0.28481	0.00102	0.99968
2.3	8.01451	1.2705	0.29172	0.00123	0.99948
2.2	8.42436	1.24972	0.3225	0.00152	0.99969
2.1	8.69167	1.26333	0.33663	0.00175	0.99964
2	9.10178	1.29649	0.35362	0.00213	0.99954
1.9	9.52387	1.31592	0.36916	0.00255	0.99941
1.8	9.98038	1.33018	0.38624	0.00305	0.99927

Table S3. Best fitted parameters (χ_T , χ_S , τ and α) with the extended Debye model for compound **1** between 1.8 and 3.8 K measured at 1400 Oe. The agreement factor R^2 is given in the last column.

T / K	$\chi_T / \text{cm}^3 \text{mol}^{-1}$	$\chi_S / \text{cm}^3 \text{mol}^{-1}$	α	τ / s	R^2
3.8	5.44792	1.163	0.09658	1.03E-04	0.99998
3.7	5.58262	1.09687	0.10155	1.12E-04	0.99998
3.6	5.73178	1.17004	0.09933	1.27E-04	0.99996
3.5	5.88575	1.21196	0.10277	1.41E-04	0.99996
3.4	6.05055	1.23573	0.10575	1.56E-04	0.99996
3.3	6.22319	1.28582	0.10181	1.75E-04	0.99996
3.2	6.41065	1.28501	0.11251	1.92E-04	0.99994
3.1	6.60871	1.3544	0.11317	2.16E-04	0.99994
3	6.82013	1.36851	0.11898	2.37E-04	0.99993
2.9	7.04311	1.43875	0.1178	2.62E-04	0.9999
2.8	7.28368	1.41947	0.12692	2.83E-04	0.99988
2.7	7.54199	1.52861	0.12858	3.15E-04	0.99988
2.6	7.8187	1.56153	0.13354	3.42E-04	0.99986
2.5	8.1237	1.60823	0.14014	3.70E-04	0.99983
2.4	8.45131	1.64421	0.14708	3.95E-04	0.99979
2.3	8.81841	1.68777	0.15339	4.23E-04	0.99976
2.2	9.22159	1.79157	0.15488	4.54E-04	0.9998
2.1	9.52261	1.83591	0.15794	4.70E-04	0.99977
2	9.97611	1.91648	0.16229	4.96E-04	0.99974
1.9	10.45902	2.01722	0.16461	5.19E-04	0.99977
1.8	10.96885	2.11901	0.16501	5.38E-04	0.99968