Supporting information for

Dysprosium Single-Molecule Magnets Involving 1,10-Phenantroline-5,6-dione Ligand

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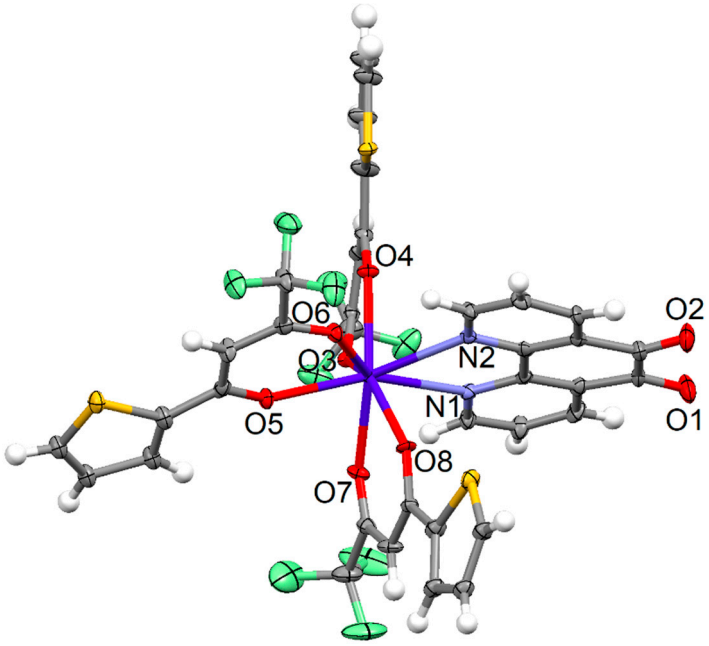

Figure S1. ORTEP view of **1**. Thermal ellipsoids are drawn at 30% probability.

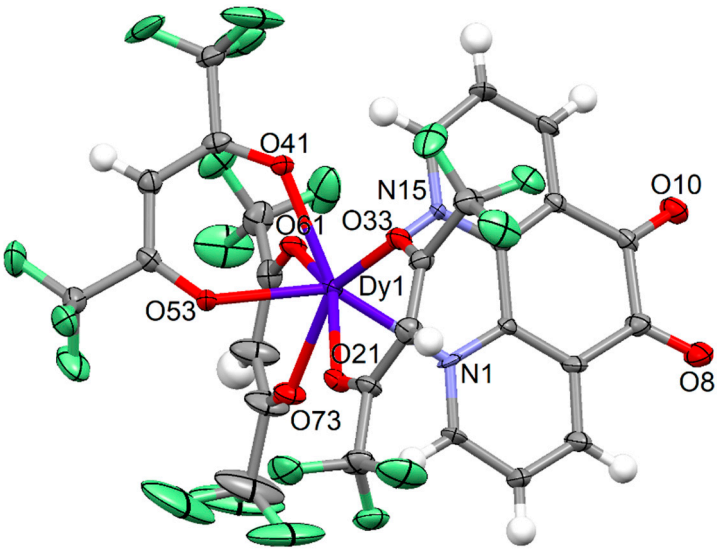

Figure S2. ORTEP view of **2**. Thermal ellipsoids are drawn at 30% probability.

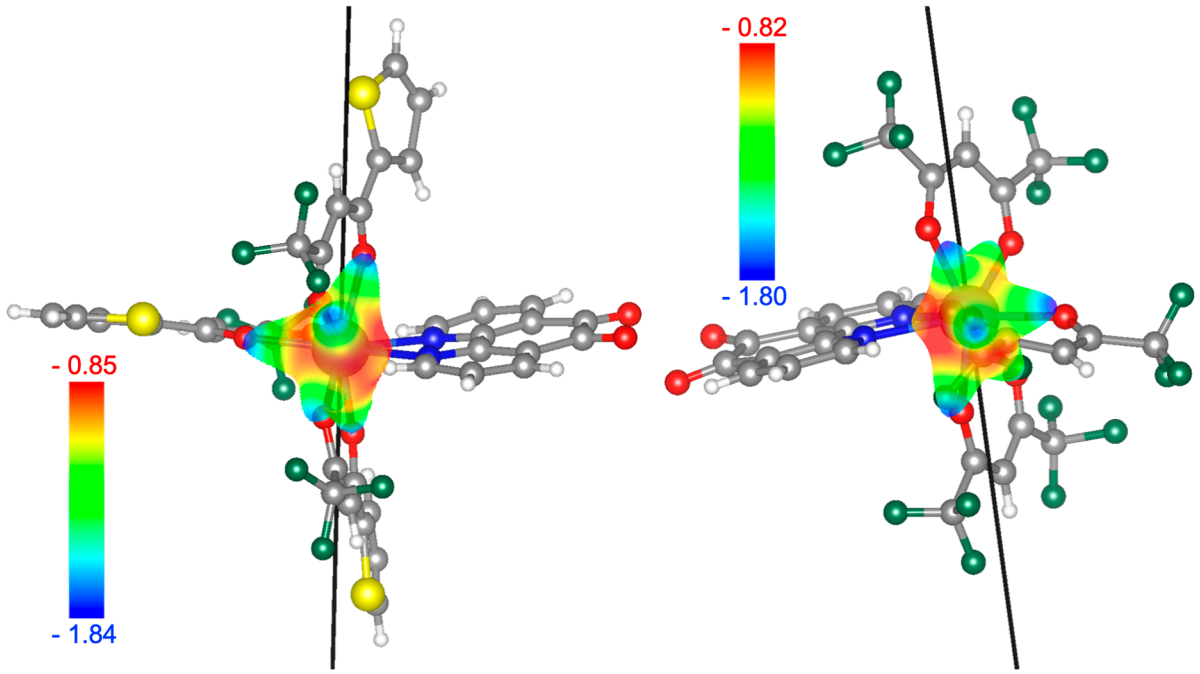

Figure S3. Molecular Electrostatic potential centered on the lanthanide ion for **1** (left) and **2** (right). The orientation of the ground state anisotropy axis is represented as a dark line.

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 ν of the oscillating field ($\omega = 2\pi\nu$). Typically, only the temperatures for which a maximum on the χ_M'' vs. ν curves, have been considered (see figure S3 here below for an example). The best fitted parameters τ, α, χ_T, χ_S are listed in Table S2 to S7 with the coefficient of determination R^2.

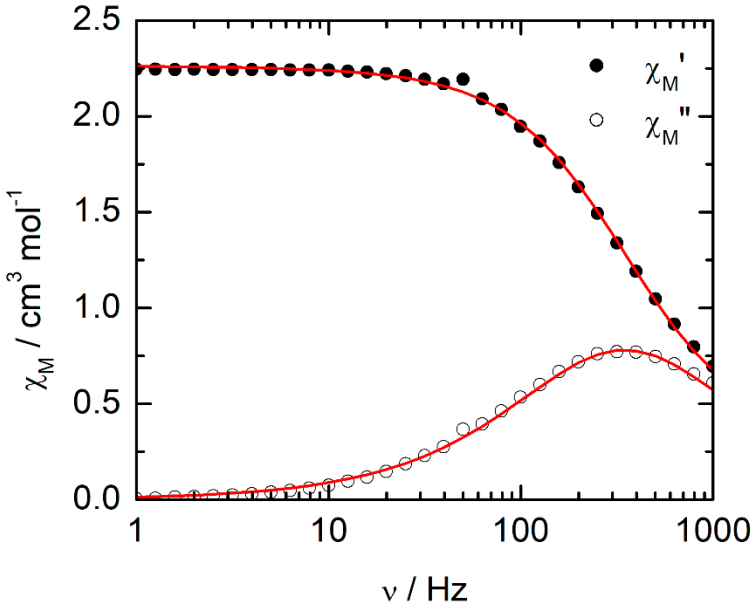

Figure S4. Frequency dependence of in-phase (χ_M') and out-of-phase components (χ_M") at 5 K for **1** in zero applied magnetic field. Red lines are the best fitted curves.

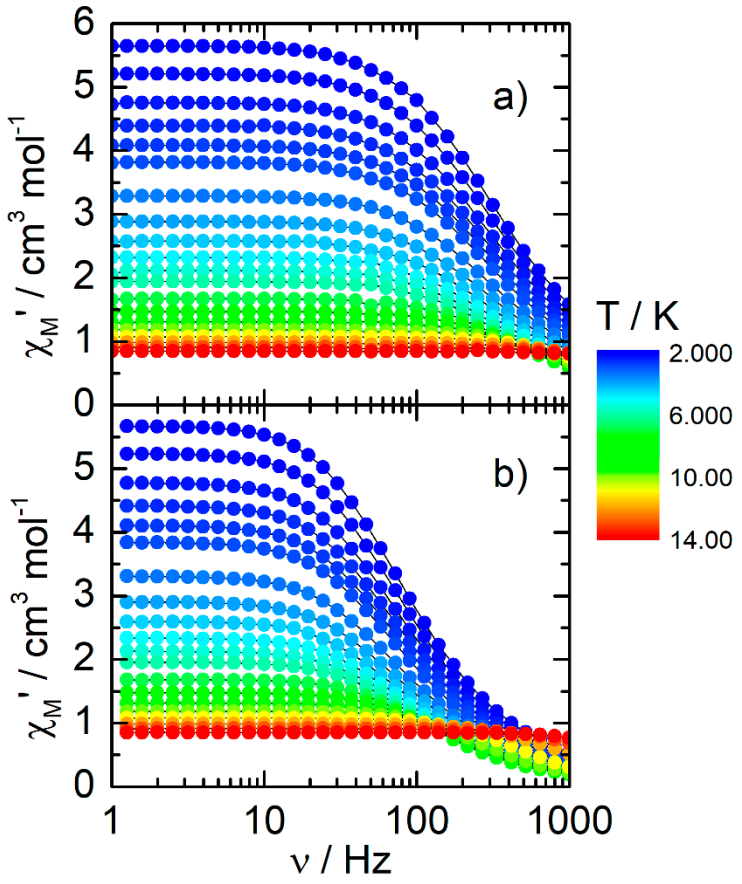

Figure S5. Frequency dependence of χ_M' in the temperature range 2-14 K for **1** (a) and **2** (b) in zero applied magnetic field.

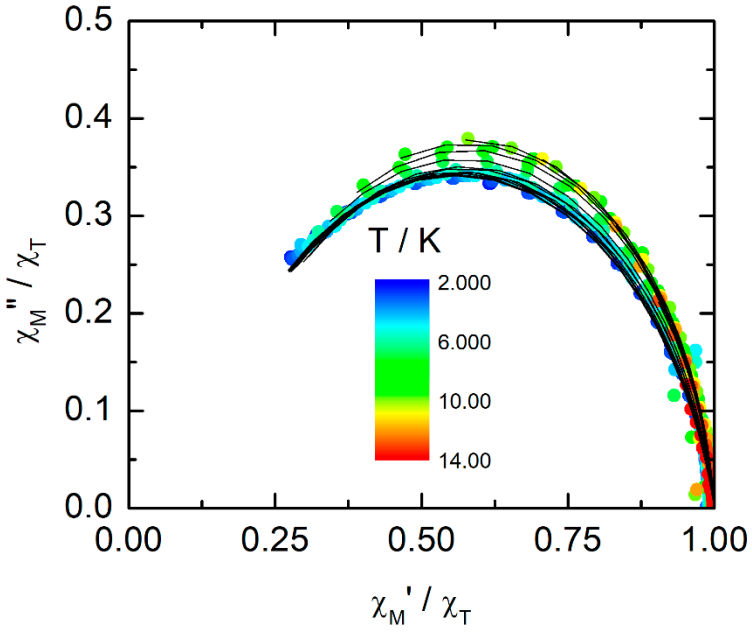

Figure S6. Normalized Argand plot between 2 and 14 K for **1** in zero applied magnetic field.

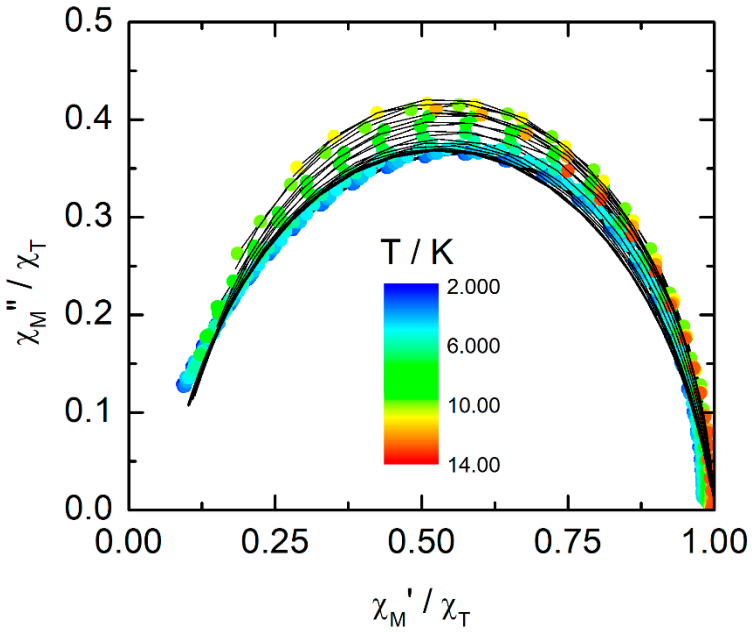

Figure S7. Normalized Argand plot between 2 and 14 K for **2** in zero applied magnetic field.

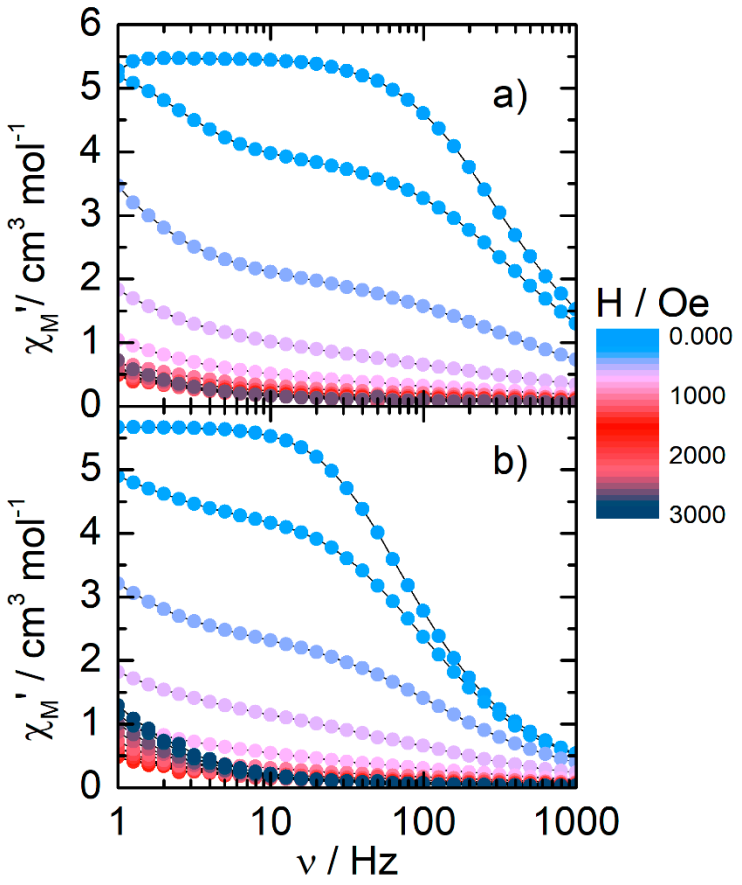

Figure S8. Frequency dependence of χ_M' at 2 K at various external fields for **1** (a) and **2** (b).

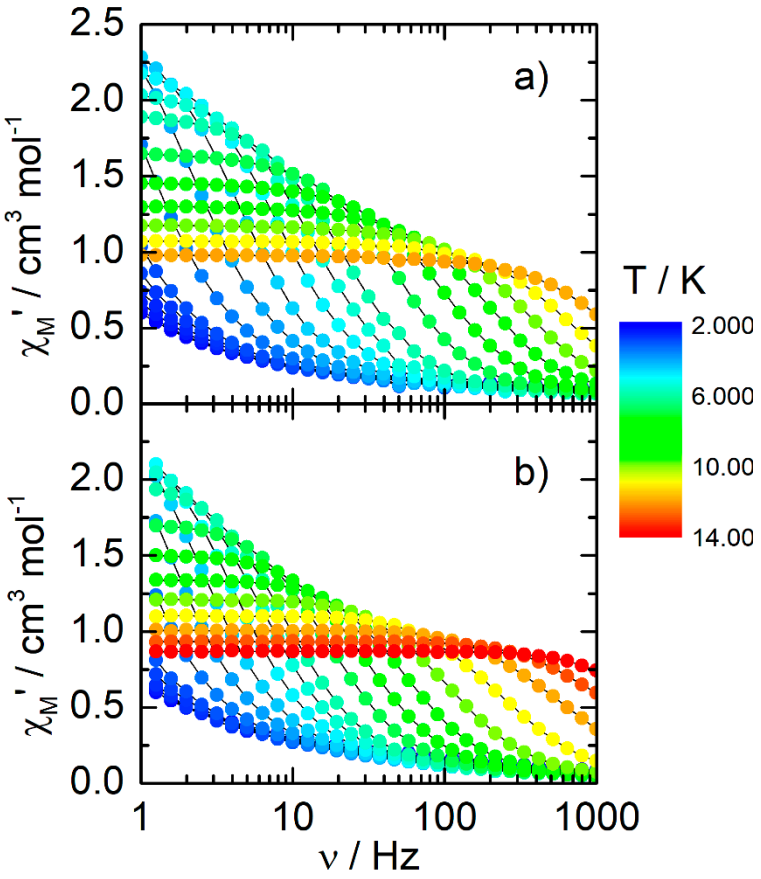

Figure S9. Frequency dependence of χ_M' in the temperature range 2-14 K for **1** (a) and for **2** (b) in 1200 and 1000 Oe applied magnetic field respectively.

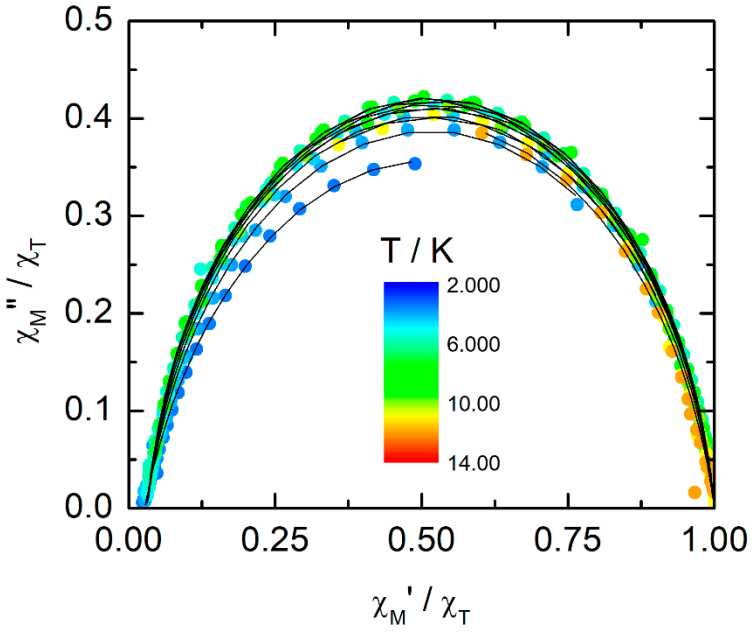

Figure S10. Normalized Argand plot between 2 and 14 K for **1** in 1200 Oe applied magnetic field.

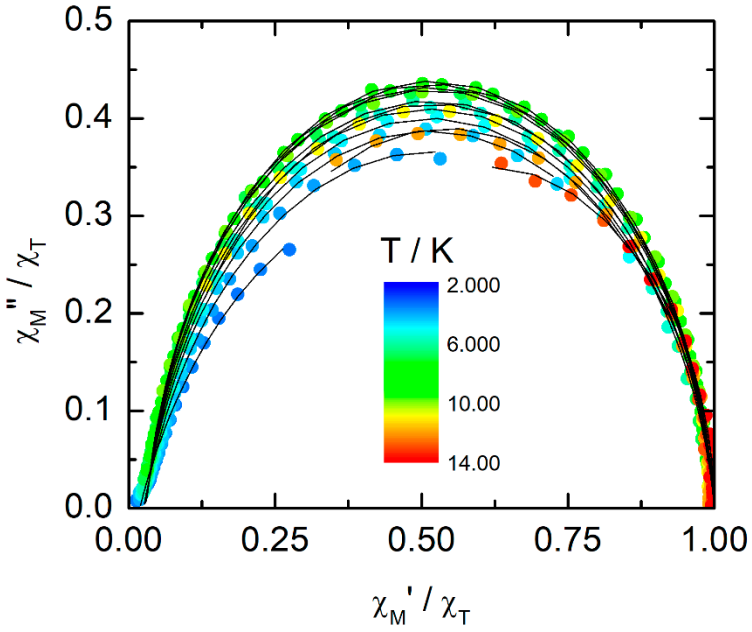

Figure S11. Normalized Argand plot between 2 and 14 K for **2** in 1000 Oe applied magnetic field.

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

Compound	[Dy(tta)$_3$(**L**)] (**1**)	[Dy(hfac)$_3$(**L**)] (**2**)
Formula	$C_{36}H_{18}DyF_9O_8N_2S_3$	$C_{27}H_9DyF_{18}O_8N_2$
M / g.mol^{-1}	1036.2	993.9
Crystal system	Tetragonal	Tetragonal
Space group	I4 (N°79)	I-4 (N°82)
Cell parameters	a = 20.4956(18) Å b = 20.4956 Å c = 19.670(2) Å	a = 19.478(2) Å b = 19.478 Å c = 18.374(2) Å
Volume / Å^3	8262.9(17)	6971.0(19)
Z	8	8
T / K	150(2)	150(2)
2θ range /°	4.14 ≤ 2θ ≤ 55.10	6.10 ≤ 2θ ≤ 54.85
ρ_{calc} / g.cm^{-3}	1.666	1.894
μ / mm^{-1}	2.051	2.289
Number of reflections	25394	29549
Independent reflections	9339	7946
R_{int}	0.0446	0.0458
$Fo^2 > 2\sigma(Fo)^2$	8119	7170
Number of variables	517	505
R_1, ωR_2	0.0543, 0.1538	0.0581, 0.1038

Table S2. Computed energies (cm^{-1}), *g*-tensor and wavefunction composition (only contributions above 10% are reported here) of the ground state multiplet $^6H_{15/2}$ in the effective spin ½ model for **1**.

KD	**E**	***g***	**wavefunction**
1	0.0	0.00 0.01 19.43	92% |± 15/2>
2	160.1	0.31 0.35 15.79	76% |± 13/2> + 22% |± 9/2>
3	224.7	1.64 2.25 16.12	34% |± 1/2> + 27% |± 3/2> + 12% |± 5/2> + 12% |± 11/2>
4	257.5	8.15 5.41 0.13	40% |± 7/2> + 26% |± 11/2> + 19% |± 1/2>

5	286.9	3.00 6.21 12.18	49% |± 5/2> + 28% |± 3/2>
6	378.1	0.07 0.32 18.41	29% |± 9/2> + 26% |± 11/2> + 16% |± 7/2> + 10% |± 13/2>
7	404.1	0.05 0.33 15.53	24% |± 9/2> + 17% |± 11/2> + 15% |± 3/2> + 15% |± 1/2> + 11% |± 7/2>
8	454.5	0.12 0.23 17.99	22% |± 7/2> + 20% |± 5/2> + 18% |± 1/2> + 16% |± 3/2> + 13% |± 9/2>

Table S3. Computed energies (cm^{-1}), *g*-tensor and wavefunction composition (only contributions above 10% are reported here) of the ground state multiplet $^{6}H_{15/2}$ in the effective spin ½ model for **2**.

KD	**E**	***g***	**wavefunction**
1	0.0	0.01 0.01 19.56	95% |± 15/2>
2	135.1	0.31 0.74 17.32	29% |± 1/2> + 26% |± 3/2> + 18% |± 5/2> + 12% |± 13/2>
3	168.0	1.54 2.99 12.53	59% |± 13/2> + 20% |± 9/2>
4	224.5	8.71 7.47 4.75	35% |± 7/2> + 34% |± 11/2> + 13% |± 5/2>
5	276.0	1.89 2.99 12.06	31% |± 9/2> + 21% |± 11/2> + 14% |± 5/2> + 12% |± 7/2>

6	302.0	0.93 3.85 13.56	34% |± 5/2> + 25% |± 7/2> + 20% |± 3/2>
7	377.8	0.01 0.17 19.24	49% |± 1/2> + 33% |± 3/2> + 12% |± 5/2>
8	436.0	0.02 0.10 19.55	30% |± 11/2> + 29% |± 9/2> + 16% |± 7/2> + 14% |± 13/2>

Table S4. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model **1** at 0 Oe in the temperature range 2-13 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
2	5.68478	0.7481	0.15942	5.19486E-04	0.99982
2.2	5.23992	0.70231	0.15776	5.21130E-04	0.99986
2.4	4.77969	0.63939	0.15718	5.20433E-04	0.99982
2.6	4.42086	0.59875	0.15809	5.22492E-04	0.99982
2.8	4.10927	0.56588	0.1557	5.20870E-04	0.99985
3	3.84009	0.53542	0.15552	5.18593E-04	0.99984
3.5	3.30845	0.47929	0.14725	5.06964E-04	0.99985
4	2.90548	0.42707	0.14658	4.95959E-04	0.99985
4.5	2.59485	0.37434	0.14569	4.82237E-04	0.99976
5	2.33614	0.36695	0.13695	4.73086E-04	0.99973
5.5	2.12305	0.35318	0.12362	4.53914E-04	0.99964
6	1.95125	0.32514	0.12085	4.26822E-04	0.99978
7	1.67423	0.2788	0.10891	3.62803E-04	0.99968
8	1.46852	0.26842	0.08239	3.00001E-04	0.99989
9	1.30945	0.24234	0.07397	2.30505E-04	0.9998
10	1.17926	0.23524	0.063	1.68926E-04	0.99962
11	1.07484	0.20672	0.07396	1.12414E-04	0.99993
12	0.98077	0.16323	0.0922	6.63012E-05	0.99992
13	0.90809	0.23663	0.08225	4.80595E-05	0.99984

Table S5. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model **2** at 0 Oe in the temperature range 2-13 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
2	5.33323	0.33291	0.15023	0.00183	0.99913
2.2	4.86618	0.29727	0.15194	0.00184	0.99908
2.4	4.49975	0.27917	0.15206	0.00184	0.99911
2.6	4.18517	0.26063	0.15221	0.00185	0.99911
2.8	3.91344	0.24312	0.15256	0.00184	0.99911
3	3.36901	0.21454	0.15155	0.00182	0.99911
3.5	2.96029	0.1913	0.14938	0.00177	0.99907
4	2.64647	0.16923	0.14699	0.00172	0.99894
4.5	2.37575	0.16337	0.13883	0.00165	0.99912
5	2.16233	0.15559	0.1317	0.00156	0.9992
5.5	1.98471	0.14534	0.12515	0.00145	0.99922
6	1.70313	0.13254	0.10774	0.00123	0.99934
7	1.48981	0.12193	0.08961	0.00101	0.99956
8	1.32414	0.11321	0.07345	0.00078	0.99971
9	1.19234	0.09683	0.06046	0.00055	0.99984
10	1.0841	0.08647	0.05558	0.00033	0.99992
11	0.98807	0.10569	0.05694	0.00018	0.99995
12	0.91498	0.17828	0.04026	0.000104	0.99998
13	5.33323	0.33291	0.15023	0.00183	0.99913

Table S6. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model **1** at 1200 Oe in the temperature range 3.5-12 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
3.5	3.49259	0.09511	0.19485	0.176280	0.99958
4	2.87914	0.08648	0.14248	0.070770	0.99972
4.5	2.53814	0.08013	0.11674	0.035930	0.9997
5	2.2803	0.07203	0.10464	0.019670	0.99996
5.5	2.07444	0.06565	0.09405	0.011780	0.99988
6	1.91118	0.05949	0.09374	0.007430	0.99995
7	1.64764	0.05218	0.08861	0.003330	0.99994
8	1.45077	0.0438	0.09026	0.001640	0.99993
9	1.29703	0.03949	9.94E-02	0.000866	0.9998
10	1.17149	0.0381	1.03E-01	0.000465	0.99986
11	1.06906	0.03464	1.19E-01	0.000249	0.99988

12	0.97632	0.01131	1.40E-01	0.000118	0.99978

Table S7. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model **2** at 1000 Oe in the temperature range 3.5-14 K.

T / K	χ_T / cm^3 mol^{-1}	χ_S / cm^3 mol^{-1}	α	τ / s	R^2
3.5	4.51369	0.08456	0.25282	0.35564	0.99694
4	3.24405	0.08284	0.1802	0.12244	0.99843
4.5	2.76546	0.07581	0.14382	0.06483	0.99887
5	2.45856	0.07056	0.1229	0.03865	0.99953
5.5	2.22053	0.06458	0.10722	0.02498	0.99963
6	2.02865	0.06083	0.09423	0.01703	0.99974
7	1.73473	0.05287	0.07715	0.00883	0.99979
8	1.51677	0.04922	0.06737	0.00499	0.99984
9	1.34887	0.04766	0.06092	0.00282	0.99989
10	1.2173	0.04092	0.07123	0.00147	0.99986
11	1.10983	0.04729	0.09053	0.000681	0.9998
12	1.01143	0.08682	0.10109	0.000289	0.99979
13	0.93475	0.21432	0.0608	0.000156	0.99992
14	0.86881	0.28254	0.0177	0.000082	0.99998