Supporting Information for

Study of the nuclear spin and dilution influence over the slow relaxation in a 3d4f heterobimetallic Single-Molecule Magnet

Jessica Flores González, Bertrand Lefeuvre, Bastien Degraeve, Olivier Cador,* Fabrice Pointillart*

Univ Rennes, CNRS, ISCR (Institut des Sciences Chimiques de Rennes) - UMR 6226, 35000 Rennes, France

Table S1. Crystallographic data for **$DyZn_2$**, **$^{162}DyZn_2$**, **$^{163}DyZn_2$**, **$(Dy@Y)Zn_2$**, **$^{162}(Dy@Y)Zn_2$** and **$^{163}(Dy@Y)Zn_2$**.

Compounds	$[(Zn\mathbf{L})_2DyCl_3]\cdot 2H_2O]$ **$DyZn_2$**	$[(Zn\mathbf{L})_2{}^{162}DyCl_3]\cdot 2H_2O]$ **$^{162}DyZn_2$**	$[(Zn\mathbf{L})_2{}^{163}DyCl_3]\cdot 2H_2O]$ **$^{163}DyZn_2$**
Crystal system	monoclinic	monoclinic	monoclinic
Cell parameters	a = 16.6(1) Å b = 23.3(2) Å c = 14.6(1) Å α = 90° β = 104.8(1)° γ = 90°	a = 16.00(3) Å b = 22.24(5) Å c = 14.17(3) Å α = 90° β = 104.59(5)° γ = 90°	a = 16.21(2) Å b = 22.66(2) Å c = 14.25(1) Å α = 90° β = 105.06(3)° γ = 90°
Volume / Å^3	5490(137)	4883(30)	5053(16)
T / K	293(2)	150(2)	150(2)
Compounds	$Dy_{0.1}Y_{0.9}(Zn\mathbf{L})_2$ **$(Dy@Y)Zn_2$**	$^{163}Dy_{0.1}Y_{0.9}(Zn\mathbf{L})_2$ **$(^{163}Dy@Y)Zn_2$**	$^{162}Dy_{0.1}Y_{0.9}(Zn\mathbf{L})_2$ **$(^{162}Dy@Y)Zn_2$**
Crystal system	monoclinic	monoclinic	monoclinic
Cell parameters	a = 16.114(2) Å b = 22.449(3) Å c = 14.233(2) Å α = 90° β = 104.371(5)° γ = 90°	a = 15.94(3) Å b = 22.39 (5) Å c = 14.11(3) Å α = 90° β = 104.58(7)° γ = 90°	a = 16.17(3) Å b = 22.62(5) Å c = 14.28(2) Å α = 90° β = 104.71(6)° γ = 90°
Volume / Å^3	4987(2)	4874(28)	5053(27)
T / K	150(2)	150(2)	293(2)

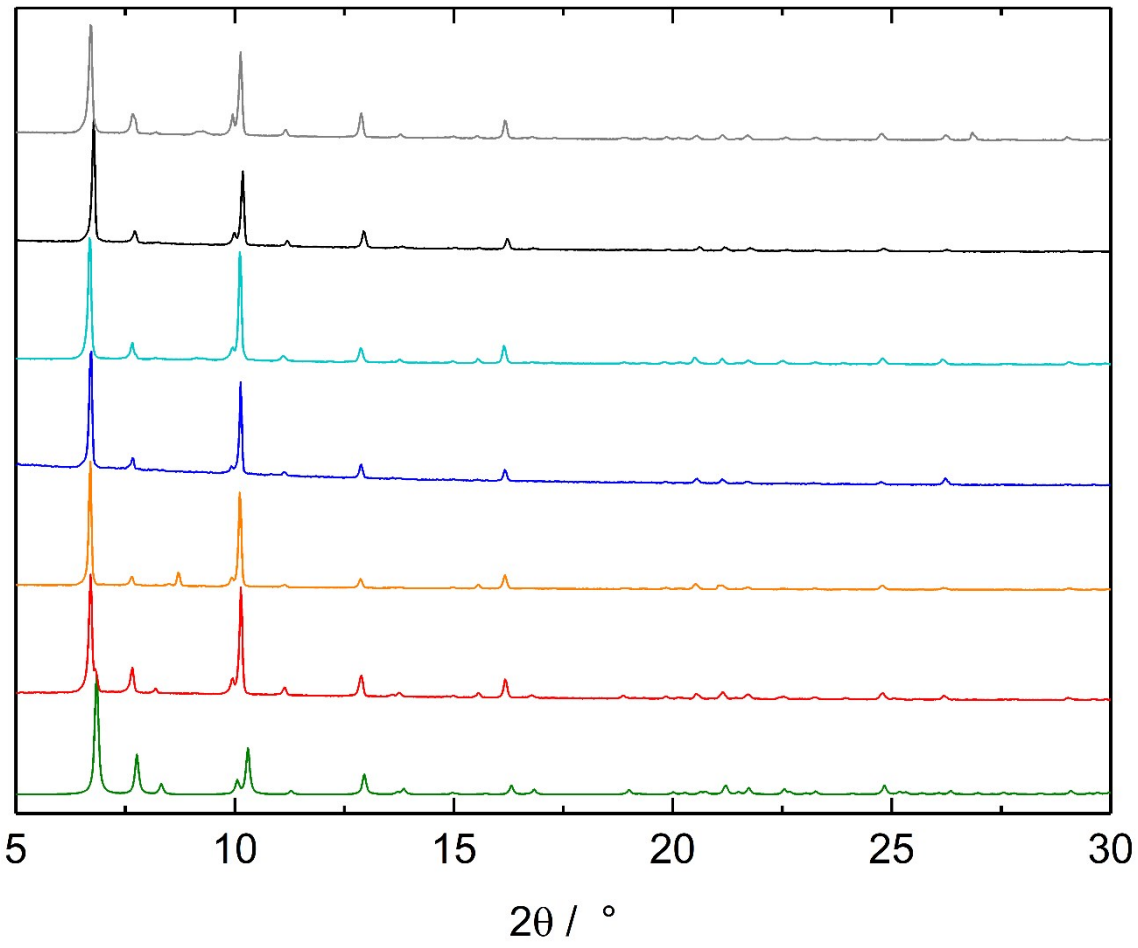

Fig. S1 Powder X-Ray Diffraction pattern of ^{162}**DyZn$_2$** (red), ^{163}**DyZn$_2$** (blue), **DyZn$_2$** (black), (**^{162}Dy@Y)Zn$_2$** (orange), (**^{163}Dy@Y)Zn$_2$** (light blue), and **(Dy@Y)Zn$_2$** (grey) with the simulated pattern from single crystal crystallographic structure (green).

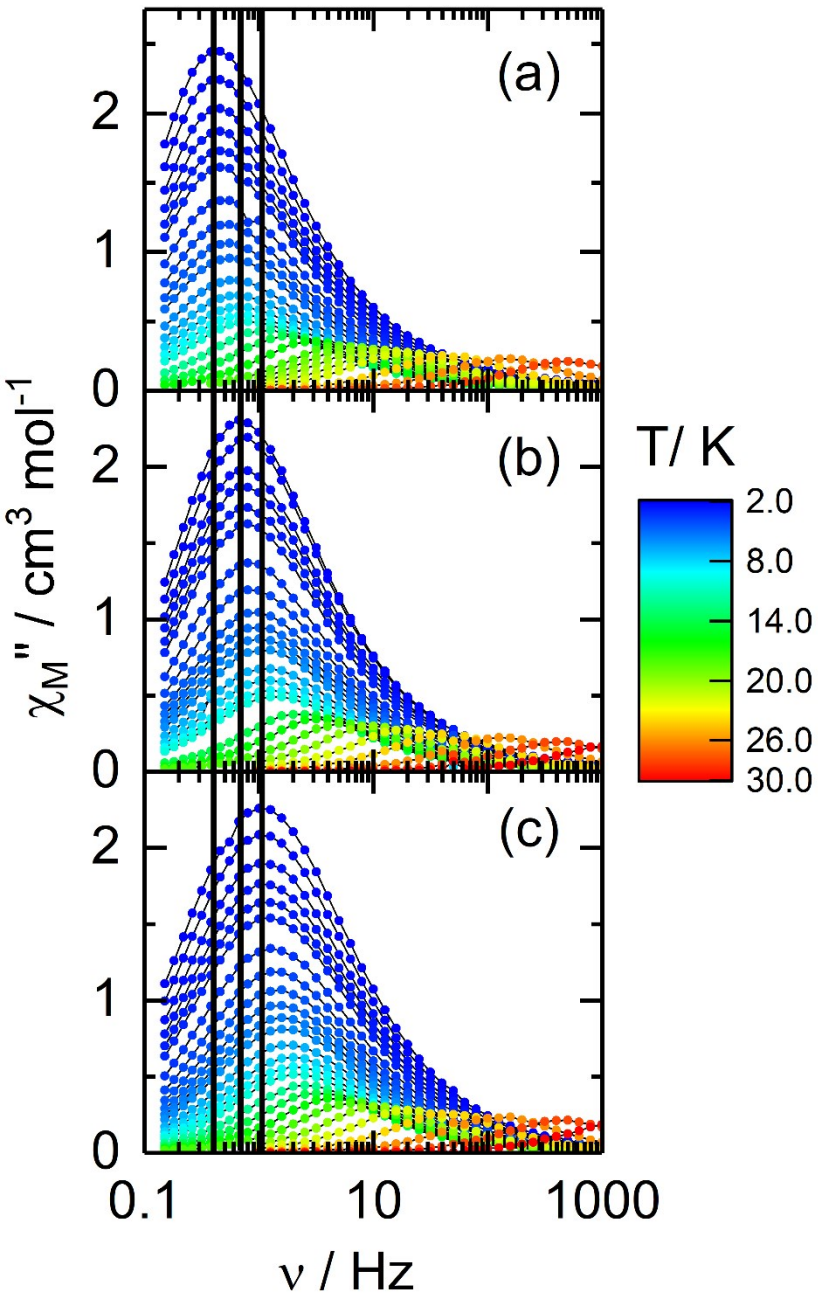

Fig. S2 Frequency dependence of the out-of-phase component of the ac susceptibility measured in zero dc field for a) ^{162}**DyZn$_2$**, b) **DyZn$_2$**.and c) ^{163}**DyZn$_2$**.

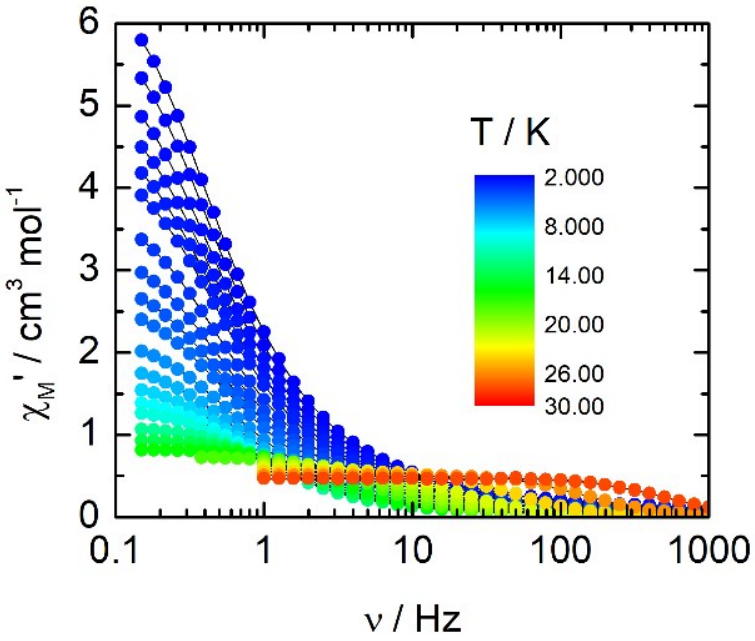

Fig. S3 Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for ^{162}**DyZn$_2$**.

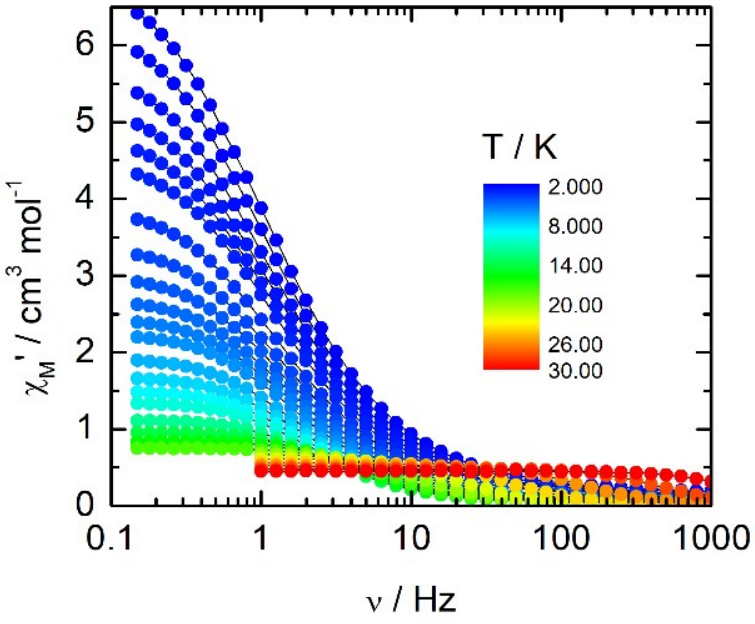

Fig. S4 Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for ^{163}**DyZn$_2$**.

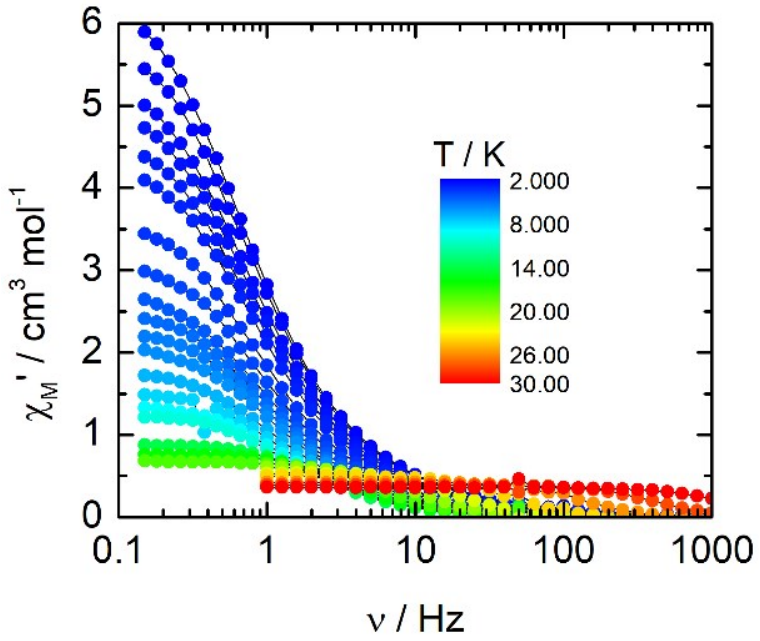

Fig. S5 Frequency dependence of the in-phase component of the ac susceptibility measured in zero dc field for **$DyZn_2$**.

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 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 Fig. here below for an example). The best fitted parameters τ, α, χ_T, χ_S are listed in Table S4 to S6 with the coefficient of determination R^2.

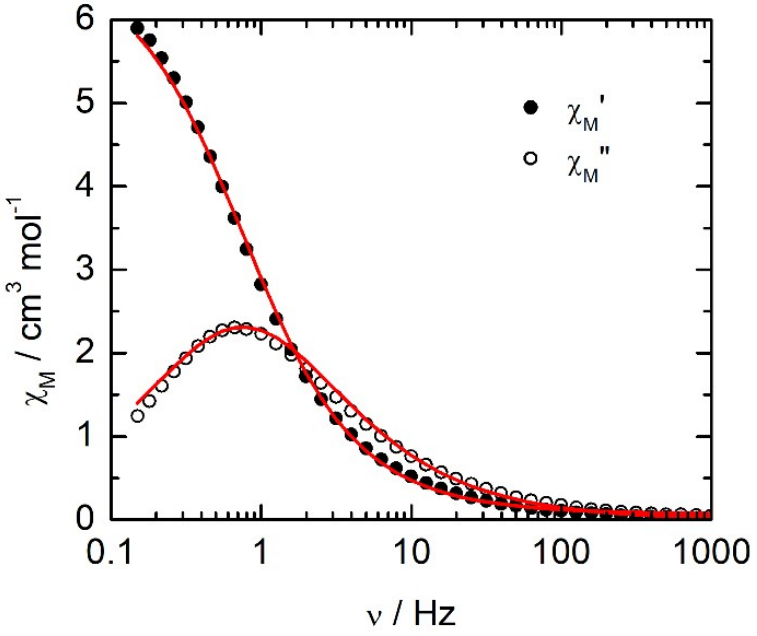

Fig. S6 Frequency dependence of the in-phase (χ_M') and out-of-phase (χ_M'') components of the ac susceptibility measured at 2 K and in zero field with the best fitted curves (red lines) with extended Debye model for **$DyZn_2$**.

Table S2. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **$DyZn_2$** at zero field in the temperature range 2-30 K.

T / K	χ_S / $cm^3\,mol^{-1}$	χ_T / $cm^3\,mol^{-1}$	τ / s	α	R^2
2	0.06364	6.76759	0.20956	0.2313	0.99883
2.2	0.05261	6.31192	0.20076	0.23460	0.99909
2.4	0.04870	5.73907	0.19732	0.23041	0.99909
2.6	0.04202	5.40171	0.19248	0.22864	0.99898
2.8	0.03456	5.00450	0.18790	0.22855	0.99899
3	0.03359	4.65074	0.18308	0.22564	0.99907
3.5	0.02683	3.87878	0.17187	0.21653	0.99889
4	0.02286	3.35228	0.16090	0.21216	0.99861
4.5	0.00178	2.92428	0.15163	0.21654	0.99871
5	0.01456	2.65793	0.14625	0.20724	0.99862
5.5	0.00122	2.41672	0.13780	0.20455	0.99845
6	0.01119	2.19950	0.13152	0.19935	0.99887
7	0.01094	1.86396	0.12377	0.19289	0.99852
8	2.00E-17	1.60925	0.11741	0.19878	0.99136
9	1.00E-18	1.42252	0.10714	0.17877	0.99874
10	1.00E-17	1.30001	0.10092	0.16527	0.99785
14	2.00E-19	0.90082	0.06211	0.10913	0.99650
16	1.00E-19	0.77292	0.04185	0.06794	0.99656
18	1.00E-19	0.68093	0.02686	0.05004	0.99684
20	1.00E-20	0.61398	0.01577	0.00990	0.99523
22	2.00E-19	0.54277	0.00829	0.00000	0.99783
24	1.00E-17	0.50052	0.00363	0.01740	0.99444
26	1.00E-17	0.42284	0.00120	0.00000	0.99478
28	1.00E-17	0.38701	0.00040	0.00764	0.99243
30	2.00E-15	0.36664	0.00012	0.04032	0.99161

Table S3. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **^{162}DyZn$_2$** at zero field in the temperature range 2-30 K.

T / K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ / s	α	R^2
2	0.18631	7.2569	0.14328	0.27431	0.99984
2.2	0.17554	6.64902	0.13978	0.2691	0.99982
2.4	0.16166	6.01877	0.135	0.26446	0.99979
2.6	0.15356	5.5293	0.13086	0.25913	0.99977
2.8	0.14612	5.12011	0.12692	0.25358	0.99975
3	0.13878	4.76358	0.1229	0.24863	0.99972
3.5	0.12309	4.06371	0.11432	0.23734	0.99968
4	0.11311	3.5335	0.10681	0.22577	0.99965
4.5	0.10103	3.13599	0.10037	0.22017	0.99947
5	0.09566	2.80741	0.09529	0.21193	0.99956
6	0.0844	2.32908	0.08713	0.19861	0.99952
7	0.07559	1.99164	0.08062	0.18796	0.99955
8	0.06709	1.73955	0.0749	0.17877	0.99951
9	0.06351	1.54245	0.07028	0.16678	0.99947
10	0.0622	1.38657	0.06593	0.15664	0.99931
12	0.05537	1.14269	0.05586	0.13316	0.99938
14	0.05011	0.97606	0.04462	0.1029	0.99948
16	0.04466	0.85155	0.03303	0.07653	0.99935
18	0.04218	0.75363	0.02241	0.05173	0.99967
20	0.0364	0.68224	0.01404	0.0433	0.99953
22	0.03485	0.61906	0.00783	0.02487	0.99936
24	0.03245	0.56625	0.00349	0.02133	0.99968
26	0.03119	0.52446	0.0012	0.02461	0.99899
28	0.03378	0.48537	3.59E-04	0.02677	0.9998
30	0.05261	0.45309	1.14E-04	0.01256	0.99958

Table S4. Best fitted parameters (χ_T, χ_S, τ and α) with the extended Debye model for compound **$^{163}DyZn_2$** at zero field in the temperature range 2-28 K.

T / K	χ_S / cm^3 mol^{-1}	χ_T / cm^3 mol^{-1}	τ / s	α	R^2
2	0.17806	7.45396	0.36148	0.25266	0.99862
2.2	0.16695	6.82998	0.35441	0.25241	0.9986
2.4	0.15048	6.19792	0.34558	0.25354	0.99858
2.6	0.1409	5.70086	0.33728	0.25413	0.99868
2.8	0.13374	5.26501	0.32849	0.25199	0.99868
3	0.12719	4.90592	0.32259	0.25321	0.99857
3.5	0.11278	4.18015	0.30443	0.2529	0.99861
4	0.10178	3.63887	0.28842	0.249	0.99878
4.5	0.0904	3.22248	0.275	0.24879	0.9988
5	0.08482	2.89185	0.26382	0.24677	0.99874
6	0.06861	2.40127	0.24437	0.24534	0.99874
7	0.06362	2.04674	0.22742	0.23813	0.99867
8	0.05851	1.77966	0.21065	0.2284	0.99865
9	0.05422	1.57321	0.19198	0.21708	0.99857
10	0.05125	1.40587	0.17124	0.20153	0.99849
12	0.04521	1.13776	0.12313	0.15308	0.99844
14	0.04242	0.96055	0.08153	0.10621	0.99878
16	0.03439	0.83136	0.04959	0.07454	0.99925
18	0.03012	0.73865	0.02894	0.05778	0.99921
19	0.02664	0.7036	0.0219	0.05958	0.9988
20	0.02656	0.66321	0.01622	0.03949	0.99948
21	0.02637	0.62872	0.0118	0.03095	0.99958
22	0.02151	0.60139	0.00807	0.03391	0.99974
24	0.02139	0.55098	0.00331	0.03245	0.99972
26	0.02256	0.50763	0.00109	0.03554	0.99972
28	0.02723	0.47036	3.28E-04	0.03235	0.99973

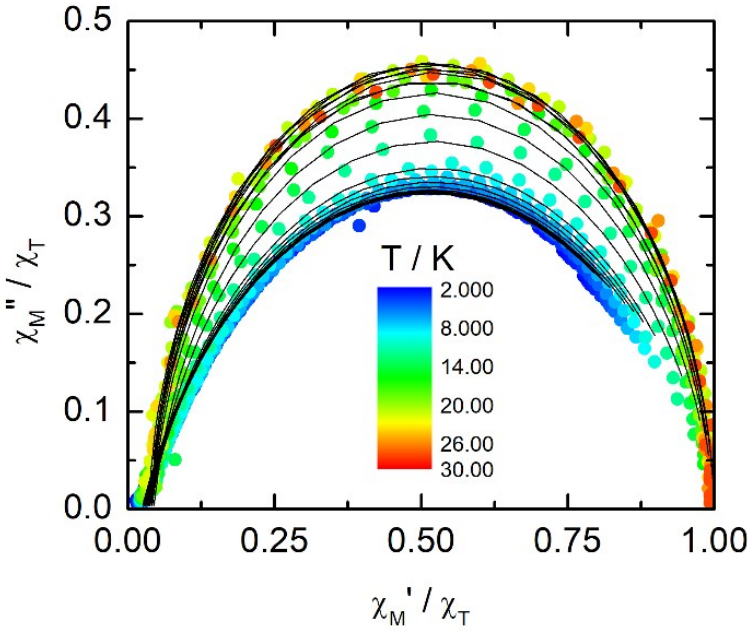

Fig. S7 Normalized Cole-Cole plots in zero field for $^{162}DyZn_2$ between 2 and 28 K.

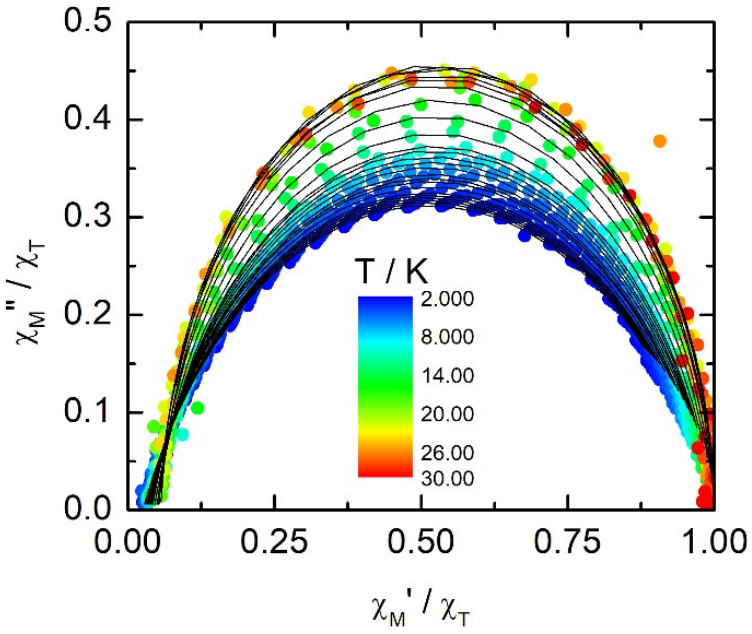

Fig. S8 Normalized Cole-Cole plots in zero field for $^{163}DyZn_2$ between 2 and 28 K.

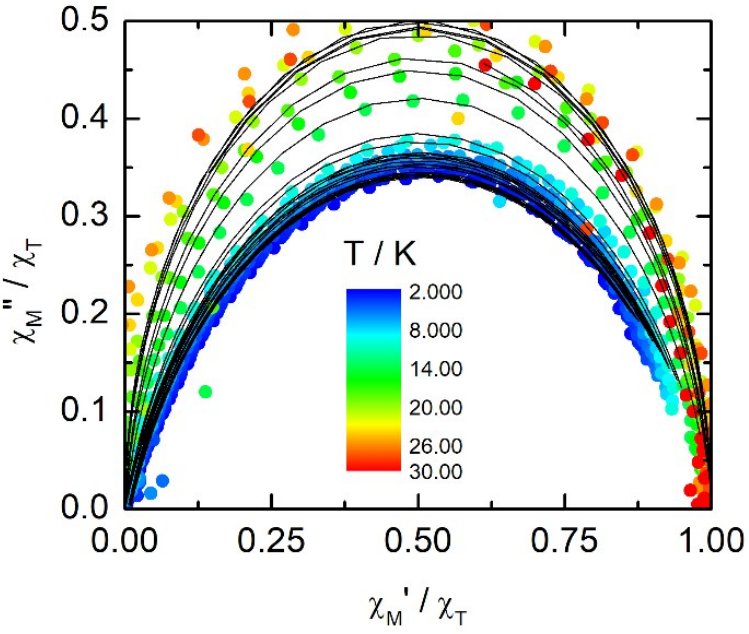

Fig. S9 Normalized Cole-Cole plots in zero field for **$DyZn_2$** between 2 and 30 K.

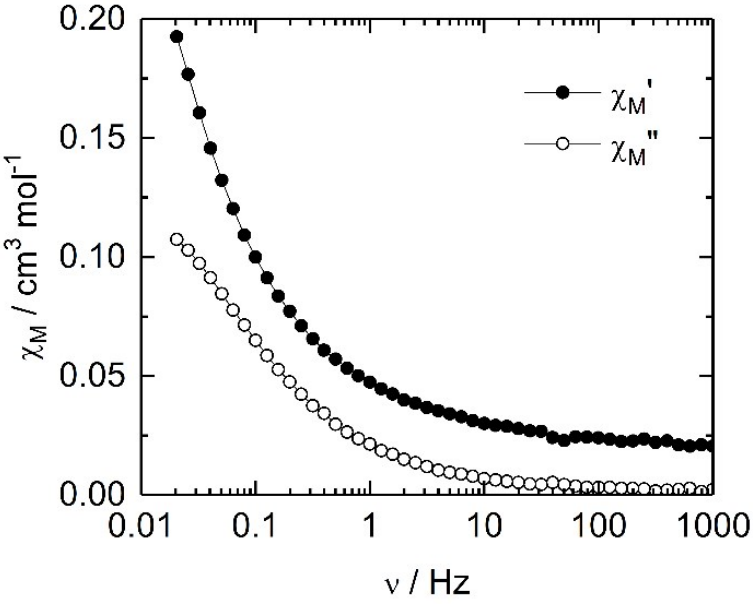

Fig. S10 Frequency dependence of the in-phase (χ_M') and out-of-phase (χ_M'') components of the ac susceptibility measured at 2 K and zero field for **(Dy@Y)Zn_2**.

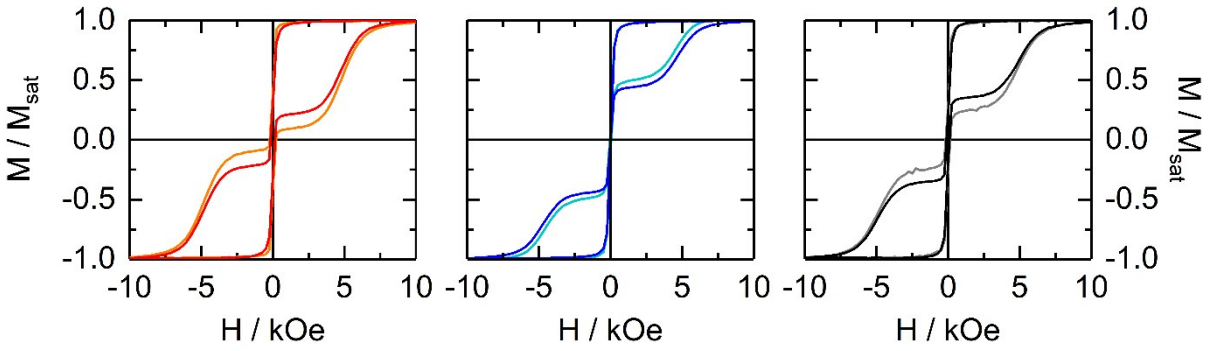

Fig. S11 Comparison of the different condensed systems with their analogue diluted. *N*ormalized magnetic hysteresis loops at 0.5 K and at a sweep rate of 16 Oe s^{-1} for **$DyZn_2$** (black), **$^{162}DyZn_2$** (red), **$^{163}DyZn_2$** (blue); and (b) the diluted **(Dy@Y)Zn_2** (grey), **^{162}(Dy@Y)Zn_2** (orange) and **^{163}(Dy@Y)Zn_2** (light blue).

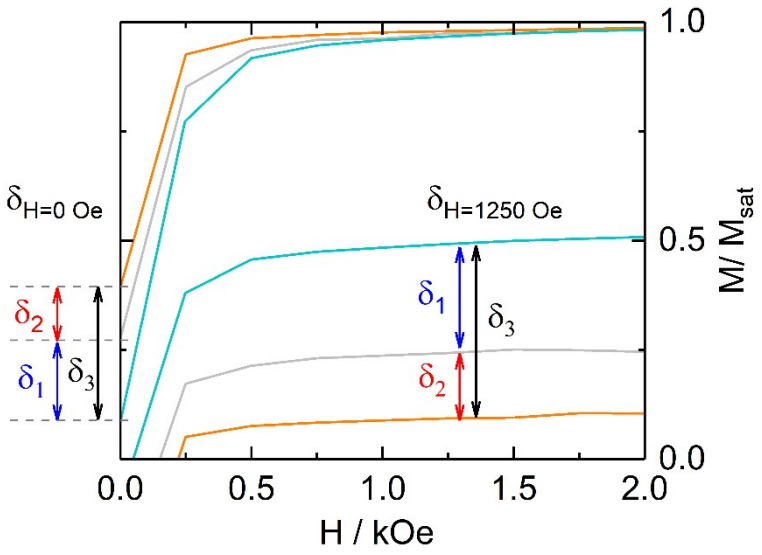

Fig. S12 Zoom of the normalized magnetic hysteresis loops at 0.5 K and at a sweep rate of 16 Oe s^{-1} for **(Dy@Y)Zn_2** (grey), **^{162}(Dy@Y)Zn_2** (orange) and **^{163}(Dy@Y)Zn_2** (light blue) for the definition of δ_1, δ_2 and δ_3 parameters.

Table S5. δ_1, δ_2 and δ_3 parameters extracted from the normalized magnetic hysteresis loops for **(Dy@Y)Zn_2**, **^{162}(Dy@Y)Zn_2** and **^{163}(Dy@Y)Zn_2** at 0.5 K.

	H = 0 Oe	H = 1250 Oe	Δ*
δ_1 (**^{163}(Dy@Y)Zn_2-(Dy@Y)Zn_2**)	0.1783	0.2469	0.0686
δ_2 (**^{162}(Dy@Y)Zn_2-(Dy@Y)Zn_2**)	0.1271	0.1463	0.0192
δ_3 (**^{163}(Dy@Y)Zn_2-^{162}(Dy@Y)Zn_2**)	0.3058	0.3932	0.0874

*Δ is defined as the absolute difference between δ_n (n = 1-3) determined at H = 0 Oe and H = 1250 Oe.