Supplementary Information

Two-dimensional frameworks built from Single-Molecule Magnets

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Experimental details

Complex **1**: $Mn(ClO_4)_2 \cdot 6H_2O$ (0.330g, 0.900mmol) and $saoH_2$ (0.122g, 0.900mmol) were dissolved in MeOH (20mL). MeONa (1.000mmol) was then added under stirring (solution A) and the solution stirred for another 40 min. A methanolic solution (10mL) containing $tmaH_3$ (0.025g, 0.100mmol) was then added to solution A and the solution stirred for about 1 min. The resulting solution left undisturbed. X-ray quality, dark green, crystals of **1** were obtained in 40% yield. Elemental analysis for **1**: Calculated (%) for $C_{55.25}H_{51.25}O_{24.5}N_6Mn_6$ (vacuum dried complex **1**): C: 43.63; H: 3.60; N: 5.53; Found: C: 43.58; H: 3.55; N: 5.59.

Complex **2**: To a methanolic solution of $Mn(ClO_4)_2 \cdot 6H_2O$ (0.330g, 0.900mmol), $saoH_2$ (0.122g, 0.900mmol) and $tmaH_3$ (0.025g, 0.100mmol), MeONa (1.000mmol) was added in small portions under stirring to form a dark green slurry solution. The solution turns to clear dark green after a stirring period of 14 mins. The resulting solution left undisturbed to form dark green crystals of **2** in 40% yield. Elemental analysis for **2**: Calculated (%) for $C_{51.33}H_{48}O_{22.67}N_6Mn_6$ (vacuum dried complex **2**): C: 42.78; H: 3.36; N: 5.83; Found: C: 42.72; H: 3.28; N: 5.90.

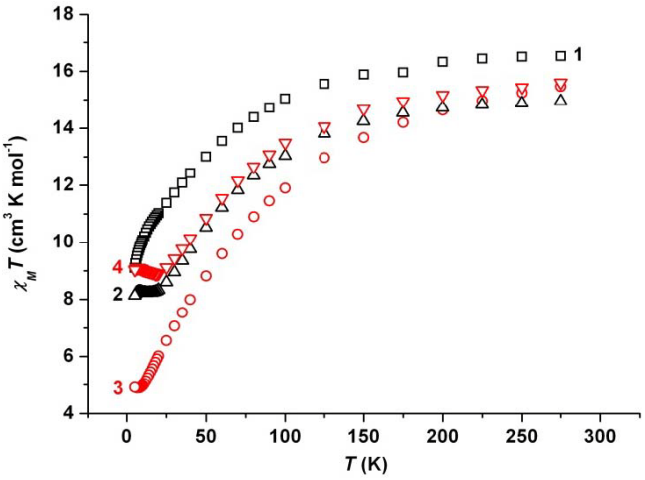
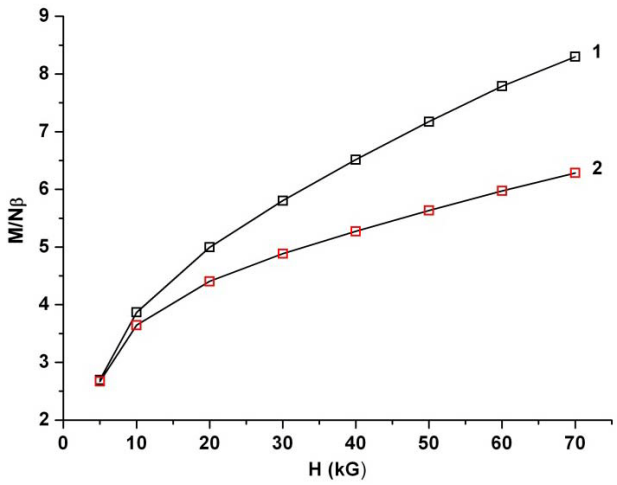


Fig. S1 Plot of $\chi_M T$ versus T for complexes **1** and **2**. The data could not be successfully fitted because each complex has more than one crystallographically independent [Mn_6] in the crystal. For comparison the data for [Mn_6] with similar torsion angles, reference 3c, are also plotted as **3** and **4**.

Fig. S2 Plot of magnetisation ($M/N\mu_B$) versus field (H) at 2 K for **1** and **2**. The data for each could not be successfully fitted using a simple model that assumes only population of the ground state. The reason, as with most members of this family, is the presence of multiple low-lying excited states and the existence of more than one crystallographically independent [Mn_6] in the crystal.



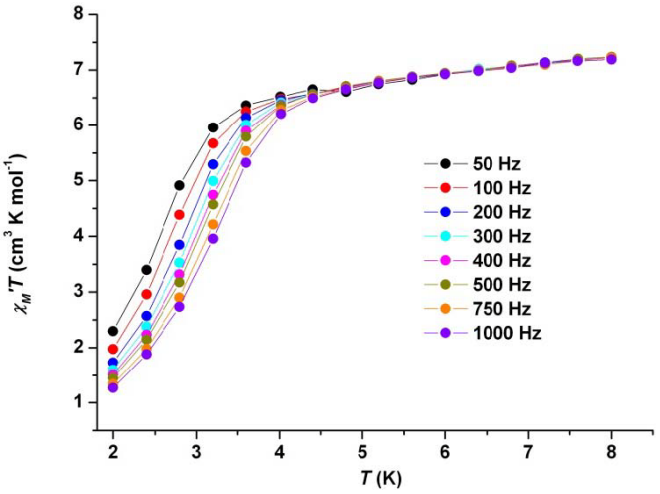
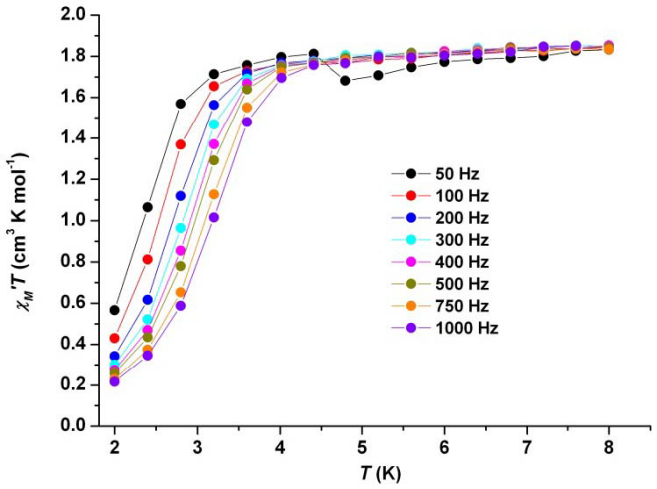


Fig. S3 Plot of $\chi_M T$ versus T for complex **1** at the indicated temperature and frequency ranges.

Fig. S4 Plot of $\chi_M T$ versus T for complex **2** at the indicated temperature and frequency ranges.



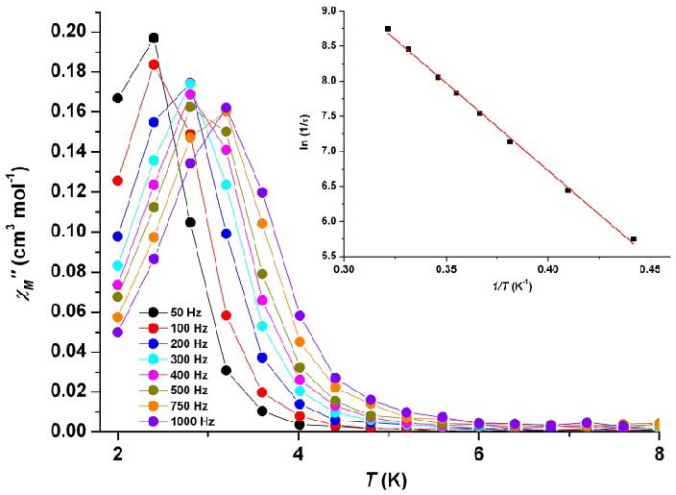


Fig. S5 Plot of χ_M'' versus T for complex **2** at the indicated temperature and frequency ranges. Inset: Arrhenius plots constructed from the χ'' data to afford $\tau_0 = 5.6\times10^{-8}$ s and $U_{eff} \approx 24$ K.

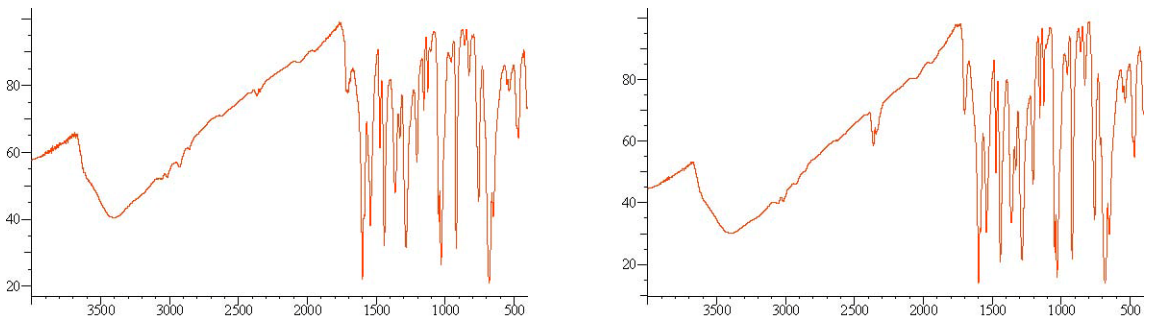


Fig. S6 The IR spectra (KBr disks) of complex **1** (left) and **2** (right) in the 4000 – 400 cm^{-1} region.

Table S1: Dimensions of the unique hydrogen bonds (distances in Å and angles in °) for **1**.

D[a]-H···A[b]	D[a]···A[b]	H···A[b]	<D[a]HA[b]
O32K-H32K···O1N [3-x,3-y,-z]	2.698(7)	1.88(1)	164(1)
O1W-H1W···O33K [3-x,3-y,-z]	2.694(7)	1.86(1)	176(1)
O53D-H53D···O1B [1-x,2-y,1-z]	2.660(7)	1.84(1)	163(1)
O1Q-H1Q···O	2.773(7)	1.94(1)	169(1)

[a]D = donor atom.
[b]A = acceptor atom.