Supporting Information for

Coexistence of Magnetization- and Dielectric Relaxations in A Single-Chain Magnet

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Synthesis of $[Mn^{III}_3O(Meppz)_3(EtOH)_4(OAc)]$ (**1**)

Method A: Ammonium or sodium acetate (0.1 mmol) was added to the ethanol-acetonitril (2:1) solution of $[Mn^{III}_3O(Meppz)_3(EtOH)_5Cl]$ (**2**, 0.1 mmol), the mixture was stirred until the acetate salts completely dissolved. The solution was filtered and filtrate was left undisturbed, and dark blue block crystals of 1 crystallized in one week. Yield: > 90%.

Method B: An ethanol solution (15 mL) containing H_2Meppz (0.1 mmol), $Mn(OAc)_2 \cdot 4H_2O$ (0.1 mmol) and NaOEt (0.2 mmol) was stirred for 15 min, then acetone (5 mL) was added to the turbid solution. The solution was stirred for another 5 min and then filtered, the filtrate was left undisturbed and slow evaporation of solvents gave crystals of **1** in yield of ~ 50%.

Method C: H_2Meppz (0.1 mmol) and NaOEt (0.2 mmol) were dissolved in ethanol solution (8 mL) and put in one arm of H-tube, another ethanol solution (8 mL) containing $Mn(OAc)_2 \cdot 4H_2O$ (0.1 mmol) was put in another arm of the H-tube. Pure Ethanol was carefully added to the H-tube. Large dark blue block crystals of **1** crystallized in one month at the middle tunnel. Yield: ~ 50%.

Synthesis of $[Mn^{III}_3O(Meppz)_3(EtOH)_5Cl]$ (**2**)

An ethanol solution (15 mL) containing H_2Meppz (0.1 mmol), $MnCl_2 \cdot 4H_2O$ (0.1 mmol) and NaOEt (0.2 mmol) was stirred for 15 min. The solution was filtered and the filtrate was left undisturbed, slow evaporation of solvent gave dark blue crystals of **2** in yield of ~ 70%.

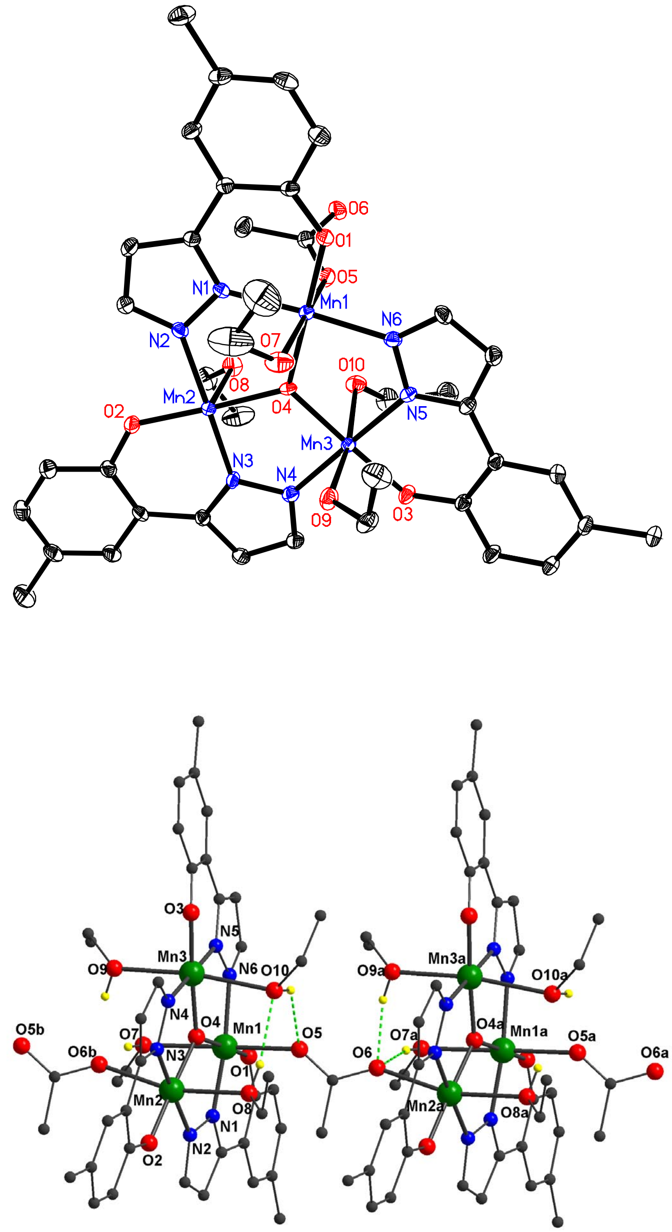

Figure S1 Coordination environments of manganese ions in **1** and the hydrogen bonds between acetate group and ethanol molecules.

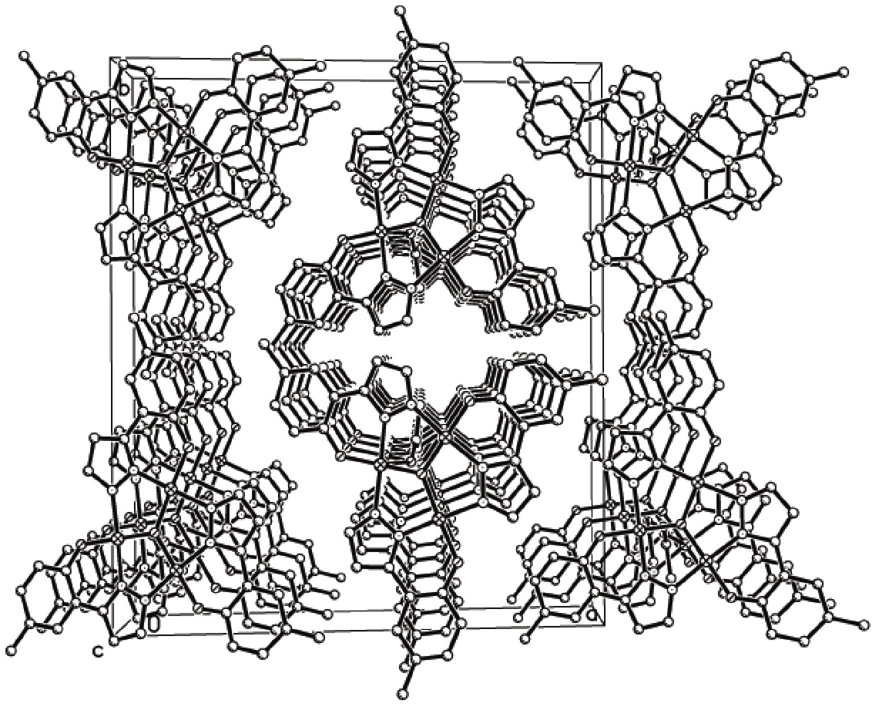

Figure S2 Three-dimensional arrays of the chain of **1** viewed along the *c* axis.

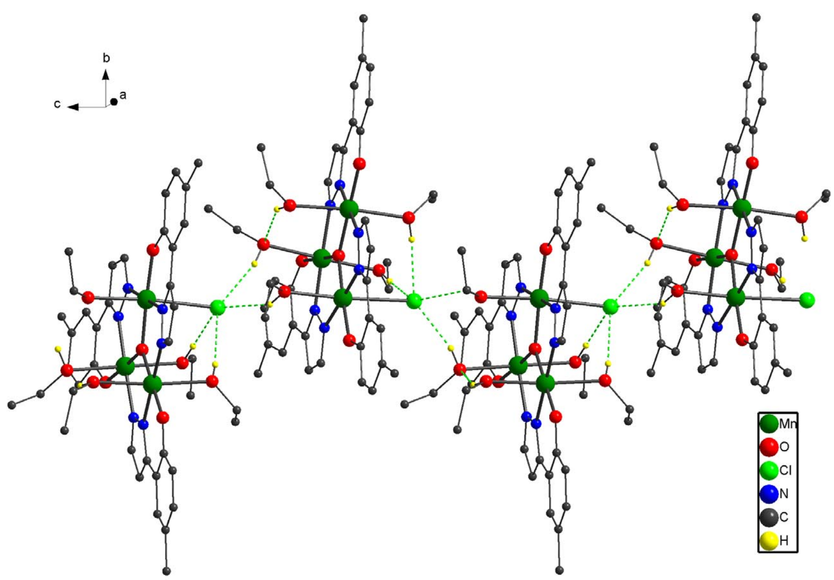

Figure S3 The structure of **2** showing the trinuclear units were connected with each other through O–H···Cl hydrogen bonds.

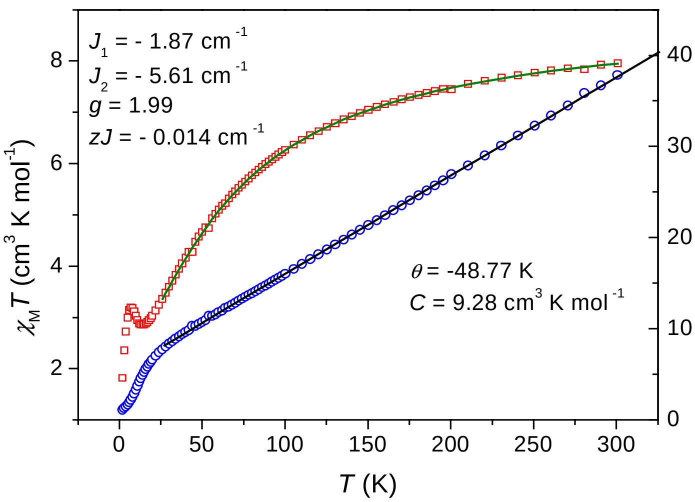

Figure S4 The $\chi_M T$ and $\chi_M^{-1} \sim T$ plots of **1**.

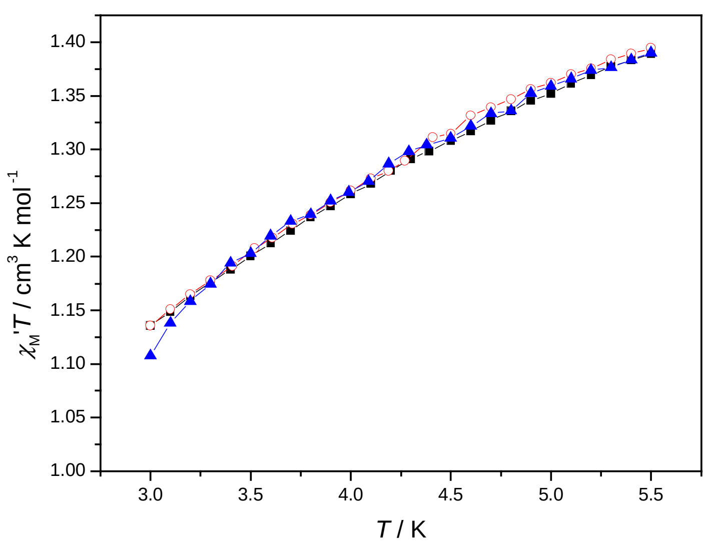

Figure S5 The $\chi_M'T \sim T$ plots of **1** at 1 Hz (■), 10 Hz (○) and 50 Hz (▲).

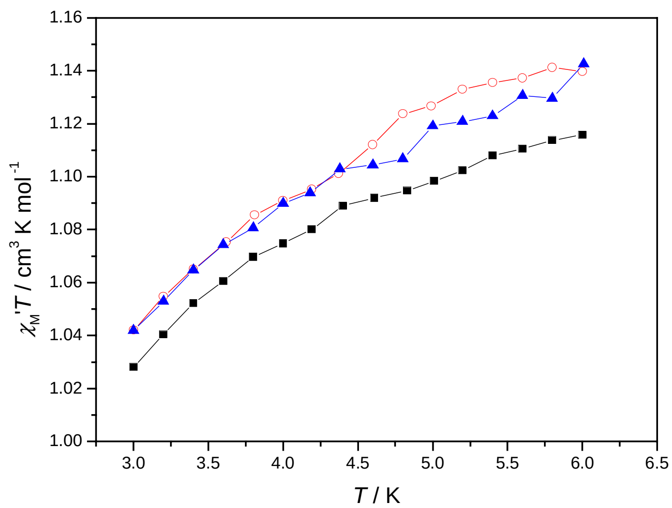

Figure S6 The $\chi_M'T \sim T$ plots of **2** at 1 Hz (■), 10 Hz (○) and 50 Hz (▲).

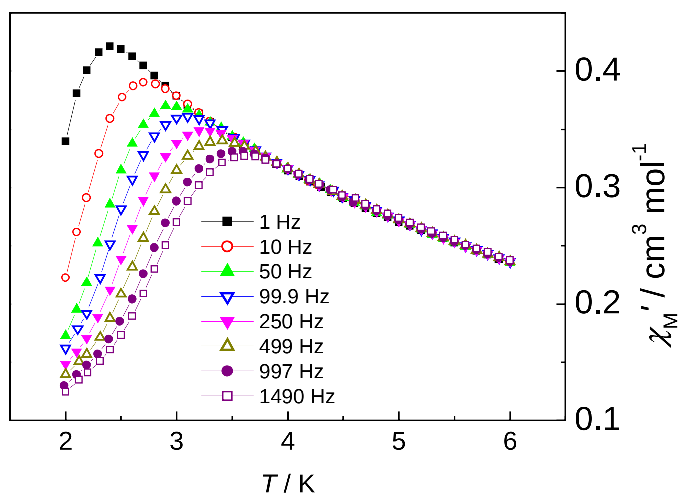

Figure S7 The in-phase (χ_M') AC susceptibility signal of **1**.

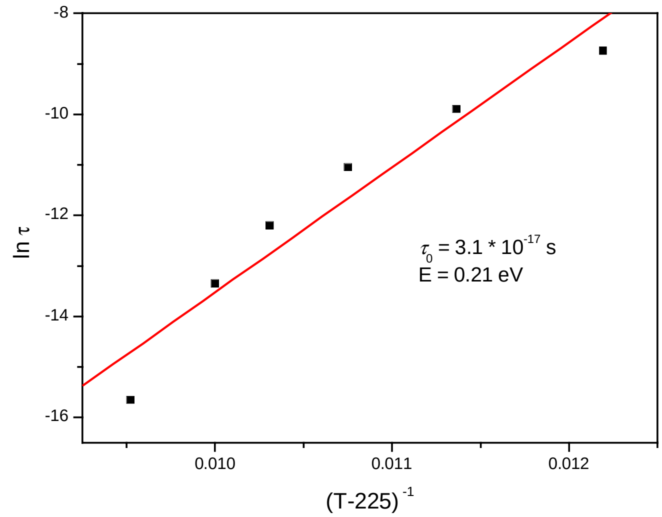

Figure S8 The Vogel-Fulcher fit for complex **1**.