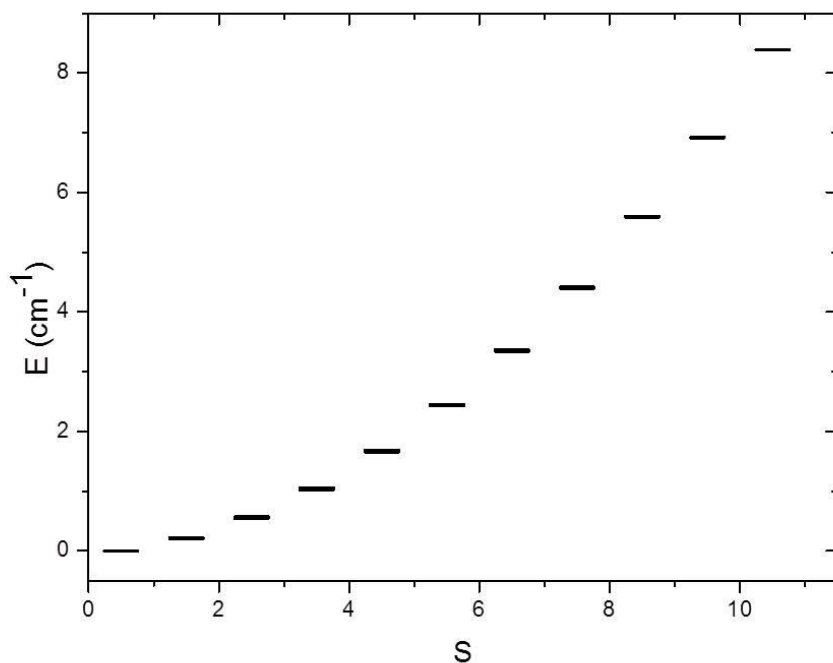
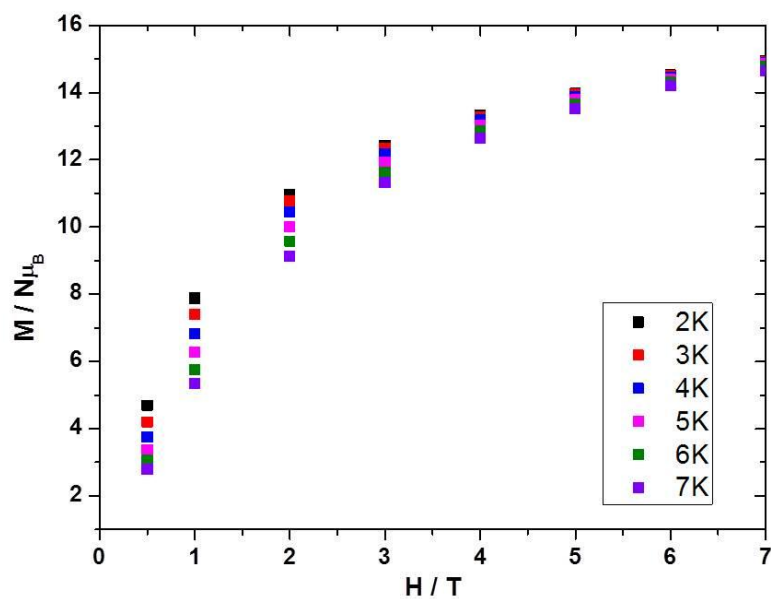


## Supporting Information

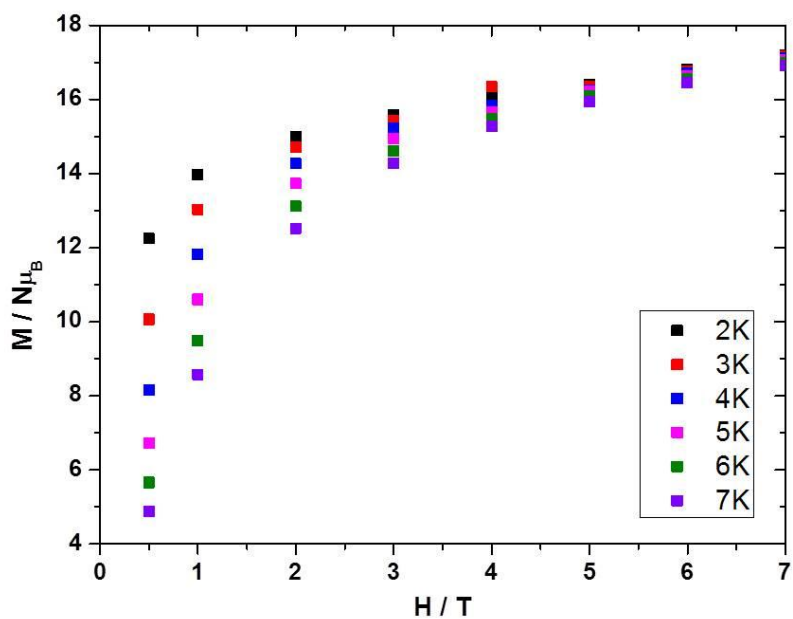
Ross McLellan,<sup>a</sup> Maria A. Palacios,<sup>b</sup> Christine M. Beavers,<sup>c</sup> Simon J. Teat,<sup>c</sup> Euan K. Brechin<sup>\*b</sup> and Scott J. Dalgarno<sup>\*a</sup>



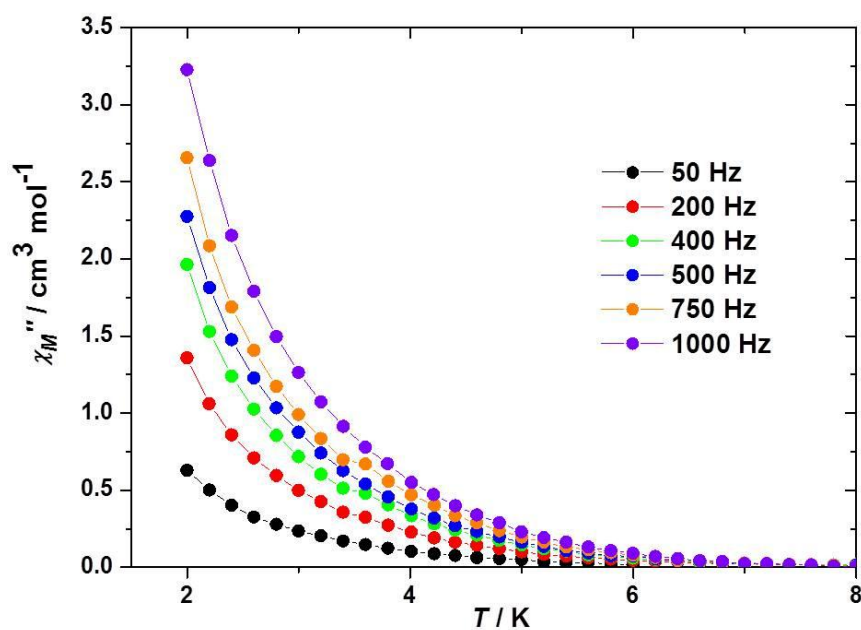
**Figure S1.** Plot of energy versus total spin ( $S$ ) state for complex **1**. The ground state is an  $S=1/2$  state but with a band of low lying states in close proximity.



**Figure S2.** Plot of magnetisation ( $M/N\mu_B$ ) versus field ( $H$ ) for complex **2**.

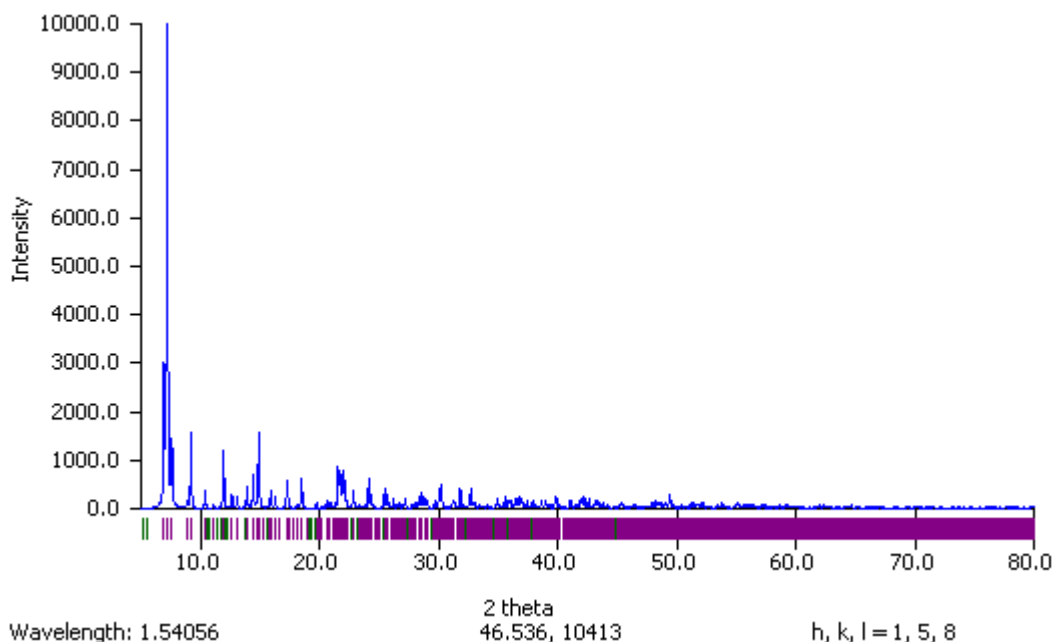


**Figure S3.** Plot of magnetisation ( $M/N\mu_B$ ) versus field ( $H$ ) for complex 3.

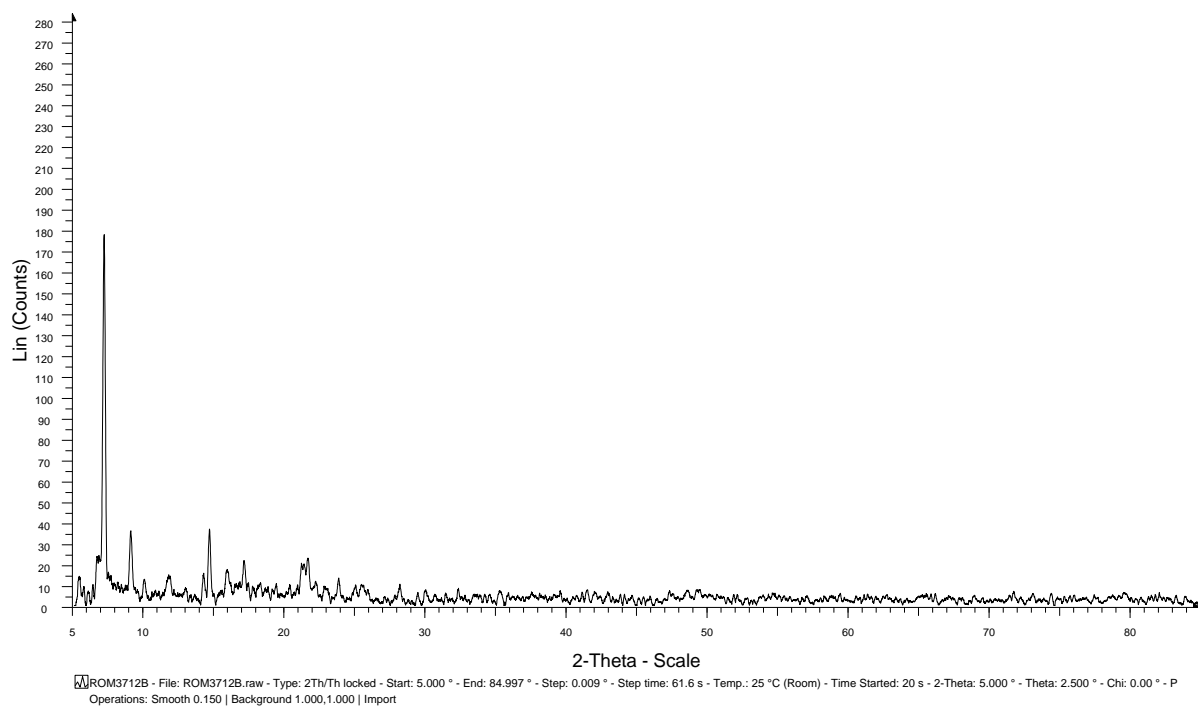


**Figure S4.** Plot of the out-of-phase ( $\chi_M''$ ) ac susceptibility versus temperature for complex 3.

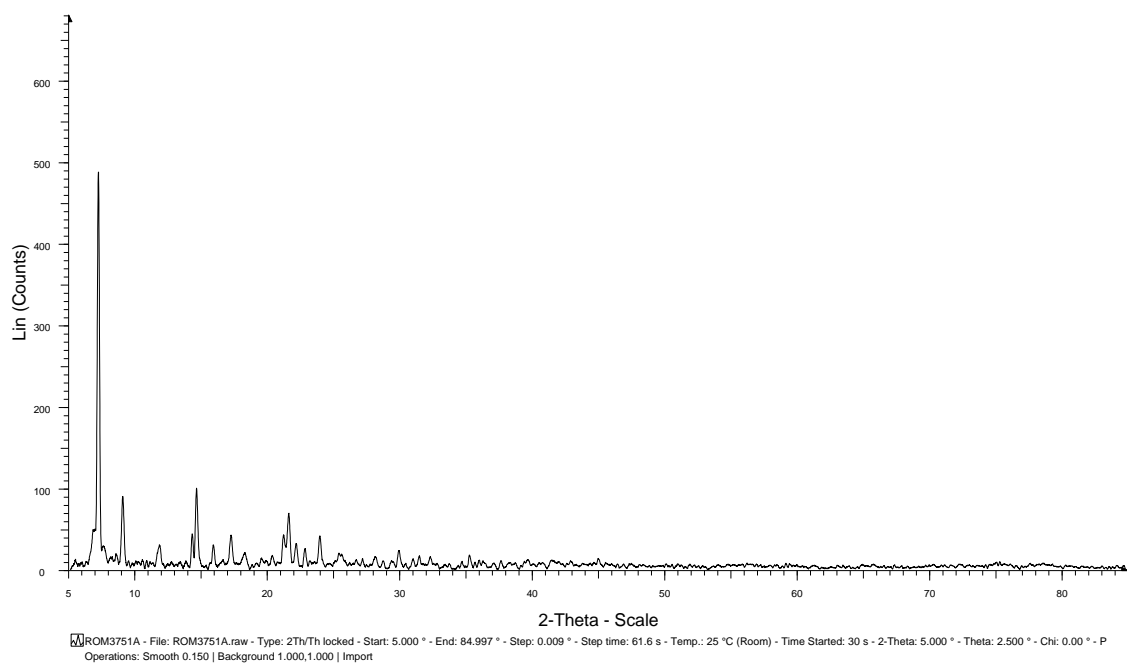
**S5.** *Synthesis of*  $[\text{Gd}^{\text{III}}_3(\text{LH}_4\text{-3H})_4(\text{DMF})_4]\cdot(\text{MeOH})\cdot(\text{MeCN})_{0.25}\cdot(\text{H}_2\text{O})_{0.5}$ , **1**:  $\text{LH}_4$  (200 mg, 0.536 mmol) and  $\text{Gd}(\text{NO}_3)_3\cdot 6\text{H}_2\text{O}$  (484 mg, 1.072 mmol) were dissolved in a 1:1 v/v mixture of MeOH/DMF. After stirring for 10 min.  $\text{Et}_3\text{N}$  (0.3 mL) was added, followed by stirring for a further hour. After filtration, crystals of **1** were grown by diffusion with MeCN. Yield 78 mg, 25% based on  $\text{LH}_4$ . Elemental analysis (%) calculated for dried crystalline **1**,  $\text{C}_{105.75}\text{H}_{139.5}\text{O}_{21.5}\text{N}_{xx}\text{Gd}_3$ : C, 55.55%; H, 6.17%; N, 2.60%. Found: C, 55.22%; H, 6.47%; N, 2.49%. Synthesis of **2** and **3**: These were synthesised in an analogous manner using  $\text{Tb}(\text{NO}_3)_3\cdot 6\text{H}_2\text{O}$  and  $\text{Dy}(\text{NO}_3)_3\cdot 5\text{H}_2\text{O}$ , respectively, in place of  $\text{Gd}(\text{NO}_3)_3\cdot 6\text{H}_2\text{O}$ .



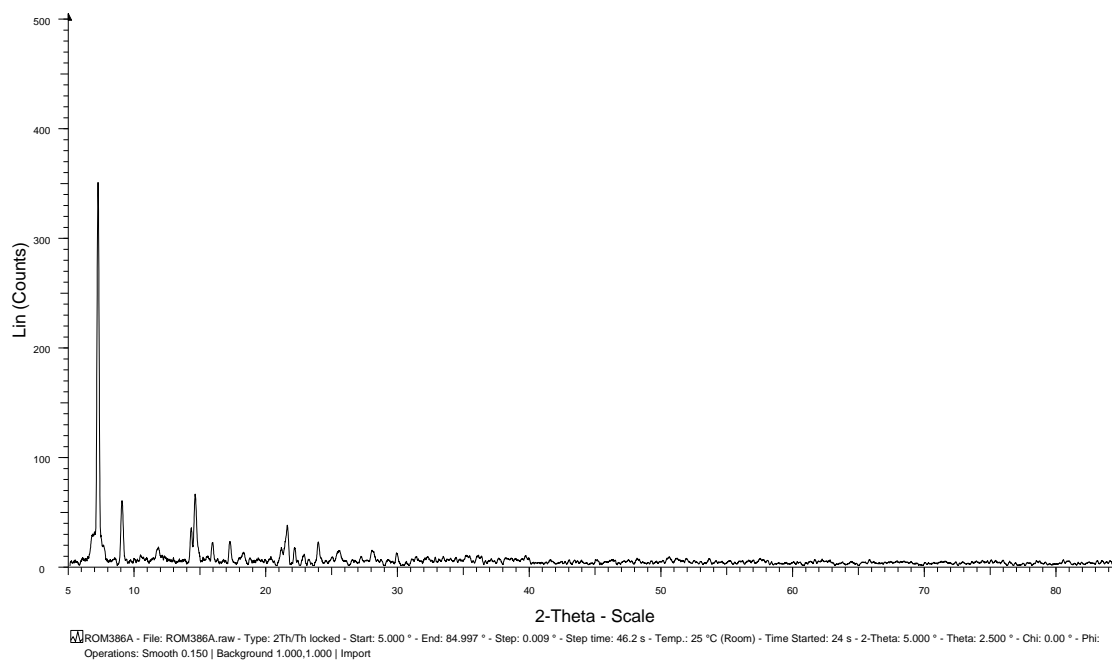
**Figure S6.** Calculated PXRD pattern for **1**.



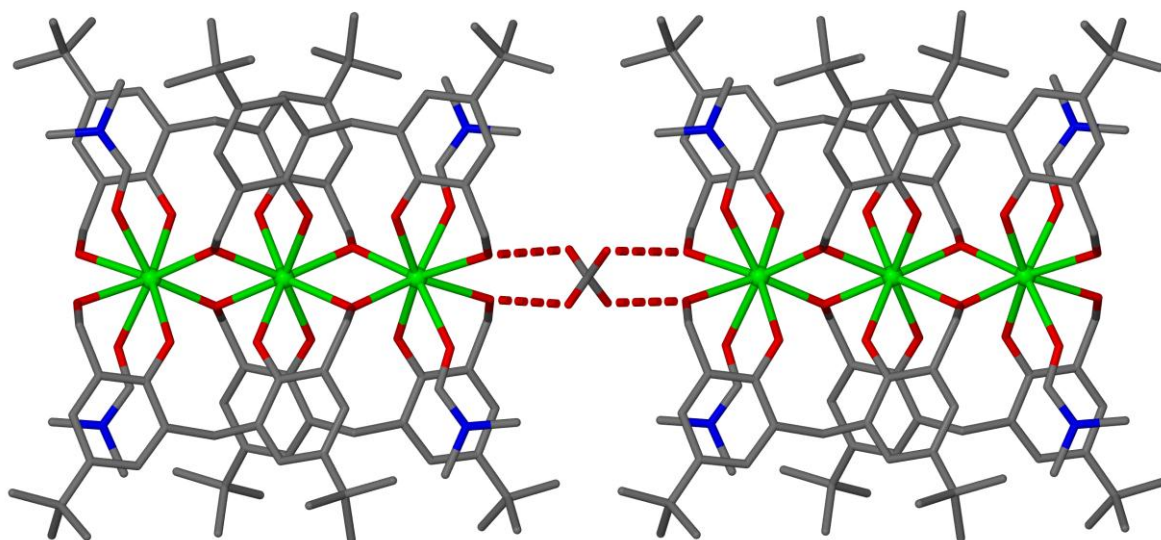
**Figure S7.** Experimental PXRD pattern for **1**.



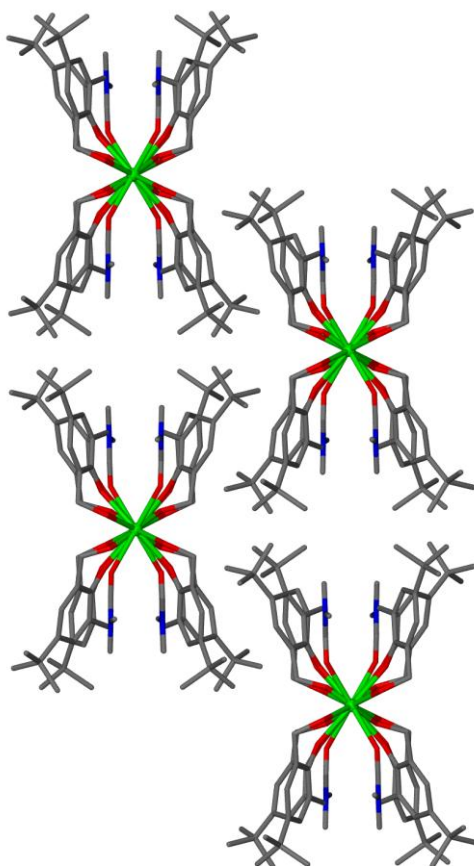
**Figure S8.** Experimental PXRD pattern for **2**.



**Figure S9** Experimental PXRD pattern for **3**.



**Figure S10.** View along *b* axis showing intermolecular interactions between disordered MeOH and neighbouring clusters. Distance between nearest Ln centres is  $\sim 9.6\text{\AA}$ .



**Figure S11.** Views in the *ac* plane showing the extended packing of Ln<sub>3</sub> clusters. The distances between nearest Ln centres are  $\sim 12.6\text{\AA}$ .