

Supporting Information

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- Additional figures relating to the extended structure of **1**.

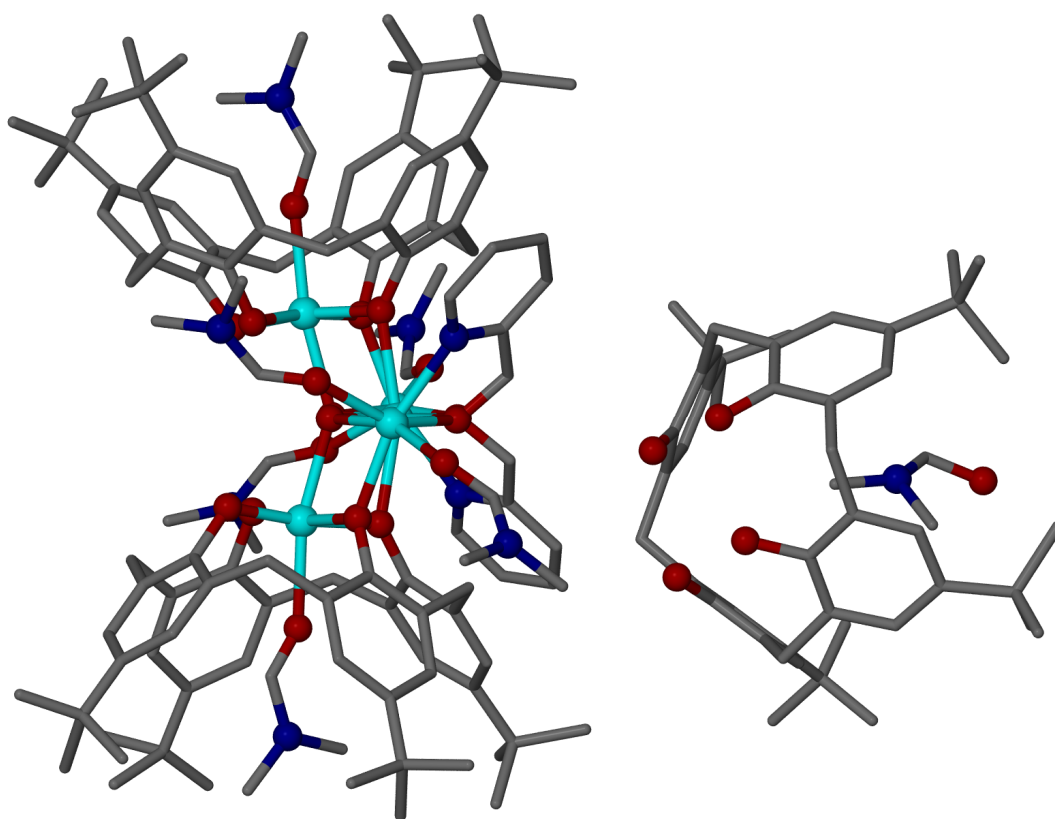


Figure S1. Orthogonal arrangement of the (TBC[4]-H) anion relative to the cation in the asymmetric unit of **1**. Hydrogen atoms and (disordered) ligated / co-crystallised solvent molecules omitted for clarity. Colour code: C – grey, N – dark blue, O – red, Mn – pale blue.

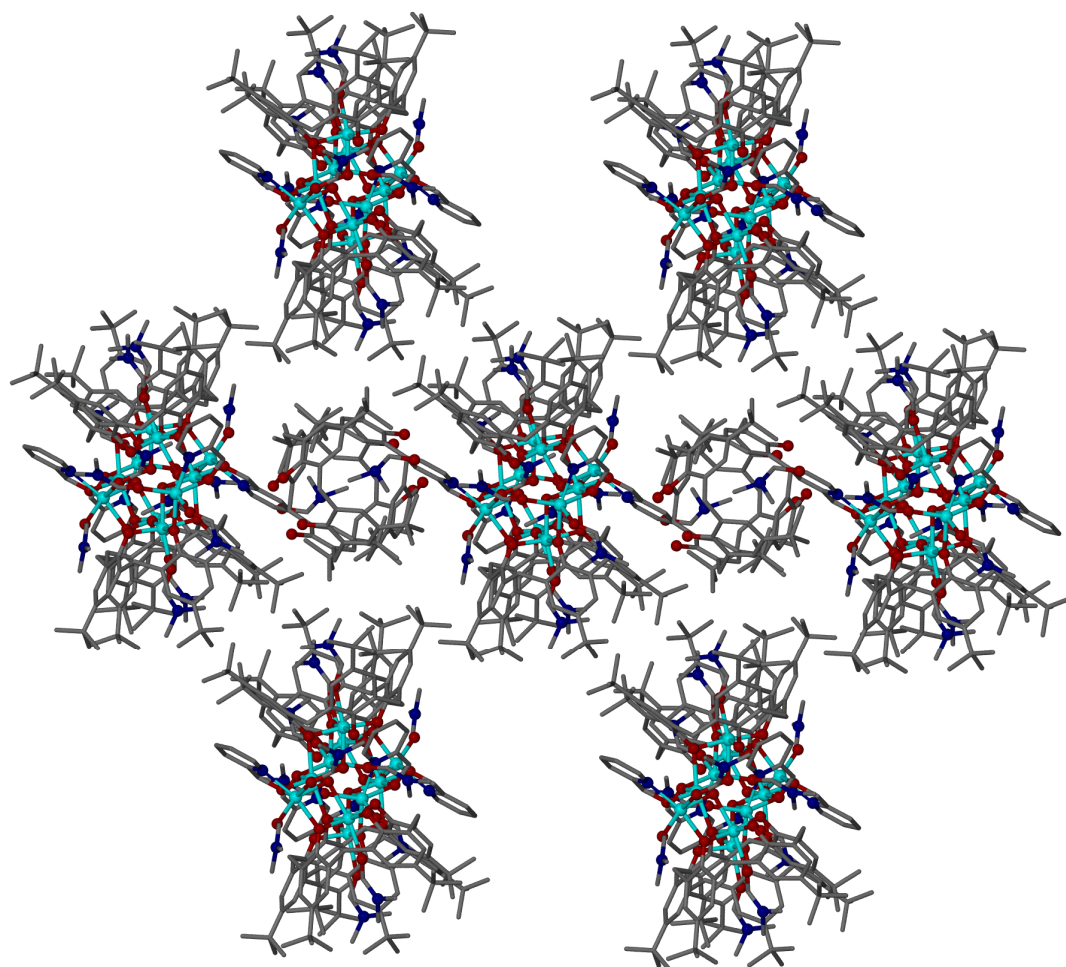


Figure S2. Extended structure of **1** showing the layer-like structure formed by cations. Hydrogen atoms and (disordered) ligated / co-crystallised solvent molecules omitted for clarity. Colour code as for Figure S1.

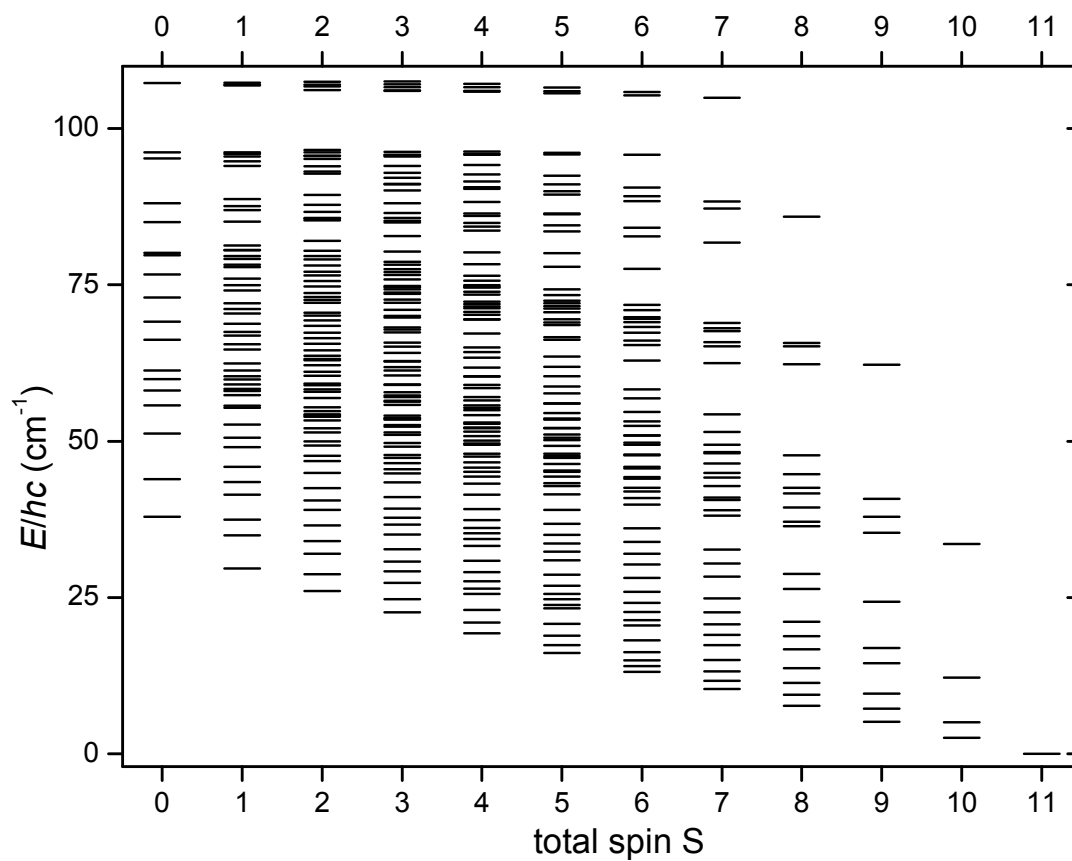


Figure S3. Zero-field energy spectrum of **1**, obtained by diagonalisation of the isotropic part of spin-Hamiltonian (1) with $J_1 = +0.348 \text{ cm}^{-1}$ and $J_2 = +2.553 \text{ cm}^{-1}$.

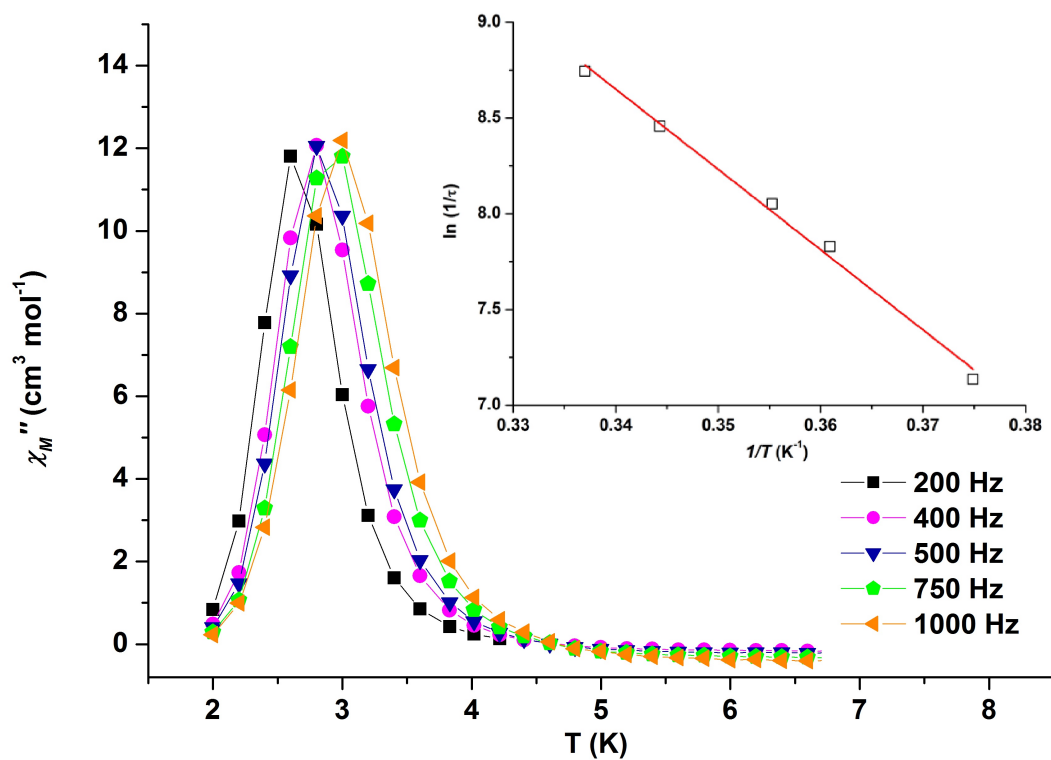


Figure S4. Ac susceptibility studies carried out on crystalline samples of **1** in the 1.8-10.0 K range in a 3.5 G field oscillating at frequencies up to 1000 Hz. Insert: Arrhenius plot constructed from these data afford $U_{\text{eff}} = 42$ K with $\tau_0 = 1.125 \times 10^{-10}$ s