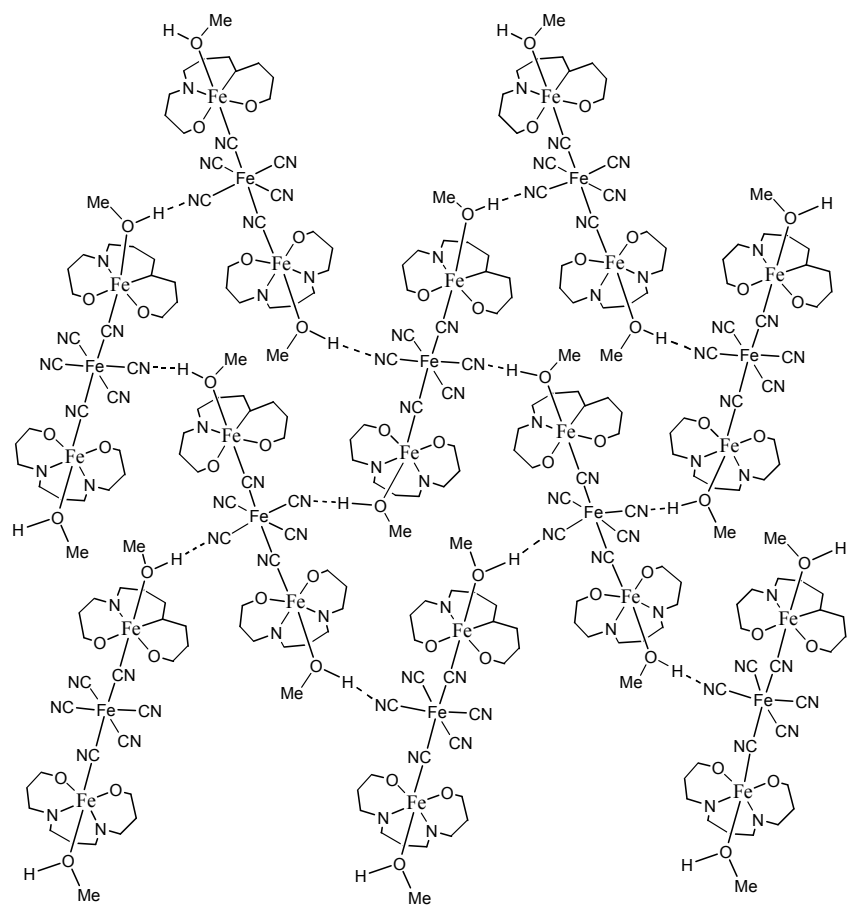


Supplementary materials

Scheme S1 Schematic diagram of **1**.



Scheme S2 Schematic diagram of **2**.

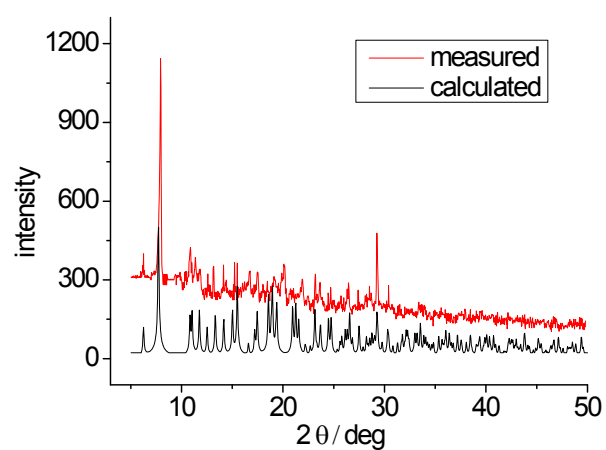


Fig S1. X-ray powder diffraction patterns for **1**.

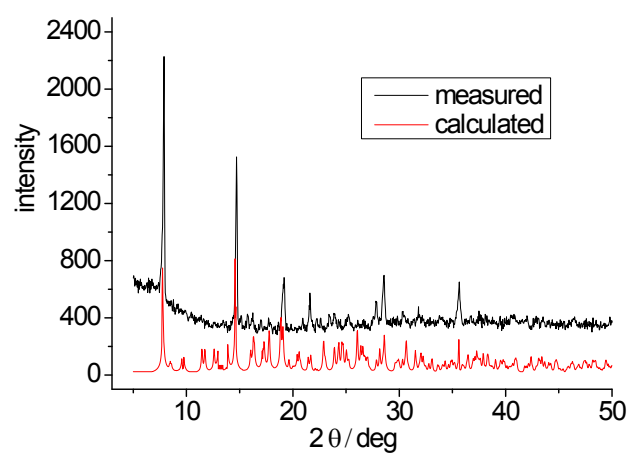


Fig S2. Powder XRD pattern for **2**.

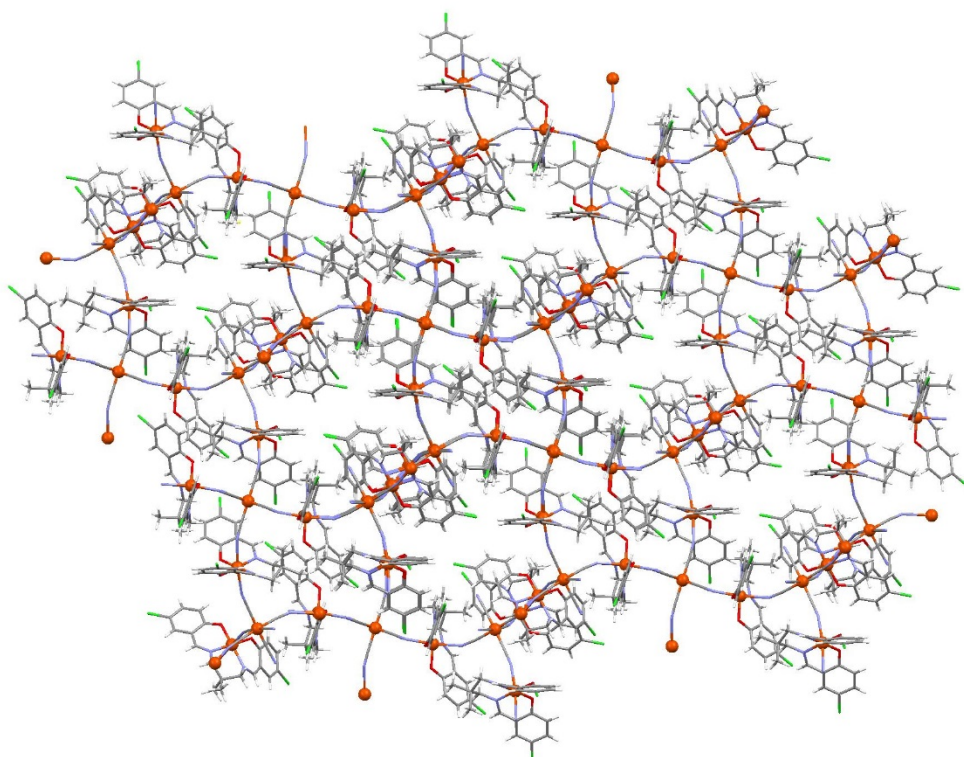


Fig.S3. A three-dimensional network structure in the bc-plane of the compound **1**. $[\text{NEt}_4]^+$ and solvent molecules have been omitted for clarity.

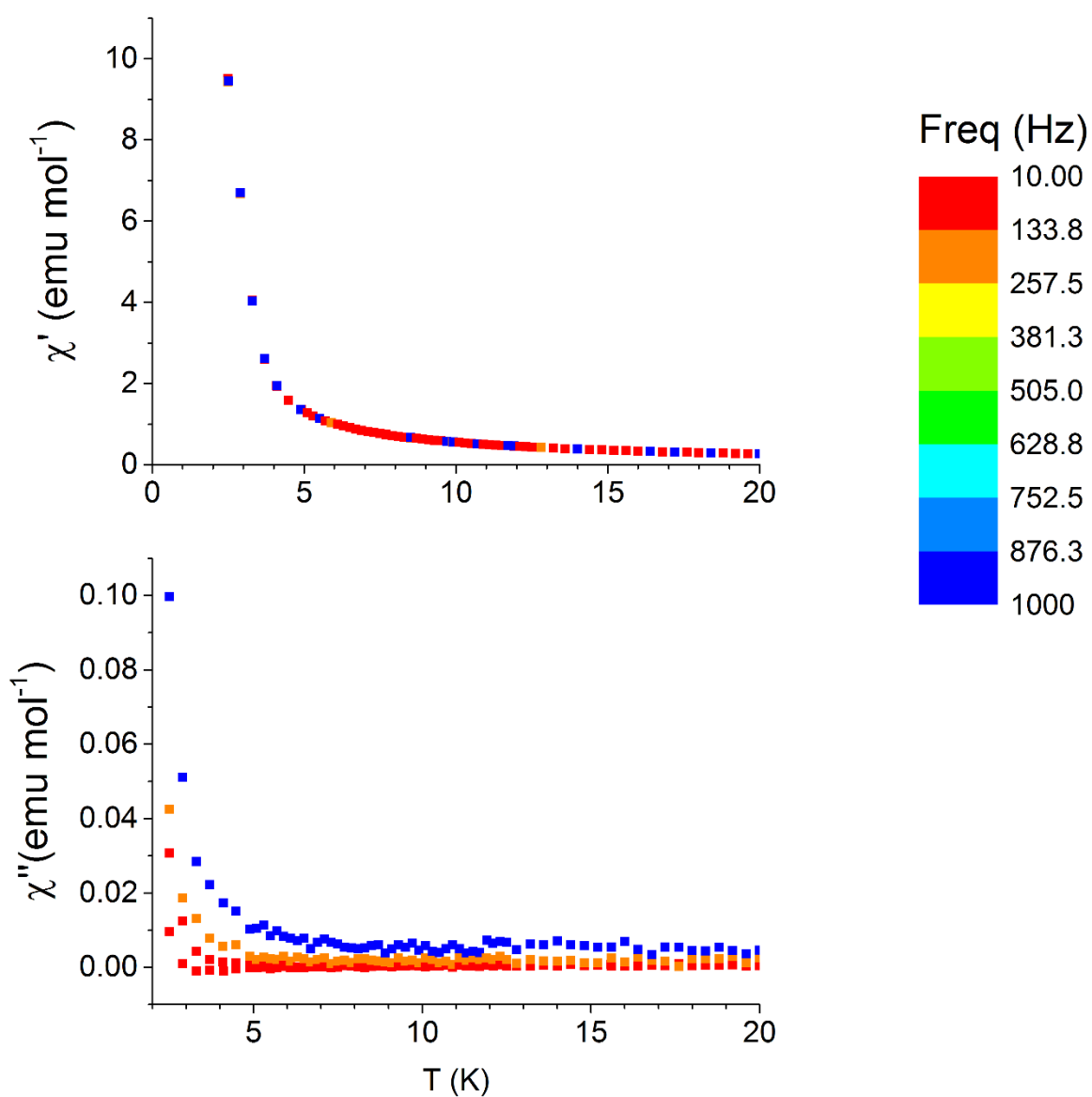


Fig. S4 (a) Temperature dependence of the real (χ') (upper) and imaginary (χ'') (lower) parts of the AC susceptibility for **1** at different frequencies and in zero static field.

Table S1. Structural and magnetic data for **1** and **2** and a series of related compounds

Compound	Struct.	$d_{\text{Fe-Ncyano}} (\text{\AA})$	$\text{Fe-N}\equiv\text{C} (^{\circ})$	$J (\text{cm}^{-1})$	Ref.
$[\text{NEt}_4][\text{Fe}^{\text{III}}(5\text{-CIL1})_2]\text{Fe}^{\text{III}}(\text{CN})_6 \cdot (\text{MeOH}) \cdot (\text{H}_2\text{O})$, 1	3D	2.360(4) – 2.414(4)	156.7(4) – 165.6(4)	F	This work
$[\text{NEt}_4][\text{Fe}^{\text{III}}(5\text{-CIL2})_2(\text{MeOH})_2]\text{Fe}^{\text{III}}(\text{CN})_6$, 2	trinuclear	2.106(4)	152.9(4)	F	This work
$[\{\text{Fe}^{\text{III}}(\text{salen})\}_3\{\text{Fe}^{\text{III}}(\text{CN})_6\}(\text{MeOH})_2]_n \cdot 3n\text{H}_2\text{O}$	2D	2.08(1) – 2.17(1)	150(1) – 168 (1)	F	14