# Electronic Supplementary Information

## The detail of structure refinements

The $tzdc^{3-}$ and protonated dimethylamine $H_2N(CH_3)_2^+$ groups in **1** and **2** are disordered over three crystallographically equivalent sites imposed by the threefold symmetry of the cubic space group $P2_13$. Therefore, their geometries were restrained, while the related atoms are constrained to have equivalent anisotropic atomic displacement parameters (O1 with O4, O2 with O3, C1 with C4, C2 with C3, N1 with N3, C5 with C6, etc.) during the refinements. According the results of elemental analyses and thermogravimetric (TG) analyses, the site of occupancy of the lattice water molecules in **1** and **2** are fixed to be 1/6. Both the *R* factor and weight *R* factor of **2** are larger than those of **1**, mainly due to the more serious disorder in **2** at the higher temperature for the diffraction data collection. In addition, the crystal may be slightly defective and/or twining.

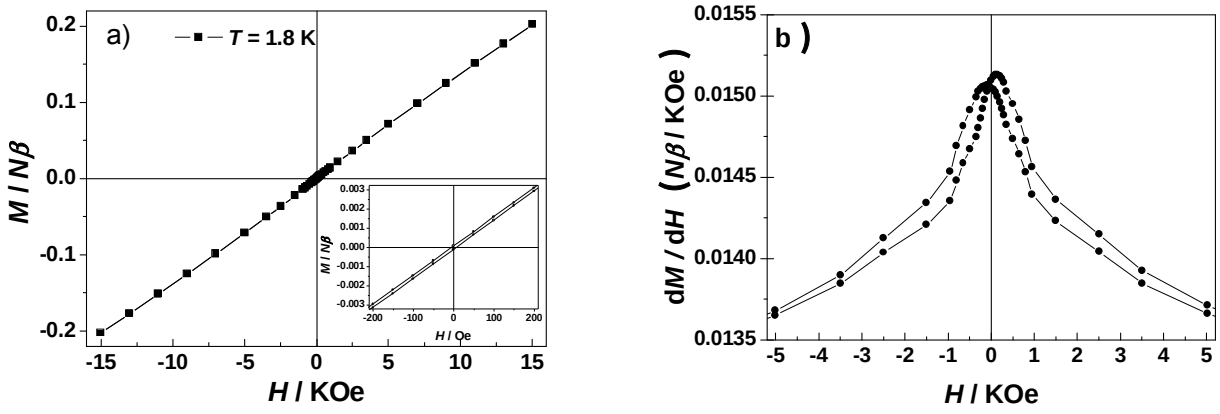


**Figure S1.** The hysteresis loop (a) and its first-order derivative (b) of **1** at 1.8 K. The first-order derivative of *M-H* curve does not overlap, with two peaks at about -100 and 100 Oe, clearly confirming the presence of small open loop which is consistent with its spin-glassy behaviour.

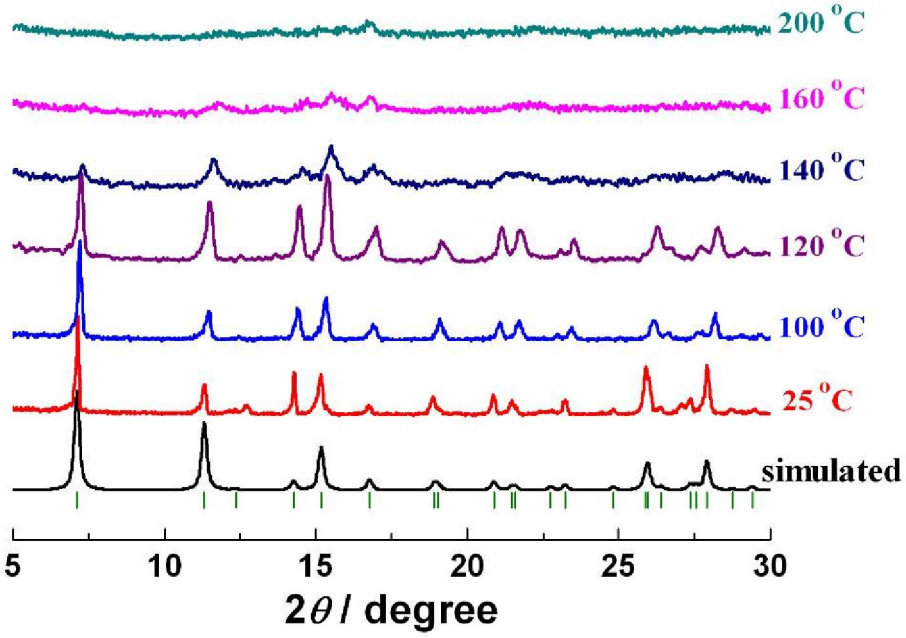


**Figure S2**. The powder X-ray diffraction pattern of **3** at different temperatures. The small green sticks represent the positions of the peak.