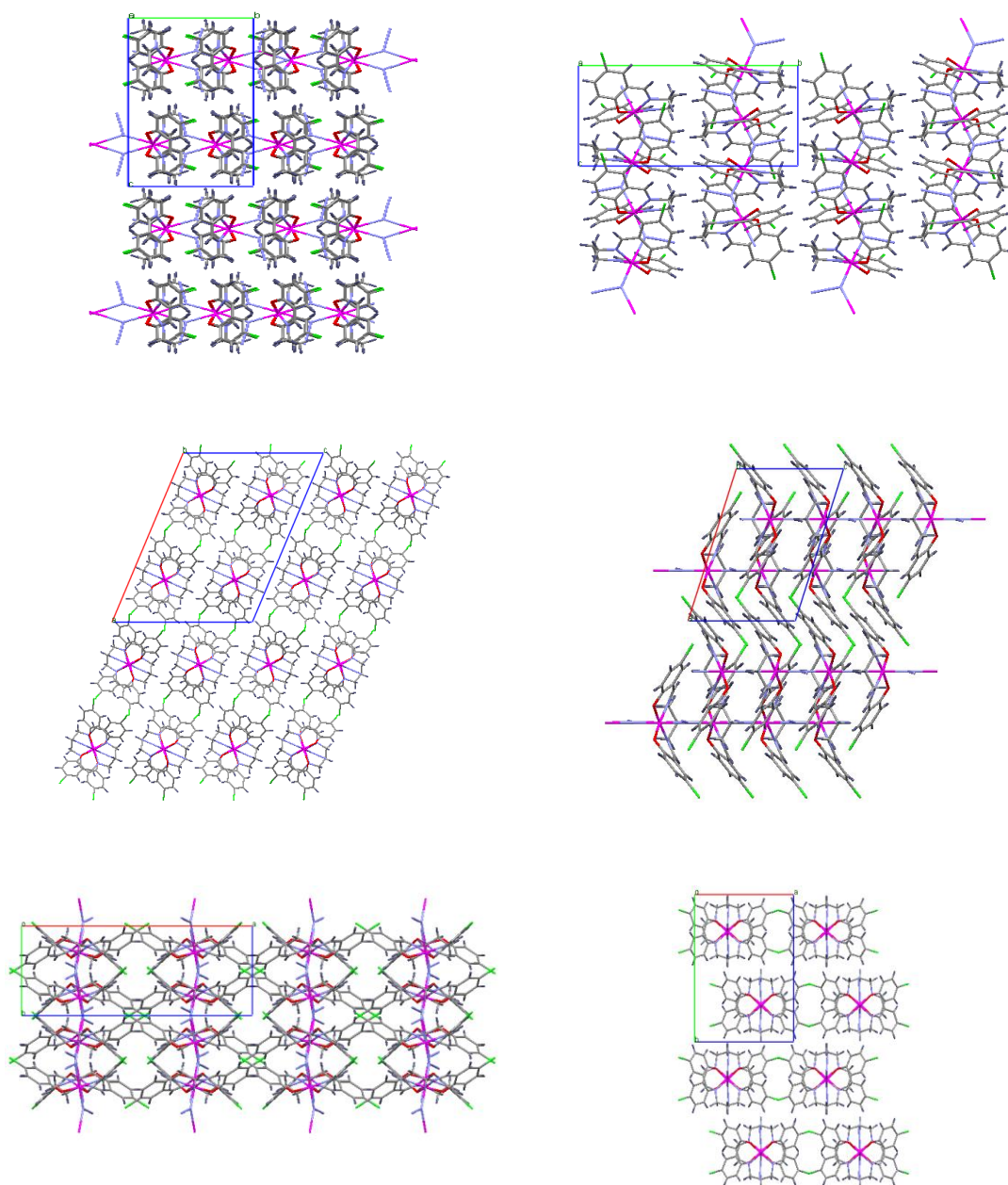
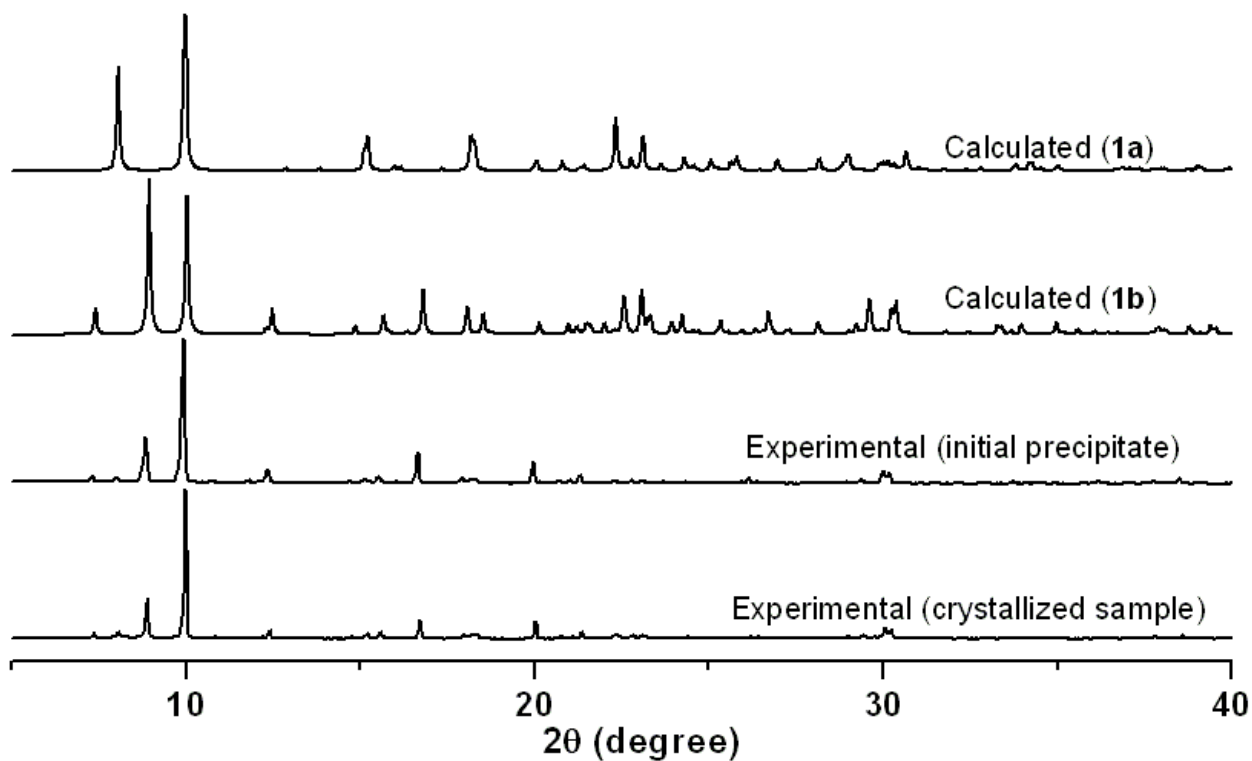


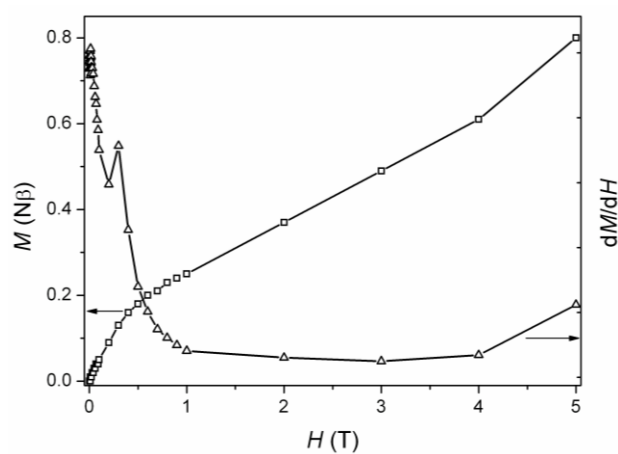
## Supplementary Figures



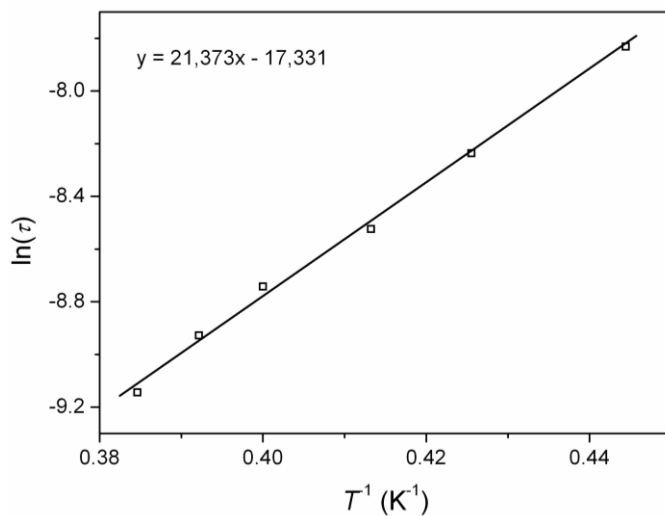
**Fig. S1** Comparison of the different views of the packing of the chain polymers in the two polymorphs, **1a** (left) and **1b** (right). From top to bottom, the views are projections on *bc*-plane, *ac*-plane and *ab*-plane, respectively. Colour code for atoms: blue, N; red, O; grey C; purple, Mn, green Cl.



**Fig. S2** Comparison of calculated powder XRD (**1a**, **1b**) pattern with experimental powder XRD pattern of compound  $[\text{Mn}(\text{5-Clisalpn})\text{N}_3]_n$  (**1**)

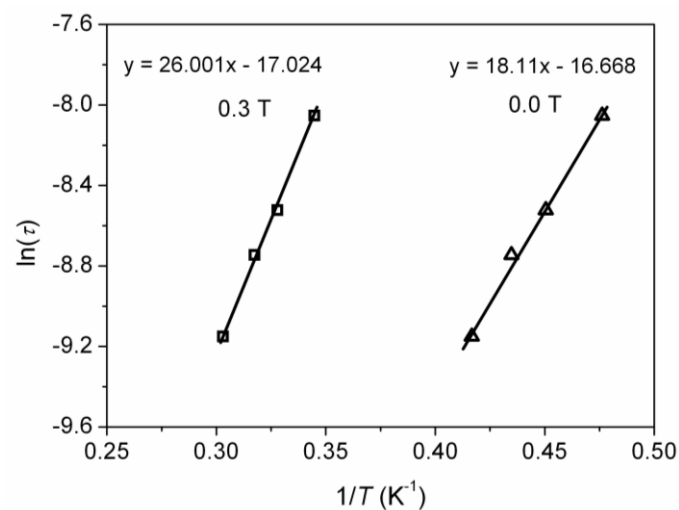


**Fig. S3** Plots of  $M$  versus  $H$  and of the derivative of  $M$  against  $H$  for **1** at 2 K. Solid lines between the experimental data points are shown as a guide to the eye, they do not result from fitting.



**Fig. S4** Plot of the logarithm of the relaxation time  $\tau$  versus  $1/T$  for **1** at zero dc field yielding

$U_{\text{eff}} = 21.4$  K and  $\tau_0 = 2.97 \times 10^{-8}$  s.



**Fig. S5** Plot of the relaxation time  $\tau_0$  versus  $1/T$  for **2** at zero ( $U_{\text{eff}} = 18.2$  K,  $\tau_0 = 5.77 \cdot 10^{-8}$  s) and 0.3 T dc field ( $U_{\text{eff}} = 26.0$  K,  $\tau_0 = 4.05 \cdot 10^{-8}$  s).