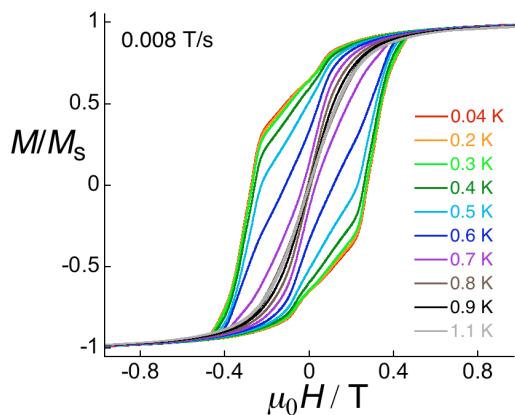


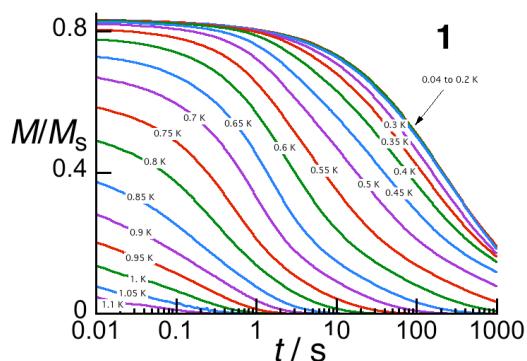
# Supporting Information

## One-dimensional coordination polymers of antiferromagnetically-coupled [Mn<sub>4</sub>] Single-Molecule Magnets

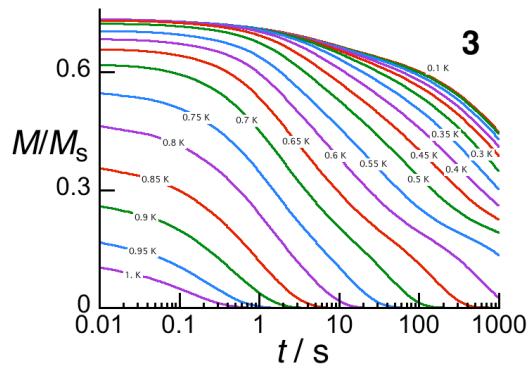
Lollita Lecren, Olivier Roubeau, Yang-Guang Li, Xavier Le Goff, Hitoshi Miyasaka, Florent Richard, Wolfgang Wernsdorfer, Claude Coulon, and Rodolphe Clérac



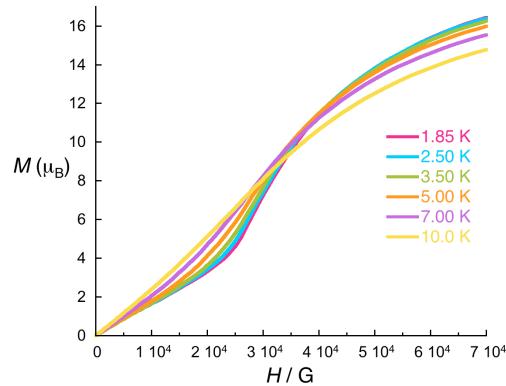
**Figure S1.**  $M$  vs.  $H$  hysteresis loops measured on an oriented single crystal of **3** at different temperatures with a field sweep rate of 0.008 T/s. The applied field is here oriented in the average easy direction of the single crystal. The data are normalized to the saturation magnetization ( $M_S$ ) at 1.4 T.



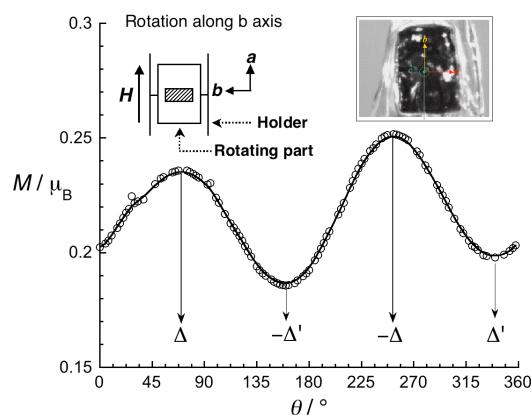
**Figure S2.** Relaxation of the magnetization of **1** at different temperatures. Data are normalized to the saturation magnetization ( $M_S$ ) at 1.4 T.



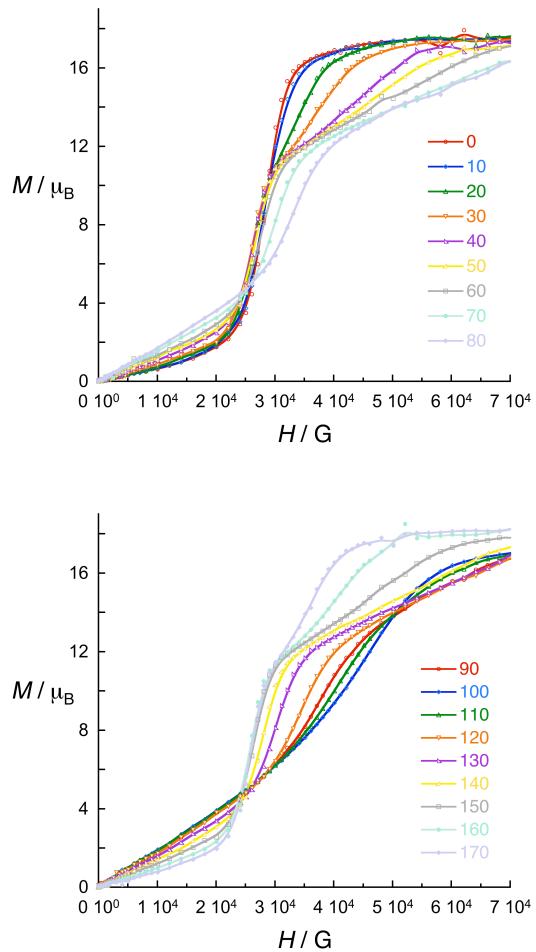
**Figure S3.** Relaxation of the magnetization of **3** at different temperatures. Data are normalized to the saturation magnetization ( $M_S$ ) at 1.4 T.



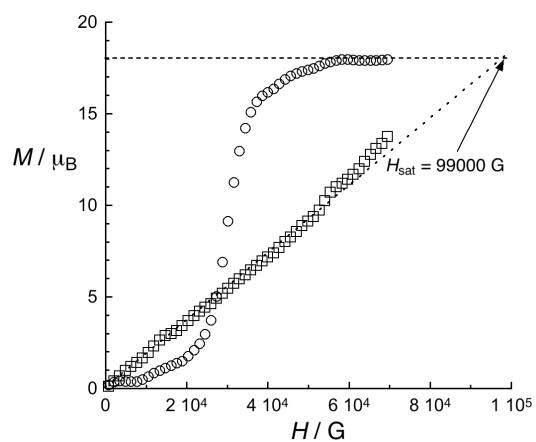
**Figure S4.** Magnetization vs dc magnetic field data on a powder sample of **4** at increasing temperatures.



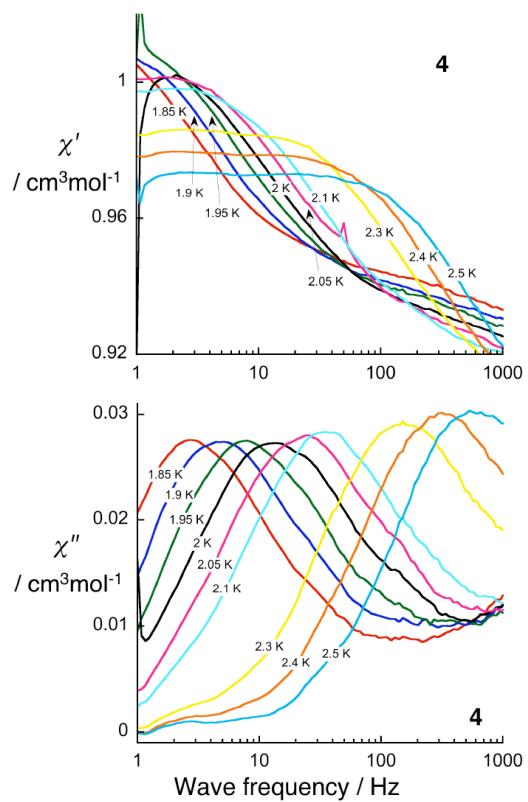
**Figure S5.** Rotation figure around the monoclinic magnetic  $b$  axis for a single crystal of **4** at 1000 Oe and 1.8 K.



**Figure S6.** Magnetization vs. dc magnetic field data for various orientations ( $\square$ , defined in the text) of the applied field in the  $(b, \Delta)$  plane perpendicular to  $\Delta'$  hard axis of a single crystal of **4** at 1.8 K.



**Figure S7.** Magnetization vs. dc magnetic field data in the easy and hard directions for a single crystal of **5** at 1.85 K. Extrapolation of the hard direction data to the saturation value of  $18 \mu_B$  gives a saturation field  $H_{\text{sat}}$  of 9.9 T.



**Figure S8.** Frequency dependence of the real (top) and imaginary (bottom) ac susceptibilities of **4** at various temperatures.

**Analytical expression used for the fitting of temperature dependent magnetic properties of [Mn<sub>4</sub>].**

Van Vleck expression of the susceptibility:

$$\chi_{tetra} = \frac{\sum S_T(S_T+1)(2S_T+1)\exp(-E(S_T)/k_B T)}{\sum (2S_T+1)\exp(-E(S_T)/k_B T)}$$

with

$$E(S_T) = \sum J_{wb} [S_T(S_T+1) \sum S_A(S_A+1) \sum S_B(S_B+1)] \sum J_{bb} [S_A(S_A+1)]$$

and

| S | S <sub>T</sub> | S <sub>B</sub> | S <sub>A</sub> | J <sub>bb</sub> | J <sub>wb</sub> | Numerator | Denominator |
|---|----------------|----------------|----------------|-----------------|-----------------|-----------|-------------|
| 9 | 9              | 5              | 4              | 40              | 20              | 1710      | 19          |
| 8 | 8              | 5              | 4              | 22              | 20              | 1224      | 17          |
| 8 | 8              | 5              | 3              | 30              | 12              | 1224      | 17          |
| 8 | 8              | 4              | 4              | 32              | 20              | 1224      | 17          |
| 7 | 7              | 5              | 4              | 6               | 20              | 840       | 15          |
| 7 | 7              | 5              | 3              | 14              | 12              | 840       | 15          |
| 7 | 7              | 5              | 2              | 20              | 60              | 840       | 15          |
| 7 | 7              | 4              | 4              | 16              | 20              | 840       | 15          |
| 7 | 7              | 4              | 3              | 24              | 12              | 840       | 15          |
| 7 | 7              | 3              | 4              | 24              | 20              | 840       | 15          |
| 6 | 6              | 5              | 4              | -8              | 20              | 546       | 13          |
| 6 | 6              | 5              | 3              | 0               | 12              | 546       | 13          |
| 6 | 6              | 5              | 2              | 6               | 6               | 546       | 13          |
| 6 | 6              | 5              | 1              | 10              | 2               | 546       | 13          |
| 6 | 6              | 4              | 4              | 2               | 20              | 546       | 13          |
| 6 | 6              | 4              | 3              | 10              | 12              | 546       | 13          |
| 6 | 6              | 4              | 2              | 16              | 6               | 546       | 13          |
| 6 | 6              | 3              | 4              | 10              | 20              | 546       | 13          |
| 6 | 6              | 3              | 3              | 18              | 12              | 546       | 13          |
| 6 | 6              | 2              | 4              | 16              | 20              | 546       | 13          |
| 5 | 5              | 5              | 4              | -20             | 20              | 330       | 11          |
| 5 | 5              | 5              | 3              | -12             | 12              | 330       | 11          |
| 5 | 5              | 5              | 2              | -6              | 6               | 330       | 11          |
| 5 | 5              | 5              | 1              | -2              | 2               | 330       | 11          |
| 5 | 5              | 5              | 0              | 0               | 0               | 330       | 11          |
| 5 | 5              | 4              | 4              | -10             | 20              | 330       | 11          |
| 5 | 5              | 4              | 3              | -2              | 12              | 330       | 11          |
| 5 | 5              | 4              | 2              | 4               | 6               | 330       | 11          |
| 5 | 5              | 4              | 1              | 8               | 2               | 330       | 11          |

|   |   |   |   |     |    |     |    |
|---|---|---|---|-----|----|-----|----|
| 5 | 5 | 3 | 4 | -2  | 20 | 330 | 11 |
| 5 | 5 | 3 | 3 | 6   | 12 | 330 | 11 |
| 5 | 5 | 3 | 2 | 12  | 6  | 330 | 11 |
| 5 | 5 | 2 | 4 | 4   | 20 | 330 | 11 |
| 5 | 5 | 2 | 3 | 12  | 12 | 330 | 11 |
| 5 | 5 | 1 | 4 | 8   | 20 | 330 | 11 |
| 4 | 4 | 5 | 4 | -30 | 20 | 180 | 9  |
| 4 | 4 | 5 | 3 | -22 | 12 | 180 | 9  |
| 4 | 4 | 5 | 2 | -16 | 6  | 180 | 9  |
| 4 | 4 | 5 | 1 | -12 | 2  | 180 | 9  |
| 4 | 4 | 4 | 4 | -20 | 20 | 180 | 9  |
| 4 | 4 | 4 | 3 | -12 | 12 | 180 | 9  |
| 4 | 4 | 4 | 2 | -6  | 6  | 180 | 9  |
| 4 | 4 | 4 | 1 | -2  | 2  | 180 | 9  |
| 4 | 4 | 4 | 0 | 0   | 0  | 180 | 9  |
| 4 | 4 | 3 | 4 | -12 | 20 | 180 | 9  |
| 4 | 4 | 3 | 3 | -4  | 12 | 180 | 9  |
| 4 | 4 | 3 | 2 | 2   | 6  | 180 | 9  |
| 4 | 4 | 3 | 1 | 6   | 2  | 180 | 9  |
| 4 | 4 | 2 | 4 | -6  | 20 | 180 | 9  |
| 4 | 4 | 2 | 3 | 2   | 12 | 180 | 9  |
| 4 | 4 | 2 | 2 | 8   | 6  | 180 | 9  |
| 4 | 4 | 1 | 4 | -2  | 20 | 180 | 9  |
| 4 | 4 | 1 | 3 | 6   | 12 | 180 | 9  |
| 4 | 4 | 0 | 4 | 0   | 20 | 180 | 9  |
| 3 | 3 | 5 | 4 | -38 | 20 | 84  | 7  |
| 3 | 3 | 5 | 3 | -30 | 12 | 84  | 7  |
| 3 | 3 | 5 | 2 | -24 | 6  | 84  | 7  |
| 3 | 3 | 4 | 4 | -28 | 20 | 84  | 7  |
| 3 | 3 | 4 | 3 | -20 | 12 | 84  | 7  |
| 3 | 3 | 4 | 2 | -14 | 6  | 84  | 7  |
| 3 | 3 | 4 | 1 | -10 | 2  | 84  | 7  |
| 3 | 3 | 3 | 4 | -20 | 20 | 84  | 7  |
| 3 | 3 | 3 | 3 | -12 | 12 | 84  | 7  |
| 3 | 3 | 3 | 2 | -6  | 6  | 84  | 7  |
| 3 | 3 | 3 | 1 | -2  | 2  | 84  | 7  |
| 3 | 3 | 3 | 0 | 0   | 0  | 84  | 7  |
| 3 | 3 | 2 | 4 | -14 | 20 | 84  | 7  |
| 3 | 3 | 2 | 3 | -12 | 12 | 84  | 7  |
| 3 | 3 | 2 | 2 | 0   | 6  | 84  | 7  |
| 3 | 3 | 2 | 1 | 4   | 2  | 84  | 7  |
| 3 | 3 | 1 | 4 | -10 | 20 | 84  | 7  |
| 3 | 3 | 1 | 3 | -2  | 12 | 84  | 7  |
| 3 | 3 | 1 | 2 | 4   | 6  | 84  | 7  |
| 3 | 3 | 0 | 3 | 0   | 12 | 84  | 7  |
| 2 | 2 | 5 | 4 | -44 | 20 | 30  | 5  |
| 2 | 2 | 5 | 3 | -36 | 12 | 30  | 5  |
| 2 | 2 | 4 | 4 | -34 | 20 | 30  | 5  |
| 2 | 2 | 4 | 3 | -26 | 12 | 30  | 5  |
| 2 | 2 | 4 | 2 | -20 | 6  | 30  | 5  |

