## Two Azido-Bridged Homospin Fe(II)/Co(II) Coordination Polymers Featuring Single-Chain Magnet Behavior

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T/K	au / s	α
3.4	5.13 × 10 <sup>-2</sup>	0.59
3.5	$1.95 \times 10^{-2}$	0.55
3.6	9.59 × 10 <sup>-3</sup>	0.52
3.7	$5.15 \times 10^{-3}$	0.49
3.8	$2.99  imes 10^{-3}$	0.48
3.9	$1.73 \times 10^{-3}$	0.48
4.0	$9.98  imes 10^{-4}$	0.47
4.1	$5.74 \times 10^{-4}$	0.47
4.2	3.41 × 10 <sup>-4</sup>	0.47
4.3	$1.86 \times 10^{-4}$	0.48
4.4	$9.05  imes 10^{-5}$	0.48
4.5	$4.32 \times 10^{-5}$	0.48
4.6	$1.96 \times 10^{-5}$	0.47

Table S1. The fitting results for 1 under zero dc field by a generalized Debye model.

 Table S2. The fitting results for 2 under zero dc field by a generalized Debye model.

T/K	τ / s	α	
2.0	$1.06 \times 10^{-1}$	0.66	
2.1	5.46 × 10 <sup>-2</sup>	0.65	
2.2	$2.82 \times 10^{-2}$	0.64	
2.3	1.36 × 10 <sup>-2</sup>	0.63	
2.4	6.66 × 10 <sup>-3</sup>	0.62	
2.5	3.33 × 10 <sup>-3</sup>	0.60	
2.6	$1.71 \times 10^{-3}$	0.59	
2.7	$8.82 \times 10^{-4}$	0.58	
2.8	$4.43 \times 10^{-4}$	0.57	
2.9	$2.05 \times 10^{-4}$	0.56	

T/K	τ / s	α
3.5	$1.36 \times 10^{-1}$	0.52
3.6	$6.19 \times 10^{-2}$	0.50
3.7	$2.96 \times 10^{-2}$	0.47
3.8	$1.75 \times 10^{-2}$	0.47
3.9	$1.07 \times 10^{-2}$	0.48
4.0	$6.23 \times 10^{-3}$	0.47
4.1	$3.96 \times 10^{-3}$	0.49
4.2	$2.51 \times 10^{-3}$	0.51
4.3	$1.45 \times 10^{-3}$	0.55
4.4	$6.99  imes 10^{-4}$	0.61
4.5	$3.34 \times 10^{-4}$	0.65

**Table S3**. The fitting results for 1 at 3.5-4.5 K under 1000 Oe dc field by a generalized Debye model.

**Table S4**. Temperature dependence of the relaxation time for **1** at 6.59-7.21 K obtained from  $\chi''$  vs *T* plots under 1000 Oe dc field.

T / K	au / s
6.59	1.61 × 10 <sup>-3</sup>
6.79	$8.12  imes 10^{-4}$
6.99	$3.21 \times 10^{-4}$
7.11	$2.29 \times 10^{-4}$
7.21	$1.59 \times 10^{-4}$



**Figure S1**. The packing diagram of 1 showing edge-to-edge (green line) and face-to-face (violet line)  $\pi \cdots \pi$  interactions. All the counterions and hydrogen atoms are omitted for clarity. Colour codes: Fe(II), orange; O, red; C, grey; N, light blue.



**Figure S2**. The packing diagram of **2** showing edge-to-edge (green line) and edge-to-face (orange line)  $\pi \cdots \pi$  interactions. All the counterions, lattice solvents and hydrogen atoms are omitted for clarity. Colour codes: Co(II), violet; O, red; C, grey; N, light blue.



Figure S3. The experimental and calculated powder XRD patterns for 1.



Figure S4. The experimental and calculated powder XRD patterns for 2.



**Figure S5.** Temperature dependent  $\chi^{-1}$  plots for 1 measured at 1000 Oe dc field. The red lines represent the Curie-Weiss fit to the data.



**Figure S6.** Temperature dependent  $\chi^{-1}$  plots for **2** measured at 1000 Oe dc field. The red lines represent the Curie-Weiss fit to the data.



**Figure S7.** Field dependence of the magnetization for **1** between 2 and 15 K. Solid lines are guides for the eyes.



**Figure S8.** First field derivative of the magnetization as a function of the applied dc field for **1** between 2 and 15 K. Solid lines are guides for the eyes.



**Figure S9.** Temperature dependence of the magnetic susceptibility for **1** as a function of applied dc field. Solid lines are guides for the eyes.



**Figure S10.** (*T*, *H*) phase diagram for **1**. The data are obtained from the location of the maximum for dM/dH vs *H* plots (•) and  $\chi$  vs *T* plots (•). The line is guide for the eyes.



**Figure S11.** Field dependence of the magnetization for **2** between 2 and 5 K. Solid lines are guides for the eyes.



**Figure S12.** ZFC and FC magnetization versus temperature curves of **1** measured with an applied dc field of 50 Oe. Solid lines are guides for the eyes.



**Figure S13.** ZFC and FC magnetization versus temperature curves of **2** measured with an applied dc field of 50 Oe. Solid lines are guides for the eyes.



**Figure S14.** Temperature dependence of the in-phase and out-of-phase ac susceptibility for **1** under zero dc field. Solid lines are guides for the eyes.



**Figure S15.** Temperature dependence of the in-phase and out-of-phase ac susceptibility for **2** under zero dc field. Solid lines are guides for the eyes.



**Figure S16.** Frequency dependence of the in-phase and out-of-phase ac susceptibility for **1** under zero dc field. Solid lines are guides for the eyes.



**Figure S17.** Frequency dependence of the in-phase and out-of-phase ac susceptibility for **2** under zero dc field. Solid lines are guides for the eyes.



Figure S18. Cole-Cole plots of 1 under zero dc field. The lines represent the fit to the data.



Figure S19. Cole-Cole plots of 2 under zero dc field. The lines represent the fit to the data.



Figure S20. Temperature dependence of the relaxation time for 1 under zero dc field. The lines represent the fit by Arrhenius Law.



**Figure S21.** Temperature dependence of the relaxation time for **2** under zero dc field. The lines represent the fit by Arrhenius Law.



**Figure S22.** Temperature dependence of the in-phase and out-of-phase ac susceptibility for **1** under 1000 Oe dc field. Solid lines are guides for the eyes.



**Figure S23.** Frequency dependence of the in-phase and out-of-phase ac susceptibility for 1 at 3.5-5.0 K (a, b) and 5.2-7.6 K (c, d) under 1000 Oe dc field. Solid lines are guides for the eyes.



**Figure S24.** Cole-Cole plots of **1** at 3.5-4.5 K with an applied 1000 Oe dc field. The lines represent the fit to the data.



**Figure S25.** Temperature dependence of the relaxation time for **1** at 3.5-4.5 K under 1000 Oe dc field. The lines represent the fit by Arrhenius Law.



**Figure S26.** Temperature dependence of the relaxation time for **1** obtained from  $\chi''$  vs *T* plots via  $\tau^1 = 2\pi v$  under 1000 Oe dc field. The lines represent the fit by Arrhenius Law.



**Figure S27.** The magnetic hysteresis loops of **1** measured between 2 and 4 K with a field sweep rate of 20 Oe/s. Solid lines are guides for the eyes.



**Figure S28.** The magnetic hysteresis loops of **2** measured between 2 and 3 K with a field sweep rate of 20 Oe/s. Solid lines are guides for the eyes. Inset: Enlarged hysteresis loops of **1** at 2-3 K.