Electronic Supplementary Information

Spin Crossover and Photomagnetic Behaviors in One-Dimensional

Looped Coordination Polymers

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Section1. Powder X-ray diffraction patterns



Fig. S1 Experimental and simulated PXRD patterns of compound 1.



Fig. S2 Experimental and simulated PXRD patterns of compound 2.



Fig. S3 Experimental and simulated PXRD patterns of compound 3.

Section2. Thermogravimetric analyses



Fig. S4 TG curves of compounds 1, 2 and 3.

Section3. Additional structural figures



Fig. S5 The asymmetric unit (a), the crystal packing mode in the *bc* plane (b), the structure of chain Fe1 (c), and the crystal color at 100 and 240 K (d) in compound **3**. Color code, Fe: sky-blue, S: yellow, N: blue, C: grey, Se: violet. Hydrogen atoms are omitted for clarity.



Fig. S6 ORTEP plot with 50% thermal ellipsoids of compounds **1** (a, b), **2** (c, d) and **3** (e, f). Color code, Fe: sky-blue, S: yellow, N: blue, C: grey, B: pink, Se: violet. Hydrogen atoms are omitted for clarity.



Void volume 12.16 Å³ 0.3 % of unit cell volume

Fig. S7 View of the voids in the framework of compound 1.



Void volume 517.09 Å³ 16.9 % of unit cell volume

Fig. S8 View of the voids in the framework of compound 2.



Void volume 287.23 Å³ 31.3 % of unit cell volume

Fig. S9 View of the voids in the framework of compound 3.



Fig. S10 The Fe^{...}Fe distance in intra-chain (red) and the nearest Fe^{...}Fe distance in inter-chain (blue) at 120 and 293 K of compound **1**.



Fig. S11 The Fe^{...}Fe distance in intra-chain (red) and the nearest Fe^{...}Fe distance in inter-chain (blue) at 100 and 293 K of compound **2**.



Fig. S12 The Fe^{...}Fe distance in intra-chain (red) and the nearest Fe^{...}Fe distance in inter-chain (blue) at 100 and 240 K of compound **3**.

Section4. Magnetic properties and DSC measurement



Fig. S13 The $\chi_M T$ versus *T* plots (per Fe unit) of compound 1 in the cooling and heating modes between 300 and 2.0 K.



Fig. S14 The $\chi_M T$ versus *T* plots (per Fe unit) of compound **2** in the cooling and heating modes between 300 and 2.0 K.



Fig. S15 The exo- and endo-thermic broad peaks of compound 1 (Because of the gradual SCO behavior, the phase transition signal of compound 2 was not observed).



Fig. S16 The $\chi_M T$ versus *T* plots (per Fe unit) of compound **3** from 270 to 2.0 K.



Section5. IR spectra

Fig. S17 IR spectra of compounds 1, 2 and 3.

Wavenumber (cm⁻¹)



Fig.S18 Variable-temperature IR spectra of compounds 1 (a) and 2 (b).



Section6. Structural parameters



Fig.S19 Variable-temperature unit-cell parameters of compounds 1 (a) and 2 (b).

Compound 1	120 K	293 K	
$\langle \mathbf{d}_{ ext{Fe1-N}} angle$, $\mathbf{\AA}^a$	1.977	2.178	
$\langle { m d}_{ m Fe2-N} angle$, ${ m \AA}^a$	2.004	2.182	
$\Sigma_{\rm Fe1},{ m deg}^b$	19.0	24.5	
$\Sigma_{\rm Fe2},{\rm deg}^b$	33.2	36.9	
Compound 2	100 K	293 K	
$\langle \mathbf{d}_{ ext{Fe1-N}} angle$, $\mathbf{\AA}^a$	2.046	2.203	
$\Sigma_{\rm Fe1},{\rm deg}^b$	24.5	30.0	
Compound 3	100 K	240 K	
$\langle \mathbf{d}_{ ext{Fe1-N}} angle$, $\mathbf{\AA}^a$	2.196	2.204	
$\Sigma_{\rm Fe1},{ m deg}^b$	10.80	12.50	

Table. S1 Structural Parameters of compounds 1, 2 and 3.

 $a(d_{\text{Fe-N}})$: the average Fe–N bond lengths. $b\Sigma_{\text{Fe}}$: the sum of the deviations of N–Fe–N angles from 90°.