Supporting Information

Novel two-step Fe-Au type spin-crossover behavior in a Hofmann-like complex Fe(4-methylpyrimidine)$_2$[Au(CN)$_2$]$_2$

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χ_M vs T plot

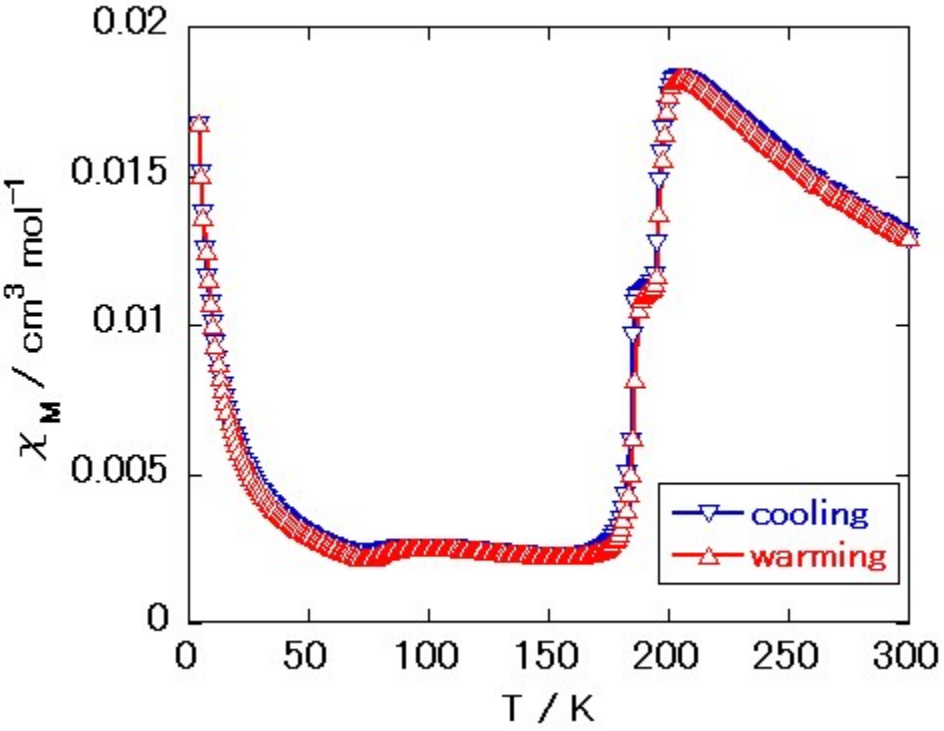


Figure S1. χ_M vs T plot of **1**

Additional crystallographic data

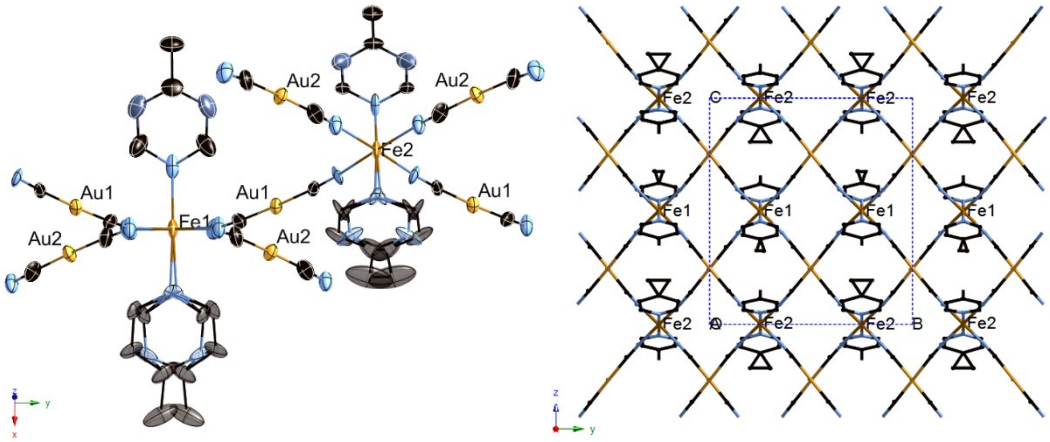
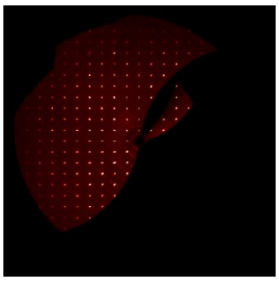
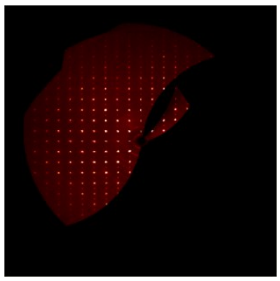
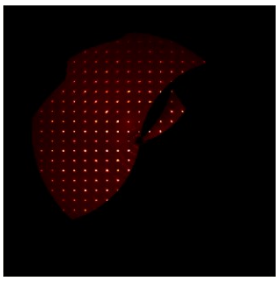
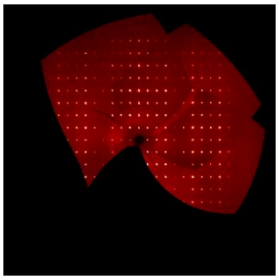
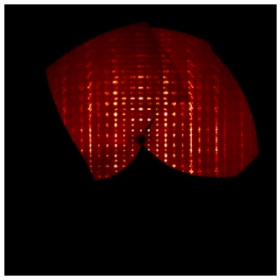
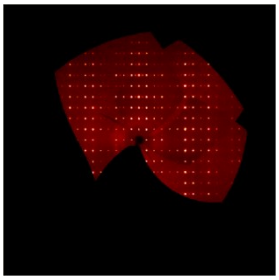
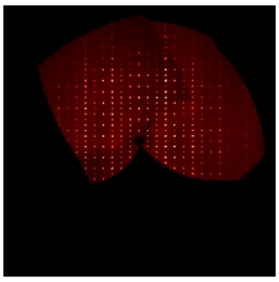
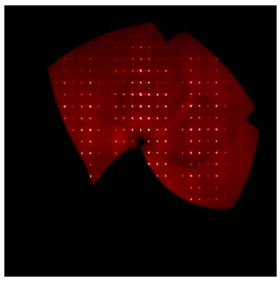
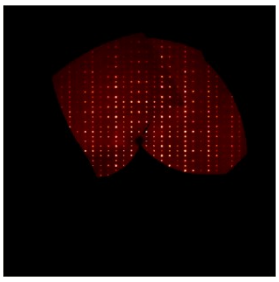


Figure S2. Crystal structure of complex **1** as determined by XRD at 190 K. (Black: C, aqua: N, brown: Fe, yellow: Au, and indigo: disordered between C and N by two orientations.) Left: ORTEP structure. Right: Packing structure along the [001] direction.

Table S1. The crystallographic parameters of **1**

Empirical formula	$C_{14}H_{12}Au_2FeN_8$		
Formula weight	742.1		
Crystal size	0.38 x 0.25 x 0.12 mm^3		
Temperature / K	298(2)	190(2)	150(2)
Crystal system	Orthorhombic	Monoclinic	Orthorhombic
a / Å	9.5856(7)	9.554(4)	9.5273(9)
b / Å	15.5741(11)	13.698(6)	14.8094(15)
c / Å	13.7797(10)	15.262(7)	13.5467(13)
β / deg.	90	90.387(7)	90
V / $Å^3$	2057.1(3)	1997.4(14)	1911.4(3)
Space group	*Pbcm*	$P2_1/m$	*Pbcm*
Z	4	4	4
D_{calc}	2.396	2.468	2.579
F(000)	1344	1344	1344
Reflections collected	15279	14676	14002
Independent reflections	3307 [R_{int} = 0.0368]	6186 [R_{int} = 0.0894]	3082 [R_{int} = 0.0407]
Data/ restraints / parameters	3307 / 6 / 141	6186 / 68 / 254	3082 / 42 / 142
Final R_1, R_w ($I > 2s$)	0.0295, 0.0660	0.0802, 0.1853	0.0231, 0.0507
Final R_1, R_w (all data)	0.0550, 0.0749	0.1673, 0.2247	0.0410, 0.0567
Goodness-of-fit on F^2	1.050	1.114	1.065

Table S2. The reconstructed reciprocal lattice of **1**

	298 K	190 K	150 K
0kl plane			
h0l plane			
hk0 plane			

Powder X-ray Pattern

Powder X-ray diffraction measurement of complex **1** is measured by Rigaku X-RAY DIFFRACTOMETER. The spectrum shows in Figure S2.

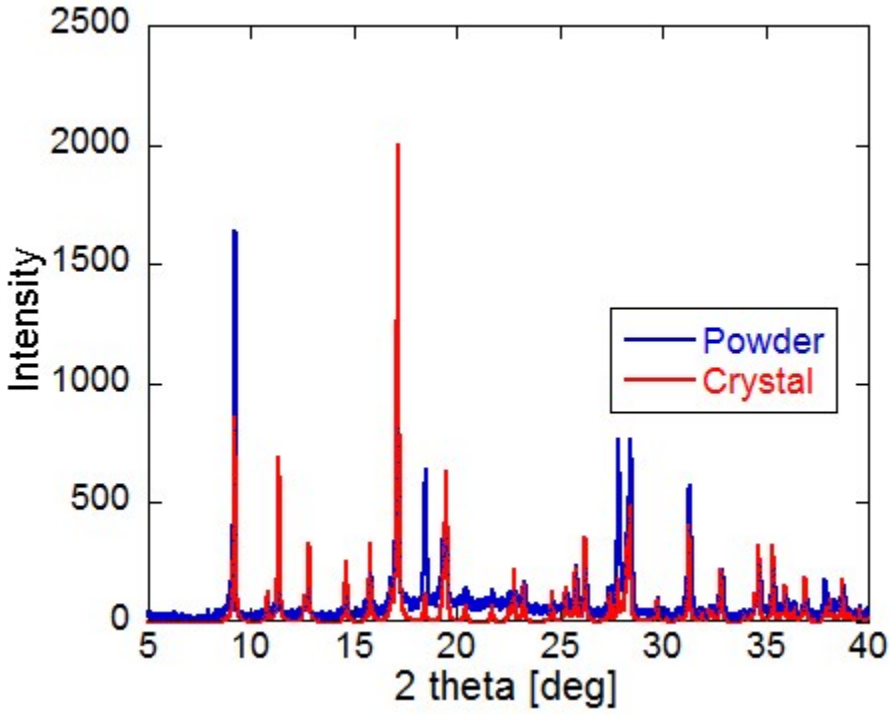


Figure S3. Powder X-ray diffraction measurement of **1** (blue: observed, red: calculated).

TG analysis

Thermal gravimetry of complex **1** is measured by Hitachi High-Technologies Corporation TG/DTA6200. The result shows in Figure S3.

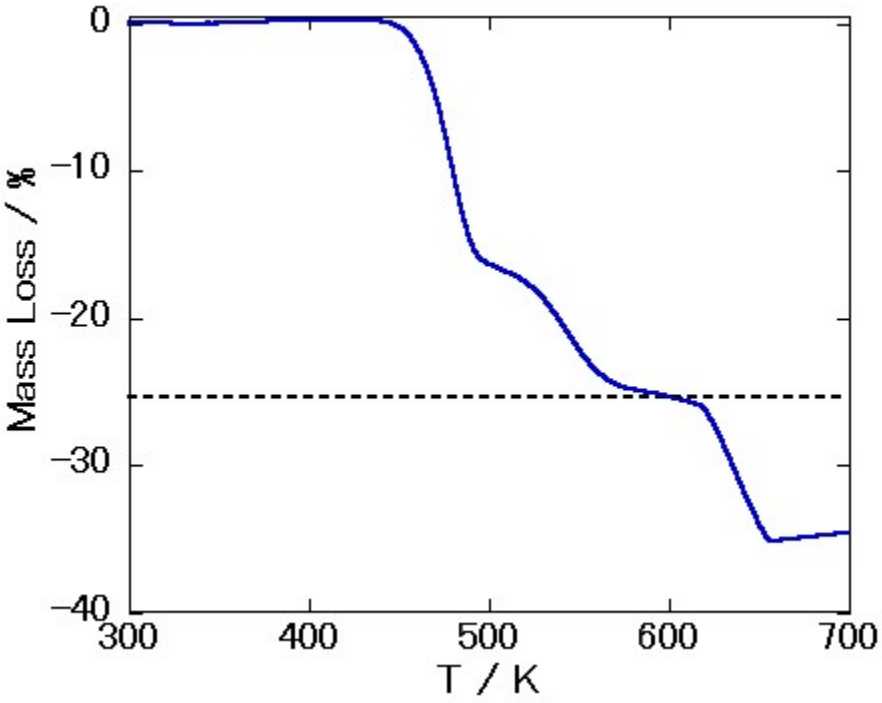


Figure S4. TG analysis of 1.

DSC analysis

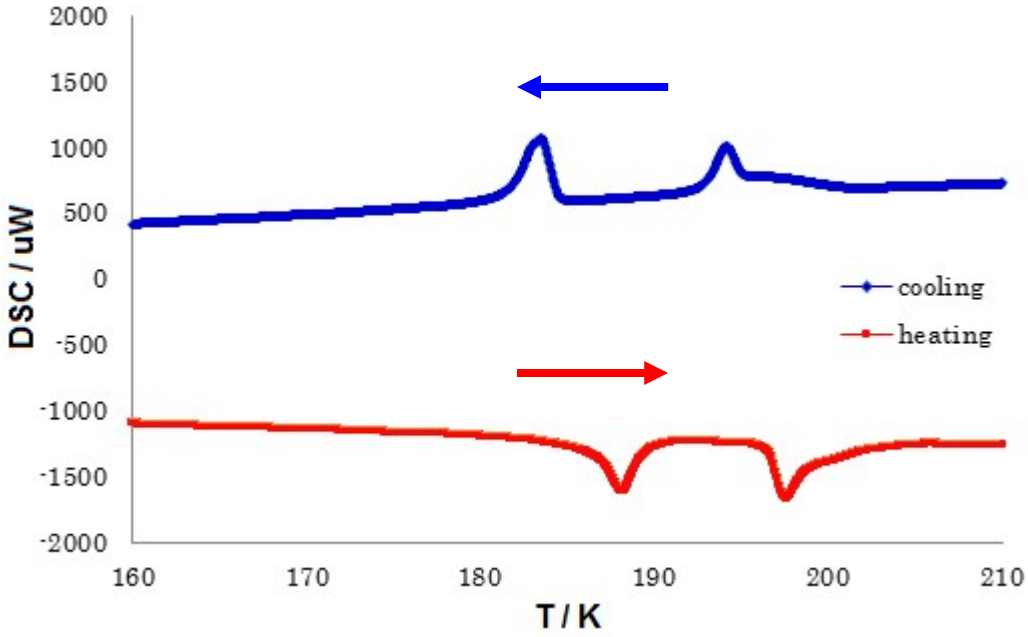


Figure S5. DSC analysis of 1.