Supporting Information

## **Cyanide-Bridged Bimetallic 3D Hoffman-Like Coordination Polymers** with Tunable Magnetic Behaviour

Jin-Yan Li, Zhao-Ping Ni, Zheng Yan, Ze-Min Zhang, Yan-Cong Chen, Wei Liu and Ming-Liang Tong $^*$ 

	300 K	225 K	150K
Fe-N1	2.115(7)	1.946(5)	1.930(5)
Fe-N2	2.128(8)	1.939(5)	1.928(5)
Fe-N3	2.198(7)	2.027(5)	2.005(4)
Ag-C1	2.048(9)	2.066(7)	2.055(6)
Ag-C2 <sup>[a]</sup>	2.060(10)	2.050(7)	2.040(6)
N2 <sup>[b]</sup> -Fe-N1	91.4(3)	90.8(2)	90.77(19)
N2-Fe-N1	88.6(3)	89.2(2)	89.23(19)
N2 <sup>[b]</sup> -Fe-N3	89.5(3)	89.2(2)	89.38(18)
N1-Fe-N3 <sup>[b]</sup>	90.0(3)	89.9(2)	89.81(18)
N2-Fe-N3	90.5(3)	90.8(2)	90.62(18)
N1-Fe-N3	90.0(3)	90.1(2)	90.19(18)
C1-Ag-C2 <sup>[a]</sup>	173.9(5)	173.4(3)	173.2(3)
Fe1-N1-C1	171.1(9)	176.3(6)	176.0(5)
Fe1-N2-C2	169.1(8)	174.0(6)	175.0(5)
Ag-C1-N1	176.1(10)	174.4(6)	173.8(5)
Ag <sup>[c]</sup> -C2-N2	175.4(10)	174.0(7)	171.2(5)

 Table S1. Selected bond lengths [Å] and angles [°] for 1.

Symmetry codes: a) -x+3/2, y+1/2, -z+1/2; b) -x+3/2, -y+1/2, -z+1; c) -x+3/2, y-1/2, -z+1/2.

	273 K	100K
Fe-N1	2.111(9)	1.940(6)
Fe-N2 <sup>[a]</sup>	2.123(9)	1.930(5)
Fe-N3	2.178(10)	2.005(6)
Au-C1	1.961(12)	1.972(6)
Au-C2	1.987(13)	1.971(7)
N2 <sup>[b]</sup> -Fe-N1	89.9(4)	89.9(2)
N2 <sup>[b]</sup> -Fe-N1 <sup>[c]</sup>	90.1(4)	90.1(2)
N2 <sup>[b]</sup> -Fe-N3 <sup>[c]</sup>	89.4(4)	89.5(2)
N1-Fe-N3 <sup>[c]</sup>	90.1(4)	90.7(2)
N2 <sup>[b]</sup> -Fe-N3	90.6(4)	90.5(2)
N1-Fe-N3	89.9(4)	89.3(2)
C1-Au-C2	176.7(5)	176.2(3)
Fe1-N1-C1	174.4(12)	178.1(7)
Fe1 <sup>[d]</sup> -N2-C2	167.1(10)	174.2(6)
Au-C1-N1	177.7(15)	175.0(7)
Au-C2-N2	176.4(11)	174.6(7)

Table S2. Selected bond lengths [Å] and angles [°] for 2.

Symmetry codes: a) -x+3/2, y+1/2, -z+3/2; b) x, -y+1, z-1/2; c) -x+3/2, -y+3/2, -z+1; d) -x+3/2, y-1/2, -z+3/2.

Table S3. Selected bond lengths [Å] and angles [°] for 3 at 150 K.

Fe-N1	2.217(3)	Ni-C9	1.880(4)
Fe-N3	2.194(3)	Ni-C10	1.870(3)
Fe-N4 <sup>[a]</sup>	2.144(2)		
N4 <sup>[a]</sup> -Fe-N3	90.89(10)	N3 <sup>[c]</sup> -Fe-N1	95.10(11)
N4 <sup>[a]</sup> -Fe-N3 <sup>[c]</sup>	89.11(10)	N3-Fe-N1	84.90(10)
N4 <sup>[d]</sup> -Fe-N1	90.98(10)	Fe-N3-C9	138.3(3)
N4 <sup>[a]</sup> -Fe-N1	89.02(10)	Fe <sup>[e]</sup> -N4-C10	160.3(3)

Symmetry codes: a) -x+2, y-1/2, -z+1/2; b) x, -y+5/2, z+1/2; c) -x+2, -y+2, -z+1; d) x, -y+5/2, z+1/2; e) -x+2, y+1/2, -z+1/2.

	1			2	
<i>T</i> [K]	300	225	150	273	100
<fe-n></fe-n>	2.147(8)	1.971(5)	1.954(5)	2.137(10)	1.958(6)
Fe-N <sub>ax</sub>	2.198(7)	2.027(5)	2.005(4)	2.178(10)	2.005(6)
Fe-N <sub>eq</sub>	2.122(8)	1.943(5)	1.929(5)	2.117(9)	1.935(6)
Fe···Fe	16.622(2)	16.317(9)	16.240(7)	16.7493(16)	16.4515(8)

Table S4. Selected interatomic distances [Å] at variant temperatures for 1 and 2.



Fig. S1 View of a portion of 3D structure in 1.



Fig. S2 View of a fragment of the 3D interpenetrated framework and the corresponding topological network in 1.



Fig. S3 Thermogravimetric analysis of compounds 1 (left) and 2 (right).



**Fig. S4** Temperature dependence of  $\chi_{\rm M}T$  for **3**.