Supporting Information

An alternative strategy to construct Fe(II)-based MOFs with multifarious structures and magnetic behaviors

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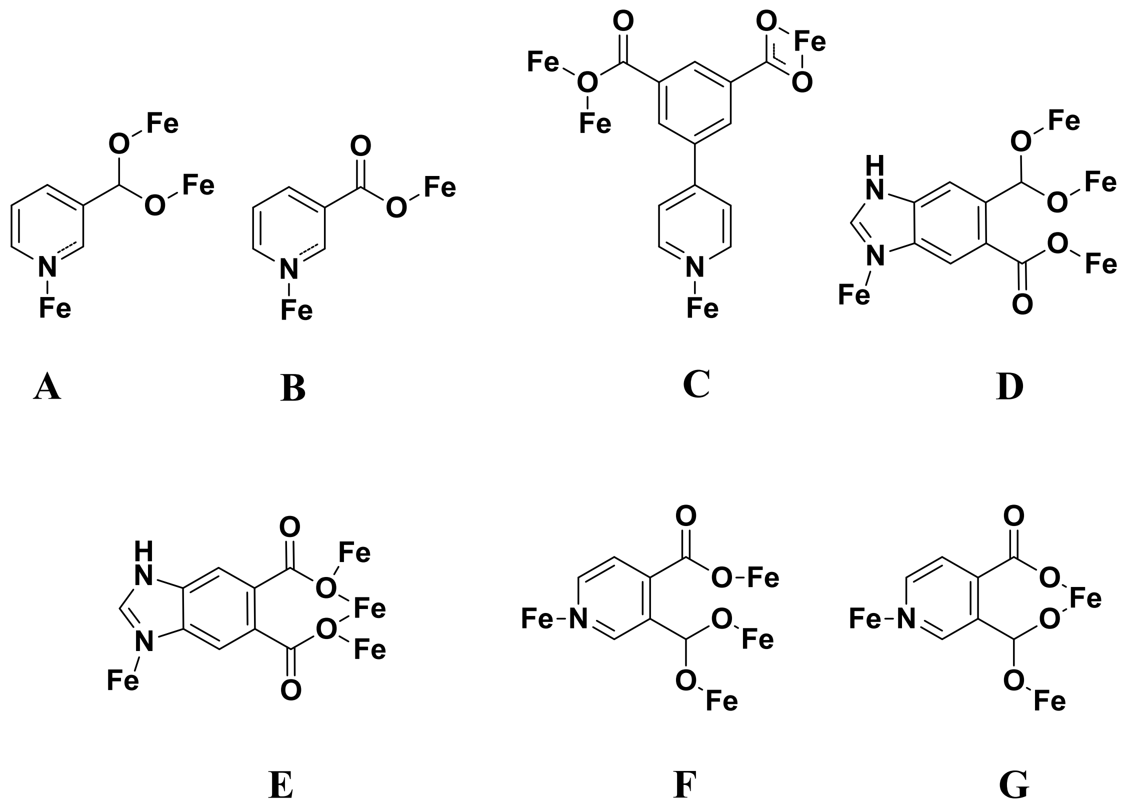
1. Coordination modes



Scheme S1 The coordination modes of heterocyclic carboxylic acids in **1-6**

2. Crystallographic Data Tables

Table S1 Selected bond lengths (Å) and angles (º) for **1**

Fe1—O7^{i}	2.0485 (13)	O8—Fe2^{i}	2.1653 (14)
Fe1—O5^{ii}	2.0955 (13)	O5—Fe1^{ii}	2.0955 (13)
Fe1—O2^{iii}	2.1569 (14)	O7—Fe1^{i}	2.0485 (13)
Fe1—N2	2.1859 (17)	O3—Fe2^{iv}	2.0712 (13)
Fe1—N1	2.1883 (17)	Fe2—N4	2.1855 (17)
Fe1—O9	2.2220 (14)	Fe2—O9	2.2073 (14)
Fe2—O3^{iv}	2.0712 (13)	Fe2—N3	2.2312 (17)
Fe2—O1^{iii}	2.0784 (13)	O1—Fe2^{iii}	2.0784 (13)
Fe2—O8^{i}	2.1653 (14)	O2—Fe1^{iii}	2.1569 (14)
O7^{i}—Fe1—O5^{ii}	179.46 (6)	Fe2—O9—Fe1	112.07 (6)
O7^{i}—Fe1—O2^{iii}	94.12 (6)	O7^{i}—Fe1—N1	91.87 (6)
O5^{ii}—Fe1—O2^{iii}	86.00 (6)	O5^{ii}—Fe1—N1	87.61 (6)
O7^{i}—Fe1—N2	89.48 (6)	O2^{iii}—Fe1—N1	87.69 (6)
O5^{ii}—Fe1—N2	90.42 (6)	N2—Fe1—N1	94.41 (6)
O2^{iii}—Fe1—N2	175.78 (6)	O7^{i}—Fe1—O9	91.53 (5)
O2^{iii}—Fe1—O9	88.64 (5)	O5^{ii}—Fe1—O9	89.00 (5)
N2—Fe1—O9	89.06 (6)	O1^{iii}—Fe2—N4	93.00 (6)
N1—Fe1—O9	175.17 (5)	O8^{i}—Fe2—N4	87.14 (6)
O3^{iv}—Fe2—O1^{iii}	175.79 (6)	O3^{iv}—Fe2—O9	90.16 (5)
O3^{iv}—Fe2—O8^{i}	86.32 (6)	O1^{iii}—Fe2—O9	88.54 (5)
O1^{iii}—Fe2—O8^{i}	97.69 (6)	O8^{i}—Fe2—O9	90.21 (5)
O3^{iv}—Fe2—N4	88.48 (6)	N4—Fe2—O9	177.09 (5)
O9—Fe2—N3	90.94 (6)	O3^{iv}—Fe2—N3	88.63 (6)
C6—O1—Fe2^{iii}	124.37 (12)	O1^{iii}—Fe2—N3	87.38 (6)
C6—O2—Fe1^{iii}	126.24 (12)	O8^{i}—Fe2—N3	174.83 (6)
C13—N3—Fe2	118.31 (13)	N4—Fe2—N3	91.59 (6)

Symmetry codes: (i) $-x-1, -y+1, -z-1$; (ii) $-x-1, -y+2, -z$; (iii) $-x, -y+2, -z$; (iv) $-x-1, -y+2, -z-1$.

Table S2 Selected bond lengths (Å) and angles (º) for **2**

Fe1—O1^{i}	2.0339 (18)	O1—Fe1^{v}	2.0339 (18)
Fe1—O4^{ii}	2.0820 (18)	O1—Fe1^{iii}	2.3313 (18)

Fe1—N1	2.128 (2)	O4—Fe1^{iv}	2.0820 (18)
Fe1—O6	2.1353 (19)	O3—Fe1^{iv}	2.3480 (18)
Fe1—O1^{iii}	2.3313 (18)	Fe1—O3^{ii}	2.3480 (18)
O1^{i}—Fe1—O4^{ii}	159.21 (7)	C1—O1—Fe1^{v}	123.98 (14)
O1^{i}—Fe1—N1	106.51 (7)	C1—O1—Fe1^{iii}	122.77 (14)
O4^{ii}—Fe1—N1	92.62 (7)	Fe1^{v}—O1—Fe1^{iii}	102.96 (7)
O1^{i}—Fe1—O6	95.90 (7)	O1^{i}—Fe1—O1^{iii}	77.04 (7)
O4^{ii}—Fe1—O6	90.85 (7)	O4^{ii}—Fe1—O1^{iii}	93.88 (6)
N1—Fe1—O6	93.61 (8)	N1—Fe1—O1^{iii}	94.02 (8)
O6—Fe1—O3^{ii}	89.59 (8)	O6—Fe1—O1^{iii}	170.82 (6)
O1^{iii}—Fe1—O3^{ii}	86.12 (7)	O1^{i}—Fe1—O3^{ii}	101.26 (7)
C8—O4—Fe1^{iv}	95.84 (13)	O4^{ii}—Fe1—O3^{ii}	59.04 (6)
C11—N1—C12	117.2 (2)	N1—Fe1—O3^{ii}	151.55 (7)
C11—N1—Fe1	121.24 (15)	O4—C8—O3	121.2 (2)
C12—N1—Fe1	121.19 (15)	C8—O3—Fe1^{iv}	83.63 (13)

Symmetry codes: (i) $x-1$, $y-1$, $z-1$; (ii) $x-1$, y, $z-1$; (iii) $-x+2$, $-y+2$, $-z+1$; (iv) $x+1$, y, $z+1$; (v) $x+1$, $y+1$, $z+1$.

Table S3 Selected bond lengths (Å) and angles (º) for **3**

Fe1—O2^{i}	1.999 (2)	N2—Fe1^{iii}	2.144 (2)
Fe1—O1^{ii}	2.047 (3)	O2—Fe1^{iv}	1.999 (2)
Fe1—O4	2.125 (2)	O1—Fe1^{ii}	2.047 (3)

Fe1—O5	2.141 (2)	Fe1—N2^{iii}	2.144 (2)
O2^{i}—Fe1—O1^{ii}	145.32 (7)	O2^{i}—Fe1—N2^{iii}	103.58 (9)
O2^{i}—Fe1—O4	87.67 (8)	O1^{ii}—Fe1—N2^{iii}	110.88 (9)
O1^{ii}—Fe1—O4	86.30 (8)	O4—Fe1—N2^{iii}	106.56 (8)
O2^{i}—Fe1—O5	92.98 (9)	O5—Fe1—N2^{iii}	89.75 (8)
O1^{ii}—Fe1—O5	83.55 (9)	C8—O4—Fe1	134.36 (15)
O4—Fe1—O5	163.06 (7)	Fe1—O5—H5A	120.0
C7—N2—Fe1^{iii}	116.88 (16)	Fe1—O5—H5B	120.0
C5—N2—Fe1^{iii}	137.88 (16)	H5A—O5—H5B	120.0
C9—O1—Fe1^{ii}	134.90 (17)	C9—O2—Fe1^{iv}	131.62 (18)

Symmetry codes: (i) x, y−1, z; (ii) −x+1, −y, −z+1; (iii) −x, −y, −z+1; (iv) x, y+1, z.

Table S4 Selected bond lengths (Å) and angles (º) for **4**

Fe1—O2^{i}	2.051 (2)	O1—Fe1^{iv}	2.1145 (19)
Fe1—O3^{ii}	2.0897 (19)	O2—Fe1^{i}	2.051 (2)
Fe1—N1	2.102 (2)	O3—Fe1^{ii}	2.0897 (19)
Fe1—O1^{iii}	2.1145 (19)	O3—Fe1^{iv}	2.1365 (19)
Fe1—O3^{iii}	2.1365 (19)		
O2^{i}—Fe1—O3^{ii}	104.34 (8)	N1—Fe1—O1^{iii}	91.88 (8)
O2^{i}—Fe1—N1	93.42 (8)	O2^{i}—Fe1—O3^{iii}	88.78 (8)
O3^{ii}—Fe1—N1	102.39 (8)	O3^{ii}—Fe1—O3^{iii}	119.68 (4)
O2^{i}—Fe1—O1^{iii}	167.54 (8)	N1—Fe1—O3^{iii}	135.89 (8)
O3^{ii}—Fe1—O1^{iii}	85.48 (7)	O1^{iii}—Fe1—O3^{iii}	79.59 (7)
C10—O3—Fe1^{ii}	125.34 (16)	C9—O1—Fe1^{iv}	119.12 (16)
C10—O3—Fe1^{iv}	113.87 (15)	C9—O2—Fe1^{i}	117.54 (17)
Fe1^{ii}—O3—Fe1^{iv}	118.68 (8)	C2—N1—Fe1	131.88 (18)
C1—N1—Fe1	121.94 (19)		

Symmetry codes: (i) −*x*+1, −*y*, −*z*+2; (ii) −*x*+1, −*y*+1, −*z*+2; (iii) *x*+1/2, −*y*+1/2, *z*−1/2; (iv) *x*−1/2, −*y*+1/2, *z*+1/2.

Table S5 Selected bond lengths (Å) and angles (º) for **5**

Fe—O3^{i}	2.023 (2)	Fe—O2^{iii}	2.280 (2)
Fe—O5	2.109 (3)	O2—Fe^{iii}	2.280 (2)
Fe—O6	2.145 (2)	O1—Fe^{iv}	2.189 (2)
Fe—N	2.168 (2)	O3—Fe^{v}	2.023 (2)
O3^{i}—Fe—O5	97.03 (10)	C3—N—Fe	119.78 (19)

O3^{i}—Fe—O6	171.22 (10)	C7—N—Fe	122.80 (18)
O5—Fe—O6	90.10 (10)	O3^{i}—Fe—N	96.32 (9)
O3^{i}—Fe—O2^{iii}	90.57 (8)	O5—Fe—N	93.47 (11)
O5—Fe—O2^{iii}	170.75 (9)	O6—Fe—N	88.33 (10)
O6—Fe—O2^{iii}	81.89 (9)	O3^{i}—Fe—O1^{ii}	89.15 (9)
N—Fe—O2^{iii}	90.92 (8)	O5—Fe—O1^{ii}	86.35 (10)
O1^{ii}—Fe—O2^{iii}	88.50 (8)	O6—Fe—O1^{ii}	86.18 (9)
C1—O1—Fe^{iv}	128.75 (19)	N—Fe—O1^{ii}	174.51 (7)
C1—O2—Fe^{iii}	123.13 (16)	H10B—O5—H10A	114 (5)
C2—O3—Fe^{v}	141.11 (19)	Fe—O6—H9B	119 (4)
Fe—O5—H10B	126 (4)	Fe—O6—H9A	102 (3)
Fe—O5—H10A	116 (3)	H9B—O6—H9A	110 (5)

Symmetry codes: (i) $x-1, y, z$; (ii) $x-1, y+1, z$; (iii) $-x+2, -y, -z$; (iv) $x+1, y-1, z$; (v) $x+1, y, z$.

Table S6 Selected bond lengths (Å) and angles (º) for **6**

Fe—O4^{i}	2.101 (2)	Fe—N	2.187 (2)
Fe—O2^{ii}	2.1238 (19)	O4—Fe^{iv}	2.101 (2)
Fe—O3^{i}	2.126 (2)	O2—Fe^{iii}	2.1238 (19)
Fe—O5	2.1367 (19)	O3—Fe^{iv}	2.126 (2)
Fe—O6	2.151 (2)		
O4^{i}—Fe—O2^{ii}	174.62 (7)	C5—N—Fe	123.92 (18)
O4^{i}—Fe—O3^{i}	82.85 (8)	C4—N—Fe	118.38 (17)
O2^{ii}—Fe—O3^{i}	96.97 (8)	O4^{i}—Fe—N	90.82 (8)
O4^{i}—Fe—O5	94.68 (8)	O2^{ii}—Fe—N	94.52 (7)
O2^{ii}—Fe—O5	79.99 (7)	O3^{i}—Fe—N	98.33 (8)

O3^{i}—Fe—O5	82.29 (8)	O5—Fe—N	174.50 (8)
O4^{i}—Fe—O6	83.64 (8)	O6—Fe—N	90.54 (8)
O2^{ii}—Fe—O6	95.66 (8)	C7—O2—Fe^{iii}	119.64 (16)
O3^{i}—Fe—O6	163.92 (8)	C7—O3—Fe^{iv}	134.61 (18)
O5—Fe—O6	90.17 (8)	C1—O4—Fe^{iv}	123.08 (18)

Symmetry codes: (i) x, $-y+3/2$, $z+1/2$; (ii) $-x+1$, $y-1/2$, $-z+3/2$; (iii) $-x+1$, $y+1/2$, $-z+3/2$; (iv) x, $-y+3/2$, $z-1/2$.

Table S7 Bond lengths (Å) and angles (°) of hydrogen bonds for **2**

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠(D-H···A)
O(5)-H(5B)···O(2)	0.90	2.14	2.987(4)	156
O(6)-H(6A)···O(5)	0.90	1.98	2.748(4)	142
O(6)-H(6B)···O(3)	0.90	1.96	2.777(3)	150
C(11)-H(18A)···O(3)	0.96	2.39	3.336(3)	167

Table S8 Bond lengths (Å) and angles (º) of hydrogen bonds for **3**

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠(D-H···A)
N(1)-H(1B)···O(3)	0.88	1.90	2.776(4)	175
O(5)-H(5B)···N(1)	0.95	2.35	3.277(4)	165
C(7)-H(7A)···O(3)	0.95	2.44	3.259(5)	144
C(7)-H(7B)···O(4)	0.95	2.31	3.212(5)	159

Table S9 Bond lengths (Å) and angles (º) of hydrogen bonds for **4**

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠(D-H···A)
N(2)-H(2A)···O(4)	0.86	1.96	2.809(3)	169
C(1)-H(1)···O(4)	0.68(4)	2.52(4)	3.136(4)	152
C(4)-H(4)···O(2)	0.83(3)	2.38(3)	3.035(3)	137

Table S10 Bond lengths (Å) and angles (º) of hydrogen bonds for **5**

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠(D-H···A)
O(6)-H(9A)···O(2)	0.70(4)	2.02(4)	2.695(3)	163
O(6)-H(9B)···O(7)	0.63(5)	2.17(4)	2.801(4)	177
O(5)-H(10A)···O(7)	0.67(4)	2.16(4)	2.821(4)	172
O(5)-H(10B)···O(4)	0.71(5)	2.12(5)	2.789(4)	159

Table S11 Bond lengths (Å) and angles (º) of hydrogen bonds for **6**

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠(D-H···A)
O(5)-H(5A)···O(1)	0.82	1.95	2.766(3)	171
O(5)-H(5C)···O(2)	0.90	2.14	2.759(3)	125
O(6)-H(6A)···O(1)	0.82	2.07	2.827(3)	154
O(6)-H(6B)···O(1)	0.90	2.48	2.980(3)	115
C(3)-H(3A)···O(4)	0.93	2.45	3.313(4)	155
C(4)-H(4A)···O(1)	0.93	2.53	3.320(4)	142
C(5)-H(5B)···O(3)	0.93	2.34	3.181(4)	150

3. Additional Structural Figures

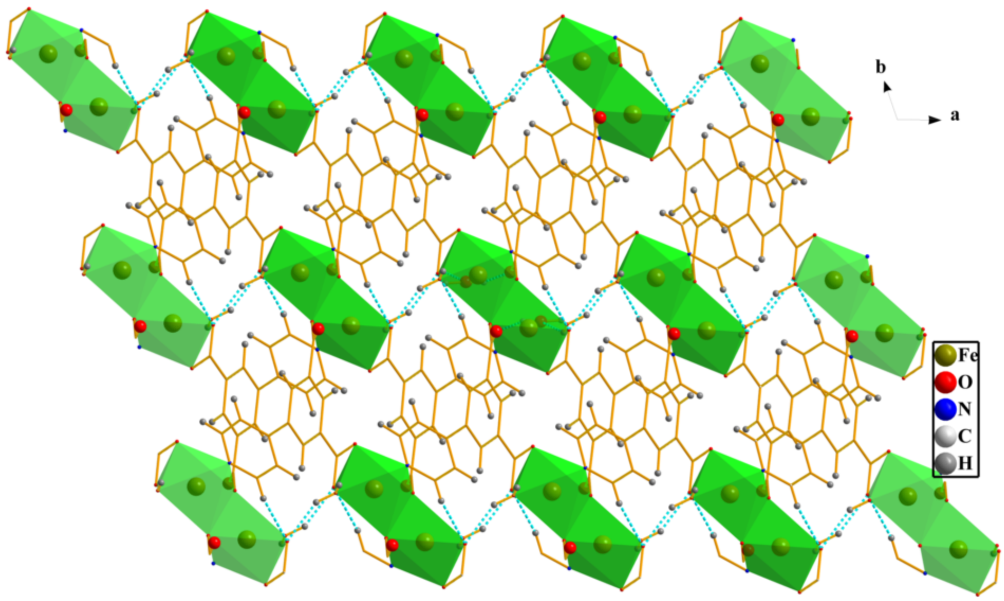


Fig. S1 H-bonding interactions in the 3D supramolecular framework of **2**

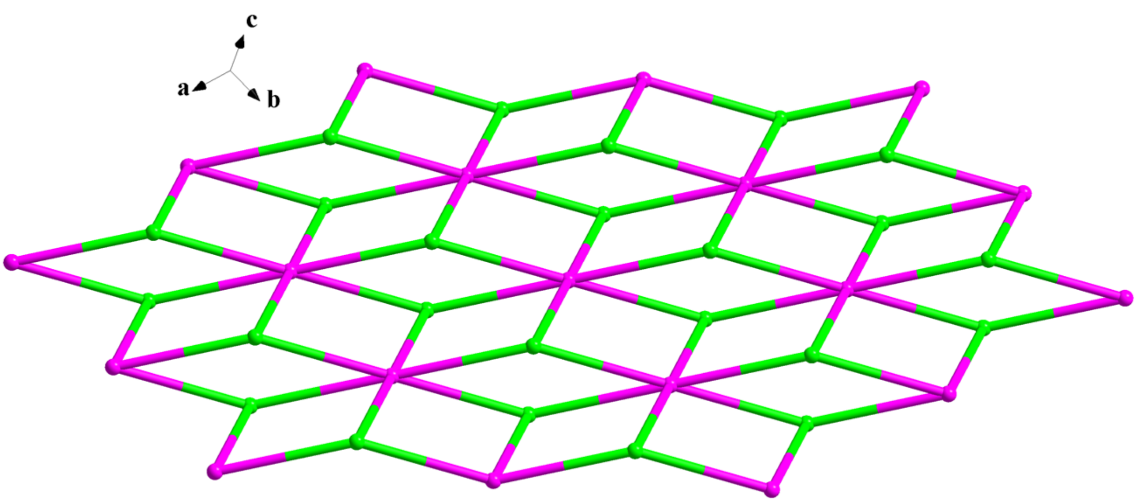


Fig. S2 The (3,6)-connected 2D framework with a *kgd* topology network in **2**

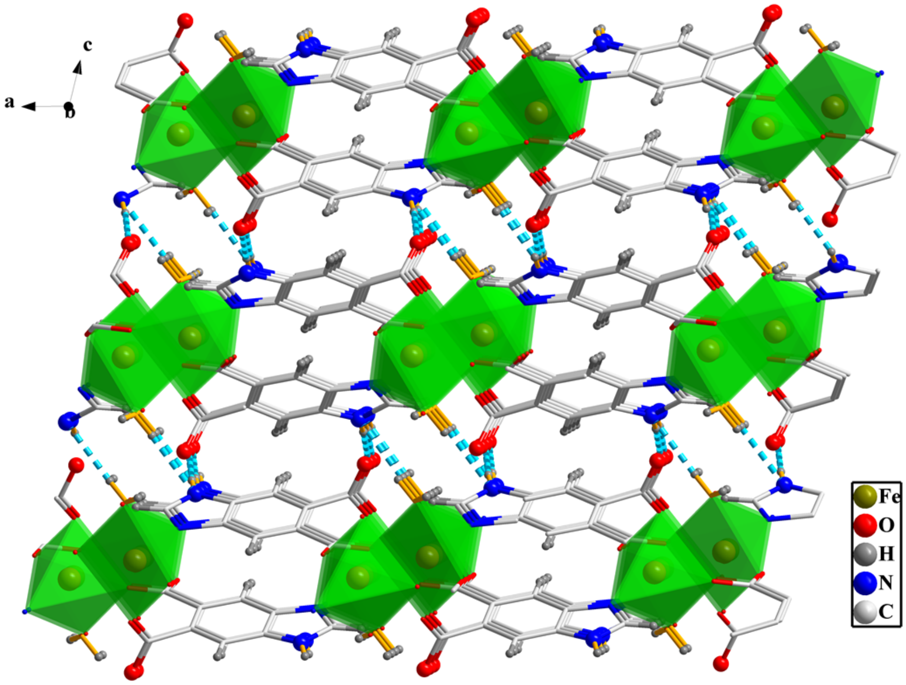


Fig. S3 H-bonding interactions in the 3D supramolecular framework of **3**

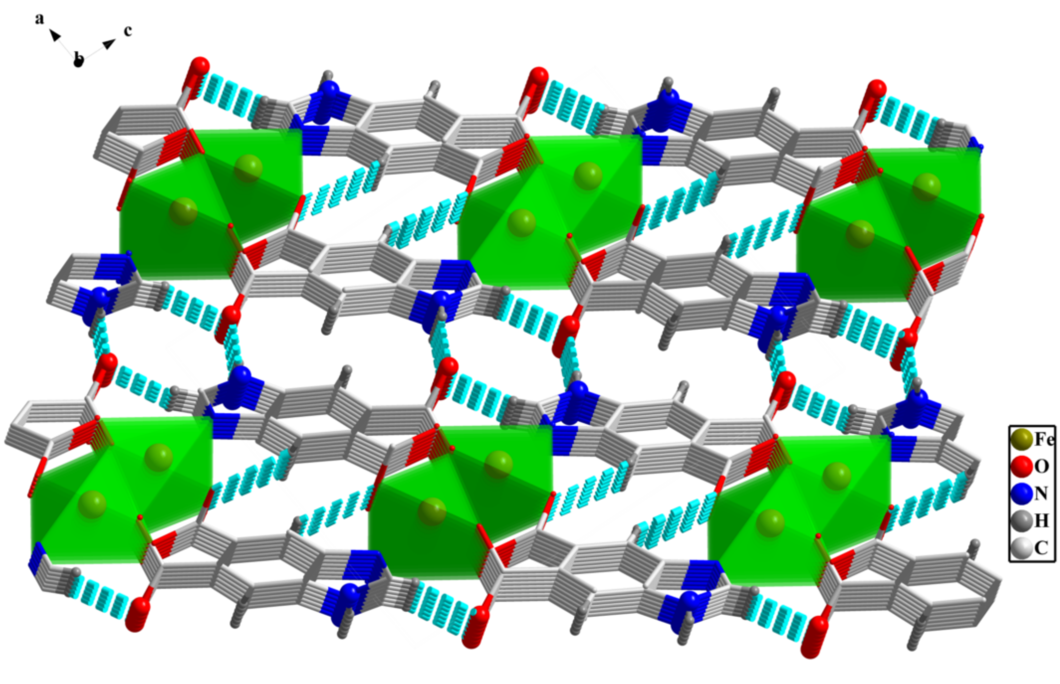


Fig. S4 H-bonding interactions in the 3D supramolecular framework of **4**

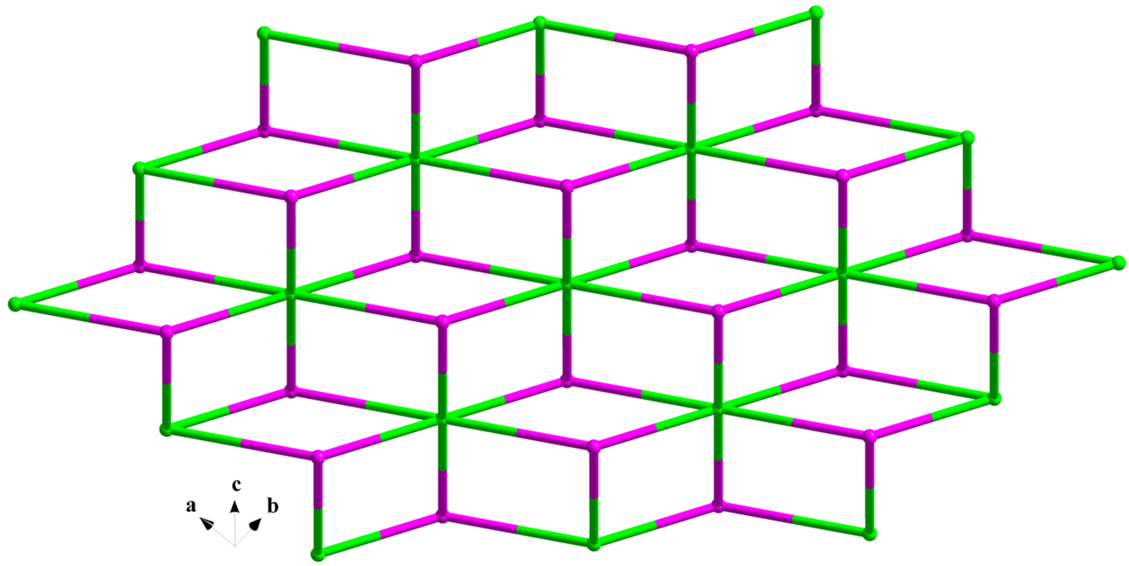


Fig. S5 The (3,6)-connected 2D framework with a *kgd* topology network in **5**

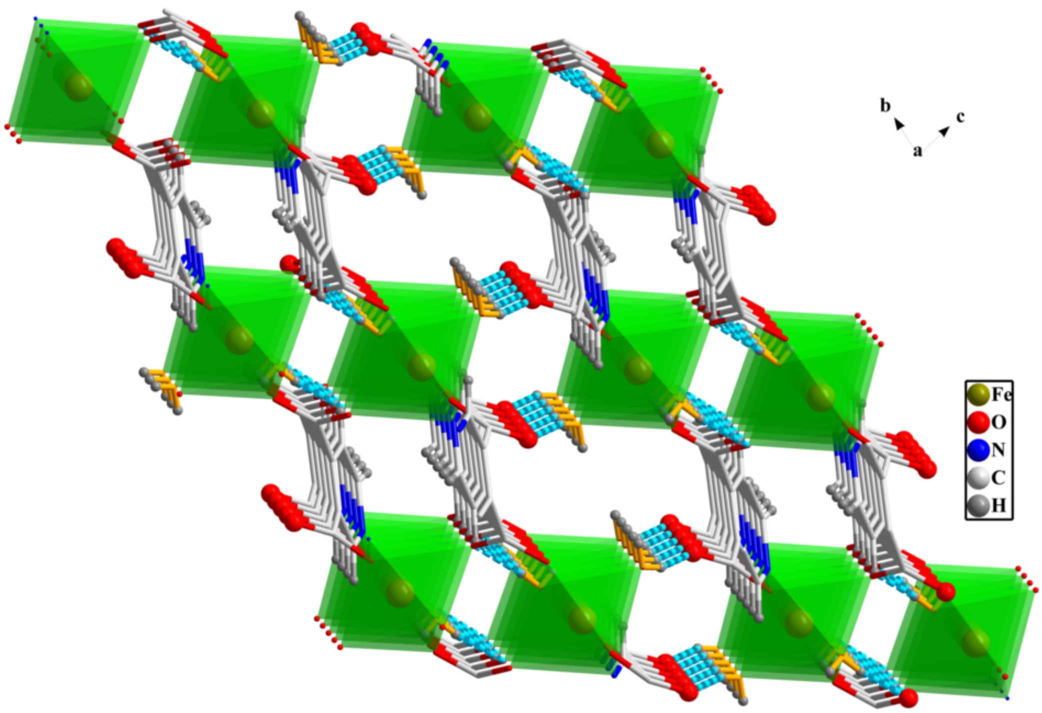


Fig. S6 H-bonding interactions in the 3D supramolecular framework of **5**

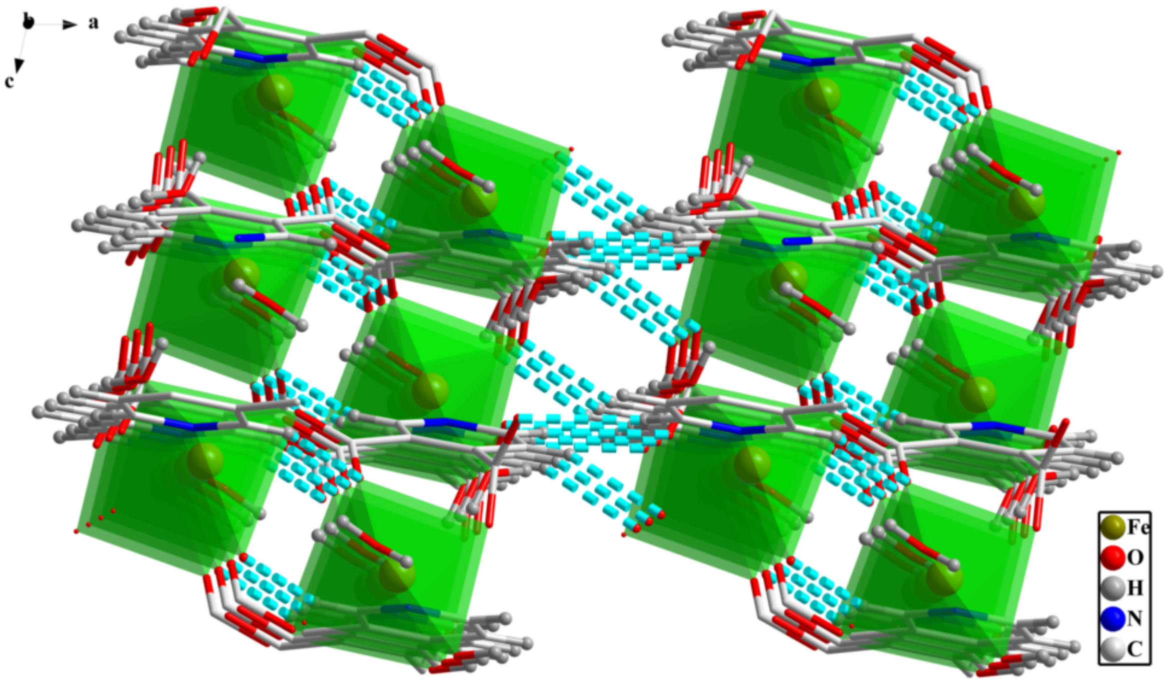


Fig. S7 H-bonding interactions in the 3D supramolecular framework of **6**

4. Powder X-ray Diffraction

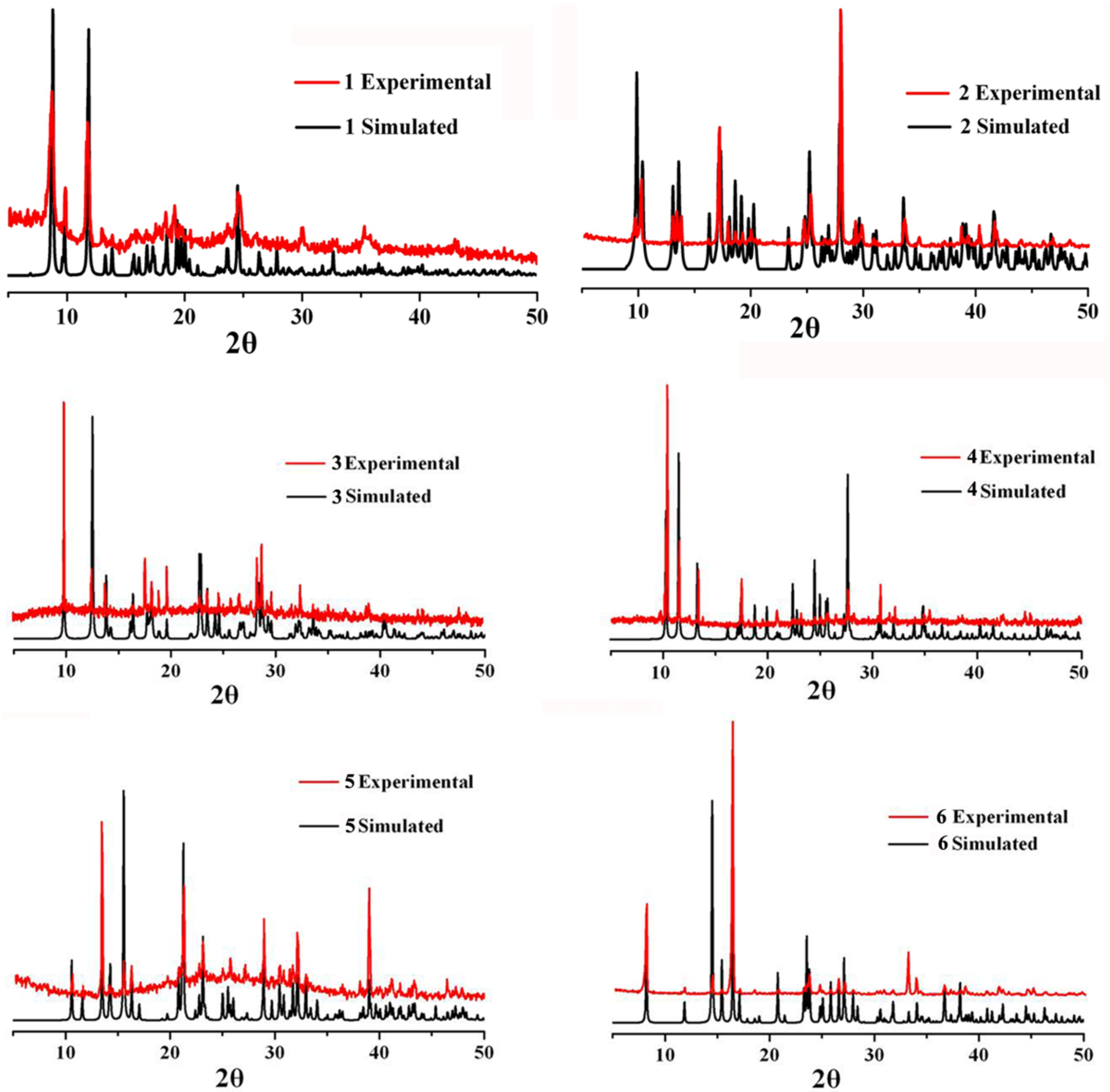


Fig. S8 Simulated and experimental XRD powder patterns of **1-6**

5. Thermogravimetric Analysis

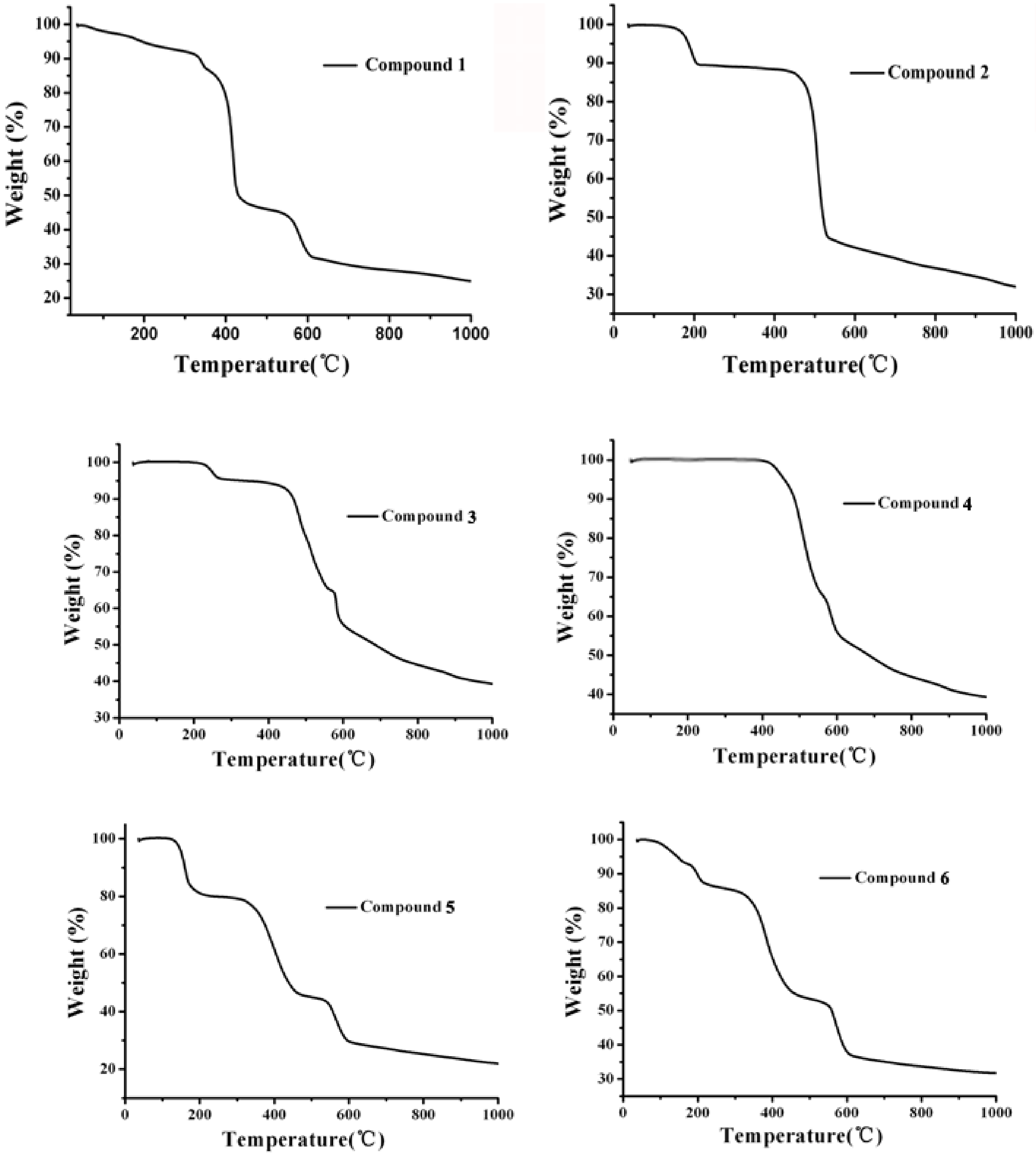


Fig. S9 TG curves of compounds **1-6**

6. Magnetic Figures

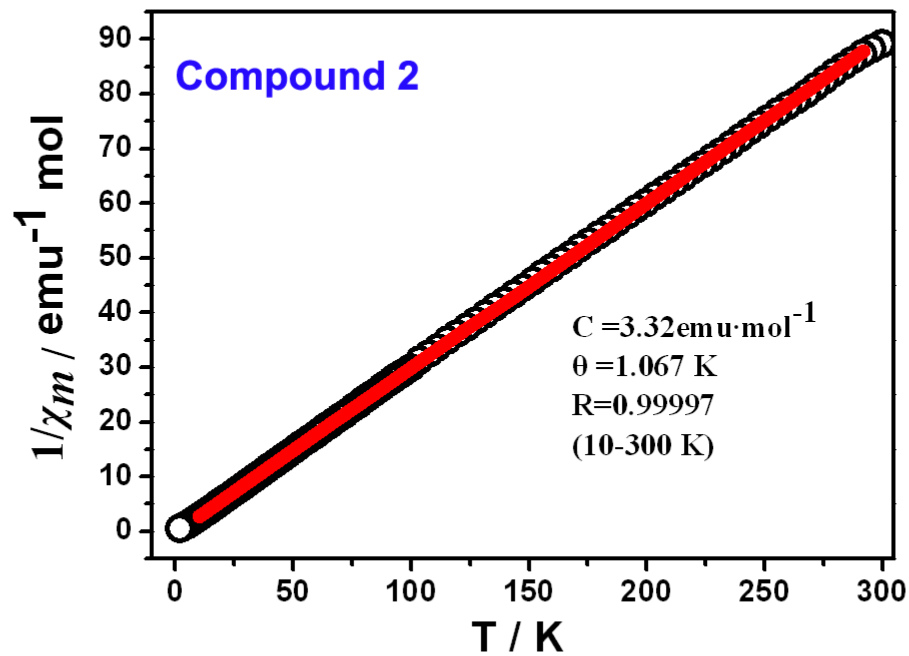


Fig. S10 The red line shows the Curie–Weiss fitting for **2**

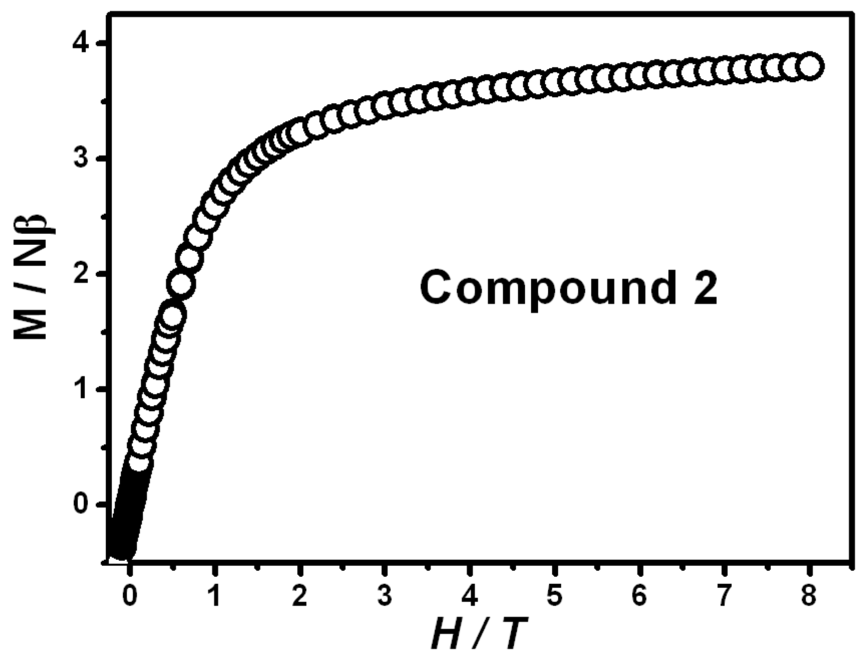


Fig. S11 The *M* vs. *H* plot for **2** measured at 2 K.

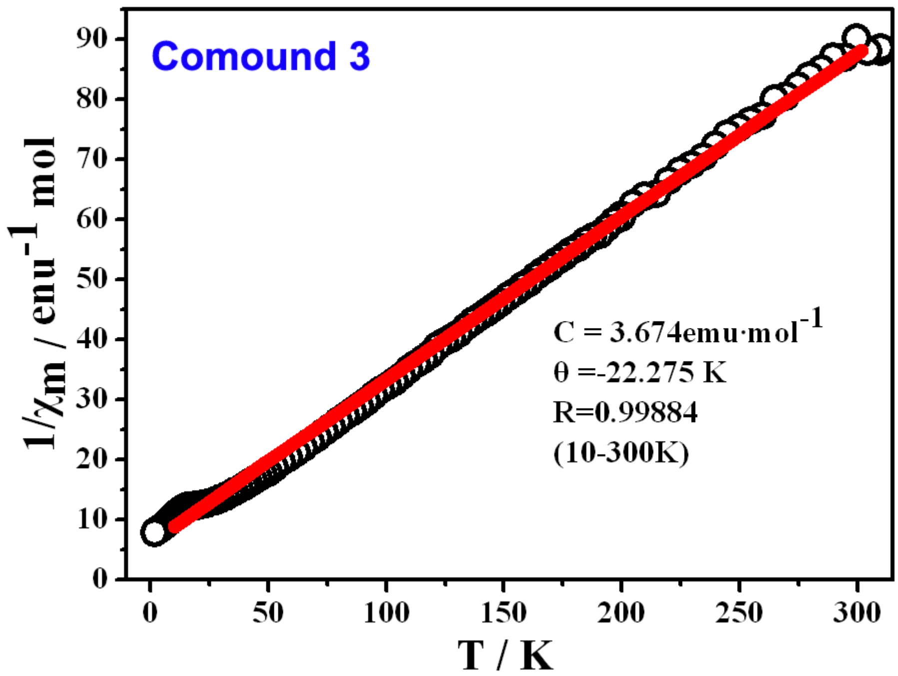


Fig. S12 The red line shows the Curie–Weiss fitting for **3**

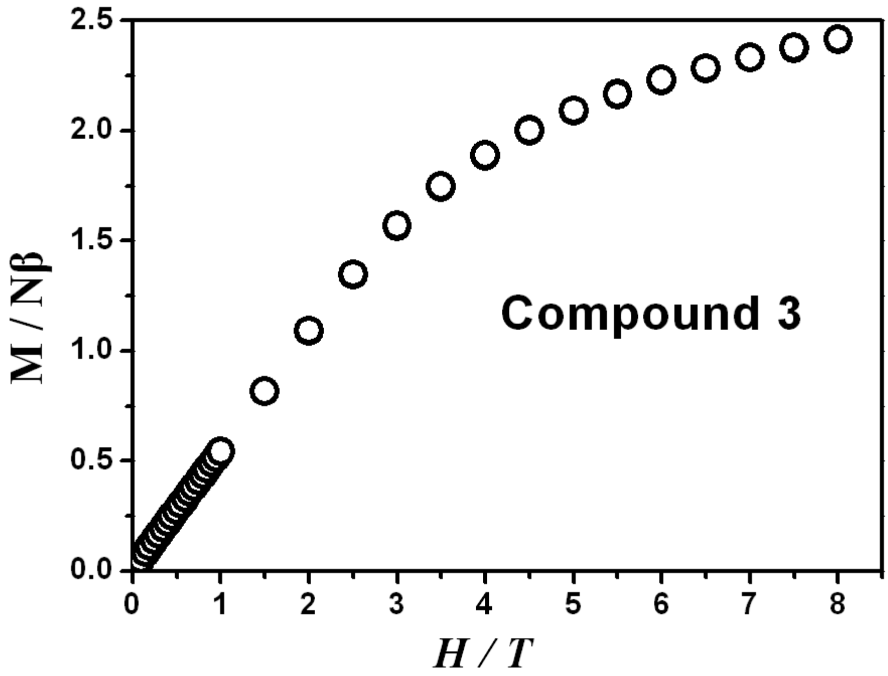


Fig. S13 The *M* vs. *H* plot for **3** measured at 2 K.

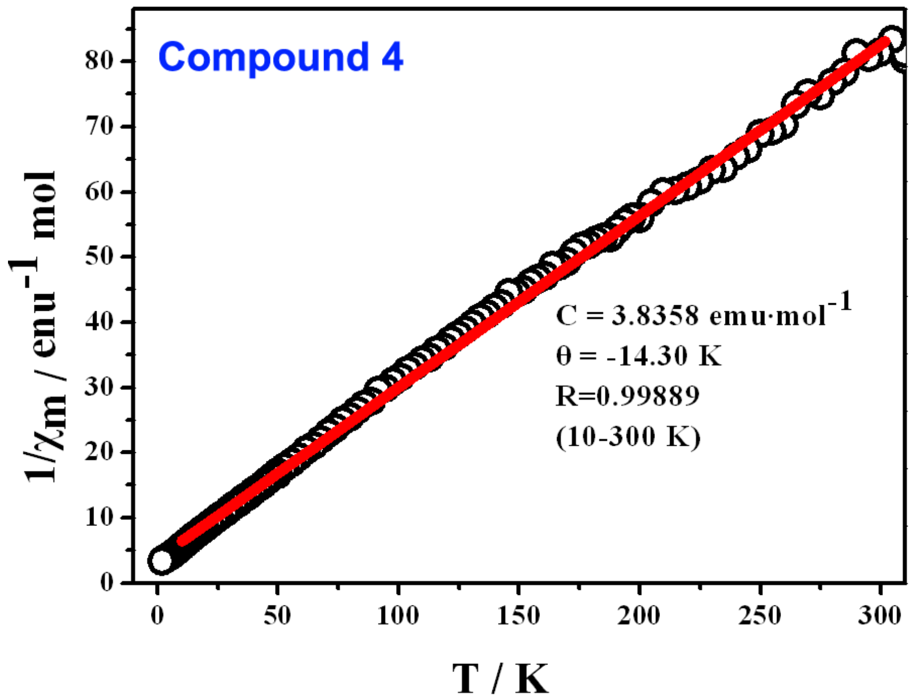


Fig. S14 The red line shows the Curie–Weiss fitting for **4**

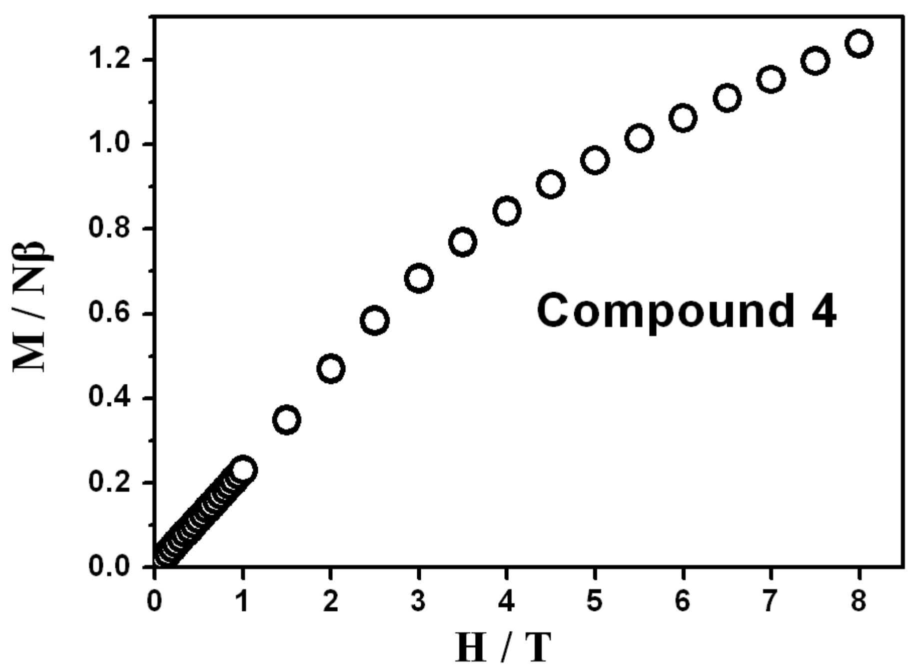


Fig. S15 The *M* vs. *H* plot for **4** measured at 2 K.

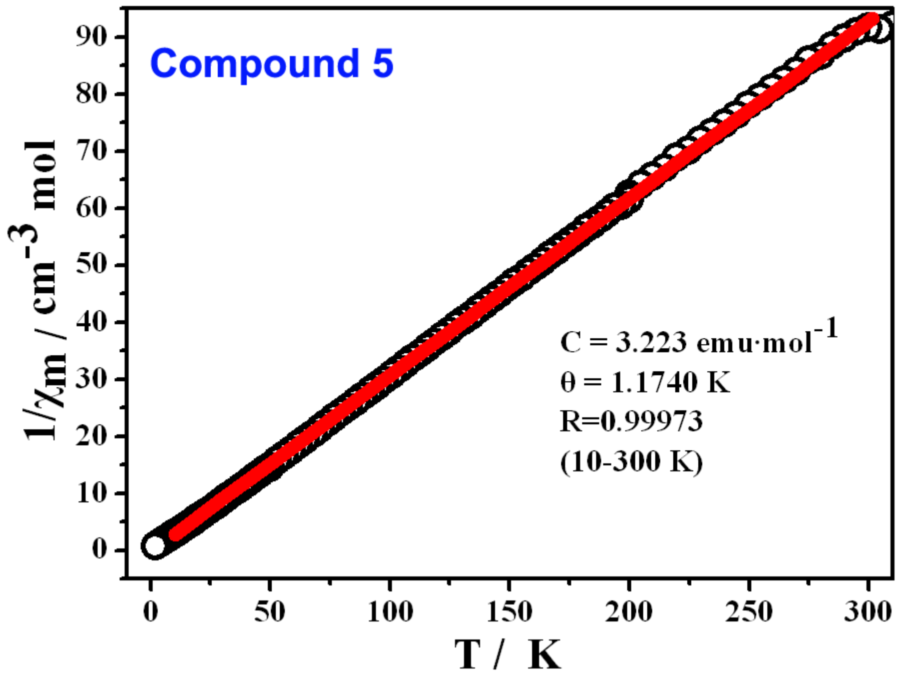


Fig. S16 The red line shows the Curie–Weiss fitting for **5**

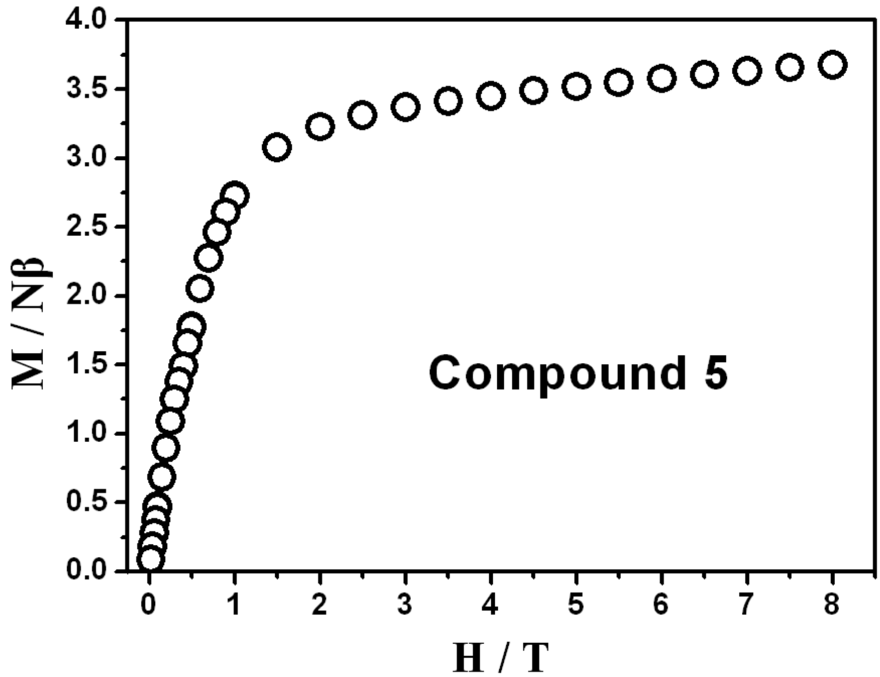


Fig. S17 The *M* vs. *H* plot for **5** measured at 2 K.

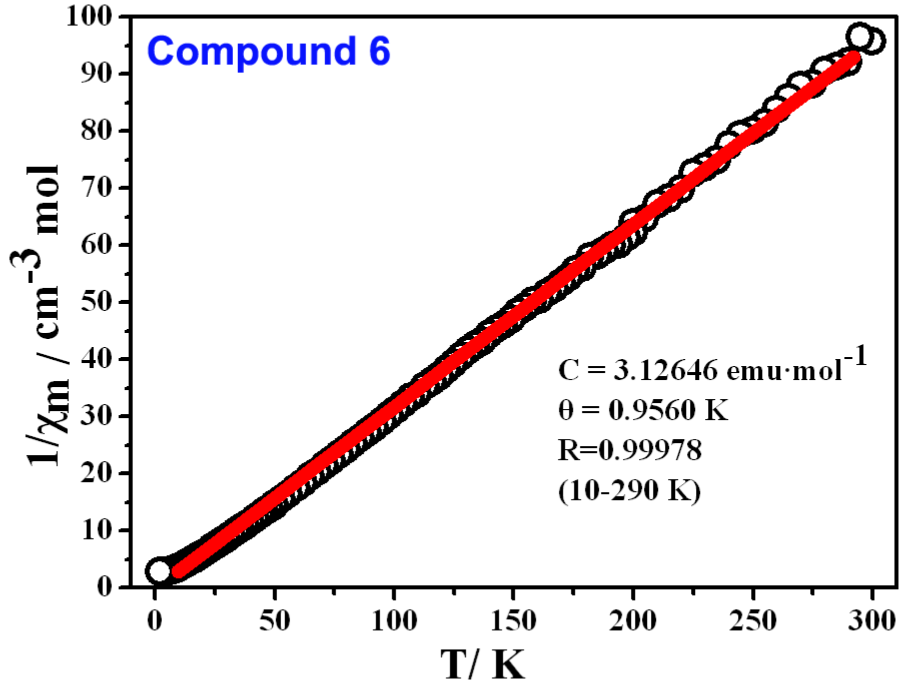


Fig. S18 The red line shows the Curie-Weiss fitting for **6**

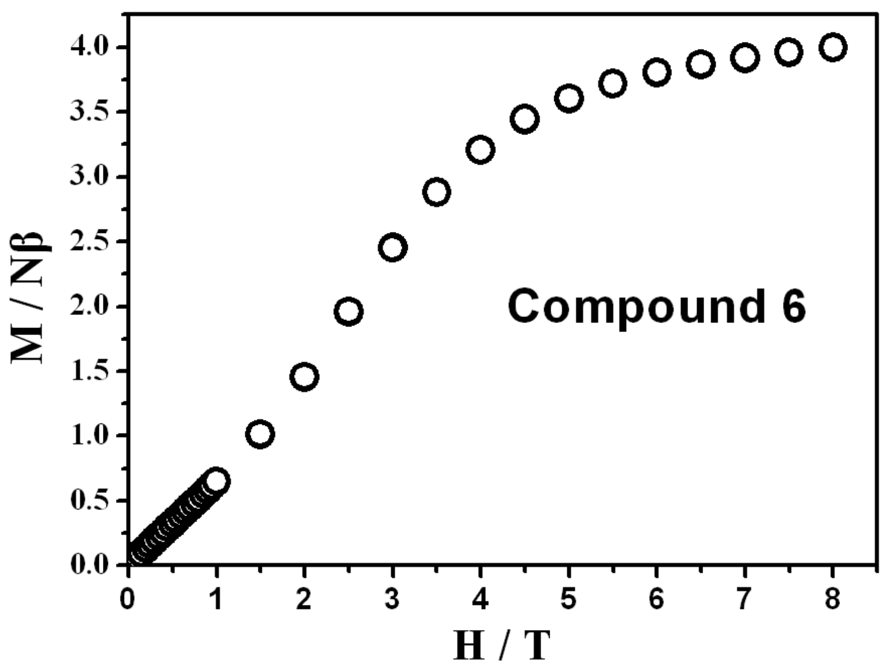


Fig. S19 The *M* vs. *H* plot for **6** measured at 2 K

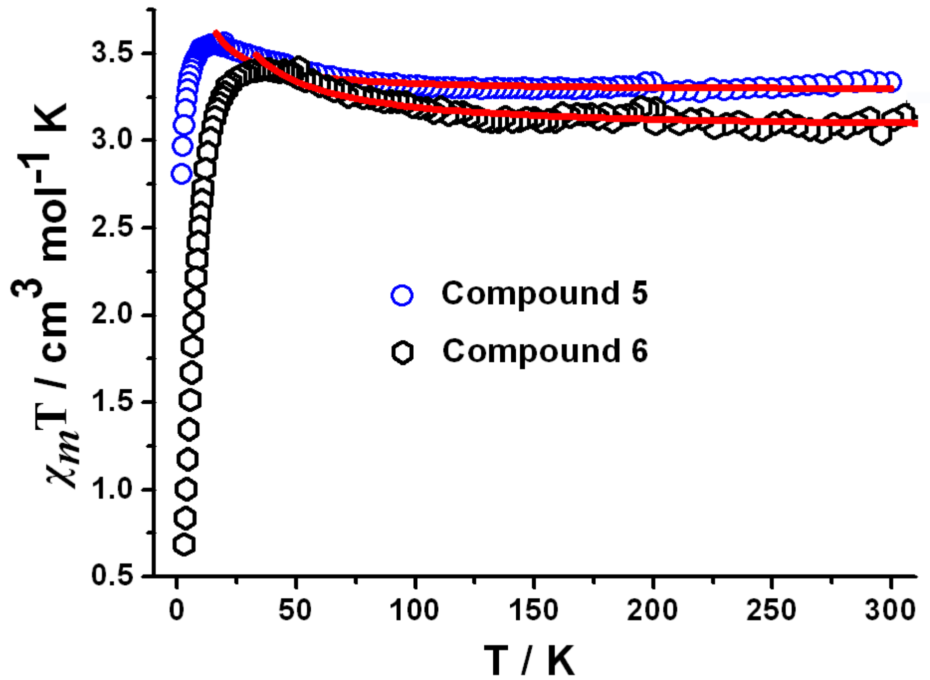


Fig. S20 The red line shows the $\chi_m T$ and fitting for **5** and **6**

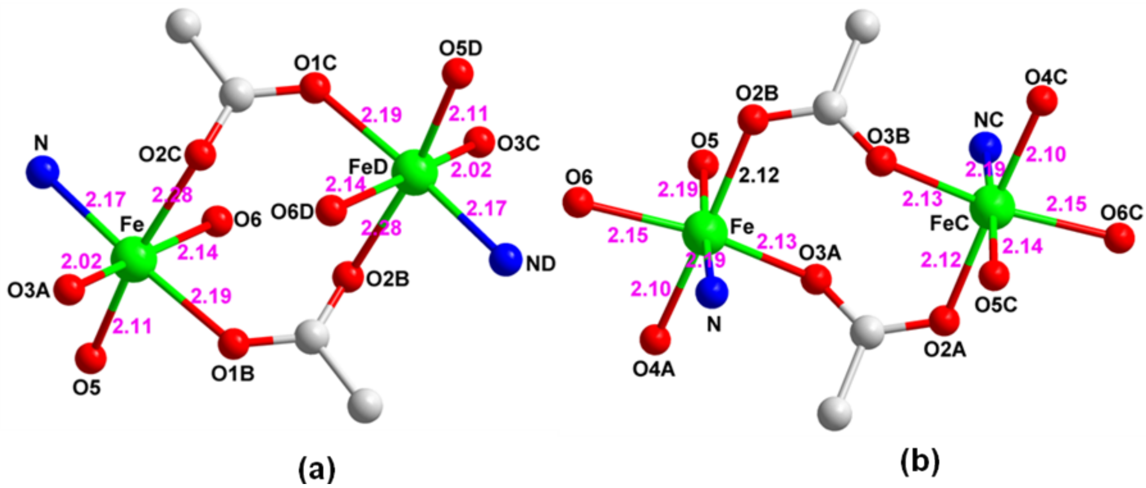
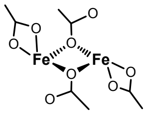
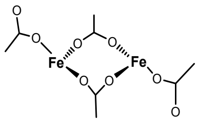
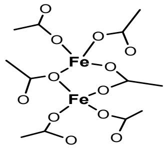
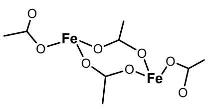
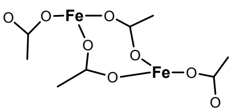


Fig. S21 (a) The *syn-anti* equatorial-axial configuration for **5**. (b) The *syn-anti* the equatorial-equatorial fashion for **6**

Table S12 The exchange couplings of compounds **2**-**6**

Bridging mode	**compounds**	**type**	**Fe-Fe Disatance/(Å)**	**∠Fe-O-Fe**	***g***	***J*(cm^{-1})**
	**2**	bis(μ_2-OR)	3.42	102.96	2.09	0.33
	**3**	bis(*syn*-*syn*)	3.40		2.03	-0.72
	**4**	*syn*-*anti*/ μ_2-OR	3.64	118.9	2.25	-2.76
	**5**	bis(*syn*-*anti*)	4.80		2.09	0.31
	**6**	bis(*syn*-*anti*)	4.70		2.01	0.85